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Transitions from spontaneous emission to stimulated emission and superfluorescence of biexcitons confined in CuCl quantum dots

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ABSTRACT

Time-resolved photoluminescence spectra of biexcitons in CuCl quantum dots embedded in NaCl matrix have been measured by two configurations of optical Kerr gate method. As for a backward configuration, a transition from spontaneous to stimulated emission is observed with increase in the excitation density. On the other hand, by a configuration collecting signals from the edge of the sample in the direction of the excitation length, an essential change in the time profile of biexciton luminescence from a conventionally exponential to a pulse-shaped according to the increasing excitation density reasonably suggests a shift from spontaneous emission to superfluorescence of biexciton luminescence.

1. Introduction

Biexcitons play an important role in nonlinear optical properties of semiconductor materials, especially in nanostructures. Intensive attentions have been paid during two recent decades to examine basic properties as well as relating phenomena of biexcitons confined in nanostructures [1–3]. In a material with an appropriate biexcitonic binding energy, which is even significantly enhanced in low-dimensional structures due to spatial confinement effect, resonant two-photon excitation is a powerful tool to reveal biexciton properties in fundamental research. For an ensemble of quantum dots (QDs) with inhomogeneous broadening due to size distribution, resonant two-photon excitation using an excitation light with narrow spectral width corresponds to size-selective excitation of the biexcitons, which can unveil the size-dependent biexciton dynamics. In addition, the resonant two-photon excitation of the biexcitons for the ensemble of the QDs induces a complete population inversion between the biexciton and exciton levels. This population system originates lasing emission of the biexcitons [4,5] and superfluorescence (SF) induced from coherent coupling among many excited two-level systems [6,7]. In this literature, we report transitions from spontaneous emission to stimulated emission and superfluorescence of the biexcitons confined in CuCl QDs in NaCl matrix. Two types of the transition can be distinguished by a change in time profiles with the increasing excitation density. Stimulated emission shows a quite fast decay profile but SF exhibits a pulse-shaped profile with a delay time, which is related with a formation of macroscopic dipole moments.

2. Experimental details

Two samples of CuCl QDs embedded in NaCl matrix with average sizes of 3 nm and 5.5 nm were fabricated by the transverse Bridgman method [8] and maintained at 4 K and 8 K, respectively, in a cryostat. Time-resolved photoluminescence (PL) measurements were performed by means of the optical Kerr gate method based on a regenerative-amplified pulse laser system (an output wavelength of 800 nm, a pulse width of ~2 ps and a repetition of 1 kHz). The amplified light was divided into two beams; one was used as a gate light irradiated onto the Kerr medium, while the other was utilized as the pump light for an optical parametric amplifier. The excitation light obtained by a fourth harmonic generation of the signal beam from the optical parametric amplifier was tuned to a photon energy corresponding to resonant two-photon excitation of biexcitons. For a backward configuration using the 3 nm sample, the excitation light of 3.188 eV was focused onto the sample with a spot size of 100 μm. For the configuration collecting PL from the edge of the sample in the direction of the excitation length, the excitation light of 3.195 eV was irradiated

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onto the surface of the sample with a stripe of $700 \times 30\ \mu m^2$. The PL signal was focused on an optical fiber connected to a 50 cm spectrometer, which is equipped with a liquid nitrogen-cooled CCD array. In both the experiments, toluene was used as the Kerr medium, and the time resolution of the experimental system was approximated to be 2.4 ps.

3. Results and discussion

3.1. Transition from spontaneous to stimulated emissions of biexciton luminescence

A typical time-integrated PL spectrum of biexciton luminescence from CuCl QDs measured under the resonant two-photon excitation is shown in Fig. 1. Two biexciton luminescence bands located at ~3.16 and ~3.17 eV are denoted as M and BM, respectively. In the previous report, the M band was suggested to relate to the free biexcitons, while the BM to the bound biexcitons since the BM band becomes relatively weak compared to the M band with the increasing temperature [9]. However, the photon energy of M band is not equal to the absorption energy from the exciton to biexciton state [10,11]. Therefore, we consider that the M band is also a kind of bound state, such as a surface-bound state inherent in the quantum dots. Because the observed characteristics of the BM band are almost the same as those of the M band, discussions of the BM band will be omitted in the following. The excitation density dependence of intensity of the M band is displayed in the inset of Fig. 1. The intensity shows a superlinear dependence on the excitation density with the power factors of 1.9 as long as the excitation density is smaller than $\sim 2.5\ mJ\ cm^{-2}$. In the high excitation density region, meanwhile, the dependent power factor increases dramatically up to a value of 8.8. Such a kind of the behavior is usually caused by an amplification process. As the number of excited dots is large enough, which should be fulfilled under a high excitation density, and one-round propagation time of the emission light through the excited volume is fairly shorter than the lifetime of the biexcitons in QDs, a luminescence signal from a dot experiences an amplification process during the propagation through the excited volume. As a result, not only a great enhancement in the PL intensity, but also a significant increase in the decay rate is expected. Therefore, an additional examination on the excitation density dependence of the decay time is needed for more reliable confirmation of existence of an amplification process in the high excitation density region.

The time profiles of the M band, which basically consist of one rise and one decay component, at two different excitation densities are shown in Fig. 2a. The rise and decay times of the M band then are extracted by a decovolution analysis and then depicted as a function of the excitation density in Fig. 2b. The decay time is almost unchanged, estimated to be about 80 ps, provided that the excitation density is smaller than $2.5\ mJ\ cm^{-2}$. In the high excitation density region, the decay time exhibits a decrease with the increasing excitation density. This tendency in the high excitation density region is consistent with expectations subsequently resulting from

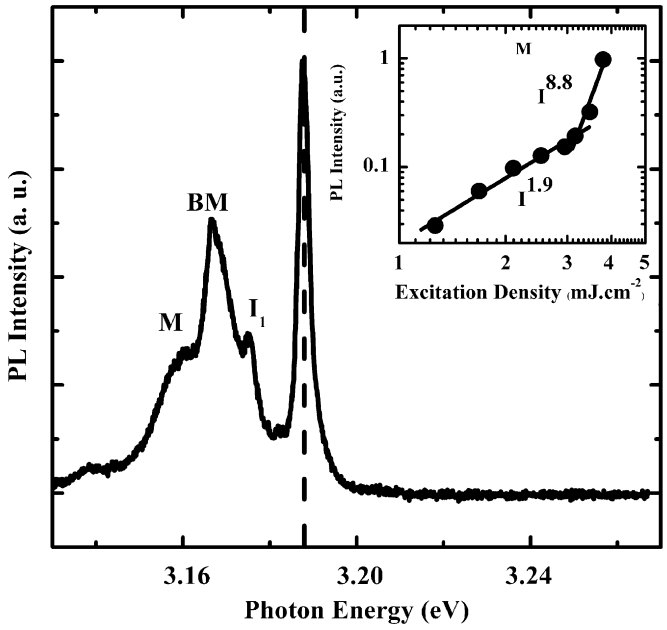

Fig. 1. A typical time-integrated PL spectrum of the biexciton luminescene under the resonant two-photon excitation of 3.188 eV (dash line). The I_1 band is associated with an exciton bound to a neutral donor. The inset shows the excitation density dependence of the PL intensity of the M band (solid circles) obtained with a backward configuration. The solid lines are the fitting curves with different power factors in the different excitation density regions.

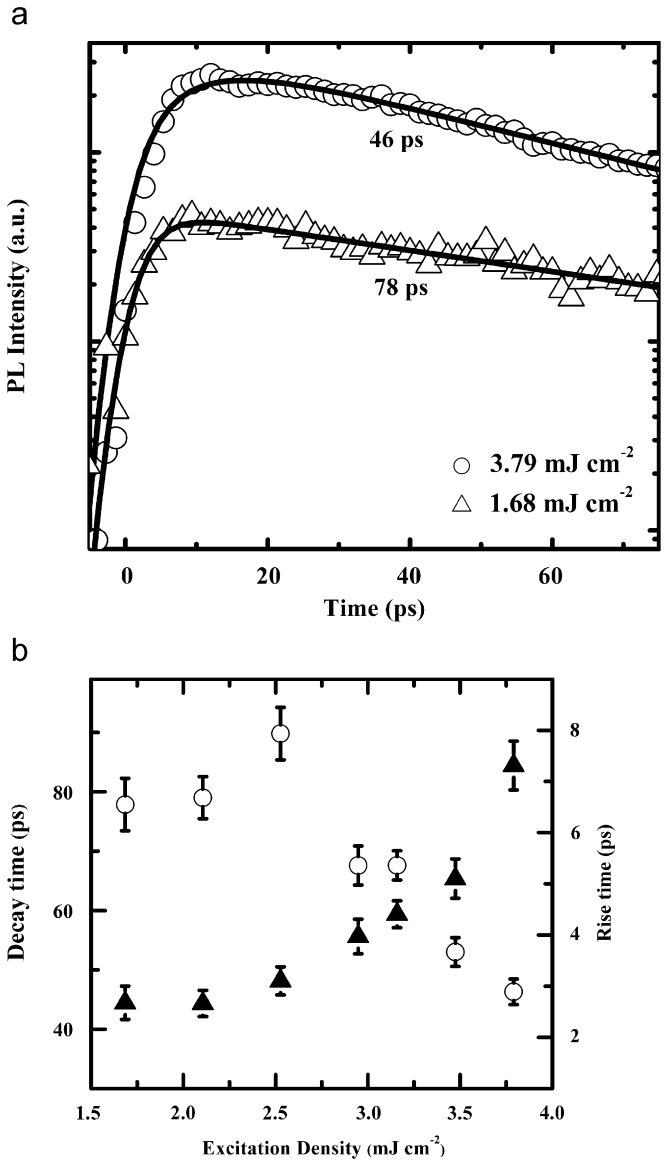

Fig. 2. (a) The time profiles of the M band at two different excitation densities (open circles and triangles). The solid lines are the convoluted fitting curves basically consisting of one rise and one decay component with the time profile of the scattered excitation pulse. (b) The decay time (open circles) and rise time (solid angles) as a function of the excitation density with the excitation photon energy of 3.188 eV.