Exciton lifetime in InAs/GaAs quantum dot molecules

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The exciton lifetimes T_1 in arrays of InAs/GaAs vertically coupled quantum dot pairs have been measured by time-resolved photoluminescence. A considerable reduction of T_1 by up to a factor of ≈ 2 has been observed as compared to a quantum dots reference, reflecting the inter-dot coherence. Increase of the molecular coupling strength leads to a systematic decrease of T_1 with decreasing barrier width, as for wide barriers a fraction of structures shows reduced coupling, whereas for narrow barriers all molecules appear to be well coupled. The coherent excitons in the molecules gain the oscillator strength of the excitons in the two separate quantum dots halving the exciton lifetime. This superradiance effect contributes to the previously observed increase of the homogeneous exciton linewidth, but is weaker than the reduction of T_2. This shows that as compared to the quantum dots reference pure dephasing becomes increasingly important for the molecules.

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The development of high-quality semiconductor quantum structures of varying dimensionality has allowed attention to be focused on coupling of these systems. Some interest in such activities arises from application side as the resulting functional units have the potential to form building blocks of a new generation of electronic and optoelectronic devices. Some interest arises also from basic physics as it gives detailed insight into quantum-mechanical coupling, for example. The most simple structure is a pair of quantum dots (QDs), coupled by tunneling. For their fabrication, a variety of techniques may be applied, such as double-cleaved edge overgrowth,[1] lateral patterning of double quantum wells,[2] or gating of two-dimensional electron gases.[3]

Here we focus on vertically coupled dot structures fabricated by self-assembly, which seems ideally suited for their realization: when growing two QD layers in close vicinity, the strain that surrounds a dot in the first, lower lying layer enforces the growth of a second dot on top of the first.[4] Although vertical stacking is well established, spectroscopic studies of these structures are just at their beginning.[5] Spectra taken on arrays suffer from inhomogeneous broadening. Only recently has it become possible to observe a well-resolved confined shell structure in photoluminescence of InAs/GaAs quantum dot molecules (QDMs). This progress has been made possible by extending the conventional Stransky-Krastanow growth scheme by an In-flush.[6] Through this modification the dot geometries in the two layers become very similar, while the size of the upper dot is considerably larger than that of the lower one in the nonextended fabrication scheme. In particular, the homogeneity of the dot height, which is the most crucial point for reducing the ensemble broadening, is improved.

A proof of quantum-mechanical coupling in these vertically aligned QDs was provided by exciton fine structure studies: pronounced anticrossings were observed in the magnetic-field dispersion of the fine structure because of field-induced state hybridizations.[7] In addition, systematic dependencies of a variety of spectroscopic quantities, such as orbital and spin-energy splittings, diamagnetic shifts, etc., on barrier width were observed.[8,9] The entirety of these results strongly support tunneling as the coupling mechanism in the QDMs, through which spatial coherency of the wave function is established.

Besides coherency in space, also temporal coherency of the excitons has already been addressed in arrays of InAs/GaAs QDMs, by measuring the dephasing with four-wave mixing.[10] For the parameters by which dephasing in three-dimensionally confined geometries can be characterized, such as homogenous linewidth, zero-phonon line weight, and activation energies, systematic dependencies on barrier width have been observed. Most importantly, the homogeneous linewidth at cryogenic temperatures increases strongly with decreasing barrier width by a factor of ≈ 6 as compared to the QDs reference. In the dots the dephasing time is about 600 ps, and it is reduced to slightly more than 100 ps only for molecules with narrow 4 and 5 nm barriers. For QDs it has been shown that the exciton dephasing is ultimately limited by the radiative lifetime of the excitons, and pure dephasing may not be important, depending on the dot structure under study.[11] One point of the present paper is to assess the importance of pure dephasing in molecular structures. For this purpose we complement the previous four-wave-mixing studies by exciton lifetime measurements. By doing so, we also address the effect of spatial coherency of the exciton wave function on its oscillator strength.

Details of the QDM sample fabrication with barrier widths of $d=4$, 5, 6, 7, 8, and 16 nm barriers can be found elsewhere.[6,12] The QDM samples were mounted on the cold finger of a microscope cryostat, which allowed a variation of the sample temperature down to 5 K. For optical excitation a mode-locked Ti:sapphire laser pumped by a frequency-doubled $Nd:YVO_4$ laser was used. The system provides trains of pulses with a duration of ≈ 150 fs at a repetition rate of 76 MHz. The laser wavelength was 780 nm, corresponding to an excitation energy slightly above the GaAs barrier. Also, experiments with excitation wavelengths around 850

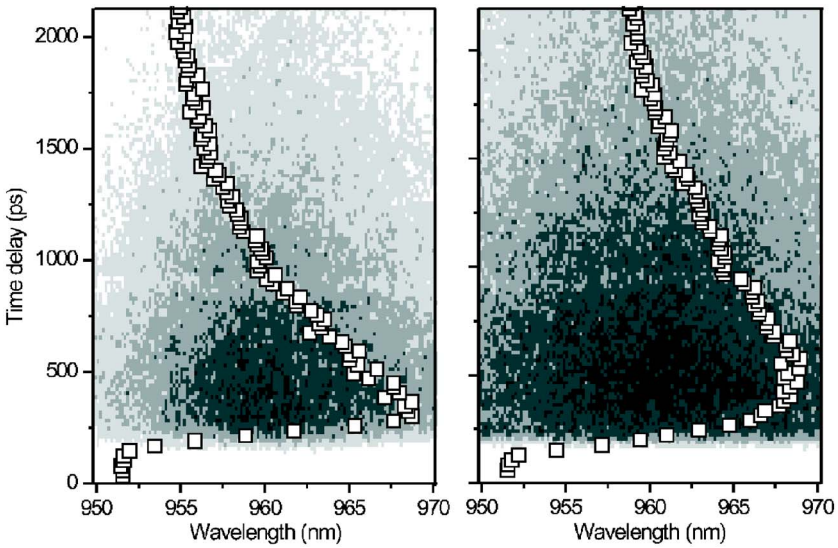

FIG. 1. (Color online) Gray scale contour plots of the photoluminescence emission decay from the QDs reference sample (right panel) and the QDM sample with a 4 nm wide barrier (left panel) at $T=10$ K. The signal was integrated in both cases for 60 s. Symbols are characteristic traces at the center of the emission band. The peak intensity has been normalized to unity. Note that there is some intensity reduction around 900 ps because of reduced sensitivity of the streak screen.

nm (energetically below the GaAs barrier into the wetting layer) were performed, but no significant changes of exciton dynamics were observed. The laser was focused to a spot size of 10 μm. From the structural density of $\sim 10^{10}$ cm^{-2}, we estimate that a few thousand molecular structures were addressed spectroscopically. The excitation power was 40 Wcm^{-2}, small enough to avoid many body effects and to address single excitons. The emission was spectrally analyzed by a single-grating monochromator ($f=0.5$ m, 300 rules per millimeter) and detected by a Hamamatsu synchroscan streak camera equipped with a Peltier-cooled S1-tube. To enhance sensitivity it contains a double multichannel plate. With this configuration a time resolution of about 30 ps was obtained.[13]

Figure 1 shows streak camera screens recorded at $T=10$ K for the QDs reference (the right panel) and the $d=4$ nm barrier QDMs (the left panel): normalized contour plots of the photoluminescence emission as function of the time delay after pulsed excitation (the vertical axis) and the emission wavelength (the bottom axis) are given. From this comparison, the luminescence decay time in the molecules is much shorter than in the QDs. We also find that within the inhomogeneously broadened emission band in both cases the decay time does not vary strongly. For the 4 nm barrier QDM sample the average tunnel splitting is larger than 30 meV and the excitons relax fast into the ground state, so that only emission from this level is detected. The lifetimes are about the same within the luminescence band, showing that the variation of exciton oscillator strength with emission energy is small.

Excitonic luminescence from both tunnel-split exciton states is observed for tunnel splittings smaller than 30 meV because of an acoustic phonon relaxation bottleneck.[9] When addressing arrays, this splitting can be resolved in the inhomogeneously broadened spectra only for the 5 nm barrier sample. For the samples with wider barriers it is too small to be resolved. Also for these samples, no significant variations of the exciton decay time are observed over the range of emission energies. This demonstrates that the lifetimes of the electron-hole complexes are the same in the two states.

The decay is rather smooth for all samples, except for some wiggles around 0.9 ns, where the sensitivity of the streak camera is reduced. Some additional weak wiggles appear in the decay of the molecules, which might reflect variations of the exciton lifetime at a given energy, in particular, for the wide-barrier samples. They do not show any systematic dependence when varying the detection energy (as can be seen from the contour plots in Fig. 1). When averaging over a finite energy range they are smeared out and will not be considered further in the following.

For better comparison, Fig. 2 shows decay curves (the symbols) taken at energies in the center of the emission bands of QDMs with 4, 6, and 8 nm wide barriers and from

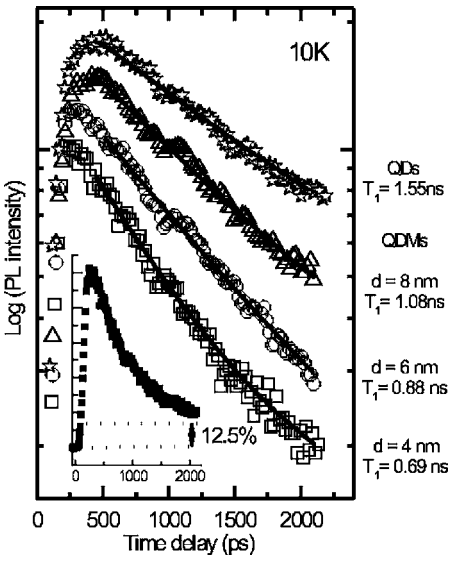

FIG. 2. Photoluminescence decay traces for the QDs reference and the 8, 6, and 4 nm barrier QDMs samples on a logarithmic scale (vertically shifted for clarity), each taken at energies in the center of the inhomogeneously broadened emission peaks. Symbols give experimental data, lines are monoexponential fits (see the text for details). The inset shows the $d=4$ nm data on a linear scale to stress the existence of a background of roughly constant intensity (the dotted line).

the QDs reference as a function of delay time ($T \approx 10$ K). For better visualization the traces have been shifted vertically. The intensity has been normalized and is given on a logarithmic scale. The samples differ slightly in the luminescence rise time (see below), which is short as compared to the decay time, for which the differences are much more pronounced. To determine the decay form, the spectra have to be considered in more detail: after rise of the signal all traces seem to follow an exponential dependence at short delays. For longer delays deviations occur, as can be seen, for example, from the trace for the $d=4$ nm sample, which is shown in the inset of Fig. 2 on a linear scale. Aside from the fast decay, there is a slow component that appears to be almost constant within the time range of interest.

As the optical excitation was done nonresonantly with a linearly polarized laser pulse, we expect a quick depolarization of the photoexcited carriers and, in particular, of the holes, since spin-relaxation times above barrier are rather short.[14] After relaxation spin-bright and spin-dark excitons in the ground state, therefore, contribute to the decay dynamics.[20] The bright states will determine the short decay behavior, while dark states can radiate only after a spin-flip process. Generally, a double exponential behavior is therefore expected. The typical spin-flip time for holes is considerably shorter than for electrons, but should still exceed 10 ns at low temperatures in quantum dots.[15] The dark excitons, therefore, will appear as a background that is almost constant during the times considered here. From the data for the 4 nm barrier sample, we find that this constant background is 12.5% of the maximum signal strength, as obtained from an extrapolation of the data toward long times. When subtracting this background, the luminescence kinetics indeed follows to a good approximation a single exponential decay.

To keep the number of fit parameters minimal, we also assumed the same dark exciton background for the other samples, so that the decay curves were analyzed by a fit function of form: $c_1 \exp(-t/T_1) + c_0$ with $c_0 = 0.125$. The solid lines in Fig. 2 give the resulting fits from which reasonable agreement with the experiment is seen. The decay times T_1 obtained by these fits are indicated at each trace. For the QDs sample a relatively slow decay of 1.55 ± 0.1 ns is observed, which can be understood from the small dot volume given by the dimensions of 20 nm diam and 2 nm height. Because of the three-dimensional confinement, the exciton wave function is extended in momentum space with a width that can be estimated by the inverse dot size. Only the components within the light cone can couple to the light field and decay radiatively. The small dot size leads to a considerable wave function spread beyond the light cone, which results in a long T_1 time. For the molecule samples T_1 is strongly reduced as compared to this value. For the 8 nm barrier QDM sample T_1 is 1.08 ± 0.1 ns, and for the 4 nm barrier sample T_1 is 0.69 ± 0.1 ns, about a factor of 2 smaller than for the QDs.[16]

Before discussing the dependence of T_1 on the barrier thickness in more detail we briefly report on its temperature behavior. Figure 3 shows the evolution of exciton lifetime with increasing temperature from 10 to 160 K for the QDs and the QDMs with a 4 nm barrier. The other QDMs samples have temperature dependencies similar to the $d=4$ nm sample (not shown). In both cases the decay time is about constant up to temperatures of 60 K. This is known as a signature of the quasi-zero-dimensional confinement of excitons whose thermalization in k space is prohibited due to a δ-like density of states,[17] preventing the characteristic linear (square-root) temperature dependence of T_1 in quantum wells (quantum wires) to develop.[18,19] For the QDs the lifetime does not change up to even higher temperatures, while for the molecules a considerable increase is observed, which is probably due to thermal recycling of the carriers, that is, repeated excitation into higher lying molecule states with subsequent relaxation into the ground state. It is not fully clear why it appears for the molecules and not for the dots. It could arise from reduced level splitting, as the energy spacing between the tunnel split states for all QDM barrier widths is smaller than the splitting between the QD p and s shells.

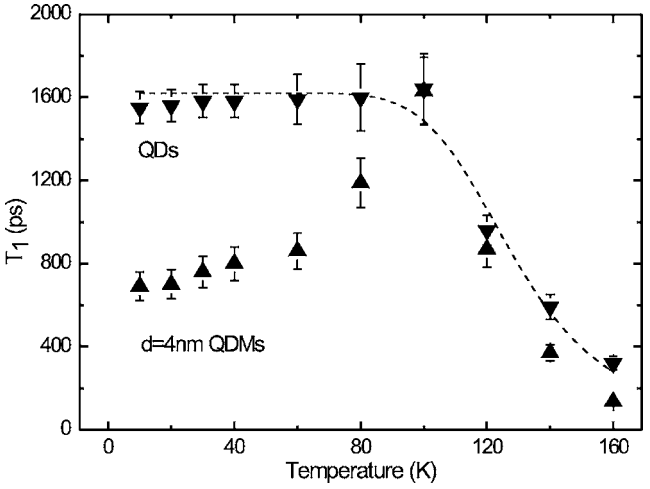

FIG. 3. Temperature dependence of the decay time T_1 from the QDMs sample with a 4 nm wide barrier (the up triangle symbols) and the QDs reference sample (the down triangle symbols). The line is a guide for the eye and follows Eq. (1) given in the text.

We note that the integrated PL intensity does not change in the temperature range up to 100 K, and the optical output is comparable for the QD and the QDM samples. From this observation we derive that nonradiative decay channels are of negligible importance in these samples, so that the population lifetime can be assumed to be equal to the radiative lifetime.

At higher temperatures above 100 K a strong decrease of T_1 occurs, which is accompanied by a quenching of photoluminescence. We ascribe this regime to thermal emission of carriers out of the dots. Taking into account a constant radiative recombination rate $1/T_{\mathrm{rad}}$ (equal to $1/T_1$ at low T) and a thermal emission rate $\exp(-E_A/kT)/T_{\mathrm{esc}}$ where E_A and T_{esc} are the activation energy and the effective escape time, respectively, one can fit the QD data according to[21]

$$T_1 = \frac{T_{\mathrm{rad}}}{1 + \frac{T_{\mathrm{rad}}}{T_{\mathrm{esc}}} \exp\left(-\frac{E_A}{kT}\right)}, \tag{1}$$

A value $E_A = 95 \pm 10$ meV is obtained which is close to half the barrier height between the molecule ground states and the

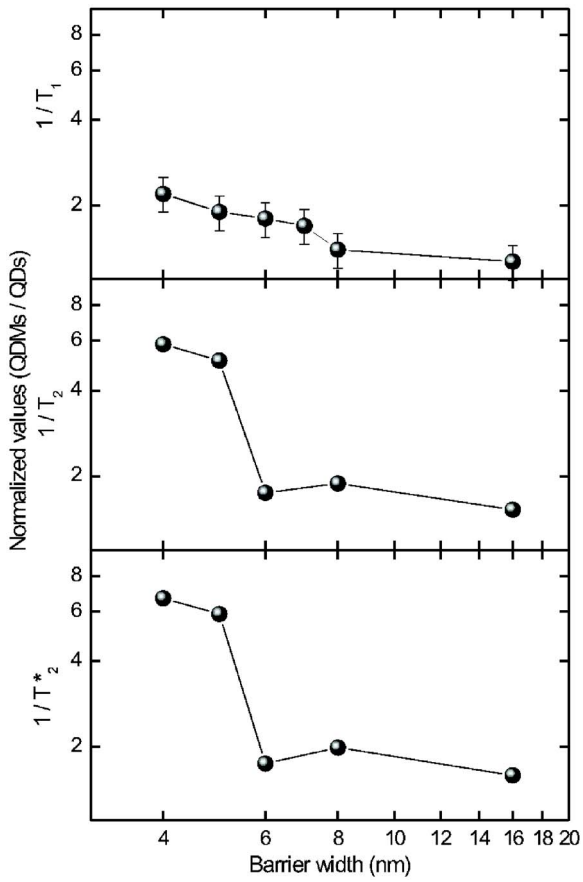

FIG. 4. (Color online) Dependence of different decay rates on the QDM barrier width normalized by the corresponding rate of the QDs reference (plotted on a log-log scale): The upper panel gives the exciton population decay rate $1/T_1$ (error bars include the errors when evaluating T_1 in QDMs and QDs), and the middle panel gives the zero-temperature exciton dephasing rate $1/T_2$ for $T \rightarrow 0$, from Ref. 10. From these quantities the pure dephasing rate $1/T_2^*$ is calculated according to Eq. (2) and shown in the lower panel. For comparison, all the rates are shown on the same vertical scale.

GaAs barrier. Such a result is expected for electron-hole pairs from Boltzmann statistics assuming thermal equilibrium between dots and surrounding.[21]

The upper panel in Fig. 4 shows the PL decay rates at low T as function of the molecule barrier width. The values displayed are normalized by the corresponding rate of the QDs reference. For all molecule samples, also for the wide barrier cases, T_1^{-1} is considerably larger than for the dot structures. The enhancement is increasing with decreasing barrier width. The radiative lifetime is inversely proportional to the oscillator strength of an exciton, which is determined by the overlap of the electron and hole wave functions and by its coherence volume. In QDs the coherence volume is closely related to the geometrical dot size.[22] Simply speaking, the exciton collects oscillator strength from every crystal unit cell into which its wave function spreads.

To understand the increase of the decay rate in the molecules, we have to consider how quantum coherent coupling of two QDs affects the oscillator strength. This problem was addressed theoretically by Bryant[23] for the case of laterally confined excitons placed in symmetrically coupled quantum wells: The ground state of an exciton in a QDM is a coherent superposition of excitons in both QDs with twice the oscillator strength of one exciton in a single QD. As a consequence we expect a reduction (an increase) of the exciton lifetime (the decay rate) by a factor of 2. The experimental value for the decay rate of 2.2 ± 0.3 obtained for the QDMs with the narrowest barrier of 4 nm agrees with this prediction, giving additional proof for an exciton wave function that is extended over the molecular structure, for which the two QDs must be coherently coupled.[24] Different penetration of electron and hole wave functions into the barrier separating the dots might lead to a reduction of oscillator strength, which is, however, expected to be considerably smaller than the effect from doubling the coherence volume.[25]

Based on these arguments, however, we would expect a decay rate increase independent of barrier width, provided coherent coupling is established in a QDM. Instead a rather smooth increase with decreasing barrier width is observed, starting from the 8 nm sample with a ratio of about 1.4. Previous studies indicate that coupling is established also for the wide barrier molecules.[7,9] Here two points have to be noted: (i) the measured decay times represent averaged values obtained from ensemble studies; (ii) the doubling of oscillator strength is expected for the case of symmetric molecules only.

The reduced decay rate value most probably arises from deviations from ideal molecule symmetry resulting in variations of the interdot coupling: Asymmetries arise from differences of the two dots that form the molecule regarding geometry and composition or from displacement of the dots relative to each other. Such asymmetries were identified as origin of the complicated exciton fine structure patterns reported in Ref. 7 on similar QDMs. From these studies we find a considerable variation of the molecule coupling in the samples with wide barriers of 7 and 8 nm. Even though it is difficult to develop a criterion for a statistical overview, the variety of fine-structure magnetic-field dispersions obtained on many single QDMs suggests that about 40% of the molecules show asymmetries that are so weak that the fine structure is identical to the one of an ideal molecule. For another 40% of the structures the asymmetries result in anticrossings in the field dispersion, demonstrating quantum-mechanical coupling of reduced strength. For the final fraction of molecules, the fine structure data do not permit us to make a clear assignment as to whether the dots are coupled or not, but at least the coupling is strongly reduced. When decreasing the barrier width, the effects of asymmetries become less and less important, as is expected from the increased tunnel splittings. The fine structure data for the 4 and 5 nm barrier samples suggest that all of the molecules are strongly coupled.

Let us now discuss the effect of these variations of molecule coupling on the exciton oscillator strength. The case of ideal symmetry has already been discussed above.[23] Any symmetry reduction will also reduce the oscillator strength. To give two examples: the tunnel coupling is basically suppressed, when the dot asymmetry is bigger then the energy gain through formation of a bonding orbital. In this case of two decoupled dots, one expects the same exciton decay rate

as for the QDs reference. If, on the other hand, the electron states are tunnel coupled but the holes are localized because of smaller tunnel matrix elements due to larger mass, the overlap of the carrier wave functions is strongly reduced leading to exciton decay rates even smaller than in the QD case.

Summarizing these discussions, the barrier width dependence of the decay rate becomes understandable. For the strongly coupled systems with 4 and 5 nm barrier, the effects of asymmetry are negligible and a lifetime reduction by a factor of about 2 is expected, in reasonable agreement with experiment. The decay rate measured for the wide barrier samples represents a statistical average to which "ideal" molecular structures contribute with an increase of T_1^{-1} by a factor of 2 as well as structures with reduced coupling with a T_1^{-1} below 2, that could be even smaller than the value for the QDs. Nevertheless, the fact that the decay rate is roughly a factor 1.4 larger than for the QDs even for the widest barrier sample, shows that most of the dot pairs are molecules. The smooth increase of T_1^{-1} reflects the decreasing importance of asymmetries on the molecule coupling, i.e., an increasing fraction of structures in the array resembles ideal molecules, as expected from the increased coupling strength.

The effect of asymmetries has to some extent been simulated in the calculations of Bryant,[23] who studied the exciton coherence in coupled dots also under the influence of an electric field along the molecule axis. Application of a bias leads to carrier localization in one of the dots and, thus, has similar consequences as a geometry induced asymmetry of the molecules.[27] The calculations show that the oscillator strength resists weak bias changes due to the electron-hole Coulomb correlations but for fields large enough to induce carrier redistribution, there is a strong drop in oscillator strength, in agreement with our qualitative arguments.

Now let us discuss, how the population decay rates T_1^{-1} compare to the exciton dephasing rates T_2^{-1} measured in Ref. 10 at 10 K. These rates are connected through

$$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{T_2^*}, \tag{2}$$

where $(T_2^*)^{-1}$ is the pure dephasing rate without change of state population, for example, due to relaxation between fine structure states, virtual transitions involving phonons, etc. The middle panel of Fig. 4 gives the barrier width dependence of T_2^{-1} for the QDMs. Given are the T_2^{-1} that are obtained from extrapolation of the four-wave-mixing data to zero temperature and are about constant below 20 K.[10] Again the data are normalized by the corresponding rate of the QDs reference. As mentioned, it has been shown that for QDs the dephasing is ultimately limited by the radiative decay of the excitons.[11]

For the QDs reference we obtained a radiative decay time of $\approx$1.6 ns, from which a dephasing time of more than 3 ns would be expected in absence of pure dephasing. However, the experimentally measured dephasing time is $\approx$600 ps only, showing that the dephasing is not radiatively limited for the QDs under study, but pure dephasing plays an important role. The origin of this pure dephasing cannot be clearly assessed from the present experiments. Similar observations had already been made on other QD samples.[28] In general, the width of the zero-phonon line at low temperatures that results from T_2^{-1} is determined by virtual phonon transitions (assuming negligible phonon linewidths),[29] whereas real phonon-assisted transitions can be neglected due to a confined level splitting that is large as compared to the thermal energy k_BT. These virtual transitions might vary with the dots under study and might provide an explanation for the difference between coherence and population decay rates in some QD samples. Also a finite phonon linewidth might become relevant for explaining the pure dephasing (see below).

Similar to the population decay rate, also for the dephasing rate an increase with decreasing barrier width has been observed in the molecules. However, this increase is much stronger than for the QDs. For the narrow barrier sample it reaches a factor of almost 6, as compared to the doubling of T_1^{-1}. Hence the difference between coherence and population decay rates becomes even bigger for the molecules as compared to the dots. This can be seen from the bottom panel of Fig. 4, which gives the pure dephasing rates normalized by the corresponding QDs reference value. Vice versa, the exciton superradiance in the molecules only contributes partly to the observed enhanced dephasing. Mechanisms for pure dephasing also become more important in the molecules. For example, the cross section for virtual scattering processes might become larger. Furthermore, one can imagine that also real phonon-assisted transitions play a role.[33] In the molecules, the level splitting is reduced due to the tunnel splitting of each state. This splitting is smaller than the QD state splitting for all barrier widths. In particular for the hole, small splittings are expected, so that real transitions might occur even at low T.

An alternate explanation might be provided by the recent observation of a finite zero-phonon linewidth that depends on the dot surrounding. In single QD luminescence on laterally patterned samples it has been observed that the enhancement of linewidth with temperature is increased with decreasing size of the mesastructures in which the dots were isolated for spectroscopy. By detailed comparison to calculations, this dependence was traced to exciton scattering with phonons, whose linewidth is determined by the lateral surface roughness.[30] This result underlines the importance of interfaces in the surrounding of the QDs on the exciton dephasing. As compared to dot structures, the QDMs are subject to further interfaces due to the implemented barrier. Certainly these interfaces will have roughness, due to which the phonon linewidth might be increased as compared to the QD case.[31] With decreasing barrier width this factor could become increasingly important due to penetration of carriers into the barrier.

Indications that the tunnel splitting considerably alters the carrier-phonon dynamics are also found in the rise of the photoluminescence signal. The rise time is $\approx$200 ps for the QDs. A rise time of comparable length is observed for the wide barrier molecules, whereas for the narrow barrier QDMs it is significantly faster ($<$120 ps). The rise is determined by the relaxation of carriers from above the GaAs barrier into the ground states of the confined geometries. For excitation into the wetting layer, basically the same times

were observed, so that the rise is obviously dominated by the relaxation within the confined geometries. In the wide barrier samples, the tunnel-induced splitting is quite small ($\approx$10 meV), so that the different shells are still energetically well separated. Therefore no large modification of relaxation is expected as compared to the dots. For the narrow barrier samples, the tunnel-induced splitting becomes of the same order of magnitude as the shell splitting. Thus the discrete QD spectrum is transformed by the molecule formation into a spectrum where the states lie more densely on the energy axis, potentially facilitating carrier relaxation.

In conclusion, we have demonstrated exciton superradiance induced by coherent tunnel coupling in self-assembled InAs/GaAs QDMs and have clarified its relation to exciton dephasing. To obtain more detailed insight it would be interesting to study two-dot molecules, in which the carrier distribution can be controlled on a detailed level by application of an electric field along the molecule axis.[27] It will be also interesting to see whether more than two-dot structures can be coherently coupled to obtain a further enhancement of the exciton oscillator strength. By placing such an arrangement into a high-finesse resonator, the regime of strong coupling of exciton and photon with a sizeable Rabi-splitting among the hybridized states might be entered.[32]

Note added in proof. We note that recently a theoretical work on phonon induced dephasing of excitons in QDMs has been reported.

ACKNOWLEDGMENTS

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the oscillator strength from all crystal unit cells, into which it spreads (see text), also the barrier might contribute to the oscillator strength, so that it is enhanced by more than a factor of 2 through the molecule formation.

[25] Studies of the Stark shift of excitons with a bias applied along the growth axis show that in dome-shaped QDs the electron-hole overlap is considerably reduced. For the In-flushed QDs with heights of $\approx$2 nm, we do not expect a strong displacement of electron and hole. How this overlap is changed by the molecule formation is not fully clear at the moment and requires further studies.[26]

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