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Quantum dissipation and broadening mechanisms due to electron-phonon interactions in self-formed InGaN quantum dots

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Quantum dissipation and broadening mechanisms in Si-doped InGaN quantum dots are studied via the photoluminescence technique. It is found that the dissipative thermal bath that embeds the quantum dots plays an important role in the photon emission processes. Observed spontaneous emission spectra are modeled with the multimode Brownian oscillator model achieving an excellent agreement between experiment and theory for a wide temperature range. The dimensionless Huang-Rhys factor characterizing the strength of electron-LO-phonon coupling and damping constant accounting for the LO-phonon-bath interaction strength are found to be ~ 0.2 and 200 cm^{-1} , respectively, for the InGaN QDs. © 2006 American Institute of Physics.

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Semiconductor quantum dots (QDs) have recently emerged as promising building blocks for quantum information processing apparatuses¹ and novel optoelectronic devices.² Thanks to the discrete nature of their structures, quantum dots are often referred to as artificial atoms. Unlike bona fide isolated atoms, however, semiconductor quantum dots cannot be separated from surrounding matrices that embed them. As a result, electron-phonon coupling plays an important role in determining optoelectronic properties of these nanostructures.³ In particular, it serves as a major dephasing mechanism for electronic coherence in quantum dots.³ Understanding electron-phonon interactions in semiconductor QD systems is therefore of both fundamental and technological interest.^{3–11}

Bastard and co-workers have proposed that polaronic states composed of strongly coupled excitons and LO phonons are the elementary quasiparticles that allow a coherent description of electronic excitation, relaxation, and luminescence processes in self-assembled semiconductor dots.^{4–6} Resonantly enhanced electron-LO phonon coupling may occur while the energy separation between discrete states of QDs is turned to match the characteristic energy of a LO phonon of the surrounding matrix via the band engineering method. Transitions within the excitonic polaron system in dots are argued to be facilitated by coupling of the exciton-LO-phonon polaron to a phonon thermostat, a scenario

which calls for application of the multimode Brownian oscillator (MBO) model. Fonoberov *et al.* employed a nonadiabatic approach to give a quantitative account of the photoluminescence (PL) spectrum of a II–VI semiconductor quantum-dots system incorporating the exciton-phonon interactions.⁷ They concluded that measured values of the Huang-Rhys parameter can only be explained within the nonadiabatic theory taking into account the phonon-assisted optical transitions. In addition to electron-LO phonon coupling, the role of acoustic phonons in various interband optical processes of semiconductor QDs has been discussed in length.^{12–14} It is currently recognized that the interactions of strongly confined electrons with acoustic phonons results in a broadened and non-Lorentzian zero-phonon line (ZPL) in luminescence spectra at low temperatures. It is also concluded that acoustic phonons should be taken into consideration for a quantitative interpretation of the luminescence spectra of semiconductor QD systems. However, a microscopic description remains elusive to quantitatively account for the contribution of acoustic phonons to the broadening of the ZPL of the QD fundamental transitions.¹³

Mukamel and co-workers introduced the MBO model to describe electronic relaxation of a two-level system attached to a few primary oscillators which are in turn linearly coupled with a bath of secondary phonons.¹⁵ Theoretical formulations have been developed to interpret ultrafast, time-resolved nonlinear spectroscopic measurements on solvation dynamics in liquids or solids.^{16–20} Rapid technological developments in making InGaN/GaN layered quantum structures²¹ provide an excellent opportunity to model

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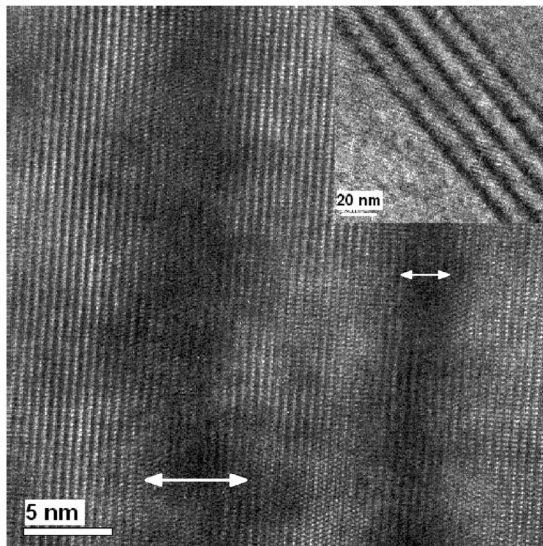


FIG. 1. Images of the InGaN/GaN QD sample obtained by cross-sectional transmission electron microscopy. The inset shows five periods of InGaN dot layers. These self-formed, indium-rich nanodots possess two representative average sizes: ~ 5 nm (long double arrow) and ~ 2 nm (short double arrow).

electron-phonon interactions in solid QD systems by applying the MBO model. It has been found that the self-formed InGaN QDs act as efficient luminescent centers in InGaN/GaN layered structures.²² Compared with InGaAs/GaAs QD material, InGaN/GaN QDs have stronger electron-phonon interactions due to the larger polarity and the existence of stronger piezoelectric field.

In this letter we report a study on electron-phonon interactions that combines experimental and theoretical efforts to arrive at a satisfactory understanding of electronic relaxation and fluorescence phenomena in Si-doped InGaN/GaN quantum dots. Photoluminescence (PL) spectra have been measured for the quantum dots, and results have been modeled with the MBO model.^{15,18} Excellent agreement obtained between theory and experiment demonstrates the presence of strong electron-phonon coupling in InGaN/GaN QDs. Crucial for understanding quantum dissipation and broadening mechanisms in nanostructures, the Huang-Rhys factor and the damping constant have been determined characterizing the electron-phonon coupling in those QDs.

Adopted for the study is a Si-doped InGaN/GaN QDs grown by metalorganic vapor phase epitaxy on a sapphire substrate. The structure consists of five periods of 12 nm GaN barriers and InGaN QD layers. The barrier layers are Si doped with a density of $\sim 10^{18}$ cm⁻³. The variable-temperature PL measurements of the samples were performed in a high-resolution PL setup described elsewhere.²³ The standard lock-in amplification technique was employed in the PL measurements.

Images of the InGaN/GaN QD sample obtained via cross-sectional transmission electron microscopy (TEM) are shown in Fig. 1. From the inset, five layers of InGaN islands can be seen to form. The high-resolution TEM images demonstrate that these InGaN QDs possess a bimodal size distribution. As indicated by double arrows in Fig. 1, two representative average sizes of the bimodal distribution are ~ 2 to ~ 5 nm. This leads to the observation of two well-separated zero-phonon emission lines as will be elaborated on later.

Based on the seminal work of Lax,²⁴ Kubo,²⁵ and Caldeira and Leggett,²⁶ the MBO model provides a convenient

theoretical framework to describe the coupling of nuclear coordinates to optical excitations and electronic relaxation channels in solids. In the MBO model, an electronic two-level system interacts linearly with a few primary nuclear coordinates that are in turn coupled linearly to a bath of secondary harmonic oscillator with a given spectral density. Three key parameters of the MBO model are the frequency of primary oscillator, the damping coefficient γ characterizing the coupling strength between the primary oscillator and the bath oscillators, and the Huang-Rhys factor S reflecting the electron-LO-phonon coupling strength.²⁷ It is known that the characteristic energy of the LO phonon (A_1 -LO mode) in GaN is about 92 meV (~ 740 cm⁻¹).²³ We thus adopted this value as the frequency of the primary oscillator which controls the energy separation between the ZPL line and one-phonon peak (and spacings between adjacent phonon sidebands). The damping constant γ , which accounts for the coupling strength of the primary oscillator and the bath modes, controls the broadening of the ZPL and its phonon sidebands. An often-adopted approach to treat inhomogeneous broadening in optical transitions of an ensemble of two-level systems such as those in InGaN/GaN QDs is to assume a Gaussian-type density-of-states distribution with a width W ,²⁸

$$f(\omega_{eg}) = \exp[-2W^{-2}(\omega_{eg} - \omega_{eg}^0)^2]. \quad (1)$$

The PL line shape can be calculated from

$$I_{\text{PL}}(\omega) = \frac{1}{\pi} \int_0^\infty f(\omega_{eg}) d\omega_{eg} \operatorname{Re} \int_0^\infty \exp[i(\omega - \omega_{eg} + \lambda)t - g^*(t)] dt, \quad (2)$$

where $\lambda = S\hbar\omega_{\text{LO}}$ and $g^*(t)$ is the complex conjugate of the line broadening function $g(t)$,

$$g(t) = -\frac{1}{2\pi} \int_{-\infty}^\infty d\omega \frac{C''(\omega)}{\omega^2} [1 + \coth(\beta\hbar\omega/2)] \times (e^{-i\omega t} + i\omega t - 1), \quad (3)$$

where $\beta = 1/k_B T$, and k_B is the Boltzmann constant. The spectral response function for a Markovian bath coupled linearly to the primary oscillator of frequency ω_{LO} with a coupling strength γ has the form¹⁸

$$C''(\omega) = \frac{2\lambda\omega_{\text{LO}}^2\omega\gamma}{\omega^2\gamma^2 + (\omega_{\text{LO}}^2 - \omega^2)^2}. \quad (4)$$

Here a frequency-independent damping coefficient was taken implying Ohmic-type dissipation of the phonon thermostat. In general, coupling of the primary oscillator to the bath modes can be nonlinear. For weak coupling, linear electron-phonon interactions are usually taken.¹⁸⁻²⁰ For the InGaN/GaN QDs system studied in the present work, it is obviously a weak coupling case because of the Huang-Rhys factor $S < 1$.

Figure 2 displays two representative PL spectra (open circles) of the sample measured at 5 K (upper panel) and 100 K (lower panel). The peaks denoted by ZPL1 and ZPL2 are assigned to the zero-phonon lines associated with the ground states of InGaN QDs with a bimodal distribution, which is supported by time-resolved PL measurements. LO phonon sidebands are on the lower energy side of the corresponding zero-phonon lines. The energy separations between

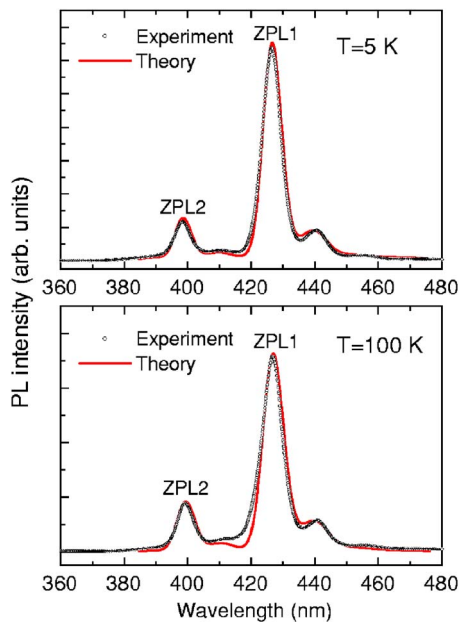


FIG. 2. (Color online) Representative PL spectra (open circles) of the sample measured at 5 K and 100 K. The solid lines are the theoretical curves. The parameters used in the calculations are $\omega_{LO}=740\text{ cm}^{-1}$, $\gamma=200\text{ cm}^{-1}$, and $S=0.20$. Adopted W -parameter values are directly determined from the experimental spectra: 165 cm^{-1} at 5 K, 20 K, and 40 K; 172 cm^{-1} at 60 K; 174 cm^{-1} at 80 K; and 180 cm^{-1} at 100 K and 120 K.

the two zero-phonon lines and their corresponding LO phonon sidebands are $\sim 90\text{ meV}$ matching the characteristic energy of the A_1 -LO mode of GaN.²³ Calculation of the zero-phonon lines and their corresponding LO phonon sidebands is facilitated by employing the underdamped MBO model with a Markovian bath.^{15,18,20} The parameters used in the calculations are $\omega_{LO}=740\text{ cm}^{-1}$, $\gamma=200\text{ cm}^{-1}$, and $S=0.20$. Calculated PL spectra (solid lines) of InGaN/GaN QDs at 5 K (upper panel) and 100 K (lower panel) are found to be in satisfactory agreement with measured ones.

As mentioned earlier, the Huang-Rhys factor is a key parameter characterizing electron-LO-phonon coupling strength, which has been proven to govern the relative intensities of the ZPL line and its successive sidebands. Accurate determination of the Huang-Rhys factor from the experimental spectra is thus crucial to understanding electron-phonon coupling system. We adopted a number of values for the Huang-Rhys factor of the InGaN/GaN QDs to calculate the PL spectra while the values of other parameters are kept unchanged. The measured and calculated integrated intensity ratios of the first-order LO phonon sideband and the zero-phonon line (ZPL1) are depicted in Fig. 3 as a function of temperature. It is found that when $S=0.2$ an excellent agreement between theory and experiment is achieved in the interested temperature range.

In summary, combining experimental and theoretical efforts, luminescence spectra of the interacting electron-phonon system in the Si-doped InGaN/GaN QDs have been studied revealing that bath modes from the surrounding matrices play a strong role in the broadening of the PL spectral lines in the QD system. Our results show that the multimode Brownian oscillator model with Ohmic dissipation can be used to model experimental PL spectra of QD systems with strong electron-phonon coupling.

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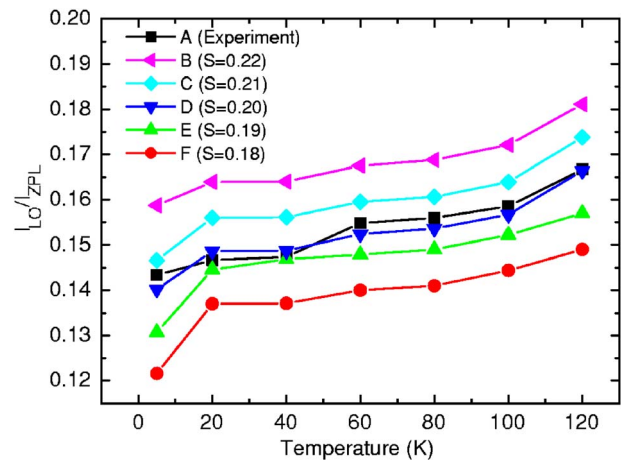


FIG. 3. (Color online) The measured and calculated intensity ratio of the first-order LO phonon sideband and the zero-phonon line (ZPL1) vs temperature. Direct comparison of experiment with theory makes one to determine the Huang-Rhys factor $S=0.20$.

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