Comment on "Dynamics of Ripening of Self-Assembled II-VI Semiconductor Quantum Dots"

In a recent Letter, Lee *et al.* [1] reported on the ripening of CdSe self-assembled quantum dots (QDs). The authors observed postdeposition ripening using an ambient air atomic force microscope (AFM) on a time scale of hours. The observations were interpreted in terms of the theory of Ostwald ripening (OR) as an intrinsic material property of the CdSe/ZnSe heterosystem. The interpretation of *ex situ* taken AFM images from II-VI QD ensembles has been discussed previously, and several papers addressed the problem of an unambiguous assignment of the ambient air surface morphology to the as-grown CdSe QD structure [2–4]. The purpose of this Comment is to show that the observations of Lee *et al.* are not suitable for concluding their statements about the role of OR. Instead, our experimental findings unequivocally demonstrate that CdSe QDs do not ripen on laboratory time scales.

Our investigation is based on an ultrahigh vacuum (UHV)-AFM connected with the molecular beam epitaxy growth chamber. The CdSe-on-ZnSe QDs were grown as described previously [5]. After QD formation, revealed by the transition from a streaky to spotty reflection high energy electron diffraction pattern, the sample was cooled down to room temperature and transferred into the UHV-AFM. There, the uncapped surface was investigated in noncontact mode, starting 15 min after growth stop and continuously scanning the same sample area for a period of about 120 h. Figures 1(a)–1(d) clearly show that the topography of the sample surface remains entirely unchanged within 5 days. The QDs have a density of about 7×10^{10} cm^{-2} and background-corrected heights between 1.5 and 3.5 nm. Their lateral extension seen in the AFM images of about 30 nm is not tip radius deconvoluted and represents thus an upper limit. The QDs are situated on the CdSe remnant layer, which exhibits a long-ranged "cloudy" roughness. We note that this roughness, corresponding to that of the 1 μm thick ZnSe buffer, is not resolved in phase images, as presented by Lee *et al.* Neither the density nor the geometric dimensions of the CdSe QD ensemble change over the examined time period. The occurrence of an apparent, very weak, double image laterally shifted in the scanning direction is an experimental artifact, caused by a small deformation of the tip. In a long-term experiment, we have confirmed the QD stability over a period of 50 days. Exposure of the uncapped structure to air has no influence on the QDs and their stability; subsequently taken AFM images demonstrate, however, the appearance of new entities with much larger sizes, comparable to the ones described in Ref. [1]. It is clear from our data that these features are not intrinsically correlated with the CdSe surface.

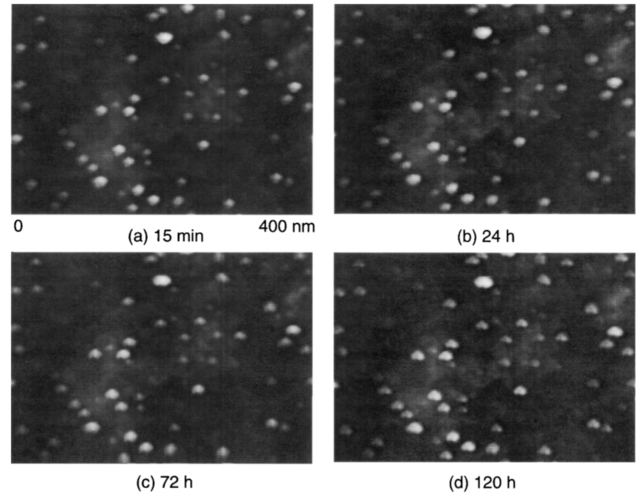

FIG. 1. *In situ* UHV-AFM images of CdSe self-assembled QDs on a ZnSe buffer. Topography of sample surface (a) directly (15 min), (b) 24 h, (c) 72 h, and (d) 120 h after QD formation.

The absence of OR indicates that—after a part of strain is relaxed through QD formation—the kinetic barrier for the edge detachment of single atoms dominates over the reduction of surface energy. A more detailed report on our findings and conclusions about the growth mechanism and stability of II-VI QDs is in progress.

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