Proceedings of the International Conference on Synthetic Metals
To be published in Synthetic Metals
Gastein, Austria
July 15-21, 2000

RECEIVED
SEP 12 2000
OSTI

ANL/CHM/CP-102620

Unconventional cyclotron resonance in the organic superconductor β"- $(ET)_2SF_5CH_2CF_2SO_3$

R.S. Edwards [a,1], J.A. Symington [a], E. Rzepniewski [a], A. Ardavan [a], J. Singleton [a],
J.A. Schlueter [b]

[a] *Clarendon Laboratory, Oxford University Department of Physics, Parks Road, Oxford, OX1 3PU, UK*
[b] *Argonne National Laboratory, Argonne, IL 60439, USA*

Abstract

We have measured angle-dependent magnetoresistance oscillations and the interplane millimetre-wave conductivity of the organic superconductor β"-$(ET)_2SF_5CH_2CF_2SO_3$ as a function of the orientation of the applied magnetic field. We observe harmonics of the cyclotron frequency in the real-space velocity of quasiparticles orbiting the Fermi surface (FS). The harmonic amplitudes depend on the field orientation, providing a new way to measure the quasi-two-dimensional FS topology.

Key words: Organic superconductors, AC transport measurements, magnetic measurements

---

Angle dependent magnetoresistance oscillation (AMRO) measurements were made on the organic superconductor β"-$(ET)_2SF_5CH_2CF_2SO_3$ [1] (T_c=5.4 K) at temperatures between 1.34 K and 4.2 K. The sample was rotated around two mutually perpendicular axes; the polar angle θ between **B** and the normal to the sample's **bc** plane and the azimuthal angle ϕ. Figure 1(a) shows the θ dependence of the magnetoresistance in β"-$(ET)_2SF_5CH_2CF_2SO_3$ for a field of 10 T, a temperature of 1.34 K and several values of ϕ. The data show clear AMROs with sharp maxima, suggesting that they are caused by quasi-two-dimensional FS sections. The temperature dependence of the Shubnikov de Haas oscillations at $\theta = 0°$ is shown in figure 1(b). An effective mass m* of $2.0 \pm 0.1\ m_e$ was obtained.

Fitting the data to a non-elliptical FS section [2], as shown in 1(b, inset);

$$\left(\frac{k_x}{a}\right)^n + \left(\frac{k_y}{b}\right)^n = 1 \qquad (1)$$

gives values $n = 1.1$, $a = 0.226 \times 10^{10}$ and $b = 0.039 \times 10^{10}$. Comparison of the predicted FS [1] and figure 1(b, inset) show that the quasi-two-dimensional FS pocket is very different from that predicted by band structure calculations [1].

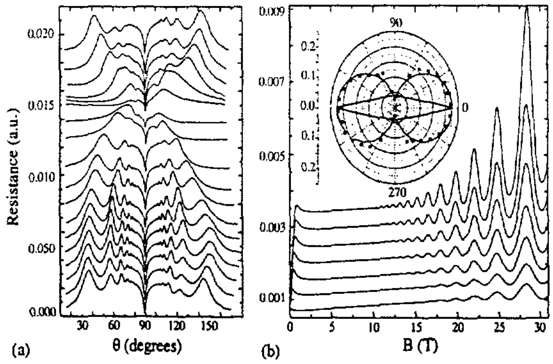

Fig. 1. (a) and (b)
AMRO measurements and fitted FS (inset)

High frequency conductivity measurements were made using millimetre-wave radiation provided by a millimetre-wave vector network analyser (MVNA) [3], and resonant cavity techniques. The sample was placed inside a rectangular cavity resonating in the TE_{102} mode at 71.2 GHz [4]. Our system enables the cavity to be rotated with

---

[1] E-mail: r.edwards1@physics.ox.ac.uk

respect to an external magnetic field, changing θ. The sample can also be rotated inside the cavity to change ϕ. Samples are aligned in the cavity so that the oscillating H field is parallel to the highly conducting planes; induced currents must flow in the interplane (low conductivity) direction, leading to a skin depth greater than the sample size. Since the conductivity anisotropy is large, the millimetre-wave dissipation is dominated by the interplane conductivity.

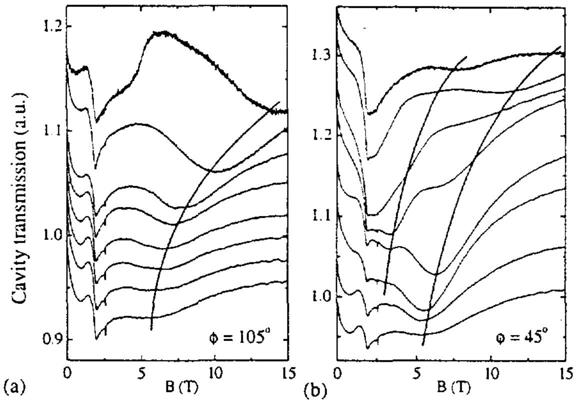

Fig. 2. (a) and (b)
Millimetre-wave measurements for two ϕ angles

Figure 2(a) and (b) shows field sweeps for $\phi = 105°$ and $45°$, between 0 and 15 T at a fixed frequency of 71.72 GHz and a temperature of 1.5 K, with $\theta = 0°$ to $70°$. The sweeps are symmetrical about $\theta = 0°$. The data have been normalized and offset for clarity. The feature visible at 2 T on all sweeps is a background of the experimental set-up.

At intermediate fields several broad resonances can be seen. For $\phi = 105°$ one resonance moves to higher fields with increasing angle θ. The position of this resonance behaves with angle in the manner expected of a cyclotron resonance,

$$\frac{\omega}{B} = A\cos(\theta - \theta_0) \qquad (2)$$

Values of $A/\omega = 0.173 \pm 0.003\,T^{-1}$ and $\theta_0 = 2° \pm 2°$ are found, leading to an effective mass m^*_{CR} of $2.26 \pm 0.03 m_e$.

For all azimuthal orientations studied except $\phi = 105°$, another resonance appears at approximately half the field of the first resonance at higher values of θ. This is the second harmonic of the fundamental resonance. At some azimuthal angles a third harmonic is seen appearing at $\theta \approx 60°$.

We propose a new theory explaining the behaviour of the harmonics as follows [5]. For a general magnetic field orientation, quasiparticles follow orbits around the FS such that the z-component of their real-space velocity, v_z, oscillates. If the angle θ is small, the orbit remains within the first Brillouin zone in the k_z-direction and v_z oscillates at the cyclotron frequency, ω_c. As θ increases, the orbit can extend over several Brillouin zones in the k_z-direction and v_z acquires oscillatory components at harmonics of ω_c. The threshold values of θ at which each harmonic becomes important depend on the ellipticity of the Fermi surface and its azimuthal orientation. Analysis of our millimetre-wave data predicts an ellipticity of 10.5:1, in excellent agreement with AMRO data (9:1) and an orientation that also agrees strongly with that found from AMRO data. The ratio of the **c** to the **a** and **b** axes can also be found.

Comparing the mass determined from the temperature dependence of magnetic oscillations m^*, to the mass as measured in a millimetre-wave experiment m^*_{CR}, we see that $m^*_{\mathrm{CR}} > m^*$ for this material. Thus it appears that the predictions [6] of enhancement of the effective mass derived from magnetic quantum oscillations over that measured in a cyclotron resonance experiment do not hold. Hubbard-model calculations have recently been carried out which contradict the simple theories [7] and which appear to support the above experimental observations. In these calculations it was found that the relationship between these two masses depends strongly upon the characteristics of the material measured (e.g. band filling) and in some cases m^* can be exceeded by m^*_{CR}.

References

[1] U. Geiser, J.A.Schluter, H.H.Wang, A.M.Kini, J.M.Williams, P.P.Sche, H.I. Zakowicz, M.L. VanZile, J.D.Dudek, P.G. Nixon, R.W. Winter, G.L. Gard, J. Ren, M.-H. Whangbo, J. Am. Chem. Soc. 118, 9996 (1996)

[2] M.-S. Nam, this conference.

[3] MVNA manufactured by *ABmm*, 52 Rue Lhomond, 75005 Paris, France.

[4] A. Ardavan, J.M. Schrama, S.J. Blundell, J. Singleton, W. Hayes, M. Kurmoo, P. Day, P. Goy, PRL 81, 713 (1998)

[5] R.S. Edwards et. al, to be published.

[6] K.F. Quader, K.F. Bedell, G.E. Brown, Phys. Rev. B 36 (1987) 156; A.J. Legett, Annals of Physics 46 (1968) 76; W. Kohn, Phys. Rev. 123 (1961) 1242.

[7] K.Kanki and K. Yamada, J. Phys. Soc. Jpn. 66, 1103 (1997)