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**Phonon dispersion in silicon nanocrystals**

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**Abstract.** Due to their compatibility with CMOS technology, silicon nanocrystals (NC) are a key element of integrated nanodevices as single-electron transistors. The physical simulation of electron transport in these devices is a challenge and to correctly include the effects of electron-phonon coupling like relaxation into the ground state or collisional broadening of energy levels, the knowledge of phonon modes in silicon NCs is required. With this aim in view, we present a theoretical approach to the determination of phonon dispersion in NCs. It is based on the adiabatic bond charge model. While in small diameter NC the phonon modes are strongly discretized, the density of states becomes quasi continuous and bulk-like for diameter larger than approximately 2.5 nm. A special attention is paid to boundary conditions and two cases are considered: the surface atoms are either free to move or rigidly fixed. In the former case surface modes are formed at low frequency and in the latter case stationary modes appear near 11 THz. By projecting the NC modes on the basis of bulk modes, one can show the increasing correlation between these modes when increasing the dot size.

1. **Introduction**

To fully assess the potentialities of electronic devices based on semiconductor quantum dots (single-electron transistor, flash memory...), it is important to understand precisely the mechanisms of electron-phonon interaction in semiconductor nanocrystals and their consequences on the electron dynamics. According to several authors, the discrete nature of electron levels in quantum dots (QDs) is expected to give rise to the phenomenon called phonon bottleneck: an electron on a excited level cannot relax to the fundamental level by inelastic phonon scattering unless the energy between the levels is exactly equal to the phonon energy, which is likely to yield slow relaxation effects [1]. However, more detailed analyses have predicted that the phonon bottleneck should weaken or even disappear because of multiphonon effects [2] and collisional broadening [3]. Studies of the consequences of collisional broadening on the electron relaxation in semiconductor quantum dots have been reported for GaAs QDs using bulk phonon energies [3-6]. Nevertheless, as for electrons, the strong confinement in nanocrystals modifies the phonon states and considering the correct phonon dispersion is certainly important to accurately describe the electron-phonon coupling.

The present work, dedicated to the calculation of phonon dispersion in silicon nanocrystals, is a part of a study aiming at adding the electron-phonon relaxation in a silicon quantum dot-based device simulator [7] and investigating its influence on current-voltage characteristics [8].

In Section 2, we present the main features of the simulation method based on the Weber’s Adiabatic Bond Charge Model (ABCM) [9] and applied here to silicon nanocrystals. The ABCM has been shown to give the best results for diamond and zinc-blende type crystals [10] and has been successfully used to calculate the phonon dispersion for bulk materials [11, 12], for surfaces [13] and...
more recently for various silicon nanostructures including lattices of nanocrystals [15]. In Section 3, the results obtained for silicon nanocrystal are presented and discussed as a function of NC radius by comparison with bulk properties.

2. Model
The frequencies $\omega$ and the amplitudes of vibrations $\mathbf{u}$ in nanocrystals are given by $M \omega^2 \mathbf{u} = \mathbf{D} \mathbf{u}$ where $M$ is the mass of the silicon atom and $\mathbf{D}$ is the dynamical matrix [16, 18], which is constructed within the frame of the Adiabatic Bond Charge Model using Weber’s interaction parameters [9]. It should be reminded that in a finite nanocrystal the notion of wave vector is meaningless and we cannot make use of any translational invariance for the calculation.

For an $N_{Si}$-atom nanocrystal, the dimension of the dynamical matrix is $3N_{Si} \times 3N_{Si}$, and the determination of the eigenvalues and eigenvectors of the dynamical matrix may require a heavy computational task.

A special attention is paid to boundary conditions since in practical applications the silicon nanocrystal should be embedded in silicon dioxide. As the specific vibrational properties of the atoms situated at the oxide interface are unknown, two extreme cases are treated: the atoms are considered either fully free to move (free boundary conditions) or kept motionless (clamped boundary conditions). The study of the Si/SiO$_2$ interface characteristics is beyond the scope of this paper. These two types of boundary conditions have already been discussed for silicon nanowires [19] and thin films [20].

In our model, surface atoms are bonded only to atoms of the nanocrystal: they have less than four nearest neighbours and are therefore surrounded by less than four bond charges. This implies, when treating the surface atom case, to modify the interactions and the magnitude of the atom charge accordingly.

3. Results
Frequencies and amplitude of vibration have been calculated for silicon nanocrystals of various sizes. Due to the lack of experimental data on phonon dispersion in silicon nanocrystals, our calculations can only be compared with phonon dispersion in bulk silicon to verify at least the coherence of the model.

For each nanocrystal, we determine the phonon density of states (DOS) and compare it to the bulk DOS. When the nanocrystal radius increases the DOS should become closer and closer to that of bulk [15, 21]. To complete the analysis, a projection method is applied to compare the nanocrystal modes with bulk modes. Bulk silicon modes $\mathbf{u}_B(\mathbf{q}, \omega_B)$ form a basis set on which the nanocrystal modes $\mathbf{u}_{NC}(\omega_{NC})$ are projected: $\mathbf{u}_{NC}(\omega_{NC}) = \sum_{\mathbf{q}, \omega_B} C(\mathbf{q}, \omega_B, \omega_{NC}) \mathbf{u}_B(\mathbf{q}, \omega_B)$. If the nanocrystal modes are strongly correlated to the bulk modes, the projection coefficient $C(\mathbf{q}, \omega_B, \omega_{NC})$ must be high and localised around a given frequency. The projection coefficients are plotted as a surface plot superimposed on the bulk dispersion curve in the (1,0,0) direction.

In the case where the surface atoms are free, it can be seen from figure 1a that the global shape of the nanocrystal DOS tends to the bulk DOS calculated with the ABCM. However, for two frequency ranges (from 0 to 2 THz and from 13 to 14 THz) the dot modes do not correspond to bulk modes. On the figure 2a the projection of dot modes on bulk modes is represented. The coherence seems good, except for the same frequency ranges where the peaks are delocalized over the whole range of wave vectors: these modes are correlated to all the bulk modes at the same time. These are specific modes of the nanocrystal.

In the other case, when the surface atoms are fixed to their equilibrium positions, it must be noted from figure 1b that the global shape of the nanocrystal DOS is close to the bulk DOS, except in the 10-11 THz frequency range where an additional peak is observed. The projection of dot modes in figure 2b confirms the existence of this specific range of modes uncorrelated to bulk modes.
While studying GaP nanocrystals, Fu [21] demonstrated the existence of surface-like modes. Such modes have also been highlighted by Tütüncü and Srivastava for InP and GaAs surfaces [13, 14]. As shown in figure 3a for one of the specific modes observed under free boundary conditions ($\nu = 1.4$ THz), the vibration amplitude is maximal near the surface of the nanocrystal: surface-like
modes actually appear and are responsible for the additional low-frequency peak in the DOS (figure 1a).

In the case of clamped boundary conditions (figure 3b for $\nu = 10.5$ THz), it appears that the vibration amplitude exhibits a succession of nodes and antinodes as a function of radial position, which is very specific of the unexpected peak of modes in the DOS in the 10-11 THz range (figure 2b).

4. Conclusion
Using the Adiabatic Bond Charge Model, we have calculated the phonon dispersion of silicon nanocrystals. The boundary conditions are chosen to be either free or clamped.

In the free standing boundary conditions case, the results are coherent with the bulk case, except for a frequency range which has been demonstrated to correspond to surface vibrations. In the clamped boundary conditions case, the DOS is still close to the bulk one but another specific low-frequency range appears with mode amplitudes characterized by nodes and antinodes as a function of the radial distance.

Based on this phonon mode calculation, the study of electron-phonon coupling and the computation of scattering rates in nanocrystals are currently underway.

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