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LETTER TO THE EDITOR

The importance of edge states in the quantum Hall regime of the organic conductor α-(BEDT–TTF)$_2$KHg(SCN)$_4$

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Abstract. We report measurements of the longitudinal magnetoresistance ρ_{zz} and magnetization of α-(BEDT–TTF)$_2$KHg(SCN)$_4$ in pulsed magnetic fields of up to 50 T and temperatures down to 400 mK, using samples of different purity. Below 2 K the amplitude of the Shubnikov–de Haas oscillations in ρ_{zz} is found to decrease dramatically with *falling* temperature. This effect is shown to coincide with quasipersistent eddy current resonances in the magnetization, which are a signature of the quantum Hall effect. Evidence is provided for the existence of a novel interplane conduction mechanism involving highly metallic edge states with supressed scattering at the surface of the sample (a so-called 'chiral Fermi liquid'), operational when the chemical potential is between Landau levels in the bulk of the material.

The charge-transfer salts α-(BEDT–TTF)$_2$MHg(SCN)$_4$ (M = K or Tl) have been the subject of much recent work [1, 2]. At magnetic fields above ~25 T, these materials are believed to possess a Fermi surface comprising a slightly warped quasi-two-dimensional (Q2D) hole cylinder plus a pair of quasi-one-dimensional (Q1D) electron sheets [2]. In magnetic fields $B \geqslant 30$ T applied perpendicular to the Q2D planes and at temperatures $T \lesssim 2$ K, the Landau level density of states (DOS) associated with the hole cylinder consists of very sharply resolved peaks; the Landau level spacing at 30 T is $\hbar\omega_c \approx 1.4$ meV, whereas the bandwidth W in the direction perpendicular to the Q2D planes is $W \approx 0.2$ meV and the scattering rate $\hbar/\tau$ is $\leqslant$0.1 meV in the highest-quality samples [3]. In contrast, the Q1D sheets cannot undergo Landau quantization; hence their contribution to the DOS is a continuous function of energy E. This means that the chemical potential μ oscillates at high magnetic fields [3]; close to half-integer values of F/B, where F is the de Haas–van Alphen (dHvA) frequency, μ is pinned to a Landau level, rising with it as B increases [3]; away from these values, μ remains within the Q1D component of the DOS between Landau levels over relatively extended regions of B. This behaviour of μ has a very dramatic effect on the physical properties of the α-(BEDT–TTF)$_2$MHg(SCN)$_4$ salts, particularly when μ is *between* Landau levels; at these fields the resistivity component [4] ρ_{zz} becomes very large [3], quantized Hall plateaux [5, 6] are observed in ρ_{xy} and the average in-plane resistivity $\rho_{||} \approx \frac{1}{2}(\rho_{xx} + \rho_{yy})$ becomes sufficiently small for quasipersistent induced currents to flow [5, 7].

In this high-field, low-temperature regime, the Lifshitz–Kosevich formalism used to treat magnetic quantum oscillations in conventional metals is inappropriate [3]; nevertheless, with proper attention to the oscillatory behaviour of μ, numerical models can be used to predict experimental dHvA and Shubnikov–de Haas (SdH) oscillations reliably [3]. However, a number of authors [8] have presented examples of resistivity data for α-(BEDT–TTF)$_2$MHg(SCN)$_4$ (M = K, Tl) which show very anomalous behaviour, in some instances reporting SdH oscillations which *decrease* in amplitude with decreasing temperature, contrary to all expectations [3]. The most recent work of this kind [9] attributes such behaviour to the presence of the quantum Hall effect (QHE), although the connection was only notional. In this letter, we report the first systematic investigation of this anomalous behaviour. Our data strongly suggest that the apparent collapse of the SdH oscillations can be attributed to highly metallic states at the sample surfaces.

Measurements were performed on single crystals of α-(BEDT–TTF)$_2$KHg(SCN)$_4$ in pulsed magnetic fields of up to 50 T at Leuven [10] and NHMFL (LANL) [11]; the samples used were from the same batch as those in [2]. Sample temperatures between 0.4 and 10 K were provided using ^{3}He and ^{4}He cryostats. The magnetoresistance was measured using alternating currents ranging from 200 nA to 5 mA at frequencies from 100 kHz to 1 MHz in order to ensure that the effects described below were not artefacts of the measurement technique or due to sample heating; alternating current transport techniques are used in pulsed fields to avoid mechanical and electrical problems [12]. The magnetic susceptibility was measured using an inductive method described elsewhere [13]. When necessary, adhesive was applied to the samples in order to prevent them from moving during the magnetic field pulse. Eddy current heating of the sample was not found to be a significant factor, as was verified [13] by applying different field sweep rates $\partial B/\partial t$.

Figure 1(a) displays the four-wire resistance of two crystals of α-(BEDT–TTF)$_2$KHg(SCN)$_4$ (samples A and B); in this measurement the current has been applied between contacts on the top and bottom (large) faces of the crystals (i.e., in the interplane z direction, perpendicular to the highly conducting Q2D planes [2]). The voltage was also measured using contacts on opposite faces, so that the measured resistance [5, 14] is proportional to ρ_{zz}. Note that the samples enter a spin-density-wave ground state below $\approx$25 T, resulting in a hump in the resistance (the so-called 'kink') and a change in the character of the oscillations [2]. Above this field, the anomalous behaviour [9] is immediately apparent in the raw resistance data. At temperatures between 4 K and 2 K, the SdH oscillations in ρ_{zz} increase in amplitude with decreasing temperature, roughly as expected; however, below 2 K, the oscillation amplitude *falls* with decreasing temperature, the effect being more acute in sample B.

Figure 1(b) shows the pick-up coil voltage detected in the inductive measurements of the susceptibility carried out on the same samples. In the high-field state above 25 T, the signals appear to consist of two oscillatory components with the same frequency, a series of broad oscillations which reverse sign between upsweeps and downsweeps of the field and much sharper oscillations which do not change sign. The sharp oscillations have been observed previously [7], and occur when μ is between Landau levels; at such fields, $\rho_{\parallel}$ becomes very small [5, 6], enabling a changing magnetic field to induce a quasi-persistent (on the ms timescale of a pulsed-field experiment) circulating current, a so-called 'eddy-current resonance' [15]. These eddy-current resonances are a characteristic feature of the QHE [6, 7, 15], as deep minima in $\rho_{\parallel}$ are accompanied by QHE plateaux in ρ_{xy}. The voltage induced in the pick-up coil [13] is given by $v = \lambda(\partial M/\partial B)\partial B/\partial t$, where λ is a constant depending on the coil. In the case of the dHvA signal, $\partial M/\partial B$ is independent of the sign of $\partial B/\partial t$, whereas the induced currents will reverse sign when $\partial B/\partial t$ reverses. Thus, the signal due to the dHvA oscillations will be of the opposite sign in upsweeps and downsweeps of the field, whereas the component due to the eddy-current resonances

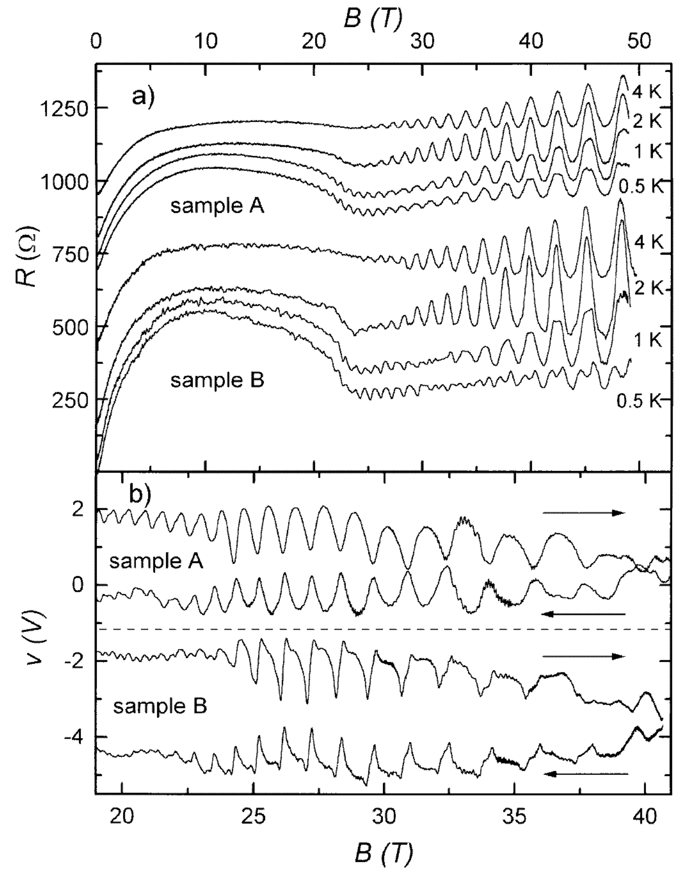

Figure 1. (a) Measured four-wire resistance (proportional to ρ_{zz}) of α-(BEDT–TTF)$_2$KHg(SCN)$_4$ samples A and B; the traces are offset for clarity. (b) Induced voltage from susceptibility measurements of the same samples at 500 mK; the arrows indicate the direction of sweep of the magnetic field. In both (a) and (b) the magnetic field is normal to the sample Q2D planes.

will always have the same sign [7]. Thus, by adding and subtracting the susceptibilities $\partial M/\partial B$ measured on the upsweeps and downsweeps of the magnetic field, the eddy-current and de Haas–van Alphen components respectively can be extracted from the signal [16]. The results of such a procedure are shown in figure 2 for samples A and B. Figure 2(a) shows that whilst eddy current resonances occur in both samples, they have a much greater prominence in sample B. In figure 2(b), the waveforms in figure 2(a) have been numerically integrated using Fourier decomposition/recomposition methods to obtain the constituent components of the magnetization. Whilst the dHvA component has a rounded saw-tooth form characteristic of Q2D metals [3], the eddy-current resonance magnetization is much more spiked in appearance [15]. The application of the numerical model of [3] allows the Dingle temperatures to be extracted from the dHvA data, revealing that sample B ($T_D = 0.4(2)$ K) is of higher purity than sample A ($T_D = 1.7(1)$ K).

Returning to the behaviour of ρ_{zz} in figure 1, it therefore appears that the stronger anomalous effect occurs in the higher-quality sample B, which exhibits more prominent features due to the QHE. We shall now examine the behaviour of ρ_{zz} in more detail.

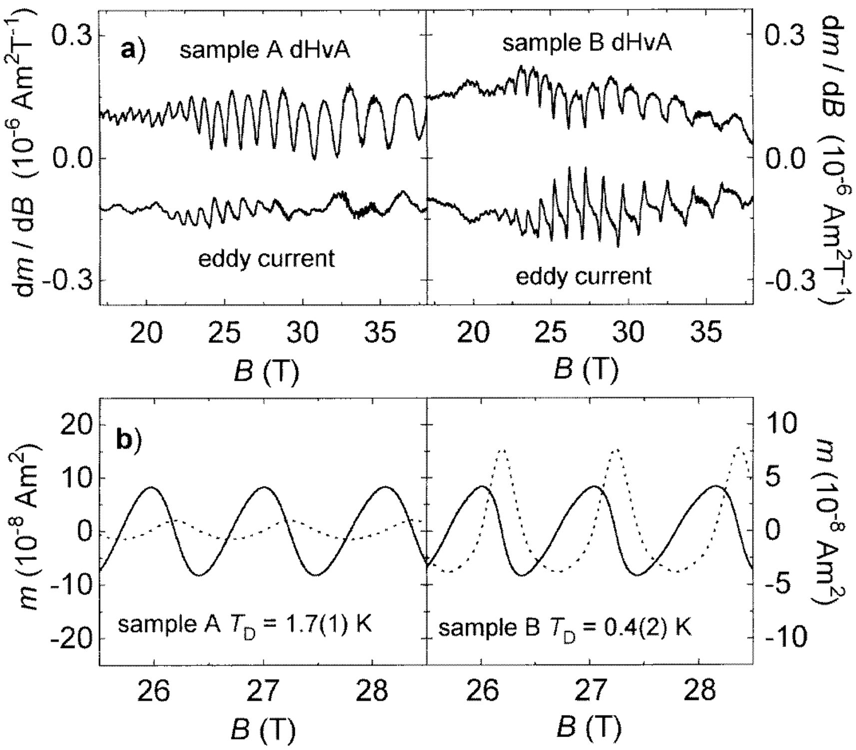

Figure 2. (a) Measured susceptibility $\partial M/\partial B$ separated into de Haas–van Alphen and eddy-current contributions for samples A and B at a temperature of 500 mK. (b) Magnetization of samples A and B, showing de Haas–van Alphen (solid curves) and eddy-current (dashed curves) contributions.

Figure 3(a) shows ρ_{zz} for sample B with the normal to the Q2D planes tilted by 20° with respect to the field. Although tilting the sample changes the form of the background (i.e. nonoscillatory) magnetoresistance [2], below 2 K, the SdH oscillation amplitude still falls with decreasing temperature, suggesting that the anomalous behaviour of ρ_{zz} is associated solely with the Landau quantization. Figure 3(b) shows values of ρ_{zz} at the peak (circles) and minimum (squares) nearest to 40 T as a function of temperature for sample B, compared to their expected variation (solid lines) deduced from numerical calculations known to predict the behaviour of ρ_{zz} reliably [3]. Note that whereas the ρ_{zz} minimum follows the expected curve, the maximum behaves very differently. Maxima in ρ_{zz} occur when μ is between Landau levels, i.e. at the same fields as the eddy-current resonances and QHE plateaux.

Figures 3(c) and 3(d) again demonstrate that the amount of attenuation of the ρ_{zz} maxima is associated with the strength of the manifestation of the QHE. In sample B (figure 3(d)), which shows stronger eddy-current resonances (figure 2), the ρ_{zz} oscillation amplitude can become *negative* at the lowest temperatures, i.e. the 'peaks' actually become lower than the 'dips', over certain ranges of field (e.g. between 25 T and 32 T in figure 1(a)). In contrast, sample A (figure 3(c)), which exhibits weaker eddy-current resonances (figure 2(a)), shows a smaller, though still considerable, deviation from the expected behaviour. Finally, figures 3(c) and 3(d) also show the amplitudes of the dHvA oscillations from the two samples, which vary with temperature as expected. This shows that the attenuation of ρ_{zz}

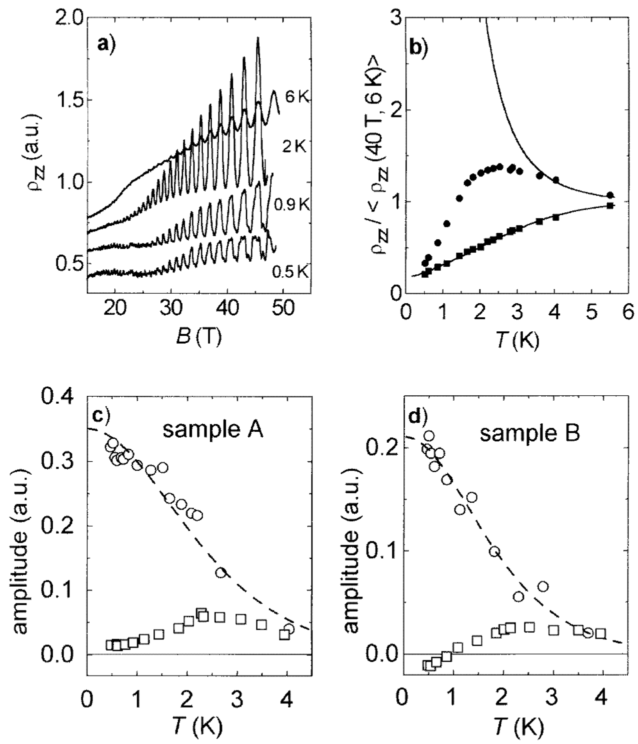

Figure 3. (a) The resistivity component ρ_{zz} of α-(BEDT–TTF)$_2$KHg(SCN)$_4$ sample B; in this figure the normal to the Q2D plane makes an angle of 20° with the magnetic field; data are offset for clarity. (b) The value of ρ_{zz} at the resistivity peak (circles) and minimum (squares) closest to 40 T for sample B as a function of temperature; the data are normalized to the 6 K value of ρ_{zz} at 40 T and the curves are model predictions. (c, d) Fourier amplitudes of the oscillations in ρ_{zz} (squares) and the de Haas–van Alphen oscillations (circles) for samples A (c) and B (d) with the magnetic field perpendicular to the Q2D planes. The Fourier transforms were carried out in the field range of 24 T to 40 T. The dashed curve is a model prediction.

oscillations is purely a magnetotransport phenomenon, and not due to a change in state of the samples, such as a phase transition.

The data in figure 3 illustrate the care which must be taken in interpreting experimental resistivity results. In [9], only the modulus of the Fourier amplitude of the SdH oscillations was plotted; this was observed to go through a minimum at ∼1 K, which was interpreted as resulting from competition between two sets of SdH oscillations with differing phase. However, the data in figure 3 show that it is only the peaks in ρ_{zz} that are strongly affected. If due attention is paid to the phase information in a Fourier transform, this results in the amplitude of the oscillations passing *through* zero to become negative (figure 3(d)).

A well known mechanism which causes anomalous behaviour of the measured resistivity in anisotropic materials is ‘current jetting’ [18]. Figure 4(a) contrasts two- and four-contact measurements of ρ_{zz} (a two-contact measurement involves using common current and voltage contacts) made using contacts on opposite faces of the crystal [19]. Both two- and four-contact measurements are very similar, showing that ‘current jetting’, which only affects four-contact measurements, cannot be responsible for the anomalous behaviour of ρ_{zz}. As the unexpected behaviour of ρ_{zz} occurs only at the points at which μ is between

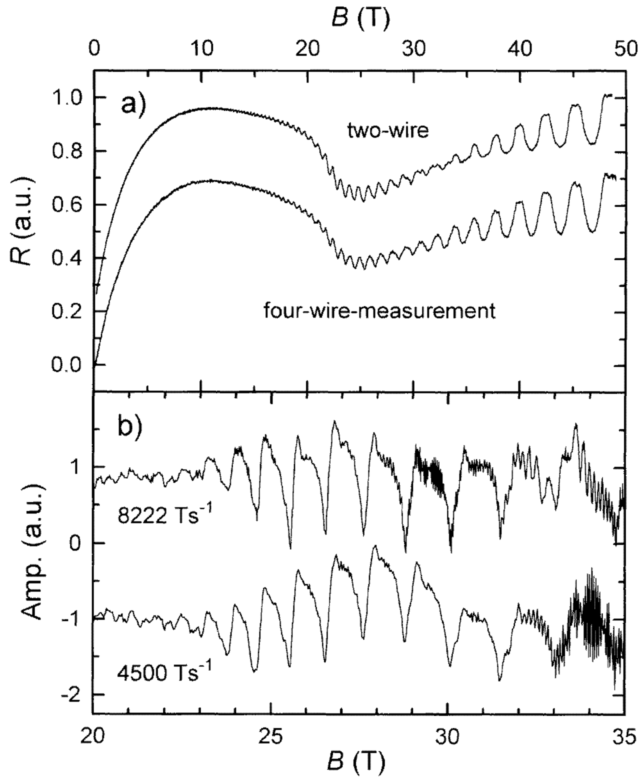

Figure 4. (a) Measured two-wire and four-wire resistances of sample A at 500 mK. The four-wire data are the same as those in figure 1. (b) Susceptibility measurements at 500 mK for two different field sweep rates $\partial B/\partial t$. Note that the eddy-current resonances are virtually unchanged in size, indicating that the induced current has saturated. In both (a) and (b) the magnetic field is normal to the sample Q2D planes.

Landau levels, it appears that some mechanism which 'shorts out' the conventional current paths in the interplane z direction operates at these particular fields. It has been proposed [20] that the conductivity of two-dimensional (2D) systems which exhibit the QHE is strongly affected under such conditions by the presence of metallic states at the sample edges, formed by the bending up of Landau levels [21] so that they cross μ. We shall now present data which indicate that such edge states are important in the BEDT–TTF salts.

Evidence for the involvement of states at the sample edges comes from the manner in which the current flowing in the eddy current resonances saturates in α-(BEDT–TTF)$_2$KHg(SCN)$_4$. Figure 4(b) shows $\partial M/\partial B$ for sample B at two different field sweep rates $\partial B/\partial t$; note that the eddy-current resonances are virtually unchanged in size. If it is assumed that the current flows chiefly around the sample edges, the saturated value can be estimated [7] from the peak-to-peak amplitude of the eddy-current resonances and the sample area (0.7 mm^3) to be $I \approx 0.16$ A, or 1.8 μA per layer. This is similar to the value observed in α-(BEDT–TTF)$_2$TlHg(SCN)$_4$, which was found to be within a few per cent of that expected for the breakdown of the QHE [7].

Although the observed saturated current is close to theoretical expectations, the current limiting mechanism in BEDT–TTF salts must be rather different from those applied to

2D semiconductor systems, which require large Hall electric fields E_H in order for the QHE to break down. For example, for inter-Landau-level Zener transitions [22] to occur, $E_H \gtrsim \hbar\omega_c/l_B L^{\frac{1}{2}}$, where l_B is the magnetic length and L is the Landau level index. In α-(BEDT–TTF)$_2$KHg(SCN)$_4$ at 40 T this would require $E_H \sim 10^5$ V m^{-1}. However, if one uses the saturation current $I \sim 1.8$ μA estimated above with the well known quantized Hall resistance [6] to derive the corresponding Hall voltage V_H, one obtains $V_H \sim 1$ mV. In a typically sized sample (diameter $\sim$ 0.5–1 mm), this implies that the average value of E_H is $\sim$2 V m^{-1}, far too small to cause Zener transitions. Similarly, spontaneous phonon emission [23] requires that the drift velocity E_H/B of the centres of the cyclotron orbits be greater than the speed of sound in the material. Substituting the average value of E_H ($\sim$2 V m^{-1}) and $B \sim 40$ T shows that this is impossible.

There are two (related) ways of connecting the observed current saturation with states at the edges of the sample; the first involves considering the type of scattering event which can lead to the breakdown of the QHE. Models based on so-called ‘extended scattering centres’ (hard-wall scatterers) [24], the size of which exceeds the cyclotron radius, have been shown to require much smaller electric fields than than those required for Zener breakdown or phonon emission. However, the *highly metallic* nature of the BEDT–TTF salts [3] means that such large-scale scattering potential variations can only exist at the edges of the samples. Inter-edge-state scattering is known to lead to a re-equilibration of the population of the edge states [25]; the maximum extent to which the edge states can become unequally populated due to such a process is $\frac{1}{2}\hbar\omega_c$. This is about 1 meV in the α-phase BEDT–TTF salts at 40 T, which compares favourably with the total breakdown Hall voltage measured in α-(BEDT–TTF)$_2$KHg(SCN)$_4$ ($\sim$1 mV, this work) or in α-(BEDT–TTF)$_2$TlHg(SCN)$_4$ ($\sim$0.5 mV [7]).

The second indication of the importance of edge states comes from estimates of the Landau level curvature at the sample surfaces. BEDT–TTF salts are three-dimensional crystallographic systems. In a magnetic field applied perpendicular to the conducting Q2D planes, the surface states will therefore extend over all of the edge surfaces of the sample, forming a so-called chiral Fermi liquid [20]. However, the interplane bandwidth is small compared to the Landau level spacing [3] at $\sim$40 T, so that the bending of the Landau levels at the edges of BEDT–TTF salt samples will be very similar to that that in the purely 2D case [26, 27]; e.g. in the case of a hard-wall edge potential [26], the Lth Landau level will be elevated to an energy $\epsilon \approx \hbar\omega_c(2L + \frac{3}{2})$ at the surface (cf $\epsilon = \hbar\omega_c(L + \frac{1}{2})$ within the bulk of the sample). As the surface states are confined within a distance $x \approx l_B L^{\frac{1}{2}}$ of the sample edge, this results in an effective edge electric field of $(1/e)\,\partial\epsilon/\partial x \sim \hbar\omega_c(L+1)/(l_B L^{\frac{1}{2}}) \sim 2\times 10^6$ V m^{-1} at 40 T. Hence, this edge electric field is much greater than the Hall field in BEDT–TTF salts, even in the case of pulsed magnetic fields. This is in marked contrast to the situation in most 2D semiconductor systems, where the breakdown of the QHE occurs in a regime where the Hall potential dominates [27] (i.e. $eV_H \gg \hbar\omega_c$), and again suggests that the edge states play a much more important role in BEDT–TTF salts.

When μ is in the gaps between Landau levels, the bulk resistivity components are strongly modified, with ρ_{xx} tending to small values [7] (such that the eddy-current resonances can occur) and ρ_{zz} becoming very large due to the virtual vanishing of the interplane component of the electronic velocity [3]. However, at the edges of the sample, Landau levels will cross μ, providing a surface metallic region which effectively shorts out the bulk of the sample; the existence of this region has already been shown to be important in the breakdown of the QHE. However, the fact that the surface states are confined within a distance $l_B L^{\frac{1}{2}}$ of the sample edge implies that they only occupy a fraction $f \sim 10^{-4}$

of a typical sample cross section. In order to account for the observed attenuation of the ρ_{zz} peaks in figure 1 the longitudinal edge-state conductivity must be some 10^6 times the typical bulk conductivity. It has been argued that the chiral nature of the edge states strongly inhibits backscattering events, so that charged impurities no longer impede the flow of edge currents [21].

If we assume that the interplane transport is dominated by surface effects at low temperatures when μ is between Landau levels, then ρ_{zz} at such fields provides a measure of the edge-state resistivity. The linear dependence of the peak height (see especially the data for sample B) on temperature therefore implies that $\rho \; \alpha \; T^1$, suggestive of non-conventional Fermi liquid behaviour (the expectation [18] would be T^2). Using the Drude relationship $\sigma_{zz} \sim ne^2\tau/m_z^*$, where $n \sim 1.6 \times 10^{26}$ m^{-3} is the electron density, τ^{-1} is the effective scattering rate and m_z^* is the interplane effective mass ($\sim 10^3$ times the in-plane effective mass owing to the high degree of anisotropy [3]), one obtains $\hbar/\tau \sim k_{\mathrm{B}}T$.

In summary, a suppression of the SdH oscillations in the resistivity component ρ_{zz} with *decreasing* temperature is observed in samples of the charge-transfer salt α-(BEDT–TTF)$_2$KHg(SCN)$_4$ which display manifestations of the QHE. The degree of suppression of the SdH oscillations has been unambigously linked with the strength of the induced eddy-current resonances in the same samples. We have shown that an earlier tentative explanation for the effect [9] involving a competition between two independent sets of oscillations is untenable. Instead, it is only the peaks of the oscillations in ρ_{zz} which are affected; the minima follow the predicted behaviour. In very high-quality samples this eventually leads to a phase inversion of the oscillations whereby the resistivity 'maxima' fall below the 'minima' in ρ_{zz}. This demonstrates the existence of a mechanism which bypasses conventional interplane current paths whenever the chemical potential lies between Landau levels. Studies of the breakdown of the QHE in the same samples suggest that highly metallic surface states, constituting a chiral Fermi liquid, assume an important role in the conduction at these fields and are probably responsible for the anomalous behaviour of ρ_{zz}.

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