Single-particle excitations and many-particle interactions in quantum wires and dots

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We have investigated electronic excitations in GaAs-Al_xGa_{1-x}As quantum wires, and dots by resonance Raman spectroscopy. We find for quantum dots and quantum wires three types of excitations; single-particle, spin-density, and charge-density excitations (SPE's, SDE's, and CDE's, respectively). Our experiments show that SPE's are enhanced under conditions of extreme resonance, which indicates that energy-density fluctuations are responsible for the excitation of unscreened SPE's. The high intensity of the SPE's allows a detailed analysis of collective effects in the zero- and one-dimensional electron systems. $\left[S0163-1829(96)51848-5 \right]$

In high-quality modulation-doped $GaAs-Al_xGa_{1-x}As$ heterostructures nearly perfect two-dimensional (2D) electron systems with quantized energy levels can be realized. With sophisticated technology it is possible to produce lateral structures and reduce the dimensionality even further and fabricate one-dimensional $(1D)$ quantum wires and zero $dimensional$ (OD) quantum dots, the latter with a totally discrete energy spectrum.¹ These systems have attracted great interest since they allow, in specially tailored structures, a detailed investigation of quantum and, in particular, manybody effects. Raman spectroscopy is an ideal tool to study these many-body effects since, with polarization-dependent selection rules, one can map out different many-body effects.² For a parallel polarization of incoming and Ramanscattered photons one measures so called charge-density ex $citations (CDE's), which reflect the collective excited states$ of the electron system and which are renormalized due to the dynamic charge and spin redistribution. In highly symmetric systems CDE's are the only excitations observed in direct far-infrared absorption.¹ In Raman spectroscopy one can additionally measure in crossed polarization collective spindensity excitations, which are affected only by the dynamic spin redistribution. It was very surprising with respect to the so far accepted theory³ that Pinczuk *et al.*⁴ observed on high-quality two-dimensional systems, in addition to CDE's and SDE's, an excitation that exhibited all features of an unrenormalized single-particle excitation. This excitation is observed in both polarizations. These results have been confirmed by a number of authors⁵—we see it in our 2D samples, too—however, the scattering mechanism was so far not clear. There are also very interesting Raman experiments on quantum wires and dots.^{6–13} Strenz *et al.* reported on SPE in quantum wires and observed SPE in dots.¹¹ Onedimensional electron systems which are nearly in the 1D quantum limit have been investigated by Schmeller *et al.*⁹ They found that the relative energy renormalizations of the 1D intersubband SDE and CDE with respect to the simultaneously observed corresponding SPE are quite similar to those found for 2D systems.⁴ However, the mechanism and conditions for the excitation and strength of collective effects

for quantum dots and wires are even less clear. Very recently, an interesting paper on quantum dots appeared.¹⁴ The authors could not resolve SDE, and stress in the conclusion the importance of a distinction between SPE and SDE for a full understanding of quantum dots. We have prepared quantum dots and wires. We observe, in one and the same sample, all three types of excitations and can thus directly probe the finite collective energy renormalizations both for CDE and SDE. We find that for wells, wires, and dots the strength of the observed excitations depends strongly on the incoming laser energy, i.e., excitation close to the bandgap resonance drastically increases the strength of the SPE with respect to CDE and SDE. We take this as evidence that under conditions of extreme resonance energy-density fluctuations¹⁵ are responsible for the scattering by nearly unscreened SPE. We find further that, for wires at large wave vector *q*, another single-particle effect, e.g., Landau damping has an important influence on the excitations.

Quantum dot and wire arrays have been prepared by deep-mesa etching, 7,13 i.e., etching through the active layer of a 25-nm-wide, one-sided modulation-doped single quantum well (SQW). The electron density and mobility of the unstructured sample were about 7.5×10^{11} cm⁻² and 3.5×10^5 cm² V⁻¹ s⁻¹, respectively. The period of the array was $a = 500$ nm (and $a = 800$ nm for another set of samples) for the wires and $a = 800$ nm for the dots. The geometrical width of the wires was $t=170$ nm ($t=270$ nm) and $t=240$ nm for the dots. In all samples we still had several 1D subbands or 0D levels occupied. The Raman experiments were performed at $T=12$ K using a closed cycle cryostat. Experiments at $T=2$ K show no significant change in the excitation spectrum. The energy of the exciting Ti:sapphire laser was in the range of transitions from various confined hole states to the first excited electron state of the unstructured SQW. The power densities were below 10 W cm $^{-2}$. The spectra were analyzed using a triple Raman spectrometer with a liquid nitrogen cooled charge-coupled device camera.

In Fig. 1 we show a series of Raman spectra on quantum wires with 170 nm geometrical width for different laser energies E_L . In Fig. 2 we show measurements on quantum dots

FIG. 1. Polarized spectra of a quantum wire sample with 170 nm geometrical width for a series of different laser energies. The wave vector $q=0.8\times10^5$ cm⁻¹ is transferred parallel to the wire direction. The subscripts *i* of the labels give the change Δn in the one-dimensional quantum number *n* for the transitions, which contribute predominantly to the observed excitations. Some corresponding transitions are sketched in the inset.

for different polarizations. Several lines are observed. We identify lines which are only observed in parallel polarization of incident and scattered light (polarized spectra) as CDE, those observed only in crossed polarization (depolarized spectra) as SDE. Lines observed in both polarizations are labeled, in accordance with Refs. 4 and 9, as SPE. The index Δn indicates the involved change in lateral quantum number (see below).

The first observation that we would like to point out is that the SPE's are strongly enhanced if we tune the exciting laser line E_L close to the band-gap resonance. We find that this enhancement of the SPE occurs for all systems, quantum wells, wires, and dots. We have carefully analyzed this behavior by recording extensive series of spectra for different closely spaced E_L , as shown in Fig. 1. A quite similar occurance of SPE under conditions of extreme resonance, which means that E_L is very close to the bandgap energy, was observed in *n*-doped bulk GaAs by Pinczuk *et al.*¹⁶ The authors could explain this behavior by explicit inclusion of the hole bands in the calculation of the scattering cross section. This kind of scattering mechanism, which stems from energy band dispersion effects, was first introduced by P. Wolff as scattering by energy-density fluctuations.^{15,17} Also in *p*-doped GaAs-Al_xGa_{1-x}As quantum well structures it has been found that energy-density fluctuations are the dominant scattering mechanism in the inelastic light scattering by intersubband hole excitations at the E_0 gap.¹⁸ In all our samples, we find that CDE and SDE could also be observed at laser energies far above the effective band gap due to an excitonic three-step scattering mechanism which was proposed by Danan *et al.*¹⁹ (e.g., CDE_2 in Fig. 1). In contrast, SPE's only occur if the laser energy is tuned close to the effective band gap and their intensities drastically decrease away from the band gap resonance energy as demonstrated in Fig. 1 for the wires. For two different laser energies this is shown in Fig. 2 for a dot sample with 240 nm dot diameter. This clearly demonstrates that conditions of extreme resonance are necessary for the observation of single-particle excitations. We therefore draw the conclusion that energydensity fluctuations as considered by Wolff¹⁵ and Klein¹⁷ are

FIG. 2. Polarized and depolarized spectra of the quantum dot sample for different exciting laser energies E_L . The subscripts *i* of the labels give the change Δn in the zero-dimensional quantum number *n* for the transitions, which contribute predominantly to the observed excitations. The corresponding transitions are sketched in the inset. The increasing background is due to luminescence, which is particularly pronounced for the extreme resonance condition at E_L =1558.9 meV.

responsible for the observed SPE's.

The SPE's reflect directly the relative position of energy levels in the quantum dot. The label Δn indicates, as shown in the inset of Fig. 2, the change in the energy level index of the involved transitions. The transition energies of the collective CDE's and SDE's are of course determined, in a model of Hartree and random-phase approximation (RPA), by contributions from all transitions from levels below the Fermi energy to levels above the Fermi energy. We have performed calculations (see below) and find that different excitations are predominantly determined by a certain Δn . This Δn we use to label the excitations. The excitation $CDE₁$ is not resolved in Fig. 2. This transition has odd parity and is thus, in first approximation, forbidden.

In Fig. 3(a) we summarize the q dependence of the observed excitations in the dots. It shows that the energetic positions of the observed 0D excitations do not depend on the transferred wave vector q . This is a clear signature of zero-dimensional behavior. The arrows in Fig. $3(a)$ indicate the energy renormalizations of the collective excitations with $\Delta n=2$ with respect to the SPE₂. For these excitations the ratio $(E_{SPE}-E_{SDE})/(E_{CDE}-E_{SPE})$ is about 0.12. Interestingly, this ratio seems to increase with increasing Δn . While the energetic positions of the observed 0D excitations do not depend on *q*, the intensities do. Within the measured *q* range, the intensities of the observed excitations increase with increasing *q*.

In quantum wires which were nearly in the 1D quantum limit SDE, SPE, and CDE have been investigated by Schmeller *et al.*⁹ The interesting point in our investigations is that we have systems with several occupied subbands. For such systems it has been assumed that the exchange contributions to the energy renormalizations of collective excitations could probably be cancelled.^{20,21} Our investigations show that the exchange interaction in quantum wires with several occupied subbands has a small but finite value with

FIG. 3. (a) Wave-vector dispersions of the experimental excitations in a dot sample. The arrows indicate the energy renormalizations of the $\Delta n=2$ collective excitations with respect to the corresponding $SPE₂$. (b) Wave-vector dispersions of the observed excitations in a quantum wire sample. SDE are marked with open circles, SPE with filled circles, and CDE with filled squares. The circles, SPE with filled circles, and CDE with filled squares. The FIG. 4. Depolarized (a) and polarized (b) Raman spectra of a shaded area marks the calculated single-particle continua.

respect to the direct Coulomb interaction. For the case of wires the situation is generally more involved, because here the electrons can move freely along the wire direction. In Fig. 4 we show for a series of different *q* vectors depolarized and polarized Raman spectra of electronic excitations in the same quantum wire sample as displayed in Fig. 1. These spectra were recorded at laser energies well above the band gap, so that SPE do not occur in these spectra. Here *q* is transferred parallel to the wire direction (see inset). In this scattering configuration the Raman allowed excitations with even parity are most prominent in the spectra (see also Fig. 1). In Fig. 4(a) the intraband SDE ($\Delta n=0$) can be observed. Its energy tends to zero with vanishing *q*. The polarized spectra [Fig. 4(b)] show the corresponding 1D intraband plasmon ($\Delta n=0$).The behavior of the collective $\Delta n=2$ intersubband excitations in dependence on the wave vector *q* is strikingly different for the SDE's and CDE's. The $SDE₂$ shows strong Landau damping with increasing q , if it enters the continuum of $SPE₂$ [Fig. 4(a)], whereas the $CDE₂$ is almost free of Landau damping [Fig. 4(b)]. Here *n* means the one-dimensional subband quantum number. In Fig. $3(b)$ the *q* dispersions of all observed excitations in the quantum wire sample are shown. The shaded areas mark the calculated continua of $SPE_{\Delta n}$. The continua have been calculated under the simplified assumption of a parabolic effective lateral potential, neglecting terms with q^2 , where we have taken into account that each continuum consists of a range of *m* narrow continua for the transitions with $\Delta n = m$, respectively. The quantization energy (3.5 meV) and one-dimensional electron density $(7.5 \times 10^6 \text{ cm}^{-1})$ have been determined by microwave magnetic-depopulation measurements which were performed under nearly the same illumination conditions as the Raman experiments. Figure $3(b)$ shows that the intersubband SDE $_{\Delta n}$ with $\Delta n=1,2,3$ (open circles) enter the corresponding single-particle continua at very small wave vectors *q*. This explains the strong effect of Landau damping that is observed in Fig. $4(a)$ for SDE₂. In contrast, the corresponding $CDE_{\Delta n}$ do not enter these singleparticle continua in the experimental accessible *q* range and

quantum wire sample for different wave vectors *q* at a laser energy E_L =1600.3 meV. The wave vector *q* is transferred parallel to the wires (see inset). The notation (s, p) in (a) means that the polarization direction of the incident light was perpendicular (*s*) and that of the scattered light was parallel (p) to the wire direction. Correspondingly, in (b) both polarizations were parallel to the wire direction. The line at 15 meV in the polarized spectra is due to hot luminescence.

are hence not Landau damped [see also Fig. $4(b)$]. A closer look onto the behavior of the experimentally observed SPE_{Λ_n} (filled circles) shows that the single-particle subband structure is more complicated than the parabolic assumption suggests. The experimental energies at $q=0$ do not exactly fit with the energies in the parabolic model. Furthermore the measured dispersion shows an upcurvature. By selfconsistent subband calculations, we find indeed that the effective potential is not parabolic. We note here that our calculations yield 9 occupied one-dimensional subbands for the quantum wire sample. A comparison of the respective excitation energies yields a ratio $(E_{SPE}-E_{SDE})/(E_{CDE}-E_{SPE})$ < 0.2 for the observed excitations in the quantum wire sample, so similar as in the case of dots. Thus, also 1D systems with several occupied 1D subbands show finite exchange contributions.

In conclusion we have observed all types of elementary electronic excitations—SPE, SDE, and CDE—in one and the same quantum dot (wire) sample. We find that scattering by SPE is strongly enhanced under conditions of close band-gap resonant excitation, which demonstrates that the scattering by energy-density fluctuations is responsible for the observation of quasiunscreened SPE. The huge intensity of SPE allows a detailed analysis of many-particle interactions in the zero- and one-dimensional electron systems. Our investigations yield a typical ratio $(E_{SPE}-E_{SDE})/(E_{CDE}-E_{SPE}) \approx 0.1$ for the observed excitations. This holds for quantum wire as well as for quantum dot samples with several occupied levels or subbands.

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