ELECTRON MAGNETIC RESONANCE FERMI SURFACE IMAGING:
APPLICATIONS TO ORGANIC CONDUCTORS AND Sr$_2$RuO$_4$

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We report detailed angle dependent studies of the metallic state microwave (40 to 200 GHz) magneto-conductivity of single crystal samples of the $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ organic charge density wave conductor, and the perovskite superconductor Sr$_2$RuO$_4$. We observe series’ of resonant absorptions which we attribute to periodic orbit resonances - a phenomenon closely related to cyclotron resonance. By performing measurements on several samples, and in different electromagnetic field configurations, we are able to couple to different orbital modes (+ harmonics), which derive from deformations (warpsings) of the quasi-one and quasi-two-dimensional Fermi surfaces of these compounds. These studies provide vital information concerning interlayer dispersion which, in turn, affects the Fermi surface nesting characteristics which are believed to play a crucial role in the low temperature physics of these exotic materials.

1 Introduction

A detailed knowledge of the Fermi surface (FS) topologies of low-dimensional conductors is an essential starting point for understanding the mechanisms that drive the various electronic instabilities which result in, e.g. the magnetism or superconductivity in these systems. For example, recent theoretical studies have shown that the symmetry of the superconducting state in quasi-2D (Q2D) and quasi-1D (Q1D) systems is extremely sensitive to the nesting characteristics of the FS (see Refs. [1,2]). Although there exists an extensive array of experimental techniques for probing FS topology, few possess the necessary resolution to profile the small (often < 1%) deformations (warpsings) that arise due to weak dispersion along the low conductivity axis (axes) of Q2D (Q1D) systems.
We have recently developed new methods for determining the FS topologies of quasi-low-dimensional systems using a millimeter-wave spectroscopic technique. A novel type of cyclotron resonance (CR) is predicted to occur, which is fundamentally different from the conventional CR observed in normal metals. This technique, which was first considered by Osada et al., essentially corresponds to high frequency ($\omega \sim \omega_c$) Angle-dependent Magneto-resistance Oscillations (AMRO). The periodic $k$-space motion (over the FS) induced by the application of a magnetic field, translates into periodic modulations of the real space quasiparticle velocities as they traverse either Q1D or Q2D warped FSs in a plane perpendicular to the applied field; in particular, the velocity components along the low-conductivity directions undergo dramatic oscillations. GHz measurements then couple resonantly to these real space oscillations, resulting in so-called Periodic Orbit Resonances (POR). Just as in the DC case, different angle dependences are predicted for the Q1D and Q2D cases. However, additional harmonic AMRO/POR series are expected, corresponding to the higher harmonic content of the FS warping. Thus, GHz AMRO offer a novel means for extracting minute, albeit essential, details of the FS topologies of low-dimensional conductors.

2 Experimental

Single crystals of $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ and the perovskite Sr$_2$RuO$_4$, with dimensions ranging from $2\times1\times0.05$ to $0.5\times0.5\times0.05$ mm$^3$, have been used in these investigations. Both materials possess layered structures, and exhibit Q2D conducting behavior. However, as we will show, the POR observed in Sr$_2$RuO$_4$ reflect its Q2D FS while, in the organic compound, a Q1D POR behavior is seen.

Measurements were performed using a cavity perturbation technique covering the frequency range from 18 to 200 GHz; for experimental details see Ref. [6]. A single sample was placed within a cylindrical resonator such that a combination of in-plane and interlayer currents were excited; a detailed discussion of the electrodynamics is published elsewhere. Interlayer currents penetrate deep (100 $\mu$m – 1 mm) into the sample due to the low conductivity in this direction. Dissipation then depends on the ratio of the interlayer and in-plane conductivities, as well as the relative dimensions of the sample. For this experimental geometry, we have shown unambiguously that this dissipation is dominated by the interlayer conductivity for the highly anisotropic organic conductors, whereas the measured dissipation is equally sensitive to the in-plane (conventional CR) and interlayer (POR) conductivities for the slightly less anisotropic Sr$_2$RuO$_4$ system.

A combination of the resistive magnets at the National High Magnetic Field Laboratory (up to 33 T) and a superconducting split-pair (7 T) were used in these investigations. The magnetic field was either swept at constant orientation with respect to the sample, or the orientation of a fixed magnetic field is swept relative to the cavity containing the sample.
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Figure 1(a): Scaled angle dependent dissipation (+ fits) measured in Sr$_2$RuO$_4$ (T = 2 K), showing several POR branches, plus harmonics. b) Angle dependent oscillations in the magneto-conductivity, at T = 2 K, of α-(BEDT-TTF)$_2$KHg(SCN)$_4$; note the field dependence of the dissipation (∝ conductivity) maxima.

3 Results and discussion

Figure 1(a) shows angle dependent POR for Sr$_2$RuO$_4$. Several branches (+ harmonics) are observed, corresponding to the three well known FSs (α, β, and γ), with effective masses of $m_\alpha = 4.3 \ m_o$, $m_\beta = 5.76 \ m_o$, and $m_\gamma = 9.73 \ m_o$. A detailed account of the analysis of this data may be found in Ref. [8], including a determination of the symmetry of the FS warping. The important point to note is the fact that the measured effective masses scale as $1/\cos \theta$, where $\theta$ is the angle between the applied field and the normal to the conducting layers. This confirms the Q2D nature of the FSs of Sr$_2$RuO$_4$, and settles a recent controversy regarding the origin of the resonances observed in the mm-wave magneto-conductivity of this compound.$^8,9$

Figure 1(b) shows the first measurements (a preliminary study) of dissipation versus angle for α-(BEDT-TTF)$_2$KHg(SCN)$_4$. The sharp peaks correspond to conductivity resonances, or PORs. The data show symmetry about $\approx 40^\circ$ and $\approx 130^\circ$, corresponding approximately to the field applied perpendicular and parallel to the highly conducting layers. The first thing to note is the amazing similarity between this data, and published DC AMRO data.$^{10}$ However, most importantly, we observe a systematic shift in the positions (angles) of the conductivity resonances for the two
magnetic field strengths. In the DC case, the AMRO conductivity peaks are observed at field independent angles. The reason for the shifts in our data is due to the fact that there exist two similar time scales in the problem, both of which exceed the scattering time – namely the FS traversal period, which depends on the field strength, and the microwave oscillation period. It has been predicted that the AMRO minima (conductivity maxima) depend on the ratio of $f/B$, where $f$ is the measurement frequency and $B$ the applied field strength.\textsuperscript{4,5} For the DC case, this ratio is always zero, and the angles at which AMRO minima are observed do not depend on the field strength.

Another point to note is the strong dependence of the POR frequency on angle in the vicinity of $40^\circ$ (field $\perp$ layers); a 70\% increase in the magnetic field ($\propto$ POR frequency) causes the conductivity peaks to move from only about $18^\circ$ to $33^\circ$ either side of $40^\circ$ (see dashed lines). For the 2D case, the POR frequency should scale as $1/\cos \theta$, where $\theta$ is measured relative to $40^\circ$ in our set up, i.e. for the 2D case, the observed shift in the peaks would be brought about by only a 13\% increase in the field strength. In fact, although preliminary, our measurements are characteristic of Q1D POR, in agreement with AMRO measurements.\textsuperscript{10} These are the first measurements of their kind, and a work very much in progress. Future measurements of this kind hold the promise to accurately image minute details of the FSs of a wide range of low-dimensional conductors.

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References