Large-area epitaxial CdTe(100) films grown on GaAs(100) substrates: MBE growth and substrate temperature effect

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Abstract

The epitaxial growth of high-quality thin films provides a powerful method for the rapid preparation of new materials and discovery of materials with promising properties. Here, we report the effect of substrate temperature on the epitaxial growth of high-quality and large-area epitaxial CdTe films on GaAs(100) by molecular beam epitaxy. RHEED patterns as a function of the substrate temperature showed sharp streaky lines. A CdTe epilayer grown at 320 °C showed the narrowest full-width at half-maximum (FWHM) of 180 arcsec from (004) reflection in X-ray rocking curve measurements. The smooth and flat surface observed in the AFM image showed the excellent uniformity of the thin film, and the surface roughness decreased monotonically with the substrate temperature. From the substrate temperature dependence of the (A^0, X) emission line, the luminescence intensity was found to increase with increasing substrate temperature, while the FWHM decreased with substrate temperature. Our experimental data suggest that the substrate temperature plays an important role in establishing hetero-epitaxial film growth for semiconductor devices.

1 Introduction

II-VI compound semiconductors have attracted considerable attention because of their wide use in optoelectronic devices [1–4]. In particular, CdTe has shown great potential in solar cell and radiation detector applications [5–7]. In solar cell applications, CdTe ensures stable performance, its n-type and p-type form with a band gap of ~ 1.5 eV, which is suitable for solar energy conversion, and it has a high absorption coefficient ($> 10^4$ cm^{-1}) in the visible light region [7]. For radiation detection, CdTe has a high resistivity (~ 10^9 Ω•cm) and fairly high mobility-life product (i.e., $\mu\tau$) that enable high charge collection efficiency and excellent energy

resolution [8]. However, high-quality, large-area CdTe single-crystal growth is very difficult and expensive [9–12]. These factors severely limit the application and development of CdTe in solar cells and radiation detectors. Large-area single-crystalline CdTe epitaxial layers grown on semiconductor substrates are a promising method for solving this problem and are potentially applicable to solar cell development and radiation detectors.

To date, various substrates, such as Si, Ge, InSb, and GaAs, have been used for epitaxial CdTe film growth [13–18]. Among them, GaAs have a large lattice mismatch with CdTe (14.6%), but it has been found to be an appropriate substrate because of the easy of using large GaAs wafers, and well-established detailed GaAs surface reconstruction analysis and clean processes [19–23]. However, even on GaAs substrates, high-quality CdTe films are still difficult to grow because various factors can affect the direction and crystal quality of CdTe films. Deoxidation, the surface stoichiometry of the substrate, and growth temperature in the heat treatment process of the GaAs substrate affect the direction of the CdTe film [24–27]. For the past 40 years, molecular beam epitaxy (MBE) has been one of the most important growth methods for high-quality CdTe single-crystalline

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films [28–30]. In the growth of a high-quality and large-area CdTe epitaxial layer, the substrate temperature is one of the main growth parameters affecting the film properties within a wide expansion [31–36]. However, there is no detailed study on the effect of substrate temperature on the epitaxial growth of large-area CdTe thin films grown on GaAs substrates by molecular beam epitaxy (MBE).

In this article, we report the epitaxial growth of large-area single-crystalline CdTe epitaxial films on a GaAs substrate by MBE. Epitaxial growth and surface configuration were monitored using in situ high-energy electron diffraction (RHEED). We presented the effect of the substrate temperature on the surface structure, crystalline quality, and surface morphology of CdTe epilayers.

2 Experimental procedure

Large-area single-crystalline CdTe epitaxial layer was grown on a GaAs(100) substrate by molecular beam epitaxy (V80H, Oxford Co.) in the temperature range of 200−320 °C. Epi-ready semi-insulating 2-inch GaAs (001) substrates were initially outgassed in the preheating chamber before being loaded into the growth chamber, and thermal annealing at 600 °C was performed for 30 min to remove the remaining impurities and oxidized layer. The base pressure of the growth chamber was maintained at 1×10^{-9} Torr, and elemental Cd (6 N) and Te (7 N) were evaporated using an effusion cell (Effucell Co.). The growth rate of CdTe was confirmed through quartz crystal microbalance (QCM), and the growth rate was approximately 0.5 Å/s. The thickness of the CdTe epitaxial layers was 2 μm. Epitaxial growth and

surface structure were monitored in situ using RHEED. The crystalline quality and structure of the epitaxial films were investigated using $\theta-2\theta$ X-ray diffraction (XRD, Bruker). The surface morphology of the CdTe epilayers was examined by atomic force microscopy (AFM, SII Nano Technology). Photoluminescence (PL) spectra were measured on a Labram HR 800 spectrometer (Horiba Scientific) using a 375 nm excitation source at 10 K.

3 Results and discussion

3.1 Surface structure

Figure 1 shows the RHEED patterns of the CdTe layers grown on 2-inch GaAs substrates as a function of the substrate temperature. To investigate the effect of substrate temperature on the surface structure, changes in the RHEED patterns were focused on pattern shape and surface reconstruction. All the epi-ready GaAs substrates were annealed without the presence of arsenic flux, and a two-dimensional RHEED pattern when the substrate temperature was 500 °C or higher, indicating the desorption of the intrinsic oxide layer. For the samples grown at 200 and 250 °C (Fig. 1c, d), the non-reconstructed GaAs(100) surface was maintained through the low-temperature growth process. On the other hand, samples grown at 280 °C and 320 °C (Fig. 1e, f) showed unique 1/2 ordered striped lines (marked with red arrows) distinguished from the primary [0$\bar{1}$1] [23] azimuth along the [0$\bar{3}$1] direction [18]. As shown in Fig. 1e, f, we confirmed that the surface structure was reconstructed to be the (8×1) morphologies when the substrate temperature was

Fig. 1 RHEED patterns of 2-μm-thick CdTe epitaxial layers grown on GaAs(100) with different substrate temperatures of **a** 2-inch GaAs(100) substrate, **b** GaAs, **c** 200, **d** 250, **e** 280, and **f** 320 °C, measured with the incident electron beam along the GaAs the substrate [0$\bar{1}$1] direction
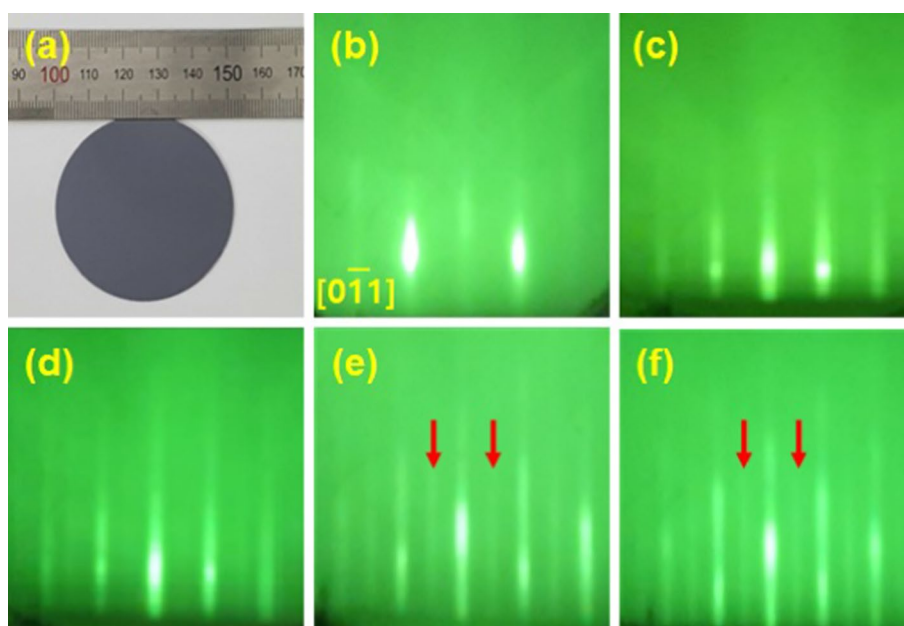
above 300 °C [18, 37, 38]. Furthermore, all the RHEED patterns as a function of the substrate temperature showed sharp streaky lines, indicating the high-quality single-crystalline features of the samples.

3.2 Crystalline structure and quality

Figure 2a shows the θ–2θ XRD patterns of the 2-µm-thick CdTe epitaxial layers as a function of the substrate temperature. All the spectra exhibit similar features, and only sharp (200) and (400) peaks were detected, indicating that the epitaxial layers were completely in the (001) direction [39, 40]. The structural properties of the CdTe single-crystalline layers grown at different substrate temperatures were investigated using XRD measurements. Figure 2b shows the substrate temperature dependence of the lattice constant perpendicular to the film ($a_\perp$) plane calculated from the XRD data of the CdTe layers.

The lattice constants were larger than the known bulk value of 6.482 Å and decreased with increasing growth temperature. This indicates that compressive strain acts in a direction parallel to the film plane and gradually weakens as the substrate temperature increases [32]. In addition, the thickness of the samples used in this study was approximately 2.0 µm, which is much thicker than the few monolayers CdTe critical thickness; the lattice mismatch strain at the interface is considered to be almost relaxed [32]. CdTe epilayers grown on GaAs substrates suffer from biaxial strain due to the lattice constant mismatch and difference in thermal expansion coefficients between CdTe (4.8×10^{-6} K^{-1}) and GaAs (5.7×10^{-6} K^{-1}). Therefore, the thermal strain induced by the difference in the coefficient of thermal expansion between the epilayers and substrates acts as a tensile strain on the CdTe layer; this increases the substrate temperature, resulting in a decrease in the lattice constant [32].
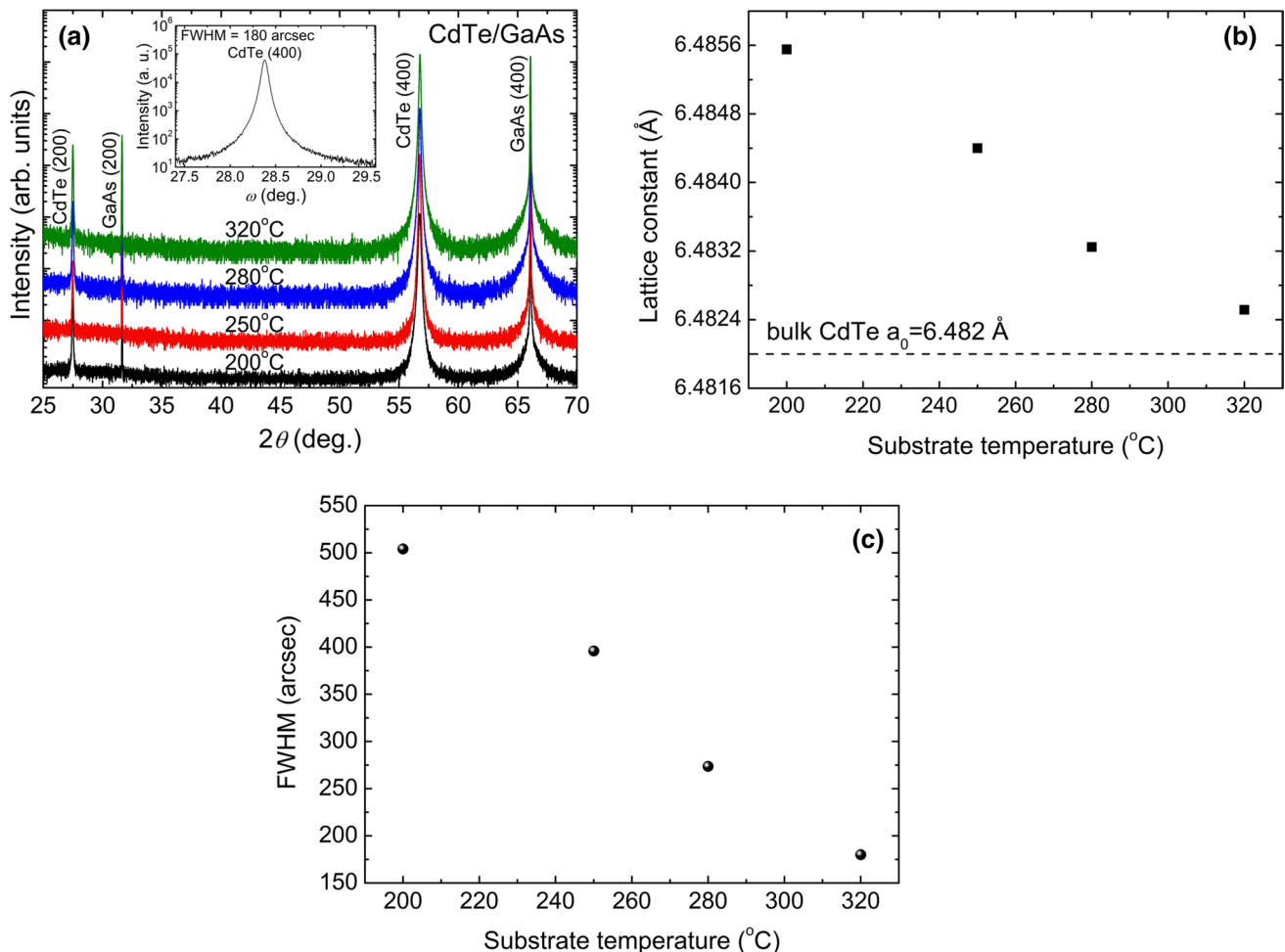

Fig. 2 a θ–2θ XRD spectra of the CdTe epilayer grown on GaAs(100) as a function of the substrate temperature. The inset shows the X-ray rocking curve from the (400) reflection of the CdTe epilayer grown at 320 °C. **b** Lattice constant perpendicular to the plane of the CdTe layers ($a_\perp$) with the substrate temperature. **c** FWHM of the X-ray rocking curves from CdTe(400) reflection with the substrate temperature.

The FWHM values measured from the X-ray rocking curve of the CdTe (400) reflection as a function of the substrate temperature are shown in Fig. 2c. The FWHM is a good indicator of the crystallinity of the CdTe epitaxial layer grown on the GaAs substrate [40, 41]. Crystalline defects, such as threading dislocations, misfit, and twins, in the grown epilayer broaden the diffraction peak and hence increase the FWHM [42]. The decrease in the FWHM with the substrate temperature indicates that the defect density decreases with the distance from the interface and remains low as the substrate temperature increases, resulting in epilayers with high crystalline quality. For the epilayer grown at 320 °C, the narrowest FWHM of 180 arcsec with a thickness of 2 μm is smaller than that of CdTe epilayers grown directly on GaAs (100) by MBE [18, 40] and MOVPE [43, 44], revealing that high crystalline quality CdTe epilayers with low dislocation densities can be achieved.

3.3 Surface morphology

AFM analysis was performed to investigate the effect of the substrate temperature on the surface shape and roughness. Figures 3a–d show the AFM images of the CdTe epilayers as a function of the substrate temperature in a scanning area of 10 μm × 10 μm. As shown in Fig. 3e, the surface roughness decreases monotonically from 2.8 to 1.5 nm with increasing substrate temperature, similar to the FWHM

result. Furthermore, the smooth and flat surface observed in the AFM image indicates that an epitaxial CdTe layer with excellent uniformity was grown and is in good agreement with the results of the RHEED pattern.

3.4 Photoluminescence measurement

Figure 4a shows the 10 K PL spectrum obtained from the epitaxial CdTe layer grown at 200, 250, 280, and 320 °C. The excitonic emission peaks of the CdTe layers were similar to those found in the PL spectra reported by other investigators. The dominant emission peak observed at approximately 1.59 eV is associated with acceptor-bound exciton (A^0, X) [17, 45, 46], and a high-energy shoulder weaker than the (A^0, X) peak was detected at 1.594 eV, which is attributable to donor-bound exciton (D^0, X) [47, 48]. The emission peaks at approximately 1.550 and 1.555 eV are attributed to donor acceptor pair recombination and the second-order longitudinal optical (LO) phonon replica of the free exciton (FE) transition [17, 49, 50], respectively. The weak emission peak observed at approximately 1.534 eV is attributed to the third LO-phonon replica of the FE [49, 50]. Finally, at the low-energy region, a broad emission peak located at approximately 1.495 eV, which can be due to the second LO-phonon replica of the DAP [51, 52]. Meanwhile, because the FWHM of the PL peak is related to defects in the epitaxial layer surface and the strong exciton peak intensity is
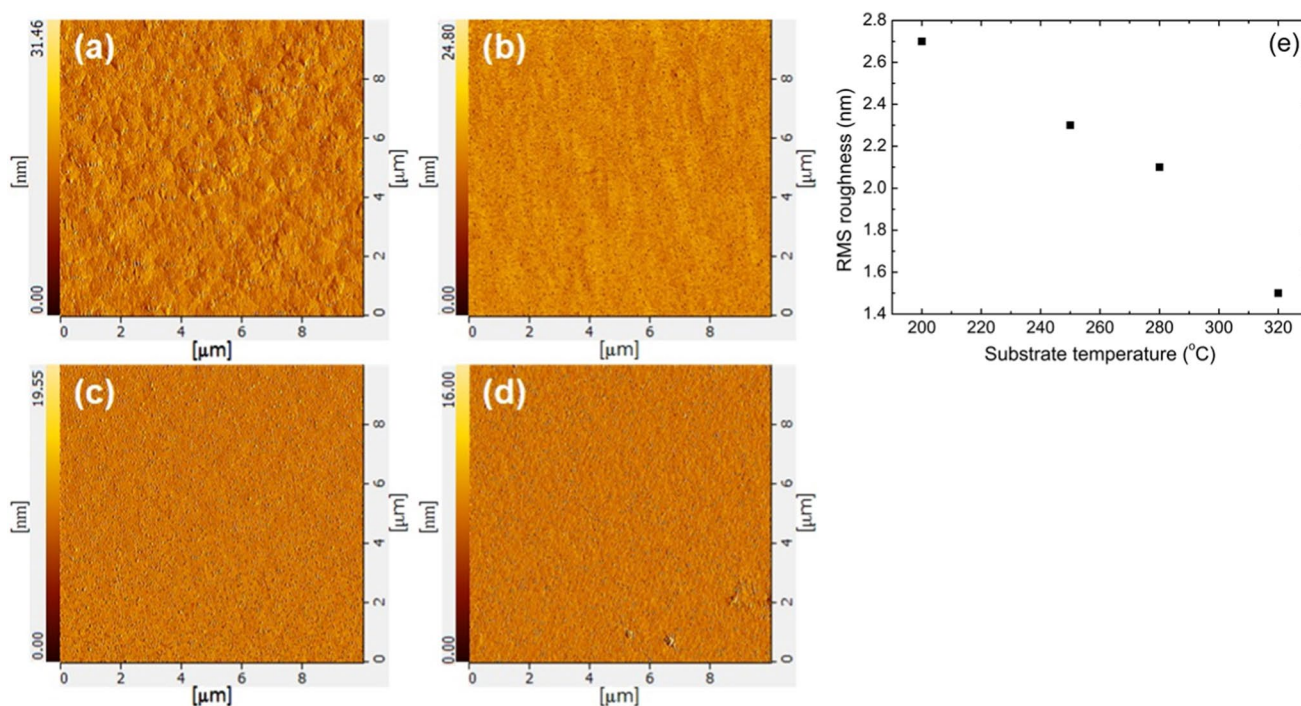

Fig. 3 Atomic force images of the epitaxial CdTe layers grown at **a** 200, **b** 250, **c** 280, and **d** 320 °C. **e** RMS roughness of the CdTe surface as a function of the substrate temperature
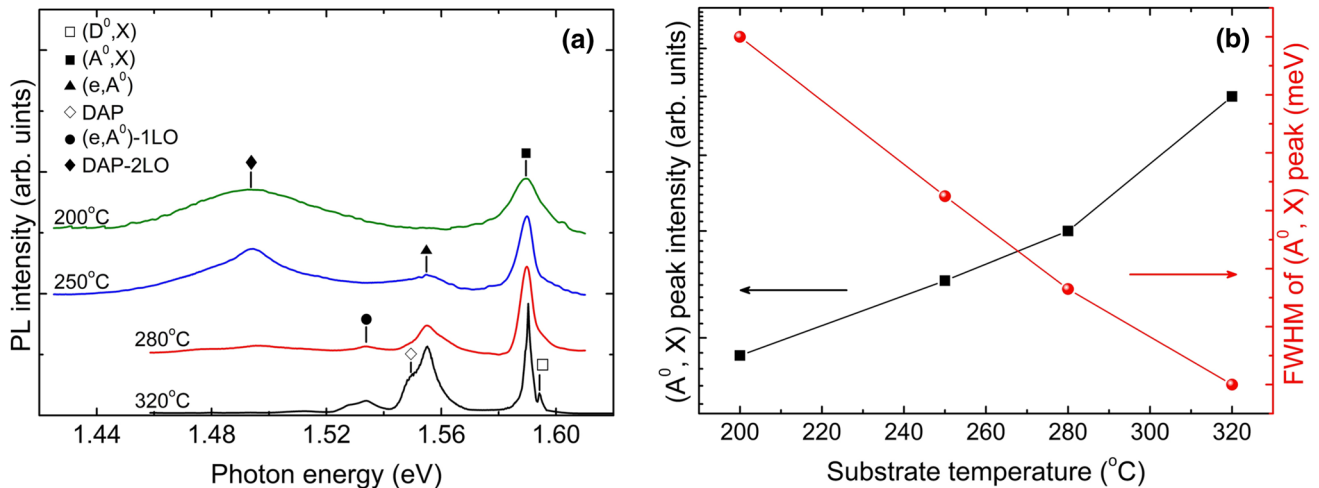
Fig. 4 **a** PL spectra of CdTe epilayers grown on a GaAs substrate as a function of substrate temperature of 200, 250, 280, and 320 ℃ at 10 K. **b** The emission intensity and the FWHM of the (A^0, X) emission lines as a function of substrate temperature

associated with highly ordered crystals, it can be used as an indicator of the crystallinity of the epitaxial layer. Figure 4b shows the (A^0, X) emission peak FWHM and intensity of the CdTe epilayers with the substrate temperature. From the substrate temperature dependence of the bound exciton, the luminescence intensity was found to increase with increasing substrate temperature, while the FWHM decreased with substrate temperature. The CdTe epilayer grown at 320 °C had a minimum value of 2.1 meV, which is similar to the minimum of 2.0 meV observed for CdTe epilayers grown directly on GaAs and InSb substrates using MBE.

4 Conclusion

We investigated the effect of substrate temperature on the high-quality epitaxial growth of single-crystalline CdTe on GaAs using MBE. The effects of the substrate temperature on the surface structure, crystallinity, and morphology of the CdTe epitaxial layers were investigated. All of the RHEED patterns as a function of the substrate temperature showed sharp 2D streaky lines with bright zero-order specular spots, indicating the high-quality single-crystalline features of the samples. The lattice constants perpendicular to the film $(a_\perp)$ plane were larger those of the bulk CdTe and decreased with increasing growth temperature. The AFM surface morphology of the epitaxial layers become smoother and flatter with increasing substrate temperature, and as the substrate temperature increased, the surface roughness decreased monotonically from 2.8 to 1.5 nm. From the substrate temperature dependence of the (A^0, X) exciton emission line, the luminescence intensity was found to increase with increasing substrate temperature, while the FWHM decreased with substrate temperature. Our results showed that tuning the

substrate temperature is an effective method for improving of large-area single-crystalline CdTe epitaxial layer crystallinity.

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