Resonant electronic Raman scattering: A BCS-like system

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In this paper we investigate the resonant intersubband Raman scattering of two-dimensional electron systems in GaAs-AlGaAs single quantum wells. Self-consistent calculations of the polarized and depolarized Raman cross sections show that the appearance of excitations at the unrenormalized single-particle energy are related to three factors: the extreme resonance regime, the existence of degeneracy in intersubband excitations of the electron gas, and, finally, degeneracy in the interactions between pairs of excitations. It is demonstrated that the physics that governs the problem is similar to the one that gives rise to the formation of the superconducting state in the BCS theory of normal metals. Comparison between experiment and theory shows an excellent agreement.

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I. INTRODUCTION

The study of quantum electron liquids in structured semiconductor systems presents a wealth of phenomena as a result of the interplay between size quantization and fundamental electron interactions. Prominent examples are as follows: (a) the Fermi edge singularity near the absorption limit of modulation-doped quantum wells [1]; (b) the fractional quantum Hall effect where strong correlations between two-dimensional electrons give rise to fractional charge particles [2]; and (c) the existence of the quantum spin Hall effect where a state of matter with different topological properties of conventional insulators is obtained in quantum wells [3]. Even the nature of the electron system may be a controversial issue, as in the case of intrasubband Raman spectra of doped quantum wires. Some authors claim that their nature reflects a Luttinger liquid model [4] and others a Fermi liquid [5].

The effects of the electronic interactions are conveniently investigated by means of inelastic light scattering or Raman scattering. Its selection rules on the incoming and outgoing light polarizations allows one to study the intersubband charge- and spin-density excitation mechanisms (CDE and SDE) [6]. The first, the polarized case, gives information of the collective charge-density excitations (CDEs) resulting from direct Coulombian as well as exchange-correlation interactions. In the latter, the depolarized case, only collective spin-density excitations (SDEs) due to exchange-correlation effects are present. Such a picture breaks down when the incoming or outgoing laser light energy matches an optical gap of the host semiconductor material. The electron gas, allied to the aforementioned collective excitations, presents also *anomalous* excitations whose energies turn out to be close to the bare electronic transitions. By this means, such excitations are called single-particle excitations (SPEs). They were first observed by Pinckzuk *et al.* [7] in three-dimensional (3D) *n*-doped GaAs with incident photon energies near the split-off edge of GaAs and subsequently in structured doped semiconductors ranging over from 2D to 1D, including inter- or intrasubband processes [8–17]. Experimentally, it is consensual that the extreme resonance regime is a necessary condition for the appearance of SPEs. Such an interpretation was confirmed theoretically by Das Sarma and Wang [12] in 2D and 1D systems by means of a resonant random-phase approximation. Further, Anjos, and Ioriatti [13], and Arantes and Anjos [14] have shown that SPEs are in fact not renormalized collective excitations. Honoring the International Year of Light (2015), we would like to revisit the highly complex theme of electronic Raman scattering which has been a source of discussions for almost 40 years [18].

The goal of this paper is to show in a clear and transparent way the physical situation of the existence of collective and single-particle excitations which, we believe, has not been completely understood, aggregating substantial information about this phenomenon. In order to accomplish this, we show that electronic Raman scattering belongs to the same class of problems such as a set of forced, coupled, and damped harmonic oscillators or BCS superconductors. Besides, we present the result that the *intersubband* SPEs are actually *coherent* collective excitations in electron systems. Our findings show that their origins rely on three conditions: first, that the laser energies should be in resonance with interband transitions (i.e., transitions involving the valence band and the conduction band states through the optical gap in question); second, that in the conduction band a set of approximately degenerate intersubband excitations exists; and, finally, that the interactions between pairs of excitations (i.e., Coulomb and/or exchange-correlation effects) should be degenerated. With the three conditions satisfied, we show that SPEs correspond to unrenormalized collective excitations. The comparison of theoretical and experimental data performed for both polarization geometries results in excellent agreement.

The experimental sample consists of a 25 nm wide, one-sided modulation-doped GaAs-$Al_{0.33}Ga_{0.67}As$ quantum well grown by molecular beam epitaxy. It has a 40 nm AlGaAs:Si doped barrier region that is separated by a 20 nm AlGaAs spacer layer from the quantum well. Our self-consistent calculations indicate that a doping layer with a total electronic density of 8.81×10^{11} cm^{-2} corresponds to a quantum well with one occupied subband with an electron density of 7.27×10^{11} cm^{-2}. The Raman spectra were obtained at 12 K using a Ti:sapphire laser and a dye laser with DCM special. These

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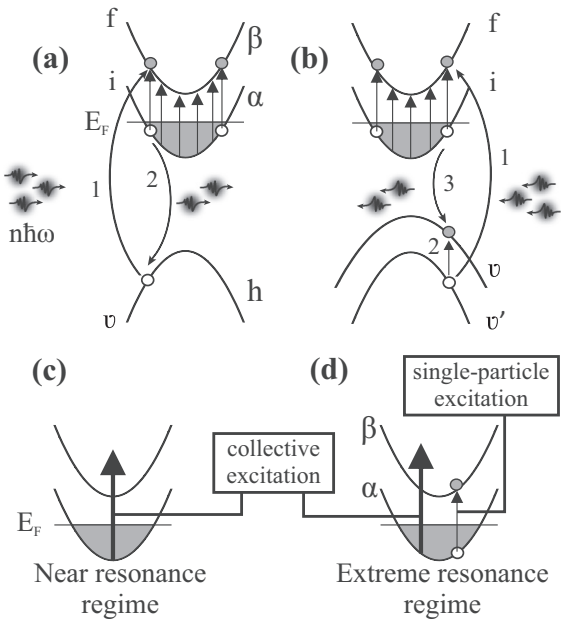

FIG. 1. Illustrations of (a) an electron gas second-order inelastic light scattering mechanism in quantum wells which involves two interband optical transitions and (b) a third-order inelastic light scattering mechanism which involves interband optical transitions (first and third step) and intersubband transitions induced by Coulomb interactions between the electron-hole pairs (or excitons) and the Fermi sea (second step). (c) Resultant excitation in the near resonance regime and (d) in the extreme resonance regime.

correspond to situations where the incoming laser energy is resonant with electronic transitions through the fundamental gap of the GaAs as well as resonant with transitions involving the GaAs split-off gap.

II. SECOND- AND THIRD-ORDER INELASTIC LIGHT SCATTERING: A COMPREHENSIVE APPROACH

The inelastic light scattering in electronic structures as understood up to now can be described via second- or third-order perturbation theory. The diagrams that address such processes may involve intrasubband [15] or intersubband transitions [17]. In this article, only cases involving intersubband transitions will be considered and, from our point of view, described as in Fig. 1.

In the first step in the second-order process [Fig. 1(a)], n photons promote N electrons to empty states in the conduction band. Holes are left in the valence band. In the second step, N electrons from the Fermi sea recombine with the holes in the valence band emitting radiation. In the third-order process, the first and third steps are analogous to the second-order one. Nevertheless, there is a second step in which a hole or electron is scattered into valence or conduction subbands, respectively. This occurs due to Coulombian coupling of electron-hole pairs or excitons with the Fermi sea via longitudinal charge-density fluctuations. In both processes, the net effect in the near resonance regime is the production of collective excitations in the conduction band, as shown Fig. 1(c). However, in the extreme resonance regime, besides the renormalized excitations, there are also unrenormalized ones, which are called single-particle excitations [Fig. 1(d)].

It is important to note that in this article we are considering a backscattering geometry situation in which the incident and scattered radiation are in the same direction. This means that intrasubband excitations will not be taken into account.

One of the claims to be supported by us is that, independent of the problem being of second order or third order, the physics involved in the process is equivalent. These statements will be put forward as premises to be proved.

As is well known, the inelastic light cross section is given by [19,20]

$$\frac{\partial^2\sigma}{\partial\Omega\partial\omega} \sim \frac{\omega_S}{\omega_L} G(\omega), \tag{1}$$

where

$$G(\omega) = \sum_F |M_{FI}|^2 \delta(E_F - E_I - \hbar\omega) \tag{2}$$

is the dynamic structure factor, and M_{FI} is the matrix element of the effective scattering operator of the light that describes the transition from the many-body state $|I\rangle$ with energy E_I to the final state $|F\rangle$ with energy E_F. $\hbar\omega_L$ and $\hbar\omega_S$ correspond to the incident (laser) and scattered photon energies. The scattering operator may be written in the Heisenberg picture as

$$M(0) = \sum_{if} \gamma_{fi} c_f^\dagger c_i. \tag{3}$$

It represents a charge-density operator with a resonant factor. The $c_f^\dagger$ (c_i) are the creation (destruction) operators of the final (initial) one-electron conduction band states. Depending on the type of process, if it is second [19] (2) or third order [20] (3), the resonant factor is written as

$$\gamma_{fi}^{(2)} = \frac{1}{m} \sum_v \frac{\langle f|\mathbf{p}\cdot\mathbf{A}_L|v\rangle\langle v|\mathbf{p}\cdot\mathbf{A}_S|i\rangle}{E_f - E_v - \hbar\omega_L} \tag{4}$$

or

$$\gamma_{fi}^{(3)} = \sum_{vv'} \frac{\langle f|\mathbf{p}\cdot\mathbf{A}_L|v'\rangle\langle v'|\widehat{U}|v\rangle\langle v|\mathbf{p}\cdot\mathbf{A}_S|i\rangle}{(E_f - E_{v'} - \hbar\omega_L)(E_i - E_v - \hbar\omega_S)}, \tag{5}$$

where $|v\rangle$ and $|v'\rangle$ represent one-electron intermediate states, $E_{i,f,v,v'}$ are state energies, $\mathbf{A}_L$ and $\mathbf{A}_S$ are vector potentials of incident (laser) and scattered light, $\mathbf{p}$ is the momentum of the electron, and $\widehat{U}$ is the Coulomb interaction potential.

In what follows, we will show that, regardless of the process order, the theoretical Raman line will be in entire accordance with the experimental one performed in the fundamental gap (third-order process) or split-off gap (second-order process) of the quantum well studied. In fact, the resonant factor will be important in Raman efficiency measurements [15,17]. In this way, we will approximate Eq. (5) by Eq. (4) considering $\widehat{U}$ constant and performing the experiment resonant with the incoming laser. Our interest is to strengthen the theory [10,13,14,21], which we believe explains the nature of SPE.

Model from a generalized self-consistent field theory

An important and very useful result to inelastic light scattering is the application of the dissipation-fluctuation theorem

[6]. Cross sections may be expressed in terms of a Fourier transform of correlation functions. It states that the dynamic structure factor $G(\omega)$ is proportional to the imaginary part of a response function. By means of the integral representation of Dirac delta function, we can express Eq. (2) as

$$G(\omega) = \frac{1}{2\pi}\int dt\, e^{i\omega t}\langle I|M^{\dagger}(t)M(0)|I\rangle, \tag{6}$$

where $M^{\dagger}(t) = e^{i\frac{H}{\hbar}t}M^{\dagger}(0)e^{-i\frac{H}{\hbar}t}$ is the time-dependent operator in the Heisenberg picture. Therefore,

$$G(\omega) \propto -\frac{\hbar}{\pi}\mathrm{Im}\langle\Psi(t)|M^{\dagger}(0)|\Psi(t)\rangle_{\omega}, \tag{7}$$

where

$$|\Psi(t)\rangle = e^{-i\frac{E_0}{\hbar}t}|0\rangle + \sum_n a_n^{(1)}(t)e^{-i\frac{E_n}{\hbar}t}|n\rangle,$$

$$a_n^{(1)}(t) = \frac{1}{i\hbar}\int_{-\infty}^{t} dt'\langle n|M(t')|0\rangle e^{i(\omega_n-\omega_0)t'}.$$

In Eq. (7), $\langle M^{\dagger}(0)\rangle_{\omega}$ is the Fourier amplitude at $-\omega$ of the expectation value in the state $|\Psi(t)\rangle$ for a time-dependent perturbation $M(0)e^{-i\omega t}$ in the linear response regime [13] and where $\lim_{\eta\to 0}\frac{1}{x+i\eta} = \frac{1}{x} - i\pi\delta(x)$ was used.

The electronic Raman scattering is a two-photon process described by the coupling between light and matter through $\mathbf{p}\cdot\mathbf{A}_L = \hat{\mathbf{e}}_L\cdot\mathbf{p}e^{i\mathbf{k}_L\cdot\mathbf{r}}$ and $\mathbf{p}\cdot\mathbf{A}_S = \hat{\mathbf{e}}_S\cdot\mathbf{p}e^{i(-\mathbf{k}_S)\cdot\mathbf{r}}$, where $\hat{\mathbf{e}}_{L(S)}$ is the incident (scattered) radiation polarizations and $\mathbf{k}_{L(S)}$ is the incident (scattered) light wave vector. The single-particle states can be written within the effective mass approach as $|b\rangle = e^{i\mathbf{k}_{\parallel}\cdot\rho}\psi_{(b)}(z)u^{(b)}(\mathbf{r})/\sqrt{A}$, where $\psi_{(b)}(z)$ are envelope functions, $u^{(b)}$ Bloch functions in the Kane model, b refers to the conduction (i,f) or valence band (v), and A is the sample area. From these considerations we rewrite Eq. (4) as

$$\gamma_{\beta\alpha}^{(2)} = \left[(\hat{\mathbf{e}}_L\cdot\hat{\mathbf{e}}_S)\delta_{\sigma_\alpha,\sigma_\beta} + i(\hat{\mathbf{e}}_L\times\hat{\mathbf{e}}_S)\langle\sigma_\alpha|\boldsymbol{\sigma}|\sigma_\beta\rangle\right] \times\frac{P_{cv}^2}{3m_0}\sum_h \frac{\langle\beta|e^{i\mathbf{k}_L\cdot\mathbf{r}}|h\rangle\langle h|e^{-i\mathbf{k}_S\cdot\mathbf{r}}|\alpha\rangle}{E_g+\epsilon_\beta+\epsilon_h-\hbar\omega_L+i\eta}, \tag{8}$$

where now, in Eq. (8), $|\alpha\rangle(|\beta\rangle,|h\rangle) = e^{i\mathbf{k}_{\parallel}\cdot\rho}\psi_{(b)}(z)/\sqrt{A}$. The spin states in the conduction band are labeled $|\sigma_\alpha\rangle$, $|\sigma_\beta\rangle$, $\boldsymbol{\sigma} = (\sigma_x,\sigma_y,\sigma_z)$ is the Pauli spin matrix vector, P_{cv} are the appropriate interband matrix elements, m_0 is the bare electron mass, E_g is an optical gap of GaAs, η is the gap damping, and $\epsilon_{\beta(h)} = \hbar\omega_{\beta(h)} + \hbar^2\mathbf{k}_{\parallel}^2/2m^*_{\beta(h)}$ are single-particle energies with $\mathbf{k}_{\parallel}$ representing 2D wave vectors. Note that Eq. (8) provides the selection rules of the inelastic light scattering by electron density fluctuations. The first term, if the incident and scattered light polarizations are parallel, corresponds to CDE. The second term will be different from zero if the two polarizations are perpendicular to each other. It corresponds to the SDE. From Eq. (8) the effective scattering operator is given by

$$M(0) = \sum_{\alpha\beta}\gamma_{\beta\alpha}^{(2)}(c_{\beta\uparrow}^{\dagger}c_{\alpha\uparrow} \pm c_{\beta\downarrow}^{\dagger}c_{\alpha\downarrow}), \tag{9}$$

$$\gamma_{\beta\alpha}^{(2)} = \frac{\left[P_{cv}^2\right]}{3m_0}\sum_h \frac{\langle\beta|e^{i\mathbf{k}_L\cdot\mathbf{r}}|h\rangle\langle h|e^{-i\mathbf{k}_S\cdot\mathbf{r}}|\alpha\rangle}{E_g+\epsilon_\beta+\epsilon_h-\hbar\omega_L+i\eta}, \tag{10}$$

where the sign + (−) identifies the CDE (SDE) and ↑ (↓) represents spin up (down) with the spin operations performed in the σ_z representation. In this work, $|\mathbf{k}_L| = |\mathbf{k}_S| = k_z$ i.e., we deal only with intersubband processes.

To calculate the response of a nonuniform electron gas submitted to the action of an external potential, we use a time-dependent local density approximation (TDLDA) [22–24] based calculation. In the case where a spin symmetry breakdown occurs, a time-dependent local spin-density approximation [25] (TDLSDA) should be used. This is the case of a discontinuity in the electron density, as, for example, when a second subband is populated in the quantum wells [26–29].

The basic idea of the self-consistent field theory approximation is to assume that a many-body system responds to a total effective field as a system of independent particles. The external potential acts on the system and induces a charge-density fluctuation as a response to the applied field. This induced fluctuation produces an induced potential. In this way, the total potential acting on the system is composed by the external and the induced potential and is given by

$$\delta V^{\mathrm{tot}} = \gamma_{\beta\alpha}^{(2)} + \delta V^{\mathrm{ind}}(\mathbf{r},t), \tag{11}$$

where

$$\delta V^{\mathrm{ind}}(\mathbf{r},t) = \int\left[\frac{e^2}{\varepsilon_l(\omega)|\mathbf{r}-\mathbf{r}'|} + U_{\mathrm{xc}}(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}')\right]\delta n(\mathbf{r}',t)d\mathbf{r}'. \tag{12}$$

The first term in Eq. (12) corresponds to the direct Coulomb interaction (the Hartree term) and the second term includes many-body exchange-correlation effects (U_{xc}). $\varepsilon_l(\omega) = \varepsilon_\infty(\omega^2-\omega_{\mathrm{LO}}^2)/(\omega^2-\omega_{\mathrm{TO}}^2)$ is the lattice dielectric function that contains the bulk GaAs frequencies of the longitudinal (ω_{LO}) and transversal (ω_{TO}) optical phonons, dielectric constant ε_∞, and where the phonon lifetime is neglected. The induced density fluctuation in the time-coordinate representation is given by $\delta n(\mathbf{r},t) = \langle\hat{\psi}^{\dagger}(\mathbf{r},t)\hat{\psi}(\mathbf{r},t)\rangle_t = \sum_{\alpha\beta}\psi_\alpha^*(\mathbf{r})\psi_\beta(\mathbf{r})\langle c_{\alpha\uparrow}^{\dagger}c_{\beta\uparrow} \pm c_{\alpha\downarrow}^{\dagger}c_{\beta\downarrow}\rangle_t$, where $\hat{\psi}^{\dagger}$ and $\hat{\psi}$ are field operators. From Eqs. (11) and (12) and solving the Heisenberg equation of motion for the expected values of the electron-hole pairs [30], we have obtained

$$\langle c_{\alpha\uparrow}^{\dagger}c_{\beta\uparrow} \pm c_{\alpha\downarrow}^{\dagger}c_{\beta\downarrow}\rangle_\omega = \frac{4\hbar\omega_{\beta\alpha}n_\alpha[1-n_\beta]}{\hbar^2\left(\omega^2-\omega_{\beta\alpha}^2+i\zeta_{\beta\alpha}\omega\right)}\delta V_{\beta\alpha}^{\mathrm{tot}}, \tag{13}$$

where

$$\delta V_{\beta\alpha}^{\mathrm{tot}} = \gamma_{\beta\alpha}^{(2)} + \sum_{\gamma\delta}\frac{4\hbar\omega_{\delta\gamma}n_\gamma[1-n_\delta]C_{\beta\alpha,\delta\gamma}}{\hbar^2\left(\omega^2-\omega_{\delta\gamma}^2+i\zeta_{\delta\gamma}\omega\right)}\delta V_{\delta\gamma}^{\mathrm{tot}}. \tag{14}$$

In Eq. (14), n_γ (n_δ) denotes the Fermi 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 of the transition, and

$$C_{\beta\alpha,\delta\gamma} = \frac{2\pi e^2}{\varepsilon_l(\omega)k_zA}\int dz\int dz'\varphi_{\beta\alpha}(z) \times[e^{-k_z|z-z'|} - U_{\mathrm{xc}}(z)\delta(z-z')]\varphi_{\delta\gamma}(z). \tag{15}$$

Equation (15) comprehends the two-dimensional Coulomb interactions [9,31] between pairs of excitations $(\beta\alpha,\delta\gamma)$ for

the case of CDEs in the conduction subband. For the case of SDEs, only the second term on the right-hand side of Eq. (15) should be considered. $U_{xc}(z)$ are functional derivatives for CDEs or SDEs obtained from the uniform electron gas data [9] and $\varphi_{\beta\alpha} = \psi_\beta(z)\psi_\alpha(z)$. Now, associating to each pair of transitions the harmonic coordinate

$$x_{\beta\alpha} = \frac{\sqrt{4\hbar\omega_{\beta\alpha}n_\alpha[1-n_\beta]}\delta V^{\rm tot}_{\beta\alpha}}{\hbar^2\left(\omega^2 - \omega^2_{\delta\gamma} + i\zeta_{\delta\gamma}\omega\right)}, \tag{16}$$

Eq. (14) can be rewritten as a matrix whose elements are given by

$$\hbar^2(\omega^2 + i\zeta_{\beta\alpha}\omega)x_{\beta\alpha} = \sum_{\delta\gamma} U_{\beta\alpha,\delta\gamma}x_{\delta\gamma} + N_{\beta\alpha}\gamma^{(2)}_{\beta\alpha}, \tag{17}$$

where

$$U_{\beta\alpha,\delta\gamma} = N_{\beta\alpha}C_{\beta\alpha,\delta\gamma}N_{\delta\gamma} + (\hbar\omega_{\beta\alpha})^2\delta_{\beta\alpha,\delta\gamma} \tag{18}$$

and $N_{\beta\alpha} = \sqrt{4\hbar\omega_{\beta\alpha}n_\alpha[1-n_\beta]}$.

In previous work, Anjos and Ioriatti [13] have shown that with an appropriate choice of the generalized coordinate it is possible to separate the SPEs and collective contributions in the inelastic light scattering cross section. SPEs will have poles on the bare frequency transitions and the collective excitations will have poles in the eigenfrequencies of the coupled system. As a consequence, in the near resonance condition, only excitations with a collective character will be present. Only under extreme resonance conditions will the SPE expression be appreciable. Owing to a canonical transformation, Eq. (17) is an analog to the equation of motion of a set of forced, damped, and coupled harmonic oscillators. The laser acts as an external potential and can be identified as an "external force." The "laser force" is expressed by $\gamma^{(2)}_{\beta\alpha}$. The electron gas oscillates due to the periodic external potential. Scattering by impurities provides a damping to the system ($\zeta_{\beta\alpha}$). Each excitation interacts with all the others, implying in a coupled system via the Coulomb interaction ($U_{\beta\alpha,\delta\gamma}$). Thus, the electronic Raman scattering can be identified as the problem of an infinite set of harmonic oscillators. In this sense, $x_{\beta\alpha}$ is the "displacement" of the oscillators. In fact, this problem can be mapped with the Newton's pendulum, as shown in Fig. 2.

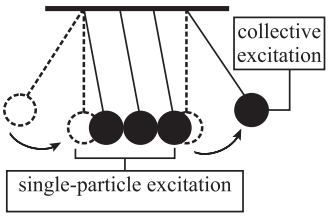

FIG. 2. Newton's pendulum. Mechanical analog of the Raman scattering in an extreme resonance regime of a 2D electron gas. The gravitational potential energy corresponds to the laser pump. When this energy is transferred to the system, two kinds of oscillations appear: (i) $N-1$ ones with small amplitudes—they are identified as single-particle excitations; and (ii) one which receives most of the energy and transferred momentum. The analog of this excitation is the two-dimensional plasmon (collective excitation: CDE or SDE).

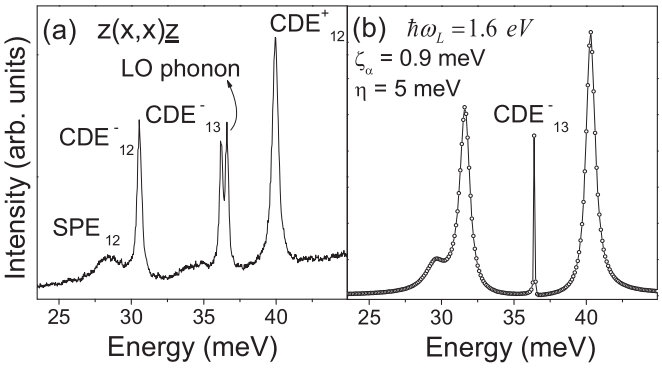

FIG. 3. (a) Experimental and (b) calculated polarized resonant Raman spectra of collective (CDE) and single-particle (SPE) excitations.

From Eqs. (15) and (18) 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 and consequently find the cross sections.

III. DISCUSSION

In Fig. 3, we present the experimental and theoretical results for the case of CDEs for an incident laser energy of 1.6 eV. The first low-energy structure appears at the unrenormalized transition energy from the first to the second subband, and as a consequence, it is identified as SPE_{12}. Then, there is a sequence of very sharp peaks. With the exception of the one at 36.6 meV [Fig. 3(a)], which is the excitation of the LO phonon of the undoped GaAs cap layer, the remaining spectral profiles represent collective CDEs. Due to the weak coupling between the excitations originating in different subbands, we coined them with the label of the subbands that contribute mainly to their formation. Moreover, the splitting of the CDEs due to the coupling with the LO phonon are considered. In this way, the structures around 30 and 40 meV are identified as CDE^-_{12} and CDE^+_{12}, respectively. An interesting structure is the sharp one near the LO phonon. Our calculations identify it as the CDE^-_{13}.

To show that this is so, Fig. 4 shows the collective spectra where the dielectric function is taken as a constant or with the fully phonon dependence considered. As the energy of the collective mode is much higher than the energy of the phonon, it couples badly with it, resulting in that the CDE^-_{13} has an energy slightly redshifted from the phonon energy. The opposite occurs with the CDE^+_{13}. From Fig. 4 one can see also that the mode CDE_{12} couples strongly with the phonon as they have comparable energies. We note that the CDE^+_{13} cannot be observed in the experiment as it is obscured by the strong luminescence signal. Nevertheless, this does not occur when the laser pump matches the split-off gap (not shown).

Figure 5 displays the results of the SDE with the same incident light energy as was given previously. In the range of energies presented, there are two structures. The sharp one is related to the collective excitation and is labeled as SDE_{12}. It is redshifted in comparison to the respective SPE due to exchange-correlation effects. The comparison between the experimental and theoretical differences in energy of the two structures is worth noting. They differ by 0.68 meV.

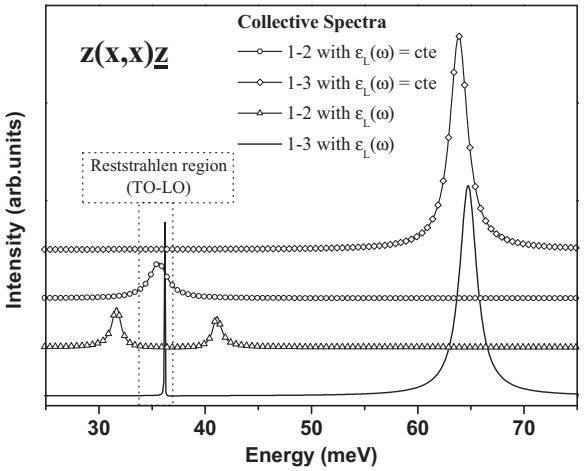

FIG. 4. Calculated polarized spectra with and without taking into account the coupling of the CDE with the optical phonons. The coupling is introduced via the frequency-dependent dielectric function.

This shows that the time-dependent LDA parametrization underestimates the value of the exchange-correlation effects, which corroborates with the results obtained by Gammon *et al.* [9]. This also helps us to understand the small discrepancies in the energy position of the collective structures originating from the first two subbands for the CDE. Concerning the discrepancy of the energy of the SPE (1.3 meV) presented in both mechanisms, it may rely on the fact that in our calculations the effect of the laser in the electronic density was not taken into account, i.e., the potential profile of the quantum well may be modified as well as a band gap renormalization may occur.

Another way to understand physically why the solution of the set of equations presented here gives rise to SPEs and CDEs is the formal correspondence between the electronic Raman scattering and the phenomenon leading to the formation of the superconducting state in the BCS theory of normal metals [32]. As stated by Cooper in his Nobel lecture [32]: "The fundamental qualitative difference between the superconducting and normal ground state wave function is produced when the

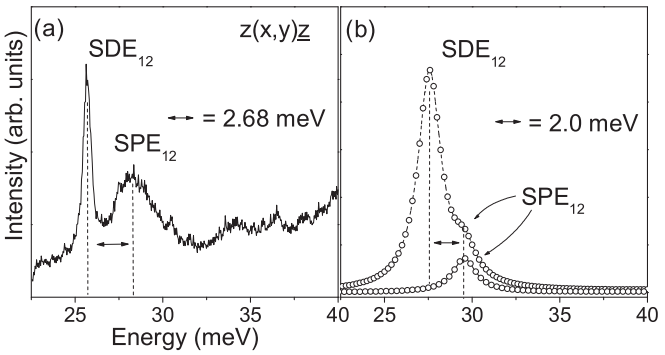

FIG. 5. (a) Experimental and (b) calculated depolarized resonant Raman spectra showing the spectral lines of the collective (SDE) and single-particle (SPE) excitations for an incident laser energy of 1.6 eV. The estimation of the exchange-correlation effects by the redshift of the SPE is also shown.

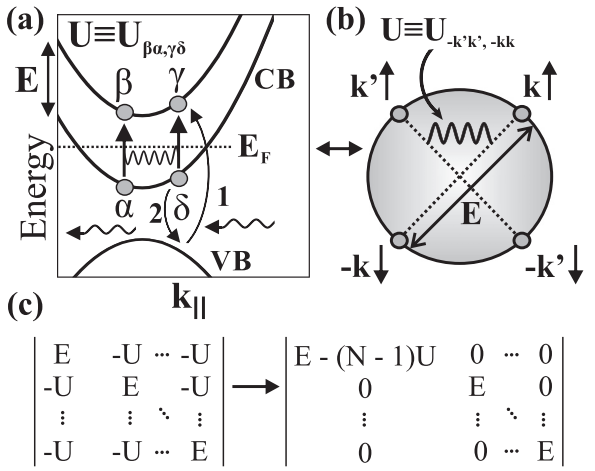

FIG. 6. Correspondence between resonant Raman scattering and the formation of the superconducting state in the BCS theory. (a) Second-order inelastic light scattering mechanism involving the split-off valence band (VB) and conduction band (CB) of GaAs. The electronic transitions $\alpha \rightarrow \beta$ ($\delta \rightarrow \gamma$) are degenerate ($E$) and coupled by a Coulomb interaction ($U_{\beta\alpha,\delta\gamma}$). The coupling is the same for any two pairs of transitions. (b) Formation of the superconducting state in the BCS theory where $\mathbf{k}$ ($\mathbf{k}'$) represent states and $\uparrow$ ($\downarrow$) spin states. The electron-phonon interaction ($U_{-\mathbf{k}'\mathbf{k}',-\mathbf{k}\mathbf{k}}$) in the Fermi surface is responsible for a degenerate coupling between the two electron pairs (Copper pairs). (c) The schematic matrix common to both situations. The diagonalization results in a single level separated from the others with energy given by $(N-1)U$ (superconducting state $\equiv$ CDE or SDE) and $(N-1)$ levels with degenerate energy (normal states of the metal $\equiv$ SPE). From this analogy, SPEs are in reality collective unrenormalized excitations.

large degeneracy of the single-particle electron levels in the normal state is removed." In normal metals, the Hamiltonian matrix which results from an attractive two-body interaction [see U in Figs. 6(b) and 6(c)] is as follows. The diagonal elements are formed by degenerated Cooper pairs with energy (E). The off-diagonal elements are constant and represent the attractive interactions mediated by phonons. Diagonalizing this matrix will result in a single level identified as the superconductor state. The other $N-1$ eigenvalues will be degenerated and are identified as normal metal states. This diagonalization represents a removal of the degeneracy. We stress that the behavior presented is independent of the physical phenomenon. Any matrix with the structure shown in Fig. 6(c) will present $N-1$ degenerate eigenvalues and one eigenvalue different from the others. Such an analogy is shown in Fig. 6 considering the excitations involving the first two subbands. In Fig. 6(a) the oscillator $\alpha \rightarrow \beta$ represents one excitation in the conduction band. The oscillators are coupled with each other via Coulomb interactions (CDE or SDE). Dealing with intersubband processes where the lateral momentum transferred by the light is zero, we assure that all the transitions along $k_{\parallel}$ will be degenerated in energy and equally coupled [see U in Figs. 6(a)–6(c)] by Eqs. (15) and (18) for any two pairs of excitations involving the two subbands. Therefore, we have a problem of N oscillators with degenerate diagonal terms (E) and all off-diagonal elements equally coupled (U), as shown in Fig. 6(c). The same occurs with the Cooper pairs mediated

by the phonon interaction at the Fermi surface of a metal in a superconductor. They are separated by $E = 2\epsilon_F$, where ϵ_F is the Fermi energy. The BCS potential ($U_{-\mathbf{k}'\mathbf{k}',-\mathbf{k}\mathbf{k}}$) corresponds to the SDE potential ($U_{\beta\alpha,\delta\gamma}$), i.e., attractive. For the CDE potential the behavior is opposite, i.e., repulsive. Therefore, for both cases, one has to diagonalize the matrix shown in Fig. 6(c).

For an $N \times N$ matrix the resulting diagonalization will furnish N states such that one of the states will have its energy renormalized by the product $(N-1)U$. This state corresponds to the coherent collective excitation of charge/spin, or, following our analogy, corresponds to the superconducting state. The remaining $(N-1)$ states are not renormalized. They are the SPEs or the normal states of the metal. The crucial point is that such excitations are revealed only when the process is in a condition of extreme resonance, the condition in which circumstances the SPEs will have a non-negligible oscillator strength [13]. We remark that the renormalization of the collective excitations are area independent. The matrix **U** depends on the inverse of the planar area of the sample as shown in Eq. (15). $N = \sum_{\mathbf{q}} 1$ is clearly area dependent. Therefore, as the collective excitations depend on the product of $N \to \infty$ and U, it results in a renormalization which is area independent or, in other words, finite.

Now, we want to address a final comment. Although the experiments shown were performed in the fundamental gap (third-order processes), the same Raman profiles are obtained when the laser pump occurs in the split-off gap (second-order processes, not shown). This reinforces all analogies and discussions performed in the present article.

IV. CONCLUSIONS

In summary, we have investigated the origins of the intersubband elementary excitations in modulation-doped quantum wells. It was shown that the physics that govern the problem is similar to the one that gives rise to the formation of the superconducting state in the BCS theory of normal superconductors and the Newton's pendulum. In particular, there are three criteria that should be fulfilled in order to experimentally observe SPE. The incident light should be in extreme resonance with the interband transitions involved, the set of intersubband excitations in the conduction band should be approximately degenerate, as well as the coupling between pairs of intersubband excitations. In such a case, it is unambiguously demonstrated that the nature of the SPE resides in unrenormalized collective excitations.

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