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Coherence and Dephasing in Self-Assembled Quantum Dots

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Abstract—We measured dephasing times in InGaAlAs self-assembled quantum dots at low temperature using degenerate four-wave mixing. At 0K, the coherence time of the quantum dots is lifetime limited, whereas at finite temperatures pure dephasing by exciton-phonon interactions governs the quantum dot coherence. The inferred homogeneous line widths are significantly smaller than the line widths usually observed in the photoluminescence from single quantum dots indicating an additional inhomogeneous broadening mechanism in the latter.

Quantum dots; exciton coherence; dephasing; homogeneous and inhomogeneous line widths.

I. INTRODUCTION

With recent progress in the fabrication of self-assembled semiconductor quantum dots (SAQDs), the realization of a solid-state system with atom-like electronic and photonic properties has become feasible [1]. Along with the technological and scientific challenges associated with the fabrication and characterization of semiconductor quantum dots, their application potential has been widely forecast and discussed [2]. Many of the predicted properties of quantum dots (QDs) rely on a two-level (or few-level) picture of their electronic structure. The application of QDs in semiconductor optoelectronic devices like lasers and amplifiers has been triggered by predictions of lower thresholds, higher differential gain, less temperature dependence, lower chirp, higher modulation bandwidth etc. [3] In these applications, large ensembles of quantum dots are needed, and the relation between the homogeneous and the inhomogeneous broadening of the QD optical transitions plays an important role [4]. Another exciting application of the two-level nature of QDs would be as building blocks in quantum logic devices [5], where the coherence/dephasing time of the individual QDs is a crucial fundamental parameter. An equally important technological challenge is the control and positioning of the individual dots [6].

The dephasing time of QD transitions can be measured in frequency space as the homogeneous line width in e.g. photoluminescence (PL) spectra of single quantum dots [7,8]. Alternatively, it is measured in time domain by transient four-wave mixing (TFWM), or photon echo, experiments on an ensemble of quantum dots [9,10]. Both techniques have been pursued, and for weakly confined quantum dots formed by well-width fluctuations in narrow quantum wells this has led to consistent results for the low-temperature dephasing times of QD excitons [11,12]. For the more strongly confined excitons in SAQDs, however, four-wave mixing experiments have shown very long low-temperature dephasing times approaching 1ns [9,10], whereas single-dot PL has shown line widths corresponding to dephasing times more than an order of magnitude shorter [7,8].

In the present article we shall demonstrate that special care needs to be taken in order to ensure that the true homogeneous line width is measured in single-dot PL experiments consistent with the very long dephasing time measured in photon echo experiments [13]. One of the difficulties in performing nonlinear four-wave mixing experiments on quantum dot ensembles is the relatively small interaction volume normally available. This can be overcome by performing the QD TFWM in a waveguide geometry using heterodyne detection to separate the signal from the incident beams [4,10] or by conventional two-beam TFWM on stacked multi-layer QDs [9]. In this work we shall concentrate on the latter.

II. EXPERIMENTAL

For the single-dot PL investigation, we used a single layer of MBE-grown In0.53Al0.47As SAQDs embedded in Al0.53Ga0.47As barriers. Thin layers of AlAs were placed 20nm above and 100nm below the dot layers to prevent carrier diffusion to the surface and the GaAs substrate. In order to limit the detection area without disturbing the physical environment of the dots, a thin gold film with sub-micron sized apertures, fabricated by electron-beam lithography, was deposited on the surface of the sample. Another sample with ten layers of In0.53Al0.47As SAQD was grown for TFWM and time-resolved PL (TR-PL) experiments. Here an antireflection coating was applied to the sample back to prevent multiple reflections of the exciting beams.

PL was excited and detected through a microscope objective (µPL) located inside a closed-cycle He cryostat. As excitation, we used two different continuous-wave light sources; a He-Ne laser (632.8nm), and a frequency doubled Nd:YAG laser (531nm). For high-resolution spectroscopy, the PL was dispersed in a 2-m Littrow spectrometer and detected.
with a cooled CCD camera. In TFWM and TR-PL experiments
the sample was cooled in a liquid helium cryostat and excited
with 120-fs pulses from a Ti:sapphire laser. We used the $2k_1-k_1$
transmission geometry and detected the signal using a silicon
photo diode, with a low-noise preamplifier, and lock-in
techniques. In TR-PL the sample was excited in the barriers
and the PL was collected, dispersed, and synchronously
detected using a streak camera with 2.5-ps time resolution.

III. RESULTS

A. Luminescence

The PL spectrum at 10K from a 500-nm aperture over a
single layer of QDs is shown in the left inset of Fig. 1. The
spectrum consists of a large number of well-resolved single
lines. The section of the spectrum in the rectangular box is
recorded at high spectral resolution and shown in Fig. 1. The
lines are well approximated with Lorentzians as shown with
the solid line. The width (FWHM) is extracted for 54 lines and
shown in the line-width histogram in the right inset of Fig. 1.
Other apertures show similar line-width statistics. The most
commonly measured line width is about 65-eV. When
corrected for the spectral resolution (20-eV), we obtain a
typical dot line width of about 45-eV, which is in good
agreement with published data on line widths from self-
organized quantum dots [7,8].

In order to be able to correct for the temperature differences
in single-dot and TFWM experiments, we have performed
temperature dependent single-dot PL measurements. We find
that the widths of the single-dot lines up to a temperature of
approximately 40K are well described by the linear relation

$$\Gamma(T) = \Gamma(0) + aT$$

(1)

where $a = 0.5$-meV/K and $\Gamma(0)$ is described by a distribution
peaking at 40-meV.

B. Time-resolved luminescence

In narrow quantum wells the homogeneous line width of
localized excitons is dominated by lifetime broadening at the
lowest temperatures. The question is whether the same holds
true for SAQDs. We have performed time resolved PL
spectroscopy on the ten-layer sample. This allows us to
evaluate the PL decay time for individual spectral components
across the entire luminescence band. We show in Fig. 2 the
time-integrated PL spectrum together with the spectrally
resolved decay times. The data show a spectral dependence of
the PL decay times from 650 ps at the high-energy side to
850 ps at the low-energy side of the PL spectrum. The time-
integrated PL spectrum coincides with the PL obtained under
continuous-wave excitation. The PL spectrum and PL decay
times are nearly independent of excitation density over two
orders of magnitude. We therefore conclude that the decay
times observed are the lifetimes $T_1$ of the dot ground states.
This allows us to calculate the contribution to the
homogeneous line width from population decay to be about
1-meV.

C. Single-dot spectroscopy

In the following we investigate the properties of the single
dot spectra in more detail. The PL spectra consist of a number
of sharp lines, which are easily separated spectrally due to the
large inhomogeneous broadening of the QD ensemble. A small
part of such a spectrum is shown in the inset of Fig. 3 for three
different excitation powers (531-nm excitation). At low power,
we observe the ground-state transition lines ($X_1, X_2, X_3$), which
increase linearly in intensity with the excitation power.
Eventually, the intensities of these lines saturate and
simultaneously additional lines appear (e.g. $XX_1, XX_2$) which
increase quadratically with excitation power. We attribute
these additional lines to biexciton states (not necessarily
originating from the same dots as the exciton lines shown
here).

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Figure 1. Time-resolved PL. The left axis shows the intensity of the time-
integrated PL, and the right axes show the measured decay times and
corresponding homogeneous broadenings at different spectral positions.

Figure 2. High-resolution PL spectrum at 10K through a 500-nm aperture. The
solid line is a Lorentzian fit to the data. The left inset shows all the lines from
the 500-nm aperture. The right inset shows the line width statistics.

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Figure 3. Single-dot spectroscopy. The $\mu$PL spectra consist of a number of
sharp lines, which are easily separated spectrally due to the
large inhomogeneous broadening of the QD ensemble. A small
part of such a spectrum is shown in the inset of Fig. 3 for three
different excitation powers (531-nm excitation). At low power,
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these additional lines to biexciton states (not necessarily
originating from the same dots as the exciton lines shown
here).
We focus our attention on the narrowest lines observed in the μPL spectra, e.g. the line \( X_1 \) shown in Fig. 3. We determine the spectrometer response function by recording a narrow line from a low-pressure Xe lamp. Both lines are well represented by Lorentzians with the widths of 23μeV and 17μeV (FWHM), respectively. Assuming that the measured PL line is a convolution of two Lorentzians, we obtain a deconvoluted line width of 6μeV for the QD emission. This value is in excellent agreement with the homogeneous broadening measured in the same type of dots at 10K using TFWM [9], as we shall discuss in the next section. In similar TFWM experiments, Borri et al. found homogeneous line widths as low as 2μeV at 7K in dots where the excitons are more strongly confined [10].

The TFWM experiments indicate that the dot ensemble is well characterized by a single dephasing time, suggesting that all dots have roughly the same homogeneous broadening. In the low-temperature μPL spectra, however, a distribution of line widths is found [9,14], where only a fraction of the line widths are approaching the value measured in TFWM. For other transitions, such as \( X_2 \) and \( X_3 \) in Fig. 3, an additional broadening mechanism is therefore present. This may be spectral fluctuations, as have been observed in real time in III-V [15] and II-VI [16] quantum dots and attributed to time-varying charge configurations in the vicinity of the dots. We observe minor variations (<3μeV) in the position of the PL lines in successively recorded spectra on a timescale of minutes, which is larger than the integration time used here (10-30s), and therefore does not influence the line width. We note also that the broader lines typically have a Gaussian line shape, indicative of random fluctuations, in contrast to the narrower lines that have a Lorentzian shape. Thus, we conclude that dot-dependent spectral fluctuations, possibly due to charge fluctuations in the vicinity of the dots and occurring on a timescale faster than we can detect using our present set-up, are responsible for the observed distribution of line widths.

D. Transient four-wave mixing

The TFWM results are recorded at an excitation density of 190 W/cm², which is more than one order of magnitude larger than we usually apply to quantum well samples. This is due to the reduced absorbance of the QD layers, leaving only a small fraction (<20%) of the quantum dots excited. The TFWM intensity still varies with excitation intensity to the third power in agreement with the underlying \( \chi^{(3)} \) process.

The TFWM signal recorded at three different spectral positions of the laser (a, b, and c) and at 5 K is shown in Fig. 4. The inset shows the 5-K PL spectrum and the spectral positions of the laser for the three TFWM traces. A dominating signal at pulse overlap (\( \tau = 0 \)) is common for the three TFWM traces. This signal is due to a non-resonant two-photon transition to the GaAs substrate and is unrelated to the quantum dots. For longer delays, a much weaker signal is observed. This weaker signal is seen to depend on the detuning of the laser with respect to the QD PL peak. For positive detuning (c) the TFWM signal is dominated by a fast component for short delays, and a much slower component for longer delays. The influence of the fast component is seen to diminish for more resonant tuning (b) and it disappears for negative detuning (a). The slow component, on the other hand, is observed to persist across the entire QD distribution.

The TFWM data are fitted well by a bi-exponential decay demonstrating that two distinct processes contribute to the TFWM signal. To obtain more quantitative information of the origin of the two processes we have recorded the TFWM signal for a number of different spectral positions of the laser. Each of these TFWM traces is fitted with a bi-exponential decay, and two decay times and two amplitudes are extracted.

Figure 3. PL line from a single QD (filled circles) compared with the spectrometer response (open circles). Both lines are fitted by Lorentzian functions (solid lines). The inset shows spectra, recorded at different excitation powers. Exciton (X) and biexciton (XX) lines are indicated.

Figure 4. TFWM data for three different detunings of the laser (a, b, and c) as indicated in the inset showing the PL spectrum and the laser spectrum centered at (c).
IV. SUMMARY

We have in TFWM experiments observed ultra-long coherence times of the dot ground state, which are more than one order of magnitude longer than found for localized excitons in narrow quantum wells. We confirm that dephasing times in excess of 1ns are attainable in self-assembled quantum dots, which should enable the demonstration of quantum logic operations in the solid state. We have also shown that a pure dephasing-limited line width can be observed in PL from III-V single quantum dots under proper excitation conditions.

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