Cyclotron resonance studies of the molecular conductor $d_{2}[1,1;0]$-(DMe-DCNQI)$_2$Cu

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Abstract

We have observed cyclotron resonance in the three dimensional organic conductor $d_{2}[1,1;0]$-(DMe-DCNQI)$_2$Cu. An effective mass of $(3.35\pm0.2)m_e$ is obtained, in excellent agreement with the effective mass deduced from de Haas-van Alphen measurements. This result rules out the possibility that electron-electron interactions play an important role in these materials.

Keywords: Organic conductors, Metal-insulator phase transition, Many-body and quasiparticle theories

1. Introduction

Recently, an organic conductor containing Cu ions (DMe-DCNQI)$_2$Cu (DMe-DCNQI = 2,5-dimethyl-N,N'-dicyanoquinodimine) has been synthesised.$^1$ A 1D Fermi surface is formed by the overlapping $pt$-orbitals of the stacked DCNQI molecules.$^2$ In addition, a 3D energy band is expected due to the Cu ions, which form a seven fold diamond-like lattice and are found to be in a mixed valence state.$^3$ Recent de Haas-van Alphen (dHvA) studies by Uji et al.$^4$ have confirmed the existence of a 3D Fermi surface, in agreement with bandstructure calculations.$^2$

The partially deuterated $d_{2}[1,1;0]$-(DMe-DCNQI)$_2$Cu salt shows re-entrant metal-insulator-metal (M-I-M) phases.$^3$ The I-phase is believed to arise from a static ordering of the Cu ions. An enhancement of the electronic specific heat coefficient ($\gamma$) close to the I-phase has been interpreted by some groups as evidence that electron-electron interactions (EEIs) drive the M-I and I-M transitions.$^5$ However, other groups fail to observe an enhancement of $\gamma$ and, so, the issue as to whether EEIs are significant is still controversial.$^6$

A dHvA measurement yields a quasiparticle effective mass, $m^*$, which includes renormalizations due to electron-phonon interactions and EEIs.$^7$ In contrast, cyclotron resonance (CR) probes the dynamical mass, $m_d$, which is insensitive to EEIs.$^3$ Therefore, a comparison between dHvA and CR masses is an obvious way to assess the role of EEIs in the title compound. Such a comparison is particularly important, since tight binding bandstructure calculations only predict the ratios of effective masses corresponding to different Fermi surface sections, $i.e.$ they do not reliably produce the absolute values.$^8$

2. Experimental details

The partially deuterated $d_{2}[1,1;0]$-(DMe-DCNQI)$_2$Cu salt was chosen for this study because of its re-entrant M-I-M behaviour. A Millimeter-wave Vector Network Analyzer$^9$ (MVNA) was configured to monitor the reflected phase and amplitude of radiation coupled to a resonant cavity containing the samples. An oversized cylindrical copper cavity was used, providing many modes in the desired frequency range. A Styrofoam sample holder, containing four needle-like samples, was placed close to the bottom of the cavity so as to excite a.c. currents in the $ab$-plane of the samples; the applied magnetic field was directed normal to the $ab$-plane. Cylindrical waveguides coupled radiation from the MVNA to the cavity, through a $^4$He cryostat situated in the magnet bore. An optical beam splitter separated the incident and reflected waves. The samples and cavity were maintained at 1.2 K, at the maximum magnetic field position within a 20 tesla Bitter-type magnet. All measurements were performed at a fixed frequency, while sweeping the magnetic field.

3. Results and data analysis

We utilize the cavity perturbation technique, in the skin depth regime,$^{10,11}$ to evaluate the complex surface impedance, $Z_S = R_S + iX_S$, of the samples. In this way, we can relate changes in phase ($\Delta \phi$) and amplitude ($\Delta A$), of the radiation reflected from the cavity, to changes in the cavity resonance frequency ($f_0$) and quality factor ($Q$);$^{11} f_0$ and $Q$ are, in turn, related to $R_S$ and $X_S$. Figure 1 shows an example of such a data transformation.

![Figure 1](image-url)
The above analysis is valid when the penetration depth (skin depth) is much smaller than the sample dimensions.\textsuperscript{10} We estimate the skin depth of our $d_2[1,1,0]$-(DMe-DCNQI)$_2$Cu samples to be about 1 $\mu$m at liquid helium temperatures;\textsuperscript{12} the smallest sample dimension is about 100 $\mu$m. It is for this reason that we consider the surface impedance, $Z_S$, of the sample. $Z_S$ is related to the complex conductivity ($\sigma = \sigma_1 - i\sigma_2$) by the following expression:\textsuperscript{10}

$$Z_S = \left( \frac{i\mu_0 \omega}{\sigma_1 - i\sigma_2} \right)^{1/2}.$$  \hspace{5cm} (1)

For the measurement geometry described in section 2, it should be possible to observe CR due to a single closed orbit ($\alpha$-orbit) on the Fermi surface.\textsuperscript{4} We use the following semi-classical expression for the conductivity to understand the behaviour of $R_S$ and $X_S$, as seen in Figure 1.

$$\sigma = \frac{1}{2} \sigma_0 \left[ \frac{1 + i(\omega - \omega_c)^2}{1 + (\omega - \omega_c)^2} \tau - \frac{1 + i(\omega + \omega_c)^2}{1 + (\omega + \omega_c)^2} \tau \right]$$  \hspace{5cm} (2)

In the above expression, $\sigma_0$ is the zero field d.c. conductivity, $\omega_c (= eB/m_0$) is the cyclotron frequency, $\omega$ is the radiation frequency and $\tau$ is the relaxation time; $m_0$ is the CR mass due to the $\alpha$-orbit and $B$ is the applied magnetic field. Eq. 2 includes both resonant (left hand term in brackets) and non-resonant (right hand term in brackets) contributions to the conductivity; this is due to the fact that the radiation in the cavity is a superposition of left and right circularly polarised components. No special considerations are made about the exact shape of the Fermi surface, i.e. Eq. 2 is generalised for a free electron gas.

The fits in figure 1 (dotted lines) have been obtained by substituting Eq. 1 into Eq. 2 and then adjusting $m_0$, $\tau$ and two prefactors.\textsuperscript{11} In doing so, we obtain a CR mass $m_0 = (3.34 \pm 0.2)m_e$ and a scattering rate $\tau^{-1} = 2 \times 10^{10}$ s$^{-1}$. Agreement between experiment and analysis is good over the entire frequency range studied (70 - 130 GHz); this can be seen in figure 2.

The average CR mass obtained from the data in figure 2 is $m_0 = (3.34 \pm 0.2)m_e$. To within the experimental errors, this value agrees with the effective mass deduced from dHvA studies,\textsuperscript{4} i.e. $m_0 = (3.3 - 3.4)m_e$.

4. Discussion and conclusions

We comment briefly on the unusual form of the phase and amplitude variation seen in Figure 1, which is not typical for CR; it is more common to observe a Lorentzian resonance lineshape, as seen, e.g., in semiconductors. The reason for this is because, in the present case, we are dealing with a highly conducting sample in a resonant cavity and, under these conditions, it is the boundary condition between the sample surface and the surrounding vacuum which determines the cavity response. This type of result has been observed previously in experiments on semimetallic Bismuth involving a resonant microwave bridge technique.\textsuperscript{13}

The fact that exactly the same values for the effective mass are obtained from dHvA and CR measurements seems to rule out the possibility that EEd's play an important role in $(DMe-DCNQI)_2$Cu. This result, coupled with the dHvA results,\textsuperscript{4} which indicate that the effective mass, $m^*$, is the same for deuterated and undeuterated salts, suggests that EEd's play no significant role in the entire $(DMe-DCNQI)_2$Cu family. Therefore, we conclude that the large effective mass is simply due to hybridisation between a narrow Cu 3$d$ band and the $\pi$ band due to the DMe-DCNQI molecules.

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References

12. $\sigma_1 \sim 3 \times 10^4$ $\Omega$cm, R. Kato, private communication.