PHYSICAL REVIEW B **86**, 241402(R) (2012)

Evidence for longitudinal coherence in fast atom diffraction

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(Received 21 August 2012; published 13 December 2012)

Angular distributions for grazing scattering of keV H atoms from an $Al_2O_3(11\bar{2}0)$ surface were recorded. These distributions reveal defined diffraction patterns which can be understood in terms of quantum scattering from well-ordered surfaces. From the observation of so-called Laue circles, we conclude a high degree of longitudinal coherence for fast atom diffraction at surfaces which allows one to resolve periodicity intervals of several 100 Å. We demonstrate this feature in scattering experiments from the reconstructed (12×4) phase of an $Al_2O_3(11\bar{2}0)$ surface obtained after annealing at temperatures of about 2000 K.

DOI: 10.1103/PhysRevB.86.241402 PACS number(s): 79.20.Rf, 68.49.Bc, 79.60.Bm

In recent years, diffraction patterns were observed in the angular distributions for collisions of fast atoms with surfaces at grazing angles of incidence.[1–3] The suppression of electronic excitations of the target surface[4] and momentum transfer to surface atoms[5,6] under these scattering conditions is important to preserve quantum coherence for observing diffraction patterns for the scattered projectiles. In the experiments, the angular resolution for coherent scattering can be made so high that diffraction peaks are resolved despite the unfavorable matching of the *de Broglie wavelength* of about 10^{-2} Å (or less) associated to the center-of-gravity motion of the fast atomic projectiles with typical periodicity intervals at surfaces of crystals of the order of Å.

This method has been successfully demonstrated in studies on the structure of insulator,[1–3] semiconductor,[7] and metal surfaces[8,9] as well as ordered thin films[10] and adsorbed atoms deposited on clean substrates.[11,12] Attractive features of fast atom diffraction (FAD) are the extreme sensitivity to the topmost surface layer, negligible modification of the surface structure, and no charging effects at insulator surfaces.[3] Based on the efficient detection of atoms with kinetic energies of some 100 eV by means of a position-sensitive detector,[13] complete diffraction patterns can be recorded in times of typically minutes. This is more efficient than for the closely related method of thermal energy atom scattering (TEAS).[14–16] Making use of FAD, the vertical displacements between the different ionic species at the surface, the so-called rumpling, for LiF(001) as well as for MgO(001) surfaces was determined with unprecedented accuracy.[17–19] For a monolayer silica film on Mo(112), the analysis of FAD patterns supported one of two conflicting structural models and provided accurate information on the positions of topmost surface atoms.[10]

The diffraction patterns are closely related to features of grazing scattering from surfaces where the impact of fast atoms with the surface proceeds in terms of surface channeling,[20,21] i.e., a steering of projectiles by surface atoms in terms of small-angle collisions. The diffraction patterns observed in experiments on FAD reported so far are the outcome of scattering along low indexed directions in the surface plane.

In Fig. 1, we display a sketch of the collision geometry and the relevant angles for scattering of fast atoms from an ordered surface. The projectile beam is scattered from the surface under a grazing angle of incidence Φ_{in} with respect to the surface, Γ denotes the azimuthal angle in the surface plane between the beam axis and a low index direction of the crystal. For $\Gamma = 0^\circ$, scattering proceeds in the regime of *axial surface channeling* along low indexed strings formed by topmost surface atoms,[20,21] where the effective interaction potential for atoms with the solid results from an averaging along those strings. This averaged potential shows a corrugation across individual axial channels and forms a one-dimensional grating normal to the direction of the incident beam and the axial channels with periodicity interval $d_\perp$.

Then, in the regime of quantum scattering, peaked intensities at azimuthal angles Ψ are found obeying the Bragg condition n $\lambda_{dB} = d_\perp \sin\Psi$ with $\lambda_{dB} = h/Mv$ being the de Broglie wavelength associated with the center-of-gravity motion of a particle of mass M and velocity v, and h is Planck's constant. Since for scattering of fast light atoms or molecules (H, H_2, He) λ_{dB} is less than two orders of magnitude smaller than typical periodicity lengths $d_\perp$ at surfaces, Bragg peaks are separated by intervals of the azimuthal angle of about 0.1° only. It turns out that in the regime of quantum scattering, such small angular splittings can be resolved.

In most cases, the diffraction patterns observed for scattering of fast atoms from different types of surfaces show streaks, i.e., defined azimuthal splittings elongated in a direction normal to the surface. This observation was attributed to the lack of (longitudinal) coherence in the effective interaction along the axial channel and the beam direction.[1] Similar as for reflection of high-energy electron diffraction (RHEED),[22] i.e., scattering of keV electrons from surfaces under grazing incidence, the streaked structure of diffraction peaks can be ascribed to effects of surface imperfections on coherent scattering. As a consequence, all FAD experiments performed so far were interpreted in terms of *transverse coherence* only, whereas for the motion along the beam, scattering is described classically.[5] Furthermore, it was an open issue as to what extent the periodic corrugation along atomic strings is averaged out in the regime of axial channeling.

In this Rapid Communication, we present data for scattering of fast H atoms from an Al_2O_3 surface where we found evidence that the *longitudinal coherence* is preserved in FAD. This important aspect for quantum scattering of fast atoms from surfaces allows one to resolve the periodicity intervals of the interaction potential normal *and* parallel to the incident beam with a substantial enhancement in resolution by more than one order of magnitude.

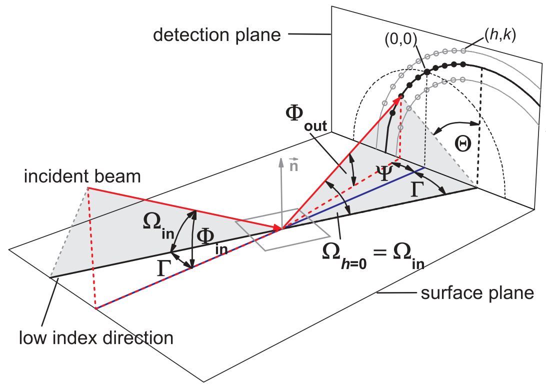

FIG. 1. (Color online) Sketch of collision geometry for grazing scattering of fast atoms from crystal surface (surface plane). Angular distributions are recorded behind target normal to scattered beam (detection plane).

In the experiments, beams of well-collimated 0.3 to 1 keV H atoms were scattered under a grazing angle of incidence Φ_{in} ranging from 0.5° to 1° from a clean and flat $Al_2O_3(11\bar{2}0)$ surface at a base pressure of some 10^{-11} mbar. Angular distributions for scattered atoms were recorded by means of a position-sensitive microchannel plate detector of 48-mm active diameter[13] mounted 0.66 m behind the target. This detector has a detection efficiency of several tens of percent for neutral atoms at energies used here. Since the incident and outgoing projectiles were neutral with beam intensities of some 10^3 atoms per second, effects of charging up and modification of the insulator target were negligible. Most standard surface-science techniques may fail here because of charging problems.

Preparation of the target surface via sputtering and annealing up to temperatures of 1300 K was not successful to obtain a well-ordered surface as manifested by blurred angular distributions.[20] In following preparation procedures as reported in literature,[23,24] we found defined angular distributions for annealing the sample up to temperatures of 2000 K. For this kind of preparation a (12 × 4) superstructure is reported with a surface unit cell formed by basis vectors $\mathbf{a}_1$, $\mathbf{a}_2$ along $[\bar{1}101]$, $[\bar{1}100]$ with opening angle $\beta = 57.5^{\text{o}}$ and $a_1 = 5.12$ Å, $a_2 = 8.24$ Å; i.e. for the (12 × 4) phase, the mesh is 61.4 Å × 33.0 Å.

In Fig. 2, we show patterns as observed with the detector for scattering of 300-eV H atoms ($\lambda_{\text{dB}} = 0.0165$ Å) from $Al_2O_3(11\bar{2}0)$ for an azimuthal rotation of the target with respect to $[\bar{1}100]$ by angles $\Gamma = 0°$ [panel (a)], 3.6° [panel (b)], and 8.1° [panel (c)] under $\Phi_{\text{in}} = 0.92°$, 0.80°, and 0.68°, respectively. As discussed in more detail below, the angular distributions for a rotation of the target from the low index direction, i.e., $\Gamma \neq 0°$, reveal Laue circles of higher orders.

For scattering along the $[\bar{1}100]$ azimuth, i.e., $\Gamma = 0°$, a defined pattern of Bragg peaks is observed where the angular splittings correspond to a periodicity interval of the interaction potential transverse to the $[\bar{1}100]$ channel $d_\perp = a_1 \sin\beta = 4.32$ Å in accord with the surface unit cell.[23,24] The corresponding periodicity length for the reconstructed (12 × 4) phase $d_\perp = 12 a_1 \sin\beta = 51.8$ Å results in splittings

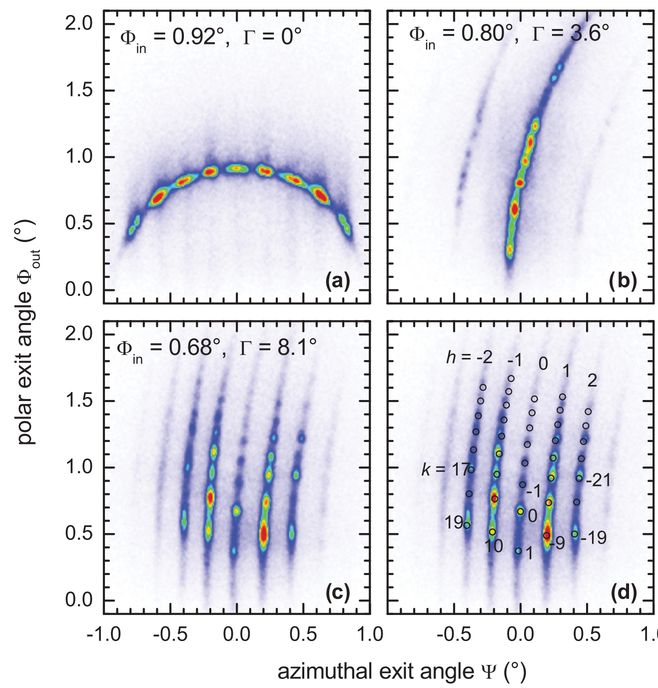

FIG. 2. (Color online) Angular distributions of scattered 300-eV H atoms as recorded with position-sensitive detector 0.66 m behind target. (a) $\Phi_{\text{in}} = 0.92°$, $\Gamma = 0°$; (b) $\Phi_{\text{in}} = 0.80°$, $\Gamma = 3.6°$; (c) $\Phi_{\text{in}} = 0.68°$, $\Gamma = 8.1°$; (d) calculated positions of diffraction spots for $\Phi_{\text{in}} = 0.68°$, $\Gamma = 8.1°$; h, k denote diffraction orders, Γ is azimuthal angle with respect to $[\bar{1}100]$.

for diffraction spots which can not be resolved for $\Gamma = 0°$ (see also discussion following).

The diffraction patterns are located on a defined circle which is different from the vertically streaked patterns observed for scattering of H atoms from other types of surfaces in previous studies using FAD.[3,25] This we ascribe here to a high longitudinal coherence in the scattering process where the circular shape is the outcome of energy conservation for the projectile motion normal with respect to atomic strings in the regime of axial surface channeling.[21]

In close analogy to RHEED or thermal He atom diffraction, this circle defines the zeroth-order *Laue circle* which is present for sufficient longitudinal coherence only. Laue circles of finite orders can not be observed for FAD because their radii are too large in view of small-angle scattering for $\Gamma = 0°$ [cf. Fig. 2(a)]. RHEED experiments have shown that higher-order Laue circles can be efficiently detected for scattering with an azimuthal offset from a low indexed channel.[22] This feature can be also observed for FAD as shown in Figs. 2(b) and 2(c) for $\Gamma = 3.6°$ and 8.1° where the angular interval between Laue circles shrinks with increasing azimuthal angle of rotation. In particular, in the angular distribution for $\Gamma = 8.1°$ [Fig. 2(c)], several orders of Laue circles are present where the defined spots are Bragg peaks owing to the superstructure of the surface which can be resolved under these scattering conditions.

For elastic scattering from the concepts of scattering theory, diffraction spots appear, if the scattering vector (difference of outgoing and incoming wave vectors) in the surface plane coincides with the reciprocal lattice vector $\mathbf{g}_{hk} = h\mathbf{a}_1^* + k\mathbf{a}_2^*$ (*Ewald construction*). For an azimuthal rotation of the target by an angle Γ with respect to $\mathbf{a}_1$, polar and azimuthal angles of diffraction spots Φ_{hk} and Ψ_{hk} follow from[26]

$$\sin\Psi_{hk} = \left[\frac{k}{a_1}\frac{\cos\Gamma}{\sin\beta} - \frac{h}{a_2}(\sin\Gamma + \cos\Gamma\cot\beta)\right]\lambda_{\mathrm{dB}}, \quad (1)$$

$$\cos\Phi_{hk} = \left[\cos\Phi_{\mathrm{in}} + \lambda_{\mathrm{dB}}\frac{k}{a_1}\frac{\sin\Gamma}{\sin\beta} + \lambda_{\mathrm{dB}}\frac{h}{a_2}(\cos\Gamma - \sin\Gamma\cot\beta)\right]\frac{1}{\cos\Psi_{hk}}. \quad (2)$$

In Fig. 2(d), we have indicated positions of diffraction peaks calculated with $12a_1$ and $4a_2$ of the (12×4) superstructure from Eqs. (1) and (2) for the experimental data shown in Fig. 2(c), i.e., $\Phi_{\mathrm{in}} = 0.68^\circ$ and $\Gamma = 8.1^\circ$. Experiment and calculations reveal Bragg peaks up to order $k = 25$ on Laue circles of order $h = -3$ to $+3$. The peak at $(h = 0,\ k = 0)$ denotes specular reflection.

In combining Eqs. (1) and (2) at $\Phi = 0^\circ$, one obtains for the effective radius Ω_h of the Laue circle of order h the longitudinal Bragg condition

$$\cos\Omega_h = \cos\Phi_{\mathrm{in}}\cos\Gamma + \lambda_{\mathrm{dB}}h/d_\parallel \quad (3)$$

with the periodicity interval parallel to $[\bar{1}100]$ $d_\parallel = 4a_2 = 33.0$ Å for the reconstructed $Al_2O_3(11\bar{2}0)$ surface.

For an azimuthal rotation of the target by Γ we derive from Eq. (3) a separation of *Laue circles* by $\Delta\Omega \approx (\sin\Omega_{\mathrm{o}})^{-1}\ \lambda_{\mathrm{dB}}/d_\parallel$ with $\Omega_{\mathrm{o}} = \Omega_{\mathrm{in}} = \sqrt{\Phi_{\mathrm{in}}^2 + \Gamma^2}$ being the incidence angle with respect to a low indexed axial string (cf. Fig. 1). For 300-eV H atoms scattered under $\Gamma = 0^\circ$ and $\Phi_{\mathrm{in}} = 0.9^\circ$, one obtains for the superstructure ($d_\parallel = 4a_2$) $\Delta\Omega \approx 16\lambda_{\mathrm{dB}}/a_2 = 1.8^\circ$. This is clearly beyond the angular spread of beams in the regime of channeling. However, for a rotation of the target by, e.g., $\Gamma = 9^\circ$, one finds $\Delta\Omega \approx 1.6\ \lambda_{\mathrm{dB}}/a_2 \approx 0.18^\circ$. Then, Laue circles of low orders can be detected as demonstrated in Fig. 2(c).

The presence of longitudinal coherence in FAD allows one to resolve periodicity lengths along the scattered beam up to several 100 Å. This transfer width is more than one order of magnitude higher than in previous FAD studies showing transverse coherence only and may even exceed the resolution in scattering of He atoms at thermal energies.[23] As a specific example, we show in Fig. 3 intensities projected onto the radius of Laue circles for scattering of 300-eV H atoms with $\Phi_{\mathrm{in}} \approx 0.7^\circ$ and $\Gamma = 3.6^\circ$, 7.2°, 8.1°, and 9.9°. The experimental splittings in the data are consistent with the value of $4a_2 = 33.0$ Å given in literature.[24] We deduce from Eq. (3) that for constant angles Φ_{in} and Γ, the radii of adjacent Laue circles Ω_h of order h scale as $\cos\Omega_{h+1} - \cos\Omega_h = \lambda_{\mathrm{dB}}/d_\parallel$. From the linear dependence by plotting this difference as function of λ_{dB} we derive from data for different projectiles energies $d_\parallel = (33.3 \pm 0.8)$ Å. A more refined analysis concerning the very complex structure of the reconstructed Al_2O_3 surface would comprise the intensities of diffraction spots and is beyond the scope of this work.

The azimuthal rotation of the target leads also to a dispersion of Bragg peaks on a given Laue circle which amounts for a range of angles around specular reflection to an angle $\Delta\Theta(\Gamma) = \cos\Gamma\ \sqrt{1 + \tan^2\Gamma/\sin^2\Phi}\ \Delta\Theta(\Gamma = 0^\circ)$. Then, the splittings between Bragg peaks for $\Gamma = 8.1^\circ$ are enhanced compared to $\Gamma = 0^\circ$ by a factor of about 10 and the Bragg peaks for the superstructure can be resolved [cf. Figs. 2(b) and 2(c)].

This feature is illustrated in Fig. 4 by the projection of intensities on Laue circles with radius Ω_h for the data shown in Fig. 2(c) ($\Phi_{\mathrm{in}} = 0.68^\circ$, $\Gamma = 8.1^\circ$). In the plots of intensities as function of angle Θ for three Laue circles of orders $h = -1$ (left panel), $h = 0$ (middle panel), and $h = +1$ (right panel), individual Bragg peaks for the superstructure are clearly

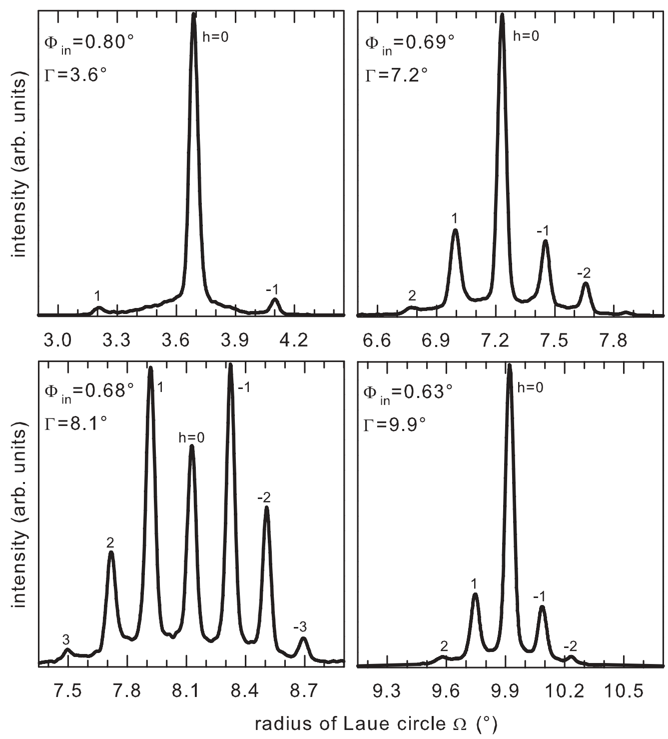

FIG. 3. Intensity projected on radius of Laue circle in terms of angle Ω for scattering of 300-eV H atoms from $Al_2O_3(11\bar{2}0)$ surface. Upper left panel: $\Phi_{\mathrm{in}} = 0.80^\circ$, $\Gamma = 3.6^\circ$; upper right panel: $\Phi_{\mathrm{in}} = 0.69^\circ$, $\Gamma = 7.2^\circ$; lower left panel: $\Phi_{\mathrm{in}} = 0.68^\circ$, $\Gamma = 8.1^\circ$; upper right panel: $\Phi_{\mathrm{in}} = 0.63^\circ$, $\Gamma = 9.9^\circ$. h denotes order of Laue circle.

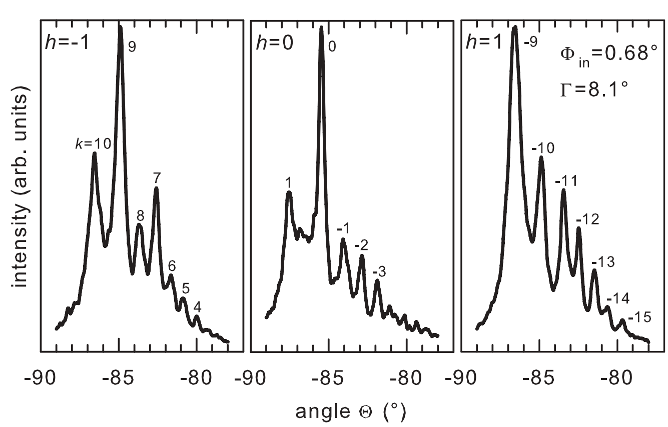

FIG. 4. Intensity projected on Laue circle of order $h = -1$ (left panel), $h = 0$ (middle panel), and $h = +1$ (right panel) for scattering of 300-eV H atoms from $Al_2O_3(11\bar{2}0)$ surface under $\Phi_{\mathrm{in}} = 0.68^\circ$ and $\Gamma = 8.1^\circ$. k denotes order of Bragg peaks.

resolved which is not achieved for $\Gamma = 0^\circ$ [cf. Fig. 2(a)]. From fits of Gaussian line shapes to the data, we obtain in the present state of analysis of data $d_\perp = (50.1 \pm 3.5)$ Å. This value is in good agreement with a periodicity length of 51.8 Å, which follows from the unit cell of the reconstructed surface.

In conclusion, we have demonstrated in experiments on diffraction effects for fast H atoms scattered from a well-ordered $Al_2O_3(11\bar{2}0)$ surface under a grazing angle of incidence that the longitudinal coherence in the collision with the surface persists as manifested by the presence of defined Laue circles. Compared to similar previous studies,[25] showing transverse coherence only, the resolution of periodicity intervals (transfer width) can be enhanced by more than one order of magnitude. Thus, this observation can be considered as an important contribution for establishing the diffraction of fast atoms as a powerful analytical tool in surface science. Since it was believed that the corrugation of the potential along the direction of the fast atoms is averaged out, the effect of the longitudinal periodicity of the potential in the diffraction patterns came very surprising. We hope to stimulate theoretical work towards a deeper understanding of this feature.

We thank the Deutsche Forschungsgemeinschaft DFG (Wi 1336) for financial support and K. Maass, J. Sölle, and G. Lindenberg for their assistance in the preparation of the experiments.

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