Vibrational Spectroscopy 87 (2016) 193–198

Contents lists available at ScienceDirect

Vibrational Spectroscopy

journal homepage: www.elsevier.com/locate/vibspec

Electronic excitations in narrow quantum wells via intersubband Raman scattering: Theoretical considerations☆

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ARTICLE INFO

Article history:
Received 10 May 2016
Received in revised form
12 September 2016
Accepted 12 September 2016
Available online 17 October 2016

Keywords:
Raman scattering
Electron gas
Collective excitations
Single-particle excitations
Narrow quantum wells

ABSTRACT

In this work a generalized self-consistent field theory was applied to investigate the elementary excitations of two-dimensional electron gas formed from narrow quantum wells via resonant intersubband Raman scattering. The developed model considers the existence of equally coupled and degenerated excitations of the electron gas and allows to observe that in extreme resonance regime the plasma oscillations splits into two contributions: a set of renormalized collective excitations (plasmons) and unrenormalized electronic transitions (single-particle excitations). Our results show that the asymmetries which appear in the Raman profile of doped narrow quantum wells can be interpreted as the entrance or exit of resonance of collective modes overlapped with single-particle transitions.

1. Introduction

The inelastic light scattering is a powerful tool that has been widely used in the study of the semiconductor materials and it has become an indispensable technique for the understanding of fundamental physical processes [1–3]. The investigation of quantized electronic systems as quantum wells, wires and quantum dots via Raman spectroscopy is quite attractive since it allows to obtain information concerning many-body effects of interacting particles.

By means of electronic Raman scattering is possible to grasp the nature of collective excitations in quasi-2D systems which are known as Charge Density Excitations (CDE) and Spin Density Excitations (SDE). CDE are plasmonic oscillations arising from the coupling between charge fluctuations via Coulombian and exchange-correlation interactions while SDE occurs only when exchange-correlation effects are present. Both depend on selection rules that are associated with polarizations of the light [4]. The CDE is active when the laser energy is resonant with a semiconductor optical gap and the incoming and outgoing light polarizations are parallel to each other (polarized spectra). The SDE is active when the laser energy is resonant with a semiconductor optical gap and the incoming and outgoing light polarizations are perpendicular to each other (depolarized spectra). Nevertheless, when the laser matches interband transitions energy of the material (extreme resonance regime), in addition to the collective excitations, emerges transitions of the electron gas noninteracting-like known as Single-Particle Excitations (SPE). It is largely accepted that the observation of the SPE are related to extreme resonance regime irrespective of the dimensionality of the electron gas system [4–6], which makes then a fascinating phenomenon. However, the physics of such transitions is still not completely understood [7]. On the other hand, an approach developed in previous works has shown in a complete and well-founded way that the SPE resides, in fact, in unrenormalized collective excitations [8–11]. In addition, in Ref. [11], an analogy is presented between resonant electronic Raman scattering and the forced coupled harmonic oscillators problem, as well as, a correspondence with the formation of the superconducting state in BCS theory of normal metals.

This article aims to provide a theoretical interpretation on the behavior of the electronic Raman results found in Ref. [1], where for GaAs narrow quantum wells formed from the semiconductor sequence AlGaAs/AlAs/GaAs/AlAs/ AlGaAs, the Raman spectra are more influenced by SPE. Two wells were studied: one with 10 nm and the other with 17 nm GaAs width. For the 10 nm wide GaAs single quantum well (extreme case) only the SPE are seen in Raman spectra because collective excitations (CDE and SDE) are too small and overlapped by the SPE peak. A qualitative comparison between

☆ Selected paper from for IV Encontro Brasileiro de Espectroscopia Raman (EnBraER), in Juiz de Fora, December 06–09, 2015.

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http://dx.doi.org/10.1016/j.vibspec.2016.09.008
0924-2031/

theoretical and experimental data is performed which allows to interpret the asymmetries of Raman profile lines as the entrance or exit of resonance of collective modes overlapped with single-particle transitions.

2. Generalized self-consistent field theory

To calculate the response of a nonuniform electron gas submitted to the action of an external potential we use a formalism based on *time dependent local density approximation* (TDLDA) [12,13]. The fundamental idea of the self-consistent field theory approximation is to assume that the system of many-electrons responds to an effective field as a system of independent particles.[1] Therefore, an external potential acting on the system induces a charge density fluctuation as response to the applied field. This induced fluctuation produces an induced potential. This new potential acts on the system again producing a new total potential (effective potential, V^{eff}), and so on *ad infinitum*. In other words, this process is self-consistent and can be represented by Eq. (1),

$$V^{eff}(\mathbf{r},t) = V^{ext}(\mathbf{r},t) + V^{ind}(\mathbf{r},t), \tag{1}$$

where $V^{ext}(\mathbf{r},t)$ is the external potential (laser). The induced potential is described by [8,12,14]

$$V^{ind}(\mathbf{r},t) = \int \left[\frac{e^2}{\varepsilon_l(\omega)|\mathbf{r}-\mathbf{r}'|} + U_{exc}(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}') \right] \delta n(\mathbf{r}',t)d\mathbf{r}'. \tag{2}$$

The first term in Eq. (2) corresponds to the direct term (Coulomb interactions), the second include many-body effects (exchange and correlation). $\varepsilon_l(\omega) = \varepsilon_\infty(\omega^2 - \omega_{LO}^2)/(\omega^2 - \omega_{TO}^2)$ is the GaAs frequency-dependent lattice dielectric function that contains the bulk frequencies of the longitudinal (ω_{LO}) and transverse (ω_{TO}) optical phonons (the phonon lifetime was neglected) with dielectric constant ε_∞. The induced density fluctuation in the time-coordinate representation is given by,

$$\delta n(\mathbf{r},t) = \langle \hat{\psi}^\dagger(\mathbf{r},t)\hat{\psi}(\mathbf{r},t)\rangle_t = \sum_{\alpha\beta} \psi_\alpha^*(\mathbf{r})\psi_\beta(\mathbf{r})\langle \hat{c}_\alpha^\dagger \hat{c}_\beta\rangle_t, \tag{3}$$

where $\hat{\psi}^\dagger$ and $\hat{\psi}$ are field operators. The coefficients and $\hat{c}_\alpha^\dagger(\hat{c}_\beta)$ in Eq. (3) are wave functions and fermion creation (destruction) operators of single-particle conduction subband states. Taking the Fourier transform of Eq. (1) and solving the Heisenberg equation of motion for expectation values $\langle \hat{c}_\alpha^\dagger \hat{c}_\beta\rangle_t$ in Eq. (3) [15,16],

$$V_{ji}^{eff} = V_{ji}^{ext} + \sum_{mn} C_{ij,mn}\Pi_{mn}^0 V_{mn}^{eff}, \tag{4}$$

where

$$C_{ij,mn} = \frac{2\pi e^2}{Ak_z\varepsilon_l(\omega)} \int\int dz dz' \zeta_{ij}(z') e^{-k_z|z-z'|}\zeta_{mn}(z) + \frac{1}{A}\int dz[-U_{exc}^{CDE}(z)]\zeta_{mn}(z) \tag{5}$$

and

$$\Pi_{mn}^0 = \sum_{\mathbf{k},\sigma} \frac{f_m(\mathbf{k}) - f_n(\mathbf{k}+\mathbf{q})}{\epsilon_m(\mathbf{k}) - \epsilon_n(\mathbf{k}+\mathbf{q}) + \hbar\omega}. \tag{6}$$

$C_{ij,mn}$ is the matrix element of the Fourier transform which couples the charge density fluctuations between the subbands ij and mn. The first term in Eq. (5) represents contributions of the direct Coulombian interactions (Hartree term) and the second represents the exchange-correlation effects for the CDE. For the SDE, only the second term on the right side in Eq. (5) should be considered. The explicit expressions for the functional derivatives U_{exc}^{CDE} and U_{exc}^{SDE} are obtained from [17]. In Eq. (5), A is the area and the wave functions that describe the confinement in the z direction (written in terms of the envelope wave functions) are $\zeta_{ij}(z') = \zeta_i(z')\zeta_j(z')$. Π_{mn}^0 is the response function of the noninteracting electronic system, $f_{m(n)}(\mathbf{k})$ is the Fermi–Dirac distribution and $\epsilon(\mathbf{k})$ is the dispersion relation in the parabolic band approximation and $\mathbf{k},\mathbf{q} \in k_x k_y$. In this work, we considered only intersubband transitions where there is no lateral momentum transferred by light ($\mathbf{q}\to 0$). In this way, we can rewrite Eq. (6) as

$$\chi_{nm}^0 \equiv \lim_{\substack{q\to 0 \\ T\to 0\,\mathrm{K}}} (\Pi_{mn}^0 + \Pi_{nm}^0) = \frac{2N_{nm}\hbar\omega_{nm}}{\hbar^2(\omega^2 - \omega_{nm}^2)}. \tag{7}$$

Eq. (7) takes into account both upward and downward transitions. $\hbar\omega_{nm} = (\epsilon_n - \epsilon_m)$ is the bare electronic transition energy, $\Pi_{mm}^0 = 0$ and $N_{nm} \equiv \sum_{\mathbf{k},\sigma} f_m(\mathbf{k}) - f_n(\mathbf{k}) = \sum_{\mathbf{k},\sigma} 1$ is the number of electrons that contributed to each transition $m\to n$. The energy transferred to the electron system by the light is $\hbar\omega = \hbar(\omega_L - \omega_S)$ where $\hbar\omega_L$ and $\hbar\omega_S$ corresponds to the incident (laser) and scattered photon energies.

The idea employed in Ref. [8] is to map the inelastic light scattering of an electron gas into a problem of a set of forced damped harmonic oscillators. The damping of the transitions (i.e., scattering by impurities) is considered when we replace $\omega\to\omega + i\gamma$. Thus, $\hbar^2(\omega + i\gamma)^2 \simeq (\hbar\omega)^2 + i\Gamma\hbar\omega$, where $\Gamma \equiv 2\hbar\gamma$ is the damping related to each transition. Therefore, we associate a harmonic coordinate to each transition pair defined as

$$x_{ji} \equiv \frac{\sqrt{2N_{ji}\hbar\omega_{ji}}}{\hbar^2(\omega^2 - \omega_{ji}^2 + i\Gamma_{ji}\hbar\omega)} V_{ji}^{eff}. \tag{8}$$

In this way, Eq. (4) can be rewritten in the following way,

$$V_{ji}^{eff} = V_{ji}^{ext} + \sum_{\substack{mn \\ n>m}} C_{ij,mn}\chi_{nm}^0 V_{nm}^{eff}$$

$$\hbar^2(\omega^2 + i\Gamma_{ji}\hbar\omega)x_{ji} = \sqrt{2N_{ji}\hbar\omega_{ji}}V_{ji}^{ext} + \sum_{mn} U_{ij,mn}(\omega)x_{nm}, \tag{9}$$

where

$$U_{ij,mn}(\omega) \equiv C_{ij,mn}(\omega)\sqrt{4N_{ji}N_{nm}\hbar\omega_{ji}\hbar\omega_{nm}} + (\hbar\omega_{ji})^2\delta_{ij,mn}. \tag{10}$$

From Eqs. (5) and (10) one can see that the matrix U is real and symmetric. Therefore, its eigenvectors constitute a base which can be used to solve the equation of x via LU decomposition.

The expression for the inelastic light scattering which connects experiment and theory is the differential scattering cross section [18] given by

$$\frac{\partial^2\sigma}{\partial\Omega\partial\omega} = r_0^2\left(\frac{\omega_S}{\omega_L}\right)S(\omega), \tag{11}$$

where $r_0^2 = e^2/mc^2$ is the classical electron radius and

$$S(\omega) = \left\langle \sum_F |\langle F|\hat{M}_{eff}|I\rangle|^2\delta(E_F - E_I - \hbar\omega)\right\rangle_I, \tag{12}$$

is known as dynamic structure factor. $\hat{M}_{eff}$ is the effective operator for a transition between the many-body state $|I\rangle$ with energy E_I to the final state $|F\rangle$ with energy E_F and $\langle\ \ \rangle_I$ an average over the initial

[1] Calculations developed here follow the approach used in Refs. [8,11].