Tunneling density of states of granular metals

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We investigate the effect of Coulomb interactions on the tunneling density of states (DOS) of granular metallic systems at the onset of Coulomb blockade regime in two and three dimensions ($d=2,3$). Using the renormalization group technique we derive the analytical expressions for the DOS as a function of temperature T and energy ε. We show that samples with the bare intergranular tunneling conductance g_T^0 less than the critical value $g_T^C=(1/2\pi d)\ln(E_C/\delta)$, where E_C and δ are the charging energy and the mean energy level spacing in a single grain, respectively, are insulators with a *hard gap* in the DOS at temperatures $T\to 0$. In $3d$ systems the critical conductance g_T^C separates insulating and metallic phases at zero temperature, whereas in the granular films g_T^C separates insulating states with the hard (at $g_T^0<g_T^C$) and soft (at $g_T^0>g_T^C$) gaps. The gap in the DOS begins to develop at temperatures $T^*\sim E_C g_T^0 \exp(-2\pi d g_T^0)$ and reaches the value $\Delta\sim T^*$ at $T\to 0$.

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Granular materials, a focus of the current mesoscopic physics, is a unique testing area for general concepts of disordered systems (see Refs. 1–4). One of the remarkable fundamental features of granular metals is the strong on-site Coulomb repulsion that leads to the suppression of transport at low temperatures and to an insulating ground state at low bare intergranular conductances $g_T^0<g_T^C=(1/2\pi d)\ln(E_C/\delta)$, where d is the dimensionality of the granular array, E_C and δ are the charging energy and the mean energy level spacing in a single grain, respectively. Good progress in understanding this insulating state has recently been made;[4] yet, despite the impressive advance, the satisfactory picture of the Mott (metal-insulator) transition that occurs at $g_T^0=g_T^C$, and of the physics of its vicinity, is still lacking. One of the fundamental questions that remains open is the suppression of the tunneling density of states that always accompanies Mott transition. In this Report, we develop a quantitative approach that enables us to investigate Mott transition in granular metals and derive the associated tunneling density of states in its vicinity.

The density of states (DOS) is a fundamental quantity that determines most of the properties of the system involved, and the electronic transport is a key phenomenon where the manifestations of the essential DOS features may be most pronounced. A general technique to treat transport properties of granular metals in the high temperature regime $T>g_T\delta$ was developed recently in Ref. 4. It was, in particular, shown that the conductivity of granular metals can be described in terms of the renormalized temperature dependent intergranular tunneling conductance given by the following expression

$$g_T(T)=g_T^0-(1/2\pi d)\ln[g_T^0E_C/T], \tag{1}$$

which holds as long as $g_T(T)>1$. The conductivity of the sample is related to the tunneling conductance as $\sigma(T)=2e^2g_T(T)a^{2-d}$, where a is the granule size and the factor of 2 is due to the spin. From Eq. (1) follows that at temperature

$$T^*\sim E_C g_T^0 e^{-2\pi d g_T^0}, \tag{2}$$

the renormalized conductance, $g_T(T)$, is strongly suppressed and approaches small values where renormalization group breaks down. Equation (1) is valid only at temperatures $T>g_T\delta$ (Ref. 5) whereas in the opposite case, $T<g_T\delta$, the conductance renormalization (1) is saturated and the system behaves essentially as a homogeneous disordered metal.[6] Comparing two relevant energy scales T^* and $g_T\delta$, one concludes[6] that if (i) $T^*<g_T\delta$ (or, equivalently, $g_T^0>g_T^C$) then, the renormalized conductance is still large at temperatures $T\sim g_T\delta$ and the low temperature phase of the system is similar to that of the disordered metals. Alternatively, if (ii) $T^*>g_T\delta$ (or $g_T^0<g_T^C$), the conductance of the system becomes significantly suppressed at $T\sim T^*$ reflecting thus the onset of the Coulomb blockade regime. In the latter case one expects that at $T\sim T^*$ the Coulomb gap begins to develop (reaching its maximal value at zero temperature) and, as a result, a noticeable suppression of DOS even at finite temperatures, $T\sim T^*$ occurs.

In this Report, we consider the tunneling DOS of granular metals with the bare tunneling conductance $g_T^0\sim g_T^C$ at the onset of Coulomb blockade regime at temperatures $T>T^*$ [case (ii) above]. We show that Coulomb blockade strongly suppresses the tunneling DOS, $\nu(T)$. For $3d$ granular samples $\nu(T)$ is given by

$$\frac{\nu(T)}{\nu_0}=\left[1-\frac{1}{6\pi g_T^0}\ln\frac{g_T^0E_C}{T}\right]^{3A}, \tag{3a}$$

whereas for granular films we obtain

$$\frac{\nu(T)}{\nu_0}=\left[\frac{g_T^0E_C}{T}\right]^{1/\pi}\left[1-\frac{1}{4\pi g_T^0}\ln\frac{g_T^0E_C}{T}\right]^{4g_T^0}. \tag{3b}$$

Here ν_0 is the DOS for noninteracting electrons and $A=0.253$ is the dimensionless constant. Equations (3) hold for

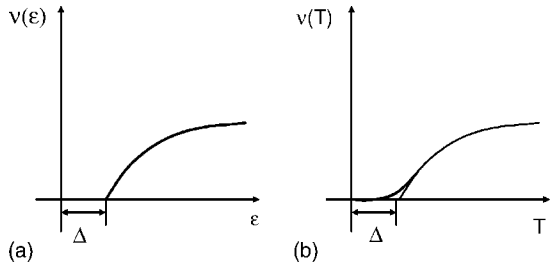

FIG. 1. Schematic behavior of the tunneling density of states as a function of (a) energy ε at zero temperature and (b) temperature T at $\varepsilon=0$.

temperatures $T>\max(T^*, g_T\delta)$ where the temperature T^* is given by Eq. (2). We note that according to Eqs. (3) the DOS vanishes exactly at the same temperature T^* as the renormalized conductance $g_T(T)$ in Eq. (1).

We show that the results (3) can be generalized to finite frequency by substitution $T\to\max\{T,\varepsilon\}$. In this case Eqs. (3) can be applied even for $T\to 0$ provided ε is larger than the characteristic energy Δ that coincides with the temperature T^*

$$\Delta \sim E_C g_T^0 e^{-2\pi d g_T^0}. \tag{4}$$

From Eqs. (3) one can see that the tunneling DOS is strongly suppressed at energies $\varepsilon\sim\Delta$. Thus, one concludes that for $g_T^0<g_T^C$ the system is an insulator at zero temperature with the Coulomb gap Δ given by Eq. (4). Although the tunneling DOS behavior $\nu(\varepsilon)$ and $\nu(T)$ in Fig. 1 in the region T, $\varepsilon<\Delta$ cannot be directly derived from our formulas [and therefore there is no reasons to expect that $\nu(\varepsilon)$ and $\nu(T)$ should be the same outside the applicability domain] in this region the behavior of the tunneling DOS can be obtained on a qualitative level with the help of physical arguments. One notes that the scale Δ represents the Mott gap. Thus, at zero temperature the tunneling DOS, $\nu(\varepsilon)$ is strictly zero for $\varepsilon<\Delta$ as shown in Fig. 1(a). At the same time the temperature dependence of the tunneling DOS, $\nu(T)$ for $T<\Delta$ must have the following activation form:

$$\nu(T) \sim \nu_0 e^{-\Delta/T}, \quad T \ll \Delta, \tag{5}$$

as shown in Fig. 1(b).

Equation (1) generalizes straightforwardly to finite frequencies by the substitution $T\to\max\{T,\omega\}$; this allows us to relate the frequency dependent conductivity, $\sigma(\omega)$ with the tunneling density of states at zero temperatures, $T=0$: For $3d$ granular samples we obtain the following scaling relation:

$$\frac{\nu(\omega)}{\nu_0} = \left[\frac{\sigma(\omega)}{\sigma_0}\right]^{3A}, \tag{6a}$$

where $\sigma_0=2e^2 g_T^0 a^{2-d}$ is the high temperature conductivity with e being the electron charge. For granular films we get the following expression:

$$\frac{\nu(\omega)}{\nu_0} = \left(\frac{\Delta}{\omega}\right)^{1/\pi}\left(\frac{e\sigma(\omega)}{\sigma_0}\right)^{4g_T^0}, \tag{6b}$$

where $e\approx 2.7182$. Equations (6) are useful for the comparison of our predictions with the experimental data.

Now we turn to the quantitative description of our model and derivation of Eqs. (3): We consider a d-dimensional array of metallic grains. The motion of electrons inside the grains is diffusive and they can tunnel between grains. We assume that in the absence of the Coulomb interaction, the sample would be a good metal.

The system of weakly coupled metallic grains is described by the Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{H}_c + \hat{H}_t. \tag{7a}$$

The term $\hat{H}_0$ in Eq. (7a) describes noninteracting isolated disordered grains. The term $\hat{H}_c$ describes the Coulomb repulsion and is given by

$$\hat{H}_c = \frac{e^2}{2}\sum_{ij}\hat{n}_i C_{ij}^{-1}\hat{n}_j, \tag{7b}$$

where i stands for the granule number, C_{ij} is the capacitance matrix, and $\hat{n}_i$ is the operator of electrons number in the i-th granule. The last term on the right-hand side, (rhs) of Eq. (7a) is the tunneling Hamiltonian

$$\hat{H}_t = \sum_{ij,p,q} t_{ij} a_{i,p}^{\dagger} a_{j,q}, \tag{7c}$$

where $a_{i,k}^{\dagger}(a_{i,k})$ are the creation (annihilation) operators for an electron in the state k of the i-th grain and t_{ij} is the tunneling matrix element corresponding to the points of contact of i-th and j-th granules.

As it was shown in Ref. 5, at temperatures $T>g_T\delta$ the model given by Eq. (7) can be effectively described in terms of the functional proposed by Ambegaokar, Eckern, and Schön (AES) in Ref. 7

$$S = S_c + S_t, \tag{8a}$$

where S_c is the charging part of the action

$$S_c = \sum_{ij}\int_0^{\beta} d\tau \frac{d\phi_i}{d\tau}\frac{C_{ij}}{2e^2}\frac{d\phi_j}{d\tau}, \tag{8b}$$

and the second term on the right-hand side of Eq. (8a) describes tunneling between the granules

$$S_t = 2\pi g_T \sum_{ij}\int_0^{\beta}\frac{T^2 d\tau d\tau'}{\sin^2[\pi T(\tau-\tau')]}\sin^2\left[\frac{\phi_{ij}(\tau)-\phi_{ij}(\tau')}{2}\right]. \tag{8c}$$

Here $\phi_{ij}(\tau)=\phi_i(\tau)-\phi_j(\tau)$ is the difference between the phases of the i-th and j-th granules. In the metallic regime we may neglect winding numbers in the phases. We use the renormalization group technique to calculate DOS. The charging part of the action, S_c in Eq. (8a) determines the upper frequency cutoff.

In terms of the functional approach based on the action (8a), DOS is expressed as[4]

$$\frac{\nu(\omega)}{\nu_0} = T\,\mathrm{Im}\int_0^\beta \frac{d\tau e^{i\omega_n\tau}}{\sin \pi T\tau}\langle e^{-i[\phi_i(\tau)-\phi_i(0)]}\rangle_{\omega_n\to -i\omega}, \qquad (9)$$

where analytical continuation from bosonic Matsubara frequencies $\omega_n = 2\pi Tn$ is assumed on the rhs of Eq. (9). Following the standard renormalization group procedure we separate the field ϕ into slow, ϕ_s and fast, ϕ_f parts and integrate over the fast field in Eq. (9). The fast fields belong to the infinitesimal volume dS in the configuration space of the quasimomentum, $\mathbf{q}$ and frequency ω that represents the energy shell $d\Lambda$. Using the one-loop approximation we obtain the following RG equation for the conductance g_T:[4]

$$\frac{dg_T(\Lambda)}{d\Lambda} = \frac{1}{2\pi\Lambda d}. \qquad (10)$$

Performing the integration in Eq. (10) we obtain Eq. (1). With the same accuracy for the flow equation of the density of states we get

$$d\ln(\nu/\nu_0) = a^d\int_{dS}\frac{d\omega}{2\pi}\frac{d^dq}{(2\pi)^d}G_\phi(\omega,q). \qquad (11)$$

Here the Green's function $G_\phi(\omega,q)$ of the phase fields ϕ is defined on the scales Λ

$$G_\phi(\omega,q) = \frac{1}{2g_T(\Lambda)}\frac{1}{|\omega|E_q}. \qquad (12)$$

In Eq. (12) we introduced the notation $E_q = 2\Sigma_{\mathbf{a}}[1-\cos(\mathbf{qa})]$ with $\{\mathbf{a}\}$ being the lattice vectors. For simplicity we assume the periodic arrangement of grains. The integration in Eq. (11) is going over the infinitesimal volume dS in the $(\omega,\mathbf{q})$ configuration space that corresponds to the energy interval $d\Lambda$. The proper way to chose a particular form of dS depends on the dimensionality of the sample: For $3d$ samples the integrals over the quasimomentum converge and one can simply choose $dS = (2\pi/a)^3 d\Lambda$. This leads to the following differential equation:

$$d\ln(\nu/\nu_0) = \frac{A}{2\pi}\frac{d\Lambda}{\Lambda g_T(\Lambda)}, \qquad (13)$$

where $A = a^3\int d^3q/(2\pi)^3 1/E_q \approx 0.253$ is the numerical constant. Integrating over Λ in Eq. (13) in the range $(T, g_T^0 E_C)$ we obtain Eq. (3a) for DOS of the $3d$ granular metals.

The $2d$ case is different since the direct integration over the quasimomentum, $\mathbf{q}$ in Eq. (11) would lead to the infrared divergence. In this case it is natural to introduce the infinitesimal volume, dS in the following way:

$$\int_{dS} d\omega d^2q = \int d\omega d^2q\,\delta(|\omega|E_q - \Lambda)d\Lambda, \qquad (14)$$

such that on the energy shell Λ the propagator (12) will not be divergent since $\omega E_q = \Lambda$. This complication does not manifest itself in the renormalization equation for conductance (10) since the conductance renormalizations come only from distances of the order of the grain size.[4] Using Eq. (14), performing the integration over the ω, q in Eq. (11) and taking into account the fact that the upper cutoff for frequency, ω is $g_T E_C(q)$, where for $2d$ granular samples the charging energy, $E_C(q)$ is given by the expression $E_C(q) = \pi E_c/qa$, with the logarithmic accuracy we obtain the following equation:

$$d\ln(\nu/\nu_0) = \frac{1}{4\pi^2}\frac{1}{\Lambda g_T(\Lambda)}\ln\left[\frac{g_T E_C}{\Lambda}\right]d\Lambda. \qquad (15)$$

Integrating Eq. (15) over the variable Λ in the interval $(T, g_T E_C)$ we obtain DOS as given by Eq. (3b) for $2d$ granular samples. Equation (3b) was derived for long range Coulomb interaction. For short range Coulomb interaction (this may happen due to the presence of external screening) the upper cutoff for ω-integration in Eq. (11) is q-independent and is given by $g_T E_C$. In this case the extra factor $1/2$ will appear in the right-hand side of Eq. (15) and as a consequence the final result for the DOS in Eq. (3b) will be modified as $\nu(\omega)/\nu_0 \to [\nu(\omega)/\nu_0]^{1/2}$. In the limit of large tunneling conductance, $g_T^0 \gg g_T^C$ one reproduces the result of Ref. 4 for the tunneling DOS of granular metals.

Although our theory applies to $1d$ granular arrays, the results in this case should be taken with caution when compared with experimental data: the problem is that the conductivity of $1d$ system is usually controlled by the weakest junction and thus must be described by the conductance distribution function[8] rather than by the average value of the conductance. Thus even small fluctuations of conductance could be important in $1d$ case, especially close to the predicted transition at $g_T^0 \sim g_T^C$; we, however leave the detailed analysis of this situation to the forthcoming publication.

In conclusion, we have investigated the effect of Coulomb blockade on the tunneling DOS of granular metals in the limit of large tunneling conductance between the grains. We have determined the critical value of tunneling conductance $g_T^C = (1/2\pi d)\ln(E_C/\delta)$ below which the granular metal becomes an insulator with a "hard" gap at zero temperature. For $3d$ samples this value of critical conductance corresponds to a metal-insulator transition, as granular samples with $g_T^0 > g_T^C$ are metallic at zero temperature.[6] This value of $g_T^C = (1/2\pi d)\ln(E_C/\delta)$ explains the long known puzzling fact that in $3d$ systems the metal insulator transition occurs at $g_T \approx 0.1$.

The situation is different for $2d$ granular systems since in this case even samples with $g_T^0 > g_T^C$ are insulators at temperatures $T \to 0$ due to interaction and quantum effects,[6] similar to those that take place in homogeneously disordered metals.[9–11] Nevertheless even in $2d$ case the critical value of conductance g_T^C represents the boundary between two physically different regimes at temperatures $T \to 0$: Samples with $g_T^0 < g_T^C$ represent the "hard" insulators, with a hard gap in the DOS, while samples with $g_T^0 > g_T^C$ are insulators with a soft gap in the DOS similar to homogeneously disordered metals.

Our results have a logarithmic accuracy, i.e., the appearing in the theory logarithm should be large. The fact that the critical tunnelling conductance g_T^C can be even less than one for realistic values of ratio E_c/δ due to extra coefficient $1/2\pi d$ does not invalidate our theory as long as $\ln(E_c/\delta) \gg 1$.

We hope that our analytical results for tunneling DOS of granular metals given by Eqs. (3) will stimulate further experiments on transport and thermodynamic properties of granular metals.

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