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Kita, Takashi
Masuda, Yoshitaka
Mori, Takayoshi
Wada, Osamu

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Long-wavelength emission from nitridized InAs quantum dots

Takashi Kita,[a)] Yoshitaka Masuda, Takayoshi Mori, and Osamu Wada
Department of Electrical and Electronics Engineering, Faculty of Engineering, Kobe University, Rokkodai 1-1, Nada, Kobe 657-8501, Japan

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A technique to grow InAs quantum dots (QDs) to extend the emission wavelength into 1.3 μm range has been developed. We performed nitridation after growing InAs QDs by molecular-beam epitaxy. During nitridation, the reflection high-energy electron diffraction keeps chevron patterns, as well as streak rods, coming from the wetting layer. A longer-wavelength emission line with a narrower spectral linewidth compared with those of InAs QDs has been observed. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1627943]

Semiconductor quantum dots (QDs), such as an InAs QD, are a promising material for laser diodes, because the laser characteristics can be improved dramatically due to their atomlike density of states.[1–3] Continuous-wave laser action of the QD laser diode has been achieved around 1 μm. To extend the emission wavelength into between 1.3 and 1.55 μm, several kinds of approaches, such as stacking,[4–6] alternating deposition,[7] and strain reduction by a capping layer,[8,9] have been proposed. On the other hand, recently, nitride (N)-containing III–V semiconductors have attracted much interest for long-wavelength laser material grown on GaAs substrates,[10] because of the large band-gap bowing parameters of 18 eV for GaNAs (Ref. 11) and 4.2 eV for InNAs.[12,13] By utilizing this characteristic, GaInNAs QDs have been fabricated to achieve between 1.3 and 1.55 μm room-temperature emission.[14] However, with increasing N concentration, the spectral linewidth becomes large and a reduction occurs in the photoluminescence (PL) intensity. We have demonstrated QD growth by using nitraidation of the InAs QDs. The nitridized InAs QDs show a longer-wavelength emission line with a narrower spectral linewidth compared with that of InAs QDs.

Self-assembled InAs QDs were grown on GaAs(001) substrates by a solid-source molecular-beam epitaxy (MBE). Active N species were created in radio-frequency (rf) plasma source from ultrapure N_2 gas. The nitrogen partial pressure was 5×10^{-5} Torr. The rf power was set at 230 W. A 120-nm-buffer layer of GaAs was grown at 530 °C. InAs deposition was performed at 450 °C. The deposition rate of InAs was 1 monolayer (ML)/min, which was estimated from the reflection high-energy electron diffraction (RHEED) oscillations during the homoepitaxial growth. The transition from two-dimensional (2D) wetting layer growth to three-dimensional island growth is determined from the change in the RHEED pattern. At the growth transition, the RHEED pattern exhibits chevron patterns representing the QDs and streak rods coming from the wetting layer. The amount of the deposited InAs at the growth transition is about 1.8 MLs. After growing the InAs QDs, we performed nitridation of the QDs in 8 s. The 8 s irradiation of the activated N species corresponds to the amount to prepare the (3×3) nitridized surface of the GaAs(001). The RHEED patterns in the [−110] azimuth before and after nitridation of the InAs QDs are shown in Figs. 1(a) and 1(b), respectively. During the nitridation, the RHEED patterns keep the chevron patterns and the streak rods. This result shows that the surface morphology does not have any damage by the nitridation process. After the nitridation, the sample was capped by a 50-nm-thick GaAs layer. The substrate temperature was kept at 450 °C during the nitridation and the following GaAs cap growth. To compare the emission property, we grew a sample with a 2D InAs thin film instead of the InAs QDs by using the same process.

PL measurements have been performed to characterize the nitridized InAs QDs. The sample was excited by the 488 nm line of an Ar-ion laser. The PL signal dispersed by a 320-mm-single monochoromator was detected by a liquid-nitrogen-cooled InGaAs array.

Figure 2 shows PL spectra at 4 K for QDs [Fig. 2(a)] before and [Fig. 2(b)] after nitridation. The InAs QDs shows a broad PL at 1.05 μm, which is the same as data reported in the previous articles. The 830 nm emission comes from GaAs. After nitridation, the QDs emit a relative narrow-band PL at 1.18 μm together with a broad signal at 1.06 μm. The sample with the nitridized 2D InAs shows a PL peak at 1.06 μm. The result is shown in Fig. 2(c). The PL wavelength of Fig. 2(c) coincides with the broad signal observed in Fig. 2(b). Thus, the 1.06 μm signal in Fig. 2(b) corresponds to the PL from the nitridized 2D InAs. Since nitridation partially exchanges As atoms by N, the InAs wetting layer with less than 2 MLs thickness is expected to become an InNAs wetting layer. On the other hand, the longer-wavelength emission line at 1.18 μm in Fig. 2(b) can be attributed to the

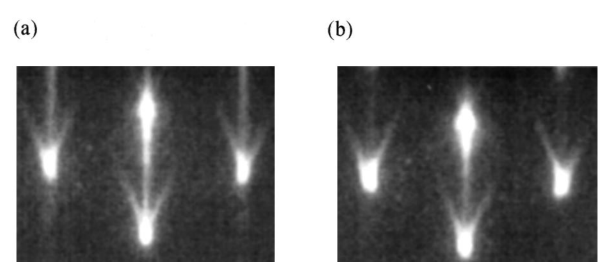

FIG. 1. RHEED patterns in the [−110] azimuth before (a) and after (b) nitraidation of InAs QDs.

[a)]Electronic mail: kita@eedept.kobe-u.ac.jp

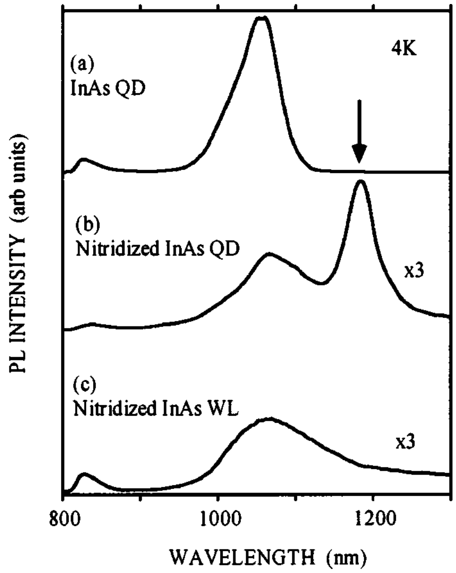

FIG. 2. PL spectra of InAs QDs (a), nitridized InAs QDs (b), and nitridized InAs wetting layer (c).

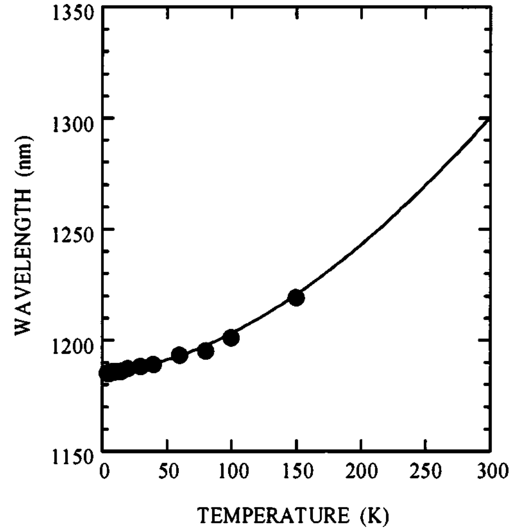

FIG. 3. Temperature dependence of PL peak wavelength for nitridized InAs QDs. The solid line indicates a caluculated result according to the Varshni empirical relation.

nitridized InAs QDs. The spectral linewidth is narrower than that of the InAs QDs.

Two origins for the longer-wavelength emission can be considered; (1) N atoms are doped uniformly into InAs QDs by the nitridation, i.e., formation of InNAs QDs, and (2) a nitridized thin layer covering the InAs QD acts as a strain-reducing layer. The transition energy of the InNAs QD is reduced by the large band-gap bowing parameters. The observed redshift of the QD PL by the nitridation is about 120 meV. If we use the previously reported band-gap change for InNAs (Ref. 13) and ignore a change in the QD size by the nitridation, the nitrogen concentration x in InN_xAs_{1-x} can be roughly estimated to be around 0.03. On the other hand, if nitridation only takes place at the QD surface and a resulting thin InNAs thin layer is formed, the thin layer can reduce the internal compressive strain in the InAs QDs when $a_{InAs} > a_{InNAs} > a_{GaAs}$, where a_{InAs}, a_{InNAs}, and a_{GaAs} are the lattice constants of InAs, InNAs, and GaAs, respectively. The strain reduction in the QD reduces the transition energy.[8,9]

The temperature dependence of the PL-peak wavelength of the nitridized InAs QDs is plotted in Fig. 3. The PL intensity rapidly decreases above $\sim$200 K. The reason is not yet clear. However, our nonannealed sample is considered to have nonradiative centers caused by crystal defects as reported for conventional GaInNAs.[15,16] Although the PL intensity becomes weak above $\sim$200 K, the wavelength data extrapolated to room temperature are close to 1.3 μm. The band-gap difference between 4 and 150 K, $\sim$30 meV, is almost the same as values for bulk InAs and GaAs. Since the N-containing InAs has a small temperature coefficient as compared with the bulk InAs, the observed bulklike temperature dependence of the band gap excludes the possibility of InNAs QDs. Therefore, the formation of the InNAs thin layer covering the InAs QDs seems to be the more probable reason for the longer-wavelength PL.

Based on this discussion, let us consider the narrowing of the PL bandwidth. Since the surface/volume ratio of the QD is larger for smaller size QDs, the internal compressive strain in the smaller size QDs is reduced by the InNAs thin layer in a more pronounced way. Therefore, PL signal from the smaller size QDs shifts significantly toward the longer-wavelength side. On the other hand, the smaller size QD has the higher transition energy. Thus, the significant redshift in the smaller size QDs with the higher transition energies causes narrowing of the PL bandwidth.

In summary, we have demonstrated growth of InAs QDs by using nitridation in MBE. A-longer wavelength emission line with a narrower spectral linewidth compared with those of InAs QDs has been observed. These results suggest that the developed growth technique of nitridized InAs QDs can be applicable to optical communication wavelength lasers.

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