Formation of self-assembled GaAs/AlGaAs quantum dots by low-temperature epitaxy

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(Received 6 July 1998; accepted for publication 31 August 1998)

We report the direct formation of self-assembled GaAs/AlGaAs quantum dots by low-temperature molecular beam epitaxy. To drive a three dimensional growth mode, the (1×1) AlGaAs surface was exposed alternately to the Ga and As sources. The resulting GaAs nanocrystals having [111] facets were clearly identified by high-resolution transmission electron microscopy. The emission spectra also confirmed the formation of dots. The transition to a three-dimensional growth mode is attributed to the limited surface migration of Ga adatoms on the AlGaAs surface, which has excess As at low substrate temperature. © 1998 American Institute of Physics. [S0003-6951(98)01544-7]

Semiconductor quantum dots (QDs) have been of interest not only for high efficient device applications but also for the formation mechanism depending on growth conditions. One of the recent advances in the fabrication of zero-dimensional QDs makes use of the natural consequences in the growth modes for heteroepitaxial growth. It is well known that thermodynamic considerations of the initial stage of epitaxy have led to the distinction of three different growth modes. In the Frank-van der Merwe (FM) mode the epitaxial material grows in a continuous two dimensional (2D) layer-by-layer way. The Stranski–Krastanov (SK) mode is an intermediate case, in which the growth of 2D wetting layer is followed by three dimensional (3D) islanding. In the Volmer–Weber (VW) mode, the formation of clusters and islands is a dominant process which results in the 3D growth.

For self-assembled QDs on a lattice-mismatched layer, most studies have exploited the transition from the 2D to the 3D growth mode which is driven by the reduction in the strain energy associated with an elastic deformation of the island morphology in the SK growth mode. However, few methods have been available for the direct formation of dots on a nearly or exactly lattice-matched layer such as GaAs/AlGaAs heterostructure. To drive the 3D islands growth for such system, the VW growth mode is expected to be more desirable than the SK mode. The VW mode is likely to be realized by either surfactant mediation or control of the external growth parameters under conditions that are far from equilibrium. Droplet epitaxy is based on incorporating the V-column element into III-column element droplets on a sulfur-passivated surface of the substrate. The inertness of the sulfur-passivated surface to the adorption of foreign atoms can drive a transition from the 2D to the 3D growth mode.

In this study we proposed and demonstrated the low-temperature (LT) droplet epitaxy for the direct formation of self-assembled GaAs QDs on AlGaAs. The LT droplet epitaxy employs the (1×1) semiconductor surface which provides the advantage over surfactant mediated droplet epitaxy that it does not require a sulfur source.

The samples studied in this work were grown on semi-insulating (001)-oriented GaAs substrates by molecular beam epitaxy (MBE) using reflection high energy electron diffraction (RHEED) to monitor the growth. After the growth of 1-μm-thick GaAs buffer layer at 580 °C with an As/Ga beam equivalent ratio of 20, the substrate temperature was ramped down to 250 °C. Then following a 200-nm-thick Al0.34Ga0.66As layer, the exposure of Ga beam of several equivalent monolayers (eq-ML) was initially performed, and an As beam was exposed immediately after the Ga exposure to form GaAs QDs. An 100-nm-thick Al0.34Ga0.66As layer was grown at the same substrate temperature in order to make buried structures, and followed by uncapped GaAs QDs for the measurement of surface morphology.

Figure 1 shows the RHEED patterns obtained in the (110) and (1  10) azimuth during the GaAs QD growth. As the substrate temperature was decreased to 250 °C, the surface structure of AlGaAs changed from a (2×4) to a c(4×4) pattern. Immediately after the deposition of LT-AlGaAs, the (1×1) surface, normally observed in LT-MBE growth, was evident. Exposure of the Ga beam gave rise to a halo pattern caused by the formation of Ga droplets as shown in Fig. 1(a). With a subsequent exposure of As beam without the Ga beam, the halo pattern is replaced by spotty features with streaks along the [111] azimuth as shown in Fig. 1(b). These sequential changes of the RHEED patterns indicated that the 3D VW growth mode instead of the 2D FM mode occurs on the LT-AlGaAs surface. When the Ga exposure was per-
formed on the c(4×4)-AlGaAs layer grown at normal temperature, the halo pattern also appeared at 250 °C. However, during the As exposure the halo pattern disappeared and changed to (1×1) streaks, indicating that appreciable facet evolution did not occur. The experimental result suggests that the surface structure of AlGaAs plays a significant role for the facet formation in the initial stage of island growth. Additionally, the simultaneous exposure of Ga and As beams shows the (1×1) surface where the 2D layer-by-layer growth proceeds. From the RHEED observation, it is worthy to note that the sequential exposure as well as the LT-grown surface is necessary to form 3D islands.

After the growth, we carried out AFM measurements under ambient condition to investigate the surface morphology. In Fig. 2, for the sample with the Ga exposure of 4.5 eq-ML, the square-based GaAs islands represented by bright columnar structure were seen in multimodal shapes with a density of ~210/μm². The size and density strongly depend on the growth condition. By increasing the substrate temperature or decreasing the As pressure up to quasiequilibrium condition, the GaAs island became elongated in the [110] direction corresponding to the surface migration of Ga adatoms.

The cross-sectional image of a GaAs dot of 15 eq-ML was measured by using high-resolution transmission electron microscopy (HRTEM) with an energy of 300 keV. An uncapped sample was chosen because, in a HRTEM image of a buried structure, it was difficult to distinguish GaAs from AlGaAs. We found that the facet planes of the dots are {111}, consistent with the RHEED observation, and that the GaAs wetting layer was absent, indicating that the growth occurs in the VW mode. The Ga droplets had hardly migrated on the LT-AlGaAs surface, and the base dimension was preserved during the As beam supply. RHEED observations demonstrate the important role of the LT-grown (1×1) surface and sequential exposure for the formation of facets.

Since the formation of 3D islands is strongly related to the surface diffusion of Ga adsorbates, the role of the low substrate temperature is important. Because the surface diffusion constant D is given by \( D = D_0 \exp(-E_e/kT) \), the Ga migration length \( L = \sqrt{D_t} \) on the AlGaAs(001) surface decreases exponentially with the substrate temperature. When the Ga droplets initially form on the AlGaAs layer, the adatoms are so immobile that they cannot jump over local energy barriers. Insufficient mass transport causes the surface to be rough and to grow according to the 2D mode. However, it is not a sufficient condition to drive the 3D growth, because simultaneous exposure results in a 2D layer-by-layer growth. More important is the effect of excess As on the island formation on the LT-AlGaAs surface, because the Ga diffusion length increases with decreasing As pressure. Although the detailed (1×1) surface structure of LT-GaAs is not fully understood, it is expected that the LT-GaAs surface contains excess As, and that this limits the Ga migration length. Under the same low substrate temperature and Ga flux but with reduced As pressure, elongation of the GaAs dots is observed, implying the enhancement of the Ga migration length associated with the lower As concentration on the surface. Considering the desorption temperature of As around 250 °C, we also expect that the Ga migration length on a (1×1) surface with a large As concentration is much shorter than that on a c(4×4) surface. Thus, we do not expect the formation of {111} facets to appreciably occur on the c(4×4) surface. At high As atmosphere as well as at low temperature, 3D VW growth can proceed in a special way resulting in the creation of dots on the surface. The change in the lattice constant of LT-GaAs due to the excess As is less than 0.1%, which is not enough to give rise to the stress-driven 3D transition. Our experimental result indicates that the stress hardly influences the formation of 3D islands and that the wetting layer was not found in Fig. 3.

Microphotoluminescence investigations with a spot diameter of ~1 μm were carried out at 77 K with an excitation energy of 1.959 eV. The emission spectra of a 4.5 eq-ML sample with a square base of around 40 nm and a height of 2 nm is shown in Fig. 4. First, the peak with the energy of 1.606 eV is far from the band gap of LT-AlGaAs whose emission intensity was almost below the detection limit. The full width at half maximum of the photoluminescence (PL) peak is 33 meV and larger than that of the emission peak from the 4.5-ML-thick quantum well (QW) grown at normal...
temperature (NT). As shown in Fig. 4(a), the line shape follows a Gaussian profile, reflecting the size distribution of the dot ensemble. The confinement effect arose mainly along the growth direction, not the lateral directions, because of the large base size of the dots. However, the peak energy of the emission spectra was higher than the value calculated for a 2-nm-thick QW by 10 meV. This blue shift might be caused by the dome shape of the QDs where the top dimension is reduced. As the excitation power density is decreased, the broad featureless emission spectra exhibit several sharp peaks. With the excitation power of 0.2 μW, the PL spectra revealed a number of sharp lines which might be considered as exciton states in dots of different sizes. It should be noted that the line width of these peaks (about 1–5 meV) is still larger than that of the spectral noise. On the other hand, the luminescence intensity of QDs is much weaker than that of NT-QW due to the low density of dots and an increased number of defects in LT growth, but it is much stronger than that of LT-QW by about two orders of magnitude. The enhanced luminescence efficiency due to the three-dimensional confinement provides an additional evidence for the formation of QDs.

In conclusion, we demonstrated the formation of GaAs dots on AlGaAs with the alternating supply of Ga and As beam source at low temperature. The LT process provides a useful fabrication technique for various nanostructures. Although the features of the QDs are thought to originate from the thermodynamic stability, further investigations on the nucleation site of Ga droplets and the role of the point defects are necessary to fully understand the surface structure and optical properties of LT-grown GaAs/AlGaAs QDs.

The authors would like to thank Dr. N. Koguchi at the National Research Institute for Metals in Japan for his helpful discussions. This work was supported by the Korean Ministry of Science and Technology (Project Code: SP-KS-05-A).

9 From the estimation of the distance between two GaAs QDs, the surface diffusion constant $D$ is calculated to be of the order of $10^{-2}$ cm$^{2}$/s, which is smaller than that of the As-adsorbed surface.