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Continuous-time random-walk model for superionic conductors

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We formulate a continuous-time random-walk model for the motion of conducting ions in β-alumina which accounts for correlation effects as well as free and jump diffusion mechanisms in a unified way. The calculated frequency-dependent conductivity and incoherent intermediate scattering function agree with experimental results, suggesting that the model provides a good description of the underlying processes, including sublattice disorder effects.

I. INTRODUCTION

In recent years the properties of superionic conductors have been intensively studied both from the theoretical and experimental points of view. These materials show a solid-phase ionic conductivity close to that of a molten salt.[1] A striking feature of β-alumina–type conductors is the behavior of its ac conductivity, which first increases with frequency, peaking at the microwave or far-infrared range, to vanish finally as[2] $1/\omega^2$. Also of interest is the incoherent intermediate scattering function (or dynamic structure factor), which for many superionic conductors present a complicated, non-Lorentzian line shape.[3,4]

Diverse models had been proposed to explain this behavior. Those based on a Fokker-Planck equation description of a particle diffusing in a periodic potential proved to be particularly successful,[5] although its solution in terms of continued fractions is somewhat cumbersome and must be worked generally in numerical form.

On the other hand jump-diffusion models present a simpler, though less realistic, description of the process.[6] In these models the ions are pictured as remaining for a mean residence time τ_0 at some interstitial site, jumping to another one with a flight time $\tau_1 \ll \tau_0$. Different experiments[7,8] have shown that this last assumption is not fulfilled by many real materials. Moreover, several studies have suggested that ion-ion correlations are of relevance to ionic transport in superionic conductors (e.g., in β-alumina[9]), as the ions are forced to distribute themselves over a large number of (nonequivalent) sites then forming clusters (sublattice or liquid disorder). Ion-ion and ion-lattice interactions may then introduce correlations between successive hops, making the single-particle hopping picture too crude.[10,11]

Recently it has been shown[12] that a more careful formulation of the random-walk (RW) model could overcome some of its drawbacks. The introduction of an appropriate waiting time density in the continuous-time random-walk (CTRW) theory of Montroll and Weiss[13] allows for considering residence and flight times of arbitrary relative size. In the limit $\tau_1 \ll \tau_0$ the jump-diffusive behavior is recovered, while taking $\tau_1 \gg \tau_0$ leads to the free-diffusion Einstein mechanism. A simplified model of the correlations between jumps can be introduced by formulating a Lorentz-gas model,[14,15] in which the probability of a given jump depends on the previous one. Selection of this correlation allows us to span the full range of behaviors, from the "bounce-back" to the "caterpillar" mechanisms.[10]

One of such models is that of Cáceres,[12] which, in spite of being one-dimensional, agrees fairly well with experimental results for bounce-back-dominated superionic conductors.

In this work we shall construct a similar model for ionic transport in β-alumina, but taking into account the true dimensionality of the material and the structure of the interstitial sites lattice. In Sec. II we use the resolvent-matrix method to find an expression for the generating function of a correlated RW on this lattice, and introduce the residence and flight times through an appropriate waiting time density. In Sec. III we calculate the velocity autocorrelation function using linear response theory and impose the sum rule and Einstein's relation for the static limit to obtain the ac ionic conductivity of the material. In Sec. IV we calculate analytically the incoherent intermediate scattering function from the ionic density-density correlation function. Finally, in Sec. V we discuss briefly the results obtained.

II. THE MODEL

Na β-alumina is composed of parallel oxygen planes with a triangular lattice structure, separated by intervening spinel structure blocks.[16] Na ions move in these planes avoiding the oxygen ions by hopping between interstitial sites which form an hexagonal, two-dimensional lattice.[2] As only half of these sites (the so-called Beevers-Ross sites) are minima of the ionic potential, the effective lattice for the ionic motion is triangular and has the same structure factor as the oxygen one [see Fig.

1(a)]. We take as basis vectors $\hat{\mathbf{e}}_1=\frac{1}{2}\Delta(\sqrt{3}\hat{\mathbf{x}}-\hat{\mathbf{y}})$ and $\hat{\mathbf{e}}_2=\frac{1}{2}\Delta(\sqrt{3}\hat{\mathbf{x}}+\hat{\mathbf{y}})$, where $\Delta=5.595$ Å is the lattice parameter.[17]

For a Na ion hopping on this triangular lattice, correlation effects make the probability of going to a given neighboring site to depend on the site from which the particle comes. We assign it probabilities p for the particle to go right through, q for turning back, $r(s)$ for making a turn of 60° (120°) to the left, and $r'(s')$ for a turn of 60° (120°) to the right [see Fig. 1(b)]. Probability conservation requires $p+q+r+r'+s+s'=1$. Different assignments for these parameters allows for modeling correlations from the caterpillar $(p\simeq 1)$ to the bounce-back $(q\simeq 1)$ mechanisms. For the totally symmetrical case $p=q=r=r'=s=s'$ we recover the uncorrelated (Markovian) RW model, as will be shown in Sec. III.

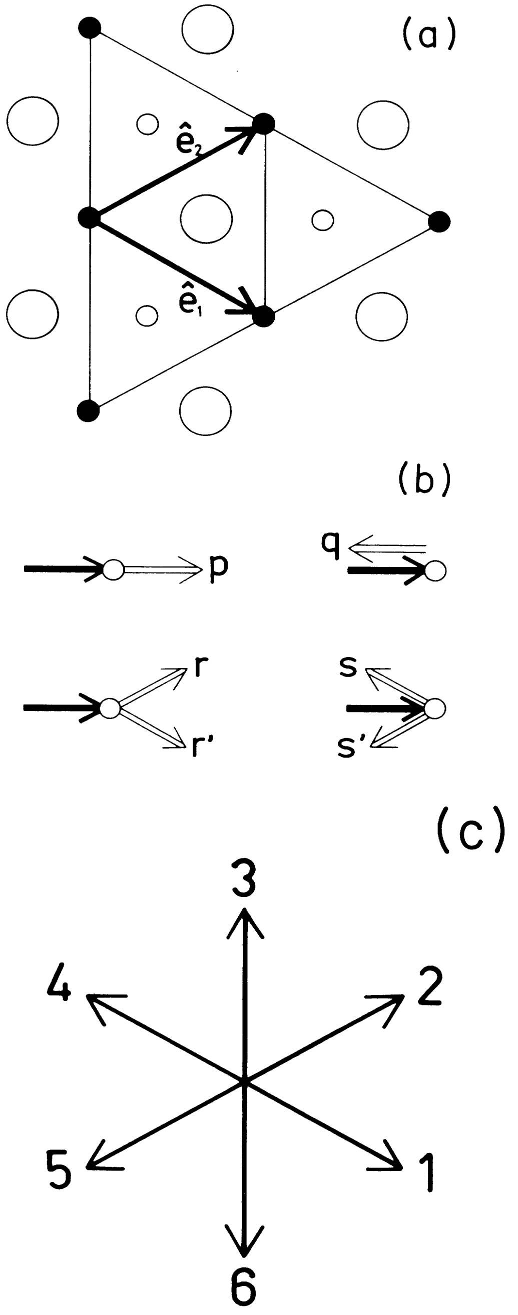

FIG. 1. (a) Interstitial sites lattice structure of Na β-alumina. Large open circles represent oxygen atoms, small circles the interstitial sites (solid circles are Beevers-Ross sites). $\Delta=5.595$ Å. Basis vectors $\hat{\mathbf{e}}_1$ and $\hat{\mathbf{e}}_2$ are shown. (b) Definition of parameters p,q,r,r',s,s'. Solid arrows represent the arrival direction, open ones the outgoing direction. (c) Numbering of the six possible flight directions for a Na ion on an interstitial site.

This model is equivalent to a two-dimensional lattice Lorentz gas.[14] The probability $P_n^i(\mathbf{s})$ $(i=1,\ldots,6)$ for the ion to be at site $\mathbf{s}=s_1\hat{\mathbf{e}}_1+s_2\hat{\mathbf{e}}_2$ (s_1,s_2 integers) in the nth step having come from direction i is

$$P_n^i(\mathbf{s})=\sum_{\mathbf{s}'}\sum_{j=1}^{6}\psi_{ij}(\mathbf{s}-\mathbf{s}')P_{n-1}^j(\mathbf{s}'),\quad i=1,\ldots,6 \tag{1}$$

where $\psi_{ij}(\mathbf{s})$ are the elements of the 6×6 matrix $\Psi(\mathbf{s})$ characterizing the transitions, and we have assumed translational invariance. The flight directions are numbered as shown in Fig. 1(c). Fourier transforming Eq. (1), we get

$$P_n^i(\mathbf{k})=\sum_{j=1}^{6}\psi_{ij}(\mathbf{k})P_{n-1}^j(\mathbf{k})\ , \tag{2}$$

where

$$P_n^j(\mathbf{k})=\sum_{\mathbf{s}}e^{i\mathbf{k}\cdot\mathbf{s}}P_n^j(\mathbf{s})$$

and similarly for $\psi_{ij}(\mathbf{k})$, $\mathbf{k}$ are the vectors of the reciprocal lattice (we keep track of transformations by explicitly writing the arguments of all functions). Defining $\epsilon_1=\exp(i\mathbf{k}\cdot\hat{\mathbf{e}}_1)$, $\epsilon_2=\exp(i\mathbf{k}\cdot\hat{\mathbf{e}}_2)$, and $\epsilon_3=\exp[i\mathbf{k}\cdot(\hat{\mathbf{e}}_2-\hat{\mathbf{e}}_1)]$, the matrix $\Psi(\mathbf{k})$ takes the form

$$\Psi(\mathbf{k})=\begin{bmatrix} p\epsilon_1 & r\epsilon_1 & s\epsilon_1 & q\epsilon_1 & s'\epsilon_1 & r'\epsilon_1 \\ r'\epsilon_2 & p\epsilon_2 & r\epsilon_2 & s\epsilon_2 & q\epsilon_2 & s'\epsilon_2 \\ s'\epsilon_3 & r'\epsilon_3 & p\epsilon_3 & r\epsilon_3 & s\epsilon_3 & q\epsilon_3 \\ q\epsilon_1^* & s'\epsilon_1^* & r'\epsilon_1^* & p\epsilon_1^* & r\epsilon_1^* & s\epsilon_1^* \\ s\epsilon_2^* & q\epsilon_2^* & s'\epsilon_2^* & r'\epsilon_2^* & p\epsilon_2^* & r\epsilon_2^* \\ r\epsilon_3^* & s\epsilon_3^* & q\epsilon_3^* & s'\epsilon_3^* & r'\epsilon_3^* & p\epsilon_3^* \end{bmatrix}, \tag{3}$$

where the asterisk denotes complex conjugation. Equations (1) or (2) must be solved with the appropriate initial conditions $P_0^i(\mathbf{s})$ or $P_0^i(\mathbf{k})$, respectively.

We now define the (marginal) probability to be at $\mathbf{s}$ independently of the arrival direction as

$$P_n(\mathbf{s})=\sum_{j=1}^{6}P_n^j(\mathbf{s})\ . \tag{4}$$

Then the discrete-time-generating function

$$G(\mathbf{k},z)=\sum_{n=0}^{\infty}z^nP_n(\mathbf{k})$$

is given by[18]

$$G(\mathbf{k},z)=\frac{P_0(\mathbf{k})+\sum_{m=1}^{5} z^m\left[P_m(\mathbf{k})-\sum_{n=0}^{m-1} P_n(\mathbf{k})\alpha_{m-n}(\mathbf{k})\right]}{1-\sum_{m=1}^{6} z^m\alpha_m(\mathbf{k})}\ , \tag{5}$$

where the marginal initial conditions $P_m(\mathbf{k})$ $(m=0,\ldots,5)$ are obtained from $P_0^i(\mathbf{k})$ through Eqs. (2) and (4), and the elements $\alpha_j(\mathbf{k})$ of the resolvent matrix are (minus) the coefficients of λ^{6-j} in the characteristic polynomial $p(\lambda)=\det(\lambda\mathbb{1}-\Psi)$ of the matrix $\Psi(\mathbf{k})$.

We now introduce the characteristic times τ_0 and τ_1. To this end we use the CTRW formalism,[13] defining a "telegraphic" waiting time density $\phi(t)$ for the jumps which is independent of the flight direction. Its Laplace transform $\phi(u)$ reads[12]

$$\phi(u)=\frac{\nu_1}{\nu_1+(\nu_0+u)u}\ . \tag{6}$$

If we define the probability that the particle remains fixed at s from time 0 to t as $[1-\int_0^t\phi(t')dt']$, it can be shown that the residence time is $\tau_0\approx\nu_0/\nu_1$. On the other hand, by using the associated generalized master equation,[19] the flight time can be written as $\tau_1\approx\nu_1^{-1/2}$.

The Fourier-Laplace-transformed probability density $P(\mathbf{k},u)$ for this CTRW is then[13]

$$P(\mathbf{k},u)=\frac{1}{u}[1-\phi(u)]G(\mathbf{k},\phi(u))\ , \tag{7}$$

which is precisely the ionic density-density correlation function. The incoherent intermediate scattering function $S(\mathbf{k},\omega)$ for this system can be easily obtained from Eq. (7).[20]

III. CALCULATION OF THE ac CONDUCTIVITY $\sigma(\omega)$

The velocity autocorrelation function $D(t)=\langle v(t)v(0)\rangle$ can be calculated from Eq. (7). Using the Kubo-Lax linear response theory[13] we write

$$D(\omega)=\left[\frac{u^2}{2}\langle s^2(u)\rangle\right]_{u=i\omega}\ , \tag{8}$$

where the average runs over the whole lattice. Equation (8) involves also an average over initial conditions in thermodynamic equilibrium, so we must take $P_0^i(\mathrm{s})=\frac{1}{6}$ $(i=1,\ldots,6)$.

We can obtain the relevant averages from the probability density, Eq. (7), as

$$\langle s_i(u)s_j(u)\rangle=-\left[\frac{1}{u}[1-\phi(u)]\frac{\partial G(\mathbf{k},\phi(u))}{\partial k_i\partial k_j}\right]_{\mathbf{k}=0}\ , \tag{9}$$

where $k_i=\mathbf{k}\cdot\hat{\mathbf{e}}_i$, $i=1,2$. Taking $r+s=r'+s'$ to ensure that there are no preferred left or right rotations and using $s^2=s_1^2+s_2^2+s_1s_2$, some lengthy but straightforward (computer) algebra leads from Eqs. (5) and (6) to

$$\left.\frac{\partial G(\mathbf{k},\phi(u))}{\partial k_i\partial k_i}\right|_{\mathbf{k}=0}=-2\left.\frac{\partial G(\mathbf{k},\phi(u))}{\partial k_i\partial k_j}\right|_{\mathbf{k}=0}=\frac{2\Delta^2}{3}\,\frac{A\phi(u)+1}{A\phi(u)-1}\,\frac{\phi(u)}{[1-\phi(u)]^2}\ , \tag{10}$$

where $i,j=1,2$, $i\neq j$, and we have defined the "correlation parameter" $A=p-q+\frac{1}{2}(r-s+r'-s')$. The parameter A can be interpreted as the difference between the outgoing "probability currents" in the forward and backward directions at a given site.

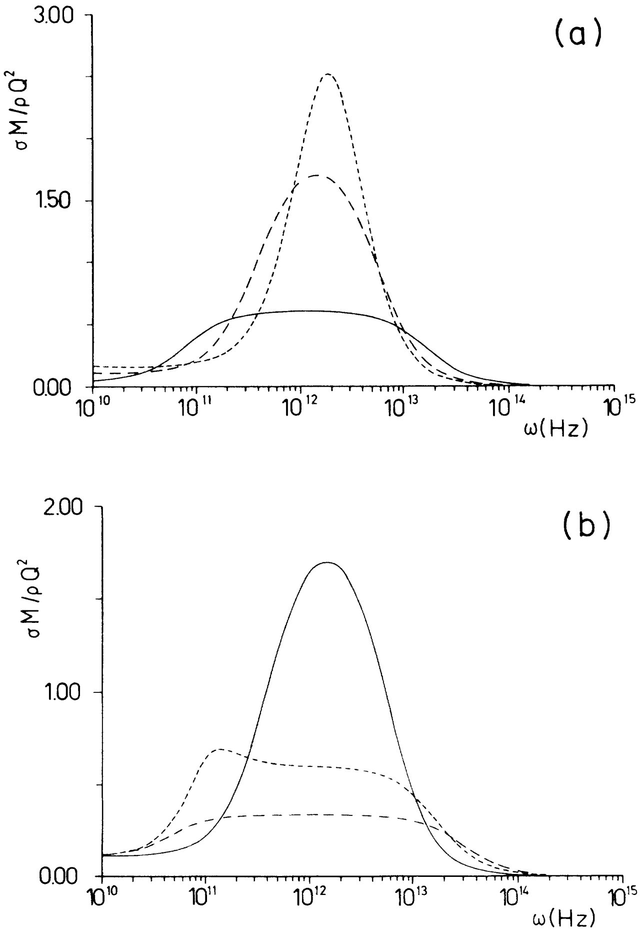

FIG. 2. Calculated ionic conductivity $\sigma(\omega)$ of Na β-alumina. $U=0.10$ eV. (a) No preferred rotations. $A=-0.88$; $T=300$ K (solid line), $T=500$ K (dashed line), $T=800$ K (dotted line). (b) Preferred rotations allowed. $T=500$ K; $A=-0.88$, $B=0$ (solid line), $A=-0.36$, $B=0.45$ (dashed line), $A=0$, $B=0.83$ (dotted line).

Substituting Eqs. (9) and (10) in Eq. (8) leads to

$$D(u)=\frac{\nu_1\Delta^2}{2(\nu_0+u)}\left[\frac{\nu_1(1+A)+(\nu_0+u)u}{\nu_1(1-A)+(\nu_0+u)u}\right] . \tag{11}$$

From Eq. (11) the frequency-dependent correlation function $D(\omega)$ can be obtained by the replacement $u\rightarrow i\omega$. We note that in the totally symmetrical case the correlation parameter vanishes, giving the known result

$$D(u)=\tfrac{1}{2}\nu_1\Delta^2\frac{1}{\nu_0+u} \tag{12}$$

for a Brownian motion with a velocity correlation time $\tau_c=\nu_0^{-1}$.

We now relate the parameters ν_0 and ν_1 to physical quantities. Assuming that the model satisfies the sum rule and Einstein's relation for the static limit, we find

$$\mathrm{Re}D(\omega)|_{\omega=0}=D_0e^{-U/kT} , \tag{13a}$$

$$D(t)|_{t=0}=\frac{kT}{M} , \tag{13b}$$

where $D_0=2\times10^{-4}$ cm^2 s^{-1} is the high-temperature diffusion coefficient, $U\approx0.10$ eV the height of the potential barrier between Beevers-Ross sites, and M the mass of the moving ions.[21] Noticing that $\lim_{t\rightarrow0^+}D(t)=\lim_{u\rightarrow\infty}uD(u)$, a comparison of Eqs. (11) and (13b) gives

$$\nu_1=\frac{2kT}{M\Delta^2} . \tag{14a}$$

Taking $u=i\omega=0$ in Eq. (11) and comparing with Eqs. (13a) and (14a), we find

$$\nu_0=\frac{1+A}{1-A}\frac{kT}{MD_0}e^{U/kT} . \tag{14b}$$

For the general case ($A\neq0$), the ac conductivity of the material is obtained from Eq. (11) as the real part of $\rho Q^2D(u=i\omega)/kT$, i.e.,

$$\sigma(\omega)=\frac{\rho Q^2}{M}\mathrm{Re}\left[\frac{1}{(\nu_0+i\omega)}\times\left[\frac{\nu_1(1+A)+(\nu_0+i\omega)i\omega}{\nu_1(1-A)+(\nu_0+i\omega)i\omega}\right]\right] , \tag{15}$$

where ρ is the (number) density of carrier ions and Q the ionic charge. This result is very similar to that of the one-dimensional model of Ref. 12, partly justifying its simplifications. As in that model we are left with only one free parameter A describing the hopping correlations. Adjustment of this parameter then allows us to span the full range of behaviors from bounce-back ($A<0$) to caterpillar ($A>0$), making the model quite general.

The conductivity given by Eq. (15) is plotted in Fig. 2(a) as a function of frequency for different values of the temperature from 300 to 800 K. The value of A selected corresponds to a bounce-back dominated situation, as that of β-alumina. The ion mass corresponds to Na and we take $U=0.10$ eV.

For completeness, we present the corresponding result for a situation in which preferred left or right rotations are allowed. Taking for simplicity $r=s$ and $r'=s'$, the ac conductivity now results

$$\sigma(\omega)=\frac{\rho Q^2}{M}\mathrm{Re}\left[\frac{1}{(\nu_0+i\omega)}\left[\frac{f_+(\omega)f_-(\omega)-\nu_1^2B^2}{f_-^2(\omega)+\nu_1^2B^2}\right]\right] , \tag{16}$$

where $f_\pm(\omega)=\nu_1(1\pm A)+(\nu_0+i\omega)i\omega$ and we have introduced the "asymmetry parameter" $B=\sqrt{3}(r-r')$, which can be interpreted as the difference between the outgoing probability currents in the left and right directions at a given site. Equation (15) is recovered in the case $B=0$.

We plot in Fig. 2(b) the conductivity given by Eq. (16) for a temperature of 500 K. The values of A and B selected correspond to different situations from bounce-back- to right-turning-dominated ones. The ion mass is that of Na and we take $U=0.10$ eV.

IV. CALCULATION OF THE SCATTERING FUNCTION $S(\mathbf{k},\omega)$

We now proceed to calculate the incoherent intermediate scattering function for this system as[20]

$$S(\mathbf{k},\omega)=\frac{1}{\pi}\mathrm{Re}P(\mathbf{k},u)|_{u=i\omega} . \tag{17}$$

From Eqs. (3)–(5) and using symmetrical initial conditions $P_0^i(\mathbf{s})=\frac{1}{6}$ ($i=1,\ldots,6$) as before, we can explicitly cast the generating function in the form

$$G(\mathbf{k},z)=\frac{\beta_1c_1c_2c_3+\gamma_1(c_1c_2+c_2c_3+c_3c_1)+\delta_1(c_1+c_2+c_3)+\eta_1}{\beta_2c_1c_2c_3+\gamma_2(c_1c_2+c_2c_3+c_3c_1)+\delta_2(c_1+c_2+c_3)+\eta_2} , \tag{18}$$

where $c_1=\cos(\mathbf{k}\cdot\hat{\mathbf{e}}_1)$, $c_2=\cos(\mathbf{k}\cdot\hat{\mathbf{e}}_2)$, and $c_3=\cos[\mathbf{k}\cdot(\hat{\mathbf{e}}_2-\hat{\mathbf{e}}_1)]$ and the parameters β_i, γ_i, δ_i, and η_i ($i=1,2$) are polynomials in z of at most sixth degree. Taking for simplicity $r=r'=s=s'$ (since this does not affect the results in any fundamental way), these parameters are

$$\begin{aligned}
&\beta_1=12\mathcal{A}\mathcal{B}^2z^3 ,\\
&\gamma_1=-4(\mathcal{C}\mathcal{A}^2z^2-\mathcal{A}-\mathcal{B})\mathcal{B}z^2 ,\\
&\delta_1=(\mathcal{C}\mathcal{A}^2z^2+\mathcal{A}+4\mathcal{B})(\mathcal{C}\mathcal{A}z^2+1)z ,\\
&\eta_1=-3(\mathcal{C}\mathcal{A}z^2+1)^2 ,\\
&\beta_2=12(\mathcal{A}+1)\mathcal{B}^2z^3 ,\\
&\gamma_2=-6\{[(p+q)\mathcal{A}+\mathcal{C}]\mathcal{A}z^2+2(p+r)\}\mathcal{B}z^2 ,\\
&\delta_2=3\{[(1-2r)\mathcal{A}+\mathcal{C}]\mathcal{A}z^2+2p\}(\mathcal{C}\mathcal{A}z^2+1)z ,\\
&\eta_2=-3(\mathcal{C}\mathcal{A}z^2+1)^2(\mathcal{A}z^2+1) ,
\end{aligned} \tag{19}$$

where we have defined $\mathcal{A}=p-q$, $\mathcal{B}=p-r$, and $\mathcal{C}=p+q-2r$.

The scattering function is obtained by simply replacing $\phi(u)$ for z in Eq. (19) and substituting Eqs. (7) and (18) in Eq. (17). This procedure is straightforward and gives an analytical expression for $S(\mathbf{k},\omega)$ as a ratio of polynomials in ω with $\mathbf{k}$-dependent coefficients, but the resulting formula is lengthy and we will not give it here. Instead, we plot the final result as a function of frequency in Fig. 3(a) for $\mathbf{k}\cdot\hat{\mathbf{e}}_1=\pi$, $\mathbf{k}\cdot\hat{\mathbf{e}}_2=0$, and some values of the temperature from 300 to 800 K. The values selected for p, q, and r correspond to a bounce-back-dominated situation which gives the same value of A as in Fig. 2(a). In Fig. 3(b) we plot $S(\mathbf{k},\omega)$ for the same values of $\mathbf{k}$ and temperature and a more extreme situation, corresponding to $A=-0.97$. In both cases $B=0$ (no preferred rotations).

V. CONCLUSIONS AND REMARKS

We have used a non-Markovian CTRW approach to model a system with sublattice disorder and noninstantaneous jumps. Our model differs both with the one employed by Huberman *et al.*[21] (in which oscillatory motion of the ions in the lattice potential was used to account for the correlations) and with that of Pietronero *et al.*[22] (in which a generalized master equation with transition rates modulated by additional *dynamical* variables was used for the same purpose).

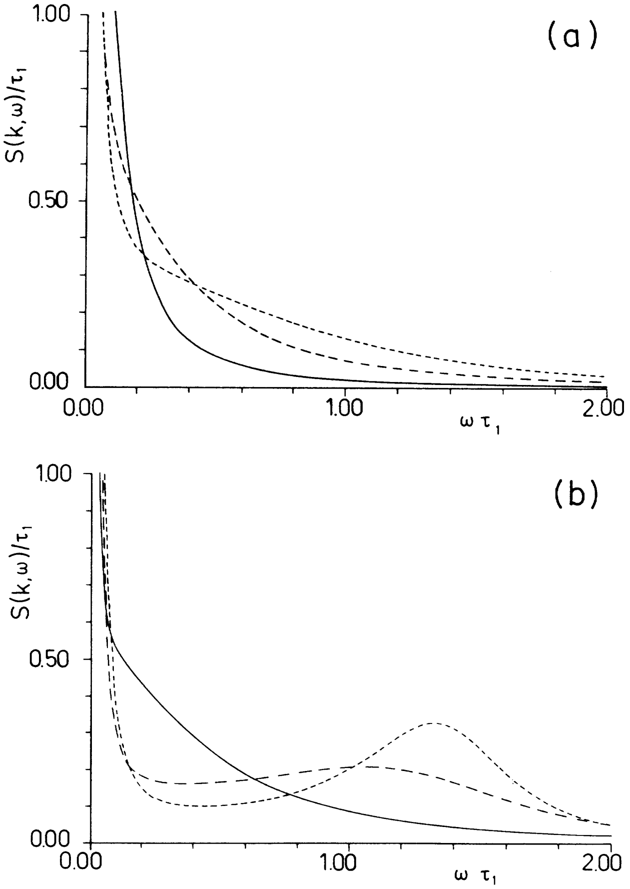

FIG. 3. Calculated incoherent intermediate scattering function $S(\mathbf{k},\omega)$ of Na β-alumina for $\mathbf{k}\cdot\hat{\mathbf{e}}_1=\pi$, $\mathbf{k}\cdot\hat{\mathbf{e}}_2=0$. $U=0.10$ eV; $T=300$ K (solid line), $T=500$ K (dashed line), $T=800$ K (dotted line). (a) $p=r=r'=s=s'=0.02$, $q=0.90$. (b) $p=r=r'=s=s'=0.005$, $q=0.975$.

Figure 2(a) shows that the expression obtained for the ionic conductivity [Eq. (15)] is in good qualitative agreement with experimental results for β-alumina.[1,2] It shows a steadily growing behavior for small ω, a strong departure from Drude-like response to show a peak at microwave frequencies, and it finally vanishes as $1/\omega^2$ for $\omega\rightarrow\infty$ as in free-ion-like models.[23] The peak is centered roughly in the 1-THz region, which corresponds to the inverse of the flight time τ_1 for the corresponding values of the physical parameters ($\tau_1^{-1}\approx v_1^{1/2}\approx 10^{12}\ \mathrm{s}^{-1}$ for Na ions at 500 K), as would be expected.

We want to stress the similarity of this result with that of the one-dimensional model of Ref. 12. Calculations on a two-dimensional square lattice as the one in Ref. 14 gave also the same behavior. This suggests that the detailed lattice structure is relatively immaterial for the ac conductivity, at least as long the lattice is regular and we do not allow for preferred rotations.

It could be argued that a strong magnetic field (external or local) can bias right or left rotations, distorting the resonance peak as shown in Fig. 2(b). However, a simple calculation shows that for the carrier ions to have a mean gyration radius of about 50 lattice spacings, which corresponds to an asymmetry parameter as little as $B=0.02$, the required fields would be roughly 10^3 T, making the observation of these phenomena in Na β-alumina and like materials very difficult.

Figure 3(a) shows that the calculated scattering function $S(\mathbf{k},\omega)$ presents a strong quasielectric line which has a non-Lorentzian line shape. As can be seen from Eqs. (7), (17), and (18), expanding $S(\mathbf{k},\omega)$ in partial fractions, it is a superposition of Lorentzian lines. This corresponds to the results of continuous models as that of Ref. 5 in the large friction limit. In Fig. 3(b) we see that a more extremely bounce-back-dominated situation leads to the appearance of an inelastic peak, as in continuous models with low friction. As expected, $S(\mathbf{k},\omega)$ *does* depend on the lattice structure.

As a final remark we want to stress the conceptual and mathematical simplicity of the model presented here. It allowed us to account for such features as lattice structure, correlation effects, and the existence of two different time scales within a single framework. Introduction of the physical quantities is straightforward, leaving for the conductivity a single free parameter which models the different degrees of sublattice disorder. Calculations are straightforward and can be done analytically. Despite its simplicity, it reproduces with realism the main features of the observed ac response and scattering function.

We think this model can be used with profit for other similar systems, reformulating it in terms of the corresponding lattice structures and transport mechanisms. Work on this line is in progress.

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