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Nonlinear optical absorption in graphene via two-photon absorption process

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ABSTRACT

Nonlinear optical absorption phenomena in monolayer graphene for scattering between carriers and *K*-optical phonons are studied via investigating the phonon-assisted cyclotron resonance (PACR) effect. The positions of the PACR peaks are indicated. Only principal transitions make a significant contribution to the optical absorption power. We found that with the increase of the magnetic field, the optical absorption spectrum increases in magnitude and also gives the blue-shift in both one- and two-photon absorption processes. Besides, the half-width is found to be almost independent from the change of temperature, but proportional to the square root of magnetic field, and this agrees with the previous experimental observations.

1. Introduction

Graphene is a single sheet of carbon atoms arranged in a honey comb lattice. Having been fabricated [1] and experimentally demonstrated to be a zero-gap system with massless Dirac particles [2,3], graphene has been expected to be a source of exotic systems and new physics because of its fascinating physical properties [4]. Graphene has been taken into great consideration not only because of its potential for applications in nano-electronic devices in the future [5,6], but also because of its exceptional electronic properties. One of its fascinating physical properties results from the unusual Landau levels (LLs) in a perpendicular quantizing magnetic field. It is known that in traditional 2D-materials, the LLs are proportional to a non-negative integer and equally spaced. However, in graphene the LLs are not equally spaced in energy but proportional to both the square root of the magnetic field and the LL index [7–11]. This unusual LL structure of graphene has been measured indirectly by investigating the transitions between LLs through the use of cyclotron resonance [12–17]. In addition, recently, Pound et al. [18] have proposed a new formula for discrete states in graphene. Their results showed that the phonon-assisted electron transitions between different LLs with phonon emission and absorption can be displayed via a new set of peaks in the density of states. Although these transitions involved optical phonons, they did not describe the electron–phonon interaction. It is known that in graphene, the electron–optical phonon interaction plays an important role to understand the electron transitions between different LLs [19–21], to understand the absorption or emission of optical phonons by electrons [22], and to cause an increase to the electromagnetic absorption [23]. Based on the development of the theory of the fine structure of PACR in graphene, Zhu et al. [24] have demonstrated that even for small electron–phonon coupling, the PACR is governed by the peaks resulting from the electron–phonon interaction, which establish an asymmetric doublet around the optical phonon frequency.

On the other hand, for the half-width (half width at half maximum – HWHM), Jiang et al. [25] have experimentally measured and found that the half-width at $T=4.2$ K appeared to be increasing with the increase of the magnetic field B, but its B-dependence could not be further appraised in their works. Besides, Li et al. found that the line width (full-width at half maximum) increased linearly with energy [26], this implies that the half-widths were found to be proportional to the square root of magnetic field. In another work, besides finding that both the peak position and the area showed a linear increase with the square root of the magnetic field, Orlita et al. [27] also experimentally measured that the width increased nearly linearly with $\sqrt{B}$ in the range of weak magnetic field while in stronger magnetic fields, it increased nonlinearly with the square root of the magnetic field. Moreover, in this work, the authors also proved that in graphene, the thermal activation is not an important scattering mechanism due to the slightly temperature-dependent half-width. However, the results

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mentioned above have been obtained only with one-photon process while the two-photon one has not been done.

In this work, we focus on optical absorption power and half-width in graphene via one- and two-photon absorption processes through the use of the PACR effect. Our results are appropriately compared to the recent experimental and some earlier theoretical results. The paper is organized as follows: In Section 2, the optical absorption power describing the PACR effect and resonant conditions are presented. The numerical results are reported and discussed in Section 3. Finally, a conclusion is given in Section 4.

2. Optical absorption power in graphene

We consider a configuration where a graphene sheet is in the (xy) plane and the uniform static magnetic field with strength B is applied along the z-direction. The normalized wave function and the corresponding energy for a carrier (electron and hole) in the Landau gauge for the vector potential $\mathbf{A}=(Bx, 0)$ are written as [10,19,28]

$$|\alpha\rangle = \psi_{nX}(\mathbf{r}) = \frac{C_n}{\sqrt{L}} e^{-iyX/a_c^2} \begin{pmatrix} S_n \phi_{|n|-1}(x-X) \\ \phi_{|n|}(x-X) \end{pmatrix}, \tag{1}$$

$$\varepsilon_\alpha = \varepsilon_n = S_n \hbar\omega_c \sqrt{|n|}, \quad n = 0, \pm 1, \pm 2, \ldots, \tag{2}$$

where the electronic states for a carrier are specified by the set of quantum numbers $\alpha=(n, X)$ with n being the LL index and $X=k_y a_c^2$ being the coordinate of the center of the carrier orbit; $\mathbf{r}=(x, y)$ is the 2D spatial coordinate; $C_n=[(1+\delta_{n,0})/2]^{1/2}$; L is the linear dimension of the system; $a_c=(\hbar/eB)^{1/2}$ is the magnetic length; and $\hbar\omega_c=\gamma\sqrt{2}/a_c$ is the effective magnetic energy with $\gamma=a\gamma_0\sqrt{3}/2$ being the band parameter, and a being the lattice constant. Besides, $S_n=+1$ and -1 stand for the conduction and valance bands, respectively. We neglect the Zeeman spin-splitting in the present study because of its significant weakness. In Eq. (1), $\phi_{|n|}(x)$ is the normalized harmonic-oscillator function, which is given as

$$\phi_{|n|}(x) = \frac{i^{|n|}}{\sqrt{2^{|n|}|n|!\sqrt{\pi}a_c}} e^{-x^2/2a_c^2} H_{|n|}\left(\frac{x}{a_c}\right), \tag{3}$$

where $H_n(x)$ is the n-th Hermite polynomial.

Using perturbation approximation methods, the optical absorption power can be calculated by relating it to the transition probability of the absorption of photons with frequency Ω as follows [29,30]:

$$P(\Omega) = \frac{F_0^2\sqrt{\varepsilon}}{8\pi} \sum_\alpha W_{\alpha,\alpha'} f_\alpha, \tag{4}$$

where F_0 is the intensity radiation; ε is the dielectric constant of the medium; $f_\alpha \equiv f_n$ is the Fermi–Dirac function; and $W_{\alpha,\alpha'}$ is the transition probability due to carrier–photon–phonon interaction in graphene, which is approximated by Fermi's golden rule

$$W_{\alpha,\alpha'} = \frac{2\pi}{\hbar} g_s g_v \sum_{\alpha'} \sum_{\mathbf{q},\mu} |V(\mathbf{q})|^2 g(\theta) |J_{nn'}(q)|^2 \Delta(\varepsilon_n, \varepsilon_{n'}), \tag{5}$$

where $g_v=2$ and $g_s=2$ are the valley and spin degeneracy, respectively; $\mathbf{q}=(q_x, q_y)$ is the phonon wave vector; μ denotes the phonon mode; $g(\theta)=(1+\cos\theta)/2$ is the overlap integral of spinor wave function; and

$$|V(\mathbf{q})|^2 = \frac{\hbar D_{op}^2}{2\rho L^2 \omega_\mu}, \tag{6}$$

$$|J_{nn'}(q)|^2 = C_n^2 C_{n'}^2 \frac{m!}{(m+j)!} e^{-u} u^j \left[L_m^j(u) + S_n S_{n'} \sqrt{\frac{m+j}{m}} L_{m-1}^j(u) \right]^2, \tag{7}$$

where D_{op} is the deformation potential constants, ρ is the areal mass density [31], $u=a_c^2 q^2/2$, $m=\min(|n|, |n'|)$, $j=||n'|-|n||$, and $L_m^j(u)$ being the associated Laguerre polynomials [28]. The quantity $\Delta(\varepsilon_n, \varepsilon_{n'})$ in Eq. (5) in the two-photon absorption process $(l=1, 2)$ is given by [29,32–34]

$$\Delta(\varepsilon_n, \varepsilon_{n'}) = \frac{a_0^2 q^2}{4} [N_\mu \delta(Y_1^-) + (N_\mu+1)\delta(Y_1^+)]$$

$$+ \frac{a_0^4 q^4}{64} [N_\mu \delta(Y_2^-) + (N_\mu+1)\delta(Y_2^+)], \tag{8}$$

where a_0 is the dressing parameter, N_μ is the distribution function of phonons with frequency ω_μ, and we have denoted

$$Y_l^\pm = \varepsilon_{n'} - \varepsilon_n \pm \hbar\omega_\mu - l\hbar\Omega \quad (l=1, 2). \tag{9}$$

Although there are four optical phonon modes in graphene, we will only consider K-optical phonon interaction ($\mu=K$) because the resonant behavior resulting from K- and Γ-optical phonons are similar [10]. For high optical phonon energy and small occupation number of optical phonons (N_K â^a_i 1), we neglect phonon absorption in comparison with emission. Substituting Eq. (5) into Eq. (4) and reducing, we have

$$P(\Omega) = \frac{F_0^2 D_{op}^2 a_0^2 \sqrt{\varepsilon}}{32\pi\rho\omega_K} (N_K+1) \sum_{n,n'} C_n^2 C_{n'}^2 f_n \int_0^\infty q^3\, dq |J_{nn'}(u)|^2$$

$$\times \left[\delta(\varepsilon_{n'} - \varepsilon_n + \hbar\omega_K - \hbar\Omega) + \frac{a_0^2 q^2}{16} \delta(\varepsilon_{n'} - \varepsilon_n + \hbar\omega_K - 2\hbar\Omega) \right]. \tag{10}$$

Using Eq. (7) and straightforwardly integrating over q (see Appendix (A.1)–(A.4), we obtain the following expression of the optical absorption power in graphene:

$$P(\Omega) = \frac{F_0^2 D_{op}^2 a_0^2 \sqrt{\varepsilon}}{8\pi\rho\omega_K a_c^4} (N_K+1) \sum_{n,n'} C_n^2 C_{n'}^2 f_n [A_1 \delta(\varepsilon_{n'} - \varepsilon_n + \hbar\omega_K - \hbar\Omega)$$

$$+ A_2 \delta(\varepsilon_{n'} - \varepsilon_n + \hbar\omega_K - 2\hbar\Omega)], \tag{11}$$

where A_1 and A_2 are dimensionless quantities defined as

$$A_1 = 2m + j - S_n S_{n'} \sqrt{m(m+j)}, \tag{12}$$

$$A_2 = \frac{a_0^2}{8a_c^2} [2 + 6m^2 + j(j+6m) - 2S_n S_{n'} (2m+j)\sqrt{m(m+j)}]. \tag{13}$$

The delta functions in Eq. (11) imply the energy conservation law, which show the PACR condition in graphene from the selection rule

$$l\hbar\Omega = \varepsilon_{n'} - \varepsilon_n + \hbar\omega_K \quad (l=1, 2), \tag{14}$$

where $l=1$ and 2 correspond to the one- (linear) and two-photon (nonlinear) absorption processes, respectively. In the absence of an electromagnetic field, $\Omega=0$, this condition is reduced to the magneto-phonon resonance: $\varepsilon_n - \varepsilon_{n'} = \hbar\omega_K$ [18,19]. Because of the stranger LLs in graphene, there is no general selection rule for