Acoustic plasmons and indirect intersubband excitations in tunneling-coupled $GaAs-Al_rGa_{1-r}As double quantum wells$

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We investigate tunneling-coupled GaAs-Al_xGa_{1-x}As double quantum wells (DQWs) by resonant inelastic light scattering. External gates allow us to control the carrier density and the spatial symmetry of the DQWs. By tuning the DQW potential to an asymmetric state with respect to the tunnel barrier, we are able to observe, besides the optical plasmon, an acoustic intraband plasmon of the tunneling-coupled bilayer system. For strongly asymmetric DQW potentials we find a crossover of the intersubband plasmon between the tunnelingsplit subbands from a direct to an indirect excitation of the bilayer system. Experimental excitation energies compare well with calculations in the random-phase approximation.

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In recent years there has been a growing interest in tunneling-coupled bilayer systems. Besides interaction effects, the additional degree of freedom which comes into play in these systems due to the tunneling coupling, is commonly known as the so-called pseudospin. The ground state of these systems is determined by the tunneling-split symmetric and antisymmetric single-particle states. New phenomena are expected due to intra- and interlayer Coulomb interactions. In the past decade, quite a number of experimental¹⁻⁷ and theoretical⁸⁻¹⁶ papers appeared concerning tunneling-coupled systems. Inelastic light scattering (ILS) has proven to be a very powerful method for the investigation of the electronic elementary excitations in semiconductor structures. Most optical experiments so far have been performed with symmetric tunneling-coupled bilayer systems.^{3,4} Here, theory predicts two excitations, an optical intraband plasmon and an intersubband excitation, 13,14 which, e.g., has been studied in Refs. 3 and 4. We have shown recently that the excitation spectrum is more subtle, in particular, that in addition an intraband acoustic plasmon exists that can be excited in an *asymmetric* bilayer system, only. In this communication we report on the observation of this mode.

It is well known¹⁷ that the longitudinal collective spectrum of a spatially separated, two-component twodimensional plasma consists of two modes: The acoustic plasmon (AP) where the carriers in both layers oscillate out of phase parallel to the layers, and, the optical plasmon (OP) where both layers oscillate in phase. The coupling between the layers is mediated by Coulomb interaction, only. At long wavelengths, the energy of the OP is proportional to \sqrt{q} and the energy of the AP goes linear in q , where q is the wave vector parallel to the layers. It was shown¹⁷ that at large spatial separation of the two layers, the AP can move outside of the continua of possible intraband single-particle transitions. An experimental observation of coupled-layer plasmons by inelastic light scattering was reported by Fasol *et al.*¹⁸ on GaAs-Al_{*x*}Ga_{1-*x*}As samples containing five layers in parallel. In Coulomb-coupled double quantum wells, the observation of AP's and OP's was reported by Kainth *et al.*¹⁹

In a two-dimensional (2D) electron system, microscopically, the electronic excitations can be of an intrasubband type, which means that, macroscopically, the electron system oscillates in the plane of the 2D system, or, they can be intersubband excitations, where the electrons oscillate perpendicular to the 2D plane. A peculiar feature of ILS is that, due to polarization selection rules, one can distinguish between collective spin-density excitations (SDE's) and chargedensity excitations $[$ (CDE's), i.e., plasmons]. The former appear in a depolarized scattering configuration, i.e., crossed polarizations of incoming and scattered light, and their energies are renormalized due to exchange-correlation effects. CDE's are visible in polarized scattering geometry, and, for typical electron densities of the two-dimensional electron system in the range of 10^{11} cm⁻², the direct Coulomb interaction leads to a blueshift, the so-called depolarization shift, of the CDE's with respect to the SDE's. A particular strength of the ILS method is that by angle-resolved measurements a defined and finite wave vector *q* can be transferred to the electron system.

In a previous theoretical work¹⁶ we have shown that in a tunneling-coupled bilayer system the low-energy CDE's can in a distinct way be influenced by the symmetry of the bilayer structure: In the general case of two-subband occupation, three low-energy CDE's exist, two intrasubband plasmons (AP and OP) and an intersubband plasmon (ISP), which originates from intersubband transitions between the tunneling-split ground-state subbands. In this paper we present an experimental investigation of the low-energy CDE's in modulation-doped GaAs- $Al_xGa_{1-x}As$ double quantum wells (DQW's) using resonant ILS. We were able to observe, to the best of our knowledge for the first time, the AP in a tunneling-coupled DQW. Semitransparent gates allow us to tune both the carrier densities and spatial symmetry of the DQW. By tuning the DQW potential from a symmetric to an asymmetric shape, we were able to detect the AP of the tunneling-coupled system. Very interestingly, we also found an intriguing behavior of the ISP. It exhibits a crossover from a direct to an indirect excitation of the DQW. We assume that this crossover takes place when the asymmetry of the potential is strong enough that, essentially, the wave function of

FIG. 1. Polarized (light gray) and depolarized (gray) spectra of electronic excitations in an asymmetric tunneling-coupled GaAs- $Al_xGa_{1-x}As$ DQW. The inset shows a sketch of the DQW potential and wave functions.

the lower subband is localized in one well, and, the wave function of the upper subband is localized in the other well.

The ILS experiments were performed at about $T=4.2$ K in a He exchange gas cryostat. For excitation, a Ti:sapphire laser was used, which was tuned conveniently above the E_0 gap of the DQW system for resonant excitation. The signals were analyzed in a triple Raman spectrometer equipped with a liquid-nitrogen-cooled charge-coupled device camera. The samples are modulation-doped GaAs- $Al_{0.33}Ga_{0.67}As$ DQW's. They consist of two 15-nm-wide GaAs quantum wells, separated by a 1-nm $Al_xGa_{1-x}As$ tunneling barrier. The AlGaAs barriers on both sides of the DQW are modulation doped using Si delta layers. In the top barrier layer, two deltadoping layers were grown, separated by a 28-nm AlGaAs and a 20-nm spacer layer to the upper GaAs well. In the lower barrier, one delta layer, separated by a 41-nm spacer from the lower GaAs well, was grown. Semitransparent titanium gates were deposited on top of the samples. By applying a voltage between the DQW and the gate, the carrier density and the self-consistent potential of the DQW in the growth direction could be tuned.

Figure 1 displays Raman spectra of the low-energy excitations of a DQW for large wave-vector transfer $q=1.35$ $\times 10^5$ cm⁻¹. Since the three peaks, indicated in Fig. 1, appear dominantly only in the polarized spectrum, we can identify them as CDE's and rule out single-particle excitations. Remarkably, the excitation at 19 meV is much broader than the excitations at 2 meV and 9 meV, which have very similar linewidths. From considerations which will be discussed below, we find that the potential of the DQW structure in this experiment was strongly asymmetric with respect to the tunneling barrier (see inset of Fig. 1). The interpretation of the CDE's displayed in Fig. 1 follows from the measured wavevector dispersion, which is plotted in Fig. 2 (symbols). The lowest-energy excitation (full squares in Fig. 2) shows a linear *q* dependence and is therefore interpreted as the AP. The OP exhibits a square-root-like behavior (full circles in Fig.

FIG. 2. Experimentally determined mode positions of the lowenergy CDE's depending on the wave-vector transfer *q* parallel to the DQW for the same sample as shown in Fig. 1. The hatched regions mark the continua of intra- and intersubband single-particle transitions.

2), and the highest-energy excitation, the ISP between the tunneling-split subbands, depends only weakly on q (open symbols in Fig. 2). The hatched regions mark the continua of possible single-particle transitions. Note that two subbands are occupied in this sample. It is somehow unexpected and very interesting that the AP does not seem to show any significant effect of Landau damping, even though its energy is inside the intraband single-particle continuum (see Fig. 2). We think that this might have something to do with the mechanism of Landau damping: The AP is a collective excitation of the total tunneling-coupled system. The concerning region of intraband single-particle transitions, where its energy falls into $(Fig. 2)$, is, however, the continuum of the lower subband, only. The effect of Landau damping on the ISP will be discussed further below. The thick solid lines in Fig. 2 are calculated within the random-phase approximation $%$ (for details of the calculation see Ref. 16). For simplicity, we have modeled in the calculations the DQW structure by two electronic delta layers, separated by an effective tunneling barrier. The parameter which determines the tunneling coupling is the splitting of the two lowest subbands, Δ_{SAS} , for the case of a symmetric DQW potential. This is a fit parameter in our calculations. The second parameter, $sin(\delta)$ $=\Delta_{SAS}/\Delta$, where Δ is the splitting of the two lowest subbands in the general (asymmetric) case, expresses the symmetry of the DQW potential: For $sin(\delta)=1$, the DQW potential is symmetric, and, for $0 \le \sin(\delta) \le 1$, the potential is asymmetric with respect to the tunneling barrier. The third parameter in the calculation shown in Fig. 2 is the total carrier density n_{tot} of the DQW. For the sample displayed in Fig. 2, we get $n_{tot} = 8.6 \times 10^{11}$ cm⁻², $\Delta_{SAS} = 3.65$ meV, and $\sin(\delta) = 0.205$, which means that the DQW potential is significantly asymmetric (see also the sketch in the inset of Fig. 1). We emphasize that the observation of AP's in multilayer structures, reported so far $(e.g., Refs. 18 and 19)$, were exclusively obtained on Coulomb-coupled structures, without tunneling coupling. Our theoretical considerations in Ref. 16

FIG. 3. Polarized Raman spectra for different gate voltages V (in steps of 0.25 V), applied between the DQW and a front gate. The wave-vector transfer is fixed at $q=1.35\times10^5$ cm⁻¹. On the righthand side, the shape of the DQW potential is sketched.

showed that, in a tunneling-coupled bilayer, the AP should have a finite energy, and hence should be observable in experiment, for an *asymmetric* DQW potential, only. For the symmetric case, its energy tends to zero (is exactly zero for delta layers).

To confirm our interpretation, we have performed experiments, where we have tuned the symmetry of the DQW potential by applying different gate voltages between the DQW and the front gate. Figure 3 shows a series of polarized Raman spectra for different gate voltages *V* between -1.25 V and $+3.25$ V. At $V=-1.25$ V, the energy of the ISP is minimal (\approx 7.5 meV) and there is no AP (lowest-energy excitations in Fig. 3) visible at that gate voltage. From both we conclude that the potential is symmetric in this gate-voltage range. By tuning *V* towards positive values, the carrier density in the DQW increases, and, at the same time, the potential becomes more and more asymmetric (see schematic pictures on the right-hand side of Fig. 3). The energy of the OP is determined dominantly by the total carrier density of the DQW. Since in the experiment displayed in Fig. 3 the wave vector q is fixed, the energy of the OP is essentially proportional to $\sqrt{n_{tot}}$. In that sense, the OP serves as a direct monitor of the electron density in the experiment. With increasing carrier density and potential asymmetry (between *V* $=$ -1.25 V and *V*=0.75 V in Fig. 3), the energy and the linewidth of the ISP increases rapidly. At about $V=0.5$ V, its intensity drops down within a relatively small voltage range. We interpret this as a crossover of the ISP from a direct excitation of the DQW structure to an indirect excitation: For

a strongly asymmetric potential, the wave function of the lowest subband is located dominantly in one well (e.g., the left one, as sketched in the inset of Fig. 1), and the wave function of the second subband is located in the other well. In this special situation, for an intersubband excitation, the electrons have to tunnel between the two layers. The strongly reduced overlap of the wave functions leads to the reduced intensity of the ISP. We note that even for the symmetric case $(V=-1.25$ V in Fig. 3) the linewidth of the ISP is, by a factor of about 2.5, larger than the linewidths of the intraband excitations. This is an effect of Landau damping of the ISP at finite *q*: The experiment in Fig. 3 is performed at a relatively large wave-vector transfer $q \approx 1.35 \times 10^5$ cm⁻¹, where the ISP enters the continuum of intersubband singleparticle transitions and is hence Landau damped $(cf. Fig. 2)$. By tuning the gate voltage towards positive values, both the electron density and the splitting of the lowest subbands Δ increase. Both lead to a broadening of the intersubband single-particle continuum. This could account for the increasing linewidth of the ISP in the range -1.25 V \lt *V* \leq + 1.00 V. That this is a reasonable assumption can also be seen by comparing the maximum linewidth of the ISP in Fig. 3 to the width of the intersubband single-particle continuum at $q=1.35\times10^5$ cm⁻¹ in Fig. 2, which is about 6 meV.

For voltages $V \ge 1$ V in Fig. 3, the carrier density and the asymmetry of the potential seem to decrease again, which can be deduced from the decreasing energies of the OP and the ISP in that voltage range. For voltages $V > 2.5$ V, the system should be again in the symmetric state. This behavior is somehow intriguing, since, naively, one would expect that for increasing positive voltages the density should increase continuously. We believe that the observed decrease of the density in the DQW is due to the opening of a bypass in the upper barrier layer: For $V \approx 1$ V, the conduction band of the delta-doped layer in the top AlGaAs barrier touches the

FIG. 4. Measured mode positions of CDE's in a DQW depending on the gate voltage. (a) – (c) display polarized spectra for the marked gate voltages. The open squares mark the positions of very broad excitations, which occur at very large voltages, only, and which are presumably due to electronic excitations within the top barrier layer.

Fermi energy and electrons flow into this bypass. This leads to a reduction of the electron density in the DQW, behind the bypass, until it reaches the flat band case, i.e., a symmetric DQW potential. Note that, due to the alloyed contacts, the bypass and the DQW are directly connected. To summarize this behavior, we have plotted in Fig. 4 the observed mode positions versus gate voltage. For very high voltages, *V* >6 V (not shown in Fig. 3), the density seems to increase again, as can be seen from the increasing energy of the OP (full circles in Fig. 4). We assume that in this voltage range, with a bypass in the upper barrier layer, the DQW potential is always fairly symmetric, since the entire voltage drops between the front gate and the bypass, which is above the DQW. Comparing Figs. $4(a)$ and $4(c)$ supports this interpretation: For both cases the total carrier density in the DQW is the same, since the energy of the OP is the same. For Fig. $4(a)$, the DQW potential is strongly asymmetric (see discussion above). Therefore, the AP has a finite energy and is visible at about 2 meV. From Fig. $4(c)$ we find that the AP should have an energy which is at least smaller than 1 meV,

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since it is not observable in the measured low-energy range. This is in agreement with the theoretical considerations of Ref. 16. Moreover, this is also, besides the polarization selection rules shown in Fig. 1, proof that the observed lowenergy excitation is an AP. For the case that it would be a single-particle excitation, its energy should only depend on the carrier density and not the symmetry of the DQW. Therefore, it should then also be visible in Fig. $4(c)$, which is not the case.

In conclusion, we have investigated the low-energy CDE's in tunneling-coupled DQW's depending on the carrier density and symmetry of the DQW. For asymmetric potentials, we have observed the AP of the tunneling-coupled system at low energies and found a crossover of the ISP from a direct to an indirect excitation of the coupled bilayer system.

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