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Nonlinear de Haas–van Alphen Oscillations in Charge-Density-Wave Systems

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In charge- or spin-density-wave systems, interaction between Landau levels and many-body effects produces nonlinear de Haas–van Alphen oscillations. This yields, *inter alia*, frequencies which are difference frequencies between ordinary oscillations with cyclotron mass equal to the sum of the ordinary masses. The new oscillations may have an amplitude as large as 10% of the ordinary ones, but they are in a new region of the spectrum where they can easily be detected. The effect is larger for weakly stable desntiy waves.

There have been recent measurements of the de Haas–van Alphen[1,2] (dHvA) and the de Haas–Shubnikov[3] effects in transition-metal chalcogenide layer compounds. Since it is known that these substances have charge-density-wave (CDW) ground states,[4] it is of great interest to explore the interrelationships between the quantized Landau levels and the CDW's.

The usual theory[5,6] of the dHvA effect shows that, for a given band structure, each extremal closed cross section of the Fermi surface gives rise to a well-defined frequency in the oscillatory free energy and hence in the magnetization and magnetic susceptibility. The study of the dHvA spectrum of a given metal serves thus as an invaluable tool in determining its Fermi surface.

The classical theory of the dHvA effect[5,6] predicts that each frequency appears together with its higher harmonics, but, for noninteracting electrons, there is no mixing of frequencies. For interacting electrons, Shoenberg[7] and Pippard[8,9] have shown that, since the electrons experience the total magnetic induction $\vec{B}$ rather than the applied field $\vec{H}$, a nonlinear equation for the magnetization appears, with its attendant frequency mixing. This magnetic interaction effect has been observed in various metals.[9-11]

The presence of either CDW's or spin-density waves[12-16] gives rise to another mechanism for the nonlinear coupling of dHvA frequencies. In the presence of stable density waves, the electronic spectrum, i.e., the energy–k-vector relationship for quasiparticles, is determined self-consistently by the many-body collective properties of the metal. These properties manifest themselves through an energy-gap parameter Δ which satisfies a nonlinear integral equation. In the presence of a magnetic field $\vec{H}$ the kernel of the integral equation is modified. Therefore, through Δ and the one-electron energy spectrum, the dHvA effect is also modified with the appearance of a new, nonlinear behavior. In particular, it is possible and likely that in CDW ground-state systems, observed small frequencies are in fact differences between ordinary dHvA frequencies.[2] If interpreted as normal dHvA signals, these difference frequencies would correspond to small cross-sectional areas of the Fermi surface which do not exist in the actual case.

In the absence of a magnetic field, the trial free energy of a CDW system, $F_{\mathrm{CDW}}(\Delta)$, is minimized by the equilibrium value Δ_C of the energy gap (order) parameter so that

$$F_C(\Delta_C) = F_{\mathrm{CDW}}(\Delta_C) - F_N < 0, \tag{1}$$

where F_N is the free energy of the normal state. The trial function $F_C(\Delta)$ satisfies the stability conditions

$$\left(\frac{dF_C}{d\Delta}\right)_{\Delta=\Delta_C} = 0; \quad \left(\frac{d^2F_C}{d\Delta^2}\right)_{\Delta=\Delta_C} \equiv \alpha\rho > 0. \tag{2}$$

In (2) the quantity ρ is a density of electronic states at the Fermi level defined below and the equation defines the dimensionless positive number α.

In the presence of a magnetic field, and neglecting both Landau diamagnetism and electron spins, we have

$$F(\Delta, H) = F_C(\Delta) + F_{\mathrm{Osc}}(\Delta, H), \tag{3}$$

where the last term is responsible for the dHvA oscillations. If we now assume cylindrical Fermi surfaces (in the spirit of the two-dimensional properties of the layer structures[3,17]) we obtain

$$F_{\mathrm{Osc}}(\Delta,H)=\rho\sum_i\sum_{\nu=1}(-1)^\nu\hbar\omega_c k_B T\nu^{-1}\frac{\cos[(\hbar c\nu/|e|H)A_i+\theta_{i\nu}]}{\sinh(2\pi^2 k_B T m_i/\hbar\omega_c)}, \tag{4}$$

where i indicates extremal (closed) cross-sectional areas $A_i(\Delta)$ of the Fermi surface with effective cyclotron mass $m_i(\Delta)$ measured in units of the free-electron mass m;

$$\omega_c \equiv |e|H/mc,$$

ν indicates the harmonic order, $\theta_{i\nu}$ is an arbitrary phase, and

$$\rho\equiv 4\pi\Omega\Delta k_z m/\hbar^2, \tag{5}$$

(Ω = volume of the crystal; Δk_z = height of the Brillouin zone perpendicular to the layers).

The equilibrium value of $\Delta(H)$ is obtained by minimizing (3) with respect to Δ. This yields

$$\Delta(H)=\Delta_C+\delta\Delta(H),$$

where, to first order,

$$\delta\Delta=-(dF_{\mathrm{Osc}}/d\Delta)_{\Delta=\Delta_C}[(d^2F_C/d\Delta^2)_{\Delta=\Delta_C}]^{-1}, \tag{6}$$

and

$$F=F_C(\Delta_C)+F_{\mathrm{Osc}}(\Delta_C,H)-\tfrac{1}{2}[(dF_{\mathrm{Osc}}/d\Delta)_{\Delta=\Delta_C}]^2[(d^2F_C/d\Delta^2)_{\Delta=\Delta_C}]^{-1}. \tag{7}$$

When (4) and (2) are introduced into (7) we obtain oscillatory terms which vary periodically with inverse magnetic field with frequencies which are characteristic of the areas $(n_iA_i\pm n_jA_j)$. In all cases the areas are for $\Delta=\Delta_C$, and n_i and n_j are integers. If we call amplitudes $f(n_iA_i\pm n_jA_j)$ the factors in front of the cosine functions in F, the most important amplitudes are given by

$$f(A_1)=\rho\frac{\hbar\omega_c k_B T}{\sinh(2\pi^2k_BTm_1/\hbar\omega_c)}; \tag{8}$$

$$f(2A_1)=\rho\frac{\hbar\omega_c k_B T}{2\sinh(4\pi^2k_BTm_1/\hbar\omega_c)}\left(\left|1+\cos\chi_{2A_1}\frac{k_BT}{\alpha\hbar\omega_c}\left\{\left[\frac{\hbar^2}{m}\left(\frac{dA_1}{d\Delta}\right)_{\Delta=\Delta_C}\right]^2+\left[2\pi^2k_BT\left(\frac{dm_1}{d\Delta}\right)_{\Delta=\Delta_C}\right]^2\right\}\right|\right); \tag{9}$$

$$f(A_1\pm A_2)=\rho\frac{(k_BT)^2}{\alpha\sinh[(2\pi^2k_BT/\hbar\omega_c)(m_1+m_2)]}\left\{\left[\frac{\hbar^2}{m}\left(\frac{dA_1}{d\Delta}\right)_{\Delta=\Delta_C}\right]^2+\left[2\pi^2k_BT\left(\frac{dm_1}{d\Delta}\right)_{\Delta=\Delta_C}\right]^2\right\}^{1/2}$$
$$\times\left\{\left[\frac{\hbar^2}{m}\left(\frac{dA_2}{d\Delta}\right)_{\Delta=\Delta_C}\right]^2+\left[2\pi^2k_BT\left(\frac{dm_2}{d\Delta}\right)_{\Delta=\Delta_C}\right]^2\right\}^{1/2}. \tag{10}$$

In all the above equations we have kept only lowest order contributions in $\delta\Delta$ and we have assumed that $2\pi^2k_BTm_i/\hbar\omega_c>1$. In (9) χ_{2A_1} is a phase which depends on the phases of θ_i of (4).

Although the CDW's modify the harmonic content of the ordinary dHvA oscillations, as shown in (9), the main effect is the appearance of the new frequencies as given by (10). In particular $f(A_1-A_2)$ may correspond to a small frequency even though both A_1 and A_2 are normal (large) areas.

In order to illustrate the results above we have calculated the amplitudes for a particular example. We have taken a two-dimensional dispersion relation $E(\vec{k})$ of the form

$$E(\vec{k})=[\hbar^2v_F^2(|\vec{k}|-k_0)^2+\Delta^2]^{1/2}-E_0, \tag{11}$$

which corresponds to a BCS-type model discussed by Fedders and Martin[13,18] and Rice.[14,18] The relation (11) yields at the Fermi level $E=0$, two concentric circles (cylinders) of radii

$$k_\pm=k_0\pm[E_0^2-\Delta^2]^{1/2}/\hbar v_F. \tag{12}$$

Of these surfaces the inner one corresponds to holes and the outer one to electronlike quasiparticles. In Fig. 1 we show results for $f(A_1)$, $f(2A_1)$,

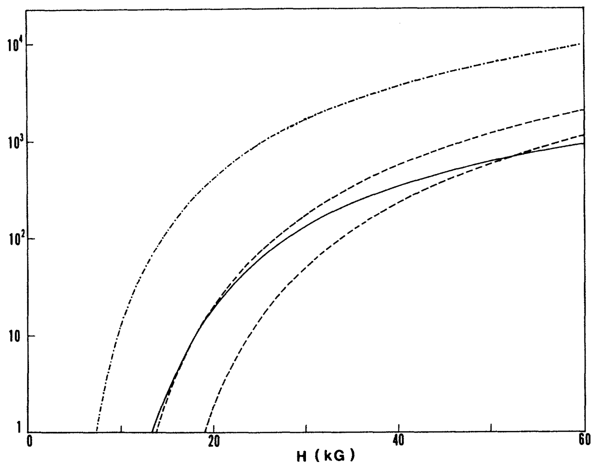

FIG. 1. The amplitude of the oscillatory free energy for the example given in the text. The abscissas are in kilogauss and the ordinates in arbitrary units. The dash-dotted curve corresponds to the fundamental areas A_1 and A_2. The dashed curves correspond to the second harmonics $2A_1$ and $2A_2$; the upper curve is for $\chi_{2A}=0$ in formula (9) (constructive interference) and the lower curve for $\chi_{2A}=\pi$ (destructive interference). The full line corresponds to both A_1+A_2 and A_1-A_2.

and $f(A_1 \pm A_2)$ for the case

$$\alpha=0.2, \quad T=4°\text{K}, \quad \Delta=0.02 \text{ eV},$$

$$E_0=0.04 \text{ eV}, \quad v_F=10\hbar k_0, \quad k_0=0.172 \text{ Å}^{-1},$$

which are realistic parameters for the layer compounds. In this model $(A_1-A_2)/A_1=0.06$, i.e., the frequency of the difference oscillation is only 6% of the A_1- or A_2-related frequencies.

It is interesting to note that the A_1-A_2 frequency of Fig. 1, which is very small, has a temperature dependence characteristic of a "heavy" mass (m_1+m_2). For the example given $m_1=m_2\cong 0.115$ and, for a typical field of 50 kG, $f(A_1-A_2)$ is a factor of 10 smaller than either $f(A_1)$ or $f(A_2)$. However, since the oscillations are in a completely different region of the spectrum, they should be easily observable.

The numerical example reported here is for admittedly rather small but reasonable "ordinary" cyclotron masses. For masses one order of magnitude larger (~ 1.0 m), the difference frequency at $T=4°$K and $H=50$ kG would have an amplitude only 10^{-5} that of the fundamental frequencies.

One point is worth remarking. The electronic spectrum at the Fermi level is particularly sensitive to the appearance of CDW or spin-density waves. In fact these density waves are stable precisely because they introduce gaps in the energy spectrum at the Fermi level. The energy and wave function of the electrons near E_F, and therefore the area and structure of the electron orbits as seen by the dHvA effect, are thus more strongly affected by the density waves than by other electronic parameters.

As a concluding remark, it is important to notice that the amplitude of the nonlinear effect, as given by (10), is inversely proportional to the parameter α defined by (2). Since α is a measure of the stability of the CDW, the new dHvA oscillations should be more easily observable for small α's, i.e., for less stable CDW systems.

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