Raman Scattering by Strained and Relaxed Ge Quantum Dots

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We present the results of an investigation of fundamental vibrations in Ge/Si structures with strained and relaxed Ge quantum dots (QDs) performed using resonant Raman spectroscopy. Self-assembled strained Ge/Si QDs were grown at different substrate temperatures by molecular beam epitaxy on Si(001) substrates. The introduction of an ultra-thin SiO_2 layer before Ge deposition was used to form relaxed Ge QDs. Resonant size-selective Raman scattering allows to make unambiguous assignment of optical phonons localized in the relaxed Ge QDs. Influence of confinement effect, strain and atomic intermixing in the QDs on the vibrational spectrum of Ge QD structures can be studied using resonant Raman scattering.

In this paper we present the results of an investigation of fundamental vibrations in self-organised Ge/Si structures with a single layer of relaxed Ge quantum dots (QDs) performed using resonant Raman spectroscopy. For the Ge/Si system the introduction of an intermediate SiO_2 layer of nanometer thickness before Ge deposition is used to form Ge QD's with sizes less than 7nm [1]. This method has been used to fabricate relaxed Ge QDs.

The existence of Ge QDs is evident from the plan-view and cross-sectional high resolution transmission electron microscopy (HRTEM) experiments. Figures 1a and 1b present the plan-view HRTEM images of a sample with relaxed Ge QDs. A two mode distribution of QDs in size is clearly observed from the figure. The lateral sizes of the QDs determined from the images are in the ranges of 100-200 nm and 3-6 nm.

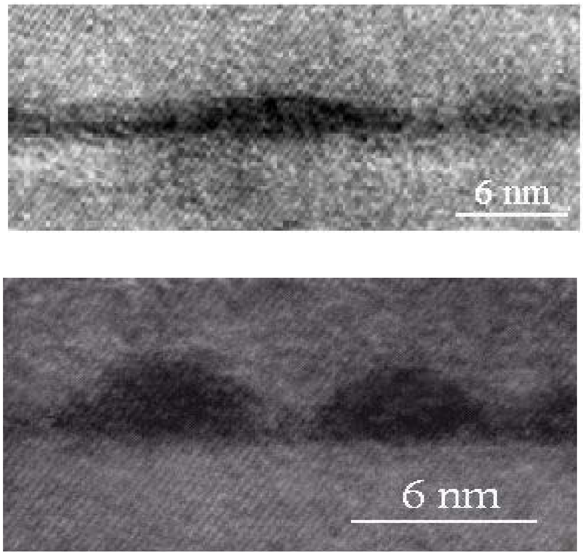

Fig 1. Cross-sectional HRTEM image of: a)- a layer of strained Ge/Si QDs b)- a layer of relaxed Ge QDs.

Samples wizth srtrained Ge QDs under investigation were grown by molecular beam epitaxy utilising the Stranski-Krastanov growth mode on Si(001) substrates at substrate temperatures varied in the range of 300 –650 ^oC. The period of structures consisting of Ge and Si layers with a nominal thickness of 0.7-1.4nm and 10-40nm, respectively, was n=7-10. At elevated temperatures in addition to hut clusters dome-like QDs occur. An average dot base size was found to be approximately 15 (50)nm with a QD height of 2 (10)nm for hut clusters (domes) .

A typical Raman spectrum of such a Ge dot structure is shown in Fig.2. Strong peaks in the spectrum are observed at 295, 406 and 520 cm^{-1} , and attributed to Ge-Ge, Ge-Si vibrational modes, and the Si phonon predominantly stemming from the (001)-oriented Si substrate, respectively. The broad feature near 480 cm^{-1} corresponds to local Si-Si vibrations and indicates Ge-Si intermixing at the interface. Phonon confinement causes a downward shift of the Ge optical phonon frequency with respect to its value in Ge bulk (300 cm^{-1} at 295K). The position of the Ge phonon localized in Ge QDs

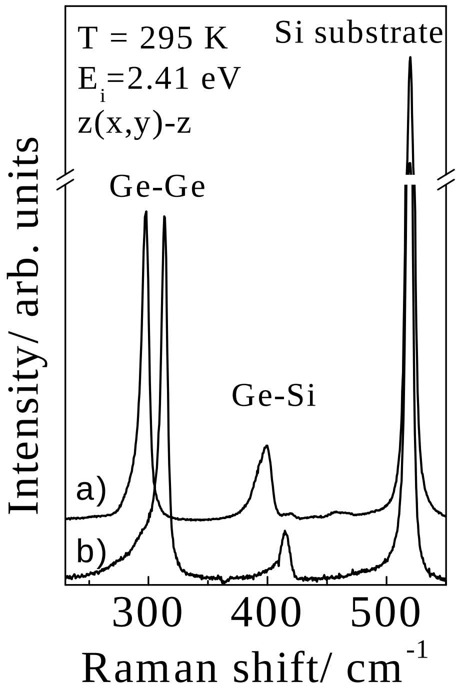

Fig.2. Raman spectrum of a Ge QD structure grown on a thin silicon oxide layer measured in $z\left(y,x\right)\bar{z}$ scattering geometry at an excitation energy E_i=2.47eV.

further shifts towards lower frequencies with increasing

excitation energy while the Raman line broadens as well.
The Raman efficiencies and the frequency positions of the Ge optical phonon as a function of excitation energy for the samples with different nominal Ge coverages are presented in Fig.3. The maximum of the Raman efficiency at 2.35 eV (1nm of nominal Ge coverage) is attributed to the E_0 resonance in Ge QDs. The shoulder located at about 2.2 eV is likely due to the E_1 resonance in relaxed Ge QDs. The energy of the E_0 transitions in Ge bulk amounts to 0.9 eV while in Ge QDs it reaches a value of about 2.5 eV due to electronic confinement. The frequency position of the Ge optical phonon under non-resonant conditions (e.g. at 1.83 eV) corresponds to the bulk value of relaxed Ge. It shifts downwards with increasing excitation energy (from 2.5 to 2.7 eV) indicating the presence of a QD size distribution. Since the QD structures under investigation have a two mode size distribution, it is most

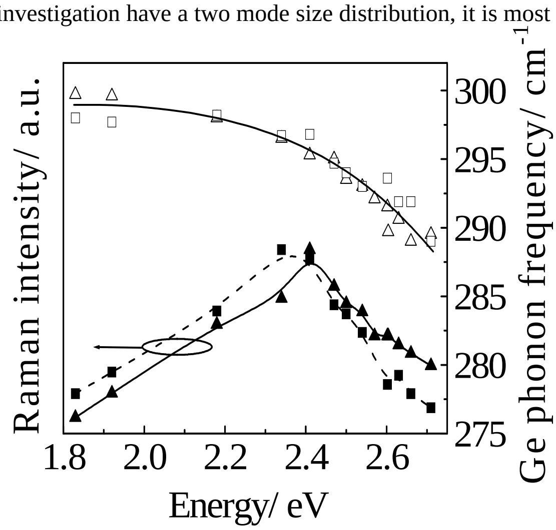

Fig. 3. Raman intensity and frequency positions of Ge phonons for a Ge QD structure with a nominal Ge coverage of 1nm (squares) and 2nm (triangles) grown on a thin silicon oxide layer measured in $z\left(x,y\right)\bar{z}$ geometry. Solid and dashed lines are guides to the eye

probable that the large Ge islands for which the confinement effect is negligible contribute effectively to the Raman scattering. With increasing excitation energy (above 2.3 eV) the smaller QDs are more involved in the scattering process. Raman scattering from smaller Ge QDs for which the E_0 resonance is at a higher energy, is size-selectively enhanced by the resonance of the exciting laser energy and the confined electronic 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 maximum of the Raman efficiency with a larger nominal thickness of the Ge QD layer (2 nm) shifts towards higher energy (2.42 eV) and indicates the formation of small-sized Ge QDs.

The built-in strain and atomic intermixing in the QDs structures grown at different substrate temperatures were studied also by means Raman spectroscopy. The frequency position of optical phonons observed in Raman spectra of the structures shifts towards lower frequency with increasing growth temperature that was attributed to higher intermixing of Ge and Si atoms and hence a strain relaxation in the QDs grown at elevated temperatures.

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Reference:

1. A. A. Shklyaev, M. Shibata, and M. Ichikawa, Phys. Rev. **B62**, 1340 (2000)