Critical state in a low-dimensional metal induced by strong magnetic fields

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We present the results of magnetotransport and magnetic torque measurements on the α-(BEDT-TTF)$_2$KHg(SCN)$_4$ charge-transfer salt within its high-magnetic-field phase, in magnetic fields extending to 33 T and temperatures as low as 27 mK. While the experimentally determined phase diagram closely resembles that predicted by theoretical models for charge-density waves in strong magnetic fields, the phase that occurs at fields above $\sim$23 T, which is expected to be either a modulated charge-density wave phase or a charge/spin-density wave hybrid, exhibits unusual physical properties that are most atypical of a density wave ground state. Notably, the resistivity undergoes a dramatic drop below $\sim$3 K within this phase, falling in an approximately exponential fashion at low temperatures, while the magnetic torque undergoes extensive hysteresis. This hysteresis, which occurs over a broad range of fields and gives rise to a large negative differential susceptibility $\partial M/\partial B$ on reversing the direction of sweep of the magnetic field, is strongly temperature dependent and also has several of the physical characteristics predicted by critical-state models normally used to describe the pinning of vortices in type II superconductors. Such a behavior appears therefore only to be explained consistently in terms of persistent currents within the high-magnetic-field phase of α-(BEDT-TTF)$_2$KHg(SCN)$_4$, although the origin of these currents remains an open question.

I. INTRODUCTION

Of all the quasi-two-dimensional (Q2D) organic charge-transfer salts that exist,[1] those of the composition α-(BEDT-TTF)$_2M$Hg(SCN)$_4$, where BEDT-TTF stands for bis(ethylenedithio)tetrathiafulvalene and where M=K, Tl, Rb, or NH$_4$, have been, perhaps, the most difficult to understand.[2,3] While the M=NH$_4$ salt is a superconductor with a transition temperature $T_c\sim1$ K,[4] the M=K, Tl, and Rb salts all undergo a transition into a more magnetoresistive state with a reconstructed Fermi surface below $T_p\sim8$ (in the M=K and Tl salts) or 10 K (in the M=Rb salt).[2,3,5] The underlying physical reason for the behavioral differences between these isostructural M=NH$_4$ and M=K, Tl, and Rb salts remains a contemporary issue. The M=K salt has, nevertheless, been shown to become superconducting under uniaxial stress applied perpendicular to the conducting layers,[6,7] and it has further been suggested that the M=K and Rb salts could exhibit filamentary superconductivity at ambient pressure.[8–10]

Unquestionably, the main physical traits of the M=K, Tl, and Rb salts at ambient pressure, at moderately low temperatures $T\lesssim T_p$ and at magnetic fields $B<B_k$, where B_k ($\approx$23 T in the M=K salt) is known as the kink transition field,[11] are more typical of a density wave (DW) ground state.[5,12,13] Yet, no direct evidence for a superlattice structure has been found. Most of the more recent experimental and theoretical surveys point toward a charge-density wave (CDW) rather than a spin-density wave (SDW) being the more likely candidate.[14–17] The low-temperature ($T<T_p$) ground state is strongly magnetoresistive.[5,12,13,18,19] However, this behavior subsides rapidly as B_k is approached.[11] The nature of the high-magnetic-field phase ($B>B_k$) at low temperatures has since been the subject of speculation.[15–23] The only fully established fact is that the transition field B_k is distinctly first order, as evidenced by pronounced hysteresis effects at $\sim$23 T in many measured physical properties.

From a theoretical perspective, B_k lies remarkably close to the critical field B_c at which one would expect a CDW, with a transition temperature of 8 K, to approach the Pauli paramagnetic limit.[15,17] A first order transition could be expected,[15,17] with the material transforming at higher fields either into a normal metal,[17] a spatially modulated CDW,[15] or a CDW-SDW hybrid phase.[24] The latter two, in particular, represent the CDW analog of the Fulde-Ferrel phase that is anticipated to occur in type II superconductors with suppressed orbital effects over the equivalent region of the phase diagram.[25,26] If such a phase is actually realized at high magnetic fields, its physical properties have not yet been the subject of a thorough investigation. Experimentally, data have been published that could be considered consistent with the existence of a different thermodynamic phase above B_k at low temperatures,[13,18,27] albeit that the earlier magnetic data were discussed largely in the context of SDW's, with no reference to CDW's. Yet other data concerning the unusual behavior of the interlayer magnetotransport and induced currents in the magnetization in pulsed magnetic fields have been notionally connected with the quantum Hall effect (QHE).[21–23] While a definitive experiment has not been performed that can firmly establish either of these latter two scenarios at high magnetic fields ($B > B_{\mathrm{k}}$), one cannot dispute the fact that this high-magnetic-field phase is exotic.

With a view to understanding more about the high-magnetic-field phase (at fields above B_{k}), in the present paper we describe the results of extensive measurements of the interlayer magnetotransport and magnetic torque on several α-(BEDT-TTF)$_2$KHg(SCN)$_4$ crystals over a broad range of temperatures ($20\ \mathrm{mK} < T < 10\ \mathrm{K}$) and in continuous magnetic fields of up to 33 T. The experimentally determined phase diagram closely follows the theoretical models of McKenzie[15] and Zanchi *et al.*[24] However, the observed behavior of the magnetotransport and magnetic torque, at magnetic fields above $\sim 23\ \mathrm{T}$, is not typical of a DW system, nor does it appear to be compatible with recent ideas based on the QHE.[21–23] For example, as the sample is cooled in a magnetic field, the resistivity undergoes an abrupt drop at $\sim 3\ \mathrm{K}$, at all fields $B \gtrsim B_{\mathrm{k}}$, with the drop being particularly pronounced at integral Landau level filling factors, falling exponentially by roughly two orders of magnitude. The occurrence of a change in slope in the magnetic torque at precisely the same temperature, on field-cooling the sample, confirms the existence of a different low-temperature thermodynamic phase below $T_{\mathrm{c}} \lesssim 3\ \mathrm{K}$, as originally suggested by Kartsovnik *et al.*[18] At lower temperatures still (i.e., $T \lesssim 2\ \mathrm{K}$), the field dependence of the magnetic torque develops a pronounced hysteresis at all fields above B_{k}, which increases approximately exponentially with decreasing T, but with the hysteresis being particularly strong at half integral Landau level filling factors. Only at much lower temperatures ($T \sim 27\ \mathrm{mK}$) does the hysteresis become more pronounced at integral filling factors, whereby it comes into closer agreement with existing pulsed magnetic field experimental data.[21,23] While this magnetic hysteresis that persists throughout the entire high-magnetic-field phase is difficult to explain in terms of conventional DW ground states or the QHE, it has many of the features of a critical-state model like those used to explain magnetic hysteresis in type II superconductors.[28,29] In particular, the hysteresis is characterized by a large negative differential susceptibility $\partial M/\partial B \ll 0$ on reversing the direction of sweep of the magnetic field, with the width of the hysteresis loop eventually approaching a saturation limit that depends weakly on the rate of change of magnetic field but strongly on temperature. While there is no solid reason for expecting the low-temperature high-magnetic-field phase of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ to be superconducting, these results do appear to indicate that the currents previously reported to be induced within the conducting planes in pulsed magnetic fields[21,23] are, in fact, persistent (at least on the time scale of Bitter magnet experiments), and occur at arbitrary Landau level filling factors. In the absence of any existing theoretical model that can explain such effects, some speculative ideas based on CDW's are proposed in the concluding section of this paper.

II. EXPERIMENT

The samples chosen for this study, referred to hereafter as 1, 2, and 3, were grown using conventional electrochemical means,[30] while static magnetic fields extending to $\sim 33\ \mathrm{T}$ were provided by the National High Magnetic Field Laboratory, Tallahassee. Temperatures between $\sim 20\ \mathrm{mK}$ and $\sim 1.6\ \mathrm{K}$ were provided by a top-loading dilution refrigerator, while higher temperatures were obtained using a ^{3}He refrigerator. Magnetotransport measurements were made using standard four-wire techniques with low-frequency ($\sim 10\ \mathrm{Hz}$) currents of between 1 and 10 μA applied perpendicular to the conducting layers. For the magnetic torque measurements, the samples were mounted on the moving plate of a phosphor bronze capacitance cantilever torque magnetometer, which was itself attached to a rigid but rotatable platform in such a way that the axes of torque and rotation were parallel to each other, but perpendicular to the applied magnetic field B. The angle between B and the normal to the capacitance plates was approximately the same as the angle θ between B and the normal to the conducting planes of the sample. The capacitance of $C \sim 1.3\ \mathrm{pF}$ was measured by means of a ratio transformer energized at 5 kHz and 30 V (rms); the largest change in C observed throughout the experiment at the highest magnetic field was $\sim 0.04\ \mathrm{pF}$, corresponding to a net angular displacement of $\sim 0.1°$. Torque interaction effects at $\theta = 7°$ (the angle at which most of the measurements were made) were therefore not a significant factor. To eliminate possible artifacts due to time constants of the instrumentation setup or the data acquisition system, the capacitance was measured with the time constant of the lock-in amplifier set to a low value of 10 ms. Furthermore, both the output of the lock-in amplifier and the shunt voltage, which determines the current flowing in the Bitter magnet coils, were measured using a digitizer with a resolution of $\sim 50\ \mu$s, while the field was swept slowly at $\sim 8\ \mathrm{mT\,s^{-1}}$.

III. TEMPERATURE-DEPENDENCE OF THE MAGNETORESISTANCE

Examples of the interlayer magnetoresistance measured in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ sample 1 at selected temperatures are shown in Fig. 1. The magnetoresistance displays the usual behavior observed for this material, reaching a maximum at $\sim 10\ \mathrm{T}$ before falling again in a linear fashion as the kink transition field is approached.[11–13,30] As with these earlier studies, the oscillations within the low-magnetic-field DW phase exhibit a pronounced second harmonic. Above the kink transition, at temperatures higher than $\sim 3\ \mathrm{K}$, the oscillations grow rapidly in amplitude with increasing field. The behavior of the interlayer Shubnikov–de Haas (SdH) wave form over this range of temperatures is well understood in these materials.[31,32] At integral Landau level filling factors, when the chemical potential μ is situated in a Landau gap, the SdH maxima increase with decreasing temperature in an insulating-like fashion. Conversely, at half-integral filling factors, the resistivity of the minima behaves in a metallic fashion. This is entirely consistent with the theoretically predicted behavior of a quasi-two-dimensional metal in a magnetic field[31,32] and is further consistent with de Haas–

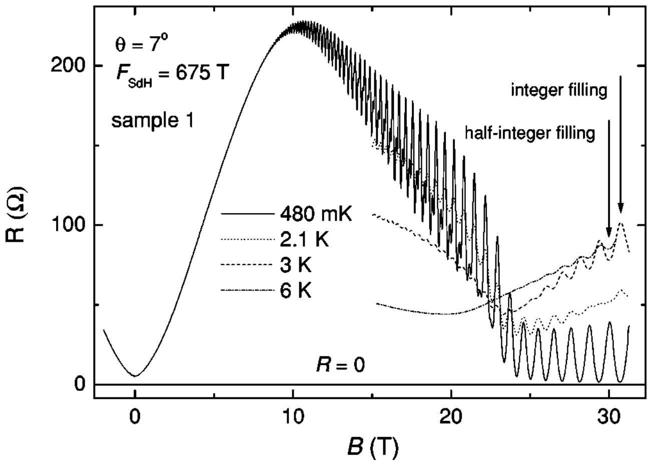

FIG. 1. The magnetoresistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ sample 1 at selected temperatures, with the SdH frequency being 675 T at $\theta = 7°$. The ''phase-inversion'' effect is particularly pronounced in this sample at fields above the kink transition field, with the inverted SdH minima having resistivity values $\sim$4 times lower than the minimum resistivity at zero field.

van Alphen (dHvA) measurements.

What is not predicted by the simple magnetotransport model is the abrupt inversion of the wave form at low temperatures, resulting in the high-temperature ($T \gtrsim 3$ K) SdH maxima becoming minima at low temperatures ($T \lesssim 3$ K), having a resistivity several times lower than that of the sample at zero field. Figure 1 does not constitute the first observation of this effect; previous studies had reported this effect in both continuous and pulsed magnetic fields.[22,23] Because no phase inversion is observed in the dHvA effect,[22,23] which is entirely a thermodynamic function of state, it must be concluded that the Landau level structure remains largely unchanged over the same temperature range. It was the occurrence of this phase inversion only in the magnetotransport that was attributed to the effect of a chiral Fermi liquid,[22,23] following a suggestion that the QHE may be taking place at high magnetic fields in this material.[21] Because the interlayer resistance of the bulk of the sample becomes insulator-like at integral filling factors, according to magnetotransport theory,[31,32] higher-conductivity chiral Fermi liquid states were proposed to take over the majority of the interlayer conductance. For this conjecture to explain the most recent data (i.e., Fig. 1), the interlayer conductivity of the edge states, which occupy $\sim$1 part in 10^4 of the sample cross section, would have to be strongly temperature dependent and also attain a conductivity at least 10^6 times higher than the field-averaged (or background) conductivity of the bulk.

Since this conjecture was made, the interlayer conductance of chiral surface states has been measured directly in semiconductor superlattices that at the same time quite definitively exhibit the QHE.[33,34] Not only is the conductance of these states found to be temperature independent, as predicted by chiral Fermi liquid theory,[35,36] but their net conductance is also observed not to be particularly high, resulting in only a weak suppression of the SdH maxima. This behavior is therefore quite different from that observed in α-(BEDT-TTF)$_2$KHg(SCN)$_4$. Given, also, that flat quantized Hall plateaus proportional to h/ie^2 have not been observed in any of the α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ salts,[19] the arguments involving chiral Fermi liquids in these salts become considerably weakened.[22,23]

A simpler pattern emerges when the resistivity, at both integral and half-integral filling factors, is plotted versus temperature in Fig. 2. For two different samples, the resistivity varies weakly with temperature for $T \gtrsim 3$ K, but then undergoes an abrupt drop for $T \lesssim 3$ K. This drop is particularly pronounced at integral filling factors (i.e., when μ is situated in the Landau gap of the Q2D pockets), but is also clearly discernible at half-integral filling factors (i.e., when μ is situated in the middle of a Landau level). This observation, again, appears to be incompatible with the notion of a chiral Fermi liquid,[22,23,35,36] which is expected to manifest itself only at integral filling factors. It could, however, be consistent with the interpretation of Kartsovnik *et al.* in terms of a new high-magnetic-field low-temperature phase.[18] In sample 1, at integral filling factors, the resistivity is observed to fall by as much as two orders of magnitude between 3 K and 490 mK, reaching 1.6 Ω at the lowest temperature. While 1.6 Ω

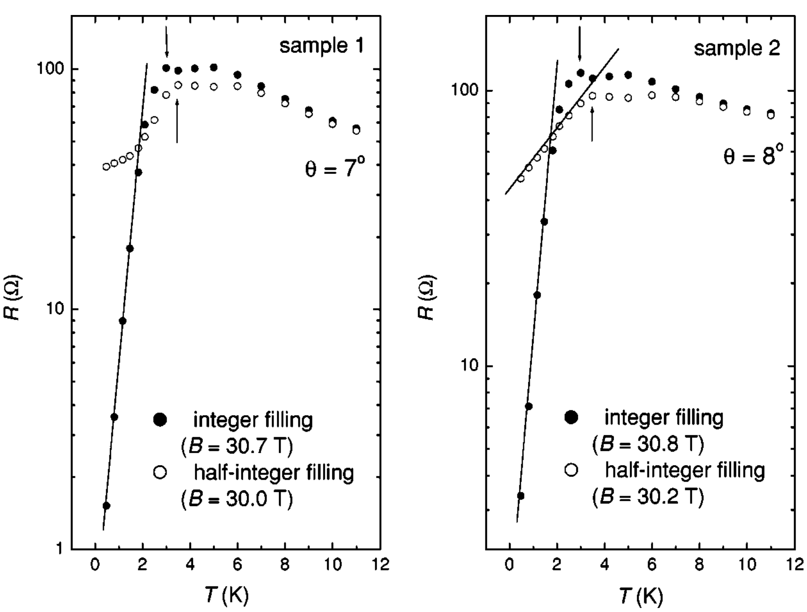

FIG. 2. The resistance versus temperature at both integral and half-integral filling factors for two different samples (sample 1 and sample 2) plotted on a logarithmic scale. In both samples, the resistance varies weakly with temperature until it drops abruptly at $\sim$3 K, falling in an exponential fashion. While the effect is more abrupt at integral filling factors, a sharp kink is also observed at half-integral filling factors. Arrows indicate where the different slopes of the resistivity intersect when plotted on a linear (rather than logarithmic) scale.

is not astonishingly low for a metal, it is measured in the least conducting direction of a compensated semimetal, which has an interplane:in-plane electrical bandwidth anisotropy of order $1:10^4$ to $1:10^5$, and is $\sim$2000 times lower than the room temperature value of 3.6 kΩ for this sample; this is even in the presence of an $\sim$30 T magnetic field.

If the low-temperature high-magnetic-field phase is a CDW, as predicted by mean field theory,[15,24] then the abrupt drop in resistivity on entry into this CDW phase is certainly without precedent.[37] Following a reduction in the number of carriers, transitions into DW ground states are invariably followed by an immediate increase in resistivity, or, more subtly, by a shoulder feature in the resistivity, should a significant fraction of the original Fermi surface survive the DW order.[37] The latter situation could apply to α-(BEDT-TTF$)_2$KHg(SCN$)_4$ (Ref. 5) within its low-magnetic-field DW phase, for which the continued presence of a Fermi surface causes the resistivity to retain a metallic behavior on cooling. Only by application of a magnetic field $B \lesssim 23$ T does the shoulder feature develop into a resistivity increase on entry into the low-magnetic-field phase.[5,13,16,18]

The change in the behavior of the resistivity on cooling α-(BEDT-TTF$)_2$KHg(SCN$)_4$ at high magnetic fields $B \gtrsim 23$ T is considerably different from that occurring within the low-magnetic-field DW phase. Phenomenalogically, the interlayer resistivity in Fig. 2, within the low-temperature high-magnetic-field phase, obeys an approximate $\rho \propto \exp(T/T_0)$ law, with $T_0 \sim 520$ mK at integral filling factors for both samples 1 and 2. It therefore follows that the resistivity remains finite, even on transferring the samples into a dilution refrigerator at temperatures as low as 20 mK at the same magnetic field. Clear trends can, however, be observed. In Fig. 1, for example, the resistivity minima become increasingly deep with increasing field, and it has also been noted that the depth of the minima is strongly sensitive to sample quality.[23] Such an exponential law is, nevertheless, quite unlike that of an ordinary metal. The only materials that exhibit qualitatively similar resistive drops followed by an approximate exponential law are filamentary or granular superconductors such as those of the form La_2CuO_{4-y},[38] or Ba-La-Cu-O.[39] It remains to be established whether this apparent similarity is anything more than superficial.

IV. TEMPERATURE DEPENDENCE OF THE MAGNETIC TORQUE

The temperature dependence of the magnetotransport alone cannot be considered as proof for a transition into a different thermodynamic phase. This usually requires measurement of a thermodynamic function of state, such as specific heat or magnetization. Specific heat measurements have thus far confirmed the existence of one second order transition into a low-temperature phase at low magnetic fields ($B < B_k$).[40] Within this same phase, magnetic moment measurements, made using a superconducting quantum interference device (SQUID) in fields of 5 T, have shown that the susceptibility drops most notably when the field is oriented within the conducting planes.[41] Such a behavior could be expected for either a CDW or a SDW phase following the net loss in Pauli paramagnetism accompanying the opening of gaps on the Fermi surface.[37] Only a very weak change, however, is observed when the magnetic field is oriented perpendicular to the conducting layers. Sasaki *et al.* notionally connected this anisotropy with antiferromagnetism associated with a SDW ground state,[41] although there has since not been any compelling evidence supporting this hypothesis.[14,42]

An alternative, yet rather trivial, explanation could be that, when the field is oriented perpendicular to the conducting layers, the loss in Pauli paramagnetism is balanced by a loss in Landau diamagnetism following the partial opening of gaps on the Q2D Fermi surface pockets due to a CDW. Gaps on the Q2D pocket are, after all, predicted by *all* models of the reconstructed Fermi surface.[43] Perhaps the strongest evidence for the latter explanation is that the magnetic susceptibility is completely isotropic for fields oriented within the planes, as demonstrated by the magnetic torque measurements of Christ *et al.*[44] This detail would be difficult to explain in terms of antiferromagnetism, but is quite easy to explain in terms of Landau diamagnetism, which, in a Q2D conductor, manifests itself only perpendicular to the conducting layers.

The essential advantage of magnetic torque measurements is that they are nominally sensitive only to the magnetic anisotropy, with the net torque being given by the product

$$\boldsymbol{\tau} = \mathbf{M} \times \mathbf{B}. \tag{1}$$

Thus, if we consider the susceptibility to be resolved into its components perpendicular to the conducting layers $\chi_\perp \equiv M_\perp / B$ and parallel to the conducting layers $\chi_\parallel \equiv M_\parallel / B$, the net magnetic torque is given by

$$\tau = \frac{1}{2}[\chi_\parallel - \chi_\perp] B^2 \sin 2\theta, \tag{2}$$

where θ is the angle between the magnetic field and the normal to the conducting layers (i.e., the **b** axis of the crystal). With the first term of Eq. (2) being dominant and negative, according to Sasaki *et al.*,[41] the change in anisotropy on entering the low-temperature DW phase exerts a net negative torque on the sample, which attempts to align the **b** axis of the sample more closely with B. This is exactly the signature observed by Christ *et al.*[44] For a constant anisotropy $\chi_\parallel - \chi_\perp$, the negative torque should increase in a manner that is proportional to B^2. The fact that this is the case only at low magnetic fields[45] could be considered as further supportive evidence for our explanation in terms of Landau diamagnetism, whereby the Landau diamagnetic contribution from the Q2D pocket returns gradually at higher magnetic fields due to magnetic breakdown across the CDW gaps.

Even though the anisotropy becomes reduced at high magnetic fields, a change in magnetic torque on field-cooling the sample is still visible in Fig. 3. This is perhaps assisted by the increasing sensitivity ($\propto B^2$) of torque magnetometry with field. While the reason for the continued anisotropy at high magnetic fields is not obvious, the abrupt change in slope in fields as high as $\sim$30.1 T clearly indicates the continuing presence of a thermodynamic phase boundary. The field of 30.1 T was chosen because, at this field, μ is exactly that for a half-integral filling factor, thereby eliminating any possible contribution to the torque from the dHvA signal.[18] If our interpretation of the magnetic measurements of Christ

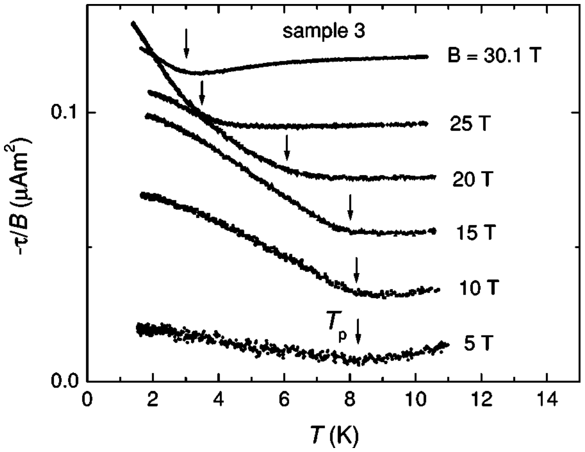

FIG. 3. Field-cooled magnetic torque measurements made at various different values of magnetic field for $\theta \sim 20°$, indicating that the change in slope continues to be discernible even at the highest available static magnetic fields. The torque has been normalized by $1/B$.

et al.[44,45] and Sasaki *et al.*[41] is correct, this could imply that the loss of Landau diamagnetism on entering the low-temperature, high-magnetic-field phase continues to be the dominant effect.

V. HYSTERESIS IN THE MAGNETIC TORQUE

One of the most intriguing aspects of this material, at high magnetic fields, is its magnetic hysteresis. An example of the magnetic torque of sample 3, measured in both rising and falling magnetic fields at 27 mK, is shown in Fig. 4.[46] Apart from the fact that the temperature is lower in the current work, the hysteresis that occurs within the high-magnetic-field phase is very similar to that observed by Christ *et al.*[47,48] Note, however, that at the time the original measurements were made, it was widely believed throughout the organic conductor community that the ground state of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ is antiferromagnetic,[41] although definitive evidence for antiferromagnetic ordering was never found. It was, however, noted by Christ *et al.* that α-(BEDT-TTF)$_2$KHg(SCN)$_4$ does not behave like a normal metal at high magnetic fields.[48] In the following section we show that the irreversible magnetization within the high-magnetic-field low-temperature phase is quite unlike that to be expected for any kind of magnetic ordering.

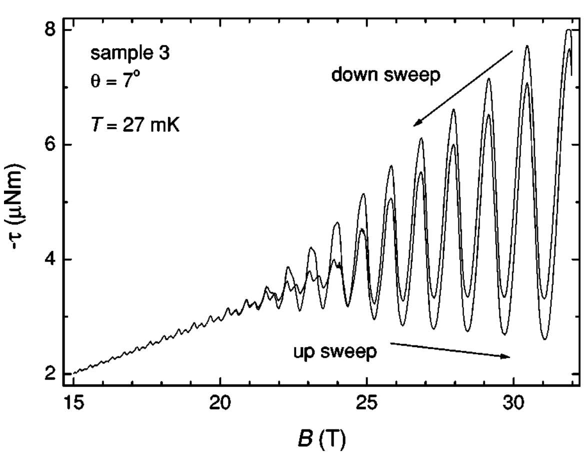

FIG. 4. The magnetic torque measured for sample 3 in rising and falling magnetic fields between 15 and 32 T. The lower (higher) of the two curves corresponds to the up (down) sweep, as indicated by the lower (upper) arrow. The temperature in the dilution refrigerator was 27 mK.

Consideration of the fact that only the anisotropy of the magnetic susceptibility gives rise to magnetic torque is the key to understanding the behavior of this material. Hence, because the dHvA effect is the oscillatory component of the Landau diamagnetism, thereby involving only orbital effects within the conducting planes, it manifests itself as large oscillations of the magnetic torque that are rather straightforward to interpret. At low magnetic fields ($B < B_k$), the oscillations exhibit the usual double-peaked structure that has been interpreted both as spin splitting[13,30,41,49,50] and as the frequency-doubling effect that accompanies a pinned CDW or SDW phase.[17] This is well known to be an effect observed at small angles ($\theta \lesssim 20°$) in this material (as well as the M = Tl and Rb salts), which occurs only within the low-magnetic-field DW phase. On increasing the field at these low temperatures (i.e., $T = 27$ mK), the kink transition is observed to be extremely abrupt, with a small but reproducible spike feature signaling the transformation into the high-magnetic-field regime at $B \sim 24.2$ T in sample 3. Above this field, the dHvA oscillations develop the characteristic triangular form that has been observed within the high-magnetic-field phase.[45,47,49]

The sudden change in the wave form that occurs at ~ 24.2 T on the rising magnetic field and at ~ 21.4 T on the falling magnetic field is indicative of an abrupt first order phase transition. Because the magnetic torque is entirely reversible within the low-magnetic-field DW phase, provided the field does not exceed ~ 24.2 T, both the dHvA oscillations and the monotonic background torque remain unchanged when the direction of sweep of the magnetic field is reversed. Similarly, when the field sweep direction is reversed within the high-magnetic-field phase, the dHvA oscillations continue to have the same triangular form, provided that the field is not swept below ~ 21.4 T. This type of behavior indicates that the low- and high-magnetic-field phases are immiscible, with the formation of domains being energetically unfavorable. Because it costs energy to mix the two phases, the low-magnetic-field DW phase is ''supercooled'' on increasing the magnetic field until the free energy difference between the two phases is no longer sustainable. To help understand this behavior in α-(BEDT-TTF)$_2$KHg(SCN)$_4$, it is instructive to consider a simplified model where the zero temperature free energies of the low- and high-magnetic-field regimes are approximated as

$$F_0 \approx -g_{1\mathrm{D}}\left[\frac{\Delta_0^2}{2} - h^2\right] \tag{3}$$

and

$$F_x \approx -g_{1\mathrm{D}}\frac{\Delta_x^2}{2}, \tag{4}$$

respectively, with $g_{1\mathrm{D}}$ being the density of Q1D states and $h = g\mu_{\mathrm{B}}B/2$.[15,17,24] We assume, to first order, that the free energy of the high-magnetic-field phase is not affected by magnetic field. Here, the subscript 0 denotes the CDW phase that is stable at low magnetic fields, while x denotes the spatially modulated or CDW-SDW hybrid phase.[15,24] If we then assume the free energy of the domain term $F_{\mathrm{mix}} \approx \epsilon y(1-y)$ to be approximately parabolic (i.e., the lowest-order even function of $y+1/2$), with y being the fraction of the material in the low-magnetic-field phase, then no domain structure can ever be stable provided $\epsilon \geqslant 0$. Since the total free energy is

$$F_{\mathrm{tot}} \approx yF_0 + (1-y)F_x + \epsilon y(1-y), \tag{5}$$

it then follows that, on increasing the magnetic field within the low-magnetic-field regime, for which $y=1$, the material cannot ''snap'' into the high-magnetic-field phase until $\partial F_{\mathrm{tot}}/\partial B \lesssim 0$. Conversely, on decreasing the magnetic field from within the high-magnetic-field regime, for which $y=0$, the material cannot snap into the low-magnetic-field phase until $\partial F_{\mathrm{tot}}/\partial B \gtrsim 0$. The kink transitions for rising and falling magnetic fields are therefore

$$h_{\mathrm{k}} = \sqrt{\frac{\Delta_0^2 - \Delta_x^2}{2} \pm \frac{\epsilon}{g_{\mathrm{1D}}}}, \tag{6}$$

with the $\pm$ sign corresponding to the direction of sweep of the magnetic field. The abruptness of the transitions should be accompanied by the release of latent heat, although this cannot be detected in the present isothermal experiment. If we assume the strength of the coupling to be similar in the two ordered regimes, then the ratio of order parameters Δ_x/Δ_0 is equal to the ratio of transition temperatures $T_{\mathrm{c}}/T_{\mathrm{p}} \sim 3/8$, and we obtain $\Delta_0 \sim 2.05$ meV and $\Delta_x \sim 0.75$ meV on inserting $B_{\mathrm{c}} \equiv B_{\mathrm{k}} \sim 23$ T. This is consistent to $\sim 50\%$ with the value obtained by application of the BCS relation $2\Delta = 3.52 k_{\mathrm{B}} T_{\mathrm{p}}$. Meanwhile, for $1/g_{\mathrm{1D}} \sim 30$ meV, we obtain $\epsilon \sim 24$ μeV.

The hysteresis that starts at magnetic fields above ~ 24.5 T has quite a different form from that associated with the kink transition. Since (1) the dHvA oscillations have the same shape and size between rising and falling magnetic fields and (2) the hysteresis takes place continuously over a wide interval in field (from ~ 24.5 T up to the highest available field of ~ 33 T), this type of hysteresis cannot be connected with the phase transition that takes place at B_{k}. While it is plausible that this hysteresis could be associated with yet another first order phase transition that takes place over an extended interval of field, in order for this to be the case, either (1) the free energies of two coexisting phases would have to be very similar but with slightly different dependences on B or μ, or (2) the mixing term ϵ would have to be very large (and negative). It is a large negative term of this type that is responsible for causing the normal/superconducting mixed phase of a type II superconductor to be stable over a very wide magnetic field interval $B_{\mathrm{c1}} < B < B_{\mathrm{c2}}$. Given that a CDW, of some form, is expected to persist to high magnetic fields in α-(BEDT-TTF)$_2$KHg(SCN)$_4$,[15,24] the high-magnetic-field phase would then have to consist of either (1) two coexisting but thoroughly mixed (i.e., $\epsilon \leqslant 0$) CDW, CDW-SDW hybrid, or normal metallic phases, with different anisotropic magnetic susceptibilities, or (2) one phase for which the nesting vector **Q**, and therefore the anisotropic susceptibility, experiences hysteresis as B is swept. Indeed, it is predicted by Zanchi *et al.*[24] that **Q** continually shifts with B within the high-magnetic-field CDW$_x$ phase, and the pinning of this CDW or CDW-SDW could conceivably lead to magnetic hysteresis. However, were this to be realized, the differences in Fermi surface topology between rising and falling magnetic fields should ultimately manifest themselves in the dHvA oscillations. This is to be expected since the peak-to-peak amplitude of the dHvA wave form is directly proportional to the number of Q2D states, while the gradient on the falling side of the oscillation (i.e., at integral filling) is proportional to the density of background states[31] [e.g., unnested quasi-one-dimensional (Q1D) states]. As it turns out, however, there is no detectable change in the peak-to-peak height, nor in the falling slope of the dHvA oscillations between rising and falling fields in Fig. 4, at least for $B \gtrsim 26$ T. In fact, from the fraction of the wave form $\gamma = g_{\mathrm{2D}}/(g_{\mathrm{2D}} + g_{\mathrm{1D}})$ $\sim 68\%$ over which the dHvA magnetization increases with field,[31] it is rather straightforward to infer that the density of background (Q1D) states is $(47 \pm 5\%)$ that of the Q2D pocket, in both rising and falling fields. Thus, the Q1D Fermi surface sheets appear to have the same density of states, within experimental uncertainly, as they do prior to their nesting.[43] This could imply that the Q1D states are not nested (or ''ungapped''). Indeed, it has been a common conclusion of *all* quantum oscillation and angle-dependent magnetoresistance oscillation measurements that the Fermi surface appears to be unreconstructed within the high-magnetic-field phase.[20,31] Alternatively, the order parameter could just simply be pinned to the chemical potential μ within the high-magnetic-field phase, which would then prevent us from being able to detect a change in the density of states of the Q1D Fermi surface sheets. This is always true in superconductors, but could also be true in certain types of DW system (e.g., the Bechgaard salts) for which there is a considerable amount of free energy to be gained by varying **Q** in order to help suppress the oscillations of μ.[51] One can therefore argue that if **Q** is sufficiently free to vary as it is in the Bechgaard salts, so as to cause no detectable difference in the density of states between rising and falling magnetic fields, then pinning of the CDW or CDW-SDW cannot be that significant. The fact that no hysteresis is observed in the magnetotransport either, lends further support to the notion of there being no change in the electronic structure between rising and falling magnetic fields.

To carry this argument further, in Fig. 5 we have integrated the hysteresis in Fig. 4 to arrive at the total energy E_{l} (solid line) lost as a result of the hysteresis incurred by sample 3 (of volume ~ 0.8 mm^3). Note that the losses associated with the kink transition in the vicinity of $B_{\mathrm{k}} \sim 23$ T are minimal,[52] but those within the high-magnetic-field phase greatly exceed the nominal free energy F_x (dotted line) of the high-magnetic-field phase by approximately a factor of 30. This shows quite clearly that the hysteresis cannot be attributed to a change in electronic structure between rising and falling magnetic fields, otherwise it would constitute a violation of the second law of thermodynamics.[53] Of course, were there actually a difference in the electronic structure between rising and falling magnetic fields, we could only expect the differences in Landau diamagnetic sus-

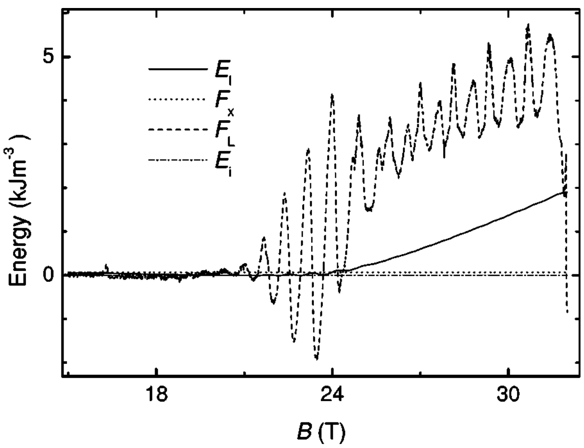

FIG. 5. A plot of various forms of energy associated with the high-magnetic-field phase as described in the text. F_x (dotted line) is the nominal free energy of the CDW_x phase (assuming the simplest model), E_l (solid line) is the integrated energy associated with the hysteretic losses, F_L (dashed line) is the equivalent Landau diamagnetic energy (should the hysteresis be caused by Landau diamagnetism), and E_i (dash-dotted line) is the approximate inductive stored energy (should the hysteresis be due to persistent currents).

ceptibility to give rise to a detectable hysteresis in the magnetic torque. Once again, if we calculate the free energy $F_L = \chi_L B^2/2$ associated with the Landau diamagnetic susceptibility χ_L that would be necessary in order to account for the experimentally observed hysteresis, we arrive at an estimate for F_L (dashed line) that greatly exceeds F_x. The incorporation of a diamagnetic term of this magnitude into Eq. (4) for F_x would therefore make the energy of the CDW_x phase much greater than that of the normal metal so that the formation of this phase at low temperatures would no longer be energetically possible.

The only other possibility, therefore, is that the hysteresis originates from some form of dynamic magnetism involving the ferromagnetic alignment of spins or circulating currents. Whenever magnetic hysteresis occurs, whether it is caused by ferromagnetism, metamagnetism, vortex pinning in a type II superconductor, or even induced currents in a quantum Hall system, the tendency is always to retain magnetic flux within the sample. Using the field-cooled magnetization as a point of reference (or the magnetization averaged between up and down sweeps), the magnetization is always slightly more diamagnetic on the rising field and slightly more paramagnetic on the falling field. Only the anisotropy of the magnetic susceptibility gives rise to magnetic torque. Therefore, the sign of the hysteresis observed here implies that it involves magnetic moments that are predominantly oriented perpendicular to the conducting planes, or, equivalently, currents flowing within the conducting planes. Clearly, α-(BEDT-TTF)$_2$KHg(SCN)$_4$ contains no magnetic ions with partially filled *d*- or *f*-electron shells, so we can eliminate this rather trivial source of ferromagnetism or metamagnetism. This, in fact, becomes a moot point in the following section, since ferromagnetism and metamagnetism never give rise to situations where $\partial M/\partial B < 0$ upon reversing the direction of sweep of the magnetic field, nor do they give rise to a critical state.

The hysteresis in Fig. 4 is therefore easier to explain in terms of induced currents. Orbital Landau diamagnetic currents, as shown above, would cost too much energy. However, the inductive stored energy $E_i = LI^2/2$ of persistent currents of the size required to account for the experimentally observed hysteresis is very low. For example, on estimating the sample inductance as $L \sim \pi r \mu_0 \sim 2$ nH for radius $r \sim 0.5$ mm and a uniform current of $I \sim 0.5$ A (required in order to account for the observed hysteresis), we arrive at an energy $E_i \sim 0.3$ J m^{-3} (dash-dotted line in Fig. 5) that is significantly less than F_x. Hence, only an induced current could give rise to magnetic hysteresis of the size observed experimentally without significantly perturbing the stability of the ground state. The amount of energy dissipated resistively on establishing the critical state is limited only by the maximum magnetic field gradient or Hall potential gradient that the sample can sustain, and not by the intrinsic free energy of the ground state.

Thus, the present data appear to indicate that the currents that were first reported in pulsed magnetic fields[21,23] are, in fact, persistent, at least on the experimental time scale of resistive Bitter magnet experiments. What is surprising in the present study, however, is that the hysteresis occurs at arbitrary filling factors and not just at integral filling factors. This therefore appears not to support the original explanation in terms of the QHE. Because the pulsed magnetic field experiment was sensitive only to the oscillatory component of the magnetic susceptibility, the amount of information that could be extracted from that measurement was comparatively limited. There was no indication, at that time,[21,23] of any significant dc component to the magnetic hysteresis.

VI. IRREVERSIBLE PROCESSES

The most compelling evidence for induced currents is that provided by the irreversibility of the magnetic torque on stopping or reversing the sweep direction of the magnetic field. In most metals, including those with DW phases, the magnetization is usually reversible, as is the case, for example, within the low-magnetic-field DW phase (i.e., see Fig. 4). On sweeping the field back and forth within the high-magnetic-field phase, on the other hand, even over a very small interval in field, the magnetic torque of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ is observed to be entirely irreversible, giving rise to a hysteresis loop of the form shown in Fig. 6(a). The nature of the hysteresis loop has several of the characteristic features of a critical-state model, such as the Bean model[28,29] that is used to describe magnetic hysteresis caused by vortex pinning in type II superconductors. This becomes particularly evident in Fig. 6(b) when the monotonic background and dHvA contributions are subtracted; the background is obtained by averaging data taken on full up and down sweeps of the magnetic field. In Fig. 6(b) we can see that, on reversing the sweep direction of the magnetic field, the magnetic hysteresis increases gradually until a critical value is reached. In type II superconductors, this would be proportional to the critical current density j_c. In Fig. 6(b), the induced magnetization corresponding to the critical state is that obtained by performing full up and down sweeps [also shown in Fig. 6(b)]. By ''critical state'' we imply that the sample has a tendency to trap flux exactly like a type II

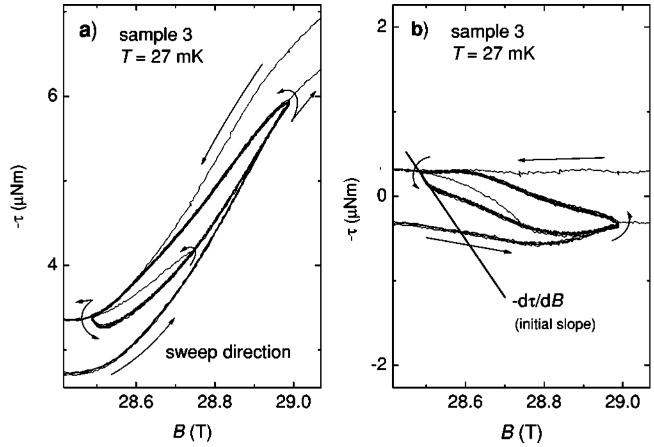

FIG. 6. (a) An example of a hysteresis loop observed in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ as a result of sweeping the magnetic field back and forth several times over the same interval at different rates between 8 mT s^{-1} and 50 mT s^{-1}. Part of the hysteresis obtained by sweeping the magnetic field over the entire range between 24.4 and 32 T is also shown. (b) The same hysteresis after subtracting the dHvA and monotonic background magnetic torque.

superconductor. While the unit of trapped flux in type II superconductors is the vortex, which contains exactly one flux quantum, it is unknown what the equivalent unit of trapped flux could be in the high-magnetic-field phase of α-(BEDT-TTF)$_2$KHg(SCN)$_4$.

Since a magnetic critical state is known only to occur in type II superconductors, this is the only type of system with which comparisons can be made. It follows from the critical-state model, as normally applied to type II superconductors, that as soon as the direction of sweep of the magnetic field is reversed the initial slope of the magnetization with respect to magnetic field is given by

$$\chi' = \frac{\partial M}{\partial B} = -\frac{f[1-\eta]}{\mu_0}, \tag{7}$$

where $-1/\mu_0$ corresponds to perfect diamagnetism, η is the demagnetization factor, and f is the volume fraction of the sample in which persistent currents flow, until a critical state is achieved. If we consider the anisotropic hysteresis to originate from currents flowing only within the conducting planes and that $\eta \sim 0.5$ (as for a cylinder), upon taking the initial slope of the magnetic torque in Fig. 6(b) we obtain $f \sim 1\%$ for sample 3 (of volume ~ 0.8 mm^3). This implies that the induced currents are effective at screening the externally changing magnetic field from at least 1% of the sample, or that the critical current is extremely inhomogeneous. This value of $f \sim 1\%$ would, of course, be underestimated were any of the current to flow perpendicular to the conducting planes, in what is normally the least conducting direction of the organic metal. The value of 1% should therefore be considered as a lower limit. That it is necessary to sweep the magnetic field by ~ 0.4 T before the polarity of the current in the sample is completely reversed could either imply that the local field difference in parts of the sample reaches strengths of order 0.4 T, or that shape effects, which give rise to demagnetization, are important.

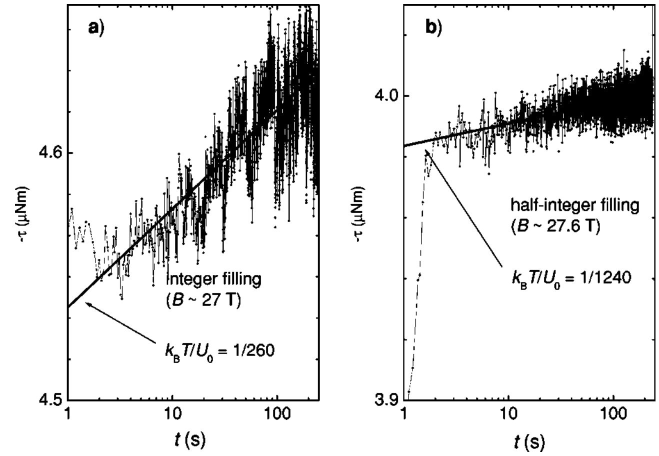

FIG. 7. Relaxation of the magnetic torque observed at an integral filling factor (a) and half-integral filling factor (b), with time t plotted on a logarithmic scale. At half-integral filling factors, the moment appears to decay logarithmically, as expected for a type II superconductor.

Another property of critical-state models, as applied to type II superconductors, is that they often give rise to relaxation phenomena. In type II superconductors, this either results from thermally assisted flux flow or quantum flux creep.[29,54,55] One way to assess whether such effects occur in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ is to monitor the dependence of the width of the hysteresis loop on sweep rate. Nonlinear current-voltage characteristics have already been detected in pulsed magnetic field experiments.[21,56] On sweeping the field at much slower rates between 8 mT s^{-1} and 50 mT s^{-1} in Fig. 6, the hysteresis increases by no more than 10%, implying that the relaxation rate is rather low. A more notable degree of relaxation of the magnetic torque is observed by stopping the sweep of the magnetic field abruptly and then observing changes over several minutes. At integral filling factors [Fig. 7(a)], the magnetic torque decays much more rapidly than at half-integral filling factors [Fig. 7(b)] and is considerably more noisy. On fitting the Anderson-Kim flux-creep model,[29] as normally applied to most type II superconductors, (making the substitution of τ for j),

$$\frac{\tau}{\tau_c} \approx \frac{j}{j_c} \approx \left[1 - \frac{k_B T}{U_0} \ln \frac{t}{t_0}\right], \tag{8}$$

the best fit is obtained at half-integral filling factors, with the characteristic parameter $k_B T/U_0$ for logarithmic decay being of order 10^{-3}. This is certainly within an order of magnitude of the experimental observations for most type II superconductors.[29] Thus, it appears to be the case that the irreversible magnetic properties of α-(BEDT-TTF)$_2$KHg(SCN)$_4$, at high magnetic fields and low temperatures are very similar to those of a type II superconductor.

VII. FIELD AND TEMPERATURE DEPENDENCE OF THE HYSTERESIS

A further analogy can be made with extreme type II superconductors on consideration of the temperature dependence of the width of the hysteresis loop. In many extreme

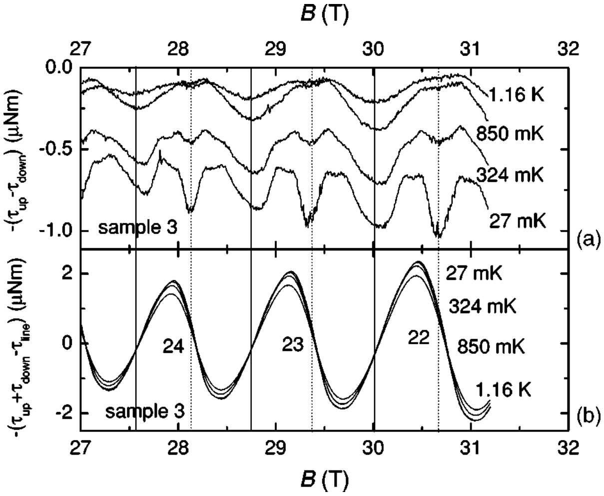

FIG. 8. (a) The hysteresis observed in α-(BEDT-TTF)$_2$KHg(SCN)$_4$, measured in the dilution refrigerator at several different temperatures, obtained by subtracting the down sweep data from the up sweep. The wave form of the hysteresis is complicated, being largest at half-integral filling factors at higher temperatures, then becoming more pronounced at integral filling factors at the lowest temperature. (b) The dHvA signal extracted from the same data by summing up and down sweeps. The monotonic background magnetic torque has been subtracted for clarity.

type II superconductors, this is found to be strongly dependent on temperature, caused, for example, by the dependence of the effective vortex pinning potential $U(T)$ on T.[29,54] Such a behavior is also observable for the critical state of α-(BEDT-TTF)$_2$KHg(SCN)$_4$, as shown in Fig. 8(a), albeit the hysteresis contains an oscillatory component. While no hysteresis can be detected at $T\sim4$ K, it is already quite pronounced by 1.16 K, particularly at half-integral filling factors. Half-integral filling factors, realized whenever $F/B+1/2$ assumes an integer value, are depicted in Fig. 8 by solid vertical lines. Note that the oscillatory component of the width of the magnetic hysteresis loop oscillates in quadrature with the dHvA oscillations [extracted in Fig. 8(b)], but in phase with the density of states, with the hysteresis and the density of states at μ both exhibiting maxima at half-integral filling factors. As was discussed in the preceding section, it cannot be the dHvA oscillations themselves that are hysteretic because (1) the dHvA and oscillatory component of the hysteresis effects are at quadrature with respect to each other and (2) the actual dHvA amplitude is only relatively weakly dependent on temperature over the same range, 27 mK$<T<$1.15 K, shown in Fig. 8(b). This, together with the absence of any hysteresis in the SdH wave form, is, again, consistent with our hypothesis in terms of induced currents.

As $T\rightarrow0$, the hysteresis becomes double peaked, with a second sharper and more strongly temperature-dependent peak emerging at integral filling factors. The occurrence of maxima in the hysteresis at both integral and half-integral filling factors, in the present work, is thermodynamically consistent with the hysteresis in the magnetothermal oscillations observed by Fortune *et al.*[57] at the highest magnetic fields, although it is likely that an experimental time constant was involved in their measurements, given the nature of the experimental technique.[57] Sharp peaks in the hysteresis were

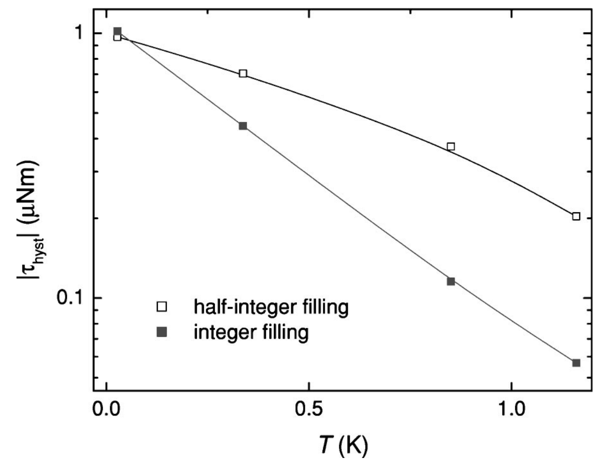

FIG. 9. A logarithmic plot of the maxima in the hysteresis versus T at half-integral filling factors ($B\sim30.1$ T) and integral filling factors ($B\sim30.7$ T).

observed only at integral filling factors in pulsed magnetic fields,[21,23] however, and it was this ''coincidence'' that led to the data being interpreted in terms of the QHE. When plotted as a function of temperature in Fig. 9, the hysteresis is strongly dependent on temperature at both integral and half-integral filling factors, with the form of the T dependence being approximately exponential, and therefore having some similarity to that typically observed in extreme type II superconductors.[29,54]

VIII. DISCUSSION AND CONCLUSION

Having identified the primary physical effects that enable us to determine the boundaries between the different phases, a tentative phase diagram for α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at least for fields applied within $\theta\sim20°$ of the **b** axis of the crystals, is shown in Fig. 10. Solid symbols in Fig. 10 have been chosen for data points extracted from thermodynamic data, which are often the most reliable indicator of a thermodynamic phase transition. Solid squares denote the change in

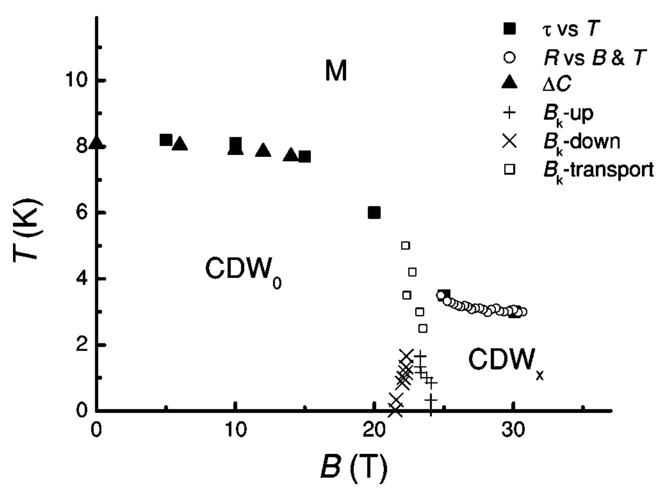

FIG. 10. A tentative phase diagram for α-(BEDT-TTF)$_2$KHg(SCN)$_4$ in a magnetic field, for small values of θ. The points have been extracted from both magnetotransport and magnetic torque measurements discussed in this work, as well as specific heat measurements of Kovalev *et al.*

slope of the field-cooled magnetic torque extracted from Fig. 3. This agrees rather well with the resistive transition into the low-temperature phase at high magnetic fields (indicated by open circles), as, for example, can be seen in both samples 1 and 2 in Fig. 2. Note that this transition is not sample dependent; only the extent to which the resistivity decreases at low temperatures is sample dependent, varying somewhat between samples 1 and 2. However, there is some indication that the phase boundary might oscillate with magnetic field in phase with the total density of states. At low magnetic fields, the field-cooled magnetic torque data agree rather well with the transition into the low-magnetic-field DW phase obtained from specific heat measurements,[40] represented here by solid triangles.

In the vicinity of the kink transition, the second order phase boundary, on cooling, becomes more difficult to extract from resistivity data, owing to the competition between different types of resistivity behavior from the low-magnetic-field DW and exotic high-magnetic-field phases. For this reason, these points, depicted here as open squares, are taken from the nonhysteretic kink observed in the magnetoresistance at higher temperatures between 2.5 K and 5 K. The hysteretic kink transition at lower temperatures, on the other hand, is extracted from the magnetic torque measurements made in the dilution refrigerator, denoted here by vertical crosses on the rising magnetic field and diagonal crosses on the falling magnetic field.

The phase diagram that emerges here is similar to that recently obtained by Kartsovnik,[58] based on a revision of an earlier phase diagram by the same author and co-workers.[18] Because we have restricted our determination of phase boundaries to those that can be either reproduced by measurement of the specific heat[40] or observed as an abrupt change in slope of the magnetic torque with temperature, it should be noted that our phase diagram is somewhat different from that obtained by Sakaki *et al.*[13] The phase diagram depicted in Fig. 10 can be summarized as follows: At temperatures above ~ 8 K, α-(BEDT-TTF)$_2$KHg(SCN)$_4$ behaves like an ordinary organic metal, transforming into a DW phase at low temperatures. The absence of any evidence for static antiferromagnetically configured spins,[14,42] together with the close similarity of our experimentally determined phase diagram to that predicted by both McKenzie[15] and Zanchi *et al.*,[24] implies that the low-temperature low-magnetic-field phase is likely to be a CDW, referred to by theorists as the CDW$_0$ phase. The first order transition that occurs on sweeping the magnetic field between the high- and low-magnetic-field, low-temperature phases is mostly consistent with the arguments of McKenzie.[15] What is particularly interesting in the current work is that, while the high-magnetic-field phase (denoted CDW$_x$) is expected on theoretical grounds to be a modulated CDW phase or mixed CDW-SDW hybrid, its transport and magnetic properties are quite unlike those observed in all known DW systems.[37]

Rather, we have shown that the magnetic behavior of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at high magnetic fields has many of the characteristic features of a critical-state model, closely resembling an extreme type II superconductor.[29,54,55] This critical-state behavior, together with the abrupt drop in resistivity at low temperatures, clearly cannot be explained by any of the existing models,[15,24,37] nor does it appear to be connected with the QHE.[21–23] This certainly provokes the question as to whether α-(BEDT-TTF)$_2$KHg(SCN)$_4$ undergoes a transition at high magnetic fields into a field-induced superconducting state. However, there are many physical reasons for rejecting such an idea. While the presence of a CDW at low magnetic fields would appear to indicate that electron-phonon interactions are relevant in this material, a CDW ground state is invariably always more stable than an electron-phonon mediated superconductor in strong magnetic fields. Both CDW's and singlet-paired superconductors are Pauli limited at a critical field B_c. However, nearly all superconductors have the additional disadvantage of being suppressed by orbital effects at much lower magnetic fields than that at which the Pauli critical field is expected to occur.[29] Thus, were the ground state of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ to result from the competition between superconductivity and CDW's, as one might gather from the fact that the $M = NH_4$ salt is superconducting and that α-(BEDT-TTF)$_2$KHg(SCN)$_4$ becomes superconducting under uniaxial stress, the fact that the CDW has already ''won'' at $B=0$, makes the emergence of superconductivity in an applied field seem somewhat remote. Furthermore, while several models have been proposed for ''reentrant'' superconductivity in very strong magnetic fields,[55,59–61] a common prerequisite of these models is that the material is already a stable superconductor at $B=0$. On the other hand, qualitatively similar irreversible magnetic and magnetoresistive effects are observed in quite a different charge-transfer salt in fields $B \gtrsim 40$ T, namely, κ-(BEDT-TTF)$_2$I$_3$, which is well known to be a superconductor at low magnetic fields, yet does not exhibit any form of density wave.[62]

Perhaps the fact that the experimental phase diagram of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ reproduces that obtained theoretically[15,24] for CDW's, implies that we should look for an explanation for the unusual physical effects in terms of CDW physics. In fact, further parallels with the phase diagram of Zanchi *et al.*[24] are found on studying its dependence on the orientation of the magnetic field.[63] Then again, we have also shown that pinning of the field-dependent **Q** vector within the CDW$_x$ phase appears not to be a viable mechanism for causing the experimentally observed hysteresis. While the mean field theories for CDW's and SDW's appear to work well in weak magnetic fields,[37] this in no guarantee that they should continue to work in very strong magnetic fields. The present theory[15,24] makes no allowance for the possibility of the magnitude of the CDW$_x$ order parameter being spatially modulated, nor does it take into consideration the possibility of currents.

On reflection, it is worth noting that the CDW was originally proposed by Fröhlich as an early theory for superconductivity.[64] While this model was subsequently shown not to be that appropriate for superconductors, CDW sliding can occur in bulk Q1D systems, provided that the CDW pinning potential is overcome by a sufficiently large electric field.[37] Only in ringlike nanoscale systems, which are too small to contain impurities, have persistent CDW currents been proposed to exist.[65] Given that vortices, located at the nodes in the order parameter, around which currents circulate, are nanoscale entities smaller than the typical interimpurity separation,[66] the question that needs to be postulated is whether an allowance for currents and nodes in the CDW_x order parameter could lead to a mixed CDW_x–normal-metallic phase that adopts the physical characteristics of the mixed phase of a type II superconductor. This would certainly have the potential to explain the irreversible magnetic properties of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ (i.e., the observation of a critical state and the large negative differential susceptibility $\partial M/\partial B$ on changing the sign of $\partial B/\partial t$), and perhaps also the pronounced drop in the interlayer magnetotransport. The high-magnetic-field phase of the CDW therefore requires considerably more theoretical attention before we can properly understand α-(BEDT-TTF)$_2$KHg(SCN)$_4$. What needs to be established experimentally is whether the observed critical state can imply anything other than persistent currents.

ACKNOWLEDGMENTS

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