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Express Letter

Improvement of Optical Properties of Air-Exposed Regrowth Interfaces Embedded in InAs Quantum Dots and GaAs/AlGaAs Quantum Wells by Atomic Hydrogen

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We report an atomic hydrogen cleaning effect on surface oxide and contaminants on airexposed interfaces embedded in GaAs quantum well (QW) and InAs quantum dot (QD). A partly grown GaAs QW and a GaAs buffer layer for InAs QDs were air-exposed and hydrogen cleaned. After this procedure, we directly regrew a GaAs QW and InAs QDs as an active layer. Removal of surface oxide was monitored by reflection high-energy electron diffraction. The cleaned surface showed $\beta(2 \times 4)$ reconstruction. The photoluminescence properties of GaAs/AlGaAs QWs and InAs/GaAs QDs showed no degradation compared with those of the reference samples, even though the air-exposed interfaces were included in the active regions. [DOI: 10.1143/JJAP.43.L103]

KEYWORDS: atomic hydrogen etching/cleaning, GaAs quantum wells (QWs), InAs quantum dots (QDs), regrowth, photoluminescence (PL), molecular beam epitaxy (MBE)

Recently progress in semiconductor epitaxial growth demands regrowth technique for application not only to nano-structures fabrication such as nano-hole, trench, site-controlled quantum dots (QDs), and quantum wires on patterned surfaces but also to monolithic integration of electronic and photonic devices.[1–4] The main issue related to all such regrowth techniques is the introduction of interface contamination and defects at the restart interface, especially in the case of *ex situ* chemical processing. Simply exposing the GaAs surface to air introduces interface defects exceeding $2 \times 10^{11}/\mathrm{cm}^2$,[5] accordingly much higher concentrations may exist on processed surfaces. To overcome these problems, it is necessary to find out *in situ* etching/cleaning methods. There have been many reports on hydrogen cleaning of the regrowth interface which have been characterized by reflection high-energy electron diffraction (RHEED), Auger electron spectroscopy,[6,7] atomic force microscope,[4,8] secondary ion-microprobe mass spectroscopy,[9] two dimensional electron mobility[10] and photoluminescence (PL).[4,9] However, the quality of the hydrogen-cleaned regrowth interface is not satisfactory. In most cases, regrowth interfaces investigated so far are situated outside of active regions even though they are very close.[4,9,10]

In this letter, we show that well controlled atomic hydrogen cleaning can improve the PL of quantum wells (QWs) and QDs, in which the air-exposed interfaces are embedded in the active layers, to the level of the continuously grown QWs and QDs.

The growth, atomic hydrogen cleaning, and regrowth were performed in an ultrahigh vacuum molecular beam epitaxy (MBE) chamber (Riber 32P). We prepared two air-exposed samples, QW and QDs, and the reference samples for each. The samples were grown on undoped-GaAs (001) substrates by using group-III solid source (Ga, Al and In) and a valved arsenic cracker with As_4 molecules. Atomic hydrogen was generated by a hot tungsten cracker and fed on sample surface.[7] Atomic hydrogen cleaning was performed at 420°C and at a H_2 pressure of 3×10^{-6} Torr in the MBE chamber.

The structure of reference and regrowth QW was 500 nm GaAs buffer layer on the substrate, 50 nm $Al_{0.3}Ga_{0.7}As$ barrier, 5 nm GaAs active layer, 30 nm $Al_{0.3}Ga_{0.7}As$ barrier and 10 nm GaAs cap. All layers were grown at 580°C. In the case of regrowth QW, the air-exposed and hydrogen-cleaned interface was embedded in the active layer. For the purpose of air-exposure and cleaning, after 3 nm growth of GaAs active layer, the sample was taken out from the chamber and kept in the atmosphere in clean room for one hour. It was reloaded again in the MBE chamber and cleaned by atomic hydrogen. Then the rest of the QW, 2 nm GaAs layer was grown followed by 30 nm $Al_{0.3}Ga_{0.7}As$ barrier and 10 nm GaAs cap. The structure of QDs is as follows, on the substrate 500 nm buffer layer was grown and air-exposed for one hour. After hydrogen cleaning in the growth chamber, InAs QDs were grown to the nominal thickness of 2 MLs at the growth temperature of 400°C and at the growth rate of 0.14 ML/s. After that, 10 nm GaAs cap was grown. We also grew the same QD structure as a reference sample except air-exposure. The oxide removal from the GaAs surface was confirmed by the observation of RHEED pattern. The optical properties of the regrowth and the reference samples were measured by PL at 10 K. In PL measurement, an argon ion laser with a wavelength of 488 nm was used, and its intensity was 5 mW focused into a diameter of roughly 1 mm.

During the atomic hydrogen supply, the surface oxides are changed to volatile species such as water molecules as well as As_2, GaOH, and Ga_2O.[11,12] Without atomic hydrogen irradiation, the RHEED pattern of air-exposed surface showed a halo with tiny weak spots, indicating that the surface was covered with an amorphous oxide layer. The surface was then exposed to atomic hydrogen at different substrate temperatures (T_s) ranging from 200 to 450°C. At T_s 420°C, a clear RHEED pattern with $\beta(2 \times 4)$ reconstructions were observed. Generally, GaAs (001) surface shows a variety of reconstructed structure depending on surface stoichiometry. The structure changes from the most As-rich $c(4 \times 4)$ phase through the (2×4), (3×1), and (4×6) phases, and finally to the Ga-rich (4×2) phase as the As coverage is decreased. The reconstruction $\beta(2 \times 4)$ is As-stabilized surface with the As coverage 0.75 ML, which is usually observed on the surface under As-rich condition at T_s around 550°C.[13] As increasing T_s to 450°C, the surface reconstruction was changed to weak (2×4), indicating the degradation of surface morphology. Therefore, we consider

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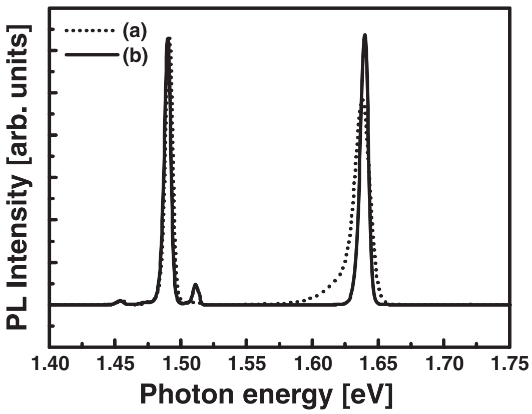

Fig. 1. PL spectra measured at 10 K from QW grown on the air-exposed and hydrogen-cleaned surface (a) and reference (b).

that the appearance of clear $\beta(2 \times 4)$ at 420°C is a good indicator for the cleaned surface and used it as a monitor of hydrogen cleaning. After obtaining optimum reconstruction, we proceeded to regrow the QW and the QD.

Figure 1 shows the PL spectra from GaAs/AlGaAs QWs with air-exposed regrowth interface (a) and of reference (b). The PL signal from the QW with regrowth interface is very strong even though the regrowth interface is embedded in the middle of active region, indicating that the atomic hydrogen effectively cleaned up the interface contaminants and oxide layer under optimum etching/cleaning conditions. The FWHM of the regrowth QW is 12 meV and approximately twice as much as that of the reference. However, this is ordinal value for the QW of 5 nm width which has monolayer fluctuations.[14)]

Figure 2 shows the PL spectra from QDs grown on the air-exposed and hydrogen-cleaned surface (a) and of reference (b). In the case of QDs, the emission peak from InAs QDs grown on hydrogen-cleaned interface is redshifted from that of the reference sample by 43 meV with a drastic decrease in FWHM from 67 to 36 meV. The redshift in the PL peak position and the decrease in FWHM could be due to the increase in QD size and the improvement of QD size uniformity, respectively. These can be attributed to the increase in the In adatom migration length on $\beta(2 \times 4)$ compared to $c(4 \times 4)$ GaAs surface. Because the $\beta(2 \times 4)$ surface structure still remained during InAs QDs grown on this surface. The integral intensity of the regrowth sample is approximately twice as much as that of the reference, which might be due to the passivation effect of hydrogen.[15)]

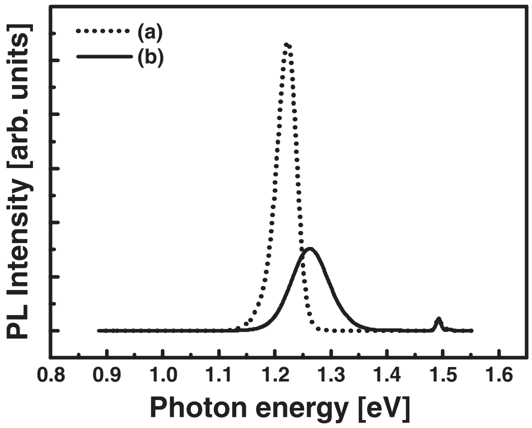

Fig. 2. PL spectra measured at 10 K from QD grown on the air-exposed and hydrogen-cleaned surface (a) and reference (b).

Table I. The optical parameters taken from *Gaussian* fitting of PL spectra at 10 K.

Samples	PL (eV)	FWHM (meV)	Intensity ratios
QW Reference	1.6395	6	1
QW Re-grown	1.6381	12	0.94
QD Reference	1.2632	67	1
QD Re-grown	1.2206	36	1.89

The peak positions and full width at half maximums (FWHM) are summarized in Table I. As shown in Table I the optical properties of the regrowth samples do not compare unfavorably with those of references, indicating that the atomic hydrogen works very well in cleaning the surface contaminants and oxide without introducing non-radiative recombination centers.

To the best of our knowledge, there are no report indicating strong PL of GaAs/AlGaAs QWs and InAs QDs directly grown on air-exposed and cleaned interface as an active layer, although there have been reported on QWs which have regrowth interfaces outside of barriers.[4,9)]

In conclusions, we have developed optimum cleaning method for the air-exposed GaAs surface by using atomic hydrogen without significant degradation of optical proprieties. Nondegraded PL intensities were observed from QWs and QDs which had air-exposed regowth interfaces in active regions. In addition, we confirmed that the atomic-hydrogen cleaned GaAs (001) surfaces had a stoichiometric $\beta(2 \times 4)$ reconstructions at T_s around 420°C.

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