Resonant Raman scattering in an InAs/GaAs monolayer structure

J. Maultzsch, S. Reich, A. R. Goñi, C. Thomsen

Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstr. 36, 10623 Berlin, Germany

Abstract We studied the excitation-energy dependence for Raman scattering on the GaAs optical phonon modes in an InAs/GaAs heterostructure. The GaAs LO phonon scattering exhibits a resonance with the InAs optical transitions, the Raman profiles depending on the polarization of the incoming light. We interpret our results as due to incoming resonance with E1-HH1 and the E1-LH1 transition of the monolayer. In addition, we demonstrate that the scattering intensity is influenced by an inhomogeneous exciton linewidth due to fluctuations in the well width.

1 Introduction

The vibrational properties of low-dimensional semiconductor structures have been widely studied by Raman spectroscopy.[1] Resonant-Raman experiments yield information also about the electronic properties as the scattering intensity is enhanced if the incoming or outgoing photon energy matches an allowed electronic transition. We consider a monolayer structure from zincblende-type materials as characterized by the point group D_{2d}. The reduction of dimensionality leaves the symmetry of the electron state (E1) at the condcuction band minimum unchanged (A_1), whereas the degeneracy of the hole states at the Γ point is lifted. The heavy hole state has E symmetry and is twofold degenerate; the light hole state has B_2 symmetry.

We present resonant-Raman scattering experiments on the GaAs optical phonon modes in an InAs/GaAs monolayer structure for different scattering geometries. We also investigated the dependence of the Raman profile on the exciton coherence lifetime as previously described by Shields *et al.*[2]

2 Experiment

The sample consisted of a single InAs layer (effective thickness about 1.5 ML), embedded in bulklike GaAs with two AlGaAs cladding layers forming a waveguide. The InAs- monolayer fundamental transition is about 100 meV below the GaAs bandgap. The light and heavy-hole state are separated energetically by a few meV [3].

Raman experiments were performed at low temperatures (2-40 K) in backscattering geometry with the light propagating along the monolayer plane in $(\bar{1}10)$ direction and polarized parallel or perpendicular to the monolayer. The inset of Fig. 1 shows a schematic picture of the setup and the coordinate system used. Raman spectra were excited with a Ti-Sapphire laser, tuned from 1.41 eV to 1.435 eV. The scattered light was analyzed by a triple-grating spectrometer and detected with a charge-coupled-device (CCD) camera.

3 Results

In Fig. 1 we show Raman spectra recorded at 5 K with an excitation energy of 1.424 eV. The lowest trace shows a spectrum in $y'(x',x')\bar{y}'$ scattering geometry. We find the GaAs LO phonon (296 cm^{-1}), which for bulk GaAs is forbidden in this configuration, to be 10–20 times stronger than the allowed TO (273 cm^{-1}) mode. For $y'(z,z)\bar{y}'$ geometry and crossed polarizations the LO scattering is observed as well, although much weaker. Note that the Raman tensor of the LO phonon is not symmetric; the scattering intensity is different in the two crossed polarizations. By varying the excitation energy we found a resonance of the GaAs LO-phonon with the InAs optical transitions, while the TO scattering intensity was almost constant. Fig. 2 shows Raman profiles in $y'(x',x')\bar{y}'$ and $y'(z,z)\bar{y}'$ geometries, along with a photoluminescence (PL) and a photoluminescence excitation (PLE) spectrum. The maxima of the Raman profiles are at 1.424 eV for $y'(x',x')\bar{y}'$ and at 1.428 eV for $y'(z,z)\bar{y}'$ configuration; we observed the same energy shift of $\Delta = 4$ meV between the two cross-polarized spectra. The position of the Raman profile depends only on the polarization of the incoming light, i.e., 1.424 eV for (x',x') and (x',z) and 1.428 eV for (z,z) and (z,x') polarized light. The PLE peak in Fig. 2 at 1.439 eV corresponds to the HH1-exciton of the InAs monolayer. The photoluminescence maximum is shifted by about 20 meV to lower energy due to the relaxation of free carriers into localized, lower-energy states. Likewise, the Raman profiles are shifted to the *red* with respect to the PLE peak.

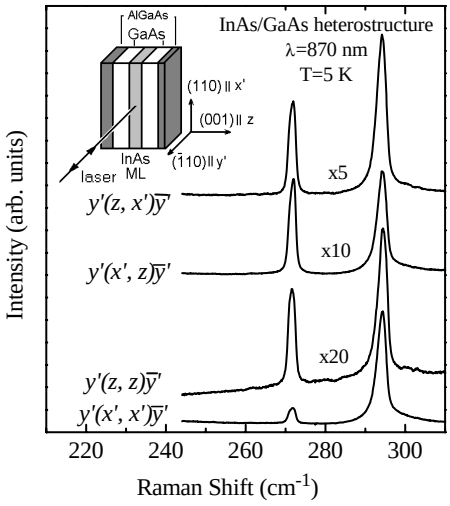

Fig. 1 Raman spectra recorded at T=5 K with an excitation energy of 1.424 eV. The scattering geometry is given in Porto's notation. The inset shows a sketch of the sample and the scattering geometry.

4 Discussion

As was shown by Shields *et al.* for a GaAs/AlAs multiple quantum well, the energetic separation between the Raman

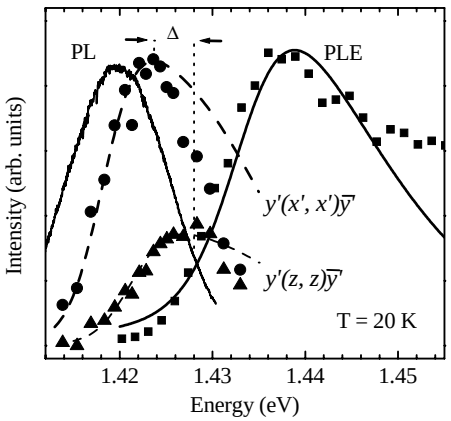

Fig. 2 Photoluminescence (PL), photoluminescence excitation (PLE) spectra, and Raman profiles of the 1.5 ML InAs/GaAs heterostructure. The PL and PLE spectra were recorded at 20 K with the light polarized parallel to the monolayer plane (x'). The Raman profiles show the scattering intensity of the GaAs LO-phonon mode normalized to the TO scattering intensity as a function of incoming photon energy. The solid and dashed lines were calculated with Eq. 2 and 3, respectively.

profile and the PLE peak can be attributed to an inhomogeneous broadening by fluctuations in the well width.[2] They assumed that an exciton at energy E_h with a homogeneous linewidth Γ_h can scatter with equal probability to all energetically lower, wider-well regions. Then $\Gamma_h(E_h)$ increases with E_h as the number of exciton states below E_h. Taking a Gaussian distribution $G(E_h)$ of homogeneously broadened exciton states centered at E_i and with a half-width Γ_i [2]

$$\Gamma_h(E_h) = \int_{-\infty}^{E_h} G(E, E_i, \Gamma_i)\, dE = \Gamma_0 + \frac{1}{2}\Gamma_1 \{1 + \mathrm{erf}(\frac{E_h - E_i}{\sqrt{ln2}\Gamma_i})\}, \quad (1)$$

where $\mathrm{erf}(x)$ is the error function, Γ_0 the low-energy limit of the homogeneous linewidth Γ_h, and Γ_1 the difference between the low and the high-energy limit. In the approximation that the PLE spectrum resembles the absorption, the PLE intensity is given by

$$I_{PLE}(E) \propto \int_{-\infty}^{\infty} \frac{\Gamma_h \cdot G(E_h, E_i, \Gamma_i)}{\pi[(E_h - E)^2 + \Gamma_h^2]} dE_h. \quad (2)$$

The Raman scattering intensity for incoming resonance with the Gaussian-like distributed exciton states of energy E_h may be written as

$$I_{RRS}(E) \propto \int_{-\infty}^{\infty} \frac{G(E_h, E_i, \Gamma_i)}{\pi[(E_h - E)^2 + \Gamma_h^2]} dE_h. \quad (3)$$

The PLE spectrum and the resonant Raman profile calculated with Eq. 2 and 3 are shown in Fig. 2. The parameters were $E_i = 1.444$ eV for the $y(x', x')\bar{y}$ Raman profile, $\Gamma_i = 14.5$ meV, $\Gamma_0 = 2 \cdot 10^{-6}$ eV, and $\Gamma_1 = 1 \cdot 10^{-2}$ eV. It is nicely seen, that the energy shift between the PLE and the Raman profile peak is reproduced well by this calculation. We find though, that the measured Raman profile is less asymmetric than the calculated one. A possible reason is that the measured PLE spectrum resembles more the exciton continuum state than the ground state. The energetic difference between the Raman profiles and the PLE peak should actually be lowered by the exciton binding energy. A smaller energy difference in our calculations results in a less asymmetric Raman profile, as we find experimentally.

The energy shift $\Delta = 4$ meV for different polarizations of the incoming light can be explained by an incoming resonance with the two different optical transitions in the monolayer. Due to the symmetry of the electronic states x' polarized light interacts mainly with the E1-HH1 exciton, z polarized light with the E1-LH1 exciton of the monolayer [3]. Fröhlich-induced LO scattering then is allowed for $y'(x', x')\bar{y}'$ and $y'(z, z)\bar{y}'$ configuration in resonance with the E1-HH1 (lower excitation energy) and the E1-LH1 transition (higher excitation energy), respectively. In cross-polarized configuration mixing of the heavy and light-hole states can lead to Fröhlich-induced scattering involving defects [4]. Taking into account the energy separation between the heavy-hole and light-hole state, we calculated the $y'(z, z)\bar{y}'$ Raman profile in Fig.2 with the same parameters as given above, but with the center E_i of the distribution at $E_i = 1.448$ eV. This difference between the heavy- and the light-hole state (4 meV) obtained by fitting the Raman profiles agrees well with the value determined by luminescence experiments.[3]

5 Conclusion

In conclusion, we studied Raman scattering by the GaAs LO phonons in an InAs/GaAs monolayer structure in resonance with the InAs optical transitions. The Raman profiles were found to depend on the polarization of the incoming light due to incoming resonance with the E1-HH1 and the E1-LH1 transition. This is consistent with the symmetry of the electronic states in the monolayer predicted by group theory. We were able to reproduce well the measured Raman profiles and their peak energies together with PLE spectra by a calculation taking into account the influence of the exciton lifetime on the scattering intensity. Compared to the superlattice studied by Shields [2] our monolayer shows larger well-width fluctuations as evidenced by the larger Γ_1 which we obtained from our fits to the data.

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