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Memory effects in transport through a hopping insulator: Understanding two-dip experiments

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We discuss memory effects in the conductance of hopping insulators due to slow rearrangements of many-electron clusters leading to formation of polarons close to the electron hopping sites. An abrupt change in the gate voltage and corresponding shift of the chemical potential change populations of the hopping sites, which then slowly relax due to rearrangements of the clusters. As a result, the density of hopping states becomes time dependent on a scale relevant to rearrangement of the structural defects leading to the excess time-dependent conductivity.

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I. INTRODUCTION

Memory effects in low-temperature transport properties of hopping insulators have been reported in several systems.[1–4] After excitation from equilibrium by, e.g., a sudden change in gate voltage, V_g, the conductance of the system increases, independent of the sign of the change. This excess conductance, $\Delta\sigma$, may persist for long times after the excitation forming the so-called memory cusp; see Ref. 5 for a review.

Several concepts were used to explain the memory cusps in the dependence of the conductance, G, versus the gate voltage, V_g. The so-called *intrinsic* mechanism is based on the assumption that the memory effects are due to complex dynamics in the strongly correlated system of interacting electrons.[6–8] It is a natural assumption since hopping insulators lack strong metallic screening and the long-range Coulomb interaction can be decisive. In Ref. 9 the connection between the glasslike behavior and Coulomb gap was argued. Another scenario, the so-called *extrinsic*, assumes that the observed conductance relaxations are due to the influence of slowly relaxing atomic configurations acting on the conducting channels. It was first advocated in Ref. 10 to explain the occurrence of a $G(V_g)$ cusp in granular gold films.

The mechanism behind the memory effects in hopping insulators is far from being fully understood. In particular, we are not aware of a fully convincing explanation of the experimental results[5] on the "double-dip" structure of the $G(V_g)$ dependences and their relaxations. Several dips can be grown in the dependence of conductivity of the gate voltage by the application of different gate voltages for a long time. Recent experiments[11] aimed at studies of the influence of the sample lateral dimensions on the glassy properties show that there are reproducible conductance fluctuations having apparently different time scales comparing with the memory cusps.

Recently we suggested a simple extrinsic model allowing for the "two-dip" behavior of the conductance of a structurally disordered hopping insulator.[12] According to this model, the memory is supported by two-state dynamic structural defects present in any medium with sufficient amount of structural disorder.[13] The two-level defects get polarized by the electrons and, in turn, form a *polaron gap* at the hopping sites decreasing hopping conductance. The slow dynamics of conductance is then due to slow rearrangement of polaron clouds around the hopping sites. An important feature of this extrinsic model qualitatively explaining logarithmic relaxation and memory effects[3,4] is the presence of a set of fluctuators possessing dipole moments, uniform density of states, and logarithmically uniform spectrum of relaxation times. Such fluctuators can also have intrinsic electronic nature.[14] Recent experiments[5,11] can help determine which particular mechanism—"extrinsic" or "intrinsic"—is responsible for the observed behavior using the temperature dependence of the "typical" relaxation time defined by the memory deep equilibration. We will show that the "chessboard" electronic fluctuators suggested in Ref. 15 to interpret $1/f$ noise in hopping conductivity also lead to the nonequilibrium conductivity behavior similar to that observed experimentally. As we will see below, this time is sensitive to the minor deviations of the relaxation-time distribution from its $\propto 1/\tau$ shape. The model[15] turns out to be able to explain the observed *increase* in the typical relaxation time with temperature, as well as some other observed features.

II. MAIN CONCEPTS

To calculate the time-dependent conductance of the system, one needs to know the time-dependent density of hopping states (DOS) at the Fermi level. The DOS is modified by rearrangement of the populations of the sites neighboring the sites belonging to the hopping cluster. This rearrangement driven by hopping electrons leads to decrease, U, in the energy of a hopping site. The polaron shifts U are different for different sites and can be characterized by a distribution function, $\mathcal{F}(U,t)$. This distribution slowly depends on time due to rearrangement of the polaron clouds.

The analysis below is similar to our previous work.[12] A site can be brought from the ground to an excited state by placing or removing an electron that can take place only if the excitation energy, ε, exceeds the polaron gap. Thus the formation of the polaron gap excludes all the states with $U \geq \bar{\varepsilon}$. Here $\bar{\varepsilon}$ is the typical excitation, its equilibrium value

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being the width of the hopping band, ε_h. For an equilibrium state created by sudden change $\delta V_g \gg \varepsilon_h$ of the gate voltage $\bar{\varepsilon} \approx \delta V_g$. Thus, $\bar{\varepsilon} \sim \max\{\varepsilon_h, \delta V_g\}$. The effect of the polaron cloud on the conductivity can be estimated as

$$\frac{\delta G(t)}{G} \sim -\int_{\bar{\varepsilon}}^{\infty} \mathcal{F}(U,t)dU. \tag{1}$$

This result is straightforward—the sites inside the polaron cloud cannot be occupied, and the density of states is *less* than the universal one. Thus the relative decrease in the density of states due to polarons can be estimated as a relative volume occupied by the polaron cloud.

To find $\mathcal{F}(U,t)$ one needs to specify the slow relaxing aggregates producing the polaron shift U. Following previous studies of $1/f$ noise in hopping conductivity,[15] we assume that slow dynamics is due to chessboard electroneutral clusters having $2N$ sites placed at a distance $\approx r$ between each other. The relaxation rate, τ, of a given aggregate depends both on the number of sites, $2N$, in the aggregate and on the typical distance, r, between the sites and energy difference E between its lowest-energy levels. We are interested in the domain where $E \lesssim T$ since in the opposite case the cluster resides in its ground state. Let us define the distribution $P(N,r,E)$ such that $P(N,r,E)dN\,dr\,dE$ is the number of the clusters per unit volume having the parameters within the region $(N+dN, r+dr, E+dE)$. To estimate this distribution, we will take into account that the typical aggregate volume is Nr^3, while the energy bandwidth for small energies is equal to $\sqrt{N}e^2/\kappa r$. Thus, the *total* density of states, $\mathcal{W}(N,r,E)$, of all aggregates with the intersite distances r' larger than some r is given as $\mathcal{W}(N,\rho,E) \sim \lambda^N / T_0 N^{3/2} a^3 \rho^2$. Here λ is a probability to add additional pair of sites to the aggregate; $\rho \equiv r/a$, where a is the localization length, while $T_0 \equiv e^2/\kappa a$. In this way we get the expression for the partial density as $P(N,\rho) \sim \lambda^N / T_0 N^{3/2} a^3 \rho^2$. Since we are interested in the case $E \lesssim T \ll T_0$ the partial density is E independent. Let us first consider the Efros-Shklovskii (ES) regime[16] of variable range hopping (VRH). In this case the relaxation rate for cluster rearrangement can be expressed by the interpolation formula[15]

$$\tau^{-1}(N,\rho) = \nu_0 (e^{-N^{2/3}\xi^2/\rho} + e^{-N\rho}), \tag{2}$$

where $\xi \equiv (T_0/T)^{1/2}$. The first contribution corresponds to formation of a "domain wall" in the aggregate, while the second one corresponds to coherent tunneling transitions leading to recharging of all aggregate sites. The distribution of relaxation rates can be calculated as

$$\mathcal{P}(\tau) = \int dN\, d\rho\, P(N,\rho)\,\delta[\tau(N,\rho) - \tau]. \tag{3}$$

As shown in Ref. 15, the integral is dominated by the values $N=N_c$ and $\rho=\rho_c$, where

$$N_c(\tau) = [\ln(\nu_0\tau)/\xi]^{6/5}, \quad \rho_c(\tau) = \xi/[N_c(\tau)]^{1/6}. \tag{4}$$

The quantities N_c and ρ_c characterize the most important clusters among those switching during the time $\sim \nu^{-1}$. Since the number of electrons in cluster N_c depends logarithmically on all relevant parameters and it cannot be very large because other relevant parameters are exponentially sensitive to it, we set $N_c \sim 1$ following Ref. 15. Substituting Eq. (4) into Eq. (3) we obtain

$$\mathcal{P}(\tau) \sim \frac{P_0}{\tau} \frac{1}{(\nu_0 \tau)^{\alpha}}. \tag{5}$$

Here $P_0 \equiv 1/(T_0 a^3 \xi^3)$ and $\alpha(\tau) \sim \xi^{-6/5} \ln^{1/5}(\nu_0\tau) \ll 1$.

Note that at $\delta V_g \gg \varepsilon_h$ the typical distance $R \approx e^2/(\kappa \delta V_g)$ corresponding to the polaron shift $\sim \delta V_g$ produced by a nearest neighbor turns out to be less than the hopping length $r_h = a\xi$, as well as a typical size $\sim r_h$ of the aggregate. Consequently one can treat the interaction between the site belonging to the percolation cluster and a fluctuator as a contact one. Thus, $U(R) \sim e^2/\kappa R$, where R is the distance between the hopping site and its nearest neighbor belonging to the fluctuator. As result, the contribution of the clusters with relaxation time τ to the distribution of polaron shifts is

$$\mathcal{F}_\tau(U) = \frac{8\pi R^2 \mathcal{P}(\tau)}{d \ln U/dR} = \frac{8\pi e^6}{\kappa^3 U^3}\mathcal{P}(\tau). \tag{6}$$

Here we have taken into account that only aggregates with $E < U$ form the polaron as well as the fact that each of the $2N$ sites of the aggregate can be coupled to the hopping site. The proper distribution $\mathcal{F}(U,t)$ is determined by the manipulation protocol. For example, if the system is brought to some state at time t_0 by a sudden change in the gate voltage then the polaron clouds are formed by all the fluctuators, which have changed their states by the observation time, t. Consequently, $\mathcal{F}(U,t) = \int_{t_0}^{t} \mathcal{F}_\tau(U) d\tau$.

III. DISCUSSION

By now we were discussing the ES regime of VRH. One can expect that the number of the metastable aggregates strongly decreases within the Mott regime. Indeed, aggregates are constructed from the sites where the intersite Coulomb energies are of the order of single-particle energies. This is not the case for typical hopping sites in the Mott VRH regime where the spread in the energies of the localized states is rather due to extrinsic disorder than to Coulomb interaction. Because of this spread it is less probable to find a set of sites forming a two-state aggregate. The addition factor entering the probability λ for adding a pair of sites to an aggregate can be estimated as the ratio of the width of the Coulomb gap, $\Delta_C \sim T_0^{3/2} T_M^{-1/2}$, to the typical hopping band in the Mott regime, $\varepsilon_M = T_M^{1/4} T^{3/4}$. Here T_M is the characteristic temperature of the Mott VRH, $\sigma \propto e^{-(T_M/T)^{1/4}}$; we have defined Δ_C as the temperature of crossover between regimes of ES and Mott VRH. The ratio $\Delta_C/\varepsilon_M = (\Delta_C/T)^{3/4}$ is an additional factor entering the probability λ. Deeply in the Mott regime, $T \gg \Delta_C$, this factor is small. Since $T_M \propto 1/g_0$ where g_0 is the Mott density of states Δ decreases with decrease in g_0.

To compare our prediction with experimental results of Refs. 3, 5–7, 11, 17, and 18 obtained using InO films, we assume that in the absence of Coulomb interaction their DOS, g_0, would be energy independent at the energies less

than or of the order of both room temperature, T_r, and the shift in the chemical potential, $\delta\mu$, due to variation in the gate voltage, δV_g, within the dip. This DOS consists of localized and extended states split by the mobility edge, ε_m. The closer μ is to ε_m, the larger the localization length is and, consequently, the hopping conductance. Different samples have different g_0 and different μ with respect to ε_m. This picture is conventional for materials with large amount of disorder.

The authors of Ref. 7 determine the carrier concentration from the Hall coefficient at $T=T_r$. Then the found concentration, n_r, is just the concentration of the extended carriers, and one can estimate DOS as $g_0 \approx n_r/T_r$. The width of the dip is related with the shift in chemical potential as $\delta V_g = g_0 \delta\mu/C \approx \delta\mu$, where C is capacitance. Thus the width of the dip is $\propto g_0$ and, for a given position of the chemical potential, it is correlated with n_r found in Ref. 7. On the other hand, the samples with the same g_0 but different positions of the chemical potential have different resistance—the lower μ the lower is the localization length and the larger is the resistance. From a phenomenological point of view, the samples with the same g_0 but larger resistance can be characterized as "more dirty." For density of states $g_0=(4\times 10^{18}\ \text{cm}^{-3})/(300\ \text{K}) \sim 10^{32}\ \text{cm}^{-3}\ \text{erg}^{-1}$, which roughly corresponds to the threshold of the memory effect in Refs. 5 and 7, the Coulomb gap can be estimated as $\varepsilon_C = (g_0 e^6/\kappa^3)^{1/2} \sim 3\times 10^{-15}$ erg. At the same time, $T=4$ K corresponds to a crossover between the Mott and ES VRH regimes, and at this temperature $\xi \sim 5$ and $\varepsilon_h \sim \varepsilon_C$. Thus, we explain the observed correlation of Refs. 5 and 7 between pronounced decrease in the memory effect and the decrease in the dip width by the decrease in the density of the metastable aggregates in the Mott regime.

According to Eq. (1) the relative change in the conductance is $\propto \bar{\varepsilon}^{-2}$. Thus the shape of the dip can be cast in an interpolation formula,

$$\frac{\delta G}{G} \sim -\frac{(e^2/\kappa)^3 P_0 Q(t)}{(C\delta V_g/g_0)^2+\varepsilon_h^2}, \quad Q(t) \equiv \int \frac{d\tau(\nu_0\tau)^{-\alpha}}{\tau}, \tag{7}$$

where the limits of integration are determined by the manipulation protocol. The temperature dependence of the dip magnitude is given by the product $\xi^{-9/5}\varepsilon_h^{-2} \propto T^{-0.1}$. Showing the same trend as in experiment[5] it is still much weaker. Assuming $\ln \nu_0\tau \sim 20$, we estimate the height of the dip as $\xi^{1/5}(\ln \nu_0\tau)^{-6/5} \sim 0.05$ that is in agreement with experiments.

To analyze time dependence of the dip, we take into account that the parameters of the system (such as concentration and localization length) are somewhat different for different gate voltage. It is clearly demonstrated by the fact that the $G(V)$ curves have a systematic slope (subtracted in the course of studies of the dip). Let us for simplicity assume that the bonding parameter ξ depends on the gate voltage, say, through the localization length. As it is seen from experiment, G increases with increase in concentration, i.e., with V_g. Consequently, we can assume that ξ decreases with increase in V_g. Correspondingly, the parameters of the aggregates also depend on V_g and are different for the aggregates responsible for different dips in the two-dip experiment.

In the well-known double-dip experiments[5] the typical relaxation time is defined as follows. First, the gate voltage in a gated sample is rapidly changed from some initial to some final value V_{g1} (we have denoted the time of this variation as $\tau_{\min}$). Then it is kept constant until some time, which we will denote as $t_{\max}$. During this time the conductivity slowly (apparently logarithmically) decreases to some value, $G_0 - \delta G(t_{\max},\tau_{\min})$. Then the gate voltage is swept to some other value, V_{g2}, and kept constant, the conductivity decreasing with time forming a new dip, $G_0 - \delta G(t,\tau_{\min})$. Here with a logarithmic accuracy we ascribed the same estimate $\tau_{\min}$ for the fast process of switching from V_{g1} to V_{g2}. Let us assume that the shift of μ due to the variation in V_g is less than the typical single-particle energies of the sites forming the aggregates. Then at the new value of V_g the aggregates responsible for the polaron gap at $V_g=V_{g1}$ stay at the same configuration of the occupation numbers. However, the occupation of the sites forming the percolation cluster at $V_g=V_{g1}$ at $V_g=V_{g2}$ is completely changed provided that the shift of the chemical potential is larger than ε_h. Thus the aggregates responsible for the first dip start to relax. However at the times $t<\tau_{\max}$ the slow aggregates still preserve the configuration corresponding to $V_g=V_{g1}$; thus the first dip is partly restored if the gate voltage is returned to the value V_{g1}. The depth of the restored dip at the time t is expected to be $\delta G(\tau_{\max},t)$. The relaxation time, $\bar{\tau}$, is defined according to equality of the depths of the "old" and "new" dips. This condition corresponds to the equality $\bar{\tau}$ is calculated according to the following procedure:

$$\int_{\tau_{\min}}^{\bar{\tau}} \mathcal{P}(\tau)d\tau = \int_{\bar{\tau}}^{\tau_{\max}} \mathcal{P}(\tau)d\tau. \tag{8}$$

Since $\mathcal{P}(\tau) \approx \text{const}\times \tau^{-1}(\nu_0\tau)^{-\alpha}$ Eq. (5) then up to the lowest approximation in α, $\bar{\tau}_0 = \sqrt{\tau_{\max}\tau_{\min}}$. The next iteration provides the correction $\bar{\tau}_1/\bar{\tau}_0 = -\frac{\alpha}{8}\ln^2(\tau_{\max}/\tau_{\min})$, which leads to decrease in $\bar{\tau}$ with temperature increase. However, it is more sensitive to the possible dependence of the parameter α on the *gate voltage*. The first (initial) dip and the second one correspond to different gate voltages and one can expect that they are formed by the states with different localization lengths. Consequently, the values of α are different. Denoting them as $\alpha_{1,2}$ for the first and the second dips, respectively, and assuming that $|\alpha_1-\alpha_2| \ll \alpha$ we arrive at the second temperature-dependent correction $\bar{\tau}_2/\bar{\tau}_0 \approx [(\alpha_1-\alpha_2)/4\alpha]\ln(\nu_0\tau_0)(\tau_{\max}/\tau_{\min})^{\alpha/2}$. As follows from experiments[7,17] the conductance for the second dip is larger than for the first one, which indicates the smaller value of ξ for the second dip. Since $\alpha \propto \xi^{-6/5}$ one concludes that $\alpha_1 > \alpha_2$. Thus the correction $\bar{\tau}_2$ is *positive* and increases with temperature both due to increase in α and ν_0. This trend can qualitatively explain the observed weak increase in $\bar{\tau}$ with temperature. A similar conclusion can be made for another protocol[5,18] for determining a typical relaxation time. As we have seen, increase in resistance or ξ is correlated with decrease in the exponent α and subsequently slowing down the time evolution. This can be a qualitative explanation of the observation in Ref. 3, slowing down the time evolution with increase in disorder.

Now let us discuss an effect of external magnetic field.[5] One can imagine two possible mechanisms: (i) shrinkage of the wave function manifesting itself as a positive addition $\propto H^2$ to the hopping exponent[16] and (ii) spin effect related to a presence of doubly occupied centers. In the latter case the magnetic field aligns spins of singly occupied sites, which blocks the spin-conserving hops between the singly occupied sites. One can expect that at $\mu g H \ll \varepsilon_h$ the latter mechanism leads to a positive magnetoresistance, its magnitude being proportional to the relative contribution of the doubly occupied sites. Both mechanisms can be accounted for by a field dependent increase in the tunneling length ρ entering the second item on the right-hand side of Eq. (2). That would, in turn, lead to the decrease in the exponent α.

In the experiments,[3] the observed magnetoresistance is only weakly dependent on the magnitude of resistance and decreases with resistance increase. This behavior seems to be contradictory to the wave shrinkage mechanism since in that case the magnetoresistance would dramatically increase with an increase in the hopping exponent ξ. In addition, the shrinkage effect is expected to be small for materials with small localization length. Thus, it is the spin mechanism that probably dominates.

Interestingly, the double-dip memory effects are not observed in standard semiconductor materials. We believe that the reason is that the ES regime of variable range hopping (VRH) in such materials either occurs at very low temperatures (less than a few Kelvins), which implies weak heat withdrawal or corresponds to very large resistances. Both of these factors seem to be disadvantageous for typical memory experiments.

To conclude, our model qualitatively explains the following experimentally observed features of the memory effect: (i) double-dip behavior of the conductance as a function of gate voltage, (ii) suppression of the above phenomenon at small carrier concentrations due to possible crossover to the Mott regime of VRH, (iii) rather counterintuitive slowing down of the time evolution (expressed through the effective relaxation time $\overline{\tau}$) with temperature increase, (iv) slow power-law relaxation tending to logarithmic with increase in resistance, (v) qualitative dependences of memory dips on temperature and electron concentration, and (vi) slowing down the relaxation with increase in external magnetic field and degree of disorder characterized by increase in resistance.

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