Ion-induced low-energy electron diffraction

T. Bernhard, Z. L. Fang, and H. Winter*

Institut für Physik, Humboldt Universität zu Berlin, D-12489 Berlin-Adlershof, Germany.

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H^+ and He^+ ions with an energy of 25 keV are scattered under a grazing angle of incidence from a clean and flat Cu(001) surface. For specific azimuthal orientations of the crystal surface with respect to low index directions in the surface plane we observe the ion induced emission of electrons with a conventional LEED (low energy electron diffraction) setup. By operating the instrument in an energy dispersive mode we find intensity distributions of emitted electrons which can unequivocally be ascribed to diffraction effects at the target surface.

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The historical experiment on diffraction of low energy electrons at a well ordered crystal surface by Davisson and Germer [1] was an important contribution to the development of quantum mechanics, which provided support for the concept of matter-waves proposed by de Broglie [2]. Nowadays the diffraction of low energy electrons (LEED) is one of the standard tools in surface physics for the characterization of periodic structures at crystal surfaces. Over the years, technical and conceptual developments of LEED have approached a high level of sophistication for the study of complex structures at surfaces and allow one to characterize defects of crystal surfaces in great detail [3,4].

Basic concepts of LEED comprise defined inital states for incident particles, i.e., collimated beams of electrons with well-defined energies in the 100 eV domain, equivalent to de Broglie wavelengths of typically Ångstroms (10^{-10} m). Detection of elastically scattered electrons results furthermore in defined final states in the collision with the crystal surface. These specific features form the basis of the success of this method: relatively simple use and a diffraction pattern which can be analyzed in terms of established concepts of scattering theory; standard and sophisticated LEED instruments are commercially available.

Coherent scattering for electron is also found for other excitation mechanisms as, e.g., for photoemission [5]. Of particular interest for the present work is the recent debate on electron diffraction effects on angle resolved energy spectra for ion induced electron emission from crystal surfaces [6,7]. In those spectra features are present at electron energies of typically 10 eV which were first interpreted in terms of excitation and subsequent decay of surface and bulk plasmons [8–10]. In conflict with this explanation are in specific cases relatively intense peaks and their pronounced energy shift with angle of detection [6,7] as well as the presence of those features at low ion energies, where the direct excitation of plasmons by ions is not possible owing to insufficient momentum transfer [11]. In further experimental studies on this problem, it was revealed that most spectral features disappear for a polycrystalline target [6].

*Author to whom correspondence should be addressed. Electronic address: winter@physik.hu-berlin.de

As a result, the structures in electron spectra were alternatively attributed to diffraction effects for electrons excited by impinging ions. A first quantitative estimate on ion induced diffraction effects for low energy electrons was recently presented by Niehaus and co-workers [7] for grazing scattering of protons from Al(111) and Cu(110) surfaces. By considering Bloch waves excited by ions in specific directions parallel to the crystal surface, enhanced intensities in electron spectra could be interpreted by a coherent scattering process.

In referring to conventional LEED, a clear demonstration of such electron diffraction phenomena for ion induced electron emission would be the presence of diffraction spots in the angular distributions of scattered electrons which are directly related to the two-dimensional reciprocal lattice of a crystal surface. In this paper we report on evidence for the presence of peaks in the angular distributions of electrons induced by ion impact on a crystal surface. Based on simple concepts of classical scattering theory, these peaks can be attributed to specific reflexes for electron diffraction at the well-ordered structure of a Cu(001) surface.

In our experiments 25 keV H^+ and He^+ ions are scattered under a grazing angle of incidence $\Phi_{in}=1.6°$ from a clean and flat Cu(001) surface. The surface is prepared by cycles of grazing sputtering with 25 keV Ar^+ ions and subsequent annealing at temperatures of about 770 K. The angular distributions of electrons ejected by ion impact are recorded using a commercially available Spot Profile Analysis LEED system (SPA-LEED) [12,13]. This instrument allows one to obtain angular distributions of electrons emitted normal to the surface via deflection to the small aperture (0.1 mm) of a channeltron detector by means of an electric octupole field. In our measurements the setup is used for detection of electrons only, and the low energy electron gun is switched off. A suppressor electrode in the detector unit—installed for reducing contributions of inelastically scattered electrons in conventional LEED—provides the operation of the system as a high pass filter for energies of detected electrons. The acceptance cone of the entrance aperture of the instrument for emitted electron amounts to about ±25°. In the conventional mode of operation the angle for incident electrons with wave vector $\vec{k}_{in}$ and for elastically scattered electrons with $\vec{k}_{out}$ is varied by the electric octupole field resulting in the scanning

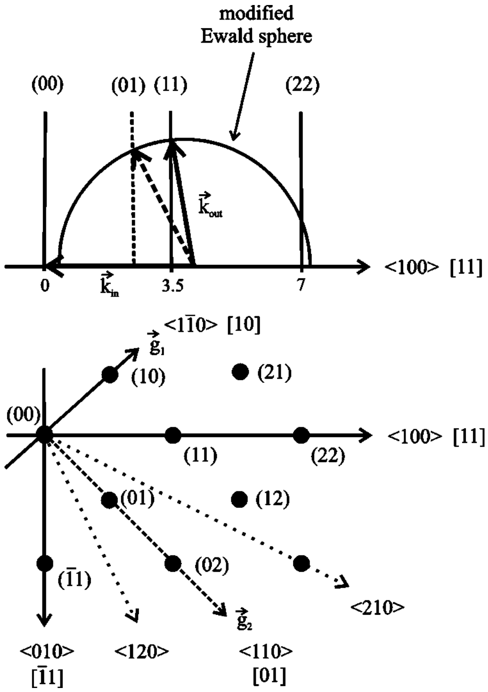

FIG. 1. Ewald construction in reciprocal lattice space. Upper panel, plane normal to surface; lower panel, plane parallel to surface. Owing to surface potential, momentum vectors k_{in}, k_{out} are different in length. Solid lines and arrows hold for direction along ⟨100⟩, etc., dashed lines and arrow for ⟨110⟩, etc., dotted lines ⟨120⟩, etc.

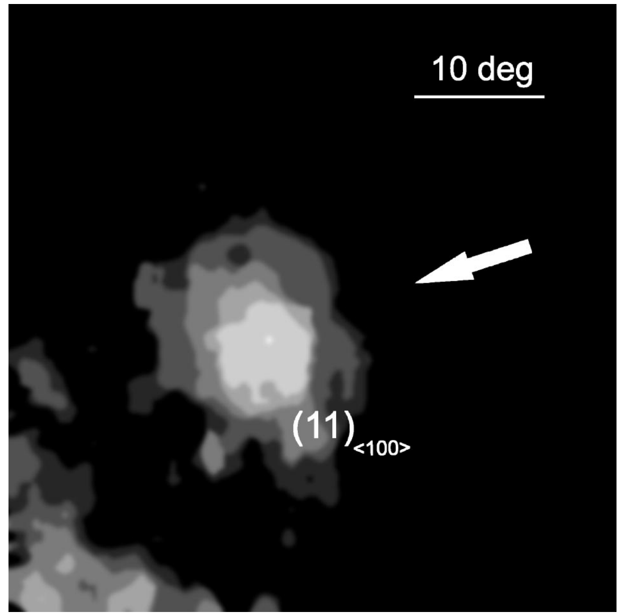

FIG. 2. Difference of 2D-intensity distributions recorded for electron energies of 52 eV and 57 eV with SPA-LEED for 25 keV protons scattered under $\Phi_{in}=1.6°$ from Cu(001) along ⟨100⟩. Arrow indicates direction of incident beam, tip of arrow direction normal to surface. Lower gray-scale level (black) at about 50% of maximum intensity.

of the scattering wave vector $\vec{K}=\vec{k}_{out}-\vec{k}_{in}$. From the slight separation of the apertures of electron gun and detector results a fixed offset angle of about 8° from a perfect antiparallel orientation of the wave vectors for incident and elastically scattered electrons.

Low energy electrons are excited during grazing ion scattering from the target and are detected with respect to energy and angle; i.e., $\vec{k}_{out}$ is scanned by means of the SPA-LEED instrument. Making use of the energy suppressor unit allows us to separate a specific interval of energies for scattered electrons by measurements at two different energies and taking the difference of the data. This feature is crucial here, because initial energies of electrons produced via ion impact are much poorer defined than in a conventional LEED experiment using electrons from a low energy electron gun. Therefore the energy of "elastically" scattered electron has to be fixed here in the exit channel.

The angular positions of diffraction spots for scattered electrons can be derived from concepts of scattering theory by means of an Ewald construction in reciprocal lattice space [14]. Electrons with initial momentum vectors $\vec{k}_{in}$ oriented along the direction of the incident ion beam are preferentially excited for grazing impact [15]. In this respect we follow suggestions on the production of Bloch waves parallel to the surface by Niehaus and co-workers [7]. This feature results in specific diffraction pattern for elastically scattered electrons that clearly differ from those obtained for large angle impact using an electron gun. Owing to the surface potential ($V\approx 12$ eV for Cu(001) [16]) initial and final momenta change from (atomic units are used) $k_{in}=\sqrt{2(E_e+V)}$ to $k_{out}=\sqrt{2E_e}$, with E_e being the final electron energy (see modified Ewald construction in Fig. 1).

Figure 1 shows a sketch of a plane in reciprocal lattice space normal (upper panel) and parallel (lower panel) to the surface with the simple square unit cell $\vec{g}_1=\sqrt{2}/a\times[10]$ and $\vec{g}_2=\sqrt{2}/a\times[01]$ with $a=3.61$ Å being the lattice constant of a copper crystal and $[10],[01]$ chosen along $\langle 1\bar{1}0\rangle,\langle 110\rangle$ of the fcc lattice. The solid vertical bars illustrate the Laue conditions for the (00), (11), and (22) reflexes (g_1,g_2). Striking feature of the construction is the direction of the (11) intensity spot about normal to the surface for the settings of our experiments; i.e., a Cu(001) surface and electron energies of some tens of eV (55 eV chosen in Fig. 1). This angular range for diffraction spots is favorable for the realization of experiments, since the LEED instrument can be installed to detect electrons within a solid angle centered along the surface normal and can be mounted directly on top of the target surface. Furthermore, predominant forward scattering of electrons excited by ion impact under grazing incidence [15] reduces the background from inelastically scattered electrons.

In Fig. 2 we display a LEED pattern obtained with 25 keV H^+ ions scattered under $\Phi_{in}=1.6°$ along the low index ⟨100⟩ azimuthal direction in the (001) plane. For electron energies between 52 and 57 eV we find a pronounced intensity spot at $12°\pm 2°$ from the surface normal in direction of the scattered beam (marked by arrow, tip indicates surface normal). The position of the diffraction spot is in accord with $11°\pm 1°$ derived from the construction sketched in Fig. 1. The angular and energy spread of electrons in the initial excitation via ion impact are considered as the main mecha-

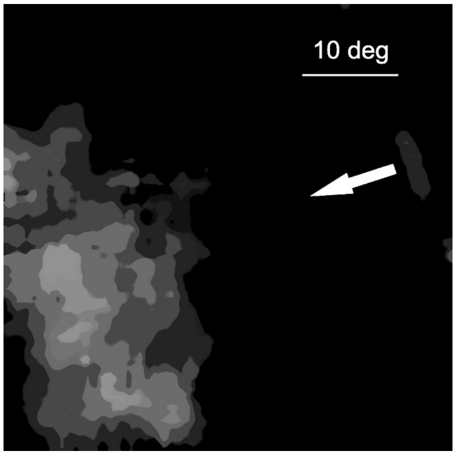

FIG. 3. Same as Fig. 2 for scattering along ⟨110⟩.

nism for a substantial blur compared to bright and sharp spots in conventional LEED. The signal in the lower left corner is attributed to enhanced (incoherent) forward scattering.

In Fig. 3 we display data for scattering along ⟨110⟩. The Laue condition for this case is sketched in Fig. 1 by a dashed bar and momentum vector $\vec{k}_{out}$. The (01) spot for 55 eV electrons should appear under an angle of 29.7° with respect to the surface normal which is at the limit of the angular acceptance of our setup. The intensity pattern present in the center of Fig. 2 has disappeared, but a weak structure is observed in the lower left corner which we partly attribute to the (01) spot, background from preferential forward scattering, and problems of imaging with SPA-LEED at larger acceptance angles [17].

For the same azimuthal orientation of the target we recorded intensity distributions for lower electron energies. Figure 4 shows the difference of distributions for 20 eV and 27 eV obtained with 25 keV He^+ ions (similar results are obtained with H^+ ions). The data reveals a diffraction spot shifted by about 10° from the normal direction (tip of arrow) which compares well with the estimate from the Ewald construction. Of specific interest are two further spots found symmetrically with respect to the central spot and the ⟨110⟩ direction of the incident beam. The positions of these spots are in accordance with coherent scattering along the ⟨210⟩ and ⟨120⟩ azimuth. This observation might be explained by the feature of enhanced secondary electron scattering in crystals along low index directions owing to momentum matching in "interzone transitions" [18]. A more detailed discussion on this new type of information on ion-solid interactions available from the analysis of electron diffraction effects will be presented in a forthcoming paper.

In passing we note that no indication for a diffraction pattern is observed for ion scattering from part of the target holder made from polished polycrystalline stainless steel with no long-range monocrystalline order (see also Ref. [6]).

A direct demonstration of diffraction effects on ion induced electron spectra is performed in experiments where the SPA-LEED instrument is replaced by an electron spectrometer (CLAM2, VG-instruments) with its angular acceptance

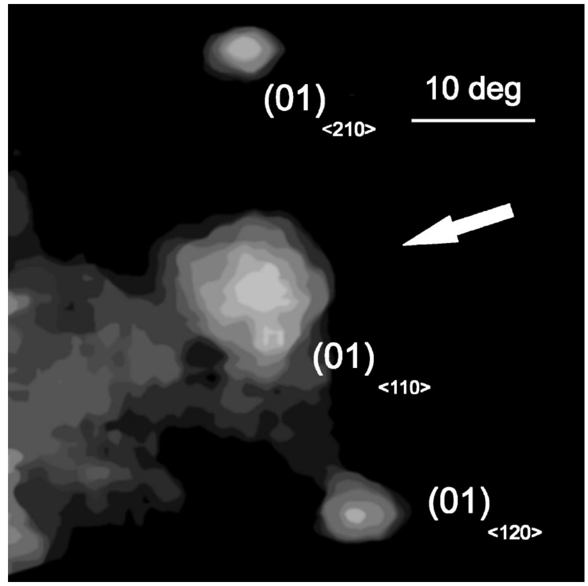

FIG. 4. Difference of two-dimensional intensity distributions recorded for electron energies of 20 eV and 27 eV with SPA-LEED for 25 keV He^+ ions scattered under $\Phi_{in}=1.6°$ from Cu(001) along ⟨110⟩.

along a direction tilted by 12° with respect to the surface normal, i.e., the direction of the (11) spot for scattering along ⟨100⟩. The energy spectra shown in Fig. 4 reveal a pronounced broad peak at about 55 to 60 eV for scattering of 25 keV protons under $\Phi_{in}=1.6°$ along ⟨100⟩, whereas for scattering along ⟨110⟩ (peak is expected here at about 15 eV) and "random" directions (±38° from ⟨100⟩) this peak disappears and only a weak structure in the spectra at "random" ascribed to *CVV* Auger electrons from the Cu surface is present.

In summarizing, the experimental data presented in our work gives clear evidence and support for the presence of diffraction effects in ion induced electron emission. Assum-

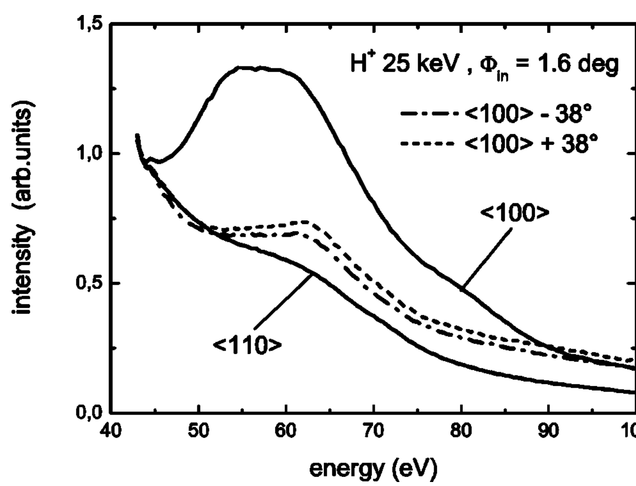

FIG. 5. Electron spectra recorded with CLAM2 spectrometer for scattering of 25 keV protons under $\Phi_{in}=1.6°$ from Cu(001). Solid curves, ion beam incident along ⟨100⟩, ⟨110⟩; dashed curve, along ⟨100⟩+38°; dashed-dotted curve, ⟨100⟩−38°.

ing the preferential excitation of electrons along the incident beam direction (primarily in direct binary encounter with projectiles), the diffraction pattern can be deduced from a modified Ewald construction. The unambiguous demonstration of the presence of diffraction effects in ion induced electron emission has important implications for this field.

Electron spectra recorded for the impact of ions on crystal surfaces are affected in a substantial manner. Structures or peaks in those spectra may originate from basic physical processes as, e.g., Auger transitions or plasmon decay, however, they can also stem from diffraction effects at crystal surfaces (cf. Fig. 5). In this respect, our work removes the uncertainty in the presence of those effects concluded from previous studies based on electron spectroscopy only. This also implies that available experimental data has to be checked with respect to effects caused by electron diffraction.

Making use of diffraction effects allows one to investigate coherent scattering of electrons excited by ion impact from a detailed analysis of the diffraction spot profiles. This provides detailed new studies on electron emission processes, since defined initial states of excited electrons can be fixed by the diffraction conditions.

We agree that the SPA-LEED instrument used has its limitations for the type of work reported here, but it was sufficient for a first direct demonstration of ion induced LEED effects. In this respect we attribute a fair amount of the broad angular widths of the observed spots (about 5°) to the instrumental resolution. An optimized detection scheme adopted to this regime of electron emission as, e.g., position and time-of-flight resolved detection with larger solid angle should be employed in future studies on this elementary process concerning ion-solid interactions.

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