Where is the Luttinger Liquid in One-Dimensional Semiconductor Quantum Wire Structures?

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(Received 10 April 2000)

We present the theoretical basis for analyzing resonant Raman scattering experiments in one-dimensional systems described by the Luttinger-liquid fixed point. We make experimentally testable predictions for distinguishing Luttinger liquids from the Fermi liquid and argue that presently available quantum wire systems are not in the regime where Luttinger-liquid effects are important.

PACS numbers: 71.10.Pm, 73.20.Dx, 73.20.Mf, 78.30.–j

It is theoretically well established [1–3] that a one-dimensional interacting electron system (1DES) is not a Fermi liquid (FL). Unlike a Fermi liquid, the interacting 1DES has neither sharp fermionic quasiparticle excitations nor a discontinuity in the electron momentum distribution function. The elementary excitations are charge e, spin zero bosons and spin $1/2$ charge 0 "semions" (fractional statistics objects), and the fermion is a composite of these. Interacting 1DES have been generically termed Luttinger liquids (LL) [2] and have been the subject of extensive theoretical study over the last 40 years and particularly over the last decade. Despite the intense theoretical interest, there have been few convincing experimental demonstrations of the predicted LL behavior in real 1DES. The power-law density of states observed in tunneling into edges of quantized Hall systems [4] have been interpreted in terms of the theoretically expected "chiral Luttinger liquid" behavior of edge states. The origin of the differences between the observed and expected exponents is presently an area of active inquiry. Photoemission experiments on Mott insulating oxides have been interpreted in terms of the "holon" and "spinon" excitations of a charged Luttinger liquid [5].

A 1DES which is of particular interest both for fundamental physics and for technology is the system formed in GaAs-based semiconductor quantum wire (QWR) structures. Modern materials growth and fabrication techniques have produced nearly ideal 1DES in which the electron may move freely only along the length of the wire. The transverse motion is quantized with the quantum 1D subbands separated by several meV. It is possible to have low enough carrier densities so that at low temperatures only the lowest 1D subband is occupied by electrons. Such GaAs QWR based 1DES should be ideal systems for the study of interacting electrons in one dimension because they are free from complications arising from band structure, lattice effects, and crossovers to three-dimensional behavior which often make interpretations of experimental data difficult in more traditional 1DES based on organic compounds.

It is surprising, therefore, that no definitive LL behavior has been reported in GaAs QWR systems, and, in fact, the 1D Fermi gas/liquid model seems to "work" operationally very well in describing and explaining the observed 1DES experimental properties in GaAs QWR [6,7]. Part of the reason for the apparent absence of the expected LL behavior is undoubtedly the fact that in weakly interacting 1DES, *at finite temperatures and in the presence of impurity scattering,* the actual quantitative difference between a LL and a FL is not large [7], although the conceptual difference between the two is huge. A more fundamental issue is that the differences between a Luttinger liquid and a Fermi liquid are most obvious in the one-electron spectrum, while the difference in particle-hole properties are much less pronounced. This perhaps accounts for the fact that one of the most important probes of QWR structures, resonant inelastic light scattering or Raman scattering spectroscopy (RRS) [6,8], which probes the particle-hole spectrum, has not yet observed any definitive indications of LL behavior in QWR systems.

In RRS experiments, light is absorbed at one frequency and reemitted at another, creating one or more particle-hole pairs. In the so-called polarized geometry with the incident and outgoing photons having the same polarization (so that no spin is transferred to the QWR), RRS experiments in GaAs QWRs consistently [6,8] show two peaks which indeed look qualitatively very similar [9] to the spectra for the corresponding 2D and 3D systems. In these higher dimensional systems, the two peaks have a clear and generally accepted Fermi liquid interpretation [9]. The higher energy peak is associated with the plasmon or charge density excitation (CDE), a collective density excitation of the electron gas, and the lower energy spectral peak is associated with incoherent particle-hole pair excitations (SPE). In the QWR materials, the lower energy peak occurs at an (approximate) excitation energy of $\nu \sim q v_F$, where q is the excitation momentum and v_F is the 1D Fermi velocity obtained from the band structure of the QWR. An interpretation of the lower peak as an SPE contribution seems therefore natural [9]. However, there is a strong theoretical objection to this interpretation: in a one-dimensional system there is asymptotic spin-charge separation; in the long-wavelength, low temperature limit, the charge excitations live at the plasmon frequency, and cannot contribute

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to excitations at the SPE energy. The signal observed in this q, ν range must be due to the chargeless spin excitations of the LL; in particular, it is possible to combine two $S = 1/2$ excitations into a $S = 0$ object, creation of which is allowed by the Raman selection rules, as first noted by Schulz [10]. Irrelevant operators arising from band curvature lead to a coupling, which vanishes in the long-wavelength limit, between the $S = 1/2$ excitations and the charge degrees of freedom, but this effect causes negligible corrections to our results (or those of other workers [10,11]).

Sassetti and Kramer (SK) presented a qualitative theory of this effect [11]. They showed that although the leading contribution to the RRS matrix element corresponds to coupling the light to the electron density operator, there is a subleading term (which becomes more important near resonance) which may be interpreted as a coupling of light to the energy density fluctuations of the electrons in the QWR. These have a contribution from the spin excitations, which qualitatively explains the data. The SK theory did not calculate the spectral weights of the RRS peaks, and breaks down too close to resonance. Most importantly, the SK calculation is logically inconsistent, because it uses an expression for the RRS matrix element which is correct only if the conduction band is a FL not a LL. Thus SK uses FL matrix elements but LL excitations.

In this paper we present an essentially complete treatment of RRS in a one-dimensional electron gas. We obtain a precise expression for the energy transferred to the QWR in a RRS experiment, valid at all values of the difference of the energy from resonance, and evaluate it quantitatively in several experimentally relevant limits. We show which features of the data contain information about the LL exponents, obtain expressions for the relative amplitudes of the SPE and CDE peaks, determine line shapes, and discuss the LL to FL crossover. Most importantly, we derive the correct RRS matrix element, and show how it is affected by LL correlations.

Resonant Raman scattering is a two-photon process in which a photon is absorbed, transferring an electron from the valence (V) band to the conduction (c) band and a photon is emitted, transferring an electron from the conduction band back to the valence band. We assume that the valence band is initially filled, and assume there is no excitonic interaction between conduction and valence band states. The excited valence hole is then described by a single-particle Hamiltonian, which we write as H_V (note this need not have quantization of transverse momentum), while the conduction band is described by some interacting Hamiltonian which we denote H_{LL}. We denote the photon absorption and emission by $P_{1,2}$, respectively. The RRS process is described by the following Hamiltonian:

$$H = H_V + H_{LL} + \hat{P}_1 + \hat{P}_2, \tag{1}$$

where the photon-in (P_1) and photon-out (P_2) terms are

$$\hat{P}_1 = e^{-i(\Omega+\nu/2)t} \sum_{p,s} c^\dagger_{p+q/2,s}(t) v_{p,s}(t) + \text{H.c.} \tag{2}$$

$$\hat{P}_2 = e^{i(\Omega-\nu/2)t} \sum_{p,s} v^\dagger_{p,s}(t) c_{p-q/2,s}(t) + \text{H.c.} \tag{3}$$

with c and v the annihilation operators for electrons in conduction and valence band states, respectively. Note that the operator $v^\dagger_{p,\sigma}$ creates an eigenstate of H_V with energy E^V_p while the $c^\dagger_{p,\sigma}$ operators do *not* create eigenstates of H_{LL}. The absorbed (emitted) photon energy and momentum are set $\Omega \pm \nu/2$ and $\pm q/2$, respectively.

We now use the standard methods of time-dependent perturbation theory to calculate the amplitude, $a_n(t_0)$, for the system at some time t_0 to be in some excited state $|n\rangle$ of QWR, but with no holes in the valence band. We assume the system is in its ground state at $t = 0$. Our neglect of any excitonic interaction between conduction and valence band simplifies the calculation, and we obtain

$$a_n(t_0) = \frac{1}{L} \sum_{r,s} \int dR\, e^{-iqR} \int_0^{t_0} dT\, e^{i\nu T} \langle n|\hat{O}_{rs}(R,T)|0\rangle \tag{4}$$

with

$$\hat{O}_{rs}(R,T) = \int dx \int_0^T dt\, \phi(x,t) \psi_{rs}(R + x/2, T + t/2) \times \psi^\dagger_{rs}(R - x/2, T - t/2), \tag{5}$$

where r and s are band and spin indices (± 1), and

$$\phi(x,t) = e^{i\Omega t} \sum_p e^{i(E^V_p t - px)}. \tag{6}$$

Equations (4) and (5) are our fundamental new results: they show that the RRS process acts to create a particle-hole pair at a spatial separation x and temporal separation t. These are determined by the average photon frequency Ω and the valence-band properties encoded in E^V_p. Further, if interactions are present in the conduction band, the states created by $\psi^\dagger$ and by ψ are not eigenstates of H_{LL} and therefore the matrix element is itself modified by interactions. SK [10] and also [9] considered a matrix element with $t = 0$ (but with x dependence) and neglected the renormalization of the matrix element.

Equations (4) and (5) may be substantially simplified in the limit of greatest physical interest. We linearize the valence-band energy about the conduction band Fermi momentum, writing $E^V = -\Delta - v^V(rp - p_F)$ for branch r and define $\omega_R = \Omega - \Delta$ as the photon frequency with respect to the resonance energy, Δ. The p integral gives $\delta(x + v^V t)$. Finally we write the conduction band operators in terms of the bosons which create eigenstates of H_{LL}, and normal order in the boson basis, obtaining

$$\hat{O}_{rs}(R,T) = L \int_0^T dt\, e^{i\omega_R t} G^c_{rs}(-rv^V t, t) :e^{i\Phi_{rs,\rho}(R,-rv^V t;T,t)}:: e^{i\Phi_{rs,\sigma}(R,-rv^V t;T,t)}:, \tag{7}$$

where

$$\Phi_{rs,\rho}(R,x;T,t) = 2\sum_{p>0} e^{-\epsilon p/2}\sqrt{\frac{\pi}{pL}}\{-\sinh\theta_\rho \sin[p(rx - v_\rho t)/2][b^\dagger_{-rp}e^{ip(rR+v_\rho T)} + \text{H.c.}] + \cosh\theta_\rho \sin[p(rx - v_\rho t)/2][b^\dagger_{rp}e^{-ip(rR-v_\rho T)} + \text{H.c.}]\}, \quad (8)$$

$$\Phi_{rs,\sigma}(R,x;T,t) = 2s\sum_{p>0} e^{-\epsilon p/2}\sqrt{\frac{\pi}{pL}}\{\sin[p(rx - v^c_F t)/2][\sigma^\dagger_{rp}e^{-ip(rR-v^c_F T)} + \text{H.c.}]\}. \quad (9)$$

Here b^+ and σ^+ create charge and spin excitations, respectively, and $v_\rho = v^c_F e^{-2\theta_\rho}$ is the plasmon velocity, where the exponent $e^{-2\theta_\rho} = \sqrt{1 + 2g/\pi v^c_F}$ is defined for the short-ranged interaction, g. $G^c(-rv^V t, t)$ is the exact conduction band Green's function at spatial separation $-rv^V t$, and temporal separation t. We have assumed that the interactions are negligible in the spin sector and therefore the spin excitation velocity is just the Fermi velocity. As long as v^V, the valence-band velocity at the conduction band p_F is different from the spin and charge velocities of Luttinger liquid, G^c is a decaying function of t. In the noninteracting case, $G^c \sim 1/t$; interaction leads to a faster decay: $G^c \sim 1/t^{1+\alpha}$ with the LL exponent $\alpha = \sinh^2\theta_\rho > 0$ for short-ranged interactions; G^c decays faster with the physically relevant long-ranged interactions. This faster decay of G^c is the mathematical expression of the renormalization of the RRS vertex by the interactions which produce the Luttinger liquid behavior. As we will now show, it has important consequences for various aspects of the RRS spectra, and, in particular, for the dependence of the CDE and SPE energies on the difference of the average photon energy from resonance.

We defer to a subsequent paper a full evaluation of the RRS correlation function, which is computationally demanding and not very illuminating, and present here the results of expanding Eq. (7) in terms of boson operators. The essential point is that if the combination of $e^{i\omega_R t}G^c(-rv^V t, t)$ decays rapidly as t increases (large ω_R, i.e., off-resonance, or large α, i.e., strong interaction), then the t integral is dominated by small times and an expression in power of bosons is rapidly convergent. We will show below that the first order term, one-boson result, gives the main contribution to CDE spectrum and dominates the whole off-resonance spectrum and the second order term, two-boson (spinon) result, gives the peak at SPE energy near resonance, but it still has relatively small weight as compared to the first order CDE.

Expanding the exponentials, keeping only the one-boson term and integrating explicitly, gives the one-boson transition rate as a delta function at $\nu = qv_\rho$ with the spectral weight ($\alpha < 1$)

$$W_1 = \frac{2L\Gamma^2(-\alpha)}{qv^2_\rho}\left|\left(\frac{\omega_R - \omega_q}{E_0}\right)^\alpha - \left(\frac{\omega_R + \omega_q}{E_0}\right)^\alpha\right|^2, \quad (10)$$

where $\omega_q \equiv qv_\rho/2$, neglecting v^V for simplicity. E_0 is the energy scale depending on the interaction range and roughly of the order of Fermi energy, E^c_F. For $\omega_q \ll |\omega_R|$, $W_1 \propto |\omega_R|^{2\alpha-2}$, while for $\omega_R = 0$, $W_1 \propto \sin^2(\pi\alpha/2)$. Thus LL effects enter the CDE portion (one boson) of the spectrum in two ways (for short-ranged interaction): first, far from resonance, it changes the frequency dependence of spectral weight from ω_R^{-2}, the noninteracting result, to $\omega_R^{-2+2\alpha}$ (note that all other higher order bosonic contribution decays much faster; this confirms the validity of the bosonic expansion we mentioned above). Second, on resonance ($\omega_R = 0$) it changes the value to be nonzero due to finite interaction strength.

To second order, two new effects appear. In the density spectrum, branch mixing processes lead to a continuum absorption beginning at the CDE threshold, $\nu = qv_\rho$. In addition, an $S = 0$ combination of spin excitations may be excited via two spinons $\langle\sigma\sigma, \sigma\sigma\rangle$ (note that there is *no* first order contribution in spin channel due to the selection rule of RRS in the polarized spectroscopy), and gives the so-called SPE mode at $\nu = qv^c_F$.

In Fig. 1, we show the spectrum from one and two bosons for different resonance energy. One sees that (i) the overall spectral weights decay very fast off resonance, and (ii) the SPE peak is generated at $\omega \sim 0.2E^c_F$ by the two-boson contribution near resonance. But as compared with the CDE peak at plasmon energy (about $0.57E^c_F$) the SPE peak is still very small. This striking result arises from the fact that the contribution of one spin boson in the first order is forbidden by the specific selection rule of polarization in depolarized RRS spectroscopy. (iii) At the higher energy side above the CDE peak, there is some continuum structure which is not shown in the range of Fig. 1. This continuum is from the interaction between different branches of charge bosons, and is not interesting because it goes to zero near the plasmon energy and its higher energy behavior is off the experimentally measurable region. (iv) When including three or higher order boson contribution (not shown in this paper), we will see the mixture of charge boson and spin boson in a form like $\langle\sigma\sigma\rho, \rho\sigma\sigma\rangle$, which will appear at the energy between qv^c_F and qv_ρ, plasmon energy, as a continuum structure. A detailed analysis shows that this is relatively small and has no special structure compared to the first two order result. While Fig. 1 is for a specific value of α (= 0.3) we show in Fig. 2 the calculated charge boson and spin boson RRS spectral weights at resonance and away from resonance. In general, the LL theory predicts much smaller spectral weight for the lower energy SPE mode than the FL theory [9] (which neglected matrix element renormalization) at resonance. This is particularly true since our best estimate for the Luttinger

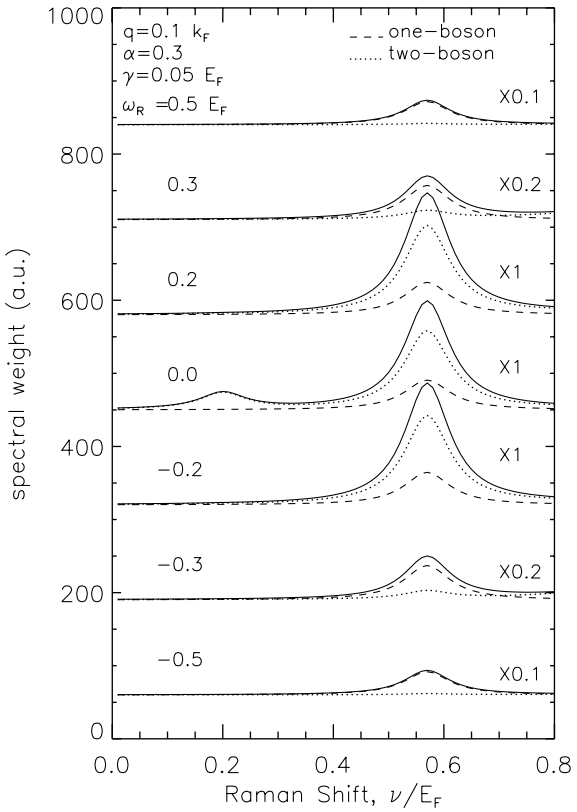

FIG. 1. Calculated polarized RRS spectra for various resonance condition, ω_R. One- and two-boson contributions have been plotted separately in order to show their relative contributions (see text). A finite broadening γ has been used to depict the results. Note that the overall spectral weights decrease dramatically off-resonance, as indicated by the individual scale factors on the right side of each plot.

exponent of the experimental system [6] (obtained from the CDE energy dispersion) is $\alpha \sim 0.4$.

As compared with the experimental result, which shows possible comparable spectral weight of SPE with CDE [6], we find that the LL theory result induced by resonance effects does not explain the experimental results quantitatively, even though we could recover the SPE peak through the coupling of two spinons in LL [11]. The perturbative results presented here are confirmed by a nonperturbative spectral weight analysis along similar lines to that leading to Eq. (15) of Ref. [10] and will be presented in a future paper. Therefore, in contrast to Ref. [10,11], we believe that the existing experimental results [6] are not proof of LL behavior but are in the high energy crossover regime where, in fact, a FL description may be more appropriate for the RRS data than the LL description which is an asymptotic low energy description. This explains the spectacular quantitative success of the FL RRS theory developed in Ref. [9].

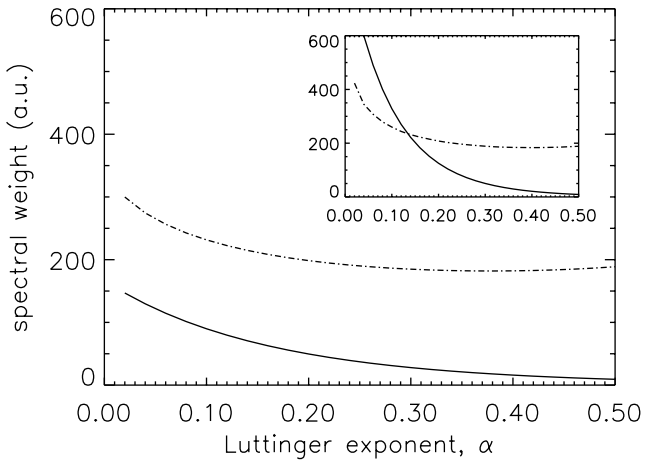

FIG. 2. Spectral weights for the low energy (solid curve) and the high energy (dashed) RRS peaks in the LL theory plotted as a function of the Luttinger exponent α: $\omega_R = 0$ (main); 0.1 (inset). When $|\omega_R|$ larger than 0.1 the low energy ("SPE") weights are always much smaller than the high energy (CDE) weights over the whole range of α.

In conclusion, we provide the correct LL theory for the RRS spectra calculation, and obtain some meaningful and interesting results to study the possible origin of LL features in the RRS spectra of 1D QWR systems. We also develop a useful bosonic expansion method to study the two-particle correlation function. Finally, we find that the LL theory cannot quantitatively explain the experimental data [6] most likely because the RRS experiments are not in the asymptotic low energy LL regime.

This work was supported by the U.S.-ONR, the U.S.-ARO, and the NSF (A. J. M.).

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