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Electric field dependence of 'forbidden' resonant Raman scattering in finite quantum wells: 'forbidden' electronic transitions

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Abstract. Using a finite-potential-barrier model we have analytically solved the Luttinger Hamiltonian with electric field terms included. In this way, the mixed valence band envelope functions have been shown to be a powerful tool for the analysis of several physical phenomena. In particular, the enhancement of the so-called 'forbidden' Raman scattering has been calculated for the GaAs- $Ga_{1-x}Al_xAs$ superlattices, and very good agreement with the available experimental data has been found. Calculations and experiments show that Raman scattering due to normally (in the absence of an electric field) 'forbidden' transitions can dominate that due to 'allowed' transitions in large electric fields.

Recently, the effect of external perturbations in the electronic states of quantum wells (Qws) and superlattices (SLS) has been the subject of a large number of studies. Greater attention has been devoted to external magnetic [1-3] and electric [4-9] fields and uniaxial external pressure [8, 10, 11]. Experimental data had usually been obtained by photoluminescence [12, 13], optical absorption [14], electroreflectance [15] and by photocurrent spectroscopy [16], but very few results on resonant Raman scattering (RRS) [6]. However, in this work we are interested in the explanation of some RRS results and, in particular, the case of an electric field perpendicular to the semiconductor layers, where the main results are shifts of the electronic transitions towards lower energies on increase in the electric field [14, 17]. This holds for the 'allowed' transitions (when $\Delta n =$ 0, where n is the conduction and valence subband index). Another important effect, also due to the electric field, is the appearance of 'forbidden' transitions ($\Delta n \neq 0$) [13, 18], ascribed to the mixing of the valence subband states when their intensities were increased by the applied electric field. This was attributed to a field-induced change of the heavy- and light-hole mixing and also to the opposite electron-hole polarizations which increases the probabilities of forbidden transitions.

In this work, we present a theoretical study of the 'forbidden' Raman peaks (when the electric field $F \neq 0$), which reflects the advantages of RRS in comparison with other conventional techniques. Firstly, the spatial localization of transitions, depending on the excitonic resonances, is better than that obtained by photoluminescence [6, 19]. Secondly, as has been mentioned by Tejedor *et al* [6], the fact that RRS is a single-step process with three simultaneous small denominators [20] produces a higher energy sensitivity than other physical processes such as photoluminescence, which is a 'onedenominator' process. The work mentioned above [6], including comparative measurements of RRs and photoluminescence in GaAs-Ga_{1-x}Al_xAs, provides a good example of the preceding comment, showing that the intensity of the resonance peaks has a dependence on F which is more complicated than the simple quenching observed in luminescence [9,21]. Our aim consists of the theoretical analysis of such a RRS structure, completing previous works [4, 19] where only 'allowed' transitions were included, neglecting the effects of the mixed hole wavefunctions and phonon structures which, in fact, are important in the study of the first-order Raman tensor.

The valence subband mixing has been extensively studied [22–26] through the envelope function description using the Luttinger Hamiltonian, based on the effective-mass approach. With the electric field term included, we rewrite the 4×4 Hamiltonian in the modified form

$$H = \frac{\frac{3}{2}}{\frac{1}{2}} \begin{bmatrix} a_{+} & c & b & 0 \\ c^{*} & a_{-} & 0 & -b \\ b^{*} & 0 & a_{-} & c \\ 0 & -b^{*} & c^{*} & a_{+} \end{bmatrix}$$
(1)

where

$$a \pm = E_{v} - \frac{1}{2}(\gamma_{1} \pm \gamma_{2})(k_{x}^{2} + k_{y}^{2}) - \frac{1}{2}(\gamma_{1} \mp 2\gamma_{2})k_{z}^{2} + Fz \qquad (2a)$$

$$b = \sqrt{3\gamma_3(k_x - ik_y)k_z} \tag{2b}$$

$$c = (\sqrt{3}/2)[\gamma_2(k_x^2 - k_y^2) - 2i\gamma_3 k_x k_y)$$
(2c)

and z is the electric field direction, perpendicular to the semiconductor layers. For the [0, 1, 0] direction in the k-space, the system is determined by

$$\alpha_{3/2} + \beta \partial^2 / \partial z^2 + Fz) \varphi_{3/2} + c\varphi_{-1/2} + \delta \partial \varphi_{1/2} / \partial z = 0$$

$$c\varphi_{3/2} + (\varepsilon_{-1/2} + \rho \partial^2 / \partial z^2 + Fz) \varphi_{-1/2} - \delta \partial \varphi_{-3/2} / \partial z = 0$$

$$-\delta \partial \varphi_{3/2} / \partial z + (\varepsilon_{1/2} + \rho \partial^2 / \partial z^2 + Fz) \varphi_{1/2} + c\varphi_{-3/2} = 0$$

$$\delta \partial \varphi_{-1/2} / \partial z + c\varphi_{1/2} + (\alpha_{-3/2} + \beta \partial^2 / \partial z^2 + Fz) \varphi_{-3/2} = 0$$
(3)

with four coupled differential equations, having

$$\alpha_{j} = -E_{jm} - \frac{1}{2}(\gamma_{1} + \gamma_{2})k_{y}^{2}$$

$$\varepsilon_{j'} = -E_{j'm} - \frac{1}{2}(\gamma_{1} - \gamma_{2})k_{y}^{2}$$

$$\beta = \frac{1}{2}(\gamma_{1} - 2\gamma_{2})$$

$$\rho = \frac{1}{2}(\gamma_{1} + 2\gamma_{2})$$

$$\delta = -i\sqrt{3}\gamma_{3}k_{y}$$
(4)

and E_{jm} and $E_{j'm}$ the eigenvalues to be determined (the subscripts j, j' stand for the pure n heavy- and m light-hole energy levels at $k_{\parallel} = 0$. If F = 0, the j and j' states are

degenerates) and γ_i the standard Luttinger parameters (see [25]). Setting the auxiliary functions

$$\phi_{14} = \varphi_{3/2} + \varphi_{-3/2} \qquad \phi_{41} = \varphi_{3/2} - \varphi_{-3/2}$$

$$\phi_{23} = \varphi_{-1/2} + \varphi_{12} \qquad \phi_{32} = \varphi_{-1/2} - \varphi_{1/2} \qquad (5)$$

we can rewrite the system (3) with two coupled differential equations, in which case

$$(\alpha_{j} + \beta \partial^{2}/\partial z^{2} + Fz)\phi_{14} + c\phi_{23} + \delta \partial \phi_{23}/\partial z = 0$$

(\varepsilon_{j'} + \rho \delta^{2}/\partial z^{2} + Fz)\phi_{23} + c\phi_{14} - \delta \partial \phi_{14}/\partial z = 0. (6)

The next step consists of defining the new variables

$$r_{i} = -(\beta F^{2})^{-1/3} (\alpha_{i} + Fz)$$
(7a)

$$s_i = -(\rho F^2)^{-1/3} (\varepsilon_{j'} + Fz) \tag{7b}$$

where r_i and s_i (the subscript *i* denotes inside the well) are not independent ones, but they are related by the constraint

$$s_{i} = (\beta/\rho)^{1/3} r_{i} + (-\rho F^{2})^{-1/3} (\alpha_{j} - \varepsilon_{j'} + \Delta E_{h}) = \kappa_{1} r + \kappa_{2}$$
(8)

where $\Delta E_{\rm h} = E_{jm} - E_{j'm'}$. From (6) and (7) we have that

$$\phi_{14}'' - r\phi_{14} + \lambda\phi_{23} + \omega\phi_{23}' = 0 \qquad \ddot{\phi}_{23} - s\phi_{23} + (\lambda/\kappa_1)\phi_{14} - \omega\kappa_1^2\dot{\phi}_{14} = 0 \qquad (9)$$

where the primes and dots indicate r_i and s_i derivatives, respectively, and

$$\omega = (\delta/\beta)(-\beta/F)^{1/3} \qquad \lambda = (c/\beta)(-\beta/F)^{2/3}.$$
 (10)

We know, through the different methods used up to now (e.g. TBA, Gaussian wave combination and probe series) that the character of the hybridized valence band states in the k-space varies from pure heavy-hole (or light-hole) states (at k = 0) to dominant light-like (or heavy-like) hole states, the mixing being modulated by the external field. We also know that the field-dependent states at k = 0, in the absence of hybridization, are described by Airy functions. Therefore, we try the following ansatz for ϕ_{14} and ϕ_{23} :

$$\phi_{14} = C(r,s)\mathcal{F}_i(r) + [1 - C(r,s)]\mathcal{G}_i(s)$$
(11a)

$$\phi_{23} = [1 - C(r, s)]\mathcal{F}_i(r) + C(r, s)\mathcal{G}_i(s)$$
(11b)

where $\mathcal{F}_i(r) = aA_i(r) + bB_i(r)$ and $\mathcal{G}_i(s) = cA_i(s) + dB_i(s)$ are the pure heavy and light states respectively, A_i and B_i are the standard Airy functions [27] and a, b, c and d are four arbitrary constants. The coefficient C will act like a mixing function limited from 0 to 1. Now, we substitute equations (11a) and (11b) into the coupled system (9), in which case we have

$$C'[2(\mathcal{F}'_{i} - \mathcal{G}'_{i}) - 2\omega(\mathcal{F}_{i} - \mathcal{G}_{i})] + C[(\mathcal{F}''_{i} - \mathcal{G}''_{i}) - \omega(\mathcal{F}'_{i} - \mathcal{G}'_{i}) - 2\lambda(\mathcal{F}_{i} - \mathcal{G}_{i})]$$
$$= -\mathcal{G}''_{i} + r\mathcal{G}_{i} - \lambda\mathcal{F}_{i} - \omega\mathcal{F}'_{i}$$

and

$$C'[-2(\mathscr{F}'_{i} - \mathscr{G}'_{i}) - 2\omega\kappa_{1}(\mathscr{F}_{i} - \mathscr{G}_{i})] + C[-(\mathscr{F}'_{i} - \mathscr{G}'_{i}) - \omega\kappa_{1}(\mathscr{F}'_{i} - \mathscr{G}'_{i}) + 2(\lambda/\kappa_{1})(\mathscr{F}_{i} - \mathscr{G}_{i})] = -\mathscr{F}''_{i} + (s/\kappa_{1}^{2})\mathscr{F}_{i} - (\lambda/\kappa_{1}^{3})\mathscr{G}_{i} + \omega\kappa_{1}\mathscr{G}'_{i}.$$

$$(12)$$

So, from the system (12) we get

$$C(r,s) = [C_1(r,s) + C_2(r,s)] / [C_3(r,s) + C_4(r,s)]$$
(13)

where

$$C_{1}(r,s) = [(\mathscr{F}'_{i} - \mathscr{G}'_{i}) + \omega\kappa_{1}(\mathscr{F}_{i} - \mathscr{G}_{i})][\mathscr{G}''_{i} - r\mathscr{G}_{i} + \lambda\mathscr{F}_{i} + \omega\mathscr{F}'_{i}]$$

$$C_{2}(r,s) = [(\mathscr{F}'_{i} - \mathscr{G}'_{i}) + \omega(\mathscr{F}_{i} - \mathscr{G}_{i})][\mathscr{F}''_{i} - (s/\kappa_{1}^{3})\mathscr{F}_{i} + (\lambda/\kappa_{1}^{3})\mathscr{G}_{i} - \omega\kappa_{1}\mathscr{G}'_{i}]$$

$$C_{3}(r,s) = (\mathscr{F}_{i} - \mathscr{G}_{i})\{-\omega(1 + \kappa_{1})(\mathscr{F}'_{i} - \mathscr{G}'_{i}) - \omega(\mathscr{F}_{i} - \mathscr{G}_{i})]\}$$

$$-2\lambda[(\kappa_{1}^{3} - 1)/\kappa_{1}^{3}][(\mathscr{F}'_{i} - \mathscr{G}'_{i}) - \omega(\mathscr{F}_{i} - \mathscr{G}_{i})]\}$$

$$C_{4}(r,s) = \omega(1 + \kappa_{1})(\mathscr{F}'_{i} - \mathscr{G}'_{i})^{2}.$$
(14)

The following steps consist of removing ϕ_i from ϕ_{ki} and applying the usual boundary conditions across the FQW interfaces to obtain the energy eigenvalues. The system can be numerically evaluated starting from the well known energy values in absence of valence band mixing [9]. Having obtained the energy levels we normalize the wavefunctions for several k-values near the Γ point (we are interested in the energy levels in the proximity of the Brillouin zone centre as we can see in the study of Raman scattering. The reason for such k-values is based on a negligible optical phonon momentum and on three different field intensities for a sample consisting of a 230 Å GaAs layer and a 250 Å $Ga_{0.57}Al_{0.43}As$ layer. The Luttinger parameters are the same as those in [9] and [25]. Because the Luttinger Hamiltonian does not allow state mixing at such a point, we are going to use $k_{\parallel} = 10^{-3} \text{ Å}^{-1}$, very close to the direct gap. Figure 1 shows the evolution of the mixing rate between different states obtained by computing $\langle V_i | hh \rangle$ and $\langle V_i | hh \rangle$ as external field functions at $k_{\parallel} = 10^{-3} \text{ Å}^{-1}$, where $|\text{hh}\rangle$ and $|\text{lh}\rangle$ are pure heavy- or lighthole states at k = 0. One can observe that the state mixing is nearly negligible at the chosen point. Figure 1 also shows the comparison with the mixing rate obtained by a TBA model [28]. It should be noted that, at $k \neq 0$, each heavy-hole state is mixed with all the light-hole states and vice versa; this mixture is found to be sufficiently strong only for the second hh and first lh-like states. The energies of these two states are closer than those of the other states, crossing at the field determined by $\Delta E_{\rm h} = 0$. A more realistic solution to the problem could be obtained from the Hamiltonian proposed by Bauer and Ando [3] including the electron-hole interaction, but it does not seem to be amenable to analytical treatment (the Coulomb interaction slightly diminishes the valence and conduction levels).

Our interest centres on analysing the Raman tensor and, in order to calculate this, we follow the same theoretical treatment as Tejedor and Hernández-Cabrera [21], based on the formalism of Manuel *et al* [30] and Martin [31]. In order to compare with experimental data, the back-scattering configuration and second mode of the LO phonon have been selected. The present method allows us to simulate any experimental configuration and every phonon mode and polarization by introducing the appropriate expressions from [31]. As we have said, we shall neglect the electron-hole interaction and other excitonic effects. Using this method we can introduce these effects subsequently by changing the envelope functions. We shall take into account a single FQw, the results being qualitatively identical for a multiple FQw structure. We shall perform a similar analysis to that in [30] when the external field is present. In the absence of field, the

F dependence of 'forbidden' resonant Raman scattering



Figure 1. Square of the spatial wavefunction of the three upper valence band states for three different external electric fields: (a) this work (for $k_{\parallel} = 10^{-3} \text{ Å}^{-1}$) for $F = 0 \text{ kV cm}^{-1}$ (----), 5 kV cm⁻¹(---) and 10 kV cm⁻¹(---); (b) from [28] (for $k_{\parallel} = 0$) for V = 0 V; V = 0.075 V (---) and V = 0.15 V (---).

forbidden part R_F of the Raman tensor is obtained by considering the electron-phonon Fröhlich interaction H_{e-ph} in a first-order approach, giving

$$R_{\rm F}(\omega_i) \simeq \sum_j \frac{\Delta \chi_j}{q} \left(q_t^2 \frac{m_{\rm e,j} - m_{\rm h,j}}{2\omega_0 m_{\rm e,j} m_{\rm h,j}} + q_z^2 (M_{\rm e,j} - M_{\rm h,j}) \right)$$
(15)

where ω_0 is the frequency of the optical phonon, q is the phonon wavevector with components q_t and q_z parallel and perpendicular to the layers, $m_{e,j}$ and $m_{h,j}$ are the electron and hole effective masses of the *j*th exciton and

$$M_{\mathbf{k},j} = \int \int \varphi_{\mathbf{k},j}(z) z^2 \varphi_{\mathbf{k},j}^*(z) \,\mathrm{d}z \tag{16}$$

k standing for e or h for electrons and holes, respectively. Also,

$$\Delta \chi_j(k_{\parallel}) = \chi_j(E_j, k_{\parallel}) - \chi_j(E_j - \hbar \omega_o, k_{\parallel}) \approx \int \frac{\rho(E, k) \, \mathrm{d}E}{(E - E_j - \mathrm{i}\Gamma_j)(E - E_j + \omega_o - \mathrm{i}\Gamma_j)} \tag{17}$$

where E_j and Γ_j are the energy transition and the damping factor, respectively. The terms $E - E_j - i\Gamma_j$ and $E - E_j + \omega_o - i\Gamma_j$ determine the positions of the two resonant peaks. The position of the lower-energy peak, corresponding to an n = j electron-hole exciton level, is termed the 'incoming' resonance. The higher-energy peak, on the other hand, is shifted from the incoming resonance by ω_o and it is the so-called 'outgoing' resonance. Both peaks were confirmed by optical absorption measurements. The factor $\Delta \chi_j$ determines the R_F energy dependence by the two-dimensional (2D) nature of the

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electronic states. Thus, the Raman intensity can be calculated from $\rho(E, k_{\parallel})$, the joint density of states, for a given superlattice configuration [29, 30], even without detailed knowledge of the electron-phonon interaction. Allowing optical dipole transitions only between E_{nv} and E_{mc} levels, $\rho(E, k_{\parallel})$ is a step function. Under these conditions, we can obtain equation (15) in a straightforward manner.

For low fields, the problem can be solved by the first-order perturbation approach, where H_{e-ph} and H_F (Hamiltonian due to the electric field) are not mixed and H_F does not contribute to the resonance of the phonon peaks of the Raman spectrum. In this approach, equations (16) and (17) remain valid and the effect of the electric field is reduced to the introduction of the Airy functions and phonon modes. For higher fields, second-order perturbation is required, so that a mixture of H_F and H_{e-ph} is possible, and the electric field directly affects the resonance of the phonon peaks. The second-order result is described by the first-order results corrected [20, 21] by

$$\sum_{n} \frac{-eF}{E_{n} - \omega_{i} - i\Gamma} \int \varphi_{en}(z_{e}) \varphi_{hn}(z_{h})(z_{e} - z_{h}) \varphi_{en}^{*}(z_{e}) \varphi_{hn}^{*}(z_{h}) dz_{e} dz_{h}$$
(18)

plus a similar term having a shift $\hbar \omega_o$ in the denominator.

The RRS cross section is directly related to the square of the Raman tensor and is proportional to the scattering volume V. It is convenient to refer this cross section to the volume of either the unit cell or an atom. Thus, different values for the cross section of solids may be given by different workers. This problem is circumvented by using, instead of the cross section, the scattering efficiency S defined by dropping V. This quantity, with the dimensions of an inverse length, represents the ratio of the scattered to the incident power for a unit path length within the solid. The scattering efficiency S multiplied by the scattering length and corrected for reflection losses in entering and leaving the sample gives the total observable scattered intensity. The scattering length may be limited by absorption in the solid, particularly in materials with strong optical absorption and near resonances. In this case, it is customary to use the back-scattering geometry. The effective scattering efficiency S^* is a dimensionless quantity obtained from S, corrected by the absorption coefficients and the reflectivities of the incident and scattered radiation and the plane parallel sample thickness [20, 32]. In this work, we have compared the scattering efficiency (obtained by means of the Raman tensor) with experimental RRS intensities, in arbitrary units,

There is a substantial improvement with respect to the results in [20, 32] when mixing effects are included; here we use the previously obtained mixed envelope functions and the different optical phonon modes. This means that we can obtain valuable matrix elements from any conduction and valence band with $n \neq n'$, and resonance appears not only for the $E_{nc-nb,l}$ exciton, but also for any $E_{nc-n'b,l}$. Another important effect is the k_{\parallel} -dependence of $\rho(E, k_{\parallel})$ and of the effective masses. The latter can be neglected in the Brillouin zone centre vicinity. The potential function of the electron in the electrostatic field associated with the LO modes has been taken from [31]. In recent work, Huang *et al* [33] show that such a potential function can be constructed including every phonon structure. Calculations have been done for two different k_{\parallel} -values: 0 and 10^{-3} Å⁻¹. Figure 2 shows the RRS spectra of the GaAs LO phonon. In order to compare directly with experimental measurements [6], we have chosen a logarithmic scale for the Raman intensities, an energy scale from 1.55 to 1.65 eV and the same well width used in [6] in the 24 kV cm⁻¹ RRS example. One may observe the behaviour of the $E_{14}(1c - 4hh)$,



Figure 2. RRS spectra (on a logarithmic scale) for a GaAs (230 Å)–Ga_{0.57}Al_{0.43}As (250 Å) superlattice with a 24 kV cm⁻¹ applied electric field: —, this work; \bullet , experimental data from [6].



Figure 3. RRS spectra (on a logarithmic scale) for three different applied electric fields: ______, 10 kV cm⁻¹; ______, 50 kV cm⁻¹; ______, 100 kV cm⁻¹. The excitonic peaks are labelled as follows ((i) and (o) indicate the 'in' and 'out' resonances for each exciton): 1, $E_{11}(i)$; 2, $E_{11}(o)$; 3, $E_{13}(i)$; 4, $E_{14}(i)$; 5, $E_{23}(i)$; 6, $E_{31}(i)$; 7, $E_{23}(o)$; 8 $E_{33}(i)$; 9, $E_{34}(i)$; 10, $E_{33}(o)$; 11, $E_{34}(o)$.

 $E_{21}(2c - 1hh)$ and $E_{24}(2c - 4hh)$ peaks when the field increases. All of them are 'forbidden' peaks. We must draw attention to the excellent agreement of our calculations with experiments in a 24 kV cm⁻¹ field.

Since photoluminescence allows the use of a wider energy range than RRS, we have represented in figure 3 the whole RRS curve from 1.40 to 1.65 eV (we cannot predict the actual RRS intensity without experimental measurements, but we can make provisions for the interpretation of possible mechanisms such as electron-phonon interaction through the power of the envelope functions). In order to show the influence of the 'forbidden' peaks over previous results, we have selected the same field intensities as in [9].

To sum up, we have here obtained analytical envelope functions representing superlattice valence band states under the action of an electric field. Through these functions we have calculated the diagonal elements (Fröhlich) structure of the Raman tensor in superlattices, proving the worth of such hybridized functions with respect to preceding work and comparing our results with experimental Raman measurements. Our scheme is useful in the study of the experiments related to the band mixing and can be generalized to any layered semiconductor, experimental configurations and phonon polarizations. In this way, the non-diagonal terms of the Raman tensor (deformation-potential interaction) can be investigated.

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