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Nonlinear cyclotron resonance of a massless quasiparticle in graphene

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We consider the classical motion of a massless quasiparticle in a magnetic field and under a weak electromagnetic radiation with the frequency ω. Due to the nonparabolic linear energy dispersion, the particle responds not only at the frequency ω but generates a broad frequency spectrum around it. The linewidth of the cyclotron resonance turns out to be very broad even in a perfectly pure material which allows one to explain recent experimental data in graphene.

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Graphene is a new two-dimensional (2D) material[1,2] consisting of a monolayer of carbon atoms packed in a dense honeycomb lattice.[3,4] The band structure of electrons in graphene has two bands (electron and hole) which touch each other at six corners of the hexagon-shaped Brillouin zone.[5] If graphene is undoped and the temperature is zero, the lower (hole) band is fully occupied while the upper (electron) band is empty. The Fermi level goes through these six so-called Dirac points. Near these points the states of electrons and holes are described by the effective Dirac equation with the vanishing effective mass of the quasiparticles, and their energy spectrum is linear,

$$\mathcal{E}_{\pm}(\mathbf{p}) = \pm Vp = \pm V|\mathbf{p}|. \qquad (1)$$

Here $\mathbf{p}=(p_x,p_y)$ is the electronic momentum, counted from the corresponding Dirac points, and $V \approx 10^8$ cm/s is the material parameter. The linear (massless) energy dispersion [Eq. (1)] and the Dirac nature of graphene quasiparticles result in its amazing physical properties which attracted much attention in the past years.

Being placed in a magnetic field $\mathbf{B}$, a charged particle rotates in the perpendicular to $\mathbf{B}$ plane with the cyclotron frequency ω_c and absorbs the energy of an external electromagnetic wave if $\omega \approx \omega_c$, where ω is the radiation frequency [the cyclotron resonance (CR)]. The width of the CR absorption line $\delta\omega$ is usually determined by the scattering of electrons and by the radiative decay rate. In typical semiconductor 2D electron systems, e.g., in GaAs quantum wells, the linewidth of the CR is small as compared to the CR frequency already in magnetic fields on order of 0.1 T (see, for example, Ref. 6).

The magneto-optical response of graphene has been theoretically studied, within the linear-response approach, in a number of publications.[7–11] Intensive experimental studies of the CR in graphene (as well as other electrodynamic phenomena) have been hampered until recently by the absence of graphene flakes of sufficiently large area. With the progress of technology, however, graphene samples of sufficiently large dimensions are becoming available now,[12,13] so the growth of experimental activity on the graphene electrodynamics is to be expected. First experimental studies of the CR in single-layered graphene have been already published in Ref. 14 (the infrared spectroscopy) and Ref. 15 (the photoconductive response); for the CR data in bilayer graphene and in thin graphite layers see Refs. 16 and 17. In both CR experiments[14,15] the CR line was found to be extremely broad. Although the experiments have been done in very strong magnetic fields (up to 18 T), the quality factor of the CR line $B_c/\delta B_c \simeq \omega/\delta\omega$ has only slightly exceeded unity. Following the traditional interpretation of the CR linewidth one had to conclude that in Refs. 14 and 15 one has dealt with extremely disordered graphene samples. As will be shown below, however, the cyclotron resonance of the massless quasiparticles [Eq. (1)] has very unusual physical properties, and *the CR line can be very broad even in perfectly pure graphene*.

Consider the classical motion of a quasiparticle with spectrum (1) in a uniform magnetic field $\mathbf{B}=(0,0,B)$ and in the presence of an external electromagnetic wave. The equation of motion and the initial condition read as

$$\frac{d\mathbf{p}}{dt} = -\frac{e}{c}\mathbf{v}\times\mathbf{B} - e\mathbf{E}(t), \quad \mathbf{v} = V\frac{\mathbf{p}}{p}, \qquad (2)$$

$\mathbf{p}|_{t=0}=\mathbf{p}_0$. We assume that the radiation frequency ω coincides with the cyclotron frequency ω_c and that the external electric field $\mathbf{E}_0(t)=E_0(t)(\cos\omega t, \sin\omega t)$ is circularly polarized in the direction coinciding with the sense of the cyclotron rotation of the particle. The spectrum of the incident radiation $S_{\rm inc}(\Omega)$ thus consists of only one spectral line, $S_{\rm inc}(\Omega) \propto \delta(\Omega-\omega)$, and the particle is under the perfect CR conditions. For the sake of clarity we further neglect all the dissipative processes such as the scattering of electrons by impurities and phonons, as well as the radiative decay. We also assume that the external electric field E_0 is weak, $\mathcal{F} \equiv eE_0/\omega p_0 \ll 1$, meaning that the energy absorbed by the particle during one oscillation period is small as compared to its average energy. Usually this corresponds to the linear-response regime.

Figure 1(a) shows the results of the numerical solution of Eq. (2) for a standard (massive) particle (in this case $\mathbf{v}=\mathbf{p}/m$ and $\omega_c=eB/mc$). Under the chosen conditions, the massive particle is *always* in resonance with the external field, it continuously absorbs the radiation energy, and the absolute value of its momentum p linearly grows with time. Rotating with the frequency ω the massive particle re-emits (scatters) the radiation of the incident wave at the same frequency ω, and the spectrum of the scattered waves $S_{\rm scat}(\Omega)$ is also proportional to $\delta(\Omega-\omega)$.

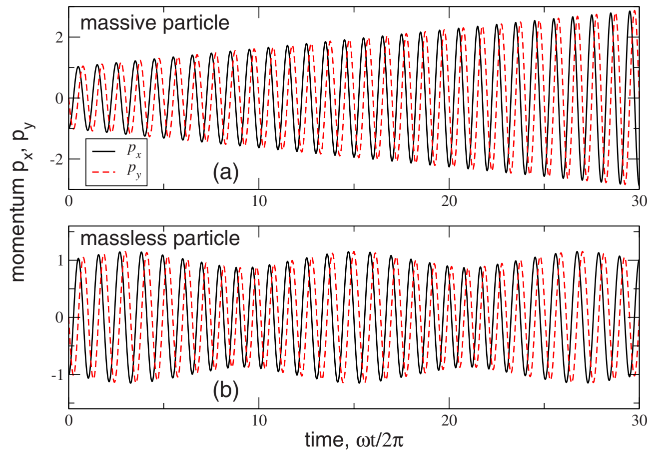

FIG. 1. (Color online) Time dependencies of the momentum of (a) the massive and (b) the massless quasiparticles under the CR conditions. The electric-field parameter is $\mathcal{F}$=0.01. Notice the difference in the vertical axis scale on plots (a) and (b).

The motion of the massless particle is essentially different.[18] Its cyclotron frequency $\omega_c(\mathcal{E})=eBV/pc=eBV^2/\mathcal{E}c$ depends on its energy. Initially, when the external field is switched on, the particle is also in resonance with the external radiation, $\omega=\omega_c=eBV/p_0c$ and starts to get energy from the wave. But, as soon as its energy increases, $p>p_0$, it gets out of the resonance and ceases to absorb the radiation energy. Its energy $\mathcal{E}=Vp$ and the absolute value of the momentum p decreases, and the particle again turns out to be in the resonance with the wave. Then the process repeats itself [Fig. 1(b)], and the particle energy oscillates in time with the period depending on the external field amplitude E_0. The time dependence of the momentum of the massless particle is *not a harmonic function* with only one Fourier component $\Omega=\omega$. It also contains other (higher and lower) harmonics.

We introduce, instead of $p_x(t)$ and $p_y(t)$, variables $p(t)$ and $\phi(t)$ according to the formula

$$\begin{pmatrix} p_x(t) \\ p_y(t) \end{pmatrix} = p(t)\begin{pmatrix} -\sin[\omega t+\phi(t)] \\ \cos[\omega t+\phi(t)] \end{pmatrix} \tag{3}$$

and investigate the time dependencies of the momentum $p(t)$ and the phase $\phi(t)$. Equation (2) is then rewritten as

$$\dot{p}(t) = eE_0 \sin \phi(t), \tag{4}$$

$$p(t)\dot{\phi}(t) = -\omega p(t) + eVB/c + eE_0 \cos \phi(t), \tag{5}$$

and the initial conditions are $p|_{t=0}=p_0$ and $\phi|_{t=0}=\phi_0$. Figures 2(a) and 2(b) show the time dependencies of $p(t)$ and $\phi(t)$ at different values of the electric-field parameter $\mathcal{F}=eE_0/\omega p_0$. Both functions are modulated, and the modulation frequency, as well as the amplitude of the momentum oscillations, decreases with $\mathcal{F}$. In contrast, the amplitude of the phase oscillations remains independent of the electric field E_0 [Fig. 2(b)].

The behavior of the $p(t)$ and $\phi(t)$ oscillations also depends on the initial phase ϕ_0 [Figs. 2(c) and 2(d)]. One sees that not only the oscillation period and amplitude may depend on ϕ_0 but even the overall shape of oscillations; see the case $\phi_0=\pi$ [the green dash-dotted curve in Figs. 2(c) and 2(d)]. The linear contribution to the phase [Fig. 2(d)] means the frequency shift since $\phi(t)=-\alpha t+\tilde{\phi}(t)$ leads to $\omega t+\phi(t)=(\omega-\alpha)t+\tilde{\phi}(t)$.

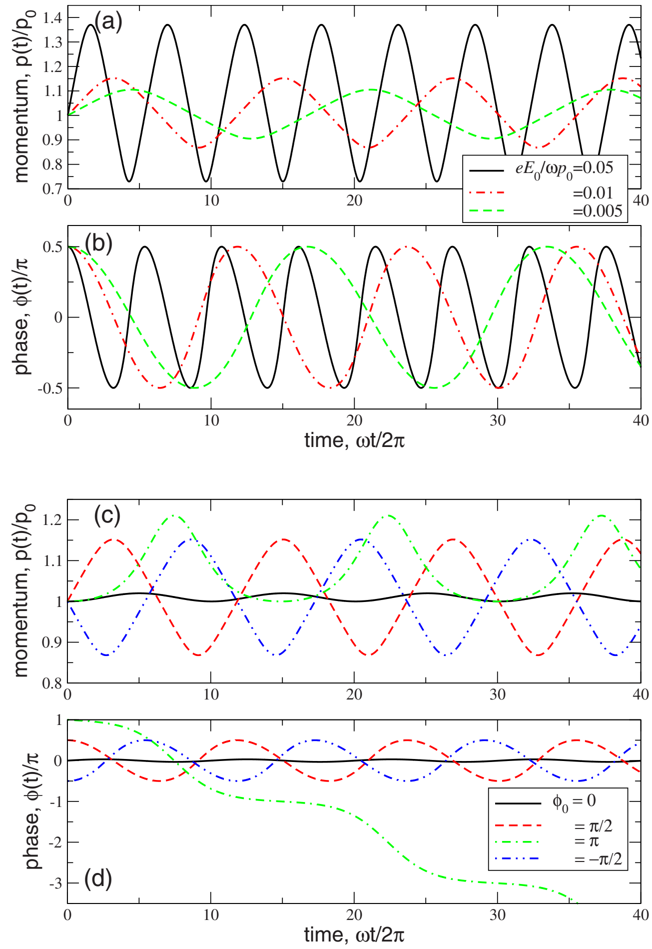

FIG. 2. (Color online) Time dependencies of the momentum $p(t)$ and the phase $\phi(t)$ at (a) and (b) different values of the electric field and $\phi_0=\pi/2$ and (c) and (d) under different initial conditions $\phi|_{t=0}=\phi_0$ and at $\mathcal{F}$=0.01.

So far we discussed the behavior of the momentum of the massless quasiparticle. The experimentally measured value, however, is the velocity

$$\mathbf{v}(t) = V\frac{\mathbf{p}(t)}{p(t)} = V\begin{pmatrix} -\sin[\omega t+\phi(t)] \\ \cos[\omega t+\phi(t)] \end{pmatrix}. \tag{6}$$

Its absolute value $|\mathbf{v}(t)|=V$ remains constant, independent of how big or small the particle momentum is. As the phase $\phi(t)$ remains to be modulated even in low fields [Fig. 2(b)], the Fourier transformation of $\mathbf{v}(t)$ shows a broad spectrum around the central frequency $\Omega=\omega$ at $\mathcal{F}\ll 1$ (Fig. 3). Since the velocity of particles determines the current and the amplitudes of the scattered electromagnetic waves, Fig. 3 shows, in fact, the Fourier spectrum of the scattered waves $S_{scat}(\Omega)$. We emphasize that we discuss the cyclotron resonance of the massless quasiparticle *in the absence of any scattering*.

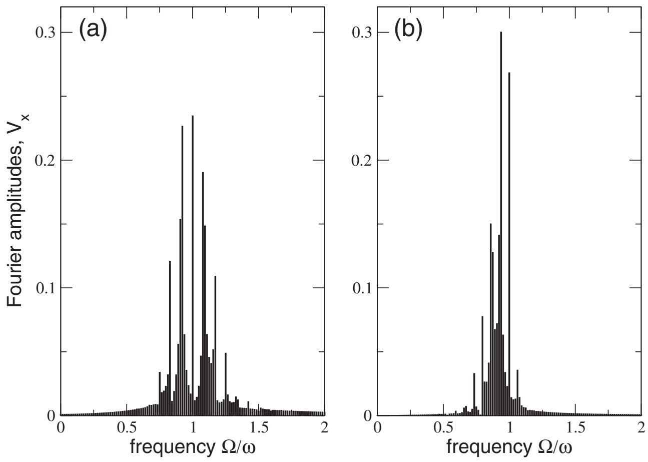

FIG. 3. Frequency spectra of the velocity v_x for $\mathcal{F}=eE_0/\omega p_0=0.01$ and (a) $\phi_0=\pi/2$ and (b) $\phi_0=\pi$.

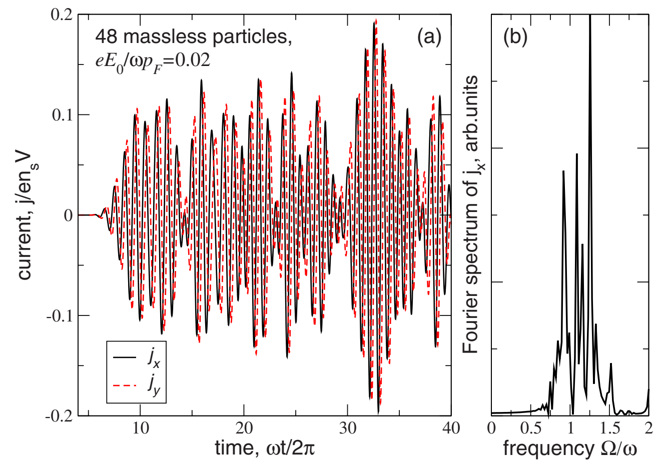

FIG. 4. (Color online) (a) Time dependence of the current in a system of 48 massless particles. It is assumed that the particles are Fermi distributed in the **p** space at $t=0$ and that the electric field is zero at $\omega t/2\pi<5$ and smoothly grows up to a constant value at $5<\omega t/2\pi<10$. (b) Fourier spectrum of the j_x component of the current shown in (a).

Under certain conditions the phase $\phi(t)$ and the spectrum $S_{scat}(\Omega)$ can be found analytically. If $\mathcal{F}\ll 1$ we substitute $p(t)=p_0[1+q(t)]$ with $|q(t)|\ll 1$ and rewrite Eqs. (4) and (5) as

$$\dot{q}(t)=\omega\mathcal{F}\sin\phi,\quad \dot{\phi}(t)=-\omega q+\omega\mathcal{F}\cos\phi. \tag{7}$$

Assuming that at $\mathcal{F}\ll 1$ the last term in the second equation [Eq. (7)] can be neglected as compared to the first one (this is confirmed by the result [Eq. (9)]) we reduce problem (7) to the nonlinear pendulum equation,

$$\ddot{\phi}(t)=-\omega^2\mathcal{F}\sin\phi, \tag{8}$$

well known in the nonlinear physics.[19] If the initial phase ϕ_0 is small, $\phi_0\lesssim 1$, we get

$$\phi(t)=\phi_0\cos(\omega\sqrt{\mathcal{F}}t),\quad q(t)=\phi_0\sqrt{\mathcal{F}}\sin(\omega\sqrt{\mathcal{F}}t),\quad \phi_0\lesssim 1. \tag{9}$$

The amplitude and the phase of the momentum oscillations are thus modulated in this case with the frequency $\omega\sqrt{\mathcal{F}}$ proportional to the square root of the electric field.

Substituting the phase $\phi(t)$ from Eq. (9) to Eq. (6) one can calculate the Fourier spectrum of the velocity $\tilde{\mathbf{v}}(\Omega)=\int\mathbf{v}(t)e^{-i\Omega t}dt/2\pi$. For example, for the v_y component we get

$$\tilde{v}_y(\Omega)=\frac{V}{2}\sum_{k=-\infty}^{\infty}i^kJ_k(\phi_0)\delta(\Omega-\omega-k\omega\sqrt{\mathcal{F}})+\frac{V}{2}\sum_{k=-\infty}^{\infty}(-i)^kJ_k(\phi_0)\delta(\Omega+\omega-k\omega\sqrt{\mathcal{F}}), \tag{10}$$

where J_k are Bessel functions. The spectrum $S_{scat}(\Omega)$ consists of an infinite number of satellite harmonics at the frequencies $\Omega=\omega\pm k\omega\sqrt{\mathcal{F}}$ with $k=1,2,\ldots$. The amplitudes of these harmonics $J_k(\phi_0)$ are determined only by the initial phase of the particle and do not depend on the electric field.

So far we discussed the response of only one quasiparticle and found that it is complicated and very different even for particles with the same p_0 but different initial phases ϕ_0. In a real graphene system one deals with many particles with different initial phases and initial momenta p_0 having the Fermi distribution. The electromagnetic response of the whole many-particle system will be a superposition of partial responses of all the individual particles and will therefore be very complicated. The time dependence of the current [see example in Fig. 4(a)] looks very chaotic, and the scattered wave spectrum $S_{scat}(\Omega)$ [Fig. 4(b)] consists of many narrow peaks distributed around $\Omega\sim\omega$. In systems with strong disorder, such as in currently available graphene samples, each of these narrow lines will be broadened, which would lead to a single broad peak. With the improvement of the graphene samples' quality in future the multiline structure of the broad CR lines should become apparent.

Our results show that *the linear-response theory is not applicable to pure graphene in finite magnetic fields*. Linearizing the classical or quantum kinetic equations, one straightforwardly gets the linear current-vs-field dependence and hence the scattered wave spectrum $S_{scat}(\Omega)$ directly proportional to the incident wave spectrum $S_{inc}(\Omega)$. This is not the case in graphene; therefore, in general, the methods of the nonlinear dynamics and the theory of chaos should be used in this material. Mathematically, the nonanalyticity of the graphene response originates from the singular Lorentz-force term in equation of motion (2). This singularity disappears at $B=0$; therefore, in zero magnetic field the linear-response theory is valid (cf. Refs. 20 and 21).

The spectrum of Landau levels of a *massive* particle is equidistant, $E_N=\hbar\omega_c(N+1/2)$. If at initial instant such a particle is on the Nth Landau-level, photons with the frequency $\omega=\omega_c=eB/mc$ will cause consecutive transitions $N\to(N+1)\to(N+2)\to\cdots$, which corresponds to the continuous growth of the classical orbit radius [Fig. 1(a)]. In contrast, the Landau-level spectrum of a *massless* quasiparticle in graphene is nonequidistant,[22] $E_N=\pm V\sqrt{2\hbar eB|N|/c}$. In the quasiclassical limit $N\gg 1$ the energy of the inter-Landau-level transitions corresponds to the classical energy-dependent cyclotron frequency,

$$E_{N+1} - E_N \approx \hbar \frac{eBV^2}{cE_N} = \hbar \omega_c(E_N), \quad N \gg 1. \tag{11}$$

The photons with the frequency $\omega = \omega_c(E_N)$ are now in resonance with only two energy levels and will cause Rabi oscillations $N \leftrightarrow (N+1)$.[23] Transitions to the higher-energy levels $N+2$, $N+3$, etc., will be hampered by the frequency detuning but will be possible in stronger electric fields. The occupation numbers of the Landau levels, and hence of the induced current, will therefore oscillate with a frequency dependent on the electric field [cf. Figs. 1(b) and 2]. This qualitative quantum-mechanical consideration also shows that the response of a massless particle should be nonmonochromatic, with the electric-field-dependent CR linewidth and agrees with our quasiclassical results.

The nonlinear effects in the cyclotron resonance are also known in the relativistic physics of ordinary particles, e.g., electrons.[18,24] But the nonlinear CR in graphene substantially differs from that of relativistic electrons. The response of real electrons is always linear in weak electric fields, while in graphene, due to the vanishing effective mass of its quasiparticles, the area of applicability of the linear-response theory shrinks down to zero (in finite magnetic fields). Numerous nonlinear effects known from the high-energy plasma physics[19] should therefore be possible to observe in pure graphene under easily accessible conditions.

To summarize, we have shown that graphene is essentially nonlinear material. Being irradiated by the monochromatic wave, graphene scatters the radiation in a broad frequency range. As a consequence, the CR line in graphene turns out to be very broad even in the absence of scattering, which may explain the broad CR lines observed in the experiments.[14,15]

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[1] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, M. I. Katsnelson, I. V. Grigorieva, S. V. Dubonos, and A. A. Firsov, Nature (London) **438**, 197 (2005).
[2] Y. Zhang, Y.-W. Tan, H. L. Stormer, and P. Kim, Nature (London) **438**, 201 (2005).
[3] M. I. Katsnelson, Mater. Today **10**, 20 (2007).
[4] A. K. Geim and K. S. Novoselov, Nature Mater. **6**, 183 (2007).
[5] P. R. Wallace, Phys. Rev. **71**, 622 (1947).
[6] I. V. Kukushkin, V. M. Muravev, J. H. Smet, M. Hauser, W. Dietsche, and K. von Klitzing, Phys. Rev. B **73**, 113310 (2006).
[7] N. M. R. Peres, F. Guinea, and A. H. Castro Neto, Phys. Rev. B **73**, 125411 (2006).
[8] V. P. Gusynin and S. G. Sharapov, Phys. Rev. B **73**, 245411 (2006).
[9] V. P. Gusynin, S. G. Sharapov, and J. P. Carbotte, Phys. Rev. Lett. **98**, 157402 (2007).
[10] Y. Barlas, R. Côté, K. Nomura, and A. H. MacDonald, Phys. Rev. Lett. **101**, 097601 (2008).
[11] T. Morimoto, Y. Hatsugai, and H. Aoki, Phys. Rev. B **78**, 073406 (2008).
[12] R. R. Nair, P. Blake, A. N. Grigorenko, K. S. Novoselov, T. J. Booth, T. Stauber, N. M. R. Peres, and A. K. Geim, Science **320**, 1308 (2008).
[13] K. S. Kim, Y. Zhao, H. Jang, S. Y. Lee, J. M. Kim, K. S. Kim, J.-H. Ahn, P. Kim, J.-Y. Choi, and B. H. Hong, Nature (London) **457**, 706 (2009).
[14] Z. Jiang, E. A. Henriksen, L. C. Tung, Y.-J. Wang, M. E. Schwartz, M. Y. Han, P. Kim, and H. L. Stormer, Phys. Rev. Lett. **98**, 197403 (2007).
[15] R. S. Deacon, K.-C. Chuang, R. J. Nicholas, K. S. Novoselov, and A. K. Geim, Phys. Rev. B **76**, 081406(R) (2007).
[16] E. A. Henriksen, Z. Jiang, L. C. Tung, M. E. Schwartz, M. Takita, Y.-J. Wang, P. Kim, and H. L. Stormer, Phys. Rev. Lett. **100**, 087403 (2008).
[17] M. L. Sadowski, G. Martinez, M. Potemski, C. Berger, and W. A. de Heer, Phys. Rev. Lett. **97**, 266405 (2006).
[18] C. S. Roberts and S. J. Bushbaum, Phys. Rev. **135**, A381 (1964).
[19] R. Z. Sagdeev and G. M. Zaslavsky, *Nonlinear Physics: From the Pendulum to Turbulence and Chaos*, 4th ed. (CRC Press, Boca Raton, FL, 1988).
[20] S. A. Mikhailov, Europhys. Lett. **79**, 27002 (2007).
[21] S. A. Mikhailov and K. Ziegler, J. Phys.: Condens. Matter **20**, 384204 (2008).
[22] Y. Zheng and T. Ando, Phys. Rev. B **65**, 245420 (2002).
[23] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics (Nonrelativistic Theory)* (Elsevier, Oxford, 1994), problem to Sec. 40.
[24] A. E. Kaplan, Phys. Rev. Lett. **48**, 138 (1982).