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Fabrication and electrical properties of the monolayer of oxidized nanometer-size metallic granules

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Abstract. A new method for the fabrication of 10 nm metallic granules is suggested, which is based on fission of liquid drops produced by laser ablation. A monolayer of such granules was deposited on the insulating substrate and then partly oxidized in air. It is shown that the conductivity of this layer, measured in lateral geometry, occurs due to electron tunneling between oxidized metallic granules having the charging energy as high as 300 meV.

Introduction

Several techniques have been suggested [1, 2] to fabricate the structures comprising nanometer-size metallic particles separated by tunnel barriers. The conductivity of 3D [1], 2D [2], and quasi-1D [3] conductors made of such granulated materials have been studied intensively to show that single-electron charging energy of individual particle is one of the main parameter determining the transport properties even at relatively high temperature. As it follows from recent consideration [4], such materials are promising for the fabrication of single-electron devices operating at room temperature. In this work we report on new technique for fabrication of low size dispersion 10 nm Cu granules which were first embedded on the insulating substrate in one monolayer and then partly oxidized to form the inter-granular barriers of native Cu oxide. The I–Vs and temperature dependence of conductivity of such layer were studied in lateral geometry.

1 Fabrication of experimental samples

Used in this work the method of nanometer-size Cu granules formation is based on fission of charged liquid metallic drops initially produced by laser ablation. The schematic of the experimental setup is shown in Fig. 1. The metallic Cu target was placed on rotating mount fixed inside the vacuum chamber (grounded) which was pumped down to 10^{-5} Pa. The DC voltage (3 kV) was supplied to the anode placed 7 cm apart from the target. The beam from pulsed AIG Nd³⁺ laser was introduced into the chamber through the side port and focused on the target surface to produce the power density about 10^9 W/cm². Finally, the alumina substrate was fixed near the anode as shown in Fig. 1.

The laser ablation of the Cu target results in flashing out the Cu particles with the dimensions wide spread in the range from the atomic size up to few microns. The largest particles (maternal drops) are liquid, which was checked in special experiment. These particles get charged in the plasma cloud and may become to be unstable if the Coulomb repelling force is higher than the surface tension. The instability threshold is

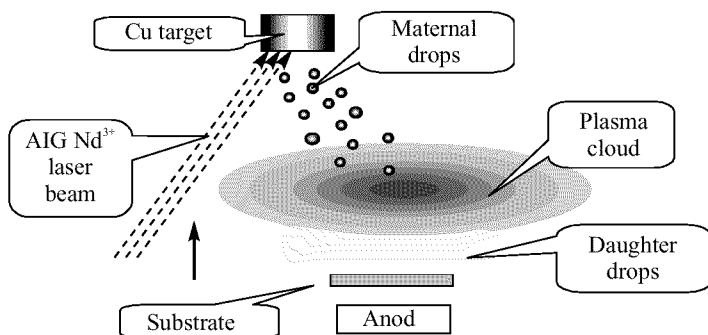


Fig 1. Schematic of experimental setup for Cu granules formation.

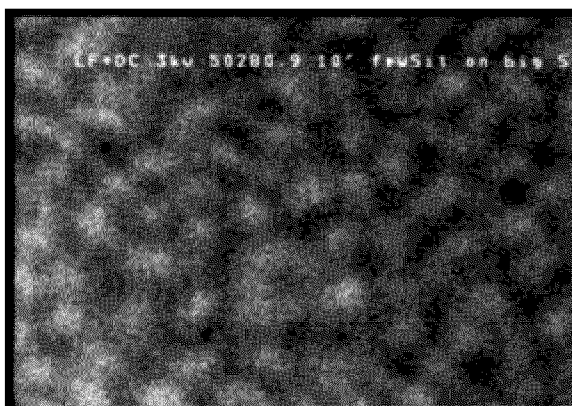


Fig 2. SEM image of fabricated monolayer (plan view).

given by Rayleigh condition [5]:

$$\frac{Q^2}{16\pi\alpha R^3} \geq 1, \quad (1)$$

where α is the surface tension, R and Q are the radius and the charge of maternal drop. Charged up to Rayleigh limit (1) each maternal drop ejects daughter drops which are also unstable and produce the next generation of daughter drops. This process of drop fission stops when the drop size comes to a certain minimum value. This final size is controlled by the rate of autoelectronic charge emission from the drop surface and is determined by the material work function. As shown in Fig. 1, the daughter drops are collected at the substrate surface. In our experiments the minimum size of Cu drops was about 10 nm and one monolayer of these granules was deposited. Certain amount of small (atomic size) Cu particles also reaches the substrate.

When the structure was taken out from the vacuum chamber it was exposed in air during some time. In such a situation, the Cu granules (and the background of atomic-size particles) were naturally oxidized in the atmosphere. Thus, the structure containing Cu granules covered by native Cu oxide was formed. SEM image of the resulting structure is shown in Fig. 2 (plain view). As one can see in this figure the granules

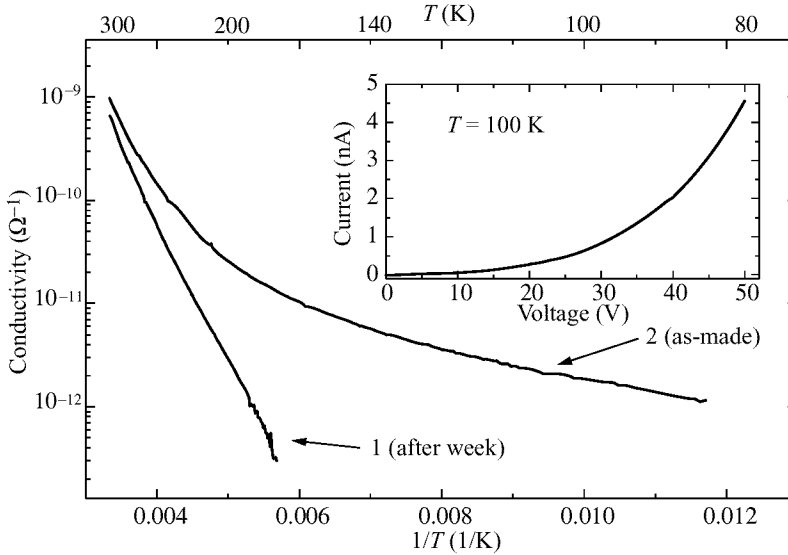


Fig 3. I-V curve and temperature dependences of structure conductivity.

have well defined size, about 10 nm, and they are dense packed, closely contacting the neighbors.

Taken from the chamber, the structure was deliberately etched by Ar^+ ions and then Cr contacts ($400 \times 400 \mu\text{m}^2$) were deposited on the top of the layer, the gap between the contacts being $5 \mu\text{m}$. The conductivity of the layer was measured in lateral geometry.

2 Experimental results and discussion

The inset in Fig. 3 shows the example of dc I-V curve measured at 100 K. At voltages below ~ 10 V the characteristic is linear but at higher voltages the exponential-like behavior is observed. This voltage corresponds to intergranular voltage drop about kT at 100 K and thus separates so-called low-field and high-field regimes [1]. Fig. 3 shows also the temperature dependencies of conductivity measured in low-field regime. The curve (1) was obtained after the same sample was kept in the room ambient during one week, while the curve (2) was measured just after the sample preparation. In both cases the temperature-activated conductivity is observed. The slope of curve (1) corresponds to the activation energy equal to 300 meV, which is associated with the charging energy of a metallic granule having the diameter and effective dielectric constant product $d\epsilon = 5$ nm. In well-oxidized structure such granules may exist inside the oxide shell. As one can see in Fig. 3, the conductivity of as-made layer (curve 2) has strictly different behavior: at low temperatures the conductivity is higher and the activation energy is small, about 30 meV (in the range 90–130 K). Apparently, at low temperatures the conductivity of as-made sample reveals the input of another transport mechanism. This mechanism may be electron hopping between some weakly localized states. We believe these states are associated with the background of small Cu particles, which “contaminate” as-made sample and disappear in due course of film oxidation.

3 Conclusion

We have fabricated 2D layer of closely packed nanometer-size metallic granules. After appropriate oxidation of this layer, pure exponential temperature dependence of low-field conductivity was observed with the activation energy as high as 300 meV which is well consistent with the charging energy of metallic granules residing inside the shell of native oxide. Such a behavior of conductivity is assumed to result from very low size dispersion in the system, which is of particular interest for further design of single-electron devices.

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