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Hopping conduction in some fractal systems

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Abstract. The DC and AC hopping conduction in some fractal systems are studied. The DC conductivity is analysed in some partial fractal systems where both localized and superlocalized states exist. The AC conductivity $\sigma(\omega)$ is shown to scale as $\omega\left[\ln(\nu_{ph}/\omega)\right]^{(D+2-\zeta)/\zeta}$ and is proportional approximately to ω^s with $s = 0.88$ for three-dimensional percolation. The ambiguities in the analysis of conductivity data are discussed and it is pointed out that the measurement of some characteristic length scales is very important in analysing the hopping mechanism and the origin of localization from experimental results.

1. Introduction

The hopping conductivity of insulators at low temperatures can usually reflect the conducting mechanism and can in principle be used to determine the physical origin which is responsible for the insulating character of the material. Some disordered materials possess fractal structure over a certain range of length scales and the introduction of fractal concepts has been successful in the identification of many physical properties in topologically disordered systems. It is therefore of importance to study the hopping conduction in fractal systems.

Electronic states in fractals have wavefunctions decaying faster than exponentially (Lévy and Souillard 1987)

$$\psi(r) \sim \exp\left[-\left(\frac{r}{L}\right)^{\zeta}\right] \tag{1}$$

where L is the (Euclidean) localization length and $\zeta > 1$ is the superlocalization exponent. This superlocalization character of wavefunctions may have important consequences. Deutscher and co-workers (1987) have extended Mott's variable-range hopping idea to fractals and found that the DC conductivity is characterized by an exponent related to ζ and the fractal dimension D. This hopping mechanism in fractals was later discussed more carefully by Harris and Aharony (1987). Their results are applicable to systems with large correlation length where all the electronic states are superlocalized. There are materials where the correlation length is not very large so that both localized and superlocalized states exist. The hopping conduction in such systems is one problem that we would examine here. The hopping conductivity for alternating currents in fractals is studied for the first time in this paper. It is found that $\sigma(\omega)$ scales as $\omega\left[\ln(\nu_{ph}/\omega)\right]^{(D+2-\zeta)/\zeta}$ and is proportional approximately to ω^s with $s = 0.88$ for three-dimensional percolation. Possible dimensional crossover behaviour of hopping conduction in some partial fractal systems is elucidated.

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The ambiguities in the analysis of conductivity data are discussed and we point out that the measurement of some characteristic length scales is very important for a reliable analysis.

This paper is organized as follows. In section 2 we study the DC conductivity in systems with both localized and superlocalized electronic states. Section 3 is devoted to the study of the effect of superlocalization on the AC hopping conductivity and in section 4 we discuss the possible dimensional crossover of the hopping conduction in some partial fractal systems. Conclusions of this paper are given in section 5.

2. DC hopping conductivity in some partial fractal systems

The behaviour of the DC conductivity at low temperatures is usually of the type $\exp[-(T_0/T)^\beta]$ and is thus characterized by the exponent β. There have been several conducting mechanisms leading to different values of β.

(i) $\beta = 1$, which is for the thermally activated hopping of carriers at the mobility edges into extended states or carriers excited into localized states at the band edges.

(ii) $\beta = 1/(d+1)$, known as the Mott relation (Mott 1968, 1969), characterizes the variable-range hopping (VRH) of carriers between the electronic states at the Fermi level with Anderson (strong) localization. Here $d > 1$ is the space dimension.

(iii) $\beta = \zeta/(D+\zeta)$ (Deutscher *et al* 1987), where D is the fractal dimension and ζ is the superlocalization exponent, results from the VRH of carriers between electronic states with superlocalized wavefunctions in fractals.

If the Coulomb interaction between electrons is important, one has $\beta = \frac{1}{2}$ (Efros and Shklovskii 1975) for Euclidean systems and $\beta = \zeta/(\zeta+1)$ (van der Puttern *et al* 1992) in the fractal cases. For clarity we shall not address the problem of the Coulomb interaction effect here.

Fractal geometry is believed to be relevant within a certain range of length scales in real materials: $a < r < \xi$, where a is the size of the smallest unit and ξ is the correlation length. At length scales greater than ξ, the system can be regarded as homogeneous and the usual Euclidean scaling is resumed. It is thus important to compare the localization length L with the correlation length ξ. If $L < \xi$, the electron feels the self-similar character of the system and the wavefunction decays following (1) up to ξ and continues to fall off exponentially at larger distances. Otherwise the electrons do not feel the fractal nature of the system and the wavefunction decays exponentially. VRH in the fractal regime can be expected if the correlation length ξ is large enough so that all the electronic states are superlocalized. Another quantity, the optimal hopping distance r_h, is also important in determining the exponent β. If r_h is found to be much larger than ξ, the exponents D and ζ have to be replaced by d and unity, i.e., their values in the homogeneous regime (Deutscher *et al* 1987). Therefore the VRH in the fractal regime appears only if

$$a \ll L \ll r_h \ll \xi \tag{2}$$

as pointed out by Aharony *et al* (1993a).

There are materials in which electronic states at different sites possess different localization length. An illustrative example is the granular materials, where the localization length L is proportional to the granular size x. When the grain size is changed, L/x does not vary (Deutscher *et al* 1987). If the system is composed of grains with different size, the localization lengths of individual electronic states may differ from each other significantly. When the correlation length is not very large or small (which is often true for practical materials), one expects that the localized (with $L > \xi$) and superlocalized (with $L < \xi$)

electronic states coexist. The character of VRH in such systems may be different from that in the Euclidean case or pure fractal regime (where the correlation length scale ξ is large enough so that all the states are superlocalized).

Following the Mott argument, the hopping probability p is proportional to the product of two terms:

$$p \sim |\psi(r)|^2 \exp[-\Delta E(r)/k_B T]. \tag{3}$$

The first term is essentially the probability of finding the electron at distance r from its initial site (Zallen 1983), and the second one represents the Boltzmann factor for the phonon-mediated hopping process. It is obvious that the hopping probability is dependent upon the character of localization of the initial state. We assume that the localized and superlocalized electronic states are randomly distributed in energy as well as in space, with a uniform distribution given by $N(E_F)$, the density of states at energies close to the Fermi energy E_F, and restrict the following calculations to the range $r_h < \xi$, since the Mott relation is recovered for $r_h > \xi$. Then one has (Deutscher *et al* 1987)

$$\Delta E(r) = \frac{1}{a^{d-D} N(E_F) r^D} \tag{4}$$

where D is the fractal dimension.

In the optimization of equation (3), we encounter two different cases depending on whether the initial state is localized or superlocalized.

Case A. The initial state is localized with localization length $L_1 > \xi$. Then substituting $\psi(r) \sim \exp(-r/L_1)$ and equation (4) into equation (3), one can find the optimal hopping distance

$$r_A = \left[\frac{L_1 D}{2k_B T a^{d-D} N(E_F)}\right]^{1/(D+1)} \tag{5}$$

and the exponent

$$\beta = \frac{1}{D+1}. \tag{6}$$

Case B. $\psi(r) \sim \exp[-(r/L_2)^\zeta]$ with $\zeta > 1$ and $L_2 < \xi$. Then one has

$$r_B = \left[\frac{L_2^\zeta D}{2\zeta k_B T a^{d-D} N(E_F)}\right]^{1/(D+\zeta)} \tag{7}$$

and

$$\beta = \frac{\zeta}{D+\zeta}. \tag{8}$$

In order to see which process is more favourable, we discuss the variation of the quantity p_{Am}/p_{Bm} with temperature, where p_{Am} denotes the optimal hopping probability of case A, and p_{Bm} that of case B. It is related to r_A and r_B by

$$p_{Am}/p_{Bm} = \exp\left[-\frac{2}{L_1} r_A \left(1 + \frac{1}{D}\right) + \frac{2}{L_2^\zeta} r_B^\zeta \left(1 + \frac{\zeta}{D}\right)\right]. \tag{9}$$

Therefore p_{Am} equals p_{Bm} if $r_A = (L_1/L_2) r_B^\zeta (1 + \zeta/D)/(1 + 1/D)$, or

$$T = T_0 \equiv \frac{D}{2k_B a^{d-D} N(E_F)} \times \left[\left(\frac{1+\zeta/D}{1+1/D}\right)\left(\frac{1}{\zeta}\right)^{\zeta/(D+\zeta)} L_1^{D/(D+1)} \left(\frac{1}{L_2}\right)^{D\zeta/(D+\zeta)}\right]^{(D+\zeta)(D+1)/D(\zeta-1)} \tag{10}$$

and case A (which is characterized by the exponent $\beta = 1/(D+1)$) will be more favourable if $T \ll T_0$. On the contrary, case B (with $\beta = \zeta/(D+\zeta)$) becomes the dominant process when $T \gg T_0$. In the intermediate-temperature region, one expects a complicated crossover behaviour. This kind of crossover may bring about difficulties in analysing the experimental data, as will be discussed in section 4.

3. Frequency dependence of the hopping conductivity in fractals

The AC conductivity of a wide variety of materials varies as ω^s with s of the order of 0.8 (see Mott and Davis 1979). The two-site or pair approximation has been shown (Pollak and Geballe 1961) to give such a power law frequency dependence in a large frequency range. We shall now examine the implication of fractals on the frequency dependence of the hopping conductivity. The study here follows the development of Mott and Davis (1979).

Adapted to our case, the analysis of Mott and Davis is as follows.

Rewriting equation (3), the mean jump time between two sites of distance R apart is

$$\frac{1}{\tau} = \nu_{ph} \exp\left[-2\left(\frac{r}{L}\right)^{\zeta}\right] \exp\left(-\frac{\Delta W}{k_B T}\right) \tag{11}$$

where ν_{ph} is the frequency of the phonon involved in the hopping process.

The number of electrons participating in the hopping is $N(E_F)k_BT$ per unit volume. Supposing that only hops of energy $\sim k_BT$ make an important contribution, then the last factor in equation (11) is of order unity. The number of vacant states into which an electron can jump is then $N(E_F)k_BT$. The important hops have $\omega\tau \sim 1$, that is for which the hopping distance is R, where

$$R = L\left[\frac{1}{2}\ln\left(\frac{\nu_{ph}}{\omega}\right)\right]^{1/\zeta} \tag{12}$$

and a range of ΔR is taken to give a significant contribution where

$$\Delta R = \frac{1}{2\zeta}\frac{L^{\zeta}}{R^{\zeta-1}}. \tag{13}$$

The hopping distance R here must satisfy $R < \xi$, otherwise the hop does not feel the existence of fractal geometry and the usual Euclidean scaling is recovered. Considering that the number of available empty states is proportional to $R^{D-1}\,\Delta R$, we have

$$\sigma(\omega) \sim k_BT\omega R^2 R^{D-1}\,\Delta R$$

or, replacing R and ΔR with the expressions given in equations (12) and (13),

$$\sigma(\omega) \sim k_BT\frac{1}{\zeta}\left(\frac{L}{2^{\zeta}}\right)^{D+2}\omega\left[\ln\left(\frac{\nu_{ph}}{\omega}\right)\right]^{(D+2-\zeta)/\zeta}. \tag{14}$$

One can see that ζ and D enter the exponent again, as in the DC conductivity. If we set $D = d$ and $\zeta = 1$, equation (14) then resumes the Euclidean scaling, i.e., $\sigma(\omega) \sim \omega\left[\ln\left(\nu_{ph}/\omega\right)\right]^{d+1}$.

The frequency dependence can usually be written as $\sigma(\omega) \propto \omega^s$, where s is a weak function of frequency if $\omega \ll \nu_{ph}$, which means that $\ln\sigma(\omega)$ versus $\ln\omega$ should scale approximately linearly with slope s given by

$$s = \mathrm{d}\ln\sigma(\omega)/\mathrm{d}(\ln\omega) = 1 - \frac{1}{\zeta}(D+2-\zeta)\Big/\ln\left(\frac{\nu_{ph}}{\omega}\right). \tag{15}$$

A plot of $\log\sigma(\omega)$ versus $\log\omega$ for various values of ν_{ph} is given in figure 1. The curve of s against $\log(\nu_{ph}/\omega)$ is shown in figure 2. The data used in the calculation are those of 3D percolation: $\zeta = 1.36$ (Aharony *et al* 1993a) and $D = 2.5$. One can see that if ν_{ph}/ω is of the order of 10^8, s is about 0.88, a value close to 0.8, which is often observed (Mott and Davis 1979). The fractal dimension dependence of the exponent s, on the other hand, is not easy to determine from equation (15) since ζ is usually relevant to D. However, under the argument $\zeta = d_{min}$ (Aharony *et al* 1993a) where d_{min} describes the scaling of the average minimal path along the bonds of the fractal, ζ does not change significantly when D changes. Therefore one can take ζ as a constant approximately and see that s changes linearly with the fractal dimension D.

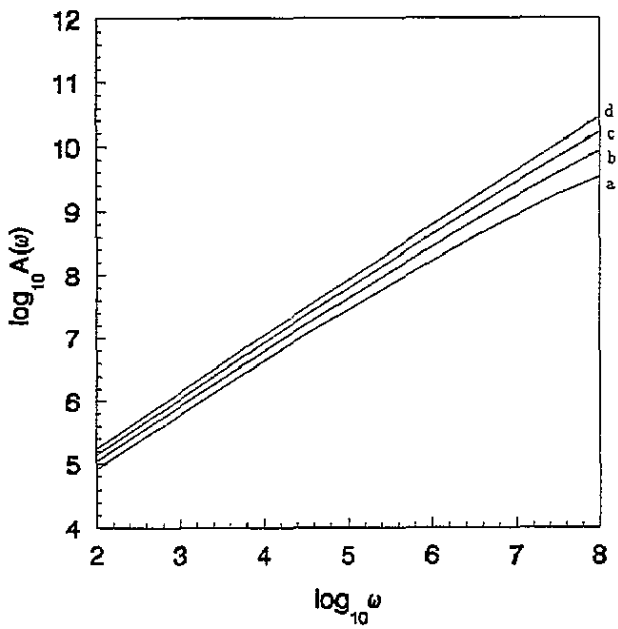

Figure 1. A plot of $\log A(\omega)$ against $\log\omega$ for various values of ν_{ph} where $A(\omega) = \omega[\ln(\nu_{ph}/\omega)]^{(D+2-\zeta)/\zeta}$. Curves a, b, c and d correspond to $\nu_{ph} = 10^{10}$, 10^{11}, 10^{12} and 10^{13} Hz respectively, and the 3D percolation values for D and ζ are used.

In the above analysis, it has been supposed that the electron goes through the barrier between the two sites, instead of over it. It should also be noted that equation (14) is deduced subject to two assumptions (Mott and Davis 1979): (1) $k_BT \ll E_F$ and (2) the resonance energy of centres of distance R apart is less than k_BT.

Equation (14) characterizes the hopping of carriers between superlocalized states. If there are both localized and superlocalized states in one material, one expects $\sigma(\omega) \sim \omega[\ln(\nu_{ph}/\omega)]^{D+1}$ for the hopping from localized states in the fractal regime, and $\omega[\ln(\nu_{ph}\omega)]^{(D+2-\zeta)/\zeta}$ for that from superlocalized states. However, the effective exponent s is not expected to vary significantly for these two cases. It is therefore not easy to see whether the hopping occurs for the localized or superlocalized states for a specific material from experimentally observed $\sigma(\omega) \sim \omega^s$ results.

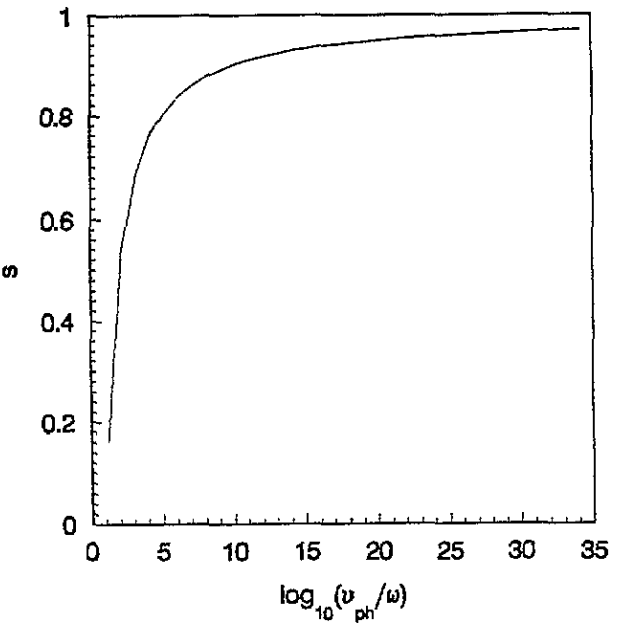

Figure 2. The exponent s of 3D percolation as a function of ν_{ph}/ω. One can see that s is about 0.88 for $\nu_{ph}/\omega = 10^8$.

4. Discussion

In real materials with fractal structure, there is always a correlation length ξ which separates the fractal regime from the Euclidean regime. If the characteristic length of a certain physical quantity changes with external conditions—such as temperature, pressure—to cross ξ, the crossover from the bulk Euclidean to fractal geometry is reflected in the change of the quantity. This dimensional crossover has several implications. The phonon (magnon)–fracton crossover which was first discussed by Alexander and Orbach (1982) may be an important one. Gordon *et al* (1987) have also found that the superconducting transition of a three-order Sierpinski wire network exhibits this crossover. This kind of dimensional crossover is essentially identical to the 3D to 2D crossovers observed in many high-T_c cuprates (see, for example, Gao *et al* 1993) and some layered magnets; the difference is that in those cases the crossovers occur between integer dimensions. Since the hopping distance of DC conduction is dependent on the temperature, and that of AC conduction is sensitive to the frequency ω, it is very likely that the dimensional crossover could occur for the conductivity from Euclidean scaling to fractal scaling.

These crossovers indeed make it very difficult to discuss the hopping mechanism and the origin of the localization from experimentally observed conductivity data. People used to fit the experimental results with one theory and extract some exponents. Unfortunately, this kind of fit is often model dependent. It is possible to obtain puzzling different exponents based upon different theories. Aharony *et al* (1993a, b) have shown that by allowing for a pre-exponential temperature-dependent factor, the data taken on carbon black polymer composites (van der Puttern *et al* 1992) may support percolation as well as non-percolation exponents. Besides, the exponents of different mechanisms usually do not vary with each other significantly, which makes it dangerous to fit experiments with a single power law over a wide temperature range to find an effective value of β. Considering these, we

feel that some characteristic length scales should be determined from auxiliary experiments before a reliable fit. These length scales include the correlation length ξ (below which the system can be regarded as fractal), the localization length L (the state is localized if $L > \xi$ and superlocalized if $L < \xi$) and the hopping distance r_h (the hop feels the existence of the self-similar feature if $r_h < \xi$). Among the three lengths, the correlation length ξ can be measured by recording light scattering or neutron spectroscopy (Courtens *et al* 1989). The localization length can be evaluated through a magnetoresistance experiment (Sin *et al* 1984) since the value of the external magnetic field necessary to eliminate the strong localization effects is a function of the localization length. The conducting mechanism will follow if one can manage to measure the hopping distance r_h directly. The length is, however, not easy to determine to the present author's knowledge. It has to be obtained together with the hopping mechanism from experiments self-consistently by a careful fit. However, the determination of L and ξ can be very helpful for one knows immediately the character of localization from them and whether a hopping mechanism is possible or not. The importance of comparing the length scales has been noticed by Deutscher *et al* (1987). However, they did not emphasize the significance of carrying out the comparison by experiment in determining the hopping mechanism and further analysing the origin of localization.

The DC conductivity results of carbon black polymer composites have been controversially discussed (Aharony *et al* 1993b, a, Michels *et al* 1993, van der Puttern *et al* 1992). Michels and co-workers (van der Puttern *et al* 1992, Michels *et al* 1993) analysed the data in a generalized VRH picture and argued that their data can be regarded as evidence for VRH between superlocalized states on a fractal by assuming Coulomb-dominated hopping, while Aharony *et al* 1993a, b felt that the exponent depends strongly on the analysis, and therefore confirms no theory. They further discussed the importance of including a pre-exponentially temperature-dependent factor in analysing the data. Although the prefactor is important in a quantitative fit to experiments, we think that in this situation it is somewhat more helpful to determine the characteristic length scales. If ξ and L can be assessed from auxiliary experiments, it is easily clarified whether the hopping distances obtained by Michels *et al* are within the fractal regime (i.e., if $r_h \ll \xi$ holds) and this can at least be used to exclude some mechanisms.

It is noteworthy that to study the superlocalization phenomena experimentally, the DC conduction is more advantageous than the AC conduction since the DC conductivity is more sensitive to the superlocalization property. Polymers and porous materials could be interesting systems to study the superlocalization property. On the other hand, to compare with experiments more quantitatively, computer simulation studies are necessary. Recently, Ortuño and Ruiz (1992) have performed Monte Carlo simulation of the conductivity of a two-dimensional localized interaction system. Their results revealed clearly two distinct regimes, corresponding to nearest-neighbour and variable-range hopping and they estimated the transition temperature from the observed crossover. Similar work on hopping conduction in fractals has not been seen and research in this direction will be worthwhile.

5. Conclusions

We have studied the DC and AC conductivity in some fractal systems. In partial fractal substances where both localized and superlocalized electronic states exist, the DC hopping conductivity may cross over from one scaling to another with change of temperature. The frequency dependence of the hopping conductivity in fractals is given for the first time.

It is shown that $\sigma(\omega)$ scales as $\omega\left[\ln(\nu_{ph}/\omega)\right]^{(D+2-\zeta)/\zeta}$ and is proportional approximately to ω^s with $s = 0.88$ for the three-dimensional percolation. We demonstrate the possible dimensional crossover of hopping conduction in some partial fractal systems. It is pointed out that the measurement of some characteristic length scales is important in determining the hopping mechanism and further analysing the origin from experimental results.

In this paper we have made the assumption of a smooth density of states near the Fermi level. This and some other simplifications might be not very realistic for a fractal. Therefore the discussions presented in this paper can be further improved if one knows the exact distribution of the density of states near the Fermi level and some other information for real fractal materials. These effects deserve further research.

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