Diffusion of particles in an inhomogeneous medium: High-friction limit

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We develop an inverse friction expansion for the diffusion of particles in an inhomogeneous medium based on the Mori formalism starting from a microscopic Hamiltonian. The friction is related to the vibrational correlation function of the substrate. An analytic formula is obtained for the diffusion constant in the high-friction limit.

I. INTRODUCTION

The classical diffusive motion of a particle in an external potential, either random or periodic, is an important problem central to many different areas of physics.[1] For example, the diffusion constant of surface adatoms is a crucial parameter in determining the rate of surface kinetic processes such as thin film growth or catalytic reactions. The diffusive motion in a periodic potential can also be mapped to such diverse problems as transport in superionic conductors and the current voltage characteristic of a Josephson tunneling junction.[2] The theoretical approach to understanding this important problem has been mainly along two lines. The first is a numerical molecular dynamics or Monte Carlo simulation method.[3] The second method is an analytic study based on the Kramers equation which is a special Fokker-Planck equation describing the Brownian motion in a potential.[2,4] The key ingredient in this equation is a frictional damping force in addition to the adiabatic external potential. This frictional force arises from coupling to the electronic or vibrational excitations of the background medium. It is often treated just as a phenomenological parameter. The Kramers equation is sufficiently complicated so that a closed analytic solution for the diffusion constant is still unobtainable. In the high-friction limit, the Kramers equation reduces to the Smoluchowski equation which contains the distribution function for the position coordinates only. For the one-dimensional (1D) case, several different groups[2,5,6] have independently arrived at the analytic solution of the Smoluchowski equation. This solution provides much insight into the nature of diffusion at different temperatures. No comparable solutions have been obtained for motion in higher dimensions.

In this paper, starting from a microscopic lattice-dynamic Hamiltonian, we use the Mori projection operator formalism[7] to derive a set of equations for different correlation functions which is equivalent to the Kramers equation. In the process, an analytic expression for the frictional damping force is also derived, relating it to the vibrational properties of the background medium. We then develop a continued fraction expansion for the velocity-velocity correlation function. This is actually an expansion in inverse powers of the frictional damping force. In the high-friction limit, we are able to obtain an analytic solution for arbitrary dimensions which reduces

to the well known solution in the 1D case. Finally, we examine the solution for the 2D case in some detail particularly for a periodic potential in view of its importance for understanding surface adatom diffusion.

II. GENERAL FORMULATION

We start from a basic Hamiltonian describing the motion of a particle interacting with N background particles vibrating about their equilibrium positions. The interaction of the tagged particle with the background provides both the inhomogeneous adiabatic potential as well as the damping force,

$$H = H_s = \frac{\mathbf{P}^2}{2m} + V[\mathbf{R}, \{\mathbf{R}_l\}] , \tag{1}$$

where

$$H_s = \sum_l \frac{\mathbf{P}_l^2}{2M} + W[\{\mathbf{R}_l\}] .$$

The first term H_s in (1) describes the kinetic and potential energy of the background. The second and third correspond to the kinetic energy of the tagged particle and its interaction with the background. At this stage all the interaction can be completely general and need not be restricted to the usual pairwise-interaction approximation.

We now introduce the projection operators. First, a projection operator $\mathcal{P}_b$ is defined. When operated on an arbitrary variable A, $\mathcal{P}_b$ corresponds to taking the partial thermal average over the background degrees of freedom, i.e.,

$$\mathcal{P}_b A = Z_b^{-1} \Pi_l \int d\mathbf{P}_l d\mathbf{R}_l e^{-\beta H} A . \tag{2}$$

Here $Z_b = \Pi_l \int d\mathbf{P}_l d\mathbf{R}_l e^{-\beta H}$. It is also useful to introduce an adiabatic potential $V_A(\mathbf{R})$ defined in the following way:

$$\exp[-\beta V_A(\mathbf{R})]$$
$$= Z_s^{-1} \Pi_l \int d\mathbf{P}_l d\mathbf{R}_l \exp[-\beta(H_s + V[\mathbf{R}, \{\mathbf{R}_l\}])] , \tag{3}$$

with $Z_s = \Pi_l \int d\mathbf{P}_l d\mathbf{R}_l \exp(-\beta H_s)$. This adiabatic potential represents a potential experienced by the diffusing particle at point $\mathbf{R}$ averaged over the background vibra-

tional degrees of freedom. The density function $n(\mathbf{R})$ for the tagged particle can be expressed as

$$n(\mathbf{R})=\left[\int e^{-\beta V_A(\mathbf{R})}d\mathbf{R}\right]^{-1}\exp[-\beta V_A(\mathbf{R})] \ . \qquad (4)$$

According to these definitions, a thermal average of a quantity involving only the coordinates of the tagged particle can be expressed in terms of $n(\mathbf{R})$ as

$$\langle f(\mathbf{R})\rangle = \int n(\mathbf{R})f(\mathbf{R})d\mathbf{R} \ .$$

Next, we introduce a set of orthogonal variables $\{A_n\}$ and their corresponding projection operators $\{\mathcal{P}_n\}$ of the diffusing particle:

$$\mathbf{A}_0=\{e^{i\mathbf{k}\cdot\mathbf{R}}\} \ ,$$

$$\mathbf{A}_1=\left\{\frac{\mathbf{P}}{m}e^{i\mathbf{k}\cdot\mathbf{R}}\right\} \ ,$$

$$\cdots \qquad\qquad\qquad\qquad\qquad (5)$$

$$\mathbf{A}_n=(1-\mathcal{P}_{n-1}-\mathcal{P}_{n-2})\mathcal{P}_b\dot{\mathbf{A}}_{n-1} \ .$$

In (5), the projection operators are defined as

$$\mathcal{P}_n\mathbf{A}=\mathbf{A}_n\chi_{nn}^{-1}(\mathbf{A}_n,\mathbf{A}) \ , \qquad (6)$$

with

$$\chi_{nn}=(\mathbf{A}_n,\mathbf{A}_n) \ ,$$

where the scalar product (A,B) is defined as the thermal average of A^*B. It is easy to verify from these definitions that the A's are orthogonal to each other. Note that in our definition, each basic variable A_n has actually an infinite number of components corresponding to different wave vectors and Cartesian coordinates of the momentum vector. At this point, it is also useful to introduce the Liouville operator L defined as

$$iL\,\mathbf{A}=\frac{d\,\mathbf{A}}{dt} \ . \qquad (7)$$

We now define the various time-dependent correlation functions $S(t)$ as

$$S_{nn'}(t)=(\mathbf{A}_n(t),\mathbf{A}_{n'}(0)) \ , \qquad (8)$$

and the Laplace transform of these correlation functions are given as

$$S_{nn'}(z)=\int_0^\infty e^{izt}S_{nn'}(t)dt$$

$$=\left(\mathbf{A}_n,\frac{i}{z-iL}\mathbf{A}_{n'}\right) \ . \qquad (9)$$

These correlation functions contain all the relevant information about the system under study. For example, the diffusion tensor is the zero frequency limit of the velocity-velocity self-correlation function with wave vector $k=0$ and $k'=0$:

$$D_{\alpha\beta}=\int_0^\infty\langle v_\alpha(t)v_\beta\rangle dt$$

$$=\lim_{z\to 0}S_{11}^{\alpha\beta}(z;\mathbf{k}=0,\mathbf{k}'=0) \ . \qquad (10)$$

For the special case of a periodic background, instead of the general set of wave vectors, only the relevant reciprocal lattice vectors need to be considered.

The Mori formalism now allows us to develop an equation of motion for these correlation functions.[8,9] The resulting equation is

$$[-iz\chi^{-1}+b+\Sigma(z)]S(z)=1 \ , \qquad (11)$$

with

$$b=i\chi^{-1}(\mathbf{A},L\,\mathbf{A})\chi^{-1} \ ,$$

and

$$\Sigma(z)=\chi^{-1}\left[QL\,\mathbf{A},\frac{i}{z-QLQ}QL\,\mathbf{A}\right]\chi^{-1} \ .$$

In this equation, Q is the projection operator into the space orthogonal to that spanned by the basic variables $\{A_n\}$. That is,

$$Q=1-\sum_{i=0}^\infty\mathcal{P}_i \ . \qquad (12)$$

The set of equations in (11) is equivalent to the Kramers equation. However, since we start from a lattice-dynamic Hamiltonian, there is an additional bonus here in the fact that instead of a phenomenological damping constant, we have a memory function which contains the damping effects. As we shall see in the next section, the memory function can be related to the vibrational properties of the background.

III. MEMORY FUNCTION

The memory function contains the details of the coupling of the adatom motion to the background vibration. In this section we examine the most important component—the $(1,1)$ element of the memory function Σ in detail. From (11), we have

$$\Sigma_{11}(z)=\frac{k_BT}{m}\chi_{11}^{-1}\gamma(z)\chi_{11}^{-1} \ , \qquad (13)$$

with

$$\gamma(z)=\frac{1}{mk_BT}\int_0^\infty e^{izt}dt\,(QL\,\mathbf{P}e^{i\mathbf{k}\cdot\mathbf{R}},e^{-iQLQt}QL\,\mathbf{P}e^{i\mathbf{k}'\cdot\mathbf{R}}) \ . \qquad (14)$$

It is easy to see that to lowest order in the displacement of the background particles, we have

$$QL\,\mathbf{P}e^{i\mathbf{k}\cdot\mathbf{R}}=-(1-\mathcal{P}_b)\frac{\partial V}{\partial\mathbf{R}}e^{i\mathbf{k}\cdot\mathbf{R}} \ . \qquad (15)$$

The $k=0$ component of (15) corresponds simply to the frictional force on the tagged particle due to the vibrational motion of the background. The projection operator Q serves to separate out the regular force on the tagged particle due to the interaction with the background at the equilibrium position. There is one limiting case when γ can be simplified considerably. This is when the time scale of the diffusive motion is much longer than

the time scale for the vibrational motion of the background. Under this circumstance, the time dependence of the coordinates of the diffusing particle can be neglected in the memory function and replaced by its initial value. This is the initial value approximation (VIA).[4] In this approximation, γ decouples into the product of averages involving the tagged particle and the background particles separately.

To simplify the memory function further, we now consider the case of pairwise interaction and a harmonic approximation for the background vibrational motion. In this case the adiabatic potential $V_A(\mathbf{R})$ is given by the expression

$$V_A(\mathbf{R}) = \sum_l \int d\mathbf{r}\, v(\mathbf{R} - \mathbf{R}_l^0 - \mathbf{r}) W_l(\mathbf{r}) , \tag{16}$$

where the Debye-Waller factor

$$W_l(\gamma) = (8\pi^3 U_l^{\|2} U_l^{\perp})^{-1/2} \exp\left[-\frac{1}{2}\left[\frac{x^2+y^2}{U_l^{\|}} + \frac{z^2}{U_l^{\perp}} \right] \right] \tag{17}$$

has been introduced. Note that Eq. (16) is only valid when the equilibrium position $\{\mathbf{R}_l^0\}$ is assumed to be independent of $\mathbf{R}$, i.e., when small relaxation around the diffusing particle is neglected. The expression for γ can then be simplified to

$$\gamma_{\alpha\beta}(z) = \frac{1}{mk_B T}\sum_{ll'} C_{ll'}^{\gamma\delta}(z)\int n(\mathbf{R}) d\mathbf{R}\, v_{\text{eff}}^{\alpha\beta}(\mathbf{R} - \mathbf{R}_l^0)$$

$$\times v_{\text{eff}}^{\gamma\delta}(\mathbf{R} - \mathbf{R}_{l'}^0)e^{-i(\mathbf{k}-\mathbf{k}')\mathbf{R}} , \tag{18}$$

where

$$v_{\text{eff}}^{\alpha\beta}(\mathbf{R} - \mathbf{R}_l^0) = \int d\mathbf{r}\nabla_R^\alpha \nabla_R^\beta v(\mathbf{R} - \mathbf{R}_l^0 - \mathbf{r}) W_l(\mathbf{r}) ; \tag{19}$$

here

$$C_{ll'}^{\gamma\delta}(z) = \int_0^\infty e^{izt} dt(u_{l\gamma}, e^{-iQLQt}u_{l'\delta}) . \tag{20}$$

In the expression in (20), the time dependence of the background particles is still governed by the complicated Liouville operator QLQ. In the weak coupling approximation, we can neglect the influence of the diffusing particle on the vibrational motion of the background. This amounts to replacing the operator QLQ by the simpler operator L describing the dynamics of the background alone.

Further progress can be made by noting that

$$\Sigma_{11}(z) = \frac{k_B T}{m}\chi_{11}^{-1}\gamma(z)\chi_{11}^{-1}$$

$$= \frac{m}{k_B T}\chi_{00}^{-1}\gamma(z)\chi_{00}^{-1} . \tag{21}$$

Now, according to (4), $\chi_{00}(\mathbf{k},\mathbf{k}')$ is just given as

$$\chi_{00}(\mathbf{k},\mathbf{k}') = \langle e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} \rangle$$

$$= \int n(\mathbf{R})e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} d\mathbf{R} . \tag{22}$$

The inverse of the infinite matrix χ_{00} is easily done first in real space and then transformed back into k space. This gives

$$\chi_{00}^{-1}(\mathbf{k},\mathbf{k}') = \frac{1}{L^{2d}}\int n^{-1}(\mathbf{R})e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} d\mathbf{R} . \tag{23}$$

In (23), L is the linear dimension of the whole system and we are imposing a periodic boundary condition. For the study of diffusion, eventually we shall take the limit of $L \to \infty$ before taking the limit $z \to 0$. Combining (18), (21), and (22), we obtain

$$\Sigma_{11}^{\alpha\beta}(z;\mathbf{k},\mathbf{k}') = \frac{m}{L^{2d}k_B T}\int n^{-1}(\mathbf{R})\eta_{\alpha\beta}(\mathbf{R},z)e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} d\mathbf{R} , \tag{24}$$

where

$$\eta_{\alpha\beta}(\mathbf{R},z) = \frac{1}{mk_B T}\sum_{\substack{ll' \\ \gamma\delta}} C_{ll'}^{\gamma\delta}(z)v_{\text{eff}}^{\alpha\gamma}(\mathbf{R}-\mathbf{R}_l^0)v_{\text{eff}}^{\beta\delta}(\mathbf{R}-\mathbf{R}_{l'}^0) . \tag{25}$$

Finally, it has also been shown that within the present approximation,[4] the memory function takes the diagonal form $\Sigma_{nn'} = \delta_{nn'}\Sigma_n$. We shall not look at the higher components Σ_n with $n > 1$ in detail since they are not needed in the high-friction limit.

IV. INVERSE FRICTION EXPANSION

Starting from the general set of equations (11) for the correlation functions $S(z)$, we can now develop a continued fraction expansion for $S_{11}(z)$ which is directly related to the diffusion constant. As we shall see below, this expression is actually an expansion in inverse powers of the friction. The leading term corresponding to the high-friction limit has a very simple form. Let us define

$$-iz\chi_{ii}^{-1} + \Sigma_{ii} = a_i(z) . \tag{26}$$

Then from (11), we obtain the set of equations

$$a_0 S_{01}(z) + b_{01}S_{11}(z) = 0 ,$$

$$b_{10}S_{01}(z) + a_1 S_{11}(z) + b_{12}S_{21}(z) = 1 ,$$

$$\vdots \tag{27}$$

$$b_{n,n-1}S_{n-1,1}(z) + a_n S_{n1}(z) + b_{n,n+1}S_{n+1,1}(z) = 0$$

$$\text{for } n > 1 .$$

Now we introduce the functions $B_n(z)$ defined as

$$S_{n1}(z) = B_{n-1}S_{n-1,1}(z) . \tag{28}$$

Then we have from (26),

$$S_{11}(z) = [a_1 - b_{10}a_0^{-1}b_{01} + b_{12}B_1(z)]^{-1} , \tag{29}$$

and for $n > 1$,

$$B_{n-1}(z) = -[a_n + b_{n,n+1}B_n(z)]^{-1}b_{n,n-1} . \tag{30}$$

Equations (29) and (30) together constitute a continued

fraction expansion for the correlation function $S_{11}(z)$. Note that for calculating the diffusion constant, we need to take the limit $z \to 0$. In this case, the a's are proportional to the friction memory function $\Sigma_{ii}(z)$. Therefore, the continued fraction expansion (29) and (30) is equivalent to an inverse friction expansion. In the high-friction limit, we can drop all the $B_n(z)$ in the continued fraction expansion for $n > 1$; the result for $S_{11}(z)$ is then the simple expression

$$S_{11}(z) = [a_1(z) - b_{10}a_0^{-1}(z)b_{01}]^{-1} . \tag{31}$$

Equation (31) is a formal solution in the high-friction limit, so it is equivalent to a solution of the corresponding Smoluchowski equation. The added bonus of the present method is that we also have a microscopic expression for the frictional force.

V. ANALYTIC SOLUTION FOR THE DIFFUSION CONSTANT IN A PERIODIC POTENTIAL

The simplicity of the formal high-friction solution (31) for $S_{11}(z)$ is somewhat misleading. The matrices $a(z)$ and b appearing in (31) are all of infinite dimensions. The row or column labels correspond to the infinite set of wave vectors k as well as the Cartesian coordinate indices. The usual approach here is to take a finite set of k vectors, invert the matrix, and then test the convergence for a larger set of $\mathbf{k}$ vectors.[4,9] However, as we show in this section, for special cases it is possible to perform the infinite matrix inversion in (31) exactly and obtain a closed analytic expression for the diffusion constant.

The special case we are interested in here is a particle diffusion in a d-dimensional background potential $V_A(\mathbf{R})$ with hypercubic symmetry. The physically interesting cases are of course diffusion in $d = 1$ (linear), $d = 2$ (square), and $d = 3$ (cubic) lattices. Let a be the basic period along any principal symmetry axis; then instead of the general set of $\mathbf{k}$ vectors, we can simply take the set of reciprocal lattice vectors

$$G_{n_1, \ldots, n_d} = \frac{2\pi}{a}(n_1, n_2, \ldots, n_d) \tag{32}$$

for all integer values of $n_1, n_2, \ldots, n_d$.

The matrix elements in (31) are

$$a_1^{\alpha\beta}(z; \mathbf{G}, \mathbf{G}') = -iz\chi_{11}^{\alpha\beta}(\mathbf{G}, \mathbf{G}') + \Sigma_{11}^{\alpha\beta}(z; \mathbf{G}, \mathbf{G}') , \tag{33}$$

and it is shown in the Appendix that

$$(b_{10}a_0^{-1}b_{01})_{\mathbf{G},\mathbf{G}'}^{\alpha\beta} = \frac{-i}{z}G_\alpha \chi_{00}^{-1}(\mathbf{G}, \mathbf{G}')G_\beta' . \tag{34}$$

It is now easy to see from the general structure of the matrix that

$$\lim_{z \to 0} S_{11}^{\alpha\alpha}(z; \mathbf{G} = 0, \mathbf{G}' = 0)$$

$$= \Sigma_{11}^{-1}(z = 0; \tilde{\mathbf{G}}, \tilde{\mathbf{G}}')|_{\tilde{G} = 0, \tilde{G}' = 0} , \tag{35}$$

where $\tilde{G}, \tilde{G}'$ are now defined in the subspace of all reciprocal lattice vectors orthogonal to the direction $\hat{x}_\alpha$, i.e.,

$$\tilde{\mathbf{G}} \cdot \hat{\mathbf{x}}_\alpha = 0 . \tag{36}$$

This implies that the inverse of Σ_{11} is given as

$$\Sigma_{11_{\alpha\alpha}}^{-1}(z = 0, \tilde{\mathbf{G}}, \tilde{\mathbf{G}}')$$

$$= \frac{k_B T}{m}L^2 \int e^{-i(\tilde{G} - \tilde{G}') \cdot \mathbf{R}_\perp}d\mathbf{R}_\perp$$

$$\times \left[\int dx_\alpha n^{-1}(\mathbf{R})\eta_{\alpha\alpha}(\mathbf{R}, z = 0) \right]^{-1} . \tag{37}$$

The diffusion constant is obtained from the $\tilde{G} = 0$ and $\tilde{G}' = 0$ element of Σ_{11}^{-1}. For the symmetry of our lattice it is obviously independent of the particular direction $\hat{x}_\alpha$ and is given by

$$D_{\alpha\alpha} = \frac{k_B T}{m}L^2 \int d\mathbf{R}_\perp \left[\int dx_\alpha n^{-1}(\mathbf{R})\eta_{\alpha\alpha}(\mathbf{R}, z = 0) \right]^{-1} . \tag{38}$$

Equation (38) is the central result of this paper. It is equivalent to an analytic solution of the Smoluchowski equation with a microscopic friction. The results for the 1D case have been obtained independently by many groups using different techniques.[5] It is remarkable that such a simple analytic expression also exists for arbitrary dimensions.

VI. SURFACE DIFFUSION

In this section, we concentrate on a more detailed discussion of the result (38) for the diffusion constant in the $d = 2$ case which is relevant for studying surface diffusion. For $d = 2$, (38) reduces to

$$D_{xx} = \frac{k_B T L^2}{m} \left[\int e^{-\beta V_A(\mathbf{R})}d\mathbf{R} \right]^{-1}$$

$$\times \int dy \left[\int dx e^{\beta V_A(\mathbf{r})}\eta_{xx}(\mathbf{R}, z = 0) \right]^{-1} . \tag{39}$$

To analyze the behavior of diffusion constant D as a function of temperature, we first look at the somewhat simplistic case where the friction η_{xx} is a constant independent of position. In the first integral over the coordinate x in (39), obviously the region near the maxima of $V_A(x, y)$ as a function of x for a given y gives the dominant contributions at low temperatures. However, in the subsequent integration over y, only the smallest of these maxima as a function of y will give the dominant contribution. Thus the saddle-point region of the potential $V_A(\mathbf{R})$ contributes predominantly to the double integral in (39). In the partition function $\int e^{-\beta V_A(\mathbf{R})}d\mathbf{R}$, regions near the minima of $V_A(\mathbf{R})$ give the dominant contributions. Combining all these considerations, we obtain the following low temperature expansion for the diffusion constant:

$$D = \frac{k_B T}{m\eta}e^{-\beta\Delta} . \tag{40}$$

Here Δ is the barrier between the saddle point and the

minimum of the potential $V_A(\mathbf{R})$. Equation (40) displays the familiar Arrhenius behavior of activated diffusion often assumed in a phenomenological analysis. To our knowledge, this is the first time that such as Arrhenius form has been derived analytically starting from a microscopic Hamiltonian. For the more general case where the friction is position dependent, obviously the result will be more complicated. It is clear from the above discussions that the value of the friction at the saddle point of $V_A(\mathbf{R})$ plays the most crucial role in determining the value of the diffusion constant. This is in agreement with results of previous numerical calculations.[4]

From our derivation of (40) and the general expression (39), we can conclude that the Arrhenius behavior for diffusion only holds at low temperatures such that $kT \ll \Delta$. As the temperature becomes comparable with the diffusion barrier, Eq. (39) indicates we should start to see significant deviations from the Arrhenius behavior. Finally, in the limit $kT \gg \Delta$, the barrier no longer plays any role in the diffusion and we recover from (39) the following behavior for the diffusion constant:

$$D = \frac{kT}{m\eta} . \tag{41}$$

This is of the same form as diffusion in a uniform viscous medium.[7] This kind of continuous crossover from Arrhenius behavior at low temperatures to unactivated diffusion at high temperatures has been observed experimentally as well as in molecular dynamics simulation studies.[3,10]

VII. SUMMARY

In this paper, we have developed a formalism for studying the diffusion of a particle in an inhomogeneous background starting from a microscopic lattice-dynamic Hamiltonian. We use the Mori projection-operator formalism to develop a coupled set of equations for various physical correlation functions which is equivalent to the Kramers equation. The added bonus is a microscopic expression for the frictional force. We then develop an inverse friction continued fraction expansion for the velocity autocorrelation function in the case of a periodic background. This allows us to obtain an analytic expression for the diffusion constant in the high-friction limit for arbitrary dimensions. The analysis of our result for diffusion in two dimensions shows explicitly the crossover from Arrhenius behavior at low temperatures to nonac-

tivated behavior at high temperatures.

The advantage of a microscopic formalism and an expression for the memory function is that we can go systematically beyond the limit of slow diffusion and high-friction limit discussed in this paper. Among many interesting questions not addressed here are the finite coverage effect of interacting particles, higher order corrections to the high-friction limit, and going beyond the initial value approximation when the diffusion time scale is comparable with the background vibrational time scale. Work in these directions is in progress.

APPENDIX

According to the definition of b and a in (11) and (26),

$$b_{10}^{\alpha} a_0^{-1} b_{01}^{\beta} = -\frac{i}{z} \left[\frac{k_B T}{m} \right]^{-2} \chi_{00}^{-1} \omega_{10}^{\alpha} \chi_{00}^{-1} \omega_{01}^{\beta} \chi_{00}^{-1} , \tag{A1}$$

where

$$\omega_{10}^{\alpha} = (\mathbf{A}_1^{\alpha}, L \, \mathbf{A}_0) , \tag{A2}$$

and

$$\omega_{01}^{\beta} = (\mathbf{A}_0, L \, \mathbf{A}_1^{\beta}) . $$

Using the result

$$L A_0(\mathbf{G}) = \mathbf{G} \cdot \mathbf{A}_1(\mathbf{G}) , \tag{A3}$$

the expression for $\omega_{10}^{\alpha}(\mathbf{G}_1, \mathbf{G}_2)$ becomes

$$\omega_{10}^{\alpha}(\mathbf{G}_1, \mathbf{G}_2) = \chi_{11}^{\alpha\delta}(\mathbf{G}_1, \mathbf{G}_2) G_{2\delta} \tag{A4}$$

(repeated indices imply a summation here), and similarly,

$$\omega_{01}^{\beta}(\mathbf{G}_1, \mathbf{G}_2) = G_{1\delta} \chi_{11}^{\delta\beta}(\mathbf{G}_1, \mathbf{G}_2) . \tag{A5}$$

However,

$$\chi_{11}^{\alpha\beta} = \frac{k_B T}{m} \delta_{\alpha\beta} \chi_{00} . \tag{A6}$$

Substitution of (A4), (A5), and (A6) into (A1) finally yields the result that appeared in Eq. (34),

$$(b_{10} a_0^{-1} b_{01})_{\mathbf{G}, \mathbf{G'}}^{\alpha\beta} = -\frac{i}{z} G_{\alpha} \chi_{00}^{-1}(\mathbf{G}, \mathbf{G'}) G'_{\beta} . $$

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