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Non-ohmic transport behavior in ultra-thin gold films

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ABSTRACT

Structure and local lateral electrical properties of Au films of thicknesses ranging from 10 to 140 nm are studied using conductive atomic force microscopy. Comparison of current maps taken at different thicknesses reveals surprising highly resistive regions (10^{10}–10^{11} Ω), the density of which increases strongly at lower thickness. The high resistivity is shown to be directly related to discontinuities in the metal sheet. Local *I–V* curves are acquired to show the nature of electrical behavior relative to thickness. Results show that in Au films of higher thickness the electrical behavior is ohmic, while it is non-ohmic in highly discontinuous films of lower thickness, with the transition happening between 34 and 39 nm. The non-ohmic behavior is explained with tunneling occurring between separated Au islands. The results explain the abrupt increase of electrical resistivity at lower thin film thicknesses.

1. Introduction

Thin metal films are of great interest in a vast variety of technologies in optics and microelectronics. In energy efficient and smart windows application, Au thin films are used as selectively transparent coating and as transparent electrodes in metal–insulator–semiconductor MIS solar cell devices [1]. As a result electrical characterization of these film structures at the nano-scale is of great importance.

Electrical resistivity of thin metal films differs markedly from that of their bulk counterpart, moreover, it increases as the film thickness decreases [2]. As metal film thickness approaches the electron mean free path (EMFP) the abrupt increase in resistivity is often attributed to surface scattering and grain boundary scattering [3,4]. It is difficult to separate the influence of surface scattering from other factors related to the film structure [5]. Morphology, nucleation and growth of Au thin film, in relation to the film preparation process and the electronic properties of the films, were extensively studied by means of ex situ TEM [6], and STM microscopy [7,8]. More recently, the kinetic growth of Au film was described by in situ STM during deposition [9] and annealing process [10]. In reported studies the rapid increase in resistivity at lower thickness is associated to the transition from a continuous to a discontinuous film structure [11,12].

Structural investigation of sputtered and evaporated Au thin films have shown to be discontinuous before a certain threshold at a thickness of some hundred angstroms, and only when the film thickness is greater than 30 nm do the films become completely continuous [11]. Non-ohmic behavior of discontinuous thin Au films studied theoretically in the past, usually attributes to different mechanisms of electron-transport between separate metal islands [13]. Morris [14] proposed a tunneling model of conduction to interpret the non-ohmic behavior in discontinuous films. Dittmer argued that this behavior is related to substrate assisted tunneling, where small amounts of the metal exist in the first layer of the isolating substrate and assist the tunneling between the metal islands [15].

The vast majority of previous studies the experiments are conducted vertically to the film plane [16,17] or at the macro-scale [18] making it difficult to correlate the electrical transport behavior to the local structure of the thin film. Recently, non-ohmic behavior of Au multi nanoparticle arrays was reported to be dependent on the nanoparticle sizes [19]. In this work we study lateral (in-plane) electrical conduction behavior in Au thin films at the nano-scale using conductive AFM. Lateral conduction incorporates the effect of topography features in the plane on the current path. Using lateral conduction we show the effect of granular structure and inherent discontinuities at different thicknesses on electrical conductivity of Au thin films. We relate this effect to tunneling in lateral conduction at nano-scale leading to non-ohmic behavior occurring at film thicknesses below 39 nm.

2. Experiment

2.1. Sample preparation

Physical Vapor Deposition is used to deposit thin Au films, 10–140 nm thick, on soda–lime glass slides substrate. The sput-

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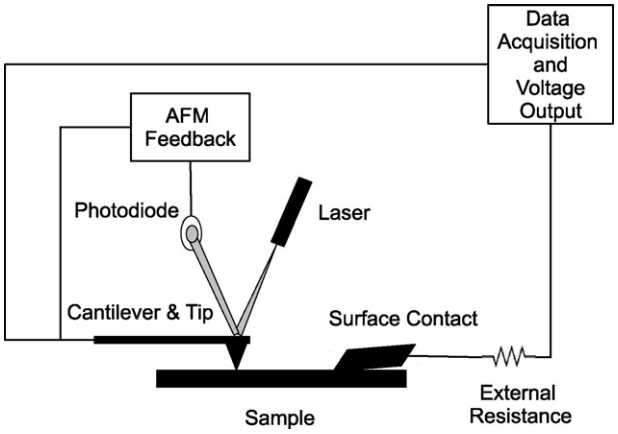

Fig. 1. Experimental setup. C-AFM system with surface current sensing setup on sample.

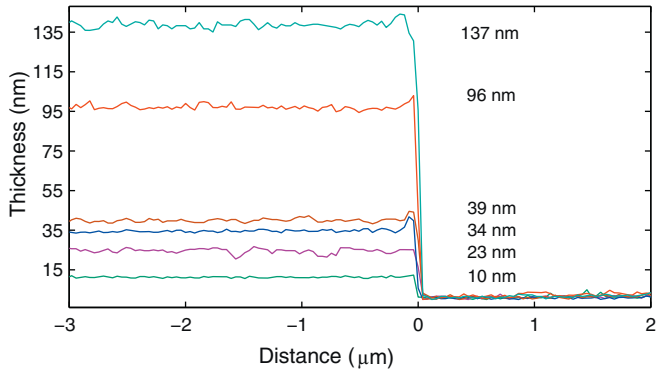

Fig. 2. AFM cross sections for the Au films in this study.

tering targets are 99.999% Au. The samples are prepared under 1 m Torr residual Argon pressure and 50 W RF power during deposition. The deposition rate of sputtered material is ~0.5 A/s for all samples.

2.2. Experimental setup

An Asylum Research MFP-3D dual-gain Conductive Atomic Force Microscope (C-AFM) is employed for measuring the electrical and morphological properties of the samples simultaneously while in contact mode. The used probes are conductive tips of 15 nm Ir coating that acts as a conductive layer to insure both good electrical conduction and mechanical resistance. Underlying the Ir coating is a 5 nm Ti coating adhesive layer deposited over Si. The cantilever has an Al reflective layer of 15 nm. The cantilever's spring constant is in the range of 2 nN/nm. The tip estimated radius is 23 nm. The C-AFM module has a current sensitivity in the range of 1 pA–10 nA. In this experiment lateral resistivity collected from the surface of the Au samples is measured as shown in the setup in Fig. 1. An external resistance of 1 GΩ is attached to obtain current maps in the current sensitivity range of the device, and more importantly to prevent the heating and changing the structure of the conductive tip by the Joule effect due to the resulting high current density as suggested by Parkhi and Gross [20].

3. Results and discussion

Au film thicknesses are independently determined using AFM step heights of scratched regions. Fig. 2 shows the cross sections of the different films. Highly reproducible electrical and topographical 1 μm × 1 μm area scans are produced using C-AFM on samples of the shown thicknesses.

Fig. 3(a) and (b) shows two topographical sample images. The different topography maps show a granular structure with difference in the grain size as the thickness increases. Fig. 3(c) shows how the grain structure profile of the film changes with thickness. The grain width ranges from ~20 nm to ~150 nm increasing with thickness. The results appear to be in good agreement with morphological studies conducted on Au thin films using TEM [12]. The difference in the grain structure is related to the kinetic growth at different stages during the film deposition. At the beginning, the grains are small, and as more metal is deposited the grains start to grow in size and get connected to other grains leading to a more continuous film structure [12]. Unfortunately, the transition between continuous and discontinuous film structure cannot be solely determined from the AFM morphology study.

Cleaning scans procedure [21,22] was used to insure the removal of the tip contaminations prior to conducting the C-AFM scans. Several topographical scans were done for this purpose. As a result current images and measurements obtained were highly reproducible. Fig. 4 shows current maps obtained for thicknesses of 10, 34, 39 and 137 nm applying voltages ranging from 1 to 5 V. The maps show light colored areas and dark areas. The light areas represent high level currents, which when given a constant applied sample voltage, yields lower resistance. The dark areas, on the other hand, represent lower currents, hence higher resistance. The different maps show the presence of highly resistive areas in all thicknesses. By comparing the current maps with their corresponding topographical ones, the highly resistive areas are found to be randomly distributed over the surface, and are not related to certain topographical features.

The resistance of the darker areas is estimated by dividing the applied voltage by the measured current yielding resistances in the order of 10 to 100 GΩ which is one to two orders of magnitude larger than the external resistance added. To explain the origin of the high resistance reported, the metal/metal contact resistance between the tip and the Au surface is estimated by means of Sharvin's formula: [23]

$$R = \frac{4\rho l}{3\pi a^2} \tag{1}$$

where ρ, l and a, denote the resistivity, the EMFP of the metal, and the contact radius, respectively. The mean resistivity of contact between the Ir probe coating and the Au surface is estimated by: $\rho_{mean} = (\rho_{Au} + \rho_{Ir})/2$ to be 34.62 nΩ m. The geometrical resolution of current measurements is estimated by calculating the area of the tip/sample mechanical contact using Hertzian model [24]. The contact radius is estimated to be ~2 nm,[1] which corresponds to a mechanical contact area of 12 nm^2. One can estimate the contact resistance to be ~100 Ω given the EMFP for Au of 38 nm [5]. This result implies that the high resistance observed cannot be related to tip/surface contact resistance.

At the same time the high resistance, above 10 GΩ, cannot be the contact resistance between Au grains. Using Eq. (1), this would lead to an unreasonable contact radius in the order of 10^{-3} Å. Hence,

[1] The radius of the contact zone a is given by the following equations

$$a = \left(\frac{4}{3}\frac{FR_t}{E}\right)^{1/3} \quad \text{with} \quad \left\{ \begin{array}{c} F = k\,\Delta D \\ \frac{1}{E} = \frac{1-\nu_1^2}{E_1} + \frac{1-\nu_2^2}{E_2} \end{array} \right\} \tag{2}$$

where F is the tip/sample applied force, k the lever spring constant, ΔD the lever deflection. R_t is the tip apex radius, E_1, E_2, ν_1 and ν_2, denote the Youngs moduli and the Poissons ratios of the tip and the gold respectively. With typical parameters F = 150 nN, R_t = 70 nm, $\nu_1 = \nu_2$ = 0.33, E_1 = 528 GPa (iridium) and E_2 = 79 GPa (gold).