Quantum heat-bath theory of dechanneling

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Dechanneling of a particle moving swiftly in a low-index direction of a crystal is caused by its interaction with its environment, viz., the electrons and phonons of the crystal. At temperatures which are not too low the relaxation of fluctuations in the environment is rapid enough that the environment may be regarded as a quantum heat bath. In this case the Heisenberg equations for the system yield in a semiclassical limit a Fokker-Planck equation describing the dynamics of the particle prior to dechanneling. The transverse diffusion coefficient may be written compactly as a time-averaged force-force correlation function. This viewpoint of dechanneling suggests how the conventional theory may be modified to describe the low-energy regime, as well as highlighting the dechanneling problem as a prototypical example of a particle-heat bath system with nonlinear dissipation. The problem thus has wider relevance to the study of particle-environment systems in general.

I. INTRODUCTION

When a well directed beam of particles is aligned with a low-index axis or plane of a crystal, its penetration depth is found to be much larger than for random crystal orientations. In these low-index orientations the trajectory of a particle through the crystal avoids direct passage towards atomic centers. Instead the impact parameter remains sufficiently large that weak scattering off successive atomic centers gives rise to a guiding and focusing effect that maintains the net forward motion of the particle for many lattice spacings. This phenomenon is referred to as channeling, and has found a number of practical applications, including its utilization in crystal structure analysis, and in the diagnostic technique known as Rutherford backscattering, amongst others.[1] Basic theoretical work on channeling is concerned primarily with calculations of the effective transverse confinement potential holding the particle in the channel, and of the friction forces acting on the particle from the surroundings.[1,2] From these, other quantities of interest in the channeling effect may be computed. More recently, the channeling effect has been investigated in bent crystals, where the possibility is opened, for example, of measuring magnetic moments of the channeling particles.[3]

A channeling particle does not remain indefinitely in its state of forward motion, but at some point may suffer a collision strong enough to remove it from the channel. This is called dechanneling. Since dechanneling causes a decrease in the number of channeling particles, it influences the quantitative interpretation of channeling experiments. As such, the study of dechanneling has also attracted considerable interest. In the classical theory of dechanneling due to Lindhard,[4] the channeling particle is assumed to suffer a succession of small random deflections transverse to the channeling direction. As a result, a phenomenological diffusion equation may be written

for the distribution function of the particle's transverse energy. Dechanneling occurs when the transverse energy exceeds the effective confinement energy of the channel. Beloshitskii and Kumakhov,[5] and Bonderup et al.[6] later gave more rigorous interpretations of the Lindhard theory by showing that it can be derived, under suitable approximations and still in a classical framework, from a Fokker-Planck description of the scattering processes within the channel. Quantum-mechanical considerations were introduced into the problem by Ohtsuki,[7] and a dechanneling theory based on these ideas was presented by Kitagawa and Ohtsuki.[8] In this approach, the particle is treated semiclassically, but its interactions with the crystal are described in quantum-mechanical terms. For a heavy channeling particle the classical and quantum approaches are equivalent under certain conditions.[9] However, the quantum approach permits a deeper study of the physical processes affecting the problem. Both the classical and the quantum-mechanical approaches have steadily been improved and refined.[10-13]

In the present work a theory of dechanneling motivated from the quantum heat-bath viewpoint[14-16] is presented, and it is shown that this gives a more complete description of the problem than conventional theories which are recovered in limits of this new description. The basic premise is that the particle motion along a channeling direction in the host crystal is in many ways equivalent to the dynamics of an irreversible system, e.g., the motion of a Brownian particle. The host crystal acts in two ways on the itinerant particle, presenting both a friction force on it slowing it down, as well as a random force causing diffusive behavior. The particle for its part dissipates kinetic energy to the crystal, energy which for a large crystal has a vanishing probability of returning to the particle. (In reality the crystal is finite, but because the crystal also exchanges heat with its surroundings, the net effect is the same as taking an infinite crystal.) It is

clear that the crystal has a much more dominant effect on the particle than the particle has on the crystal. In the limit where the equilibrium properties of the crystal are barely affected by the presence of the particle, we may regard it as a heat bath. This results in a considerable computational simplification which is used in constructing the theory.

Since the work of Caldeira and Leggett,[14] the most popular method of handling heat-bath problems in which dissipation is an important consideration has been through the use of Feynman's path integral. In this method the density matrix of the particle-environment system is written as a path integral (in either real time or imaginary time depending on the problem at hand), following which the environment variables are traced out by performing the relevant functional integrals over cyclic paths, leaving just the reduced density matrix for the particle alone. Since only Gaussian functional integrals can be evaluated explicitly, the type of heat baths that are normally used are restricted to those that can be expressed as a sum of simple harmonic oscillator degrees of freedom. However, this idealization is not always possible, as in the dechanneling problem where the environment is very complicated. Although a path-integral treatment may, in principle, be possible for this problem, quantities of interest can be obtained directly from a Fokker-Planck equation, and this Fokker-Planck equation is obtained more straightforwardly by alternative means.

A major complication in the analysis is that the dynamics of the channeling particle does not permit linear-response arguments to be invoked, except at very low particle velocities where, in fact, treatment of ionization effects becomes problematic. Indeed, in a region of great interest, the Bethe-Bloch regime, the friction force acting on the particle is proportional to v^{-2}, which is clearly nonlinear. The difficulty in handling nonlinear quantum systems in general prevents a complete and rigorous solution to the dechanneling problem. However, the more modest objective of obtaining a Fokker-Planck equation is attainable,[12,13] and a derivation of such an equation is sufficient for the further study of dechanneling. The overall strategy employed in this particular work will be to write the Heisenberg equations for the system of particle plus environment in the form of a generalized Langevin equation assuming a heavy channeling particle and high temperatures, and then to transform to an approximate Fokker-Planck description assuming that the crystal acts as a heat bath. The Heisenberg equations at the starting point of the analysis form a complete quantum-mechanical description of the particle together with its interactions with the crystal environment. The final Fokker-Planck equation is a contracted description for an essentially classical particle undergoing stochastic motion according to macroscopic friction and diffusion coefficients. These latter parameters are expressed as expectations of quantum-mechanical operators associated with the underlying quantum heat bath. The nonlinearity of the problem is manifested through the inapplicability of the usual fluctuation-dissipation theorem to the Fokker-Planck description.

The study of dechanneling is thus formulated in terms of a particle in a heat-bath problem. The corresponding Fokker-Planck description is possible for a heavy particle in a high-temperature limit where memory effects in the environment are largely suppressed. Similar Fokker-Planck equations have been derived before, but the method used here is different. The theory reduces to the conventional Lindhard energy diffusion equation in the Bethe-Bloch regime, and the analysis clarifies some of the implicit assumptions made in the Lindhard theory, such as in the treatment of the environment. Memory effects in the environment can be introduced into the theory perturbatively, but the resulting differential equations are considerably more difficult to solve. The particle in a heat-bath approach also suggests a possible method by which the conventional dechanneling theory can be modified to describe very low-energy particles. At very low energies, ionization effects are important, and this has hitherto precluded any reliable analysis of dechanneling in this regime.

In Sec. II, the influence of the crystal on the channeling particle is expressed in terms of a quantum force operator. Using a Markov approximation for this force, dynamical equations governing the time evolution of the particle and force are written. In Sec. III, a semiclassical approximation for the particle is used to obtain a Fokker-Planck description of the particle-crystal system. Regarding the crystal as a heat bath, projective techniques are applied to eliminate the force variable from the problem resulting in a reduced Fokker-Planck equation with a diffusion coefficient expressed as the time average of a force-force correlation function. In Sec. IV, the total force operator is expanded to low orders in the interaction potential, and explicit expressions obtained for the stopping power and diffusion coefficient. In Sec. V, the equivalence of the present theory to the conventional quantum formulation of dechanneling is demonstrated, and the force-force correlation function is used to write down the plasmon, phonon, and core electron contributions to the diffusion coefficient. Finally in Sec. VI, a method of extending dechanneling theory to the low-energy regime (where the Lindhard theory is not valid) is suggested, and the relevance of the analysis to wider particle-environment problems is discussed. Mathematical details of the projective techniques used in Sec. III are given in the Appendix.

II. PARTICLE-ENVIRONMENT SYSTEM

In this section we define the Hamiltonian for the problem, and write down the Heisenberg equations for the particle operators. The influence of the environment on the particle is expressed in terms of a quantum force operator. A separation of time scales argument is advanced to isolate slowly varying and rapidly fluctuating components of this force. The particle Heisenberg equations then resemble the Langevin equations for a Brownian particle in a potential. The Mori-Zwanzig projection operator formalism is used as a stepping stone to obtain a linearized relaxation equation for the fluctuating component of the force in a high-temperature, Markov limit.

The resulting equations allow us to obtain a Fokker-Planck description of the problem subsequently in Sec. III. Finally, we comment on the validity of the separation of time scales argument.

A. Heisenberg equations for particle

Consider a charged particle moving at fast but nonrelativistic speeds along a low-index direction of a perfect unstrained crystal with no external applied fields. The total Hamiltonian of the system comprises the particle, electron, and lattice terms together with the respective couplings. Forces acting on the particle are due to interactions with the electrons and the lattice. These include the force from the periodic crystal potential (or channel potential) arising from elastic particle-electron and particle-lattice scattering, and the friction and random forces arising from inelastic scattering. We wish to express these forces in terms of the above interactions, and thereby elicit dynamical equations governing the system evolution.

The particle-electron interaction is

$$\hat{W}_e = -\sum_{\mathbf{r}_j} \frac{e^2}{|\hat{\mathbf{r}} - \hat{\mathbf{r}}_j|}, \tag{2.1}$$

where $\hat{\mathbf{r}}$ is the position of the particle, and its charge is assumed to be $+e$. The summation extends over the positions of all electrons in the crystal. The particle-lattice interaction is

$$\hat{W}_L = \sum_{\mathbf{R}_j} U_L(\hat{\mathbf{r}} - \hat{\mathbf{R}}_j), \tag{2.2}$$

where $U_L(\mathbf{r})$ is the particle-nucleus potential, and the sum is over all lattice nuclei positions. A monatomic basis is assumed since a more complex basis does not change the basic arguments. In Eqs. (2.1) and (2.2) the electrons and nuclei in the crystal appear in separate sums. However, since only the more weakly bound electrons of any particular lattice ion will generally be able to interact strongly with the charged particle, in practice the sum in Eq. (2.1) can be restricted to include only these latter electrons, and the potential $U_L(\mathbf{r})$ in Eq. (2.2) modified to suit.

It is convenient to replace the two interactions above by a single particle-environment interaction. Writing $V_{\mathbf{q}} = 4\pi e^2/q^2$ and $U_{\mathbf{q}}$ as the qth Fourier components of the Coulomb potential and $U_L(\mathbf{r})$, respectively, and $\hat{\rho}_{\mathbf{q}} = \sum_j e^{-i\mathbf{q}\cdot\hat{\mathbf{r}}_j}$ and $\hat{\eta}_{\mathbf{q}} = \sum_j e^{-i\mathbf{q}\cdot\hat{\mathbf{R}}_j}$ as the electron density and lattice density operators, we introduce the vectors

$$\Phi_{\mathbf{q}} = \begin{pmatrix} -V_{\mathbf{q}} \\ U_{\mathbf{q}} \end{pmatrix}, \quad \hat{\zeta}_{\mathbf{q}} = \begin{pmatrix} \hat{\rho}_{\mathbf{q}} \\ \hat{\eta}_{\mathbf{q}} \end{pmatrix}. \tag{2.3}$$

Further denoting the scalar product of $\Phi_{\mathbf{q}}$ and $\hat{\zeta}_{\mathbf{q}}$ as $\Phi_{\mathbf{q}} \cdot \hat{\zeta}_{\mathbf{q}}$ we have

$$\hat{W} = \hat{W}_e + \hat{W}_L = \sum_{\mathbf{q}\neq 0} \Phi_{\mathbf{q}} \cdot \hat{\zeta}_{\mathbf{q}} e^{i\mathbf{q}\cdot\hat{\mathbf{r}}}. \tag{2.4}$$

The $\mathbf{q} = 0$ component in the summation in Eq. (2.4) is omitted since the crystal is charge neutral in absence of the particle.

The total Hamiltonian for the system is

$$\hat{H} = \hat{T}_p + \hat{H}_E + \hat{W}, \tag{2.5}$$

where $\hat{T}_p = \hat{p}^2/2m$, m and $\hat{\mathbf{p}}$ are the mass and momentum of the particle, and $\hat{H}_E$ is the Hamiltonian for the environment and can be written solely in terms of electron and phonon operators. $\hat{W}$ will contribute to both elastic and inelastic scattering of the particle. Denoting these contributions as $\hat{U}$ and $\hat{H}_i$, respectively, we rewrite $\hat{H}$ as

$$\hat{H} = \hat{H}_p + \hat{H}_E + \hat{H}_i, \tag{2.6}$$

where $\hat{H}_p = \hat{T}_p + \hat{U}$ depends only on particle operators. Note that $\hat{U}$ is defined to be that part of $\hat{W}$ independent of electron or phonon operators, $[\hat{U}, \hat{H}_E] = 0$.

With the Hamiltonian $\hat{H}$, the Heisenberg equations for the particle operators are

$$i\hbar\partial_t \hat{\mathbf{r}}(t) = [\hat{\mathbf{r}}(t), \hat{H}], \quad \hat{\mathbf{r}}(0) = \hat{\mathbf{r}}, \tag{2.7a}$$

$$i\hbar\partial_t \hat{\mathbf{p}}(t) = [\hat{\mathbf{p}}(t), \hat{H}], \quad \hat{\mathbf{p}}(0) = \hat{\mathbf{p}}, \tag{2.7b}$$

where in the Heisenberg representation

$$\hat{\mathbf{r}}(t) = e^{i\hat{H}t/\hbar}\hat{\mathbf{r}}e^{-i\hat{H}t/\hbar}, \tag{2.8}$$

etc., and ∂_t denotes a time derivative. Evaluating the commutators in Eq. (2.7) gives

$$\partial_t \hat{\mathbf{r}}(t) = \hat{\mathbf{p}}(t)/m, \tag{2.9a}$$

$$\partial_t \hat{\mathbf{p}}(t) = \hat{\mathbf{F}}(t). \tag{2.9b}$$

$\hat{\mathbf{F}}(t)$ is defined as the force operator, which we may write as

$$\hat{\mathbf{F}}(t) = \hat{\mathbf{F}}^{el}(t) + \hat{\mathbf{F}}^{in}(t), \tag{2.10}$$

where $\hat{\mathbf{F}}^{el} = -i\hbar^{-1}[\hat{\mathbf{p}}, \hat{U}]$, and $\hat{\mathbf{F}}^{in} = -i\hbar^{-1}[\hat{\mathbf{p}}, \hat{H}_i]$. If any $\hat{\mathbf{p}}$ dependence of $\hat{U}$ or $\hat{H}_i$ is negligible, we may write $\hat{\mathbf{F}} = -\partial_{\mathbf{r}}\hat{U} - \partial_{\mathbf{r}}\hat{H}_i$. The force operator $\hat{\mathbf{F}}(t)$ is itself governed by a Heisenberg equation. We could of course try to solve approximately the heirarchy of Heisenberg equations. However, a more physically appealing approach lends itself if we notice that by replacing $\partial_{\mathbf{r}}\hat{H}_i$ by a sum of friction and "random" components, the above equations resemble the Langevin equations of a Brownian particle in a potential $\hat{U}$. We argue that $\partial_{\mathbf{r}}\hat{H}_i$ can indeed be written this way. Stochastic methods then lead to tractable evolution equations in a limit where the particle is treated semiclassically.

Application of stochastic methods invariably entails the use of the Mori-Zwanzig projection operator formalism.[17,18] For the force operator $\hat{\mathbf{F}}(t)$, this results in a generalized Langevin equation with a friction force

comprising $\hat{\mathbf{v}}(t)$ convoluted with a complicated memory term, where $\hat{\mathbf{v}} = \hat{\mathbf{p}}/m$ is the particle velocity and we will use v for the magnitude of the semiclassical velocity. The friction force, or stopping power, for a heavy particle is generally a complicated function of the particle velocity.[2,19,20] For very low velocities a linear dependence of stopping power on v is observed. For fast particles the stopping power exhibits the characteristic Bethe-Bloch v^{-2} dependence arising from inelastic scattering with electrons. At intermediate velocities there is a crossover region between the above two limits. At much higher velocities, relativistic corrections become important. How this complete velocity dependence may be extracted from the generalized Langevin equation is not obvious.

Other methods also exist for obtaining Langevin equations for a quantum system. Schmid[21] used a path-integral approach to obtain a generalized Langevin equation. However, the problems with path-integral approaches have been mentioned already in the Introduction: it is not clear how the general Hamiltonian of Eq. (2.5) should be handled by this method. Ford, Lewis, and O'Connell[22] showed that a generalized Langevin equation can be written rather directly from the Heisenberg equations when the interaction part of the Hamiltonian can be written linear in terms of time-dependent operators. Indeed, Hu and O'Connell[15] used this approach to study a system of electrons and phonons interacting with a "center-of-mass" coordinate ("particle"), bearing much resemblence to the system under study here. However, their treatment of phonons is simpler than here, and the Hamiltonian they used is not sufficient to recover all known results on diffusion coefficients. Applying their methods to the Hamiltonian of Eq. (2.5) leads to difficulties since it is not clear how to manipulate the more general form of the phonon operators.

Rather than proceed by one of these methods, we will separate the force operator into systematic and random components directly by a different method. The Mori-Zwanzig procedure will be utilized subsequently to motivate a study of the random force, which has somewhat simpler properties than the full force $\hat{\mathbf{F}}(t)$.

B. Quantum force operator

Before continuing it is useful to consider an analogy with a simple example from nonequilibrium thermodynamics.[23] When a quantity Q is in equilibrium, the fluctuations δQ it exhibits about its equilibrium value $\overline{Q}$ may be ascribed to the influence of a random force, e.g., from a heat bath. Away from equilibrium, Q still feels the random force, but in addition experiences a friction force which drives it back towards $\overline{Q}$. In the linear regime this description is embodied in the Langevin equation, and is one particular motivation for the use of the Mori-Zwanzig approach. In the present context of a particle in a crystal, the equilibrium state is one in which the particle is nominally at rest, i.e., $\langle \hat{\mathbf{v}} \rangle = 0$, in the crystal at a finite fixed temperature. In this state, the particle is influenced by random thermal forces which maintain its vi-

brational motion at a level above its quantum-mechanical zero-point motion (excluding any possible activated diffusion, etc.). The amplitude of this vibration is smaller the more massive the particle. The nonequilibrium state of interest corresponds to the particle in forward motion. Now, in addition to the random force, the particle experiences a friction force due to inelastic scattering from electrons and phonons, which drives it back towards a state of nominal rest.

In what follows, we denote the expectation of an operator $\hat{A}$ as

$$\langle \hat{A}(t) \rangle = \mathrm{Tr}\,\hat{\rho}_0 \hat{A}(t), \quad \hat{\rho}_0 = \hat{\rho}_E \hat{\rho}_p, \quad (2.11)$$

where $\hat{\rho}_0$ is the density matrix of the system at time $t = 0$, $\hat{\rho}_E = e^{-\beta \hat{H}_E}/\mathrm{Tr}\,e^{-\beta \hat{H}_E}$, and $\hat{\rho}_p$ is the initial density matrix of the particle. For simplicity we assume the particle to enter the crystal in a momentum eigenstate $|\mathbf{K}_0\rangle$, so $\hat{\rho}_p = |\mathbf{K}_0\rangle\langle\mathbf{K}_0|$, although this initial condition is readily generalized. In the analysis to follow in later sections, we will consider only the motion of a heavy channeling particle. The wave packet associated with such a particle has finite extent (is "small"), and a semiclassical approximation may be made. In other words, we will approximate quantum-mechanical operators associated with the heavy particle, such as its position and velocity, by their corresponding expectations.[24] From Eq. (2.10) we see that the total force operator has components arising from elastic-scattering processes and from inelastic-scattering processes. We therefore examine the effect of these components on the motion of a semiclassical particle as defined above.

Consider the commutator of the total particle energy operator $\hat{H}_p(t)$ with $\hat{H}$:

$$[\hat{H}_p(t), \hat{H}] = \tfrac{1}{2} i\hbar \left\{ \hat{\mathbf{v}}(t) \cdot \hat{\mathbf{F}}^{\mathrm{in}}(t) + \hat{\mathbf{F}}^{\mathrm{in}}(t) \cdot \hat{\mathbf{v}}(t) \right\}. \quad (2.12)$$

Taking the expectation of Eq. (2.12),

$$\partial_t \langle \hat{H}_p(t) \rangle = \mathbf{v} \cdot \langle \hat{\mathbf{F}}^{\mathrm{in}}(t) \rangle, \quad (2.13)$$

where $\mathbf{v} = \langle \hat{\mathbf{v}}(t) \rangle$ is the semiclassical velocity of the particle. Equation (2.13) tells us that the total particle energy is influenced only by $\hat{\mathbf{F}}^{\mathrm{in}}$. Hence $\hat{\mathbf{F}}^{\mathrm{in}}$ can only arise from inelastic-scattering processes as originally intimated. On the other hand, the particle kinetic energy operator gives

$$\partial_t \langle \hat{T}_p(t) \rangle = \mathbf{v} \cdot \langle \hat{\mathbf{F}}^{\mathrm{el}}(t) + \hat{\mathbf{F}}^{\mathrm{in}}(t) \rangle. \quad (2.14)$$

Since we expect the particle kinetic energy to be affected both by inelastic processes and by the crystal potential (interaction with which is elastic), $\langle \hat{\mathbf{F}}^{\mathrm{el}}(t) \rangle$ may be identified as the force arising from the crystal potential.

Further, we separate $\hat{\mathbf{F}}^{\mathrm{in}}(t)$ into two components, $\hat{\mathbf{S}}_p(t)$ arising by virtue of the net forward motion of the particle, and $\hat{\mathbf{f}}(t)$ being completely independent of this motion:

$$\hat{\mathbf{F}}^{\mathrm{in}}(t) = -\hat{\mathbf{S}}_p(t) + \hat{\mathbf{f}}(t). \quad (2.15)$$

Since $\hat{\mathbf{S}}_p(t)$ is dependent on the velocity, we see that it should have some relation to the friction force (this is shown below), and $\hat{\mathbf{f}}(t)$ will be any remaining contribu-

tion to $\hat{\mathbf{F}}^{\text{in}}(t)$. A simplification of the problem is possible if we can separate the time scales of variations in $\hat{\mathbf{S}}_p(t)$ and $\hat{\mathbf{f}}(t)$. We discuss in Sec. II D under what conditions this may be achieved. $\hat{\mathbf{f}}(t)$ turns out to be rapidly varying, whereas $\hat{\mathbf{S}}_p(t)$ varies much more slowly with time. Assuming this separation of time scales, we define the time average over a time interval ε as

$$\overline{A(t)}_\varepsilon = \frac{1}{\varepsilon} \int_{t-\varepsilon/2}^{t+\varepsilon/2} dt' \, \hat{A}(t'). \qquad (2.16)$$

The time ε is sufficiently long that the average of $\hat{\mathbf{f}}(t)$ vanishes over this interval, $\overline{\mathbf{f}(t)}_\varepsilon = 0$, but short enough that the mean particle velocity does not vary significantly, so $\overline{\mathbf{S}_p(t)}_\varepsilon = \hat{\mathbf{S}}_p(t)$. Then we have $\overline{\mathbf{F}^{\text{in}}(t)}_\varepsilon = -\hat{\mathbf{S}}_p(t)$.

Using $\partial_t = \mathbf{v} \cdot \partial_{\mathbf{r}}$, and averaging Eq. (2.13) over ε,

$$\langle \hat{\mathbf{S}}_p(t) \rangle = -\partial_{\mathbf{r}} \overline{E}, \qquad (2.17)$$

where $\overline{E} = \overline{\langle H_p(t) \rangle}_\varepsilon$ is the total energy of the particle once we have smoothed out any rapid fluctuations arising from the force $\hat{\mathbf{f}}(t)$. Therefore $\partial_{\mathbf{r}} \overline{E}$ is the net rate of change in particle energy with displacement, which is just the stopping power. Denoting the magnitude of this stopping power as S_p, we write $\langle \hat{\mathbf{S}}_p(t) \rangle = \mathbf{u} S_p$ (there is no minus sign here because $\overline{E}$ decreases with $\mathbf{r}$), with $\mathbf{u} = \mathbf{v}/v$ being a unit vector pointing in the direction of motion of the particle. Thus we call $\hat{\mathbf{S}}_p(t)$ the friction force operator, and $\hat{\mathbf{f}}(t)$ the random force operator. In principle, we could also expect a rapidly varying component of force arising from elastic-scattering processes. However, Lindhard shows that for a sufficiently fast moving particle this is not so.[4] In Sec. IV we obtain an expression for $\langle \hat{\mathbf{S}}_p(t) \rangle$ which we show in Sec. V to be identical to the usual expression for stopping power.

C. Dynamical equation for random force

The random force $\hat{\mathbf{f}}(t)$ is effectively $\hat{\mathbf{F}}^{\text{in}}(t)$ evaluated for the state of equilibrium defined by $\langle \hat{\mathbf{v}}(t) \rangle = 0$, and as such should exhibit only equilibrium fluctuations. This permits a very useful approximation to describe the dynamics of $\hat{\mathbf{f}}(t)$. We use Mori's analysis to rewrite the Heisenberg equation that governs $\hat{\mathbf{f}}(t)$, viz.,

$$i\hbar \partial_t \hat{\mathbf{f}}(t) = [\hat{\mathbf{f}}(t), \hat{H}], \qquad (2.18)$$

as the generalized Langevin equation[17,25]

$$\partial_t \hat{\mathbf{f}}(t) = -\int_0^t dt' M(t-t') \hat{\mathbf{f}}(t') + \hat{\xi}_f(t), \qquad (2.19\text{a})$$

$$\langle \hat{\xi}_f(t) \hat{\mathbf{f}}(0) \rangle_K = 0, \qquad (2.19\text{b})$$

$$\langle \hat{\xi}_f(t) \hat{\xi}_f(t') \rangle_K = M(t-t') \langle \hat{\mathbf{f}}(0) \hat{\mathbf{f}}(0) \rangle_K, \qquad (2.19\text{c})$$

with the initial condition $\hat{\mathbf{f}}(0) = \hat{\mathbf{f}}$. Note that Eqs. (2.18)

and (2.19) are identically equivalent. A further term (the frequency matrix) is absent from Eq. (2.19a) due to the Hermiticity of $\hat{\mathbf{f}}(t)$. Here $M(t)$ is the memory function (matrix), and $\hat{\xi}_f(t)$ has the appearance of a stochastic quantity. The Kubo scalar product of quantities $\hat{\mathbf{A}}$ and $\hat{\mathbf{B}}$ (scalar or vector) is defined[25,26]

$$\langle \hat{\mathbf{A}}(t) \hat{\mathbf{B}}(t') \rangle_K = \beta^{-1} \int_0^\beta d\overline{\beta} \langle \hat{\mathbf{A}}(t - i\hbar\overline{\beta}) \hat{\mathbf{B}}(t') \rangle$$
$$- \langle \hat{\mathbf{A}}(t) \rangle \langle \hat{\mathbf{B}}(t') \rangle. \qquad (2.20)$$

If $\hat{\mathbf{A}}$ and $\hat{\mathbf{B}}$ are vectors then $\hat{\mathbf{A}}\hat{\mathbf{B}}$ is the outer product. Equation (2.19c) is an expression of the fluctuation-dissipation theorem for the random force fluctuations. The angular brackets appearing in Eq. (2.20) are expectations as defined in Eq. (2.11).

There are two features of Eqs. (2.19) that make an exact solution of the problem very difficult. These are the Kubo scalar product, and the memory function $M(t)$, neither of which can be evaluated exactly. However, we argue below that at "high" temperatures the Kubo scalar product simplifies to an ordinary correlation function, and $M(t)$ becomes effectively a δ function. These result from the main consequence of the high-temperature condition, that is that the thermal energy $1/\beta = k_B T$ becomes large compared to other energy scales, and certain quantum processes in the environment become suppressed (short lived). As one example, in metals, quantum corrections to the conductivity tend to zero at temperatures above $\sim$4 K, at which point the bulk macroscopic condictivity is given by just the classical Drude formula. As another example, in superconducting materials, superconductivity is quenched above the critical temperature. Thus, at sufficiently high temperatures, in practice exceeding only a few Kelvin, all such quantum coherence effects in the environment are largely dominated by thermal fluctuations.

Considering only high temperatures of course means that the emerging theory will only be applicable at these temperatures, but this is not a problem since channeling experiments are not usually conducted at extremes of temperature. In any case, energy dissipated by the channeling particle will presumably cause local heating sufficient to smear many of the quantum effects in the environment in the immediate vicinity of the particle, even at extremely low temperatures. Thus the high-temperature condition may actually be valid at lower ambient crystal temperatures than the arguments below imply.

The force fluctuations $\hat{\mathbf{f}}(t)$ arise from the local environment of the moving particle. The arguments above show that coherent excitations in the environment, by which we mean fluctuations in the electron or lattice density operators, are very short lived at the sort of temperatures under consideration. In this case, imagine the environment to be divided into identical blocks (or cells) of coherent regions, with the force $\hat{\mathbf{f}}(t)$ on the particle being due to blocks lying on or near the trajectory of the particle. The size of a block will depend on the characteristic "coherence length" of the fluctuations.[27] At relatively high temperatures this will be of the order of the

interatomic spacing. Assume that the coherent fluctuations within these blocks have a characteristic lifetime τ_ρ, which is dependent on the temperature through scattering rates of excitations within the block. Two important points are evident regarding these coherent excitation fluctuations. First is that fluctuations in spatially separate blocks are uncorrelated, since the fluctuation is spatially confined to within a block. Second is that temporally consecutive fluctuations within the same block are also uncorrelated, since the fluctuation is also temporally confined to within a time duration τ_ρ.[27] Since different fluctuations are uncorrelated, we see that over sufficiently long time scales the fluctuations develop a Markovian behavior, meaning that one fluctuation becomes independent of previous fluctuations for a sufficiently long time interval between them. We will use this observation below.

Consider one particular block lying near the trajectory of the particle. As the particle moves past this block, it experiences a force fluctuation for a duration of time τ_ρ due to a coherent excitation fluctuation (i.e., electron or lattice density fluctuation) within the block. If the particle is moving so rapidly that it leaves the vicinity of the block within the time τ_ρ, then the actual force fluctuation it feels only has a significant magnitude for a time τ_m which is less than τ_ρ. If however τ_ρ is sufficiently small, then the particle will feel this force for the duration $\tau_m \approx \tau_\rho$. Thus τ_m is the effective time duration over which the particle feels the force due to a coherent excitation fluctuation within a block. Because of this, and recalling the arguments of the previous paragraph, the random force $\hat{\mathbf{f}}(t)$ felt by the particle for times much longer than $t_0 + \tau_m$ becomes independent of (i.e., uncorrelated to) the force at time t_0, so the "memory" of the random force goes to zero $M(t) \to 0$ for $t \gg \tau_m$. In other words, over time scales much longer than τ_m the particle does not see the full details of the memory effects but only the effective memory function

$$M(t) = 2M\delta(t), \quad t \gg \tau_m. \tag{2.21}$$

This constitutes a Markov approximation for the random force fluctuations. Additionally, the very short lifetimes of the coherent excitation fluctuations imply that the real part of M becomes very large, dominating the imaginary part. Thus putting $\operatorname{Re} M = \lambda$, Eqs. (2.19) and (2.21) together then give

$$\langle \hat{\mathbf{f}}(t)\hat{\mathbf{f}}(0)\rangle_K = \langle \hat{\mathbf{f}}(0)\hat{\mathbf{f}}(0)\rangle_K e^{-\lambda t}, \tag{2.22}$$

which represents the long-time relaxation behavior of the second moment of $\hat{\mathbf{f}}(t)$. The parameter λ gives a measure of the degree of correlation between coherent fluctuations at different times.

A further simplification arises in the high-temperature limit of the Kubo scalar product defined in Eq. (2.20). Recall in this limit that the thermal energy $1/\beta$ becomes large compared to other energy scales, i.e., $1/\beta \gg \hbar\omega$ where $\hbar\omega$ represents the characteristic energy scale of quantum processes in the environment. Thus, the typical energies contributing strongly to the Kubo scalar prod-

uct are those for which $1/\bar{\beta} \sim \hbar\omega$. Since $\bar{\beta} \leq \beta$, the integrand in Eq. (2.20) may therefore be approximated by its $\bar{\beta} \approx 0$ limit, which leads to

$$\begin{aligned} \langle \hat{\mathbf{f}}(t)\hat{\mathbf{f}}(0)\rangle_K &\approx \langle \hat{\mathbf{f}}(t)\hat{\mathbf{f}}(0)\rangle_c \\ &\equiv \langle \hat{\mathbf{f}}(t)\hat{\mathbf{f}}(0)\rangle - \langle \hat{\mathbf{f}}(t)\rangle\langle \hat{\mathbf{f}}(0)\rangle. \end{aligned} \tag{2.23}$$

This is the high-temperature limit of the Kubo scalar product. Another way of viewing this would be to consider the lifetimes of coherent excitations in the environment. These lifetimes are dominated by the strongest scattering mechanism operative in a particular temperature range, and may be expressed in the form $\tau_\rho \propto T^{-d}$ for some power d, and some temperature range. Generally $d \gtrsim 1$ for scattering mechanisms in three dimensions[28,29] so τ_ρ will decrease more rapidly with temperature than does β. Since $\tau_m \lesssim \tau_\rho$, we see that for sufficiently high temperatures, the inequality $\tau_m \lesssim \hbar\beta$ should approximately hold. Now $\bar{\beta} \leq \beta$ in the integrand of Eq. (2.20), so $\tau_m \lesssim \hbar\bar{\beta}$; and since Eq. (2.22) is applicable over times $t \gg \tau_m$, we obtain the condition $t \gg \hbar\bar{\beta}$. Inserting this into the expression for $\langle \hat{\mathbf{f}}(t)\hat{\mathbf{f}}(0)\rangle_K$ gives again Eq. (2.23) as above. In fact, Eq. (2.23) also follows from Eq. (2.20) upon taking $\hbar \to 0$, suggesting a classical limit. However, this is not a true classical limit, since the expectations are still of quantum-mechanical operators, and may be referred to instead as a classical approximation.

Collecting the above results together, we have for the high-temperature behavior of the second moment of $\hat{\mathbf{f}}(t)$,

$$\langle \hat{\mathbf{f}}(t)\hat{\mathbf{f}}(0)\rangle_c = \langle \hat{\mathbf{f}}(0)\hat{\mathbf{f}}(0)\rangle_c e^{-\lambda t}. \tag{2.24}$$

Such behavior is characteristic of an underlying Ornstein-Uhlenbeck process,[30]

$$\partial_t \hat{\mathbf{f}}(t) = -\lambda \hat{\mathbf{f}}(t) + \hat{\xi}(t), \tag{2.25a}$$

$$\langle \hat{\xi}(t)\hat{\mathbf{f}}(0)\rangle_c = 0, \tag{2.25b}$$

$$\langle \hat{\xi}(t)\hat{\xi}(t')\rangle_c = 2\lambda\langle \hat{\mathbf{f}}(0)\hat{\mathbf{f}}(0)\rangle_c \delta(t - t'), \tag{2.25c}$$

where $t - t' \gg \tau_m$, and $\hat{\xi}(t)$ is a Gaussian white noise source. Equation (2.25) describes the small "equilibrium" fluctuations of the force operator when we consider the channeling of a fast particle at not too low temperatures. An analogous equation governs the long-time velocity diffusion of a Brownian particle, for example.

D. Separation of time scales

The above analysis shows under what conditions the separation of time scales we alluded to earlier arises. At high temperatures and for a fast moving particle, the force $\langle \hat{\mathbf{f}}(t)\rangle$ felt by the particle fluctuates over a time τ_m. On the other hand, because the friction force felt by a fast moving particle is comparatively weak (proportional to v^{-2} in the Bethe-Bloch regime), the mean particle veloc-

ity only changes significantly in an interval many times the duration τ_m. Thus whereas $\langle \hat{\mathbf{f}}(t) \rangle$ changes over a time scale τ_m, $\langle \hat{\mathbf{S}}_p(t) \rangle$ changes over a time scale much longer than this. We define ε in Eq. (2.16) to lie somewhere between these two limits.

III. FORMULATION OF DECHANNELING

The Heisenberg equations for the particle operators $\hat{\mathbf{r}}$ and $\hat{\mathbf{p}}$ provide a real-space description of possible trajectories the particle may follow. If we confine ourselves to initial conditions resulting in particle motion along a low-index direction of the crystal, the problem at hand is to find the probability of the particle dechanneling at any particular time or depth. We will show that from the dynamical equations for $\hat{\mathbf{r}}$, $\hat{\mathbf{p}}$, and $\hat{\mathbf{f}}$ derived in the previous section, suitable approximations yield a Fokker-Planck equation from which quantities of interest may be calculated.

When the forward momentum of the particle is much greater than its transverse momentum, the Lindhard continuum potential is applicable,[4] and we may separate the motion of the particle into components parallel and transverse to the channeling direction, with no coupling between the two. Dechanneling then requires us to study the transverse component of the particle motion. Define $\mathbf{n}_\mu$ to be unit vectors perpendicular to the channeling direction. For planar channeling, the index μ takes only one value, and $\mathbf{n}_\mu$ is the direction normal to the plane. For axial channeling, μ can take two values. The corresponding vectors are orthonormal, $\mathbf{n}_\mu \cdot \mathbf{n}_\nu = \delta_{\mu\nu}$, and span the plane normal to the channeling axis. Projecting Eqs. (2.9) and (2.25) in the direction of $\mathbf{n}_\mu$, and denoting the vector with components $\hat{r}_\mu$ as $\hat{\mathbf{r}}_\perp$ (and likewise for $\hat{p}_\mu$, $\hat{f}_\mu$, etc.):

$$\partial_t \hat{\mathbf{r}}_\perp(t) = \hat{\mathbf{p}}_\perp(t)/m, \qquad (3.1a)$$

$$\partial_t \hat{\mathbf{p}}_\perp(t) = \hat{\mathbf{F}}_\perp^{el}(t) - \hat{\mathbf{S}}_\perp(t) + \hat{\mathbf{f}}_\perp(t), \qquad (3.1b)$$

$$\partial_t \hat{\mathbf{f}}_\perp(t) = -\lambda \hat{\mathbf{f}}_\perp(t) + \hat{\xi}_\perp(t), \qquad (3.1c)$$

$$\langle \hat{\xi}_\mu(t) \hat{\xi}_\nu(t') \rangle_c = 2\lambda D_{\mu\nu}^f \delta(t - t'), \qquad (3.1d)$$

where $D_{\mu\nu}^f = \langle \hat{f}_\mu(0) \hat{f}_\nu(0) \rangle_c$, and $t - t' \gg \tau_m$.

Although it appears that we may go directly now to a Fokker-Planck equation, the fact that none of $\hat{\mathbf{r}}_\perp$, $\hat{\mathbf{p}}_\perp$, or $\hat{\mathbf{f}}_\perp$ commute with one another makes a suitable distribution function difficult to define, although it should reduce to something resembling a Wigner function after "integrating out" the force $\hat{\mathbf{f}}_\perp$. Rather, we proceed as follows. We assume a heavy channeling particle. The wave packet associated with this particle will have little extent, and we can replace the operators $\hat{\mathbf{r}}_\perp$ and $\hat{\mathbf{p}}_\perp$ by the classical variables $\mathbf{r}_\perp$ and $\mathbf{p}_\perp$, which are the mean position and momentum of the particle wave packet. This corresponds

to a semiclassical approximation for the particle.[24] We also replace the force operators and $\hat{\xi}_\perp(t)$ by their expectations. From the discussion in the previous section, we may write $\langle \hat{\mathbf{F}}_\perp^{el}(t) \rangle = -U_c'(\mathbf{r}_\perp)$, and

$$-\langle \mathbf{n}_\perp \cdot \hat{\mathbf{S}}_p(t) \rangle \approx -(S_p/p_0)p_\perp = -\gamma p_\perp, \qquad (3.2)$$

where the initial momentum of the particle is $p_0 = \hbar K_0$, and the forward momentum is taken to be much larger than $p_\perp$. With $\langle \hat{\mathbf{f}}_\perp(t) \rangle = \mathbf{f}_\perp(t)$ and $\langle \hat{\xi}_\perp(t) \rangle = \xi_\perp(t)$, we obtain from Eq. (3.1) the Fokker-Planck equation[31] for the distribution function $P(\mathbf{r}_\perp, \mathbf{p}_\perp, \mathbf{f}_\perp)$:

$$\partial_t P = (L_a + L_i + \lambda L_b)P, \qquad (3.3a)$$

$$L_a = -\frac{\mathbf{p}_\perp}{m} \cdot \frac{\partial}{\partial \mathbf{r}_\perp} + \frac{\partial}{\partial \mathbf{p}_\perp} \cdot [\gamma \mathbf{p}_\perp + U_c'(\mathbf{r}_\perp)], \qquad (3.3b)$$

$$L_i = -\mathbf{f}_\perp \cdot \frac{\partial}{\partial \mathbf{p}_\perp}, \qquad (3.3c)$$

$$L_b = \frac{\partial}{\partial \mathbf{f}_\perp} \cdot \left(\mathbf{f}_\perp + D_f \frac{\partial}{\partial \mathbf{f}_\perp} \right). \qquad (3.3d)$$

For simplicity, we approximate the crystal as being isotropic about the channeling direction, and write the force diffusion coefficient as (no summation over the repeated indices)

$$D_f = D_{\mu\mu}^f = \langle \hat{f}_\mu(0) \hat{f}_\mu(0) \rangle_c. \qquad (3.4)$$

More general situations can be handled by continued use of the diffusion tensor.

We next introduce the heat-bath concept into the problem. Till now we have regarded the particle as a subsystem interacting with its environment (the crystal), the particle and environment comprising the total system. Now, the kinetic energy of the particle is negligible compared to the internal energy of the crystal. Further, the relatively high temperatures we assume imply correspondingly large relaxation rates for density fluctuations (large τ_ρ), in addition to significantly decreased correlations between the fluctuations (large λ). So energy lost by the particle is distributed rapidly amongst the many degrees of freedom of the crystal. Consequently, we see that the equilibrium properties of the crystal are only slightly disturbed by the action of the particle, whereas the particle is markedly influenced by the crystal. The crystal thus has a property resembling that of a heat bath: it drives the particle towards an equilibrium ("rest") state, but is in turn affected negligibly by the particle.

The force fluctuations on the particle arising from this heat bath are just given by $\mathbf{f}_\perp$. The equilibrium distribution function for $\mathbf{f}_\perp$ is described by the steady-state equation

$$L_b \rho_{eq}(\mathbf{f}_\perp) = 0, \qquad (3.5)$$

giving

$$\rho_{\text{eq}}(\mathbf{f}_\perp) = \frac{1}{\sqrt{2\pi D_f}} e^{-f_\perp^2/2D_f}. \tag{3.6}$$

Assuming the crystal to be a heat bath requires that the distribution of the force fluctuations remains approximately $\rho_{\text{eq}}(\mathbf{f}_\perp)$ irrespective of the presence of the particle. In this approximation, we may eliminate the environment variable $\mathbf{f}_\perp$ from the problem altogether by projecting the evolution of the system onto a state in which the $\mathbf{f}_\perp$ distribution is always $\rho_{\text{eq}}(\mathbf{f}_\perp)$.[32-34] The large λ limit implied by the high temperature we have assumed further allows this projection to be implemented as an expansion in λ^{-1}. To lowest order in λ^{-1}, the evolution equation for the reduced probability density $\sigma(\mathbf{r}_\perp, \mathbf{p}_\perp) = \int d\mathbf{f}_\perp P(\mathbf{r}_\perp, \mathbf{p}_\perp, \mathbf{f}_\perp)$ is (see Appendix):

$$\left(\frac{\partial}{\partial t} + \frac{\mathbf{p}_\perp}{m} \cdot \frac{\partial}{\partial \mathbf{r}_\perp} - U_c'(\mathbf{r}_\perp) \cdot \frac{\partial}{\partial \mathbf{p}_\perp} \right) \sigma$$

$$= \frac{\partial}{\partial \mathbf{p}_\perp} \cdot \left(\gamma \mathbf{p}_\perp + D_p \frac{\partial}{\partial \mathbf{p}_\perp} \right) \sigma, \tag{3.7a}$$

where

$$D_p = D_f/\lambda. \tag{3.7b}$$

In the limit $\lambda \to \infty$, higher-order corrections to Eq. (3.7a) may be ignored.

Equation (3.7a) represents the Fokker-Planck equation for an essentially classical particle. It is only valid at "high" temperatures as discussed in Sec. II C. The presence of the heat bath manifests itself in the appearance of diffusion and friction terms, which are expressed as thermal averages of quantum-mechanical operators associated with the environment. The semiclassical nature of the description is evident in the presence of classical ("local") variables describing the particle, and quantum variables describing the influence of the environment on the particle. Equation (3.7a) is analogous to Kramers' generalization of the Liouville equation to include stochastic effects,[35] corresponding to the Fokker-Planck equation for a Brownian particle in a potential $U_c(\mathbf{r}_\perp)$. The difference is that γ and D_p are not related simply through the Einstein relation, reflecting the fact that Eq. (3.7a) does not emerge from usual linear-response arguments. Note that because $\xi_\perp(t)$ in Eq. (3.1c) is just a white noise source, the above equation can be obtained in the limit $\lambda \to \infty$ by the direct elimination of $\mathbf{f}_\perp$ in Eq. (3.1) using the steady-state condition $\partial_t \mathbf{f}_\perp(t) = 0$, and then transforming to a Fokker-Planck equation, giving again the above. Higher-order corrections when λ is large but finite may be found as shown in the Appendix.

Fokker-Planck equations of the above form have appeared before in dechanneling theory[5,8,12] although the above has been obtained by altogether different means. We regard $U_c(\mathbf{r}_\perp)$ as the continuum potential well known in channeling theory.[4,36] The diffusion coefficient may be written more in the form of a transport coefficient as

$$D_p = \lim_{\omega \to 0} \int_0^\infty dt e^{i\omega t} \langle \hat{f}_\mu(t) \hat{f}_\mu(0) \rangle_c. \tag{3.8}$$

This expression is valid for time scales much longer than τ_m although the equation itself is independent of time. Equation (3.7b) follows from Eq. (3.8) upon using Eq. (2.24). The equivalence of Eq. (3.8) for the diffusion coefficient to the expression given by the Ohtsuki-Nitta theory is demonstrated in Sec. V.

Equation (3.7) as it stands is still complicated to solve. In most channeling problems, the velocity of the channeling particle is high (in the Bethe-Bloch regime) and the friction coefficient γ is quite small. This is equivalent to the underdamped (i.e., low friction) limit of Brownian motion, and corresponds to when the energy of the particle changes slowly. In this case, Eq. (3.7) may be simplified by transforming the diffusion in $\mathbf{r}_\perp, \mathbf{p}_\perp$ space into a diffusion in the space of the transverse particle energy. This reduction was studied originally by Kramers[35] by transforming the equation to action-angle variables, and then averaging over the angle variable. Stratonovich[37] has also studied this problem. In the context of channeling, the transformation to an energy variable was studied by Beloshitskii and Kumakhov.[5] Their result is the energy diffusion equation written by Lindhard.[4] In principle, the underdamped limit also describes the case for very low channeling particle velocities, where the friction coefficient can also be quite small.

IV. EXPANSION OF FORCE OPERATOR

The analysis in Sec. II left us with the quantities $U_c'(\mathbf{r}_\perp)$, S_p, and D_p for which we did not obtain explicit expressions. Our aim in this section is to approximate these quantities by constructing a perturbation expansion of the force operator to low orders in the interaction potential $\hat{W}$. For the stopping power this is a reasonable procedure since a fast particle couples only weakly to the environment, so the Born approximation should be valid. Reasoning along fluctuation-dissipation lines, we may expect D_p to also be adequately expressed in a Born approximation, since D_p is the zero-frequency component of the spectral density of force fluctuations, and S_p represents the dissipation. (Note, however, that the usual fluctuation-dissipation theorem[26] does not apply to fast channeling particles.) This expectation is justified a posteriori by the good agreement observed between theory and experiment.[38,39]

A perturbation expansion of the expectation of the force operator is readily developed. For convenience we consider the time evolution of the particle-crystal system from the infinite past and use the interaction

$$\hat{W}_\eta = g e^{-\eta|t|} \hat{W}, \quad \eta \to 0^+. \tag{4.1}$$

By using this form of interaction we ignore any transient effects as the particle enters the crystal. We have also in-

troduced an expansion parameter g into Eq. (4.1) to help us keep track of powers of the coupling coefficients in the interaction. We will later set this equal to 1. The interaction here is expressed very generally, and when performing explicit calculations later in Sec. V, we will find it more useful to write the interaction in specific forms appropriate for the actual processes under consideration.

Equation (4.1) allows the force operator to be written

$$\hat{F}(t) = \hat{S}^{\dagger}(t)\overline{F}(t)\hat{S}(t), \qquad (4.2)$$

with the S matrix defined

$$\hat{S}(t) = \hat{T}\exp\left(-i\hbar^{-1}\int_{-\infty}^{t} dt'\,\overline{W}_{\eta}(t')\right), \qquad (4.3)$$

where $\hat{T}$ is the Wick time-ordering operator, and the overbar denotes an operator in the interaction picture:

$$\overline{W}_{\eta}(t) = e^{i(\hat{T}_p + \hat{H}_E)t/\hbar}\hat{W}_{\eta}e^{-i(\hat{T}_p + \hat{H}_E)t/\hbar}. \qquad (4.4)$$

Noting that the force operator may be written $\hat{F}(t) = -\partial_{\mathbf{r}}\hat{W}_{\eta}(t)$ which is first order in g, we see that the expectation of the force may be found from

$$\langle \hat{W}_{\eta}(t)\rangle = \mathrm{Tr}\,\hat{\rho}_0\hat{S}^{\dagger}(t)\overline{W}_{\eta}(t)\hat{S}(t). \qquad (4.5)$$

A compact diagrammatic expansion of this expectation is possible using the Keldysh method[40,41] (with a few simplifying assumptions concerning the environment Hamiltonian $\hat{H}_E$ to ensure that Wick's theorem works smoothly). Although we have recourse to this, the fact that we are interested only in low orders of the expansion in $\hat{W}_{\eta}$ affords a simplification which avoids any further complications. To this end we expand the S matrix to first order in g, $\hat{S}(t) \approx 1 + g\hat{S}_1(t)$, obtaining

$$\hat{W}_{\eta}(t) \approx g\hat{W}^{(1)} + g^2\hat{W}^{(2)} = \overline{W}_{\eta}(t) + 2\,\mathrm{Re}[\overline{W}_{\eta}(t)\hat{S}_1(t)].$$

$$(4.6)$$

This gives directly the low-order expansion of $\hat{F}(t)$ that we require, since from this we can extract expressions for $U'_c(\mathbf{r})$, S_p, and D_p.

We consider the matrix elements of $\hat{W}_{\eta}(t)$ with respect to the environment states $|n\rangle$, where $\hat{H}_E|n\rangle = \hbar\omega_n|n\rangle$. Diagonal matrix elements of $\hat{W}^{(1)}$ give no change of environment state, and so should contribute only to the crystal potential. To verify this, we find the expectation of $\hat{W}^{(1)}$ (putting $g = 1$):

$$\langle \hat{W}^{(1)}\rangle = \sum_{\mathbf{q}}\Phi_{\mathbf{q}}\cdot\langle \hat{\zeta}_{\mathbf{q}}\rangle_E e^{i\mathbf{q}\cdot\mathbf{r}}, \qquad (4.7)$$

where $\mathbf{r} = \langle\hat{\mathbf{r}}\rangle$, and $\langle\cdots\rangle_E$ denotes a thermal average with respect to the environment density matrix $\hat{\rho}_E$. We may show that this corresponds to the unperturbed crystal potential acting at the point $\mathbf{r}$ by using a cumulant expansion of $\langle\hat{\zeta}_{\mathbf{q}}\rangle_E$. Denote the Bravais lattice vectors by $\overline{\mathbf{R}}_j$ (which coincide with the mean lattice ion positions for the simple lattice assumed), and consider the density operator

$$\hat{\zeta}'_{\mathbf{q}} = \sum_j e^{-i\mathbf{q}\cdot\overline{\mathbf{R}}_j}e^{-i\mathbf{q}\cdot\hat{\mathbf{s}}_j}, \qquad (4.8)$$

where $\hat{\mathbf{s}}_j$ denotes either an electron position relative to $\overline{\mathbf{R}}_j$, or a lattice ion displacement, and the sum is over all electrons and lattice ions. The expectation of $e^{-i\mathbf{q}\cdot\hat{\mathbf{s}}_j}$ may be evaluated by a cumulant expansion:

$$\langle e^{-i\mathbf{q}\cdot\hat{\mathbf{s}}_j}\rangle_E = e^{W(\mathbf{q})} = \exp\left(\sum_n \frac{1}{n!}G_n(\mathbf{q})\right), \qquad (4.9)$$

where $G_n(\mathbf{q})$ are the cumulants (with $\hat{X} = -i\mathbf{q}\cdot\hat{\mathbf{s}}_j$)

$$G_1(\mathbf{q}) = \langle\hat{X}\rangle_E, \qquad (4.10a)$$

$$G_2(\mathbf{q}) = \langle\hat{X}^2\rangle_E - \langle\hat{X}\rangle_E^2, \qquad (4.10b)$$

$$G_3(\mathbf{q}) = \langle\hat{X}^3\rangle_E - 3\langle\hat{X}^2\rangle_E\langle\hat{X}\rangle_E + 2\langle\hat{X}\rangle_E^3, \qquad (4.10c)$$

and so on.[42] The quantity $W(\mathbf{q})$ comprises expectations of operators, and is independent of the index "j," so

$$\langle\hat{\zeta}'_{\mathbf{q}}\rangle_E = e^{W(\mathbf{q})}\sum_{\mathbf{G}}\delta_{\mathbf{q}-\mathbf{G}}, \qquad (4.11)$$

where $\mathbf{G}$ is a reciprocal-lattice vector. Thus $\langle\hat{W}^{(1)}\rangle$ may be written as a sum over only reciprocal-lattice points, and must correspond to the unperturbed crystal potential at a point $\mathbf{r}$. This is the lowest-order contribution to $U_c(\mathbf{r})$.

The off-diagonal matrix elements of $\hat{W}^{(1)}$ do not contribute to the trace, and so do not contribute to any expectation values. However, these matrix elements do contribute to the diffusion coefficient D_p, and hence give the random force operator to lowest order in g:

$$\hat{\mathbf{f}}^{(1)}(t) = -\sum_{n\neq n'}|n\rangle\langle n|\partial_{\mathbf{r}}\overline{W}_{\eta}(t)|n'\rangle\langle n'|. \qquad (4.12)$$

Although this does not contribute to $\mathbf{f}(t) = \langle\hat{\mathbf{f}}(t)\rangle$ directly, $\hat{\mathbf{f}}^{(1)}(t)$ affects the time evolution of $\mathbf{f}(t)$ through the strength of the fluctuations in $\xi(t)$. Note that the above expression is not to be interpreted as a solution to the Langevin equation, Eq. (2.25), which it clearly is not,

but as yielding the Born approximation for D_p when inserted into the force-force correlation function, Eq. (3.8). We use Eq. (4.12) to explicitly evaluate the diffusion coefficient in the next section.

We will ignore off-diagonal matrix elements of the second-order term $\hat{W}^{(2)}$, since these are higher-order contributions to D_p. Instead we look at the expectation,

$$\langle \hat{W}^{(2)} \rangle = 2 \, \mathrm{Re} \, \mathrm{Tr} \, \hat{\rho}_0 \overline{W}_\eta(t) \hat{S}_1(t)$$

$$= -2 \, \mathrm{Re} \, i\hbar^{-1} \sum_{nn'} p_n \int_{-\infty}^{t} dt' \langle \mathbf{K}_0 | \langle n | \overline{W}_\eta(t) | n' \rangle$$

$$\times \langle n' | \overline{W}_\eta(t') | n \rangle | \mathbf{K}_0 \rangle. \tag{4.13}$$

As before, the $n = n'$ term in this can arise only from elastic processes. Excluding this term leaves the inelastic friction force,

$$S_p = -\langle \mathbf{u} \cdot \partial_\mathbf{r} \hat{W}^{(2)} \rangle_{\mathrm{in}}, \tag{4.14}$$

where $\mathbf{u} = \mathbf{v}/v$, and "in" indicates that we exclude the $n = n'$ term in the averaging. We show in the next section that Eq. (4.14) is equivalent to the stopping power formula in the Ohtsuki-Nitta theory. The components of $-\langle \partial_\mathbf{r} \hat{W}^{(2)} \rangle_{\mathrm{in}}$ orthogonal to $\mathbf{u}$ contribute to the random force $\mathbf{f}(t)$.

V. DIFFUSION COEFFICIENTS

In this section we show that the force-force correlation function given in Eq. (3.8) reduces to the Ohtsuki-Nitta expression for the diffusion coefficient in a simple limit. The Ohtsuki-Nitta expressions are the standard expressions of the conventional quantum theory of dechanneling, and have been shown to reduce under certain conditions to the classical expressions of the Lindhard theory.[9] Using the force-force correlation function we then derive the plasmon, phonon, and core electron contributions to the diffusion coefficient. Finally, we show that Eq. (4.14) reproduces the correct stopping power formula in the Ohtsuki-Nitta theory. To facilitate the analysis, we assume that interactions between the valence electrons, phonons, and core electrons can be neglected, so the environment state vector can be factorized as

$$|n\rangle = |\psi\rangle |\phi\rangle |\chi\rangle, \tag{5.1}$$

where $|\psi\rangle$ is a many-particle state for the valence electron population, and $|\phi\rangle$ and $|\chi\rangle$ are the corresponding states for the phonon gas and the core electron population.

A. Correspondence with Ohtsuki-Nitta theory (Refs. 12 and 13)

Instead of the diffusion coefficient D_p, we consider the diffusion coefficient per unit length $D = D_p/v$, where v is the semiclassical velocity of the particle. The v arises when we substitute t in the Fokker-Planck equation by z, the distance traveled along the channeling direction, with $v = dz/dt$. D is a diagonal component of the dif-

fusion tensor. For dechanneling studies it turns out to be more useful to define a "transverse" diffusion coefficient $D_\perp$ which is the sum of the diagonal components of the diffusion tensor, since this enters directly into the Lindhard theory.[13] Thus from Eq. (3.7), to lowest order,

$$D_\perp = \frac{1}{2v} \int_{-\infty}^{\infty} dt \langle \hat{\mathbf{f}}_\perp(t) \cdot \hat{\mathbf{f}}_\perp(0) \rangle, \tag{5.2}$$

with Eq. (4.12) defining the random force operator

$$\hat{\mathbf{f}}_\perp(t) = -i \sum_\mathbf{q} \sum_{n \neq n'} \mathbf{q}_\perp e^{i\hat{H}_0 t/\hbar} |n\rangle H_{nn'}(\mathbf{q})$$

$$\times e^{i\mathbf{q} \cdot \hat{\mathbf{r}}} \langle n' | e^{-i\hat{H}_0 t/\hbar}, \tag{5.3a}$$

$$H_{nn'}(\mathbf{q}) = \langle n | \Phi_\mathbf{q} \cdot \hat{\zeta}_\mathbf{q} | n' \rangle, \tag{5.3b}$$

where $\hat{H}_0 = \hat{T}_p + \hat{H}_E$. Inserting this into Eq. (5.2) gives

$$D_\perp = -\frac{1}{2v} \int_{-\infty}^{\infty} dt \sum_{\mathbf{q}\mathbf{q}'} \mathbf{q}_\perp \cdot \mathbf{q}'_\perp \Phi_\mathbf{q} \Phi_{\mathbf{q}'}$$

$$\times K_{\mathrm{in}}(\mathbf{q}, -\mathbf{q}') J(\mathbf{q}, \mathbf{q}'), \tag{5.4a}$$

$$K_{\mathrm{in}}(\mathbf{q}, -\mathbf{q}') = \langle \hat{\zeta}_\mathbf{q}(t) \hat{\zeta}^\dagger_{-\mathbf{q}'}(0) \rangle_{\mathrm{in}}, \tag{5.4b}$$

$$J(\mathbf{q}, \mathbf{q}') = \langle \mathbf{K}_0 | e^{i\mathbf{q} \cdot \hat{\mathbf{r}}(t)} e^{i\mathbf{q}' \cdot \hat{\mathbf{r}}(0)} | \mathbf{K}_0 \rangle, \tag{5.4c}$$

where

$$\hat{\zeta}_\mathbf{q}(t) = e^{i\hat{H}_E t/\hbar} \hat{\zeta}_\mathbf{q} e^{-i\hat{H}_E t/\hbar}, \tag{5.4d}$$

$$\hat{\mathbf{r}}(t) = e^{i\hat{T}_p t/\hbar} \hat{\mathbf{r}} e^{-i\hat{T}_p t/\hbar}. \tag{5.4e}$$

The scalar product $\Phi_\mathbf{q} \cdot \hat{\zeta}_\mathbf{q}$ is implicit in the above. $K_{\mathrm{in}}(\mathbf{q}, -\mathbf{q}')$ is a density-density correlation function, and $\langle \cdots \rangle_{\mathrm{in}}$ is defined

$$\langle \hat{A}\hat{B} \rangle_{\mathrm{in}} = \sum_{n \neq n'} p_n \langle n | \hat{A} | n' \rangle \langle n' | \hat{B} | n \rangle, \tag{5.5}$$

with $p_n = e^{-\beta\hbar\omega_n} / \mathrm{Tr} \, e^{-\beta\hat{H}_E}$ being the statistical weight of an environment state $|n\rangle$, and $\hat{H}_E |n\rangle = \hbar\omega_n |n\rangle$. Writing $K_{\mathrm{in}}(\mathbf{q}, -\mathbf{q}')$ in real space as $K_{\mathrm{in}}(\mathbf{r}, \mathbf{r}')$, we note that it has the following invariance properties:

$$K_{\mathrm{in}}(\mathbf{r} + \mathbf{a}_z, \mathbf{r}' + \mathbf{a}_z) = K_{\mathrm{in}}(\mathbf{r}, \mathbf{r}'), \tag{5.6a}$$

$$K_{\mathrm{in}}(\mathbf{r} + \mathbf{a}_\perp, \mathbf{r}' + \mathbf{a}_\perp) = K_{\mathrm{in}}(\mathbf{r}, \mathbf{r}'), \tag{5.6b}$$

where $\mathbf{a}_z$ is an arbitrary vector pointing along the channeling direction (recall the continuum potential), and $\mathbf{a}_\perp$ is a lattice vector pointing transverse to the channeling direction. With $\mathbf{q} = (\mathbf{q}_\perp, q_z)$ and $\mathbf{q}' = (\mathbf{q}'_\perp, q'_z)$, Eqs. (5.6) give

$$\mathbf{q}_\perp + \mathbf{q}'_\perp = \mathbf{g}, \quad q_z + q'_z = 0, \tag{5.7}$$

where $\mathbf{g}$ is a reciprocal-lattice vector such that $\mathbf{a}_z \cdot \mathbf{g} = 0$. This notation is appropriate for axial channeling, but is easily modified to describe planar channeling. Now

$$\hat{T}_p|\mathbf{K}\rangle = E_{\mathbf{K}}|\mathbf{K}\rangle, \quad E_{\mathbf{K}} = \hbar^2 K^2/2m \qquad (5.8)$$

for an arbitrary particle state $|\mathbf{K}\rangle$. Inserting a decomposition of unity into Eq. (5.4c), and with $e^{i\mathbf{q}\cdot\hat{\mathbf{r}}}|\mathbf{K}\rangle = |\mathbf{K}+\mathbf{q}\rangle$,

$$J(\mathbf{q},\mathbf{q}') = \sum_{\mathbf{K}} e^{i(E_{\mathbf{K}_0} - E_{\mathbf{K}})t/\hbar}$$
$$\times \langle \mathbf{K}_0|\mathbf{K}+\mathbf{q}\rangle\langle \mathbf{K}+\mathbf{q}|e^{i\mathbf{g}\cdot\hat{\mathbf{r}}}|\mathbf{K}_0\rangle. \qquad (5.9)$$

Using the semiclassical substitution $\hat{\mathbf{r}} \to \mathbf{r}$, the diffusion coefficient may then be written,

$$D_\perp = \frac{1}{2v} \int \frac{d^3q}{(2\pi)^3} \sum_{\mathbf{g}} \hbar^2 \mathbf{q}_\perp \cdot (\mathbf{q}_\perp - \mathbf{g})\Gamma(\mathbf{q},\Omega)e^{i\mathbf{g}\cdot\mathbf{r}},$$
$$(5.10a)$$

$$\Gamma(\mathbf{q},\Omega) = \frac{2\pi}{\hbar} \sum_{n\neq n'} p_n H_{nn'}(\mathbf{q})H_{n'n}(-\mathbf{q}+\mathbf{g})$$
$$\times \delta(\hbar\Omega - \hbar\omega_{n'n}), \qquad (5.10b)$$

where the $\mathbf{q}$ summation has been approximated by an integral, $\omega_{n'n} = \omega_{n'} - \omega_n$, and $\hbar\Omega = E_{\mathbf{K}_0} - E_{\mathbf{K}_0-\mathbf{q}}$ is the energy lost by the particle. Finally, taking the zero-temperature limit of $\Gamma(\mathbf{q},\Omega)$, i.e., $p_n \to \delta_{n0}$, making a change of variables $\mathbf{q} \to -\mathbf{q}+\frac{1}{2}\mathbf{g}$, and approximating the $\mathbf{g}$ summation by an integral gives Nitta's expression for the diffusion coefficient,[12,43]

$$D_\perp = \frac{1}{2v} \int \frac{d^3q}{(2\pi)^3} \int \frac{d^2g}{(2\pi)^2} \hbar^2(q_\perp^2 - \tfrac{1}{4}g^2)$$
$$\times \sum_{n\neq 0} S_n(\mathbf{q}-\tfrac{1}{2}\mathbf{g}, \mathbf{q}+\tfrac{1}{2}\mathbf{g})e^{i\mathbf{g}\cdot\mathbf{r}}, \qquad (5.11)$$

where

$$S_n(\mathbf{Q},\mathbf{Q}') = \frac{2\pi}{\hbar} H_{0n}(-\mathbf{Q})H_{n0}(\mathbf{Q}')\delta(\hbar\Omega - \hbar\omega_{n0}) \quad (5.12)$$

is Ohtsuki's inelastic-scattering factor.[7,44] $\Gamma(\mathbf{q},\Omega)$ is the finite-temperature equivalent of this inelastic-scattering factor summed over n.

Note that from Eq. (5.10) we may also write the diffusion coefficient in the alternative form,

$$D_\perp = \frac{1}{2v} \int \frac{d^3q}{(2\pi)^3} \sum_{\mathbf{g}} \mathbf{q}_\perp \cdot (\mathbf{q}_\perp - \mathbf{g})\Phi_{\mathbf{q}}\Phi_{-\mathbf{q}+\mathbf{g}}$$
$$\times S_{\text{in}}(\mathbf{q},\Omega)e^{i\mathbf{g}\cdot\mathbf{r}}, \qquad (5.13)$$

where

$$S_{\text{in}}(\mathbf{q},\Omega) = \int_{-\infty}^{\infty} dt\, e^{i\Omega t} \langle \hat{\zeta}_{\mathbf{q}}(t)\hat{\zeta}_{\mathbf{q}-\mathbf{g}}^\dagger(0)\rangle_{\text{in}} \qquad (5.14)$$

becomes the usual dynamic structure factor for density-density correlations, $S(\mathbf{q},\Omega)$, when $\mathbf{g} = 0$ and $\langle\cdots\rangle_{\text{in}}$ is replaced by an ordinary thermal average. This forms a more convenient starting point for some of the calculations below.

B. Plasmon contribution

Approximating the valence electrons by a homogeneous free-electron gas we write[45]

$$H_{nn'}(\mathbf{q}) = -\langle \psi_n|V_{\mathbf{q}}\hat{\rho}_{\mathbf{q}}|\psi_{n'}\rangle, \qquad (5.15a)$$

$$V_{\mathbf{q}} = 4\pi e^2/q^2, \qquad (5.15b)$$

$$\hat{\rho}_{\mathbf{q}} = \sum_{\mathbf{k}\sigma} \hat{c}_{\mathbf{k}+\mathbf{q}\sigma}^\dagger \hat{c}_{\mathbf{k}\sigma}, \qquad (5.15c)$$

where the $|\psi_n\rangle$ are many-particle (Fock) states of the electron gas, and $\hat{c}_{\mathbf{k}\sigma}^\dagger$ and $\hat{c}_{\mathbf{k}\sigma}$ are creation and annihilation operators for states with momentum $\mathbf{k}$ and spin σ. For a homogeneous system only the $\mathbf{g} = 0$ term contributes to the $\mathbf{g}$ summation in Eq. (5.13), so the transverse diffusion coefficient is given by

$$D_\perp = \frac{1}{2v} \int \frac{d^3q}{(2\pi)^3} q_\perp^2 V_{\mathbf{q}}^2 S(\mathbf{q},\omega), \qquad (5.16)$$

where

$$S(\mathbf{q},\omega) = \int_{-\infty}^{\infty} dt\, e^{i\omega t} \langle \hat{\rho}_{\mathbf{q}}(t)\hat{\rho}_{\mathbf{q}}^\dagger(0)\rangle \qquad (5.17)$$

is the dynamic structure factor at finite temperature, and

$$\omega = \Omega \approx K_0 v = q_z v, \quad \hbar q \ll \hbar K_0. \qquad (5.18)$$

The summation in $\langle\cdots\rangle_{\text{in}}$ has been extended to include $n = n'$ since $\langle\psi_n|\rho_{\mathbf{q}}|\psi_n\rangle = 0$, so the angular brackets in Eq. (5.17) denote an ordinary thermal average. Also the momentum transfer in the scattering process, $\hbar q$, is assumed to be negligible compared to the initial momentum of the particle, $\hbar K_0$.

Approximating $S(\mathbf{q},\omega)$ by its zero-temperature, long-wavelength limit where the contributions to the polarization of the crystal are solely from plasmon excitations,[46]

$$S(\mathbf{q},\omega) \approx \frac{\pi\hbar\omega_p}{V_{\mathbf{q}}}\delta(\omega - \omega_p), \qquad (5.19)$$

where ω_p is the plasma frequency. Inserting this into Eq. (5.16) with $\omega = q_z v$,

$$D_\perp = \frac{e^2\hbar\omega_p}{4v} \left\{ q_c^2 - \left(\frac{\omega_p}{v}\right)^2 \ln\left[\left(\frac{q_c v}{\omega_p}\right)^2 + 1\right] \right\}, \qquad (5.20)$$

where $q_c = \omega_p/v_F$ is a wave-vector cutoff,[46] with v_F being the Fermi velocity. This formula was derived originally by Kitagawa and Ohtsuki.[8] The plasmon contribution to the diffusion coefficient is generally not significant and is usually ignored.

C. Phonon contribution

In the harmonic approximation, the lattice ion displacement $\hat{\mathbf{u}}_i = \hat{\mathbf{R}}_i - \overline{\mathbf{R}}_i$ is expanded to linear order in

phonon creation and annihilation operators. Thus[47]

$$H_{nn'}(\mathbf{q}) = \langle \phi_n | U_\mathbf{q} \hat{\eta}_\mathbf{q} | \phi_{n'} \rangle, \tag{5.21a}$$

$$\hat{\eta}_\mathbf{q}(t) = \sum_j e^{-i\mathbf{q}\cdot\mathbf{R}_j} e^{-i\mathbf{q}\cdot\hat{\mathbf{u}}_j(t)}, \tag{5.21b}$$

$$\hat{\mathbf{u}}_j(t) = \sum_{\mathbf{k}\lambda} \left(\frac{\hbar}{2NM\omega_{\mathbf{k}\lambda}} \right)^{1/2} \mathbf{e}_{\mathbf{k}\lambda}(\hat{a}_{\mathbf{k}\lambda} e^{-i\omega_{\mathbf{k}\lambda}t}$$
$$+ \hat{a}_{-\mathbf{k}\lambda}^\dagger e^{i\omega_{\mathbf{k}\lambda}t}) e^{i\mathbf{k}\cdot\mathbf{R}_j}, \tag{5.21c}$$

where N and M are the number of ions in the crystal and their mass (a monatomic basis is assumed), and $\hat{a}_{\mathbf{k}\lambda}^\dagger$ and $\hat{a}_{\mathbf{k}\lambda}$ are creation and annihilation operators for phonon states of wave vector $\mathbf{k}$, mode λ, energy $\hbar\omega_{\mathbf{k}\lambda}$, and polarization $\mathbf{e}_{\mathbf{k}\lambda}$. $|\phi_n\rangle$ are Fock states for the phonon system. The polarization vector satisfies $\mathbf{e}_{\mathbf{k}\lambda}^* = \mathbf{e}_{-\mathbf{k}\lambda}$.

Recall that the definition of the diffusion coefficient $D_\perp$ involves only off-diagonal matrix elements. This corresponds to the inclusion of only inelastic processes in the evaluation of $S_{\text{in}}(\mathbf{q},\omega)$. However, it turns out to be easier to first compute $S(\mathbf{q},\omega)$, and then find $S_{\text{in}}(\mathbf{q},\omega)$ by subtracting off the elastic-scattering contribution. This means we do the calculation with diagonal matrix elements initially included, and then subsequently subtract out their contribution.

Thus we start by finding

$$S(\mathbf{q},\omega) = \int_{-\infty}^{\infty} dt\, e^{i\omega t} \langle \hat{\eta}_\mathbf{q}(t) \hat{\eta}_{\mathbf{q}-\mathbf{g}}^\dagger(0) \rangle. \tag{5.22}$$

Inserting Eq. (5.21b), the density-density correlation function in Eq. (5.22) becomes

$$K(\mathbf{q},\mathbf{q}-\mathbf{g}) = \sum_{jl} e^{-i\mathbf{q}\cdot\mathbf{R}_j} e^{i(\mathbf{q}-\mathbf{g})\cdot\mathbf{R}_l}$$
$$\times \langle e^{-i\mathbf{q}\cdot\hat{\mathbf{u}}_j(t)} e^{i(\mathbf{q}-\mathbf{g})\cdot\hat{\mathbf{u}}_l(0)} \rangle. \tag{5.23}$$

We use the Einstein approximation wherein each lattice ion is assumed to vibrate independently of all others. Anharmonicities may be treated by using a cumulant expansion of the expectation in Eq. (5.23), although to lowest order this just gives the Einstein approximation again.[2,47] If we neglect anharmonic effects from the outset, we may evaluate $K(\mathbf{q},\mathbf{q}-\mathbf{g})$ as follows. For operators $\hat{A}$ and $\hat{B}$ which both commute with $[\hat{A},\hat{B}]$, the Baker-Campbell-Hausdorff formula reduces to

$$e^{\hat{A}} e^{\hat{B}} = e^{\hat{A}+\hat{B}} e^{[\hat{A},\hat{B}]/2}. \tag{5.24}$$

In the Einstein approximation $\hat{\mathbf{u}}_j(t)$ and $\hat{\mathbf{u}}_j(0)$ are easily shown to satisfy the requirement for Eq. (5.24) to hold. Then Eq. (5.24) together with

$$\langle \hat{a}_{\mathbf{k}\lambda}^\dagger \hat{a}_{\mathbf{k}'\lambda'} \rangle = \delta_{\mathbf{k}\mathbf{k}'}\delta_{\lambda\lambda'}(e^{\beta\hbar\omega_{\mathbf{k}\lambda}} - 1)^{-1}, \tag{5.25}$$

gives

$$K(\mathbf{q},\mathbf{q}-\mathbf{g}) = Ne^{-W_\mathbf{q} - W_{\mathbf{q}-\mathbf{g}} + 2W(\mathbf{q},\mathbf{q}-\mathbf{g})}, \tag{5.26a}$$

$$W_\mathbf{q} = \sum_{\mathbf{k}\lambda} \frac{\hbar}{4NM\omega_{\mathbf{k}\lambda}} |\mathbf{q}\cdot\mathbf{e}_{\mathbf{k}\lambda}|^2 \coth \tfrac{1}{2}\beta\hbar\omega_{\mathbf{k}\lambda}, \tag{5.26b}$$

$$2W(\mathbf{q},\mathbf{q}-\mathbf{g}) = \sum_{\mathbf{k}\lambda} y_{\mathbf{k}\lambda} \cosh \omega_{\mathbf{k}\lambda}(\tfrac{1}{2}\beta\hbar - it), \tag{5.26c}$$

$$y_{\mathbf{k}\lambda} = \frac{\hbar\mathbf{q}\cdot\mathbf{e}_{\mathbf{k}\lambda}(\mathbf{q}-\mathbf{g})\cdot\mathbf{e}_{\mathbf{k}\lambda}^*}{2NM\omega_{\mathbf{k}\lambda} \sinh \tfrac{1}{2}\beta\hbar\omega_{\mathbf{k}\lambda}}. \tag{5.26d}$$

$W_\mathbf{q}$ is the Debye-Waller factor, and $W(\mathbf{q},\mathbf{q}) \to W_\mathbf{q}$ when $t \to 0$.

Now using a standard trick in the Einstein approximation[47,48]

$$e^{y\cosh x} = \sum_{n=-\infty}^{\infty} e^{nx} I_n(y), \tag{5.27}$$

where $I_n(y)$ is a modified Bessel function of the first kind, gives

$$S(\mathbf{q},\omega) = 2\pi Ne^{-W_\mathbf{q} - W_{\mathbf{q}-\mathbf{g}}} e^{\beta\hbar\omega/2}$$
$$\times \prod_{\mathbf{k}\lambda} \sum_{n=-\infty}^{\infty} I_n(y_{\mathbf{k}\lambda})\delta(\omega - n\omega_{\mathbf{k}\lambda}). \tag{5.28}$$

The term proportional to $\delta(\omega)$ in this expansion, i.e., the $n = 0$ term, is the elastic-scattering contribution, and must be subtracted off to obtain $S_{\text{in}}(\mathbf{q},\omega)$. For a large crystal $N \to \infty$, so $y_{\mathbf{k}\lambda} \to 0$. Now

$$I_n(y) = \sum_{r=0}^{\infty} \frac{(y/2)^{n+2r}}{r!\Gamma(r+n+1)}, \tag{5.29}$$

so as $y \to 0$, retaining only the $r = 0$ term in the expansion,

$$I_n(y) \approx \frac{1}{n!} \left(\frac{y}{2} \right)^n. \tag{5.30}$$

The larger the n, the faster this tends to zero. This shows that for large crystals only processes involving a small number of phonons (i.e., small n) are important. Since the phonon energies are negligible compared to the energy of the channeling particle we may therefore make the replacement $\delta(\omega - n\omega_{\mathbf{k}\lambda}) \approx \delta(\omega)$. Then using Eq. (5.27) again,

$$S(\mathbf{q},\omega) \approx 2\pi Ne^{-W_\mathbf{g}}\delta(\omega), \tag{5.31}$$

from which

$$S_{\text{in}}(\mathbf{q},\omega) = 2\pi N \left[e^{-W_\mathbf{g}} - e^{-W_\mathbf{q} - W_{\mathbf{q}-\mathbf{g}}} \prod_{\mathbf{k}\lambda} I_0(y_{\mathbf{k}\lambda}) \right] \delta(\omega). \tag{5.32}$$

For $N \to \infty$ we have $I_0(y_{\mathbf{k}\lambda}) \to 1$. Inserting $S_{\text{in}}(\mathbf{q},\omega)$ into Eq. (5.13), changing variables $\mathbf{q} \to -\mathbf{q} + \tfrac{1}{2}\mathbf{g}$, and doing the q_z integral,

$$D_\perp = \frac{N}{2v^2} \int \frac{d^2\mathbf{q}_\perp}{(2\pi)^2} \sum_\mathbf{g} (q_\perp^2 - \tfrac{1}{4}g^2) U_{-\mathbf{q}_\perp + \mathbf{g}/2} U_{\mathbf{q}_\perp + \mathbf{g}/2}$$

$$\times \left[e^{-W_\mathbf{g}} - e^{-W_{-\mathbf{q}_\perp + \mathbf{g}/2}} e^{-W_{\mathbf{q}_\perp + \mathbf{g}/2}} \right] e^{i\mathbf{g} \cdot \mathbf{r}}, \quad (5.33)$$

which is Nitta's result.[12] Nitta,[12] and Ohtsuki and Nitta[13] show that this expression reduces to the Lindhard formula for single phonon scattering[4] when $|\mathbf{r}_\perp|$ is large, and to the Kitagawa-Ohtsuki formula[8] when $|\mathbf{r}_\perp|$ is small. The preceding analysis is different from that usually given, and reveals additional steps required to justify the final result.

D. Core electron contribution

We describe the core electrons as a Slater determinant of single particle Hartree-Fock orbitals $|\alpha\rangle$. In a tight-binding approximation, the overlap of orbitals centered on adjacent atoms is ignored. The core electron density operator is

$$\hat{\rho}_\mathbf{q} = \sum_{j=1}^N \sum_{l=1}^{N_e} e^{-i\mathbf{q} \cdot (\overline{\mathbf{R}}_j + \hat{\mathbf{s}}_l)}, \quad (5.34)$$

where the j and l summations are over the lattice ion and core electron positions, N and N_e are the number of lattice ions and the number of electrons on a lattice ion, and $\hat{\mathbf{s}}_l$ is the position of the lth electron relative to the lattice ion position $\overline{\mathbf{R}}_j$. Since the core electron excitation energies are generally large compared to the thermal energy β^{-1} but small compared to the energy of the channeling particle, we approximate $|\chi_n\rangle$ by the ground state $|\chi_0\rangle$, and put $\delta(\Omega - \omega_{nn'}) \approx \delta(\Omega)$. Then the diffusion coefficient may be written

$$D_\perp = \frac{1}{2v} \int \frac{d^3\mathbf{q}}{(2\pi)^2} \sum_\mathbf{g} \mathbf{q}_\perp \cdot (\mathbf{q}_\perp - \mathbf{q}) V_\mathbf{q} V_{-\mathbf{q} + \mathbf{g}}$$

$$\times \langle \chi_0 | \hat{\rho}_\mathbf{q}^{in} \hat{\rho}_{-\mathbf{q} + \mathbf{g}}^{in} | \chi_0 \rangle e^{i\mathbf{g} \cdot \mathbf{r}} \delta(\Omega), \quad (5.35)$$

where "in" denotes that only off-diagonal matrix elements are taken. The ground state $|\chi_0\rangle$ may be written as the Slater determinant

$$\langle \mathbf{r}_1 \mathbf{r}_2 \cdots | \chi_0 \rangle = \frac{1}{\sqrt{N_e!}} \det |\langle \mathbf{r}_j | \alpha \rangle|, \quad (5.36)$$

whence

$$D_\perp = \frac{N}{2v^2} \int \frac{d^2\mathbf{q}_\perp}{(2\pi)^2} \sum_\mathbf{g} (q_\perp^2 - \tfrac{1}{4}g^2)$$

$$\times V_{-\mathbf{q}_\perp + \frac{1}{2}\mathbf{g}} V_{\mathbf{q}_\perp + \frac{1}{2}\mathbf{g}} \left[\sum_\alpha f_{\alpha\alpha}(\mathbf{g}) \right.$$

$$\left. - \sum_{\alpha\beta} f_{\alpha\beta}(-\mathbf{q}_\perp + \tfrac{1}{2}\mathbf{g}) f_{\beta\alpha}(\mathbf{q}_\perp + \tfrac{1}{2}\mathbf{g}) \right] e^{i\mathbf{g} \cdot \mathbf{r}}, \quad (5.37)$$

where $f_{\alpha\beta}(\mathbf{q}) = \langle \alpha | e^{-i\mathbf{q} \cdot \hat{\mathbf{s}}} | \beta \rangle$ is the Compton scattering factor.[2] Nitta, Ohtsuki, and Kubo[49] show that in a par-

ticular limit, this expression reduces to the Lindhard formula for the contribution of close electron scattering.

E. Stopping power

Here we express Eq. (4.14) for S_p in a form recognizable as the stopping power. The identity

$$\lim_{\eta \to 0^+} \frac{1}{\omega - i\eta} = \mathcal{P} \frac{1}{\omega} + i\pi\delta(\omega), \quad (5.38)$$

where $\mathcal{P}$ denotes the principle part, allows Eq. (4.14) to be written as

$$S_p = -\frac{1}{v} \int_{-\infty}^\infty dt \sum_{\mathbf{q}\mathbf{q}'} \mathbf{v} \cdot \mathbf{q} \Phi_\mathbf{q} \Phi_{\mathbf{q}'} K_{in}(\mathbf{q}, -\mathbf{q}') J(\mathbf{q}, \mathbf{q}').$$

$$(5.39)$$

$K_{in}(\mathbf{q}, -\mathbf{q}')$ and $J(\mathbf{q}, \mathbf{q}')$ are defined in Eq. (5.4). The arguments leading to Eq. (5.10) then give

$$S_p = -\frac{1}{v} \int \frac{d^3\mathbf{q}}{(2\pi)^3} \sum_\mathbf{g} \hbar \mathbf{v} \cdot \mathbf{q} \Gamma(\mathbf{q}, \Omega) e^{i\mathbf{g} \cdot \mathbf{r}}, \quad (5.40)$$

with $\Gamma(\mathbf{q}, \Omega)$ as in Eq. (5.10b). From Eq. (5.18) we may write for a fast particle

$$\hbar \mathbf{v} \cdot \mathbf{q} \approx \hbar\Omega = \hbar(\omega_{n'} - \omega_n), \quad (5.41)$$

whence in the zero-temperature limit, and putting $E_n = \hbar\omega_n$,

$$S_p = \frac{1}{v} \int \frac{d^3\mathbf{q}}{(2\pi)^3} \sum_\mathbf{g} \sum_{n \neq 0} (E_n - E_0)$$

$$\times S_n(\mathbf{q} - \tfrac{1}{2}\mathbf{g}, \mathbf{q} + \tfrac{1}{2}\mathbf{g}) e^{i\mathbf{g} \cdot \mathbf{r}} \quad (5.42)$$

which is the formula for the stopping power in the Ohtsuki-Nitta theory.[9] $S_n(\mathbf{Q}, \mathbf{Q}')$ is Ohtsuki's inelastic-scattering factor defined in Eq. (5.12). The similarity in structure between the formulas for $D_\perp$ and S_p [Eqs. (5.10) and (5.40)] suggests that some form of fluctuation-dissipation result should hold, but this will not be of the usual form since we are not in the linear-response regime. We do not examine this point further here.

VI. CONCLUSIONS

A formulation of dechanneling theory based on a particle in a heat-bath approach has been presented. Rather than considering a density matrix as has been done in the past,[9] we started instead from the Heisenberg equations for the particle position and momentum operators. This has the great utility of allowing ready visualization of the problem in the semiclassical limit of these equations. It was argued that the force operator could be separated into a component arising from the crystal potential, a friction component arising by virtue of the particle motion, and a random component independent of the particle motion. It was further argued that at high tempera-

tures and for a fast particle, the Heisenberg equation for the random component effectively becomes a Langevin equation. A Fokker-Planck equation was then obtained, from which the force variable was eliminated by regarding the crystal as a heat bath. The resulting reduced Fokker-Planck equation allows various quantities of interest in dechanneling theory to be calculated.

This Fokker-Planck equation is valid only for a heavy particle, and at temperatures typically above a few Kelvin, where coherent quantum processes in the environment have very short lifetimes. Much of the quantum-mechanics in the initial Heisenberg equations is not present in the final theory. For a heavy channeling particle, quantum-mechanical corrections to its behavior are small, so a semiclassical approximation allows the particle to be treated essentially classically. The high-temperature condition simplifies the otherwise unsolvable particle-environment interaction, allowing only an effective Markov limit of this interaction to be retained. This limit also allows explicit reference to the environment to be removed from the problem altogether. The final contracted description of the problem thus achieves an effective decoupling of the dynamics of the particle from that of the environment. The dynamics of the particle is governed by an essentially classical stochastic equation, which is what we are primarily interested in. However the environment is still governed by quantum dynamics, and although the corresponding equations do not appear explicitly in the final theory, the quantum nature of the dynamics is nevertheless implicit in the parameters describing the influence of the environment on the particle, namely the friction and diffusion coefficients. Indeed, these parameters are expressed in terms of thermal averages of environment operators [see Eqs. (5.10) and (5.40)], clearly showing the continued quantum dynamical nature of the environment, despite the appearance of the parameters in a classical equation. The heat-bath approximation tells us that we are in effect neglecting the influence of the particle on the dynamics of the environment which is assumed to be in a state of equilibrium always.

The point of view used in this work has led to some useful insights into the dechanneling problem. The analysis clarifies how the Lindhard theory treats the environment. In particular, only the Markov limit of the quantum properties of the environment are retained: the electron and lattice ion density fluctuations are assumed to have an infinitesimal lifetime, and memory effects are ignored. The transverse diffusion coefficient in this limit may be written in a very compact form as the zero-frequency component of the spectral density of random force fluctuations, clearly showing the neglect of memory information. An expansion of the force operator to low orders is sufficient to yield expressions for both the stopping power and the diffusion coefficient that are finite-temperature generalizations of standard expressions. We also remark that the damping coefficient in the reduced Fokker-Planck equation obtained here is related very simply to the stopping power S_p, which should be expected on physical grounds. In other theories, such a relationship is much less transparent.

There are several points we did not touch upon in the analysis. Since the arguments have assumed a strongly localized (semiclassical) channeling particle, the theory is generally only valid for heavy ions, ignoring ionization effects. Delocalization of the particle wave packet, such as would occur for electrons or positrons, is usually treated within a density-matrix framework, although then we lose the physical picture of particle trajectories, so useful in the above analysis. Such delocalization might, for example, lead to quantization of the transverse motion of the particle, which will introduce modifications to the theory. Since our effort has been spent in developing a particular viewpoint of the dechanneling process, we have also not expanded the discussion to include the very low-energy regime, where ionization processes are important, or to the very high-energy regime, where relativistic contributions are important. However, we expect the underlying heat-bath concept to be valid in these limits too.

For relativistic energies, the only significant modification we expect is an additional contribution to the transverse diffusion coefficient due to photon emission.[13] The low-energy regime is more difficult to analyze since the Born approximation is not applicable to low velocity particles. This has hitherto precluded any reliable extension of the Lindhard theory to this regime. However, the analogy to Brownian motion that we have consistently utilized offers a possible way forward. At low energies the stopping power is linear in velocity, given by Firsov's phenomenological expression.[50] For linear dissipation, postulating a fluctuation-dissipation theorem to hold (although we have not proved this) allows us immediately to express the transverse diffusion coefficient in terms of the friction coefficient using Einstein's relation. As a complication, we cannot easily neglect random elastic scattering at low energies as we did when discussing the quantum force operator in Sec. II. Such scattering is caused by the deviation from the continuum Lindhard potential of the effective potential felt by the particle, and its effect may be approximated by an additional contribution to the transverse diffusion coefficient. These observations allow us to write a Fokker-Planck equation for low-energy channeling particles entirely in keeping with the heat-bath concept. We will not discuss this procedure or its validity further here.

The environment used in this work is considerably more complicated than those found in typical heat-bath problems. There is no obvious way of expressing the particle-electron and particle-lattice interactions in a form suitable for exact functional integration. This makes the general problem of a quantum particle interacting with a quantum environment difficult to solve. However, in the semiclassical limit we have shown that progress can be made in a rather straightforward manner from the system Heisenberg equations, provided a separation of time scales argument can be applied to isolate a rapidly fluctuating component of the force acting on the particle. Without this rapidly fluctuating force, we have simply the Ehrenfest equations. For the dechanneling problem, a separation of time scales is possible at high temperatures and when the friction force on the particle changes only slowly. In this case the system Heisen-

berg equations can be written in the form of tractable Langevin equations.

From these equations, it is of course possible to write down a path-integral representation of the probability density function. This will only describe a semiclassical particle in the Markov limit of the original environment. In standard path-integral treatments with idealized heat baths, phenomenological parameters are often introduced to make contact with experiment. By contrast, in the treatment presented here, details of the original environment appear in the microscopic definitions of the friction force and diffusion coefficient. The friction force determines the dissipation present in the particle-environment interaction, and may be either linear or nonlinear. The analysis suggests that the high-temperature Markov limit of a complex environment may be described by an effective heat bath of simple harmonic oscillator degrees of freedom (boson heat bath) with parameters determined from the original environment parameters. Such a description is much simpler than the path integral for the full density matrix obtained from the original particle-environment interaction, and makes the present analysis relevant to the study of very general particle-environment systems.

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APPENDIX: ELIMINATION OF BATH VARIABLE

The object is to eliminate the force variable $\mathbf{f}_\perp$ from the Fokker-Planck equation

$$\frac{\partial}{\partial t} P(\eta, \mathbf{f}_\perp, t) = (L_a + L_i + \lambda L_b) P(\eta, \mathbf{f}_\perp, t), \quad \text{(A1a)}$$

$$P(\eta, \mathbf{f}_\perp, 0) = \rho_{eq}(\mathbf{f}_\perp) \sigma(\eta, 0), \quad \text{(A1b)}$$

with L_a, L_b, and L_i given in Eq. (3.3), and η denoting $(\mathbf{r}_\perp, \mathbf{p}_\perp)$. Also $\rho_{eq}(\mathbf{f}_\perp)$ is the equilibrium force distribution given in Eq. (3.6). The crystal is taken to be in thermal equilibrium before the entry of the particle, so the initial condition assumes the factorized form shown. We want to obtain a reduced evolution equation for the particle distribution function $\sigma(\eta, t)$. A number of elimination schemes designed to handle such a reduction are available, including Laplace transform methods,[30,51] a Chapman-Enskog expansion,[32] and cumulant expansions,[33,34] amongst others. In the present case a cumulant expansion of the reduced evolution operator is readily developed, and is used below.

To effect the reduction we use the projection operator $\mathcal{P}$:

$$\mathcal{P}g(\eta, \mathbf{f}_\perp, t) = \rho_{eq}(\mathbf{f}_\perp) \int d\mathbf{f}_\perp g(\eta, \mathbf{f}_\perp, t). \quad \text{(A2)}$$

A useful representation of $\mathcal{P}$ may be obtained from the Fokker-Planck equation $\partial_t \rho(\mathbf{f}_\perp, t) = L_b \rho(\mathbf{f}_\perp, t)$. This has an operator solution $\rho(\mathbf{f}_\perp, t) = e^{L_b t} \rho(\mathbf{f}_\perp, 0)$. L_b is dissipative (it has only negative eigenvalues), so $\rho(\mathbf{f}_\perp, t)$ decays to an equilibrium distribution $\rho_{eq}(\mathbf{f}_\perp)$ as $t \to \infty$. Using this, together with $\mathcal{P}\rho(\mathbf{f}_\perp, 0) = \rho_{eq}(\mathbf{f}_\perp)$, gives the operator identity

$$\mathcal{P} = \lim_{t \to \infty} e^{L_b t}. \quad \text{(A3)}$$

$\mathcal{P}$ is now readily shown to satisfy

$$[\mathcal{P}, L_a] = 0, \quad \text{(A4a)}$$

$$\mathcal{P}e^{\lambda L_b t} = e^{\lambda L_b t}\mathcal{P} = \mathcal{P}. \quad \text{(A4b)}$$

It is convenient to transform Eq. (A1) using

$$\overline{P}(\eta, \mathbf{f}_\perp, t) = e^{-\lambda L_b t} P(\eta, \mathbf{f}_\perp, t). \quad \text{(A5)}$$

Taking the time derivative of $\overline{P}(\eta, \mathbf{f}_\perp, t)$, and using Eq. (A1) gives the evolution equation,

$$\frac{\partial}{\partial t}\overline{P}(\eta, \mathbf{f}_\perp, t) = \left[L_a + \overline{L}_i(t)\right] \overline{P}(\eta, \mathbf{f}_\perp, t), \quad \text{(A6)}$$

where $\overline{L}_i(t)$ and L_i are related through the similarity transformation $\overline{L}_i(t) = e^{-\lambda L_b t} L_i e^{\lambda L_b t}$. Thus, the overbar denotes a quantity in an "interaction" representation, by analogy with the interaction picture of quantum-mechanics. The solution to Eq. (A6) may formally be written using a time-ordered exponential,

$$\overline{P}(\eta, \mathbf{f}_\perp, t) = \hat{T} \exp\left(L_a t + \int_0^t dt' \overline{L}_i(t')\right) P(\eta, \mathbf{f}_\perp, 0), \quad \text{(A7)}$$

with $\hat{T}$ being the time-ordering operator. Applying the projection operator $\mathcal{P}$ to both sides of Eq. (A5), and using Eq. (A4) gives

$$\sigma(\eta, t) = \rho_{eq}(\mathbf{f}_\perp)^{-1} \int d\mathbf{f}_\perp \overline{P}(\eta, \mathbf{f}_\perp, t), \quad \text{(A8)}$$

whereupon from Eq. (A7) and the initial condition Eq. (A1b),

$$\sigma(\eta, t) = \left\langle \hat{T} \exp\left(L_a t + \int_0^t dt' \overline{L}_i(t')\right) \right\rangle_f \sigma(\eta, 0), \quad \text{(A9)}$$

where for any operator $A(\mathbf{f}_\perp)$,

$$\langle A(\mathbf{f}_\perp)\rangle_f = \int d\mathbf{f}_\perp A(\mathbf{f}_\perp)\rho_{eq}(\mathbf{f}_\perp). \quad \text{(A10)}$$

Although we can now proceed directly with the cumulant expansion of the expectation appearing in Eq. (A9), evaluation of the cumulants is eased by a change of representation. Denote the eigenstates of L_b and its adjoint $L_b^\dagger$ as $|\phi_n\rangle$ and $\langle\psi_n|$, respectively. The $|\phi_n\rangle$ and $\langle\psi_n|$ form a biorthogonal set with $\langle\psi_m|\phi_n\rangle = \delta_{mn}$. The explicit

forms of the $n = 0$ eigenstates are $|\phi_0\rangle \to \rho_{\text{eq}}(\mathbf{f}_\perp)$ and $\langle\psi_0| \to 1$. This allows the expectation in Eq. (A9) to be written $\langle A\rangle_f = \langle\psi_0|A|\phi_0\rangle$. Further, putting $\hat{U} = e^{v^2/4D_v}$ consider the similarity transformation

$$L_B = \hat{U}L_b\hat{U}^{-1} = -B_\mu^\dagger B_\mu, \qquad \text{(A11)}$$

where

$$B_\mu = \frac{f_\mu}{2\sqrt{D_f}} + \sqrt{D_f}\frac{\partial}{\partial f_\mu}, \qquad \text{(A12a)}$$

$$B_\mu^\dagger = \frac{f_\mu}{2\sqrt{D_f}} - \sqrt{D_f}\frac{\partial}{\partial f_\mu}. \qquad \text{(A12b)}$$

For planar channeling, the indices μ in Eqs. (A11) and (A12) can be ignored. For axial channeling, μ can take two possible values and the convention of summation over repeated Greek indices is used. L_B is self-adjoint, $L_B^\dagger = L_B$, and its eigenstates are the harmonic oscillator states $|n\rangle = \hat{U}|\phi_n\rangle$ and $\langle n| = \langle\psi_n|\hat{U}^{-1}$, with $\langle m|n\rangle = \delta_{mn}$. B_μ and $B_\mu^\dagger$ have the properties,

$$[B_\mu, B_\nu^\dagger] = \delta_{\mu\nu}, \qquad \text{(A13a)}$$

$$B_\mu^\dagger|n\rangle = \sqrt{n+1}|n+1\rangle, \qquad \text{(A13b)}$$

$$B_\mu|n\rangle = \sqrt{n}|n-1\rangle, \quad B_\mu|0\rangle = 0, \qquad \text{(A13c)}$$

and are the creation and annihilation operators for the harmonic oscillator. For axial channeling the $|n\rangle$ are two-dimensional harmonic oscillator states, and we require a further index μ on the n (which we have suppressed).

Thus, introducing factors of unity, $1 = \hat{U}^{-1}\hat{U}$, inside the expectation, Eq. (A9) may be written equivalently as

$$\sigma(\eta,t) = \langle 0|\hat{T}\exp\left(L_at + \int_0^t dt'\overline{L}_I(t')\right)|0\rangle\sigma(\eta,0),$$
$$\text{(A14)}$$

where

$$\begin{aligned}\overline{L}_I(t) &= \hat{U}e^{-\lambda L_b t}L_i e^{\lambda L_b t}\hat{U}^{-1} \\ &= -\alpha_\mu\left(\overline{B}_\mu(t) + \overline{B}_\mu^\dagger(t)\right),\end{aligned} \qquad \text{(A15)}$$

and $\alpha_\mu = \sqrt{D_f}\partial/\partial p_\mu$. The explicit time dependence of the operators $\overline{B}_\mu(t) = e^{-\lambda L_b t}B_\mu e^{\lambda L_b t}$ and $\overline{B}_\mu^\dagger(t) = e^{-\lambda L_b t}B_\mu^\dagger e^{\lambda L_b t}$ is found most simply from the corresponding "Heisenberg" equations with initial conditions $\overline{B}_\mu(0) = B_\mu$ and $\overline{B}_\mu^\dagger(0) = B_\mu^\dagger$, giving

$$\overline{L}_I(t) = -\alpha_\mu(B_\mu e^{-\lambda t} + B_\mu^\dagger e^{\lambda t}). \qquad \text{(A16)}$$

With this change of representation, Eq. (A14) may be rewritten in terms of a cumulant expansion,

$$\sigma(\eta,t) = \hat{T}\exp\left(\sum_{n=1}^\infty \int_0^t dt_1 F_n(t_1)\right)\sigma(\eta,0), \qquad \text{(A17a)}$$

where

$$\begin{aligned}F_n(t_1) &= \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \cdots \int_0^{t_{n-1}} dt_n \\ &\quad \times \langle\phi(t_1)\phi(t_2)\cdots\phi(t_n)\rangle_c,\end{aligned} \qquad \text{(A17b)}$$

$$\phi(t) = L_a + \overline{L}_I(t), \qquad \text{(A17c)}$$

and the subscript c in Eq. (A17b) denotes a cumulant average (often called "ordered" cumulants to distinguish them from other types of cumulants). Such expansions in the context of stochastic processes have been studied in detail by Kubo,[52] van Kampen,[53] and Fox.[54] A general prescription for expressing these cumulants in terms of ordinary moments has been given by van Kampen.[53] Thus, for example, the first three cumulants are

$$\langle\phi(t_1)\rangle_c = \langle 1\rangle, \qquad \text{(A18a)}$$

$$\langle\phi(t_1)\phi(t_2)\rangle_c = \langle 12\rangle - \langle 1\rangle\langle 2\rangle, \qquad \text{(A18b)}$$

$$\begin{aligned}\langle\phi(t_1)\phi(t_2)\phi(t_3)\rangle_c &= \langle 123\rangle - \langle 12\rangle\langle 3\rangle - \langle 13\rangle\langle 2\rangle \\ &\quad - \langle 1\rangle\langle 23\rangle + \langle 1\rangle\langle 2\rangle\langle 3\rangle + \langle 1\rangle\langle 3\rangle\langle 2\rangle,\end{aligned}$$
$$\text{(A18c)}$$

where $\langle 12\rangle = \langle 0|\phi(t_1)\phi(t_2)|0\rangle$, etc. For cummutating operators, the above reduce to the rules given in Eq. (4.10).

Utilizing the above rules, the first four terms of the expansion in Eq. (A17) are

$$F_1(t) = L_a, \qquad \text{(A19a)}$$

$$F_2(t) = \lambda^{-1}\alpha_\mu\alpha_\mu(1 - e^{-\lambda t}), \qquad \text{(A19b)}$$

$$F_3(t) = \lambda^{-2}\alpha_\mu[L_a, \alpha_\mu]\left[1 - (1 + \lambda t)e^{-\lambda t}\right], \qquad \text{(A19c)}$$

$$F_4(t) = \lambda^{-3}\alpha_\mu[L_a, [L_a, \alpha_\mu]]\left[1 - (1 + \lambda t + \tfrac{1}{2}\lambda^2 t^2)e^{-\lambda t}\right].$$
$$\text{(A19d)}$$

Here we continue using the convention of summation over repeated Greek indices. We do not study the properties of the sum of terms $F_n(t)$, but we expect the series to be asymptotic in large λ due to a cluster property of the cumulants.[54] In this large λ limit, and retaining only the first two terms, the time derivative of Eq. (A17) gives the desired equation,

$$\frac{\partial}{\partial t}\sigma(\eta,t) = \left(L_a + \lambda^{-1}\alpha_\mu\alpha_\mu\right)\sigma(\eta,t). \qquad \text{(A20)}$$

This equation is valid over time scales much longer than λ^{-1}.

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