Slow relaxation of conductance of amorphous hopping insulators

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Abstract

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

1. Introduction

A peculiar transport memory effect has been observed in many hopping insulators [1–5]: shifting the system either from the equilibrium or from its steady transport state produced by, e.g., a sudden change of the gate voltage, $\delta V_g$, increases the conductance $\sigma$ of the system, the effect not depending on the sign of the change. The $\sigma(\delta V_g)$ dependence shows a characteristic memory cusp (see [6] for experimental details and a review) which may persist for a long time. Explanations of this appealing observation can be grouped into two major classes which are often referred to as intrinsic and extrinsic mechanisms. The former one attributes memory effects to slow dynamics of strongly correlated electrons subject to quenched disorder and thus forming a Coulomb glass [7–9]. The connection between the features of a glassy phase and the formation of a Coulomb gap was revealed in [10], suggesting that within the locator approximation the correlated electronic system maps to the Sherrington–Kirkpatrick spin glass. A direct relation between the long range electron correlations and the formation of the exponential distribution of deep electron states, which is characteristic of glassy systems, was also demonstrated in [11]. In a Coulomb glass, memory effects reflect delayed formation of the Coulomb gap in the Efros–Shklovskii (ES) regime of the variable range hopping (VRH) [12].Recently we have demonstrated how ‘slow’ many-electron fluctuators can be formed in a Coulomb system and analyzed their influence on the $1/f$ noise [13]. Such fluctuators can also lead to slow relaxation and memory effects.

An extrinsic scenario assumes that electronic memory effects are caused by slowly relaxing atomic configurations thus influencing conducting channels and was first proposed in [14] to explain the $G(V_g)$-cusp in granular Au films. A possibility that polaron effects may be responsible for slow relaxation in hopping conductors was also discussed in [5, 15].

The Coulomb glass is formed well below the temperature $T_0$ entering the ES law for the VRH as

$$\sigma = \sigma_0 e^{-(T_0/T)^{1/2}}. \quad (1)$$

Experiment [16] has shown that in this regime the $1/f$ noise intensity is strongly correlated with $T_0$, proving the electronic nature of the low-frequency noise. However, the conductance memory cusps and their relaxation (aging) were observed in the systems which do not exhibit ES behavior, but are apparently subject to structural disorder [6, 17]. Various memory effects were observed in metallic granular structures [2, 5, 14], which also possess a high degree of structural disorder. This calls for careful examination of whether the memory effects and the ‘two-cusps’ $G(V_g)$-dependence [6] can be explained as a result of slow relaxation in the structurally disordered atomic matrix.
In this paper we present a model that may serve as a step towards a quantitative description of memory cusps based on an extrinsic mechanism. We show that due to slow relaxation of atomic structure, polaron clouds, which form near the hopping sites, suppress the bare electron density of states (DOS). Changing gate voltage shifts chemical potential, removes the polaron screening and, thus, increases the hopping conductivity. As the atomic structure adjusts itself with time to the new position of chemical potential, the conductivity relaxes to its quasi-stationary magnitude.

Atomic structural relaxation is attributed to two-level systems (TLSs) [18, 19]. The TLS model successfully describes thermodynamics and kinetics of amorphous solids at low temperature. It suggests that there exist atoms or groups of atoms undergoing tunneling motion that can be characterized by the broad universal distribution of their parameters. Since all materials where the memory effects in conductivity were observed are strongly disordered, one would expect that TLSs should exist there similarly to other glasses and disordered materials [19]. These two-level systems interact with conducting electrons because they possess a dipole moment. In this manuscript we examine the effect of electron–TLS interaction on the non-equilibrium conductivity. We show that this interaction results in the non-equilibrium behavior of conductivity which is qualitatively equivalent to the experimental observations, i.e. increase in conductivity after gate voltage application with its subsequent logarithmic relaxation to the equilibrium value. This theory uses the previous work [20, 21], where the similar non-equilibrium behavior of the dielectric constant in amorphous solids [21, 22] has been explained using the TLS interaction. Since TLS parameters are quite universal from material to material [19], we can use these parameters for quantitative estimates which show that our theoretical predictions are consistent with existing experimental data.

The paper is organized as following. In section 2 we define the conditions where our consideration is applicable, i.e. electron–electron interaction can be neglected, while electron–TLS interaction is significant. In section 3 the non-equilibrium behavior of electron density of states caused by their interaction with TLSs is derived. In section 4 the non-equilibrium behavior of conductivity is described and compared with the experimental data. The results of the manuscript are summarized in section 5.

2. When can the electron–electron interaction be neglected?

In this manuscript we ignore the effect of electron–electron interaction on the non-equilibrium behavior of conductivity. This is possible only under specific conditions when the electronic interaction is weak compared to their characteristic energies and there is no slow relaxation within the electronic subsystem. According to various considerations [12, 13], the slow relaxation in the electronic subsystem can take place only below some critical temperature \( T_G \), which is defined as the electronic glass transition temperature in [12]. This transition temperature is defined by the Coulomb gap energy

\[ k_B T_G \sim \Delta_C. \]  

Below we set \( k_B = 1 \).

The Coulomb gap depends on the electronic density of states \( g_0 \), dielectric constant \( \kappa \), system dimension \( d \) and electron localization radius \( a \). We believe that the dimensionless parameter

\[ \chi = g_0 a^{d-1} e^2 / \kappa = g_0 a^d T_0 \]

is small, i.e.

\[ \chi = g_0 a^d T_0 < 1. \]  

where the characteristic temperature \( T_0 \) in the Efros–Shlovskii hopping conductivity equation (1) is given by

\[ k_B T_0 \sim \frac{e^2}{\kappa a}. \]

Then the Coulomb gap is defined as

\[ \Delta_C = T_0 \chi^{-1}. \]  

If temperature exceeds the Coulomb gap energy then electronic relaxation is fast. It is also useful to notice that for electronic excitations with typical energy \( E \) their characteristic interaction \( U_E \sim e^2 (g_0 E)^2 \) is smaller than the energy \( E \) until \( E \sim T < \Delta_C \) and the condition \( U_E \sim E \) serves as the definition of the Coulomb gap. So when \( T > \Delta_C \) the interaction of representative excitations with energy \( E \geq T \) is less than their energy so it can be treated as weak.

If the system is near metal dielectric transition [16] and the electron localization radius is large, then one can possibly have the opposite limit \( \chi = g_0 a^{d-1} e^2 / \kappa > 1 \). This is the case which takes place in silicon MOSFETs investigated in [16]. Under those conditions only the Efros–Shlovskii variable range hopping law equation (1) is observed. We believe this is not the case for the systems of interest [1–5], where different conductivity behavior is observed that is closer to the Mott’s variable range hopping law, which suggests \( \chi < 1 \) [23].

Therefore it is not possible to interpret the memory effects in conductivity at \( k_B T > \Delta_C \) without the involvement of electronic interaction with extrinsic slowly relaxing defects like TLSs. Our theoretical study is restricted to this ‘high temperature’ situation. Since the temperature \( T_G \) can change within the range of 1–100 K [16] our assumption that the experimental temperature 4 K exceeds the Coulomb gap energy equation (5) does not conflict with common sense. We believe that at least some of the experiments [1–5] have been performed in this temperature range. Unfortunately, existing experimental data do not permit us to answer the question as to whether the experimental temperature is above or below the Coulomb gap.

Although the absence of the Efros–Shkovskii conductivity temperature dependence equation (1) [3] agrees with our assumption that the electronic interaction is not important, this does not prove the absence of Coulomb gap and electronic glassy state [23]. We therefore suggest additional experiments that can help to investigate the Coulomb gap in the system which is a necessary prerequisite of the Coulomb glass state. Such experiments can be made, for instance, using scanning tunneling microscopy [25]. If the Coulomb
gap in the density of electronic states is not observed at experimental temperatures, then one should expect that the extrinsic mechanism is in charge for the memory behavior in conductivity.

3. Electron density of states affected by electron–TLS interaction

Our consideration is based on the concept of ‘two-level systems’ (TLSs) [18] taking as its central hypothesis the assumption that in a system with quenched disorder a certain number of atoms (or groups of atoms) can occupy one of (at least) two equilibrium positions. These atoms therefore move in a double-well potential created by their environment and characterized by the asymmetry energy (difference in energy minima) and by the height and the width of the separating barrier. The atoms comprising the TLSs change their configuration either by tunneling through- or by thermally activated hopping over the barrier. Randomness in the heights and widths of the TLS barriers gives rise to an exponentially broad distribution of structural relaxation times.

A localized electric charge polarizes the neighboring TLSs, which thus acquire an electric dipole moment, \( \mu \), and screen the original bare charge, see figure 1(a). The interaction energy of TLSs with the localized charge is estimated as

\[
U(\mathbf{r}) \sim e(\mathbf{\mu} \cdot \mathbf{r})/\kappa r^3, \quad r \gg a, \tag{6}
\]

where \( \mathbf{r} \) is the vector connecting the center of this TLS with the position of localized electron state, \( a \) is the localization length and \( \kappa \) is the material dielectric constant. Polarized TLSs form a polaron cloud around the localized electron and create a polaron gap for electronic excitations, implying that states with the single-electron energy \( \phi < \sum_i U(\mathbf{r}_i) \) cannot be excited any more.

In what follows we will find the correction to the electronic DOS, \( g_0 \), due to electron–TLS interactions in the lowest approximation in the TLS density. To this end we first determine the change in DOS due to a single neighboring TLS characterized by the given energy splitting, \( E \), and the relaxation time, \( \tau \). The target correction to the electron DOS is then obtained by averaging of the single TLS contribution with respect to all possible neighboring TLS positions and \( E \) and \( \tau \).

Consider the correction to the density of electronic states with some energy \( \varepsilon \) caused by the electron–TLS interaction. Since the concentration of TLS is small we can assume that only one most closely located TLS is important, while the probability that two TLSs are significant is much smaller than unity. Then we consider electrons interacting with the single TLS using the standard Hamiltonian

\[
\hat{H}_{\text{pair}} = \phi n + Un\sigma^z + \Delta\sigma^z, \tag{7}
\]

where \( n = 0, 1 \) is the electron population operator, the spin \( 1/2 \) \( z \)-projection operator \( \sigma^z = \pm 1/2 \) describes two states of the TLS, \( \phi \) is the electronic energy in some localized state, \( \Delta \) is the energy of the two-level system and \( U \) is the charge–dipole interaction of the electron with the TLS. In thermal equilibrium the electron excitation energy \( \varepsilon = \phi + U\sigma \) can take values \( \phi \pm U/2 \) with the probabilities defined by the equilibrium Boltzmann factors

\[
p^+ = \frac{\exp\left(-\frac{\Delta}{\kappa\tau}\right) + \exp\left(-\frac{\Delta - U/2 - \phi}{\kappa\tau}\right)}{\exp\left(\frac{\Delta}{\kappa\tau}\right) + \exp\left(-\frac{\Delta - U/2 - \phi}{\kappa\tau}\right) + \exp\left(\frac{\Delta + U/2 - \phi}{\kappa\tau}\right) + \exp\left(-\frac{\Delta + U/2 - \phi}{\kappa\tau}\right)}, \tag{8}
\]

\[
p^- = \frac{\exp\left(\frac{\Delta}{\kappa\tau}\right) + \exp\left(\frac{\Delta + U/2 - \phi}{\kappa\tau}\right)}{\exp\left(\frac{\Delta}{\kappa\tau}\right) + \exp\left(-\frac{\Delta - U/2 - \phi}{\kappa\tau}\right) + \exp\left(\frac{\Delta + U/2 - \phi}{\kappa\tau}\right) + \exp\left(-\frac{\Delta + U/2 - \phi}{\kappa\tau}\right)},
\]

respectively. The time-dependent correction to the density of states taken at a certain time \( t \) can be associated only with those TLSs which have relaxation time \( \tau \) longer than the time of the experiment \( t \). That time must also have an upper restriction by some maximum TLS relaxation time \( \tau_{\text{max}} \) which serves as the upper cutoff in the TLSs’ logarithmically uniform distribution over their relaxation times \( P(\Delta, \tau) = P_0 \Theta(\tau_{\text{max}} - \tau)/\tau \). Within the logarithmic accuracy, one can account for the above constraints introducing integrated time-dependent TLS density at \( t < \tau_{\text{max}} \) as

\[
P(t) = P_0 \ln(t/\tau_{\text{max}}). \tag{9}
\]

Here \( \tau_0 \) is the characteristic minimum time defined by the field sweep rate. This expression clearly demonstrates the nature of the logarithmic time dependence of TLS contribution to the electronic density of states similarly to previous work [20].

The correction to the density of states of the particular electron with energy \( \varepsilon \) caused by its interaction with neighboring TLSs can be expressed as

\[
\delta g_\varepsilon = \frac{1}{V} \sum_{ij} \left( P^+_{ij} \delta(\varepsilon - \phi_i - U_{ij}/2) + P^-_{ij} \delta(\varepsilon - \phi_i + U_{ij}/2) - \delta(\varepsilon - \phi_i) \right), \tag{10}
\]

where the sum is taken over all pairs made of an electron \( i \) and a TLS \( j \) characterized by energies \( \phi_i \) and \( \Delta_j \), respectively, \( U_{ij} \) is their interaction and \( V \) is the system volume. Probabilities \( P^\pm_{ij} \) are defined using equation (8) with the substitutions \( \phi = \phi_i \), \( \Delta = \Delta_j \) and \( U = U_{ij} \). The summation in equation (10) can be
replaced with integration over electron and TLS energies and the distance $R$ between electron and TLSs. This yields

$$\langle \delta g(\varepsilon) \rangle = g_0 P(t) \int d\mathbf{r} \int d\mathbf{r}' \int d\phi \int d\phi' \int d\Delta$$

$$\times \left\{ (1 + e^{-\frac{t}{\tau}}) \delta(\varepsilon - \phi - U/2) + e^{\frac{t}{\tau}} (1 + e^{-\frac{t}{\tau}}) + (1 + e^{-\frac{t}{\tau}}) \delta(\varepsilon - \phi + U/2) + e^{\frac{t}{\tau}} (1 + e^{-\frac{t}{\tau}}) - \delta(\varepsilon - \phi) \right\},$$

(11)

where $g_0$ is the electronic density of states at energy $\varepsilon$ taken in the absence of interaction with TLSs and $d$ is the system dimension. Remember that $U = e(\mu, R)/(\varepsilon R^3)$ is the dipole charge interaction between electron and TLS possessing dipole moment $\mu$ and averaging is performed with respect to random directions of a TLS dipole moment. After integration over electronic energy $\phi$ and TLS energy $\Delta$ in equation (11) we obtained

$$\langle \delta g(\varepsilon, t) \rangle = g_0 P(t) T \int d\mathbf{r} \int d\mathbf{r}' \left[ \ln \left( \frac{\cosh(\varepsilon/T) + 1}{\cosh(\varepsilon/T) + \cosh(U/T)} \right) \right].$$

(12)

The most important electrons are those contributing to the hopping conductivity. These electrons have energy $\varepsilon$ of order of the hopping energy $\varepsilon_h$, which always exceeds the thermal energy. Therefore assuming $\varepsilon > T$ one can approximate the logarithm under the integral in leading order in $1/T$ as

$$\ln \left( \frac{\cosh(U/T) + 1}{\cosh(U/T) + \cosh(U - \varepsilon)} \right) \approx -\frac{1}{2} \exp(-\varepsilon/|U|),$$

The final expression for the correction is similar to the one used in earlier works [20, 24] for the Coulomb gap and the dipole gap in the density of TLS states. However, our derivation of the correction is more general because our expression equation (12) can be used at arbitrary temperature while the earlier derivation is valid only in the low temperature limit. Finally the correction to electronic density of states can be expressed as

$$\langle \delta g(\varepsilon, t) \rangle \approx -2 g_0 P_0 \varepsilon \mu \frac{\varepsilon}{\kappa} \frac{\mu}{\kappa} \frac{T}{\mu} \ln \left( \frac{t}{\tau_m} \right),$$

where $\mu$ is the typical dipole moment of a TLS and $\tau_m$ is some characteristic minimum time associated with the gate voltage application. Only electrons with energy $\varepsilon \sim \varepsilon_h = T^{3/4} a^{-3/4} e^{-1/4}$ influence the hopping conductance. We believe that at $\varepsilon \sim \varepsilon_h \sim 30$ K equation (13) is still applicable.

4. Analysis of experimental data

Equation (13) can be used to interpret the experimental data only if all relevant TLSs were initially out of equilibrium. In other words, all electrons contributing to hopping must be surrounded by non-equilibrium TLSs. The equilibrium can be broken due to the application of the gate voltage. This can be realized when all electrons contributing to hopping are 'newcomers', i.e. they are brought to the vicinity of the Fermi level by application of the gate voltage, $V_g$. For that, the shift of the Fermi energy by the gate voltage must exceed the hopping energy $\varepsilon_h$. Then those electrons break the equilibrium in their neighboring TLSs randomly changing their energies by the scale of their interaction with those TLSs. All relevant TLSs with energy of order of $\varepsilon_h$ coupled to entering electrons (or holes) by an interaction having the same order of magnitude, experience the jump in their energy induced by entering electrons. This moves them all out of equilibrium. Their relaxation leads to the polaron shift of electron energy out of the Fermi energy thus reducing the conductivity. Thus the condition $\Delta E \approx \varepsilon_h$ defines the width of the cusp in the non-equilibrium conductivity as a function of the gate voltage. One can show using [7] that under those conditions the shift of TLS energy induced 'directly' by the gate voltage is still smaller than $\varepsilon_h$ because of the small TLS dipole moment $\mu \approx 2 \times 10^{-18}$ erg/cm$^{3/2}$ [19]. We can compare this estimate with the experiment extracting $\varepsilon_h$ from the expression for the conductivity, $\sigma \approx \sigma_0 \exp(-\varepsilon_h/T)$. Let us put $\sigma_0$ equal to the minimal metallic conductivity ($\sim 10^{-4}$ $\Omega^{-1}$) and use available experimental results of [3] for the representative sample with resistance of $3.8$ M$\Omega$ at $T = 4.1$ K. The assumption about $\sigma_0$ is justified by the experimental observations (see [29] and references therein).

Then the hopping energy is $\varepsilon_h = T \ln(\sigma_0/\sigma) \approx 2.1 \times 10^{-3}$ eV. This value agrees qualitatively with the shift of the Fermi energy 3–5 meV associated with the value of the gate voltage $V_g$ at which the memory cusp is affected as was estimated earlier in [7]. Thus the suggested mechanism agrees with the experimental observations.

The correction to the conductivity can be estimated as

$$\langle \delta V_{g,x} \rangle \approx \frac{P_0 e \mu}{\kappa} \left( \frac{e \mu}{\kappa} \right)^{1/2} \ln \left( \frac{t}{\tau_m} \right).$$

(14)

The time-dependent factor $\zeta = \ln(t/\tau_m)$ contains the measurement time as $t$ and the inverse sweep rate, $\tau_m \gg \tau_0 = T_0 e^{(\varepsilon_h/T)T/4}$, and affects only the amplitude of the peak, but not its shape. Here $\tau_0$ is the characteristic time of the variable range hopping and the pre-exponential factor $\tau_0$ is of the order of 1 ps at experimental temperatures. This can explain the experimentally observed independence of the dip shape on the sweep rate [7].

Similar considerations apply to the ES VRH with the proper renormalization of the hopping parameters. Now the typical energy scale optimizing the hopping rate is $\varepsilon_h = (T_0/T)^{1/2}$, where $T_0 = \beta e^2/k a$, $\tau_n = T_0 e^{(\varepsilon_h/T)T/4}$, and $\beta \approx 2.8$ is a numerical factor [23].

Let us now discuss the available experimental data in the light of the above theory. According to our previous estimates, $\varepsilon_h \approx 2.1 \times 10^{-3}$ eV. The typical dipole moment can be estimated as $\mu \approx 2 \times 10^{-18}$ erg/cm$^{3/2}$, which is close to typical dipole moments of TLSs in glasses [19, 20].

We are not aware of independent measurements of the TLS density, $P_0$, in the materials under consideration. In principle, this quantity can be determined, e.g. by measurement of the low-frequency dielectric constant at $T \approx 1$ K, where it should depend on the temperature logarithmically, the slope of the logarithmic dependence being $P_0 \mu_{\ell}^2/\kappa$. This dimensionless quantity turns out to be almost the same, $\approx 0.3 \times 10^{-3}$, in
many materials with strong quenched disorder, see [19–22] and references therein. Apparent universality of this quantity in such materials was attributed to the interaction between TLSs [21, 26]. Assuming that the material studied in [3] belongs to the same ‘universality class’ as materials with strong quenched disorder, we estimate \( P_\text{TLS} \approx 0.6 \times 10^{13} \text{ erg}^{-1} \text{ cm}^{-3} \), as in most oxide glasses [19]. We set \( \kappa \approx 10 \) following [9].

Now we can check our assumptions regarding the 3D arrangement of TLSs forming the polarons and regarding the lowest approximation in the TLS density. Estimating the polaron radius, \( r_p \), as the length at which the electron–TLS interaction, \( e\mu/kT \), is comparable with the typical electron energy \( \epsilon_h \), we get \( r_p \approx 1 \) nm which is much less than the sample thickness \( \ell \). The average number of the TLSs forming polarons, i.e. located within the polaron radius and having \( E \lesssim \epsilon_h, N \sim (4\pi/3) P_\text{TLS} \rho_p^3 \), turns out to be \(< 10^{-2} \), i.e. much less than 1. Thus the lowest approximation in the TLS density is valid.

Using the above estimates and equation (14) we get \( \delta\sigma(t)/\sigma \approx 0.02 \ln(t/t_0) \). Thus we predict a logarithmic relaxation rate of conductivity \( r = d \ln(\sigma)/d \ln(t) \approx 0.02 \). According to the experimental data [3, 8] at \( T = 4.1 \) K and for sample thickness equal to 10 nm the conductivity changes by about 8% during two decades in time so we can estimate it as \( r \approx 0.015 \). Thus our theory agrees with experimental data reasonably well.

5. Discussion and conclusion

We have presented a simple model of slow dynamics of hopping conductance in structurally disordered hopping insulators. It takes into account rearrangements of the dynamic structural defects, TLSs, leading to the formation of polarons close to the electron hopping states. The model qualitatively explains both the logarithmic relaxation and memory effects, and provides quantitatively reasonable estimates of the time-dependent non-equilibrium change in conductivity, \( \delta\sigma(V_\text{g}, t) \) (see, e.g. [6], figure 2).

The dependencies of this quantity on different parameters—electron concentration, controlled by the gate voltage; magnetic field [3] and various protocols of breaking down the system equilibrium—are encoded in the logarithmic factors \( \ln(\sigma/\sigma_0) \) and \( \ln(t/t_0) \) while the temperature dependence enters as a power law through the energy \( \epsilon_h \): \( \delta\sigma \propto \epsilon_h^{-3/2} \). That leads to the main temperature dependence \( \propto T^{-9/8} \) and \( \propto T^{-3/4} \) for the Mott and ES VRH, respectively. Thus our theory explains the fast increase of the non-equilibrium raise of conductivity with decreasing temperature.

The following note is in order. We demonstrated that ‘slow’ excitations induced by structural disorder can indeed be responsible for the double-dip memory effect in hopping semiconductors in analogy to those in glasses [21, 22]. The effect we considered is due to polarons formed by the structural excitations. Yet, one should bear in mind that a similar polaron effect appears in pure electronic models. In particular, the polarons formed from pair excitations were considered in [27]. In its turn, ‘electronical polarons’ can be formed from slow relaxing electronic ‘aggregates’ discussed in [13]. The presence of structural TLSs in In\( _{\text{FO}} \) films is indirectly supported by the fact that the low-frequency noise in these materials does not increase significantly under the disorder-driven metal-to-insulator transition [28].

Further experimental verification is necessary to decide whether the non-equilibrium behavior is associated with the extrinsic or intrinsic model. A regular approach to attain this goal should be based on techniques affecting differently the structural and electronic degrees of freedom. One of the possibilities is measurement of the AC linear response such as simultaneous measurements of the attenuation and velocity of the acoustic waves in transverse magnetic fields. The expected effect is caused by transitions in structural two-well configurations, while the electronic transitions are strongly suppressed by the magnetic field [30]. Alternative experimental verification can be performed using scanning tunneling microscopy [25] as described in section 2.

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