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Stark effect in GaAs-$Al_xGa_{1-x}As$ quantum wells: Light scattering by intersubband transitions

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The electric field dependence of $c_0 \to c_1$ intersubband transitions of photoexcited electrons has been studied in a 264-Å GaAs-$Al_{0.3}Ga_{0.7}As$ quantum-well structure using Raman spectroscopy. Experimental results for the Stark shifts are in very good agreement with theoretical predictions. The intensity of the intersubband peak increases rapidly with applied field due to parity mixing. In contrast to the enhanced broadening shown by exciton resonances, the width of $c_0 \to c_1$ is nearly independent of the field. This feature is attributed to effects of structural disorder.

Studies of the effect of electric fields perpendicular to the layers on the electronic properties of quantum-well structures (QWS's) have recently attracted much attention.[1-17] The interest is motivated in part by device applications (e.g., high-speed optical modulators)[2,8,10] which are based on the pronounced shifts of heavy-hole exciton resonances induced by the field. Quantum confinement allows well-defined resonances at shifts that are orders of magnitude larger than in the bulk, where field ionization limits the shifts to a small fraction of the binding energy.[3,6,7,16] Experimentally, the Stark effect has been investigated in GaAs-$Al_xGa_{1-x}As$ QWS's by use of optical absorption,[3,9,10] photocurrent,[13,14] excitation spectroscopy,[17] and photoluminescence[4,5,7,12,15] (PL) techniques. In this work, we report a Raman scattering (RS) study of the electric-field dependence of $c_0 \to c_1$ *intersubband* transitions of photoexcited electrons [c_0 (c_1) denotes the lowest (first excited) well state associated with the conduction band]. Our results are relevant to recent theoretical works on intersubband optical absorption[18] and on the possibility of using these excitations as the bases for field-tunable far-infrared photodetectors.[19] Stark shifts derived from our measurements are in very good agreement with theoretical calculations. The $c_0 \to c_1$ Raman peak shows an increase in intensity with applied field due to broken inversion symmetry, and a nearly field-independent width. The latter finding differs from the behavior shown by exciton resonances exhibiting field-enhanced broadening.[8,12,13] Disorder originating in structural defects is shown to account for this difference.

The sample used in our experiments was grown by molecular-beam epitaxy on a Si-doped (001) GaAs substrate in the following sequence: 0.3-μm Si-doped ($n = 2\times10^{18}$ cm^{-3}) GaAs buffer layer, 0.19-μm $Al_{0.3}Ga_{0.7}As$, 30 uncoupled GaAs wells with thickness $L = 264$ Å (198-Å-thick $Al_{0.3}Ga_{0.7}As$ barriers), and 0.19 μm $Al_{0.3}Ga_{0.7}As$ clad layers; unless indicated, the layers are nominally undoped. A semitransparent Schottky contact was formed by evaporating Au to build a 150-Å-thick film on top of the sample. Raman scattering measurements were performed at $T = 2$ K using a cw LDS698 (pyridin 1) dye laser, pumped by an Ar^+ laser. The excitation region covering the HH5 exciton resonance[20] [associated with the fourth excited conduction and heavy-hole (HH) states of the quantum wells] was investigated in detail. The laser beam was focused to a spot 200 μm in radius; powers were in the range 0.1–0.15 W. The estimated power density at the interface between the QWS and the clad layer is $P \approx 6-10$ $W cm^{-2}$; these values are inferred from a determination of the reflectivity of the coated sample ($\cong 70\%$) and the scattering intensity on and off the electrode. Raman spectra were recorded in the $z(x',x')\bar{z}$ and $z(x',y')\bar{z}$ backscattering configurations, where z is normal to the layers, and x' and y' are along the [110] and [1$\bar{1}$0] directions.

Figure 1 shows RS data of the QWS for different external dc voltages V_{ext}. The peak at 18.9 meV ($V_{ext} = 0$) is due to $c_0 \to c_1$ intersubband transitions of electrons. A calculation using the band-gap discontinuities determined by Miller *et al.*[21] predicts $c_0 \to c_1 = 18.4$ meV. In the range of V_{ext}'s investigated, $-3V \leq V_{ext} \leq 0.9$ V, the full width at half maximum (FWHM) of $c_0 \to c_1$ remained nearly constant at $\cong 3$ meV. With increasing $|V_{ext}|$, the intersubband peak grows in intensity and shifts to higher energies, except for $0 \leq V_{ext} \leq 0.2$ V. Because the shift and intensity must be even functions of the electric field, it follows that -0.2 V is the built-in voltage of the structure.

The observation of the $c_0 \to c_1$ transition indicates that the lowest subband in the sample is partially occupied. This is due to photoexcitation,[22] as revealed by results on the P dependence of the scattering (not shown). A crude estimate based on parameters from the literature[7] gives a steady-state electron concentration $\sigma \sim 2\times10^9$ cm^{-2} ($P = 10$ $W cm^{-2}$). σ can also be determined from the positions of the intersubband peak in the (x',x') and (x',y') configurations.[23] The bare transition energy is given by the latter, while (x',x') exhibits a shift proportional to σ due to depolarization effects.[23] Our data reveal no appreciable shifts (< 0.25 meV), which implies $\sigma < 5\times10^9$ cm^{-2}. This upper limit is consistent with the estimate above.

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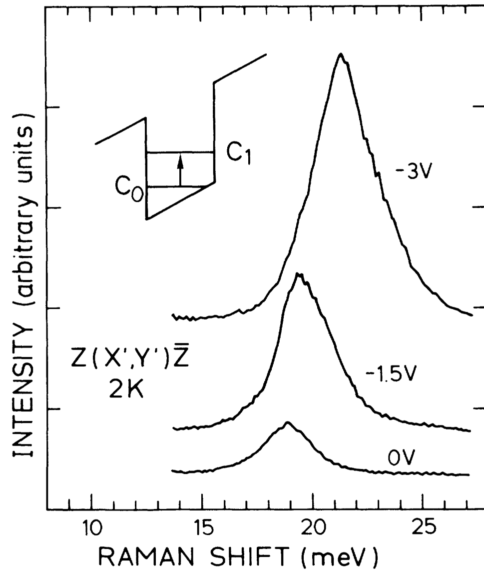

FIG. 1. Raman spectra of the 264-Å GaAs-$Al_{0.3}Ga_{0.7}As$ QWS showing the $c_0 \rightarrow c_1$ intersubband transition at different external voltages. The laser energy is $\omega_L = 1.685$ eV. Counting rates at the maxima are typically $\sim$400 counts $s^{-1} W^{-1}$. The inset shows a schematic energy diagram (not to scale).

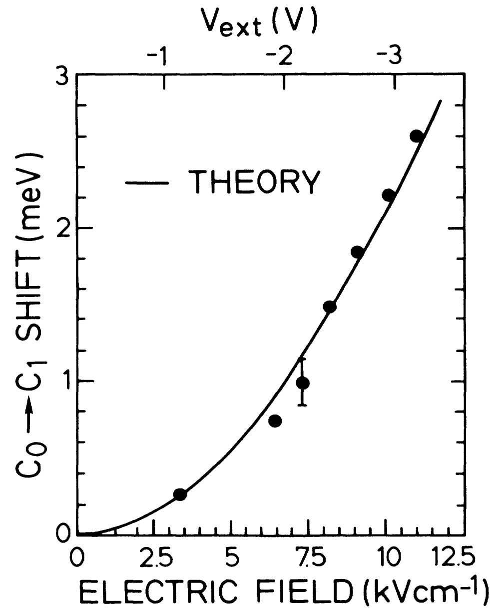

FIG. 2. Comparison between measured (solid circles) and calculated $c_0 \rightarrow c_1$ Stark shifts.

In the range of fields studied (see below), the electron-hole recombination time does not vary enough[7,15] to explain the increase in intensity shown by $c_0 \rightarrow c_1$ (which might be attributed to a larger steady-state population). At zero field, $c_0 \rightarrow c_1$ is strictly forbidden in RS because the states have different parities.[23] This indicates that the scattering enhancement results from parity mixing due to the field as in field-induced absorption by forbidden excitons.[10]

The V_{ext} dependence of the shift of $c_0 \rightarrow c_1$ is shown in Fig. 2, together with results of calculations. Theory and experiment are in very good agreement if a length of 2.7 μm is used to convert voltages into fields. In the calculations, the eigenenergies of quasi-bound-electron states were obtained numerically. We used the procedure described in Ref. 11 to avoid imaginary corrections to the energy due to tunneling, which are negligible in our case.

The observation of a nearly field-independent width of $c_0 \rightarrow c_1$ is a significant feature of our results. This behavior differs significantly from the reported[8,12,13] field-induced enhanced broadening of exciton resonances, which is mainly determined by interface roughness and interwell size fluctuations[12] (tunneling effects are relatively unimportant in most cases studied[2-5,7-10,12-15] except at very high fields; they are vanishingly small in our structure). Structural defects lead to a clear differentiation between the field-dependent widths of excitons and intersubband transitions. The argument is simple; broadening due to fluctuations in the average well width L, and also due to island roughness under conditions of localization,[24] is approximately given by

$$\Gamma(E) = \frac{\partial \Omega(E)}{\partial L} \delta L, \tag{1}$$

where E is the field and Ω is the energy of the corresponding excitation. For excitons[11,12] Γ *increases* with E while it *decreases* for intersubband transitions. Calculations using the parameters of our structure and $\delta L = 2.83$ Å (one monolayer) give $\Gamma = 0.4$ meV ($E = 0$) and $\Gamma = 0.3$ meV ($E = 12$ kV cm^{-1}). Because δL does not commonly exceed 3–4 monolayers,[24] it is clear that other mechanisms (such as impurity scattering) contribute to the $c_0 \rightarrow c_1$ width.

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