Orbital Diamagnetism of Two-Dimensional Electrons

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Different direct perturbation theories show that the Peierls substitution leads to quantitative and qualitative errors in calculations of the energy difference with and without magnetic field for both nearly free and tight-binding two-dimensional electrons. The sources of these errors are clarified. The lowering of the electron kinetic energy in a magnetic field is shown to be in general an artifact of the Peierls substitution. Spontaneous orbital ferromagnetism is shown to be impossible. Credibility of the usual theory of Landau diamagnetism for two dimensions is reestablished.

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The so-called "Peierls substitution,"[1]

$$\varepsilon(\mathbf{k}) \to \varepsilon(-i\nabla - e\mathbf{A}),$$

or

$$t(\mathbf{m}-\mathbf{m}') \to t(\mathbf{m}-\mathbf{m}')\exp\left[ie\int_{\mathbf{m}}^{\mathbf{m}'}\mathbf{A}(\mathbf{r})d\mathbf{r}\right], \qquad (1)$$

is usually adopted for a slowly varying magnetic field **A** in the vicinity of the bottom or top of an energy band $\varepsilon(\mathbf{k})$.[2] Here t is a hopping integral.

Hasegawa, Lederer, Rice, and Wiegmann,[3] through the substitution Eq. (1), claim that the energy of two-dimensional noninteracting spinless electrons on a lattice has an absolute minimum when the flux per plaquette, φ (in units hc/e), exactly equals the electron density ν per cell. They also claim "the failure in this case of the usual theory of Landau diamagnetism." This has been conformed by other workers,[4-7] using the same substitution. In particular, Abanov and Khveshchenko,[6] using the Peierls substitution in the nearly-free-electron limit, obtain a negative energy difference ΔE, with $\varphi=\nu$ and without a magnetic field, proportional to ν^3, thus confirming that "the system of two-dimensional lattice fermions has a tendency toward a spontaneous generation of orbital currents."

However, the Peierls substitution is a rather uncontrolled approximation,[8] so that the reliability of these results remains in doubt. Moreover, Nicopoulos and Trugman[9] using second-order perturbation theory in a crystal field showed that for many weak potentials the lowest-energy state is in fact at zero magnetic field.

In this Letter we calculate the total energy of two-dimensional electrons in a magnetic field with the Peierls substitution and compare this with direct perturbation theories: First, we discuss nearly free electrons for the whole region of concentration; second, we treat the tight-binding limit. We clarify the limits of the Peierls substitution. For nearly free electrons we show that the Peierls substitution is an asymptotic approximation, which leads to serious errors in the energy difference both in the low-concentration limit and in the high-concentration one. We show that the kinetic energy with a magnetic field is higher than without it, both at low and at high concentrations. For a strong periodic potential such that the tight-binding limit may be used, we generalize the Peierls approach,[1] taking into account the diamagnetic atomic shift and the dependence of the *value* of the hopping integral, Eq. (1), on the magnetic field. Using an exact solution for a model crystal field we show that a strong magnetic field shifts the atomic level and diminishes the hopping integral, increasing the total energy. We underline that the usual theory of the orbital diamagnetism, based on the existence of uncompensated surface diamagnetic currents, is also credible for two-dimensional electrons.

The energy spectrum of the nth Landau subband for nearly free electrons is given by

$$\det\left\|(\varepsilon-\varepsilon_n^{(0)})\delta_{kk'}-\langle nk'|V|nk\rangle-\sum_{k'',n'\neq n}\langle nk'|V|n'k''\rangle\langle n'k''|V|nk\rangle/\omega(n-n')\right\|=0, \qquad (2)$$

where $|nk\rangle$ is an unperturbed wave function in the Landau gauge $\mathbf{A}=(0,Hx,0)$, k is the y component of the wave vector, $\omega=eH/m$, and $\varepsilon_n^{(0)}=\omega(n+\frac{1}{2})$ denotes a Landau level, which is φN-fold degenerate, with N the number of unit cells. A first-order splitting of Landau levels exists due to degeneracy. Moreover, the Fourier components $v(\mathbf{g})$ of the crystal field $V(\mathbf{r})$ do not act independently in a magnetic field. For example, using Eq. (2) one can obtain for the one-particle ground-state energy

$$\begin{aligned}\varepsilon_0(k)={}&\omega/2+2v\exp(-\pi/2\varphi)\cos(ak/\varphi)\\&-(2v^2/\omega)\exp(-\pi/\varphi)\{\mathrm{Ei}(\pi/\varphi)-\ln(\pi/\varphi)-C+[\mathrm{Ei}(-\pi/\varphi)-\ln(\pi/\varphi)-C]\cos(2ak/\varphi)\}.\end{aligned} \qquad (2')$$

From this expression one can see that for a totally filled lowest Landau level ($\varphi=\nu$) the first-order correction and the interference term of different Fourier components in the second order do not contribute to the total energy:

$$E(H)-E^{(0)}(0)=-(mNa^2v^2/\pi)\exp(-\pi/v)[\mathrm{Ei}(\pi/v)-\ln(\pi/v)-C]\,, \tag{3}$$

where $E^{(0)}(0)=\pi v^2N/ma^2$ is the energy of free electrons in zero field, $\mathrm{Ei}(x)$ is the integral exponent, $C=0.577216$, and the simplest form

$$V(\mathbf{r})=2v\cos(2\pi x/a) \tag{4}$$

is assumed, which has zero matrix elements between different degenerate states, with a the lattice constant, and $h=c=1$.

Thus, the total energy has a power-series expansion in v with the leading term second order in v, and different Fourier components act independently in leading order for a filled lowest Landau level, or for any integer number of filled Landau levels.

Equation (3) was obtained in a slightly different form by Nicopoulos and Trugman.[9] In the low-density limit, $v \ll 1$, which is the same as the short-wavelength limit since one Landau level is always filled, one can use the asymptotic representation of $\mathrm{Ei}(x)$:[10]

$$E(H)-E^{(0)}(0)=-(mNa^2v^2/\pi)\left[\sum_{n=1}^{N^*}(n-1)!\,(v/\pi)^n+R_{N^*}\right], \tag{5}$$

in the sense that the correction R_{N^*} is small,[11]

$$R_{N^*}<(\pi/2N^*)^{1/2}N^*!\,(v/\pi)^{N^*}\,. \tag{6}$$

For zero field one can integrate the second-order one-particle spectrum

$$\varepsilon(\mathbf{k})=k^2/2m-m\sum_{\mathbf{g}}|v(\mathbf{g})|^2/(\mathbf{k}\cdot\mathbf{g}+g^2/2) \tag{7}$$

up to $k<k_F$, where k_F is the Fermi wave vector, or use the expression for the static dielectric constant of two-dimensional free electrons[12] to obtain

$$E(0)-E^{(0)}(0)=-(mNa^2v^2/2\pi)\{1-\Theta(\pi/4v-1)(1-4v/\pi)^{1/2}\}\,,$$

or, at $v<\pi/4$,

$$E(0)-E^{(0)}(0)=-(mNa^2v^2/\pi)\sum_{n=1}^{\infty}(v/\pi)^n(2n-2)!/n!\,(n-1)!\,. \tag{8}$$

Thus one can see from Eqs. (5) and (8) that at $v \ll 1$ both energies coincide up to the fourth power of concentration:

$$\Delta E=E(H)-E(0)=-(mNa^2v^2/\pi)v^4/\pi^4\,. \tag{9}$$

To obtain the second-order energy $E^P(H)$ with the Peierls substitution one has to change $\mathbf{k}$ in Eq. (7) into $-i\nabla-e\mathbf{A}$ and calculate the diagonal matrix element of the second term, with the following result:

$$E^P(H)-E^{(0)}(0)=-(mNa^2v^2/\pi)(v/2)^{1/2}\,\mathrm{Im}\,w((\pi/2v)^{1/2})\,, \tag{10}$$

where $w(z)=\exp(-z^2)[1-\mathrm{erf}(-iz)]$ is an error function.[10] For $v \ll 1$ the Peierls substitution gives

$$E^P(H)-E^{(0)}(0)=-\frac{mNa^2v^2}{\pi}\left[\frac{v}{\pi}+\sum_{n=2}^{N^*}(2n-3)!!\left(\frac{v}{\pi}\right)^n+R'_{N^*}\right], \tag{11}$$

which differs from the exact expression, Eq. (5), starting from the term proportional to v^3 (see also Ref. 6). Thus

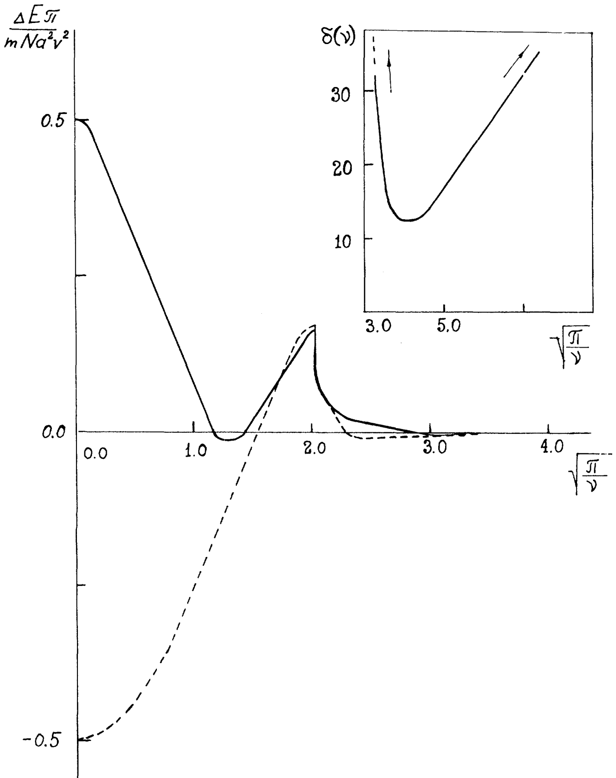

FIG. 1. Solid line: energy difference with ($\varphi=v$) and without the magnetic field (the difference between the solid and dotted curves in Ref. 9); dashed line: the same energy difference, calculated with the "Peierls substitution." Inset: The relative error of the "Peierls substitution" at low concentrations v.

the Peierls substitution overestimates the energy difference ΔE even in the region of low concentration, where it is usually assumed to be a good approximation. The relative error $\delta(\nu)$ of the Peierls substitution for ΔE is divergent at small ν:

$$\delta(\nu)=(\Delta E^P-\Delta E)/\Delta E=\pi/\nu\rightarrow\infty\,. \tag{12}$$

At large ν, the Peierls substitution, Eq. (10), gives the wrong sign of ΔE. For nearly free electrons the Peierls substitution turns out to be qualitatively (Fig. 1) and quantitatively (Fig. 1, inset) wrong.

To clarify the nature of the error we write the second-order correction in the form

$$\varepsilon_n^{(2)}=\mathrm{Re}\langle nk|V(\varepsilon_n^{(0)}-H+i\delta)^{-1}V|nk\rangle\,, \tag{13}$$

with $H=(-i\boldsymbol{\nabla}-e\mathbf{A})^2/2m$ and $\delta\rightarrow 0$. One might try to expand the resolvent $(\varepsilon_n^{(0)}-H)^{-1}$ in powers of the fluctuations of H. This expansion runs into problems, however, if matrix elements of V exist to states with $\varepsilon_n^{(0)}$. Ignoring this problem and simply substituting

$$(\varepsilon_n^{(0)}-H+i\delta)^{-1}=-(\langle H\rangle-\varepsilon_n^{(0)}-i\delta)^{-1}\sum_{n=0}^{\infty}\frac{(H-\langle H\rangle)^n}{(\langle H\rangle-\varepsilon_n^{(0)})^n}\,, \tag{14}$$

where $\langle H\rangle=\omega(n+\frac{1}{2})+g^2/2m$ is the energy in the state

$$|\mathbf{g}\rangle=\exp(i\mathbf{g}\cdot\mathbf{r})|nk\rangle\,,$$

one obtains a second-order energy correction $\tilde{\varepsilon}^{(2)}$,

$$\tilde{\varepsilon}_n^{(2)}=-m\mathrm{Re}\left(\sum_{\mathbf{g}}|y(\mathbf{g})|^2\langle nk|[g^2/2+\mathbf{g}\cdot(-i\boldsymbol{\nabla}-e\mathbf{A})]^{-1}|nk\rangle\right.$$
$$\left.+\sum_{\mathbf{g}'\neq\mathbf{g}}v(\mathbf{g})v(\mathbf{g}')^*\langle nk|\exp[i(\mathbf{g}-\mathbf{g}')\cdot\mathbf{r}][g^2/2+\mathbf{g}\cdot(-i\boldsymbol{\nabla}-e\mathbf{A})]^{-1}|nk\rangle\right)\,, \tag{15}$$

which differs from the correct $\varepsilon^{(2)}$. The first term in Eq. (15) is the second-order energy with the Peierls substitution and the second one is the interference of different Fourier components of a periodic potential, which does not contribute to the total energy in case of a filled Landau level. From this analysis it becomes clear that the Peierls substitution does not treat the problem correctly, which arises because of degeneracy. Furthermore, it uses an asymptotic expansion of the resolvent Eq. (14), which makes some sense only in the region of small fluctuations:

$$\langle\mathbf{g}|\{2m(H-\langle H\rangle)/g^2\}^n|\mathbf{g}\rangle<1\,,$$

which is the same as

$$\varphi/\pi<1\,.$$

For large flux $\varphi/\pi>1$, the Peierls substitution is meaningless.

At a low concentration, the correct energy difference ΔE, being negative, has an extremely low value, Eq. (9). Thus, any correction which varies with concentration slower than ν^4 will be dominant in this region. That is the case for a three-dimensional correction, which for a one-particle spectrum,

$$\varepsilon(\mathbf{k})=k_{\parallel}^2/2m+2t_{\perp}[1-\cos(k_{\perp}a_{\perp})]\,,$$

gives a positive energy difference that is concentration independent (see, for details, Ref. 13):

$$\Delta E_3=E_3^{(0)}(H)-E_3^{(0)}(0)=3mNa^2t_{\perp}^2/2\pi\,, \tag{16}$$

where $t_{\perp}$ and $a_{\perp}$ are the interplane hopping integral and the interplane distance, correspondingly. The superscript (0) means that electrons are free in the plane.

In the case of tight-binding electrons the atomic wave function at site $\mathbf{m}$ in a magnetic field can be expressed through a simple gauge transformation as

$$\psi_{\mathbf{m}}(\mathbf{r})=\exp(ie\chi_{\mathbf{m}})\psi_0(\mathbf{r}-\mathbf{m})\,, \tag{17}$$

where $\chi_{\mathbf{m}}=\frac{1}{2}\mathbf{H}\times\mathbf{m}\cdot\mathbf{r}$ and ψ_0 is the atomic orbital for $\mathbf{m}=0$ in a magnetic field, for which we now use the symmetric gauge $\mathbf{A}=\frac{1}{2}\mathbf{H}\times\mathbf{r}$. Replacing the atomic pseudopotential $V(\mathbf{r})$ by a harmonic-oscillator form,

$$V(\mathbf{r})=m\omega_0^2(x^2+y^2)/2\,,$$

with ω_0 the atomic level in zero field, one obtains from the Schrödinger equation

$$\{-\Delta/2m-(e/m)\mathbf{H}\cdot\mathbf{L}+e^2A^2/2m+V(\mathbf{r})\}\psi_0=\varepsilon(H)\psi_0\,,$$

$$\varepsilon(H)=(\omega_0^2+\omega^2/4)^{1/2}\,, \tag{18}$$

$$\psi_0(\mathbf{r})=\exp\{-(x^2+y^2)/2\lambda^2\}/\lambda\sqrt{\pi}\,, \tag{19}$$

where $\lambda^{-4}=a^{-4}+(4\lambda_H)^{-4}$, a_0 is the radius of the atomic orbital in zero field, $\lambda_H^{-2}=2\pi\varphi/a^2$, and $\mathbf{L}$ is the orbital angular momentum. The diamagnetic shift of the atomic level, Eq. (18), dominates, as was mentioned by Nicopoulos and Trugman.[9] Here, using the exact model solution, Eq. (19), we show that the reduction of the hopping integral will also raise the kinetic energy. With Eq. (19) one finds for the nearest neighbors $m_x'-m_x=a$, $m_y'-m_y=0$,

$$\tilde{t}=\int\exp\{i(e/2)[\mathbf{H}\times(\mathbf{m}-\mathbf{m}')\cdot\mathbf{r}\}\times\psi_0(\mathbf{r}-\mathbf{m})V\psi_0(\mathbf{r}-\mathbf{m}')d\mathbf{r}$$
$$=\sum_{\mathbf{g}}v(\mathbf{g})\exp[ig_xa/2-g_x^2\lambda^2/4-(g_y+\pi\varphi/a)^2\lambda^2/4-a^2/4\lambda^2]\,. \tag{20}$$

One can see from Eq. (20) that a strong magnetic field indeed gives exponential reduction of the hopping integral due to localization of atomic orbitals, Eq. (19). For the simple form of $V(\mathbf{r})$, Eq. (4), we obtain

$$\tilde{t}/t = \exp(-3a_0^2\pi^2\varphi^2/8a^2) \tag{21}$$

for $\pi\varphi < a^2/a_0^2$, and

$$\tilde{t}/t = \exp(-\pi\varphi/2) \tag{22}$$

for $\pi\varphi > a^2/a_0^2$.

In conclusion, we have shown that the kinetic energy of electrons in a magnetic field is higher than without a field, both for nearly free two-dimensional electrons in a wide range of their concentration and for tight-binding electrons. Even if in some narrow region of concentration (Fig. 1) ΔE is negative, this fact does not mean the violation of the Landau theory of orbital diamagnetism, which assumes that electrons are diamagnetic in the limit $H \to 0$. Both perturbation theory and the Peierls substitution give positive ΔE in this limit. The magnetic susceptibility in finite fields is a differential oscillating characteristic, which does not depend on the value or the sign of the total energy.

We point out that the Peierls substitution does not adequately describe the total energy with a magnetic field either in the nearly-free-electron case or in the tight-binding one.

Our conclusions have an indirect bearing on the occurrence of flux phases, proposed by Kalmeyer and Laughlin.[14] There, the ground-state wave function in Eq. (1) is used as a trial wave function, the field in Eq. (1) being fictitious and having no self-energy ($H^2/8\pi$ of a *real* field for $\varphi = \nu$ would be a factor of 10^5 higher than the electron energy). On the other hand, the claimed "lowering of the kinetic energy in a real field" has been used as support for the occurrence of these flux phases.[3-7] Our results show that this argument is unjustified, being an artifact of the Peierls substitution outside its range of validity.

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