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LETTER TO THE EDITOR

A new calculation of $1/f$ noise in disordered systems with hopping transport

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Abstract. The noise spectrum in hopping conduction systems is known to be closely related to the conductivity, as shown by scaling relationships between the two. The frequency-dependent conductivity and DC conductivity are related by scaling formulations as well. Percolation theoretical frameworks, such as used here, generate automatically relationships between frequency-dependent and DC transport properties. Under application of a DC field, the power spectrum of the (flicker) noise is usually inversely proportional to the frequency, proportional to the square of the applied field and, in crystalline germanium at low temperature, T, it has been found to be independent of T. Charge transport is traditionally represented in terms of a random impedance network in which all pairs of sites are connected by resistors. By considering cluster polarization effects of large clusters of resistors on the critical, blocking resistances on the percolation path, it is shown here that hopping conduction systems at low temperatures should generate a universal flicker noise proportional to the applied field squared, the inverse of the frequency, and independent of T. The cluster polarization effects considered arise from charge transported through large numbers of resistors in sequence, a process which, as it turns out, appears to generate much more visible effects in the noise than in the conductivity.

1. Introduction

1.1. *Relationship with conduction*

The source of $1/f$ noise, observed in a wide variety of condensed matter, has been debated without resolution [1, 2]. The wide range of systems in which power spectra appear to approximate a $1/f$ dependence (or $f^{-[1+\delta]}$, $|\delta| < 1$) has suggested to many observers the relevance of an underlying mechanism of great generality. Although the present work is designed to address *directly* only the noise spectrum in specific hopping conduction systems, it will nevertheless not be possible either to ignore this fundamental uncertainty or to avoid controversy regarding the best treatment of disorder.

Experimentally, the power spectrum, $J(\omega)$, of the noise is generally [2] determined in the presence of a DC field, F. The contribution, $\Delta J(\omega)$, to the power spectrum resulting from the application of an external field is usually $\Delta J(\omega) \propto F^2/\omega$. In impurity conduction in crystalline germanium experimental results [3] show that $\Delta J(\omega) \propto F^2/\omega$ is independent of temperature, T, as well. I try to clarify this result here.

Hopping conduction systems have been the prototypical systems for application of percolation theory to transport [4]. Such percolation theoretical formulations have recently been shown [5–8] to handle very low frequency conduction phenomena which are not well suited for treatment in effective-medium theories [9]. Thus a significant additional

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motivation is provided for application of percolation theoretical treatments to calculations of noise in hopping systems since it is precisely the low frequency portion of the noise spectrum which is of interest here.

In order to apply percolation theory, otherwise known as critical rate analysis, conduction must proceed primarily along paths of low resistance in an otherwise highly resistive random network. These paths are constructed [4] from random associations of relatively low resistance resistors. The blocking resistance on these paths has the value R_c, which is calculated by applying the condition [10],

$$\int_0^{R_c} W(R)\mathrm{d}R = \alpha_c \tag{1}$$

with α_c a number, in a well known example [10] equal to 2.7, and $W(R)\mathrm{d}R$ the number of resistances connected to an arbitrary site with resistance values between R and $R+\mathrm{d}R$. This condition states essentially that emplacement into the network of all resistors with resistance values less than or equal to R_c just generates an interconnected, infinitely long path. In this type of treatment the DC conductivity, $\sigma_{DC} \propto 1/R_c$. In strongly inhomogeneous systems scaling of the AC conductivity with the DC conductivity and a critical frequency, ω_c, proportional to the DC conductivity results because a cross-over from local (at high frequencies) to non-local relaxation occurs [11] at ω_c. The magnitude of the conductivity is pinned to the DC value by the same percolation condition on relevant resistance values at this frequency. Both R_c and ω_c will turn out to be relevant to formulations of $\Delta J(\omega)$ as well. Together with the proposition below, these scaling arguments provide a basis for understanding recent scaling formulations of the noise spectrum which involve the ohmic conductivity [12].

1.2. Basis for calculations and new perspective

The calculations given are based on the (existing) proposition [13] that $1/f$ noise originates from alterations in the resistance of the DC network produced by polarization charge transport. The difference between the current and previous calculations [13] is that I treat large-scale cluster polarization currents. I use expressions for the current, resistance and capacitance of such chains which were derived originally to calculate the polarization of polymers. This choice of theoretical inputs is compatible with a substitution of the backbone portion of the cluster for the entire cluster. The currents relevant for the present calculations involve charge transport at low frequencies through long chains of resistors over and above the charge transported separately through individual resistors on time scales derived from the individual resistance values. This type of cluster polarization has been shown to be responsible for additional contributions to the real part of the dielectric constant at frequencies below ω_c, corresponding to critical percolation [5–8] (with prior [14] and subsequent [15] agreement with experiment). Such cluster polarization effects may be important in a wide variety of insulators which have a non-zero DC conductivity at finite temperatures due to hopping conduction. The topology of random impedance networks including the connectivity of the individual portions of the network plays a role in the calculations; these particular aspects may be relevant for $1/f$ noise in a wider range of systems than those considered here.

2. The impedance network representation

The Nyquist theorem for the noise in a resistive circuit element was derived [16] in an inductive picture of conduction (appropriate to e.g. metallic conduction) in which the correlation time of a random current contribution is determined by a scattering time [5]. It may be equally applied to capacitive phenomena such as hopping conduction with the correlation time replaced by a relaxation time. The Miller–Abrahams (MA) [17] random impedance network which the following calculations exploit, is equivalent to the regime of ohmic transport described by the coupled set of first-order differential equations,

$$\mathrm{d}f_i/\mathrm{d}t = \sum_j f_j w_{ji}[1 - f_i] - w_{ij}[1 - f_j]. \tag{2}$$

f_i is the probability that site i is occupied (supposed equal to the mean occupation in the case of Fermi statistics) and w_{ij} is the conditional probability per unit time that, given site i is occupied and site j is empty, an electron can tunnel from site i to site j. Both the f and the w in this equation are exponential functions of random variables related to site energies, while only the w are exponential functions of site separations [17]. Stochastic microscopic transport described by this system of first order equations, clearly consistent with microscopic exponential decay of charge fluctuations, may be equivalently represented in terms of a random impedance network in the ohmic regime [17, 18]. This MA impedance network consists of resistors and capacitors, whose connections with (2), physical interpretations and values in the ohmic transport regime are given below. Later calculations involving non-ohmic effects are based on lowest-order (in the applied field) modifications of the ohmic network.

The individual resistances describe [17] the difficulty of transferring charge from e.g. site i to j (resistor R_{ij}) while the capacitors describe [18] the ability to alter the charge on site i (capacitor C_i). The R are connected between each pair of sites, $i;j$, while, for purposes of calculating the AC conductivity, the C are connected between each site and a generator of the external potential. The values,

$$R_{ij}^{-1} = \frac{e^2}{kT} f_i w_{ij}[1 - f_j] \tag{3}$$

and

$$C_i = \frac{e^2}{kT} f_i[1 - f_i] \tag{4}$$

of the resistances and capacitances are obtained through the linearization of (1) in an applied field. The statistical occurrence of the individual resistors is given in terms of the equilibrium occurrence of the w_{ij} and f_i. The relaxation time [19, 20], τ, of the subunit consisting of the sites i and j is $R_{ij}C_iC_j/(C_i + C_j)$. In the system studied here, variable range hopping [21], the typical resistance lengths are $a(T_0/T)^{1/4}$, and typical site energies are confined to within $kT(T_0/T)^{1/4}$ of the Fermi energy. Here, T_0 is a reference temperature related to the density of states, and a is the localization radius of the electronic wave functions.

The stochasticity in the microscopic transport is an obvious choice for generating the noise in the resistors, R_{ij}. Provided whatever charge transport involving i and j is restricted to occur between these sites (e.g. at high frequencies if all other resistors connected to either site i or j are much larger) the relaxation of charge fluctuations on these two sites can be described by the single differential equation appropriate to an RC series network, i.e. (2) with j restricted to a single site. The power spectrum of a resistor connecting such a pair of sites is well approximated by the Nyquist formula [16],

$$J(\omega) = RkT/\pi \qquad \omega \leqslant \tau^{-1} \tag{5}$$

$$J(\omega) = 0 \qquad \text{otherwise} \tag{6}$$

with τ the relaxation time given above. The reason why this representation is adequate for the present purpose is that the equivalent resistors in such strongly disordered systems are exponential functions of random variables. The logarithmic frequency scale appropriate for 'strong' disorder compresses $J(\omega)$ sufficiently that the distinction between the exact form (a Lorentzian) and a sharp cut-off at a maximum frequency is unimportant when the superposition of power spectra of independent resistors is calculated. Such a superposition generates an approximate $1/f$ noise spectrum at frequencies greater than ω_c whenever the individual resistance values are exponential functions of random variables [22, 23]. At ω_c the contributing resistance values are R_c, and $J(\omega_c) \approx kTR_c \propto 1/\sigma_{DC}$. The approximate result, $J(\omega) \propto 1/\omega$ for $\omega > \omega_c$ is consistent with the scaling formulation, $J(\omega) \propto [(\omega_c)/(\omega)]1/\sigma_{DC}$. For $\omega < \omega_c$ resistors $R < R_c$ do not contribute substantially to an unstimulated noise, and potential fluctuations due to resistors (or clusters of resistors) with $R > R_c$ are either shorted or screened by the smaller resistors so that $J(\omega)$ becomes constant. For calculations of the flicker noise at low frequencies we will be interested nevertheless in long chains of resistors $R < R_c$, and their interaction with the DC current. The transport/relaxation characteristics of 1D chains were determined by Pollak and Pohl [24]. The relaxation time of the chain is roughly [24],

$$\tau_{N,R} = N^2RC/\pi^2 = NRNC/\pi^2 \tag{7}$$

explicitly demonstrating the physical result that N times the charge per characteristic resistor must be transferred through N times the number of resistors. The factor π^2, appropriate for large N, is extraneous in the limit $N \to 1$. Equation (7) actually gives the largest relaxation time appropriate for such a chain of N resistors and $N+1$ capacitors; the smaller relaxation times correspond to charge transport through portions of the chain [24], and their effects can show up at different frequencies (different N). Thus the enhanced relaxation time reflects that the currents denoted as cluster currents account for additional transfer of charge (not just the sum of the charges transferred through individual resistors) on a much longer time scale. Consistent with the transport of charge through N resistors is the observation [24] that the equivalent resistance of the chain is NR.

3. Application of a DC field

As for non-linear conduction, effects due to application of a DC field are many and varied. Identifying the most important effect(s) can be aided by comparison with experiment. Individual resistors change their resistance value, clusters of resistors change their cluster resistance values and relaxation times, and fluctuations in both the polarization and DC currents influence each other.

The addition of a DC field reduces the relaxation times of individual resistors and clusters of resistors [24]. To lowest order, this reduction is by a term proportional simultaneously to the square of the field and the zero-field relaxation time. This effect is stronger for clusters than for individual resistors (and thus is larger at low frequencies) because of the increased relevant length scales of the energy of the external field, and because the relaxation times of large clusters of resistors *in the absence of a DC field* are strongly enhanced over those of the largest resistors in the cluster. As a consequence, the noise arising from large clusters of resistors is shifted to a higher frequency, enhancing the noise spectrum at that frequency [25]. An interesting consequence of this physical result is that the enhancement of the power spectrum at any frequency *by this means* is very closely related to the noise at that frequency in the absence of an external field. In some respects this result is desirable, in others it is not. Since scaling formulations of the flicker noise [12] demonstrate its intimate connection with the conductivity, itself related to the unstimulated noise by the fluctuation-dissipation theorem, a close relationship between the forms of the flicker noise and unstimulated noise must exist. However, it can be shown that using such an argument as a basis for a calculation scheme leads to a contribution to the flicker noise which extends only to frequencies incrementally below ω_c with the increment an increasing function of the external field [25]; such a restriction is not noted experimentally [26] and this mechanism is rejected.

Fluctuations in the energies of sites on the percolation path produce fluctuations in the resistance of the critical network. Such fluctuations (due to cluster polarization effects) will be shown here to occur at arbitrarily low frequencies for arbitrarily small fields, and are the ones of greatest interest. This type of mechanism has been suggested [13, 26] to be the most likely to produce the observed $1/f$ noise; *what is new here is the explicit means to calculate the magnitudes of the fluctuations in terms of concrete cluster polarization effects*, as well as the resulting quality of agreement with experiment.

Fluctuations in the critical resistance, ΔR_c, lead to contributions to $\Delta J(\omega) \approx kT\Delta R_c$. Changes in individual site energies lead to changes in R_c through alteration of the f, while changes in energy gradients produce changes in R_c through an alteration of the w. For simplicity we look only at effects on the f. The changes in site energy are proportional to the square of the applied field because it is an interaction energy between two distinct induced charges, one on the DC cluster, the second on the backbone of a large, but finite cluster nearby.

Consider the Coulomb interaction between the induced charge, $\Delta\rho_2$, on the end of a chain of N critical resistors (and $N + 1$ associated capacitors) and an induced charge, $\Delta\rho_1$, on a site, j, on the critical path. I choose $R = R_c$ in such chains because, at critical percolation, clusters of all sizes exist, in contrast to off-critical resistors for which exponential functions cut off cluster distributions at large cluster sizes. In the presence of a DC field, F, the occupation of site j on the DC path may be altered by $\exp[e(eFl)/kT]$, where l is the distance between resistors, R_c. l is the same for the separation of critical resistors on a large cluster, and very nearly the same value for their bulk separation. The induced charge is given by the lowest order expansion of an exponential occupation factor both on the DC path and on large (unconnected) clusters, and for a site on the DC path is $\Delta\rho_1 \propto e(eFl)/kT$. Similarly, the additional charge at one end of the chain of resistors is $\Delta\rho_2 \propto [e(NeFl)/kT]NC$, where NC is the (parallel) capacitance of the chain, and $NeFl$ is the potential difference across the chain. Each of the $N + 1$ capacitors for T not too low, have (in variable-range hopping) $C = e^2/kT$, meaning that the charge generated per critical resistor is the ratio of the field energy, eFl to the thermal energy, kT, times the electronic charge, and is also representable as the product of the capacitance and the potential, $CV = (e^2/kT)Fl$. However [27] at very low T, Coulomb repulsion on a chain of resistances inhibits charge generation, and involves a ratio of a Coulomb energy per critical resistor to kT. Thus when the thermal energy available to an electron is no longer enough to offset the self-energy per electron, the self-energy limits the charge generation. The reason why the self-energy per electron is the relevant energy is that an average is performed over many possible hopping transitions, each of which involves a probability of electronic transfer which is much smaller than one. The resulting modification to the capacitance is

$$C = \frac{e^2}{kT + \frac{e^2}{\varepsilon l}} \tag{8}$$

where ε is the background dielectric constant of the medium. These induced charges produce an interaction energy as large as

$$\Delta E_{int} = \frac{\Delta\rho_1 \Delta\rho_2}{\varepsilon r} \tag{9}$$

where r is the minimum reasonable separation. A value of r smaller than l appears to be unreasonable, because l gives the separation of the critical resistance values, so I choose l. $\Delta R = R_c \exp(\Delta E/kT) - R_c$ is now,

$$\Delta R = \left[\frac{eFl}{kT}\right]^2 N^2 \frac{e^2}{\varepsilon l \left(kT + \frac{e^2}{\varepsilon l}\right)} R_c. \tag{10}$$

In the low temperature limit, $kT < \frac{e^2}{\varepsilon l}$, this expression yields

$$\Delta J(\omega) = kT R_c \left[\frac{eFl}{kT}\right]^2 N^2. \tag{11}$$

But the condition on the largest N allowed is frequency dependent since $N^2 R_c C = \tau$ cannot exceed $1/\omega$, allowing the substitution $N^2 \to \omega_c/\omega$, and

$$\Delta J(\omega) = kT R_c \left[\frac{eFl}{kT}\right]^2 \frac{\omega_c}{\omega}. \tag{12}$$

Since the cross-sectional area taken up by a chain of N resistors of separation l is l^2, the number of such clusters available for influencing the DC current on any given segment of the DC cluster must be proportional to $[\frac{a}{l}]^2$ (with a the localization length)) and $\Delta J(\omega) \to \Delta J(\omega)[\frac{a}{l}]^2$.

The scaling form of (12) is identical to that in the text between (6) and (7) except for the additional factor $(eFl)^2/(kT)^2$, the ratio of the square of the field energy per critical resistor to the square of the thermal energy. Although various statistical effects have been ignored in this discussion, and only the largest reasonable effect has been calculated, result (12) will turn out to be in accord with experiment in crystalline germanium [3], and its appearance in this type of scaled form involving the critical resistance and critical frequency suggests its relevance to general formulations of the flicker noise in hopping conduction systems.

4. Conclusions

The following results have been obtained:

(i) in the absence of an external field $J(\omega_c) \propto R_c kT$ in all systems, allowing approximate scaling formulations of $J(\omega)/J(\omega_c)$, where $J(\omega_c) \propto 1/\sigma(\omega_c) \propto 1/\sigma_{DC}$,

(ii) an additive enhancement of $J(\omega)$ (for sufficiently low T, a condition dependent on relevant Coulomb interaction energies compared with kT) which is proportional to ω^{-1} and (to lowest order) to the square of the applied field, i.e.

$$\Delta J(\omega, F) \propto \frac{\omega_c}{\omega} \frac{R_c kT}{(l/a)^2} \left[\frac{eFl}{kT}\right]^2 \tag{13}$$

(iii)

$$J(\omega_c, F) \propto \frac{R_c e^2 F^2 a^2}{kT} = \frac{F^2 a^2}{\omega_c}$$

(the latter equality does not follow in spatially random systems, i.e. nearest neighbour hopping).

The specific result (iii) in the case of VRH (variable-range hopping) is compared with recent experimental observations [3] of the noise spectrum in *crystalline* germanium, although some uncertainty in the interpretation arises. While conduction in crystalline germanium is assumed to proceed by electronic 'hopping' through impurity states at temperatures low enough to exclude band hopping, it is usually assumed [28] that the mechanism for the DC conduction is by 'nearest neighbour' hopping [20] (SR systems) at the highest temperatures (for which band hopping is negligible), variable-range [28] hopping at lower temperatures, and variable-range hopping in a Coulomb gap [29] at still lower temperatures. Without specific data for the conductivity, it is not possible to state conclusively whether the variable-range hopping model is appropriate. Nevertheless, the results were interpreted by Shlimak *et al* [3] in terms of the hopping model calculations of Shklovskii [13] and coworkers. The result at low frequencies from (41) is (ignoring numerical constants),

$$\Delta J(\omega, F) = \frac{F^2 a}{\omega} = \frac{F^2 a^2}{\omega a}. \tag{14}$$

The noise spectrum was reported by Shlimak *et al* [3] to be proportional to the square of the electric field, F, the inverse of the frequency, ω, and to be temperature independent, in agreement with the result here. Since Shlimak *et al* [3] demonstrate a cross-over from a strong temperature dependence (at higher T) to the temperature-independence (at lower T), this may represent a cross-over from nearest-neighbour to variable-range hopping; in such a case a cross-over from an exponential temperature dependence to a result independent of T appears to duplicate their results. (As pointed out in section 3, the exponential T dependence in SR systems arises from the fact that R_c is exponentially T dependent, but $\omega_c = [R_c]^{-1}$ is not, in contradistinction to VRH.)

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