

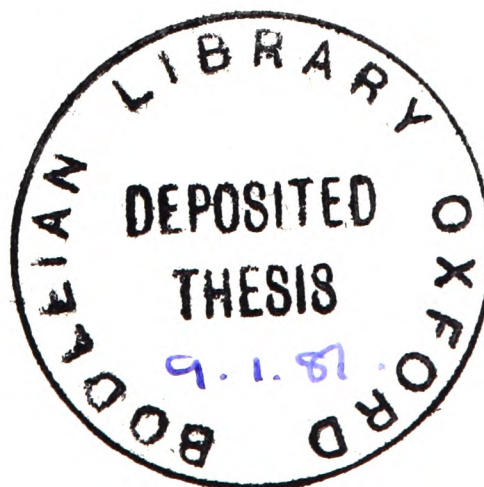
SOME APPLICATIONS OF
NUCLEAR ORIENTATION

by

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A Thesis submitted for the degree of Doctor of
Philosophy in the University of Oxford

October 1980



ABSTRACT

This thesis describes Nuclear Orientation and radio-frequency magnetic susceptibility experiments on the enhanced nuclear antiferromagnet holmium vanadate (HoVO_4) from 1 K down to about 1 mK and in applied fields of up to 2.4 tesla.

Adiabatic demagnetisation of single crystals of HoVO_4 allowed a mean nuclear spin temperature of about 1 mK to be reached. R.f. magnetic susceptibility measurements at approximately 2.4 mK and in an applied field of about 1.5×10^{-2} T identified the spin 'flop' phase of the nuclear antiferromagnet HoVO_4 . The Néel temperature T_N was determined to be 4.0 (2) mK which is in good agreement with the value of 4.8 mK calculated for the dipole-dipole interaction of the enhanced magnetic moments at the Ho site.

Gamma-ray anisotropy measurements of $^{166\text{m}}\text{HoVO}_4$ showed that in low applied fields ($B \lesssim 0.5 \times 10^{-2}$ T) and temperatures of about 1 mK the antiferromagnetic domains were uniformly distributed in the a - a' plane of the tetragonal zircon crystal structure. This was interpreted in terms of pinning of the nuclei due to crystalline imperfections. The spin 'flop' phase was identified in an applied field of 1.2×10^{-2} T which is in good agreement with the susceptibility measurements. Demagnetisation experiments down to the relatively high residual fields of 4.2×10^{-2} T and 7.4×10^{-2} T and temperatures of about 2 mK showed that the effective field at the nucleus was much smaller than the applied field thus indicating that there was some antiferromagnetic ordering even at these relatively high fields.

The magnetic dipole moment of $^{166\text{m}}\text{Ho}$ was estimated to be $3.60 (6) \mu_N$ from gamma-ray anisotropy measurements of some

gamma transitions of known NO parameters. This value is in good agreement with the value of $3.69 \mu_N$ calculated for the Nilsson configuration $[6337/2]_n [5237/2]_p$ for the extra-core neutron and proton respectively. Mixing ratio (δ) measurements for gamma-ray transitions between the ($K^\pi = 2^+$) γ -vibrational and ($K^\pi = 0^+$) ground state rotational bands of the daughter nucleus ^{166}Er showed that M1 transition probabilities are predominantly via $\Delta K = 1$ band mixing which is in good agreement with systematics for deformed nuclei in this mass region.

Nuclear refrigeration experiments on a powdered compressed HoVO_4 - copper pill allowed a lattice temperature of 3.9 (3) mK to be reached. Thermal contact measurements on a similar pill showed that the effective area of contact between the salt particles and copper was only about 30 cm^2 which was a small fraction of the total surface area available. Preliminary work on a HoVO_4 - gold pill enabled a ^{60}Co Co thermometer to be cooled to 6.0 (1) mK.

ACKNOWLEDGEMENTS

I would like to thank the following:

Prof. B. Bleaney for extending to me the facilities of the Clarendon Laboratory and for many invaluable discussions, suggestions and sustained interest in this work.

Prof. E.W.J. Mitchell for extending to me the facilities of the Clarendon Laboratory.

Dr. A.L. Allsop for his patient supervision, skilful help with the running of low temperature equipment and critical discussions on various aspects of this thesis.

Dr. N.J. Stone for many helpful suggestions and involvement with the experiments at every stage.

Dr. G.J. Bowden, especially for his invaluable theoretical skills and help with the analysis and interpretation of the data.

Prof. H. Suzuki, Dr. D.H. Chaplin and Dr. I.A. Campbell for help with some of the experiments.

Mr. S. Hornung, Mr. A.H. Wood and Mr. D.W. Murray for their cheerful co-operation at all times.

Mrs. B.M. Wanklyn and Dr. S.H. Smith for growing the high quality crystals of HoVO_4 .

Mr. M.R. Wells for doing the N.M.R. measurements and for many helpful discussions.

Mr. F.R. Wondre for help with the x-ray equipment of the Materials Preparation Group.

Mr. G. Read for skilfully shaping some of the crystals.

The other members of the technical staff of the Clarendon Laboratory for their unending patience and friendly co-operation,

especially Messrs. H.S. Locke and G. Mathews of the Research Workshop and Messrs. J. Siertsema and T.J. Hastings of the Electronic Workshop.

The Royal Society, the Clarendon Laboratory and the Scott Foundation for Maintenance Grants.

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CHAPTER 1

EXPERIMENTAL TECHNIQUES AND DATA ANALYSIS

1.1 Introduction

This chapter describes the low temperature apparatus used in making the measurements of this thesis. It consisted mainly of a ^3He - ^4He dilution refrigerator (DR400, built by Oxford Instruments) with associated magnets for the Nuclear Orientation and Refrigeration experiments. A brief account is also given of the gamma-ray spectroscopy detectors and associated electronics used in the Nuclear Orientation experiments.

1.2 The ^3He - ^4He Dilution Refrigerator

Designed and built by Oxford Instruments, it is capable of maintaining stable temperatures down to 12 mK. Typically it takes about two days to assemble and cool to liquid helium temperatures. However, once in operation it can maintain stable mixing-chamber temperatures for weeks provided the system is kept topped up with liquid refrigerants. The temperature of the mixing-chamber is altered by changing the current through a resistive heater thermally anchored to it. This particular dilution refrigerator has a cooling power of approximately $3\mu\text{W}$ at 25 mK.

Lounasmaa (1) has discussed in detail the operating principles of the ^3He - ^4He dilution refrigerator. The following is a brief description of the bare essentials. Below about 0.9 K a mixture of ^3He and ^4He separates out into two phases, a ^3He rich (concentrated) phase and a ^4He rich (dilute) phase. If ^3He is made to move from the concentrated to the dilute phase across the phase boundary, cooling will occur due to the difference in entropy between the two phases. In a dilution refrigerator the phase separation occurs in the mixing-chamber which is the coldest part

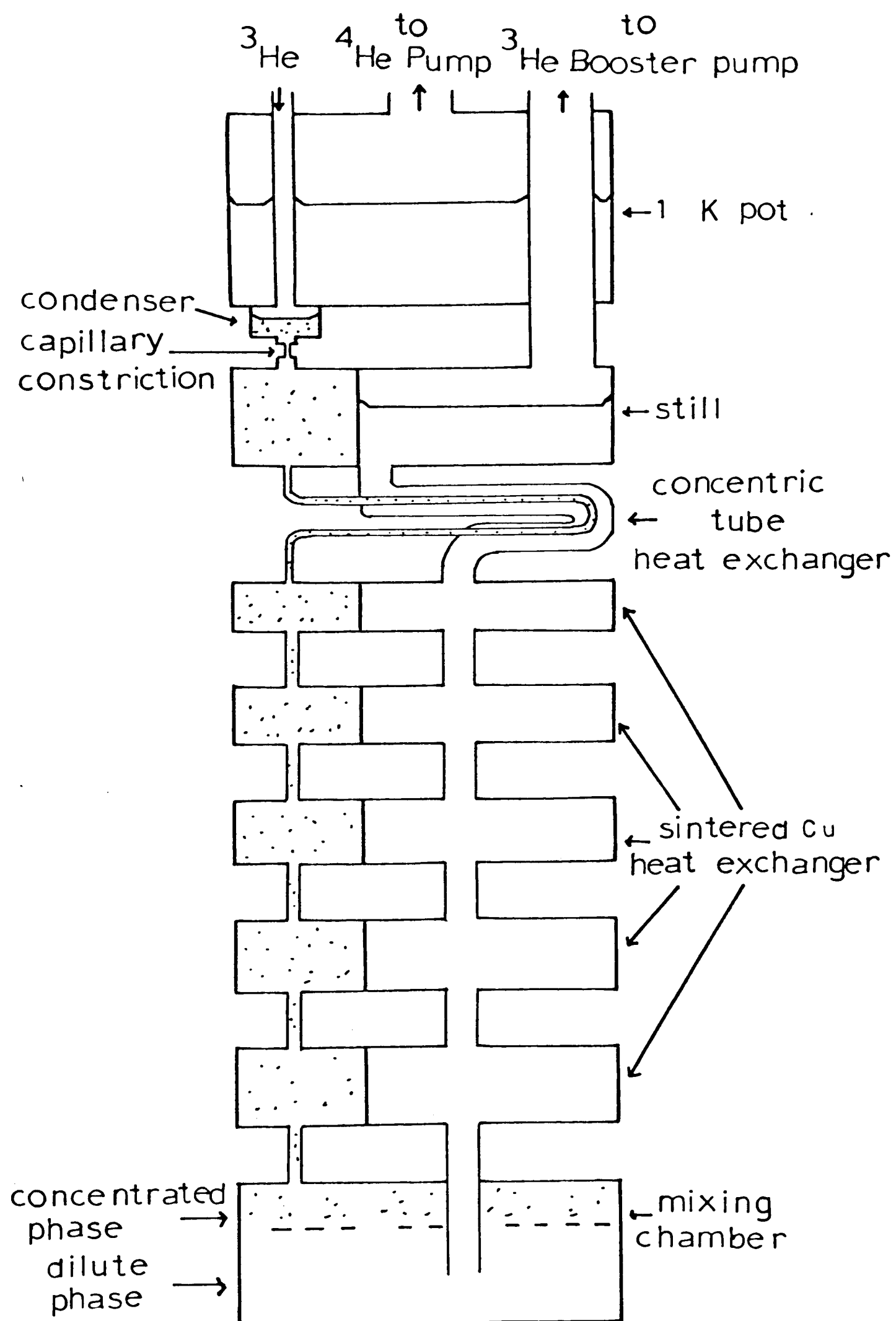


Fig. 1.1 Layout of the dilution unit

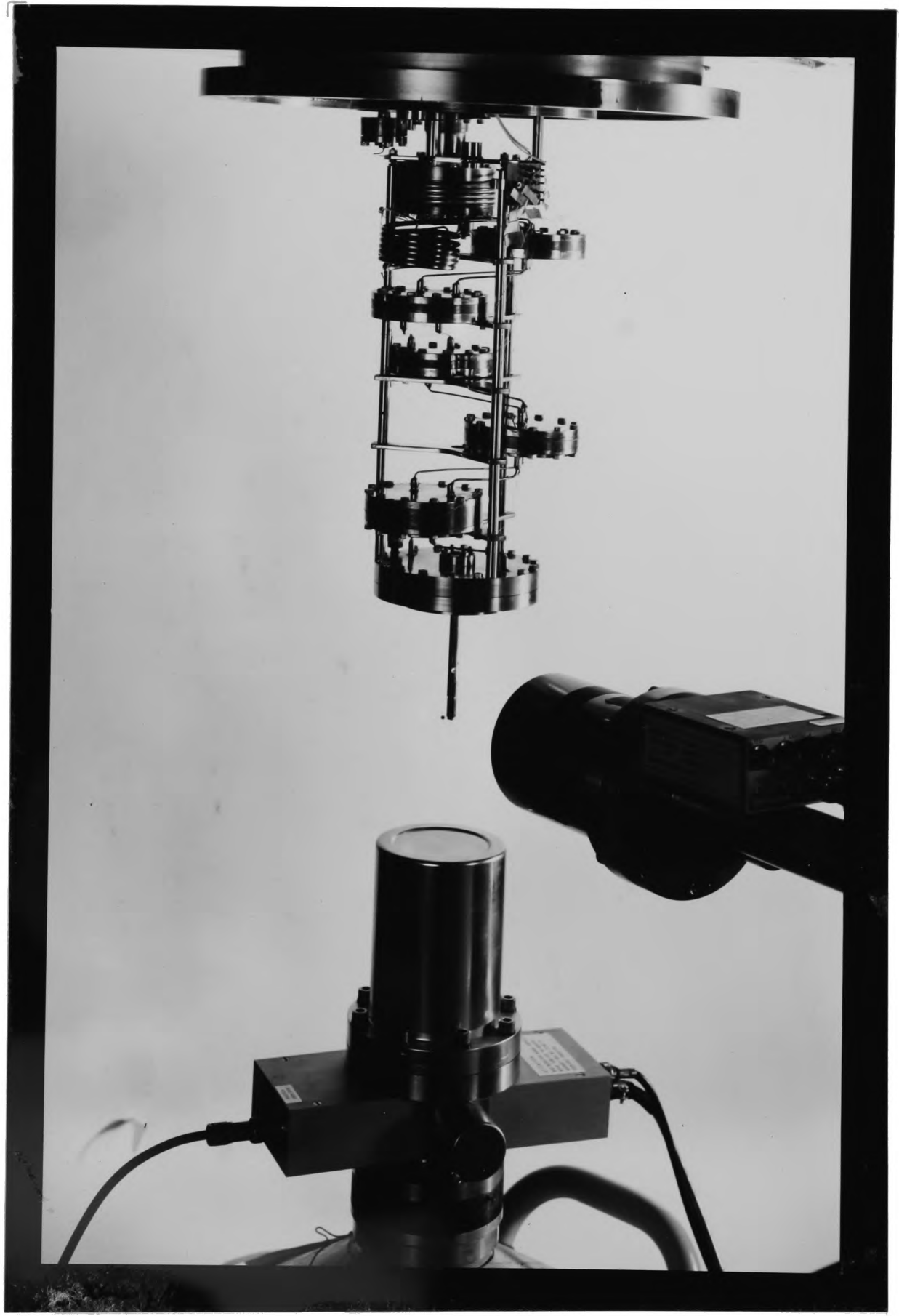


Plate 1 The ^3He - ^4He dilution refrigerator

of the dilution unit. ^3He is made to cross the boundary by pumping on the dilute phase. The osmotic pressure set up on depletion of ^3He causes ^3He to move from the concentrated to the dilute phase thus cooling the liquid. In practice, the pumping is done on a separate chamber (still) maintained at about 0.6 K for optimum circulation of ^3He (see Fig. 1.1). The returning ^3He is pre-cooled by passing through a ^4He pumped 1 K pot and condensed through a constriction in a capillary. The liquid ^3He is cooled further by passing through a concentric tube heat-exchanger and five step heat-exchangers containing sintered copper. While the cooling power can be improved by increasing the circulation rate, the increased viscous heating reduces efficiency unless the heat-exchangers are also increased in size.

1.3 The 'Top-Loading' Facility

The DR400 has a facility which allows samples to be changed while the dilution refrigerator is in operation. This allowed experiments to be done on radioisotopes with half-lives as short as 10 hours (2). The refrigerator has an access tube running down through the Inner Vacuum Can through which sample rods can be inserted. 'Top loading' of a sample consists of coupling the sample rod by a left-handed thread on to a specially designed 'top-loading syphon'. This is inserted through a vacuum lock until its lower end is just above the 1 K pot, and pre-cooled to 4.2 K by pumping liquid helium through the syphon. Radiation baffles at the 1 K pot are then swung out of the way, and after de-coupling from the liquid helium supply the syphon is lowered and the cold finger screwed into the mixing-chamber. Further screwing then dis-engages the syphon which can be removed through the vacuum

lock and the 1 K radiation baffles closed.

In the r.f. susceptibility measurements, the need for wiring of electrical leads meant that the refrigerator had to be assembled for each experiment. This was also true for experiments where samples were attached to the cold finger by means of silver paint. Here the link was fragile and so there was danger of the sample being dislodged. The 'top-loading' system was therefore not used for these experiments and all assembly was at room temperature.

1.4 Thermometry

1.4.1 Magnetic Thermometer

The magnetic susceptibility of cerium magnesium nitrate (CMN) obeys the Curie-Weiss law

$$\chi = c/(T - \Delta) \quad 1.1$$

where C and Δ are constants. In principle, knowing the magnetic susceptibility at two fixed points is enough to calibrate a CMN magnetic thermometer, given that Δ is known. For a paramagnetic system

$$\Delta = (N-d) c \quad 1.2$$

where N is a term representing the dipolar interactions between the electronic spins, d is the demagnetisation factor and c is the Curie constant per unit volume of specimen. For CMN above about 10 mK, N is $\sim 1/3$ and $c = 1.01 \times 10^{-2}$ (S.I.), so that for a spherical specimen ($d = 1/3$), theory predicts $\Delta_0 = -0.27$ mK for a spherical sample of CMN magnetised perpendicular to the trigonal axis (3), which is

in good agreement with some measurements (4). However, there is some controversy with regard to the temperature scale of CMN (4) and our own measurements on a single crystal CMN cylinder (Sample 1) of demagnetisation factor 0.175 gave $\Delta = -3.3(3)$ mK. This gave $N = -0.153(20)$, in some disagreement with the expected value of $1/3$. This measured value was used to calculate Δ for a second cylindrical sample (Sample 2) of demagnetisation factor 0.058. This calculated Δ was used to calibrate the carbon resistor of section 1.4.2.

The susceptibility of the CMN crystal was measured using an r.f. technique (see also section 4.2.4). The sample was placed inside a coil which formed the inductive part of a self-resonant L-C circuit and had a Q factor of about 100 at low temperatures. Stable oscillations in the tank circuit were maintained by an oscillator at about 400 kHz (5) and the frequency read on a frequency meter. The coil was constructed by winding a single layer of 200 turns of 46 SWG enamelled copper wires on a PTFE sleeve 7 mm diameter and fixed in position using GE varnish. It had an inductance of $\sim 30 \mu\text{H}$. A 3300 pF polystyrene capacitor formed the capacitive part of the tank circuit. The tank circuit was linked to the oscillator by means of a specially constructed r.f. co-axial cable (see section 1.8).

Changes in the susceptibility of the sample produce changes in the resonant frequency $\delta\nu$ and are given by the expression

$$\delta\nu = -\frac{1}{2}\nu_0 f \delta\chi_{\text{r.f.}} \quad 1.3$$

where ν_0 is the frequency without the sample, f is the filling

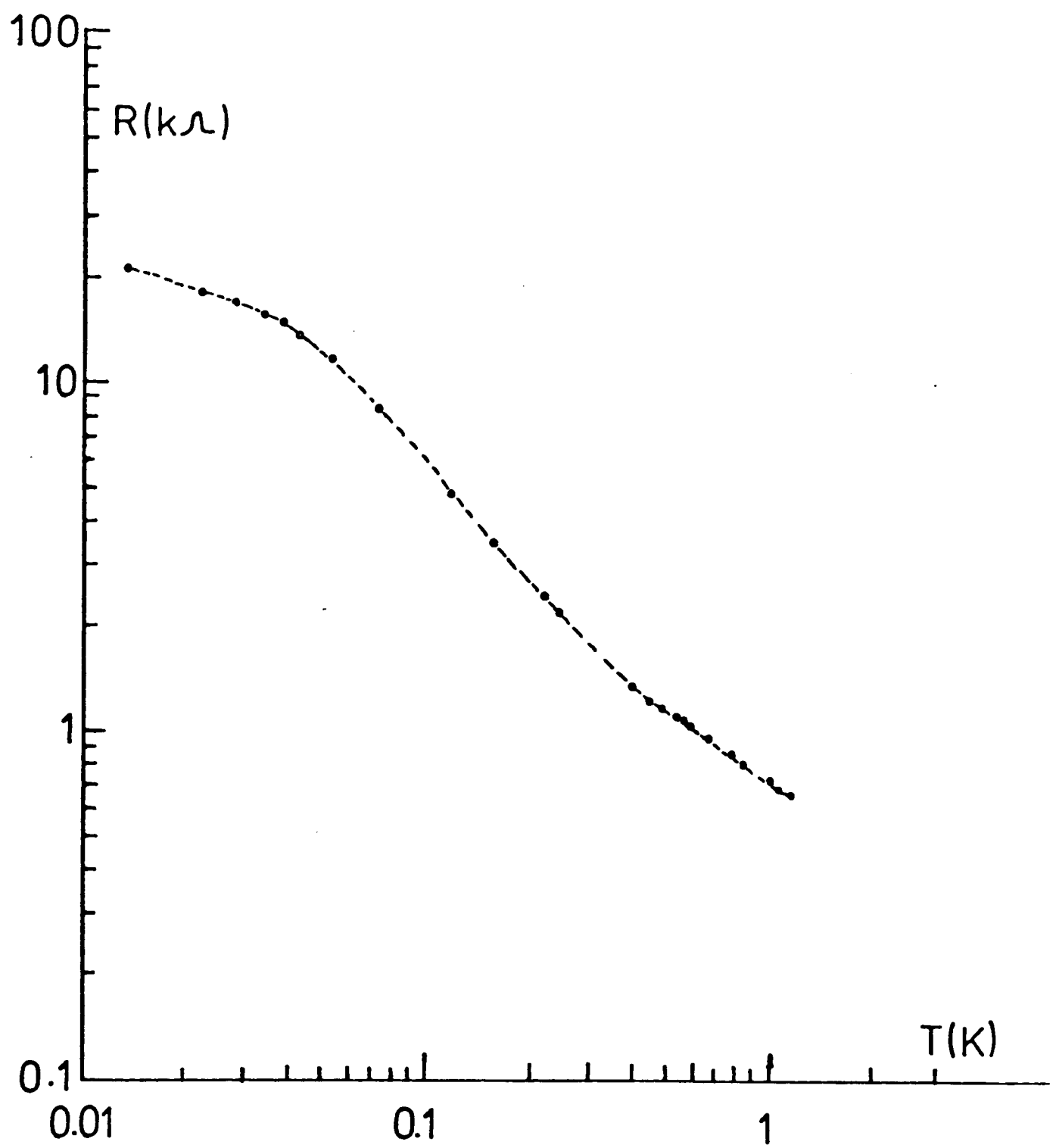


Fig. 1.2 Carbon resistor calibration

factor and $\delta\chi_{\text{r.f.}}$ the r.f. susceptibility change. As the frequency shift $\delta\nu$ is directly proportional to the susceptibility change $\delta\chi_{\text{r.f.}}$.

$$\delta\nu \propto 1/(T - \Delta) \quad 1.4$$

This allowed the calibration of the CMN thermometer in terms of frequency shifts.

1.4.2 Carbon Resistor Thermometer

The outer coating of a 220Ω $\frac{1}{2}\text{W}$ Speer carbon resistor was ground down and ^{the resistor} attached to the mixing-chamber using silicone grease. Owing to poor thermal contact, the resistor was found to be unreliable below about 50 mK. It was calibrated using a CMN single crystal (Sample 2). The r.f. susceptibility of the crystal was measured at 1.2 K and 13.9 mK with temperatures derived from vapour pressure of ^4He and a ^{60}Co Co Nuclear Orientation (NO) thermometer respectively (see below). As explained in section 1.4.1, this was sufficient to define a temperature scale for the magnetic thermometer. The error in the calibration is estimated to be 5% between 50 and 500 mK. The calibration is plotted out in Fig. 1.2.

1.4.3 Nuclear Orientation Thermometry

Spatial anisotropy of gamma-ray emission from a radioactive impurity in a metal host can be used to measure temperature if the orientation parameters are known. In such a case, absolute temperatures can be measured with a precision generally limited by statistical accuracy. Three different thermometers were used and their relative merits are discussed

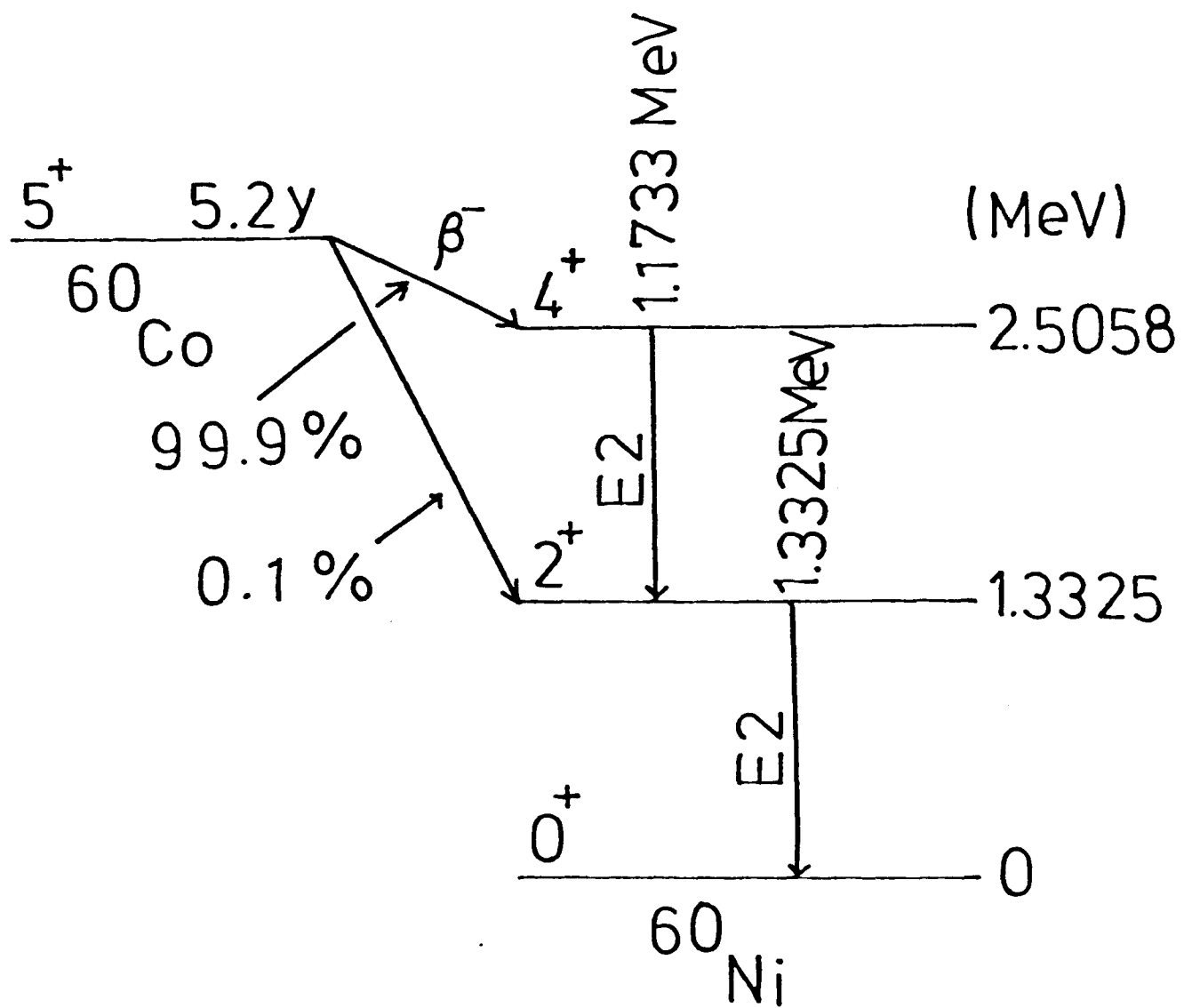
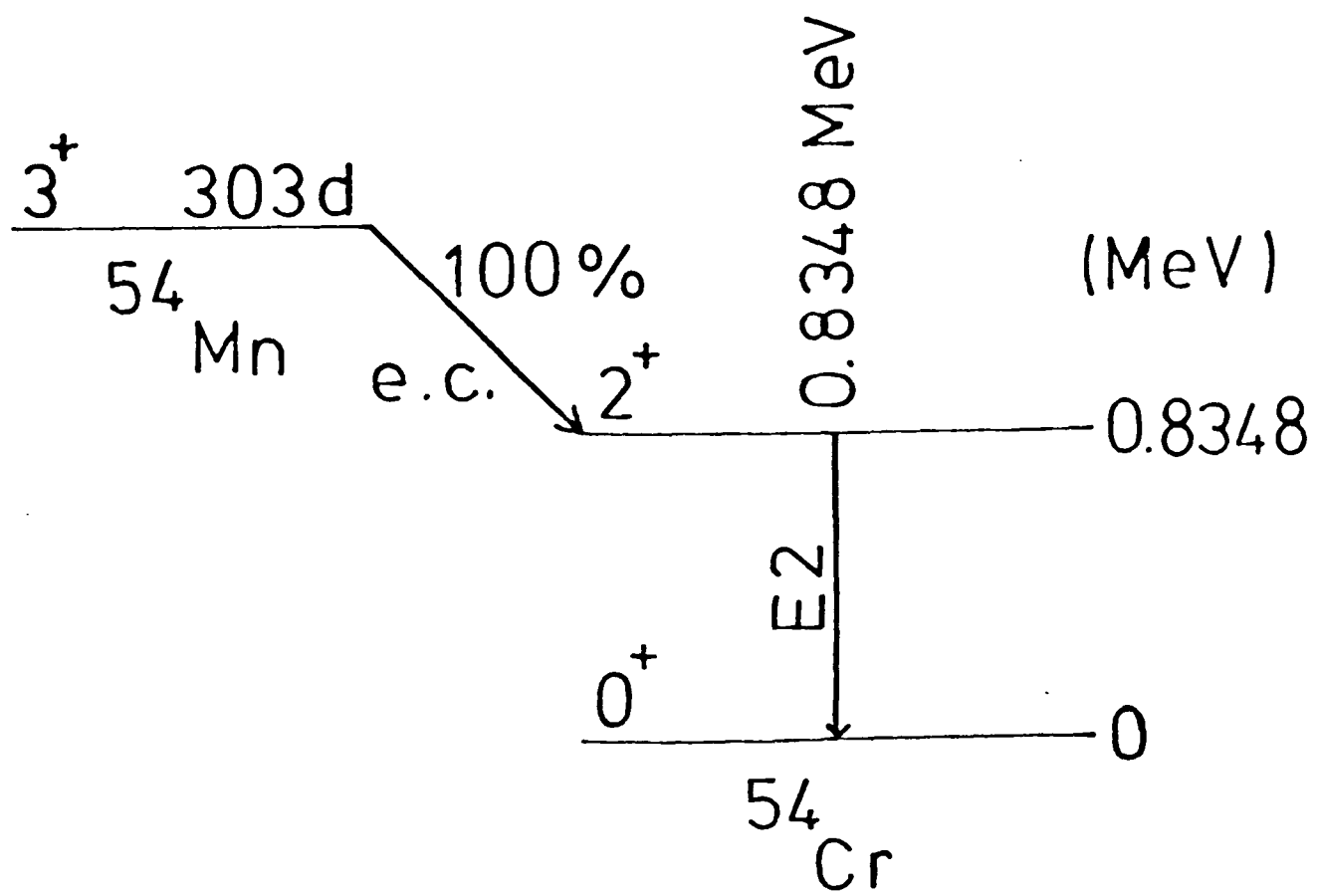


Fig. 1.3 Decay schemes of ^{54}Mn and ^{60}Co

below.

(a) ^{60}Co Co This was produced by neutron irradiation of a needle-shaped single crystal of h.c.p. cobalt. The decay scheme of ^{60}Co is shown in Fig. 1.3 (6). It has a nuclear dipole moment of $3.79\mu_N$ (7), a hyperfine field of -21.9 tesla in h.c.p. cobalt (8) and well known NO parameters. It has a single easy axis of magnetisation (c-axis) which means that it does not require a field to polarise the nuclei. It was therefore used in the susceptibility experiments where measurements had to be made in zero applied field (for example CMN calibration). However, cobalt has a large nuclear heat capacity peaking at about 10 mK which reduced the efficiency of some of the nuclear refrigeration experiments which used this thermometer.

(b) ^{60}Co Fe This was produced by thermal diffusion of ^{60}Co activity into a polycrystalline iron foil at about 1500°C for several hours. Although it has a negligible nuclear heat capacity at mK temperatures, a relatively large applied field of 0.5 tesla is required to saturate the magnetic domains. It could not therefore be used in the susceptibility measurements. Moreover, its use in the refrigeration experiments was limited by the large nuclear heat capacity of the copper cold finger in applied fields of 0.5 tesla. ^{60}Co in iron has a hyperfine field of -28.72 tesla (9). This is larger than that of ^{60}Co Co between 10 and 50 mK, but it does lose some sensitivity below about 5 mK.

(c) ^{54}Mn Cu This is a Kondo system below $T_K = 0.06 \pm 0.02\text{K}$ (10, 11) with a hyperfine field proportional to the applied field in low fields. It saturates in high fields to give a maximum field of 2.78 tesla (11). It was prepared by thermal diffusion for about ten hours at 800 °C. However, a sample

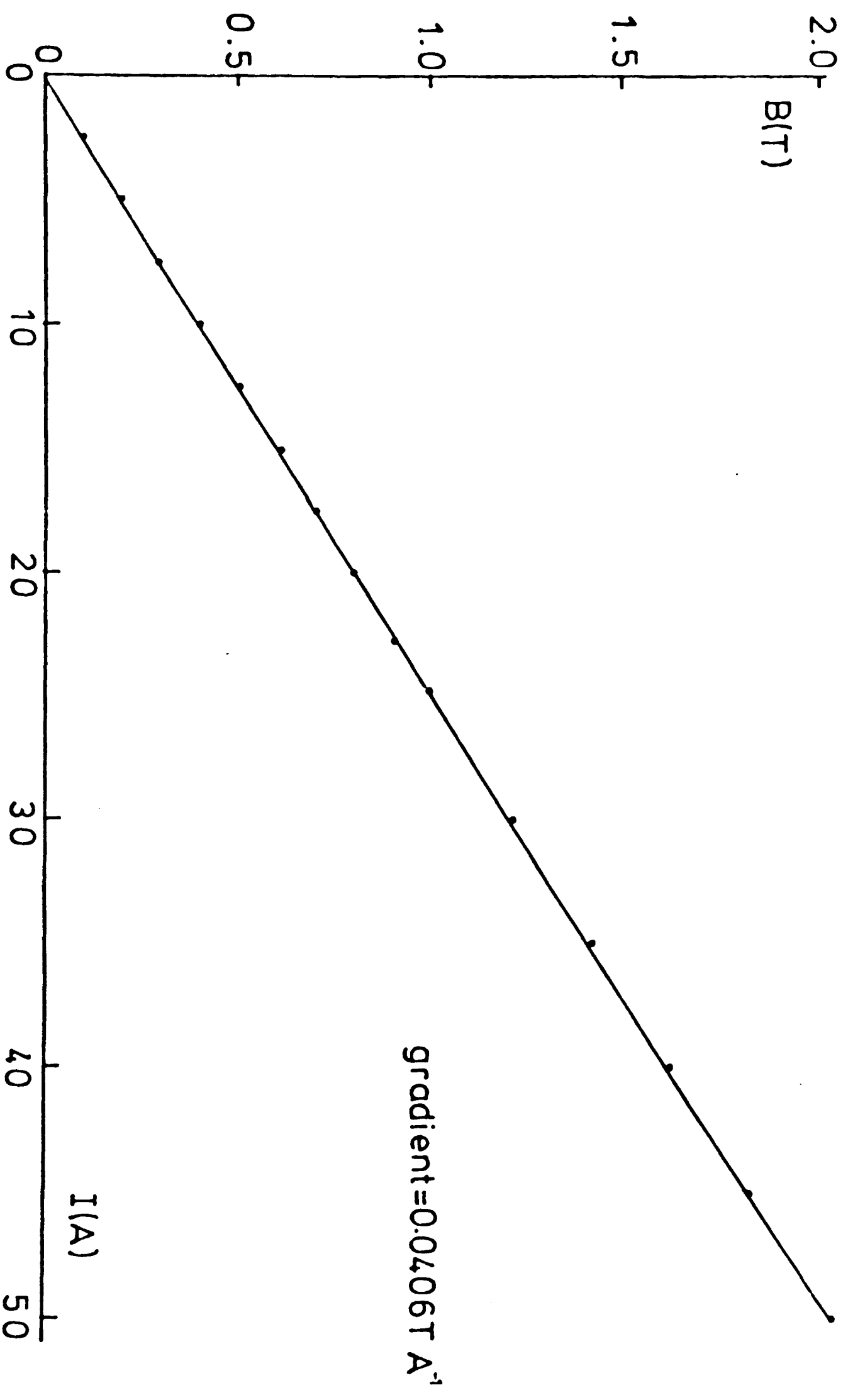


Fig. 1.4 Calibration of superconducting magnet 1

prepared in this way was found to be contaminated with about 200 ppm of iron which made the hyperfine field temperature dependent (11). The thermometer was calibrated against a ^{60}Co Co thermometer by D.W. Murray (see section 7.6.1). ^{54}Mn has a simple decay scheme (Fig. 1.3) and low radioactive heating (35 pW per μCi) (1) which makes it an ideal choice in nuclear refrigeration experiments.

1.5 Superconducting Magnets

Two superconducting magnets were used in the r.f. susceptibility and NO experiments. Both were driven by a constant current supply built in the Laboratory Electronic Workshop.

1.5.1 Magnet 1

This was designed and built in the Magnet Workshop of the Clarendon Laboratory by S. Hornung (2). It is a split Helmholtz coil with no field compensation. The windings are multistrand 33-gauge niobium-titanium wire. Fig. 1.4 is a calibration curve for the magnet, measured using a Hall probe of accuracy 1%. The maximum current used in these experiments was 60 A and the measured magnet constant was 0.0406(5)

T A^{-1} . It had a 'remanent' field after cycling of about $-2 \times 10^{-4}\text{T}$ in the central region. This low 'remanent' field made possible reliable low field measurements of r.f. susceptibility and gamma-ray anisotropy (Chapters 4 and 5). All the low field measurements were made using this magnet.

1.5.2 Magnet 2

This was built by R. Fox (12) also from niobium-titanium wire but had a separate field compensating coil. The magnet

produces a field of 2 tesla for 44 A through its windings with a homogeneity of about $\pm 1\%$ within $\pm 1/2$ cm of the centre. It was used in the experiments on nuclear refrigeration and the measurement of the nuclear moment of $^{166\text{m}}\text{Ho}$.

1.5.3 Heat Switch Magnet

This is a small uncompensated magnet built by D.W. Murray to operate the tin heat switch for nuclear refrigeration experiments (Chapter 8). It has a magnet constant of $3.15 \times 10^{-2} \text{ T A}^{-1}$. About 1 A was sufficient to drive the tin wires normal.

1.5.4 Magnetiser Magnet (SS1)

This was used to magnetise the $\text{HoVO}_4\text{-Cu}$ and $\text{HoVO}_4\text{-Au}$ pills in the nuclear refrigeration experiments. It produces a field of 0.0552 T A^{-1} with a 5% homogeneity over ± 2 cm of the centre.

1.6 The Gamma-Ray Detectors and Associated Electronics

The gamma-ray detectors were the Ge(Li) type and had a F.W.H.M. of about 2.3 keV and efficiency of about 15% at 1.3 MeV. Two such detectors were available mounted axially and equatorially respectively. They were manufactured by Philips and are fitted with a pre-amplifier and H.T. power supplies. The pulses are amplified using Canberra 2010 Spectroscopy Amplifiers and fed into a Nuclear Data 2200 Multichannel Analyser (MCA) with 4096 channel memory. The general layout is shown in Fig. 1.5. Where two gamma-ray detectors were used, the pulses were summed and stored in the MCA using a 'router' (see below).

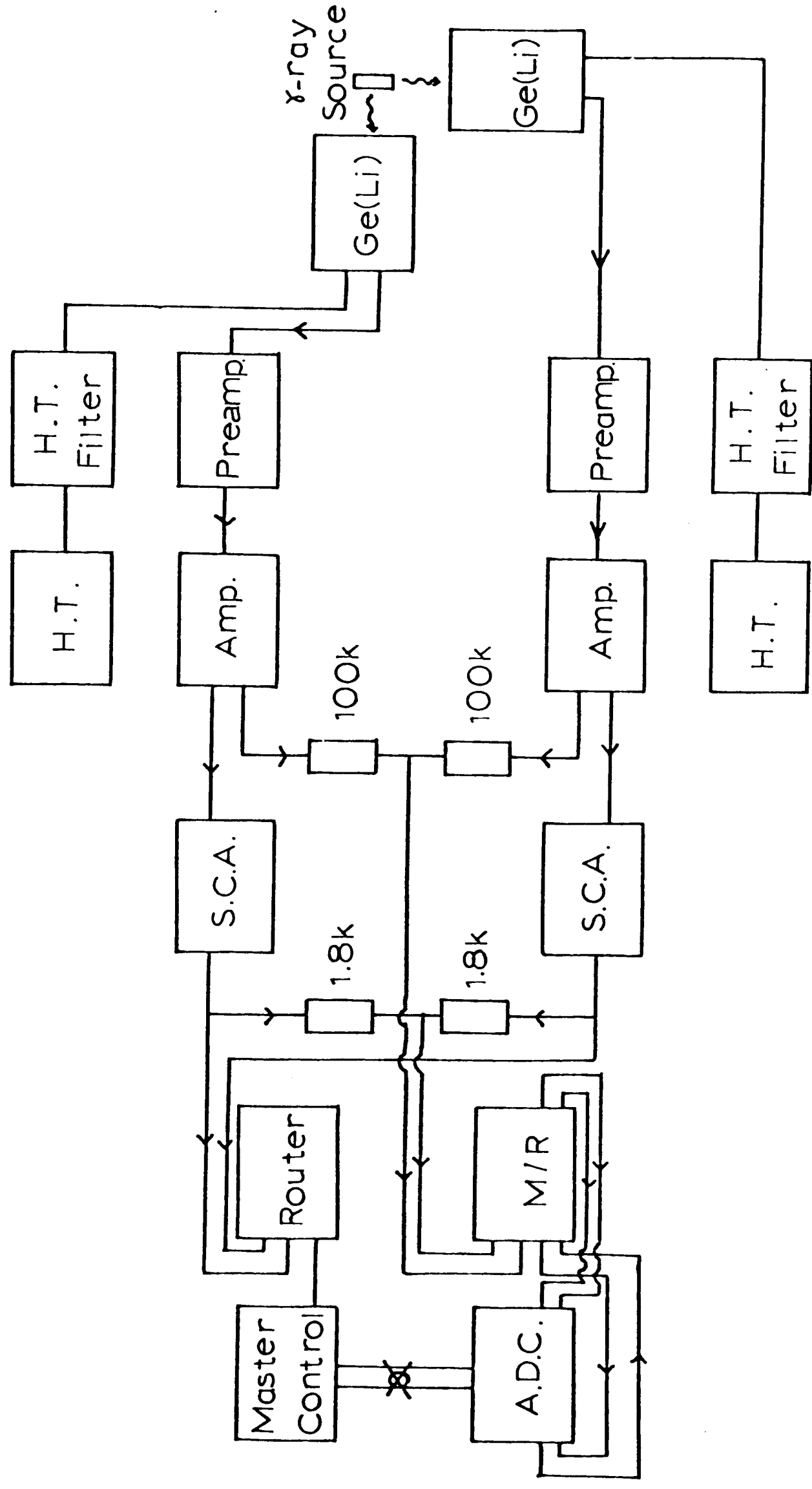


Fig. 1.5 Block diagram of the 'Router' and Ge(Li) detector electronics

1.6.1 The 'Router'

A system was built by S. Hornung (2) which made possible simultaneous storing of pulses from two detectors using the two halves of the same MCA. The main amplifier produces two pulses, a prompt bipolar and a delayed ($2\mu\text{s}$) monopolar. The monopolar pulses from the two amplifiers are summed using the 100 k Ω resistors (Fig. 1.5) and then fed through the Mixer/Rejector (M/R) into the Analogue to Digital Converter (ADC) of the MCA. The prompt bipolar pulses, however, are fed into two separate single channel analysers which produce square logic pulses. These logic pulses are fed into the 'router' which provides the necessary information for the MCA to allocate the gamma-ray pulses into the correct halves of the MCA. The logic pulses are also summed and fed into the M/R which prompts it to send coincidence pulses to the ADC only when the pulses do not overlap. This prevents overlapping gamma-rays from the two detectors from being analysed. The routing system is shown in Fig. 1.5.

Some deterioration of peak-width was observed using the 'router'. For this reason, only the axial detector was used in the nuclear moment determination of $^{166\text{m}}\text{Ho}$ (Chapter 6).

1.6.2 The Coincidence Correction

A small correction has to be made to take into account loss of counts due to coincidence of gamma-ray pulses from the two detectors when the router is used. If the pulses were to arrive within a time $\tau \sim 8\mu\text{s}$ both pulses would be rejected (see section 1.6.1). The loss in count rate from detector 1 is directly proportional to the count rate in detector 2; any reduction in count rate from one detector would increase the count rate from the other.

The count rate N_a from detector A is given by the expression

$$N_a = N_a^0 (1 - N_e \tau) \quad 1.5$$

where N_a^0 is the count rate in the absence of detector E and N_e is the count rate in detector E. The change in count rate in detector A, δN_a , due to a change in count rate in E, δN_e , is given by the expression

$$\delta N_a = -N_a^0 \tau \delta N_e \quad 1.6$$

The relative change in count rate in A is

$$\frac{\delta N_a}{N_a^0} = -\tau \delta N_e \quad 1.7$$

In the NO experiments of Chapter 5, the maximum change in count rate summed over a whole spectrum was ~ 20 counts s^{-1} which required a coincidence correction of only 0.02%. This negligible correction was ignored in the analysis of the NO data to be discussed in Chapter 5.

1.6.3 Storage of Data

In the earlier experiments, the data were punched on to paper tape. However, a new system was built by J. Siertsema of the Clarendon Laboratory Electronic Workshop which allowed the data from the MCA to be stored on magnetic tape using a NOVA 1200 computer. On readout, the data from the MCA are fed through a specially designed interface into another interface. From here the data are transferred into

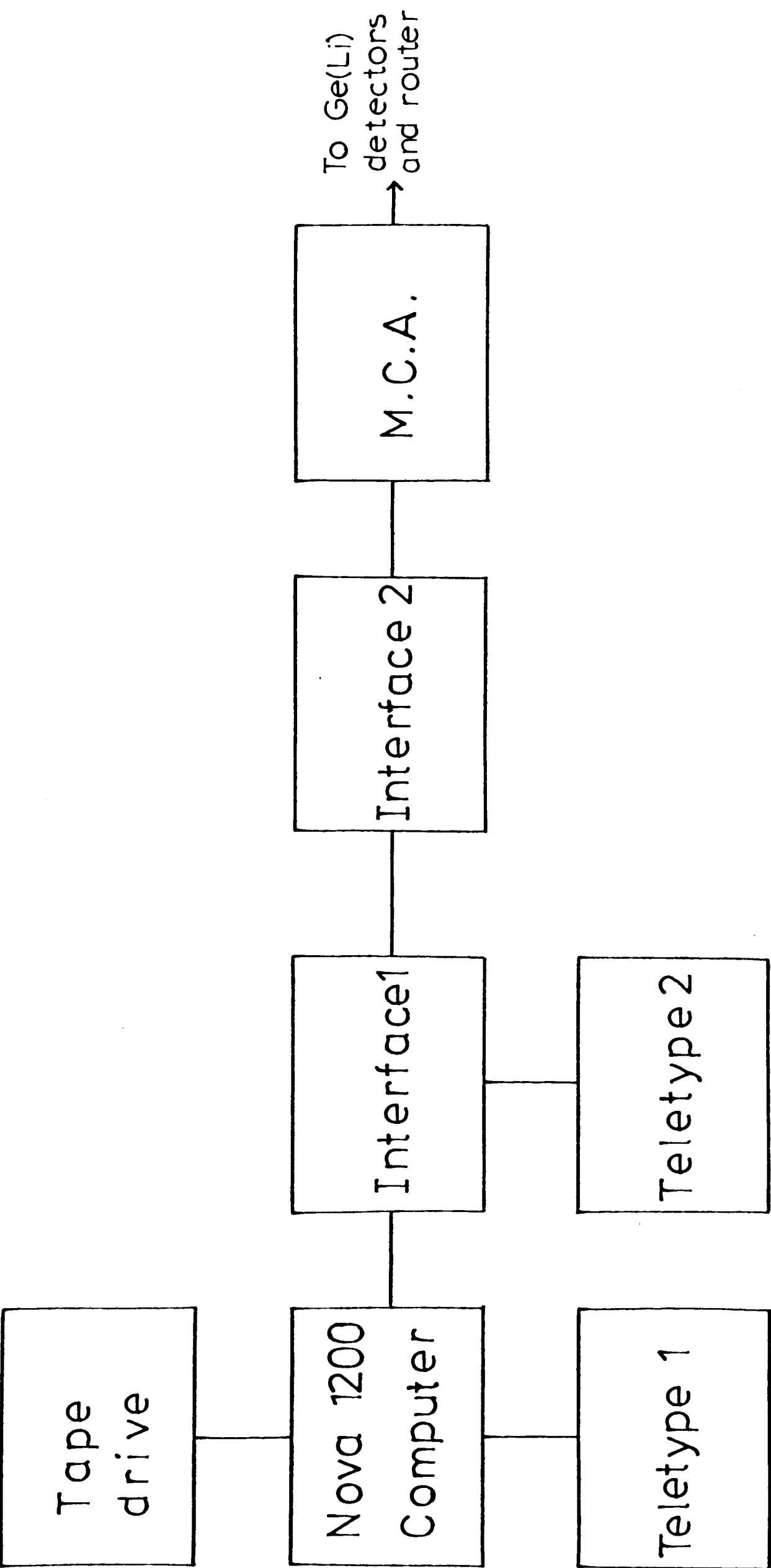
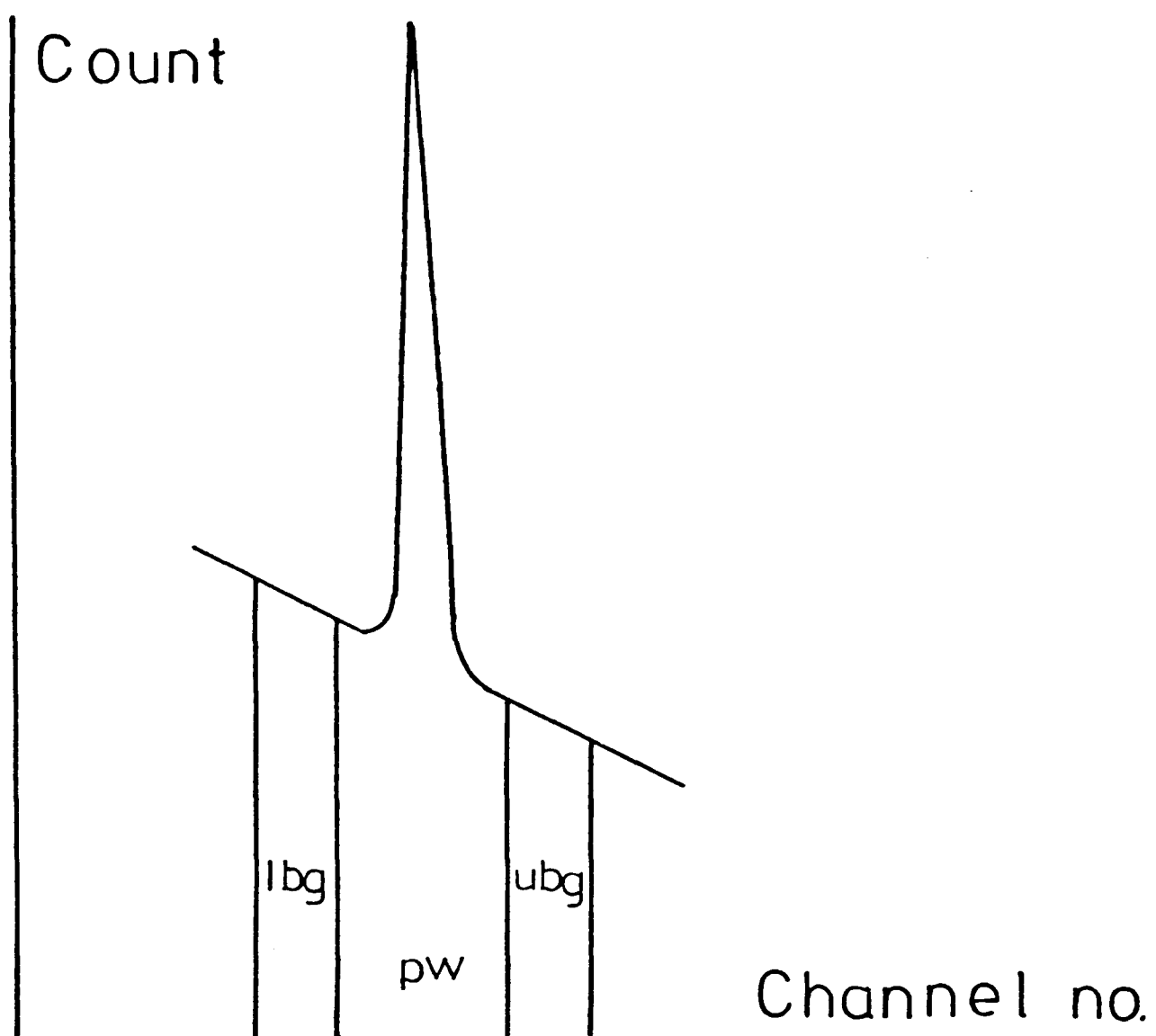


Fig. 1.6 Block diagram of the data storage system



lbg = lower background

pw = peak window

ubg = upper background

Fig. 1.7 Photo-peak 'window' and background settings

the computer memory. The computer then transfers the data on to a 9 track 800 b.p.i. tape in a tape drive in blocks of 256 channels. Using this system it is possible to acquire up to 90 spectra, each of 4096 channels in succession, the limit being set by the length of magnetic tape used. In addition, data can be analysed while the MCA is in the acquire mode using the Teletype link to the computer. The whole system is controlled by an 'Interface Program' (written by J. Siertsema) which is read into the computer. The system is shown in Fig. 1.6.

1.7 Data Analysis

The data stored on magnetic tape was analysed using a BASIC computer program developed by D.W. Murray (13). For the analysis of the gamma-ray spectra, a typical spectrum was printed out using a Teletype and the photo-peaks identified by calibrating their energies using well-known transitions. Peak and background 'windows' were then set for each peak and fed into the computer. The computer program summed the counts in the peak 'window' and subtracted the appropriate background counts by fitting a straight line through the background 'windows' (see Fig. 1.7). This was found to be sufficiently accurate for these experiments but care had to be taken to exclude background peaks from the 'window' settings. The program corrected for small shifts in peak positions by fitting a quadratic to the highest three channels and finding the maximum. The peak was then shifted by an appropriate fractional number of channels and the summation made for the new set of 'windows'. An interpolation procedure was used at the edge of the peak 'windows'.

The statistical error in an individual count, ξ , is

given by the expression

$$\xi = (N + B)^{1/2} \quad 1.8$$

where N and B are the counts in the peak and background 'windows' respectively. Where several counts were taken, an average was worked out and the statistical error was reduced by a factor

$$(\text{Number of counts})^{1/2}$$

The counts in the photo-peak were then divided by a 'warm' count for the same acquisition period from which anisotropies were evaluated

$$\text{Anisotropy (\%)} = \frac{(W_c - W_w)}{W_w} \times 100 \quad 1.9$$

c = cold
w = warm

'Cold' refers to the oriented state and 'warm' to the isotropic distribution. The interpretation of the gamma-ray anisotropies is discussed in Chapter 3.

1.8 Radio-frequency Co-axial Cable

An approximately 75 Ω impedance co-axial cable was constructed which made possible r.f. susceptibility measurements and Nuclear Magnetic Resonance on Oriented Nuclei (NMR/ON) attempts. The inner cable was constructed from 1mm German-silver tubing and was pushed into a 1/4 inch stainless steel tube fitted to the bottom of the 1 K pot. The inner tube was electrically isolated from the outer by means of PTFE

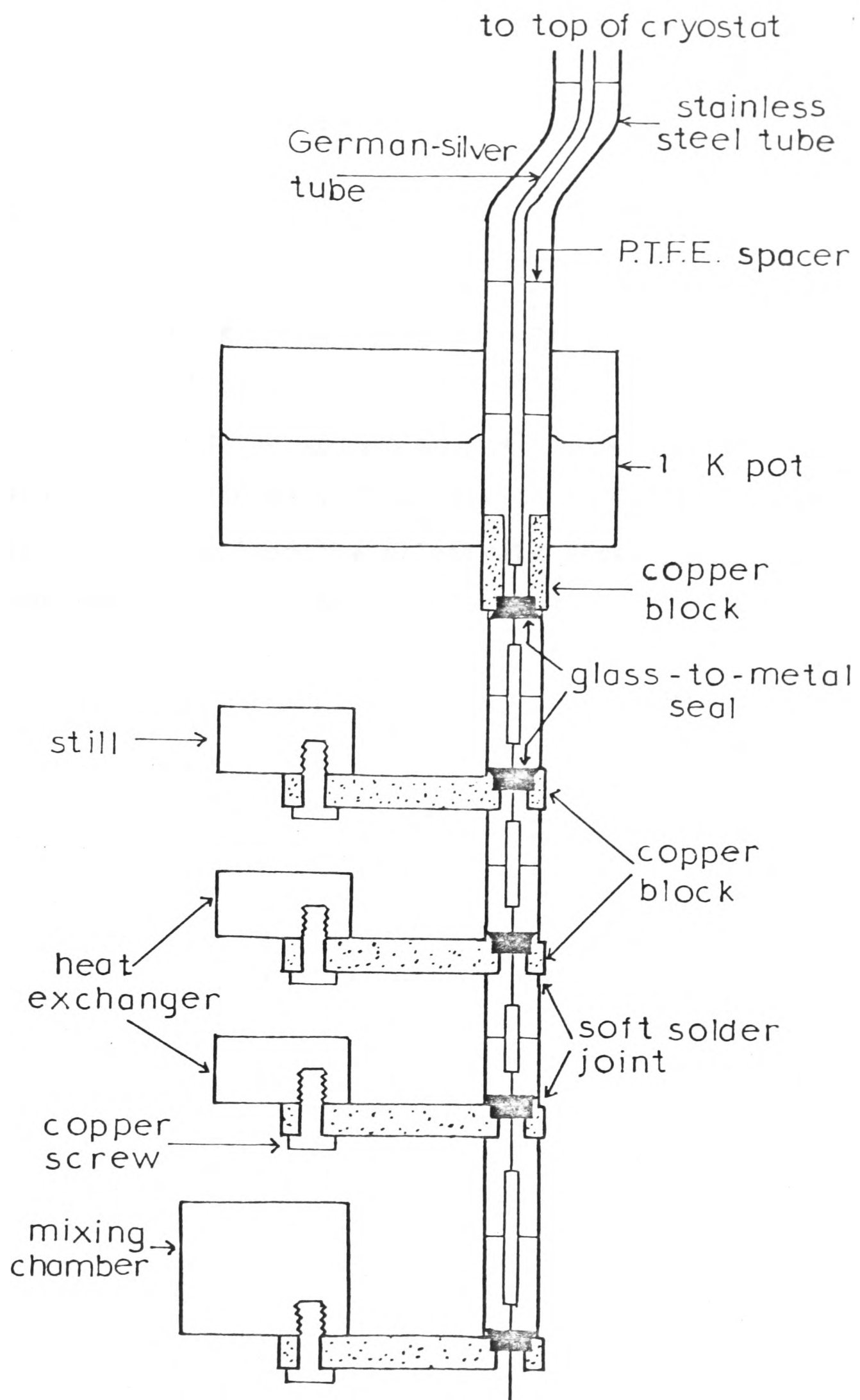


Fig. 1.8 Schematic diagram of the r.f. co-axial cable

spacers. Thermal contact between the inner and outer tubes was established by means of miniature glass-to-metal seals (supplied by Cathodeon Limited, Nuffield Road, Cambridge CB4 1TF). Good thermal anchoring was necessary to reduce the heat leak into the dilution unit. The cable was thermally anchored at 4 K, the 1 K pot, the still, two heat-exchangers and the mixing-chamber by means of copper blocks and glass-to-metal seals as shown in Fig. 1.8. This arrangement was found to produce negligible heat leaks into the dilution unit. For the NMR/ON experiments, a separate miniature co-axial cable was connected at the bottom of the 1 K pot and was taken outside the 1 K shield. This was to reduce heat input into the dilution unit from the relatively large resistive heating produced in the cables.

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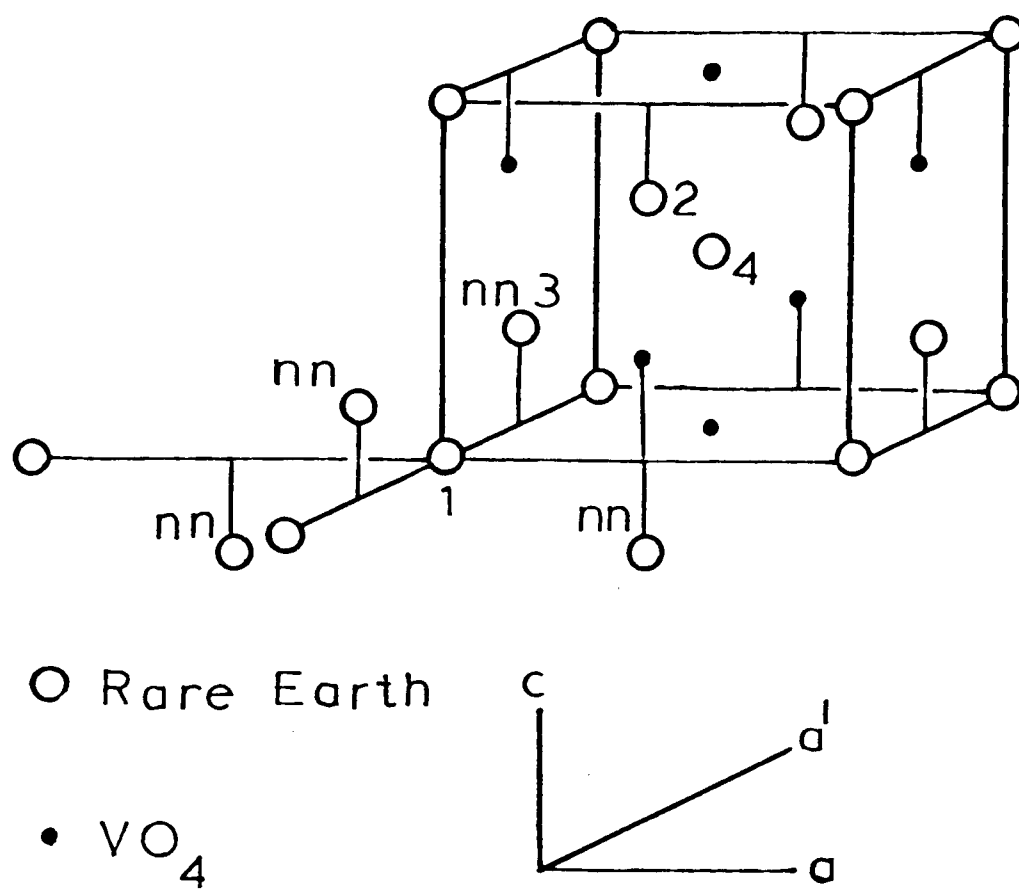
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CHAPTER 2

ENHANCED NUCLEAR MAGNETISM

IN HoVO_4 - THEORY



nn = nearest neighbour

Fig. 2.1 HoVO_4 crystal structure

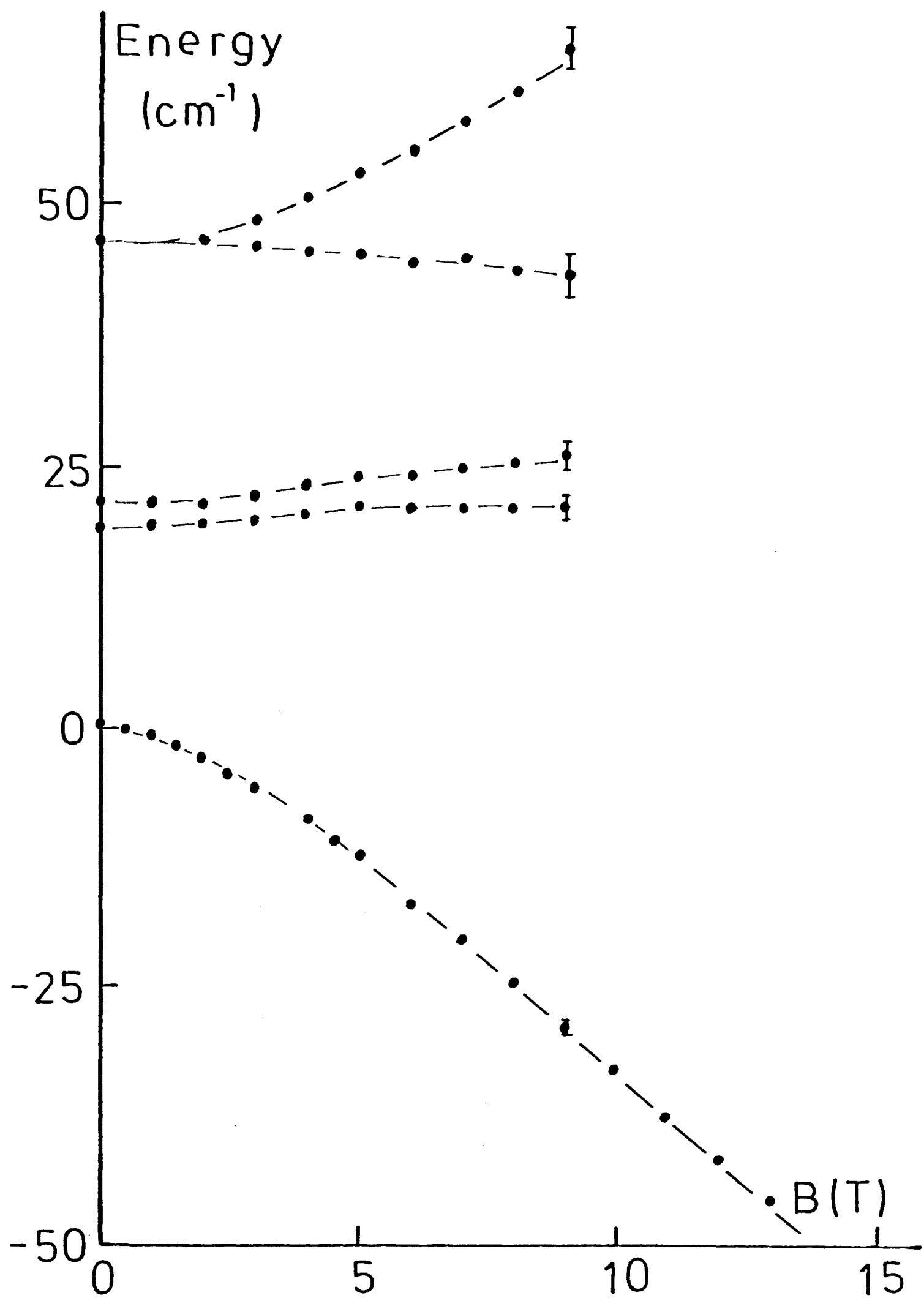


Fig. 2.2 HcVO₄ energy levels in an applied field B || a-axis

2.1 Introduction

Holmium vanadate, HoVO_4 , is one of a number of rare earth compounds with the formula RXO_4 where $X = \text{P}, \text{V}$ or As and R is a rare earth ion. Most of them have the tetragonal zircon structure with space group D_{4h}^{19} (1). There are four equivalent rare earth ions per unit cell which are shown in Fig. 2.1. Their positions are shown in Table 2.1.

Table 2.1

Lattice positions of rare earth ions in the non-primitive unit cell

<u>Rare Earth Ion</u>	<u>Lattice Position (a, a', c)</u>
1	(0,0,0)
2	($\frac{1}{2}, 0, \frac{3}{4}$)
3	(0, $\frac{1}{2}$, $\frac{1}{4}$)
4	($\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$)

At 25°C , HoVO_4 has the cell dimensions $a = 0.71214$ nm and $c = 0.62926$ nm (1).

The ground state of Ho^{3+} is $^5\text{I}_8$ which splits in the crystal field of tetragonal symmetry into nine singlets and four doublets. Optical spectroscopy by Battison et al (2) has shown that the ground state is a singlet with a doublet at 21 cm^{-1} and a singlet and doublet at 47 cm^{-1} . Fig. 2.2 shows how the lowest few levels split in a magnetic field applied parallel to the crystal a-axis. In zero field the ground state is mainly $|J_z = 0\rangle$ with small admixtures of $|J_z = \pm 4\rangle$ and $|J_z = \pm 8\rangle$. However, an applied field parallel to the a-axis mixes in $|J_z = \pm 1\rangle$ states into the ground state owing to the fact that the doublet at 21 cm^{-1} is mainly $|J_z = \pm 1\rangle$. This produces a second-order Zeeman splitting in the ground state (see below).

Magnetic susceptibility measurements by Swithenby (3) and Loftus (4) and Nuclear Magnetic Resonance (NMR) work by Bleaney et al (5) showed that the mixing of $|J_z = \pm 1\rangle$ into the singlet ground state produced a large 'enhancement' of the nuclear moment (~ 175) via the hyperfine interaction. This suggested the possibility of using HoVO_4 in nuclear adiabatic demagnetisation experiments at temperatures readily available using a dilution refrigerator. Although exchange effects between the nuclei are small (5), the large 'enhanced' nuclear moment was expected to cause nuclear ordering to occur in the mK instead of the μK range, predicted for 'ordinary' nuclear paramagnets.

2.2 The Nuclear Spin Hamiltonian

Bleaney et al (5) have discussed the nuclear Hamiltonian for Ho^{3+} in HoVO_4 . The following is a brief summary. In the crystal field approximation, the Hamiltonian for Ho^{3+} can be written as

$$\mathcal{H} = H_{\text{c.f.}} + g_J \mu_B \underline{B} \cdot \underline{J} - g_I \mu_n \underline{B} \cdot \underline{I} + A_J \underline{I} \cdot \underline{J} + H_Q \quad 2.1$$

The first term is the crystal field interaction. The other terms are the electronic Zeeman, nuclear Zeeman, nuclear magnetic hyperfine and electric quadrupole hyperfine interactions respectively. It is clear that for the singlet ground state the electronic Zeeman interaction vanishes in first order as $\langle J_z \rangle = 0$. Second order perturbation theory gives for the ground state, $|0\rangle$

$$\Delta E_2^O = \sum_{i \neq 0} \frac{\langle 0 | g_J \mu_B \underline{B} \cdot \underline{J} - g_I \mu_n \underline{B} \cdot \underline{I} + A_J \underline{I} \cdot \underline{J} | i \rangle \langle i | g_J \mu_B \underline{B} \cdot \underline{J} - g_I \mu_n \underline{B} \cdot \underline{I} + A_J \underline{I} \cdot \underline{J} | 0 \rangle}{(E_0 - E_i)} \quad 2.2$$

where ΔE_2^O refers to the second order shift in energy, and the summation is over all the excited states $|i\rangle$ in the ground manifold J . This gives a quadratic Zeeman interaction in the x (a or a' crystal axes) direction given by

$$(\Delta E_2^O)_x^{\text{electronic Zeeman}} = -B_x^2 g_J^2 \mu_B^2 \sum_{i \neq 0} \frac{\langle 0 | J_x | i \rangle \langle i | J_x | 0 \rangle}{(E_i - E_0)} \quad 2.3$$

As J_x mixes in states of $|J_z = \pm 1\rangle$ into the ground state, the second order electronic Zeeman shift will not be zero. Equation 2.3 can be rewritten as

$$(\Delta E_2^O)_x^{\text{electronic Zeeman}} = \frac{-B_x^2}{2} a_x g_J^2 \mu_B^2 \quad 2.4$$

$$\text{where } a_x = 2 \sum_{i \neq 0} \frac{\langle 0 | J_x | i \rangle \langle i | J_x | 0 \rangle}{(E_i - E_0)} \quad 2.5$$

This gives a 'Van-Vleck' electronic susceptibility χ_{VV} at low temperatures given by

$$\chi_{VV} = a_x g_J^2 \mu_B^2 \quad 2.6$$

This has the constant value of $2.32 \mu_B$ /tesla below about 1K (5). Cross terms between the electronic Zeeman and hyperfine interactions give rise to an 'enhanced' nuclear Zeeman term in second order, given by the expression

$$(\mathcal{H})_x^{\text{nuclear Zeeman}} = -(a_x g_I \mu_B A_J + g_I \mu_n) B_x I_x \quad 2.7$$

for an applied field in the x-direction. Equation 2.7 can be rewritten to give

$$(\mathcal{H})_x^{\text{nuclear Zeeman}} = - (K + 1) g_I \mu_n B_x I_x \quad 2.8$$

$$\text{where } K, \text{ the 'enhancement factor'} = \frac{a_x g_J \mu_B A_J}{g_I \mu_n}$$

For HoVO_4 $K \sim 175$ and is independent of the isotope of holmium used. A similar term can also be written for the y direction. As J_z has no matrix element between $|J_z = 0\rangle$ and $|J_z = \pm 1\rangle$, the nuclear Zeeman term in the z direction is expected to be almost unenhanced. In fact Bleaney et al (5) estimated an enhancement factor $K \sim 3$ in the z direction. In NMR work it is convenient to write Equation 2.8 in the form

$$(\mathcal{H})_x^{\text{nuclear Zeeman}} = - \gamma_{\perp} \hbar B_x I_x \quad 2.9a$$

where $\gamma_{\perp}$ refers to the plane perpendicular to the c-axis and

$$\gamma_{\perp} = (1 + K) g_I \mu_n / \hbar \quad 2.9b$$

The full nuclear Hamiltonian in a field is given by

$$\mathcal{H}_n = - \gamma_{\parallel} \hbar B_z I_z - \gamma_{\perp} \hbar (B_x I_x + B_y I_y) + P [I_z^2 - \frac{1}{3} I(I+1)] \quad 2.10$$

where the electric quadrupole interaction has axial symmetry about the z-axis. Equation 2.10 can be rewritten to give

$$\mathcal{H}_n = - \gamma_{\parallel} \hbar B_x I_x - \gamma_{\perp} \hbar (B_y I_y + B_z I_z) - \frac{P}{2} [I_z^2 - \frac{1}{3} I(I+1)] + \frac{P}{4} (I_+^2 + I_-^2) \quad 2.11$$

where now the x-axis defines the quadrupole direction. It

will be found convenient to use Equation 2.11 in some of the NO work to be discussed in Chapter 5.

NMR measurements on ^{165}Ho (5) gave

$$(\chi_{\perp}/2\pi) = 1526 (3) \text{ MHz T}^{-1}$$

$$|P| = 25.9 (3) \text{ MHz}$$

$$\text{and } (\chi_{\parallel}/\chi_{\perp}) \sim 10^{-2}$$

2.3 The 'Enhancement Factor' in Applied Fields

As the applied field along an 'enhancement' direction is increased, the induced electronic moment will start to saturate. This causes the 'enhancement factor' to decrease with applied field. Bleaney et al (6) have shown from their r.f. susceptibility measurements that

$$K_{\chi}(B) = K_{\chi}^0 (1 + 0.0466 B^2)^{-\frac{1}{2}} \quad 2.12$$

where $K_{\chi}(B)$ and K_{χ}^0 are the 'enhancement factors' in an applied field B (tesla) and zero field respectively. Table 2.2 shows the relative change of $K_{\chi}(B)$ for various applied fields.

Table 2.2

Calculated $K_{\chi}(B)/K_{\chi}^0$ as a function of applied field

B (tesla)	$K_{\chi}(B)/K_{\chi}^0$ ($= \chi_{\perp}^B / \chi_{\perp}^B = 0$)
0	1
0.5	0.994
1.0	0.978
1.5	0.951
2.0	0.918

The nuclear Hamiltonian (Equation 2.10) can be used in fields

of up to 2 Tesla by putting in the corrected value of $\gamma_{\perp}^B$ from Table 2.2. As this correction to $\gamma_{\perp}$ is small below about 0.1 tesla, the 'zero field' value was used throughout the NO studies of Chapter 5. But in the nuclear moment measurement of ^{166m}Ho , where large applied fields were used, Table 2.2 was used to correct for this effect.

2.4 The Nuclear Electric Quadrupole Interaction

In HoVO_4 the nuclear electric quadrupole interaction is the sum of three terms.

2.4.1 The Pseudo-Quadrupole Term

This arises from the cross term

$$(A_J \underline{I} \cdot \underline{J})^2$$

of Equation 2.2 which gives rise to a pseudo-quadrupole term

$$P_{pq} = \frac{A_J^2 a_x}{2} \quad 2.13$$

As A_J is proportional to the nuclear g factor, Equation 2.13 implies that the pseudo-quadrupole term scales as the square of the nuclear g factor. For ^{165}Ho Bleaney et al (5) estimate $^{165}P_{pq} = + 35.6 \text{ MHz}$. Taking $I = 7/2$, $g_I = (4.125/3.5)$ for ^{165}Ho (8) and $I = 7$, $g_I = (3.60/7)$ for ^{166m}Ho (see Chapter 6) the pseudo-quadrupole term for ^{166m}Ho is estimated to be

$$^{166m}P_{pq} = + 6.8 (1) \text{ MHz}$$

2.4.2 The Electric Field Gradient (e.f.g.) due to the 4f Electrons

The 4f electrons from the Ho^{3+} ion generate an e.f.g. which produces a quadrupole interaction term P_{4f} given by the

equation (9)

$$P_{4f} = \frac{-3e^2 Q \langle r^{-3} \rangle}{4I (2I-1)} \langle J \parallel \alpha \parallel J \rangle \langle 3J_z^2 - J(J+1) \rangle \quad 2.14$$

where Q and I are respectively the electric quadrupole moment and spin of the nucleus and $\langle J \parallel \alpha \parallel J \rangle$ is a reduced matrix element.

2.4.3 The e.f.q. due to the Lattice

The electric quadrupole interaction due to the lattice scales with

$$Q/I(2I-1)$$

Thus both the lattice and 4f electrons produce a quadrupole splitting which scales the same way. This enables us to compute the total electric quadrupole splitting for ^{166m}Ho from ^{165}Ho given that the spins and moments of the two isotopes are known.

The total quadrupole splitting P ($=P_{\text{lattice}} + P_{4f} + P_{\text{pq}}$) for ^{165}Ho is equal to ± 25.9 (3) MHz while the pseudo-quadrupole term $^{165}P_{\text{pq}} = + 35.6$ MHz. This gives for the sum of the 4f and lattice contributions.

$$^{165}P_{4f} + ^{165}P_{\text{lattice}} = -9.7 \text{ or } -61.5 \text{ MHz}$$

for positive and negative sign of ^{165}P respectively.

Assuming that $^{166m}Q = + 5.3$ (3) barns (see section 6.8)

and $^{165}Q = 3.5$ barns (10) we obtain

$$^{166m}P_{4f} + ^{166m}P_{\text{lattice}} = -3.4$$
 (2) or - 21.6 (12) MHz

for positive and negative sign of ^{165}P respectively. Thus

$$^{166}\text{m}_\text{P} = ^{166}\text{m}_\text{P}_{\text{pq}} + ^{166}\text{m}_\text{P}_{4\text{f}} + ^{166}\text{m}_\text{P}_{\text{lattice}} = + 3.4 (3) \text{ MHz}$$

$$\text{or } - 14.8 (13) \text{ MHz.}$$

NO measurements (Chapter 5) showed unambiguously that $^{166}\text{m}_\text{P}$ is positive. We therefore adopt the positive value

$$^{166}\text{m}_\text{P} = + 3.4 \text{ MHz}$$

which is the value used throughout the thesis. This means that

$$^{165}\text{P} = + 25.9 \text{ MHz}$$

Strictly speaking, both P_{pq} and $\text{P}_{4\text{f}}$ are field dependent. However, in low applied fields ($B < 0.1\text{T}$) the effect is small and so the low field values were used in the NO measurements of Chapter 5. In high fields P is but a small perturbation on top of the nuclear Zeeman term. Thus the quadrupole term can be taken to be constant to a good approximation in all fields.

2.5 The Local Field at the Ho^{3+} Site

Bleaney et al (5) have shown that the field acting at the rare earth site has to be corrected for the Lorentz, demagnetisation, local dipole and exchange field effects. The total field at an ion, B_i , is given by the equation

$$B_i = B_i^0 + \left[\mu_0 \left[\left(\frac{1}{3} - d \right) \frac{N\mu_B}{V} \right] D_d + D_{\text{exch.}} \right] \frac{m}{\mu_B} \quad 2.15$$

where B_i^0 is the applied field, d the demagnetisation factor, N/V the number of ions per unit volume, m the total magnetic moment in Bohr Magnetons, D_d the local dipolar field summed over a spherical volume for a moment of one μ_B and $D_{\text{exch.}}$ a

term to include the effect of the electronic exchange.

Equation 2.15 can be written in the form

$$B_i = B_i^0 (1 - f_s \chi)^{-1} \quad 2.16$$

where f_s is a semi-empirical constant determined for a particular sample from NMR measurements and χ is the total susceptibility. At low temperatures χ must include both the electronic and nuclear susceptibilities. The values of f_s determined from NMR at 4.2K for the samples used in this thesis are discussed in Section 4.2.

2.6 Predictions of Nuclear Ordering in HoVO₄

Bleaney (7) has discussed in detail the possible nuclear ordered structures in HoVO₄. The following is a brief account of the main ideas propounded therein.

2.6.1 Nuclear Ferromagnetism

The measured susceptibility is given by the expression

$$\chi_{\text{meas.}} = M/B_0 \quad 2.17$$

where B_0 is the applied field and M is the induced moment.

From Equations 2.16 and 2.17 it follows that

$$\chi^{-1} = \chi_{\text{meas.}}^{-1} + f_s \quad 2.18$$

where χ is the total 'intrinsic' susceptibility (= nuclear + electronic contributions). It must be emphasised that Equation 2.18 is quite general and does not assume any particular temperature dependence for χ . For ferromagnetic ordering

$$\chi_{\text{meas.}}^{-1} = 0 \quad \text{so that}$$

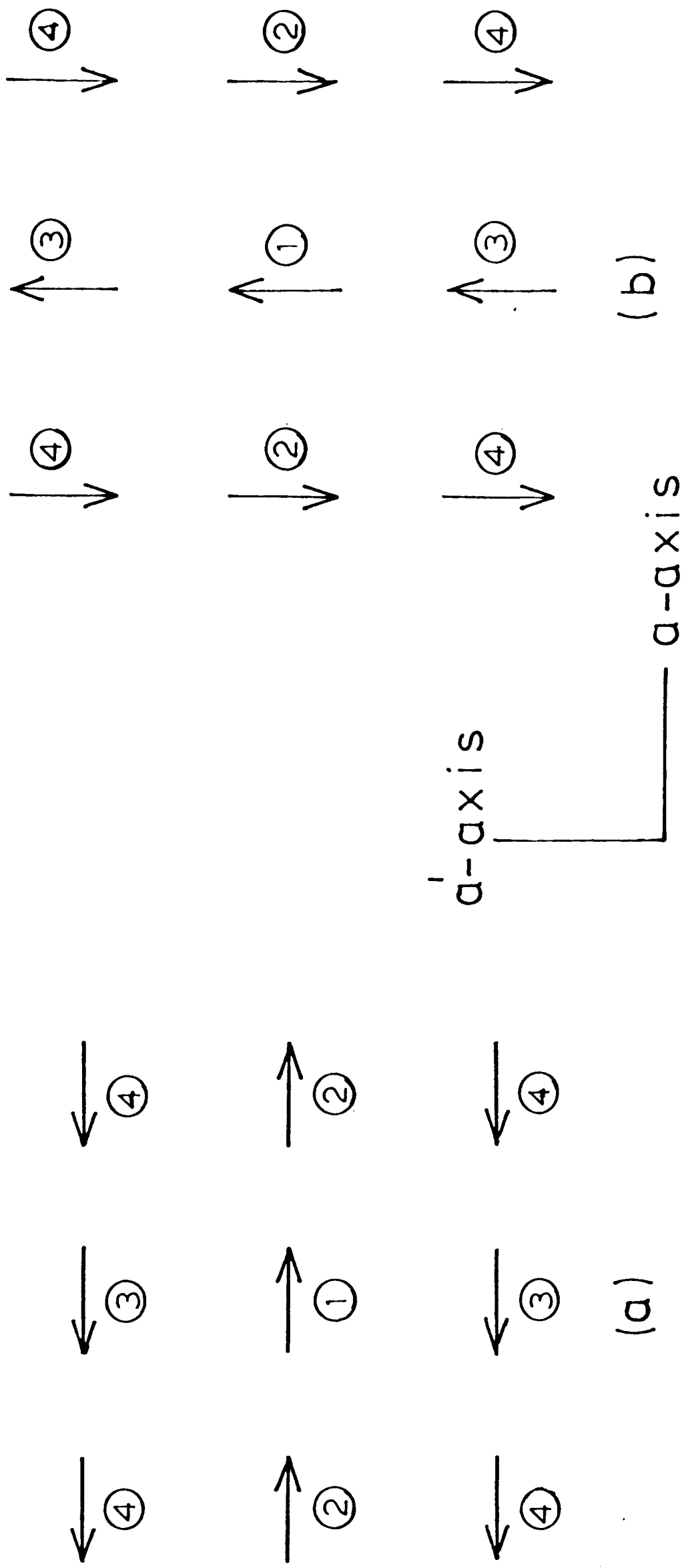


Fig. 2.3 Two equivalent nuclear antiferromagnetic arrangements in HVO_4

$$\chi^{-1} = f_s \quad 2.19$$

From Equation 2.18 f_s is a function of sample shape and for one of the samples used $f_s = 0.02$ which corresponds to $T_c \sim 1\text{mK}$.

2.6.2 Nuclear Antiferromagnetism

In HoVO_4 , the dominant interaction between the Ho^{3+} ions is dipole-dipole with some exchange via the O^{2-} ions. As the enhancement is restricted to the a - a' plane, we shall consider nuclear ordering in this plane only.

Fig. 2.1 shows the four nearest neighbours (nn) for ion 1. Although they do not lie in the same plane it is quite convenient to draw them to be so. Fig. 2.3 shows two equivalent antiferromagnetic arrangements which produce a large positive field at site 1. In order to compute this field it is necessary to do a dipole sum for each lattice. It is clear from Fig. 2.3 that lattice 2 in (a) produces the same field on site 1 as lattice 3 in (b). Thus there are only four different sums. These are listed in Table 2.3 for a moment of $1\mu_B$ at each site, where e denotes 'end on' position and b the 'broadside on' position.

Table 2.3

Dipolar sum for $1\mu_B$ at each site

Site		Dipole field ($T\mu_B^{-1}$)
1	a	$\frac{1}{4}B_1 - 0.0024$
4	d	$\frac{1}{4}B_1 + 0.0020$
2 or 3	{ e	$\frac{1}{4}B_1 + 0.0442$
	{ b	$\frac{1}{4}B_1 - 0.0336$

$B_1 = + 0.0436 \text{ tesla} \mu_B^{-1}$ is the Lorentz field.

For the arrangements (a) and (b), the total (antiferromagnetic) field $B_{AF} = a-d+e-b$. This gives a field of $0.0734 \text{ T} \mu_B^{-1}$ for the arrangement of Fig. 2.3. Exchange interaction with the four nn does not alter this field because two nn are parallel and two nn are anti-parallel. All other antiferromagnetic arrangements can be shown to be energetically unfavourable.

For the ferromagnetic case, the field at the ion B_F is given by

$$B_F = a+d+e+b = +0.0588 \text{ T} \mu_B^{-1} \quad 2.20$$

Inclusion of exchange effects reduces B_F to $0.0388 \text{ T} \mu_B^{-1}$. Thus in zero applied field the antiferromagnetic arrangement of Fig. 2.3 is the energetically favoured configuration.

2.6.3 The Néel Temperature T_N

The total magnetisation is given by the equation

$$M = (\chi_n(T) + \chi_e) \mu_B B \quad 2.21$$

Where χ is in $\mu_B \text{ T}^{-1}$. For the antiferromagnetic arrangement of spins, the field on the ion, B_i , is given by

$$B_i = 0.0734 M / \mu_B \quad 2.22$$

At the ordering temperature,

$$\frac{M}{\mu_B} = (\chi_n(T) + \chi_e) \frac{0.0734 M}{\mu_B} \quad 2.23$$

from which

$$(0.0734)^{-1} = \chi_n(T) + \chi_e \quad 2.24$$

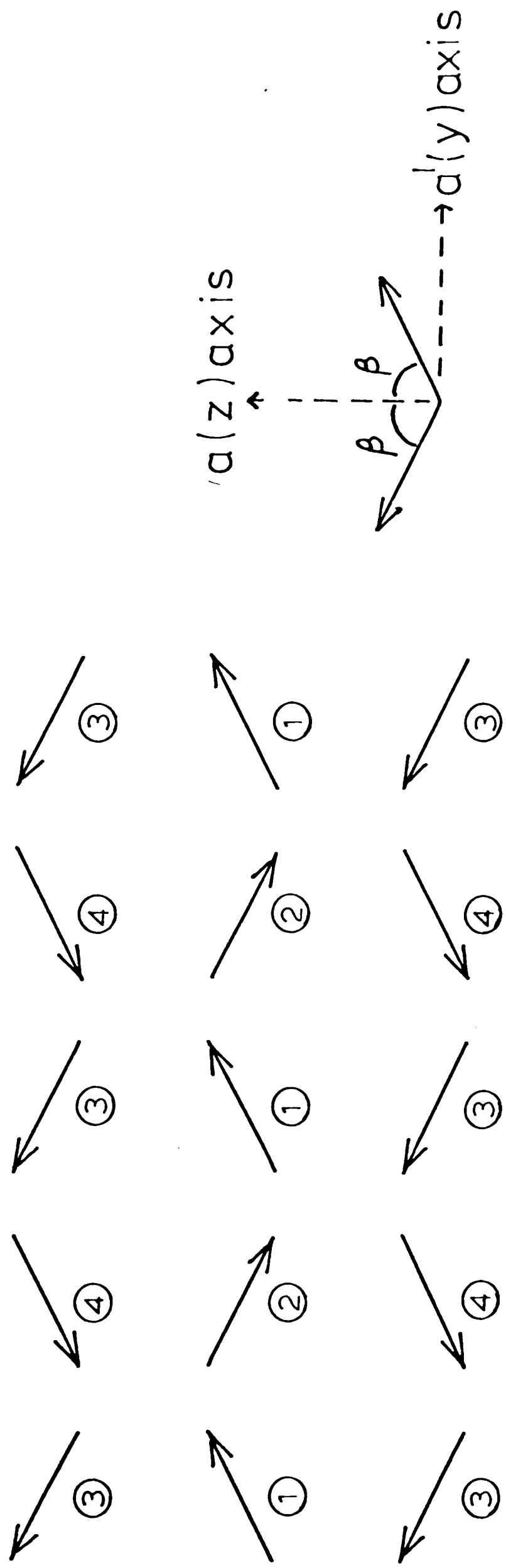


Fig. 2.4 'Puckered' spin structures in nuclear antiferromagnet HcVO_4

T_N is then that temperature at which Equation 2.24 is satisfied. By computing the nuclear susceptibility $\chi_n(T)$ as a function of temperature and using Equation 2.24, T_N was calculated to be 4.8 mK. As explained in section 2.6.2, exchange interaction due to the four nn cancels out for the antiferromagnetic arrangements of Fig. 2.3. This is also true for the four next nearest neighbours (nnn). However, the eight next nearest neighbours (nnnn) produce a net exchange field at the nucleus. Bleaney (7) estimates that this would reduce the Néel temperature from 4.8 to 4.6 mK.

2.6.4 'Puckered' Spin Structures

Bleaney (7) has also discussed some of the 'puckered' or 'canted' spin structures in the a-a' plane. These are antiferromagnetic in nature, 'canted' at an angle β to the a or a' axis with no net magnetisation in zero field (Fig. 2.4). It turns out from dipole energy considerations that in zero field all such 'puckered' spin structures are equally likely. It can be shown that exchange interaction with the four nn does not alter this conclusion. This means that in zero field the antiferromagnetic domains are uniformly distributed in the a-a' plane, given that there is no magnetic anisotropy in this plane.

However, on application of a small field the spin 'flip' structure is energetically favoured, in the absence of any anisotropy in the a-a' plane. As the applied field is increased, the spins will rotate until in an applied field of $\sim 0.016T$ they are all parallel. They are then in the ferromagnetic or 'flip' phase. The effect these structures have on the γ -ray anisotropy below T_N is discussed in Chapter 5.

2.7 Some Calculations of the Nuclear System

2.7.1 Nuclear Entropy and Susceptibility Calculations

The nuclear Hamiltonian (Equation 2.9) was used to calculate the entropy and the magnetic susceptibility of the nuclear system. A computer program was written in collaboration with G.J. Bowden to enable these calculations to be made. The programme was written in FORTRAN and run on the University 1906A ICL computer. The matrix elements for the nuclear Hamiltonian were first computed. Diagonalisation of the eight by eight matrix gave the nuclear energy levels. The entropy per ion, S , was calculated using the equation

$$S = -k \text{Tr} [\rho \ln \rho] \quad 2.25$$

where ρ , the density matrix is given by the expression

$$\rho = e^{-\beta \mathcal{H}} / \text{Tr} [e^{-\beta \mathcal{H}}] \quad 2.26$$

and Tr denotes a trace. The isothermal susceptibility in the direction x , χ_T^x is given by the expression

$$\chi_T^x = \beta (\gamma_L \hbar)^2 \left\{ \langle I_x \{ I_x \} \rangle - \langle I_x \rangle \langle I_x \rangle \right\} \quad 2.27$$

where

$$\langle I_x \{ I_x \} \rangle = \sum_i \left\{ \rho_{ii} \sum_{i'} I_x(i i') I_x(i' i) + \beta^{-1} \sum_{\substack{k \\ E_k \neq E_i}} \frac{(\rho_{ii} - \rho_{kk}) I_x(i i') I_x(i' k)}{E_k - E_i} \right\} \quad 2.28$$

with a similar expression for the y direction. The matrix element $I_x(i i)$ refers to the representation in which $\mathcal{H}$ is diagonal. This means that $I_x(i i)$ must be calculated from the Unitary transformation which diagonalises the Hamiltonian of Equation 2.10. Thus if V represents the eigenvectors of $\mathcal{H}$

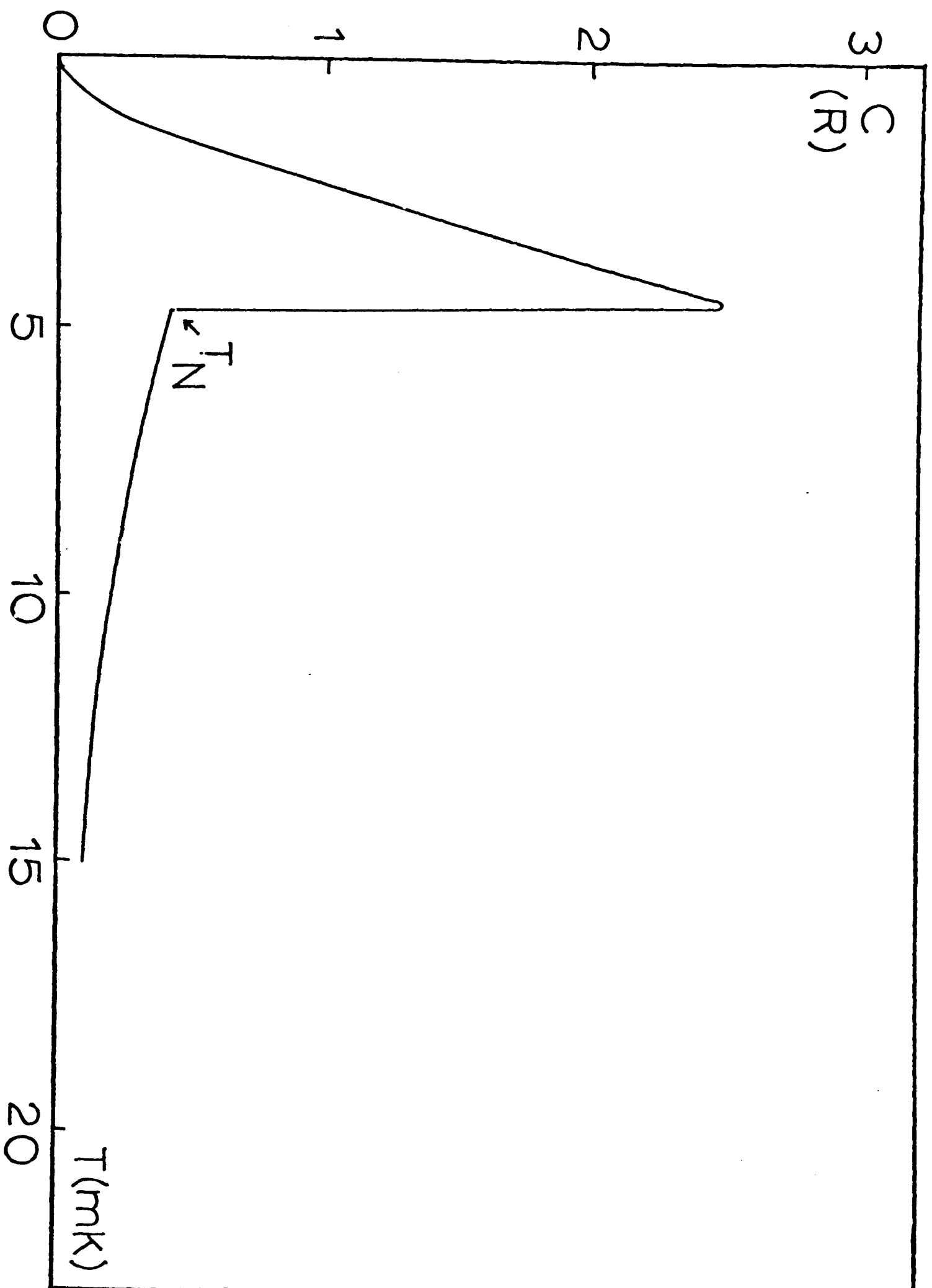


Fig. 2.5 Nuclear heat capacity of HoVO_4 at low temperatures

$$I_{x(ik)} = \sum_j V_{ij}^+ V_{jk} \langle i | I_x | k \rangle \quad 2.29$$

where the matrix element $\langle i | I_x | k \rangle$ refers to the Zeeman representation.

The adiabatic susceptibility in the x-direction is given by the expression

$$\chi_S^x = \chi_T^x - \beta (\gamma \hbar)^2 \frac{[\langle I_x \rangle \langle H \rangle - \langle I_x \rangle \langle H \rangle][\langle H I_x \rangle - \langle I_x \rangle \langle H \rangle]}{\langle H^2 \rangle - \langle H \rangle^2} \quad 2.30$$

where $\langle I_x \rangle \langle H \rangle$ is given by an expression similar to Equation 2.28. In zero applied field χ_S is identically equal to χ_T .

2.7.2 Nuclear Specific Heat

The nuclear specific heat at constant field C_B was computed from the expression

$$C_B = T \frac{\Delta S}{\Delta T} \quad 2.31$$

The computed specific heat curve is shown in Fig. 2.5, calculated using Equation 2.31 for $\Delta T/T = 0.01$. Below the Néel temperature a 'self-consistent' mean field calculation was used (see below). The discontinuity in the specific heat near T_N is typical of magnetic ordering in a mean field approximation. The high temperature 'tail' is due to the large quadrupole specific heat which is normally absent in simple ordering systems.

2.7.3 'Self-Consistent' Mean Field Calculations

Although the analysis of section 2.6.3 gave the Néel temperature T_N , a computer calculation was made to evaluate some of the magnetic properties of the system below the ordering temperature.

Fig. 2.4 shows a general canted structure of the nuclear spins below T_N . The crystal axes a' and a were arbitrarily taken to be in the y and z directions respectively with the quadrupole interaction directed along the x -axis (not shown in the figure). Eq. 2.11 is the relevant nuclear Hamiltonian with the component B_x set to zero.

In general, the field acting on an ion in the 'puckered' spin arrangement has both B_y and B_z components. This requires a slight modification of Equation 2.15. For the 'puckered' structure directed at an angle β to the a -axis

$$M_{a'} = (M_n + M_e) \sin \beta \quad 2.32a$$

$$M_a = (M_n + M_e) \cos \beta \quad 2.32b$$

where $M_{a'}$ and M_a are the moments resolved along the a' and a axes respectively. Here $\beta = \pi/2$ corresponds to the anti-ferromagnetic 'flop' phase and $\beta = 0$ corresponds to the paramagnetic or ferromagnetic phase. The total field acting on an ion in the direction of z is given by the expression

$$B_i^z = B_i^0 + B_i^z \left\{ \mu_0 \left[\left(\frac{1}{3} - d \right) \frac{N \mu_B}{V} \right] + D_d + D_{\text{exch.}} \right\} \frac{(M_n + M_e) \cos \beta}{\mu_B} \quad 2.33$$

This is identical to Equation 2.15 if $(M_n + M_e) \cos \beta$ is substituted for m . Equation 2.33 can be written to give

$$B_i^z = B_i^0 + B_i^z \frac{D_z (M_n + M_e) \cos \beta}{\mu_B} \quad 2.34$$

D_z is the sum of the demagnetising, Lorentz, local dipole and exchange fields. For example, for Sample 2 (section 4.2.3)

$D_z = -0.0043 T \mu_B^{-1}$. To calculate B_i^y it is necessary to set the demagnetisation, Lorentz and exchange terms to zero. This is because the net moment is zero in the antiferromagnetic 'flop' phase. Also the exchange due to the four nn cancel out. Thus

$$D_y = 0.0734 T \mu_B^{-1} \quad 2.35$$

as explained in section 2.6.2. This gives for the total field at the ion in the y direction

$$B_i^y = D_y (M_n + M_e) \sin \beta / \mu_B \quad 2.36$$

The electronic and nuclear moments are given by the expressions

$$M_e = g_J \mu_B \langle \underline{J} \rangle \quad 2.37a$$

$$M_n = (\gamma_I \hbar) \langle \underline{I} \rangle \quad 2.37b$$

where $\langle \rangle$ indicates an expectation value. These expressions can be substituted into Equations 2.36 and 2.34 to compute the ionic fields. Multiplying Equations 2.34 and 2.36 by the electronic susceptibility χ_e^0 ($= 2.32 \mu_B T^{-1}$) the following equations are easily derived:

$$M_i^{ez} = \chi_{ez} [\mu_B B_i^z + D_z M_n \cos \beta] \quad 2.38a$$

$$M_i^{ey} = \chi_{ey} [D_y M_n \sin \beta] \quad 2.38b$$

where $\chi_{ez} = (1 - D_z \chi_e^0)^{-1} \chi_e^0 \quad 2.39a$

and $\chi_{ey} = (1 - D_y \chi_e^0)^{-1} \chi_e^0 \quad 2.39b$

Here M_i^{ez} and M_i^{ey} are respectively the components of the electronic moment in the z and y directions.

The components of the 'effective' field at the nuclei

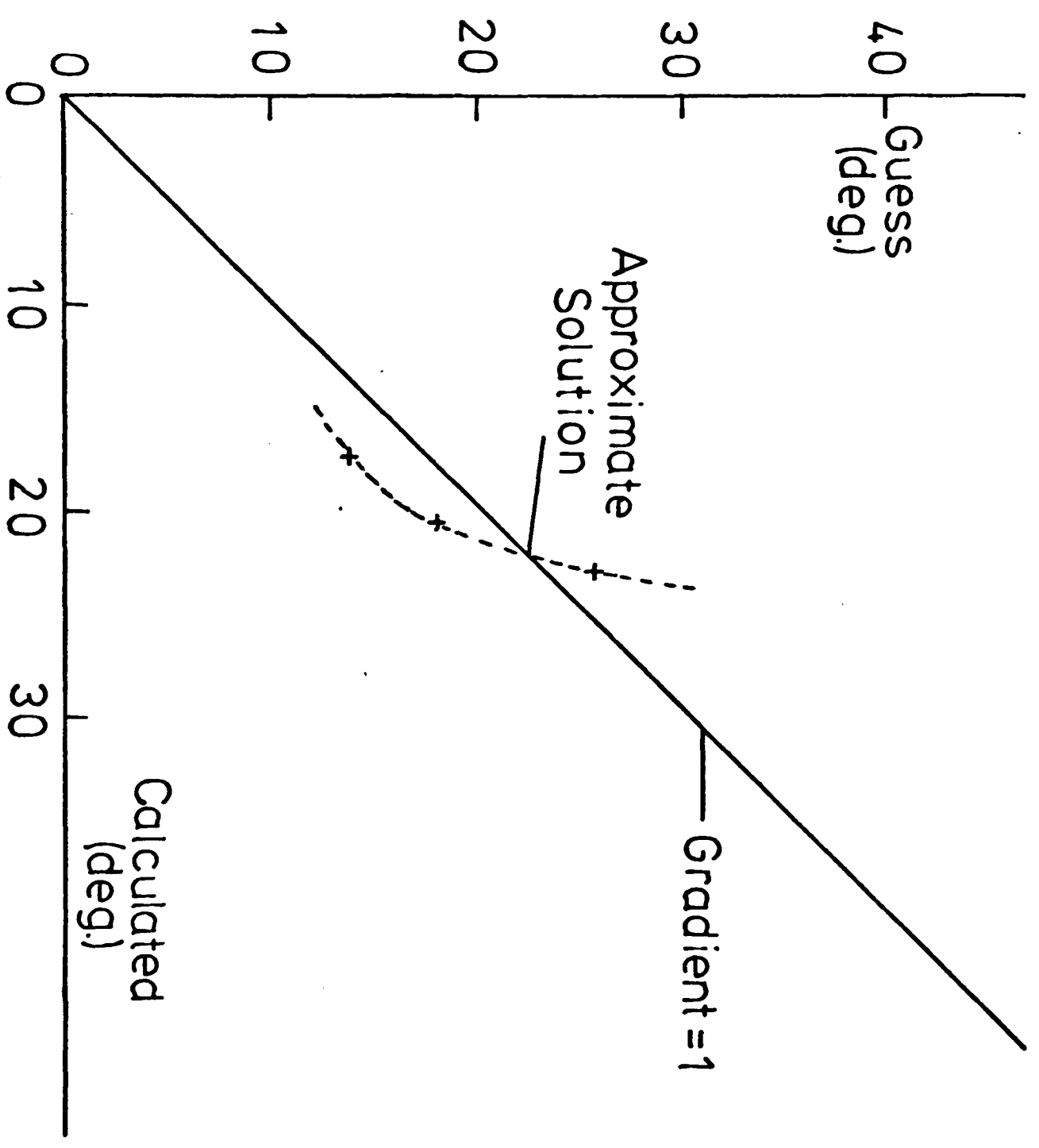


Fig. 2.6 Self-consistent calculation: method of solution

are given by the expressions

$$B_z^n = B_i^0 + D_z (M_i^{ez} + M_n) \cos \beta / \mu_B + A_J' M_i^{ez} \quad 2.40a$$

$$B_y^n = D_y (M_i^{ey} + M_n) \sin \beta / \mu_B + A_J' M_i^{ey} \quad 2.40b$$

where $A_J' = \frac{A_J}{(g_J B g_I \mu_n)}$

Substituting Equations 2.38a and 2.38b into Equations 2.40a and 2.40b to eliminate the electronic components of the magnetic moment, we get

$$B_z^n = B_i^0 + \chi_{ez} (A_J' + D_z / \mu_B) (\mu_B B_i^0 + D_z M_n \cos \beta) + D_z M_n \cos \beta / \mu_B \quad 2.41a$$

$$\text{and } B_y^n = \chi_{ey} (A_J' + D_y / \mu_B) (D_y M_n \sin \beta) + D_y m_n \sin \beta / \mu_B \quad 2.41b$$

It should be noted that B_y^n and B_z^n refer to 'effective' fields at the nucleus. For this particular choice of axes the nuclear Hamiltonian is given by

$$\mathcal{H} = -g_I \mu_n B_y^n I_y - g_I \mu_n B_z^n I_z - \frac{1}{2} P [I_z^2 - \frac{1}{3} I(I+1)] + \frac{1}{4} P [I_+^2 + I_-^2] \quad 2.42$$

where the 'bare' nuclear g factors are used in the nuclear zeeman term. The net nuclear moment per ion is given by the Brillouin function

$$M_n = (\chi_I \hbar) \text{Tr} [I e^{-\beta \mathcal{H}}] / \text{Tr} [e^{-\beta \mathcal{H}}] \quad 2.43$$

A 'self-consistent' solution is equivalent to solving Equations 2.41 to 2.43 to give consistent values for β and M_n .

The method of solution used is illustrated in Fig. 2.6 (for β only). A 'guess' value of β was used to compute the

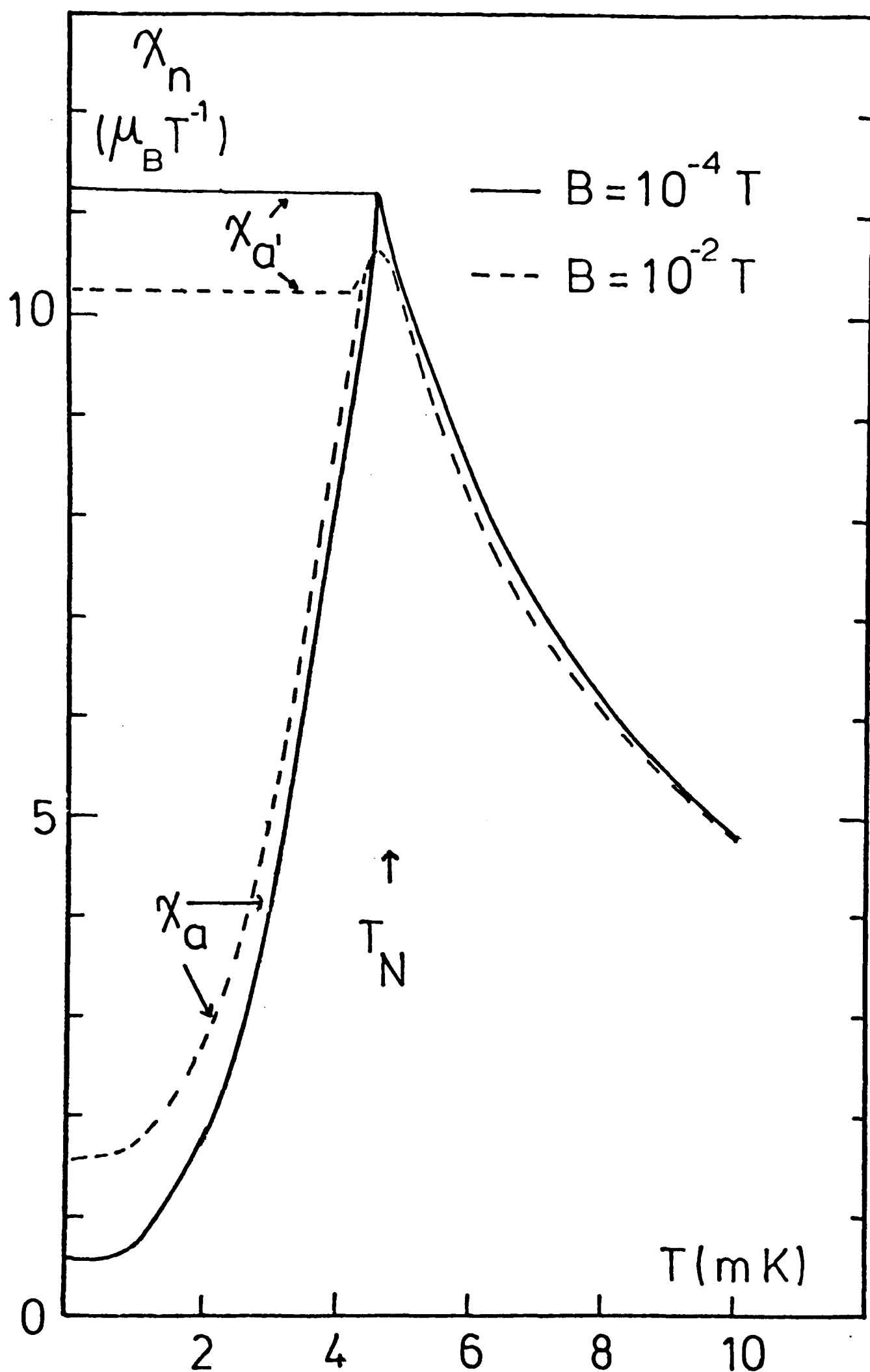


Fig. 2.7 Nuclear susceptibility in small applied fields

nuclear field using Equation 2.40. The nuclear moment was then evaluated using Equations 2.42 and 2.43 from which β was evaluated. The 'calculated' value of β was then used as a 'guess' value and the procedure repeated. In this way three 'guess' and 'calculated' values were generated. An approximate value for β was deduced by evaluating the point of intersection of a quadratic curve through these points and a straight line of slope unity through the origin. By going through this procedure a number of times a sufficiently accurate 'self-consistent' value of β was obtained. A similar procedure was used to evaluate the nuclear moment M_n .

2.8 Some Magnetic Properties of the Nuclear Antiferromagnet

HoVO₄

The 'self-consistent' mean field calculations were used to compute some magnetic properties of the nuclear antiferromagnet HoVO₄. These are illustrated in Figs. 2.7 to 2.9.

2.8.1 Nuclear Susceptibility

Fig. 2.7 shows the calculated nuclear susceptibility as a function of temperature for several applied fields along the a-axis. χ_a and χ_a , are the nuclear susceptibilities parallel and perpendicular to the applied field respectively. As expected for an ordinary antiferromagnet, χ_a , is independent of temperature while χ_a decreases with temperature below T_N .

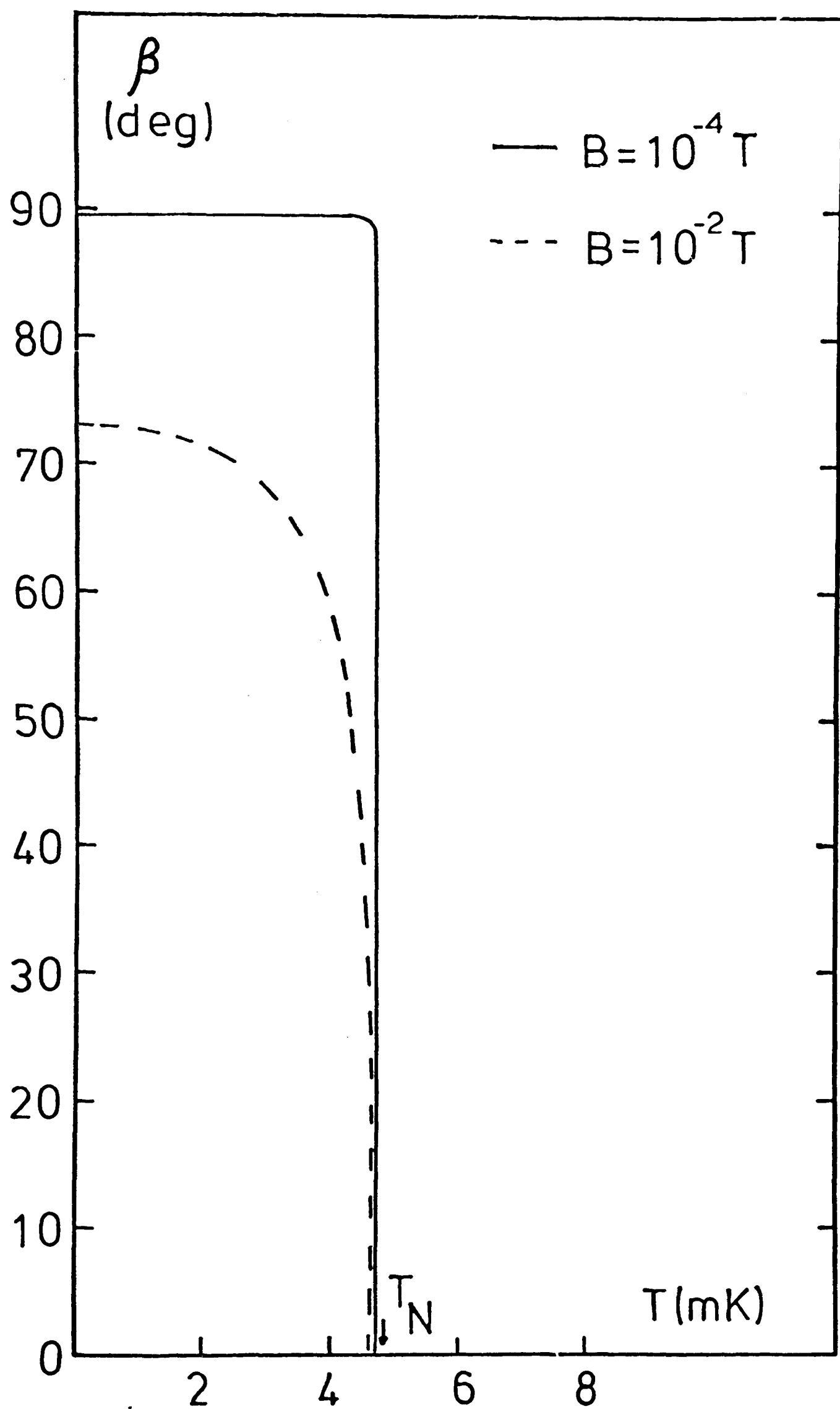


Fig. 2.8 Canting angle vs temperature

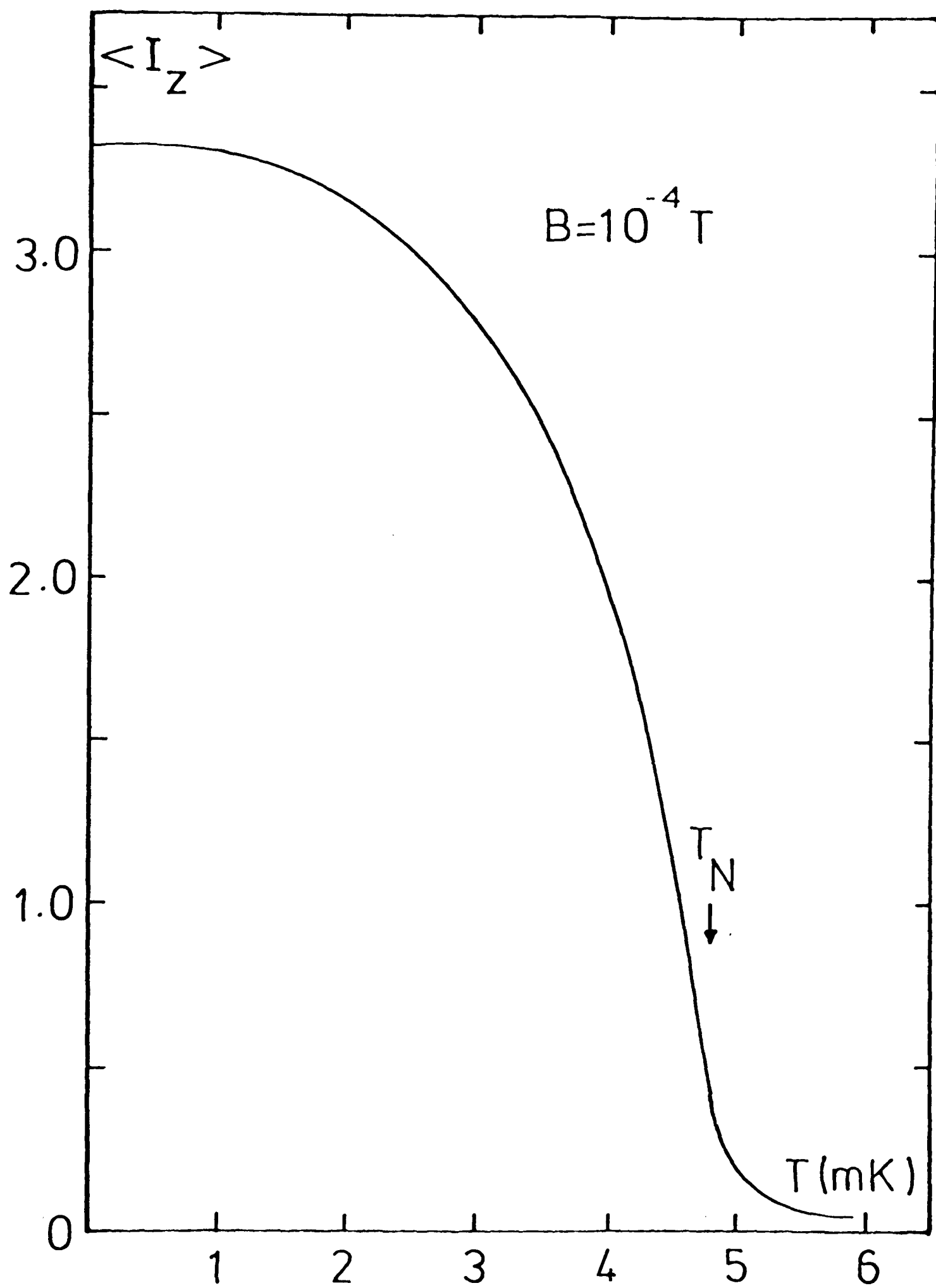


Fig. 2.9 $\langle I_z \rangle$ vs temperature

2.8.2 Spin 'Flop' Phase

The calculations showed that in a small applied field of $1 \times 10^{-4} \text{ T}$ the spins were in the 'flop' phase below T_N as predicted in Section 2.6.4. Fig. 2.8 shows how the 'canting angle' β varies as a function of temperature for two applied fields. As expected, this angle decreases on increasing field and temperature.

2.8.3 The Molecular Field

In zero applied field, the ionic field was evaluated to be 0.034 tesla (7) at 0K. Fig. 2.9 shows $\langle I_z \rangle$ as a function of temperature in an applied field $B_z = 1 \times 10^{-4} \text{ T}$. It can be seen that the saturation value is slightly below the expected value of $7/2$, possibly due to the quadrupole interaction.

Chapter 2

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CHAPTER 3

NUCLEAR ORIENTATION THEORY FOR A NON-AXIALLY-
SYMMETRIC NUCLEAR ENSEMBLE

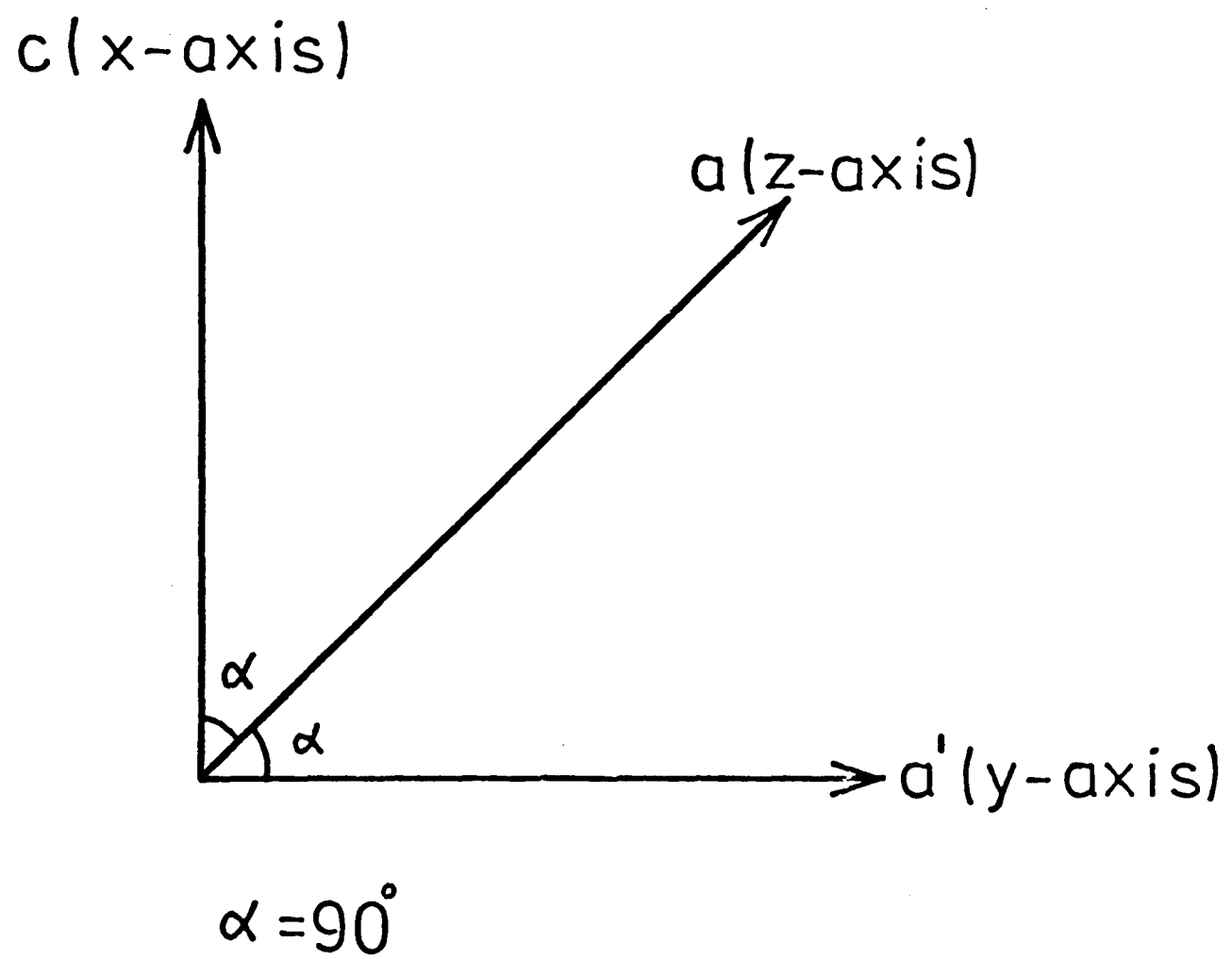


Fig. 3.1 Spatial relation between the crystal and statistical tensor axes

3.1 Introduction

This chapter describes the theory relevant to the analysis of gamma-ray anisotropy at low temperatures. For HoVO_4 , the axis of the electric quadrupole interaction does not coincide with the Zeeman axis. This necessitated the use of generalised statistical tensors of Fano (1, 2), which are introduced in this chapter. The transformation properties of these statistical tensors are discussed with particular reference to 'canted' spin structures of the antiferromagnet HoVO_4 .

3.2 The Nuclear Hamiltonian

It is convenient to use the nuclear Hamiltonian of Equation 2.11, with the applied field along the crystal a-axis (z co-ordinate) and the axially symmetric quadrupole interaction along the c-axis (x-co-ordinate) (Fig. 3.1). This gives

$$\mathcal{H} = -\gamma_I B_Z I_Z - \frac{1}{2}P [I_Z^2 - \frac{1}{3}I(I+1)] + \frac{1}{4}P [I_+^2 + I_-^2] \quad 3.1$$

For $^{166\text{m}}\text{HoVO}_4$, $(\frac{\gamma_I}{\hbar}) \simeq 660 \text{ MHz/tesla}$ and $^{166\text{m}}P = +3.4(3) \text{ MHz}$

(see Section 6.4.1).

3.3 Nuclear Orientation

Nuclear Orientation or polarisation occurs when the nuclear energy states become unequally populated. This is generally produced in practice by cooling the nuclei to temperatures where the splitting of the nuclear levels becomes of order kT . Most of the Nuclear Orientation to date has taken advantage of the large magnetic hyperfine fields generated at impurity sites in a ferromagnetic host. In HoVO_4 , the

mixing of the electronic states produces a large 'enhancement' of the applied field at the nucleus via the hyperfine interaction. This produces significant nuclear polarisation at accessible temperatures and magnetic fields. However, a non-axial electric quadrupole interaction makes the analysis of the angular distribution of emitted gamma-rays at low temperatures complicated, especially in low applied fields where the electric quadrupole and magnetic Zeeman interactions become comparable in magnitude. The general formulation of the angular distribution of gamma-ray emission as discussed for example by Steffen and Alder (3) is a convenient starting point for evaluation of NO statistical tensors for the case of a non-axial quadrupole interaction. This was the procedure used in this work and is discussed more fully in References 4 and 5. The following is a brief summary.

For a nuclear system, the angular distribution of emitted gamma radiation in a solid angle $\delta\Omega$ is given by the expression.

$$W'(\theta, \phi) = \frac{\delta\Omega}{2\pi^2} \sum_{\lambda \text{ even}} (2I_i+1)^{\frac{1}{2}} (2\lambda+1)^{-\frac{1}{2}} A_{\lambda}(\gamma) \rho_q^{\lambda}(I_i) Y_{\lambda q}^*(\theta, \phi)$$

3.2

where I_i is the spin of the oriented state and λ is the rank of the statistical tensor describing the orientation of the parent nucleus. Owing to parity conservation, the sum is only over $\lambda = \text{even}$ for gamma radiation. For beta radiation, where this is not necessarily true, odd terms have to be considered. The highest multipolarity studied in this work was 2, which required terms up to $\lambda = 4$.

3.4 The A_λ (λ) Parameter

This is the product of the deorientation parameter U_λ and the F_λ transition coefficient (see below).

3.4.1 The Deorientation Parameter U_λ

U_λ describes the effect of unobserved transitions between the oriented state and the initial state I_i of the observed transition. These unobserved transitions reduce the 'anisotropy' of the observed transition by an amount which is related to the spins and multipolarities of the preceding (unobserved) transitions.

If the spins of the levels of the unobserved transition are I_1 and I_2 , then for a multipolarity L the deorientation parameter U_λ is given by the expression

$$U_\lambda (I_1 I_2 L) = (-1)^{I_1 + I_2 + L + \lambda} [(2I_1 + 1)(2I_2 + 1)]^{\frac{1}{2}} \left\{ \begin{matrix} I_1 & I_2 & L \\ I_2 & I_2 & \lambda \end{matrix} \right\} \quad 3.3$$

where $\left\{ \begin{matrix} I_1 & I_2 & L \\ I_2 & I_2 & \lambda \end{matrix} \right\}$ is a 6 - j symbol (6)

A preceding transition may be of mixed multipolarity and it is therefore convenient to define a mixing ratio δ where

$$\delta = \frac{\langle I_2 || L'' || I_1 \rangle}{\langle I_2 || L' || I_1 \rangle} \quad 3.4$$

The reduced matrix elements $\langle || || \rangle$ are transition probabilities for the multipolarities considered and $L'' = L' + 1$. The definitions and derivations of these reduced matrix elements have been discussed by Krane and Steffen (7). The highest multipolarity considered in this work was 2. Thus all δ s in this thesis refer to mixing ratios of E2 and M1 only. For a mixed transition, the deorientation parameter U_λ is given by the expression

$$U_{\lambda} = \frac{U_{\lambda} (I_1 I_2 L') + \delta^2 U_{\lambda} (I_1 I_2 L'')}{1 + \delta^2} \quad 3.5$$

where δ is as defined above. It should be noted that the sign convention for δ used in this work is that of Krane and Steffen (7). Where there are several transitions before the observed transition, the deorientation parameter is given by the product of the deorientation parameters of all preceding transitions. Thus

$$U_{\lambda} = U_{\lambda}^{(1)} U_{\lambda}^{(2)} \dots U_{\lambda}^{(m)} \quad 3.6$$

It often happens that the initial level of the observed transition is fed by several different cascades in which case the effective U_{λ} is determined by suitable weighting of the individual U_{λ} s in proportion to the intensities of the transitions considered. Thus:

$$U_{\lambda} = \frac{\sum_{i=1}^N W_i U_{\lambda}^i}{\sum_{i=1}^N W_i} \quad 3.7$$

where W_i and U_{λ}^i are the gamma-ray intensity and deorientation parameter respectively for the i^{th} feed.

3.4.2 The coefficient F_{λ}

The coefficient F_{λ} describes the angular distribution associated with the particular multipolarity of the observed transition. It is a function of the multipolarities and initial and final spins of the states involved. Consider a mixed transition between the initial state I_i and final state I_f of multipolarity L'' and L' where $L' = L'' - 1$. It is then convenient to define

$$F_{\lambda}^I (L^I L'' I_f I_i) = (-1)^{I_i + I_f + 1} [(2I_i + 1)(2L^I + 1)(2L'' + 1)(2\lambda + 1)]^{\frac{1}{2}} \\ \begin{pmatrix} L^I & L'' & \lambda \\ 1-1 & 0 & \end{pmatrix} \times \left\{ \begin{pmatrix} L^I & L'' & \lambda \\ I_i & I_i & I_f \end{pmatrix} \right\} \quad 3.8$$

where $\begin{pmatrix} L^I & L'' & \lambda \\ 1-1 & 0 & \end{pmatrix}$ is a 3-j symbol and $\left\{ \begin{pmatrix} L^I & L'' & \lambda \\ I_i & I_i & I_f \end{pmatrix} \right\}$ is a 6-j symbol (6)

The coefficient F_{λ} for a mixed transition is given by the expression

$$F_{\lambda} = \frac{F_{\lambda}^I (L^I L^I I_f I_i) + 2\delta F_{\lambda}^I (L^I L'' I_f I_i) + \delta^2 F_{\lambda}^I (L'' L'' I_f I_i)}{1 + \delta^2} \quad 3.9$$

where F_{λ}^I is defined by Equation 3.8. F_{λ} is sensitive to the magnitude and sign of δ through the middle term of Equation 3.9. This makes possible accurate evaluation of δ in NO studies from a measurement of F_{λ} (see section 7.3).

Tabulations of U_{λ} and F_{λ} for various initial and final spins and multipolarities exist (8) which were used in the NO studies of Chapter 6.

3.5 $Y_{\lambda q}(\theta, \phi)$

$Y_{\lambda q}(\theta, \phi)$ are the spherical harmonics of rank λ and can be written in the form

$$Y_{\lambda q}(\theta, \phi) = \lambda^q + |q| \left[\frac{2\lambda+1}{4\pi} \left(\frac{\lambda - |q|}{\lambda + |q|} \right) \right]^{\frac{1}{2}} P_{\lambda}^{|q|}(\cos\theta) e^{iq\phi} \quad 3.10$$

where P_{λ}^q is a Legendre polynomial.

3.6 The Statistical Tensors ρ_q^{λ}

The statistical tensors of rank λ , ρ_q^{λ} , can be calculated using the equation

$$\rho_q^\lambda(I_i) = \sum_m (-)^{I_i+m} \langle I_i -m | I_i m' | \lambda q \rangle \langle I_i m | \rho | I_i m' \rangle \quad 3.11$$

where ρ is the density matrix and $\langle I_i -m | I_i m' | \lambda q \rangle$ is a Clebsch-Gordan coefficient, defined for example by Edmonds (9). The density matrix is given by the equation

$$\rho = e^{-\beta H} / \text{Tr} [e^{-\beta H}] \quad 3.12$$

where $\beta = 1/k T$. Here T refers to the equilibrium nuclear spin temperature, which may be different from the lattice temperature. The density matrix (Equation 3.11) is not in general diagonal, as m need not be equal to m' . However, it is easy to compute $\langle I_i m | \rho | I_i m' \rangle$ once the unitary matrix U which reduces the Hamiltonian (Equation 3.1) to diagonal form is known. If ρ_E represents the density matrix in diagonal form, then

$$\rho = U \rho_E U^{-1} \quad 3.13$$

where the matrix ρ contains the elements of the density matrix in the Zeeman representation.

The statistical tensors ρ_q^λ are defined with respect to a particular set of orthogonal axes. On rotation of the co-ordinate axes through the Euler angles α , β and γ , the statistical tensors are given by the transformation equation

$$(\rho_q^\lambda)' = \sum_{q''} D_{q'' q}^\lambda(\alpha, \beta, \gamma) \rho_{q''}^\lambda \quad 3.14$$

where the prime refers to the new set of axes and $D_{q'' q}^\lambda(\alpha, \beta, \gamma)$ are the rotation matrices, defined for example by Edmonds (9).

For the Hamiltonian considered (Equation 3.1), all statistical tensors with odd q vanish. It can also be shown that

$$\rho_q^{\lambda*} = (-)^q \rho_{-q}^\lambda \quad 3.15$$

By expanding the spherical harmonics Y_q^λ and using Equation 3.15 (even q only), it can be shown that

$$W(\theta, \phi) = \frac{\delta\mathcal{N}}{4\pi} \left\{ 1 + A_2(\gamma) (2I_i + 1)^{\frac{1}{2}} \left[\rho_0^2 \frac{1}{2} (3 \cos^2 \theta - 1) + \rho_2^2 \left(\frac{3}{2}\right)^{\frac{1}{2}} (1 - \cos^2 \theta) \cos 2\phi \right] + A_4(\gamma) (2I_i + 1)^{\frac{1}{2}} \left[\rho_0^4 \frac{1}{8} (35 \cos^4 \theta - 30 \cos^2 \theta + 3) + \rho_2^4 \frac{1}{2} \left(\frac{5}{2}\right)^{\frac{1}{2}} (1 - \cos^2 \theta) (7 \cos^2 \theta - 1) \cos 2\phi + \rho_4^4 \left(\frac{35}{32}\right)^{\frac{1}{2}} (1 - \cos^2 \theta)^2 \cos 4\phi \right] \right\} \quad 3.16$$

neglecting terms greater than $\lambda = 4$. As $\lambda = 4$ was the highest order considered in the analysis of gamma-rays emitted by $^{166m}\text{HoVO}_4$, Equation 3.16 is adequate for our purposes. It is convenient to define a normalised 'anisotropy' of gamma radiation by the equation

$$\text{Anisotropy} = W(\theta, \phi) - 1 = A_2(\gamma) (2I_i + 1)^{\frac{1}{2}} \left[\rho_0^2 \frac{1}{2} (3 \cos^2 \theta - 1) + \rho_2^2 \left(\frac{3}{2}\right)^{\frac{1}{2}} (1 - \cos^2 \theta) \cos 2\phi \right] + A_4(\gamma) (2I_i + 1)^{\frac{1}{2}} \left[\rho_0^4 \frac{1}{8} (35 \cos^4 \theta - 30 \cos^2 \theta + 3) + \rho_2^4 \frac{1}{2} \left(\frac{5}{2}\right)^{\frac{1}{2}} (1 - \cos^2 \theta) (7 \cos^2 \theta - 1) \cos 2\phi + \rho_4^4 \left(\frac{35}{32}\right)^{\frac{1}{2}} (1 - \cos^2 \theta)^2 \cos 4\phi \right] \quad 3.17$$

In Equation 3.17, $W(\theta, \phi)$ is normalised to give unity at high temperatures where the emitted gamma radiation is isotropic.

3.7 Solid Angle Correction Factor Q_λ

The 'measured' anisotropy is attenuated by a factor Q_λ due to the finite solid angle subtended by gamma-ray detectors used in experiments. These solid angle correction factors have been tabulated for several geometries for Ge (Li) detectors (10). They are a function of distance from source and the energy and multipolarity of the emitted gamma-ray. The values used in these experiments were calculated by D.W. Murray (11). Table 3.1 gives the solid angle corrections for the Ge (Li) detectors used, for typical values of distance from source and

gamma-ray energies.

Table 3.1

Q_λ , Solid angle correction factors

Distance from source (cm)	Gamma-ray energy (keV)	Q_2	Q_4
7	1000	0.9581	0.8650
11	1000	0.9801	0.9346
15	1000	0.9883	0.9614

Finally, we write for the measured (normalised) anisotropy of gamma-radiation.

$$\begin{aligned}
 W(\theta, \phi) - 1 = & Q_2 A_2(\gamma) (2I_i + 1)^{\frac{1}{2}} \left[\rho_0^2 \frac{1}{2} (3 \cos^2 \theta - 1) + \rho_2^2 \left(\frac{3}{2} \right)^{\frac{1}{2}} (1 - \cos^2 \theta) \right. \\
 & \left. \cos 2\phi \right] + Q_4 A_4(\gamma) (2I_i + 1)^{\frac{1}{2}} \left[\rho_0^4 \frac{1}{8} (35 \cos^4 \theta - 30 \cos^2 \theta + 3) + \right. \\
 & \left. \rho_2^4 \left(\frac{5}{2} \right)^{\frac{1}{2}} (1 - \cos^2 \theta) (7 \cos^2 \theta - 1) \cos 2\phi + \rho_4^4 \left(\frac{35}{2} \right)^{\frac{1}{2}} (1 - \cos^2 \theta)^2 \right. \\
 & \left. \cos 4\phi \right]
 \end{aligned}
 \tag{3.18}$$

Where Q_2 and Q_4 are the solid angle correction factors appropriate to the detector geometry.

It is clear from Equations 3.16-3.18 that for gamma-rays of multipolarity 1 (A_2 term only) the statistical tensors ρ_0^2 and ρ_2^2 fully describe the orientation of the parent nucleus. For such a case, anisotropy data from two detectors are both necessary and sufficient to get complete information about the orientation of the parent nucleus, provided that the geometries of the detectors are not equivalent. However, if A_4 terms are also present, a further three detectors are needed to evaluate the terms ρ_0^4 , ρ_2^4 and ρ_4^4 . This difficulty was avoided in the NO experiments of Chapter 5 by choosing a pure E1 gamma-ray (712 keV).

For the particular case of the 712 keV gamma-ray of

$^{166\text{m}}\text{HoVO}_4$ ($A_2(\gamma) = 0.273$, $A_4(\gamma) = 0$) we have, using Equation 3.18.

$$\begin{aligned} W_a(0) - 1 &= Q_2 \cdot 1.0573 \rho_0^2 \\ W_a'(\pi/2, \pi/2) - 1 &= Q_2 (-0.5287 \rho_0^2 - 1.2949 \rho_2^2) \\ W_c(\pi/2, 0) - 1 &= Q_2 (-0.5287 \rho_0^2 + 1.2949 \rho_2^2) \end{aligned} \quad 3.19$$

where $W(\theta, \phi)$ are the measured anisotropies. Equation 3.19 can be used to determine ρ_0^2 and ρ_2^2 from the measured anisotropies.

3.8 Evaluation of $\bar{\rho}_0^2$ and $\bar{\rho}_2^2$ for 'Canted' Spin Structures

Equation 3.18 applies to co-linear (ferromagnetic or simple antiferromagnetic) spin structures only. For 'canted' spin structures (Fig. 2.4) we define an average 'measured' statistical tensor

$$\bar{\rho}_q^2 = \frac{1}{2} [(\rho_q^2)' + (\rho_q^2)'] \quad 3.20$$

where $(\rho_q^2)'$ and $(\rho_q^2)''$ are statistical tensors for two spins 'canted' at an angle β and defined with respect to the same (unprimed) frame of reference (Fig. 3.1). However, in this frame of reference the direction of the spin does not coincide with the z-axis. This necessitates re-writing the nuclear Hamiltonian (Equation 3.1) to include a Zeeman term along the y direction. But it is simpler to use the transformation properties of the statistical tensors themselves (Equation 3.14). This was the procedure used in this work (4).

In order to evaluate the average statistical tensors $\bar{\rho}_q^\lambda$, the statistical tensors $(\rho_q^\lambda)'$ and $(\rho_q^\lambda)''$ are transformed from the common unprimed frame of reference to their own 'intrinsic' frames of reference using Equation 3.14 with the Euler angles $\alpha = \frac{1}{2}\pi$, $\gamma = -\frac{1}{2}\pi$ and $\beta = \pm\beta$. For the particular

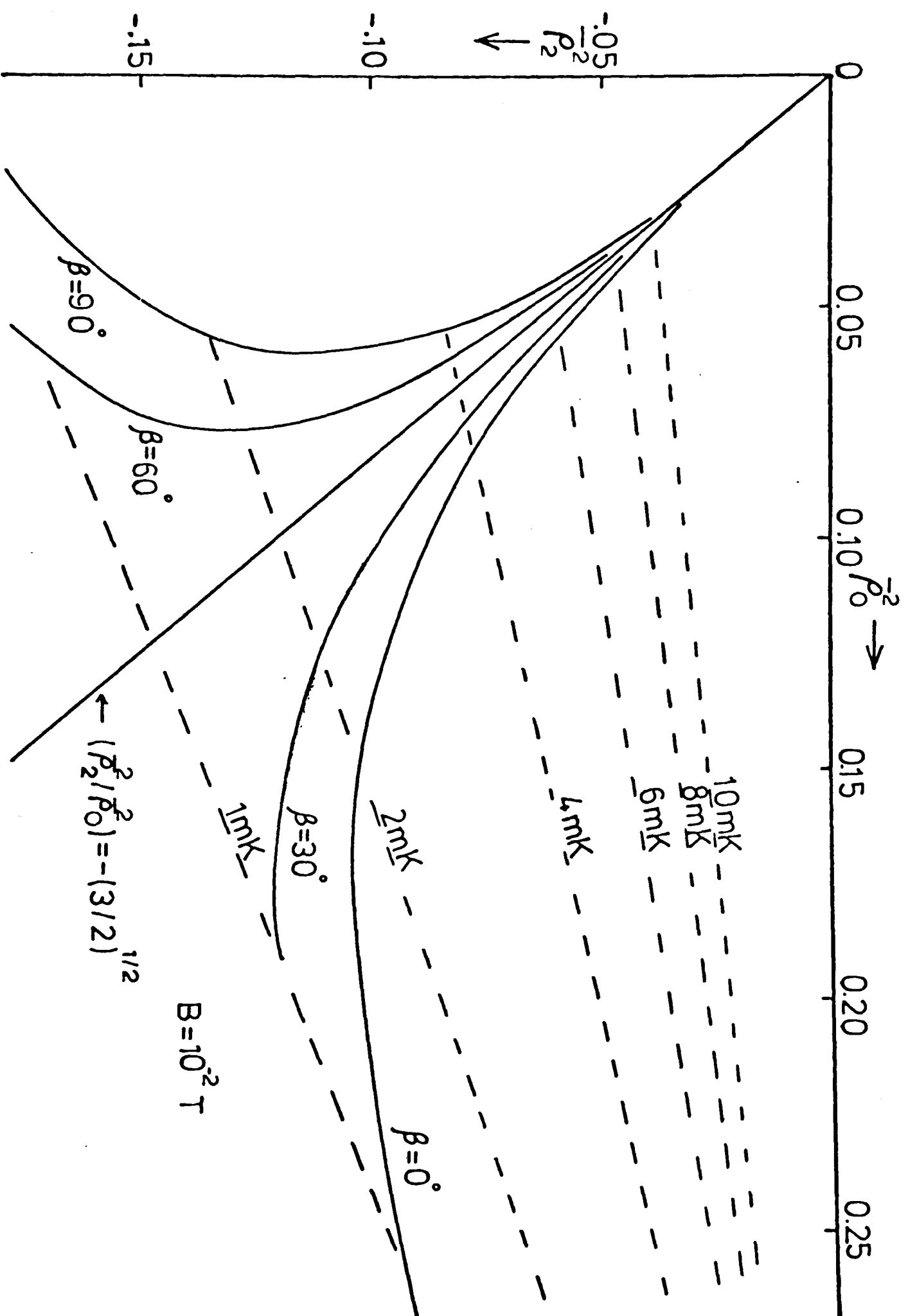


Fig. 3.2 Statistical tensor plot of $\bar{\rho}_2^2$ vs $\bar{\rho}_0^2$

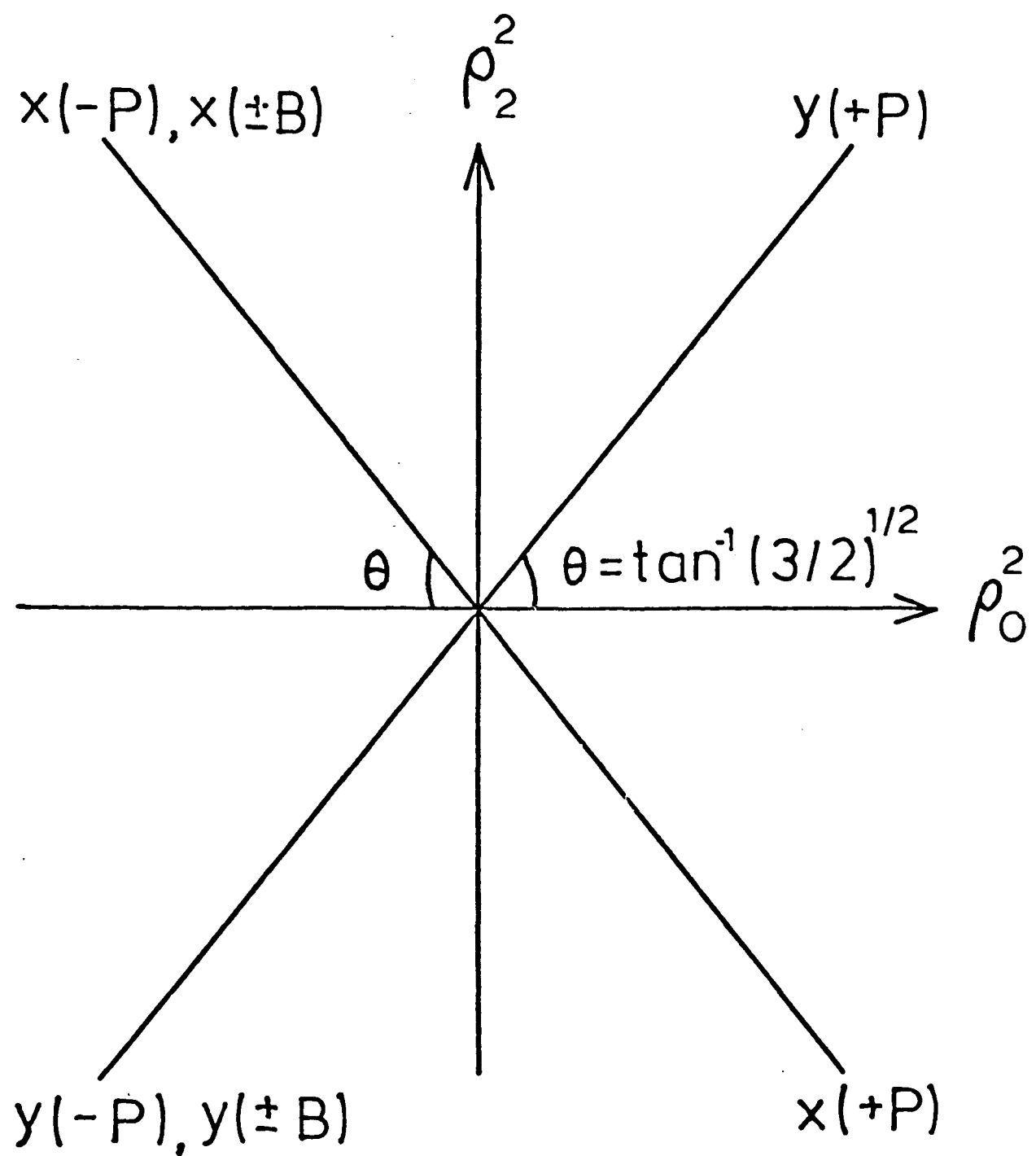


Fig. 3.3 Statistical tensor diagram for pure magnetic and pure quadrupole interactions

case of $\lambda = 2$

$$\begin{aligned} \bar{\rho}_2^{-2} &= \rho_2^2 (1 - \frac{1}{2} \sin^2 \beta) - \rho_0^2 \frac{1}{2} \left(\frac{3}{2}\right)^{\frac{1}{2}} \sin^2 \beta \\ \text{and } \bar{\rho}_0^{-2} &= -\rho_2^2 \left(\frac{3}{2}\right)^{\frac{1}{2}} \sin^2 \beta + \rho_0^2 \frac{1}{2} (3 \cos^2 \beta - 1) \end{aligned} \quad 3.21$$

for a canting angle β . Putting $\beta = 45^\circ$ in Equation 3.21

$$\bar{\rho}_2^{-2} / \bar{\rho}_0^{-2} = -\left(\frac{3}{2}\right)^{\frac{1}{2}} \quad 3.22$$

For an isotropic distribution of spin directions in the a-a' plane we have for the mean values

$$\overline{\sin^2 \beta} = \overline{\cos^2 \beta} = \frac{1}{2}$$

which is equivalent to $\beta = 45^\circ$. Thus Equation 3.22 is also true for an isotropic distribution of spin directions in the a-a' plane. In fact Equation 3.22 is also true for the 'pure' quadrupole case, where the Zeeman interaction term is totally absent. This ambiguity is resolved by analysing gamma radiation of higher multipolarity, for example 2. In section 3.9, the characteristics of statistical tensors of rank 4 is discussed with special reference to 'canted' spin structures.

Fig. 3.2 shows some theoretical calculations for $\bar{\rho}_0^{-2}$ and $\bar{\rho}_2^{-2}$ for several values of β for an applied field of $1 \times 10^{-2} \text{ T}$. The straight line of slope $-(\frac{3}{2})^{\frac{1}{2}}$ corresponds to the case of a 'pure' quadrupole interaction. The isotherms in the figure can be shown to be straight lines and can be used to evaluate the 'canting' angle β from the measured values of $\bar{\rho}_0^{-2}$ and $\bar{\rho}_2^{-2}$.

Fig. 3.3, reproduced from Allsop et al (4), shows how ρ_0^2 and ρ_2^2 vary for pure quadrupole and magnetic Zeeman interactions directed along various principal axes. Thus $x(B)$, $y(B)$ and $z(B)$ denote a pure magnetic hyperfine interaction directed along the x, y and z axes respectively. Similarly $x(P)$, $y(P)$

and $z(P)$ correspond to pure axially symmetric quadrupole interaction along z , y and z axes respectively. It is clear from the figure that the sign of the gradient of the lines is sensitive to the sign of the quadrupole interaction P , but not to the sign of the Zeeman interaction.

3.9 The Statistical Tensors ρ_0^4 , ρ_2^4 and ρ_4^4

For gamma radiation of multipolarity 2, statistical tensors up to rank 4 must be considered. The measured anisotropy from statistical tensors of rank 4 is given by the expression

$$W(\theta, \emptyset) - 1 = A_4(\gamma) Q_4 (2I_i + 1)^{\frac{1}{2}} \left[\rho_0^4 \frac{1}{8}(35\cos^4 - 30\cos^2 + 3) + \rho_2^4 \left(\frac{5}{2}\right)^{\frac{1}{2}} (1 - \cos^2)(7\cos^2 - 1) \cos 2\emptyset + \rho_4^4 \left(\frac{35}{2}\right)^{\frac{1}{2}} \frac{1}{4} (1 - \cos^2)^2 \cos 4\emptyset \right] \quad 3.23$$

(see Equation 3.16). This gives

$$\begin{aligned} W_a(0) - 1 &= Q_4 A_4(\gamma) (2I_i + 1)^{\frac{1}{2}} \rho_0^4 \\ W_a(\pi/2, \pi/2) - 1 &= Q_4 A_4(\gamma) (2I_i + 1)^{\frac{1}{2}} \left[.375\rho_0^4 + .7906\rho_2^4 + 1.046\rho_4^4 \right] \\ W_c(\pi/2, 0) - 1 &= Q_4 A_4(\gamma) (2I_i + 1)^{\frac{1}{2}} \left[.375\rho_2^4 - .7906\rho_2^4 + 1.046\rho_4^4 \right] \end{aligned} \quad 3.24$$

for the measured gamma anisotropy along the principal crystal directions. By proceeding in the same manner as indicated in section 3.8, it can be shown that for a spin structure 'canted' at an angle β to the a -axis (Fig. 2.4).

$$\begin{aligned} \rho_4^4 &= \frac{1}{8}(1 + 6\cos^2\beta + \cos^4\beta)\rho_4^4 - \frac{7^{\frac{1}{2}}}{4}\sin^2\beta(1 + \cos^2\beta)\rho_2^4 + \\ &\quad \frac{1}{8}\left(\frac{35}{2}\right)^2\sin^4\beta\rho_0^4 \end{aligned}$$

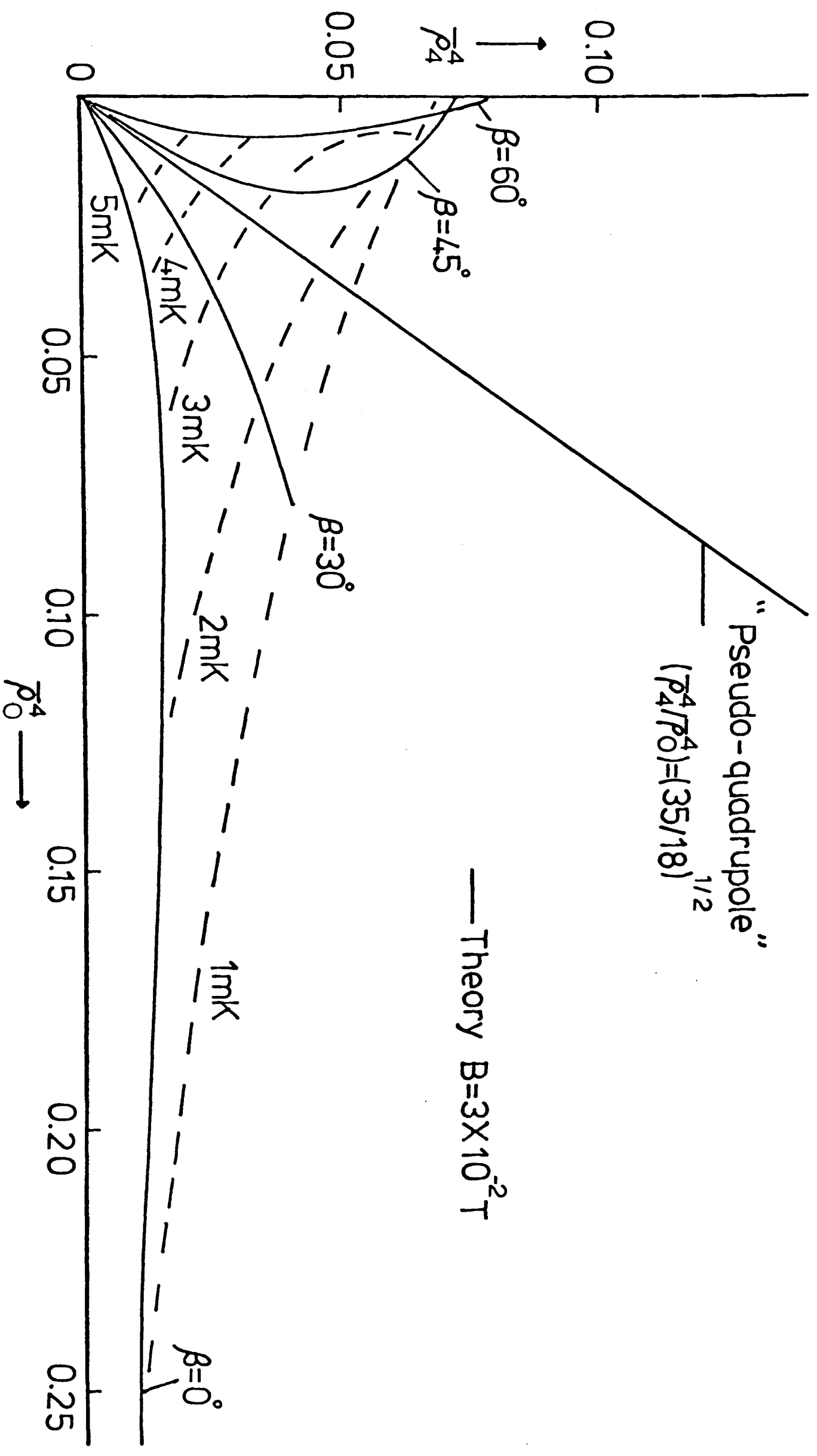


Fig. 3.4 Statistical tensor plot of $\bar{\rho}_L^4$ vs $\bar{\rho}_O^4$

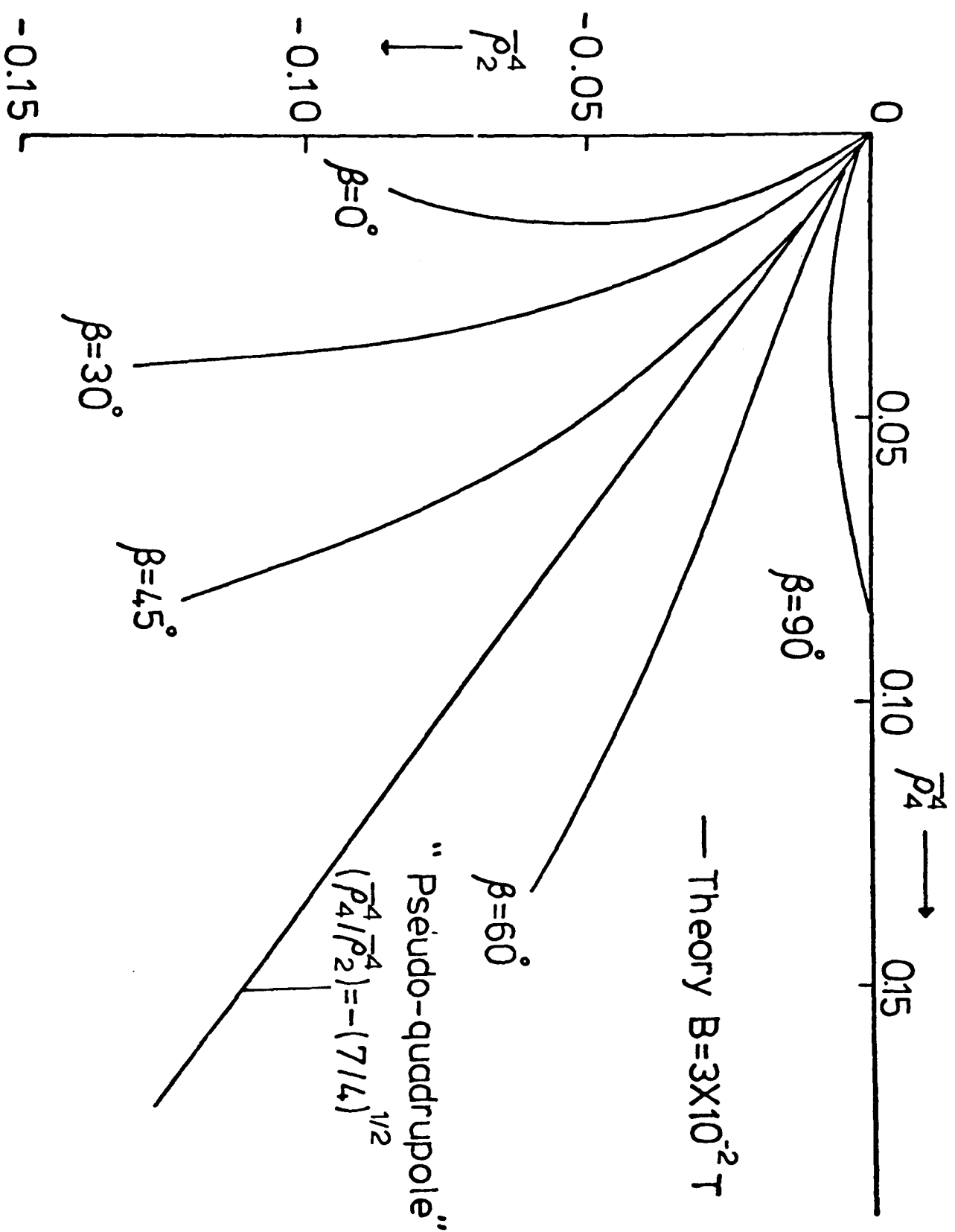


Fig. 3.5 Statistical tensor plot of $\bar{\rho}_4^{-4}$ vs $\bar{\rho}_2^{-4}$

$$\begin{aligned}\bar{\rho}_2^{-4} &= -\frac{7}{4} \sin^2 \beta (1 + \cos^2 \beta) \rho_4^4 + \frac{1}{2} (7 \cos^4 \beta - 6 \cos^2 \beta + 1) \rho_2^4 - \frac{1}{4} \left(\frac{5}{2}\right)^{\frac{1}{2}} \\ &\quad (1 - \cos^2 \beta) (7 \cos^2 \beta - 1) \rho_0^4 \\ \bar{\rho}_0^{-4} &= \frac{1}{4} \left(\frac{5}{2}\right)^{\frac{1}{2}} \sin^4 \beta \rho_4^4 - \frac{1}{2} \left(\frac{5}{2}\right)^{\frac{1}{2}} (1 - \cos^2 \beta) (7 \cos^2 \beta - 1) \rho_2^4 + \frac{1}{8} (35 \cos^4 \beta \\ &\quad - 30 \cos^2 \beta + 3) \rho_0^4\end{aligned}$$

3.25

Here $\bar{\rho}_4^{-4}$, $\bar{\rho}_2^{-4}$ and $\bar{\rho}_0^{-4}$ are the 'measured' average statistical tensors for the 'canted' spin structures and ρ_4^4 , ρ_2^4 and ρ_0^4 are defined with respect to the 'intrinsic' co-ordinate system of the spins. Two separate plots are required to completely specify these orientation tensors as a function of β and nuclear spin temperature T . Furthermore, the isotherms are not straight lines (cf $\bar{\rho}_0^{-2}$ vs $\bar{\rho}_2^{-2}$ plots) but are sensitive to the ratio of the nuclear quadrupole and Zeeman splittings. Figs. 3.4 and 3.5 are respectively plots of $\bar{\rho}_4^{-4}$ vs $\bar{\rho}_0^{-4}$ and $\bar{\rho}_4^{-4}$ vs $\bar{\rho}_2^{-4}$ for an applied field of $3 \times 10^{-2} T$ for several canting angles β . These curves represent the gamma-ray orientation tensors for $^{166m}\text{HoVO}_4$ for canting angle β in the antiferromagnetic phase. The quadrupole splitting P for $^{166m}\text{HoVO}_4$ was taken to be +3.4 MHz and the nuclear Zeeman splitting was taken to be 660 MHz tesla⁻¹. From the diagram, it is clear that in each case, straight lines define a 'pure' quadrupole interaction along the x-axis. However, for a spin structure canted at 45° to the applied field ($\beta = 45^\circ$), curves are obtained whose shapes depend critically on the ratio of the nuclear quadrupole to the Zeeman interactions. For the case of a uniform distribution of spin directions in the a - a' plane, that is with all canting angles equally probable, straight lines are also obtained with the same slope as the 'pure' quadrupole, but with the isotherms displaced away from the origin. This is

presumably because the emission pattern from a uniform distribution of spin directions in the a - a' plane is identical to a positive quadrupole interaction P , directed along the axis perpendicular to the plane containing the spins. Thus an accurate knowledge of spin temperature could in principle differentiate between the three cases outlined above, given that the orientation tensors are also known. Measurements of statistical tensors of rank 4 (section 5.8.2) clearly showed that for $^{166}\text{mHoVO}_4$ the spins were not canted at 45° to the direction of the applied field at a temperature of about 1 mK. However, in the absence of an independent measurement of spin temperature, it was not possible to distinguish between a 'pure' quadrupole interaction and a uniform distribution of spins in a plane perpendicular to the quadrupole axis.

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Chapter 3

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CHAPTER 4

A RADIO-FREQUENCY MAGNETIC SUSCEPTIBILITY STUDY
OF THE NUCLEAR ANTIFERROMAGNET HoVO_4

4.1 Introduction

This chapter describes radio-frequency (r.f.) magnetic susceptibility measurements on single crystals of HoVO_4 from 1 K down to about 1 mK and in fields of up to 2.4 tesla (1). These measurements extended to lower temperatures the work of Loftus (2), who showed that HoVO_4 obeys Curie's law down to about 100 mK.

4.2.1 Experimental Details

The single crystal samples of HoVO_4 were prepared by a flux-growth process in the Materials Preparation Group of the Clarendon Laboratory (3). A brush consisting of about 200 47 SWG enamelled copper wires was uniformly varnished around the crystal specimen and hard-soldered to the copper cold finger which was thermally linked to the mixing-chamber of the dilution refrigerator by means of a screw coupling. The r.f. magnetic susceptibility measurements were made using the same technique as discussed in section 1.4.1. The HoVO_4 crystal was pushed into a PTFE sleeve about 7 mm in diameter and fixed in place by nylon thread. The coil was constructed by winding a single layer of 200 turns of enamelled 46 SWG copper wires uniformly round the PTFE sleeve and fixed in position using GE-7041 varnish. A 3300 pF polystyrene capacitor formed the capacitive part of the L-C self-resonant circuit. The L-C circuit had a Q factor of about 100 at 4 K and resonated at approximately 450 kHz. The frequency of oscillation was stable to about 1 Hz but small shifts of order 10 Hz were observed on topping up the helium bath, which were small compared to the shifts due to the magnetic sample. The experimental arrangement is sketched

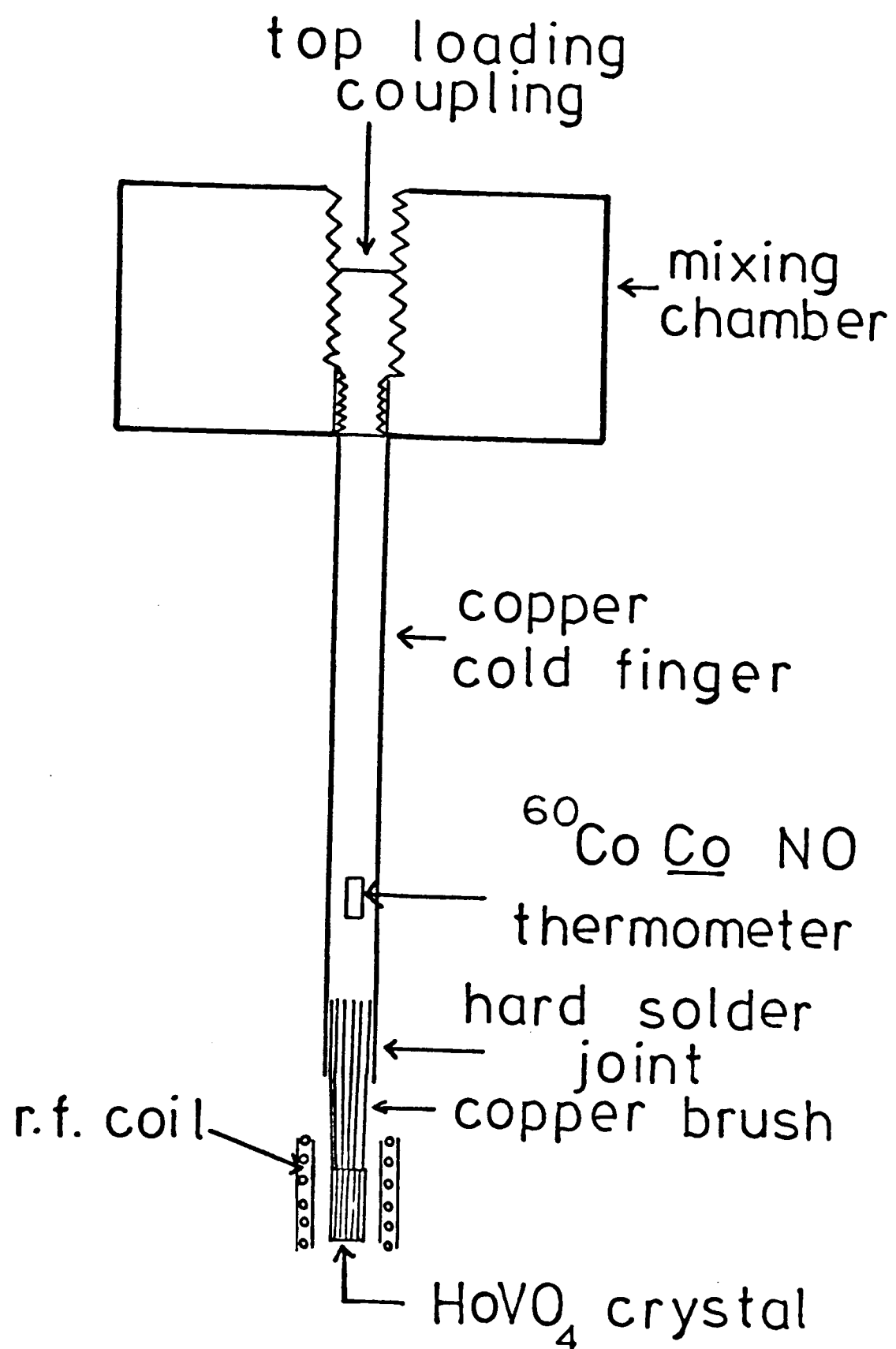


Fig. 4.1 Sketch of the r.f. susceptibility sample arrangement

in Fig. 4.1.

4.2.2 Samples

Two different samples were used in these experiments.

4.2.3 Sample 1

This was constructed by polishing down two single crystals and glueing them together with Araldite to give a cylinder approximately 5 mm long and 2.5 mm in diameter with the axis along an 'enhanced' (a) direction. NMR of ^{165}Ho at 4.2 K gave a nuclear Zeeman splitting, $(\frac{\gamma_L}{2\pi})$ of 1562 MHz/tesla. Bleaney et al (4) have shown that the local field at the Ho^{3+} ion is altered by demagnetisation and exchange effects (section 2.5). However, this shift in the field scales as the bulk magnetisation of the sample so that

$$\frac{B_{\text{ion}}}{B_{\text{app.}}} = (1 - f_s \chi)^{-1} \approx 1 + f_s \chi \quad (f_s \chi \ll 1) \quad 4.1$$

where χ is the sum of the electronic and nuclear susceptibilities and f_s is constant for a particular shape. The electronic susceptibility is constant below about 1 K and has the value

$$\chi_e = 2.32 \mu_B / \text{tesla} \quad (4)$$

For this particular sample $f_s = 0.0108$ from which the demagnetisation factor was calculated to be 0.19 (S.I. units). This is close to the estimated value of 0.14.

4.2.4 Sample 2

This was roughly rectangular, 1 mm x 2 mm x 3 mm with the c-axis along the longest dimension. The single crystal was irradiated in a thermal neutron flux of $1.5 \times 10^{14} \text{ n cm}^{-2}$

s^{-1} for 6 hours at the U.K.A.E.A. experimental reactor D.I.D.O. and subsequently annealed in air for 6 hours at 1300°C . The crystal which was originally greenish-yellow turned dark brown on irradiation. It did not revert to its natural colour on annealing and it is possible that defects and dislocations were not expelled by the heat treatment. It was found, however, that annealing in air at 1100°C , heating and cooling the crystal at the controlled rate of $50^{\circ}\text{C hr}^{-1}$, brought the crystal back to its natural colour. Unfortunately all the measurements were made before the controlled anneal and therefore the effect this had on the data will be discussed where appropriate in the text. The orientation of the crystal was checked using the Laue x-ray camera facility of the Materials Preparation Group of the Clarendon Laboratory.

Thermal contact was made in the same manner as for Sample 1 and the crystal a-axis was oriented parallel to the applied field. NMR of the sample in this geometry at 4.2 K gave ($\frac{\gamma_L}{2\pi}$) of 1510 MHz/tesla, which corresponds to a demagnetisation factor of 0.56 and f_s of -0.005.

4.2.5 Rare Earth Impurities in HoVO_4

The crystals were grown from 99.99% pure rare earth oxide. Neutron irradiation produced activities from which approximate rare earth impurity contents were calculated and are tabulated in Table 4.1.

Table 4.1
Rare earth impurities in irradiated HoVO₄

Rare earth impurity	Half life	Estimated content (ppm of H _o ³⁺)	β heating per μCi of ^{166m} Ho (nW)
¹⁶⁰ Tb	72.1 d	250	1
¹⁵² Eu	1.3 y	10	0.2
¹⁵⁴ Eu	8.5 y		0.05
¹⁷⁰ Tm	129 d	10	0.02
¹⁶⁹ Yb	32 d	600	0.1
Total			1.4

The crystals also contained varying amounts of silver from contamination of the platinum crucibles used in the crystal growing. The effect these impurities had on the magnetic behaviour of the nuclear antiferromagnet HoVO₄ is discussed in section 5.9.

4.2.6 Susceptibility Measurement

A highly stable oscillator was built which responded to the changes in the inductance of a parallel tuned L-C circuit (5). The resonant frequency ν of a loss-free oscillating circuit of capacitance C and inductance L is given by the expression

$$\nu = \frac{2\pi}{\sqrt{LC}}$$

The inductance of the coil is proportional to the magnetic permeability of the medium surrounding it. Thus

$$\nu = \frac{2\pi}{(L_o(1+\chi(\nu))C)^{1/2}}$$

where L_0 is the inductance of the coil in free space, f is the filling factor which is a function of the sample geometry and $\chi(\nu)$ is the r.f. magnetic susceptibility. Expanding to first order

$$\nu = \nu_0 \left(1 - \frac{1}{2} f \chi(\nu) \right) \quad (f \chi(\nu) \ll 1) \quad 4.2a$$

where ν_0 is the resonant frequency in the absence of the sample. It was not possible to measure ν_0 as the sample could not be removed from the coil during the experiment. It is convenient to express $\chi(\nu)$ as a function of frequency shift with respect to ν_0 . Thus:

$$\chi(\nu) = \frac{2(\nu_0 - \nu)}{f \nu_0} = \frac{-2 \delta \nu}{f \nu_0} \quad 4.2b$$

For the cylindrical sample f was estimated to be 0.091 from the measured frequency shifts and known nuclear susceptibility. This method afforded a very sensitive measurement of the temperature dependent nuclear susceptibility. For example, the nuclear susceptibility of Sample 1 produced a frequency shift of 3 kHz at 36 mK, which was stable to about 1 Hz.

The resonant frequency ν is related to the Q factor by the expression

$$\nu = \nu_0' \left(1 - 1/Q^2 \right)^{1/2} \quad 4.3$$

A rough calculation showed that eddy current losses due to the copper brush inside the coil produced a negligible reduction in Q of the circuit. This is consistent with our measurements which could not detect any reduction in Q due to the copper brush within an accuracy of about 20. Even if magnetoresistive effects of the copper were to change the Q of the circuit by ~ 10 , the frequency shift (Equation 4.3)

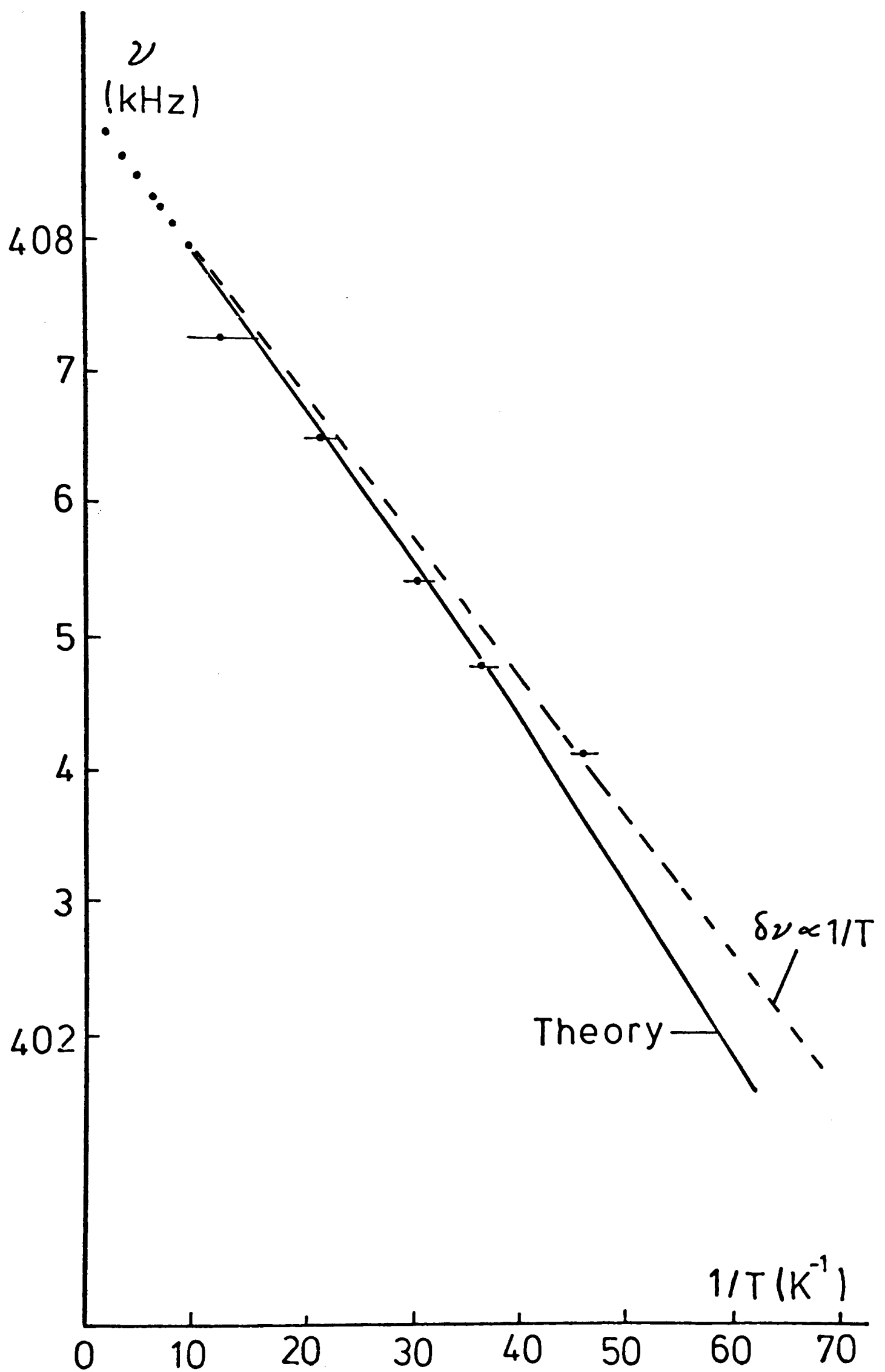


Fig. 4.2 resonant frequency vs reciprocal temperature

would be only 5 Hz at $\nu = 400$ kHz. This is consistent with the measurements of section 4.3.2.

4.2.7 Thermometry

A calibrated 220Ω $\frac{1}{2}$ W Speer carbon resistor was used to measure temperatures above about 50 mK. Details of its calibration are presented in section 1.4. Below 50 mK, the carbon resistor lost thermal contact with the mixing-chamber and so a single crystal of h.c.p. ^{60}Co Co NO thermometer was used at the lowest temperatures.

4.3 Measurements on Sample 1

4.3.1 Temperature Dependence of the r.f. Susceptibility

The resonant frequency ν of the coil circuit was measured as a function of temperature. Below about 40 mK, thermal relaxation times were long and therefore the frequency was recorded as a function of time at these temperatures. Thermal relaxation times, τ , could then be calculated by fitting the frequency curves to an exponential of the form

$$\nu = \nu_e (1 - e^{-t/\tau}) \quad 4.4$$

where τ is a relaxation time and ν_e is the equilibrium frequency (see section 4.3.2). The frequencies are plotted against reciprocal temperature in Fig. 4.2. As the susceptibility is directly proportional to the frequency shift from an appropriate origin (Equation 4.2b), this is equivalent to plotting susceptibility against temperature. A straight line would then indicate a Curie law. However, at the lowest temperatures, there is substantial deviation from Curie's law because of the nuclear quadrupole splitting P ($= 1.24$ mK).

Also the field acting on the nucleus has to be corrected for demagnetisation, dipolar and electronic exchange effects (see below). The nuclear susceptibility in zero field was evaluated from a perturbation expansion of the nuclear Hamiltonian to second order. The calculated values were then corrected in the following way:

The measured susceptibility is given by the expression

$$\chi_{\text{meas.}} = M / B_{\text{app.}} \quad 4.5$$

Using Equation 4.1

$$\chi_{\text{meas.}}^{-1} = \chi^{-1} - f_s \quad 4.6$$

Where the 'intrinsic' susceptibility χ is given by the sum of the nuclear and electronic susceptibilities

$$\chi = \chi_e + \chi_n \quad 4.7$$

and $\chi_e = 2.32 \mu_B / \text{tesla (4)}.$

The baseline frequency ν_0 is defined so as to include the electronic part of the susceptibility which is temperature independent below 1 K. The frequency shift $\delta\nu$ is then directly proportional to the difference between the measured susceptibility at the particular temperature considered and at 1 K

$$\delta\nu \propto \chi_{\text{meas.}}^T - \chi_{\text{meas.}}^e \quad 4.8$$

Here $\chi_{\text{meas.}}^e$ and $\chi_{\text{meas.}}^T$ are the electronic and total susceptibilities respectively. Combining Equations 4.6, 4.7 and 4.8.

$$\delta\nu \propto [(\chi_e + \chi_n)^{-1} - f_s]^{-1} - \chi_{\text{meas.}}^e \quad 4.9$$

where $\chi_{\text{meas.}}^e = \left(\frac{1}{2.32} - f_s \right)^{-1} = 2.38$ for the cylindrical sample (Sample 1). The 'intrinsic' and measured nuclear susceptibilities are summarised in Table 4.2 for various temperatures for Sample 1. Equation 4.9 can be used to

Table 4.2

Measured and theoretical nuclear susceptibilities below 100 mK

T (mK)	χ_n ($\mu_B T^{-1}$)	$\chi_{\text{meas.}}^n$ ($\mu_B T^{-1}$)
50	0.88	0.93
45	0.98	1.04
40	1.11	1.18
35	1.27	1.36
30	1.50	1.60
25	1.82	1.95
20	2.32	2.49
15	3.17	3.44

derive the nuclear susceptibility χ_n once $\delta\nu$ is known or vice versa. But the filling factor f_s cannot be measured directly and so Equation 4.9 was used to compute a theoretical curve which is also shown in Fig. 4.2. From the figure it is clear that below about 25 mK ($\frac{1}{T} = 40 \text{ K}^{-1}$) there were marked deviations from the theoretical fits. While it was difficult to assess the accuracy of the asymptotic values (see Equation 4.4) owing to the long relaxation time at the lowest temperatures, the deviations were large enough to suggest genuine deviations from the theoretical fits, computed using the nuclear Hamiltonian of Equation 2.11.

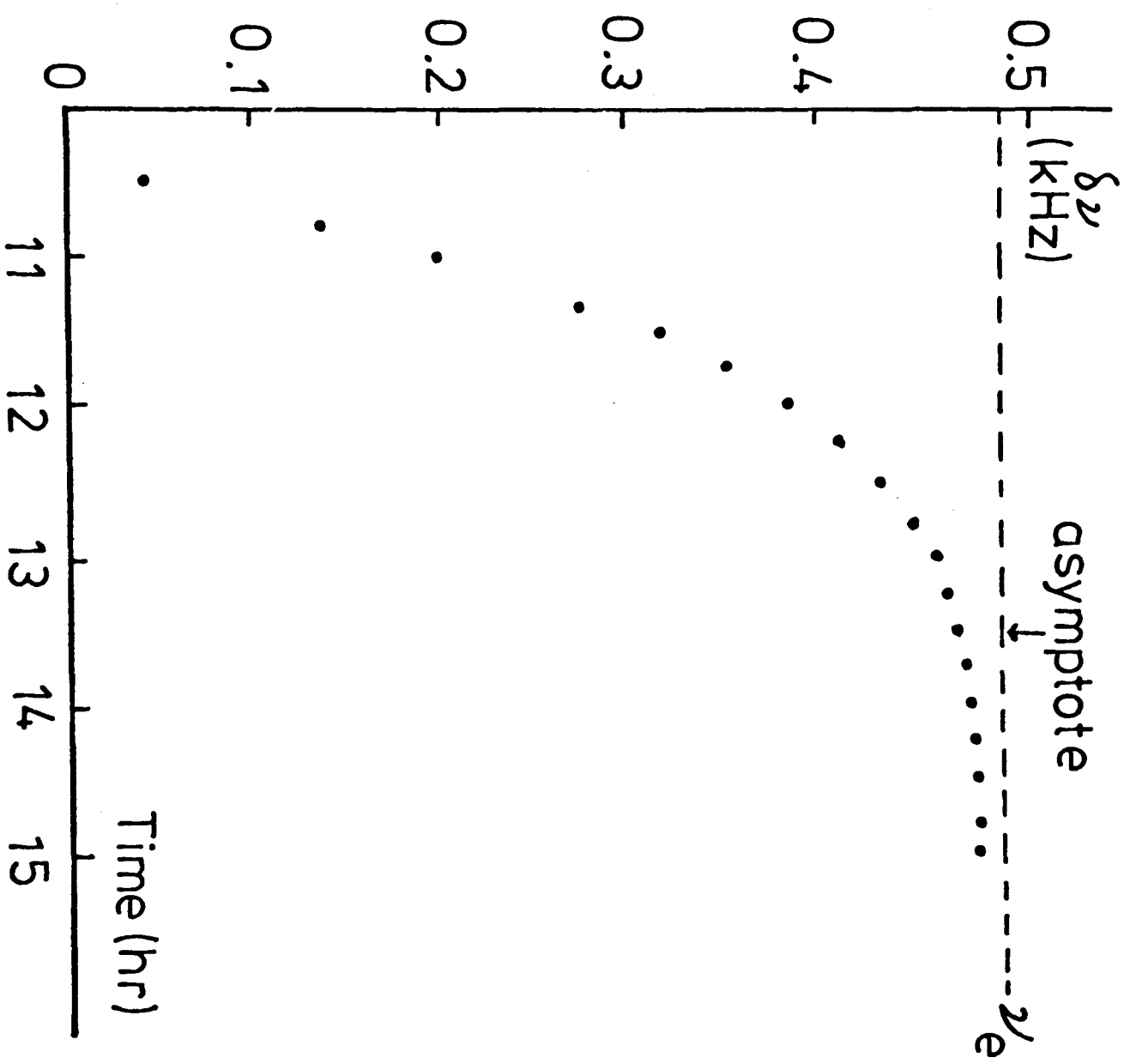


Fig. 4.3 Thermal relaxation at low temperatures

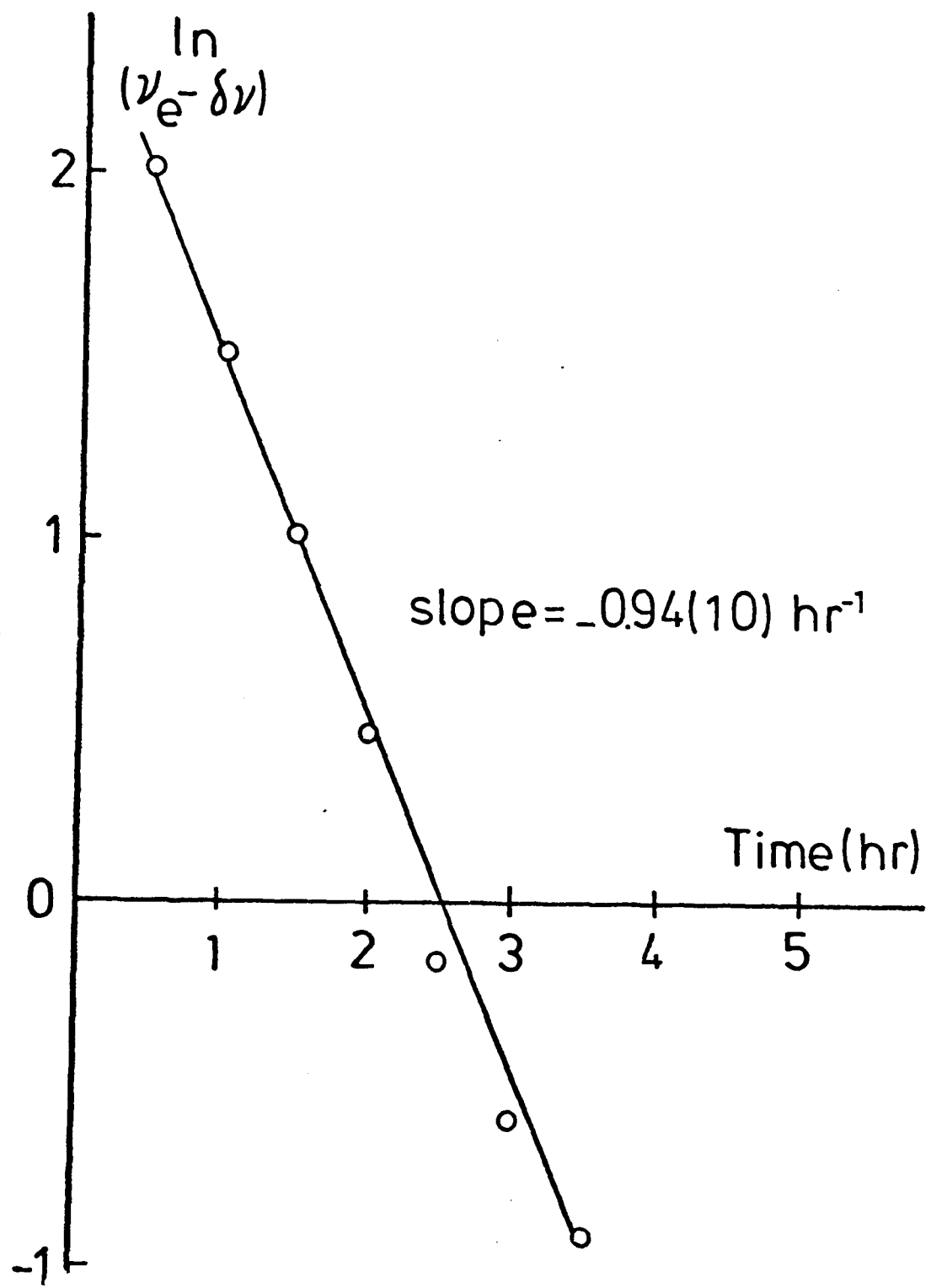


Fig. 4.4 Method for measurement of thermal relaxation time

4.3.2 Nuclear Spin-Lattice Relaxation Times

Loftus (2) concluded from his measurements that the nuclear spin-lattice times for ^{165}Ho were at most a few seconds at about 0.1 K. Similar results were obtained in this work at the highest temperatures but below about 50 mK relaxation times were longer and could be determined quite accurately. The temperature of the mixing-chamber of the dilution refrigerator was controlled by changing the current through the heater windings and the approach to thermal equilibrium of ^{165}Ho nuclei was monitored using the r.f. susceptibility. By fitting the frequency to Equation 4.4, the relaxation time τ was determined. Figs. 4.3 and 4.4 show one such fit at 34 mK. The measured relaxation times are plotted in Fig. 4.5 and are observed to be strongly temperature dependent.

At mK temperatures, the 'direct' process is expected to dominate the spin-lattice relaxation process. In this process, exchange of energy takes place directly between a nucleus and a single phonon of the appropriate energy (6). Bleaney et al (4) estimate

$$\tau_1^{-1} \sim 10^{-30} \nu^3 \coth(h\nu / 2 k T) \quad 4.10$$

for the 'direct' process from their NMR measurements on HoVO_4 at liquid helium temperatures. Here τ_1 (sec.) is the nuclear spin-lattice relaxation time for the 'direct' process and ν is a nuclear resonance frequency (Hz). This is also plotted in Fig. 4.5 for a typical NMR frequency of 150 MHz and gives relaxation times which are too long at the higher temperatures by several orders of magnitude. A faster relaxation process is indicated from these measurements, the mechanism of which

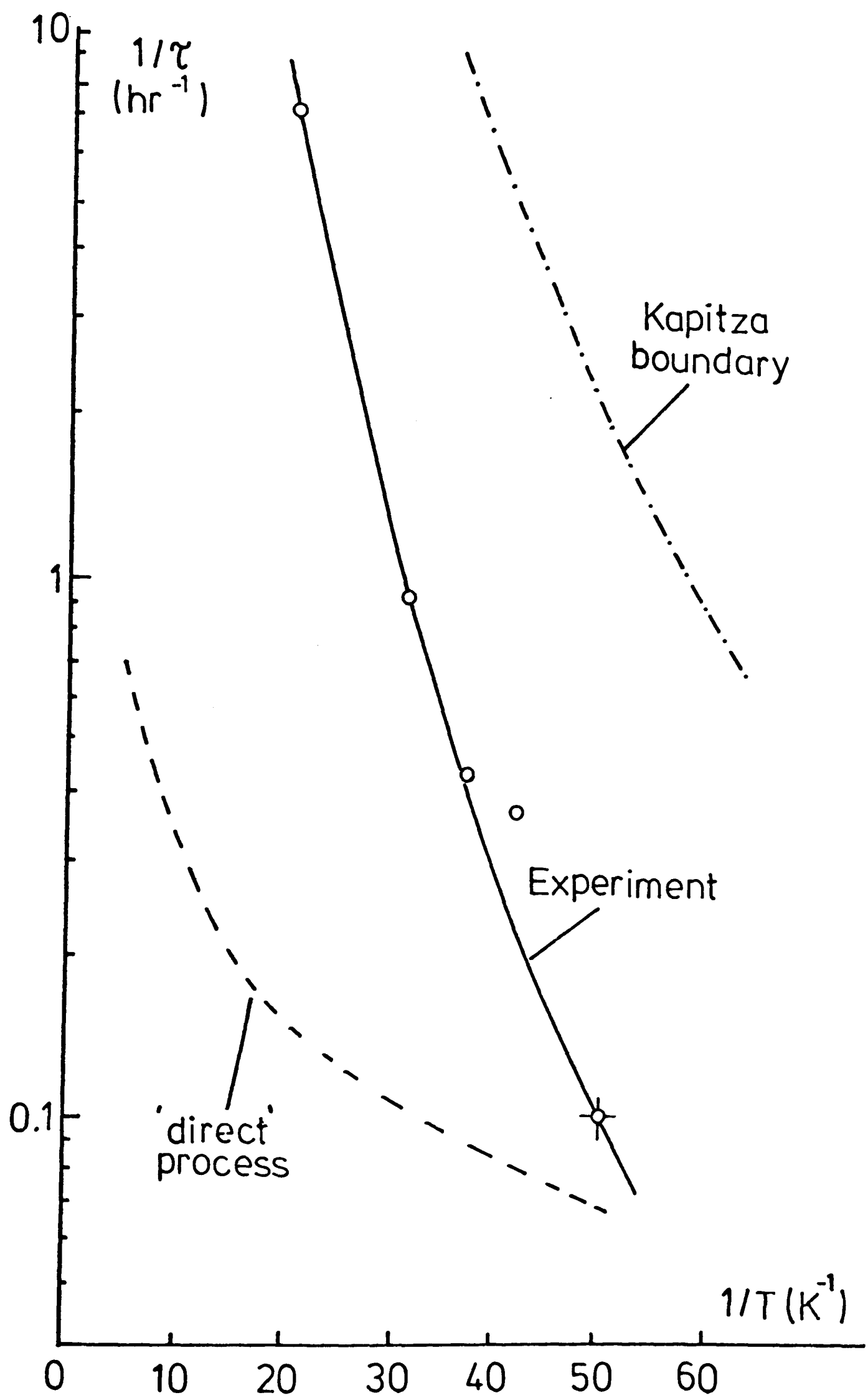


Fig. 4.5 Thermal relaxation time for HoVO_4 at low temperatures

is not clear.

At very low temperatures, the nuclear heat capacity C_S is very much greater than the lattice heat capacity C_L for HoVO_4 . Consequently, the 'effective' (measured) relaxation time τ_o is given by the expression (7).

$$\tau_o = \tau_1 \frac{C_L + C_S}{C_L} \approx \tau_1 \frac{C_S}{C_L} \quad (C_L \ll C_S) \quad 4.11$$

where τ_1 is given by Equation 4.10. This gives relaxation times τ_o which are even longer than the measured τ_s .

The crystal was attached to the 'cold finger' using GE-7031 varnish as explained in section 4.2.1. The phonon mismatch at the insulator-metal interface produces a boundary thermal resistance. Little (8) has considered in detail this boundary resistance in terms of the velocity of sound and phonon distribution in the two media. He has shown theoretically that in general heat transport across such a boundary is given by the expression

$$\dot{Q} = \frac{A}{R_K} \Delta T \quad (\Delta T \ll T) \quad 4.12$$

where $\dot{Q}$ is the rate of heat flow, A the surface area, R_K the boundary resistance $\propto T^{-3}$ and ΔT the temperature drop across the interface. Equation 4.12 has also been shown to be approximately true in many experiments (for example see Reference 9). If the total heat capacity of the solid is C , then the approach to thermal equilibrium is governed by the equation

$$C \delta T = - \frac{A}{R_K} \Delta T \delta t \quad 4.13$$

if we assume perfect thermal conduction inside the solid.

The approach to equilibrium is then exponential with a

characteristic time τ_K , where

$$\tau_K = \frac{C R_K}{A} \quad 4.14$$

For HoVO_4 at these low temperatures most of the heat capacity is nuclear, and is derived from the electric quadrupole splitting of the levels of $^{165}\text{HoVO}_4$, P. In zero field it is approximately given by the equation

$$C = \frac{21 P^2}{2 k T^2} \quad (R) \quad 4.15$$

where T is the temperature. For sample 1:

$$v, \text{ the volume of the crystal} = 0.025 \text{ cm}^3$$

$$\text{Molar volume} = 48 \text{ cm}^3$$

$$\text{Total surface area of contact} = 0.39 \text{ cm}^2$$

Putting these values in Equations 4.14 and taking $R_K = 15 T^{-3} \text{ K s cm}^2 \text{ J}^{-1}$ (9)

$$1/\tau_K \sim 6.8 \times 10^8 T^5 \text{ hrs}^{-1}$$

This is also plotted in Fig. 4.5 and gives relaxation times which are much shorter than the experimental values. This could be explained if the actual area of contact was much smaller than the estimated value of 0.39 cm^2 . Although the enamelled copper wires were uniformly distributed around the crystal, it is quite possible there was only point contact between the two. A reasonably good fit to the data was obtained using an 'effective' surface area $\frac{1}{20} A$.

It does seem that boundary resistance dominates relaxation processes at these low temperatures in HoVO_4 and so it was not possible to determine τ_1 , the nuclear spin-lattice relaxation time. Only an upper limit could be deduced from these measurements.

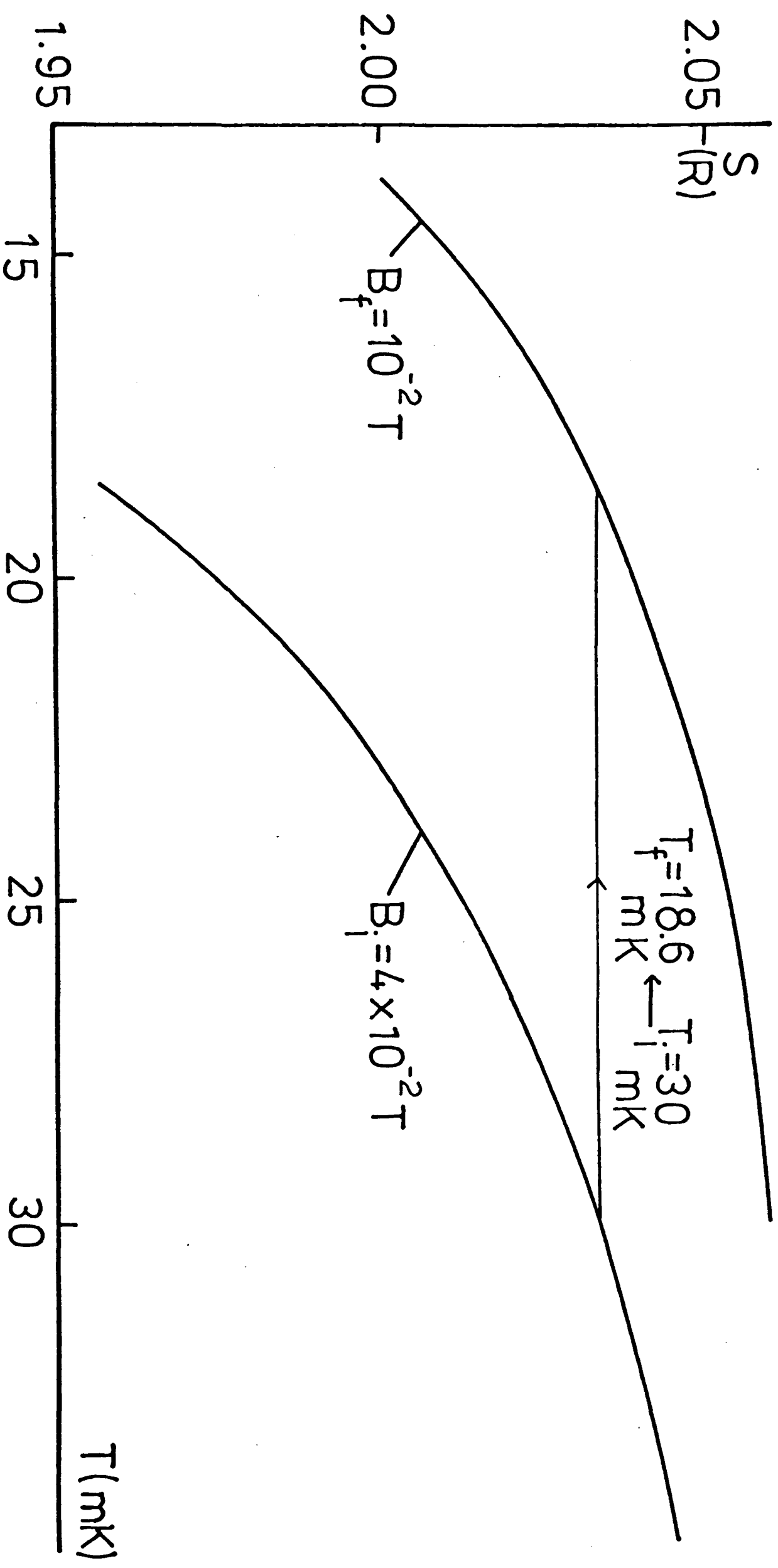


Fig. 4.6 Method for determination of T_f (method (a))

4.3.3 Adiabatic Demagnetisation to Zero Field

A series of approximately adiabatic demagnetisations were done from $T_i = 36$ mK and a number of initial magnetic fields B_i down to $B_f = 2 \times 10^{-3} T$, which was the field due to the leakage current in the power supply. The h.c.p. ^{60}Co Co NO thermometer was used throughout to measure the initial temperature which was maintained at 36 mK by passing a constant current through the mixing-chamber heater of the dilution refrigerator. The frequency, ν , was recorded immediately after demagnetisation, from which nuclear susceptibilities were determined using Equation 4.9 using a filling factor $f = 0.091$. Two methods were used to estimate the temperatures reached after demagnetisation.

(a) Entropy calculations (section 2.7.1) were made using the nuclear Hamiltonian (Equation 2.11) and the final temperature reached, T_f , deduced from the condition that entropy was conserved during demagnetisation. This is justified because the demagnetisations were done in a time short compared with the thermal relaxation time at 36 mK. The method is illustrated in Fig. 4.6. The entropies below the nuclear ordering temperature, T_N , were computed using a Mean Field (MF) approximation details of which are given in section 2.7.1.

(b) At sufficiently high starting temperatures, the temperature reached after adiabatic demagnetisation is given by the expression

$$T_f = \frac{B_f}{B_i} T_i \quad \begin{array}{l} i = \text{initial} \\ f = \text{final} \end{array} \quad 4.16$$

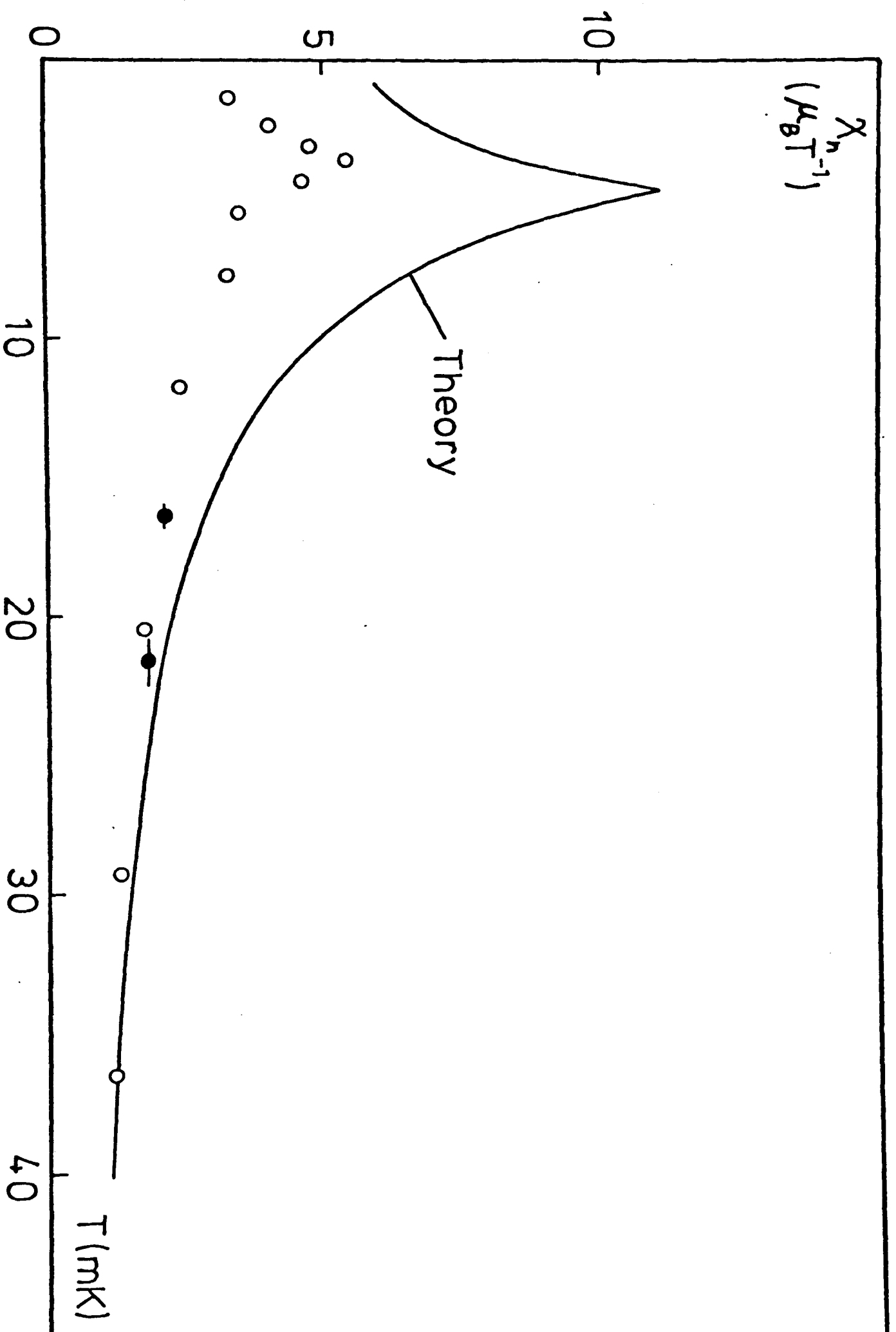


Fig. 4.7 Nuclear r.f. susceptibility vs temperature (NO thermometry for filled in circles)

The final field B_f includes terms to take into account dipolar, exchange and electric quadrupole interactions and is given by the expression (10).

$$B_f^2 = B_{app.}^2 + B_{dip.}^2 + B_{exch.}^2 + \frac{4P^2}{\gamma_h^2} \quad 4.17$$

Where P is the quadrupole interaction and γ_h is the nuclear Zeeman splitting for $^{165}\text{HoVO}_4$. The last three terms can be lumped together to give for $^{165}\text{HoVO}_4$

$$B_f^2 = B_{app.}^2 + (3.56 \times 10^{-2})^2 \text{ tesla}^2 \quad (10)$$

However, at these temperatures (~ 36 mK), the quadrupole splitting is not small compared to the temperature T and therefore Equation 4.17 is not strictly true. This is especially true in low fields where kT is of order the nuclear splittings. For example the entropy calculations for demagnetisations from 2.2×10^{-2} and $4.2 \times 10^{-2}T$ gave $B_{eff.}$ of 2.95×10^{-2} and $2.93 \times 10^{-2}T$ respectively. Using the measured frequency shifts $\delta\nu$ soon after demagnetisation from 36 mK, the nuclear susceptibilities were evaluated using Equation 4.8. From the values obtained for demagnetisations from $2.2 \times 10^{-2}T$ and $4.2 \times 10^{-2}T$ and using the theoretical calculations for the susceptibility, T_f could be determined. Using Equation 4.16 $B_{eff.}$ was calculated to be 5.1×10^{-2} and $4.9 \times 10^{-2}T$ for the two fields. These relatively large values for $B_{eff.}$ indicate a genuine departure from the computed susceptibility curves at the lowest temperatures (Fig. 4.2). For this reason, the entropy calculations of method (a) were used to determine temperatures, T_f . The corrected nuclear susceptibilities (in a field $B_f = 2 \times 10^{-3}T$) are plotted as a function of temperature in Fig. 4.7.

The observed susceptibilities are well below the calculated values: this is probably due to the inadequacy of the MF approximation (see below). The peak in the susceptibility at 4.0 ± 0.2 mK indicates nuclear anti-ferromagnetic ordering. The theoretical curve in Fig. 4.7 is for an antiferromagnet in the MF approximation with domains uniformly distributed in the a - a' plane. There is good experimental evidence to suggest that this is so in applied fields $\lesssim 10^{-2}$ T (see Chapter 5).

In HoVO_4 , the nuclear antiferromagnetic ordering is predominantly due to the dipole-dipole interaction between the enhanced moments of the four nearest neighbours (nn) with a small amount of indirect antiferromagnetic exchange via the O^{2-} ions (section 2.6, 11). This is to be contrasted with most electronic systems where ordering is primarily exchange-induced, that is via the overlap of the electronic orbits. The dipole-dipole interaction between two ions is given by the expression

$$\mathcal{H}_{dd} = \frac{\mu_0}{4\pi} \left[\frac{\underline{\mu}_i \cdot \underline{\mu}_j}{r_{ij}^3} - 3 \frac{(\underline{\mu}_i \cdot \underline{r}_{ij})(\underline{\mu}_j \cdot \underline{r}_{ij})}{r_{ij}^5} \right] \quad 4.18$$

The MF approximation is then equivalent to assuming that each ion is in an 'average' magnetic field generated by the other ions around it, so that

$$\mathcal{H}_{dd} = \frac{\mu_0}{4\pi} \left[\frac{\underline{\mu}_i \cdot \langle \underline{\mu}_j \rangle}{r_{ij}^3} - 3 \frac{(\underline{\mu}_i \cdot \underline{r}_{ij})(\langle \underline{\mu}_j \rangle \cdot \underline{r}_{ij})}{r_{ij}^5} \right] \quad 4.19$$

While the above approximation should be a good one for systems where the ordering is essentially long-range, like HoVO_4 , it does ignore short-range correlation effects.

De Jongh and Miedema (12) have reviewed in some detail these correlation effects in an illuminating discussion.

Quoting from Fisher (13), they define a correlation function between two spins

$$\Gamma_{\underline{r}}(T) = \langle S_o^z S_{\underline{r}}^z \rangle / \frac{1}{3} S(S+1) \quad 4.20$$

($\underline{r} = 0, 1, 2 \dots \infty$)

where the angular brackets $\langle \rangle$ denote an expectation value of the operator product, $\frac{1}{3} S(S+1)$ is a normalisation factor and $\underline{r}$ is the position of its neighbours. These correlation functions and their derivatives can then be used to calculate the thermodynamics of the ordering transition. All the $\Gamma_{\underline{r}}(T)$ s apart from Γ_{∞} have an inflexion point at T_c (Γ_{∞} itself vanishes at T_c). If the magnetic interactions are between the nearest neighbours only, the magnetic exchange energy can be expressed as

$$U_m(T) = - N_o z |J| \langle S_o^z S_1^z \rangle \quad 4.21$$

where N_o is the total number of spins, z the number of nearest neighbours, J the exchange constant and $\langle S_o^z S_1^z \rangle$ is the correlation function between the nearest neighbours. The specific heat capacity C_m , which is $\frac{\partial U_m}{\partial T}$, shows a ' λ -type' anomaly at T_c as Γ_1 has a point of inflexion at this temperature. Moreover, it has a high temperature 'tail' ($T > T_c$) which is indicative of short-range order. On the other hand, the MF approximation shows no such tail. Experiments on many magnetic substances have shown ' λ -type' anomalies near T_c and high temperature 'tails' in the specific heat, thus indicating the importance of short-range order in magnetic systems (12). The specific heat of HoVO_4 in the mK region (in zero applied field) is primarily due to the electric quadrupole interaction which has the same temperature dependence ($C_B \propto T^{-2}$) as the high temperature 'tail'

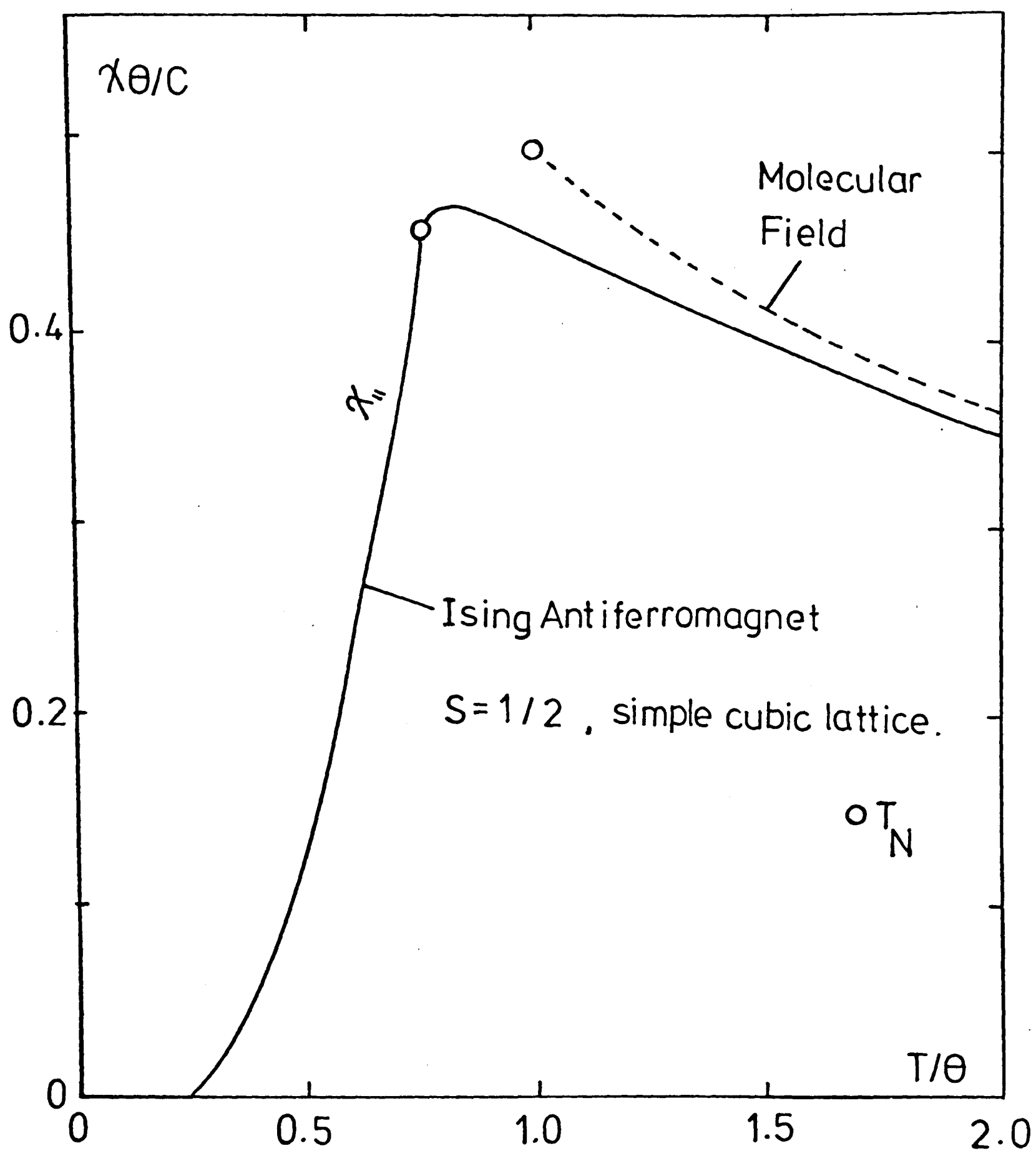


Fig. 4.8 Magnetic susceptibility calculation for an Ising antiferromagnet

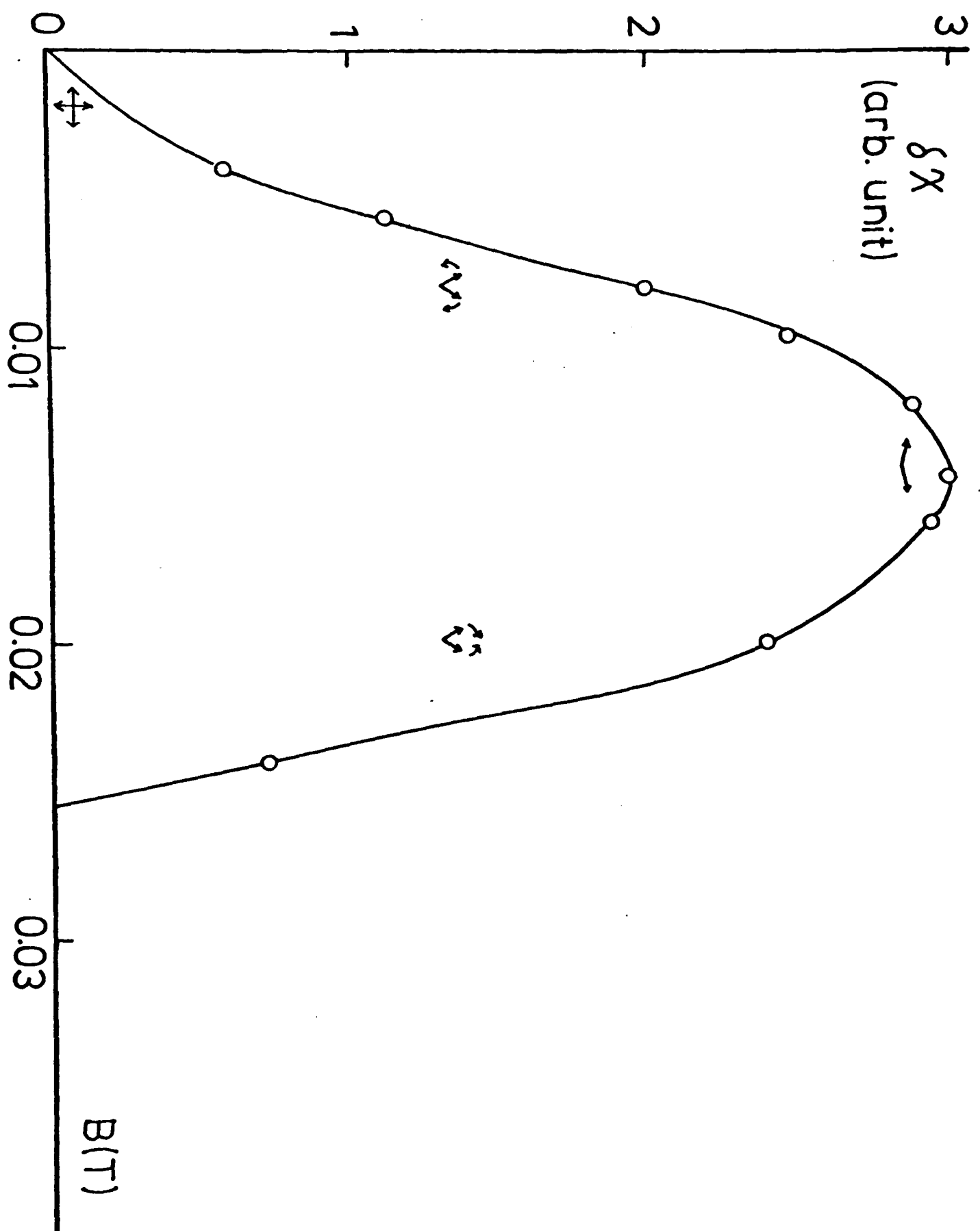


Fig. 4.9 Relative r.f. susceptibility shift vs applied magnetic field

due to short-range order above T_N . This meant that it was not possible to come to any general conclusions regarding the strength of short-range order in HoVO_4 from the nuclear refrigeration experiments of Chapter 8.

Sykes and Fisher (14) have calculated the parallel susceptibility $\chi_{\parallel}$ for a 2 dimensional (2-D) Ising anti-ferromagnet in a simple cubic lattice for $S = \frac{1}{2}$ using an approximate method. This is shown in Fig. 4.8 along with the MF prediction. It is clear from the curves that the MF approximation gives a value for the Néel temperature, T_N , which is high compared to an Ising model. Moreover, the susceptibility above T_N is larger for the MF model. These conclusions are broadly in line with the HoVO_4 measurements. An exact Ising model calculation for HoVO_4 is made difficult by its high spin value ($I = 7/2$) and large electric quadrupole interaction. However, the above arguments indicate that a 2-D Ising model calculation would give a lower T_N and smaller susceptibilities than the corresponding MF predictions.

4.3.4 Adiabatic Susceptibility Below the Ordering Temperature

The r.f. susceptibility was measured as a function of magnetic field below the ordering temperature and is plotted in Fig. 4.9. No hysteresis effects were observed in the magnetisation curve and the mean spin temperature was estimated to be 2.4 ± 0.4 mK from entropy calculations. This temperature is independent of small applied fields and the error includes a contribution from gradual warm up of the specimen during the course of the experiment which took about twenty minutes. The r.f. susceptibility shows a maximum which can be interpreted as follows:

The spins are uniformly distributed in the a - a' plane

in small applied fields. On application of a small magnetic field, ($B_f \ll 3.3 \times 10^{-2} T$, the field at the ion in the ordered state at $T = 0K$) the spins rotate into the 'flop' phase. This is the energetically favoured phase in an applied field (section 2.6.4). In a field of about $1.5 \times 10^{-2} T$ the antiferromagnetic spin 'flop' is complete. Further increases in the B field tends to cause the spins to 'close up' into the ferromagnetic phase, which reduces the susceptibility. In zero applied field, the measured susceptibility is an average of χ_a and $\chi_{a'}$; on applying a field, the 'flop' produces more of $\chi_{a'}$, thus increasing the measured susceptibility ($\chi_{a'} > \chi_a$). The process is represented diagrammatically also in Fig. 4.9. It is not clear why such a relatively large applied field is required to cause the spins to 'flop'. Gamma-ray anisotropy measurements (see Chapter 5) indicate an 'anisotropy field' of about $1.2 \times 10^{-2} T$. It seems likely that the crystal breaks up into domains below the Néel temperature with crystalline imperfections forming pinning centres. The consequences of this model are discussed in section 5.9.

The magnetisation is given by integrating the differential isothermal susceptibility χ_T , with respect to the magnetic field B

$$M_T(B) = \int_0^B \frac{\partial M_T}{\partial B} dB = \int_0^B \chi_T dB \quad 4.22$$

In principle expressing χ_T as a function of χ_S and integrating with respect to B using Equation 4.22, the magnetic moment can be evaluated. However, this functional relationship is not known below T_N and so the moment was not evaluated.

4.3.5 Adiabatic Susceptibility in a Field

R.f. susceptibility measurements were made as a function of applied field at about 200 mK and at the lowest temperature obtainable, about 16 mK. The results at 200 mK were similar to those of Loftus (2) and afforded a check on our r.f. susceptibility measurements. At the highest temperatures, thermal relaxation times were short enough for the lattice and the nuclear spins to be at a common temperature during magnetisation. However, this was not so at the lowest temperatures where thermal relaxation times were of order many hours (see section 4.3.2.).

From thermodynamics

$$\chi_T = \left(\frac{\partial M}{\partial B} \right)_T, \quad \chi_S = \left(\frac{\partial M}{\partial B} \right)_S \quad 4.23$$

where χ_T and χ_S are the isothermal and adiabatic susceptibilities respectively. Using the thermodynamic equation for a magnetic system (For a definition of U see for example Ref. 15)

$$dU = TdS + BdM \quad 4.24$$

and Maxwell's relations, the expressions

$$\frac{\chi_T}{\chi_S} = \frac{C_B}{C_M} \quad 4.25 \quad \text{and} \quad C_B - C_M = \frac{T}{\chi_T} \left(\frac{\partial M}{\partial T} \right)_B^2 \quad 4.26$$

are easily derived. Here C_B and C_M are the specific heat capacities at constant field and magnetisation respectively. To proceed further one requires some knowledge of the nuclear Hamiltonian.

For HoVO_4 , the heat capacity C_B is made up of Zeeman, quadrupole and dipole terms and is given by the expression

$$C_B = \frac{I(I+1)}{3kT^2} (\chi_1 \hbar)^2 (B^2 + B^2 + B_{\text{int.}}^2) \quad 4.27$$

(per ion)

where $\gamma_L \hbar$ is the nuclear Zeeman splitting, the quadrupole splitting

$P = \frac{\gamma_L \hbar B_p}{2}$ and the dipolar interaction $B_{int.}^2 = 1.23 \times 10^{-4} \text{ (tesla)}^2$ (10). At the relatively high temperature of 200 mK, Curie's law is accurately obeyed as the quadrupole splitting is only 1.24 mK. In this case the magnetisation in low fields is given by the expression

$$M = \frac{I(I+1) B (\gamma_L \hbar)^2}{3 kT} \quad (\text{per ion}) \quad 4.28$$

Using Equations 4.25, 4.26 and 4.28

$$C_B - C_M = \frac{I(I+1) (\gamma_L \hbar)^2 B^2}{3 kT^2} \quad (\text{per ion}) \quad 4.29$$

Using Equations 4.25, 4.26 and 4.27

$$\chi_T / \chi_S = C_B / C_M = \frac{B^2 + B^2 + B_{int.}^2}{B^2 + B_{int.}^2} \quad 4.30$$

$$\text{or} \quad \chi_S = \chi_T \frac{B^{*2}}{B^2 + B^{*2}} \quad 4.31$$

$$\text{where } B^{*2} = B_p^2 + B_{int.}^2 = 1.27 \times 10^{-3} \text{ tesla}^2.$$

At temperatures of order 200 mK there is no appreciable depopulation of the Zeeman levels in fields of about 0.2 T and so the isothermal susceptibility χ_T is virtually field independent and can be assumed to be constant at a particular temperature. The adiabatic susceptibility χ_S is field dependent and is given by Equation 4.31.

The susceptibility at 200 mK was fitted as a function of field using Equation 4.31 with B^* as a free parameter. As only relative susceptibilities could be measured by this

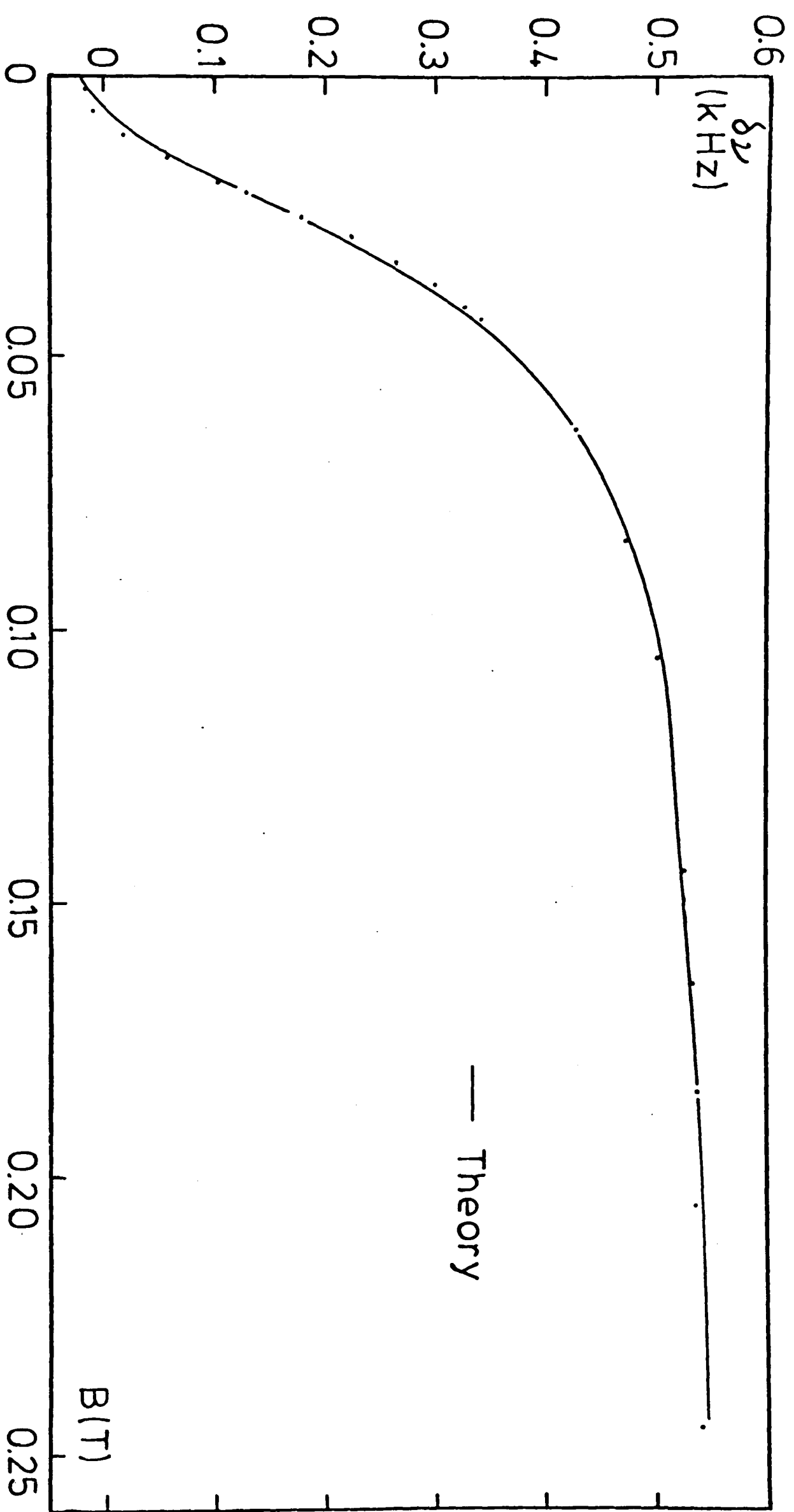


Fig. 4.10 Frequency shift with respect to an arbitrary origin vs magnetic field at $T \sim 200$ mK

technique, Equation 4.31 was rewritten to give in terms of frequency shifts

$$\chi_s(B) = \text{const} (408150 - \nu) \frac{B^{*2}}{B^2 + B^{*2}} \quad 4.32$$

where 408150 is a baseline which corresponded to zero nuclear susceptibility. The best fit was obtained for $B^* = 3.4(1) \times 10^{-2}$ T. A correction of +2.5% should be applied to take into account demagnetisation effects. This takes B^* up to $3.5(1) \times 10^{-2}$ T which brings it into good agreement with the measurements of Bleaney et al who quote a figure of 3.56×10^{-2} T (10). The results are plotted in Fig. 4.10. The good agreement obtained indicates that the bulk of the field dependent χ_s is from the ^{165}Ho nuclei.

Analysis of susceptibility data at $T \sim 15$ mK was complicated by the long thermal relaxation time at this temperature in low applied fields. Two extreme models are applicable.

- (a) The nuclear spins are in thermal contact with the lattice during magnetisation
- (b) The nuclei are essentially decoupled from the lattice throughout the magnetisation process.

In view of the long relaxation time at 15 mK, (b) is expected to be a better approximation than (a). A computer programme was written to calculate χ_s and entropies for various fields and temperatures using the nuclear Hamiltonian known in this temperature range (Equation 2.11). The appropriate entropy conditions were then used to evaluate temperatures and isentropic susceptibilities from which the frequency shifts were evaluated. These theoretical fits are

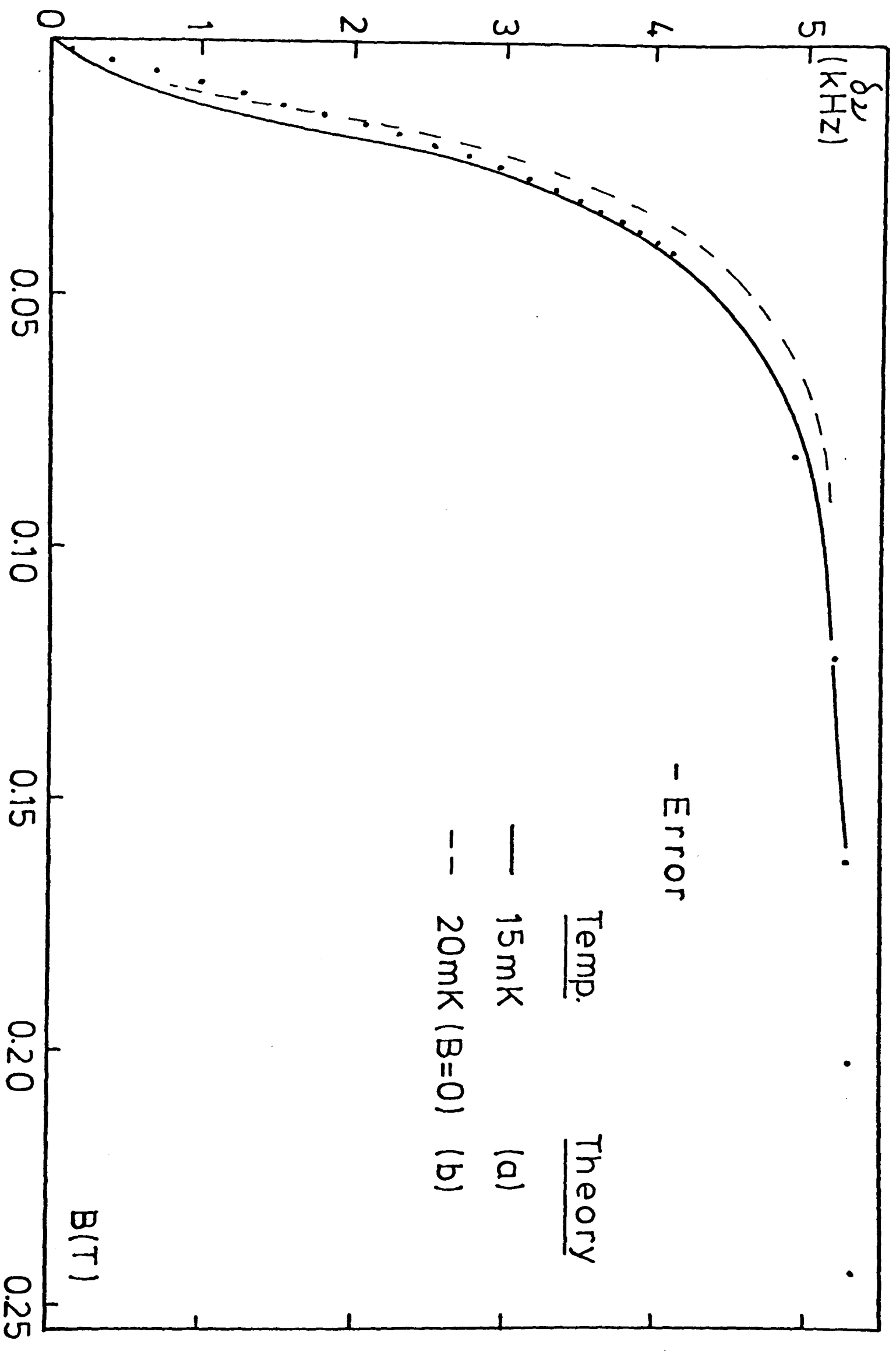


Fig. 4.11 Frequency shift with respect to an arbitrary origin vs magnetic field at $T \sim 15$ mK

plotted in Fig. 4.11 along with the experimental points. Although none of the curves fit the data over the whole field range, model (b) gives a better fit for fields $B < 0.03$ T while (a) seems to be better at the higher fields. This could be explained if spin-lattice relaxation times were shorter in higher fields causing some equilibration to the lattice in higher fields. However, relaxation times are expected to get longer in larger applied fields due to greater separation of the nuclear energy levels which require more energetic phonons to extract the heat of magnetisation from the nuclear system.

4.4 Measurements on Sample 2

Most of the measurements made on this sample were on gamma-ray anisotropy and are discussed in Chapter 5. The rest of the chapter will be devoted to susceptibility measurements on this sample.

Owing to radioactive self-heating, there was a large temperature gradient across the crystal. Although the 'cold finger' was at 12 mK, anisotropy measurements on the 712 keV gamma-ray from ^{166m}Ho indicated a mean crystal temperature of 39 ± 1 mK (section 5.3). R.f. susceptibility measurements, extrapolated from high temperatures gave an average crystal temperature of 50 ± 5 mK. The discrepancy between the two measurements may be due to the difference in thermal averaging: viz. the gamma-ray anisotropy for an E1 transition is proportional to $\langle M_z^2 \rangle$ while the r.f. susceptibility is proportional to $\langle M_z \rangle$, where M_z is the nuclear moment.

Adiabatic demagnetisation from 1.8 T and initial temperature of 39 mK (see above) down to small applied (final) fields were made and the gamma-ray anisotropy and r.f. susceptibility

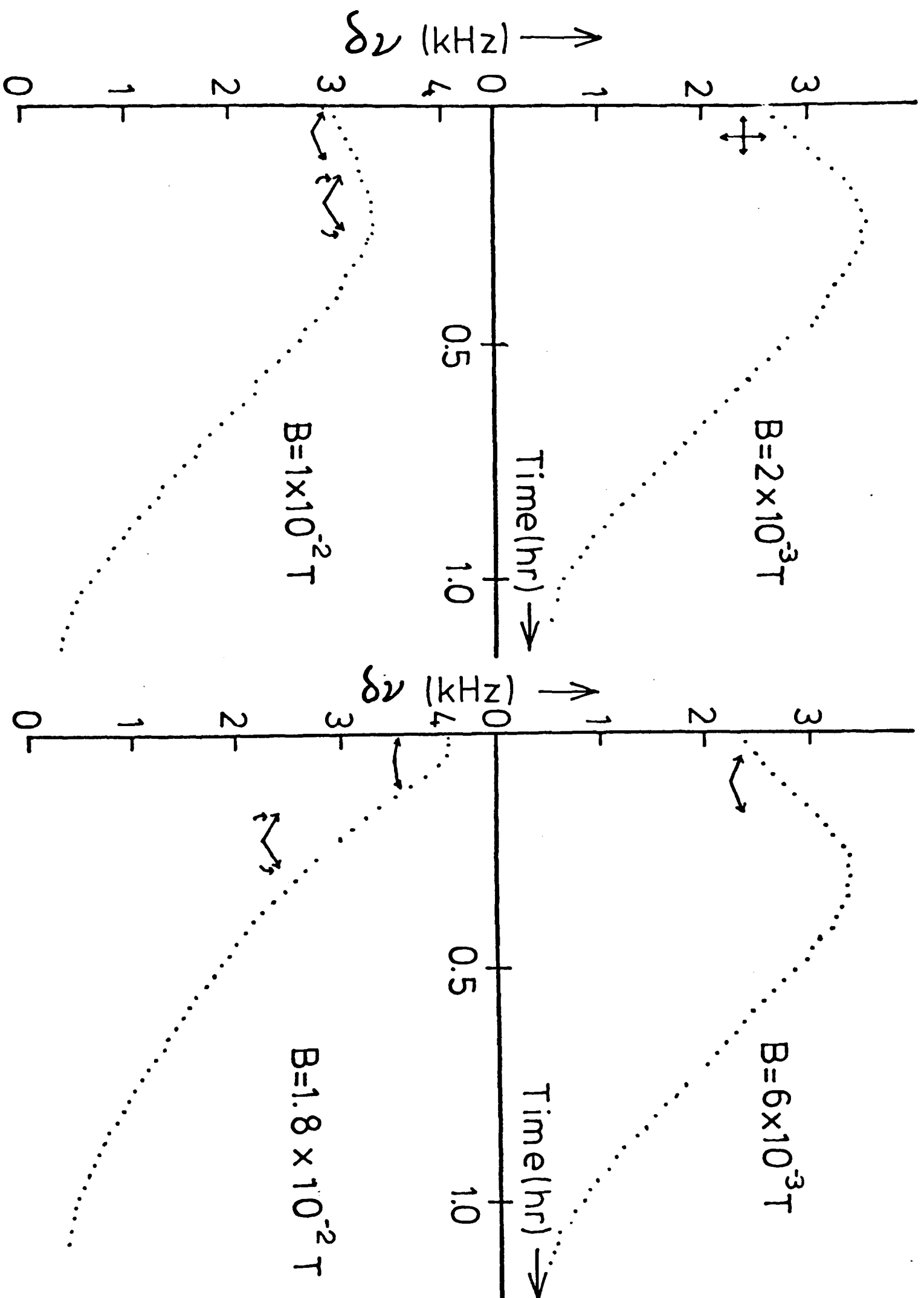


Fig. 4.12 Resonant frequency shift vs time after demagnetisation

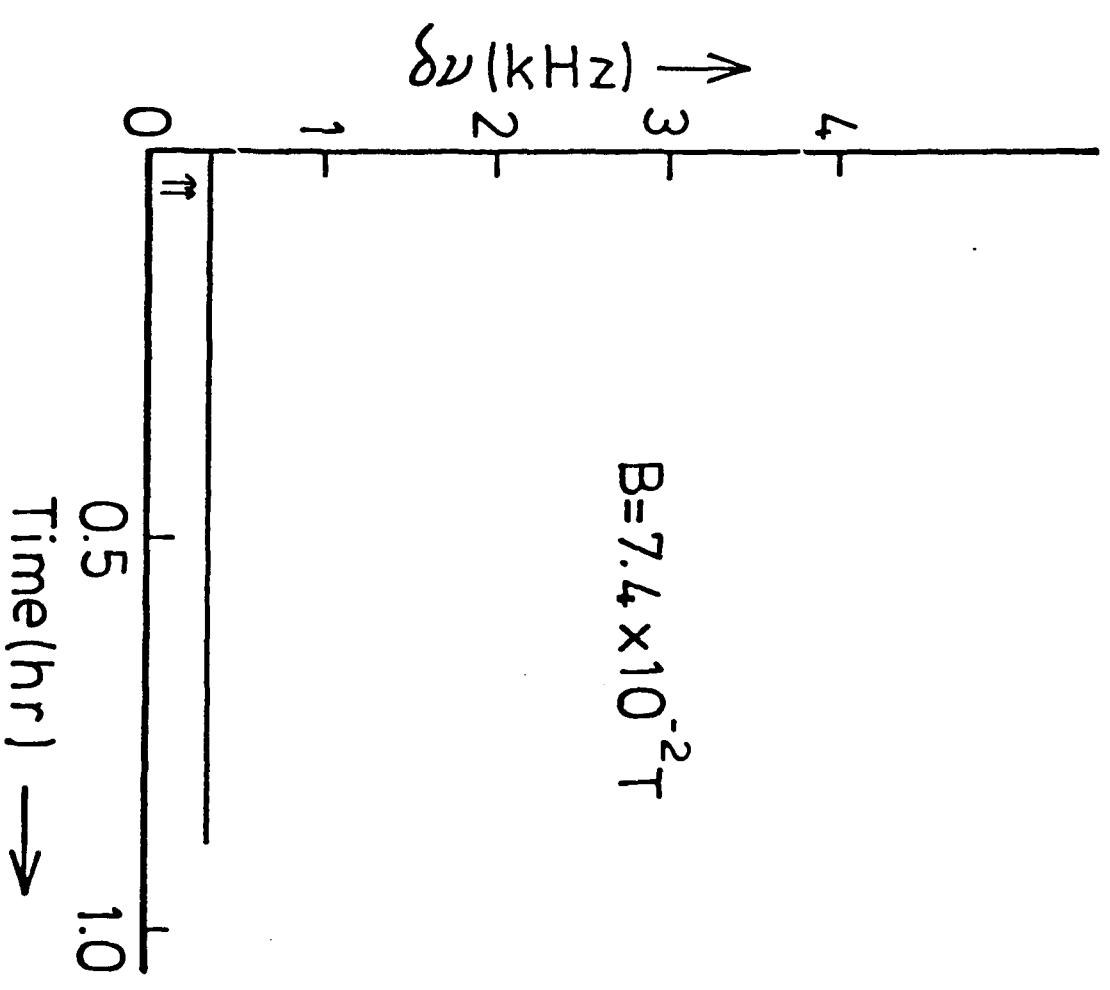
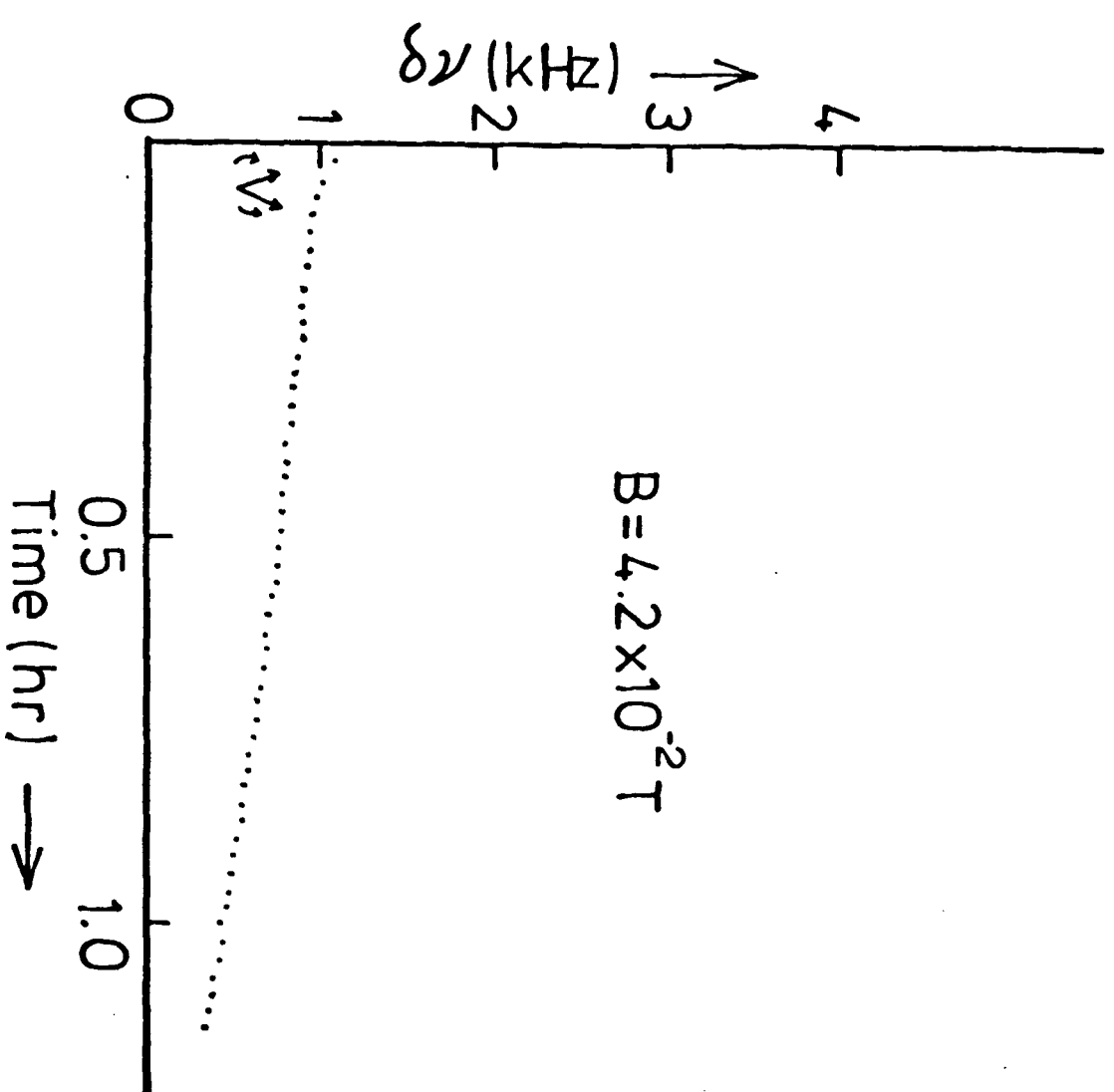


Fig. 4.13 Resonant frequency shift vs time after demagnetisation in relatively large applied fields

were recorded simultaneously. The r.f. susceptibility data are plotted in Figs. 4.12 and 4.13. The ordinate is in units of kHz and is proportional to the uncorrected nuclear susceptibility while the abscissa is in units of hours after demagnetisation.

The following analysis which follows closely section 4.3.4 is consistent with gamma-ray anisotropy measurements which are presented in Chapter 5. As explained in section 4.3.4, the antiferromagnetic domains are uniformly distributed in the a - a' plane in small applied fields, but gradually rotate into the 'flop' phase as the field is increased further. As a result, the measured susceptibility increases to a maximum in an applied field of about 1.8×10^{-2} T. On increasing the field further, the antiferromagnetic ordering is destroyed and the spins 'close up' into the ferromagnetic phase. Thus χ_S is almost independent of temperature in an applied field of 7.4×10^{-2} T. It should be noted that the temperature reached after demagnetisation is independent of small final applied fields. ($B_{app} \ll 3.4 \times 10^{-2}$ T) (see also Chapter 5). This means that any increase of χ_S with applied field is not just due to a higher spin temperature. The possible spin structures are also indicated in Figs. 4.12 and 4.13.

4.5 Conclusions

R.f. susceptibility measurements down to about 1 mK confirmed that HoVO_4 orders antiferromagnetically with $T_N = 4.0$ (2) mK, close to the value of 4.8 mK predicted from the magnetic dipole-dipole interaction of the enhanced nuclear moments (section 2.6). The nuclear antiferromagnetic spin

'flop' phase has been identified in an applied field of about 1.5×10^{-2} T but the exact origin of this relatively large 'anisotropy field' remains unknown. Crystalline defects are thought to produce pinning of Ho nuclei in random directions thus producing a uniform distribution of spin directions in the a-a' plane. On application of a field, the pinning was overcome by interactions between the Ho nuclei. The MF approximation gave susceptibilities which were too high. An Ising 2-D model was predicted to give a better fit to the data but an exact calculation was not attempted due to the high spin ($I = \frac{7}{2}$) and large electric quadrupole interaction ($P = 1.24$ mK).

Thermal relaxation was dominated by boundary resistance between the crystal and the dilution refrigerator and so it was not possible to measure any nuclear spin-lattice relaxation times. An upper limit was obtained, however, which could not be explained by the 'direct' process.

References

Chapter 4

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CHAPTER 5

A NUCLEAR ORIENTATION STUDY OF NUCLEAR ANTIFERRO-
MAGNETIC ORDERING IN HoVO₄

5.1 Synopsis

This chapter describes NO experiments on $^{166\text{m}}\text{HoVO}_4$ down to approximately 1 mK, in several small applied fields. The nuclear antiferromagnetic spin 'flop' phase was identified in a field of about 1.2×10^{-2} T. In zero applied field, the results were consistent with a uniform distribution of spins in the a-a' plane. Even in relatively large applied fields ($\sim 4 \times 10^{-2}$ T), there was some pinning of the spins below about 5 mK so that the 'effective' field at the nucleus was somewhat less than the applied field. This indicated that antiferromagnetic ordering was not completely destroyed in an applied field as large as 4×10^{-2} T.

5.2 Introduction

In the past, magnetic ordering in solids has been investigated by neutron diffraction and Nuclear Orientation. For the latter, the nuclei have to be cooled to very low temperatures. Additionally, the magnetic ion must have a suitable radioactive isotope of known NO parameters. For these reasons NO studies on magnetic ordered systems have been few with notable exceptions.

Turrell et al (1) and Gorling et al (2) demonstrated the spin 'flop' phase in $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ and $\text{MnCl}_2 \cdot \text{H}_2\text{O}$ from measurements of gamma-ray anisotropy of ^{54}Mn incorporated as a dilute impurity in the solid. More recently Marshak and Turrell (3) have reported a study on the helical spin structure of ^{165}Ho metal, from the gamma-ray anisotropy of $^{166\text{m}}\text{Ho}$.

While neutron diffraction is of more general application,

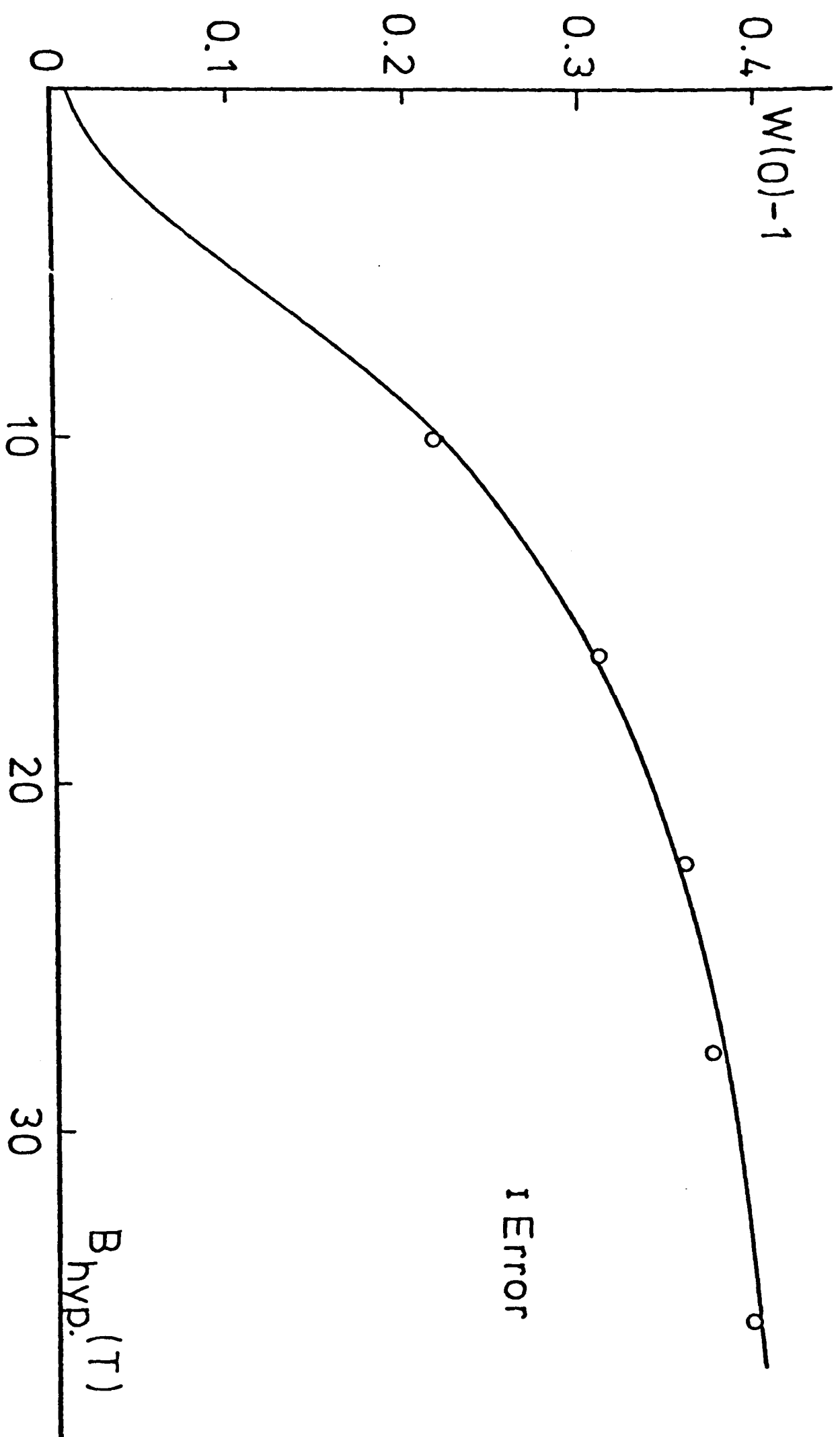
it can only be used successfully in cases where magnetic scattering is comparable with nuclear scattering. Nevertheless, many electronic magnetic ordered systems have been studied by this technique. Recently, in an ingenious experiment, Abragam and co-workers (4) identified several nuclear ordered structures in lithium hydride by this technique. Although nuclear magnetic scattering is negligible as compared with pure nuclear scattering, due to the weakness of nuclear magnetism, the strong interaction responsible for nuclear scattering is spin-dependent. This means that for a polarised nucleus, the scattering is not isotropic but is a function of the polarisation and the angle between the nuclear spin and neutron. The effect, which is large for protons, enabled Abragam et al (5) to obtain strong extra Bragg peaks as the nuclear spins ordered.

The stable isotope of holmium, ^{165}Ho , which is 100% abundant, is easily converted to the long-lived isomeric state $^{166\text{m}}\text{Ho}$ ($t_{1/2} = 1.2 \times 10^4 \text{ y}$, $I = 7^-$) by thermal neutron irradiation. The short-lived state ^{166}Ho ($t_{1/2} \sim 24 \text{ h}$, $I = 0$) soon decays to the stable isotope ^{166}Er . Although $^{166\text{m}}\text{Ho}$ has a rather complicated gamma decay spectrum, sufficient information is available from measurements (6) for the 712 and 810 keV gamma-rays to be used to measure the Nuclear Orientation tensors of Chapter 3. Most of the analysis in this Chapter refers to the 712 keV gamma-ray (pure E1) with some data of poorer quality from the 810 keV gamma-ray (E2 + M1). The two sets of data were found to be broadly self-consistent within the limits of experimental error.

5.3 Experimental Details

As explained in Chapter 4, single crystals of HoVO_4 were irradiated using a thermal neutron flux, and partially annealed in air at 1200°C (section 4.2.4). In the early experiments, (Series 1) the crystal was thermally anchored to the mixing chamber of the dilution refrigerator by means of 200 47SWG enamelled copper wires (see Chapter 4 for further details). This allowed simultaneous measurement of r.f. magnetic susceptibility and gamma-ray anisotropy. The anisotropy measurements of the 712 keV gamma-ray as a function of applied magnetic field was used to estimate the mean $^{166\text{m}}\text{Ho}$ nuclear spin temperature. Fig. 5.1 shows one such set with the 'best fit' calculated for percentage in good sites $z = 88$ (2.5)% (see section 5.4) and spin temperature of $^{166\text{m}}\text{Ho} = 39$ (1) mK. The nuclear magnetic dipole moment of $^{166\text{m}}\text{Ho}$ was taken to be $3.60 \mu_N$ (see Chapter 6) and $A_2(\gamma)$ for the 712 keV gamma-ray was calculated to be 0.273. The rather poor percentage in good sites is thought to be due to lattice imperfections e.g. defects, still remaining due to imperfect annealing.

To investigate the temperature gradient across the crystal, a separate experiment was done with a similar crystal attached to the copper 'cold finger' by means of high conductivity silver paint (supplied by Acheson Colloids). A $^{54}\text{MnNi}$ NO thermometer was fixed to the top face of the crystal, also using silver paint, but making sure that the thermometer was cooled by contact to the crystal only. The radioactive heating in the thermometer was small and it was hoped to measure accurately the lattice temperature of the top face of the crystal in this way. The measurements



1 Error

Fig. 5.1 Anisotropy of the 712 keV gamma-ray vs magnetic field at 39 mK

indicated that the top face of the crystal was at a temperature of 51 (3) mK although the 'cold finger' was at about 13 (1) mK (see section 4.4). Thus a substantial temperature gradient was found to exist across the crystal.

In a later set of experiments (Series 2) made months later, the crystal was attached to the 'cold finger' by means of high conductivity silver paint. Due to reduction in radioactive self-heating from the decay of the principal impurity ^{160}Tb ($t_{1/2} = 72\text{d}$), a mean crystal temperature of approximately 30 mK was obtained in this set of experiments. Moreover, on adiabatic demagnetisation low temperatures were maintained for a longer period of time. By using a lower applied field in the second series of Runs, approximately the same ratios B_i/T_i were achieved in both sets of experiments. However, as we shall see later, lower temperatures were reached in Series 2.

Several adiabatic demagnetisations were made from applied fields of 2.4 T and 1.8 T for Series 1 and Series 2 respectively. Typically, the magnetic field was left on for about 4 hours so that the heat of magnetisation could be removed from the crystal. The field was then reduced to the required low value in about 10 minutes using a linear sweep unit. The detectors were then moved into position and 400 second counts started immediately. (The Ge (Li) detectors were very sensitive to magnetic fields and had to be shielded from the stray field of the polariser magnet). In the early experiments, data from the relevant channels were punched on to paper tape at the end of each count and analysed using the NOVA 1200 computer. Later, when a new system had been devised and built by J. Siertsema of the Electronic Workshop of the

Clarendon Laboratory (section 1.6.3), the data were directly stored on magnetic tape. When the crystal had returned to 'base' temperature, it was heated to about 100 mK and normalising or 'warm' counts were taken in zero applied field.

5.4 Data Analysis

Gamma-ray pulses from the axial and equatorial Ge (Li) detectors were summed and fed into the full 4096 channel memory of the MCA. 400 second counts were taken until the anisotropy in both detectors had dropped to a low value. In general, about ten counts were found to be sufficient.

The anisotropies were evaluated from the expression

$$W(\theta, \phi) - 1 = \frac{\text{'cold' count} - \text{'warm' count}}{\text{'warm' count}} \quad 5.1$$

(see section 1.7) where 'warm' refers to high temperatures, where the gamma-ray emission is isotropic. A small correction ($\sim 1\%$) had to be made to the 'warm' count at 100 mK to take into account gamma-ray anisotropy due to the electric quadrupole interaction. As explained in Chapter 3, we can unite for the 712 keV gamma-ray

$$\begin{aligned} W_a(0) - 1 &= Q_2 \, 1.0573 \, \rho_0^2 \\ W_{a'}(\pi/2, \pi/2) - 1 &= Q_2 \, (-0.5287 \, \rho_0^2 - 1.2949 \, \rho_2^2) \\ W_c(\pi/2, 0) - 1 &= Q_2 \, (-0.5287 \, \rho_0^2 + 1.2949 \, \rho_2^2) \end{aligned} \quad 5.2$$

where W_a , $W_{a'}$, and W_c are the anisotropies measured along the a , a' , c crystal axes respectively, and ρ_0^2 , ρ_2^2 are statistical tensors describing the orientation of the parent nucleus.

After the first series of experiments, it was found at disassembly that there was a slight misorientation of the

crystal so that the equatorial detector (a'-axis) was at $\phi = 11^\circ$ (2), $\theta = 90^\circ$. By putting in the appropriate values for the spherical polar co-ordinates θ and ϕ , the following corrected equations are easily derived (Equation 3.17)

$$\begin{aligned} W'_a - 1 &= Q_2 \cdot 1.0573 \rho_0^2 \\ W'_{a'} - 1 &= Q_2 (-0.5287 \rho_0^2 - 1.2006 \rho_2^2) \\ W'_c - 1 &= Q_2 (-0.5287 \rho_0^2 + 1.2006 \rho_2^2) \end{aligned} \quad 5.3$$

The primes refer to the directions of the detectors. Equations 5.2 and 5.3 were used to evaluate statistical tensors ρ_0^2 and ρ_2^2 . It should be noted that the measured orientation tensors are thermal averages over the whole crystal. Where several 'canting' angles existed through the crystal, the orientation tensors were averages over the possible angles.

5.5.1 Percentage in Good Sites

As stated in section 5.3, anisotropy data from the 712 keV gamma-ray could be interpreted to show that only about 88% of the nuclei experienced the full magnetic hyperfine field. In this simple model the rest (12%) of the nuclei are assumed to experience no hyperfine field at all. This model has been found to be a good approximation for impurities in metal hosts. NMR measurements on $^{165}\text{HoVO}_4$ at 42K by M.R. Wells of the Clarendon Laboratory showed that the quadrupole-split lines were broadened as a result of neutron irradiation. This is consistent with the percentage in good sites, z , being less than 100%. However, it is not clear how this would affect the e.f.g. at the nucleus. As discussed in section 2.4, the e.f.g. at a holmium nuclear site is the sum of three contributions. These are respectively the

pseudo-quadrupole interaction arising from a second order term in the nuclear Hamiltonian and terms from the 4f electrons and lattice. All three terms make significant contributions to the e.f.g. and may change in different ways as a result of neutron irradiation. Thus it is not possible to predict the 'effective' percentage in good sites for the electric quadrupole interaction. We therefore set z to be 100% for the electric quadrupole interaction for the NO data of this chapter. If this were not true the estimate of temperatures would be systematically high by about 30% at 1 mK.

5.5.2 Nuclear Spin Temperature of ^{166m}Ho and ^{165}Ho

The question arises as to whether the nuclear species ^{166m}Ho and ^{165}Ho reach a common spin temperature in applied fields up to about 1 tesla. A calculation by Bleaney (7) indicated that cross-relaxation between the two nuclear species should be fast enough to ensure a common spin temperature even in fields as large as 1 tesla. We therefore take the ^{165}Ho nuclear spin system to be a large heat bath in thermal equilibrium with the ^{166m}Ho nuclei. In any case, in low fields of ~ 50 mT, dipolar broadening is expected to ensure fast cross-relaxation between the two nuclear species due to crossing of nuclear energy levels. Thus we can safely assume that in the low field demagnetisation experiments to be discussed the two spin systems were at a common temperature.

5.6 Accuracy of NO Temperature Measurement

There were several factors which contributed to the uncertainty in the temperature estimated from the gamma-ray anisotropy of 712 keV gamma-ray. First of all, there was the intrinsic statistical accuracy associated with the gamma-

ray emission which produced an uncertainty in temperature of about 15% at 1 mK for a 400 second count. This error is shown in the form of error bars on the appropriate figures. The electric quadrupole splitting for $^{166\text{m}}\text{Ho}$ is known to an accuracy of about 10%. This gave a total error of $\sim 17\%$ at 1 mK. As explained in section 5.4, there could be a further systematic error due to some of the nuclei not experiencing the full e.f.g.

5.7 Nuclear Orientation Results

A series of adiabatic demagnetisations were made from the initial conditions, $B_i = 2.4$ T and $T_i = 39$ mK for Series 1 and $B_i = 1.8$ T, $T_i = 30$ mK for Series 2, down to several low residual fields B_f . Although the nuclear spin entropy removed in the two series of experiments was comparable, lower temperatures were reached in Series 2, perhaps because the demagnetisations were more nearly adiabatic in the latter owing to reduction in radioactive self-heating. The following sections describe these experiments and unless otherwise stated, relate to data from the 712 keV gamma-ray only. Except for the relatively high field Runs of 1.8×10^{-2} T and 4.2×10^{-2} T, the data from the two series of experiments will be presented separately.

Series 1

5.7.1 $B_f = 2 \times 10^{-3}$ T

Two demagnetisations were made to the lowest residual current of the magnet power supply. The gamma-ray detectors were positioned along the crystal axes a and c in the first experiment (Run 3) and along a and a' in the second (Run 4).

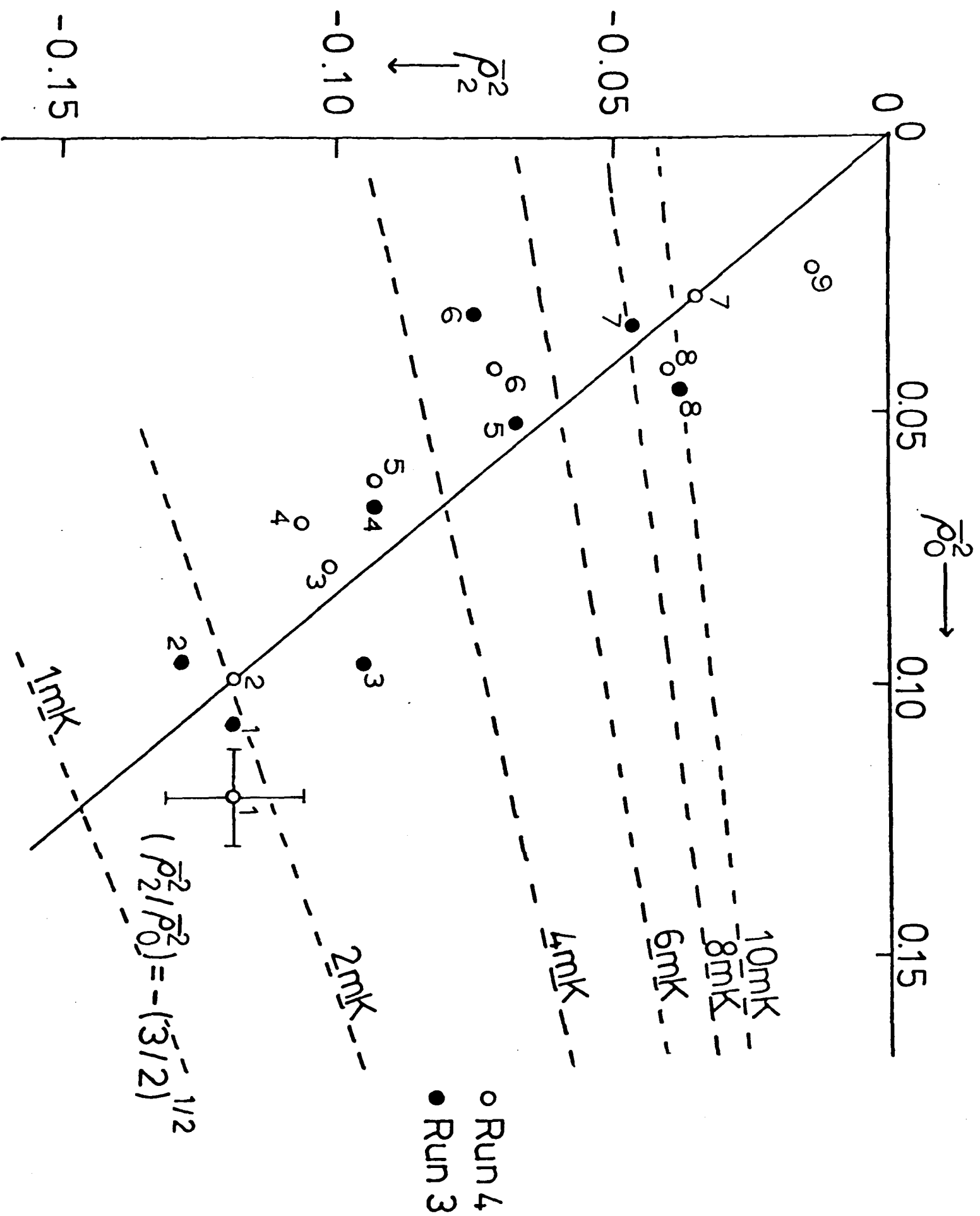


Fig. 5.2 Statistical tensor plot of $\bar{\rho}_2^2$ vs $\bar{\rho}_0^2$ for $B_f = 2 \times 10^{-3}$ T

The average statistical tensors ρ_0^2 and ρ_2^2 were extracted from each set of anisotropy data and are presented in Fig. 5.2., where each data point represents a 400 sec. count and the numbers denote the sequence in which they were made. Both sets of data points lie approximately on a straight line of slope

$$\bar{\rho}_2^2 / \bar{\rho}_0^2 = - \left(\frac{3}{2}\right)^{\frac{1}{2}}$$

As explained in section 3.8, this corresponds to the spins either canted at 45° to the field in the a-a' plane, or uniformly distributed in the a-a' plane. $\bar{\rho}_2^2$ and $\bar{\rho}_0^2$ measurements are unable to distinguish between these two possible spin structures. In fact, a pure quadrupole interaction along the crystal c-axis also gives a straight line of the same slope.

In an effort to resolve this ambiguity, gamma-ray anisotropy data from the 810 keV gamma-ray were also analysed. This gamma-ray has a large A_4 (γ) term which allows measurement of statistical tensors of rank 4. However, from Equation 3.24 it is clear that a small misorientation of the crystal with respect to the detectors can cause a relatively large error in the measured statistical tensors of rank 4. For this reason, data from Series 1 were not used to analyse the fourth rank tensor component of the 810 keV gamma-ray. Section 5.8.2, (Series 2) which discusses the 810 keV gamma-ray, shows that the data are consistent with the nuclear spins uniformly distributed in the a-a' plane, but not with a canting angle $\beta = 45^\circ$.

Also shown in Fig. 5.2 are the isotherms for $^{166m}\text{P} = + 3.4 \text{ MHz}$. The average nuclear temperature reached after

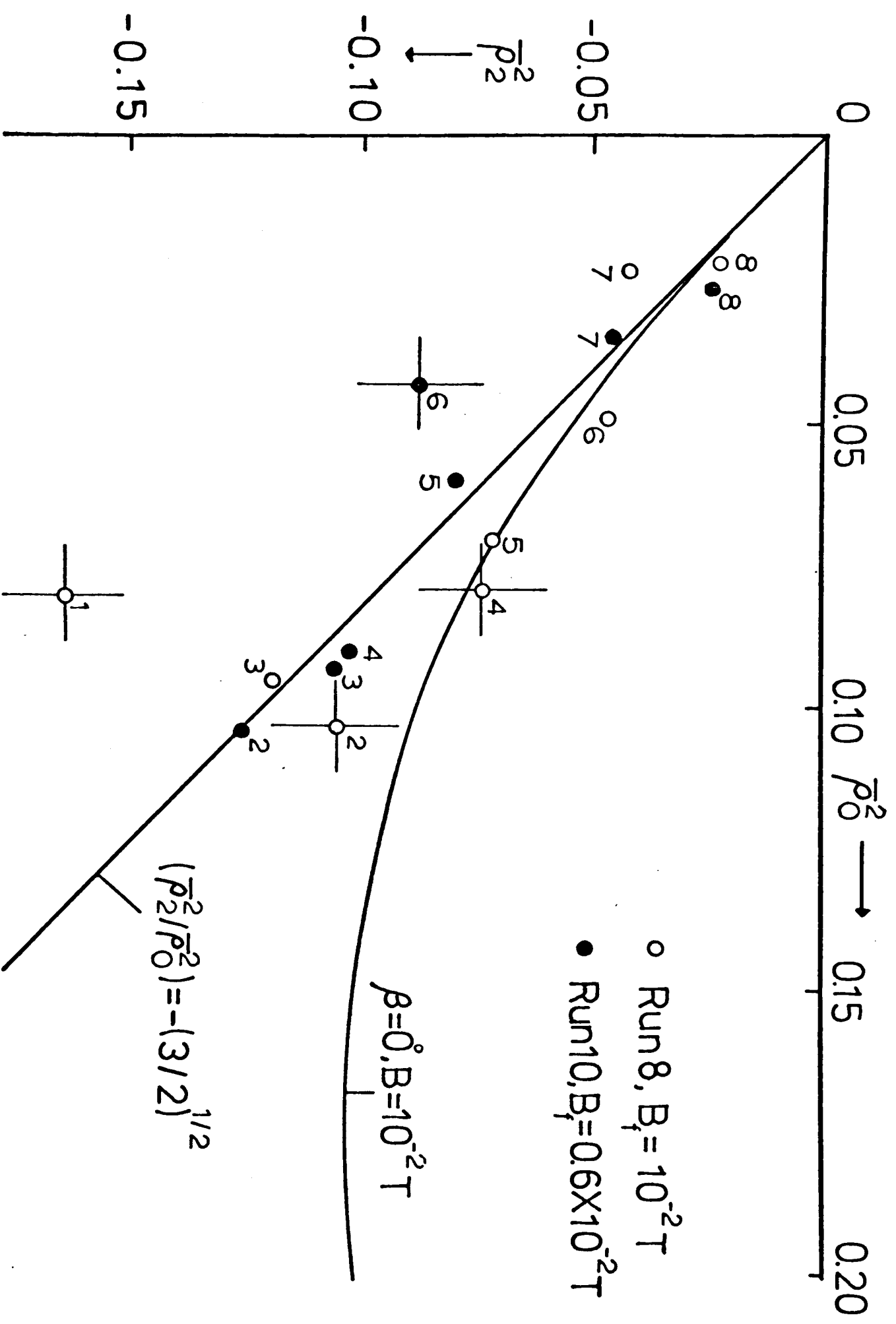


Fig. 5.3 Statistical tensor plot of $\bar{\rho}_2^2$ vs $\bar{\rho}_0^2$ for $B_f = 10^{-2} \text{ T}$ and $B_f = 0.6 \times 10^{-2} \text{ T}$

adiabatic demagnetisation, T_f , was 1.8 (3) mK, which is somewhat higher than the value of 1.0 (1) mK calculated from entropy consideration. The discrepancy may be due to imperfect adiabatic conditions owing to the relatively large radioactive self-heating in these experiments.

5.7.2 $B_f = 1.0 \times 10^{-2}$ T

In this experiment (Run 8), a residual field of 1.0×10^{-2} T was maintained in the superconducting magnets and the counters were placed along the a and c axes respectively. The extracted $\bar{\rho}_0^2$ and $\bar{\rho}_2^2$ parameters are plotted in Fig. 5.3. The plot shows that apart from the first data point, all the data points fall approximately on a straight line with slope

$$\bar{\rho}_2^2 / \bar{\rho}_0^2 = -\left(\frac{3}{2}\right)^{\frac{1}{2}}$$

The first point clearly shows that the nuclear spins have started to 'flop', that is the average canting angle β is greater than 45° . The lowest temperature reached was about 1 mK, but the crystal warmed up rapidly to nearly 2 mK in about 400 seconds. The lower temperature reached in this Run as compared with Runs 3 and 4 may be due to better starting conditions. A calculation (Fig. 2.5) showed that the nuclear heat capacity drops off rapidly below the Néel temperature. This could explain the rapid warm up of the spins from 1 mK to 2 mK in 400 seconds.

Even at the relatively low temperature of 2 mK, a field of 1.0×10^{-2} T is apparently unable to cause the spins to 'flop' appreciably in direct contradiction with the predictions of section 2.6.4.

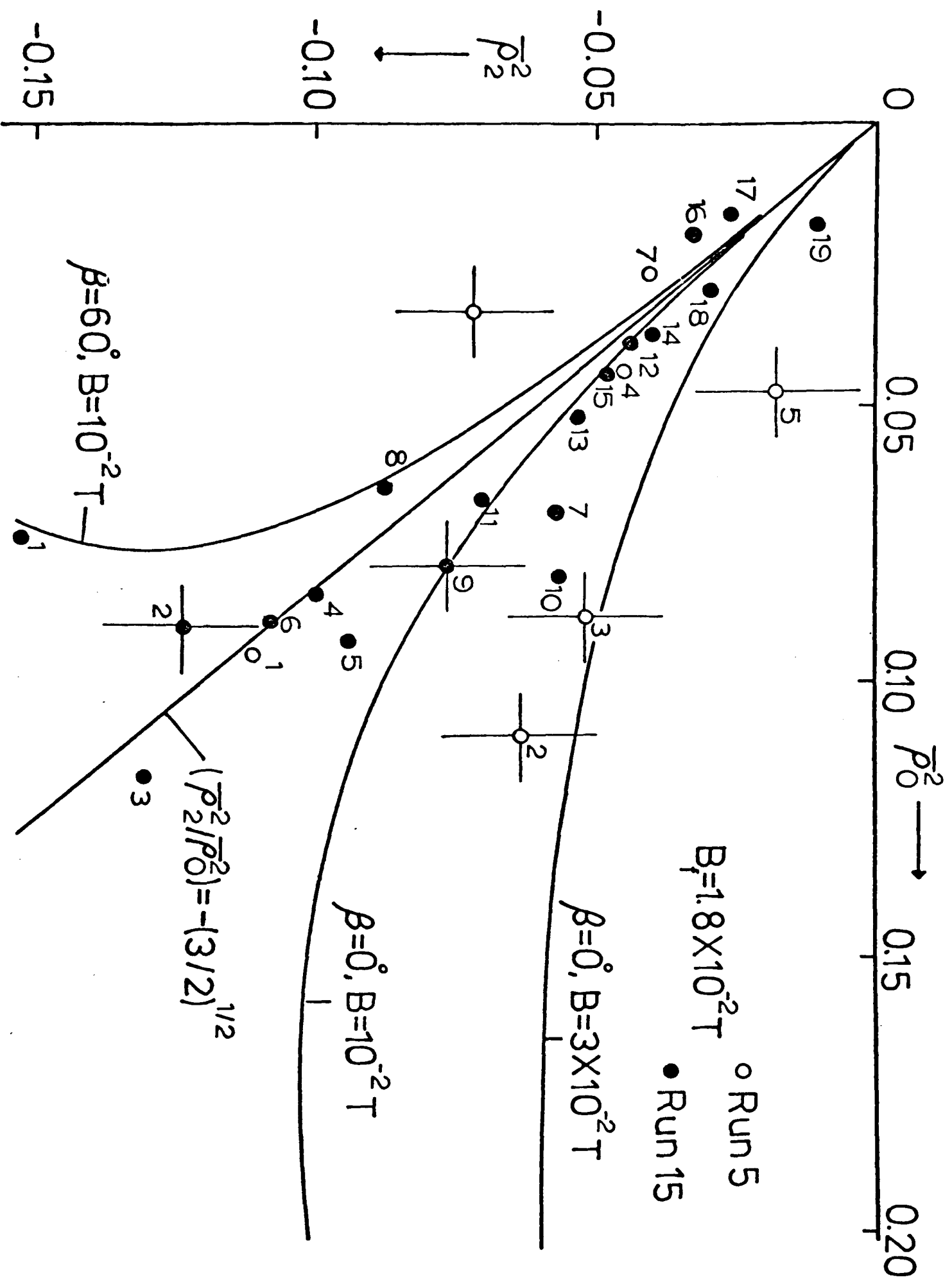


Fig. 5.4 Statistical tensor plot of $\bar{\rho}_2^2$ vs $\bar{\rho}_0^2$ for $B_f = 1.8 \times 10^{-2} T$

$$5.7.3 \quad \underline{B_f = 0.6 \times 10^{-2} \text{ T}}$$

In order to observe the spin 'flop' a demagnetisation to $0.6 \times 10^{-2} \text{ T}$ was done (Run 10) with the same detector geometry as section 5.7.2. Unfortunately, the first five minutes of counting time were lost and so the spin 'flop' was not observed. The data are also presented in Fig. 5.3. The lowest temperature reached is estimated to be $<2 \text{ mK}$, taking into account the first five minutes lost. Entropy calculations showed that the nuclear spin entropy was insensitive to small applied fields. Although the spins canted appreciably, the entropies were unaltered even in fields as large as $1.6 \times 10^{-2} \text{ T}$. One can therefore assume that the temperature reached on adiabatic demagnetisation is independent of small applied fields. Once again, the data points fall on the 'pseudo-quadrupole' line.

5.8 High Field Runs

Several relatively large residual field Runs were made in both series of experiments and are presented together for convenience.

$$5.8.1 \quad \underline{B_f = 1.8 \times 10^{-2} \text{ T}}$$

A total of two Runs were made down to an applied field of $1.8 \times 10^{-2} \text{ T}$. The lowest temperature reached in Series 1 was just above 2.0 mK while about 1.5 mK was reached in Series 2 (Fig. 5.4). Theory (section 2.6.4) suggests that at 0K an applied field of about $1.6 \times 10^{-2} \text{ T}$ is enough to drive the nuclear spins into the ferromagnetic ('flip') phase. At a finite temperature ($T > 0\text{K}$) the nuclear spins are not fully polarised and so the spin 'flip' will occur in a larger applied

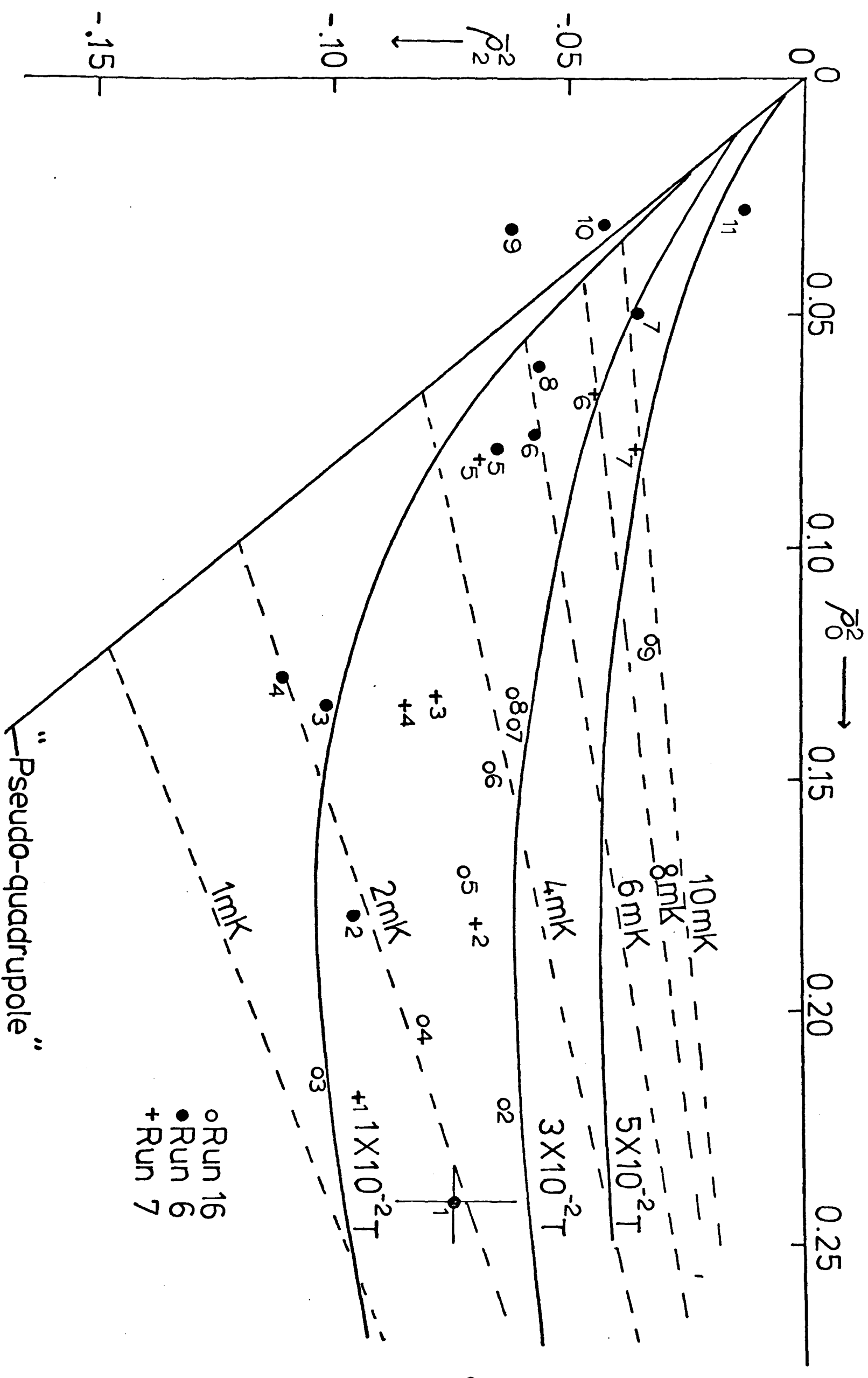


Fig. 5.5 Statistical tensor plot of $\bar{\rho}_2^2$ vs $\bar{\rho}_0^2$ for $B_I=4.2 \times 10^{-2}$ T

field. This could explain the results for these Runs, which indicate that the spins are in the antiferromagnetic phase in an applied field of 1.8×10^{-2} T. In fact the first data point of Series 2 gives a canting angle $>45^\circ$, that is it is partially 'flopping'. Also plotted in the figure are theoretical curves for applied fields of 1×10^{-2} T and 3×10^{-2} T for $\beta = 0$ (ferromagnetic phase). If the spins were in the ferromagnetic phase, the points would lie about half-way between these two curves, which is clearly not the case at the lowest temperatures.

$$5.8.2 \quad \underline{B_f = 4.2 \times 10^{-2} \text{ T}}$$

Two Runs (6, 7) were made in the first Series and one (Run 16) in the second. For Runs 6 and 7, the detectors were placed along a and c crystal directions respectively, while for Run 16 the detectors were directed along the axes a and a'. The data from these Runs, which were broadly self-consistent, are plotted in Fig. 5.5. The lowest temperature reached was 1.5 (3) mK. An entropy calculation gave a value for T_f of 1.2 mK, in good agreement with the measured value. In the entropy calculation, it was assumed that the spins were in the paramagnetic phase and a small correction was made to take into account the difference between the field at the ion and the applied field.

Inspection of Fig. 5.5 shows that the data points lie on a curve, approximately half-way between the theoretical curves for $B = 1.0 \times 10^{-2}$ T and $B = 3 \times 10^{-2}$ T for a canting angle $\beta = 0^\circ$ (ferromagnetic phase). The MF model of Chapter 2 predicts that in an applied field $B > 1.6 \times 10^{-2}$ T, the spins would be in the 'flip' (ferromagnetic) phase, that is

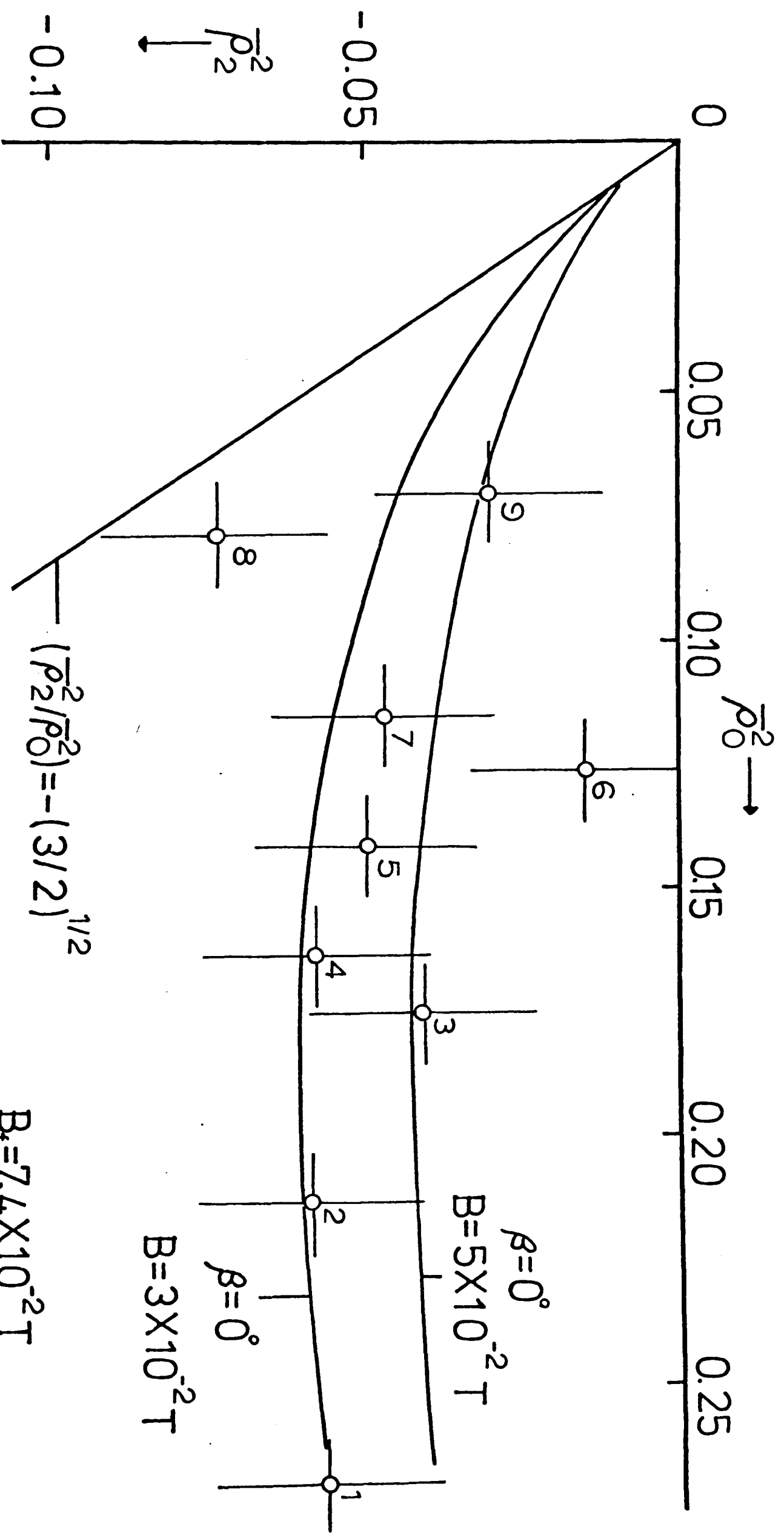


Fig. 5.6 Statistical tensor plot of $\bar{\rho}_2^2$ vs $\bar{\rho}_0^2$ for $B_f = 7.4 \times 10^{-2} \text{ T}$

with the spins oriented parallel to the applied field. If this were true, the field at the ion is expected to be close to the applied field (apart from a correction of about 1% for demagnetisation effects), and the data points would lie on a curve for $B = 4.2 \times 10^{-2}$ T, $\beta = 0$. Since this is not so, it is clear that there is some nuclear antiferromagnetic ordering even at these relatively high fields. As the statistical tensors are sensitive to the ratio of the quadrupole parameter P to the nuclear Zeeman interaction term, one could obtain better agreement with theory by increasing the value of P , perhaps by a factor of two. However, this would increase all the temperature estimates by approximately the same factor which would disagree with the entropy calculations. In any case, it seems unlikely that the quadrupole parameter is in error by this amount.

5.8.3 $B_f = 7.4 \times 10^{-2}$ T

Fig. 5.6 is a plot of $\bar{\rho}_2^2$ vs $\bar{\rho}_0^2$ for a demagnetisation to a residual applied field of 7.4×10^{-2} T. The two detectors were placed parallel to the crystal a and c axes respectively. From the statistical tensor plot, the lowest temperature reached is estimated to be 2.4 (5) mK in good agreement with an entropy calculation of 2.0 mK. Once again, the magnetic field at the ion is appreciably smaller than the applied field of 7.4×10^{-2} T. Reasonable agreement with theory is obtained if B is taken to be $4 (1) \times 10^{-2}$ T, with $\beta = 0$.

5.9 Series 2

The spin 'flop' phase was identified in this series of experiments, with data for $B_f = 1.2 \times 10^{-2}$ T showing it most

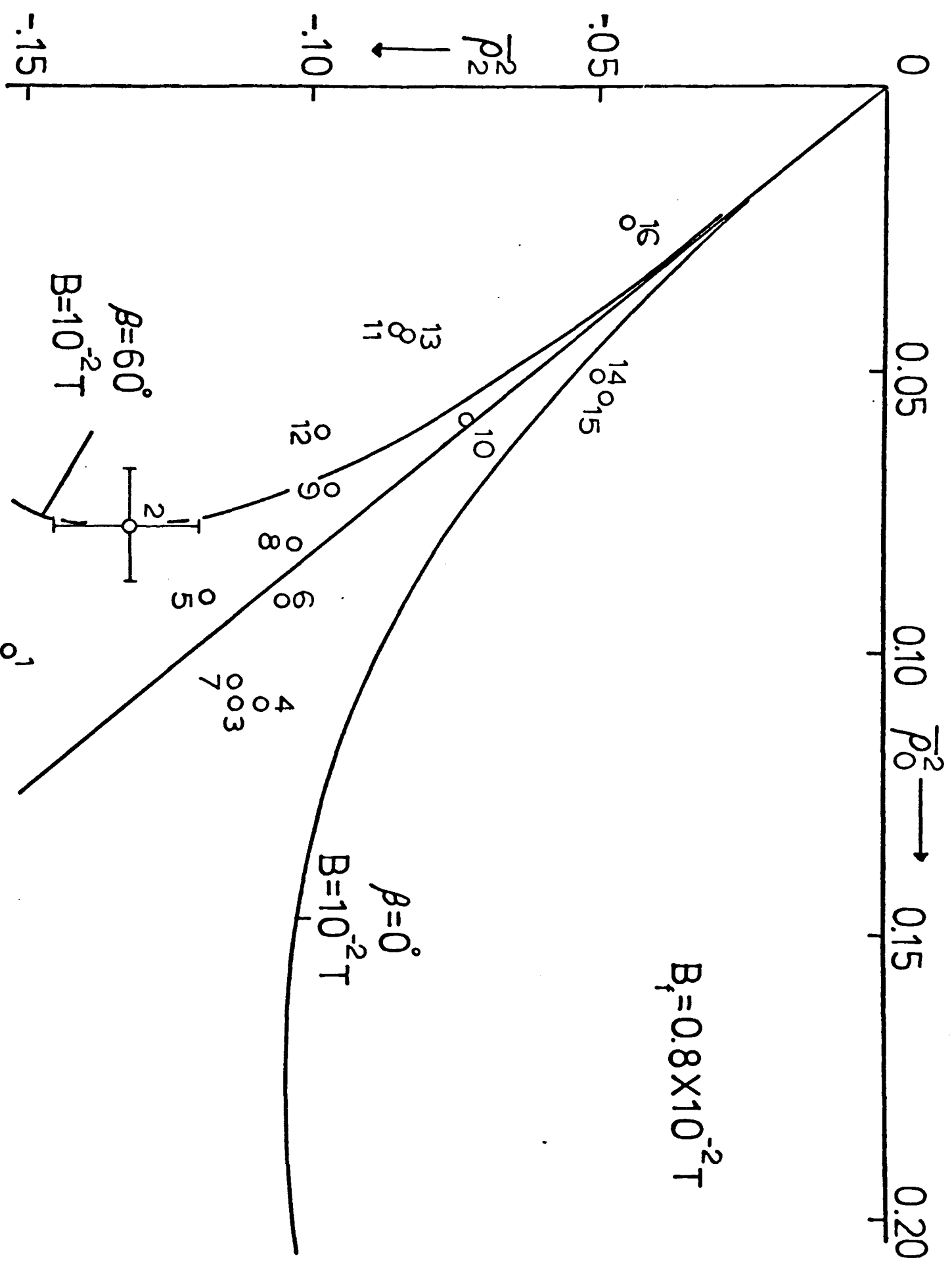


Fig. 5.7 Statistical tensor plot of $\bar{\rho}_2^2$ vs $\bar{\rho}_0^2$ for $B_f = 0.8 \times 10^{-2} T$

clearly. The 810 keV gamma-ray was analysed to get more information on the 'canted' spin structure of the nuclear system. This is discussed in section 5.9.2.

$$5.9.1 \quad \underline{B_f = 0.8 \times 10^{-2} \text{ T}}$$

The $\bar{\rho}_2^2$ vs $\bar{\rho}_0^2$ plot (Fig. 5.7) showed that the lowest temperature reached was 1.2 (3) mK, in good agreement with entropy calculations. The first data point, to the left of the 'pseudo-quadrupole' line, clearly indicates that the spins are in the 'flop' phase ($\beta > 45^\circ$). The rest of the points fall approximately on a straight line of slope

$$\frac{\bar{\rho}_2^2}{\bar{\rho}_0^2} = -\left(\frac{3}{2}\right)^{\frac{1}{2}}$$

Also plotted on the same figure is a theoretical curve for $B = 1 \times 10^{-2} \text{ T}$, $\beta = 0$ representing spins ferromagnetically aligned parallel to the applied field. Clearly also if the spins were subject only to the external applied field at these temperatures, the data points would fall close to this curve.

5.9.2 Measurement of $\bar{\rho}_4^4$, $\bar{\rho}_2^4$ and $\bar{\rho}_0^4$ (Statistical Tensors Rank 4)

As explained in section 3.8, measurements of $\bar{\rho}_0^2$ and $\bar{\rho}_2^2$ are intrinsically incapable of distinguishing between spins 'canted' at 45° to the applied field in the a-a' plane and a pure quadrupole interaction along the c-axis. Anisotropy data from the 810 keV gamma-ray was therefore analysed to resolve this ambiguity.

The 810 keV gamma-ray anisotropy at 1 mK has a substantial

rank 2 component ($A_2(\gamma) = 0.075(8)$) which had to be subtracted from the measured anisotropies to obtain the rank 4 component ($A_4(\gamma) = 0.43$). Moreover, the temperature reached on demagnetisation was not low enough to show much effect from rank 4 component. For these reasons, the data were of a lower quality as compared with the 712 keV gamma-ray. Nevertheless, important conclusions could be drawn as we shall see below.

As no zero field measurements were made in Series 2, the lowest field Run available ($B_f = 0.8 \times 10^{-2}$ T) was chosen for analysis. The anisotropy from the 712 keV gamma-ray was used to correct for the rank 2 component. It is clear from Equation 3.17 that ρ_q^2 components scale with $A_2(\gamma)$. Thus the contribution from the second rank tensor component for the 810 keV gamma-ray could be estimated from the measured values for the 712 keV gamma-ray, given that the respective $A_2(\gamma)$ values are known. This was the procedure used.

The fourth rank tensor component was further analysed to give estimates for the orientation tensors ρ_q^4 . The measured anisotropy from statistical tensors of rank 4 is given by the expression (Equation 3.24).

$$\begin{aligned}
 W_a' (0) - 1 &= Q_4 A_4(\gamma) (2I_i + 1)^{\frac{1}{2}} \rho_0^4 \\
 W_a' (\pi/2, \pi/2) - 1 &= Q_4 A_4(\gamma) (2I_i + 1)^{\frac{1}{2}} [0.375 \rho_0^4 + 0.7906 \rho_2^4 \\
 &\quad + 1.046 \rho_4^4] \quad 5.4 \\
 W_c' (\pi/2, 0) - 1 &= Q_4 A_4(\gamma) (2I_i + 1)^{\frac{1}{2}} [0.375 \rho_0^4 - 0.7906 \rho_2^4 + \\
 &\quad 1.046 \rho_4^4]
 \end{aligned}$$

Equation 5.4 shows that the gamma-ray anisotropy measured along the crystal a-axis gives a unique value for ρ_0^4 . However,

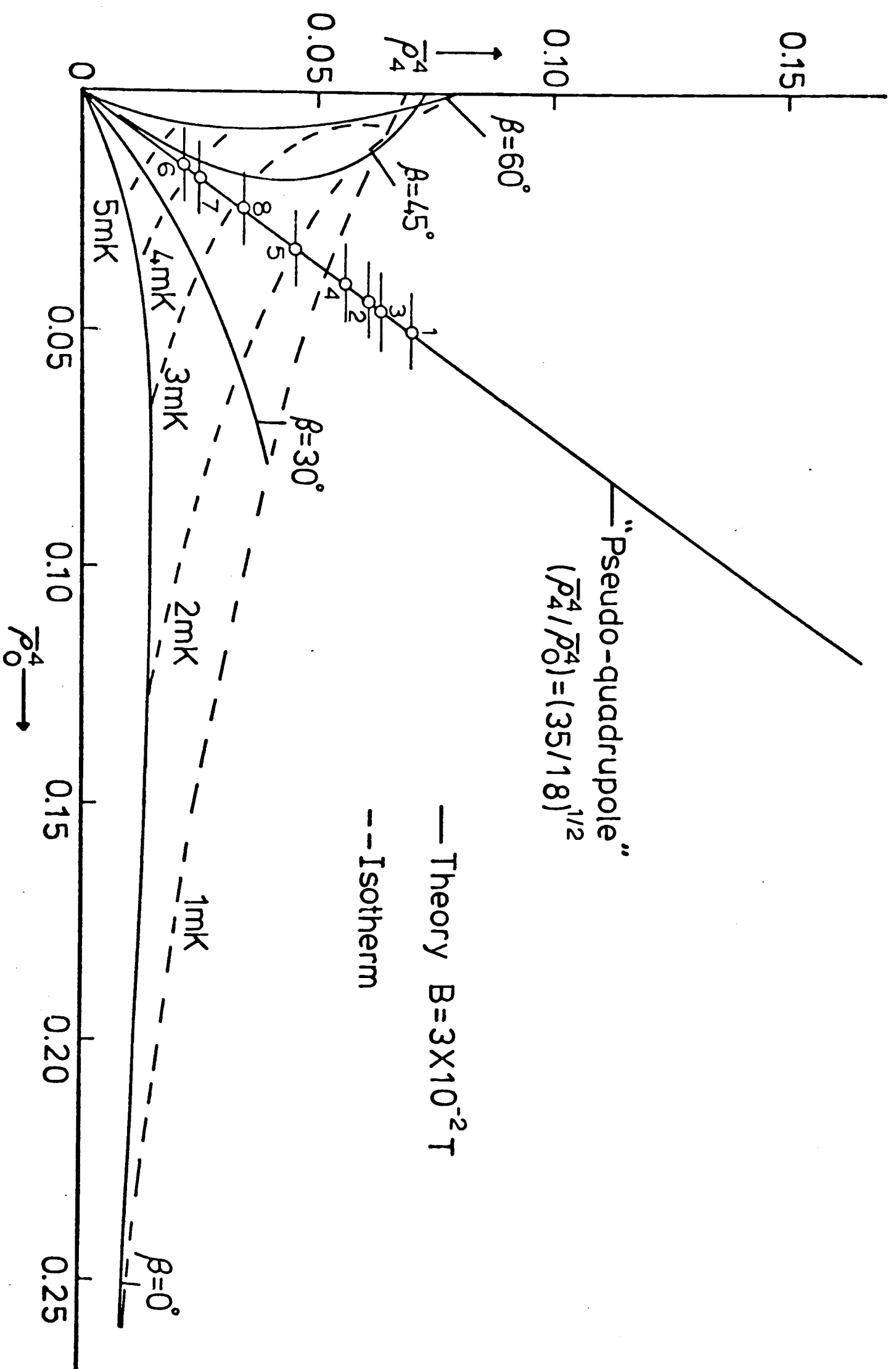


Fig. 5.8 Statistical tensor plot of $\bar{\rho}_4^4$ vs $\bar{\rho}_0^4$ for $B_f = 0.8 \times 10^{-2} T$

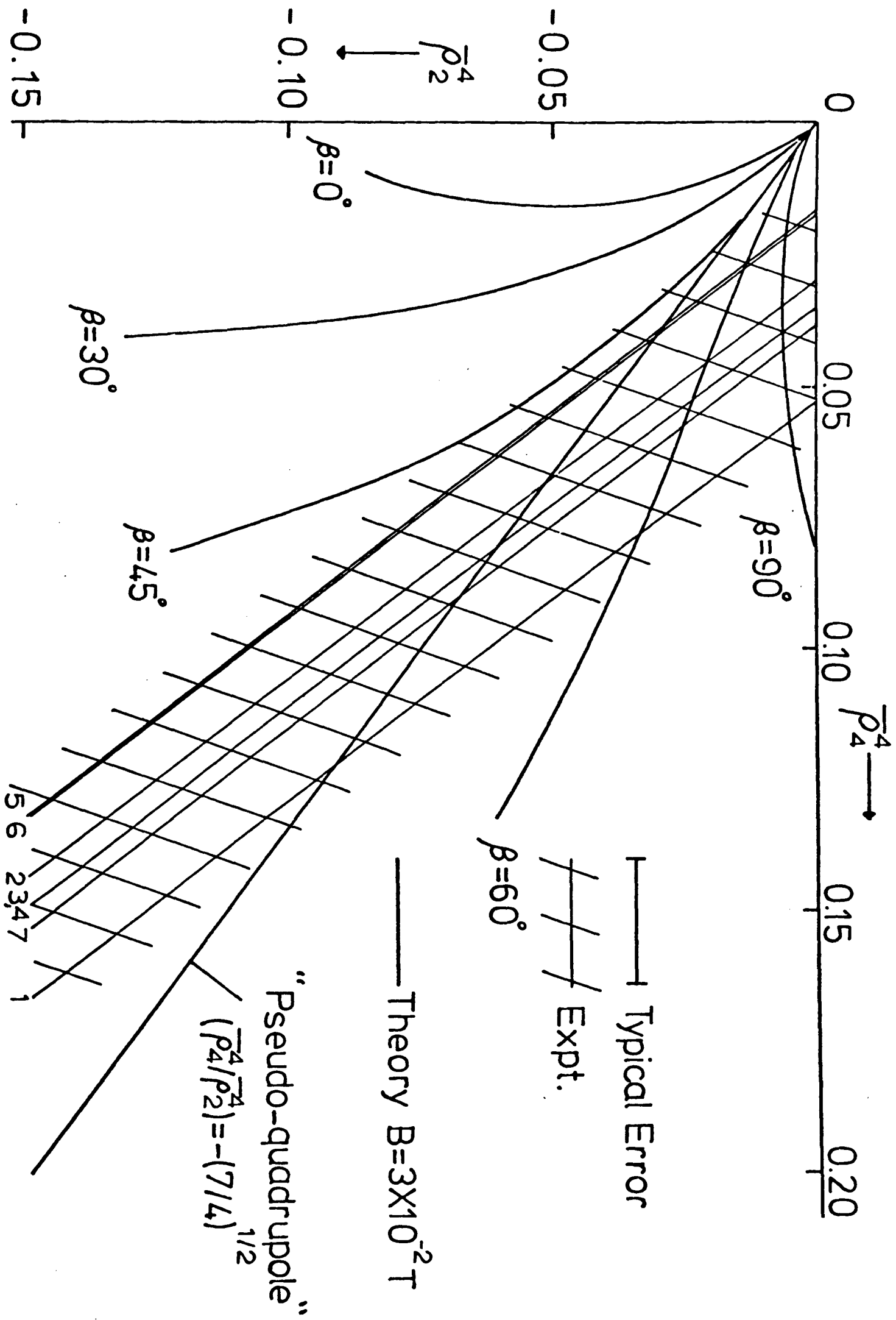


Fig. 5.9 Statistical tensor plot of $\bar{\rho}_2^4$ vs $\bar{\rho}_4^4$ for $B_f = 0.8 \times 10^{-2} \text{ T}$

as only two detectors were used, unique values for ρ_2^4 and ρ_4^4 could not be obtained from our measurements. The measured $\bar{\rho}_0^4$ values are plotted in Fig. 5.8. It should be noted that the $\bar{\rho}_4^4$ component for each data point is totally arbitrary but is made to lie on the straight line representing a pure quadrupole interaction along the crystal c-axis. The reason for this is indicated below. Also plotted are theoretical curves for canting angles $\beta = 30^\circ$, 45° and 60° for an applied field $B = 3 \times 10^{-2}$ T. Calculations show that the field acting on the nuclei is $\sim 3 \times 10^{-2}$ T below the Néel temperature (section 2.8.3). Thus the curves for $B = 3 \times 10^{-2}$ T represent the gamma-ray emission pattern for different canting angles β , for the antiferromagnet $^{166m}\text{HoVO}_4$. It should, however, be stressed that although these curves are sensitive to the ratio of the quadrupole interaction to the Zeeman term, the general conclusions which follow do not depend on its exact ratio.

The curve for $\beta = 45^\circ$ has a maximum $\bar{\rho}_0^4$ of ~ 0.02 , which is much smaller than the measured maximum value of ~ 0.05 . In fact, below a spin temperature of about 1.5 mK, $\bar{\rho}_0^4$ becomes negative. Clearly, our measurements show that the spins are not 'canted' at 45° to the applied field but are uniformly distributed in the a-a' plane. Such a distribution would give data points which fall on a straight line of slope

$$\bar{\rho}_4^4 / \bar{\rho}_0^4 = \frac{1}{3} \left(\frac{35}{2}\right)^{\frac{1}{2}}$$

Assuming zero magnetic field at the nuclei, the lowest temperature reached is estimated to be 1.4 (2) mK, in good agreement with temperatures deduced from the 712 keV gamma-ray data. For spins uniformly distributed in the a-a' plane with a

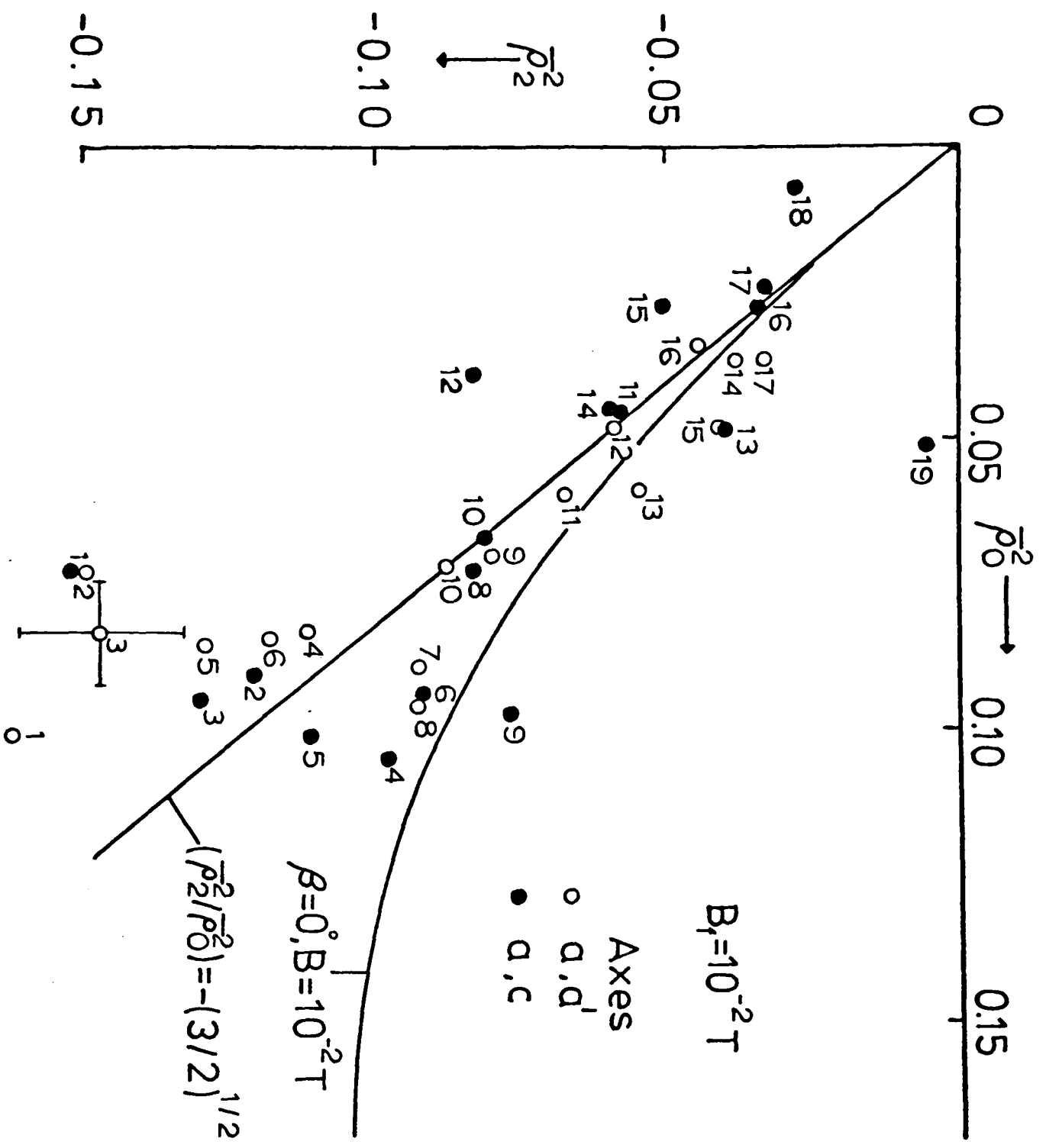


Fig. 5.10 Statistical tensor plot of ρ_2^{-2} vs ρ_0^{-2} for $B_f = 10^{-2} \text{ T}$

magnetic field at the ion of 3×10^{-2} T, the final temperature is estimated to be 1.7 (3) mK, which is somewhat higher than the entropy prediction.

Data from the equatorial detector (a'-axis) were also analysed by subtracting the second rank tensor components by scaling equatorial data for the 712 keV gamma-ray. Subtraction of $\bar{\rho}_0^4$ deduces from the 810 keV gamma-ray axial data then gave a series of loci in the $\bar{\rho}_2^4 - \bar{\rho}_4^4$ plane as shown in Fig.

5.9. The straight lines so obtained intersect the line of slope

$$\bar{\rho}_4^4 / \bar{\rho}_2^4 = -\sqrt{7/4}$$

and the lowest temperature reached is estimated to be 0.5 ($^{+5}_{-2}$) mK. Once again the loci for the lowest temperatures do not intersect the curve for $\beta = 45^\circ$, thus showing that a 'canting' angle of 45° is unlikely.

5.9.3 $B_F = 1.0 \times 10^{-2}$ T

Two Runs were made with the gamma-ray detectors along a and a', and along a and c in the second. The derived average statistical tensors $\bar{\rho}_0^2$ and $\bar{\rho}_2^2$ plotted in Fig. 5.10, are found to be broadly self-consistent. The first four data points clearly indicate the spin 'flop' phase ($\beta > 45^\circ$). The (average) lowest temperature reached in the two Runs was 1.0 (2) mK, again in good agreement with entropy calculations. Fig. 5.11 is a plot of the measured anisotropy along the a and a' axes as a function of time after the end of demagnetisation. For a uniform distribution of spins in the a-a' plane, the anisotropy along the two

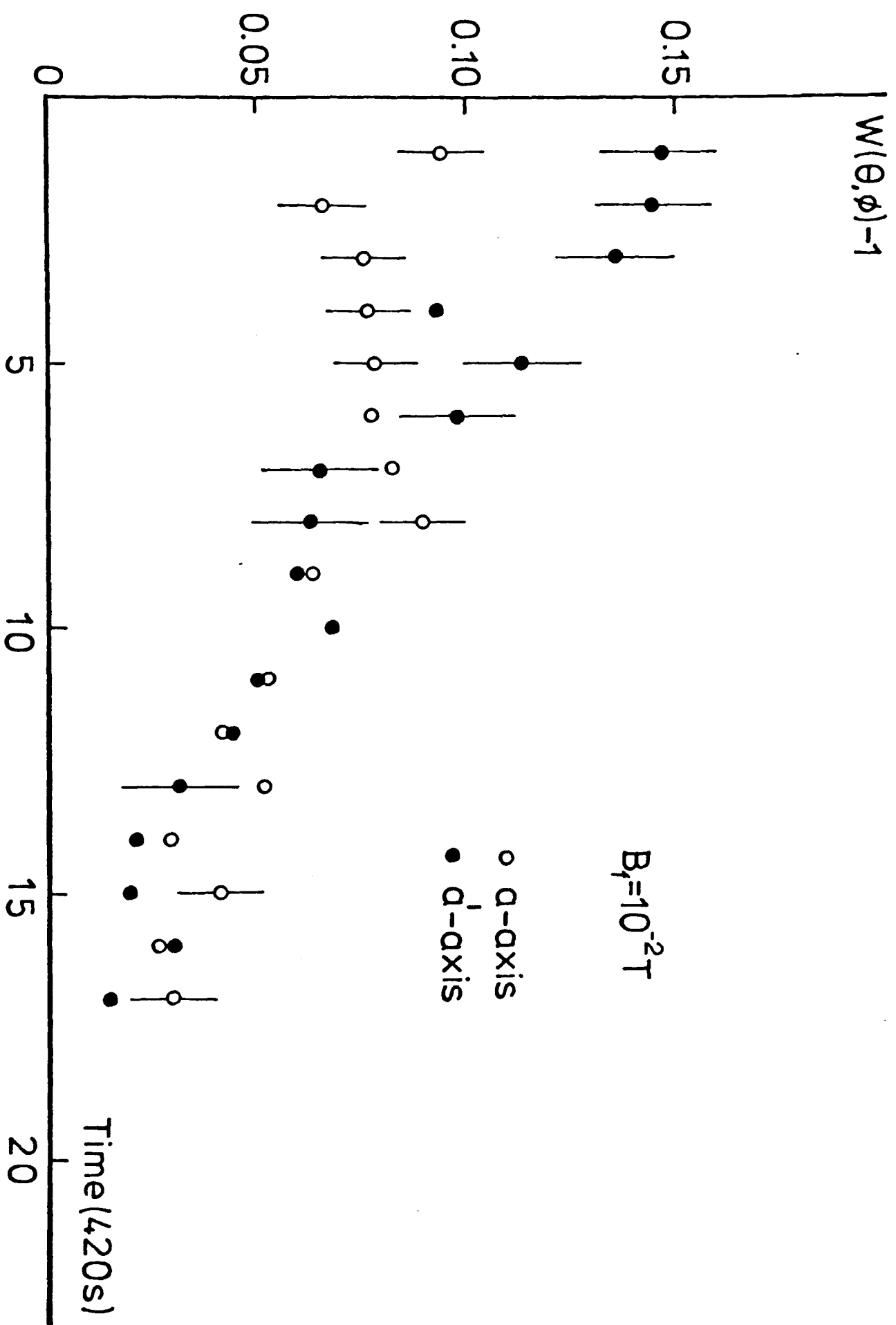


Fig. 5.11 Anisotropy of the 712 keV gamma-ray vs time after demagnetisation for $B_f = 10^{-2} \text{ T}$

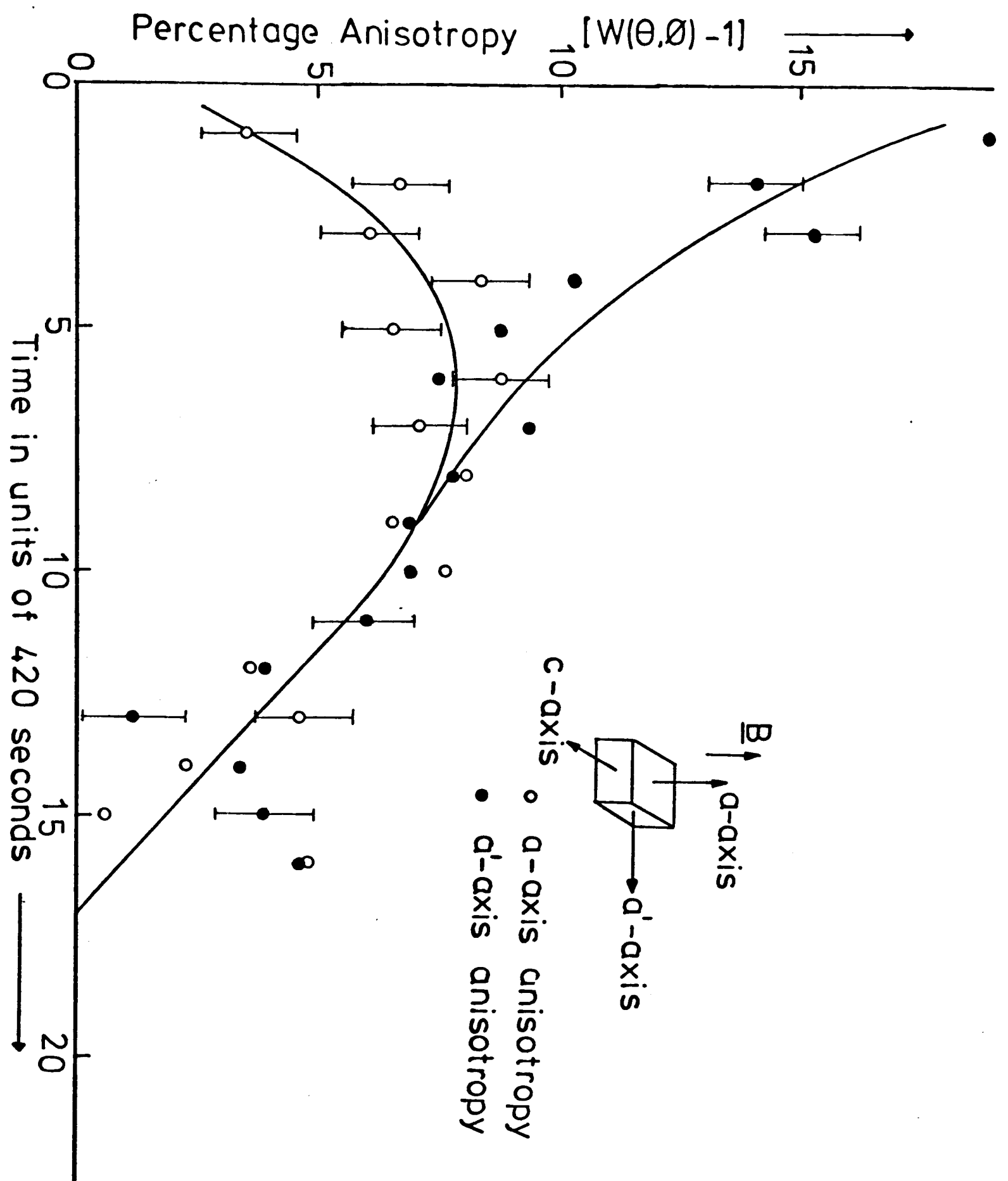


Fig. 5.12 Anisotropy of the 712 keV gamma-ray vs time after demagnetisation for $B_f = 1.2 \times 10^{-2}$ T

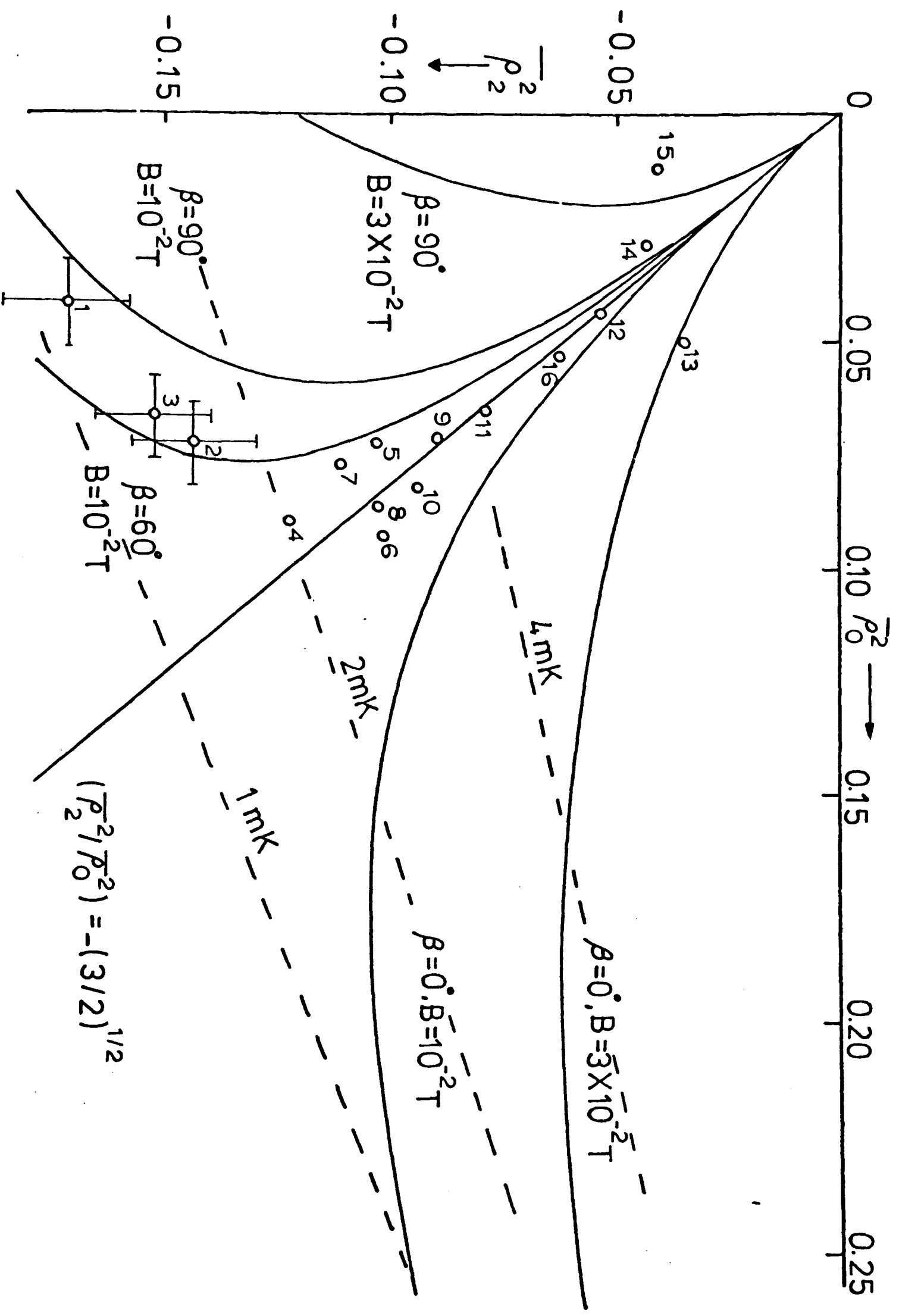


Fig. 5.13 Statistical tensor plot of $\bar{\rho}_2^2$ vs $\bar{\rho}_0^2$ for $B_f = 1.2 \times 10^{-2} T$

directions would clearly be identical. The difference in count rate may be interpreted to show that the spins are preferentially oriented parallel to the a' axis ('flop' phase). This increases the count rate along the a' axis at the expense of the count rate along the a axis. After about six counts of 420 seconds, the count rates along the two directions became identical owing to the warming up of the spins.

$$5.9.4 \quad \underline{B_f = 1.2 \times 10^{-2} \text{ T}}$$

This Run identified the nuclear antiferromagnetic spin 'flop' most clearly. Fig. 5.12 is a plot of the measured anisotropy along the crystal directions a and a' as a function of time after end of demagnetisation. The difference in count rates along the two directions is quite striking: at the lowest temperature the anisotropy along the a' axis is about three times that along the a axis. As the nuclei warm up, they lose polarisation and also start to rotate in to the direction of the applied field. As a result, the count rate along the a direction at first increases and then starts to decrease, thus explaining the peak in the count rate along this direction. The count rate along a' , however, is decreasing all the time. After about six counts of 420 seconds, the count rate along the two directions have become roughly equal, indicating that the spins have become isotropic in the a - a' plane. It should be noted that if the spins were to close up into the paramagnetic phase, the count rate along these two directions would not be equal.

Fig. 5.13 is a plot of the average statistical tensors $\bar{\rho}_0^2$ and $\bar{\rho}_2^2$, calculated from the anisotropy data. From the

isotherms sketched on the diagram, we estimate the lowest temperature reached to be about 1.3 mK. Also plotted are curves for applied fields of 10^{-2} T and 3×10^{-2} T for various values of β . The best fit to the data is obtained for $\beta \approx 55^\circ$ for $B = 3.3 \times 10^{-2}$ T. The statistical tensor curves are quite sensitive to the applied field B : thus for $B = 10^{-2}$ T, $\beta \approx 75^\circ$. A calculation (section 2.7.2) gave $\beta = 69^\circ$ for an applied field $B = 1.2 \times 10^{-2}$ T, somewhat larger than our measured values. The reasons for this apparent reduction in the effective field at the nucleus are discussed in the next section.

5.10 Discussion

From NO studies on the 712 and 810 keV gamma-rays from ^{166m}Ho , we have identified an antiferromagnetic spin 'flop' phase in an applied field of 1.2×10^{-2} T and a temperature of approximately 1 mK. We also note that this value for the applied field required to induce the spin 'flop' is in good agreement with r.f. susceptibility measurements on $^{165}\text{HoVO}_4$, which gave a peak in the susceptibility in an applied field of approximately 1.5×10^{-2} T (section 2.4). The NO data also showed that in small applied fields ($< 5 \times 10^{-3}$ T) and temperatures of order 1 mK, the spins were uniformly distributed in the a - a' plane with no preferred axis of magnetisation.

At first sight, it might seem that the above results can be explained in terms of magnetic anisotropy in the a - a' plane. For HoVO_4 , the anisotropy energy, E_a may be written in the form

$$E_a = \frac{K}{4} \sum_{i=1}^4 \sin^2 (2 \theta_i) \quad 5.5$$

where K is a constant, θ_i is the angle between the direction of spin magnetisation and the 'easy' axis and the summation is over the four sublattices that make up the nuclear ordered structure. From symmetry arguments, the 'easy' axis is either along one of the 'enhanced' directions (a or a') or at 45° to it. If the former were true, the spin 'flip' phase would be preferred in small applied fields. The spins would then 'flop' as the field is increased. For the latter case, the spins would 'cant' with $\beta = 45^\circ$ in small applied fields. The NO anisotropy data is clearly incompatible with both these cases. We therefore conclude that simple ideas of magnetic anisotropy cannot explain these results.

It seems likely that as the crystal is demagnetised to below T_N (4.0 mK), it breaks up into magnetic domains, effectively giving a uniform distribution of spins in the basal plane. It is tempting to conjecture that the domains form around paramagnetic impurities (mainly rare-earth), but the following analysis shows that this is probably untrue. The main rare-earth impurities Yb and Tb (see section 4.2.5), in a crystalline field of tetragonal symmetry are expected to have a singlet and a Kramers doublet ground state respectively, with no anisotropy in the basal plane. This presumably precludes pinning of spins in any particular direction in the basal plane. What seems probable is that crystalline imperfections (defects, dislocations, etc.) cause pinning of spins in particular preferred directions when the crystal breaks up into domains. On this basis, one would expect the non-irradiated sample to have larger domains than the irradiated one. Presumably, this would not alter the field required to cause the spin 'flop' to occur. This is precisely

what is observed: both the non-irradiated and irradiated samples showed a spin 'flop' at approximately the same applied field.

It is not possible to deduce the shape or size of magnetic domains from the measurements of this chapter. However, a rough estimate of the average pinning energy per ion can be made from dipole energy considerations. In zero applied field, the average spin 'canting' angle of 45° is stabilised by a pinning mechanism. On application of a magnetic field of 1.2×10^{-2} T, the spins have a measured 'canting' angle $\beta \approx 55^\circ$ for a local field at the ion B of 3.3×10^{-2} T. If the total moment at the ion is m ($\approx 0.46 \mu_B$ at 0K), then for a canting angle β the total dipolar energy E_d in an applied field B is given by the equation

$$E_d = -0.0734 m^2 \sin^2 \beta - mB \cos \beta \quad 5.6$$

For a uniform distribution of spins in the basal plane, this gives

$$\bar{E}_d = -0.0734 m^2 \langle \sin^2 \beta \rangle = -0.0367 m^2 \quad 5.7$$

Substituting $\beta = 55^\circ$ in Equation 5.6

$$(E_d)_{\beta = 55^\circ} = -0.0493 m^2 - 0.00688 m \quad 5.8$$

Assuming that at this field the pinning energy E_p is just overcome by the spin 'flopping', we get

$$E_p \sim 0.0127 m^2 + 0.0688 m$$

Substituting $m = 0.46 \mu_B$, $E_p \sim 82$ MHz (~ 4 mK), a rather large figure. This figure should be taken only as a rough

indicator of the strength of the pinning, as it is doubtful whether the approximations are valid for applied fields comparable with the ordering field.

5.11 Zero-Point Motion

The relatively high field Runs of 4.2×10^{-2} T and 7.4×10^{-2} T showed that the effective field at the ion was only about half the applied field. Also, even in an applied field as large as 1.8×10^{-2} T, the spins were antiferromagnetically aligned with $\beta \gtrsim 45^\circ$. These results suggest that the transition from the antiferromagnetic to the paramagnetic phase is not sharp.

It has been known for some time (8) that the ground state for an antiferromagnet has an energy term which does not vanish at absolute zero. This reduces the effective saturation magnetisation. Davis (9) has computed the reduction in saturation magnetisation for several crystal structures and spin values. It turns out that the spin reduction is greatest for low spin and low dimensionality systems. Recent magnetic measurements on the 2-D antiferromagnet Rb_2MnF_4 ($S = 5/2$) indicate that the reduction

$$S - \langle S_z \rangle$$

is of order 0.2 (10,11) in good agreement with the calculations of Davis. For $^{165}\text{HoVO}_4$ ($I = 7/2$) the reduction is expected to be smaller, perhaps 5%. This is apparently too small to explain the observed reduction in local field of $\sim 50\%$ for the high field Runs of 4.2×10^{-2} T and 7.4×10^{-2} T.

5.12 Conclusions

Clearly, further experiments need to be done to map out

the different phases of the magnetic system more accurately. By applying small applied fields in different directions in the basal plane, it should be possible to investigate magnetic ordering in this plane (if any). Moreover, using gamma-ray detectors oriented in three different directions, $A_4(\gamma)$ terms from the 810 keV gamma-ray could be determined. This would give additional information on the 'canted' spin structures.

As indicated in the introduction to this chapter, in favourable cases it is possible to study nuclear antiferromagnetism by the technique of neutron diffraction. Neutron scattering by a nucleus of spin I can be interpreted in terms of scattering lengths a_+ and a_- , associated with the compound nuclei of spin $I + \frac{1}{2}$ and $I - \frac{1}{2}$ respectively. For an unpolarised nucleus, the total coherent scattering cross-section σ , from an unpolarised neutron beam is given by the expression (12)

$$\sigma = 4\pi \left(\frac{I+1}{2I+1} a_+ + \frac{I}{2I+1} a_- \right)^2 \quad 5.9$$

where $I/(2I+1)$ and $(I+1)/(2I+1)$ are weighting factors for the scattering lengths a_- and a_+ respectively. However, for a nucleus of polarisation P , there is also an extra coherent scattering cross-section $\delta\sigma$, given by the expression (13)

$$\delta\sigma = 4 \left[\frac{P (a_+ - a_-) I}{(2I+1)} \right]^2 \quad 5.10$$

where the nuclear polarisation P is defined by the expression

$$P = \left| \frac{\langle I_z \rangle}{I} \right| \quad 5.11$$

Equation 5.10 gives the total scattering cross-section

relevant to the observation of nuclear ordering in HoVO_4 . For a regular crystalline structure, the diffracted beam produce Bragg peaks whose intensities $|F|^2$ are given by the expression

$$|F|^2 = \left[\frac{P (a_+ - a_-) I}{2I+1} \right]^2 G^2 \quad 5.12$$

where G is a geometrical factor which depends on the orientation of the diffracting planes.

For ^{165}Ho , neutron diffraction measurements of Herpin et al (13) give

$$(a_+ - a_-) = -0.34 (4) \times 10^{-12} \text{ cm}.$$

The above scattering length compares favourably with the total coherent scattering length for an unpolarised ^{165}Ho nucleus

$$b_{\text{Ho}} = 0.85 (2) \times 10^{-12} \text{ cm}$$

measured by Koehler et al (14). Thus nuclear antiferromagnetism in HoVO_4 could be studied by neutron diffraction. Extra Bragg peaks should appear below 4 mK, as the nuclear spins begin to order. The average size of the magnetic domains could be estimated from the width of these Bragg peaks (4). However, there could be problems of heating of the lattice due to absorption of neutrons. This would reduce the data accumulation time.

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Chapter 5

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CHAPTER 6A MEASUREMENT OF THE NUCLEAR MAGNETIC DIPOLE MOMENT
OF $^{166}\text{m}_{\text{Ho}}$

6.1 Synopsis

This chapter describes the measurement of the nuclear magnetic dipole moment of ^{166m}Ho . The measured value of $3.60 (6) \mu_N$ is in good agreement with a value of $3.69 \mu_N$ estimated assuming a Nilsson configuration $[633 \ 7/2]_n$ $[523 \ 7/2]_p$ for the last neutron and proton.

6.2 Introduction

The calculation of the electric quadrupole splitting ^{166m}P for $^{166m}\text{HoVO}_4$ requires knowledge of the magnetic dipole moment of ^{166m}Ho (section 2.4). The published value of $4.0 (6) \mu_N$ (1) has a fairly large experimental error. The aim of the present work was to try to improve on the experimental uncertainty of the above measurement. This experiment used high resolution Ge (Li) detectors compared with poor resolution NaI (Tl) detectors of the earlier experiment. This enabled many more gamma-ray transitions from the decay of ^{166m}Ho to be studied in this work. A Nuclear Orientation study of the energy levels of ^{166}Er is presented in Chapter 7.

6.3 Experimental Details

It is necessary to know the nuclear spin temperature accurately in order to measure the magnetic dipole moment μ . In metals, good thermal contact and conduction generally ensure that the specimen and NO thermometer are at the same temperature. However, as explained in Chapter 4, the nuclear spins of ^{166m}Ho were at a temperature substantially higher

than the 'cold finger' temperature of 12 mK. This was due to the Kapitza boundary resistance at the metal-insulator interface. This boundary resistance is empirically found to strongly depend on temperature ($\propto T^{-3}$). It was hoped that by going to a relatively high temperature of between 50 and 100 mK, this could be reduced to a negligible amount without seriously reducing the size of gamma-ray anisotropy. However, NO thermometers in common use (section 1.4.3) lose sensitivity at these temperatures. This necessitated calibrating the magnetic susceptibility of a single crystal of CMN as a function of temperature in the range 12 mK to 1 K. Owing to the strong field dependence of the magnetic susceptibility of CMN at these low temperatures, care had to be taken to reduce the stray field from the polariser magnet. This is discussed below.

6.3.1 Sample Construction

A single crystal specimen of $^{166\text{m}}\text{HoVO}_4$ of activity $2\text{ }\mu\text{Ci}$ was silver painted onto the 'cold finger' thermally anchored to the dilution refrigerator, with its a-axis parallel to the applied magnetic field. A needle-shaped single crystal of h.c.p. ^{60}Co Co was soft-soldered next to the HoVO_4 crystal with its (long) c-axis parallel to the applied field.

6.3.2 Thermometry

A single crystal of CMN was polished to give a cylinder approximately 4.5 mm long and 3.3 mm diameter, with its axis perpendicular to the trigonal axis. It had an approximate demagnetisation factor of 0.18 (S.I.). The crystal specimen was pushed into a coil which formed the inductive part of an L-C tank circuit. The r.f. susceptibility was measured using

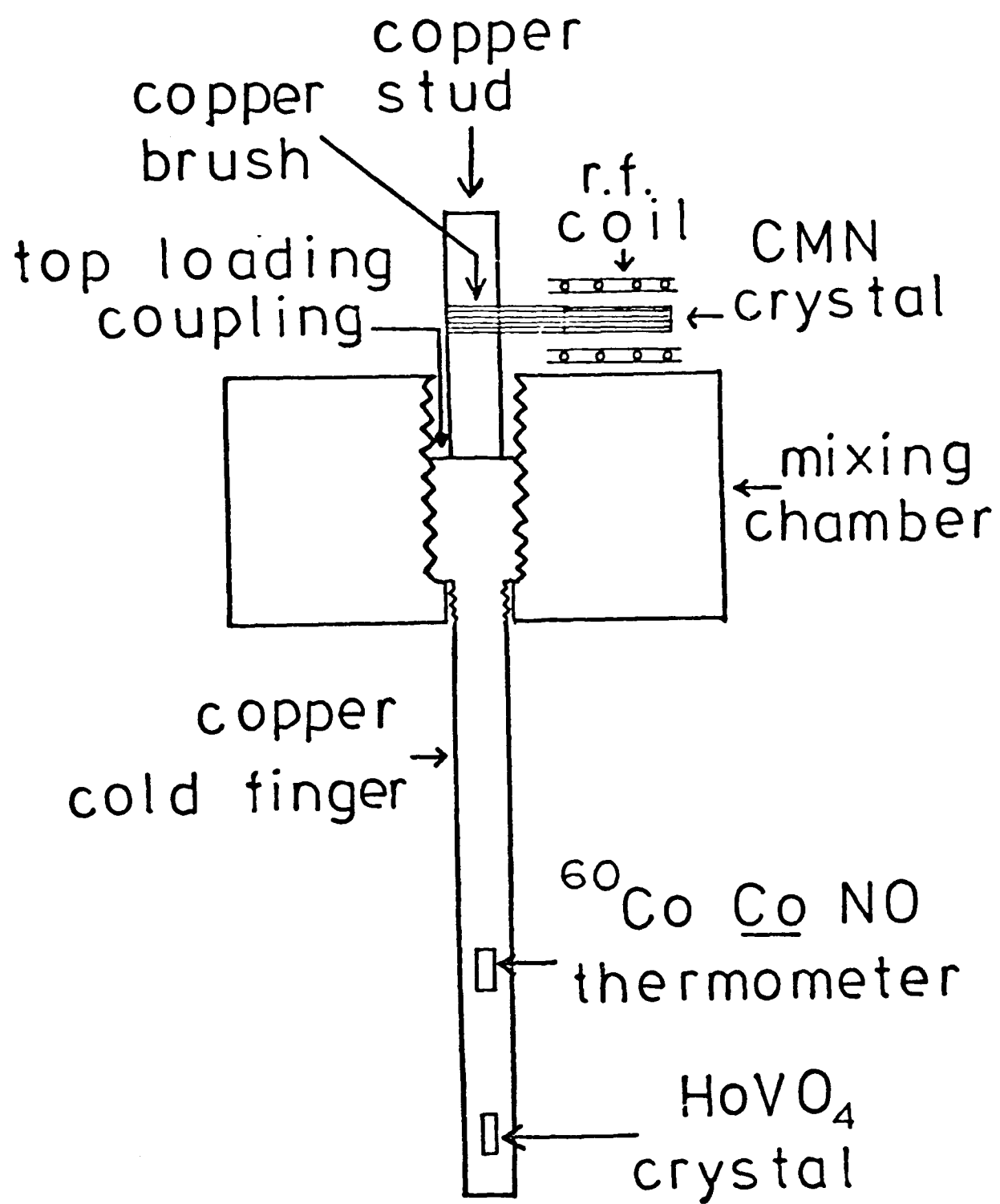


Fig. 6.1 Schematic diagram of the sample arrangement

the same oscillator circuit as discussed in section 1.4.1. A brush of 47 SWG enamelled copper wires was attached to the crystal specimen with GE varnish. Thermal contact was made to a copper bush by means of (non-superconducting) cadmium solder. The copper bush, which had a left-handed thread at one end, was screwed into an adaptor thermally anchored to the mixing chamber of the dilution refrigerator. A diagram of the relevant parts of the experimental arrangement is shown in Fig. 6.1. The superconducting solenoid (Magnet 2, section 1.2) used to polarise the $^{166\text{m}}\text{Ho}$ nuclei was field compensated so that the field at the CMN crystal located 30 cm from it was very small. To further reduce the effective stray field, the CMN crystal was fixed with its axis parallel to the horizontal plane. The r.f. susceptibility of the crystal was studied as a function of polariser field at the base temperature of the dilution refrigerator. No saturation effects were found for a field at the centre of the polariser coil of 2 T.

The r.f. susceptibility of the CMN crystal was determined as a function of temperature, measured using the NO ^{60}Co Co thermometer. The specimen temperature was changed by heating the mixing chamber. A high temperature point at about 600 mK was determined using a calibrated Speer carbon resistor. As is well known, the magnetic susceptibility of CMN is given by the Curie-Weiss expression

$$\chi = C / (T - \Delta) \quad 6.1$$

where C and Δ are constants. As explained in section 1.4.1, there is some uncertainty about the magnitude and temperature dependence of Δ . By plotting $1/\chi$ against T, Δ was determined

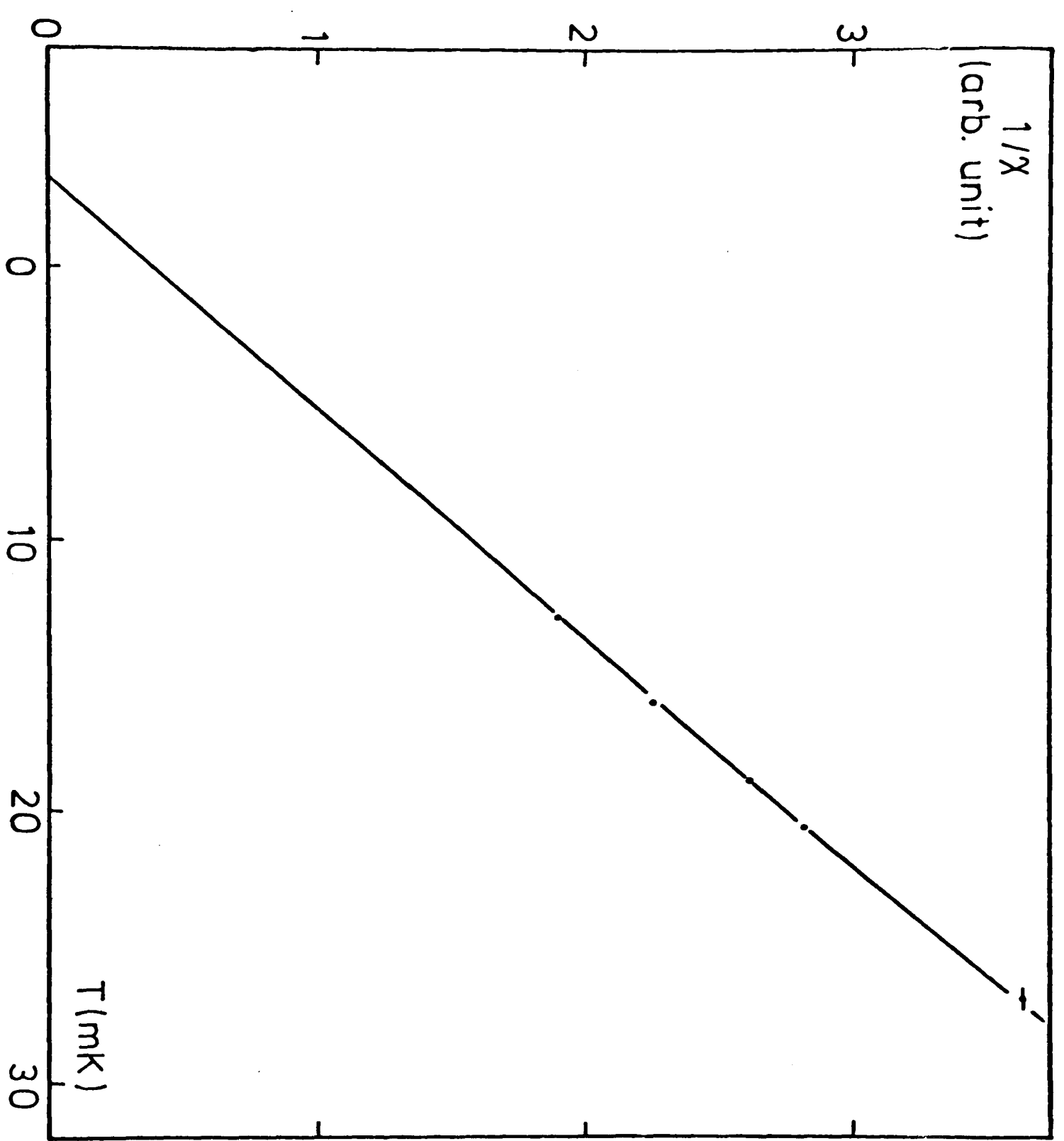


Fig. 6.2 Determination of Δ for CMN thermometer

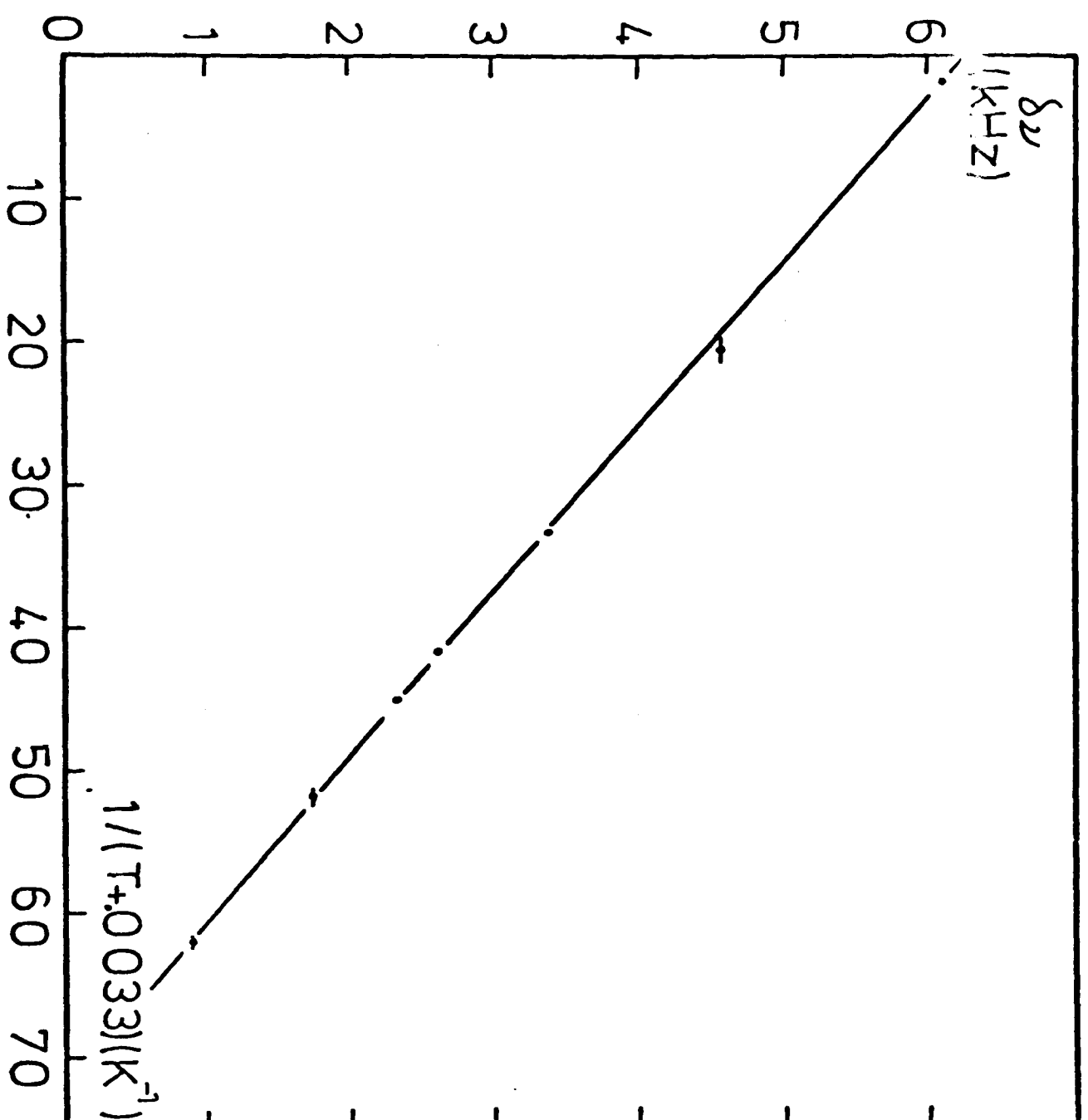


Fig. 6.3 CMN magnetic thermometer calibration

for this assembly to be $-3.3 (3)$ mK (Fig. 6.2). Values of $1/\chi$ are given in terms of frequency shifts because absolute measurements of r.f. susceptibilities could not be made using this technique. The negative value of Δ is consistent with CMN being an antiferromagnet. However, a calculation of (section 1.4.1) for a cylinder of demagnetisation factor $d = 0.18$ gave $\Delta = + 1.32 (7)$ mK, somewhat at variance with the measured value. The source of this discrepancy is unknown. The measured value of Δ was used to fit the susceptibility measurements using Equation 6.1. The resulting fit, shown in Fig. 6.3, was used to estimate all temperatures for the NO results. The accuracy of the CMN thermometer calibration is estimated to be about 1% at about 50 mK. At the lowest temperatures, thermal equilibrium times for the CMN crystal were several hours owing to poor thermal contact. The asymptotic value at each temperature was estimated from a plot of resonant frequency as a function of time.

6.3.3 Gamma-ray Counting System

As explained in section 1.6, two gamma-ray detectors were available for the experiment. However, it was found that summation of pulses from both detectors and use of the 'Router' (section 1.6.1) increased the width of the photo-peak. To maintain good peak resolution, a single (axial) detector was used in these experiments. The detector was placed with its axis along the polariser direction about 12.5 cm from the single crystal of HoVO_4 . The pulses from the detector after amplification were fed into the full 4096 channel memory of the MCA. After each acquisition period of either 1000 or 2000 seconds, the full spectrum was stored on magnetic tape using

a system devised by J. Siertsema (section 1.6.3).

6.4 The Experiment

After the CMN thermometer had been calibrated at several temperatures, the mixing chamber was heated to about 50 mK and the temperature monitored using the calibrated CMN thermometer. 1000 and 2000 second spectra were recorded on magnetic tape for several applied fields. For each field point, the first three 1000 second counts were discarded, as the nuclear spin system was still equilibrating with the 'cold finger'. The crystal was then heated to about 60 mK and several more measurements were made. The data were analysed using the NOVA 1200 computer and the details of the procedure used are described fully in section 1.7. 'Warm' or normalisation counts were taken at the beginning of the experiment only, owing to liquid helium in the main bath running out before the end of the experiment. The two sets of data for the 712 keV gamma-ray were analysed separately, but as they gave similar values for the magnetic dipole moment μ , it was decided to treat them as a single set. Table 6.1 gives the data for the 712 keV gamma-ray. As explained in section 2.3, the 'enhancement factor' is field dependent. This required the applied field $B_{app.}$ to be corrected using Table 2.1 and the particulars are summarised in Table 6.1. The effective field at the nucleus, $B_{hyp.}$, is given by the relation

$$B_{hyp.} = (K+1) B_{corr.}$$

where K is the 'enhancement factor' in zero applied field. NMR of ^{165}Ho at 4.2 K by M.R. Wells gave, for this particular crystal

Table 6.1
Temperature and magnetic field data for NO of

<u>$^{166}\text{m}_{\text{Ho}}$</u>					
$B_{\text{app.}}$	$B_{\text{corr.}}$	T	$B_{\text{corr.}}/T$	$B_{\text{hyp.}}/T$	711.7 keV Anisotropy
(T)	(T)	(mK)	(TK^{-1})	(TmK^{-1})	$W(o)-1$ (%)
0	0	48.7(5)	0	0	1.2(4)
0.25	0.25	48.7(5)	5.13	0.89	6.3(3)
0.50	0.497	48.3(5)	10.3	1.79	16.4(4)
1.00	0.978	52.1(5)	18.8	3.26	28.0(2)
1.50	1.427	51.0(5)	28.0	4.86	35.5(3)
2.00	1.836	49.8(5)	36.9	6.40	39.4(4)
0.25	0.25	65.2(4)	3.83	0.67	3.2(2)
0.50	0.497	65.0(4)	7.65	1.33	10.2(4)
0.75	0.740	64.7(4)	11.4	1.99	17.9(3)
1.00	0.978	64.2(4)	15.2	2.64	24.6(4)
1.25	1.206	59.8(4)	20.2	3.50	29.5(4)
1.50	1.427	63.8(4)	22.4	3.88	31.3(3)
1.75	1.626	61.4(4)	26.7	4.64	34.2(4)
2.00	1.836	62.8(4)	29.2	5.08	35.6(3)

$$(^{165} \gamma_1 / 2\pi) = 1557 \text{ MHz tesla}^{-1}$$

Using Equation 2.9b, $K_{B=0}$ was calculated to be 172.3. In all the figures for gamma-ray anisotropy, $B_{\text{hyp.}}/T$ is plotted against anisotropy.

6.5 Measurement of the Magnetic Dipole Moment of $^{166\text{m}}\text{Ho}$

A number of gamma-rays of known multipolarity from the decay of $^{166\text{m}}\text{Ho}$ were analysed to yield the magnetic dipole moment μ . The parameters U_2 , F_2 , U_4 and F_4 were calculated assuming the published decay scheme (2) using a computer programme developed by D.W. Murray (3). $^{166\text{m}}\text{Ho}$ ($I=7^-$) decays to 6^- levels of ^{166}Er by β^- emission. Consequently, the β -transition is pure $J_\beta = 1$ and the U_2 , U_4 coefficients for the β transition are known quite accurately. Incomplete knowledge of mixing ratios of several of the gamma transitions produced uncertainties in U_2 and U_4 coefficients and were taken into account in the data analysis. The solid angle corrections Q_2 and Q_4 were calculated from tabulations for a Ge (Li) detector of the same geometry, 12.5 cm from the source (4). The anisotropy data for each gamma-ray were analysed using a weighted least squares method with the nuclear Zeeman splitting $\gamma_L^{B=0}$ and percentage in good sites z as free parameters. The nuclear Hamiltonian

$$\mathcal{H} = - \gamma_L \hbar B_z I_z - \frac{1}{2}P [I_z^2 - \frac{1}{3}I(I+1)] \quad 6.2$$

was used throughout, and the off diagonal terms (see Equation 3.1) I_+^2 and I_-^2 were ignored as they are small compared to the diagonal term in the quadrupole interaction

$$-\frac{1}{2}P [I_z^2 - \frac{1}{3}I(I+1)]$$

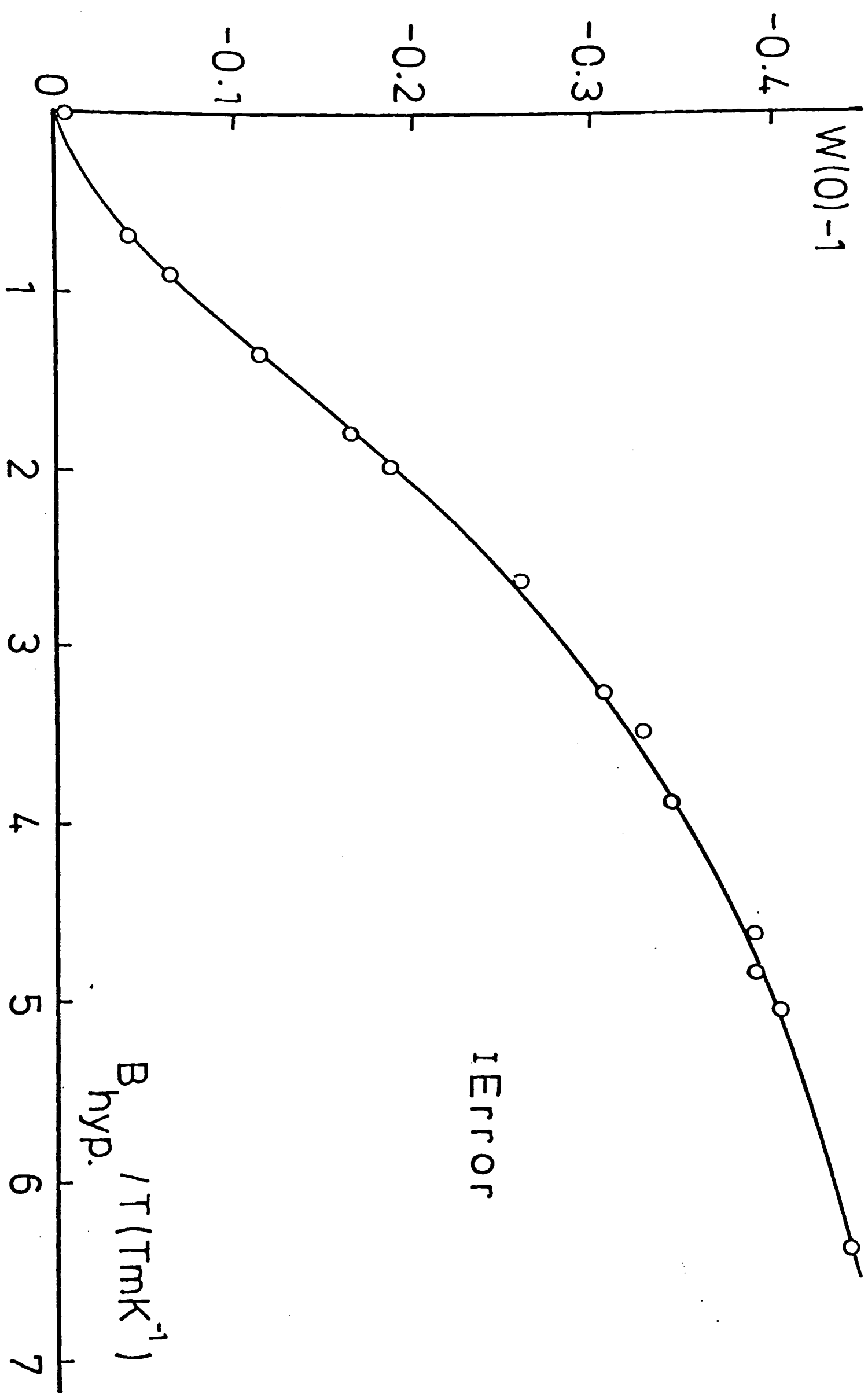


Fig. 6.4 Anisotropy of the 184 keV gamma-transition

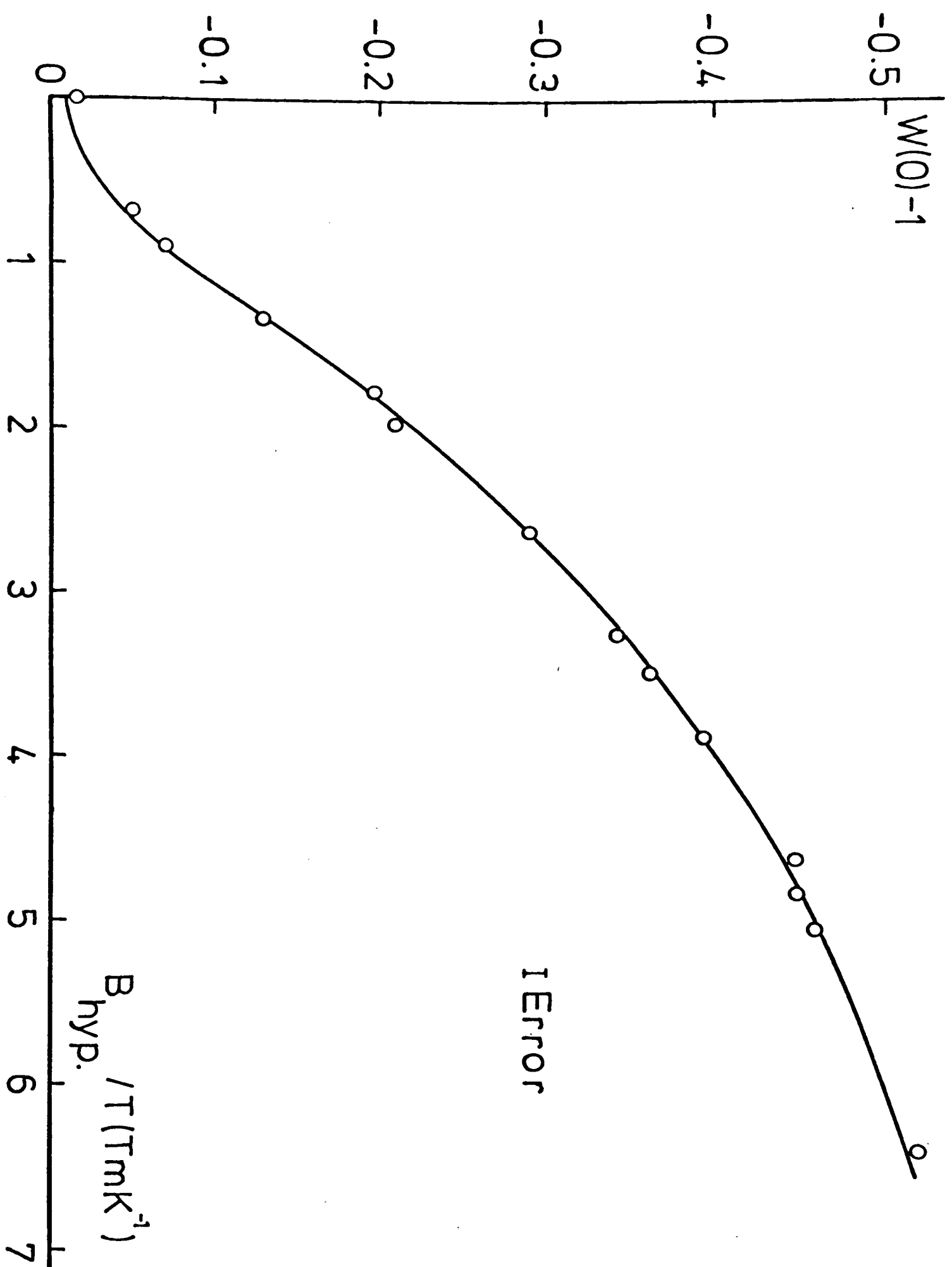


Fig. 6.5 Anisotropy of the 280 keV gamma transition

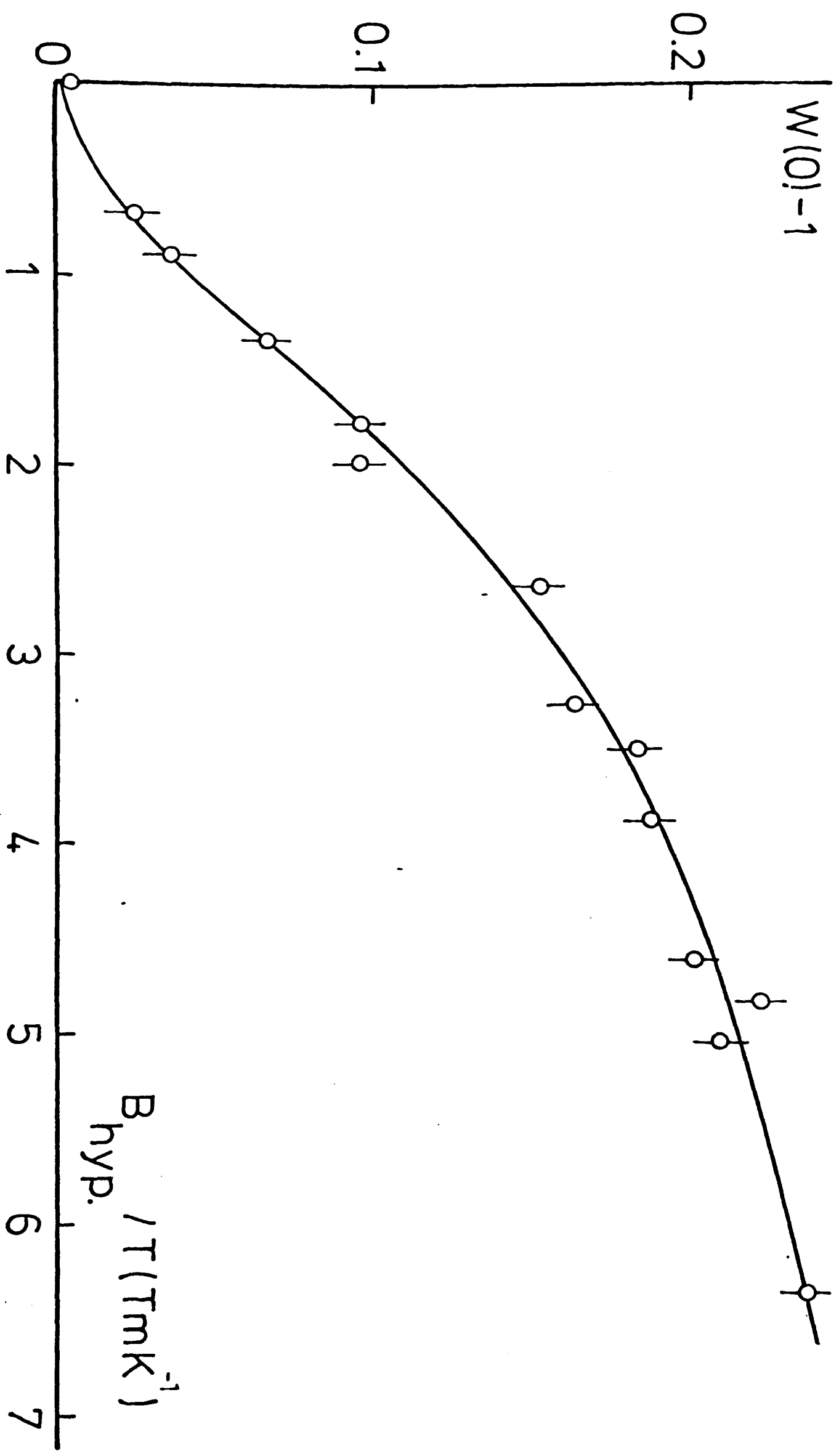


Fig. 6.6 Anisotropy of the 411 keV gamma-transition

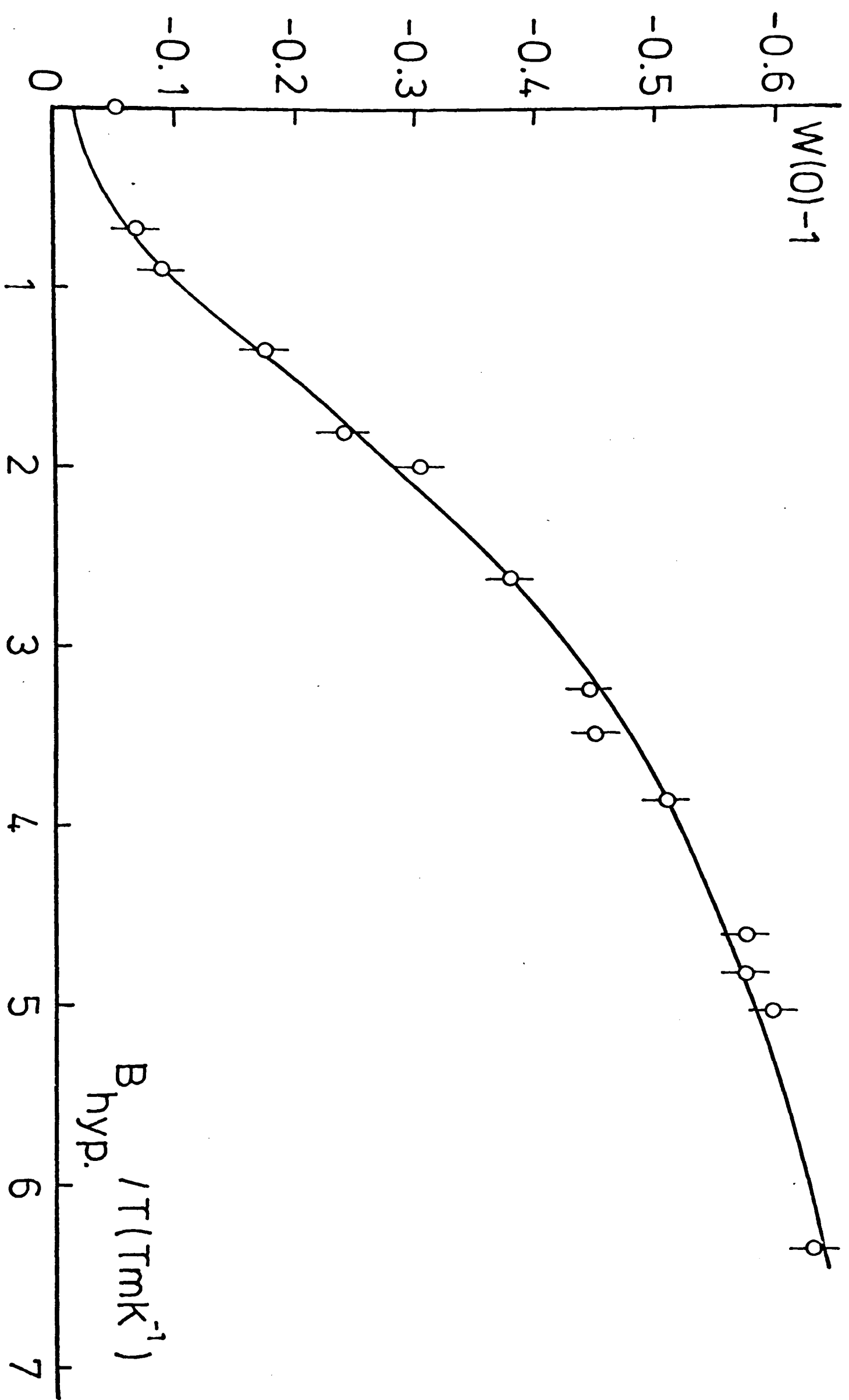


Fig. 6.7 Anisotropy of the 571 keV gamma transition

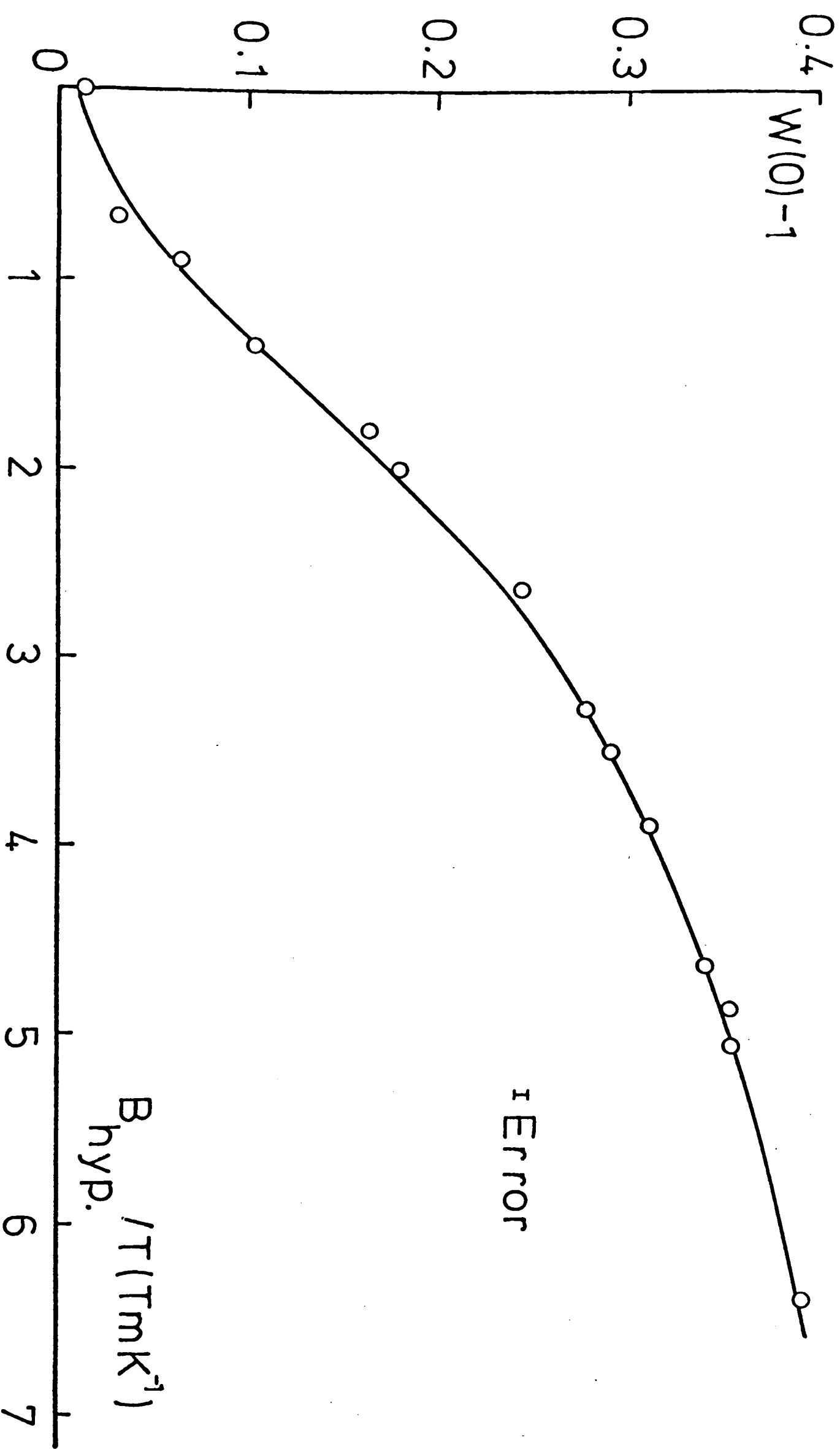


Fig. 6.8 Anisotropy of the 712 keV gamma-transition

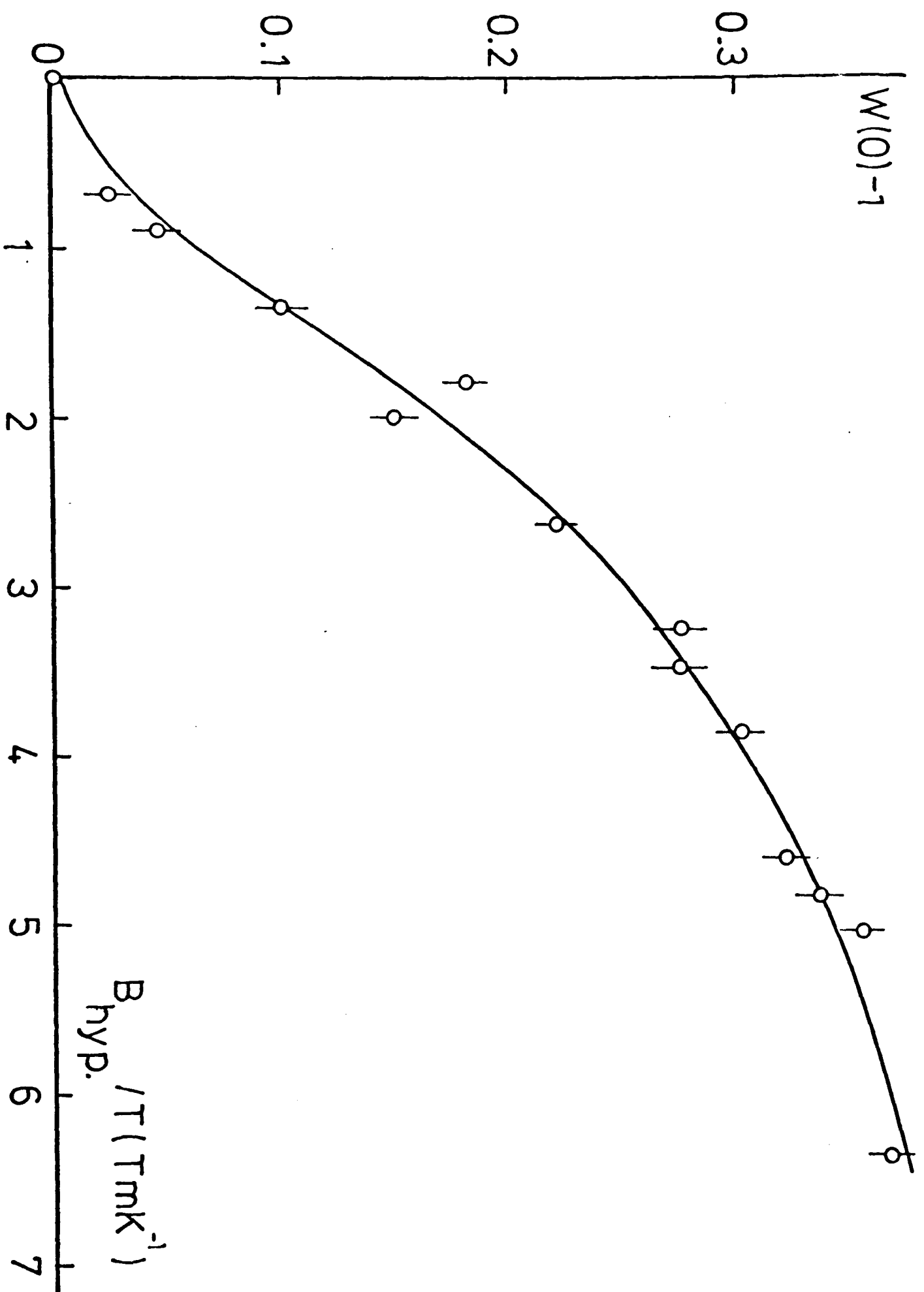


Fig. 6.9 Anisotropy of the 752 keV gamma-transition

for large magnetic fields ($\frac{\mu_B}{I} \gg P$). The electric quadrupole interaction P was taken to be +3.4 MHz (see section 2.4). The results are summarised in Table 6.2 with the quoted experimental error corresponding to an increase in reduced χ^2 of 1. (The reduced χ^2 was computed by dividing the total χ^2 by the number of degrees of freedom). The data are presented in Figs. 6.4-6.9, with the smooth curves representing the 'best fit'. The results for the nuclear Zeeman splitting for ^{166m}Ho are expressed as ratios

$$^{166m}\chi_{\perp} / ^{165}\chi_{\perp}$$

as the exact value for the splitting is a function of sample shape. The weighted average values for this ratio and the percentage in good sites z are:

$$\begin{aligned} ^{166m}\chi_{\perp} / ^{165}\chi_{\perp} &= 0.436 \text{ (6)} \\ z &= 95.4 \text{ (17)} \end{aligned}$$

Taking μ and I for ^{165}Ho as $4.125 \text{ (44)} \mu_N \text{ (5)}$ and $7/2$ respectively, we obtain for ^{166m}Ho :

$$\mu = 3.60 \text{ (6)} \mu_N$$

This is in excellent agreement with the value of $3.65 \text{ (13)} \mu_N$ obtained by Marshak (6) from NO measurements on ^{166m}Ho in Ho metal. The fact that z is less than 100% may indicate imperfect annealing as has been discussed in section 5.4.

6.6 The Nuclear Energy Level Scheme for ^{166}Ho

Thermal neutron capture, conversion electron spectroscopy and nuclear reactions have established a total of sixteen low-lying rotational bands of ^{166}Ho (7, 8), with $I = 6^+$ the

Table 6.2

Experimental values of $^{166}\chi_1/^{165}\chi_1$ and z for some transitions of known multipolarity

gamma-ray (keV)	Intensity (%)	$I_i \rightarrow I_f$	Multipolarity	($^{166m}\chi_l/^{165}\chi_l$)	z (%)	Values taken U_{2F2}	U_{4F4}
184.4	73.2	$4_g^+ \rightarrow 2_g^+$	pure F2	0.432(11)	91.9(30)	-.3006(37)	-.0507(40)
280.5	29.8	$6_g^+ \rightarrow 4_g^+$	pure F2	0.441(12)	95.9(46)	-.3137(65)	-.0809(79)
410.9	11.6	$6^- \rightarrow 7_\gamma^+$	pure E1	0.444(32)	91.2(40)	0.1718	
571.0	5.8	$6^- \rightarrow 6_\gamma^+$	pure E1	0.427(24)	100.0(25)	-.4295	
711.7	58.7	$6^- \rightarrow 5_\gamma^+$	pure E1	0.439(14)	95.5(15)	0.2733	
752.3	13.1	$6^- \rightarrow 5_\gamma^+$	pure E1	0.432(32)	92.1(40)	0.2733	

highest spin detected. If it is assumed that the odd neutron and proton in ^{166}Ho can be described separately by the Nilsson wave functions $\mathcal{N}_n$ and $\mathcal{N}_p$ respectively, then K is given by the equation

$$K = \left| \mathcal{N}_n \pm \mathcal{N}_p \right|$$

Here K is a quantum number which describes the internal motion of the nucleus. By taking $\mathcal{N}_n$ and $\mathcal{N}_p$ as observed in ^{165}Dy ($\mathcal{N}_n = \frac{7}{2}^+ [633\uparrow]$) and ^{165}Ho ($\mathcal{N}_p = \frac{7}{2}^- [523\uparrow]$) respectively, the lowest lying levels are predicted to be

$$\begin{aligned} K &= 0^- \quad [633\uparrow - 523\uparrow] \\ \text{and } K &= 7^- \quad [633\uparrow + 523\uparrow] \end{aligned}$$

This assignment is expected for the 99th neutron $[633\uparrow]$ and the 67th proton $[523\uparrow]$ in their lowest Nilsson orbitals. Investigation of the decay of the 80 hour (half-life) ^{166}Dy has established the $K = 0^-$ rotational band (9-15). But the $K = 7^-$ rotational band has not been conclusively established experimentally, although there is strong internal evidence that the ground state of ^{166m}Ho is 7^- as outlined above (7, 8). Our experiments are consistent with this.

6.7 The Nuclear Magnetic Dipole Moment of ^{166m}Ho

In the collective rotational model of the nucleus, the nuclear magnetic dipole moment μ , for the ground state, can be written in the form

$$\mu = (g_R + K_p g_p + K_n g_n) I / (I+1) \quad 6.2a$$

Equation 6.2a for the moment is composed of three parts.

Firstly, there is a contribution g_R from the collective motion

of the core nucleons. If the assumption is made that the core particles are just as likely to be up as down

$$g_R \approx Z/A \quad 6.2b$$

where Z is the nuclear charge and A is the total number of nucleons in the core. The second and third contributions, $K_p g_p$ and $K_n g_n$, are from the extra-core proton and neutron respectively. The rotational bands in ^{166}Ho are fairly well understood (section 6.6) with the three lowest bands probably associated with the configuration

$$\mathcal{J}_n \mathcal{J}_p = [633 \ 7/2]_n [523 \ 7/2]_p$$

for the extra-core neutron and proton respectively. The ground state of ^{166m}Ho is then associated with the spins of the odd neutron and proton coupled parallel to give the rotational band levels

$$K = K_n + K_p = 7^-$$

To calculate the magnetic dipole moment using Equation 6.2a, it is necessary to estimate the particle g factors g_p and g_n . This can be done from neighbouring isotopes with the same proton and neutron configuration. Table 6.3 (16) gives these g factors, where $(g_K)_{\text{calc.}}$ refers to a calculation made using the particle-rotor model.

Table 6.3

Single particle magnetic g factors for ^{165}Ho and ^{167}Er

Nucleus	Orbit	$(g_K)_{\text{abs.}}$	$(g_K)_{\text{calc.}}$
^{165}Ho	$(523 \ \frac{7}{2})_p$	1.35	1.53
^{167}Er	$(633 \ \frac{7}{2})_n$	-0.26	-0.39

Using the values of g_K from Table 6.3, the nuclear magnetic dipole moment of ^{166m}Ho is calculated to be

$$\mu_a = 3.69 \mu_N$$

$$\mu_b = 3.84 \mu_N$$

where μ_a and μ_b refer to the values computed using $(g_K)_{\text{obs.}}$ and $(g_K)_{\text{calc.}}$ respectively. The value of g_K was computed using Equation 6.2b. These calculated values are in good agreement with the measured value of $3.60 (6) \mu_N$.

6.8 The Electric Quadrupole Moment of ^{166m}Ho

Both the 4f and lattice contributions to the electric quadrupole interaction scale with the spectroscopic quadrupole moment (section 2.4), which has not been determined experimentally. The following estimate of Q was used in the evaluation of the 4f and lattice contributions to ^{166m}P of section 2.4.

In the rotational model of the nucleus, the spectroscopic static electric quadrupole moment Q is defined in terms of the intrinsic moment Q_0 by the equation.

$$Q = \frac{3K^2 - I(I+1)}{(I+1)(2I+3)} Q_0 \quad 6.3$$

for a particular band K of spin I . The quantum number K represents the angular momentum of the intrinsic motion of the nucleus and has a fixed value for a particular band based on a given intrinsic state. The quantum number I is the total angular momentum of the nucleus. For the ground state, I is generally equal to K . This gives

$$Q = \frac{I(2I-1)}{(I+1)(2I+3)} Q_0 \quad 6.4$$

The intrinsic moment Q_0 is a function of the deformation of the nucleus and is given by the expression

$$Q_0 = \frac{4}{3} \left\langle \sum_{K=1}^Z r_K^2 \right\rangle \delta \quad 6.5$$

where the sum is over all the protons of the nucleus and δ is the distortion parameter. For a spheroidal nucleus, with uniform charge distribution, δ is defined by the equation.

$$\delta = \frac{3}{2} \frac{(R_3)^2 - (R_1)^2}{(R_3)^2 + 2(R_1)^2} \quad 6.6$$

where R_3 and R_1 are the nuclear radii parallel to and perpendicular to the nuclear symmetry axis respectively. Clearly

δ is a measure of departure from sphericity of the nucleus.

From Equation 6.5, it can be inferred that addition of neutrons does not drastically alter the intrinsic quadrupole moment Q_0 .

Typically the distortion parameter δ is between 0.25 and 0.35 for nuclei in the region $150 < A < 180$ (17) and Q_0 is also

positive (prolate shape). From measured values of Q_0 for $A = 166$ (18) Q_0 is estimated to be +7.9 (4) barns for ^{166m}Ho .

This gives for ^{166m}Ho ($K = I = 7$)

$$Q = +5.3 (3) \text{ b.}$$

6.9 Nuclear Magnetic Resonance on Oriented $^{166m}\text{HoVO}_4$

A number of NMR/ON attempts were made on $^{166m}\text{HoVO}_4$ at dilution fridge temperatures in applied fields between 0.5 and 1 T and radio-frequencies between 280 and 400 MHz. In some experiments, the magnetic field was swept through a wide range while keeping the radio-frequency constant, either with frequency modulation (FM) of depth 0-5 MHz, or without any modulation. In others, the magnetic field was kept constant and the r.f.

swept through a range of frequencies with FM alternately on and off. Both Ge(Li) and scintillation (NaI (Tl)) counters were used with single channel analyser 'windows' set on the 184 and 712 keV gamma-rays. No genuine resonance effects were observed in any of the experiments. Bleaney has suggested (19) that cross-relaxation times between the nuclear species ^{165}Ho and $^{166\text{m}}\text{Ho}$ could be as short as 1 ms, thus making it difficult to saturate the ($^{166\text{m}}\text{Ho}$) resonance with the relatively low r.f. powers ($B_1 \sim 0.3 \mu\text{T}$) available in these experiments.

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Chapter 6

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CHAPTER 7A NUCLEAR ORIENTATION STUDY OF THE ENERGY BANDS OF ^{166}Er

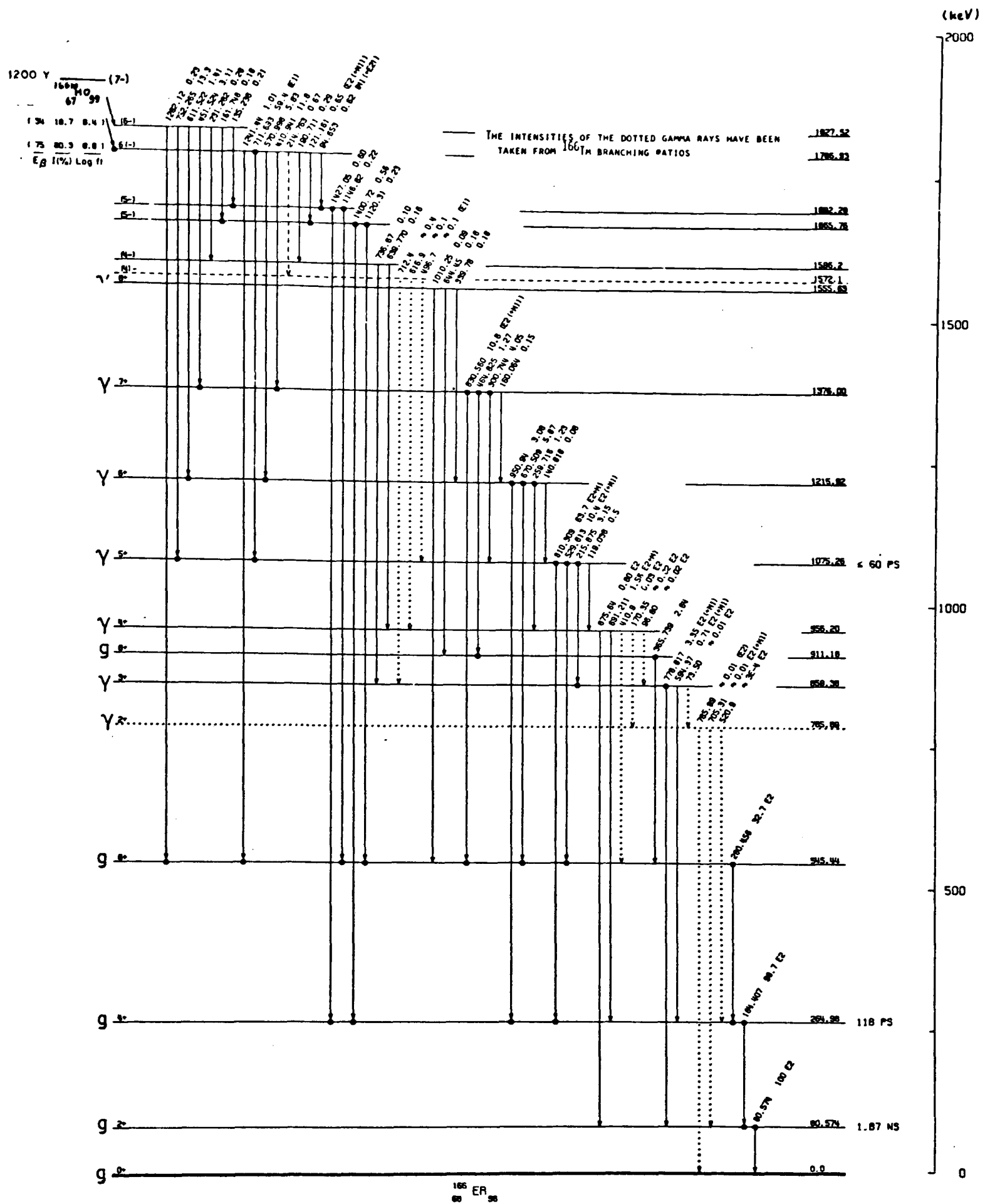


Fig. 7.1 The decay scheme of ^{166m}Ho

7.1 Introduction

This chapter is a continuation of the NO experiment of Chapter 6 and discusses measurements of multipolarities and mixing ratios of a number of the stronger gamma-ray transitions of the nucleus ^{166}Er . In particular, a 'Mikhailov type plot' of the reduced magnetic dipole transition probabilities $B(M1)$ for the transitions between 2^+ γ -band and 0^+ ground state rotational band indicated that mixing between the bands is predominantly $\Delta K = 1$.

7.2 The Nuclear Energy Levels of ^{166}Er

The energy levels of ^{166}Er have been studied both from the decay of ^{166}Ho and ^{166}Tm (1) and from nuclear reaction experiments on Dy (2). The ground (g) state is a rotational band based on $K^\pi = 0^+$ with a well-developed γ -vibrational band $K^\pi = 2^+$ closely above it. These bands along with the complete decay scheme of $^{166\text{m}}\text{Ho}$ are shown in Fig. 7.1. There is also an octupole vibrational band but the levels above 3^- have not been positively identified by experiment (3, 4). From the $\Delta K = 2$ selection rule, the interband transitions between the ground state and γ -vibrational bands are expected to be almost pure electric quadrupole (E2). This is confirmed both by gamma-ray intensity and angular distribution measurements of the interband transitions (1, 2, 5, 6, 7). Reich and Cline showed from their intensity measurements (1) of the interband transitions that there was some mixing between the ground state and γ -vibrational bands. This they attributed to $\Delta K = 2$ band mixing and estimated

$$\xi_{\gamma} = 9.4 \times 10^{-4}$$

for the band mixing parameter ξ_{γ} . $\Delta K = 2$ band mixing involves direct coupling between $K = 0$ ground state and $K = 2$ γ -vibrational bands. Another type of band mixing ($\Delta K = 1$) occurs via interaction with a third $K = 1$ band through first order Coriolis effects. Both types of band mixing produce magnetic dipole (M1) admixtures for gamma transitions between the two bands. Our measurements of δ for the interband transitions indicated that M1 transition probabilities are predominantly via $\Delta K = 1$ band mixing, consistent with available experimental data for deformed nuclei in this mass range (8).

7.3 Transitions Between the γ -Vibrational and Ground State Rotational Bands

As explained in the previous section, interband transitions between the ($K = 2^+$) γ -vibrational and the ($K = 0^+$) ground state rotational bands in ^{166}Er are predominantly E2 with small admixtures of M1. Measurement of the mixing ratio δ for the interband transitions gives information regarding the nature of band mixing. Some of these transitions were strong enough for evaluation of $U_2 F_2$ ($= A_2$) and $U_4 F_4$ ($= A_4$).

As discussed in section 3.4.2, both F_2 and F_4 are sensitive to the sign and magnitude of δ . This enables NO measurements in general to give good precision for δ . For each of the above gamma-transitions, the gamma-ray anisotropy data were fitted with δ as the only free parameter. Equations 3.5 and 3.8 were incorporated into a weighted least-squares programme to work out U_{λ} and F_{λ} as a function of the variable δ . The average measured values for μ and z (section

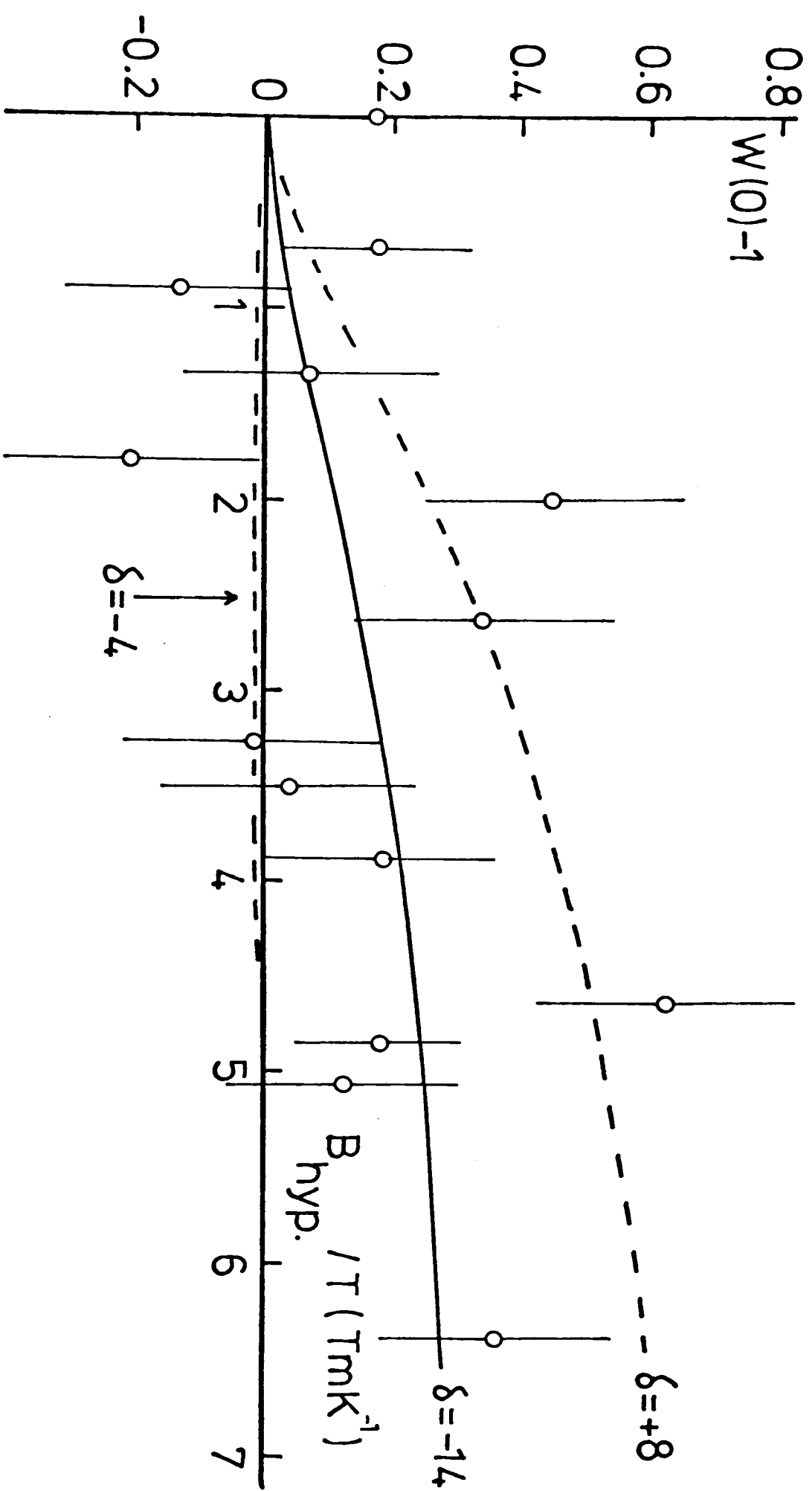


Fig. 7.2 Anisotropy of the 594 keV gamma-transition

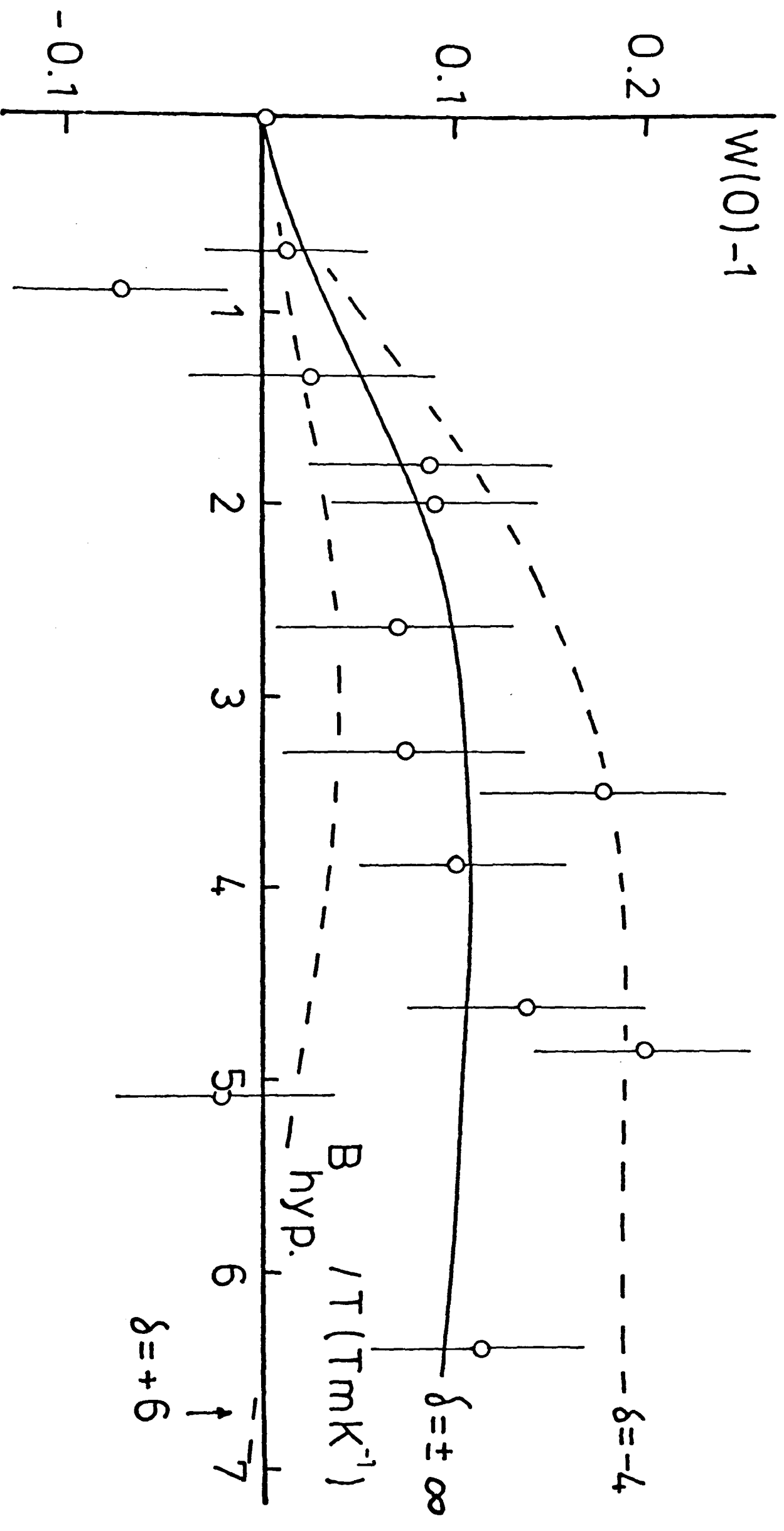


Fig. 7.3 Anisotropy of the 691 keV gamma-transition

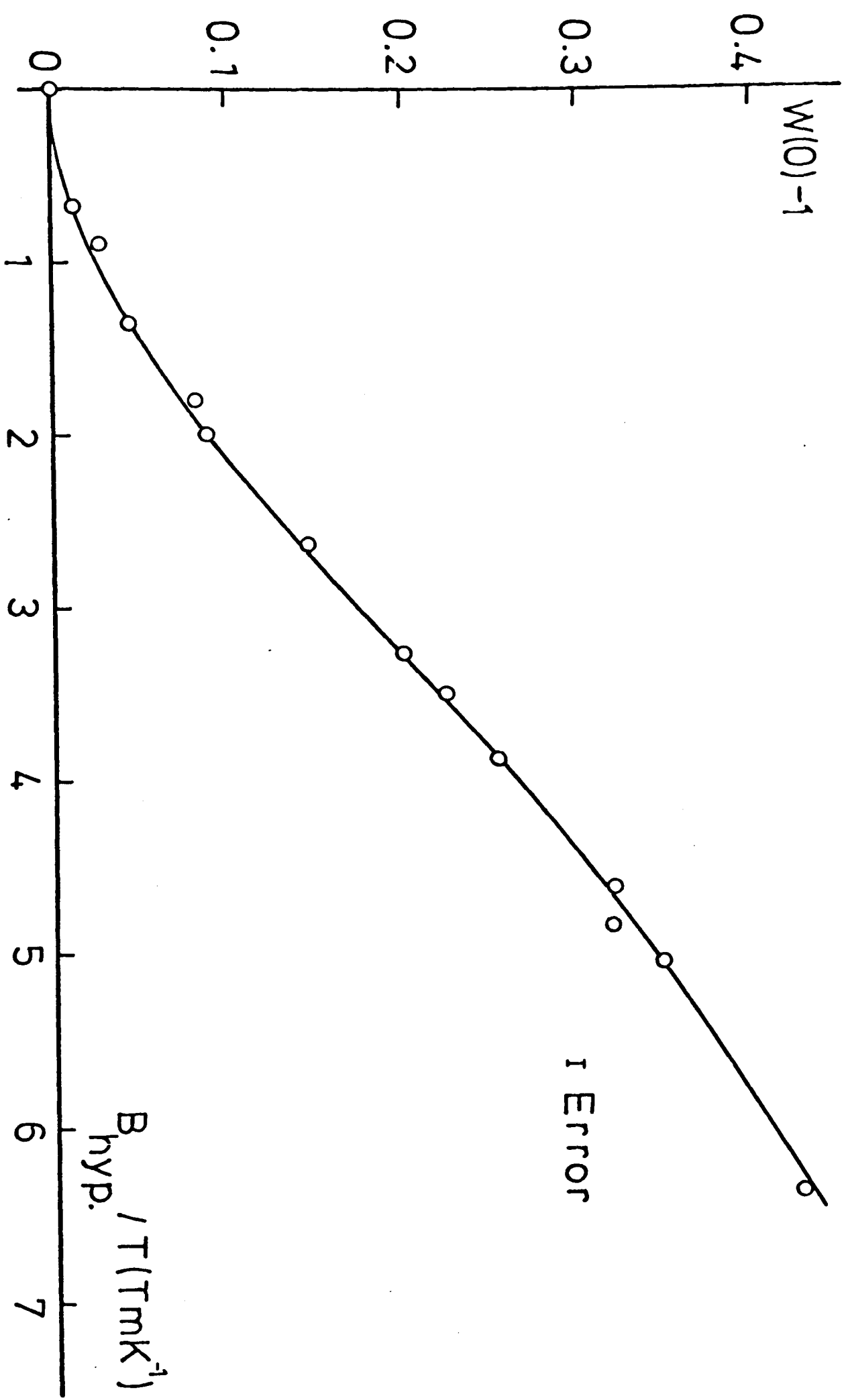


Fig. 7.4 Anisotropy of the 810 keV gamma-transition

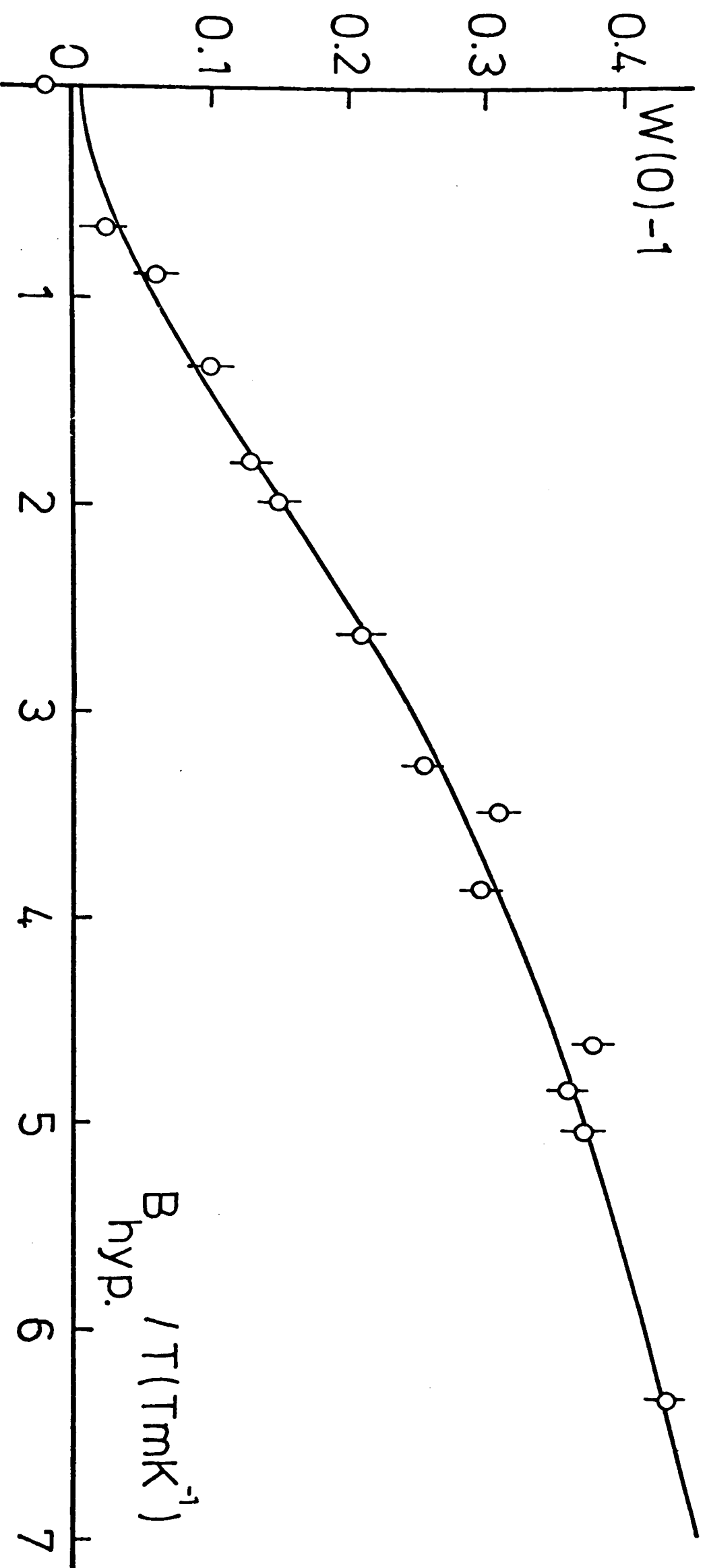


Fig. 7.5 Anisotropy of the 530 keV gamma-transition

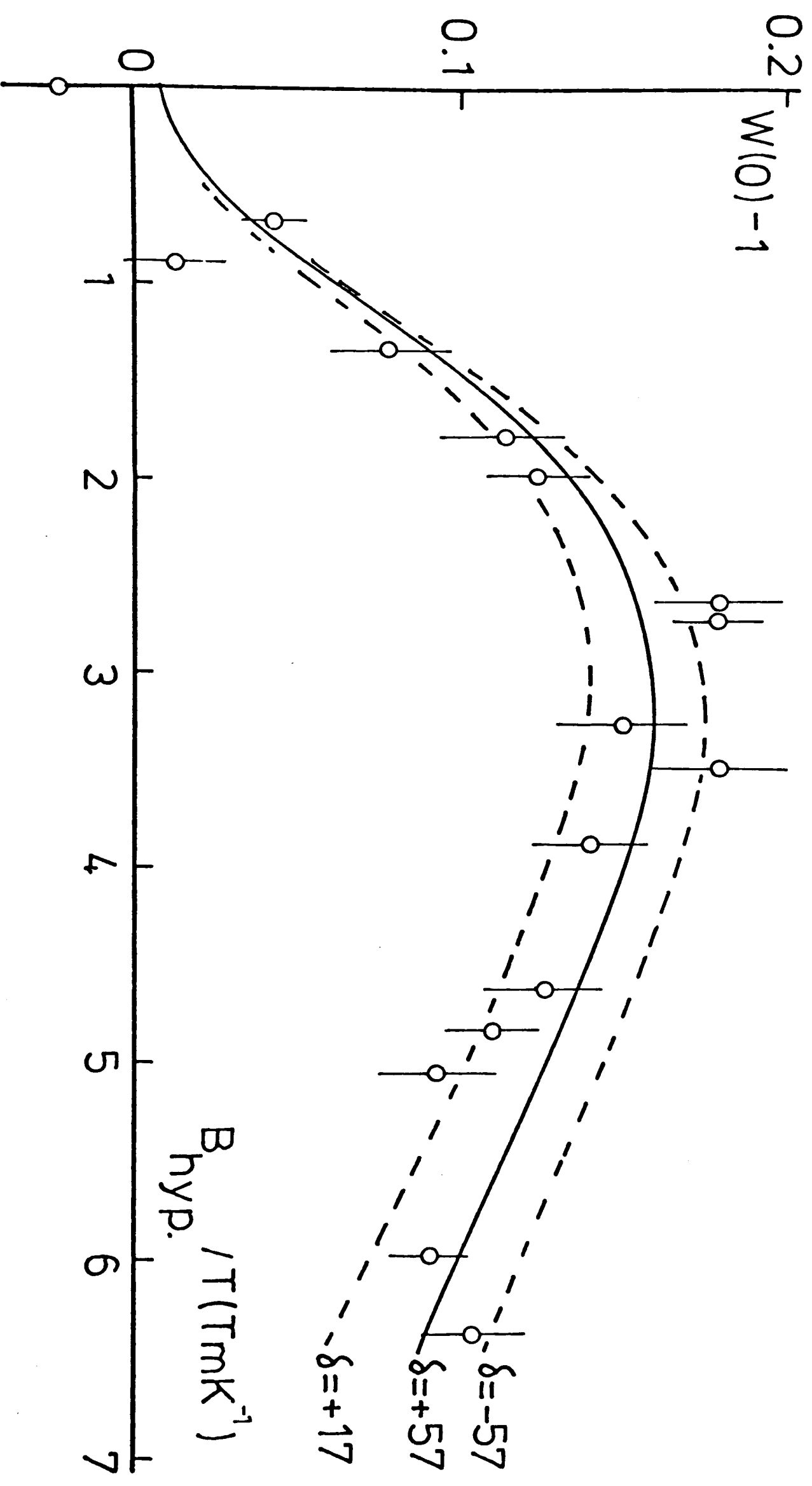


Fig. 7.6 Anisotropy of the 671 keV gamma-transition

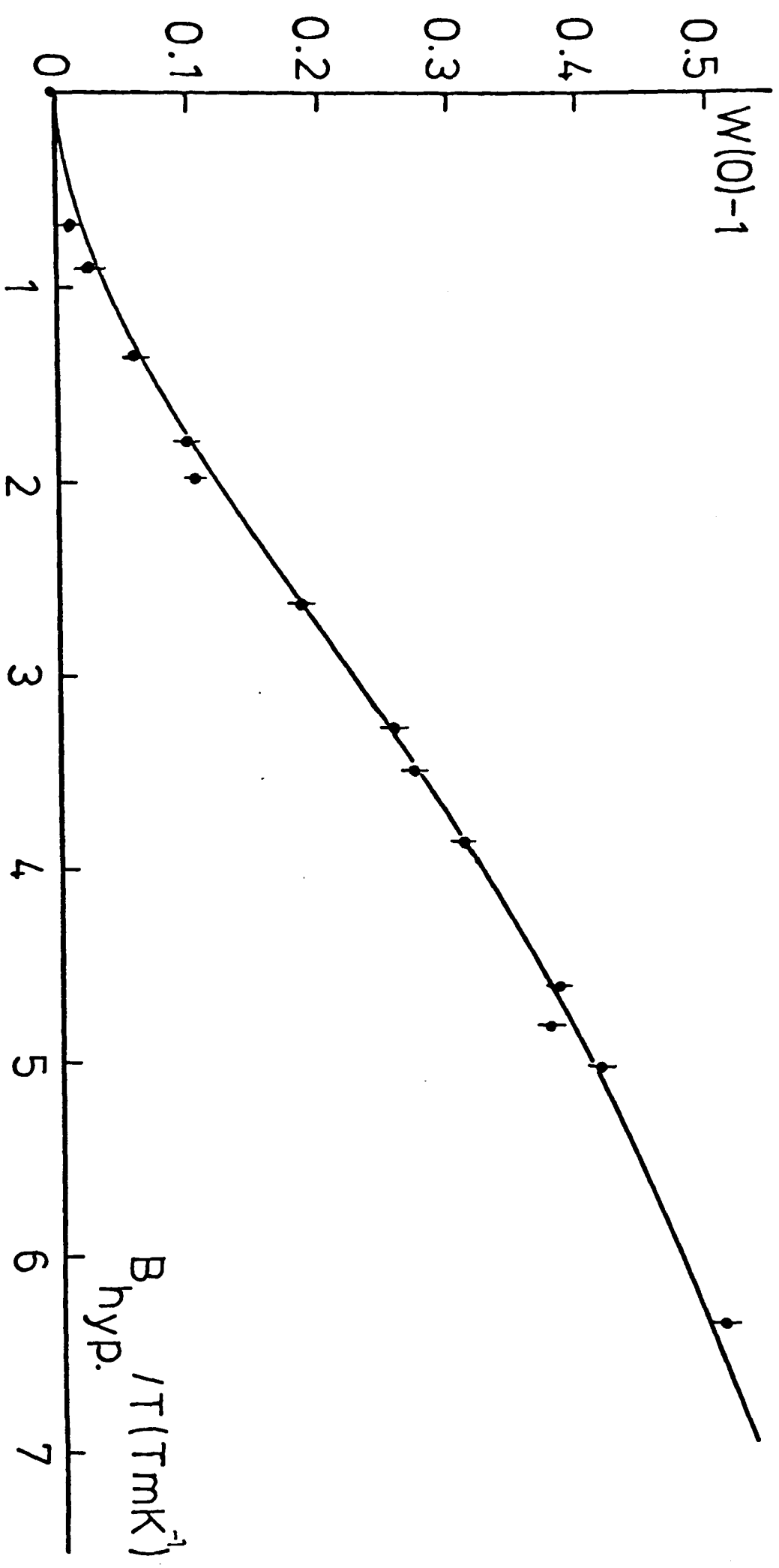


Fig. 7.7 Anisotropy of the 831 keV gamma-transition

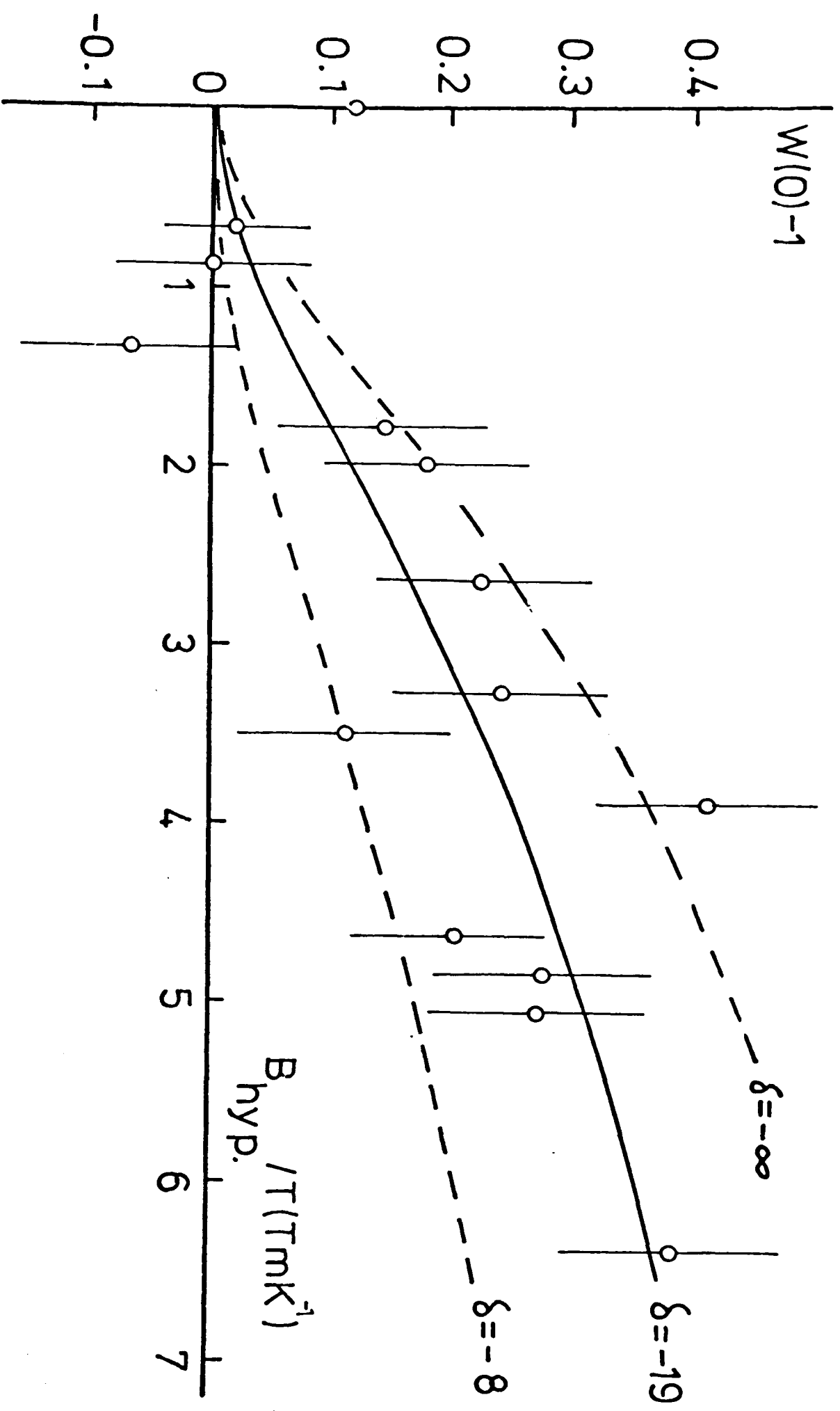


Fig. 7.8 Anisotropy of the 465 keV gamma-transition

6.5) were used throughout. One or more minima in χ^2 were found which corresponded to the solution for δ . The experimental error in δ was estimated by allowing the reduced χ^2 to increase by 1.

The particulars for the measurements made in this work are summarised in Table 7.1, where for completeness data from other sources have also been included. It should be mentioned that owing to contamination by ^{152}Eu ($t_{1/2} = 3.4\text{y}$) in the source, the 779 keV gamma-ray ($3_{\gamma}^+ \rightarrow 2_{\text{g}}^+$) could not be studied in this work. The 691 keV gamma-ray was contaminated by unresolved gamma-rays from the decay of impurities $^{110\text{m}}\text{Ag}$ ($t_{1/2} = 253\text{d}$) and ^{152}Eu . This contamination amounted to about 30% of the intensity of the 691 keV gamma-ray and was taken into account in the data analysis of the photo-peak. As a result of the contamination the uncertainty on δ was rather large. The data are presented in Figs. 7.2-7.8 with the full curves representing the 'best fit' and the dashed curves denoting the error limits. Apart from $\Delta I = 0$, all the interband transitions show negative δ . Fits for the 465 and 594 keV gamma-rays gave alternative small positive values of δ which have been discarded as all the interband transitions are expected to be almost pure E2. Accuracy of these measurements was not good enough to distinguish between large positive and large negative values of δ for $\Delta I = 0$ transitions. The values of δ for the 465 and 530 keV gamma-rays obtained in this work are in slight disagreement with those of West et al (2). However, the measured values of $U_2\text{F}_2$ and $U_4\text{F}_4$ of West et al are in good agreement with ours. The discrepancy is thought to be due to a different fitting procedure used in the angular distribution measurements of the aforementioned.

Table 7.1

Mixing ratios for the interband transitions between the γ -vibrational and ground state bands

Transition $I_i \longrightarrow I_f$	E (keV)	U_F 2 2	This work U_{4F4}	δ/E_1 (MeV ⁻¹)	Ref. 8 δ/E (MeV ⁻¹)	Ref. 2 δ/E (MeV ⁻¹)
$2^+_{\gamma} \longrightarrow 2^+_g$	705.3					
$3^+_{\gamma} \longrightarrow 2^+_g$	778.8		contaminated		-27($^{+13}_{-54}$)	
$3^+_{\gamma} \longrightarrow 4^+_g$	594.4	0.19(19) 0.16(21)	0.003(3) 0.061(1)	-24($^{+17}_{-\infty}$)>+13.5		-26 ($^{+17}_{-\infty}$)
$4^+_{\gamma} \longrightarrow 4^+_g$	691.2	0.21($^{+9}_{-10}$)	-0.210(3)	$\pm\infty$ (<-5) >+9	-5($^{+2}_{-4}$)	
$5^+_{\gamma} \longrightarrow 4^+_g$	810.3	0.075(8)	0.431(1)	-28(4)		-104($^{+71}_{-\infty}$)
$5^+_{\gamma} \longrightarrow 6^+_g$	529.8	0.212(10)	0.171(1)	-77($^{+19}_{-34}$)	-9.5 (5)	
$6^+_{\gamma} \longrightarrow 6^+_g$	670.5	0.256(19)	-0.367(1)	+85($^{+\infty}_{-60}$) <-85	-1.7 ($^{+5}_{-11}$) +9.4 ($^{+\infty}_{-43}$)	
$7^+_{\gamma} \longrightarrow 6^+_g$	830.6	0.130(12)	0.414(1)	-25(4)	-45 ($^{+20}_{-\infty}$)	
$7^+_{\gamma} \longrightarrow 8^+_g$	464.8	0.15 ($^{+9}_{-11}$)	0.211	-41($^{+24}_{-\infty}$)	-6.7 ($^{+20}_{-32}$)	
		0.23(8)	0	+0.09(6)		
$8^+_{\gamma} \longrightarrow 8^+_g$	644.4					-1.24(30) +2.5 ($^{+16}_{-9}$)
$9^+_{\gamma} \longrightarrow 9^+_g$	839.8					-12.3($^{+32}_{-\infty}$)

7.4 Band Mixing in ^{166}Er

As indicated in section 7.2, band mixing is responsible for small M_1 admixtures in transitions between the γ -vibrational and ground state rotational bands (9a). Band mixing also perturbs E2 transition rates between and within bands, and level spacings between energy levels within a given band.

Mikhailov (10) has shown that in general the reduced E2 transition probabilities between states in the γ -vibrational and ground state bands can be expressed by the equation

$$B(E2; K=2, I_i \rightarrow K=0, I_f) = 2 M_1^2 \frac{\langle I_i 22 - 2 | I_f 0 \rangle^2}{(1 + a_2 I_f (I_f + 1))} \quad 7.1$$

where $a_2 = -M_2/M_1$

and the angular brackets denote a Clebsch-Gordan coefficient.

I_i and I_f are respectively the initial and final state spins.

Equation 7.1 is quite general and makes no assumptions regarding the nature of the interband mixing. If there were no band mixing at all, a_2 and therefore M_2 would vanish. For $\Delta K = 2$ interband mixing it can be shown that (9b)

$$M_2 = \xi_\gamma \left(\frac{15}{8\pi} \right)^{1/2} eQ_0 \quad 7.2$$

where Q_0 is the intrinsic quadrupole moment for the band and ξ_γ is a parameter describing the extent of admixture of the two bands. Thus measurements of $B(E2)$ can give the extent of band mixing through Equations 7.1 and 7.2. A 'Mikhailov plot' of

$$[B(E2)]^{1/2} / \langle I_i 22 - 2 | I_f 0 \rangle \quad (\text{y-axis})$$

versus

$$I_f(I_f+1) - I_i(I_i+1) \quad (\text{x-axis})$$

would give a straight line of intercept $2M_1^2$ on the y-axis and slope $2M_1^2 a_2$. These parameters can be related to ξ_γ and Q_0 via Equation 7.2. The E2 transition probabilities for the interband and intraband transitions can be written in the form

$$B(E2; I2 \rightarrow I'0) = 2 | \langle I22-2 | I'0 \rangle |^2 | \langle 0 | M'(E2, -2) | 2 \rangle |^2 \quad 7.3a$$

$$B(E2; IK \rightarrow I'K) = | \langle IK20 | I'K \rangle |^2 | \langle K | M'(E2; 0) | K \rangle |^2 \quad 7.3b$$

where the quantities in the angular brackets to the right are matrix elements for interband and intraband transitions respectively. The intrinsic quadrupole moment for a band $Q_0(K)$ is given by the expression

$$Q_0(K) = |e|^{-1} \left(\frac{16\pi}{5} \right)^{\frac{1}{2}} \langle K | M'(E2; 0) | K \rangle \quad 7.4$$

where e is the electronic charge and the matrix element is the same as in Equation 7.3b. The intraband transition probability $B(E2)$ can be computed using Equations 7.3b and 7.4 given that Q_0 is known. The interband transition probability from an initial state I_i is then calculated from $B(E2)$ for an intraband transition from the same initial state I_i and the ratio of the measured branching ratios for the two transitions. This was the procedure used by Reich et al (1) in computing $B(E2)$ for many of the interband transitions. For transitions from the 2_γ^+ state, $B(E2)$ was calculated from the measured (absolute) Coulomb excitation probability

$$B(E2; 00 \rightarrow 22)$$

between 0_g^+ and 2_γ^+ ($= 0.13 (1) e^2 b^2$, Ref. 7) and the expression

$$B(E2; I_i \rightarrow I_f) = \frac{2I_f + 1}{2I_i + 1} B(E2; I_f \rightarrow I_i) \quad 7.5$$

Table 7.2

Table of B(E2) values for the 'Mikhailov plot'

Transition		B(E2)		
$I_i \longrightarrow I_f$		(e^2b^2)	$I_f(I_f+1)-I_i(I_i+1)$	$(B(E2))^{1/2}/\langle I_i 22-2 I_f 0 \rangle$
				(eb)
$2^+_{\gamma} \longrightarrow 2^+_g$		0.048(3)	0	0.412(11)
$2^+_{\gamma} \longrightarrow 0^+_g$		0.026(1)	-6	0.361(10)
$2^+_{\gamma} \longrightarrow 4^+_g$		0.0047(5)	14	0.573(32)
$4^+_{\gamma} \longrightarrow 2^+_g$		0.0104(11)	-14	0.295(16)
$4^+_{\gamma} \longrightarrow 4^+_g$		0.058(11)	0	0.409(39)
$4^+_{\gamma} \longrightarrow 6^+_g$		0.015(4)	22	0.71(10)
$5^+_{\gamma} \longrightarrow 4^+_g$		0.034(6)	-10	0.327(27)
$5^+_{\gamma} \longrightarrow 6^+_g$		0.046(7)	12	0.505(41)
$6^+_{\gamma} \longrightarrow 4^+_g$		0.0055(8)	-22	0.236(18)
$6^+_{\gamma} \longrightarrow 6^+_g$		0.060(9)	0	0.405(31)
$7^+_{\gamma} \longrightarrow 6^+_g$		0.026(4)	-14	0.295(23)
$7^+_{\gamma} \longrightarrow 8^+_g$		0.055(9)	16	0.525(41)
$8^+_{\gamma} \longrightarrow 8^+_g$		0.068(14)	0	0.431(46)
$3^+_{\gamma} \longrightarrow 2^+_g$		0.050(5)	- 6	0.36(2)
$3^+_{\gamma} \longrightarrow 4^+_g$		0.033(2)	8	0.47(1.5)

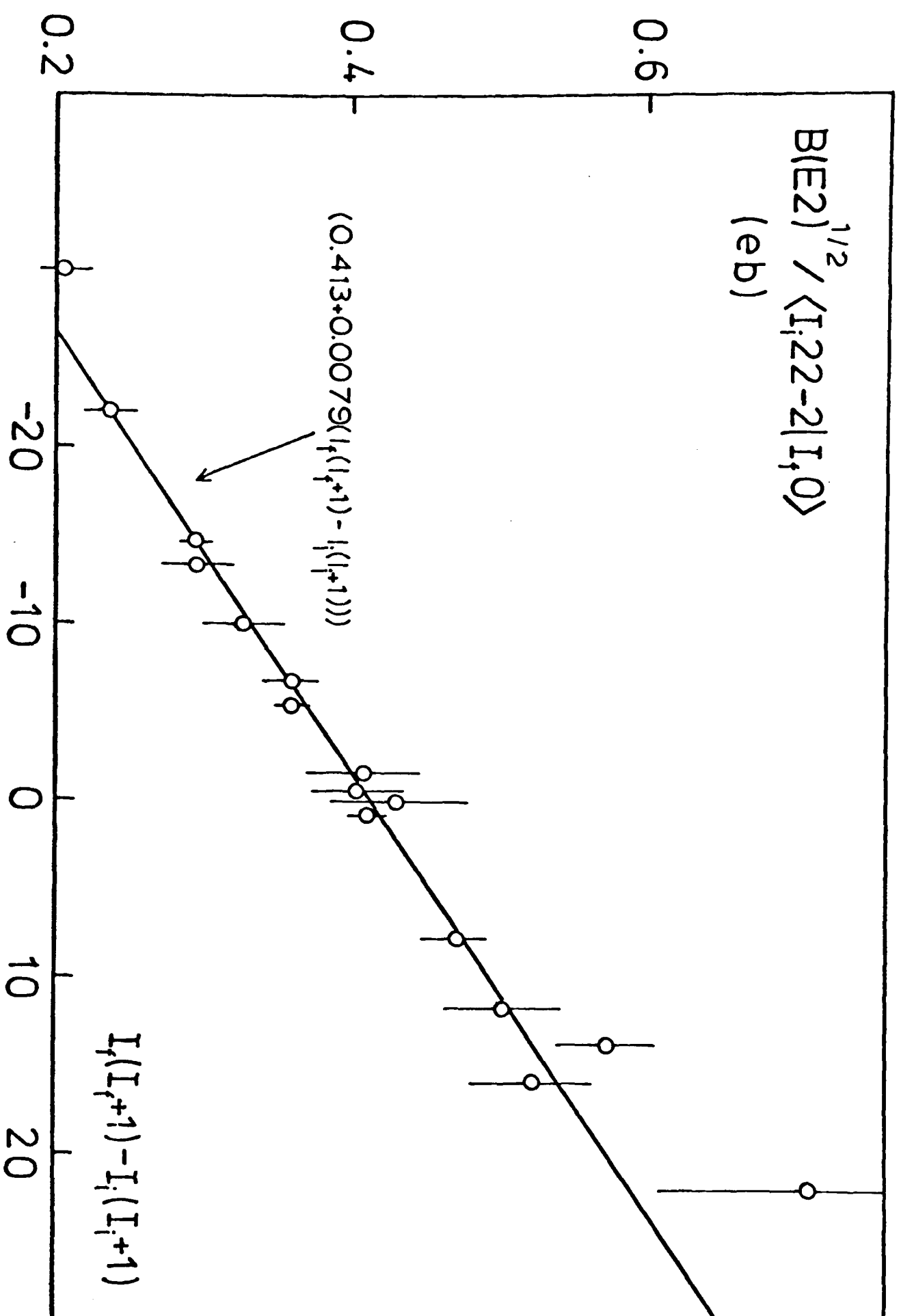


Fig. 7.9 The 'Mikhailov plot' of reduced E2 transition probabilities for interband transitions in ^{166}Er

For the transitions $3_{\gamma}^{+} \rightarrow 2_{\text{g}}^{+}$ and $3_{\gamma}^{+} \rightarrow 4_{\text{g}}^{+}$, the $B(E2)$ values were estimated as no intraband transitions are observed from the 3_{γ}^{+} state. The calculated values for (emission) $B(E2)$ are summarised in Table 7.2, where the parameters for the 'Mikhailov plot' are also tabulated. The 'Mikhailov plot' for $B(E2)$ for ^{166}Er is shown in Fig. 7.9 with a least squares fit through the points. A weighted least squares fit yielded the following estimates for M_1 and M_2 :

$$\begin{aligned} M_1 &= 0.454 (3) \text{ eb} \\ M_2 &= -8.7 (4) \times 10^{-3} \text{ eb} \end{aligned}$$

in good agreement with the results of Reich et al who first applied this analysis to ^{166}Er (1).

7.5 Evaluation of M1 Transition Probabilities for the Inter-band Transitions

The coupling between the γ -vibrational and ground state bands can give rise to M1 transitions between the bands, and the reduced matrix elements for such transitions is given by the expression (9c)

$$\begin{aligned} \langle K=2, I_i || \mu(M1) || K=0, I_f \rangle &= (2(2I_f+1)I_f(I_f+1))^{\frac{1}{2}} \\ \langle I_f || I_i || I_i \rangle &= (M_1 + M_2(I_i(I_i+1) - I_f(I_f+1))) \end{aligned} \quad 7.6$$

For $\Delta K = 2$ band mixing

$$M = -2 \sqrt{2} \left(\frac{3}{4\pi} \right)^{\frac{1}{2}} \frac{e\hbar}{2M} (g_K (K=2) - g_R) \epsilon_{\gamma}$$

$$M_1 = -4M_2 \quad 7.7$$

$$\text{and } M_2 = \frac{1}{2}M$$

Equation 7.6 once again is quite general and makes no assumptions regarding the origin of the band mixing. For the case that the

M1 transitions arise from admixtures of a $K = 1$ band, $M_2 = 0$.

Thus plotting

$$\begin{aligned} & \langle K = 2, I_i || \mu(M1) || K = 0, I_f \rangle / (2(2I_f+1) I_f (I_f+1))^{1/2} \\ & \times \langle I_f 111 || I_i 2 \rangle \end{aligned}$$

against

$$I_i (I_i+1) - I_f (I_f+1)$$

for the interband transitions could give a straight line of slope

$$0 \text{ for } \Delta K = 1$$

$$\text{and } M_2 \text{ for } \Delta K = 2$$

To calculate the reduced matrix element of Equation 7.6, we use the expression

$$\delta/E \text{ (MeV)} = 0.835 \frac{\langle I_f || \mu(E2) || I_i \rangle}{\langle I_f || \mu(M1) || I_i \rangle} \quad 7.8$$

where E2 and M1 reduced matrix elements for emission are in units of electron barns (eb) and nuclear magnetons (μ_N) respectively and

$$B(\pi\lambda, I_i \rightarrow I_f) = \frac{1}{2I_i+1} |\langle I_f || \mu(\pi\lambda) || I_i \rangle|^2 \quad 7.9$$

Also

$$\langle I_i || \mu(M1) || I_f \rangle = (-)^{I_f-I_i} ((2I_i+1)B(M1, I_i \rightarrow I_f))^{1/2} \quad 7.10$$

Thus in terms of reduced transition probabilities $B(M1)$, the 'Mikhailov type plot' is equivalent to plotting

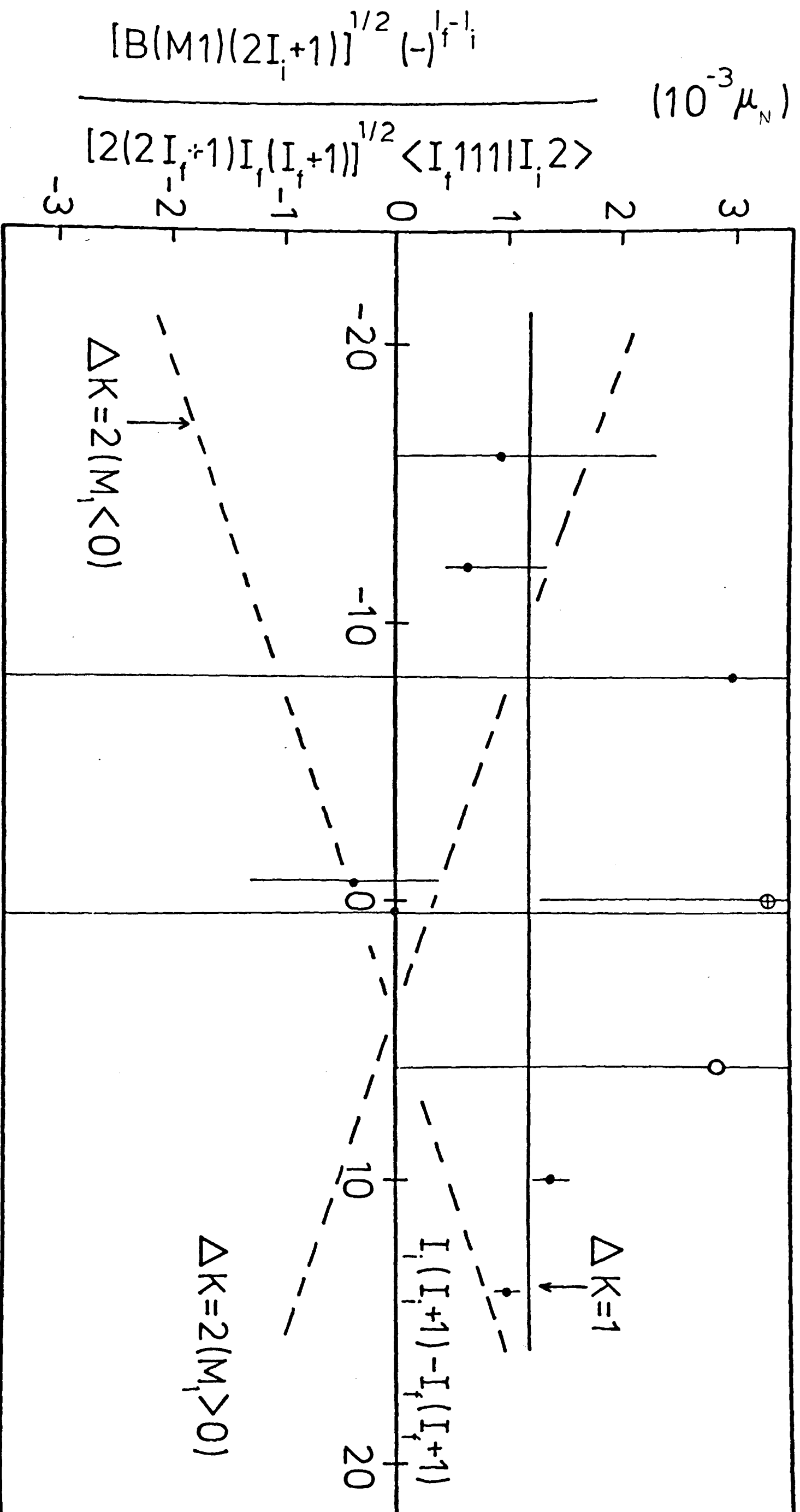


Fig. 7.10 The 'Mikhailov-type plot' of reduced M1 transition probabilities for interband transitions in ^{166}Er (filled in circles this work, $\oplus$ ref. 8, $\circ$ ref. 2)

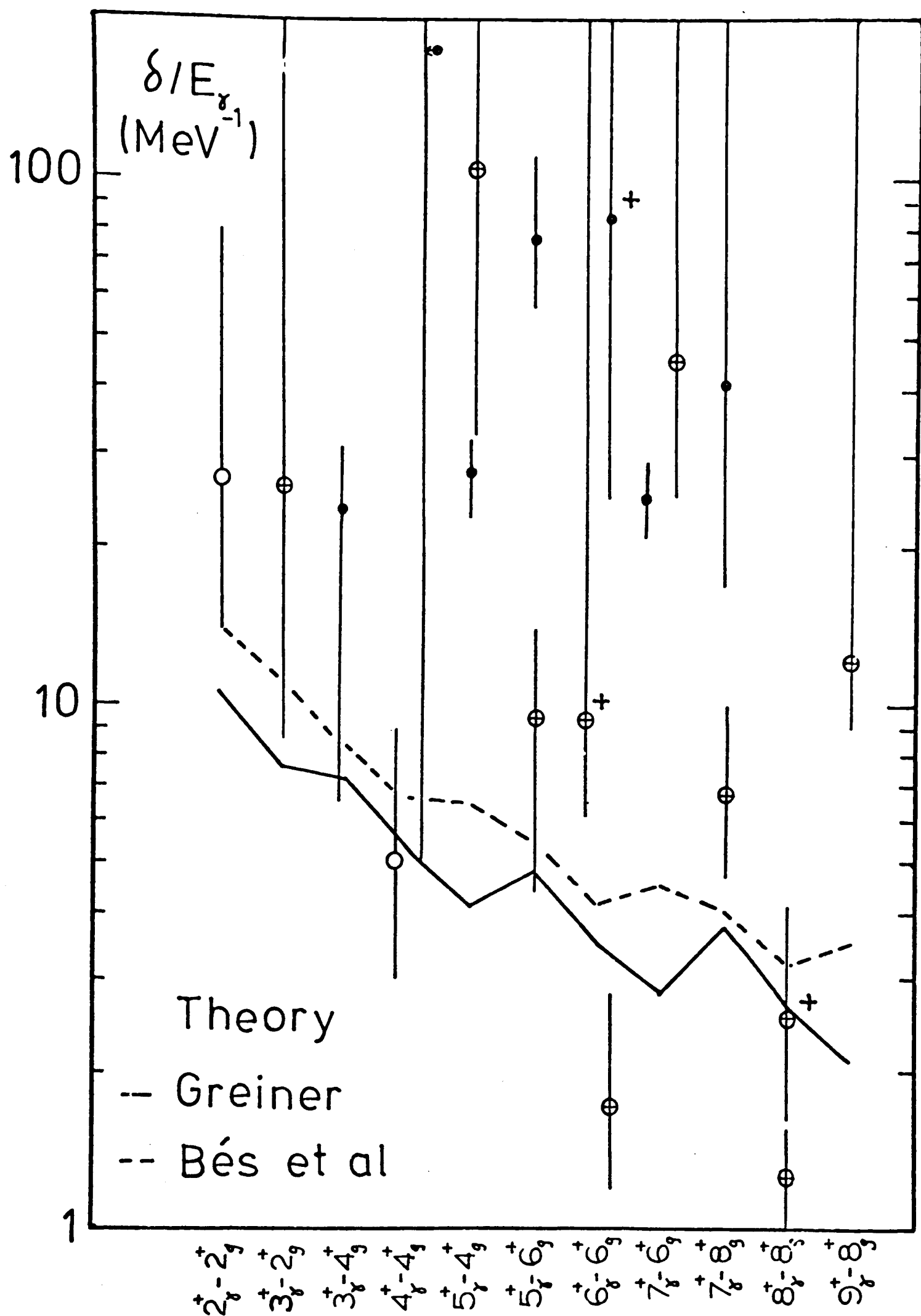


Fig. 7.11 Reduced mixing ratios for transitions between $(\pi=2^+)$ γ -vibrational band and $(K^\pi=0^+)$ ground state rotational band (filled in circles this work, $\oplus$ ref. 2, $\circ$ ref. 8)

$$((2I_i+1) B(M1, I_i \rightarrow I_f))^{\frac{1}{2}} (-)^{I_f-I_i} / (2(2I_f+1) I_f (I_f+1))^{\frac{1}{2}}$$

$$x < I_f \parallel I_i 2 >$$

against $I_i (I_i+1) - I_f (I_f+1)$.

Equations 7.8 and 7.10 define a phase convention which gives positive (negative) values for (emission) $(B(M1))^{\frac{1}{2}}$ for positive (negative) δ (Equation 7.8). The calculations for $B(M1)$ are summarised in Table 7.3. The 'Mikhailov type plot' for $B(M1)$ is shown in Fig. 7.10 with theoretical lines for $\Delta K = 1$ and $\Delta K = 2$ band mixing. The theoretical line for $\Delta K = 2$ was derived from Equation 7.7 with the following estimates for ξ_γ and $|g_K - g_R|$ taken from the measurements of Reich and Cline (1)

$$|g_K - g_R| \approx 0.11$$

$$\xi_\gamma \sim -10^{-3}$$

The uncertainty in the sign of $g_K - g_R$ gives two lines for $\Delta K = 2$ mixing. The theoretical line for $\Delta K = 1$ seems to be in better agreement with the data points than either of the lines for $\Delta K = 2$. This is consistent with the systematics for nuclei in this region and is discussed by Krane (8). From our measurements, we estimate

$$M_1 \approx 1.2 \times 10^{-3} \mu_N$$

A review by Krane (8) discusses a number of attempts to calculate the magnitude of the mixing ratio δ for interband transitions. Notable among these are theories by Bès et al (11) and Greiner (12), which predict positive and negative values of δ respectively for interband transitions between a

γ -vibrational band and ground state band. These calculations

Table 7.3

Table of B(M1) values for the 'Mikhailov type plot'

$I_i \longrightarrow I_f$	$I_i(I_i+1) - I_f(I_f+1)$	$B(M1)$ ($10^{-6}\mu_N^2$)	$\frac{(B(M1)(2I_i+1))^{\frac{1}{2}}(-)^{I_f-I_i}}{(2(2I_f+1)I_f(I_f+1))^{\frac{1}{2}} < I_f^{111} I_i 2 >}$ ($10^{-3}\mu_N$)	Ref.
$2^+_2 \gamma$	0	4 ($^{+13}_{-3}$)	3.4 ($^{+400}_{-20}$)	8
$3^+_2 \gamma$	6	49 ($^{+460}_{-49}$)	2.9 ($^{+50}_{-29}$)	2
$3^+_3 \gamma$	-8	39 ($^{+420}_{-39}$)	3.0 ($^{+80}_{-80}$)	This work
$4^+_4 \gamma$	0	0 ($^{+1600}_{-0}$)	0 ($^{+9}_{-5}$)	This work
		1600 ($^{+2900}_{-1100}$)	9.5 ($^{+60}_{-40}$)	8
$5^+_4 \gamma$	10	30 (7)	1.4 (1.5)	This work
$5^+_5 \gamma$	-12	5.4 ($^{+60}_{-30}$)	0.65 (20)	This work
$6^+_6 \gamma$	0	5.8 ($^{+120}_{-58}$)	-0.38 ($^{+80}_{-90}$)	This work
$7^+_7 \gamma$	14	29 (6)	1.0(1)	This work
$7^+_8 \gamma$	-16	23 ($^{+110}_{-23}$)	0.95($^{+130}_{-95}$)	This work
$8^+_8 \gamma$	0	7600 ($^{5000}_{-7600}$)	-7.6 ($^{+30}_{-40}$)	8

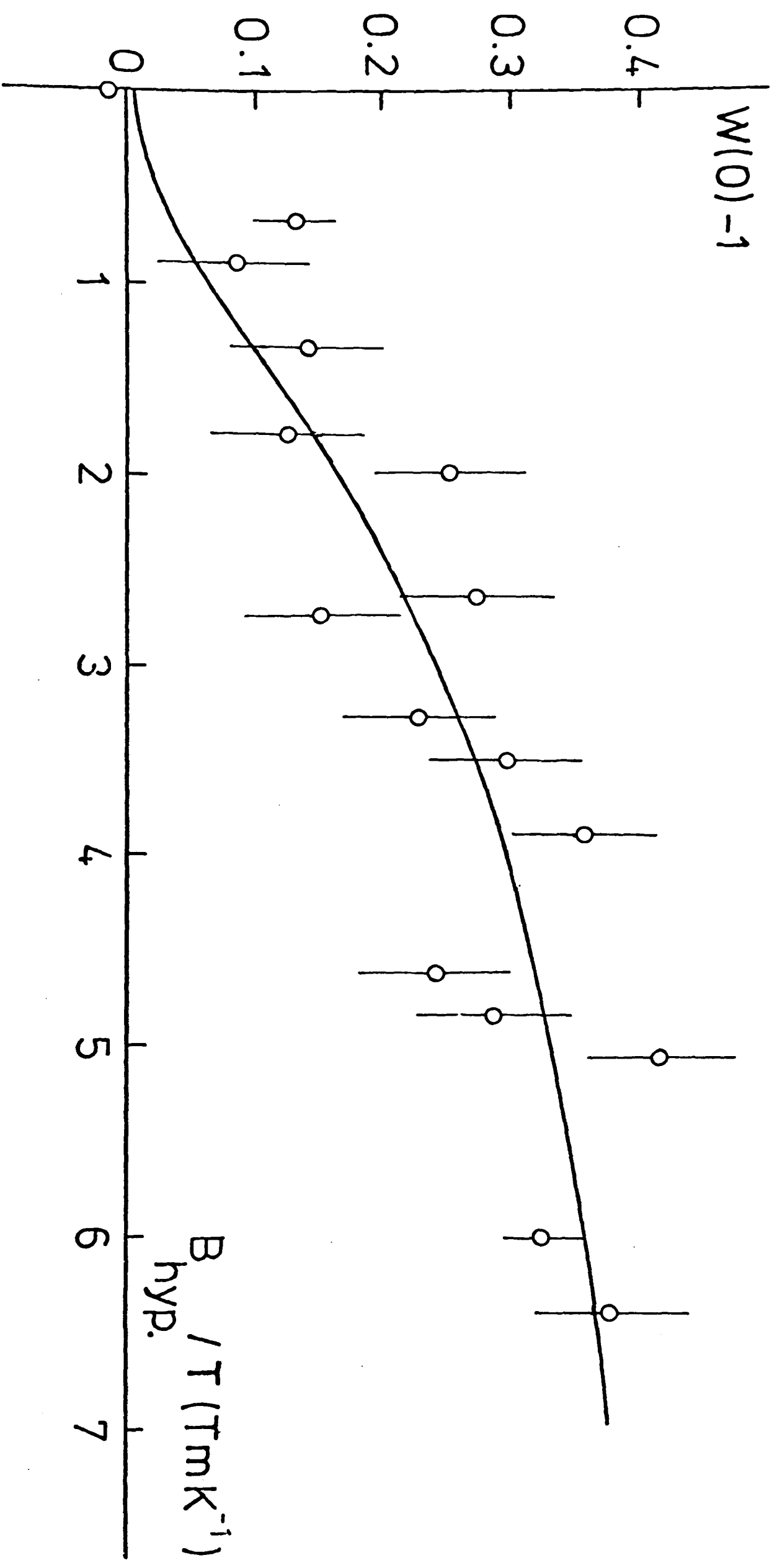


Fig. 7.12 Anisotropy of the 1401 keV gamma-transition

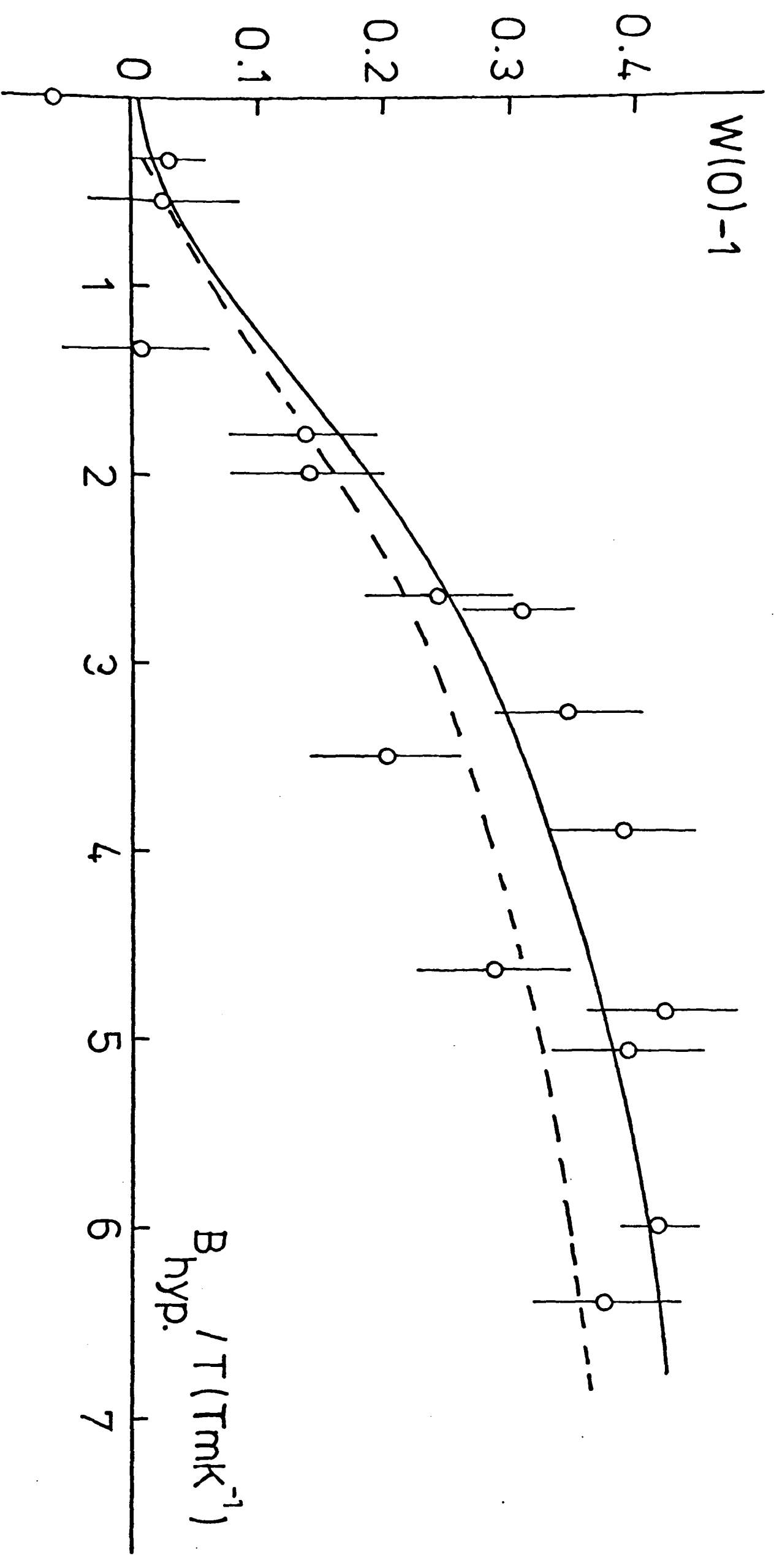


Fig. 7.13 Anisotropy of the 1427 keV gamma-transition (dashed curve theory for $I_1=5^-$)

(Fig. 7.11) give values for δ/E which are rather smaller than those measured in this work.

7.6 The $K^\pi = 2^-$ Octupole Vibrational Band

Nuclear reaction experiments have indicated the presence of several octupole excitation modes based on the $K^\pi = 2^-$ band at 1458 keV and a rotational band based on the $K^\pi = 4^-$ band at 1572 keV (3, 4). On the basis of gamma-ray intensity measurements, Burke et al (4) have assigned spins and parities 2^- , 3^- and 4^- for the 1458, 1514 and 1596 keV levels respectively. These assignments for spins and parities predict $I = 5^-$ for the 1666 and 1692 keV levels. Both these levels are weakly fed in the decay of ^{166m}Ho and have been studied in this work. The levels de-excite to give gamma-rays of energy 1401 and 1427 keV respectively. The data for these gamma-rays are plotted in Fig. 7.12 and 7.13 respectively, with a smooth ('best fit') curve through each set of points. Below is a table of measured U_2F_2 values for each gamma-ray along with predictions for initial spins 4^- , 5^- and 6^- .

Table 7.4

U_2F_2 and U_4F_4 values for $K^\pi = 2^-$ octupole vibration band

$I_i \rightarrow I_f$	E_γ (keV)	Measured		Theory					
		U_2	F_2	U_2F_2	U_4F_4	U_2F_2	U_4F_4	U_2F_2	U_4F_4
		2	2	$(I_i = 4^-)$		$(I_i = 5^-)$		$(I_i = 6^-)$	
$(5^-)4^+$	1400.7	0.256(45)	-0.38	0	0.250(23)	0	-.28 to -.8	-.1 to +.4	
$(5^-)4^+$	1427.0	0.291(50)	-0.38	0	0.250(23)	0	-.28 to -.8	-.1 to +.4	

Table 7.4 shows that $I_i = 5^-$ gives the best fit to the data thus confirming the spin designations of the Nuclear Data

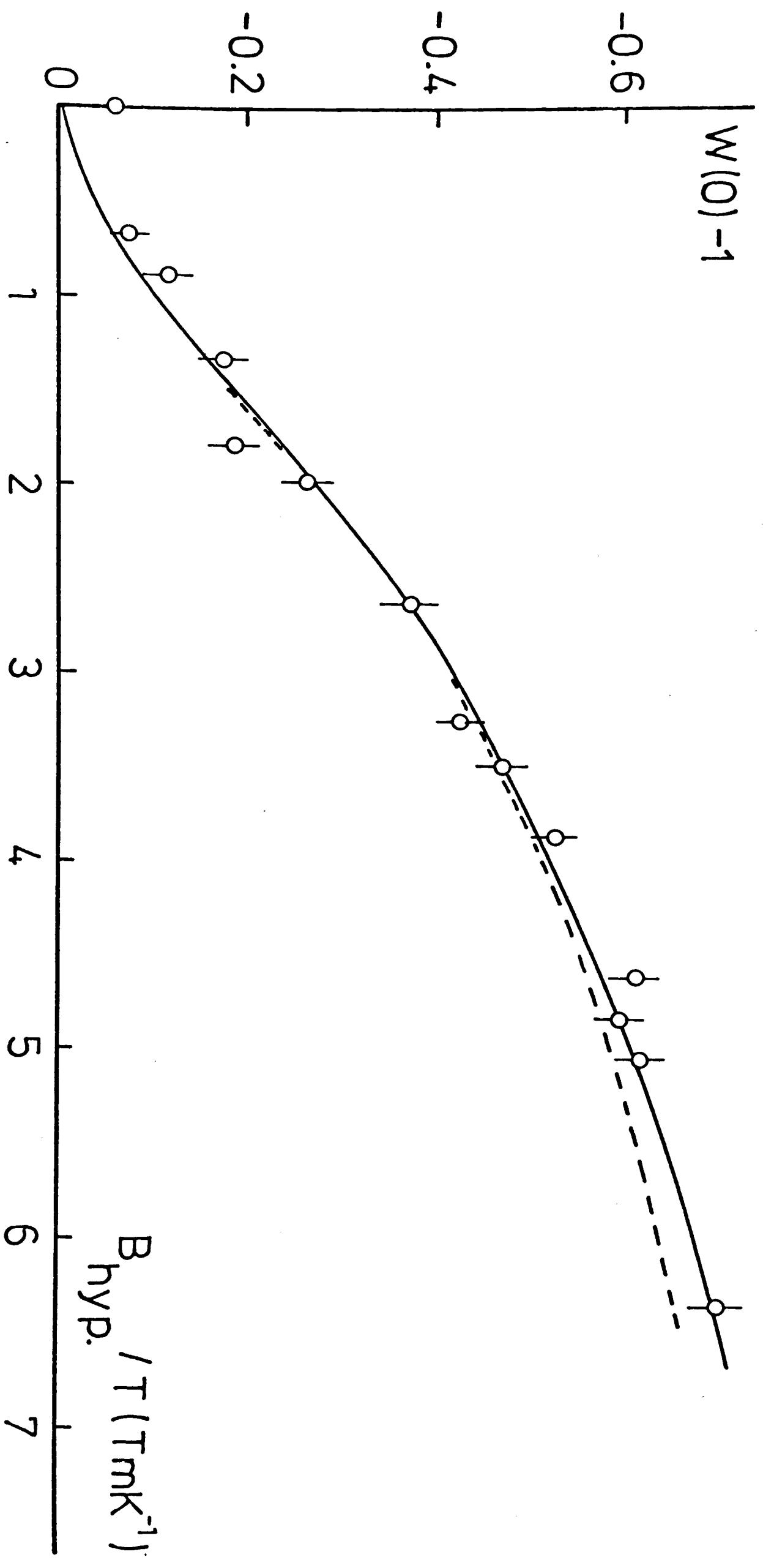


Fig. 7.14 Anisotropy of the 215 keV gamma-transition (dashed curve theory)

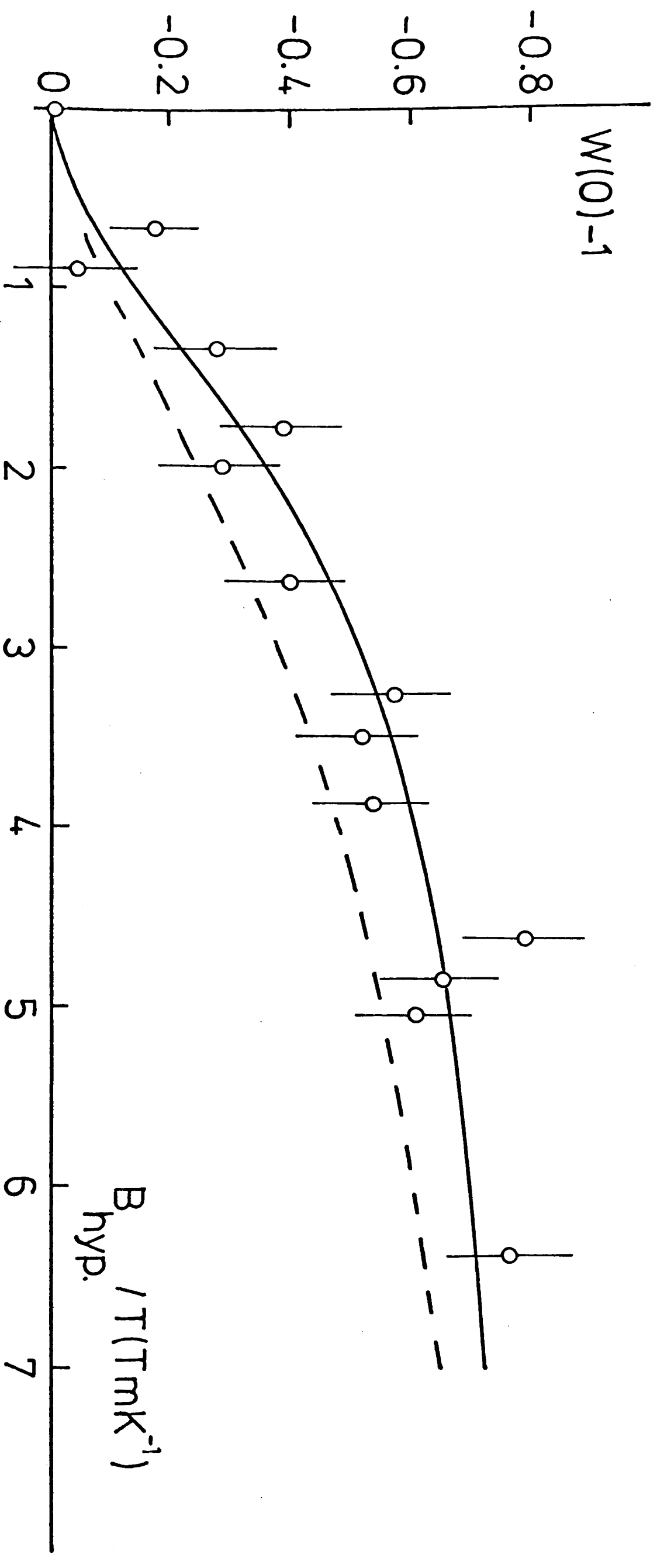


Fig. 7.15 Anisotropy of the 260 keV gamma-transition (dashed curve theory)

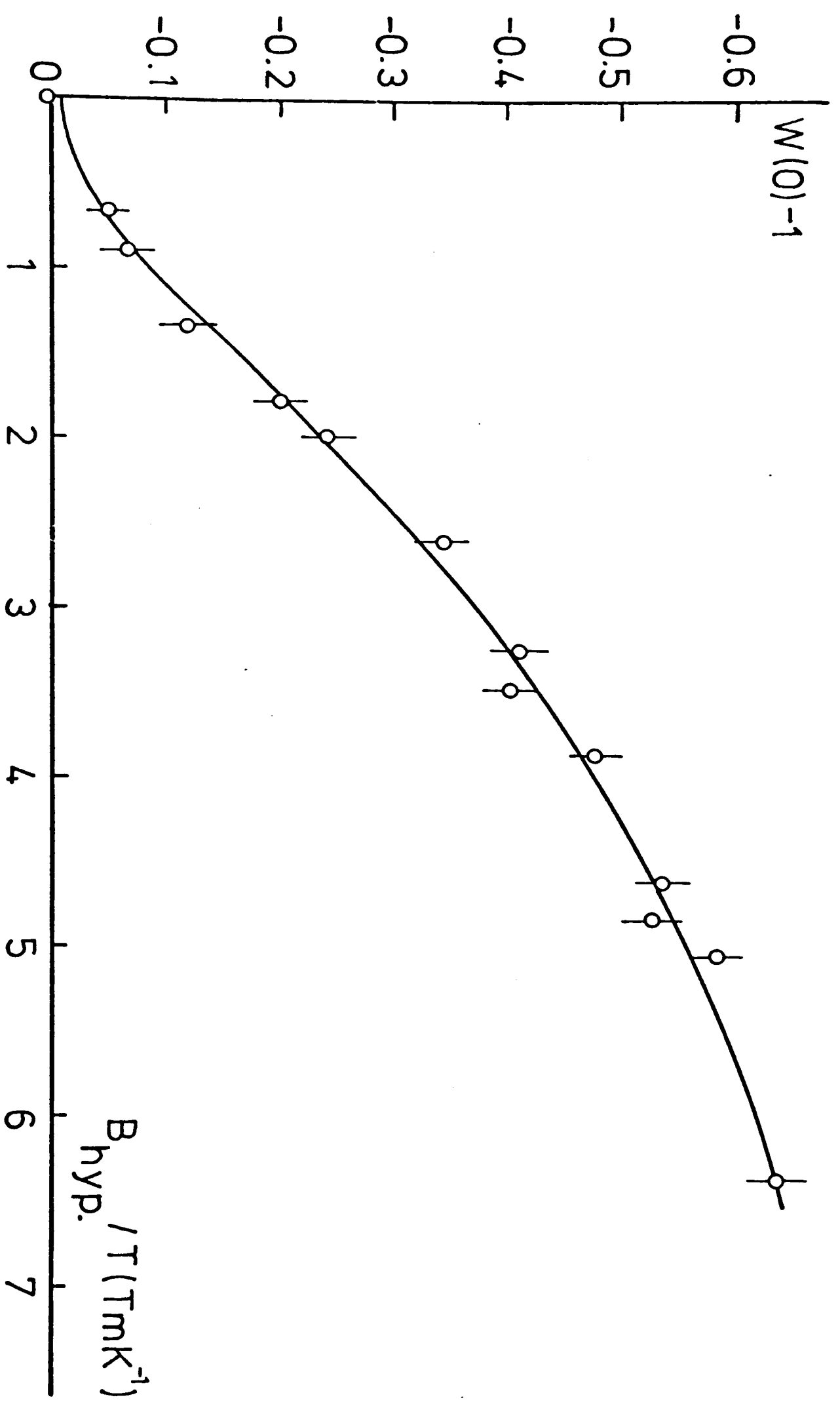


Fig. 7.16 Anisotropy of the 301 keV gamma-transition

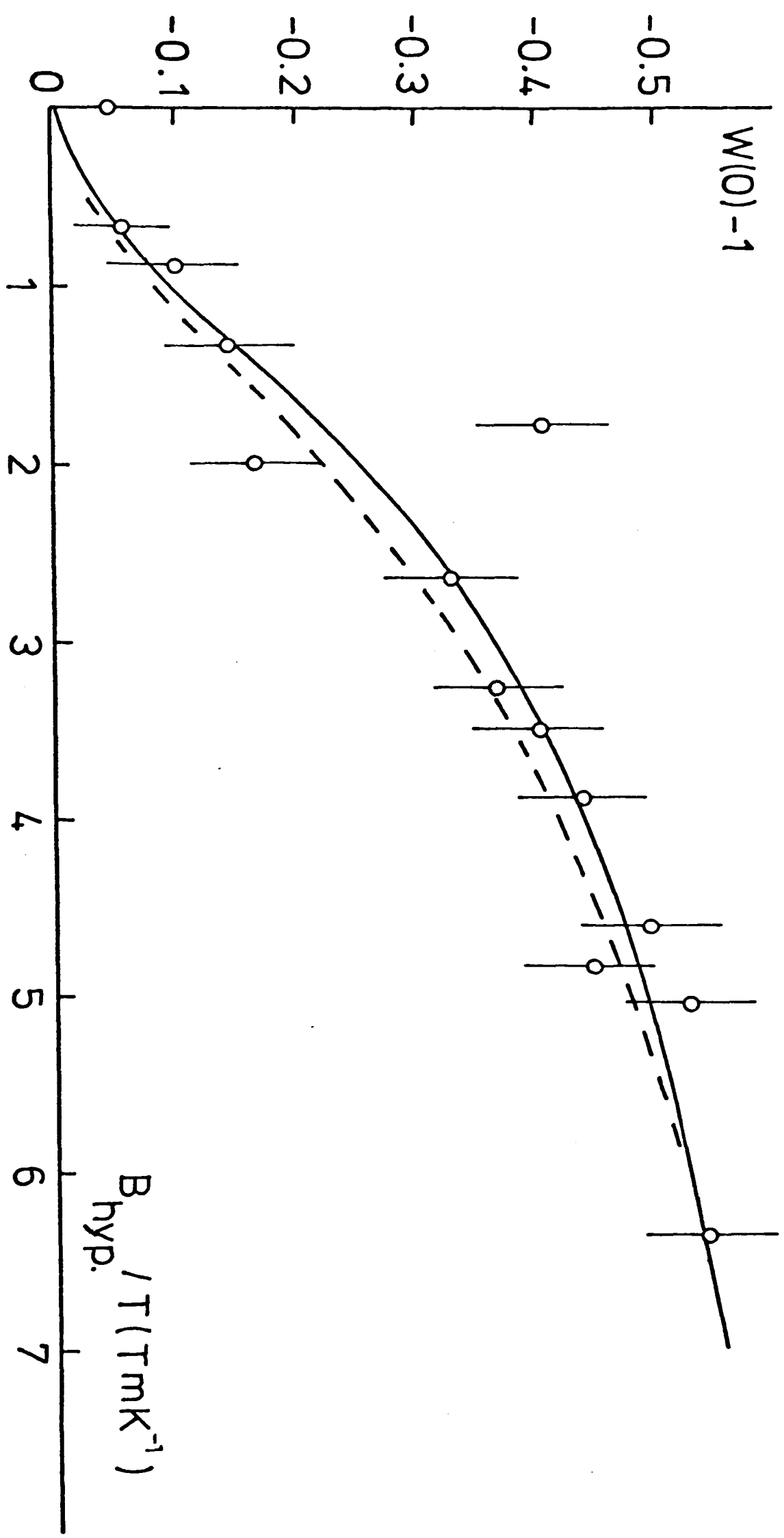


Fig. 7.17 Anisotropy of the 366 keV gamma-transition (dashed curve theory)

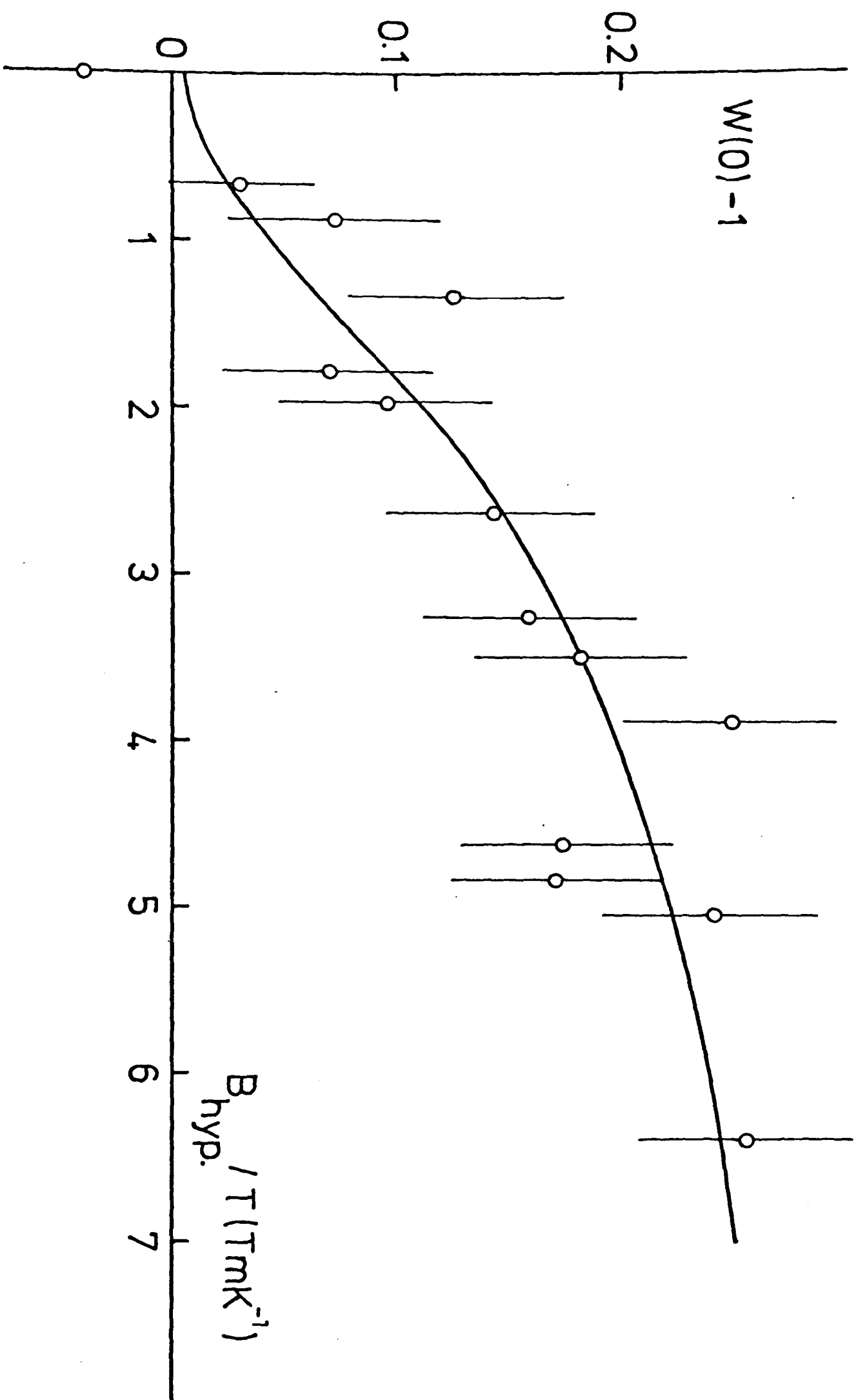


Fig. 7.18 Anisotropy of the 452 keV gamma-transition

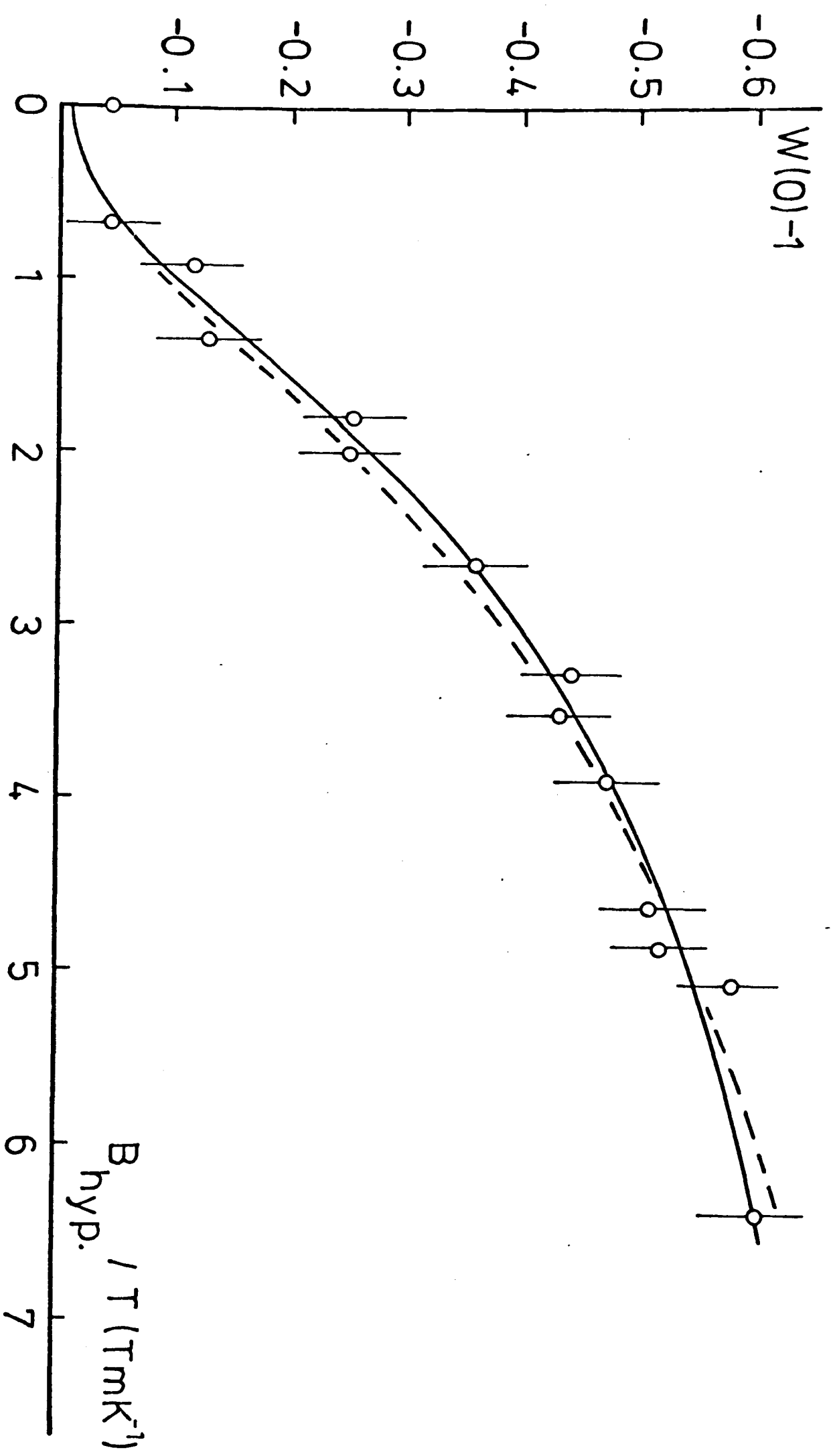


Fig. 7.19 Anisotropy of the 951 keV gamma-transition (dashed curve theory)

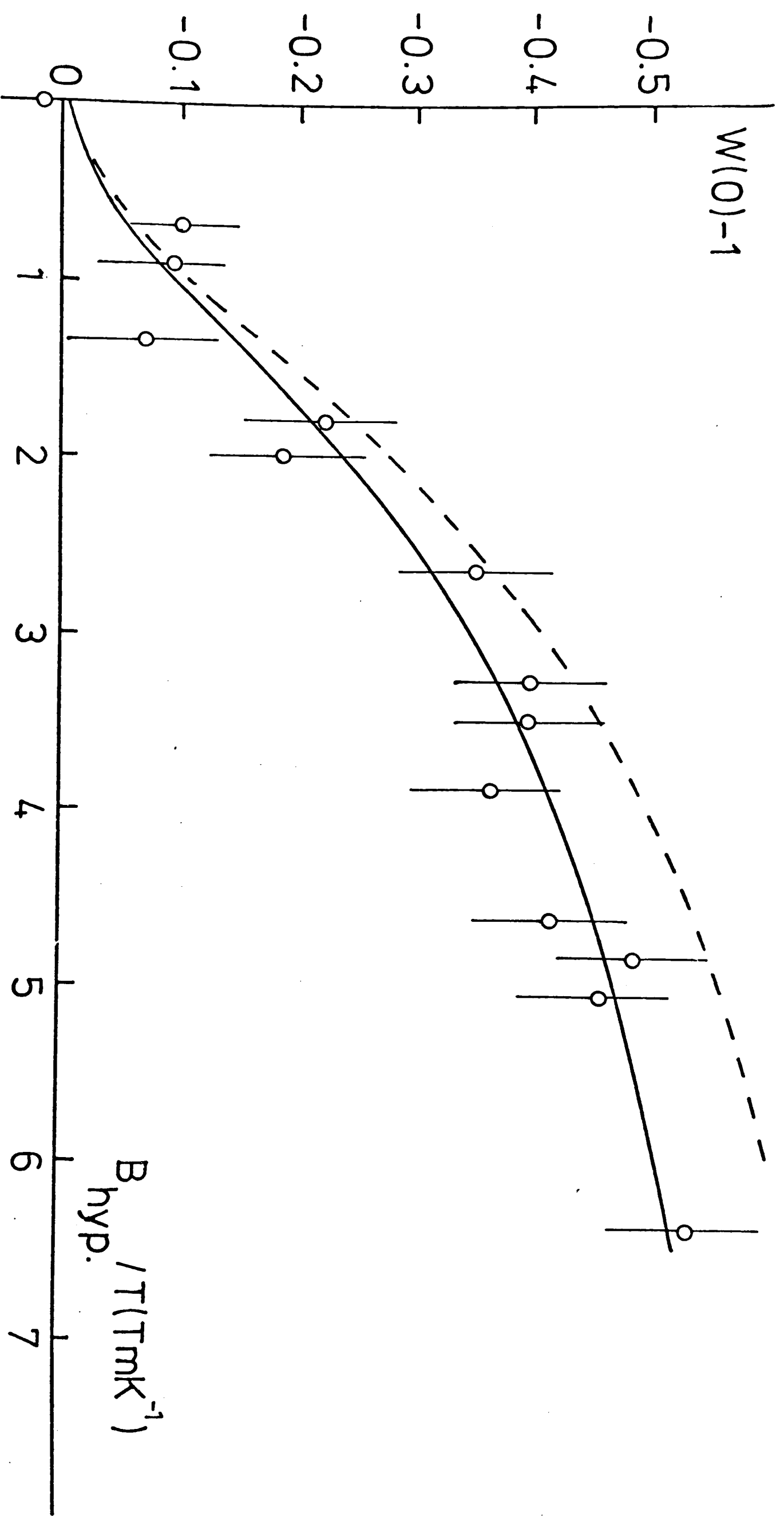


Fig. 7.20 anisotropy of the 1241 keV gamma-transition (dashed curve theory)

Sheets (13).

7.7 Other Gamma-Ray Transitions

A number of other transitions were studied which confirmed the known spins and parities of some of the levels of the γ -vibrational and ground state bands. The results for U_2F_2 and U_4F_4 are summarised in Table 7.5 which also contains theoretical values. For the E2 transitions both U_2F_2 and U_4F_4 were allowed to vary in the least-squares fitting program. The measured values of U_2F_2 and U_4F_4 are in broad agreement with the predictions, within the limits of experimental error. The measured value of U_2F_2 for the 260 keV gamma-ray is outside the estimated error limits, which correspond to about 1 standard deviation. Any unresolved peak within the 260 keV photo-peak 'window' would reduce the magnitude of the anisotropy effects; but the magnitude of the measured value is bigger than the theoretical predictions. Moreover, both the initial and final state spins for the gamma-ray are well established. The source of the discrepancy is not known and may be due to a systematic error in the data analysis. The data for the above transitions are plotted in Figs. 7.14 - 7.20, with the full curves denoting the 'best fit'.

Table 7.5

Some measured U_2F_2 and U_4F_4 coefficients for ^{166}Er

Transition $I_i \longrightarrow I_f$	E (keV)	Intensity (%)	Measured		Theory		Multipolarity	Comments
			U_2F_2	U_4F_4	U_2F_2	U_4F_4		
$6^- \longrightarrow 4^-$	214.8	0.55	-0.41(6)	-0.1(2)	-0.389(1)	-0.186(1)	E2	E2 confirmed
$5^+_{\gamma} \longrightarrow 3^+_{\gamma}$	215.9	2.6						
$6^+_{\gamma} \longrightarrow 4^+_{\gamma}$	259.7	1.1	-0.57(12)	0.15(29)	-0.362(1)	-0.143(1)	E2	see text
$7^+_{\gamma} \longrightarrow 5^+_{\gamma}$	300.7	3.8	-0.34(5)	-0.22(10)	-0.367	-0.151	E2	E2 confirmed
$8^+_{\gamma} \longrightarrow 6^+_{\gamma}$	365.7	2.5	-0.39(5)	+0.01(10)	-0.328(15)	-0.10(20)	E2	E2 confirmed
$6^- \longrightarrow 7^-_{\gamma}$	451.5	3.1	+0.17(5)		0.1718		E1	E1 confirmed
$6^- \longrightarrow 6^+_{\gamma}$	611.5	1.4		unresolved doublet			E1	
$6^+_{\gamma} \longrightarrow 4^+_{\gamma}$	950.9	3.0	-0.41(6)	-0.03(14)	-0.367	-0.151	E2	E2 confirmed
$6^- \longrightarrow 6^+_{\gamma}$	1241.4	1.0	-0.36(6)		-0.43		E1	E1 confirmed

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Chapter 7

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CHAPTER 8

NUCLEAR REFRIGERATION USING THE ENHANCED
NUCLEAR PARAMAGNET HoVO_4

8.1 Introduction

The first nuclear cooling experiment performed in 1956 by Kurti et al (1) cooled copper nuclei to a few μK although the conduction electrons and lattice within the metal remained at about 10 mK. Those early experiments were successful because the conduction electrons were essentially decoupled from the nuclei owing to the long spin-lattice relaxation times at mK temperatures. It was not until 1965 that March and Symko (2) managed to cool conduction electrons in copper to about 7 mK by adiabatic demagnetisation. It is usual to distinguish between cases where only the nuclei are cooled and cases where the conduction electrons and/or lattice are also cooled. The former is termed nuclear cooling while the latter is referred to as nuclear refrigeration.

Andres and Bucher have investigated several intermetallic compounds of praseodymium for nuclear refrigeration and their work has been reviewed by Andres (3). These 'Van Vleck' compounds have enhancement factors of order 10-20. PrNi_5 ($T_c \sim 0.6 \text{ mK}$), for example, has been cooled to 0.5 mK from dilution fridge temperatures (4).

Nuclear refrigeration using non-metallic crystals is restricted by poor thermal contact and long relaxation times. In principle, thermal contact can be improved by using small crystals, thereby increasing the surface area. Using powdered specimens of the electronic paramagnet cerium magnesium nitrate (CMN) in glycerol-water solution, samples have been cooled to about 2 mK by adiabatic demagnetisation. A large surface area of contact ($\sim 1000 \text{ cm}^2$) between the CMN and the sample is obtained by using copper foils, incorporated into the slurry. Rosenblum et al (5) used a pill consisting of compressed CMN

and gold to cool specimens to about 3 mK. This was the technique used in this work, where the lowest lattice temperature reached was 3.9 (3) mK.

8.2 Thermal Contact and Nuclear Refrigeration

As explained in Chapter 4, Kapitza boundary resistance between an insulator and a metal makes cooling of specimens using adiabatic demagnetisation of an insulator difficult. However, the situation can be improved by using fine powders which serve to increase the effective area of contact. Incorporating the powder particles into a copper matrix allows good heat extraction from the nuclear refrigerant. The rate of heat flow across a metal-insulator interface, $\dot{Q}$, is given by the equation (6)

$$\dot{Q} = \frac{A \delta T}{R_K} \quad \text{where} \quad \delta T \ll T \quad 8.1$$

A is the effective area of contact, δT is the temperature drop across the interface and R_K the boundary resistance is proportional to T^{-3} . Taking

$$R_K \sim 15 T^{-3} \text{ K s cm}^2 \text{ J}^{-1} \quad (7)$$

for a typical metal-insulator interface

$$\dot{Q} = \frac{AT^3}{15} \delta T \quad \text{K s cm}^2 \text{ J}^{-1} \quad 8.2$$

Equation 8.2 can be used to estimate the temperature difference

δT between the insulating powder and the surrounding copper matrix (of much finer particle size), if it is assumed that there is negligible thermal resistance within the crystallites themselves. For spherical crystallites of diameter d_p , the contact area A between the crystallites and the copper powder is given by the approximate relation

$$A \simeq \frac{6Vf}{d_p} \quad 8.3$$

where V is the total volume of powder and f the filling factor takes into account spaces within the pill. For a pill containing 1 cm^3 of salt of average particle diameter $5 \mu\text{m}$, $A \sim 10^4 \text{ cm}^2$ and the temperature gradient δT is of order

$$0.02 \text{ mK per nW of powder}$$

at 4 mK for $f = 1$. This gives a temperature drop between the pill and 'cold finger' of 0.1 mK for a typical heat leak of 5 nW. It should be emphasised that these estimates are very approximate but are probably accurate to within a factor of two. However, thermal resistance measurements (section 8.6.4) showed that for compressed HoVO_4 -Cu pills the effective area of contact was only 30 cm^2 .

8.3 Experimental Details

8.3.1 The HoVO_4 -Copper Pill

Single crystals of HoVO_4 were ground down to between $1\text{-}10 \mu\text{m}$ diameter using an agate pestle and mortar. The powder was then treated with dilute nitric acid for about 12 hours to remove traces of lead fluoride flux trapped during crystal growth. After washing the treated powder in de-ionised water, it was dried in an oven at about 60°C for several hours. The powder was then mixed with $1\text{-}5 \mu\text{m}$ high purity (99.998%) copper powder (supplied by Materials Research Company Limited) under a dry nitrogen atmosphere so as to give a mixture of 40% HoVO_4 and 60% copper by volume. The mixing was done for about two hours using absolute alcohol as the wetting agent. After drying, the mixture was kept sealed under vacuum to prevent oxidation of the copper.

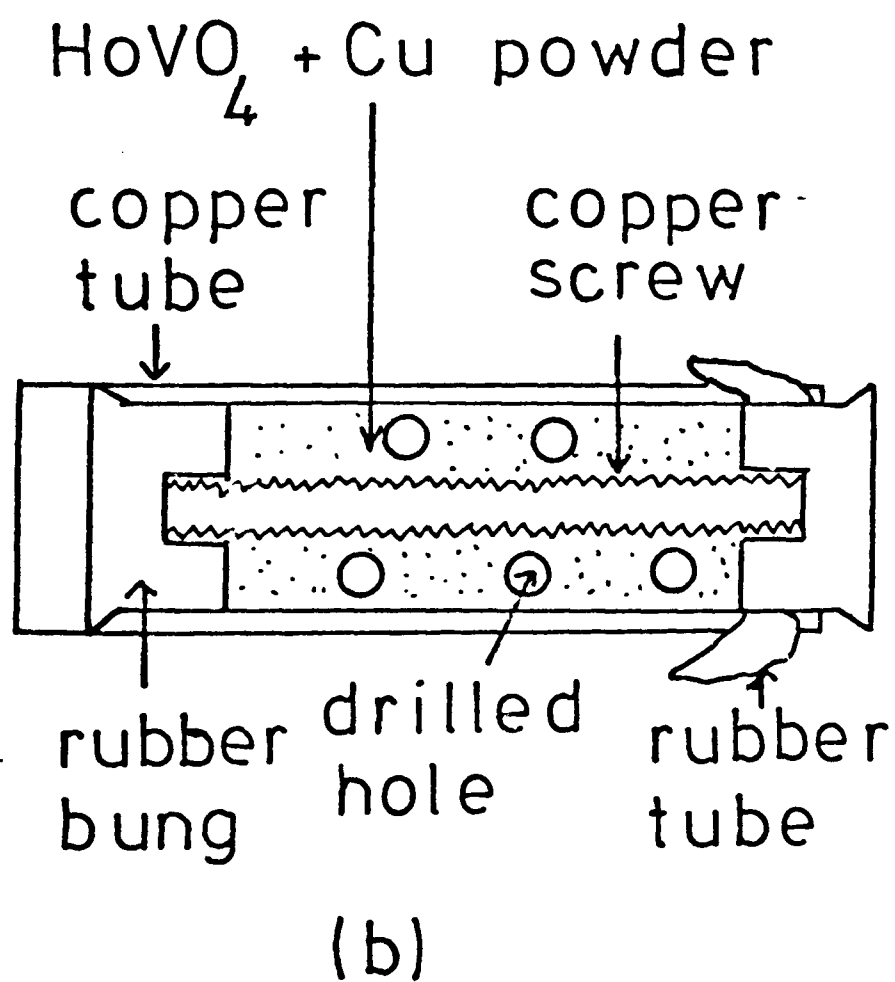
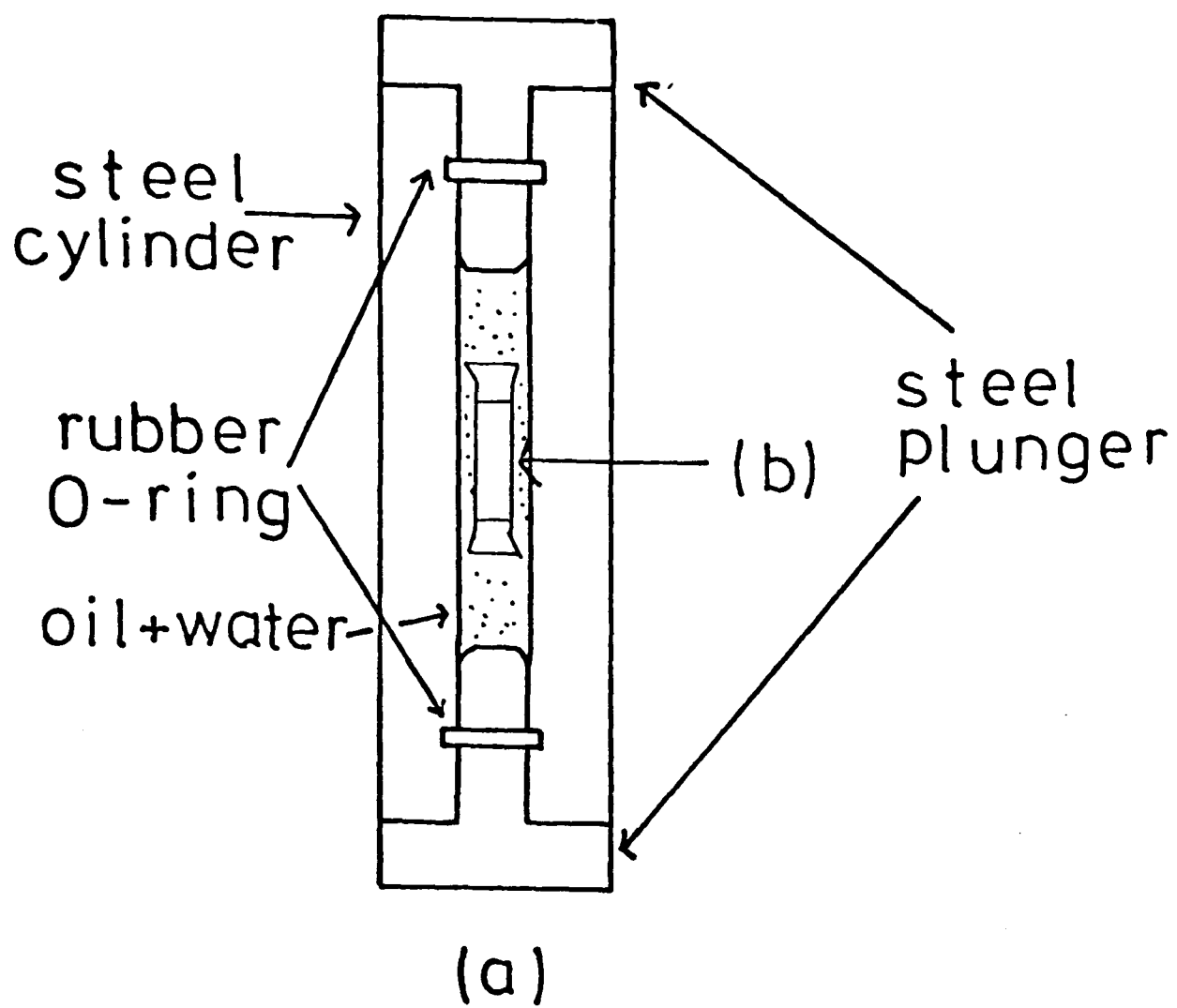


Fig. 8.1 Schematic diagram of the cold-press technique

The powdered HoVO_4 -copper mixture was compressed into a solid cylinder approximately 2 cm long and 1 cm diameter by a technique which is illustrated in Fig. 8.1. The powder was packed into a rubber sleeve, sealed at both ends, and cold pressed inside a steel cylinder at about 1900 atmospheres using a small hand press. Hydrostatic pressure was transmitted to the powder through an oil-water emulsion contained in the steel cylinder and sealed at both ends by means of rubber O-rings located in grooves in the metal. Thermal contact to the pill was by means of a single 6BA copper screw which was located centrally in the powdered mixture before pressing.

Typically, a filling factor of 80-90% was obtained in these pills. The pills were fragile but could be heat-treated to improve mechanical strength. This was done in a hydrogen atmosphere at about 600°C . However, this produced cracks inside the pill.

8.3.2 Superconducting Heat-Switch

In nuclear refrigeration experiments, it is necessary to be able to thermally isolate the nuclear refrigerant from the pre-cooling stage during and after demagnetisation. This is conveniently achieved in practice by using a superconducting heat-switch, usually in the form of wires.

A good superconducting heat-switch must have good thermal conductivity in the 'on' or normal state and negligible conductivity in the 'off' or superconducting state. Table 8.1 summarises the measurements of March and Symko (2) for lead, tin and zinc heat-switches.

Table 8.1

Metal	Critical field (T)	Thermal conductivity in the normal state (Watt m ⁻¹ K ⁻¹)	Thermal conductivity in the super- conducting state (Watt m ⁻¹ K ⁻¹)
Lead	0.08	310 T	4.4 T ³
Tin	0.03	500 T	5.0 T ³
Zinc	0.005	400 T	1.1 T ³

Table 8.1 shows that of the three elements considered, zinc has the lowest conductivity in the superconducting state and good conductivity in the normal state. However, it has a low critical field which could cause problems due to stray field from the main magnetiser magnet (SS1) of the nuclear stage. Tin, on the other hand, has a reasonable critical field but rather a large thermal conductivity in the 'off' state. Tin is however readily available in wire form with high (5N) purity. All heat switches considered in this chapter were constructed from twenty to thirty 0.5 mm 5N purity tin wires. They were twisted together and soldered with indium to a copper stud. Care was taken to try to prevent formation of indium-tin eutectic which has poor thermal conductivity. Nevertheless, it was found that some of the heat switches made in this way had poor thermal conductivity in the normal state which may have been due to formation of the indium-tin eutectic. It is also possible that the strains produced by twisting the wires reduced their thermal conductivity. The heat-switch was about 2 cm long. A small superconducting magnet (section 1.5.3) provided the necessary field to drive the tin heat-switch normal. About 1.2 A was passed through the magnet using a small power

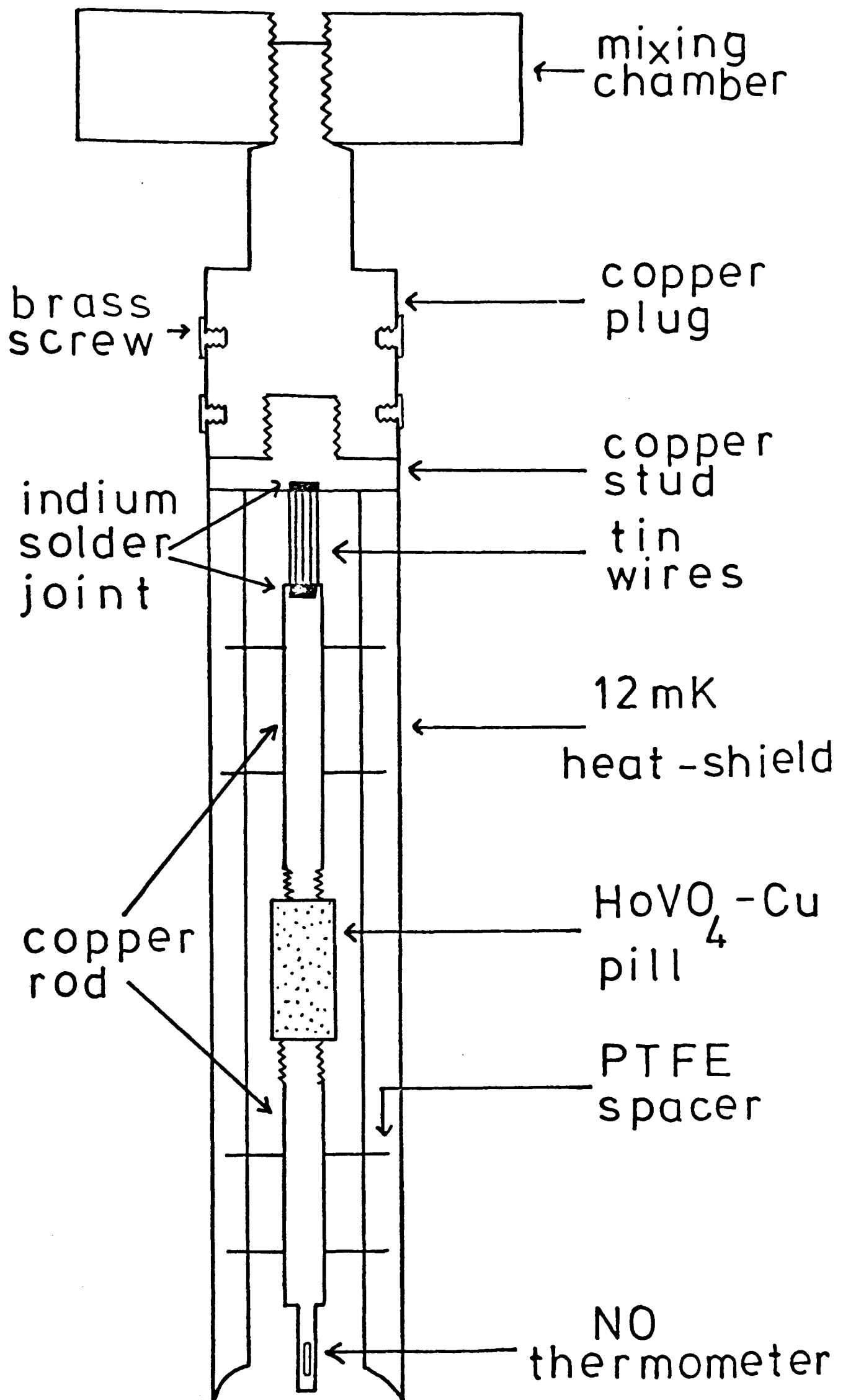


Fig. 8.2 Layout of the HcVO₄-copper nuclear refrigeration stage



Plate 2 The HoVO_4 -Cu nuclear refrigeration stage



supply and sweep unit. Typically the current sweep time was five minutes.

8.3.3 The HoVO_4 -Copper Nuclear Refrigeration Stage

Fig. 8.2 shows the layout of the nuclear refrigeration stage. A copper rod, annealed at 600°C to improve low temperature thermal conductivity, was screwed to each of the HoVO_4 pill contact screws (c.f. 8.3.1). The heat-switch was soldered between one of these rods and a copper stud which was thermally and mechanically anchored to the mixing chamber of the dilution refrigerator through screwed couplings to an intermediate copper plug. The NO thermometers were soldered using indium to the lower copper rod, which was filed down to give a flat surface. A 14 mK heat shield was constructed from a brass tube of Rollet size 8. To reduce possible eddy current heating in the heat shield, the tube was slotted to give three segments which were then copper-plated to a thickness of order $50\ \mu\text{m}$ for good thermal conductivity. The heat-switch was mechanically and thermally anchored to the copper plug using brass screws. The nuclear stage was mechanically supported by means of four PTFE spacers $1/2\ \text{mm}$ thick which had holes for the copper rods. Some of the experiments to be discussed involved electrical heating of the lower copper rod. For this purpose, $312\ (\pm 1\%) \Omega$ of 47 SWG insulated Constantan wire was non-inductively wound round the lower copper rod and fixed in position using GE-7031 varnish. The wire leads were thermally anchored all along the 14 mK shield to reduce heat leak into the pill assembly. The total heat leak into the pill assembly through the leads was estimated to be $0.003\ \text{nW}$, and therefore quite

negligible. The nuclear stage tended to vibrate with the refrigerator unit. Despite this, total heat input with the superconducting switch 'open' into the nuclear stage was only about 5 nW.

8.4 Nuclear Orientation Thermometry for the Nuclear Stage

Several factors have to be considered in the choice of an NO thermometer for a nuclear demagnetisation stage. They are the following:

(1) Heat Capacity

The host metal of the NO thermometer should have as small a heat capacity as possible, otherwise a large proportion of the cooling power of the nuclear refrigerant will be wasted on cooling the thermometer. Both lattice and electronic heat capacities are negligible at such low temperatures. However nuclear heat capacities can be quite substantial, for example h.c.p. single crystal cobalt has a heat capacity of $8.3 \text{ J K}^{-1} \text{ mole}^{-1}$ at 10 mK, a substantial heat load on all but the most powerful nuclear refrigerant systems. A typical ^{60}Co Co thermometer (3×10^{-4} moles) has a heat capacity of $2 \mu\text{J mK}^{-1}$ at 10 mK and $1.5 \mu\text{J mK}^{-1}$ at 5 mK. This must be compared with $8 \mu\text{J mK}^{-1}$ and $32 \mu\text{J mK}^{-1}$ at 10 and 5 mK respectively for a typical (10^{-2} moles) HoVO_4 -copper pill used in these experiments. The total heat load due to the cobalt thermometer is calculated by integrating the heat capacity between the two temperatures T_i and T_f (see section 8.6.3(a)). The above figures indicate that the cobalt thermometer could be a substantial heat load. On the other hand, ^{54}Mn and ^{60}Co impurities in nickel and iron have negligible heat capacities at mK temperatures.

(ii) Spin-Lattice Relaxation Times

The radioactive species must have a fast relaxation time so that it responds quickly to changes in temperature of the 'cold finger' to which the thermometer is attached. For dilute impurities in ferromagnets, the spin-lattice relaxation time T_1 is given by the Korringa relation

$$\frac{1}{T_1} = \frac{\hbar \omega_0}{2kC_K} \coth \frac{\hbar \omega_0}{2kT_L} \quad 8.4$$

where C_K is the Korringa constant for the impurity, $\hbar \omega_0$ the nuclear Zeeman splitting and T_L the lattice temperature. Typically T_1 varies from 0.1-100 sec at 10 mK for NO thermometers. It should be mentioned that the thermal relaxation time for a NO thermometer also depends on its heat capacity and on how well it is thermally linked to the refrigerant. For the refrigeration experiments of this chapter, relaxation was limited by thermal resistance between thermometer and pill (see below). The characteristic thermal relaxation time for the thermometer, τ_K , is related to its heat capacity C , Kapitza boundary resistance R_K and area of contact with the nuclear refrigerant A by the equation (Equation 5.13).

$$\tau_K = \frac{C R_K}{A} \quad 8.5$$

For the ^{60}Co Co thermometer used in these experiments:

$$C \sim 1.5 \times 10^{-3} \text{ J K}^{-1}$$

$$R_K \sim 15 \text{ T}^{-3} \text{ K s cm}^2 \text{ J}^{-1}$$

Taking the effective surface area of contact A as 30 cm^2 (section 8.6.4)

$$\gamma_K \sim 6000 \text{ s}$$

at 5 mK. The above estimate is not incompatible with the nuclear refrigeration measurements of this chapter.

(iii) Magnetic Properties of the Host Metal

Many NO thermometers use ferromagnetic hosts. These are often polycrystalline foils and need an applied field of 0.1-1 T to saturate the magnetic domains. Thermal contact to the nuclear stage was by means of copper rods which have a nuclear heat capacity proportional to B^2 . Therefore it is an advantage to use NO thermometers which do not require an applied magnetic field. Polycrystalline iron foils are magnetically soft and about 0.2 T is enough to saturate them. On the other hand, nickel is magnetically hard and about 1 T is required to saturate nickel foils. In this respect, ^{60}Co in single crystal h.c.p. cobalt is useful in that no applied field is necessary to orient the domains. The domains naturally align themselves parallel to the crystal c-axis.

(iv) Thermometer Sensitivity

The sensitivity of a NO thermometer is a function of temperature, nuclear Zeeman splitting and spin. In this respect the Kondo system ^{54}Mn Cu is ideal, as the magnetic hyperfine field can be altered at will by changing the applied field (8, 9).

(v) Radioactive Heating

^{54}Mn and ^{60}Co are the most commonly used radioactive impurities. ^{54}Mn decays by electron capture and therefore has a low radioactive heating of about 35 pW per μCi (10). On the other hand, ^{60}Co decays by β^- emission and has the

relatively large heating of 750 pW per μCi . This makes ^{54}Mn sources superior to ^{60}Co in cases where low heat input is important (e.g. nuclear refrigeration experiments).

(vi) Summary

On balance, $^{54}\text{Mn-Cu}$ is the most suitable NO thermometer for nuclear refrigeration experiments. Unfortunately, the $^{54}\text{Mn Cu}$ thermometer used in these experiments had an estimated 200 ppm iron impurity but once calibrated could be used down to about 4 mK (section 8.6.1). ^{60}Co in single crystal h.c.p. cobalt has useful sensitivity at 4 mK but has a relatively large nuclear heat capacity at mK temperatures which is a disadvantage. $^{60}\text{Co Fe}$ while useful at 4 mK requires an applied field of about 0.8 T to align the domains.

8.5 Nuclear Refrigeration Experiments

A typical nuclear refrigeration experiment consisted of the following:

- (a) The heat-switch magnet was energised to about 1.2 A in a few minutes. This drove the superconducting tin wires normal and allowed heat from the pill to be conducted into the mixing chamber.
- (b) The pill was magnetised by passing a current through the main magnetiser coils, SS1. Typically the field was left on for four hours to allow the pill to cool to between 15 and 20 mK. The actual time taken depended on the thermal conductivity of the tin wires.
- (c) The polariser magnet was swept to the appropriate current.
- (d) The current in the heat-switch magnet was reduced to zero in two or three minutes when the pill was ready for demagnetisation.

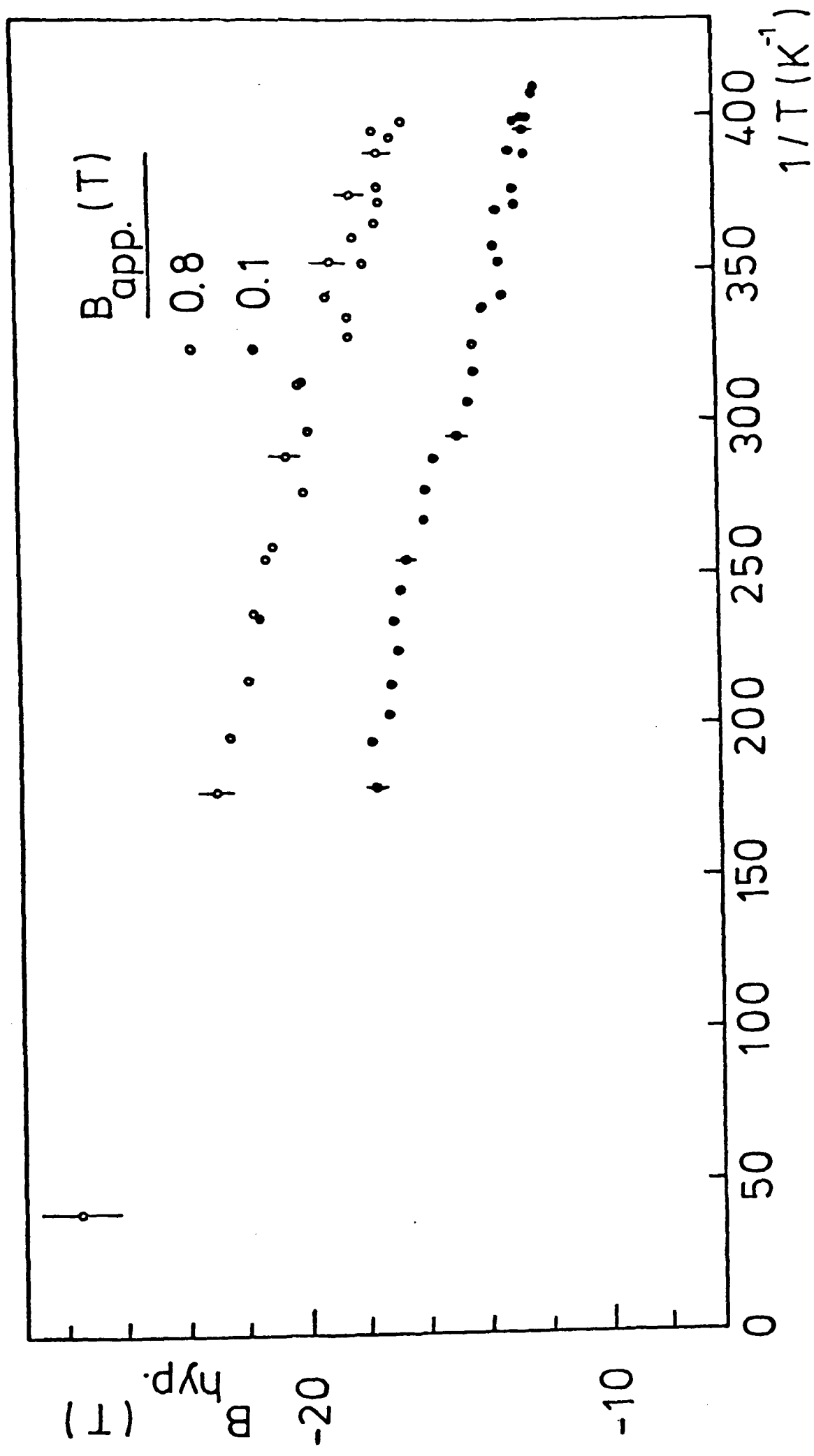


Fig. 8.3 Magnetic hyperfine field of the $^{54}\text{Mn Cu NO}$ thermometer vs reciprocal temperature

(e) The pill was demagnetised to zero field at a controlled rate. Several linear sweep rates were available, the slowest being 0.1 A min^{-1} . A typical slow demagnetisation was the following:

- (i) Down to 16 A (0.88 T) at 1.5 A min^{-1}
- (ii) 16 A - 8 A (0.44 T) at 0.75 A min^{-1}
- (iii) 8 A - 5 A (0.28 T) at 0.3 A min^{-1}
- (iv) 5 A - 0 A at 0.1 A min^{-1} .

A typical fast demagnetisation involved (i) and (ii) and then down to zero current at rate(ii).

8.6 Nuclear Refrigeration with HoVO_4 -Copper Pills

8.6.1 Experiment 1

This was the first of the nuclear refrigeration experiments on compressed HoVO_4 -copper pills and also showed the best performance. The pill had a mass of 8.69 g corresponding to an estimated HoVO_4 content of 2.6 g (0.01 mole). The NO thermometers $^{54}\text{Mn Cu}$ and $^{60}\text{Co Fe}$ were soldered with indium to the copper 'cold finger', thermally attached to the compressed pill. As explained in section 8.4, the $^{54}\text{Mn Cu}$ thermometer had an iron content of 200 ppm, which caused the hyperfine field to be temperature dependent. The hyperfine field was calibrated against a $^{60}\text{Co Co}$ thermometer by D.W. Murray and the results are plotted in Fig. 8.3 (section 1.4.3). Compton and Williams (8, 9) have shown that the hyperfine field B_{hyp} at constant applied field can be fitted to the expression

$$B_{\text{hyp}} = A_0 + \frac{A_1}{T} \quad 8.6$$

where A_0 and A_1 are constants. Such fits to the data (Fig.

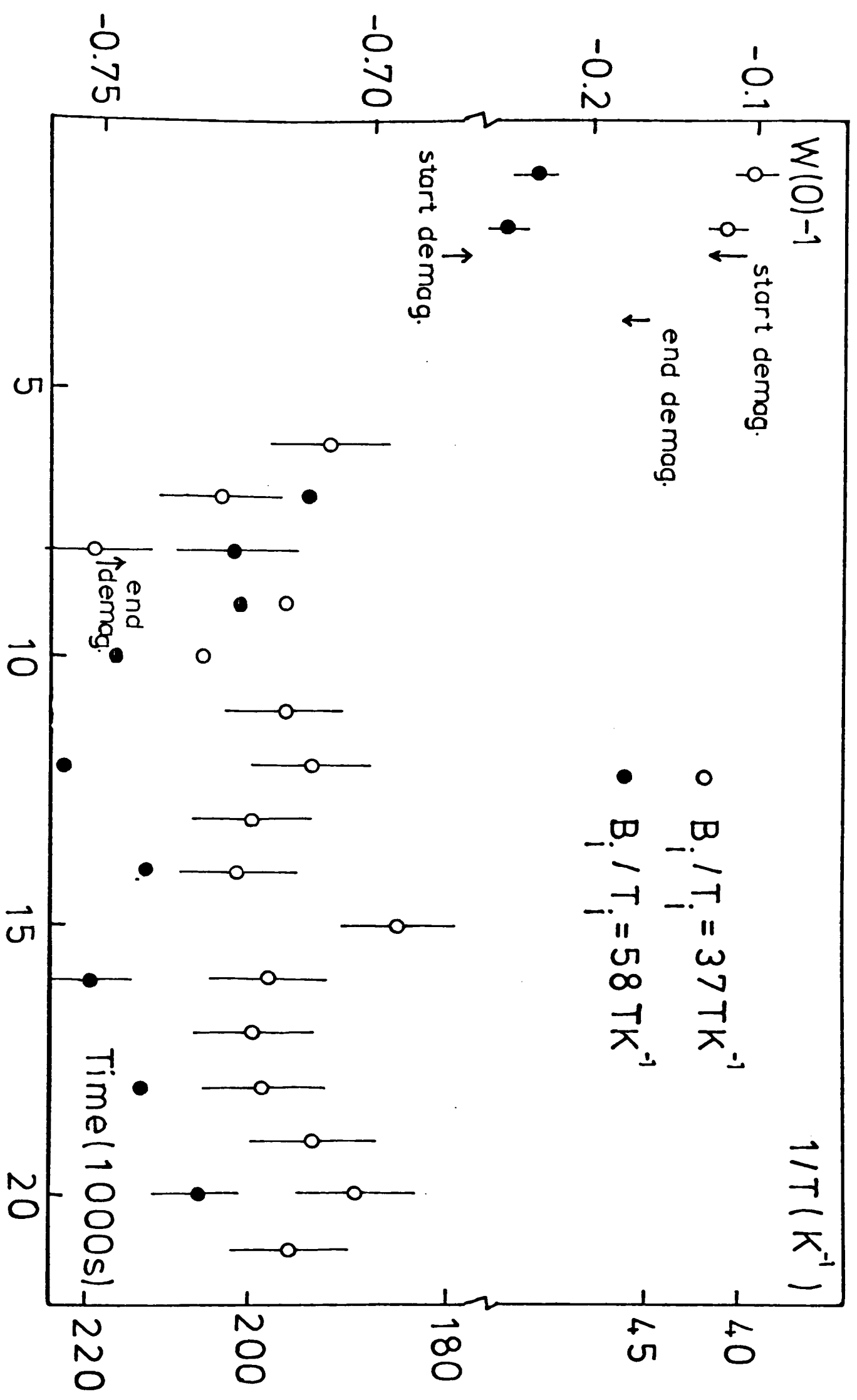


Fig. 8.4 Nuclear refrigeration of HoVO_4 -copper pill-Experiment 1 part (a)

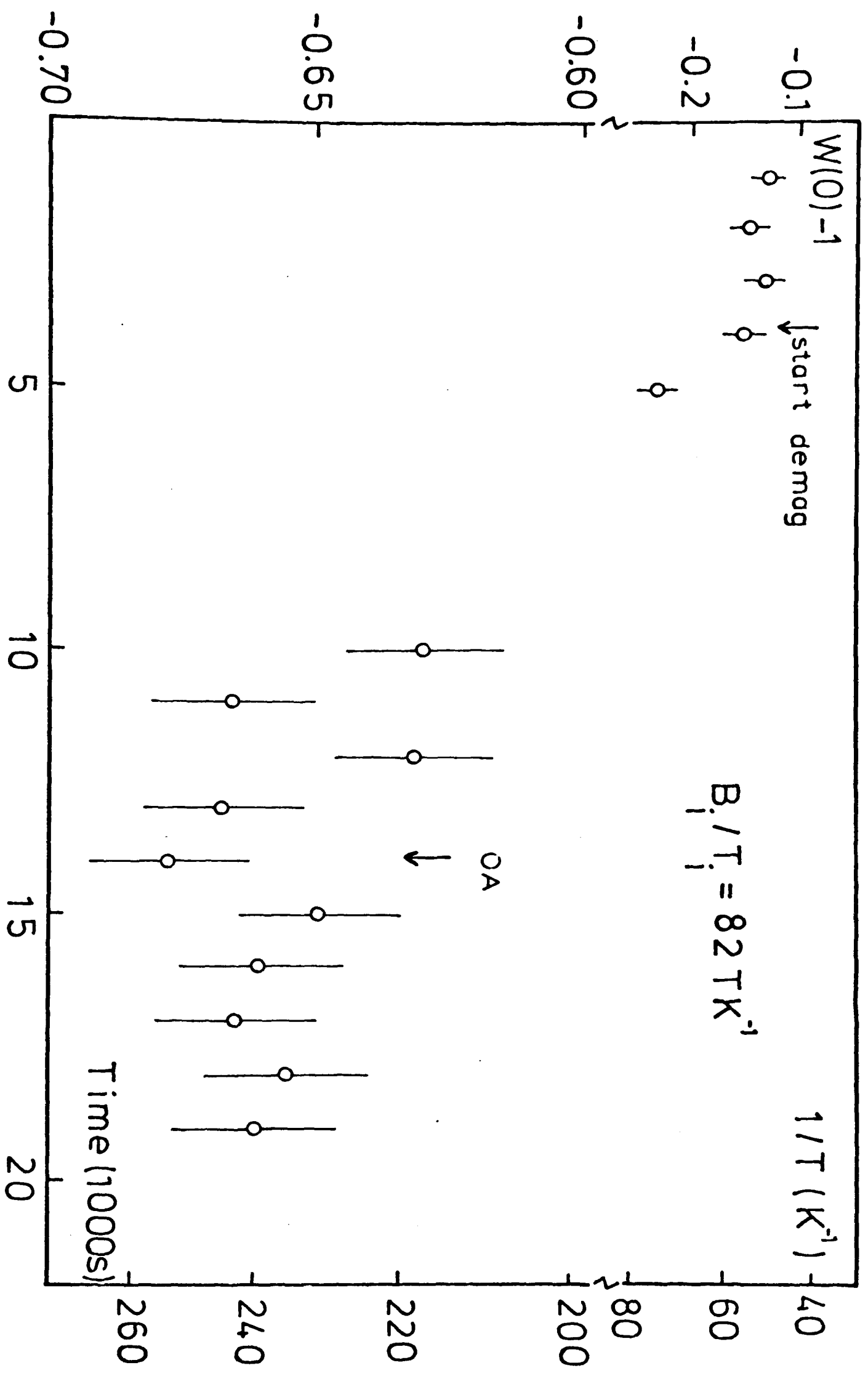


Fig. 8.5 Nuclear refrigeration of HoVO_4 -copper pill-Experiment 1 part (b)

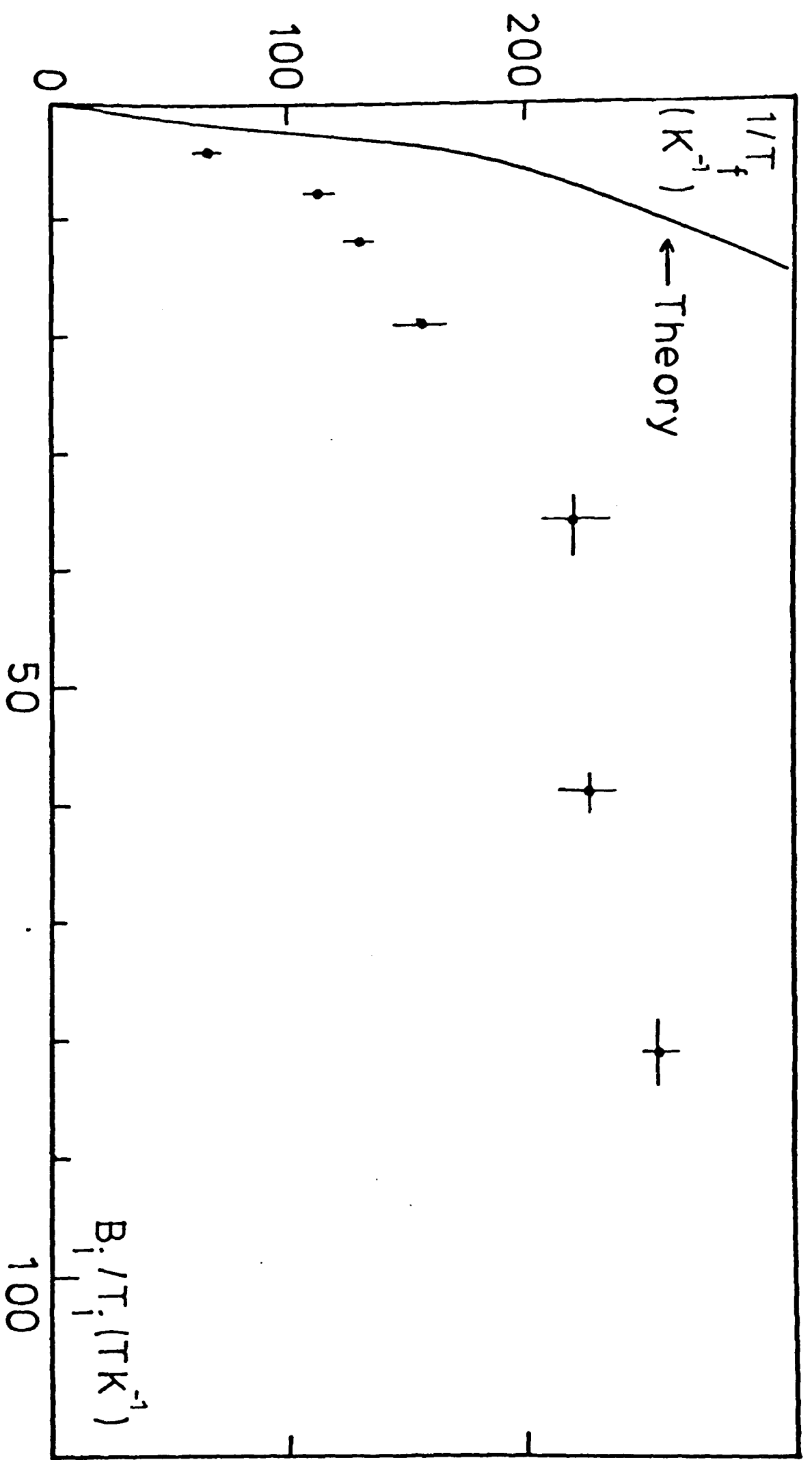


Fig. 8.6 Reciprocal temperature after demagnetisation vs B_i/T_i

8.3) were used to deduce temperatures down to 3.9 mK. The ^{60}Co Fe thermometer was badly shaped and was unsaturated even in 0.8 T. Thus the ^{54}Mn Cu thermometer had to be used throughout to determine temperatures. Adiabatic demagnetisations were made from several values of B_i/T_i down to $B_f = 2 \times 10^{-3}$ T. Different sweep rates and polariser fields were tried to investigate optimum conditions for demagnetisation of the pill. The details are summarised in Table 8.2. The lowest temperature reached was 3.9 (3) mK and as expected this was for the demagnetisation with the largest value of B_i/T_i and smallest polariser field (see below). The best three Runs are plotted in Figs. 8.4 and 8.5. The results for the demagnetisations are summarised in Fig. 8.6 where $1/T_f$ is plotted against B_i/T_i . The solid line in the figure represents an idealised calculation for perfect isentropic demagnetisation of a randomly oriented polycrystalline sample of HoVO_4 . The calculation assumed that the crystallites were in thermal equilibrium at all times during demagnetisation and no account was taken of extraneous heat capacities and residual heat leaks. Not surprisingly, the measured values of T_f were well above those calculated. The following is a discussion of the possible reasons:

(a) Apart from the pill, the bulk of the heat capacity of the nuclear stage was the nuclear heat capacity of the cold finger. At 5 mK this is estimated to be 15% of that of the pill for a polariser field of 0.8 T, reducing to less than 1% for 0.1 T. Indium has a quadrupole heat capacity of $\sim 1/T^2$ J K $^{-1}$ mole $^{-1}$ (11). The NO thermometers were soldered using about 100 mg of indium which had an estimated heat capacity of only about $0.003 \mu\text{J mK}^{-1}$ at 5 mK. On the other hand, the

Table 8.2

Refrigeration of HoVO₄-copper compressed pill

B_i (T)	T_i (mK)	B_i/T_i (T K ⁻¹)	T_f (mK)	Polariser field (T)	Demagnetisation rate
1.62	19.8	82	3.9(3)	0.1	slow
1.01	27.2	37	4.5(2)	0.8	fast
1.01	17.4	58	4.4(2)	0.8	slow
0.21	18.0	11.7	7.7(3)	0.8	fast
0.14	18.3	7.7	8.8(4)	0.8	fast
0.40	21.7	18.6	6.3(3)	0.8	fast
0.14	32	4.2	15.0(7)	0.8	fast

slow $\equiv$ down to 10A at 0.75A min⁻¹, 10A-5A at 0.3A min⁻¹,
5A-0A at 0.1A min⁻¹.

fast $\equiv$ at a constant 0.75A min⁻¹.

$$(1 \text{ A} \quad \equiv \quad 0.055\text{T})$$

heat capacity of the pill is calculated to be about $30 \mu\text{J mK}^{-1}$ at 5 mK. Thus extraneous heat capacities are not expected to drastically reduce the efficiency of the pill.

(b) Our measurements indicated that there was a residual heat leak into the pill of order 5 nW (section 8.6.4). Assuming that the total heat leak into the pill is equivalent to 5 nW for one hour at a pill temperature of 4 mK and using the equation

$$\delta S = \frac{\delta Q}{T} \quad 8.7$$

the increase in entropy of the salt pill due to the heat leak is estimated to be

$$\delta S = 0.004 \text{ J K}^{-1}$$

Entropy calculations for the pill (0.01 mole HoVO_4) at 4 mK give

$$S = 0.1 \text{ J K}^{-1}$$

Finally, from the polycrystalline entropy curves, it can be shown that the above entropy increase would produce warming up of the pill of about 0.05 mK, which is quite small.

(c) Eddy current heating in the pill during a typical demagnetisation is estimated to be $0.1\text{--}1 \mu\text{J}$ for electrical resistivity in the range $\rho = 1.6 \times 10^{-9} \Omega \text{ m}^{-1}$ – $1.6 \times 10^{-9} \Omega \text{ m}^{-1}$ (for a typical solid copper compressed pill $\rho \sim 0.5 \times 10^{-8} \Omega \text{ m}^{-1}$ at 4 K). This is quite negligible when compared with the enthalpy of the pill.

(d) Fig. 8.4 shows that the ^{54}Mn Cu thermometer reached its lowest temperature about an hour after the end of the demagnetisation for both slow and fast rates of demagnetisation.

This time constant depends not only on the internal equilibrium time for the pill but also on the thermal relaxation time of the cold finger and NO thermometers attached to it.

These time constants, given by Equation 8.5, reflect the poor thermal contact within and out of the pill and are expected to diminish the efficiency of the pill.

(e) Owing to the boundary resistance between the pill and the cold finger, residual heat input into the cold finger due to radioactive heating from the NO thermometers, mechanical vibrations etc. produced a temperature gradient between the pill and thermometer. This is thought to be the dominant limitation to low temperatures obtainable using a nuclear refrigeration system of the type described here. Experiments on a pill similar to the one described here indicated that the effective surface area between the HoVO_4 crystallites and the copper matrix was only of order 30 cm^2 (c.f. section 8.6.4). Using the heat flow equation for a Kapitza boundary (Equation 8.2), an effective surface area $A = 30 \text{ cm}^2$ gives a temperature drop across the interface of

$$4 \text{ mK per nW of heat input at } 4 \text{ mK}$$

This gives a typical temperature drop across the salt-copper interface of about 4 mK for a typical radioactive heat leak from a ^{60}Co Co thermometer of 1 nW into the cold finger.

These estimates indicate that the low temperature performance of the HoVO_4 -copper pill can be explained in terms of a relatively small surface area of contact between the HoVO_4 crystallites and copper matrix.

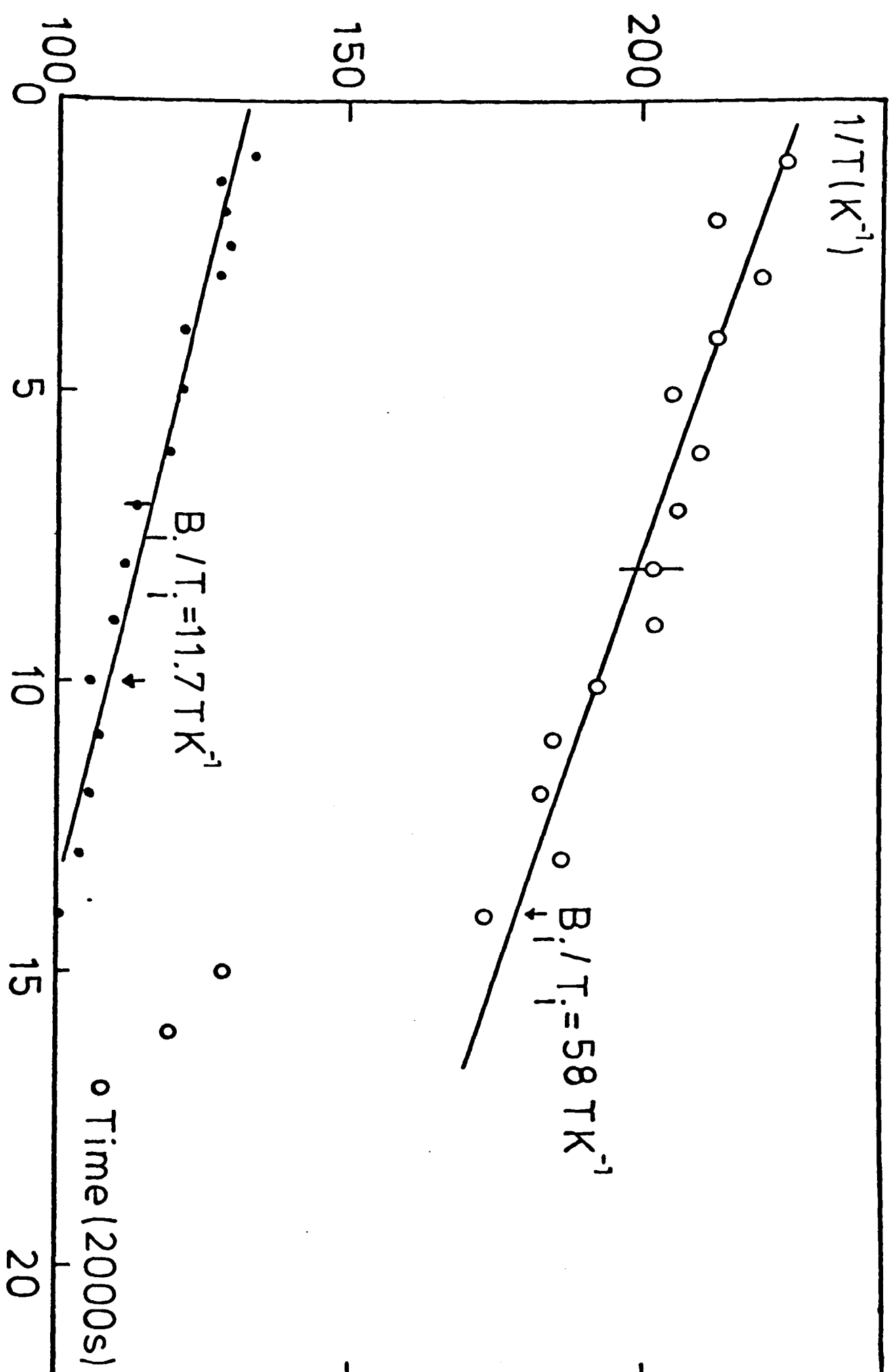


Fig. 8.7 Reciprocal temperature vs time after demagnetisation

8.6.2 Warm-Up Rate

The heat leak into the pill produced a temperature rise, given by the equation

$$\dot{Q}dt = mC_B dT \quad 8.8$$

where m , C_B are respectively the number of moles and molar heat capacity of HoVO_4 . A calculation showed that the heat capacity of HoVO_4 has an anomaly close to the Néel temperature (~ 4 mK), which cannot be expressed by a simple algebraic expression. However, at high temperatures ($T \gg T_N$) the specific heat capacity is given by the expression

$$C_B = \frac{\alpha_o}{T^2} \quad 8.9$$

where $\alpha_o = 2.7 \times 10^{-4} \text{ J mole}^{-1} \text{ K}$ for HoVO_4 . Substituting Equation 8.9 into Equation 8.8

$$\dot{Q}dt = \frac{A}{T^2} dT \quad 8.10 \quad \text{where } A = \alpha_o m$$

Taking $m = 0.01$, $A = 2.7 \times 10^{-4} \text{ J K}$ for the HoVO_4 -Cu pill, and integrating Equation 8.10 gives

$$\dot{Q} t = -A \left(\frac{1}{T_1} - \frac{1}{T_2} \right) \quad 8.11$$

Plotting $1/T$ against t would give a straight line of slope $-\dot{Q}/A$. From the slope $\dot{Q}$ can be determined assuming that A is known. Two such plots are shown in Fig. 8.7 in the temperature region $1/T = 100\text{--}220 \text{ K}^{-1}$. Straight lines were fitted and the results are summarised in Table 8.3.

Table 8.3

Warm-up rate for HoVO₄-copper pill

$1/T_i$ (K ⁻¹)	$1/T_f$ (K ⁻¹)	T_N (mK)	Nuclear refrigerant	Warm-up rate (K ⁻¹ Hr ⁻¹)	Estimated heat-leak (nW)
130	100	4.0(2)	HoVO ₄ -Cu	-4.3(4)	5
220	170			-6.1(6)	
125	225	0.6	PrNi ₅	- 80	4

The heat leaks were computed from the relation

$$\text{slope} = \frac{-\dot{Q}}{\dot{A}} \quad 8.12$$

For comparison, also shown in Table 8.3 are results for a small PrNi₅ nuclear refrigeration stage (~ 0.02 mole) built in the laboratory (12). It can be seen that although the heat leaks for the two systems were similar, the warm-up rate for the HoVO₄-copper pill was smaller, due to the larger heat capacity. Thus HoVO₄ is a useful nuclear refrigerant in the temperature range $1/T = 100\text{-}220 \text{ K}^{-1}$. It is worth pointing out that the straight lines obtained in Fig. 8.7 indicate that to a good approximation the specific heat capacity of HoVO₄ is given by Equation 8.9. This is in direct contradiction with a calculation (see Fig. 2.5), which showed a sharp specific heat anomaly at about 4.8 mK, the Néel temperature. While the calculation was made in the Mean Field approximation for a single crystal of HoVO₄, it should also be valid for a polycrystalline sample. Thus these measurements push the specific heat anomaly below about 4.2 mK.

8.6.3 Experiment 2

In this experiment, a ⁶⁰Co Co crystal was soldered to the

cold finger with indium and used to measure temperatures. This was thought necessary at the time because of uncertainty about the low temperature behaviour of the ^{54}Mn Cu thermometer used in Experiment 1. In all other respects, this experiment was identical to Experiment 1. The details are summarised in Table 8.4.

Table 8.4

Results of refrigeration Experiment 2

B_i (T)	T_i (mK)	B_i/T_i (T K $^{-1}$)	T_f (mK)	Polariser field (T)	Demagnetisation rate
2.4	16.9(6)	143(5)	4.7(1)	0.1	45A-8A 1.5A min $^{-1}$ 8A-0A 0.75A min $^{-1}$
1.6	17.0(6)	92(3)	4.5(1)	0.1	30A-10A 0.75A min $^{-1}$ 10A- 5A 0.3A min $^{-1}$ 5A- 0A 0.1A min $^{-1}$
2.4	16.7(6)	144(5)	4.6(1)	0.1	45A-30A 1.5A min $^{-1}$ 30A- 0A 0.75A min $^{-1}$
1.1	16.9(6)	64(3)	4.6(1)	0.1	20A-7.5A 0.75A min $^{-1}$ 7.5A-5A 0.3A min $^{-1}$ 5A -0A 0.1A min $^{-1}$

$$(1\text{A} \equiv 0.055 \text{ T})$$

Table 8.4 shows that the lowest temperature reached was 4.5(1) mK which is slightly higher than the 3.9 mK reached in Experiment 1. The reasons may be the following:

(a) As discussed in section 8.4, cobalt has a substantial nuclear heat capacity which peaks at about 10 mK. The heat capacity of the cobalt thermometer used is estimated to be $2\mu\text{J mK}^{-1}$ and $1.5\mu\text{J mK}^{-1}$ at 10 and 5 mK respectively. The lowest temperature was recorded about half an hour after the end of the demagnetisation. The enthalpy of the cobalt thermometer between 4 and 10 mK is of order $10\mu\text{J}$. If it is

assumed that the cobalt thermometer cooled between 4 and 10 mK by exchange of heat with the nuclear refrigerant, the pill would warm up by about 0.3 mK. This is within a factor of two of the observed discrepancy between Experiments 1 and 2.

(b) There may have been some deterioration in contact properties of the pill due to oxidation of the copper and the effects of thermal cycling. This would explain the reduced performance of Experiment 2. It is worth noting that T_f is insensitive to B_i/T_i and demagnetisation rates for the large values B_i/T_i used in these experiments. This is consistent with Experiment 1. This indicates that irreversible processes dominate within the pill.

8.6.4 Thermal Contact in HoVO₄-Copper Pills

In principle it is possible to measure thermal resistance between the pill and cold finger by applying known amounts of electrical power to a heater located on the cold finger (section 8.3.3) and measuring the change in temperature. In practice, the temperature of the pill is not constant during the heating period owing to its finite heat capacity and the time taken for equilibrium to be established is of order hours.

A number of experiments were made with ^{60}Co Co as thermometer. In the first experiment, with the heater assembled, the lowest temperature recorded was a disappointing 7.3 mK. Passing a current through the heater warmed up the pill disastrously and no thermal resistance measurements could be made. Examination of the pill assembly after the run showed that the poor result was probably due to poor thermal contact between the pill and cold finger at the screw joint.

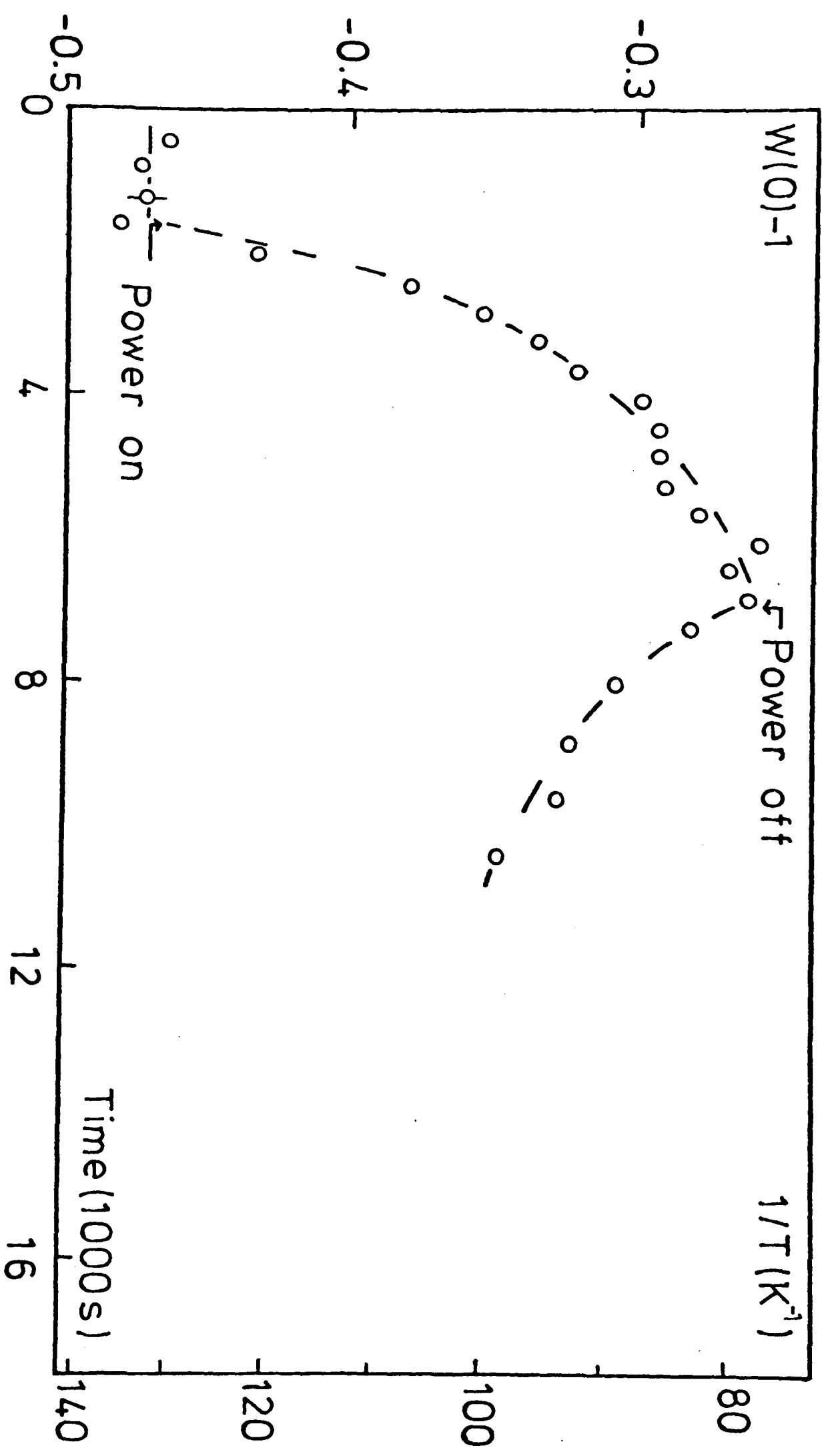


Fig. 8.8 Measurement of effective surface area of contact of HoVO_4 -copper pill

The screw thread was damaged while efforts were made to improve contact. A second pill was constructed, 0.75 cm diameter, 2.1 cm long and mass = 6.8 g. It had an estimated HoVO_4 content of 2 g corresponding to about 0.007 mole. This was about 25% less than the HoVO_4 content of the first pill. The pill was screwed into the copper cold finger and then soldered with indium to improve thermal contact.

The performance of the new pill was poor. The best results are presented in Table 8.5, where each of the values represents the best of several demagnetisation Runs. The poor performance of the pill may be due to slow oxidation of the copper powder, despite being kept under vacuum.

Table 8.5

Refrigeration experiments on pill 2

B_i (T)	T_i (mK)	B_i/T_i (T K ⁻¹)	T_f (mK)
0.8	26.3(7)	32	7.7(1)
1.6	17.7(4)	92	6.8(1)

The poor thermal contact and long relaxation time constants are illustrated in Fig. 8.8. 4.6 nW of power was applied to the heater and the temperature of the cobalt thermometer monitored every 400 seconds. Unfortunately, the time constants involved were long and equilibrium was not established. The cold finger warmed up from 8 to 12 mK with a time constant of order an hour. On switching the power off, the thermometer cooled down immediately, showing that the pill had not warmed right up. Using Equation 8.2 for heat flow across a Kapitza boundary, with $\dot{Q} = 4.6$ nW, $T = 5$ mK and $\delta T \sim 4$ mK, the effective contact area is estimated to be 30 cm², which is a

small fraction of the total available surface area (section 8.2). This rough estimate indicates that the poor performance of the pill can be explained in terms of poor thermal contact between the crystals and the copper matrix. It can be inferred from the above analysis that the first pill worked better because of a larger surface area of contact, as the heat leaks are unlikely to be very different in the two cases.

Due to poor thermal contact between the pill and copper cold finger, heat capacity measurements of the pill could not be made with any degree of reliability.

8.7 The HoVO₄-Gold Pill

The deterioration of the performance of the HoVO₄-copper pills could be explained by oxidation of the fine copper powder. On the other hand, gold powder is highly stable against oxidation and can be obtained in fine powder form. A HoVO₄-gold pill was therefore constructed from 99.95% 0.5-1 μ m gold dust (supplied by Johnson-Matthey Chemicals) and HoVO₄ powder, using the cold press technique outlined in section 8.3.1. A gold-plated 99.9% purity copper foil was inserted into the HoVO₄-gold mixture before pressing, for thermal contact to the pill. The foil was 0.5 cm wide and 0.1 mm thick. Before gold-plating the foil was heat-treated (see below). The pill was roughly cylindrical in shape and had a mass of 6.0 g and filling factor of 0.8. Its estimated HoVO₄ content was 1.0 g. Thermal contact to the pill was by means of 0.1 mm thick and 0.5 cm wide copper foils. Two such foils were annealed at 900°C with a continuous air leak of 2×10^{-4} torr pressure going over it. This served to oxidise

the iron impurities in the foil and thus improve its conductivity at low temperature by about a factor of 10 (13). Typically the copper foils had an electrical conductivity at 4 K about 200 times that at room temperature before heat treatment and 2000 times after heat treatment. Copper foils were used in this experiment to reduce extraneous heat capacities to a minimum. The heat-switch consisted of 12 strands of 5N 0.5 mm diameter tin wires soldered to the copper foils with indium.

The lowest temperature reached with the HoVO_4 -gold pill was 6.0(1) mK as measured by a ^{60}Co Co thermometer. The demagnetisation results are summarised in Table 8.6. The integrated heat leak in Table 8.6 is for a heat leak of 5 nW. Although the ratio B_i/T_i were similar in all three experiments, the lowest temperature reached was dependent on the rate of demagnetisation.

Table 8.6

Results for demagnetisations of the HoVO_4 -
gold pill

B_i (T)	T_i (mK)	B_i/T_i (T K ⁻¹)	T_f (mK)	Demagnetisation rate (A min ⁻¹)		Estimated heat leak during demagnetisation (μJ)
1.66	17.3	96	6.2(5)	30A-8A	1.5	0.2
				8A-3A	0.75	
				3A-0A	0.3	
1.66	18.6	89	6.0(5)	30A-10A	0.75	0.5
				10A- 5A	0.3	
				5A- 0A	0.1	
1.66	19.2	86	8.4(2)	30A-15A	0.3	1
				15A- 0A	0.1	

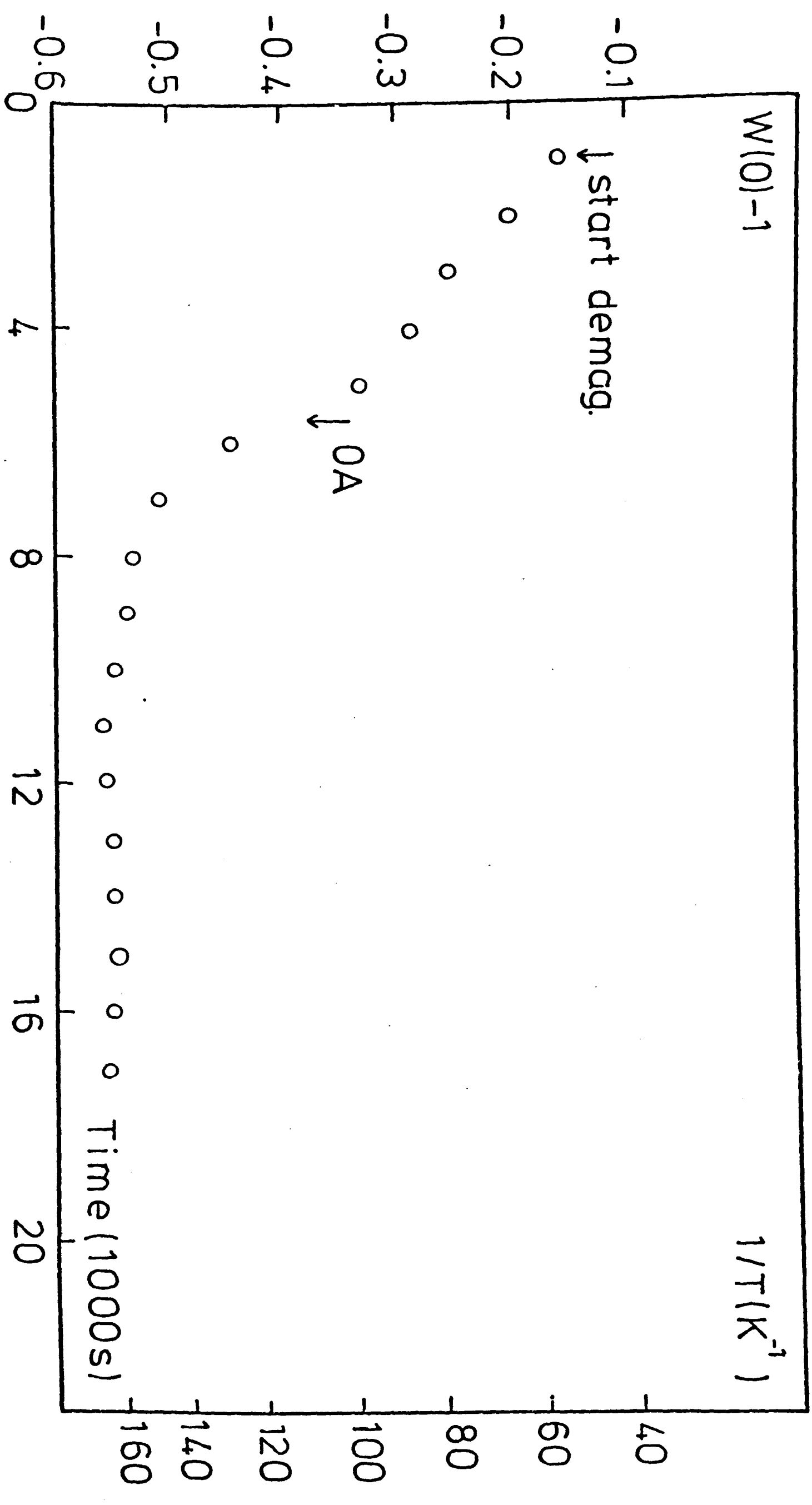


Fig. 8.9 Nuclear refrigeration of HeVO_4 -gold pill

The estimated heat leaks are too small to explain the large discrepancy between the values of T_f for the second and third demagnetisations. The pill had an estimated heat capacity of about $10 \mu\text{J mK}^{-1}$ at 5 mK and thus no appreciable heating of the pill due to heat leaks during demagnetisation is expected. It is possible that the tin heat-switch was faulty and had appreciable thermal conduction in the 'off' (superconducting) state. Fig. 8.9 shows that the temperature of the cobalt thermometer reached its lowest value some 1/2-1 hour after the end of the demagnetisation. This was typical for the HoVO_4 -copper pills too. It should be pointed out that the HoVO_4 -gold pill had about 1/3 of the HoVO_4 content of the HoVO_4 -copper pills. This would definitely make the gold pill more sensitive than the copper pill to heat leaks.

8.8 Conclusions

A relatively simple nuclear refrigeration system has been described which used the enhanced nuclear paramagnet HoVO_4 . The lowest temperature reached was 3.9(3) mK using a compressed HoVO_4 -copper pill. The pill had a typical warm-up rate of $5 (\text{K Hr})^{-1}$ in the temperature range 4-10 mK. This warm-up rate is slow compared with a Pr Ni_5 nuclear stage of about the same molar content. This makes HoVO_4 -copper pills useful in the above temperature range. Preliminary experiments on a HoVO_4 -gold pill allowed thermometers to be cooled to 6.0(1) mK.

The problem of thermal contact between the insulating HoVO_4 crystallites and samples attached to the copper cold finger has not been fully solved. Thermal resistance measurements on the HoVO_4 -copper pill indicated that the effective area of contact between the pill and the cold finger was about

30 cm², which was only a small fraction of the total surface area available. This suggests that only a small fraction of the HoVO₄ crystallites were in thermal contact with the copper powder surrounding them. The HoVO₄-gold pill was only about a third of the HoVO₄-copper pill in salt content, which made the former more sensitive to stray heat leaks and heat capacities. This could explain the poor performance of the HoVO₄-gold pill.

In these experiments mixing was done by hand in an agate mortar. It may be possible to produce more uniform mixing by using a mill.

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