Journal of Alloys and Compounds 509 (2011) 3155–3159

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Electrical and optical characteristics of Au/PbS/n-6H–SiC structures prepared by electrodeposition of PbS thin film on n-type 6H–SiC substrate

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ARTICLE INFO

Article history:
Received 2 September 2010
Received in revised form 1 December 2010
Accepted 3 December 2010
Available online 13 December 2010

Keywords:
Silicon carbide
Schottky barrier modification
XRD
PbS

ABSTRACT

To realize Schottky barrier height (SBH) modification in the Au/n-6H–SiC Schottky diodes, lead sulfide (PbS) thin films were grown on n-6H–SiC by electrodeposition method. At first, XRD experiments were performed to investigate the crystal structure of the PbS film electrodeposited on n-6H–SiC. It has been deduced from the diffraction profile that the PbS thin film has a crystal structure more strongly oriented along the [2 0 0] direction. An optical energy band gap value of 1.42 eV for the PbS film was obtained from its optical absorption spectra. Then, we have prepared Au/PbS/n-6H–SiC Schottky barrier diodes (SBDs) with interface layer and reference Au/n-6H–SiC/Ni SBDs. The SBH enhancement has been succeeded by the PbS interlayer, influencing the space charge region of the SiC. The SBH values of 1.03 and 0.97 eV for the samples with and without the interfacial PbS layer were obtained from the forward bias current–voltage (*I–V*) characteristics. The SBH increase in the Au/PbS/n-6H–SiC SBD with the interfacial PbS layer has been attributed to the fact that the interface states contain a net negative interface charge in metal/n-type semiconductor contact due to the presence of the interfacial PbS layer.

1. Introduction

Metal–silicon carbide (SiC) contacts play very important roles in many high-performance devices in optoelectronic, high-temperature, high-frequency, and power applications [1–3]. Schottky barrier height (SBH) in metal/semiconductor (MS) is a fundamental parameter for the rectifying contacts [4–10]. The presence of another semiconductor thin film with nanometer thickness at the MS interface in the Schottky contacts introduces a new method in the control of fundamental device parameters [10–15]. Thereby, the presence of an interfacial layer at the MS structures plays an important role in the determination of the characteristic parameters of the devices [14–18]. Meanwhile, the SBH modification is one of the essential conditions for the device development. Many attempts have been performed to realize the modification of the SBH or the continuous control of the SBH using an interfacial layer or a chemical passivation procedure at certain MS interfaces [18–24]. Much of these activities have been focused on understanding and controlling the parameters such as the interface potential barriers [14–22]. For example, Güllü and Türüt [10] have prepared Al/phenosulfonphthalein (=PSP)/semiconductor junctions by coating of an organic film to p-Si substrate, and they [10] have shown that the junctions exhibit diode-like characteristics and they have achieved the modification of the SBH of Al/p-Si diode by the PSP organic material. Aydogan et al. [11] have fabricated Au/n-type ZnO/n-Si diodes with rectifying properties by the electrochemical deposition technique and they have found a value of 0.59 eV for this structure although the SBH value for the conventional Au/n-Si diodes is 0.82 eV. Mangal et al. [14] have reported fabrication of GaAs based metal–insulator–semiconductor solar cell with polyaniline, using a polymeric material as an insulator, and they [14] have measured the photovoltaic response of Al/polyaniline/GaAs diode. Furthermore, Roberts and Evans [25] have achieved the control of the interfacial potential barrier for metal/n-GaAs diodes using thin interlayer of the organic semiconductor, tin phthalocyanine (SnPc).

In this paper, ultra thin films of lead sulfide (PbS) have been electrodeposited on either a transparent indium tin oxide (ITO) coated quartz or n-6H–SiC substrates. The PbS thin films grown on ITO electrodes have been used for both determining the band gap value and estimating the thickness of PbS by means of the optical absorption measurements. Furthermore, the PbS thin films on the n-6H–SiC have been grown to form Au/PbS/6H–nSiC/Ni Schottky diodes, where the Au–SiC interface has been modified by a thin layer of the PbS. Electrochemical deposition is a useful technique to obtain highly oriented semiconductive thin films and is an alternative to conventional deposition methods because it is economic, operates at ambient temperatures and pressure, and provides film

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doi:10.1016/j.jallcom.2010.12.028

thickness control. The current–voltage (I–V) characteristics of the fabricated Au/PbS/6H–nSiC/Ni and reference Au/6H–nSiC/Ni SBDs have been measured to experimentally examine whether the thin PbS layer can be successfully used to vary effective SBH. The XRD measurements of the samples have been carried out to determine the crystal structure.

2. Experimental procedures

6H–nSiC wafer with axis orientation and 300 ± 25 μm thickness was used in this study. The n-SiC wafer was chemically cleaned using the RCA cleaning procedure (i.e. 10 min boil in $NH_3 + H_2O_2 + 6H_2O$ followed by a 10 min $HCl + H_2O_2 + 6H_2O$ at 60 °C) before making contacts. The ohmic contact was made by evaporating Ni on the back surface of the SiC substrate, then, it was annealed at 950 °C for 5 min in N_2 atmosphere.

The PbS thin film was electrochemically deposited on the front surface of the SiC substrate with the ohmic contact. The PbS electrodeposition was carried out according to method given in Ref. [26]. This method is based on the underpotential deposition (UPD) of either Pb or S at the same time from the saturated solution of PbS containing excess of PbS as a source of Pb^{2+} and S^{2-} at elevated temperatures. In this method, a highly oriented structure of PbS is supposed to form on the electrode. Electrodeposition procedure was achieved in 0.1 M acetate buffer solution (pH 4.5) including saturated lead sulfide. Cell temperature was fixed at 90 °C by using a thermostat system. The preparation of lead sulfide was achieved by addition of 0.01 M Na_2S ($Na_2S{\cdot}9H_2O$, Reagent grade, Aldrich) into 0.01 M Pb^{2+} ($Pb(CH_3COO)_2{\cdot}3H_2O$, Merck) solution with rapid stirring while nitrogen is purged from the solution. All of the electrolyte solutions were prepared in a Milli-Q ultra pure water (conductivity ≤5.5 $\mu S\,m^{-1}$). The solutions were deoxygenated by passing dry argon through the electrochemical cell for 30 min prior to each experiment. The cyclic voltammetry and potential-controlled electrolysis experiments were performed with a Gamry potentioscan system connected to a three-electrode cell. Indium tin oxide (ITO) coated quartz (10 $\Omega\,cm^{-2}$) and SiC substrates were used as working electrode. An Ag/AgCl (3 M NaCl) (Bioanalytical Systems) electrode served as reference electrode, and a Pt wire electrode was used as counter electrode. The PbS/SiC substrate with the ohmic contact was inserted into vacuum system to form the Au/PbS/6H–nSiC/Ni structures. The Schottky contacts on the PbS layer have been formed by evaporating Au as dots with diameter of about 1.0 mm (the diode area = 7.85×10^{-3} cm^2). The thickness of Au was approximately 50 Å, and a hard mask prior to evaporating to deposit gold film was used. Thus, the reference Au/6H–nSiC/Ni SBD was also formed. The current–voltage (I–V) characteristics of these structures were measured using a HP 4140B picoampermeter at 300 K and in the dark.

The powder X-ray diffractograms (XRD) of prepared samples were recorded using a Rigaku Miniflex powder X-ray diffractometer with Cu Kα radiation (λ = 1.5405 Å). Optical absorption spectra of PbS thin films on the ITO-coated quartz substrates were acquired by using a Shimadzu UV-3101 UV–vis–NIR spectrometer at room temperature.

3. Results and discussion

3.1. The optical absorption and XRD measurements

Fig. 1 presents the vis–NIR optical absorption spectra of PbS film on ITO-coated quartz electrode. PbS film was prepared after 5 min of electrodeposition at −550 mV constant potential from the saturated solution of PbS containing excess of PbS in 0.1 M acetate buffer solution (pH 4.5) at the cell temperature of 90 °C. Electrodeposition potential of −550 mV was determined by using cyclic voltammetry experiment according to UPD of both Pb and S on ITO-coated quartz electrode.

The optical band gap energy of the PbS can be obtained by the following relation [26–29]

$$\alpha h\nu = A(h\nu - E_g)^m, \quad (1)$$

where α is the linear absorption coefficient, ν is the frequency, h is Planck's constant, and m depends on the nature of optical transition. Specifically, m is 1/2, 3/2, 2, and 3 for transitions directly allowed, directly forbidden, indirectly allowed and indirectly forbidden, respectively. A is constant involving the properties of the bands and E_g is defined as the optical energy band gap between the valence band and the conduction band.

The direct energy gap, E_g can be obtained from plot of $(\alpha h\nu)^2$ versus $h\nu$ (Fig. 2). A direct energy gap value of 1.42 eV for the PbS film was obtained by extrapolating the linear portion of the $(\alpha h\nu)^2$ versus $h\nu$ plot in Fig. 2. Alanyalıoglu et al. [26] have reported that the band gap of PbS increases from 0.41 to 1.85 eV with decreasing film thickness from 30.5 to 1.1 nm due to quantum confinement effect. Similar results have also been observed for CdS [30,31] and Sb_2Te_3 [32] and ZnS thin films [33]. On the basis of reference [26], it can be estimated that the thickness of the PbS film is less than 10 nm.

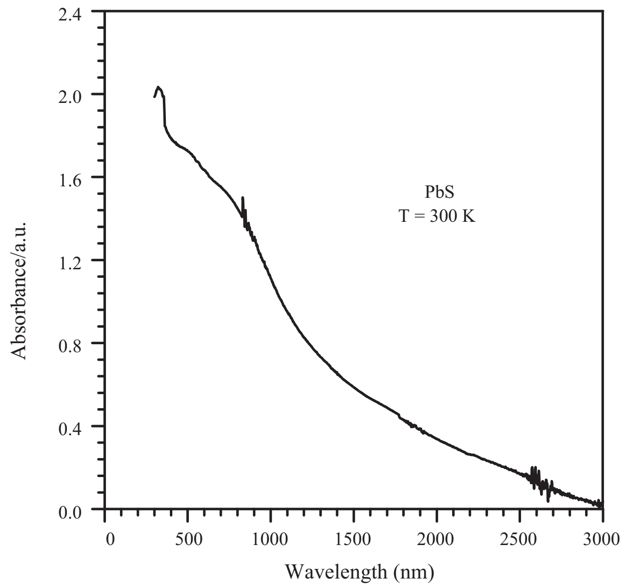

Fig. 1. The optical absorption spectra of PbS film on ITO-coated quartz substrate at room temperature.

We have attempted to prepare the Au/PbS/n-6H–SiC structure by electrodeposition of PbS on SiC electrode from 0.1 M acetate buffer solution (pH 4.5) by applying −550 mV constant potential for 5 min at the cell temperature of 90 °C. Fig. 3 shows the XRD pattern of prepared Au/PbS/n-6H–SiC structure. Interplanar spacing values (d) and corresponding ($h\,k\,l$) planes of the Au/PbS/n-6H–SiC structure are also presented in Table 1. From the diffraction profiles, it is obvious that two peaks are observed for the PbS film. A strong peak

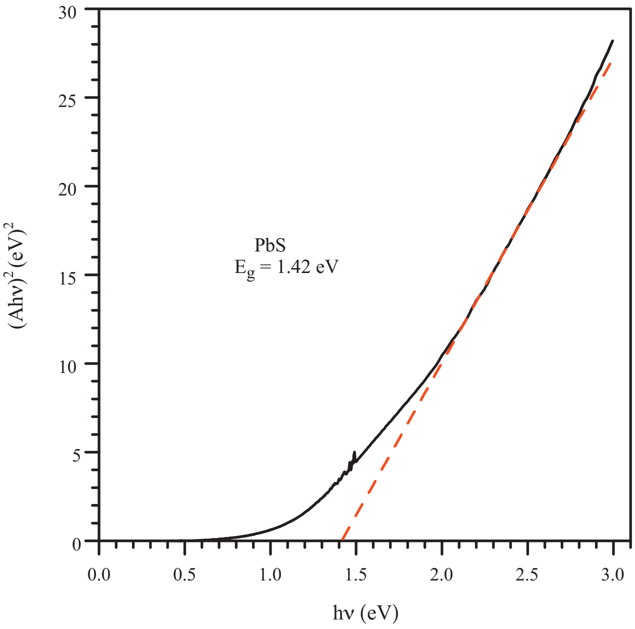

Fig. 2. $(Ah\nu)^2$ versus $h\nu$ plot for direct band gap of the PbS film.

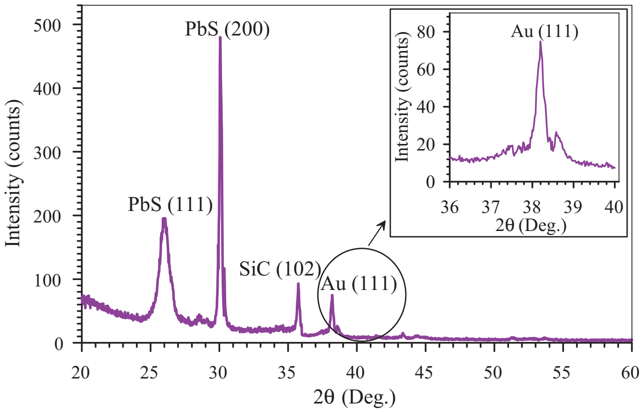

Fig. 3. The XRD plot of the Au/PbS/n-6H–SiC structure with interfacial layer.

at $2\theta = 30.06^\circ$ corresponds to (2 0 0) crystal face and a weak peak at $2\theta = 25.96^\circ$ corresponds to (1 1 1) crystal structure. It is clear that PbS has the preferential orientation along cubic (2 0 0) plane when the intensities of these two peaks are compared with each other. Oznuluer et al. [34] have prepared PbS thin films by using electrodeposition technique and they have indicated that the formation of PbS film follows a transition from a thermodynamically controlled orientation to a kinetically preferred orientation of (2 0 0) direction on the electrode after a critical thickness. In our best knowledge (1 1 1) direction of PbS in the X-ray diffractogram takes place due to electrodeposition of ultra thin film of PbS that is estimated to be less than 10 nm from optical studies. Moreover, observation of a peak of (1 1 1) direction other than cubic (2 0 0) direction in the X-ray diffractogram of the PbS may also be due to ultra thin film of PbS. We believe that PbS film structure is independent of the surface structure after a critical thickness and we expect a uniform PbS film structure on SiC substrate. Furthermore, PbS thin films on polycrystalline Au and single crystal Au(1 1 1), and ITO electrode have been studied by us using morphological techniques. It has been observed that the PbS film grows in highly uniform cubic structure on all of these electrodes after a critical thickness [29]. Gaiduk et al. [35] and Stancu et al. [36] have reported that the morphology and the thickness of the PbS layers strongly depend on the chemical nature of substrate studied.

The XRD peak at $2\theta = 35.74^\circ$ is attributed to SiC(1 0 2) structure [37]. Puiso et al. [38] and Choudhury and Sarma [39] have prepared PbS thin films on Si(1 0 0) and Si(1 1 1) single crystal substrates by successive ionic layer adsorption and reaction (SILAR) and electrodeposition techniques, respectively. They [37–39] have obtained the similar diffraction peaks of SiC in the XRD results. The peak at $2\theta = 38.3^\circ$ associates with (1 1 1) direction of Au contact [26]. The inset of Fig. 3 shows the X-ray diffractogram of gold for a comparison.

In addition to explanations above, it may be explained why the XRD peak intensities of SiC and Au are much less than that of the PbS peak (2 0 0) though the thickness of SiC and Au films are much higher. The XRD peak intensities of SiC and Au can be much less than that of PbS peak (2 0 0) because the SiC layer is under the PbS thin film, and the thickness of Au is approximately 50 Å which is less than the thickness of 10 nm of the PbS thin film.

Table 1
Summary of Fig. 3.

Au/PbS/n-6H–SiC	2θ (°) (measured)	d (Å) standard	d (Å) measured	h k l
PbS	25.96	3.429	3.429	1 1 1
	30.06	2.969	2.970	2 0 0
SiC	35.74	2.516	2.510	1 0 2
Au	38.20	2.355	2.355	1 1 1

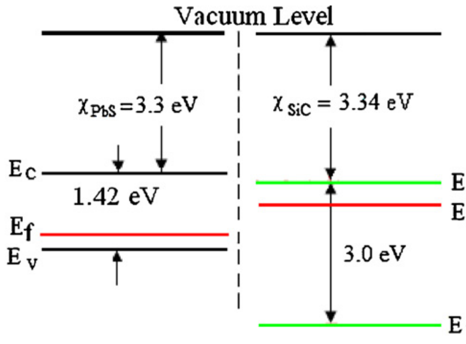

Fig. 4. The energy-band diagram of the p-PbS and n-6H–SiC before contact.

3.2. Current and voltage characteristics

According to the thermionic emission (TE) theory, the current expression in Schottky barrier diodes (SBDs) can be given by [40–42]

$$I = AA^*T^2 \exp\left(-\frac{\Phi_b}{kT}\right)\left[\exp\left(\frac{qV}{nkT}\right) - 1\right], \tag{2}$$

where

$$I_0 = AA^*T^2 \exp\left(-\frac{\Phi_b}{kT}\right) \tag{3}$$

is called the saturation current, A is the effective area of diode, A^* is the effective Richardson constant and 156 A cm^{-2} K^{-2} for n-type 6H–SiC semiconductor [1], T is the temperature in Kelvin, q is the elementary charge, k is Boltzmann's constant, n is the ideality factor that is unity for an ideal diode and it is determined from the slope of the linear region of the forward bias ln I–V characteristic by means of the relation

$$n = \frac{q}{kT}\frac{dV}{d(InI)} \tag{4}$$

However, n has usually a value greater than unity. High values of n can be ascribed to the presence of an interfacial thin layer between metal–semiconductor, and therefore, to the bias-dependent of the BH [16,19]. Φ_b is the zero-bias BH in eV. The BH Φ_b can be obtained by extrapolation of the forward or reverse bias current–voltage curve to zero applied voltage by the following equation:

$$\Phi_b = kTIn\left(\frac{AA^*T^2}{I_0}\right) \tag{5}$$

Fig. 4 shows the energy-band diagram of the p-PbS and n-6H–SiC before contact. PbS has a bulk electron affinity of about 3.3 eV [43,44]. The effective mass of electrons and holes of PbS is the same and PbS is a p-type semiconductor, the Fermi level is expected to lie above the valence band [44]. Fig. 5 shows the forward and reverse bias I–V characteristics of the reference Au/n-type 6H–SiC and electrochemically prepared Au/PbS/n-6H–SiC/Ni structures at 300 K. By means of Eqs. (4) and (5), the values of the ideality factor and Φ_b for the reference Au/n-type 6H–SiC were calculated as 1.07 and 0.97 eV, respectively. The values of the ideality factor and Φ_b for the Au/PbS/n-6H–SiC were also calculated as 1.37 and 1.03 eV, respectively. An ideality factor value of 1.37 can be originated from the presence of the PbS layer between Au and SiC layers [40–42,44–49]. The increase of 60 meV in the BH value may be ascribed to the presence of the PbS interlayer that modifies the effective BH by influencing the space charge region of the SiC [40–42,44–49]. As

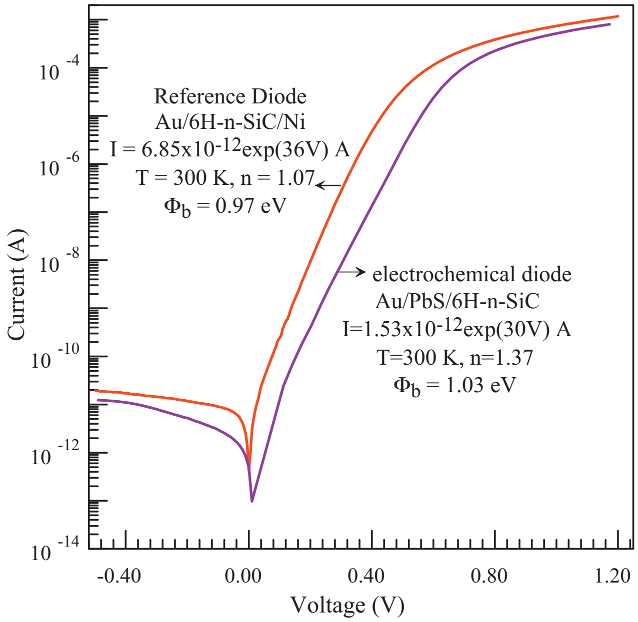

Fig. 5. The forward and reverse bias current–voltage characteristics of reference Au/6H-n-SiC and Au/PbS/6H-n-SiC structures.

well-known, the interfacial layer in MS contacts plays an important role in the determination of the BH of the devices, and it can be a sensitive probe useful in establishing processes for minimizing surface states, surface damage and contamination. Thus, this process may ultimately increase the quality of devices fabricated using the semiconductor. Thereby, it is known that the PbS film forms a physical barrier between the metal and SiC substrate, it protects the SiC surface from the metal [40–42,45].

As mentioned in Refs. [40,46,47], the semiconductor surface in the MS contacts can be affected by the deposition of the contact metal. That is, due to the noninvasive nature of the PbS/SiC contacts or to a relatively undisturbed semiconductor surface at the PbS/SiC contacts compared to the M/SiC contact obtained by the direct deposition of the metal on the SiC substrate, the energy dependence of the interface state densities may prove useful in determining the processes involved in SBH formation and metal/insulator/semiconductor diode surface properties. Therefore, it can be said that directly deposition of metals on the semiconductors can generate large numbers of the interface states at the semiconductor surface that strongly influence the properties of the resulting Schottky barrier, and thus the deposition of the nonreactive materials onto the semiconductor substrate does not result in the same drastic modification of the semiconductor surface. Thus, it can be said that the PbS layer causes a significant modification of interface states. As can be seen, the barrier height may be tuned by using thin films with nanometre thickness. That is, by means of the choice of the interlayer thickness, the device can be designed to exhibit the desired properties [50].

We can explain as follows that the PbS interlayer modifies the effective BH by influencing the space charge region of the SiC [40–42,45]. It has been indicated above that the BH values of 0.97 eV for the reference sample (Au/n-6H–SiC) and 1.03 eV for the Au/PbS/n-6H–SiC sample have been obtained. It has been stated that the increase of 60 meV for the Au/PbS/n-6H–SiC with respect to the value of 0.97 eV for the reference sample may be ascribed to the presence of the PbS interlayer which modifies the effective BH by influencing the space charge region of the SiC. In metal/n-type semiconductor contacts, in the absence of the interface states, the negative charge Q_m on the metal surface must be equal the positive space charge Q_{sc} because the junction is electrically neutral. In the presence of the interface states, the neutrality condition becomes $Q_m + Q_{sc} + Q_{ss} = 0$ [40–42,45]. The occupancy of the interface states is determined by the Fermi level which affects the change of the space charge Q_{sc} in the depletion layer of the semiconductor, and thus the modification of the SBH in the diode with the PbS interlayer. The difference of 60 mV between SBH of both diodes indicates the presence of a negative interfacial charge Q_{ss} in the PbS/n-6H–SiC interface [40–42,45]. In such a case, the positive space charge Q_{sc} must be greater than if the interface state charges were absent [40–42,45]. This means that the SBH will increase due to the increase in the negative interface state charges in metal/n-type semiconductor contacts. Thus, we may assume that the SiC surface has surface states and the deposition of PbS leads to new bond formation and interface states between PbS and SiC.

4. Conclusions

We have successfully prepared Au/PbS/n-6H–SiC Schottky diodes by using electrodeposition technique. Thickness of the PbS film has been predicted as less than 10 nm by regarding to reference [26]. The BH values of 1.03 and 0.97 eV for the Schottky structures with and without the interfacial PbS layer have been obtained from the forward bias *I–V* characteristics, respectively. Thus, the BH modification has been succeeded using the PbS thin layer. The difference between the BH values of both structures has been ascribed to the PbS interlayer to modify the effective BH by changing the space charge of the SiC. It has been supposed that the space charge has changed due to the fact that the interface states have a net negative charge in the Au/PbS/n-SiC interface.

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