

DYNAMICS OF IONIC MOTION IN POLYMERIC IONIC CONDUCTORS

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Carrier transport in disordered systems is often treated theoretically using random hopping models. When the host medium is a polymer above its glass transition temperature such theories have to be modified to account for microscopic structural changes in the polymer on the experimental timescale. This paper describes the results obtained to date for models that take into account this dynamic nature of the disorder, and their applications to ionic motion in polymeric ionic conductors.

1. Introduction

There is substantial evidence that transport of charge carriers in highly viscous liquids as polymeric ionic conductors above their glass transition temperature is dominated by microscopic segmental motions of the host medium [1]. Therefore, while hopping models have been very useful in studies of diffusion and conduction in a variety of solid-state disordered systems [2], such models cannot be used without modification to describe carrier transport in polymer electrolytes. Instead most workers have used theories based on free volume [3] or configuration entropy [4] concepts. While such theories incorporate the thermodynamics of the host polymer in an explicit way, they are not well suited for treating dynamical phenomena. Recently, several attempts were made to generalize hopping models so as to take into account the dynamics of the host.

Two relatively simple models which take into account the host dynamics in addition to the carrier hopping have been investigated in the past few years. In the first model, due to Druger, Nitzan, and Ratner (DNR) [5-8], the host dynamics is described by a series of instantaneous "renewal" events. These events occur at random times governed by a renewal time distribution $f(t)$, whose first moment $\tau_{\text{ren}} = \int_0^{\infty} t f(t) dt$ is the average renewal time. In the renewal process the network bonds and/or sites are reassigned according to the (static) distribution which determines the random nature of the system. Thus, for a random bond model with a fraction p of available bonds, the positions of these available bonds are reassigned at each renewal event, whereupon the carrier motion proceeds on the newly defined lattice.

In the second model, due to Harrison and Zwanzig (HZ) [9], individual bonds rather than the whole lattice are changed randomly in time. In both models the host dynamics is assumed to be spatially uncorrelated (i.e. changes in one location occur independently of those in another location) and also uncorrelated with the position and state of the hopping carrier.

While the HZ model seems to be more realistic than the DNR model (both are oversimplified due to the assumptions stated above), the latter has yielded some exact solutions of quite general nature while the HZ model has been solved only within an effective medium approximation (EMA). Remarkably, the EMA result for the HZ model is very similar to the corresponding exact result of the DNR model.

In addition, recent studies of the so-called stirred percolation problem [10-13] (where charge carriers move only within one component of a microemulsion formed from two immiscible liquids) and of diffusion of a

tracer particle in a system of other particles characterized by a different jump rate [14] are closely related to the work described below. Refs. [10–14] focus on scaling relations near the underlying lattice percolation threshold.

2. Diffusion and conductivity in dynamically disordered systems

The following discussion focuses on the diffusion coefficient D associated with a single moving particle in a dynamically disordered network. The corresponding conductivity σ is obtained from D using the Nernst-Einstein relation

$$\sigma = (nq^2/k_B T)D, \tag{1}$$

where n is the density, q is the charge of the (assumed non-interacting) carriers, and T is the temperature. We begin with the DNR model.

Consider first dc diffusion. For the dc diffusion coefficient DNR derive the remarkably simple result [5]

$$D = n_d \overline{\langle r^2 \rangle}_{\text{ren}} / \tau_{\text{ren}}, \tag{2}$$

where $n_d = 2d$ (d is the dimensionality), τ_{ren} is the average renewal time, and $\overline{\langle r^2 \rangle}_{\text{ren}}$ is the mean-square displacement during a renewal period (in the presence of drift r should be replaced by $r - \langle r \rangle$); $\langle \ \rangle$ denotes the usual ensemble average while the bar denotes an average over the renewal time distribution. Thus, if $\langle r^2 \rangle_0(t)$ is known as a function of time t for any system with static disorder $\overline{\langle r^2 \rangle}_{\text{ren}}$ is obtained from

$$\overline{\langle r^2 \rangle}_{\text{ren}} = \int_0^\infty dt f(t) \langle r^2 \rangle_0(t). \tag{3}$$

The subscript “0” in $\langle r^2 \rangle_0$ denotes that this quantity corresponds to the statically disordered system. If τ_{ren} is very short (relative to the hopping rate w) this just yields the mean-square displacement in the averaged network (for a bond percolation network having transition rates w with probability p and 0 with probability $1-p$, the averaged network rates are all given by wp). If $\tau_{\text{ren}} \gg t_{\text{obs}} \gg w^{-1}$ (t_{obs} is the observation time) this is the static hopping limit where the dynamics is controlled by w and p . If $t_{\text{obs}} \gg \tau_{\text{ren}} \gg w^{-1}$ the dynamics is affected by τ_{ren} . In particular, for a percolation network below the percolation limit, the dynamics is controlled by τ_{ren} and p .

As an explicit example consider a one-dimensional random chain with bonds available and unavailable with probabilities p and $1-p$, respectively. This system is below the percolation threshold for $p < p_c = 1$, and

$$\langle x^2 \rangle(t) \rightarrow a^2 p / (1-p)^2 \tag{4}$$

for $t \rightarrow \infty$, where a is the lattice constant. Provided that τ_{ren} is much larger than the time it takes to fill the average cluster, the diffusion coefficient D is given by

$$D = pa^2 / 2(1-p)^2 \tau_{\text{ren}}. \tag{5}$$

For $d > 1$, below the percolation limit we get similarly

$$D \sim |p - p_c|^{-x} / \tau_{\text{ren}} \tag{6}$$

($x = \nu(2 - \beta/\nu) \simeq 2.5$ for $d=2$, 1.35 for $d=3$) as long as τ_{ren} is long relative to the time it takes to fill the average finite cluster. As $p \rightarrow p_c$ from below, τ_{ren} becomes short relative to this time and we have a cross-over to the behavior described by eqs. (2) and (3), where $\langle r^2 \rangle(t)$ should be calculated on the percolating cluster [11,13]. No further transitions are expected when p increases beyond p_c .

Turning now to ac diffusion our starting point is the following expression due to Scher and Lax [15]

$$D(\omega) = n_d \lim_{\epsilon \rightarrow 0^+} (i\omega)^2 \int_0^{\infty} dt \exp(-i\omega t - \epsilon t) \langle r^2 \rangle(t) . \quad (7)$$

The original DNR evaluation of the frequency-dependent diffusion [6] was carried out for a particular (Poisson) distribution of renewal times. The probability that the system undergoes N renewal events during time t is taken to be

$$P(N, t) = [(\lambda t)^N / N!] \exp(-\lambda t) \quad (8)$$

so that the waiting-time distribution for the time between renewals is exponential

$$f(t) = \lambda \exp(-\lambda t) \quad (9)$$

and

$$\tau_{\text{ren}} = \lambda^{-1} . \quad (10)$$

For this system DNR found the useful relation [6]

$$D(\omega) = n_d (\lambda + i\omega)^2 \int_0^{\infty} dt \exp[-(\lambda + i\omega)t] \langle r^2 \rangle_0(t) , \quad (11)$$

where again $\langle r^2 \rangle_0$ is evaluated in the static disorder limit. Eq. (11) implies that

$$D(\omega) = D_0(\omega - i\lambda) \quad (12)$$

so that if D_0 is known explicitly as a function of ω in the static disorder limit, $D(\omega)$ in the renewing system (with the renewal time distribution (9)) becomes immediately available. Another interesting result is obtained by comparing eqs. (7) and (11) to yield the following relation between the mean-square displacements in the renewing and in the equivalent static-disorder system

$$\langle r^2 \rangle(t) = \langle r^2 \rangle_0(t) \exp(-\lambda t) + \lambda \int_0^t (2 + \lambda t - \lambda \tau) \exp(-\lambda \tau) \langle r^2 \rangle_0(\tau) d\tau . \quad (13)$$

Finally note that for $\omega = 0$, eq. (11) yields eq. (2) with $\tau_{\text{ren}} = \lambda^{-1}$ and $\langle r^2 \rangle_{\text{ren}}$ given by eqs. (3) and (9).

The simplicity of the relations (eqs. (2) and (12)) between the transport properties of the dynamically disordered system and the same properties of the static system results from the great simplification associated with the model assumptions that describe the medium evolution as a random series of uncorrelated global renewal events. Any property whose total magnitude is given by additive contributions from the processes occurring in different renewal periods may be calculated from the same property for the static system by averaging over the times in which the renewal events occur. Such a procedure was recently carried out in great generality by Druger [8]. He considered a general observable $G(t)$ whose time evolution may be written as a sum of contributions from different renewal periods

$$G(t) = g_0(\tau_0) + \sum_{i=1}^N g(\tau_i) \quad (14)$$

with $\tau_N = t$ while τ_n , $n=0, \dots, N-1$ are the times between renewal events where $g_0(0) = g(0) = 0$. In principle the function $g_0(x)$ may be different from $g(x)$, because g_0 describes the evolution of G starting from a random instant of time while g corresponds to a process which starts immediately following a renewal. In a similar way the two time distributions: $\phi(\tau) d\tau$, the probability for a renewal to occur in the time interval $(\tau, \tau + d\tau)$ measured from some random time, and $\psi(\tau) d\tau$, a similar probability distribution for τ measured from the previous

renewal, may in principle be different. Assuming that these distributions as well as the properties $g_0(\tau)$ and $g(\tau)$ of the static system are known, Druger [8] obtained the following expression for the Laplace transform \hat{G} of G :

$$\langle \hat{G}(i\omega) \rangle^2 = \frac{1}{(i\omega)^{-2}} \left[\frac{\hat{\phi}(i\omega)}{\hat{\Psi}(i\omega)} L \left(\Psi \frac{dg_0}{dt} \right) + i\omega L \left(\Phi \frac{dg}{dt} \right) \right], \quad (15)$$

where

$$\Phi(t) = \int_0^\infty \phi(\tau) d\tau, \quad \Psi(t) = \int_0^\infty \psi(\tau) d\tau \quad (16a)$$

and (for a function $h(t)$)

$$L[h(t)] = \hat{h}(i\omega) = \int_0^\infty dt \exp(-i\omega t) h(t). \quad (16b)$$

The result (15) is the most general consequence of the DNR model and may be shown to yield the results (2), (11), (12) and (13) in the appropriate limits. In particular it should be noticed that even though the earlier results (2) and (11)–(13) were obtained for a random bond model, the result (15) is based only on the uncorrelated nature of the renewal events, which serves to show that also our former results are valid for any type of randomness.

We now turn to the HZ model [9]. The mathematical statement of this model is given by eqs. (17), (18):

$$\frac{d\mathbf{P}}{dt} = - \sum_{\mu} \sigma_{\mu}(t) \mathbf{V}_{\mu} \cdot \mathbf{P}, \quad (17a)$$

$$\mathbf{P} = \sum_i P_i |i\rangle, \quad (17b)$$

$$\mathbf{V}_{\mu} = (|i\rangle - |j\rangle)(\langle i| - \langle j|), \quad (17c)$$

where $\mu = (ij)$ is a bond index. If σ , the transition rate associated with bond μ , were time independent, this would have been the regular master equation for a random walk in a random bond system. However, the probability $f_{\mu}(\sigma, t)$ that the bond μ will be in state σ is itself governed by a master equation

$$\frac{\partial f_{\mu}(\sigma, t)}{\partial t} = \sum_{\sigma'} M_{\mu}(\sigma, \sigma') f_{\mu}(\sigma', t) \quad (18)$$

with

$$M_{\mu}(\sigma, \sigma) = - \sum_{\sigma' \neq \sigma} M_{\mu}(\sigma', \sigma).$$

HZ considered only the two-bond-state system, i.e. when the bond can be either available ($\sigma = 1$) or broken ($\sigma = 0$). For this case evaluation of the diffusion coefficient in the effective medium approximation yields [9]

$$D(\omega) = D_0(\omega - 1/\tau), \quad (19)$$

where

$$p/\tau = M(1,0), \quad q/\tau = M(0,1), \quad p+q=1. \quad (20)$$

Eq. (19) is identical to the DNR result (12). This suggests that this result is valid in more general circumstances than those implied by the DNR or the HZ model assumptions. Indeed, a similar result is obtained in the scaling

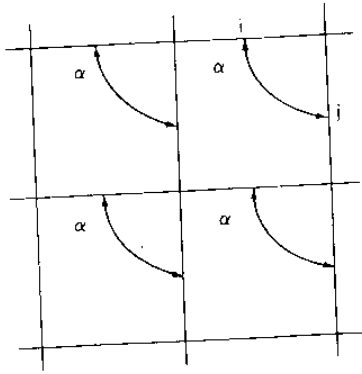


Fig. 1. The correlated bond rearrangement model.

treatment of the stirred percolation model of Grest et al. [11]. It may also be shown that the HZ effective medium approach, when applied to a fluctuating random site (rather than a random bond) model also leads to the same result. Eqs. (12) or (19), however, are no longer valid in more general circumstances. For example, in the case where each bond can be in several states ("grey" transport, where σ can attain several values between zero and one) effective medium theory leads to a diffusion rate which depends on $n-1$ relaxation times (where n is the number of the bond states).

In the systems considered above, the underlying polymer motion was modeled in the simplest possible way: in the DNR model the random bond network is rearranged completely at each renewal event. In the HZ model each bond fluctuates between its open and closed states independently from the other bonds. It is of interest to ask to what extent bond-bond correlations in the renewal processes affect the diffusion through the network. Harris et al. [16] considered this problem for a one-dimensional DNR-type model, and showed, by simple simulations, that if the bond renewals are restricted to occur between nearest neighbor sites, the dc conductivity is reduced relative to that for random renewal, since the carrier must occasionally wait for the (slower) renewal chain of events before it can proceed along the chain. More recently, this question was taken up by Granek and Nitzan [17], who have considered an extension of the HZ model in two dimensions. In this generalized model there are two kinetic processes: each bond can fluctuate between its open and closed position with rate $1/\tau$ as in the HZ case. In addition, pairs of adjacent bonds can interchange status (see fig. 1) so that if one of them is closed and the other is open a rate process with rate α can transform them to the opposite (open-closed) configuration. In the extreme case where $1/\tau=0$ the α process corresponds to a gate-like segmental motion, which may alternately close one of two available pathways. This scheme is somewhat reminiscent of the Grotthus model for proton transport in ice which is facilitated by rotation of the water molecules.

The basic network entity of this model is a two-bond unit (e.g. the ij pair in fig. 1) which can be in one of four states (11), (10), (01), or (00). These units evolve according to the stochastic rate equation

$$\partial f / \partial t = \mathbf{\Omega} \cdot \mathbf{f}, \quad (21)$$

where $\mathbf{f}(t)$ is the four-state probability vector whose components $f(\lambda\mu, t)$ are the probability to find the pair of bonds in state $\lambda\mu$ ($\lambda\mu=00, 10, 01, 11$), and where the rate matrix $\mathbf{\Omega}$ is

$$\mathbf{\Omega} = \begin{pmatrix} -2p/\tau & q/\tau & q/\tau & 0 \\ p/\tau & -(\tau^{-1} + \alpha) & \alpha & q/\tau \\ p/\tau & \alpha & -(\tau^{-1} + \alpha) & q/\tau \\ 0 & p/\tau & p/\tau & -2q/\tau \end{pmatrix} \quad (22)$$

with p/τ and q/τ (with $q=1-p$) the rates at which bonds are created and destroyed independently and with

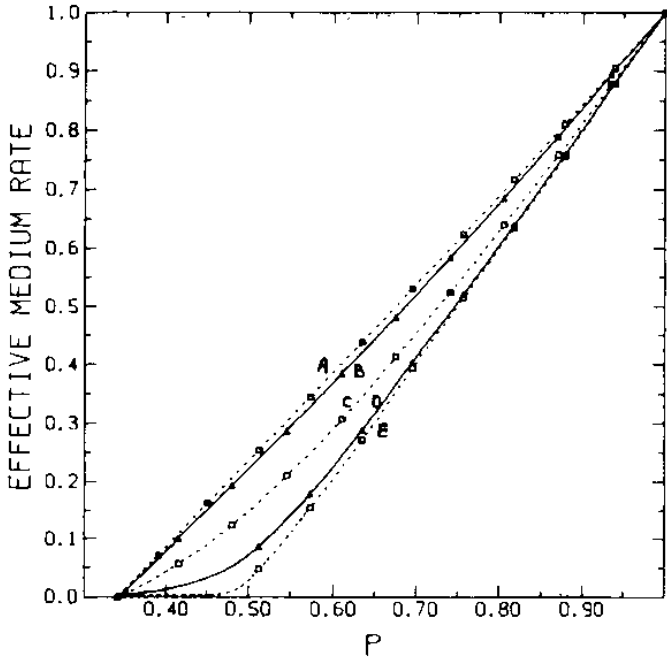


Fig. 2. Effective medium rate as a function of the bond filling factor p for the adjacent bond interchange model $1/\tau=0$. Curves (A)–(E) correspond to different values of α as follows (in units of the single bond hopping rate): (A) $\alpha=15.0$, (B) $\alpha=3.0$, (C) $\alpha=0.25$, (D) $\alpha=0.025$, (E) $\alpha=0.004$. $\omega=0$ in all cases.

the rate α as described above, i.e. the rate for interbond exchange. The rate matrix (22) describes a situation in which the equilibrium fraction of unbroken bonds is p .

From the percolation theory point of view the limit $\tau=\infty$ constitutes a new percolation problem where on top of the usual statically disordered bond network we have introduced a dynamical interchange between the state of adjacent bonds. Effective medium solution of this problem may be obtained [17] by generalizing the approach of Harrison and Zwanzig [9]. For the two-dimensional square lattice we find that the effective medium percolation threshold (which is $p=\frac{1}{2}$ in the static, $\alpha=0$ limit) is downshifted to $p=0.341$. This threshold does not depend on the magnitude of α . Some results for the p dependence of the effective medium hopping rate are shown in fig. 2 for $1/\tau=\omega=0$ and for different values of α . Note that all rates, α , $1/\tau$, and the resulting effective medium rate, are expressed in units of the single-bond hopping rate. When $1/\tau$ and/or ω are different from 0 the threshold moves to $p=0$ (fig. 3); however, the general form of the curves does not change.

These results clearly show the role of bond motions in particle diffusion on a disordered matrix. For typical cases where α and $1/\tau$ are $\approx O(1)$ or larger the matrix motions quantitatively change the diffusion rate above the percolation threshold. Below the percolation threshold diffusion is practically dominated by these matrix motions. Correlations in matrix motion, expressed by the magnitude of α relative to $1/\tau$, affect the rate quantitatively but do not change the qualitative behavior obtained in earlier work for the case of uncorrelated renewal events.

3. Relation to the physical world

The relations given above allow the results of extensive work on static disorder hopping and percolation models to be applied in studying dynamic disorder transport. The main problem incurred in the application of these results to actual systems is our ignorance concerning the parameters which determine the dynamics of the host network. Even with this limitation, the results of the previous section provide a convenient framework for analyzing experimental systems [18].

Another approach attempted by us some time ago [7] is to try to identify the correspondence between the parameters of the dynamic disorder hopping (DDH) theories and more-traditional approaches such as free

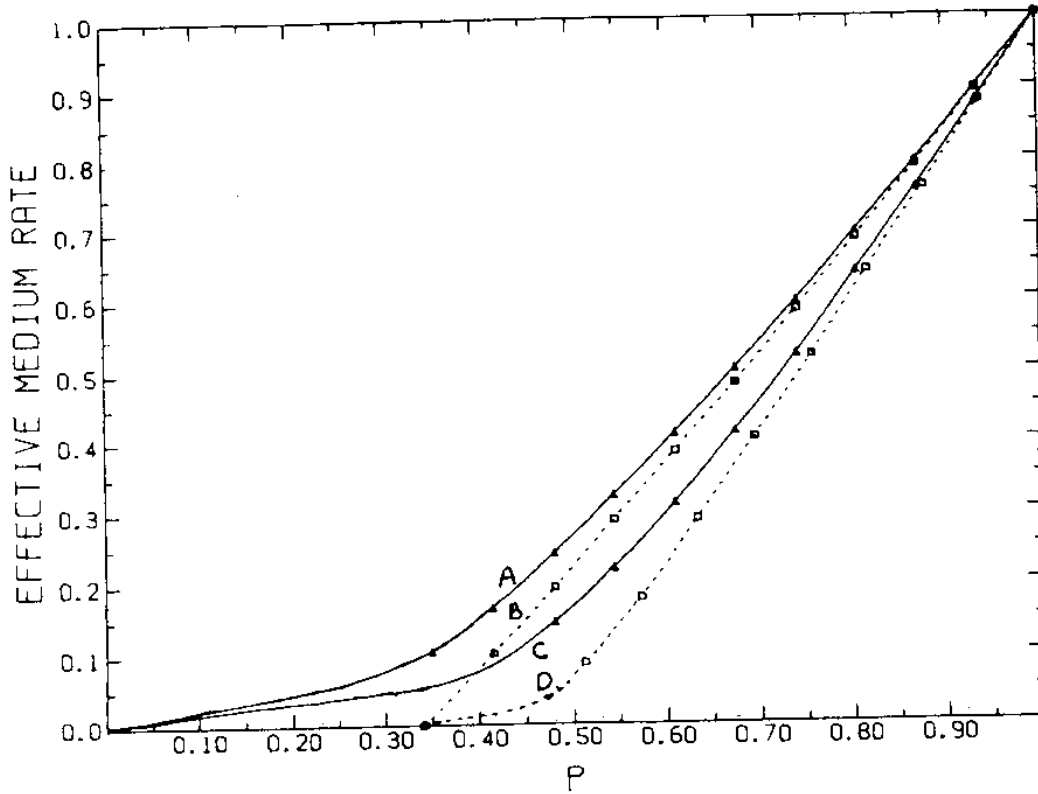


Fig. 3. Same as fig. 2. Curves (A)–(D) correspond to different values of α and $1/\tau$ as follows: (A) $1/\tau=0.1$, $\alpha=3.0$; (B) $1/\tau=0$, $\alpha=3.0$; (C) $1/\tau=0.1$, $\alpha=0.025$; (D) $1/\tau=0$, $\alpha=0.025$. $\omega=0$ in all cases. Curves (B) and (D) are the same as the corresponding curves in fig. 2.

volume theory [3] (FVT), which has been extensively used to fit experimental data for transport and relaxation in polymers. A somewhat modified version of the procedure of ref. [7] is the following: if we adopt FVT for the motion of the polymer segments, τ_{ren} can be estimated from

$$\tau_{ren} \simeq (a_{seg}/u_{seg}) \exp(\gamma V_{seg}^*/V_f), \quad (23)$$

where V_{seg}^* is the smallest volume needed for segment motion, of order equal to the characteristic volume of a segment, V_f is the free volume [3], a_{seg} is the linear dimension associated with a typical segment, and u_{seg} is the thermal velocity related to the segment mass and to the temperature. γ is a constant of order $\lesssim 1$. Eq. (23) is obtained by equating the FVT expression for the diffusion coefficient associated with local segmental motion with the expression $D_{seg} = a_{seg}^2/\tau_{ren}$. This local segmental motion becomes mostly frozen below the glass transition temperature of the polymer.

For an ion moving in this polymer network we are interested not in local motion but in long-range motion by hopping. The polymer host plays the role of a random network for this motion and we identify [7] the filling factor p of this network with

$$p = \exp(-\gamma_1 V_1^*/V_f), \quad (24)$$

where γ_1 is another constant and where V_1^* is the minimum volume needed for the ion hopping. p vanishes for $V_f \rightarrow 0$ and becomes unity as $V_f \rightarrow \infty$. Assuming that $p < p_c$ and that the individual ion hopping rates are fast relative to the τ_{ren} scale, eq. (6) is relevant for the dc diffusion of the ion. This leads to

$$D_{ion} = \frac{[p_c - \exp(-\gamma_1 V_1^*/V_f)]^{-x}}{(a_{seg}/u_{seg}) \exp(\gamma V_{seg}^*/V_f)}. \quad (25)$$

The temperature-dependent terms in (25) are $u_{seg} \sim \sqrt{T}$ and $V_f = V_0 + \alpha(T - T_g)$, where T_g is the glass transition temperature, α is the thermal expansion coefficient of the polymer above the glass transition temperature

(the constant V_0 may be further expanded with respect to pressure). Eq. (25) thus provides the expected temperature dependence of the ion diffusion coefficient and (by eq. (1)) of the conductivity. It should be kept in mind though that this oversimplified treatment has neglected important temperature-dependent processes such as ion-ion interaction and ion pairing.

The main qualitative prediction of eq. (25) is that the temperature (and pressure) dependence of D_{ion} should follow the temperature and pressure dependence of intrinsic polymer properties (e.g. viscosity) since they are controlled according to (25) by the polymer motion. This prediction is often confirmed by experiments [1]. We recall (see section 2) that other regimes exist where the ion motion is affected also by the (temperature-dependent) ion hopping rate.

4. Conclusions

In this paper we have summarized the results obtained thus far on the DDH problem. We have seen that under some model assumptions general relations exist between the transport properties in such systems and the same properties of the corresponding static systems. While the relevance to experimental systems is obvious and can be demonstrated even at this stage, it is also evident that further elaborations of the theory are needed in order to make it a quantitative tool. In particular the following questions still remain open.

(a) How do interactions between mobile ions affect the diffusion and conductivity? In framework ionic conductors mobility was found to depend strongly on such (screened coulombic) interactions [19]. In polymeric electrolytes where mobility is dominated by host motion such interactions may still be extremely important and can modify the transport behavior in an essential way. It should be remembered that in addition to theoretical difficulties in treating such many-body problems, the experimental situation is also complicated by the fact that varying the ion concentrations in the polymer host in an effort to understand the role of interionic interactions often strongly affects the motion of the host polymer, since increased concentrations of charged ions produce a weak crosslinking effect on the chains [20]. Nevertheless, in most polymer electrolyte systems of current experimental interest the overall ionic concentration lies in the range 1–10 molar. At these very high concentrations, nearly those of solvated molten salts, interactions among ions are clearly very important.

(b) What are the implications of DDH models for properties of the polymer network other than ionic motions? The arguments in section 3 suggest that the polymer segmental motion and the motion of an ion inside the polymer may be described within the same theoretical framework. The ability to describe polymer mechanical properties using parameters of the DDH theory will be an important step towards establishing a firm theoretical connection between mechanical and charge-transport properties of the host polymer. Efforts to construct such theories are currently underway.

(c) How can the results of experimental studies of such properties as microwave conductivity [21], dielectric relaxation, Brillouin scattering [22] linewidths, inelastic neutron scattering [23], or NMR relaxation times [24] be amalgamated into the dynamic disorder models? Properties such as the bond switch rate α , or the renewal time τ_{ren} , are clearly and closely related to these relaxation properties, but a more detailed understanding of the nature of the ionic conduction process is needed before any facile assignment of a relaxation time to a renewal process can be ventured [1,3].

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References

- [1] H. Cheradame, in: IUPAC macromolecules, eds. H. Benoit and P. Rempp (Pergamon Press, Oxford, 1982);
C. Berthier, in: Polymer electrolyte reviews, eds. J.R. MacCallum and C.A. Vincent (Elsevier, Amsterdam, 1987);
M.A. Ratner, in: Polymer electrolyte reviews, eds. J.R. MacCallum and C.A. Vincent (Elsevier, Amsterdam, 1987).
- [2] H. Bottger and V.V. Bryksin, Hopping conduction in solids (Akademie Verlag, Berlin, 1985).
- [3] D.F. Shriver and M.A. Ratner, Chem. Rev., to be published;
M.H. Cohen and D. Turnbull, J. Chem. Phys. 31 (1959) 1164,
M. Watanabe and N. Ogata, in: Polymer electrolyte reviews, eds. J.R. MacCallum and C.A. Vincent (Elsevier, Amsterdam, 1987);
A. Killis, J.F. Le Nest, H. Cheradame and A. Gandini, Makromol. Chem. 183 (1982) 2835.
- [4] G. Adam and J.H. Gibbs, J. Chem. Phys. 43 (1965) 139;
C.A. Angell, Solid State Ionics 18/19 (1986) 72.
- [5] S.D. Druger, A. Nitzan and M.A. Ratner, J. Chem. Phys. 79 (1983) 3133.
- [6] S.D. Druger, M.A. Ratner and A. Nitzan, Phys. Rev. B31 (1985) 3939.
- [7] S.D. Druger, M.A. Ratner and A. Nitzan, Solid State Ionics 9/10 (1983) 1115.
- [8] S.D. Druger, in: Transport and relaxation processes in random materials, ed. M.F. Shlesinger (World Scientific, Singapore, 1986).
- [9] A.K. Harrison and R. Zwanzig, Phys. Rev. A32 (1985) 1072.
- [10] M. Lagues, J. Phys. (Paris) 40 (1979) L331.
- [11] G.S. Grest, I. Webman, S.A. Safran and A.L.R. Bug, Phys. Rev. A33 (1986) 2842.
- [12] A.R. Kerstein and B.F. Edwards, Phys. Rev. 33 (1986) 3353.
- [13] A.L.R. Bug and Y. Gefen, Phys. Rev. A35 (1987) 1301.
- [14] R. Kutner and K.W. Kehr, Phil. Mag. A48 (1983) 199.
- [15] H. Scher and M. Lax, Phys. Rev. B7 (1973) 4491.
- [16] C. Harris, A. Nitzan, M.A. Ratner and D.F. Shriver, Solid State Ionics 18/19 (1986) 151.
- [17] R. Granek and A. Nitzan, to be published.
- [18] S.M. Ansari, M. Brodwin, M. Stainer, S.D. Druger, M.A. Ratner and D.F. Shriver, Solid State Ionics 17 (1985) 101.
- [19] S.H. Jacobson, M.A. Ratner and A. Nitzan, J. Chem. Phys. 77 (1982) 5752;
Y. Boughaleb, R.O. Rosenberg, M.A. Ratner and A. Nitzan, Solid State Ionics 18/19 (1986) 160;
U. Thomas and W. Dieterich, Z. Physik B62 (1986) 287;
G.S. Grest and M.H. Cohen, Advan. Chem. Phys. 48 (1981) 455.
- [20] J.F. Le Nest, A. Gandini, H. Cheradame and J.P. Cohen-Addad, Macromolecules, to be published.
- [21] T. Wong, M. Brodwin, B. Papke and D.F. Shriver, Solid State Ionics 5/6 (1981) 889.
- [22] L.M. Torell and C.A. Angell, unpublished;
L. Borjesson, S.W. Martin, L.M. Torell and C.A. Angell, Solid State Ionics 18/19 (1986) 631.
- [23] C. Poinsignon and C. Berthier, Abstracts First Int. Symp. Polymer Electrolytes, St. Andrews, Scotland, 1987.
- [24] A. Killis, J.F. Le Nest, H. Cheradame and J.P. Cohen-Addad, Polym. Bull. 6 (1982) 351;
M. Minier, C. Berthier and W. Gorecki, J. Phys. (Paris) 45 (1984) 739.