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Superfluorescent pulsed emission from biexcitons in an ensemble of semiconductor quantum dots

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Abstract
Picosecond time-resolved photoluminescence from biexcitons in CuCl quantum dots (QDs) embedded in a NaCl matrix has been measured using an optical Kerr gate method. Ultrafast pulsed emission from the biexciton states was observed for the first time, only under resonant two-photon excitation of biexcitons. This implies that complete population inversion between the biexciton and exciton states is necessary in order to trigger the pulsed emission. In addition, the nature of the dependence of the time profiles of the pulsed emission on the excitation intensity reveals that the peak intensity is directly proportional to the square of the number of excited QDs. We conclude that this phenomenon is caused by superfluorescence, that is, the cooperative spontaneous radiative decay of many isolated excited states coupled by a resonant electromagnetic wave. Such a phenomenon has been observed for the first time in an ensemble of semiconductor QDs in this study. The results presented in this paper show that it is possible to control the microscopic coherent dynamics of electronic excited states in a QD ensemble.

1. Introduction

In 1954, Dicke theoretically predicted cooperative spontaneous emission from an ensemble of N isolated two-level systems when the distance between any given pair of two-level systems is shorter than the emission wavelength [1]. This optical phenomenon is called superradiance. When all the N two-level systems are in the excited state, that is, when complete population inversion occurs, the peak intensity and width of the time profile of the superradiance pulse are proportional to N^2 and $1/N$, respectively [1–3]. Thus far, most studies on pulsed emissions such as the one mentioned above have focused on a particular phenomenon known as superfluorescence [3]. Superfluorescence occurs due to self-induced synchronization among isolated transition dipole moments through a fluorescence radiation field [4]. Superfluorescence has been reported mainly for atoms and molecules [5, 6], and those embedded in the solid state [7–10]. On the other hand, Dicke's theory predicts

different phenomena under low-excitation conditions—when one of the N two-level systems is excited, the transition rate of systems during superradiance is enhanced to N times that observed for an isolated two-level system [1]. The following theory has primarily been applied to study 'exciton superradiance' in condensed matter such as semiconductor quantum wells [11, 12], quantum dots (QDs) [13] and J-aggregates [14]: the transition rate of an exciton (a bound electron–hole pair) is enhanced by the accumulation of the dipole moments of the atoms present in the volume where coherence of center-of-mass motion of the exciton exists.

The superradiance described in the above reports is based on coupling between atoms or molecules. However, owing to the quantization of the energy levels of electrons, holes and excitons [15], semiconductor QDs can be regarded as an isolated quantum system, in addition to being regarded as an ensemble of atoms; hence, an ensemble of semiconductor QDs may possibly exhibit superradiance or superfluorescence in the same manner as atoms and molecules do. Furthermore,

as a consequence of the radiative time of the exciton being shorter than that for electronic transitions in atoms and molecules, ultrafast radiation or ultrashort pulsed emission can be expected to be typical of cooperative emission from semiconductor QDs. Thus far, the superradiance phenomenon under low-excitation conditions has been reported only for CdSe QDs [16]. To the best of our knowledge, however, there have been no reports on superfluorescence in QD ensembles, which is classified as superradiance under high-excitation conditions.

One of the problems encountered in generating superfluorescence in a QD ensemble is the difficulty in achieving complete population inversion for certain pairs of levels through conventional excitation methods. For example, if the exciton state and the ground state are chosen as the two levels for complete population inversion, a high excitation intensity is necessary to generate excitons in all the QDs. However, since excitons in QDs are randomly generated following the Poisson distribution, complete population inversion is prevented. We propose a new strategy for achieving complete population inversion in a QD ensemble via the resonant two-photon excitation of a biexciton [17, 18]. Since the resonant two-photon excitation of the biexciton requires the excitation photon energy to be less than the exciton energy by half the biexciton binding energy, the exciton state at the initial stage is completely unoccupied (figure 1). Consequently, complete population inversion is realized between the biexciton and exciton states. Furthermore, cascade relaxation from the biexciton to the exciton states in a QD reduces the probability of transition from the exciton to the biexciton while mutual interaction between excitons generates biexcitons in bulk crystals. Therefore, the entire biexciton population generated during the initial stage can be kept intact while generating superfluorescence.

CuCl QDs are appropriate for achieving complete population inversion between the biexciton and exciton states through resonant two-photon excitation [18, 19]. This is because the biexciton binding energy in CuCl QDs [20–22] is sufficiently large to distinguish between one- and two-photon absorption bands. Thus far, we have reported highly efficient laser emission from the biexciton states in CuCl QDs embedded in a NaCl matrix under two-photon excitation of the biexcitons [23, 24]. Furthermore, we have observed pulsed emission from the biexcitons through time-resolved photoluminescence (PL) measurements, which suggest the possibility that the emission exhibits superfluorescence [25]. In this paper, we report the dependence of the time profiles of the PL from the biexcitons upon the excitation intensity. We observed that the peak intensity of the pulsed emission is proportional to the square of the number of excited QDs. This dependence is consistent with the nature of superfluorescence.

2. Experimental details

Samples of CuCl QDs embedded in NaCl matrices were fabricated by the transverse Bridgman method [26]. The nominal concentration of CuCl was about 1 mol%, and the
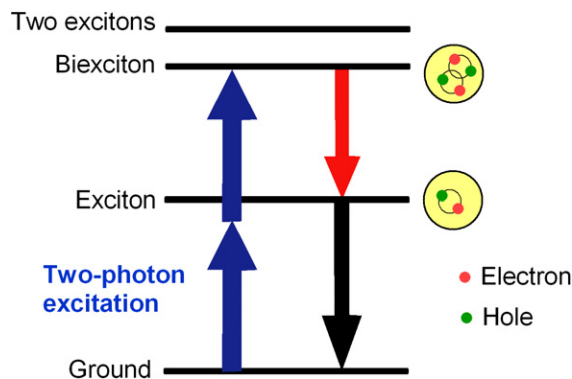

Figure 1. Energy level diagram for the exciton and biexciton states. Blue arrows represent two-photon excitation to the biexciton state. Red and black arrows represent the radiative relaxation of the biexciton and exciton, respectively. The excitation photon energy for resonant two-photon excitation to the biexciton is lower by half the biexciton binding energy compared to the exciton energy.

average dot radius was approximately 5.5 nm, which was estimated by observing the blue-shift of the PL energy of free excitons at 77 K. The sample was cleaved to yield platelets about ~500 μm thick. Time-resolved luminescence measurements were performed by means of an optical Kerr gate method using a pulse laser system based on a regenerative amplifier; the pulse laser system had an auto-correlation pulse width of 220 fs, a wavelength of 800 nm and a repetition rate of 1 kHz. The laser beam from the regenerative amplifier was split into two beams—one served as a gate light to the Kerr medium, while the other was used as the pump light for an optical parametric amplifier. The excitation light was obtained using the fourth-harmonic generation of the signal beam from the optical parametric amplifier. The wavelength of the excitation light was tuned to match the photon energy corresponding to two-photon direct excitation of the biexcitons (3.192 eV) or the resonant excitation energy of the excitons (3.209 eV). The spectral width of the excitation light was ~12 meV. The time resolution of the measurement system was ~1.2 ps. The excitation light was focused on the sample in the shape of a stripe (of length ~820 μm and width ~60 μm) using a cylindrical lens. PL emitted from an edge of the sample was analyzed using a spectrometer equipped with a charge-coupled device. The sample was maintained at 15 K in a He-flow cryostat.

3. Results and discussion

The time-integrated PL spectrum under resonant two-photon excitation of biexcitons is shown in figure 2, wherein the excitation photon energy (3.192 eV) is lower than the exciton energy in the bulk crystals (3.204 eV). Two PL bands, denoted as M and BM in the figure, were observed, and these have been attributed to free and bound biexcitons, respectively, in a previous report [27]. In our former measurements under a backward scattering configuration, we found that the PL band corresponding to the exciton, which is generally the bound exciton band at low temperatures, appeared at the
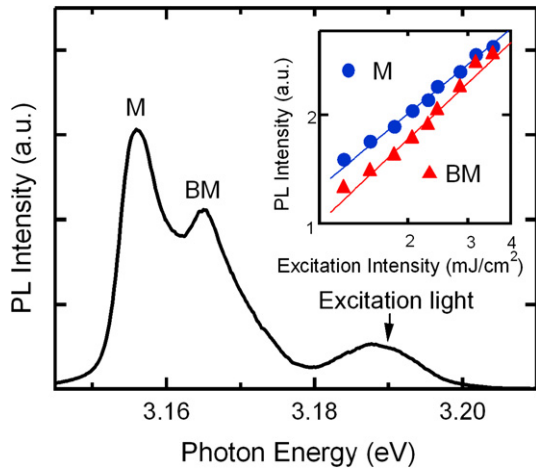
Figure 2. The time-integrated PL spectrum under resonant two-photon excitation of the biexcitons. The PL peaks denoted as M and BM are attributed to the free and bound biexcitons, respectively [27]. The inset shows the dependence of the PL intensities of the M and BM bands on the excitation intensity (blue circles and red triangles). The solid lines indicate the dependence on the excitation intensity to the power of 2.6 and 3.0 (blue and red lines), respectively, for the M and BM bands.

higher energy side of the excitation energy [18]. However, we could not observe the abovementioned PL band in the present experiment. Either the bound exciton band overlapped with the spectrum formed from scattered excitation light or it disappeared due to reabsorption of the tail of the exciton absorption band in the sample. As the excitation intensity increases, the number of generated biexcitons is expected to show a quadratic dependence on the excitation intensity. However, the PL intensities of the generated biexcitons show a power dependence greater than the quadratic dependence

(>2) on the excitation intensity, as shown in the inset in figure 2. This result implies that the PL intensity of the biexciton increases effectively along the direction of the stripe of the excitation volume.

Figures 3(a) and (b) show contour maps of the time-resolved PL spectra that were obtained using excitation light with intensities of 0.92 mJ cm^{-2} and 3.5 mJ cm^{-2}, respectively. At 0.92 mJ cm^{-2}, the M and BM bands show decay times of $\sim$50 ps and $\sim$40 ps, respectively, in contrast to the previously reported values of 125 ps and 70 ps under band-to-band excitation [27]. The short decay times observed in this experiment probably resulted from the amplified spontaneous emission, facilitated by long propagation lengths and population inversion between the biexciton and exciton states. On the other hand, at the intensity of 3.5 mJ cm^{-2}, the time taken to reach the maximum PL for the M band is greater than at 0.92 mJ cm^{-2}. Furthermore, the decay times for the M and BM bands under the latter excitation intensity are shorter than those under the former. It is difficult to attribute the increase in the delay time before the maximum PL is reached to the effect of the stimulated emission, because the propagation time of the luminescence in the excited stripe line, about 4.1 ps, was less than the delay time of $\sim$20 ps for the M band and $\sim$12 ps for the BM band. We shall discuss the abnormal behavior of the abovementioned PL time profiles later. Further, at 3.5 mJ cm^{-2}, a new PL band with a time profile of a pulse at 3.172 eV before the formation of the BM band. Since this new PL band pulse appears in a very short time, we could not observe it in the time-integrated PL spectra. Hereafter, we refer to this band as the FM band and we focus on its features. The biexciton cannot be generated by circularly polarized light through the two-photon absorption process because the total angular momentum of the biexciton is $J = 0$ [17, 18, 28]; therefore, in order to confirm that the FM band originated from
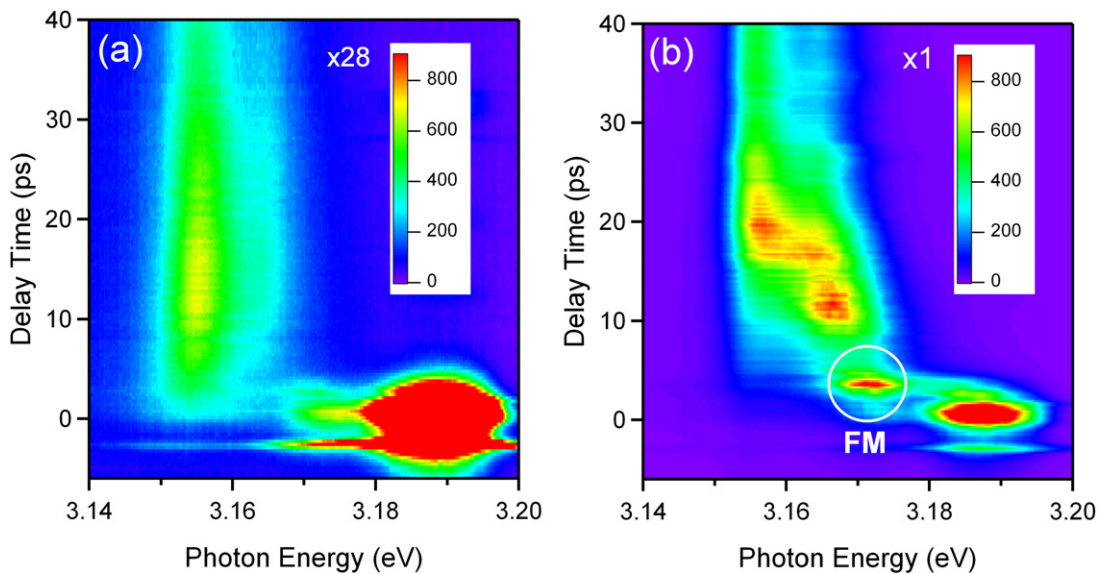

Figure 3. Time-resolved PL contour maps for resonant two-photon excitation of biexcitons. The excitation intensities corresponding to (a) and (b) are 0.92 mJ cm^{-2} and 3.5 mJ cm^{-2}, respectively. A pulse from the scattered excitation light (3.187 eV) is observed at -3 ps and 0 ps from the edge and inside the sample, respectively. In (a), the formation and decay of M (3.155 eV) and BM (3.165 eV) bands is observed. In (b), a new pulsed emission denoted by FM is observed before the formation of the M and BM bands. The scale of the intensity in each figure is normalized to see the change in the PL intensity clearly.
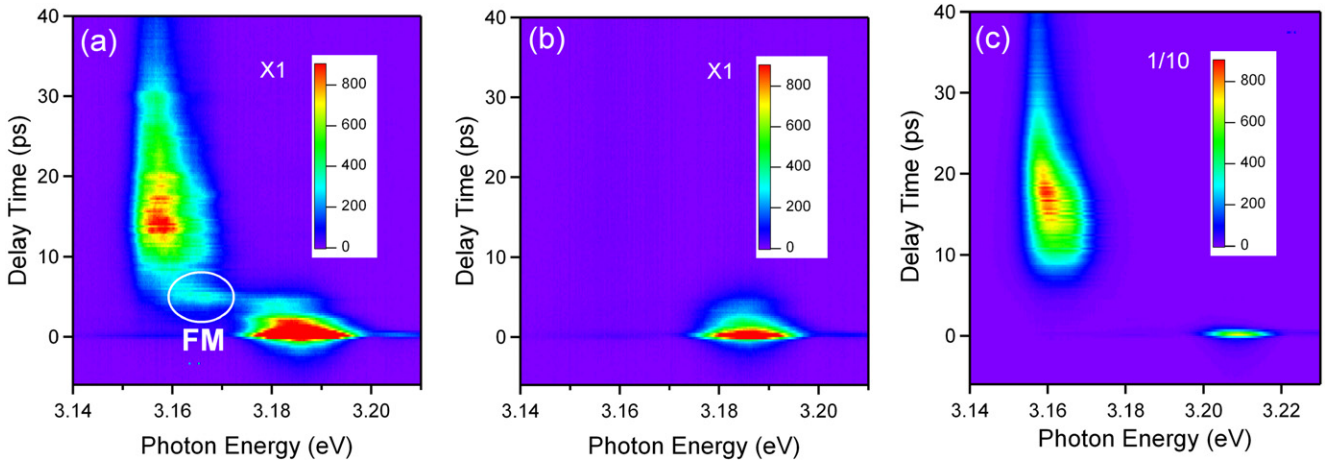
Figure 4. (a), (b) Time-resolved PL contour maps under resonant two-photon excitation of the biexciton corresponding to excitation by linearly polarized and circularly polarized light, respectively, at 2.0 mJ cm^{-2}. (c) The time-resolved PL contour map under resonant excitation of the excitons. The energy of the photons used for excitation is 3.209 eV and the excitation intensity is 2.3 mJ cm^{-2}. The scale of the intensity in each figure is normalized to see the change in the PL intensity clearly.
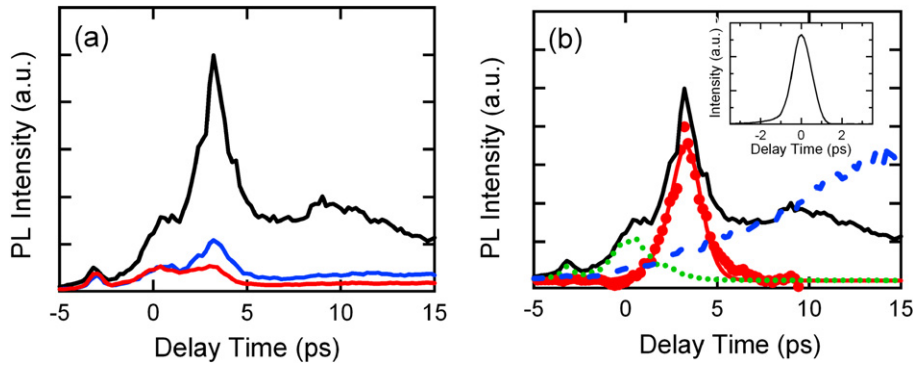

Figure 5. Time profiles of the FM band. (a) The time profiles obtained from experiments with excitation intensities of 3.5 mJ cm^{-2} (black line), 2.80 mJ cm^{-2} (blue line), and 2.3 mJ cm^{-2} (red line). (b) The extraction of time profiles of the FM band from the time profile obtained in the experiment (black-solid curve). The green-dotted curve and blue-dashed curve represent the contribution of the scattered excitation light and the rising part of the BM band, respectively. The time profile of the FM band extracted from the profile obtained from the experiment is shown by red circles. The red-solid curve represents the calculated values obtained by assuming that the time profile of superfluorescence is given by the convoluted result of equation (1) and the time window function of the measurement shown in the inset.

the biexciton state, the polarization of the excitation light was examined. Figures 4(a) and (b) show the contour maps of the time-resolved PL spectra obtained using linearly polarized and circularly polarized excitation light, respectively. It is clear that along with the M and BM bands, the FM band also disappeared when the excitation light was circularly polarized. This result confirms that the FM band originated in the biexciton states. Figure 4(c) shows the contour map of the time-resolved PL spectra obtained with the resonant excitation of the exciton band; the excitation photon energy was set to 3.209 eV. The M band is dominant relative to the BM band, and its time profile shows a slow rise to the PL peak with a very short decay time. However, the FM band did not appear under resonant excitation of the excitons. Consequently, it is hypothesized that complete population inversion between the biexciton and exciton states is necessary to produce the FM band. The above results suggest that the FM band may be due to superfluorescence.

Time profiles of the FM band for different excitation intensities are shown in figure 5(a). The peak at delay time $t \sim -3$ ps is due to partial scattering of the excitation light at the sample edge, while that at $t = 0$ ps is due to the scattering of the primary excitation beam inside the sample. The FM band appears at a delay time of $t \sim 3$ ps, and its intensity increases superlinearly with the excitation intensity, indicating that the FM band is a consequence of luminescence from the actual biexciton state and not the scattering of the excitation light. In order to confirm that the FM band exhibits superfluorescence originating in the biexciton state, the time profile of the FM band was obtained as explained below.

The time profiles of the FM band were extracted carefully from the raw time profiles by considering the contributions of the tails of the band corresponding to the scattered excitation light and the BM band. Figure 5(b) shows the extracted time profile of the FM band (red circles) obtained by subtracting the sum of the normalized profiles of the scattered excitation light at 0 ps (green-dotted curve) and the rise of PL in the BM band (blue-dashed curve) from the experimental profile (black-solid curve). The extracted time profile of the FM band was
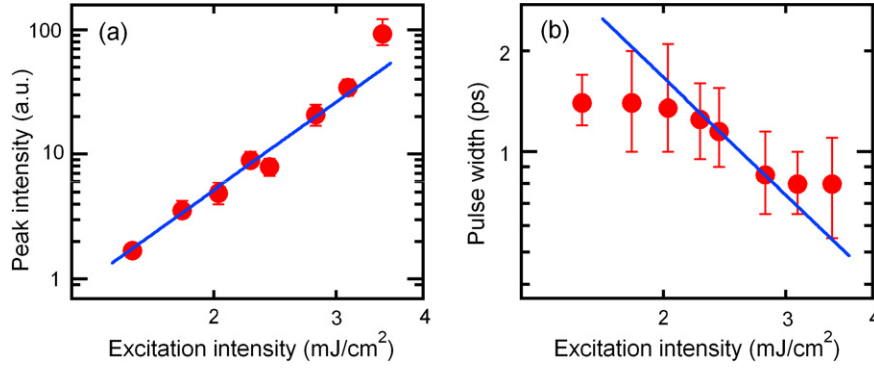
Figure 6. (a), (b) The dependence of peak intensity I_0 and pulse width τ_r for the FM band, respectively, on the excitation intensity, obtained by fitting the time profiles (red circles) to a curve as mentioned in the text. The blue lines show the predicted dependences in ideal superfluorescence under the assumption that the number of excited dots is proportional to the square of the excitation intensity (peak intensity $\propto I_{exc}^4$ and pulse width $\propto I_{exc}^{-2}$).

fitted to a curve that was the convolution of the time window function shown in the inset and an ideal time profile of the superfluorescence with an infinite dephasing time [3], as given below:

$$I(t) \propto I_0 \, \text{sech}^2\left(\frac{t - \tau_0}{\tau_r}\right), \qquad (1)$$

where I_0 is the peak intensity, τ_0 is the delay time of the peak intensity and τ_r is the pulse width. In figure 5(b), the values of the PL intensity for the FM band calculated using (1) are shown by the solid-red curve. The dependences of I_0 and τ_r on the excitation intensity in the above analysis are shown in figures 6(a) and (b), respectively. For superfluorescence from the QD ensemble, I_0 and τ_r are related to N in the following manner:

$$I_0 \propto N^2 \qquad (2)$$

$$\tau_r \propto 1/N. \qquad (3)$$

The experiments showed that the increase in I_0 had almost a fourth-power dependence on the excitation intensity. This dependence clearly follows the relation expressed in equation (2), under the assumption that the density of excited QDs is proportional to the square of the excitation intensity under two-photon excitation. In addition, beyond $\sim$2 mJ cm^{-2}, τ_r was found to decrease with increasing excitation intensity. This dependence qualitatively agrees with the superfluorescent behavior. These results strongly suggest that the FM band can be attributed to superfluorescence from the biexciton states. It is to be noted that the observed delay time remained almost constant, although it was expected to decrease with an increase in the number of excited QDs. In the present experiments, insufficient time resolution of the measurement led to difficulties in the evaluation of the excitation intensity dependence of the delay time since no change in the latter could be observed. Furthermore, the experimental time profiles were obtained by averaging many pulses, while the characteristics of superfluorescence had to be evaluated for individual pulses [29]. Therefore, we are unable to discuss the quantitative agreement of the delay time τ_0 with the superfluorescent behavior.

In order to induce superfluorescence, the dephasing time needs to be longer than both τ_r and τ_0. In CuCl QDs embedded

in a NaCl matrix, exciton superradiance, which occurs in a shorter time than the dephasing time of the exciton, has been observed to have a radiative time of more than $\sim$100 ps [13]. Furthermore, the dephasing time of excitons for CuCl QDs in glass has been reported to be 130 ps [30]. From these reports, it can be concluded that the exciton dephasing time in the CuCl QD is sufficiently long for superfluorescence to occur. Although the dephasing time for the biexcitons has not been reported, we assume that it is longer than the duration of the presence of the FM band, for the following reasons. The biexciton–acoustic phonon scattering time in bulk crystals was reported to be $\sim$3.2 ps at 20 K [31], which was estimated using the spectral width of the two-photon absorption band of biexcitons. On the other hand, the biexciton dephasing time according to a four-wave-mixing measurement was 16 ps at 5 K [32]. In general, the zero-phonon line width of the biexciton is expected to be narrower in QDs than in bulk crystals because quantization of the acoustic phonon energy in the QD results in a decrease in the biexciton–acoustic phonon interaction. This mechanism to narrow the spectral width has been reported for excitons in the CuCl QDs [33]. In addition, collisions among biexcitons, which cause dephasing of the biexcitons under high-density excitation, can be neglected in QDs because it is improbable that more than two biexcitons exist in a QD. Consequently, it is reasonable that the dephasing time of a biexciton in CuCl QDs is sufficiently long to produce superfluorescence.

For cooperative spontaneous emission, it is necessary that the QDs possess a common transition energy to interact through the electromagnetic field. Therefore, one has to take into account the inhomogeneously broadened spectral width of biexciton luminescence due to the QD size distribution. In our experiments, the QD density was estimated from the averaged dot radius of 5.5 nm and CuCl concentration of 1 mol% in the fabrication process to be $\sim$1.3 × 10^{16} cm^{-3}, and consequently the number of QDs per effective wavelength ($\sim$260 nm in NaCl) was estimated to be $\sim$230 at 3.172 eV. Given the homogeneous spectral width of $\sim$200 μeV at 15 K [34] and inhomogeneous broadening of the exciton band by $\sim$15 meV, we estimate that several QDs only interact with one another if the exciton luminescence shows superfluorescence. However,

the inhomogeneous spectral width of the PL energy of the biexciton was found to be much less than that of the excitons for the size distribution because the increase in the biexciton binding energy for the smaller QD cancels the blue-shifting of the exciton energy [19, 20]. Consequently, we concluded that the QD density is sufficient to generate superfluorescence by facilitating interaction between the QDs.

Under two-photon excitation of biexcitons, in addition to the FM band, the ordinal PL bands—the M and BM bands—are capable of exhibiting superfluorescence owing to complete population inversion. When the excitation intensity was increased, it was seen that the PL time profiles of these bands changed from exponential decays to time profiles that shared similarities with the pulsed PL. This transition in the time profile may be reasonably interpreted as a transition from amplified spontaneous emission to superfluorescence, although a more detailed analysis based on numerical calculations is necessary to confirm this.

Finally, we discuss the origin of the FM band. The FM band was observed for the first time in the present measurements, although investigations on biexciton properties in CuCl QDs have been carried out over the last two decades. Thus far, for one-photon excitation of the excitons, the PL bands corresponding to the biexciton state were reported to be the M and BM bands, which were attributed to free and bound biexcitons, respectively [27]. However, in our recent experiments, detailed analysis of the PL spectra under resonant two-photon excitation of the biexciton revealed that the M band is not luminescence from the biexciton state generated directly by the excitation light [19]. Consequently, it is reasonable that the FM band originates from the free biexciton state, and it emerges only in an effective light amplification system by overcoming the rapid transition into the M or BM bands.

4. Conclusions

We measured the time-resolved PL spectra of biexcitons in CuCl QDs using optical Kerr gate spectroscopy. Under resonant two-photon excitation of biexcitons, pulsed emission—in a band termed the 'FM band'—was observed from the biexciton state for the first time. The FM band appeared only under resonant two-photon excitation of biexcitons, therefore we concluded that complete population inversion between the biexciton and exciton states is necessary to produce the FM band. In addition, the dependence of the time profiles of the FM band on the excitation intensity was of a superfluorescent nature when we assume that the number of excited QDs increases with the square of the excitation intensity. Consequently, we concluded that the FM band is produced by superfluorescence from the biexciton states in CuCl QDs. This is the first observation of superfluorescence in a semiconductor QD assembly.

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