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Temperature dependent optical properties of single, hierarchically self-assembled GaAs/AlGaAs quantum dots

M. Benyoucef · A. Rastelli · O. G. Schmidt · S. M. Ulrich · P. Michler

Abstract We report on the experimental observation of bright photoluminescence emission at room temperature from single unstrained GaAs quantum dots (QDs). The linewidth of a single-QD ground-state emission (≈ 8.5 meV) is comparable to the ensemble inhomogeneous broadening (≈ 12.4 meV). At low temperature ($T \leq 40$ K) photon correlation measurements under continuous wave excitation show nearly perfect single-photon emission from a single GaAs QD and reveal the single photon nature of the emitted light up to 77 K. The QD emission energies, homogeneous linewidths and the thermally activated behavior as a function of temperature are discussed.

Keywords GaAs quantum dots · Hierarchical self-assembly · Single dot spectroscopy · Room temperature luminescence · Photon correlation

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During the last decade, much attention has been paid to the fabrication of semiconductor quantum dots (QDs) and to their optical properties. QDs have large oscillator strengths, narrow spectral linewidths, long-term stability, and can be easily integrated inside device structures (e.g., pillar microcavities) [1–4]. Single photon generation using single self-assembled QDs

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obtained by Stranski–Krastanow (SK) growth mode has been demonstrated at low temperature [5, 6]. Single photons are useful for applications in quantum cryptography and quantum computation. Understanding the temperature dependence of QD emission is essential for making efficient devices, in particular at room temperature, where most devices operate.

Recently, a novel growth technique was used to fabricate self-assembled GaAs/AlGaAs QDs [7]. These QDs offer several advantages compared to SK grown QDs: The grown material is ideally unstrained with sharp interfaces and emits light in the visible range. Because of the limited intermixing between QD and barrier material, the determination of the QD structural properties is affected by much less uncertainties compared to SK QDs. This renders the calculation of the QD optical and electronic properties easier [7–10] and makes these QDs an ideal playground to understand basic QD properties [11]. Moreover, the size and shape of the QDs can be substantially varied by tuning the growth parameters [7, 8, 12, 13].

The variation of the emission energy, the full width at half maximum (FWHM), and the quenching of the photoluminescence (PL) intensity of SK-grown QDs with increasing temperature have been extensively studied [see e.g., Refs. 14–16]. Peter et al. [17] have studied the asymmetric phonon sidebands on both the exciton (X) and biexciton (XX) emission lines in single GaAs monolayer fluctuation QDs. They showed that these sidebands are due to a nonperturbative coupling to the acoustic phonons. Because of the small lateral confinement in such QDs it is not possible to study their emission at temperatures higher than about 35 K. The origin of QD emission broadening and quenching at high temperature is still subject of debate.

Moreover, only few studies of high temperature photon emission statistics from SK-grown QDs [18–20] have been performed. In this work we study the temperature dependence of the emission of a single, hierarchically self-assembled GaAs/AlGaAs QD. At room temperature we observe bright PL emission with FWHM of ≈ 8.5 meV. Remarkably, the width of a single-QD emission is comparable to the ensemble inhomogeneous broadening of ≈ 12.4 meV. By combining a micro-photoluminescence (μ-PL) and a Hanbury-Brown and Twiss (HBT) correlation setup, we demonstrate a single photon emitter from the excitonic transition up to 77 K. Also, the QD-PL emission energies, homogeneous linewidths and the thermally activated behavior are discussed.

The investigated samples were grown on GaAs (001) substrate by a solid-source molecular beam epitaxy system equipped with an $AsBr_3$ etching unit. The GaAs QDs are obtained by overgrowing a GaAs surface containing self-assembled nanoholes [12] with 7 nm $Al_{0.45}Ga_{0.55}As$, 2 nm GaAs, 100 nm $Al_{0.35}Ga_{0.65}As$, 20 nm $Al_{0.45}Ga_{0.55}As$ and 20 nm GaAs [7]. The 2-nm-thick GaAs layer fills up the nanoholes in the underlying AlGaAs barrier leading to the formation of inverted GaAs QDs below a thin GaAs quantum well (QW). For single-QD and ensemble investigations, the QD density is chosen as 1×10^8 cm^{-2} and 4×10^9 cm^{-2}, respectively. To perform single QD PL spectroscopy, 1 μm^2 mesa structures were fabricated by optical lithography and wet etching. The sample was mounted in a cold-finger helium flow cryostat which can be moved by computer-controlled xy-linear translation stages for exact positioning with a spatial resolution of 50 nm. For the excitation of our sample structure, the laser light was focused by a microscope objective (with numerical aperture $NA = 0.6$) to a spot diameter of 2 μm. The same microscope objective was used to collect the QD emission. The collected luminescence was then spectrally filtered by a 0.5 m focal length monochromator equipped with a liquid nitrogen cooled charge coupled device (CCD) for PL measurements. The samples were excited by a continuous wave (cw) laser emitting at 532 nm (see Ref. [21] for further details). For photon statistics measurements, the PL light is sent to a modified HBT correlation setup [22]. The HBT consisted of a 50/50 non-polarizing beam splitter and two single-photon counting avalanche photodiodes (SAPDs) each providing a time resolution of ~700 ps. The SAPDs output signals were used to trigger the start and stop channels of a time-to-amplitude converter the output of which was stored in a PC-based multichannel analyzer. In this way, a histogram $n(\tau)$ of photon

correlation events as a function of the time delay $\tau = t_{stop} - t_{start}$ was recorded.

Figure 1(a) compares the room temperature PL emission from an unstrained single GaAs QD located at 1.545 eV (solid line) and a PL spectrum from ensemble GaAs QDs peaked at 1.528 eV (dashed line) from a different sample with high QD-density taken at excitation power of ≈ 480 Wcm^{-2} with integration times of 1 s. The PL lines located at 1.429 eV, 1.581 eV, and 1.708 eV are assigned to bulk GaAs, the first excited state of the single GaAs QD, and the quantum well, respectively. The PL peak energy of the ensemble shows a redshift compared to the single QD because the high-density sample is characterized by slightly larger QDs. The ensemble spectrum was shifted to higher energy by 17 meV for comparison. The ground state PL linewidth of the single GaAs QD at room temperature is ≈ 8.5 meV, which is comparable to the inhomogeneous linewidth of the ensemble GaAs QDs (≈ 12.4 meV). This is a surprising result, since it is commonly assumed that the FWHM of a QD ensemble is mainly determined by the size/composition fluctuations of the QDs. While this remains true at low temperature, Fig. 1(a) shows that the room-temperature emission-linewidth of our QDs is dominated by the homogeneous broadening of the single QD emission. Such a result, which is attributed to the good size homogeneity of our QDs ($\pm 6\%$ in height [8]),
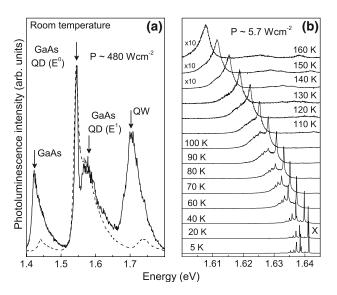

Fig. 1 (a) PL spectra measured at room temperature from a single GaAs QD (solid line) and from an ensemble of GaAs QDs (dashed line). For the excitation of the ensemble sample, the laser light was focused using a 5 × microscope objective to a spot diameter of 20 μm and about 10^4 QDs were being excited. The ensemble spectrum was shifted to higher energy by 17 meV for comparison. (b) PL spectra of a single GaAs QD as a function of the temperature

suggests the possibility of using these QDs with large surface density as efficient active region (e.g., in a microcavity laser), which could allow for plenty of new application relevant research as well as fundamental physics studies.

The single GaAs QD PL spectra as a function of the temperature taken at ≈ 5.7 Wcm^{-2} excitation power are shown in Fig. 1(b). The graphs show a clear shift of the center of the excitonic luminescence to longer wavelengths due to the bandgap reduction with increasing temperature. The PL spectrum taken at low temperature consists of resolution-limited well separated sharp emission lines with no background. The X line is well visible up to 100 K, above this temperature X overlaps with multiexcitonic lines. Below 130 K, the emission is rather intense so integration times of 1 s were sufficient to obtain reasonable signal-to-noise ratios. The temperature increase causes the PL linewidth to increase, since the contribution of the phonon sidebands becomes larger.

In Fig. 2, we display a typical temperature variation of the peak position energy and the homogeneous linewidths of the X PL-line of a single GaAs QD ($T \leq 100$ K) and QD ground-state ($T > 100$ K) deduced from the Lorentzian profile taken at ≈ 5.7 Wcm^{-2} excitation power. The reasons of choosing this power for the temperature dependence measurements are: (a) It was the minimum power of
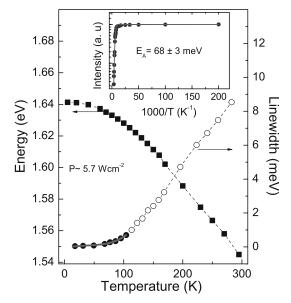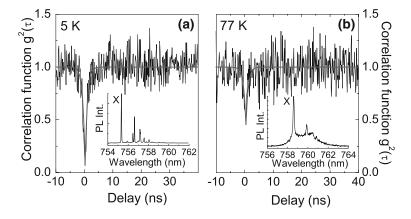
obtaining a reasonably good PL signal up to room temperature and (b) from the power dependence measurements taken at 5 K no effect was found on the linewidths of the X PL line for powers up to ≈ 7 Wcm^{-2}. The PL peak energy shows a redshift of about 97 meV as the temperature increases from 5 K to 295 K due to the bandgap shrinkage. In contrast with the behavior of InGaAs QDs [23], the redshift of the PL peak energy of GaAs QD ground-state (X) with rising temperature follows the thermal shrinkage of the bulk GaAs and is well fitted using the empirical Varshini equation (not shown here). We believe that the disagreements found in Ref. 23 have to be attributed to strain-related phenomena. Taking into account the spectral resolution of our setup (90 μeV) we obtain for the X an intrinsic linewidth Γ of 24 ± 15 μ eV at 5 K ($\Gamma = \sqrt{\Gamma_m^2 - \Gamma_{res}^2}$ [24], where Γ is the intrinsic linewidth, Γ_m is the measured linewidth, and Γ_{res} is the spectral resolution of our setup). This value increases only by a few μeV as the temperature is raised to 40 K. Furthermore, as the sample temperature is increased, the PL spectra become broader due to the contribution of mainly two mechanisms: (a) Phonon coupling which leads to the appearance of a broad background that emerges at both sides of the zero-phonon line which is assigned to the phonon sidebands [25] and (b) phonon scattering, which gives rise to the broadening of the excitonic zero-phonon transition [26]. As already mentioned above, the X line is clearly visible up to 100 K. Therefore, for a more quantitative analysis of the coupling mechanism the zero-phonon line is separated from the phonon sidebands by fitting only the central part of the X line by a Lorentzian. The linewidth data are fitted using the following relation, which describes the temperature dependence of the excitonic peak broadening in quantum wells and bulk semiconductors [27] and which has also been used for QDs [28, 29]: $\Gamma = \Gamma_0 + \gamma_{Ac}T + \gamma_{Op}(e^{\hbar\omega_{LO}/k_BT} - 1)^{-1}$, where $\Gamma_0 = 84 \pm 13$ μ eV is the zero K linewidth, the second term (γ_{Ac}) gives the acoustic phonon scattering, and the third term gives the scattering with optical phonons. $\hbar\omega_{LO}$ represents the energy of the longitudinal optical (LO) phonon of GaAs. The phonon broadening exhibits a linear variation for $T \leq 40$ K. The linear term in the above equation has been reported by many research groups [see e.g., Refs. 15 and 28]. In the quantum well case, the linear temperature variation accounts for the exciton absorption of acoustic phonons of energies much smaller than k_BT [30]. The physical origin of the observed linear term in the temperature dependence of the homogeneous linewidth in QDs is still under debate. At low temperature,

Fig. 2 Temperature dependence of PL energy positions (■) and homogeneous linewidth of the exciton PL line (●) and the dot ground-state (○) in a single GaAs QD. The solid line is a fit of the experimental data in the range 0–100 K. The inset is an Arrhenius plot of the integrated PL intensity as a function of reciprocal temperature

Fig. 3 Autocorrelation measurements under cw laser excitation obtained from the exciton photon of a single GaAs QD at (**a**) 5 K and (**b**) 77 K. The insets display the corresponding PL spectra

we found an acoustic-phonon broadening $\gamma_{Ac} = 1.0 \pm 0.1$ μeV K^{-1} in a single QD, which suggests that the linewidth does not depend strongly on the temperature in this range. This value is slightly larger compared to the values obtained from InGaAs QDs reported by Bayer et al. [15] and Borri et al. [31] but smaller than the value obtained by Urbaszek et al. [24]. At higher temperature, a slight contribution of the phonon side-bands to the linewidth data deduced from the Lorentzian fit of the central peak of the X PL-line cannot be excluded. We estimate an upper limit for optical-phonon broadening (γ_{Op}) in a single QD to be 30 ± 2 meV from the fit of the above equation which suggests that a strong electron-LO-phonon interaction occurs.

The integrated PL intensity of the ground state transition of the single QD is shown in the inset of Fig. 2. The integrated intensity remains almost constant up to 100 K and shows an exponential quenching at higher temperatures. An activation energy $E_A = 68 \pm 3$ meV is derived from the data fit of the integrated intensity using [32]: $I = I_0/[1 + C\exp(-E_A/k_BT)]$, where I is the integrated PL intensity, I_0 is the PL intensity at 5 K, C is the transition rate (the ratio of the thermal escape rate to radiation recombination rate), and E_A is the activation energy. The measured activation energy is nearly half of the total barrier height [33] of 140 meV (i.e., the sum of the barrier heights for electrons and holes). A possible explanation is that the carriers behave as correlated electron-hole pairs [34].

Autocorrelation measurements have been performed to demonstrate single photon generation as a function of temperature under cw excitation of the single GaAs QD. Figure 3(a), (b) show the measured normalized correlation function $g^{(2)}(\tau)$ of the (X) QD emission at 5 K and 77 K, respectively. The corresponding PL spectra are shown in the insets. Both traces exhibit a clear dip in the correlation counts for

the time delay $\tau = 0$ ns, indicating a strong photon antibunching. The $g^{(2)}(\tau)$ is fitted by a function of the form: $g^{(2)}(\tau) = 1 - a\exp(-|\tau|/t_m)$, where a accounts for the background present in the measurements and t_m is the antibunching time constant. The values of $1 - a = g^{(2)}(0)$ obtained from the fit are 0.06 for the trace (a) which shows a nearly perfect single photon emitter and 0.45 for trace (b). In both cases, $g^{(2)}(0) < 0.5$ is a signature of a single quantum emitter. The measured $g^{(2)}(0)$ does not reach its theoretical value of zero because of the presence of a weak uncorrelated background at low temperature. At high temperatures, a stronger background contributes to the PL spectra due to the acoustic phonon sidebands [25] and the presence of the transitions involving holes in the excited states [20], which leads to the reduction of the photon antibunching from the X line. In the presence of a background the value of the $g^{(2)}(0)$ is increased by a factor of $1 - \rho^2$, where $\rho = S/(S + B)$ is the ratio of signal S to background B counts [35]. From the PL spectra (insets of Fig. 3) ρ was determined to be 0.97(0.75) for $T = 5$ K (77 K). The resulting values for $g_B^{(2)}(0)$ are 0.06 (0.44), which are in good agreement with the $g^{(2)}(0)$ values.

In summary, we have studied the temperature dependence of the luminescence of single unstrained self-assembled GaAs quantum dot (QD) structures. Single QD spectroscopy showed that the QDs are characterized by intense room temperature PL emission. Surprisingly, it was found that at room temperature the linewidth of the single QD is comparable to the ensemble inhomogeneous broadening. The single quantum emission nature was demonstrated at elevated temperatures by photon correlation measurements. The QD PL emission energies, homogeneous linewidths and the thermally activated behavior were discussed.

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