Strain Relaxation and Misfit Dislocation Interactions in PbTe on PbSe (001) Heteroepitaxy

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Strain relaxation of heteroepitaxial PbTe layers on PbSe (001) by misfit dislocation formation is shown to take place near equilibrium without kinetic barriers. The comparison of the experimental data with different strain relaxation models shows that mutual dislocation interactions are of crucial importance for the strain relaxation process. This results in a faster relaxation than predicted by the Frank and van der Merwe model for non-interacting dislocation arrays.

Introduction

Strain relaxation is a key issue in lattice-mismatched heteroepitaxy [1], [2]. It occurs via formation of coherent three-dimensional (3D) islands in the Stranski-Krastanow growth mode or by the formation of interfacial misfit dislocations. The latter is usually described by the Matthews-Blakeslee [3] or Frank-van der Merwe [4] strain relaxation models, which yield the critical layer thickness as well as average dislocation densities in thermodynamic equilibrium. For the case of semiconductors, however, many experimental works have demonstrated that the actual course of strain relaxation is strongly affected by the dislocation kinetics [1], [2], which are determined by various processes such as nucleation, multiplication, thermally activated glide as well as blocking of dislocations. These processes impose kinetic barriers for misfit dislocation formation and therefore strongly retard the relaxation process. In the present work, we actually demonstrate the opposite behavior for PbTe on PbSe (001) heteroepitaxy with a lattice-mismatch of 5.2%. We not only find that a near-equilibrium strain state is formed in the layers but even more that the actual course of strain relaxation is significantly faster than predicted by the Mathews-Blakeslee (MB) or Frank-van der Merwe (FdM) models. This remarkable fact is shown to be due to the strong influence of mutual dislocation interactions on the energetics of the dislocation network. Comparing our results with recent models that have incorporated these dislocation interactions [5], we still find a significant deviation of our experimental data from the model calculations. Our result can be explained quite well under the assumption that the dislocation self-energy is essentially independent of the layer thickness.

Experimental and Results

The PbTe/PbSe samples were grown by molecular beam epitaxy. First, several µm thick PbSe buffer layers were deposited onto (001) oriented PbSe substrates at a temperature of 380°C. Then, PbTe layers were grown on top with thicknesses ranging from 1 to 50 monolayers (ML), where 1 ML corresponds to a layer thickness of 3.23 Å. *In situ* reflection high-energy electron diffraction (RHEED) was employed to monitor the growth process and determine the strain-state of the layers as a function of layer thickness. Complementary investigations were carried out using ultra-high vacuum STM as described in Ref. [6]. Here we focus on the question of how strain relaxation is influenced by the growth conditions, which were varied over a wide range of substrate tem-

peratures of 70 to 360°C and growth rates of 0.12 to 1.13 ML/sec. Complementary post-growth annealing studies were carried out for layers for which the growth was interrupted at layer thicknesses between 1 and 10 ML.

Figure 1 (a) shows the RHEED patterns recorded at a PbTe layer thickness of 14 ML. The elongated shape of the diffraction streaks indicates that a 2D surface is retained during PbTe growth in spite of the rather large lattice-mismatch. This is in agreement with our previous scanning tunneling microscopy studies [6]. Due to the small scattering angles, the lateral spacing of the diffraction streaks directly corresponds to the reciprocal space lattice vector, which is inversely proportional to the in-plane surface lattice constant. To determine the course of strain relaxation, intensity line profiles were recorded across the diffractions streaks as indicated by the white line in Fig. 1 (a) and shown in Fig. 1 (b). The evolution of these profiles as a function of layer thickness is displayed in the grey scale image of Fig. 1 (c). At a PbTe layer thickness of 1 ML, clearly the streaks start to shift inwardly, signifying the onset of strain relaxation. Using the initial PbSe surface with its bulk lattice parameter of $a_{0,sub}$ = 6.124 Å as a reference, the relative change of the streak spacing directly yields the relaxed strain ε_{rel} of the layer as a function of layer thickness *h.*

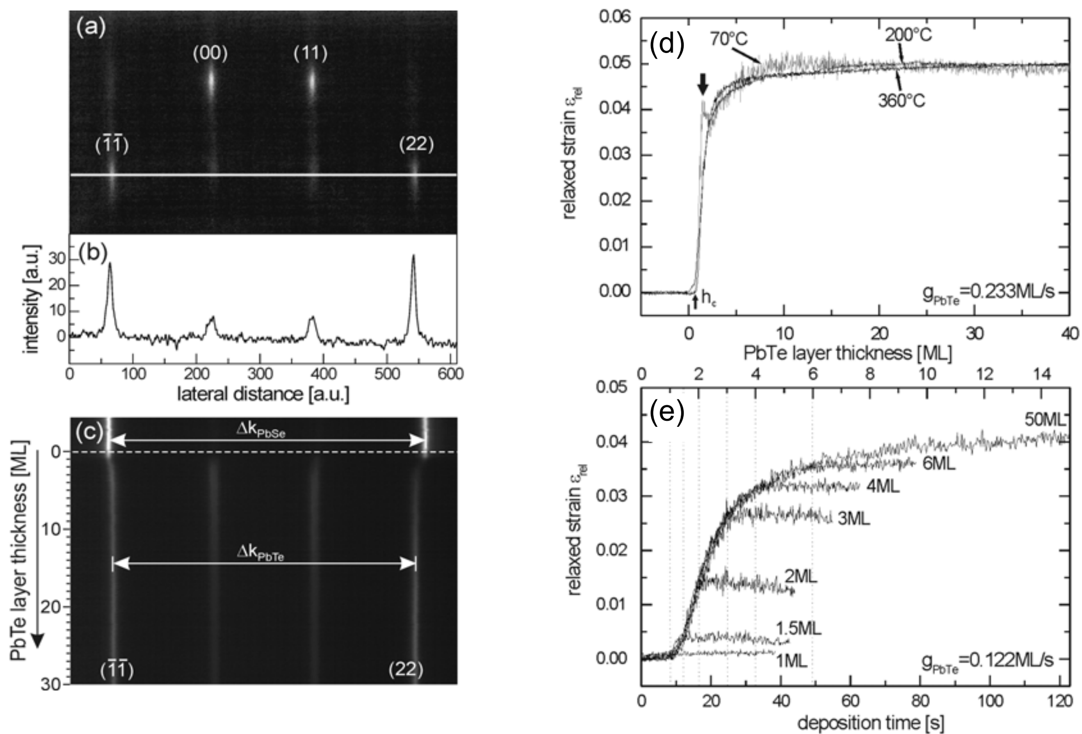

Fig. 1: Left hand side: (a) RHEED pattern of 14 ML PbTe on PbSe (001) recorded along the [100] azimuth. (b) Intensity profile across the (-1-1) and (22) streaks as indicated by the white line in (a). (c) Grey-scale image of the intensity profiles as function of the PbTe layer thickness. Right hand side: Strain relaxation curves of PbTe/PbSe (001) as a function of the epilayer thickness measured at different deposition temperatures (d) and during post-growth annealing (e) at 360°C after interruption of the growth at 1, 1.5, 2, 3, 4 and 6 ML.

The resulting relaxation curves for PbTe epilayers deposited at different growth temperatures of 70, 200 and 360 °C are shown in Fig. 1 (d). In all cases, the onset of strain relaxation occurs at a critical thickness of 1 ML, and no retardation of strain relaxation occurs as the temperature decreases. Furthermore, when interrupting the growth at

PbTe thicknesses of 1, 1.5, 2, 3, 4 and 6 ML (see Fig. 1 (e)), no further post-growth relaxation was observed during subsequent sample annealing. Identical results were obtained when the growth rate was varied between 0.12 to 1.13 ML/sec. This clearly demonstrates that strain relaxation in the PbTe/PbSe (001) heteroepitaxial system is not influenced by dislocation kinetics but takes place at near-equilibrium conditions.

To gain more insight on the strain relaxation process, the course of strain relaxation was modeled using the FdM approach in which the total energy of the layer/substrate system is minimized with respect to the dislocation density [4]. This facilitates the introduction of dislocation interactions [5]. Even more, the nearly perfect square network of pure edge misfit dislocations formed in the PbTe layers [6] represents a nearly ideal realization of the dislocation configuration envisioned by the original FdM model. This is illustrated by the STM image of a 9 ML PbTe layer displayed as insert in Fig. 2, in which the dark lines represent the grid of subsurface edge dislocations with a Burgers vector of $b = ½[110]a_0$. Neglecting surface and interface energies, the total energy density e_{tot} of a 2D epilayer on a non-compliant substrate is given by the sum of the coherency energy e_{coh} due to the biaxial strain in the layer and the self-energy e_{MD} of the misfit dislocation network. For relaxed layers, the initial misfit strain ε_0 is reduced by $-(\rho_{MD} b_{||,edge})$ by misfit dislocations, where $b_{||,edge}$ is the edge component of the Burgers vector within the interface plane (here equal to b) and ρ_{MD} is the 1D dislocation line density.

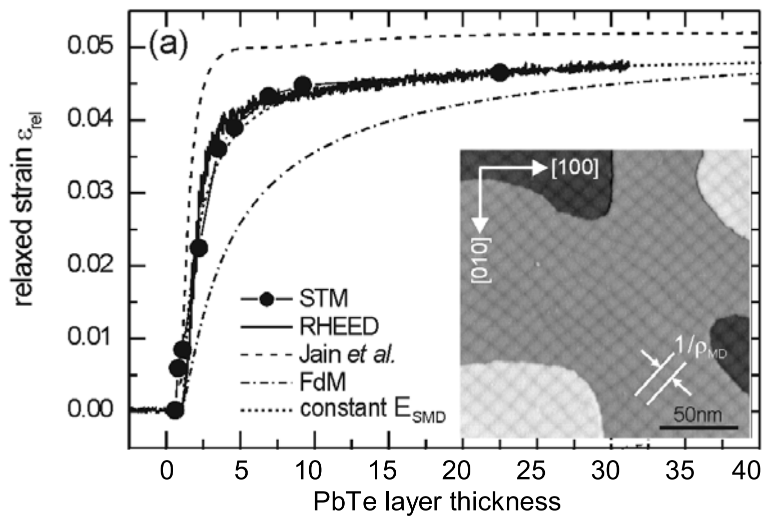

Fig. 2: Comparison of PbTe/PbSe (001) relaxation curves measured by RHEED (solid) and STM (dots) with the FdM model for non-interacting dislocations (dash-dotted), the model of Jain et al. for interacting dislocation arrays (dashed), and the model of constant dislocation energy for δ = 0.05 (dotted). Insert: STM image of a 9 ML PbTe layer on PbSe (001), where the dark lines indicate the quasi-periodic subsurface network of edge dislocations.

Thus, the coherency energy can be written as $e_{coh} = B\, h\, (e_0 - \rho_{MD}\, b_{||,edge})^2$ where the elastic energy factor $B = 2\mu(1 + \nu)/(1 - \nu)$ contains the shear modulus μ and Poisson ratio ν of the epilayer. The self-energy of the dislocation network e_{MD} is calculated by summation of the self-energies E_{SMD} of each misfit dislocation as $e_{MD} = 2\, \rho_{MD}\, E_{SMD}$, where the factor of two accounts for the fact that two orthogonal misfit dislocation arrays are required to relax the biaxial lattice-mismatch.

In the original FdM model, the self-energy of each isolated single misfit dislocation is given by the logarithmic expression $E_{SMD} = Kb^2 (4\pi)^{-1} \ln(h\alpha/b)$ where $K = \mu/(1-\nu)$ is the dislocation energy coefficient, here for pure edge dislocations, and the parameter α accounts for the energy at the dislocation core, which is usually chosen as 4 for semiconductors. Minimizing the total energy e_{tot} with respect to the dislocation density ρ_{MD} yields the critical thickness h_c as well as the equilibrium relaxed strain ε_{rel} as a function of layer thickness as:

$$h_c = \frac{Kb^2}{4\pi B \varepsilon_0 b_{||,edge}} \cdot \ln(h_c \alpha / b) \qquad \varepsilon_{rel}^{FdM}(h) = -\varepsilon_0 \left[1 - \frac{h_c}{h} \frac{\ln(h\alpha / b)}{\ln(h_c \alpha / b)} \right] \qquad (1)$$

Figure 2 shows the comparison of the calculated strain relaxation ε_{rel} with the data obtained by RHEED and STM. For the latter, ρ_{MD} was directly determined from the average dislocation spacing [6] observed in the STM images as indicated in the insert of Fig. 2. Whereas both experimental results fall onto the same line, the calculated $\varepsilon_{rel}(h)$ is considerably lower, *i.e.*, the experimentally observed strain relaxation is much *faster* than predicted by the FdM model. This is in spite of the fact that the calculated critical thickness of h_c = 1ML is in excellent agreement with the experimental value.

To resolve the disagreement, the expression for the dislocation self-energy has to be modified in order to include the mutual dislocation interactions that become important when the dislocation spacing is comparable to the layer thickness. Here we follow the calculation of Jain *et al.* [5], who have derived the self-energy of a periodic array of interacting dislocations. The minimization of the total energy as a function of dislocation density can then be done only numerically. To obtain the same critical thickness as in the FdM model and the experiments, a slightly larger core parameter α has to be used. The result of this model is plotted as dashed line in Fig. 2. In this case, a very fast initial strain relaxation is predicted, similar as observed in the experiments. However, the model yields an almost complete strain relaxation already at 5 ML layer thickness, where the experimental values are still considerably lower. This indicates that the Jain model overestimates the effect of dislocation interactions.

To resolve this problem, we propose a new model that assumes a constant dislocation self-energy of $E_{SMD} = Kb^2 / 4\pi$ independent of the layer thickness. In this case, straightforward explicit expressions for h_c and ε_{rel} are obtained, with

$$h_c = K\, b^2 (4\pi\, B\, b_{||,edge}\, \varepsilon_o)^{-1} \quad \text{and} \quad \varepsilon_{rel} = \varepsilon_0 \left[(1 - \delta) - h_c / h\right] \qquad (2)$$

where δ is a small correction parameter to account for a possible residual strain at large layer thicknesses. As is demonstrated by the dotted line in Fig. 2 with this new functional dependence the experimental data can be exactly reproduced.

Conclusions

In conclusion, dislocation interactions are of great importance for the strain relaxation process in heteroepitaxial layers. For PbTe on PbSe (001) system we therefore find a significantly faster strain relaxation as compared to the FdM model for non-interacting dislocations. This can be explained only by the effect of the dislocation interactions on the formation energies of the dislocations. Using a model in which the dislocation self-energies are assumed to be constant as a function of layer thickness, an excellent agreement with the experimental data is obtained. This work was supported by the FWF and the Academy of Sciences of Austria.

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