Zero-Phonon Line Broadening and Satellite Peaks in Nanowire Quantum Dots: The Role of Piezoelectric Coupling

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We investigate the influence of the one-dimensional character of the phonon modes in a catalytically grown GaAs nanowire on the absorption spectrum of an embedded quantum dot, focusing on the contribution from the piezoelectric coupling. In general, the reduced dimensionality of the phonons leads to spectral side peaks and a zero-phonon line broadening due to the energetically lowest (acoustic) phonon mode. While the deformation potential predominantly couples to radial modes, the piezoelectric interaction can also couple strongly to modes of axial character, leading to additional absorption features. The contribution of the piezoelectric coupling to the zero-phonon line is negligible.

INTRODUCTION

Recent epitaxial progress in the fabrication of catalytically grown nanowires as well as the possibility to include heterostructures has led to an increased interest in quantum dots (QDs) in nanowires. QDs are interesting for the study of quantum information and cryptography, requiring a good control of the dephasing mechanisms. Since the QDs are embedded in a strongly confined solid state matrix, it is important to take into account the influence of the geometry of the system on the phonon modes which offer an important decoherence channel.

Recently, the influence of the phonons interacting with an electronic two-level system via the deformation potential coupling was investigated in a GaAs wire. Here, we extend this analysis to include the interaction with phonons via the piezoelectric coupling which may be of particular interest for wurtzite nanowires. In comparison with bulk modes which lead to continuous phonon side bands attached to the zero-phonon line (ZPL), the interaction with wire phonons leads to a set of phonon side peaks. These reflect the multitude of phonon modes arising due to the spatial confinement in the lateral direction. In addition, while bulk phonons do not lead to a finite linewidth of the excitonic transition without resorting to higher order processes, the lowest (acoustic) wire phonon mode leads to a temperature dependent ZPL broadening. In this paper, we focus on the piezoelectric interaction which can lead to additional absorption features as compared to the case of the deformation potential coupling due to different selection rules in the coupling mechanisms.

THEORY

In the following, we give a brief outline of the calculation of the absorption spectrum and the phonon modes following Ref. as well as the QD parameters used. The different electron-phonon coupling mechanisms are considered in Section ...

Hamiltonian: We consider an interband two-level system (conduction level $|c\rangle$, valence level $|v\rangle$) interacting with the wire phonon modes via a diagonal coupling after a $\delta$-pulse excitation by a classical light field. After the initial excitation, the system is described by the Hamiltonian

$$H = H_0 + H_{el-ph};$$

the first term,

$$H_0 = (\varepsilon_v a_v^\dagger a_v + \varepsilon_c a_c^\dagger a_c) + \sum_{q\kappa} \hbar \omega_{q\kappa} b_{q\kappa}^\dagger b_{q\kappa},$$

describes the kinetics of the electronic two-level system and the phonons. Here, $a_i^\dagger(a_i)$ denotes the creation (annihilation) operator of an electron in state $|i\rangle$ with energy $\varepsilon_i$, while $b_{q\kappa}^\dagger(b_{q\kappa})$ denotes the creation (annihilation) operator of a phonon with quasi-momentum $q$ and dispersion $\omega_{q\kappa}$, where the modes are labeled by $\kappa$. The interaction with the phonons is given by

$$H_{el-ph} = -a_c^\dagger a_c \sum_{q\kappa} (g_{vv}^{q\kappa} - g_{cc}^{q\kappa}) (b_{q\kappa} + b_{-q\kappa}^\dagger).$$

Here, $g_{ii}^{q\kappa} = \int d^3 x \varphi_i^*(x)V_{q\kappa}(x)\varphi_i(x)$ is the electron-phonon coupling element with the interaction potential $V_{q\kappa}(x) = \sum_{q\kappa} V_{q\kappa}(x)e^{iq\kappa\cdot\mathbf{x}}(b_{q\kappa} + b_{-q\kappa}^\dagger)$, and the interaction is taken to be diagonal. No higher order phonon terms are considered here.

Absorption: The absorption is determined by the imaginary part of the susceptibility via the macroscopic polarization $P(t) \sim d_{vc}(t) + c.c.$, where $d_{vc}$ is the interband dipole moment. The microscopic polarization $p(t) = \langle a_c^\dagger a_v \rangle$ is calculated via the independent Boson
model (IBM) which can be solved analytically for an arbitrary number of phonon modes. The solution for $t > 0$ is given by

$$p(t) = p(0)e^{-i(\omega_{\text{disp}}+\Delta)t} \times$$

$$\times \exp \left[ - \int d\omega J(\omega) \left( 4n(\omega) \sin^2 \frac{\omega t}{2} + 1 - e^{-i \omega t} \right) \right],$$

with the transition energy $\omega_{\text{disp}} = \varepsilon_c - \varepsilon_v$, the Bose-Einstein distribution $n(\omega) = [\exp(h\omega/k_B T) - 1]^{-1}$ (temperature $T$), and the polaron shift and the phonon spectral density, respectively given by

$$\Delta = - \int d\omega J(\omega),$$

$$J(\omega) = \sum_{\eta} \frac{|q^\eta_{\text{p}} - q^\eta_{\text{c}}|^2}{\hbar^2} \delta(\omega - \omega_{\eta}).$$

**Phonon modes:** The phonon modes are calculated within an isotropic continuum model based on Ref. [16]. Assuming free-surface boundary conditions, the solutions are uniquely determined, taking the general quantized form

$$u_i(\mathbf{x}) = \frac{1}{\sqrt{N}} \sum_{\eta} u_{\eta i}(\mathbf{x}) e^{iqz(b_{\eta i} + b^\dagger_{\eta i})}$$

($i = r, \varphi, z$; $N$: number of unit cells). In the following discussions, we restrict to compressional modes ($u_\varphi = 0$). Torsional modes ($u_\varphi 
eq 0$) do not couple via the deformation potential coupling; for the piezoelectric coupling, this is taken to be an approximation. Furthermore, modes with a $\varphi$-dependence (so-called flexural modes) are disregarded, since we assume a QD model which is azimuthally symmetric, in which case the coupling to these modes vanishes.

We consider a zinc-blende lattice in the (111) growth direction. The resulting lattice corresponds, in a nearest-neighbor approximation, to the wurtzite lattice [17]. Calculations of strain in nanowires have shown that such a transformation is a good approximation [18]. The orientation is taken into account in the phonon modes via the velocities of sound which are now taken along the (111) direction. The dispersion relation as well as two exemplary modes are shown in Fig. 1.

**Quantum dot model and wire parameters:** For the QD model, we take a spherically symmetric charge distribution with harmonic potential $\varphi_i(\mathbf{x}) \propto e^{-(r^2+z^2)/2\alpha^2}$, with Gaussian radii $a_r = 5.8$ nm and $a_z = 3.19$ nm. For the wire, we use GaAs parameters: deformation potentials $D_c = -14.6$ eV, $D_v = -4.8$ eV, mass density $\rho_m = 5370$ kg/m$^3$, longitudinal and transverse sound velocities $v_1 = 5400$ m/s, $v_2 = 2800$ m/s, dielectric constant $\varepsilon_s = 12.9$, and wire radius $R = 25$ nm. The piezoelectric constants for wurtzite GaAs are not known. We thus calculate the constants from the zinc-blende value $e_{14}$, again using the transformation described above [17]. Using the value $e_{14} = -0.16$ Cm$^{-2}$ [10], we obtain an estimate for the wurtzite piezoelectric constants $e_{15} = e_{31} = 0.09$ Cm$^{-2}$ and $e_{33} = -0.18$ Cm$^{-2}$. We note that larger values for $e_{14}$ can be found in the literature [20] and that the transformation formula is an approximation, and thus the effect of the piezoelectric coupling could certainly be enhanced compared to the results shown here.

**ELECTRON-PHONON COUPLING MECHANISMS**

We consider two electron-phonon coupling mechanisms: the deformation potential and the piezoelectric coupling. In the following, we discuss and compare the coupling elements and the resulting phonon spectral densities. We restrict the discussions to the six energetically lowest phonon modes.

**Deformation potential:** The coupling of the electronic system to the phonons via the deformation potential is just given by the divergence of the displacement [15]:

$$V^i(\mathbf{x}) = D_i \nabla \cdot \mathbf{u}(\mathbf{x}),$$

where $D_i$ is the deformation potential. The calculated coupling constants are shown in Fig. 2(left), while the corresponding spectral density is shown in Fig. 2(right). Modes 3 and 6 are almost purely radial for $a \approx 0$ (cf. Fig. 1), thus they couple strongly via the deformation potential. However, due to the density of states, the spectral density of mode 3 is relatively weak. The axial modes 4 and 5 only couple weakly via the deformation potential for $a \approx 0$ due to the restriction to diagonal strain elements $S_{ij} = (\partial_i u_j + \partial_j u_i)/2$ in Eq. 8.

**Piezoelectric coupling:** For the piezoelectric coupling, the displacement strain causes an electrostatic potential
which is determined via
\[
\varepsilon_0 \varepsilon_s \Delta \phi(x) = \nabla \cdot \mathbf{P}^{\text{piezo}},
\]
where \( \mathbf{P}^{\text{piezo}} = \varepsilon_{k,ij} S_{ij} \) (piezoelectric constants \( \varepsilon_{k,ij} \)) 21, 22, 23. For the wurtzite structure, only three independent piezoelectric constants remain, leading to the polarization
\[
\mathbf{P}^{\text{piezo}}(x) = \left( \varepsilon_{15} \left( \partial_x u_x(x) + \partial_z u_z(x) \right) \right) \mathbf{e}_r
+ \left\{ \varepsilon_{31} \left( \partial_x u_x(x) + \frac{1}{r} u_r(x) \right) + \varepsilon_{33} \partial_z u_z(x) \right\} \mathbf{e}_z.
\]
Assuming that the potential vanishes for \( r \to \infty \) using \( \varepsilon_s = 1 \) outside the wire, we obtain an analytical solution for the electron-phonon potential \( V(x) = -\varepsilon_0 \Delta \phi(x) \).

In Figs. 3(left) and 3(right), the coupling elements and the corresponding spectral density is shown, respectively. Compared to the deformation potential coupling, modes 4 and 5 couple strongly here for \( q \approx 0 \) since the piezoelectric potential [Eq. (10)] takes into account the nondiagonal strain elements \( S_{ij} \) which favor axial modes for small \( q \) (cf. Fig. 1). This interplay between the deformation potential and the piezoelectric coupling can be seen nicely when comparing the spectral densities of the two coupling mechanisms, shown in Fig. 4(left).

**ABSORPTION SPECTRA**

We now calculate the absorption within the IBM for the one-dimensional phonon modes. In Fig. 4, the absorption spectrum for (left) a QD interacting with the wire modes is compared to (right) a QD exposed to bulk phonons for different temperatures. In both cases, a phenomenological (radiative) dephasing of \( T_2 = 1 \mu\text{eV} \) is used 24, and the system is excited resonantly on the polaron shifted transition energy \( E = \hbar (\omega_{\text{pol}} + \Delta) \). In Fig. 4(right), the different coupling mechanisms are compared in the absorption for the wire modes.

**Side peaks:** Compared to the case of bulk phonons, where the linear dispersion of the acoustic phonon mode leads to continuous side bands attached to the ZPL, the discrete wire modes lead to a series of phonon side peaks. The side bands are strongly temperature dependent, where higher temperatures lead to both increased dephasing as well as a broader spectrum due to multi-phonon scattering processes. Furthermore, at low temperatures, the absorption/emission asymmetry can be seen. The peaks in the absorption spectra

FIG. 2: (left) Coupling elements \( |g^{\text{coupl}}_{\phi \epsilon} - g^{\text{coupl}}_{\phi \epsilon}|^2(q) \) \( (L = 1 \text{ nm}) \) and (right) phonon spectral density \( J(\omega) \) for the deformation potential coupling for the energetically lowest phonon modes.

FIG. 3: (left) Coupling elements \( |g^{\text{coupl}}_{\phi \epsilon} - g^{\text{coupl}}_{\phi \epsilon}|^2(q) \) \( (L = 1 \text{ nm}) \) and (right) phonon spectral density \( J(\omega) \) for the piezoelectric coupling for the energetically lowest phonon modes.

FIG. 4: Comparison of (left) the phonon spectral densities \( J(\omega) \) and (right) the absorption spectra at \( T = 77 \) K for the deformation potential and piezoelectric coupling.

FIG. 5: Absorption spectrum of a QD interacting with (left) wire phonons via the deformation potential and the piezoelectric coupling and (right) bulk phonons via the deformation potential coupling for different temperatures.
correspond to the extrema in the dispersion, where the density of states has a singularity, whereas their strength is determined by the magnitude of the coupling at the extrema. In addition to the radial modes 3 and 6 which couple strongly via the deformation potential, the axial modes 4 and 5 can be seen in the absorption which interact via the piezoelectric coupling. The dependence of the scattering mechanism on the mode character can be seen more clearly in Fig. 4(right), where the two mechanisms are compared.

ZPL broadening: As was shown in Ref. [6], the phonon mode extending to vanishing energy leads to a temperature dependent broadening of the ZPL. This can be seen nicely in Fig. 5(left) for increasing temperature. For the piezoelectric coupling, no considerable contribution to the ZPL broadening is found due to the form of the piezoelectric potential for \( q \to 0 \).

CONCLUSION

We have studied the interaction of a quantum dot in a catalytically grown GaAs nanowire via the deformation potential and piezoelectric electron-phonon coupling. In general, the electron-phonon interaction leads to discrete side peaks, which are strongly temperature dependent, and a broadening of the zero-phonon line. Since the deformation potential couples strongly only to radial modes, the piezoelectric coupling can become important for modes which are mainly axial and thus can play an important role for the determination of the optical spectrum. The inclusion of the piezoelectric coupling does not lead to an appreciable contribution to the zero-phonon line broadening. It should be mentioned that larger values of the piezoelectric constants than used here have been reported in the literature, and thus the influence of the piezoelectric coupling could become more important. Furthermore, the piezoelectric coupling is strongly dependent on the quantum dot geometry, and thus the relative as well as the absolute strengths of the coupling constants must be considered for each case.

As an outlook, the size and shape dependence of the piezoelectric coupling to the one-dimensional wire phonon modes must be further investigated. In addition, the torsional modes, which are neglected in this investigation, should be considered to ascertain their interaction strength via the piezoelectric coupling.

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