Defense Technical Information Center Compilation Part Notice

ADP013174

TITLE: Size-Selective Raman Scattering in Self-Assembled Ge/Si Quantum Dot Superlattices

DISTRIBUTION: Approved for public release, distribution unlimited
Availability: Hard copy only.

This paper is part of the following report:

TITLE: Nanostructures: Physics and Technology International Symposium [9th], St. Petersburg, Russia, June 18-22, 2001 Proceedings

To order the complete compilation report, use: ADA408025

The component part is provided here to allow users access to individually authored sections of proceedings, annals, symposia, etc. However, the component should be considered within the context of the overall compilation report and not as a stand-alone technical report.

The following component part numbers comprise the compilation report:
ADP013147 thru ADP013308

9th Int. Symp. "Nanostructures: Physics and Technology"
St Petersburg, Russia, June 18–22, 2001

Size-selective Raman scattering in self-assembled Ge/Si quantum dot superlattices

A. G. Milekhin†, A. I. Nikiforov†, O. P. Pchelyakov†, S. Schulze‡ and D. R. T. Zahn‡

† Institute of Semiconductor Physics, RAS, 630090 Novosibirsk, Russia
‡ Institut für Physik, Technische Universität Chemnitz,
D-09107 Chemnitz, Germany

Abstract. Self-organised Ge quantum dot (QD) superlattices having properties of two- and zero-dimensional structures were investigated by Raman spectroscopy. Longitudinal optical Ge (LO) and Ge-Si (L) phonons and folded acoustic (LA) phonons superimposed with a strong continuous emission were studied under resonant conditions. The measured phonon frequencies of folded LA phonons up to 15^{th} order are in a good agreement with those calculated using the Rytov model. The low frequency continuous emission can be explained in terms of a breakdown of crystal momentum conservation for resonant Raman processes involving acoustic phonons. A frequency enlargement of continuous emission band and a downward shift of LO Ge phonons with increasing excitation energy (2.54–2.71 eV) are attributed to electron and phonon size-confinement in the small Ge QDs resonantly contributing to the scattering process.

Introduction

Recently, Raman spectroscopy was applied for characterisation of vibrational modes in self-assembled Ge QDs superlattices [1, 2]. It was found, that these structures behave very similar compared to the planar superlattices exhibiting folded acoustical phonons and LO and TO phonons in the Raman spectra [2, 3]. However, a weakening of selection rules in Ge QD superlattices due to the non-conserving momentum transfer is expected. Very recently, Cazayous *et al.* [4] has reported on observation of continuous emission even from the single layer with the large-size Ge QDs (dot base size of 170 nm) attributed to the breakdown of the wave vector conservation law due to the loss of translational invariance. Nevertheless, no detailed study of Ge QD superlattices, containing small-size QDs where three-dimensional confinement is especially important has been performed yet.

In this paper we report on size-selective Raman scattering in self-organised Ge/Si superlattices with small-size Ge QDs performed under resonant conditions.

1. Experimental

Samples were grown by molecular beam epitaxy of Ge and Si layers utilising Stranski-Krastanov growth mode on Si(001) substrates. The growth temperature of the silicon layers was 800 and 500°C before and after deposition of Ge layer, respectively. The Ge quantum dot layers were grown at 300°C. The set of samples under investigation consists of Ge and Si layers with nominal thickness of 1.4 nm and 30 nm, respectively. The samples were delta-doped with boron atoms ($N_b = 6 \times 10^{11}$ cm^{-2}) and ($N_b = 1.5 \times 10^{11}$ cm^{-2}) in the middle of Si layers for samples A and B, respectively. However, no noticeable influence of the doping on the Raman spectra was found.

A dot base size of 15 nm, a QD height of 1.5 nm and period of the structure of 38.5±1.0 nm were found using a cross-sectional high-resolution transmission electron microscopy (HRTEM).

The Raman scattering experiments were performed using lines of an Ar^+, Kr^+ and HeNe lasers in the range of 1.83–2.71 eV. The scattering geometries employed were z(xx)-z and z(xy)-z, where the labels x, y, z refer to [100], [010], [001] directions, respectively. Raman spectra were measured in Stokes and anti-Stokes region.

2. Low frequency region

According to the selection rules for Raman scattering in planar Ge/Si superlattices grown on Si(001) the LO phonons can be seen in z(yx)-z scattering geometry while the LA phonons can be observed in z(xx)-z geometry. As in the case of the planar Ge/Si superlattices, the experimental Raman spectra of Ge QD superlattices (sample A) measured in z(xx)-z geometry and shown in Fig. 1 reveal the a number of periodic oscillations (up to 15^{th} order) assigned to folded LA phonons in the Ge dot superlattice. The doublets of the folded phonons are not resolved because of the small splitting value (of about 1 cm^{-1}). These oscillations are superimposed with a broad continuous emission with a maximum at about 40 cm^{-1}.

The folded acoustical phonons observed can be well described using the Rytov model [5]. The calculated acoustic phonon dispersion curve is shown in the inset to Fig. 1. The horizontal line corresponds to the scattering wave vector used in experiment. It can be seen, that the agreement with the Rytov model is excellent and no adjusting of parameters is required. The period of structures deduced from the calculation is found to be 37.9 nm that corresponds very well to the average value evaluated from the HRTEM images.

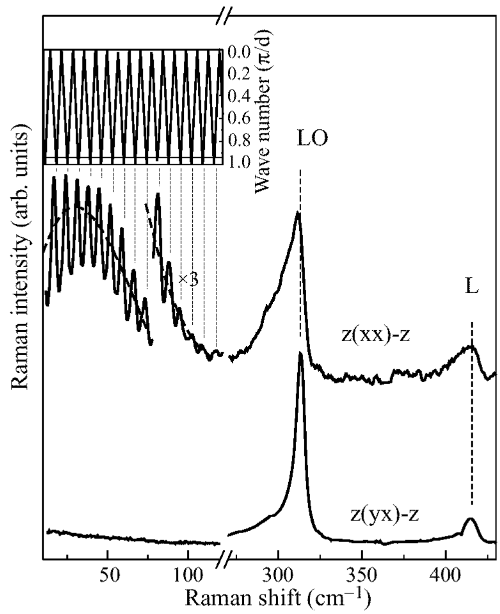

Fig. 1. Raman spectra of the sample A taken at $E_i = 2.41$ eV. The inset shows the calculated dispersion of LA phonons for the planar Ge/Si superlattice. Calculated Raman spectra are shown by dashed lines.

The Raman spectra were measured in Stokes and anti-Stokes regions with various excitation energies. The continuous emission and the scattering efficiency of the folded acoustic phonons is strongly enhanced for an excitation energy of 2.34 eV. Moreover, the observed emission band shifts toward higher free value of 25 cm^{-1} for excitation energy of 2.61 eV. The origin of the continuous emission observed can be understood using a model based on interaction between bulk-like acoustic phonons and confined electronic states. The electronic (or hole) confinement causes a breaking up of translational invariance and leads to Raman scattering by phonons originating from the whole acoustic dispersion branch [6, 7]. Following to Mlayah *et al.* [7] the scattering intensity under resonant conditions is calculated. Due to the particular shape and size of QDs (the dot base size is about one order of magnitude larger than the QD height) only the confinement along the growth axis can be considered. Coupling between Ge layers is neglected. The calculated Raman spectrum with an average value of Ge dot height of 1.2 nm is shown in Fig. 1 by a dashed line. The maximum and the spectral shape of the calculated emission band is consistent with those observed in experiment. The frequency enlargement of continuous emission observed at excitation energy of 2.61 eV corresponds to the Raman response from the QDs of the smaller size (0.8 nm). Indeed, the frequency position of continuous emission band is a measure (Fourier transform) of electronic confinement. Thus, strong electronic confinement in the small-size Ge QDs is very likely responsible for the frequency enlargement of continuous emission band resonantly enhanced at higher excitation energies.

3. Optical spectral region

In optical spectral range, the experimental Raman spectra of Ge QD superlattices (sample A) shown in Fig. 1 reveal the LO phonons in Ge QDs at 315 cm^{-1} in the z(yx)-z scattering geometry. The appearence of LO phonons in the "prohibited" z(xx)-z geometry manifests the lifting of the Raman selection rules for the planar superlattices due to QD formation. Frequency position of LO phonons corresponds to fully strained Ge QD layers where the biaxial compressive strain of Ge bonds is $\sim$ 4% [2]. Weaker feature at 417 cm^{-1} (labelled as L) was attributed to the longitudinal Ge-Si vibrational modes.

While the experimental facts considered above can be satisfactorily explained using various two-dimensional models, behaviour of optical phonons in Ge QD superlattices probed with different laser excitation lines can be understood only in terms of the zero-dimensional confinement. Figure 2 shows the Raman efficiencies and the frequency positions for the

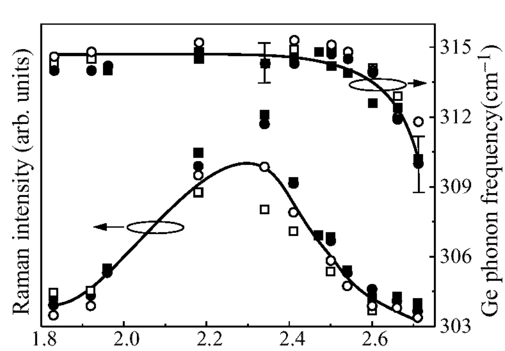

Fig. 2. Raman intensity and frequency positions of LO Ge phonons for the samples A (squares) and B (circles) measured in z(xy)-z (open) and z(xx)-z (solid) geometry.

LO Ge phonons displayed as a function of excitation energy. The resonance peak at about 2.34 eV that is very similar to the resonance observed in Ref. [1] is attributed to the E_1 exciton in Ge quantum dots. The position of the LO phonons localised in Ge QDs shifts towards lower frequency with the excitation energy (from 2.5 to 2.7 eV). This shift amounts to 4–5 cm^{-1} and indicates the presence of a QD size distribution in Ge dot superlattices. Raman scattering from smaller Ge QDs for which the E_1 exciton is at the higher energies, is size-selectively enhanced by the resonance of the exciting laser energy and the confined excitonic states. The size-confinement effect of optical phonons in Ge QDs which is stronger for the QDs with a small size, gives rise to a shift of optical phonons towards lower frequencies due to a negative dispersion of optical phonons in Ge.

The interdiffusion at Ge-Si interface in the structures under investigation was found to be very small [2] and cannot be responsible for this effect. A good agreement of calculated and experimental Ge phonon frequency positions as well as a small broadening of Ge optical phonons (~ 6 cm^{-1}) suggests that the fluctuation of strain for different QDs is small. Thus, in our samples, the atom intermixing and strain variation can be neglected.

In conclusion, we have carried out a detailed analysis of the Raman spectra of Ge QD superlattices. The folded LA phonons seen in the spectra can be very well described by Rytov theory usually applied for conventional superlattices. The weakening of selection rules due to non-conservation of crystal momentum gives rise to the low frequency continuous emission. The confined electron-hole transitions which lead to an increase of the E_1 exciton energy with decreasing Ge QD size allow size-selective Raman scattering under resonance conditions. Strong electronic confinement in small size Ge QDs is very likely responsible for frequency enlargement of continuous emission.

References

[1] S. H. Kwok, P. Y. Yu, C. H. Tung, Y. H. Zhang, M. F. Li, C. S. Peng and J. M. Zhou, *Phys. Rev. B* **59**, 4980 (1999).

[2] A. Milekhin, N. Stepina, A. Yakimov, A. Nikiforov, S. Schulze and D. R. T. Zahn, *Eur. Phys. J. B* **16**, 355 (2000).

[3] A. Milekhin, N. Stepina, A. Yakimov, A. Nikiforov, S. Schulze, T.Kampen and D. R. T. Zahn, *to published in Poceedings of ICPS-10, Osaka, Japan, sept. 2000*, (2001).

[4] M. Cazayous, J. R. Huntzinger, J. Groenen, A. Mlayah, S. Christiansen, H. P. Strunk, O. G. Schmidt and K. Eberl, *Phys. Rev. B* **62**, 7243 (2000).

[5] S. M. Rytov, *Akoust. Zh.* **2**, 71 (1956).

[6] T. Ruf, V. I. Belitsky, J. Spitzer, V. F. Sapega, M. Cardona and K. Ploog, *Phys. Rev. Lett.* **71**, 3035 (1993).

[7] A. Mlayah, A. Sayari, R. Grac, A. Zwick, R. Carles, M. A. Maaref and R. Planel, *Phys. Rev. B* **56**, 1486 (1997).

[8] F. Cerdeira, C. J. Buchenauer, F. H. Pollak and M. Cardona, *Phys. Rev. B* **5**, 580 (1972).