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Classical Wigner theory of gas surface scattering

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The scattering of atoms from surfaces is studied within the classical Wigner formalism. A new analytical expression is derived for the angular distribution and its surface temperature dependence. The expression is valid in the limit of weak coupling between the vertical motion with respect to the surface and the horizontal motion of the atom along the periodic surface. The surface temperature dependence is obtained in the limit of weak coupling between the horizontal atomic motion and the surface phonons. The resulting expression, which takes into account the surface corrugation, leads to an almost symmetric double peaked angular distribution, with peaks at the rainbow angles. The analytic expression agrees with model numerical computations. It provides a good qualitative description for the experimentally measured angular distribution of Ne and Ar scattered from a Cu surface. © 2008 American Institute of Physics. [DOI: [10.1063/1.2954020](https://doi.org/10.1063/1.2954020)]

I. INTRODUCTION

The scattering of atoms by metal surfaces has been employed as a probe of surface properties such as diffusion, vibrational relaxation, and structure.¹ Rare gas atoms such as He, Ne, and Ar typically do not penetrate into the bulk and so the information obtained from the scattering process relates mainly to the surface layer. Since these atoms are relatively inert, they act as good probes for a large variety of atoms and molecules adsorbed on the surface. Experimental studies of the scattering of He, Ne, and Ar from copper vicinal surfaces have been reported in a series of papers.^{2–6} Further studies of Ne and Ar atoms scattered from a variety of surfaces have been reported in Refs. 7–13.

The initial theoretical studies of helium atom scattering by metal surfaces predated the experiments.¹⁴ Due to the light mass of the He atom, it necessitates a quantum mechanical treatment. A general coupled channel method of calculating diffraction intensities for a given geometry was developed by Wolken.¹⁵ More approximate methods have been used to invert measured angular distributions into force fields describing the interaction of the elastically¹⁶ and inelastically¹⁷ scattered atoms from copper vicinal surfaces.

Miller and co-workers,^{18–21} Doll,²² and Guantes *et al.*²³ utilized semiclassical *S*-matrix theory to study He atom scattering from surfaces. In contrast to a purely classical dynamics approach, the semiclassical theory could account for the important features of the diffraction pattern. Two recent reviews of classical, semiclassical, and quantum methods may be found in Refs. 24 and 25. For noble gas atom scattering the corrugated Morse potential (CMP) proposed by Armand and Manson⁴ has been found²⁶ to be in satisfactory agreement with experimental results. Further work by Gorse *et al.*⁵

has led to the introduction of a modified corrugated Morse potential (MCMP) for the low corrugated (110) and (113) faces.

Due to their heavier masses, the scattering of Ne and especially Ar is readily described in terms of classical models. Brako²⁷ developed a classical model in which the vertical motion (*z* direction) is coupled linearly to the phonon bath of the surface. He derived an analytical expression for the angular distribution, as well as for the energy and momentum transfer. However, he did not consider the effect of corrugation on the scattering. As a result he also ignored the coupling between the phonon bath and the horizontal motion of the atom along the surface. The classical rainbow effect which is caused by surface corrugation was then studied by Horn *et al.*²⁸ The effect of surface corrugation was further studied through the introduction of the so called “washboard model” of Tully.²⁹ However, as in the paper of Horn *et al.*²⁸ the classical collision dynamics was simplified by assuming an impulsive collision model. A further refinement of the washboard model with applications to the scattering of Ne and Ar atoms may be found in Ref. 30. Most recently, Manson and co-workers^{31,32} have employed the theory of Brako²⁷ to analyze the experimental results for the scattering of Ar atoms from Ru(0001). Detailed molecular dynamics investigation of Ar atom scattering on Ni(001) may be found in Ref. 33 and Ar on Pt(111) in Ref. 34.

The purpose of the present study is to derive and apply a classical theory for the angular distribution that takes into account surface corrugation and surface temperature and does not use an impulsive approximation for the dynamics. The formalism we will use is known as the classical Wigner approximation,^{35–38} in which the initial conditions of the phonon bath and the incident wavepacket are treated quantum mechanically while the ensuing dynamics is obtained from the classical equations of motion. The same approach has been called “mixed quantum classical theory”^{39,40} or the

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“linearization approximation”⁴¹ since it is a linearized limit of a semiclassical initial value representation treatment of the same problem.⁴²

The central result of this paper is that the angular distribution is given by the expression

$$P(\theta) = \frac{1}{\pi} \int_{-\infty}^{\infty} dZ \sqrt{\frac{1}{2\pi\sigma^2}} \times \exp\left(-\frac{Z^2}{2\sigma^2}\right) \frac{H(K^2 - (\theta + \theta_{i0} + \Delta\theta_1 + Z)^2)}{\sqrt{K^2 - (\theta + \theta_{i0} + \Delta\theta + Z)^2}}, \quad (1.1)$$

where $H(x)$ is the Heaviside function and θ_{i0} is the incident scattering angle. The distribution is determined by three parameters—a shift parameter $\Delta\theta$, the rainbow angle K (relative to the specular scattering angle $-\theta_{i0}$), and the thermal width as given in terms of the variance σ^2 that (in the high temperature classical limit) is linearly proportional to the surface temperature. This expression is valid provided that the following conditions hold: (a) the incident wavepacket is localized in momentum space, (b) the scattering is classical in nature, (c) the coupling between the vertical and the horizontal motion of the scattered particle is weak, and (d) the frictional force exerted on the particle by the surface is weak.

In Sec. II we provide the framework and the working model underlying the theory. We then provide in Sec. III the expression for the angular distribution first in the absence and then in the presence of interaction with the phonon bath. The detailed derivations are given in Appendixes A thru C. The analytic theory is then applied in Sec. IV to a model in which the potential in the vertical direction is taken to be the Morse potential. The results are compared with numerically exact simulations. We find excellent agreement, further justifying the analytic theory. We then apply the theory to the scattering of Ne and Ar from a Cu surface. Good qualitative agreement is found for both systems.

We end with a discussion of further extensions of the theory, as well as the possibility of solving the same problem but within a semiclassical initial value representation treatment of the scattering dynamics. Such a treatment would also be valid for the scattering of He where due to the small mass, quantum interference effects cannot be ignored.

II. THEORETICAL FRAMEWORK

A. The thermal angular distribution

We consider the scattering of a particle with mass M from a surface. In the vertical z direction the particle interacts with the surface via a potential $V(z)$, which would typically have the form of a Morse potential. The potential along the x direction is periodic. For simplicity we ignore the y coordinate. When the particle is sufficiently distant from the surface, it is a free particle. When it comes closer to the surface, there is an interaction between the vertical and horizontal motion. We also assume that the interaction with the phonon bath comes from coupling between the horizontal

motion and the phonons of the surface; the vertical motion is not directly coupled to the bath. We then use the following model Hamiltonian:

$$H = \frac{p_x^2 + p_z^2}{2M} + V(z) + V_l f(z) \cos\left(\frac{2\pi x}{l}\right) + \frac{1}{2} \sum_{j=1}^N \left[p_j^2 + \omega_j^2 \left(x_j - \frac{c_j \sqrt{M}}{\omega_j^2} x g(z) \right)^2 \right], \quad (2.1)$$

where l is the periodic lattice length and $p_j, x_j, j=1, \dots, N$ are the mass weighted momentum and position operators of the bath degrees of freedom, which are linearly coupled to the system motion. The function $f(z)$ gives the coupling due to the corrugation and $g(z)$ couples the harmonic bath to the horizontal motion when the particle is close to the surface.

The bath Hamiltonian (in mass weighted coordinates and momenta) is defined to be

$$H_B = \frac{1}{2} \sum_{j=1}^N (p_j^2 + \omega_j^2 x_j^2). \quad (2.2)$$

Initially, the atom is sufficiently distant from the surface so that it does not interact with it. It is then described by a Gaussian wave function $|\psi\rangle$. The exact quantum mechanical expression for the angular distribution is written in terms of a correlation function with factorized initial conditions as

$$P(\theta) = \lim_{t \rightarrow \infty} P(\theta, t) = \lim_{t \rightarrow \infty} \text{Tr} \left(\frac{e^{-\beta H_B}}{Z_B} |\psi\rangle \langle \psi| \hat{K}^\dagger(t) \delta(\theta - \hat{\theta}) \hat{K}(t) \right). \quad (2.3)$$

Here, $\hat{K}(t)$ is the exact quantum mechanical propagator ($\hat{K}(t) = \exp(-i\hat{H}t/\hbar)$), $\delta(x)$ is the Dirac “delta” function, and the angular operator is defined as the angle with respect to the normal to the surface,

$$\hat{\theta} = \tan^{-1} \left(\frac{\hat{p}_x}{\hat{p}_z} \right). \quad (2.4)$$

B. The classical Wigner approximation

The Wigner representation of the thermal harmonic bath is

$$\rho_{B,W}(\mathbf{p}, \mathbf{x}) \equiv \left\langle \frac{e^{-\beta H_B}}{Z_B} \right\rangle_W = \prod_{j=1}^N \left(\nu_j / (\pi \hbar) \exp \left(-\frac{\nu_j}{\hbar \omega_j} (p_j^2 + \omega_j^2 x_j^2) \right) \right), \quad (2.5)$$

with

$$\nu_j \equiv \tanh \left(\frac{\hbar \beta \omega_j}{2} \right), \quad (2.6)$$

and the distribution is normalized.

The initial wave function is chosen to have the Gaussian form

$$\langle x, z | \psi \rangle = \left(\frac{\gamma}{\pi} \right)^{1/2} \exp \left(-\frac{\gamma}{2} [(x - x_0)^2 + (z - z_0)^2] \right) + \frac{i}{\hbar} p_{x_0} (x - x_0) + \frac{i}{\hbar} p_{z_0} (z - z_0). \quad (2.7)$$

The momenta p_{x_0} and p_{z_0} define the incident scattering angle

$$\theta_{i0} = \tan^{-1} \left(\frac{p_{x_0}}{p_{z_0}} \right) \quad (2.8)$$

and the radial initial momentum

$$p_0^2 = p_{x_0}^2 + p_{z_0}^2. \quad (2.9)$$

The Wigner representation of $|\psi\rangle\langle\psi|$ is

$$\rho_S(p_x, p_z, x, z) = \left(\frac{1}{\pi\hbar} \right)^2 \exp \left(-\gamma [(x - x_0)^2 + (z - z_0)^2] - \frac{(p_x - p_{x_0})^2 + (p_z - p_{z_0})^2}{\hbar^2 \gamma} \right), \quad (2.10)$$

and in this representation the angular distribution then becomes

$$P(\theta) = \lim_{t \rightarrow \infty} \int_{-\infty}^{\infty} \prod_{j=1}^N \frac{dp_j dx_j}{2\pi\hbar} \int_{-\infty}^{\infty} dp_x dp_z dx dz \rho_{B,W}(\mathbf{p}, \mathbf{x}) \times \rho_S(p_x, p_z, x, z) \delta \left(\theta - \tan^{-1} \left(\frac{p_x(t)}{p_z(t)} \right) \right), \quad (2.11)$$

where the notation $p_z(t), p_x(t)$ stands for the time evolution. In the classical Wigner approximation which will be considered henceforth in this paper, this time evolution is given by the classical equations of motion.

C. The classical dynamics

Using the shorthand notation $V(x, z) = V(x) + V_1 f(z) \cos(2\pi x/l)$, defining a spectral density

$$J(\omega) = \frac{\pi}{2} \sum_{j=1}^N \frac{c_j^2}{\omega_j} \delta(\omega - \omega_j) \quad (2.12)$$

and an associated friction function

$$\eta(t) = \frac{2}{\pi} \int_0^\infty d\omega \frac{J(\omega)}{\omega} \cos(\omega t), \quad (2.13)$$

one finds that the equations of motion for the system variables are the generalized Langevin equations (GLEs)

$$M\ddot{x}_t + \frac{\partial V(x_t, z_t)}{\partial x_t} + M g(z_t) \int_{-t_0}^t dt' \eta(t-t') \left[\frac{d[x_t' g(z_t')]}{dt'} \right] = \sqrt{MF(t)} g(z_t), \quad (2.14)$$

$$M\ddot{z}_t + \frac{\partial V(x_t, z_t)}{\partial z_t} + M g'(z_t) x_t \int_{-t_0}^t dt' \eta(t-t') \times \left[\frac{d[x_t' g(z_t')]}{dt'} \right] = \sqrt{MF(t)} g'(z_t) x_t, \quad (2.15)$$

where the trajectory is initiated at the time $-t_0$. The initial wavepacket is chosen such that z_0 is sufficiently far away from the surface. For the chosen value of the width γ , we can safely assume that for all initial conditions the phonon bath is not coupled to the atomic motion, that is, $g(z)=0$. The Gaussian noise function is therefore dependent only on the initial conditions of the phonon bath,

$$F(t) = \sum_{j=1}^N c_j \left(x_j \cos[\omega_j(t+t_0)] + \frac{p_j}{\omega_j} \sin[\omega_j(t+t_0)] \right). \quad (2.16)$$

It has zero mean and its correlation function is proportional to the time dependent friction function.

III. CLASSICAL WIGNER THEORY FOR THE ANGULAR DISTRIBUTION

A. Scattering in the absence of dissipation

In this subsection we will present an analytical expression for the angular distribution in the absence of interaction with the bath and in the limit of weak corrugation—the potential parameter V_1 that couples the vertical to the horizontal motion will be considered to be small. The dynamics is solved to leading order in V_1 . In this way we obtain an expression for the momentum added (or subtracted) to the horizontal motion as a result of this coupling. Through energy conservation this also leads to a shift in the vertical momentum. We then use these results to derive an expression for the angular distribution, in the limit that the initial wavepacket is very narrow in the momentum space or, more specifically, in the limit that the width parameter in the incident wavepacket $\gamma \rightarrow 0$.

As shown in Appendix A, using a perturbation theory for the dynamics, in which the small parameter is the corrugation strength V_1 , which is assumed to be much smaller than the well depth V_0 , one finds that for a given trajectory, the shift in the scattering angle induced by the corrugation is given by the expression

$$\delta\theta_t = \frac{\delta p_x(x, p_x, p_z)}{p_z} \equiv K(p_z, p_x) \sin \left(\frac{2\pi}{l} \left[x + \frac{p_x t_0}{M} \right] \right). \quad (3.1)$$

As shown below, the rainbow angles are found to be $-\theta_i \pm K$, where $K(p_z, p_x)$ is given in terms of the potential parameters and the known unperturbed trajectory in the vertical direction, which depends on the initial momentum in the vertical direction,

$$K(p_z, p_x) = \frac{4\pi V_1}{p_z l} \int_0^\infty dt' \cos \left(\frac{2\pi p_x t'}{lM} \right) f[z(t')]. \quad (3.2)$$

To obtain the angular distribution we must integrate over the initial coordinates and momenta. The details of the integration are presented in Appendix B. The only additional

approximation introduced is that the width parameter γ of the initial wavepacket is assumed to be small as compared to the lattice length l , such that $\gamma l^2 \ll 1$. One then finds that

$$P(\theta) = \frac{1}{\pi} \frac{H(K^2 - (\theta + \theta_{i0})^2)}{\sqrt{K^2(p_0, \theta_{i0}) - (\theta + \theta_{i0})^2}}, \quad (3.3)$$

where $H(x)$ is the Heaviside function, $H(x) = 1$ if $x > 0$ and is 0 otherwise. The scattered angular distribution is a function that has maxima at the rainbow angles $\theta + \theta_{i0} = \pm K(p_0, \theta_{i0})$ and a minimum at the specular angle for which

$$P(-\theta_{i0}) = \frac{1}{\pi |K(p_0, \theta_{i0})|}. \quad (3.4)$$

Outside of the range $-|K| \leq \theta + \theta_{i0} \leq |K|$ the angular distribution vanishes.

B. Scattering with weak dissipation

Typically, the friction exerted on the incident atom will have a phonon component that can be characterized with two parameters—a friction strength parameter (η) and a cutoff frequency ω_c that reflects the Debye frequency of the crystal. The spectral density would be thus typically Ohmic with an exponential cutoff frequency. This gives rise to memory in the time dependent friction. However, one may expect that the memory effect on the dynamics will be negligible since the cutoff frequency is typically much larger than the atom surface frequencies. We therefore simplify the consideration of the dynamics and specify it for purely Ohmic friction such that

$$\eta(t) = 2\eta\delta(t). \quad (3.5)$$

As in the dissipationless case, we proceed in Appendix C to use perturbation theory to derive an expression for the change in final scattering angle for a trajectory with specified initial conditions for the system variables and a given realization of the noise. In addition to considering the corrugation parameter V_1 as small relative to V_0 , here we also assume that the friction is weak, allowing for a first order perturbation theory in these parameters.

The straightforward application of perturbation theory then leads to the following result for the final momentum in the horizontal (x) direction:

$$\Delta p_x = p_z K(p_x, p_z) \sin\left(\frac{2\pi}{l} \left[x + \frac{p_x}{M} t_0\right]\right) + \Delta p_{x,1} + \Delta p_{x,2}. \quad (3.6)$$

Here, the rainbow angle function K is defined as in the dissipationless case [see Eq. (3.2)]. In addition one finds a friction induced change in the momentum

$$\Delta p_{x,1} = -\eta p_x \int_0^\infty dt g^2(z_t) \quad (3.7)$$

and a noise induced change

$$\Delta p_{x,2} = 2\sqrt{M} \sum_{j=1}^N c_j h_j \left(x_j \cos(\omega_j t_0) + \frac{p_{j1}}{\omega_j} \sin(\omega_j t_0) \right), \quad (3.8)$$

where h_j is the cosine Fourier transform of the vertical motion dependent coupling function

$$h_j \equiv \int_0^\infty dt g(z_t) \cos(\omega_j t). \quad (3.9)$$

As in the dissipationless case, the shift in the vertical momentum is determined by the energy balance. However, here one has to take also into account the energy gained by the bath during the collision,

$$\Delta E_B = \langle \Delta E_B \rangle + \delta E_B. \quad (3.10)$$

The energy loss to the bath has two parts—the average energy loss to the bath $\langle \Delta E_B \rangle$ and the fluctuational energy loss δE_B . Explicit expressions for the energy losses in terms of the system and bath dynamics are given in Appendix C. The resulting change in the vertical momentum after the scattering event is then found to be

$$\Delta p_z = \frac{p_x}{p_z} \Delta p_x + \frac{M \Delta E_B}{p_z}. \quad (3.11)$$

Expanding the final scattering angle to first order with respect to the changes in the horizontal and vertical momenta, one finds that the change in the scattering angle is

$$\delta \theta_i \equiv K(p_x, p_z) \sin\left(\frac{2\pi}{l} \left[x + \frac{p_x}{M} t_0\right]\right) + \Delta \theta_1 + \Delta \theta_2, \quad (3.12)$$

where the angular shift is

$$\Delta \theta_1 = \frac{\Delta p_{x,1}}{p_z} \cos^2 \theta_i \quad (3.13)$$

and the angular fluctuation is

$$\Delta \theta_2 = \frac{\Delta p_{x,2}}{p_z} \cos^2 \theta_i. \quad (3.14)$$

To obtain the angular distribution it remains to integrate over the system phase space variables and to average over the bath. The detailed derivation is provided in Appendix C; here we provide only the essential results. One finds that the angular distribution has a form that is similar to the angular distribution in the absence of dissipation, except that now it is broadened by a Gaussian distribution that reflects the interaction of the particle with the phonon bath of the surface. The expression for the angular distribution is

$$P(\theta) = \frac{1}{\pi} \int_{-\infty}^{\infty} dZ \frac{H(K^2 - (\theta + \theta_{i0} + \Delta \theta_1 + Z)^2)}{\sqrt{K^2(p_0, \theta_{i0}) - (\theta + \theta_{i0} + \Delta \theta_1 + Z)^2}} \times \sqrt{\frac{1}{2\pi\sigma^2}} \exp\left(-\frac{Z^2}{2\sigma^2}\right), \quad (3.15)$$

where the temperature and friction dependent variance of the distribution is expressed in the continuum limit as

TABLE I. Parameters for helium scattering.

	θ_{i0} (deg)	E_0 (meV)	Mass (a.u.)		
Beam parameters	-32.5	63.0	7296.12		
Cu(110) potential	V_0 (meV)	V_1/V_0	α ($\text{\AA}^{-1}$)	1 ($\text{\AA}$)	
Surface parameters	6.35	0.03, 0.0885	1.05	3.6	
Bath parameters	ω_c (a.u.)	η (a.u.)	ζ ($\text{\AA}$)		
	0.001 061 17	0.0002 ω_c	2.645		
Wavepacket parameters	γ (a.u.)	x_0 (a.u.)	z_0 (a.u.)	p_{x0} (a.u.)	p_{z0} (a.u.)
	0.020 933 9	-25.4828	40.0	3.1249	-4.9051

$$\sigma^2 = \frac{4\hbar M}{\pi p_0^2} \int_0^\infty d\omega J(\omega) \frac{\left(\int_0^\infty dt g(z_t) \cos(\omega t) \right)^2}{\tanh \frac{\hbar \beta \omega}{2}}. \quad (3.16)$$

Equations (3.15) and (3.16) are the central results of this paper. The coupling to the thermal bath induces a shift in the peaks through the term $\Delta\theta_1$ [cf. Eq. (3.13)] and a broadening from the Gaussian averaging. The variance of the distribution is readily obtained in the continuum limit through the spectral density. Note that the variance is quantum mechanical; it does not vanish in the limit of zero temperature but includes the zero point fluctuations of the bath. This reflects the classical Wigner approximation, in which the initial thermal conditions are treated quantum mechanically and it is only the dynamics that is treated classically.

IV. APPLICATIONS

A. Analytic results for a Morse oscillator potential in the absence of dissipation

In the following we will use the known analytic dynamics of the Morse oscillator to obtain an explicit expression for the rainbow angle function $K(p_0, \theta_{i0})$. The model potential we use is

$$V(x, z) = V_0 [1 - \exp(-\alpha z)]^2 - V_0 + V_1 \times \exp(-2\alpha z) \cos\left(\frac{2\pi x}{l}\right), \quad (4.1)$$

where the function coupling the vertical motion to the horizontal motion has been specified to be $f(z) = \exp(-2\alpha z)$. The initial vertical energy of the particle when it is far from the surface is denoted as E_z and the initial vertical momentum is p_z . As already noted above, the origin of time was chosen as the point at which the trajectory reaches the turning point (z_{tp}) so that at time $t=0$ the vertical velocity vanishes. Therefore

$$E_z = V(z_{tp}) = \frac{p_z^2}{2M}. \quad (4.2)$$

In the absence of coupling, the equation of motion for the z coordinate is

$$M\ddot{z} = -2\alpha V_0 (1 - \exp(-\alpha z)) \exp(-\alpha z). \quad (4.3)$$

One can then find by inspection that the time dependence of the classical trajectory is given by the relation

$$\exp(\alpha z_t) = \frac{V_0}{E_z} \left(\left(\sqrt{1 + \frac{E_z}{V_0}} \right) \cosh(\Omega t) - 1 \right), \quad (4.4)$$

where the energy dependent frequency is given in terms of the energy and potential parameters as

$$\Omega^2 = \frac{2\alpha^2 E_z}{M}. \quad (4.5)$$

The resulting expression for the rainbow angle function is

$$K(p_0, \theta_{i0}) = \frac{-4\pi V_1 E_z^2}{p_0 l \cos \theta_{i0} (E_z + V_0) V_0} \int_0^\infty dt' \cos\left(\frac{2\pi p_x t'}{lM}\right) \times \frac{1}{\left(\cosh(\Omega t') - \sqrt{\frac{V_0}{E_z + V_0}} \right)^2}. \quad (4.6)$$

In the limit of high incident energy as compared to the well depth of the Morse potential ($E_z \gg V_0$), one obtains the result,

$$K(p_x, p_z) \simeq -\frac{2\pi V_1}{\alpha l V_0} \int_0^\infty dt' \frac{\cos\left(\frac{2\pi \tan(\theta_{i0})}{l\alpha} t'\right)}{\cosh^2(t')} = \frac{2\pi V_1}{\alpha l V_0} \frac{\pi^2 (\tan \theta_{i0})}{l\alpha \sinh\left(\frac{\pi^2 |\tan(\theta_{i0})|}{l\alpha}\right)}. \quad (4.7)$$

B. Analytic results for a Morse oscillator potential in the presence of dissipation

The horizontal motion will typically be coupled to the surface phonon bath when the approaching atom is close to the surface. For the Morse potential model used above, it is thus reasonable to choose the coupling function as a Gaussian centered at the minimum of the Morse potential

$$g(z) = \exp(-z^2/\zeta^2). \quad (4.8)$$

As can be inferred from Eq. (4.4) the time t^* at which the atom goes through the minimum ($z=0$) is

$$\cosh(\Omega t^*) = \sqrt{1 + \frac{E_z}{V_0}}. \quad (4.9)$$

At this time,

$$\left(\frac{dz}{dt}\right)^2 \Big|_{t=t^*} = \frac{\Omega^2}{\alpha^2} \left(1 + \frac{V_0}{E_z}\right), \quad (4.10)$$

so that we may approximate

$$\exp(-z^2/\xi^2) \approx \exp\left(-\frac{\Omega^2}{\xi^2 \alpha^2} \left(1 + \frac{V_0}{E_z}\right) (t - t^*)^2\right). \quad (4.11)$$

It then follows that

$$\begin{aligned} h_j &= \int_0^\infty dt g(z_t) \cos(\omega_j t) \\ &\approx \int_{-\infty}^\infty dt \cos(\omega_j [t + t^*]) \exp\left(-\frac{\Omega^2}{\xi^2 \alpha^2} \left(1 + \frac{V_0}{E_z}\right) t^2\right) \\ &= \frac{\xi \alpha \sqrt{\pi} \cos(\omega_j t^*)}{\Omega \sqrt{1 + \frac{V_0}{E_z}}} \exp\left(-\frac{\xi^2 \alpha^2 \omega_j^2}{4 \Omega^2 \left(1 + \frac{V_0}{E_z}\right)}\right). \end{aligned} \quad (4.12)$$

and

$$\Delta p_{x,1} = -\eta p_x \int_0^\infty dt g^2(z_t) \approx -\frac{\eta p_x \xi \alpha \sqrt{\pi}}{\Omega \sqrt{2 \left(1 + \frac{V_0}{E_z}\right)}}. \quad (4.13)$$

We thus have that

$$\Delta \theta_1 = \frac{\Delta p_{x,1}}{p_z} \cos^2(\theta_{i0}) = -\frac{\eta \xi \alpha \sqrt{\pi} \sin(\theta_{i0}) \cos(\theta_{i0})}{\Omega \sqrt{2 \left(1 + \frac{V_0}{E_z}\right)}}. \quad (4.14)$$

Using the definition of the spectral density, specifying to Ohmic friction with an exponential cutoff

$$(J(\omega) = \eta \omega \exp(-\omega/\omega_c))$$

and taking the classical limit for the thermal bath distribution ($\hbar \rightarrow 0$), we then have

$$\begin{aligned} \sigma^2 &= \frac{4M \cos^4(\theta_{i0})}{\beta p_z^2} \sum_j \frac{c_j^2 h_j^2}{\omega_j^2} \\ &= \frac{2M \eta \xi^2 \cos^4(\theta_{i0})}{\beta E_z (E_z + V_0)} \int_0^\infty d\omega \cos^2(\omega t^*) \exp\left(-\frac{\xi^2 \alpha^2 \omega^2}{2 \Omega^2 \left(1 + \frac{V_0}{E_z}\right)} - \frac{\omega}{\omega_c}\right) \\ &= \frac{1}{\beta E_z} \frac{\sqrt{\pi M} \eta \xi \cos^4(\theta_{i0})}{\sqrt{E_z + V_0}} \exp\left(\frac{E_z + V_0}{M \omega_c^2 \xi^2}\right) \left(1 - \operatorname{erf}\left(\sqrt{\frac{E_z + V_0}{M \omega_c^2 \xi^2}}\right)\right) + \frac{\sqrt{\pi M} \eta \xi \cos^4(\theta_{i0})}{\beta E_z \sqrt{E_z + V_0}} \\ &\quad \times \operatorname{Re} \left[\exp\left(\frac{E_z + V_0}{M \omega_c^2 \xi^2} (1 - 2i\omega_c t^*)^2\right) \left(1 - \operatorname{erf}\left((1 - 2i\omega_c t^*) \sqrt{\frac{E_z + V_0}{M \omega_c^2 \xi^2}}\right)\right) \right]. \end{aligned} \quad (4.15)$$

For Ohmic friction the result is simpler,

$$\begin{aligned} \sigma^2 &= \frac{1}{\beta E_z} \frac{\sqrt{\pi M} \eta \xi \cos^4(\theta_{i0})}{\sqrt{E_z + V_0}} \\ &\quad \times \left[1 + \exp\left(-\frac{2t^{*2} \Omega^2 \left(1 + \frac{V_0}{E_z}\right)}{\xi^2 \alpha^2}\right) \right]. \end{aligned} \quad (4.16)$$

C. Numerical verification

In this subsection we will compare the analytic expression for the angular distribution with numerical simulations, in which the dissipative bath is represented in terms of a finite number of oscillators, which closely mimic the continuum limit. The resulting friction kernel in the simulation is thus not strictly Ohmic; however, as already noted, the

memory time is sufficiently short, so it does not really make a big difference. The parameters used for the comparison with simulation roughly describe the scattering of He from the Cu(110) surface. The relevant scattering, potential, initial wavepacket, and frictional parameters are provided in Table I.

We first present in Fig. 1 results obtained for scattering without dissipation and two values of the corrugation strength parameter $V_1 = 0.03V_0, 0.0885V_0$. The initial distance of the wave packet center from the surface is sufficiently large to ensure that the wavepacket is in the asymptotic region where the interaction with the surface is negligible while the impact parameter is taken to be zero. The analytic results are obtained by averaging the expression for the angular distribution given in Eq. (B5) over the Gaussian distribution of the initial wavepacket. As can be seen from the

FIG. 1. Comparison between the angular distributions for classical helium atom scattering obtained numerically (solid line) and using the analytical expression (dashed line) in the absence of dissipation for two different corrugation amplitudes. The inset gives a schematic profile of the scattering.

figure, for the weaker corrugation, the agreement is nearly perfect; it worsens slightly as the corrugation strength increases. We note especially that for the stronger corrugation, the angular distribution is not perfectly symmetric about the specular angle. This asymmetry, already noted by Tully,²⁹ which appears both in the simulation and in the analytic theory is a result of the initial Gaussian distribution, which is chosen to be Gaussian in the Cartesian momenta and so is not symmetrically distributed in the angular space. The asymmetry grows as the corrugation strength increases, increasing the magnitude of the difference between the rainbow angle and the specular angle.

The numerical results with dissipation were obtained by assuming an Ohmic spectral density with a cutoff frequency. For the parameters used, it was sufficient to use only 20 bath modes. The discretization was carried out as in Ref. 44 by assuming that there are N_b+1 bath oscillators and that the maximal bath frequency is infinity for the N_b+1 th mode. The N_b bath oscillator frequencies and coupling coefficients are then determined from the relations

$$\omega_j = -\omega_c \ln\left(1 - \frac{j}{N_b+1}\right), \quad (4.17)$$

FIG. 3. Comparison between the angular distributions for classical He atom scattering obtained numerically (solid lines) and using the analytical expression (dashed lines) at different temperatures for very weak corrugation $V_1/V_0=0.03$.

FIG. 2. Temperature dependence of the variance of the thermal bath (σ^2) for the parameters of He atom scattering as given in Table I in units of deg^2 .

$$c_j = \sqrt{\frac{2\omega_j^2 \eta \omega_c}{\pi(N_b+1)}}. \quad (4.18)$$

We first plot in Fig. 2 the temperature dependence of the variance [cf. Eqs. (C26), (C27), and (4.15)] for $V_1/V_0=0.03$ at three different levels of treatment. The dashed line denotes the values obtained for Ohmic friction with the exponential cutoff (the friction strength and cutoff frequency are as given in Table I). The solid line represents the values from the classical limit ($\hbar \rightarrow 0$) approximation [cf. Eq. (4.15)]. The points denote the values evaluated by using the simpler expression [Eq. (4.16)] for purely Ohmic friction. The quantum variance has a finite nonzero value at zero temperature while the classical approximation vanishes at $T=0$ K. However, this difference is noticeable only at very low temperature. It is also evident that the differences between the Ohmic and cutoff spectral densities are not very large.

In Figs. 3 and 4 we present results for the angular distribution as a function of temperature. Figure 3 shows the results for the weak corrugation case. The analytic results here include averaging over the initial wavepacket. The vari-

FIG. 4. Comparison between the angular distributions for classical He atom scattering obtained numerically (solid lines) and using the analytical expression (dashed lines) at different temperatures for a stronger corrugation $V_1/V_0=0.0885$.

ance has been computed for the spectral density of the discretized harmonic bath of 20 oscillators, rather than the continuum limit so as to make the comparison with the numerical simulations as close as possible. The simulated results were obtained by Monte Carlo averaging over initial conditions; the sample size used was 2.5×10^8 points. As is evident from the figure, the analytical expression is indeed a very good description for the classical angular distribution. Increasing the temperature causes a smearing out of the rainbow peaks, ultimately leading to a unimodal function.

The same results are shown also in Fig. 4, but this time for the larger value of the corrugation parameter. Again, the analytic expression provides a good representation of the true distribution; however the differences are more noticeable than for the weaker corrugation shown in Fig. 3. The larger corrugation causes the distance between the rainbow peaks to grow. As a result, even at high temperature one remains with a bimodal distribution.

D. Experimental application

The He atom is very light. Its angular distribution is distinctly nonclassical; even at low temperatures, the maxi-

mum of the distribution is found at the specular angle.³ However the angular distributions for both Ne and Ar scattered from the Cu surface and observed in the scattering plane are much more classical in nature. Already for Ne, one observes two peaks located at the rainbow angles, although these are modulated by nonclassical oscillations due to quantum diffraction. For Ar, there are no oscillations; one distinctly observes the double peaked structure as derived from the present classical theory.

In Fig. 5 we show fits of the analytic expression [Eq. (1.1)] to the experimental results for both Ne and Ar. The parameters used for the analytical fit are given in Table II. As is evident from the figure, the functional form provides a reasonable fit to the experimental results. The fact that the fits are not perfect reflects a number of approximations used in our derivation. Especially for Ne scattering, the classical dynamics cannot capture the diffraction oscillations, but only the overall envelope of the angular distribution. The low amplitude of the experimental high angle shoulder in the case of Ar scattering cannot be explained at the present level of the theory; we do note that we have ignored the third degree of freedom (the motion along the y direction) and this might have some influence on the resulting distribution, especially when the scattering angle is large.

FIG. 5. Analytical curve (dashed line) fitted to the experimentally obtained scattering distribution (solid line) of (a) Ar and (b) neon atoms. The experimental data are adapted from Ref. 3.

TABLE II. Parameters for experimental fits.

Parameters		Argon	Neon
Expt. Beam	E (meV)	63.75	63.38
	θ_{i0} (deg)	-28.5	-32.5
	M (a.u.)	72 820.41	36 785.7
Surface characteristics ^a	V_0 (meV)	50.8	12.70
	V_1/V_0	0.03	0.03
	T (K)	105	105
	$\Delta\theta$ (deg)	-0.5	-0.1
Fit parameters for analytic curve	σ^2 (deg ²)	11.3	7.0
	K (deg)	15.3	18.0

^aVide Ref. 3.

V. DISCUSSION

We have presented an analytical treatment of the classical Wigner theory of scattering from a corrugated surface. The derived expression is valid in the limit of weak coupling between the vertical and horizontal motions of the scattered particle as well as weak coupling with the surface phonon bath. Although we did not include any coupling between the vertical motion and the phonon bath, this is not essential. Such a coupling would only serve to increase the energy loss to the bath, leading to a further broadening of the distribution due to the thermal motion. The theory presented has been derived for Ohmic friction; however extension to a more realistic form of the friction which would include a cutoff frequency determined by the Debye frequency of the surface is straightforward and would not cause a significant change in the results.

The resulting angular distribution has two peaks, reflecting the rainbow scattering angles. The distribution is broadened and smeared out by interaction with the surface phonons. Increasing the temperature tends to turn the distribution into a unimodal one. Direct comparison of the theory with numerical simulations shows excellent agreement provided that the parameters are indeed in the weak interaction limit specified by the theory. As the coupling is increased, one finds that the qualitative form of the distribution is still well described by the analytic expression.

As already mentioned in Sec. I, there have been previous attempts at deriving a theory for the classical scattering. The major improvements of our theory are that (a) we include the surface corrugation; (b) coupling to the phonons is included via the horizontal motion; (c) we do not invoke impulsive collisions, but expressly use the potential of interaction and classical mechanics to derive the rainbow angles, energy loss, etc.; (d) when fitting to experimental results, the parameters may be inverted to provide an estimate of the frictional force felt by the scattered atom.

The analytic theory has been derived for in plane scattering. One may extend it also to include the coupling of the vertical motion to the phonon bath as well as inclusion of the second dimension of the surface, that is, to provide a three dimensional theory. These are topics for ongoing and future work. The present theory could also include the classical dynamics of a diatom scattered from a metal surface such as N_2 , for which experimental results are available.³ One may use the present theory also to determine the differential angular and energy distribution as well as sticking probabilities.

The fact that the analytical treatment for the angular distribution is in excellent agreement with the numerically exact classical Wigner computation of the distribution suggests that one should extend the present treatment to also study the quantum diffraction, using the semiclassical initial value representation (SCIVR) formalism for the angular scattering. It would be of interest to understand whether the SCIVR approach can account for the experimentally observed diffraction pattern especially in the case of the scattering of the He atom. Preliminary computations show that indeed the SCIVR angular distribution for He is peaked at the specular angle.

Deriving an analytic expression within the SCIVR formalism is a much more formidable challenge, as it is necessary to derive explicit expressions also for the action of the trajectories.

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APPENDIX A: THE CHANGE IN THE SCATTERING ANGLE

The origin of time is chosen as the point at which the unperturbed trajectory in the vertical direction reaches the turning point (z_{tp}) so that at time $t=0$ the vertical velocity vanishes. From Hamilton's equations we have that

$$p_x(t) = p_x + \frac{2\pi V_1}{l} \int_{-t_0}^t dt' \sin\left(\frac{2\pi}{l} x(t')\right) f[z(t')]. \quad (A1)$$

We assume that the corrugation parameter V_1 is small so that to leading order in the coupling constant V_1 , the motion in the horizontal direction is that of a free particle,

$$x(t) \simeq x + \frac{p_x}{M}(t + t_0). \quad (A2)$$

The coupling function $f(z(t))$ is by construction symmetric in time. The momentum at the time t_0 is then given within this perturbation theory by the expression

$$\begin{aligned} p_x(t_0) &\simeq p_x + \frac{4\pi V_1}{l} \sin\left(\frac{2\pi}{l} \left[x + \frac{p_x}{M} t_0\right]\right) \\ &\quad \times \int_0^\infty dt' \cos\left(\frac{2\pi p_x t'}{lM}\right) f[z(t')] \\ &\equiv p_x + \delta p_x. \end{aligned} \quad (A3)$$

Using the conservation of energy and the known shift in the final momentum in the horizontal direction, we deduce to first order the shift in the vertical momentum,

$$p_z(t_0) = -p_z + \frac{p_x}{p_z} \delta p_x. \quad (A4)$$

The incident scattering angle is by definition

$$\theta_i = \tan^{-1}\left(\frac{p_x}{p_z}\right). \quad (A5)$$

The final scattering angle may now be written as

$$\begin{aligned} \theta(t_0) &= -\theta_i - \delta\theta_i \\ &\equiv \tan^{-1}\left(\frac{p_x + \delta p_x(x, p_x, p_z)}{-p_z + \frac{p_x}{p_z} \delta p_x}\right) \\ &\simeq -\theta_i - \frac{\delta p_x(x, p_x, p_z)}{p_z}. \end{aligned} \quad (A6)$$

We thus have that the change induced in the scattering angle

due to the interaction of the vertical and horizontal motions is as given in Eq. (3.1).

APPENDIX B: DERIVATION OF THE ANGULAR DISTRIBUTION IN THE ABSENCE OF DISSIPATION

In this appendix we provide in some detail the manipulations that lead to the expression for the angular distribution, as given in Eq. (3.3). The angular distribution is obtained by averaging the angular shift $\delta\theta_i$ as given in Eq. (3.1) over the initial conditions. For this purpose, we first integrate over the horizontal coordinate. Changing variables from x to $y=x+\zeta$ (using the notation $\zeta=p_x t_0/M$) we find that

$$\begin{aligned} & \int_{-\infty}^{\infty} dx e^{-\gamma(x-x_0)^2} \delta(\theta + \theta_i + \delta\theta_i) \\ &= \int_{-\infty}^{\infty} dy e^{-\gamma(y-\zeta-x_0)^2} \delta\left(\theta + \theta_i + K \sin\left(\frac{2\pi y}{l}\right)\right) \\ &= \frac{1}{2\pi} \int_{-\infty}^{\infty} d\kappa \int_{-\infty}^{\infty} dx e^{-\gamma(x-\zeta-x_0)^2 + i\kappa[\theta + \theta_i + K \sin(2\pi x/l)]}. \end{aligned} \quad (\text{B1})$$

The width γ is now chosen such that $\sqrt{\gamma}l \ll 1$. This then means that $\gamma(x-\zeta-x_0)^2$ can be considered to be a constant as x changes over the interval l . We can then use the following approximation:

$$\begin{aligned} & \int_{-\infty}^{\infty} dx e^{-\gamma(x-y)^2 + i\kappa K \sin(2\pi x/l)} \\ &\simeq \sum_{j=-\infty}^{\infty} \exp\left(-\gamma\left[l\left(j+\frac{1}{2}\right)-y\right]^2\right) \\ &\quad \times \int_0^l dx \exp\left(i\kappa K \sin\left(\frac{2\pi}{l}x\right)\right) \\ &\simeq J_0(\kappa K) \int_{-\infty}^{\infty} dx \exp(-\gamma(x-y)^2) \\ &= \sqrt{\frac{\pi}{\gamma}} J_0(\kappa K), \end{aligned} \quad (\text{B2})$$

where in the second line, the summation has been approximated by an integral. We thus have that

$$\begin{aligned} & \int_{-\infty}^{\infty} dx \exp(-\gamma(x-x_0)^2) \delta(\theta + \theta_i + \delta\theta_i) \\ &\simeq \frac{1}{2\pi} \sqrt{\frac{\pi}{\gamma}} \int_{-\infty}^{\infty} d\kappa \exp(i\kappa[\theta + \theta_i]) J_0(\kappa K) \\ &= \sqrt{\frac{1}{\pi\gamma}} \frac{H(K^2 - (\theta + \theta_i)^2)}{\sqrt{K^2 - (\theta + \theta_i)^2}}, \end{aligned} \quad (\text{B3})$$

where $H(x)$ is the Heaviside function, $H(x)=1$ if $x>0$ and 0 otherwise.

Since this result is independent of the vertical coordinate z , we can readily integrate also over the vertical coordinate so that

$$\begin{aligned} P(\theta) &= \lim_{t \rightarrow \infty} \left(\frac{1}{\pi \hbar \sqrt{\gamma}} \right)^2 \int_{-\infty}^{\infty} dp_x dp_z \\ &\quad \times \exp\left(-\frac{(p_x - p_{x_0})^2 + (p_z - p_{z_0})^2}{\hbar^2 \gamma}\right) \\ &\quad \times \frac{H(K^2 - (\theta + \theta_i)^2)}{\sqrt{K^2 - (\theta + \theta_i)^2}}. \end{aligned} \quad (\text{B4})$$

In the limit that $\gamma \rightarrow 0$ we then find [changing to radial momenta $p^2 = p_x^2 + p_z^2$, $p_0^2 = p_{x_0}^2 + p_{z_0}^2$, $\theta_{i0} = \tan^{-1}(p_{x_0}/p_{z_0})$] the desired result as also given in Eq. (3.3),

$$\begin{aligned} P(\theta) &= \int_0^{\infty} \frac{p dp}{\pi} \int_0^{2\pi} d\theta_i \delta[p - p_0 \cos(\theta_i - \theta_{i0})] \\ &\quad \times \delta[p_0 \sin(\theta_i - \theta_{i0})] \frac{H(K^2 - (\theta + \theta_i)^2)}{\sqrt{K^2(p, \theta_i) - (\theta + \theta_i)^2}} \\ &= \frac{1}{\pi} \frac{H(K^2 - (\theta + \theta_{i0})^2)}{\sqrt{K^2(p_0, \theta_{i0}) - (\theta + \theta_{i0})^2}}. \end{aligned} \quad (\text{B5})$$

APPENDIX C: DERIVATION OF THE ANGULAR DISTRIBUTION IN THE PRESENCE OF DISSIPATION

1. The change in the horizontal momentum

For Ohmic friction, the motion in the horizontal x direction is governed by the Langevin equation

$$\begin{aligned} M\ddot{x}_t - \frac{2\pi V_1}{l} f(z_t) \sin\left(\frac{2\pi x_t}{l}\right) + M g(z_t) \eta[\dot{x}_t g(z_t) \\ + x_t \dot{z}_t g'(z_t)] = \sqrt{M} F(t) g(z_t), \end{aligned} \quad (\text{C1})$$

from which it follows that

$$\begin{aligned} p_x(t_0) &= p_x + \frac{2\pi V_1}{l} \int_{-t_0}^{t_0} dt \sin\left(\frac{2\pi x_t}{l}\right) f(z_t) \\ &\quad - M \eta \int_{-t_0}^{t_0} dt g(z_t) \frac{d}{dt} [x_t g(z_t)] \\ &\quad + \int_{-t_0}^{t_0} dt \sqrt{M} F(t) g(z_t). \end{aligned} \quad (\text{C2})$$

Therefore, to first order in the corrugation strength V_1 and the noise strength, using the fact that initially the particle is far from the surface so that the coupling function $f(z)=0$ and that it is symmetric with respect to time, we have that

$$\begin{aligned} p_x(t_0) &\simeq p_x + \frac{2\pi V_1}{l} \int_{-\infty}^{\infty} dt \sin\left(\frac{2\pi}{l} \left[x + \frac{p_x}{M}(t+t_0)\right]\right) f(z_t) \\ &\quad - \eta p_x \int_0^{\infty} dt g^2(z_t) + \sqrt{M} \int_{-t_0}^{t_0} dt F(t) g(z_t). \end{aligned} \quad (\text{C3})$$

Given the unperturbed trajectory in the vertical direction, one can readily evaluate both the frictional shift

$$\Delta p_{x,1} = -\eta p_x \int_0^\infty dt g^2(z_t) \quad (C4)$$

as well as the noise induced shift

$$\Delta p_{x,2} = 2\sqrt{M} \sum_{j=1}^N c_j h_j \left(x_j \cos(\omega_j t_0) + \frac{p_j}{\omega_j} \sin(\omega_j t_0) \right), \quad (C5)$$

with

$$h_j \equiv \int_0^{t_0} dt g(z_t) \cos(\omega_j t). \quad (C6)$$

This allows us to write down that

$$\begin{aligned} p_x(t_0) &\simeq p_x + p_z K(p_x, p_z) \sin\left(\frac{2\pi}{l} \left[x + \frac{p_x}{M} t_0 \right]\right) \\ &\quad + \Delta p_{x,1} + \Delta p_{x,2} \\ &\equiv p_x + \Delta p_x, \end{aligned} \quad (C7)$$

where $K(p_x, p_z)$ is as defined in Eq. (3.1).

2. The change in the vertical momentum

As noted above, the shift in the vertical momentum is determined by the energy balance. In addition to the change in momentum in the horizontal direction, one must also take into consideration the loss of energy to the bath.

The average energy loss to the bath is given by the expression⁴³

$$\begin{aligned} \langle \Delta E_B \rangle &= \frac{M}{2} \int_{-t_0}^{t_0} dt_1 \int_{-t_0}^{t_0} dt_2 \frac{d[x_{t_1} g(z_{t_1})]}{dt_1} \\ &\quad \times \eta(t_1 - t_2) \frac{d[x_{t_2} g(z_{t_2})]}{dt_2} \\ &= M \eta \int_{-t_0}^{t_0} dt_1 \left(\frac{d[x_{t_1} g(z_{t_1})]}{dt_1} \right)^2, \end{aligned} \quad (C8)$$

where the second equality is specified for Ohmic friction. The fluctuational energy loss is given by the expression

$$\delta E_B = -\sqrt{M} \int_{-t_0}^{t_0} dt \frac{d[x_t g(z_t)]}{dt} F(t). \quad (C9)$$

Since the function $g(z)$ that couples the horizontal motion to the bath is typically localized around the minimum of the potential energy surface or at the turning point for the vertical motion, the term with dg/dt may be neglected in Eq. (C9), so that

$$\langle \Delta E_B \rangle \simeq \frac{\eta p_x^2}{M} \int_{-t_0}^{t_0} dt g^2(z_t) = -\frac{p_x \Delta p_{x,1}}{M} \quad (C10)$$

and

$$\delta E_B \simeq -\frac{p_x}{\sqrt{M}} \int_{-t_0}^{t_0} dt g(z_t) F(t) = -\frac{p_x \Delta p_{x,2}}{M}. \quad (C11)$$

Energy conservation then implies that

$$\frac{p_x^2(t_0) + p_z^2(t_0)}{2M} = \frac{p_x^2 + p_z^2}{2M} - \Delta E_B, \quad (C12)$$

so that the shift in the vertical momentum is given by

$$\Delta p_z = \frac{p_x}{p_z} \Delta p_x + \frac{M \Delta E_B}{p_z}. \quad (C13)$$

3. The change in the scattering angle

Given the shifts in the horizontal and vertical momenta, it is then straightforward to determine the change in the final scattering angle

$$\begin{aligned} \theta(t_0) &= -\theta_i - \delta\theta_i \\ &\equiv \tan^{-1} \left(\frac{p_x + \Delta p_x}{-p_z + \frac{p_x \Delta p_x + M \Delta E_B}{p_z}} \right) \\ &\simeq -\theta_i - \frac{\cos^2(\theta_i)}{p_z} \left(\Delta p_x \left(1 + \frac{p_x^2}{p_z^2} \right) + \frac{M p_x \Delta E_B}{p_z^2} \right), \end{aligned} \quad (C14)$$

so that

$$\delta\theta_i \equiv K(p_x, p_z) \sin\left(\frac{2\pi}{l} \left[x + \frac{p_x}{M} t_0 \right]\right) + \Delta\theta_1 + \Delta\theta_2. \quad (C15)$$

The angular shift is then

$$\Delta\theta_1 = \frac{\Delta p_{x,1}}{p_z} \cos^2 \theta_i, \quad (C16)$$

while the angular fluctuation is

$$\Delta\theta_2 = \frac{\Delta p_{x,2}}{p_z} \cos^2 \theta_i. \quad (C17)$$

4. The temperature dependent angular distribution

We now carry out the integration over the horizontal variable. The procedure is the same as in the case without dissipation and we find

$$\begin{aligned} &\int_{-\infty}^{\infty} dx \exp(-\gamma(x - x_0)^2) \delta(\theta + \theta_i + \delta\theta_i) \\ &\simeq \sqrt{\frac{1}{\pi\gamma}} \frac{H(K^2 - (\theta + \theta_i + \Delta\theta_1 + \Delta\theta_2)^2)}{\sqrt{K^2 - (\theta + \theta_i + \Delta\theta_1 + \Delta\theta_2)^2}}. \end{aligned} \quad (C18)$$

Integrating over the vertical variable, letting the width parameter of the Gaussian wavepacket $\gamma \rightarrow 0$, changing to radial momenta $p^2 = p_x^2 + p_z^2$, $p_0^2 = p_{x_0}^2 + p_{z_0}^2$, $\theta_{i0} = \tan^{-1}(\frac{p_{x_0}}{p_{z_0}})$, we find that

$$\begin{aligned} P(\theta) &= \frac{1}{\pi} \int \prod_{j=1}^N \frac{dp_j dx_j}{2\pi\hbar} \rho_{B,W}(\mathbf{p}, \mathbf{x}) \\ &\quad \times \frac{1}{\sqrt{K^2(p_0, \theta_{i0}) - (\theta + \theta_{i0} + \Delta\theta_1 + \Delta\theta_2)^2}}, \end{aligned} \quad (C19)$$

where it is understood that all variables in the expression are

now functions of the incident energy and scattering angle only and we have omitted the Heaviside function for the sake of brevity.

To carry out the integration over the bath coordinates, we change variables from p_j, x_j to

$$p_j(0) = p_j \cos(\omega_j t_0) - \omega_j x_j \sin(\omega_j t_0), \quad (\text{C20})$$

$$x_j(0) = x_j \cos(\omega_j t_0) + \frac{p_j}{\omega_j} \sin(\omega_j t_0). \quad (\text{C21})$$

Since

$$dp_j(0)dx_j(0) = dp_j dx_j \quad (\text{C22})$$

and

$$p_j^2(0) + \omega_j^2 x_j^2(0) = p_j^2 + \omega_j^2 x_j^2, \quad (\text{C23})$$

the change of variables simplifies the integration over the bath. The dependence on the bath comes from the term $\Delta\theta_2$, which is only dependent on the $x_j(0)$'s. Defining a "generalized bath angle,"

$$Z \equiv \Delta\theta_2 = \frac{2\sqrt{M}}{p_z} \cos^2(\theta_{i0}) \sum_{j=1}^N c_j h_j x_j(0), \quad (\text{C24})$$

we note that

$$\begin{aligned} P(\theta) &= \frac{1}{\pi} \int \prod_{j=1}^N \frac{dp_j dx_j}{\pi} \nu_j \exp\left(-\frac{\nu_j}{\hbar \omega_j} (p_j^2 + \omega_j^2 x_j^2)\right) \frac{H(K^2 - (\theta + \theta_{i0} + \Delta\theta_1 + \Delta\theta_2)^2)}{\sqrt{K^2(p_0, \theta_{i0}) - (\theta + \theta_{i0} + \Delta\theta_1 + \Delta\theta_2)^2}} \\ &= \frac{1}{\pi} \int_{-\infty}^{\infty} dZ \frac{H(K^2 - (\theta + \theta_{i0} + \Delta\theta_1 + Z)^2)}{\sqrt{K^2(p_0, \theta_{i0}) - (\theta + \theta_{i0} + \Delta\theta_1 + Z)^2}} \sqrt{\frac{1}{2\pi\sigma^2}} \exp\left(-\frac{Z^2}{2\sigma^2}\right), \end{aligned} \quad (\text{C25})$$

where the temperature and friction dependent variance of the distribution is

$$\sigma^2 = \frac{2\hbar M}{p_0^2} \cos^2(\theta_{i0}) \sum_j \frac{c_j^2 h_j^2}{\nu_j \omega_j}. \quad (\text{C26})$$

One finds that the variance is readily expressed in the continuum limit by noting that

$$\sum_j \frac{c_j^2 h_j^2}{\nu_j \omega_j} = \frac{2}{\pi} \int_0^{\infty} d\omega J(\omega) \frac{\left(\int_0^{\infty} dt g(z_t) \cos(\omega t)\right)^2}{\tanh \frac{\hbar \beta \omega}{2}}. \quad (\text{C27})$$

Equations (C25)–(C27) are the central result of this Appendix.

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