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기판온도에 따른 PbTe 박막의 구조 및 전기적 물성

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Structure and Electrical Properties of PbTe Thin Film According To The Substrate Temperature

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요 약

Pulsed laser deposition법에 의하여 양질의 PbTe 박막을 다양한 기판온도에 따라 성장시켰다. XRD패턴으로부터 각 온도에서의 PbTe층들은 결정화가 되어있음을 알 수 있었다. 또한 PbTe 박막의 XRD 피크들은 (h00)의 방향성을 나타내고 있다. Pb의 재증발로 인하여 400℃ 이상에서는 PbTe 박막은 결정성의 박막으로 형성되지 않았다. AFM 이미지로부터 박막의 표면은 작은 granular 결정들과 평탄한 매트릭스로 구성되어 있음이 관찰되었다. 기판온도의 증가에 따라 표면의 입자들이 커지는 것을 알 수 있었다. Hall-effect 측정으로부터 300℃에서 성장한 PbTe 박막의 전기적 특성은 3.68 x $10^{18}cm^{3}$의 캐리어 농도와 148 cm^2/Vs 의 Hall 이동도를 나타내었다.

Abstract

PbTe thin films of high quality were deposited on HF-treated Si(100) substrates at various substrate temperature by pulsed laser deposition technique. XRD patterns showed that PbTe layers were well-crystallized to a cubic phase with (h00) preferred orientation with the substrate temperature up to 300℃. PbTe films could not form at substrate temperature above 400℃ because of reevaporation of the Pb. According to AFM image, the surface of films was composed of small granular crystals and flat matrix. According to the increase of substrate temperature, the grain size at film surface becomes larger. By Hall-effect measurement, the carrier concentration and Hall mobility of n-type PbTe films grown by T_{sub}=300℃ were 3.68 x $10^{18}cm^{3}$ and 148 cm^2/Vs, respectively.

I. Introduction

The pulsed laser deposition(PLD) method has been proven as a suitable method for the growth of highly crystalline inorganic thin film[1]. By now, PLD method has been applied to many materials, including semiconductor, metal, and dielectric material.

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The chalcogenide compounds based on the IV-VI semiconductor groups are found to have interesting electrical and magnetic characteristics[2-4]. Moreover, the materials have an advantage in the preparation process compared with the metal oxides. The growth process of metal oxide needs the oxidizing gas but the transition metal chalcogenide does not, which is an important factor because the oxidizing gas may damage the surface of thin film. The preparation of other transition metal chalcogenide films of sulfides and selenides such as NiS, NbS_2, $NbSe_2$, and MoS_2 using a pulsed laser ablation was previously reported[5,6].

Lead chalcogenide systems have considerable scientific interest because of their potential for device applications. These materials are narrow IV-VI semiconductors with NaCl structures. PbTe has been extensively investigated in the bulk crystal form because of its thermoelectric and infrared detection properties. Among lead chalcogenide compounds, PbTe despite of semiconductor is prepared crystallized thin film at room temperature. But the crystallinine structure of PbTe thin film obtained at the various substrate temperatures is not fully investigated. The high conductivity is necessary for device application. Thus, we investigated the relation of crystallinity and conductivity. In this letter, we reported the structure and electric properties of PbTe thin film at a various substrate temperature. The film characterizations were performed by x-ray diffraction(XRD), atomic force microscopy(AFM), and Hall-effect measurements.

II. Experimental

The PbTe thin films were prepared by 193 nm ArF excimer laser deposition in vacuum ($1x10^{-6}$ Torr) onto a Si(100) substrate. Figure 1 shows a schematic representation of the apparatus for pulsed laser ablation used in this experimental. The substrate was pretreated with HF prior to film deposition. The pulsed laser with a repetition rate of 3-5 Hz was focused onto the target with an energy density of about 1.5 J/cm^2. The distance between the target and the substrate was 4 cm. The films were deposited on the substrates positioned opposite against the target in the vacuum chamber. The film were deposited at various substrate temperature(T_{sub}) from room temperature to 400 ℃ during the formation of films. The growth rate was approximately 1-2 $nmsec^{-1}$ and the film thickness was about 100 nm. The film growth was monitored with an oscillating quartz-crystal thickness monitor in-site. The achieved film thickness was measured with an optical microscope ex-site. The crystal structure and crystallographic orientation of the films were identified with an X-ray diffractometer(XRD) at CuKα radiation. The surface morphology and roughness of the film was studied by atomic force microscopy(AFM). The electrical properties of the films were determined by Hall-effect measurements using Van der Pauw technique at room temperature, in which the dc magnetic field strength were done at 3KG. Aluminum films were evaporated on to the PbTe films under high vacuum for ohmic contact.

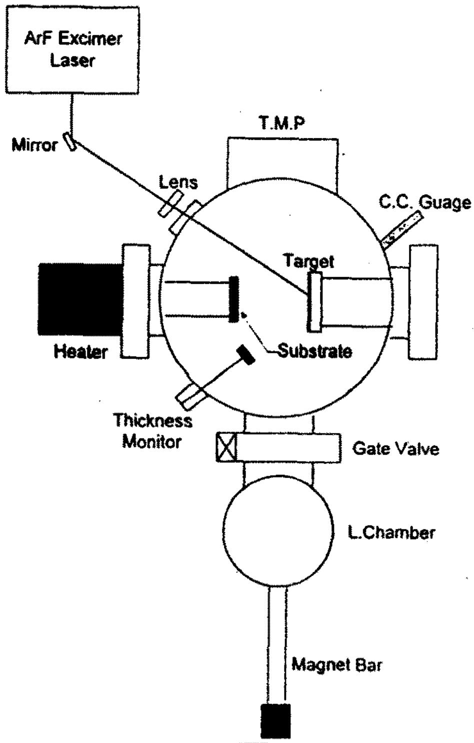


Fig. 1. Schematic diagram of pulsed laser deposition system.

III. Results

Figure 2 shows the XRD patterns of the PbTe film prepared at various substrate temperatures(T_{sub}) from room temperature to 300℃. The XRD patterns showed that PbTe films of each T_{sub} appeared to be crystallized as illustrated in figure 2(a), (b) and (c). And as indicated by the strong (h00) peaks in all films of this figure, the PbTe films of each T_{sub} become a-axis oriented. PbTe films could not be prepared at $T_{sub.}$ above 400℃, probably due to the reevaporation of Pb. Usually, TiO_2 films are prepared at a substrate temperature of 1000℃, while stoichiometric $PbTiO_3$ film grown at 450℃[7]. In this case, PbO was formed by oxidizing Pb, and $PbTiO_3$ film grown at significantly higher temperature than PbTe films. Therefore, it is reasonable to think that PbTe films can be obtained at low growth temperatures because of the low melting temperature(m.p. 330℃) of Pb and non-oxidizing atomsphere.

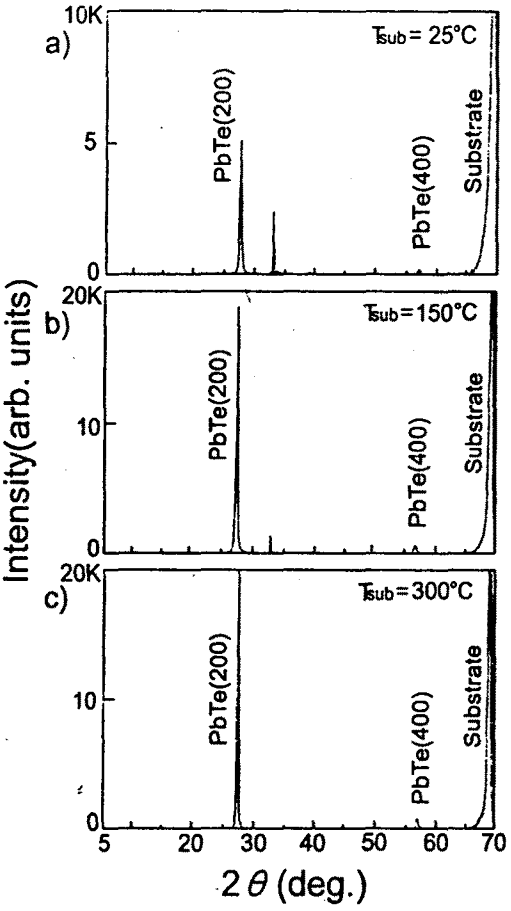


Fig. 2. X-ray diffraction pattern of PbTe film on Si(100) at substrate temperature of (a)25℃, (b)150℃, (c)300℃.

The crystallinity of the PbTe films was estimated by the intensity of (200) x-ray diffraction peak shown in Figure 2, and FWHM of (200) diffraction profile on $2\theta-\theta$ as shown in Figure 3. In Fig.2, the maximum intensities were 5000, 18000 and 25000 cps with $T_{sub.}$ of 25, 150 and 300℃, respectively; the intensity of (200) peak increase with the increase $T_{sub.}$ The rocking curve full width at half-maximum(FWHM) were 0.236°, 0.169°, 0.161° at the substrate temperatures. The FWHM decreases with increasing $T_{sub.}$ for ~0.3° of FWHM, it is known that grains are well aligned with their a-axes normal to the substrate. Consequently, it was concluded that PbTe thin film of high crystallinity can be obtained at T_{sub} =300℃.

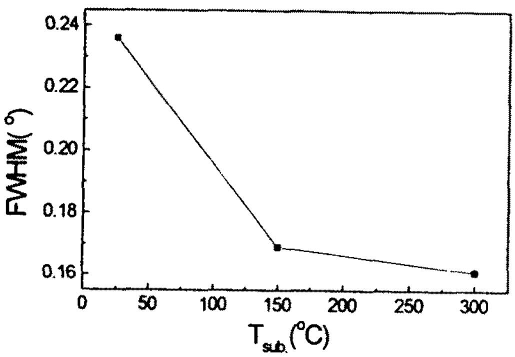


Fig. 3. The rocking curve full width at half-maximum (FWHM) and XRD peak intensity for the (200) peak of PbTe film on Si at each substrate temperature.

Figure 4 shows AFM images of PbTe films whose corresponding XRD patterns are shown in Figure 2. The film grown at $T_{sub.}$=25℃ has smooth surface with the roughness of 2Å. The film grown at $T_{sub.}$=150℃ is composed of small round grains dispersed in flat matrix. The particles size is about 0.5㎛. When the $T_{sub.}$ increased above 300℃, the grains size became larger. The film has wedge-shape grains whose typical size is approximately 1x2㎛, as shown in Figure 4(c). Thus, an increase of the grain size can be achieved by increasing of the substrate temperature. In general, the larger grain size of thin films would lead to the improvement in the electronic properties.

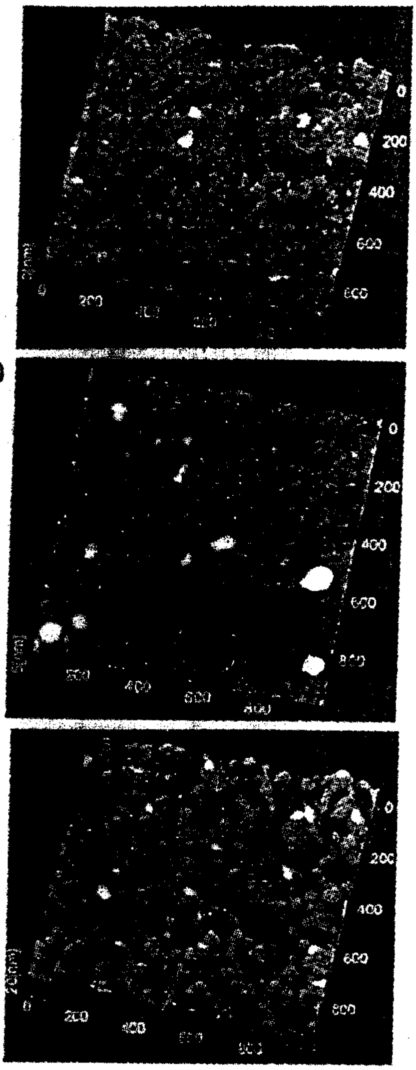
(a)
(b)
(c)


Fig. 4. AFM images(scan area 10×10㎛) of PbTe film on Si substrate at temperature of (a)25℃, (b)150℃, (c)300℃.

The electrical properties of the PbTe films obtained at various $T_{sub.}$ by Hall-effect measurement were summarized as shown in Table I. From the result of measurement, we can conclude that PbTe thin film is an n-type semiconductor. PbTe films have the carrier concentration of 7.06×10^{16}, 3.68×10^{18}, and 2.76×10^{19} cm^{-3}, whose corresponding microstructures are XRD pattern shown in Figure 2(a), 2(b) and 2(c), respectively. The corresponding Hall mobilities are 59, 100, 148 cm^2/Vs, respectively. The low mobility of the film grown at $T_{sub.}$=25℃ are probably caused by the small grain size of the surface. The increase of mobility as a function of substrate temperature is a commonly observed characteristic for semiconductor films due to the grain boundary scattering of the charge carriers. The conductivity(1.12 $\Omega^{-1}cm^{-1}$) of PbTe film prepared at T_{sub}=300℃ was larger than those at T_{sub}=25 and 150 ℃(0.87x10^{-2} and 0.26x10^{-3} $\Omega^{-1}cm^{-1}$, respectively). The carrier concentration, Hall mobility and conductivity of PbTe thin film were increased according to the increase in T_{sub}. It is evident that the electrical properties of these films are strongly influenced by the film growth temperature.

Table. I. Electrical properties of PbTe films on Al_2O_3 substrate at each substrate temperature.

T_{sub} (℃)	film thickness (㎛)	conductivity ($\Omega^{-1}cm^{-1}$)	carrier concentration (cm^{-3})	mobility (cm^2/vs)
25	0.5	1.12	7.06×10^{16}	59
150	0.5	0.87×10^{2}	3.68×10^{18}	100
300	0.5	0.26×10^{3}	2.76×10^{19}	148

Ⅳ. Conclusion

The PbTe thin film on HF-treated Si(100) were grown by ArF excimer laser ablation. XRD pattern of PbTe films at various substrate temperatures revealed cubic PbTe with a strongly preferred (h00) orientation. Semiconductive PbTe thin film was obtained even at substrate temperature of room temperature. In this film, AFM images showed that the surface was composed of small granular crystal and flat matrix with a roughness of 2 Å. As $T_{sub.}$ increases, the grain size increases and the morphology changes to wedge-shape. The electrical properties of PbTe films grown at various substrate temperature were strongly influenced by the film growth temperature; the carrier concentration, Hall mobility and conductivity of PbTe thin films were coincidently increased with increasing T_{sub}.

Acknowledgements

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