PHYSICAL REVIEW B VOLUME 34, NUMBER 10 15 NOVEMBER 1986

Doubly resonant LO-phonon Raman scattering via the deformation potential with a single quantum well

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(Received 3 July 1986)

Doubly resonant Raman scattering (DRRS) due to LO phonons where the incoming and outgoing photons are resonant with the $n=1$ light- and heavy-hole free excitons, respectively, has been observed at low temperature with a very-high-quality single GaAs quantum well of estimated thickness 28.3 Å. The measured circular and linear polarization characteristics of this DRRS show that it is dominated by the deformation-potential process modified by considerable spin relaxation and an admixture of about 8% Fröhlich scattering. In contrast to bulk GaAs, interference between these two processes is not observed or expected for DRRS.

Doubly resonant Raman scattering (DRRS) where the incoming and outgoing photons are both resonant with heavy-hole exciton transitions that differ in energy by $\hbar\omega_{LO}$ has been observed previously with GaAs-$Al_xGa_{1-x}As$ quantum wells.[1] A theoretical treatment of this intrinsic single-LO-phonon first-order DRRS led to the conclusion that the observed strength could be accounted for by the Fröhlich (electrostatic polar) interaction[2] (F process) and that the deformation-potential contribution[2] (D process) was negligible in this case. DRRS has an important advantage from a theoretical point of view in that it involves Raman processes with well-defined initial and final exciton states. This Communication describes the first demonstration of intrinsic DRRS due to the deformation potential. The DRRS observed involves a light hole[1,3] and was obtained with a very-high-quality single GaAs quantum well clad by $Al_{0.3}Ga_{0.7}As$ barriers where the well thickness L, estimated to be 10 ML wide (28.3 Å), is such that the observed $n=1$ heavy- and light-hole exciton transitions E_{1h} and E_{1l}, respectively, satisfy the condition $E_{1l}-E_{1h}=\hbar\omega_{LO}$. Experimental determinations of the DRRS Raman intensities for various polarizations indicate that this DRRS is dominated by the D process.

The single quantum well was grown by molecular-beam epitaxy (MBE) at 600 °C in a Gen II MBE station on a (100) semi-insulating GaAs substrate mounted on a rotating substrate holder. The MBE growth was interrupted for two minutes at each of the two GaAs-$Al_{0.3}Ga_{0.7}As$ interfaces. Earlier work[4,5] has shown that this type of growth interruption can result in photoluminescence characteristics that reflect vastly improved interfaces as is the case here.

Photoluminescence (PL) and excitation spectra at ≈ 6 K demonstrate the high quality of this quantum well. The spectra were obtained in the backscattering direction 24° off normal incidence with a detection resolution full width at half maximum (FWHM) of 0.2 meV. Excitation was with a tunable cw dye laser (FWHM ≈ 0.38 meV). Circular and linear polarization techniques were employed to observe optical alignment and circular polarization.

The PL and excitation spectra both show multiple well-resolved peaks due to changes in well thickness by one-monolayer (ML) steps. An excitation spectrum is shown in Fig. 1 with detection set at 1.688 eV, the low-energy side of the E_{1h} exciton emission peak for the well of estimated width 11 ML. The E_{1h} and E_{1l} exciton peaks are clearly seen for both the 10- and 11-ML parts of the well. The FWHM of ≈ 3.5 meV of the E_{1h}(11-ML) peak is much less than the observed 16-meV change in E_{1h} for a change in L of 1 ML. This demonstrates that the inter-

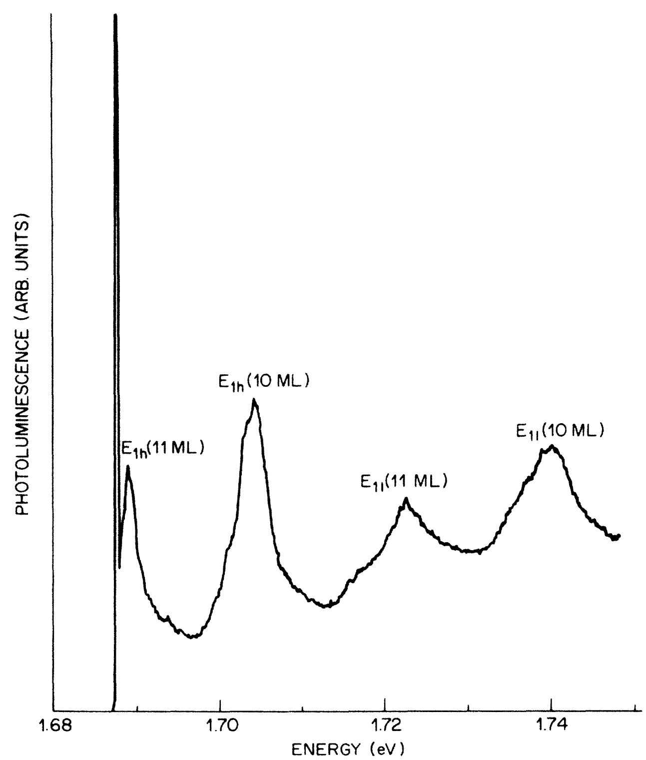

FIG. 1. Excitation spectrum at 6 K for a single GaAs quantum well with large area regions 10 and 11 ML wide. The $n=1$ heavy- and light-hole excitons, E_{1h} and E_{1l}, respectively, are labeled. The configuration was $z(y,x)\bar{z}$ and the excitation intensity ≈ 24 W/cm^2. The energy separation E_{1l}(10 ML)-E_{1h}(10 ML) $=\hbar\omega_{LO}$.

faces consist of very smooth ML steps large in lateral extent compared to the excitons. Of most interest for the present work is the near coincidence of E_{1l}(10 ML)-E_{1h}(10 ML) with $\hbar\omega_{LO}$=36.7 meV for bulk GaAs (Ref. 6) which should therefore give rise to DRRS.

PL and excitation spectra demonstrating DRRS are shown in Fig. 2. Directions in the plane of the well are defined by $\hat{x}\parallel[100]$, $\hat{y}\parallel[010]$, $\hat{x}'\parallel[110]$, and $\hat{y}'\parallel[1\bar{1}0]$; and $\hat{z}\parallel[001]$ is normal to the well.[2,3] The data in Fig. 2 are for excitation-detection configurations $z(y,x)\bar{z}$ and $z(x,x)\bar{z}$. Before recording these spectra, the DRRS signal for $z(y,x)\bar{z}$ was optimized by adjusting the detected E_{1h}(10-ML) photon energy and the exciting E_{1l}(10-ML) photon energy E_p for the maximum intensity. This led to excitation at E_p=1.7402 eV and detection at 1.7045 eV, which gives an energy difference of 35.7 meV±0.3 meV. For the PL spectra, the detector sensitivity was reduced by a factor of 10 for the E_{1h}(11-ML) luminescence. The roughly comparable strengths of the 10- and 11-ML peaks in Fig. 1 coupled with the very strong tendency to emit at E_{1h}(11 ML) even for resonant excitation at E_{1l}(10 ML) as shown in Fig. 2, is convincing evidence of extensive lateral motion of the excitation from the 10-ML parts of the well to the regions where L=11 ML. The temperature dependence and the excitation spectrum of the PL peak at 1.686 eV show that it is due to bound E_{1h}(11-ML) excitons.

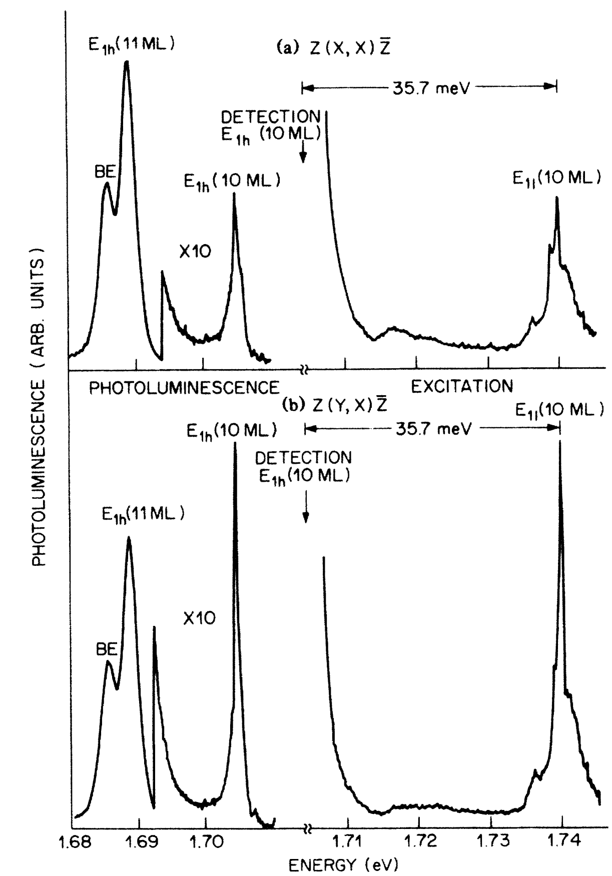

FIG. 2. Photoluminescence and excitation spectra at 6 K showing DRRS with excitation at 1.7402 eV (E_{1l}) for the PL spectra and detection at 1.7045 eV (E_{1h}) for the excitation spectra. The polarization configurations (a) $z(x,x)\bar{z}$ and (b) $z(y,x)\bar{z}$ with the larger DRRS signals from the $z(y,x)\bar{z}$ configuration show that the deformation-potential contribution is dominant. The peak in PL labeled BE is due to bound E_{1h}(11-ML) excitons.

The sharp DRRS peak, FWHM of ≈0.5 meV, shows up most clearly in the PL which has therefore been utilized to estimate its strength. This is done by subtracting the nonresonant part of the E_{1h}(10-ML) emission. A summary of the various DRRS polarization intensities estimated in this manner is given in Table I. The different configurations were obtained with the same sample, incident beam, and detection system, by changing only the input and output polarizers. Note that the negative circular polarization (0.21-0.79)/(0.21+0.79)=−0.58 and the optical alignment

$$\frac{I(z(x,x)\bar{z})-I(z(y,x)\bar{z})}{I(z(x,x)\bar{z})+I(z(y,x)\bar{z})}=-0.56$$

are consistent with DRRS via the D process, which by itself would give −1 for both these quantities. The observed DRRS width and the laser excitation width suggest a true FWHM for the process of ≈0.3 meV assuming Gaussian broadening.

The E_{1l}(11-ML)-E_{1h}(11-ML) splitting as obtained from Fig. 1 is 33.5 meV, 2 meV smaller than the corresponding 10-ML quantity. A FWHM of ≈3 meV was measured for the DRRS from the 10-ML regions of the well. This suggests that DRRS may also occur with the two excitons from the 11-ML parts of the well. However, 33.9 meV is the energy of the TO phonon of bulk GaAs (Ref. 6) which as shown earlier[3] takes part only in (x,z) and (y,z) spectra of quantum wells, and hence DRRS is not expected in the backscattering geometry used here. In fact, DRRS was not obseved from the 11-ML parts of the well.

To interpret the data we postulate a fraction $(1-f)$ due to the intrinsic D process, the remaining fraction f due to various (intrinsic, impurity assisted, bandmixing assisted) F processes, and relaxation times τ_0,τ_1,τ_2, representing, respectively, the virtual exciton lifetime, the decay time for population differences in the exciton polarization states, and the dephasing time for coherence of these states. The relevant F processes probably involve impurity scattering

TABLE I. Measured intensities of the single-LO-phonon DRRS for various excitation-detection polarization configurations. The measured values are normalized to make $I(z(+,+)\bar{z})+I(z(+,-)\bar{z})=1.00$.

Configuration	Intensity
$z(+,+)\bar{z}$	0.21
$z(+,-)\bar{z}$	0.79
$z(x,x)\bar{z}$	0.23
$z(y,x)\bar{z}$	0.82
$z(x',x')\bar{z}$	0.84
$z(y',y')\bar{z}$	0.95
$z(y',x')\bar{z}$	0.18

and produce, unlike the D process, LO phonons of nonzero momentum along the well (FI process). The internal consistency of the data can be checked against the two conservation relations[1] (omitting for brevity $z,\bar{z}$),

$$I(x,x)+I(y,x)=I(+,+)+I(+,-)$$
$$=I(y',x')+\tfrac{1}{2}[I(x',x')+I(y',y')] \quad (1)$$

Upon putting in numbers from Table I this becomes

$$(1.05)\approx(1.00)\approx(1.075) ,$$

which gives an indication of the accuracy of the data. A simple model of relaxation leads to the relations

$$\frac{\tau_0}{\tau_2}=\frac{2I(y',x')}{1-2I(y',x')}=0.56 , \quad (2)$$

$$f=I(x,x)+[I(x,x)-0.5](\tau_0/\tau_2)=0.078 , \quad (3)$$

$$\frac{\tau_0}{\tau_1}=\frac{I(+,+)-f}{0.5-I(+,+)}=0.46 , \quad (4)$$

assuming the normalization $I(+,+)+I(+,-)=1$ and the validity of Eq. (1). Neither band mixing nor exchange coupling can explain the data; a relaxation model is required.

The interference between the two mechanisms is measured by the quantity

$$\tfrac{1}{4}[I(y'y')-I(x',x')]\lesssim[f(1-f)]^{1/2}(1+\tau_0/\tau_2)^{-1} , \quad (5)$$

which from Table I becomes

$$(0.03)\lesssim(0.17) .$$

The limit on the right is for the case (not expected) where both mechanisms involve the same phonon. The numbers indicate that interference is negligible in the present case. This is consistent with the following facts: (a) the FI processes cannot interfere with the D process because of their nonzero phonon momentum along the well, and (b) if the quantum well has reflection symmetry in the xy plane, the intrinsic F process cannot interfere with the D process in the present case because they require phonons of opposite parity.

The LO-phonon energy observed here $\hbar\omega_{LO}=35.7$ meV lies below the bulk GaAs value 36.7 meV. Previously, with much wider wells ($L\approx140$ Å) we observed[1] the bulk value in DRRS. However, for GaAs-AlAs superlattices a lowering of energy has been seen experimentally[7] and predicted theoretically.[8] In the present case of a single quantum well embedded in a $Al_{0.3}Ga_{0.7}As$ alloy, the phonon may not be a truly confined GaAs mode but may be a wave packet of alloy modes whose shape is given by (for the D process) the product of the $1h$ and $1l$ confinement wave functions. We note that LO for the alloy[9] is about 1.5 meV below that of GaAs, and the narrow well ($L\approx28.3$ Å) should have considerable penetration into the barrier. We calculate that the product of hole confinement functions gives a weighting of 20% for the alloy and 80% for the GaAs, so we cannot account in this way for a downward shift larger than 0.5 meV. The remaining 0.5 meV must come from the presence of high momentum components in the phonon, which because of the dispersion of the LO branch have lower energy than $\hbar\omega_{LO}$.

It is a pleasure to acknowledge several useful discussions with Dr. Aron Pinczuk and the support of Dr. A. S. Jordan.

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