Multiexciton transients in a single quantum dot

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The relaxation dynamics of a multiple exciton complex (multiexciton) confined in a semiconductor quantum dot has been investigated. Emission signals from a single self-organized GaAs/Al0.3Ga0.7As quantum dot are temporally resolved with picosecond time resolution. The emission spectra consisting of the multiexciton structures are observed to depend on the delay time and the excitation intensity. Quantitative agreement is found between the experimental data and the calculation based on a model describing the successive relaxation of multiexcitons.

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1. Introduction

Semiconductor quantum dots (QDs) are promising candidates for the fundamental units for realizing quantum information processing [1]. One efficient approach for the implementation of quantum operations is to exploit the optical excitation (exciton) in QDs. Excitonic Rabi oscillations have been demonstrated in single QDs [2–4], QD-based photodiodes [5], and ensembles of QDs [6], showing that the exciton state is an ideal two-level system, and single quantum bit (qubit) operations are realized. Observation of the long dephasing time which exceeds several hundred picoseconds [7–9] supports the possibility of sufficient numbers of quantum manipulations using ultrashort optical pulses. Practical quantum computations also require the preparation of a large number of the qubits which consist of interacting two-level systems. Spatially coupled QDs (or an array of QDs) ideally meet this requirement [10], although creation of such

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structures with sufficiently high definition is currently not achievable even with a state-of-the-art fabrication technique. Another efficient approach to realize multiple qubit operations is to exploit the multiexciton complex confined in a QD [11]: a QD can contain several carriers (up to 6–10 in prototype III–V QDs), and these carriers form a new correlated state, known as a multiexciton. In this framework, each excitonic qubit is individually accessible by using the polarization and energy degrees of freedom of light, and entanglement of these states relies on the Coulomb correlation between carriers [11, 12].

Multiexciton effects have been explored by means of a single QD (SQD) photoluminescence (PL) technique [13–18], where a variety of emission lines are observed with increasing photoinjected carriers via few-particle effects. Theoretical investigations based on an effective mass model [16, 19], a Monte Carlo approach [20], and a pseudo-potential one [21] show that these calculations successfully reproduce the multiexciton spectra. Although formation of multiexcitons is now an established issue, only a few attempts have been devoted to study the relaxation kinetics of multiexcitons [22, 23]. For implementation of multi-qubit processing within a SQD, a clear understanding of the multiexciton transients is obviously required. In this contribution we report on ultrafast nonlinear carrier relaxation inside a SQD [24]. The multiexcitonic emissions are resolved with picosecond time resolution and interpreted in terms of the successive transition between the correlated few-exciton states. Quantitative analysis on the multiexcitonic emission allows us to determine the temporal dependence of the carrier population confined in a single dot. This permits us to show a strict correlation between the rise-time characteristics of the SQD emission and the number of photoinjected carriers. We demonstrate that the intra-dot carrier–carrier scattering process is responsible for the efficient carrier relaxation in semiconductor QDs.

2. Experimental

We focus on GaAs/Al_{0.3}Ga_{0.7}As self-assembled QDs grown by modified droplet epitaxy (MDE) [25, 26]. MDE is a non-conventional growth method for self-assembling QDs even in lattice-matched systems [26]. By modifying the surface reconstruction and the adatom mobility it is possible to obtain QD samples with a density as low as \( \approx 6 \times 10^8 \) cm\(^{-2}\). Surface and cross sectional high-resolution scanning electron microscope images demonstrate the formation of pyramidal shaped nanocrystals of 16 nm height and 20 nm base [27].

The optical measurement was performed with a fs mode-locked Ti-sapphire laser of 76 MHz repetition rate. A second-harmonic beam of the laser output (\( \lambda = 400 \) nm) was used for excitation. This beam was loosely focused on the sample by a conventional lens of \( f = 200 \) mm. The emitted signal was collected by a microscope objective of N.A. = 0.5. The present configuration allows one to irradiate the sample homogeneously inside the detection spot, and to determine the power density with precise accuracy. The spatial resolution was \( \sim 1.2 \) \( \mu \)m, causing \( \sim 7 \) QDs to be collected on average. The signal was dispersed by a polychromator, and detected by a synchronously scanning streak camera. The temporal and spectral resolutions were 15 ps and 0.8 meV, respectively. Emission spectra with stationary excitation were also observed in the initial stage of the experiments, adopting a continuous wave (cw) \( \text{Ar}^+ \) laser for excitation. The sample was attached to a cold finger of a He-flow cryostat. All experiments were performed at 3.5 K.

3. Results and discussions

The emission spectra obtained with conventional lens optics after stationary (cw) excitation are presented in Fig. 1A. The PL bands at \( \sim 1.93 \) and \( \sim 1.96 \) eV arise from an Al\(_{0.3}\)Ga\(_{0.7}\)As barrier layer. The doublet structure of the barrier emission is induced by impurity-related effects. A broad emission band is found at the lower-energy side of the barrier PL, corresponding to the emission from ensembles of QDs. Large spectral broadening reflects the inhomogeneous distribution of the size and shape of the QDs. The energy of the emission band agrees with a result of effective mass calculation, applied to the relevant sample structure [28].
Fig. 1. A, Stationary emission spectra using conventional optics. B, MicroPL spectra with the same experimental conditions.

Note that the observed emission corresponds to the grand-state transition, i.e. recombination of the electron and the hole both occupied in the lowest-energy confinement state (s-shell state). Excited-state emission (p-shell transition) will be observed to be around 80 meV higher than the ground state emission, appearing when the sample is irradiated more strongly [28, 29]. The microPL spectrum at weak cw excitation is shown in Fig. 1B. The broad emission band seen in the ‘macro’ PL spectra changes into the sharp split lines, which originates from isolated QDs. The number of emission lines (6 ∼ 10) agrees with the estimated number of QDs inside the detection area with a micro objective lens.

The dependence of the SQD emission on excitation intensity is exhibited in Fig. 2. At weak excitation, a single line due to recombination of a single exciton (denoted by 1X) is solely observed. With increasing excitation, a new emission line, 2X, appears at a lower energy of 1X, reflecting the formation of a two-exciton complex (biexciton). The relevant emission corresponds to the transition of a biexciton (two electrons and two holes bound in the s-shell) leaving a single exciton as a final state. The energy split between 1X and 2X determines the biexciton binding energy to be 0.8 meV, which is relatively small compared to that of the bulk GaAs (∼1.5 meV). Reduction of the biexciton binding energy in the QD structure is explained in terms of the penetration of electron wavefunctions outside a QD [20]. At stronger excitation, other emission lines (3X, 4X, . . .) are observed at several meV lower than 1X and 2X. In this case, more than three carriers are injected into a dot, so that the p shell state begins to be occupied. Transitions take place on the annihilation of the electron–hole pairs occupying the s-shell and p-shell states, while the p-shell related emission is out of our spectral range. (The inter-shell transitions such as the recombination of the s-shell electron and the p-shell hole are forbidden in QDs with cylindrical symmetry.) The correlation energy for multiexcitons is roughly given by the summation of the two-particle exchange energies. As a result, multiexciton lines shift to lower energy with increasing population of the confined carriers. Such a strongly excited behavior is similar to the bandgap renormalization effect seen in bulk semiconductors.
Typical transient microPL spectra are shown in linear color scale in Fig. 3. Similarly to the cw data, the spectra consist of split PL bands, which originate from emissions of individual QDs. It is evidently found that each of the PL bands have a finite linewidth which is larger than the spectral resolution of our apparatus. In addition, the emission does not indicate a monotonic decay, and the spectral feature (width and energy) is seen to change with time. Such significant behavior is evidence of the multiexciton relaxation involved in photoinjected QDs.

Figure 4 shows a series of time-resolved SQD emission at various excitation powers, $P_{\text{exc}}$. These emissions arise from the ground state recombination of an isolated SQD. As is clearly shown, a highly nonlinear behavior and peculiar temporal dynamics appear on the SQD emission. At low excitation, $P_{\text{exc}} = 47 \text{ nJ cm}^{-2}$, Fig. 4A exhibits a narrow line, defined as A, and a weak band, defined as B, at the low-energy side of A. Both components decrease monotonically with time, although the B band decays much faster than the A line. With increasing excitation power to $150 \text{ nJ cm}^{-2}$ (Fig. 4B), the B band appears as strong as A, and an additional rapid-decay component, defined as C, emerges at lower energy. Simultaneously, the temporal shape of A tends to be stretched, and the finite rise profile is evidently resolved. After nearly 500 ps the spectral evolution is almost identical to that of the lower excitation case. On increasing the irradiation, as in Fig. 4C, the broad C band dominates the transient stage of emission, and the rise of A and B is found to be slower. It is worth noting that the present features are common to all the SQD emissions, independent of their energy or dot size.

The temporal behavior of SQD emission is characterized by the transient PL spectra reported in Fig. 5A. We selected four spectra from the PL trace at $P_{\text{exc}} = 150 \text{ nJ cm}^{-2}$, indicated by the white arrows in Fig. 4B. The spectra show that: (i) the broad C band together with the B band dominates the early stage of the emission; (ii) the C band rapidly fades away with time; and finally, (iii) in the long-time limit, the B band
Fig. 3. Typical time-resolved microPL signals in GaAs QDs at 150 nJ cm\(^{-2}\) pulse excitation.

Fig. 4. Temporally and spectrally resolved images of a SQD emission at various excitation power densities: \(P_{\text{exc}} = 47\) nJ cm\(^{-2}\) (panel A), 150 nJ cm\(^{-2}\) (panel B), and 470 nJ cm\(^{-2}\) (panel C). Three emission components characteristic to the SQD spectra are denoted by A, B, and C.
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Fig. 5. Panel A: transient SQD emission spectra at $P_{\text{exc}} = 150 \text{ nJ cm}^{-2}$ for various delay times, which are presented by the white arrows in Fig. 4B. Panel B: high-resolution SQD spectra with cw excitation at 2 kW cm$^{-2}$ (solid line) and 70 W cm$^{-2}$ (dotted line). Panels C and D: temporal evolution of the SQD emission bands of A, B, and C, at $P_{\text{exc}} = 47$ and 150 nJ cm$^{-2}$, respectively. Spectral windows for the signal integration are shown in A.

is totally replaced by the A line. Temporal development of these emission bands are reported in Fig. 5C and D for two different excitation powers. In the transient spectra, we also resolve a red shift of the A line ($\sim 0.8 \text{ meV}$, presented by vertical lines in Fig. 5A and a narrowing of the B band with increasing time. These observations suggest that A and B originate from multiple components which are not spectrally resolved. This speculation is supported by the high-resolution SQD spectra with cw excitation, reported in Fig. 5B. In fact, the high-resolution spectra exhibit two lines split by 0.8 meV in the A-line region, while four lines characterize the B-band region. When the irradiation is weak enough, the highest-energy line appears alone, and with increasing irradiation, other lower-energy lines take place. Note that a very similar spectral profile is obtained for Fig. 5A and B, although a different QD was captured.

The transient emissions are therefore characterized by three components: the long lasting, high energy A line at 1828 meV, the B line at 1823 meV, and the fast and broad C band whose energy depends on the excitation power and delay time. This peculiar evolution can be attributed to multieexcitonic effects. The presence of more than one exciton in the QD determines the splitting and the red shift of the emission energy of the QD ground state transition. In fact, the carrier exchange interaction leads to a progressive decrease of emission energy when increasing the number of excitons in the QD, although a certain reduction is expected due to correlation effects [21]. Thus, when the number of excitons increases, more lines appear in the low-energy side of the fundamental single exciton transition. It is worth noting that all the emissions originate from the recombination of an electron (and a hole) confined in the same lowest QD state, while their energy is modified by the carrier–carrier correlation. Within this framework we attribute the doubly degenerate A line to single exciton and biexciton recombinations, and the four-fold degenerate B band to the recombinations when the total number of excitons is between three and six. A larger number of excitons captured in the QD give rise to the C band. These attributions reproduce the power dependence of the SQD emissions well, as will be discussed later.
To perform a quantitative discussion, we analyze the time evolution of the SQD PL using a model based on the successive transitions between the multiexciton states. Since our observations suggest that the transient SQD spectrum is solely determined by the population of excitons at a specific time, the emission dynamics are described by a set of rate equations [22],

$$\frac{d\rho_i}{dt} = -\Gamma_i \rho_i + \Gamma_{i+1} \rho_{i+1},$$  \hspace{1cm} (1)

where $\rho_i(t)$ is the probability of finding $i$ excitons inside the QD, and $\Gamma_i$ is the $iX$ transition rate—hereafter $iX$ indicates the recombination with $i$ excitons inside the QD. We also adopt the realistic assumption that the number of photoinjected carriers are statistically distributed, and it is given by the Poisson distribution, so that

$$\rho_i(t = 0) = e^{-\bar{\rho} \frac{\rho_i}{i!}},$$  \hspace{1cm} (2)

where $\bar{\rho}$ represents the average number of photoinjected carriers [18].

The efficiency of photoinjection was extracted by analyzing the time-integrated (TI) intensities of the SQD emissions. Variations of the TI intensities for the A-, B-, and C-emission bands are presented in Fig. 6A, together with the calculations based on the above model. The best fit to the data gives an efficient carrier-capturing cross section, $\xi$, of $1.85 \times 10^{-11}$ cm$^2$. With the use of $\xi$, the number of photoinjected carriers is given by the multiplication of $\xi$ with the photon flux, or the carrier density at the sample surface. The present procedure allows us to determine the carrier population inside the QD. It was found that the magnitude of $\xi$ is consistent with a result of the PL yield measurement of the present sample, and of the same order with that of the InAs/GaAs QDs reported in Ref. [22].

To interpret the temporal evolution of the SQD emission, $\rho_i(t)$ is solved numerically. For simplicity we assume that the transition rate of the multiexciton state is given by the summation of the relevant
single-carrier recombination rates. In this treatment, dynamics of up to 10 excitons are specified using the three lowest inter-shell (s-, p-, and another higher-shell) transition rates as free parameters. In Fig. 6B and C, the best fits of the data Fig. 5C and D are presented, where the transition times for the s-, p-, and the higher energy shell are derived as 400, 600, and 300 ps, respectively, by an accuracy of 20%. Excellent agreement between calculation and the data supports the validity of the assumption that the multiexciton transition strength, i.e. overlapping between the few electron–hole pairs, is not strongly modified by the carrier–carrier correlation.

4. Conclusions

We have determined the nonlinear carrier dynamics in a SQD by means of PL measurements with picosecond time resolution. Effects of the multiexciton dynamics leads to strong nonlinearities in the SQD emission. The observed behavior stems from successive relaxation dynamics of multiexcitons. Moreover, the efficient intra-dot relaxation has been shown to be due to the nonlinear interaction between carriers inside a QD. We believe that the investigation of transient SQD emissions allows us to obtain a deeper understanding of the ultrafast carrier dynamics in zero-dimensional semiconductor systems.

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