ANISOTROPIC RADIATIVE DECAY OF InP SELF-ASSEMBLED QUANTUM DOTS

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Strong optical anisotropy of the photoluminescence (PL) spectra and time resolved PL spectra is observed in InP self-assembled quantum dots (SADs) embedded in GaInP matrix. From the TEM study, the origin of the optical anisotropy appears to be due to the formation of composition modulation planes in GaInP matrix. We also studied the size dependence of the radiative decay time. In the vicinity of the PL peak from InP SADs, slowdown of the PL decay time was observed on decreasing the detection energy. It is found that the PL decay time becomes faster again at the lower energy side of the PL peak from InP SADs suggesting increase in the oscillator strength with increasing size.

Semiconductor self-assembled quantum dots (SADs) formed through the Stranski-Krastanov mode have recently attracted much interest from the view point of both fundamental physics and device applications. In order to clarify the electronic structures and the relaxation processes of the confined exciton, many kinds of optical measurements have been performed such as photoluminescence (PL), μ-PL, cathodoluminescence, Raman scattering, time resolved PL.[1−5] Since III-V compound SADs contain few structural defects, the PL decay time measurement would provide valuable information about the radiative decay time and the oscillator strength of the confined exciton.

In this short paper, we report the size and polarization dependence of the PL decay time in InP SADs. The origin of the optical anisotropy and the relaxation processes of the confined exciton are discussed.

InP quantum dots embedded in GaInP were grown on a Si-doped (100) oriented GaAs substrate by using a gas-source molecular beam epitaxy system.[6] An edge of the sample was glued on a sample holder and the sample was immersed directly into liquid He. The excitation light source was the SHG of Ti:sapphire laser ($\sim$3.1 eV). The PL is analyzed by a 50-cm single monochromator and detected by a charge coupled device camera. The

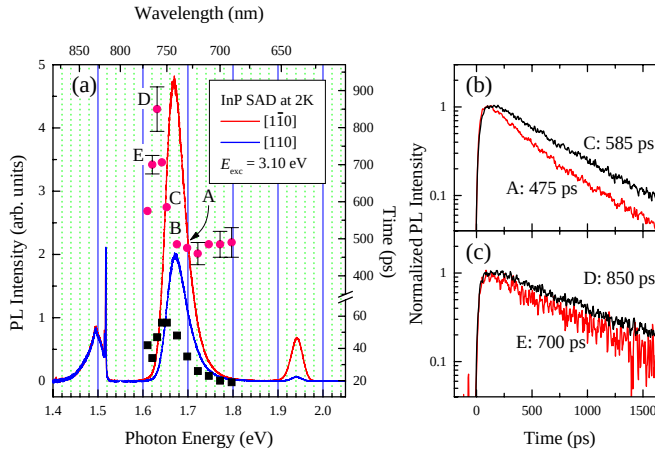

Figure 1. (a): Polarization dependence of PL spectra at 2 K under the band-to-band excitation observed for [1$\bar{1}$0] (red curve) and [110] (blue curve) polarization. Squares and circles denote the rise and decay time, respectively. (b) and (c): Time resolved PL decay curves observed at A:1.698 eV, C: 1.653 eV, D: 1.631 eV and E: 1.621 eV, which were measured without a polarizer. The symbols A to E correspond to those in Fig. 1(a).

measurement system of the PL decay time is composed of a 25-cm subtractive double monochromator and a streak camera. The time resolution of this system is better than 7 ps.

The red and blue curves in Fig. 1(a) show the PL spectra at 2 K observed along the [1$\bar{1}$0] and [110] directions of the GaAs substrate, respectively. The PL bands observed at 1.52 eV and 1.94 eV arise from the GaAs substrate and the GaInP matrix, respectively. The PL peak due to the radiative decay of the confined excitons in the SADs is observed at around 1.67 eV. The state filling effect of this PL peak is observed for the excitation powers above 20 nJ/cm^2. As pointed out by Castrillo *et al.*, the observed PL decay time of the excited state exciton becomes slower under the high power excitation, because if the ground state of the exciton is filled the excitons of the excited states cannot relax to the ground state.[2] In our experiments, in order to eliminate this effect and to observe the size dependence of the PL decay time in SADs, the PL decay measurements were performed under weak excitation ($\sim$7 nJ/cm^2).

The intensity of the PL peak coming from the GaAs substrate does not depend on [1$\bar{1}$0] and [110] polarizations because bulk GaAs has cubic (T_d) symmetry. The PL bands of the GaInP matrix and the InP SADs, however,

show distinct polarization dependence, although both the GaInP and InP are considered to have the same symmetry as GaAs. The PL spectrum was not sensitive to the polarization of the laser under the band-to-band excitation, since the memory of polarization is lost before the carriers excited in GaInP matrix relax into the quantum dots. The degree of polarization at the PL peak of InP and GaInP was about 40 % and 80 %, respectively.

From the TEM study, the origin of the optical anisotropy in GaInP matrix is considered to be due to the formation of composition modulation planes along the $[1\bar{1}0]$ direction.[6] In this case, since GaInP matrix has C_{2v} or lower symmetry, the optical anisotropy of the PL in GaInP is observed. The PL spectra from SADs embedded in an anisotropic matrix would show polarization dependence because of anisotropic strain.

In order to study the size dependence of the radiative decay time, the time resolved PL curves are obtained over a wide energy range around the PL peak of the InP SADs. The typical behavior of the unpolarized PL decay curve is shown in Fig. 1(b) and 1(c). Each PL decay curve is normalized at the peak. The PL decay time τ_d and the rise time τ_r are estimated by fitting the observed curve as $I(t) = [1 - \exp(-t/\tau_r)] \exp(-t/\tau_d)$. τ_d and τ_r thus obtained are shown in Fig. 1 by circles and squares, respectively. The PL decay time observed above 1.7 eV is about 480 ps. With the decrease of the detection energy, the PL decay time increases [A→C in Fig. 1(a) and 1(b)], and has the maximum value at 1.631 eV [D in Fig. 1(a) and 1(c)]. This result indicates that the PL decay time increases as the size of the quantum dots increases. In order to explain the results qualitatively, we might need to consider the relaxation (i) from the higher energy states to the ground state, and/or (ii) from small size SADs to large ones as well. The decay time decreases again on decreasing the detection energy [D→E in Fig. 1(a) and 1(c)]. This appears to be related to the theoretically expected increase in the oscillator strength with increasing size.[7]

The polarization dependence of the PL decay time was studied around the PL peak of the InP SADs as shown in Fig. 2, and clear anisotropy of the PL decay time was observed at the PL peak and at the higher energy side of the PL peak. The PL decay curve observed at the lower energy side of the PL peak was almost the same for both polarizations [curve C in Fig. 2]. With the increase of the detection energy, the polarization dependence of the PL decay time becomes stronger [curve C→curve A in Fig. 2]. The PL decay time observed for $[1\bar{1}0]$ polarization at 1.698 eV [curve A] is about 40 % faster than that observed for [110] polarization.

A clear polarization dependence of the PL decay time in GaInP matrix is also observed (Figure is not shown here). At the PL peak of GaInP, the PL

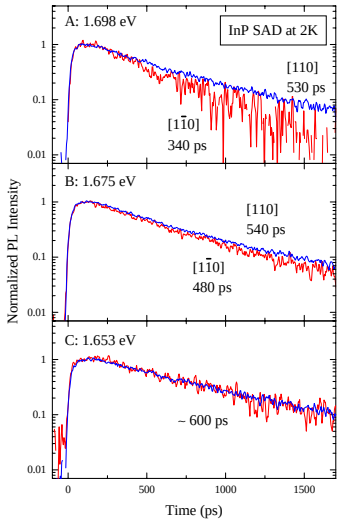

Figure 2. The time resolved PL curves observed at A: 1.698 eV, B: 1.675 eV and C: 1.653 eV. Data shown by the blue and red curves were observed for [110] and [1$\bar{1}$0] polarization, respectively. The symbols A, B and C correspond to those in Fig. 1(a). With the increase of the detection energy [curve C→curve A], the anisotropy of the PL decay time becomes clear.

decay time was 620 ps for [1$\bar{1}$0] polarization and 790 ps for [110] polarization. Both the GaInP matrix and the InP SADs show faster decay when observed for [1$\bar{1}$0] polarization than for [110] polarization. This explains why the PL observed for [1$\bar{1}$0] polarization shows stronger intensity.

Acknowledgments

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