Nearly elastic scattering and the trajectory approximation

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We consider the scattering of an incident particle by bosons. We assume the coupling is linear in the boson coordinates, but can be quite general in the incident particle coordinate. We show that when the final distribution is close to elastic, the independent-boson model applies to this problem. It yields a loss distribution which is simply the exponentiation of the first-order distorted-wave Born approximation (DWBA). Furthermore, we demonstrate that in this regime, for short wavelengths of the scattered particle, this exponentiated Born approximation reduces to the trajectory approximation (TA). Thus it includes both the DWBA and the TA as special cases. We also give a simple recipe for estimating the error this approximation makes. We illustrate these results on a simple one-dimensional model involving a single oscillator, and discuss their relation to previous studies of overlapping regimes. This approximation and accompanying error estimate should prove very useful in analyzing multiphonon effects in atom-surface scattering.

I. INTRODUCTION

There exists, as yet, no generally solvable theory to describe the scattering of neutral atoms from solid surfaces, even within the Born-Oppenheimer approximation for electronic motion.[1] The problem consists of a continuum of phonons in a semi-infinite crystal interacting with a single-incident atom, and poses an interesting theoretical challenge. Many approximations have been used in different regimes,[2] some without compelling justification. In particular, the trajectory approximation (TA) has been used[3] to analyze scattering results in the semiclassical regime, i.e., where all quantum corrections to classical results are small. In this regime a large number of phonons are excited, as $E_i, E_f, |E_f - E_i| \gg \hbar\omega_D$, where E_i and E_f are the initial and final energies of the incident particle, respectively, and ω_D is the Debye frequency of the target. Furthermore, the de Broglie wavelength of the incident particle is short, so that the Wentzel-Kramers-Brillouin (WKB) approximation is valid for the eigenstates of the potential felt by the incident particle when the lattice is rigid.[4] However, the TA has recently been shown to be inaccurate for semiclassical scattering by Jensen, Chang, and Kohn[5] (hereafter JCK). They found that, for an extremely simple model, the TA fails except when the mean energy transfer is small.

In this paper, we discuss the general problem of nearly elastic scattering of an incident particle by a boson bath. To make clear what we mean by nearly elastic, we consider a Hamiltonian of the form

$$H = H_{\text{bath}} + H_{\text{inc}} + V \ , \tag{1}$$

where H_{bath} describes the bosons, H_{inc} the single incident particle, and V the coupling between them. In general, H_{inc} contains both the kinetic energy of the incident particle and some one-body potential. For example, in atom-surface scattering, this potential is usually chosen to be the static reflection potential, i.e., that potential experienced by the incident atom while all the surface atoms are kept at their equilibrium positions. We denote the eigenstates of H_{inc} by $|\mathbf{k}\rangle$, where $\mathbf{k}$ is the wave vector of the incoming part of the wave function, far from the scattering region. If in Eq. (1) V were zero, the scattering would be purely elastic, i.e., no energy transfer would occur between the incident particle and the bath. Furthermore, the expectation value for the number of particles in state $|\mathbf{k}\rangle$ would be unity for $\mathbf{k}=\mathbf{k}_i$, the initial wave vector, and zero for all others, throughout the duration of the collision. Now for nonzero V, by nearly elastic we mean that this expectation value is dominated by states at or close to $\mathbf{k}_i$ at all times during the collision. For short wavelengths of the incident particle, nearly elastic is equivalent to requiring that the particle's recoilless trajectory is not strongly modified by the coupling to H_{bath}. Note that in nearly elastic scattering, the number of phonons excited in the collision is not restricted, but their cumulative effect on the state of the incident particle must be small.

We illustrate this concept by studying scattering in one dimension from a single oscillator via a purely repulsive interaction. Figure 1 shows some schematic loss spectra for this system. The energy loss of the particle is plotted along the horizontal axis, and the probability of that loss is represented by a vertical column. If the interaction were turned off, then the loss spectrum would be just a column of unit height at the origin. In the presence of the interaction, nearly elastic scattering occurs when ΔE is small in this simple model. However this does not necessarily mean the scattering is weakly inelastic, i.e., has a large elastic fraction as shown in Fig. 1(a). In fact, Fig. 1(b) has the same mean energy loss as Fig. 1(a), although here the scattering is strongly inelastic, i.e., the elastic fraction is negligible.

We solve the nearly elastic scattering problem for interactions which are linear in the displacements of the bosons. We find the independent-boson model[6] applies to this case, leading to a loss spectrum which is just that of linearly driven oscillators. The driving forces are given

simply by the matrix elements of the interaction, evaluated on the uncoupled scattering states. For atom-surface scattering, these are just the matrix elements occurring in the distorted-wave Born approximation. Because of the form of the final spectrum [see Eq. (11)], we call this the exponentiated Born approximation (EBA).

The EBA is an old idea, applied to surface scattering by many previous authors. Unlike the (distorted-wave) Born approximation, it is not an expansion in the number of phonons excited. In the early 1980's, Brenig and coworkers[7–12] thoroughly investigated the application of the EBA to the problem of atoms scattering from phonons at a surface. There it was called the static approximation. However, as early as 1971, Beeby[13] had used it in an estimate of the Debye-Waller factor in this case, and Mueller-Hartmann, Ramakrishnan, and Toulouse[14] had applied it to atom scattering from bosonized electronic excitations. It has also been combined with various other approximations, e.g., short wavelengths of the incoming atom,[15,16] or hard-wall interactions.[17] In fact, it has recently been applied to the scattering of 70-meV He from Pt, with considerable success.[18] Nevertheless, the EBA itself has never been justified on more than intuitive grounds, nor has its entire regime of validity been specified.

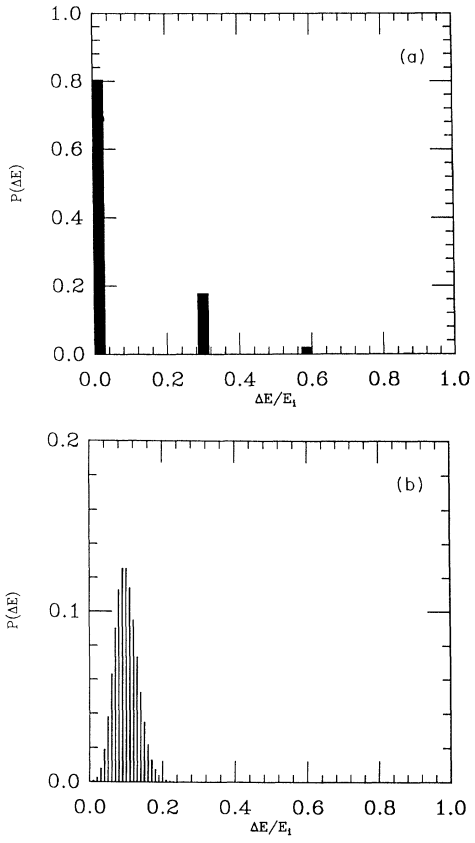

FIG. 1. Schematic quasielastic loss spectra, calculated in the simple model described in Sec. V. The probability is plotted vs energy loss of the incident atom. In (a) there is a large elastic fraction, but in (b) it is negligible, although both have the same $\overline{\Delta E}$.

This paper contains several important results, of both practical and formal significance. To state our formal results precisely, we introduce at this point the term "quasielastic" scattering, which we distinguish from "nearly elastic" scattering. By quasielastic, we mean that $\overline{\Delta E}$ is infinitely small relative to all other energy scales in the system. Quasielastic scattering includes, as special cases, weakly inelastic scattering, semiclassical scattering with small energy transfer, and quasiadiabatic scattering. Brenig argues[7] that the EBA is valid when the scattering is quasielastic. We rigorously demonstrate that quasielasticity is a sufficient requirement for the EBA's validity for the class of Hamiltonians described in Eq. (1).

We can now also quantify what we mean by nearly elastic. Since the EBA yields the leading corrections to elastic scattering, we call the scattering nearly elastic when the error made by the EBA is small, say less than 10%. In fact, our formal derivation provides a simple recipe for estimating the error involved in the use of the EBA for Hamiltonians of the type described by Eq. (1). Thus, in any given situation one can now say just how small $\overline{\Delta E}$ needs to be to make the EBA approximately true, and what is the relevant energy to compare it with.

Our method for estimating the error in the EBA may be easily applied to the full atom-scattering problem so that in real experiments one now has *a priori* condition which, if satisfied, tells one that theoretical calculations based on the EBA can be trusted. This should prove to be of great benefit to the comparison of theory with experiment, given the large number of parameters involved,[19] and the complexity of realistic calculations.[20]

Furthermore, we find that by using a result of Burke and Kohn,[4] the EBA reduces to the TA when $\lambda_i, \lambda_f \ll d$, where λ_i and λ_f are the initial and final wavelengths of the particle, and d is the smallest length scale in the system, in the same sense as used above for quasielastic scattering. Thus, d might be the length scale on which the repulsive part of the potential varies, or the attractive part, etc. Note that short wavelengths do not necessarily imply semiclassical scattering, in the sense defined above. The short-wavelength condition, allowing the WKB approximation in evaluating the scattering matrix elements, can be satisfied independently of the requirement for multiphonon scattering. For example, in quasiadiabatic scattering, the wavelength of the incident particle becomes shorter as its mass grows, keeping E_i and the interaction fixed, but the number of phonons excited remains finite for a cold surface.[4] In fact, our proof also demonstrates the validity of the TA for quasiadiabatic scattering from a linear interaction. This result had been postulated by Burke and Kohn,[4] but only proven for weakly inelastic scattering. This confirmation has already been mentioned in a preliminary report.[21]

Our proof of the validity of the EBA for quasielastic scattering is very general and applies to scattering from a boson bath in any number of dimensions. However, to illustrate our results, we consider scattering from the simple one-dimensional model discussed in Fig. 1. Such a model is one of the simplest possible examples of a system described by Eq. (1) which contains the feature of energy

transfer from the incident particle to the target. The model is *not* intended as a realistic model of all atom-surface scattering. Indeed, it fails to include parallel momentum transfer, diffraction, an attractive well, or a continuum of phonons, all of which can play vital roles in atom-surface scattering.[7-10] The purpose of this model is merely to show how our proof and error estimate work through a very simple example, and develop insight into what nearly elastic means in this simple case. Thus we find conditions for the validity of the EBA which apply *only* to this simple model. However, even here we find that the EBA is valid in a wider range of parameter space than either the distorted-wave (DWBA) or the TA, and a future publication will apply our method for estimating the error to a realistic treatment of atom-surface scattering, including both an attractive well and a phonon continuum, and giving concrete criteria for the validity of the EBA and the TA.

In our one-dimensional model we use an exponential repulsion, linearized in the oscillator's displacement. For this problem, the final distribution within the EBA is simply a Poisson distribution, characterized by a single number, the Debye-Waller exponent η, where $\exp(-\eta)$ is the fraction of purely elastic scattering. We estimate the error involved in the EBA, and show how it approaches zero as any quasielastic limit is approached. We find results consistent with the previous studies of JCK for semiclassical scattering on a similar model.

The bulk of this paper is concerned with a very general proof of the validity of EBA for quasielastic scattering described by Hamiltonians of the form given in Eq. (1). This includes most practical cases of atom-surface scattering. We begin with some formal definitions and a hand-waving argument to give credence to our claims. In Sec. III, we construct a formal proof of our result, using simple operator algebra. In Sec. IV, we give our estimate for the error in the EBA, and discuss its implications for nearly elastic scattering in realistic atom-surface systems. Only in Sec. V do we reintroduce the simple model mentioned above, apply our error estimate, and give conditions for the validity of the EBA for this model. Finally, in Sec. VI we discuss the implications of our results for this field.

II. SIMPLE ARGUMENT

Before presenting a hand-waving argument to demonstrate the plausibility of our claims, we first introduce some essential definitions. For definiteness, we work within the framework of an atom scattering from vibrations in a three-dimensional surface, but emphasize that our conclusions apply generally to a Hamiltonian with the features stated in the Introduction. In this and the next section, we use the notation $\hbar=1$; lowercase bold letters indicate three-dimensional vectors, upper case bold indicate two-dimensional vectors parallel to the surface, the subscript z indicates the component perpendicular to the surface, and italics indicate the length of a vector. Thus, $\mathbf{k}=(\mathbf{K},k_z)$, and $k=|\mathbf{k}|$. The only exception to this is in the position coordinate of the incident atom, denoted $\mathbf{r}=(\mathbf{R},z)$. We also denote the Fourier transform of a function $F(t)$ by $\tilde{F}(\omega)=\int_{-\infty}^{\infty} dt\, e^{i\omega t}F(t)$.

For this problem we have

$$H_{\text{bath}}=\sum_{\mathbf{Q}}\omega_{\mathbf{Q}}a_{\mathbf{Q}}^{\dagger}a_{\mathbf{Q}}\ , \tag{2}$$

$$H_{\text{inc}}=\sum_{\mathbf{k}}\epsilon_k c_{\mathbf{k}}^{\dagger}c_{\mathbf{k}}\ , \tag{3}$$

and

$$V=\sum_{\mathbf{p},\mathbf{k}}V_{\mathbf{p},\mathbf{k}}c_{\mathbf{p}}^{\dagger}c_{\mathbf{k}}a_{\Delta\mathbf{K}}+\text{H.c.}\ , \tag{4}$$

with $\Delta\mathbf{K}=\mathbf{P}-\mathbf{K}$. This model has phonon modes of parallel wave vector $\mathbf{Q}$, energy $\omega_{\mathbf{Q}}$, and creation and annihilation operators $a_{\mathbf{Q}}^{\dagger}$ and $a_{\mathbf{Q}}$. These are regarded as periodic functions of their arguments, with the period of the two-dimensional surface reciprocal lattice. The sum over all branches with parallel wave vector $\mathbf{Q}$ is implicit. The operators $c_{\mathbf{p}}^{\dagger}$ and $c_{\mathbf{k}}$ create and destroy incident particles in distorted-wave states labeled $\mathbf{p}$ and $\mathbf{k}$, respectively. These states are scattering states from the static reflection potential $U(\mathbf{r})$, i.e., the potential experienced by the incident atom when the surface atoms are held fixed. They are normalized so that the incoming part of the wave function in real space is just $\exp(i\mathbf{k}\cdot\mathbf{r})/\sqrt{\Omega}$, far from the surface, where Ω is the volume of a large box containing the entire system. In general, their outgoing parts include diffracted beams. Their energies are just $\epsilon_k=k^2/2m$ in the continuum. We also implicitly include in the sum over $\mathbf{k}$ in Eq. (3) any bound states of $U(z)$ which, if they exist, we assume to be at discrete energies and of finite number.

We often work in the interaction representation, in which the time dependence of the operators is generated by the noninteracting Hamiltonian for the entire system, $H_0=H_{\text{bath}}+H_{\text{inc}}$. In this picture, the particle-surface interaction may be written as

$$V_I(t)=\sum_{\mathbf{p},\mathbf{k}}V_{\mathbf{p},\mathbf{k}}c_{\mathbf{p}}^{\dagger}(t)c_{\mathbf{k}}(t)a_{\Delta\mathbf{K}}(t)+\text{H.c.}\ , \tag{5}$$

where $c_{\mathbf{k}}(t)=c_{\mathbf{k}}e^{-i\epsilon_k t}$ and $a_{\mathbf{Q}}(t)=a_{\mathbf{Q}}e^{-i\omega_Q t}$.

The zero-temperature energy-loss spectrum (the generalization to finite temperatures and angular distributions is straightforward) may then be expressed as

$$P(\Delta E)=\langle\phi_f|\delta(H_{\text{bath}}-\Delta E)|\phi_f\rangle\ , \tag{6}$$

where $|\phi_f\rangle$ is the final state of the complete system (after the scattering has occurred). The dynamics of the scattering system are contained in how $|\phi_f\rangle$ is generated from the initial state of the system $|\mathbf{k}_i,0\rangle$, where $\mathbf{k}_i$ is the initial wave vector of the incident atom and $|0\rangle$ represents the ground state of the crystal. We write

$$|\phi_f\rangle=S|\mathbf{k}_i,0\rangle\ , \tag{7}$$

where S is the scattering matrix due to the interaction V and can, at least formally, be expressed as a power series in V.

Using these definitions, we may present our hand-waving argument. In the interaction representation we write, for the effect of the potential on the initial state of the system,

$$V_I(t)|\mathbf{k}_i,0\rangle=\sum_{\mathbf{k}} V^*_{\mathbf{k}_i,\mathbf{k}} e^{i(\epsilon_p-E_i-\omega_{\Delta\mathbf{K}})t}|\mathbf{k},\Delta\mathbf{K}\rangle\ , \tag{8}$$

where $|\mathbf{k},\mathbf{Q}\rangle$ is a product state with the incident atom in state $\mathbf{k}$ and a single phonon of parallel momentum $\mathbf{Q}$ ($=\Delta\mathbf{K}$ up to a surface reciprocal-lattice vector) excited in the target. However, in the quasielastic regime the state of the incident particle is almost unchanged, so that $|\mathbf{k},\mathbf{Q}\rangle\simeq|\mathbf{k}_i,\mathbf{Q}\rangle$. The same reasoning applies when the state $|\mathbf{k}_i,\mathbf{Q}\rangle$ is acted upon by the potential, and again the wave vector of the incident atom remains approximately unchanged. We extend this argument to all scattering processes throughout the collision. Thus, in our calculation, we may write, approximately

$$V_I(t)\simeq\sum_{\mathbf{p}} V_{\mathbf{p},k_i} e^{i(\epsilon_p-E_i-\omega_{\Delta\mathbf{K}})t} a_{\Delta\mathbf{K}}+\text{H.c.} \tag{9}$$

or, in the more familiar Schrödinger representation,

$$V\simeq\sum_{\Delta\mathbf{K}} U_{\Delta\mathbf{K}}(t)a_{\Delta\mathbf{K}}+\text{H.c.}\ , \tag{10}$$

where $U_{\Delta\mathbf{K}}(t)=\sum_{p_z} V_{\mathbf{p},\mathbf{k}_i} e^{-i(E_i-\epsilon_p)t}$ and $\mathbf{P}=\mathbf{K}+\Delta\mathbf{K}$. The particle is no longer coupled to the surface phonons, and the full Hamiltonian just describes a set of oscillators that are linearly driven by the potential. The exact solution to this problem is given simply by[6]

$$\mathcal{N}(\tau)=\exp\left[-\sum_{\Delta\mathbf{K}}|\tilde{U}_{\Delta\mathbf{K}}(-\omega_{\Delta\mathbf{K}})|^2(1-e^{-i\omega_{\Delta K}\tau})\right]\ , \tag{11}$$

and $P(\Delta E)=\tilde{\mathcal{N}}(\Delta E)/2\pi$, where $\tilde{\mathcal{N}}(\omega)$ is the Fourier transform of $\mathcal{N}(\tau)$, and

$$\tilde{U}_{\Delta\mathbf{K}}(\omega)=2\pi V_{\mathbf{p},\mathbf{k}_i}\delta(\omega+E_i-\epsilon_p) \tag{12}$$

is the Fourier transform of $U_{\Delta\mathbf{K}}(t)$. In this last result, $\mathbf{p}$ is uniquely determined by energy and momentum conservation,

$$\mathbf{P}=\mathbf{K}+\Delta\mathbf{K}\ ,\quad p_z=\sqrt{k^2-P^2+2m\omega}\ . \tag{13}$$

Because of the form of Eq. (11), we call this the exponentiated Born approximation, as the matrix elements $\tilde{U}_{\Delta\mathbf{K}}(-\omega_{\Delta\mathbf{K}})$ are just those appearing in the distorted-wave Born approximation. This completes our handwaving derivation for the validity of the EBA for quasielastic scattering. This result will now be derived more carefully.

III. VALIDITY OF THE EBA FOR QUASIELASTIC SCATTERING

In this section we show that the EBA is valid for quasielastic scattering from a linear interaction. The proof involves finding the final distribution of states of the bosons, and deducing the distribution of states of the incident particle by conservation rules.

We begin with some formal scattering theory, and illustrate how it yields the correct loss spectrum for a linearly driven oscillator. We then apply this formalism to the full scattering problem, and show how the interaction behaves just as a time-dependent driving force on the bosons when the scattering is quasielastic. This occurs because boson creation and annihilation processes become statistically independent in this case. In the next section we will discuss how this limit is achieved for a simple model.

We employ a formalism that is particularly well suited to solving the driven oscillator problem.[21] The scattering matrix introduced in Eq. (7) is simply the $t\to\infty$ limit of the evolution operator for the complete system, $U(t,t_0)\equiv e^{-iH(t-t_0)}$, where H is the total Hamiltonian for the entire system and t_0 is taken to $-\infty$. In the interaction representation we write

$$U_I(t,t_0)\equiv e^{iH_0t}U(t,t_0)e^{-iH_0t_0}=e^{-iG(t,t_0)} \tag{14}$$

and it is straightforward to show that $G\equiv G(\infty,-\infty)$ has the following expansion in powers of the potential in the interaction representation,[22]

$$G=G^{(1)}+G^{(2)}+G^{(3)}+\cdots\ , \tag{15}$$

where

$$G^{(1)}=\int_{-\infty}^{\infty}V_I(t)dt\ , \tag{16}$$

$$G^{(2)}=-\frac{i}{2}\int_{-\infty}^{\infty}dt\int_{-\infty}^{t}dt'[V_I(t),V_I(t')]\ , \tag{17}$$

etc. The series becomes increasingly complex as one goes to higher orders, but here we use only the property that each $G^{(n)}$ is a linear combination of terms, each of which involves the commutation of n $V_I(t)$'s together. The Fourier transform of the energy-loss spectrum defined by Eq. (6) is then (up to a factor of 2π)

$$\mathcal{N}(\tau)=\langle\mathbf{k}_i,0|e^{i(H_{\text{bath}}+W)\tau}|\mathbf{k}_i,0\rangle\ , \tag{18}$$

where

$$W=e^{iG}H_{\text{bath}}e^{-iG}-H_{\text{bath}}\ . \tag{19}$$

To actually calculate W, we expand in powers of G to find $W=W^{(1)}+W^{(2)}+\ldots$, where

$$W^{(n)}=\frac{i^n}{n!}\{G^n,H_{\text{bath}}\} \tag{20}$$

and $\{G^n,H_{\text{bath}}\}\equiv[G,[G,\ldots,H_{\text{bath}}]\ldots]$, i.e., G commuted with H_{bath} n times. Finally, once W has been found, $\mathcal{N}(\tau)$ may be expressed as a cumulant expansion,[23]

$$\ln\mathcal{N}(\tau)=\gamma^{(1)}(\tau)+\gamma^{(2)}(\tau)+\ldots\ , \tag{21}$$

where

$$\gamma^{(1)}(\tau)=-i\int_0^{\tau}d\tau'\langle W(\tau')\rangle_c\ , \tag{22}$$

$$\gamma^{(2)}(\tau)=-\int_0^{\tau}d\tau'\int_0^{\tau'}dt''\langle W(\tau')W(\tau'')\rangle_c\ , \tag{23}$$

and so forth, where the cumulants are evaluated on the initial state $|\mathbf{k}_i,0\rangle$ and the τ dependence of $W(\tau)$ is generated by H_{bath} alone, i.e., $W(\tau)=e^{iH_{\text{bath}}\tau}We^{-iH_{\text{bath}}\tau}$. The cumulants are defined in Ref. 23, and involve expectation values with means subtracted. For example, $\langle W(\tau)\rangle_c=\langle W(\tau)\rangle$, $\langle W(\tau)W(\tau')\rangle_c=\langle W(\tau)W(\tau')\rangle-\langle W(\tau)\rangle\langle W(\tau')\rangle$, etc. Much of this formalism is simi-

lar to that used by Brenig.[7]

We demonstrate how this rather elaborate scheme works by applying it to a simple system of linearly driven oscillators, with

$$V_I(t)=\sum_{\mathbf{Q}} U_{\mathbf{Q}}(t)[a_{\mathbf{Q}}(t)+a^{\dagger}_{\mathbf{Q}}(t)] \ . \tag{24}$$

Using the commutation relations of the phonon operators, it is clear from Eqs. (16) and (17) that the only relevant contribution to G is $G^{(1)}$. [All others vanish except $G^{(2)}$, which is just a c number, and therefore does not contribute to W in Eq. (19)]. For the same reasons, only the first two terms in the expansion of W [Eq. (20)] survive, namely

$$W^{(1)}=i\sum_{\mathbf{Q}} \omega_{\mathbf{Q}}[\tilde{U}_{\mathbf{Q}}(-\omega_{\mathbf{Q}})a_{\mathbf{Q}}-\tilde{U}^{*}_{\mathbf{Q}}(-\omega_{\mathbf{Q}})a^{\dagger}_{\mathbf{Q}}] \ , \tag{25}$$

$$W^{(2)}=\sum_{\mathbf{Q}} \omega_{\mathbf{Q}}|\tilde{U}_{\mathbf{Q}}(-\omega_{\mathbf{Q}})|^2 \ . \tag{26}$$

As W contains, at most, linear powers of phonon operators, all cumulants vanish except the first and second,[24] whose sum yields

$$\gamma(\tau)=\sum_{\mathbf{Q}} |\tilde{U}_{\mathbf{Q}}(-\omega_{\mathbf{Q}})|^2(e^{-i\omega_{\mathbf{Q}}\tau}-1) \ , \tag{27}$$

which, when substituted into Eq. (21), gives the loss spectrum already quoted in Sec. I [see Eq. (11)].

We are not ready to tackle the full scattering problem. We use the Hamiltonian introduced in Sec. I [see Eqs. (2)–(4)], so the potential in the interaction representation is given by Eq. (5). For convenience, we rewrite this as

$$V_I(t)=\sum_{\mathbf{k},\Delta\mathbf{K}} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \bar{V}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},\omega)\rho(\mathbf{K},\epsilon_k;\Delta\mathbf{K},\omega) \times e^{i\omega t}a_{\Delta\mathbf{K}}(t)+\text{H.c.} \ , \tag{28}$$

where we have transformed the sum over p_z into an integral over energies. In the above, we have relabeled $\mathbf{k}$ with the set $(\mathbf{K},\epsilon_k)$ and $\mathbf{p}$ with $(\mathbf{K}+\Delta\mathbf{K},\epsilon_k+\omega)$. We have defined $\rho(\mathbf{K},\epsilon_k;\Delta\mathbf{K},\omega)=c^{\dagger}_{\mathbf{p}}c_{\mathbf{k}}$, and $\bar{V}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},\omega)=(mL_z/p_z)V_{\mathbf{p},\mathbf{k}}$, where L_z is the box length in the z direction. Thus, $\bar{V}$ is the interaction matrix element evaluated on distorted-wave states, normalized to unit incident flux. A factor $L_z/(2\pi)$ appears in transforming from discrete to continuous p_z, and a factor m/p_z appears in the transformation to energies from wave vectors.

With this form for the potential, we may calculate the contributions to G, as given by Eqs. (16) and (17). We find

$$G^{(1)}=\sum_{\mathbf{k},\Delta\mathbf{K}} \bar{V}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},\omega_{\Delta\mathbf{K}})\rho(\mathbf{K},\epsilon_k;\Delta\mathbf{K},\omega_{\Delta\mathbf{K}})a_{\Delta\mathbf{K}} +\text{H.c.} \tag{29}$$

and

$$G^{(2)}=\sum_{\mathbf{k},\Delta\mathbf{K},\Delta\mathbf{K}'} \int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi} \times\bar{C}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},\omega;\Delta\mathbf{K}',\omega') \times\rho(\mathbf{K},\epsilon_k;\Delta\mathbf{K}+\Delta\mathbf{K}',\omega+\omega') \times\tilde{A}(\Delta\mathbf{K},\omega;\Delta\mathbf{K}',\omega') \ , \tag{30}$$

where

$$\bar{C}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},\omega;\Delta\mathbf{K}',\omega')=\bar{V}(\mathbf{K}+\Delta\mathbf{K}',\epsilon_k+\omega';\Delta\mathbf{K},\omega) \times\bar{V}(\mathbf{K},\epsilon_k;\Delta\mathbf{K}',\omega') -\bar{V}(\mathbf{K}+\Delta\mathbf{K},\epsilon_k+\omega;\Delta\mathbf{K}',\omega') \times\bar{V}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},\omega) \tag{31}$$

is the difference of matrix elements produced by the commutator in Eq. (17) and

$$\tilde{A}(\Delta\mathbf{K},\omega;\Delta\mathbf{K}',\omega')=-\frac{i}{2}\int_{-\infty}^{\infty}dt\int_{-\infty}^{t}dt'[a_{\Delta\mathbf{K}}(t)+a^{\dagger}_{-\Delta\mathbf{K}}(t)][a_{\Delta\mathbf{K}'}(t')+a^{\dagger}_{-\Delta\mathbf{K}'}(t')]e^{i\omega t+i\omega' t'} \tag{32}$$

is the combination of phonon operators surviving in $G^{(2)}$. When we say surviving, we refer to the fact that any parts of $G^{(2)}$ which are diagonal in both particle and boson space do not contribute to the final distribution, just as in the driven oscillator case. Such terms have been dropped from Eq. (30).

The important feature of $G^{(2)}$ for our purposes is in the matrix element combination given by Eq. (31). Note that if $\bar{V}$ were independent of its first arguments, i.e., depended only on the energy and momentum transfer, and not on the initial state of the particle, then $G^{(2)}$, and all higher-order contributions, would vanish identically. This is the typical case to which the independent boson model is applied,[6] and corresponds to a structureless interaction. For our class of problems, this will not generally be true. However, for quasielastic scattering, we expect the dominant states occurring in the integrals to be close in $\mathbf{k}$ space to the initial state. We expand the matrix elements about the initial state of the particle, i.e.,

$$\bar{V}(\mathbf{K}_i+\Delta\mathbf{K}',E_i+\omega';\Delta\mathbf{K},\omega)=\bar{V}(\mathbf{K}_i,E_i;\Delta\mathbf{K},\omega)+\Delta\mathbf{K}'\left.\frac{\partial\bar{V}(\mathbf{K},E_i;\Delta\mathbf{K},\omega)}{\partial\mathbf{K}}\right|_{\mathbf{K}=\mathbf{K}_i}+\omega'\left.\frac{\partial\bar{V}(\mathbf{K}_i,E;\Delta\mathbf{K},\omega)}{\partial E}\right|_{E=E_i}+\cdots \ . \tag{33}$$

Thus we have expanded in small changes in the initial state of the particle, keeping the energy and momentum exchange fixed. We show below that the EBA is recovered by replacing $\bar{V}(\mathbf{K}_i+\Delta\mathbf{K}',E_i+\omega';\Delta\mathbf{K},\omega)$ by $\bar{V}(\mathbf{K}_i,E_i;\Delta\mathbf{K},\omega)$ in Eq. (31) and wherever it appears in the definition of the $G^{(n)}$'s.

Recall that in the Introduction we defined quasielastic scattering as being infinitely close to elastic scattering. Clearly the replacement described above becomes exact in this limit. Also, we can now see what is required for nearly elastic scattering. The derivative terms in Eq. (33) must not significantly affect the state distribution. Thus, in any particular situation, the sensitivity of the matrix elements to the initial state of the particle will determine just how large an error is made in using the EBA, and so how wide a region of parameter space can be called nearly elastic. Note also that since Eq. (30) contains sums and integrals over virtual intermediate states, we require the cumulative effect of the dropped terms to be small.

By inserting this form for $\bar{V}(\mathbf{K}_i+\Delta\mathbf{K}',E_i+\omega';\Delta\mathbf{K},\omega)$ in $G^{(2)}$ in Eq. (30) we find that $\bar{C}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},\omega;\Delta\mathbf{K}',\omega')$ vanishes so that $G^{(2)}$ is identically zero, and, in fact, all $G^{(n)}=0$ for $n>1$. Thus, for quasielastic scattering, G is almost identical in form to the driven oscillator case. Similar reasoning applies to the calculation of W, and we find a simple generalization of the linearly driven oscillator result,

$$W^{(1)}=i\sum_{\mathbf{k},\Delta\mathbf{K}}\omega_{\Delta\mathbf{K}}\{\bar{V}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},\omega_{\Delta\mathbf{K}})\rho(\mathbf{K},\epsilon_k;\Delta\mathbf{K},\omega_{\Delta\mathbf{K}})a_{\Delta\mathbf{K}} - \bar{V}^*(\mathbf{K},\epsilon_k;\Delta\mathbf{K},-\omega_{\Delta\mathbf{K}}) \times\rho(\mathbf{K},\epsilon_k;\Delta\mathbf{K},-\omega_{\Delta\mathbf{K}})a^\dagger_{\Delta\mathbf{K}}\} \quad (34)$$

$$W^{(2)}=\sum_{\mathbf{k}}c^\dagger_{\mathbf{k}}c_{\mathbf{k}}\sum_{\Delta\mathbf{k}}\omega_{\Delta\mathbf{K}}|\bar{V}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},-\omega_{\Delta\mathbf{K}})|^2 . \quad (35)$$

Finally, the same approximation, Eq. (33), may also be used to show the cumulant expansion is just as in the linearly driven oscillator case, i.e., only the first and second cumulants are nonzero. The only difference with the driven oscillator problem is that W is now an operator on incident particle states. But the approximation of Eq. (33) is equivalent to allowing matrix elements with differing particle energies to commute, i.e., to having consecutive phonon creation and annihilation events statistically independent. Then, by the basic theorem on cumulants,[23] the only surviving cumulants are those that survive for the driven oscillator problem. Thus we find

$$\gamma(t)=\sum_{\Delta\mathbf{K}}|\bar{V}(\mathbf{K}_i,E_i;\Delta\mathbf{K},-\omega_{\Delta\mathbf{K}})|^2(e^{-i\omega_{\Delta\mathbf{K}}t}-1) , \quad (36)$$

where the cumulant has been evaluated on the initial state $|\mathbf{k}_i,0\rangle$. Note that the only approximation used to achieve this result was the replacement described after Eq. (33).

To include the angular distribution in this proof, Eq. (6) is generalized to an operator which also projects out the final direction of the scattered particle, and the argument given above goes through. To include surfaces at finite temperatures, one simply sums over a thermally weighted distribution of initial phonon states in Eq. (11), to each of which the above argument applies. This leads to a simple generalization of the above results. We define the driving function as

$$\tilde{D}_{\mathbf{k}}(\Delta\mathbf{K},\omega)=(2\pi)^3\{|\bar{V}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},-\omega)|^2\times[1+n(\omega)]g(\Delta\mathbf{K},\omega) +|\bar{V}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},\omega)|^2\times n(-\omega)g(\Delta\mathbf{K},-\omega)\} . \quad (37)$$

Here, the density of states of bosons with momentum $\Delta\mathbf{K}$ is denoted $g(\Delta\mathbf{K},\omega)$, and $n(\omega)$ is the Bose occupation factor, at the surface temperature. This equation includes all branches of phonons in the target and even static corrugation may be included through the details of $g(\Delta\mathbf{K},\omega)$. Then the final distribution, calculated within the EBA, is just

$$P_{\mathrm{EBA}}(\Delta\mathbf{K},\epsilon)=\int_{-\infty}^{\infty}\frac{dt}{2\pi}\int\frac{d^2R}{4\pi^2}e^{i\epsilon t+i\Delta\mathbf{K}\cdot\mathbf{R}}\times\exp[D(\mathbf{R},t)-D(\mathbf{0},0)] , \quad (38)$$

where $D(\mathbf{R},t)$ is the inverse Fourier transform of the driving function, and the normalization has been chosen such that $\int_{-\infty}^{\infty}d\epsilon\int d\Delta\mathbf{K}P_{\mathrm{EBA}}(\epsilon,\Delta\mathbf{K})=1$. This is the complete solution to the scattering problem within the EBA.

In discussing the EBA distribution, we first note that, for weakly inelastic scattering, the exponential $\exp[D(\mathbf{R},t)]$ may be expanded about 1, yielding a distribution

$$P(\Delta\mathbf{K},\epsilon)=[1-D(\mathbf{0},0)]\delta(\epsilon)\delta^2(\Delta\mathbf{K}) +\tilde{D}_{\mathbf{k}}(\Delta\mathbf{K},\omega)/(2\pi)^3 . \quad (39)$$

This is just the familiar distorted-wave Born approximation result.

Next, we discuss how, when the wavelengths involved are short, the TA is recovered. For asymptotically short wavelengths, the wave functions involved in evaluating the driving function $D_{\Delta\mathbf{K}}$ may be approximated by their WKB forms.[25] Furthermore, it can be shown[4] that the difference in their phases, as a function of z, becomes simply $\omega_{\Delta\mathbf{K}}t(z)$, where $t(z)$ is the inverse of $z_{\mathrm{cl}}(t)$, the elastic classical trajectory. The resulting matrix elements are precisely the Fourier transform of the classical force, as prescribed by the TA. This result was earlier argued to be true by Sedlmeir and Brenig.[8] When inserted into the driving function [Eq. (37)], the EBA loss spectrum is precisely that of the (recoilless) TA, for a linear coupling.[3]

These results for the matrix elements are derived in detail in Appendix C of Ref. 4 for quasiadiabatic scattering. In that limit, $m\to\infty$, with E_i and the interaction fixed. Thus, $\overline{\Delta E}\to 0$, as the scattering becomes increasingly adiabatic, and the EBA becomes valid. Furthermore, $k_i d\to\infty$, so the EBA becomes the TA. So our proof of the EBA for quasielastic scattering, when combined with previous results, is also a proof of the TA for quasiadiabatic scattering from a linear coupling. Previously this

had only been shown to be valid for weakly inelastic scattering, and speculated to be valid more generally.[4]

IV. ERROR ESTIMATE FOR THE EBA

Next we use our proof to make an estimate of the error involved in employing the EBA. We point out that the spectrum prescribed by the EBA may always be calculated for a given system. It simply will not be a good approximation to the true spectrum unless conditions close to those of the above proof prevail. The essential assumption of the proof, Eq. (33), is that we may neglect errors due to the dependence of the matrix elements on the particle state occurring within the calculation. These will vanish in any limit which approaches elastic scattering. However, we would like to be able to estimate the size of these errors to get some insight into how close to elastic one needs to be.

The earliest point in the proof where we use this assumption is when we neglect $G^{(2)}$ in favor of $G^{(1)}$. We therefore use the ratio of these two terms as an estimate of the error, which we denote by Δ. We write

$$\Delta^2=\langle\phi|[G^{(2)}]^2|\phi\rangle/\langle\phi|[G^{(1)}]^2|\phi\rangle\ , \tag{40}$$

where $|\phi\rangle$ is some typical state of the system during the collision process. We use squares to ensure that the operators have diagonal matrix elements. In $G^{(2)}$, we include only those terms which either create or destroy two phonons simultaneously, as the other parts yield relaxations which do not contribute to final loss distribution. For a typical state of the system, we take the particle wave vector to be $\mathbf{k}_i$, as we expect our estimate to be meaningful only when the scattering is nearly elastic. We characterize the boson state by $\bar{n}$ excitations, where $\bar{n}$ is the mean number of excitations in the bath after the collision. This should yield a crude estimate of the error involved in using the EBA, and should be sufficient for determining qualitative trends. We will say the EBA "works" when Δ is smaller than some chosen small value, say 10%. The Appendix contains the details of how this estimate is made for the simple model introduced in the next section.

To understand intuitively why Δ estimates the error in the EBA, consider the prescription for calculating $G^{(2)}$ given in Eqs. (30)–(32). We can see that $G^{(2)}$ is an amplitude for correlated two-phonon emission and annihilation by the incoming particle. The EBA excludes the contribution from such events. Thus the particle during the collision has no "memory" of previous events, as it only ever emits or annihilates one phonon at a time, and always as if from the initial state of the particle.

The remainder of this section consists of a brief qualitative discussion of how Δ becomes small in the regimes where the EBA works. The most obvious regime in which Δ is small is when the scattering is weakly inelastic. Here, $\bar{V}\ll 1$, and, as $G^{(2)}$ is proportional to $\bar{V}^2$, while $G^{(1)}$ is proportional to $\bar{V}$, this means Δ is proportional to $\bar{V}$. As stated above, the EBA becomes the simple first-order distorted-wave Born approximation, and is valid here.

However, another quite distinct regime also exists in which Δ becomes small. The operator $G^{(2)}$ involves the commutator of various $\bar{V}$ operators [Eq. (31)] which may cancel one another, even when $|\bar{V}|$ is not small. In particular, if $\bar{V}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},-\omega)$ were independent of $\mathbf{K}$ and ϵ_k, then $G^{(2)}$ (and therefore Δ) would vanish. Thus Δ will be small when $\bar{V}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},-\omega)$ does not depend strongly on $(\mathbf{K},\epsilon_k)$ in the vicinity of $(\mathbf{K}_i,E_i)$. To be precise, if $\epsilon_k=E_i+\omega$ and $\mathbf{K}=\mathbf{K}_i+\Delta\mathbf{K}$, then $\bar{V}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},-\omega)$ need only be approximately independent of $(\epsilon_k,\mathbf{K})$ for those values of ω and $\Delta\mathbf{K}$ for which $\bar{V}(\mathbf{K},\epsilon_k;\Delta\mathbf{K},-\omega)$ is nonnegligible (relative to, say, its peak value). To illustrate this, Fig. 2 contains plots of $\bar{V}$ (for the simple model to be described in the next section) as a function of energy transfer. The model is one dimensional, so that parallel momentum plays no role. κ is a dimensionless measure of the initial wave vector. The specific values of the parameters used in the plots are given in Sec. V. Each plot contains $\bar{V}$ for two values of κ, differing by the same amount in each figure. The higher peak is always associated with the larger value of κ. In (a), $\kappa=1$ for one line, and $\kappa=0.7$ for the other. Thus, a decrease of 0.3 in κ leads to a substantial change in the matrix elements. Furthermore, the peaks are relatively broad, giving a large contribution to the integrals in Eq. (30). In (b), the lines are drawn for $\kappa=2.3$ and 2, and the difference is much smaller. Finally, in (c), $\kappa=6.3$ and 6, the difference is negligible, and the matrix elements are much more sharply peaked about 0, leaving a much narrower region in which they need be independent of ϵ_k. Thus, for an initial κ of 6.3, if the scattering only involves changes in κ of about 0.3, all matrix elements (of a given energy transfer) involved in the collision are about the same. Then the commutator in Eq. (31) is very small, and the corrections to EBA negligible. This conclusion is clearly not true for an initial κ of 1, where the matrix elements depend strongly on the particular value of κ.

A final important point must be made here. The relative insensitivity of $\bar{V}$ to changes in k is not a sufficient condition for the validity of the EBA, as it implies only that the change in matrix elements is small for a *single* excitation of the surface. However, in some regimes, there may be a large number of bosons excited, leading to a substantial net change in k and therefore a breakdown in the EBA. Indeed, as is discussed in some detail in Sec. V, this is exactly what happens as the semiclassical limit is approached, so that the EBA is *not* generally valid in this limit. All the above information is, of course, implicitly carried in the estimate of Δ.

We conclude this section with a brief discussion of how Δ might be expected to behave for realistic atom scattering from surfaces, simply to highlight some of the differences from the results of the simple model discussed below. For example, that model does not contain an attractive well. In the presence of such a well, especially one whose depth D is much larger than the energy of the incident particle, considerable sticking could occur.[9] Then, the mean energy transfer would be comparable to the initial energy. However, this energy transfer would still be small relative to the effective incident energy as given by the Beeby corrections,[1] E_i+D, so that the EBA approximation in Eq. (33) would still be good. In such a

case, the naive criterion of small fractional energy transfer would be too restrictive, as argued by Brenig.[7] Similarly, the presence of resonances connected with bound states of the well[12,26] and the presence of a continuum of phonons[11] will modify statements of when the EBA applies, but in all cases a calculation of Δ as given in Eq. (40) will indicate how well the EBA is working in any given situation. In fact, the effect of such features will form the basis of a future paper. For now, however, we study the simple model to illustrate how Δ works.

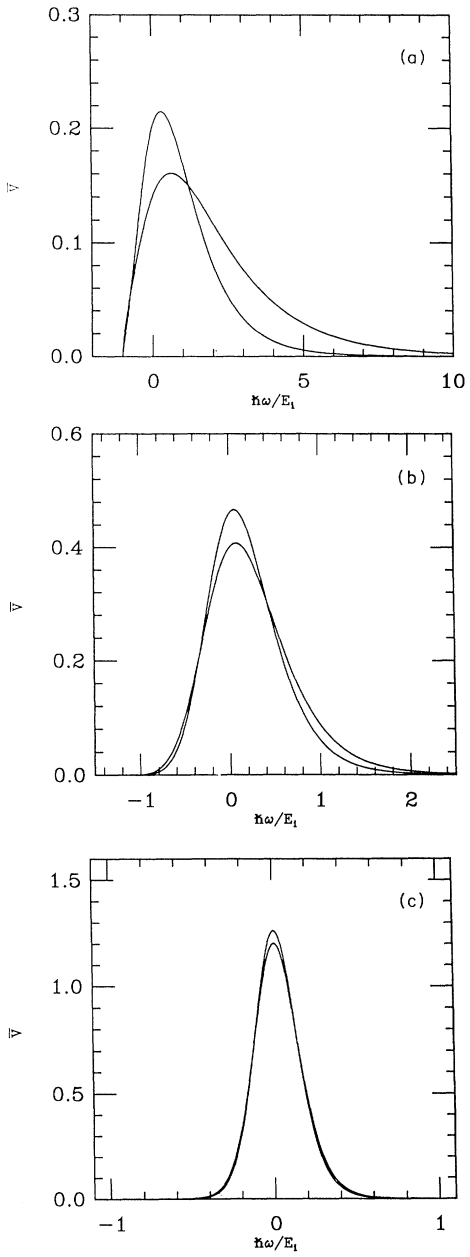

FIG. 2. Plots of $\bar{V}$ as a function of energy transfer for the model described in Sec. V. Each plot displays two values of κ, differing by 0.3. The more strongly peaked function is always the larger κ. In (a), $\kappa=1, 0.7$; in (b), $\kappa=2.3, 2$; in (c), $\kappa=6.3, 6$.

V. A SIMPLE MODEL

To illustrate the proof of the previous section, we study the properties of a simple one-dimensional model. This keeps the number of independent parameters manageable,[19] while allowing an intuitive understanding of why EBA works where it does. In this model, one sees the EBA spanning a region of parameter space which includes both that of the TA and of the DWBA. The model allows for a transfer of energy from an incident particle to a target, but little else. It contains no directions parallel to the surface, so that we need no longer keep track of parallel momentum. It tells us nothing about situations with an attractive well, nor with a continuum of phonons. It is not intended as a realistic model of atom-surface scattering. The results of this section cannot be taken as applying to all cases of atom-surface scattering. Such complications will be dealt with in a future publication.

To simplify the expression of results, we change notation slightly. We now restore $\hbar$ explicitly, and we always use k and E to denote initial values of the incident atom's wave vector and energy, respectively.

In the spirit of JCK, we consider an incident particle scattering from a single oscillator via a (linearized) repulsion. The Hamiltonian is

$$H=\frac{P^2}{2M}+\tfrac{1}{2}M^2\omega_0^2Z^2+\frac{p^2}{2m}+V(z)+F(z)Z \ . \tag{41}$$

Here, P, M, Z and ω_0 denote the oscillator's momentum, mass, position, and frequency, respectively; p, m, and z are the same quantities for the incident atom; and $V(z-Z)$ is the repulsive interaction between them, with $F(z)=-dV/dz$. The position and momenta are quantum-mechanical operators. The matrix elements of the interaction are then

$$\bar{V}(\epsilon_k;\omega)=F_k(\omega)u_0 \ , \tag{42}$$

where $F_k(\omega)=|\langle k'|F(z)|k\rangle|$, $k'=\sqrt{k^2+2m\omega/\hbar}$, and $u_0^2=\hbar/2M\omega_0$ is the zero-point mean-square displacement of the oscillator. The states $|k\rangle$ are here normalized to unit incident flux. Note that $\bar{V}(\epsilon_k;\omega)$ vanishes for all $\omega<-\epsilon_k/\hbar$. This yields a zero-temperature driving function [see Eq. (37)] of

$$\tilde{D}(\omega)=F_k^2(-\omega)u_0^2\delta(\omega-\omega_0) \ , \tag{43}$$

from which the EBA loss spectrum can be easily calculated, using Eq. (11),

$$P(\Delta E)=e^{-\eta}\sum_{n=0}^{[E/\hbar\omega_0]}\frac{\eta^n}{n!}\delta(\Delta E-n\hbar\omega_0) \ , \tag{44}$$

where $\eta=F_k^2(-\omega_0)u_0^2$. Spectra appearing in this paper are simply bar charts, in which the heights of the bars represent the weights of the δ functions. This yields the following moments for the loss spectrum, if either $\hbar\omega_0\ll E$ or $\eta\ll 1$:

$$\overline{\Delta E}=\eta\hbar\omega_0 \ , \quad \overline{(\Delta E)^2}=\eta(\eta+1)\hbar^2\omega_0^2 \ . \tag{45}$$

Since $\eta=\overline{\Delta E}/\hbar\omega_0$, under these conditions, the Debye-Waller exponent is equal to the mean number of excita-

tions of the oscillator. Note that, if $\eta \ll 1$, the distorted-wave Born approximation is recovered by expanding in powers of η, while for $\eta \gg 1$ and $\hbar\omega_0 \ll E$, the well-known Gaussian form for the loss spectrum occurs.[6]

We next discuss the specific case of an exponential repulsion, $V(z)=V_0 e^{-z/d}$, and $F(z)$ evaluated on the one-dimensional distorted eigenstates yields just the Mott-Jackson form[27]

$$F_k(\omega)=\hbar k 2\pi\omega\tau\sqrt{\sinh^{-2}[\pi(k-k')d]-\sinh^{-2}[\pi(k+k')d]}\ , \tag{46}$$

where $\tau=d/v$ is a measure of the collision time, as v is the initial velocity of the incident atom. In general, this problem can be characterized by three dimensionless parameters. As we are particularly interested in studying the behavior of this model close to the classical limit, we choose two of these parameters to be independent of $\hbar$, i.e., they characterize the classical limit of this model. In particular, we use the same parameters as JCK. We define $\alpha=1/(\omega_0\tau)$, a measure of how close to adiabatic the scattering is, i.e., $\alpha \ll 1$ means that the time for the collision to take place is much longer than the oscillator's period. We also define $\gamma=d_{\max}/d$, where $d_{\max}=p_i/(M\omega_0)$ and p_i is the initial momentum of the incident particle. As shown in JCK, for very small displacements of the oscillator during the collision, $d_{\max}$ is an upper limit on its displacement during the collision. Thus, γ is a measure of how close one is to scattering from a rigid potential, so that when $\gamma \ll 1$, the oscillator hardly moves from its equilibrium position during the collision. Finally, we choose $\kappa=kd$ as a dimensionless measure of the wave vector. In terms of these parameters, $\eta=(\kappa\gamma/2)f^2(\kappa,\alpha)$, where $f(\kappa,\alpha)=F_k(-\omega_0)/k$ is the dimensionless driving force and is given by [see Eq. (46)]

$$f(\kappa,\alpha)=\frac{2\pi}{\alpha}\left[\sinh^{-2}\left\{\pi\kappa\left[1-\left[1-\frac{2}{\kappa\alpha}\right]^{1/2}\right]\right\}-\sinh^{-2}\left\{\pi\kappa\left[1+\left[1-\frac{2}{\kappa\alpha}\right]^{1/2}\right]\right\}\right]^{1/2}. \tag{47}$$

The spectra in Fig. 1 were calculated with $\eta=0.22$ and $E=3.5\hbar\omega_0$ in (a), and $\eta=10$ and $E=100\hbar\omega_0$ in (b). The plots of $\bar{V}$ versus $\hbar\omega/E_i$ in Fig. 2 were made with $\gamma=0.1$.

To see how the TA can be recovered from EBA for this model, we consider the limit $\kappa\to\infty$, keeping α fixed. Then the driving force $f(\kappa,\alpha)$ becomes

$$f_{\mathrm{TA}}(\alpha)=\frac{2\pi}{\alpha\sinh(\pi/\alpha)}\ . \tag{48}$$

This is just the result given by the recoilless TA for this system, as the classical recoilless trajectory is

$$z_{\mathrm{cl}}(t)=d\ln\left[\frac{V_0}{E}\cosh\left[\frac{t}{2\tau}\right]\right]. \tag{49}$$

Then the Fourier transform of $m\ddot{z}_{\mathrm{cl}}(t)$ is kf_{TA}, where f_{TA} is given by Eq. (48).

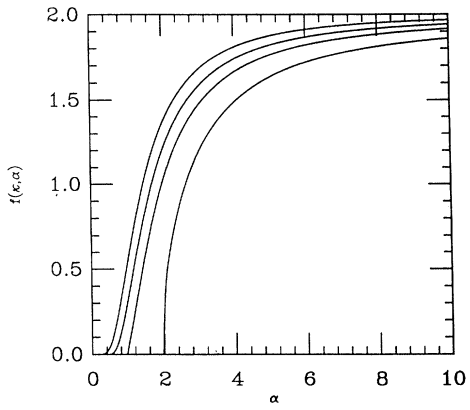

FIG. 3. Plot of $f(\kappa,\alpha)$ [given by Eq. (47) in the text] vs α for $\kappa=1, 2, 4, \infty$, from right to left. The curve $\kappa=\infty$ is just $f_{\mathrm{TA}}(\alpha)$.

It is straightforward to show that f_{TA} is a good approximation to the exact EBA result whenever $\kappa \gg 1$ and $\kappa \gg 1/\alpha$. This is demonstrated in Fig. 3, a plot of $f(\kappa,\alpha)$ versus α for various values of κ. For any given value of $\alpha(>1)$, the error is $O(1/\kappa\alpha)$. Of course, just because TA and EBA coincide for a given set of parameters does not imply that they are valid. However, *any* limit in which the driving function tends to its TA form and in which the EBA is valid, is a limit in which the TA is valid. As we will see below, such limits include special cases of semiclassical scattering, but also include any quasiadiabatic scattering.

Now we consider regimes of validity of the EBA. In general, we expect the EBA to be valid near any limit in which all the moments of the final distribution are tending to their elastic values. We consider two specific limits

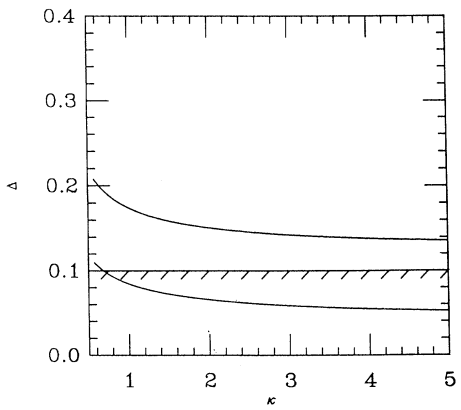

FIG. 4. Plots of Δ (estimated error in the EBA) vs κ (reduced wave vector) for fixed values of α and γ. The upper curve is $\alpha=5$, $\gamma=0.3$, and the lower is $\alpha=5$, $\gamma=0.1$. The EBA works beneath the hatched line, i.e., where $\Delta \leq 10\%$.

to illustrate this property, and compare our results to previous studies.

A. The semiclassical regime

The classical limit is defined by letting $\hbar \to 0$, keeping all classical quantities fixed. In terms of our dimensionless parameters, the classical limit is found by taking $\kappa \to \infty$, keeping α and γ fixed. We then define the semiclassical regime as one in which κ is large but finite, and consider corrections to classical results as a power series in $1/\kappa$. As noted in the Introduction, this implies that wavelengths are short, i.e., $\lambda \ll d$ ($\kappa \gg 1$), allowing the WKB approximation for the matrix elements given by Eq. (48). It also implies that the mean number of phonons excited, $\overline{\Delta E}/\hbar\omega_0 (=\eta)$, is much greater than one, so that the scattering is dominated by multiphonon events, and that the initial energy is much greater than the oscillator energy spacing, i.e., $E_i \gg \hbar\omega_0$ ($\kappa \gg 1/\alpha$). JCK found that, in the semiclassical regime, the TA generally failed, except when either α or γ was much smaller than 1. They pointed out that these regimes corresponded either to near adiabaticity or to small displacements of the oscillator relative to the potential range during the collision, respectively.

We find the same results for EBA. Figure 4 is a plot of Δ, our estimated error for the EBA, as a function of κ, keeping α and γ fixed. The higher curve is $\alpha=5$, $\gamma=0.3$, and its asymptotic (and therefore classical) value is above 10%. The lower curve is $\alpha=5$, $\gamma=0.1$, and its classical value is below 10%. Thus the EBA (and therefore the TA) fails in the semiclassical regime for the first values of the parameters, but works for the second. Figure 5 is a plot of constant error ($\Delta=0.1$) for $\kappa \to \infty$. The asymptotic values of the two curves plotted in Fig. 4 are represented by squares. Only for sufficiently small values of α or γ does the EBA work. As the EBA becomes the TA in this limit, this is to be expected, and our results are consistent with those of JCK. However, JCK found that the TA worked in these regimes only in their calculation of the mean-square width of the loss distribution, defined as $\delta^2 E = (\Delta E)^2 - \Delta E^2$. They also studied the so-called mean energy shift, defined as the leading $[O(\hbar)]$ corrections to ΔE in the semiclassical limit. They found that as $\Delta E/E \to 0$, the shift vanished for both the exact calculation and the TA, but that their ratio did not approach one. Applying their recipe for the shift [their Eq. (21)] to the recoilless classical trajectory, we also find no shift. Furthermore, if we look at the loss spectrum generated by f_{TA}, the mean energy loss is exactly the classical value, and again the shift is zero. But if we consider corrections to f_{TA}, expanding the exact $f(\kappa, \hbar\omega_0/E)$ by about $\kappa \to \infty$, we find a shift proportional to $\Delta E/E$. This indicates that it is beyond the range of validity of the EBA to correctly calculate this shift. Thus, the numerical result of JCK, that the ratio of the exact shift to the shift in the TA does not approach one as $\Delta E/E \to 0$, does not contradict our result that the TA is valid for semiclassical quasielastic scattering.

The EBA is more general than the TA, insofar as it also works beyond the semiclassical regime. In Fig. 6 we show just how the classical limit is approached. The almost straight diagonal line in Fig. 6 is a plot of η as a function of κ, for constant α (=5) and Δ (=0.1). To keep Δ constant, γ grows as κ grows, finally leveling out at 0.23, consistent with the previous figure. To the right of this curve, the EBA works, while to the left, it fails. The lower curve is a similar plot for the distorted-wave Born approximation, while the curve to the right of the figure is the same for the TA. It asymptotically approaches the EBA curve as $\kappa \to \infty$. How these errors are calculated is described in detail in the Appendix. Essentially, the error in these approximations is a sum of the error made by the EBA with the error made relative to the EBA. Note that

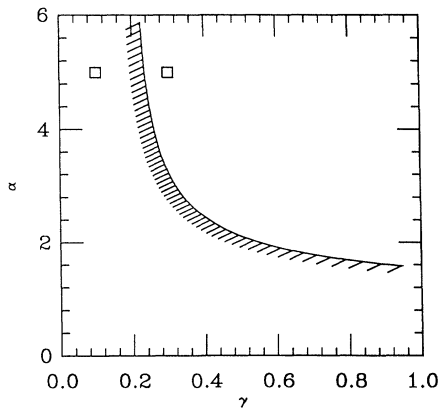

FIG. 5. Plot of constant error, $\Delta=0.1$, for the EBA in the semiclassical regime, $\kappa \to \infty$ (actually, $\kappa=31.83$). The EBA (and thus the TA) works to the left of and beneath this curve. The squares represent the $\kappa \to \infty$ limit of the two curves plotted in Fig. 4.

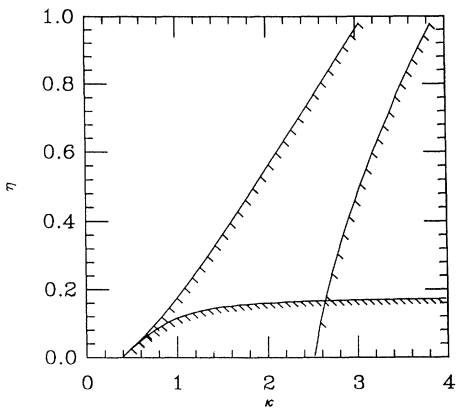

FIG. 6. Plots of constant error ($\Delta=0.1$) for fixed α (=5), for various approximations, in the (η,κ) plane. η is the Debye-Waller exponent, i.e., $\exp(-\eta)$ is the probability of elastic scattering, and κ is the reduced wave vector. Each approximation works to the right of (and below) its hatched constant error line. The curve entirely in the region $\eta<0.2$ is for the distorted-wave Born approximation, the curve with $\kappa>2$ is for the trajectory approximation, and the third curve is for the EBA. The TA curve approaches the EBA curve asymptotically as $\kappa \to \infty$. How these errors were calculated is described in the Appendix.

along the curves the external parameter γ is made to vary (in different ways on each curve) so as to keep the error made in each approximation fixed along its curve. Consider now the distorted-wave Born approximation curve. For these conditions, there is a maximum value of η, above which the DWBA fails. In fact, one cannot meaningfully have $\eta > 1$ in the distorted-wave Born approximation, because that would imply a greater than unity probability of exciting the oscillator into its first excited state. The definition of error we use for the distorted-wave Born approximation, which s given in the Appendix, is designed to preclude this possibility, which is why the distorted-wave Born approximation curve in this figure never grows beyond $\eta=0.2$ as $\kappa\rightarrow\infty$. Thus, the distorted-wave Born approximation is limited to weakly inelastic scattering. In fact, as $\kappa\rightarrow\infty$, the loss spectrum becomes a Gaussian with negligible probability for the oscillator to end in either the ground state or first excited state, and so cannot be well approximated by the distorted-wave Born approximation. The EBA can, of course, treat situations where $\eta \gg 1$, as is the case here for large κ. Similarly, the TA only works for sufficiently short wavelengths, i.e., there is a minimum κ below which it fails. On the other hand, the EBA includes all the parameter space covered by the other two approximations, and more. There is a trade off between scattering strength and wave vector. In this figure, the area enclosed by the three curves is a region in which only the EBA works. Thus, the EBA is valid for ranges of parameters in which neither of the other two approximations apply.

Figure 7 is a plot of the fractional energy loss as a function of κ, for $\alpha=5$ and $\Delta=0.1$, i.e., the same conditions as the previous figure. It illustrates the quasielastic nature of the EBA. Note that only for κ larger than about 3 does the criterion for the validity of the EBA become just that the fractional energy transfer is small. For small κ, clearly a more stringent requirement takes over. For fixed α and γ, as $\kappa\rightarrow 0.4$, the value which corresponds to the incident particle having just enough to excite the oscillator to its first state, the strength of the scattering tends to zero. So, the EBA becomes equivalent to the DWBA here. On the other hand, the virtual excitations contributing to $G^{(2)}$ are not cut off at this value of κ, so that Δ would become very large as its denominator vanishes. Thus, the line of constant Δ drops to zero at $\kappa=0.4$. This is simply saying that in this vicinity the first-order (distorted-wave) Born approximation is no good, as the second-order terms dominate. Finally, we mention that the DWBA cannot yield an accurate picture of the entire spectrum for the largest values of κ shown here for the reasons given in the discussion of Fig. 6. However, as noted by Brenig,[7] the first moment of that distribution, i.e., the mean energy loss, *is* the same in the DWBA and the EBA, and is therefore correctly given by the DWBA in the parameter regime below the hatched line of this figure.

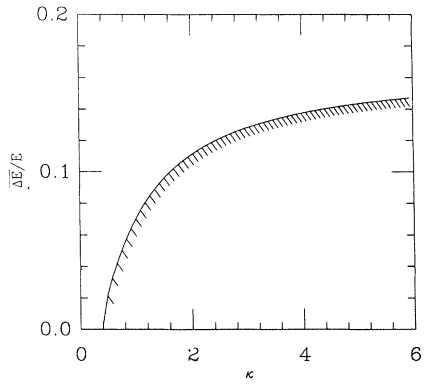

FIG. 7. Same as Fig. 6, except plotted in the (fractional energy loss, κ) plane, and only the EBA curve is plotted here.

B. The quasiadiabatic regime

This is the regime studied by Burke and Kohn, in which $m\rightarrow\infty$, with E, d, M, and ω_0 fixed. In the limit, the scattering is completely adiabatic, and therefore totally elastic. It is corrections to this limit which are called quasiadiabatic. In dimensionless parameters, we again let $\kappa\rightarrow\infty$, but now keeping $\kappa\alpha$ and γ/κ fixed. This is equivalent to keeping $\hbar\omega_0/E_i$ and u_0/d fixed. As this limit is approached, $\Delta\bar{E}\rightarrow 0$, so we expect the EBA to work for any values of the fixed parameters for sufficiently large κ. This is illustrated in Fig. 8, which consists of plots of Δ versus κ for several values of the fixed parameters. In all cases, the curves tend to zero error exponentially as $\kappa\rightarrow\infty$. Thus, the EBA is valid for quasiadiabatic scattering in general.

We note here that this simple model displays a qualitatively different behavior to that of real surfaces, because real surfaces contain acoustic modes with frequencies ranging continuously down to zero. Summing over those modes would change the exponential decay of Fig. 8 into a power-law behavior. However, as long as the number of phonons excited during the collision remains finite (as

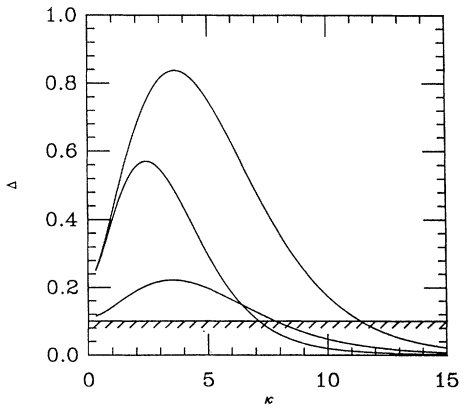

FIG. 8. Plots of Δ (EBA error) vs κ (reduced wave vector) as the adiabatic limit is approached. Both the ratio of oscillator spacing to incident energy and the ratio of zero-point displacement to interaction range are held fixed, at values of 0.2 and 0.2 for the highest-peaked curve, 0.3 and 0.2 for the next-highest, and finally at 0.2 and 0.1 for the lowest.

was shown to be the case for realistic systems by Burke and Kohn) the EBA will be valid in the quasiadiabatic regime.

VI. CONCLUSIONS

We have rigorously shown that the EBA works for particle scattering from a boson bath when the scattering is nearly elastic in the sense defined in the Introduction, in agreement with Brenig's claim.[7] We have seen how it reduces to the recoilless trajectory approximation when the incident particle's wavelength is sufficiently short, and to the distorted-wave Born approximation when the scattering is sufficiently weak. We have investigated the semiclassical regime, and found results consistent with JCK.[5] We have also studied the quasiadiabatic regime, and verified the conjecture of Burke and Kohn.[4] We have developed an explicit formula for estimating the error involved in the EBA and have used it to show that the EBA is valid in regimes not covered by either the TA or the DWBA. We have used a simple one-dimensional model to illustrate how our error estimate works. Because of the simplicity of this model, it cannot be taken as representative of realistic systems, but our method of estimating the error can and will be applied to the full atom-surface scattering problem. However, many questions concerning the TA remain unanswered.

Perhaps the most important of these concerns the correct treatment of problems in which no single state dominates, i.e., in which there is no unique elastic state in the decoupled problem. A solution to this problem would greatly extend the usefulness of the EBA to such areas as sticking, diffraction, etc. Another important issue is the question of when the TA with recoil is a useful approximation. JCK found, for their simple model, the TA was valid only for small energy transfer. However, the present proof suggests that all that is really required for the validity of the full TA is that $\hbar\bar{\omega} \ll E$, where $\hbar\bar{\omega}$ is the mean excitation energy per phonon, and E is the energy of the incident particle at any time during the collision. More complex systems may allow this condition to be satisfied, and employ the full TA, without reducing to the EBA. Another problem is to generalize the discussion to interactions which are not linear in the surface atom displacements. In particular, it seems likely the TA is valid for quasiadiabatic scattering in which the surface atoms undergo large distortions, even if the interaction is nonlinear about the equilibrium positions, by linearizing the potential about the adiabatically varying equilibrium positions. Nourtier[15] addresses many of these questions, but only within the approximation of short wavelength of the incident particle. The present study suggests these questions might also be tractable for arbitrary wavelengths, especially using the formalism developed here.

To conclude, we note the EBA is valid for a large variety of atom-surface scattering experiments, and may be used to calculate multiphonon scattering with relatively little further work than the distorted-wave Born approximation. It should provide a useful tool for understanding atom-surface scattering, and a more detailed study of the error it makes under realistic scattering conditions will follow this work.

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APPENDIX: ERROR ESTIMATES FOR THE SIMPLE MODEL

The error estimate for the EBA given by Eq. (40) is designed as a qualitative indicator of when the EBA works. It is fairly crude, and as such, we make several further approximations (for calculational convenience only) in applying it to the simple model of Sec. V. In that model, we may write, for $G^{(1)}$ and $G^{(2)}$,

$$G^{(1)} = Aa + A^{\dagger}a^{\dagger}, \quad G^{(2)} = Baa + B^{\dagger}a^{\dagger}a^{\dagger}, \tag{A1}$$

where A, given by Eq. (29) applied to this model, is just

$$A = \sum_k \bar{V}(\epsilon_k;\omega_0)\rho(\epsilon_k;\omega_0) \tag{A2}$$

while B comes from performing the time integrals in Eq. (32), and inserting the result in Eq. (30), to yield

$$B = \frac{i}{2}\sum_k \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{\bar{C}(\epsilon_k;\omega,2\omega_0-\omega)}{\omega_0-\omega}\rho(\epsilon_k;2\omega_0), \tag{A3}$$

where

$$\bar{C}(\epsilon_k;\omega,2\omega_0-\omega) = \bar{V}(\epsilon_k+2\omega_0-\omega;\omega)\bar{V}(\epsilon_k;2\omega_0-\omega) - \bar{V}(\epsilon_k+\omega;2\omega_0-\omega)\bar{V}(\epsilon_k;\omega) \tag{A4}$$

is the analog of Eq. (31) for this model. Note that in Eq. (A1) we have already excluded the relaxation terms in $G^{(2)}$, which are of the form $a^{\dagger}a$ in the oscillator operators. Then,

$$\langle \phi[G^{(1)}]^2|\phi\rangle \approx (2\eta+1)\langle k_i|AA^{\dagger}|k_i\rangle, \tag{A5}$$

and

$$\langle \phi|[G^{(2)}]^2|\phi\rangle \approx 2(\eta+\tfrac{1}{2})^2\langle k_i|BB^{\dagger}|k_i\rangle. \tag{A6}$$

But $\langle k_i|AA^{\dagger}|k_i\rangle$ is just η, so that the error made by the EBA is approximated as

$$\Delta_{\mathrm{EBA}} \approx \left[\frac{\eta+\frac{1}{2}}{\eta}\right]^{1/2} g(\kappa,\hbar\omega_0/E_i), \tag{A7}$$

where

$$g(\kappa,\hbar\omega_0/E_i) = P\int_{-\infty}^{E_i+\omega_0} \frac{d\omega}{2\pi\omega}\bar{V}(E_i;\omega_0-\omega) \times \bar{V}(E_i+\omega_0-\omega;\omega_0+\omega), \tag{A8}$$

and P indicates a principal value integral. Note that the upper limit in the integration is due to the vanishing of

the first matrix element for energy losses greater than the initial energy. This last expression for $g(\kappa,\hbar\omega_0/E_i)$ comes from shifting the origin of the frequency integral in Eq. (A3) by ω_0, and then using detailed balance on the matrix elements (which are real, because the scattering states are)

$$\bar{V}(E;\omega)=\bar{V}(E+\omega;-\omega) \tag{A9}$$

in the second half of the commutator in Eq. (A4).

To estimate the errors in the distorted-wave Born approximation and the TA, we assume the total error is the root square sum of the error made by the EBA and the error made relative to the EBA i.e.,

$$\Delta_{\rm tot}=\sqrt{\Delta^2_{\rm EBA}+\Delta^2_{\rm rel}}\ . \tag{A10}$$

To estimate the relative error, we simply use the error made in the value of the Debye-Waller exponent, as this is a measure of the strength of the scattering, and completely characterizes the final distribution in this model in the EBA. We write

$$\Delta^2_{\rm rel}=\left[\frac{\eta_{\rm EBA}-\eta_{\rm approx}}{\eta_{\rm EBA}}\right]^2 . \tag{A11}$$

Note that, for the DWBA, the Debye-Waller exponent may be deduced from flux conservation to be

$$\eta_{\rm DWBA}=-\ln(1-\eta)\ , \tag{A12}$$

as η, which is given after Eq. (45) is the text and represents the total probability of inelastic scattering in this approximation. These formulas were used to estimate the errors in the figures.

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