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Strong intermediate-state effects in exciton-mediated electronic intrasubband scattering in multiple-quantum-well structures

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Electronic instrasubband excitations are studied by means of resonant inelastic light scattering by a two-dimensional electron gas. The different effect of sharp excitonic intermediate states on depolarized and polarized scattering gives direct evidence for the collective behavior of the spin-density excitation in contrast to the band of single-particle excitations. The observed spin-density energy (0.9 meV) is at significantly lower energy than the single-particle energy (1.1 meV). This is due to the Coulomb exchange interaction in the electron gas.

Modulation-doped heterostructures have become a model system with which to study many-body effects in the free-electron gas. Since the work of Pinczuk *et al.*[1] and Abstreiter and Ploog,[2] inelastic light scattering has been used as a powerful technique to study the energies and the character of the elementary excitations of the two-dimensional electron gas.[3] The method has recently attracted new interest after the discovery of huge enhancements of the *inter*subband scattering cross section at photon energies resonant with quantum-well excitons[4] and the observation of separated single-particle excitations (SPE) and spin-density excitations (SDE) in *inter*subband scattering.[5,6]

In *inter*subband scattering, the collective character of the SDE shows up in the much smaller width of the SDE compared with the band of SPE.[5] On the other hand, due to inherent Landau damping, the *intra*subband peak of the SDE has almost the same width as the SPE. No definite selection rules exist for the SPE (Ref. 7). Hence the question arises whether other indications of the collective character of the SDE can be observed, permitting clear distinction between SDE and SPE. There is great interest in resolving SDE and SPE also in *intra*subband scattering, because the relative position of the two excitation peaks would allow direct measurement of the magnitude of the exchange Coulomb interaction in the ground state of the electron gas.[8-10]

In this Rapid Communication, we show that a suitable probe of the collective character of the SDE are intermediate-state effects in light-scattering spectra where the laser photon is resonant with quantum-well excitons. For low-energy *intra*subband excitations (≈ 1 meV), this is a nearly doubly resonant process, which gives rise not only to huge resonant enhancements, but also to strongly different spectra for polarized and depolarized conditions. The specific differences can be traced back to the collective and single-particle character of the excitations involved. In previous work[11,12] on *intra*subband scattering, the resonances with excitons have not been investigated.

The sample examined here is made up by ten asymmetrically modulation-doped GaAs quantum wells (of 250-Å width), grown by molecular-beam epitaxy. The 2500-Å-wide $Al_{0.3}Ga_{0.7}As$ barriers are δ doped at a distance of 400 Å below the well. The electron density under illumination is $n = 2\times10^{11}$ cm^{-2} (Ref. 13) and the mobility is 1×10^5 cm^2/Vs (for 77 K and for $n = 1.4\times10^{11}$ cm^{-2} in the dark). The incident power density is below 5 W/cm^2 and the temperature of the sample holder is 4.2 K. The spectra are recorded in the near-backscattering configuration under oblique incidence of the laser on the sample. Depolarized and polarized inelastic light-scattering spectra are measured in the $z(x'y')\bar{z}$ and $z(x'x')\bar{z}$ configurations, respectively, z corresponds approximately to the $\langle 001\rangle$-growth axis and x' and y' represent the $\langle 110\rangle$ and $\langle 1\bar{1}0\rangle$ directions, respectively. The selection rules[7,14] predict depolarized scattering by SDE and polarized scattering by charge-density excitations (CDE). The SPE are allowed for both polarizations.[7,14]

Figure 1 shows the photoluminescence (PL) and the photoluminescence-excitation (PLE) spectra. The main

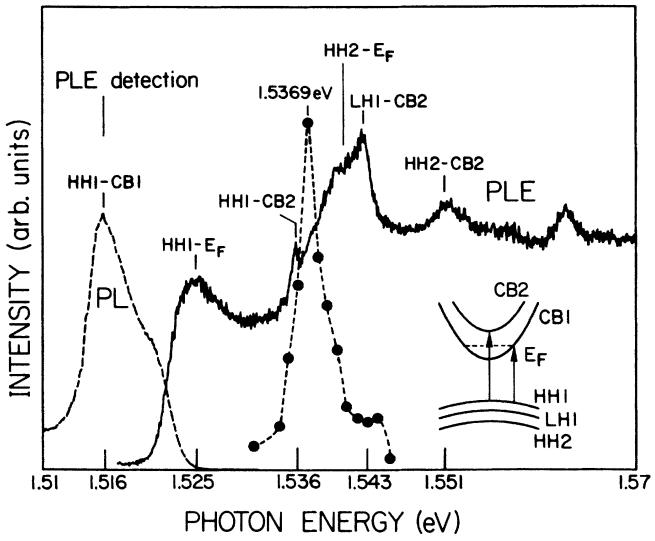

FIG. 1. The photoluminescence (dashed line) and the photoluminescence excitation spectra (solid line) are shown. The solid dots give the measured Raman intensity of the SDE as a function of the laser photon energy.

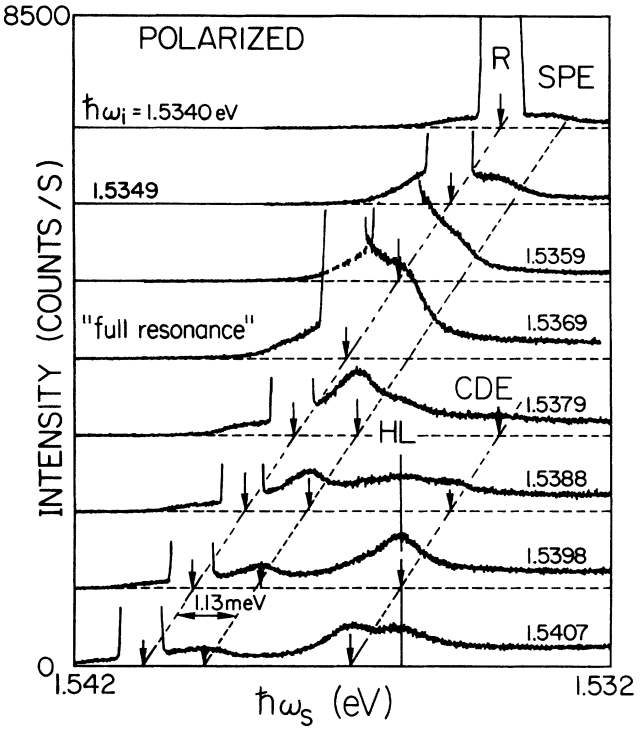

FIG. 2. Polarized Raman spectra for different laser photon energies $\hbar\omega_i$, a carrier concentration n of 2×10^{11} cm^{-2} and a scattering wave vector of 1.02×10^5 cm^{-1}. The peak at $\hbar\omega_s = 1.536$ eV represents hot luminescence. The charge-density excitation and the single-particle excitation are seen at Stokes shifts of 3.85 and 1.13 meV, respectively. The different spectra are given for laser photon energy steps of approximately 1 meV and are offset by 1000 counts/s. Hence, the position of the Rayleigh peak (R) and the Raman peaks lie near a straight line, which is shown as a guide to the eye.

intermediate state in our Raman experiments is the exciton derived from the heavy-hole band (HH1) and the empty second conduction band (CB2), the corresponding band-to-band transition being shown in the inset in Fig. 1. This exciton is allowed, because parity is broken by the electrical fields in these asymmetrically modulation-doped wells. It shows up as a sharp peak in the PLE spectrum at $\hbar\omega_{\text{HH1-CB2}} = 1.536$ eV. Hence, the position of the upper subband CB2 (20 meV above CB1) is well above the Fermi energy (7 meV) and only one subband is occupied. The transition from the HH1 band to the Fermi edge gives rise to the Fermi-edge singularity[15] in the PLE spectrum at $\hbar\omega_{\text{HH1-}E_F} = 1.525$ eV and is shown in the inset. The broad peak in Fig. 1 at about $\hbar\omega_{\text{HH2-}E_F} = 1.54$ eV is attributed to a Fermi-edge singularity with the second heavy-hole band (HH2). The solid dots show the measured Raman intensity of the SDE as a function of laser photon energy. The maximum of the scattering cross section is at 1.5369 eV. For this energy, the scattered (outgoing) photon energy is ≈ 1.536 eV and is in resonance with $\hbar\omega_{\text{HH1-CB2}}$. The enhancement of the Raman intensity is in this case very strong, because, aside from the resonance of the outgoing photon, the ingoing photon is resonant with excited exciton states.

Figures 2 and 3 show the resonant behavior in Raman scattering of the *intra*subband excitations as a function of laser photon energy $\hbar\omega_i$, for polarized and depolarized conditions, respectively. The peak at $\hbar\omega_s = 1.536$ eV is due to hot luminescence (HL) from the HH1-CB2 exciton. The CDE is seen in the polarized spectra at a Stokes shift of 3.9 meV. Low-energy peaks are observed for the polarized and depolarized case at 1.1 and 0.9 meV, respectively. In full resonance at $\hbar\omega_i = 1.5369$ eV, the polarized excitation transforms into a broad structure, whereas a peak is still clearly visible in the depolarized spectrum. For $\hbar\omega_i = 1.5379$ eV, the greatly enhanced Raman-scattering process is dominant compared to the HL, the latter being only a small shoulder of the Raman peak. The relaxation to $\hbar\omega_{\text{HH1-CB2}}$, giving rise to HL, could be due to any excitation (e.g., acoustical phonons), but the fact that the Raman scattering by electronic excitations at $\hbar\omega_i = 1.5379$ eV is stronger than HL indicates that the peak in full resonance is also due to electronic excitations. The polarized excitation is attributed to the scattering by SPE: Its line shape is composed of independent electron-hole excitations of the same wave vector but of different energies. Those electron-hole transitions corresponding to a scattering of the initial excitonic state to $\hbar\omega_{\text{HH1-CB2}} = 1.536$ eV are resonantly enhanced: this explains the observed change in line shape, when passing through resonance. The observation of the density of states of SPE, modified by intermediate-state effects, is predicted[14] for parallel polarizations and for full band-to-band resonance (no excitonic effects). The depolarized peak is attributed to SDE. The SDE is a collective excitation which has a definite energy for a given wave vector, from which very little variation in peak position for the depolarized case is expected as observed. A rather small cross section, if at all, is observed for depolarized SPE. This is not unreasonable. Polarized scattering by SPE arises in resonance due to a cancellation of the large dielectric screening.[14] For depolarized scattering, screen-

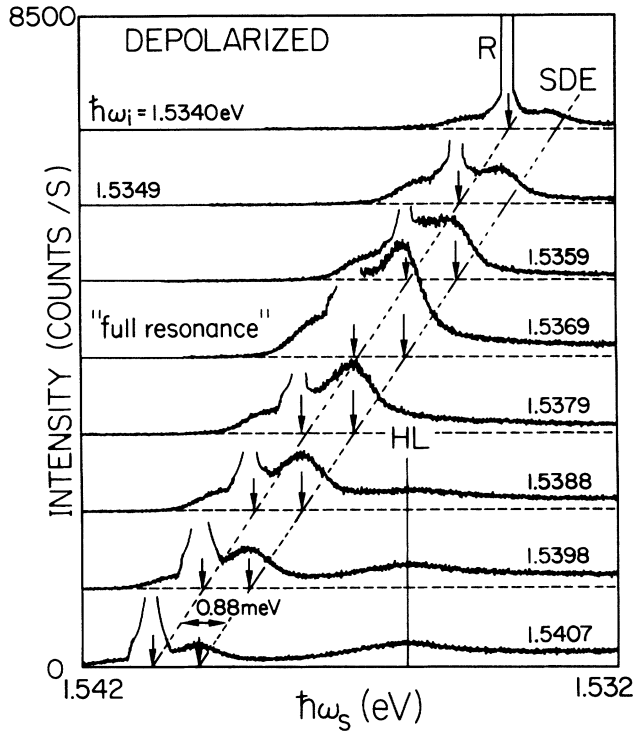

FIG. 3. Depolarized Raman spectra for different laser photon energies $\hbar\omega_i$, the parameters are the same as in Fig. 2. The spin density excitation is seen at a Stokes shift of 0.88 meV.

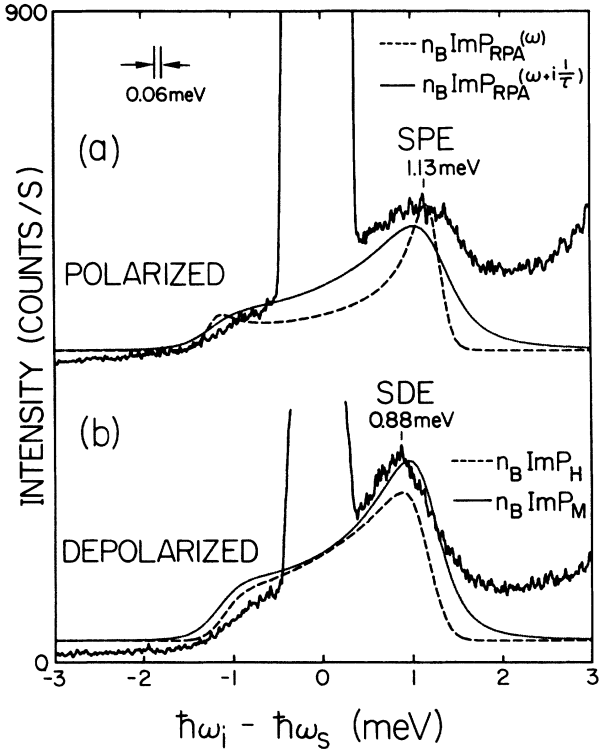

FIG. 4. (a) Measured polarized and (b) depolarized Raman spectra for a laser photon energy of $\hbar\omega_i = 1.5407$ eV. The solid and dotted theoretical lines are fits as described in the text.

ing by spin-density fluctuations is in any case small for energies in the SPE range, giving rise to the SDE. Our experimental findings correspond to very recent results on *inter*subband scattering, where it has been found that resonant scattering by SPE is mainly polarized.[6]

Over the whole range of laser photon energies shown in Figs. 2 and 3, an energy difference is seen between SPE and SDE. In Fig. 4, this is compared with theory, using the spectra recorded with $\hbar\omega_i = 1.5407$ eV. An electron-gas temperature of 9 K is used. The experimental SPE is approximated in Fig. 4(a) by the density of states (DOS) of SPE, the DOS being proportional to the imaginary part of the random phase approximation (RPA) expression for the polarization $P_{\mathrm{RPA}}(\omega,k)$.[10] We take account of a finite SPE relaxation time τ by adding an energy-independent imaginary part to each of the SPE energies. Hence, the Raman intensity I_R is taken proportional to $n_B(\omega, T)\mathrm{Im}[P_{\mathrm{RPA}}(\omega+i/\tau,k)]$, where $n_B(\omega,T)$ is the thermal population factor. The solid and dashed theoretical lines in Fig. 4(a) show this fit for $\hbar/\tau = 0.3$ meV and infinite τ, respectively. The corresponding theoretical peak positions are 1.03 and 1.15 meV. In Fig. 4(b), we fit the SDE peak. In order to explain the experimental energy difference between SDE and SPE, a generalization of the RPA, introduced by Hubbard,[8-10,16] is used:

$$I_R \propto n_B \mathrm{Im}[P_{\mathrm{RPA}}/(1+\gamma_H P_{\mathrm{RPA}})] = n_B \mathrm{Im}P_H .$$

γ_H represents exchange screening of the *intra*subband Coulomb vertex and is used as a fit parameter. The dashed line in Fig. 4(b) shows the result for $n\gamma_H = 2.5$ meV, where n is the carrier density. The theoretical peak is at 0.88 meV. This value $n\gamma_H$ is an upper estimate, because a finite relaxation time also introduces a splitting between SPE and SDE. This can be discussed using an approximation due to Mermin[12,17] for the polarization:

$$I_R \propto n_B \mathrm{Im}\left[\frac{(1+i/\tau\omega)P_{\mathrm{RPA}}(\omega+i/\tau,k)}{1+(i/\tau\omega)P_{\mathrm{RPA}}(\omega+i/\tau,k)/P_{\mathrm{RPA}}(0,k)}\right]$$

$$= n_B \mathrm{Im}P_M .$$

The solid theoretical line in Fig. 4(b) shows the fit obtained for $\hbar/\tau = 0.3$ meV. The peak is 0.97 meV. Using a still larger value of $\hbar/\tau = 0.5$ meV, the peak energies (not shown) for the SPE and the SDE become 0.96 and 0.84 meV, respectively, the splitting being even in that case much smaller than the experimental splitting of 0.25 meV. This shows that the above-discussed Hubbard-type correction is necessary. Our result $n\gamma_H \approx 2.5$ meV is 1.7 times larger than the value $n\gamma_H = 1.5$ meV, obtained recently[5] for the *inter*subband vertex (at $k = 0$) in comparable structures. This increase is in agreement with the local-spin-density expression for the exchange parameter:[6,8] it is explained by the almost doubled overlap integral between the two electronic states involved.

Using the parameters of Fig. 4, the RPA results in an energy of 4.14 meV for the CDE. The effect of the Hubbard-type correction of RPA on the CDE energy is found to be negligible.

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