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Carrier density and DC conductivity of ultrathin aluminum films

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During the last two decades, the size of electronic devices and systems has been decreased continuously and recently nanometer scale range was attained. Unusual properties and performances have been obtained with nanostructured materials. Ultrathin metal films are an important part in electronic devices and systems and their electrical conductivity has been paid some attention [1, 2].

There have been many studies demonstrating the thickness dependence of thin metal films on electrical conductivity [3–5]. Some authors attributed the different electrical conductivity of ultrathin metal films from bulk metals to the scattering of electrons by surfaces [6, 7] and grain boundaries [8, 9] in films, as well as to quantum size effects [10, 11]. Also, a microstructure transition was proposed as another reason for size effect of ultrathin metal films on electrical conductivity [12–14]. In this article, the dependence of direct current (DC) conductivity or the carrier density of ultrathin aluminum films is attempted.

Ultrathin aluminum films were deposited by DC magnetron sputtering with a cylindrical target made of high-purity aluminum in a high vacuum system pumped to a base pressure of 4×10^{-4} Pa. The polyester foil substrates were pretreated ultrasonically by ethanol before deposition. The purity of Ar gas for sputtering was 99.999 wt%. The gas flow was controlled using electronic mass flow meter and the gas pressure was measured using a capacitance vacuum gauge. The titanium target was sputter cleaned for 3 min, before the deposition under the established conditions. A programmable timer controlled the dwelling time that substrates spent under target. The pressure in the vessel during deposition was 8×10^{-2} Pa. The voltage and the current of the target were 400 V and 4 A, respectively. During the film growth, the deposition parameters were maintained as constant as possible. All of the ultrathin aluminum films were obtained under the same experimental conditions.

The DC conductivity of ultrathin aluminum films was obtained from multiplying the electrical resistance and thickness of the films. The sheet resistance of the films was measured with the four probe resistance method from *in situ* measurement by obtaining the voltage drop across the film, as well as the current through the film outside of vacuum vessel, to make sure the change of DC conductivity of ultrathin aluminum films during their growth process can be recorded in detail simultaneously. Details of the measurement were described elsewhere [12, 13].

In a separate set of experiments, the thickness dependence of ultrathin aluminum films on carrier density was obtained using the Hall effect measurement with a HL5500 PC Hall effect measurement system.

Fig. 1 shows the evolution of DC conductivity with thickness of ultrathin aluminum films. Ohmic behavior is first observed for the film when the film thickness is 2.1 nm and DC conductivity of the films increases with their thickness until 50 nm. It increases abruptly when the film thickness is in the range of 5 and 35 nm, and remains relatively constant when the film thickness is up 50 nm. At the same time, a marked decrease in DC conductivity by many orders on magnitude can be found as the film thickness is reduced below 4.2 nm. It is worth to point out that even the maximum value of DC conductivity of ultrathin aluminum films is much lesser than that of bulk aluminum $3.8 \times 10^7/\Omega$ m [15], which can be attributed to the scattering of electrons by surfaces and grain boundaries in the film.

Fig. 2 shows the thickness dependence of ultrathin aluminum films on carrier density. The carrier density of aluminum films increased with film thickness until 55 nm, also a marked decrease in carrier density by many orders on magnitude can be found as the film thickness is reduced below 14.4 nm. These values on carrier density of ultrathin aluminum films are much

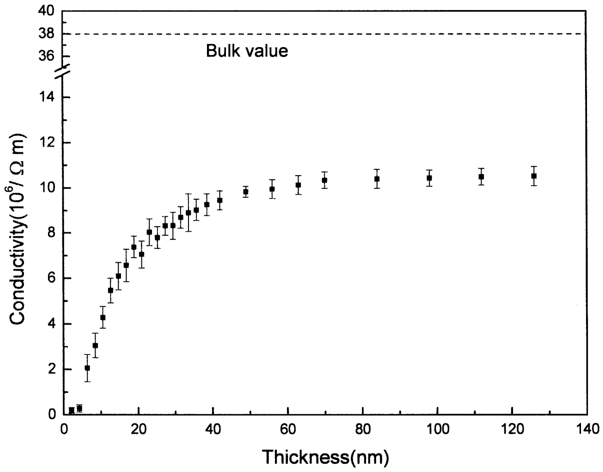

Figure 1 Thickness dependence of ultrathin aluminum films on DC conductivity.

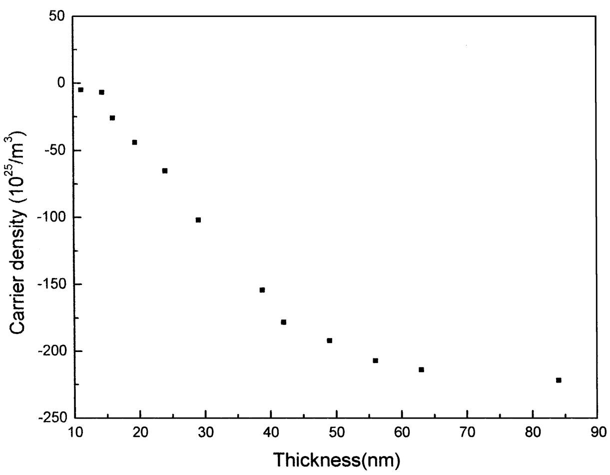

Figure 2 Thickness dependence of ultrathin aluminum films on carrier density.

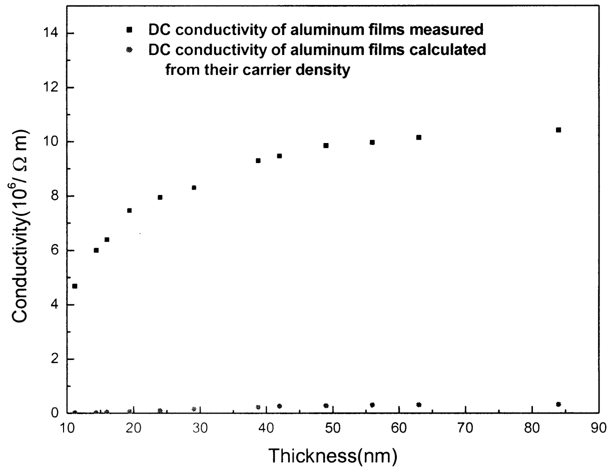

Figure 3 Comparison of ultrathin aluminum films on DC conductivity between measured value and calculated value from their carrier density.

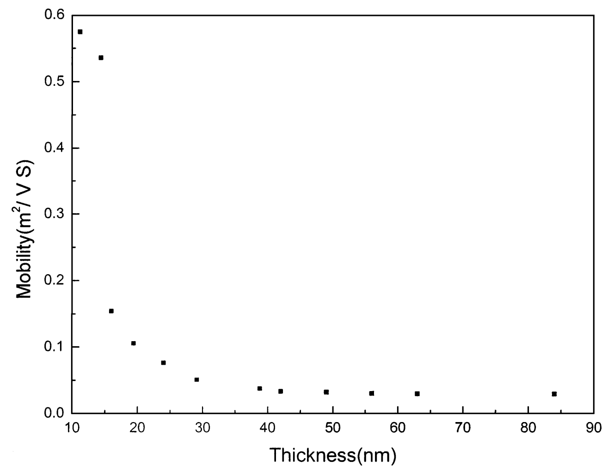

Figure 4 Thickness dependence of ultrathin aluminum films on carrier mobility.

less than that of bulk aluminum $1.8 \times 10^{29}/\text{m}^3$ [15, 16].

It is well known that electrons in a metal are distributed over a number of energy bands. All the electrons are free to move through the lattice. Only those which are contained in incompletely filled energy band can contribute to the resultant current and are to be regarded as free electrons. The precise number of free electrons depends on the detailed configuration of energy bands [7]. The alternation of energy bands and the limitation by so much grain boundaries in ultrathin aluminum film cause an abrupt decrease in the carrier density. Generally, the size of metal grains increases with increase in film thickness [17], resulting in decreased limitation by boundaries and in increased carrier density [7, 18]. The alternation of microstructure during aluminum film growth is one of the reasons for variation of carrier density.

As the conductivity (σ) is determined by the free carrier density (n) and the carrier mobility (μ):

$$\sigma = ne\mu$$

The DC conductivity of the ultrathin aluminum films is predicted with their carrier density and shown as Fig. 3. In the calculation, μ_e is taken as 0.9×10^{-3} $\text{m}^2\text{V}^{-1}\text{S}^{-1}$ [19]. It is obvious from the comparison that the formula fails to explain the experimental data. The deviation indicates that carrier mobility of ultrathin aluminum films cannot be a constant, there must be a dependence of this parameter on the film thickness.

By the formula, the thickness dependence of ultrathin aluminum films on carrier mobility is obtained and shown in Fig. 4. The carrier mobility of ultrathin aluminum films decreases with increase in the film thickness. It is noticeable that all the carrier mobility values are much more than that of bulk aluminum, 0.9×10^{-3} $\text{m}^2\text{V}^{-1}\text{S}^{-1}$ [19]. This should come from the decrease in carrier density. As carrier mobility depends on the average time between two collision, the decrease in carrier density of ultrathin aluminum films leads the increase in average time between carrier collisions, which result in the increase in the carrier mobility. In fact, the size of metal grains increased with the film thickness. In this sense, the carrier mobility should be increased with the film thickness as the limitation of grain boundary to electrons decreased, but the increase in carrier density will play an opposite effect on carrier mobility. The experimental results in Fig. 4 show the limitation: the grain boundary plays a more important role when the film thickness is no more than 15 nm, as the carrier density changes only a little and the carrier density play more important role on carrier mobility when the film thickness is more than 20 nm.

From all the results discussed above, carrier density plays an important role on DC conductivity and carrier mobility of ultrathin aluminum films.

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