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2006 J. Phys.: Conf. Ser. 51 319

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High-field studies of the slow thermal death of interlayer coherence in quasi-two-dimensional metals

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Abstract. The interlayer magnetoresistance ρ_{zz} of the organic metal κ -(BEDT-TTF)₂Cu(NCS)₂ has been studied in fields B of up to 45 T and at temperatures T from 0.5 K to 50 K. The peak in ρ_{zz} seen in exactly in-plane fields, a definitive signature of interlayer coherence, remains to T 's exceeding the Anderson criterion for incoherent transport by a factor ~ 25 . Angle-dependent magnetoresistance oscillations (AMROs) due to Fermi-surface orbits are suppressed by rising T , with a T^2 dependence suggesting electron-electron scattering.

Many interesting compounds possess quasi-two-dimensional (Q2D) electronic bandstructure; examples include crystalline organic metals [1, 2, 3, 4], cuprates [5] and layered ruthenates [6]. Such systems may be described by a tight-binding Hamiltonian in which the ratio of the interlayer transfer integral t_{\perp} to the average intralayer transfer integral t_{\parallel} is $\ll 1$ [1, 4]. The question arises as to whether the interlayer charge transfer is coherent or incoherent in these materials, *i.e.* whether or not the Fermi surface is three dimensional (3D), extending in the interlayer direction. Various criteria for interlayer incoherence have been proposed, including [7]

$$k_{\text{B}}T > t_{\perp}, \quad (1)$$

where T is the temperature. In such a picture, thermal fluctuations are proposed to “wipe out” details of the interlayer periodicity [7].

Interlayer incoherence is used as a justification for a number of theories which are thought to be pivotal in the understanding of reduced-dimensionality materials (see e.g. [7, 8, 9]). It is therefore important to test assertions such as Eq. 1. To this end, we have made measurements of the magnetic-field-orientation dependence of the resistance of the crystalline organic metal κ -(BEDT-TTF)₂Cu(NCS)₂ [9] using fields of up to 45 T. This material was chosen because its Fermi surface is well known [9], previous low- T experiments have demonstrated that the interlayer transfer integral is $t_{\text{a}} = 0.065 \pm 0.07$ meV [10, 11] and standard laboratory T 's allow the inequality in Eq. 1 to be exceeded by orders of magnitude ($t_{\text{a}}/k_{\text{B}} \approx 0.5$ K).

Interlayer coherence is detected using a phenomenon known as the “coherence peak” or “SQUIT (Suppression of QUasiparticle Interlayer Transport)peak” [1, 2, 4], a maximum in the interlayer component of the magnetoresistance ρ_{zz} observed when the field \mathbf{B} lies exactly in

the intralayer plane. This occurs because of the effective interlayer velocity averaging caused by closed orbits on the side of the Fermi surface; these can exist if, *and only if*, the interlayer transport is coherent, *i.e.* the Fermi surface extends in the interlayer direction (Fig. 1(a)).

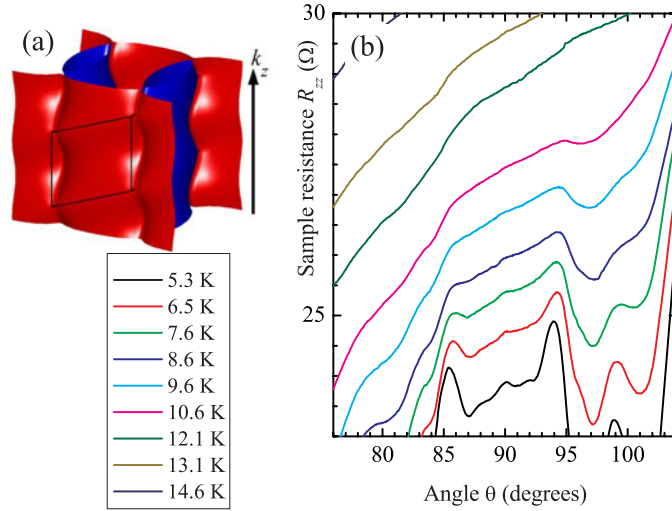


Figure 1. (a) 3D Fermi surface of κ -(BEDT-TTF)₂Cu(NCS)₂ based on low- T SQUIT data. The interlayer warping has been exaggerated for visibility [10]; the corrugations allow closed orbits to occur on the Fermi surface in an exactly in-plane field. (b) Interlayer resistance R_{zz} ($\propto \rho_{zz}$) of a κ -(BEDT-TTF)₂Cu(NCS)₂ sample as a function of field (\mathbf{B}) orientation ($\theta = 90^\circ$ is \mathbf{B} in-plane; $\theta = 0$ is \mathbf{B} normal to the planes). The rotation plane is defined by $\phi = 160^\circ$ (see [10] for details of the coordinates); here, the ρ_{zz} features are due to orbits on the Q1D Fermi-surface sections (red in Fig. 1(a)). $B = 45$ T; see inset for T values.

Figure 1(b) shows ρ_{zz} data for κ -(BEDT-TTF)₂Cu(NCS)₂ for $5.3 \text{ K} \leq T \leq 14.6 \text{ K}$ (analogous data for lower T are in Ref. [10]). The SQUIT peak is clearly visible close to $\theta = 90^\circ$; its angular width is consistent with the above-mentioned value of $t_a \approx 0.065 \text{ meV}$ [10]. In spite of the small size of t_a , the SQUIT peak, demonstrating interlayer coherence, continues to be observable up to at least 13.1 K, exceeding the criterion in Eq. 1 by a factor ~ 25 [12].

Having demonstrated that the Fermi surface of κ -(BEDT-TTF)₂Cu(NCS)₂ remains 3D up to at least $T \approx 13 \text{ K}$ [12], it is informative to look at the T -dependence of other features in ρ_{zz} , such as angle-dependent magnetoresistance oscillations (AMROs). Fig. 2(a) shows ρ_{zz} of κ -(BEDT-TTF)₂Cu(NCS)₂ over a wider angular range than Fig. 1 and for a different plane of rotation, defined by the angle ϕ [10]. For this ϕ , the AMROs and SQUIT are due chiefly to orbits on the Q2D sections of the Fermi surface (blue sections in Fig. 1(a)); the AMROs are hence “Yamaji oscillations” and can be indexed accordingly [9, 10]. Note that whilst the background magnetoresistance does not show a dramatic T -dependence, the AMROs decrease in amplitude rapidly, as shown in Fig. 2(b). By contrast, the amplitude of the SQUIT peak varies more slowly with T . Elsewhere [13] we show that this difference can be understood in terms of the nature of the orbits responsible for the different features in ρ_{zz} .

To extract quasiparticle scattering rates from AMROs, numerical calculations of ρ_{zz} are made using the Boltzmann-transport approach and model Fermi surface (Fig 1(a)) of Ref. [10]. A comparison of data and simulation is shown in Fig 3. It is found that the amplitudes of the simulated AMRO and their B -dependence can be made to fit those of the experimental data by scaling the scattering rate τ^{-1} , an input parameter of the model. Once this has been done,

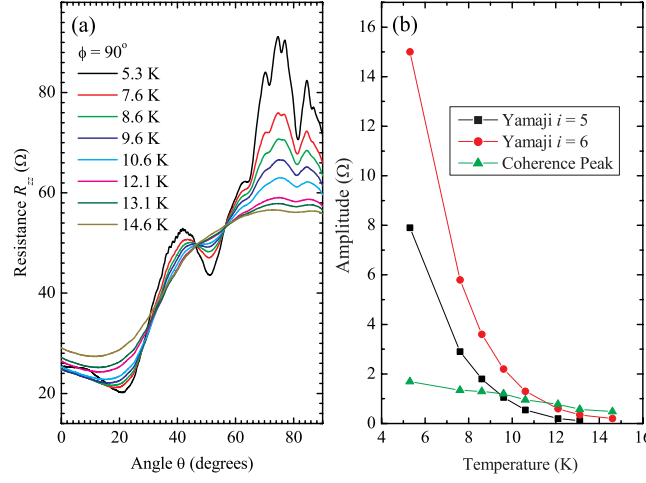


Figure 2. (a) R_{zz} for κ -(BEDT-TTF)₂Cu(NCS)₂ at several T and $B = 45$ T; $\phi = 90^\circ$ so that features in ρ_{zz} are chiefly due to the Q2D Fermi-surface sections. (b) The amplitude of various features in ρ_{zz} versus T , including the $i = 5$ and 6 Yamaji AMROs (see Ref. [10] for information on the indexing) and the SQUIT (coherence) peak.

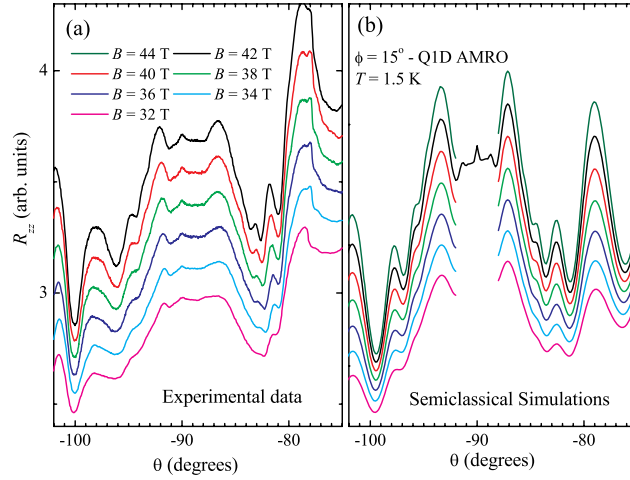


Figure 3. Comparison of experimental ρ_{zz} data (a) and a numerical simulation (b) using the model of Ref. [10]. $T = 1.5$ K and $\phi = 15^\circ$; the B values are given in the inset key.

the experimental AMRO amplitudes can be plotted against the orbit frequency ω (known from the bandstructure [9, 10]) multiplied by τ . As can be seen in Fig. 4, the experimental AMRO amplitudes lie on a “universal curve” for each ϕ , giving confidence in the approach.

Once the τ^{-1} values are extracted, it is found that they follow a T -dependence of the form $\tau^{-1} = \zeta + \chi T^n$, with $n \approx 2$ (see *e.g.* Fig. 4, inset) and a $T = 0$ scattering rate close to that measured by other means [15]. This strongly suggests that the T -dependent attenuation of the AMROs is due to electron-electron scattering. A T^2 dependence of the scattering rate has been previously inferred from $B = 0$ resistivity measurements [14]. However, problems in deconvolving the in-plane resistivity component $\rho_{||}$ from ρ_{zz} in experimental data [9, 15], and the influence

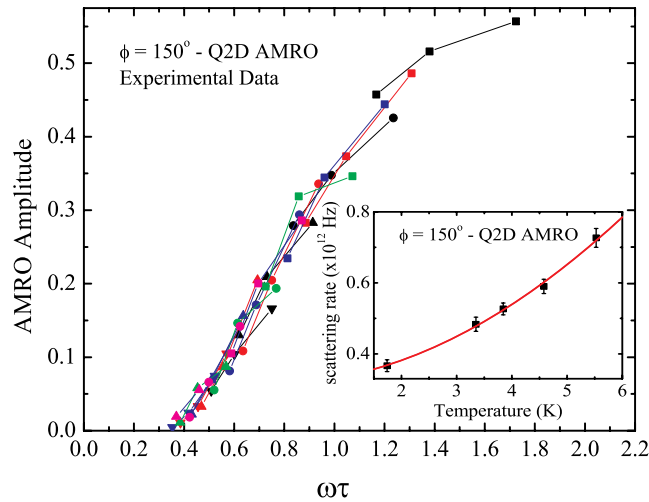


Figure 4. Experimental AMRO amplitudes for $\phi = 150^\circ$ plotted as a function of the orbit angular frequency ω times scattering time τ . The Yamaji AMRO indices [9] i are $i = 2$ (square), $i = 3$ (dot), $i = 4$ (triangle) and $i = 5$ (inverted triangle). Temperatures are 1.7 K (black), 3.4 K (red), 3.8 K (blue), 4.6 K (green) and 5.5 K (purple). The inset shows the T -dependence of τ^{-1} for this ϕ , fitted to a function of the form $\zeta + \chi T^n$, with $n = 1.8 \pm 0.4$.

of the superconducting transition on the T -dependence of the measured resistivity [9, 16] has meant that this attribution could not be considered conclusive. By contrast, the T -dependent AMRO provide a stringent and unambiguous gauge of the scattering rate of the normal-state quasiparticles, allowing the electron-electron scattering mechanism to be definitively identified.

This work was supported by US Department of Energy (DoE) grants LDRD 20040326ER and LDRD 20030084DR. Work at NHMFL is performed under the auspices of the National Science Foundation, DoE and the State of Florida.

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