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Amorphous-Te-mediated self-organization of CdSe/ZnSe nanostructures

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Abstract

Deposition of amorphous Te on a strained epitaxial CdSe layer at room temperature, and its subsequent desorption in the temperature range of 200–310 °C results in novel morphological reorganization of the CdSe surface. Dashes, long chains of quantum dots (QDs), and lateral QD molecules have been observed in atomic force microscopy images of CdSe surfaces, after such a treatment. Te admixture of the QD-layer may not be ruled out. The morphology of the CdSe (Te) surface is strongly governed by the thickness of the deposited amorphous Te layer, but shows almost no dependence on the temperature at which this layer is desorbed. The observed self-ordering of nanostructures over length scales of \sim 600 nm by this process strongly suggests that Te leads to an enormous enhancement of adatom migration. While at low temperature, photoluminescence (PL) consists of emissions from localized excitons in both the observed large QD-based nanostructures and a residual inhomogeneous CdSe(Te) layer, at room temperature the entire contribution is apparently from the former. This leads to a redshift of the spectra, with increase in temperature from 7 to 300 K, by as much as 600 meV. © 2006 Elsevier B.V. All rights reserved.

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In heteroepitaxy nanometer-sized islands are formed when the growth of an adequately strained epi-layer takes place in the Stranski-Krastanow (SK) mode. In this growth mode, elastic relaxation of misfit strain leads to formation of islands beyond a critical thickness of the growing epi-layer. Exploiting this phenomenon, dense ensembles of uniform and coherent (defect-free) quantum dots QDs of several III-V, IV-VI, and IV-IV semiconductor combinations have been fabricated. Unfortunately however, the occurrence of SK growth still remains dubious for the II-VI systems, i.e. CdSe on ZnSe [1,2]. Consequently, several alternative approaches have been developed to induce/enhance QD formation in this system [3–7], one of which, as originally proposed by Tinjod et al., is by depositing and re-desorbing an amorphous group-VI layer onto a supercritically strained two-dimensional layer of the island-forming II-VI material [3]. Deposition and subsequent desorption of a Te- (Se-)amorphous layer in case of CdTe (CdSe) growth on ZnTe (ZnSe) has been demonstrated, by direct imaging techniques, to result in the formation of large, three-dimensional islands [8,9].

In this work, instead of amorphous Se, we used amorphous Te to induce islanding of CdSe on ZnSe. Substitution of Se with Te as the amorphous layer forming material led to the formation of a variety of self-ordered morphologies. While for CdSe coverage of less than ~ 3 monolayers (ML), a low density of large isolated QDs are formed, for CdSe coverage of 3 ML, long dashes, chains, and aligned lateral QD-molecules result. In the latter case, the thickness of the deposited amorphous Te layer appears to govern strongly the final morphology of the CdSe surface. To the contrary, the temperature at which the amorphous layer is desorbed plays no significant role. Room temperature photoluminescence (PL) spectra of these samples are centered at $\sim 2.0 \,\text{eV}$ and reach values down to 1.6 eV. Compared to conventional-molecular beam epitaxy (MBE)-grown QD-layers of comparable

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CdSe coverages, the PL spectra of these samples are redshifted by more than 200 meV [10].

All samples were grown by MBE, in a Riber 2300 MBE chamber equipped with elemental Zn, Cd, Se, and Te effusion cells. Before the growth of the II-VI layers, an undoped GaAs buffer, 200 nm thick, was grown atop the oxide-desorbed, epi-ready GaAs:Si (100) substrates, in another inter-connected Riber 2300 MBE chamber. The typical steps in the growth of the samples are as follows: A ZnSe buffer, 45–50 nm thick, is first grown at 280 °C. The sample is then cooled down to $T_{\rm G} = 230 \,^{\circ}{\rm C}$ under Se flux and a few monolayers (ML) of CdSe are grown by conventional MBE. The as-grown CdSe surface is then cooled down to room temperature and amorphous Te layers, of different thicknesses $t_{\rm D}$, are deposited at a rate of 1 nm min⁻¹. Subsequently, the amorphous-Te-capped CdSe surface is heated up to temperatures between $T_{\rm D} = 200$ and 310 °C, whereby the amorphous cap layer desorbs, leaving behind a re-organized CdSe surface, possibly alloyed with some Te.

The entire process was monitored in situ by reflection high energy electron diffraction (RHEED). High resolution X-ray diffraction (HRXRD) was carried out with a fourcrystal diffractometer using a $\text{CuK}_{\alpha 1}$ source. Atomic force microscopy (AFM) was performed in contact mode in atmospheric ambient. Photoluminescence (PL) spectra were recorded at 7 and 300 K, using a frequency doubled Ti-sapphire laser (420 nm emission line) or a laser diode (405 nm) for excitation, respectively.

Fig. 1(a) shows the surface of a pseudomorphic ZnSe layer, onto which an amorphous Te layer, of thickness $t_{\rm D} = 15 \,\rm nm$ was deposited and subsequently desorbed at $T_{\rm D} = 280 \,^{\circ}$ C. No features are formed and a flat surface with a rms roughness of 0.7 nm results. However, when the surface of a $\Theta = 2.5 \,\text{ML}$ CdSe layer is similarly treated, isolated QDs, 3-5 nm high, 40 nm in base diameter, and with a very low areal density, are formed [Fig. 1(b)]. For $\Theta = 3 \,\mathrm{ML}$ CdSe, a rich variety of self-ordered nanostructures are observed. Dashes, up to 600 nm long and aligned along one of the $\langle 110 \rangle$ axes, are formed for $t_{\rm D} = 1$ nm [Fig. 1(c)]. Increasing $t_{\rm D}$ to 5 nm results in the ordering of long chains of QDs, with QD-heights reaching values of up to 6 nm [Fig. 1(d)]. For $t_D = 60$ nm, lateral QDmolecules, aligned along one of the $\langle 110 \rangle$ axes, are assembled [Fig. 1(e)]. The heights of the QD-pairs vary between 12 and 16 nm, but some of them are as high as 30 nm. In all the above cases $T_{\rm D}$ varied between 280 and 310 °C, except for $t_{\rm D} = 1$ nm, for which the amorphous cap desorbed at about 200 °C. Fig. 1(f) corresponds to a surface for which $t_{\rm D} = 60 \,\mathrm{nm}$ and $T_{\rm D}$ was maintained at 220 °C. Aligned pairs of QDs, similar to those observed in Fig. 1(e) are observed. In this case however, the QDs within the pairs are slightly smaller and closer to each other. Based on the above trends, we observe that the final morphology of the CdSe surface is governed strongly by t_D but is almost independent of $T_{\rm D}$. We note that the observed nonformation of nanostructures on the ZnSe surface proves



Fig. 1. (Color Online) (a) AFM image of a ZnSe surface ($\Theta = 0$) after capping with amorphous Te layers of thickness $t_{\rm D} = 15$ nm and decapping at $T_{\rm D} = 280$ °C. (b)–(f): AFM images of CdSe surfaces corresponding to different values of Θ (ML), $t_{\rm D}$ (nm), and $T_{\rm D}$ (°C). (b) 2.5/15/280 (c) 3/1/200 (d) 3/5/280 (e) 3/60/310 (f) 2.5/60/220.

that the self-organized morphology of the CdSe QD-layers are related to strained CdSe (Te) nanostructures and not to mere droplets of incoherent Te.

Fig. 2 shows the PL spectra corresponding to $\Theta = 2.5 \,\mathrm{ML}$ and $t_{\mathrm{D}} = 15 \,\mathrm{nm}$, and three samples with $\Theta = 3 \text{ ML}$ and $t_{\rm D} = 1$, 5, and 60 nm ($T_{\rm D} = 310 \,^{\circ}\text{C}$), respectively. For all samples the low-energy tail reaches values below 2.0 eV. A bimodal behavior of the spectrum is evident for $\Theta = 2.5 \text{ ML}$, with the peak maximum at 2.45 and a shoulder at 2.2 eV. The maximum corresponds to emission from rough residual layer-like regions between the QDs, while the shoulder represents the emission from the QDs themselves. At room temperature, the PL linewidth is reduced and the maximum is drastically redshifted by $\sim 400 \text{ meV}$. It is attributed solely to the QDs (see inset of Fig. 2). For the sample with $\Theta = 3 \text{ ML}$ and $t_{\rm D} = 1$ nm, the PL maximum lies at 2.35 eV. Increasing $t_{\rm D}$ to 5 nm leads to a 200 meV redshift of the emission. This is attributed to reduced confinement energy of excitons

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