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# Epitaxy and superlattice growth

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We have performed a rigid-lattice, atomistic, model calculation for bcc-fcc epitaxy. We have calculated the energy of an fcc (111) plane in contact with a bcc (110) substrate as a function of relative position, orientation, vertical displacement, and ratio of fcc/bcc lattice parameters. The results show two orientations for which deep minima exist in the dependence of energy on the fcc/bcc lattice parameter ratio. Our results are in qualitative agreement with earlier Moiré pattern studies. The calculations were made with two different atom-atom potentials; the results are found to be independent of the long-range attractive part of the potential. This implies that for rigid lattices the energetics is only determined by the relative atomic sizes of the substrate and overlayer. A comparison with experimental results indicates that the growth of metallic superlattices is controlled not only by epitaxial energetics but also by the stacking of the various constituents.

The study of epitaxial crystal growth is of fundamental as well as of technological interest. Extensive experimental work has been done to study the growth of noble gases and organic materials on graphite,[1] the growth of semiconductors and metals[2] on a variety of substrates, and the growth of semiconducting[3] and metallic[4] superlattices. The theoretical work has been somewhat limited to the study of epitaxy using Moiré patterns,[5] and phenomenological energetics calculations[6] and studies relating to the effect of substrate-induced strains on the orientation of monolayer adsorbates.[7]

We present here a rigid-lattice atomistic model for bcc (110)–fcc (111) epitaxy. We have studied the energy of fcc (111) plane on a bcc (110) substrate as a function of position, orientation, vertical displacement and ratio of fcc/bcc lattice parameters. The model shows the existence of two well defined angles for which minima exist in the energy versus fcc/bcc lattice parameters ratio. These results are independent of the range of the attractive part of the atom-atom potential and, therefore, imply that for rigid lattices the energetics is only determined by the relative atomic sizes of the substrate and overlayer.

Comparison with experimental fcc-bcc epitaxy and fcc-bcc superlattice growth is also presented. This comparison indicates that size effects are very important for orientational epitaxy and that they could explain a large body of experimental data. The comparison with experimental results on fcc-bcc superlattice growth indicates that the type of study presented here explains, perhaps partially, superlattice growth although a full theory would require introducing the effect of lattice vibrations. In addition, it is found that superlattice growth is strongly affected by the relative stacking of planes.

In our model calculation, the substrate consists of three bcc (110) atomic planes properly stacked with respect to each other as part of a proper bcc structure and one fcc (111) plane. The bcc (110) planes are infinite and the fcc (111) plane is limited to a disk of finite radius. Since both lattices are taken to be rigid, the only interaction potential needed is that between an fcc atom and a bcc atom. Two potentials have been used: (a) a short-ranged, exponentially decaying potential

$$V(r)=4\exp[-\tfrac{8}{3}(r^2-1)]\{\exp[-\tfrac{8}{3}(r^2-1)]-1\} \quad (1)$$

and (b) a standard Lennard-Jones ($L$-$J$) potential

$$V(r)=4\left(\frac{1}{r^{12}}-\frac{1}{r^6}\right) . \quad (2)$$

To a very high accuracy, the repulsive part and the depth of both potentials are the same; they only differ in the long-range part as shown in Fig. 1. The manner of writing Eqs. (1) and (2) shows that our unit of length is the sum of the radii of the two types of atoms and the depth of the potential is the unit of energy.

The total energy, per particle of the fcc disk, was calculated and minimized with respect to a displacement in the $x,y$ plane which is taken to be the plane of the two rigid structures, the interplanar distance for fixed rotation angle $\theta$ and ratio $P$ of fcc-bcc lattice parameters. Calculations were per-

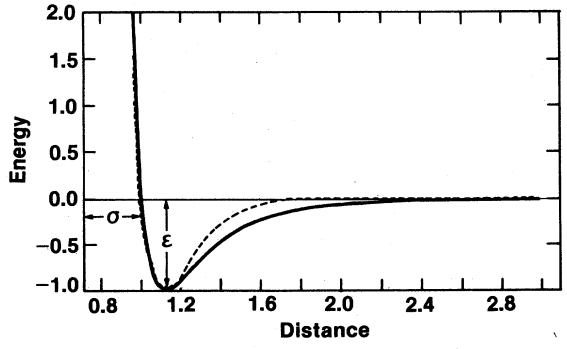


FIG. 1. Lennard-Jones (solid line) and exponential (dashed line) potential used in the calculation. The length scales in the calculation are determined by the hard-core radius $\sigma$ and energies by the depth of the potential $\epsilon$.

 

formed to ensure that (1) the size of the fcc plane is large enough to avoid edge effects, (2) that symmetric situations ($\theta$, $60°-\theta$, $60°+\theta$, etc.) give identical results, (3) that cutoff errors in the potential do not influence the final results, and (4) that the minimization procedure does not affect the results.

We found that the relative position of the fcc and bcc lattices in the plane does not affect the final energy and that the distance between the fcc and bcc lattices always converged to a value between 0.94 and 0.97. Therefore, the only two important variables are (1) ratio of the lattice parameters and (2) the orientation of one lattice with respect to the other.

Figure 2 shows constant energy contours for the exponential potential in the $P$-$\theta$ plane ($P$=fcc/bcc lattice parameter). These results show that there is a deep energy minimum ($E_{\min}\sim -3.8$) at $\theta\simeq 5°$ and $P=1.33$ and a shallower minimum ($E_{\min}=-3.5$) at $\theta\simeq 0°$ and $P=1.16$. This is shown much more clearly (Fig. 3) in a plot of the energy versus $P$ at constant $\theta$ for the $L$-$J$ potential. Minima exist only on the vicinity of $\theta=0$ and $\theta=5°$; at any other angle the energy is essentially independent of $P$. It is quite remarkable that the results obtained using the $L$-$J$ potential are hardly different from the results found for the exponential potential. However, the relative depths at $\theta\cong 0°$ and $\theta\cong 5$ shown in Fig. 3 are smaller than in the case of the exponential potential by about 4%. This is the only effect which results from the $-1/r^6$ attraction in the $L$-$J$ potential compared with the exponential attractive part shown in Fig. 1.

The theoretical study of fcc-bcc epitaxy has received limited theoretical attention. Bruce and Jaeger[5] studied the epitaxy of fcc on bcc and bcc on fcc crystals with the help of Moiré diagrams. In this method, the crystal planes in question, bcc (110) and fcc (111) in our case, are drawn to scale on transparencies and the relative orientations are derived from the dependence of the Moiré fringe spacing on the orientation angle. It is claimed that those orientations are preferred for which the Moiré fringe spacings is large. This is because there is less unfavorable overlap of atoms in this situation. A further modification in this model implies that

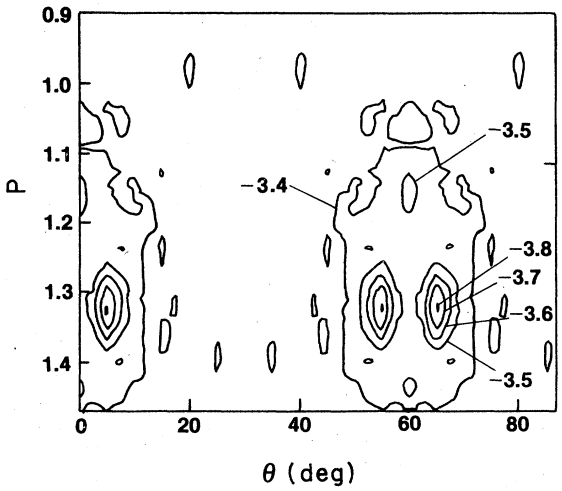


FIG. 2. Constant energy per particle, contour plots in the $P$-$\theta$ plane for the exponential potential. $P$=fcc/bcc lattice parameter. $\theta$ is the angle between fcc/bcc.

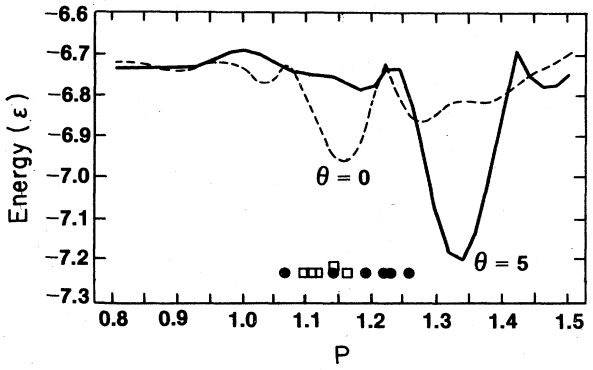


FIG. 3. Energy vs ratio of fcc/bcc lattice parameters for the Lennard-Jones potential. $P$=fcc/bcc lattice parameter. The symbols close to the $x$ axis indicate the systems that have been attempted to grow in superlattices. Closed symbols indicate combinations that do not and open symbols combinations that do exhibit superlattice growth in the sense described in the text. Systems from left to right ●: Nb/Ni, □: Nb/Cu, □: W/Ni, □: Mo/Ni, ●: W/Cu, □: Mo/Cu, □: V/Ni, ●: V/Cu, ●: Cr/Ni, ●: Cr/Cu, and ●: Fe/Cu.

the orientation adopted is that for which unfavorable areas are more "elongated."

Later, van der Merwe[6] used a rigid model to calculate the interfacial potential energy. In this model, a potential is used which has the symmetry of the substrate and where the parameters are adjusted to conform to the symmetry of the surface layer. Our model calculation confirms some of the assumptions of these theories, namely, that the position *in* the plane and the distance *between* the fcc and bcc planes will not affect the qualitative conclusions.

The agreement between the three treatments is quite remarkable. With quite high accuracy, all these models predict that there is a deep minimum at $P=1.33$ for $\theta=5.3$ (Kurdjumov-Sachs, KS, orientation) and two minima at $\theta=0$ for $P\simeq 1.13$ and 1.42 (Nishiyama-Wasserman, NW, orientation).

The value of these types of calculation relies on the possibility of studying the relative importance of different terms which determine the epitaxy energy. Such calculations can also help in guiding further experimental studies. The theoretical results presented earlier can be compared to experimental studies of thin ($\sim$1–2 monolayers) single epitaxial films, thick single epitaxial layers, and superlattices. A quite comprehensive comparison with thin and thick epitaxial films has been performed by Bruce and Jaeger[5] and by van der Merwe.[6]

Up to now it has not been possible to make detailed quantitative comparisons between theory and experiment in the field of the growth of superlattices. In fact, systematics of the growth of superlattices is quite limited and further work is necessary to check the importance of the various growth parameters.

We have attempted to grow in a *crystalline* superlattice, twelve different fcc/bcc combinations. The samples were grown using a sputtering technique described elsewhere.[8] Briefly, the substrates [90° and 0° sapphire, mica, NaCl, MgO, glass, Ge (111), Ge (100), Si (111), Si (100), KCl, and $BaF_2$] were held on a rotating, heated (20–350 °C), temperature controlled ($\pm 2$ °C) platform. The materials were sputtered from two high-rate magnetron sputtering guns which were shielded from each other so that no over-