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Inelastic atom–surface scattering with large numbers of phonons

J R Manson

Department of Physics and Astronomy, Clemson University, Clemson, SC 29634, USA

Abstract. We consider the interaction of a neutral atomic projectile colliding with a solid surface in the limit in which large numbers of vibrational quanta are exchanged, while at the same time the projectile itself may be moving with momenta corresponding to quantum mechanical motion.

The diffuse multiphonon background observed in energy resolved He–surface scattering is now understood [1, 2]. In most He scattering experiments, the conditions are quantum mechanical: sharp features such as diffraction lines and single-phonon peaks can be resolved in the scattered intensity. However, for some experiments the inelastic scattering is so strong that elastic and single-quantum features cannot be resolved [3]. Such behaviour occurs under near-classical conditions of large projectile momenta and high surface T. However, large numbers of vibrational quanta can be involved when part of the system is in the quantum regime. Examples are a small momentum projectile colliding with a very hot surface, or with a surface made up of low-mass atoms. A quite different example would be a fast semiclassical projectile colliding with a surface at very low T where quantum mechanical zero-point motion would dominate the phonon exchange. We demonstrate here that for all of these cases, simple closed-form expressions for the scattering intensity can be obtained. The scattered intensity is proportional to the transition rate for scattering from the incident projectile state $\boldsymbol{k}_i$ to the final state $\boldsymbol{k}_f$ given by the generalized golden rule

$$w(\boldsymbol{k}_f, \boldsymbol{k}_i) = \frac{2\pi}{\hbar}\left\langle\!\!\left\langle \sum_{n_f} |T_{fi}|^2 \delta(E_f - E_i)\right\rangle\!\!\right\rangle \tag{1}$$

where $\langle\langle\rangle\rangle$ signifies an average over initial crystal states and the sum is over final crystal states. For the multiphonon part of the scattered intensity, the energy exchange is usually dominated by low-frequency modes, and the transition matrix element contributions from each surface unit cell:

$$T_{\boldsymbol{k}_f,\boldsymbol{k}_i}(t) = \sum_{l,\kappa} \exp\{-i\boldsymbol{k}\cdot[\boldsymbol{r}_l + \boldsymbol{r}_\kappa + \boldsymbol{u}_{l,\kappa}(t)]\}\tau^{\kappa}_{\boldsymbol{k}_f,\boldsymbol{k}_i}. \tag{2}$$

Equation (2) is a quick-collision approximation i.e. the transition matrix depends on the surface displacement only through the relative phase arising from the path length. Its validity for He scattering has been established through a number of calculations [1, 2].

In the large-phonon-number limit, when the Debye–Waller exponent $2W(\boldsymbol{k})/6 \gg 1$ where $2W(\boldsymbol{k}) = \langle\langle|\boldsymbol{k}\cdot\boldsymbol{u}_{l,\kappa}(t)|^2\rangle\rangle$, the differential reflection coefficient (1) becomes

$$\mathrm{d}R/\mathrm{d}\Omega_f\,\mathrm{d}E = (m^2|\boldsymbol{k}_f|/8\pi^3\hbar^5 k_{iz})|\tau_{fi}|^2(\hbar\pi/\omega_0 k_B T)^{1/2}\exp[-(E+\hbar\omega_0)^2/4k_B T\hbar\omega_0] \tag{3}$$

where $\hbar\omega_0 = \hbar^2 k^2/2M$ with k the total momentum exchange and M the mass of the crystal atoms. The scattering intensity is Gaussian-like in the energy exchange E, but shifted to the energy loss side by the amount $\hbar\omega_0$. However, this function is not a true Gaussian

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because ω_0 is a function of k. It is limited on the energy loss side by the fact that the projectile may lose no more energy than it had initially, and it is skewed towards the energy gain side by the energy dependence of the ω_0. The width of the differential reflection coefficient in energy exchange is roughly $2\sqrt{k_B T\hbar\omega_0}$, and the peak amplitude decays with increasing temperature as $(k_B T\hbar\omega_0)^{1/2}$. This behaviour of the width and peak intensity of the differential reflection coefficient is understandable in terms of the unitarity condition, which guarantees the equality of the number of scattered particles to the number of incident particles. At higher T the intensity spreads over a large range of E; consequently in order to preserve the number of particles, the peak must decrease. Equation (3) is the semiclassical limit for a point particle scattering from a lattice of centres, in which case the coherence region shrinks to where the incoming projectile scatters with only a single surface atom. There is a second classical limit that is applicable in the case of a projectile that is large compared to the inter-atomic lattice spacing, when the surface can be considered to be a continuum. This equation is obtained from (1) by replacing the summation by an integral, which then can be evaluated in the classical limit by the method of steepest descents. The final result is

$$\begin{aligned} dR/d\Omega_f\,dE = {} & (m^2|k_f|/4\pi^3\hbar^5 k_{iz} S_{uc})|\tau_{fi}|^2 v_R^2(\hbar\pi/\omega_0 k_B T)^{3/2} \\ & \times \exp\{-[(E+\hbar\omega_0)^2 + 2\hbar^2 v_R^2 K^2]/4k_B T\hbar\omega_0\} \end{aligned} \tag{4}$$

where v_R is a weighted average of surface phonon velocities parallel to the surface. There is a difference between the two classical limits (3) and (4). In the latter the temperature dependence of the peak in the inelastic intensity varies as $1/T^{3/2}$ as opposed to $1/T^{1/2}$, and the extra Gaussian behaviour appears in parallel momentum exchange.

There is another case in which a semiclassical closed-form expression can be obtained, i.e. when the particle momentum is so great that $2W$ (or equivalently, the number of phonons exchanged) is large, but the surface is at low temperature where zero-point motion is important. In this case the differential reflection coefficient is again a skewed Gaussian

$$\begin{aligned} dR/d\Omega_f\,dE = {} & (m^2|k_f|/4\pi^3\hbar^5 k_{iz} S_{uc})|\tau_{fi}|^2 v_R^2[8\pi/3\omega_0\omega_D\Omega(T)]^{3/2} \\ & \times \exp\{[2(E+\hbar\omega_0)^2 + 4\hbar^2 v_R^2 K^2]/3\hbar^2\omega_0\omega_D\Omega(T)\} \end{aligned} \tag{5}$$

where ω_D is the Debye frequency and $\Omega(T) = 8\int_0^1 x^3[n(\hbar\omega_D x/k_B T) + \frac{1}{2}]\,dx$ approaches unity as $T \to 0$.

We note that the classical limit exhibits the approach to equilibrium for the scattering of a low-energy beam of particles from a very hot surface [4]. The Gaussian function in equation (4), in the limit $E_f \gg E_i$, is an exponential in E_f. Using the energy exchange equation $E_f = 4\mathcal{E}mM/(M+m)^2$ for a pairwise collision with the crystal atom of energy $\mathcal{E}$ the scattered intensity becomes

$$dR/d\Omega_f\,dE \propto T^{-3/2}\exp(-\mathcal{E}/k_B T). \tag{6}$$

This shows that the particles leave the surface with an energy distribution having the exponential dependence of the Maxwell–Boltzmann distribution function.

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