Field-Induced Dynamic Diamagnetism in a Charge-Density-Wave System

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ac susceptibility measurements of the charge-density-wave (CDW) compound α-(BEDT-TTF)$_2$-KHg(SCN)$_4$ at magnetic fields, $\mu_0 H > 23$ T, above its Pauli paramagnetic limit, reveal unambiguously that the magnetic hysteresis observed previously within this CDW phase is diamagnetic and can only be explained by induced currents. It is argued that the ensemble of experimental techniques amounts to a strong case for dissipationless conductivity within this phase.

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Dynamic diamagnetism, caused by persistent currents induced by changing magnetic fields, is well known to occur both in quantum Hall systems [1], for which the current and Hall electric field are exactly orthogonal, and in type-II superconductors, for which the current is intrinsically related to the pinned vortex density gradient [2]. Thus far, no further mechanisms for persistent currents in bulk macroscopic systems have been discovered. However, phenomena bearing qualitatively similar physical characteristics to persistent currents [3] have been detected at fields above, what is thought to be, the Pauli paramagnetic limit of the charge-density-wave (CDW) system α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ (M = K or Tl) [4–8]. When the total differential susceptibility $\chi' = \partial M/\partial H \sim 1\%$ departs significantly from perfect diamagnetism (i.e., $\chi' \equiv -100\%$) [3], however, proving the existence of dissipationless conductivity, in only a small fraction of a sample, is by no means a trivial matter. Thus far, it can be argued only that the observations exhibit many physical inconsistencies [3] with the behaviors that should be expected for the various CDW [5,6], antiferromagnetic [9,10], or quantum Hall [11–13] ground states that have been proposed to exist in this material since its discovery.

In this Letter, we report the results of ac susceptibility measurements within the high magnetic field phase of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ that further exemplify the growing disparity between physical observations and the proposed models [3,5,6,9–13]. By using standard ac susceptibility techniques, the real and imaginary contributions χ' and χ'' are determined unambiguously, revealing a large ratio $\chi''/\chi' > 1$ that firmly establishes the existence of induced currents. By itself, this ac susceptibility measurement indicates quite directly that the in-plane resistivity becomes significantly lower than that ordinarily expected for a low carrier density metal within a high magnetic field CDW phase. Furthermore, the dependence of these currents on temperature, time, and Landau level filling factor is qualitatively and quantitatively similar to the hysteresis observed in the magnetic torque [3], thereby indicating that these phenomena have a common origin. When considered together, these experiments amount to compelling evidence for the existence of dissipationless conductivity and persistent currents within the high magnetic field phase of α-(BEDT-TTF)$_2$KHg(SCN)$_4$, realized for at least part of the sample.

The sample, in roughly the form of a cube of volume $V \approx 1.2 \times 10^{-10}$ m^3, was chosen from the same batch as used for earlier studies of the high magnetic field phase [3] and was placed on the end of detection coils, originally designed to measure small samples in rapidly changing magnetic fields [14], thereby having a linear response to frequencies as high as 1 MHz. Both the sample and the coils were immersed in either gaseous or liquid ^{3}He throughout the experiment, with dc magnetic fields extending to ~31 T, applied along the **b** axis of the crystal, provided by the National High Magnetic Field Laboratory, Tallahassee. The ac field component, also applied along the **b** axis, was generated using a solenoid wound around the outside of the plastic ^{3}He refrigerator immersed in superfluid ^{4}He. A detectable signal was obtained using frequencies as low as $\omega/2\pi \sim 10^4$ Hz, with an ac field of $h_0/\sqrt{2} \sim 43$ G rms being achieved without significantly heating the superfluid bath. In performing this ac susceptibility experiment, the induced voltage was related to the magnetic susceptibility by the formula

$$v' + iv'' = cV[\chi' + i\chi'']\omega h_0, \qquad (1)$$

where $c \sim 0.010 \pm 0.005$ H m^{-1} is the geometrical coupling factor of the sample to the coils. In order to provide a secondary calibration of c (in addition to that evaluated numerically), and to ensure that the phase of the susceptibility was determined correctly, the entire detection system was tested on samples of superconducting κ-(BEDT-TTF)$_2$Cu(NCS)$_4$ of comparable dimensions.

Concentrating initially on the ac susceptibility results in the absence of a dc magnetic field, the temperature T dependence of χ' and χ'' for the α-(BEDT-TTF)$_2$KHg-(SCN)$_4$ sample is shown in Fig. 1(a). While no pronounced change in χ' is detectable above the level of the noise, a change in χ'' is observed, indicating that the

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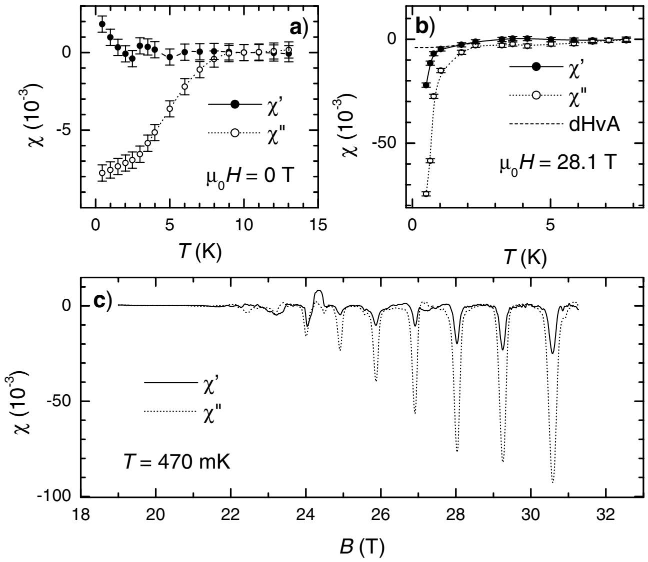

FIG. 1. The temperature dependences of χ' and χ'' measured (a) at $\mu_0 H = 0$ and (b) at $\mu_0 H \sim 28.1$ T (i.e., at an integral filling factor). Note that the maximum diamagnetic de Haas–van Alphen contribution to χ' (dashed line) is both small and relatively weakly T-dependent [16]. (c) The field dependence of χ' and χ'' within the high magnetic field phase at $T \sim 470$ mK.

susceptometer functions effectively as a contactless means for determining the residual in-plane resistivity. Given the difficulties encountered when attempting to determine ρ_{xx} or ρ_{yy} when using standard four probe transport techniques [11,15], this is already a useful result. Adopting the cylindrical sample approximation, whereby $\rho_{\parallel} = \frac{1}{2}(\rho_{xx} + \rho_{yy}) \approx -\mu_0 r^2 \omega / 8\chi''$, on inserting the minimum value of $\chi'' \sim -7.5\%$ and $r \sim 250$ μm into this expression, we arrive at the estimate $\rho_{\parallel} \sim 8 \pm 4$ $\mu\Omega$ cm. Note that the skin depth, $\delta_{\parallel} = \sqrt{2\rho_{\parallel}/\mu_0\omega} \sim 1400 \pm 350$ μm, comfortably exceeds the sample radius, therefore implying that the oscillating electromagnetic field is easily able to penetrate at this frequency.

When a dc magnetic field $\mu_0 H$ is introduced, both χ' and χ'' become undetectably small as $\rho_{\parallel}$ increases and the signal-to-noise ratio deteriorates; the major source of noise originating from the power supplies used to generate the current for the Bitter magnet coils. The lack of any significant signal is to be expected in this material, since the largest normal monotonic contribution to the magnetic susceptibility for this 2D metal, i.e., the Pauli paramagnetic susceptibility $\chi_P = \mu_0 e^2 m^* k_z / 8\pi^2 m_e^2 \sim 5 \times 10^{-6}$ (where $m^*/m_e \sim 3.5$ is the mass renormalization for the entire density of states, m_e is the free electron mass, and k_z is the periodicity of the Fermi surface perpendicular to the conducting layers, which is present in the formula following the summation over many layers), is beyond the sensitivity of this susceptometer. Note also that, since $2\pi F h_0 / H^2 \approx 0.03 \ll 1$ (where $\mu_0 F \sim 670$ T is the characteristic quantum oscillation frequency for this material), the optimum conditions for observing the de Haas–van Alphen effect in this material are far from realized at $\sim$30 T [16]. In spite of this, a strong signal both in χ' and χ'' develops above the kink transition field $\mu_0 H_k \sim 23$ T in Fig. 1(b), particularly at integral filling factors, as shown in Fig. 1(c).

A negative χ' within the high magnetic field phase of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ (i.e., $\mu_0 H > \mu_0 H_k$) implies that the magnetism is primarily diamagnetic [17], therefore indicating that the salient physics involves the motion of itinerant electrons [18]. Changes in the Landau diamagnetism, associated with CDW pinning of the field-dependent CDW nesting vector [5], has been proposed as one of the possible candidates for magnetic hysteresis within this phase [3,6,19]. However, for Landau diamagnetism $\chi_L = -\mu_0 e^2 k_z / 24\pi^2 m^*$ to account for a diamagnetic response of this magnitude would require an incredibly low effective mass of $m^* \lesssim 2 \times 10^{-5} m_e$. Clearly, an electron (or hole) pocket with an effective mass this low could not exist at fields above $\sim$23 T; otherwise its characteristic cyclotron energy $\hbar\omega_c \sim 100$ eV would greatly exceed the electronic bandwidth of the metal. The ac susceptibility results in Fig. 1 can therefore be explained only by the induction of currents. In fact, a situation where $\chi''/\chi' > 1$ (i.e., the imaginary component exceeds the real component), can be realized only at this low a frequency (i.e., 10 kHz) by induced currents [18], since their resistive dissipation is the only conceivable means by which the hysteretic energy losses $\mu_0|\chi''|h_0^2/2$ can exceed the intrinsic change in free energy $\mu_0|\chi'|h_0^2/2$. The underlying physical reason for this is that the energy consumed by currents is supplied directly by the induced emf (v_{emf}) as the externally applied magnetic field is changed, rather than by intrinsic reconfigurations of the ground state.

Diamagnetism due to currents, which manifests itself both in the real and the imaginary susceptibility, should occur in any system for which the resistivity becomes sufficiently small at low temperatures. Such effects certainly occur in quantum Hall systems [1], in the vicinity of a Hall plateau, and in superconductors [2], although, for practical reasons, only the latter are regularly the subject of ac susceptibility measurements. As the resistivity starts to drop on decreasing T, χ'' is the first to decrease, owing to the fact that the complex penetration depth $\tilde{\lambda}$ [20] (not to be confused with the London penetration depth) is larger than the sample dimensions. As the resistivity falls further, $\tilde{\lambda}$ eventually becomes smaller than the sample dimensions, causing χ' to drop at the expense of χ'' as the sample behaves more and more like an ideal inductor. Only if $\tilde{\lambda}$ approaches the London penetration depth λ_L, and $\chi'' \equiv 0$, can the entire sample be considered to be dissipationless.

Clearly, this limit is far from being realized with the present measurements on α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at $T \gtrsim 470$ mK and $\mu_0 H \sim 28.1$ T, as evident in Fig. 1(b). To investigate how χ' and χ'' develop further would require an extension of this ac susceptibility technique to dilution refrigerator temperatures. While this cannot yet be accomplished with the existing apparatus, the drop in χ'' does, however, precede the drop in χ', as T is lowered, enabling the mean bulk resistivity at $T \sim 470$ mK and $\mu_0 H \sim 28.1$ T to be estimated. By applying the cylindrical approximation once again, we arrive at an estimate $\bar{\rho}_{\parallel} \sim 0.8 \pm 0.4\ \mu\Omega$ cm at $\mu_0 H \sim 28.1$ T that is now half that of room temperature Cu. This is certainly unusual for a metal with $\sim 10^5$ times fewer carriers than Cu, and is even more unusual for what is thought to be a CDW ground state. Because the skin depth, $\delta_{\parallel} \sim 440 \pm 110\ \mu$m, is now comparable with the sample diameter, $2r \sim 500\ \mu$m, a fraction of the ac field becomes screened from the sample interior. In the limit $\delta_{\parallel} > r$, the maximum fraction of the sample screened by the induced currents can be estimated by using the expression, $\chi' \approx -r^4/96\delta_{\parallel}^4$, for an infinitely long cylinder, with which we obtain $\chi' \sim -0.1 \pm 0.1\%$. Thus, even by overestimating this quantity, we fail to account for the depth of χ' observed experimentally. This could imply that the minimum in-plane resistivity is extremely inhomogeneous at integral filling factors, with some parts of the sample having local resistivities orders of magnitude lower than $0.8 \pm 0.4\ \mu\Omega$ cm in order to locally screen the oscillatory magnetic field. Another possible explanation for the large depth of χ' could be the nonlinear current-versus-voltage (I-V) characteristics of the material, whereby the impedance of the sample varies strongly throughout the ac cycle (see discussion below), vanishing only when the oscillatory induced voltage v_{emf} crosses through zero.

While the present ac susceptibility measurements at $T \gtrsim 470$ mK, by themselves, do not prove the existence of dissipationless conductivity, they do prove that a low resistivity state is achieved at high magnetic fields in α-(BEDT-TTF)$_2$KHg(SCN)$_4$, at least at integral filling factors, which readily enables the induction of sizeable currents of order ~ 0.5 A in ac fields of ~ 10 kHz and in pulsed magnetic fields [11]. Not until the dynamic diamagnetism observed in this work and that giving rise to diamagnetic hysteresis in the magnetic torque of Ref. [3] are shown to have a common origin is the case for persistent currents established. While we do not yet have access to a single experimental technique that can bridge all experimental regimes, the features observed by each separate technique do exhibit striking similarities that would be difficult to attribute to independent mechanisms. For example, the currents observed here are strongly dependent on T, for $T \lesssim 2$ K, as are those detected in pulsed magnetic fields [11,13], as is the diamagnetic hysteresis observed in the magnetic torque in dc fields [3]. This is most clearly indicated in Fig. 2, where the dynamic magnetizations $|M|$ obtained using ac and dc techniques are compared. Although the magnitudes of $|M|$ are different, possibly owing to the nonlinear I-V characteristics of the material (see below), their dependences on T are similarly strong. In the magnetic torque measurements, $|M|$ is extracted directly from the hysteresis in Fig. 8a of Harrison *et al.* [3], while in this paper $|M| \approx \sqrt{\chi'^2 + \chi''^2}\, h_0$. Surely, both techniques must be measuring the *same* effect.

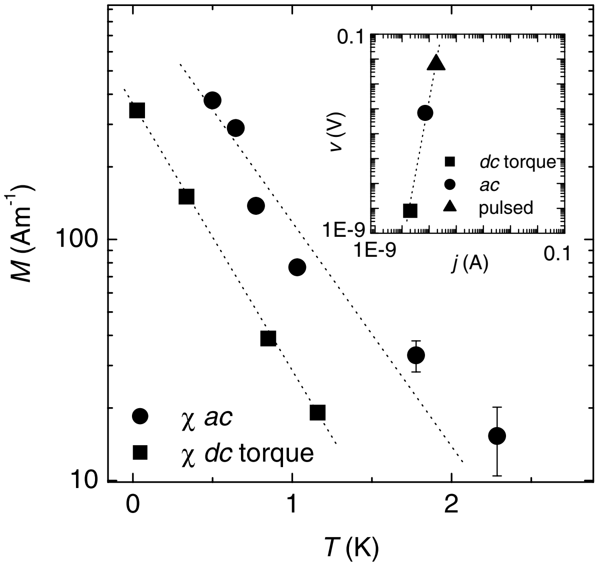

FIG. 2. The estimated dynamic magnetization $|M|$ plotted versus T, extracted from the ac susceptibility measurements, in this work, and the magnetic torque measurements. The inset shows the current-versus-voltage (I-V) characteristics of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at $\mu_0 H \sim 30$ T estimated from three different experimental techniques, as described in the text. In the present experiments, it is the zero-to-peak amplitude of the moment that is considered with the rate of change of magnetic field being ~ 270 T s^{-1}. Dotted lines have been drawn where appropriate to guide the eye.

Furthermore, in all cases [3,11,13], the largest diamagnetic response, at the lowest temperatures, occurs at integral filling factors and, in all cases, the diamagnetic response is observed to be most strongly time dependent at integral filling factors. This latter observation is evidenced in the present ac susceptibility experiments by the presence of a significant imaginary component, while, in the pulsed magnetic field and static magnetic field torque measurements [3,11,13], it is evidenced by the nonlinear I-V characteristics, or, equivalently, by the observation of a critical state [3]. This is yet further exemplified, in the magnetic torque measurements [3], by the slow logarithmic decay of the hysteresis with time. Perhaps the most remarkable observation is that the magnitude of the current extracted from each measurement technique is approximately the same (to within an order of magnitude). On combining all three different experiments, we now have the opportunity to construct an I-V plot in the inset of Fig. 2 that extends over six decades of voltage $v_{emf} = a\mu_0 \partial H/\partial t$, where $\mu_0 \partial H/\partial t$ is the rate of change of magnetic field and a is the cross-sectional area of the sample. The current per layer, on the other hand, is easily estimated by using the approximate formula $j \approx bM$, where $b = 2\pi/k_z$ is the interlayer separation. Despite having used widely varying techniques, a weak but steady dependence of j on v_{emf} is apparent.

In conclusion, we have shown that the diamagnetic features observed in α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ (where M = K or Tl) [11,13] can only be attributed to induced currents. These currents have similar physical characteristics to the hysteresis observed in magnetic torque measurements in purely dc magnetic fields, thereby suggesting that these same currents become persistent at sufficiently low temperatures in, at least, part of the sample. Since critical currents are common both to the quantum Hall effect [1] and superconductivity [2], and therefore probably to any other type of current-carrying ground state, identifying the mechanism responsible for persistent currents, here, is a matter that meets with considerable difficulty. That the currents occur predominantly at integral filling factors, when the chemical potential μ is situated in a Landau gap, and are accompanied by a significant Hall potential gradient [21] is notionally consistent with an explanation in terms of the quantum Hall effect [11–13]. Given that α-(BEDT-TTF)$_2$KHg(SCN)$_2$ possesses a simple quasi-two-dimensional Fermi surface that is approaching the quantum limit at $\mu_0 H \gtrsim 23$ T; this is a reasonable thing to expect. The hysteresis in the magnetic torque is, however, much too extensive to be accounted for by persistent currents that should only be induced when μ is situated in between Landau levels [1,3]. Thermodynamic measurements, on the other hand, indicate that a low temperature phase exists at high magnetic fields that is different from the normal metallic phase [3,22,23]. The boundaries of this phase are consistent with theoretical models of the CDW phase diagram [5,6]. However, these mean field theories far from anticipate even a good metal at high magnetic fields, let alone the occurrence of persistent currents. It has also been noted [3] that the magnetic hysteresis within the high magnetic field phase of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ exhibits a critical state. Such behavior is precedented only by type-II superconductors in strong magnetic fields [2]. Given that the critical state persists throughout the region of H-T space theoretically associated with CDW$_x$ [5,6], one cannot exclude the possibility of the condensate [24] being somehow involved in generating persistent currents at fields above the Pauli paramagnetic limit. For this to be realized, however, the CDW phase would have to be quite unlike any other form of CDW phase that has been found to exist [3,25]; it would have to be both radically depinned and capable of forming something analogous to a vortex state.

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Note added.—Since submitting this Letter, magnetic measurements made in the 45 T hybrid magnet at the National High Magnetic Field Laboratory, Tallahassee [26] have shown that these currents persist to fields far in excess of 40 T.

[1] See standard texts; for example, T. Chakraborty, in *The Quantum Hall Effects: Integral and Fractional, Second Edition,* Springer Series in Solid-State Sciences Vol. 85 (Springer-Verlag, Berlin, 1995).

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[4] The Pauli paramagnetic limit is the term given to the field at which the Zeeman splitting of the nested Fermi surface sheets becomes comparable to the zero field CDW gap $2\Delta_0$.

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[17] Note that the diamagnetic spikes in χ' cannot be accounted for by the falling slope of the de Haas–van Alphen signal, which has a maximum value of $\mu_0 e^2 k_z F^2 / 2\pi^2(1 - \gamma)\hbar m^* H^2 \sim 0.4\%$ at $T \rightarrow 0$ (see Fig. 2a in Ref. [3]).

[18] Note that the frequency used for this study corresponds to $\sim 4 \times 10^{-8}$ meV or 0.4 μK. It is inconceivable that a resonant gap in the electronic structure could occur at this low energy.

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