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PERSISTENT CURRENTS AT FIELDS ABOVE 23 T

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Experimental studies made on organic conducting salts of the composition α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ (where M = K, Tl and Rb) indicate that they exhibit persistent currents at magnetic fields exceeding 23 T. The presence of currents cannot be explained by the quantum Hall effect, while superconductivity seems unlikely. All indications point towards a new type of dissipationless current flow involving relative gradients in the pinning of a CDW and quantized orbital magnetism.

In high purity two-dimensional itinerant electron systems, Landau quantization often leads to an orbital magnetization that dominates all other contributions at high magnetic fields.[1] This is certainly established to be true in the majority of molecular conducting systems,[2] where the present experimental techniques do not lend themselves favourably to the detection of spin paramagnetic effects. Thus, if magnetic hysteresis is observed at high magnetic fields, one can assume with confidence that it is the orbital magnetism that is hysteretic; whether it corresponds to (1) first order phase transitions across which the orbital magnetism changes or (2) persistent currents.

Molecular conductors of the form α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ (where M = K, Tl or Rb) have become known to be plagued by hysteresis effects at high magnetic fields.[3] Most notable, is an abrupt change in the pattern of Landau quantization at and around fields of 23 T (for M = K), thought to be of the type (1) above. A change in shape and size of the de Haas-van Alphen (dHvA) oscillations[4] results from a change in the electronic structure caused by a transition between commensurate and incommensurate charge-density wave (CDW) phases.[5,6] Although this transition is driven primarily by changes in the spin paramagnetism of the groundstates,[7,8] it is the ancillary change in orbital magnetism that is detected experimentally.[3,4]

Another form of hysteresis comes into prominence at fields exceeding 23 T (for M = K), manifesting itself as a vertical offset in the dHvA oscillations between rising and falling fields.[3] This can be shown to be of type (2), above, by monitoring the behaviour of the irreversible magnetization as the magnetic field is cycled.[3,9,10] Figure 1A depicts a model hysteresis loop for a cylindrical sample of radius r within which currents flow at a critical current density j_c.[11] On the rising magnetic field (point 1) the irreversible magnetization is saturated at a negative value $M_{sat} = -j_c r/3$. On reversing the direction in which the applied magnetic field $\mu_0 H$ is swept (point 2), the irreversible magnetization changes according to the susceptibility $\chi' = -\eta$. As the magnetic field changes further, the polarity of the currents becomes

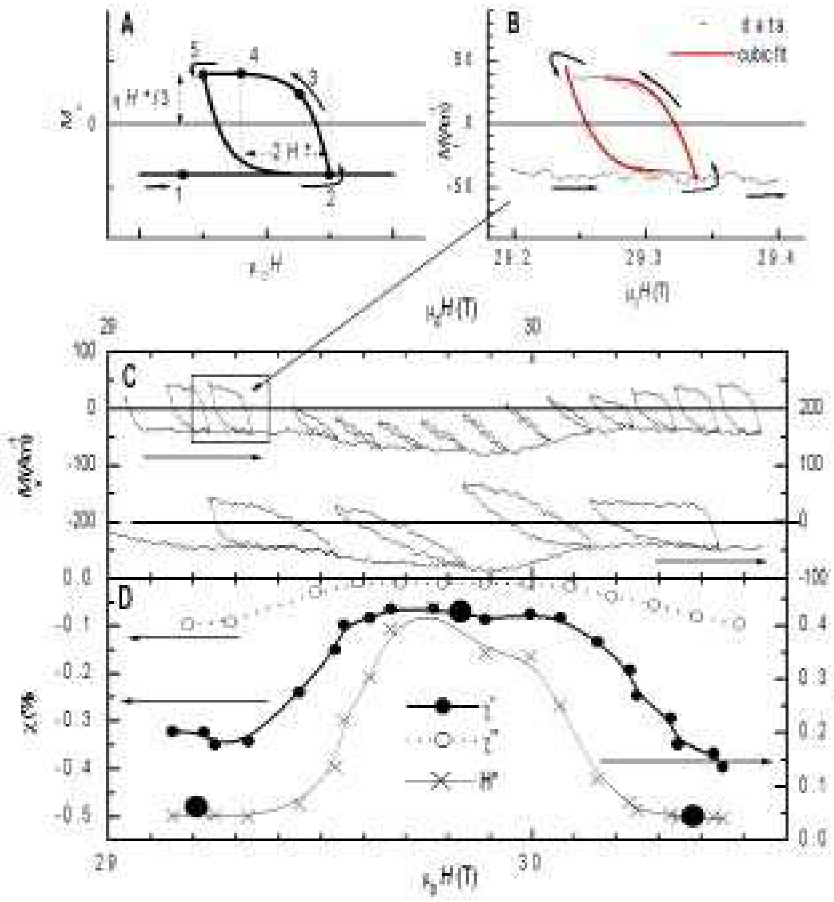

Figure 1. The irreversible magnetic properties of α-(BEDT-TTF)$_2$KHg(SCN)$_4$. (A) Model irreversible magnetization M_{ir} due to currents, as observed in type II superconductors. The dependence of M_{ir} on the magnetic sweep history as explained in the text. (B) An example of M_{ir} measured in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at 85 mK using magnetic torque with the angle between the applied magnetic field and the normal to the conducting planes being 18^o. (C) A series of loops measured over an extended range of field. (D) χ' χ" and H* extracted from M_{ir} in (C). Enlarged symbols correspond to the model calculations.

reversed within the sample, penetrating inwards in the form of a concentric flux-reversal front. The inductive response of the sample collapses quadratically, leading to a cubic lineshape in the irreversible magnetization (point 3). Finally the magnetization saturates again at $M_{sat} = +j_c r/3$ (point 4), having taken an interval in field equivalent to twice the coercion field $H^* = \eta M_{sat}$ to fully reverse all currents. Figure 1B shows actual data taken from magnetic torque measurements made on a sample of α-(BEDT-TTF)$_2$KHg(SCN)$_4$.[10] The cubic formula derived by Bean for type II superconductors fits the change in the irreversible magnetization in the flux reversal regime perfectly well,[11] implying that the inductive response of the sample collapses quadratically as in the simple model in Figure 1A.

The primary implication of the fits in Figure 1B is that the sample sustains a uniform gradient in the orbital magnetization $\nabla \times \mathbf{M}_{orb}$, with its orientation depending on the magnetic field history. By Maxwell's equations, this gradient is irreducibly equivalent to a current density $\mathbf{j}$ that attains a critical value $\mathbf{j}_c$. The origin of the current is subject to debate. Two kinds of effect, namely the quantum Hall effect[12] and superconductivity,[13] are commonly known to give rise to persistent or long-lived currents in metals with magnetic fields present. Both appear to be unlikely for the following reasons:

Quantum Hall effect:[12] Long-lived currents, here, occur orthogonal to a Hall electric field that is sustained owing to the absence of quasiparticle scattering effects orthogonal to the current. The failure of transport experiments to detect a persistent Hall electric field in static magnetic fields weighs heavily against the quantum Hall effect as a likely mechanism. Furthermore, the absence of quasiparticle scattering processes orthogonal to the current is normally achieved when the chemical potential μ is situated in a Landau gap. This cannot be the case at all filling factors at all magnetic fields: note that in Figure 1C, hysteresis loops are observed in α–(BEDT-TTF)$_2$KHg(SCN)$_4$ at all filling factors and at all fields above 23 T. It should also be noted that currents are carried by surface states in the quantum Hall effect: they are not expected to permeate the bulk as the experiments in Figure 1B indicate.

Superconductivity:[13] Persistent currents, here, are sustained in equilibrium when the vortex density gradient is held in place by vortex-impurity interactions. The magnetic behaviour in Figure 1B and transport behaviour in Reference 14 are both consistent with an inhomogeneous superconducting phase. The primary argument against such an explanation is that superconductivity tends to be destroyed by a magnetic field. Superconductivity is induced by a magnetic field only in rare instances where an internal antiferromagnetic exchange field is compensated by the applied field.[15] Evidence for significant internal magnetic fields have not been reported in any of the α–(BEDT-TTF)$_2$*M*Hg(SCN)$_4$ salts.[16,17] The electron *g*-factors are also too high to enable more exotic forms of field-induced superconductivity.[18]

Should neither of the above conventional mechanisms satisfactorily explain the experimental data in Figure 1, more exotic explanations must be considered, even if these explicitly involve the CDW groundstate. A mechanism involving the spontaneous sliding of CDWs (a variant of Fröhlich superconductivity) has been considered.[14] The extreme anisotropy of such a phase could account for the survival of the persistent currents to very high magnetic fields (at least approaching 100 T). It, nevertheless, requires the existence of unprecedented quantum depinning effects.

Manifestly similar effects to Fröhlich superconductivity can result even if the CDW is pinned. A model has recently been proposed that can explain why certain aspects of the behaviour of α–$(BEDT\text{-}TTF)_2M Hg(SCN)_4$ salts exhibit parallels with both the quantum Hall effect and type II superconductivity.[19] Orbital magnetism resulting from Landau quantization is an essential component of this model. Organic conducting salts of the form α–$(BEDT\text{-}TTF)_2M Hg(SCN)_4$ are subject to Landau quantization owing to the existence of a quasi-two-dimensional section of Fermi surface in addition to the quasi-one-dimensional sheets that become nested.[20] As a consequence of the strong variations in the density of states with field accompanying Landau quantization, carriers flow back and forth between the sections of Fermi surface as the magnetic field is swept.[21] The optimum nesting vector **Q** on the quasi-one-dimensional section of Fermi surface therefore undergoes oscillations in response to oscillations in the number of carriers between the quasi-one-dimensional sheets, causing the CDW to become periodically stretched and compressed like a concertina. Magnetic hysteresis results when this process is impeded by the pinning of the CDW to impurities.[19]

The hysteresis, here, is parameterized by a differential chemical potential $2\Delta\mu$ between the two Fermi surface sheets: a change in the chemical potential $-\Delta\mu$ of the quasi-two-dimensional Fermi surface pocket is compensated by an equal and opposite change $\Delta\mu$ in that of the quasi-one-dimensional Fermi surface sheets.[19] Rather than being uniform, however, the differential chemical potential exists in the form of a gradient, $2\nabla(\Delta\mu)$. If this were not the case, the hysteresis would be accompanied by a surface current that would exceed the critical current density. This is related to the characteristic CDW sliding threshold electric field by means of the formula $\mathbf{j}_c = (\mathbf{E}_t \times \mathbf{z})/\rho_{xy}$.[19] Such nehaviour results from the fact that the chemical potential gradients on the two sections of Fermi surface exist in equilibrium and oppose each other, while the gradient that exists on the quasi-one dimensional Fermi surface sheets is electrostatically limited such that $\nabla(\Delta\mu) < e\mathbf{E}_t$. The relation between $\mathbf{j}_c$ and $\mathbf{E}_t$, described above, is similar to the relation between **j** and **E** in the quantum Hall effect,[12] except that, here, there exists *zero* net electric field across the sample. A consequence of this behaviour is that the current becomes uniformly distributed within the interior of the sample upon saturation of the magnetization. The manner in the sample responds to changes in magnetic field is therefore similar to that of a type II superconductor, giving rise to hysteresis loops consistent with the Bean model, like those in Figure 1B.[11]

Figure 1C shows a series of hysteresis loop measurements made on a sample of α–$(BEDT\text{-}TTF)_2KHg(SCN)_4$.[10] Several quantities can be extracted from these data: the irreversible susceptibility χ', the losses incurred by the hysteresis χ", the coercion field H^* and the irreversible saturation magnetization M_{sat}. The quantities χ', χ" and H^*, thus extracted, are shown in Figure 1D. χ' and H^* are epsciallysensitive to the Landau level filling factor F/B, but can be accounted for by the model described in Reference 19. On reversing the direction of sweep of the magnetic field, CDW pinning initially prevents the CDW from sliding, except in the regions closest to the sample surface. The temporal dHvA susceptibility of the quasi-two dimensional Fermi surface becomes characteristic of a system in which there are no quasi-one dimensional Fermi surface states.[21] It is because the pinning disrupts the process be which quasiparticles flow back and forth between the two Fermi surface sheets that the susceptibility becomes hysteretic.[19] This effect can be modeled and estimates of χ' made from such a model (calculated in Reference 19) at integral and half-integral filling factors are indicated by oversized filled circular symbols in Figure 1D. The agreement between experiment and theory can be seen to be rather good.

The experimental results displayed in Figure therefore amount to compelling evidence for persistent currents occurring orthogonal to the charge polarization field of a CDW. The mechanism that gives rise to these currents appears to be unprecedented and quite unlike that giving rise to superconductivity or the quantum Hall effect.

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