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Inelastic light scattering by spin-density, charge-density, and single-particle excitations in GaAs quantum wires

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We report the observations of inelastic light scattering by the elementary excitations of the one-dimensional electron gas. The quantum wires are fabricated from a modulation-doped single $GaAs/Al_xGa_{1-x}As$ quantum well by electron-beam lithography and shallow electron-cyclotron-resonance reactive-ion etching. In spectra of intersubband excitations we observe a clear separation of spin-density and charge-density collective modes. The splitting associated with the lateral potential of the wires is also seen in transitions to states of the first excited level of the parent quantum well. Determinations of electron-electron interactions in intersubband excitations of the one-dimensional electron gas show large corrections due to the exchange terms (or vertex corrections) that represent the excitonic binding of the particle-hole pairs.

Narrow quantum wires of width less than 100 nm are currently obtained by state-of-the-art semiconductor nanofabrication technologies.[1,2] The behavior of free carriers in such nanostructures presents fundamental characteristics due to their motional confinement to one dimension. Electron-electron interactions in one dimension are expected to change drastically both elementary excitations and screening, and the question of whether the one-dimensional (1D) electron system is better described as a Fermi liquid or as a Luttinger liquid is still open.[3,4] Intersubband transitions and elementary excitations of the 1D electron gas have been studied by far-infrared (FIR) optical absorption[5,6] and inelastic light scattering.[7,8] FIR measurements can detect charge-density excitations (CDE's), if they carry an electrical dipole moment.[5,9,10] In contrast, inelastic light scattering experiments are specially well suited to observe CDE's,[8,11] spin-density excitations (SDE's),[8,12] and single-particle excitations (SPE's),[7,8,12] which can be identified by simple polarization selection rules.[13]

The observation of spin-density excitations that are well separated from the single-particle continuum reveals the strength of vertex corrections (or excitonic shifts) due to exchange terms of the Coulomb interaction in 2D systems.[14,15] These terms represent the excitonic binding of the particle-hole pair states in intersubband excitations.[16] The strength of the exchange interaction depends strongly on the confinement of the electron gas and on its density. In two dimensions and at very low densities exchange effects are predominant over the Hartree terms,[17] whereas in the high-density regime with more than one subband occupied by electrons, SDE's are suppressed.[18] Recent light scattering work in semiconductor quantum wires found indications for an enhancement of excitonic shifts in one dimension.[11,12] However, in these experiments well-defined intersubband spin-density excitations were not observed in the absence of an external magnetic field and the strength of vertex corrections in one dimension could not be assessed.

In this paper we report observations of excitations of the one-dimensional electron gas in GaAs quantum wires by resonant inelastic light scattering. The free electrons are close to the quantum limit, because only two 1D subbands are occupied. Extremely sharp 1D intersubband SDE's and CDE's are identified by clear polarization selection rules. The 1D subband spacings are obtained from single-particle intersubband excitations that occur in the spectra. We also observe excitations due to transitions from the occupied 1D subbands to the states of the first excited level of the parent quantum well. The splitting of such quasi-2D-intersubband excitations is consistent with the subband spacings obtained in spectra of 1D intersubband excitations. These observations allow quantitative determinations of shifts due to electron-electron interactions in the energies of 1D intersubband excitations. Our results indicate that in the 1D quantum limit vertex corrections are about as significant as the depolarization shifts due to direct terms of the Coulomb interaction. Furthermore, intrasubband SDE's are clearly observed, which show the 1D characteristic line shape, and display a linear dispersion as a function of the wave vector parallel to the wires. From the dispersion we obtain the Fermi energy of the system.

The samples were prepared following the method described in Ref. 19, starting from a modulation-doped $GaAs/Al_xGa_{1-x}As$ single quantum well (SQW) grown by molecular-beam epitaxy. The GaAs well was 250 Å wide and the $Al_{0.16}Ga_{0.84}As$ barriers extend 500 and 850 Å below and above, respectively. The top barrier was delta-doped with two layers of silicon at 250 and 800 Å from the well. The mobility and carrier density of the as-grown SQW at 4.2 K were 2.4×10^6 $cm^2/V\,s$ and 3.12×10^{11} cm^{-2}. The first confined electron state (E_0) and the second one (E_1) are separated by 21 meV. Lines with 2000-Å period and 700 Å width were patterned by electron-beam lithography into a polymethylmethacrylate (PMMA) resist layer covering the sample. The resulting PMMA stripes acted as a mask for the subsequent etching process in an electron-cyclotron-resonance reactive ion etcher. The etching parameters were chosen to remove the Si doping in the gaps between the lines without violating the SQW itself. Lines of charged Si donors remaining in the top barrier produce the periodic electrostatic potential that confines the carriers into a type-II lateral superlattice [Fig. 1(a)]. This was established by photoluminescence measurements that show large redshifts due to spatially indirect electron-hole recombination.[19,20] The confining potential was found to be modified by photoexcited carriers under strong illumination. From these measurements we infer a lateral potential modulation of approximately 10 meV under the condition of the reported experiments. The results presented below show that the quantum wires have a Fermi energy of 5.0 ± 0.2 meV obtained from the dispersion of intrasubband SDE's and a subband spacing of 3.0 ± 0.5 meV. This results in largely two occupied subbands with densities $n_0 = 6.1 \pm 0.1 \times 10^5$ cm^{-1} and $n_1 = 4.3 \pm 0.4 \times 10^5$ cm^{-1}.

Inelastic light scattering experiments were performed at 1.7 K using a tunable dye laser in resonance with excitonic transitions between the first confined hole state and the second quasi-2D electron state [E_1 in Fig. 1(b)]. The incident laser power was kept below 1 W/cm^2. Spectra were measured in the conventional back-scattering geometry. Wave vectors along the wires were applied by tilting the sample normal with respect to the laser beam by an angle Θ around an axis perpendicular to the wires. The resulting wave-vector component is given by $q = (4\pi/\lambda)\sin\Theta$, where λ is the wavelength of the incident light. Incident and scattered light were linearly polarized parallel (H) or perpendicular (V) to the wires. Spectra taken with parallel polarizations of incident and scattered beams exhibit CDE's, whereas SDE's are only

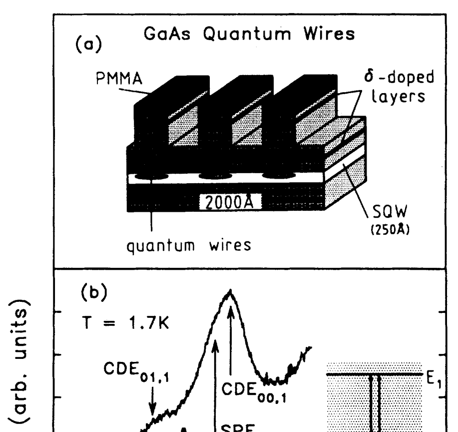

FIG. 1. (a) Schematic structure of the quantum wires. The lateral carrier confinement is achieved by shallow dry etching through the openings of the PMMA resist mask. (b) Polarized and depolarized light scattering spectra corresponding to 2D-like transitions to the first excited subband due to the confinement in the growth direction, as shown in the inset.

active in spectra with crossed polarizations (depolarized spectra). The SPE's show up in both types of spectra under extreme resonance conditions.

1D confinement effects due to the lateral potential are revealed by additional features that occur in the spectra in the energy range of quasi-2D intersubband transitions. Both polarized and depolarized spectra show a main peak [see Fig. 1(b)], labeled $\mathrm{CDE}_{00,1}$ and $\mathrm{SDE}_{00,1}$, at energies close to the ones of CDE's and SDE's of the parent quantum well. Therefore, they are assigned to the collective excitations associated with transitions between the lowest state of the quantum well, which is split into 1D subbands $E_{00}, E_{01}, \ldots$ by the wire potential, and the first excited 2D subband E_1.[21] The small peak on the high-energy side of the $\mathrm{SDE}_{00,1}$, energetically coincident with the low-energy shoulder of the $\mathrm{CDE}_{00,1}$ peak, is assigned to the corresponding single-particle transition ($\mathrm{SPE}_{00,1}$) in analogy to the 2D case.[14] The interesting features are the ones labeled $\mathrm{CDE}_{01,1}$ and $\mathrm{SDE}_{01,1}$ which are absent in the spectra of the as-grown SQW. They are assigned to charge-density and spin-density excitations corresponding to transitions between the first excited 1D subband and E_1. From these observations and assuming that the corresponding single-particle transition $\mathrm{SPE}_{01,1}$ has an energy in between both collective modes, we infer for the 1D subband spacing a value of 2–3 meV.

We consider now the observations of 1D intersubband and intrasubband excitations. Figures 2 and 3 show sets of depolarized (VH) and polarized (VV) spectra measured at the smallest wave vector (for $\Theta = 0$ and $k \leq 2 \times 10^{-4}$ cm^{-1}). The features that remain stationary are due to inelastic light scattering. The dependence of spectral intensities and line shape on incident photon enegy is due to strong resonant enhancements when photon energies overlap with excitonic transitions of the GaAs quantum well.[22] In Figs. 2 and 3 there is also a weak luminescence that moves through the spectra as a broad (~2 meV) background. This signal is clearly seen

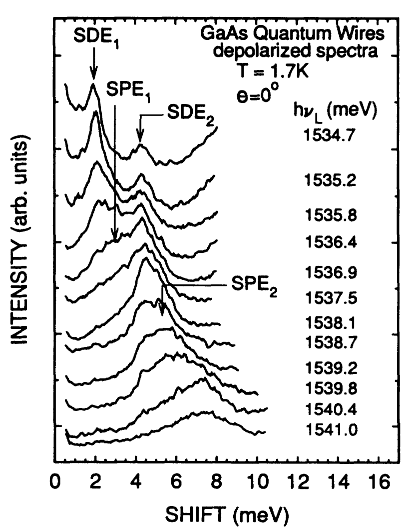

FIG. 2. Depolarized resonant light scattering spectra of 1D intersubband excitations for different incident photon energies.

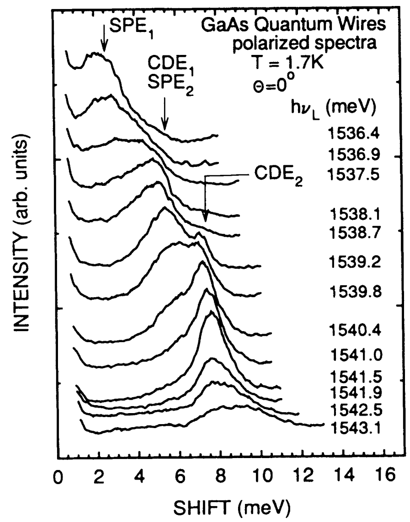

FIG. 3. Polarized resonant light scattering spectra of 1D intersubband excitations for different incident photon energies.

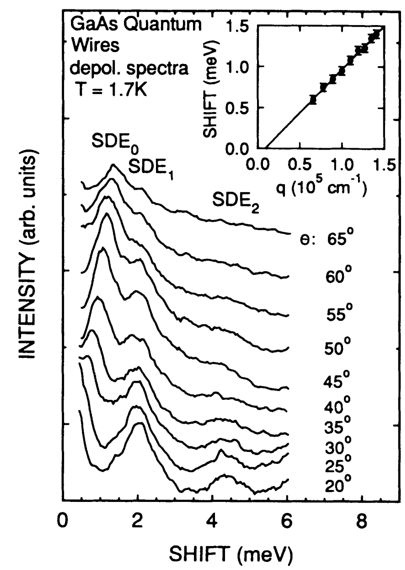

FIG. 4. Depolarized light scattering spectra of the quantum wire sample for different tilt angles Θ, corresponding to different wave vectors q along the wires. The inset shows the wave-vector dispersion of the intrasubband spin-density excitation (SDE_0).

in the off-resonance spectra measured at the highest incident photon energies. The prominent peaks (labeled SDE_1 and SDE_2) in the depolarized spectra at 2.0 and 4.4 meV are interpreted as 1D intersubband SDE's and the one at 7.4 meV in polarized spectra as 1D intersubband CDE's. The subscript $\Delta n = 1, 2$ labels the change in 1D subband quantum number. These are the collective excitations associated with the lowest-energy transitions between states confined by the lateral potential of the quantum wires.

The 1D intersubband single-particle transitions are also observed in both types of spectra. For crossed polarizations (see Fig. 2) the SPE_1 and SPE_2 appear as the small shoulders at the high-energy side of their corresponding SDE's around 3.0 and 5.5 meV, respectively. Two weak features also occur at the same energies but in the polarized spectra of Fig. 3. Again a subband spacing of $\approx$3 meV is obtained from these measurements in good agreement with the light scattering data in the energy range of quasi-2D transitions. We note that the collective excitation CDE_1 is expected close to the energy of the SPE_2 and they probably merge together in the 5.5-meV peak. One possible reason for the weakness of the CDE_1 feature compared to CDE_2 might be parity conservation, because for a symmetric potential a two-photon process like inelastic light scattering is only sensitive to even transitions ($\Delta n = 2, 4, \ldots$).

We have measured in the same sample both collective CDE's and SDE's as well as intersubband single-particle excitations of the 1D electron gas which allows the determination of the direct and exchange terms of the Coulomb interaction. The energies of the collective modes can be expressed in terms of the single-particle transition energy ($E_{\rm SPE}$), the depolarization shift ($W_{\rm dep}$), and excitonic shift ($W_{\rm xc}$) as follows: $(E_{\rm CDE})^2 = (E_{\rm SPE})^2 + (W_{\rm dep})^2 - (W_{\rm xc})^2$ and $(E_{\rm SDE})^2 = (E_{\rm SPE})^2 - (W_{\rm xc})^2$. With the experimental values found for the $\Delta n = 2$ transition ($E_{\rm SDE} = 4.4$ meV, $E_{\rm SPE} = 5.5$ meV, $E_{\rm CDE} = 7.4$ meV), one obtains a ratio $(W_{\rm xc}/W_{\rm dep}) =$ 0.55. This value is similar to the one of the $\Delta n = 1$ transition (0.44) but it is slightly lower than the ratio of 0.68 extracted from the 2D intersubband transition energies measured in the unpatterned region of the sample, in contrast with other results on quantum wires.[8]

The different strength of the vertex corrections measured here as compared to the results of Ref. 8 might be related to the actual shape of the wire potential in each case. Given the low electron densities in the quantum wires, we consider a parabolic potential model. This connects the measured potential modulation and subband spacing consistently, as the leading term of a Fourier expansion of the lateral potential with curvature given by the subband spacing yields a modulation height of about 11 meV. Thus there are three well-confined 1D subbands of which two are densely populated. In this case 1D intersubband excitations are well defined and appear as very sharp peaks (down to 0.7 meV) compared to the broader spectral features observed in much shallower wires of Ref. 8, where the second 1D state is more extended due to the superlattice effect. On the other hand, the larger occupation of the second 1D subband here is the cause for the reduction of vertex corrections attributed to the cancellation between contributions from electrons in both 1D subbands.[18]

We have also investigated dependence of the spectral features on wave vector along the wires. Figure 4 shows depolarized spectra for different tilt angles. At low angles the intersubband SDE_1 and SDE_2 are clearly observed. These excitations are dispersionless and their linewidths exhibit the characteristic behavior with increasing wave vector due to Landau damping in the SPE continuum. Starting at 25° a sharp feature is apparent at low energies. This peak, which is only observed in depolarized spectra, is assigned to 1D intrasubband spin-density excitations (labeled SDE_0). It displays a linear dispersion (see inset to Fig. 4) that extrapolates to -0.1 meV at zero wave vector. This is a measure of intrasubband Coulomb vertex corrections. Similar shifts (0.25 meV) due to exchange interactions[15] have been observed in 2D samples with comparable average electron densities. The Fermi energy of the 1D system can be extracted from the slope of the SDE_0 because this triplet spin mode is expected at energies[4] $\hbar\omega = \hbar q v_F$, where v_F is the Fermi velocity. In this way we have obtained $E_F = 5.0 \pm 0.2$ meV. Finally, we point out that the observation of intrasubband SDE speaks for a small smearing of the Fermi surface giving additional evidence that the 1D electron gas behaves as a Fermi liquid.[12,23]

In conclusion, we have observed various excitations of electrons in GaAs quantum wires by inelastic light scattering. The formation of 1D subbands due to the lateral confinement is established by a structure in the former 2D intersubband transitions as well as in 1D intersubband and intrasubband excitations. We achieved measurements of 1D intersubband spin-density excitations. This shows that all basic excitations of a 1D electron system can be observed in inelastic light scattering experiments. The energetic position of the intersubband and intrasubband SDE is shifted from the SPE by vertex corrections due to exchange and correlation electron-electron interactions. This effect is of similar strength as in the case of two-dimensional electron systems in $GaAs/Al_xGa_{1-x}As$ quantum wells. However, further experiments will have to reveal the exact size of this contribution in its dependency on the number of occupied subbands.

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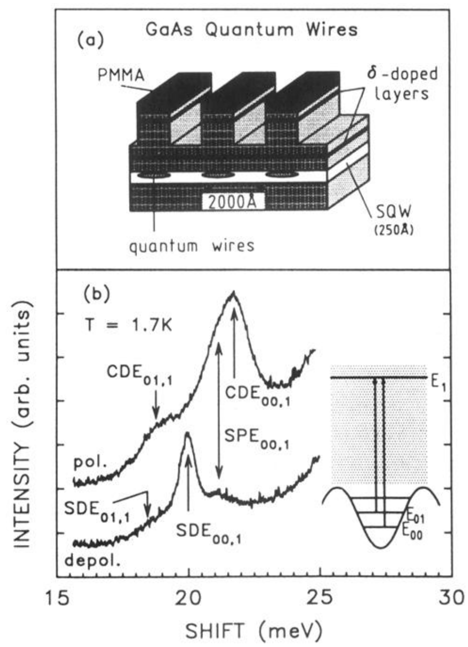

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