Collective and single-particle excitations in Raman scattering of multilayer δ-doped systems

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(Received 19 June 2000; revised manuscript received 25 August 2000; published 29 December 2000)

We report theoretical calculations of resonant Raman cross sections via spin- and charge-density mechanisms. The theory clearly demonstrates that the sufficient conditions for the appearance in extreme resonance regime of simultaneous collective and single-particle modes in the nonuniform electron gas of multilayer δ-doped systems resides in the existence of degenerate and equally coupled intersuband excitations. A comparison between theoretical and experimental data lead us to believe that the disorder inherent to the doping process should be relevant in order to give the correct intensity between the collective and single-particle modes.

DOI: 10.1103/PhysRevB.63.035404 PACS number(s): 73.20.Mf, 78.30.Fs, 71.45.−d, 73.21.−b

Among the various spectroscopic probes, inelastic light scattering is an especially powerful technique. By means of polarization selection rules,[1] one can excite collective charge- and spin-density excitations (CDE's and SDE's). The first are observed when the incoming and scattered light polarizations are parallel to each other (polarized geometry). Their energy modes are renormalized with respect to single-particle excitations (SPE's) due to direct and exchange Coulomb interactions. SDE's occur when the polarizations are perpendicular (depolarized geometry), and are affected only by exchange interactions resulting in redshifted modes with respect to the single-particle energies. An intriguing fact occurs when Raman measurements are performed in a regime where the incoming laser energy matches an optical gap of the host semiconductor, the *extreme resonance* regime. In this case, Raman spectra of quantum wells,[2] quantum wires and dots,[3] and even of *n*-GaAs,[4] exhibit, in addition to collective CDE's and SDE's, unexpected unscreened SPE's. Additionally to intersubband SPE, intrasubband SPE was also reported.[5–7] From theoretical point of view (also experimentally) it is consensual that for a correct description of the experiments the valence band states should be taken in account. In fact, recently SPE's were obtained in calculations performed by means of a resonant random-phase approximation[8] in one dimensional (1D) and 2D systems, and also in quantum dots using the TDLDA (time-dependent local-density approximation).[9] Both papers stressed the relevance of the valence band in the resonant process, and suggested that so-called single-particle excitations are in reality the collective ones. Reinforcement of this interpretation was also reported for quantum wires treated within the Luttinger model, where low energy peaks in polarized spectra commonly associated with SPE's, were interpreted as intrasubband collective spin-density excitations.[10] Although the puzzle of simultaneous SPE's and collective excitations has been the subject of a great amount of work, in our opinion there is still a lack of information concerning the physics of the phenomenon. This is the main subject of the present work. We propose in a clear physical basis an *intersubband* theory of resonant Raman scattering which provides *sufficient* conditions for the signature of SPE in an otherwise collective spectra. Such conditions rely on the existence of degenerate and equally coupled intersubband excitations of an electron gas. We show that the very nature of SPE is related to the existence of *unrenormalized* collective excitations.

Our theoretical TDLDA-based studies, conducted in near and extreme resonance regimes, deal with the existence of single-particle and collective excitations of δ-Si:GaAs multilayer systems. Such well-known structures, for relatively high silicon planar doping, result in periodical space-charge potentials along the growth direction which confine a nonuniform electron gas around the donor sheets. Their shallow-well confinement makes them adequate for resonant Raman studies performed in the $E_0+\Delta_0$ spin-split edge. For incident light perpendicular to the doping sheets and resonant with the $E_0+\Delta_0$ optical gap, our theoretical polarized and depolarized cross sections show that the Raman spectra will be composed of collective and single-particle intersubband contributions. Calculations performed for a particular δ-doped structure agree with our available experimental data and with those previously reported,[11] providing a complete understanding of the spectral line shapes. In the depolarized case, a collective term could not be observed due to small exchange-correlation contributions which confers to the spectrum a single-particle character. On the other hand, in the polarized case, Coulombian coupling effects give rise to single-particle excitations similar to those of the depolarized spectra, as well as to a definite ''pure'' collective mode. Comparison between theoretical and experimental data also lead us to believe that the disorder inherent to the doping process should be relevant in order to give the correct intensity between the collective and single-particle excitations. The process can be understood as a back transference of oscillator strengths to the single-particle transitions caused by the lost of coherence of the ''pure'' collective modes.

The electronic system considered is an 11-period δ-doping superlattice grown along the *z* direction with a unit cell $d=500$ Å and a sheet carrier density of $n_s=1.0\times10^{12}$ cm^{-2}. Density-functional theory calculations in the local-density approximation (LDA) result in a conduction

band formed of three occupied minibands. The first is a flat 2D band confining 76% of the electrons. The remaining two are nearly free-electron (NFE) bands with 21% and 3% of the total number of available electrons, respectively, the latter being half filled.[12] For the intermediate spin-split hole states, the same conduction-band boundary conditions were adopted, with the assumption that holes are subjected only to the Hartree part of the self-consistent potential.

To calculate the scattering cross sections, we extended the Hamilton and McWhorter formula[13] within a time-dependent LDA.[14,15] Based on such an extension, zero-temperature cross-sections may be expressed in terms of autocorrelation functions of the form

$$G(\omega)=\int \frac{dt}{2\pi} e^{i\omega t}\langle M^{\dagger}(t)M(0)\rangle, \tag{1}$$

where $M^{\dagger}(t)$ is the appropriate scattering operator in the Heisenberg representation. These in turn are related via a fluctuation-dissipation theorem to the response function $\langle M^{\dagger}(0)\rangle_{\omega} e^{-i\omega t}$ of the system submitted to a time-dependent perturbation of the form $M(0)e^{-i\omega t}$; that is,

$$G(\omega)=-\frac{\hbar}{\pi}\mathrm{Im}[\langle M^{\dagger}(0)\rangle_{\omega}], \tag{2}$$

where $\langle M^{\dagger}(0)\rangle_{\omega}$ is the Fourier amplitude at $-\omega$ of the expectation value of the effective Schrödinger operator $M(0)=\Sigma_{\alpha\beta}[\delta V^{ext}_{\beta\alpha}]_{c,s}(c^{\dagger}_{\beta\uparrow}c_{\alpha\uparrow}\pm c^{\dagger}_{\beta\downarrow}c_{\alpha\downarrow})$,[1] in the evolving many-particle state $|\Psi(t)\rangle$ originally the ground state at $t=-\infty$. In the preceding equations $\hbar\omega=\hbar(\omega_L-\omega_S)$ is the light energy transferred to the system, with $\omega_L(\omega_S)$ being the incident (scattered) light frequency. Since we are only concerned with intersubband excitations, we assume a light wave-number transfer, $|\mathbf{q}|=|\mathbf{k}_L-\mathbf{k}_S|$ and an incoming and scattered light wave number, $|\mathbf{k}_L|=|\mathbf{k}_S|=q/2$ along the growth direction of the superlattice, the z axis. The above considerations show that the power spectrum of the density fluctuations may be obtained from a knowledge of the response function

$$\langle M^{\dagger}(0)\rangle^{c,s}_{\omega}=\sum_{\alpha\beta}[\delta V^{ext}_{\beta\alpha}]^{*}_{c,s}\langle c^{\dagger}_{\alpha\uparrow}c_{\beta\uparrow}\pm c^{\dagger}_{\alpha\downarrow}c_{\beta\downarrow}\rangle_{\omega}, \tag{3}$$

where the fermion operators stand to states α and β in the conduction band along the growth direction (n,k_z), with spin up (down), and to two-dimensional plane-wave states, parallel to the doping planes, with wave vectors $\mathbf{k}_{\parallel}$. $c(s)$ and $+(-)$ refer to the charge- (spin-) density mechanism, with enhancement factors given by

$$[\delta V^{ext}_{\beta\alpha}]_{c,s}=\frac{[P^2_{cv}]_{c,s}}{3m_0}\sum_{h}\frac{\langle\beta|e^{(1/2)iqz}|h\rangle\langle h|e^{(1/2)iqz}|\alpha\rangle}{E_g+i\gamma_g+\epsilon_{\beta}+\epsilon_h-\hbar\omega_L}, \tag{4}$$

where the h's are the valence-band intermediate states, E_g is the split-off gap broadened by a phenomenological damping constant γ_g, and $[P^2_{cv}]_{c,s}$ are the interband matrix elements for the appropriate scattering geometry.[16] $\epsilon_{\beta}(\epsilon_h)$ stand for the conduction (valence) subband energies. To simplify the notation in the following, $[\delta V^{ext}_{\beta\alpha}]_{c,s}=\delta V^{ext}_{\beta\alpha}$.

In order to solve Eq. (3), we observe that the expected value on its right-hand side is just the induced density fluctuation of the charge or spin, which in the time-coordinate representation is written as

$$\delta n(\mathbf{x},t)=\sum_{\alpha\beta}\psi^{*}_{\alpha}(\mathbf{x})\psi_{\beta}(\mathbf{x})\langle c^{\dagger}_{\alpha\uparrow}c_{\beta\uparrow}\pm c^{\dagger}_{\alpha\downarrow}c_{\beta\downarrow}\rangle_t. \tag{5}$$

The central idea behind our generalized self-consistent-field approximation is to assume that the many-body system responds to the total field

$$\delta V^{tot}(t)=\delta V^{ext}(t)+\delta V^{ind}(t), \tag{6}$$

as a system of independent particles. Here $\delta V^{ext}(t)$ is the weak external perturbation, and $\delta V^{ind}(t)$ the induced (charge or spin) fluctuation. In the static case where these fluctuations are treated exactly within the formalism of density-functional theory $(\omega=0)$, induced potentials are given in terms of the functional derivatives of the electron-electron energy density. Besides direct Coulomb interactions (the Hartree term), these include many-body effects due to exchange and correlation. Even without a formal justification, we take these potentials as representative of a time-dependent situation $(\omega\neq 0)$. This means that the induced charge-density potential is given by

$$\delta V^{ind}(\mathbf{x},t)=\frac{1}{\epsilon_L(\omega)}\int\frac{e^2}{|\mathbf{x}-\mathbf{y}|}\delta n(\mathbf{y},t)d\mathbf{y}+U_{cd}(\mathbf{x})\delta n(\mathbf{x},t), \tag{7}$$

with $\epsilon_L(\omega)$ being the frequency-dependent lattice dielectric function where the phonon lifetime was neglected.[15] On the other hand, the induced potential due to spin-density fluctuations is given by

$$\delta V^{ind}(\mathbf{x},t)=U_{sd}(\mathbf{x})\delta n(\mathbf{x},t). \tag{8}$$

In Eqs. (7) and (8), $U_{cd}(\mathbf{x})$ and $U_{sd}(\mathbf{x})$ are functional derivatives whose expressions may be obtained from the uniform electron-gas data.[17]

Besides the problem of solving the equations of motion for expectation values of the electron-hole pairs of Eq. (5), we were also aware of the fact that the local current must be conserved, i.e., that

$$\nabla\cdot\mathbf{J}+\frac{\partial}{\partial t}n=0. \tag{9}$$

In order to take such a fact in account, the following possible current-conserving scheme is proposed. First we consider equations of motion in the absence of damping, that is,

$$i\hbar\frac{\partial}{\partial t}\langle c^{\dagger}_{\alpha}c_{\beta}+c^{\dagger}_{\beta*}c_{\alpha*}\rangle_t=\hbar\omega_{\beta\alpha}\langle c^{\dagger}_{\alpha}c_{\beta}-c^{\dagger}_{\beta*}c_{\alpha*}\rangle_t \tag{10}$$

and

$$i\hbar\frac{\partial}{\partial t}\langle c_\alpha^\dagger c_\beta - c_{\beta*}^\dagger c_{\alpha*}\rangle_t = \hbar\omega_{\beta\alpha}\langle c_\alpha^\dagger c_\beta + c_{\beta*}^\dagger c_{\alpha*}\rangle_t + 2(n_\alpha - n_\beta)\delta V_{\beta\alpha}^{tot}, \tag{11}$$

where $n_{\alpha(\beta)}$ denotes the Fermi occupation number of the initial (occupied) and final (unoccupied) conduction-band states. In these equations we have considered that if α (which here includes also spin coordinate) corresponds to a state with energy ϵ_α and an orbital wave function $\psi_\alpha(\mathbf{x})$, then the state $\alpha*$ corresponds to a one-particle state with an orbital wave function $\psi_\alpha^*(\mathbf{x})$ with the same energy ϵ_α. Note also that $\delta V_{\beta\alpha}^{tot}(t) = \delta V_{\alpha*\beta*}^{tot}(t)$. To include damping, we keep the first of these equations without modification, and modify the second by the inclusion of an Ohmic relaxation term $\gamma_{\beta\alpha}$ which is associated with the damping of each pair transition with energy $\hbar\omega_{\beta\alpha} = \epsilon_\beta - \epsilon_\alpha$, that is,

$$i\hbar\frac{\partial}{\partial t}\langle c_\alpha^\dagger c_\beta - c_{\beta*}^\dagger c_{\alpha*}\rangle_t = \hbar\omega_{\beta\alpha}\langle c_\alpha^\dagger c_\beta + c_{\beta*}^\dagger c_{\alpha*}\rangle_t + 2(n_\alpha - n_\beta)\delta V_{\beta\alpha}^{tot} - i\hbar\gamma_{\beta\alpha}\langle c_\alpha^\dagger c_\beta - c_{\beta*}^\dagger c_{\alpha*}\rangle_t. \tag{12}$$

Since

$$\frac{\partial}{\partial t}n(\mathbf{x},t) = -i\frac{\omega}{2}\sum_{\alpha\beta}\langle c_\alpha^\dagger c_\beta + c_{\beta*}^\dagger c_{\alpha*}\rangle_t \psi_\alpha^*(\mathbf{x})\psi_\beta(\mathbf{x}) \tag{13}$$

and

$$\boldsymbol{\nabla}\cdot\mathbf{J} = \frac{i}{2}\sum_{\alpha\beta}\omega_{\beta\alpha}\langle c_\alpha^\dagger c_\beta - c_{\beta*}^\dagger c_{\alpha*}\rangle_t \psi_\alpha^*(\mathbf{x})\psi_\beta(\mathbf{x}), \tag{14}$$

it is clear that the equation of motion of Eq. (10) is simply a statement of conservation of the local current. Solving Eqs. (10), (12), (13), and (14) for $\langle c_\alpha^\dagger c_\beta\rangle_t$, we found that the induced density fluctuations are given by

$$\delta n(\mathbf{x},t) = \sum_{\alpha\beta}\Phi_{\beta\alpha}(\mathbf{x})\frac{2\hbar\omega_{\beta\alpha}n_\alpha(1-n_\beta)}{\hbar^2(\omega^2 - \omega_{\beta\alpha}^2 + i\gamma_{\beta\alpha}\omega)}\delta V_{\beta\alpha}^{tot}(t), \tag{15}$$

where $\Phi_{\beta\alpha}(\mathbf{x}) = \psi_\alpha^*(\mathbf{x})\psi_\beta(\mathbf{x})$.

In the next step we associate a harmonic coordinate $x_{\beta\alpha}$ with each pair transition $(\beta\alpha)$,

$$x_{\beta\alpha} = \frac{\sqrt{2\hbar\omega_{\beta\alpha}n_{\beta\alpha}}}{\hbar^2(\omega^2 - \omega_{\beta\alpha}^2 + i\gamma_{\beta\alpha}\omega)}\delta V_{\beta\alpha}^{tot}, \tag{16}$$

where $n_{\beta\alpha} = n_\alpha(1-n_\beta)$. Since pair transitions interact via their associated density fluctuations, the various amplitudes $x_{\beta\alpha}$ are coupled to one another, resulting such that Eq. (6) may be rewritten as

$$\hbar^2(\omega^2 - \omega_{\beta\alpha}^2 + i\gamma_{\beta\alpha}\omega)x_{\beta\alpha} = \sum_{\delta\gamma}U_{\beta\alpha,\delta\gamma}x_{\delta\gamma} + \sqrt{2\hbar\omega_{\beta\alpha}n_{\beta\alpha}}\delta V_{\beta\alpha,}^{ext}, \tag{17}$$

where the matrix elements $U_{\beta\alpha,\delta\gamma}$ for the case of charge-density fluctuations are defined as

$$U_{\beta\alpha,\delta\gamma}(\omega) = \sqrt{4\hbar\omega_{\beta\alpha}\hbar\omega_{\delta\gamma}n_{\beta\alpha}n_{\delta\gamma}}\int\int\Phi_{\beta\alpha}^*(\mathbf{x})\Phi_{\delta\gamma}(\mathbf{y}) \times\left[\frac{e^2}{\epsilon_L(\omega)|\mathbf{x}-\mathbf{y}|} + U_{cd}(\mathbf{x})\delta(\mathbf{x}-\mathbf{y})\right]d\mathbf{x}d\mathbf{y}. \tag{18}$$

Therefore, the problem is reduced to that of finding the response of a set of driven linearly coupled damped harmonic oscillators. Instead of considering the general situation where the external potential may depend on coordinates that run perpendicular to the superlattice axis, we restricted ourselves to the case of external fields varying only along the superlattice axis. Since in this case the density fluctuations are functions of z only, the induced potentials produce a collective oscillation within a pair of subbands $(\alpha\beta)$ in which all vertical transitions participate with equal amplitudes. Accordingly, we define a collective harmonic coordinate $\overline{x}_{\beta\alpha}$ by

$$\overline{x}_{\beta\alpha} = \frac{\sqrt{2\hbar\omega_{\beta\alpha}N_{\beta\alpha}}}{\hbar^2(\omega^2 - \omega_{\beta\alpha}^2 + i\gamma_{\beta\alpha}\omega)}\delta V_{\beta\alpha}^{tot}, \tag{19}$$

in which $N_{\beta\alpha} = 2\Sigma_{\mathbf{k}_\parallel}n_\alpha(\mathbf{k}_\parallel)[1-n_\beta(\mathbf{k}_\parallel)]$, and where the factor 2 arises from the sum over the spin configurations. From now on, the subscripts α and β denote quantum numbers for motion along the superlattice axis, and

$$\delta V_{\beta\alpha}^{tot} = f_{\beta\alpha}^{[1]} + \delta V_{\beta\alpha}^{ind}, \tag{20}$$

with

$$f_{\beta\alpha}^{[1]} = \frac{\sum_{\mathbf{k}_\parallel}n_\alpha(\mathbf{k}_\parallel)[1-n_\beta(\mathbf{k}_\parallel)]\delta V_{\beta\alpha}^{ext}(\mathbf{k}_\parallel)}{\sum_{\mathbf{k}_\parallel}n_\alpha(\mathbf{k}_\parallel)[1-n_\beta(\mathbf{k}_\parallel)]}. \tag{21}$$

With these definitions, the equation of motion for $\overline{x}_{\beta\alpha}$ remains the same as that previously given for $x_{\beta\alpha}$, provided the factor $n_{\beta\alpha}$ is replaced by $N_{\beta\alpha}$ and the factor $\delta V_{\beta\alpha}^{ext}$ by $f_{\beta\alpha}^{[1]}$. Similar equations are obtained for spin-density fluctuations. They are obtained simply by dropping the Coulomb term and replacing the potential $U_{cd}(\mathbf{x})$ by its equivalent $U_{sd}(\mathbf{x})$ in the expression previously given for the coupling matrix U.

Returning to the question of determining the scattering cross sections, from the previous equations we now have that,

$$\langle M^\dagger(0)\rangle_\omega^{c,s} = \sum_{\alpha\beta\mathbf{k}_\parallel}[\delta V_{\beta\alpha}^{ext}(\mathbf{k}_\parallel)]^*\frac{2\hbar\omega_{\beta\alpha}2n_\alpha(\mathbf{k}_\parallel)[1-n_\beta(\mathbf{k}_\parallel)]}{\hbar^2(\omega^2 - \omega_{\beta\alpha}^2 + i\gamma_{\beta\alpha}\omega)} \times[\delta V_{\beta\alpha}^{ext}(\mathbf{k}_\parallel) + \delta V_{\beta\alpha}^{tot} - f_{\beta\alpha}^{[1]}]. \tag{22}$$

Using the definitions

$$f^{[2]}_{\beta\alpha}=2\hbar\omega_{\beta\alpha}N_{\beta\alpha}\frac{\sum_{\mathbf{k}_\parallel} n_\alpha(\mathbf{k}_\parallel)[1-n_\beta(\mathbf{k}_\parallel)]|\,\delta V^{ext}_{\beta\alpha}(\mathbf{k}_\parallel)|^2}{\sum_{\mathbf{k}_\parallel} n_\alpha(\mathbf{k}_\parallel)[1-n_\beta(\mathbf{k}_\parallel)]} \tag{23}$$

and

$$f_{\beta\alpha}=\sqrt{2\hbar\omega_{\beta\alpha}N_{\beta\alpha}}\frac{\sum_{\mathbf{k}_\parallel} n_\alpha(\mathbf{k}_\parallel)[1-n_\beta(\mathbf{k}_\parallel)]\delta V^{ext}_{\beta\alpha}(\mathbf{k}_\parallel)}{\sum_{\mathbf{k}_\parallel} n_\alpha(\mathbf{k}_\parallel)[1-n_\beta(\mathbf{k}_\parallel)]}, \tag{24}$$

we obtain the final expression

$$\langle M^\dagger(0)\rangle_\omega=\sum_{\alpha\beta}\frac{(f^{[2]}_{\beta\alpha}-|f_{\beta\alpha}|^2)}{\hbar^2(\omega^2-\omega^2_{\beta\alpha}+i\gamma_{\beta\alpha}\omega)}+\sum_{\alpha\beta} f^*_{\beta\alpha}\bar{x}_{\beta\alpha}, \tag{25}$$

where $\bar{x}_{\beta\alpha}$ are the vector components of the following matrix equation:

$$[\hbar^2\omega^2-\bar{\mathbf{U}}(\omega)+i\mathbf{\Gamma}\hbar\omega]\cdot\bar{\mathbf{x}}=\mathbf{f}. \tag{26}$$

In Eq. (26), the components of the matrix $\mathbf{\Gamma}$ are associated with the damping $\gamma_{\beta\alpha}$, and the elements of the vector $\mathbf{f}$, identified as the external pumping of our oscillator scheme, are given by Eq. (24). The expression of the matrix $\bar{\mathbf{U}}$ corresponds to that of the matrix $\mathbf{U}$ joined with the diagonal matrix composed of the square of the intersubband energies. As we assumed an external potential of the form $\delta V^{ext}_{\beta\alpha}\propto e^{iqz}$ and wave functions with periodic boundary conditions in the P periods of the multilayer system, Bloch momentum conservation assures that only excitations pairs $(i=\beta\alpha)$ and $(j=\delta\gamma)$ with wave numbers differing from each other by $q+g$ are coupled. In this case, the elements of the matrix $\bar{\mathbf{U}}$ for the charge-density mechanism, for instance, becomes

$$\bar{U}_{i,j}=\frac{\sqrt{4\hbar\omega_i\hbar\omega_jN_iN_j}}{P\bar{A}d}\left[\sum_g\frac{4\pi e^2}{\epsilon_L(\omega)}\frac{\rho^*_i(g)\rho_j(g)}{|q+g|^2}+d\int_{cell}U_{cd}(z)\Phi^*_i(z)\Phi_j(z)dz\right]+\delta_{i,j}(\omega_i)^2. \tag{27}$$

In the above equation, $\bar{A}$ is the area of the cross section normal to the superlattice (SL) axis, d is the length of the unitary cell, and $g=(2\pi/d\times\text{integer})$ is the wave number of the reciprocal lattice. For the expressions of $\Phi_j(z)$ and $\rho_j(g)=\int_{cell}e^{-i(q+g)z}\Phi_j(z)dz$, each wave function $\psi(z)$ was normalized in the unit cell.

Analyzing Eq. (25), one can see that the first term in the right-hand side has poles on the bare frequencies transitions, so it represents the contributions of the *single-particle excitations*. The second, having poles in the eigenfrequencies of the coupled system, represents the *collective* response of the electron gas.

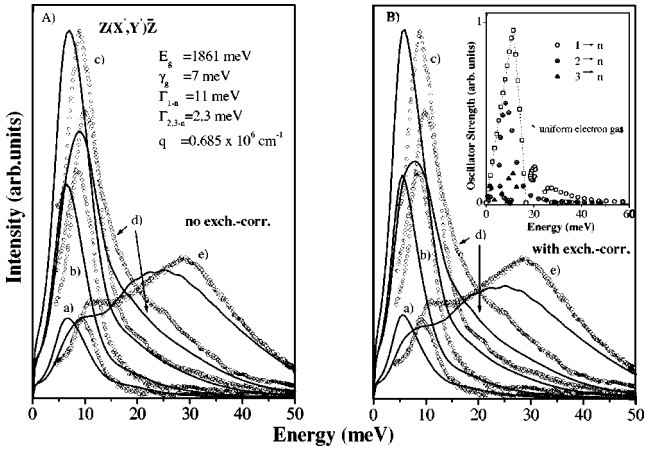

FIG. 1. Depolarized spectra of a δ-doped SL without (A) and with (B) excitonic effects. The experimental (circles) and the theoretical (lines) curves were obtained for (a) 1855 (b) 1863, (c) 1872, (d) 1880, and (e) 1897 meV incident laser energies. The inset corresponds to the oscillator strength in near resonance regime without many-body corrections. The symbols indicate the minibands from which the transitions originate. The empty squares are the results for the corresponding uniform system.

Based on the expressions of Eqs. (23) and (24), the numerator of the single-particle term is clearly expressed as a variance. This variance will be different from zero only in the extreme resonance regime where each $\mathbf{k}_\parallel$ will furnish different contributions in the energy denominators of the $\delta V^{ext}_{\beta\alpha}(\mathbf{k}_\parallel)$ due to the in-plane valence-band dispersion. Therefore the occurrence of SPE can be interpreted as the result of a deviation from the mean value of the resonant energy denominators. In near-resonance conditions, on the other hand, all the energy denominators of $\delta V^{ext}_{\beta\alpha}(\mathbf{k}_\parallel)$ are replaced by an average one with a consequent elimination of the intermediate valence-band states through completeness.[12,15] Thus the entire dependence of the external potential on $\mathbf{k}_\parallel$ is lost, canceling the single-particle term. Our result therefore consistently shows that in the near-resonance regime only collective excitations are present.

In Fig. 1 we show theoretical $(T=0\ \text{K})$ and experimental $(T=6\ \text{K})$ depolarized Raman data for various incident laser energies without [Fig. 1(A)] and with [Fig. 1(B)] exchange-correlation effects. The theoretical results are the result of the plot of the imaginary part of Eq. (2), and were fitted to the experimental ones only by adjusting the peak intensity of the curve labeled (c). The remaining intensities are automatically considered as a result of this fit. The numerical parameters used in our calculations are shown in Fig. 1(A) (a detailed discussion of their values was given in Ref. 12). The first aspect readily seen from the spectra is that their line shapes are extremely dependent on the incoming laser energy, which characterizes a resonant process. Moreover, they consist of basically two structures: one localized on the low-energy side of the spectra, and a broad one on the high-energy side. The origins of such structures are easily identi-

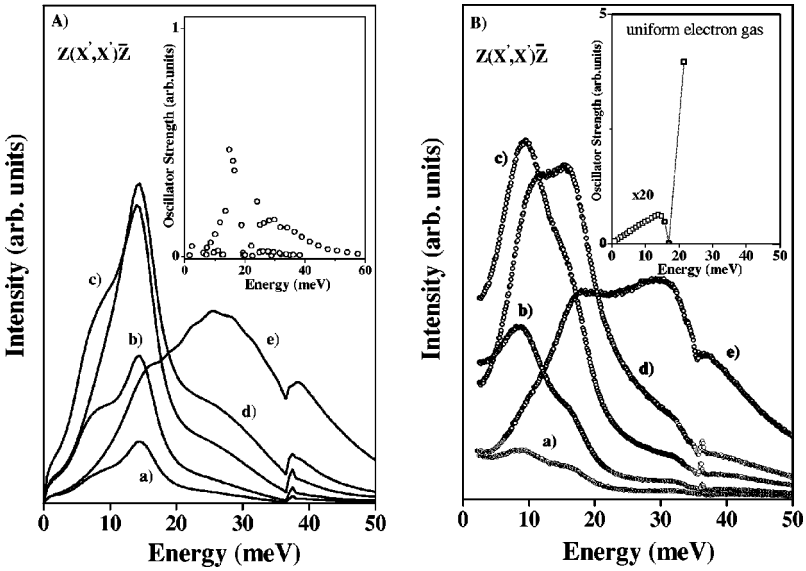

FIG. 2. Theoretical (A) and experimental (B) polarized spectra of the same sample with the same incident laser energies of Fig. 1. The inset of Fig. 2(A) indicates the spectral distribution of oscillator strength in conditions of near resonance without excitonic effects. The inset of Fig. 2(B) shows the respective distribution for the 3D gas.

fied by the spectral distribution of the oscillator strength in the near-resonance regime without exchange-correlation interaction, shown in the inset of Fig. 1(B). For clarity, in this and in the following oscillator strength plots, transitions with very small strengths were suppressed. As mentioned above, in the near-resonance regime the response function is given only by the collective term of Eq. (25). Therefore, we are led to look for eigenvalues and eigenvectors of the imaginary part of the matrix $\overline{\mathbf{x}}$. This was done through an expansion of $\overline{\mathbf{x}}$ in terms of the spectral decomposition of the matrix $\overline{\mathbf{U}}$, which is real and symmetric.[18] In near resonance and without considering many-body effects, the diagonalization is trivial (not for the polarized case, where Coulomb interactions are present), since in this case the matrix $\overline{\mathbf{U}}$ is already diagonal. As a result two sets of transitions arise. The first is a low-energy one originating from transitions of the NFE bands. As shown in the inset, these excitations are bounded by transitions of the correspondent uniform electron gas, which confers on them the status of 3D transitions. The second set, unbounded results from transitions that originate from the flat miniband states, therefore, with a 2D character. Such a set will resonate at higher laser energies due to the spatial separation of electrons and holes. Strictly speaking, what we did was to obtain a collective spectrum with zero interaction, and this, as we will see in the following, is sufficient to understand the depolarized spectra.

Returning to the extreme resonance regime, one can see that the discussion conducted above completely explains the plots of Figs. 1(A) and 1(B). Without (with) many-body effects, both plots present basically two resonant structures in the same region of the near-resonance regime. Moreover, they are obtained totally (mainly) by means of the single-particle term of Eq. (25). Therefore the connection between *single-particle* excitations and *collective unrenormalized excitations* in the depolarized spectra is straightforward. This means that the signature of excitonic effects is small in our structure and that, contrarily to what occurs in quantum wells, one cannot resolve the spin-density collective excitations that come from the second term of Eq. (25). In reality many-body effects introduced a redshift of approximately 1 meV on the curves, as well as slightly different intensities. Nevertheless, we observed that scattering intensities without many-body effects systematically furnished better results [compare, for example, the intensity of the curve (*d*) in the graphs]. We attribute this to the presence of disorder inherent in the doping process which basically produces two effects. The first is a damping of the excitations, taken in account through our phenomenological spectral function. The second, caused by in-plane potential fluctuations, results in a breakdown of momentum conservation rules. Such a breakdown would result in the loss of the in-plane coherence of the excitations, producing random fluctuations of charge which on average decrease the coupling between the intersubband transitions. Consequently the strength of the collective excitations should be reduced or, from another perspective, an enhancement of the single-particle contributions should be observed. In fact, such fluctuations invalidate the plane-waves description assumed in the normal plane of the superlattice axis. Instead, a superposition of these should be used. This would introduce additional phases into the scattering operator, resulting in a enhancement of the variance in Eq. (25). This would also explain why we previously obtained an excellent result compared with the experiment, even without taking many-body effects into account.[12] Such a transference of oscillator strengths will be more evident when we present the results of the polarized spectra. In summary, the line shapes of the depolarized spectra at extreme resonance are basically constituted by two sets of not renormalized intersubband collective excitations or, as usually termed, *single-particle excitations*.

Turning now to the polarized case, Fig. 2 presents theoretical (A) and experimental (B) Raman data where the same parameters of the SDE case are used. In spite of the difference in intensity between the structure around 15 (theory) and 16 meV (experiment) the two sets of spectra are almost identical. To reveal the nature of the spectra, the inset of Fig. 2(A) displays the correspondent oscillator strengths in the near-resonance regime, where only Coulombian interaction

is considered. Comparing the oscillator strengths of the SDE and CDE mechanisms, one immediately see that the effect of the Coulombian coupling is basically to deform the low-energy distributions, forming a pronounced peak. Pursuing the same line of reasoning, the inset of Fig. 2(B) shows the spectrum of oscillator strengths of the corresponding uniformly doped 3D gas. In this respect, a comparison of this plot with the 3D depolarized case shows that almost all the oscillator strength is taken from the region of *single-particle* excitations, and transferred to a collective excitation (plasma) mode on the high-energy side. In near resonance only this *pure* collective mode is excited. Nevertheless, the *single-particle modes* or again, the *unrenormalized collective modes*, exist, and can be excited in the extreme resonance regime. The same situation occurs with the superlattice modes in the spectral region of the NFE bands. The coupled electron system tries to transfer oscillator strength to a pure collective mode. Obviously this cannot be done in the same way as in a uniform 3D gas, since the electrons feel the effect of the confining potential. Now we have sufficient elements to understand the nature of the structures on the low-energy side of the extreme resonance polarized plots. The first, around 9 meV, corresponds to resonant 3D *single-particle* modes, and comes mainly from contributions of the single-particle term of Eq. (25). The second, around 16 meV, arises basically from a collective plasmalike excitation derived from the NFE gas. Previously such a peak was interpreted as being due to intersubband transitions.[11] Concerning the difference of theoretical and experimental intensities of the plasmalike excitation, our explanation is much the same as the one given for the depolarized case; the discrepancy is due to the breakdown of momentum conservation caused by the presence of disorder, with a consequent partial transference of oscillator strength back to the single-particle modes. Momentum conservation is assumed in the coupling matrix $\overline{\mathbf{U}}$, so it is not surprising to find a greater intensity between the theoretical and experimental collective modes in Fig. 2. Naturally the effect is larger than in the depolarized case, because the Coulombian coupling is also larger.

Considering the higher-energy structure in polarized spectra, the interpretation is almost the same as in the depolarized case, i.e., the spectra are composed of a set of resonant collective intersubband transitions with single-particle character (an exception around the LO-phonon region to be discussed below). This may be seen from comparison of the oscillator strengths of SDE's and CDE's in the region around 30 and 60 meV. Basically the form of the structure in this region remains the same. In this situation the matrix $\overline{\mathbf{U}}$ has diagonal elements much greater than the off-diagonal elements (Coulombian coupling), which are composed of superpositions of localized and delocalized states. Concluding, the polarized spectra in the extreme resonance regime is composed of basically three structures. Two, on the low- and high-energy sides, have single-particle character; the third, intermediate, structure represents a collective plasmalike mode.

Now we are going to discuss the very nature and the conditions for the appearance of single-particle excitations. Let us take a pair of subbands. As already seen, the geometry of the experiment assures us that the induced potential produces a collective oscillation within a pair of subbands in which all vertical transitions participate with equal amplitudes. Therefore, for *each* pair of subbands we have to find the in-plane response function of a set of N equally coupled and degenerate oscillators. In this case, the matrix $\overline{\mathbf{U}}$ is constituted of N degenerate diagonal elements and constant off-diagonal elements, say, $\overline{U}_{ij}=V$, where i and j represent each possible vertical pair transition. Diagonalizing such a matrix, the N-fold degenerate states will be split into an $(N-1)$-fold renormalized eigenstates lowered in energy to $E=-V$ and a single nondegenerate higher state with a renormalization energy $E=(N-1)V$. The number of states N is directly proportional to the transversal area of the sample. The opposite occurs in the coupling matrix elements, that depend of the inverse of the area [see Eq. (27)]. As a result, basically *no* energy renormalization is produced in the $(N-1)$-degenerate excitations, which establishes their *single-particle character*, although collective in their very nature. On the other hand, the single mode has a net renormalization, as the product of N and V is independent of the area. This attributes a ''pure'' *collective character* to such an eigenstate. The system in this work has sets of intersubband transitions such as just described. Nevertheless, the physics of the phenomenon is essentially the same. If this collective system is nearly equally coupled and degenerate, we will have single-particle and collective excitations. This is what is shown in the graphics of the oscillator strengths. In the 3D polarized case, for example, the condition of equal degeneracy and coupling is fulfilled. Therefore, we have well-defined collective and single-particle excitations. For the polarized δ-doped case we have the conditions reasonable satisfied in the nearly 3D part of the system, and poorly satisfied in the 2D part of the system. It is important to note that the degree of degeneracy is linked to the value of the coupling V. We remark that such a formalism is identical to the one which leads to the formation of Cooper pairs in the superconducting phase of metals.[19] There, electron pairs in the Fermi surface can scatter to states around the Fermi energy due to Pauli's exclusion principle. Those states have nearly the same energy and the same coupling, which configures the same situation as that of our oscillator scheme.

A final feature must be discussed in CDE spectra in the region of the LO phonon, where asymmetric lines appear. In conditions of *near* resonance these lines were interpreted[20] as due to the interference of the LO phonon and a continuum of single-particle transitions. Here the asymmetries appear when the frequency of the excitation matches that of the LO phonon, taken in account via lattice dielectric function. This will cause an infinite coupling which results in an absence of transitions in this region.

In summary, we provided a resonant theory for inelastic light scattering cross sections for both charge- and spin-density mechanisms in the nonuniform electron system of a periodically δ-doped superlattice that shows that the presence of SPE's are related to the existence of nearly degenerate and equally coupled transitions in the electron gas. Their very nature reside in zero renormalized collective

excitations. In near resonance the SPE oscillator strengths are transferred to the *pure* collective mode. Nevertheless, SPE's exist, and can emerge in conditions of extreme resonance regime. Our calculations suggest that an additional contribution to the single-particle cross sections may be given as a result of disorder effects which break down momentum conservation rules. In order to avoid effects of disorder and also an excess of transitions, we suggest the use of ''cleaner'' structures like quantum wells which clearly possess degenerate and equally coupled excitations in the scattering geometry proposed in this paper.

One of the authors (V. A.) gratefully acknowledges Dr. M. J. V. Bell and Dr. L. A. O. Nunes for furnishing the measurements, and CNPQ and FAPESP, Brazil, for financial support.

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