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Improvement of InAs quantum-dot optical properties by strain compensation with GaNAs capping layers

X. Q. Zhang,[a] S. Ganapathy, I. Suemune,[b] H. Kumano, and K. Uesugi
Research Institute for Electronic Science, Hokkaido University, Kita-12, Nishi-6, Sapporo 060-0812, Japan and CREST, Japan Science and Technology Corporation, Japan

Yoichi Nabetani and Takashi Matsumoto
Department of Electrical Engineering, University of Yamanashi, 4-3-11 Takeda, Kofu, Yamanashi 400-8511, Japan

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Two kinds of self-assembled InAs quantum dots (QDs) grown on GaAs (001) substrates were studied. One is capped with GaAs layers and the other with GaNAs strain-compensating layers. Photoluminescence (PL) measurements on the two kinds of InAs QDs showed distinct dependence on the selection of the capping layers. The homogeneity and luminescence efficiency of the InAs QDs were much improved when the net strain was reduced with GaNAs layers. These results demonstrate the importance of net strain compensation for the improved optical quality of InAs QDs. [DOI: 10.1063/1.1629803]

InAs quantum dots (QDs) have been intensively investigated, and practical applications such as semiconductor lasers[1] and optical amplifiers[2] are being developed. Single-photon emitters (SPEs) based on QDs are another distinct application which utilizes the intrinsic nature of QDs.[3] In the applications to SPEs for quantum cryptography or quantum computation, the quantum efficiencies of SPEs in generating single photons and transferring them to receivers are one of the key issues. In this regard, the optical qualities or quantum efficiencies of InAs QDs are important. As is well known, InAs QDs are self-assembled on GaAs surfaces with the Stranski-Krastanow (SK) growth mode. This naturally induces residual compressive strain within the QDs. Capping InAs QDs with GaAs layers extends the compressive strain into the capping layers, which makes the spatial alignment of the QDs grown in the neighboring layers possible.[4] The relation of the compressive strain and the luminescence peaks of the InAs QDs has been extensively studied.[5,6] On the other hand, application of tensile-strained GaNAs capping layers to InAs QDs was recently proposed,[7,8] which led to longer wavelength emission up to 1.55 μm. The reduction of the compressive strain around InAs QDs with tensile-strained GaNAs capping layers was identified with transmission-electron-microscopy cross-sectional observations.[7,9] The purpose of this paper is to study how optical properties such as luminescence efficiencies and homogeneity of the QDs depend on the selection of the capping layers. The role of the capping layers in the optical properties of InAs QDs was studied by comparing two kinds of InAs QDs: One is InAs QDs capped with conventional GaAs layers and the other is QDs capped with GaNAs strain-compensating layers (SCLs).[7] It will be demonstrated that the InAs QDs capped with GaNAs SCLs show much improved optical properties. These observations are discussed from the viewpoint of net strain compensation with GaNAs SCLs.

Self-assembled InAs QDs were grown on GaAs (001) substrates by metalorganic molecular-beam epitaxy. The precursors used were triethyl gallium (TEGa), triethyl indium (TEIn), trisdimethylaminoarsenic (TDMAAs), and monomethylhydrazine (MMHy) with the corresponding beam equivalent pressures (BEP) of 8×10^{-4}, 8×10^{-5}, 3×10^{-3}, and $(5-20)\times10^{-3}$ Pa. 100-nm-thick GaAs buffer layers were grown on GaAs substrates at 550 °C and then the substrate temperature was lowered to 400 °C. About 2 monolayers (MLs) of InAs were deposited at an average growth rate of 0.1 ML/s with a pulsed supply of TEIn and a continuous supply of TDMAAs. Transition from the two-dimensional growth mode to the SK growth mode was confirmed with reflection high-energy electron diffraction observations by the change from streaky to spotty patterns. Atomic force microscopy observations of InAs QDs without capping showed high QD density of $\sim1\times10^{11}$ cm^{-2}. The average dot height was about 3 nm and the average base diameter was about 25 nm. After the growth of the first stack of InAs QDs, a 10-nm-thick GaAs capping layer or a 10-nm-thick GaNAs SCL with different nitrogen (N) concentrations (0–2.7%) was grown. Subsequently, an additional 10-nm-thick GaAs layer was grown to initialize the surface for the growth of the second stack of InAs QDs. Two stacks of InAs QDs were grown following this procedure to check the homogeneity of the QDs in the neighboring stacks. The N concentration in the GaNAs layers was estimated with symmetric (004) and asymmetric (224) high-resolution x-ray diffraction measurements to evaluate the elastic distortion of the layers,[10] and with the assumption of Vegard's law between the lattice constants of the constituent binary compounds. The photoluminescence (PL) properties of the two-stacks of InAs QDs were measured with the excitation of a second harmonic of a yttrium aluminum garnet (YAG) laser at the wavelength of 532 nm between 18 K and room temperature in a closed-cycle He cryostat.

[a]Present address: Institute of Optoelectronic Technology, Northern Jiaotong University, Beijing 100044, People's Republic of China.
[b]Author to whom correspondence should be addressed; electronic mail: isuemune@es.hokudai.ac.jp

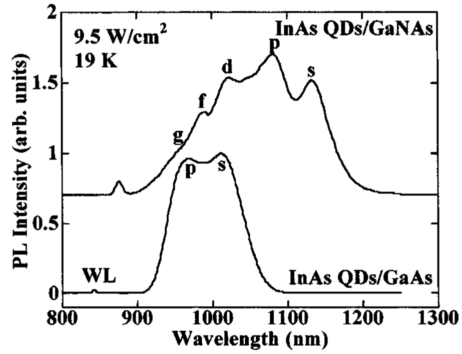

FIG. 1. PL spectra measured with the excitation intensity of 9.5 W/cm^2 at 19 K on InAs QDs capped with $GaN_{0.007}As_{0.993}$ and GaAs.

The PL spectra were measured on the two kinds of InAs QDs. One is the two-stacks of InAs QDs embedded in GaAs (InAs QDs/GaAs sample), and the other is the two-stacks of InAs QDs embedded in the GaNAs SCL with a N concentration of 0.7% (InAs QDs/GaNAs SCL sample). In spite of the high QD density of $\sim 1 \times 10^{11}$ cm^{-2}, the subpeaks originating from QD energy states were clearly observed as shown in Fig. 1. The two spectra were measured under the same excitation intensity. The observation of higher excited states in the InAs QDs/GaNAs SCL sample demonstrates that the band filling effect is enhanced by capping InAs QDs with GaNAs SCLs. This suggests a smaller contribution of nonradiative recombinations in the InAs QDs/GaNAs SCL sample.

The temperature dependence of the subpeaks in the two samples, of which the peaks were estimated by Gaussian fitting of the observed PL spectra,[8] is shown by the closed circles in Fig. 2. As a reference, the temperature dependence of the GaAs energy gap[11] is also shown in Fig. 2 by shifting the energy by 0.212 eV for display purposes. The lowest-state peaks denoted as "s" in the two cases shows the distinct difference in their temperature dependences. The one for the InAs QDs/GaAs sample shown in Fig. 2(a) exhibited an S-shaped temperature dependence, while the one in the InAs QDs/GaNAs SCL sample shown in Fig. 2(b) exhibited a monotonic temperature dependence. The S-shaped temperature dependence has been reported with GaInN/GaN multiple quantum wells (QWs),[12] a GaInNAs/GaAs single QW (Ref. 13, and references therein), and it was explained by carrier localization in inhomogeneously broadened systems. The PL linewidth of the "s" state in the InAs QDs/

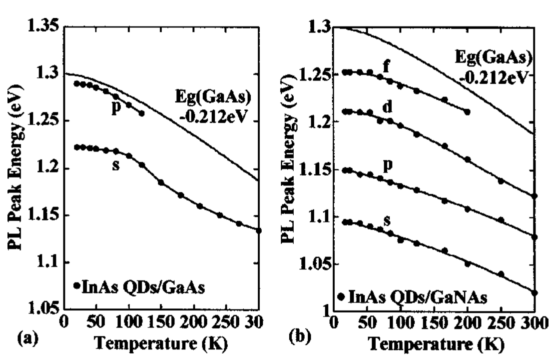

FIG. 2. Temperature dependence of the subpeak energies in the two samples: (a) InAs QDs capped with GaAs and (b) InAs QDs capped with $GaN_{0.007}As_{0.993}$.

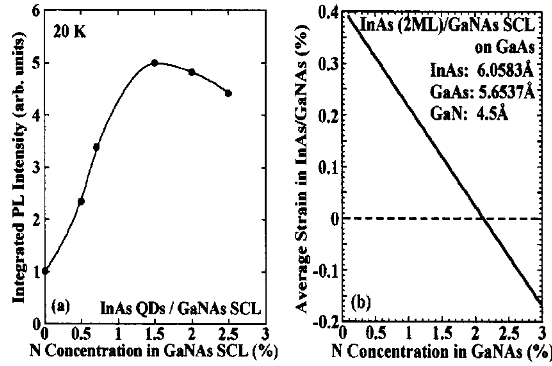

FIG. 3. (a) Integrated PL intensities of the InAs QDs/GaNAs SCL samples plotted against the N concentrations in the GaNAs capping layer. (b) Estimated average strain in the InAs/GaNAs system grown on GaAs substrates.

GaAs sample showed a minimum at around 150 K (not shown) where the redshift of the emission peak was more enhanced, in agreement with the observations in earlier work.[12] This phenomenon was explained with carrier localization in disordered systems at the lower temperature and carrier hopping among inhomogeneously distributed QDs at the higher temperature.[14] The PL linewidth of the "s" state in the InAs QDs/GaNAs SCL sample showed a monotonic temperature dependence (not shown) in accordance with the monotonic temperature dependence of the emission peak shown in Fig. 2(b).

The observed difference in the temperature dependence of the "s" lowest-state emission of the InAs QDs demonstrates that the "disordering" of the InAs QDs is enhanced with the GaAs capping while the "disordering" is reduced by capping with the GaNAs SCLs.

InAs QDs/GaNAs SCL samples with different N concentrations were also prepared, and the observed integrated PL intensities are plotted against the N concentrations in Fig. 3(a). The PL intensity was increased with an increase of the N concentration in the GaNAs SCL capping layers, and the increase was up to five times that observed with the GaAs capping layer, where the maximum increase was observed with a N concentration of ~1.5%. To understand the observed N concentration dependence of the PL intensity, the average strain in one period of the InAs QDs/GaNAs SCL was calculated. The lattice constants of InAs, GaAs, and cubic GaN used were 6.0583, 5.6537, and 4.50 Å, respectively. The average strain in the InAs/GaNAs system grown on GaAs substrates was evaluated by calculating the average lattice constant in one stack of the 10-nm-thick GaNAs layer and the InAs QDs layer with a simple approximation of the InAs QDs and the wetting layers with a 2-ML-thick uniform InAs layer. As shown in Fig. 3(b), the average compressive strain induced by the InAs "layer" is compensated by the increase of N concentration in the GaNAs SCL, and the average strain is reduced to zero near a N concentration of 2.1%. Although the details of the QD structures were not included in the present calculation, the improvement of the luminescence efficiency and the reduction of the average strain in the system show reasonable correlations with each other.

To study further the dependence of the InAs QD optical properties on the capping layers, the luminescence from each QD state transition was separated from the observed overall

TABLE I. Comparison of thermal activation energy of each transition and the corresponding PL peak separations for InAs QDs capped with $GaN_{0.007}As_{0.993}$ and GaAs. E_a is the thermal activation energy, E_B is the band gap energy of the $GaN_{0.007}As_{0.993}$ barrier, and E_{WL} is the PL peak from the InAs wetting layers.

		f (meV)	d (meV)	p (meV)	s (meV)
InAs QDs/$GaN_{(0.7\%)}As$	E_a	84	112	157	222
	$\Delta E_i = E_B - E_i$	77	119	181	236
	$E_B = 1.33$ eV				
InAs QDs/GaAs	E_a	—	—	83	165
	$\Delta E_i = E_{WL} - E_i$	—	—	140	208
	$E_{WL} = 1.43$ eV				
InAs QDs/GaAs	E_a	—	—	—	105
	$\Delta E_i = E_{WL} - E_i$	—	—	—	120
(Ref. 15)	$E_{WL} = 1.307$ eV				

spectra with the Gaussian fitting method discussed in Ref. 8. The thermal activation energy of each transition was estimated from the Arrhenius plots of the integrated PL intensities. The estimated thermal activation energies E_a and the relation to the energy differences of the QD state PL subpeaks with the GaNAs barrier (or wetting layers) are summarized in Table I. E_B for the InAs QDs/$GaN_{0.007}As_{0.993}$ sample is the band gap energy of the $GaN_{0.007}As_{0.993}$ barrier.[8] The reasonable agreement between the estimated thermal activation energies and the energy differences between the QD state PL subpeaks and the GaNAs barrier shows that the quenching mechanism of the PL intensities is the thermionic emission of carriers from the QD states to the GaNAs barriers, followed by carrier migration in the barrier layers and final nonradiative recombination. In this case, the thermal activation energies are given by the energy difference between the barrier energy and the corresponding QD state transition energies following the Maxwell-Boltzmann statistics of the populated carriers.

A similar comparison was made for the InAs QDs/GaAs sample and the results as well as the similar report from Ref. 15 are also shown in Table I. E_{WL} is the PL peak energy observed from the InAs wetting layers, and the energy separations of the PL subpeaks and the wetting layers were compared with the thermal activation energies. The InAs QDs/GaAs sample as well as the similar report of Ref. 15 showed lower activation energies than the PL subpeak energy separations. This suggests that capping InAs QDs with GaAs induces additional nonradiative recombination centers, which lower the thermal activation energies.

The above observations show that InAs QD homogeneities and luminescence quantum efficiencies are improved with a reduction of the average strain in the materials system by compensating the compressive strain induced by the InAs QDs with tensile strain in the GaNAs capping layers. Similar improvements of the optical properties with net strain compensation have been reported for strained quantum wire structures.[16] On the other hand, improved InAs QDs homogeneity has also been reported by capping with GaInAs layers.[17] The basic idea in capping InAs QDs with GaInAs layers is the reduction of the interface mismatch between InAs and GaInAs. But there remains the problem in this method that net strain remains in the system, and multiple-stacking of the InAs QDs layers causes degradation of crystalline quality due to the strain accumulation. The present method of capping InAs QDs with tensile-strained GaNAs reduces the net strain in the materials system, and the capping layer dependence discussed in this paper would be more pronounced with a larger number of stacking InAs QD layers. As for the interface strain between InAs and GaNAs, it will increase with increase in the N concentration in GaNAs, and the excess interface strain will degrade the QD optical properties. This is the reason why the PL intensity in Fig. 3(a) starts to decrease with a lower N concentration than that calculated to be optimum to realize the zero average strain in Fig. 3(b).

The actual strain distribution around the InAs QDs is more complex.[18] One of the authors (Y.N.) has been investigating the strain field within the framework of an atomic scale calculation by the valence-force field model, and a difference of the strain distribution around QDs with and without capping layers was identified. However, it is not easy to discuss the homogeneity and the luminescence efficiency of the InAs QDs dealt with in this paper with these numerical strain calculations, since a consideration of the average strain needs the calculation of a large three-dimensional volume including the GaNAs layers, and this makes the numerical calculation time overwhelming.

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