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Letter to the editor

Formation of self-assembled ZnTe quantum dots on ZnSe buffer layer grown on GaAs substrate by molecular beam epitaxy

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Abstract

Self-assembled ZnTe quantum dot structures were grown by molecular beam epitaxy on GaAs substrates with a 200 nm ZnSe buffer layer. Surface morphology was studied by atomic force microscopy. A three-dimensional Volmer–Weber growth mode was identified. Two types of dots were observed. Strong photoluminescence observed at 1.9–2.2 eV was attributed to emission from the large type II ZnTe quantum dots. Emission from the smaller ZnTe quantum dots was observed at an energy of around 2.26 eV. The density of the larger and smaller dots was approximately $10^8/cm^2$ and $10^9/cm^2$, respectively. © 2002 Elsevier Science B.V. All rights reserved.

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Advances in the fabrication technique of quantum dot structures (QDs) have realized single photon detection [1] and memory cell [2] production, using a QD field-effect transistor. The fabrication of a QD laser also has become feasible [3,4]. Recently, GaN QDs in an AlN matrix have been grown on Si (1 1 1) by molecular beam epitaxy (MBE). A dot of appropriate size has been demonstrated to emit white light [5]. A light-emitting diode has also been made from II–VI compound semiconductor QDs [6]. Despite the intense attention paid to the fabrication of opto-electronic devices based on QD and the novel physical properties of material QD systems, time evolution of dot shape during and after growth remains a problem [7,8]. Neither the growth dynamics nor the phase diagram of the whole II–VI compound semiconductor family has been thoroughly explored. Two to three-dimensional, Stranski–Krastanow (SK) growth mode has been

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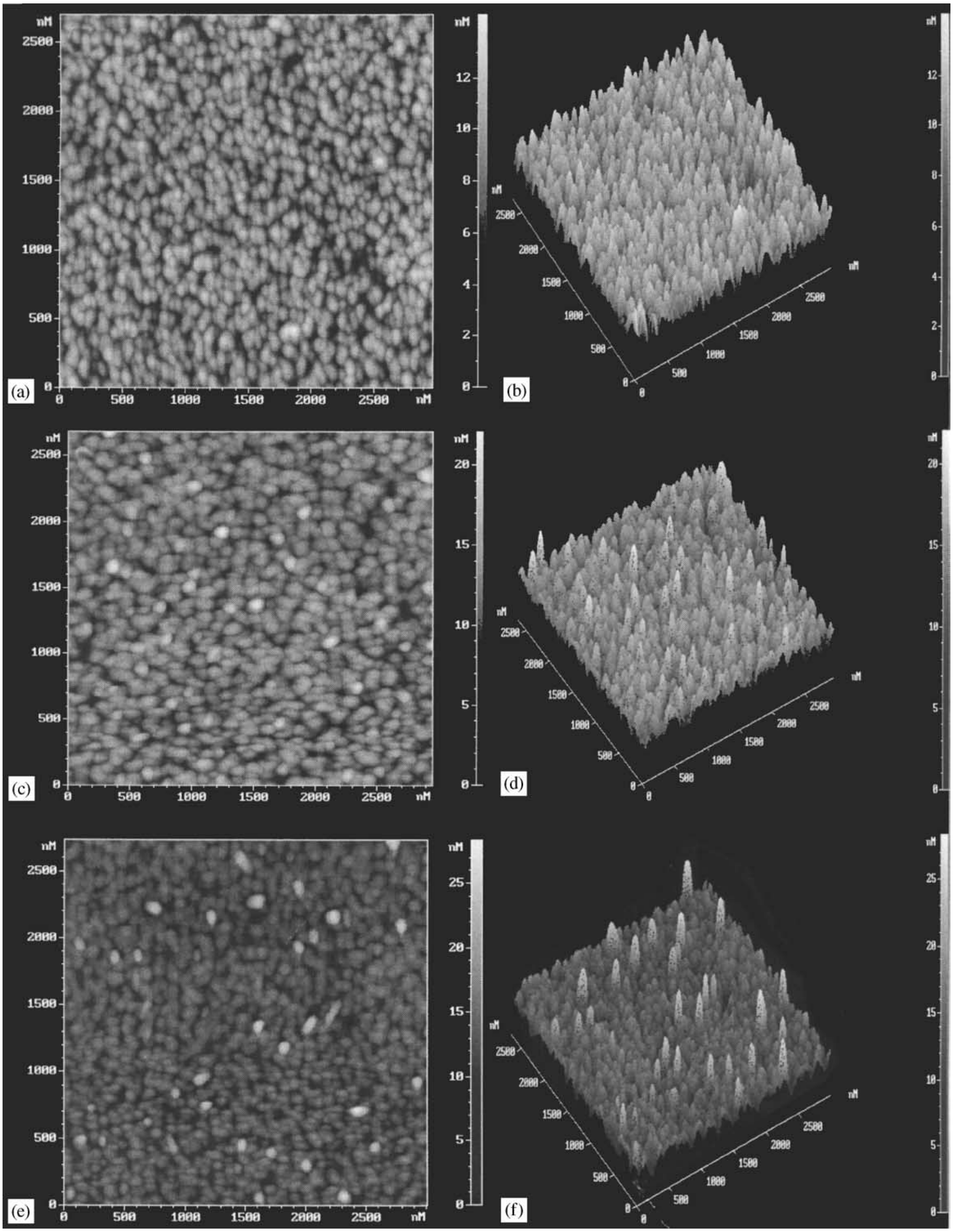

Fig. 1. AFM images of samples 1, 4 and 8. (a), (c) and (e) are plane views of sample 1, 4 and 8, respectively. (b), (d) and (f) are three-dimensional views of samples 1, 4 and 8, respectively.

observed for CdSe/ZnSe/GaAs [9], MnTe/CdTe/GaAs [10], and ZnTe/GaAs [11] QD systems. While, a three-dimensional Volmer–Weber (VW) growth mode has been observed only for ZnSe/ZnS/GaAs QD [12] grown by metal organic chemical vapor deposition (MOCVD). Among the II–VI compound semiconductor QD structures so far studied, only the CdS/ZnSe (ZnSSe) QD structure exhibits type II band alignment [6]. In this study, type II ZnTe QDs were grown in a ZnSe matrix by MBE. The morphology of QD was examined by atomic force microscopy (AFM). A Volmer–Weber growth mode was also identified. According to our results, photoluminescence (PL) studies closely correspond to the AFM results.

Self-assembled ZnTe QDs were grown on a ZnSe buffer layer by applied EPI 620 MBE system. A substrate temperature of 300°C was used. A ZnSe buffer layer with a thickness of approximately 200 nm was grown. The growth rate of the ZnSe buffer layer was about 0.056 nm/s. The growth rate of ZnTe dots was about 0.026 nm/s. The average coverage of ZnTe for samples one to eight was 1.3, 1.9, 2.6, 2.9, 3.2, 3.5, 3.9 and 4.8 monolayers (MLs), respectively. Two sets of samples were grown. The first set was grown for PL measurement. A ZnSe capping layer of about 5 nm was grown following the growth of the ZnTe dots. The other set of samples, for AFM study, were grown without a ZnSe capping layer. The 325 nm line of a He–Cd laser was used to generate the PL spectra. The samples were mounted on the cold finger of a closed-cycle refrigerator, whose temperature was controlled between 10 and 300 K. A SPEX 1403 double-grating spectrometer equipped with a thermal electric-cooled photomultiplier tube was used to analyze the PL spectra. Slit widths were set to 100 μm to yield a spectral resolution better than 0.1 meV.

Fig. 1 shows the plane view and three-dimensional view of samples 1, 4, and 8, obtained by AFM. The formation of self-assembled QDs is observed. In Figs. 1(a) and (b), the average dot size of sample 1, with an average ZnTe coverage of 1.3 MLs, is around 80 nm in diameter and 12 nm high. The dot density of sample 1 is approximately $9.0 \times 10^9/\mathrm{cm}^2$. The total volume of ZnTe dots, obtained by AFM, is very close to that estimated from the growth rate. As shown in Figs. 1(c) and (d), when the average coverage increases to 2.9 MLs, another group of larger ZnTe dots is formed at the cost of a decrease in the number of smaller dots. The average larger dots are 120 nm in diameter and 21 nm high. The density of the smaller dots decreases from $9.0 \times 10^9/\mathrm{cm}^2$ (sample 1) to $4.0 \times 10^9/\mathrm{cm}^2$ (sample 4). The density of the larger dots of sample 4 is approximately $3.6 \times 10^8/\mathrm{cm}^2$. Fig. 1(c) shows ripening of the smaller dots. The two types of dots remain as the average ZnTe coverage increases to 4.8 MLs, as shown in Figs. 1(e) and (f). Fig. 2 shows summary plots of dot density versus ZnTe coverage. The solid squares represent smaller dots and the open squares represent larger dots. Solid and dotted lines guide the viewer. Fig. 2 reveals that the dot density of smaller dots gradually increases as the average ZnTe coverage increases either from 1.3 to 1.9 MLs or from 2.9 to 4.8 MLs. The dot density of larger dots does not increase with average ZnTe coverage. The additional ZnTe coverage increases

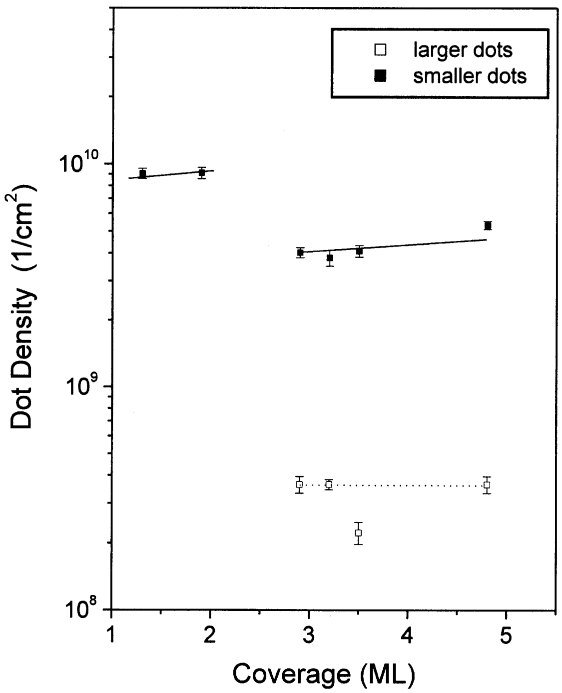

Fig. 2. ZnTe quantum dot density versus coverage. The solid squares represent smaller dots. The open squares represent larger dots. Solid and dotted lines guide the viewer.

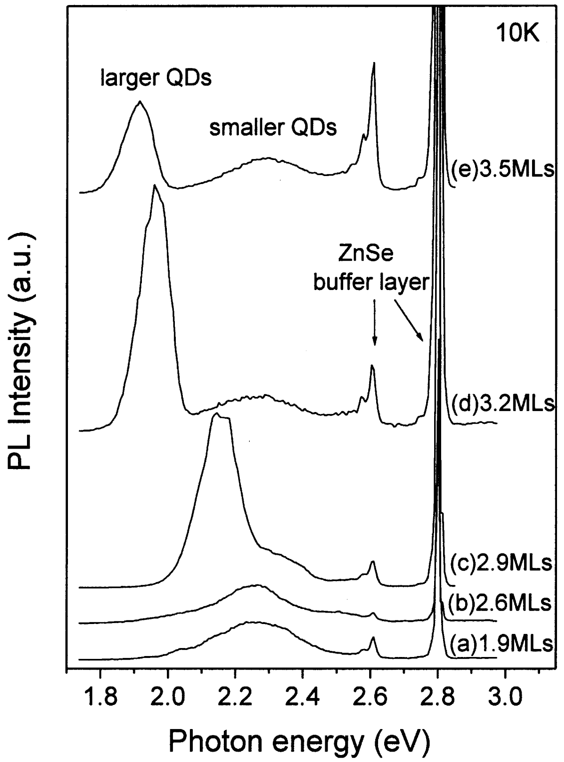

Fig. 3. Photoluminescence spectra of ZnTe quantum dots at 10 K. The average coverage of ZnTe is (a) 1.9 MLs, (b) 2.6 MLs, (c) 2.9 MLs, (d) 3.2 MLs, and (e) 3.5 MLs.

the dot height from 21 nm for sample 4 to 24 nm (sample 8).

Fig. 3 presents the PL spectra. For the 1.9 MLs sample, PL from the near band edge emission of the ZnSe buffer layer is observed near 2.8 eV. The PL structure near 2.6 eV is identified as the defect state of the ZnSe buffer layer. A broad PL structure, which is a signature of PL from ZnTe QD with various dot sizes, is observed at 2.26 eV. An additional peak is observed on the lower energy side of the PL spectrum as the average ZnTe coverage increases to 2.9 MLs. Further increases in the average ZnTe coverage shift the lower energy peak to a markedly lower energy, as shown in the PL spectra of the 3.2 and 3.5 MLs samples. However, the energy of the higher energy peak of ZnTe QD does not move with increasing average ZnTe coverage. The PL results are consistent with the appearance of ZnTe QD with larger dots, which was observed in AFM. The PL

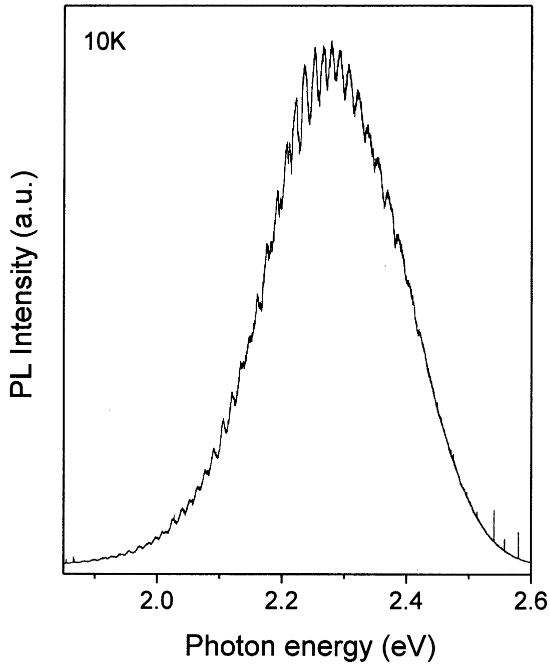

Fig. 4. High-resolution photoluminescence spectrum of sample 3 (2.6 MLs).

results further imply that the smaller dot size of ZnTe QD does not change with average coverage. The size of larger dots increases with average ZnTe coverage, and the PL position shifts to a lower energy as the dot size increases. Also, the energies of both smaller and larger dots are lower than that of the ZnTe epilayer, implying type II band alignment of ZnTe QD grown in the ZnSe matrix. A crucial conclusion from the AFM and PL studies is that ZnTe QD grown on a ZnSe buffer layer (AFM work) or in a ZnSe matrix (PL work) presents Volmer–Weber three-dimensional growth. No wetting layer is observed from the AFM and PL, to result from the two-dimensional S–K growth mode.

Fig. 4 shows a high-resolution PL spectrum of the 2.6 MLs sample. Typical oscillations, reflecting the broad distribution of dot sizes, are present in a broad PL structure of ZnTe QD [13,14]. The energy separation between adjacent peaks ranges from 11 to 18 meV. This separation coincides with neither the longitudinal nor the transverse optical phonon energy, excluding the possibility that the appearance of peaks is due to phonon-related emission. Varying either the dot diameter or the dot height changes the peak position of the oscillations. The lower/higher energy side corresponds to the PL of the largest/smallest QD of the QD group with the smaller dot sizes.

In summary, ZnTe QD structures were grown on GaAs substrates with a ZnSe buffer layer. A three-dimensional Volmer–Weber growth mode was also identified. Two types of dots were observed as well. The PL study demonstrates type II band alignment for ZnTe QD grown in a ZnSe matrix.

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