# Misfit accommodation in heteroepitaxy by inclined stacking faults

M. Henzler, C. Homann, U. Malaske, and J. Wollschläger

*Institut für Festkörperphysik, Universität Hannover, Appelstrasse 2, D-30167 Hannover, Germany*

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For accommodation of the misfit between growing film and substrate an interesting mechanism is proposed. A periodic arrangement of inclined stacking faults may in certain cases provide an average lattice constant with much smaller misfit. It is shown that two experiments are well described with this mechanism. New experiments with KCl films on NaCl(100) [and old ones with Xe films on Si(111)7×7] require stacking faults in the film to describe the structure of the film as observed by spot profile analysis of low-energy electron diffraction.

## I. INTRODUCTION

Frequently, heteroepitaxial growth is perfectly accomplished when the growing film has the same structure and lattice constant as the substrate. The success of heteroepitaxial devices with III-V compounds depends heavily on the defect-free structure of the films, so that no gap states are introduced by dislocations or other defects. It has been shown, however, in many examples also for metals and insulators, that a small misfit or even a large misfit between the lattice constants of film and substrate may not hinder epitaxial growth. Usually epitaxy is described as perfect films with defects like dislocations (if present) at the interface. The accommodation of the misfit, however, may occur also in quite different ways including defects not only at the interface, but also periodically in the film. Depending on the corrugation of the substrate and on the type of bonding, the film may keep its bulk lattice constant and adjust the orientation perfectly to the substrate (which has been called "floating mode"[1]). For a strong corrugation of the substrate the film may adapt the substrate lattice structure and constant ("pseudomorphic mode").[2] For increasing film thickness the film needs more and more elastic strain energy, so that relaxation via the formation of interface dislocations decreases the total energy.[2] In fcc crystals the stacking fault in a (111) direction requires only a small amount of energy. It is the purpose of this paper to discuss the possibility of a special structure of a thin film with stacking faults that may have a lower energy due to avoidance of elastic strain and misfit dislocations than a film with strict bulk periodicity including strain and/or interface dislocations. Experiments will be discussed that are best explained with the existence of a periodic array of inclined stacking faults.

## II. MODEL CONSIDERATIONS

For a small corrugation of the surface and a nondirectional bonding the first atom deposited onto a perfect lattice [like fcc (111) or fcc (100)] should be found in a highly coordinated hollow site. For the second atom an attractive interaction with the first one is to be considered. If the lattice constant $a$ for the growing film is slightly larger than the lattice constant $b$ of the substrate [misfit $m = (a-b)/b > 0$], then the neighboring site may not be occupied in the central position. A monolayer may form a rather perfect film with its

own lattice constant $a$. If the misfit $m$ is approximated by $m \approx 1/M$ with $M$ integer, then after a distance of $M \times a \approx a \times b/(a-b)$ again a perfect site for the film atom is possible. For all atoms in between, an increasingly large deviation from the optimum site is necessary. So, only for a small corrugation, a strong binding to the substrate and a strong binding within the film this floating mode may be observed. This film, however, has to show some modulation of height close to a sinusoidal shape with a lateral periodicity of the moiré pattern $a \times b/(a-b)$, which is detectable with scanning tunneling microscopy (STM) directly and with low-energy electron diffraction (LEED) via the satellite spots close to each diffraction spot due to the new periodicity (Fig. 1). For the present case of a misfit of 11.7% the moiré distance is about nine rows of KCl on ten rows of NaCl.

For the second and further monolayers a nearly perfect binding is possible. Only the approximately sinusoidal bending of the film requires more and more elastic energy. Therefore another arrangement may be more favorable. Stacking faults within the film (with an inclination to the substrate) of course cause some energy, which, however, is not very large for the (111) glide planes of the fcc structure.

On the other hand, the fit to the substrate may be improved considerably by introducing stacking faults inclined to the substrate. As an example, in Fig. 2 a cross section of a film with [111] orientation in a $(0\bar{1}1)$ plane with a stacking fault is shown. If a vector across the stacking fault is added to the vector $[\bar{2}11]$, a new periodicity may be produced within the (111) plane, since this vector 1/6 of $[\bar{4}11]$ is described by a vertical shift of 1/9 of $[111]$ (or 1/3 of the layer distance) and a lateral shift of 5/18 of $[\bar{2}11]$. If these stacking faults are repeated periodically, a small inclination of the
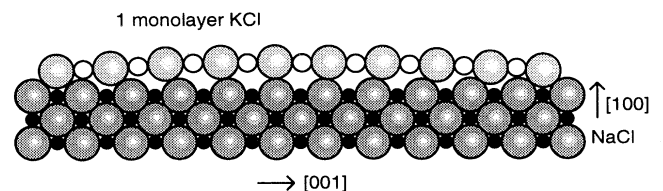
1 monolayer KCl



FIG. 1. Model of a cross section through a NaCl (100) film along a (010) plane with 1 ML of KCl in the "floating mode." Chlorine, sodium, and potassium ions are marked in gray, black, and white, respectively.
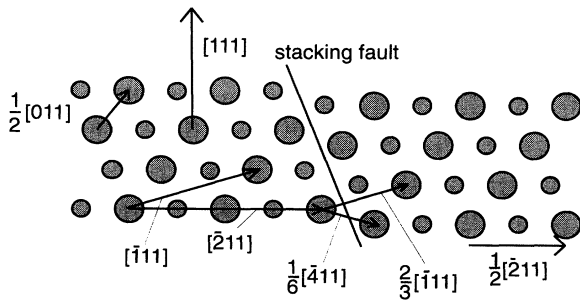
FIG. 2. Model of a cross section through an fcc(111) crystal with a stacking fault in a $[1\bar{1}1]$ direction. Big dots show atoms in the front layer; smaller dots show those in the second layer.

(111) plane of the film towards the substrate is produced.

Such an inclination is avoided when a second set of stacking faults with opposite inclination is introduced. This case will be demonstrated on a fcc(100) plane (Fig. 3). Since here a stacking fault along (111) is compensated by a stacking fault along $(\bar{1}11)$, the base layer of the film is strictly in the (100) plane, since the vector 5/6 $[011]$ of Fig. 3 is given by 2/3 $[111]$ + 2/3 $[\bar{1}11]-1/2[011]$. Nevertheless, the lateral shift across the stacking faults of 2/3 of the row distance may reduce the misfit considerably. This will be demonstrated by the following experimental results.

## III. EXPERIMENTAL RESULTS

Monolayers of KCl have been deposited onto epitaxial films of NaCl(100) at about 150 K (Ref. 3) by thermal evaporation of KCl and annealing up to 400 K. The lattice constant of KCl (0.629 nm) is considerably larger than that of NaCl (0.563 nm). For a coverage of roughly 1 ML the LEED pattern of the film as recorded with a high-resolution LEED system[4] shows the KCl lattice constant. Additionally, all spots have satellites in the distance of the misfit [11.7% misfit produces satellites with a distance of 10% of the surface Brillouin zone (BZ)]. The arrangement of Fig. 1 is therefore the structure of the film. The intensity of the satellites relative to the intensity of the center peak of the spot increases with electron energy. This dependence may be used to derive the modulation of the monolayer.[5] Here the peak to peak height of the modulation is $h=0.028\pm0.003$ nm. Also, the submonolayer regime showed this structure. The quanti-
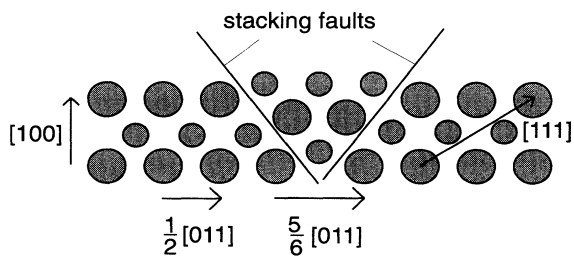


FIG. 3. Model of a cross section through an fcc(100) crystal with two stacking faults along [111] and [$1\bar{1}\,\bar{1}$]. Big dots show atoms in the front layer; smaller dots show those in the second layer.
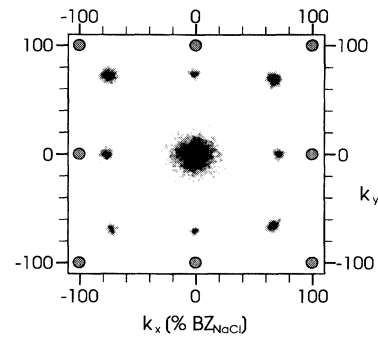


FIG. 4. LEED pattern of a 2.2-ML KCl film on NaCl(100) with the (00) beam in the center and the first-order spots of the film. The positions of the first-order spots of NaCl as seen for the bare film are shown schematically at the 100% positions as gray dots. Electron energy is 70 eV, corresponding to a phase of 4.3.

tative analysis of the submonolayer data, however, is difficult due to the different form factor of NaCl.

For a film thickness from 2 to about 3 ML a drastic change of the LEED pattern was observed: the main first-order diffraction beam was neither at the NaCl position (here taken as the position of 100% BZ), nor at the KCl position (90% BZ), it was found at about 75% BZ (Fig. 4). Since no modification of KCl or any mixture with NaCl with a lattice constant of 4/3 of NaCl is known, a stacking fault structure is proposed (Fig. 5). If after every three atomic rows in the [011] direction a double stacking fault as shown in Fig. 3 is incorporated, a KCl film with ideal distances in all nearest-neighbor distances (including neighbors across the stacking fault) with a periodicity of 1.632 nm is obtained, which is only 2.5% larger than four distances of the NaCl substrate. This small misfit may be reduced further when some stacking faults are incorporated after two atomic rows of KCl. This new periodicity produces a $4\times4$ superstructure. A calculation of the pair-correlation function in the KCl film shows that the peak at 75% should be dominant. A strictly periodic structure should also produce, at some energies, measurable intensities at the 1/4, 1/2, and full position, which was not observed in the narrow useful energy range. Therefore some deviations from strict periodicity are necessary for a full description of the experimental findings. A step
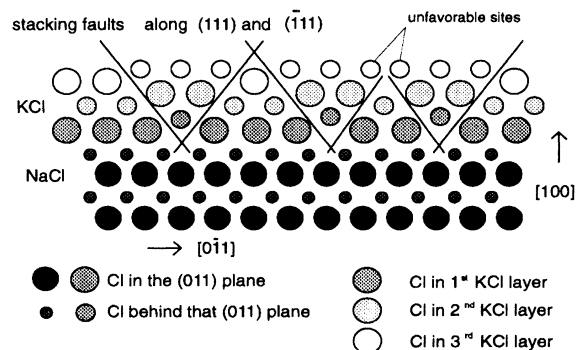


FIG. 5. Cross section through an NaCl(100) film with 3 ML of KCl showing the proposed misfit accommodation with double stacking faults. Only the chlorine ions are shown.

height analysis using the energy dependence of the satellite intensity provides a prominent step height of 1/3 layer distance, which is compatible with the stacking fault model of Fig. 4.

For more than 3 ML again the KCl lattice constant is found, although with a large half width, showing that the stacking faults are overgrown or recrystallized with many defects.

## IV. DISCUSSION

The comparison of Fig. 5 with Fig. 1 clearly shows the improvement of the fit by the periodic stacking fault array. Whereas in Fig. 1 the atoms in the KCl film are shifted against the perfect site from 0 to 50%, in Fig. 5 the interface atoms are shifted only 10% at the most. The atoms of the second layer are in perfect sites with respect to the nearest neighbors, where the surface has the structure equivalent to the interface layer. Also, the atoms of the third layer are at exact bulk distances, as long as three rows are at the interface. As is seen on the right half of Fig. 5, over a region with only two rows at the interface between stacking faults the third layer atoms are too close. This might be the reason that the film is still compressed by 2.5%, although a mixture with two-row distances at the interface would have reduced the misfit. The regular three-row structure therefore should be stable only up to a thickness of three layers in this case.

The LEED pattern shows all four first-order spots at 75% in the $[0\bar{1}1]$ and $[01\bar{1}]$ directions. There has to be therefore a square network of stacking faults with three rows in both directions between stacking faults. The crossings of the stacking faults provide no more sites, which are perfect with respect to nearest neighbors. Obviously the deviations are small, so that, nevertheless, the stacking fault structure is favorable.

The above interpretation for the KCl/NaCl system may be generalized in the following way: For heteroepitaxy of fcc(100) (including the NaCl and the diamond structure) a misfit $m = 1/M$ may be accommodated by islands of about $n = 1/(3m) - 2/3$ rows. Those islands are connected by double stacking faults as shown in Fig. 5. The maximum number of layers in this structure is given by the number of rows between the stacking faults. The rows fit into the substrate with a maximum shift of about 1/6 of the lattice constant compared to the worst case, in Fig. 1, of 1/2. An arbitrary misfit may be accommodated by an appropriate mixture of $n$ and $n + 1$ rows or by a residual strain of about $m^2$, which is much smaller than for the pseudomorphic case with the strain equal to the misfit.

The only reported case of heteroepitaxial growth including a periodic array of stacking faults is the epitaxy of Xe on Si(111) $7 \times 7$.[6] Since the $7 \times 7$ structure shows a strong corrugation[7] and six Xe distances are rather close to the width of the $7 \times 7$ unit mesh, the observed LEED pattern has been explained with a stacking fault every seven Si distances in all three directions $\{2\bar{1}\,\bar{1}\}$ of the (111) surface, as indicated in Fig. 2. The inclination of the Xe(111) layers of $3°$ against the Si(111) plane has been observed.[6]

Recently another example seems to fit exactly the model presented here: for epitaxy of Cu on Ni(100) the STM image shows stripes elevated by 1/6 of the lattice constant with a width equal to the film thickness.[8] A careful analysis should prove the stacking fault model.

The stacking fault model may present a means of misfit accommodation, which is a necessary intermediate structure during heteroepitaxial growth of very thin films. For thicker films the lattice has to rearrange in some way, since a crossing of the stacking faults should be very unfavorable. Understanding and controlling or avoiding the intermediate stacking fault structure may help to produce perfect epitaxial layers even for systems with a large misfit.

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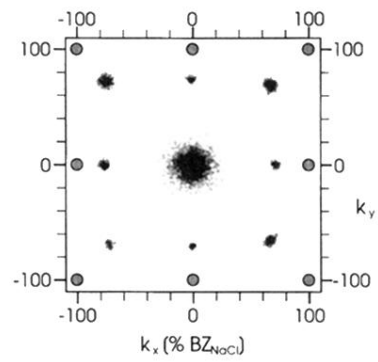
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