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Leveling and rebuilding: An approach to improve the uniformity of (In,Ga)As quantum dots

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We report on an approach to improve the uniformity of a single layer of (In,Ga)As quantum dots (QDs) grown by molecular-beam epitaxy. The key concept of our approach is to level and rebuild the (In,Ga)As QDs during insertion of a short period GaAs/InAs superlattice between the (In,Ga)As QD layer and the GaAs capping layer. For optimized layer thickness and number of superlattice periods this process results in uniform (In,Ga)As QDs with narrow photoluminescence line width of 20 meV at 4.5 K. © 2002 American Institute of Physics. DOI: 10.1063/1.1506780

Fabrication of (In,Ga)As quantum dots (QDs) by molecular-beam epitaxy (MBE) has been extensively studied in order to obtain high quality low-dimensional quantum structures. However, the random nucleation process during the formation of QDs undesirably leads to large size fluctuations, which, consequently, result in broad photoluminescence (PL) peaks, typically with line widths of 40–100 meV. A small PL line width is, however, of importance for QD lasers to reduce the threshold current. Stacked QD layers grown by MBE have been demonstrated to exhibit a narrower PL line width but stacking can only improve the uniformity of QDs in the layers above the first layer of dots, which serve as seeds to affect the nucleation of QDs in the next layer by modifying the strain distribution. The uniformity does not change for QDs in the first layer. Little research has been done to improve the uniformity of a single layer of QDs. In this letter, we present an approach to improve the uniformity of a single layer of (In,Ga)As QDs. By this approach, the PL line width at 4.5 K is lowered from 38 to 20 meV.

The samples were grown by solid source MBE on semi-insulating GaAs (100) substrates. After oxide desorption at 580 °C, a 300 nm thick GaAs buffer layer was deposited. Then the substrate temperature was lowered to 500 °C for growth of the (In,Ga)As QD layer formed by 2.1 nm In0.35Ga0.65As. Between the (In,Ga)As QD layer and the 10-nm-GaAs capping layer, a short period GaAs/InAs superlattice (SL) was inserted with a growth interruption time of 10 s between each layer. A series of samples was grown with a number of SL periods N ranging from 0 to 20 and a different thickness of GaAs and InAs in the SL. Thereafter, the substrate was heated up to 580 °C for growth of 100 nm GaAs. The (In,Ga)As QD layer and GaAs/InAs SLs were repeated on top at 500 °C for atomic force microscopy (AFM) measurements. The growth rates for GaAs and InAs were 0.24 and 0.13 μm/h, with the arsenic beam equivalent pressure kept at 8–9 × 10⁻⁶ Torr. During growth of the (In,Ga)As QDs and GaAs/InAs SLs, the surface was monitored by reflection high-energy electron diffraction (RHEED). For PL measurements, the samples were excited by a Nd:YAG laser in a cryostat (4.5 K) at an excitation power density of 256 mW/cm². The surface morphology of the samples was characterized in air by tapping mode AFM.

The AFM image of the (In,Ga)As QD layer formed by 2.1 nm In0.35Ga0.65As is shown in Fig. 1(a). After capping the QD layer by 2 ML GaAs, the surface morphology is that

FIG. 1. AFM images (a) (In,Ga)As QDs formed by 2.1 nm In0.35Ga0.65As, and (b) the (In,Ga)As QDs capped by 2 ML GaAs. The height scale is 5 nm.

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shown in Fig. 1(b). The AFM results are consistent with the
RHEED observation. For the surface morphology shown in
Fig. 1(a), the RHEED pattern viewed along the [01\bar{1}] azimuth consists of well-defined chevrons which evolve after 1.7 nm In\textsubscript{0.35}Ga\textsubscript{0.65}As. During growth of the GaAs capping layer, the chevrons fade away gradually. The RHEED pattern changes to short streaky diffraction rods with weak intensity within deposition of 2 MLs GaAs. The transition of the RHEED pattern indicates that the QDs level during the capping process (the original QD height is 3 nm), resulting in a relatively flat surface with mounds elongated along the [01\bar{1}] direction, as shown in Fig. 1(b). The evolution of the RHEED pattern during capping the (In,Ga)As QDs is very similar to the case of capping InAs QDs reported in Ref. 10. For deposition of only 0.5 MLs InAs on the 2 ML GaAs, we find complete rebuilding of the (In,Ga)As QDs on the surface, indicated by the change of the RHEED pattern from weak streaky diffraction rods back to clear chevrons. It is interesting that the evolution of the RHEED pattern described above is identically repeated during each period of the GaAs/InAs (2 ML/0.5 ML) SLs.

It is obvious that both processes, leveling and rebuilding of QDs involve a large amount of mass transport. When leveling In atoms migrate away from the (In,Ga)As QDs while the migration direction is opposite during QD rebuilding. Leveling is driven by strain relief between the QDs and the GaAs capping layer. Indium and gallium atoms diffuse from the QDs over a large area, resulting in a rather flat surface morphology with shallow (2–3 ML high) mounds, as evidenced in Fig. 1(b). The lateral extension of the mounds by far exceeds the dot-to-dot distance. Hence, net migration of In atoms is expected from the location of larger QDs to that of smaller ones. Nucleation of the following QDs on top of the leveled QDs involves collection of In atoms around their base (the QDs form for 0.5 ML InAs while the critical layer thickness on GaAs is 1.7 ML). Therefore, for QDs rebuilding on smaller leveled QDs, a portion of In atoms is provided from neighboring larger leveled QDs in the preceding layer. By this way, exchange of In atoms from larger to smaller QDs occurs, which results in distinct improvement of the uniformity of the QD structure in SL growth. The improvement is directly confirmed by the PL spectra shown in Fig. 2 and the surface morphology in Fig. 3(a). (In,Ga)As QDs on GaAs show a PL peak centered at 960 nm with line width of 38 meV. The PL line width of the (In,Ga)As QDs decreases to 26 meV for the sample with two periods GaAs/InAs SL inserted between the QD layer and GaAs capping layer, and the PL peak position redshifts to 1006 nm. Note that remarkable improvement is obtained by insertion of two periods GaAs/InAs SL with total thickness as small as 1.4 nm. The narrowest PL peak of 20 meV width at 1017 nm and with the highest peak intensity is achieved when the number of SL periods is increased to 10. Further increase of the number of SL periods results in PL line broadening and surface roughening when the GaAs/InAs SL becomes too thick. The PL peak shift to longer wavelength with increase of the number of SL periods is attributed to strain reduction and increasing size of the QDs during SL growth, while the narrowing of the PL line is mainly assigned to improved size uniformity rather than strain reduction or size increase as discussed in the following.

A sample with 6 nm In\textsubscript{0.20}Ga\textsubscript{0.80}As, i.e., the same In composition as the average one of the GaAs/InAs (2 ML/0.5

![Fig. 2. PL spectra at 4.5 K of samples with different number of periods (N) of GaAs/InAs (2 ML/0.5 ML) SLs inserted between the (In,Ga)As QD layer and the GaAs capping layer. The inset shows the dependence of the PL line width on N.](image)

![Fig. 3. AFM images of samples with (a) 2.1 nm and (b) 1.4 nm In\textsubscript{0.35}Ga\textsubscript{0.65}As layers capped by ten periods GaAs/InAs (2 ML/0.5 ML) SLs. The height scale is 7 nm.](image)
ML) SL, inserted between the (In,Ga)As QD layer and the 10 nm GaAs capping layer reveals a PL peak at 992 nm with line width of 42 meV. Strain relief of the QDs due to the GaAs/InAs SL which is comparable to that due to the (In,Ga)As layer is, thus, excluded as the reason for the reduction of the PL line width. Moreover, replacing the ten periods GaAs/InAs (2 ML/0.5 ML) SLs by 20 periods GaAs/InAs (1 ML/0.25 ML) SLs causes the PL line width to increase from 20 to 29 meV. One ML GaAs and 0.25 ML InAs is too thin to fully establish the repeated processes of leveling and rebuilding QDs, as indicated by the almost unchanged spotty RHEED pattern during growth. Finally, also for a sample with a five periods GaAs/InAs (2 ML/0.75 ML) SL inserted between the (In,Ga)As QD layer and the GaAs capping layer a broad PL peak at 1080 nm with line width of 51 meV is obtained. Here, the large amount of InAs breaks the balance between leveling and rebuilding the QDs, and the RHEED pattern also stays spotty during growth. Although in these last two examples, the increase of the dot size can be assumed to be the largest due to the maintenance of a dot morphology during overgrowth, the PL line width increases. Hence, we conclude that the PL line narrowing for the sample with GaAs/InAs (2 ML/0.5 ML) SL is due to improvement of the (In,Ga)As QD size uniformity enabled by the highest degree of mass transport between the (In,Ga)As dots during QD leveling and rebuilding, as indicated by the RHEED pattern to change repeatedly from streaky to spotty.

Further information on the overgrowth process is provided by comparing Figs. 3(a) and 3(b) of the surface morphology of the ten periods GaAs/InAs (2 ML/0.5 ML) SLs grown on 2.1 nm (sample A) and 1.4 nm (sample B) In$_{0.35}$Ga$_{0.65}$As. For sample A, high density and uniform (In,Ga)As QDs are formed while the surface of sample B is very flat except for a low density of large mounds elongated along the [011] direction. The difference is due to the formation of (In,Ga)As QDs after the growth of 2.1 nm In$_{0.35}$Ga$_{0.65}$As, while the 1.4 nm In$_{0.35}$Ga$_{0.65}$As layer remains flat. The PL spectra for samples A and B are shown in Fig. 4. For sample B, the narrow peak (12 meV) at 942 nm is assigned to the (In,Ga)As quantum well in the flat area while the broad peak at 990 nm is from the large mounds, as shown in Fig. 3(b). For sample A, only a narrow PL peak (20 meV) at 1017 nm from the (In,Ga)As QDs is observed. Clearly, the initial surface status strongly affects the growth of the GaAs/InAs SLs. Note that both the average In composition of the GaAs/InAs (2 ML/0.5 ML) SL of 0.20 and each InAs layer thickness of 0.5 MLs are below the critical value for dot formation. Hence, leveling and rebuilding of the QDs during

![FIG. 4. PL spectrum (solid line) at 4.5 K for the sample with ten periods GaAs/InAs (2 ML/0.5 ML) SL inserted between the (In,Ga)As QD layer formed by 2.1 nm In$_{0.35}$Ga$_{0.65}$As and the GaAs capping layer. The dashed line shows the PL spectrum of the sample with the same structure but the In$_{0.35}$Ga$_{0.65}$As layer thickness of 1.4 nm.](image-url)