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Optical Quenching Mechanism in InAs Quantum Dots in an $Al_{0.95}Ga_{0.05}As$ Matrix

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InAs quantum dots (QDs) were grown grown in an $Al_{0.95}Ga_{0.05}As$ matrix by using the molecular-beam epitaxy technique. Photoluminescence (PL) measurements were made as functions of the magnetic fields and the temperature. Two prominent PL transitions were observed from QDs and defects in the matrix layer at 5 K. In magnetic fields, the transition from QDs does not change its spectral shape at magnetic fields up to 15 T, whereas the defect-related transition shows a blue-shift at magnetic fields above 8 T. By varying the temperature from 5 K to room temperature, the transition from QDs persists up to $\sim$ 200 K and the defects-related transition quenches quickly near 70 K. The activation energies obtained by using an Arrhenius fitting of the PL intensities indicate that the excitons dissociated by thermal energy transfer into higher energy levels.

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I. INTRODUCTION

Due to an atomic-like density of states, epitaxially grown self-assembled semiconductor quantum dots (QDs) can be applied to devices such as single-electron transistors and single-photon generators for quantum information and computing [1, 2], efficient light emitting diodes [3,4] and solar cell devices [5]. Understanding the basic physical properties of QDs is important before applying them to such devices. Especially, understanding the optical transitions of excitons in QDs is an important subject in order to interpret the working mechanism of optoelectronic devices or single-photon devices.

Application of magnetic fields to a low-dimensional semiconductor quantum structure can be widely used to understand exciton dynamics. An exciton (X) has a hydrogen (H) analogy because of its being a bound state of a positive (a hole) and a negative (an electron) charge. In semiconductor quantum structures, due to the reduced dimensionality, not only the significant enhancement of exciton binding energy but also the formation of charged- [6] and bi-excitons [7] expand scientific interests. In the presence of a magnetic field, an exciton undergoes a diamagnetic shift, which is quadratic in the magnetic field and inversely proportional to the effective reduced mass of the exciton. Therefore, if an exciton is confined in a narrow region or is deeply bound, the diamagnetic shift is restricted due to the heavy effective reduced mass [8–10]. Charged excitons (X^- and X^+) have an analogy to ionized hydrogen, H^- and H_2^+. In such a system, spin singlet and triplet states show strong magnetic-field dependences. Therefore, application of an external magnetic field can make it possible to understand the exciton dynamics in QDs.

In this work, we report the photoluminescence emission properties of InAs QDs in an $Al_{0.95}Ga_{0.05}As$ matrix grown on a GaAs substrate. Two pronounced photoluminescence (PL) emission peaks emerge and are identified as transitions of InAs QD excitons and defects in the matrix. In the presence of a magnetic field, the exciton transition does not change its spectral shape at magnetic field up to 15 T. This is due to the strong confinement effect, which significantly enhances the exciton's effective mass. By increasing the temperature from 5 K to room temperature, we found that the defect transition quenched quickly around 70 K whereas the transition from QDs persisted to $\sim$ 200 K. The Ahrrenius fitting indicates that the PL intensity quenching of the exciton recombination reflects the thermal dissociation of the excitons. Once the exciton is dissociated by the thermal energy, the carriers in the QDs transfer to the defect level

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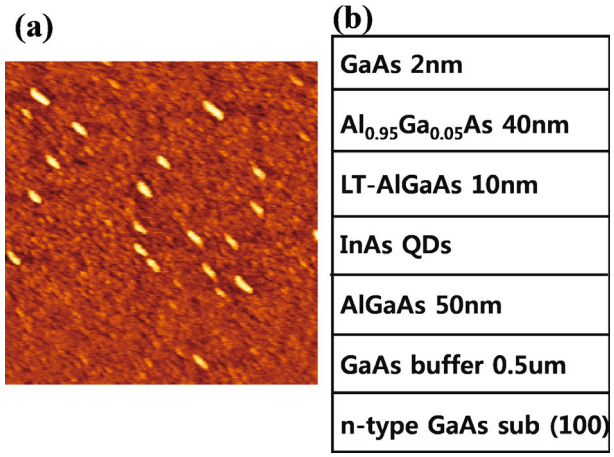

Fig. 1. (Color online) (a) Atomic force microscopy (AFM) image (1 μm $\times$ 1 μm) of the InAs QDs. The shapes of the QDs are slightly elongated along the [1$\bar{1}$0] direction. The average size of the QDs is 12 $\times$ 40 nm for the height and the length, respectively. (b) Sample structure used for this study. InAs QDs were grown in an $Al_{0.95}Ga_{0.05}As$ matrix.

with further increasing temperature.

II. EXPERIMENT

The sample was grown by using a molecular-beam epitaxy system equipped with a cryogenic pump and an ion getter pump. An n-typed GaAs (100) substrate was thermally cleaned at a substrate temperature of 620 °C under an arsenic tetramer (As4) flux at a base pressure of 1 $\times$ 10^{-5} Torr and was monitored using reflection high-energy electron diffraction (RHEED) pattern. At a growth temperature of 600 °C, a 500-nm-thick GaAs buffer layer and a 50-nm-thick $Al_{0.95}Ga_{0.05}As$ layer were grown. Approximately 1.7 monolayers of InAs QDs were deposited, followed by a 30-s annealing at 470 °C under an As4 flux at a base pressure of 3 $\times$ 10^{-6} Torr. To avoid indium evaporation during the growth of barrier at the high-temperature, we deposited a 10-nm low-temperature (470 °C)-grown $Al_{0.95}Ga_{0.05}As$ layer prior to the 40-nm-thick $Al_{0.95}Ga_{0.05}As$ barrier layer under As4 at a flux of 1 $\times$ 10^{-5} Torr. Finally, a $\sim$ 2-nm-thick GaAs capping layer was grown under an As4 flux at a base pressure of 1 $\times$ 10^{-5} Torr to prevent oxidation by the atmosphere. The substrate temperature was increased during the growth of the capping layer from 470 to 580 °C. The growth rates of InAs QDs, AlAs and GaAs (for the AlGaAs wetting layer) were 0.063 monolayers/s, 0.25 nm/s and 0.013 nm/s respectively. The growth rate of the GaAs capping layer was 0.2 nm/s. In order to confirm the size and the density distributions of QDs, we grow a separate InAs QD sample under the same growth conditions, but without a capping layer for AFM measurement. As seen in Fig. 1(a), the QDs are elongated along the [1$\bar{1}$0] direction due to the lateral strain fields originated from the lattice mismatch between AlGaAs and InAs [11–13].

For the temperature-dependent PL measurements, a 50-cm spectrometer with a 1200-line/mm grating equipped with a liquid-nitrogen-cooled charge-coupled device was used to record the PL spectra. The 532-nm line of a Nd-YAG laser was used as the excitation source for the PL measurements. To maintain stable excitation laser power during the long period of the temperature dependent PL measurements, we placed a laser power stabilizer (Thorlabs LCC3111H/M) in the path of the laser beam. To vary the sample temperature (5 K $\sim$ 300 K) during PL measurements, we placed the sample on the cold finger of a double-stage closed-cycle refrigerator. To avoid abrupt temperature change, instead of using a heater, we turned off the refrigerator at 5 K and naturally increased the temperature to room temperature. For the magneto-PL measurements, the sample was placed at the center of a 15-T superconducting magnet. A 1-mm-diameter multimode optical fiber was employed to carry external laser light to the sample and to bring the PL signal from the sample to a 25-cm spectrograph equipped with a CCD detector. The sample was immersed in a ^{4}He dewar to maintaine temperature at 4.2 K during the magneto-PL measurements. The PL spectra was taken in 0.1-T steps from 0 to 15 T.

III. RESULTS AND DISCUSSION

Figure 2 exhibits the PL transitions under magnetic fields from 0 to 15 T. At B = 0, the peak labeled as L centered at $\sim$ 1.3515 eV is associated with the excitonic recombination in the InAs QDs. The transition energy from the InAs QDs used for this study is significantly lower than the value previously reported by other groups (1.7 $\sim$ 1.8 eV) [14–18]. Because the QD size distribution and the shape of the sample used for this study are different from the others. The broad line-width of the spectrum reflects the ensemble of various QD sizes. To discuss the U-peak centered at $\sim$ 1.4866 eV, we had to consider that the InAs QDs had been capped with a low temperature (470 °C)-grown $Al_{0.95}Ga_{0.05}As$ matrix. A low-temperature-grown matrix contains numerous defects, hence, the U-peak is attributed to a recombination from the defect level in the AlGaAs matrix. We do not consider the effect of the $Al_{0.95}Ga_{0.05}As$ X-band because the energy level of our sample is significantly lower than the energy of the X-band.

With increasing magnetic fields, the L-peak changed neither its intensity nor the position of its peak energy. This means that the diamagnetic behavior of the excitons in the InAs QDs is similar to that of deeply bound excitons, which have heavy effective masses. The transition from the defects level (U-peak) shows spectral broaden-

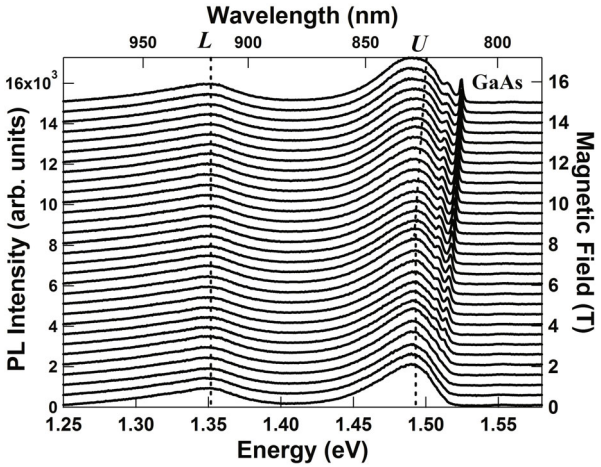

Fig. 2. PL spectra in magnetic fields. Low-temperature (4 K) magneto-PL spectra are displayed for magnetic fields from 0 to 15 T in 0.5-T steps. The L-peak does not change its spectral shape up to a magnetic field of 15 T whereas the U-peak shows a slight blue-shift with spectral broadening. At magnetic fields above 2 T, two new peaks, identified as being due to GaAs bulk free and GaAs donor bound excitons, emerge along the higher energy side of the U-peak.

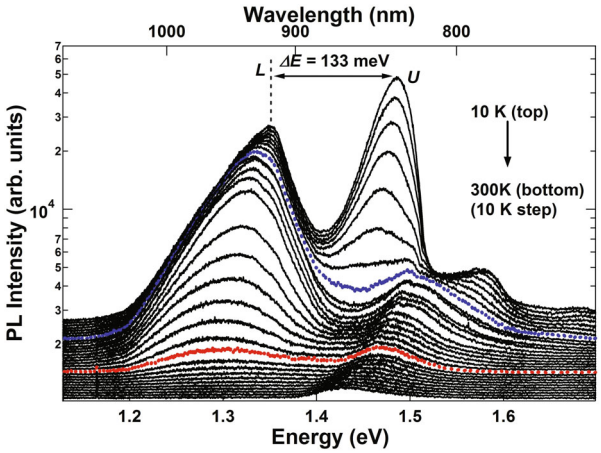

Fig. 3. (Color online) Temperature-dependent PL spectra from 10 K to 300 K in 10-K steps. The L-peak persists to 200 K (red line). However, the U-peak completely quenches at $\sim$ 70 K (blue line) and a new peak identified as being due to the GaAs bulk exciton emerges at the higher energy side of the U-peak and continues to 300 K.

ing at magnetic fields above 8 T and the position of the peak energy exhibits a small blue-shift. This is due to the fact that the carriers in the defect level have a relatively lighter effective mass than the carriers in the InAs QDs confined in a narrow region. At a magnetic field of 2 T, new transitions emerged on the higher energy side of the U-peak and showed diamagnetic shifts with further increasing magnetic fields. These transitions are identified as the GaAs bulk free exciton (FX) and the GaAs donor bound exciton (DX) from the GaAs substrate. The appearance of the transitions from the GaAs substrate can be explained in such a way that even though the substrate is blocked by AlGaAs barriers, external magnetic fields can strongly enhance the oscillator strength of the GaAs bulk excitons.

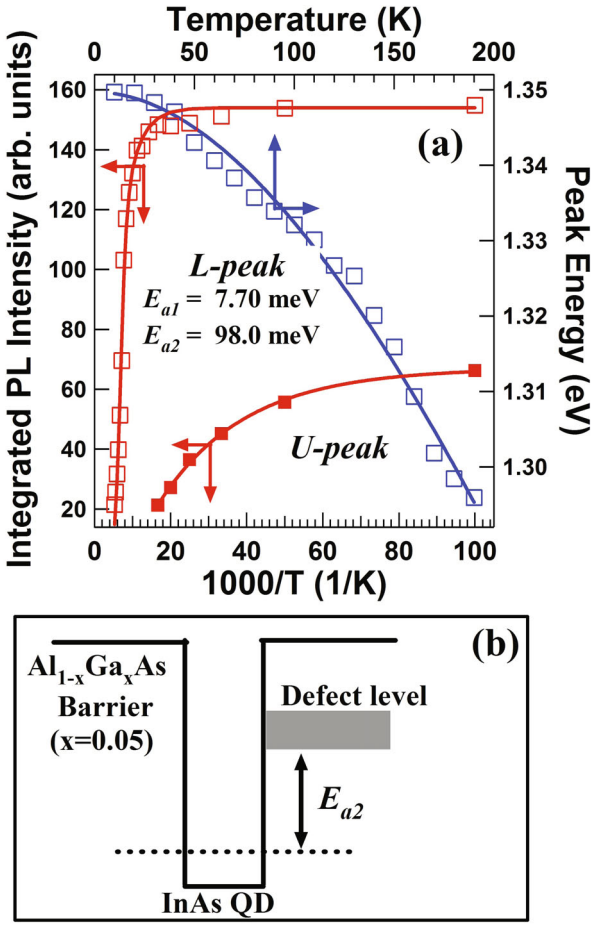

Fig. 4. (Color online) (a) Integrated intensity (left axis) vs. inverse temperature (bottom axis) and the peak energy position (right axis) vs. temperature (top axis). (b) Carriers in the QDs can transfer to a higher energy level in the AlGaAs defect.

In order to investigate the thermal quenching mechanism, we carried out PL measurements while increasing the temperature from 10 K to room temperature, as shown in Fig. 3. The peak energy separation between the L- and the U-peaks is about 135 meV at 10 K. With increasing temperature, the L-peak persists up to 200 K whereas the U-peak disappears quickly at $\sim$ 70 K. A new peak identified as the GaAs bulk free exciton appears at $\sim$ 1.5 eV (see blue-broken line for the 80-K spectrum). This GaAs free exciton survives to room temperature. Due to thermal broadening, the spectrum of the GaAs free exciton at a high-temperature is rather broader than the high magnetic field one for which the magnetic field induces wavefunction localization.

Figure 4(a) shows the temperature dependences of the

PL peak energy and the PL intensity of the L-peak. In the figure, the markers and the solid lines indicate the experimental and the fitted data, respectively. The peak transition energy follows the Varshni relation fitting ($E_g = E_0 - \alpha T^2/(T+\beta)$, solid line), which is typical behavior due to bandgap reduction with increasing temperature [19]. To analyze the thermal dissociation of InAs QD excitons, we attempted to fit the integrated intensity of the L-peak by using the two-step Arrhenius equation [20,21],

$$I = \frac{I_0}{1 + C_1 \exp(-E_{a1}/k_B T) + C_2 \exp(-E_{a2}/k_B T)}, \quad (1)$$

where I_0 is the areal intensity at 0 K, k_B is the Boltzmann constant, and C_i and E_{ai} (i=1, 2) are the ratio of the thermal escape to the recombination rates and activation energies, respectively . The low-temperature activation energy for the QD exciton transition ($E_{a1} \sim$ 7.7 meV) is attributed to the thermal dissociation of the QD excitons, which reflects the binding energy of the excitons formed in the InAs QDs. The high-temperature activation energy, ($E_{a2} \sim$ 98.0 meV) is related with the carriers in the QDs being thermally transferred to the higher energy levels in the defects. Figure 4(b) depicts the thermal transfer of the dissociated carriers in the QD to a higher energy level in the AlGaAs matrix . The two-step Arrhenius process indicates that with increasing temperature, initially the thermally disturbed exciton can be dissociated into free carriers. With further elevating temperature, the thermally dissociated carriers can transfer to higher energy levels in the AlGaAs defects, which leads to PL-intensity quenching. On the other hand, the U-peak intensity simply fitted by using an exponential function,

$$I = I_0 exp(-B/k_B T), \quad (2)$$

where B is fitting parameter. This means that the quenching mechanism of the U-peak is due to the simple thermalization induced by the increasing non-radiative recombination with increasing temperature.

IV. CONCLUSION

We measured the PL transitions from InAs QDs buried in an $Al_{0.95}Ga_{0.05}As$ matrix in high magnetic fields to 15 T at 4 K and the temperature dependence from 5 K to room temperature at B = 0. In the presence of a high magnetic field, the transition from the QD excitons labeled as L changes neither its intensity nor its transition energy at magnetic fields up to 15 T. This is due to the fact that the excitons formed in the QD act as deeply bound carriers with a heavy effective-mass. However, the transition from the defect level in the AlGaAs matrix, labeled as U exhibits spectral broadening and a slight blue-shift. When the temperature is increased from 10 K to room temperature, the U-peak quenches quickly around 70 K whereas the L-peak persist up to 200 K. The two-step Arrhenius equation indicates that the low-temperature activation energy for the L-peak, which reflects the exciton binding energy formed in the QD, is $\sim$ 7.7 meV. The high-temperature activation energy ($\sim$ 98.0 meV) is associated with the carriers that transfer from the QD levels to the defect level. Unlike the quenching mechanism of the L-peak, the quenching mechanism of the U-peak is attributed to the increasing rate of the non-radiative recombination with increasing temperature.

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