Advances in the molecular-beam epitaxial growth of artificially layered heteropolytypic structures of SiC

Andreas Fissel,[a] Bernd Schröter, Ute Kaiser, and Wolfgang Richter
Friedrich-Schiller-Universität Jena, Institut für Festkörperphysik, Max-Wien-Platz 1, D-07743 Jena, Germany

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The controlled growth of SiC heteropolytypic structures consisting of hexagonal and cubic polytypes has been performed by solid-source molecular-beam epitaxy. On on-axis substrates, 4H/3C/4H−SiC(0001) and 6H/3C/6H−SiC(0001) structures were obtained by first growing the 3C−SiC layer some nanometer thick at lower substrate temperatures ($T=1550$ K) and Si-rich conditions and a subsequent growth of α-SiC on top of the 3C−SiC layer at higher T (1600 K) under more C-rich conditions. On off-axis substrates, multiheterostructures consisting of 4H/3C- or 6H/3C-stacking sequences were also obtained by first nucleating selectively one-dimensional wire-like 3C−SiC on the terraces of well-prepared off-axis α-SiC(0001) substrates at low $T(<1500$ K). Next, SiC was grown further in a step-flow growth mode at higher T and Si-rich conditions. After the growth, many wire-like regions consisting of 3C−SiC were found also within the hexagonal layer material matrix indicating a simultaneous step-flow growth of both the cubic and the hexagonal SiC material. © *2000 American Institute of Physics.* [S0003-6951(00)01039-1]

The growth of structures consisting of different SiC polytypes is a challenge for new applications of the semiconductor material SiC, such as resonant tunneling or other heterojunction devices. It offers also the potential of increasing flexibility in the design of SiC-based electronic structures.[1] Moreover, superlattices of SiC polytypes with confined electrons in a two-dimensional gas may also be of interest for future applications.[2,3] However, no suitable heterostructures were obtained so far because of the high defect level resulting from the heteropolytypic growth, such as twin boundaries, and, moreover, because the growth of such structures demands definite nucleation conditions for each of the polytype forming the structure. We report on growth conditions of SiC essential to realize multiheterostructures of different polytypes.

The experiments were performed by solid-source molecular beam epitaxy (MBE) at substrate temperatures (T) between 1200 and 1600 K. Several experiments were performed by varying T, growth rate (R), the Si/C ratio and the substrate off-angle. The experimental conditions of solid-source MBE are described elsewhere.[4–6] The obtained structures were investigated by cross-sectional transmission electron microscopy (TEM), electron channeling (EC) patterns, and photoluminescence (PL) spectroscopy.

Before the growth experiments, the substrate surfaces were prepared *ex situ* by plasma etching and wet chemical treatment.[7] *In situ* the samples were prepared by sublimation etching at 1600 K in a Si flux of $10^{14}\,\mathrm{cm^{-2}\,s^{-1}}$. Whereas no ordered step structure was found after *ex situ* preparation, a well-developed step array was obtained after this treatment, with steps typically of 2–6 monolayers (ML) in height in case of 6H−SiC and of 1–4 ML in case of 4H−SiC, respectively. Afterwards a SiC-buffer layer was grown via step flow at the same T and a carbon flux of $10^{13}\,\mathrm{cm^{-2}\,s^{-1}}$, corresponding to $R=7.5$ nm/h, a rate low enough to prevent nucleation also on nominal on-axis substrates.[6,8] The final step morphology mostly consists of steps one unit cell in height.

The most serious problem in the growth of SiC heterostructures is the occurrence of incoherent twin (double-position) boundaries (DPB) when 3C−SiC is involved. Recently, we have already[6,8,9] shown that the growth of 3C−SiC can be significantly improved by an alternating supply of Si and C at medium T (1430 K) on on-axis α-SiC(0001). The

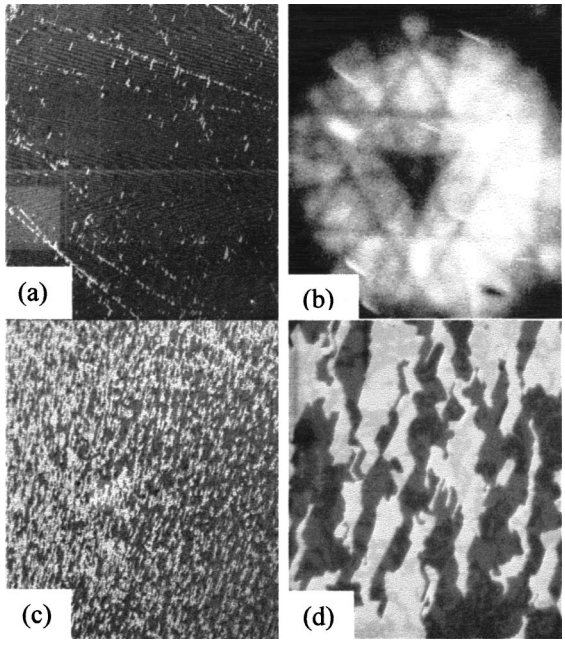

FIG. 1. EC micrographs (400×400 μm) of 3C−SiC layers grown by solid-source MBE on on-axis 4H−SiC(0001) at a growth rate of 30 μm/h at 1550 (a) and 1475 K (c) and (d) (40×40 μm) (lighter parts represent the other cubic domain); (b) corresponding EC pattern of the single domain layer grown at 1550 K.

[a]Electronic mail: p5anfi@rz.uni-jena.de

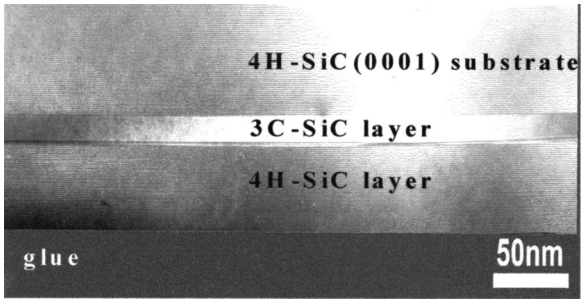

FIG. 2. TEM micrograph of a 4H/3C/4H–SiC(0001) heterostructure grown at 1550 (3C–SiC) and 1600 K (4H–SiC) by solid-source MBE.

diameter of DPB domains was found to be increased to some hundreds μm. That already demonstrated the importance of adatom mobility and nucleation conditions for the growth of single domain 3C–SiC layers. A further improvement of the 3C–SiC layer quality was achieved now by a further increase in T and improvement of the surface quality, as demonstrated in Fig. 1. In this way single domain 3C–SiC was obtained on α-SiC(0001) also by a continuous supply in dimensions of some mm also for layers only some nanometer (nm) thick. 3C–SiC in twin position was found to be only associated with surface imperfections like scratches or dislocations. This indicates a coherent nucleation of 3C–SiC in one orientation at more equilibrium-like conditions and on well-ordered surfaces. Furthermore, as already demonstrated for the growth of 3C–SiC on 6H–SiC(0001),[10] the 3C–SiC layers grow pseudomorphic on hexagonal SiC. That means, there is no difference in the in-plane lattice constant between the cubic layer and the hexagonal substrate material, whereas the lattice constants in the growth direction of the hexagonal and cubic SiC are different.

Growing SiC at 1600 K under more C-rich conditions on the 3C–SiC layer at the same growth rate results either in the formation of 4H–SiC (Fig. 2) on top of the 3C layer grown on 4H–SiC(0001) or of 6H–SiC when the 3C layer was grown on 6H–SiC(0001). The regions of homogeneous formation of the hexagonal polytypes on 3C–SiC were found to be in the range of some μm, which is likely limited in the moment by bulk-related defects, such as dislocations and micropipes. A double-heterostructure consisting of 6H/3C/6H/3C/6H–SiC(0001) was also achieved (Fig. 3). The preferential formation of the hexagonal polytypes at low supersaturations and C-rich conditions agrees very well with estimations performed within the framework of nucleation theory.[11] However, the formation of either only a 4H/3C heterostructure on 4H–SiC or of 6H/3C on 6H–SiC indicates also an influence of the strain within the layers on the structure of polytype grown. The more C-rich conditions in the Si/C ratio can easily be checked by the occurrence of the $(\sqrt{3}\times\sqrt{3})$ superstructure in the reflection high-energy electron diffraction (RHEED) pattern, which is formed by a Si adlayer of 1/3 ML on top of the SiC surface[12] and appears before surface graphitization.[6] In Fig. 4 are given conditions for the occurrence of the superstructure as function of T and the applied Si flux, which is used in excess in the Si/C ratio during the growth under more C-rich conditions.

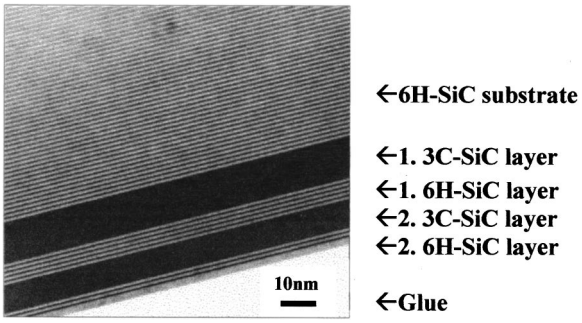

FIG. 3. TEM micrograph of a 6H/3C/6H/3C/6H–SiC(0001) doubleheterostructure grown at 1550 (3C–SiC) and 1600 K (6H–SiC) by MBE.

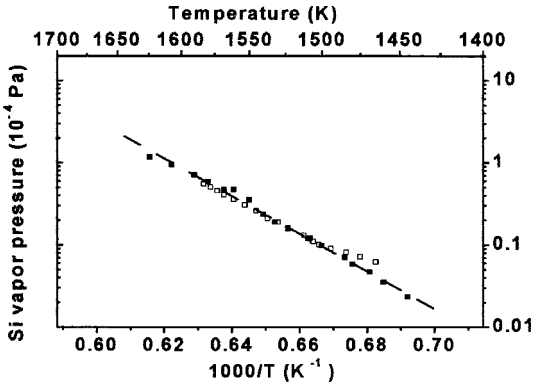

FIG. 4. Arrhenius plot of equilibrium Si vapor pressure to stabilize the $\sqrt{3}\times\sqrt{3}$ surface superstructure on α-SiC(0001). The dotted line indicates the occurrence of the superstructure and corresponds to an activation energy of 4.5 eV. Above this line, the conditions becomes Si rich, whereas below this line graphite will be formed.

On off-axis substrates, 3C–SiC was grown by an alter-

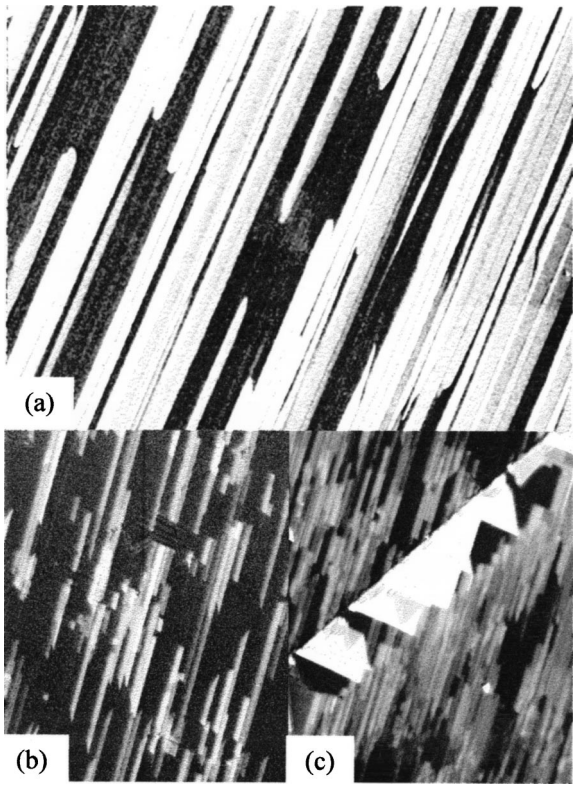

FIG. 5. EC contrast micrograph of (a) 3C–SiC wire-like nuclei formed on terraces of 0.9° off-axis 6H–SiC(0001) at 1500 K; 1.6-μm-thick SiC layer grown at 1600 (b) and at 1550 K (c) on 3.5° off-axis 4H–SiC(0001) after nucleating of wire-like 3C–SiC at 1350 K (light parts correspond to 3C–SiC).

nating supply at lower T down to 1300 K to improve the layer quality and to prevent the step-flow growth, respectively. The nucleation of SiC islands results in the formation of wire-like regions of 3C–SiC on the terraces of α-SiC(0001). Depending on the off-angle and the growth conditions (T,R) either nucleation takes place on many of the terraces (low off-angle, low T, high R) or only on some larger ones ($>1°$, higher T, low R). After further growth at high T (1600 K) and Si-rich conditions, such wire-like regions were also found within the hexagonal layer material, as demonstrated in Fig. 5. These wires also consist of 3C–SiC as revealed by EC and PL spectroscopy. 3C–SiC formed by nucleation at defects (scratches) and grown on the terraces are clearly distinguished by its intensity and shape. This indicates a simultaneous growth of hexagonal and cubic parts.

In summary, the growth of new artificially layered heterostructures of SiC consisting of a hexagonal and cubic polytype has been demonstrated by means of solid-source MBE. There are two different ways depending on the substrate orientation. On well-oriented substrates, at first 3C–SiC was nucleated and subsequent grown at 1550 K. 3C–SiC layers nearly free of DPB were obtained in this way on well-prepared surfaces. Dependent on the substrate polytype, on the 3C–SiC layer either 4H– or 6H–SiC was grown afterwards at higher T and at C-rich conditions. On off-axis substrates, 3C–SiC was nucleated at even lower T to prevent a stepflow growth. At certain conditions (off-axis angle) only a selective nucleation occurs, leading to wire-like low-dimensional structures. In a second step, the SiC layers were grown further by a step-flow growth mode of both the cubic nuclei and the hexagonal substrate material. In the result of such a growth process a multiheterostructure was formed consisting of many cubic and hexagonal lamellae.

Actually, the physical properties of this new kind of heterostructure are under investigation besides the improvement of the heterostructure homogeneity.

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