Influence of Landau Level Broadening on the de Haas-van Alphen Effect

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Dingle's treatment of the influence of Landau level broadening on the de Haas-van Alphen effect is extended by the method of Lifshitz and Kosevich to include a Fermi surface of arbitrary shape. It is shown that a momentum- or energy-dependent linewidth can influence the period, phase, and amplitude of the magnetization oscillations, although crude estimates indicate that in many cases the effects would be quite small.

INTRODUCTION

HE influence of Landau level broadening on the de Haas-van Alphen (DHVA) effect has been considered by Dingle¹ for free electrons, where the effect of a Lorentzian line shape parameterized by a "collision time" τ takes the form of the factor $\exp(-m^*c/\tau eH)$ in the oscillatory magnetization. We shall show by the method of Lifshitz and Kosevich² (hereafter referred to as LK) that this result is applicable for an arbitrary Fermi surface, provided the effective mass m^* is defined as equal to $\partial S/2\pi \partial E$, where S is the appropriate extremal cross-sectional area inclosed by the Fermi surface in momentum space. We shall also show that kinematical or dynamical effects which cause the parameter τ to be momentum or energy dependent may influence the period and phase as well as the amplitude of the oscillations. In particular, by allowing $1/\tau$ to contain a term proportional to the momentum in the direction of the applied magnetic field (the z axis), the relative importance of electrons with different p_z is shifted so that the effective area determining the frequency of the oscillatory magnetization is no longer the extremum. However, as indicated by crude estimates, such effects may generally be quite small.

With the methods of quantum field theory, Bychkov³ has treated the effect of elastic scattering of free electrons by impurities. He has concluded that the Dingle factor is applicable if $\tau = 4\tau'/\pi$, where τ' is the mean free-collision time in the absence of an applied field, provided that $1/\nu_c \tau' \gg h\nu_c/\zeta$, where ν_c is the cyclotron frequency and ζ the Fermi energy. For $1/\nu_c \tau' \leq h\nu_c/\zeta$, the effect of impurities can also reduce to the Dingle factor if other conditions are met.³

PROOF FOR ARBITRARY FERMI SURFACE

Following Dingle,¹ let us suppose that the broadening of the nth Landau level can be described by a Lorentzian distribution function given by

$$dN = \left(\frac{\hbar}{\pi\tau}\right) \frac{dE}{(E - E_n)^2 + (\hbar/\tau)^2}.$$
 (1)

This has been normalized on the interval $(-\infty,\infty)$, which can also be taken as $(E_n - E_0, \infty)$ with negligible error, provided that the ground-state energy E_0 and energy-level spacing are much larger than \hbar/τ . Such broadening can be caused by the finite lifetime of the individual states or the lowering of the free-particle symmetry by the crystal field.^{4,5} Consequently, it could be energy-dependent, although the exact form it would take is as yet uncertain. We shall consider Eq. (1) as a phenomenological basis for our treatment.

A system of noninteracting fermions has a free energy which from the standpoint of the grand canonical ensemble can be written as a sum over quantum states λ available to each particle⁶

$$F = -kT \sum_{\lambda} \ln\{1 + \exp[\zeta - E(\lambda)/kT]\} - \zeta N, \quad (2)$$

where ζ is the Fermi energy. Diamagnetic properties of the system are predicted by retaining the summation over the E_n levels, each with a degeneracy L^2eH/hc (we assume normalization for a cube of side L; each level must include an integration over the states lying below and above in energy as given by Eq. (1). The sum over p_z can be approximated by an integral with the usual degeneracy factor L/h. For the present we shall ignore the spin contribution, which has been considered by Cohen and Blount⁷ and references given by them. Then

⁷ M. H. Cohen and E. I. Blount, Phil. Mag. 5, 115 (1960).

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¹ R. B. Dingle, Proc. Roy. Soc. (London) A211, 500, 517 (1952).
² I. M. Lifshitz and A. M. Kosevich, Zh. Eksperim. i Teor. Fiz.

^{29, 730 (1955) [}English transl.: Soviet Phys.-JETP 2, 636 (1956)].

Yu. A. Bychkov, Zh. Eksperim. i Teor. Fiz. 39, 1401 (1960) [English transl.: Soviet Phys.—JETP 12, 977 (1961)].

⁴ P. G. Harper, Proc. Phys. Soc. (London) A68, 874, 879 (1955); A. D. Brailsford, ibid. A70, 275 (1957).

⁵ J. Zak (to be published).

⁶L. Landau and I. M. Lifshitz, Statistical Physics (Pergamon Press, Ltd., London, 1958), p. 152.

Eq. (2) is explicitly written as

$$F = \frac{-kTVeH}{\hbar^2 c} \left(\frac{\hbar}{\pi\tau}\right) \int_{-E_0}^{\infty} d\xi \sum_{n=0}^{\infty} \int_{-\infty}^{\infty} dp_z$$
$$\times \ln\left\{1 + \exp\left[\frac{\zeta - E_n(p_z) - \xi}{kT}\right]\right\} \left[\xi^2 + (\hbar/\tau)^2\right]^{-1} - \zeta N. \quad (3)$$

The lower limit on the ξ integration is a finite number, so that all integrals converge. We arbitrarily choose it to insure that all energies included in the "sum" are positive; later we shall replace it by $-\infty$, since $E_0 \gg \hbar/\tau$. Application of the Poisson formula to the sum over "n" [cf. LK Eq. (2.3)] permits us to write for the oscillatory part of F:

$$(F)_{\text{osc}} = \frac{-2kTVeH}{\hbar^2 c} \left(\frac{\hbar}{\pi\tau}\right) \operatorname{Re} \int_{-E_0}^{\infty} d\xi \sum_{j=1}^{\infty} \int_{-1/2}^{\infty} dn \int_{-\infty}^{\infty} dp_z \\ \times \ln\left\{1 + \exp\left[\frac{\zeta - E_n(p_z) - \xi}{kT}\right]\right\} \\ \times \exp(i2\pi jn)[\xi^2 + (\hbar/\tau)^2]^{-1}.$$
(4)

The integral over "n" can be changed to one over E by using the Bohr-Sommerfeld quantum condition for motion in the plane perpendicular to H [LK Eq. (1.3)]:

$$\oint P_y dQ_y = (n+\gamma)h, \qquad (5)$$

which reduces to the quantum condition for the area contained within the projection of each particle's trajectory on the transverse plane in momentum space,

$$S(E, p_z) = (n + \gamma) heH/c.$$
(6)

Equation (4) then becomes

$$(F)_{\text{osc}} = \left\{ \frac{-2kTV}{\hbar^3} \left(\frac{\hbar}{\pi \tau} \right) \operatorname{Re} \int_{-E_0}^{\infty} d\xi \sum_{j=1}^{\infty} \int_{0}^{\infty} dE \int_{S>0} dp_z \right. \\ \left. \times \frac{\partial S}{\partial E}(E, p_z) \ln \left[1 + \exp \left(\frac{\zeta - E - \xi}{kT} \right) \right] \right. \\ \left. \times \frac{\exp \left[2\pi i j (cS/heH - \gamma) \right]}{\xi^2 + (\hbar/\tau)^2} \right].$$
(7)

By making the further change of variable $E'=E+\xi$, (F)_{ose} can be seen to have the same form as Eq. (7), but with the E' integration taken over the interval (ξ, ∞) . Since the maximum contribution of the integrand comes from the region $E\approx\xi\gg\hbar/\tau$ and $\xi\leq\hbar/\tau$, we approximate the lower limit of the E' integration by zero, leaving

$$(F)_{\rm osc} = \left\{ \frac{-2kTV}{h^3} \left(\frac{\hbar}{\pi\tau}\right) \operatorname{Re} \int_{-E_0}^{\infty} d\xi \sum_{j=1}^{\infty} \int_{0}^{\infty} dE' \int_{S>0} dp_z \right. \\ \left. \times \frac{\partial S}{\partial E} (E' - \xi, p_z) \ln \left[1 + \exp\left(\frac{\xi - E'}{kT}\right) \right] \right. \\ \left. \times \frac{\exp\{2\pi i j \left[cS(E' - \xi, p_z) / heH - \gamma \right] \right\}}{\xi^2 + (\hbar/\tau)^2} \right\}.$$
(8)

Exchanging orders of integration and summation and integrating by parts, we find

$$(F)_{\text{osc}} = \left\{ \frac{-2V}{\hbar^3} \left(\frac{\hbar}{\pi \tau} \right) \operatorname{Re} \sum_{i=1}^{\infty} \int_0^{\infty} dE' f \left(\frac{E' - \zeta}{kT} \right) \right. \\ \left. \times \int_0^{E'} dE'' \int_{S>0} dp_z \int_{-E_0}^{\infty} d\xi \frac{\partial S}{\partial E''} (E'' - \xi, p_z) \right. \\ \left. \times \frac{\exp[icjS(E'' - \xi, p_z)/\hbar eH - i2\pi j\gamma]}{\xi^2 + (\hbar/\tau)^2} \right\}, \quad (9)$$

where $f(x) = (1+e^x)^{-1}$ is the Fermi function. The integrals can be approximated by noting that the primary contribution of interest from the integrand is in the region $E'' \approx E'$ and $\xi \approx 0$ where $(\partial S/\partial p_z)_{p_m} = 0$. This latter condition defines " p_m ." Then the area in the phase factor can be expanded as

$$S(E''-\xi, p_z) \approx S(E', p_m) + \frac{\partial S}{\partial E}(E', p_m) [E'' \quad \xi - E'] + \frac{1}{2} \frac{\partial^2 S}{\partial p_z^2}(E', p_m) [p_z - p_m]^2. \quad (10)$$

With the assumptions that the ξ dependence of τ and $\partial S/\partial E''$ can be neglected and that $E_0 \gg \hbar/\tau$ so that the lower limit of the ξ integral is effectively $-\infty$, the ξ integration can be readily performed:

$$\begin{pmatrix} \frac{\hbar}{\pi\tau} \end{pmatrix} \int_{-\infty}^{\infty} d\xi \exp\left\{-\frac{icj}{\hbar eH} \frac{\partial S}{\partial E}(E', p_m)\xi\right\} [\xi^2 + (\hbar/\tau)^2]^{-1}$$
$$= \exp\left\{-\frac{cj}{\tau eH} \frac{\partial S}{\partial E}(E', p_m)\right\} = \exp\{-j/\nu_c^*\tau\}, \quad (11)$$

where $\nu_c^* = eH/m^*c$ and $m^* = \partial S/2\pi \partial E$. If τ is independent of E' and p_z , this factor can be removed from the integral and be recognized as the factor first derived by Dingle.¹ It also can conveniently be written as $\exp(-2\pi^2 jkT_D/\beta^*H)$, where T_D is defined to be the "Dingle temperature"—previously called the "x factor" —and β^* is $e\hbar/m^*c$. The remaining integrals are performed as in the treatment of LK, and with the insertion

of the $\cos(j\pi gm^*/2m)$ from the spin dependence as defined by Cohen and Blout,⁷ we obtain as our end result

$$(F)_{\rm osc} = 2VkT \left(\frac{eH}{hc}\right)^{3/2} \left|\frac{\partial^2 S}{\partial p_z^2}\right|_{\varsigma, p_m}^{-1/2} \times \sum_{j=1}^{\infty} \frac{\exp(-2\pi^2 jkT_D/\beta^*H)}{j^{3/2}\sinh(2\pi^2 jkT/\beta^*H)} \times \cos\left[\frac{jc}{\hbar eH}S(\varsigma, p_m) - 2\pi j\gamma \mp \frac{\pi}{4}\right] \times \cos\left[\frac{jc}{\hbar eH}S(\varsigma, p_m) - 2\pi j\gamma \mp \frac{\pi}{4}\right] \times \cos\left(j\pi gm^*/2m\right).$$
(12)

The upper sign is used when $S(\zeta, p_m)$ is a maximum, the lower for a minimum. As usual, the magnetization is given by $M = -(\partial F/\partial H)_{T,V}$.

ENERGY AND MOMENTUM-DEPENDENT BROADENING

The lifetimes of electronic states in metals at low temperature are determined primarily by the frequency of collision with impurities and lattice imperfections. Since a single particle moving through a cloud of fixed, hard-core scattering centers has a mean free path independent of its speed, its collision time is inversely proportional to its speed or momentum. In addition, lattice broadening of levels may contribute an energy-dependent term to the linewidth, which is additive to $1/\tau$ at least in the limit $\hbar/\tau \ll \beta^* H.^4$ As a simple phenomenological description which is mathematically tractible, we assume

$$1/\tau = 1/\tau_0 + \alpha_0 |p_z| + \alpha_1(E).$$
 (13)

The ξ integral in Eq. (9) can be performed as before, leaving the p_z integral, which for $\alpha_0=0$ was evaluated by the method of stationary phase about the point p_m where $(\partial S/\partial p_z)_{p_m}=0$.

Near this point $S(E'', p_z)$ can be expanded as

$$S(E^{\prime\prime},p_z)\approx S_1(E^{\prime\prime})-\epsilon(E^{\prime\prime})[p-p_m]^2, \qquad (14)$$

where $\epsilon > 0$ if S has a maximum at p_m . For $\alpha_0 \neq 0$, the stationary point is moved off the real axis by an amount $i\alpha_0 \hbar m^*/2\epsilon$, and the integral can be evaluated by the method of steepest descent. Continuing the evaluation of the remaining integrals in Eq. (9) by asymptotic approximations, we find the equivalent to Eq. (12):

$$(F)_{osc} \approx 2VkT \left(\frac{eH}{hc}\right)^{3/2} |2\epsilon(\zeta)|^{-1/2} \operatorname{Re}\left[1 - \frac{i\hbar}{2\pi} \left(\frac{\partial\alpha_1}{\partial E}\right)_{\zeta}\right] \sum_{j=1}^{\infty} j^{-3/2} \exp\{-j\left[1/\tau_0 + \alpha_0 p_m + \alpha_1(\zeta)\right]/\nu_c^*\} \\ \times \left\{\sinh\left[\frac{2\pi^2 kT}{\beta^* H}\right] \cos\left[\frac{\pi kT}{\nu_c^*} \left(\frac{\partial\alpha_1}{\partial E}\right)_{\zeta}\right] - i\cosh\left[\frac{2\pi^2 kT}{\beta^* H}\right] \sin\left[\frac{\pi kT}{\nu_c^*} \left(\frac{\partial\alpha_1}{\partial E}\right)_{\zeta}\right]\right\}^{-1} \\ \times \exp\left\{\frac{icj}{\hbar eH}S_1\left[1 - \frac{(\alpha\hbar m^*)^2}{4\epsilon S_1}\right] - i2\pi j\gamma \mp i\frac{\pi}{4}\right\} \cos\left(\frac{j\pi gm^*}{2m}\right).$$
(15)

Since the actual form of $\alpha_1(E)$ is unknown, we must resort to crude estimates to determine the importance of the correction terms in Eq. (15). Assuming a linear dependence for $\alpha_1(E)$, we let $(d\alpha_1/dE)_{\zeta} \approx 1/\tau_0\zeta$, so that for $\zeta \approx 10^{-2}$ eV, $hd\alpha_1/dE \approx \hbar/\tau_0\zeta \approx 10^{-13}/\tau_0$. As DHVA experiments have been conducted on materials with τ_0 as small as 10^{-12} sec,⁸ this term can be appreciable for such extreme cases. The other energy-dependent correction term is

$$\frac{kT}{\nu_c^*} \left(\frac{d\alpha_1}{dE} \right)_{\zeta} \approx \frac{kT}{\zeta \nu_c^* \tau_0} \approx 2\pi^2 \left(\frac{kT_D}{\zeta} \right) \left(\frac{kT}{\beta^* H} \right),$$

which becomes $10^{-1}(kT/\beta^*H)$ if $T_D \approx 1^{\circ}$ K. This term is therefore more important at lower fields, where $kT/\beta^*H \approx 1$. It is thus possible that if $(d\alpha_1/dE)_{\xi}$ is large, these terms could make corrections to the phase or frequency of magnetization oscillation, depending upon the field dependence of $\alpha_1(E)$.

We shall now consider the explicit frequency correction term, $(\alpha_0 \hbar m^*)^2/4\epsilon S_1$. For an ellipsoid of revolution, $S = 2\pi m^* (E - p_z^2/2m_z)$, and with $\alpha_0^2 \approx 1/\tau_0^2 m_z \zeta$, we find this term is about $(\hbar/4\pi\tau_0 \zeta)^2 \approx (10^{-14}/\tau_0)^2$ and consequently is of second order and negligible. However this term might be noticeable if $\partial \tau/\partial p_z$ were anomalously large at the Fermi surface.

In conclusion, we wish to emphasize that the above estimates necessarily have been crude, because the energy and p_z dependence of the linewidths are unknown; however, our results indicate that such dependence could lead to noticeable effects other than an exponential decrease in amplitude.

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⁸ D. Shoenberg, Phil. Trans. Roy. Soc. London A245, 1 (1952).