Influence of Inelastic Processes on Fast-Atom-Surface Diffraction

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Abstract. Diffraction of fast helium atoms at alkali-halide surfaces under grazing angles of incidence shows intriguing diffraction patterns. The persistence of quantum coherence is remarkably strong, even though high surface temperatures and high (keV) kinetic energies of the incident atoms would strongly suggest the dominance of dissipative and decohering processes. The main source of decoherence is the excitation or absorption of surface vibrations upon impact. The momentum transfer between the surface and the incident helium atom depends on the amplitude of the thermal vibrations of the surface atoms and the energy of the incident particle. We present an ab initio simulation of the quantum diffraction of fast helium beams at a LiF (100) surface in the ⟨110⟩ direction, and compare with recent experimental diffraction data.

1. Introduction

Ever since the pioneering experiments by Estermann and Stern [1], diffraction of massive particles scattered at surfaces has remained one of the most impressive illustrations of the quantum wave nature. Estermann and Stern observed interference patterns of slow (“thermal”) helium atoms and $H_2$ molecules scattered off alkali halide surfaces. In order to investigate the quantum-classical crossover, interference of much more massive objects like fullerenes scattered on gratings has been studied successfully [2, 3]. In matter-wave interferometry of large and complex biomolecules, de Broglie wavelengths as small as few picometer ($10^{-12} \text{ m}$) have been reached [3]. The same regime of small de Broglie wavelengths $\lambda_{dB} \to 0$, can also be approached by using lighter particles with a considerably larger kinetic energy. Recently, sub-picometer wavelengths have been accessed by scattering studies of fast atoms with keV energies at surfaces [4, 5, 6] with $\lambda_{dB}$ as small as $\approx 200$ femtometers. In spite of the remarkably small wavelength, the diffraction patterns obtained by these experiments do not only show clear evidence of coherent wavepacket interference, but even provide detailed structural information about the scattering surface. This is all the more remarkable, as thermal fluctuation amplitudes of the surface atoms are much larger than $\lambda_{dB}$. Especially at high surface temperatures, thermal displacement of surface atoms leads to a considerably enhanced momentum transfer between the incident atom and the surface. The collisions of projectiles at keV energies with the surface would strongly suggest the dominance of dissipative and decohering processes.
Figure 1. Helium atoms scattered at the LiF-surface at a grazing angle of incidence $\theta$ (a). In 100-direction, the He atoms interact with longitudinal chains of either Li or F atoms (b). Averaging the He-LiF potential along the longitudinal direction yields an effective two-dimensional potential.

In this communication we present first examples of ab initio simulations of fast helium atom diffraction ($^4$He) at a LiF(100) surface. We analyze the suppression of decoherence in grazing-incidence scattering within an open quantum system (OQS) approach and quantitatively reconstruct experimental diffraction images [4, 5].

2. Method

Fast atom scattering at surfaces under small grazing angles ($\theta_m \approx 1^\circ$) is characterized by widely disparate kinetic energies parallel (Fig. 1a) ($E_\parallel = E_0 \cos^2 \theta_m \approx E_0$) and normal ($E_\perp = E_0 \sin^2 \theta_m \approx E_0 \theta_m^2$) to the surface. This leads to vastly different projected de Broglie wavelengths

$$\lambda_{dB,\perp} = 2\pi/k_\perp = 2\pi \hbar/\sqrt{2ME_\perp},$$

where $k_\parallel = k_x$ and $k_\perp = k_z$ are parallel and normal components of the wave vector of the incident beam, respectively. While $\lambda_{dB,\perp}$ is comparable to the lattice spacing ($a = 2.01\,\text{Å}$ for LiF) $\lambda_{dB,\parallel}$ is orders of magnitude smaller. The fast motion along the surface leads to an approximate separability of the longitudinal and the transverse degrees of freedom and a weak coupling during the collision between them [7]. By averaging the atom-surface potential along the direction of the incident atom (x-direction), the scattered particle experiences an effective two-dimensional atomic string potential $V_{str}(y, z)$ (Fig. 1b).

Projecting the outgoing wave onto the plane transverse to the beam direction $|\langle k_y k_z | \Psi_{out} \rangle|^2$, two-dimensional diffraction patterns (see Fig. 1a) emerge. The remarkable persistence of quantum coherence in the presence of an energetic collision with keV energies and lattice vibrations at elevated temperatures in the experiment ($T \approx 620 \,\text{K}$ [4, 6]) is the key to quantitatively analyzing diffraction patterns and extracting structural and dynamical information.

Starting point of the open quantum system approach (OQS) [2] is the reduction of the Liouville-von Neumann equation

$$i\frac{d\rho(t)}{dt} = [H, \rho(t)],$$

for the density operator $\rho$ of the entangled system of atomic wavepacket and surface to a Lindblad
equation
\[ \frac{d\sigma(t)}{dt} = -i [H_S, \sigma(t)] + R\sigma(t), \]
for the reduced density operator
\[ \sigma(t) = Tr_R [\rho(t)] = \sum_i \{i|\rho(t)|i\}, \]
describing the translational motion of the atomic wavepacket where all “reservoir” degrees of freedom (\(R\)) representing the LiF surface have been traced out. The partial trace in Eq. (4) includes both the average over thermal excitation of the surface as well as collisional excitations of the surface by the interacting beam, which is the root cause for decoherence represented in Eq. (3) by the so-called relaxation operator \(R\).

Under surface channeling conditions, the longitudinal motion is effectively decoupled from other degrees of freedom (except for cases of resonant coherent excitations [14], i.e. frequency matching of the longitudinal corrugation with internal degrees of freedom of the projectile). We therefore separate the system Hamiltonian in longitudinal and transverse parts.

\[ H_S = \frac{P^2}{2m} + V_{str}(y, z) + \frac{P^2}{2m} \]

The Hamiltonian contains the effective transverse interaction potential of atom with the LiF surface, \(V_{str}(y, z)\). It is obtained by averaging the full potential \(V_{surf}(\mathbf{r})\)

\[ V_{str}(y, z) = \frac{1}{d} \int_{x_0}^{x_0+d} dx \, V_{surf}(x, y, z), \]

where \(d\) is the space between two adjacent F of Li atoms.

The initial density matrix of the atomic beam prior to entanglement with the solid,

\[ \sigma(t = 0) = \int d^3p \, |\Psi_{p_i}\rangle\langle\Psi_{p_i}| \]
corresponds to an integral over wavepackets representing the incoherent energy- and angular spread of the initial beam.

In the presence of an environment with a very large number of degrees of freedom, the quantum-to-classical crossover can be understood within the framework of decoherence theory [1], or alternatively, elements beyond standard unitary quantum dynamics, including the frequently involved “collapse” of the wavefunctions [8, 9]. We solve the Lindblad equation by a quantum trajectory Monte Carlo (QTMC) method [10] which is an extension of similar techniques in quantum optics [12]. In the present case of free-particle wavepackets, the QTMC reduces to an ensemble average over solutions of a stochastic linear Schrödinger equation. The evolution of the wavepacket is given by a sequence of collisions and propagation in the effective potential. Each stochastic realization of the wavepacket at the time \(t = t_N\) is given by

\[ |\Psi_i(t)\rangle = \prod_{j=1}^N U_{coll}(q_j, t_j) U_{cont}(t_j, t_{j-1}) |\Psi_i\rangle, \]

where \(|\Psi_i\rangle\) is the initial wavepacket at \(t_0 = 0\). The evolution operator for an impulsive stochastic collisional momentum exchange \(q_j\) with the surface is given by

\[ U_{coll}(q_j, t_j) = \exp (i \vec{q}_j \vec{r}), \]

while the continuous evolution in between two “kicks” during the interval \([t_j, t_{j-1}]\) is given by

\[ U_{cont}(t_j, t_{j-1}) = \exp (-i H_S (t_j - t_{j-1})), \]

with the system Hamiltonian \(H_S\).
3. Atom-surface momentum transfer and decoherence

Momentum transfers \( \{ \vec{q}_j \} \) as well as collision times \( \{ t_j \} \) are stochastic sequences determined by collisional interactions with the environment. The resulting diffraction pattern in the transverse plane is calculated as an ensemble average over stochastic realizations.

The momentum transfer distribution functions in longitudinal \((x-)\) direction and in the transverse \((y,z-)\) directions entering Eq. 9 are obtained along sequences of binary collisions between helium and lithium and fluorine by averaging over the random displacement of the thermally excited lattice atoms. The mean square displacements of the atoms are given by \[ \langle u^2 \rangle = \frac{3\hbar T}{M k_B \Theta^2_D}, \] where \( \Theta_D \) is the surface Debye temperature, \( M \) the mass of the corresponding lattice atom and \( T \) the temperature of the surface. The Debye temperature was estimated as \( \Theta_D = 530K \).

As shown in Fig. 2, the \( z \)-component of the momentum transfer \((\Delta k_z)\) is considerably larger than the momentum transfer in the other two directions. All the momentum transfer distributions become wider as the energy of the incident atom increases, since faster incident atoms lead to larger momentum transfer between the atom and the surface. In the range of 0.5keV to 2keV however, this increase is only moderate. The widths of the momentum transfer distributions remains in the same order of magnitude throughout this energy range (Fig. 2).

The momentum transfer in the transverse plane \((\Delta k_y, \Delta k_z)\) displaces the momentum of each wavepacket. As shown in Fig. 2, the \( z \)-component of the momentum transfer \((\Delta k_z)\) is considerably larger than the momentum transfer in the other two directions. This leads to an elongation of the diffraction peaks in \( z \)-direction in the resulting diffraction picture (see Fig. 4). The effect of \( \Delta k_x \) is more subtle: it causes velocity and kinetic energy changes of the parallel motion. The energy exchange with the surface (of the order of \( \approx eV \)) results in random phases in the continuous time evolution.

Diffraction images can be successfully reconstructed only when decay of coherence across the beam profile due to encoding of localization information ("which path" information) is accounted for. Neglecting first the initial beam divergence, we consider an incident Gaussian wavepacket (Fig. 3) of the form

\[ \Psi_{in}(\vec{r}) = e^{ik_{∥}x} \Psi_{⊥}(y, z) \] (12a)

with

\[ \Psi_{⊥}(x, y) \propto e^{-ik_{⊥}z} e^{-y^2/2\sigma^2_y} e^{-(z-z_0)^2/2\sigma^2_z}. \] (12b)

\( \sigma_{y,z} \) are the transverse coherence length of the incident beam determined by the width of the zeroth-order diffraction peak \( (\sigma^m_y = \sigma^m_x \approx 2L \frac{\lambda_{min}}{D}) \). The latter is controlled by the collimation slit of width \( D \) positioned at a distance \( L \) in front of the surface. The transverse coherence length is large compared to the lattice spacing. In fact, Eq. (12) is close to a delocalized plane wave. We expand the initial wavepacket in terms of a coherent superposition of narrow Gaussian wavepackets (Fig. 3a) each of which is propagated according to Eqs. (8 to 10). The random evolution phases acquired due to collisions occurring at different instances in time (i.e. different \( x \) coordinates along the string) causes a reduction of the transverse coherence length (Fig. 3b) along the surface normal, \( \sigma^z_{\text{out}} \), of the projected wavepacket, monitored after its center reached distance from the surface \( z = z_0 \) again.

Two portions of the wave packet which are initially separated by the offset \( \delta z \) hit the surface at different positions on the longitudinal \((x-)\) axis (see Fig. 3). If both trajectories suffer the same amount of momentum transfer \( \delta k \) during the impact on the surface, their phase difference
after the impact is $\phi \approx \delta k \delta x$. Only if $\phi \ll 2\pi$, the two paths will interfere constructively. The coherence width of the wavepacket in $z$-direction (perpendicular to the surface) can therefore be roughly estimated as

$$\delta z = \delta x / \theta_{\text{in}} \approx \pi \theta_{\text{in}} / \delta k$$

(13)

A more detailed numerical analysis of the reduction of the coherence width of the incident wavepacket is shown in Fig. 3. The width of the wavepacket is reduced dramatically due to stochastic momentum transfer with the LiF surface. The coherent width of the wavepacket depends on the energy of the incident He atom and on the surface Debye temperature. As higher energies of the incident atom lead to larger momentum transfer, it also causes stronger decoherence effects and narrower resulting wavepackets. Higher surface Debye temperatures on the other hand correspond to smaller thermal oscillation amplitudes of the surface atoms, which leads to less momentum exchange between the He atom and the surface, and, in turn, to weaker decoherence effects. Although the effect of the energy and the surface Debye temperature on the coherence width of the wavepacket is clearly visible, this effect is only moderate in strength. In a wide parameter range, the resulting wavepackets can be approximated by Eq. 12b, with $\sigma_z \approx 0.5 \text{au.}$, which translates into a FWHM of $\sqrt{8 \log 2} \sigma_z \approx 1.2 \text{au.}$.

**Figure 2.** Probability density functions of momentum loss of a He atom hitting a LiF surface with a temperature of 620K. Momentum transfer distribution functions of the x-component (longitudinal direction, a), the y-component (b) and the z-direction (perpendicular to the LiF surface, c) are shown for different energies of the incident He atoms.

We determine $V_{\text{str}}(y, z)$ as the string average along the $\langle 110 \rangle$ direction of the full He-LiF surface potential $V_{\text{surf}}(\vec{r})$ calculated for a large LiF cluster on the multi-configuration self consistent field level, using the quantum chemistry code Columbus [15]. The string potential
along the \((110)\) direction is periodic with \(V_{\text{str}}(y + d, z) = V_{\text{str}}(y, z)\), where \(d\) is the spacing between adjacent strings of \(F\).

4. **Calculating diffraction patterns**

Due to the periodicity of the effective two-dimensional atomic string potential \((V_{\text{str}}(y, z) = V_{\text{str}}(y + d))\), where \(d\) is the space between two adjacent \(F\) or \(Li\) atoms, all the interference maxima correspond to Bragg peaks.

\[
k^{(n)}_{by} = \frac{2\pi}{d}, \quad n = 1, 2, 3, \ldots
\]  

(14)

A typical diffraction pattern at a well-defined incident particle energy shows modulations of the intensity such that not all the Bragg peaks are simultaneously visible. Whether or not a particular Bragg position \(k^{(n)}_{by}\) features an interference maximum at a certain energy depends on the shape of the effective atomic string potential, in particular on its corrugation amplitude.

A key parameter characterizing the diffractive scattering induced by \(V_{\text{str}}\) is the corrugation amplitude of the string potential, \((\Delta z)_{\text{str}}\) i.e. the maximum variation of the normal \((z)\) coordinate of the equipotential surface \(E_1 = V_{\text{str}}(y, z)\). For a wide interval of normal energies \((0.5 \text{ eV} \leq E_{\perp} \leq 10 \text{ eV})\), within which a converged potential for a large cluster can be generated, \((\Delta z)_{\text{str}}\) is remarkably weakly dependent on \(E_{\perp}\) and essentially constant \((\Delta z)_{\text{str}} \approx 0.3\AA\). As some of the experimental diffraction data are taken at even smaller perpendicular energies for \((E_{\perp} < 0.5 \text{ eV})\), where the ab initio potential calculation becomes unreliable, we have extrapolated \(V_{\text{str}}\) to low-energy surfaces subject to the constraint that \((\Delta z)_{\text{str}}\) remains constant. Imposition of such a constraint has proven remarkably successful when compared with experimental data.

Reconstruction of the experimental diffraction pattern requires, in addition to decoherence, incoherent averaging over the geometric angular divergence taken to be rectangular, \(W_R\), and energy spread taken to be approximately, Lorentzian, \(W_L\).

\[
\langle I(\theta_{\text{out}}, \phi_{\text{out}}) \rangle = \int d\theta' \int d\phi \int dE' I(\theta', \phi, E') \times W_L(E' - E) W_R(\theta' - \theta_{\text{out}}) W_R(\theta' - \theta_{\text{out}}).
\]  

(15)

We compare the resulting diffraction patterns to experimental results [4] for scattering of He atoms \((E_0 = 1 \text{ keV})\) on a LiF(001) surface aligned along \((110)\) direction under a grazing angle of incidence \(\phi_{in} = 1.1^\circ(E_{\perp} \approx 0.38 \text{ eV})\). We find a near-perfect reconstruction of the experimental diffraction pattern without any adjustable parameter (Fig. 4). The positions of the main maxima are obtained from the interference condition for a periodic potential along the \(y\)-direction,

\[
\sin \alpha_n = \frac{k_y}{k_\perp} = n\frac{\lambda_{dB\perp}}{d}, \quad n = 0, \pm 1, \pm 2, \ldots
\]  

(16)

The two outermost maxima are the so-called rainbow peaks. Between the rainbow peaks, there are intricate substructures, formed by additional maxima (“supernumeraries”). These structures are well reproduced by the simulation.

Summarizing, fast keV atomic collisions with insulating alkali-halide surfaces give rise to remarkably sharp and well-resolved diffraction images. Despite de Broglie wavelengths as small as one hundred femtometer, quantum interference persists. Due to the weak coupling of transverse and longitudinal degrees of freedom decoherence is effectively suppressed in grazing incidence collisions. Ab initio quantum trajectory Monte Carlo simulations accurately reproduce the diffraction pattern and allow to extract structural surface information such as buckling at an unprecedented level of detail. This technique also promises application to other surfaces and structures relevant for material sciences.
Figure 3. A delocalized wavepacket hits the surface that is considerably wider than the size of the unit cell. Different components of the wavepacket (indicated in (a) by narrower wavepackets) are subject to different random momentum changes, which lead to a partial loss of coherence. The coherent width of the wavepacket is therefore drastically reduced (b). The effective coherence width of the wavepacket moderately depends on the energy of the incident particle as well as on the surface Debye temperature (c).

Figure 4. Experimental (from Ref. [4]) (a) and theoretical (b,c) diffraction patterns for scattering of 1keV He from LiF under $\theta = 1.1^\circ$. In b), with surface buckling $\delta z = 0.053\text{Å}$, c) without surface buckling $\delta z = 0$. Note the different number of interference maxima.

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