dc voltage effect on elementary excitations of a two-dimensional electron gas

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Using a generalized self-consistent field theory we obtained the intersubband spectra of charge-density excitations for the nonuniform electron gas of a GaAs-$Al_xGa_{1-x}As$ single quantum well submitted to dc gate voltages. The simulations performed with incoming laser light in extreme resonance conditions with the split-off gap of the GaAs demonstrate the collective character of the single-particle excitations and the different regimes of coupling between the charge-density excitations and the longitudinal optical phonon of GaAs.

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Inelastic light scattering is a standard tool to study the elementary excitations of quantum electron gases in semiconductor nanostructures. The richness of the technique allows one to estimate many-body effects, as the Coulomb and exchange-correlations interactions, by means of selection rules on the incoming and outgoing laser light polarizations.[1,2] Such estimations become possible due to the wealth of lasers now available, which allows one to perform experiments in which the differences between the incoming and outgoing laser light are in resonant conditions with an electronic or phononic excitation of the host semiconductor. Concerning the electronic excitations (intersubband or intrasubband) it is largely accepted that they can be described by a second- or third-order process of matter-light interaction when the electron gas is in the scope of Fermi liquid theory.[3] A different approach is theoretically expected in one-dimensional (1D) systems where the Luttinger model is assumed to be valid.[4] Unquestionably experimental realizations of a such model are still lacking although studies are conducted with potential candidates such as carbon nanotubes, semiconductor quantum wires, and 1D molecular systems.[5]

One of the most largely observed, although not fully understood, phenomenon in resonant Raman spectra of electron gases of semiconductors nanostructures is the so-called single-particle excitations (SPE's) process. These excitations occur irrespective of the dimensionality of the electron gas system (exceptions may occur in 1D systems as stated above) with their energy apparently equal to the bare electronic transitions. Such an excitation was not expected, as previous theories predicted collective behavior of the electron gas, i.e., collective charge- or spin-density excitations (CDE or SDE), whose energies were shifted from SPE due to the many-body effects.[5–11] Experimentally[5] and more recently also theoretically[9,10] it is agreed that the extreme resonance condition is a condition necessary for the appearance of SPE. Such conditions are reached with laser light resonant with the interband transitions, i.e., transitions involving valence- and conduction-band states through an optical gap. Therefore, a successful theory should involve the valence-band states in the calculations.

The subject of SPE, CDE, and SDE was also investigated by Anjos and Ioriatti[10] in GaAs:Si multilayer δ-doped systems. There it was shown that in order to obtain the signature of SPE, further requirements should be satisfied, not only the extreme resonance regime. Such conditions rely on the existence of degenerate and equally coupled intersubband excitations. The comparison between theory and experiment for depolarized Raman spectra (spin-density mechanism) and for various incident laser energies was excellent. Nevertheless in the case of polarized spectra (charge-density mechanism) the agreement was only qualitative, concerning the difference in intensity between SPE and CDE.[10,12] It was argued that such a difference comes from the fact that in δ-doping superlattices the electron gas coexists in the same spatial region of the dopants.[13] Their presence gives rise to fluctuations in the confinement potential with consequent breakdown of momentum conservation rules. As a result, the coherence of the collective charge-density excitations is lost, causing a partial transference of oscillator strength back to the single-particle modes.

In order to avoid the problems mentioned in the preceding paragraph, one may use the one-sided modulation-doped quantum wells. Such structures presents themselves as suitable structures to investigate the elementary excitations in the resonant Raman scattering. The spatial separation between the dopants and the electron gas ensures extremely high mobilities, which inhibits the back transference of oscillator strength. Nevertheless even in such structures there is controversy concerning the nature of the elementary excitations. As a matter of fact, in a recent experimental work[14] the inelastic light scattering of a modulation-doped GaAs-$Al_xGa_{1-x}As$ quantum-well structure was studied with respect to a continuously varied dc bias applied between a top and back gate contact. There, the interaction between the longitudinal-optical (LO) phonon of the GaAs and the charge-density excitations was studied. In the measured

spectra a very sharp structure near the LO-phonon energy was present. Its nature was attributed to a renormalized (softened) LO-phonon mode due to the reduction of the transverse optical (TO)–LO splitting by means of the electron gas. On the other hand, a previous work, also in the resonant Raman scattering of a GaAs-$Al_xGa_{1-x}As$ quantum well (without bias) interpreted the peak near the LO phonon of the GaAs as due to an upper CDE–LO-phonon coupled mode.[3] Such apparent contradictory interpretations motivated the present work.

In this work we use a generalized self-consistent field theory to investigate theoretically the elementary excitations of the resonant intersubband polarized Raman scattering of the electron gas of a modulation-doped GaAs-$Al_xGa_{1-x}As$ quantum well submitted to a dc bias. The spectra are obtained with incoming light resonant with interband transitions between the valence states of the split-off gap of the GaAs and states of the conduction band. We show that the nature of SPE is related to nonrenormalized collective physical process. Also, we show that under suitable electronic densities and suitable resonant conditions a branch of an CDE–LO-phonon mode structure should appear near the LO phonon of GaAs. This structure represents the weak coupling regime between the charge-density excitations and the LO phonon in opposition to the strong one also shown.

The sample considered consists of a one-sided modulation-doped GaAs-$Al_{0.33}Ga_{0.67}As$ quantum well. From the $Al_xGa_{1-x}As$ buffer barrier, it has a 245-Å-wide GaAs quantum well, a $Al_xGa_{1-x}As$ spacer layer of 200 Å, and an intentionally doped layer of ionized donor impurities uniformly distributed over a layer of 400 Å, with a density of $N_D=0.62\times10^{18}$ cm^{-3}. At the top of the structure a gate voltage is applied, and the potential of the surface, is then given by $\phi_s=\phi_{BS}-V_g$, where $\phi_{BS}=1000$ meV is the contact Schottky potential. In the structure considered we have to take into account a residual distribution of ionized acceptor impurities uniformly distributed over the system of $N_A=1.0\times10^{14}$ cm^{-3}. Figure 1 presents the self-consistent potential profiles for gate voltages, $V_g=-360$, -280, -200, -80, 0, 80, 200, 280, and 360 meV, respectively. From Fig. 1 we see that the increase of the gate voltage results in a decrease of the surface potential, which, by its turn, leads the system to accumulate charge into the barrier, the region between the gate contact and the well. In such cases, for the gate voltage above a determined value, the deformation of the potential of the barrier is strong enough to accumulate charge in this region. This is shown in Fig. 2, where the behavior of the electronic density for the gate voltages of Fig. 1 is presented. For our case we have confinement into the barrier for the following gate voltages: $V_g=200$ mV, $V_g=280$ mV, and $V_g=360$ mV. However, for all cases shown here, the first subband (ground state) of the system is localized into the well. Moreover, with the parameters used to configure the sample we were not able to populate a second subband in the well via the tuning of the gate voltage. Our trials to populate a second subband in the well resulted in a populated subband into the barrier which increases with the increase of the gate voltage. It is interesting to note also in Fig. 2 that a depletion of the electrons from the barrier by means of the gate voltage decrease does not imply an increase of the density in the well. This is due to the passivation of the acceptor impurities along the structure.

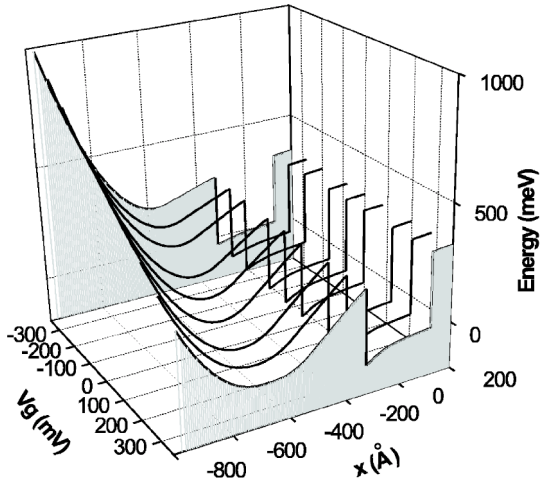

FIG. 1. The calculated potential profiles of a one side gated modulation-doped quantum well with gate voltages, $V_g=-360$, -280, -200, -80, 0, 80, 200, 280 and 360 mV.

Figure 3 shows the energy dispersion of various subbands and the total density in the well as a function of the previous gate voltages. In the figure the Fermi level is considered as the reference level. For gate voltages of $V_g=280$ and 360 mV the third subband disappears (goes to the barrier). This, as we shall see below, will affect profoundly the behavior of the Raman profiles, as the Stokes energies of interest in this paper correspond to transitions between the first to the second and third subbands. In the figure we see clearly, as mentioned in the previous paragraph, the reduction of the electronic density in the quantum well for negative gate voltages albeit the complete depletion of electrons from the barrier.

In Fig. 4 the energy transitions of the various subbands as a function of the gate voltages are plotted. In particular the transitions $E_{01}$and E_{02} will represent the energies of the SPE in the polarized Raman scattering bellow. Let us turn now to

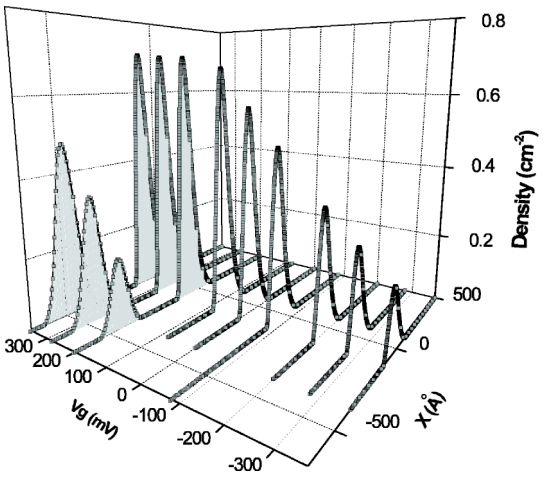

FIG. 2. Calculated 2D electronic density in the well in function of the same gate voltages of Fig. 1.

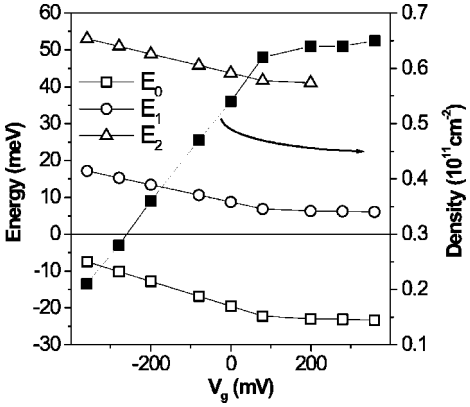

FIG. 3. Calculated subbands energy dispersions and electronic density of the quantum well in function of the same gate voltages of Fig. 1.

the calculations of the Raman scattering cross sections.

The inelastic light scattering spectra will be obtained with incident laser energy tuned above the $E_0+\Delta_0$ gap of GaAs ($\sim$1.86 eV). Around this value and around the 2D electron density here calculated, Danan *et al.*[3] observed a broad band in the Raman efficiency of the collective spin-density excitations. Such a band typifies a second-order process. Therefore here we will not consider third-order processes involving excitonic transitions that give rise to very sharp peaks in the Raman efficiency for both charge- and spin-density excitation mechanisms. Nevertheless, apart from different Raman efficiencies between the various elementary excitations, no other characteristics of the spectra are changed between the second- and third-order process. This strongly suggests that the conclusions obtained in this work may remain valid even in the domain of the third-order processes.

In order to calculate the polarized Raman cross section, suppose that our nonuniform electron gas is submitted to a small time-dependent perturbation $M(0)e^{-i\omega t}$. In time t the mean value of $M^{\dagger}(0)$ will be given by

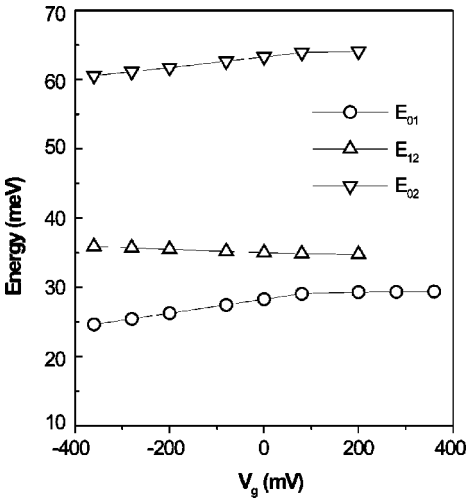

FIG. 4. Calculated subband energy transitions as a function of the same gate voltages of Fig. 1.

$$\langle \Psi(t)|M^{\dagger}(0)|\Psi(t)\rangle, \tag{1}$$

where $|\Psi(t)\rangle$ is the time-dependent many-body wave function expanded in terms of the single-particle states of the quantum well until first order in the perturbation. Taking from Eq. (1) the term-dependent of $-\omega$ one can arrive at

$$\int_{-\infty}^{\infty} \frac{dt}{2\pi} e^{i\omega t}\langle 0|M^{\dagger}(t)M(0)|0\rangle = -\frac{\hbar}{\pi}\mathrm{Im}\langle \Psi(t)|M^{\dagger}(0)|\Psi(t)\rangle_{\omega}. \tag{2}$$

The knowledge of the right side of the Eq. (2) will furnish the desired Raman cross section due to the fact that the Fourier transform of the correlation function, on the left-hand side, is proportional to the Raman cross section.

In second quantization formalism, the effective scattering operator for the charge-density mechanism is given by[10]

$$M(0) = \sum_{\alpha\beta} \gamma_{\beta\alpha}(c^{\dagger}_{\beta\uparrow}c_{\alpha\uparrow} + c^{\dagger}_{\beta\downarrow}c_{\alpha\downarrow}), \tag{3}$$

where $c^{\dagger}_{\beta}$ (c_{α}) are creation (destruction) fermion operators of the final (initial) one-electron conduction subband states with spin up ($\uparrow$) or down ($\downarrow$). Such states have wave functions $\psi_{\alpha(\beta)}(z)e^{i\mathbf{q}\cdot\rho}/\sqrt{A}$ and energies $\epsilon_{\alpha(\beta)}=\hbar\omega_{\alpha(\beta)}+\hbar^2q^2/2m$, where $\mathbf{q}$ represent 2D wave vectors perpendicular to the growth direction (z) and A is the sample area. In Eq. (3)

$$\gamma_{\beta\alpha} = \frac{[P^2_{cv}]}{3m_0}\sum_h \frac{\langle\beta|e^{i\mathbf{k}_L\cdot\mathbf{r}}|h\rangle\langle h|e^{-i\mathbf{k}_S\cdot\mathbf{r}}|\alpha\rangle}{E_g+\epsilon_{\beta}+\epsilon_h-\hbar\omega_L+i\eta}, \tag{4}$$

corresponds to an external potential (resonant factor) involving interband excitations between the split-off hole (h) and conduction band states. It can be interpreted as a generalized force produced in the electron system by the optical pumping. Such analogy gives to the operator in the right side of Eq. (3) the status of a generalized displacement as the product of the "force" and the "displacement" will reproduce the effective Hamiltonian acting upon the system. In Eq. (4), $[P^2_{cv}]$ are the appropriate interband matrix elements involving the Γ_7 split-off hole and Γ_6 Bloch wave functions, m_0 is the bare electron mass, E_g is the $E_0+\Delta_0$ band gap of GaAs, η is a phenomenological damping, $\hbar\omega_L$ is the energy of the incident light, and $|\mathbf{k}_L|=|\mathbf{k}_S|=k$ are the moduli of the incident (scattered) light wave vector $[\mathbf{k}=(\mathbf{k}_{\parallel},k_z)]$. In this work we deal only with the intersubband process with no lateral momentum transfer therefore in the calculations $\mathbf{k}_{\parallel}\rightarrow\mathbf{0}$.

From Eq. (2) one can see that we have to calculate expected values of the Fourier transform of the induced density fluctuations. This is done by means of the one-body density matrix in the linear regime, which gives

$$\langle c^{\dagger}_{\alpha\uparrow}c_{\beta\uparrow} + c^{\dagger}_{\alpha\downarrow}c_{\beta\downarrow}\rangle_{\omega} = \frac{4\hbar\omega_{\beta\alpha}n_{\alpha}[1-n_{\beta}]}{\hbar^2(\omega^2-\omega^2_{\beta\alpha}+i\zeta_{\beta\alpha}\omega)}\delta V^{\mathrm{tot}}_{\beta\alpha}, \tag{5}$$

where

$$\delta V^{tot}_{\beta\alpha} = \gamma_{\beta\alpha} + \sum_{\gamma\delta} \frac{4\hbar\omega_{\delta\gamma} n_\gamma [1 - n_\delta] C_{\beta\alpha,\delta\gamma}}{\hbar^2(\omega^2 - \omega^2_{\delta\gamma} + i\zeta_{\delta\gamma}\omega)} \delta V^{\rm tot}_{\delta\gamma} \qquad (6)$$

is the total potential acting on the system composed by the external potential plus the induced potential. In Eq. (6), n_γ (n_δ) denotes the Fermi occupation number of occupied (unoccupied) conduction subband states, $\hbar\omega = \hbar(\omega_L - \omega_S)$ is the energy transferred to the electron system by the light, $\hbar\omega_{\delta\gamma}$ is the bare electronic transition, $\zeta_{\delta\gamma}$ is the damping associated with the lifetime of the transition, and

$$C_{\beta\alpha,\delta\gamma} = \frac{2\pi e^2}{\varepsilon_l(\omega) k_\parallel A} \int dz \int dz' \varphi_{\beta\alpha}(z) \times [e^{-k_\parallel |z-z'|} - U_{xc}(z)\delta(z-z')] \varphi_{\delta\gamma}(z), \qquad (7)$$

represents the Coulombian and exchange-correlations interactions[7] between a pair of excitations where $\varepsilon_l(\omega) = \varepsilon_\infty(\omega^2 - \omega^2_{\rm LO})/(\omega^2 - \omega^2_{\rm TO})$ is the dielectric function which takes in account the coupling of the excitations with the optical phonons and $\varphi_{\beta\alpha} = \psi_\beta(z)\psi_\alpha(z)$. Now, associating to each pair of transition a generalized coordinate **x** whose components[10] are given by

$$x_{\beta\alpha} = \frac{\sqrt{4\hbar\omega_{\beta\alpha} n_\alpha [1 - n_\beta]}\, \delta V^{\rm tot}_{\beta\alpha}}{\hbar^2(\omega^2 - \omega^2_{\beta\alpha} + i\zeta_{\beta\alpha}\omega)}, \qquad (8)$$

Eq. (6) can be rewritten as a matrix whose elements are given by

$$\hbar^2(\omega^2 + i\zeta_{\beta\alpha}\omega) x_{\beta\alpha} = \sum_{\gamma\delta} U_{\beta\alpha,\delta\gamma} x_{\beta\alpha} + N_{\beta\alpha}\gamma_{\beta\alpha}, \qquad (9)$$

where

$$U_{\beta\alpha,\delta\gamma} = N_{\beta\alpha} C_{\beta\alpha,\delta\gamma} N_{\delta\gamma} + (\hbar\omega_{\beta\alpha})^2 \delta_{\beta\alpha,\delta\gamma} \qquad (10)$$

and $N_{\beta\alpha} = \sqrt{4\hbar\omega_{\beta\alpha} n_\alpha [1 - n_\beta]}$. From Eqs. (7) 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[15] and consequently finding the cross sections.

Figure 5 presents the results for the charge-density excitations for incident laser energy of 1.945 eV and for the selected $V_g = -360$, -200, -80, 80, 280, and 360 mV. In order to arrive at this result, around 20 valence subbands were calculated. Nevertheless, from these, on average, four subbands were used as only these effectively have a non-negligible superposition with the three conduction subbands considered in the well. Also, indirect transitions (quantum-well-barrier transitions) were discarded based on the fact that their oscillator strengths are around six orders of magnitude less than the direct ones.

Based on the results of the Fig. 4 one can identify in Fig. 5 two structures whose energies correspond to the energy of the bare transitions of the quantum-well electron gas. By this reason, they are identified as SPE. The first, with low energy, originates from transitions from the first to the second subband, which we label as $\rm SPE_{01}$. From Fig. 4 and for the gate voltages presented in Fig. 5, its energy values are given by $E_{01} = 24.64$, 26.24, 27.45, 29.26, 29.32, and 29.38 meV, respectively. Note that $\rm SPE_{01}$ resonance behavior is directly

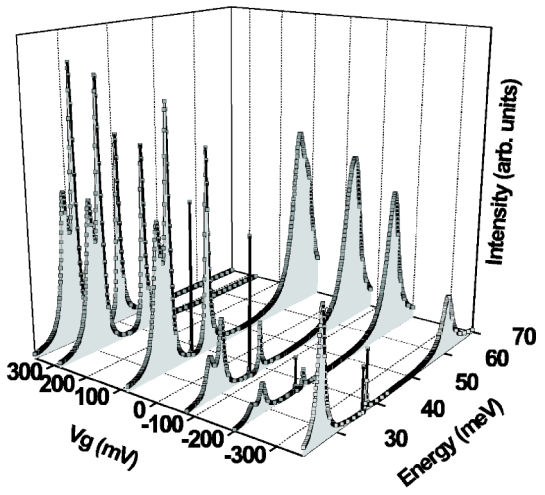

FIG. 5. Calculated polarized resonant Raman spectra of the QW as a function of selected gate voltages showing the spectral lines of the collective (CDE) and single-particle (SPE) excitations for incident laser energy of 1.945 eV.

proportional to the increase of the gate voltage and that for $V_g = -360$ mV it cannot be observed. Such resonant behavior is accounted for by the different superposition of conduction-valence band states and by the resonant energy denominator of Eq. (4). The second excitation, $\rm SPE_{02}$, corresponds to transitions from the first to the third subband with voltage- ($V_g = -360, -200, -80, 80$ mV) dependent energy values $E_{02} = 60.54$, 61.70, 62.63, and 63.88 meV given by Fig. 4. The excitation is not present for gate voltages $V_g = 280$ and 360 mV (Figs. 4 and 5) due to the fact that at such voltages the third subband goes to the barrier. Like $\rm SPE_{01}$, for $V_g = -360$ mV, $\rm SPE_{02}$ cannot be observed for every gate voltage. This is illustrated for clarity in Fig. 6, and is a fingerprint of the near resonant Raman spectra where only collective modes are expected to be present. The interesting point in our calculations is that SPE's are derived from the imaginary part of the expected value of an effective scattering operator which can be rewritten in terms of a generalized coordinate **x** [see Eq. (9)], which is essentially a collective term, as every

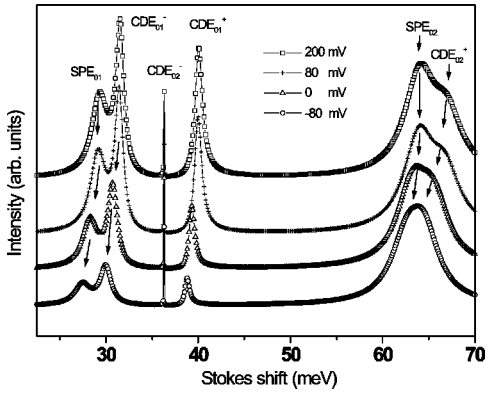

FIG. 6. Calculated polarized resonant Raman spectra of the QW as a function of selected gate voltages showing the merging of the $\rm CDE^+_{02}$ and $\rm SPE_{02}$ peaks when the extreme resonance regime is left. Other excitations are also shown.

possible transition is coupled via the many-body matrix **U**. This demonstrates that the SPE can be in reality collective not renormalized excitations.

The remaining spectral profiles in Fig. 5 represent collective CDE–LO-phonon coupled modes that present two types of coupling, strong and weak ones. In the former case, the plasmalike excitations should be near the LO phonon of the GaAs and by this reason will interact strongly with the phonon giving rise to modes similar to the L_+ and L_- in three-dimensional systems. This is the case for transitions originated from the first to the second subbands in the quantum well and labeled E_{01} in Fig. 4. Accordingly, we identify them as collective modes CDE_{01}^{-} and CDE_{01}^{+} in Fig. 6. We remark that the electron-phonon coupling is introduced via the dielectric function $\varepsilon_l(\omega)$ in Eq. (7) where the phonon lifetime was not considered. This was done in order to have a real matrix **U**.

The weak coupling regime represents the coupling between the collective modes basically from the first and third subbands with the LO phonons. Therefore we labeled them as CDE_{02}^{-} and CDE_{02}^{+}. The first mode, seen in Fig. 5 and 6, is a very thin peak with energy of 36.4 meV, which is almost the energy of the LO phonon (36.5 meV) and may be the peak which nature was attributed to a renormalized (softened) LO-phonon mode.[14] From Fig. 6, one can observe that its behavior does not follows the general tendency of the others peaks with increasing gate voltage, i.e., the reduction of their energy. At the high-energy side we have the CDE_{02}^{+} mode, which is represented by a wing in the SPE_{02} spectra. Figure 6 shows that with the decrease of gate voltage it tends to merge with single-particle excitation, which means that one is leaving the extreme resonance condition and performing a transition to the near-resonance regime that, as stated above, will present only collective excitations. Therefore we consider that for $V_g=-80$ mV almost all single-particle oscillator strength was transferred to the collective mode CDE_{02}. A similar fact occurs with SPE_{01} (see Fig. 5), nevertheless, for a smaller value of the gate voltage.

In summary, we presented a generalized self-consistent field theory in order to investigate the elementary excitations of polarized intersubband Raman spectra of modulation doped quantum wells subjected (or not) to applied gate voltages. Contrary to what was expected, the decreases of the gate voltages will not be implied in an enhancement of the population of the carriers in the well, as the extra carriers will be extracted from the well due to the passivation of the acceptor levels along the structure. Concerning the Raman spectra, it was demonstrated that single-particle excitations may be not renormalized collective excitations. They are present in the Raman spectra in a regime of extreme resonance where the incoming laser light matches the interband conduction-band–valence-band transitions. In our case, the split-off conduction band undergoes transitions. Otherwise, only collective excitations will be present. In our system, the collective excitations involve two coupling regimes with the LO phonon of the GaAs, the weak and the strong one. The first involves transition energies (the first to the third subband) around the double of the energy of the phonon. As a consequence they give rise to a quasiphonon and a quasiplasmon in the spectra. From our results, these excitations will be unambiguously present in the Raman spectrum when they are in their resonance regime. The strong coupling regime represents mixed electronic and LO-phonon excitations when the plasmalike electronic excitation energy and the phonon energy are comparable. Such modes are routinely present in one-sided polarized resonant Raman spectra, which serves as a check of our theory. As a final remark, we would like to state that although our paper derives firm conclusions concerning the nature of the elementary excitations in resonant Raman scattering of 2D systems, one should take extra care when comparing Raman efficiencies and absolute positions of the excitations predicted here and those due to a real experiment. We expect some deviations resulting from the role of the illumination[14] and possibly also from the band gap renormalization effect,[12,14] which alter the electronic density of the well and were not contemplated in this work.

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