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Electrical transport properties of polycrystalline SnO_2 thin films

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ABSTRACT

We study the electrical transport properties of degenerate tin dioxide thin films with thickness larger than 250 nm. Our samples have a rutile structure and the effective mass of electron is 0.31 m_0, which is obtained from the variation in optical bandgap. The temperature dependence of the Hall mobility indicates that the ionized impurity scattering is the dominant elastic scattering mechanism for electrons. In the low temperature range, the clear negative magnetoresistivity is observed, which can be attributed to the three-dimensional weak localization (WL) effect. By applying the three-dimensional WL theory, we extracted the electron dephasing length, which decreased on increasing temperature. Unexpectedly, the temperature dependence of the extracted dephasing length is found to be dominated by the electron-electron scattering in the small-energy-transfer process and the electron-phonon scattering has negligible contribution. This can be attributed to the low electron concentration in our samples.

1. Introduction

Due to the coexistence of high conduction and optical transparency, metal oxides such as In_2O_3, ZnO, CdO, and SnO_2 have been widely studied for many years [1,2]. Among them, SnO_2 with a wide bandgap of 3.6 eV has attracted renewed attention recently because of its potential applications for gas sensor [3], ultraviolet sensor [4,5], and high mobility thin-film transistor [6]. In these applications, the fundamental electrical property of this material is crucial for designing the devices [7]. Experimentally, it is found that different growth conditions can significantly change its electrical property, which may be related to the different concentration of donors such as tin interstitials and/or oxygen vacancies [8,9]. Until now, SnO_2 thin film has been reported to be insulator or degenerate semiconductor by different groups. In insulating thin films, various electrical transport mechanisms were found, which included thermal activation [10–12], hopping transport [7,11], and tunneling transport [13]. In degenerate SnO_2 thin films, meanwhile, negative magnetoresistivity (MR) was observed at low temperature near zero magnetic field, which was attributed to the two-dimensional weak localization (WL) effect [14,15].

This effect is caused by the constructive interference of two phase-coherent electronic waves propagating in opposite directions along a closed trajectory [16]. From the WL effect, one can experimentally determine the electron dephasing length (L_φ) which is a critical quantity for developing quantum-interference nanodevices [17,18]. When L_φ value is smaller than the film thickness, the two-dimensional WL effect should appear. On the contrary, when L_φ value is larger than the film thickness, the three-dimensional WL effect is expected to occur. However, there has been no report about the negative MR originating from the three-dimensional WL effect in unintentionally doped SnO_2 films until now. In this paper, we study the electrical transport properties of polycrystalline SnO_2 thin films with thickness larger than 250 nm. At low temperature, the negative MR is observed, which can be attributed to the three-dimensional WL effect. The obtained L_φ shows a decreasing on increasing temperature, which can be described by the electron-electron scattering.

2. Experiments

SnO_2 samples were grown on glass substrates by atmospheric-pressure chemical vapor deposition, and the detailed processes are similar with that reported in Ref. [19]. One sample, which we refer to as sample A, has a thickness of ~270 nm and the other sample with the thickness of ~480 nm is referred to sample B. Crystal structure was determined by X-ray diffraction (XRD)

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(Bruker-AXS, D8 Advance) with Cu Kα radiation at room temperature. The optical transmittance of the films was measured using a Perkin Elmer Lambda 800/900 UV/visible spectrometer. The surface morphology of the films was examined by scanning electron microscopy (SEM) (JSM-5610LV). The temperature and magnetic field dependence of the resistivity was measured in the Van-der-Pauw geometry under perpendicular magnetic fields.

3. Results and discussion

Fig. 1 shows XRD spectra for both our samples. The grown films are polycrystalline with six peaks of the rutile structure. From the positions of these peaks, one can obtain the lattice constant. The obtained lattice constants a and c are 0.476 (0.477) and 0.319 (0.317) nm for sample A (sample B), which are consistent with the reported values [10,20]. Fig. 2 shows the UV/visible transmission spectra of our samples. Evidently, these films all exhibit an optical transparency of 70–80% in the visible range. From the transmission spectra, one can obtain the optical absorption coefficient by using the relation $\alpha = -(\ln T_{tr})/d$ (here, T_{tr} is the transmittance rate and d is the thickness of the thin film). The obtained α is related to the photon energy in a direct-bandgap material by the expression [21] $\alpha h\nu = A(h\nu - E_g)^{1/2}$, where $h\nu$ is the incident photon energy, A is a constant, and E_g is the bandgap. Inset of Fig. 2 plots the $(\alpha h\nu)^2$ against $h\nu$, in which one can determine E_g from the tangent (blue lines) to the absorption edge. The bandgaps determined by this way are 3.80 eV and 3.76 eV for sample A and sample B, respectively. These values are comparable to the reported results [6,22].

It is noteworthy that the bandgap value in sample A is wider than in sample B, indicating that the bandgap widening occurs. This is usually observed in degenerate semiconductor, which results from the Burstein-Moss (BM) effect [23]. The bandgap widening due to this effect can be calculated as

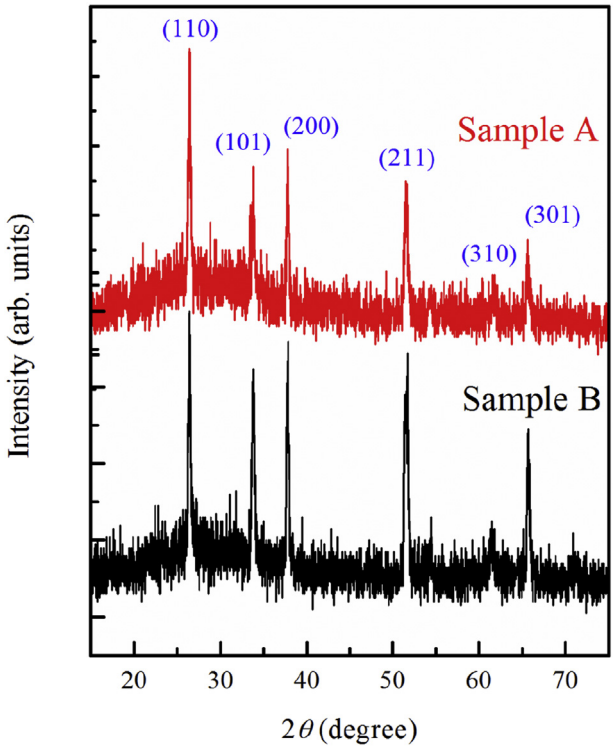

Fig. 1. XRD patterns for two SnO_2 films grown on glass.

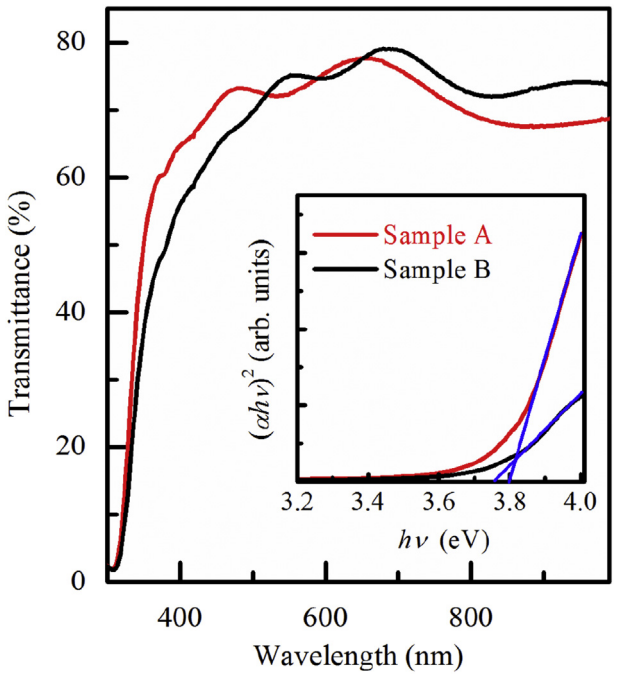

Fig. 2. Room temperature optical transmission spectra of the SnO_2 films. Inset shows $(\alpha h\nu)^2$ as a function of photon energy $h\nu$, where α, h, and ν are respectively absorption coefficient, Planck constant, and photon frequency. The energy gap was obtained by extrapolating the linear part.

$$\Delta E_g^{BM} = \frac{\hbar}{2m_{eh}^*}\left(3\pi^2 n\right)^{2/3}, \tag{1}$$

where n is the carrier concentration, and $m_{eh}^* = 1/(1/m_e^* + 1/m_h^*)$ with m_e^* and m_h^* representing the effective mass of electron and hole, respectively [24]. Meanwhile, the bandgap narrowing due to the electron-electron scattering and the electron-impurity scattering cannot be neglected in SnO_2 films [25], which is given by

$$\Delta E_g^N = \hbar\nu_v - \hbar\nu_c, \tag{2}$$

where $\hbar\nu_v$ and $\hbar\nu_c$ are the self-energies in the valence and conduction bands. Then, the measured bandgap shift can be expressed by

$$\Delta E_g = \Delta E_g^{BM} - \Delta E_g^N. \tag{3}$$

Our samples are degenerate semiconductors (see discussed below). The Hall effect measurements indicate that our samples are n-type and the electron concentrations are 2.4×10^{26} and 1.8×10^{26} m^{-3} at 300 K, respectively. The smaller value of the electron concentration in the thicker sample suggests that the donor concentration decreases with increasing the thickness of film. Similar result is also reported in SnO_2 film prepared by the magnetron sputtering [26]. Substituting these values and ΔE_g (0.04 eV) into Eq. (3) with $m_h^* = 1.0m_0$ (m_0 is the mass of free electron) and $\Delta E_g^N = 0.065$ eV [25], we find that $m_e^* = 0.31m_0$, which is in line with the reported values [25,27].

Fig. 3(a) shows the temperature dependence of resistivity for both samples. As seen in the figure, both samples exhibit a negative temperature coefficient over the whole temperature range of 13–300 K, indicating a semiconductor behavior. However, we note that the resistivity ratio of $\rho(13\,K)/\rho(300\,K)$ is smaller than 1.4, hinting that our samples may be degenerate semiconductors.