Observation of Resonant Impurity States in Semiconductor Quantum-Well Structures

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(Received 30 January 1985)

Resonant Raman scattering experiments on GaAs(Si doped)-$Al_xGa_{1-x}As$ quantum-well structures show transitions involving the ground state of the donors and narrow resonant donor states derived from higher conduction subbands. These new impurity-related features, which occur at slightly higher energies than the associated conduction intersubband excitations, have been studied as a function of power density, temperature, and well width.

PACS numbers: 73.40.Lq, 71.55.Fr, 78.30.Gt

The properties of shallow donors and acceptors in semiconductor multiple-quantum-well (MQW) structures have recently attracted a great deal of attention.[1-20] In particular, numerous theoretical publications[1-12] have discussed the dependence of the impurity binding energy (E_B^0) on the well width (L), confinement-induced splittings of degeneracies, and broadening of the impurity spectrum originating in the nonequivalency of different positions within a well. Experimentally, these effects have been investigated by use of photoluminescence,[14-16,20,21] resonant Raman scattering[17] (RRS), and infrared[18,19] techniques. An important feature of quantum confinement of impurities that has so far escaped experimental confirmation is the expected occurrence of a ladder of *resonances,* i.e., impurity states derived from higher subbands overlapping in energy with the continuum.[10] These localized states are expected to strongly influence the transport properties of MQW structures by serving as hot-electron traps. The possibility that this trapping mechanism could lead to negative differential conductance has been previously considered in the literature.[22] Here, the layered structures show a major advantage over bulk semiconductors because their parameters have a wide range of tunability.

Resonant levels are known to couple only weakly to subband states in the closely related problem of excitons in MQW structures.[23] In this Letter, we report RRS data on GaAs(Si-doped)-$Al_xGa_{1-x}As$ MQW structures revealing the first clear evidence of narrow impurity resonances. The Raman spectra show, both, transitions into resonant donor states and intersubband excitations of photoexcited electrons. This allows a very precise determination of binding-energy differences that can be compared with theoretical predictions.

Two samples grown by molecular beam epitaxy on (001) GaAs were studied. They consist of thirty periods of 125-Å-thick $Al_xGa_{1-x}As$ and GaAs layers of thicknesses $L=238$ Å ($x=0.23$) and $L=460$ Å ($x=0.24$). The approximation that considers isolated quantum wells is valid here because of the relatively large thickness of the $Al_xGa_{1-x}As$ layers. Si donors were incorporated during growth at the center of the GaAs slabs with a dopant concentration of $n=5\times10^{15}$ cm^{-3}. The width of the donor spike in the samples is nominally $\approx L/3$. RRS experiments were performed with use of laser energies ω_L resonant with the optical gap derived from the $E_0+\Delta_0$ gap of GaAs. At $E_0+\Delta_0$, the scattering involving states derived from the conduction band is strongly enhanced.[24] Data were recorded in the $z(x',x')\bar{z}$ and $z(x',y')\bar{z}$ backscattering configurations, with x' and y' denoting [110] and [$\bar{1}$10] directions and z normal to the layers. The $z(x',x')\bar{z}$ geometry allows scattering by charge-density fluctuations (CDF).[24] Spin-density fluctuations (SDF) are allowed in the $z(x',y')\bar{z}$ configuration.[24]

The main result of our work is the observation of *doublets* associated with intersubband transitions of electrons in SDF spectra. Such SDF doublets are not observed in either undoped-[25] or modulation-doped[24] (donors in $Al_xGa_{1-x}As$) MQW structures and, consequently, stem from the presence of impurities in the well material, i.e., GaAs. The double-peak features can only be seen at low temperatures, for very low values of the power density P ($\lesssim$5–30 W cm^{-2}). The sample with $L=460$ Å shows $\epsilon_0\rightarrow\epsilon_1$ and $\epsilon_0\rightarrow\epsilon_2$ doublets at E_{01} (E'_{01}) = 8.0 (10.5) meV and E_{02} (E'_{02}) = 21.6 (23.4) meV, where ϵ_n is the energy of subband n. For this structure, a calculation using the band-gap discontinuities determined by Miller, Kleinman, and Gossard[26] predicts $\epsilon_1-\epsilon_0=6.9$ meV and $\epsilon_2-\epsilon_0=18.3$ meV. The $L=238$-Å sample exhibits two lines at E_{01} (E'_{01}) = 21.8 (24.0) meV associated with $\epsilon_0\rightarrow\epsilon_1$ excitations (the calculation gives $\epsilon_1-\epsilon_0=21.5$ meV). The $\epsilon_0\rightarrow\epsilon_2$ doublet is not well resolved in this sample. A relatively weaker feature

ascribed to $\epsilon_0 \rightarrow \epsilon_2$ scattering was observed at ~ 58 meV (the calculated value is 60.8 meV).

The temperature and P dependence of the doublets are very similar. They are, however, markedly different for the two components. Figure 1 shows the $\epsilon_0 \rightarrow \epsilon_1$ doublet in the $L = 460$-Å structure as a function of P. The intensity of the lower-energy feature E_{01} (normalized to the intensity of the longitudinal-optical phonon of GaAs) increases roughly linearly with P, for $P < 100$ W cm^{-2}. E'_{01} is no longer resolvable for P greater than ~ 50 W cm^{-2}. Below that value, its normalized intensity remains constant within experimental error. The normalized intensity of E_{02} in this sample, and of E_{01} in the $L = 238$-Å structure, is also proportional to P. The corresponding E'_{02} and E'_{01} features are comparatively less well defined, appearing already as shoulders for $P \gtrsim 10$ W cm^{-2}. The position of the primed components of the doublets does not depend on P. The unprimed companions show only minor shifts ($\lesssim 0.4$ meV) in the range extending up to $P \approx 3 \times 10^3$ W cm^{-2}.

The behavior of $z(x',x')\bar{z}$ CDF spectra (not shown) is different in several respects. First, CDF doublets could only be observed at the lowest end of the P range investigated. In this region, CDF and SDF spectra are basically the same except for differences in the relative intensity of the peaks. At intermediate densities, however, the CDF structures show a complex line shape that cannot be described as a simple superposition of two lines. At slightly higher P, the spectra in the two configurations exhibit only the unprimed peaks and become again almost identical. For still larger powers the CDF features shift to higher energies and broaden substantially.

The temperature dependence of the SDF spectrum of the $L = 460$-Å structure is shown in Fig. 2. The doublets associated with $\epsilon_0 \rightarrow \epsilon_1$ and $\epsilon_0 \rightarrow \epsilon_2$ transitions appear on top of a broad band due to $E_0 + \Delta_0$ luminescence. The feature of interest here is the strong decrease in the intensity of E'_{01} and E'_{02} with increasing temperature. The normalized intensity of the unprimed components does not vary much. Similar results were obtained for CDF spectra.

The dependence of the unprimed lines on power density and the fact that their positions are very close to the theoretical values indicate that they are due to intersubband transitions of photoexcited electrons occupying the lowest conduction well state. This assignment is supported by a comparison with early Raman results[25] on undoped GaAs-$Al_xGa_{1-x}As$ MQW struc-

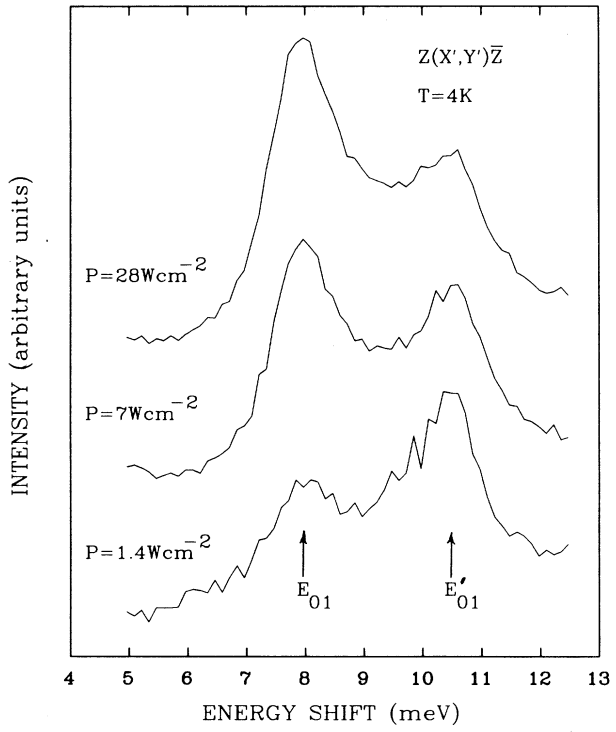

FIG. 1. Raman spectra of the MQW structure with $L = 460$ Å for different power densities, showing the doublet associated with $\epsilon_0 \rightarrow \epsilon_1$ transitions. The intensity has been normalized to the LO phonon of GaAs. Data were obtained for $\omega_L = 1.882$ eV.

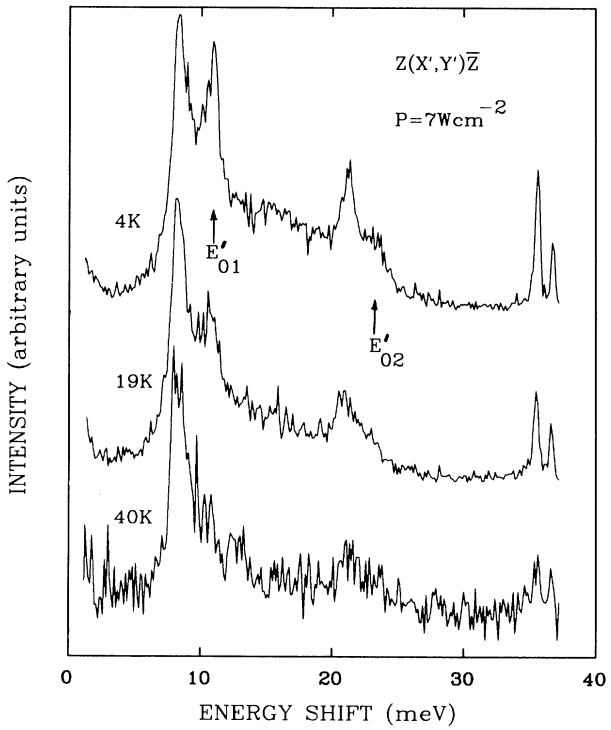

FIG. 2. Spectra of the $L = 460$-Å sample at several temperatures. Arrows indicate the position of donor-related features. The laser energy is $\omega_L = 1.882$ eV.

tures showing this form of scattering. The temperature behavior is also consistent with this interpretation.

The higher-energy components of the doublets are ascribed to transitions from the ground state of the Si donors (in the lowest subband) to the lowest-lying resonant impurity states derived from higher subbands. This identification is substantiated as follows. First, the rapid quenching of E'_{01}, E'_{02} with increasing temperature is similar to that of the $1s \rightarrow 2p$ donor feature in infrared absorption spectra.[19,27] Also, for $T = 40$ K, we calculate that $\sim 80\%$ of the impurities should be ionized. Scattering by donor states is also consistent with the lack of P dependence of the normalized intensity and the absence of doublets in modulation-doped[24] and undoped[25] MQW structures. Second, the position and line shape of the primed components rule out an assignment of transitions into band states. These transitions should give rise to a continuum with a low-energy cutoff positioned at E_B^0 (≈ 7.0 meV for $L = 460$ Å)[4] above the intersubband excitations. Third, our data are in very good agreement with reported calculated values of the binding energy of the *lowest* resonant level associated with a given subband, for donors at the center of the wells. In our interpretation, the separation of the two components of the doublets is equal to the difference between E_B^0 and the binding energy of the resonant state. The theoretical values of $E_B^0 - E_B^1 = 1.94$ meV, $E_B^0 - E_B^2 = 1.5$ meV (for $L = 460$ Å), and $E_B^0 - E_B^1 = 2.2$ meV (for $L = 238$ Å), obtained from a scaling of the results in Ref. 10, are very close to the measured splittings of 2.5, 1.8, and 2.2 meV, respectively.[28] The narrow width of the resonant transitions is also consistent with the theoretical prediction[10] of negligible coupling between the continuum and resonant donor levels.[29] The actual width reflects more likely the distribution of impurity sites in the wells rather than coupling effects.

Finally, we consider briefly the differences between CDF and SDF spectra (a more detailed analysis of the data will be discussed elsewhere). Within the random-phase approximation, SDF are totally unscreened.[24] The positions of the peaks in the SDF geometry should then correspond to *bare* transition energies which, in good agreement with our findings, do not depend on the concentration of photoexcited carriers, i.e., on P. Screening enters of course in CDF. The shift to higher energies of CDF lines at very large power densities is a well-known result of collective effects that become observable at relatively large concentrations of photoexcited electrons.[24,25] The behavior of CDF spectra at low P's, however, is not well understood. Tentatively, we associate the observed complex line shapes with a range of photoexcited-carrier densities for which the screening of the impurity transitions by the carriers is important. A clear understanding of the spectral line shapes will likely provide some information on the magnitude of this effect, which is a topic of current theoretical interest.[13]

The work was supported by the U.S. Army Research Office under Contracts No. DAAG-29-83-K-0131 and No. DAAG-29-82-K-0057, and the U.S. Office of Naval Research.

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[27]Preliminary Raman data on the sample with $L = 238$ Å show a similar temperature dependence for the $1s \rightarrow 2s$ peak.

[28]Final-state interaction effects, as considered by T. Ando for GaAs-$Al_xGa_{1-x}As$ heterojunctions [J. Phys. Soc. Jpn. **51**, 3893 (1982)], do not appear to be of importance in our case because of the very low density of photoexcited carriers (ρ). Values of ρ can be determined from the positions of intersubband peaks in SDF and CDF spectra (Ref. 24); the measured shifts were $\lesssim 1$ cm^{-1} giving an upper limit of $\rho \sim 5 \times 10^9$ cm^{-2}.

[29]For an impurity at the center of the well, the first resonance does not couple to the lowest subband because these states have different parity (see Ref. 10). In our samples, however, some coupling is expected because of the finite width of the impurity spike.