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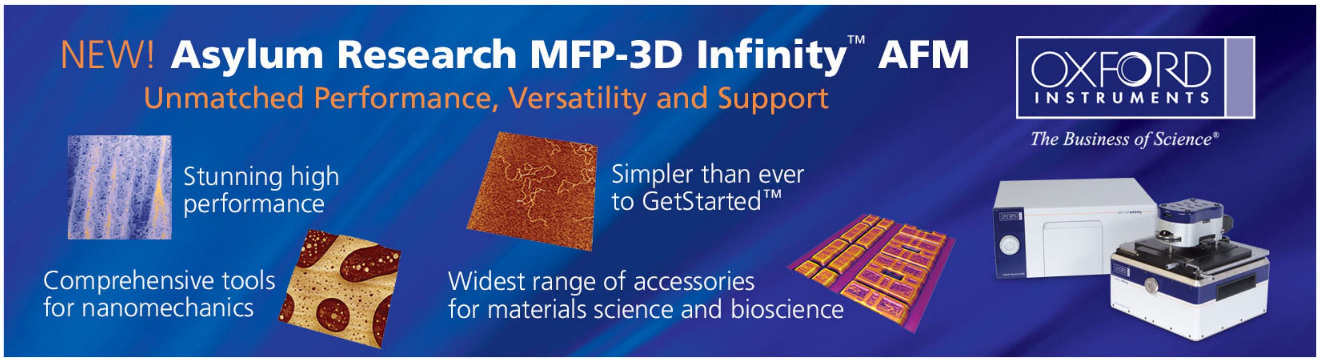

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1/*f* noise in micrometer-sized ultrathin indium tin oxide films

Sheng-Shiuan Yeh,[1] Wei-Ming Hsu,[1] Jui-Kan Lee,[1] Yao-Jen Lee,[2] and Juhn-Jong Lin[1,3,a)]
[1]*NCTU-RIKEN Joint Research Laboratory and Institute of Physics, National Chiao Tung University, Hsinchu 30010, Taiwan*
[2]*National Nano Device Laboratories, Hsinchu, 30078, Taiwan*
[3]*Department of Electrophysics, National Chiao Tung University, Hsinchu 30010, Taiwan*

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We have measured the low-frequency noises of ultrathin indium tin oxide films to investigate the effect of post annealing on the noise level. The noises obtained obey an approximate $1/f$ law in the frequency range $f \approx 0.1$–20 Hz. The microstructures and grain sizes of our films were altered by adjusting the annealing conditions. An enhancement of the noise level was observed for those samples comprising smaller grains, where numerous grain boundaries exist. This enhancement in the noise level is ascribed to atomic diffusion along grain boundaries or dynamics of two-level systems near the grain boundaries. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4821938]

Indium tin oxide (Sn-doped indium oxide or ITO) films are visible-light transparent materials with high electrical conductivity. They have widespread applications in liquid crystal displays, organic light-emitting diodes, and solar cells.[1–4] Recently, ITO films have been used as electrodes[5,6] and active channel layers[6] in fully transparent thin-film transistor (TFT) devices. For a material to be practically employed in miniature electronic devices, the level of the flicker noise ($1/f$ noise) is a key issue because a large $1/f$ noise could potentially hinder the device performance.[7] Thus far, there has been no report on the $1/f$ noise in ITO ultrathin films in the literature. While post thermal annealing is essential to the improvement of the optical transmission of ITO films,[8] it could, on the other hand, deteriorate the electrical conductivity.[9] In this work, we have carried out measurements of the $1/f$ noise in ultrathin ITO films. In particular, the effect of post annealing on the noise level is studied.

Our ITO ($In_{91.8}Sn_{8.2}O_{150-\delta}$) films were fabricated by rf sputtering deposition on glass substrates. They were supplied by the AimCore Technology Corporation.[10] In optical applications, thin films are required due to the demand of high transparency. Thus, we used 21-nm thick ultrathin ITO films in this study. Our samples were patterned using the electron beam lithography with a negative tone resist NEB-22 (Sumitomo Chemical Co.), followed by etching in a 10% aqueous solution of hydrochloric acid (HCl).[11] Each sample contained six terminals, as depicted in Fig. 1. This sample configuration was designed for the low frequency noise measurement using the ac bridge technique.[12,13] The dimensions of our samples were typically 10–20 μm in width and 40–100 μm in length. The sample volumes are listed in Table I. It was previously reported that post annealing in oxygen atmosphere could improve the blue-light transmittance of ITO,[14] we thus performed thermal annealing of our films in a continuous oxygen gas flow in a furnace. The samples were heated at a rate of 10 °C/min, maintaining at a final temperature of either 350 or 400 °C for 1 h, and then cooled to room temperature. The ac bridge noise measurement technique[12,13] modulated the low-frequency signals (i.e., the $1/f$ noise) to an optimal frequency of the preamplifier. The low-frequency signals were then demodulated using the lock-in technique. Only the noise but not the bias was measured by balancing the bridge circuit. A Stanford Research model SR560 amplifier, a model SR830 lock-in amplifier, and a model SR785 dynamic signal analyzer were utilized in our measurements. A modulation frequency $f_0 = 3$ kHz was chosen to minimize the noise contribution from the SR560 amplifier. A schematic diagram of our measurement circuit is shown in Fig. 1. Our data were all taken at room temperature.

It is known that the microstructures of ITO films can be altered after thermal annealing.[8,15] Hsu *et al.*[8] have recently observed modifications of the crystallinity of ITO films which were subject to various annealing temperatures and time durations. While a relatively low annealing temperature could lead to columnar nanograins, a relatively high annealing temperature could result in a transformation to equiaxed nanograins. The former structure contained comparatively few grain boundaries, while the latter contained numerous grain boundaries. In addition, Gulen *et al.*[15] have reported observations of variation of the grain size with post annealing temperature. Figures 2(a)–2(c) show the scanning electron microscope (SEM) images of our as-grown and post-annealed ultrathin ITO films. We found that the average

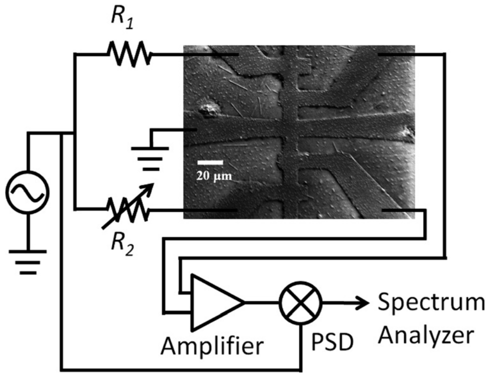

FIG. 1. An SEM image of sample C and a schematic diagram for our low-frequency noise measurement setup. The ballast resistor (R_1) has a fixed resistance of approximately 5 times that of the sample and the adjustable resistor (R_2) is used to balance the bridge.

[a)] Electronic mail: jjlin@mail.nctu.edu.tw

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TABLE I. Values for relevant parameters of ultrathin indium tin oxide films at 300 K.

Film	Annealing Temperature	Average grain Diameter (nm)	Sample volume ($\times10^{-17}$ m^3)	R (Ω)	ρ ($\mu\Omega$ cm)	n_c ($\times10^{26}$ m^{-3})	N_c ($\times10^9$)	α	γ
A	As-grown	10	1.2	540	281	5.0	6.0	1.3	0.011
B	350 °C	8.6	2.1	1120	646	2.2	4.5	1.1	0.018
C	400 °C	7.4	5.3	4280	1510	2.2	12	1.0	0.047

grain diameters are ≈10, ≈8.6, and ≈7.4 nm in films A (as-grown), B (annealed at 350 °C), and C (annealed at 400 °C), respectively. The corresponding grain size histograms are plotted in Figs. 2(d)–2(f). Clearly, our post annealing processes resulted in a reduction of the average grain size. The amount of reduction was more pronounced for the sample subject to a higher annealing temperature.

Figure 3(a) shows the voltage noise power spectrum density, S_V, at several bias voltages for sample A. It is seen that S_V obeys a $1/f^\alpha$ law with $\alpha \approx 1.3$. Furthermore, S_V possesses a V^2 bias voltage dependence, see Fig. 3(b). (Similar properties were also observed for samples B and C.) A widely accepted theoretical model for the origin of the 1/*f* noise considers the two-level systems (TLSs) which may be charge traps or nearby atomic positions in a conductor.[16,17] If TLSs fluctuate via thermal activation processes, the power spectrum density of resistance fluctuations, $S_R(f)$, can be expressed as the integration of individual power spectrum density of the thermal resistance fluctuations for a single TLS, weighted by the distribution of the activation energy in the TLSs. If the distribution of the activation energy varies slowly compared to the thermal energy, S_R should possess approximate $1/f$ dependence. In accordance with S_R, the measured S_V for an ohmic conductor can be described by the empirical Hooge expression[18]

$$S_V = \gamma \frac{V^2}{N_c f^\alpha} + S_V^0, \tag{1}$$

with $\alpha \approx 1$. The prefactor γ characterizes the normalized magnitude of the $1/f$ noise. It was found that $\gamma \approx 10^{-3}$–10^{-2} in typical metals,[17] and $\gamma \approx 10^{-7}$–10^{-2} in semiconductors.[19] N_c is the total number of charge carriers in the sample, and S_V^0 is the power spectrum density of the background noise. In our measurement setup (Fig. 1), $V = 2V_{\rm rms}$, where $V_{\rm rms}$ is the root-mean-square voltage drop across one half of the sample.[12,13] Our observation of a quadratic dependence of S_V on V is consistent with the resistance fluctuations and ohmic nature of our ultrathin ITO films.[9,20]

The γ value for a given sample can be extracted from the slope in a S_V versus V^2 plot according to Eq. (1), provided that the value of N_c is known. In our previous studies,[9,21] we had obtained the carrier concentration n_c from the temperature dependent thermoelectric power and the Hall effect measurements. We found a decrease in the n_c value

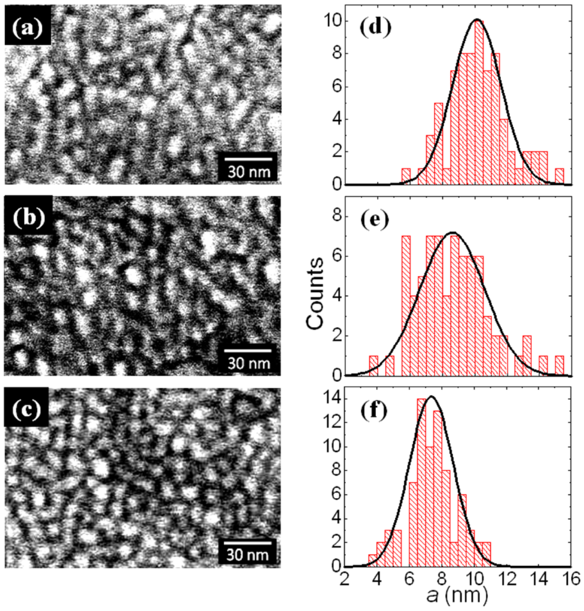

FIG. 2. (a)–(c) SEM images of samples A–C, as indicated. The pictures were taken in a Hitachi S-4800 field emission scanning electron microscope. (d)–(f) The corresponding histograms of grain size for samples A–C. The curves depict Gaussian distributions.

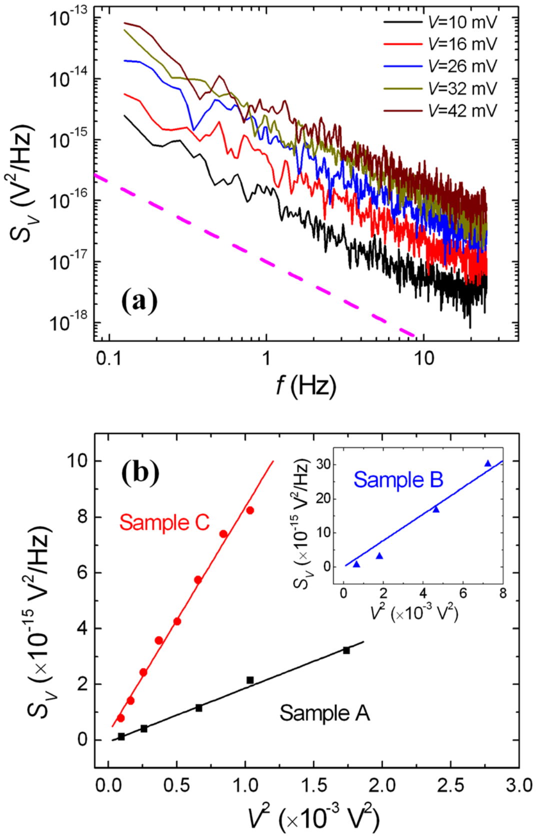

FIG. 3. (a) The voltage power spectrum densities of sample A under various bias voltages, as indicated. The dashed line is drawn proportional to $1/f^{1.3}$ and is a guide to the eye. (b) S_V ($f = 1$ Hz) versus V^2 for three ultrathin ITO films, as indicated. The straight lines are linear fits.

after thermal annealing in an oxygen atmosphere,[9] which could be ascribed to the oxygen vacancies in ITO being compensated.[22] Taking these factors into account, our extracted γ values (listed in Table I) fall in the range 0.01–0.05, which are in close consistency with those in metals with high concentration of defects.[23] We have plotted the variation of γ values with resistivity ρ in Fig. 4. It is worth noting that the measured ρ in ITO was dominated by the electron scattering from defects and grain boundaries while the electron scattering from phonons contributed only a few percent.[21,24]

We found that our extracted γ value is largest in the sample annealed at 400 °C but smallest in the as-grown film. In particular, we observe that γ increases with increasing ρ. A positive correlation between γ and ρ has previously been reported.[13,23,25] Pelz and Clarke[13] employed electron irradiation to introduce defects in Cu films. The introduction of additional defects raised the value of ρ and a subpopulation of mobile defects,[13,26] resulting in a larger γ. Fleetwood and Giordano[23] performed thermal annealing to vary the amount of defects in AuPd films. They found a higher defect level and a larger ρ would cause a larger γ. van den Homberg *et al.*[25] also found that, compared to single-crystalline Al films, the presence of extra grain boundaries in polycrystalline films enhanced both ρ and γ values. In particular, the authors attributed the increase in the noise level to the atomic diffusion along grain boundaries. Similar results had been found in Al films by Koch *et al.*,[27] and in Au films by Verbruggen *et al.*[28]

In our ITO films, the variation of γ cannot be ascribed to the variation of the number of oxygen vacancies, because our thermal annealing in an oxygen atmosphere would reduce the amount of oxygen vacancies[22] and lead to a suppressed γ. Instead, our observation of the change in grain size (Fig. 2) can shed light on the microscopic mechanism for the variation of γ. Accepting the aforementioned explanation that there exist atomic diffusions along grain boundaries[25,27,28] or TLSs near grain boundaries, we immediately conclude that a sample with more grain boundaries (and hence more fluctuation centers) will have a higher $1/f$ noise level.[19] In the present work, sample A possesses the largest grain size, rendering the least amount of grain boundaries, which thus causes a small value of γ. In contrast, sample C possesses the smallest grain size and numerous amounts of grain boundaries, which thus leads to a large value of γ. The case of sample B lies in between.

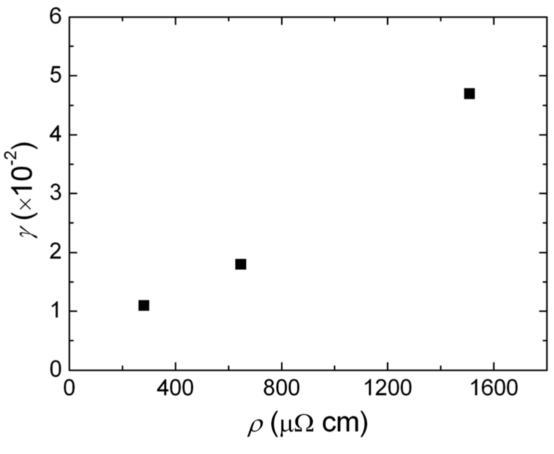

FIG. 4. Noise level γ as a function of resistivity ρ.

To explain the post-annealing induced modification of ρ in our ITO films, both the variation of n_c and the change in microstructures should be taken into account. Compared to sample A, the n_c value in sample B was reduced by a factor of ≈2.3, while the ρ value increased by a factor of ≈2.3. This implies that the change in ρ was dominantly originated from the change of n_c. In this case, the grain size was only reduced by a factor of ≈15%. In sample C, the n_c value was reduced by a factor of ≈2.3 from that in sample A, while the ρ value was increased by a factor of 5.4. Thus, the reduction of grain size (by ≈27%) and the accompanied increased amounts of grain boundaries should contribute significantly to the increase of ρ. Such marked effects of grain boundaries in sample C are consistent with our observation of an enhanced γ value in this particular film, as discussed. This experiment suggests that γ is a much more sensitive parameter than ρ to the (dynamic) microstructures of grain boundaries.

In conclusion, we have measured the $1/f$ noise in ultrathin ITO films and studied the effect of post thermal annealing on the noise level γ. An increase in the γ value was found, which can be ascribed to an increase in the amounts of grain boundaries. Microscopically, this can be ascribed to the atomic diffusion along grain boundaries or dynamics of TLSs near the grain boundaries. For applications in transparent electronic devices, while ultrathin ITO films are desired, they may possess notable granularity[20] and hence be accompanied with a high low-frequency noise level. The issue how to minimize the amount of grain boundaries deserves serious attention.

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