Depression of positive magneto-conductance due to anti-weak localization effect in annealed In$_2$O$_3$-ZnO thick films

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Abstract. We investigated the magneto-conductivity $\Delta$ in three dimensional indium zinc oxide films with different resistivity $\rho$ prepared by postannealing in air. The weak localization theory was fitted to data of $\Delta$ at temperatures below 50K by the use of suitable inelastic scattering time $\tau$ and spin-orbit(S-O) scattering time $\tau_s$. We found the $\rho$ dependences of both times $\tau$ and $\tau_s$ in a range $1.5\times10^{-4}\Omega^{\rho} < \rho < 4\times10^{-4}\Omega^{\rho}$. As $\rho$ increases, the ratio $\tau_s/\tau$ increases from $\approx 0.005$ to $\approx 0.5$ and the $\Delta$ at low temperatures changes from positive to negative values. We suggest a picture that the annealing in air brings the change of the S-O scattering from light to heavy atoms, namely, oxygen to indium and/or zinc atoms.

1. Introduction

Transparent materials exhibiting high electrical conductivity have been extensively investigated [1-3]. Recently, there is a great deal of interest in indium zinc oxide films [4]. From the viewpoint of the electrical transport properties at low temperatures in the low carrier systems, indium zinc oxide films are suitable material to investigate the quantum effects of the electron weak localization (WL) [5]. Regarding the magneto-conductivity $\Delta$, the $\Delta$ takes negative or positive values depending on the strength of the ratio $\tau_s(T)/\tau_o$ [6-9]. The conditions $\tau_s(T)/\tau_o <<$ and $\tau_s(T)/\tau_o >>$ give a positive $\Delta$ and a negative $\Delta$, respectively. Because the value of $\tau_s$ depends on not only the dirtiness of the specimens but also the atomic number $Z$ as $\tau_o \propto Z^{-\lambda}$, where $\tau$ is the elastic scattering time [10], it is expected that we can obtain a useful information about the S-O scattering atom from $\Delta$. 

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2. Experimental

Amorphous (In$_2$O$_3$)$_{1-x}$(ZnO)$_x$ films with 350nm thickness were prepared by deposition on glass substrates with use of the DC-magnetron sputtering. To obtain the films with different $\rho$, we annealed amorphous films in air for one hour on a hot plate preheated to several annealing temperatures. In Table 1, the typical characteristics are summarized.

Table 1. Characteristics of indium zinc oxide film at 2.0K. The values of $\rho$ and $n$ in parentheses are data at 300K.

| No | $\tau_{a}$ (deg) | $\rho$ in $\Omega$ m | $n$ in m$^{-3}$ | $L_{m}^{2}$ in m$^{2}$ | $(\tau_{a}/\tau_{m})$ |
|----|------------------|---------------------|-----------------|---------------------|---------------------|
| 1  | 25               | $3.7 \times 10^{4}$ | $5.8 \times 10^{10}$ | $4.0 \times 10^{12}$ | $0.005$             |
| 2  | 250              | $4.3 \times 10^{4}$ | $1.4 \times 10^{10}$ | $1.0 \times 10^{13}$ | $0.5$               |
| 3  | 275              | $1.3 \times 10^{4}$ | $(5.9 \times 10^{10})$ | $1.0 \times 10^{14}$ | $0.51$              |
| 4  | 280              | $1.0 \times 10^{3}$ | $(5.9 \times 10^{10})$ | $3.6 \times 10^{16}$ | $0.37$              |

3. Results and discussions

To understand low-temperature transport properties due to the WL effect, we measured $\rho(H)$ at various temperatures. We define $\Delta$ as $\Delta = \Delta(H) = \langle \rho(H) \rangle - \langle \rho(0) \rangle$. The theoretical expression $\Delta$ for three dimensional (3D) systems is given as follows [8,11],

$$
\Delta = e^{2} / (2\pi \hbar) f \left[ \left( 1/4 \tau_{m} + 1/\tau_{c} \right) (h / DeH) \right] / 2 - \left[ (h/4\tau_{m}DeH) / 2 \sqrt{eH/\hbar} \right],
$$

where $f(x) \approx \left[ \sqrt{2 + x} - \sqrt{x} \right] - \left[ (1/2 + x)^{-1} + 3/2 + x \right]^{-1} + 2.03 + x$ and $D$ is the diffusion constant.

Figure 1(a) shows the $\Delta(H)$ for the film No.1 at 2.0K $\leq T \leq$ 0K. The solid lines are calculated from above equation, regarding $L_{m}^{2}(T)$ = $3\tau_{m}(T)$ as a fitting parameter, where $D\tau_{m}$ is assumed to be independent of temperature. It is clearly shown that the theory can well describe the data in the wide ranges of $H$ and $T$. We can consider this film is a typical weak localization specimen, taking account of the small value of the ratio of $\tau_{a}(2K)/\tau_{m} = 5 \times 10^{-5}$.

Figure 1(b) shows the $\Delta(H)$ of the film No.4. The solid lines also show theoretical ones. The temperature dependence of $\Delta$ is completely different from that of the film No.1. At low temperatures, the values of $\Delta$ are negative. Increasing temperature, $\Delta$ changes from negative to positive values. This means that the localization condition $\tau_{m} \ll \tau_{a}$ is not sufficiently satisfied at low temperatures. When temperature increases above 10K, the magnitude of $\Delta$ decreases keeping the positive values. These behaviors seem to give a sign that the localization turns to the anti-localization in annealed films. In the inset, data $L_{m}^{2}(T) = 3\tau_{m}(T)$ obtained by fitting procedure are shown. The value of $\tau_{m}$ monotonically decreases with increase of temperature. Theoretically, $\tau_{m}(T)$ for impure 3D systems is given as follows [12], $\tau_{m}(T) \approx \left[ 3/4 (E_{F})^{1/2} \hbar (k_{F} / K_{B} T)^{1/2} \right]$. However, it seems that the experimental data is given as $\tau_{m}(T) \propto T^{-1}$ shown by the solid line. For the reason of a discrepancy between the theory and experimental data, we can consider a additional inelastic scattering mechanism independent of temperature as discussed by Y.W Hsu et al. [13].

Figure 2 shows the $1/\rho$ dependence of characteristics of $L_{m}^{2}(2.0K)$ and $L_{m}^{2}$. Although both values essentially decrease with decrease of $1/\rho$, the $L_{m}^{2}$ strongly changes than $L_{m}^{2}$. This indicates
that heat treatment in air depresses the localization effect and brings the anti-localization effect in the electrical conductive oxide materials.

**Figure 1.** $\Delta \tau (H) = \rho (H) - \rho (0)$.

The solid lines are calculated to fit data. (a) Data for the film No.1 at several temperatures. (b) Data for the film No.4. The inset shows the $D\tau$, for the film No.4. The solid line shows $D\tau \propto T^{-1}$.

**Figure 2.** $1/\rho$ dependence of $L_{in}^2 = \tau_{in}$ and $L_{so}^2 = \tau_{so}$. Solid and dotted lines are calculated from the free electron model for $L_{in}^2$ and $L_{so}^2$, respectively. The inset shows the $\rho \cdot n$ plots. Solid line shows the relation of $\rho \propto n$.

We show a brief discussion on the dirtiness dependence of $L_{in}^2 = \tau_{in}$ and $L_{so}^2 = \tau_{so}$. To find the $1/\rho$ dependence of both characteristics, we use some relations derived from the assumption of the free electron model, i) $\nu \propto r \propto (1/n)^{1/3}$, ii) $\rho = (1/n)\nu (1/\ell) \propto n^{-3/2} (1/\ell)$ and iii) $D \propto (1/\ell)$. Using these relations, we obtain $\rho$ dependence $L_{in}^2 = D\tau_n \propto n^{-2} \rho^{-1/2}$ and $L_{so}^2 = D\tau_s \propto n^{-3} \rho^{-1/3}$. From the experimental result between $n$ and $\rho$ shown in the inset in Fig.2, we can express approximately $n \propto \rho^{-1/2}$ as shown by the solid line. Using this relation, we obtain two characteristics as a function of $\rho$ as $L_{in}^2 \propto \rho^{-4}$ and $L_{so}^2 \propto \rho^{-3}$ which are shown by solid and dotted line, respectively. As for the $1/\rho$ dependence of $L_{in}^2$, it seems that the present model can well explain the experimental result. However, the data of $L_{so}^2$ show the stronger $1/\rho$ dependence than that expected from the model.

In the above simple model for the $1/\rho$ dependence of $L_{so}^2$, we have not argued the atomic number $Z$. The discrepancy between the model and the experimental result suggests that it is necessary to take into account of $Z$, namely $S-O$ scattering atoms. The positive $\Delta$ due to the electron localization in the film No.1 means that the $S-O$ scattering comes from light atoms. In the oxide materials, the conductive electrons are generally understood to originate from the oxygen vacancies. It is reasonable to consider that vacancies may also act as the $S-O$ scattering to show the localization effect with positive $\Delta$. When the film is annealed at high temperatures, these oxygen vacancies are annihilated to decrease the carriers. Therefore, we believe that the number of the light
atom decreases due to the annihilation of oxygen vacancies. As a result, the contribution of heavy-
atom from excess indium and/or zinc relatively increases to shorten the S-O scattering time $\tau_s$.

4. Conclusions

We observed change of the sign of $\Delta$ from negative to positive ones for high-resistivity indium zinc oxide films. From the analysis of $\Delta$ by the weak localization theory, we found that the ratio $\tau_s/\tau_o$ increases with decrease of $1/\rho$. We consider that the strong depression of length of $\tau_s$, comparable to $\tau_s$, in films with high resistivity originates from the change of spin-orbit scattering atoms from the oxygen to indium and/or zinc ones.

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