Diamagnetism of band electrons in macroscopically inhomogeneous crystals

T. A. Onishchenko
M. V. Lomonosov Moscow State University

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It is shown that the orbital motion of band electrons in a macroscopically inhomogeneous crystal can produce anomalously strong diamagnetism.

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Macroscopic inhomogeneity of a crystal, which is associated, for example, with deformations and electric fields, leads to a dependence of the electron spectrum on the space coordinates. If the depth of the space modulation of energy greatly exceeds the band width $\mathscr{E}_0$ and if the forbidden energy bands E_g are sufficiently broad, then the wave functions of electrons will be localized. Such localization of electrons of the partially filled bands may produce a diamagnetism several orders of magnitude greater than the Landau diamagnetism.

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We shall illustrate this by using a simple model—a cubic crystal in which the energy spectrum of conduction electrons has the form

$$\mathcal{E}(k_x, k_y, k_z) = \mathcal{E}(k_x) + \mathcal{E}(k_y) + \mathcal{E}(k_z), \quad \mathcal{E}\left(k + \frac{2\pi}{a}\right) = \mathcal{E}(k),$$

$$\int_{-\pi/a}^{\pi/a} \mathcal{E}(k)\, dk = 0$$

where a is the size of the unit cell. The band electrons in strong-coupling approximation and in the model crystal potential $V(x) + V(y) + V(z), V(x + a) = V(x)$ have such a spectrum. We place the crystal in a uniform electric field $\mathbf{E}||OX$ and a magnetic field $\mathbf{B}||OZ$. We shall write the Hamiltonian disregarding the spins

$$\mathcal{H} = \mathcal{E}\left(k_x, k_y - \frac{eB}{\hbar c}x, k_z\right) + Fx, \quad F = -eE.$$

If $E_g^3 F^{-2}\left|\frac{\partial^2}{\partial k_x^2}\mathcal{E}(k_x)\right|^{-1} \gg 1$, then we can ignore the interband transitions in the electric field; suppose that $\left(\frac{\hbar c}{eB\sqrt{a}}\right)^{2/3} \gg \frac{\mathcal{E}_0}{|F|} \gg a$, and the mean free path $l > \mathcal{E}_0/|F|$. At $B = 0$ the energy levels are

$$E_{N, K_y, K_z} = FaN + \mathcal{E}(K_y) + \mathcal{E}(K_z), \tag{1}$$

and the corresponding wave functions are localized in x in the regions $\mathcal{E}_0/F$ in magnitude with the centers at the points $x = aN$, where N is an integer. We shall calculate the level shift (N, K_y, K_z) with an accuracy to B^2 after the magnetic field B is turned on. We shall represent $\mathcal{H}$ in the form $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 + \mathcal{H}_2$:

$$\mathcal{H}_0 = \mathcal{E}\left(k_x, k_y - \frac{eB}{\hbar c}aN, k_z\right) + iF\frac{\partial}{\partial k_x},$$

$$\mathcal{H}_1 = -\frac{eB}{\hbar c}\frac{\partial \mathcal{E}\left(k_y - \frac{eB}{\hbar c}aN\right)}{\partial k_y}\left(i\frac{\partial}{\partial k_x} - aN\right), \tag{2}$$

$$\mathcal{H}_2 = \frac{1}{2}\left(\frac{eB}{\hbar c}\right)^2\left[\frac{\partial^2}{\partial k_y^2}\mathcal{E}\left(k_y - \frac{eB}{\hbar c}aN\right)\right]\left(i\frac{\partial}{\partial k_x} - aN\right)^2.$$

The eigenfunctions $\mathcal{H}_0$ have the form

$$\Psi_{N, K_y, K_z}(k_x, k_y, k_z) = \left[\frac{a}{2\pi}\delta\left(K_y + \frac{eB}{\hbar c}aN - k_y\right)\right.$$

$$\left.\times\, \delta\left(K_z - k_z\right)\right]^{1/2} \exp\int_{-\pi/a}^{k_x}\left[-FaN + \mathcal{E}(k')\right]\frac{i}{F}dk'_x, \tag{3}$$

and the energy levels coincide with Eq. (1). We shall regard $\mathscr{H}_1$ and $\mathscr{H}_2$ as perturbations

$$\Delta E_{N,K_y,K_z} = \Delta E^{(1)} + \Delta E^{(2)} + \Delta E^{(3)} = \langle N, K_y, K_z | \mathscr{H}_1 | N, K_y, K_z \rangle$$

$$+ \sum'_{N',K'_y,K'_z} |\langle N', K'_y, K'_z | \mathscr{H}_1 | N, K_y, K_z \rangle|^2 [E_{N,K_y,K_z} - E_{N',K'_y,K'_z}]^{-1} + \langle N, K_y, K_z | \mathscr{H}_2 | N, K_y, K_z \rangle. \quad (4)$$

Only the matrix elements $\langle N+m, K_y - \frac{eB}{\hbar c} am, K_z | \mathscr{H}_1 | N, K_y, K_z \rangle$, $m = \pm 1, \pm 2, \ldots$, whose magnitude is independent of the sign of m, are nonvanishing in $\mathscr{H}_1$; $\Delta E^{(1)} = 0$. Because of unboundedness and equidistance in N of the spectrum of E_N, K_y, K_z, $\Delta E^{(2)} \frac{1}{|F|a} \left| \mathscr{E}_0 x (\frac{eB}{\hbar c} a \frac{\mathscr{E}_0}{F})^2 \right|^2 \ll \Delta E^{(3)}$.

$$\Delta E_{N,K_y,K_z} \approx \Delta E^{(3)} = \langle N, K_y, K_z | \mathscr{H}_2 | N, K_y, K_z \rangle$$

$$= \frac{\mathscr{E}_0}{2} \left(\frac{eB}{\hbar c} a \frac{\mathscr{E}_0}{F} \right)^2 A(K_y), \quad (5)$$

$$A(K_y) = (2\pi a \mathscr{E}_0^3)^{-1} \frac{\partial^2 \mathscr{E}(K_y)}{\partial K_y^2} \int_{-\pi/a}^{\pi/a} [\mathscr{E}(k)]^2 dk, \quad |A(k_y)| \sim 1.$$

We shall not analyze the different electron scattering processes whose allowance would lead to the appearance of electric current which does not change Eq. (5) when $l > \mathscr{E}_0/|F|$. Suppose that the concentration $n \ll a^{-3}$, $T \ll \mathscr{E}_0$, and $\mathscr{E}_0(k_0) = \min\{\mathscr{E}(k)\}$. Thus, only the states with $|K_{y,z} - k_0| \ll a^{-1}$ are occupied; moreover, the occupation numbers η_N, K_y, K_z are independent of N because of electroneutrality of the system, and the energy gain of the system after B is turned on is

$$\frac{1}{V} \Delta E = \frac{1}{2} \mathscr{E}_0 \left(\frac{eB}{\hbar c} a \frac{\mathscr{E}_0}{F} \right)^2 n A(k_0), \quad A(k_0) > 0. \quad (6)$$

In the absence of macroscopic inhomogeneity, the electric field in this case, $\Delta E/V$, is determined by the Landau diamagnetism[1] and differs from Eq. (6) by a factor of the order of $(F/\mathscr{E}_0)^2 n^{-2/3} \sim 10^{-6}$ for $\mathscr{E}_0/F \sim 10^{-4}$ cm, $n \sim 10^{21}$ cm^{-3}.

We regard $\mathscr{H}_1$ and $\mathscr{H}_2$ as perturbations, since such a method of calculation is the most obvious, although it formally requires unjustifiably more rigorous constraints on the value of B. But we can easily see that $\mathscr{H}_1$, which contains $i\frac{\partial}{\partial k}$ in the first power, can be calculated exactly and $\mathscr{H}_2$ can be regarded as a perturbation. An exact calculation of $\mathscr{H}_1$ does not shift the E_{N,K_y,K_z} level under consideration. A calculation using this method gives the same results for Eqs. (5) and (6). The matrix elements of $\mathscr{H}_2$ are

much smaller than the level spacing $|F|\,a$ for $\mathscr{E}_0\left(\frac{eB}{\hbar c}a\frac{\mathscr{E}_0}{F}\right)^2 \ll |F|\,a$, which was predicted.

We shall now consider a cubic crystal with narrow valence and conduction bands $\mathscr{E}_0^{v,c} \ll E_g$ for $T \sim E_g$ in the magnetic field $B \| OZ$. Suppose that

$$\mathcal{H}^{v,\,c} = \mathcal{E}^{v,\,c}(k_x, k_y, k_z) + \Phi^{v,\,c}(x), \quad \Phi^c(x) - \Phi^v(x) = E_g(x)$$

for $T = 0$ and $B = 0$ because of deformations and other reasons and that the modulation depth $E_g(x)$ in x is much larger than $\mathscr{E}_0^{v,c}$. The strong-coupling approximation is carried out according to the parameter $\mathscr{E}_0/E_g$, and

$$\mathcal{E}^{v,c}(k_x, k_y, k_z) = -\mathcal{E}_0^{v,\,c}[\cos k_x a + \cos k_y a + \cos k_z a] + 0\left(\frac{\left(\mathcal{E}_0^{v,c}\right)^2}{E_g}\right).$$

For simplicity, we assume that the remaining bands are separated from the bands under consideration by gaps that are much larger than E_g. The bands under consideration can be the impurity bands—a donor and an acceptor band; we assume in this case that the impurities are packed in a cubic superlattice whose edges of the unit cell $a^v = a^c = a$ are oriented along the axes of the coordinates.

The method of calculating this model will be described more accurately in another paper; we present only the results here. At $T = 0$ the system is a dielectric and at $T \sim E_g$ both bands are partially filled and space charges and additional electric fields are produced; if they are taken into account, then the Hamiltonian will have the form

$$\mathcal{H}^v = \mathcal{E}^v(k_x, k_y, k_z) - \frac{1}{2}E_g(x), \quad \mathcal{H}^c = \mathcal{E}^c(k_x, k_y, k_z) + \frac{1}{2}E_g(x) \tag{7}$$

when $T \sim E_g$ and $B = 0$. The magnetic properties of the system under consideration depend on x, and the variation of the thermodynamic potential $\Omega = -T\Sigma_\alpha \ln\left(1 + \exp\frac{\mu - E_\alpha}{T}\right)$ as a result of turning on the local field B, which is reduced to a unit volume, has the form $\frac{1}{V}\delta\Omega = \frac{1}{V}(\delta\Omega^v + \delta\Omega^c)$

$$\frac{\delta\Omega^{v,\,c}}{V} \approx \frac{\mathcal{E}_0^{v,c}}{8a^3}\left(\frac{eB}{\hbar c}\,a\,\frac{\mathcal{E}_0^{v,c}}{F(x)}\right)^2 \frac{\mathcal{E}_0^{v,c}}{T}\left[1 + \exp\left(\frac{1}{2T}E_g(x)\right)\right]^{-1}\left[1 + \exp\left(-\frac{1}{2T}E_g(x)\right)\right]^{-1} = D^{v,c}B^2, \quad F(x) = \frac{1}{2}\frac{\partial}{\partial x}E_g(x). \tag{8}$$

The result (8) is correct if the conditions $\left(\frac{\hbar c}{eB\sqrt{a}}\right)^{2/3} \gg \frac{\mathscr{E}_0}{|F|} \gg a$, $\frac{\mathscr{E}_0}{|F|} < l$, $\left|\frac{\partial^2 E_g}{\partial x^2}\right| \ll \frac{F^2}{\mathscr{E}_0}$ which are evidently violated near the extrema $E_g(x)$ in which $F = \frac{1}{2}\frac{\partial}{\partial x}E_g(x) = 0$, are satisfied. However, these regions, in which this approach cannot be used, can occupy a relatively insignificant part of the crystal volume. Allowance for the spins doubles $D^{v,c}$ in Eq. (8), $D_s^{v,c} = 2D^{v,c}$.

The magnetic susceptibility corresponding to (8) is $4\pi X = -8\pi(D_s^v + D_s^c) \times [1 + 8\pi(D_s^v + D_s^c)]^{-1} \sim 0.9$ for $a = 10^{-7}$ cm, $\frac{\mathscr{E}_0}{F} = 2 \times 10^{-3}$ cm, $\frac{\mathscr{E}_0}{T} = 0.1$, $T \sim \frac{E_g}{2}$, and $T^0 = 300$ K. A calculation of the model with $\mathscr{E}_0^v \ll E_g$, $\mathscr{E}_0^c \gg E_g$, and $a^v \gg a^c$, i.e., a macroscopically inhomogeneous crystal with an impurity donor band v and a conduction band c, gives similar results.

The aim of this study is to prove that nearly ideal diamagnetism, which is not associated with electron-electron interaction, can exist in principle. The obtained effect does not appear to be a low-temperature effect.

It is possible that this type of diamagnetism has been observed in the experiment.[2]
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[1]L. D. Landau, Zs. Phys. **64**, 629 (1930).
[2]N. B. Brandt, S. V. Kuvshinnikov, A. P. Rusakov, and M. V. Semenov, Pis'ma Zh. Eksp. Teor. Fiz. **27**, 37 (1978) [JETP Lett. **27**, 33 (1978)].

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