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Monte Carlo simulation of epitaxial growth

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A Monte Carlo–based method for simulation of epitaxial crystal growth is reported. The improvement of our method, in comparison with simulations based on the solid-on-solid model, is the possibility of simulating crystal growth without the assumption of a discrete crystal-lattice structure. Our model requires only the specification of a Hamiltonian. The method is used for studying growth in a two-dimensional cross section through the substrate and the epilayer up to a width of 80 atoms. Two different potentials have been chosen for the simulations: (a) the Lennard-Jones potential and (b) a directional Lennard-Jones potential to describe covalent systems. The epitaxial growth of strained layers was simulated to study the incorporation of dislocations at the interface, and island and layer growth in the nucleation stage were investigated.

I. INTRODUCTION

Advances in crystal-growth technologies, in particular in the field of epitaxial growth from the vapor phase,[1] have stimulated the search for realistic theoretical models of crystal growth. With the aid of modern computer technology, it has become possible to perform *ab initio* calculations with realistic interaction potentials.

In a number of computer simulations, the growth of lattice-matched crystals is described by an Ising model.[2–5] These models are based on a predefined lattice structure, and have been used successfully to study the surface roughening with increasing temperature,[3] the transition between island and layer growth,[4] and the propagation of spiral dislocations.[5] Recently, more sophisticated simulations have been published which allow the simulation of lattice-mismatched structures.[6–8]

However, calculations using a continuous space for the simulation are quite rare.[9–12] Due to the complexity of continuous-space models, their application has to be restricted to very small three-dimensional crystals or two-dimensional cross sections of crystals. Continuous-space models using molecular-dynamics (MD) and Monte Carlo (MC) methods provide some advantages in comparison to discrete-space models, because the assumption of the Hamiltonian is sufficient to describe the physical behavior of the system.

The most often used potential for the simulation of crystal growth is of Lennard-Jones (LJ) type, which has been used for MC (Ref. 9) and MD (Ref. 10) simulations. The LJ potential has been selected for our calculations, because it saves a lot of CPU time, due to its short-range nature. We used a directional LJ (DLJ) potential in two dimensions, where the particles are characterized by their locations and the orientation of the bonds.[11]

At present, there is no general rule for the choice of the simulation model. With the model presented in this paper it is, in principle, possible to tackle the problem of lattice mismatch, substrate orientation, substrate temperature dependence, surface morphology, incorporation of dislocations, and the transition from island to layer growth for heteroepitaxy, including the growth of binary and ternary compounds on binary substrates.

II. DESCRIPTION OF THE MODEL

We use a continuous-space MC method to simulate epitaxial crystal growth in a two-dimensional cross section through the substrate and the epilayer parallel to a (001) plane of a zinc-blende structure. In this cross section the four tetrahedral bonds of each atom are oriented in the crystal lattice as shown in Fig. 1(a). It is evident that the projection of the tetragonal zinc-blende lattice into the plane gives a fourfold symmetry. Therefore we set up a DLJ potential reflecting the fourfold symmetry. This restriction to two dimensions was applied to the simulation purely to save CPU time.

In our model each particle is represented by two two-dimensional vectors in the plane of the cross section, namely its location $\mathbf{r}$ and its orientation $\mathbf{e}$ with $|\mathbf{e}|=1$. The pairwise interaction potential is characterized by three parameters: the binding energy E_0, the binding length r_{equ}, and the parameter β which describes the covalent character of the bond. The potential energy of two interacting particles is given by

$$E_{12}(r,\varphi_1,\varphi_2)=\begin{cases} E_0\dfrac{r^{-12}-(1-\beta)\alpha r^{-6}-\beta f(\varphi_1)f(\varphi_2)\alpha r^{-6}}{\alpha^2}\,, & r<r_{\text{cutoff}}\\ 0, \quad r\ge r_{\text{cutoff}}\,. \end{cases}$$

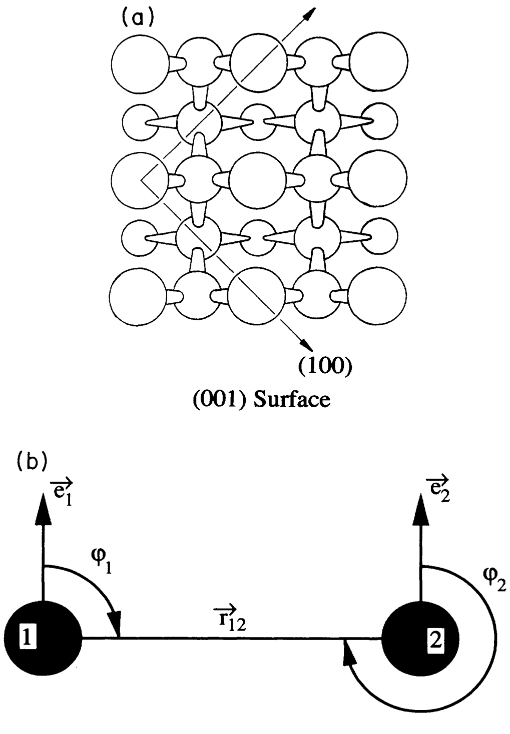

FIG. 1. (a) (001) surface of a zinc-blende structure with the usual crystallographic directions and the corresponding bonds. (b) Schematic drawing of two particles at a distance of r_{12}, with their orientations $\mathbf{e}_1, \mathbf{e}_2$ and the definition of the angles φ_1, φ_2.

The pairwise interaction energy is a function of the variables r_{12}, φ_1, and φ_2 which are shown in Fig. 1(b). The distance r_{12}, the angular dependence f, the parameter α, and the cutoff radius r_{cutoff} are taken as

$$\mathbf{r} \equiv \mathbf{r}_1 - \mathbf{r}_2 , \quad r \equiv |\mathbf{r}| ,$$

$$\cos(\varphi_n) \equiv \frac{\mathbf{r}\cdot\mathbf{e}_n}{r} , \quad f(\varphi_n) \equiv \cos^2(2\varphi_n) , \quad \text{with } n \in \{1,2\} ,$$

$$\alpha \equiv 2 r_{\text{equ}}{}^{1/6} , \quad r_{\text{cutoff}} \equiv 2.5 \, r_{\text{equ}} .$$

A graphical representation of the potential energy for $\beta=0.5$ is given in Fig. 2. The interaction potential can be tuned by the parameter β from the pure Lennard-Jones potential with $\beta=0$ to the pure directional Lennard-Jones potential with $\beta=1$. Since different species of atoms are allowed, E_0, r_{equ}, and β are specific for the atom species involved in the interaction. Therefore, we denote these parameters by $E_{0,ij}$, $r_{\text{equ},ij}$, and β_{ij} to describe an interaction of an atom of kind i with an atom of kind j. To simulate the growth of compound epilayers on compound substrates our model allows one to select $E_{0,ij}$, $r_{\text{equ},ij}$, and β_{ij} for n different kinds of atoms. That means, in principle, $3n(n+1)/2$ parameters describing the interaction of atoms can be set at will.

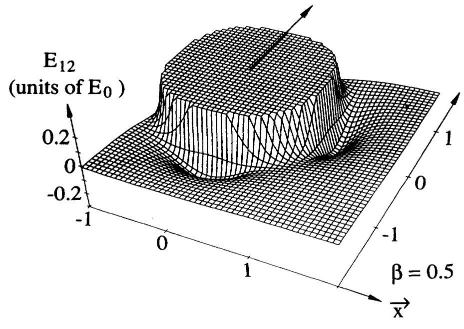

FIG. 2. Pairwise binding energy $E_{12}(r,\varphi,0)$ in units of E_0 for $\beta=0.5$.

To save CPU time we set

$$r_{\text{equ},ij} = (r_{\text{equ},ii} + r_{\text{equ},jj})/2$$

and $\beta_{ij}=\beta_{ii}=\beta_{jj}=\beta$. The symbols ○ and × denote the two kinds of atom according to the representation of atoms in the lattice pictures. By definition, ○ indicates a substrate atom while × indicates an atom of the epilayer.

To simulate the growth of a crystal we use periodic-boundary conditions in the horizontal direction. We start by placing some layers of substrate atoms in the distance of $r_{\text{equ},\text{○○}}$. The substrate is equilibrated by means of a conventional importance sampling MC method;[6] this means that one particle is selected at random and moved to a position within a rectangle with a horizontal length of $2.5 \, r_{\text{equ},\text{○○}}$ and a height of $r_{\text{equ},\text{○○}}$. The orientation $\mathbf{e}$ of the particle is chosen at random also. This move and rotation is accepted with the probability $p=\exp(-\Delta H/k_B T)$. This procedure, commonly called the "MC step," is repeated until an acceptable approach to the equilibrium is achieved. After the substrate is equilibrated, another particle is placed near the surface at a random position and the system is equilibrated again. After some particles are deposited, expectation values of the position of each particle are calculated. The difference between the actual and the average position is used as an indication of the stability of the lattice. This procedure is repeated until the required number of particles is deposited.

We present the results in three different ways. Since we are working with a two-dimensional cross section, it is possible to show a picture of the whole lattice.

Since we are interested in the vertical distribution of the particles, we plot $N_{<h}(h)$, defined as the number of particles located below the height h, as a function of h. One can calculate $N_{<h}(h)$ from $n(h)$, the number of particles at the height h.

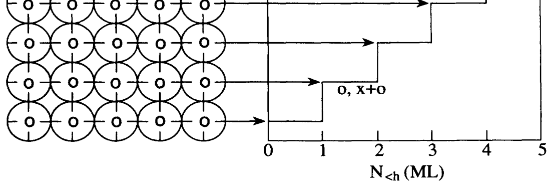

FIG. 3. Correlation between lattice picture and h-versus-$N_{<h}$ plot.

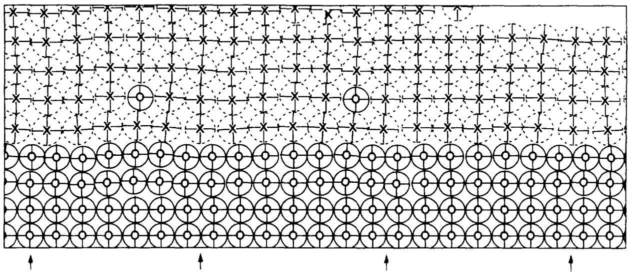

FIG. 4. Typical part of the simulated lattice of an epilayer with a misfit of 14% simulating the situation present in CdTe on GaAs. The other parameters are $\beta=0.5$, $E_{0,\times\times}=E_{0,\times\circ}=E_{0,\times\times}$, and $k_BT=0.04E_{0,\times\times}$. The resulting dislocations are marked by arrows.

$$N_{<h}(h)=\int_0^h n(h')dh' .$$

To emphasize the direct correlation to the lattice pictures, $N_{<h}$ is drawn on the horizontal axis of Fig. 3, while h is drawn on the vertical axis as defined by the growth direction. Three curves are given in Fig. 3, one for the substrate particles only (labeled $\circ$), one for the epilayer atoms (labeled $\times$), and one for the whole system (labeled $\times+\circ$).

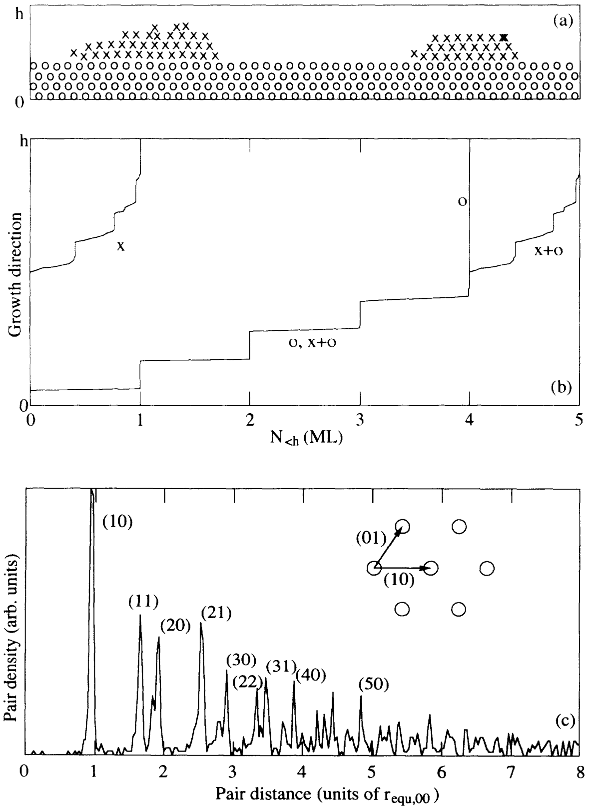

FIG. 5. Result of a simulation to demonstrate island growth by selecting a weak bond between epilayer and substrate. The parameters are $\beta=0$, $r_{\mathrm{equ},\times\times}=0{,}95\ r_{\mathrm{equ},\circ\circ}$; $E_{0,\times\circ}=0.5\ E_{0,\times\times}$; $E_{0,\circ\circ}=E_{0,\times\times}$: (a) typical part of the lattice, (b) h-versus-$N_{<h}$ plot, and (c) pair density vs distance in units of $r_{\mathrm{equ},\circ\circ}$.

To demonstrate the lattice quality we plot the pair density in arbitrary units as a function of the particle distance r. The resulting figure shows a series of peaks, each corresponding to a translation vector in the two-dimensional lattice. The horizontal axis is always scaled in lattice constants of the substrate ($r_{\mathrm{equ},\circ\circ}$), which gives us the possibility of identifying the lattice misfit and the long-range order in the lattice.

By setting the parameter β between 0 and 1, we could choose between LJ and DLJ potential. Whenever we set $\beta=0$ we obtained a hexagonal lattice structure, while for $\beta=0.5$ the results showed a quadratic lattice.

III. RESULTS AND DISCUSSION

To test our model we simulated the growth of a structure with a mismatch of 14% using DLJ with $\beta=0.5$ and $r_{\mathrm{equ},\times\times}=1{,}14\ r_{\mathrm{equ},\circ\circ}$. The binding energies were chosen $E_{0,\times\times}=E_{0,\times\circ}=E_{0,\times\times}$ and $T=0.04E_{0,\times\times}k_B^{-1}$. The result is depicted in Fig. 4, which shows a typical section of the lattice but the simulation was calculated for a substrate width of 70 atoms. The resulting dislocations are marked by arrows. It is evident that the mean distance between two dislocations is seven substrate atoms. It is thus in excellent agreement with the TEM observations of CdTe/GaAs heterostructures.[13]

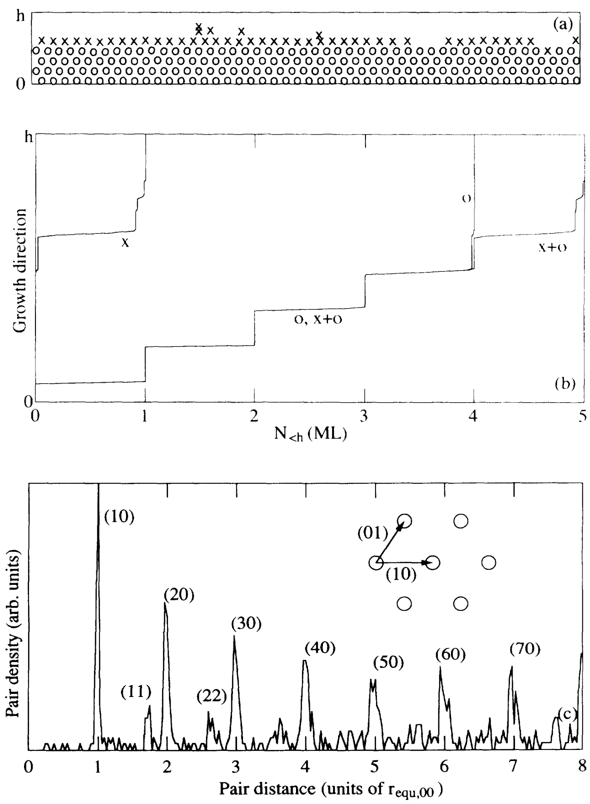

FIG. 6. Result of a simulation to demonstrate layer growth by selecting a strong bond between epilayer and substrate. The parameters are $\beta=0$, $r_{\mathrm{equ},\times\times}=r_{\mathrm{equ},\circ\circ}$; $E_{0,\times\circ}=2\ E_{0,\times\times}$; $E_{0,\circ\circ}=E_{0,\times\times}$: (a) typical part of the lattice, (b) h-versus-$N_{<h}$ plot, and (c) pair density vs distance in units of $r_{\mathrm{equ},\circ\circ}$.

In the next set of simulations we investigated the transition from island to layer growth. The substrate width was kept constant during all simulations with 80 atoms and the temperature was set to $T=0.04\,E_{0,\times\times}k_B^{-1}$. The ratio of the binding energy between substrate and epilayer atoms $E_{0,\times\circ}$ to the binding energy between epilayer atoms $E_{0,\times\times}$ was varied in seven steps from 0.5:1 to 2:1. For all the simulated systems we found a strong influence of the ratio of the binding energies on the nucleation stage. Out of 48 simulations we show two typical results. For a weak binding between substrate and epilayer $E_{0,\times\circ}=0.5\,E_{0,\times\times}$, $E_{0,\circ\circ}=E_{0,\times\times}$ and a lattice-mismatched (5%) system, we obtained the results shown in Fig. 5. For a strong bond between substrate and epilayer ($E_{0,\times\circ}=2.0\,E_{0,\times\times}$, $E_{0,\circ\circ}=E_{0,\times\times}$) and exact lattice match, the result is shown in Fig. 6. The lattice pictures Figs. 5(a) and 6(a) show clearly either island or layer growth. The h-versus-$N_{<h}$ plot in Fig. 5(b) reflects the island growth in the steps of the curve labeled ×, which shows that more than one lattice plane is needed to accommodate the number of atoms for a full monolayer. On the other hand, the h-versus-$N_{<h}$ plot in Fig. 6(b) labeled × shows that almost the full monolayer coverage is reached within one lattice plane.

Figures 5(c) and 6(c) show the pair density for island and layer growth. The insets define the unit cells with the base vectors for the identification of the translation vectors. Within an island one can find pairs for almost all translation vectors while in a single layer there exist only pairs for translation vectors parallel to the interface. Therefore, we find peaks for the translation vectors (11), (21), (22), and (31) in Fig. 5(c), whereas in Fig. 6(c) the (50), (60), and (70) peaks are clearly resolved. The lattice mismatch of 5% is evident by a shift of all peaks in Fig. 5(c) in comparison to the scale, which is given in lattice constants of the substrate.

In this paper we have demonstrated the use of a continuous-space Monte Carlo method for studies of island and layer growth by using different binding energies for the deposited atoms to the substrate. The restriction to a two-dimensional cross section allows only a simplified picture of the real growth mechanisms. However, we could show that our results describe the realistic situation as shown, for example the incorporation of dislocations at the CdTe/GaAs heterointerface.

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