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Tomonaga-Luttinger features in the resonant Raman spectra of quantum wires

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The differential cross section for resonant Raman scattering from the collective modes in a one-dimensional system of interacting electrons is calculated nonperturbatively using the bosonization method. The results indicate that resonant Raman spectroscopy is a powerful tool for studying Tomonaga-Luttinger liquid behavior in quasi-one-dimensional electron systems.

One-dimensional (1D) electron systems are important paradigms for studying elementary excitations. In these systems, electron-electron correlations can be treated exactly with the bosonization technique within the Tomonaga-Luttinger model.[1,2] Especially, one can rigorously show that the energetically lowest excitations are collective.[3] The only existing modes are charge- and spin-density excitations (CDE's and SDE's), with frequency–wave-number dispersions that are renormalized by the Coulomb repulsion and the exchange interaction, respectively.[4–7] In particular, Landau-quasiparticle excitations are absent in such non-Fermi liquids, since their lifetime is vanishingly small.

One can also calculate correlation functions, say $C(\varepsilon)$, which are experimentally observable. As a function of the variable ε, typical power-law behaviors have been predicted. Schematically,

$$C(\varepsilon)\propto\varepsilon^{\mu(g)}, \tag{1}$$

where $\mu(g)$ is in general a noninteger exponent that contains the interaction parameter g. Well known examples are photoemission and one-photon absorption.[8] Similar to the Fermi liquid, the Tomonaga-Luttinger liquid appears to be of fundamental importance in modern condensed matter theory. Therefore, directly measuring such behavior is extremely important. Unfortunately, straightforward experimental evidence is still missing, in spite of considerable efforts performed on very different materials including quasi-1D conductors and superconductors.[6] Also, predictions obtained by mapping fractional quantum Hall states to a Luttinger liquid[9] have been found very difficult to confirm, as well as Luttinger-liquid features in the dc conductance of quantum wires.[10] Only recently has evidence for Luttinger behavior been detected in the transport properties of nanotubes,[11] and in resonant tunneling through an electron island in a single-mode quantum wire.[12]

A very powerful technique for studying the electronic excitations is Raman scattering.[13–16] For energies far above the fundamental absorption edge (off-resonance), peaks in the Raman cross section corresponding to CDE's and SDE's have been identified for parallel and perpendicular polarizations of incident and scattered light, respectively. In resonant Raman scattering, for photon energies near the fundamental absorption edge, polarization-insensitive structures have been found. They have been interpreted as ''single-particle excitations'' since their dispersion corresponds roughly to that of the pair excitations of noninteracting electrons.

Especially in recent experiments on semiconductor quantum wires, these polarization-insensitive features have been the subject of detailed investigations in the regions of the intra- as well as intersubband transitions.[17–21] By applying the bosonization method to the excitations in quantum wires, the physical nature of the intrasubband SPE features has been clarified: when approaching resonance, higher-order spin-density correlation functions give rise to sharp structures in the cross section also in parallel polarization, with a dispersion law close to that of the SDE.[22]

Together with the findings at photon energies far from resonance—collective CDE's and SDE's in parallel and perpendicular polarization, respectively—the successful interpretation of the SPE structures suggests that Raman spectroscopy should be very promising for testing the Tomonaga-Luttinger model for quantum wires.

In the present paper, we demonstrate that this is indeed the case. We evaluate the differential cross section near resonance in both polarizations. We show that the strengths of the peaks associated with the higher-order SDE's behave according to power laws similar to Eq. (1) when changing the photon energy and/or the temperature. This can by no means be obtained by mean-field approaches such as the random phase approximation (RPA). Confirming our predictions experimentally would directly indicate that quantum wires are non-Fermi liquids.

In general, the electronic Hamiltonian of quantum wires consists of contributions of several subbands. For describing pair excitations with small wave numbers q, the subbands can be simplified to two branches denoted by $\lambda=\pm$ with linear dispersions near the Fermi wave numbers $\pm k_{\mathrm{F}}$ and assumed to differ only in ''confinement energies'' ϵ_j, measured from the minimum of the bulk conduction band,

$$\epsilon_j^{\lambda}(k)=E_{\mathrm{F}}+\epsilon_j+\hbar v_{\mathrm{F}}(\lambda k-k_{\mathrm{F}}), \tag{2}$$

with the wave vector component k in the direction of the wire. The electron-electron interaction contains terms that couple all of the subbands. In addition, there are matrix elements that mix only states within a given subband. They describe backward and forward scattering processes. While intraband forward scattering can be easily treated within the bosonization approach,[23] backward scattering, including the interband matrix elements, lead to severe complications, especially near $q\approx 0$ and $T=0$.[24] However, for describing Raman scattering, we are *not* interested in the behavior at extremely small q. This can be used to justify a transformation that decouples the intra- from the interband excitations. Eventually, the Hamiltonian can be written as a quadratic form in the corresponding charge and spin densities.[25] In order to demonstrate the main results of the present paper, we need only to consider the intrasubband modes, say, within the lowest subband, $j=0$.

The bosonization technique consists of replacing the standard Fermion fields $c_s^{\lambda}(k)$, associated with spin $s=\pm$ and branch λ, by boson fields $\Phi_s^{\lambda}(x,y)$. For instance,

$$c_s^{\lambda\dagger}(k+q)c_s^{\lambda}(k)=\frac{i\lambda}{2\pi Ly}\int_{-\infty}^{\infty}dx\,dy\,y^{-1}e^{i[y(k-\lambda k_{\mathrm{F}}+q/2)+xq]}\times e^{-i\Phi_s^{\lambda\dagger}(x,y)}e^{-i\Phi_s^{\lambda}(x,y)}, \tag{3}$$

$$\Phi_s^{\lambda}(x,y)\equiv\frac{4\pi\lambda}{\sqrt{2}L}\sum_{q<0}\frac{e^{-\mathrm{i}\lambda qx}}{q}\sin\left(\frac{qy}{2}\right)[\rho^{\lambda}(\lambda q)+s\sigma^{\lambda}(\lambda q)], \tag{4}$$

with $\rho^{\lambda}=\rho_{+}^{\lambda}+\rho_{-}^{\lambda}$ and $\sigma^{\lambda}=\rho_{+}^{\lambda}-\rho_{-}^{\lambda}$ the charge and spin densities, respectively, where $\rho_s^{\lambda}(q)=\Sigma_k c_s^{\lambda\dagger}(k+q)c_s^{\lambda}(k)$.

This can be used to evaluate in a closed form the Fourier transform of the correlation function

$$\chi(q,t)=\mathrm{i}\Theta(t)\langle[N^{\dagger}(q,t),N(q,0)]\rangle, \tag{5}$$

which contains the generalized density operator

$$N(q)=\sum_{k,\lambda,s}\frac{\gamma_s}{D(k,q)}c_s^{\lambda\dagger}(k+q)c_s^{\lambda}(k). \tag{6}$$

The imaginary part of the former gives the differential cross section. The quantity γ_s denotes an effective optical transition probability. For simplicity, we assume equal transition probabilities for parallel and perpendicular polarizations of incoming (polarization $\boldsymbol{e}_{\mathrm{I}}$) and outgoing (polarization $\boldsymbol{e}_{\mathrm{O}}$) light, independent of s,

$$\gamma_s=\gamma(\boldsymbol{e}_{\mathrm{I}}\cdot\boldsymbol{e}_{\mathrm{O}}+\mathrm{i}s|\boldsymbol{e}_{\mathrm{I}}\times\boldsymbol{e}_{\mathrm{O}}|). \tag{7}$$

The denominator

$$D(k,q)=E_{\mathrm{c}}(k+q)-E_{\mathrm{v}}-\hbar\omega_{\mathrm{I}} \tag{8}$$

contains the energy of incident photons $\hbar\omega_{\mathrm{I}}$, a dispersionless valence band energy E_{v}, and a single-subband conduction band $E_{\mathrm{c}}(k)=\epsilon_0^{\lambda}(k)$ [cf. Eq. (2)]. At first glance, this seems to be oversimplified in view of realistic, say $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$, quantum wires. However, it is sufficient to explain our main results, which can be straightforwardly generalized to several subbands. It is clear from Eqs. (3) and (4) that (i) $N(q)$ contains all powers of the charge- and spin-density operators and (ii) the cross section can be evaluated *nonperturbatively*.

Out of resonance, when $\hbar\omega_{\mathrm{I}}$ is much larger than the energy gap, $E_{\mathrm{g}}\equiv E_{\mathrm{c}}(0)-E_{\mathrm{v}}$, the energy denominator is approximately constant. The first and second terms in Eq. (7) give rise to peaks in the Raman spectra associated with CDE's and SDE's, respectively, when inserted into Eq. (6). This is the ‘‘classical selection rule.’’

Closer to resonance, when the photon energy approaches E_{g}, higher-order correlations become important. They violate the above-noted selection rule. This can be seen by expanding $D(k,q)^{-1}$ in powers of $\hbar v_{\mathrm{F}}\lambda(k-\lambda k_{\mathrm{F}})/(E_{\mathrm{g}}+E_{\mathrm{F}}-\hbar\omega_{\mathrm{I}})$. Especially, in parallel polarization, a peak related to a higher-order SDE has been predicted. For large photon energies, its intensity behaves as $(E_{\mathrm{g}}+E_{\mathrm{F}}-\hbar\omega_{\mathrm{I}})^{-4}$, in contrast to the $(E_{\mathrm{g}}+E_{\mathrm{F}}-\hbar\omega_{\mathrm{I}})^{-2}$ behavior predicted for the SDE in perpendicular polarization.[22] For $\hbar\omega_{\mathrm{I}}$ very close to resonance, the nonperturbative bosonization method leads to the characteristic nonanalytic dependencies on photon energy and temperature, as will be shown now.

In order to determine the correlation function (5) one needs the Heisenberg operators of the charge and spin densities in the subspace of the intraband modes of the lowest subband. For simplicity, we assume the dispersions of the charge and spin modes to be approximated by $\omega_{\rho}(q)=v_{\rho}(q)|q|$, with $v_{\rho}(q)=v_{\mathrm{F}}[(1/g_{\rho}-1)\exp(-|q|/q_{\mathrm{int}})+1]$, and $\omega_{\sigma}(q)=v_{\sigma}|q|$, with $v_{\sigma}=v_{\mathrm{F}}/g_{\sigma}$, respectively. This is justified since the experimentally relevant region corresponds to $|q|\ll q_{\mathrm{int}}$. The parameters g_{ρ} and g_{σ} describe the strengths of Coulomb and exchange interactions, respectively. Generally, $g_{\sigma}\approx 1>g_{\rho}>0$.[22] The cutoff q_{int} reflects the finite range of the repulsive interaction in the dispersion of the CDE.

By inserting Eq. (3) into Eqs. (6) and (5) one can perform the thermal average. By taking into account translational invariance along the wire, the cross section can be written in a closed form as a triple integral that can computed numerically. However, the essential physics can be extracted by the following approximation. First, we consider contributions $\chi(q,t)\propto\exp[i\omega_{\sigma}(q)t]$. These generate peaklike structures in the Raman cross section near the frequency of the SDE. We obtain

$$\mathrm{Im}\,\chi(q,\omega)\approx\delta(\omega-\omega_{\sigma})[(\boldsymbol{e}_{\mathrm{I}}\cdot\boldsymbol{e}_{\mathrm{O}})^2\mathcal{I}_1+|\boldsymbol{e}_{\mathrm{I}}\times\boldsymbol{e}_{\mathrm{O}}|^2\mathcal{I}_2], \tag{9}$$

where $\mathcal{I}_1(q,\omega_{\mathrm{I}},T)$ and $\mathcal{I}_2(q,\omega_{\mathrm{I}},T)$ are the peaks strength in parallel and perpendicular polarization, respectively. Correspondingly, when selecting $\chi(q,t)\propto\exp[\mathrm{i}\omega_{\rho}(q)t]$ (since v_{ρ} approximately constant for small q), we get

$$\mathrm{Im}\,\chi(q,\omega)\approx\delta(\omega-\omega_{\rho})(\boldsymbol{e}_{\mathrm{I}}\cdot\boldsymbol{e}_{\mathrm{O}})^2\mathcal{I}_0. \tag{10}$$

Equations (9) and (10) constitute our first general, important result: while the SDE gives rise to a peaklike structure in both polarizations, the CDE appears as a peak only in parallel and *not* in perpendicular configuration, even near resonance. This can be most easily seen by considering the lowest-order term that is proportional to $\sigma\rho$ in perpendicular polarization and this *cannot* give rise to a peak at the frequency of the CDE.[22]

Furthermore, one can prove a general theorem, namely, that the terms in a power-law expansion of $N(q)$ that contribute near the frequency of the CDE in perpendicular polarization (i) contain at least one spin density operator, and (ii) consist always of a product of an *odd* number of spin-density operators multiplied by a product of charge-density operators. Terms of this kind will not produce a peak in the corresponding cross section at the frequency of the CDE. When calculating the correlator, there is always a residual pair of spin-density operators, $\sigma(t)\sigma(0)$, which remains time-dependent and destroys the coherence of the associated CDE terms. This annihilates any spurious CDE peak in the cross section.

In the following, we consider only the structures related to the SDE. Similar results can be extracted for $\mathcal{I}_0$, Eq. (10). The intensities of the former are (for $g_\sigma=1$)

$$\mathcal{I}_1(q,\omega_{\rm I},T)=\frac{Lq\gamma^2}{12(\hbar v_{\rm F})^2}\left[\frac{q^2}{2}+\left(\frac{\pi}{\beta\hbar v_\sigma}\right)^2\right]\left|\frac{d\mathcal{S}}{dQ}\right|^2, \tag{11}$$

with $\beta^{-1}=k_{\rm B}T$ ($k_{\rm B}$ is the Boltzmann constant), and

$$\mathcal{I}_2(q,\omega_{\rm I},T)=\frac{Lq\gamma^2}{(\hbar v_{\rm F})^2}|\mathcal{S}(Q,T)|^2. \tag{12}$$

The integral

$$\mathcal{S}(Q,T)=\int_0^\infty dy\, e^{iQy}F(y) \tag{13}$$

depends on the ''reduced photon wave number'' $Q=(E_{\rm g}+E_{\rm F}-\hbar\omega_{\rm I}+\hbar v_{\rm F}q/2)/\hbar v_{\rm F}$. The function

$$F(y)=\frac{1}{(1+q_{\rm int}^2y^2)^\mu}\left[\frac{\beta\hbar v_\sigma}{\pi y}\sinh\left(\frac{\pi y}{\beta\hbar v_\sigma}\right)\right]^{-1/2}\times\left[\frac{\beta\hbar v_\rho}{\pi y}\sinh\left(\frac{\pi y}{\beta\hbar v_\rho}\right)\right]^{-2\mu-1/2} \tag{14}$$

contains the exponent

$$\mu=(g_\rho+1/g_\rho-2)/8 \tag{15}$$

typical for Tomonaga-Luttinger correlation functions.[6–8] Remarkably, it contains the parameter of the charge interaction, though it describes SDE-related features. This indicates that physically the higher-order SDE's in parallel configuration are ''dressed'' by CDE's.

Equations (11) to (14) constitute our second important prediction: the dependencies of the intensities of the SDE peaks in resonant Raman scattering on the energy of incident photons and/or the temperature in parallel and perpendicular polarizations are governed by nonrational exponents that are characteristic for the Tomonaga-Luttinger liquid and contain the strength of the repulsive interaction between the electrons.

Let us identify in more detail the parameter regions where this ''Tomonaga-Luttinger behavior'' can be expected to be most clearly detectable. There are three characteristic wave numbers: the inverse of the range of the interaction $q_{\rm int}$, the wave number of the elementary excitation q, and the wave number corresponding to the temperature, $q_\beta=1/\beta\hbar v_{\rm F}$. We assume $q_{\rm int}\gg q_\beta>q$ since below $q_{\rm int}$ we expect the most important interaction-induced effects. We consider interactions of experimental relevance which correspond to $g_\rho>g_0$ with g_0 such that $\mu(g_0)=1/2$, i.e., $g_0\approx0.2$ and $g_\sigma=1$.

For $Q>q_{\rm int}$, $\mathcal{I}_n\propto(q_{\rm int}/Q)^{4/n}$ ($n=1,2$) we are still far from resonance.[22] For $q_{\rm int}>Q$ we are near resonance. As long as $Q>q_\beta$ the dependence on the temperature of the integral $\mathcal{S}(Q,T)$ does not affect the result,

$$\mathcal{I}_n\propto\left(\frac{q_{\rm int}}{Q}\right)^{4(1/n-\mu)}. \tag{16}$$

For $q_\beta>Q$ one obtains a dependence on temperature

$$\mathcal{I}_n\propto\left(\frac{q_{\rm int}\hbar v_{\rm F}}{k_{\rm B}T}\right)^{4(1/n-\mu)}. \tag{17}$$

For all of interaction parameters discussed, the ratio $\mathcal{I}_1/\mathcal{I}_2$ behaves independently of the interaction as β^2 or Q^{-2}, though the energy and temperature dependencies contain the interaction parameter. For $g_\rho<g_0$, the behavior is similar, but cannot be treated analytically.

Traditionally, inelastic light scattering of interacting electrons has been analyzed within RPA. This seems to work well for the nonresonant case as it gives for quantum wires similar results for the dispersion as the present approach. In RPA, the cross section is related to the electronic polarizability. By expanding into a power series in terms of the interaction, one finds that the first term, often denoted as $\Pi_2(q)$, which is independent of the interaction, contains an energy denominator $D(k,q)^{-2}$. This is the only contribution in *perpendicular* polarization.[16] It gives a peak at the frequency of the pair excitations of the noninteracting electrons, $v_{\rm F}|q|$.

In *parallel* polarization, and far from resonance, Π_2 can be absorbed into a geometrical series in the interaction. This yields only one pole—corresponding to a peak in the Raman cross section—at the frequency of the CDE. When approaching resonance, such that the k dependence of $D(k,q)$ has to be taken into account, Π_2 contributes separately,[15] and produces an additional pole at the energy of the noninteracting electron-hole pair. The corresponding peak intensity, however, does not show any nonanalytical power-law behavior.

In the Tomonaga-Luttinger approach, the low-energy excitations are collective. There are no modes at the energies of noninteracting electron-hole pairs. The energetically lowest excitations are SDE's with energy $\hbar v_\sigma|q|$.

In principle, the renormalization of excitation frequencies could be achieved within a self-consistent perturbational approach, generalized to include exchange interaction, *but* taking into account consistently exchange self-energy and *in addition* exchange vertex corrections in Π_2. However, in order to obtain the above nonanalytical behavior of the intensity of SDE peaks when approaching resonance, these corrections should include the Coulomb interaction *to infinite order*, as seen in Eq. (15). Thus, in perturbative language, self-energy and vertex corrections are responsible for the non-analytic power-law behaviors of the spectra close to resonance. This does not contradict the well-known result that far from resonance the sum of the two terms exactly cancel due to Ward identities[6,26] Indeed, the latter cannot be applied in the presence of k-dependent vertices.

Currently, the existence of the SPE in the experiments on quantum wires are well established and consistent with our above reported findings. Unfortunately, experimental data do not include systematic studies of the dependencies of the peak intensities on photon energy and/or temperature. Such studies, however, should be highly desirable since they are expected to contribute to solving a fundamental question of modern many-body physics, namely, in how far electronic correlations beyond mean fields are important for describing *correctly* the low-energy CDE's and SDE's of clean quasi-1D electron systems.

In summary, we have pointed out that resonant Raman scattering is a powerful tool for experimentally investigating Tomonaga-Luttinger behavior in quasi-1D electron systems. We have shown that, when approaching resonance, SDE-induced peaks appear in both parallel and perpendicular polarizations of incident and scattered photons. In contrast, the CDE cannot produce peaks in perpendicular polarization. We have quantitatively determined the nonanalytical behavior of the intensity of the peaks in the resonant Raman spectra that are due to SDE's. The measurement of these nonanalytical dependencies on photon energy and/or temperature predicted above would be decisive for discovering fundamental non-Fermi liquid behavior in clean quantum wires and represents major challenges for experiment.

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