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Quantum galvanomagnetic effects in the organic metal α-(BEDT-TTF)$_2$TlHg(SCN)$_4$

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The transverse magnetoresistance and Hall effect of the organic metal α-(BEDT-TTF)$_2$TlHg(SCN)$_4$ have been studied in static magnetic fields of up to 33 T applied perpendicular to the highly conducting planes. The Hall resistivity is found to exhibit strong oscillations caused by the coexistance of quasi-one-dimensional and quasi-two-dimensional states in the electronic system. In higher quality crystals, plateaulike features appear at magnetic fields above ~30 T, which are consistent with recent claims of the quantum Hall effect in this material. [S0163-1829(97)52324-1]

The quantum Hall effect (QHE) tends to be regarded as a property of two-dimensional (2D) semiconductor systems such as GaAs-(Ga, Al)As heterostructures or Si metal-oxide-semiconductor field-effect transistors.[1–3] However, recent pulsed magnetic field measurements of the quasi-two-dimensional (Q2D) organic metals α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ (M=K, Tl) (Refs. 4 and 5) revealed sharp negative spikes in the magnetization, apparently due to quasipersistent eddy currents; it was suggested[4] that these were associated with the deep minima in the resistivity component ρ_{xx} that accompany the quantum Hall effect plateaus[1,2] in ρ_{xy}. The pulsed-field magnetization studies stimulated numerical calculations[6] of ρ_{xx} and ρ_{xy} employing a Fermi surface consisting of a Q2D cylinder and a pair of weakly-warped quasi-one-dimensional (Q1D) sheets; the α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ (M=K, Tl) salts are thought to possess such a Fermi surface in their high-field states.[7,8] These calculations indicated that the field-dependent behavior of ρ_{xx}, ρ_{yy}, and ρ_{xy} of the charge-transfer salts could under certain circumstances be rather different from those of the 2D semiconductor systems, chiefly due to the additional presence of the Q1D Fermi-surface components.[6] Nevertheless, at sufficiently high magnetic fields, QHE plateaus were predicted to occur[6] in ρ_{xy} in the charge-transfer salts. In this paper, we describe measurements of the transverse magnetoresistance and the Hall effect of single crystals of α-(BEDT-TTF)$_2$TlHg(SCN)$_4$ designed to check the calculations of Ref. 6 and to establish the possibility of observing the quantum Hall effect.

Most previous resistivity studies of the M=K, Tl salts have involved measurements of ρ_{zz}, the resistivity component in the direction perpendicular to the highly conductive ac planes.[7] The reasons for this are twofold. First, the resistivity in the interplane direction is usually around three orders of magnitude larger (and therefore easier to measure) than that in the intraplane direction, reflecting the extreme anisotropy of the band structure.[8–10] Second, the single crystals tend to be irregular in shape, so that the apparent measured in-plane resistivity ρ_m is a combination of all of the components of the resistivity tensor. For example, in the simplest measurement geometry, i.e., four contacts on the same ac face of the crystal, the current is concentrated near this face, leading to an apparent increase of the in-plane resistivity; in Ref. 11 it was noted that for samples in which $t/l \geqslant (\rho_{||}/\rho_{zz})^{1/2}$, where t and l are the sample thickness and distance between the current contacts respectively, and $\rho_{||} \approx \frac{1}{2}(\rho_{xx}+\rho_{yy})$, the measured resistivity is $\rho_m \approx 2(t/l)(\rho_{||}\rho_{zz})^{1/2}$.

Bearing these considerations in mind, two single crystal samples of α-(BEDT-TTF)$_2$TlHg(SCN)$_4$, prepared using standard electrochemical techniques,[12] were selected for this study. Sample A was very thin, having dimensions

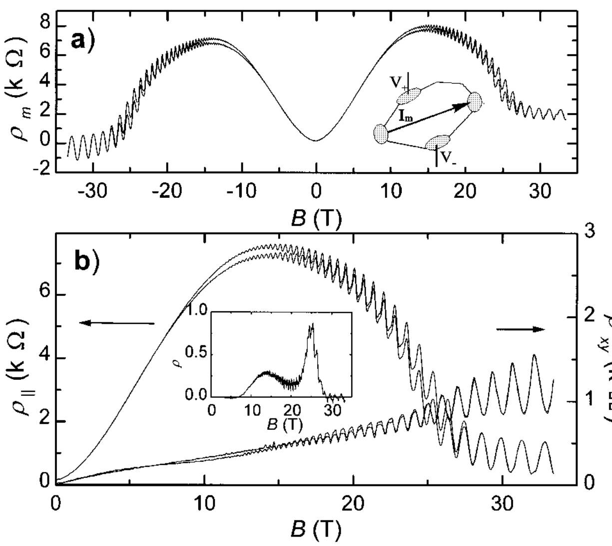

FIG. 1. (a) The measured resistivity ρ_m for sample A (expressed in two-dimensional resistivity units) for both rising and falling positive and negative polarity sweeps of the magnetic field. Sample geometry and contact configuration are shown in the inset. (b) The transverse resistivity $\rho_{||}$ and the Hall resistivity ρ_{xy} extracted from the data in (a). The inset displays the hysteresis in between rising and falling magnetic fields.

$\approx 1\times 1\times 0.008$ mm^3; it was chosen because $(\rho_{zz}t^2/\rho_{||}l^2)\ll 1$, minimizing the mixing of the resistivity components referred to in the previous paragraph. Sample B had less ideal dimensions $\approx 1.7\times 1.5\times 0.08$ mm^3; however, pulsed-field magnetization studies had previously shown that sample B exhibited the quasipersistent currents[4] thought to be characteristic of the QHE. In order to further minimize mixing between in-plane and interplane resistivity components, platinum wires were connected to the *edges* (rather than the faces) of both samples using graphite paint. Steady magnetic fields were provided by a Bitter magnet at the National High Magnetic Field Laboratory in Tallahassee,[13] and one of the in-house ^{3}He cryostats was used to cool the samples to 700 mK. Resistance measurements were carried out using ac currents of 5–20 μA and 20–300 Hz.

Figure 1(a) shows the measured resistivity ρ_m for sample A, for both positive and negative magnetic fields applied perpendicular to the ac plane. ρ_m has been obtained by multiplying the measured resistance by t/b, where b is the interplane lattice constant. The current and voltage contacts were arranged so that ρ_m contains contributions from both $\rho_{||}$ and ρ_{xy} [see inset to Fig. 1(a)]; this is obvious from the raw data, the Hall component causing the lack of symmetry of ρ_m about zero field (Fig. 1). The two resistivity components can be separated because of their differing dependence on the *sign* of the magnetic field; $\rho_{||}$ is the average of the ρ_m data for positive and negative fields, and ρ_{xy} is half their difference. Figure 1(b) shows $\rho_{||}$ and ρ_{xy} data extracted in this manner.

Our chief preoccupation in this paper will be the behavior of ρ_{xy} in high magnetic fields. However, before examining this in detail, we shall briefly discuss the $\rho_{||}$ data. Below $\sim$4 T, $\rho_{||}$ rises approximately quadratically with field. At higher fields, $\rho_{||}$ deviates from quadratic behavior and rises to a maximum (about 40 times larger than the zero field value) at $\sim$12 T, and hysteresis between up and down sweeps of the magnetic field is shown between $\sim$10 T and $\sim$28 T; in all of these respects, the behavior of $\rho_{||}$ in this sample is qualitatively similar to that of ρ_{zz} observed in other measurements of α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ (M=K, Tl) crystals.[7,14,15] At fields of between 20 and 28 T, $\rho_{||}$ drops steeply in the well-known ''kink,'' a first order phase transition between the low-field, low-temperature spin-density-wave (SDW) state known to occur in the M= K, Tl, Rb salts and what is thought to be a metallic phase characterized by the unreconstructed Fermi surface, consisting of a Q2D hole pocket and a pair of Q1D electron sheets.[4,7] The hysteresis reaches its largest magnitude at fields around the kink transition[16] [Fig. 1(b), inset], with a weaker maximum occurring below. Shubnikov–de Haas oscillations of frequency 670 T are clearly observable at all fields, in agreement with previous studies;[4,14,19] these correspond to carrier orbits about the closed Q2D section of the Fermi surface.[7]

In contrast to $\rho_{||}$, ρ_{xy} exhibits weaker structure throughout the SDW regime. Taking the slope of the approximately linear portion of ρ_{xy} below $\sim$4 T, a carrier density of $\sim 8\times 10^{25}$ m^{-3} is obtained, corresponding to $\sim 1.6\times 10^{17}$ m^{-2} per layer, somewhat higher than the value found in Ref. 20 for α-(BEDT-TTF)$_2$KHg(SCN)$_4$. The density found in this paper represents half of the cross-sectional area of the Q2D hole pocket responsible for the 670 T frequency Shubnikov–de Haas oscillations, but this poor agreement is hardly surprising, given the complex Fermi surface topology[7] of the α-phase BEDT-TTF salts within the SDW phase (see Ref. 20 for a discussion of the Hall effect in metals with complex Fermi surfaces). Above $\sim$4 T, the Hall coefficient starts to decrease in size, becoming $\sim$30% smaller by 10 T.

Oscillations in ρ_{xy}, of frequency 670 T, begin to be observable above $\sim$15 T, and become rather strong at fields above the kink transition. Above the kink, ρ_{xy} becomes greater than $\rho_{||}$, an additional sign that the Fermi surface has taken on a simpler topology.[1,2,21] In this regime, meaningful comparisons can be made with the model calculations of Ref. 6, carried out for a Fermi surface consisting of Q1D and Q2D components. For such a system, ρ_{xy} is given by[6]

$$\rho_{xy}\sim \frac{\sigma_{xy}}{\sigma_{xy}^2+(\sigma_{xx}+\sigma_{1D})\sigma_{yy}}, \tag{1}$$

where σ_{xx} and σ_{yy} are the conductivities associated with the Q2D states, and σ_{1D} is the conductivity of the Q1D states, whose velocities are assumed to be aligned along the x direction. When the chemical potential μ is situated in a gap between the well-resolved Landau levels of the Q2D states, σ_{xx} and σ_{yy} effectively vanish, and it can be shown that[6] $\rho_{xy}\sim 1/\sigma_{xy}=R_H=B/eN_{2D}$, N_{2D} being the total number of Q2D states; if the Landau levels of the Q2D states are sharp and well separated, a QHE plateau can then develop over a finite range of magnetic field around this point,[6] as in the 2D semiconductor systems. The existence of QHE plateaus requires μ to be pinned in states distinct from those of the mobile (Q)2D carriers; in the semiconductor case, these are the localized states at the edges of the Landau levels, whereas in the charge-transfer salts this role is taken by the Q1D part of the Fermi surface.

When μ is in the center of the nth Landau level, the conductivity of the Q2D states will reach a maximum value[22] $\sigma_{xx} \approx \sigma_{yy} \approx (n+\frac{1}{2})e^2/(\pi^2\hbar) \sim 1/(\pi R_H)$. If the Q1D states are assumed to behave in a Drude-like manner at high magnetic fields,[6] this results in very deep minima in ρ_{xy}. However, if the Q1D states are assumed to be localized at high fields, ρ_{xy} just exhibits a steady increase between the QHE plateaus,[6] as it does in the 2D semiconductor systems. The Q1D states were also found[6] to have a very distinct effect on ρ_{yy}. Whereas ρ_{xx} exhibits minima (maxima) when μ is between (in) Landau levels, the presence of Drude-like Q1D states leads to very large maxima in ρ_{yy} when μ is between Landau levels, i.e., the oscillations in ρ_{xx} and ρ_{yy} are π out of phase.[6] It is only when the conductivity σ_{1D} of the Q1D states becomes small that the behaviors of ρ_{xx} and ρ_{yy} become similar. Thus in Fig. 1(b), the strong oscillations in ρ_{xy} indicate that the Q1D states play some part in the high-field conductivity. However, the oscillations in $\rho_{\parallel}$, which is an average of ρ_{xx} and ρ_{yy}, are π out of phase with those in ρ_{xy}, indicating that σ_{1D} must be relatively small compared to σ_{xx} and σ_{xy} at these high fields. This observation is consistent with the observation of quasipersistent induced currents in the magnetization studies,[4] a prerequisite for which *both* ρ_{xx} and ρ_{yy} should be very small when μ is between Landau levels.[6]

While the peaks in ρ_{xy} should approach the QHE values, the uncertainty of sample thickness means that the experimental values cannot be quantitatively compared with theoretical expectations. Furthermore, the absence of flat plateaus at the tops of the peaks indicates that sample A is probably of insufficient quality for the QHE to be seen at sufficiently low fields. Harmonic analysis of the Shubnikov–de Haas oscillations of sample A supports this assertion; the oscillations of lower quality samples show only a small higher harmonic content.[10] However, in sample B, there is evidence for the emergence of QHE plateaus. The much greater thickness of sample B means that the current path will be less straightforward, and that measurements of ρ_m will probably contain some component of ρ_{zz}. Figure 2, showing measurements for three different contact configurations, illustrates this problem. The recordings were made on adjacent field sweeps with the sample remaining *in situ*. The data are the average of negative and positive sweeps; the label $\rho'_{\parallel}$ indicates that the result is liable to contain contributions from both the true $\rho_{\parallel}$ and ρ_{zz} components. It is evident that the differing forms of $\rho'_{\parallel}$ cannot be identified with any particular crystallographic axis; it is more likely that they result from complex current paths, perhaps caused by defects and inclusions. This is particularly evident in Fig. 2(c), in which the sign of $\rho'_{\parallel}$ reverses; such an effect can only be due to the effective current path reversing with respect to the voltage terminals, caused by the conflation of conductances that change at different rates with magnetic field. This is analogous to the phenomenon of ‘‘current jetting’’ described by Pippard.[21] Note that Fig. 2(c) also shows the best evidence for the admixture of ρ_{zz} with $\rho_{\parallel}$; as the magnetoresistance oscillations in these two components are in antiphase,[6,10] the resultant

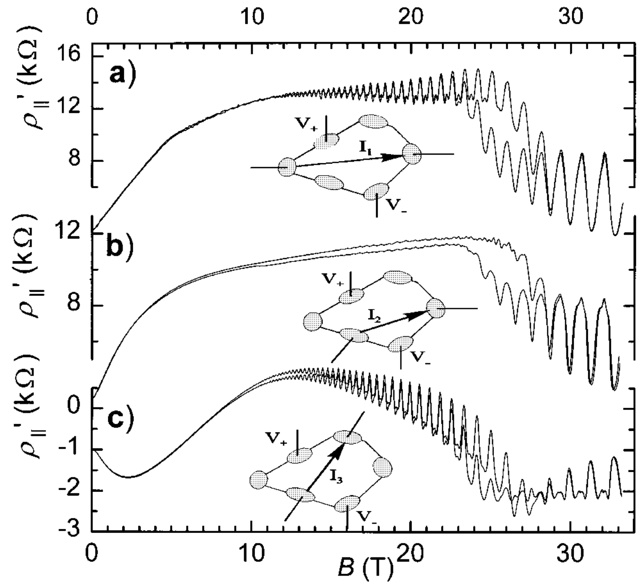

FIG. 2. (a)–(c) The transverse magnetoresistivity $\rho'_{\parallel}$ measured in sample B for different arrangements of the contact geometry. The insets show the respective contact configurations.

oscillations in $\rho'_{\parallel}$ have a double-peaked structure. However, in Figs, 2(a) and 2(b), $\rho'_{\parallel}$ appears to be chiefly determined by $\rho_{\parallel}$.

In contrast, measurements of ρ_{xy} tend to be rather independent of the current distribution. Figure 3 compares $\rho'_{\parallel}$ [Fig. 3(c)] [contact configuration as in Fig. 2(a)] with two recordings of ρ_{xy} [Fig. 3(a)] [contact configuration as in Fig. 2(a)] and Fig. 3(b) [contact configuration as in Fig. 2(b)] and typical pulsed magnetic field magnetization data obtained with the same sample [Fig. 3(d)]. While the absolute value of ρ_{xy} in Figs. 3(a) and 3(b) differs, probably reflecting inhomogeneities in the sample (e.g., regions of different thickness, voids, cracks), the qualitative behavior of

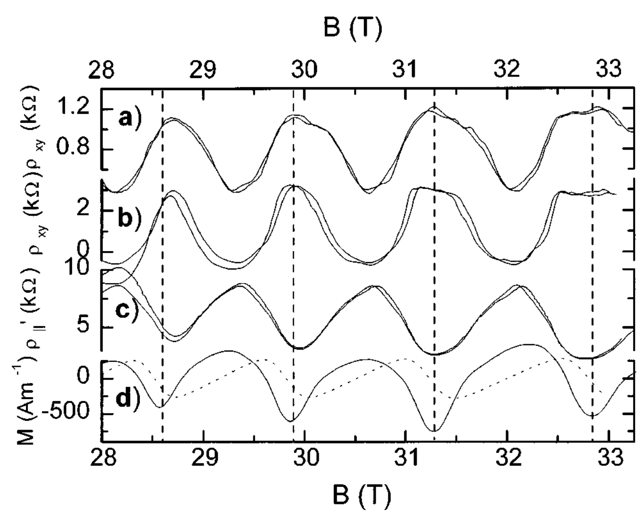

FIG. 3. The Hall resitivity ρ_{xy} in sample B with (a) contact configuration as in Fig. 2(a) and (b) contact configuration as in Fig. 2(b). It is compared in (c) with $\rho'_{\parallel}$ [contact configuration as in Fig. 2(a)] and (d) with the magnetization signal from Ref. 4. The dotted line in (d) shows the calculated magnetization in the absence of quasipersistent eddy currents.

ρ_{xy} in the two traces is very similar and possesses the predicted phase relationship with $\rho_{\|}$ (see the discussion above and Refs. 6 and 10). Furthermore, starting from fields of $\sim$30 T, the field around which the material is believed to be completely transformed into its high field state,[16] plateaus start to emerge at the tops of the oscillations in ρ_{xy}, in agreement with the model predictions that the QHE plateaus should be observed.[6] As expected, the plateaus also coincide with the positions of the eddy current resonances in the magnetization [the strong negative dips in Fig. 3(d)], which are attributed to the deep minima in $\rho_{||}$ that accompany the QHE plateaus.[4]

Unquestionably it was the minima in $\rho_{||}$ that led to the eddy current resonances observed in the magnetization in Ref. 4. However, owing to the inhomogeneities within the sample and contributions to $\rho'_{||}$ from ρ_{zz}, it is not possible to infer to what extent ideal conductivity is reached within regions of the sample. Poorer quality regions of a sample have the effect of adding both parallel and series resistances to the magnetoresistance of the higher quality regions, and so can therefore obscure some of the detail. However, the fact that one of the minima in $\rho'_{||}$ goes below the zero field resistance in Fig. 2(a) is suggestive of the fact that ideal conductivity may be taking place throughout some regions of the sample.

In summary, the results presented in this paper provide direct experimental verification of the recent model predictions,[6] showing that pronounced oscillations of the Hall resistivity occur in a metallic system consisting of both Q2D and Q1D states. At high magnetic fields in the higher quality sample, we observe the emergence of plateaus in the Hall resistivity. This latter observation adds substantial weight to our previous assertion that the QHE occurs in this material at high magnetic fields.[4]

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