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The Phase Diagram of α-$(ET)_2TlHg(SCN)_4$: An Electrodynamic Investigation

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Abstract

We have probed the phase diagram of α-$(ET)_2TlHg(SCN)_4$ using a mm-wave technique. A substantially different behavior is seen for the conductivity parallel and perpendicular to the 2D layers as the various phase boundaries are cut. We compare our findings with published data and discuss our results in terms of recent models for the low-temperature phase diagram of this family of organic metals.

Keywords: Magnetotransport, Conductivity, Organic conductors

1. Introduction

α-$(ET)_2TlHg(SCN)_4$ belongs to a family of organic conductors that have been the subject of extensive experimental and theoretical investigation [1]. Their electronic structures are highly anisotropic due to a physical structure which consists of layers of ET molecules separated by anion sheets. Conduction occurs predominantly within the ET layers, while the conductivity normal to these layers is some three orders of magnitude lower. Band structure calculations predict a 300 K Fermi surface (FS) consisting of a quasi two-dimensional (Q2D) hole pocket and a pair of quasi one-dimensional (Q1D) electron sheets [1].

Below ~10 K, α-$(ET)_2TlHg(SCN)_4$ undergoes a phase transition which is believed to be driven by a nesting instability between the Q1D FS. There has been considerable debate as to whether this instability results in a modulation of the charge or spin density. In either case, a reconstruction of the room temperature FS is expected to occur.

In this investigation, we take a subtle approach to this problem. For certain geometries, and in high magnetic fields, it is possible to measure an AC conductivity which scales with the scattering time τ - as opposed to the classical DC magnetoresistance which scales with mobility $e\tau/m^*$. In doing so, we find evidence for a new low temperature phase at magnetic fields above the so-called "kink" transition.

2. Experimental details

We use a Millimeter-wave cavity perturbation technique (described elsewhere [2]). Experiments were performed in two configurations in which currents were either excited within the highly conducting layers, or normal to this direction. Due to the huge anisotropy in the conductivity, these two cases probe very different quantities. For the in-plane configuration, dissipation occurs due to a surface resistance R_S. For the inter-plane configuration, currents are excited throughout the bulk of the sample and dissipation is due to the real part of the conductivity σ_1.

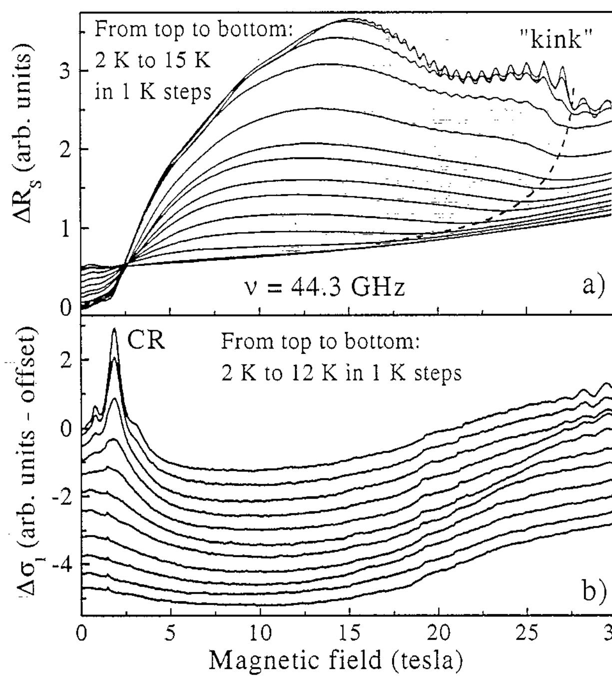

Fig. 1: a) In-plane, and b) inter-plane measurements of the field dependent electrodynamic response of α-$(ET)_2TlHg(SCN)_4$.

3. Results

Fig. 1 shows the in-plane and inter-plane electrodynamic response of α-$(ET)_2TlHg(SCN)_4$. Fig 1a shows many features which are well known from DC measurements [1]; *e.g.* at low temperatures, R_S rises sharply in field, reaching a maximum at a field of ~15 T; then, at about 27 T, there is a sharp "kink" in R_S. The dashed line shows the temperature dependence of this "kink". At the lowest temperatures, quantum oscillations are seen.

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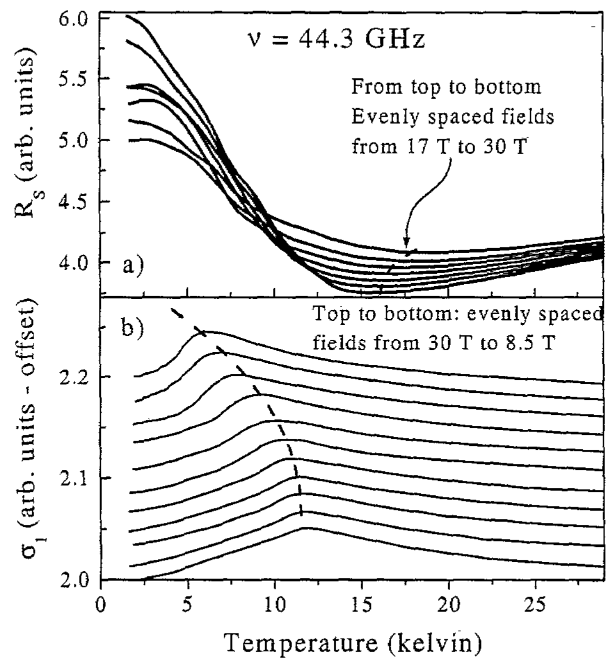

Fig. 2: T dependence of a) in-plane R_s, and b) inter-plane σ_1.

The main oscillation frequency (670 T) agrees with published data [1]. There is also a much slower oscillation (not periodic in 1/B), which is not understood. At very low fields (~2 T), there is kink in R_S which we attribute to cyclotron resonance (CR) [2,3].

The data in Fig. 1b is inverted with respect to the data in Fig. 1a, since the conductivity rather than the surface resistance is probed. The most striking differences between Figs 1a and b are the absence of any obvious "kink", the very strong CR feature at low fields, and the relative weakness of the quantum oscillations.

Fig. 2 show the temperature dependence of in-plane R_S, and inter-plane σ_1, for a range of magnetic fields. The slopes of all of the curves change sign at a field dependent temperature, indicating possible phase transitions. However, the field dependence is entirely different for the two figures (see dashed curves).

4. Discussion

In order to understand the differences in the in-plane and inter-plane responses, we appeal to a semiclassical description for the AC conductivity. A general form for the diagonal components is

$$\sigma_{rr} = \frac{Ne^2\tau}{m_{rr}} \sum_{n=0}^{\infty} C_{r,n} \times \frac{1 + i(\omega \pm n\omega_c)\tau}{1 + (\omega \pm n\omega_c)^2 \tau^2},$$

where r denotes the specific component (*i.e.* x, y or z), the $C_{r,n}$ are coefficients which depend on the FS topology, and $\omega_c = eB/m_{xx}$ is the cyclotron frequency [4]. It is assumed that the DC magnetic field is applied parallel to the least conducting (z-) axis.

It is well known that the effective masses in the α-phase ET salts are of order m_e [4]. Thus, at fields above 15 T, we make the assumption $n\omega_c >> \omega$ (for $n > 0$). The coefficient $C_{r,0}$ is equal to zero for the in-plane Q2D conductivity (*i.e.* $r = x$ or y), since it has no open orbit contribution. In contrast, $C_{r,0}$ is finite for σ_{zz}, and any Q1D contribution to σ_{rr}. If we also assume that $\omega\tau > 1$, which must be the case since we see CR, then the real part of the high-field in-plane conductivity, σ_{xx}, is given approximately by:

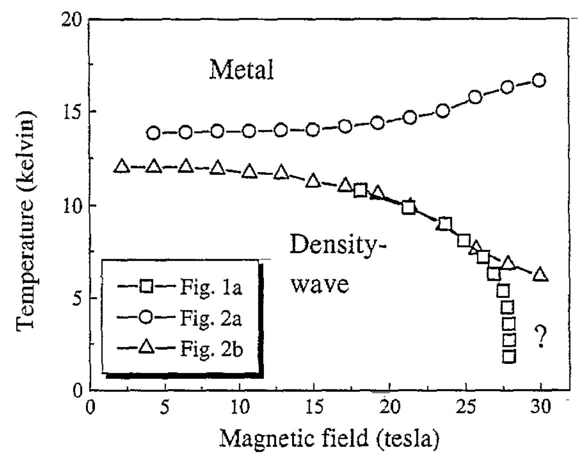

Fig. 3: Phase diagram obtained from the data in Figs 1 and 2.

$$\sigma_{xx} = \frac{Ne^2\tau}{m_{xx}} \sum_{n=1}^{\infty} C_{r,n} \times \frac{1}{n^2(\omega_c\tau)^2};$$

this expression is also valid for the DC conductivity. In contrast, the inter-plane AC conductivity additionally requires the term,

$$\sigma_{zz,0} = \frac{Ne^2\tau}{m_{zz}} C_{z,0} \times \frac{1}{1+(\omega\tau)^2},$$

in the summation. In fact, this term should dominate σ_{zz} at high fields, since $n\omega_c >> \omega > 1$. Thus, we see that σ_{xx} scales with the mobility $e\tau/m_{xx}$, whereas the σ_{zz} is dominated by the $n = 0$ term which does not depend on the in-plane effective mass, m_{xx}.

Now we can account for the differences in the two sets of data; we will do so with reference to the phase diagram in Fig. 3. At the "kink", the mobility must change, but not τ, since the "kink" is not seen in σ_{zz}, *i.e.* m_{xx} must be renormalized at the "kink". In contrast, the phase line indicated by triangles marks a point where either m_{zz} is renormalized, or m_{xx} and τ are renormalized equivalently so that the mobility is unchanged.

The region in the phase diagram labeled with a question mark was previously believed to belong to the high temperature metallic phase. From these studies, it is apparent that this is not the case. A phase diagram rather similar to this has been proposed by McKenzie [5] for a charge-density-wave (CDW), where the "kink" separates two different CDW phases. The line indicated by circles in Fig. 3 has been seen by other authors [6]. However, It is not clear whether this is a real phase boundary or due to magnetoresistance.

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