Pergamon

NanoStructured Materials, Vol. 6, pp. 783-786, 1995

0965-9773/95 $9.50 + .00

0965-9773(95)00176-X

MODELLING OF CONDUCTION IN GRANULAR METALS

A. Kolek
Department of Electrical Engineering
Rzeszów University of Technology, W. Pola 2, 35-959 Rzeszów, Poland

Abstract *- The Charging energy model of conduction in granular metals has been investigated. In this approach electrical conduction is modelled by a resistor lattice built from conductances, $G_{ij}=G_0 exp(2\chi s_{ij}+E_{ij}/kT)$. χ is the decay rate of the electron wave function in an insulator, k is Boltzmann constant and s_{ij} and E_{ij} are intergrain separation and intergrain charging energy respectively. The critical path method used to find the conductivity, σ, of the model gives the temperature dependence of $\sigma \sim exp[-(T_1/T)^{1/2}]$ in agreement with widely observed experiments. The temperature, T_1, was expressed in terms of the model's parameters, $T_1 = 6A\chi B_c/z\rho k$. Here A is the width of the distribution of tunnelling distances, B_c is the constant of order 1.5 in three dimension, z is the coordination number of the lattice and ρ is the density of grain charging energies. Numerical simulations of the model has been performed. Data have been carefully analysed. The influence of finite size of the model and preexponential terms appearing in the formula for σ has been considered. An excellent agreement between the critical path analysis and numerical data has been found in the limit $A\chi/\rho \gg 1$.*

INTRODUCTION

Granular metals are metal-insulator composites formed from small metallic and insulating grains of nanometre size. They are usually produced as films grown by co-evaporation or co-sputtering. The commonly observed feature of granular metals is a dependence of electrical conductivity, σ, on temperature, T,

$$\sigma \sim \exp[-(T_1/T)^{1/2}]. \qquad [1]$$

To explain the relation [1] charging energy model of conduction has been recently proposed (1,2). In this model nearest neighbour sites i, j in a regular lattice are connected by conductances $G_{ij}=G_0\exp(2\chi s_{ij}+E_{ij}/kT)$. Here χ is decay rate of electron wave function in the insulator, k is Boltzmann constant and s_{ij} and E_{ij} are intergrain separation and intergrain charging energy respectively. The latter can be expressed in terms of grain charging energies E_i and E_j. Grain charging energy E_i is the energy required to place an electron (charge e) on an initially neutral grain. For instance for a separate grain of diameter D_i embedded in an insulating host of dielectric constant ε we have $E_i=e^2/\varepsilon D_i$. When only hops of electrons between charged and neutral grains (what is a dominant process) are considered $E_{ij}=(E_i+E_j+|E_i-E_j|)/2=\max(E_i,E_j)$. To model randomness of the system s_{ij} and E_j are assumed to be (independent) random variables which obey distributions $N(s_{ij})$ and $\rho(E_j)$ respectively. Then $N(s)$ is supposed to be uni-

form on lattice spacing, A, $N(s)=z/A$ where z is the coordination number of the lattice. $\rho(E)$ has the shape with a flat part near $E=0$ and quickly goes to 0 when E exceeds its median value E_0. This type of $\rho(E)$ is a result of considerations which take into account both a distribution of grain sizes and random electrostatic potential which appears due to the distribution of the electrical charge (2). Monte Carlo simulations of this model has been performed on simple cubic lattice and show that, indeed, relation [1] can been observed in a limited temperature range (2). Analytical solution of the model is also possible. It was shown (3) that equation [1] is the characteristic dependence of the model and predictions for temperature T_1 upon the model's parameters has been established. The scope of the paper is to check whether theoretical and numerical results agree and thus answer the question whether analytical treatment of the model was correct. In the next section we shortly review results of analytical analysis of the model.

CRITICAL PATH ANALYSIS

Critical path analysis bases on the observation that overall conductance of any resistor network that is built from conductances, which take values distributed over several orders of magnitude, is determined by the so-called critical conductance, G_c. The latter is defined in such a way that the network's bonds with $G_{ij} \leq G_c$ form the tiniest subnetwork which percolates through the whole network. Thus for all the bonds in this subnetwork we have

$$\frac{s_{ij}}{s_m} + \frac{E_i + E_j + |E_i - E_j|}{2E_m} \leq 1, \qquad [2]$$

where $s_m=\ln(G_0/G_c)/2\chi$ and $E_m=\ln(G_0/G_c)/kT$. Thus s_m and E_m have the meanings of maximum tunnelling distance and maximum grain energy allowed for bonds and sites which form the percolating subnetwork respectively. On the other hand it is well known that to form percolating cluster it is required on average at least B_c occupied bonds per lattice site. B_c takes value which depends only on the dimensionality of the system. For example $B_c \cong 1.5$ for all 3D lattices. Thus we can find critical conductance of the network calculating the average value of occupied bonds (i.e. those that belong to the percolating subnetwork) per lattice site (1)

$$\frac{\int_0^{E_m} dE_i \rho(E_i) \int_0^{E_m} dE_j \rho(E_j) \int_0^{maxs_{ij}} ds_{ij} N(s_{ij})}{\int_0^{E_m} dE_i \rho(E_i)} = B_c. \qquad [3]$$

Above $maxs_{ij}$ is the maximum distance which satisfies condition [2] for a given set of E_i and E_j. In the temperature range for which E_m lies on the flat part of $\rho(E)=\rho$ near $E=0$ integrals on the left hand side of equation [3] can be calculated. We get $zs_mE_m\rho/3A=B_c$ which immediately leads us to the relation $G_c \sim \exp[-(T_1/T)^{1/2}]$ with the temperature

$$T_1 = 6A\chi B_c/z\rho k. \qquad [4]$$

Thus, if we note that $\sigma \sim G_c$, we show in an analytical way that the charging energy model being considered is able to explain experimental relation [1]. In the next section we present the results of Monte Carlo simulations which were performed in order to check wether or not relation [4] is fulfilled.

NUMERICAL SIMULATIONS

To test results [1] and [4] obtained in the previous section computer simulations have been carried out. The simulations were performed for the number of the model's parameters $A\chi$, and ρ. For each set of those parameters several hundred simple cubic lattices of size L=15 were generated and their conductances were computed for various temperatures. Then the data were averaged (<·>) and arranged in log<G> versus $T^{-1/2}$ plots as it is shown in figure 1a. As it can be seen, in a quite wide temperature range data follow the straight line i.e. relation [1] is indeed fulfilled. The temperature T_1 can be described from the slope of the line. To calculate T_1 only high temperature data should be used. In this temperature range different kinds of averaging give nearly the same values of averaged conductance. This means that percolation correlation length, ξ, is shorter than the size of the model. On the contrary, at very low temperatures ξ is longer than L and different averaging procedures lead to different values of <G>. This obvious observation lets us learn however that more detailed analysis of the model should include the influence of ξ on the conductance of the model. It was shown that

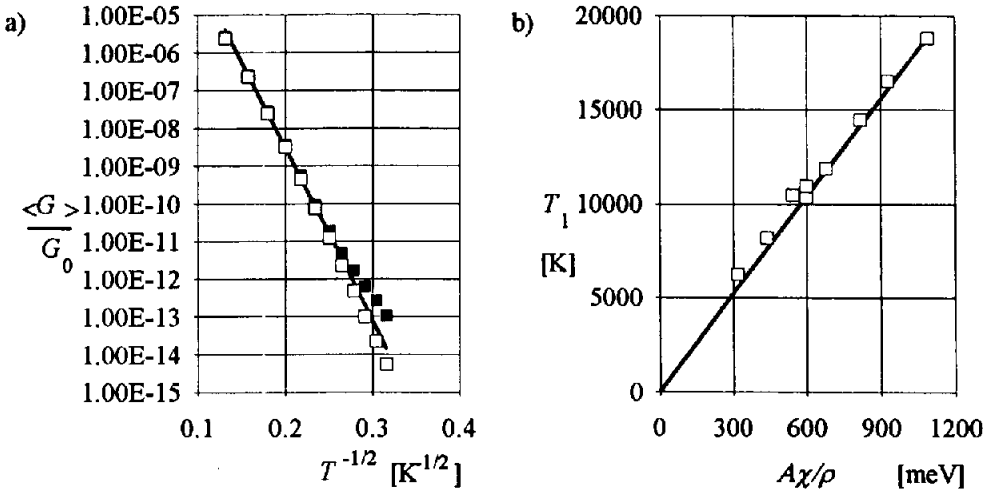

Figure 1. a) Results of numerical simulations. Data shown in the figure are for the model in which $A\chi$=15, and ρ=1/41 [meV]$^{-1}$ were used. Filled squares refer to arithmetic averaging whereas the empty ones are for harmonic averaging of conductances obtained from several hundreds realisations of the model. b) Comparison of the theoretical result of equation [4] (solid line) with numerical simulations (squares). Each point refers to temperature T_1 obtained from the least square fitting of the data like in figure 1a with equation [6]. In the fitting procedure only data for which arithmetic and harmonic averages coincide were used.

taking this effect into account results in the preexponential term emerging in equation [1]. For example in the general problem of the network in which conductances take values G_{ij}=exp(-λx) where x is the random variable uniformly sampled over 0 and 1 and $\lambda \gg 1$, the percolation correlation length scales like λ^y and thus the conductivity of the network is given by (4)

$$\sigma=\xi^{-1}G_c \sim \lambda^{-y}G_c=\lambda^{-y}\exp(-\lambda x_c), \qquad [5]$$

where x_c denotes value of the percolation threshold, i.e. the percentage of bonds required to form percolating cluster. The value of the exponent y is still an object of controversy. Whereas most theoretical works show $y=\nu$, where ν is the percolation correlation length exponent, $\nu \cong$ 0.89 (5), the most recent numerical simulations give y=0.6 (4). Our simulations give y=0.76 (6) and this value will be used in further calculations.

Let us turn back to the model being analysed. As we have shown in the previous section the network's critical conductance is given by $G_c \sim \exp[-(T_1/T)^{1/2}]$. Thus we can use $\lambda=(T_1/T)^{1/2}/x_c$ to determine correlation length in our model. We get $\xi \sim T^{-y/2}$, which eventually leads us to the conductivity of the model

$$\sigma \sim T^{y/2}\exp[(T_1/T)^{1/2}], \qquad [6]$$

and this is the relation we should fit with the data from numerical simulations. Temperature T_1 can be easily determined from the least square analysis of $\ln(\langle G \rangle T^{y/2})$ versus $T^{-1/2}$ data. Values of T_1 calculated in such a way for simulations of the model with various values of $A\chi$, and ρ are shown in figure 1b. In this figure the theoretical result of equation [4] is also displayed as the solid line with the slope of $6B_c/zk$. The agreement is excellent in the limit of $A\chi/\rho \gg 1$ which is also the condition of the application of the critical path method to solve percolation problems.

ACKNOWLEDGEMENTS

The work was supported in part by Stefan Batory Foundation. Helpful discussions with A.W. Stadler and A. Kusy are also gratefully acknowledged.

REFERENCES

1. P. Sheng and J. Klafter, Phys. Rev. B 27, 2583 (1983).
2. L.F. Chen, P. Sheng, B. Abeles and M.Y. Zhou, *Physical Phenomena in Granular Metals*, 187, MRS Symp. Proc. 195 ed G.D. Cody, T.H. Geballe and P. Sheng, Pittsburg, PA:MRS (1990). M. Zhou, P. Sheng, L. Chen and B. Abeles, Phil. Mag. B 65, 867 (1992). P. Sheng, Phil. Mag. B 65, 357 (1992).
3. A. Kolek, J. Phys.: Condens. Matter 6, 469 (1994).
4. S. Tyč and B.I. Halperin, Phys. Rev. B 39, 877 (1989). P. LeDoussal Phys. Rev. B 39, 881 (1989). A.E. Morozovskii and A.A. Snarskii, Zh. Eksp. Teor. Fiz. 104, 4059 (1993).
5. D. Stauffer, *Introduction to Percolation Theory*, Taylor&Francis, London (1985).
6. A. Kolek, A.E. Morozovskii and A.A. Snarskii in preparation