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Fluctuator model of memory dip in hopping insulators

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Abstract We show that the non-equilibrium dynamic in two-dimensional electron glasses close to metal-dielectric transition is sensitive to electric fields confinement inside the sample, which leads to a nearly thermally activated conductance behavior and a strong non-equilibrium conductance response to the gate voltage, including a memory dip in a field dependence of conductance.

1 Introduction

Low temperature universal behaviors of dielectric constant in amorphous solids and DC conductivity in doped semiconductors can be used for highly sensitive low temperature thermometry (see Review [1] and references therein). For instance the temperature raise in ion-implanted silicon Si:P:B semiconductor thermistors (bolometers) detected via the change in conductance and caused by the cosmic x-ray absorption can be used to accurately measure the energy of corresponding photon. Absorption of an x ray increases the temperature of the semiconductor and this increase is detected via the conductance change. [2,3] The performance of bolometers is limited by non-equilibrium "glassy" slow dynamics including slow relaxation and $1/f$ noise. In this paper we discuss the common effects in slow relaxation existing both in amorphous solids and some semiconductors and suggest the general mechanism of these phenomena. Understanding of the nature of non-equilibrium dynamics can be useful to find the ways to reduce its destructive outcomes.

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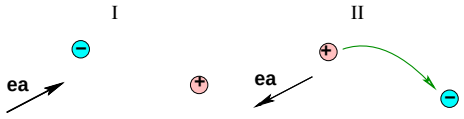

Fig. 1 Reduction of charge carrier DOS due to the interaction of electron (blue circle with "−" sign inside) and configurational transition, i. e. dipolar fluctuator, shown by the black arrow and having a dipole moment *ea*. If the interaction of fluctuator and electron is very large, the escape of electron in a direction shown by the blue arrow, must be accompanied by the simultaneous transition of the fluctuator (state I to state II). This "clustering" effect eliminates the electron contribution to the system conductance.

Slow relaxation in glassy materials is associated with transitions between local minima in configurational space separated by high potential barriers. Reasonably broad distribution of transition barrier heights and lengths results in a logarithmic relaxation because transition rates depend on these parameters exponentially.

At low temperature $(0-20\mathrm{K})$ this logarithmic relaxation is observed in a variety of disordered materials. Particularly, a sudden application of an external electric field to amorphous dielectrics results in a fast increase of dielectric constant with its subsequent logarithmic relaxation back to equilibrium [4,5]. Similar behaviour of conductance is observed after sudden application of a gate voltage in certain two-dimensional hopping insulators including indium oxide [6,7,8], ultrathin films of Bi and Pb [9], granular aluminum [10,11,12] and nickel [13].

Theoretical model, based on the Efros-Shklovskii Coulomb gap theory, [14] was proposed by one of the authors in 1995 [15] to describe the slow non-equilibrium dynamics in amorphous solids induced by the DC field application. [4,5] The attempt to extend this model to the Coulomb glasses has been made in Refs. [16,17]. Below we briefly review these old works and discuss the possible improvements in theory associated with the low-dimensional field confinement in quasi- two-dimensional samples. We show that this confinement effect can remarkably enhance the strength of the non-equilibrium system response.

2 Model of fluctuators

To characterize the non-equilibrium dynamics one has to model configurational transitions. In dielectric glasses at low temperature relaxation and aging phenomena are associated with two-level systems [18] (TLS) formed by tunneling transitions of atoms or groups of atoms between close energy minima separated by varying potential barriers. TLS possess a universal statistics with respect to their energies, E, and relaxation times, τ,

$$P(E,\tau)=\frac{P_0}{\tau}. \tag{1}$$

TLS interact with conducting electrons because they possess some dipole moment μ.

In a more complicated case of electronic glass one can introduce the model of fluctuators [16,17] possessing TLS statistics Eq. (1) and representing local system tunneling rearrangements between close energy minima. Since the transition rate grows exponentially with the number of entities participating in the transition one can expect that the number of participants is of order or less than 10; otherwise

the transition takes too long to be observed. Then such transitions must occur in small spatial domains because far separated transitions are weakly coupled and should take place independently. One can characterize these local fluctuators by some typical dipole moment μ_* to describe their interaction with electrons.

In dielectric glasses the fluctuators, TLS, can be only of structural nature. In electronic glasses they can be of electronic or structural nature, or involve both electron and structural rearrangements. The concept of electronic glass state where localized electrons are frozen in deep local energy minima formed by an external disorder and a long-range electronic interaction has been suggested long time ago [19,20,21] and supported by the extension of spin-glass models to the electronic glass [22]. Several microscopic models have been suggested to account for the formation of fluctuator in the disordered system of interacting localized electrons (see e. g. Refs. [23,24,25]). It is not clear whether the experimental data in crystalline semiconductors indicate the presence of fluctuators there. The $1/f$ noise universal scaling with variable range hopping parameters reported in relatively thin ion-implanted Si:P,B samples [26] can be, indeed, interpreted using the fluctuator model. [25] However, this noise has been remarkably (6 times) suppressed in 1500 nm thick samples annealed at high temperature for a long time so that the impurity atoms diffuse uniformly through the entire thickness. [27]. Therefore it is not clear whether $1/f$ noise is the property of the bulk material or it is associated with the non-uniform distribution of dopants within the sample. The formation of electronic glassy state is not strongly supported by the numerical studies as well. [28,29]

In spite of the nature of fluctuators we can use a TLS like model Eq. (1) assuming a fluctuator density to be a constant, P_*, and characteristic dipole moments to be a constant, μ_*. It is convenient to characterize their relationship by the dimensionless product $P_*\mu_*^2/\kappa_{in} \approx \chi_*$, so one has

$$P_* \approx \frac{\chi_* \kappa_{in}}{\mu_*^2}. \tag{2}$$

In amorphous solids χ_* is a universal parameter of order of $10^{-3} - 10^{-4}$, which can have a weak (logarithmic) temperature dependence. [5,30] As we will see below, the similar assumption leads to a quantitatively valid estimate of the non-equilibrium raise of conductance.

3 Field confinement

In previous studies [16,17,22,25,29,31,32,33,34] the glassy behavior was investigated in the standard variable-range hopping regime, where conductance behaves as $G \propto e^{-(T_0/T)^n}$ $1/4 \leq n \leq 1/2$ [14]. It turns out that the conductance of many materials showing glassy behavior possesses a stronger temperature dependence close to the Arrhenius law, $n \approx 1$. [9,8,12]

We suggest that this behavior is a consequence of a field confinement in 2 dimensions ($2D$) [35,36]. It takes place if the dielectric constant of the film of interest, κ_{in} exceeds that of the environment, $\kappa_{in} \gg \kappa_{ex1} = (\kappa_1 + \kappa_2)/2$ (κ_1, κ_2 are dielectric constants of materials surrounding our sample from two sides). This definitely

takes place in materials close to the metal-insulator transition [8,12] where the dielectric constant approaches infinity. One should note that the actual screening of the Coulomb interaction is associated with the polarization of electronic states rather than real hopping processes formally making static dielectric constant equal to infinity.[37] This is so called resonant contribution [38] similar to the one in dielectric glasses [39].

In the confinement regime the interaction of electrons differs from the standard Coulomb interaction, $e^2/(\kappa_{in} r)$ [35,36] (see also Ref.[40], where the field confinement was considered for Ti oxide based materials). Particularly, the interaction of two electrons at short distance $r \leq d/2$ can be expressed with the logarithmic accuracy as

$$U_C(r) = \frac{e^2}{\kappa_{in} r} + 2\Delta_*,\ \Delta_* \approx \frac{e^2}{\kappa_{in} d} \ln\left(\frac{\kappa_{in}}{\kappa_1}\right). \tag{3}$$

This interaction results in the hard Coulomb gap Δ_* in a DOS leading to the conductance behavior [35,36]

$$G \approx G_0 e^{-\frac{\Delta_*}{k_B T}}, \tag{4}$$

Granular materials can be described by the model of interacting localized electrons in the case of a grain size small compared to the localization length in accordance with the theory of cotunneling [41,42].

Conductance activation energy in granular Al changes from 20 to 40K from most conductive to less conductive samples of the thickness $d \approx 200$Å.[12] Accordingly, using Eq. (3) we find that internal dielectric constant changes in these samples from 30 to 100 always exceeding the external dielectric constant $\kappa_1 \approx 5$ by at least an order of magnitude.

The confinement model can be verified experimentally modifying the environmental dielectric constant, κ_1. For example placing the ice possessing high dielectric constant, $\kappa_{ice} \approx 90$, on the top of the sample should completely destroy thermally activated behavior, Eq. (4), for most "dielectric" samples. Also the activaion energy should decrease with the sample thickness, Eq. (3). The similar trend is seen in granular Al films [43]. Indeed samples of thickness 10 and 20 nm having similar preexponential factors in Eq. (4) have activation energies different by approximately the factor of 2 [43], i. e. for $G_0 \approx 0.8 \cdot 10^{-5}\Omega$ one has $\Delta_{*10}/k_B = 31$K and $\Delta_{*20}/k_B = 14$K, while for $C_0 \approx 0.18 \cdot 10^{-5}\Omega$ one has $\Delta_{*10}/k_B = 57$K and $\Delta_{*20}/k_B = 23$K, respectively. Therefore we believe that our model is relevant for granular Al.

4 Non-equilibrium Dynamics

The only parameter in the definition of conductance Eqs. (3), (4) sensitive to the slow relaxation of fluctuators is the internal dielectric constant, κ_{in}. Following Eq. (4) one can express the non-equilibrium part of conductance in terms of the related nonequilibrium contribution to the dielectric constant, which depends on time and gate voltage, as

$$\frac{\delta G(V,t)}{G} \approx \frac{\Delta_*}{k_B T} \frac{\delta \kappa_{in}(V,t)}{\kappa_{in}}. \tag{5}$$

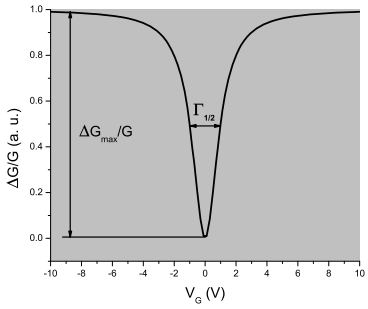

Fig. 2 Typical shape of memory dip

It is quite natural to expect that the resonant dielectric constant of Anderson insulators with fluctuators behaves similarly to that in amorphous solids [4,5,15] giving rise to the experimentally observed non-equilibrium conductance behavior.

A general mechanism for the non-equilibrium relaxation of conductance, earlier used to interpret experimental data for dielectric constant in glasses [5,15], can be described as following [16,17]. Electronic DOS decreases due to interaction of fluctuators with electrons in localized states (see interaction of electron with local fluctuator shown in Fig. 1). If this interaction is sufficiently strong then the electron needs the large energy of that interaction to leave or enter this state. At low temperature such process becomes forbidden and the only possible excitation of electron must be accompanied by the fluctuator transition occurring very rarely compared to electron hopping time. Therefore this electron cannot efficiently respond to the external electric field, i. e., contribute to hopping conductivity or dielectric constant. Thus the equilibration of the system after its disturbance, e. g. by the gate voltage, results in a slow reduction of conductance by means of trapping of low energy electrons, primarily released after the application of a gate voltage, by relaxing fluctuators. [5]

Important experimental data characterizing glassy behavior were obtained in the memory dip measurements [8,12]. Memory dip is seen in conductance after a fast sweep of the gate voltage around its equilibrium value, $V_g = 0$ where it was hold before for tens of hours (see V-shaped conductance dependence on that voltage in Fig. 2). The greater the voltage the stronger the system departs from equilibrium increasing the conductance, according to the previously described scenario. Memory dip can be characterized by its half-width $\Gamma_{1/2}$ and depth $\delta G_{max}/G$.

The temperature dependence of conductance, G, memory dip parameters, $\Gamma_{1/2}$, and $\delta G_{max}/G$ contain significant information about the nature of the electronic glassy state. Here we suggest the theoretical model of electronic glass involving the $2D$ field confinement, which is capable to interpret several observed behaviors at intermediate temperature, $4\text{K} \leq T \leq 20\text{K}$, including (A) linear temperature dependence of a memory dip half-width and its universality with respect to a sample conductance [12] (cf. [8]) and (B) nearly T^{-2} dependence of $\Delta G/G$ slightly varying for samples with different conductances [12].

Consider the effect of fluctuators on the dielectric constant determining the memory dip Eq. (5). The dielectric constant is associated with the polarization of electron-hole pairs of various lengths r, smaller than the hopping length. Similarly to Ref. [15] the part of dielectric constant most sensitive to its interaction with fluctuators is associated with low-energy electron-hole pairs separated by intermediate distance, r, such that $a < r < d/2$. It is harder to disturb shorter dipoles due to their large tunneling splitting, $\Delta_0 \geq k_B T_0 \gg k_B T$, while longer pairs do not make important contribution due to the "hard" Coulomb gap at corresponding energies caused by the field confinement in $2D$. Let those dipoles be characterized by the density function $F(\Delta, r)$ depending on their energy, Δ and size r. This size determines the tunneling amplitude of electron between two localized states, $\Delta_0 \approx E_0 e^{-r/a}$, $E_0 \approx k_B T_0$. Then one can approximate the contribution of interest to the dielectric constant as (c. f. [14,38])

$$\Delta\kappa_{in} \approx \frac{2\pi e^2}{3} \int_a^{\frac{d}{2}} r^4 dr \int_{-\infty}^{\infty} d\Delta \frac{\Delta_0^2 F(\Delta, r)}{E^3} \times \tanh\left(\frac{E}{2k_B T}\right);\ E = \sqrt{\Delta^2 + \Delta_0^2}. \tag{6}$$

This contribution is identical to the resonant contribution of two level systems to the dielectric constant in amorphous solids [5,15]. The relaxational contribution does not affect the effective Coulomb interaction as demonstrated in Ref. [37] using both analytical and numerical studies.

In the absence of fluctuators the density function of sufficiently large dipoles, $a < r < d$ with the energy, $E \sim k_B T$, can be approximated by [14,38]

$$F(E, r) \approx \frac{3}{10\pi^2} \left(\frac{\kappa_{in}}{e^2}\right)^6 \left(\frac{e^2}{\kappa_{in} r} + E\right)^5 \approx \frac{3}{10\pi^2} \frac{\kappa_{in}}{e^2 r^5}, \tag{7}$$

where a small energy, $E \sim k_B T$, can be approximately neglected.

Consider the change in dipolar density and, correspondingly, conductance induced by the gate voltage sweep to some value V_g. This gate voltage creates electric field inside the sample, which can be estimated as $F_{DC} \approx V_g \kappa_1 / (\kappa_{in} d_{tot})$, where d_{tot} is the thickness of insulating layer separating the sample from the gate electrode. In the granular Al $d_{tot} \sim 10\text{nm} \gg d$. This estimates is not valid at very small gate voltage where the distance between injected electrons exceeds the sample thickness d. However for granular Al samples of 200 nm thickness at $T > 4\text{K}$ this is not the case for gate voltages of interest [12].

The interaction of dipoles with fluctuators reduces their density of states [15,17]. Only interaction exceeding the thermal energy is important. We assume, that the distance between the fluctuator and closest charge of the dipole is smaller than the size of the dipole r, which is necessary to make the interaction larger than the thermal energy. This is true for $r \sim d/2$ and $T > 4\text{K}$ [11]. Then the correction to the dipole density due to fluctuators with relaxation times τ within the domain, $t_{min} < \tau < t_{max}$,can be expressed as [16,17]

$$\frac{\delta F(E, r)}{F(E, r)} \approx -\frac{32\pi}{15} \frac{\Delta}{E} P_* E_0 a^3 \left[\frac{E_0}{E}\right]^{1/2} \ln\left(\frac{t_{max}}{t_{min}}\right). \tag{8}$$

The time dependence is due to a logarithmically uniform distribution of fluctuator relaxation time (Eq. (1)), t_{max} stands for the time the system spent after being cooled down and t_{min} is some characteristic minimum time, determined by the rate of a gate voltage application.

Substituting Eq. (8) into Eq. (6), performing the straightforward integrations and substituting the final result into Eq. (5) we obtain the following expression for the fluctuator correction to the conductance

$$\frac{\delta C}{C} \approx -\frac{32\sqrt{2}\chi_*}{75}\frac{\Delta_*}{k_BT}\sqrt{\frac{E_0}{k_BT}}\ln\left(\frac{d}{2a}\right)\ln\left(\frac{t_{max}}{\tau_{min}}\right). \tag{9}$$

This equation describes the aging effect. It can be also used to describe the conductance time dependence after the application of a large gate voltage bringing all relevant fluctuators out of equilibrium. Then the minimum time should be replaced with the waiting time t and the sign of the expression should be reverted. The temperature dependence of the conductance logarithmic relaxation rate $r = d\ln(C)/d\ln(t)$ is close to the power law $r \propto T^{-3/2}$ that is consistent with the observations of Ref.[9] in ultrathin films of Bi and Pb.

Consider the shape of the memory dip (Fig. 1). The energy change of fluctuator with the dipole moment μ associated with the application of a gate voltage V_g is given by $\delta E = F_g\mu\cos(\theta) \approx V_g\kappa_1\mu\cos(\theta)/(\kappa_{in}d_{tot})$, where θ is the angle between directions of the dipole moment and the external electric field F_g, d_{tot} is the distance between sample and gate electrode and k_1 is the dielectric constant of the insulating layer separating the gate electrode from the sample (see Refs. [12,8] for details). Only fluctuators with sufficiently small energy E, $0 < E < \delta E$, will be really disturbed from their equilibrium states. This condition sets the upper constraint δE for integration over energy in Eq. (6). Also the time dependent logarithm $\ln(t_{max}/t_{min})$ in Eq. (6) should be modified. It is determined by the contribution of fluctuators removed from equilibrium by the gate voltage, which possess the relaxation time smaller than the maximum time t_{max}, determined by the time the sample was kept at fixed temperature after cooling and larger than the minimum time, t_{min}, determined by the gate voltage sweep rate, r_s, as $t_{min} \sim k_BT/(er_s)$. Then the memory dip can be described by the equation

$$\frac{\delta C(V_g)}{C} \approx -\frac{32\sqrt{2}\chi_*}{75}\frac{\Delta_*}{k_BT}\ln\left(\frac{d}{2a}\right)$$
$$\times\ln\left(\frac{t_{max}r_se\kappa_1}{k_BTd_{tot}\kappa_{in}}\right)\left[\sqrt{\frac{E_0}{k_BT}}-2\sqrt{\frac{E_0d_{tot}\kappa_{in}}{V_gea\kappa_1}}\right]. \tag{10}$$

Using this result one can estimate the half width of the memory dip as

$$\Gamma_{1/2} \approx 16\frac{k_BT}{e}\frac{\kappa_{in}d_{tot}}{\kappa_1 a}. \tag{11}$$

‘ This result agrees with the experimentally observed linear temperature dependence of the memory dip width at temperatures $T \geq 4$K [12,8]. At lower temperatures the dependence is getting stronger. In our opinion this is because the estimate for gate voltage induced electric field, $F_g \approx V_g^{(1/2)}\kappa_1/(\kappa_{in}d_{tot})$, is no longer valid. One

can use that macroscopic expression if the density of electrons injected due to the application of a gate voltage exceeds one electron per the cubic sample thickness. This is satisfied for granular Aluminium samples down to 4K [10], but fails at lower temperatures, where this field is defined by the injected electron, closest to the given fluctuator. One can show that at low temperatures a width of memory dip is proportional to squared temperature, $V_g^{(1/2)} \sim \frac{(k_B T)^2}{e} \frac{d^2 d_1 \kappa_{in}^2}{e^2 \kappa_1 a^2}$.

The quantitative universality of the memory dip halfwidth discovered in [12] for samples with conductances different by orders of magnitude requires understanding. It is important that in the vicinity of a metal-insulator transition the ratio of two diverging parameters, ε_{in}/a, is expected to be approximately constant on the dielectric side both in accordance with the theoretical analysis and experimental data [45]. One can naively expect that at the scale of localization length the characteristic kinetics and Coulomb energies should be close to each other, $E_0 = e^2/(\kappa_{in} a) \approx \hbar^2/(m a^2)$, which makes the ratio, $a/\varepsilon_{in} \sim \hbar^2/(m e^2) \approx 0.5\text{Å}$, universal. It expresses the Bohr's radius, a_B (here m is the effective mass of electron, which is taken to be equal to the bare electronic mass).

Under these assumptions Eq. (11) results in the universal dependence of the memory dip halfwidth on the gate voltage $\Gamma_{1/2} = \eta T$ with the proportionality coefficient coefficient η depending only on the thickness d_{tot} and dielectric constant κ_1 of the layer separating the sample and the gate electrode. For granular Aluminium sample one has $\kappa_1 \approx 9$, $d_{tot} = 1000\text{Å}$ which yields $\eta = 0.29\text{V K}^{-1}$ in excellent agreement with the experimental result $\eta \approx 0.25\text{V K}^{-1}$ [12].

The same approach does not work so good for the half-width of the memory dip reported in Ref. [8] for In_2O_{3-x} sample having a relatively small resistance of $200\text{k}\Omega$. We believe that this is because the localization length in this almost conducting sample exceeds the thickness of the sample $d_{InO} \sim 30\text{Å}$ and therefore one should replace the localization length a in Eq. (11) with the sample thickness. Assuming that the localization length exceeds the sample thickness by an order of magnitude one can make the experimental data consistent with the theory. Other data reported in Ref. [8] correspond to very low temperatures. However, in granular Aluminium one can estimate, fitting activation energy $20 - 40\text{K}$ with Eq. (3) and using the relationship, $a \approx 0.5\kappa_{in}(\text{Å})$, that the localization length is small enough, $15\text{Å} < a < 50\text{Å}$, in all samples of thickness 200Å so our estimate is justified.

The temperature dependence of the depth of the memory dip in Eq. (10) in the limit of $V_g \to \infty$ is expressed by the power law, $\Delta C/C \propto T^{-3/2}$, and additional weak temperature dependencies of the dimensionless parameter χ_* and the logarithmic factor $\ln\left(\frac{t_{max} r_s k_{ex}}{k_B T d_{tot} \kappa_{in}}\right)$. Integration of these three dependences could be responsible for the T^{-2} behavior of the memory dip depth reported in Ref. [11].

According to the experiment [11] the absolute value of the relative correction reaches few percents at $T \sim 4K$ for less conducting sample, which is consistent with Eq. (10) if one set $\chi_* \sim 5 \cdot 10^{-4}$ in agreement with the typical values of that parameter in amorphous solids [5].

5 Conclusion

Thus we suggested the model of slow dynamics in electronic glasses due to fluctuators behaving similarly to TLS in amorphous solids. The model is consistent with existing experimental data both qualitatively and quantitatively. Theoretical predictions, Eqs. (10), (11) can be tested varying system parameters including electron localization length, sample thickness and temperature.

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