Spin-orbit effects in the (e,2e) scattering from a W(110) surface and thin gold layer

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Abstract. Spin-effects were studied in the (e,2e) scattering from W(110) single crystal and thin gold layer on W(110). Intensity asymmetry observed in the spectra using spin-polarised incident beam were identified as spin-orbit interaction in the valence electron states involved in the scattering with the contribution of orbital momentum of incident electrons.

1. Introduction

Incident kinetic electrons impinging on a solid surface can be scattered elastically and inelastically from the surface. The energy distribution of scattered and ejected electrons forms a secondary emission spectrum, which consists of: i) a sharp maximum of elastically scattered electrons, ii) a part of the spectrum that contains electrons scattered with some energy loss, and finally iii) a broad and almost featureless maximum of “true secondary” or “ejected” electrons. In conventional single-electron spectroscopy all these parts of the spectrum are analyzed separately and independently. However there might be an internal relationship between parts ii) and iii). This relationship can be revealed if two electrons resulting from the scattering of an incident electron from a surface are detected and analyzed. This type of experiment is referred to as (e,2e) scattering experiment [1,2] and the electron spectroscopy involving detection and analysis of two electrons represents a simplest example of a multi-electron spectroscopy – two-electron spectroscopy of surfaces. When the incident electron beam is spin-polarized then two spectra can be recorded: i) (I^+) - with the polarization vector of the primary beam parallel to the quantization axis and ii) (I^-) – with polarization vector anti-parallel to the quantization axis. We chose the quantization axis to be perpendicular to the scattering plane which contains the normal to the surface and two detectors (figure 1).

In the case of a heavy metal (for example, W or Au) the spin-orbit interaction in the incident electron state inside the sample as well as the spin-orbit interaction in the valence electron state of the sample can be visualized by the spin-polarized two-electron spectroscopy. It allows to study the spin-dependent electron scattering dynamics as well as spin-dependent electronic structure of surfaces.

In this paper we present a comparative analysis of spin-dependent features in the (e,2e) spectra of the W(110) crystal and Au(111) film on the W(110) substrate.
2. Experimental details

Experimental set-up consists of a scattering chamber and a spin-polarized electron source chamber connected through a valve. In both chambers the vacuum is maintained in the $10^{-11}$ Torr range. The scattering chamber is equipped with instruments for preparation and characterization of surfaces and thin films such as: Low Energy Electron Diffraction (LEED), Auger Electron spectrometer, ion sputtering gun, electron beam evaporators, mass-spectrometer, and quartz microbalance.

![Figure 1](image.png)

**Figure 1.** Geometry of the experiment. R1 and R2 are the symmetry planes. The $[1\bar{1}0]$ direction of the crystal runs along the y axis.

The geometrical arrangement of the experiment is shown in figure 1. The (e,2e) spectrometer is described elsewhere [3]. In brief, the incident electron impinges onto the sample surface and two outgoing electrons are detected in coincidence and their momenta are measured using time-of-flight technique [4]. This technique requires a reference point on the time scale. Therefore the incident beam is pulsed with the pulse width of less than 1 ns and repetition rate of 4 MHz. The coincidence electronics allows the detection of two time-correlated electrons within the time window of 200 ns. To ensure that the two electrons are resulting from an interaction of a single incident electron with the sample a very low incident current ($\approx 10^{-14}$ A) was used providing less than one electron per pulse in average.

For the electron detection we used two position-sensitive detectors based on 75 mm in diameter Micro Channel Plates (MCP). Position sensitivity allows the measurement of the electron angular distribution and the correction of the electron flight distance for the time-of-flight energy measurement [4].

The spin-polarized electron source is based on the photo-emission from a strained GaAs photocathode with negative affinity. Photoelectrons excited by circularly polarized light with the wavelength 836 nm are initially longitudinally polarized. They pass through a 90° deflector such that the emergent beam is now transversely polarized. The polarization P of the electron beam is measured in a separate experiment and is estimated to be $(66 \pm 2)$ %. It can be reversed from P to $-P$ by changing the sense of circular polarization of the laser light incident on the GaAs photo-cathode. The polarization of the incident beam is chosen to be perpendicular to the scattering plane.
When the coincidence conditions between the two arms of the (e,2e) spectrometer are switched off each of its arms can be used as a Spin-Polarized Electron Energy Loss Spectrometer for recording electron energy loss spectra at two mirror symmetric angles.

Comparison of SOI in W(110) and Au(111)

![Figure 2. Binding energy distributions for W(110) and Au(111) – a). Corresponding spin-resolved energy sharing distributions – b).](image)

As samples we studied a W(110) surface and thin films of Au grown on this surface.

The W(110) crystal was mounted on a rotatable manipulator with the [110] direction along the rotational axis and perpendicular to the scattering plane that contains the normal to the sample surface and the detectors (see figure 1).

The tungsten crystal was cleaned in vacuum prior to measurements and to a gold film deposition using a standard procedure [5] including oxygen treatment at 10^-7 Torr oxygen pressure and 1400 K sample temperature followed by a few high-temperature flashes up to 2300 K. The cleanliness of the surface was monitored by Auger Electron Spectroscopy and Low Energy Electron Diffraction (LEED).

The Au film was evaporated from a small piece of gold wire melted on a V-shaped tungsten filament heated resistively. The layer-by-layer growth of such a film was confirmed by MEED oscillations [6]. The quality of the film was demonstrated by observing quantum well states in such a layer of gold using photoelectron spectroscopy [7]. The thickness of the gold layer (2 to 4 ML in our experiment) was estimated using Auger peak intensity as a function of deposition time as well as
3. Experimental results and discussion

Two high-Z metals were chosen to study spin-orbit interaction in the (e,2e) reaction: i) single crystal of W(110) and ii) thin film of Au deposited on the W(110) substrate. To explain the way the experimental results are presented we recall that the energy conservation law in the (e,2e) reaction implies that \( E_b = (E_1 + E_2) - E_o \), where \( E_b \) is the binding energy of the valence electron, \( E_o \) is the primary electron energy and \( E_1 \) and \( E_2 \) are the energies of the two detected electrons. Besides, the momentum conservation for the component parallel to the surface implies that \( k_{b||} = k_{1||} + ng_{||} + k_{2||} - k_{0||} \) (indices \( b,1,2,0 \) have the same meaning as for energies, \( g_{||} \) - surface reciprocal lattice vector and \( n = 0 \) for energies and geometry used in our experiments). The number of correlated pairs plotted as a function of the \( x \)-component of the bound electron momentum is called \( K_x \) - distribution. Thus, energy and momentum conservation allow the valence electron involved in the collision to be localized in energy-momentum space in contrast to a single-electron spectroscopy (EELS), where the result of a scattering event is integrated over the unresolved second electron state.

Our experimental (e,2e) spectra \( I^+ \) and \( I^- \) measured for two polarizations of the incident beam can be presented in various coordinate systems. Then the asymmetry defined as \( A = (I^+ - I^-)/(I^+ + I^-) \) can be calculated. We present in figure 2 the binding energy spectra (a) for tungsten and the gold layer

**Figure 3.** Binding energy spectra for W(111) measured at different primary energies (a) and corresponding asymmetry spectra of \( K_x \) - distributions (b).
excited by 27 eV spin-polarized primary electrons. For a particular binding energy (band of binding energy) one can plot an energy sharing distribution. These distributions can be constructed for two opposite polarizations of the incident beam and then the asymmetry can be calculated. In such a way the asymmetry of these distributions for various binding energies were calculated. It was found that the most pronounced asymmetry for W(110) is located at about 1 eV below the Fermi level and for Au film it is located at about 4 eV below the Fermi level. The spin-resolved energy sharing distributions and the spin-difference spectra for the tungsten and gold layer are shown in figure 2b. One can see that the sharing distributions for these two samples are different. On the other hand the shapes of the spin-difference curves are quit similar: in both cases the curves change sign at \( E_1 - E_2 = 0 \). This shape is typical for spin-orbit interaction and consistent with the symmetry consideration. Indeed, the reflection in the R1 symmetry plane (figure 1) reverses the spin of the incident electron (because the spin is an axial vector) and interchanges of the two outgoing electrons. It implies that \( \Gamma'(E_1 - E_2) = \Gamma(E_2 - E_1) \). In other words the spin-asymmetry will remain the same if we mirror reflect the curve in R1 plane and then flip it vertically. This “left-right” symmetry of the curves is typical for spin-orbit-induced asymmetry.

![Figure 4](image)

**Figure 4.** Binding energy spectra of Au(111) film recorded at different primary energies (a) and corresponding asymmetry spectra (b) at binding energy shown by black rectangular in (a).

We note here that in the case of tungsten a substantial density of states below the Fermi level (where the maximum of asymmetry is observed) is formed by d-states, whereas in the Au film the d-states start to contribute to the density of states only at 4 eV below the Fermi level (where the maximum asymmetry is observed for the gold layer). Thus, the energy location of the observed
asymmetry indicates that the spin-orbit effect shows up when the d-states of the valence band are involved in the (e,2e) scattering.

Spin-orbit effects in the (e,2e) reaction on W(100) were analyzed in [9]. In this work the collision of an incident electron with a valence electron of the target was treated in a distorted-wave Born approximation formalism with exchange, and the four relevant quasi-one-electron states being solutions of the Dirac equation. To obtain detailed insight into the origin of the individual features of the (e,2e) spectra of W(100) and their spin asymmetry, additional calculations were performed where each of the four relevant wave-functions was modified artificially such that spin-orbit coupling and the elastic scattering amplitudes from the ion-core planes were selectively switched off. The calculated spin asymmetry of the (e,2e) cross-section on W(100) was found to be due to the spin-orbit coupling in the valence electron states mainly. Even though these results are obtained for a W(100) surface the approach and qualitative explanation of the spin-asymmetry in the (e,2e) spectra must be valid for the W(110) surface and thin Au(111) film. Our intention was to check to what extent the spin-asymmetry in the energy sharing distributions does not depend on the primary electron energy and is only determined by the spin-orbit interaction in the valence state of the target.

![Figure 5](image-url)

**Figure 5.** Asymmetries of single-electron spectra (left column) and two-electron spectra (right column) of a W(110) crystal for various azimuth orientations. Azimuth = 0 corresponds to the crystal orientation of figure 1.

We measured (e,2e) spectra on W(110) and Au(111) at various primary energies and analyzed spin-asymmetry in the energy sharing distributions (or \( K_x \) – distributions). In the case of tungsten the shape of asymmetry in the energy sharing distributions (at binding energy just below the Fermi level) remains the same in a wide energy range from 32 eV down to 15 eV, although the size of asymmetry changes. However when the primary energy decreased to 13 eV the asymmetry changed the sign (see
In the case of the Au film the change of the sign in the spin-asymmetry occurs even at higher energy, about 22 eV (see figure 4).

These results indicate that the spin-orbit-induced asymmetry in the (e,2e) spectra of W(110) and Au(111) is not determined by the spin-orbit interaction in the valence state of the target only, but also by a contribution of the orbital momentum of the incident electron.

For the two-fold symmetry surface of W(110) we measured single-electron spectra as well as coincidence spectra for various azimuthal orientation of the surface for primary energy of 27 eV and normal incidence. It turns out that the spin-asymmetry in both single- and two-electron spectra depend on the profile of the potential in the scattering plane (figure 5). The comparison of spectra a) and c) shows that at some electron energies (or combination of the energies in the electron pair) the asymmetry even changes the sign when the azimuth of the sample orientation changes by 90°. The comparative analysis of the asymmetry in the single- and two-electron spectra shows that if the asymmetry is observed in a single-electron spectrum at particular secondary electron energy \( E_1 \), then in the corresponding coincidence spectrum the asymmetry shows up in the energy sharing distribution at the combination of energies where one of the electrons has the energy \( E_1 \). Thus the spin features of the secondary emission can be linked to the spin features of the energy loss part of the secondary emission spectrum.

4. Conclusions
Experimental results presented in the paper show that the spin-orbit-induced asymmetry in the energy sharing distribution:
- depends on the binding energy of the valence electron involved in the collision;
- sensitive to the anisotropy of the scattering potential;
- depends on the primary electron energy;

The spin-orbit-induced asymmetry observed in a single-electron spectrum (secondary emission spectrum) is observed also in the sharing distribution of the (e,2e) spectrum but with larger value. These findings allow drawing the following qualitative picture of the spin-dependent effects in the (e,2e) scattering from non-magnetic heavy-atom metals. An incident electron shares orbital momentum with the valence electron of the target forming a “quasi-particle” while scattering back to the surface. The “spin-filter” effect [10,11] on the surface potential barrier of the sample contributes to the observed intensity asymmetry in the (e,2e) spectra.

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References
[1] Kirschner J, Artamonov OM, Terekhov AN, Phys. Rev. Lett. 69 (1992) 1711
Kirschner J, Artamonov OM, Samarim SN, Phys. Rev. Lett. 75 (1995) 2424
[2] Iacobucci S, Marassi L, Camilloni R, Nannarone S, Stefani G, Phys. Rev. B51 (1995-II) 10252
[3] Artamonov OM, Samarim SN, Kirschner J, Appl. Phys. A65 (1997) 535
[4] Samarim SN, Artamonov OM, Waterhouse DK, Kirschner J, Morozov A, Williams JF, Rev. Sci. Instr. 74 (2003) 1274
[5] Cortenraad R, Ermolov SN, Semenov VN, Denier van der Gon A. W, Glebovsky VG, Bozhko SI, and Brongersma HH, J. Crystal Growth 222 (2001) 154
Zakeri Lori K, Peixoto TRF, Zhang Yu, Prokop J, Kirschner J, Surface Science 604 (2010) L1
[6] Knoppe H and Bauer E, Phys. Rev. B 48 (1993) 5621
[7] Shikin AM, Rader O, Prudnikova GV, Adamchuk VK, and Gudat W, Phys. Rev. B 65 (2002) 075403
[8] Augustus PD and Jones JP, Surf. Sci. 64 (1977) 713
[9] Gollisch H, Xiao Yi, Scheunemann T and Feder R, *J. Phys. C: Condens Matter* 11 (1999) 9555
[10] Oepen HP, Hünlich K, and Kirschner J, *Phys. Rev. Lett.* 56 (1986) 496
[11] Samarin S, Artamonov OM, Sergeant AD, and Williams JF, *Surf. Sci.*, 579/2-3 (2005) 166