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Transport Properties of Square Lattice of Metallic Nanogranules Embeded in Insulator

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ABSTRACT

We have made theoretical studies of transport properties of square lattice of metallic nanogranules embedded insulting layers. We have developed an extension of the classical Sheng-Abeles model for a single layer of identical spherical particles located in sites of a simple square lattice with three possible charging states of granule and three kinetics processes, creation of a pair on neighbor granules, recombination of such a pair and charge translation from a charged to neighbor neutral granule. This model neglecting the effect of disorder within a layer and of multilayered structures, revealed a variety of possible kinetic and thermodynamical regimes. Effective kinetic equations for averaged charge densities were derived for the characteristics area of the granular sample, the contact areas beneath metallic currents leads and free area between these leads. From these kinetic equations, it was shown that the tunnel conduction in the free area did not produce any notable charge accumulation and the conduction regime was purely ohmic. Some conduction in the contact area became impossible without charge accumulation, leading to a generally non-ohmic conduction regime, since the contact area dominated in the overall resistance. The calculated I-V curves and temperature dependences were found in a good agreement with available experimental data and obtained theoretical results.

KEYWORDS Transport, Lattice, Nanogranules, Embedded Insulator, Disorder, Kinetic Equations, Tunnel Conduction Charge Accumulation.

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INTRODUCTION

A number of physical mechanisms still need better understanding for transport phenomena in films are still a great challenge and, at present various studies address them [1-3]. The main reason is that granular systems reveled certain characteristics which cannot be obtained either in classical conduction regime, in metallic electrolyte or gas discharge conduction or in the hopping regime in doped semiconductors or in common tunnel junctions. Their specifics are mainly determined by the drastic difference between the characteristics time of an individual tunneling event $\sim \frac{\hbar}{\varepsilon_F} \sim 10^{-5}$ seconds and the interval between such events on the same granule $\sim \frac{e}{jd^2} \sim 10^{-3}$ seconds at typical currents density $j \sim 10^{-3}$ A/cm^2 and granule diameter d ~ 5.0 nm. Other important moments are the sizable coulomb charging energy $E_C \sim \frac{e^2}{\varepsilon_{eff} d}$, typically ~ 10 meV and the fact that the tunneling rates across the layer may be even several orders of magnitude slower [4] than along it. The interplay of these factors leaded to unusual macroscopic effects including a peculiar slow relaxation of electric charge discovered in experiments on tunnel conduction through granular layers and granular films [5-6]. The specifics can be contrasted with well studied process of tunnel conduction in the variable range hopping regime [7]. The latter approach is more adequate for tunneling between atomic localized states, e.g. in doped semiconductors with shallow dopant levels where the hopping range is defined by the effective localization radius and can extend over many periods of crystalline lattice. Actually, nanostructured granular films are of a considerable interest for modern technology due to their peculiar physical properties like giant magneto-resistacne [8], coulomb blockade [9-10] or high density magnetic memory [11] properties that are impossible for continuous materials. Due to dynamical accumulation of charge and not to thermic excitation of charge carrier the conductance was obtained [12]. Kulik and Shekhter [13] presented tunnel conduction through granular media.

METHOD

We have considered a system of identical spherical metallic nanogranules of diameter d, located in sites of simple square lattice of period within a layer of thickness $b \sim a$ of insulating host with a dielectric constant ε. In the charge transfer process each granule can bear different number of σ of electrons in excess or deficit of the constant number of positive ions and the resulting excess charge σe defines a coulomb charging energy $\sim \sigma^2 E_C$.

At modulate temperatures $T \lesssim E_C / k_B$, the consideration can be limited only to the ground neutral state $\sigma = 0$ and single charged states $\sigma \pm 1$. Actually for low metal contents will separated small grains and typical granule size 'd' ~ 3 nm in a medium with effective dielectric constant $\varepsilon_{eff} \sim 25$, we estimated $E_C \sim 20\ meV$ and since the energy difference between single charged and double charged states is $3E_C$, the relative smallness of tunnel probability to this state was $\sim \exp(-3E_C / k_B T)$ and the effective temperature limitation was $T \lesssim 3E_C / k_B \sim 660\,K$. This assured the adopted single charge restriction for the whole temperature range upto at least room temperatures. For a three dimensional granular array, E_C was considered under the assumption of a constant ratio between the mean space 'S' and granule diameter 'd' in the form $E_c = e^2 f\left(s/d\right)/\left(\varepsilon d\right)$ where the dimensionless function .

$$f(z) = \frac{1}{\left(1 + \frac{1}{2z}\right)}$$

The complete dielectric response of three dimensional insulating host with the dielectric constant ε and metallic particles with the volume fraction $f < 1$ and diverging dielectric constant $\varepsilon_m \to \infty$ can be characterized by the effective value $\varepsilon_{eff} = \frac{\varepsilon}{(1-f)}$.

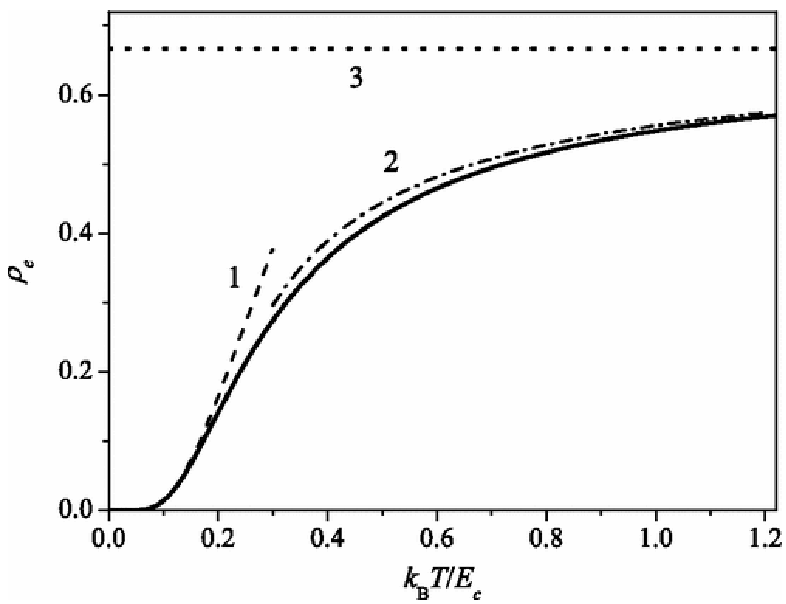
For planar lattice of granules, the analogous effective constant can be estimated, summing the energy $\frac{e^2}{(\varepsilon d)}$ of charged granule at the $n = 0$ site and energy of its interaction with electric dipolar moments $\approx \left(\frac{e}{\varepsilon_{eff}}\right)\left(\frac{d}{2n}\right)^3 n$, induced by the coulomb field from this charge in macroscopic dielectric approximation on all the granules at the sites $n = a\left(n_1, n_2\right)$.

$$E_c = \frac{e^2}{d}\left[\frac{1}{\varepsilon} - \frac{\alpha}{\varepsilon_{eff}^2}\left(\frac{d}{a}\right)^4\right] = \frac{e^2}{\varepsilon_{eff} d}.$$

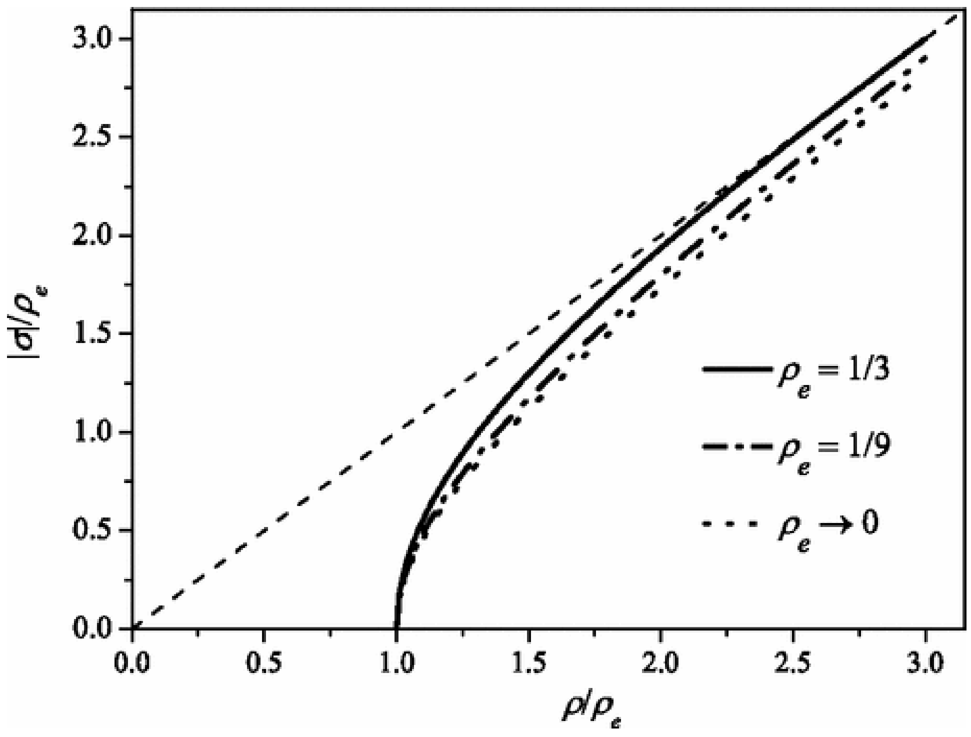
Where constant $\alpha = \frac{\pi}{4}\sum_{n \neq 0} n^{-4}$.

RESULTS AND DISCUSSION

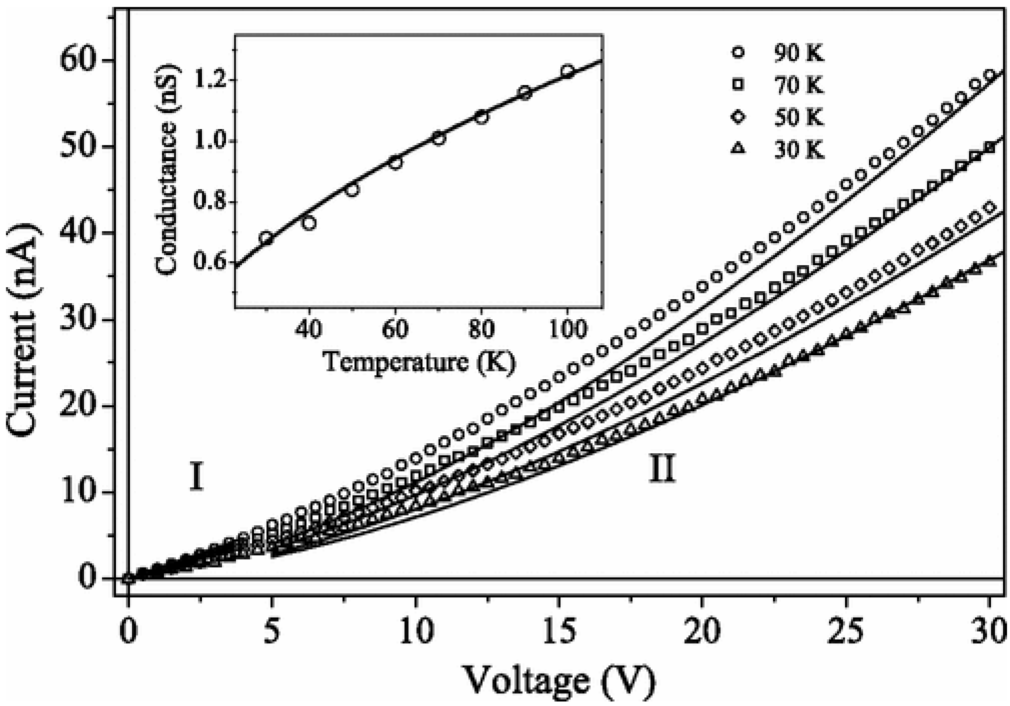
Graph (1) shows that in the presence of electric field $F_r \neq 0$ the local equilibrium is perturbed and the system generates current and generally accumulate charge. Graph (2) shows that for moderate temperatures, $T \lesssim E_C / k_B$, where the neglect of multiple charged state is justified, this dependence is reasonably close to the simplest low temperature form $\sigma = \sqrt{\rho^2 - \rho_e^2}$. It was found that the evident consequence of long range character of coulomb field in free area where possible due to screening effects by the metallic contacts and to the related short range fields. There is practically no charge accumulation and hence no diffusive contribution to the current in free area. Thus steady state of free area in out of equilibrium conditions is characterized by the ohmic conductivity g_e. The conduction in the overall system resulted from matching the considered process in contact and free area. To evaluate the global resistance of the circuit in series it is necessary to add the contributions of both areas to it. Choosing $T = 50K$ the ohmic conductance of the free area G_{FA} was calculated through the formula $G_{FA} = g(\rho_e) L'b/l \approx \omega 1.5 \times 10^{-18}$ seconds. In the contact area we have estimated the conductance using the formula $G_{CA} = \omega 8.0 \times 10^{-22}$ seconds. We have found that any choice of ω, the conductance of the contact area was about 4 orders of magnitude smaller than that of the free area and dominated the global resistance of the system. The effective value of the parameter V_0 giving the best fit to the experimental data notably higher than obtained by above formula for a single layer system. We have obtained the single value $V_0 \approx 0.5V$ whereas the bet fit for 10 layer experimental sample which is shown in graph (3). This difference was effectively accounted by the simple multiplicative factor $\alpha \approx 6$ the multilayer factor so $V_{\exp} = \alpha V_0$ assured both the agreement for regimes I and II of I-V curves and boundary $V \sim V_{\exp}$ between them, which is shown in graph (3). The obtained results were compared with previously results of theoretical and experimental works and were found in good agreement.



Graph 1: Equilibrium density ρ_e of charge carriers in the function of temperature (solid line). Curve 1 (dashed line) corresponds to the low-temperature asymptotics $\rho_e \approx 2\exp(-E_c / 2k_B T)$ and curve 2 (dashed-dotted line) to the high-temperature asymptotic $\rho_e \approx \rho_\infty - E_c / 9k_B T$, converging to the limit $\rho_\infty = 2/3$ (dotted line).



Graph 2: The charge density σ in function of the carrier density ρ for different temperatures (corresponding to different thermal equilibrium value ρ_e).



Graph 3: I-V characteristics for a granular sample at different temperatures, compared with the theoretical curves for Regimes I and II. (Inset) Temperature dependence of ohmi conductance G_0, measured data (circles) vs. calculated results.

CONCLUSION

We have studied the transport properties of square lattice of metallic nanogranules embedded in insulator. We have found that in this model with three possible charging states $\pm$ or 0 of a granule and three kinetic processes creation or recombination of pair and charge transfer between neighbor granules, the mean field kinetic theory was developed. We have also found the interplay between charging energy and temperature between applied electric field and the coulomb fields by the nano compensated charge density. The resulting charge and current distributions were found to be essentially differ in the free area, between the metallic contacts or in the contact area beneath those contacts. Thus the steady state *dc* transport is compatible only with zero charge density and ohmic resistivity. The obtained results were found in good agreement with previously obtained results.

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