Comparison of (C25) and (C30) gives

$$g_\alpha(\text{n.o.})=\frac{2i\alpha}{\pi}(4\pi\beta\zeta)^{1/2}Q_\alpha(X,X'). \quad \text{(C31)}$$

Insertion of (C31) into (C9) yields

$$I_1{}^{xx}=-6\frac{q^2}{p_F{}^2}\frac{\partial}{\partial\xi}\frac{\partial}{\partial\xi'}\eta^{3/2}\sum_\alpha\frac{\alpha^2}{\alpha^2+\gamma^2}Q_\alpha(X,X')|_{\xi=\xi'}. \quad \text{(C32)}$$

We introduce the Cohen-Harrison-Harrison[10] notation

$$s_\alpha(X)=\int_0^{\pi/2}[J_n'(X\sin\theta)]^2\sin^3\theta d\theta, \quad \text{(C33)}$$

and note that

$$\sum_{\alpha=-\infty}^{\infty}[J_\alpha'(X)]^2=\tfrac{1}{2}.$$

Our result can be written

$$I_1{}^{xx}(\text{n.o.})=\left[-1+3\gamma^2\sum_{\alpha=-\infty}^{\infty}\frac{s_\alpha(X)}{\alpha^2+\gamma^2}\right]\eta^{3/2}. \quad \text{(C34)}$$

Now we wish to express this in terms of $X_0=qv_F/\omega_c$ instead of X. Using $\eta=1-\delta_2$ and $s_\alpha(X)=s_\alpha(X_0)-X_0s_\alpha'(X)\frac{1}{2}\delta_2$, we obtain a "nonoscillating" contribution

$$I_1{}^{xx}(\text{n.o.})=-1+\tfrac{3}{2}\delta_2+3\gamma^2\sum_{\alpha=-\infty}^{\infty}\frac{s_\alpha(X_0)}{\alpha^2+\gamma^2}$$
$$-\tfrac{3}{2}\delta_2\sum_{\alpha=-\infty}^{\infty}\frac{\gamma^2}{\alpha^2+\gamma^2}\left[3s_\alpha+X_0\frac{\partial}{\partial X_0}s_\alpha(X_0)\right]. \quad \text{(C35)}$$

Combination of (C22) and (C35) gives Eq. (3.45) of the text.

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Size Effects on the Diamagnetic Susceptibility of a Free-Electron Gas*

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We consider a noninteracting electron gas constrained in one dimension by a harmonic-oscillator potential as a model of a metallic film with specular surfaces. The diamagnetic response of this system to an applied field is investigated to study the effect of sample size on both the de Haas–van Alphen (dHvA) oscillations and the steady susceptibility. When the diameters of the dominant cyclotron orbits become comparable to the effective sample "thickness," we find large departures from the familiar H^{-1} law for the dHvA oscillations. However, the steady part of the susceptibility appears to remain independent of size.

I. INTRODUCTION

THIS paper is devoted to an investigation of a model for size effects on the diamagnetic susceptibility of a degenerate free-electron gas. In particular, we study the film geometry shown in Fig. 1, in which the magnetic field is taken to be parallel to the surface of a film of thickness L. The problem is to elucidate the corrections to both (a) the steady part of the diamagnetic susceptibility and (b) the oscillatory de Haas–van Alphen (dHvA) structure which arise when the ratio $\zeta=2R_c/L$ is nonzero. (R_c is the cyclotron radius of an electron at the Fermi surface travelling in a plane perpendicular to the applied field.) We shall restrict our attention to a model (described below) which is essentially equivalent to specular reflection from the surfaces of the sample. This assumption raises the question of the applicability of such a calculation to real systems, where diffuse reflection will certainly play a role. However, recent experiments by Koch *et al.*[1] indicate that a specular boundary may be obtainable, at least for certain groups of electrons. The physical effect of the boundaries is to lift the degeneracy associated with a particular Landau level[2] which arises from cyclotron orbits centered at different positions across

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[1] F. Koch (private communication) has shown that in several experiments on the transport properties of metals such as Sn and In the results are consistent only with highly specular surface scattering.

[2] C. Kittel, *Quantum Theory of Solids* (John Wiley & Sons, Inc., New York, 1963).

the thickness of the film. As we shall see, when the parameter $\zeta \gtrsim 1$, the dHvA structure deviates appreciably from the bulk result of periodicity in H^{-1}. On the other hand, the steady (nonoscillatory) part of the susceptibility is seen to be rather insensitive to ζ. We find that the Landau result χ_L obtains whether or not the orbits are closed in the film.

Previous calculations[3] of the initial susceptibility for a constrained degenerate electron gas have found χ_L to within a size-independent numerical factor. This strange result (that this numerical factor is size-independent and different from unity) will be shown to arise from the discreteness of the energy levels in a finite system, and is of no fundamental importance.

Our model of a thin film is formed by placing the electrons into a one-dimensional harmonic-oscillator potential of classical frequency Ω. At absolute zero temperature and in the absence of a magnetic field, the electrons fill this well to some Fermi energy $\mu(0)$, which is determined by the electron density. Since only states near the Fermi surface contribute to the magnetization, we define an effective thickness L by

$$\tfrac{1}{2}m\Omega^2L^2=p^2\mu(0)\,, \tag{1.1}$$

where p is a coefficient of order unity which is adjusted to give the usual free-electron relation between density and chemical potential in the limit $L\to\infty$. Such a model was used by Friedman[3] for the case of a nondegenerate electron gas where he, indeed, found the Landau result. The special feature of this model which makes it useful is that the constraining potential is commensurate with the effective potential arising from the magnetic field; thus the exact energy spectrum can be found by an elementary transformation. We may then calculate the diamagnetic susceptibility without recourse to approximate treatments of the surface (e.g., WKB approximation), even for specular reflection. Section II is devoted to a calculation of the internal energy and field-dependent Fermi energy for this model. Section III then discusses the diamagnetic behavior of the system.

II. ENERGY CALCULATIONS

With the model described in the Introduction and the geometry of Fig. 1, the single-particle Hamiltonian for the constrained noninteracting electron gas is

$$\mathcal{H}=(2m)^{-1}[\mathbf{P}-(e/c)\mathbf{A}]^2+\tfrac{1}{2}m\Omega^2y^2\,, \tag{2.1}$$

where the vector potential $\mathbf{A}$ may be taken in the transverse gauge

$$\mathbf{A}=-H(y,0,0)\,. \tag{2.2}$$

Assuming an infinite sample in the x-z plane, the Hamiltonian is separable in Cartesian coordinates and the wave functions $\psi(\mathbf{r})$ may be expressed as

$$\psi(\mathbf{r})=\exp[i(k_xx+k_zz)]\chi(y)\,, \tag{2.3}$$

[3] L. Friedman, Phys. Rev. **134**, A336 (1964), and references therein.

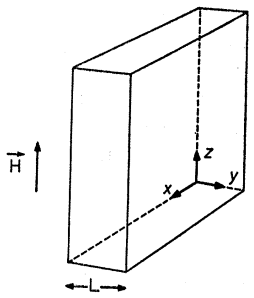

FIG. 1. Slab geometry considered in this work. The field is applied parallel to the z axis in the plane of the sample.

where (k_x,k_z) are the in-plane momenta. The linear transformation

$$y=y'-\hbar\omega_ck_x/m\omega^2 \tag{2.4}$$

reduces the Schrödinger equation to the form of a one-dimensional harmonic oscillator

$$-\frac{\hbar^2}{2m}\frac{d^2\chi}{dy'^2}+\left(\frac{1}{2}m\omega^2y'^2-\frac{1}{2}\frac{\hbar^2\omega_c^2}{m\omega^2}k_x^2\right)\chi(y')=E'\chi(y')\,, \tag{2.5}$$

where $\omega_c=eH/mc$ is the cyclotron frequency and

$$\omega^2=\omega_c^2+\Omega^2. \tag{2.6}$$

The total energy E of a given eigenstate is then

$$E=E'+(2m)^{-1}\hbar^2(k_x^2+k_z^2)\,, \tag{2.7}$$

where E' has the well-known form

$$E'=(n+\tfrac{1}{2})\hbar\omega-\tfrac{1}{2}(\hbar^2\omega_c^2/m\omega^2)k_x^2\,,\quad n=0,1,2\cdots. \tag{2.8}$$

Thus the single-particle energy levels are

$$E(n,k_x,k_z)=(n+\tfrac{1}{2})\hbar\omega+(2m)^{-1}\hbar^2k_z^2+(2m)^{-1}\hbar^2k_x^2(\Omega/\omega)^2. \tag{2.9}$$

This result exhibits explicitly the removal of the degeneracy of the Landau levels corresponding to classical orbits whose centers are at different positions across the thin dimension of the film. In the limit of an infinitely thick sample ($\Omega\to 0$) the levels coalesce and states with different centers (i.e., different quantum numbers k_x) become degenerate. The corresponding wave functions are as usual Hermite polynomials. However, since the energy spectrum suffices to calculate the bulk magnetic moment, we shall not discuss the eigenfunctions further in this paper.

The Pauli exclusion principle requires that a given orbital state is at most doubly occupied (spin up or down). For the problem at hand, we shall neglect the Pauli paramagnetism which can easily be included, but would unnecessarily complicate the equations. Thus spin shall only be included for counting (i.e., we shall assume that the up- and down-spin states for a given orbital are degenerate). We shall also, for simplicity, perform all calculations at absolute zero temperature—this restriction can also be easily removed. Then as electrons are added to the system, they will fill up the doubly degenerate orbital levels described by (2.9) until

all the electrons in the metal are accommodated. The maximum energy of any electronic state is then the chemical potential (or Fermi energy). Since these states near the Fermi energy play the dominant role in the magnetization, the Fermi energy must be rather carefully calculated. At 0°K, the total number of electrons is given by

$$N=2\sum_{n,k_x,k_z}{}' 1, \tag{2.10}$$

where the prime indicates that the summation is performed over the occupied states, i.e., $E(n,k_x,k_z)<\mu$. Replacing the summation over the continuous quantum numbers k_x and k_z by integrals, (2.10) can be rewritten

$$N=(S/2\pi^2)\sum_{n<\bar{n}}\int\int dk_x dk_z, \tag{2.11}$$

where $\bar{n}$ is the value of the quantum number n of the most energetic occupied level, and S is the area of the film. Using the energy spectrum (2.9), the integrals and summation can easily be performed to give

$$N=(mS/4\pi\hbar^2)(\omega/\Omega)(\bar{n}+1)[\mu-\tfrac{1}{2}\hbar\omega(\bar{n}+1)], \tag{2.12}$$

which, together with the condition

$$(\bar{n}+\tfrac{1}{2})\hbar\omega\lesssim\mu, \tag{2.13}$$

determines the chemical potential μ. Now the effective film thickness L can be found by using (1.1) and requiring that in the limit $L\to\infty$ (and in zero field) the relation between the Fermi energy and electron density derived from (2.12) agrees with the equivalent relationship for a free-electron gas:

$$N=(LS/3\pi^2)[2m\mu(0)/\hbar^2]^{3/2}. \tag{2.14}$$

This fixes the coefficient p in (1.1) to be $\frac{3}{8}\pi$. Then for a given effective film thickness, (1.1) uniquely determines the corresponding oscillator frequency. This procedure relates our model to a physical film.

Once the Fermi energy is formed, we can calculate the total internal energy of the electron gas by summing the energy spectrum (2.9) over all occupied states. It is convenient to write U in the form

$$U=U_n+U_x+U_z, \tag{2.15}$$

corresponding to the three terms in (2.9), where

$$U_i=\sum_{n,k_x,k_z}{}' E_i. \tag{2.16}$$

Each term is calculated in a fashion similar to that for the electron number N (i.e., summing over n and integrating over k_x and k_z). The results are

$$U_n=\sum_{n,k_x,k_z}{}'(n+\tfrac{1}{2})\hbar\omega=(mS/8\pi\hbar)(\omega^2/\Omega)(\bar{n}+1)\times[(\bar{n}+1)\mu-\tfrac{2}{3}\hbar\omega(\bar{n}+\tfrac{1}{2})(\bar{n}+\tfrac{3}{2})] \tag{2.17}$$

and

$$U_x=U_z=(mS/16\pi\hbar)(\omega^2/\Omega)(\bar{n}+1)[(\mu^2/\hbar\omega)-\mu(\bar{n}+1)+\tfrac{1}{3}\hbar\omega(\bar{n}+\tfrac{1}{2})(\bar{n}+\tfrac{3}{2})],$$

which gives

$$U=(mS/2\pi\hbar^2)(\omega^2/\Omega)(\bar{n}+1)\times[\mu^2-\tfrac{1}{3}(\hbar\omega)^2(\bar{n}+\tfrac{1}{2})(\bar{n}+\tfrac{3}{2})]. \tag{2.18}$$

We now define the dimensionless quantity ϵ, which describes how far below the Fermi surface lies the level $(\bar{n}+\frac{1}{2})\hbar\omega$, by

$$\epsilon=(\mu/\hbar\omega)-(\bar{n}+\tfrac{1}{2}), \tag{2.19}$$

where ϵ has the property that $1\geqslant\epsilon\geqslant 0$. In terms of ϵ, (2.12) and (2.18) can be written as

$$N=(mS/2\pi\hbar^2)(\omega/\Omega)(\hbar\omega)[(\mu/\hbar\omega)^2-(\epsilon-\tfrac{1}{2})^2] \tag{2.20}$$

and

$$U=(mS/3\pi\hbar^2)(\omega/\Omega)(\hbar\omega)^2[(\mu/\hbar\omega)+\tfrac{1}{2}-\epsilon]\times[(\mu/\hbar\omega)^2+(\mu/\hbar\omega)(\epsilon-\tfrac{1}{2})+\tfrac{1}{2}\epsilon(1-\epsilon)]; \tag{2.21}$$

the energy per electron becomes

$$u\equiv U/N=\tfrac{2}{3}\mu(H)+\tfrac{1}{3}\rho(H), \tag{2.22}$$

where the energy $\rho(H)$ is

$$\rho(H)\equiv\hbar\omega\epsilon(1-\epsilon)(\mu/\hbar\omega-\tfrac{1}{2}+\epsilon)^{-1}.$$

In the limit of zero field and $L\to\infty$, the energy per electron in this model becomes $\frac{2}{3}\mu$, which is somewhat different than the standard free-electron-gas result of μ. The discrepancy arises from the somewhat different densities of states in the two models. In the subsequent calculations based on the energy per particle (2.22), this slight departure from a perfectly-free-electron gas should be kept in mind.

III. DIAMAGNETISM

The magnetic moment per electron M/N is now formed from

$$M/N=-\partial u/\partial H. \tag{3.1}$$

Before actually computing $M(H)$, it is useful to discuss the behavior of the parameters ϵ and $\bar{n}$ as a function of magnetic field. Consider some initial state of the system when the field has a value H_i and there is a corresponding ϵ_i and $\bar{n}_i$, where the subscript i denotes the initial values. As the field increases, ϵ decreases from the value ϵ_i because the spacing between levels increases. At some particular value of the field H_n, ϵ becomes zero, i.e., $(\bar{n}_i+\frac{1}{2})\hbar\omega(H_n)=\mu(H_n)$, which is the condition that the energy level in question (of a "belly" orbit) passes through the Fermi surface. As the belly-orbit Landau level passes through the Fermi surface, ϵ goes to zero and then abruptly changes to 1 as the number of occupied levels $\bar{n}$ decreases by 1. An explicit representation for ϵ can easily be obtained if the critical frequencies ω_n for the coincidence of the highest level with the

Fermi level is defined by

$$\mu(H_n)=(\bar{n}+\tfrac{1}{2})\hbar(\omega_n^2+\Omega^2)^{1/2}, \tag{3.2}$$

i.e.,

$$\omega_n=eH_n/mc. \tag{3.3}$$

Using (2.12), (2.19), and (3.2), ϵ becomes

$$\epsilon=\tfrac{1}{2}\bar{n}\omega^{-2}(\omega_n^2-\omega_c^2). \tag{3.4}$$

Thus when $\omega_c\to\omega_n$, $\epsilon\to 0$. It is easily verified that this would be equivalent to $\epsilon=1$ (at $H=H_n$) had we chosen to write (3.4) in terms of $\bar{n}-1$ and $\omega_{\bar{n}-1}$. It is also important to notice from (2.19) that while ϵ is a discontinuous function of the field, both the chemical potential and $\rho(H)$ are continuous. Throughout the subsequent calculation of the magnetization, it will be convenient to study a given period of ϵ, i.e., the field domain between the passage of one Landau level through the Fermi surface until the next. The field derivative of the chemical potential in a given interval is then

$$\partial\mu/\partial(\hbar\omega_c)=(\omega_c/\omega)(\bar{n}+\tfrac{1}{2}+\epsilon)+\omega\partial\epsilon/\partial\omega_c, \tag{3.5}$$

where [from (3.4)]

$$\partial\epsilon/\partial\omega_c=-\bar{n}\omega_c\omega^{-4}(\omega_n^2+\Omega^2). \tag{3.6}$$

Combining (3.5) and (3.6), we get

$$\partial\mu/\partial(\hbar\omega_c)=(\omega_c/2\omega)[1+\bar{n}\omega^{-2}(\omega_c^2-\omega_n^2)]. \tag{3.7}$$

Similarly, one easily shows that

$$\partial\rho/\partial(\hbar\omega_c)=(\omega_c/2\omega)[1-3\omega^2(\omega_n^2+\Omega^2)^{-1}]+\bar{n}(\omega_c/4\omega)\omega^{-2}(\omega_n^2-\omega_c^2)\times[1+3\omega^2(\omega_n^2+\Omega^2)^{-1}], \tag{3.8}$$

which then gives [from (2.22) and (3.1)]

$$M=\beta(\omega_c/\omega)(\omega_n^2-\omega_c^2)(\omega^2+\Omega^2)^{-1}\times[(\bar{n}/2\omega^2)(\omega_n^2-\omega_c^2)-1], \tag{3.9}$$

where β is the Bohr magneton. This can conveniently be rewritten as

$$M=-\beta\bar{n}^{-1}\epsilon(1-\epsilon)\omega_c\omega(\omega_n^2+\Omega^2)^{-1}, \tag{3.10}$$

which is our main result. It is interesting to note that this gives a closed form [together with (3.4)] for the field dependence of the magnetization associated with a given period, i.e., for a particular Landau level. It now remains to analyze the consequences of this formula; this shall be the scope of the remaining discussion.

Since $\epsilon\gtrless 0$, the magnetization is always diamagnetic with minima at $\epsilon=0$, where the magnetization is zero. This occurs when the highest Landau level lies just at the Fermi surface and hence has no partial occupancy. This is then an essentially classical situation wherein the Bohr–van Leeuwen theorem states that there should be no magnetization, in agreement with (3.10). If we take the zeros of M to denote the positions of the dHvA oscillations, the periodicity may be determined from

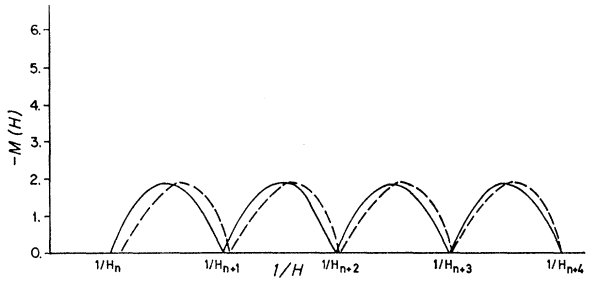

FIG. 2. Plot of $M(H)$ as a function of H^{-1} from (3.10) for $L=10^{-4}$ cm, $\zeta\cong 10^{-1}$. The dashed curve is $M(H)$ for $\zeta=0$, i.e., a bulk sample. The sketched curve is only for a few oscillations near $H\simeq 10^6$ Oe.

(2.19) and (2.20) by fixing the total number of electrons in the system. Setting $\epsilon=0$, we find

$$\hbar^2(\omega_n^2+\Omega^2)\bar{n}(\bar{n}+1)=\text{const}\cong\mu_{\min}^2, \tag{3.11}$$

where $\mu_{\min}$ is the value of the Fermi energy when the highest Landau level (in zero field) passes through the Fermi surface. It has been assumed in this expression that $\mu_{\min}\gg\hbar\Omega$, which is always the case in degenerate systems. The size effect on the dHvA period can now be obtained directly from (3.10) if we use the parameter $\zeta=2R_c/L\cong 2\Omega/\omega_c$. Then for orbits which are small compared to the film thickness, $\zeta\ll 1$, we find in the degenerate region $\bar{n}\gg 1$ the standard result,

$$\bar{n}\hbar\omega_n\cong\mu_{\min}, \tag{3.12}$$

for the appearance of nodes in $M(H)$; i.e., there is a periodicity in H^{-1}. However, in the large orbit limit $\zeta\gg 1$, we find that

$$\bar{n}\hbar\omega_n\cong(\bar{n}^{-2}\mu_{\min}^2-\hbar^2\Omega^2)^{1/2}. \tag{3.13}$$

In order to more easily see how the periodicity changes with ζ, it is convenient to consider the difference in fields for two adjacent nodes in M, i.e.,

$$\omega_{n-1}^2-\omega_n^2\cong 2\hbar^{-2}\mu_{\min}^2=2\bar{n}^{-1}(\omega^2+\Omega^2)\cong p^2v_f^2(1+4\zeta^2)/4\bar{n}\zeta^2L^2. \tag{3.14}$$

Thus for small ζ,

$$\omega_{n-1}^2-\omega_n^2\simeq(pv_f)^2(4\bar{n}\zeta^2L^2)^{-1}, \tag{3.15}$$

which is independent of thickness but proportional to $\bar{n}^{-1}$. On the other hand, for large orbits,

$$\omega_{n-1}^2-\omega_n^2\simeq(pv_f)^2(\bar{n}L^2)^{-1}, \tag{3.16}$$

which is clearly dependent on size and varies only as $\bar{n}^{-1}$. This shows the onset of a type of geometric resonance with standing modes across the film. This type of behavior is readily observable in Figs. 2 and 3, where the magnetization curve is sketched for some different values of ζ.

The relative maxima in the $M(H)$ curve occur very nearly at $\epsilon=\frac{1}{2}$, as is seen from (3.10), because the only rapidly varying function of field in this expression is ϵ. Then if ω_n^* denotes the value of ω_c where $M(H)$ is

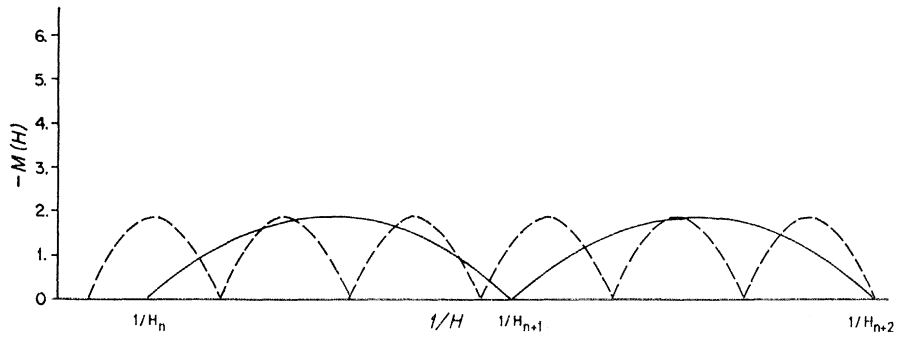

FIG. 3. Plot of $M(H)$ or H^{-1} for $L=10^{-4}$ cm but a lower field $H\simeq 10^5$ Oe corresponding to $\zeta\cong 1$. The dashed curve is the $\zeta=0$ result for comparison. Notice the large periodicity change but relative constancy of the amplitude of the oscillations.

maximum (M_n^*) in the nth period, we find, at the maximum,

$$M_n^*\cong(4\bar{n})^{-1}\beta\omega_n^*(\omega_n^*+\Omega^2)^{1/2}(\omega_n^2+\Omega^2)^{-1}$$
$$\cong-(4\mu_{\min})^{-1}\beta\hbar\omega_n^*(\omega_n^{*2}+\Omega^2)(\omega_n^2+\Omega^2)^{-1}. \quad (3.17)$$

However, ω_n^* and ω_n are always very close in field, and thus M_n^* is nearly independent of size. Similarly, the steady (monotonic) component of the magnetization is independent of size because $\langle M\rangle$ in a given period is

$$\langle M\rangle\cong-\beta(6\bar{n})^{-1}\omega_n(\omega_n^2+\Omega^2)^{-1/2}\cong-\beta^2H/3\mu_{\min}. \quad (3.18)$$

Figures 2 and 3 plot $M(H)$ from (3.10) for two cases, respectively: $\zeta=0.1$, which corresponds nearly to the bulk situation, and $\zeta=1$, which exhibits the large periodicity changes. It is evident that the amplitude of the oscillations is independent of ζ, and in fact the steady susceptibility also maintains its bulk (Landau) value independent of ζ.

The last point to be mentioned is the fact that the initial susceptibility, i.e., $\partial M/\partial H|_{H\to 0}$, does not give very precise information about the system. This is again seen from (3.10). The initial magnetization depends on $\epsilon(0)$, i.e., the value of ϵ for zero field. This, however, is a delicate function of the size of the sample and the electron density. Basically the spatial quantization gives rise to a quasidiscrete spectrum even in zero field. Thus effectively the initial susceptibility corresponds to some point on a dHvA oscillation and hence may even be paramagnetic, i.e., $\chi>0$. It is only by studying many periods that some meaningful results can be obtained about geometric effects.

In conclusion, we have shown, for our particular model of a thin film, that the steady diamagnetism as well as the amplitudes of the dHvA oscillations is rather insensitive to the ratio R_c/L. However, we do find large changes in the dHvA periodicity when this ratio becomes comparable to unity. While we believe that our artificial model gives, at least qualitatively, the correct behavior for specular reflection, it must be emphasized that diffuse surface scattering could completely obscure the striking effects on the dHvA oscillations. However, we expect our result, that the average diamagnetism is essentially unaffected by specular scattering of electrons from the surface, to remain valid even in the diffuse-scattering situation.

Further investigations on this problem will include the effects of (a) finite-temperature effects, (b) impurity scattering, and (c) electron-electron interactions.

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