

Collective interband excitations in the Raman spectra of quantum wires

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A unifying theory for the collective interband excitations of the interacting electrons in a quantum wire with several occupied subbands is developed by using the bosonization method. Interband spin- and charge-density excitations for two subbands are treated in detail. The results are used to clarify the physical nature of the interband modes detected in recent Raman scattering experiments, especially the so-called single-particle excitations. [S0163-1829(99)00611-6]

In spite of considerable effort, the excitation spectra of interacting electrons in clean quantum wires with several occupied subbands have not yet been fully understood. Only very limited physical understanding of the results of the available Raman experiments—the standard tool for detecting elementary excitations¹—has been achieved by using mean-field theories as the random phase approximation (RPA) or the Hartree-Fock approach (HFA).^{2–4}

On the other hand, for only one occupied subband, results for the one-dimensional Luttinger-liquid model^{5,6} have been shown not only to be consistent with experimentally observed *intraband* charge- and spin-density excitations (CDE and SDE),^{7–11} but also to account for the so-far unexplained intraband “single-particle excitations” (“SPE”).^{12,13} Generalizations of this approach to include several subbands¹⁴ have been found to be difficult. Treating two coupled subbands led to some picture for the ground-state properties and for the energetically lowest excitations.^{15–17} Neglecting certain interaction matrix elements, *intraband* excitation spectra were obtained.^{13,18} An attempt to generalize the bosonization technique of the Luttinger-liquid model for the *interband* features in the Raman spectra of several subband quantum wires has not yet been attempted. A consistent theory that can account for both the CDE and SDE, as well as the “SPE” in the energy region of the *interband* modes presently does not exist.

In this paper, we describe a novel approach to treat the *interband* excitations of quantum wires by using approximations that are similar to the *bosonization method* used previously for the explanation of the *intraband* modes, particularly concerning the “SPE.” We describe the main results and show that they can be successfully applied to explain the positions and the heights of the peaks in the Raman spectra of state-of-the-art quantum wires. A full report of the highly nontrivial calculations and a detailed discussion of the results in comparison with the experiments will be given elsewhere.¹⁹

We consider quasi-1D confined interacting electrons with effective mass m . The one-particle spectrum consists of parabolic bands, $E_j(k) = E_j + \hbar^2 k^2 / 2m$, where E_j ($j = 1, 2, 3, \dots$) are the confinement energies. They parametrize the confinement potential. We assume that the Fermi energy

E_F is adjusted such that the Fermi velocities $v_{Fj} = \sqrt{2(E_F - E_j)/m}$ of n occupied bands are approximately the same. This can be realized in experiment. For example, for a boxlike confinement potential, the $n+1$ st subband is energetically far away from the n th band and it is possible to adjust E_F well in between. When the bands are energetically almost equidistant—the case of parabolic confinement—one can suspect that with more than the lowest n subbands occupied, the experimentally observed *interband* excitations will be dominated only by the transitions between the lowest bands, in analogy with the previous findings for the *intraband* excitations.¹² Although we have not included screening due to higher subbands, we find our results consistent with existing experiments, especially regarding the *resonant behavior* not treated previously.

As discussed below in more detail, we use an assumption that is equivalent to neglecting the interplay between *intra* and *interband* excitations. This leads to a quadratic Hamiltonian that can be diagonalized in the presence of the interaction. For the latter, we take into account Coulomb and exchange matrix elements. All the modes are found to be collective CDE's and SDE's. We calculate the cross section for Raman scattering far from and closer to resonance. For simplicity, we apply our theory to the case of only two occupied subbands, $j = 1, 2$.

We find two pairs of branches corresponding to *interband* CDE and SDE. Near $q = 0$, only two of these—one CDE and one SDE—have nonvanishing weights in the nonresonant Raman cross section. The other two are, at $q = 0$, degenerate. These contribute to the Raman cross section near resonance, when the energy of the incident photons $\hbar\omega_i$ approaches E_G , the gap energy. Additionally, the usual selection rules—CDE in the polarized, SDE in the depolarized configurations of incident and scattered photons—cannot be applied near resonance giving rise to an interpretation of the experimentally observed *interband* “SPE” in terms of combined *inter* and *intraband* SDE's.

All of our results are consistent with the existing Raman data. Furthermore, we predict new structures that suggest to reinterpret the Raman spectra of quantum wires in the region of the *interband* transitions.

In order to describe the interband excitations, we rewrite the Hamiltonian of the noninteracting electrons

$$H_0 = \sum_{j,s,k} E_j(k) c_{js}^\dagger(k) c_{js}(k), \quad (1)$$

[$c_{js}^\dagger(k)$, $c_{js}(k)$ Fermion operators, s spin quantum number, k wave number] as a quadratic form in the densities

$$\rho_{ij,s}(q) \equiv \sum_k c_{is}^\dagger(k+q) c_{js}(k) \equiv \rho_{ij,s}^{(-)}(q) + \rho_{ij,s}^{(+)}(q). \quad (2)$$

Here, $\rho^{(+)}$ and $\rho^{(-)}$ correspond to the independent branches of the spectrum with $k > 0$ and $k < 0$, respectively, with an extension of the Hilbert space to states with negative energies.⁵

Of crucial importance for the diagonalization of the Hamiltonian is that commutation relations of the form

$$[H_0, \rho_{ij,s}^{(\lambda)}(q)] \propto \rho_{ij,s}^{(\lambda)}(q) \quad (3)$$

can be constructed. From Eqs. (1) and (2) one can show that (neglecting terms $\propto q^2$)

$$[H_0, \rho_{ij,s}^{(\lambda)}(q)] = (E_i - E_j) \rho_{ij,s}^{(\lambda)}(q) + \frac{\hbar^2 q}{m} \sum_{k,\lambda=\pm} k c_{is}^{(\lambda)\dagger}(k+q) c_{js}^{(\lambda)}(k). \quad (4)$$

For the *intra*band densities $i=j$ we obtain the required commutator if we assume that in the sum on the right-hand side $k \approx \lambda k_{Fi}$, consistent with the linearization of the dispersion near k_{Fi} . We do not consider this case in the following as the results have been described previously.^{12,13}

For $i \neq j$, we replace $k \approx \lambda(k_{F1} + k_{F2})/2$ which is equivalent to neglecting multiple pair excitations between inter and intra band modes.¹⁹ The commutators of the densities are ($\lambda = \pm$)

$$[\rho_{12,s}^{(\lambda)}(q), \rho_{ii,s}^{(\lambda')}(q')] = (-1)^i \delta_{\lambda\lambda'} \delta_{ss'} \rho_{12,s}^{(\lambda)}(q+q'), \quad (5)$$

$$\begin{aligned} & [\rho_{12,s}^{(\lambda)}(q), \rho_{21,s}^{(\lambda')}(-q')] \\ &= \sum_k [c_{1s}^{(\lambda)\dagger}(k+q-q') c_{1s}^{(\lambda)}(k) - c_{2s}^{(\lambda)\dagger}(k-q') \\ & \quad \times c_{2s}^{(\lambda)}(k-q)] \delta_{\lambda\lambda'} \delta_{ss'}. \end{aligned} \quad (6)$$

Relations (5) and (6) imply that, strictly, interband and intra-band modes are *not* decoupled. However, they may be decoupled when considering expectation values in the ground state. By assuming $\langle \rho_{12,s}^{(\lambda)} \rangle = 0$ and $\langle c_{is}^{(\lambda)\dagger}(k) c_{is}^{(\lambda)}(k') \rangle = \delta_{k,k'} n_{is}^{(\lambda)}(k)$, where $n_{is}^{(\lambda)}(k)$ is the Fermion particle number, one can show the right-hand side of Eq. (6) to be $\delta_{\lambda\lambda'} \delta_{ss'} \delta_{q,q'} (L/2\pi) (k_{F1} - k_{F2} - \lambda q)$ (with L the system length) similar to the one-band case.⁵ Commutator (6) has now the form necessary for the formal description of the interband excitations as bosons.

The kinetic energy associated with the interband excitations can then be cast into the quadratic form

$$H_0 = \frac{\hbar v}{L} \sum_{q,\lambda} [\rho_{12}^{(\lambda)}(q) \rho_{21}^{(\lambda)}(-q) + \sigma_{12}^{(\lambda)}(q) \sigma_{21}^{(\lambda)}(-q)], \quad (7)$$

where $v = (v_{F1} + v_{F2})/2$, and

$$\rho_{12} = \frac{1}{\sqrt{2}} (\rho_{12,\uparrow} + \rho_{12,\downarrow}), \quad \sigma_{12} = \frac{1}{\sqrt{2}} (\rho_{12,\uparrow} - \rho_{12,\downarrow}). \quad (8)$$

For the interaction, we start from the general form

$$\sum_{ss'} \sum_{ijlm} \sum_{qkk'} \hat{V}_{ijlm}(q) c_{is}^\dagger(k+q) c_{js}^\dagger(k') c_{ls}(k'+q) c_{ms}(k).$$

The matrix elements $\hat{V}_{ijlm}(q)$ are obtained by projecting the three-dimensional Coulomb potential $V(\mathbf{r})$ on the subbands and Fourier transforming with respect to x .^{3,13} By using the Fermion operators corresponding to left and right spectral branches [cf. Eq. (4)], one can decompose the interaction into many nontrivial contributions. Important for the interband excitations are

$$H_\rho = \frac{1}{2L} \sum_{q,\lambda,\lambda'} [\hat{V} + (\delta_{\lambda\lambda'} - 1) \hat{V}_{ex}] \rho_q^{(\lambda)} \rho_{-q}^{(\lambda')}, \quad (9)$$

$$H_\sigma = - \frac{\hat{V}_{ex}}{2L} \sum_{q,\lambda \neq \lambda'} \sigma_q^{(\lambda)} \sigma_{-q}^{(\lambda')}. \quad (10)$$

Here, we defined density operators $\nu_q^{(\lambda)} = \sum_{i \neq j} \nu_{ij}^{(\lambda)}(q)$ ($\nu = \rho, \sigma$), and abbreviated the interaction matrix elements by $\hat{V} \equiv \hat{V}_{ijij}(0) \propto e^2/\epsilon_0$ ($i \neq j$), and $\hat{V}_{ex} \approx \hat{V}(2k_F)$, with $k_F = (k_{F1} + k_{F2})/2$. Only the CDE feel the Coulomb repulsion.

The total Hamiltonian for the interband excitations $H_0 + H_\rho + H_\sigma$ being quadratic in the densities, can be diagonalized separately for CDE and SDE by generalizing the method used previously by Penc and Solyom for the intra-band excitations.²⁰ One obtains two quartic equations that are biquadratic. There are two CDE's, r_\pm , and two SDE's, s_\pm corresponding to energies

$$E_\pm^\nu(q) = E_\pm^\nu(0) \left(1 \pm A_\pm^\nu \frac{\hbar^2 q^2 v^2}{E_0^2} \right). \quad (11)$$

The energy scales are given by

$$E_\pm^\rho(0) = E_0 \sqrt{1 + (1 \pm 1) V \mp V_{ex}}, \quad (12)$$

$$E_\pm^\sigma(0) = E_0 \sqrt{1 \mp V_{ex}}, \quad (13)$$

the constants by $A_\pm^\rho = [2 + (2 \pm 1)V]/2V$ and $A_\pm^\sigma = -1/V_{ex}$. The above expressions hold for not too large q and to the order $O(V_{ex}^2)$. We redefined $V_{ex} = 2\hat{V}_{ex}/\hbar v$, and $V = 2\hat{V}/\hbar v$. The complete excitation spectra, evaluated numerically by solving the secular equations are shown in Fig. 1.

Now we discuss the significance of the results for interpreting the above mentioned Raman scattering experiments on quantum wires. The interband contribution to the differential cross section is given by the dynamic auto-correlation function of the generalized density^{1,12}

$$N(q) = \sum_{i \neq j, k, \lambda, s} c_{is}^{(\lambda)\dagger}(k+q) c_{js}^{(\lambda)}(k) \frac{\Gamma}{D_i(k, q)}. \quad (14)$$

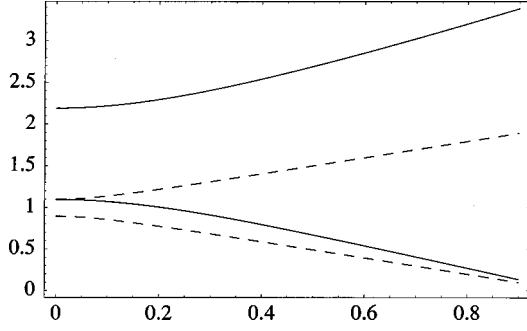


FIG. 1. The interband excitation energies E/E_0 for the two-band CDE (full lines) and SDE (dashed lines) as a function of $h\nu q/E_0$ ($V=1, V_{ex}=0.1$). Energetically highest CDE (SDE) is $r_+(s_-)$.

Here, $\Gamma = (\gamma_1 \mathbf{e}_1 \cdot \mathbf{e}_0 + i s \gamma_2 |\mathbf{e}_1 \times \mathbf{e}_0|)$ contains information about the relative polarization of incident (\mathbf{e}_1) and scattered (\mathbf{e}_0) light. The constants $\gamma_{1,2}$ are mean transition probabilities. The quantities

$$D_i(k, q) = E_G^0 - \hbar \omega_1 + E_i + \frac{\hbar^2(k+q)^2}{2m} \quad (15)$$

are the energy denominators due to the second-order perturbation theory in the electromagnetic field, with E_G^0 the energy gap of the bulk material. The crucial point for the understanding of the Raman spectra is the expansion of D_i^{-1} in powers of $(k - \lambda k_F + q/2)/[E_G^i - \hbar \omega_1 + \lambda \hbar \nu q/2]$, with $E_G^i = E_G^0 + E_i + m v^2/2$.

The lowest order term

$$N(q) = \sum_{i \neq j, \lambda} \frac{\gamma_1 (\mathbf{e}_1 \cdot \mathbf{e}_0) \rho_{ij}^{(\lambda)}(q) + i \gamma_2 |\mathbf{e}_1 \times \mathbf{e}_0| \sigma_{ij}^{(\lambda)}(q)}{E_G^i - \hbar \omega_1 + \hbar \lambda \nu q/2} \quad (16)$$

leads to the ‘‘classical selection rule,’’ namely that CDE and SDE appear only in the polarized and depolarized configurations, respectively. For photon energies such that, $|E_G^i - \hbar \omega_1| \gg \hbar \nu q$ one obtains by summing over the branch index λ in Eq. (16) that only symmetric combinations of charge and spin densities, $\nu_s = \nu_{ij}^{(+)} + \nu_{ij}^{(-)}$, ($\nu = \rho, \sigma$) are observable. Closer to resonance, $E_G^i \approx \hbar \omega_1$, the corresponding antisymmetric densities, $\nu_a = \nu_{ij}^{(+)} - \nu_{ij}^{(-)}$, in Eq. (16) become dominant.

In order to see which of the above collective modes appear in the spectra, we have to express $\nu_{s,a}$ in terms of the eigenmodes r_{\pm} and s_{\pm} . The results of the diagonalization shows that, basically, ρ_s is given by r_+ for all wave numbers. Similarly, $\sigma_a \approx s_-$. Thus, off resonance for all q , the collective charge mode r_+ will appear in the Raman spectrum in parallel polarization, while the collective spin mode s_- will be present only near resonance.

For the other densities, the dependence on the wave number is important. Near $q=0$, σ_s is dominated by s_+ , while with increasing wave number, $\sigma_s \approx s_-$. This means that in the off-resonant Raman spectra the weight of the spin modes is shifted from E_+^{σ} towards E_-^{σ} . The crossover is determined by the exchange interaction V_{ex} . For the antisymmetric charge density, we find correspondingly $\rho_a \approx r_-$ ($q \approx 0$). It

TABLE I. Collective interband modes observable in Raman scattering in lowest order.

| | Nonresonant | Resonant | |
|----------------|-------------|-----------|-------------|
| E_+^{ρ} | all q | large q | polarized |
| E_-^{ρ} | | small q | polarized |
| E_+^{σ} | small q | | depolarized |
| E_-^{σ} | large q | all q | depolarized |

becomes r_+ for larger q . As a consequence, the weight of the respective CDE peak shifts from E_-^{ρ} to E_+^{ρ} near resonance. The crossover is determined by the Coulomb repulsion energy V .

We have summarized these results in Table I, together with the polarization rules.

In addition, due to the higher order terms in the expansion of the energy denominator $D_i(k, q)$, structure will appear in the resonant spectra, which does not obey the conventional ‘‘selection rules.’’ Mixed combinations of intra and interband SDE s_- -modes become observable in *parallel* polarization. They are commonly associated in experiments with ‘‘SPE.’’ In *perpendicular* polarization only higher order contributions consisting of simultaneously propagating CDE and SDE are predicted.

Additional features may arise due to hybridization of inter and intraband modes. In Ref. 3 the effect of hybridization between different branches of the excitation spectra has been studied as a result of an asymmetry in the confinement potential. In contrast, we want to point out here, that even for symmetric confinement, interaction-induced hybridization between intraband and interband modes will occur when considering corrections to the ‘‘mean-field’’ approximation [cf. Eq. (6)]. This is of particular importance in view of the fact that signatures of the hybridization have been observed in experiment.⁷

Quantum wires in the two-band limit have been studied experimentally by Goñi *et al.*⁷ Signatures of the interband CDE’s have been detected by these measurements, which have to be assigned energetically to the modes denoted above as r_{\pm} [at energy $E_{\pm}^{\rho}(0)$].³ The structure denoted as ‘‘SPE’’ has to be associated with the above-mentioned combination of inter and intraband SDEs. In particular, the *interband* ‘‘SPE’’ appearing in the spectra at energies near and slightly above E_0 (≈ 3 meV) has to be associated with s_- . In addition, by analyzing the data in Figs. 2 and 3 of Ref. 8 ($q \approx 0$), we find full consistency with the predicted crossover behavior when approaching resonance (Table I).

More recent experiments on quantum wires with several subbands occupied^{10,11} reveal considerably rich structure concerning especially interband transitions. Signature in the polarized out-of-resonance Raman spectra of the high-energy, so-called depolarization-shifted CDE between nearest-neighbored subbands has been reported in this work. Also, signatures of the corresponding SDE’s were detected in the depolarized spectra at small wave numbers. They appear roughly at an energy about a factor of two smaller. From this, we estimate our above interaction matrix element $V \approx 1$, which corresponds roughly to the energetic interband distance. Closer to resonance, a branch of excitations (asso-

ciated in that work with ‘‘SPE’’) has been found in both polarizations corresponding to energies which increase with increasing wave number. According to our findings, these can be associated with the antisymmetric SDE s_- which contains the exchange interaction. Consistent with this interpretation, its dispersion is $\propto q^2$ but only in the region $hqv < E_0\sqrt{V_{ex}}$ [cf. Eq. (11)]. From the very small energy differences between these and the SDE s_+ observed in the depolarized spectra,¹¹ which have a very small negative curvature, we estimate V_{ex} to be at least an order of magnitude smaller than V . More detailed analysis of the experimental data, and more experiments on quantum wires with two occupied subbands, would provide clearer evidence for the two energetically low-lying excitation branches E_-^p and E_+^s , which have negative slopes as a function of q .

Comparing with previous theoretical RPA work, which applies only out-of-resonance, we find our present results consistent with the earlier findings for the CDE.³ Some investigation of intersubband SDE has been performed within a HFA.⁴ Our present theory extends these results consider-

ably and clarifies their physical meaning. We provide an analytical framework that treats both, intra and interband excitations on an equal and transparent footing. Furthermore, due to the bosonization technique, our theory allows a systematic and quantitative study of the behavior of the peak strengths in the Raman spectra *when approaching resonance*. Most importantly, our model provides insight into the physical nature of the excitations observed in the Raman spectra, and we have demonstrated that the ‘‘SPE’’ in quantum wires can be consistently understood within the framework of collective excitations.

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