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Coupling of excitons with free electrons in light scattering from GaAs quantum wells

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We observe unexpectedly narrow peaks (~1–2 meV) in the resonant enhancement of inelastic light scattering by a two-dimensional electron gas. The experiments are carried out in high-mobility modulation-doped GaAs quantum wells. Scattering by spin-density and charge-density intersubband excitations is enhanced through incoming and outgoing channels. Such excitonic resonances are much sharper than the Fermi energy (10–20 meV). To explain these observations we propose new mechanisms for light scattering by excitations of the electron gas. They are based on the coupling between excitons and the Fermi sea through Coulomb exchange and direct interactions.

The presence of free electrons has been shown to alter significantly the optical properties of semiconductor quantum-well structures. Experimental and theoretical studies of absorption and emission spectra, especially in GaAs-GaAlAs two-dimensional (2D) systems, have recently attracted considerable interest.[1-10] The renormalization of the band gap is explained by many-body exchange of correlation effects.[2-4,9] Electron-hole interactions must also be taken into account to explain the line shape of emission and absorption spectra of modulation-doped quantum wells.[1,5-10] The enhancement of oscillator strength near the absorption edge has been interpreted in terms of a hole correlated with the whole Fermi sea,[6,11] giving rise to a singularity[11] as observed earlier in x-ray spectroscopy of metals.[12]

The results presented in this paper show that resonant inelastic light scattering from modulation-doped GaAs quantum wells probes the interactions between excitons and the Fermi sea. These interactions manifest themselves in the strong enhancement of the scattering intensities of charge-density and spin-density intersubband excitations of the 2D electron gas at photon energies resonant with quantum-well excitons. Although light scattering by excitations of the Fermi sea have been studied extensively,[13] our results are the first in which excitonic enhancements are identified. These results are evidence of a new mechanism of inelastic light scattering by the electron gas. The direct term of the Coulomb interaction enters in the light scattering cross section of charge-density excitations and the exchange term in that of spin-density excitations. This new light scattering mechanism provides a unique insight into the electron-electron interactions in doped semiconductor quantum wells.

The light scattering experiments were performed on several one-sided modulation-doped quantum-well structures, grown by molecular-beam epitaxy, with well widths in the range 200 Å $\leq L_W \leq$ 250 Å. The Si dopants are introduced on the top $Ga_{0.7}Al_{0.3}As$ layer and separated from the GaAs quantum well by a wide undoped spacer layer 300 Å $\leq d \leq$ 400 Å. The areal density n_s of the electron gas at the GaAs-$Ga_{0.7}Al_{0.3}As$ interface is in the range $(3\text{–}6.5)\times 10^{11}$ cm^{-2} and the low-temperature mobility is within $(3.5\text{–}9)\times 10^5$ cm^2/V sec. These mobilities are higher than previously reported in GaAs quantum wells.[14] The band bendings in the asymmetric wells are similar to those of single heterojunctions in which the highest electron mobilities have been reported.[14] Because of the confinement of the valence states, the exciton transitions could be measured by conventional photoluminescence (PL) and photoluminescence excitation (PLE) techniques. Inelastic light scattering spectra were recorded in the conventional polarized $z(x'x')\bar{z}$ and depolarized $z(x'y')\bar{z}$ configurations, where z is the axis normal to the layers and light polarizations x' and y' correspond respectively to $\langle 110\rangle$ and $\langle 1\bar{1}0\rangle$ directions. Light scattering by charge-density and spin-density fluctuations was observed in polarized and depolarized configurations, respectively.[13] The measurements were made at $T = 3$ K, using tunable cw dye lasers operating in the range 720–820 nm. Typical power densities were less than 5 W/cm^2.

Figure 1 shows spectra obtained in the range of the lowest intersubband excitations of a 200-Å asymmetric

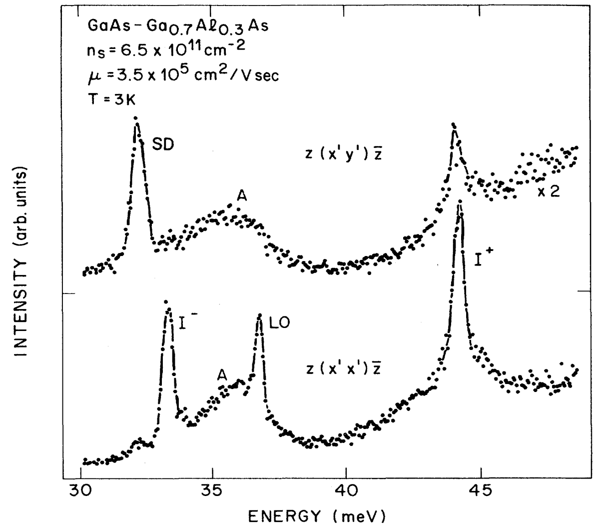

FIG. 1. Depolarized $z(x'y')\bar{z}$ and polarized $z(x'x')\bar{z}$ inelastic light scattering spectra of intersubband excitations.

quantum well with $n_s = 6.5 \times 10^{11}$ cm^{-2} and $\mu = 3.5 \times 10^5$ cm^2/V sec. The depolarized spectrum exhibits a sharp line at 32.1 meV. There is also a broader band centered at 35.3 meV labeled A in Fig. 1. The sharp line is absent in polarized spectra. Such a selection rule identifies this transition as the spin-density (SD) intersubband excitation with an energy close to the subband spacing E_{01}.[13] The width of the line is only 0.6 meV. In higher mobility samples, the measured linewidths are as low as 0.3 meV. These are the narrowest intersubband excitations reported.[13] The narrower linewidths, as compared to earlier results, are probably a consequence of reduced inhomogeneous broadening and reduced carrier scattering in the asymmetric wells. In the polarized spectrum we observe the three peaks labeled I^-, LO (longitudinal optical phonon), and I^+. The I^- and I^+ lines correspond to the two branches of the coupled mode of charge-density intersubband excitation and LO phonons. The energy of the I^- collective mode is very close to that of the GaAs TO phonon (33.7 meV). The energy shift of the I^+ mode from SD is due to the large depolarization field effect in this asymmetric well. The broad band A is more intense in parallel polarization. Although close in energy, band A does not follow the polarization selection rules that apply to light scattering by spin- or charge-density fluctuations of free electrons.[13] We have determined that line A is not associated with impurity-related intersubband excitations, such as those reported by Gammon *et al.*[15] Work in progress indicates that it originates from scattering by single-particle intersubband excitations.[16]

Figure 2(a) shows the profile of resonant enhancement of the intensity of the SD excitation in the photon energy range between 1.54 and 1.61 eV. For lower energies, the strong luminescence of the Fermi sea obscures the light scattering signal. Surprisingly, sharp peaks are observed when the *incident* (incoming resonance) or the *scattered* (outgoing resonance) photon energies are equal to the energies of excitons measured in PLE spectra. The notation E_nH_m (E_nL_m) refers to the exciton formed with an electron in the nth excited conduction subband and a heavy (light) hole in the mth excited valence subband. Figure 2(b) shows a schematic diagram of the quantum well, displaying the low-lying conduction and valence energy levels. The strong peak at 1.572 eV corresponds to a resonant enhancement when the laser photon energy is equal to that of the E_1H_2 exciton. The associated outgoing resonance is observed at 1.604 eV. The E_1L_1 incoming is also visible but much weaker than the E_1H_2. The other sharp structures in Fig. 2(a) are the outgoing resonances at E_1H_0, E_1L_0, and E_1H_1.

Resonant profiles of lines I^- and I^+ in polarized spectra (not shown) also exhibit sharp resonance peaks at the energies of excitons. In the sample studied above, the incoming resonances of the I^+ modes are obscured by the strong luminescence at the fundamental energy gap. The I^- mode shows also a strong incoming resonance at the E_1H_2 exciton. The strength of the outgoing resonance is about 1 order of magnitude smaller. In most of the excitonic resonances we find that the intensities of light scattering by charge- and spin-density fluctuations are comparable.

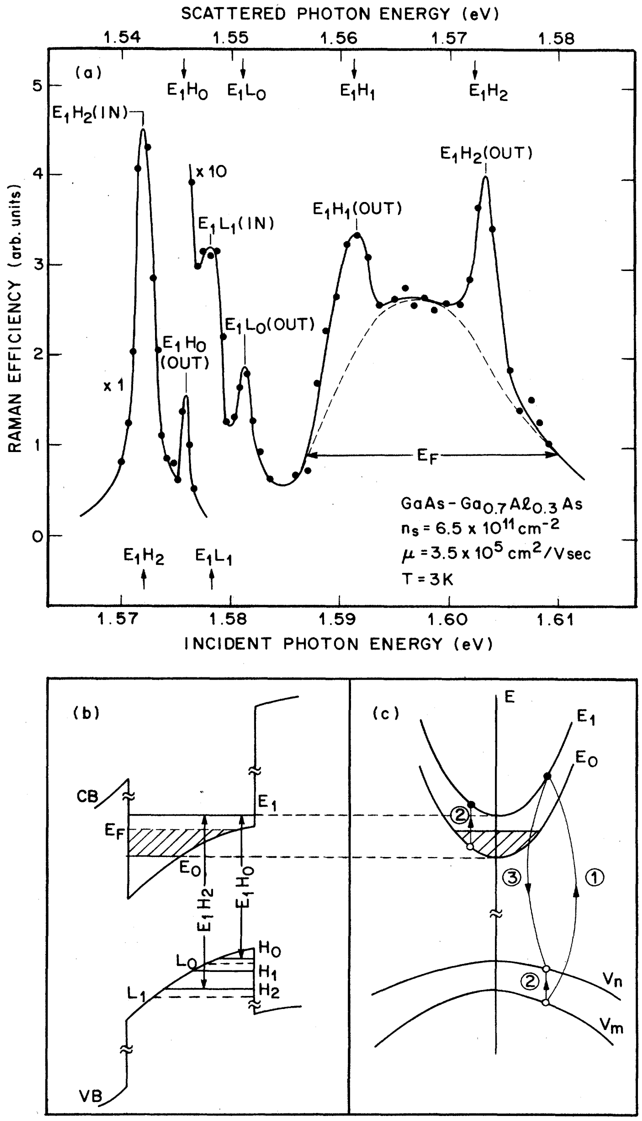

FIG. 2. (a) Resonant enhancement profile of light scattering by the spin-density intersubband excitation. The energies of excitons obtained in PLE are shown by the arrows. (b) Schematic structure of the quantum well, showing the asymmetric band bending and lowest conduction and valence confined states. (c) Diagram of resonant light scattering by intersubband excitations in the 2D wave-vector space. Virtual creation and annihilation of excitons are represented as single electron and hole events.

Resonant light scattering by electronic excitations of the 2D electron gas has been previously described by processes in which photons couple directly to states of the Fermi sea.[13,17] In the two virtual optical transitions an electron is promoted from the valence band to an excited conduction subband and an electron below the Fermi level recombines with the remaining hole.[17] This description is supported by experimental results of resonant light scattering in modulation-doped GaAs quantum wells obtained when the incident photon energy is close to the $E_0 + \Delta_0$ energy gap of GaAs ($\sim$1.9 eV).[18,19] A distinctive feature of this type of inelastic light scattering is the width of the profile of resonant enhancement. Since any electron in the Fermi sea can participate in the virtual transitions, the width is comparable to the Fermi energy. The broad band at 1.595 eV in Fig. 2(a) is assigned to this mechanism. The other peaks in the resonant profile of charge- and spin-density excitations are much sharper than the Fermi energy.

To explain the unexpected sharp incoming and outgoing resonances we propose new third-order inelastic light scattering processes in which two optical transitions are associated with intermediate exciton states. Light scattering by spin-density intersubband excitations takes place because of exchange coupling of exciton with the Fermi sea. This is the interaction that accounts for polarization anomalies observed in optical processes of modulation-doped quantum wells.[6] The coupling generates an intersubband excitation and the exciton associated with the incoming photon is scattered to a different state before it annihilates and the scattered photon is emitted. To discuss the new mechanism we consider the exciton as a superposition of electron-hole pair states. Exciton scattering is thus described by single conduction or valence state events. Figure 2(c) shows the case in which it is the hole in the exciton state that makes a transition from a valence subband V_m to another subband V_n. We emphasize that this mechanism conserves wave vector, as suggested by the sharpness of the SD peak (<0.6 meV).

In general, we can write the intensity of the resonantly scattered light as[20]

$$I \propto \left| \sum_{vv'} \left(\frac{M^*_{v'}(H_e)_{vv'}M_v}{(E_v-\hbar\omega_L+i\eta)(E_{v'}-\hbar\omega_S+i\eta)} + \frac{M^*_v(H_e)_{v'v}M_{v'}}{(E_{v'}-\hbar\omega_L+i\eta)(E_v-\hbar\omega_S+i\eta)} \right) \right|^2 , \tag{1}$$

where M_v is the optical matrix element for the creation of exciton in state $|v>$, of energy E_v. $(H_e)_{vv'}$ is the matrix element for the interaction between the Fermi sea and the exciton in state $|v>$, making a transition to state $|v'>$. A phenomenological broadening η has been introduced. The simplified model proposed by Ruckenstein, Schmitt-Rink, and Miller[6] considers an infinitely heavy hole with spin that is coupled to a Fermi sea of N electrons through direct (U) and exchange (J) contact interactions written as

$$H_e = -\frac{U}{N}\sum_{\substack{nn'\\kk'\\s}} a^\dagger_{n\mathbf{k}s}a_{n'\mathbf{k}s} - \frac{J}{4N}\sum_{\substack{nn'\\k'\\ss'}} a^\dagger_{n\mathbf{k}s}\langle s|\sigma_z|s'\rangle a_{n'\mathbf{k}s'} . \tag{2}$$

$a_{n\mathbf{k}s}$ ($a^\dagger_{n\mathbf{k}s}$) are annihilation (creation) operators for an electron of momentum $\mathbf{k}$ and spin s subband n and σ_z is the Pauli matrix. Here, we shall be concerned only with intersubband excitations $n=0$, $n'=1$. The exchange contribution in Eq. (2) involves the terms ($a^\dagger_{l\mathbf{k}\uparrow}a_{0\mathbf{k}'\uparrow} - a^\dagger_{l\mathbf{k}\downarrow}a_{0\mathbf{k}'\downarrow}$) and gives rise to fluctuations in SD via $0\rightarrow 1$ intersubband excitations. The contribution of the direct term contains ($a^\dagger_{l\mathbf{k}\uparrow}a_{0\mathbf{k}\uparrow} + a^\dagger_{l\mathbf{k}\downarrow}a_{0\mathbf{k}'\downarrow}$) and therefore represents fluctuations in charge density of the coupled I^+ and I^- modes.

Polarization selection rules for SD excitations can be derived from Eq. (1). We assume the general form

$$H_{\text{exc}} = -J\sum_{i<N}\mathbf{S}\cdot\sigma_i = -J\sum_{i<N}S_z\sigma_{iz}$$

for the exchange interaction, where $\mathbf{S}$ is the spin operator for the hole and i labels the states of the Fermi sea. We find that the light scattering tensor has only off-diagonal terms. It predicts spectra in the depolarized configuration, in agreement with experiment.

In the vicinity of E_1H_2, hereafter denoted $|v_0\rangle$, the dominant contribution to the scattered intensity is due to the first term if $\hbar\omega_L \sim E_{v_0}$ and due to the second term if $\hbar\omega_S \sim E_{v_0}$. In principle the summation is extended over all possible intermediate exciton states. In order to extract contributions from the dominant terms in Eq. (1) we simplify the expression to a single term. In this case, the intensity ratio between the incoming and outgoing resonances of the SD excitation is

$$\frac{I_{\text{inc}}}{I_{\text{out}}} \propto \left| \frac{E_v - E_{v_0} - E_{01}}{E_v - E_{v_0} + E_{01}} \right|^2 , \tag{3}$$

where I_{inc} and I_{out} are obtained by setting $\hbar\omega_L$ or $\hbar\omega_S$ equal to E_{v_0} in Eq. (1), respectively. The experimental value is $I_{\text{inc}}/I_{\text{out}} = 20 \pm 5$ which yields $E_v = 1.552$ eV $\pm$ 1.5 meV. This value for the energy of the intermediate state agrees well with the energy of the E_1L_0 or E_1H_0 exciton in Fig. 2(a). Therefore, we conclude that diagrams similar to those shown in Fig. 2(c) dominate the evaluation of the scattering cross section under the conditions of this experiment. Excitonic transitions of the type $E_1H_2 \rightarrow E_1L_0$ caused by the Coulomb interaction with the Fermi sea are made possible by valence-band mixing.[7] The analysis of incoming and outgoing resonances of light scattering by charge-density intersubband excitations yields similar results.

In conclusion, we have identified a new type of resonant inelastic light scattering mechanism by intersubband excitations. In these processes intermediate excitonic states are dominant. Resonant inelastic light scattering is shown to give an insight into the response of a 2D electron gas to the presence of a photocreated hole carrying charge and spin. It goes beyond what is observed in absorption and emission spectroscopy by separating direct and exchange contributions. In particular, our observations give further evidence of significant electron-hole exchange interaction, which remains to be explained theoretically. The resonant enhancements are of an order of magnitude stronger than resonant enhancement at the $E_0+\Delta_0$ band gap. This enables us to study intersubband excitations with very low power density and negligible heating of the electron gas.

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