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Multiphonon resonance Raman scattering in Landau-quantized graphene

Zi-Wu Wang*, Zhi-Qing Li

Tianjin Key Laboratory of Low Dimensional Materials Physics and Preparing Technology, Department of Physics, Tianjin University, Tianjin, 300354, China

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ABSTRACT

We theoretically investigate multiphonon resonance Raman scattering between the Landau levels in graphene on the polar substrate using the Huang-Rhys's model. We not only present the single and multiple surface optical (SO) phonons scattering, but also propose the combined multiphonon scattering, which is composed of the SO phonon and longitudinal acoustic phonon. We find that the combined multiphonon scattering has a blue-shift behavior with increasing the magnetic field, differing from these SO phonon resonance scattering occurred at specific magnetic field values. This behavior may be used to explain the changing shoulder of the Raman spectrum of optical phonon resonance scattering in experiments. The theoretical model could be expanded to analyze the fine structure of Raman spectrum in two-dimensional materials.

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1. Introduction

The non-equidistant Landau level (LL) separation is an unique property of graphene when the perpendicular magnetic field is presented, which displays a number of interesting optical properties [1,2], such as the selection rule of the emission of polarized light [3,4], a giant Faraday rotation [5]and generation of entangled photon states [6]. In particular, the LL separation can be tuned from the infrared to terahertz regimes by manipulating the external magnetic field, which may be used as the infrared and terahertz sources for many electric devices [7–12]. Therefore, the studies of the non-radiative relaxation channels between LLs, competing with the optical transitions processes, are of crucial for the exploration of optical properties in the Landau-quantized graphene (LQG).

Two types of non-radiation channels were mainly considered until now. One is the Auger scattering, which prevails at high carriers concentration and has been proved in experiments [13,14]. Another is the different optical phonons scattering, such as the longitudinal optical (LO) phonons [3,15], the surface optical (SO) phonons induced by the supporting substrates [16] and the out-of-plane phonon at the Γ point [17], which plays a dominant role when the LL's separation matching the optical phonon energy. This is known as the magnetophonon resonance effect, which has been identified widely in experiments by the magneto-exciton transition [18], magneto-optical conductivity [19] and magneto-Raman scattering [15,20,21]. In particular, the SO phonon mode induced by the supporting substrate has aroused more interesting in the structure of graphene/polar substrate due to the charge carriers in graphene coupling strongly with the SO phonon [20,22,23]. However, theory predicted that this type of channel will be strongly suppressed as the optical phonon energy mismatches the LL's separation, which is also called the relaxation bottleneck effect. In order to break this effect, we have proposed the combined two-phonon scattering composed of one optical phonon and one acoustic phonon in previous work [24,25]. Its relaxation time is fast and very close to the Auger scattering. Auger scattering has been extensively proved in experiments, but the progress of this combined two-phonon scattering is very slow due to the lack of the proper platform and effective ways.

In the present paper, we theoretically propose the Raman scattering with multiphonon processes in LQG based on the Huang-Rhys's model [26–28]. We present the single SO phonon and the combined two-phonon scattering ($\hbar\omega_{SO} + \hbar\omega_{LA}$), in which $\hbar\omega_{SO}$ and $\hbar\omega_{LA}$ are the energy of one SO phonon and one longitudinal acoustic (LA) phonon modes, respectively, as well as two-SO phonon ($2\hbar\omega_{SO}$) and the combined three-phonon scattering ($2\hbar\omega_{SO} + \hbar\omega_{LA}$). We study the intensity of Raman scattering and present the modulations of the intensity of the scattering by the external magnetic

* Corresponding author.
E-mail address: wangziwu@tju.edu.cn (Z.-W. Wang).

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filed, temperature, polarizability of substrate and internal distance between graphene and substrate. Our theoretical results indicate that the LQG provides a good platform to identify these multi-phonon scattering, especially for the combined two-phonon scattering in experiments.

2. Theoretical model

We consider that a uniform magnetic field **B** is perpendicular to the monolayer graphene sheet laying on the polar substrate. In the frame of symmetric gauge for **B**, the energies are quantized into Landau levels schemed in Fig. 1 and the corresponding eigenfunction and eigenenergy of the single free carrier can be solved analytically (see the supplementary materials).

In general, Raman scattering involves electronic excitations as intermediate states. In LQG, the Landau level can be acted as the intermediate state for the resonance Raman scattering. The electromagnetic field of the incident and scattered photon interact primarily with these Landau levels, and emission of phonons occurs due to electron-phonon interaction. Based on the Huang-Rhys's model [26–28], the cross section of multiphonon Raman scattering can be schematically represented as

$$R_n \propto \left| \sum_{j,k,\nu} \frac{\mu_{ij}(\omega_i) A_n \left| \left\langle \psi_j \middle| \left\langle \chi_j(Q_{j,\nu}) \middle| H_{ep}^{\nu} \middle| \chi_k(Q_{k,\nu}) \right\rangle \middle| \psi_k \right\rangle \middle| \mu_{ki}(\omega_s) \right|}{[\hbar\omega_i - (E_j - E_i) - i\Upsilon][\hbar\omega_i - \hbar\omega_s - n\hbar\omega_\nu - i\Upsilon]} \right|^2, \tag{1}$$

with

$$\mu_{ij}(\omega_i) = \frac{\hbar e^2 V_F^2}{2\varepsilon_0 \omega_i} \begin{cases} \dfrac{1}{2} & (j=0 \quad or \quad i=0) \\ \dfrac{1}{4} & (j\neq 0 \quad and \quad i\neq 0) \end{cases}, \tag{2}$$

which describes the transition process between LLs for the incident photon following the optical selection rule [3,4,6], in which e is the carrier charge, V_F is the Fermi velocity, ε_0 is the permittivity of vacuum and ω_i is the frequency of the incident photon. The scattering process $\mu_{ki}(\omega_s)$ has the similar form. The detail approximation for $\mu_{ij}(\omega_i)$ and $\mu_{ki}(\omega_s)$ are given in supplementary materials. ψ_j (ψ_k) and E_j (E_i) is the eigenfunction and eigenvalue of LL,

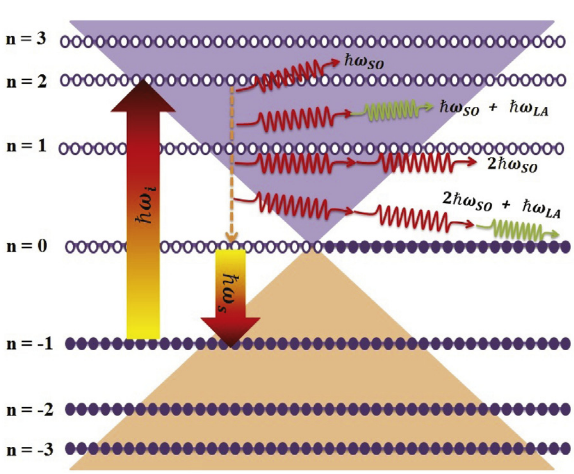

Fig. 1. The schematic diagrams for the distributions of the Landau levels in graphene and a possible channel of Raman scattering with different multiphonon resonance processes. $\hbar\omega_i$ and $\hbar\omega_s$ stand for the incident photon and scattered photon, respectively. (A colour version of this figure can be viewed online.)

respectively. $\chi_j(Q_{j,\nu})$ is the harmonic oscillation describing the lattice vibration with the equilibrium position $Q_{j,\nu}$. n is the scattering phonon number and $\hbar\omega_\nu$ is the phonon energy with the mode ν (ν = SO,LA), Υ is the broadening factor. In this paper, the linear electron-SO phonons and -LA phonons couplings H_{ep}^{ν} are taken into account and can be given

$$H_{ep}^{\nu} = \sum_q M_\nu(q) e^{iq\cdot r} Q_{\nu,q} = \sum_q \mathscr{M}^{\nu}(q\cdot r) Q_{\nu,q}, \tag{3}$$

with the coupling element [19,29]

$$M_{SO_\lambda}(q) = \sqrt{\frac{e^2\hbar\omega_{SO,\lambda}}{2\mathbb{A}\varepsilon_0}\left(\frac{1}{\kappa_0+1} - \frac{1}{\kappa_\infty+1}\right)} e^{-qz_0},$$

for SO phonon mode including two branches ($\lambda = 1, 2$) and

$$M_{LA}(q) = i\sqrt{\frac{\hbar D^2 q^2}{\mathbb{A}\rho\omega_{LA}}},$$

for LA phonon mode in the deformation potential mechanism [30], in which $\mathbb{A}$ is the area of the monolayer graphene, k_∞ (k_0) is the high (low) frequency dielectric constant, z_0 is the internal distance between the graphene and the substrate, ρ is the mass density, D denotes the deformation potential constant, ω_{LA} is the LA phonon frequency. In the Franck-Condon approximation, the term of the phonon scattering in Eq. (1) can be rewritten as

$$\begin{aligned} & A_n \sum_\nu |\langle\psi_j|\langle\chi_j(Q_{j,\nu})|H_{ep}^{\nu}|\chi_k(Q_{k,\nu})\rangle|\psi_k\rangle|^2 \approx A_n \\ & \sum_{\nu,q} |\langle\psi_j|\mathscr{M}^{\nu}(q\cdot r)|\psi_k\rangle|^2 |\langle\chi_j(Q_{j,\nu})|\chi_k(Q_{k,\nu})\rangle|^2 \\ & = \left|\mathscr{M}_{jk}^{SO} + \mathscr{M}_{jk}^{LA}\right|^2 \quad \times A_n \sum_\nu |\langle\chi_j(Q_\nu + \Delta_{j,\nu})|\chi_k(Q_\nu + \Delta_{k,\nu})\rangle|^2, \end{aligned} \tag{4}$$

where $\mathscr{M}_{jk}^{\nu}$ is the transition matrix between the intermediate state j and k, $\Delta_{j,\nu} = \langle\psi_j|\mathscr{M}^{\nu}(q\cdot r)|\psi_j\rangle$ denotes the shift of the equilibrium position of lattice vibration induced by the electron state ψ_j. In general, multiphonon Raman scattering was calculated based on the complicated high-orders perturbation process, which is replaced by the simple overlap integral of the lattice vibration in the Huang-Rhys's model. Calculating the thermal average over (A_n) the initial states j and summing over the final states k, Eq. (4) can be converted into

$$\begin{aligned} & \left|\mathscr{M}_{jk}^{SO} + \mathscr{M}_{jk}^{LA}\right|^2 \left[\frac{N_{SO}+1}{N_{SO}}\right]^{\frac{P}{2}} \left[\frac{N_{LA}+1}{N_{LA}}\right]^{\frac{1}{2}} \\ & \times \exp[-F_{SO}(2N_{SO}+1)] I_P \left[2F_{SO}\sqrt{N_{SO}(N_{SO}+1)}\right] \\ & \times \exp[-F_{LA}(2N_{LA}+1)] I_1 \left[2F_{LA}\sqrt{N_{LA}(N_{LA}+1)}\right], \end{aligned} \tag{5}$$

where $N_\nu = 1/[\exp(\hbar\omega_\nu/K_B T) - 1]$ is the Bose occupation function of phonon number, $F_\nu = \sum_q (\omega_{\nu,q}/2\hbar)\Delta_{jk,\nu}^2$ is the Huang-Rhys factor standing for the strength of the lattice relaxation and $\Delta_{jk,\nu} = \Delta_{j,\nu} - \Delta_{k,\nu}$ describes the shift of the lattice oscillator position before and after the transition, I_P and I^1 are the modified Bessel functions, $P = 1, 2, 3\cdots$ denotes the number of the scattering SO phonons. Throughout this paper, we assume that the broadening factor Υ = 5 meV, the deformation potential constant $D = 30$ eV for the LA phonon with the linear dispersion $\omega_{LA} = c\ q$ ($c = 7.6\times 10^3 m/s$)[30]. The SO phonon mode has a single frequency and the adopted parameters for the SO phonon energies and polar

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