Asymmetric exclusion model for mixed ionic conductors

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The ionic conductivity of mixed alkali glasses exhibits a deep minimum as a function of the relative concentrations of the two alkali ions. To study this behaviour we consider a simple one-dimensional model for asymmetric diffusion of two kinds of particles. Different particles are assumed to repulse each other. We consider two versions of the model: with or without overtaking of particles. For the case of perfect repulsion we find exact expressions for the stationary current. The model with weaker repulsion is studied by means of numerical simulations. The stationary current as a function of the ratio of particle concentrations is found to exhibit a minimum, related to correlations existing in this system.

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I. INTRODUCTION

Interesting physical effects have been observed in studying the behaviour of glassy ionic conductors. The conductivity of mixed alkali systems exhibits a strong dependence on the ratio of the alkali concentrations with a deep minimum at a ratio close to 1 \cite{1}. In some experiments the minimum conductivity is a few orders of magnitude smaller than the conductivity of the pure system. An interesting model explaining the anomalous conductivity has recently been introduced in \cite{2} (see also \cite{3}, \cite{4}). The model is based on the assumption that cations in glasses create and maintain their own local environment which, due to memory effects, produce a strong dependence of the conductivity on the relative concentrations of the two ions. Numerical studies of this model have been presented.

Problems of this kind can be understood within the framework of lattice gas models. Recently an extensive effort has been invested in studying particles hopping in a preferred direction with stochastic dynamics and hard core interactions. These are simple examples of non-equilibrium macroscopic systems \cite{5}, \cite{6} which exhibit very interesting collective phenomena such as phase transitions. These models have been used to study hopping conductivity or diffusion in narrow pores \cite{4}, \cite{7}. Furthermore, they are related to growth processes \cite{9}–\cite{11}. Some exact results for particular models are known \cite{8}–\cite{21}.

The aim of this paper is to discuss a lattice gas model for asymmetric particle hopping which is related to the behaviour of mixed ionic conductors. The model describes the dynamics of two kinds of particles moving in the same direction under the influence of a driving field. Here, we do not consider site memory effects but rather assume that there exists a repulsive interaction between the two types of ions. Our model, studied in $d = 1$ dimensions, is simpler than the one introduced in \cite{4}.

We consider two versions of the model. In the first version the two kinds of particles jump independently to the right along a one-dimensional lattice without overtaking each other. In the second version different kinds of particles are allowed to overtake. This may incorporate the effect of the higher space dimension into the one-dimensional model.
II. THE MODEL

To model the dynamics of a system with mixed charge carriers we consider a one-dimensional lattice of length $L$. Each site can take one of three states: it may be occupied by a particle of type $a$ or by a particle of type $b$, or it may be vacant (occupied by a hole $e$). We assume that both kinds of particles contribute to the conductivity of the system. The $a$- and $b$- particles may be identified with different alkali ions. The particles are assumed to undergo an asymmetric exclusion dynamics. The asymmetry is caused by a strong electric field. Throughout this paper we assume periodic boundary conditions.

The total number of $a$- ($b$-) particles is assumed to be equal to $\rho_a L$ ($\rho_b L$). We define $r$ as the ratio of concentration of $a$-particles to the sum of both particle concentration.

$$r = \frac{\rho_a}{\rho} \quad \text{with} \quad \rho = \rho_a + \rho_b . \quad (2.1)$$

To be specific we consider stochastic asymmetric exclusion dynamics with interaction between the $a$- and $b$-particles. Two versions of the model are studied. In the first version no overtaking of $a$- and $b$-particles is allowed. A particle can move to its right if the site on its right is vacant. The probability of making the step during the time interval $dt$ depends on whether an $ab$-bond is broken ($\alpha dt$), created ($\beta dt$), neither broken nor created ($1 dt$) or one bond is broken and another one created ($\alpha \beta dt$). The interaction between the two types of particles is thus introduced by the two parameters $\alpha$ and $\beta$. The possible steps defining the dynamics are given by the following processes:

$$xaex \Rightarrow xeax \quad \text{with rate} \quad 1 \quad (2.2)$$
$$baex \Rightarrow beax \quad \text{with rate} \quad \alpha \quad (2.3)$$
$$xaeb \Rightarrow xeb \quad \text{with rate} \quad \beta \quad (2.4)$$
$$baeb \Rightarrow beab \quad \text{with rate} \quad \alpha \beta \quad (2.5)$$
$$ybey \Rightarrow yeb \quad \text{with rate} \quad 1 \quad (2.6)$$
$$abey \Rightarrow aeb \quad \text{with rate} \quad \alpha \quad (2.7)$$
\[ ybea \Rightarrow yeba \quad \text{with rate } \beta \quad (2.8) \]

\[ aeba \Rightarrow aeba \quad \text{with rate } \alpha \beta \quad (2.9) \]

No other steps are allowed. In these equations the occupation of the four sites \( i-1, i, i+1, i+2 \) is given and the dynamical step takes place between sites \( i \) and \( i+1 \). We denote by \( x \) a site which is occupied by either an \( a \)-particle or by a hole \( e \), and \( y \) represents a site which is either occupied by a \( b \)-particle or by a hole \( e \). The interaction between the two kinds of particles is attractive for \( \alpha < 1 \), \( \beta > 1 \) and is repulsive for \( \alpha > 1 \), \( \beta < 1 \). The other regions of the \( \alpha, \beta \)-plane describe dynamics with competing interactions.

One can look at \( d > 1 \) dimensional systems as described by coupled chains which are directed along the driving field. The coupling between the chains allows for hopping of particles from one chain to another. As a result overtaking of \( a \)- and \( b \)-particles in a single chain can take place by hopping via neighbouring chains. One may therefore model certain aspects of higher dimensional systems by studying a one-dimensional model with overtaking. The rate \( \gamma \) of overtaking events is expected to be low. We thus consider a second version of the model where the following steps, involving overtaking, are permissible in addition to those given by Eqs. (2.2)-(2.9):

\[ ea(e)be \Rightarrow eb(e)ae \quad \text{with rate } \gamma \quad (2.10) \]

\[ aa(e)be \Rightarrow ab(e)ae \quad \text{with rate } \beta \gamma \quad (2.11) \]

\[ ba(e)be \Rightarrow bb(e)ae \quad \text{with rate } \alpha \gamma \quad (2.12) \]

\[ ea(e)ba \Rightarrow eb(e)aa \quad \text{with rate } \alpha \gamma \quad (2.13) \]

\[ aa(e)ba \Rightarrow ab(e)aa \quad \text{with rate } \alpha \beta \gamma \quad (2.14) \]

\[ ba(e)ba \Rightarrow bb(e)aa \quad \text{with rate } \alpha^2 \gamma \quad (2.15) \]

\[ ea(e)bb \Rightarrow eb(e)ab \quad \text{with rate } \beta \gamma \quad (2.16) \]

\[ aa(e)bb \Rightarrow ab(e)ab \quad \text{with rate } \beta^2 \gamma \quad (2.17) \]

\[ ba(e)bb \Rightarrow bb(e)ab \quad \text{with rate } \alpha \beta \gamma . \quad (2.18) \]

Another set of allowed processes are obtained from (2.10)-(2.18) by interchanging \( a \) and \( b \).
The symbol \((e)\) means, there can be a hole between the \(a\)- and the \(b\)- particle, which stays at the same position during the process. Note that the processes with a hole between the particles involve a next-nearest-neighbour interchange.

**III. MEAN FIELD APPROXIMATION**

In this section we discuss the stationary current in mean field approximation (MFA). The mean field approximation gives the same stationary current for both versions of the model. The stationary current of \(a\) particles in MFA can be written

\[
j_a = \rho_a (1 - \rho_b)^2 (1 - \rho) + (\alpha + \beta) \rho_a \rho_b (1 - \rho_b)(1 - \rho) + \alpha \beta \rho_a \rho_b^2 (1 - \rho)
\]  

(3.1)

The expression for the \(b\)-particle current is similar. In terms of the ratio \(r\) introduced in Eq. (2.1) we get for the total current \(j = j_a + j_b\)

\[
j = \rho (1 - \rho) + r (1 - r) \rho^2 (1 - \rho) \{2(\alpha + \beta - 2) + (\alpha \beta - \alpha - \beta + 1) \rho\}
\]  

(3.2)

The current exhibits a minimum in the conductivity if the relation

\[
2(\alpha + \beta - 2) + (\alpha \beta - \alpha - \beta + 1) \rho < 0
\]

(3.3)

is satisfied.

Typical currents \(j\) as a function of \(r\) for different values of \(\rho\) are given in Fig.1. Although the current exhibits a minimum at \(r = 1/2\) (which is most pronounced for \(\alpha = \beta = 0\)), its value at the minimum is rather close to the current of the pure system (\(j(1) = j(0) = \rho(1 - \rho)\)), unlike the experimental results. In the next section we calculate the exact current for \(\beta = 0\) and demonstrate that the current at \(r = 1/2\) can become much lower than its mean field value.

**IV. EXACT RESULTS AND SIMULATIONS**
A. The model without overtaking

We now discuss the first version of the model (rules (2.2) - (2.9)). Let us view the ring as a chain with sites 1, ..., L and denote a configuration by $\underline{n} = \{n_i\}$ where $n_i = a$, $b$ or $e$ if site $i$ is occupied by either an $a$-particle, $b$-particle or a hole, respectively. Furthermore we define a frame $\underline{F}$ as the sequence of $a$- and $b$-symbols which is obtained from a configuration $\underline{n}$ by removing all $e$-symbols. For example the frame associated with the configuration $abebbeaeb...$ is $abbaab...$.

The dynamics of the first version of the model allow for jumps of $a$- or $b$-particles into holes but do not allow for exchange of position of two particles. It may be viewed as the dynamics of holes moving to the left on a lattice certain sites of which are occupied by particles. Consequently, the frame $\underline{F}$ changes in time only by means of cyclic permutations. The latter ones are possible because of the periodic boundary conditions assumed throughout this work. The dynamics are thus nonergodic, and the final state depends on the initial frame $\underline{F}^{(i)}$ defined by the initial configuration $\underline{n}^{(i)}$.

The case $\alpha = \beta = 1$. Although the experimentally observed anomaly is not expected here, a short discussion of this case seems instructive. There is neither repulsion nor attraction between the $a$- and $b$-particles for this choice of the parameters $\alpha$ and $\beta$. The stationary probability distribution $p_{st}(\underline{n})$ for this process is similar to the one for exclusive diffusion of one kind of particle which is well known [20]. It assigns the same probability to all configurations with the initial frame $\underline{F}^{(i)}$ or its cyclic permutations. The stationarity of this distribution can be easily checked by counting the number of incoming and the number of outgoing states for a given configuration. (An incoming state is a configuration which can change to the configuration of interest during one hopping event. Outgoing states are created by a single event taking place in the given configuration.) Note that the form of the stationary distribution depends strongly on the boundary conditions. The above discussion
is correct only for periodic boundary conditions.

Obviously, since there is no interaction between the $a$- and $b$-particles beyond the hard core term, the current $j$ does not depend on the ratio $r$. As in the case of one kind of particles it is exactly the same as calculated in MFA:

$$j(r, \rho) = \rho(1 - \rho) .$$  \hfill (4.1)

The case $\beta = 0$ ($\alpha \neq 0$). This is a nontrivial but exactly soluble case, in which perfect repulsion between the $a$- and $b$-particles takes place. Since $\beta = 0$, no $ab$-bonds (including $ba$-bonds) are created. But since $\alpha \neq 0$ $ab$-bonds may be broken. Hence the system runs into configurations for which the number of $ab$-bonds is minimal. The stationary state is thus characterized by the number $\rho_{bo} L$ of still existing $ab$-bonds.

To study the stationary distribution in detail let us first define the quantity $\rho_{ab}^{(i)}$ as

$$\rho_{ab}^{(i)} = n_{ab}^{(i)}/L$$

where $n_{ab}^{(i)}$ is the number of $ab$-bonds in the initial frame $F^{(i)}$. (Note that due to the periodic boundary conditions we have to include in $n_{ab}^{(i)}$ bonds which may exist between site $L$ and site 1.) The number of $ab$-bonds in the frame does not change in time for the only allowed changes of the frame are cyclic permutations. A configuration, however, has generally less $ab$-bonds than the frame, since holes may be located between the particles of such a bond. In order to allow for a configuration without any $ab$-bond the system needs at least as many holes as number of $ab$-bonds in the frame. Hence, if $1 - \rho \leq \rho_{ab}^{(i)}$, the system runs into a configuration with a density $\rho_{bo} = \rho_{ab}^{(i)} - 1 + \rho$ of $ab$-bonds and all holes are stuck between $a$- and $b$-particles. Since any change of configuration takes place by means of a hopping hole, the configuration the system runs into does not change in time and has a vanishing current. On the other hand, if $1 - \rho > \rho_{ab}^{(i)}$, the number of holes exceeds the number of $ab$-bonds in the frame. The system evolves into configurations with no $ab$-bonds, i.e., where $a$- and $b$-particles are separated by at least one hole. The number of holes free to
hop is given by $n_f = \rho_f L$ with

$$\rho_f = 1 - \rho - \rho_{ab}^{(i)} \quad (4.2)$$

It turns out that for $1 - \rho > \rho_{ab}^{(i)}$ the distribution assigning the same probability to all configurations with no $ab$-bonds and with the frame $F^{(i)}$ or its cyclic permutations is stationary. This can be seen by noting that the number of incoming states for any such configuration is equal to the number of states to which the configuration can evolve.

These simple considerations enable one to derive a general expression for the current $j(r, \rho)$. Let us consider the case of nonvanishing current ($1 - \rho > \rho_{ab}^{(i)}$). A jump between two neighbouring sites, say site 1 and site 2, occurs with rate 1 if the left site is occupied by a particle and the right one by a free hole (a hole which is not stuck between an $a$- and an $b$-particle). Therefore $j(r, \rho)$ equals the probability of finding $ae_f$ or $be_f$ at sites 1 2, where $e_f$ denotes a free hole. It reads

$$j(r, \rho) = \operatorname{prob} (n_1 = a \text{ or } n_1 = b) \operatorname{prob} (n_2 = e_f | n_1 = a \text{ or } n_1 = b). \quad (4.3)$$

Here, the second term in the right hand side of this equation denotes the conditional probability of finding a free hole at site 2 given that site 1 is occupied by a particle. Due to translational invariance one has $\operatorname{prob} (n_1 = a \text{ or } n_1 = b) = \rho$. The probability $\operatorname{prob} (n_2 = e_f | n_1 = a \text{ or } n_1 = b)$ is given by the ratio of the number of configurations which have a free hole at site 2 and a particle at site 1 to the total number of configurations with a particle at site 1. The configurations contributing to these numbers may have different frames. However, all these frames can be obtained from $F^{(i)}$ by cyclic permutation. Thus they all have the same number of $ab$-bonds, and we may restrict ourselves to configurations with the frame $F^{(i)}$. The above probability is given by $\operatorname{prob} (n_2 = e_f | n_1 = a \text{ or } n_1 = b) = X/Y$, where $Y$ is the number of configurations with the frame $F^{(i)}$ and a particle at site 1, and $X$ is the number of those configurations which in addition have a free hole at site 2. All configurations to be counted may be constructed by first inserting a hole in between any pair of $ab$-particles in the frame $F^{(i)}$. One then has to
distribute the remaining \( n_f = \rho_f L \) free holes in between the particles in a way that site 1 is occupied by a particle. Thus \( Y \) is equal to the number of ways of distributing \( n_f \) indistinguishable holes in \( n = n_a + n_b \) states. It is given by

\[
Y = \binom{n+n_f-1}{n_f}^{-1}.
\]

Out of these configurations \( X = Y - \binom{n+n_f-2}{n_f}^{-1} \) have a free hole at site 2. The second term on the right hand side of this equation gives the number of configurations with no free hole at site 2. The current \( j(r, \rho) \) for \( 1 - \rho > \rho_{ab}^{(i)} \) is therefore given by:

\[
j(r, \rho) = \rho \left\{ \binom{n+n_f-1}{n_f} - \binom{n+n_f-2}{n_f} \right\} \binom{n+n_f-1}{n_f}^{-1}.
\]

Simplifying the binomials results in the following expression for the current:

\[
j(r, \rho) = \begin{cases} \frac{\rho \rho_f}{\rho_f + \rho_f - L} & \text{for } 1 - \rho > \rho_{ab}^{(i)} \\ 0 & \text{otherwise} \end{cases}
\]

(4.4)

The density of free holes, \( \rho_f \), is a function of \( \rho \) and of \( \rho_{ab}^{(i)} \), as expressed in (4.2). In the thermodynamic limit the \( L^{-1} \)-term in the expression for the current vanishes.

Consider now the case of random initial conditions in which the initial configuration \( n_{(i)} \) is created by uniformly distributing \( \rho_a L \) a-particles and \( \rho_b L \) b-particles on a lattice of length \( L \). The average number of ab-bonds in the frame \( n_{(i)} \) corresponding to this initial condition is \( 2r(1 - r)\rho L \). For \( L \to \infty \) the relative fluctuations of this quantity vanish. Consequently, we find for any initial configuration with a- and b-particles uniformly scattered on a large lattice:

\[
\rho_{ab}^{(i)} = 2r(1 - r)\rho.
\]

(4.5)

Combining this with Eq. (4.2) and (4.4) we obtain:

\[
j(r, \rho) = \begin{cases} \frac{\rho^{1-\rho^2}1+2r(1-r)}{1-2r(1-r)} & \text{for } \rho < \rho_c(r) \\ 0 & \text{otherwise} \end{cases}
\]

(4.6)

where

\[
\rho_c(r) = 1/[1 + 2r(1-r)]
\]

(4.7)

is the critical density. The expressions for the current \( j \) and the critical density \( \rho_c \) are independent of \( \alpha \) (as long as \( \alpha \neq 0 \)). Fig.1 shows the current as a function of \( r \) for different densities \( \rho \). For densities \( \rho < \rho_c(r = 1/2) = 2/3 \) the current is nonzero for any value \( r \).
This result is rather different from the mean field current (3.2). While the latter is a function of $\alpha$ the exact expression for the current $j$ is independent of $\alpha$. Fig. 1 compares the exact result with the mean field current (3.2) for $\alpha = 0$. This value is chosen since it corresponds to the most pronounced minimum of the conductivity in mean field approximation. The discrepancy between mean field and exact results indicates that the deep minimum is related to correlations.

_The case $\beta > 0$ ($\alpha \neq 0$)_ . For nonvanishing $\beta$ the stationary distribution is more difficult to calculate and one has to resort to numerical simulations of the model. However, a few properties of the $j(r, \rho)$-function can be seen easily. For $r = 0$ and $r = 1$ the current is equal to the mean field current given in Eq. (4.1) since in both cases only one kind of particles performs exclusive diffusion. The stationary solution for that process is known to be uncorrelated [5]. Furthermore the $j(r)$-function is symmetric with respect to reflection about the $r = 1/2$-line because the model is defined in way that a- and b-particles play the same role.

The stationary current for $\beta > 0$ does not vanish for high densities, as in the case $\beta = 0$ since there are always possible hopping events. Consequently, we expect a qualitatively different behaviour for $\rho > \rho_c$ as compared to the $\beta = 0$-case. Replacing $\beta = 0$ by a finite $\beta$ the current is increased for any value of $\rho$ and $r$.

Computer simulations of the process were performed by letting particles hop stochastically on a lattice of $L = 1000$ sites. Averages are calculated as time averages for a particular realization as is done in experiments. Besides, time averages and ensemble averages coincide if the stationary probability distribution is chosen properly, i.e. in the subspace of the phase space which is actually reached by the system.

We have carried out simulations for the case $0 < \beta << 1$ and $\rho = 0.7 > \rho_c$ which seems to reflect the experimental results for the current [1] quite well. The parameter $\alpha$ is chosen to be 1 but it is supposed to play a minor role as long as $\alpha \neq 0$. Results are shown in
Fig. 2. Tuning $\beta$, which depends on the temperature via $\beta = \exp(-\delta E/k_B T)$, any ratio $j(r = 0)/j(r = 0.5)$ can be obtained for $\rho > \rho_c$. Here $\delta E$ is the energy barrier one has to overcome by creating an $ab$-bond. The model is therefore capable of exhibiting the deep minimum of the conductivity which is observed experimentally.

B. The model with overtaking

We next study the second version of the model (rules (2.2)-(2.18)) where $\gamma \neq 0$. Since overtaking is allowed in these dynamics, the frame undergoes noncyclic permutations of particles unlike in the $\gamma = 0$ case.

Let us discuss the case of perfect repulsion ($\beta = 0$) in detail. By simple state-counting it can be shown that a distribution assigning the same probability to any occurring configuration is stationary. But while only configurations with the initial frame $F^{(i)}$ and its cyclic permutations are allowed for the first version of the model, here there is a broader distribution of frames which are reached by the dynamics. As in the previous version, for sufficiently large density $\rho$ the model is expected to run into a configuration in which all holes are stuck between $a$- and $b$-particles. This state has a vanishing current. On the other hand, for densities lower than some critical density $\rho_c(r)$ the number of holes exceeds the number of $ab$-bonds in the initial frame. The system therefore has free holes which generate nonvanishing current.

For densities satisfying $\rho \leq \rho_c(r)$ rules (2.10)-(2.18) allow for changes in the frame via interchanges of $a$- and $b$-particles. Any frame with a number of bonds less than the number of holes can be created during the dynamics. The stationary probability distribution $p_{st}$ assigns the same probability to any configuration without $ab$-bonds.

To calculate the current associated with the stationary distribution we first consider the current averaged over all configurations whose frames have $n_{ab} = \rho_{ab} L$ $ab$-bonds. As it was
shown in Section IV A the current averaged over all configurations with $\rho$ and $\rho_{ab}$ is given by

$$j(\rho, \rho_{ab}) = \frac{\rho \rho_f}{\rho + \rho_f - L^{-1}}$$  \hspace{1cm} (4.8)$$

where

$$\rho_f = 1 - \rho - \rho_{ab}$$  \hspace{1cm} (4.9)$$
is the density of the free holes. The steady state current is thus obtained by averaging $j(\rho, \rho_{ab})$ over all possible $\rho_{ab}$ or $n_{ab}$. Since all allowed configurations have the same weight one has to find the number of configurations corresponding to $n_{ab}$. To this end we consider first the number of frames associated $n_{ab}$. We then calculate the number of ways of distributing $n_f = \rho_f L$ free holes on a given frame, and obtain the probability of having a frame with $n_{ab}$ $ab$-bonds. Note that, as explained at the beginning of Section IV A, a frame is defined by taking a configuration $\underline{n} = \{n_1, ..., n_L\}$ and removing all holes. This defines a sequence of $A$ intervals of $a$-particles alternating with $B$ intervals of $b$-particles. The first and the last interval may either be of the same or of different type. If the intervals at both ends of the lattice are the same, say $a$, type then $A = n_{ab}/2 + 1$ and $B = n_{ab}/2$. Similarly if both end intervals are of $b$-type then $A = n_{ab}/2$ and $B = n_{ab}/2 + 1$. On the other hand if the two end intervals are of different types then $A = B = n_{ab}/2$. The number of ways of arranging $n_a = r \rho L$ $a$-particles in $A$ groups is given by $\binom{n_a - 1}{A - 1}$, and similarly the number of possibilities of arranging the $n_b = (1 - r) \rho L$ $b$-particles in $B$ groups is $\binom{n_b - 1}{B - 1}$. Therefore in each of the above cases the number of possible frames is $\binom{n_a - 1}{A - 1} \binom{n_b - 1}{B - 1}$. We now have to find the number of ways of distributing $n_f = \rho_f L$ free holes in each frame. In the case where both ends of the frame are of the same type this number is given by $\binom{L - n_{ab}}{n_f}$ while in the case where the two ends are different it is given by $[\binom{L - n_{ab}}{n_f} + \binom{L - n_{ab} - 1}{n_f}]$. Thus the statistical weight associated with $n_{ab}$ takes the form
\[ f(n_{ab}) = \left[ \begin{array}{c} n_{ab} - 1 \\ n_{ab}/2 \\ \end{array} \right] \left[ \begin{array}{c} n_{a} - 1 \\ n_{ab}/2 - 1 \\ \end{array} \right] + \left[ \begin{array}{c} n_{ab} - 1 \\ n_{ab}/2 - 1 \\ \end{array} \right] \left[ \begin{array}{c} n_{b} - 1 \\ n_{ab}/2 \\ \end{array} \right] \left( L - n_{ab} \right) \frac{n_f}{n_f} + 2 \left[ \begin{array}{c} n_{ab} - 1 \\ n_{ab}/2 - 1 \\ \end{array} \right] \left[ \begin{array}{c} n_{a} - 1 \\ n_{ab}/2 - 1 \\ \end{array} \right] \left( L - n_{ab} \right) \frac{n_f}{n_f} \]

\[ \frac{2L}{n_{ab}} \left( \frac{L - n_{ab} - 1}{(1 - \rho)L - n_{ab}} \right) \left( \frac{(1 - \rho)L - 1}{n_{ab}/2 - 1} \right) \left( (1 - \rho)\rho L - 1 \right) \frac{n_{ab}/2 - 1}{n_{ab}/2} \]

Equations (4.10)-(4.11) give the stationary current for a lattice of arbitrary length \( L \). In the thermodynamic limit the weight function \( f(n_{ab}) \) is sharply peaked around \( n_{ab} = \rho_{ab} L \) where \( \rho_{ab} \) is a solution of the following equation:

\[ 0 = r(1 - r)\rho(1 - \rho) - \frac{1}{2}(1 - \rho)[1 - \rho + 4r(1 - \rho)\rho] \rho_{ab}^* + \left[ \frac{1}{2} - \frac{3}{4} \rho + r(1 - \rho)\rho \right] (\rho_{ab}^*)^2 \]  

(4.12)

This yields \( \rho_{ab}^* \) is a function of \( \rho \) and \( r(1 - r) \) which has its maximum at \( r = 1/2 \) for any given value of \( \rho \). For example, solving Eq.(4.12) for \( r = 1/2 \) we find

\[ \rho_{ab}^*(r = \frac{1}{2}, \rho) = \frac{1}{2} - \frac{1}{2} \sqrt{1 - 2\rho(1 - \rho)} \]  

(4.13)

Hence, for an infinite lattice the number of bonds in the frame is \( \rho_{ab}^* L \) and the stationary current is

\[ j(r, \rho) = \begin{cases} \rho Z^{-1} \sum_{k=0}^{\infty} \frac{1 - \rho - 2kL^{-1}}{1 - 2k + 1} f(2k) & \text{for } \rho < \rho_c(r) \\ 0 & \text{otherwise} \end{cases} \]  

(4.11)

Let us now consider the random initial conditions defined in Section IV A and try to estimate the critical density \( \rho_c(r) \) below which the current of the system is nonzero. Clearly, if the concentration \( \rho_{ab}^{(i)} = 2r(1 - r)\rho \) of \( ab \)-bonds in the initial frame \( F^{(i)} \) satisfies

\[ \rho_{ab}^{(i)} = \rho_{ab}^*(r, \rho) \]  

(4.14)
\( \rho^{(i)}_{ab} < 1 - \rho \), the system has free holes which generate ergodic dynamics leading to a current \( j \) as given by Eqs. (4.13) and (4.14). This yields a first lower bound for the critical density \( \rho_c(r) > 1/[1 + 2r(1 - r)] \). However, due to the particle exchange mechanism which exists for \( \gamma > 0 \) the system may exhibit a finite current even for \( \rho^{(i)}_{ab} \gtrsim 1 - \rho \). This may easily be seen by considering the limit \( \gamma \ll 1 \). In this limit and for time scales shorter than \( 1/\gamma \), basically all initially free holes are caught by \( ab \)-bonds, and thus stop moving. However for longer time scales, where particle exchange processes take place, some of the holes which are stuck in \( ab \)-bonds are released. For example a sequence \( abab \) may evolve into \( aabb \) thus reducing the number of \( ab \)-bonds by two and creating free holes. And a sequence \( aabbaabb \) may in principle evolve into \( aaaaabbb \), but this requires the existence of free holes to begin with. If the only holes in this sequence are those stuck in the \( ab \)-bonds, particle exchange processes do not take place. However, if in addition there are some free holes, the sequence may change. If we take into account only changes which do not require the existence of free holes the number of \( ab \)-bonds in the frame is reduced and becomes \( \tilde{\rho}_{ab} L \) with

\[
\tilde{\rho}_{ab} = \rho^{(i)}_{ab} - [4r^2(1 - r)^2 + o(r^3(1 - r)^3)] \rho .
\]

(4.15)

We thus expect that for \( \tilde{\rho}_{ab} < 1 - \rho \) the system exhibits a nonvanishing current. This yields the following lower bound for \( \rho_c(r) \):

\[
\rho_c(r) \gtrsim 1/[1 + 2r(1 - r) - 4r^2(1 - r)^2] + o(r^3(1 - r)^3) .
\]

(4.16)

The current \( j(r, \rho) \) is shown in Fig.3 as a function of \( r \) for various values of \( \rho \). In the figure \( \rho_c(r) \) is determined by Eq. (4.10) to second order in \( r(1 - r) \).

The dynamics of the hopping holes have a time scale of order 1 whereas the reordering of the frame has a characteristic time much larger than one since we consider the case \( \gamma \ll 1 \). Hence we observe the following scenario: The system runs into the stationary state of the first version of the model (see Fig. 1) which decays very slowly to the final state (Fig. 3).
V. CONCLUSION

A simple model describing the transport properties of mixed ionic conductors has been introduced and analyzed. The model exhibits a minimum conductivity for equal concentrations of the two species of particles \( r = 1/2 \), a result which is compatible with experimental observations in mixed alkali glasses. A mean field approximation yields the correct qualitative behaviour of the conductivity, but it fails to explain the very low conductivity for \( r = 1/2 \). It has been demonstrated by exact solution and numerical simulations in the strong repulsion limit that the conductivity corresponding to the model is indeed very small at \( r = 1/2 \) in accordance with experimental observations.

The model can be extended to study the case in which particles move into both directions with arbitrary rates. The exact results obtained above for the case \( \beta = 0 \) are easy to generalize: The probability distribution which assigns the same probability to all configurations with no bonds is stationary even when hops in both directions take place. Moreover, the rates \( p \) and \( q \) of hopping to the right and left, respectively, may be taken as time dependent without changing the stationary state. The current is then \([p(t) - q(t)]j\), where \( j \) is given by Eq.(4.4) or Eq.(4.6) for \( \gamma = 0 \) and by Eq.(4.11) or Eq.(4.14) when overtaking is included, i.e. for \( \gamma \neq 0 \). Obviously, the conductivity does not depend on the frequency if the driving field is harmonic.

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Captions to the figures

Fig.1: Stationary current \( j \) for the model without overtaking as a function of the ratio \( r \) for \( \beta = 0 \) and different values of the density \( \rho \), solid lines: exact results (\( \alpha \) arbitrary), dashed lines: mean field approximation (\( \alpha = 0 \)); (1): \( \rho = 0.15 \); (2): \( \rho = 2/3 \); (3): \( \rho = 0.8 \)

Fig.2: Stationary current \( j \) for the model without overtaking as a function of the ratio \( r \) for \( \rho = 0.7 \), \( \alpha = 1 \) and different \( \beta \)'s obtained by simulations (The labels are the values of \( \beta \), and the lines are interpolations.)

Fig.3: Stationary current \( j \) for the model with overtaking as a function of the ratio \( r \) for \( \beta = 0 \), arbitrary \( \alpha \) and different values of the density \( \rho \), exact results