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Heteroepitaxial growth of CdTe on a *p*-Si(111) substrate by pulsed-light-assisted electrodeposition

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Highly oriented CdTe(111) films of high-crystalline quality were grown on a Si(111) substrate by pulsed-light-assisted electrodeposition at room temperature. Strong photoluminescence peaks due to the bound exciton recombination were observed for the electrodeposited CdTe films, confirming the high quality of the films prepared by the present method. Atomic-force microscopy measurements showed an atomically ordered arrangement and demonstrated the epitaxial growth of the CdTe films. © 2002 American Institute of Physics. [DOI: 10.1063/1.1463718]

Heteroepitaxially grown compound semiconductor thin films, e.g., GaAs on Si and CdTe on Si, are very important both technologically and scientifically and have a wide range of applications such as in highly efficient solar cells and semiconductor laser diodes. Thus, there is considerable interest in developing crystal growth methods for heteroepitaxial thin films of compound semiconductors. Various growth techniques, such as molecular-beam epitaxy (MBE),¹ metal-organic chemical-vapor deposition (MOCVD),² and atomic-layer epitaxy (ALE),³ have been developed. However, these methods require very expensive apparatus and reagents, e.g., trimethylgallium, dimethylcadmium, and high temperature (>200 °C). In contrast, the electrodeposition of compound semiconductor thin films can be carried out at low cost and at low temperature, i.e., near room temperature. Unfortunately, however, electrodeposited semiconductor films are polycrystalline and of low quality.⁴ Therefore, these cannot be used for electronic devices. In the early 1990s, Stickney and his colleagues reported that CdTe thin films of high crystallinity could be prepared on a gold single crystal using electrochemical atomic-layer epitaxy (ECALE), an analogue of ALE.⁵ The ECALE method consists of the alternate underpotential deposition (UPD) of two or more elements to form a compound semiconductor. Since their paper was published, many researchers have applied this method for the heteroepitaxial growth of semiconductors such as CdS,⁶ CdTe,⁷ and GaAs (Ref. 8) on a gold single crystal. And Lincot and his colleagues reported the heteroepitaxial growth of a CdTe film done on a InP(111) substrate, which was covered by CdS film, by one-step electrodeposition.⁹ However, it is more attractive and important to grow heteroepitaxial films of compound semiconductors on a Si wafer for practical applications such as optoelectronics are concerned.

CdTe is one of the most important semiconductors and

its thin film has been used for solar cells,¹⁰ near-infrared optical devices,¹¹ and so on. The interest in growing CdTe on a Si substrate is partly motivated by its potential use as an alternative substrate for fabricating Hg_xCd_{1-x}Te infrared detectors. The epitaxial growth of a CdTe film on Si(100) and Si(111) substrates has been achieved using MBE.¹² The Si(111) substrate is more attractive than the Si(100) substrate because it has the same symmetry as epitaxial CdTe(111) and the direct growth of CdTe on Si(111) should be possible without the multidomain problem during the nucleation stage that is observed in the heteroepitaxial growth of CdTe on Si(100).

In this letter, we demonstrate that a CdTe heteroepitaxial thin film can be electrochemically grown on a *p*-Si(111) substrate under pulsed-light illumination using a very simple apparatus.

The present pulsed-light-assisted electrodeposition (PLAE) method consists of the alternate depositions of two elements to form a compound semiconductor, one by the electrochemical deposition and the other by photoelectrochemical deposition, as schematically illustrated in Fig. 1 for the growth of the CdTe layer on *p*-Si. Here, HTeO₃⁻ and the Cd-EDTA complex were used as the Te and Cd sources, respectively. The electrode potential should always be kept at the potential where Te is deposited from HTeO₃⁻ under a diffusion-limited condition, but the Cd-EDTA complex ion is not reduced to metallic Cd. The first step is the deposition of 1 ML of elemental Te in the dark [see Fig. 1(a), (i)]. After completion of the deposition of 1 ML of elemental Te on the *p*-Si substrate, which can be determined by measuring the charge passed, the electrode is illuminated by light, the energy of which is larger than the band-gap energy of Si, for a limited time. As the conduction-band electrons, which are energetic enough to reduce the Cd-EDTA complex to Cd, are generated, a cathodic current due to the Cd deposition flows, in addition to the cathodic current due to the Te depo-

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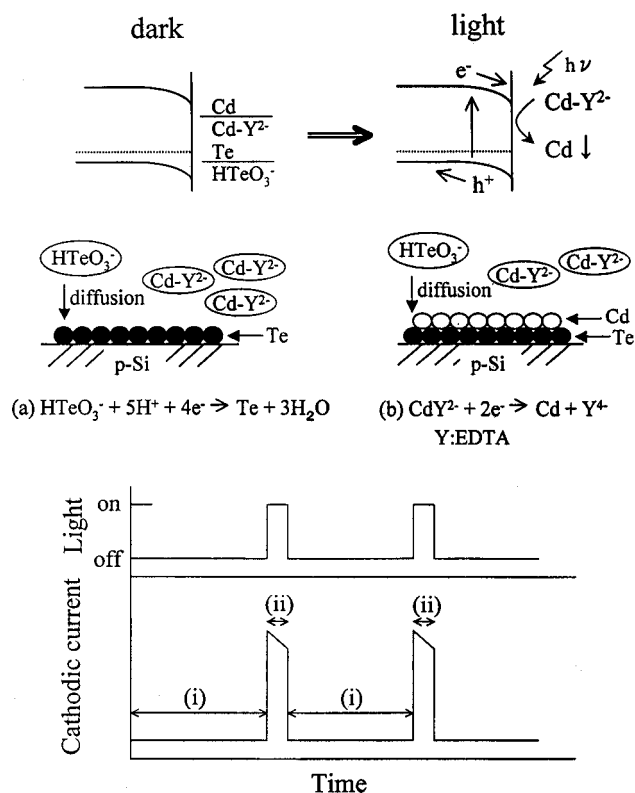


FIG. 1. Schematic representation of the pulsed-light-assisted electrodeposition of CdTe. (a) Deposition of one Te monolayer in the dark. (b) Deposition of one Cd monolayer under illumination.

sition [see Fig. 1(a), (ii)]. The amount of cathodic current is controlled by the light intensity and is generally quite higher than the one for the Te deposition in the dark. Thus, although Te deposition still takes place under illumination, it can be neglected and the Cd deposition preferentially takes place on the deposited Te layer under illumination. The illumination should be stopped after the 1 ML of Cd is deposited, which can be determined by measuring the charge passed. The Te deposition then proceeds on the deposited Cd layer in the dark. These processes should be repeated until the Cd/Te film of the desired thickness is formed. CdTe is expected to be formed by the solid-state reaction.

Deposition of the CdTe film based on the above concept was performed on a p-Si(111) substrate (1.13 cm^2), which was etched by HF solution before use, in aqueous NaOH solutions ($\text{pH}=12 \pm 0.1$) containing 100 mM of the Cd-EDTA ($\text{Cd}-\text{Y}^{2-}$) complex and 0.50–0.10 mM TeO_2 at room temperature. A conventional three-electrode two-compartment cell with an optical window was used for the preparation of the CdTe thin films. A platinum wire and a Ag/AgCl electrode were used as the counter- and reference electrodes, respectively. A potentiostat and a Coulomb meter were used for the potential control and measurement of the charge passed during the electrolysis, respectively. A 300 W Xe lamp with an IR absorbing filter was used as the light source and the pulsed illumination was provided by an optical shutter. The opening and closing of the shutter were controlled by a personal computer, to which the Coulomb meter was connected, based on the charges passed in the dark for Te deposition and under illumination for Cd deposition.

Thus, 1 ML of elemental Te (0.54 mC) was first depos-

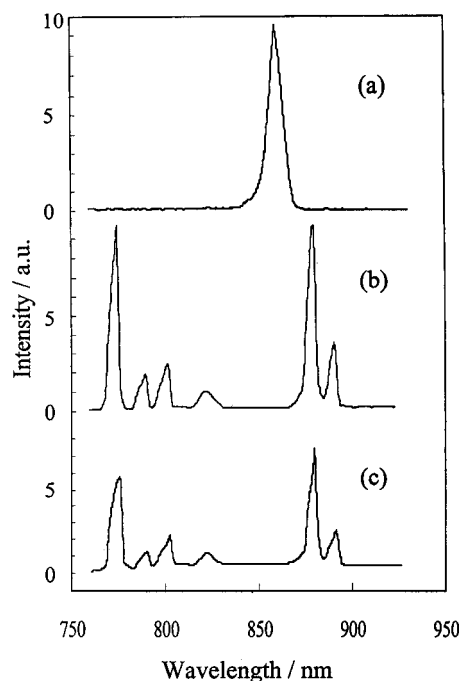


FIG. 2. Photoluminescence spectra of CdTe thin films on p-Si(111) measured at (a) 18 K, (b) 18 K, and (c) 50 K. Deposition conditions: (a) sulfuric acid solution containing 100 mM CdSO_4 and 1 mM TeO_2 . Electrodeposition = -0.45 V vs Ag/AgCl in the dark, (b) and (c) alkaline solution containing the 100 mM Cd-EDTA complex and 0.50 mM TeO_2 . Electrodeposition = -1.10 V vs Ag/AgCl under illumination.

ited on the p-Si electrode in the dark with a typical current of less than $1 \mu\text{A cm}^{-2}$ and then, the shutter was opened so that the p-Si electrode was illuminated and the Cd deposition proceeded with a typical current of $100 \mu\text{A cm}^{-2}$ until 1 ML of elemental Cd (0.27 mC) was deposited. The shutter was kept closed until another monolayer of elemental Te (0.54 mC) was deposited. The CdTe films were grown by repeating these processes. Because the photocurrent was significantly greater than in the dark, the time required for Cd deposition ($\sim 3 \text{ s}$) was much shorter than that for Te deposition ($\sim 10 \text{ min}$).

Figure 2 shows the photoluminescence (PL) spectra of a CdTe film, which was prepared by conventional electrodeposition in the dark in a sulfuric acid solution containing 100 mM CdSO_4 and 1 mM TeO_2 , measured at 18 K (a), and of a 30-ML-thick CdTe film prepared by PLAE observed at 18 K (b) and 50 K (c) by excitation with the 325 nm line of a He-Cd laser (5 mW), with the resolution of 0.5 nm. Only a broad luminescence peak near 860 nm due to the so-called defect luminescence was observed at the CdTe film grown by conventional electrodeposition in the dark [Fig. 2(a)], showing that the CdTe film was polycrystalline with many defects. On the other hand, six peaks were observed at 776, 796, 802, 820, 880, and 890 nm in the PL spectra of the CdTe film prepared by PLAE [Figs. 3(b) and 3(c)]. These peaks were smaller and broader at higher temperatures, suggesting that these PL peaks are due to radiative recombination within the CdTe film. The peak at 776 nm (1.6 eV) is attributed to the bound exciton (BE) recombination or band-to-band transition from the near-band-edge states. The peak energy is slightly greater than the reported value.¹³ This should correspond to a slight increase in the band-gap energy of CdTe

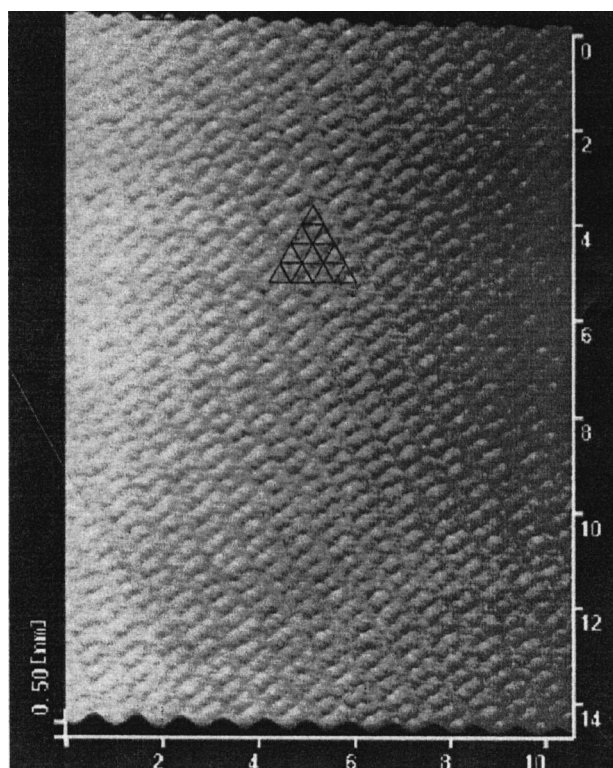


FIG. 3. AFM micrograph of the 20-ML-thick CdTe film deposited on *p*-Si(111). Deposition conditions are the same as those in Fig. 2(b). The inset triangle represents the lattice points of the (111) plane of the zinc-blende CdTe crystal.

caused by the compressive strain because of a larger lattice constant of CdTe (0.648 nm) than Si (0.543 nm). The peak at 790 nm can be assigned to a band-to-shallow acceptor radiative transition.¹⁴ The origin of the shallow acceptor may be an interstitial tellurium atom or sodium ion. The broad peak at 802 nm should be due to the strong donor–acceptor pair transition, and its phonon replica was observed near 820 nm. These PL peaks are known to be observed only when the material is of high quality such as a single crystal.¹⁵ Thus, it is clear that the crystalline quality of the CdTe film grown by PLAE is far better than that of the CdTe films prepared by conventional electrodeposition in the dark, although two peaks corresponding to the so-called defect luminescence at 880 and 890 nm were also observed for the pulsed-light-assisted CdTe film.

The intensity and full width at half maximum (FWHM) of the BE peak are used to evaluate the quality of the epitaxial layers. The FWHM of the BE peak at 776 nm of the CdTe film grown by PLAE under the present condition was 10.3 meV at 18 K, which is greater than the values usually reported for high-quality MOCVD-grown CdTe films, i.e., 5.0–7.5 meV.¹⁵ The ratio of the intensity of the BE peak to that of the defect peak at 880 nm, which is suggested to be an indicator of crystalline quality,¹⁶ is about 1 at 18 K and is greater than that of MOCVD-grown CdTe films on GaAs reported by Kisker.¹⁶ These results show that the quality of the CdTe thin films prepared by PLAE are comparable to or even better than that of conventional MOCVD-grown CdTe films. High-quality layers of CdTe were successfully grown on Si(111) by PLAE.

Figure 3 shows an atomic-force microscope (AFM) image of a 20-ML-thick CdTe film deposited on *p*-Si(111). The surface of the film is very smooth with regularly arranged gourd-shaped spots. These regularly arranged gourd-shaped spots were observed at almost every part of the film surface. The observed spots consisted of two small spots. The lattice points of the (111) plane of the CdTe crystal, which has a zinc-blende structure, are shown by the vertices of the triangle in Fig. 3. They are in good agreement with the positions of the gourd-shaped spots, and this fact implies that the CdTe films are oriented along the direction of the (111) plane. In the x-ray diffraction patterns of this film and a 50-ML-thick CdTe film, two diffraction peaks were observed at $2\theta=23.7$ and 28.4 . The peak at $2\theta=28.4$ is due to Si(111), and the broad peak at $2\theta=23.7$ is due to CdTe(111). Thus, these results and the fact mentioned in Fig. 2 confirmed that the highly oriented CdTe films of high quality can be grown on *p*-Si(111) by PLAE.

In conclusion, we have demonstrated that high-quality and highly oriented CdTe films can be grown on Si(111) by the pulsed-light-assisted electrodeposition technique. Although more studies are needed in order to obtain a high-quality epitaxial layer, this method should provide the way to extend the well-advanced silicon technology to optoelectronics applications.

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