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Luminescence properties of InAs quantum dots formed by a modified self-assembled method

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The luminescence properties of self-assembled InAs quantum dots (QDs) on GaAs (1 0 0) substrates grown by molecular beam epitaxy have been investigated using temperature-dependent photoluminescence (PL) and time-resolved PL (TRPL). InAs QDs were grown using an In-interruption growth technique, in which the indium flux was periodically interrupted. InAs QDs grown using In-interruption showed reduced PL linewidth, redshifted PL emission energy, increased energy level spacing between the ground state and the first excited state, and reduced decay time, indicating an improvement in the size distribution and size/shape of QDs.

1. Introduction

Self-assembled InAs quantum dots (QDs) have been studied intensively for various applications such as laser diodes, light-emitting diodes, detectors, and optical amplifiers. The atomic-like density of states and localized wave functions of QDs are particularly attractive characteristics for applications in lasers due to the reduced threshold current density, high differential gain, and high temperature stabilization [1–4]. One of the important QD parameters for the optimization of laser design is radiative lifetime, which depends on the QD size [5]. In order to improve and control the size and shape, density, and alignment of QDs, various efforts have been performed on self-assembled InAs QDs, for example, by inserting InGaAs or InAlAs strain reducing layer (SRL) and using In- and/or As-interruption technique during QD growth [6–11]. In our previous work [7], the optical properties of InAs QDs grown on InP substrates were improved by introducing an $In_{0.32}Ga_{0.68}As$ layer between the InAs QD layer and the InAlGaAs layer. However, the InGaAs layer can result in an increase of the temperature sensitivity of the QD layer because of the reduced energy barrier. Thus InAlAs SRL has been used to enhance the potential barrier and to increase the energy separation between the ground state and first excited state, which can lead to have good temperature stability [8,9]. InAs QDs grown by the In-interruption technique have resulted in good lateral self-alignment as well as improved optical properties [10].

In this paper, we present the optical characteristics of self-assembled InAs QDs grown on GaAs substrates by the In-interruption growth technique. The luminescence and carrier dynamics of InAs QDs as functions of temperature and emission wavelength have been studied using photoluminescence (PL) and time-resolved PL (TRPL) measurements.

2. Experimental details

InAs QD samples were grown by molecular beam epitaxy on semi-insulating GaAs (1 0 0) substrates. After depositing a 300 nm-thick GaAs buffer layer at 580 °C, InAs QDs were grown at 480 °C by the In-interruption growth technique. During QD growth, the In flux was periodically interrupted by In shutter whereas the As flux was supplied continuously. The shutter for the In source was opened for 1 s and then closed for either 9, 19, 29, or 39 s. This growth sequence was repeated 30 times during the growth of InAs QDs. The samples grown with In-interruption times of 9, 19, 29, and 39 s are denoted as S09, S19, S29, and S39, respectively. For comparison, an InAs QD sample without In-interruption was grown in a pure GaAs matrix for 30 s. This sample is denoted as S00. The total amount of In supplied for all InAs QD layers was the same (30 s) but the total growth time for each sample was varied.

PL and TRPL measurements were performed using an Edinburgh Instruments FLS 920 spectrometer at the Central lab of Kangwon

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National University. The luminescence signal was analyzed in a 0.30 m monochromator and detected by a near-infrared photomultiplier tube (NIR-PMT) detector. The InAs QD samples were excited using a cw diode laser (λ=532 nm) for PL, and a picosecond pulsed diode laser (λ=635 nm, pulse width=93 ps) for TRPL. The luminescence decays were measured by a time-correlated single photon counting system. The PL and TRPL were measured at temperatures ranging from 10 to 300 K.

3. Results and discussion

Fig. 1 shows the low temperature (10 K) PL spectra of the InAs QD samples grown by In-interruption growth technique. The PL spectra for all QD samples mainly consist of two peaks, which are attributed to the ground state (GS) and the first excited state (1ES) transitions. The emission peak positions and the values of the full-width at half maximum (FWHM) were determined using Gaussian peak fitting. As the In-interruption time increases from 0 to 19 s, the PL peak position of the GS (1ES) for the QD samples is redshifted by about 102 nm (79 nm) from 1096 (1055) to 1198 nm (1134 nm) and the energy-level spacing between the GS and the 1ES increases from 44 meV for S00 and S09 to 58 meV for S19. With further increasing In-interruption time from 19 to 39 s, however, the PL peak positions of both the GS and the 1ES are almost constant, but the PL intensities for samples S29 and S39 show an increase of ~30% and a decrease of ~60%, respectively, compared to that of sample S19 as shown in Fig. 1. Sample S09 shows the strongest PL intensity which is about 5 times stronger than the intensity of sample S39. The FWHM of the GS peak is reduced from 40 meV for S00 to 25 meV for all samples grown by In-interruption. The reduced FWHM and the redshift of the PL transition energies indicate that the size distribution, size and shape of the QDs are effectively improved by In-interruption during QD growth. Wang et al. [11] reported similar results for the effect of growth interruption on self-assembled InAs/GaAs islands. The redshift of the PL peak for InAs islands with 1.7 monolayer coverage was explained by InAs material transfer from the wetting layer (WL) into the formed islands during growth interruption, leading to an enlargement of the islands. In addition, the increase of the energy-level spacing between the GS and the 1ES can also be explained by the improved quantum effects resulting from the better aspect ratio (height/diameter) of the QDs grown by In-interruption.

The size and the surface morphology of the InAs QD grown by In-interruption technique were investigated using cross-sectional transmission electron microscope (TEM) and atomic force microscopy (AFM) measurements, respectively (not shown here) [10]. The height and lateral size (diameter) of the QD increased with increasing In-interruption time from 0 to 29 s. Bright and dark regions in the AFM images were observed for all samples. The height of the bright region was approximately 0.511 nm higher than that of the dark region. Joyce et al. [12,13] reported that two terraces were observed in the scanning tunneling microscopy images, and the WL had the rather disordered appearance characteristic of a (1×3) reconstruction, a characteristic of $In_xGa_{1-x}As$ alloy formation in the 2D growth of InAs on GaAs substrate [14,15]. The AFM images showed that the QDs grown by In-interruption technique were located in the boundary between the bright and dark regions, which consisted of InGaAs alloy formed from 2D growth of InAs on GaAs substrate [12–15]. This self-alignment of QDs can be explained by the migration of In sources to a suitable site (step edge) during the interruption time, resulting in the increase of QD size. On the other hand, the QDs for S00 were grown randomly on all regions because of a lack of migration time for the In. For S00, In sources were continuously supplied during the growth time of 30 s, and thus contribute immediately to QD formation in all regions. The results of TEM and AFM measurements [10] clearly showed improved size and size distribution of QDs grown using In-interruption, which are consistent with the PL results shown in Fig. 1.

The PL peak energies of the GS for all InAs QDs as a function of temperature are shown in Fig. 2. The solid curves in Fig. 2 are the transition energies calculated using Varshni's equation [16]; $E_g(T)=E_{g0}-\alpha T^2/(\beta+T)$, where E_{g0} is the bandgap energy of InAs at 0 K, and α and β are empirical parameters. The solid curves calculated using parameters [17] (E_{g0}=0.417 eV, α=0.276 meV/K, and β=93 K) are shifted to the energy of the GS at 10 K. While the

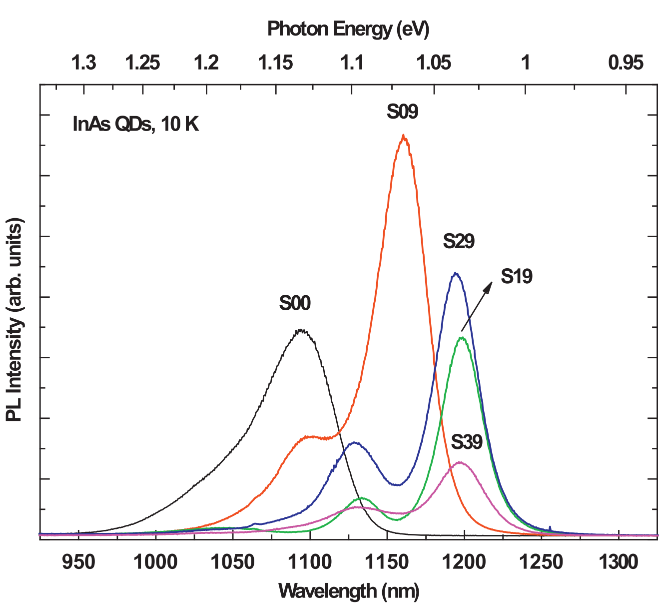

Fig. 1. Low temperature (10 K) PL spectra of InAs QD samples grown by In-interruption growth technique. In-interruption time for S00, S09, S19, S29, and S39 is 0, 9, 19, 29, and 39 s, respectively.

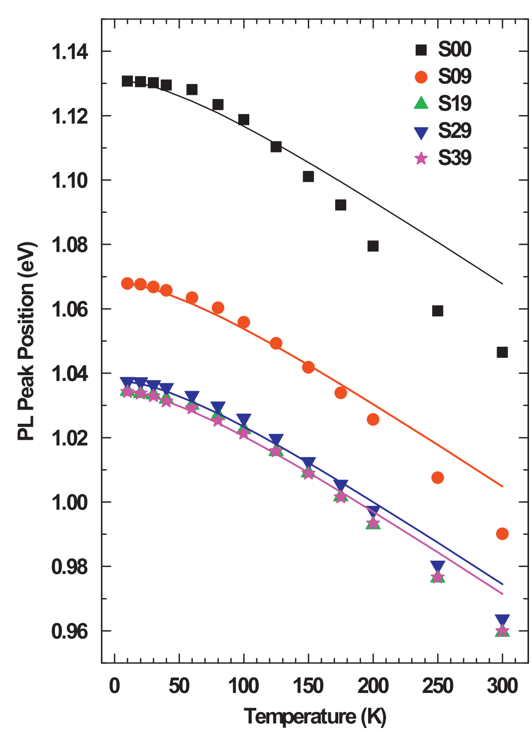

Fig. 2. Measured PL peak energies for InAs QD samples at various temperatures. The solid curves are the bandgap energies calculated using Varshni's equation.