

Dichotomy between the Hole and Electron Behavior in Multiband Superconductor FeSe Probed by Ultrahigh Magnetic Fields

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Magnetoresistivity ρ_{xx} and Hall resistivity ρ_{xy} in ultrahigh magnetic fields up to 88 T are measured down to 0.15 K to clarify the multiband electronic structure in high-quality single crystals of superconducting FeSe. At low temperatures and high fields we observe quantum oscillations in both resistivity and the Hall effect, confirming the multiband Fermi surface with small volumes. We propose a novel approach to identify from magnetotransport measurements the sign of the charge carriers corresponding to a particular cyclotron orbit in a compensated metal. The observed significant differences in the relative amplitudes of the quantum oscillations between the ρ_{xx} and ρ_{xy} components, together with the positive sign of the high-field ρ_{xy} , reveal that the largest pocket should correspond to the hole band. The low-field magnetotransport data in the normal state suggest that, in addition to one hole and one almost compensated electron band, the orthorhombic phase of FeSe exhibits an additional tiny electron pocket with a high mobility.

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Uniquely amongst the Fe-based superconductors, tetragonal β -FeSe undergoes an orthorhombic distortion at $T_s \approx 90$ K without accompanying long-range magnetic order at any temperature, and therefore provides a unique opportunity to probe the unusual electronic structure in the symmetry-broken state from which superconductivity emerges below ~ 9 K. In particular, recent advances in high-quality single crystal growth [1] and several theoretical proposals focused on the role of frustrated magnetism and nematicity [2–4] have led to renewed interest in this system as a key material of Fe-based superconductivity. Furthermore, it has been suggested that in the superconducting state FeSe may be in the crossover regime between weak-coupling Bardeen-Cooper-Schrieffer and strong-coupling Bose-Einstein condensation limits, which may cause unexpected effects [5]. To understand such intriguing properties of FeSe, the detailed experimental determination of its electronic structure is crucial.

Previous measurements of electronic properties of FeSe using both ARPES [6–9] and quantum oscillations [9–11], have suggested different interpretations of the electronic structure. In this Letter, by analyzing the high-field magnetotransport data, we reveal the details of the bulk electronic structure and the effects of scattering from which

a more complete picture of the electronic structure of FeSe can be constructed. We use a novel approach to extract this information, analyzing the quantum oscillations measured both in the resistivity ρ_{xx} and Hall effect ρ_{xy} , which allows us to deduce the origin of the observed oscillation frequencies in FeSe in fields up to 88 T. We observe a positive Hall effect suggesting that, in a two band picture, hole carriers have higher mobility than the (almost compensated) electrons at low temperatures. At higher temperatures, the Hall effect changes significantly with magnetic field, which can only be understood considering a three-band picture, with an additional small electron band with high mobility. Furthermore, the observed large, nonsaturated magnetoresistance is the consequence of the existence of a small and compensated multiband electronic structure in FeSe.

Experimental details.—Samples were grown by the KCl/AlCl₃ chemical vapor transport method [1]. In-plane transport measurements were performed using state-of-the-art extreme conditions of high magnetic fields combined with very low temperatures, in a dilution fridge in pulsed fields up to 56 T and up to 88 T in ⁴He cryostats at the LNCMI, Toulouse [12,13], and dc fields to 33 T at the HFML, Nijmegen. Low-field measurements (up to 13.5 T) were performed in a Quantum Design PPMS in Oxford.

The Hall and resistivity contributions were separated by (anti)symmetrizing the data obtained in positive and negative magnetic fields applied along the c axis. Good electrical contacts were achieved both by spot welding and Sn or In solder and to avoid heating effects electrical currents below 10 mA in pumped helium cryostats or 1 mA for the dilution refrigerator were used. Quantum oscillations were observed in four samples and two samples were measured below 1.4 K in the dilution refrigerator. One sample (S1) was also measured up to 88 T at 1.4 K.

High-field magnetotransport measurements.— Figures 1(a) and 1(c) show the field dependence of ρ_{xx} and ρ_{xy} for sample S1 at very low temperatures (below 1.3 K). We observe the transition from the superconducting to the normal state around 14 T, and a very large non-saturating magnetoresistance at low temperatures. The observation of quantum oscillations in the Hall effect is less commonly reported than Shubnikov-de Haas oscillations in resistivity, but has been recently studied in other multiband systems, such as cuprates [15] and $\text{Ca}_3\text{Ru}_2\text{O}_7$ [16], although to the best of our knowledge not previously in Fe-based superconductors.

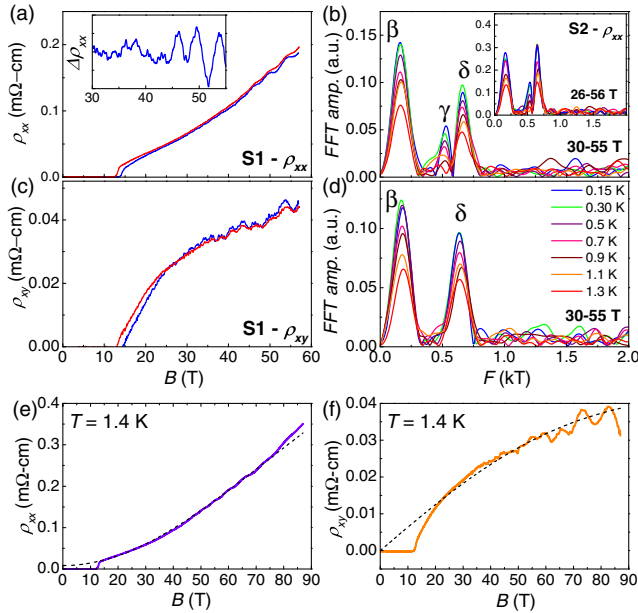


FIG. 1 (color online). High-field magnetotransport data. Magnetic field dependence of (a) ρ_{xx} and (c) ρ_{xy} for sample S1 at dilution fridge temperatures. The inset in (a) shows the oscillatory part of ρ_{xx} at 0.15 K. The corresponding fast Fourier transform (FFT) spectra of (a) and (c) using a large field window (27–56 T) is shown in (b) and (d), respectively, which identifies a series of frequencies, β , γ , and δ . Notably, the γ peak is absent (or very weak) in the Hall effect component, ρ_{xy} . The inset shows the Fourier spectrum for sample S2. (e) Resistivity and (f) Hall effect measurements at ultra-high magnetic field up to 88 T at 1.4 K for sample S1. The dashed lines correspond to a combined fit to a two-band model to the data, as described in the text and Ref. [14].

Quantum oscillations analysis.—The temperature dependence of the amplitude of quantum oscillations in resistivity is usually well described by the Lifshitz-Kosevich formula describing oscillations in thermodynamic quantities [17]. Analysis of the temperature dependence of the amplitude of oscillations, as shown in Figs. 1(b) and 1(d), allows us to determine the effective masses by fitting to the Lifshitz-Kosevich formula (see Table. S I in the Supplemental Material [14]). We find good agreement in the values of the quantum oscillation frequencies and effective masses between samples (within the error bars) and with those reported previously by Terashima *et al.* [10]. The γ orbit has a particularly heavy effective mass of $(7-8)m_e$ (m_e being the electron mass), whereas the β and δ orbits have effective masses of around $4m_e$, which suggests a different origin of the two frequencies. The field window in $1/B$ used in the pulsed field experiments (26–56 T for sample S2) is not large enough fast Fourier transform (FFT) to identify frequencies smaller than the β frequency at 180 T, as compared with studies in dc fields in which an α pocket of about 50 T has been resolved [10].

Usually, it is impossible to determine based purely on quantum oscillation data whether a given quantum oscillation frequency arises from an electronlike or holelike band, although often a comparison to band structure calculations can give a satisfactory understanding. However, here we develop a novel argument based on a compensated two-carrier model and the relative strength of oscillation peaks in ρ_{xx} and ρ_{xy} , in order to argue that the γ peak originates from an electronlike Fermi surface pocket, whereas the β and δ are associated with the minimum and maximum of the holelike band.

Our approach is based on the observation that the γ orbit is missing, or at least much weaker, in the ρ_{xy} FFT spectrum than that of ρ_{xx} , as can be directly seen by comparing the FFT spectra from sample S1 in Figs. 1(b) and 1(d). Note that in this sample, ρ_{xx} and ρ_{xy} are calculated by symmetrizing data from the same pair of contacts, which rules out any external explanation such as temperature gradients in the sample. In a multiband system, ρ_{xx} and ρ_{xy} may be written in terms of the conductivities of the various bands. It can be shown (see Ref. [14] for derivation) that in a compensated metal (i.e., equal number of holes and electrons, which stoichiometric FeSe must have) where both the electron and hole conductivities have an oscillatory component, that the Hall effect is *relatively* more sensitive to the oscillations of the more mobile carrier compared to resistivity; that is to say that if both electronlike and holelike oscillations are observed in a FFT of ρ_{xx} oscillations, the FFT spectrum of ρ_{xy} will show a relatively enhanced amplitude of the more mobile carrier. In FeSe, the sign of the Hall coefficient at low temperatures and high fields is positive, i.e., the holes are (on average) more mobile, therefore the holelike orbits are enhanced in the ρ_{xy} FFT spectrum and the missing or weak 540 T γ

peak is an electronlike orbit whereas both the β and δ frequencies are likely to be holelike frequencies.

By assigning the δ and β to the maximum and minimum of a quasi-2D pocket (as suggested in Ref. [10]), we estimate that this band would contain 0.014 electrons/Fe, equivalent to a carrier density for the hole band of $n_h = 3.6 \times 10^{20} \text{ cm}^{-3}$. We note that the observation of a positive high-field Hall coefficient which implies that the hole carriers are more mobile (on average) than the electron carriers is unusual in Fe-based superconductors, where the Hall effect is typically found to be negative (e.g., Refs. [18–21]), and quantum oscillation studies have typically found much higher amplitudes (longer scattering times) for the electron pockets than the hole pockets [22,23], and sometimes the hole pockets are not observed at all [24,25].

Low-field magnetotransport behavior.—So far we have focused on the low-temperature, high magnetic field measurements, since these are the conditions where quantum oscillations are observed. However, a lot of information at higher temperatures can be obtained from low-field studies. Figures 2(a) and 2(b) show the temperature dependence of the Hall effect and the relative transverse magnetoresistance up to 14 T of a single crystal of FeSe (S3). At high temperature, the Hall effect is observed to be linear up to 10 T, and changes sign twice above 90 K. In this high-temperature regime, the most natural model to turn to is a compensated two-band model, where the two bands represent the electron and hole pockets (charge compensation is required in a stoichiometric FeSe crystal). By simultaneously fitting ρ_{xx} and ρ_{xy} we can extract three free parameters $n = n_e = n_h$, μ_e , and μ_h . A feature of this model is that the Hall effect is strictly linear at all magnetic fields and the magnetoresistance does not saturate [14]. This model was also used for LiFeAs, where it was found that the Hall effect was negative and linear up to 14 T at all temperatures [21].

The temperature dependence of the extracted parameters, assuming an equal number of holes and electrons with the carrier density $n = n_h = n_e$ and the two (field-independent) mobilities μ_h and μ_e , is shown in Figs. 2(e) and 2(f). The carrier density is relatively constant at high temperatures, $n \sim 3 \times 10^{20} \text{ cm}^{-3}$, which is close to the low-temperature value extracted for the hole band density from quantum oscillations. Although in many iron-based superconductors the electrons are generally more mobile and therefore the Hall effect is commonly found to be negative [21], in the case of FeSe the mobilities of holes and electrons are very similar at high temperature with subtly different temperature dependency [see Fig. 2(f)]. The sign of the low-field R_H [Fig. 2(c)] changes twice above 90 K, as at some temperatures the electrons are slightly more mobile, and at other temperatures the holes are.

The three-band model.—Below the structural transition at T_s (~ 75 K), the Hall effect becomes noticeably nonlinear

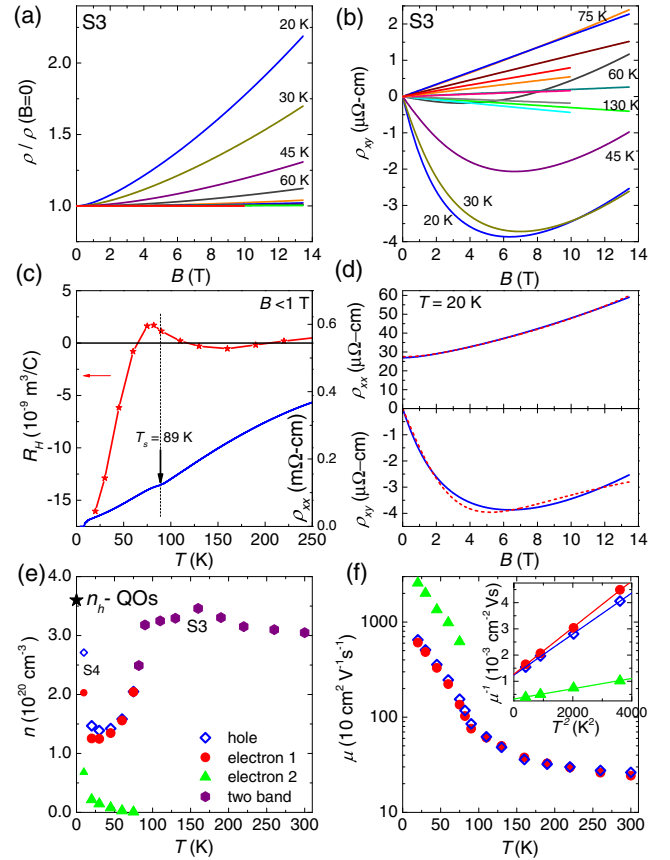


FIG. 2 (color online). High-temperature magnetotransport data. (a) Magnetoresistance measured to 13.5 T in a sample S3 at temperatures between 20–300 K. (b) Hall effect in FeSe. (c) Low-field Hall coefficient, defined by a straight-line fit to data < 1 T. The temperature dependence of the resistivity for S3 is also shown. (d) Simultaneous fit of the magnetoresistance and Hall effect using a constrained compensated three-carrier model. (e) Temperature dependence of the carrier density extracted from two- and three-carrier fits (a three-carrier model is used below 75 K which accounts for the development of the low-field non-linearity in the Hall effect data). (f) Temperature dependence of mobility. Inset: The inverse of mobility, proportional to the scattering rates, which has a $\sim T^2$ dependence in the range of 20–60 K.

and such behavior cannot be understood in a compensated two-band model. Recently, a mobility spectrum analysis of FeSe [26] demonstrated the presence of high-mobility electron carriers (beside less mobile hole and electron carriers) which could account for the nonlinear Hall effect and, in particular, the negative sign of the Hall coefficient at low fields, but becoming positive at high fields. The Hall effect in FeSe is also similar to that in annealed BaFe_2As_2 single crystals [27]. Although the Fermi surface in the magnetically reconstructed phase is completely different, there are coincidental similarities in that there is a small but compensated number of carriers and, in particular, a small number of highly mobile electronlike carriers. A three-carrier model [28] can describe well the Hall effect data in annealed single crystals of BaFe_2As_2 [27]. As FeSe

TABLE I. Parameters extracted from magnetotransport measurements on FeSe using both a compensated ($n_{e1} = n_{h1}$) and uncompensated ($n_{e1} \neq n_{h1}$) two-band model to fit the low-temperature-high field regime, as well as a three-band model to fit the low-field-high-temperature data for different samples, as discussed in the text. The carrier density n and mobility μ of each carrier can be compared with the value from quantum oscillations for the hole band of $n_h = 3.6 \times 10^{20} \text{ cm}^{-3}$.

Parameters (model)	T (K)	ΔB (T)	$h1$	$e1$	$e2$
n (two-band, S2) (10^{20} cm^{-3})	1.5	28–56	3.8(1)	4.7(1)	...
μ (two-band, S2) (cm^2/Vs)			1335	499	...
n (two-band, S2) (10^{20} cm^{-3})	1.5	28–56	3.5(1)	3.5(1)	...
μ (two-band, S2) (cm^2/Vs)			756	401	...
n (three-band, S4) (10^{20} cm^{-3})	10	0–13.5	2.71	2.03	0.68
μ (three-band, S4) (cm^2/Vs)			623	457	1843

displays a similar behavior we implement this approach but further constrain it by simultaneously fitting ρ_{xx} and ρ_{xy} for a compensated system with one hole and two electron pockets (with five free parameters for mobilities and carrier concentrations). Figure 2(d) shows the fits of this model to the data at 20 K which account well for the low-field, nonmonotonic behavior and reveal the presence of a small but highly mobile electron pocket. Thus, our analysis suggests that the Fermi surface of FeSe must have more than one electron band, and the magnetotransport behavior is better described if three carriers are present, i.e., one hole and two electron pockets.

Recent high-resolution ARPES data on FeSe [9], show that the Fermi surface deforms significantly, breaking the rotational symmetry, when the system goes from the tetragonal to the orthorhombic phase at T_s . At high temperatures, the Fermi surface is composed of two hole bands ($n_{h1} \gg n_{h2}$) and likely two electron bands (n_{e1} and n_{e2}), which are well compensated. At low temperatures, however, there is a single hole band n_{h1} with a size close to one of the electron bands n_{e1} , but the second electron band n_{e2} is rather small. In terms of magnetotransport, this scenario is equivalent to having two types of electronlike carriers, one with similar but slightly lower mobility compared to the hole bands, whereas the second electron band with n_{e2} carriers, may have lighter effective masses and consequently higher mobility. Using the three-band model from our magnetotransport data on FeSe, the new small pocket has a carrier density of $n_{e2} = 0.7 \times 10^{20} \text{ cm}^{-3}$ and high mobility with 1843 cm^2/Vs . This pocket would correspond to a very low frequency of 90(10)T in the quantum oscillations with a lighter effective mass of $\sim 1.5(5)m_e$ compared to the other bands. These values are in good agreement with those extracted for this pocket from quantum oscillations for the α pocket [10] and the small electron band from high resolution ARPES studies [9].

In the low-temperature limit, in the normal state (once the superconductivity is suppressed above 14 T) the ρ_{xx} and ρ_{xy} data are well described by a two-band model over a large field window, with the extracted values listed in Table I. We find that the hole mobility is 1.9–2.7 times larger than the electron mobility, which would be enough to

give a noticeable difference in the relative amplitudes of oscillations in ρ_{xx} and ρ_{xy} as argued above. For a perfectly compensated system, ρ_{xy} is exactly linear in the high-field limit both in the two- and three-band models, whereas experimentally there is a small amount of curvature at high fields. This could indicate a small departure from stoichiometry, or possibly a field dependence of the mobility. Another possibility concerns the field-induced Fermi surface effects caused by the Zeeman splitting whose energy scale is expected to become comparable to the effective Fermi energy of the smallest band ($\sim 2\text{--}4 \text{ meV}$ [5,10]) in the highest magnetic field range. Although no clear signature of such a Lifshitz transition is observed, the presence of a small pocket superimposed on other frequencies is hard to resolve. Despite this, our quantum oscillations analysis in very high magnetic fields based on a two-carrier model allows us to assign reliably the origin of the hole and electron pockets.

While at low fields and high temperatures the two- and three-carrier models capture the data well, the extracted temperature dependence of the carrier density shows a marked reduction below T_s [Fig. 2(e)], dropping by a factor of ~ 2 compared to the high-temperature values. Furthermore, the carrier density n_h estimated from quantum oscillations (assuming the β and δ bands form the quasi-2D hole band) is $3.6 \times 10^{20} \text{ cm}^{-3}$ —similar to the high-temperature carrier densities but substantially higher than the 20 K data. Below 20 K, the carrier density seems to recover, as shown by extracted parameters on a different sample S4 (see Ref. [14]), and furthermore the pulsed-field low-temperature data gives carrier densities from two carrier fits which are comparable with those estimated from quantum oscillations. In order to reconcile the apparent drop in carrier density below T_s with the observation from ARPES [9] and the expectation that the Fermi surface should not change significantly in the absence of any spin-density-wave order, we speculate that a strongly anisotropic scattering rate may exist on the Fermi surfaces below the structural transition. If there are strong spin fluctuations (as observed by NMR below T_s [29]), parts of the hole and electron Fermi surfaces which are well nested

by the antiferromagnetic ordering vector will have a dramatic increase in the scattering rate, similar to calculations in Ref. [30], and will be effectively *shorted out* by non-nested segments of the Fermi surface with a much longer scattering time (and which may have conventional $\sim T^2$ Fermi-liquid temperature dependence of mobility [Fig. 2(f)]). This may also be related to the anomalous quasiparticle interference in FeSe, which shows very strong in-plane anisotropy of energy dispersion [5]. One can then envisage that in our simple model which assumes isotropic scattering on all bands, the drop in the *effective* carrier density, without an actual change in Fermi surface volume, could be a manifestation of strongly anisotropic scattering, with almost half of the available carriers *missing*. However, at very low temperatures, the scattering becomes more isotropic (impurity scattering only in the $T = 0$ limit) [30]. In this limit (in which we observe quantum oscillations) we find that the carrier densities extracted from the high-field two-carrier model are comparable to those estimated from quantum oscillations, n_h [see Fig. 2(e)].

In summary, magnetotransport studies in ultrahigh magnetic fields and at low temperatures allow us to access the Fermi surface of FeSe and to determine independently the signs of the charge carriers. In the limit of isotropic scattering, our novel analysis of the quantum oscillations in the resistivity and Hall effect, suggest the presence of holelike bands with more mobile carriers than the almost compensated electronlike band. Furthermore, the high-temperature and low-field data can be described only if an additional highly mobile electron pocket is considered. We find that the magnetotransport behavior of FeSe is a direct consequence of the small multiband Fermi surface with different carrier mobilities. Further theoretical work will be required to understand the role played by anisotropic scattering in FeSe and how it can affect its normal and superconducting properties.

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