Optical properties of biexcitons in ZnS

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Characteristics of biexcitons in ZnS epitaxial layers with biaxial tensile strain have comprehensively been studied by means of photoluminescence excitation spectroscopy, magnetophotoluminescence spectroscopy, and time-resolved photoluminescence spectroscopy. Photoluminescence excitation measurement of biexcitons allowed us to observe two-photon absorption of biexcitons and to determine the biexciton binding energy of 8.0 ± 1.2 meV. Under a magnetic field, the luminescence line due to the radiative recombination of light-hole free excitons slightly shifted to the lower-energy side (about 0.3 meV at 9 T). On the other hand, the luminescence line due to the radiative recombination of biexcitons split into two components. This splitting (about 1.2 meV at 9 T) originated from the Zeeman splitting of light-hole free excitons in the final state of the optical transition for biexciton luminescence. In addition, the luminescence decay times of both light-hole free excitons and biexcitons were estimated to be about 130 and 80 ps, respectively. The observed temporal behavior was consistent with the expected recombination dynamics between excitons and biexcitons.

I. INTRODUCTION

Excitonic optical properties have extensively been studied to date in a large variety of semiconductors. In highly excited semiconductors, it is now well established that the interaction between two or more excitons causes the unique properties that are characterized by the formation of biexcitons (excitonic molecules) and the inelastic scattering of excitons.[1] Such characteristic phenomena of a dense excitonic system have been studied mainly in wide-gap semiconductors because of the advantages of excitonic nature: excitons in wide-gap semiconductors have relatively larger binding energies as well as smaller Bohr radii, so that the excitons exist stably at higher densities. As a result, the above phenomena are remarkably observed in the dense excitonic systems of wide-gap semiconductors.

From the point of view of exciton engineering, it is important to clarify nonlinear optical properties of dense excitonic phenomena. It has been pointed out that recombination processes of dense excitons in wide-gap semiconductors contribute to the formation of optical gain.[2–6] This situation is more pronounced in low-dimensional structures because of the enhancement in exciton binding and its oscillator strength due to the effect of quantum confinement.[7–12]

Among wide-gap semiconductors, ZnS has a large exciton binding energy and a small Bohr radius of about 37 meV and 2.4 nm, respectively. Therefore, ZnS is one of the most fruitful candidates to explore the intrinsic recombination processes of dense excitonic systems. The advantage of excitonic nature in ZnS is also expected to be an effective factor for applications to low-threshold ultraviolet laser diodes due to excitonic optical gain formation.[13]

In this paper we concentrate our work on the optical characteristics of biexcitons in ZnS. We present comprehensive studies on biexcitons in ZnS by means of photoluminescence excitation (PLE) spectroscopy, magnetophotoluminescence magneto-PL spectroscopy, and time-resolved PL spectroscopy.

II. EXPERIMENTAL PROCEDURE

The cubic-structured ZnS epitaxial layers used in the present work were grown on (100)-oriented GaAs substrates by means of a low-pressure metalorganic chemical vapor deposition technique with all gaseous sources.[14] Dimethylzinc at a concentration of 1.06% diluted in a He gas and hydrogen sulfide at a concentration of 10.8% diluted in a H_2 gas were used as Zn and S sources, respectively. These gases were supplied to a reaction chamber directly through the piezovalves of mass flow controllers and air-operated valves. The growth conditions were as follows: a growth temperature of 500 °C, a growth pressure of about 0.5 Torr, and a VI/II flow ratio of 18. The growth rate of ZnS epitaxial layers was estimated to be about 1 μm/h. The layer thickness was varied from 2 to 8 μm. It was found from high-resolution double-crystal x-ray diffraction measurement that the rocking curve full width at half maximum of the (400) ZnS reflection was about 600 arc sec for a 2-μm-thick layer, and decreased to 144 arc sec for an 8-μm-thick layer.[15]

PL measurement was carried out by employing a Xe-Cl excimer laser (308 nm) as an excitation source in order to perform band-to-band excitation. The pulse width and the repetition rate were 2.5 ns and 100 Hz, respectively. PL spectra were detected by a liquid-nitrogen-cooled charge-coupled-devices camera in conjunction with a 50-cm single-grating monochromator with a 2400-grooves/mm grating. The spectral resolution was better than 0.025 nm (i.e., better than 0.3 meV around 3.7900 eV). PLE measurement was performed by employing the second-harmonic light from a dye laser pumped by a frequency-doubled *Q*-switched Nd^{3+}:YAG (yttrium aluminum garnet) laser. The pulse width and the repetition rate were 3 ns and 100 Hz, respectively. PLE signals were detected by a photomultiplier in conjunction with a 1-m single-grating monochromator with a 1200-grooves/mm grating. Magneto-PL measurement was performed using an optical cryostat with a superconducting split-coil magnet up to 9 T. A time-resolved PL measure-

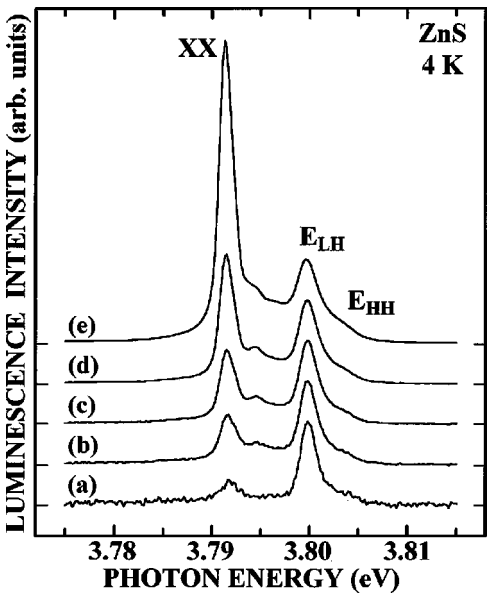

FIG. 1. Excitonic photoluminescence spectra at 4 K taken from a 6-μm-thick ZnS epitaxial layer under excitation-power densities of (a) 0.016, (b) 0.15, (c) 0.26, (d) 0.52, and (e) 1.6 kW/cm^2. Each spectrum is normalized at the peak height of the E_{LH} line.

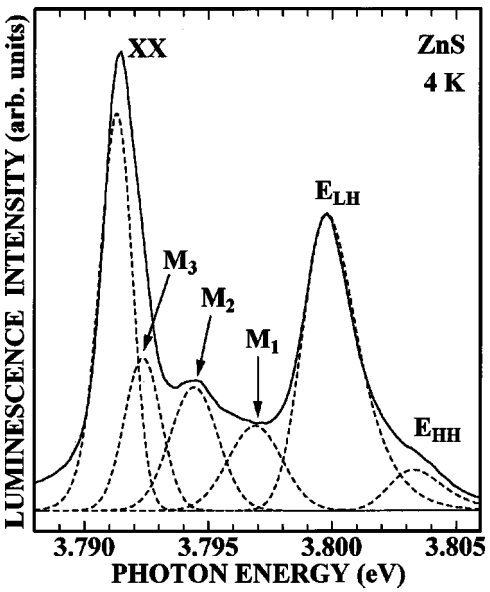

FIG. 2. Spectral deconvolution of excitonic luminescence components. The solid line shows the observed spectrum at 4 K taken from a 6-μm-thick ZnS epitaxial layer under excitation-power density of 0.52 kW/cm^2. Each luminescence component is shown by the dashed lines.

ment was also performed using a synchroscan streak camera in conjunction with a 25-cm single-grating monochromator with a 1200-grooves/mm grating. The excitation source was the third-harmonic light (267 nm) of amplified titanium sapphire laser pulses. The pulse width and the repetition rate were 200 fs and 250 kHz, respectively. The instrumental response of our system had an exponential decay-time constant of about 5 ps.

III. EXPERIMENTAL RESULTS

A. Exciton and biexciton luminescence

ZnS epitaxial layers grown on GaAs substrates are affected by biaxial tensile strain because of a larger thermal expansion coefficient of ZnS ($6.7\times10^{-6}\,\mathrm{K}^{-1}$) as compared to that of GaAs ($5.8\times10^{-6}\,\mathrm{K}^{-1}$). The strain splits the uppermost degenerate valence band into heavy-hole and light-hole bands. We have recently analyzed the tensile strain in ZnS epitaxial layers on the basis of a bimetallic strip model[16] and obtained the resultant energy splitting between heavy-hole and light-hole bands.[15,17] Experimental energy splitting was also derived from a Boltzmann deconvolution of heavy-hole and light-hole splitting of free-exciton luminescence. As a result, it was found that the experimental values approximately agreed with the calculated results.[15]

Figure 1 shows the excitonic PL spectra at 4 K taken from a 6-μm-thick ZnS epitaxial layer under excitation-power densities of (a) 0.016, (b) 0.15, (c) 0.26, (d) 0.52, and (e) 1.6 kW/cm^2. At the excitation-power density of (a) 0.016 kW/cm^2, the spectrum is dominated by the radiative recombination of light-hole free excitons (denoted by E_{LH}). This assignment is confirmed by our separate reflectance measurement.[17] The peak position is located at 3.7998 eV and the linewidth is about 2 meV. At the higher-energy side of the E_{LH} line, a weak luminescence line is also observed, which is due to the radiative recombination of heavy-hole free excitons (denoted by E_{HH}). It is found from the Boltzmann deconvolution that the peak position of the E_{HH} line is located at 3.8036 eV and the energy splitting between heavy-hole and light-hole free excitons is 3.8 meV. This value is approximately supported by the calculated value of 4.1 meV.[15] Each spectrum shown in Fig. 1 is normalized at the peak height of the E_{LH} line.

With increasing excitation-power density, the E_{LH} line grows almost linearly. The dependence of the E_{LH} intensity (I_{LH}) on the excitation intensity (I_P) gives approximately $I_{\mathrm{LH}}\propto I_P^{1.1}$. On the other hand, a luminescence line at 3.7915 eV grows superlinearly and becomes prominent. On the basis of our previous paper,[18] this line (denoted by XX) is attributed to the radiative recombination of biexcitons, especially light-hole biexcitons.[19] The dependence of the XX intensity (I_{XX}) on the excitation intensity (I_P) gives approximately $I_{XX}\propto I_P^{1.9}$. The energy separation between the E_{LH} and the XX luminescence lines is 8.3 meV. In addition to the main spectral features mentioned above, it is found from the lineshape analysis that there exist three additional luminescence components between the E_{LH} and the XX lines. These three components are denoted by M_1, M_2, and M_3, and are shown in Fig. 2 by the dashed lines. The observed spectrum shown in Fig. 2 by the solid line is identical to spectrum (d) in Fig. 1. The peak positions are located at 3.7969, 3.7944, and 3.7924 eV for M_1, M_2, and M_3, respectively. The origin of these components is not clear at the present time. However, it is found that all three components indicate the superlinear dependence on the excitation-power density. Therefore, these components may be related to the biexcitonic transitions associated with the first excited states of biexcitons or heavy-hole biexcitons.

B. Two-photon absorption of biexcitons

In order to confirm our assignment mentioned above, PLE measurements of the XX line were performed. Figure 3

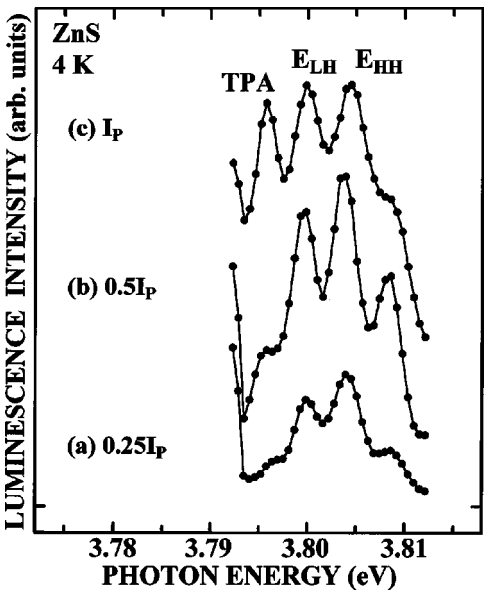

FIG. 3. Photoluminescence excitation spectra of biexcitons at 4 K taken from a 6-μm-thick ZnS epitaxial layer under excitation-power densities of (a) $0.25I_P$, (b) $0.5I_P$, and (c) I_P ($I_P = 8.5\,\mathrm{kW/cm^2}$). The detected photon energy is tuned at 3.7915 eV.

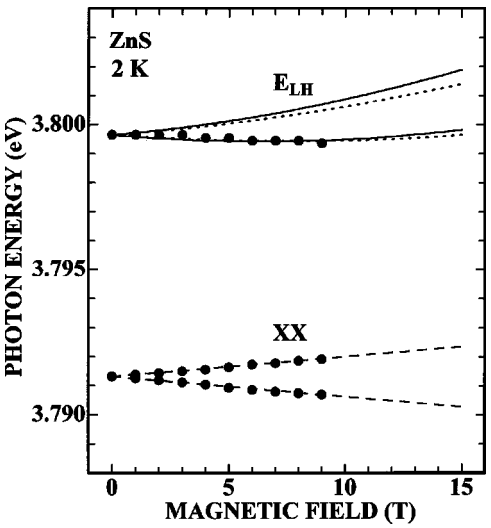

FIG. 5. Peak energies of the light-hole free-exciton luminescence line (E_{LH}) and the biexciton luminescence line (XX) as a function of magnetic field. The solid lines are due to a least-squares fit to the experimental energy shift of the E_{LH} line. The dashed lines for the XX line only represent the linear Zeeman splitting. The dotted lines for the E_{LH} line represent the calculated results.

shows the PLE spectra at 4 K taken from the 6-μm-thick ZnS epitaxial layer under excitation-power densities of (a) $0.25I_P$, (b) $0.5I_P$, and (c) I_P ($I_P = 8.5\,\mathrm{kW/cm^2}$). The detected photon energy is tuned at 3.7915 eV, which corresponds to the peak energy of the light-hole biexciton luminescence (XX). The excitation photon energy is varied by a step of about 0.6 meV (0.05 nm). In all three spectra, two clear peaks are observed at 3.7998 and 3.8039 eV. These are unambiguously attributed to the light-hole free exciton (E_{LH}) and the heavy-hole free exciton (E_{HH}) resonances, respectively. In addition, there exists an excitation-intensity-dependent peak (denoted by TPA) at the lower-energy side of the E_{LH} resonance. The peak position is located at 3.7958 eV, which is just at the center between the peaks of the E_{LH} resonance and the light-hole biexciton luminescence (XX). Therefore, the peak denoted by TPA is attributed to the two-photon biexciton resonance, which indicates the direct creation of biexcitons from a ground state by a two-photon absorption process. There also exists a peak at 3.8086 eV, which is located at 4.7 meV above the E_{HH} resonance. The origin of this peak is not clear at the present time.

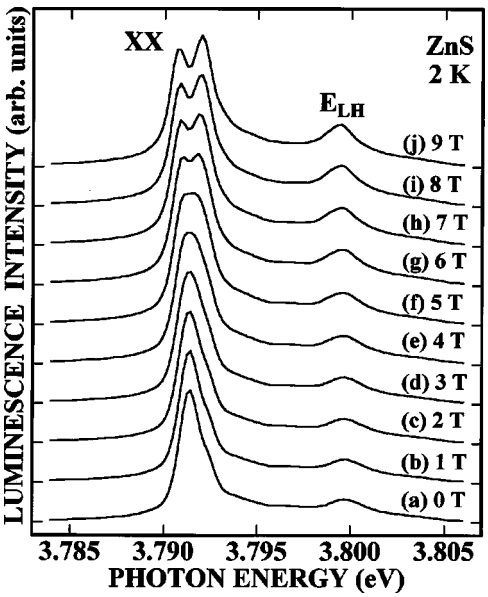

FIG. 4. Excitonic photoluminescence spectra at 2 K taken from an 8-μm-thick ZnS epitaxial layer under magnetic fields from (a) 0 to (j) 9 T in a Faraday configuration. The excitation-power density is 1.3 kW/cm^2.

C. Biexciton luminescence under magnetic field

Figure 4 shows the excitonic PL spectra at 2 K taken from an 8-μm-thick ZnS epitaxial layer under external magnetic fields from (a) 0 to (j) 9 T in a Faraday configuration. The excitation-power density is 1.3 kW/cm^2. In this sample, the luminescence linewidths of both the E_{LH} and the XX lines at 0 T are 1.7 and 1.1 meV, respectively.[15] With increasing magnetic field, the E_{LH} line indicates a slight redshift (about 0.3 meV at 9 T). On the other hand, the XX line clearly splits into two components. The splitting energy is about 1.2 meV at 9 T. It is known that the biexciton is a spin-singlet state and therefore its energy cannot be split by a magnetic field. However, the final state of the optical transition for biexciton luminescence is an exciton. Therefore, the splitting of the biexciton luminescence is attributed to the Zeeman splitting of the exciton,[20] the light-hole exciton in this case. The Zeeman splitting energy is given by $\Delta E_{\pm} = g^* \mu_B B$, where g^* is the effective g factor of exciton, μ_B is the Bohr magneton, and B is the applied magnetic field. On the basis of the lineshape analysis, the Zeeman splitting energy is evaluated as a function of the magnetic field. As a result, it is found that the Zeeman splitting energy is almost linear with the magnetic field and that the coefficient of $g^* \mu_B$ is estimated to be 1.38×10^{-4} eV/T. This value corresponds to the effective g factor of 2.38 ± 0.10 for the light-hole exciton.

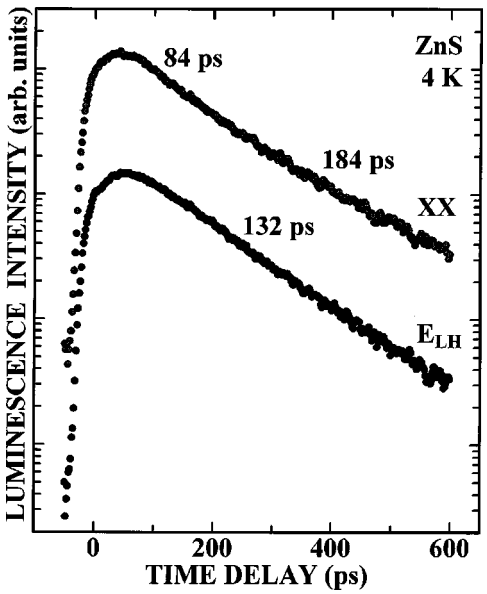

FIG. 6. Time-resolved luminescence of light-hole free exciton (E_{LH}) and biexciton (XX) lines at 4 K taken from an 8-μm-thick ZnS epitaxial layer under excitation-energy density of 0.2 μJ/cm^2.

Figure 5 shows the peak energies of the E_{LH} and the XX lines as a function of the magnetic field. The solid lines are due to a least-squares fit to the experimental energy shift for the E_{LH} line on the assumption that the observed E_{LH} line is attributed to the lower-energy component of two spin splitting ones. This fitting includes a quadratic diamagnetic shift in addition to the Zeeman splitting mentioned above, and the diamagnetic shift is evaluated to be 5.37×10^{-6} eV/T^2. The dotted lines represent the calculated result, which will be discussed later in detail. On the other hand, the dashed lines for the peak energies of the XX line only represent the linear Zeeman splitting mentioned above. Without including the quadratic term with the magnetic field, the dashed lines are in good agreement with the observed peak energies for the XX line. This result indicates that the diamagnetic shift of the biexciton is almost as large as that of the exciton (light-hole exciton).

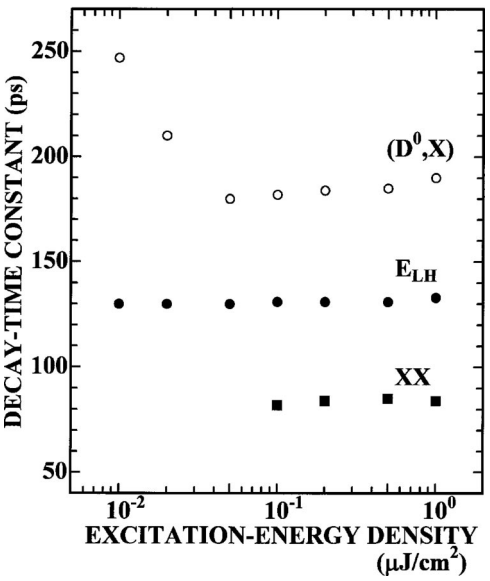

FIG. 7. Decay-time constants of light-hole free exciton (E_{LH}), biexciton (XX), and donor-bound exciton [(D^0,X)] luminescence as a function of excitation-energy density.

D. Time-resolved luminescence of biexcitons

We have also measured the recombination dynamics of excitons and biexcitons by means of time-resolved PL spectroscopy. Figure 6 shows the time-resolved luminescence of both the light-hole free exciton (E_{LH}) and the biexciton (XX) at 4 K taken from an 8-μm-thick ZnS epitaxial layer under an excitation-energy density of 0.2 μJ/cm^2. The photon flux density irradiated to the sample surface at 0.2 μJ/cm^2 is estimated to be about 2.7×10^{11} cm^{-2}. This value is almost comparable to the density at 1.3 kW/cm^2 under Xe-Cl excimer laser excitation. The luminescence intensity of the E_{LH} line decays exponentially with a decay-time constant of about 132 ps. On the other hand, the luminescence intensity of the XX line does not indicate a single-exponential decay. Then, we consider that there exist two different recombination channels at the energy position of the XX line. One is unambiguously due to the radiative recombination of biexcitons as mentioned above. The other is probably due to the radiative recombination of excitons bound to neutral donors because it is well known that the energy position of the donor-bound exciton is much too close to that of the biexciton.[18,21,22] Therefore, we simply assume a double-exponential decay for the XX line. Using a least-squares fit to the experimental data, the decay-time constants of the faster- and slower-decay components are estimated to be about 84 and 184 ps, respectively. The faster- and the slower-decay components are attributed to the radiative recombination of the biexciton and the exciton bound to neutral donors, respectively. The decay-time constants of both the E_{LH} line and the XX line as a function of excitation-energy density are summarized in Fig. 7. Within the excitation-energy densities from 0.01 to 1 μJ/cm^2, the decay-time constant of the E_{LH} line (denoted by solid circles) is almost constant. On the other hand, it is found that the XX line indicates a single-exponential decay at the excitation-energy densities below 0.05 μJ/cm^2. In this excitation range, the decay-time constant gradually decreases with increasing excitation density (denoted by open circles). Above 0.1 μJ/cm^2, the XX line indicates a double-exponential decay as mentioned in Fig. 6. The faster- and the slower-decay-time constants are denoted by solid squares and open circles, respectively. It is reasonable to understand that the luminescence line below 0.05 μJ/cm^2 is dominated by the radiative recombination of excitons bound to neutral donors [denoted by (D^0,X)]. And, above 0.1 μJ/cm^2, the contribution of biexcitons appears and becomes prominent with increasing excitation-energy density, which results in the double-exponential decay profile for the XX line. This assignment is consistent with the excitation-energy-density dependence of the luminescence intensity: it is found that the XX line indicates the superlinear dependence on the excitation-energy density above 0.1 μJ/cm^2. It is clear that the decay-time constant of the biexciton luminescence is smaller than that of the free-exciton luminescence, and is also constant within the excitation-energy densities from 0.1 to 1 μJ/cm^2. This result is consistent with the

expected recombination dynamics between excitons and biexcitons: the shorter lifetime of the biexciton as compared to that of the exciton.[23]

IV. DISCUSSION

On the basis of the PLE measurements shown in Fig. 3, we first examine the binding energy of the biexciton in ZnS. The resonance of the light-hole free exciton is located at 3.7998 eV. The two-photon resonance of the biexciton is observed at 3.7958 eV. The energy difference between the two resonances is 4.0 ± 0.6 meV. This energy difference corresponds to half of the biexciton binding energy. Therefore, we obtain the biexciton binding energy of 8.0 ± 1.2 meV. This value approximately agrees with our previous paper, in which we estimated the biexciton binding energy of 9 meV on the basis of the energy difference between the PL peaks of the exciton and the biexciton.[18]

Next, we examine the spectral features of excitons and biexcitons under a magnetic field. The theoretical calculation of effective g factors for both light-hole and heavy-hole excitons, g_{LH} and g_{HH} has been reported by Cho *et al.*[24] Using the material parameters of ZnS reported by Lawaetz,[25] we obtain the values of $g_{\mathrm{LH}}=2.02$ and $g_{\mathrm{HH}}=1.30$. Then, the Zeeman splitting for the light-hole and the heavy-hole free excitons is evaluated as follows:

$$\Delta E_{\mathrm{Zeeman}}^{\mathrm{LH}} = \pm \tfrac{1}{2} g_{\mathrm{LH}} \mu_B B = \pm 5.85 \times 10^{-5} B \text{ eV}, \quad (1)$$

$$\Delta E_{\mathrm{Zeeman}}^{\mathrm{HH}} = \pm \tfrac{1}{2} g_{\mathrm{HH}} \mu_B B = \pm 3.76 \times 10^{-5} B \text{ eV}. \quad (2)$$

In addition, the diamagnetic shift of a bulk exciton is given by

$$\Delta E_{\mathrm{diamag}} = (e^2 a_B^2 / 4\mu^*) B^2 = (4\pi^2 \epsilon^2 \hbar^4 / \mu^{*3} e^2) B^2, \quad (3)$$

where a_B is the exciton Bohr radius and μ^* is the exciton reduced mass. Using electron, light-hole, and heavy-hole effective masses of $m_e^*/m_0=0.28$, $m_{\mathrm{LH}}^*/m_0=0.23$, and $m_{\mathrm{HH}}^*/m_0=1.76$, respectively, we obtain light-hole and heavy-hole exciton reduced masses of $\mu_{\mathrm{LH}}^*/m_0=0.13$ and $\mu_{\mathrm{HH}}^*/m_0=0.24$, respectively. Then, the diamagnetic shift of the light-hole and the heavy-hole free excitons is evaluated as follows:

$$\Delta E_{\mathrm{diamag}}^{\mathrm{LH}} = 3.88 \times 10^{-6} B^2 \text{ eV}, \quad (4)$$

$$\Delta E_{\mathrm{diamag}}^{\mathrm{HH}} = 6.16 \times 10^{-7} B^2 \text{ eV}. \quad (5)$$

The dotted lines shown in Fig. 5 indicate the calculated result based on Eqs. (1) and (4), and agree well with the experimental result.

For the light-hole free-exciton luminescence, the higher-energy component of the two spin splitting ones cannot be detected clearly. This results from the splitting of the initial state for the optical transition of excitons. It is reasonable to understand qualitatively that the luminescence intensity of the higher-energy component relative to that of the lower-energy one is expected to be much reduced because of the dominant occupation of the lower-energy electronic state. For the biexciton luminescence, on the other hand, the splitting is caused by the final-state splitting for the optical transition. Then, no difference in intensity between the two spin components is expected.

V. CONCLUSIONS

We have studied the optical characteristics of the biexciton in ZnS epitaxial layers. In the PLE spectra of the biexciton, the two-photon absorption of the biexciton was clearly observed, and the biexciton binding energy was evaluated to be 8.0 ± 1.2 meV. Under a magnetic field, the splitting of the biexciton luminescence line was observed. The splitting energy was about 1.2 meV at 9 T. This splitting originated from the Zeeman splitting of the light-hole free exciton in the final state of the optical transition for biexciton luminescence. The time-resolved luminescence was also measured for both the light-hole free exciton and the biexciton. The decay-time constants of the light-hole free exciton and the biexciton luminescence were estimated to be about 130 and 80 ps, respectively. The observed temporal behavior was consistent with the expected recombination dynamics between excitons and biexcitons.

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