

# Epitaxial Growth of a Thin Film and Quantum Dots

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**Abstract:** The authors give a brief review on people's general view on the epitaxial growth of a thin film via the layer-by-layer growth mode. Then they have a remark on the self-assembling of quantum dots via the epitaxial growth to notice its complexity and nonlinear behavior. Furthermore, based on the further analysis of the experimental data published previously, the authors have an estimate of the time scale for the growth of a quantum dot and demonstrate that it is an unexpectedly rapid process. Finally, the authors propose a preliminary conceptual framework within which the formation of quantum dots can be understood in the InAs/ GaAs (001) system.

**Key words:** epitaxial growth; thin films; self-assembled quantum dots; InAs/ GaAs (001)

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## 薄膜和量子点的外延生长

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**摘要:** 首先简短地综述了人们关于外延薄膜材料层状 (layer-by-layer) 生长机制的认识; 给出了作者关于自组装量子点外延生长过程的评价和观点, 强调了量子点自组装生长过程的复杂性和非线性性质。在对已经发表过的实验数据进一步分析的基础上, 作者对一个量子点自组装生长形成所需要的时间作了一个估算, 说明这是一个非常快的过程 ( $< 10^{-4}$  s)。最后, 作者提出了一个理解量子点自组装生长过程机制的模型。

**关键词:** 外延生长; 薄膜; 自组装量子点; InAs/ GaAs (001)

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## 0 Introduction

Epitaxial growth of a material on a substrate is very important in a variety of modern practical devices. Although the growth of an epitaxial film

with thickness control exact to an atomic layer is a routine nowadays by using molecular beam epitaxy (MBE) and metallic organic chemical vapor deposition (MOCVD), many aspects of the relevant underlying physical processes remain elusive and people

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微纳电子技术第47卷第6期 321

have always been very curious to them. About thirty years ago, when people's concern was to obtain an epitaxial film with two-dimensional (2D) sharp interface, their desire in episteme on epitaxial growth may arise primarily from their intrinsic curiosity on the knowledge unknown to them, as both the MBE and MOCVD are born successful in producing a thin films in the atomic layer-by-layer growth mode, needless of any additional effort in making an atomic flat surface or a sharp interface required in a practical device. With the advent of the concept of the self-assembling of quantum dots (QDs) at about 1990's, the situation changes and the understanding of the atomic processes underlying the epitaxial growth was promoted to be compelling.

QDs are three-dimensional (3D) in structure with the characteristic length scale in nanometer. An extensive and arduous investigation on QD's has been performed over more than twenty years. However, the QD's are very complex and seems to be a real challenge to people, and the tremendous efforts in QD's investigation result in a great variety of controversies rather than a consensus in many aspects of the problem. Up to the date, the QD's intrinsic physical properties and structural properties remain to be solved, and their marvelous practical applications as expected from theory are mostly still kept in potential. In order to make a further progress in QD's investigation and application, the understanding of how the QD's epitaxial growth proceeds seems to be indispensable and necessary.

There are two purposes for this article. The first purpose is to give a review, from the author's perspective view with the emphasis on simplicity and brevity, on how people understand the epitaxial growth of a film in terms of atomic events. The second purpose is to propose a novel interpretation of the QD's self-assembling, based on the analysis of our experimental data as obtained from the InAs/GaAs (001) system in our own laboratory.

## 1 Layer-by-Layer Growth

During an epitaxial growth of a metal on a metal with a simple lattice structure and without a surface reconstruction, if the supersaturation is relatively low and the ratio of the atomic diffusion constant  $D$  to the deposition flux  $F$  is relatively high (for example,  $D/F > 10^5$ ), the epilayer growth may proceed in the layer-by-layer growth mode. Ideally, the growth process is simplified to be cyclic and successive completions of an atomic layer. The formation of a complete atomic layer on the growing surface is separated into several regimes. The first is the nucleation regime during which the adatoms from the deposition flux diffuse on the surface and meet with one another to form nuclei of a few atoms (two atoms in an irreversible growth). The adatom diffusion rate is determined by the Arrhenius relation  $D = v_0 \exp(-E_s/kT)$ , where  $v_0 = 10^{13}/s$  and  $E_s$  is the interaction energy between an adatom and the substrate surface and is 1.0 eV for metals. At the end of the nucleation regime, the areal density of nuclei approaches a saturation value, and the second regime, aggregation regime, begins. In the aggregation regime, there are almost no nucleation events and most of the deposited adatoms are incorporated into the 2D growing islands developed from the nuclei formed in the first regime. The last regime for the completion of an atomic layer is the coalescence regime, during which the relatively large 2D islands coalesce to form a complete atomic layer, and then a next atomic layer begins to form. The nucleation and aggregation regime are relatively simple and can be treated with the mean-field rate equations, kinetic Monte Carlo simulations, and scaling arguments, in addition to the direct observation by using STM. It has been well established that in these two regimes, the regularities can be summarized into several scaling laws

$$N_{\text{isl}}(\theta) \sim \theta^{-z} \quad (1)$$

$$s_{\text{av}} \sim \theta^{-z} \quad (2)$$

$$N_{\text{isl}} \sim [D/F]^{-z} \quad (3)$$

$$N_{\text{isl}}(s) = (s/s_{\text{av}})^{-z} f(s/s_{\text{av}}) \quad (4)$$

Here,  $\theta$  is the coverage as deposited,  $N_{\text{isl}}$  the total number density of 2D islands,  $s_{\text{av}}$  the average island size,  $T$  the substrate temperature,  $N_{\text{isl}}(s)$  the number density of islands containing  $s$  atoms, with the three exponents  $\alpha$ ,  $z$ , and  $\beta$  being positive in sign and one order in magnitude<sup>[1-4]</sup>.

The layer-by-layer growth occurs also for the homoepitaxial growth of semiconductors, for example, GaAs (001), although the situation on a semiconductor surface is much more complex because of the existence of the two components and surface reconstruction. The completion of a molecular layer is also reducible to atomic events, similar to the simpler case of a metal. The atomic events on a semiconductor that should be taken into account in interpreting the epitaxial growth are much more numerous than on a metal<sup>[5-6]</sup>.

As said above, the nucleation and aggregation of 2D islands on a growing surface are understandable in terms of single atomic events, such as atomic deposition, diffusion and attachment, etc. However, according to such a scenario, when the lateral dimension of a 2D island is large enough, the nucleation events should happen on the top of these islands<sup>[7]</sup>, and the 3D growth may occur. More contradictorily, in the case of homoepitaxial growth of GaAs on a vicinal (001) substrate, J. H. Neave et al<sup>[8]</sup> observed that the layer-by-layer growth could transform from step-flow mode to nucleation-growth on the terraces with decreasing the growth temperature. They interpreted the transformation in growth mode as follows: as the growth temperature is decreased, the atomic diffusion length is reduced below the terrace width on the vicinal GaAs (001) surface, and adatoms can no longer reach the atomic step edges of a terrace;

then they themselves nucleate into new 2D islands which subsequently grow in lateral dimension. Although this famous work seems to provide a direct experimental evidence for the layer-by-layer growth for GaAs/GaAs (001), it also provokes, from the author's perspective, a paradox: the growth mode can transform from step-flow to the nucleation-growth on the terraces with reducing the atomic diffusion length; similarly, the nucleation should also happen on the top of the 2D islands, which may be much larger in lateral dimension than a terrace width. In reality, this does not happen. Such a paradox demonstrates that the layer-by-layer growth cannot be completely interpreted in terms of deposition, diffusion and incorporation, and some kind of motion more complex than these individual atomic events should be involved, which needs further investigation in depth.

## 2 Self-Assembling of Quantum Dots: General Remark

As said above, the epitaxial growth of a film via the layer-by-layer mode can be reduced to a series of individual atomic events, and such a reductionism is successful in some aspects of the problem, for example, the nucleation and growth in the submonolayer regime. Scaling relations (1) - (4) are universal, regardless of the material characteristics and the detail in the epitaxial growth conditions, these are the aesthetical regularities in physics resulting from the stochastic and random individual atomic events. In comparison, the self-assembling of QD's is much more complex. QD's epitaxial growth process seems to be highly nonlinear and sensitively dependent on the growth parameters. Although a great amount of experimental data have been accumulated and a large number of theoretical discussions have been presented in the literature, little consensus or universal characteristics of QD's self-assembling have been established up to the date. Therefore, it is very difficult to

have a comprehensive review on the epitaxial growth of quantum dots as well as its physical properties.

The understanding of the epitaxial growth of quantum dots is a great challenge to people! There is a general trend to model the formation of QD's in the individual atomic events<sup>[9-18]</sup> with considering on the influence of misfit strain, as for the layer-by-layer growth. However, the former should be much complex than the latter, as the former may involve a large amount of atomic events simultaneously. Therefore, such a reductionism may not work for the QD's problem, because it cannot properly take into account of the possible complexities and collective behaviors of atoms in large number.

### 3 Self-Assembling of Quantum Dots in InAs/ GaAs (001)

The InAs/ GaAs(001) system is one of the paradigms for the fabrication of quantum dots by using the MBE method, both in fundamental research and practical application. In the conventional MBE, the InAs QDs can be grown in a large range of growth parameters, such as substrate temperature, incident flux, substrate orientation, and so on. In addition, the InAs QDs can also be fabricated by using a number of modified MBE methods, such as mobility enhanced epitaxy<sup>[19]</sup> and droplet epitaxy<sup>[20]</sup>, which are remarkably different from each other in the growth condition. Therefore, it seems almost impossible that there is a general atomic kinetic pathway, which is the purpose of many theoretical investigations, for the formation of InAs quantum dots universally applicable to such a great variety of the growth conditions. In this section, based on the analysis of our experimental data on the quantum dots in InAs/ GaAs (001)<sup>[21-23]</sup> as obtained by using the conventional MBE method at a lower growth rate of 0.1 ML/s (monolayer per second) and the substrate tempera-

ture  $T_s = 500$  and a higher rate of 1.0 ML/s with  $T_s = 520$ , we propose a novel explanation for the QD's epitaxial growth.

#### 3.1 InAs QD's Epitaxial Growth Behavior

The InAs QDs fabricated by using the two different MBE growth conditions, 0.1 ML/s at 500 and 1.0 ML/s at 520, are significantly different from each other in the epitaxial growth behavior<sup>[22]</sup>, which is demonstrated in the evolution of an ensemble of QDs with increasing InAs coverage. Fig. 1 shows the QD's areal density  $N$  (eff) versus the effective InAs coverage  $\theta_{eff} = \theta - \theta_c$ , where  $\theta$  is the nominal InAs coverage and  $\theta_c$  is the critical coverage at which the 2D/3D growth mode occurs. For the case of 0.1 ML/s and 500, the experimental data in the Fig. 1 (a) can be fitted into two successive power laws<sup>[21]</sup>,  $N \propto \theta_{eff}^{1.8}$  and  $N \propto \theta_{eff}^{0.6}$ . For the case of 1.0 ML/s and 520, as shown in Fig 1(b), the density  $N$  varies with  $\theta_{eff}$  in accordance with the exponential function  $N = N_0 e^k \theta_{eff}$  in the  $\theta_{eff}$  range of about 0 - 0.05 ML. With the effective

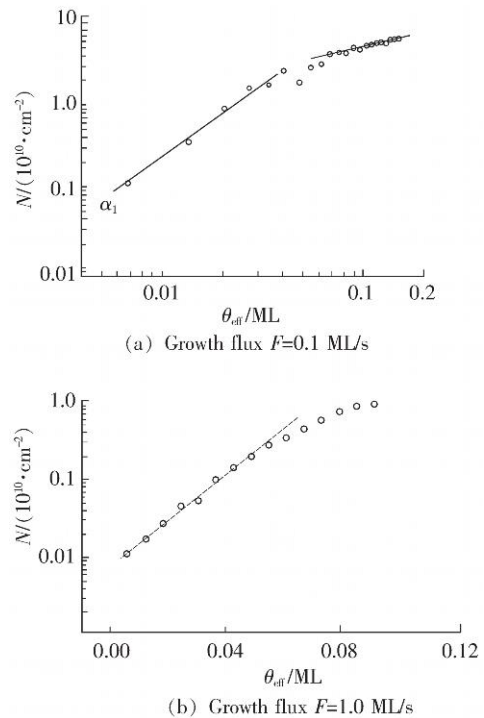


Fig. 1 Areal density of quantum dots  $N$  vs effective InAs coverage  $\theta_{eff}$   
 图1 量子点面积密度  $N$  与 InAs 有效沉积量  $\theta_{eff}$  的关系

coverage increasing above about 0.05 ML, the increasing rate of  $N$  slows down and deviates gradually from the straight line as marked in Fig. 1 (b), and there is a tendency for  $N$  to approach saturation<sup>[23]</sup>.

Fig. 2 (a), (c) and (e) are the AFM (atomic force microscope) snapshots taken at  $\theta_{eff} \sim 0.006$  ML, 0.05 and 0.09 ML, respectively, for the case of 1.0 ML/s and 520. Fig. 2 (b), (d) and (f) show the corresponding height histograms  $N(h)$ , the number of QDs of  $h$  in height. It can be seen from the figures that there are only mature quan-

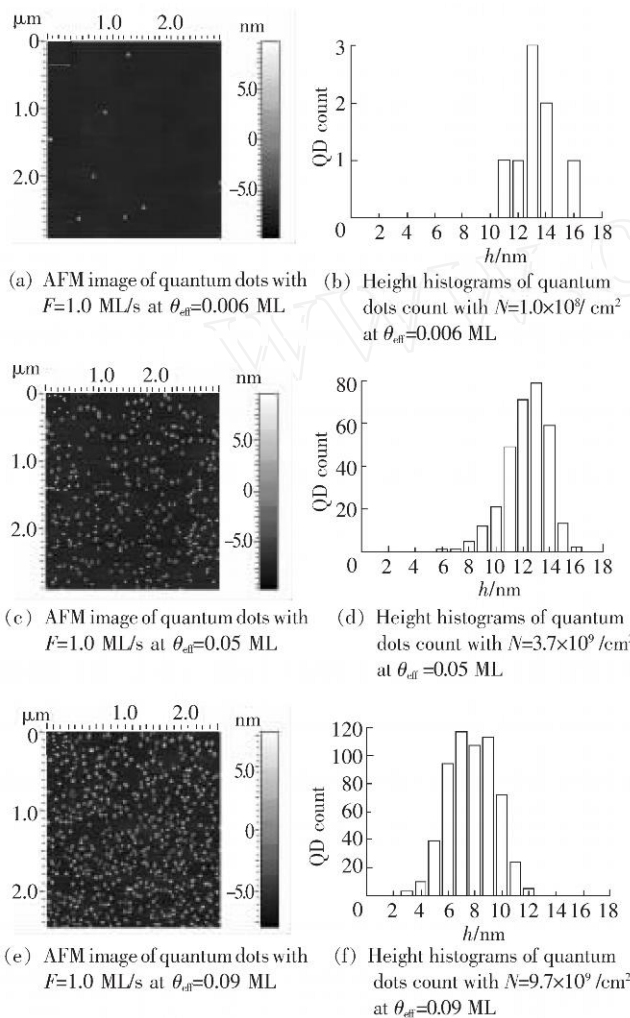


Fig. 2 AFM images of quantum dots in the case of  $F = 1.0$  ML/s at  $\theta_{eff} = 0.006, 0.05$  and  $0.09$  ML and the corresponding height histograms

图2  $\theta_{eff} = 0.006, 0.05$  和  $0.09$  ML 时的量子点 AFM 像和相应的量子点高度分布 ( $F = 1.0$  ML/s)

tum dots with  $h$  above 10 nm even at the very early growth regime with  $\theta_{eff} \sim 0$ . As comparison, Fig. 3 (a), (c) and (e) are the AFM images for the case of the flux rate 0.1 ML/s at  $\theta_{eff} = 0.004, 0.01, 0.06$  ML, respectively; Fig. 3 (b), (d) and (f) are the corresponding height histograms  $N(h)$ . In contrast to the former case of the 1.0 ML/s flux, it can be seen from these figures that there are just very low clusters with the heights below 1 nm at the very beginning. With  $\theta_{eff}$  increasing, the value of  $N(h)$  with  $h$  above

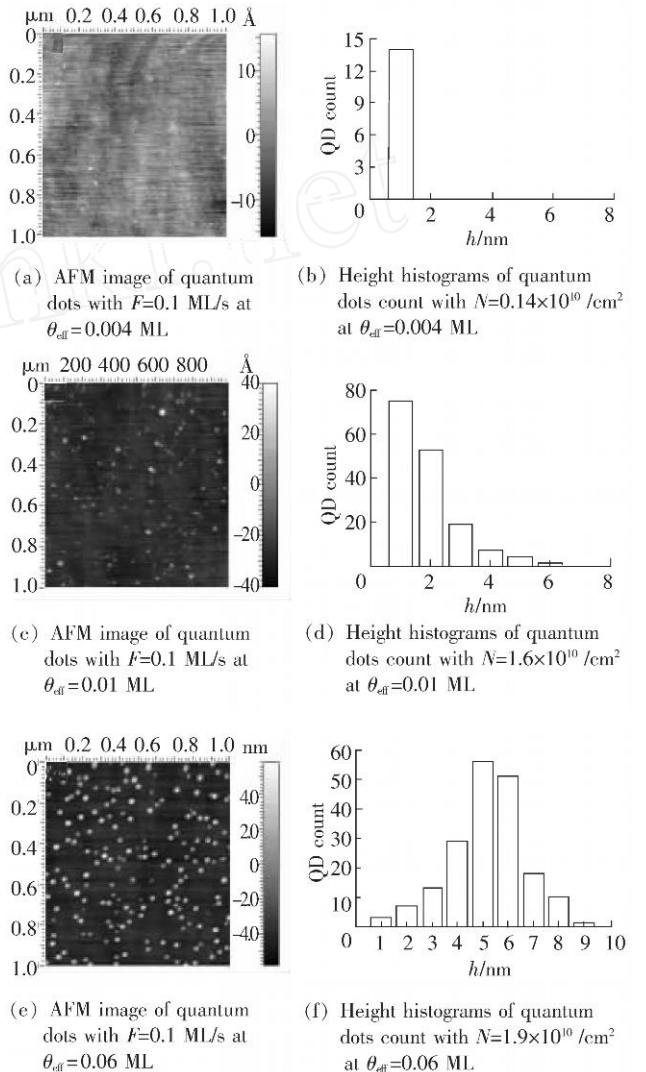


Fig. 3 AFM images of quantum dots in the case of  $F = 0.1$  ML/s at  $\theta_{eff} = 0.004, 0.01$  and  $0.06$  ML, respectively and the corresponding height histograms

图3  $\theta_{eff} = 0.004, 0.01$  和  $0.06$  ML 时的量子点 AFM 像和相应的量子点高度分布 ( $F = 0.1$  ML/s)

1 nm increases progressively, and evolves into a decreasing function of  $h$ , as shown in Fig. 3 (d); with  $\theta_{\text{eff}}$  increasing further,  $N(h)$  becomes a Gaussian, as shown in Fig. 3 (f), with the QD's average height no longer increasing any more.

The two cases of InAs QDs are significantly different from each other in QD's evolution with increasing the coverage. In the case of 0.1 ML/s, the two regimes characterized by the power laws  $N \propto \theta_{\text{eff}}^{1.8}$  and  $N \propto \theta_{\text{eff}}^{0.6}$  are apparently rather similar to the nucleation and aggregation stages of the submonolayer regime in the layer-by-layer growth mode. In the first regime, there are only QD's precursors with height below 1 nm, and they grow progressively up to a given height. In contrast, for the case of 1.0 ML/s, there is no such a progressive QD's growth process with increasing the coverage, and a quantum dot seems to grow into a mature size instantaneously at its birth moment. Another significant difference between the two cases is in the areal density at the very early stage of the QD formation: at 0.1 ML/s, the QD's precursor density is relatively large ( $\sim 10^{10}/\text{cm}^2$ ) in the beginning of the 2D/3D transition; while at 1.0 ML/s, the density of mature quantum dots is rather low ( $N_{\text{begg}} < 10^8/\text{cm}^2$ ) in the beginning. The differences in both QD's density and height at the 2D/3D transition suggest the difference between the two cases in growth behavior: in the former case, the QD nucleation and growth are local events during which the material as required comes from a limited region; while in the latter case, the formation of a quantum dot involves a relatively large area of, at least,  $\sim 1 \mu\text{m}^2$  ( $1/N_{\text{begg}}$ ). It should be noted that the formation of a quantum dot is generally regarded as a local event<sup>[24-25]</sup> confined in a region as determined by the adatom diffusion length, which is generally in the order of 10 nm in magnitude.

The situation of 1.0 ML/s is rather similar to our daily experiences with the formation of a water

droplet on an unwetted surface: if some amount of water is sprayed onto the unwetted surface, a water droplet would spontaneously form as driven by surface tension. The difference between the situation of a water droplet and that of a quantum dot is that the former can be of any macroscopic size, while each QD in the ensemble is fixed in volume to a nanometer scale. The evolution of the QD's ensemble is characterized by the exponential function  $N = N_0 e^{k \theta_{\text{eff}}}$ , which implies that quantum dots form one after another<sup>[22]</sup>, rather than simultaneously nucleate in a nucleation regime, and then simultaneously grow in an aggregation regime.

### 3.2 Time Scale for the Formation of an InAs Quantum Dot

The time scale for the events taking place on a growing surface such as GaAs (001) and Si (001) span a remarkably large range. For example, at a normal growth temperature, a surface atom finishes a vibration cycle in about  $10^{-13}$  seconds; an adatom moves from one lattice site to a neighboring site in about  $10^{-9}$  -  $10^{-7}$  seconds; the atomic attachment/detachment events at the atomic step edges on the Si (001) surface need about  $10^{-4}$  seconds<sup>[26]</sup>. The fast recovery process of the GaAs (001) surface after the growth termination requires several ten of seconds<sup>[27]</sup>; a perturbation on an unstable Si (001) with a characteristic length of 300 nm doubles its amplitude via surface diffusion in 3 hours<sup>[28]</sup>.

How long a period of time does an InAs quantum dot take for its growth from the moment of nucleation to a given full height on the growing GaAs (001) surface? This cannot be measured exactly at the present time. However, an upper limit for such a period of time may be established from our experimental data on the variation of QD density  $N$  with effective coverage, as shown in Fig. 1 (b)<sup>[22-23]</sup>. From the figure, we know that the QD density increases with coverage in accordance with  $N = N_0 e^{k \theta_{\text{eff}}}$ . The coverages  $\theta_{\text{eff}}$  can be translated

into growth time  $t$  via the relation  $x = Ft$ , where  $F$  is the deposition flux. Furthermore, the difference in the density  $N$  is easily discernible between the two AFM images taken at the regions distanced from each other by a few micrometers. Based on these experimental data, in the case of 1.0 ML/s, the period of time for an InAs quantum dot growing into a mature one with a given height of about 10 nm is less than about  $10^{-4}$  -  $10^{-5}$  seconds, comparable to the rate at which the Si atom detach or attach to an atomic step edge<sup>[26]</sup>.

Nowadays, the epitaxial growth phenomena including the self-assembling of QDs are generally understood in terms of the solid-on-solid model and are believed to be facilitated via the adatom diffusion with the diffusion constant  $D = v_0 \exp(-E_a/kT)$ . However, if the formation of a quantum dot were an aggregation or a growth processes determined by the adatom diffusion, it has to be relatively slow and should take a time at least more than one second<sup>[29]</sup>. The tremendously fast formation of an InAs quantum dot in the case of 1.0 ML/s demonstrates that there should be a much more rapid process for the mass transport than the conventional adatom diffusion.

### 3.3 A Novel Explanation for the Formation of Quantum Dots

The surface of the InAs wetting layer on GaAs (001) may be much more complex than a growing surface as usually assumed in which there are only diffusing adatoms that implements mass transport. J. M. Garcia et al<sup>[30]</sup> reported that only 50% of indium were incorporated into the crystal during the MBE deposition of a few monolayers of InAs on GaAs (001) at 470 °C due to the segregation effects, which is significantly enhanced by the mismatch strain; the remaining amount of indium was floating on the growing surface and was progressively incorporated into the crystal structure during the subsequent growth of the GaAs capping layer. In addition, based on the thermodynamic

calculation, D. J. Bottomley<sup>[31-32]</sup> proposed that the melting point of InAs on GaAs (001) could be significantly reduced by the mismatch strain, and during MBE growth, there was a molten InAs monolayer upon which the quantum dots were formed. Based on these experimental observations and theoretical calculations, there should be a large amount of In atoms or InAs molecules floating on the growing surface. In order to explain the significantly rapid formation of an InAs quantum dot, as we observed, we assume that these floating atoms or molecules have very little interaction with the wetting layer and are almost freely moving on the surface. This is a rather different state from that of an adatom on the surface. Under the action of misfit strain and surface tension, these atoms or molecules move very fast in a certain direction to accomplish the formation of a quantum dot.

Because the mass transport via the freely moving particles is much faster than the incidence flux  $F$ , the growth parameters can be taken as thermodynamic parameters. As a first-approximation, the formation of InAs QDs may be regarded as a phase transition accomplished under the condition of a local thermodynamic equilibrium. It is well known that the phase transition of a pure substance, such as water, can be a continuous or discontinuous in nature, depending on the temperature and pressure. Under the growth conditions of 0.1 ML/s and 500 °C, the QD's growth is a continuous and progressive process at first, and the formation of quantum dots may be regarded as a continuous phase transition or a second-order one. As a continuous transformation, the 2D/3D transition starts from a critical point at which the nuclei or QD's precursors are formed via statistical fluctuations in the density of floating atoms or molecules. The order parameter for the continuous transformation may be the volume of QDs per unit area or the extent of the strain relaxation in the InAs strain film via the formation of QDs. Under the



growth condition of 1.0 ML/s and 520 °C, the thermodynamic condition is equivalent to a point on the coexistence curve in the phase diagram, which significantly deviates from the critical point. The formation of QDs is a discontinuous phase transition from the phase of floating particles to a phase of an ensemble of mature quantum dots; once enough, all the InAs material on the growing surface available is instantaneously assembled into quantum dots.

## 4 Conclusion

In nature, there are many phenomena that cannot be directly solved by using a simple physical model or a set of equations. The formation of a quantum dot, involving a large number of atoms ( $> 10^4$ ), is a process that can be accomplished in less than  $10^{-4}$  seconds, as demonstrated in this article. The process may be not a conventional growth or an aggregation process that can be implemented by a series of individual atomic events. The process may involve some complex collective atomic motion. Therefore, the formation of a quantum dot should be understandable in terms of a more macroscopic way than atomic detail, as discussed in this article. The article is a step made by the authors in this way.

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#### Biography:

Wu Ju graduated from university in 1982, and received his PH. D degree in 1989. He had been working on the growth of semi-insulating GaAs single crystal and on the identification of defects and structures of epitaxial growth semiconductors since he graduated from university and worked for his doctoral thesis. He is currently an associate researcher in the lab working on the physical mechanism for the self-assembling of quantum dots in the InAs/GaAs (001), and he would like to demonstrate that the spontaneous formation of these quantum dots should be a result of an event simultaneously involving a large number of atoms, instead reducible to individual atomic motions, such as atomic diffusion and attachment, one-by-one.

## 下期部分目录预告

原位 Si 掺杂 c-BN 薄膜的研究

纳米线在新型太阳能电池中的应用研究

900 nm 三叠层隧道级联半导体激光器的结构优化

硅 MEMS 工艺及展望

立方相碳化硅 MEMS 器件研究进展

体硅热膜传感器单片集成工艺的研究

紫外固化真空负压纳米压印系统的研制

Ku 波段 GaAs 单片功率放大器研究

3D 封装及其最新研究进展

单片集成锁模量子点激光器