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Energy loss and inelastic diffraction of fast atoms at grazing incidence

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ABSTRACT

The diffraction of fast atoms at grazing incidence on crystal surfaces (GIFAD) was first interpreted only in terms of elastic diffraction from a perfectly periodic rigid surface with atoms fixed at equilibrium positions. Recently, a new approach has been proposed, referred here as the quantum binary collision model (QBCM). The QBCM takes into account both the elastic and inelastic momentum transfers via the Lamb-Dicke probability. It suggests that the shape of the inelastic diffraction profiles are log-normal distributions with a variance proportional to the nuclear energy loss deposited on the surface. For keV Neon atoms impinging on a LiF(001) surface under an incidence angle θ, the predictions of the QBCM in its analytic version are compared with numerical trajectory simulations. Some of the assumptions such as the planar continuous form, the possibility to neglect the role of lithium atoms and the influence of temperature are investigated. A specific energy loss dependence $\Delta E \propto \theta^7$ is identified in the quasi-elastic regime merging progressively to the classical onset $\Delta E \propto \theta^3$. The ratio of these two predictions highlights the role of quantum effects in the energy loss.

1. Introduction

The energy loss of keV ions at solid surfaces has been investigated in detail both from the theoretical and experimental points of view. One of the important regimes at low energy is the nuclear regime where electronic excitations play a minor role. At grazing angle, collisions of keV atoms can be gentle enough to allow scattering in a quantum regime as illustrated by clear diffraction features (see e.g. [1] for a review). The identification of elastic fast atom diffraction, characterized by the absence of energy exchange with the surface, was predicted almost ten years ago [2] but experimental evidences of a well defined Laue circle with diffraction spots size limited by that of the primary beam were scarce and hardly quantified [3–5].

A first attempt to quantitatively describe both the elastic and inelastic diffraction of fast atoms, together with the associated line profiles, was recently proposed [6]. The succession of binary collisions with surface atoms along the classical trajectory was described using an idealized trajectory giving close analytic form of the energy loss and inelastic profiles. Starting from the quantum properties of the individual surface atoms considered as harmonic oscillators during a distant binary collision, the model offers a smooth transition between the quantum and classical regimes with specific predictions on the onset for the nuclear energy loss.

After a short presentation of the QBCM, some of its assumptions are analyzed with more realistic trajectory simulation using the Ne-LiF system to better understand the limitations. Accordingly less importance will be given to an *a priori* justification since these can be discussed in view of the simulations.

2. Established theory

2.1. Elastic diffraction; the rigid lattice and the potential energy landscape

Theoretical approaches to GIFAD consider the surface as an ideal system with atoms standing still at their equilibrium positions so that the potential energy landscape (PEL) of the helium-surface is perfectly periodic. The dynamics of the projectile atomic wave-function on this PEL, *i.e.* the diffraction, has been modeled via wave-packet [10,11], transition matrix [2], semi-classical trajectories [1,12], Bohmian trajectories [13], close coupling [8,14] or multi-channels Hartree methods [15].

2.2. Reduced dimension of the PEL

Elastic diffraction of fast atoms of energy E impinging on a surface with an incidence angle θ_{in}, was early understood[10] and described by a 2D problem where an effective particle with perpendicular energy $E_\perp = E\sin\theta_{in}$ evolves in a 2D PEL $V_{2D}(y,z) = \int V_{3D}(x,y,z)dx/d_x$. Here x is taken along the low index crystal axis as depicted in Fig. 1 and d_x is the period along x. This axial channeling approximation (ASCA) was

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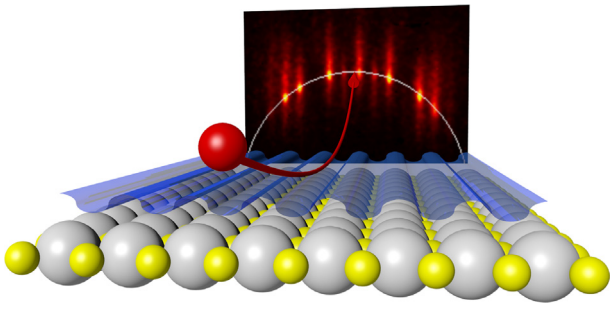

Fig. 1. Schematic view of keV Ne atoms impinging on the LiF(0 0 1) surface. The diffraction pattern is recorded on an detector [7] placed ∼ 1 m downstream. The elastic diffraction spots are localized on the Laue circle (white line) while the inelastic intensity shows elongated streaks around the Laue circle.

established with quantitative criterion well satisfied for grazing incidence keV projectile and small lattice units [14,15]. Experimentally this is evidenced by the presence of only one Laue circle.

Here we focus on inelastic processes which are described as individual binary collisions along the projectile's travel along x, *i.e.* precisely the direction neglected in ASCA. We assume that the mean properties of these trajectories are well estimated by the trajectory on the mean planar potential defined as

$V_{1D}(z) = \frac{1}{d_x d_y}\int\int V_{3D}(x,y,z)dxdy$ where d_y is the period along the direction y. Assuming an exponential form for $V_{1D}(z) \propto e^{-\Gamma z}$, the trajectory $z(t)$ is analytic and so are its first and second derivatives $p_z(t)$ and $\dot{p}_z(t)$ describing the momentum transfer to the surface per unit time or unit length. Considering that only one atom with mass m per lattice unit a receives the exchanged momentum, an energy deposition curve can be defined and integrated to produce an energy loss [2,16].

$$E_{loss} = \frac{2}{3}\mu E\Gamma a\theta_{in}^3. \tag{1}$$

where $\mu = M/m$ with M the projectile mass and m the mass of a surface atom. The energy deposition curve has a quasi-gaussian profile [6] and its full width at half maximum can be used to define the trajectory length $L \propto 1/\theta$ [10]. This important parameter L can be expressed as the number of most active binary collisions. If one assumes that all collisions are equivalent, the incident beam is deflected by an angle $2\theta_{in} = \theta_{in} + \theta_{out}$, arising from a series of N_{eq} equivalent deflections by , each of them producing a recoil energy $E_r = \mu E\delta\theta^2$ with

$$N_{eq} = \frac{6}{\Gamma a\theta_{in}}. \tag{2}$$

2.3. Thermal movement of the surface atoms

At finite temperature the crystal hosts a population of phonon giving rise to movement with an amplitude $\sigma_z^2 = \langle z^2\rangle$.

Considering the coupled oscillators and averaging over the thermal distribution, the z distribution is gaussian with [17]

$$\langle z^2\rangle = \frac{3\hbar}{2m\omega}\coth\left(\frac{T_D}{2T}\right) = \frac{3\hbar^2}{2mk_BT_D}\coth\left(\frac{T_D}{2T}\right); \tag{3}$$

where k_B is the Boltzmann constant, T_D is the Debye surface temperature so that $\hbar\omega = k_BT_D$ is the energy of a vibration quantum of the local Debye oscillator. Interpreting $\sigma_z^2 = \langle z^2\rangle$ as the variance of the probability to find a surface atoms away from the surface plane, the surface is far from being as flat as idealized in the rigid lattice model. These displacements induce deviation from the ideal trajectory and affect the coherence of the diffracted signal.

As often encountered in quantum mechanics, the situation can be approached in two ways, either in the real space as summarized below or in the momentum space described in the next section. The real space approach considers the coherence of the waves emitted by an ensemble of diffraction centers distributed around their equilibrium positions. In thermal energies atom scattering or in Xray diffraction, where the scattering takes place on a single atom, this gives rise to the Debye–Waller factor $DWF = e^{-\langle(k_z z)^2\rangle} \sim e^{-k_z^2\langle z^2\rangle}$ for the specular reflection of a wave-vector k_z. In GIFAD, the momentum exchange is spread along the successive tiny collisions with the surface atoms and each one only contributes with a dephasing $d\phi = \delta k_z z$ where δk_z can be estimated as $\delta k_z = 2k_z/N_{eq}$. All these contributions add up incoherently so that the overall dephasing is reduced. The specific DWF for GIFAD is much more favorable [16,2] $e^{-k_z^2\sigma_z^2/N_{eq}}$, indicating that the scattering takes place on a row of N_{eq} active atoms reducing by $\sqrt{N_{eq}}$ the amplitude of the thermal oscillations.

In both cases, the elastic signal corresponds to atoms at their equilibrium position, and its intensity is attenuated by thermal displacement. The fate of the incoherent signal is however less clear. Where does it appear? Under what conditions the diffraction features remain visible? In other words, how can we describe diffraction pattern in the inelastic regime? These questions are easier to address with the momentum approach at the heart of the binary quantum collision model.

3. The quantum binary collision model QBCM

The momentum approach describes the elastic scattering on the surface as a series of elastic collisions with the quantum oscillators. If a collision is elastic, the trajectory will again correspond to the classical trajectory associated with the center of the harmonic oscillator *i.e.* as if the rigid lattice description with motionless atoms at their equilibrium positions were real. If q is the momentum transferred to this surface atom, then the probability p_e to leave the wave function unchanged is $p_e = |\langle\psi|e^{iqz}|\psi\rangle|^2$. Using the Bloch theorem [18] $\langle e^{iqz}\rangle = e^{-\frac{1}{2}q^2\langle z^2\rangle}$, the elastic probability is again the standard DWF. For an isolated oscillator with pulsation ω in its ground state the probability is $p_e = e^{-E_r/\hbar\omega}$ with $E_r = \hbar^2q^2/2m$ the associated recoil energy. This is equivalent to the Lamb-Dicke probability of recoilless emission which means that in a trapping potential, the wave function may absorb a momentum q without exchanging the recoil energy E_r. In this respect, E_r is only a virtual recoil energy.

Taking into account the actual value of $\langle z^2\rangle$ on the surface given in Eq. (3), p_e reads

$$p_e = exp\left(-3\frac{E_r}{k_BT_D}\coth\left(\frac{T_D}{2T}\right)\right). \tag{4}$$

The product probability P_e that all binary collisions are elastic factorizes to outline the sum of all the virtual recoil energies $E_{loss} = \Sigma_j E_{rj}$ along the trajectory

$$P_e = exp\left(-3\frac{E_{loss}}{k_BT_D}\coth\left(\frac{T_D}{2T}\right)\right). \tag{5}$$

If a collision with the surface atom is inelastic, different properly-weighted initial and final wave functions have to be evaluated. We can also consider that the momentum dispersion induced by the inelastic collision can be evaluated from classical mechanics with thermally displaced atoms, as if the inelastic collision would project the wave function to its spatial probability distribution $P(z) = |\langle\Psi|z|\Psi\rangle|^2$ which is the gaussian distribution of Eq. (3). Since $P(z)$ is centered around the equilibrium value, the median value of the angular distribution $P(\theta_i)$ is the elastic value θ_e with a, yet unknown, width σ_{θ_i}. Considering these inelastic angular straggling as independent, their contributions are added quadratically for each inelastic collision along the trajectory. The individual inelastic contributions σ_{θ_i} can be calculated numerically or, within few assumptions on the form of the interaction potential, they can be evaluated analytically.