Modification of InAs quantum dot structure by the growth of the capping layer

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InAs quantum dots inserted at the middle of a GaAs quantum well structure have been investigated by transmission electron microscopy and scanning transmission electron microscopy. We find that the growth condition of the overlayer on the InAs dots can lead to drastic changes in the structure of the dots. We attribute the changes to a combination of factors such as preferential growth of the overlayer above the wetting layers because of the strained surfaces and to the thermal instability of the InAs dots at elevated temperature. The result suggests that controlled sublimation, through suitable manipulation of the overlayer growth conditions, can be an effective tool to improve the structure of the self-organized quantum dots and can help tailor their physical properties to any specific requirements of the device applications. © *1998 American Institute of Physics.* [S0003-6951(98)00227-7]

The fabrication of quantum dots (QDs) has been intensively studied in recent years due to promising applications such as QD lasers,[1,2] single electron transistor,[3] etc. In lattice-mismatch systems, self-organized QDs can be achieved by using a Stranski-Krastanov (SK) growth mode.[4] In such a growth mode, the initial growth is layer by layer which is called the wetting layer, and subsequent growth is via isolated islands, which lower the mismatch strain energy. Extensive study has been carried out to characterize the growth of self-organized dots. It appears that the dot density and size distribution are mostly a function of the InAs deposition. Improvements in the structure of the InAs dot assembly have concentrated on this front. There are relatively few studies[5–8] which have focused on the effect of the capping layer which is required for the passivation of the surface of the InAs quantum dots for device applications. In this letter, we show clear evidence for the role of the capping layer growth to modify the self-organized dot structure. In particular, we show that change is induced by sublimation of the InAs dots because of the incomplete coverage by the GaAs capping layer on strained surfaces. Controlled sublimation is shown to lead to elimination of the incoherent InAs particles, to narrowing of the particle size distribution and to controlled modification of the dot height. Our results suggest an avenue for the optimization of such dot structures.

The samples were grown in a molecular beam epitaxy (MBE) chamber, using a GaAs (100) wafer. The structure consists of a 0.6 μm thick undoped GaAs buffer layer, followed by a 50 nm undoped $Al_{0.33}Ga_{0.67}As$ barrier, a 20 nm undoped GaAs quantum well, a 40 nm undoped $Al_{0.33}Ga_{0.67}As$ spacer layer, a 40 nm Si-doped (1×10^{18} cm^{-3}) $Al_{0.33}Ga_{0.67}As$ layer, and finally a 17 nm GaAs cap layer. For the purpose of this letter, we need to focus only on the growth condition of the undoped GaAs quantum well structure. The first 10 nm of GaAs were grown at a substrate temperature of 580 °C as measured by an optical pyrometer. Then, InAs of various nominal thicknesses is introduced at 530 °C. The transition from pseudomorphic layer growth to SK growth mode is monitored by *in situ* scanning tunneling microscopy (STM) and atomic force microscopy (AFM) measurement.[9] The samples reported for this letter are all grown with InAs coverage of 2.15 ML where self-organized InAs dots are found by STM on the GaAs surface to have an estimated density of 10^{10} cm^{-2}. The dots are circular with a mean diameter of 26 nm and an average height of 8 nm. Three samples were grown with the same structural sequence except for the condition of the top half of the GaAs quantum well layer which acts as the capping layer for InAs dot structure. In sample 1, all of the 100 Å thick GaAs capping layer was grown at 530 °C (the same temperature of the InAs QD formation). In sample 2, the first 50 Å GaAs was grown at 530 °C and 50 Å GaAs was grown at 580 °C. In sample 3, only the first 10 Å GaAs was grown at 530 °C and the remaining 90 Å GaAs was grown at 580 °C. The substrate temperature ramping time was 2 min between 530 and 580 °C and no temperature overshoot was observed. The samples were examined in a JEOL-2000EX transmission electron microscope (TEM) operating at 200 kV and a VG-HB501 dedicated scanning transmission electron microscope (STEM) operating at 100 kV. Both plan-view and cross-section specimens were prepared by mechanical polishing followed by ion milling in a liquid nitrogen cooled stage.

Figure 1 shows plan view TEM images of the three samples, taken from the (100) zone axis. For sample 1, the InAs dots or particles are clearly resolved through the well-known strain contrast mechanism of Ashby and Brown.[10] The dot density is about half the value obtained from *in situ* STM observation. This reduction in density is accompanied by the growth of some particles, indicating some particles ripening during the growth of the capping layer. It is important to realize that the quantum dots are precipitate particles confined by the wetting layer to a plane. The behavior of such particles is very familiar in three dimensions from stud-

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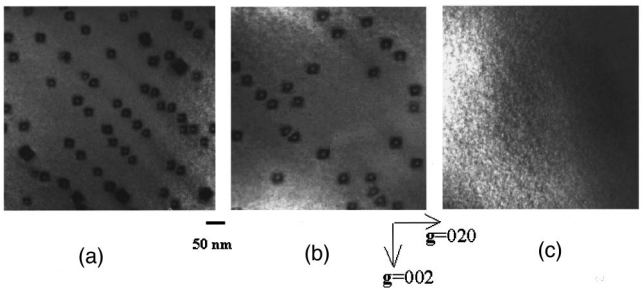

FIG. 1. Plan-view images from (100) zone axis of sample 1 (a), sample 2 (b), and sample 3 (c).

ies of two-phase materials,[11] but of course it is modified by the planar confinement and by the capping layer. We will return to this point later.

Now we describe the evolution of the dot or particle structure as the growth temperature of the capping layer is altered. As is shown from Fig. 1, the dot density is halved in sample 2, and no dot is observed in sample 3. This behavior is also clearly apparent in the cross-sectional images, which also show the characteristic wetting layer, which is present in all three samples.

A closer examination of the plan-view TEM image in Fig. 1 reveals contrast variation between InAs dots for specimen 1. The same area is imaged at higher magnification using two-beam strain contrast in Fig. 2 and two particles are found. The particles showing a line of no contrast perpendicular to the g vector[9] are coherent, those with much stronger and complicated strain contrast are incoherent, as revealed by the threading dislocation lines imaged by the TEM in the $g/3g$ weak beam dark field (DF) condition, Fig. 2(b). The number of incoherent particles in sample 1 is about 12% of the total particles present. No such incoherent particles are observed in samples 2 and 3. It is interesting that the coherent particles have insufficient strain to be visible in the weak beam image.

To determine accurately the dimensions of the dots observed, we have used the quasikinematic imaging condition suggested by Woolhouse and Brown[12] together with the incoherent annular dark field imaging technique. Figure 3 shows a centered dark field image from TEM and the annular dark field image (ADF) from STEM of the same area in sample 2. In the DF image, the strain contrast is dominant and it is not easy to identify the true dot shape. The line of no contrast exists but is seen to bend slightly upward, indicating

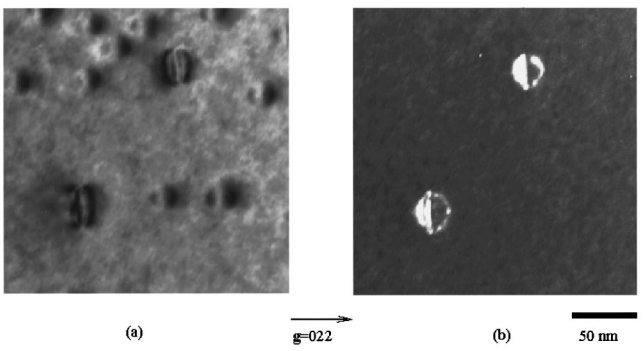

FIG. 2. Bright Field (a) and weak beam ($g/3g$) dark field from plan view of sample 1 (b).

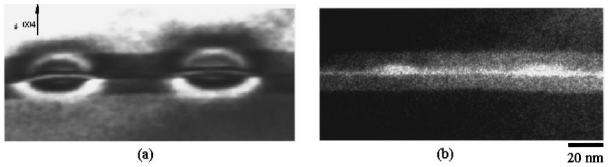

FIG. 3. Centered dark field TEM (a) and ADF from STEM (b) for comparison; both from the same area in sample 2.

that the part of the strain due to the lattice mismatch is now accommodated in the GaAs buffer layer.[4,5] There is no strain contrast in the ADF image, the real dot shape shows in the cross-section specimen by STEM, as well as the images of the InAs wetting layer and the GaAs quantum well. Through the ADF imaging mode, we confirm that no dots are present in sample 3. From these measurements, we find the average dimension of the dots is about 36 nm width and 8 nm height for sample 1, about 28 nm width and 4 nm height for sample 2, and of course, zero dimensions for sample 3.

The evolution of the self-organized dots in the three samples may be understood if we consider the adsorption sites for the subsequent GaAs deposition. The incoming adatoms see an inhomogeneous and uneven substrate consisting of the InAs wetting layer a few monolayers thick and InAs dots several nanometers in height. Most importantly, the wetting layer is fully strained to be lattice matched with the GaAs substrate, but the top of the InAs dots is partially relaxed with a lattice spacing approaching that of the bulk InAs crystal. From energetic considerations alone we, therefore, expect the GaAs overlayer growth to start preferentially on the wetting layer, if the adatoms have sufficient surface mobility. Evidence for this growth selectivity has been demonstrated for GaAlAs multilayer growth on self-organized InAs dots on GaAs(100) surface[5] and we can expect that our case is similar. As a consequence, we do not expect the top of the InAs dots to be protected by the GaAs overlayer until the equivalent thickness of the total GaAs deposition is at least equal to or in excess of the dot height, whereby the GaAs overlayer growth on the dot can be driven both by curvature of the growth front and the redistribution of the strain in the dot. For sample 1 where the overlayer thickness is greater than the dot size, this growth selectivity has no observable effect on the dot density and the dot height as is observed. For samples 2 and 3, the nominal thickness of the low temperature grown GaAs overlayers is less than the height of the uncapped InAs dots. Since the growth of the second half of the overlayer takes place at the elevated temperature where the InAs dot is not stable against evaporation,[13] we expect partial evaporation of the InAs dot. This is consistent with the observation that the dot height is reduced to 4 nm in sample 2, so that partial protection of the dot is possible from the edge of the islands, and all dots disappear in sample 3.

One feature of the evolution of the dot structure from sample 1 to sample 2 is the reduction in dot density from 5.8×10^9 to 3×10^9 cm^{-2} (counting from an area about 12 μm^2), and especially the complete absence of incoherent particles in sample 2 (Figs. 1 and 2). This suggests that the partial protection offered by the low temperature grown GaAs layer in sample 2 is not effective for some of the particles. This is possible if the particle is large so that

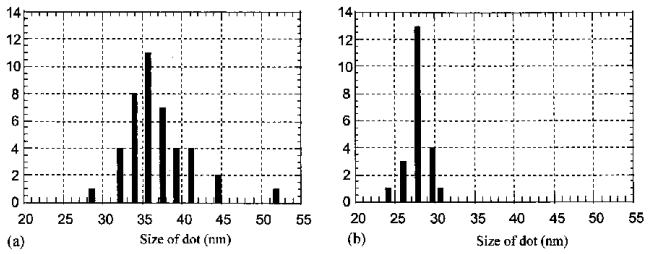

FIG. 4. Size distribution of dots from sample 1 (a) and sample 2 (b).

evaporation can proceed from the center towards the edge. We have evidence for this in the observation of some ring-shaped particles in sample 2. It is also likely that the larger particles have a more relaxed surface, hence, are more difficult for the GaAs adatoms to adhere to. This favors selective thermal desorption of large particles. This is supported by the narrower size distribution for dots found in sample 2, Fig. 4(b). The same mechanism is thought to be responsible for the disappearance of incoherent dots in sample 2 since the surface of such uncapped dots will be fully relaxed. In plan-view images, some thinned patches in the film can be seen where dots had probably formed, but had subsequently disappeared, consistent with the selective desorption process. Of course, the sublimation model outlined above may be a simplification, as Ga-In intermixing is known to be important as well, but many features of the observations can be understood in terms of the lack of protection against desorption afforded by the capping layer where the misfit with the InAs is large.

Returning to the TEM image of sample 1 shown in Fig. 1, comparison with the corresponding *in situ* STM result shows that the low temperature growth is not entirely benevolent. The density of particles is reduced as a result of overlayer growth. This is accompanied by a broadened size distribution [Fig. 4(a)] that is a sign of the coarsening process by ''Ostwald ripening,'' as described, for example, in the book by Haasen.[11] How this is influenced by the growth of the overlayer remains unanswered. On the other hand, the appearance of a significant number of incoherent particles (12%) is understandable because of preferential growth of the larger incoherent particles due to their free-energy advantage, and possibly by redistribution of the strain in the particles during the overlayer growth.

The size distribution of sample 2 (13% variation in size) is much narrower than that of coherent dots in sample 1, indicating that size distribution in sample 2 is more ideal for the fabrication of devices like QD lasers. This result coincides with the results of photoluminescence spectroscopy from these three samples, which demonstrate no InAs QD peak in sample 3, and shows that the full width at half maximum (FWHM) of the InAs QD peak from sample 2 is 35 mV, narrower than that from sample 1. Reference 7 also reports that the narrowest FWHM of the QD peak has been obtained by adjusting the thickness of the capping layer, although the corresponding images of the structure of QDs among those samples is absent.

In conclusion, we have demonstrated that InAs QDs disappear even if nominally covered by a GaAs capping layer grown at low temperature with a thickness of only 10 Å. As the thickness of the GaAs capping layer increases, the dot density surviving beneath it increases, but some of the dots start to lose coherency. Optimization of quantum dot structure is achieved by adjusting the overlayer growth condition. The result also shows that the *ex situ* observations by TEM And STEM are essential to monitor the structure in addition to the STM and AFM *in situ* observations.

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[1] S. Fafard, K. Hinzer, S. Raymond, M. Dion, J. McCaffrey, Y. Feng, and S. Charbonneau, Science **274**, 1350 (1996).
[2] F. Heinrichsdorff, M.-H. Mao, N. Kirstaedter, A. Krost, D. Bimberg, A. O. Kosogov, and P. Werner, Appl. Phys. Lett. **71**, 22 (1997).
[3] K. Yano, T. Ishii, T. Hashimoto, T. Kobayashi, F. Murai, and K. Seki, IEEE Trans. Electron Devices **41**, 1628 (1994).
[4] D. J. Eaglesham and M. Cerullo, Phys. Rev. Lett. **64**, 1943 (1990).
[5] Q. Xie, P. Chen, and A. Madhukar, Appl. Phys. Lett. **65**, 2051 (1994).
[6] X. W. Lin, J. Washburn, Z. Liliental-Weber, E. R. Weber, A. Sasaki, A. Wakahara, and Y. Nabetani, Appl. Phys. Lett. **65**, 1677 (1994).
[7] R. P. Mirin, J. P. Ibbetson, J. E. Bower, and A. C. Gossard, J. Cryst. Growth **175/176**, 696 (1997).
[8] M.-E. Pistol, N. Carlsson, C. Persson, W. Seifert, and L. Samuelson, Appl. Phys. Lett. **67**, 1438 (1995).
[9] S. J. Brown, 1996, Ph.D. thesis, University of Cambridge. Copy obtainable from the University Library.
[10] M. F. Ashby and L. M. Brown, Philos. Mag. **8**, 1083 (1963).
[11] P. Haasen, *Physical Metallurgy*, 2nd ed. (Cambridge University Press, 1986), p. 214.
[12] G. R. Woolhouse and L. M. Brown, *4th European Regional Conference on E. M.*, edited by D. S. Bocciarelli (Tipografia Poliglotta Vaticana, Rome, 1968), p. 297; see also G. R. Woolhouse, Ph.D. thesis, University of Cambridge, 1968, copy obtainable from the University Library.
[13] O. Brandt, K. Ploog, L. Tapfer, M. Hohenstein, R. Bierwolf, and F. Phillipp, Phys. Rev. B **45**, 8543 (1992).