

# Investigation of the effects of a quantum dot crystal geometry on its Brillouin spectrum

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**Abstract.** We develop a theoretical model and carry out simulation of Brillouin spectrum of three-dimensional (3D) quantum dot (QD) arrays with a high order of 3D periodicity, i.e. quantum dot crystals (QDC). The phonon spectrum of Ge/Si QDC is found from the numerical solution of the elasticity equation for the whole structure. The developed approach is valid for any QD shape and regimentation and allows to include disorder in consideration.

## Introduction

Raman and Brillouin spectroscopies have proven to be a powerful tool for investigation of arrays of semiconductor quantum dots, nanoparticles, as well as of nano- and microcrystalline multilayers. It is capable of providing information on modification of vibration spectra of such structures as well as on carrier confinement [1],[2],[3]. Of special interest are phonon confinement effects in QDC, which is a very perspective thermoelectric material because it combines the profits of phonon scattering on QDs and increased mini-band conductivity [4].

When quantum dots form a *regimented* or partially regimented array [6], the interpretation of Brillouin spectra becomes more of a challenge due to possible appearance of additional phonon dispersion branches, e.g., standing waves inside or between quantum dots, etc. [5] along with the additional effect of strain, alloying, and interdiffusion. This presents a strong motivation for theoretical investigation of Brillouin and Raman spectra of quantum dot arrays.

In this paper we outline our model based on numerical solution of the elasticity equation for the *whole structure rather than for separate dots*, which allows for accurate interpretation of Raman spectra of QDC. We argue that it is essential to consider the vibration spectrum of the whole structure in order to obtain correct peak positions and separate the effect of strain or interdiffusion from phonon confinement.

## 1 Theoretical model

To simplify the numerical solution of the elasticity equation for the heterogeneous system of QDC we restricted our analysis to orthorhombic periodicity of QDC formed by QDs of various shape. The feature size of QDC (3 nm – 9 nm) was chosen to be much smaller than the phonon mean-free path and laser wavelength

( $\lambda = 514$  nm) yet it is large enough for application of the elastic continuum approximation (see applicability limits discussed in Ref. [5]). In the same time, it allows to discretize the elasticity equation on the square grid of monolayer-size that ensures the model shape of QD to be very close to the real one. This is especially important for pyramidal, dome, and “damaged” QDs. On the other hand, the expansion of the solution domain onto the several unit cells of QDC allows us to introduce a periodical quasi-disorder in the model.

The finite-difference elasticity equations were obtained using Euler-Lagrange equations from the discretized Lagrangian of the system, which ensured the Hermiticity of the corresponding matrix constructed on bonds with determined on them material parameters (see Ref. [5] for the details). To avoid the uncertainty in definition of material parameters we assumed that they change linearly on QD boundaries. The corresponding matrix is sparse; it contains only 35 nonzero elements per every  $3N_xN_yN_z$  row. Here  $N_i$  denotes a number of nodes in  $i$ -direction. Thus we used implicitly restarted Arnoldi algorithm [7] to find its eigenvalues. The scheme developed ensures the relative error less than 4% for as little nodes as  $N = 15$  per QDC period in every direction and less than 1% at  $N = 30$ , when QDs are rectangular prisms.

After phonon dispersion is found, we obtain Raman intensities using macroscopic theory for calculating the photoelasticity tensor. It describes the phonon – photon interaction in the following way. A periodic displacement of geometrical points of the matter  $\mathbf{U}(\mathbf{r}, \mathbf{q}, \Omega) = \mathbf{u}(\mathbf{r}, \mathbf{q}, \Omega) \exp(-i\Omega\mathbf{t})$  causes the periodic change of the local strain  $\sigma_{ij}$  which, in turn, locally modulates the dielectric susceptibility  $\epsilon_{ij} = \epsilon_{ij}^0 + \sum q_{ijkl}\sigma_{kl}$  of the matter. Here  $\Omega$  is the phonon frequency,  $\mathbf{q}$  is the phonon wave vector,  $\epsilon^0$  is an unperturbed susceptibility tensor, which is diagonal in the main coordinate system of cubic semiconductors,  $q_{ijkl}$  are components of photoelastic tensor. In semiconductors of cubical symmetry there are only 2 independent non-vanishing components of the photoelasticity tensor  $q_{1111}$  and  $q_{1122}$ . Thus  $\epsilon_{ij}$  perturbed by phonons has only its diagonal components

$$\epsilon_{xx} = \epsilon_{xx}^0 + q_{1111} \frac{\partial U_x}{\partial x} + q_{1122} \frac{\partial U_y}{\partial y} + q_{1122} \frac{\partial U_z}{\partial z}, \quad (1)$$

with similar expressions for  $\epsilon_{yy}$  and  $\epsilon_{zz}$  obtained by cyclic exchange of  $x$ ,  $y$ , and  $z$ .

Electromagnetic wave with frequency  $\omega$  and wave vector  $\mathbf{k}$  in optically isotropic medium can be characterized by complex amplitude  $\mathbf{D} = \epsilon\epsilon_0\mathbf{A} \exp(i\mathbf{k} \cdot \mathbf{r})$ . Here  $\epsilon_0$  is dielectric susceptibility of vacuum,  $\mathbf{A}$  is the light polarization vector, which is perpendicular to the direction of the wave propagation,  $|\mathbf{k}| = 2\pi\epsilon^{1/2}/\lambda$ , and  $\lambda$  is the light wavelength in vacuum.

We limit our consideration to one-phonon anti-Stokes processes. Substituting expression for  $\mathbf{D}$ , taking into account that  $|\mathbf{D}_i\rangle$  and  $|\mathbf{D}_f\rangle$  correspond to electromagnetic wave in QDC with dielectric susceptibility perturbed by phonons (Eq. (1)) and applying quasi-periodic boundary conditions, we find the probability of the scattering from initial to the final state with the phonon assistance that *in effective optical medium approximation* correlates with conventional Raman tensor  $\mathfrak{R}$  for

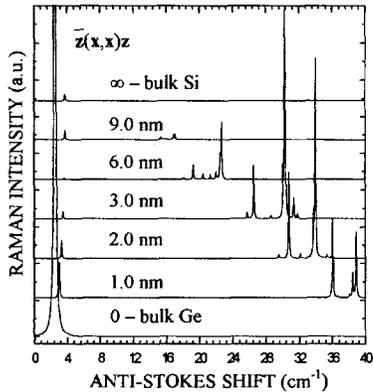


Figure 1: Modification of Raman spectrum of Ge/Si cubic QDC with  $L_x = L_y = L_z = 3.0$  nm and  $D_x = D_y = D_z$  with change of the distance between QDs.

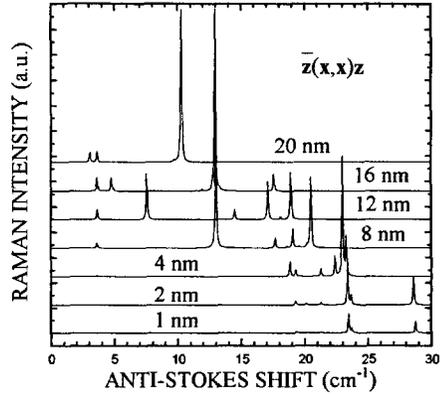


Figure 2: Modification of Raman spectrum of Ge/Si tetragonal QDC with  $L_x = L_y = 3.0$  nm,  $D_x = D_y = 9.0$  nm, and  $D_z = 2L_z$  with change of the QD height in  $[[001]]$  quasi-crystallographic direction, i.e. along the direction of light propagation.

the *whole* QDC structure  $P_{fi} \propto |\mathbf{A}^f \cdot \mathfrak{R} \cdot \mathbf{A}^i|^2$ , where

$$\mathfrak{R}_{xx} \propto \int_{EC} \exp(-i\mathbf{q} \cdot \mathbf{r}) \left[ (q_{1111} A_x^i A_x^f + q_{1122} (A_y^i A_y^f + A_z^i A_z^f)) \frac{\partial u_x}{\partial x} \right] d\mathbf{r}. \quad (2)$$

Expressions for  $\mathfrak{R}_{yy}$  and  $\mathfrak{R}_{zz}$  can be obtained by cyclic change of  $x$ ,  $y$ , and  $z$ . The intensity of Stokes peaks in experimental spectra can be found by scaling with the corresponding Boltzmann factors.

## 2 Results and discussion

It is usually assumed that in normal-incidence back-scattering configuration the Raman spectroscopy probes the zone-center phonons since transfer momentum is very small compared with the Brillouin zone size, e.g. the wavelength of light is several orders of magnitude larger than the lattice constant. The specific of Raman spectroscopy of regimented arrays of quantum dots is that the momentum  $|\mathbf{q}| \cong 2|\mathbf{k}_i|$  ( $\sim 0.085$  nm $^{-1}$  for the Ar laser) is comparable with the size of the quasi-Brillouin zone (QBZ), which is about  $0.35$  nm $^{-1}$  for QDC period along the direction of the light propagation  $D = 9.0$  nm. Thus, it is important to know the phonon states accurately when analyzing Brillouin spectra of QDC. Note, that each phonon branch changes the symmetry of the corresponding vibration in the regions of QBZ where it interacts with other branches.

A change in the inter-dot distance between cubical Ge in Si matrix (see Fig. 1) causes nonlinear redistribution of intensities. In two limiting cases of infinitely

small and infinitely large inter-dot distances QDC evolves to bulk Ge or Si, respectively. Correspondingly, only the lowest longitudinal mode is active. The upper longitudinal and mixed modes are most intensive when the symmetry breaking is highest. It is achieved when the dot size  $L$  is comparable with the inter-dot distance  $H$ . The shrinking of the QBZ with increasing  $D = L + H$  results in the red shift of these peaks since folding of the acoustic phonon dispersion branches is attained at lower energies. When the symmetry of regimentation of QDs is preserved the general structure of the Raman spectrum is the same.

Fig. 2 illustrates the effect of the dot shape, i.e. the symmetry breaking, on Brillouin spectra. The presented results are for the dots with constant base ( $L_x = L_y = 3.0$  nm and  $D_x = D_y = 9.0$  nm) and changing height of the quantum dot along  $[[001]]$  quasi-crystallographic direction. The inter-dot distance is fixed at  $H_z = L_z$ . One can see significant redistribution of the peaks intensity and strong shift of some peaks, which is a combined effect of the QBZ size decrease in  $[[001]]$  quasi-crystallographic direction and strong modification of phonon dispersion with change of the symmetry. The position of each peak can be traced to the shift of the folded acoustic and quasi-optical phonon branches as discussed in Ref. [5].

One should note here that the simulated Brillouin spectra have more complicated structure than typical doublets observed in Raman scattering from folded acoustic phonons in quantum well superlattices. The position of these peaks could not be deduced from Lamb-type models that use eigenmodes of free-standing nanocrystals.

In conclusion, our approach allows for an accurate analysis of experimental Raman spectra of 3D quantum dot arrays. It can be used to account for the effects of dot shape, regimentation, matrix materials and assist in separation of the spatial confinement effects from alloying and interdiffusion.

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