VNU Journal of Science: Mathematics - Physics

Journal homepage: https://js.vnu.edu.vn/MaP

Original Article

The Influence of an External Magnetic Field on Multi-photon Non-linear Absorption Coefficient of a Strong Electromagnetic Wave in Two-dimensional Graphene

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Received 6 May 2023
Revised 17 January 2024; Accepted 30 January 2024

Abstract: Based on the quantum kinetic equation for electrons, we theoretically studied the Quantum Multi-photon Nonlinear Absorption of a Strong Electromagnetic Wave (EMW) in two-dimensional graphene with electron-optical phonon scattering mechanism. The general multi-photon absorption coefficient was presented as a function of the temperature, the external magnetic field, the photon energy, and the amplitude of external EMW. The results show that in the presence of the magnetic field, absorption spectral lines appear consistent with the magneto-phonon resonance conditions. In which, the effect of multi-photon absorption is stronger than that of mono-photon absorption. Besides, the quantum multi-photon nonlinear absorption phenomenon has been studied from low to temperatures. This transcends the limits of the classical Boltzmann kinetic equation which is studied in the high-temperature domain. The computational results show that the dependence of Multi-photon Nonlinear Absorption Coefficient (MNAC) on the above quantities is consistent with the previous theoretical investigation. Another novel feature of this work is that the general analytic expression for MNAC shows the Half Width at Half Maximum (HWHM) dependence on the magnetic field which is in good agreement with the previous experimental observations. Thus, our estimation might give a critical prediction for future experimental observations in graphene.

Keywords: Multi-photon non-linear absorption coefficient, 2D graphene, quantum kinetic equation, strong electromagnetic wave, electron-phonon scattering, magneto-phonon resonance.

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https//doi.org/10.25073/2588-1124/vnumap.4849

1. Introduction

It has been almost 20 years since Geim and Novoselov discovered a method to separate graphene monolayers with office tape [1]. Since then, there have been many studies on the superior properties of graphene compared with other traditional semiconductors. In particular, the energy band structure of graphene exhibit zero band gap, and the electron mobility is very high, thereby opening up many new research directions, both theoretically and experimentally. Theoretically, Sarma et al. have studied the electron mobility in graphene with the electron-acoustic phonon scattering mechanism by Boltzmann transport equation [2], Ando et al., have studied magneto-phonon resonance using the Kubo formula [3, 4]. Experimentally, Tiras et al., investigated the 2D electron energy relaxations, carrier density, and electron in-plane effective mass of electrons by Shubnikov-de Haas (SdH) and Hall effect measurements [5].

The quantum nonlinear absorption effect which is one of the most significant research topics in low-dimensional electron gas systems has been studied in both theoretical and experimental aspects [6-8] by using different techniques. For graphene, many different research methods have been used to investigate the absorption effect. Hoi et al., [9] found that the explicit expression of optical absorption power in 2D Graphene under a perpendicular magnetic field and proved that the half width at half maximum of cyclotron-phonon resonance does not depend on the temperature by using the projection operator technique. Phuc and Hieu [10] presented the nonlinear optical absorption power in the presence of the perpendicular magnetic field with electron-optical phonon interaction picture via a two-photon absorption process using the perturbation approximation method. When surveying the effect of the EMW on the absorption power for a unit surface, Kryuchkov et al., [11] pointed out that in the large amplitudes of circularly polarized EMW, the absorption power is proportional to the EMW's amplitude within the full self-consistent relaxation time approximation. In all these works, the optical power has been investigated taking into account only electron-optical phonon interaction in the high-temperature range, while the electron-acoustic phonon in the low-temperature range has not been concerned. Moreover, the previous works also did not mention the influence of linearly polarized EMW on the MNAC via the multi-photon absorption processes (MPA).

In this work, we presented our calculation of the nonlinear absorption coefficient of an intense EMW in 2D graphene in which both electron-optical phonon scattering and electron-acoustic phonon scattering with the MPA process have been investigated by using the quantum kinetic equation method. This approach allows us to take into account the rate of change in unbalanced electron distribution function, and thus, shows the electron-phonon interaction picture clearly under the influence of a laser radiation field. This is the reason why this method has been used effectively in the study of EMW absorption effects in bulk semiconductors [8], Quantum wells [12], and Cylindrical quantum wires [13]. In the next section, we give the theoretical basis and basic calculations. Numerical results and discussion are presented in Sec. 3. Finally, concluding remarks are given briefly in Sec. 4.

2. Theoretical Calculations

We considered a graphite sheet where electrons move freely in the (x, y) plane. The system is subjected to a magnetic field $\mathbf{B} = (0, 0, B)$ with a vector potential $\mathbf{A'} = (Bx, 0, 0)$. Energy levels in strong magnetic field B are quantized into discrete energy levels, called Landau levels or Landau orbits. The wave function and the corresponding energy are written as [3]:

$$\Psi(\mathbf{r}) \equiv \left|n, \mathbf{k}_y\right\rangle = \frac{C_n}{\sqrt{L}} \exp\left(-i\frac{Xy}{l_B^2}\right)\begin{bmatrix} S_n \Phi_{|n|-1}(x-X) \\ \Phi_{|n|}(x-X) \end{bmatrix} = \frac{C_n}{\sqrt{L_y}} e^{ik_y y}\begin{bmatrix} S_n \Phi_{|n|-1}(x-X) \\ \Phi_{|n|}(x-X) \end{bmatrix} \tag{1}$$

$$\varepsilon_n = S_n \hbar\omega_B \sqrt{|n|} \tag{2}$$

with S_n is the sign function defined by $S_n = -1$ for an electron when $n<0, S_n = 1$ for a hole when $n>0$, and $S_n = 0$ for $n=0; C_n^2 = (1+\delta_{n,0})/2$ where δ_{ij} is the Kronecker Delta, $\delta_{ij}=1$ when $i=j$ and $\delta_{ij}=0$ when $i\neq j$. In Eq. (18) $\Phi_{|n|}(x)$ is the normalized harmonic oscillator function given by

$$\Phi_{|n|}(x) = \frac{i^{|n|}}{\sqrt{2^{|n|}|n|!\sqrt{\pi} l_B}} \exp\left[-\frac{1}{2}\left(\frac{x}{l_B}\right)^2\right] H_{|n|}\left(\frac{x}{l_B}\right) \tag{3}$$

where $n = 0, \pm1, \pm2, \ldots$ being the Landau indices. X being the coordinate of the center of the carrier orbit $X = k_y l_B^2$ with $l_B = \sqrt{\hbar/eB}$ is the radius of the ground state electron orbit in the (x, y) plane or the magnetic length. $\mathrm{H}_{|n|}(x/l_B)$ is the n-th order Hermite polynomial and $\hbar\omega_B = \sqrt{2}\gamma/l_B$ is the effective magnetic energy [3, 4] with $\gamma = \left(\sqrt{3}/2\right)a\gamma_0 = 6,46\text{eV.Å}$ is the band parameter, $\gamma_0 = 3,03\text{eV}$ is the resonance integral between nearest neighbor carbon atoms, $a = 0,246$ nm is the lattice constant.

When a strong EMW is applied to the system with the electric field vector $\mathbf{E} = (0, E_0 \sin\Omega t, 0)$ (E_0 and Ω are the amplitude and frequency, respectively), the Hamiltonian of the electron-phonon system in the second quantization representation can be written as [3, 10]

$$\begin{aligned} \mathrm{H} &= \sum_{\mathrm{n},\mathbf{k}_y} \varepsilon_{\mathrm{n}}\left(\mathbf{k}_{\mathrm{y}} - \frac{\mathrm{e}}{\hbar \mathrm{c}}\mathbf{A}(\mathrm{t})\right) \mathrm{a}^{\dagger}_{\mathrm{n},\mathbf{k}_y} a_{\mathrm{n},\mathbf{k}_y} + \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}}\left(b^{\dagger}_{\mathbf{q}} \mathrm{b}_{\mathbf{q}} + \frac{1}{2}\right) \\ &+ \sum_{\mathrm{n},\mathrm{n}'} \sum_{\mathbf{k}_y,\mathbf{q}} \mathrm{M}_{\mathrm{n},\mathrm{n}'}(\mathbf{q}) a^{\dagger}_{\mathrm{n}',\mathbf{k}_y+\mathbf{q}_y} a_{n,\mathbf{k}_y}\left(b^{\dagger}_{-\mathbf{q}} + b_{\mathbf{q}}\right) \end{aligned} \tag{4}$$

Here, $a^{\dagger}_{n,\mathbf{k}_y}$ and $a_{n,\mathbf{k}_y}$ ($b^{\dagger}_{\mathbf{q}}$ and $b_{\mathbf{q}}$) are the creation and annihilation operators of electron (phonon), respectively; $\hbar\omega_{\mathbf{q}}$ is the phonon energy, n and n' are the band indices of states $|\mathbf{k}\rangle$ and $|\mathbf{k}+\mathbf{q}\rangle$, respectively. The vector potential of laser radiation as a strong EMW $\mathbf{A}(t)$ takes the for

$$\mathbf{A}(t) = \frac{c}{\Omega}\mathbf{E}_0 \cos(\Omega t), \tag{5}$$

with $M_{n.n'}(\mathbf{q})$ (the matrix factor) as follows [10]

$$\left|\mathrm{M}_{\mathrm{n},\mathrm{n}'}(\mathbf{q})\right|^2 = |\mathrm{C}(\mathbf{q})|^2 \left|J_{\mathrm{n},\mathrm{n}'}(\mathbf{q})\right|^2 \tag{6}$$

with $\mathrm{C}(\mathbf{q})$ is the electron-phonon interaction constant which depends on the scattering mechanism $J_{n,n'}(\mathbf{q})$ given by [10]

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

with $L_m^j(x)$ is the associated Laguerre polynomial, $u = l_B^2 q^2/2, q^2 = q_x^2 + q_y^2$, $\mathrm{m}=\min(|n|,|n'|), j = \|n'|-|n\|$.

The general quantum kinetic equation for electron distribution function [8]

$$\frac{\partial f_{n,\mathbf{k}_y}(t)}{\partial t} = -\frac{i}{\hbar}\left[a^{\dagger}_{n,\mathbf{k}_y} a_{n,\mathbf{k}_y}, \mathrm{H}\right]_t \tag{8}$$

where $f_{n,\mathbf{k}_y}(t) = \left\langle a^{\dagger}_{n,\mathbf{k}_y} a_{n,\mathbf{k}_y} \right\rangle_t$ is the electron distribution function, and $\langle \ldots \rangle_t$ denotes the statistical average value at the moment t.

To investigate the optical absorption properties in the graphene sheet in the presence of an EMW, we need to have an analytic expression of the non-linear absorption coefficient [8]

$$\alpha = \frac{8\pi}{c\sqrt{\kappa_\infty} E_0^2} \left\langle \mathbf{J}_y \cdot \mathbf{E} \right\rangle_t \tag{9}$$

where κ_∞ is the high-frequency dielectric constant in graphene and c is the speed of light in a vacuum.

In Eq. (9), $\mathbf{J}_y(t)$ is the carrier current density in 2D graphene, which flows in the y-direction because the laser field is polarized in the y-direction. Its expression is given as [8]

$$\mathbf{J}_y(t) = \frac{e\hbar}{m_e} \sum_{n,\mathbf{k}_y} \left(\mathbf{k}_y - \frac{e}{\hbar c} \mathbf{A}(t) \right) f_{n,\mathbf{k}_y}(t) \tag{10}$$

Here, e and $m_e = 0.012 m_0$ [5] are the charges and the effective mass of electron in graphene, m_0 being the electron rest mass.

Solving the quantum kinetic equation, which is established for electrons in graphene for the case of an external magnetic field, using the same calculation methods as previous works [8, 12, 13], we obtain the ℓ -photon absorption coefficient

$$\alpha = \frac{8\pi^2 \Omega}{c\sqrt{\kappa_\infty} E_0^2} \sum_{\mathbf{q},n',n} \left| M_{n,n'}(\mathbf{q}) \right|^2 \overline{N_{\mathbf{q}}} \sum_{\ell} \ell J_\ell^2 \left(\frac{eE_0 q}{2m_e \Omega^2} \right) \sum_{\mathbf{k}_y} \overline{f}(\varepsilon_n) \delta\left(\varepsilon_{n'} - \varepsilon_n + \hbar\omega_{\mathbf{q}} - \ell\ddot{y}\Omega \right) \tag{11}$$

Here, $J_s(x)$ is the s th-order Bessel function of the argument x, $\delta(x)$ is the Dirac delta function. We assume that the distribution function of electrons in thermal equilibrium is the Fermi-Dirac distribution $\overline{f}(\varepsilon_n) = \left\{1 + \exp\left[(\varepsilon_n - \varepsilon_F)/(k_B T)\right]\right\}^{-1}$, in which, ε_F is Fermi energy, k_B is the Boltzmann constant, and T is the absolute temperature of the system. $\overline{N_{\mathbf{q}}}$ is the equilibrium distribution function for phonons, which is given by the Bose-Einstein distribution function $\overline{N_{\mathbf{q}}} = \left[\exp\left(\hbar\omega_{\mathbf{q}} / k_B T\right) - 1\right]^{-1}$.

In the next step, we will consider two scattering mechanisms of electrons and phonons to elucidate the physical properties involved.

2.1. Electron-optical Phonon Scattering

At the high temperature $(T > 50\ \mathrm{K})$, we assume that the dispersion of phonon follows and the electron-optical phonon interaction constant is determined by [9-11]

$$|\mathrm{C}(\mathbf{q})|^2 = \frac{\hbar^2 D_{op}^2}{2\rho L^2 (\hbar\omega_0)} \tag{12}$$

with D_{op} and ρ are deformed potential of optical phonon and two-dimensional mass density of graphene sheet.

Transforming the summations over $\mathbf{q}$ and $\mathbf{k}_y$ to integrals as follows [9, 14] and using the approximate expression of the Bessel function, we obtain the absorption coefficient for the case of electron-optical phonon interaction

$$\alpha_{op} = \frac{4\pi D_{op}^2 \hbar\Omega \overline{\mathrm{N}_{\mathbf{q}}}}{c\sqrt{\kappa_\infty} E_0^2 \rho l_B^2 \omega_0} \sum_\ell \left(\frac{eE_0}{2m_e\Omega^2} \right)^{2\ell} \frac{1}{\ell[\Gamma(\ell)]^2} \sum_{n,n'} \overline{f}(\varepsilon_n) \delta(\varepsilon_{n'} - \varepsilon_n + \hbar\omega_0 - \ell\ddot{y}\Omega) \int_0^{+\infty} q^{2\ell+1} \left| \mathrm{J}_{n,n'}(u) \right|^2 dq \tag{13}$$

with $\Gamma(\ell)$ is the Gamma function for the positive integer argument ℓ.

Using the integrals in the appendix and analytic transformations, we obtain an explicit expression of the optical absorption coefficient in the case of electron-optical phonon scattering

$$\alpha_{op} = \frac{4\pi n_0 D_{op}^2 \hbar\Omega \overline{\mathrm{N}_{\mathbf{q}}}}{c\sqrt{\chi_\infty} E_0^2 \rho l_B^2 \omega_0} \sum_{n',n} \overline{f}(\varepsilon_n) \sum_\ell A_\ell \delta(\varepsilon_{n'} - \varepsilon_n + \hbar\omega_0 - \ell\ddot{y}\Omega) \tag{14}$$

A_ℓ is the dimensionless parameter characterizing ℓ-photon absorption process.

For mono-photon absorption (1PA)

$$A_1 = \left(\frac{eE_0}{4m_e\Omega^2} \right)^2 \frac{2C_n^2 C_{n'}^2}{l_B^4} \left[2m + j + 1 - 2S_n S_{n'} \sqrt{m(m+j)} + S_n^2 S_{n'}^2 (2m + j - 1) \right] \tag{15}$$

For two-photon absorption (2PA)

$$A_2 = \left(\frac{eE_0}{4m_e\Omega^2} \right)^4 \frac{2C_n^2 C_{n'}^2}{l_B^6} \left\{ 2 + 6m(m+1) + j\left[j + 3(2m+1) \right] - 4S_n S_{n'} (2m + j) \sqrt{m(m+j)} + \right.$$
$$\left. + S_n^2 S_{n'}^2 \left\{ 2 + 6m(m-1) + j\left[j + 3(2m-1) \right] \right\} \right\} \tag{16}$$

For three-photon absorption (3PA)

$$A_3 = \left(\frac{eE_0}{4m_e\Omega^2} \right)^6 \frac{2C_n^2 C_{n'}^2}{3l_B^8} \{ (2m + j + 3) \left\{ 2 + 6m(m+1) + j\left[j + 3(2m+1) \right] \right\} + 4m(2m + j)(m + j) +$$
$$+ S_n^2 S_{n'}^2 \left\{ (2m + j + 1) \left\{ 2 + 6m(m-1) + j\left[j + 3(2m-1) \right] \right\} + 4(m-1)(2m + j - 2)(m + j) \right\} -$$
$$- 6S_n S_{n'} \left(5m^2 + 5mj + j^2 + 1 \right) \sqrt{m(m+j)} \Big\} \tag{17}$$

Expression (25) shows the extremely complex dependence of the optical absorption coefficient on the parameters related to the external field. This is due to the presence of the factor $J_{n',n}(u)$, which represents the influence of the magnetic field.

2.2. Electron-acoustic Phonon Scattering

For this scattering mechanism, we just need to do the same calculation steps as in the case of electron-optical phonon scattering and use Eq. (11). However, there is some variation in the matrix element formula as follows [2, 11, 14]

$$|C(\boldsymbol{q})|^2 = \frac{\hbar D_{ac}^2 q}{2\rho v_s L^2} \tag{18}$$

here, v_s and D_{ac} are the sound velocity in graphene and the magnitude of the deformation potential energy, respectively. Moreover, we assume that the scattering between electron and acoustic phonon is quasi-elastic, hence acoustic phonon energy $\hbar\omega_{\boldsymbol{q}} = \hbar v_s q$ is ignored in the Dirac delta function [15]. Considering the case of a non-degenerate phonon system, the equilibrium distribution function of the acoustic phonon has the form [2] $\overline{\mathrm{N}_{\mathbf{q}}} \approx k_B T / \hbar\omega_{\boldsymbol{q}}$.

By the same calculation as above, we can establish the analytic expression of the general multi-photon non-linear absorption coefficient in the acoustic phonon-electron scattering mechanism

$$\alpha_{ac} = \frac{4\pi n_0 k_B T D_{ac}^2 \Omega}{c\sqrt{\chi_\infty} E_0^2 \rho l_B^2 v_s^2} \sum_{n',n} \overline{f}(\varepsilon_n) \sum_{\ell} A_\ell \delta(\varepsilon_{n'} - \varepsilon_n - \ell\ddot{y}\Omega) \tag{19}$$

A remarkable point in Eq. (14) and Eq. (19) is that the Dirac delta functions will be divergent if their argument approaches zero. To eliminate divergence, we replace the delta functions with the Lorentzian functions as follows [15]

$$\delta(\varepsilon) = \frac{1}{\pi} \frac{\Gamma}{\varepsilon^2 + \Gamma^2} \tag{20}$$

with Γ is the dimensionless parameter characterizing the scattering strength, its expression is given [9, 15]

$$\Gamma^2 = \sum_{\boldsymbol{q}} \left(\overline{N_{\boldsymbol{q}}} + \frac{1}{2} \pm \frac{1}{2} \right) |M(\boldsymbol{q})|^2 \tag{21}$$

It can be seen that the analytic expressions of the general MNAC in the presence of a magnetic field are extremely complex. In the next section, we will give more insight into this difference through numerical computation and graphing with the help of computer programs and numerical methods.

3. Results and Discussion

In this section, we detail the numerical evaluation of MNAC in the presence of a strong magnetic field for two electron-phonon scattering mechanisms, from low temperature to high-temperature domain. The parameters used in computational calculations are as follows [2-5, 9-11, 14]: $\gamma = 6.46\text{eV}.\text{Þ}, \rho = 7.7\times10^{-8}\ \text{g/cm}^2, n_0 = 5\times10^{15}\ \text{m}^{-2}, \mathrm{D_{op}} = 1.4\times10^{9}\ \text{eV/cm}, \mathrm{D_{ac}} = 19\text{eV},$ $\mathrm{v_s} = 2\times10^{6}\ \text{cm/s}, \hbar\omega_0 = 162\text{meV}, \kappa_\infty = 4$. The value of the Fermi energy level in the Fermi - Dirac distribution function can be approximated between the Landau levels $\mathrm{n} = 0$ and electrons [16]. In other words, $\varepsilon_F = \hbar\omega_B / 2$, with $\hbar\omega_B$ is the effective magnetic energy from Eq. (19). In this paper, we consider the Landau levels from -4 to 4.

Figures 1 and 2 indicate the MNAC as a function of the effective magnetic energy (proportional to the square root of magnetic field B) at 2 different values of the photon energy. As can be seen from these figures, we see the appearance of absorption spectral lines. The absorption coefficient is only significant at these resonance peak positions. The appearance of resonance peaks can be explained by cyclotron-phonon resonance conditions $\hbar\omega_B\left(S_{n'}\sqrt{|n'|} - S_n\sqrt{|n|}\right) + \hbar\omega_0 - \ell\ddot{y}\Omega = 0$. In addition, the density of

absorption peaks becomes sparse as the effective magnetic energy increases. This can be explained by the influence of the strong magnetic field causing the absorption spectrum to be interrupted in the region where the effective magnetic energy is greater than the photon energy of an EMW.

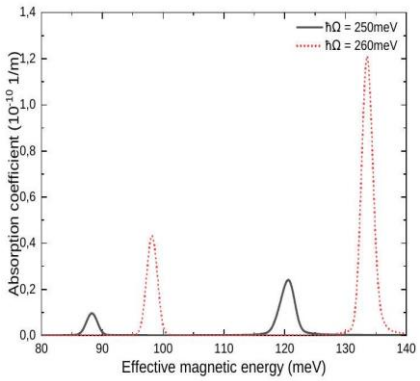

Figure 1. The dependence of MNAC on the magnetic energy (electron - optical phonon interaction). Here, $E_0 = 5\times10^5$ V / m, T = 100 K.

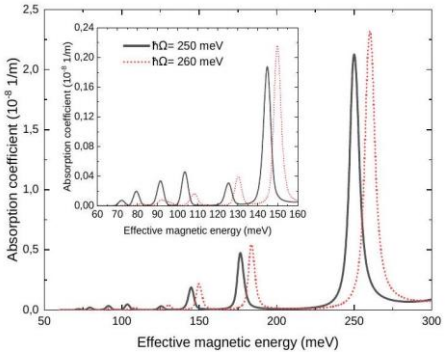

Figure 2. The dependence of MNAC on the magnetic energy (electron - acoustic phonon interaction). Here, $E_0 = 5\times10^5$ V / m, T = 20 K.

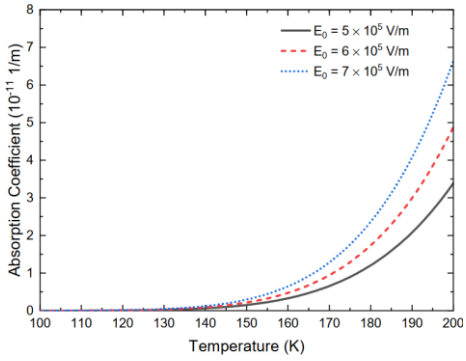

Figure 3. The dependence of MNAC on the temperature of system (electron - optical phonon interaction). Here, ħΩ = 250 meV, B = 6 T.

In Figure 3, the dependence of MNAC on the temperature of the system with three different values of the electromagnetic field strength is monotonically increasing. This is quite similar to the results obtained in a previous work [17] in the absence of strong magnetic fields. However, the presence of a magnetic field increases the absorption coefficient significantly. This can be explained by the presence of a magnetic field that forms a parabolic confinement potential where the electron motion is quantized on the Oz axis, resulting in the scattering probability and the Landau level shift between electrons, phonons and photons increased significantly, strongly affecting the multi-photon absorption coefficient in graphene.

4. Conclusion

In summary, we have theoretically investigated the Quantum multi-photon Non-linear Absorption Coefficient in monolayer graphene with two scattering mechanisms based on the quantum kinetic equation method. The new theoretical expressions for the absorption coefficient in graphene are established as functions of the external field, the photon energy of EMW, the temperature of the system, and the magnetic field. The phenomenon of electromagnetic wave absorption in graphene has been studied in detail from the low-temperature domain (the limitation of classical theories) to the high-temperature domain, giving results in good agreement with classical theories such as the Boltzmann kinetic equation. In the presence of a strong magnetic field, the absorption spectrum of electromagnetic waves is interrupted. Resonance peaks appear according to cyclotron-phonon resonance conditions. The position of the resonance peaks depends only on the magnetic field and not on the temperature of the system. The contribution of processes that absorb more than one photon is shown more clearly than with other methods. The influence of a strong magnetic field interrupts the absorption spectrum in the highly effective magnetic energy domain. This result can be used for for checking the accuracy of the data obtained in the fabrication of graphene-related electronic devices.

Acknowledgments

This research is financially supported by Vietnam National University, Hanoi - Grant number QG.22.11.

Appendix: The integrals used in the calculation

From Eqs. A1, A2, A3, and A4 of Ref. [10], we have

$$I_1 = \frac{m!}{(m+j)!}\int_0^{+\infty} e^{-u}u^{j+3}\left[L_m^j(u)\right]^2 du$$

$$= (2m+j+3)\left\{2+6m(m+1)+j\left[j+3(2m+1)\right]\right\}+4m(2m+j)(m+j)$$

$$I_2 = \frac{(m-1)!}{(m+j)!}\int_0^{+\infty} e^{-u}u^{j+3}L_m^j(u)L_{m-1}^j(u)du = -3\left(1+j^2+5mj+5m^2\right)$$

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