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Phonon Interaction in InGaAs/GaAs Quantum Dots

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ABSTRACT

The carrier-phonon interaction in self-organized In(Ga)As/GaAs quantum dots is investigated under resonant excitation of the ground-state transition. Different phonon-coupled processes are observed. The distinction between Raman scattering and hot-luminescence process has been resolved by time-dependent photoluminescence measurements. The quantum dot LO (33.8 meV) as well as an interface (36.5 meV) phonon mode is observed by resonant Raman scattering. For the QD LO phonon mode, a very short radiative lifetime of 10 ps was found.

INTRODUCTION

In recent years, carrier-phonon interactions in self-organized semiconductor quantum dots (QDs) [1] have attracted considerable attention. They are important to understand the recombination and dephasing dynamics of confined excitons. Quantum dots may lead to different phonon modes because of interface boundaries or impurity atoms in the crystal. As a result of the local character of those phonon modes, the spatial overlap between electronic states and phonons is fairly large [2]. It has been shown experimentally and theoretically that single carriers [3, 4] and excitons [5-8] in QDs are strongly coupled to longitudinal optical (LO) phonons leading to the formation of QD excitonic polarons [7]. The observation of suppressed carrier relaxation has led to the prediction of a phonon bottleneck effect [2]. Additionally, exciton-phonon interactions have an influence on Raman scattering intensities and the intensities of the LO phonon sidebands of the QD ground state luminescence.

Here, we present a combination of resonant Raman spectroscopy, and time-resolved as well as temperature-dependent photoluminescence measurements. Resonant Raman spectroscopy was performed in order to enhance the Raman cross-section. The resonant PL technique enables us to retrieve the energies of the localized phonon modes. However, PL measurements are often inappropriate to distinguish between different phonon-coupled processes like Raman scattering and hot-luminescence process. Raman scattering is expected to have a much shorter lifetime than any luminescence process. Therefore, time-resolved PL measurements were performed to enable us to differentiate between coherent and incoherent processes.

EXPERIMENTAL DETAILS

In order to investigate the influence of the QD phononic system on the electronic properties of the QD structures, resonant photoluminescence (PL) experiments were performed at low temperatures and at elevated temperatures up to room temperature. Resonant PL was excited by a continuously tunable Ti:Sa pumped optical parametric oscillator (OPO) and detected by an InGaAs photodetector. Time-resolved photoluminescence (TRPL) was carried out for the purpose of achieving lifetime constants of phonon-assisted processes. TRPL was detected by a multi-channel plate (MCP) with a time resolution better than 30 ps. The temporal width of the excitation laser pulses was 3 ps at a repetition rate of 80 MHz. Photoluminescence as well as time-resolved and temperature-dependent experiments were performed on two different In(Ga)As/GaAs quantum dot structures. Both samples were grown on GaAs substrates by metal-organic chemical vapor phase deposition (MOCVD) and consist of a single In(Ga)As QD layer.

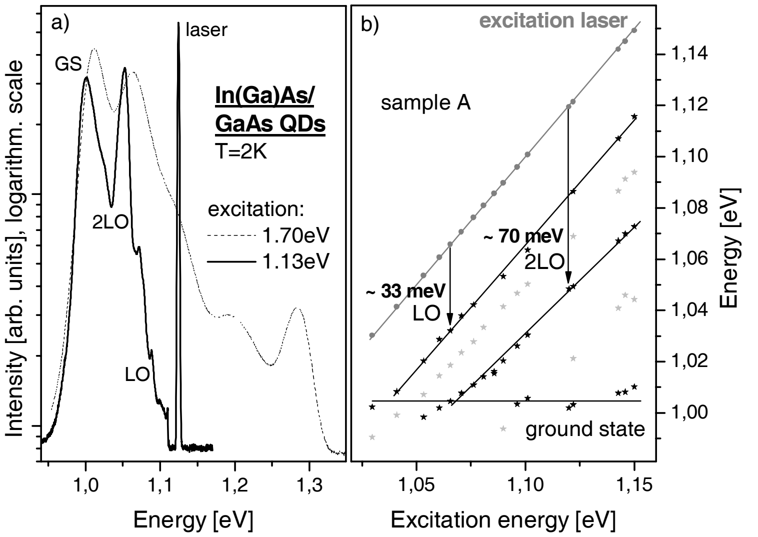

Figure 1: (a) PL spectra of In(Ga)As/GaAs QDs (sample A) for non-resonant and resonant excitation at T=2 K. (b) Detection energy versus excitation energy.

RESULTS AND DISCUSSIONS

The photoluminescence spectra of sample A for non-resonant and resonant excitation at 2 K are depicted in Fig. 1 (a). The gray spectrum was excited with 1.70 eV (non-resonant to the QD system). The ground state luminescence is centered at 1.01 eV. In addition, several other structures originating from the excited states can be observed (centered at 1.062 eV, 1.125 eV, 1.196eV, and 1.283eV). The black spectrum in Fig. 1 (a) represents a typical photo-luminescence spectrum under resonant excitation of the In(Ga)As quantum dot system with 1.13 eV. The narrow line at the high energy side of this spectrum is the laser. The ground state luminescence is centered at 1.005 eV and its FWHM is smaller in comparison to non-resonant excitation due to fluorescence line narrowing. Also, a structure at 1.065 eV can be observed. The origin of this luminescence was initially believed to be the first excited state. But this structure shifts to lower energies when the excitation laser is tuned to higher wavelengths. Since this structure shifts with the excitation energy at a constant distance of approximately 70 meV, it could be explained with a hot-luminescence process occurring at the 2LO energy. In addition to that, several other structures are shifting with the excitation energy, as depicted in Fig. 1 (b). Although the origin of two of those luminescence structures are questionable, one structure could be a hot-luminescence process occurring at the LO energy (circa 33 meV). Time-resolved photoluminescence measurements on the 2LO and LO structures revealed lifetimes comparable to the ground state transition. The intensity of those hot-luminescence structures does not remain constant. It increases with higher excitation wavelengths upon reaching the ground state luminescence. After that point, the hot luminescence strongly decreases until it disappears completely.

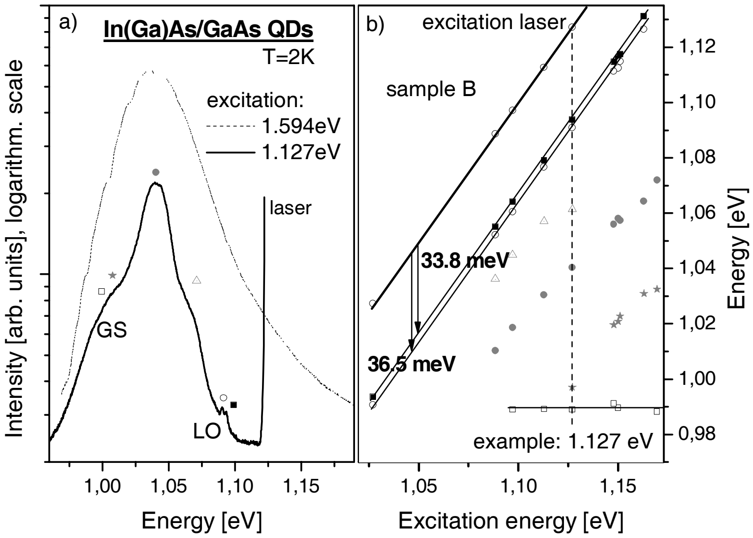

Figure 2: (a) PL spectra of In(Ga)As/GaAs QDs (sample B) for non-resonant and resonant excitation at T=2 K. (b) Detection energy versus excitation energy. The dashed line represents exemplarily the resonantly excited PL spectrum as shown in a).

Photoluminescence spectra of sample B for non-resonant and resonant excitation at 2 K are shown in Fig. 2 (a). The gray spectrum was excited with 1.594 eV (non-resonant to the QD system) and shows solely a very broad (FWHM=62 meV) ground state luminescence. The black spectrum was excited with 1.127 eV (resonant to the QD system). The ground state luminescence can be observed as a shoulder at an energy of 0.99 eV. In accordance to sample A, several other structures are shifting with the excitation energy as presented in Fig. 2 (b). Analog to sample A, a hot-luminescence process can be observed. A good example can be seen in Fig. 2 (a): The structure centered at 1.05 eV shifts with the excitation energy and has a considerable higher luminescence intensity than the ground state. Besides the hot luminescence, this sample shows unlike sample A a Raman scattering. A very sharp double structure shifts with the excitation energy at constant distances of 33.8 meV and 36.5 meV. Those energies fit to the In(Ga)As/GaAs quantum dot LO phonon mode (circa 33 meV) and the interface mode (circa 36 meV), respectively [2].

In order to distinguish between a hot-luminescence process and Raman scattering, we performed time-resolved photoluminescence measurements on our samples. The Raman scattering was expected to have a much shorter lifetime than the hot-luminescence process. TRPL measurements on sample A revealed comparable lifetimes of the ground-state luminescence and the hot-luminescence process. A fit assuming exponential rise and decay processes yields time constants of 520 ps (rise) and 750 ps (decay) for the hot-luminescence structures. In the case of sample B, the hot-luminescence structures (the gray dotted curve in Fig. 3) shows a slow hot-luminescence process solely. The analysis of this measurement pointed at lifetimes comparable to the ground state transition (rise and decay times of 270 ps and 650 ps, respectively), too.

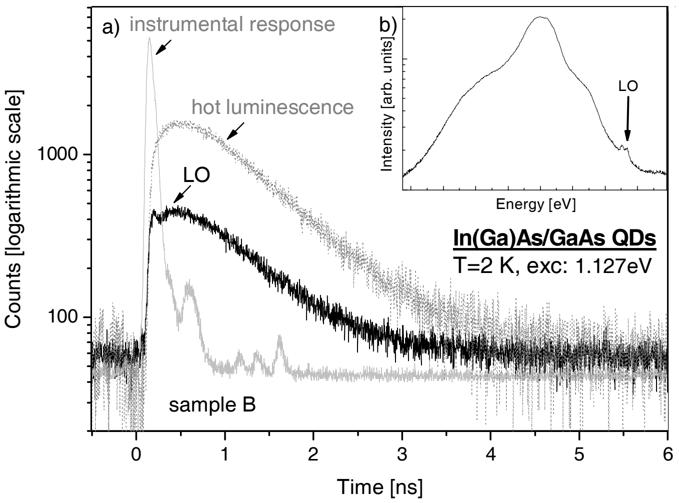

Figure 3: Time-resolved PL of the LO-phonon in In(Ga)As/GaAs QDs (sample B). The gray spectrum shows the instrumental response to the laser system. (b) PL spectrum for resonant excitation at T=2 K where the TRPL was measured. The arrow indicates the detection energy.

But the transients of the Raman scattering of the QD LO phonon mode show a different behavior (black curve in Fig. 3). The dynamical response to resonant excitation at T=2 K comprised two different components with diverse lifetimes for the QD LO phonon mode. We observed an incoherent portion as a result of the luminescence background that showed rise and decay constants in the magnitude of this sample's ground state luminescence. In addition to that, a much faster coherent portion as a consequence of the Raman scattering is visible. A fit assuming a bi-exponential decay process yields a decay time of 10 ps for the coherent portion.

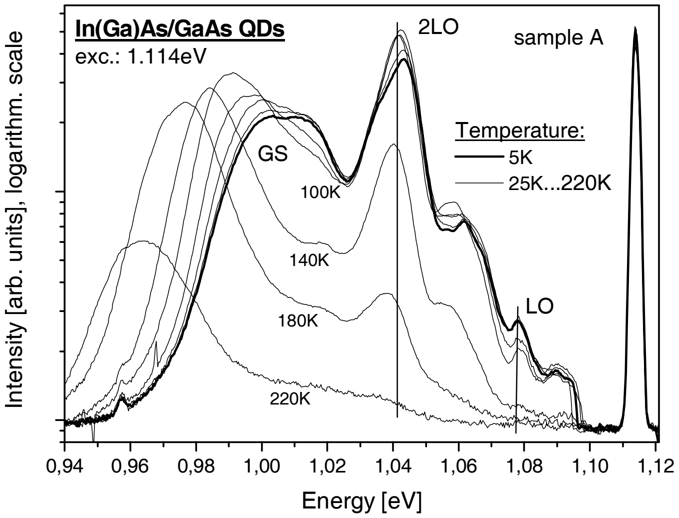

Figure 4: PL spectra of In(Ga)As/GaAs QDs (sample A) for resonant excitation at different temperatures (T=5 K...220 K).

Supplementary, temperature-dependent measurements have been performed. Photoluminescence spectra of sample A at temperatures between 5 K and 220 K are depicted in Fig. 4. All of those spectra were measured under resonant excitation for the identical excitation wavelength of 1113 nm (1.114 eV). It can be observed that the ground state luminescence increases a little until approximately 100 K. For higher temperatures the ground state intensity decreases. Because of the temperature increase, the ground state shows a red shift. For phonon-assisted processes it is expected that the luminescence intensity rises for increasing temperatures. Our measurements showed a different behavior. For temperatures until 100 K the intensity of the 2LO (at 1.043 eV) and the LO (at 1.078 eV) structures slightly increases (especially the 2LO structure). But for higher temperatures, the intensity goes down very fast. This behavior can be explained with the red shift of the ground-state luminescence. Since the luminescence of the ground state shifts to lower energies with the temperature, the hot-luminescence cross section goes down. The temperature shift of the ground state is very strong for temperatures above 100 K so that the intensity of the hot-luminescence structures decreases rapidly for higher temperatures.

CONCLUSIONS

In summary, we have reported on two different phonon-assisted charge-transfer processes occuring for self-organized In(Ga)As/GaAs quantum dots. We observed a hot-luminscence process and Raman scattering under resonant excitation of the QD ground state exciton. For one sample (B), we presented experimental evidence for the QD LO phonon mode through resonant Raman scattering with a very short radiative lifetime of 10 ps. The other sample (A) shows only hot-luminescence processes, since TRPL measurements revealed no fast Raman scattered portion.

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