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

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Abstract. The interlayer magnetoresistance $\rho_{zz}$ of the organic metal $\kappa$-(BEDT-TTF)$_2$Cu(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 $\rho_{zz}$ 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 $\sim 25$. 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, \textit{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]

$$k_B T > t_\perp,$$

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 $\kappa$-(BEDT-TTF)$_2$Cu(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 $t_a = 0.065 \pm 0.07$ meV [10, 11] and standard laboratory $T$s allow the inequality in Eq. 1 to be exceeded by orders of magnitude ($t_a/k_B \approx 0.5$ 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 $\rho_{zz}$ observed when the field $B$ 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 $\kappa$-(BEDT-TTF)$_2$Cu(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 $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ sample as a function of field (B) orientation ($\theta = 90^\circ$ is B in-plane; $\theta = 0$ is B normal to the planes). The rotation plane is defined by $\phi = 160^\circ$ (see [10] for details of the coordinates); here, the $\rho_{zz}$ 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 $\rho_{zz}$ data for $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ for $5.3$ K $\leq T \leq 14.6$ 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 $t_a \approx 0.065$ 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 $\sim 25$ [12].

Having demonstrated that the Fermi surface of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ remains 3D up to at least $T \approx 13$ K [12], it is informative to look at the $T$-dependence of other features in $\rho_{zz}$, such as angle-dependent magnetoresistance oscillations (AMROs). Fig. 2(a) shows $\rho_{zz}$ of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ over a wider angular range than Fig. 1 and for a different plane of rotation, defined by the angle $\phi$ [10]. For this $\phi$, 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 $\rho_{zz}$.

To extract quasiparticle scattering rates from AMROs, numerical calculations of $\rho_{zz}$ 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 $\tau^{-1}$, an input parameter of the model. Once this has been done,
Figure 2. (a) $R_{zz}$ for $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ at several $T$ and $B = 45$ T; $\phi = 90^\circ$ so that features in $\rho_{zz}$ are chiefly due to the Q2D Fermi-surface sections. (b) The amplitude of various features in $\rho_{zz}$ 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 $\rho_{zz}$ 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 $\omega$ (known from the bandstructure [9, 10]) multiplied by $\tau$. As can be seen in Fig. 4, the experimental AMRO amplitudes lie on a “universal curve” for each $\phi$, giving confidence in the approach.

Once the $\tau^{-1}$ 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 $\rho_{zz}$ 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 $\omega$ times scattering time $\tau$. 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 $\tau^{-1}$ for this $\phi$, 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.

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