Inelastic Light Scattering by Acoustic Phonons in Quantum Dots and Quantum Films

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Abstract—The Raman scattering spectra were analyzed in order to find and explain similarities and differences in the interaction between electrons and acoustic phonons in quantum dots and quantum films. **DOI:** 10.3103/S1062873809070028

A new field of nanoscale science, which deals with the properties of phonons in nanostructures and control of phonon dispersion in nanostructures (phonon engineering) is referred to as nanophononics. Reducing the size of electronic devices below the acoustic phonon mean free path forms new possibilities for phonon propagation and interaction.

The role of interaction of acoustic phonons with electrons in optical and electrical properties of nanostructures is especially important in the following cases: (i) when microstructures decrease in size and phonons with a larger wave vector are involved in the electron-phonon interaction, due to which electron scattering by acoustic phonons becomes more pronounced as compared to the scattering of electrons by optical phonons; (ii) when scattering by optical phonons, being dominant in the 3D case, is reduced or suppressed due to phase and spatial confinements; and (iii) when the number of defects decreases with improvement of the nanostructure technology, as a result of which the electron-optical phonon interaction weakens, and the minimum linewidth of optical spectra is determined by the electron-acoustic phonon interaction.

We begin consideration with Raman scattering in quantum dots (QDs), which has been studied more comprehensively as compared to that in quantum films. For our study, we chose Ge QDs in a dielectric matrix, because such structures are used as carriers in single-electron nonvolatile memory devices [1]. We synthesized Ge nanocrystals using the disproportionation reaction 2GeO \rightarrow Ge +GeO₂ in a helium flow at temperatures ranging from 300 to 550°C [2]. In the resulting heterophase film, Ge crystals were dispersed therein an amorphous dielectric GeO₂ matrix. Varying the synthesis conditions, we could control the size of microcrystals.

There are two characteristic types of acoustic vibrations in QDs: spheroidal and torsional, with only their lowest energy modes being active in Raman scattering. They can be distinguished by polarization measurements, because spherical vibrations lead to polarized scattering, while torsional ones result in depolarized scattering geometry. Raman spectra of QDs were measured at room temperature in the backscattering geometry using a Ramanor U-1000 spectrometer, with the electric field vector of the exciting wave oriented parallel to the scattering plane, while the vector of the scattered wave was oriented either perpendicularly to this plane (V-H polarization) or parallel to it (V-V polarization).

Most QD specimens prepared so far by diverse techniques are changed by nonuniform size distribution. Since most of the QD properties are determined by their size and shape, it is important to develop techniques capable of controlling the QD average size and distribution in a given sample. These parameters can be estimated based on the microscopic theory of Raman scattering, from the widths and positions of Raman peaks. The larger the acoustic mismatch $(\rho_2 V_2 / \rho_1 V_1$, where ρ is the density of the material and V is the sound velocity) between dots and matrix, the better acoustic cavity the OD is, and the stronger it localizes phonons. The Ge-QD structures used in single-electron nonvolatile memory device applications are embedded in a dielectric SiO₂ matrix formed by oxidation of Si layers with incorporated Ge QDs. We can find from the Raman peak widths whether Ge QDs are surrounded by amorphous SiO_2 oxide or by Si. In Fig. 1, the polarized Raman spectra of Ge QDs embedded in Si [3] and in a dielectric GeO₂ matrix [2] are compared.

It can be seen that the Raman peaks of Ge QDs in Si are less pronounced against the scattered light background, because, first, Si stronger scatters light than an amorphous GeO_2 matrix and, second, the acoustic mismatch between Ge and Si is smaller than that between Ge and GeO_2 .

The electronic states in Raman scattering play the role of the intermediate link since the photon—phonon interaction occurs via the electron subsystem.



Fig. 1. Polarized Raman scattering spectra of Ge QDs with a size of ~ 13 nm (a) located on Si after oxidation and annealing [3] and (b) embedded in a GeO₂ matrix [2].

The interaction via the deformation potential (DP) is one of the most important mechanisms of coupling between electrons and acoustic vibrations. For a homogeneous semiconductor of cubic symmetry with a conduction band minimum at the Γ point, the interaction Hamiltonian has the form

$$H_{\rm int}^{\rm PD} = D {\rm div} \mathbf{u}(\mathbf{r}), \qquad (1)$$

where D is the deformation potential constant; the lattice shift divergence **u** determines the relative deformation-induced change in the crystal volume. It can be seen from Eq. (1) that in the case of spherical nanocrystals torsional modes do not contribute to this coupling because of their transverse character. Having analyzed our Raman spectra [4, 5], we concluded that torsional vibrations of Ge QDs are observed in the case of V-H polarization. Thus, it is reasonable to suggest another interaction mechanism. Indeed, it was reported in [6] that, in addition to the usual deformation potential coupling, electron-acoustic phonon interaction may arise through the perturbation of electrons by vibrating boundaries of nanostructures. This additional interaction, known as the ripple mechanism (RM) (applicable for all nanostructure geometries), is introduced by the equation

$$\mathcal{H}_{\text{int}}^{\text{RM}} = \mathbf{u}(\mathbf{r})\vec{\nabla}U(\mathbf{r}),\tag{2}$$

where $U(\mathbf{r})$ is the confinement potential. In contrast to the bulk mechanism (1), which allows only the interaction with longitudinal phonons in isotropic media, transverse acoustic phonons contribute to the interactions (2) as well. Thus, the Raman spectra observed on torsional vibrations of Ge QDs in Fig. 1 are most likely to be caused by RM.

Let us consider now the case of Raman scattering in quantum films grown on different substrates, in view of the recent theoretical investigation of the Raman scattering by longitudinal acoustic phonons in thin Si membranes and Si films on SiO₂ substrates (SOI structures) [7]. We have studied thin strained and unstrained epitaxial Ge films deposited on Si and GaAs substrates, respectively [8]. The films were grown in a highvacuum system allowing simultaneous electron diffraction structure analysis. The experimental Raman spectra in Fig. 2 were measured in the backscattering geometry using a Ramanor U-1000 spectrometer in the V-V polarization, i.e. with the only contribution of longitudinal acoustic phonons.

It was revealed in [7] that spatial modulations of acoustic, optical, and photoelastic properties affect the shape of Raman scattering spectra. In other words, the film appears to be simultaneously three different cavities: acoustical, optical, and photoelastic; they contribute jointly to the Raman scattering efficiency. The photoelasic model was used to calculate the Raman spectra. If we consider the acoustic modes localized in an unsupported film, under stress-free boundary conditions at the upper and lower film surfaces, the eigenmodes turn out to be quantized in the wave vector, according to the expression

$$q_n = \pi n/L, \tag{3}$$

where *L* is the film thickness and *n* is an integer mode index. Within the linear dispersion regime of acoustic modes, the eigenmodes are regularly spaced in frequency, according to the relation $\omega_n = q_n V_L$ and Eq. (3). It was shown in [7] that the peaks in the Raman spectra of unsupported Si films (membranes) are related to only the acoustic cavity, while the optical and photoelastic contributions affect only the relative mode amplitude.

When a film is not freestanding but attached to a substrate, the phonon wave function partially penetrates the boundaries because of the difference between their acoustic impedances $\rho_2 V_2 / \rho_1 V_1$, where ρ is the density of the material and V is the speed of sound. The final acoustic mismatch between the film and substrate appears to result in the spatial (bound-ary-induced) confinement of acoustic phonons. However, it was pointed in [7] that resonant acoustic modes are not confined within the film since they have a similar amplitude in the substrate due to the boundary conditions. Therefore, a continuum of acoustic modes from the substrate is available at any acoustic mis-



Fig. 2. Raman scattering spectra for longitudinal acoustic phonons: (a) strained epitaxial Ge films with thicknesses of (1) 19 and (2) 10 nm on a Si substrate; (b) unstrained Ge films with thicknesses of (1) 18 and (2) 12 nm on a GaAs substrate. In panel (a), the Brillouin LA peak of the Si substrate is indicated by an arrow. The Brillouin peaks of Ge and GaAs at 3 and 2 cm⁻¹, respectively, are invisible. The spectra contain the vibrational n = 2 and n = 3, except for the case (2) in panel (a) where only the mode n = 3 is present.

match, i.e., resonant phonon modes arise from the interaction between the continuum of phonon states from the substrate and the quasi-localized modes in the film. The substrate-film system is analogous to the substrate-defect layer system. The theory of impurity modes in lattice dynamics [9] is well understood via the Green's function formulation of host-defect interactions, especially in the case of a single substitutional impurity atom of mass m_I , connected to the nearest neighbors via the force constants f_{I} . Heavy substituents with smaller f_I give rise to band modes within the continuum of the host lattice and acquire generally a resonant character. It was revealed in the theoretical study [10] that the film/substrate phonon structure changes in the presence of an oxide layer on the film. In this case, the resonant modes shift upward on the energy scale, and their number increases due to



Fig. 3. Calculated low-frequency Raman spectra of the $10\text{Ge}_2\text{Si}_2$ film (~6 nm) on the Si substrate: (a) a freestanding film uncapped with oxide and (b) a film capped with oxide [10]. The Brillouin LA peak of the Si substrate is indicated by an arrow.

the involvment of another acoustic cavitry of the oxide layer (Fig. 3). The film in Fig. 3 is an ultrathin Ge_mSi_n superlattice; its low-frequency Raman spectrum is insensitive to the layered structure and corresponds to vibrations of a single layer with averaged parameters. A comparison of our experimental spectra (Fig. 2) with the calculated ones in Fig. 3 demonstrates that the Raman spectrum of the Ge/Si film is in better agreement with that of the oxide uncapped film, whereas the spectrum of the Ge/GaAs film is close to that of the capped film.

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