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Periodic orbit resonances in layered metals in tilted magnetic fields

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The frequency dependence of the interlayer conductivity of a layered Fermi liquid in a magnetic field that is tilted away from the normal to the layers is considered. For both quasi-one- and quasi-two-dimensional systems resonances occur when the frequency is a harmonic of the frequency at which the magnetic field causes the electrons to oscillate on the Fermi surface within the layers. The intensity of the different harmonic resonances varies significantly with the direction of the field. The resonances occur for both coherent and weakly incoherent interlayer transport and so their observation does not imply the existence of a three-dimensional Fermi surface. [S0163-1829(99)51240-X]

There is considerable interest in performing frequency-dependent transport measurements on strongly correlated metals in the hope that they will provide information about the metallic state such as a direct determination of the scattering rate. Layered organic metals^{1,2} are ideal for such experiments due to their high purity and a number of experiments have been performed.^{3,4} In this paper, we show that when the magnetic field is tilted away from the normal to the layers that there are well-defined resonances in the interlayer conductivity when the frequency equals a harmonic of the frequency at which the magnetic field causes electrons to traverse the Fermi surface within the layers. This occurs for both quasi-two and quasi-one-dimensional systems. The intensity of the resonances at different harmonics varies significantly with the direction of the field. For example, it is possible to choose the field direction so one will see predominantly only odd or even harmonic resonances. In general, a three-dimensional Fermi surface is not necessary for the observation of the resonances. We also compare our results to previous theoretical work,^{5,6} which has involved more complicated band structures.

We assume a Fermi liquid within the layers with the simplest possible dispersion relation $\epsilon(k_x, k_y)$. For quasi-one-dimensional systems we take

$$\epsilon(k_x, k_y) = \hbar v_F (|k_x| - k_F) - 2t_b \cos(k_y b), \quad (1)$$

where v_F is the Fermi velocity, k_F is the Fermi wave vector, t_b the interchain hopping integral, and b the interchain distance. For the quasi-two-dimensional case, $\epsilon(k_x, k_y) = \hbar^2/2m^*(k_x^2 + k_y^2)$ where m^* is the effective mass. Solution of the semi-classical equations of motion shows that in a magnetic field B normal to the layers electrons move across the Fermi surface within the layers at a periodic orbit frequency ω_0 , which equals $e v_F b B / \hbar$ or $e B / m^*$ for the quasi-one- and quasi-two-dimensional cases, respectively.

We consider a magnetic field tilted at an angle θ away from the normal to the layers. For the quasi-one-dimensional case, we at first only consider the case where the field is confined to the $x-z$ plane, i.e., the plane containing the most and least-conducting directions. This is done for reasons of simplicity; later in the paper, we consider more general field directions for the quasi-one-dimensional case. We have calculated the frequency-dependent interlayer conductivity for two different models for the interlayer transport: coherent and weakly incoherent interlayer transport. A detailed discussion of these two models is given in Ref. 7. The former model is associated with a three-dimensional Fermi surface. The latter occurs when the intralayer scattering rate $1/\tau$ is much larger than the interlayer hopping integral t_c and so one cannot define a wave vector perpendicular to the layers. In that case the Fermi surface is only defined within the layers. In this paper, we present the results of our calculations and discuss their implications. The details of the calcu-

lations are similar to those for the dc conductivity.⁷ The scattering time τ is assumed to be the same at all points on the Fermi surface. We find that the frequency-dependent interlayer conductivity for *both* coherent (except when the field is very close to the layers) and weakly incoherent interlayer transport is, for a frequency ω ,

$$\frac{\sigma_{zz}(\omega)}{\sigma_{zz}^0} = \sum_{\nu=-\infty}^{\infty} \frac{J_{\nu}(\gamma \tan \theta)^2}{1 + (\omega - \nu \omega_0 \cos \theta)^2 \tau^2}, \quad (2)$$

where $J_{\nu}(x)$ is the ν th order Bessel function, ω_0 is the oscillation frequency associated with the magnetic field, and σ_{zz}^0 is the dc conductivity in the absence of a magnetic field. γ is a constant that depends on the geometry of the Fermi surface; it is $2t_b c / \hbar v_F$ and k_{FC} for the quasi-one- and quasi-two-dimensional cases, respectively.

A. AMRO

In the dc limit ($\omega=0$), Eq. (2) reduces to our result for the dc conductivity.^{7,8} At a fixed field, the interlayer resistivity oscillates as a function of the tilt angle; referred to as angular-dependent magnetoresistance oscillations (AMRO).² These oscillations are known as the Yamaji and Danner oscillations for the quasi-two- and quasi-one-dimensional cases, respectively. The interlayer dc resistivity is a local maximum at the angles, $\theta = \theta_{max}^m$ where

$$\gamma \tan \theta_{max}^m = \left(m - \frac{1}{4}\right) \pi \quad m = 1, 2, 3, \dots, \quad (3)$$

and a local minimum at angles, $\theta = \theta_{min}^m$ where

$$\gamma \tan \theta_{min}^m = \left(m + \frac{1}{4}\right) \pi \quad m = 1, 2, 3, \dots. \quad (4)$$

B. Drude peak

In zero field ($\omega_0=0$) or when the field is perpendicular to the layers ($\theta=0$) (2) reduces to the Drude form,

$$\sigma_{zz}(\omega) = \frac{\sigma_{zz}^0}{1 + (\omega\tau)^2}. \quad (5)$$

It is interesting that this result holds for weakly incoherent transport. This means that even when the intralayer scattering rate is so large ($1/\tau \gg t_c$) that the interlayer mean-free path is much less than the layer spacing one will still observe a Drude peak when there is a Fermi liquid within each of the layers.

C. Periodic orbit resonances

The most important property of Eq. (2), and the focus of this paper, is that the frequency-dependent conductivity has resonances at harmonics of the periodic orbit frequency,

$$\omega = n \omega_0 \cos \theta, \quad (6)$$

where n is an integer. The n th resonance has intensity $J_n(\gamma \tan \theta)^2$, which varies significantly with the orientation of the magnetic field.

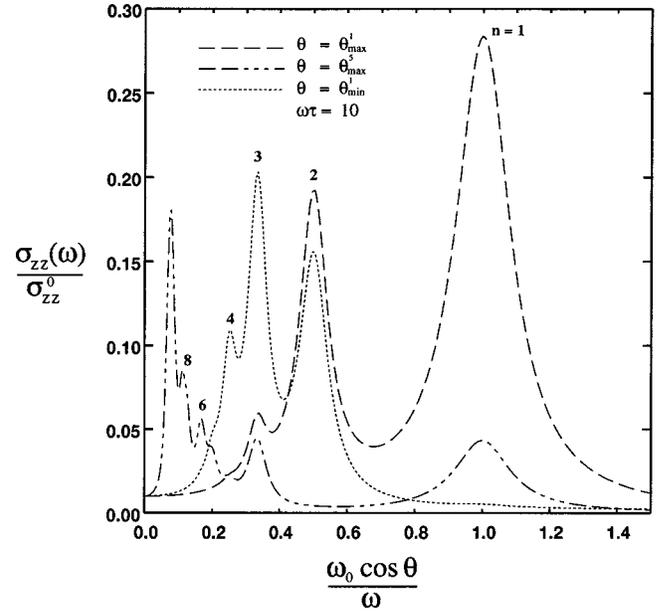


FIG. 1. Magnetic-field dependence of the interlayer conductivity at a fixed frequency ω when the magnetic field is tilted at several different angles θ away from the normal to the layers. The integers given near the peaks denote at which harmonic of the periodic orbit frequency $\omega_0 \cos \theta$ at which the resonance occurs. Note that if the field is tilted at an angle corresponding to the m th AMRO maxima (minima) peaks only occur at the odd (even) harmonics with $n < m$. The peak at the lowest field is the superposition of several harmonics. The ac conductivity $\sigma_{zz}(\omega)$ is normalized to the dc conductivity in zero field, σ_{zz}^0 . The scattering time τ is such that $\omega\tau=10$.

What is the physics behind these resonances? In the coherent case the interlayer electronic group velocity for a trajectory on the Fermi surface starting at $k_z(0)$ is

$$v_z[k_z(0), t] \sim \sin[\gamma \tan \theta \sin(\omega_0 \cos \theta t) + k_z(0)c] \\ \sim \sum_n J_n(\gamma \tan \theta) \sin(n \omega_0 \cos \theta t), \quad (7)$$

which will produce an alternating current at harmonics of the periodic orbit frequency. An ac electric field in the z direction will have resonances with this current. For the weakly incoherent case the overlap of the time-dependent wave functions between neighboring layers has the same form as the right-hand side of Eq. (7).

In a typical experiment the layered metal is placed inside a microwave cavity, which has a fixed resonance frequency ω . The magnetic field is then varied and one looks for resonances in the cavity response, which will generally be dominated by changes in the interlayer resistivity. Hence, in Fig. 1 we plot $\sigma_{zz}(\omega)$ versus ω_0 , which is proportional to the magnetic field. The figure shows a very rich structure in the field dependence in a tilted field, with resonances at many different harmonics of the periodic orbit frequency $\omega_0 \cos \theta$. Furthermore, the intensity of the different resonances is very different, depending on whether the field is at an angle corresponding to an AMRO maximum or minimum. For example, at the first AMRO minimum the first harmonic is suppressed whereas at the fifth AMRO maximum the second

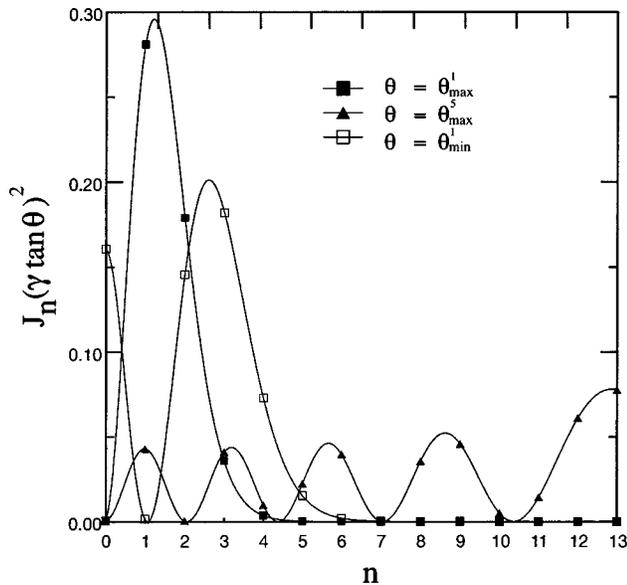


FIG. 2. The intensity of the different harmonic resonances in the interlayer conductivity the field is tilted at angle corresponding to different AMRO maxima and minima.

and fourth harmonics are suppressed. For $z > n$, the asymptotic form $J_n(z) \sim \sqrt{2/\pi z} \cos(z - n\pi/2 - \pi/4)$ holds. Hence, in general, if the field is tilted at the m th AMRO maxima (minima) one will see only the odd (even) harmonics with $n < m$. To illustrate the above discussion in Fig. 2, we plot the intensity of the resonance at the n -th harmonic as a function of n at several different angles. For example, this explains the absence of the $n=2, 4$, and 7 peaks for the fifth AMRO maximum. The peak at the lowest field in Fig. 1 represents a superposition of resonances with the $n=12, 13$, and 14 harmonics; they cannot be resolved because $\omega_0 \tau \sim 1$.

The richness of the response shown in Fig. 1 has important implications for the interpretation of experimental results, many of which contain multiple resonances. As emphasized previously by Hill,⁵ one should be cautious about assigning different resonances to different bands with different effective masses. It is quite possible that if the sample is aligned (perhaps inadvertently) so that the field is tilted away from the normal to the layers that the different features observed actually correspond to harmonics of a single band.

D. Open orbit resonances

In a quasi-two-dimensional metal the presence of periodic orbits is clearly seen in magnetic oscillations such as the de Hass-van Alphen effect. In contrast, the periodic orbits in a quasi-one-dimensional metal do not produce such oscillations, raising the question of what might be clear experimental signatures of the existence of the periodic orbits.⁹ The result (2) shows that frequency-dependent resonances in the interlayer conductivity provide a means to directly establish the periodic motion of the electrons in a quasi-one-dimensional system and to determine the Fermi velocity v_F . It has been proposed that in strongly correlated chains with weak-interchain coupling that coherence is confined to the chains.¹⁰ One would not expect periodic orbit behavior within the layers in such circumstances. Hence, detection of

periodic orbit resonances in a particular material is a possible way to rule out this proposal¹⁰ for that material.

Gorkov and Lebed¹¹ have previously proposed a method of detecting periodic orbit resonances in a quasi-one-dimensional metal using singularities in the surface impedance. However, Hill⁵ pointed out that typical experiments do not satisfy the requirement that the skin depth be less than the cyclotron radius and mean-free path. The method here involves the bulk conductivity and so is not under similar constraints. Hill⁵ considered the frequency-dependent interlayer conductivity for a three-dimensional band structure where the interlayer hopping depends on the wave vector within the layer and the intralayer Fermi surface is elliptical. He found that resonances associated with the second and third harmonics of the cyclotron frequency were possible. The present work shows that some of the effects he found associated with a complicated band structure can also be produced with a simple band structure in a tilted field.

E. Kohn's theorem

Some of the motivation for frequency-dependent measurements in layered metals has been the idea that cyclotron resonance measurements can provide information about the strength of many-body effects that lead to differences between the effective mass deduced from cyclotron resonance and that deduced from magnetic oscillations. Kohn¹² showed that for an *isotropic* three dimensional metal with the electric field *perpendicular* to the magnetic field, a cyclotron resonance experiment will measure a resonance frequency which is independent of the strength of the electron-electron interactions. In contrast, the effective mass determined from the temperature dependence of magnetic oscillations is renormalized by the interactions. Hence, a comparison of the effective mass from the two measurements provides a means to determine the importance of electron-electron interactions in a material. Kohn's argument easily follows through for a two-dimensional electron gas with the magnetic field perpendicular to the plane of the two-dimensional electron gas and the oscillating electric field parallel to the plane. However, the resonances in the interlayer conductivity discussed above are determined by the component of the magnetic field perpendicular to the layers, which is *parallel* to the direction of the oscillating electric field. One finds in that case that Kohn's argument breaks down and so without further work it is not clear whether Kohn's ideas are applicable to the interlayer conductivity of layered metals. In particular, it is quite possible that the resonances discussed here include all the effects of the interactions.

F. Typical parameter values

Typical microwave cavities operate at $\omega = 30\text{--}150$ GHz and to observe the resonances discussed here we should have at least $\omega\tau > 3$. This means the sample must be of sufficient purity that the scattering time $\tau > 2 \times 10^{-11}$ s. The values for quasi-two-dimensional (BEDT-TTF) materials deduced from the Dingle temperature (~ 1 K) for magnetic oscillations are typically about 10^{-11} s. The Dingle temperature tends to underestimate the true value and so with the best samples it should be possible to detect the effects discussed here. For the quasi-one-dimensional $(TMTSF)_2\text{ClO}_4$

Danner, Kang and Chaikin¹³ estimated $\tau=4 \times 10^{-12}$ sec from comparing the angular-dependent interlayer magnetoresistance with semiclassical calculations. If $\omega=100$ GHz, this would give $\omega\tau \sim 0.4$ and so one would need a better sample or to work in the infra-red to see a clear resonance. Furthermore, to detect the first harmonic the field needs to be tilted near the first AMRO maximum, $\theta_{max}^1=84^\circ$. If $\omega=100$ GHz, the first harmonic will occur at a field $B=\hbar\omega/(e v_F b \cos \theta_{max})=2$ T.

G. Quasi-one-dimensional systems with the field in a general direction

For the field $\vec{B}=(\sin \theta \cos \alpha, \sin \theta \sin \alpha, \cos \theta)$, the interlayer conductivity is

$$\frac{\sigma_{zz}(\omega)}{\sigma_{zz}^0} = \sum_{\nu=-\infty}^{\infty} \frac{J_{\nu}(\gamma \tan \theta \cos \alpha)^2}{1 + [\omega - \omega_0(\nu \cos \theta - c \sin \theta \sin \alpha/b)]^2 \tau^2}, \quad (8)$$

which reduces to Eq. (2) when $\alpha=0$.¹⁴ This can be used to interpret recent observations⁴ of frequency-dependent resonances in the low-temperature phase of α -(BEDT-TTF)₂KHg(SCN)₄. In order to make a clearer connection with Ref. 4 we change to angular coordinates similar to that used there, where $\vec{B}=(\sin \phi, \cos \phi \sin \psi, \cos \phi \cos \psi)$. The n th resonance will occur with intensity $J_n(\gamma \tan \phi / \cos \psi)^2$ when $\omega/\omega_0 = n \cos \phi \cos \psi - (c/b) \cos \phi \sin \psi$ which can be rewritten as $\omega \cos \psi_n / \omega_0 \cos \phi = n \sin(\psi + \psi_n)$ where $\tan \psi_n = c/nb$ and $\omega_0 \cos \phi$ is proportional to B_{\parallel} , the component of the field parallel to the $y-z$ plane. This is in a similar form to Eq. (6) in Ref. 4. If $b/c=1.3$ then $\psi_1=38^\circ$ or 142° and $\psi_2=21^\circ$. These values are in reasonable agreement with the values deduced in Ref. 4 from the two resonances observed in the experiment.

An alternative interpretation of the origin of the resonances was given in Ref. 4 in terms of a quasi-one-dimensional band structure,¹⁵ where the interlayer transport includes hopping beyond next-nearest neighbors,

$$\epsilon(k_x, k_y, k_z) = \hbar v_F (|k_x| - k_F) - \sum_{m,n} t_{mn} \cos(mk_y b + nk_z c) \quad (9)$$

and each sum is over all the integers. This can be compared to our dispersion (1), which corresponds to Eq. (9) with the only nonzero elements being $t_{10}=t_b$ and $t_{01}=t_c$, and we do not necessarily assume coherent interlayer transport. Blundell, Ardavan, and Singleton have shown⁶ that for a magnetic field in the $y-z$ plane the frequency-dependent interlayer conductivity is

$$\frac{\sigma_{zz}(\omega)}{\sigma_{zz}^0} = \sum_{m,n} \frac{(nt_{mn}/t_c)^2}{1 + [\omega - \omega_0(n \cos \theta - mc \sin \theta/b)]^2 \tau^2}, \quad (10)$$

which has resonances when $\omega = \omega_0(n \cos \theta - mc \sin \theta/b)$ where m and n are integers. The intensity of these resonances depends on the hopping matrix elements, t_{mn} . Whereas in our treatment these features arise from a component of the field in the x direction, in Ref. 4 they are attributed to the presence of warping in the Fermi surface associated with t_{11} and t_{12} .¹⁶ Also, since Eq. (8) holds for both coherent and weakly incoherent interlayer transport even the existence of a three-dimensional Fermi surface is not necessary for the observation of frequency-dependent resonances. Provided there is a Fermi surface within the layers and the intralayer momentum is conserved in transport between adjacent layers then the resonances are observable.

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¹T. Ishiguro, K. Yamaji, and G. Saito, *Organic Superconductors*, 2nd ed. (Springer, Berlin, 1998).

²J. Wosnitzer, *Fermi Surfaces of Low Dimensional Organic Metals and Superconductors* (Springer, Berlin, 1996).

³J. Singleton *et al.*, Phys. Rev. Lett. **68**, 2500 (1992); S. Hill *et al.*, Synth. Met. **55-57**, 2566 (1993); **70**, 821 (1995); A. Polisski *et al.*, J. Phys.: Condens. Matter **8**, L195 (1996); S.V. Demishev *et al.*, Phys. Rev. B **53**, 12 794 (1996); A. Ardavan *et al.*, Synth. Met. **85**, 1501 (1997); H. Ohta *et al.*, *ibid.* **86**, 2011 (1997); **86**, 1913 (1997); K. Akioka *et al.*, *ibid.* **86**, 2051 (1997); A. Polisski, J. Singleton, and N.D. Kushch, *ibid.* **86**, 2197 (1997); S. Hill *et al.*, Physica B **246-247**, 110 (1998); K. Akioka *et al.*, *ibid.* **246-247**, 311 (1998); S. Hill *et al.*, cond-mat/9905147 (unpublished).

⁴A. Ardavan *et al.*, Phys. Rev. Lett. **81**, 713 (1998).

⁵S. Hill, Phys. Rev. B **55**, 4931 (1997).

⁶S.J. Blundell, A. Ardavan, and J. Singleton, Phys. Rev. B **55**, R6129 (1997).

⁷P. Moses and R. H. McKenzie, Phys. Rev. B **60**, 7998 (1999).

⁸R.H. McKenzie and P. Moses, Phys. Rev. Lett. **81**, 4492 (1998).

⁹The rapid oscillations seen in the *TMTSF*₂*X* materials [J.S. Brooks *et al.*, Phys. Rev. B **59**, 2604 (1999), and references

therein] are not due to the periodic orbits discussed here.

¹⁰S.P. Strong, D.G. Clarke, and P.W. Anderson, Phys. Rev. Lett. **73**, 1007 (1994); D.G. Clarke and S.P. Strong, Adv. Phys. **46**, 545 (1997); D.G. Clarke, S.P. Strong, P.M. Chaikin, and E.I. Chashechkina, Science **279**, 2071 (1998).

¹¹L.P. Gor'kov and A.G. Lebed', Phys. Rev. Lett. **71**, 3874 (1993).

¹²W. Kohn, Phys. Rev. **123**, 1242 (1961).

¹³G.M. Danner, W. Kang, and P.M. Chaikin, Phys. Rev. Lett. **72**, 3714 (1994).

¹⁴As an aside, we note that if we take the dc limit of Eq. (8) then the interlayer resistivity will have minima at the magic angles given by $\tan \theta \sin \alpha = b\nu/c$, where ν is an integer. The intensity of these features depends on the component of the field parallel to the chains. Such minima have been observed in (*TMTSF*)₂PF₆ at 8.3 kbar [I.J. Lee and M.J. Naughton, Phys. Rev. B **57**, 7423 (1998)].

¹⁵T. Osada, S. Kagoshima, and N. Miura, Phys. Rev. B **46**, 1812 (1992); S.J. Blundell and J. Singleton, *ibid.* **53**, 5609 (1996).

¹⁶Generally on the grounds of quantum chemistry one would expect $t_{12} \ll t_{11} \ll t_{10} = t_c$ and so it is appealing that it is unnecessary to assign them comparable values in order to explain experimental results.