Intensity asymmetry of the (00) diffracted spin-polarized electron beam scattered from W(110): Azimuthal dependence

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Intensity asymmetry of the elastically scattered spin-polarized electrons from W(110) surface at 22 eV primary energy and at 25° angles of incidence and detection ((00) diffraction beam) was measured as a function of the azimuthal angle. Experimental results are compared with the calculations based on the relativistic multiple scattering formalism. The comparison showed fairly good agreement. These support the general theoretical approach and the input data chosen for the calculations. The information gained from our work is important for the interpretation of results of various spin-polarized electron spectroscopies as well as for design and construction of multi-layered structures with spin-active interfaces. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4812751]

Recently discovered spin-split surface states on Au(111)[1] and spin-dependent surface resonances on W(110) with Dirac cone dispersion[2,3] attracted again attention of researchers[4,5] to the structural and electronic properties of high-Z nonmagnetic surfaces that exhibit an imbalance of spin-up and spin-down states in the phase space.

One of the powerful instruments for studying intricate structural and electronic properties of surfaces is interaction of spin-polarized electrons with solid surfaces.[6,7] It was successfully applied for studying magnetic and nonmagnetic surfaces.[7] One of the techniques is based on the analysis of elastically scattered electrons. In case of crystal surfaces, it turns out to be Low Energy Electron Diffraction (LEED).

There are four basic modes of LEED application for a surface structural analysis: (i) fixed energy LEED pattern distribution analysis; (ii) I-V curve analysis or energy profile of a diffraction pattern; (iii) intensity analysis of the (00) diffraction beam at fixed primary energy as a function of the incidence angle (rocking curves); (iv) intensity analysis of the (00) diffraction beam at fixed primary energy and fixed angle of incidence as a function of the azimuthal angle (rotation curves).

When spin-polarized electrons are used, a new variable *electron spin* appears. This obviously extends information that can be extracted from measurements. In general, this means that a more precise picture of the electron scattering can be drawn and more rigorous structural analysis can be performed. The "rocking curves" and "rotation curves" techniques then have two modifications each.

In the first one, an unpolarised electron beam is scattered from the surface, and the polarization P of the elastically scattered electrons is measured as a function of the incidence angle ("rocking curves") or as a function of the azimuthal angle ("rotation curves"). In the second modification, a spin-polarized beam is scattered from a crystal surface and its intensity is measured for two opposite polarizations of the incident beam: I^+ and I^-, then asymmetry A can be calculated: $A = (I^+ - I^-)/(I^+ + I^-)$. The asymmetry A is measured then as a function of the incident angle ("rocking curves") or as a function of the azimuthal angle ("rotation curves").

In fact, there is a relationship between these two modifications. Namely, due to the time inversion symmetry there must be reciprocity between the asymmetry and the spin polarization measurements. It means that if a 100% spin-polarized electron beam is used and the asymmetry of elastic scattering is A then in a similar scattering experiment of unpolarised electrons the value of polarization P of scattered electron beam would be equal to the value of the asymmetry: $P = A$ in proper spatial symmetric conditions.[8,9]

For the last few decades tungsten crystals, mostly of W(100), W(110), and W(111) faces, have been used as bench marks in spin-polarized electron scattering experiments. It was used as a sample in the pioneering double scattering experiment by Kirschner and Feder,[10] it is used in spin detectors based on electron diffraction that is spin-sensitive. Theoretical approach for the description of spin-dependent low energy electron diffraction was tested on tungsten crystal.[9,11] First experiments on the correlated electron pair emission excited by non-polarized and spin-polarized electrons were performed on tungsten crystal.[12,13] The reasons for that are: (i) large Z, which means large spin-orbit effects that are easy to observe; (ii) possibility to prepare really clean and well-ordered crystal surface and ability to reproduce such a surface; (iii) choice to use reconstructed or non-reconstructed surface; (iv) surface of tungsten crystal is the ideal substrate for a deposition of variety of metallic, semiconductor, and insulator films.

The W(110) surface is of a particular interest because it remains unreconstructed after the cleaning procedure, possesses spin-polarized surface resonances, has two-fold symmetry and potentially more features in rotation curves.

There was an extensive experimental and theoretical work on the spin polarization of elastically scattered

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electrons from W(110) and W(001), where rotation polarization curves were measured.[14,15] That was the first application of Spin-Polarized LEED (SPLEED) for quantitative structural analysis. It was demonstrated that this analysis is very suitable for low-energy primary electrons (<80 eV). It was suggested to extend such analysis down to very low energies but above strong surface barrier resonance below 15 eV.

In our previous work,[16] we used spin-polarized incident electrons to study intensity and asymmetry profile of the (00) diffraction beam scattered from W(110) in the energy range 8–23 eV and at the incident angle of 25°. We found a prominent asymmetry feature of about 60% slightly below the emergence threshold energy for two nonspecular beams. The calculations on the basis of relativistic multiple-scattering formalism reproduced rather well the experimental counterpart.[16] This profile was measured within the scattering plane that contained [100] direction along the sample surface. Beside resonance-like feature in the asymmetry profile at about 13 eV, there was a region of energies around 21 eV and above with almost zero asymmetry indicating that at these energies the elastic scattering cross sections for spin-up and spin-down electrons are equal. We decided to measure azimuthal dependence of the asymmetry (*rotation curves*) at the energy 22 eV using the same geometry of scattering as in the case of the profile measurements.[16]

Our experimental setup is depicted in Fig. 1. The measurements were performed in UHV conditions, with the pressure in the 10^{-11} Torr range. Spin-polarized electron gun is based on the photoemission from strained GaAs photocathode activated by repetitive deposition of Cs and adsorption of oxygen in order to get a negative electron affinity of the surface.[17] A semiconductor diode laser with the wavelength of 833 nm was used for the photoelectron excitation. A liquid crystal retarder converted linearly polarized light into circularly polarized light and controlled the helicity of polarization and hence, the spin polarization of photoelectrons. Originally longitudinally polarized beam of photoelectrons was bended by a 90° spherical deflector and became transversally polarized with the spin polarization vector perpendicular to the scattering plane. The degree of polarization was measured in a separate experiment and is estimated to be (50 ± 2)%.

Incident polarized electrons impinged onto the sample surface at the angle of 25 ± 0.5° with respect to the normal to the surface and elastically scattered electrons ((00) diffracted beam) were detected at the angle of 25 ± 0.5°. We used a laser light reflection from the sample surface to estimate the change of the polar angle when the azimuth changes. Our analysis shows that the maximum wabbling angle could be up to one degree. Scattered electrons are detected by position sensitive detector base on two 75 mm in diameter micro-channel plates in chevron configuration with resistive anode. Energy distributions of scattered electrons were measured using Time-of-Flight (TOF) technique.[18] To enable this type of energy measurements, the incident beam was pulsed to define a reference point on the time scale. The pulse width was about 800 ps and the repetition rate was 4 MHz. An example of two energy distributions measured for two opposite polarizations of the incident beam is shown in Fig. 2. The width of the elastic maximum is about 0.5 eV that determines the energy resolution at 22 eV. (In the TOF method of electron energy measurements, the energy resolution depends on the energy to be measured and is better for lower energies). The W(110) crystal was cleaned using well established cleaning procedure.[19,20]

Azimuthal position of the sample was determined by the angle between the normal to the scattering plane (that coincides with the Y axis) and the $[1\bar{1}0]$ direction in the surface of the W(110) crystal sample. Uncertainty of the azimuthal position of the sample was 0.5°. Fig. 1 shows the zero azimuth position of the sample. Two spectra were recorded for every azimuthal position of the sample: (1) for the incident beam polarization “spin-up” and (2) for the incident beam polarization “spin-down.” For this purpose, the polarization vector **P** was oriented “up” (along Y axis) or “down” (“negative” direction of Y axis), and its orientation was changed every five second to avoid any artefacts of asymmetry related to the electron beam instability or gradual contamination of the surface. Accordingly two files were stored in the computer: one for “spin-up” and another for “spin-down” primary beam. Using position-sensitivity of the detector, we were able to reduce a contribution of diffusely (quasi-) elastically scattered electrons by adsorbed particles, imperfections of the surface and by phonons, which would

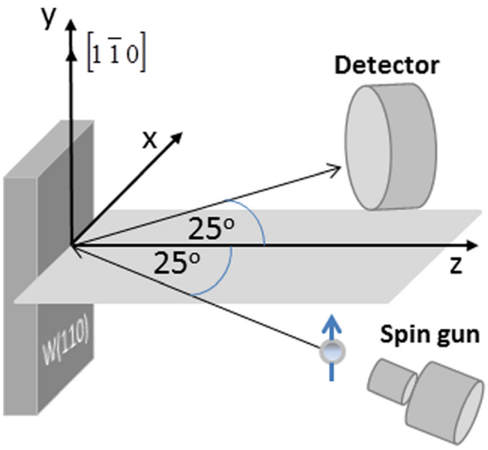

FIG. 1. Geometry of the experiment. The shown azimuthal position of the sample corresponds to the azimuthal angle “zero,” i.e., crystal axis $[1\bar{1}0]$ is parallel to the “Y” axis.

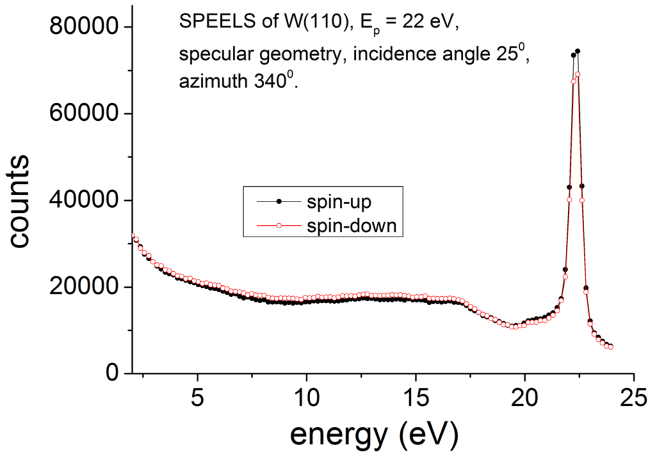

FIG. 2. Energy distributions of scattered from W(110) electrons in specular geometry for spin-up and spin-down primary beam.

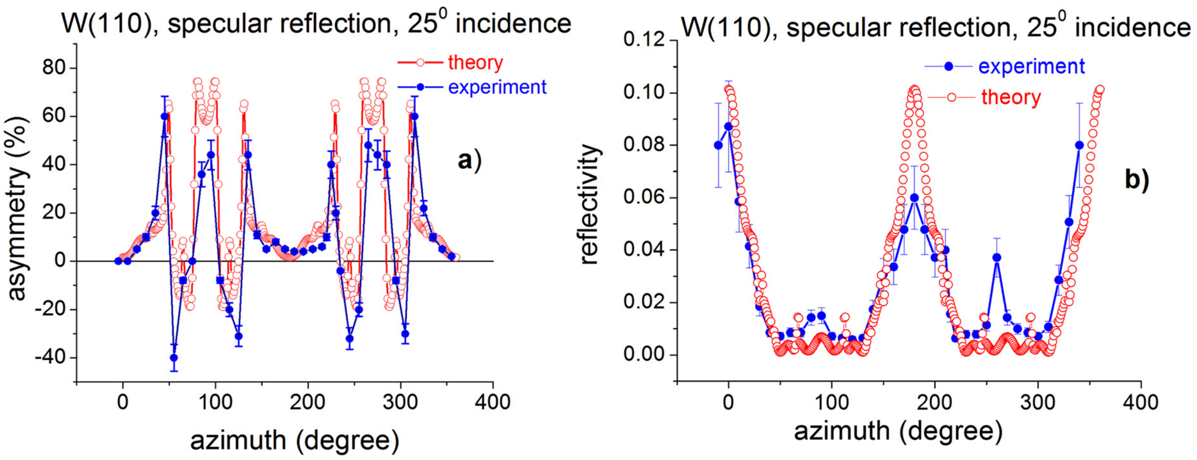

FIG. 3. Asymmetry of (00) diffraction beam as a function of azimuthal position of the sample; experiment and theory—(a); normalised intensity (reflectivity) of (00) diffraction beam as a function of azimuthal position of the sample; experiment and theory—(b). Asymmetry spectrum is normalised by the polarization of the incident beam, and the reflectivity curve is scaled (multiplied by scaling factors) for comparison with theory.

reduce the measured spin asymmetry. In order to achieve that we selected on the detector a small (R = 10 mm) area around the diffraction spot and processed only electrons detected within this area. The amplitude of the elastic maximum (sharp maximum on Fig. 2) has been taken as the intensity I of the (00) beam, and the asymmetry in this approximation was calculated as usual: $A = (I^+ - I^-)/(I^+ + I^-)$, where I^+ is intensity for “spin-up” incident beam and I^- is intensity for “spin-down” incident beam.

SPLEED calculations were performed by means of a relativistic multiple scattering formalism, which has been presented in detail in Ref. 7 and applied successfully to many surface systems (cf., e.g., Refs. 16 and 21, and references therein). As a prerequisite, we calculated the electronic structure of the ground state of W(110) by means of an *ab initio* Full-Potential Linear Augmented-Plane-Wave (FLAPW) method.[22] Using a local density approximation (LDA) for the exchange-correlation energy,[23] we applied this method to a W(110) film consisting of 11 monoatomic layers, with the first interlayer spacing reduced by 3% relative to the bulk interlayer spacing on the grounds of LEED analyses.[14,24,25] We thereby obtained, in particular, a real one-electron potential, which we used to construct the complex quasi-particle potential input needed for calculating the SPLEED intensity and asymmetry. For the imaginary part V_{0i} of the quasi-particle potential we used, as in Ref. 16, $-0.1(E + \phi)^{0.83}$ eV, where E denotes the incident electron energy and ϕ the work function, which for W(110) is $\phi = 5$ eV. The surface potential barrier with image asymptotic was chosen as described in detail in Ref. 16.

Fig. 3(a) presents a comparison between measured and calculated intensity asymmetries of elastic scattering of spin-polarized electrons from W(110) at fixed primary energy of 22 eV, fixed incidence angle of 25° and varying azimuthal angle. The comparison looks fairly well except for two angles of 90° and 270°, where calculations show higher asymmetry than experiment. The possible reason of such discrepancies might be an imperfection of the sample crystal, for example, or wabbling of the sample while changing the azimuthal angle. As above mentioned, the change of polar angle may be up to 1° when the azimuthal angle changes. Additional calculations actually revealed that the value of asymmetry at 90° and 270° reacts very sensitively to small changes of the polar angle.

The asymmetry rotation curves reflect the 2 *mm* spatial symmetry of the W(110) surface: period of 180° and mirror symmetry with respect to 0°, 90°, 180°, and 270°. This mirror symmetry dictates that at each of these azimuthal angles the curves have either a (local) maximum or minimum. Whether there is actually a pronounced minimum (like at 0° and 180°), a maximum, or a local minimum adjacent to two maxima (like in the theoretical curve at 90° and 270°), depends sensitively on the energy and the polar angle. This appears plausible from the physical origin of the spin-up and spin-down intensities, from which the asymmetry is obtained. For an electron impinging on the surface, there are first scattered spherical waves from single atoms, the amplitudes of which are spin-dependent due to spin-orbit coupling. These waves are re-scattered spin-dependently from other atoms and so on (multiple scattering). The amplitude of the outgoing plane wave (the diffracted beam) is the sum over the amplitudes from a large number of multiple scattering paths, i.e., there is very complex interference. Whether it is predominantly constructive or destructive, depends sensitively on energy and angles, and so does the spin dependence of the outgoing amplitude and thence the intensity and, consequently, the asymmetry.

Fig. 3(b) shows normalised intensity (reflectivity) of the (00) diffraction beam as a function of the azimuthal angle. Experimental curve was measured as the detector’s total count rate as a function of azimuthal angle. That includes all electrons: elastically and non-elastically scattered from W(110) in the specular direction. Then the background related to the inelastically scattered electrons was subtracted and spectrum was scaled (multiplied by a scaling factor) to compare the shape of the curve with the calculated one. It is seen from Fig. 3(b) that the main features of the experimental curve are very well reproduced by calculations. The intensity rotation curves show also the two-fold symmetry (180° period).

In conclusion, we have measured and calculated using relativistic multiple scattering formalism the rotation curves for the (00) diffraction beam from W(110) at 25° polar angle and 22 eV primary energy. Fairly good agreement between experiment and calculations supports input parameters of the theory and general theoretical approach. Given that the same parameters were used for calculating the energy profile of the (00) beam from the same surface of W(110),[16] we conclude that the chosen parameters of the scattering potential and the model of the surface potential barrier are correct. We think that above presented results can help in the interpretation of the azimuthal dependence of the spin-polarized secondary emission spectra from W(110) as well as spin-polarized (e,2 e) spectra of W(110). Indeed, since EELS spectra of a crystal sample in specular geometry result from the scattering combination (elastic + inelastic) or (inelastic + elastic), the azimuthal dependence of the elastic step of these two-step processes will influence the whole energy loss spectrum. Assuming the reciprocity of the spin polarization and the asymmetry one can expect that at the primary energy 22 eV of unpolarised incident beam and proper choice of scattering angles the polarization of scattered electrons can reach up to 60% (see Fig. 3(a)). Moreover, this polarization can be easily controlled by changing the azimuthal position of the sample.

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