Electron glass: Intervalley transitions and the hopping conduction noise

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The properties of the electron glass ground state and of those low-energy metastable states (valleys), in which the Coulomb potential at any occupied impurity is lower than that at any empty one, are studied by computer simulation. The transitions between just these states are expected to determine the low-frequency stochastic dynamics of the electron glass at low temperatures. The variation of the number of valleys, N_v, in samples with the same number of impurities, N_D, but different arrangements, the shift of the N_v distribution to greater numbers with growing N_D, the energy range of the valleys, the differences between the electron arrangements in different valleys in the same sample, and the activation energies for intervalley transitions are found. The energy range of the valleys is, at any N_D, on the order of the characteristic Coulomb energy at the mean distance between impurities. Since the number of valleys grows with N_D the mean distance between adjacent valley energies drops with N_D. Despite the small differences between the valley energies the valleys are separated by energy barriers that, in samples with high number of N_D and N_v, are distributed within a wide range. The width of this range grows with the size of the sample (N_D) and with the number of valleys in it. This is an argument in favor of the idea that just the intervalley transitions are the source of low-frequency hopping conduction noise with the $1/f$ spectrum in lightly doped semiconductors at low temperatures. [S0163-1829(98)05616-1]

I. INTRODUCTION

Strongly disordered systems constitute a very interesting and important class of physical systems: amorphous solids, semiconductors with hopping conduction, semiconductors near the semiconductor-metal transition, spin glasses, vortex glasses in high-T_c superconductors, proton glasses, metal-insulator mixtures, and many others. One of their common features is the high magnitude of the low-frequency noise whose spectrum is, as a rule, of $1/f$ type: resistance noise in amorphous solids and semiconductors with hopping conduction, magnetic noise in spin glasses, etc. (for a review see Ref. 1). The noise is a manifestation of the stochastic dynamics of the system. What common properties stemming from the frozen-in strong disorder are responsible for the specific low-frequency stochastic dynamics of these systems, which results in the intensive $1/f$ noise? To answer this question, we should study the mechanisms of low-frequency noise in various strongly disordered systems and find their common basic features.

This paper is concerned with the low-frequency hopping conduction noise in electron (Coulomb) glasses (EG), that is, in semiconductors, doped with shallow impurities and partially compensated, at low temperatures. The mean properties, including the mean transport properties, of EG are well studied (see Ref. 2). The stochastic dynamics and the resulting resistance noise are much less understood. The high magnitude of the resistance noise in strongly disordered conductors is well explained, at least qualitatively, within simple percolation models, by the strongly nonuniform distribution of current density and the electric field. However, it is well known that simple percolation models, being ''geometrical,'' give no answer to the problem of the noise spectrum.

Voss[3] measured the low-frequency noise in Si inversion layers with an impurity band, created by Na^+ ions, at low temperatures. He found that the spectral density of the noise has $1/f$ frequency dependence and proved that it results not from surface mechanisms but is an intrinsic property of the hopping conduction. Shklovskii[4] developed the first theory of the hopping conduction noise for the case of nearest-neighbor hops. According to the main idea of this theory, the noise spectrum has $1/f$ frequency dependence due to the exponentially wide spectrum of transition frequencies of the one-electron hops between impurity centers. These frequencies strongly depend on the hopping distance r: $\nu = \nu_0 \exp(-2r/a)$, where a is the effective Bohr radius of the impurities, and ν_0 is a factor that weakly depends on the parameters of the hops (the effect of the change of the energy accompanying each hop has been neglected). In Ge doped by shallow impurities $\nu_0 \sim 10^{11}-10^{12}$ s^{-1}. It is well known[2] that the dc hopping conduction is determined by the so-called critical network (critical infinite cluster, CIC). It is formed by those impurities between which all distances do not exceed $r_c + a$, where r_c is the critical percolation radius.[2] The frequencies of hops within the CIC are of the order of or higher than $\nu_c = \nu_0 \exp(-2r_c/a)$. These hops contribute to the growth of the resistance noise with decreasing frequency f at $f \geqslant \nu_c$. At $f < \nu_c$ the charge carriers within the CIC quickly equilibrate, and the resistance fluctuations are produced by hops between the CIC, on one hand, and finite clusters located in its pores. As the frequency f decreases, the infinite cluster (IC) within which the occupation of the impurities can be considered as quickly equilibrating, grows at the expense of finite clusters. At each such frequency, the resistance fluctuations, i.e., the fluctuations of the number of charge carriers in the CIC, are produced by hops between this IC (its size depends on f) and the remaining finite clusters, which are separated from the IC by distances $r(f) = (a/2)\ln(\nu_0/f) > r_c$. The greatest relaxation times in this system correspond to the electron exchange between the IC

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and such finite clusters (or single impurities) that are almost isolated, i.e., the distance between them and their nearest neighbors belonging to the IC is much greater than the mean distance between impurity centers, $\sim n_D^{-1/3}$, where n_D is the concentration of impurities. Such great distances correspond to large ''voids'' with no impurities. According to the Poisson distribution, the probability of such a void is of the order of $\exp(-4\pi n_D r^3/3)$, i.e., is steeply falling with r at $n_D r^3 > 1$. This exponential function decays faster than $\exp(-2r/a)$ at $r > r_0 = 1/\sqrt{2\pi n_D a}$. Hence, even if at higher frequencies the noise spectral density grows with decreasing frequency f as $1/f$, it deviates from $1/f$ and saturates at such frequencies at which $r(f)$ becomes greater than r_0, i.e., at frequencies

$$f_{\min} = \nu_0 \exp\left[-\frac{2}{\sqrt{2\pi n_D a^3}} \right]. \tag{1.1}$$

The one-electron-hop model of hopping resistance noise was later analyzed in more detail.[5] Numerical calculations have been performed for practically achievable values of $n_D a^3$. The frequency dependence of the spectral density was found to follow a power law $S(f) \propto f^{-\gamma}$ but with the exponent γ considerably lower than 1. No frequency range with the $1/f$ spectrum was found within these calculations.

The frequency $f_{\min}$, at which, in this model, the power-law behavior of $S_R(f)$ turns out to be a constant, depends on $n_D a^3$. Practically, in the samples used in experiments on hopping conduction and noise, this quantity cannot be too small, otherwise the resistivity becomes immeasurably high.[4,5] Usually $(n_D a^3)^{-1/2} \leqslant 30$, hence the exponential function in the right-hand side (rhs) of Eq. (1.1) cannot be smaller than $\sim 10^{-9}$. Since $\nu_0 \sim 10^{11}-10^{12}\ \mathrm{s}^{-1}$, the lower-frequency limit for $1/f$ noise produced by one-electron hops is $\sim 10^1-10^2$ Hz. However, the experimental spectrum is $1/f$ at lower frequencies.[3,6] Moreover, if the noise is found to have a $1/f$ spectrum, no low-frequency limit of this frequency dependence has ever been found in any experiment.[1] It does not mean that $1/f$ has no lower limit: for all mechanisms that are at least qualitatively substantiated, this lower-frequency limit is practically not achievable.[1] We must conclude that the mechanism of the hopping conduction noise suggested in Ref. 4 works at intermediate, but not too low, frequencies.

The same qualitative conclusion is also true for the model suggested by Kozub.[7] In this model the resistance fluctuations are produced by hops within finite clusters that change the electrical potentials at the impurities belonging to the CIC and, consequently, modulate the conduction along the CIC. In real samples with not too small $n_D a^3$ the frequency range of this noise is limited from below.

The main problem in the physics of noise is to find what transitions and between what states determine the stochastic dynamics of the system in the given frequency range. First, only those transitions that occur between states that are populated at the temperature of measurement T contribute to the spectral density of noise. Baranovskii, Efros, Gelmont, and Shklovskii[8] found, by computer simulation of a simple model of the EG, several energy minima, i.e., metastable, or pseudoground, states, very close to the lowest-energy minimum that can be considered as the ground state. The same structure of the low-energy states was found also for a more realistic model of the Coulomb glass.[9] It was also found in Refs. 8 and 9 that the differences between the energies of the ground state and these metastable states are very small, usually less than or of the order of the characteristic energy of the Coulomb interaction at the mean distance between neighboring donor impurities, $E_C = e^2 n_D^{1/3}/\kappa$, where n_D is the concentration of the major shallow impurity, and κ is the dielectric permittivity of the host crystal. The ground state and these low-lying pseudoground states are called below ''valleys.'' During the last several years several groups have developed special computational algorithms for finding, within some simplified models of the electron glass of small size, more complete sets of the lowest excited states of this many-body system.[10–14] Presumably, these excited states include the pseudo ground states.

There is a second requirement: only those transitions that have long enough relaxation times contribute to the growth of the spectral density of noise at *low frequencies*. This means that the states between which the transition occurs must be separated by a high enough activation or tunnel barrier. The relaxation times of the low-lying excited states of a model EG have been studied in Refs. 10 and 11. The authors found some extremely slow relaxation processes and associated them with relaxation from metastable states of the EG. Transitions between states not separated by barriers do not contribute to the growth of the spectral density of noise at low frequencies.

The main idea of the present paper is that the transitions between low-lying energy valleys of the EG are a real source of $1/f$ hopping conduction noise. Each intervalley transition is, in general, a reconfiguration of a great number of electrons, which can occur even by one-electron hops, but by a great number of such hops. Therefore, we studied, using computer simulation, the properties of valleys relevant to the low-frequency stochastic dynamics of the EG. In Sec. II the model and the method of computer simulation are presented. In Sec. III the statistics of the valleys is found: their number versus the size of the EG, the differences between their energies, the differences between the electron arrangements in different valleys, the dependence of these differences on the EG size and their distribution. Despite the energies of the valleys being very close to one another they are separated by high barriers. In Sec. IV the calculation of the intervalley activation energies that determine the relaxation times and, ultimately, the low-frequency stochastic dynamics of the EG are presented, and the resistance noise produced by intervalley transitions is discussed. In Sec. V some qualitative conclusions are formulated.

II. THE MODEL AND THE METHOD OF COMPUTATION

Electron glass is a system of randomly arranged major and compensating impurities. For definiteness, we assume that they are donors (total number N_D) and acceptors (total number N_A), respectively. At a given arrangement of the impurities, each state of the EG corresponds to a specific distribution of $N_D - N_A$ electrons among N_D donor sites. Since the number of electrons in a definite ith donor, $N(i)$, can be only 0 or 1, the number of such states equals N_s

$=N_D!/[(N_D-N_A)!N_A!]$. For any representative sample of the EG this number is huge. Therefore the properties of this disordered many-body system are studied by numerical simulation. In the present paper all calculations have been performed for compensation ratio 0.5, i.e., $N_A=\frac{1}{2}N_D$.

The model of an EG used for computer simulations constitutes an infinite number of identical simulation cubes, each of which contains randomly arranged impurities: N_D donors and N_A acceptors. N_D-N_A electrons are placed onto the donors. Thus, in each simulation cube there are N_A negatively charged acceptors, N_A positively charged donors, and N_D-N_A neutral donors (electrons), and the arrangements of the impurities, charged and neutral, in all cubes are identical. All energies are expressed in units $E_C=e^2n_D^{1/3}/\kappa$, where n_D is any given concentration of donors within the scope of the model, all distances are expressed in units $n_D^{-1/3}$. The side of the simulation cube in these units is $N_D^{1/3}$. It was important to study an infinite system because otherwise one could guess that the low-lying valleys result from some effect of the surface of a finite and small sample, and are not a bulk effect.

Since the energy of the EG is the energy of Coulomb interaction of the ionized impurities, and the Coulomb interaction falls off slowly, the Ewald method of summation was used (for a recent review see Ref. 15). The calculated energy E is the Coulomb energy of the entire system per one simulation cube. It can be written as follows:

$$E=\frac{1}{2}\sum_{ij} q_iq_j[(1-\delta_{i,j})\psi_{ij}+\delta_{ij}\psi], \tag{2.1}$$

where i and j number all impurities within one simulation cube, $q_i=1$ for acceptors, $q_i=-[1-N(i)]$ for donors, $N(i)$ is the occupation of the ith donor by an electron [$N(i)=1$ for a neutral donor, $N(i)=0$ for an ionized, i.e., empty, donor]. The ''potentials'' ψ_{ij} and ψ are expressed in terms of Ewald sums:

$$\psi_{ij}=\sum_{\mathbf{R}} \frac{\mathrm{erfc}(\alpha r_{ij\mathbf{R}})}{r_{ij\mathbf{R}}} + \sum_{G\neq 0} \frac{\exp(-\pi^2G^2/\alpha^2)}{\pi VG^2}\cos(2\pi\mathbf{G}\mathbf{r}_{ij}) \quad (i\neq j),$$

$$\psi=\sum_{\mathbf{R}(\neq 0)} \frac{\mathrm{erfc}(\alpha R)}{R}+\sum_{\mathbf{G}\neq 0} \frac{\exp(-\pi^2G^2/\alpha^2)}{\pi VG^2}-\frac{2\alpha}{\sqrt{\pi}}. \tag{2.2}$$

The vectors $\mathbf{R}$ and $\mathbf{G}$ are the vectors of the cubic lattice and its inverse, respectively, $\mathbf{r}_i$ is the radius vector of the ith impurity in the simulation cube, $\mathbf{r}_{ij}=\mathbf{r}_i-\mathbf{r}_j$, $r_{ij\mathbf{R}}=|\mathbf{r}_{ij}+\mathbf{R}|$, $R=|\mathbf{R}|$, and V is the volume of the unit cube. The value of the parameter α is chosen to obtain the best convergence of the Ewald series.[15] The function erfc(x) is the complementary error function.

The quantities ψ_{ij} depend on the arrangement of the impurities only and are not affected by the variation of the donors' occupancies $N(i)$. Being calculated for a given random arrangement of impurities, they can be stored and used for calculation of the energy E and the Coulomb potentials at any site,

$$\phi(i)=\sum_{j(\neq i)} q_j\psi_{ij}+q_i\psi. \tag{2.3}$$

The electron hops affect only the ''charges'' q_i of the donors. The variation of the system's energy due to an electron hop from the donor i to an empty donor j equals

$$\Delta E=\sum_{i'(\neq i,j)} q_{i'}(\psi_{ji'}-\psi_{ii'}). \tag{2.4}$$

III. THE VALLEYS AND THE DIFFERENCES BETWEEN THEM

The EG valleys were found using the same procedure that was used in Refs. 8 and 9 [the only difference is the Ewald summation, Eq. (2.1)] and in simulations of other disordered many-body systems, e.g., spin glasses. Starting from a random distribution $N(i)$ of electrons among the donor impurities, the electrons are moved, one by one, from one donor to an unoccupied donor, irrespective of the distance between them, if this transfer lowers the total energy of the system. The state (or states) found by this procedure must satisfy two requirements: (1) no one-electron transfer from an occupied donor to an empty one, even across the entire system (not only to the nearest neighbor), can result in a transition to a state with lower total energy, i.e., the variation of the system's energy by any one-electron transfer is always positive, (2) the Coulomb potential $\phi(i)$ at any ith occupied donor must be lower than that at any empty donor, j. Different starting states, in general, yield different energy minima E_m. The so found energy minima that are higher than the lowest one, considered as the ground state, are metastable, or pseudo ground states (see below). Both the ground state and these metastable states may be called valleys. Since valleys are obtained by satisfying the two requirements listed above, they differ from other states of the EG, including other metastable states, by the presence of the Coulomb gap in the spectrum of one-electron energy levels.

1. The energies and the number of valleys. The obtained energies of the valleys E_m ($m=1,\ldots,N_v$) are always negative. Their absolute values $|E_m|$, depend on the specific random arrangement of the impurities and are greater, by a factor $\sim 2-3$, than those obtained without Ewald summation. The energy of the ground state per one donor, $|E_1|/N_D$, strongly fluctuates at $N_D=50$ (from ≈ 0.6 up to ≈ 1.4) and most frequently is around 1.1. As the size of the system grows the fluctuations become smaller and the mean value slightly greater. At $N_D=500$ the energy $|E_1|/N_D$ fluctuates by $\approx \pm 10\%$ around ≈ 1.3. One may guess that at $N_D \gtrsim 1000$ the size of the simulation cube is greater than the length that determines the Coulomb energy (screening length), i.e., in this respect the simulation cube is a macroscopic self-averaging system.

The number of valleys N_v for a given N_D strongly varies for different random arrangements of the impurities. For instance, for $N_D=400$ the number of valleys was found to vary from $N_v=4$ up to 106, for $N_D=500$ from 10 up to 176. It looks like N_v is not a self-averaging quantity. Nevertheless, one may anticipate that, in samples of sufficiently large size, the relative variation of N_v is small. Using our data we can

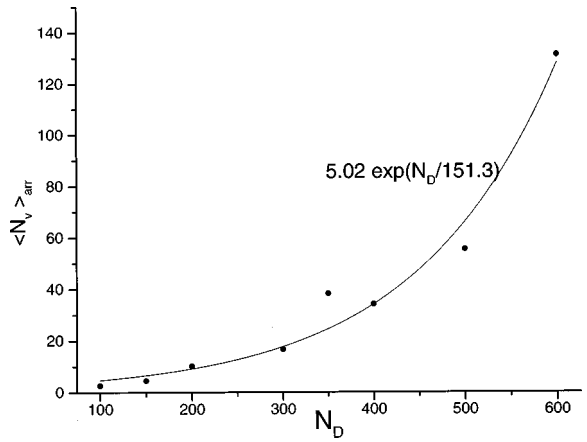

FIG. 1. The mean number of valleys $\langle N_v\rangle_{\rm arr}$ vs the size of the sample N_D. $\langle N_v\rangle_{\rm arr}$ is found, at each given N_D, by averaging over 10 different arrangements of the impurities. Exponential curve is a fit to the data.

neither prove this nor determine the size at which the crossover occurs.

There is an obvious trend: the greater the size of the system, N_D, the wider the distribution of N_v and the less probable are small N_v. Hence, on average, N_v grows with N_D. The dependence of the mean $\langle N_v\rangle_{\rm arr}$, averaged over 10 different arrangements of impurities at each N_D, on N_D is presented in Fig. 1. Exceptionally high numbers of valleys were found in one system with $N_D=400$ ($N_v=106$) and 5 systems with $N_D=500$ ($N_v=128$, 132, 144, 145, and 205).

2. The differences between the valleys. The greatest energy difference between valleys, $E_{N_v}-E_1$, is always $\sim E_C$ only. The mean distance between adjacent valley energies, $[E_{N_v}-E_1]/(N_v-1)$, for impurity arrangements with $N_v>1$, fluctuates significantly at given N_D: at $N_D=50$ it varies from ≈ 0.03 to ≈ 0.3, at higher $N_D=300-500$ it most often does not exceed ≈ 0.1, and is typically a few 0.01 (at $N_D=500$, <0.01). Much more obvious is the correlation between the mean separation of valley energies and the number of valleys, N_v, which is shown in Fig. 2. As one can see, it falls off with N_v: the valley spectra become more dense at greater N_v. In other words, the range of valley energies

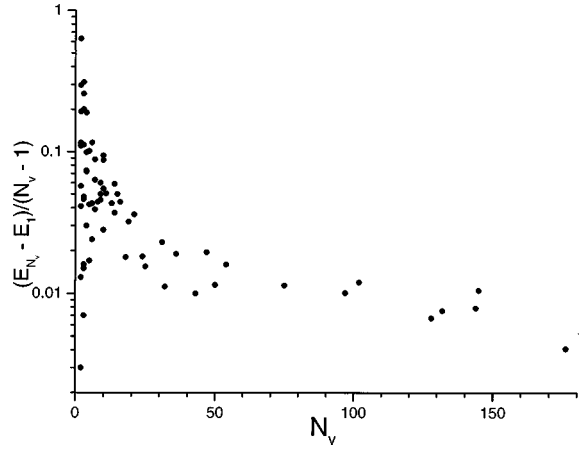

FIG. 2. The mean distance between adjacent valley energies vs the number of valleys N_v. The energies are in units E_C.

$E_{N_v}-E_1$ remains $\sim E_C$ irrespective of N_D but N_v, on average, grows with N_D, therefore the density of the valleys per unit energy also grows.

The valleys differ not only by their energies but, obviously, by the sets of the occupancy numbers $N_m(i)$, i.e., by the distribution of the N_D-N_A electrons among N_D donors. Some donors that are occupied in one valley (say, mth) are empty in another valley (say, the nth). Since the total number of electrons is the same in all valleys, the total number of donors whose occupation is different in the two valleys is always an even number. It is convenient to denote one-half of this total number as ΔN_{mn}. The electrons in the donors occupied in the mth valley and empty in the nth one may be called ''difference electrons;'' those donors that are empty in the mth valley and occupied in the nth one may be called ''difference holes.'' ΔN_{mn} is the number of ''difference electrons'' or ''difference holes.'' This number can be only $\geqslant 2$. Otherwise the transfer of the only ''difference electron'' of the higher valley to its only ''difference hole'' will result in the transition to the lower valley. This is impossible because, according to the method used to find the valleys (see above), no transition of only one electron may lower the energy of this state. The number ΔN_{mn} can be reduced, by one-electron hops, only if the energy of the system in the higher valley is first increased. This means that a transition from a higher valley to a lower one by one-electron hops always requires a nonzero activation energy (for the inverse transition the barrier includes also the difference of the energies of the two valleys).

By analogy with spin glasses, one can define, for pure states m and n, their overlap q_{mn} and the distance d_{mn} between them (analog of the Hamming distance):

$$q_{mn}=\frac{1}{N_D}\sum_i\,[2N_m(i)-1][2N_n(i)-1]=1-2\Delta N_{mn}/N_D,$$

$$d_{mn}=\frac{1}{N_D}\sum_i\,[N_m(i)-N_n(i)]^2=1-q_{mn}=2\Delta N_{mn}/N_D. \tag{3.1}$$

The greater ΔN_{mn} is the greater is the Hamming distance between them in the space of the states and the smaller is the overlap between the two valleys.

For a definite arrangement of impurities, the numbers ΔN_{mn} vary within some range. Of course, if any 3 valleys are taken, any of the three differences ΔN_{mn} must be smaller than or equal to the sum of the two others, that is, the triangle rule must hold. In fact, the equality $\Delta N_{23}=\Delta N_{12}+\Delta N_{13}$ occurs if the impurities whose occupation is different in valleys 2 and 1, and the impurities whose occupation is different in 3 and 1, do not coincide. An interesting problem is as follows: do the differences ΔN_{mn} or, equivalently, the Hamming distances between the valleys, obey also the condition of ultrametricity? Ultrametricity means that for any 3 valleys, l,m,n, the following condition is satisfied: $\Delta N_{mn}\leqslant\max\{\Delta N_{nl},\Delta N_{lm}\}$. It implies that 2 of the 3 differences are equal and are greater than or equal to the third one. If this condition holds the structure of the states is hierarchical (for a review see Ref. 16). The results show that sometimes the last inequality is satisfied (for instance, the differences are 3,

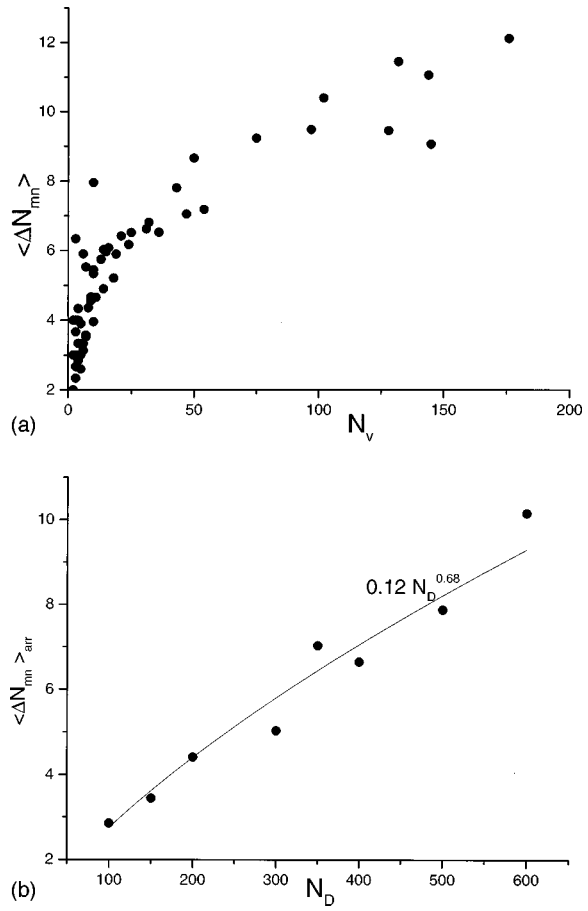

FIG. 3. (a) The mean number of ''difference electrons'' $\langle \Delta N_{mn} \rangle$ at definite arrangements of impurities vs the corresponding number of valleys N_v. (b) The mean number of ''difference electrons'' $\langle \Delta N_{mn} \rangle_{\text{arr}}$ vs N_D. $\langle \Delta N_{mn} \rangle_{\text{arr}}$ is obtained by averaging, at each given N_D, over 10 different arrangements of the impurities. The continuous curve is a power-law fit to the data.

3, and 2). However, often all three numbers are different (for instance, 2, 4, and 5), and even if two (of three) numbers are equal, they may be smaller than the third one.

Like the number of valleys, the numbers ΔN_{mn} depend on the specific arrangement of the impurities but their relative variations are much less than those of N_v. The mean $\langle \Delta N_{mn} \rangle$ is obtained by averaging over all ΔN_{mn} at a given impurity arrangement. This mean value grows, on average, with corresponding N_v, as shown in Fig. 3(a). The mean difference, $\langle \Delta N_{mn} \rangle_{\text{arr}}$, averaged over a number of random arrangements of the impurities with given N_D, grows with N_D [Fig. 3(b)]. Obviously, $\langle \Delta N_{mn} \rangle_{\text{arr}}$ cannot grow with N_D faster than linearly. The results presented in Fig. 3(b) are in agreement with this expectation.

A typical distribution of ΔN_{mn} at small N_D is shown in Fig. 4(a). The most frequent value of ΔN_{mn} is the minimum one, i.e., 2. At higher N_D the typical distribution is different [Figs. 4(b) and 4(c)]: the smallest difference $\Delta N_{mn}=2$ and the maximum ΔN_{mn} are the least probable; the most probable are the numbers in the middle between the extreme ones. As a whole, the distribution is roughly a normal one. One can see also in Figs. 4(a)–4(c) that the maximum ΔN_{mn} is $\sim 0.05 N_D$. However, even if ΔN_{mn} is a small portion of N_D, in a macroscopic sample, for any part of the sample with large enough size this number is great.

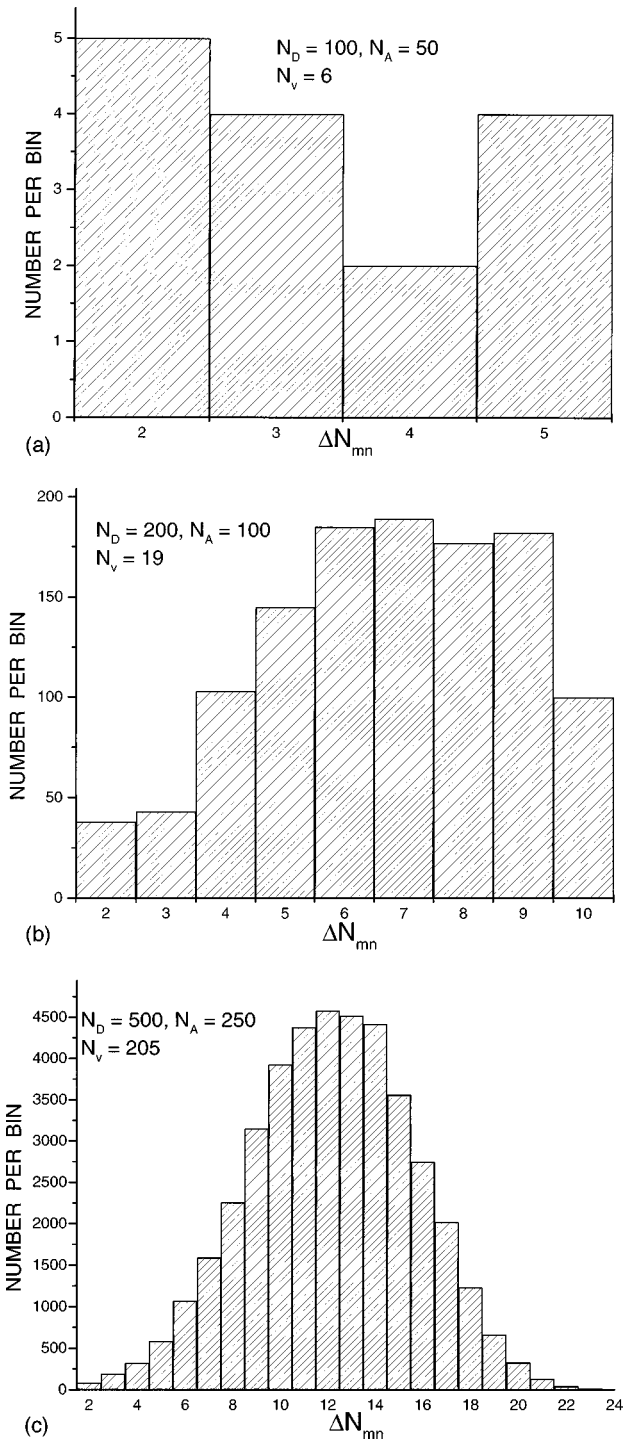

FIG. 4. The histograms of the numbers of ''difference electrons'' ΔN_{mn} for three samples with (a) $N_D=100$, $N_v=6$; (b) $N_D=200$, $N_v=19$, and (c) $N_D=500$, $N_v=205$.

The valleys are found by descent from states with random occupation of donors and, consequently, with high energy. The number of such initial states, N_{init}, must be high enough and it must grow with the size of the system, N_D. By increasing this number in excess of, say, N_D (for $N_D \leqslant 600$) no increase of the number of valleys N_v is usually found. However, for our purpose and for qualitative conclusions, it is not very important if one more or a few other valleys exist close to already found.

The most important problem is whether an energy mini-

mum exists that is considerably deeper than the already found group of valleys with close energies. In other words, does an energy gap $\gg E_C$ exist between the ground level and the next valleys? If such a minimum exists and if it is always a single one (with no close valleys) then the stochastic motion in the form of intervalley transitions does not exist in the EG at low temperatures. In numerous calculations for systems of different sizes we have never found such a minimum whose energy is lower than the energies of a group of valleys by more than the characteristic difference of the valleys' energies. During the procedure of finding the energy barriers between the valleys (see below), the system was frequently checked: into what state does it descend after being allowed to make hops to states with lower energies? The system was never found in any state deeper than the already found lowest valley.

IV. THE ENERGY BARRIERS FOR INTERVALLEY TRANSITIONS AND THE RESISTANCE NOISE SPECTRUM

We show below that, despite the variations of the valley energies at a fixed impurity arrangement being small, the valleys are separated by energy barriers of various heights. In a macroscopic system, these barriers may vary in a very broad range. Just such broad and smooth distribution of activation energies is well known to yield a $1/f$ spectrum of noise down to very low frequencies.

In an EG the notion of the energy barrier between valleys becomes definite only if a definite restriction on the number of electrons making simultaneously a hop and on the length of the hops is imposed. We assume that the hops are one electron only and each hop is performed from an occupied donor to the nearest empty one or to one of the few (for instance, 2–3) nearest empty donors. It is worth noticing that the intervalley barriers are always nonzero (see Sec. III).

The transition from one valley, m, to another one, n, is a sequence of electron hops that results, ultimately, in the ''recombination'' of the difference electrons and holes. Each such sequence is a path, in the space of states, connecting the two valleys. It has some maximum energy. The activation energy is the minimum of these maximum energies:

$$E_{mn}^{(\mathrm{act})} = \min_{\text{paths } m\to n} \{ \max_{\text{along the path}} E \}. \tag{4.1}$$

For any EG system with more than one valley, i.e., with great enough $N_D \geqslant 100$, the number of paths from one valley to another is very great. Even if, for not too low noise frequencies and hence not too high activation energies, the calculations are confined to low-energy intermediate states only and are using, for instance, the numerical methods elaborated in Ref. 11, the total number of possible paths from one valley to another through these states remains great. This makes impossible a rigorous calculation of barrier heights satisfying Eq. (4.1) and compels one to limit the number of paths checked. Of course, the results obtained in this way are merely estimates of the true activation energies. The goal is to obtain the correct order of their magnitudes and the characteristic features of their distribution. Several algorithms have been tested. The one that yields the lowest intervalley barriers is presented below.

Two valleys, say, m and n, are specified. One valley, m, is considered as the initial one, and the goal is to find the energy barrier for the transition to the target valley, n. Before each electron hop is performed, a difference electron (DE) or a difference hole (DH) is randomly chosen among all DE and DH. The DH (DE) that is nearest to the chosen DE (DH) is then found. Thus, a pair DE-DH is chosen. Then a number (M) of such, nearest to the chosen DE, empty donors that are closer to DH are found. Similarly, M such, nearest to the chosen DH, filled donors that are closer to DE are also found. A hop of the DE or the DH to any of its corresponding M chosen neighbors decreases the distance DE-DH (the results presented below are obtained with $M=3$). Of all these possible hops (in general, $2M$ hops) one is actually performed that results in the minimal change of the system's Coulomb energy ΔE. This requirement makes less probable a significant difference between the calculated energy barrier and the true one.

After each hop the set of donor occupation numbers, $N(i)$, and the set of difference electrons and holes are updated. The energy of the system in the new state is stored. Then a new pair DE-DH is randomly chosen. At $\Delta N_{mn} \gg 2$, this new pair, most probably, is different from the previous one. Some hops result in the ''recombination'' of a difference electron with a difference hole. Their number gradually tends to zero. This means that the intervalley transition is performed. Then the difference, $\Delta E_{\max}$, between the maximum energy of the system along the transition path and the initial energy E_m is found and stored.

Variation of the sequence of random numbers used to choose the pairs DE-DH (by changing the initial number of the random number generator) generates a different transition path between the same two valleys, and, in general, a different value of $\Delta E_{\max}$ is obtained. The procedure is repeated many times (in the present calculations, no less than 20 times for each pair of valleys). The lowest among the so obtained $\Delta E_{\max}$ is taken as the energy barrier, ϵ_{mn}, for the *direct* transition from the valley m to the valley n. This algorithm leaves room for various generalizations.

When the number of valleys $N_v > 2$ the actual (easiest) path from one valley, m, to another one, n, that is, the path with minimal energy barrier, may pass through other, intermediate, valleys. The *actual* activation energies $E_{mn}^{(\mathrm{act})}$ must satisfy at least the following condition for the case of one intermediate valley:

$$E_{mn}^{(\mathrm{act})} \leqslant \max\{E_{ml}^{(\mathrm{act})}, E_{ln}^{(\mathrm{act})}\} \quad \text{for any } l \neq m,n. \tag{4.2}$$

Here l numbers all valleys that can serve as intermediate ones. The condition (4.2) is similar to the condition of ultrametricity (see Sec. III).

In order to satisfy this condition the direct energies ϵ_{mn} found by the procedure outlined above have been subjected to the next procedure. For any definite pair of valleys (m,n) the value of $\max\{\epsilon_{ml}, \epsilon_{ln}\}$ has been found for all intermediate valleys $l(\neq m,n)$. The minimum value of these maxima was compared with ϵ_{mn}, and the smallest of these two quantities was recorded as $E_{mn}^{(\mathrm{act})}$. This procedure significantly reduces the calculated energy barriers as compared with the direct barriers, ϵ_{mn}.

The distributions of the activation energies $E_{mn}^{(\mathrm{act})}$ found by this method, for three definite samples with $N_D=100$, $N_D=200$, and $N_D=500$, are shown in Figs. 5(a)–5(c). The samples are the same for which data are presented in Figs. 4(a)–4(c). The most important conclusion is that the interval within which the energy barriers vary grows, on average, with the system's size N_D. For $N_D=100$ the barriers between valleys are perhaps of the same order as the typical barriers for intravalley transitions, that is, for these small sizes one cannot expect a great difference between the two types of transitions.

At higher N_D [Fig. 5(b) and 5(c)] the distribution of the activation energies roughly resembles that of the differences ΔN_{mn} for the same sample [Figs. 4(b) and 4(c)]. In particular, the distributions are the highest in the middle range between the minimum and maximum values of $E_{mn}^{(\mathrm{act})}$ and ΔN_{mn}, respectively, the extreme values being the least probable. It shows that, despite the fact that no direct (deterministic) relationship between $E_{mn}^{(\mathrm{act})}$ and ΔN_{mn} exists in this strongly disordered system, they are correlated: on average, the greater ΔN_{mn} and, consequently, the Hamming distance between two valleys is [Eq. (3.1)], the higher the barrier between them (this result is similar to that obtained, using a different method, by Vertechi and Virasoro[17] for the Ising spin-glass model with infinite range interactions). The correlation between $E_{mn}^{(\mathrm{act})}$ and ΔN_{mn} is illustrated in Fig. 6. One can see that, on average, one pair ''difference electron—difference hole'' contributes $\sim(0.06-0.15)E_C$ to the energy barrier, which is a quite plausible result.

If the method were exact the difference of energy barriers for transitions between two valleys, from m to n and from n to m, i.e., $E_{mn}^{\mathrm{act}}-E_{nm}^{\mathrm{act}}$, must coincide with the difference of the valley energies, E_n-E_m. Due to the inaccuracy of the method used these two differences, in general, do not coincide. Since, for each pair of valleys, m and n, the energy barriers E_{mn}^{act} and E_{nm}^{act} are calculated independently, the ratio $[E_{mn}^{\mathrm{act}}-E_{nm}^{\mathrm{act}}-E_n+E_m]/E_{mn}^{\mathrm{act}}$ can be considered as a measure of the accuracy of the method of energy barriers' computations. This ratio was found for all pairs of valleys. The absolute values of the ratios are typically lower than 0.25, and seldom exceeded 0.3. For many pairs of valleys the ratio was found to be $\pm$ computer zero.

An intervalley transition results in a change of the set of occupancy numbers $N(i)$. One may expect that this change produces a fluctuation of the specimen's hopping conductance, $\delta G(t)$. It stems, first of all, from the change of some resistances of the Miller-Abrahams network.[2] The conductance g_{ij} (inverse resistance) connecting two donor sites i and j (of this network) is nonzero only when one of these sites is occupied by an electron and another one is empty. The change of the occupation number of a donor impurity results in a drastic change of several conductances g_{ij} connecting this impurity to its neighbors. The specimen's conductance G is changed also due to the variation of the Coulomb potentials and to electron correlation effects.[2] No symmetry restriction is known that can reduce the hopping conduction fluctuations produced by intervalley transitions to zero. If they are not zero the spectral density is expected to grow with decreasing frequency f. In fact, according to the data presented in this section, the range of intervalley activation energies grows with the size of the system. One can therefore expect that, in a macroscopic sample, for any achievable low frequency f such intervalley transitions and, consequently, such conductance fluctuations exist which have high enough relaxation times $\gtrsim\omega^{-1}$. The low-frequency noise spectrum is expected to be of $1/f$ type due to the smooth distribution of intervalley activation energies in a broad range (see, for instance, Ref. 1). At low temperatures, the thermal motion of a Coulomb system in equilibrium and

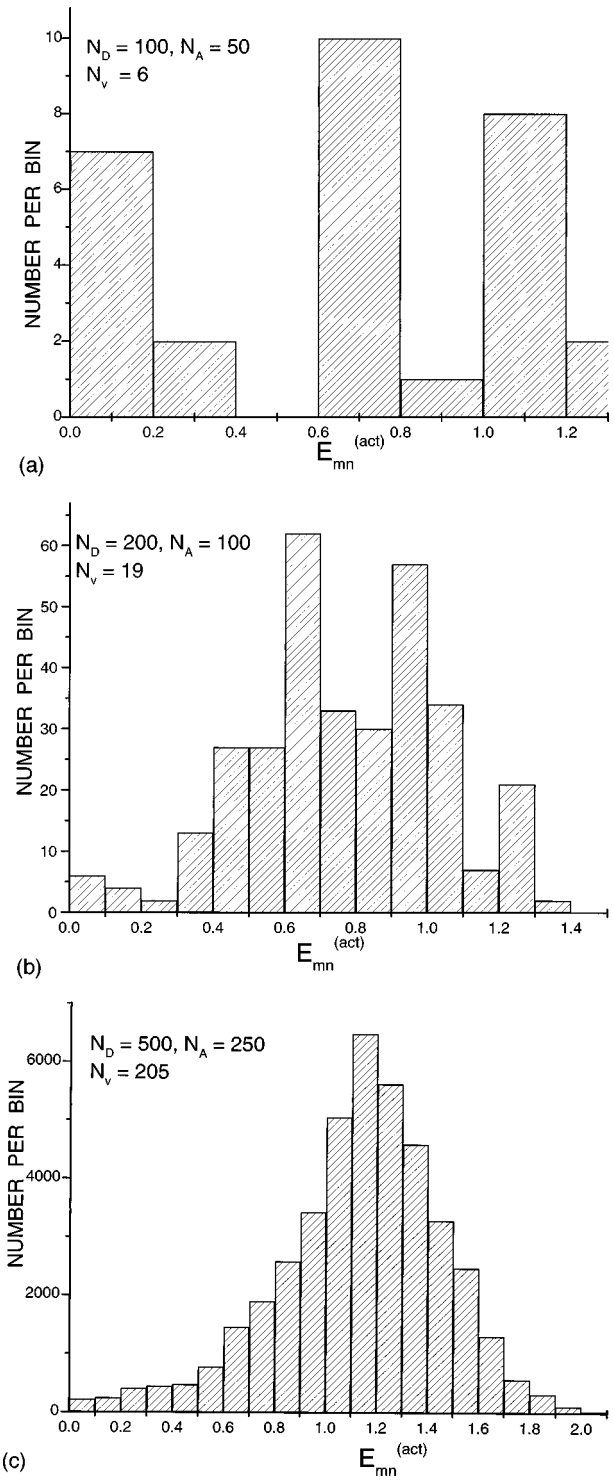

FIG. 5. The histograms of the energy barriers $E_{mn}^{(\mathrm{act})}$, in units E_C, for the same three samples as in Fig. 4.

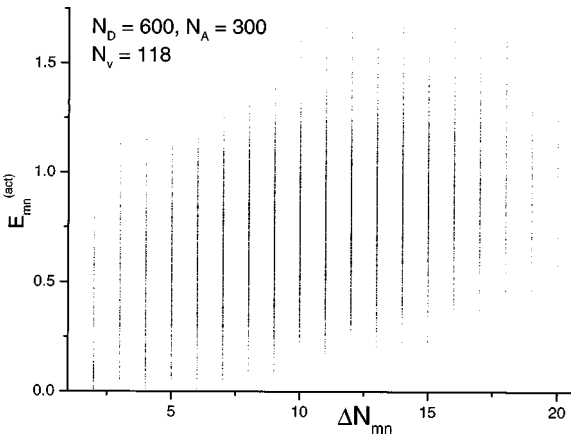

FIG. 6. The energy barriers $E_{mn}^{(\mathrm{act})}$ vs the number of difference electrons, ΔN_{mn}, for a sample with $N_D=600$, $N_v=118$. In those ranges of the barrier energy where the density of the scattered dots is high they coalesce and create continuous vertical lines.

under a small electric field is confined to a small part of the total number of states. The transition from one valley to another one with different configuration of electrons may result therefore in a significant change of the sample's resistance.

Despite the fact that the noise spectrum is qualitatively understood, the problem of its *magnitude* is not solved here. First, calculations of the conductance change by direct numerical modeling are lacking sufficient accuracy. Second, calculations based on the Markov equations for the hopping conduction (see Appendix) seem impossible due to the great number of the system's states involved. Therefore in this section only qualitative considerations are presented.

The problem of the magnitude of the conduction noise can be considered as a part of the general problem of the effect of a variation of the donors' occupancies, $N(i)$, in a given sample on its hopping conductance. It is somewhat similar to the problem of damage spreading (see Ref. 18 and references therein), i.e., the evolution in time of a relatively small initial difference of $N(i)$ (like the difference between two valleys) in two samples with identical impurity arrangements.

V. CONCLUSIONS

The low-energy EG landscape has a valley structure.[8,9] As was found in Sec. III, for all N_D used in computations, the number of valleys, N_v, in samples of the same size N_D strongly varies for different arrangements of the impurities. On average, N_v steeply grows with the size of the sample (Fig. 1).

In all samples for which the calculations have been made ($50 \leqslant N_D \leqslant 600$), the total range of valley energies is of the order of the characteristic Coulomb energy E_C. Since the number of the valleys rapidly grows with N_D, the mean energy gap between two adjacent valleys drops with N_D and N_v (Fig. 2). If this tendency holds at even higher N_D, the distribution of the valleys is expected to be a relatively very narrow peak near the ground energy. It can be considered as a highly degenerate ground level.

The difference between the electron arrangements in two valleys, m and n, is described by the number of ''difference electrons'' or ''difference holes,'' ΔN_{mn}, which is proportional to the Hamming distance between these two states [Eq. (3.1)]. On average, ΔN_{mn} grows with the size of the system and with the number of valleys in the sample (Fig. 3). The distribution of ΔN_{mn} in multivalley samples (high $N_D \gtrsim 200$) has a typical form similar to a normal distribution, the extreme values being the least probable [Figs. 4(a)–4(c)]. The maximum values of ΔN_{mn} do not exceed $\sim 0.05 N_D$.

The energy barriers for intervalley transitions have been estimated by a computer simulation of the ''recombination'' of the ''difference electrons'' with the ''difference holes'' (Sec. IV). Despite the fact that the valley energies are almost degenerate, the energy barriers for transitions between them are not small and are distributed in a range whose width grows with the size of the system. The energy barriers between valleys and the corresponding numbers of difference electrons are correlated. In particular, the distribution of the energy barriers in a given sample is similar to that of the differences ΔN_{mn} [Figs. 5(a)–5(c)]. For high N_D, both resemble a normal distribution. The growth of the energy barriers' range is determined by two major patterns: (1) the sublinear growth of typical values of ΔN_{mn} with N_D [Sec. III and Fig. 3(b)]; (2) the typical contribution of one ''difference electron—difference hole'' pair to the energy barrier is $\simeq (0.06-0.15) E_C$.

The intervalley transitions satisfy both requirements for those transitions that are a source of low-temperature low-frequency noise: (1) in any macroscopic sample and on any scale higher than several hundred major impurities there are valleys whose energy separation from the ground state is many times smaller than the thermal energy $k_B T$ at the temperature T of the experiment; (2) despite the valley energies being very close, the valleys are separated by high activation barriers whose magnitude grows, on average, with the size of the system considered. Therefore, these transitions definitely contribute to the growth of the spectral density of the hopping conduction noise with decreasing frequency at low frequencies. Due to the broad and smooth distribution of activation energies the spectrum of this noise is expected to be of $1/f$ type. However, the intervalley transitions are not expected to be the only ones that produce fluctuations of the hopping conductance in EG.

The slow (''glassy,'' nonergodic) stochastic dynamics that generates the $1/f$ hopping conduction noise manifests itself also in other low-temperature phenomena: slow relaxation of the Coulomb gap after injection of extra charge carriers at low temperatures[19] and slow field effect and persistent photoconductivity.[20,21] The intervalley transitions may contribute to these phenomena as well.

The hopping conduction noise is a particular example of noise in many-body systems with a strong frozen-in disorder. Despite all differences between two such systems, EG and spin glass, the low-frequency stochastic dynamics of these two systems are similar (for reviews of noise in spin glasses see Refs. 22 and 1). The fact that transitions between low-lying (created by the disorder) metastable states play an important role in two such systems, makes very plausible the idea that in other similar systems, including those in which the low-energy landscape has not yet been studied, just such transitions determine the low-frequency stochastic dynamics. Similar low-frequency stochastic dynamics may be found

also in proton glasses, in strongly disordered semi-insulating granular metallic films, and some other systems with frozen-in strong disorder.

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APPENDIX

Since the stochastic dynamics of the EG through hops is a Markov one the problem of hopping conduction noise can be formulated in terms of the eigenvectors and eigenvalues of the matrix of Markov equations for the variation of the probability, $P(\alpha,t)$, to find the system in a state α at the time t:

$$\frac{\partial P(\alpha,t)}{\partial t}=-\sum_{\beta}\Gamma_{\alpha\beta}P(\beta,t), \tag{A1}$$

where the matrix

$$\Gamma_{\alpha\beta}=\delta_{\alpha\beta}\sum_{\gamma}W(\alpha\gamma)-W(\beta\alpha), \tag{A2}$$

and $W(\alpha\beta)$ is the probability of transition, per unit time, from the state α to the state β.

It is well known that the eigenvectors $\Psi_k(\alpha)$ and eigenvalues λ_k of the matrix $\Gamma_{\alpha\beta}$ determine the distributions $\Psi_k(\alpha)\exp(-\lambda_k t)$, which decay in time following a purely exponential law. The eigenvector corresponding to the lowest $\lambda_0=0$ (no decay) is simply the set of the equilibrium probabilities of the states: $\Psi_0(\alpha)=w_\alpha$. All other eigenvectors ($k>0$) are relaxation modes, and the corresponding λ_k are their inverse relaxation times. These modes satisfy the relations

$$\sum_{\alpha}\Psi_k(\alpha)=0,\quad \sum_{\alpha}\Psi_k^2(\alpha)/w(\alpha)=1. \tag{A3}$$

The first relation means that any such $\psi_k(\alpha)$ is a perturbation of the equilibrium population of states that is an increase of population of some states at the expense of some other states. The second relation is the normalization equation.

The spectral density of the conductance fluctuations can be expressed in terms of these relaxation modes:

$$S_G(f)=\sum_{k>0}G_k^2\frac{4\lambda_k}{\omega^2+\lambda_k^2},\quad G_k=\sum_{\alpha}G(\alpha)\Psi_k(\alpha), \tag{A4}$$

where $G(\alpha)$ is the conductance in the state α of the system. Part of these relaxation modes may be associated with intervalley transitions. The low-frequency noise is determined by those $\lambda_k\neq 0$ that are sufficiently small for the given frequency f, i.e., $\lambda_k\leqslant\sim\omega$ and for which the corresponding G_k is significant. If the distribution of $\ln\lambda_k$, i.e., the activation barriers or tunnel exponents, is smooth in a broad range the spectral density $S_G(f)$ is of $1/f$ type.

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