Fast Atom Diffraction at Surfaces

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Abstract. Recently pronounced diffraction effects for grazing scattering of fast light atoms and molecules with energies up to some keV under axial surface channeling were observed. The rich diffraction patterns provide information on the interatomic spacings between axial surface channels and on the corrugation of the interaction potential. The latter effect can be used to study the structure of surfaces with fast atoms via an interferometric technique. The new method shows similarities to thermal He atom scattering (HAS), but has a number of advantages as simple tuning of the projectile energy (de Broglie wavelength) and, in particular, an orders of magnitude more efficient detection of scattered projectiles. As an example for the application of Fast Atom Diffraction (FAD) for studies on the structure of surfaces, we present results for the rumpling of the LiF(001) surface. The quantum coherence in the scattering process is preserved by specific features of surface channeling which is investigated in detail via the coincident detection of the diffraction patterns with the energy loss of scattered atoms. It turns out that the suppression of electronic excitations owing to the band gap of insulator surfaces play a key role for coherent scattering and the application of FAD in surface science.

1. Introduction

In recent years diffraction patterns were observed in the angular distributions for collisions of fast atoms with surfaces at grazing angles of incidence [1,2]. Specific features in the regime of surface channeling lead to quantum effects for the scattering of fast atomic projectiles on well ordered crystal surfaces [1-4] or thin films [5]. The angular resolution in the experiments can be made so high that diffraction patterns are resolved, irrespective of an unfavourable matching of the de Broglie wavelength associated to the center of gravity motion of the fast atomic particles with typical periodicity length scales of a crystal lattice. Furthermore a suppression of excitations of the target by the fast projectiles results from the relatively large distances of closest approach to the topmost surface layer. This holds in particular for insulators and semiconductors with electronic band-gap [6].

In the meantime this new regime of atomic collisions with solids was employed in investigations on a broad class of different materials. Fast Atoms Diffraction (FAD) was first observed for scattering from insulator surfaces with wide band gap [1-3] and then also for semiconductor surfaces [4], ultrathin films deposited on substrates [5], ordered layers of adsorbates [7], and even clean metal surfaces [8,9]. Attractive properties for the application of the FAD method are the extreme sensitivity for the region of the topmost layer of surfaces, the negligible particle induced damage or modification
of the surface, no charging effects for insulator surfaces, and a high efficiency of detection which allows one to record complete diffraction pattern by means of position sensitive detectors in a short time.

In this paper we discuss the basic features of FAD and demonstrate the power of the method as analytical tool in surface science. By introducing an interferometric technique to the scattering of fast atoms from surface, the rumpling – a relative shift of atoms at the topmost surface layer – of the LiF(001) surface was determined with unprecedented precision. For such studies the coherence in the quantum scattering process is crucial. We briefly outline recent progress in studies on the mechanisms leading to decoherence during the scattering process.

2. Quantum scattering in grazing collisions of fast atoms from surfaces

In figure 1 we show a sketch of the collision geometry for grazing scattering of fast atoms from a LiF(001) surface along a low indexed <110> direction. Here scattering from the topmost layer of the surface proceeds in the regime of “axial surface channeling” where the effective scattering potential results from an averaging along the strings as shown for an equipotential energy surface of 3 eV for the interaction of He atoms in figure 2. This results in an effective two-dimensional potential in a plane normal to the propagation of the projectiles. This potential has the form of a corrugated sheet (cf. right panel of figure 2) and gives rise to scattering out of the incident collision plane – with a “rainbow” at the extreme of azimuthal angular deflection – and in the regime of quantum scattering to diffraction patterns in the angular distribution of scattered projectiles as sketched in figure 1.

![Figure 1. Sketch of the collision geometry for axial surface channeling from LiF(001) surface.](image)

For a simple understanding of the diffraction patterns we show in figure 3 a sketch of the equipotential energy surface in a plane normal to the direction of the incident beam for scattering along <110>, i.e. conditions as shown in figure 1 with alternating strings formed by F- or Li+ ions. In most cases the corrugated potential plane is well approximated by a sinusoidal hard wall which can be considered here as reflection grating for the scattered fast atoms. For quantum scattering the periodicity interval d leads to Bragg scattering with constructive interference, if \( n \lambda_{db} = d \sin \Psi \), with the azimuthal angle \( \Psi \) and the de Broglie wavelength \( \lambda_{db} = h/(2ME_o)^{1/2} \) (h Planck’s constant, M mass of particle). The corrugated potential leads to a blazing effect similar as for an optical echelette grating which enhances the intensity of finite diffraction orders.
**Figure 2.** Left panel: Equipotential energy surface $V(x,y,z) = 3 \text{ eV}$ for interaction of He atom in front of LiF(001) surface. Right panel: Effective potential energy surface $V(x,z) = 3 \text{ eV}$ for grazing scattering along y direction ($<110>$). Arrow in left panel indicates direction for averaging of $V(x,y,z)$.

The motion of projectiles with kinetic energy $E_0$ in a plane normal to the incident beam proceeds with an energy $E_\perp = E_0 \sin^2 \Phi$ so that for grazing angles of incidence of $\Phi \approx 1^\circ$, $E_\perp$ amounts for keV atoms to eV energies only. Therefore projectiles are reflected in front of the topmost layer of surface atoms and do not penetrate into the bulk of the target. It is straightforward to attribute to the motion in the normal plane (cf. figure 3) with energy $E_\perp$ a de Broglie wavelength $\lambda_{dB \perp} = \frac{\lambda_{dB}}{\sin \Phi}$. For grazing scattering $\lambda_{dB \perp}$ is about two orders of magnitude larger than $\lambda_{dB}$ so that the deflection angles $\Theta$ in the angular distributions can be large as follows from the respective Bragg relation $n \lambda_{dB \perp} = d \sin \Theta$. Bragg scattering is sketched in figure 3 by classical trajectories (A, A*, A**).

**Figure 3.** Sketch of contours of equipotential energy surfaces for axial channeling of $^3\text{He}$ atoms with $E_0 = 1 \text{ eV}$ along $<110>$ of LiF(001). A, A*, A**, B represent trajectories with same final deflection angle. “rb” denotes trajectory for rainbow.

The trajectory in figure 3 labeled “rb” represents the maximum in angular deflection which gives rise to a “collisional rainbow”. In close analogy to diffraction phenomena for a slit or a grating, scattering from the unit cell determines the intensities of individual Bragg peaks. Since for a corrugation of sinusoidal shape one always finds for angles $\Theta < \Theta_{rb}$ two trajectories with the same deflection angle, interference between trajectories of type A and B determines the scattering within the unit cell, the so called “form factor”. This feature is in close analogy to the atmospheric phenomenon of “supernumerary rainbows” [3]. For deflection angles around the classical rainbow angle $\Theta_{rb}$ the resulting intensity patterns are well approximated by the Airy theory. For small deflection angles the intensities for Bragg peaks of order $n$ are given by $I(\Theta) \sim J_n^2 \left( \frac{\pi \Delta z (1+\cos \Theta)}{\lambda_{dB}} \right)$ with $J_n$ being the
Bessel function of order \( n \) and \( \Delta z \) the corrugation of the equipotential energy surface as indicated in figure 3. Thus the intensity pattern of Bragg peaks as function of \( \Theta \) or \( \lambda \), allows one to derive \( \Delta z \) from experimental data. As an example we discuss here recent work on the corrugation of the LiF(001)-surface [10].

3. Corrugation and rumpling of the LiF(001) surface from Fast Atom Diffraction

In the experiments on the rumpling of the LiF(001) surface, the azimuthal orientation of the surface was chosen such that the incident beam was impinging along a <110> direction in the surface plane. \(^3\)He atoms with energies of some 100 eV up to some keV were scattered from a LiF(001) surface under a grazing angle of incidence \( \Phi_{\text{in}} \) of typically 1°. The fast neutral He atoms were produced by resonant charge transfer of He\(^+\) ions from an ECR ion source in a gas cell in the accelerator beam line. The neutralized beam was collimated by sets of vertical and horizontal slits to a divergence of less than 0.02°. The adjustable slits are components of a two stage differential pumping system in order to maintain a base pressure in the upper \( 10^{-11} \) mbar range in our UHV scattering chamber. The target was prepared by cycles of grazing sputtering with 25 keV Ar\(^+\) ions under \( \Phi_{\text{in}} \approx 2° \) at a target temperature of about 200°C and subsequent annealing of the target at a temperature of 350°C for about 15 minutes. After a major number of preparation cycles well defined angular distributions for scattered atoms were observed which serve as an indication of a clean target surface widely free from defects.

![Intensity distributions recorded with position sensitive detector for scattering of \(^3\)He atoms from LiF(001) surface under \( \Phi_{\text{in}} = 0.99° \) along <110>. Projectile energies: (a) 0.35 keV, (b) 0.50 keV, (c) 0.65 keV, (d) 0.80 keV, (e) 1.00 keV, (f) 2.90 keV.](image)

Angular distributions were measured by means of a position sensitive micro-channelplate detector which was mounted 66 cm behind the target. With typical count rates up to 10000 counts per second, a complete diffraction pattern was recorded within minutes. This is a major advantage compared to the conceptually similar method of thermal energy atoms scattering (TEAS) with He atoms where detection efficiencies are several orders of magnitude smaller. In figure 4 we show a series of angular distributions for \(^3\)He atoms with energies between 0.3 keV (panel a) and 2.90 keV (panel f) as recorded with the position sensitive detector. The horizontal axis represents the azimuthal exit angle, the vertical axis the polar exit angle, where the circles denote the conservation of normal energy for scattering from axial strings formed by surface atoms. According to the simple Bragg relation for the...
azimuthal angle $\Psi$ the angular positions of peaks for a fixed $\Phi_m$ depend on $\lambda_{dB}$ so that with increasing projectile energy the angular positions of the peaks decrease.

Striking feature of the data in figure 4 is the relative intensity of Bragg peaks as function of $\lambda_{dB}$. For the low diffraction orders $n$, starting with order $n = 0$ at $\Psi = 0$, one finds an oscillatory behaviour for a variation of the energy and thus of $\lambda_{dB}$. For an energy of 0.35 keV pronounced peaks for the orders $n = 0$ and 2 are observed, whereas the intensity of the peak for the odd order $n = 1$ is weak. At 0.65 keV an opposite intensity diffraction pattern is found with respect to even and odd orders.

**Figure 5.** Projections of intensities into annulus as indicated by circles in figure 4 as function of deflection angle $\Theta$ for angular distributions displayed in figure 4.

Projections of data into an annulus as indicated by circles for the data sets presented in figure 4 are plotted in figure 5 as function of the deflection angle $\Theta$. This presentation of relative peak intensities allows one to analyze the data on a quantitative level. The solid curves represent best fits to the diffraction peaks to Lorentzian lineshapes where the relative intensities $I_n$ of Bragg peaks of orders $n$ are taken from the Bessel functions $J_n(\pi \Delta z (1+\cos\Theta) / \lambda_{\perp})$ with $\Delta z$ as a parameter. With this procedure one derives from data sets as shown in the panels of figure 5 the corrugation of the interaction potential as function of the normal energy $E_{\perp}$. In this respect, tuning of $E_{\perp} = E_{\perp} \sin^2\Phi_m$ can simply be achieved by changing of the projectile energy and/or the angle of incidence $\Phi_{in}$.

In figure 6 we present results from a detailed study on the corrugation of the interaction potential for scattering of $^3$He and $^4$He atoms from a LiF(001) surface along <110> under angles of incidence $\Phi_{in} = 0.71^\circ$, $0.81^\circ$, $0.96^\circ$, and $0.99^\circ$. Within the scatter of data we find that the corrugation of the potential $\Delta z$ is constant from normal energies of a some 10 meV up to about 1 eV. The solid curve in figure 6 represents the corrugation for a He-LiF(001) interaction potential calculated by Pruneda from density functional theory (DFT) [11] which is in excellent accord with the analysis of the experimental data.
Figure 6. Corrugation $\Delta z$ of the He-LiF(001) interaction potential from diffraction studies with $^3$He and $^4$He atoms scattered under $\Phi_{in} = 0.71^\circ$ (open circles), 0.81° (open diamonds), 0.96° (open triangles), and 0.99° (open squares). Solid curve: DFT calculations by Pruneda [11].

In the DFT calculations on the interaction potential for He atoms in front of the solid the structure of the surface is also obtained. It is a long standing debate to what extent the positions of lattice atoms are reconstructed in the surface region. Of particular interest is the so called “rumpling” – a slight vertical shift of the topmost Li$^+$ ions compared to F$^-$ ions of the LiF crystal. For LiF(001) a rumpling of some 1/100 Å was derived within the last decades in theoretical as well as experimental studies.

Figure 7. Normalized relative intensity of diffraction order $n = 0$ (top) and $n = 1$ (bottom) for scattering of $^3$He atoms from LiF(001) along <110> under $\Phi_{in} = 0.61^\circ$ (circles), 0.81° (squares), and 0.99° (triangles) as function of $E_{\perp}$. Curves: Wave-packet simulations based on He-LiF(001) interaction potential taking into account no rumpling (dash-dotted), rumpling of $\delta z = 0.042$ Å (solid curve), and $\delta z = 0.084$ Å (dashed curve).
We have determined the rumpling $\delta z$ of the LiF(001) surface in studies on fast atom diffraction by plotting in figure 7 the intensities of the Bragg peaks of order $n = 0$ and $n = 1$ as function of the normal energy $E$ [10]. The data are analyzed in terms of semiclassical trajectory calculations where interaction potentials derived from DFT calculations for different values of $\delta z$ are considered. As revealed from the comparison between the experimental data and the calculations, good agreement is found for assuming a rumpling of $\delta z = 0.042 \text{ Å}$, whereas for two other choices of $\delta z = 0 \text{ Å}$ (dashed-dotted curve) and $\delta z = 0.084 \text{ Å}$ (dashed curve) pronounced deviations are evident.

Figure 8. Left panel: Mean square deviation as function of $\delta z$ for fit to experiment in figure 7. Right panel: Comparison of rumpling $\delta z$ from this work with results from previous studies.

In the left panel of figure 8 we have plotted the mean square deviation between the experimental and the calculated data as function of the rumpling $\delta z$ for the Bragg diffraction orders $n = 0$ (full circles) and $n = 1$ (full diamonds). A best fit is obtained for $\delta z = 0.042 \text{ Å}$ where a change of the rumpling by $\pm 0.015 \text{ Å}$ increases the mean square deviation by about 50 percent. For the final determination one has to take into account the thermal vibrations of lattice atoms which result in a slight effective outward shift of the interaction potential with different contributions for Li$^+$ ions ($\Delta_{Li} = 0.025 \text{ Å}$) and F$^-$ ions ($\Delta_{F} = 0.017 \text{ Å}$). The corrected rumpling amounts to $\delta z = 0.05 \text{ Å}$ with an estimate of the experimental uncertainty of $\pm 0.015 \text{ Å}$. Taking into account effects caused by approximations in the DFT calculations on the interaction potential, the overall present value for the rumpling is $\delta z = (0.05 \pm 0.04) \text{ Å}$ [10]. In the right panel of figure 8 we compare our result with experimental and theoretical work over the last decades. Our result compares well with the calculations and a recent experimental analysis basis on IV-LEED, however, the uncertainty with respect to $\delta z$ could be clearly reduced.

4. Decoherence in the scattering process

For the quantum scattering process it is crucial that the coherence persists in the collision with the solid. For fast atoms this is a nontrivial issue, since the probabilities for excitations of electrons and atoms of the target atoms by keV atomic projectiles are in general considerable. Then in terms of the “which way problem” in quantum physics local excitations allow one to identify the pathway of projectiles so that quantum interference is destroyed. On the last ICPEAC-conference this issue was discussed by Aigner et al. [12]. For LiF electronic excitations are suppressed owing to the wide band gap of about 14 eV so that electronic excitations can proceed only via the production of excitons (12 eV excitation energy) or emission of electrons to vacuum. As a consequence, one finds in the energy loss spectra for scattering of atoms from a LiF surface discrete energy loss peaks. For scattering conditions used for FAD experiments, the energy loss spectrum for H atoms in panel b of figure 9 is dominated by elastic scattering of atoms. With a setup that provides the recording of angular
distributions for scattered atoms in coincidence with their energy loss we could recently demonstrate that the diffraction pattern is closely related to the suppression of electronic excitations. For elastically scattered 1 keV H atoms a well defined diffraction pattern is resolved (panel d), whereas for projectiles scattered with a discrete energy loss owing to electronic excitations no signatures of diffraction can be found [6]. This feature can be explained by the initial local capture of electrons from F\(^-\) sites of the crystal lattice at the surface.

**Figure 9.** Angular distributions for scattering of 1 keV H atoms from LiF(001) along <110> in coincidence with projectile energy loss as shown in panel (b). (a) total intensity, (c) pattern for discrete energy loss, (d) pattern for elastically scattered atoms.

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