Orbital Paramagnetism of Electrons in a Two-Dimensional Lattice

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It is shown that the orbital response of a two-dimensional electron gas in a periodic potential to a weak magnetic field is always paramagnetic when the Fermi level is sufficiently close to a saddle point of the band structure. Another instance of orbital paramagnetism is found when the Fermi level is close to a narrow indirect gap.

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Recently, there has been considerable interest in the behavior of a two-dimensional noninteracting electron gas under the influence of both a periodic potential and a magnetic field [1-4]. In the absence of the periodic potential the ground-state energy is generally *increased* by the magnetic field B, except in the special case of completely filled Landau levels where it equals the $B=0$ value. In the weak magnetic-field regime ($\mu B \ll k_B T \ll E_F$, where μ is the magnetic moment, T is the temperature, and E_F is the Fermi energy) one obtains the well-known Landau-Peierls diamagnetic susceptibility [5] $\chi_0 = -e^2/24\pi mc^2$ in two dimensions (we ignore spin for simplicity). On the other hand, Hasegawa *et al.* [1], working with a schematic tight-binding model where the effect of the magnetic field is taken into account by means of the Peierls substitution [6], find that the ground-state energy is generally *decreased* by the magnetic field, and attains an absolute minimum when there is one flux quantum per electron. In the weak magnetic-field limit, this model yields a *paramagnetic* (i.e., positive) orbital susceptibility provided that the occupation fraction of the band is between 20% and 80% [7].

The tight-binding model of Hasegawa *et al.* [1] can be and has been criticized on the basis that it does not include the diamagnetic energy of the tight-binding orbitals and the reduction of the hopping amplitude by the magnetic field. When these effects are taken into account the magnetic response appears to revert to familiar atomic diamagnetism. In the case of a weak periodic potential, it is found, treating the periodic potential in second-order perturbation theory, that the magnetic field generally increases the ground-state energy [3,4]. Consistent with this, the weak magnetic-field susceptibility is found to be diamagnetic. All these findings seem to imply that orbital paramagnetism, although possible in principle, is not realized in nature.

Actually, the case for orbital paramagnetism in two dimensions is much better than the above results suggest. This will be demonstrated in this Letter for the physical case of weak magnetic field [8] and finite periodic potential. Our theory treats the periodic potential exactly, and the magnetic field perturbatively. Thus, we are able to calculate the exact low-magnetic-field susceptibility in a given periodic potential. In contrast to this, the approaches of Refs. [3] and [4] start from a finite magnetic field, which they treat exactly, but then treat the periodic potential approximately, either by a perturbative expansion around the uniform state, or by a tight-binding model. These approaches are unable to capture all the effects of the band structure on the low-magnetic-field susceptibility. We shall see that the exact susceptibility contains terms which depend only on the properties of electrons at the Fermi surface as well as many other terms which depend on all the occupied states. Ordinarily, the two types of terms are of the same order of magnitude. However, in a two-dimensional system, when the Fermi level is close to a saddle point of the band structure, the Fermi surface term takes over, due to the logarithmic divergence of the density of states. We shall show that this contribution is always paramagnetic. The phenomenon can be physically understood as a consequence of magnetic breakdown of quasiclassical electronic orbits in the vicinity of a saddle point, where the quasiclassical approximation fails. Even if the Fermi level is not close to a saddle-point singularity, the orbital response can still be paramagnetic. An instance of this effect is found when the Fermi level approaches a narrow indirect gap. This will be explicitly demonstrated below, by solving a model periodic potential.

We start from the second-order perturbative expansion for the shift of the ground-state energy per unit area of a periodic, noninteracting system in the presence of a vector potential $\mathbf{A}(\mathbf{r})$:

$$\Delta E^{(2)} = \frac{e^2}{2c^2}\int_{\mathrm{BZ}} \frac{d^2q}{(2\pi)^2} \sum_{\mathbf{G},\mathbf{G}'} \sum_{\mu,\nu} A_\mu(\mathbf{q}+\mathbf{G}) \left[R_{\mu\nu}(\mathbf{q}+\mathbf{G},\mathbf{q}+\mathbf{G}') + \frac{\delta_{\mu\nu} n(\mathbf{G}'-\mathbf{G})}{m} \right] A_\nu(\mathbf{q}+\mathbf{G}') . \quad (1)$$

Here $R_{\mu\nu}(\mathbf{q}+\mathbf{G},\mathbf{q}+\mathbf{G}')$ is the current-current response function, $\mathbf{q}$ is a wave vector in the first Brillouin zone, $\mathbf{G}$ and $\mathbf{G}'$ are reciprocal-lattice vectors, $n(\mathbf{G})$ is a Fourier component of the electronic density, $\mathbf{A}(\mathbf{q})$ is the Fourier transform of the vector potential, and m is the electron mass. In a noninteracting system

$$R_{\mu\nu}(\mathbf{q}+\mathbf{G},\mathbf{q}+\mathbf{G}') = \int_{\mathrm{BZ}} \frac{d^2k}{(2\pi)^2} \sum_{n,n'} \frac{f(E_{n,k-q/2}) - f(E_{n',k+q/2})}{E_{n,k-q/2} - E_{n',k+q/2}} \Lambda^{\mu *}_{nn'}(\mathbf{k};\mathbf{q}+\mathbf{G}) \Lambda^{\nu}_{nn'}(\mathbf{k};\mathbf{q}+\mathbf{G}') , \quad (2)$$

where E_{nk} are band energies, $f(E)$ is the Fermi distribution function,

$$\Lambda^{\nu}_{nn'}(\mathbf{k};\mathbf{q}+\mathbf{G}) = \frac{\hbar}{m}\langle u_{n',k+q/2}|e^{i\mathbf{G}\cdot\mathbf{r}/2}(k_\nu - i\nabla_\nu)e^{i\mathbf{G}\cdot\mathbf{r}/2}|u_{n,k-q/2}\rangle \tag{3}$$

is the current vertex, and $|\psi_{n,k}\rangle = \exp(i\mathbf{k}\cdot\mathbf{r})|u_{n,k}\rangle$ are Bloch states. A crucial property of the current-current response function is

$$\sum_\nu R_{\mu\nu}(\mathbf{q}+\mathbf{G},\mathbf{q}+\mathbf{G}')(q_\nu+G'_\nu) = -\frac{1}{m}n(\mathbf{G}'-\mathbf{G})(q_\mu+G'_\mu)\,. \tag{4}$$

This follows from gauge invariance of the current response and guarantees that the continuity equation for the current is satisfied. In the limit $q\to 0$, $\mathbf{G}'=0$, Eq. (4) implies

$$R_{\mu\nu}(\mathbf{G},0) = -\frac{1}{m}n(-\mathbf{G})\delta_{\mu,\nu}\,. \tag{5}$$

Now, using the Landau gauge for the uniform magnetic field $\mathbf{A}(r) = -By\hat{\mathbf{x}}$, substituting in Eq. (1), and using Eqs. (2)-(5), we obtain the orbital magnetic susceptibility per unit area as

$$\chi = -\frac{\partial^2 E}{\partial B^2}\bigg|_{B=0} = -\frac{e^2}{c^2}\frac{1}{2}\frac{\partial^2}{\partial q^2}R_{xx}(q\hat{\mathbf{y}},q\hat{\mathbf{y}})\bigg|_{q=0}\,. \tag{6}$$

This expression provides a simple way of calculating the noninteracting susceptibility for an arbitrary band structure. Note that it is much more compact than any previous expression based on a direct expansion of the free energy [9]. Of course, in the free-electron gas limit, it reduces to the Landau-Peierls result [5].

In Eq. (2) we distinguish intraband contributions ($n=n'$) and interband contributions ($n\neq n'$). The intraband contributions can all be reduced to integrals over the Fermi line

$$\chi_{\text{intra}} = -\frac{e^2}{4\pi^2\hbar^2c^2}\sum_n{}'\oint\frac{dl_k^{(n)}}{|\nabla_k E_{nk}|}\left\{\frac{1}{12}[\partial_x^2E_{nk}\partial_y^2E_{nk} - (\partial_x\partial_yE_{nk})^2] + 2\partial_xE_{nk}\lambda^{(1)}(\mathbf{k}) - |\lambda^{(2)}(\mathbf{k})|^2\right\}, \tag{7}$$

where the primed sum is taken over the bands which intersect the Fermi level and $dl_k^{(n)}$ is the length element along the nth segment of the Fermi line. ∂_μ is a shorthand for $\partial/\partial k_\mu$. Explicit expressions for $\lambda^{(1)}(k)$ and $\lambda^{(2)}(k)$ can be given in terms of Bloch eigenvalues and wave functions:

$$\lambda^{(1)}(\mathbf{k}) = \frac{\hbar^2}{m}[\langle u_{n,k}|-i\nabla_x|u_{n,k}^{(2)}\rangle + \langle u_{n,k}^{(2)}|-i\nabla_x|u_{n,k}\rangle - \langle u_{n,k}^{(1)}|k_x - i\nabla_x + \frac{m}{\hbar^2}\partial_xE_{n,k}|u_{n,k}^{(1)}\rangle]\,, \tag{8}$$

$$\lambda^{(2)}(\mathbf{k}) = \frac{\hbar^2}{m}[\langle u_{n,k}|-i\nabla_x|u_{n,k}^{(1)}\rangle - \langle u_{n,k}^{(1)}|-i\nabla_x|u_{n,k}\rangle]\,, \tag{9}$$

where $|u_{n,k}^{(1)}\rangle$ and $|u_{n,k}^{(2)}\rangle$ are defined by the perturbative expansion

$$|u_{n,k+q\hat{y}/2}\rangle = |u_{n,k}\rangle + q|u_{n,k}^{(1)}\rangle + q^2|u_{n,k}^{(2)}\rangle + \cdots\,. \tag{10}$$

The interband contribution, on the other hand, is given by a sum over all occupied states:

$$\chi_{\text{inter}} = -\frac{e^2\hbar^2}{m^2c^2}\frac{\partial^2}{\partial q^2}\left[\sum_{\substack{n,n'\\ n\neq n'}}\int\frac{d^2k}{(2\pi)^2}\frac{f(E_{n,k})}{E_{n,k}-E_{n',k+q\hat{y}}}|\langle u_{n,k}|-i\nabla_x+k_x|u_{n',k+q\hat{y}}\rangle|^2\right]_{q=0}\,. \tag{11}$$

The second derivative can be expressed in terms of Bloch wave functions, but these expressions are quite lengthy and will be given elsewhere.

Equations (7)-(11) are very complex. The orbital susceptibility is the sum of many terms, some diamagnetic, others paramagnetic, most of which elude a simple physical interpretation. It is generally impossible to know the sign of the result without performing the calculation. A remarkable exception occurs in two-dimensional systems when the Fermi energy happens to be near a saddle point of the band structure. The shape of the constant-energy curves in the vicinity of the saddle point is given by four branches of hyperbolae as shown in Fig. 1. It is well known that the vanishing of $|\nabla E_{nk}|$ at the saddle point leads to a logarithmic divergence in the density of states. Since the second derivatives of the energy are finite at the saddle point, we see that the term within square brackets of Eq. (7) diverges logarithmically

$$\chi_{\text{intra}} \cong +\frac{e^2\hbar^2}{12c^2}\frac{N(E_F)}{|m_1||m_2|}\,, \tag{12}$$

where $N(E_F)$ is the density of states at the Fermi level and m_1,m_2 are the two effective masses at the saddle

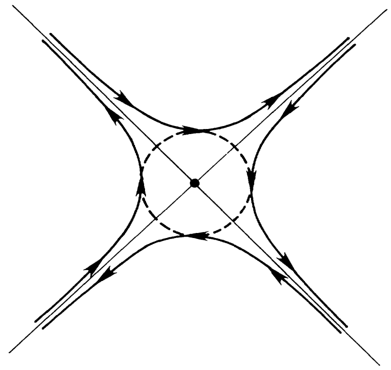

FIG. 1. Expanded view of quasiclassical electron trajectories (solid lines) near a saddle point. The broken line depicts the tunneling motion of electrons.

point [eigenvalues of the matrix $(\partial_\alpha \partial_\beta E_{nk}/\hbar^2)^{-1}$]. This term is positive definite, i.e., paramagnetic, because $m_1 m_2 < 0$. All other terms in the susceptibility remain finite as the Fermi level approaches the saddle point. In the $\lambda^{(1)}$ term of Eq. (7) the saddle-point singularity is canceled by the vanishing factor $\partial_x E_{nk}$. The $\lambda^{(2)}$ term —which is always paramagnetic—is also nonsingular at the saddle point. This is because the vanishing of $\mathbf{V}_k E_{nk}$ at the saddle point $\mathbf{k}_0$ requires the Bloch wave functions ψ_{n,k_0} to be purely real or $u_{n,k_0} = \exp(-i\mathbf{k}_0 \cdot \mathbf{r})\bar{u}_{n,k_0}$, where $\bar{u}_{n,k_0}$ is real. This fact, together with the definition of $u^{(1)}_{n,k}$ [Eq. (10)], can be used to prove that $\lambda^{(2)}(\mathbf{k}_0) = 0$ [10]. Finally, the sum over occupied states [Eq. (11)] is insensitive to Fermi-surface singularities. Thus Eq. (12) describes the exact asymptotic behavior of the susceptibility as E_F approaches the singularity.

The physical reason for the paramagnetic response near a saddle point is easily understood. In the quasiclassical approximation the electrons move along constant-energy curves shown in Fig. 1. As the electrons approach the saddle point, however, their group velocity tends to zero. In this region the quasiclassical approximation fails, and the electrons tunnel from one trajectory to the neighboring one as shown by the dashed line in Fig. 1. This is the well-known phenomenon of magnetic breakdown [11]. The resulting rotation of the electron is in a direction opposite to the classical direction of rotation. The induced magnetic moment is reversed.

At this point a few numerical calculations are required in order to assess quantitatively the size of the effect and hence the possibility of observing it in susceptibility measurements. We take as a model the separable two-dimensional potential

$$V(x,y) = 2V_0[\cos(2\pi x/a) + \cos(2\pi y/a)]\,. \qquad (13)$$

By varying the dimensionless parameter $\alpha = ma^2V_0/4\pi^3\hbar^2$ we can go from the free-electron gas regime ($V_0 \to 0$) to the tight-binding regime ($V_0 \to \infty$). The eigenvalues and eigenfunctions of this problem are given by

$$\psi_{n_x,k_x;n_y,k_y}(x,y) = \psi_{n_x,k_x}(x)\psi_{n_y,k_y}(y)\,, \qquad (14)$$

and

$$E_{n_x,k_x;n_y,k_y} = E_{n_x,k_x} + E_{n_y,k_y}\,, \qquad (15)$$

where $E_{n,k}$ and $\psi_{n,k}$ are the Bloch eigenvalues and eigenfunctions of the one-dimensional problem with periodic potential $2V_0[\cos(2\pi x/a)]$, which we can solve very accurately by numerical diagonalization on a plane-wave basis. Note that the band structure has a saddle point at $k_x = \pi/a$, $k_y = 0$ and symmetry-related points.

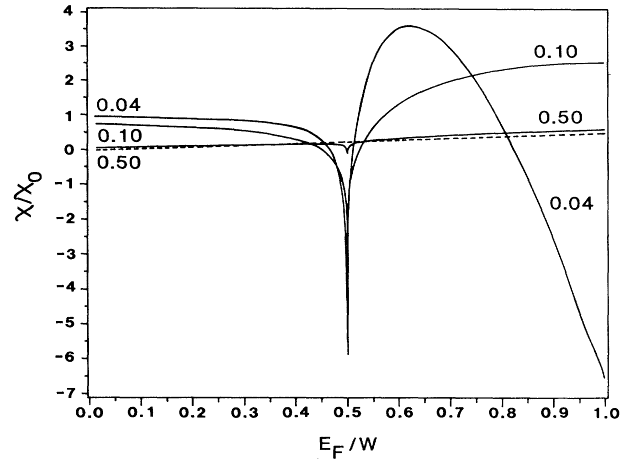

FIG. 2. Orbital susceptibility χ in units of free-electron susceptibility χ_0 as a function of Fermi energy E_F in units of the width of the lowest band W. The calculations are done for the separable model potential of Eq. (13) at three values of α (0.04, 0.10, 0.50) such that the lowest band does not overlap with other bands. The saddle point is located at $E_F/W = 0.5$. The case $\alpha = 0.04$ has a narrow indirect gap. The dashed line is the susceptibility of harmonic oscillators localized near the minima of the potential at $\alpha = 0.50$.

Figure 2 shows our results for the orbital susceptibility at various values of α. The smallest value of $\alpha = 0.04$ is chosen so that the maximum of the lowest-energy band at $k_x = k_y = \pi/a$ is barely inferior to the minimum of the next higher band at $k_x = \pi/a$, $k_y = 0$. Thus, the model has a narrow indirect gap between the two lowest bands, and we plot the susceptibility as a function of Fermi energy within the lowest band. Near the bottom of the band we obtain the diamagnetic susceptibility of a free-electron gas with effective mass $m/m^* \simeq 0.95$. As the Fermi energy increases we observe a gradual crossover from diamagnetism to paramagnetism. The paramagnetic susceptibility diverges (logarithmically) at the saddle point $E_F/W = 0.5$ as expected (W is the width of the lowest band). The location of the crossover depends sensitively on the numerical value of the nonsingular terms in Eqs. (7)-(11). These terms are diamagnetic, and tend to oppose the crossover. If we now increase the Fermi energy beyond the saddle point, we first observe a crossover from paramagnetism back to diamagnetism; then as the lowest band becomes almost completely filled we observe a second sharp reversal to paramagnetism. This unexpected behavior is entirely due to the interband term, Eq. (11). The energy denominator for the two lower bands $(E_{0,k}-E_{1,k+q\hat{y}})^{-1}$ becomes very large when $q\hat{\mathbf{y}}\rightarrow(\pi/a)\hat{\mathbf{y}}$, the wave vector connecting the maximum of the lowest band (0,0) to the minimum of the next higher band (0,1). As a result, the second derivative of this term becomes unusually large even at $q=0$ and gives a paramagnetic contribution which eventually dominates the susceptibility. Of course, this effect can only occur when the indirect gap is very narrow. In the present case, it rapidly disappears as α increases beyond 0.05 (see Fig. 2). Note that the paramagnetic orbital susceptibility both in the region of the saddle point and in the region of the narrow indirect gap can be quite large, in fact, several times larger than the Landau-Peierls susceptibility. This is therefore a large effect that should be detectable in experiments on dense materials with quasi-two-dimensional electronic structures, such as the superconducting cuprates (above T_c).

The orbital susceptibility in a tight-binding case ($\alpha=0.5$) is also plotted in Fig. 2. In this case we always observe a diamagnetic response, except in the vicinity of the saddle-point singularity where paramagnetism takes over. This result is consistent with the observation of Refs. [3] and [4] according to which one should essentially see atomic diamagnetism in the tight-binding limit. However, due to our exact treatment of the periodic potential, we also find the weak paramagnetic singularity in a narrow range of Fermi energies near the saddle point of the band structure. It is interesting to compare the exact diamagnetic susceptibility calculated from Eqs. (7)–(11) with the susceptibility of a system of independent harmonic oscillators localized near the minima of the periodic potential. The latter is shown as a dashed line in Fig. 2, at $\alpha=0.5$. Clearly, the two curves are quite similar. The agreement becomes perfect at larger values of α (the region of the saddle-point singularity shrinks to zero) giving us a nontrivial check of the correctness of our calculations.

In conclusion, I emphasize that the occurrence of a paramagnetic orbital susceptibility by no means implies that the system has a tendency toward orbital ferromagnetism or, for that matter, any other current-carrying instability. In fact, the energy required to create a weak current distribution in the noninteracting electron gas is determined, in second-order perturbation theory, by the inverse current-current response tensor [Eq. (2)], which is negative definite, ensuring stability. Thus, the question of stability is completely unrelated to the sign of the coefficient of q^2 in the small-q expansion of R_{xx} [see Eq. (6)], which determines the character (paramagnetic or diamagnetic) of the orbital susceptibility.

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