Film/Substrate Orientation Relationship in the AlN/6*H*-SiC Epitaxial System

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(Received 29 February 1996)

AlN has been grown on the (0001) basal and $(1\bar{1}00)$ prism faces of 6*H*-SiC. High resolution transmission electron microscopy studies show that, despite the very different symmetries of these two substrate planes and their polar and nonpolar natures, the film/substrate orientation relationship is identical for both systems. It is shown that this result is consistent with a geometrical method recently proposed for determining the orientation relationship between two crystals in a bicrystal. [S0031-9007(96)00923-4]

PACS numbers: 68.55.–a

With the recent advent of optoelectronic devices in the wide and direct band-gap semiconductors in blue and ultraviolet regions of the spectrum, GaN, AlN, and other III nitrides have become a major focus of interest. Single crystal films of both AlN and GaN are currently grown on different substrates such as sapphire (α-Al_2O_3), SiC, and Si by metalorganic vapor phase epitaxy (MOVPE) and molecular beam epitaxy (MBE). In early research, Yoshida *et al.* [1] and Morita *et al.* [2] deposited single crystal AlN on sapphire by gas-source MBE and metalorganic chemical vapor deposition (MOCVD), respectively. More recently, Rowland *et al.* [3], Sitar *et al.* [4], and Tanaka *et al.* [5] investigated the growth of AlN on 6*H*-SiC by plasma-assisted, gas-source molecular beam epitaxy. For epitaxial growth of GaN on sapphire and 6*H*-SiC, it has been found that an initial deposition of AlN acts as a buffer layer and improves the quality of the GaN film [6,7]. This is partly because AlN has the same crystal structure (wurtzite or 2*H*) as GaN; partly due to the fact that the AlN/sapphire or AlN/SiC lattice and thermal mismatches are smaller than either GaN/sapphire or GaN/SiC; and, perhaps more importantly, because AlN acts as a wetting agent and thus promotes better lateral growth [8]. Because of the relatively small lattice and thermal mismatches of AlN and SiC, the latter material is potentially a better substrate than sapphire for growth of GaN, and the system GaN/AlN/SiC has been recently characterized by a number of workers [9–13]. All of these systems, as well as AlN [3–5], were grown on commerically available 6*H*-SiC wafers which were parallel to the (0001) basal plane. Recently the $\{1\bar{1}00\}$ prism plane of 6*H*-SiC has also been considered as a possible substrate material [14]. In this research, we have used high resolution transmission electron microscopy (HRTEM) to study and compare the growth of AlN on orthogonal faces of single crystal 6*H*-SiC, namely, the (0001) basal plane and the $\{1\bar{1}00\}$ prism plane. These two faces have very different symmetries, however, the orientation relationship (OR) between AlN and 6*H*-SiC was found to be identical implying that, for this particular system, the orientation that the film chooses during nucleation and growth is independent of the substrate surface. Subsequently we show that this result can be predicted from a geometrical method recently developed for predicting the orientation relationship between two identical or different crystals in contact [15].

Both the 6*H*-SiC substrate and the AlN film have a $P6_3mc$ space group. The basal (0001) planes of a 6*H*-SiC crystal are related by a 6_3-fold screw axis along the *c* direction normal to the basal planes, and $\{1\bar{1}00\}$ is a *c*-glide symmetry plane of the crystal. These different symmetries can be seen in the very different atomic configurations of the (0001) and $\{1\bar{1}00\}$ faces of 6*H*-SiC, shown in the schematic projections of Figs. 1(a) and 1(b). In addition to their different symmetries, another important difference between the (0001) and $\{1\bar{1}00\}$ faces of a 6*H* (as well as the wurtzite) structure is that the former is polar whereas the latter is nonpolar. It is well known that in epitaxial growth of compound semiconductors the substrate polarity often has a large influence on the film properties [16,17].

The AlN films were grown on the (0001) Si-terminated face of 6*H*-SiC by MOVPE at 1100 °C, and on the $\{1\bar{1}00\}$ face of 6*H*-SiC by MBE at 1050 °C; for details of growth, see Refs. [3] and [12]. The crystals were subsequently investigated by HRTEM. For this purpose, cross-sectional TEM specimens were prepared from the two crystals with the foil surfaces parallel to a $\{11\bar{2}0\}$ plane of the 6*H*-SiC substrate. The microscopy was performed on a JEOL 4000EX microscope operating at 400 kV with the incident electron beam along the $\langle 11\bar{2}0\rangle$ zone axis of the substrate; this viewing direction is the most suitable for HRTEM of hexagonal semiconductors.

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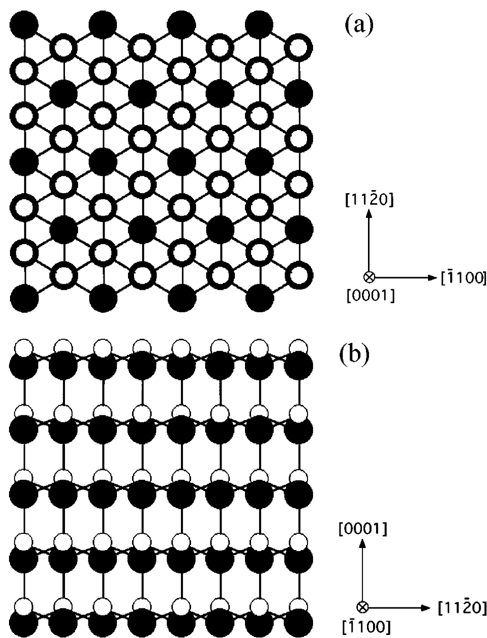

FIG. 1. Schematic projection of 6*H*-SiC on the (a) (0001) basal plane; (b) (1$\bar{1}$00) prism plane. The large open circles and small solid circles represent silicon and carbon atoms, respectively.

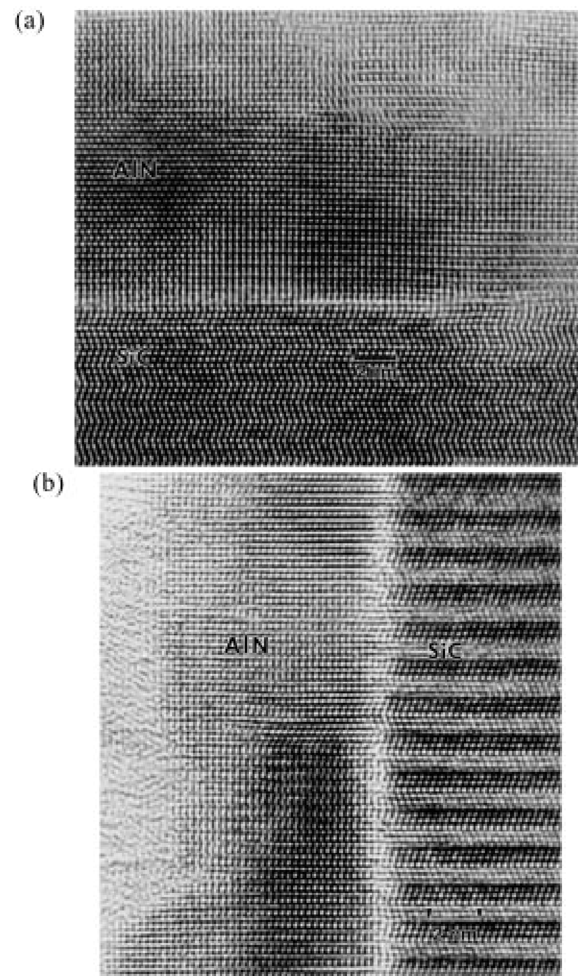

FIG. 2. Cross-sectional HRTEM micrograph of AlN film deposited on the (a) (0001) basal plane of 6*H*-SiC; (b) (1$\bar{1}$00) prism plane of 6*H*-SiC. In both figures, the incident electron beam is along the $[11\bar{2}0]_{\mathrm{AlN}}//[11\bar{2}0]_{\mathrm{SiC}}$ zone axes.

Figures 2(a) and 2(b) show HRTEM micrographs of the two composite crystals. From the inspection of these micrographs, and also the corresponding diffraction patterns along the $\langle 11\bar{2}0\rangle$ zone axis (not shown), it is immediately clear that the film/substrate OR is identical in both cases and is in fact a parallel epitaxial orientation relationship:

$$(0001)_{\mathrm{AlN}}//(0001)_{\mathrm{SiC}}\,,$$
$$\langle 11\bar{2}0\rangle_{\mathrm{AlN}}//\langle 11\bar{2}0\rangle_{\mathrm{SiC}}\,.$$

The studies of Akasaki *et al.* [8] indicate that, during deposition of AlN on the (0001) face of sapphire at ~600 °C, AlN nucleates as fine crystallites together with an amorphous layer which crystallizes at higher temperatures (~1000 °C) to form a homogeneous single-crystal film of AlN. For the much smaller lattice-mismatched system AlN/SiC and the higher growth temperature of ~1000 °C, the growth is presumably more two dimensional and takes place by the layer-by-layer mode. However, even in the this case there are indications that deposition of AlN proceeds by nucelation and subsequent growth and coalescence of (nearly 2D) island nuclei. The fact that the film/substrate OR is the same for deposition of AlN on (0001) and $\{1\bar{1}00\}$ SiC surfaces is a rather surprising result because one would expect that the underlying substrate atoms influence the aggregation of the film atoms on the substrate and the way that the 3D or 2D islands are oriented.

The OR shown by the above equation can be actually predicted from a geometrical model that was recently proposed and applied to a number of epitaxial systems [15]. This method originated from electron diffraction and HRTEM studies of a number of different heteroepitaxial systems. Inspection of the composite diffraction patterns of a bicrystal (e.g., a film/substrate system) along different zone axes revealed each time that the low index diffraction vectors in the two patterns were *nearly* aligned and of *nearly* equal length. Since a diffraction pattern is simply a section of the reciprocal lattice of a crystal, the experiments suggested that there is a *three-dimensional* near coincidence of diffraction spots of the two crystals in the reciprocal space.

In order to formulate the problem, each reciprocal lattice point *hkl*, corresponding to the reciprocal lattice vector **g**, is represented by a *sphere, g,* of radius r^* around the point. Such a reciprocal lattice sphere then corresponds to various sets of nearly parallel planes to the (*hkl*) set, and with nearly equal interplanar spacings $1/(|\mathbf{g} \pm \Delta\mathbf{g}|)$. It is then hypothesized that those orientation relationships between crystal 1 (with reciprocal lattice spheres g) and crystal 2 (with reciprocal lattice spheres G) are favored in which the sum volume of all intersections, $g \cap G$, is maximized. In order to apply this method and predict the OR between two crystals, the reciprocal lattice points of

the two crystals are calculated from their primitive unit cells, and each point is replaced by a sphere of radius r^*. Assuming that the origin of the reciprocal lattices of the two crystals are coincident in the 3D (reciprocal) space, one of the crystals (say, crystal 2) is rotated about two orthogonal axes of the other crystal (crystal 1). Each orientation of crystal 2 with respect to crystal 1 (i.e., each OR between crystal 1 and crystal 2) is specified by a set of rotation angles (θ, ϕ). At each value of (θ, ϕ), a number of reciprocal lattice spheres of the two crystals intersect, corresponding to those sets of planes in the two crystals which are near parallel and near equispaced. The volume ν_{gG} at the intersection of reciprocal lattice spheres g and G of the two crystals is calculated for all the spheres in the reciprocal space and the sum $V(\theta, \phi) = \sum \nu_{gG}$ at this OR is computed. Subsequently $V(\theta, \phi)$ is plotted versus θ and ϕ. In all the cases that have been examined, peaks are obtained at those (θ, ϕ) sets that are consistent with the expected or experimentally observed ORs [15]. Thus in the case when two identical cubic crystals are in contact (thus forming a grain boundary), all the coincidence site lattice (CSL) orientation relationships [18] are recovered; in the case of heteroepitaxial systems that were experimentally examined, such as V/MgO, V/Al_2O_3 (or Nb/Al_2O_3), and bcc/fcc metals, the computed ORs are in agreement with experimental observations [15]. It should be mentioned that, in addition to the major peaks, other peaks (of smaller amplitude) are also obtained that have not been observed in our experiments. This method may be compared with the *near-coincidence lattice site* model that has been discussed by a few authors [19–21] except that the whole treatment is in the reciprocal space, i.e., Fourier transform of the real crystal space.

In a review of the various geometric criteria that have been proposed for low-energy interfaces, and their comparison with experimental results, Sutton and Balluffi [22] concluded that none of the criteria can be regarded as wholly reliable. The method described in the present paper discusses the OR between two crystals and not the interface between them; the latter is presumably determined according to energetic reasons, i.e., those interfaces are favored which have the lowest energy. In the application of this method, the interface between two crystals does not enter in the determination of the OR between them. Rather, the preferential OR is one in which (1) a maximum number of sets of lattice planes in the two crystals are as parallel to each other as possible, and (2) these near-parallel sets of lattice planes have as close an interplanar spacing as possible.

In order to apply the above-described model to AlN/SiC, we have computed and plotted $V(\theta, \phi)$ versus θ and ϕ for this system. We shall first present some results showing the effect on the plots of varying the parameters r^* (radius of the sphere surrounding each reciprocal lattice point) and R^* (the extent in the reciprocal space within which the summation calculations are carried out). A few representative examples of these calculations are shown as three-dimensional plots of

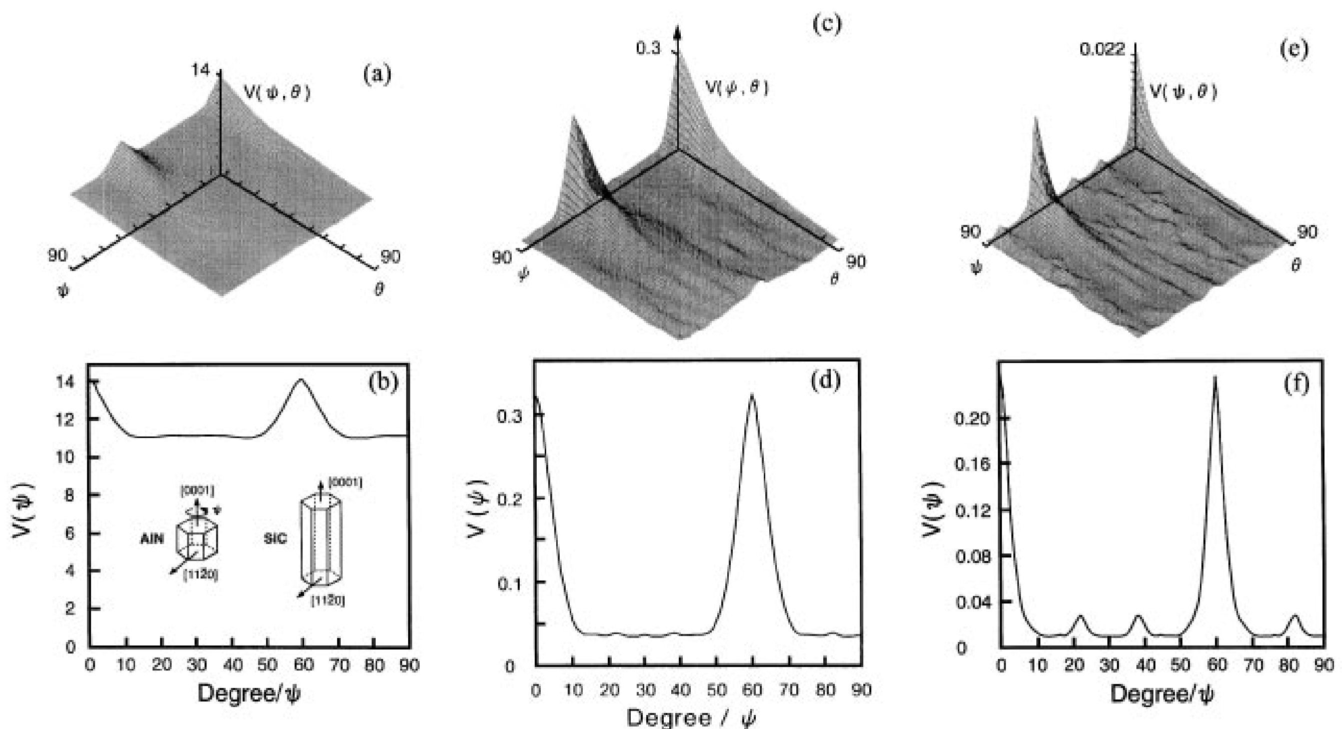

FIG. 3. (a),(c),(e) $V(\theta, \phi)$ as a function of θ and ϕ for (a) $r^* = 0.5a^*$, $R^* = 2.0a^*$, i.e., $R^*/r^* = 4$; (c) $r^* = 0.2a^*$, $R^* = 2.0a^*$, i.e., $R^*/r^* = 10$; (e) $r^* = 0.1a^*$, $R^* = 2.0a^*$, i.e., $R^*/r^* = 20$; the initial orientation $\theta = \phi = 0$ of (conventional) unit cells of the two crystals is inset in (b). (b),(d),(f) The $\theta = 0$ section of the 3D plots of (a), (c), and (e), respectively. Note the 60° periodicity of $V(\phi)$ along the ϕ axis.

$V(\theta, \phi)$ in Fig. 3 with the value of R^* kept constant at $2.0a^*$ and r^* varied between $0.5a^*$ and $0.1a^*$, where a^* is the basal edge of the reciprocal lattice unit cell of 6*H*-SiC [i.e., $a^* = |(\mathbf{b} \wedge \mathbf{c})/(\mathbf{a} \wedge \mathbf{b} \cdot \mathbf{c})|$, where $\mathbf{a} = \mathbf{b}$ and $\mathbf{c}$ are the unit cell parameters of the real lattice]. The initial orientation of the two unit cells (at $\theta = \phi = 0$) is shown in the inset in Fig. 3(b) where $r^* = 0.5a^*$; this corresponds to a ratio $R^*/r^* = 4$. In Fig. 3(c), $r^* = 0.2a^*$ corresponding to $R^*/r^* = 10$, and in Fig. 3(e), $r^* = 0.1a^*$ corresponding to $R^*/r^* = 20$. All figures are very similar and basically exhibit two predominant peaks in $V(\theta, \phi)$ corresponding to $\theta = 0$—at which the basal planes of the two crystals are parallel—and $\phi = 0$ or $\phi = 60°$—at which the $\langle 11\bar{2}0\rangle_{\mathrm{AlN}}$ directions of the two crystals are parallel. These are shown more clearly in Figs. 3(b), 3(d), and 3(f), which show two-dimensional $V(\phi)$ plots corresponding to sections of Figs. 3(a), 3(c), and 3(e) at $\theta = 0$. These computed results are precisely in accord with the experimental observations shown in Fig. 2. The peak is periodic in ϕ with a periodicity of 60°, which is as it should be for a 6-fold symmetric crystal. The results of our calculations, and the few examples shown in Fig. 3, also indicate that an increase in the value of R^*, or a decrease in the value of r^*, results in an enhancement in the peak resolution. Thus as R^*/r^* increases from 4 in Fig. 3(b) to 10 in Fig. 3(d) to 20 in Fig. 3(f), the relative amplitude of the major peaks (at $\phi = 0, 60, \ldots$) increases and they become sharper. In addition, secondary peaks at $\phi = 21.8°$ and $\phi = 38.2°$ start to appear at larger values of R^*/r^* which correspond to two new ORs; in both these cases, the basal planes of the two crystals are still parallel, i.e., $(0001)_{\mathrm{AlN}}//(0001)_{\mathrm{SiC}}$, but similar directions in these planes are no longer parallel, e.g., the $\langle 11\bar{2}0\rangle_{\mathrm{SiC}}$ is not parallel to $\langle 11\bar{2}0\rangle_{\mathrm{AlN}}$ but, instead, is parallel to a high-index $\langle uvw0\rangle_{\mathrm{AlN}}$ direction of AlN.

In conclusion, we have compared the orientation relationship between AlN films grown on two different faces of a 6*H*-SiC substrate, one parallel to its basal plane and the other parallel to its prism plane. HRTEM investigations of the two film/substrate systems show the surprising results that, despite the very different symmetries of these two surfaces and their polar and nonpolar natures, the AlN/SiC OR is identical in the two cases. This implies that in the AlN/SiC system the substrate surface does not influence the orientation of the film. This is similar to the Nb/Al_2O_3 [23] and V/Al_2O_3 [24,25] systems, where the metal grows with a unique orientation with respect to sapphire irrespective of the substrate surface on which growth is carried out. The results are consistent with a geometrical model for bicrystal orientation relationships developed in Ref. [15].

We would like to thank Ling Zhou and Dr. Fen-Ren Chien at CWRU for assistance with the theoretical calculations and TEM specimen preparation, respectively. Thanks are also due to Cheng Wang and M. D. Bremser at NCSU for carrying out the growth of AlN/SiC bicrystals. The work at CWRU was supported by Grant No. DE-FG02-93ER45496 from the Department of Energy, and in JFCC by the Frontiers Ceramics Project from the Science and Technology Agency of Japan.

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