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Layer-by-layer growth and island formation in CdSe/ZnSe heteroepitaxy

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Abstract

Layer-by-layer epitaxial growth of CdSe on ZnSe has been observed at temperatures as low as 170–240 °C. Beyond this range, Reflection high energy electron diffraction (RHEED) specular spot intensity oscillations disappear, signifying a multilayer growth mode. While conventional epitaxial growth of CdSe at ~300 °C results in formation of a rough layer-like structure, a method combining the epitaxial growth of a quasi-two-dimensional (2D) CdSe layer at 230 °C and subsequent annealing at 310 °C induces the formation of discrete CdSe quantum dots (QDs). Here we show that the formation of QDs by this method relies essentially on the layer-by-layer growth of CdSe at 230 °C, which allows an induced roughening of the as-grown 2D layer, during the subsequent annealing step. The layer-by-layer growth mode results possibly due to the nucleation of small and irregular precursor 2D clusters during growth at low temperatures, at the kink sites of which potential barriers for downward inter-layer transport of adatoms is low/absent.

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Almost all practical applications involving heterostructures require the constituent epitaxial layers to be smooth and thereby, the interfaces between two successive layers to be well defined and abrupt. Growth of smooth layers is also important for the epitaxial self-assembly of quantum dots (QDs), since roughening inhibits the formation of a supercritically strained epilayer [1], the necessary ingredient for the Stranski–Krastanow (SK) transition to take place [2]. Appreciably smooth layers of many semiconductors can be grown nowadays, thanks to the state-of-the art epitaxial growth technologies like molecular beam epitaxy (MBE), and the continued efforts in understanding the fundamental mechanisms of crystal growth. Reflection high energy electron diffraction (RHEED) serves as an invaluable tool for the real-time monitoring of the growth front during epitaxy. In conventional understanding of epitaxial growth, RHEED specular spot intensity (SSI) oscillations signify a layer-by-layer growth, wherein, according to the Gilmer–Weeks model [3], random two-dimensional (2D) clusters are nucleated by the impinging adatoms on an initially smooth surface, which subsequently grow in size, coalesce, and finally lead to the completion of the growing monolayer. This requires the adatoms of the growing species to diffuse down from top to the edges of the 2D clusters on rather short time scales. In the ideal case, growth of a new layer begins only after the completion of the preceding layer. Therefore, the growth front in layer-by-layer growth is ideally 2D. When however, the growth temperature is lowered, the RHEED-SSI oscillations are totally/partially suppressed, due to a transition to multilayer growth mode, leading to a rough growth front. The transition is attributed to the reduction in surface diffusivity of the mobile adatoms, whereby downward inter-layer transport of adatoms is hindered. In view of this understanding of epitaxial growth,

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a counter-intuitive phenomenon observed in a few cases over the last decade is the re-entrant layer-by-layer growth, characterized by the reappearance of RHEED-SSI oscillations at low temperatures, where mobility of adatoms is expected to be largely suppressed. The phenomenon has been observed in the homoepitaxial growth of Pt (1 1 1) [4], Ag(1 1 1) [5], GaAs (1 1 1)A [6], and GaAs (0 0 1) [7]. To the best of our knowledge, occurrence of such a phenomenon has so far not been explored in case of epitaxial growth of II–VI semiconductors, albeit such a study is of more significance in this case, considering (a) the severe cation-intermixing propensity [8,9] and (b) a low surface mobility of adatoms, at typical growth temperatures [10]. Here we report the observation of RHEED-SSI oscillations for CdSe heteroepitaxy on ZnSe, in a temperature range of 170–240 °C, and its disappearance above this range. We further demonstrate that formation of CdSe QDs, by a technique combining MBE growth of a CdSe layer at 230 °C and subsequent ramp-up to and annealing at 310 °C, relies fundamentally on the ability to grow relatively smooth epilayers of CdSe within the aforementioned temperature range. The origin of the layer-by-layer growth in CdSe heteroepitaxy on ZnSe, similar to the re-entrant mechanism proposed for the metal- and GaAs homoepitaxy, lies in the enhancement of adatom down-climb from top of small and corrugated precursor 2D clusters. This occurs, despite low adatom mobility, possibly due to breakdown/reduction of potential barriers at the kink sites along the periphery of the 2D clusters.

All samples were grown by MBE, in a Riber 2300 MBE chamber equipped with elemental Zn, Cd, and Se effusion cells. Before the growth of the II–VI layers, an undoped GaAs buffer, 200 nm thick, was grown atop the oxide-desorbed, epiready GaAs:Si (1 0 0) substrate, 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 230 °C under Se flux and a few monolayers (ML) of CdSe are grown by conventional MBE. The as-grown CdSe surface is then capped at 230 °C (by another 45–50 nm thick layer of ZnSe) or ramped up to 310 °C, annealed for 20 min, and then capped at 300 °C—depending on whether a smooth layer or an ensemble of QDs is envisaged, respectively. A few of the samples have also been grown at 300 °C by conventional MBE and without any growth interruption at any stage. RHEED-SSI profiles were recorded using a electron gun operating at a beam energy of 15 keV. The angle of incidence of the electron beam with respect to the substrate surface was fixed at ~3°. Photoluminescence (PL) was excited at about 5 K by an Ar^+ laser (UV multiple lines), analyzed with a 1 m spectrometer with 1200 mm^{-1} grating, and detected with a LN_2 cooled CCD camera. Atomic force microscopy (AFM) images were recorded in atmospheric ambient in contact mode, the cantilever tip radius being <10 nm.

Fig. 1 shows the RHEED-SSI profiles, recorded along the [1 0 0] azimuth during the growth of CdSe at various growth temperatures. It is evident that while at 307 °C the SSI drops monotonically, in the range of 170–240 °C, it shows an oscillatory nature. The growth rate of CdSe measured from the period of intensity oscillations is in very good agreement with that determined a priori by high-resolution X-ray diffraction [11]. However, the RHEED-SSI oscillations in the aforementioned temperature range are strongly damped. This signifies that the layer-by-layer growth mode is only partially recovered, i.e., only a certain fraction of the ultimate monolayer is built before the new layer starts to form.

The partial recovery of the layer-by-layer growth mode, as revealed by the appearance of the RHEED oscillations below a growth temperature of 240 °C, has been beneficially utilized to induce formation of CdSe islands on ZnSe.

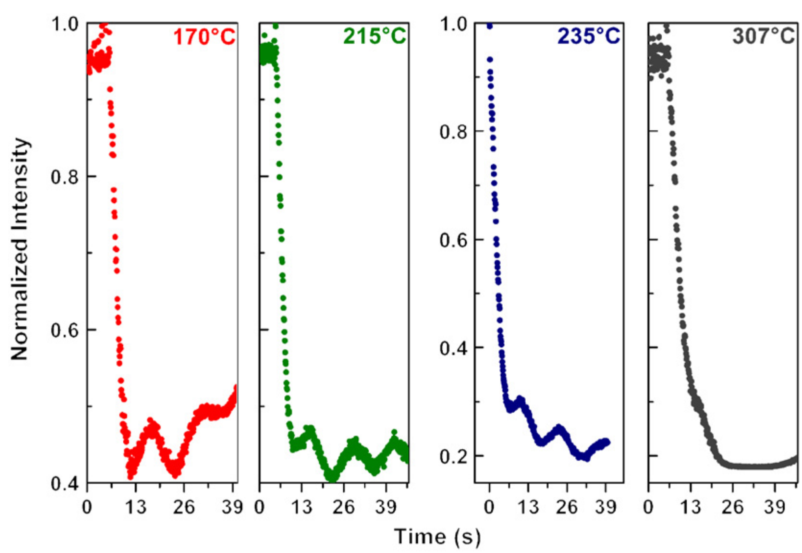

Fig. 1. (Color online) RHEED specular spot intensity profiles recorded along the [1 0 0] azimuth during the MBE growth of CdSe at different temperatures.