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CHARGE TRANSPORT IN FLUCTUATING SYSTEMS

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ABSTRACT

Mobility of charge carriers in certain liquid systems is controlled by temporal fluctuations in local conductivity. Fast transport proceeds along high mobility regions similarly to traditional percolation with the difference that these regions form and fade away with fluctuations. By making use of the idea of waiting time distribution of continuous time random walk, formulae for relative mobility as a function of the expectation value of proportion of high mobility regions are suggested. The results compare reasonably well with experimental data. Under the experimental conditions given quasi-percolation theory and the effective medium theory of traditional percolation do not differ too much numerically.

For site percolation threshold in non-fluctuating systems the expression $[(e-1)/ez]^{1/2}$ is suggested where e is the base of natural logarithm and z is the coordination number.

АННОТАЦИЯ

Подвижность носителей заряда в некоторых жидких системах контролируется временными флуктуациями местной проводимости. Быстрый транспорт протекает по областям высокой подвижности, как и в обычной перколяции, разница состоит только в том, что эти области образуются и распадают в итоге флуктуаций. Используя идею распределения времени ожидания из теории "continuous time random walk" /непрерывного блуждания/ предложены формулы для описания относительной подвижности в зависимости от ожидаемого значения доли областей высокой подвижности. Результаты расчета удовлетворительно совпадают с экспериментальными данными. В данных экспериментальных условиях цифровые величины квази-перколяции и теории эффективной среды обычной перколяции не отличаются значительно.

Предлагается выражение для порогового значения перколяции в нефлуктуирующих системах: $[(e-1)/ez]^{1/2}$, где e - основа натурального логарифма, а z - координационное число.

KIVONAT

A töltéshordozók mozgékonyságát egyes folyékony rendszerekben a lokális vezetőképességben fellépő időbeli fluktuációk szabják meg. Gyors transzport csak nagy mozgékonyságot megengedő tartományok mentén játszódhatik le, hasonlatosan a hagyományos perkolációhoz, azzal a különbséggel, hogy ezek a tartományok a rendszer fluktuációinak hatására állandóan keletkeznek és eltünnek. Felhasználva a "continuous time random walk" /folytonos idejü bolyongás/ elméletének egy fogalmát, a várakozási idő eloszlását, meghatároztuk, hogy hogyan függ a relativ mozgékonyság a nagy mozgékonyságu tartományok részarányának várható értékétől. A számitott eredmények meglehetősen jól egyeznek a kisérleti adatokkal. Az alkalmazott kisérleti körülmények között kvázi-perkoláció és a hagyományos perkoláció effektiv közeg elmélete numerikusan csak kevéssé különböznek egymástól.

Nem fluktuáló rendszerekben végbemenő hely- perkoláció küszöbértékére az $[(e-1)/ez]^{1/2}$ kifejezést javasoljuk, ahol e a természetes logaritmus alapszáma, z pedig a koordinációs szám.

INTRODUCTION

Percolation theory has been developed for the description of transport processes in spatially disordered systems like random networks, amorphous solids or composite materials /for reviews see Refs. 1-5/. In order to visualize the basic problem let us imagine a network of ohmic resistors with randomly distributed missing elements or a random mixture of conducting and isulating balls. Current can flow across such a system only if the proportion of conducting elements is high enough to form at least one contiguous channel along which the charge carriers can pervade the entire /infinitely large/ system. The most obvious common feature of all such models is the existence of a non-zero lower limit of the proportion of conducting elements below which no current can flow. This limit is called the percolation threshold.

Percolation theory deals with spatial fluctuations only, all the properties and parameters being regarded as independent of time. Hence it was an important intuitive step forward that was made by Kestner and Jortner[6] who applied one of the description of percolation, effective medium theory[7], to charge transport in hydrocarbon liquids. Here, conducting and insulating regions were thought to form due to thermodynamic fluctuations, this meaning that a conducting region can turn into an unsulating one and vice versa. Thus local conductivity changes with time at any given site.

Although having proposed an alternative description of charge mobility in liquid hydrocarbons[8] we made use of these ideas in the understanding of electron and hole mobility in certain liquid mixtures[9]. In these mixtures the charge carriers are in a high-mobility state only if they are surrounded exclusively by the molecules of one of the components or, in brief, if they are in a pure subsystem. Examples of such mixtures will be given in a later section.

Whatever the chemical nature of the mixture, pure conducting subsystems are brought about by temporal fluctuations of concentration. While our attempt to describe mobility as a function of concentration by making use of effective medium theory seemed to be successful for a number of mixtures[9] the conceptual problem of how to reconcile percolation with temporal fluctuations remained, though tacitly, unresolved.

The aim of the present paper is to give a simple description of charge carrier motion governed by temporal fluctuations in conductivity. To this end some notions of continuous time random walk theory[10-12] will be made use of. Whereas classical random walk is characterized by a constant waiting time between two subsequent jumps this theory applies a continuous distribution of waiting times the function $\Psi(t)dt$ being the probability of a jump taking place between t and t+dt.

A two-state model will be adopted, i.e. a fluctuating subsystem will be thought to be either in the conducting or in the insulating state. The results will be compared both with experimental data and with effective medium theory. By developing an analogy between the present treatment and percolation in non-fluctuating systems an expression for percolation threshold will be proposed.

MOBILITY IN FLUCTUATING SYSTEMS

Let the macroscopic system be divided into subsystems. Perennial fluctuations change their properties in such a manner that they are either in the conducting or in the insulating state. A charge carrier can progress if both the subsystem in which it resides and one of its neighbors are conducting. If either or both of the subsystems in question are insulating the carrier is trapped. The existence of more than two adjacent subsystems is disregarded.

Let the average waiting time in a conducting subsystem be denoted by τ_o. If the system consists exclusively of conducting subsystems and the diffusivity has no dispersion the waiting time distribution is exponential,

$$\Psi_o(t) = \tau_o^{-1} \exp(-t/\tau_o). \qquad (1)$$

In this case Einstein's expression holds for any frequency[10],

$$D_o = L^2/2\tau_o, \qquad (2)$$

where L is the mean displacement for a single jump.

On the average a charge carrier, moving in a system of fluctuating conductivity, is imagined to make an attempt to leave the subsystem in which it resides at each τ_o instant. Let W denote the probability of the first attempt to be successful. The probability of a jump taking place between t and $t+\tau_o$ is given by

$$\Psi(t) = (1-W)^{t/\tau_o} W. \qquad (3)$$

The quotient t/τ_o equals the average number of unseccessful attempts /cf. Chandrasekhar[13]/. The series $\Psi(t)$ can be replaced by the continuous function

$$\Psi(t) = \tau^{-1} \exp(-t/\tau), \tag{4}$$

where τ is defined as

$$\tau = -\tau_o/\ln(1-W). \tag{5}$$

Here $\Psi(t)$ is a waiting time distribution function which, if integrated between t and $t+\tau_o$, renders $\Psi(t)$ as given by Eq.(3).

The integration of $\Psi(t)$ between O and τ_o should yield the probability of the first jump being successful. Integrating $\Psi(t)$ one indeed finds

$$\int_0^{\tau_o} \Psi(t)\, dt = W, \tag{6}$$

and

$$\int_0^{\tau_o} \Psi_o(t)dt = (e-1)/e = \eta \cong 0.6321. \tag{7}$$

Thus, η is the probability of a jump taking place between O and τ_o in a system which consists of conducting subsystems only.

In view of $\Psi(t)$ being a simple exponential the diffusion constant in a fluctuating system is given by Einstein's expression for any frequency,

$$D = L^2/2\tau. \tag{8}$$

The task is now to determine W through the properties of the system. A limiting value can immediately be established. Let the expectation value of volume fraction of conducting subsystems be denoted by C /for the sake of brevity C will be called conducting concentration from here on./ The relationship

$$W(C = 1) = \eta \tag{9}$$

must hold in view of Eqs.(6) and (7) whatever the functional form of W(C).

Two different cases will be discussed: /a/ Unlimited fluctuation: the presence of a charge carrier does not influence the fluctuation of the subsystem by which it is withheld. This is the case when interaction between charge carrier and subsystem is weak. /b/ Limited fluctuation: a subsystem which has obtained a carrier by having become conducting cannot turn again into the unsulating state. This happens if the interaction between charge carrier and environment is strong enough to stabilize the conducting state.

UNLIMITED FLUCTUATION

The charge carrier on the average investigates the state of the subsystem at intervals τ_o. Let W_{cc} and W_{ii} denote the respective probabilities of finding a conducting or an insulating subsystem in the same state by two subsequent investigations whereas W_{ci} and W_{ic} denote the respective probabilities of a conducting subsystem to be found insulating or an insulating to be found conducting from one attempt to the next.

The charge carrier which landed in a subsystem at moment t can move on at the first attempt if both the subsystem in which it resides and its neighbour are conducting at $t+\tau_o$. With unlimited fluctuation present this can happen in four different ways: /i/ both subsystems are conducting at t and $t+\tau_o$; /ii/ the first one is conducting at t and $t+\tau_o$ while the second one is insulating at t and conducting at $t+\tau_o$; /iii/ the first one is insulating at t and conducting at $t+\tau_o$ while the second one is conducting at t and $t+\tau_o$; /IV/ both are insulating at t and conducting at $t+\tau_o$.

Hence W can be written as

$$W = W_{uf} = \eta[C^2W_{cc}^2 + 2C(1-C)W_{ic}W_{cc} + (1-C)^2W_{ic}^2], \tag{10}$$

where the subscript uf refers to the mode of fluctuation. This expression also complies with Eq.(9).

Assuming an equilibrium to prevail the formation and disappearance probability of a certain state must be equal, thus the equation

$$CW_{ci} = (1-C)W_{ic} \tag{11}$$

must hold. Since only two states are available a simple relationship prevails:

$$W_{ci} + W_{cc} = W_{ic} + W_{ii} = 1. \tag{12}$$

/For Eqs.(11) and (12) cf. Ref. 13./

By combining Eqs.(10-12) one finds

$$W = \eta C^2. \tag{13}$$

Inserting Eq.(13) into Eq.(5) one obtains the concentration dependent time-constant of the waiting time distribution function as

$$\tau_{uf} = -\tau_o/\ln(1-\eta C^2). \tag{14}$$

and Eqs.(2), (6) and (14) yield

$$D_{uf}/D_o = \mu_{uf}/\mu_o = -\ln(1-\eta C^2). \tag{15}$$

Here μ_{uf} and μ_o denote charge carrier mobilities at conducting concentrations C and 1, respectively. The proportionality between D and μ is assumed to prevail. The function Eq.(15) is plotted in Fig. 1.

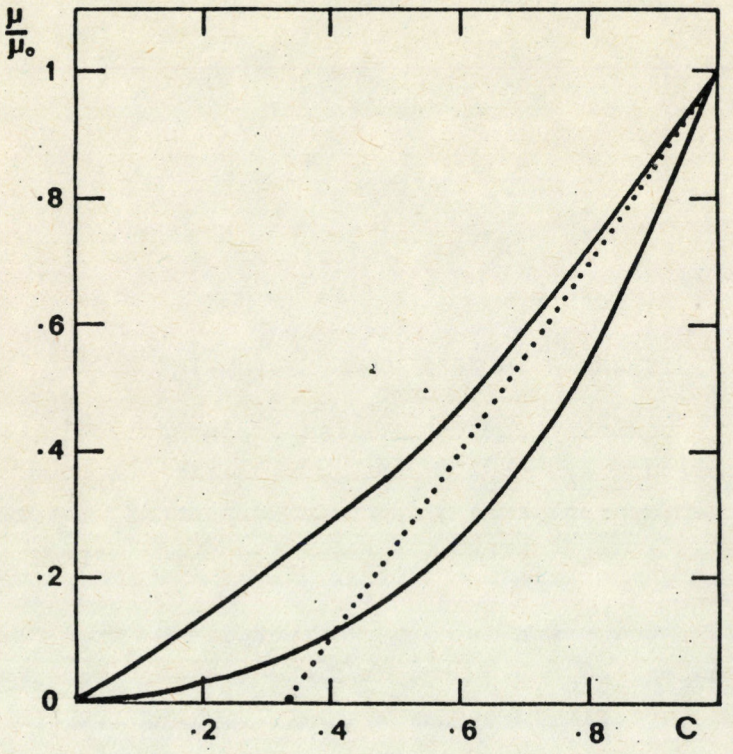

Fig. 1. Relative mobility as a function of C for unlimited /lower curve/ and limited /upper curve/ fluctuations, Eqs.(15) and (17), respectively. Dotted line: effective medium medium theory. For all cases, r = 0.

Until now it was assumed that $\mu_{uf} = 0$ if $C = 0$, that is, if the conducting concentration is zero no current can flow. If this is not the case and a non-zero mobility, μ_m, can be observed also across insulating subsystems, Eq.(15) must be modified to become

$$\mu_{uf}/\mu_o = -(1-r)\ln(1-\eta C^2) + r \qquad (15a)$$

where $r = \mu_m/\mu_o$.

LIMITED FLUCTUATION

The subsystem in which the charge carrier landed at moment t is in the conducting state with certainty at $t+\tau_o$. Thus W depends only on the probability of the neighboring subsystem being conducting at $t+\tau_o$. By recalling Eqs.(9), (11) and (12) this can be expressed as

$$W = W_{lf} = \eta[CW_{cc} + (1-C)W_{ic}] = \eta C, \qquad (16)$$

where the subscript lf refers to limited fluctuation. Combining Eqs.(2), (5), (8), and (16) one finds

$$D_{lf}/D_o = \mu_{lf}/\mu_o = -\ln(1-\eta C) \qquad (17)$$

if $\mu = 0$ at $C = 0$ and - similarly to Eq.(15a) - if $\mu_m \neq 0$

$$\mu_{lf}/\mu_o = -(1-r)\ln(1-\eta C) + r. \qquad (17a)$$

The graph of Eq.(17) is given in Fig. 1.

COMPARISON WITH EXPERIMENTS AND WITH EFFECTIVE MEDIUM THEORY

Charge migration in certain liquid mixtures depends greatly on concentration fluctuations. Three such systems, in which radiation-produced electrons or holes move faster than conventional ions, were discussed previously in terms of effective medium theory[9]. Now we re-examine these experimental results.

Similarly to Ref. 9, there are two ways to compute C from the mole fraction of the component enhancing charge motion, x. If the charge is localized and hence interacts with one single subsystem only, C is given as

$$C = C_s = x^n \qquad (18)$$

where n is the number of molecules with which the charge carrier is in direct interaction - this being taken for the size of the subsystem. If, however, the charge is delocalized and interacts with a large number of subsystems the entropy of mixing, ΔS_m, defines C by

$$C = C_r = x \exp(-\Delta S_m/k) = x \exp\{n[x\ln x + (1-x)\ln(1-x)]\} \qquad (19)$$

TRANS-DECALINE - CYCLO-HEXANE

A trans-decaline$^+$ positive ion can donate its charge to a neighboring trans-decaline but not to a cyclo-hexane molecule[14]. The positive charge interacts with one molecule only, hence n=1. The interaction energy being the ionization potential of the molecule the interaction between charge carrier and molecule must be regarded as strong thus the case of limited fluctuation, Eq.(17a), is expected to prevail. The charge carrier is localized which demands the use of Eq.(18). Experimental data and the quasi-percolation theory curve are given in Fig. 2. For comparison the effective medium theory curve[9] is also plotted. The agreement between experiment and both theories is reasonable.

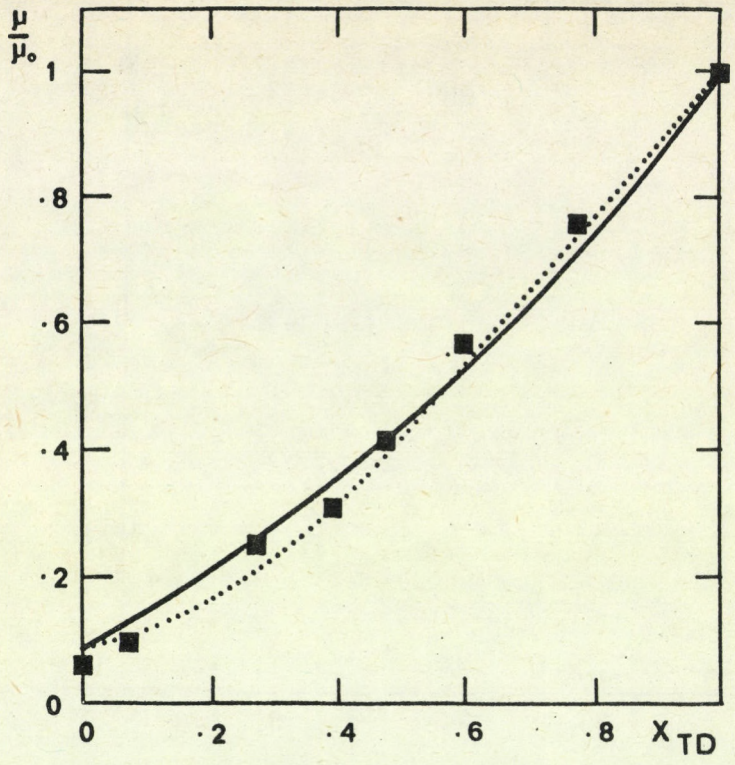

Fig. 2. Relative mobility in trans-decaline - cyclo-hexane mixtures[14] *as a function of the mole fraction of trans-decaline, x_{TD}. ——— present, using Eqs.(17a) and (18); ..... Ref. 9: both with n=1 and r=0.09.*

HEXAFLUOROBENZENE - BENZENE

Radiation-produced negative charge carriers in pure hexafluorobenzene have a mobility some 50 times higher than that of ordinary ions. The addition of an inert diluent, e.g. benzene, reduces the mobility drastically[15]. There are strong indications that the charge carrier is an electron delocalized over a number of C_6F_6 molecules[9]. Charge motion consists in the migration of an electron from a group of C_6F_6 molecules to a similar neighboring group. With a second, inert component present concentration fluctuations procure for the formation of groups consisiting exclusively of C_6F_6, i.e. of pure subsystems. The electron was shown to be bound to the group by an energy of some 3eV thus the energy per molecule is much higher than kT if the number of molecules in a group is not larger than 20. These facts compell one to use the limited fluctuation expression and the $C_s(x)$ function, Eqs. (17a) and (18).

The curves calculated by the present quasi-percolation method and by the effective medium theory are plotted in Fig. 3. together with experimental data. The agreement seems to be good it should be noted, however, that quasi--percolation was computed with n=15 whereas in the effective medium treatment n=12 was used. At present no experiment can tell which of the two figures is the more realistic one nor can we comment upon the reason of this disagreement.

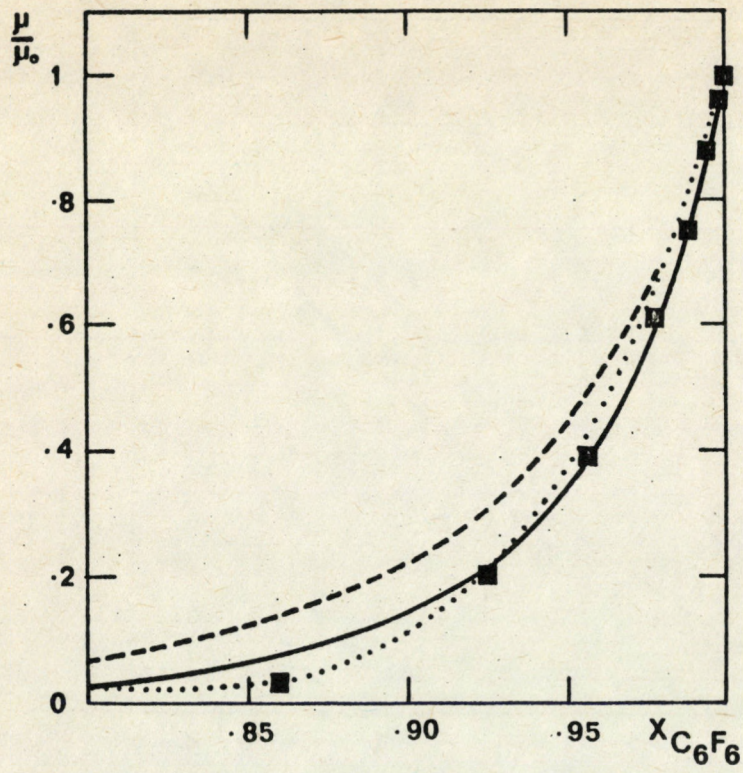

Fig. 3. Relative mobility in hexafluorobenzene - benzene mixtures[15] *as a function of the mole fraction of hexafluorobenzene, $x_{C_6F_6}$. ——— present, using Eqs.(17a) and (18) with n=16 and r=0.025; ---- present, with n=12 and r=0.025; ..... Ref. 9 with n=12 and r=0.025.*

N-HEXANE - ETHANOL

Electron mobility in n-hexane is by some two orders of magnitude higher than that in ethanol. Excess electrons in saturated liquid hydrocarbons are partially localized, i.e. a fraction P of them is localized, (1-P) is quasi--free. With mobilities for the localized and quasi-free states μ_L and μ_F, respectively, the experimental mobility is given as[8,16]

$$\mu_o = P\mu_L + (1-P)\mu_F. \qquad (20)$$

The presence of ethanol slows down electron motion because either of the two states can form in pure subsystems only. The lower limit of mobility in the mixtures equals that measured in pure ethanol and is denoted by μ_m.

In order to apply quasi-percolation to the present case one has to recall the physical differences between localized and quasi-free states. The interaction between a localized electron and the environment is strong and is limited to one molecule only[8] hence formulae (17a) and (18) with $n=n_L=1$ refer to this state. A quasi-free electron, however, being delocalized interacts with a large number of molecules and the interaction is weak hence Eqs.(15a) and

(19) must be used with $n=n_F$ which is to be determined by parameter fitting. The concentration dependent mobility in the mixture is of the form

$$\mu/\mu_o = -P(r_L-r)\ln(1-\eta C_s) - (1-P)(r_F-r)\ln(1-\eta C_r^2) + r, \qquad (21)$$

where

$$r = \mu_m/\mu_o;\ r_L = \mu_L/\mu_o \text{ and } r_F = \mu_F/\mu_o.$$

The curves calculated by Eq.(21) and by the effective medium theory[9] together with experimental points[17] are given in Fig. 4. The agreement between both theories and experiment seems to be acceptable.

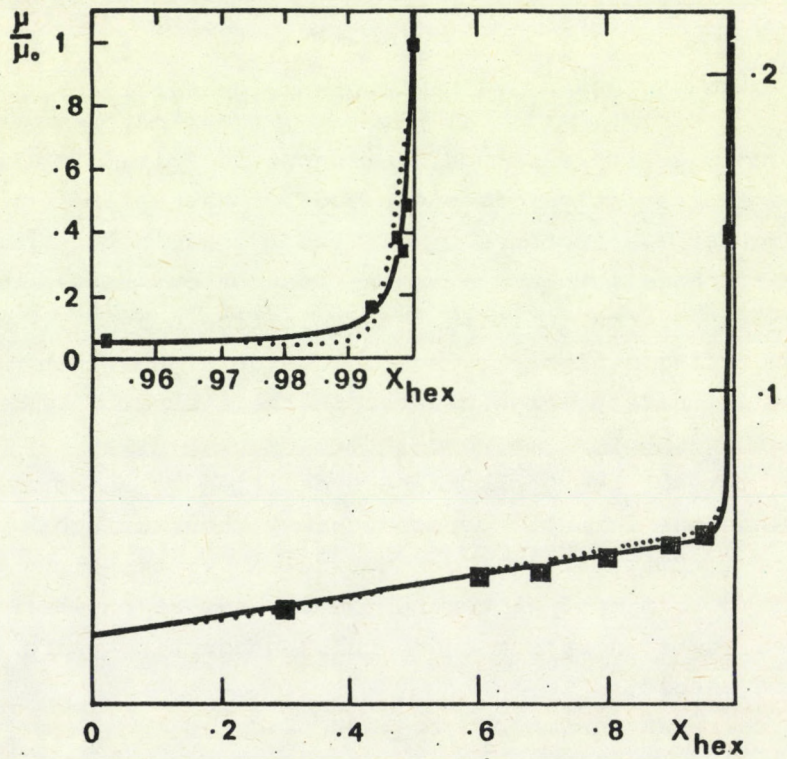

Fig. 4. Relative mobility in n-hexane - ethanol mixtures[17] as a function of the mole fraction of n-hexane, x_{hex}. —— present, using Eq.(21); ..... Ref. 9: both with $n_L=1$, $n_F=25$, $r=0.022$, $r_L=0.056$, $r_F=290.3$ and $P=0.9967$ /cf. Ref. 16./.

CONCLUDING REMARKS

A theoretical description was developed and a word coined for charge transport through media in which local conductivity fluctuates with time. Quasi-percolation differs from traditional percolation in the non-existence of any percolation threshold. This marked difference, however, is blurred if charge carriers have a finite mobility even in insulating subsystems. In that case effective medium theory and quasi-percolation coincide reasonably when using the same parameters and both of them describe the experiments adequately. This shows how good the intuitive idea of Kestner and Jortner[6] was.

Mobility as calculated by the above treatment does not have any dispersion. This is so because fluctuations were regarded to be extremely short lived, a subsystem was thought to change its state many a times during the passage of a charge carrier. This enabled us to use equilibrium assumptions on fluctuations which, in turn, made W independent of time. It must be remembered, however, that this simplification is due to an approximation which might break down at mobilities much higher or fluctuations much slower than those which prevail in the systems treated now.

A more general treatment must involve time-dependent fluctuations and must make W depend on time. Such a calculation might predict a frequency dependence of mobility, an effect not yet observed.

APPENDIX. PERCOLATION THRESHOLDS IN NON-FLUCTUATING SYSTEMS

In this Appendix a simple method, analogous in spirit to the foregoing discussion of quasi-percolation, is suggested for the estimation of percolation thresholds in space structures of different coordination numbers.

It is generally held[1], though - to the best of our knowledge - it has been strictly proven for the infinite square lattice only[18], that if the proportion of conducting subsystems is equal to or higher than the threshold there is only one infinitely large cluster in the system. It is as if a backbone of conducting subsystems were formed.

Let the structure of the backbone be simplified as an array of conducting subsystems which has the form of a space curve without branching or loops. The probability of finding pairs of conducting subsystems along any such space curve can be determined. We regard the concentration where this probability diverges, i.e. where an epidemic growth of conducting pairs sets in, as the percolation threshold.

The idea is somewhat resemblant to that of Sykes and Essam[19] who defined threshold by the concentration where functions of the mean number of clusters exhibit singularities. The reduction of the backbone to a simple space curve and the definition of the threshold in terms of conducting pairs along a space curve are rather arbitrary approximations the validity of which we have failed <u>a priori</u> to justify.

Let C now denote the volume fraction of conducting subsystems in a rigid system and z the coordination number. The probability of finding two adjacent conducting subsystems, P', can be given as

$$P' = C[1-(1-C)^z] = zC^2 + \text{higher order terms.} \qquad (A1)$$

The exclusion of branching means that the possibility of more than two adjacent conducting subsystems is excluded. We try to express this limitation by disregarding higher order terms in Eq.(A1), i.e. we write the probability as $P = zC^2$.

Let us consider a contiguous array of subsystems and evaluate the probability, $\Psi(k)$, that no conducting pair can be found from the first through the k-th subsystem and the (k+1)-th is conducting and has a conducting neighbor. Similarly to formulae of quasi-percolation, $\Psi(k)$ can be expressed as

$$\Psi(k) = (1-P)^k P = (1-zC^2)^k zC. \tag{A2}$$

This can be replaced by the continuous function $\Psi(x)$ as

$$\Psi(x) = \kappa^{-1}\exp(-x/\kappa), \tag{A3}$$

where x is a length measured along the array, $[x]/L = 2k$, L is the linear dimension of a subsystem and κ is defined as

$$\kappa = -2L/\ln(1-zC^2). \tag{A4}$$

The integration of $\Psi(x)$ between 2kL and 2(k+1)L results in $\Psi(k)$ in complete analogy with Eqs.(3) and (4).

The expectation value of the length between two conduction pairs can be evaluated as

$$\langle x \rangle = \int_0^\infty x\,\Psi(x)\,dx = \kappa \tag{A5}$$

Now we express the percolation threshold in terms of κ. Finite subsystems and the limiting case $L \to 0$ are treated separately.

Subsystems of finite dimensions. The length of a pair of subsystems is 2L. If $\kappa > 2L$ the array is insulating since this inequality means that conducting pairs are held apart by insulating subsystems. The array becomes conductive, when the relationship $\kappa = 2L$ holds, i.e. when all the conducting pairs coalesce. This defines the percolation threshold, C_c, in view of Eq.(A4) as

$$-\ln(1-zC_c^2) = 1. \tag{A6}$$

Infinitely small subsystems. The number of pairs of subsystems per unit length is $(2L)^{-1}$ whereas the number of conductive pairs is κ^{-1}. Hence the probability of a pair being conductive is $2L/\kappa$. The probability of all the pairs of subsystems per unit length being conductive is $(2L/\kappa)^{1/2L}$. Let the limit $L \to 0$ be investigated by considering also Eq.(A4),

$$\lim_{L\to 0} [-\ln(1-zC^2)]^{1/2L} = \begin{cases} 0 \text{ if } -\ln(1-zC^2) < 1 & \text{(A7)} \\ 1 \text{ if } -\ln(1-zC^2) = 1. & \text{(A8)} \end{cases}$$

There is a sudden change in the probability of an infinite conducting array to exist. It is zero until C is as low as for Eq.(A7) to be valid and becomes abruptly equal to 1 as C attains the value set by Eq.(A8). The concentration

defined by Eq.(A8) is recognized as the percolation threshold for $L \to 0$, C_c', and is seen to be the same as C_c defined by Eq.(A6).

Expressing C_c from Eq.(A6) one finds

$$C_c = [(e-1)/ez]^{1/2} \cong 0.79506z^{-1/2}. \qquad (A9)$$

Several C_c values computed by Eq.(A9) are given in Table I. together with the results of earlier numerical calculations[3]. The agreement between Eq.(A9) and the Monte-Carlo values seems to be reasonable also as far as their dependence on z is concerned. The numerical data show an approximately $z^{-0.55}$ dependence which compares well with the $z^{-0.5}$ dependence predicted by the present treatment. Effective medium theory, which cannot account for any dependence on z, yields $C_c(em) = 0.333$.

Table I.

Percolation thresholds calculated by different methods

z	4	6	8	12
C_c(Ref. 3.)	0.425[a]	0.307[b]	0.243[c]	0.204[d] 0.195[e]
C_c(present)	0.397	0.325	0.281	0.230

[a]diamond, [b]simple cubic, [c]body centered cubic, [d]hexagonally close packed, [e]face centered cubic, all referring to site percolation

The numerical computation refer to site percolation, i.e. a case where free passage is barred by certain sites becoming insulating. It is contrasted by bond percolation where the bonds which connect different sites of the system are thought to be broken. Our model of adjacent subsystems being either conducting or insulating is apparently better related to site percolation.

Although coordination number appears to be important in our present treatment dimensionality plays here no role. The representation of the backbone as a space curve with no loops is due to this fact since such a curve can usually be folded out in a plane thus the difference between two and three dimensions disappears.

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