Angle-Resolved Mapping of the Fermi Velocity in a Quasi-Two-Dimensional Organic Conductor

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We demonstrate a new method for determining the Fermi velocity in quasi-two-dimensional (Q2D) conductors. Application of a magnetic field parallel to the conducting layers results in periodic open orbit quasiparticle trajectories along the Q2D Fermi surface. Averaging of this motion over the Fermi surface leads to a resonance in the interlayer microwave conductivity. The resonance frequency is simply related to the extremal value of the Fermi velocity perpendicular to the applied field. Thus, angle dependent microwave studies enable a complete mapping of the in-plane Fermi velocity. We illustrate the applicability of this method for the highly 2D organic conductor κ-(BEDT-TTF)2I3.

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Microwave spectroscopy has been utilized as a means of studying the electrodynamic properties of metals for well over half a century, especially resonant absorption in an external dc magnetic field. Quasiparticles in such conductors usually move on closed periodic trajectories in reciprocal (k) space, or cyclotron orbits in real space. When any period associated with this motion matches the period of the external electromagnetic field, so-called cyclotron resonance (CR) occurs if the condition \( \omega_c \tau > 1 \) is satisfied, where \( \omega_c \) is the cyclotron frequency and \( \tau \) is the relaxation time; \( \omega_c \) depends on the magnetic field strength, and on the cyclotron mass (\( m_r \))—a characteristic of the Fermi surface (FS). In conventional metals, large Fermi velocities (\( v_F \sim 10^6 \text{ m/s} \)) complicate matters to the extent that CR is only observed in the anomalous skin effect regime. The theoretical foundations for this and many other electrodynamic properties of metals have been firmly established [1].

In layered conductors, the FS may be either quasi-two-dimensional (Q2D), quasi-one-dimensional (Q1D), or a combination of both. In the Q2D case, the FS is a warped cylinder with its axis perpendicular to the layers (see Fig. 1) while, in the Q1D case, the FS consists of a pair of warped sheets at ±\( k_F \). Because of this reduced dimensionality (and reduced \( v_F \sim 10^3 \text{ m/s} \)), several new effects in the microwave conductivity have been reported, one of which is the observation of multiple periodic orbit resonances (POR) in Q1D systems [3]. In this Letter, we detail a new magnetic resonance phenomenon which enables angle-resolved mapping of the in-plane Fermi velocity for a Q2D conductor. As such, this technique is complimentary to angle-resolved photoelectron spectroscopy (ARPES) [4], i.e., it can provide information concerning the in-plane momentum dependence of the density of states (\( \approx v_F \)) and quasiparticle scattering rate (\( \tau^{-1} \)), as opposed to methods based on the Shubnikov–de Haas (SdH) and de Haas–von Alphen (dHvA) effects, which measure only the momentum averages of these quantities. We illustrate the utility of this method for the \( \kappa \)-(BEDT-TTF)2I3 [5] (BEDT-TTF \( \equiv \) bis-ethylenedithio-tetrathiafulvalene) organic superconductor, which has a relatively simple and well characterized FS (see Fig. 1 and Ref. [2]). However, this technique could equally be applied to more exotic Q2D conductors.

![FIG. 1. (a) An illustration of the quasiparticle trajectories on a warped Q2D FS cylinder for a field oriented perpendicular to the cylinder axis; the warping has been greatly exaggerated for clarity. The resulting trajectories lead to a weak modulation of the quasiparticle velocities parallel to \( k_z \) and, hence, to a resonance in \( \sigma_{zz} \). (b) The thick line shows \( v_F(\phi) \) according to Eq. (1); the right angle triangle illustrates the relationship between \( v_F^\parallel(\phi) \) and \( v_F(\phi) \) [see text and Eq. (4) for explanation]. (c) The Fermi surface of \( \kappa \)-(BEDT-TTF)2I3 according to Ref. [2].](image)
(e.g., Sr₂RuO₄ [6]), provided that they possess relatively long mean free paths. As many ARPES investigations have shown [4], angle-resolved FS spectroscopies have the potential to reveal critical information concerning the various instabilities which give rise to unusual magnetic and superconducting states in low-dimensional correlated electron systems.

Quasiparticles in QD conductors move under an external magnetic field along open orbits. The motion is periodic because of the underlying periodicity of the crystal lattice. When the period of this motion coincides with the period of an appropriately polarized electromagnetic field, resonant microwave absorption occurs, i.e., the ac conductivity attains a maximum. Although a form of closed-orbit CR has been reported for several Q2D organic conductors, its origin is quite different from that in normal metals [7,8]. Electromagnetic fields penetrate easily into the bulk of the sample for current excitation normal to the layers [9], due to the very low interlayer conductivity (the anomalous skin effect is impossible to achieve in the millimeter spectral range, even for the highest conducting direction); the typical interlayer skin depth at 50 GHz is about 50–100 μm, i.e., comparable with the sample dimensions. Indeed, typical sample shapes and conductivity anisotropies result in a situation wherein the interlayer conductivity (σzz) usually dominates the electrodynamic response [9]. Thus, it has been shown both theoretically [7,8] and experimentally that the Q2D closed-orbit resonances are related to the finite warping of the FS, i.e., this effect is analogous to the QD POR, albeit that the periodic motion normal to the layers is related to the underlying cyclotron motion which is predominantly confined to within the layers.

A new effect appears if one aligns the magnetic field within the layers of a Q2D conductor, i.e., perpendicular to the Q2D FS cylinder axis. In this case, the quasiparticle motion is principally open (except for a small fraction of the total electrons—see Fig. 1(a) and [10–12]). This results in periodic motion along open orbits normal to the layers. The period of this motion depends on the magnetic field strength, B, and the velocity component (vzz) perpendicular to the field. Consequently, the period depends strongly on the in-plane wave vector kₓy. However, as we will show, averaging over the FS leads to the result that the extremal perpendicular velocity (vzz) dominates the electrodynamic response, giving rise to a singularity (a form of resonance) in the interlayer conductivity. The resonance occurs when the period of the electromagnetic field matches the periodicity of these extremal quasiparticle trajectories. Thus, one can map out the Fermi velocity by this method. This effect was originally predicted by Peschanskii and Pantoja [13], albeit within the anomalous skin effect regime. In the following, we derive some expressions for the real experimental case, where the skin depth is bigger or comparable to the sample size. Using these calculations, we show that it is also possible to map out the Fermi velocity in this limit.

We consider an energy dispersion of the form

$$E(\mathbf{k}) = \frac{h^2 k_x^2}{2m_x} + \frac{h^2 k_y^2}{2m_y} - 2t_\perp \cos(k \cdot \mathbf{R}),$$

(1)

where ‖ and m₂ are the in-plane diagonal components of the effective mass tensor, R is the real space vector characterizing the interlayer FS warping, and 4tₚ is the interlayer bandwidth. This approach assumes that the interlayer transport is coherent. However, recent theoretical studies have shown that closed-orbit POR occur for both the coherent and weakly incoherent cases [8]. Therefore, it is reasonable to assume that this would also hold for the open orbit resonance, since the physical origin of the two effects is essentially the same. For a calculation of the interlayer ac conductivity (σzz(ω)) we use the Boltzmann transport equation:

$$\sigma_{zz}(\omega) = \frac{e^2}{4\pi} \int d^2k \left[ -\frac{\partial f_0(\mathbf{k})}{\partial E(\mathbf{k})} \right] v_z(\mathbf{k},0) \times \int_{-\infty}^{0} v_z(\mathbf{k},t) e^{-i\omega t} e^{i\tau} dt,$$

(2)

where τ is the quasiparticle relaxation time. First, we assume a T = 0 limit, so that the derivative of the distribution function, f₀(k), may be replaced by δ(E⁻E₀), where E₀ is the Fermi energy. Second, we neglect the part of the Lorentz force which depends on the interlayer velocity, vz. Finally, we neglect the effect of the closed orbits. This assumption is valid provided \(\sqrt{E_F/4t_p} \gg \pi/T_p\), where T_p is the smallest period for the open orbits [10–12]. From available SdH and dHvA data for the title compound, the ratio E_F/4t_p is estimated to be larger than 10⁴, i.e., κ-(BEDT-TTF)_2I₃ is highly two dimensional. As we show later, \(\pi/T_p\) is of order unity; thus, the closed orbits may be neglected. As a result, the interlayer conductivity can be written

$$\sigma_{zz}(\omega) \approx \int \frac{d\omega_c}{\omega_c} \frac{1}{\omega_c} \frac{dS_k}{d\omega_c} \frac{1 - i\omega \tau}{(1 - i\omega \tau)^2 + (\omega_c \tau)^2} d\omega_c,$$

(3)

where dS_k is an element on the FS, \(\omega_c = eBav_{zz}/h\) is the frequency associated with a given quasiparticle trajectory on the FS, and \(\omega_c^{ext}\) is the extremal value of \(\omega_c\) (a is the interlayer spacing, and \(v_{zz}\) is the in-plane velocity perpendicular to the applied magnetic field).

Looking at Eq. (3), one can see that \(\sigma_{zz}(\omega)\) will be dominated by the extremal perpendicular velocity \(v_{zz}^{ext}\) (see Fig. 1(b)), since dS_k/dω_c diverges (i.e., dω_c/dS_k → 0) at these points on the FS. This leads to a resonance condition whenever \(\omega = \omega_c^{ext} = eBav_{zz}^{ext}/\hbar\), provided that \(\omega \tau > 1\). Measurement of \(\omega_c^{ext}\), as a function of the field orientation \(\psi\) within the xy plane, yields a polar plot of \(\omega_c^{ext}(\psi)\). The procedure for mapping \(\psi_f(\phi)\) is then identical to that of reconstructing the FS of a Q2D.
we expect the dielectric polarizability of the sample has the form within the plane of the plate. In this situation, the effect of currents perpendicular (parallel) to the layers of the sample was achieved using a 7 T superconducting magnet. All measurements were carried out at $T_c = 3.5$ K, and at a frequency of 53.9 GHz. According to published resistivity data [2], we estimate a 54 GHz skin depth for currents perpendicular (parallel) to the layers of 53 $\mu$m (4 $\mu$m). Thus, we expect $\sigma_{zz}$ to dominate any losses in the cavity [9], as required in order to detect the new open orbit resonance.

In Fig. 2 we plot the field dependence of the microwave absorption and phase shift (solid curves) for several field orientations ($\phi$) within the highly conducting $bc$ plane of the sample. To understand the shapes of the curves, we treat the sample as a thin conducting plate of thickness $d$, subjected to a microwave ac magnetic field polarized within the plane of the plate. In this situation, the effective polarizability of the sample has the form

$$\alpha(\omega) = \frac{\tanh(kd/2)}{kd/2} - 1, \quad (6)$$

where $k = (-i\omega\mu_0)^{1/2}$. The absorption is then proportional to the imaginary part of $\alpha(\omega)$, while the phase shift is proportional to the real part. While Eq. (6) is clearly only approximate, we find excellent agreement between the predicted and observed behavior; as an illustration, we have included fits to the $\phi = -54^\circ$ data in Fig. 2. The resonances are rather broad (indicated by arrows), due to the fairly small $\omega\tau$ product ( 2). While it is quite difficult to accurately determine the resonance position from the absorption data, it is relatively easy to do so using the phase shift.

In Fig. 3 we plot the experimentally determined $v^\text{ext}_\bot(\psi)$ on a polar diagram. These values were deduced by three different methods: (1) from the maximum in the phase shift (●); (2) from fits to the absorption curves (○); and (3), from fits to the phase shift curves (∗). The dashed line is a fit to the points deduced by method 1, and the solid line is the corresponding Fermi velocity [from Eq. (4)]. The value for $v_{ym}$ is $1.3 \times 10^5$ m/s and $v_{ym}$ is 0.62 ×
10^5 m/s. This anisotropy is in good agreement with the known anisotropy of the small Q2D FS for \( \kappa \)-(BEDT-TTF)_2I_3 [dark shaded region in Fig. 1(c)]. If one assumes a parabolic dispersion [Eq. (1)], it is possible to compare our data with the band parameters determined for the large Q2D \( \beta \) orbit from optical data by Tamura et al. [18]. In particular, we may estimate the effective mass along the \( \epsilon \) direction as \( m^e_\epsilon = h k^e_F / v^\text{ext}_{\text{cm}} \sim 2.5 m_e \), which compares to the value of \( 2.4m_e \) determined from the optical measurements [18].

Based on the known value, \( S_k = 5.5 \times 10^{18} \text{ m}^{-2} \), for the area of the small Q2D section of the FS in \( k \) space, we may estimate the momentum averaged cyclotron mass from the relation \( m^r = h (S_k / S_\epsilon)^{1/2} \), where \( S_\epsilon \) is the area of the FS in velocity space (Fig. 3). This method gives \( m^r = 1.7 m_e \), while the experimental value deduced from the SdH and dHvA effects is \( \sim 1.9 m_e \) [15], where \( m_e \) is the free electron mass; the value deduced from earlier closed-orbit POR measurements is \( 2.2m_e \) [16]. The mass anisotropy \( (m_\epsilon / m_e) \) deduced from the present measurements for the small Q2D FS [dark shaded region in Fig. 1(c)] is 4. From fits to the data, we estimate an isotropic relaxation time of \( \tau \sim 5 \text{ ps} \), or \( \sigma \tau \sim 2 \). Thus, our initial assumption to neglect closed orbits is fully justified. As a point of interest, if the relaxation time \( \tau \) did vary with the azimuthal angle \( \phi \), this variation could be deduced on the basis of the resonance line shapes.

In principle, one should also expect an open orbit resonance effect from the Q1D FS sections [see Fig. 1(c)], as has recently been reported for several other low-dimensional organic conductors [3]. However, no clear evidence for such behavior was found. Indeed, our current and earlier [16] microwave studies clearly show that the Q2D FS dominates the electrodynamic properties of \( \kappa \)-(BEDT-TTF)_2I_3. This likely suggests that the warping of the Q1D FS is weaker than that of the Q2D section.

Finally, we comment on the effect of a possible misalignment of the field rotation axis. Following Peschansky and Kartsovnik [11], we may assume that the conductivity will be dominated by the periodic motion along \( k_z \), rather than by any cyclotron motion in the \( k_xk_y \) plane, provided \( \sin \theta \ll 1/\omega^\text{ext}_c \tau \) (\( \theta \) is the misalignment angle). Clearly, therefore, the accuracy of our sample alignment is enough to justify our procedure for mapping \( v_F \).

In conclusion, we have demonstrated a new type of magneto-electrodynamic resonance in a layered conductor with a Q2D FS. By rotating the applied field within the layers, one can map out the angle dependence of the Fermi velocity. We illustrate the applicability of this method for the highly 2D organic conductor \( \kappa \)-(BEDT-TTF)_2I_3, for which we obtain FS parameters which are consistent with published results.

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