Resonant Raman scattering in InGaAs/AlAs quantum dots

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We report resonant Raman scattering from InGaAs/AlAs quantum dots with indium content $x = 0.3 \div 1$. The first order Raman spectra reveal resonant behaviour selective to a quantum dot size. Up to third-order multi-phonon processes have been observed in the quantum dot structure.

1 Introduction

Elementary vibrational excitations in nanostructures with self-assembled quantum dots (QD’s) attract a significant interest from both experimental and theoretical points of view. In potential optoelectronic devices based on quantum dots the knowledge of the phonon spectrum is of great importance because phonons influence the relaxation rate of photoexcited electrons and the mobility of charge carriers. Raman spectroscopy is a widely used method for studying phonons in nanostructures. First-order Raman scattering by optical, acoustic and interface phonons in InAs QDs embedded in AlAs or GaAs matrices is investigated in detail [1–6] while the data on QDs consisting of a solid solution are rather rare [7–9]. The Raman study of optical phonons localized in In$_x$Ga$_{1-x}$As QDs with $x = 0.3; 0.35$ and two-dimensional layers under resonant excitation was recently reported [7]. Folding of acoustic phonons was studied in periodical InGaAs/AlGaAs QD structures [8]. Very recently, intermixing in In$_{0.5}$Ga$_{0.5}$As/GaAs as well as in InAs/GaAs QD structures was probed using Raman spectroscopy [9]. Even less is known on multiple-phonon Raman scattering in QDs [7, 10] despite multiple-phonon processes are important for electronic relaxation and can be employed to solve the problem of degradation of photoluminescence in QDs.

This paper presents the results of resonant first-order and multiple-phonon Raman scattering by optical and interface phonons in InGaAs/AlAs QDs having a wide range of indium content ($x = 0.3 \div 1$).

2 Experimental

The structures consisting of 10 periods of InGaAs QDs embedded in AlAs were grown by molecular beam epitaxy on (001)-oriented GaAs substrates utilizing Stranski-Krastanov growth mode at the substrate temperature of 500 °C. Each period contains an InGaAs QD layer with $x = 0.3, 0.5, 0.75, 1.0$ and nominal thickness of 8.7, 5.3, 2.6, 2.25 monolayers, respectively, and a 23.5 nm-thick AlAs layer.

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3 Results and discussion

The structures under investigation were characterized by a cross-sectional transmission electron microscopy (TEM). Figure 1 shows the TEM images of samples with $x = 0.3$ and $x = 0.5$. While InAs/AlAs structures reveal a pyramid-like shape of QDs with the base length about 10 nm and the height of about 1.5 nm [1], InGaAs QDs have larger size (base size (height) of about 15 nm (3+4 nm) and 20+30 nm (4+6 nm) for $x = 0.75$ and $x < 0.75$, respectively) and lens-like shape. The fluctuation of QD size within a QD layer is about 20%. The size of QDs gradually increases from the bottom towards the top QD layers producing additional inhomogeneity.

The first-order Raman spectra (Fig. 2a) reveal features near 240±250, 270±290 and 400 cm\(^{-1}\) related to longitudinal optical InAs- and GaAs-like (denoted as $I_{LO}^\text{InAs}$ and $G_{LO}^\text{GaAs}$) phonons from InGaAs QDs and to optical phonons from AlAs layers (not shown in Fig. 2). The features located between LO and TO phonon frequencies near 240, 280 and 380 cm\(^{-1}\) were attributed to InAs-, GaAs-, AlAs-like interface phonons ($I_{IF}^\text{InAs}$, $G_{IF}^\text{GaAs}$ and $A_{IF}^\text{AlAs}$, respectively) [5]. The frequency positions of InAs- (GaAs-) like optical phonons are shifted towards lower (higher) frequencies with decreasing $x$. This shift of the optical phonons

![Figure 1](image1.png)

Fig. 1 Cross-sectional TEM images of In\(_x\)Ga\(_{1-x}\)As/AlAs QD samples with a) $x = 0.3$ and b) $x = 0.5$.

The Raman spectra were recorded at $T = 20$ K in backscattering geometries from planar surfaces excited by different lines of Ar\(^{+}\) and Kr\(^{+}\) lasers. The scattering configurations employed were $z(x)\overline{z}$ and $z(x)\overline{z}$ with $x$, $y$ and $z$ parallel to the [100], [010] and [001] directions, respectively.

![Figure 2](image2.png)

Fig. 2 Raman spectra of In\(_x\)Ga\(_{1-x}\)As/AlAs QD structures with $x = 0.3$, 0.5, 0.75, 1 measured in the $z(x)\overline{z}$ scattering geometry with 528.7 and 647.1 nm laser excitation lines at 20 K.
indicated in Fig. 2a by arrows is consistent with a two-mode behavior of optical phonons in InGaAs taking into account a compressive strain in QDs [11]. A detailed analysis of build-in strain in InGaAs/AlAs QD will be published elsewhere. The peak at the low-frequency side of the InAs-like phonon feature observed for InGaAs QDs with x = 0.3 and 0.5 is attributed to InAs-like TO phonons.

With increasing excitation energy a red shift of pure InAs and GaAs-like LO phonons in QDs is observed (Fig. 2a for the sample with x = 0.5 and Fig. 3). The intensity of InAs phonons approaches a maximum at about 2.92 eV conforming that the resonant conditions are achieved. For InAs QDs (Fig. 3a) the shift of InAs-like LO phonons is consistent with previously published data [1, 3] and is explained by selective resonant Raman scattering. According to the proposed model, large dots having relatively lower energies of electronic transitions contribute to the Raman spectra mostly for excitation lines in the red spectral range which are closer to the resonance with electronic transitions. The smaller QDs have higher energies of electronic and hole ground states. This leads to increasing relative contribution to the Raman spectra at higher excitation energies. In smaller-size dots phonon confinement effect becomes more significant and, therefore, causes a noticeable decrease of the phonon frequencies due to the negative dispersion of the optical phonons.

Raman scattering by InAs-like phonons in the case of InGaAs QDs has lower intensity compared to InAs QDs and therefore InAs-like were not observed at off-resonant conditions (at the energies above 1.92 eV). However, GaAs-like LO features due to GaAs-like LO phonons are clearly seen for different excitation energies (Fig. 3b). Moreover, their frequency position decreases with increasing excitation energy as in case of InAs QDs thus confirming selective resonant Raman scattering by InGaAs QDs.

Since the photon energy of a 647.1 nm line is larger than that of the photoluminescence (PL) band of the samples (for x=1 the maximum of PL is about 1.6 eV), outgoing resonant conditions are satisfied, i.e. the energy of the scattered light equals an electronic transition energy. Therefore, high-order resonant Raman scattering from phonons is enhanced and observed in the spectra of Fig. 2b. It is worth to mention that the high-order spectra are almost the same for both polarizations, while the one-phonon spectra reveal in addition LO phonons of AlAs in z(xx) scattering geometry (not shown here). The off-resonant Raman spectrum of the structure with x = 0.5 measured with 528.7 nm laser line is shown in Fig. 2a for comparison.

In the vicinity with $E_0$ resonance in QDs, up to third-order scattering is observed (Fig. 2b), involving both pure overtones of the first-order optical and interface InAs, GaAs and AlAs phonons and combination of phonons from the materials. The assignment of the high-order phonons is made according to their fundamental modes with an error of less than 2 cm$^{-1}$. The vibrational energies of these modes are the sum of the corresponding first-order modes. For the sample with x = 0.3 the feature denoted as 2G reproduces

![Fig. 3](image-url)  
**Fig. 3** Raman spectra of In$_x$Ga$_{1-x}$As/AlAs QD structures with a) x = 1 and b) x = 0.3 measured in the $z(xx)z$ scattering geometry with different excitation energies at 20 K.
the structure of that of the first-order GaAs-like phonons with doubled frequency. The PL signal gives rise to an intensive background gradually increasing towards higher frequencies. Note, that this background was subtracted for clarity. To our knowledge, this is the first observation of the scattering by a combination of phonons arising not from different phonon branches but from modes of QD and matrix materials. Similar results were reported only for short-period GaAs/AlAs superlattices grown along the [001] and [012] directions [12, 13]. Overtones of surface optical phonons were observed also for ZnO nanocrystallites [14] indicating a strong exciton-Fröhlich phonon coupling. The resonance features observed in multiphonon Raman spectra can be explained in the framework of a “cascade model” [15]. According to this model, a relaxation process of hot exciton comprises the following steps: (i) the absorption of the incident photon with the successive excitation of an exciton, (ii) the relaxation of this exciton into lower energy states with emission of LO phonons through a cascade-type process, and (iii) the radiative recombination of the exciton with the emission of the scattered photon with an overtone energy. Thus, the inelastic scattering occurs as a cascade process in which excitons take part only as virtual intermediate states. That is different from photoluminescence process for which the relaxation involves real exciton states in accordance with the energy and momentum conservation laws.

4 Conclusions

Resonant Raman scattering by self-organised InGaAs/AlAs QD structures was investigated. First-order Raman spectra reveal two-mode behavior for optical phonons localised in InGaAs QDs. The frequency position of InAs and GaAs-like phonons increases with increasing excitation energies. This effect is attributed to size selective Raman scattering by inhomogeneous QD array. Under the outgoing resonant condition, multiple phonon scattering occurs at combinational frequencies of optical and interface phonons of the QD and matrix materials.

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