Microstructural Properties of Single Crystalline PbTe Thin Films Grown on $BaF_2(111)$ by Molecular Beam Epitaxy *

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Single crystal PbTe thin films have been grown on BaF_2 (111) by using solid source molecular beam epitaxy. The studies of evolution of the surface morphology with the increasing growth temperature from 375 to 525° C by AFM show that PbTe epilayers exhibit smooth surface morphologies with atomic layer scale roughness and are crack free. It is found that for PbTe grown at 475° C, the morphology is dominated by triangles and the rms roughness is 3.987 nm. Compared to the rms roughnesses of 0.432 nm and 0.759 nm for the samples grown at 375 and 525° C respectively, the surface of the PbTe layer grown at 475° C is much rougher. This roughening transition is due to the interaction between the elastic relaxation and the plastic relaxation during the strain relaxation process. In contrast to the result of the morphology that the PbTe epitaxial layer grown at 375° C has most smooth surface, as observed from the line width of x-ray diffraction curves at higher growth temperature improves the crystal quality of the single-crystalline PbTe layer.

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Recently Ⅳ-Ⅵ lead chalcogenide semiconductor materials have attracted much attention because of their important applications in infrared optoelectronic devices, such as long wavelength infrared intrinsic detectors and mid-infrared lasers.[1,2] The performance of these devices depends on the quality of epitaxial materials and to optimize the eptitaxy of materials *in situ* characterization technique has been used.[3] PbTe is one of lead chalcogenide materials, which has a narrow direct and temperature sensitive bandgap at room temperature.[1] Using PbTe as the buffer layer, Springholz *et al.* has realized the self-organized growth of three-dimensional quantum-dot crystals.[4] The crystalline quality of the PbTe buffer layer is of importance for the self-organized growth of quantum dots. The growth rate and substrate temperature is considered decisive in the growth of the lead chalcogenide.[5] However, to our knowledge few studies have been devoted to the optimization of PbTe growth conditions and the microstructural properties of PbTe grown on the $BaF_2(111)$ substrate.[6] In this Letter, we report the growth of single crystal PbTe thin films on $BaF_2(111)$ substrates by using a solid source molecular beam epitaxy system at the growth temperature varied from 375 to 525° C. The microstructural properties including surface morphology, elemental compositions and crystallinity are characterized. Interesting changes of surface morphology are observed.

The PbTe film was grown on freshly cleaved $BaF_2(111)$ substrates in the growth chamber of a solid source molecular beam epitaxy (SSMBE) system. The background pressure of the growth chamber is 1.5×10^{-10} Torr. High purity (5N) polycrystal PbTe was used as the source material. The $BaF_2(111)$ substrates were prebaked at ~ 200° C for 40 min in a preparation chamber (residual pressure $P = 10^{-7}$ Torr) before transferring into the growth chamber. Before the growth initiation, the substrate was out-gassed for 10 min in the growth chamber at the temperature of 25° C higher than growth temperature, which leads to a clean BaF_2 surface free from H and O contamination. During the deposition, the vacuum of the growth chamber was in the low 10^{-7} Torr. The growth rate, calibrated by measuring the PbTe layer thickness using a TENCOR α-step profiler, was about 0.6 μm/h for all the samples. Optical Nomarski microscope was used to observe the surface defects of PbTe layers. Detailed morphological characterization and statistical analysis of defects for all the samples were performed *ex situ* by atomic force microscopy (AFM) using NT-MDT AFM with semi-contact mode. The composition analysis of the epitaxial layers was performed with energy dispersive x-ray (EDX) spectroscopy. The EDX was carried out with accelerating voltage of 25 kV. The thin film crystallinity was characterized using a high-resolution x-ray diffraction (HRXRD) system, Philips X'Pert Materials Research Diffractometer.

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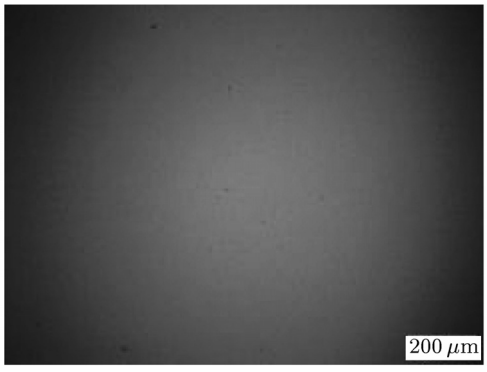


Fig. 1. Nomarski optical micrograph showing the surface morphology of the PbTe layer grown on $BaF_2(111)$ substrate.

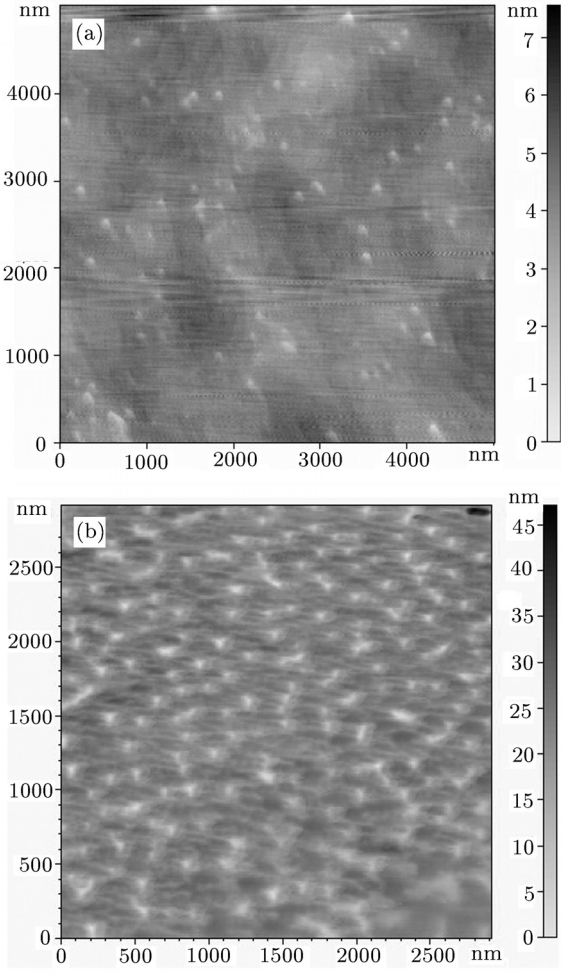


Fig. 2. AFM image shows the morphology of PbTe thin films grown at (a) 375°C and (b) 475°C. The grey scale on the right of the image identifies the derivative of the height.

Figure 1 shows the Nomarski photograph of surface morphology for a $PbTe/BaF_2$ layer grown at the substrate temperature of 475°C. It is seen that the surface is very smooth. No slip lines and cracks caused by the formation of the strain relieving dislocations have been observed, though the lattice mismatch of PbTe with BaF_2 is as large as 4.2%.

Figure 2(a) shows an AFM image of a PbTe epitaxial layer grown at 375°C. The white area that is pictured in shade of the grey presents a terraced surface structure. The black points indicating the derivative of the height tend to distribute around step of the terraced surface. The observed terraced surface structure is interpreted by growth spirals formed around threading dislocations (TD) that originate from the PbTe growth on the 4.2% lattice-mismatched $BaF_2(111)$ substrate.[7] The unfilled holes around the terraced step line are caused by the intersect of several dislocations. In an area of 5 μm × 5 μm, the rms roughness of the PbTe layer grown at 375°C is 0.432 nm, a flatness of atomic layer scale.

Figure 2(b) is an AFM image of the PbTe epitaxial layer grown at 475°C. It is seen that with elevation of growth temperature, a significant increase of the surface roughness occurs. Contrarily to the terraced surface structure of the sample grown at 375°C, the morphology of the PbTe layer grown at 475°C exhibits a rather uniformly distributed array of the triangles. The triangles have an average width of 90–100 nm and an average depth of 5–10 nm. The change of shape from holes to triangles is due to the strain relaxation via the glide dislocations in the main $\{001\}$-$\langle 110\rangle$ systems with the increasing mobility of the misfit dislocations at higher temperature.[8] The rms roughness of the sample grown at 475°C is 3.987 nm for the 3 μm ×3 μm image, almost 10 times higher than that of the sample grown at 375°C. However the rms roughness decreases with the further increasing of growth temperature. For the sample grown at 525°C, the rms roughness of the surface decreases to 0.759 nm. These surface roughness changes can be explained within the mechanisms of the strain-relaxation. In the strain-relaxation process of lead chalcogenide, both elastic relaxation by the formation of the strain-induced coherent island and the plastic relaxation by the formation of the misfit dislocation have been observed.[8] Although the two models of the strain-relaxation will lead to a surface roughening in the strained heteroepitaxial layer, the surface roughness is mainly driven by the formation of the growth islands.[8,9] At the lower substrate temperature, the misfit dislocation density is high because of insufficient high dislocation mobility. When the size and spacing of the islands is larger than the separation of the TDs, the strain-induced coherent islands will be suppressed and a smooth surface is obtained as shown in Fig. 2(a). With the increase of the dislocation mobility at an elevated growth temperature, the TDs density rapidly decreases, which corresponds to the spacing of the TDs increases. When the spacing of the islands is smaller than that of the TDs, the transition of the growth mode from 2D to 3D occurs which leads to a significant surface roughening.

Such a phenomenon is observed in the sample grown at 475 °C. When growth temperature further elevates, adatom surface diffusion lengths increase and reaches the spacing of the TDs, which leads to the decrease of surface roughness. The rms roughness measured from PbTe epilayer grown at 525°C is 0.759 nm that is much smaller than that of the sample grown at 475°C.

In order to determine the composition of the PbTe layer, we measured the energy dispersive x-ray spectra of the sample grown at 475°C. Figure 3 shows an EDX spectrum of the PbTe/BaF_2(111) structure. It can be seen that no emission of characteristic lines for other elements was observed except Pb, Te and Ba, which indicates that only Pb and Te components are deposited in the BaF_2 substrate. The signal of F element detected by EDX is very weak owe to the lower sensitivity to the light element. An approximate quantitative analysis is performed using the EDX software and the ratio of the Te/Pb in the epitaxial layer is 1:1.069, indicating Pb enriched at the higher growth temperature.

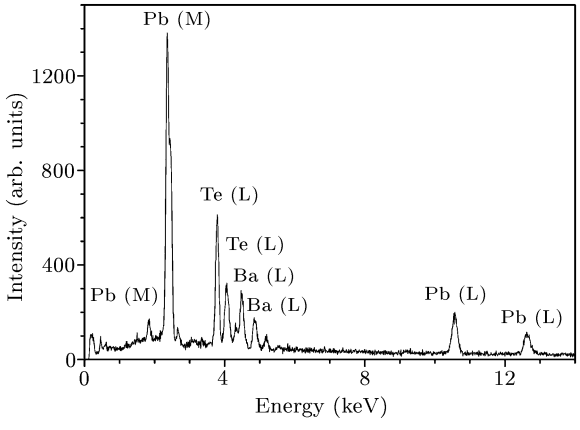


Fig. 3. Energy dispersive x-ray spectrum for the PbTe thin film grown at 475°C.

The x-ray diffraction rocking curves for the PbTe layers on BaF_2(111) grown at different substrate temperatures were measured by HRXRD and the full width at half maximum (FWHM) was obtained. The dependence of FWHM on the growth temperature is shown in Fig. 4. A typical HRXRD rocking curve is shown in the inset of Fig. 4. The FWHM of the PbTe layers varies significantly for different growth temperatures, from 95 to 361 arc seconds. It can be seen that the FWHM tends to decrease when the growth temperature increases from 375°C to 525°C. As shown in the inset of Fig. 4, the FWHM is less than 100 arc seconds for the PbTe grown at 475°C. We also find that FWHM varies slightly with temperature change at higher temperature range, whereas it rapidly increases as growth temperature decreases. This behaviour can be explained from strain relaxation, the growth kinetics and the surface adatom diffusion lengths. With increasing surface diffusion coefficient, adatoms become much more favourable to incorporate into the surface step edges. When the surface diffusion length is comparable to the average step-step distance at higher temperature, a transition of growth mode to step flow growth occurs and the layer is almost fully relaxed.[10]

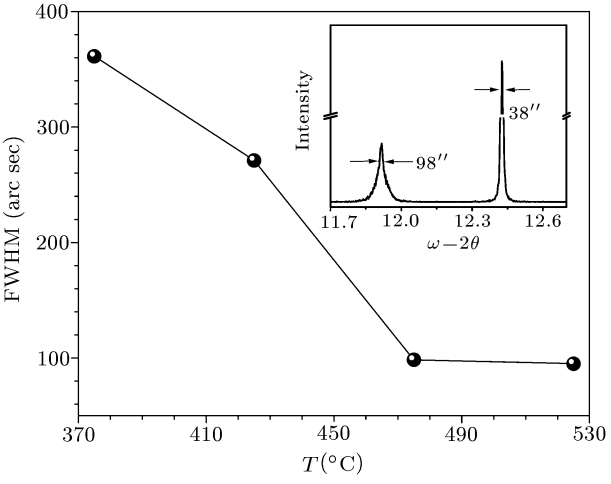


Fig. 4. The dependence of FWHM of (111) diffraction peaks for the PbTe thin films on growth temperature. The inset showing HRXRD spectrum ($\omega-2\theta$) for the PbTe thin film grown at 475°C.

These HRXRD observations suggest that crystallinity of the PbTe epitaxial layer is optimized at the growth temperatures from 475°C to 525°C. We can estimate the dislocation density ρ of the PbTe layer grown at 525°C using equation $\rho = \delta^2/4.35b^2$ in which half width δ is due only to grown-in dislocations,[11] where $\delta = 0.5\times$FWHM (the full width at half maximum of x-ray diffraction rocking curve); $b = a/2\langle 110\rangle$ is the Burgers vector of edge dislocations, where $a = 0.6462$ nm is the lattice constant of PbTe. A dislocation density of $\sim 3 \times 10^8\,\mathrm{cm}^{-2}$ is calculated for the PbTe layer grown at 525°C.

In conclusion, PbTe epitaxial layers have been grown on BaF_2(111) by using solid source molecular beam epitaxy in the growth temperatures range from 375 to 525°C. The evolution of the surface morphology of PbTe has been studied with the increasing growth temperature using AFM. PbTe epilayers exhibit smooth surface morphologies of atomic layer scale roughness and are crack free. It is found that for PbTe film grown at 475°C the morphology is dominated by triangles and the rms roughness is 3.987 nm. Compared to the rms roughnesses of 0.432 nm and 0.759 nm for the samples grown at 375 and 525°C respectively, the surface of PbTe layer grown at 475°C is about 10 times rougher. This roughening transition is attributed to the interaction between the elastic relaxation and the plastic relaxation during the strain relaxation process. The compositional analysis using EDX gives result of 1:1.069 for the ratio of Te/Pb with

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