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## Possible Fröhlich superconductivity in strong magnetic fields

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### Abstract

A brief review of some of the arguments pointing towards the possibility of organic conductors of the form  $\alpha$ -(BEDT-TTF)<sub>2</sub>MHg(SCN)<sub>4</sub> (where  $M$ =K, Tl and Rb) being candidates for Fröhlich superconductivity is given.

**Keywords:** superconductivity, high magnetic fields, one dimensional

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The presence of a magnetic field within a sample generally inhibits superconductivity [1]. In rare cases where superconductivity is observed to be induced by an external magnetic field, this occurs only because the external magnetic field compensates an internal antiferromagnetic exchange field, as recently realized in  $\lambda$ -(BETS)<sub>2</sub>FeCl<sub>4</sub> [2]. The superconductivity is therefore not field-induced with respect to the intrinsic magnetic field seen by the Cooper pairs.

Charge transfer salts of the form  $\alpha$ -(BEDT-TTF) $_2$ MHg(SCN) $_4$  (where  $M = \text{K, Tl and Rb}$ ) could potentially be the first examples of *true* field-induced superconductors [3]. A number of experimental observations are shown to be consistent with a superconducting state existing at fields exceeding  $B_k \sim 23 \text{ T}$  for  $M = \text{K}$  or  $32 \text{ T}$  [3-6] for  $M = \text{Rb}$  [6], while experiments employing  $\mu\text{SR}$  and NMR techniques find little evidence for an intrinsic magnetic field [7,8]. More unusually,  $\alpha$ -(BEDT-TTF) $_2$ MHg(SCN) $_4$  salts are thought to possess charge-density wave (CDW) ground states at low magnetic fields [9-13]. According to mean field theory, the phase that exists at fields above  $B_k$  (where properties akin to superconductivity are observed) is expected to be an exotic form of CDW where the up-spin and down-spin components of the quasi-one-dimensional Fermi surface sheets nest independently [9,14]. This invokes the question as to whether a CDW under these previously unprecedented conditions can develop some of the characteristics of a superconductor in the manner originally predicted by Fröhlich [15].

The above question results after approximately ten years of research on these materials by several different groups. The first order phase transition that takes place at  $B_k$  is thought to be caused by the low magnetic field, low temperature CDW phase reaching its Pauli paramagnetic limit [9,11,12]. At magnetic fields higher than  $B_k$ , the free energy of a conventional CDW state becomes higher than that of the normal metal or a CDW phase in which the separate spin Fermi surface sheets nest independently [4,9,14].

The existence of magnetic hysteresis in the form of an offset between rising and falling fields, at all fields exceeding  $B_k$ , has been known for some time [16]. Only recently, however, has this hysteresis been shown to be connected with the eddy currents that are observed in pulsed magnetic fields [5,6,17]. The presence of currents becomes especially clear when magnetic hysteresis measurements made using magnetic torque [6] are compared with magnetic measurements made on type II superconductors [18] in Fig. 1. Although the field is cycled over a short interval at very high magnetic fields in Fig. 1b, the basic form of the hysteresis loop remains the same as that in Fig. 1a. Notably, the susceptibility  $\mu_0 dM/dB$  is largest and diamagnetic immediately after the sweep direction of the external magnetic field is reversed. On extracting this slope  $dM/dB$  from many hysteresis loops over a wide range of magnetic fields [6], the susceptibility (thus obtained in Fig.2a [6]) is found to agree with the results of *ac* susceptibility measurements in Fig 2b [5]. Small differences in the magnitude of the

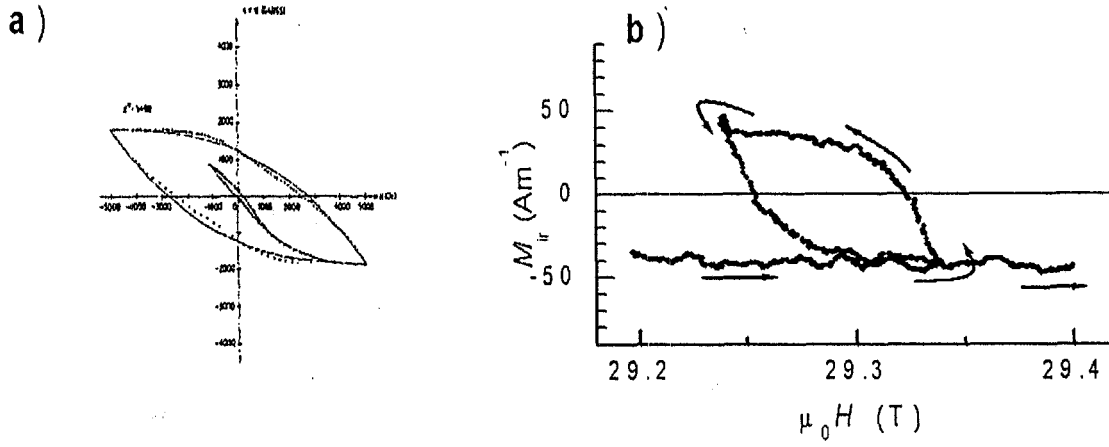


Fig. 1. Comparison of measurements of the irreversible magnetization made on lead at low magnetic fields and  $\sim 4.2$  K (a) [18] with those on  $\alpha$ -(BEDT-TTF) $_2$ KHg(SCN) $_4$  at high magnetic fields and  $\sim 80$  mK (b) [6].

susceptibility results from the fact that each of the samples in Figs. 2a and 2b are from different batches. Nevertheless, the existence of a hysteretic magnetization exhibiting precisely the same behaviour as those observed in type II superconductors combined with a strong diamagnetic response in *ac* susceptibility measurements firmly establishes the existence of persistent currents of a critical magnitude  $j_c$  [6]. The persistence of the currents at all magnetic fields above  $B_k$  to fields in excess of 60 T rules out explanations involving the quantum Hall effect or phase transitions involving changes in Landau diamagnetism. The

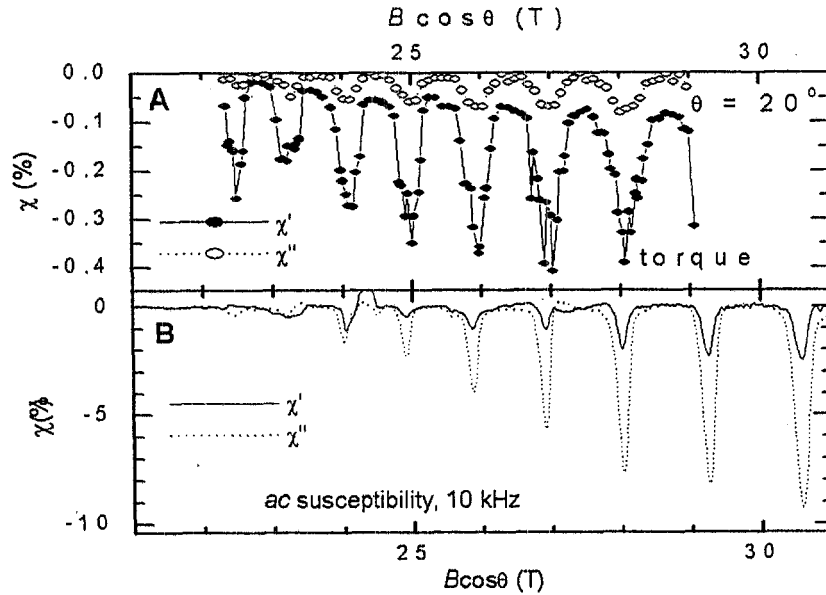


Fig. 2. Comparison of the real  $\chi'$  and imaginary  $\chi''$  components of the susceptibility extracted from measurement of the slope of the magnetic torque at  $\sim 80$  mK (a) [6] and *ac* susceptibility at  $\sim 0.5$  K (b) [5].  $\theta$  is the angle between the applied magnetic field and the normal to the conducting planes.

magnitude of the  $ac$  susceptibility is also several orders of magnitude too large to be explained in terms of Landau diamagnetism. Given that this type of behaviour is only observed in type II superconductors with pinning [18], superconductivity, of some form, appears to be the only reasonable explanation.

Oscillations in the density of states of a closed Fermi surface pocket (often referred to  $\alpha$ -pocket) appear to modulate the magnitude of the  $ac$  susceptibility in Fig. 2a in a manner that has yet to be understood. The  $ac$  susceptibility is largest at integer filling factors, when the chemical potential is situated in a Landau gap, though present at all filling factors at fields above  $B_k$ .

Were superconductivity, of some form, involved, we should expect to observe a dramatic increase in conductivity (or reduction in resistivity) at the same low temperatures where persistence currents are observed inductively. Quantum oscillations in the magnetoresistance have been known to be unusual for some time [19,20]. However, when the resistivity is plotted versus temperature  $T$  at a fixed magnetic field [3] in Figs. 3a and 3b, a dramatic drop in resistivity can be seen to occur (by as much as 2 orders of magnitude) at the same fields where the  $ac$  susceptibility is maximum, and for all current directions [3]. A lesser drop occurs at all other fields. The solid lines represent a model in which a sharp superconducting transition is broadened by gaussian. Fits to this model appear to be surprisingly good, and suggests that the form of the resistivity at temperatures below approximately 3 K has the form of a broadened superconducting transition. This could be consistent with an inhomogeneous superconducting phase.

Figure 4a shows the results of fitted values of the midpoint of the transition  $T_c$  and the width of the transition  $\Delta T_c$  plotted versus  $B$ : good fits are only obtained at fields above  $B_k$ . One can conclude from this plot that the width of the transition (in addition to the magnitude of the  $ac$  susceptibility) is modulated by the density of states of the  $\alpha$ -pocket. The thermodynamic midpoint of the transition  $T_c$ , however, appears not to be modulated at all. A  $T_c$  that is independent of the density of states of the  $\alpha$ -pocket implies that the order parameter is only associated with the quasi-one-dimensional Fermi surface sheets: the same Fermi surface sheets that nest when the low magnetic field, low temperature CDW phase is formed. There is therefore no doubt that the thermodynamic ground state at high magnetic fields is primarily

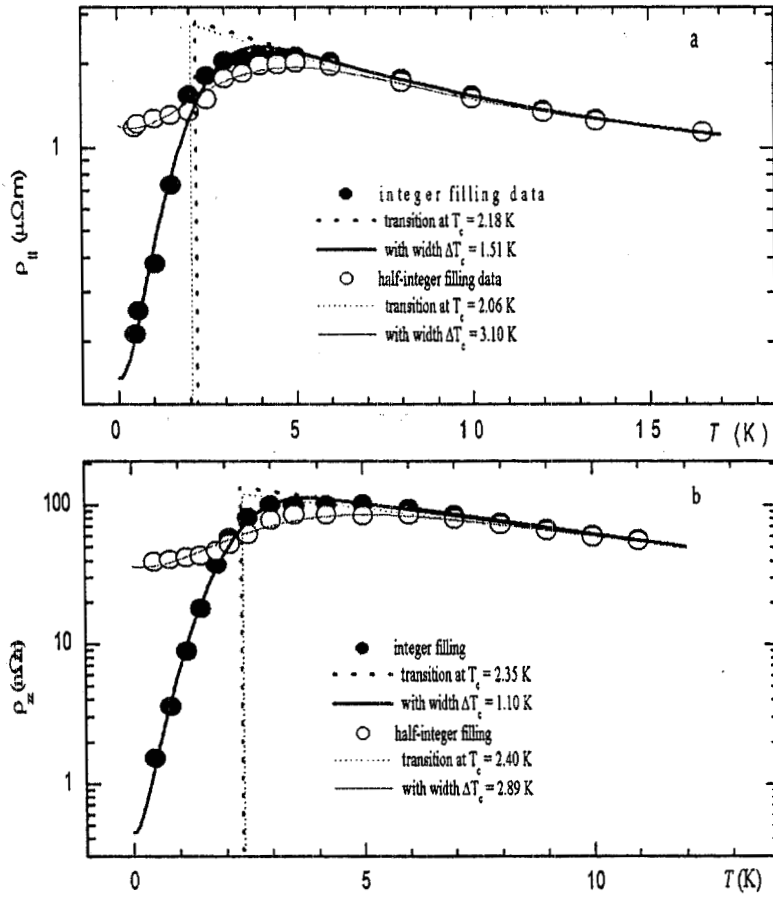


Fig. 3. Temperature dependence of the in-plane resistivity (a) of  $\alpha$ -(BEDT-TTF) $_2$ KHg(SCN) $_4$  extracted from skin depth measurements and inter-plane resistivity (b) made using conventional 4-wire techniques [3]. At integer filling (where the resistivity drop and  $ac$  susceptibility are greatest), the chemical potential is situated in a gap between Landau levels of the  $\alpha$ -pocket.

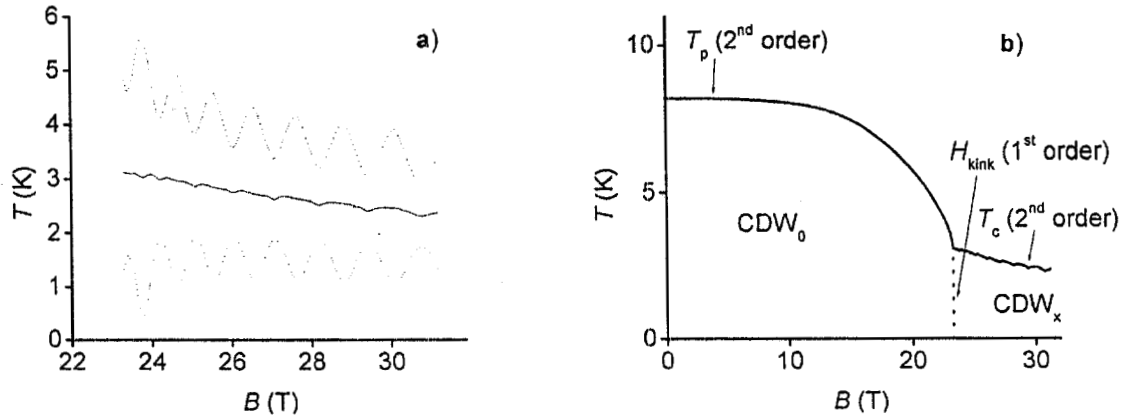


Fig. 4. (a) The transition  $T_c$  (solid line) extracted from the fits (using the method described in the text and in more detail in Ref. [3]), together with dashed lines corresponding to  $T_c + \Delta T_c/2$  and  $T_c - \Delta T_c/2$ . (b) The phase diagram after including  $T_c$  from the fits.

associated with the quasi-one-dimensional sheets. This observation is consistent with the mean field theory of CDW's [9,14]. The physical properties of this high magnetic field phase, however, closely resemble those of a superconductor, and are not at all like those of a CDW. It is for these reasons that we propose  $\alpha$ -(BEDT-TTF)<sub>2</sub>MHg(SCN)<sub>4</sub> as a strong candidate for Fröhlich [15] superconductivity. On combining the  $T_c$  data (solid line) with existing data on the transition  $T_p$  into the CDW phase [4,13] (solid line) and the first order transition field  $B_k$  (dotted line), all three lines can be seen to meet at a tri-critical point.

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