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LETTER TO THE EDITOR

Experimental evidence for Fröhlich superconductivity in high magnetic fields

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Abstract

Resistivity and irreversible magnetization data taken within the high magnetic field CDW_x phase of the quasi-two-dimensional organic metal α -(BEDT-TTF)₂KHg(SCN)₄ are shown to be consistent with a field-induced inhomogeneous superconducting phase. In-plane skin depth measurements show that the resistive transition on entering the CDW_x phase is both isotropic and representative of the bulk.

Prior to the development of the BCS theory of superconductivity [1], Fröhlich proposed a novel form of superconductivity in one-dimensional (1D) metals due to spontaneously sliding charge-density waves (CDWs) [2]. CDWs were subsequently discovered in many materials containing quasi-one-dimensional (Q1D) Fermi-surface sections [3]; in such systems, a gap opens up over part or all of the Q1D Fermi-surface section(s) due to a nesting instability [3]. However, this gap, and the pinning of the CDW to impurities, generally prevent a CDW from contributing to the electrical conductivity, so that the electrical transport properties are dominated by any remaining normal carriers [3]. Conduction by sliding CDWs due to their depinning may occur under large electric fields, but is accompanied by considerable dissipation [3]. In this respect, the organic conductor α -(BEDT-TTF)₂KHg(SCN)₄ (together with the analogue compounds in which K has been substituted by Tl or Rb) may be an exception [4]. This highly anisotropic layered metal exhibits what is thought to be a CDW with an exceptionally low transition temperature $T_p \sim 8$ K [5–9]. At applied magnetic fields ($\mu_0 H$) greater than 23 T and temperatures (T) less than 2 K, α -(BEDT-TTF)₂KHg(SCN)₄ possesses a phase which carries dissipationless currents [10, 11].

In this letter, we present compelling evidence that these currents are conveyed by the CDW itself. Measurements of the skin depth reveal that the currents propagate deep into the bulk of the material. Furthermore, the drop in resistivity with decreasing temperature is shown to be isotropic, possessing a form characteristic of a transition into an inhomogeneous superconducting phase.

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Fröhlich's original model neglects interactions with impurities [2]; these spatially pin the CDW, preventing the spontaneous sliding motion required for superconductivity [3]. This pinning is often conveniently represented by a 'particle' in a sinusoidally-varying 'washboard' potential [3]. There are, however, limits in which this model should fail. For example, if the zero point energy of the 'particle' $\varepsilon_0 = \hbar^2 Q^2/4m^*$ exceeds the potential depth V , the washboard potential can no longer confine the CDW. Here, Q is the wavevector (or nesting vector) representing the spatial periodicity of the potential and $2m^*$ is the paired electron-hole mass, enhanced due to interactions between the CDW and the crystal lattice [3]. The degree to which m^* is renormalized with respect to the effective mass characteristic of the unperturbed band (m_b) is proportional to the square of the gap that opens at the Fermi energy on formation of the CDW [3].

The energy gap in α -(BEDT-TTF)₂KHg(SCN)₄ at $T = \mu_0 H = 0$ can be estimated using the BCS relation $2\Delta_0 = 3.52k_B T_p \approx 3$ meV [2, 3, 10]. It has been suggested that this gap is closed by the Zeeman energy when $\mu_0 \mu_B H/\sqrt{2} \approx 2\Delta_0$, corresponding to $\mu_0 H \approx 23$ T [7, 9]; this causes a first-order phase transition (the 'kink' transition) into a proposed spatially modulated CDW_x phase [7, 9, 10, 12–14], (see notional phase diagram in figure 1). α -(BEDT-TTF)₂KHg(SCN)₄ is therefore unusual in two respects: (1) it exhibits the only known CDW state in which Δ_0 is sufficiently small for such a transition to have been shown to occur in experimentally-accessible magnetic fields (a similar situation may be realized in (Per)₂M(mnt)₂ (where $M = \text{Au}$ or Pt), which has thus far only been subjected to fields of less than 20 T; see [15]); and (2) the reduced gap $2\Delta_x \approx 1$ meV that characterizes the CDW_x phase [10] is two orders of magnitude smaller than those in typical CDW materials [3].

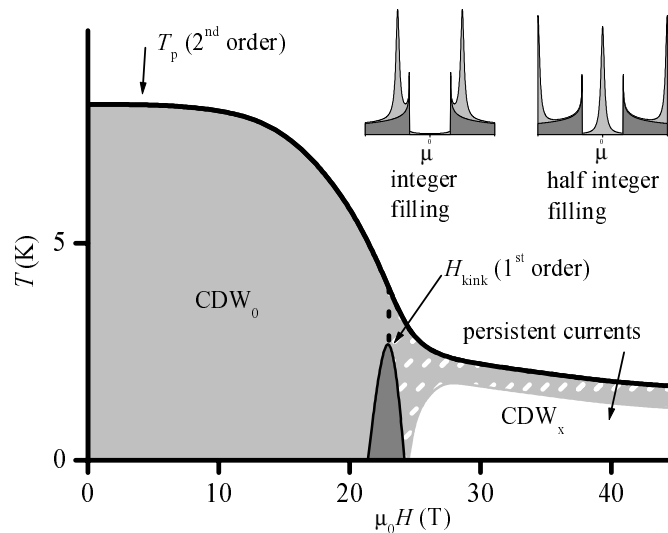


Figure 1. Notional phase diagram of α -(BEDT-TTF)₂KHg(SCN)₄ constructed from theoretical models [7, 12]) and data accumulated in this and other works [10, 13, 14, 16]). The solid line represents a second-order transition into the CDW phase (light shading) with a dotted line (together with the region of hysteresis (heavy shading)) representing a first-order transition between the proposed CDW₀ (solid shading) and CDW_x (hatched shading) phases. Persistent currents (white region) are observed only within the CDW_x phase. In the top right-hand corner, we show the density of states resulting from CDW formation within the 1D bands (heavy shading) together with the contributions from the Landau levels of the 2D band (light shading), both at integer and half integer filling factors. μ represents the position of the chemical potential.

The smallness of the gap leads to a negligibly small enhancement of m^* with respect to m_b (in contrast to that in more typical CDWs), leading to estimates of ε_0 in the range 10^2 – 10^3 meV (using $Q \approx 7 \times 10^9 \text{ m}^{-1}$ from [16]). Hence ε_0 greatly exceeds both $2\Delta_x$ and V , given that V must be less than Δ_x for the CDW to be stable [3]. These arguments lead one to expect that the CDW_x phase may undergo the spontaneous sliding necessary for Fröhlich superconductivity [2]. In the following, we will show that the resistivity and the magnetization of α -(BEDT-TTF) $_2$ KHg(SCN) $_4$ within the CDW_x phase do indeed exhibit behaviour characteristic of an inhomogeneous superconductor.

Figure 2 compares different measurements of the resistivity of α -(BEDT-TTF) $_2$ KHg(SCN) $_4$; the interplane resistivity component ρ_{zz} is measured using conventional four-terminal transport [10]. However, the highly anisotropic conductivity of α -(BEDT-TTF) $_2$ KHg(SCN) $_4$ makes it very difficult to measure the average in-plane resistivity ρ_{\parallel} using such techniques [16]. By contrast, the radio-frequency skin depth $\delta_{\parallel} = \sqrt{2\rho_{\parallel}/\mu_0\omega}$ enables both ρ_{\parallel} and the extent to which the currents penetrate the sample to be determined simultaneously [17]. The single-crystal sample is placed inside the inductor of a tunnel diode oscillator (TDO) tank circuit [17], with the oscillating magnetic field \tilde{h} perpendicular to

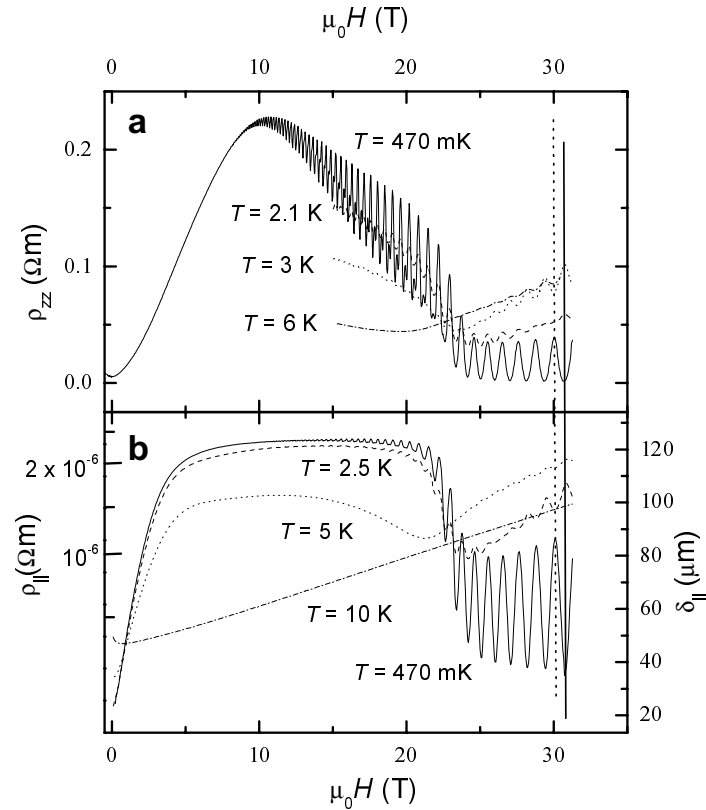


Figure 2. Examples of the interplane and in-plane resistivity components, ρ_{zz} and ρ_{\parallel} , of α -(BEDT-TTF) $_2$ KHg(SCN) $_4$ plotted versus dc magnetic field at different temperatures. ρ_{zz} (a) is measured using the conventional four-wire transport technique while ρ_{\parallel} (b) is estimated from the skin depth δ_{\parallel} (note that the ρ_{\parallel} axis is non-linear). The angle between the interplane b -axis of the sample and the dc magnetic field is $\sim 7^\circ$. The solid vertical line indicates an example of a magnetic field value where ν is an integer while the dotted line indicates one where ν is a half-integer.

the conducting layers. Screening within the sample results in a reduction in inductance, and consequently to an increase in the resonant frequency $\omega/2\pi$. Changes in frequency can be measured to high precision, with δ_{\parallel} being approximately related to ω by $\delta_{\parallel} \approx [\frac{1}{2} + (\omega - \omega_0)/\eta\omega_0]r$. Here, $\omega_0/2\pi = 38.975 \pm 0.005$ MHz is the resonant frequency without the sample, $\eta = 0.053 \pm 0.002$ is the effective filling factor of the sample with respect to the total inductance of the circuit and $r = 250 \pm 50$ μm is the effective sample radius [17].

Figure 2(b) shows ρ_{\parallel} extracted from δ_{\parallel} at several different temperatures. At temperatures $T > 8$ K, α -(BEDT-TTF)₂KHg(SCN)₄ displays the positive magnetoresistance exhibited by most organic metals [16]. At temperatures below 8 K, the magnetoresistance develops a semiconducting-type behaviour for $3 < \mu_0 H < 23$ T (i.e. ρ_{\parallel} increases with decreasing T), enabling \vec{h} to fully penetrate; however, above 23 T, the resistivity is lower, the change becoming more distinct as the temperature decreases. The Shubnikov–de Haas (SdH) oscillations in figure 2 are due to an additional two-dimensional (2D) band [18] that, while remaining largely unaffected by the CDW order, becomes quantized in a series of sharply-defined Landau levels at high magnetic fields [19]. Since the number of filled levels ν is proportional to $1/B$ (owing to an increase in the degeneracy of each level with magnetic induction $B \approx \mu_0 H$) [19], the field can be used to tune the position of the chemical potential relative to the Landau levels; at integer filling factors $\nu = F/B$, where F is the frequency of the SdH oscillations, the chemical potential resides in a Landau gap between the highest filled and lowest empty Landau level.

The data in figure 2 are very important, because they show that in almost all respects, including the detailed phase behaviour of the SdH oscillations, the variation of ρ_{\parallel} (estimated from δ_{\parallel}) with $\mu_0 H$ and T at high fields is identical to that of ρ_{zz} . (Of course ρ_{zz} is several orders of magnitude greater than ρ_{\parallel} owing to the anisotropy of the band structure [18]). We can therefore assert that the changes taking place in the resistivity as a function of $\mu_0 H$ and T are intrinsic, bulk properties of α -(BEDT-TTF)₂KHg(SCN)₄.

The temperature dependences of ρ_{zz} and ρ_{\parallel} are shown in figure 3 for both integer and half-integer ν . Remarkably, the temperature dependence of ρ_{zz} and ρ_{\parallel} at integer ν is exactly the same as a transition into an inhomogeneous superconducting phase [20–22]. In such systems, the (normally sharp) transition into the zero resistivity ground state becomes statistically broadened by a gaussian distribution with standard deviation ΔT_c about a mean critical temperature \bar{T}_c [20–22]. As can be seen in figure 3, the convolution of a sharp transition into a zero resistivity state with gaussian broadening (i.e. the error function) provides an excellent fit to both the ρ_{zz} and ρ_{\parallel} data at integer filling factors. This implies that an increasing fraction of the sample no longer contributes to the resistivity as T is lowered, in a manner consistent with the carriers (within this fraction) having condensed into a superconducting phase. As $\Delta T_c \approx 1$ K is a significant fraction of $\bar{T}_c \approx 2$ K, the transition is never entirely complete, so that the total resistivity of the inhomogeneous phase does not actually fall to zero at $T = 0$. Instead, the resistivity drops off exponentially in the limit $T \rightarrow 0$ (i.e. $\rho \propto e^{T/T_0}$) in a manner similar to that observed in several well known examples of inhomogeneous superconductors [23, 24]. This resistivity model can also be fitted to the data at half-integer filling factors with a very similar \bar{T}_c , but with the transition width ΔT_c being considerably broadened. The fact that the midpoint of the transition \bar{T}_c is virtually unaffected by the Landau-level filling factor of the 2D quasiparticles (see figure 1 inset) is consistent with the gap formation being solely determined by the Q1D sheets. The Q2D Landau density of states appears only to modulate the width of the transition, possibly as a result of its influence on the scattering rate; there is an increased density of available states at the chemical potential at half-integer filling factors (see figure 1 inset) [16].

Finally, we turn to the magnetization, which exhibits a behaviour entirely consistent with that of an inhomogeneous superconductor in the CDW_x phase. Figure 4(a) shows an example

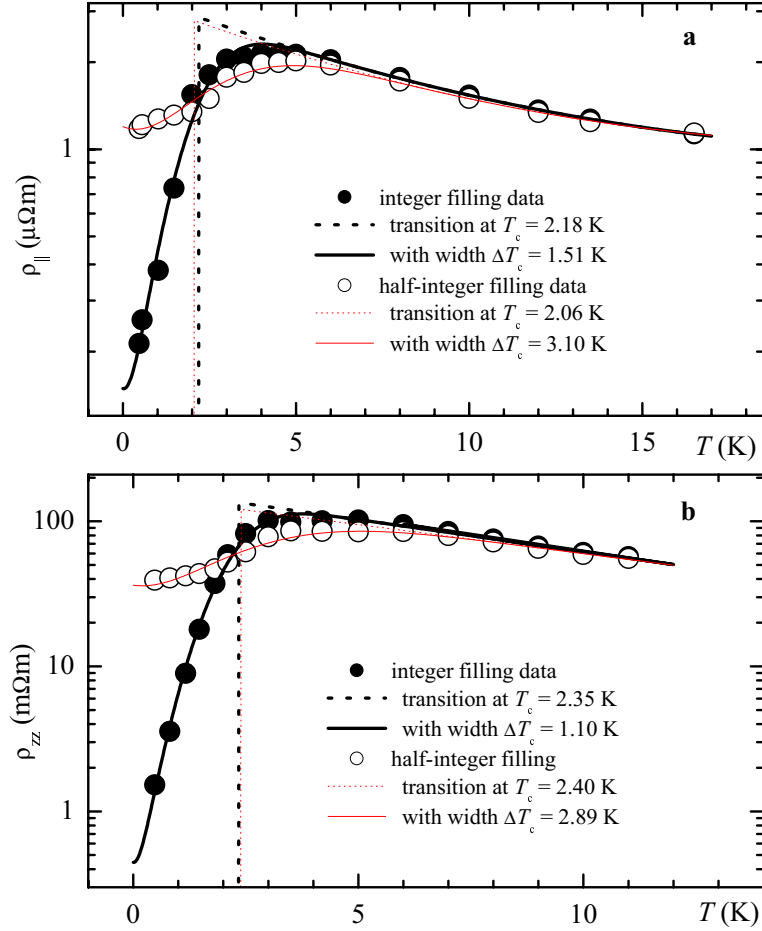


Figure 3. Temperature dependence of the resistivity. (a) The in-plane resistivity $\rho_{||}$ (filled circles) extracted from $\delta_{||}$ at integer filling factors (at $\mu_0 H \approx 30$ T), together with a resistivity extrapolation (using a polynomial fit) from within the normal phase down to the midpoint of the transition \tilde{T}_c (heavy dotted line). Below \tilde{T}_c the resistivity is zero. On multiplying the extrapolated resistivity by the function $\frac{1}{2}(\text{erf}[(T - \tilde{T}_c)/\Delta T_c] + \text{erf}[-(T - \tilde{T}_c)/\Delta T_c] + 2)$, we obtain excellent agreement (heavy solid line) with the experimental data (filled circles). The same procedure is also performed on the data at half-integer filling factors (but with open symbols and thinner lines), leading to a very similar value of \tilde{T}_c but broader transition. (b) The same analysis repeated on the interplane resistivity data taken from [10].

of a hysteresis loop in the irreversible magnetization M obtained using a torque magnetometer (see [10] for a detailed description of this technique); two important features of this loop are consistent with Bean's critical state model for vortex pinning in type II superconductors [25]. Firstly, the irreversible magnetic susceptibility $\partial M/\partial H$ is diamagnetic (i.e. negative) on reversing the direction of sweep of H [10]; this effect can only be ascribed to circulating currents [10, 11] (while circulating currents are not expected in a purely 1D CDW system, they can occur in 2D CDW systems such as α -(BEDT-TTF) $_2$ KHg(SCN) $_4$ [16, 26]). Secondly, the magnitude of the hysteresis saturates at a critical value M_{sat} enabling us to identify a critical current density j_c [10, 25]. Because the hysteresis occurs at all filling factors ν , the currents

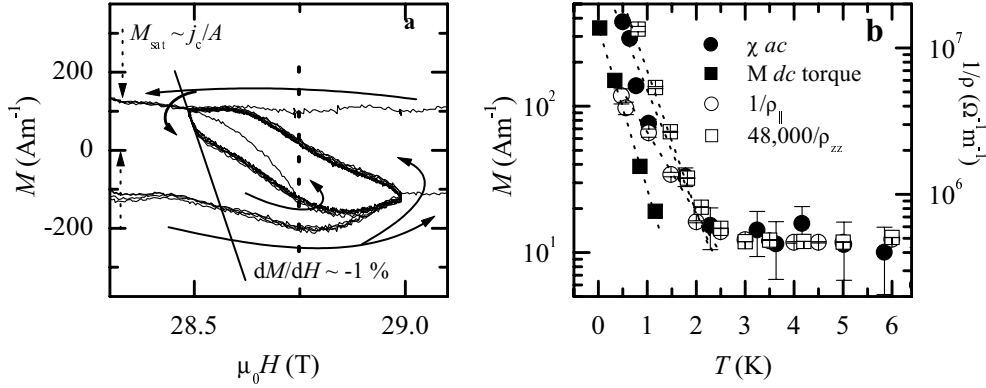


Figure 4. Examples of magnetic measurements made on α -(BEDT-TTF) $_2$ KHg(SCN) $_4$. (a) An example of a hysteresis loop in the irreversible magnetization M estimated from the magnetic torque measured at an angle of $\sim 7^\circ$ between the inplane b -axis of the sample and the magnetic field. Solid arrows indicate the direction of sweep of the magnetic field, often swept over the same interval in field to show reproducibility. The dashed arrows indicate the saturated irreversible magnetization M_{sat} , while the inclination of the solid straight line indicates the negative differential susceptibility $\partial M/\partial H$. The vertical dotted line indicates the field at which ν is half integer. (b) Comparisons of the irreversible magnetization of the currents measured by means of ac susceptibility and magnetic torque (filled symbols) with the conductivity $1/\rho$ (open symbols), versus T , with dotted lines drawn merely to guide the eye. In an inhomogeneous superconductor, both approximately scale with the fraction of the sample in which currents are conveyed without dissipation.

cannot originate from the quantum Hall effect (QHE), as earlier experiments had appeared to suggest [16]. In the limit $\nu \gg 1$ that applies here, the QHE is expected to operate only at integer filling factors [27].

Thus, our reasons for attributing these effects within the high magnetic field phase of α -(BEDT-TTF) $_2$ KHg(SCN) $_4$ to Fröhlich superconductivity [2] can be summarized as follows:

- the semiclassical model for CDW pinning is expected to fail [3]
- both the magnetization and the resistivity display effects found only in superconductors; the induced currents are consistent with Bean's critical state model [10, 25], while the resistivity has exactly the form of a transition into an inhomogeneous superconducting phase [20–22]
- these effects are observed at *all* filling factors for $\mu_0 H > 23$ T
- only a single resistive transition is observed as a function of T on entering the region in H and T over which the currents are observed, with the midpoint \bar{T}_c of this transition (being virtually independent of the 2D density of states) coinciding with the transition into the CDW $_x$ phase predicted by theory [7, 12].

The small diamagnetic fraction ($-\partial M/\partial H \sim 1\%$) [10], large skin depth ($\delta_{\parallel} > 30 \mu\text{m}$, being an appreciable fraction of r) and broadness of the resistive transition ($\Delta T_c \sim 1\text{--}3$ K) all suggest that the persistent currents are confined to many small islands or filaments throughout the sample surrounded by regions that are normal. Figure 4(b) provides further support for this picture, where it is apparent that both the size of the irreversible magnetization M and the conductivity $1/\rho$ obey an approximate e^{-T/T_0} law in the limit $T \rightarrow 0$ (see figure 4(b)). It is interesting that the transition width ΔT_c is so broad for a material that should most definitely be in the clean limit. We can infer this from the fact that the mean free path $l = v_F \tau \sim 4000 \text{ \AA}$ (estimated from the results of de Haas–van Alphen measurements

[28], where $v_F \sim 80,000 \text{ ms}^{-1}$ is the Fermi velocity) greatly exceeds the coherence length $\zeta_0 = \hbar v_F / \pi \Delta \sim 200 \text{ \AA}$ (conservatively estimated using the BCS formula [1, 3]). Should α -(BEDT-TTF)₂KHg(SCN)₄ possess a vortex state, inhomogeneity could be a consequence of the transition being broadened by a magnetic field in a similar manner to that observed in other highly anisotropic superconductors [29, 30]. Another possibility to consider is that, unlike Cooper pairs in conventional superconductors [1], a CDW charge modulation experiences additional Coulomb interactions with impurities [3]. While the Coulomb force may not necessarily be able to pin the CDW condensate if Δ is too small, it may still affect the stability of the CDW order should V become comparable to Δ (see [3]).

In conclusion, both transport and magnetization measurements within the low-temperature, high-magnetic-field CDW_x phase of α -(BEDT-TTF)₂KHg(SCN)₄ are shown to be consistent with inhomogeneous superconductivity, realized in small filaments or islands throughout the bulk of the sample. The locality of this effect with respect to the CDW phase diagram in H and T leads us to propose that Fröhlich superconductivity (or perhaps a 2D variant thereof [26]) is realized in this material at high magnetic fields.

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