

Optical transmissivity and conductivity of metal films with micro-scale holes for transparent electrodes

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In this work, we studied optical transmission and electrical conduction properties of micro-scale holey metal (Al) films fabricated on glass substrates using conventional UV lithography and wet etching processes. For the properly etched holey metal films, the measured transmittance in the visible spectrum range is uniformly above 85%, and the effective sheet resistance is in the order of $\sim 1 \times 10^{-5} \Omega\text{-cm}$; somehow they show better figure-of-merit than other typical transparent conducting layers. Additionally considering that the holes are in the micro-scale dimension, such structured metal films can be applied in many optoelectronic devices.

(Received April 10, 2010; accepted May 20, 2010)

Keywords: Transparent electrode, Metal film, Optical transmission, Conductivity

1. Introduction

High figure-of-merit transparent electrodes are important to improve the overall performance of some optoelectronic devices. As indium tin oxide (ITO) films have been widely used in commercial devices, there has been a consistent interest of researchers to develop novel materials to replace ITO so as to reduce the cost (due to scarcity of indium) and meet the need for better transparent electrodes. In the past several years, people have studied utilization of novel nanomaterials, such as carbon nanotubes [1] and graphene films [2, 3], as transparent electrode materials. In spite of their high cost and low compatibility, these materials didn't show much superior performance than ITO. More recently, unstructured or structured ultra-thin metal films were also investigated for transparent electrodes [4, 6], which are shown to be promising for both their performances and simplicity in technical processing. For example, in Ref. 6, Ghosh *et al* first deposited a 2-nm-thick Ni film on substrate, and then they patterned 100-nm-thick Cu grid on Ni, with grid period 500 μm and line-width 5~20 μm ; they achieved optical transmittance of $\sim 80\%$ and estimated sheet resistance of $\sim 6.5 \Omega/\square$. In this letter, we experimentally show that when metal films (e.g. Al) are well etched with micro-scale hole arrays, optical transmittance of $>85\%$ and sheet resistance of $\sim 1 \Omega/\square$ (directed measured) can be achieved. Due to the micro-scale dimensions in addition, such patterned metal films can be very suitable for transparency electrodes applications. Here it's also noted that, if the dimensions of the hole arrays are in the sub-wavelength scale, the transmittance will be strongly reduced or only transmissive in narrow-bands, thus they are not suitable for wide-band optical transparency [7, 8].

2. Experimental details

The holey metal films in this work were fabricated with

the following processes. Firstly, a 100-nm-thick Al film was deposited on a cleaned microscopy glass using magnetron sputtering process. Then, 500-nm-thick positive photoresist (Shipley PR1805) was spin-coated on the Al film, and exposed using the standard UV lithography (Karluss MA6/BA6 mask aligner). The patterns on the photo-mask was a square lattice array of square apertures with period of 4 μm and aperture side-length of 2 μm . After developing of the exposed photoresist, the samples were dipped into the solutions of [$\text{H}_3\text{PO}_4 : \text{C}_2\text{H}_4\text{O}_2 : \text{HNO}_3 : \text{H}_2\text{O} = 16 : 2 : 1 : 1$ (vol)] at room temperature ($\sim 20^\circ\text{C}$) to etch the underneath Al using the residual photoresist pattern as a mask. Finally, the photoresist was removed with acetone. In the above processes, by controlling the etching time, Al was etched in different levels, which resulted in Al patterns with different morphologies. I.e. if the Al are less etched, the patterns are small hole arrays; and if the Al are over-etched, the patterns are island arrays. For the above etching conditions, at an etching time of approximately 15 minutes, circular hole arrays with extremely narrow partition can be resulted, as shown in the microscopy image in Fig. 1. And this condition is considered to be optimal for the structure to be used as a transparent electrode layer. Then we characterized the optical transmission and electrical conductivity of the fabricated structures.

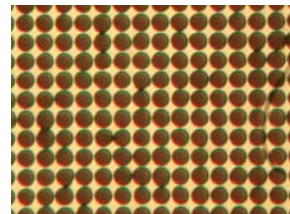


Fig. 1. Microscopy image of a metal film on glass substrate properly etched with micro-scale holes for transparent electrode application. The holes are in square lattice with period of 4 μm .

3. Results and discussion

Fig. 2 shows the transmission spectra of as-fabricated samples in different spectrum regime. In Fig. 2(a), the spectra were characterized using a UV-Visible spectrophotometer (CARY 300). All the three samples were fabricated under the optimal conditions. We can see that the transmittance is well above 85% in the visible range. It's noted that the abrupt transmittance drop in the UV range is due to strong UV-absorption of substrate glass. We can estimate from the microscopy image or roughly calculate that the holes occupy approximately 78% of the whole area, but here higher transmittance of over 85% is resulted. For this "over-transmission", there are surely minor contributions from classical diffraction effects and direct transmission of light through such thin metals (100-nm-thick). But we think it is mainly due to excitation of surface plasmons (SP) at the edges of the holes and even on in-plane metal surfaces due to their granular morphologies, which undergoes a funneling effect [9-11]. The funneling effect guide surface plasmons, coupled from otherwise obstructed incident light, to flow along the metal surfaces and transmit to the other side of the holy metal layer; further, SPs on the other side can be decoupled into transmitted light. As such, nearly 90% of the optical power impinging on the obstructed surface area, which laterally extends from the metal edge for hundreds of nanometers, can effectively transmit in the forward direction [10, 12]. Here, for our samples prepared in optimal conditions, the partition width is in the deep sub-micrometer scale, and the width of the metal area surrounded by four neighboring holes is in the order of 1~2 μm . Considering that funneling effect takes place at both opposite edges of the metal patterns, it's rational for us to accept the result of experimentally high transmission.

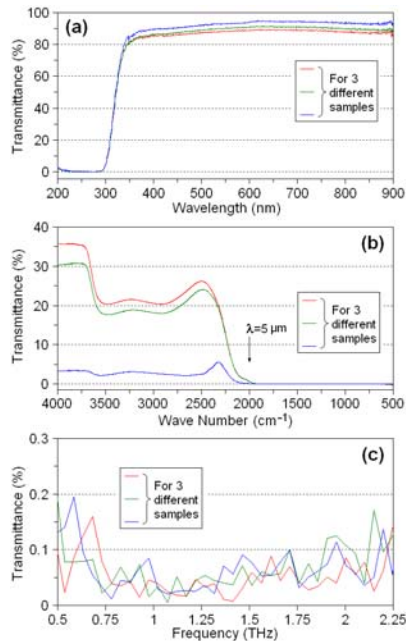


Fig. 2. Optical transmission spectra of the holy metal films in the (a) VIS-NIR range, (b) IR range and (c) THz range.

Since our project was also designed using the holy metal film to block the low-frequency electromagnetic waves, we also studied the transmission properties in the infrared (IR) and terahertz (THz) spectrum range. The infrared transmission spectra was characterized using a Fourier transform infrared spectrometer (Nicolet Avatar 330). The results are typically shown in Fig. 2(b) for samples prepared in different Al etching levels (the red and green lines correspond to samples Al-etched at nearly optimal conditions). We can clearly observe that there is a cut-off at the wavelength of around $\lambda=5 \mu\text{m}$, which is roughly in consistent with the prediction from waveguide theory for light transmission through holes. Below the cut-off wavelength, as the Al of a sample is more etched, its transmittance appears to be larger. But for all three samples, the transmittance above the cut-off wavelength is measured to be well below 1%, or even below 0.1%. The THz transmission spectra were obtained using a time-domain THz spectroscopy system (in the Key Lab of THz Photonics in the Capital Normal University, China). Extremely low transmittance is also shown in Fig. 2(c) for the samples in Fig. 2 (b).

Conductivity property of the samples was characterized with the four-probe method (with a micro-controlled four-probe stage, D41-11A/ZM, Beijing Jianzhong Machinery) and current-voltage (I-V) characterization method (with a Keithley 6514 electrometer, a Keithley 2410 sourcemeter and a probe stage). In both methods, electrical contact is via metal probes. Since tip sizes of the probes (in the order of tens of micrometers) are much larger than the cell size of the hole arrays, the measured data are believed to be effective values that are comparable with those of a continuous conducting film. Table I shows the measured sheet resistance values of three samples. The sample A is relatively less etched in Al etching, and the sample B and C are enough etched under optimal conditions. Each sample was measured for three times at different sites, and a good uniformity of conductivity was observed. As the sample area ($\sim 1 \times 1 \text{ cm}^2$) is much larger than the size of the four probes (interspacing = 1 mm), using the initial Al film thickness as the effective thickness of the conducting layer, the effective resistivities of the samples were also calculated, as shown in the table. We can see that the resistivity of as-prepared metal layer is approximately in the order of $\sim 1 \times 10^{-5} \Omega\text{-cm}$, approaching that of bulk Al ($\sim 2.9 \times 10^{-6} \Omega\text{-cm}$), and much smaller than that of typical ITO films ($\sim 10^{-4} \Omega\text{-cm}$). Fig. 3 shows the characterized I-V curve for the sample B in Table 1.

Table 1. Measured sheet resistance of holy metal films and corresponding resistivities.

Sample	Sheet Resistance R_{\square} (Ω/\square)				Resistivity ρ ($\Omega\text{-cm}$)
	1 st -site	2 nd -site	3 rd -site	Average	
A	0.43923	0.41609	0.41226	0.422527	4.23×10^{-6}
B	1.6354	1.3365	0.87694	1.282947	1.28×10^{-5}

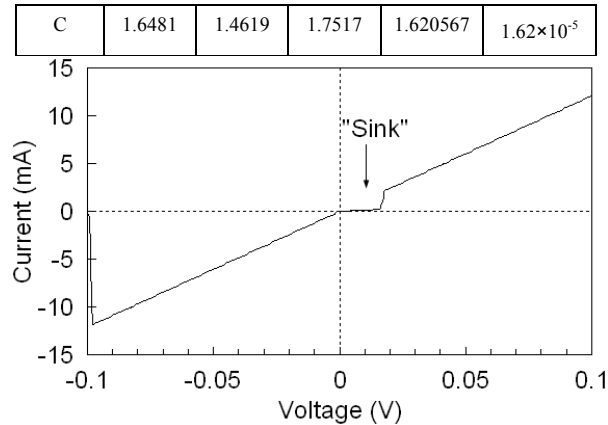


Fig. 3. I-V characteristics of an enough etched holy metal film.

In I-V characterization, the current was the mean value of 100 measurements at each voltage, and the applied voltage scans from negative to positive values with a step of 0.001 V. For the 7-mm-spacing of two probes, the measured resistance, i.e. inverse slope of the I-V curve, is 8.23Ω . Interestingly, when the polarity of the scanning voltage is just changed to the opposite, there always appears a “sink” on the I-V curve. Although we postulate it is related to a retardation effect induced by parasitic inductance and capacitance of the holy metal films, we cannot give an affirmative explanation of this phenomenon.

4. Conclusions

In summary, we studied the optical transmission and electrical conductivity properties of metal (Al) films with properly etched micro-scale holes. The transmittance in the visible range is shown to reach more than 85%, and the effective sheet resistance of the metal structure layer reaches $\sim 1 \Omega/\square$, corresponding to the effective resistivity of $\sim 1 \times 10^{-5} \Omega \cdot \text{cm}$. Since dimensions of the holes are in the micro-scale, typically well within the carriers' diffusion length in semiconductors, such structured metal films can be an alternative choice for transparent electrodes in optoelectronic devices, such as light emitting devices, solar cells, and liquid crystal displays etc.

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