Ultralong Dephasing Time in InGaAs Quantum Dots

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We measure a dephasing time of several hundred picoseconds at low temperature in the ground-state transition of strongly confined InGaAs quantum dots, using a highly sensitive four-wave mixing technique. Between 7 and 100 K the polarization decay has two distinct components resulting in a non-Lorentzian line shape with a lifetime-limited zero-phonon line and a broadband from elastic exciton-acoustic phonon interactions.

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One fundamental aspect of light-matter interaction is the time evolution of the polarization induced in a medium by a coherent light field. A common approach is to model an optical transition by an ensemble of identical, independent, two-level systems and attribute the time decay of the macroscopic polarization to a loss of amplitude of the microscopic dipoles due to population relaxation (inelastic dephasing) or to a loss of phase coherence among the dipoles (pure dephasing). Many experiments were dedicated to measuring the decay time of the macroscopic polarization (usually also affected by a distribution of the transition energies, the so called inhomogeneous broadening) in semiconductors where, however, the two-level system simplification hardly applies [1]. The achievements in fabrication of quantum dots (QDs) opened the possibility of investigating the light-matter interaction with atomiclike objects in semiconductors [2]. From the fundamental argument of energy and momentum conservation, it was predicted that, due to the discrete energy level scheme in a QD, the interaction of carriers with acoustic phonons is inhibited (phonon bottleneck), resulting in a dephasing time at low temperatures limited only by the radiative lifetime [3].

With the recent development of growth techniques, epitaxially grown QDs with high crystal quality can be fabricated [4]. The dephasing time in these QDs is widely addressed experimentally, mainly by measuring the homogeneous linewidth (inversely proportional to the dephasing time) by isolating a few dots from the inhomogeneously broadened ensemble. For excitons weakly confined by the lateral disorder in narrow GaAs quantum wells it was indeed found that the homogeneous broadening of the dot ground-state transition at low temperature is limited by the radiative lifetime [3,5]. For strongly confined In(Ga)As/GaAs self-organized quantum dots, very attractive for applications such as QD lasers [4], the homogeneous broadening at low temperature is measured to be in the range of 50 μeV, much larger than the lifetime limit (radiative lifetimes are in the nanosecond range), and thus attributed to pure dephasing [6,7]. Recently it has been pointed out for II-VI QDs that the homogeneous broadening at low temperature is no longer Lorentzian but consists of a Lorentzian zero-phonon line (ZPL), broadened by inelastic processes, and a non-Lorentzian broadband due to elastic interactions with acoustic phonons [8]. However, the radiative limit of the ZPL was not reached, probably due to an additional dynamical broadening originating from a slow spectral shift of the lines (the so called spectral diffusion), a well known problem in single-dot spectroscopy [9].

Transient four-wave mixing (FWM) spectroscopy is a powerful tool to directly measure the dephasing time even in an inhomogeneously broadened system [1]. However, while it has been reported in GaAs islands [5] and in II-VI QDs [10,11], the only time-resolved FWM on the ground-state transition of InGaAs quantum dots was reported at room temperature by us using a waveguide geometry to enhance the interaction length [12].

In this Letter, we present the first FWM in strongly confined InGaAs QDs in the full temperature range from 7 to 300 K. We find that the dephasing time at low temperature

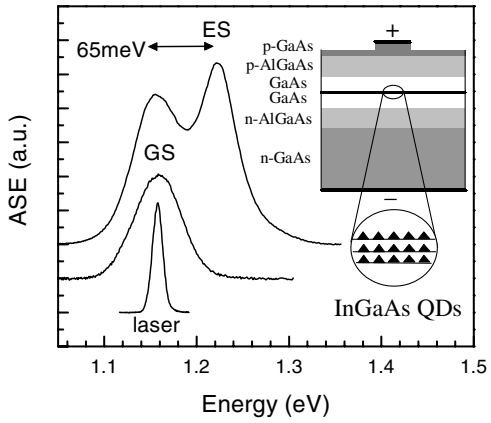

FIG. 1. Amplified spontaneous emission at 25 K at low (0.5 mA) and high (20 mA) injection current. The dot ground-state transition (GS) is inhomogeneously broadened and separated to the first excited state transition (ES) by 65 meV. The spectrum of the pulse used in the experiment is shown. The curves are vertically displaced for clarity. A scheme of the quantum dot waveguide is also given.

is *several hundred picoseconds,* which corresponds to *only a few μeV homogeneous broadening,* never observed in single-dot spectroscopy. Moreover, the polarization decay has a nonexponential dynamics with a fast and a slow component resulting in a non-Lorentzian line shape. The investigated sample is a *p-i-n* structure grown by metal-organic chemical vapor deposition containing three layers of self-organized $In_{0.7}Ga_{0.3}As$ QDs separated by 35-nm-thick GaAs spacers. Two AlGaAs cladding layers and a ridge structure of 5-μm width and 500-μm length provide optical waveguiding. The end facets were tilted to avoid multiple reflections. With electrical injection, amplified spontaneous emission (ASE) is detected and spectrally resolved, as shown in Fig. 1, at 25 K. At this temperature, thermalization among the dots does not occur and the ASE at low injection current reflects the inhomogeneous distribution of the dot ground-state transition due to the random capture of the injected carrier by the dots [4]. We infer a Gaussian inhomogeneous broadening of 60-meV full width at half maximum (FWHM) attributed to fluctuations in dot size and indium concentration. At high injection current, filling of the first optically active excited state is visible which has 65-meV energy separation from the ground-state transition. Photoluminescence on a reference sample shows a wetting layer transition $\sim$210 meV above the dot ground state, evidencing the strong confinement. QD lasers fabricated from the same structure showed efficient ground-state lasing at room temperature with low transparency current densities ($\sim 6\ A/cm^2$ per active layer) indicating the good crystal quality and the high quantum efficiency of the dots [13]. The FWM experiment is performed using an optical parametric oscillator providing Fourier-limited $\sim$150-fs pulses at 76 MHz repetition rate, with a wavelength tunable to the center of the dot ground-state transition (as shown in Fig. 1) which ranges from 1170 nm at 300 K to 1070 nm at 7 K. At this wavelength position we estimate a negligible contribution (2%) of the excited states to the optical density of states. Two pulses are coupled into the transverse electric waveguide mode with a relative delay time τ_P. The FWM signal is detected using a heterodyne technique similar to the one discussed in our previous work [12], but modified for the high-repetition rate with an improved signal-to-noise ratio. In this technique a reference pulse interferes with the FWM signal and its delay time relative to the signal is varied in order to time resolve the FWM [12]. Note that the FWM *field amplitude* is measured via its interference with the electromagnetic field of the reference pulse. The sample was held in a specially designed cryostat allowing for light coupling in and out of the waveguide with a high numerical aperture, and the FWM measurements were performed without electrical injection.

In the heterodyne technique [12] the pulse 1 (2) is frequency shifted by ω_1 (ω_2) and the FWM signal is detected at $2\omega_2 - \omega_1$. The FWM of the inhomogeneously broadened dot ensemble is a photon echo in real time, as clearly shown in Fig. 2. Here, τ_R measures the delay of the reference pulse with respect to pulse 2; i.e., pulse 1 is at $\tau_R = -\tau_P$ and we measure a FWM signal centered at [12] $\tau_R = \tau_P$. The pulse energy of pulse 2 is twice that of pulse 1, and in the range of $I_0 \leq 0.1$ pJ, corresponding to an absorption bleaching of $\sim$10% relative to the maximum absorption ($\sim 30\ cm^{-1}$, 1.5 optical density), i.e., well below one electron-hole pair excited per dot [14]. The FWM signal is in the third-order regime as shown in the right part of Fig. 2 where the time integrated FWM (TI FWM) is shown versus τ_P for different pulse energies. At low excitation intensity we clearly observe beats in the initial FWM dynamics that we assign to exciton-biexciton beats [15,16] from which we infer a biexciton binding energy of 3 meV similar to previous reports. With increasing intensity the signal scales as a third-order response and finally saturates, showing a changed exciton-biexciton beat as observed in quantum wells when fifth-order contributions are important [15].

It can be seen from Fig. 2 that for $\tau_P > 2$ ps a long-lived signal is present. In Fig. 3 we show the time dynamics of the TI FWM up to $\tau_P = 400$ ps and for different temperatures as indicated. From 300 to 125 K the dynamics is dominated by a dephasing time below 1 ps. Below 100 K a slow component appears which becomes dominant at smaller temperatures. This component has an exponential decay [17] with an increasing time constant with decreasing temperature. At $T = 7$ K the corresponding dephasing time is 630 ps, *equivalent to only 2 μeV homogeneous broadening*.

We point out that the experiment is sensitive to 3 orders of magnitude FWM electric field, i.e., *6 orders of magnitude FWM intensity,* as shown in Fig. 3 (see, for instance, the data at 50 K). Note also that the heterodyne detection

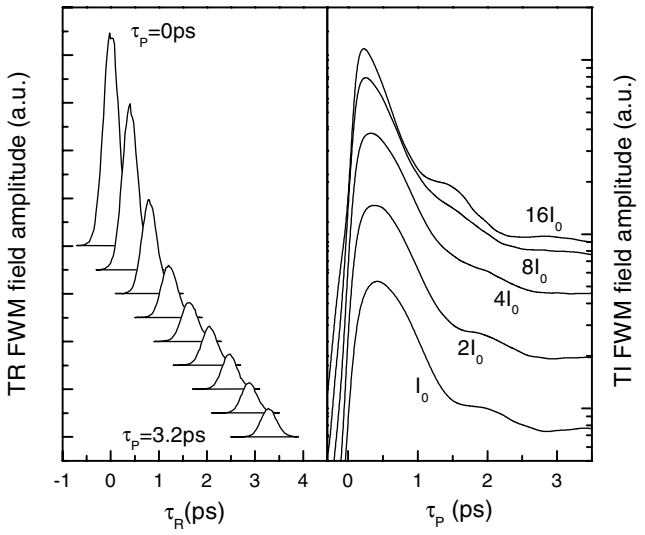

FIG. 2. Left: Time-resolved four-wave mixing field versus reference delay time at 50 K. The delay time between the exciting pulses varies in 400-fs steps from 0 to 3.2 ps, as indicated. Right: Time-integrated four-wave mixing versus delay time for different exciting intensities. The signal shown on the left part of the figure corresponds to $2I_0$.

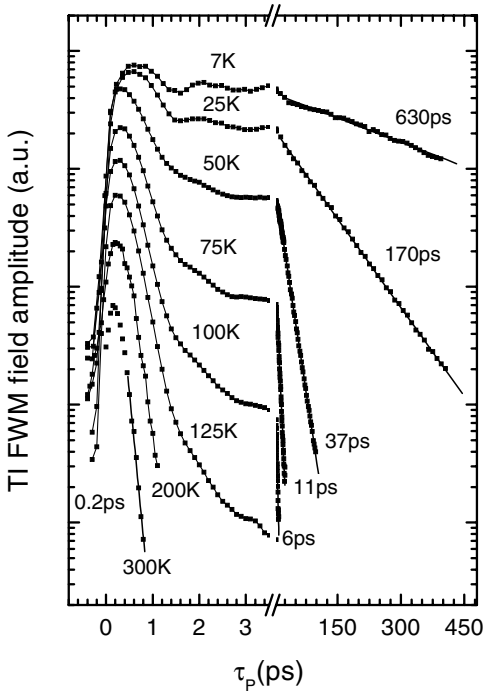

FIG. 3. Time-integrated four-wave mixing at different temperatures as indicated. The dephasing times inferred from the exponential decays are indicated together with the exponential fits.

is insensitive to nonlinear luminescence and scattered light. Moreover, FWM intrinsically overcomes the problem of spectral diffusion [9]. In FWM only the spectral shift of the emitters from the time between the pulse 1 and the formation of the echo signal will affect the echo. Since spectral diffusion is a slow process over seconds its effect is negligible in FWM. In contrast, single-dot spectroscopy that often requires long integration times can be severely affected by spectral diffusion. This might explain the observation of low temperature linewidths in InGaAs QDs in the range of 50 μeV, much larger than our finding.

In order to obtain more insight in the homogeneous broadening corresponding to the measured dephasing, we have calculated the Fourier transform of the TI FWM signal, shown in Fig. 4. Apart from the initial rise in the TI FWM and the exciton-biexciton beats, the time-integrated photon echo versus τ_P is a probe of the time evolution of the first order polarization [1,16] and its Fourier transform represents the homogeneous line shape [18]. It is instructive to see that at 50 K the long exponential decay results in a sharp Lorentzian line and the fast initial dephasing gives a weak broadband. At 75 K the sharp line broadens and the ratio between the broadband and the sharp line increases resulting in a strongly non-Lorentzian line shape similar to what was observed recently in II-VI QDs [8]. At 100 K the broadband is the dominant contribution resulting in a FWHM in the meV range [19].

We interpret the sharp Lorentzian line in Fig. 4 as the zero-phonon line broadened by inelastic processes such as radiative recombination and phonon-assisted activation into higher energy exciton states [8]. In Fig. 5 the temperature dependence of the ZPL width is shown together with a fit using 0.66 μeV radiative broadening summed to a linear and a temperature activated broadening proportional to the Bose population number. The radiative broadening is taken from the radiative lifetime of 1 ns measured in differential transmission. From the fit we obtain an activation energy of 16 $\pm$ 1 meV and a linear increase of 0.22 $\pm$ 0.02 μeV/K. The activation energy is attributed to the separation to the exciton state involving only the first excited hole level. This is consistent with the measured 65-meV separation to the optically active exciton state involving additionally the first excited state of the electron. These energy separations agree well with typical energy spacings calculated in InGaAs QDs [4,20]. The linear dependence is interpreted as the inelastic interaction with acoustic phonons with energies much smaller than k_BT even at these low temperatures, due to transitions between the different spin states of the exciton ground state. In fact, both the exchange splitting between bright and dark excitons and the splitting between the two bright exciton

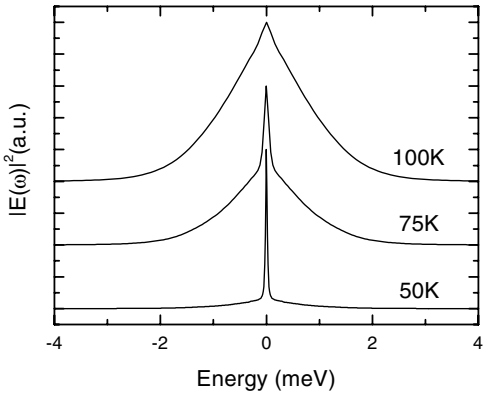

FIG. 4. Power spectrum of the time-integrated four-wave mixing field at different temperatures as indicated.

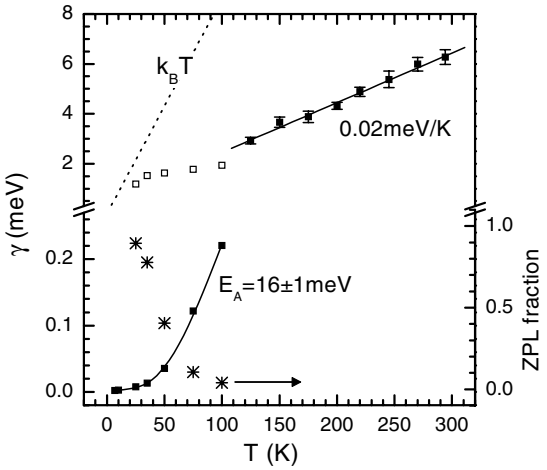

FIG. 5. Full width at half maximum homogeneous linewidths estimated from the measured dephasing. Below 100 K, the open squares are the linewidths of the broadband, while the closed squares are the linewidths of the zero-phonon line. The dotted line is the thermal energy k_BT. The stars are the ratio between the area of the zero-phonon line and the total area of the line.

states due to an asymmetry of the confinement potential in the QD plane are reported to be in the range of few hundred μeV in this type of dot [21].

The broadband in Fig. 4 is interpreted as the elastic acoustic-phonon broadening discussed in Refs. [5,8]. In this model the exciton-phonon interaction is treated in a molecular fashion where electronic excitations imply a shift in the lattice equilibrium position. In other words, the optical creation of an exciton locally distorts the lattice. The ground-state excitonic transition is described using the Frank-Condon principle and involves absorption or emission of acoustic phonons. The lattice distortion is of increasing importance with decreasing size of the confined exciton state, and it is very relevant for II-VI QDs. Our results show that also in III-V InGaAs QDs the lattice distortion is important. In Fig. 5 we show the FWHM of the broad line which we have inferred from the data shown in Fig. 4. Considering the limitations of the procedure used to deduce the line shape from TI FWM, these values can be subject to systematic errors, but they allow us to qualitatively evaluate the order of magnitude of the broadening. It is interesting to note that this line broadening only weakly depends on temperature. The cutoff energy of the acoustic modes involved in the exciton-phonon coupling is roughly determined by the inverse of the localization length and quantifies the size of the energy spread of the broad acoustic-phonon band [8]. One can qualitatively comment that, if $k_B T$ is bigger than this cutoff energy, all the phonon modes involved in the coupling are occupied and the broadening is given by the cutoff energy. When instead the temperature is low enough to inhibit the phonon population a decrease in the broadening with decreasing temperature occurs. Moreover, the ratio between the area of the acoustic-phonon band and that of the ZPL depends strongly on the phonon occupation. We find that the area of the broadband is only a 10% contribution to the total area at 25 K (see Fig. 5), while it dominates at 100 K.

Above 100 K the fast dephasing dominates the TI FWM decay up to room temperature. When fitting exponentially to these initial dynamics we infer a homogeneous broadening linearly increasing with temperature, with the slope shown in Fig. 5. Recent experiments using near field scanning microscopy showed a qualitatively similar trend in the homogeneous broadening of InGaAs quantum dots [22]. This was interpreted to be due to the interaction of excitons with acoustic and optical phonons that starts to be important above 100 K, as also discussed in Ref. [23]. Within our picture it is natural to interpret the fast dephasing above 100 K as related to the elastic acoustic-phonon scattering previously discussed. However, its change with temperature compared to the acoustic-phonon band below 100 K supports the possibility that additional processes, such as interactions with optical phonons, are coming into play.

In conclusion, we have reported the first measurement of a dephasing time of several hundred picoseconds at low temperatures in strongly confined InGaAs quantum dots, close to the radiative lifetime limit. The occurrence of such an extremely long coherence is a new result compared to what was known until now from single-dot spectroscopy and does bring a fundamental revision in the understanding of the line broadening in III-V quantum dots, as well as in their potential application in quantum information processing [24].

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[18] The phase is not known and was chosen to be constant, so that the resulting line shape is by construction symmetric.
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