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A Unified Model for Diffractive and Inelastic Scattering of a Light Atom from a Solid Surface

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Abstract

A simple model for gas-surface scattering is presented which permits treatment of inelastic effects in diffractive systems. This model, founded on an impulsive collision assumption, leads to an intensity distribution which is just a sum of contributions from n-phonon scattering events. Furthermore, by using a convenient form for the repulsive interaction potential, analytic expressions are obtained for the elastic and one-phonon intensities that are in qualitative agreement with experimental results.

I. Introduction.

Ever since the early days of quantum mechanics, there has been a continuing interest in trying to understand and describe the nature of the scattering of gaseous atoms and molecules from various types of solid surfaces. With the discovery of diffraction peaks in the scattering of a helium beam from a lithium fluoride crystal plane, theorists were able to construct the first crude models for the gas-surface interaction which gives rise to the observed diffractive phenomena.[1] Not until more recently, however, has much progress been made in the development of a global theory which would allow one actually to identify all or most of the structure yielded by experiments. Notably, Goodman[2] and later Goodman and Tan,[3] using a continuum model of the solid and obtaining transition probabilities via the method of Cabrera, Celli, Goodman, and Manson[4] (CCGM), were able to calculate a scattering distribution for the He-LiF(001) system which at least qualitatively reproduces the experimental inelastic results. Other work by Lin and Wolken,[5] who performed a close-coupling calculation, and by Metiu[6] has also helped to clarify the physics of the gas-surface collision, although both the approaches taken by these investigators require extensive numerical computation before the scattering structure can be revealed. On the other hand, the state of the theory has also benefited from the consideration of simplified scattering models which permit one to identify the particular constituent effects that give rise to the composite intensity pattern. A good example of just such an approach is to be found in the work of Weare,[7] who has examined the specific case in which the surface and gas temperatures are sufficiently low that a

first-order perturbative treatment of the inelasticity adequately describes the scattering from a smooth potential surface.

In the present work we provide a simple alternative one-dimensional surface model which manifestly displays the principal features of both the elastic and the inelastic processes. Although the basic formalism has been previously described elsewhere,[8] the model which we have adopted in fact permits an analytic determination of the scattering intensities, thereby adding substantially to the understanding of the consequences of such a formulation.

There appear to be two major stumbling blocks in evidence in the bulk of the previous inelastic studies. The first is the assumption that in the absence of phonon excitations, the crystal surface is flat.[2,7,9] Such an assumption is clearly inadequate if a unified model is to be constructed due to the fact that for a flat surface specular scattering is the only elastic process allowed. Consequently, one should select a form for the gas-surface interaction potential (the surface contour being classically just the turning-point surface for this potential at the given collision energy) which yields a version of the corrugated hardwall potential in the limit of zero phonon displacement inasmuch as such a corrugated contour is known to produce the desired diffraction peak structure.[10]

Secondly, there is always a problem involved in treating the phonon mode enumeration and averaging. Elaborate treatments, such as that by Beeby,[9] have all of the proper phonon dynamics incorporated in them; however, the difficulty of such inclusion makes those formalisms somewhat cumbersome to use while apparently adding little to the construction of a straightforward physical picture of the scattering. Furthermore it is desirable to avoid <u>ad hoc</u> averaging procedures[11] whose accuracy is hard

to evaluate. In the model described below, we have handled these modes in a very intuitive way which does indeed seem to generate the gross phonon structure but at the same time does not obscure the fundamental physics.

This work takes advantage of the widely used assumption that the scattering pattern arises as a result of a more or less purely repulsive two-body short range component of the gas-surface interaction. Such an assumption logically leads to an impulsive collision model, which itself has a basis in experimental findings,[12] so that the surface motion may be effectively decoupled from the actual collision dynamics within the interaction time interval. Equivalently, this particular model has been obtained by Weare[7] in the limit that for a given initial energy state of the solid, the translational energy of the incident gas atom is allowed to become large. By making this approximation, we do, however, necessarily restrict our formalism to the collision of light atoms such as helium with the surface, although in practice these are the very systems which are amenable to experimental study. A more detailed discussion of the impulsive collision assumption is given in Section II.

II. Impulsive Collision Approximation.

The basic problem in which we are interested is the calculation of a transition probability (i.e., a scattering intensity) from some initial wavevector $\vec{k}_i$, which describes the unperturbed motion of the incident gas atom, to a final wavevector for the scattered atom $\vec{k}_f$, with a concurrent energy loss (or gain) due to inelasticity, ΔE. Practically, since at present one cannot experimentally characterize precisely the quantum states of the solid before or after the collision, it is necessary to average appropriately over the phonon modes if one is to obtain a quantity which can actually be observed. Thus, the scattering intensity may be written in terms of an S-matrix element as

$$I_{\Delta E,\vec{k}_f\leftarrow\vec{k}_i} = \sum_{\underset{\sim}{n}_2} \sum_{\underset{\sim}{n}_1} \frac{e^{-\beta\epsilon_{\underset{\sim}{n}_1}}}{Q} \delta[\Delta E + (\epsilon_{\underset{\sim}{n}_2} - \epsilon_{\underset{\sim}{n}_1})] \left| S_{\vec{k}_f,\underset{\sim}{n}_2\leftarrow\vec{k}_i,\underset{\sim}{n}_1} \right|^2 \qquad (2.1)$$

where $\underset{\sim}{n}_1$ and $\underset{\sim}{n}_2$ label respectively the initial and final phonon states having energies $\epsilon_{\underset{\sim}{n}_1}$ and $\epsilon_{\underset{\sim}{n}_2}$. [In addition, Q is the phonon partition function, and $\beta = (k_B T_S)^{-1}$ (with k_B being Boltzmann's constant and T_S the characteristic surface temperature).]

As indicated in the Introduction, one then commonly proceeds by taking the short-range gas-surface interaction to be repulsive, the limit of which being a simple hard wall. Certainly if diffractive elastic scattering dominates the intensity pattern, then it is reasonable to assume that the collision may be modeled in zeroth order by a hard sphere rebounding elastically from an infinitely hard surface. More realistically, the surface is described as being a corrugated wall which undergoes distortions due to the excitation of phonon modes in the solid, these distortions

presumed to be a small percentage of a lattice dimension, and that this motion is slow compared to the collision time (which, of course, in the case of a perfectly hard wall is infinitesimal).

The above impulsive collision assumption may be introduced into Eq. (2.1) by writing the S-matrix element in the sudden approximation form,[13]

$$S_{\vec{k}_{f\sim 2} n_2 \leftarrow \vec{k}_{i\sim 1} n_1} = \langle \vec{k}_{f\sim 2} n_2 | e^{2i\eta(x;\underset{\sim}{q})} | \vec{k}_{i\sim 1} n_1 \rangle \quad , \tag{2.2}$$

where in this particular case the phase shift η depends not only upon the coordinate parallel to the surface plane, x, but also parametrically upon the vector of phonon normal mode displacement coordinates, $\underset{\sim}{q} = \{q_j\}$. Since we have assumed the gas-surface repulsive interaction to be well described by a hard wall potential, it then follows that the phase shift is given by the hard sphere scattering result,[14] i.e.,

$$\eta(x;\underset{\sim}{q}) = -kZ(x;\underset{\sim}{q}) \quad ,$$

where $Z(x;\underset{\sim}{q})$ is the equation of the surface contour.

But now how does one actually determine a form for this surface contour? Presumably, if the distortions which arise as a result of the excitation of the phonon modes are, as was previously suggested, sufficiently small in amplitude, then the contour should be adequately described by a truncated Taylor series, the expansion being made about the equilibrium surface position,

$$Z(x;\underset{\sim}{q}) = Z(x;0) + \frac{\partial Z(x;\underset{\sim}{q})}{\partial \underset{\sim}{q}} \Big|_{\underset{\sim}{q}=0} \cdot \underset{\sim}{q} \tag{2.3}$$

($\underset{\sim}{q} = 0$ corresponding to the undistorted surface). In order to simplify the notation somewhat, we rewrite Eq. (2.3) in the following form:

$$Z(x;\underset{\sim}{q}) = Z(x) + \underset{\sim}{\zeta}(x)\cdot\underset{\sim}{q} \quad , \tag{2.4}$$

with the vector $\underset{\sim}{\zeta}(x)$ having components given by

$$\zeta_j(x) = \frac{\partial Z(x;q)}{\partial q_j}\Big|_{\underset{\sim}{q}=0} \quad .$$

Within this expansion the product $\zeta_j(x)q_j$ may be interpreted then as being the displacement of the surface contour at x as a result of the excitation of the j^{th} normal mode, the total displacement at x being obtained by summing over all of the N modes of the surface atoms. Unfortunately it will in general not be possible to determine $\underset{\sim}{\zeta}(x)$ analytically; however, for the case which we shall examine in Section IV, these vectors may be constructed, and hence the surface contour (and thus the phase shift) may be obtained.

III. Formal Considerations.

Using the results of the previous section (Eqs. (2.1), (2.2), and (2.4)), the scattering intensity is given in the impulsive approximation by

$$I_{\Delta E,\vec{k}_f\leftarrow\vec{k}_i} = \sum_{\underset{\sim}{n}_2}\sum_{\underset{\sim}{n}_1} \frac{e^{-\beta\varepsilon_{\underset{\sim}{n}_1}}}{Q}\,\delta[\Delta E + (\varepsilon_{\underset{\sim}{n}_2}-\varepsilon_{\underset{\sim}{n}_1})]\left|\int dx\int d\underset{\sim}{q}\; e^{-i\Delta k_x x}\right.$$

$$\left. \cdot\,\phi_{\underset{\sim}{n}_2}(\underset{\sim}{q})^*\,\phi_{\underset{\sim}{n}_1}(\underset{\sim}{q})\, e^{-i\Delta k_z Z(x)}\, e^{-i\Delta k_z\,\underset{\sim}{\zeta}(x)\cdot\underset{\sim}{q}}\right|^2 , \qquad (3.1)$$

where $\phi_{\underset{\sim}{n}_1}$ and $\phi_{\underset{\sim}{n}_2}$ are, respectively, the initial and final quantum states of the solid, and Δk_x and Δk_z are the projections of $\Delta k = |\vec{k}_f - \vec{k}_i|$ parallel and perpendicular to the plane of the surface. One then notes that since the coordinate representation of the transition operator is given here by

$$T(\underset{\sim}{q}) = \int dx\; e^{-i\Delta k_x x}\, e^{-i\Delta k_z Z(x)}\, e^{-i\Delta k_z\underset{\sim}{\zeta}(x)\cdot\underset{\sim}{q}} , \qquad (3.2)$$

Eq. (3.1) may be rewritten as

$$I_{\Delta E,\vec{k}_f\leftarrow\vec{k}_i} = \sum_{\underset{\sim}{n}_2}\sum_{\underset{\sim}{n}_1} \frac{e^{-\beta\varepsilon_{\underset{\sim}{n}_1}}}{Q}\,\delta[\Delta E + (\varepsilon_{\underset{\sim}{n}_2}-\varepsilon_{\underset{\sim}{n}_1})]\langle \underset{\sim}{n}_1|T^\dagger|\underset{\sim}{n}_2\rangle$$

$$\cdot\langle \underset{\sim}{n}_2|T|\underset{\sim}{n}_1\rangle \quad .$$

Furthermore, by employing the Fourier transform identity for the delta function, this last equation may be cast into a particularly convenient form, namely the well-known correlation function expression,[8] via the following transformations:

$$I_{\Delta E,\vec{k}_f\leftarrow\vec{k}_i} = (2\pi\hbar)^{-1}\int dt\ e^{-i\Delta Et/\hbar}\sum_{\underset{\sim}{n}_2}\sum_{\underset{\sim}{n}_1}\frac{e^{-\beta\varepsilon_{\underset{\sim}{n}_1}}}{Q}\ \langle \underset{\sim}{n}_1|T^\dagger(0)|\underset{\sim}{n}_2\rangle$$

$$\cdot\langle \underset{\sim}{n}_2|T(t)|\underset{\sim}{n}_1\rangle$$

$$= (2\pi\hbar)^{-1}\int dt\ \frac{e^{-i\Delta Et/\hbar}}{Q}\ \mathrm{Tr}[e^{-\beta H_0}\ T^\dagger(0)\ T(t)]$$

$$\equiv (2\pi\hbar)^{-1}\int dt\ e^{-i\Delta Et/\hbar}\ \langle T^\dagger(0)\ T(t)\rangle \quad ,$$

where T(t) is the time-evolved transition operator T in the Heisenberg representation,

$$T(t) = e^{-iH_0t/\hbar}\ T\ e^{iH_0t/\hbar} \quad ,$$

and H_0 is the phonon Hamiltonian.

The time correlation function thus defined may be evaluated by re-expanding in terms of the T-matrix elements,

$$\langle T^\dagger(0)\ T(t)\rangle = \sum_{\underset{\sim}{n}_2}\sum_{\underset{\sim}{n}_1}\frac{e^{-\beta\varepsilon_{\underset{\sim}{n}_1}}}{Q}\ e^{-i(\varepsilon_{\underset{\sim}{n}_2}-\varepsilon_{\underset{\sim}{n}_1})t/\hbar}\ \langle \underset{\sim}{n}_1|T^\dagger|\underset{\sim}{n}_2\rangle$$

$$\cdot\langle \underset{\sim}{n}_2|T|\underset{\sim}{n}_1\rangle$$

$$= Q^{-1}\int dx\int dx'\int d\underset{\sim}{q}\int d\underset{\sim}{q}'\ e^{-i\Delta k_x(x-x')}\ e^{-i\Delta k_z(Z(x)-Z(x'))}$$

$$\cdot e^{-i\Delta k_z\underset{\sim}{\zeta}(x)\cdot\underset{\sim}{q}}\ e^{i\Delta k_z\underset{\sim}{\zeta}(x')\cdot\underset{\sim}{q}'}\ [\sum_{\underset{\sim}{n}_2}\langle \underset{\sim}{q}'|\underset{\sim}{n}_2\rangle\ e^{-i\varepsilon_{\underset{\sim}{n}_2}t/\hbar}\ \langle \underset{\sim}{n}_2|\underset{\sim}{q}\rangle]$$

$$\cdot[\sum_{\underset{\sim}{n}_1}\langle \underset{\sim}{q}|\underset{\sim}{n}_1\rangle\ e^{-\beta\varepsilon_{\underset{\sim}{n}_1}}\ e^{i\varepsilon_{\underset{\sim}{n}_1}t/\hbar}\ \langle \underset{\sim}{n}_1|\underset{\sim}{q}'\rangle] \quad . \qquad (3.3)$$

Assuming that the phonon modes are harmonic, the terms in square brackets in Eq. (3.3) are just harmonic oscillator propagators in the coordinate representation.[15] Thus the above equation may be written

$$\langle T^{\dagger}(0)\ T(t)\rangle = \int dx \int dx'\ e^{-i\Delta k_x (x-x')}\ e^{-i\Delta k_z (Z(x)-Z(x'))}\ C(x,x';t)\ , \tag{3.4}$$

the new instantaneous position correlation function $C(x,x';t)$ being given by

$$C(x,x';t) = Q^{-1} \prod_j \int dq_j \int dq'_j\ e^{-i\Delta k_z (\zeta_j(x) q_j - \zeta_j(x') q'_j)}$$

$$\cdot \left[\frac{4\pi^2\hbar^2}{m^2\omega_j^2} \sin\omega_j t\, \sin\omega_j (t + i\hbar\beta)\right]^{-1/2} \exp\left\{\frac{im\omega_j}{2\hbar \sin\omega_j t}\left[(q_j^2 + q_j'^2)\cos\omega_j t\right.\right.$$

$$\left.\left. - 2q_j q'_j\right] - \frac{im\omega_j}{2\hbar \sin\omega_j (t+i\hbar\beta)}\left[(q_j^2 + q_j'^2)\cos\omega_j (t+i\hbar\beta) - 2\, q_j q'_j\right]\right\}\ ,$$

(here m and ω_j are the mass and frequency of the j^{th} mode). The integrals over phonon mode displacement coordinates are of the general gaussian form and therefore may be done analytically, the details of which will not be given here. Suffice to say, after a substantial amount of algebra one obtains

$$C(x,x';t) = \exp\left\{-\sum_j \frac{\hbar\Delta k_z^2}{4m\omega_j}\left[(\zeta_j(x)^2 + \zeta_j(x')^2 - 2\zeta_j(x)\zeta_j(x')\cos\omega_j t)\coth\frac{\hbar\omega_j\beta}{2}\right.\right.$$

$$\left.\left. + 2i\zeta_j(x)\zeta_j(x')\sin\omega_j t\right]\right\}\ .$$

Then, recognizing that the mean square displacement of a harmonic oscillator[16] is just

$$\langle q_j^2 \rangle = \left(\frac{2m\omega_j}{\hbar} \tanh \frac{\hbar\omega_j\beta}{2}\right)^{-1} ,$$

the correlation function may be rewritten as

$$C(x,x';t) = \exp\{-\frac{1}{2}\sum_j \Delta k_z^2 \langle q_j^2 \rangle (\zeta_j(x)^2 + \zeta_j(x')^2)\}$$

$$\cdot \exp\{\sum_j \Delta k_z^2 \langle q_j^2 \rangle \zeta_j(x)\zeta_j(x')[\cos\omega_j t - i \sin\omega_j t \tanh \frac{\hbar\omega_j\beta}{2}]\} . \tag{3.5}$$

At this point one may identify the Debye-Waller factor, defined by

$$W(x) = \exp\{-\frac{1}{2}\Delta k_z^2 \sum_j \langle q_j^2 \rangle \zeta_j(x)^2\} .$$

Furthermore, if we pass to the classical limit then $\tanh \frac{\hbar\omega_j\beta}{2} \to \frac{\hbar\omega_j\beta}{2}$, with the result being that Eq. (3.5) takes the form

$$C^{CL}(x,x';t) = e^{-W(x)}\, e^{-W(x')} \exp\{\sum_j \Delta k_z^2 \langle q_j^2 \rangle \zeta_j(x)\zeta_j(x')$$

$$\cdot[\cos\omega_j t - i \frac{\hbar\omega_j\beta}{2} \sin\omega_j t]\}$$

$$= e^{-W(x)}\, e^{-W(x')} \exp\{(1 + i \frac{\hbar\beta}{2} \frac{d}{dt}) \sum_j \Delta k_z^2 \langle q_j^2 \rangle$$

$$\cdot \zeta_j(x)\zeta_j(x')\cos\omega_j t\}$$

$$= e^{-W(x)}\, e^{-W(x')} \exp\{(1 + i \frac{\hbar\beta}{2} \frac{d}{dt})\Delta W(x,x';t)\} . \tag{3.6}$$

(Note that the completely classical result ($\hbar = 0$) is obtained by totally neglecting the imaginary part of ΔW.)

While Eq. (3.6) is the exact result for the correlation function within the impulsive collision assumption, in its present form one is not able to identify easily the elementary physical processes which give rise to the observed effect. In order to reveal these processes, the last exponential is expanded in a power series, which when substituted with Eq. (3.4) into (3.1) leads to the following equation for the scattered intensity:

$$I_{\Delta E,\vec{k}_f\leftarrow\vec{k}_i} = (2\pi\hbar)^{-1}\int dt\, e^{-i\Delta Et/\hbar}\int dx\int dx'\, e^{-i\Delta k_x(x-x')}\, e^{-i\Delta k_z(Z(x)-Z(x'))}$$

$$\cdot e^{-W(x)}\, e^{-W(x')}\,\{1 + (1 + i\frac{\hbar\beta}{2}\frac{d}{dt})\Delta W$$

$$+ \frac{1}{2}\,[(1 + i\frac{\hbar\beta}{2}\frac{d}{dt})\Delta W]^2 + \ldots\,\}\quad .$$

Since the time enters into ΔW through a cosine term only, the time integral simply yields energy delta functions,

$$I_{\Delta E,\vec{k}_f\leftarrow\vec{k}_i} = \delta(\Delta E)\left|\int dx\, e^{-i\Delta k_x x}\, e^{-i\Delta k_z Z(x)}\, e^{-W(x)}\right|^2$$

$$+ \frac{1}{2}\sum_j\{[\delta(\Delta E-\hbar\omega_j) + \delta(\Delta E+\hbar\omega_j)] - \frac{\hbar\omega_j\beta}{4}[\delta(\Delta E-\hbar\omega_j)$$

$$- \delta(\Delta E+\hbar\omega_j)]\Delta k_z^2\langle q_j^2\rangle\left|\int dx\, e^{-i\Delta k_x x}\, e^{-i\Delta k_z Z(x)}\right.$$

$$\left.\cdot\, e^{-W(x)}\,\zeta_j(x)\right|^2\} + \ldots$$

$$= I_0 + I_1 + \ldots$$

(terms through first order in ΔW being shown explicitly). In this form the component elementary scattering processes may be easily recognized: the first term produces just the purely elastic spectrum with peak intensities attenuated by Debye-Waller factors, the second term represents the one-phonon inelastic event, and the remaining terms account for higher order phonon processes. Note that in the one-phonon term the phonon annihilation ($\Delta E = + \hbar\omega_j$) and creation ($\Delta E = - \hbar\omega_j$) contributions are symmetric in the completely classical result, and that even though when quantum effects begin to become significant such that the intensity symmetry is broken, the positions of the inelastic peaks are unaltered.

Due to the periodicity of the surface, it is advantageous to transform the x-integrals into sums of integrals over a unit cell. Since the details of this transformation are given in Appendix A, we will only indicate the final results here. For the component intensities one finds that

$$I_0 = Na^2\,\delta(\Delta E)\sum_{\ell}\delta(\ell - \frac{\Delta k_x a}{2\pi})\left|\frac{1}{a}\int_0^a dx\; e^{-i\Delta k_x x}\, e^{-i\Delta k_z Z(x)}\, e^{-W(x)}\right|^2$$

$$= Na^2\delta(\Delta E)\sum_{\ell}\delta(\ell - \frac{\Delta k_x a}{2\pi})\left|S_\ell^0\right|^2 \quad , \tag{3.7a}$$

and

$$I_1 = \frac{1}{2}\sum_j\{[\delta(\Delta E-\hbar\omega_j) + \delta(\Delta E+\hbar\omega_j)] - \frac{\hbar\omega_j\beta}{4}[\delta(\Delta E-\hbar\omega_j)$$

$$- \delta(\Delta E+\hbar\omega_j)\}\Delta k_z^2\langle q_j^2\rangle\left|\int_0^a dx\; e^{-i\Delta k_x x}\, e^{-i\Delta k_z Z(x)}\right.$$

$$\left.\cdot\, e^{-W(x)}\sum_n e^{-i\Delta k_x na}\,\zeta_j(x+na)\right|^2 \quad , \tag{3.7b}$$

where N is the number of unit cells within the experimental interaction zone.

Eqs. (3.7) represent the general result for the scattering intensity due to elastic and one-phonon inelastic processes. (We are ignoring for the time being any higher order processes which give rise to any effective elastic or single-phonon transitions, e.g., a two-phonon event in which the same phonon is first annihilated and then created or vice versa.) Although higher-order phonon terms may be similarly constructed, it will be convenient to restrict ourselves to consideration of only these two contributions, although there seems to be no general consensus as to the appropriateness of the one-phonon approximation.[7,9]

IV. Interaction Potential.

The formal solution given in the previous section, while perhaps somewhat illustrative, does not really provide a physical picture of the collision due to the presence of the as yet unknown ζ-vectors. In order to obtain analytical formulae which clearly reveal the scattering structure, it is necessary to adopt a model potential which at least qualitatively represents the true gas-surface interaction while at the same time permits a tractable solution to the problem. Remembering that the impulse approximation supposes that the interatomic forces are fundamentally repulsive, we take as our model potential the simple two-body form

$$V = V_0 \sum_j e^{-\alpha(x-x_j)^2} e^{-\gamma(z-z_j)} , \qquad (4.1)$$

where V_0 is some scaling parameter which also sets the units properly, the adjustable parameters α and γ are measures of the range of the potential, and (x_j, z_j) is the instantaneous position of the j^{th} surface atom.

Furthermore, let us assume that the solid atoms have only two unique vibrational frequencies, ω_x and ω_z, respectively corresponding to oscillation parallel or perpendicular to the surface plane. Such an assumption implies that we are considering some sort of modified Einstein solid, a consideration which shall be discussed in more detail in Section V. The phonon mode displacement coordinates, q_j^x and q_j^z, are therefore just the displacements of the j^{th} atom from its equilibrium position. Thus, if the coordinate system is fixed with the origin at the equilibrium position of one of the surface atoms

$$x_j = ja + q_j^x$$

$$z_j = q_j^z \quad . \tag{4.2}$$

Substituting Eq. (4.2) into (4.1) and defining the surface contour to be the classical turning-point surface for the interatomic potential, i.e., $V(Z(x,\underset{\sim}{q})) = E$, one may solve for the contour analytically,

$$Z(x,\underset{\sim}{q}) = \gamma^{-1} \ln \frac{V_0}{E} + \gamma^{-1} \ln \sum_j e^{-\alpha(x-ja-q_j^x)^2} e^{\gamma q_j^z} \quad .$$

As mentioned in the Introduction, in the static surface limit ($\underset{\sim}{q} = 0$), this contour should resemble a corrugated hard wall. Thus,

$$Z(x,0) \equiv Z(x) = \gamma^{-1} \ln \frac{V_0}{E} + \gamma^{-1} \ln \sum_j e^{-\alpha(x-ja)^2} \quad . \tag{4.3}$$

Using the Poisson sum rule,[17] the summation may be written

$$\sum_\ell \int dj \; e^{2\pi i \ell j} \; e^{-\alpha(x-ja)^2} = \frac{\sqrt{\pi}}{a\sqrt{\alpha}} \sum_\ell e^{-\pi^2 \ell^2/\alpha a^2} \; e^{\frac{2\pi i \ell}{a} x}$$

$$\cong \frac{\sqrt{\pi}}{a\sqrt{\alpha}} \left(1 + 2e^{-\pi^2/\alpha a^2} \cos \frac{2\pi x}{a}\right) \quad .$$

(Since the sum is presumed to be rapidly convergent, we retain only the $\ell = 0, \pm 1$ terms). Consequently, Eq. (4.3) becomes

$$Z(x) \cong \gamma^{-1} \ln \left(\frac{V_0}{E} \frac{\sqrt{\pi}}{a\sqrt{\alpha}}\right) + \gamma^{-1} \ln \left(1 + 2e^{-\pi^2/\alpha a^2} \cos \frac{2\pi x}{a}\right)$$

$$\cong Z_0 + \frac{2}{\gamma} e^{-\pi^2/\alpha a^2} \cos \frac{2\pi x}{a} \qquad \left(\text{assuming } e^{-\pi^2/\alpha a^2} \ll \frac{1}{2}\right)$$

$$\cong Z_0 + ha \cos \frac{2\pi x}{a} \quad , \tag{4.4}$$

where Z_0 is just a constant (and which therefore only scales Z(x)) and ha is the surface amplitude. One may easily see now that Z(x) does indeed have the canonical form of a corrugated surface and is, as required, periodic in x.

Having made the assumption that the surface atoms oscillate with the normal mode frequencies, the ζ-vectors may be easily generated:

$$\zeta_{z_j}(x) = \frac{\partial Z(x,q)}{\partial q_j^z}\Big|_{\underset{\sim}{q}=0} = \frac{e^{-\alpha(x-ja)^2}}{\sum_{j'} e^{-\alpha(x-j'a)^2}}$$

and

$$\zeta_{x_j}(x) = \frac{\partial Z(x,q)}{\partial q_j^x}\Big|_{\underset{\sim}{q}=0} = \frac{2\alpha}{\gamma} \frac{(x-ja)\, e^{-\alpha(x-ja)^2}}{\sum_{j'} e^{-\alpha(x-j'a)^2}} \qquad (4.5)$$

Before proceeding further, notice that the vector involving displacement of the surface atoms parallel to the surface, $\zeta_{x_j}(x)$, is inversely proportional to the potential parameter γ. Just by looking at the form of Eq. (4.1), it is clear that if the potential function is to mimic a strongly repulsive interaction, then γ must be large, otherwise the impulsive collision approximation is invalid. But for large γ, $\zeta_{x_j}(x) << \zeta_{z_j}(x)$, and thus the x-motion of the surface atoms may essentially be neglected with respect to motion perpendicular to the surface. This neglect of in-plane motion is used almost universally in the work of others, and therefore it is encouraging that our model shows this feature explicitly.

The same methods which lead to the simplification of the summation in Eq. (4.3), namely the use of the Poisson sum rule and retention of only the first harmonic terms, when applied to Eq. (4.5) yield

$$\zeta_{z_j}(x) = \frac{a\sqrt{\alpha}}{\sqrt{\pi}} \frac{e^{-\alpha(x-ja)^2}}{1 + 2e^{-\pi^2/\alpha a^2}\cos\frac{2\pi x}{a}} .$$

Introduction of this form into the expression for W(x) followed by application of the above summation convention then permits one to write, after some algebra,

$$W(x) = \frac{a\sqrt{\alpha}}{2\sqrt{2\pi}} \Delta k_z^2 \langle q_z^2 \rangle \frac{1 + 2e^{-\pi^2/2\alpha a^2}\cos\frac{2\pi x}{a}}{(1 + 2e^{-\pi^2/\alpha a^2}\cos\frac{2\pi x}{a})^2} . \qquad (4.6)$$

Once these last two equations have been obtained, one may construct the one-phonon scattering intensity, I_1, within the context of the preceding approximations, remembering that for our model the sums over phonon modes have been reduced to sums over individual surface atoms. Inasmuch as that calculation is not particularly instructive, we shall dispense with the details here and only indicate the result,

$$I_1 = \frac{N}{2} a^2 \Delta k_z^2 \langle q_z^2 \rangle \{[\delta(\Delta E - \hbar\omega_z) + \delta(\Delta E + \hbar\omega_z)] - \frac{\hbar\omega_z\beta}{4}[\delta(\Delta E - \hbar\omega_z) - \delta(\Delta E + \hbar\omega_z)]\} \sum_{\ell} e^{-\frac{2\pi^2}{\alpha a^2}(\ell - \frac{\Delta k_x a}{2\pi})^2} (|K_\ell^{(1)}|^2 + |K_\ell^{(2)}|^2 + |K_\ell^{(3)}|^2)$$

$$K_\ell^{(1)} = \frac{1}{a}\int_0^a dx\ F(x)$$

$$K_\ell^{(2)} = \frac{2}{a}\int_0^a dx\ F(x)\ e^{-\pi^2/2\alpha a^2}\cos\frac{\pi x}{a}$$

$$K_\ell^{(3)} = \frac{2}{a}\int_0^a dx\ F(x)\ e^{-\pi^2/2\alpha a^2}\sin\frac{\pi x}{a}$$

$$F(x) = e^{-\frac{2\pi i\ell}{a}x}\ e^{-i\Delta k_z Z(x)}\ e^{-W(x)}\ [1 + 2e^{-\pi^2/\alpha a^2}\cos\frac{2\pi x}{a}]^{-1} \quad . \tag{4.7}$$

The integrals appearing above as well as the one which appears in the expression for I_0 may all be done as indicated in Appendix B after inserting the forms for Z(x) and W(x) given by Eqs. (4.4) and (4.6). Doing so, the final result for the elastic and one-phonon intensity contributions through order μ^2 is

$$\begin{aligned} I_{\Delta E,\vec{k}_f\leftarrow\vec{k}_i} &= N\delta(\Delta E)a^2e^{-2\bar{W}}\sum_\ell \delta(\ell - \frac{\Delta k_x a}{2\pi})J_\ell(\lambda)^2 \\ &+ N\{\frac{1}{2}[\delta(\Delta E-\hbar\omega_z) + \delta(\Delta E+\hbar\omega_z)] - \frac{\hbar\omega_z\beta}{8}[\delta(\Delta E-\hbar\omega_z) \\ &- \delta(\Delta E+\hbar\omega_z)]\}a^2\Delta k_z{}^2\langle q_z{}^2\rangle e^{-2\bar{W}}\sum_\ell e^{-\frac{2\pi^2}{\alpha a^2}(\ell - \frac{\Delta k_x a}{2\pi})^2} \\ &\cdot [J_\ell(\lambda)^2 + 4\mu^2(J'_\ell(\lambda)^2 + |S_\ell|^2 + |C_\ell|^2)] \quad , \end{aligned} \tag{4.8}$$

where $\mu = e^{-\pi^2/2\alpha a^2}$ (assumed to be small)

$$\lambda = \Delta k_z h a$$

$$\bar{W} = \frac{a\sqrt{\alpha'}}{4\pi} \Delta k_z^2 \langle q_z^2 \rangle \qquad (4.9)$$

$J_\ell(\lambda)$ = Bessel function of order ℓ

$$J'_\ell(\lambda) = \frac{d}{d\lambda} J_\ell(\lambda)$$

and the integrals S_ℓ and C_ℓ are given by the following series:

$$S_\ell = \sum_{n=0}^{\infty} \frac{(i\lambda)^n}{2^n n!\pi} \sum_{m=0}^{n} \binom{n}{m} \left[-\frac{(-1)^{\ell-n+2m}}{2} \right] \left[\frac{1}{\ell-n+2m+\frac{1}{2}} + \frac{1}{\ell-n+2m-\frac{1}{2}} \right]$$

$$C_\ell = \sum_{n=0}^{\infty} \frac{(i\lambda)^n}{2^n n!\pi} \sum_{m=0}^{n} \binom{n}{m} \frac{(-1)^{\ell-n+2m}}{2} \left[\frac{1}{\ell-n+2m+\frac{1}{2}} - \frac{1}{\ell-n+2m-\frac{1}{2}} \right] ,$$

$\binom{n}{m}$ being the conventional binomial coefficient.

V. Discussion.

The final equations of Section IV provide an analytic form for the scattering intensity pattern which is produced by the assumed pairwise repulsive potential, Eq. (4.1), through first order in the inelastic phonon effects. Clearly, the basic structure is dominated by the elastic diffraction term, producting delta-function peaks whenever Δk_x is equal to a reciprocal lattice vector, although the intensities of these peaks are attenuated by a Debye-Waller factor. This primary structure is then augmented by inelastic lobes on either side of each diffraction peak. (Of course, the actual peak profile may be greatly complicated by the overlap of inelastic lobes with nearby elastic structure). Any direct broadening of the elastic peaks can only be caused in our model by "experimental" effects such as a distribution of incident velocities; broadened inelastic peaks, however, are explicitly predicted as a result of a gaussian distribution of Δk_x values which is centered about the diffraction condition. We should note, though, that if the phonon frequencies are sufficiently low, the expected peak shape would more closely resemble a sharp spike with broad inelastic "wings" near the base. But in any case we would predict that any observed diffraction peak width should correspond roughly to the width of the incident velocity distribution.

To make a quantitative comparison between the theoretical results we have obtained and experimental measurements requires considerable computational work. It would be necessary, e.g., to average our expressions over the appropriate distributions of initial velocities as well as over the finite detector width, and then one could vary the various parameters

in the interaction potential to see if the data were explicable in terms of reasonable values of the parameters. Although such calculations are desirable, they are beyond the scope of this paper, which has been to show how a simple analytical model can account qualitatively for the various aspects of diffraction and of energy transfer. With these limitations in mind, however, a qualitative comparison of Williams[18] results with our model is useful and somewhat encouraging. For example, our model does appear able to reproduce the basic structure and positions of the inelastic scattering lobes, which are seen as low bumps slightly separated from the strong elastic peaks. We also note that some asymmetry in the annihilation and creation intensities may be observed in the experimental measurements, although it appears that those results tend to show the phonon annihilation lobe (i.e., the lobe shifted toward the surface normal) to be somewhat more intense than the corresponding creation lobe, whereas our model would predict that phonon creation would be the dominant effect if any asymmetry is present. [In the limit of a very cold surface, it is clear that phonon creation should be the principal energy transfer process simply because there are relatively few excited phonon modes available for annihilation. Eq. (3.7b) explicitly yields such a result for appreciable values of β (i.e., for low surface temperature).] Although at present one really cannot elaborate further on the agreement with experiment, it should be mentioned that essentially no features are yielded by the proposed model which cannot be roughly correlated with the observed intensity structure.

Inasmuch as there does seem to be reasonable qualitative agreement between the model and experiment, it is interesting to consider why the modified Einstein description of the solid appears to be adequate. The obvious conclusion to draw is that actually only a narrow range of phonon mode frequencies contributes significantly to the scattering, and thus most solid descriptions give essentially the same results. Generally it appears that our basic assumptions, namely an Einstein solid and a periodic surface, compare very favorably with the more customary postulate of a Debye solid with a flat surface. Certainly in the limit where the two solid models do yield effectively identical phonon structures, the ability to describe both diffraction and inelastic transitions within a single unified formalism is indeed a definite advantage.

Aside from the omission of any experimental averaging as mentioned above, there is one more general feature of the gas-surface scattering problem which we have ignored. That feature involves the presence of a

long-range attractive part in the actual two-body potential. As mentioned by others, inasmuch as the detailed structure of such an attractive potential apparently has little or no bearing on the scattering pattern in the absence of surface trapping, a simple square well form for the attraction seems adequate. We have chosen to ignore the well altogether in our model by making the assumption that the only consequence of considering such a well is the addition of a momentum increment perpendicular to the surface to an approaching gas atom and the subtraction of a corresponding increment from the scattered atom. Naturally, this quasiclassical assumption leads to a change in the actual incident and predicted scattered angles, although these modified angles may, of course, be simply related to the experimentally observed angles.[19] But because the scattering pattern has the same qualitative features with or without a well, we have opted to neglect completely the presence of any attractive well.

Before concluding, we wish to stress the point that the width of the inelastic scattering lobes are, as expected, related to the degree of inelasticity present. This width, arising as a result of a gaussian distribution of Δk_x values in the I_1 term, may be correlated with the effective Debye-Waller factor, Eq. (4.9), which to order μ^2 is just a multiplicative constant. Specifically, for this particular gaussian distribution the standard deviation may be written

$$\sigma = \frac{a\sqrt{\alpha}}{2\pi} \quad .$$

Notice then that with this definition one may write the effective Debye-Waller exponent as

$$\bar{W} = \frac{\sigma}{2} \Delta k_z^2 \langle q_z^2 \rangle \quad .$$

Hence for smaller σ, i.e., for a narrower distribution in Δk_x, W is also smaller, which implies that e^{-2W} is closer to unity--this is indeed the expected concerted behavior for a system which is becoming less inelastic. Furthermore, since σ depends on the potential parameter α, one may conclude that a decrease in α, corresponding to a "loosening" of the potential in the x-direction, would simultaneously cause a reduction in the observed inelastic intensity.

Finally, in conclusion, we summarize the qualitative features of the results obtained from our scattering model:

(1) the elastic scattering peaks are infinitely sharp if the incident atomic beam is monoenergetic;

(2) the inelastic scattering peaks are broadened even for a monoenergetic beam, with the peak widths being dependent upon the interaction potential parameters and not upon either the surface temperature or the collision energy;

(3) the effective Debye-Waller factor, Eq. (4.9), shows the expected temperature dependence (inasmuch as it is a function of $\langle q_z^2 \rangle$), and appears to first order as just a multiplicative term;

(4) the surface amplitude, ha, is independent of the collision energy in the static surface limit, although the actual position of the potential turning-point contour <u>is</u> a function of E;

(5) the effect of the in-plane motion of the surface atoms is negligible as compared with the effect due to motion perpendicular to the surface plane;

(6) the symmetry of the one-phonon annihilation and creation lobes does

exhibit a temperature dependence, with the two being totally symmetric only in the high temperature limit (the positions of the lobes do not, however, show any such dependence).

Overall, the present work provides a very convenient and instructive model for the gas-surface collision problem. Although in principle the formalism allows one to treat all possible n-phonon inelastic scattering processes, we have shown that it is possible to obtain a good qualitative agreement with experiment by only examining the one-phonon effect. It is also encouraging that such results are obtainable from a one-dimensional surface model, even though it is reasonable to expect that extension could be made to a two-dimensional lattice with little difficulty. We feel particularly confident that the absence of the commonly used flat-surface assumption provides a definite advantage in that the treatment of a wider range of structured surfaces becomes possible.

Appendix A: Summation Over Unit Cells

The coordinate integrals over the interval $[-\infty,\infty]$ may be transformed to integrals over the interval $[0,a]$, where a is the unit cell length, via the identity

$$\int_{-\infty}^{\infty} dx\ f(x) = \sum_n \int_0^a dx\ f(x+na)$$

where the integer n numbers the unit cells. For example, by using this formula the elastic contribution to the intensity may be written

$$I_0 = \delta(\Delta E)\left| \sum_n \int_0^a dx\ e^{-i\Delta k_x (x+na)}\ e^{-i\Delta k_z Z(x+na)}\ e^{-W(x+na)} \right|^2 . \quad (A.1)$$

But since $Z(x) = Z(x+na)$ by assumption and since presumably $W(x) = W(x+na)$ inasmuch as functions of x only must exhibit the periodicity of the lattice (obviously any model for these functions must bear out this assumption), Eq. (A.1) becomes

$$I_0 = \delta(\Delta E)\left| \int_0^a dx\ e^{-i\Delta k_x x}\ e^{-i\Delta k_z Z(x)}\ e^{-W(x)} \sum_n e^{-i\Delta k_x na} \right|^2$$

$$= \delta(\Delta E)\left| \int_0^a dx\ e^{-i\Delta k_x x}\ e^{-i\Delta k_z Z(x)}\ e^{-W(x)} \right|^2 \sum_n \sum_{n'} e^{-i\Delta k_x a(n-n')} .$$

Defining $\bar{n} = \frac{n+n'}{2}$ and $\Delta n = n-n'$ and then resumming (noting that the sum over $\bar{n}$ just gives N, the total number of unit cells within the physical limits of the experiment),

$$I_0 = N\delta(\Delta E)\left|\int_0^a dx\, e^{-i\Delta k_x x}\, e^{-i\Delta k_z Z(x)}\, e^{-W(x)}\right|^2 \sum_{\Delta n} e^{-i\Delta k_x a \Delta n} \quad .$$

Then, by using the Poisson sum rule one finally obtains

$$I_0 = N\delta(\Delta E) \sum_{\ell} \delta\left(\ell - \frac{\Delta k_x a}{2\pi}\right)\left|\int_0^a dx\, e^{-i\Delta k_x x}\, e^{-i\Delta k_z Z(x)}\, e^{-W(x)}\right|^2 \quad .$$

Appendix B: Debye-Waller Integrals

From Eq. (3.7a) we need to calculate

$$|S_\ell^0|^2 = \left|\frac{1}{a}\int_0^a dx\, e^{-\frac{2\pi i\ell}{a}x}\, e^{-i\Delta k_z Z(x)}\, e^{-W(x)}\right|^2 .$$

Linearizing the Debye-Waller exponential,

$$|S_\ell^0|^2 \cong \left|\frac{1}{a}\int_0^a dx\, e^{-\frac{2\pi i\ell}{a}x}\, e^{-i\Delta k_z Z(x)} - \frac{1}{a}\int_0^a dx\, e^{-\frac{2\pi i\ell}{a}x}\, e^{-i\Delta k_z Z(x)}\, W(x)\right|^2$$

$$\cong |S_{\ell(s)}^0 - \Delta S_\ell^0|^2$$

$$\cong \left|S_{\ell(s)}^0\left(1 - \frac{\Delta S_\ell^0}{S_{\ell(s)}^0}\right)\right|^2 .$$

Then by resumming, with the hope of recovering some of the higher order contributions lost in the linearization, $|S_\ell^0|^2$ may be written in terms of $|S_{\ell(s)}^0|^2$, the result for the static surface limit, as

$$|S_\ell^0|^2 \cong |S_{\ell(s)}^0|^2\, e^{-2\,\mathrm{Re}\frac{\Delta S_\ell^0}{S_{\ell(s)}^0}} . \qquad (B.1)$$

Substituting Eqs. (4.4) and (4.6) for Z(x) and W(x),

$$S_{\ell(s)}^0 = \frac{1}{a}\, e^{-i\Delta k_z Z_0}\int_0^a dx\, e^{-\frac{2\pi i\ell}{a}x}\, e^{-i\lambda\cos\frac{2\pi x}{a}} = e^{-i\Delta k_z Z_0}\, e^{-i\frac{\pi}{2}\ell}\, J_\ell(\lambda)$$

and

$$\Delta S_\ell^0 = \frac{\sqrt{\alpha}}{4\pi} \Delta k_z^2 \langle q_z^2 \rangle e^{-i\Delta k_z Z_0} \int_0^a dx\, e^{-\frac{2\pi i \ell}{a}x} e^{-i\lambda \cos\frac{2\pi x}{a}} \frac{1+2\mu\cos\frac{2\pi x}{a}}{(1+2\mu^2\cos\frac{2\pi x}{a})^2} .$$

Assuming μ to be small (at constant surface amplitude, ha), one then expands the quotient above in a Taylor series and integrates term by term to obtain

$$\Delta S_\ell^0 = \bar{W}\, e^{-i\Delta k_z Z_0}\, e^{-i\frac{\pi}{2}\ell} [J_\ell(\lambda) + i(2\mu - 4\mu^2) J_\ell'(\lambda)]$$

to order μ^2.

Thus, to this order in μ, Eq. (B.1) may be evaluated as

$$|S_\ell^0|^2 \cong J_\ell(\lambda)^2\, e^{-2\bar{W}} .$$

This same approximation procedure is then used for the calculation of the integrals in Eq. (4.7), still retaining only the terms through μ^2. By this method, the integrals S_ℓ and C_ℓ in Eq. (4.8) are found to be of the form

$$S_\ell = \frac{1}{\pi} \int_0^\pi d\theta\, \sin\ell\theta\, \sin\frac{\theta}{2}\, e^{i\lambda\cos\theta}$$

$$C_\ell = \frac{1}{\pi} \int_0^\pi d\theta\, \cos\ell\theta\, \cos\frac{\theta}{2}\, e^{i\lambda\cos\theta} .$$

An expansion of the exponential followed by term-by-term integration yields the series solutions given in Section IV.

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