Spiral growth and threading dislocations for molecular beam epitaxy of PbTe on BaF₂ (111) studied by scanning tunneling microscopy

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Molecular beam epitaxy of PbTe on BaF₂ (111) is studied using UHV-scanning tunneling microscopy and atomic force microscopy. It is shown that PbTe growth is totally dominated by growth spirals formed around threading dislocations (TD) that originate from the growth on the 4.2% lattice-mismatched substrate. Due to dislocation annihilation, the TD density rapidly decreases with layer thickness, which results in a dramatic increase of the electron mobilities in the layers.

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Advances in epitaxial growth of lead-salt (IV-VI) semiconductors¹ have recently lead to room temperature operation of midinfrared lead-salt diode lasers in pulsed mode² and to cw operation at 223 K.³ While these devices are usually grown on lead-salt substrates, (111) oriented BaF₂ has proven to be an excellent alternative as substrate material for lead-salt heterostructures.⁴ Although BaF₂ is still considerably lattice-mismatched with respect to most of the lead-salt compounds, it is readily available in high crystalline quality and no significant degradation of epitaxial layers occurs during thermal cycling because of the well matched thermal expansion coefficients. In addition, while lead-salt substrates are always highly conducting, BaF₂ is optically transparent and highly insulating, which is an essential prerequisite for the study of the electronic properties of heterostructures.⁵ Due to the much larger (111) free surface energies of the lead salts, however, epitaxial growth on BaF₂ always starts in a three-dimensional (3D) Vollmer-Weber growth mode. Therefore, thick buffer layers have to be deposited in order to achieve smooth overlayers with high crystalline perfection.

In this letter, we use UHV-scanning tunneling microscopy (STM) and *ex situ* atomic force microscopy (AFM) to study the surface structure of PbTe on BaF₂ (111) grown by molecular beam epitaxy (MBE). It is shown that after coalescence of the layers, the surface structure is completely dominated by growth spirals formed around threading dislocations (TDs) that originate from the growth on the 4.2% lattice-mismatched substrate. With increasing layer thickness we observe a drastic decrease of the TD density due to dislocation annihilation processes, correlated with an increase of the low temperature carrier mobilities in the layers. In contrast, additional deposition of lattice-mismatched overlayers results in a drastic increase of the TD density after strain relaxation. This is demonstrated for the case of PbTe growth on 2.1% mismatched EuTe spacer layers.

Epitaxial growth was carried out in a Riber MBE system with standard effusion cells for PbTe, Eu, and Te.⁵ Cleaved BaF₂ (111) substrates were prebaked at 500 °C for 15 min. For PbTe growth, a substrate temperature of 340 °C and growth rates of about 1 μm/h were used. Epitaxial surfaces were studied using a BEETLE STM and a PARK AFM. For

the STM measurements, epitaxial growth was interrupted at well defined growth sequences and after rapid cooling, the samples were transferred to an attached UHV-STM chamber. Large scale STM images were mounted together from several 0.5×0.5 μm² images recorded in a step and scan mode.

Using a thin EuTe nucleation layer predeposited on the BaF₂ substrates, *in situ* reflection high-energy electron diffraction studies indicate that coalescence of the PbTe islands starts already at a layer thickness of about 100 Å.⁷ Indeed, STM images taken at 170 Å layer thickness [Fig. 1(a)] show that the 3D islands already have merged together to form a continuous PbTe overlayer. At 1000 Å layer thickness, a totally uniform and extremely smooth overlayer has formed. However, as shown in Fig. 1(c), the corresponding surface has a very peculiar surface step structure that consists of very short segments of monolayer steps, where both ends are terminated by a threading dislocation (see arrows), which connect the misfit dislocation segments at the PbTe/BaF₂ interface with the free surface. If one draws a closed loop around the TDs on the surface, there remains a net surface normal displacement of 3.7 Å, equal to the height of one monolayer. This indicates that the TDs are of screw character. In the lead-salt compounds, the Burgers vector **b** of dislocations is usually of 1/2[110] type, with a (111) surface normal component of 3.72 Å, corresponding exactly to the observed surface displacements. Whereas a single screw-type threading dislocation would produce a semi-infinite step on the surface, here these steps are terminated by a second dislocation that cancels the surface normal displacements, i.e., by a dislocation with opposite normal component of **b**.

Since in our STM images no 2D islands are observed on the surface, a pure step flow growth mode is present where adatom deposition leads only to a *lateral* growth of the steps on the surface. Since in our case, the end points of these surface steps are all *pinned* by the threading dislocations, lateral growth at the step edges leads to an increasing bowing of the step segments until a closed step loop is formed and emitted. Thus, each pair of threading dislocations acts like a continuous Frank-Read source for surface steps. Due to the very close spacing of the TDs, however, there is no fixed step connection between two adjacent dislocations but rather a continuous step exchange between pairs of neighboring dislocations. This is illustrated schematically in Fig. 2, representing the surface area marked by (A), in Fig. 1(c). Ini-

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FIG. 1. STM images ($1.7 \times 1.4 \mu\text{m}^2$) of PbTe layers on BaF₂ (111) with thicknesses of: 170 Å, 600 Å, 1000 Å, 2000 Å, 5000 Å, and 3 μm [from (a) to (f)]. The gray scale represents the derivative of the height in the horizontal direction. The screw-type threading dislocations are indicated by arrows.

tially, dislocations (1) and (2) are connected by a surface step line, and likewise dislocations (3) and (4). As atoms are deposited on the surface, each step half-loop expands as indicated by the arrows, until step (3–4) meets TD (1). At this point, the step connection between the two pairs of dislocations is exchanged [Fig. 2(b)], i.e., a half-loop is formed between TDs (2) and (4) and a short step segment between (1) and (3). As then the bottle neck in the (2–4) half-loop closes, a complete step loop is emitted from the group of dislocations leaving inside two short step segments with exchanged partners [Fig. 2(c)]. Further growth leads to the reverse step exchange process, such that the original step configuration is essentially regained [Fig. 2(d)]. Each complete

pass through this exchange cycle corresponds to the growth of *one new* monolayer on the surface. Since in each cycle the original step configuration is reproduced, no overall change in surface step structure can occur during growth.

However, the STM images for larger layer thicknesses [Figs. 1(d) to 1(f)] show that as more material is deposited on the surface the steps start to wind up around the threading dislocations. While the curvature of the surface steps at the threading dislocations remains essentially constant (see Fig. 1), the increasing separation between TDs allows more and more complete turns of the step loops around the TDs and, therefore, larger and larger growth spirals are formed. At 3 μm layer thickness [Fig. 1(f)], the surface is completely covered by large growth spirals, which are typically several μm apart. Since the average step spacing within the spirals remains essentially constant (≈ 2000 Å), the large growth spirals consist of more than ten turns. Thus, the centers of these growth spirals are elevated by more than 50 Å, which gives rise to a gentle hillock surface structure that is often observed by Normarski microscopy.

Since we observe both right and left handed growth spirals on the surface, TDs with different orientations of **b** are present in the layers. Due to their elastic strain fields, TDs with antiparallel Burgers vector are strongly attracted to each other by a force that is inversely proportional to the dislocation spacing.⁸ Provided a sufficient dislocation mobility, TDs with opposite Burgers vector, thus, move closer and closer and when they meet, dislocation annihilation occurs, which eliminates the threading arms of the misfit dislocations at the PbTe/BaF₂ interface. This mechanism leads to the reduction in TD density. Figure 3 shows the TD density determined by STM and AFM as a function of layer thickness. With in-

FIG. 2. The change of step configuration during the deposition of one monolayer illustrated for the surface area marked (A) in Fig. 1(c). The two-step segments are each pinned by two threading dislocations with opposite **b** and act as a Frank–Read source for surface steps.

FIG. 3. Threading dislocation density for PbTe layers on BaF₂ as a function of layer thickness, determined by STM (▲) and AFM (●).

creasing layer thickness, the TD density rapidly decreases from above $1.5 \times 10^9 \text{ cm}^{-2}$ for $d = 1000 \text{ Å}$, to a few times 10^6 cm^{-2} for layers of several μm in thickness, following roughly a power law dependence with a negative exponent of 1.2 (dashed line in Fig. 3). This rapid decrease implies a quite high dislocation mobility at the elevated growth temperatures, which is consistent with the fact that the lead salts are very easily deformable materials.⁹ At the same time, we observe a drastic increase of the 10 K carrier mobilities by two orders of magnitude from $2 \times 10^4 \text{ cm}^2/\text{V s}$ to above $2 \times 10^6 \text{ cm}^2/\text{V s}$ for layer thicknesses from 1000 Å to 3 μm , respectively, which clearly indicates a strong correlation between carrier mobility and TD density in PbTe epitaxial layers.⁷

Although semiconductor substrates like Si or GaAs are essentially dislocation-free, misfit dislocation formation in strained-layer heteroepitaxy always leads to a high density of threading dislocations on the surface due to nucleation and multiplication of dislocations. We have studied the latter for MBE of PbTe on relaxed EuTe layers (2.1% lattice mismatch) predeposited on thick PbTe buffer layers. The AFM surface image of such a 2000 Å PbTe layer grown on a 330 Å EuTe spacer layer is shown in Fig. 4. In comparison to the thick PbTe buffer layers, we observe a drastic *increase* of the threading dislocation density by almost two orders of magnitude to $2 \times 10^9 \text{ cm}^{-2}$, whereas for EuTe spacer layers of only 120 Å, the TD density is equal to that of the PbTe buffer layers. This observation confirms our previous strain relaxation model, in which the *kinetics* of strain relaxation for EuTe on PbTe (111) were determined mainly by TD multiplication, with an onset at about 150 Å EuTe layer thickness.⁶ In addition, there is a striking difference in the surface structure of the 2000 Å PbTe layer on EuTe compared to that of the thick PbTe layers on BaF₂. In spite of the very high TD density, no growth spirals are observed on this surface. Obviously, the dislocation terminated step segments are locked within the step train of the highly terraced surface

FIG. 4. $3 \times 3 \mu\text{m}^2$ AFM image of a 2000 Å PbTe layer on a relaxed 330 Å EuTe spacer layer predeposited on a 3 μm PbTe buffer. Dislocation multiplication during strain relaxation produces a high TD density on the surface (arrows), but due to the highly terraced surface structure the formation of growth spirals is strongly suppressed.

structure (see Fig. 4), where on the narrow terraces there is not enough space for the steps to wind up around the threading dislocations. The observed highly terraced surface structure results from the dislocation glide processes during strain relaxation,⁶ and is quite typical for strained-layer heteroepitaxy. It is why for lattice-mismatched MBE of other semiconductors, a spiral type of growth mode is not observed even for high TD densities, with the exception of the highly mismatched GaSb/GaAs system.¹⁰

In conclusion, it was shown that the surface structure of PbTe epitaxial layers on BaF₂ (111) is determined mainly by threading dislocations originating from the PbTe/BaF₂ interface. This, in combination with persisting step flow growth results in a unique spiral type of growth mode, that is usually not observed for strained-layer heteroepitaxy of other semiconductors. Furthermore, it was demonstrated that continued growth results in a drastic decrease of the threading dislocation density, leading to significant improvements in the electronic properties of the layers.

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