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De-relaxation of plastically relaxed InAs/GaAs quantum dots during the growth of a GaAs encapsulation layer

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Abstract

The morphological modifications of plastically relaxed InAs/GaAs quantum dots (QDs) are studied by transmission electron microscopy (TEM) and interpreted on the basis of energy calculations. The growth of a GaAs cap-layer over the dislocated QDs leads to a significant increase of their size, and to a decrease of their In-composition, due to In/Ga exchange and interdiffusion reactions. A mechanism of de-relaxation of the QDs leading to the disappearance of the dislocation network at the bottom InAs/GaAs interface is observed as a consequence of these structural modifications. Our calculations show that this de-relaxation process is energetically favorable below a critical In-composition of the QDs, which is reached during the growth of the GaAs cap-layer due to In/Ga interdiffusion.

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1. Introduction

The III–V semiconductors quantum dots (QDs) have been intensively studied over the past two decades. In fact, the original physical properties related to the three-dimensional confinement in these nanostructures are very interesting for a wide range of applications. Among the various material systems involved in these works, InAs/GaAs QDs appear now as a reference. The initial stage of the re-growth of GaAs over InAs/GaAs QDs has been shown to strongly affect their size, shape and composition due to In/Ga exchange and interdiffusion reactions. Thus, these effects strongly modify the composition profile of the QDs [1]. Shape transitions from QDs to ring or camel-like nanostructures have also been observed. They have been interpreted as resulting from the combined influence of the In/Ga interdiffusion, the difference in the diffusion velocities of In and Ga adatoms, and the strain relaxation at the apex of the QDs that hinders the incorporation of Ga on the top of the nanostructures [2,3]. The high lattice mismatch between InAs and GaAs (about 7%) is the driving force for the formation of QDs. This strong lattice mismatch can also lead to the formation of plastically relaxed QDs, particularly in growth conditions enhancing an efficient diffusion of the species adsorbed on the growth surface. Thus, the formation of big plastically relaxed InAs clusters on GaAs has been reported. It has also been shown that when partially covered by GaAs, these big clusters could be re-evaporated during growth stops carried out under As-rich ambient [4–6]. Apart from this particular case of re-evaporation, it is commonly admitted that once plastically relaxed, InAs QDs act as sources of dislocations that propagate through the structure during the growth, thus leading to a degradation of the QD photoluminescence yield.

In this article, we present a detailed study of the effect of the encapsulation by GaAs of plastically relaxed InAs QDs grown by low-pressure metalorganic vapor phase epitaxy (LP-MOVPE). A mechanism of de-relaxation of small

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plastically relaxed QDs during their encapsulation by GaAs is evidenced and described on the basis of transmission electron microscopy (TEM) studies.

2. Preparation of the samples

The samples studied in the following were grown by LP-MOVPE, and observed by TEM. The growth was performed on (0 0 1)-oriented semi-insulating GaAs substrates at a work pressure of 60 Torr. A GaAs buffer, a 30 nm thick $Al_{0.35}Ga_{0.65}As$ layer and a 47 nm GaAs layer were first deposited at 670 °C. After a growth interruption under arsine, during which the temperature was ramped down to the growth temperature of the QDs array (460 °C), 3 nm of GaAs were deposited, and the QDs were formed depositing 2.1 monolayers (ML) of InAs at 0.08 $ML\,s^{-1}$. For sample A, the temperature was then quickly ramped down and the growth was stopped. For sample B, the QDs were encapsulated by a 4 nm thick GaAs overlayer grown at 460 °C before to stop the growth. For a detailed description of the QDs growth conditions and mechanisms, see Ref. [7]. Both samples were then cleaved and thinned for TEM plane-view and cross-sectional observations. For plane views, the backsides of the samples were first mechanically and then chemically thinned with a bromine–methanol solution until sufficiently thin to transmit the electron beam. For the cross-sectional views, thin foils were prepared by mechanical polishing followed by milling with 2.5 keV Ar^+ ions. In the final stage, we add a reactive iodine vapor to remove most of the amorphous layers left on the thin foil surfaces by ion etching. The images were obtained in a TEM Philips CM20 "supertwin" equipped with a "Megaview" charge coupled device camera from Soft Imaging System, Munster, Germany.

3. Experimental results

A TEM plane view of sample A, taken under zone-axis bright field conditions, is shown in Fig. 1. This sample contains two types of three-dimensional structures, namely big clusters and small QDs. Big clusters have an average diameter of 90 nm and an average height of 6 nm, as deduced from atomic force microscopy experiments (not shown here). Their density is about $1.10^9\,cm^{-2}$. Small QDs present an average lateral size of 25 nm, a density of $4.10^8\,cm^{-2}$ and an average height of 3.1 nm. Both these structures present moiré fringes, typical for plastic relaxation: these moiré fringes result from the interference between the electron beams diffracted by the substrate (GaAs) and the material forming the three-dimensional structures. Both big clusters and small QDs are therefore plastically relaxed on GaAs. The simultaneous presence of big and small plastically relaxed QDs is characteristic of our LP-MOVPE growth conditions: this growth technique leads to very large surface mobility of the adatoms, enhancing and accelerating the QD plastic relaxation process. In our growth conditions, this bimodal distribution of plastically relaxed QDs is always observed at the early stage of the epitaxy, even for high InAs growth rates and low InAs thickness. The step of the moiré fringes depends on the in-plane lattice mismatch between the substrate and the material constituting the relaxed structure, i.e. on the composition of the clusters and QDs, as well as on their plastic relaxation rate. Assuming a complete plastic relaxation, we found an In composition of 88% for the big plastically relaxed clusters, and of 70% for the small relaxed QDs. The hypothesis of a complete plastic relaxation is supported by our experimental observations: the distance between subsequent dislocations in the small plastically relaxed QDs is approximately constant (Fig. 2). Partial plastic relaxation would have led to an inhomogeneous repartition of the misfit dislocations inside the QDs. A weak-beam TEM plane view of sample A is presented in Fig. 2. In these imaging conditions, dislocations for which vanishing conditions are not satisfied appear as bright lines in the cliché. In the particular conditions of Fig. 2, one observes dislocations which lines are perpendicular to *g*. This indicates that the dislocations are pure edge dislocations with Burgers vectors in the interface (Lomer–Cottrell dislocations), as expected for highly mismatched systems. Fig. 2 confirms that both big clusters and small QDs are dislocated. The total misfit dislocation density is $0.45\,cm^{-1}$, taking into account the fact that only half of the misfit dislocation network is imaged on Fig. 2 (the other half can be imaged by rotating **g** of 90°). This dislocation density was estimated by

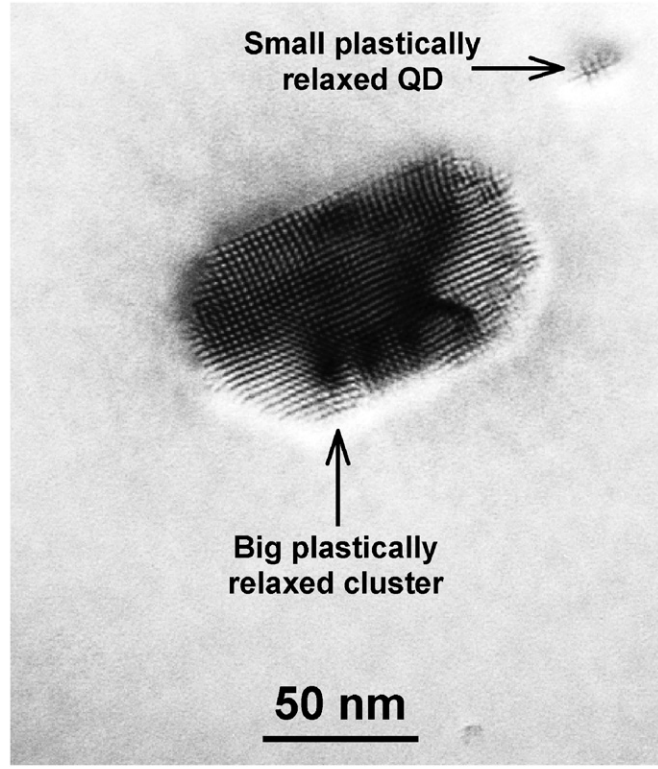

Fig. 1. TEM plane view of sample A (uncapped structure), taken under 0 0 1 zone-axis bright-field conditions.