A fractal description of the dielectric response of disordered materials

1993 J. Phys.: Condens. Matter 5 4233

(http://iopscience.iop.org/0953-8984/5/25/013)

J. Phys.: Condens. Matter 5 (1993) 4233–4242. Printed in the UK

A fractal description of the dielectric response of disordered materials

G A Niklasson

Physics Department, Chalmers University of Technology, S-41296 Göteborg, Sweden

Received 19 February 1993

Abstract. We use a generalized diffusion equation to derive theories for the dielectric response of materials exhibiting fractal dynamics. Earlier results for the relaxation of charge carriers on fractal aggregates and fractal surfaces, as well as by fractal time processes, are obtained by simple scaling arguments. It is argued that the existence of cut-offs to the fractal structures and processes leads to dielectric response functions of the Cole–Cole form for bound charge carriers and of the Davidson–Cole form for quasi-free charge carriers. A novel expression is proposed for the case of a convolution of two fractal processes. These response functions are compared with other theoretical treatments and their relevance for experiments is assessed.

1. Introduction

It is by now well established that power-law relations in the frequency and time domains are a common feature of the dielectric properties of virtually all materials [1]. The existence of these power-law relations suggests a self-similarity in the underlying physical processes and it is therefore not surprising that fractals have been invoked repeatedly as an explanation of the dielectric response (see [2] and references therein). There exists a large literature on this subject which deals with the dielectric properties of fractal aggregates [3, 4], anomalous diffusion [5, 6], percolation clusters [7–9], fractal time processes [10, 11] and fractal surfaces [12–18] as well as general fractal circuits [19]. Considerable effort has recently been devoted to the study of the relationship between the fractal approach and previous empirical and theoretical relations [20, 21].

In this paper we elucidate the relationship between the various fractal approaches to the dielectric response and propose a closed form for the response function of a fractal process with an upper cut-off. We make use of a generalized diffusion equation [22, 23] and show that in the case of fractal processes the results can be expressed in the formalism of fractional calculus [24]. The methods used to obtain the dielectric response function are briefly reviewed. Capacitive as well as conductive processes are considered. Subsequently, in section 3 we consider the special cases of fractal aggregates, fractal surfaces and fractal time. The effects of cut-offs to the fractal processes are of crucial importance for obtaining response functions that can be compared to experimental data. In section 4 we treat the effect of two coexisting fractal processes, with different cut-offs, in the same material. Various approaches to the derivation of the dielectric response in this case are possible. Finally, in section 5 the main results of the paper and their possible experimental verification are discussed.

0953-8984/93/254233+10$07.50

2. Basic formalism

The motion of charge carriers in disordered materials can often be described as a diffusive process. The motion of a single charge carrier between localized states is then modelled as a discrete random walk. Here we are interested in the average behaviour of the physical quantities, which is the result of the average motion of many charge carriers. Hence, the effects of configurational averaging on the transport process are extremely important.

The configurational averaging leads to a generalized master equation if the microscopic transport process in each configuration is taken to be described by a master equation [25]. A generalized diffusion equation can be obtained as the continuum limit of the generalized master equation [2,23,26]. The case of electrical transport under an applied electric field, $E(t)$, was treated by Leal Ferreira [23] and others (see, e.g. [22]). Butcher [22] obtained the constitutive relation for the particle current density:

$$J_p(r, t) = -\int_0^t M(t-t')\nabla n(r, t')\,dt' - (e/kT)\int_0^t M(t-t')n(r, t')E(t')\,dt' \tag{1}$$

which leads to the generalized diffusion equation for the charge carrier density $n(r, t)$:

$$\partial n(r, t)/\partial t = \int_0^t M(t-t')\nabla^2 n(r, t')\,dt' + (e/kT)\int_0^t M(t-t')\nabla n(r, t')E(t')\,dt'. \tag{2}$$

Here e is the electron charge, k is Boltzmann's constant, T is the temperature and $M(t)$ is the inverse Laplace transform of the frequency-dependent diffusivity, $M(s)$, where $s = i\omega$. Equations (1) and (2) are composed of a diffusion term and a field-dependent drift term.

We now consider the steady-state solution to equation (1) in the linear response regime at small applied fields. For many electronic and ionic conductors one may assume that the charge carrier density can be written as a constant, n_0, plus possibly short-range fluctuations due to the discrete nature of the transport process. Hence we do not consider effects due to charge injection and inhomogeneous space charge distributions. Neglecting the fluctuations in the charge carrier density we arrive at a macroscopic transport equation for the electrical current density, $J(t) = eJ_p(t)$, which can be written as

$$J(t) = e(n_0 e/kT)\int_0^t M(t-t')E(t')\,dt' = e(n_0 e/kT)M(t) * E(t). \tag{3}$$

Here we have introduced the symbol $*$ to denote a convolution. The so-called Einstein relation allows the calculation of the frequency-dependent AC conductivity from the diffusivity according to [27]

$$\sigma(s) = en_0(eM(s)/kT). \tag{4}$$

Generalized diffusion equations for fractal time processes (fractional Brownian motion) and diffusive transport on fractal structures have recently been studied in detail by Giona and Roman [28]. They find that transport properties on fractals can be accurately modelled by the solutions of fractional diffusion equations (FDEs) [29]. The FDEs are of the same form as equation (2) without the drift term and with $M(t)$ being a power law of time. This suggests that we can use the full equations (1)–(3) to describe the motion of charge carriers on a fractal structure due to a weak applied electrical field.

When the diffusivity exhibits a power-law form, i.e. when $M(t) \simeq t^{n-1}$, the formalism of fractional derivatives is very convenient to use. We denote the derivation operator by D and write for the fractional derivative, of order $-n$ [24], of a function $f(t)$

$$\mathrm{D}^{-n} f(t) = (1/\Gamma(n))\big(t^{n-1} * f(t)\big) \tag{5}$$

where Γ denotes the gamma function. This definition is valid for $n > 0$, and can easily be generalized to arbitrary n [24]. The Laplace transform of equation (5) is simply s^{-n} multiplied by the Laplace transform of $f(t)$ [24]. This is a special case of the product rule for the Laplacian of a convolution.

The charge carriers in disordered materials can be of two kinds that we call quasi-free and bound. The expression 'quasi-free' is used in order to avoid confusion with free-electron conduction in metals. The quasi-free charge carriers can move in the material over macroscopic distances and give rise to a DC conductivity at low frequencies, or alternatively to a so-called low-frequency dispersion [30]. In this case the complex resistivity, $\rho(t)$, is used as a dielectric response function. It is defined so that $\rho(s) = 1/\sigma(s)$. By rearranging equation (3) we obtain

$$E(t) = \rho(t) * J(t) \tag{6}$$

in the time domain. The corresponding equation in the frequency domain is

$$\rho(s) = E(s)/J(s). \tag{7}$$

On the other hand, for the bound charge carriers, it is more appropriate to consider the relaxation of the polarisation $P(t)$ towards the equilibrium value $P_0(t) = \chi_0 E(t)$. Here χ_0 is the static dielectric susceptibility. Since $J(t) = \mathrm{D}P(t)$ and the driving force for the process is $P_0(t) - P(t)$, the relaxation equation becomes [31]

$$\mathrm{D}P(t) \simeq M(t) * \big(P_0(t) - P(t)\big) \tag{8}$$

which is a straightforward generalization of the Debye equation. The proper response function is now the dielectric susceptibility, which can be obtained from the frequency domain equation

$$\chi(s) = P(s)/E(s). \tag{9}$$

In the next section these general formulae are applied to the cases of fractal structures and fractal time processes.

3. Fractal conduction processes

In this section, we treat conduction on fractal aggregates, conduction phenomena at fractal surfaces and fractal time processes. We give expressions for $M(t)$ or its Laplace transform $M(s)$ for these cases, in order to derive the dielectric properties from the formalism in section 2. Explicit forms are suggested for the dielectric response functions of fractal processes with an upper cut-off, both for bound and quasi-free charges.

3.1. Conduction on fractal aggregates

We consider a random walk on a fractal structure [32,33]. The length scale, $L(t)$, covered by the random walk is related to time by the random walk dimension, d_w:

$$L(t) \simeq t^{1/d_w} \simeq s^{-1/d_w}. \tag{10}$$

This leads to a time- or frequency-dependent diffusion coefficient given by

$$D(s) \simeq L^2(t)/t \sim sL^2(s) \sim s^{1-2/d_w} \tag{11}$$

where $D(s)$ is actually equal to the diffusivity $M(s)$ in equations (1)–(3). Note, however, that $D(t) \neq M(t)$. The AC conductivity for this case must be scaled in the diffusion volume [34] and is not directly proportional to $M(s)$. Taking this into account we obtain [3,34]

$$\sigma(s) \simeq (L(s))^{d_f-2} M(s) \simeq s^{1-d_f/d_w} \tag{12}$$

where d_f denotes the fractal dimension of the aggregate. The length scale exponent $d_f - 2$ arises from the cross-sectional area divided by the length of the diffusion volume. The equation analogous to equation (3), which describes conduction on a fractal aggregate, then becomes

$$J(t) \simeq \mathrm{D}^{1-d_f/d_w} E(t) = \mathrm{D}^n E(t) \tag{13}$$

where we have included the effect of scaling in the diffusion volume.

3.2. Conduction processes at fractal surfaces

Various conduction phenomena can occur at or in the immediate vicinity of fractal surfaces. These processes can be described by equation (13) but with other values of the exponent n. For the case of diffusion-limited charge transport at a fractal interface it is realized that $d_w = 2$ and hence [14, 15, 35]

$$\sigma(s) \simeq (L(s))^{d_f-1} \simeq s^{(d_f-1)/2}. \tag{14}$$

The transport equation is still equation (13) with $n = (d_f - 1)/2$.

Another case concerns charge transport on a fractal surface. There exist strong indications [36] that for a wide class of surfaces one should use $d_w = d_f$. We obtain

$$\sigma(s) \simeq M(s) \simeq s^{1-2/d_f} \tag{15}$$

and hence equation (13) is valid with $n = 1 - 2/d_f$. This result was earlier obtained by Radoev and Tenchov [37].

The dielectric response of fractal blocking electrodes has recently attracted much attention [13–17]. It seems that the behaviour of such electrodes is very dependent on the specific fractal model chosen [14, 15].

3.3. Fractal time processes

For fractal time processes [10], it is well known that the conductivity is proportional to the diffusivity and scales as [2]

$$\sigma(s) \simeq M(s) \simeq s^{1-d_t} \tag{16}$$

where $d_t (= 2/d_w)$ is the dimensionality of the process. Hence the transport equation is given by equation (13) with $n = 1 - d_t$.

A fractal time process can be described by a distribution of waiting times [10]. Electrical conduction in an exponential distribution of activation energies constitutes a simple example [2, 38]. A more complicated situation occurs when a fractal time process is operating on a fractal structure. In this case the value of the random walk dimension defined by equation (10) is itself changed by the presence of the waiting time distribution. This problem has been studied in detail by Harder *et al* [39] by use of simple scaling arguments.

3.4. Analytic response functions

This section is concluded with some arguments giving the form of the dielectric response function for one fractal process. We make use of the fact that a fractal structure cannot extend to arbitrarily large length scales, nor can a fractal time process extend to infinitely long times. In situations encountered in practice there must be an upper cut-off to the fractal region. The AC conductivity is given by a power law for fractal processes; this is described by the transport equation $J(t) \simeq \mathrm{D}^n E(t)$. Next the influence of the cut-off for bound and quasi-free charge carriers is investigated.

For the case of bound charges, the polarization is relaxing towards equilibrium and the effect of the upper cut-off is included in the equilibrium polarizability $P_0(t)$. When conduction takes place on a finite fractal aggregate the cut-off is related to the size of the aggregate. From equation (8) we obtain $\mathrm{D}P(t) \simeq \mathrm{D}^n(P_0(t) - P(t))$ and hence

$$\chi(s) = \chi_0 P(s)/P_0(s) = \chi_0/\left(1 + (s\tau)^{1-n}\right) \tag{17}$$

where τ is a characteristic relaxation time characterising the upper cut-off. Equation (17) describes a relaxation peak in the frequency domain. It was proposed long ago by Cole and Cole [40] as an empirical formula describing the relaxation of various dielectric materials.

For quasi-free charge carriers, equations (6) and (7) were employed in order to determine the complex resistivity $\rho(s)$. The transport equation can be expressed as $E(t) \simeq \mathrm{D}^{-n} J(t)$, which means that the time-dependent resistivity has a power-law form and can be written as

$$\rho(t) \simeq t^{n-1} \exp(-t/\tau). \tag{18}$$

Here an exponential cut-off to the power-law behaviour of the resistivity has been introduced. Again τ is a relaxation time corresponding to the upper cut-off. We propose the simple exponential cut-off because now equation (18) reduces to an exponential function in the non-fractal limit ($n = 1$), as it should. The Laplace transform of equation (18) gives the final result for the response function according to

$$\rho(s) = \rho_0/(1 + s\tau)^n \tag{19}$$

where ρ_0 is the DC resistivity. This equation is of the same form as the well known empirical Davidson–Cole (DC) formula [41].

An alternative choice for the time-dependent resistivity would be to take $\rho(t) \simeq \exp(-(t/\tau)^{1-n})$, which leads to the so-called stretched exponential form for the response function [42]. In this case we have a slower cut-off than in the DC formula. In cases where the two approaches give noticeably different results, it appears that experimental data favour the DC expression [21, 43].

In disordered materials there may exist a distribution of the relaxation times that characterize the upper cut-offs. Hence the Cole–Cole and Davidson–Cole functions may have to be averaged over such distributions. One example arises in percolation theory, where one has to average over a power-law distribution of cluster sizes [9, 44].

4. Combinations of two fractal processes

A situation where two power-law processes are present in the dielectric properties and operate on different time scales is frequently encountered. One example of this is the so

called low-frequency dispersion [2,30]. Different approaches have been put forward for the derivation of the response function in this situation, and we think that further work is needed. Le Mehaute and Crepy [12,31] considered two fractal processes connected in series so that

$$E(t) \simeq \mathrm{D}^{-m}G(t) \simeq \mathrm{D}^{-m}\left(\mathrm{D}^{-q}J(t)\right) = \mathrm{D}^{-m-q}J(t) \tag{20}$$

where $G(t)$ denotes an intermediate physical quantity. Equation (20) describes the composite response function as the convolution of the response functions of the fractal processes. Furthermore, it is implied that $\sigma(s) \simeq s^{m+q}$. The complete dielectric response function is in general very complex, because the two processes have both lower and upper cut-offs that can occur at different times. Evidence for the addition of power-law indices, implied by equation (20), has been found in experiments on solid polymer electrolytes [31]. For these materials, charge transfer at the electrodes dominates the impedance at high frequencies, whereas at low frequencies, the charge transfer couples to bulk diffusion according to equation (20) [31].

By introducing an exponential cut-off, as in equation (18), for one of the processes, we can derive a simple theory for the low-frequency dispersion [30]. We assume that the two fractal processes couple in the high-frequency region, and that only one of them prevails at low frequencies. The complex resistivity is then obtained from

$$\rho(t) \simeq t^{n+p-2}\exp(-t/\tau) * t^{-p} \tag{21}$$

where the power-law indices m and q in equation (20) have been chosen in order to reproduce the empirically found [2,30] asymptotic behaviour at short and long times. Note that n and p can only take values between zero and unity. The Laplace transform is easily found and gives a generalized DC (GDC) expression, namely

$$\rho(s) \simeq (s\tau)^{p-1}/(1+s\tau)^{n+p-1}. \tag{22}$$

Dissado and Hill (DH) [45,46], on the other hand, made use of a related procedure for the dielectric susceptibility response function and found that

$$\chi(t) \simeq t^{-1}\left(t^{-n-p}\exp(-t/\tau) * t^{p}\right). \tag{23}$$

We conjecture that a convolution in the susceptibility (proportional to the capacitance) corresponds to processes operating in parallel. Since the DH theory has been used extensively in the fitting of experimental data [46] it is of interest to compare the GDC and DH theories. The power-law exponents have been chosen so that the asymptotic power-law regions at high and low frequencies are equal, but differences arise in the crossover between the two power laws. This is seen in figure 1, where we depict the inverse loss tangent, $\tan\varphi = (\rho_2(\omega)/\rho_1(\omega))$, as a function of frequency, ω. Here ρ_2 and ρ_1 denote the imaginary and real parts of the resistivity, respectively. We chose a high value of $n = 0.95$ in the computation in order to study a case with a wide crossover region. It is seen that for $p = 1$, when the GDC theory becomes equal to the DC expression (19), the agreement with the DH theory is very good. However, discrepancies arise for lower values of p; in particular, the approach to the low-frequency asymptote is slower for the DH theory than for the GDC theory. The differences are studied in more detail in figure 2, where we plot the loss tangent for the cases $p = 0.5$ and $p = 0.8$. In this plot the low-frequency region is amplified and the discrepancies are readily seen. It should be possible to distinguish between the two theories by careful experiments.

It should also be mentioned that Macdonald [38] has proposed a scheme for averaging over two exponential distributions of activation energies. The distributions exist for energies less than and larger than a certain cut-off energy, respectively.

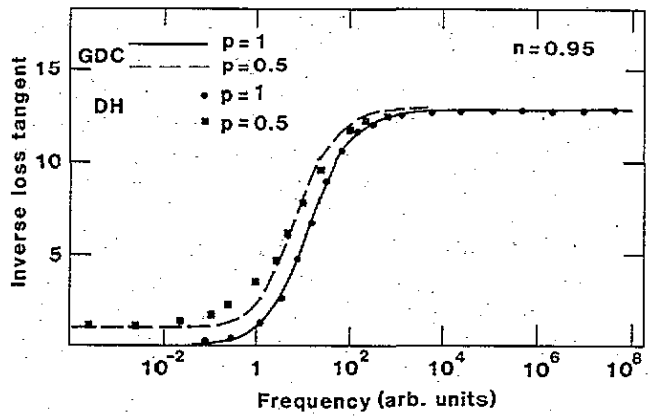

Figure 1. Inverse loss tangent as a function of frequency for the generalized Davidson–Cole (GDC) and Dissado–Hill (DH) theories. The frequency scale has been normalized so that the theories coincide at a frequency of 5 units. Calculations from the GDC and DH theories with exponents $n = 0.95$ and $p = 0.5$ and 1.0 are given by the symbols shown in the figure.

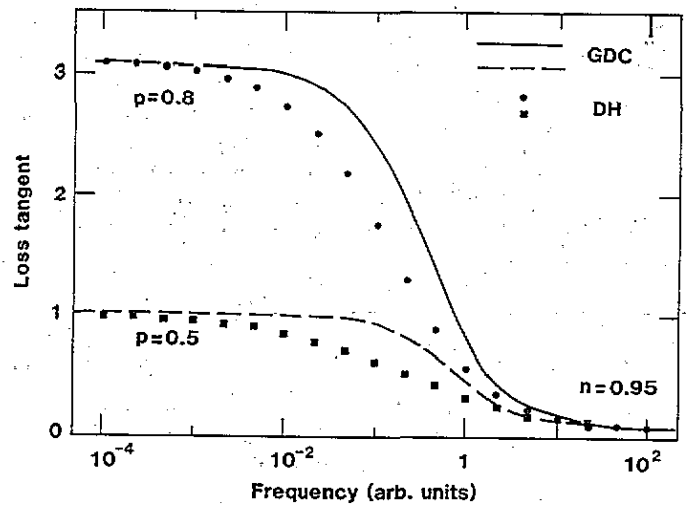

Figure 2. Loss tangent as a function of frequency for the generalized Davidson–Cole (GDC) and Dissado–Hill (DH) theories. The frequency scale has been normalized so that the theories coincide at a frequency of 5 units. Calculations from the GDC and DH theories with exponents $n = 0.95$ and $p = 0.5$ and 0.8 are given by the symbols shown in the figure.

5. Discussion

We conclude by discussing the experimental relevance of the response functions derived in sections 3 and 4. For the case of bound charge carriers, there exist some experimental problems. First, it is difficult to ascertain whether a relaxation peak is due to charge carriers or to some other effect such as dipoles. Secondly, in materials where charge carriers are assumed to be dominating the dielectric response, relaxation peaks are frequently seen superimposed on a background due to the quasi-free charges. This introduces uncertainties

in the determination of the response function, since it must be separated from that of the background. Despite this, some evidence for equation (17) actually exists. Hill [47] has analysed the dielectric response of SiO_2, Pt–SiO_2 and some chalcogenide glasses. The contributions due to bound and quasi-free charge carriers were separated. In five out of six analysed samples it seems that the relaxation peak due to bound charge carriers has a shape similar to that obtained from the Cole–Cole formula [47]. More work is needed in order to firmly establish equation (17) and to find out whether other factors may affect the dielectric response.

Various dielectric response functions that have been proposed for the case of quasi-free charge carriers were compared recently [21]. It was found that the DC theory, equation (19), gives a better agreement with experiments on thin films of Al_2O_3, silicon oxynitrides and SiO_2 [21] than alternative dielectric response functions, namely the stretched exponential and a simple averaging over a power-law distribution of transition rates (DTR). In addition the DC relation is in good agreement with experimental data for some ionic conductors [43,48]. In figure 3, we depict the inverse loss tangent for a silver-iodide–silver-borate glass [48,49] as a function of frequency. It is seen that the DC theory is in much better agreement with experiments than either the DTR or stretched exponential response functions. In all of these cases the exponent p could not be distinguished empirically from unity, hence the GDC theory in section 4 was not used. We remark that the overall similarity with the DH predictions makes the GDC theory an interesting alternative for the description of low-frequency dispersion [30].

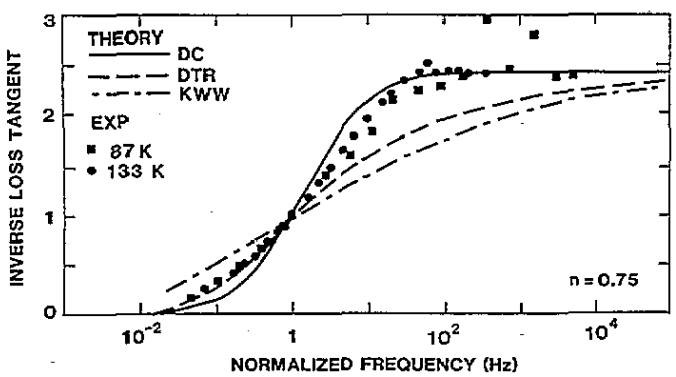

Figure 3. Inverse loss tangent as a function of frequency for $(AgI)_{0.6}(Ag_2O{:}2B_2O_3)_{0.4}$ glass. Squares denote our measurements at 87 K, while circles denote measurements from [49] at 133 K. Curves denote calculations with $n = 0.75$ using the Davidson–Cole (DC), power-law distribution of transition rates (DTR) and stretched exponential (KWW) expressions, as shown in the top left of the figure.

It should be emphasized that the approach to the dielectric response in this paper is restricted to the case when the charge-carrier density is essentially constant. For cases in which charge injection, space charges or decay of the charge-carrier density due to recombination or trapping occur, the full generalized diffusion equation (equations (1) and (2)) has to be solved. For example, it is known that the diffusivity and the particle decay exhibit different asymptotic behaviour at long times [50]. Measurements of both quantities may be used to distinguish behaviour due to fractal time and fractal structures [50].

In conclusion, dielectric response functions were derived for fractal conduction processes. These expressions can be compared to experimental data as well as to other

theoretical approaches. Several recent experiments appear to support a fractal interpretation of the dielectric response due to quasi-free and bound charge carriers.

Acknowledgments

This work was financially supported by grants from the Swedish Natural Science Research Council and the Swedish Technical Research Council. Valuable discussions with J R Stevens are gratefully appreciated.

References

[1] Jonscher A K 1983 *Dielectric Relaxation in Solids* (London: Chelsea Dielectrics)
[2] Niklasson G A 1987 *J. Appl. Phys.* **62** R1; 1989 *Physica* D **38** 260
[3] Clerc J P, Tremblay A-M S, Albinet G and Mitescu C D 1984 *J. Phys. Lett.* **45** L913
[4] Nigmatullin R R 1986 *Phys. Status Solidi* b **133** 425
[5] Gefen Y, Aharony A and Alexander S 1983 *Phys. Rev. Lett.* **50** 77
[6] Havlin S and Ben-Avraham D 1987 *Adv. Phys.* **36** 695
[7] Efros A L and Shklovskii B I 1976 *Phys. Status Solidi* b **76** 475
[8] Stroud D and Bergman D J 1982 *Phys. Rev.* B **25** 2061
[9] Clerc J P, Giraud G, Laugier J M and Luck J M 1990 *Adv. Phys.* **39** 191
[10] Shlesinger M F 1988 *Ann. Rev. Phys. Chem.* **39** 269
[11] Chen H and Wu X 1988 *Z. Phys.* B **71** 387
[12] Le Mehaute A and Crepy G 1983 *Solid State Ionics* **9–10** 17
Le Mehaute A 1984 *J. Stat. Phys.* **36** 665
[13] Liu S H 1985 *Phys. Rev. Lett.* **55** 529
Liu S H, Kaplan T and Gray L J 1986 *Solid State Ionics* **18–9** 65
[14] Nyikos L and Pajkossy T 1990 *Phys. Rev.* B **42** 709
Pajkossy T 1991 *J. Electroanal. Chem.* **300** 1
[15] Sapoval B, Chazalviel J-N and Peyriere J 1988 *Phys. Rev.* A **38** 5867
[16] Geertsma W, Gols J E and Pietronero L 1989 *Physica* A **158** 691
[17] Blunt M 1989 *J. Phys. A: Math. Gen.* **22** 1179
[18] Maritan A and Toigo F 1990 *Electrochim. Acta.* **35** 141
[19] Hill R M, Dissado L A and Nigmatullin R R 1991 *J. Phys.: Condens. Matter* **3** 9773
[20] Dissado L A and Hill R M 1989 *J. Appl. Phys.* **66** 2511
[21] Niklasson G A 1989 *J. Appl. Phys.* **66** 4350
[22] Butcher P N 1978 *Phil. Mag.* B **37** 653
Butcher P N and Clark J D 1981 *Phil. Mag.* B **43** 1029
[23] Leal Ferreira G F 1977 *Phys. Rev.* B **16** 4719
[24] Oldham K B and Spanier J 1974 *The Fractional Calculus* (New York: Academic)
[25] Klafter J and Silbey R 1980 *Phys. Rev. Lett.* **44** 55
[26] Kenkre V M and Knox R S 1974 *Phys. Rev.* B **9** 5279
[27] Scher H and Lax M 1973 *Phys. Rev.* B **7** 4491
[28] Giona M and Roman H E 1992 *J. Phys. A: Math. Gen.* **25** 2093
Roman H E and Giona M 1992 *J. Phys. A: Math. Gen.* **25** 2107
[29] Wyss W 1986 *J. Math. Phys.* **27** 2782
[30] Jonscher A K 1978 *Phil. Mag.* B **38** 587
[31] Le Mehaute A 1989 *The Fractal Approach to Heterogeneous Chemistry* ed D Avnir (New York: Wiley) p 311–28
[32] Alexander S and Orbach R 1982 *J. Phys. Lett.* **43** L625
[33] Rammal R and Toulouse G 1983 *J. Phys. Lett.* **44** L13
[34] Liu S H 1986 *Solid State Physics* vol 39 (New York: Academic) p 207
[35] Wong P-Z 1987 *AIP Conf. Proc.* **154** 304
[36] Giugliarelli G, Maritan A and Stella A L 1990 *Europhys. Lett.* **11** 101
[37] Radoev B P and Tenchov B G 1987 *J. Phys. A: Math. Gen.* **20** L159

[38] Ross Macdonald J 1985 *J. Appl. Phys.* **58** 1971; 1987 *J. Appl. Phys.* **62** R51
[39] Harder H, Havlin S and Bunde A 1987 *Phys. Rev.* B **36** 3874
[40] Cole K S and Cole R H 1941 *J. Chem. Phys.* **9** 341
[41] Davidson D W and Cole R H 1951 *J. Chem. Phys.* **19** 1484
[42] Moynihan C T, Boesch L P and Laberge N L 1973 *Phys. Chem. Glasses* **14** 122
[43] Pathmanathan K and Stevens J R 1990 *J. Appl. Phys.* **68** 5128
[44] Stauffer D 1985 *Introduction to Percolation Theory* (London: Taylor and Francis)
[45] Dissado L A and Hill R M 1983 *Proc. R. Soc* A **390** 131
[46] Dissado L A and Hill R M 1984 *J. Chem. Soc. Faraday Trans.* II **80** 291
[47] Hill R M 1985 *Thin Solid Films* **125** 277
[48] Niklasson G A, Brantervik K and Börjesson L 1991 *J. Non-Cryst. Solids* **131–3** 1096
[49] Börjesson L, Torell L M, Liu C, Martin S W and Angell C A 1987 *Phys. Lett.* **125A** 330
[50] Blumen A, Klafter J, White B S and Zumofen G 1984 *Phys. Rev. Lett.* **53** 1301