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Grazing incidence fast atom diffraction, similarities and differences with thermal energy atom scattering (TEAS)

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Grazing Incidence Fast Atom Diffraction, similarities and differences with thermal energy atom scattering (TEAS)

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Grazing incidence diffraction of fast atoms at surfaces (GIFAD) has made rapid progress and has established itself as a surface analysis tool where the effective energy E_{\perp} of the movement towards the surface is in the same range as in TEAS. To better compare the properties of these techniques, we review our results using the diffraction of helium or neon atoms impinging on a LiF(001) surface as a model system. E -scan, θ -scan, and ϕ -scan are presented where the primary beam energy E is varied between a few hundred eV up to five keV, the angle of incidence θ_i between 0.2 and 2 deg. and the azimuthal angle ϕ_i around 360 deg. The resulting diffraction charts are analyzed in terms of high and low values of the effective energy E_{\perp} . The former provides high resolution on the positions of the surface atoms and the attached repulsive interaction potentials while the second is sensitive to the attractive forces towards the surface. The recent progress on the inelastic diffraction is briefly presented.

1 Introduction, grazing incidence

Since this review paper is a part of a special issue with several contribution on atomic diffraction, we focus on the recently developed extension; grazing incidence fast atom diffraction at surfaces (GIFAD), and address the reader to other contributions for an in-depth discussion of the thermal energy atom scattering (TEAS or HAS) developed in the '70s by J.P. Toennies *et al.*¹, J. Lapujoulade *et al.*², Mattera *et al.*³ and others as reviewed in several papers^{4–9}. To better compare both techniques, we will focus on the extensively studied model system of helium or neon atoms impinging on a LiF(001) surface.

Grazing incidence fast atom diffraction was discovered fifteen years ago^{10–12} but could have been observed much earlier because it requires only a comparatively simple setup¹³: a commercial keV ion source, a custom neutralization cell, and a position-sensitive detector which are easily available since the advent of the micro-channel plate for night vision devices in the '80s. The diffraction of fast atoms under grazing incidence was predicted by Andreev¹⁴ a few years before its first observation but the paper did not attract attention, probably because it was firmly believed that the inelastic processes would lead to decoherence washing out diffraction features.

Soon after the first observation, it was realized that GIFAD has strong similarities with Reflection High-Energy Electron Diffraction

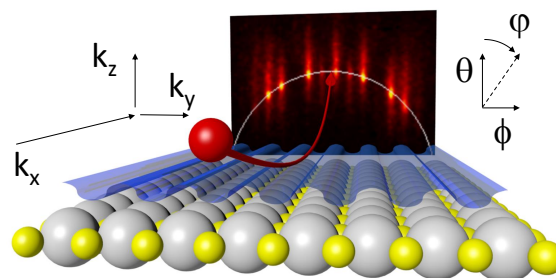


Fig. 1 Illustration of a GIFAD experiment. The primary beam of Ne neutral atoms bounces a few Å above a crystal surface. When the beam is aligned with a low index direction, it is diffracted by the well-aligned rows of surface atoms before being collected on an imaging detector.

(RHEED)¹⁵ as well as with Thermal Energies Atom Scattering (TEAS). It was immediately noted that the fast movement along the low index direction, taken here as the x axis, is strongly decoupled¹⁰ from the much slower movement in the perpendicular (y, z) plane (see Fig. 1). We use here the notation $\vec{k}_i(k_{ix}, k_{iy}, k_{iz})$ and $\vec{k}_f(k_{fx}, k_{fy}, k_{fz})$ to define the components of the projectile wave-vector respectively before and after scattering on the surface. The energy $E_{\perp} = \hbar^2(k_{iy}^2 + k_{iz}^2)/2m$ of the motion in this plane corresponds to a helium particle evolving in the axially averaged interaction potential $V_{2D}(y, z)$ (see Eq. 1). In GIFAD, the energy E_{\perp} can be tuned in a broad range of values by tuning the angle of incidence so that diffraction features have been observed from a few meV up to several eV, covering the energy range used in standard TEAS experiments.

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This decoupling from the slow to the fast motion, called axial surface channeling approximation (ASCA), has been investigated in detail^{10,16,17}, and only experimental evidence that the low index direction actually provides a reference axis in GIFAD experiments will be presented here.

The paper is organized as follows: a brief description of the specific geometry needed for fast atom diffraction, then the standard GIFAD measurements are presented in detail. First using static diffraction patterns to illustrate the elastic and inelastic diffraction. Then, assuming elastic diffraction, variations of the projectile energy and angles of incidence are presented. These are θ -scan, ϕ -scan, and E-scan. Then high and low values of the effective energies are analyzed to outline the sensitivity to surface topology and attractive forces.

2 Projectile trajectory.

The large kinetic energy used in GIFAD provides a very high detection efficiency compatible with the use of position sensitive detector allowing the full diffraction image to be recorded at once and within seconds to minutes. However, the fundamental difference with TEAS is not the energy but the grazing scattering geometry which imposes the successive interaction with many atoms of the surface. Each of these interactions taking place at large distance from the surface (few Angstrom) induces only a tiny deflection but these sum up coherently to produce a significant deflection both in the collision plane where the momentum component directed towards the surface is reversed for specular reflection and in the perpendicular direction. The smooth dashed red line in Fig. 2a represents the projectile trajectory $z(x)$ of a 1 keV helium atom impinging at 1° incidence on top of an atomic row of F^- ions of a LiF surface oriented along the $\langle 110 \rangle$ direction. Compared with

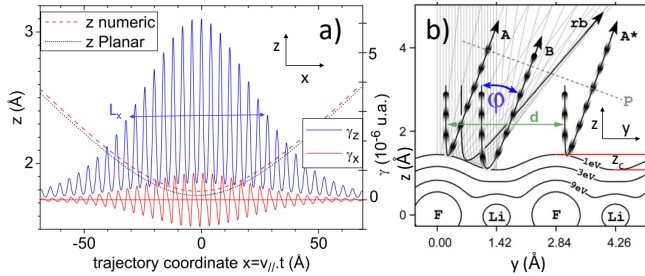


Fig. 2 a) Classical trajectory $z(x)$ of a 1 keV ^4He atom impinging at 1° incidence along the $\langle 110 \rangle$ direction on the LiF(001) surface¹⁸. The full blue and red curves represent the vertical and horizontal components of the acceleration vector. The red dashed line is calculated with the full 3D potential $V_{3D}(\vec{r})$ and is almost identical to the analytic one calculated with the mean repulsive planar potential, $V_{1D}(z)$ in eq.1. b) Projectile trajectories $z(y)$ and equi-potential lines of the 2D interaction potential $V_{2D}(y,z)$ relevant for the scattering in the (y,z) plane (taken from ref. ¹⁹).

the horizontal x scale, the vertical z scale, on the left-hand side, had to be magnified by a factor close to fifty to avoid looking like a horizontal straight line. Nevertheless, the interaction with the individual surface atoms is very well localized as illustrated with the peaks on the blue and red curves representing the acceleration in the vertical and horizontal directions respectively. These represent the force and momentum exchange between the helium pro-

jectile and the surface atoms. Along the z direction, all peaks are positive, pushing the projectile away from the surface while along x the projectile is first slowed when approaching a surface atom and accelerated when leaving resulting in a vanishingly small integral as predicted by Henkel *et al.*²⁰ and observed by Farias *et al.*²¹ in TEAS at grazing incidence. The envelope of these sharp peaks has a quasi-Gaussian envelope with a well defined standard deviation. This width represents the mean length of interaction and, neglecting the attractive forces, it scales with the inverse of the angle of incidence θ_i ^{18,22}. The more grazing the angle of incidence, the broader the interaction curves and also the weaker the amplitude because the integral is proportional to twice the initial momentum towards the surface to ensure specular reflection. In section 7, all the inelastic effects specific to GIFAD will be interpreted as due to these numerous gentle collisions. In the perpendicular (y,z) plane (Fig.2b), from ref.¹⁹), projectile trajectories resemble the one observed from standard TEAS collisions with a slow projectile coming 90° ($\varphi=0$) above the surface and bouncing on the surface atoms. However, the potential energy curves correspond to the 3D potential energy landscape used in TEAS but averaged along the low index direction as briefly discussed in the next section.

3 A GIFAD setup.

Fig. 3 shows a schematic drawing of a GIFAD setup with two opposite arms connected to the main UHV chamber hosting the crystal surface. The left-hand side is for the neutral beam production while the right-hand-side is for the detection. Both are essentially long tubes of reduced diameter leaving full access to the 2π volume above the surface. For an incidence angle θ_i and azimuthal angle ϕ_i , $\vec{k}_i = k_i(\cos \theta_i \cos \phi_i, \cos \theta_i \sin \phi_i, \sin \theta_i) \approx k_i(1, \sin \phi_i, \sin \theta_i)$.

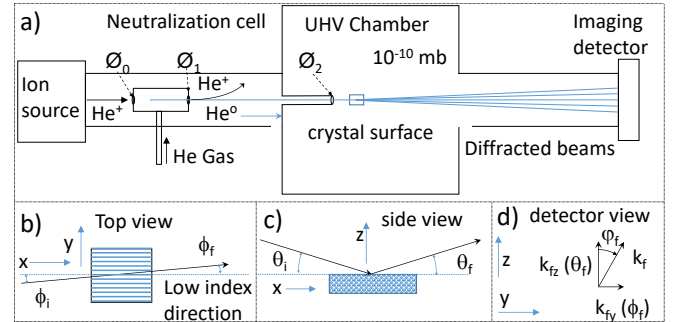


Fig. 3 Scheme of the experimental setup. a) The He^+ ions extracted from the ion source at keV energy, are converted into neutral He^0 atoms after resonant charge exchange into a gas cell. The neutral beam is collimated by two diaphragms \varnothing_1 and \varnothing_2 to control its angular spread $\delta\theta$. After reflection on the crystal surface in the UHV chamber, the diffracted beams are collected on an imaging detector ≈ 1 m downstream. b), c) and d) are schematic views of the Scattering geometry with definitions of the incidence and azimuthal angles θ and ϕ and φ .

3.1 Beam and surface properties.

Neutral atoms are produced by resonant charge exchange into a 2 cm long cell filled with helium at a pressure estimated around 10^{-3} mbar. With electron capture cross-sections on the order of 10^{-15} cm^2 , this is enough to ensure that approximately 10% of the

ion beam will be converted into a beam of neutral atoms having almost the same energy and angle as the ion beam while reducing the straggling associated with multiple collisions. Two small diaphragms of diameter \varnothing_0 and \varnothing_1 are installed at both ends of the neutralization cell to reduce leaks from the helium gas. The exit hole \varnothing_1 also defines the source size of the atomic beam. The angular spread $\delta\theta$ of the atomic beam is defined by a second diaphragm \varnothing_2 located at a distance L downstream so that the angular resolution is $\delta\theta \simeq (\varnothing_1 + \varnothing_2)/4L$. To resolve diffraction, the angular resolution should be less than the Bragg angle associated with the surface, $\phi_B \simeq G_y/k$ where k is the projectile wave number and $G_y = 2\pi/a_y$ is the reciprocal lattice vector associated with the periodic arrangement a_y of the atomic rows. G_y is typically less than a few \AA^{-1} while a 1 keV He atom wavenumber k is larger than one thousand \AA^{-1} so that the Bragg angles to be resolved are only a few mrad requiring diaphragm sizes between 50 and 10 μm . This corresponds to a transverse momentum spread $\delta k_{\perp} = k\delta\theta$ and a transverse beam coherence of δy or $\delta z \simeq \delta k_{\perp}^{-1}$ defined by the Heisenberg uncertainty relation which is usually limited below 10-20 \AA^{-1} . However, due to the grazing incidence angle $\theta_i \approx 1^\circ$ used in GIFAD, the length coherently illuminated along the x direction is $\theta_i^{-1} \sim 100$ times larger. Elastic diffraction spots having the same size as the primary beam, as visible in Fig. 4a), can only be observed if the surface illuminated coherently by the beam is itself coherent (perfectly periodic) which is rather demanding in terms of surface preparation. Fortunately, LiF can be easily cleaved with large terraces if specific color centers are produced in the bulk by controlled exposure to neutrons or gamma rays²³. Gentle annealing is enough to eliminate the color centers in UHV. Grazing incidence techniques, in general, are quite sensitive to the presence of defects. This sensitivity is heightened with non-penetrating techniques such as GIFAD because a single ad-atom or molecule encountered along the comparatively long projectile trajectory is enough to perturb the scattering. The possibility of inducing thermal desorption of impurities is therefore important.

3.2 Imaging detector.

Several position-sensitive detectors are available²⁴ to detect helium atoms around 1 keV, all based on micro-channel plates. For diffraction experiments and easy data analysis, the most important properties are a good uniformity as well as an excellent integral and differential linearity (constant pixel size). A single, large-diameter micro-channel plate together with a phosphor screen filmed by a scientific-grade camera demonstrated good results with a ≈ 70 mm active diameter on a 100 mm flange²⁵.

4 Brief theoretical aspects

So far, atomic elastic diffraction on a surface has been treated as the quantum dynamics of the projectile atom evolving in the potential energy landscape (PEL) $V_{3D}(x, y, z)$ created by all surface atoms at equilibrium positions. In other words, quantum dynamics is the tool linking the unknown PEL to the measured diffracted intensities. Assuming that the quantum scattering can be treated exactly, both theory and experiment are trying to measure the PEL

as accurately as possible and to provide physical parameters such as atomic positions or the attractive physisorption depth.

4.1 Potential energy landscape (PEL).

Potential energy landscape is the potential energy $V_{3D}(\vec{r})$ of a target atom placed at a location \vec{r} of coordinates (x, y, z) above the surface. We define here two associated interaction potentials useful to understand the dynamics and describe GIFAD results. These are the mean axial and mean planar interaction potential $V_{2D}(y, z)$ and $V_{1D}(z)$ respectively,

$$V_{2D}(y, z) = \int_{a_x} V_{3D}(\vec{r}) dx ; V_{1D}(z) = \int_S V_{3D}(\vec{r}) dx dy \quad (1)$$

where a_x is a periodic length along the low index x direction and S is a unit cell. Within the ASCA, GIFAD probes the $V_{2D}(y, z)$ interaction potential. Such an axial potential is represented in Fig. 2b) where the large circles labeled F and Li correspond to rows of fluorine and lithium atoms. In principle, several low index directions should be probed to better describe the full 3D PEL without ambiguity. $V_{1D}(z)$ is useful to describe general properties such as the mean trajectory length L or the mean interaction time τ , as well as the mean attractive well depth D .

4.2 PEL development over effective binary potentials.

Describing the PEL over the full 3D volume above a unit cell is quite demanding and a useful data reduction procedure consists of adjusting effective binary interaction potential attached to the surface atoms so that their summed contributions reproduce the calculated or inferred PEL: $V_{3D}(x, y, z) = \sum_{i,j,k} V_k(r_{ijk})$ where k is an index for each surface atom in the lattice unit and i, j are the lattice indexes. With this approach, a reduced set of k binary interaction potentials is needed together with the equilibrium positions x_k, y_k, z_k of the k atoms of the surface lattice cell. For LiF, these are the location of the Li and Fluorine ions as well as the few parameters describing the associated attractive and repulsive forces. This compact description is well suited to consider thermal displacements or fitting procedures between theory and experiment.

4.3 Quantum scattering models.

The quantum dynamics of a helium atom having keV or tens of meV energies is not fundamentally different except that since the wavelength is a hundred times smaller the typical unit volume needed to describe an oscillation of the wave-function should be typically 10^6 times smaller and the time step also should be adapted to smaller values. Using the ASCA, only the slow motion in the (y, z) plane has to be considered and wave packet propagation^{10,26-28} or close coupling approaches^{29,30} have been successfully applied. The accuracy of the axial channeling approximation was investigated in detail by extensive 3D calculations compared with the simplified 2D ASCA^{16,31,32}. All methods developed for TEAS such as Bohmian trajectories^{33,34} or multichannel quantum defect theory^{32,35} should also apply to GIFAD withing the ASCA. The close-coupling approach has shown to be particularly efficient³¹ even in a situation where almost a hundred diffraction

channels are open²⁹. We consider that quantum scattering can be solved and that a given PEL can therefore be compared with diffraction images.

4.4 Simplified scattering models.

It is often desirable to have approximate methods allowing a qualitative understanding of the measured diffraction images and classical, semi-classical, and optical methods have provided valuable results. H. Winter and A. Schüller^{19,36} as well as M.S. Gravielle and J.E. Miraglia^{37,38} have developed a semi-classical approach. Using an eikonal approximation of small-angle scattering, they could treat the entire 3D scattering problem and verify the accuracy of the ASCA with classical trajectories, as pioneered by Danailov and Karpuzov³⁹. M.S. Gravielle and J.E. Miraglia have also eliminated the so-called "rainbow divergence" appearing at the maximum lateral deflection angle⁴⁰ and demonstrated the sensitivity of the approach to the potential energy landscape⁴¹. The simplest approach is however the hard corrugated wall model (HCW) (see *e.g.* ref.⁴²). Assuming that the repulsive wall is quite abrupt, for instance, exponential, most of the deflection takes place in the vicinity of the turning point of the classical trajectory as illustrated in fig.2b). The HCW model considers a point-like specular deflection at the iso-energy potential surface $\tilde{z}(y)$ defined by $V(y, \tilde{z}(y)) = E_{\perp}$ and straight-line trajectories otherwise, for both the way in and out. The $\tilde{z}(y)$ surface acts as a corrugated mirror for the equivalent optical problem. This $\tilde{z}(y)$ function is also called the corrugation function and the difference between its maximum and minimum values is the full corrugation amplitude of z_c . Within the HCW model, the probabilities of the diffraction into the j -th diffraction channel are calculated from⁴²⁻⁴⁵

$$P_j = \frac{k_{fz}}{k_{iz}} \left| \frac{1}{a_y} \int_0^{a_y} e^{-ijG_y y - i(k_{fz} + k_{iz})\tilde{z}(y)} dy \right|^2. \quad (2)$$

The HCW model has been mainly used to link diffracted intensity to a particular corrugation function $\tilde{z}(y)$. The HCW is not fully quantitative but it is enough to track simple tendencies, for instance, that z_c decreases with energy relative to the LiF $\langle 110 \rangle$ direction while it increases along $\langle 100 \rangle$ ⁴⁶ or to identify the charge transfer between Zn and Se atoms at the surface⁴⁷.

5 Experimental elastic and inelastic diffraction.

Both fig.1 and fig.4 display tiny spots visible on the diffraction images together with longer vertical stripes extending mainly along the polar angle θ_f (z direction). We assume here that these spots correspond to elastic diffraction where no energy is exchanged with the surface while the inelastic component is attributed here to the excitation of phonons because the large band-gap efficiently prevents electronic excitation in the range of energy and incidence angle^{48,49}.

Just like any other diffraction technique, the elastic diffraction intensity reveals the periodic component of the system investigated while the inelastic one contains information on the momentum transfer associated with the processes leading to energy exchange. In other words, the elastic diffraction indicates the

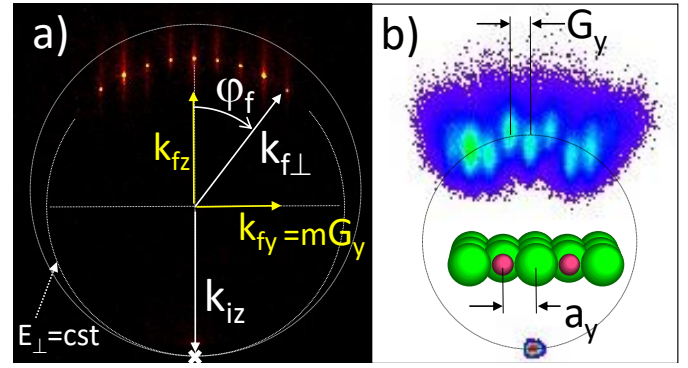


Fig. 4 a) Diffraction pattern of 300 eV He on LiF along the $\langle 110 \rangle$ direction at $\theta_f = 1.08^\circ$ ($k_{\perp} = 14.3 \text{ \AA}^{-1}$). The white cross indicates the location of the beam before target insertion. The line between this cross and the specular spot indicates the scattering plane $k_{fy} = k_{iy}$. The elastic diffraction spots fall on the Laue circle of energy conservation: $k_{fy}^2 + k_{fz}^2 = k_{iz}^2$ and exhibit diffraction along the y direction only $k_{fy} = mG_y$. b) early diffraction image from ref.⁵⁰ where elastic diffraction was not visible.

location of the equilibrium positions, that of the center of the surface atom vibrational wave function irrespective of its thermal motion while the inelastic component will be sensitive to the imperfections such as the motion associated with this vibrational movement as probed by the projectile atom.

Compared with photons or electrons, atoms have a large mass

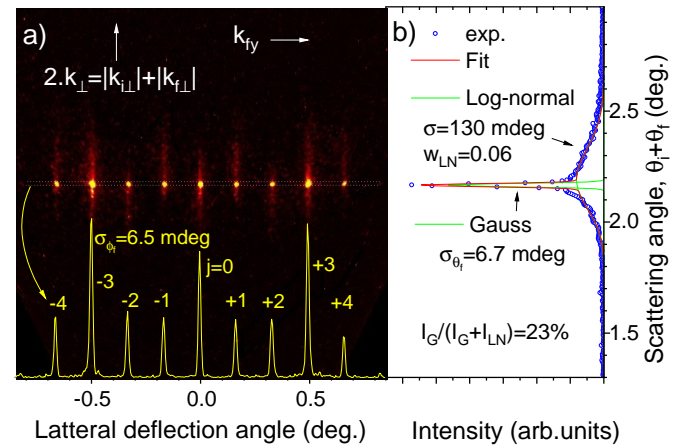


Fig. 5 Same as fig.4a) but the vertical axis is now $k_{\perp eff} = |k_{\perp}| + |k_{f\perp}|$ corresponding to the diameter of the dotted circle going through the beam in fig.4a). The Laue circle is now a horizontal line and the intensity on a 6 mdeg wide horizontal band is reported in yellow at the bottom. b) The projection of a) on the vertical axis gives the polar scattering distribution $P(\theta_f)$. It indicates two components, a narrow quasi-gaussian and a much broader one having a quasi-log-normal profile.

so that the recoil momentum transferred classically to the target atoms cannot be neglected. For an isolated target atom interacting with the helium projectile, momentum conservation implies that any projectile deflection is associated with the opposite recoil momentum of the target atom. In a solid or on a surface, the target atoms are not free to move but strongly coupled to their neighbors and this can be modeled by a Debye or Einstein model where each atom is in a harmonic oscillator where a mo-

mentum change by δk may or may not lead to a vibrational excitation. The oscillator is usually described by its quantized excitation energy $\hbar\omega_D \equiv k_B T_D$ where ω_D is the Debye frequency, k_B is the Boltzmann constant and T_D the Debye temperature. For an atom in the vibrational ground state ($T \ll T_D$), this probability is

$$p_e = e^{-E_r/\hbar\omega_D} \quad (3)$$

known as the Mössbauer-Lamb-Dicke probability or probability for recoil-less emission in photon spectroscopy. This defines the Lamb-Dicke regime needed to cool down atoms down to nano kelvin in optical lattices. Two different classical points of views can be adopted, one is to say that target atoms are displaced along z by σ_z and that this induces a path difference of $2\sigma_z$ giving a phase difference $\delta phase = 2\vec{d}\vec{k}\vec{\sigma}_z \approx 2dk_z\sigma_z$, with $\vec{d}\vec{k} = (\vec{k}_f - \vec{k}_i)$ so that the intensity scattered at Bragg angle will be affected by an amount $e^{-\delta phase^2}$ leading to the well known Debye-Waller factor. For a temperature T , the thermal Gaussian distribution of z is.

$$\sigma_z^2 = \langle z^2 \rangle = \frac{3\hbar}{2m\omega} \coth\left(\frac{T_D}{2T}\right) = \frac{3\hbar^2}{2mk_B T_D} \coth\left(\frac{T_D}{2T}\right) \quad (4)$$

where the absence of top neighbor for a surface atom is taken into account^{18,51}.

The other approach considers the momentum dk transferred to a surface atom and evaluates, with quantum mechanic, the probability that this transfer can induce a vibration excitation. Classically, these separate views would be considered as different contributions but in quantum mechanics, momentum or spatial approaches are simply different ways to evaluate the same probability. At this point, we simply consider that the elastically diffracted intensity reveals the equilibrium position of the surface target atoms, just as any elastic diffraction technique.

The simplest transform extracting the elastic and inelastic contribution would consider the center of the Laue circle as an invariant but we will see in section 7 that this is not well-adapted to inelastic scattering intensity located outside the Laue circle. Instead, it was shown^{52,53} that using the location of the direct beam (k_{iy}, k_{iz}) measured before target insertion as a reference point is crucial to define properly the scattering angle from the point of view of atomic collision (see section 7).

The transformation is the following, first, the detector coordinates are slightly rotated so that the specular plane representing the z axis is perfectly vertical. Then, for any coordinates (k_{fy}, k_{fz}) on the detector one calculates the value of k_{\perp} as the radius of the circle passing by itself and by the direct beam with a center located on the $k_{iy} = 0$ vertical line.

On the Laue circle of energy conservation, k_{\perp} is both the initial and the final value: $k_{\perp}^2 = k_{iy}^2 + k_{iz}^2 = k_{fy}^2 + k_{fz}^2$ but outside, the value defined above corresponds to a half sum of $k_{i\perp} + k_{f\perp}$.

To illustrate the transformation, we first consider the ideal situation displayed in fig.4a) where the beam is well-aligned with the low index direction $k_{iy} = 0$ (or $\phi_i = 0$) so that the energy of the motion in the perpendicular plane is simply $E_{\perp} = E \sin^2 \theta$. Fig.5a) reports the same data after the transform, the elastic diffraction spots are aligned horizontally. When projected onto the vertical direction corresponding now to k_{\perp} , the polar scattering profile al-

lows an easy determination of the relative, elastic, and inelastic intensities. The polar distribution is fitted by two components, a narrow Gaussian with $\sigma = 6.7$ mdeg and a broader log-normal profile (eq.5) with a standard deviation twenty times larger $\sigma = 130$ mdeg corresponding to a log-normal dimensionless relative width $\sigma\theta_s/\theta_s$, of 0.06 where θ_s is the full specular scattering angle $\theta_i + \theta_f$. For reasonably narrow distributions the relative width parameter w is close to the standard deviation divided by the median value $w = \sigma_{\theta}/\theta_s$.

$$LN[\theta_s; w](\theta) = \frac{A}{\sqrt{2\pi w\theta}} \exp\left(-\frac{(\ln \frac{\theta}{\theta_s})^2}{2w^2}\right) \quad (5)$$

The intensity of the Gaussian component represent 23% of all the intensity and should correspond to the absolute Debye-Waller probability. We use the width of the Gaussian distribution to define the size of the narrow band to isolate the elastic profile displayed in yellow in bottom of fig.5a). The profile can be analyzed by standard software to extract the diffraction intensities. A fit imposing equal line shape and equal spacing of the peaks provides a good accuracy on the measured intensities and line shapes. In the present case, the fig shows a quasi Gaussian line-width with $\sigma = 6.5$ mdeg comparable to the size of the primary beam *i.e.* to the angular resolution convoluted by the spatial resolution of the detector suggesting that the elastic spot size is not limited by the surface coherence length.

6 Elastic diffraction.

Fig.5b) shows that even at the specular angle a significant fraction of the intensity can be present. In this example corresponding to $E_{\perp} = 106$ meV, the inelastic intensity corresponds to 77% of all the scattered intensity and around 20% of the intensity on the Laue circle. *Stricto sensu* one should subtract this underlying 20% inelastic intensity and this can be achieved easily with well-tuned doubly differential filters such as the "Mexican hat"⁵³. However, in the regime where both elastic and inelastic diffraction are observed, their intensity distributions on the Laue circle were found to be equal¹⁸. A tentative explanation of this observation is given in section 7 describing our inelastic scattering model. In the present section, for simplicity, We will not discuss inelastic diffraction and we consider that the diffracted intensities correspond to the elastic component. We now examine the different types of measurements obtained by varying the beam parameters.

6.1 Polar angle scan (θ -scan).

A polar scan is a step-wise variation of the angle of incidence θ_i at fixed energy and azimuthal angle ϕ_i . For simplicity, we consider here that this azimuthal angle ϕ_i is zero so that the projection of the beam direction on the surface coincides with a low axis direction of the crystal as in fig.4. As the projectile energy and the angle of incidence ($\theta \ll 1$) the Bragg angle $\phi_B = \arctan(G_y/k \cdot \cos \theta_i)$ is constant with a few 10^{-4} relative variation. A visual and compact way to display the evolution of the diffracted intensities in this polar θ -scan is to plot the intensities on the Laue circle in a 2D color map. Since the diameter of the Laue circle is here pro-

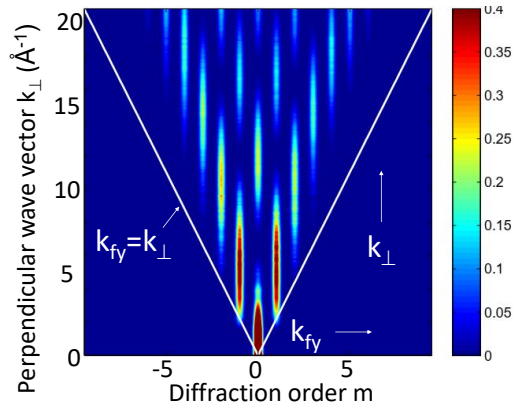


Fig. 6 Diffraction chart corresponding to a θ scan recorded with 300 eV ^4He along the $\langle 110 \rangle$ direction. The Bragg angle is fixed and the angle of incidence θ_i determines the value of k_{\perp} . 20 \AA^{-1} corresponds to $E_{\perp} = 208 \text{ meV}$ while the lowest values correspond to 2 meV .

portional to the angle of incidence, the length of the half-circle above the surface grows linearly while the incidence angle producing an overall V shape. In fig.6, the white line indicates the maximum value of the lateral deflection expressed in diffraction order. This corresponds to a projectile atom scattered parallel to the surface, with $\phi_f = \theta_i$, and therefore $\theta_f = 0$ i.e. with all its momentum k_{iz} converted in the in-plane movement along the y direction. Like many diffraction charts, the one plotted in fig.6 is highly redundant, but this is also a warrant of high reliability. This can be qualitatively understood from the hard corrugated wall model. Within the HCW, each line in fig.6 corresponds to one iso-energy corrugation function and since the shape of these curves evolves only slowly (see e.g. fig.14b), changing the value of E_{\perp} can be viewed as illuminating a given grating with different values of λ_{\perp} . The most simple case would correspond to a cosine corrugation function $\tilde{z}(y) = z_c/2 \cos(2\pi y/a_y)$ where z_c is the full corrugation amplitude and a_y the lattice parameter associated with periodic row structure of the averaged on the low index x direction. In this case, Eq. 2 indicates that the diffracted intensities are given by $I_j = J_j^2(2\pi z_c/\lambda_{\perp})$ where J_j is the Bessel function of rank j . The advantage of this corrugation function is that the whole diffraction chart can be modeled with only one parameter z_c . Each diffraction order relative intensity I_j can be seen as an independent measurement of the path difference of the two trajectories leading to a lateral deflection by $j \times \phi_B$. The diffraction chart nicely shows the progressive broadening of the diffraction peaks with the energy E_{\perp} but we now know that the width of the elastic peak should remain constant and close to the direct beam profile so that the broadening observed in earlier experiment was due to an inelastic effects.

6.2 Energy scan (E-scan).

An energy scan is a step-wise variation of the primary beam energy E while keeping the angles of incidence θ_i and azimuthal ϕ_i fixed. During an E-scan, the radius of the Laue circle is constant but the Bragg angle $\phi_B = \arctan(G_y/k \cdot \cos \theta_i)$ gets lower as the energy increases so that more diffraction orders can be observed until they can not be resolved experimentally. Fig.8 displays an E-

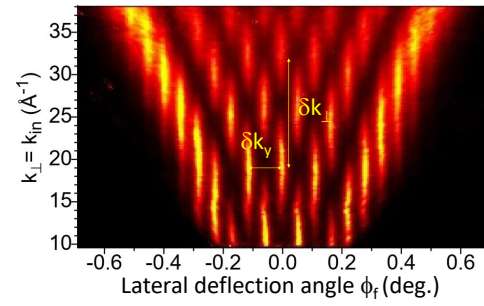


Fig. 7 diffraction charts of 500 eV neon on LiF $\langle 110 \rangle$ recorded at incidence angles θ_i between 0.25 and 1 deg. corresponding to $E_{\perp} = 9.5$ and 76 meV respectively. In the paraxial region, a pseudo period is observed $\delta k_{\perp} \sim 13.3 \text{ \AA}^{-1}$ and $\delta k_y \sim 2G_y$.

scan of neon atoms on a LiF surface along the $\langle 110 \rangle$ direction. The locations of the Bragg peaks are indicated by dotted white lines.

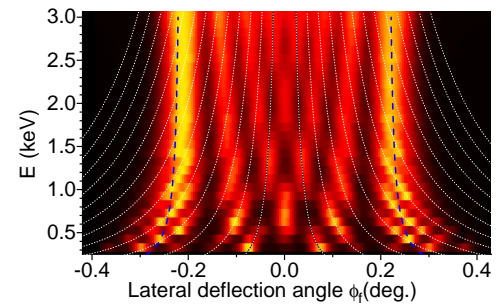


Fig. 8 diffraction charts of neon on LiF $\langle 110 \rangle$ recorded at fixed incidence of 0.42 deg and at incident energies between 300 eV and 3 keV. The energy $E_{\perp} = E_0 \times 5.37 \cdot 10^{-5}$.

One interesting aspect of the E-scan is that the part of the crystal surface illuminated remains constant allowing more reliable identification of small variation of the diffracted intensities. On a microscopic level, the length of the trajectory is also expected to remain constant as long as attractive forces can be neglected and this offers an interesting opportunity for the investigation of inelastic processes. The number of binary collisions should remain constant as well as the associated deflection angle.

6.3 λ diffraction chart

An alternate presentation of the data was proposed by Winter and Schueller¹⁹, it consists in plotting another 2D color map but as a function of the projectile wavelength λ_{\perp} irrespective of the way the data have been collected. This would be interesting mainly for data collected in the elastic regime because the ASCA tell us that the only relevant parameters are the one associated with the motion in the plane perpendicular to the low index direction. This can be the energy E_{\perp} , the wave-number k_{\perp} or the wavelength λ_{\perp} . However, the diffraction charts also display the measured width of the diffraction peaks and this was shown to be associated mainly to limited surface coherence and to inelastic processes and these depend also on the actual projectile energy (see section 7). In practise it does not really allow to combined very different

experimental condition but certainly offer a nice presentation of the nodal structures. An example is reported on fig.14a).

All the diffraction charts presented provide clear 2D picture having their own interest. These can illustrate specific features such as a slow variation of the corrugation amplitude seen as a variation of the oscillation rate of the specular intensity with k_{\perp} or a variation of the rainbow deflection angle at high or low velocity. Perhaps the most important prediction of the ASCA, is that the elastic diffracted intensities should depend only on the energy E_{\perp} irrespective of the values of E and incidence angle θ_i . Fig.21 shows that indeed the red circles and blue triangles corresponding to the θ -scan and the E-scan in fig.7 and fig.8 respectively fall on top of each other.

6.4 Azimuth scan (ϕ -scan).

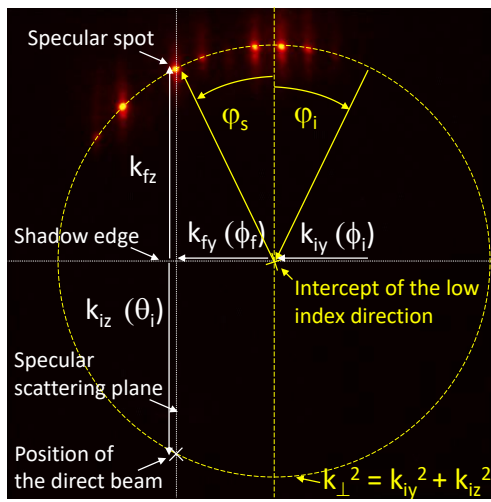


Fig. 9 Diffraction pattern of 460 eV He impinging along LiF $\langle 110 \rangle$ with $\theta_i = 0.9$ deg. and an azimuth angle $\phi_i = 0.45^\circ$. The white dotted lines correspond to the intercept with macroscopic planes ; the horizontal shadow edge of the surface plane and its perpendicular the specular scattering plane which contains both the incident and specularly reflected projectiles. It corresponds to the impact of an effective particle with a momentum $\hbar k_{\perp} = \hbar \sqrt{k_{iy}^2 + k_{iz}^2}$ with an incidence angle $\phi_i = \arctan(k_{iy}/k_{iz}) \approx 26^\circ$ with respect to the surface normal.

Fig.9 displays a typical diffraction image recorded when the azimuthal direction does not coincide with a low axis direction of the crystal. It corresponds to He atoms with an energy of 460 eV and an incidence angle $\theta_i = 0.9$ deg., close to the conditions of fig.4a) but with an azimuth angle $\phi_i = 0.45^\circ$. The elastic diffraction spot are still aligned on a circle passing by the primary beam position and the specular spot but they define a Laue circle with a center shifted by 0.45° corresponding to the intercept with the low index direction. The projection of the scattering plane on the detector, defined by the line connecting the specular spot to the location of the direct beam before target insertion is not anymore a diameter of the Laue circle which is now $2k_{\perp} = 2\sqrt{k_{iy}^2 + k_{iz}^2}$. This radius $2k_{\perp}$ corresponds to the angle between the beam direction and the crystal surface low index direction. In the (y,z) plane this angle corresponds to a particle which is no longer perpendicular to the surface but which impact

the atomic rows of the $V_{2D}(y,z)$ mean potential with an angle of incidence $\phi_i = \arctan(k_{iy}/k_{iz}) \approx \arctan(\phi_i/\theta_i)$. This experiment evidence is another clear illustration of the ASCA.

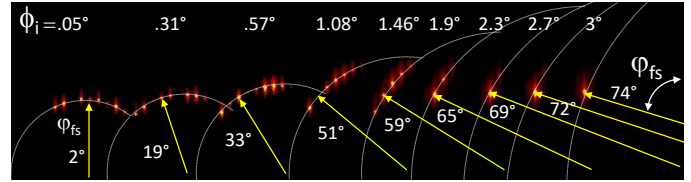


Fig. 10 Juxtaposition of nine diffraction patterns of 460 eV He corresponding to an azimuthal angle ϕ of 0.05, 0.3, 0.57, 1.1, 1.9, 2.3, 2.7 and 3 deg. relative to the $\langle 110 \rangle$ direction. This ϕ scan is also a k_{iy} scan and corresponds to a variation of the effective angle of incidence $\phi_i = \arctan(k_{iy}/k_{iz})$, but associated with an increase of the effective energy E_{\perp} due to the increase of k_{\perp} illustrated by the Laue circle. The yellow arrows indicate the specular spot with $k_{fz} = -k_{iz}$, $k_{fy} = k_{iy}$. During experiment, the location of the specular spot on the detector stays fixed.

The azimuthal scan correspond to a variation of the surface azimuthal angle ϕ_i . During an azimuthal scan several low index direction can be identified providing the basis of atomic triangulation measurements^{54,55} detailed in the next section. We focus here on the behavior of the diffraction pattern close to a given low index direction. As detailed above, according to the ASCA, an azimuthal scan corresponds to a variation of the incidence angle ϕ_i in the (y,z) plane. This is illustrated in fig.10 where the successive Laue circles indicate the progressive evolution to grazing scattering angles in the (y,z) plane. It also shows that the effective energy associated with the motion in the (y,z) plane represented by yellow arrows linking the center of the Laue circle to the specular spot increases with the azimuthal angle ϕ_i . When the incidence angle ϕ_i in the perpendicular plane becomes itself grazing, the diffracted intensities gradually reduce to a single specular peak. This is the GIFAD equivalent of the situation where a pronounced reduction of the in-plane diffraction was observed by Farías *et al.*²¹ in TEAS when the angle of incidence with the surface plane reached ≈ 20 deg. This situations has been well re-

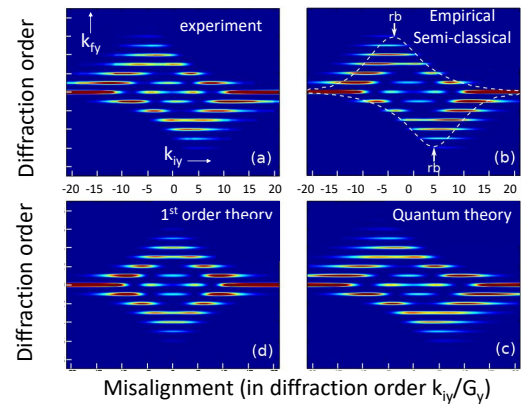


Fig. 11 a) The evolution of the diffracted intensities of 460 eV He impinging at $\theta_i = 0.93^\circ$ is reported as a function of the azimuthal angle ϕ relative to the $\langle 110 \rangle$ direction expressed in diffraction order $G_y = 0.133$ deg. b), c) and d) are for calculations from ref. ^{17,20,56} respectively.

produced both by quantum scattering calculation¹⁷ and by semi

classical trajectory calculation⁵⁷. Ruiz *et al.* also proposed a description in terms of quasi-resonance that should apply to most GIFAD situations⁵⁸. Due to the very small value of the Bragg angle in GIFAD, it can be difficult to reach the perfectly aligned condition $\phi_i \ll \phi_B$. From the experimental point of view, it is important to try to estimate first-order corrections to better compare image recorded with small values of the azimuthal angle $\phi_i \simeq \phi_B$. The HCW formula in eq.2 does not allow $k_{iy} \neq 0$, whereas simple 1st order perturbation theory by Henkel *et al.* was found to account for the progressive attenuation of the non-specular diffraction at grazing incidence ϕ_i in the (y, z) plane (large values of ϕ_i) but does not fulfill time reversal symmetry. This was empirically and qualitatively corrected by Debiossac *et al.*⁵⁶ as illustrated in fig.11c). A second-order perturbation theory was proposed by Pollak and Miret-Artés⁵⁹ that should allow even better corrections. During an azimuthal scan, the angle of incidence θ_i should remain fixed. Therefore the direction normal to the surface should coincide with the azimuthal rotation axis with a high accuracy which is particularly demanding in a backable UHV system compatible with transfer systems. In practice, recording the specular spot location during a complete 360° azimuthal scan allows a precise measurement of the residual tilt angle ϑ in the form of a cosine oscillation $\theta_s \simeq \theta_0 + \vartheta \cos \phi - \phi_0$ with a defect ϑ usually of a few tenths of a degree. Measuring the azimuth ϕ_0 for which the observed specular scattering is maximum allows an easy correction by adjusting a cosine compensation of the polar angle during an azimuthal scan to provide a stable incidence angle θ_i during rotation⁶⁰. Note that the presence of a sharp elastic diffraction spot is also very useful to identify the presence of various surface mosaicity that can arise from thermal or mechanical shock⁶¹.

6.4.1 ϕ -scan and atomic triangulation.

The azimuthal scan is usually the first experiment performed after the insertion of a new sample under UHV. This allows precise measurement of the main crystallographic directions difficult to track precisely in a transfer system. Most often, diffraction is not observed immediately because of molecular adsorption or any other reason such as limited coherence. We present here triangulation measurements offering a natural extension of the diffraction data presented above to the classical limit where diffraction does not exist. The word triangulation was introduced by Pfandzelter *et al.*⁶² to designate an azimuthal scan during which the secondary electron emission following the impact of 25 keV H^+ ions at grazing incidence was recorded. Maxima in the electron yield were assigned to low index directions of the topmost layer because, with $E_{\perp} \approx 30$ eV, the trajectories deep inside the valleys are significantly longer. By following the new direction appearing after exposure to metallic vapor, they could identify crystallographic properties of freshly grown over-layers. With atoms having an energy E_{\perp} below one eV, electron emission is not present and another contrast mechanism has been proposed adapted to GIFAD environment equipped with a position-sensitive detector recording the scattering profile^{54,55}. Feiten *et al.* monitored the maximum intensity during the azimuthal scan observing dips associated with the low index directions while Kalashnyk *et*

*al.*⁵⁵ directly monitored the width and mean position of scattering profile observing peaks of maximum width associated with low index directions together with a characteristic swing of the mean scattering plane. Both observation were recorded in conditions where diffraction was not or hardly observed and we plot below similar measurement performed with 460 eV helium¹⁷.

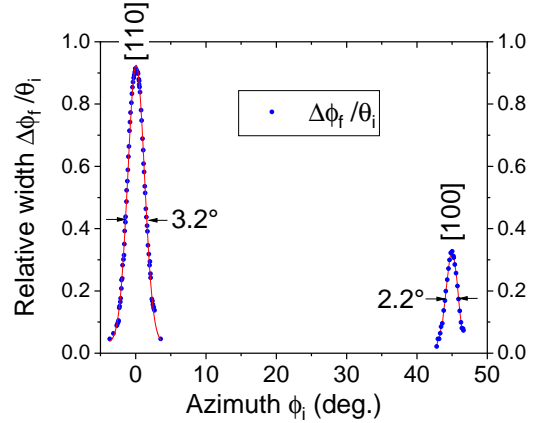


Fig. 12 For 460 eV helium on a LiF(001) surface¹⁷, the azimuthal scattering width $\Delta\phi_f$ (fwhm) divided by the incidence angle θ_i is reported as a function of the incidence azimuthal angle ϕ_i revealing the low index direction $\langle 110 \rangle$ and $\langle 100 \rangle$. The image in fig 4a) corresponds to the top of the left peak and $\Delta\phi_f$ is the width along k_y divided by the radius of the Laue circle. Both the amplitude and width of the peaks are expected to be hardly modified whether diffraction is observed or not.

Fig.12 reports the evolution of the mean scattering width $\Delta\phi_f$ evaluated automatically from the diffraction image as the standard deviation σ_{ϕ_f} times 2.35, a number that would coincide with the full width at half maximum (fwhm) for a Gaussian distribution. To propose a number that does not depend rapidly on the angle of incidence θ_i we used the scattering width $\Delta\phi_f$ divided by the angle of incidence θ_i . The two low index directions $\langle 110 \rangle$ and $\langle 100 \rangle$ appear with a very large contrast and their evaluation could be compared with quantum or classical calculation on a quantitative basis. The data around $\phi_i = 0$ correspond to the values plotted in fig.11. Here again, the width is strongly related with the rainbow scattering angle $\Delta\phi_f \sim 2\phi_r$ but it is defined without ambiguity through the variance of the measured scattering intensities. Such evaluation remain meaningful even when a limited number of diffraction orders is observed, as in fig.4a) making the rainbow angle impossible to measure accurately. The position, the amplitude, and the width of these triangulation peaks are not expected to change significantly with the beam energy or with the surface quality. Only the base level, recorded for ϕ_i value away from low index direction, is expected to rise rapidly from a value close to the width of the primary beam when diffraction conditions are good as in fig.10 up to much larger values when the surface has a limited coherence length and/or is partly covered by adsorbates.

In addition to the increased scattering width when the azimuthal angle ϕ_i approaches a low index direction, a guiding effect was demonstrated¹⁷. Using the same data, the fig.13 shows three different presentations of the mean vertical and lateral

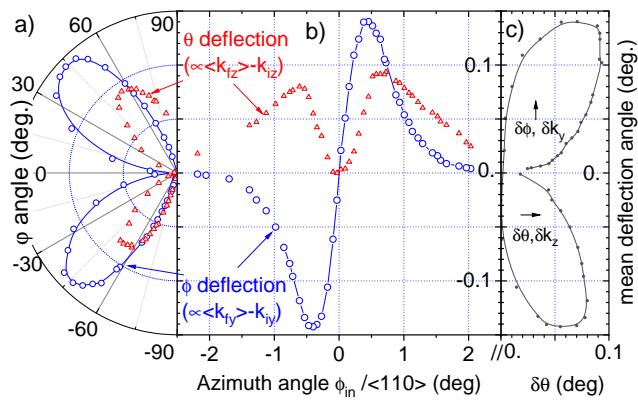


Fig. 13 For 460 eV helium on a LiF(001) surface at an incidence angle $\theta=0.48^\circ$ and variable azimuth ϕ_m , the mean lateral (ϕ_f) and vertical (θ_f) deflection referred to the specular beam are reported in three different forms. b) is a cartesian plot as a function of the azimuth angle ϕ_m referred to $\langle 110 \rangle$ direction¹⁷. a) is a polar plot as a function of the angle $\phi_i = \arctan(k_{iy}/k_{iz})$. c) describes the trajectory of the mean impact position ($(\langle \theta_f \rangle, \langle \phi_f \rangle)$) as the incident azimuth ϕ_i is varied.

deflection taking place during an azimuthal scan when passing through a low index direction. The oscillation of the mean lateral deflection ($\langle \phi_f \rangle$) was observed even without diffraction in ref.⁵⁵ but, in the inelastic regime, the weaker vertical deflection (along θ_f) is easily blurred by the broad log-normal inelastic polar scattering profile.

6.5 high perpendicular energies, the topology

For large values of the energy E_\perp of the motion in the perpendicular plane, the associated wavelength becomes comparatively small allowing a good spatial accuracy. This is true in microscopy and applies also to diffraction. Let us assume that the HCW model is exact and that one tries to measure the shape of the corrugation function $\tilde{z}(y)$ over the projected lattice cell a_y , for a given value of E_\perp . Due to the interferometric nature of diffraction, a resolution between $\lambda/10$ and $\lambda/100$ can be expected in the z direction depending on the accuracy in measuring the diffracted intensities. Along the y direction the accuracy should scale with λ_\perp meaning that the shape $\tilde{z}(y)$ along y may have n significant points where n is the number of observed diffraction peaks. In addition, at large values of E_\perp the exact contribution of attractive forces, which are still difficult to evaluate qualitatively, are reduced allowing more confidence in the evaluation of the PEL. For LiF the projected lattice parameter a_y along the $\langle 110 \rangle$ and $\langle 100 \rangle$ directions are 2.2\AA and 3.12\AA respectively while a ^4He atom with 100 meV and 1 eV energy has a wavelength of 0.45\AA and 0.14\AA respectively. The obvious limitation being that the diffraction peaks should be well-resolved to allow a good measurement of their intensities.

The first quantitative attempt to go beyond simple corrugation measurements and to use GIFAD data for topology was proposed by Schueller *et al.*²⁷ using *ab initio* DFT calculation and a wave packet propagation technique. Comparing with diffraction data in fig.14a), they proposed a rumpling of the lithium ions by $0.05\pm 0.04\text{\AA}$ below the fluorine plane as well as an overall PEL displayed in fig.14b). A striking result is that the shape of the

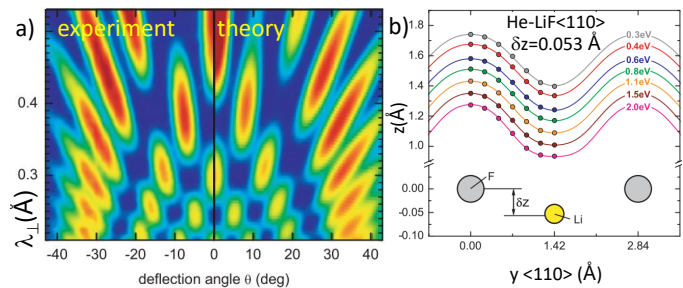


Fig. 14 a) Comparison of experimental and calculated diffraction chart of He on LiF along $\langle 110 \rangle$ as a function of the wavelength λ_\perp . b) The calculated potential energy landscape and, in the bottom the atomic positions highlighting the rumpling by 0.053\AA of the Li^+ ions.

corrugation function $\tilde{z}(y)$ associated with successive values of E_\perp appear very similar. This indicates that, in the optical limit and in this energy range, the diffraction data should correspond qualitatively to illuminating a grating of fixed shape $\tilde{z}(y)$ with a variable wavelength λ_\perp providing very high redundancy. In this case all the diffraction orders evolve smoothly and can be interpreted easily with optical models (see *e.g.* ref.^{29,63}). The central region of the diffraction chart where $\phi_f \sim 0$ corresponds to quasi specular reflection. *i.e.* to trajectories bouncing on a flat section of the PEL either on top or in the bottom of the valleys. In this paraxial region the pseudo period $\delta k_\perp \sim 13.3\text{\AA}^{-1}$ observed in fig.6 can be interpreted as due to a full corrugation amplitude $z_c = \pi/\delta k_\perp \sim 0.24\text{\AA}$ between the top and the bottom of the valley without any further assumption on the exact shape. Along the y direction, the pseudo period of two diffraction orders between successive maxima along the y direction in fig.6 indicates that the bottom sits in between the two top of the valleys. More complex shapes of the PEL could be measured²⁹ with this the optical model and with a surprising accuracy regarding its simplicity, probably because by measuring such pseudo periods one eliminates quantum corrections needed to predict the absolute locations of the maxima.

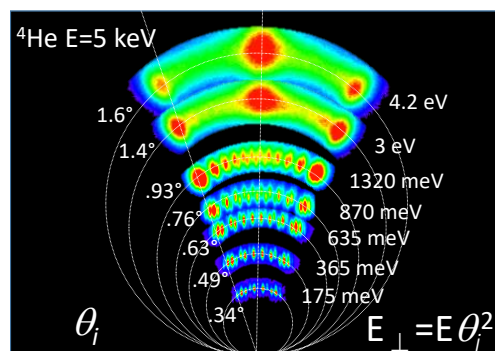


Fig. 15 Superposition of seven diffraction patterns measured with 5 keV ^4He projectiles on LiF aligned along the $\langle 110 \rangle$ direction. The angle of incidence θ_i is indicated on the left and the corresponding energy E_\perp on the right. All Bragg peaks can be resolved until $E_\perp = 1.5\text{ eV}$. Supernumerary rainbow structure (see text and fig.16) dominates above $E_\perp = 300\text{ meV}$. Above $E_\perp = 2\text{ eV}$ only the Supernumerary rainbow structure persists but slowly weakens. At 4.2 eV only broad intensity variation persist as detail in fig.17.

The progression of the scattering profiles with higher values of

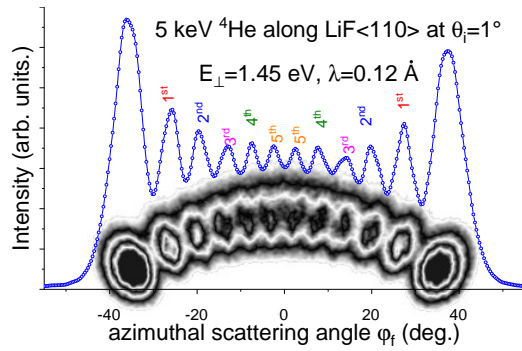


Fig. 16 The scattering profile of 5 keV ^4He atoms at $\theta_i=1^\circ$ incidence on LiF $\langle 110 \rangle$ expressed as a function of the angle ϕ_f in the perpendicular plane. Along with the external rainbow peaks at $\phi_r \simeq 36^\circ$, five additional supernumerary rainbow peaks are observed indicating a full corrugation amplitude of $z_c = 0.33 \text{ \AA}$ (see text). Close to the center, the inelastic Bragg peaks are still resolved showing a π shift from j to $j+1$ while the four diffraction orders at the outer rainbow angle ϕ_r are almost in phase but are still resolved in the natural diffraction coordinates ϕ_f .

E_\perp is reported in fig.15 using 5 keV ^4He projectiles. The inelastic diffraction dominates and clear diffraction peaks can be resolved up to $\approx 1.5 \text{ eV}$ but the scattering profiles are progressively structured in groups adopting a radial nodal structure identified as the supernumerary rainbows by analogy with the well known optical effect. These maxima (and minima) correspond here to successive phase match (and mismatch) between the two trajectories scattered at the same angle with the lattice cell. For the outermost rainbow, classical trajectories scattered at the maximum deflection angle originate from both sides of the inflection point of the corrugation function defined by $d^2z(y)/dy^2 = 0$ and can be chosen arbitrarily close to each other and are therefore always in quasi-phase match producing the genuine rainbow, ubiquitous in scattering theory.

As discussed above the path difference for the trajectories bouncing on top and bottom of the valley and contributing to specular region amount to $2 \times z_c$ so that the phase difference is expected to increase progressively from zero at the outer rainbow ϕ_r to $2k_\perp \times z_c$ in the center $\phi_f = 0$ or $\phi_f = \pi$. A maximum of the diffracted intensity will be observed each time that the phase difference is close to $2n\pi$. For a cosine corrugation function, the maxima would be those of the Bessel functions representing its Fourier transform. For a more complex shape of the corrugation function, the exact location of the maxima should allow accurate determination of the corrugation function⁶⁴. Fig.16 displays a scattering profile recorded with 1.45 eV with a black and white palette together with the intensity distribution reported corresponds to a radial projection where twelve maxima are observed with a minimum in the center. This tells us that the specular phase shift in the center should be around $5.5 \times 2\pi$ giving $z_c \approx 0.33 \text{ \AA}$, a fair estimate of the corrugation amplitude probed by a helium atom at this energy E_\perp . Independently, the location of the rainbow angle at $\phi_r = 36^\circ$ indicates that the inflection point (where $d^2z(y)/dy^2 = 0$) in the corrugation function corresponds to a derivative of $d\tilde{z}(y)/dy = \phi_r/2 = 18^\circ$. Note that the inner maxima corresponding to the fifth supernumerary rainbow are in fact

made of still resolved $j = \pm 1$ diffraction order while the dip in between corresponds to the quasi-extinct specular spot. This corresponds to the paraxial structure identified in previous section where the intensity switches from maximum to minimum for adjacent diffraction orders.

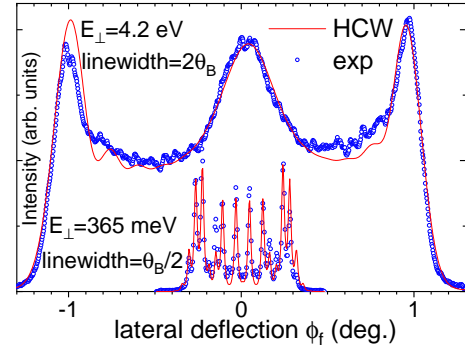


Fig. 17 The diffraction profiles for 5 keV helium along LiF $\langle 110 \rangle$ at $E_\perp = 4.2 \text{ eV}$ (top) and 365 meV (bottom) are fitted by the HCW formula of eq. 2. In both spectra, the Bragg angle is $\phi_B = 0.04 \text{ deg.}$ but the individual width of the diffraction peaks was chosen as 0.02 for the bottom spectrum and 0.08 for the top one. In this latter case, the rapid oscillation predicted by the HCW in the paraxial region is efficiently washed out.

At even higher values of E_\perp , fig.15 shows that the nodal structure also disappears progressively producing smooth scattering profiles. Similar profiles have been recorded at keV energies and were considered to be entirely classical however, most often classical trajectory simulations were struggling to reach a fair agreement. The unexpected surprise is that the optical HCW model does a good prediction in the whole range of E_\perp . Fig.17 displays the lateral deflection profile of the topmost pattern in fig.15 together with a fit by the HCW model using only two terms in the Fourier expansion of the corrugation function $\tilde{z}(y) = 0.15 \cos 2\pi y/a_y + 0.026 \cos 4\pi y/a_y$ in \AA . The outermost rainbow angle is well reproduced as well as the central broad maximum. This latter can only be present if the corrugation function is not a pure cosine. Qualitatively, it corresponds to a flattening of the bottom of the valley due to the presence of Li^+ ions visible on fig.14b). Compared with the electrons of the $2p^6 \text{ F}^-$ ions forming the valence band the density of the $1s^2$ electrons of Li^+ ions is much more compact and decays more rapidly. For these reasons, the Li^+ contribution can be almost neglected at a large distance but fig.17 clearly illustrates their contribution at a closer distance to the surface⁴⁶. The fig.17 also displays a comparable fit of a diffraction pattern recorded at lower angle of incidence θ ; *i.e.* having the same Bragg angle $\phi_B = 0.04^\circ$ (the peak separation). The main difference is that the width of the individual inelastic peaks is half a Bragg angle for the bottom one and twice the Bragg angle for the top one. In both cases, the HCW is fully coherent and predicts rapid intensity oscillations, for instance in the paraxial region, but, on top, these are efficiently washed out simply by considering a line profile $2\phi_B$ broad. The strong attenuation of oscillation amplitude of the supernumerary rainbows also surprisingly well reproduced, probably because their frequency also increases with E_\perp as illustrated by the variation between fig.16 and fig.17. The line-widths used in these fits are much larger

than the width of the primary beam indicating a dominant contribution from inelastic width discussed in section 7 and not to spot size effects as discussed by Frisco and Gravielle⁶⁵. However, from the HCW point of view, these contributions could have a similar effect on the visibility of the fringes.

6.6 low perpendicular energies, the attractive forces

At low values of E_{\perp} , the influence of the attractive forces, the Van der Waals, and, for ionic crystals, the polarisation forces become important. Since these usually have a longer range than the repulsive one, a small physisorption well with a depth D may be present. This physisorption depth plays a key role in the early stage of chemical reactions at surfaces but is difficult to measure or to calculate accurately. TEAS is a reference technique⁶⁶ with the ultimate resolution of spin-echo which allowed precise determination⁶⁷ of physisorption well by identifying resonance associated with the vibrational eigenstates of He in this shallow well. These resonances corresponding to a sharp modification of the diffracted intensities were first observed by Frisch and Stern⁶⁸ only a few years after de Broglie prediction that all particles should have an associated wavelength. Assuming that the attracted forces are directed along the z direction perpendicular to the surface, a minimum energy $E_z > D$ is required to escape from the surface attraction. A particle arriving from the vacuum necessarily has a kinetic energy larger than D above the well, but after deflection when impacting the surface the velocity component along z might not be enough to escape from the attraction to the surface. Classically, it bounces back on the surface and encounters as many additional impacts as needed until the energy along z is again larger than D . Quantum mechanics is significantly different, the allowed deflection is limited to multiple of the reciprocal lattice vector G_y and the allowed bouncing trajectories should correspond to bound states of the potential well explaining the resonant behavior. More important all different paths leading to a given diffraction order interfere, building a large amplitude resonance.

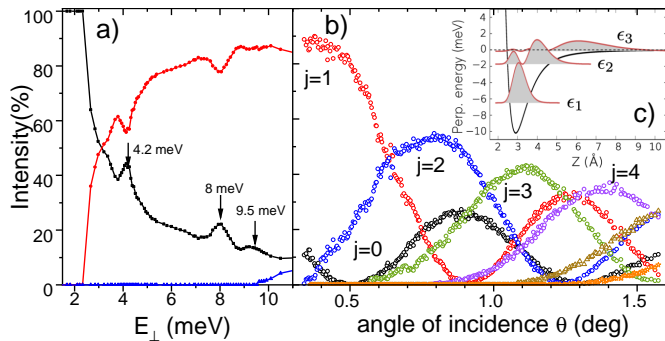


Fig. 18 For 300 eV ^4He on LiF along the $\langle 110 \rangle$ direction, the intensity of the diffraction orders j ($\pm j$ added) is reported for E_{\perp} between 2 meV and 233 meV. a) corresponds to the lowest values of E_{\perp} displaying bound state resonances while a smooth evolution is observed above in panel b). The insert is a schematic view of the mean interaction potential $V_{1D}(z)$ showing the bound states.

With GIFAD, only the three lowest resonance could be measured²⁸ and the fig.18 corresponding to the diffraction chart in

fig.6 show that the measured amplitude of the resonance is significantly reduced compared with the 100% theoretical predictions by wave packet propagation technique²⁸. This reduction was interpreted as due to the finite coherence length of the surface. The nature of the resonance on the average potential $V_{2D}(y, z)$ is similar to the one in TEAS and the time τ_b needed for a rebound is the same. However, compared with TEAS, the distance $x = v_x \tau_b$ spanned is longer by several orders of magnitude and any adatom or lack of periodicity will perturb the interference between direct and indirect scattering. The 0.5 meV width of the resonance may have different contributions but if associated with the coherence length the corresponding value would be $0.2 \mu\text{m}$ which is already quite a large value.

In conclusion, bound states resonances can also be measured with GIFAD but the large projectile velocity implies long distances between successive bumps so that a very long surface coherence is required. Even with several thousand Å coherence length only weakened resonance could be observed and only when two diffraction orders interfere producing, in theory, a 100% amplitude in the intensity associated with the resonance.

With neon atoms, the number of diffraction orders is much larger and so is the number of bound states⁶⁹ and the diffracted intensities plotted in fig.21 combining the data of the two diffraction chart in fig.8 and fig.7 do not reveal their presence. However, in the E_{\perp} scan plotted in fig.8, a clear increase of the maximum deflection angle ϕ_f is visible at the lowest values of E_{\perp} . The increase of the maximum deflection angle is often associated with an increase of the effective corrugation because for a simple shape of the corrugation potential the maximum slope is supposed to be proportional to the corrugation amplitude. Since lower values of E_{\perp} correspond to scattering taking place at a larger distance z to the surface plane, the corrugation is expected to decrease. This apparent contradiction was interpreted qualitatively as a refraction effect taking place on the way out. The projectile is acceler-

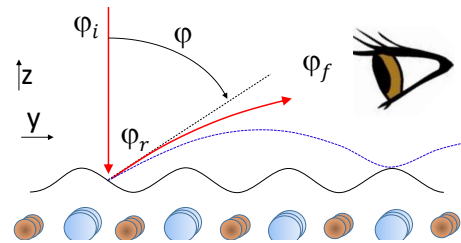


Fig. 19 Schematic view of the refraction effect. When the effective energy E_{\perp} is much larger than the well depth D , the exit angle ϕ_f is close to the rainbow angle ϕ_r . When $E_{\perp} \approx D$, the observed maximum deflection ϕ_f increases. For selected values of E_{\perp} , bound states can be temporarily populated.

ated toward the surface by an amount D so that the impact energy $E_{\perp} + D$ does not tend to zero revealing an quasi-constant distant of approach and a stable maximum deflection angle close to the rainbow angle ϕ_r . When leaving the repulsive region, the projectile enters the region where the attractive forces dominate and, assuming that these forces are directed towards the surface, the energy D has to be subtracted from the motion along z producing an increased deflection angle ϕ_f or ϕ_f as illustrated in fig.19

outlining the analogy with refraction in optics when light is emitted from a planar media with a refraction index larger than in the region of observation. The blue dashed line in fig.8 is a simple model using a value of $D = 10$ meV. A quantitative evaluation

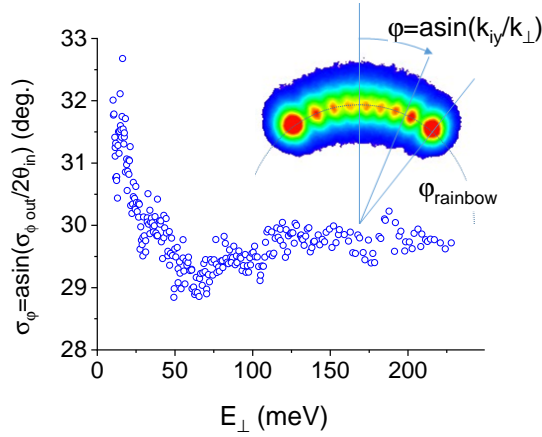


Fig. 20 For each value of k_{\perp} in fig.6 (also in fig.18b), the width σ_{ϕ} is defined as $(\sum_j P(j) \cdot j^2 \phi_B^2)^{1/2}$ where $P(j)$ is the probability distribution of diffraction order j and $\phi_B = \arcsin(G_y/k)$ is the Bragg angle. evolution of the mean exit angle σ_{ϕ} (see text) with the energy E_{\perp} . A clear increase below 50 meV is observed probably due to a refraction effect. The insert show a typical scattering profile in the semi classical regime, here $E=5$ keV, $E_{\perp}=0.8$ eV to define the angle ϕ and the particular value $\phi_{rainbow}$.

has been performed by a quantum diffraction code confirming the coarse estimation and this will be described in the next section. We try here to extend this type of measurement to helium where the increased deflection is not easily visible at low energies because only two or three diffraction orders are present (see fig.6 and fig.18a) preventing fair measurement of the rainbow scattering angle. The natural alternative allowing quantitative measurement is to use the standard deviation σ_{ϕ} or its equivalent value σ_{ϕ} (with $\phi_f = \arctan(\theta_f/\phi_f)$) which is independent from the projectile energy along x . Fig.20 reports the evolution of this mean standard scattering width with the energy E_{\perp} from the data of the θ scan plotted in fig.6. The values do not point exactly at the rainbow scattering angle but it is expected to scale in proportion. Fig.20 shows that σ_{ϕ} reaches a minimum around 50 meV before displaying a sharp increase at lower values of E_{\perp} . A comparable effect has been observed after scattering of helium on a graphene layer⁷⁰ but using a HCW fit of the data. The strategy proposed here is to present the data in a form that is model-independent and which displays a high contrast.

6.7 Quantitative comparison with quantum theory.

The comparison of experimental data to theoretical prediction can take different forms depending on which aspect is to be outlined. When the PEL is calculated without any adjustable parameter the accuracy might not allow a perfect overlap. In this case, qualitative features are searched to point as precisely as possible to the possible correction. This was the case with scattering on GaAs where the PEL obtained by *ab initio* density functional calculation showed a fair agreement only when the attractive forces were scaled down to produce a $D=8.7$ meV deep potential well

before being transformed into diffracted intensities by a quantum dynamics calculation. The comparison used to derive the rumpling of the Li^+ ions also relies on a very good qualitative agreement displayed in fig.14. This is needed because 2D color plots such as the one in fig.11 immediately reinforce the feeling of qualitative agreement probably because our vision is well trained to shape recognition in images. If in addition, a doubt is present due to approximate scattering theory, the hope of quantitative comparison rapidly degrades. The other option which became available only recently is to adjust the model to the experimental data. In ref.³⁰, the diffraction data from neon displayed in fig.7 and fig.8 were compared with a recent *ab initio* calculation of the PEL by Miraglia and Gravielle⁷¹ who kindly accepted to provide a compact description of their results in the form of effective binary interaction potentials. After the quantum scattering of helium using the close coupling code developed by Zugarramurdi and Borisov³¹, the comparison with data showed qualitative agreement but also systematic discrepancies. Together with L. Lavoine, we first tried to modify by hand each of the parameters of the binary potentials by a few percent but an optimization procedure was obviously needed. AG Borisov grab the problem from a different point of view and increased the performance of the close coupling code by a factor hundred⁷² allowing automatic search of the best fit to the data. Fig.21 reports the first quantitative agreement of GIFAD data overall diffraction orders and over a significant range of E_{\perp} . The interaction potential derived is therefore an empirical PEL but it can also be seen as a refinement of the initial calculation or a sort of data inversion, probably unique with reasonable initial conditions. A useful application would be to take pure surface electron densities and target position, as calculated by density functional suites and to tune the attractive part and, for instance, the target positions, by optimization to diffraction data.

7 Inelastic diffraction.

The previous section, devoted to elastic diffraction showed the profound similarities between GIFAD and TEAS. We now focus on the differences, among which the ability of GIFAD to observe elastic diffraction at large energy E_{\perp} where the standard Debye-Waller factor would predict negligible elastic intensity. A modified Debye-Waller factor^{51,73} was proposed soon after the first observation even if the first estimations were more a measure of the visibility of the inelastic diffraction since the elastic regime was not demonstrated with GIFAD before 2012^{61,74}. When clear elastic diffraction is observed, the Debye-Waller factor is easily measured as illustrated in fig.5b). In this figure, the inelastically scattered intensity shows very well resolved inelastic diffraction peaks corresponding to partially coherent diffraction as modeled in ref.⁵¹. It was rapidly recognized that the thermal movement of surface atoms was responsible for the inelastic diffraction but the theory was not readily adapted to GIFAD and early models trying to reproduce thermal effects did not predict elastic diffraction^{26,75} leaving open questions.

What is the modified DWF?

What are the inelastic polar and azimuthal profiles?

What are the inelastic diffraction intensities?

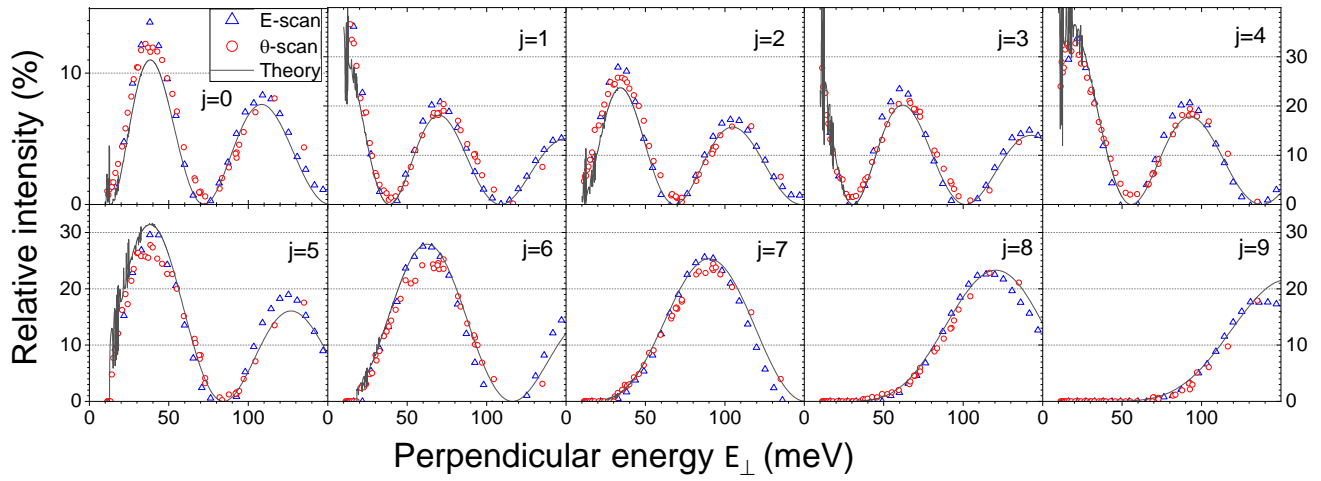


Fig. 21 The neon-LiF $\langle 110 \rangle$ diffraction data reported in the θ _scan in fig.7 and the E_scan in fig.8 are plotted for each diffraction order j , ($\pm j$ added) as a function of the energy E_{\perp} . The black line is a calculation using parameterized projectile-surface interaction potential build on the basis of the *ab initio* calculations of ref.⁷¹ optimized to the data by a new close coupling code⁷² providing the first quantitative agreement³⁰.

A first attempt to answer these questions was proposed in ref.⁵² with the quantum binary collision model (QBCM).

7.1 Classical scattering, momentum transfer, and energy loss.

As illustrated in fig.2a) the projectile trajectory in the (x, y) plane of the fast motion is at the same time very smooth and made of successive localized interaction with individual surface atoms. This suggests that the binary collision model can be applied to study the projectile trajectory. The QBCM only introduces a new aspect which is to consider the surface atom as an harmonic oscillator and to consider the collision in the sudden approximation limit. Let us start with elastic scattering. Each target atom is represented by its Debye vibrational wave-function centered at the equilibrium position and, within a sudden approximation, the binary collision transfers a momentum $\delta\vec{k} = \int_{-t_1}^{t_1} \vec{\gamma} dt$ where $t_1 = a_x/2v_x$ is the time needed to cover half the lattice unit a_x along the x direction and $v_x \simeq v$ the projectile velocity. Assuming a local exponential behavior of the binary effective interaction potential $V_{1D}(z) \propto e^{-\Gamma z}$, the momentum transfer is also expected to have an exponential behavior with the minimum distance of approach $\delta k_z \propto e^{-\Gamma z}$ producing an equivalent individual deflection angle $\delta\theta = \arcsin \delta k_z/k$. Within such simplifying assumptions the trajectory $z(x)$ on the mean 1D potential (V_{1D} in eq.1) can be calculated analytically as well as the density of momentum transfer to the surface $dk(x)/dx$. Attributing the momentum transfer to one atom with mass m_t , per lattice unit, the momentum transferred to individual surface atoms as well as the associated recoil energy $E_r = (\hbar\delta k)^2/2m_t$ can be calculated along the trajectory. The sum ΔE_{Cl} of the recoil momenta $E_r = (\hbar\delta k)^2/2m_t$ with m_t the mass of the recoiling target atom. $\Delta E_{Cl} = \propto \theta_{in}^3$ loss along a grazing trajectory^{18,51}. Where a is the lattice parameter that appears when considering the density of scattering centers on the surface. Compared with the recoil energy $4\mu E\theta^2$ that would arise from a

single binary collision at $2\theta = \theta_i + \theta_f$, ΔE_{Cl} is N_{eff} times smaller:

$$N_{eff} = \frac{6}{\Gamma a \theta_i} \quad (6)$$

This number N_{eff} is interpreted as an effective number of binary collision contributing equally by a small deflection $\delta\theta = 2\theta_i/N_{eff}$. This result obtained with the mean planar interaction potential was numerically checked to remain valid, for trajectories on a 3D surface with atoms at equilibrium positions. It should be noted that giving an infinite mass to the surface atoms *i.e.* neglecting the recoil energies, the projectile classical scattering falls exactly on a perfect Laue circle outlining the validity of ASCA in the classical regime³⁹. Classically, the surface atoms should now be left free to move due to thermal agitation. More precisely their position should display a temperature dependant Gaussian distribution. The scattering profile then becomes much broader with a marked log-normal character (eq.5) showing qualitative similarities with observed scattering profiles^{76,77}.

7.2 Quantum binary scattering.

The quantum aspect is introduced in the model by considering that the surface atoms are trapped in the Debye harmonic oscillator created by the neighboring surface atoms. The binary collision with this atom is considered in the sudden approximation and the momentum transfer δk calculated above is interpreted as the momentum transferred to the center of the atomic wave function $|\psi\rangle$ centered at the equilibrium position. In response, the probability that the oscillator undergoes a vibrational transition can be calculated exactly as $p_e = |\langle \psi | e^{i\delta k z} | \psi \rangle|^2$ which, for an atom initially in the vibrational ground state amounts to $p_e = e^{-E_r/\hbar\omega_D}$ (eq.3) which is significant only when the recoil energy is less than a vibrational energy quantum¹⁸ and identified as the Mössbauer-Lamb-Dicke probability for recoil-less photon emission characteristic of a trapped atom, for instance in an optical lattice. Over the complete trajectory, the probability that all individual binary collisions are elastic is the product of all individual probabilities

$P_e = \Pi p_e$, it factorises as $P_e = e^{-\Sigma E_r/\hbar\omega_D} = e^{-\Delta E_{cl}/\hbar\omega_D}$. Compared with the standard Debye-Waller factor used in TEAS, a factor N_{eff} appears in the exponent supporting the early interpretation by Rousseau *et al.*⁷³ and Manson *et al.*⁵¹ that the modified Debye-Waller factor adapted to GIFAD can be interpreted as a sharing of the momentum transfer $2k_{iz}$ among N_{eff} equivalent scatterers. As a result, GIFAD has demonstrated clear elastic diffraction up to a few hundred meV, and well-resolved inelastic Bragg peaks up to a few eV perpendicular energy E_{\perp} . Of course, the QBCM suffers from imperfections such as the neglect of the attractive forces but this aspect is under development and results do not contradict this simple approach. More problematic, the eigenstates of the surface vibration modes are not the localized Einstein or Debye modes but the phonon modes as identified in TEAS (see *e.g.*⁷⁸). If the localized vibrational excitation would be inside the bulk, its decomposition to the phonons modes would be an almost perfect dilution since the Fourier transform of a delta function is a white homogeneous distribution.

This might not be the case on a surface where specific vibration modes exist⁷⁹ that are localized, but using a slab approach of the surface, Schram and Heller⁸⁰ suggested that the initial excitation could indeed dilute in many phonon modes and that the apparent quasi-elastic diffraction could be due to the excitation of very low energy mode having a negligible contribution to the angular and energy straggling. It should be noted that, when applied to TEAS, the Lamb-Dicke probability p_e is exactly the Debye-Waller factor suggesting that the vision of a local excitation probability maybe meaningful?

7.3 Three different regimes.

Each binary collision is either elastic or inelastic but the probability that all N_{eff} equivalent collisions are elastic or inelastic are well separated revealing an intermediate region as illustrated in fig.22. On the left-hand side, a quasi-elastic regime can be defined where the overall elastic probability P_e is significant *i.e.* where both elastic and inelastic diffraction should be observed. The effective number of inelastic collisions can be evaluated by weighting the effective number of collisions N_{eff} (eq.6) by the probability $p_i = p_e$ that the collision is inelastic. Fig.23 shows that this quasi-elastic regime corresponds to at most one inelastic collision with a possible contribution of two collisions when the elastic fraction is below a few percent. On the right-hand side, the quasi-classical regime where almost all collisions are inelastic so that classical simulation should be valid. Note that the perturbative approach proposed here that the resulting angular straggling is simply the sum of individual ones calculated along a common trajectory evaluated on a rigid lattice may reach its limits because the individual deflections are becoming larger and larger as the distance of closest approach decreases. In between lies a broad intermediate region where elastic diffraction is not present and where inelastic diffraction progressively blurs but where classical simulation models might fail because the quantum features should continue to play a role.

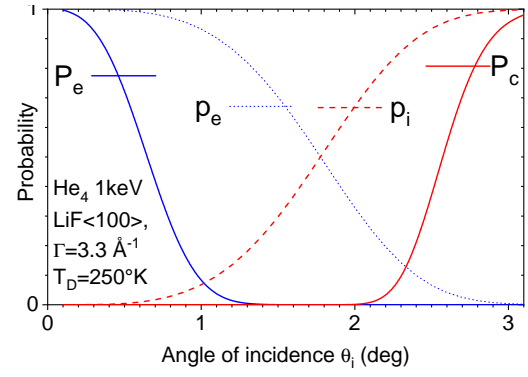


Fig. 22 The probability p_e and $P_e = \Pi_1^{N_{eff}} p_e$ of individual and overall elastic scattering respectively. The complementary inelastic probabilities $p_i = (1 - p_e)$ and $P_c = \Pi_1^{N_{eff}} p_i$ are plotted as a function of the angle of incidence θ_i for 1 keV He.

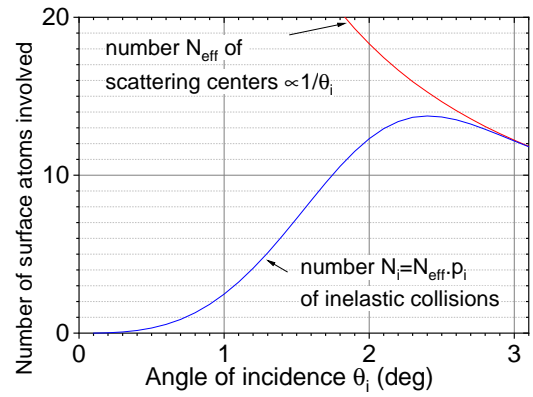


Fig. 23 For the same conditions as fig.22, the effective number of scattering centers given by eq.6 is plotted together with the mean number of inelastic collisions given by $p_i N_{eff}$ with $p_i = 1 - p_e$ given by eq.3.

7.4 inelastic scattering.

The second aspect of the QBCM is to propose a perturbative strategy to evaluate the energy and angular straggling associated with an inelastic collision. If the collision is elastic then the target atom wave function $|\psi\rangle$ is left unchanged and the transferred momentum δk leaves no possibility to localize the target atom. At variance, if the collision is inelastic, all possible vibrational excitation should be calculated together with the associated momentum transfer. The QBCM empirically considers that the classical description of collision is a good approximation of the angular and energy straggling. The target atom wave-function $|\psi\rangle$ centred at equilibrium is replaced by its position probability distribution $|\langle\psi|\vec{r}|\psi\rangle|^2$. For a harmonic oscillator at temperature T , this distribution is a Gaussian function with a standard deviation given by eq.4. Each value of \vec{r} produces a specific deflection angle. Restricting here to target movement along z , a displacement from equilibrium by δz gives a scattering angle $\delta\theta = e^{-\Gamma(z+\delta z)}$. When weighted by the Gaussian probability distribution of z this produces a log-normal distribution (eq.5) centered around the elastic value $\delta\theta_e = e^{-\Gamma z}$. The relative width $w = \delta\theta/\theta_i = \Gamma\sigma$ is independent of the angle of incidence θ_i . Considering that the thermal variation δz as much smaller than the distance z of the projectile

to the surface atom the QBCM suggests a statistical treatment of the angular and energy straggling *i.e.* that the variance of the polar inelastic scattering distribution is the sum of the variance of all inelastic events.

7.5 azimuthal and polar profiles.

The QBCM evaluates the inelastic momentum transfer from the classical trajectories. The mean classical trajectory evaluated from the mean interaction potential $V_{1D}(z)$ is enough to get significant values for the elastic and inelastic scattering probabilities but has no chance to provide a quantitative estimate of the lateral and polar shapes $\delta\phi_f$ of the inelastic diffraction peaks. When the collision is considered inelastic, we consider that the 3D classical distribution. This alone does not produce a very large broadening if only the 'on-top' trajectories are considered. To evaluate all condition for binary collisions, the momentum exchange leading to inelastic effects have to be averaged over all impact parameter inside the lattice unit. Each inelastic even provides a radial momentum transfer oriented along the internuclear axis and all contributions are added to generate an individual inelastic scattering. These are then self convoluted as many times as the number of inelastic events.

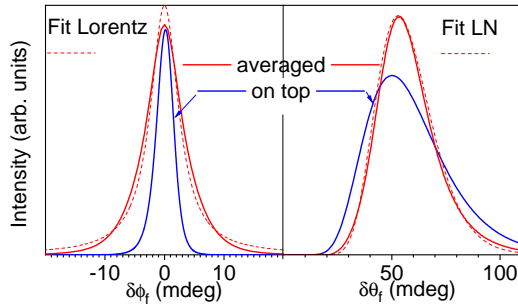


Fig. 24 Angular straggling $\sigma\theta_f$ (right) and $\sigma\phi_f$ (left) of an individual inelastic collision averaged over the lattice cell¹⁸. Compared with the "on-top" trajectory, the lateral broadening has acquired a partial Lorentzian character while the log-normal scale parameter $w \sim \sigma\phi_f/\theta_s$ describing the broadening of the polar distribution is reduced by $\approx 30\%$.

The three coordinated of the atom $|\langle\psi|\vec{r}|\psi\rangle|^2$ wave-function $|\psi\rangle$ centred at equilibrium are replace by a their position probability distribution $|\langle\psi|\vec{r}|\psi\rangle|^2$. This is true also for the polar profile, the model described above predicts a log-normal distribution but only for a single collision and for an "on top" binary collision. For multiple collisions, the polar inelastic profile should be a convolution of such log-normal distributions and though such convolution is not a log-normal profile in general the numerical convolutions turned out to keep a very strong log-normal character, probably because the relative width parameter w is small (*e.g.* $w=0.06$ in fig.5b). Still, on top" collisions are specific and transfer momentum mainly along the z direction. Taking into account a reasonable value of the inelastic momentum transfer $\delta\vec{k}$ induced by an inelastic collision with a surface atom at a given location with respect to the projectile, classical trajectories are needed. This is however comparatively easy because only a lattice site has to be inspected to evaluate the vectorial aspect of the momentum

transfer. This has been done for LiF^{18,52}, and the fig.24 shows that the resulting polar profile keeps a log-normal character but with a width reduced by 30% while the azimuthal width becomes larger acquiring large tails but significantly less than a Lorentzian profile. the θ_scan in fig.7 and the E_scan in fig.8

7.6 inelastic profiles in E_scan and θ_scan .

With regard to the inelastic diffraction, the energy E_\perp is no longer a good quantum number and, for instance, the E_scan of fig.8 and θ_scan of fig.7 which have similar elastic diffracted intensities when plotted as a function of E_\perp (fig.21) have very different inelastic scattering widths. This later is determined as the line profile providing a good fit of the inelastic diffracted intensities as illustrated in the bottom of fig.17. To first order, the inelastic line-width was found almost constant during the E_scan while, it was found to increase almost linearly with E_\perp during the θ_scan . In both cases, diffraction progressively disappears as the inelastic line-width $\sigma\phi_f$ compares with the Bragg angle ϕ_B which behave exactly opposite; constant during a θ_scan and decreasing with $\sqrt{E_\perp}$ in an E_scan . This During an E_scan , the projectile trajectory is essentially unchanged, it is only shifted closer to the surface as the energy E_\perp increases. The effective number of collisions N_{eff} is constant as well as the overall elastic deflection angle $\theta_i + \theta_s$ and the individual elastic deflection angle $\delta\theta_s = 2\theta_s/N_{eff}$. According to the QBCM, each inelastic collision has a log-normal angular distribution characterized by a constant ratio w between the elastic scattering angle and the inelastic broadening (eq.5) so that, in the quasi-elastic regime, the polar and azimuthal inelastic profiles are expected to remain constant during an E_scan . During a θ_scan , the number of binary collisions decreases with the angle of incidence θ_i (eq.6) while the overall deflection increases so that each binary collision is associated with an individual elastic deflection angles $\delta\theta_s$ scaling with θ_i^2 , *i.e.* with E_\perp in a θ_scan .

7.7 Why inelastic is almost like the elastic.

When analyzing the diffraction images, *e.g.* in fig.5b), it was noted that in spite of a sharp variation of the absolute intensity, the diffracted relative intensities above and below the Laue circle are different but show a perfect continuity with the elastic one. To interpret these inelastic diffracted intensities a wavelength is needed! It was found experimentally^{18,53} that using the half sum $k_{eff} = (k_{i\perp} + k_{f\perp})/2$ also offers a continuity in the data analysis. The inelastic diffracted intensities measured at k_{eff} closely match the elastic diffracted intensities measured at the same value of $k_{i\perp} = k_{eff}$ provided that k_{eff} is chosen within the fwhm of the inelastic scattering distribution *i.e.* not too far from specular.

Within the QBCM, this can be explained in the quasi-elastic regime where, on average, only one inelastic collision is responsible for each event of the inelastic profile. The blue peaks in the trajectory in fig.2 correspond to the acceleration γ_z and their integral correspond to the momentum transfer to the surface atoms. The recoil energy E_r should scale as the square of these values producing a distribution well-localized around the turning point of the trajectory where the distance z to the surface is minimum. The inelastic probability $p_i = 1 - p_e$ can be expanded

as $p_i \sim Er/\hbar\omega$ and is also expected to be localized around the turning point so that typical inelastic event would consist of two half elastic trajectories with the surface, the way-in with a wave-number k_i and a way-out with a wave-number k_f . In between a single inelastic event provides a small additional kick to the momentum change accumulated in both half collisions. The evolution of the elastic intensities displayed in diffraction charts (e.g. in fig.7) explains why the inelastic diffraction appears as directed upward or downward at different diffraction order or different values of E_{\perp} as in fig.4. The thermal distribution of these kicks along the y and z directions are displayed in fig.24 and the diffraction remains visible as long as this or these additional kicks along y are less than a reciprocal lattice vector.

Table 1 scaling laws, the magnification/amplification factor corresponds to $\theta = 0.5^{\circ} \simeq 10^{-2}$ rad

L	trajectory length	θ^{-1}	10^2
N_{eff}	effective number of scatterer	θ	10^{-2}
ΔE_b	gas phase binary recoil energy	θ^2	10^{-4}
ΔE_{Cl}	classical projectile energy loss	θ^3	10^{-6}
P_i	inelastic projectile scattering probability	θ^3	10^{-6}
E_r	individual recoil energy	θ^4	10^{-8}
p_i	individual inelastic probability	θ^4	10^{-8}
ΔE_{qu}	mean quantum projectile energy loss	θ^7	10^{-14}
E_{rqu}	mean quantum individual recoil energy	θ^8	10^{-16}

The table 1 indicates the important scaling laws the appear in GIFAD. Except for the first two items, all quantities have the primary energy $E \sim 10^3 eV$ as a pre-factor, however, for an angle of incidence $\theta_i = 0.5^{\circ} \sim 10^{-2}$ rad, the successive powers of θ_i allows, for instance, recoil energy E_r on the meV range. Attention should be paid that these scaling laws do not take into account the attractive part of the potential and become irrelevant for energies $E_{\perp} \leq D$ where D would be the depth of the attractive well if any. More work is needed to turn the inelastic model quantitative so that it can be exploited to derive surface Debye temperature or electron-phonon coupling⁸¹, or, reversely, the absolute surface temperature, a key parameter during growth processes. So far it has not been possible with GIFAD to identify specific phonon modes by inelastic diffraction.

8 Applications of GIFAD.

The present paper is focused on academic systems of rare gases diffracted by the LiF surface but fast atom diffraction has been observed on metals such as Ag⁸², Ni⁸³ and semi-conductors ZnSe⁴⁷, GaAs²⁹. Due to the grazing geometry identical to that of RHEED, GIFAD has shown to be compatible with MBE environment where it is mandatory to leave the space facing the surface free of any instrument to allow well-controlled evaporation⁶³. In this context, GIFAD was able to follow *in situ* and online the growth process with pronounced intensity oscillations having a maximum for each layer completion whatever the angles of incidence θ_i or ϕ_i , and whatever the primary energy⁸⁴. In general, the surface sensitivity is extreme, in the sense that like TEAS, fast atom diffraction takes place a few Å above the terminal layer without any possible penetration so that the diffraction images are not contaminated by information originating from the bulk. A property most

valuable to investigate surface reconstructions triggered by temperature²⁹, by oxygen adsorption⁸⁵ or by migration of contaminants⁸⁶, single over-layers of organic⁸⁷ or inorganic insulating⁸⁸ and conducting^{70,89} material and ultra thin films in general.

8.1 Perspective.

From the fundamental point of view, the inelastic diffraction is an important challenge, and collaboration is needed with TEAS where several theoretical approaches^{79,90,91} have been developed to encompass both elastic and inelastic diffraction. Experimentally, a combined GIFAD and HAS study of a thin overlayer, for instance of graphene⁹² would probably help the identification of specific vibration modes and allow significant progress. Regarding elastic diffraction, the recent progress³⁰ should allow a crystallographic software starting with a qualitative adjustment of surface atoms coordinates and effective binary interaction potentials by the HCW model and then final quantitative optimization by quantum scattering programs.

9 Conclusions

Due to the very efficient decoupling of the movement perpendicular and parallel to the surface, GIFAD is a variable energy diffraction technique, where E_{\perp} can be tuned between a few meV up to eV. This large energy range gives access to the physisorption regime where the depth of the attractive potential energy well can be determined and on the other side to the structural regime where atomic positions can be determined with a few pm accuracy and charge transfer processes are easily measured. It seems difficult to reach significantly lower values, such as those needed for quantum reflection⁹³ but an extension to semi-classical and classical regimes is comparatively easy, allowing complementary measurements such as the direct recoil spectrometry^{94,95} providing identification of surface contaminant. The theoretical description of the elastic regime has made significant progress so that the experimental data can now be fitted to a potential energy landscape making the corrugation function a subsidiary output with practical interest but moderate significance. The inelastic regime is much less advanced but is making rapid progress and should develop to help to diagnose imperfect situations such as those encountered during the growth process. Finally, one regret, obvious but sincere, we would have liked to visit our honored guest J.P Toennies soon after GIFAD was discovered.

Table 2 The following abbreviations are used in this manuscript:

FAD	Fast Atom Diffraction
fwhm	full width at half maximum
GIFAD	Grazing Incidence Fast Atom Diffraction
HAS	Helium Atom Scattering
HCW	Hard Corrugated Wall
PEL	Potential Energy Landscape
QBCM	Quantum Binary Collision Model
RHEED	Reflection High Energy Electron Diffraction
TEAS	Thermal Energies Atom Scattering

Conflicts of interest

There are no conflicts to declare.

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