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Pulsed-magnetic-field measurements of Hall potential oscillations in α-(BEDT-TTF)$_2$TlHg(SCN)$_4$ within the quantum Hall regime

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Using a variant of the Corbino geometry in pulsed magnetic fields of up to 60 T, we have made direct measurements of the Hall potential in α-(BEDT-TTF)$_2$TlHg(SCN)$_4$ within the quantum Hall regime. This method enables the in-plane components of the resistivity tensor, which are normally very difficult to measure, to be investigated and the nonlinear behavior of the sample's *I-V* characteristics to be studied. It is found that an increasing probability of magnetic breakdown at higher fields leads to a degradation of the quantum Hall effect. [S0163-1829(99)50116-1]

The quasi-two-dimensional (Q2D) metallic charge-transfer salts[1] α-(BEDT-TTF)$_2M$Hg(SCN)$_4$, where M=K, Tl, have attracted considerable attention over the past eight years (see Ref. 2 and references therein for a recent review). One of the remarkable features of these salts is their electronic density of states (DOS) at low temperatures in magnetic fields of $B\sim 30$ T and above, applied perpendicular to the Q2D planes. At such fields, the Fermi surface (FS) consists of a Q2D hole pocket and a pair of quasi-one-dimensional (Q1D) sheets.[2] In many samples, the Landau levels associated with the Q2D FS section are very sharply resolved; at $B\approx 30$ T, the Landau-level spacing $\hbar\omega_c \approx 1.4$ meV, while the broadening due to scattering is ~ 0.07 meV and the interplane bandwidth is ~ 0.2 meV.[3,4] In contrast, the Q1D FS sections give a continuous contribution to the DOS.[4] The form of the DOS has a profound effect on the motion of the chemical potential μ; as B is swept, μ is alternately pinned within the sharp Landau levels and the Q1D states between the Landau levels.[3,4] While μ is between Landau levels, the carriers involved in charge transport cannot perform the closed orbits necessary to cause changes in the Hall resistivity ρ_{xy}, which therefore remains "frozen" at a constant value;[5,6] it has been shown that the plateaux in ρ_{xy} thus produced correspond to a manifestation of the quantum Hall effect (QHE).[6–11] At the same fields, the resistivity component $\rho_\parallel \approx \frac{1}{2}(\rho_{xx}+\rho_{yy})$ exhibits deep minima[7,9,10] and the interplane resistivity ρ_{zz} shows large maxima.[4]

In this paper, we describe measurements of the Hall potential in α-(BEDT-TTF)$_2$TlHg(SCN)$_4$ in this high-field range, made using an analog of the Corbino geometry (Fig. 1) applied to semiconductor systems.[12] Rather than applying an external electric field, we have exploited the annular electric field caused by the changing magnetic field of a pulsed-field magnet;[9] when applied perpendicular to the sample's conducting planes, this induces a circulating current,[9,13] which produces a Hall electric field $E_{\rm H}$ perpendicular to both B and the current. $E_{\rm H}$ will be roughly radial in a cylindrical sample in which $\rho_{xx}\sim\rho_{yy}$, leading to a Hall potential $V_{\rm H}$ between the center and the outer edge of the sample. In a sample with a circular cross section of radius a,

$$V_{\rm H}\approx a^2 \frac{\partial B}{\partial t}\frac{\rho_{xy}}{\rho_\parallel}, \qquad (1)$$

as long as the charge transport is ohmic (i.e., the induced current is proportional to $\partial B/\partial t$).

Equation (1) reveals one of the motivations for measuring $V_{\rm H}$; normally, the large inter/intra-plane anisotropy of BEDT-TTF salts means that $\rho_\parallel$ and ρ_{xy} are extremely difficult to measure reliably,[14] especially at high magnetic fields.[7] However, Eq. (1) can be rearranged to give

$$\frac{\rho_{xy}}{\rho_\parallel}\approx \frac{V_{\rm H}}{a^2(\partial B/\partial t)}, \qquad (2)$$

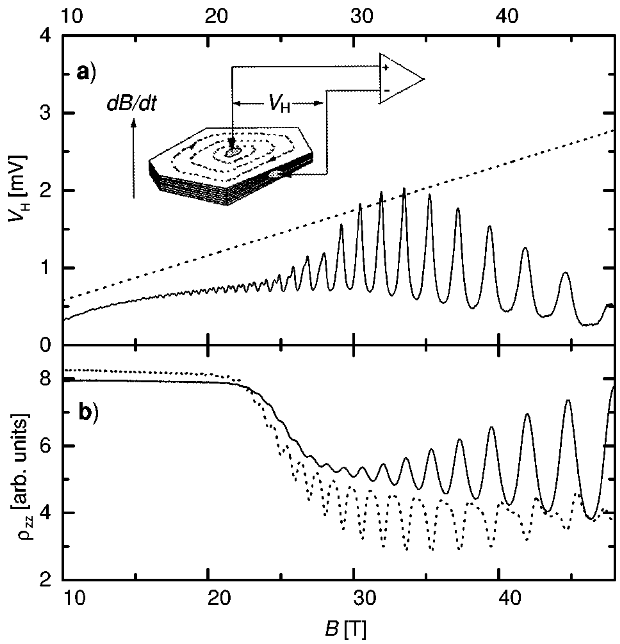

FIG. 1. (a) Measured Hall potential V_H at $T=400$ mK for rising magnetic field, applied perpendicular to the Q2D layers. The dashed line denotes the cyclotron energy divided by the electronic charge, $\hbar\omega_c/e$. (b) The interplane resistivity ρ_{zz} for the same sample at $T=2.1$ K (solid curve) and $T=400$ mK (dotted curve). The inset is a schematic of the contact arrangement used to measure the Hall potential; the dashed curves are the electric field lines for positive $\partial B/\partial t$.

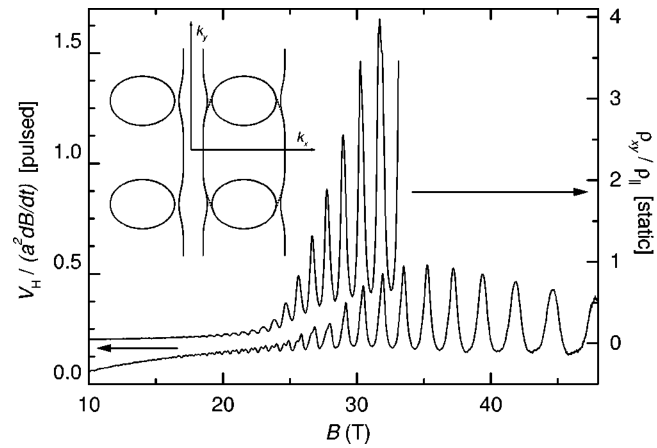

FIG. 2. $V_H/(a^2\partial B/\partial t)$ from pulsed-field measurements compared with the ratio $\rho_{xy}/\rho_\parallel$ from steady-field data ($T\sim0.5$ K). Inset: Schematic of the FS of α-(BEDT-TTF)$_2$TlHg(SCN)$_4$ within the high-field phase; the large magnetic breakdown orbit between the Q1D and Q2D segments is indicated (see Ref. 2 for a discussion of current models of the FS).

implying that a measurement of V_H will give information about the in-plane resistivity tensor components.

Ideally, a Corbino geometry sample should be a disk with a pair of ring contacts, one about a hole in the center, and the other around the edge.[12] Since single crystals of α-(BEDT-TTF)$_2$TlHg(SCN)$_4$ are of a nonideal shape, we have placed one contact on the center of the top surface and one at the edge (see Fig. 1); in contrast to conventional semiconductor systems, α-(BEDT-TTF)$_2$TlHg(SCN)$_4$ is essentially metallic, so that external contacts have little effect on the charge distribution within the sample, ameliorating the nonideal contact geometry. In addition, there is evidence[10,11] that the electronic states at the edges of samples of α-(BEDT-TTF)$_2M$Hg(SCN)$_4$, M=K, Tl, constitute a chiral Fermi liquid[15] of high electrical conductivity at low temperatures and magnetic fields greater than 30 T; this will ensure that the whole of the sample edge is essentially an equipotential. Below we will show that such a situation occurs in the present experiment. The uniformity of BEDT-TTF salts ensures that the variation of the Hall field E_H with distance r from the center is approximately the same from one layer to the next. With the contacts placed as in Fig. 1, the measured voltage should therefore be the same as that for a single layer.

Contact resistances of ~10 Ω were obtained using graphite paint; the voltage leads were connected to a high impedance amplifier. Pulsed magnetic fields of up to 60 T, with a rise time to peak field of ~10 ms, were provided by the NHMFL, Los Alamos;[16] temperatures down to 400 mK were obtained using a plastic ^{3}He refrigerator.

Figure 1(a) shows the Hall potential V_H, measured between the center and the edge of the sample.[17] The background induced voltage due to the open loop area between the wires has been subtracted. At fields below the kink transition (at $B\approx26$ T in the M=Tl salt[7,18]) marking the boundary between the low- and the high-field state, the signal displays a strongly split wave form resembling Zeeman spin splitting, in common with other types of quantum oscillations observed in α-(BEDT-TTF)$_2M$Hg(SCN)$_4$, M=K, Tl.[2,3,7,18,19] Once α-(BEDT-TTF)$_2$TlHg(SCN)$_4$ fully reaches its high-field phase[20] above $B\approx30$ T, strong oscillations occur in V_H. These result from the fact that $\rho_\parallel$ now exhibits very deep minima due to the QHE.[7,10,11] The Hall resistivity ρ_{xy} and the in-plane resistivity $\rho_\parallel$ oscillate in antiphase, with peaks in ρ_{xy}, and therefore peaks in V_H [Eq. (1)], occurring whenever μ lies between adjacent Landau levels.[6–8] Figure 1(b) shows the interplane resistivity ρ_{zz} measured on the same sample using conventional techniques.[2,7] At $T=2.1$ K, the peak positions coincide with the fields where μ lies between Landau levels; this is expected from theories modeling the bulk interplane transport.[4] However, at $T=400$ mK, the oscillations in ρ_{zz} reverse phase, indicating the presence of a chiral Fermi liquid around the edges of the sample.[10,15,21]

Figure 2 shows the measured Hall voltage V_H divided by $a^2(\partial B/\partial t)$ [see Eq. (2); $a\approx0.5$ mm for the present sample] compared with the ratio of ρ_{xy} and $\rho_\parallel$, obtained from conventional magnetotransport experiments in quasistatic fields, provided by a Bitter magnet at NHMFL, Tallahassee; lock-in techniques with an ac sample current of 10 μA and 37 Hz have been applied along with special peripheral contacts[7] to obtain $\rho_\parallel$ and ρ_{xy}. In order to compensate for the geometrical differences between the two measurements, the data sets have been scaled to ensure that the oscillations within the low-field phase (well clear of the QHE regime) are of the same size.[22] Once the high-field phase is entered, the oscillations are notably larger in the static-field data. In order to understand the difference between the pulsed- and quasistatic-field measurements, we must understand the relationship between the circulating current I in the sample and its driving electromotive force V, provided by $\partial B/\partial t$.

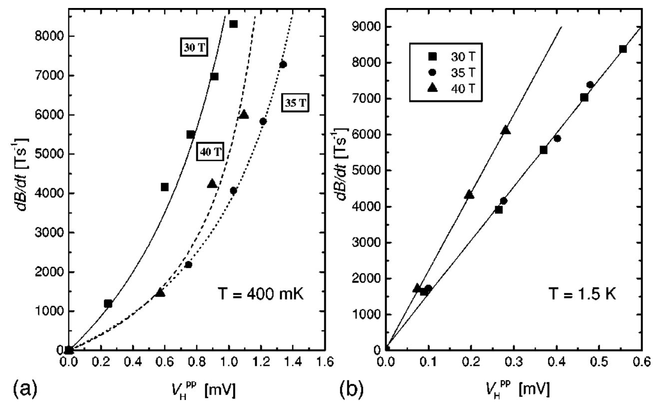

FIG. 3. (a) Peak-to-peak size $V_{\mathrm{H}}^{\mathrm{pp}}$ of the oscillation amplitude of V_{H} versus $\partial B/\partial t$ for different fields $B=30$ T, 35 T, 40 T and $T=400$ mK; the curves, which are intended only as guides to the eye, are fits to the data of the function $V_{\mathrm{H}}^{\mathrm{pp}}=(\gamma(\partial B/\partial t)V_{\mathrm{H}}^{\mathrm{lim}})/(\gamma(\partial B/\partial t)+V_{\mathrm{H}}^{\mathrm{lim}})$, which has the correct limiting behavior for $(\partial B/\partial t)\rightarrow 0,\infty$. Here $V_{\mathrm{H}}^{\mathrm{lim}}$, which represents the limiting value of V_{H} caused by the current saturation, and γ are fit parameters. (b) The same for $T=1.5$ K, this time using linear fits of the form $V_{\mathrm{H}}^{\mathrm{pp}}=\gamma(\partial B/\partial t)$.

The I-V characteristics of the in-plane resistivity in the pulsed-field measurement can be deduced by plotting V_{H} versus $\partial B/\partial t$ at a constant field, taking into account that the induced voltage at distance r from the center will be $V=(r/2)(\partial B/\partial t)$ and that $I=V_{\mathrm{H}}/\rho_{xy}$ (ρ_{xy} is constant at a particular field and temperature). Figure 3 shows such a plot for the temperatures $T=400$ mK and 1.5 K. In order to remove any systematic errors in subtracting the background induced voltage due to the open loop area between the wires, the peak-to-peak size $V_{\mathrm{H}}^{\mathrm{pp}}$ of the oscillations in V_{H} has been measured. Data are shown in Fig. 3 for fields close to $B=30$ T, 35 T, and 40 T; the sweep rate $\partial B/\partial t$ was varied by changing the voltage to which the capacitor bank was charged.[16] For $T=400$ mK, we find that I is proportional to $\partial B/\partial t$ at low magnetic fields, i.e., the sample behaves in an ohmic fashion. Within the QHE regime, however, this dependence becomes nonlinear, indicating a saturation of the induced current at a critical value [Fig. 3(a)]. This saturation will restrict the size of V_{H}, explaining why $V_{\mathrm{H}}/(a^2\partial B/\partial t)$ derived from pulsed-field measurements is smaller than the ratio $\rho_{xy}/\rho_{\parallel}$ obtained from steady-field data (see Fig. 2). In the quasistatic-field experiment, only the externally applied current (~ 10 μA) is flowing in the sample; this is typically several orders of magnitude smaller than the critical current.[9]

The current saturation is thought to be due to mechanisms such as Zener tunneling between Landau levels at the sample edges.[10,23] This leads to a dissipation of the induced current and thus to breakdown of the QHE.[9,10] The maximum Hall potential energy drop eV_{H} between the center and the perimeter of the sample is therefore limited by the Landau-level spacing $\hbar\omega_{\mathrm{c}}$. Figure 1(a) shows that between ~ 27 T and 37 T, the values of the peaks of V_{H} coincide with the cyclotron energy[24] $\hbar\omega_{\mathrm{c}}$ divided by e. This is in agreement with estimates of the peak circulating current made in magnetization experiments.[9,10]

The effects of the extreme nature of the electronic DOS (almost δ-function-like Q2D Landau levels with a smooth background of Q1D states) will be weakened by increasing the temperature; the consequent broadening of the Fermi-Dirac distribution function around μ will allow electrons to access some of the Q2D states, even when μ is between Landau levels.[4] Thus, the minima in $\rho_{\parallel}$ will be much more shallow,[6,10] greatly restricting the peak current that can flow in the sample. Hence, at 1.5 K, the peak-to-peak size of the oscillations in V_{H} is reduced[26,27] and the sample's I-V characteristics are apparently ohmic, with linear fits extrapolating through the origin [Fig. 3(b)].

An effect that will severely change the form of the electronic DOS is the onset of magnetic breakdown (MB) between the Q1D and Q2D sections of the FS (see inset to Fig. 2).[28] MB has been measured in the high-field phase of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ in both magnetization[28,29] and steady-field transport experiments,[25] revealing a breakdown field of $B_0\approx(70\pm 10)$ T. A similar value can be assumed for the $M=$Tl compound.[29,30] This has profound consequences for the likelihood of the QHE at high magnetic fields. MB provides a mechanism whereby carriers from the Q1D sheets of the FS can tunnel through to the Q2D sections and perform closed orbits, allowing them to contribute to the Hall conductivity. It also introduces Q2D structure in the DOS between Landau levels.[31,32] Therefore, both the deep minima in $\rho_{\parallel}$ and the plateaux in ρ_{xy} will be removed, destroying the QHE. Assuming B_0 to be ~ 70 T, the breakdown probability $e^{-B_0/B}$ increases[28] from $\sim 6\%$ at 25 T to $\sim 21\%$ at 45 T. The increasing importance of MB is the most likely cause of the tendency of the peak Hall potential energy to fall below $\hbar\omega_{\mathrm{c}}$ as the field increases beyond ~ 37 T. This is confirmed by the fact that the greatest nonlinearity in the I-V characteristics is seen at $B=35$ T, with the values at $B=40$ T showing a smaller current and slightly more ohmic behavior [Fig. 3(a)].

We therefore conclude that the QHE in α-(BEDT-TTF)$_2M$Hg(SCN)$_4$, $M=$K, Tl, is only possible over a restricted range of magnetic field and by a fortuitous set of circumstances. Owing to the persistence of vestiges of their low-field states,[3,7] these materials only attain the electronic DOS required to observe the QHE above $B\sim 27$ T ($M=$K) and 30 T ($M=$Tl). However, the increasing probability of MB will start to destroy the effect above $B\sim 37$ T. Hence, there is only a small field region over which plateaux in the Hall resitivity ρ_{xy} can be observed.

In summary, we have used a variant of the Corbino geometry in pulsed magnetic fields to obtain measurements of quantum oscillations of the Hall potential in a crystal of α-(BEDT-TTF)$_2$TlHg(SCN)$_4$. This enables the in-plane components of the resistivity tensor and the nonlinear behavior of the I-V characteristics within the quantum Hall regime to be studied. This can be used to establish the range of field over which the QHE is realized in α-phase BEDT-TTF charge-transfer salts.

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[1] BEDT-TTF is bis(ethylenedithio)tetrathiafulvalene or ET.

[2] A.A. House *et al.*, J. Phys.: Condens. Matter **8**, 8829 (1996); **8**, 10 361 (1996); **8**, 10 377 (1996).

[3] N. Harrison *et al.*, Phys. Rev. B **52**, 5584 (1995).

[4] N. Harrison *et al.*, Phys. Rev. B **54**, 9977 (1996).

[5] In the definition of the components of the resistivity tensor, the x and y directions are taken to be within the Q2D planes; z is in the interplane direction.

[6] N. Harrison *et al.*, J. Phys.: Condens. Matter **9**, L47 (1997).

[7] N. Harrison *et al.*, Phys. Rev. B **55**, R16 005 (1997).

[8] John Singleton, Phys. Mag. **19**, 195 (1997).

[9] N. Harrison *et al.*, Phys. Rev. Lett. **77**, 1576 (1996).

[10] M.M. Honold *et al.*, J. Phys.: Condens. Matter **9**, L533 (1997).

[11] S. Hill *et al.*, Phys. Rev. B **55**, R4891 (1997).

[12] B.I. Halperin, Phys. Rev. B **25**, 2185 (1982); R.F. Kazarinov and S. Luryi, *ibid.* **25**, 7626 (1982).

[13] The circulating current occurs because of Maxwell's third equation, $\nabla\times\mathbf{E}=-(\partial\mathbf{B}/\partial t)$.

[14] L.I. Buravov *et al.*, J. Phys. I **4**, 441 (1994).

[15] J.T. Chalker and A. Dohmen, Phys. Rev. Lett. **75**, 4496 (1995); L. Balents and M.P.A. Fisher, *ibid.* **76**, 2782 (1996).

[16] J.E. Crow *et al.*, Physica B **211**, 30 (1995).

[17] Due to its inductive nature, the size of the sample current is determined by the added serial resistance experienced around a closed loop. The spatial anisotropy of the in-plane resistivity due to the contribution of the Q1D states therefore does not introduce a dependence of the measured Hall voltage on the exact positioning of the edge contact.

[18] M.V. Kartsovnik *et al.*, J. Phys. I **4**, 159 (1994).

[19] T. Osada *et al.*, Phys. Rev. B **41**, 5428 (1990).

[20] Detailed de Haas–van Alphen studies of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ have shown that this salt is only fully transformed into the high-field state at fields of $\sim$3–4 T above its kink transition (Ref. 3). Assuming the same transition interval for α-(BEDT-TTF)$_2$TlHg(SCN)$_4$, we can conclude that the high-field state is fully established above $\sim$30 T.

[21] Surface conduction only bypasses bulk conduction at fields where μ lies *between* two Landau levels. At these fields, the lifetime of the edge states is greatly enhanced due to the nonavailability of states in the interior of the sample into which they can scatter. This leads to a suppression of the maxima in ρ_{zz} on lowering the temperature and to an eventual reversal of the oscillation phase (Ref. 10).

[22] The techniques described below can be used to show that the I-V characteristics of the sample are ohmic at low magnetic fields, justifying this renormalization.

[23] V. Nikos Nicopoulos and S.A. Trugman, Phys. Rev. Lett. **65**, 779 (1990); S. Komiyama *et al.*, Phys. Rev. B **45**, 11 085 (1992).

[24] We have assumed an effective mass of $m_\alpha^*=2m_e$ for the fundamental frequency of the Q2D hole pocket (Refs. 2, 10, and 25).

[25] M.M. Honold, D.Phil. thesis, University of Oxford, 1999.

[26] An equivalent way of considering the restoration of ohmic behavior in Fig. 3(b) is to recall that the mechanism associated with the saturation of the circulating current depends on the sample being within the quantum Hall regime (Refs. 9 and 10). An increase in temperature will lead to a degradation of the QHE (Ref. 27). The proportionality of I and V at $T=1.5$ K suggests that the QHE is already quenched at this temperature [Fig. 3(b)].

[27] *The Quantum Hall Effect*, edited by R.E. Prange and S.M. Girvin (Springer-Verlag, New York, 1990).

[28] M.M. Honold *et al.*, Phys. Rev. B **58**, 7560 (1998).

[29] Observations of the MB β frequency in pulsed magnetic fields have so far been restricted to inductive measurements of the susceptibility, carried out in the $M=$K salt (Ref. 28). Susceptibility measurements are far more sensitive with respect to the detection of high frequency oscillations than transport measurements, owing to the direct proportionality of their oscillation amplitude to the square of the frequency (Ref. 3). In the low-field state, the β frequency has already been observed in both the $M=$K and Tl salts (Ref. 30).

[30] S. Uji *et al.*, Phys. Rev. B **54**, 9332 (1996).

[31] Eventually, MB will cause the electronic DOS to become completely Q2D in nature, but with a much more complex structure containing multiple small gaps (Ref. 32).

[32] N. Harrison *et al.*, J. Phys.: Condens. Matter **8**, 5415 (1996).