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Ab initio study of In adsorption on $Al_xGa_{1-x}As$ substrates at the first stages of droplet epitaxy

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Abstract. In this work we study the adsorption of In on $Al_xGa_{1-x}As$ substrates at the first stage of droplet epitaxy with goal of explaining the anomalous behavior observed in previous experiments where an increase in the content of chemically active Al in $Al_xGa_{1-x}As$ substrates led to an increase in the surface mobility of In adatoms affecting the final distribution of InAs quantum dots. DFT simulations showed that when In adatoms directly interact with As-terminated substrates, there is no deviation from normal behavior, but when In binds with a fully formed In wetting layer, the results are in agreement with those observed in our experiments – increasing Al content led to a decrease in adsorption energy, which means that surface mobility of adatoms has increased. We assume that this specific effect of droplet epitaxy is caused by lack of a stabilizing As layer, allowing the formation of In dimer rows and direct interaction of adatoms with them.

Keywords: density functional theory, III-V, indium, surface mobility, quantum dots, droplet epitaxy, molecular beam epitaxy

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Моделирование адсорбции In на подложках $Al_xGa_{1-x}As$ в течение первых стадий капельной эпитаксии

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Аннотация. В данной работе проведены исследования адсорбции In на подложках $Al_xGa_{1-x}As$ на начальной стадии процесса капельной эпитаксии с целью объяснить аномальное поведение, наблюдающееся при росте квантовых точек этим методом – увеличение содержания химически активного Al в подложках $Al_xGa_{1-x}As$ приводит не к уменьшению, а к увеличению подвижности адатомов In, влияя на конечное распределение квантовых точек InAs. DFT моделирование показало, что при взаимодействии адатомов In непосредственно с подложкой, Al снижает поверхностную подвижность адатомов, тогда как формирование промежуточного смачивающего слоя In между подложкой и адатомами приводит к изменению зависимости на противоположную – увеличение

содержания Al приводит к уменьшению энергии адсорбции. Мы предполагаем, что данный эффект проявляется в капельной эпитаксии ввиду смены типа поверхностной структуры с As- на Me-стабилизированную, что приводит к непосредственному взаимодействию адатомов In с образовавшимся реконструированным смачивающим слоем.

Ключевые слова: теория функционала плотности, A3B5, индий, поверхностная подвижность, квантовые точки, капельная эпитаксия, молекулярно-лучевая эпитаксия

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Introduction

Use of InAs/AlGaAs quantum dots for single-photon emission requires low density (< 10^8 cm^{-2}), high uniformity and reproducibility of the size, shape and distribution of quantum dots (QD). When growing QDs in the Stranski-Krastanov (S-K) mode on the surfaces of Al containing substrates, it is expected that high chemical activity of Al will inhibit the adsorbates mobility, allowing the adsorbates to form stronger bonds with the surface [1]. This means that increase in the Al content leads to an increase in the QD density, which means that the adatom mobility decreases.

At the same time, in droplet epitaxy (DE) the effect is the opposite – as shown in our previous work [2, 3], an increase in the Al content leads to a decrease in the droplet density, which means that the adatom mobility during the wetting layer formation [4] increases. At low growth temperatures (less than 300 °C), the influence of the substrate composition is relatively small: the average diameter of the droplet structures increases from 16 to 21 nm at 150 °C and from 35 to 41 nm at 200 °C with an increase in the aluminum fraction from 0 to 1. The density decreases by 1.5 – 2 times: from $3{\cdot}10^{10}$ cm^{-2} to $2{\cdot}10^{10}$ cm^{-2} at 150 °C and from $3{\cdot}10^{9}$ cm^{-2} to $1.7{\cdot}10^{9}$ cm^{-2} at 200 °C (Fig. 1).

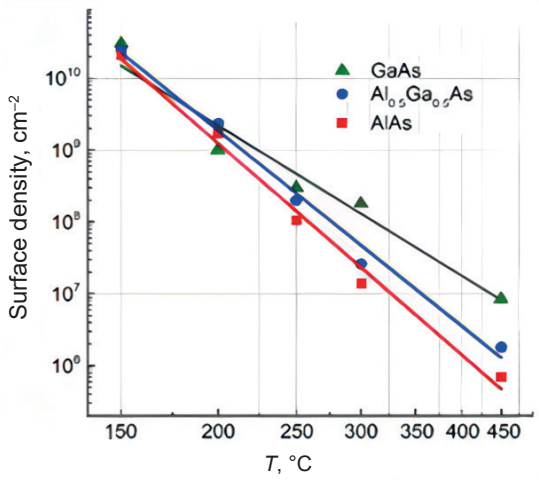

Fig. 1. Surface density of nanostructures obtained by droplet epitaxy after deposition of 3 ML of In on the GaAs, $Al_{0.5}Ga_{0.5}As$ and AlAs surfaces at different substrate temperatures

In the region of relatively high growth temperatures (high for droplet epitaxy of indium), the effect of the Al content is more pronounced. The density of indium droplet structures decreases by an order of magnitude: from $1.8 \cdot 10^8$ cm^{-2} down to $1.4 \cdot 10^7$ cm^{-2} at a substrate temperature of 300 °C and from $8.5 \cdot 10^6$ cm^{-2} to $7 \cdot 10^5$ cm^{-2} at 450 °C. In this case, the average size of droplet nanostructures doubles: from 98 to 176 nm at 300 °C and from 230 to 437 nm at 450 °C.

To see if this effect can be detected from first principles calculations, we modeled GaAs, $Al_{0.5}Ga_{0.5}As$ and AlAs covered by 0.25 ML and 1.25 ML of In (coverages representative of the first stages of droplet epitaxy), and calculated the resulting adsorption energies, which describe strength of bonding between the substrate and adatoms moving over it. The adsorption energy directly affects the adatom mobility, since the stronger the bond the adatom forms with the surface, the more difficult it becomes for it to move, which leads to a proportional decrease in the adatom mobility.

Methods

The adsorption energy was obtained using formula (1)

$$Eads_{0.25ML(1.25ML)} = E_{A3B5(+InML)+In} - E_{A3B5(+InML)} - E_{In}, \quad (1)$$

where $E_{A3B5(+InML)+In}$ is the total free energy of GaAs (covered with 1 ML of In) with an In atom on top, $E_{A3B5(+InML)}$ is the energy of GaAs (covered with 1 ML of In), and E_{In} is the energy of an indium atom in vacuum. A more negative value of *Eads* means a stronger bond between the adsorbate and the surface, which suppresses the adatom mobility, resulting in a higher density of nanostructures.

Ab initio calculations of the slab models were carried out using density functional theory (DFT). It was applied using the Vienna ab initio simulation package [5] with the projector augmented wave potential construction. For approximating the exchange-correlation functional we used Perdew's generalized-gradient-approximation (PBE) [6]. DFT+U method based on Dudarev's approach [7] was used to properly describe the interactions of p-band electrons. Simulation of parameters affected by random distribution of atoms in $Al_{0.5}Ga_{0.5}As$ alloy was achieved by use of special quasirandom structures [8]. After convergence testing K-point sampling density and cutoff energy values were set to 6x6x1 and 400 eV with higher values giving energy difference of less than 0.001 eV.

Since *Eads* would be different at different points of the surface, we calculated a potential energy surface (PES) plot, showing an approximate map of *Eads* obtained by interpolating the results of calculations over the surface on a uniform 5×5 grid. In the case of the 0.25 ML coverage calculations (Fig. 2 upper row), we put a single In adatom at grid coordinates over the As-terminated surfaces (which relax by forming As-dimer rows). In the case of a 1.25 ML coverage (Fig. 2 lower row), we study the interaction of the In adatom with the existing wetting layer (WL). This In WL also forms dimer rows during relaxation.

Results and discussion

Figure 2 shows the highest adsorption energy (which corresponds to the most favorable adsorption position) found after placing In adatoms at adsorption points forming a uniform 5×5 grid over the substrate surface for all coverages and substrates. Upper row of PES plots shows that max *Eads* changes with Al content at a low coverage of 0.25 ML. An increase in Al content leads to an increase in max *Eads* from −3.453 up to −3.823 eV for GaAs and AlAs, respectively, which means that adatom mobility decreases. This dependance matches the one seen in the S-K growth mode.

However, calculations at completely metal stabilized 1.25 ML coverage (Fig. 2 lower row), have shown that when In directly interacts with the dimer rows of the reconstructed In wetting layer (the relaxation step during the calculation results in the formation of dimer rows consisting of In adatoms), the behavior changes to one seen in our droplet epitaxy experiments, where an increase in the content of chemically active Al leads to a decrease in the adsorption energy from −4.289 down to −3.989 eV and consequently higher surface mobility. This shows that the observations obtained in our experiments are in good agreement with theory and can be explained by the nature of droplet epitaxy, in which an In wetting layer forms in the absence of a stabilizing As flux.

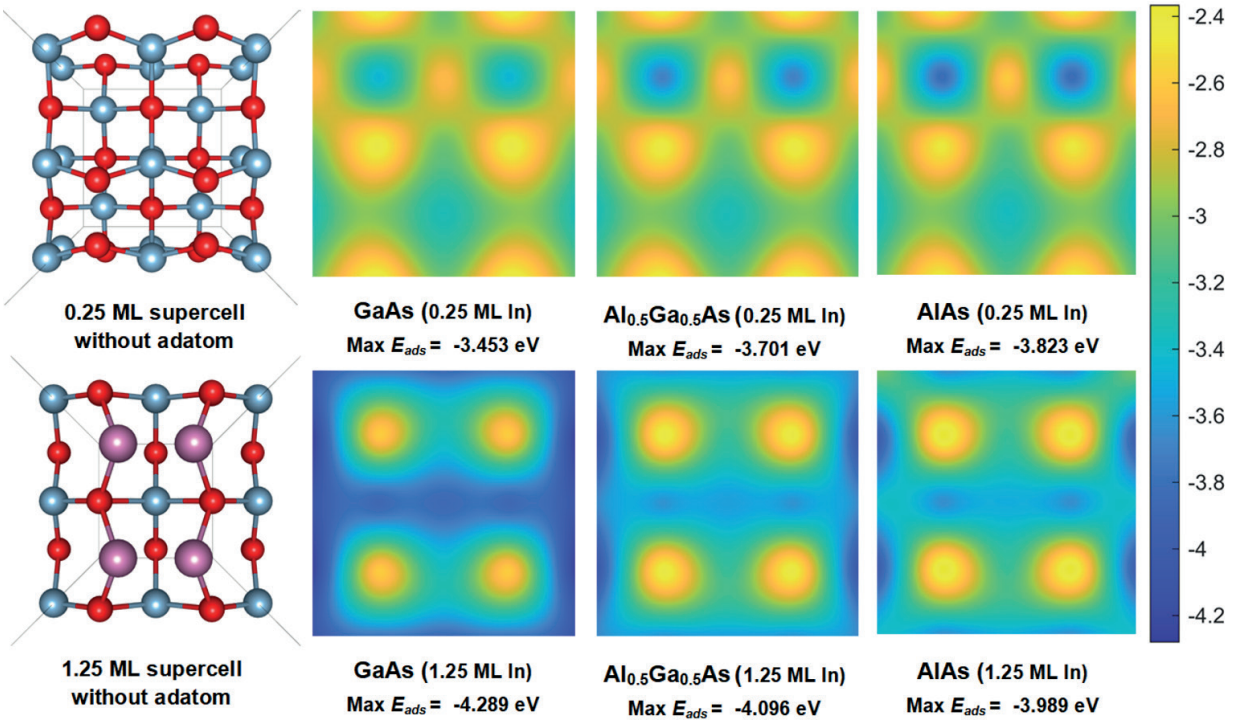

Fig. 2. PES plots showing *Eads* of In adatom on surfaces of $Al_xGa_{1-x}As$ substrates at 0.25 ML (1 In adatom) and 1.25 ML (4 In WL atoms + 1 In adatom) wetting layer coverage

This means that the change in the dependance of adatom mobility on Al content can be explained by the absence of a stabilizing As layer, which usually prevents the formation of In dimer rows and does not allow direct bonding of In adatoms to the already formed In wetting layer. These differences in the droplet epitaxy and S-K growth can be responsible for the studied effect, making them the subject of our future studies.

REFERENCES

1. **Andrews A. M., et al.,** Independent control of InAs quantum dot density and size on $Al_xGa_{1-x}As$ surfaces. Journal of Materials Science: Materials in Electronics. 19 (2008) 714–719.

2. **Balakirev S.V., et al.,** Anomalous behavior of In adatoms during droplet epitaxy on the AlGaAs surfaces. Nanotechnology. (31) (48) (2020) 485604.

3. **Chernenko N.E., et al.,** Effect of the Al content in the substrate on the In nanodroplets growth by droplet epitaxy. Journal of Physics: Conference Series. (1695) (1) (202).

4. **Dukhan D.D., Balakirev S.V., Voloshina E.N., Solodovnik M.S.,** Ab initio modelling of In wetting layer formation on As-stabilized GaAs during first stages of droplet epitaxy, St. Petersburg State Polytechnical University Journal. Physics and Mathematics. 16 (3.1) (2023) 193–197.

5. **Kresse G., Furthmüller J.,** Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set. Physical review B. 54 (16) (1996) 11169.

6. **Perdew J.P., Burke K., Ernzerhof M.,** Generalized gradient approximation made simple. Physical review letters. 77 (18) (1996) 3865.

7. **Dudarev S.L., Botton G.A., Savrasov S.Y., Humphreys C.J., Sutton A.P.,** Electron-energy-loss spectra and the structural stability of nickel oxide: An LSDA+ U study. Physical Review B. 57 (3) (1998) 1505.

8. **Mäder K.A., Zunger A.,** Short-and long-range-order effects on the electronic properties of III-V semiconductor alloys. Physical Review B. (51) (16) (1995) 10462.

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