Tunnel percolation and current path switching in a granular metal

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Abstract. By means of electron beam induced deposition we have prepared a series of granular metals consisting of tungsten nanocrystallites embedded in an insulating matrix. The metal content varied between 19 at% and 34 at%. The samples have been analyzed by voltage-current and temperature-dependent conductivity measurements. Within the insulating matrix the nm-sized metallic nanocrystallites are irregularly distributed so that the classical percolation threshold can be assumed to be at approximately 50 at% metal volume fraction. Consequently, within the classical limit no current transport at low temperatures would be expected. Nevertheless, for samples with metal volume fraction above 24 at% the temperature-dependent conductivity extrapolates to a finite value for $T = 0$. We argue that clustering of nanocrystallites to larger aggregates of enhanced tunneling probability between the cluster members may be the reason for the observed behaviour. The transport can then be considered as a tunnel percolation process between clusters. As a consequence the current is inhomogeneously distributed in the samples. From analyzing different voltage regions of the voltage-current characteristics, separated by a sample-dependent threshold voltage $V_C$, showing either reversible (at lower voltages) or irreversible (at higher voltages) behaviour, we conclude that only one current path is active at any given moment and that this current path need not be that of the highest possible conductance. In particular, in some instances it proved possible to increase the conductance by a factor of $10^6$ after having passed $V_C$.

1. Introduction

Based on the model of link percolation applied to granular metals such phenomena as inhomogenous current flow, current path switching, current path activation and re-activation can be qualitatively understood. The simplest type of a link-percolation model is that on a regular lattice in which the lattice sites are connected with a probability $p \leq 1$. For current transport to occur between contacts at opposite sides of the sample, at least one continuous path of linked sites (current path) is needed. For granular matter the “sites” are defined by metallic nanocrystallites and the links are formed by the finite tunneling probability between the crystallites. We prepared our samples of granular matter by electron beam induced deposition (EBID). This technique is known for yielding materials with an internal structure consisting of metallic nanocrystallites embedded in an insulating carbon matrix [1]. This structure is highly disordered. We measured the voltage-current characteristic, $I(V)$, and analyzed the switching behaviour in the current paths as the bias voltage was varied. We investigated samples with varying metal content between 19 at% and 34 at%.
2. Experimental
For sample preparation we utilized a dual-beam system (FEI, Nova NanoLab 600) with a Schottky-type electron emitter having an ultimate resolution of 1 nm. The precursor gas was tungsten hexacarbonyl W(CO)$_6$. We used p-doped silicon as substrate material insulated via a 300 nm thick thermally grown SiO$_2$ layer on which 100 nm thick aluminum contacts had been previously patterned by standard lithography techniques. The metal content of the samples composition was determined by in-situ energy dispersive x-ray analysis at 5 keV electron beam energy. A typical sample used for our transport measurements appears in Fig. 2.

![Figure 1. Typical EBID sample (strip) on Si(p)/SiO$_2$ chip with 100 nm thick aluminum contacts.](image)

3. Results and discussion
In the available metal content range from 19 at% to 34 at% the existence of a direct percolation path between the W nanocrystallites can most likely be excluded [2]. We observed current flow in all of our samples, which one would not expect within a simple percolation model, yet several experimental observations suggest that tunnel percolation effects, possibly between clusters of nanocrystallites, have a role to play in these materials. We suggest a tunneling percolation model that may explain some characteristics of our EBID samples, as the switching of the current paths for activation and re-activation of “destroyed” samples and the observation of enhancement of the conductance of selected samples by a factor of $10^6$.

In Fig. 2 a typical $I(V)$ characteristic is shown. In the left inset a voltage up to 1 V was applied to the inner contacts of the sample. After having achieved a current flow of 250 $\mu$A a rapid decrease of the current was observed. This seems to imply an irreversible destruction of the sample. However, after this irreversible change on the inner contacts was observed the $I(V)$ characteristic measured on the outer contacts showed a significant current flow. This suggests that the sample is neither destroyed nor can the current flow be homogeneous across the sample’s cross section. Arguably, within the tunnel percolation model the current path switched. By increasing the voltage another current path was established. This is shown in the main graph of Fig. 2. At about 4 V the current increases strongly. The smaller jumps in the characteristic do not cause a stable change of the current path. We noticed in many experiments that a current increase of at least three orders of magnitude tends to yield a stable current path. The right inset of Fig. 2 shows the $I(V)$ characteristic after a new current path was established. A voltage sweep up to 0.1 V now results in a current in the $\mu$A regime.

Fig. 3 shows the $I(V)$ characteristics of a sample which is apparently badly conducting. The left inset depicts the initial $I(V)$ curves of the sample in its original state. For comparison the $I(V)$ curve as measured between blank contact pads is also shown. This gives an indication of the isolation resistance of the SiO$_2$ layer. The main graph depicts the irreversible “activation” process, achieved by increasing the voltage beyond the sample-specific critical voltage $V_C$. A new current path is formed. The small peaks in the three curves are an indication for spurious path switching processes. The right inset depicts the $I(V)$ characteristic after the deposit has been “activated”. By comparison to the original $I(V)$ data it can be observed that the conductance has increased by a factor of $10^6$ at 0.1 V bias.
4. Conclusion
We found in granular W samples prepared by electron beam induced deposition clear evidence for current path switching. Activation of initially badly conducting samples as well as re-activation of apparently “destroyed” samples proved possible. We suggest that this can be qualitatively explained by a tunneling percolation model. Experiments aiming for a direct visualisation of the suggested current path are presently in progress.

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References
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