Spin-polarized single- and two-electron spectroscopies at various kinematics

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Abstract. We present experimental results on the application of spin-polarized single- and two-electron spectroscopy in various kinematics for studying W(110) crystal and ferromagnetic films of Co and Fe. Spin effects in the measured spectra are expressed in terms of the asymmetry $A$ defined as $A = (I^+ - I^-)/(I^+ + I^-)$, where $I^+$ and $I^-$ are spectra measured for two opposite orientations of the polarization vector of the incident beam. It is demonstrated that the asymmetry spectra depend on the kinematics of experiment and can even change the sign of the asymmetry.

1. Introduction

Spin-polarized Electron Energy Loss Spectroscopy (SPEELS) was applied for studying Stoner excitations [1-8] as well as spin waves on various ferromagnetic surfaces [9, 10]. Spin-polarized two-electron spectroscopy (SPe,2e) provided information on spin-related properties of nonmagnetic [11] and magnetic [12] surfaces. Usually the geometrical arrangement of a SPEELS measurement is characterized by a fixed angle between the incident electron beam and the axis of the detector (analyzer) [7] and the sample can be rotated around the axis perpendicular to the scattering plane containing the electron beam and the detector. In that way the angles of incidence and detection can be varied conveniently.

There are several important specific features in electron scattering from surfaces:

- The presence of the surface implies that the parallel-to-the-surface electron momentum is conserved;
- Refraction of the electron trajectories at the surface potential barrier while passing through the interface;
- Ordered distribution of scattering centres at the surface implies that electrons (or electron pairs) undergo a diffraction;
- Imbalance of spin-up and spin-down electrons in the valence band in a ferromagnetic surface leads to the intensity asymmetry of measured spectra.

Generally two regimes of the SPEELS measurements are used: i) specular geometry, when the incidence angle is equal to the detection angle; and ii) off-specular geometry when the incident angle
is not equal to the detection angle. It is assumed that in the specular geometry the mechanism of the electron energy loss is of dipole type, i.e. the energy loss does not involve a large momentum transfer and the interaction of the incident electron with the surface occurs through the electromagnetic field generated by the electron. In the second case, off-specular geometry, the electron-electron encounter occurs with substantial momentum transfer from the incident electron to the valence electron of the target. It is also assumed that in the dipole-type of scattering no exchange effects can be observed.

The aim of this work is to test various kinematical arrangements for the spin-polarized electron spectroscopies and to study the influence of the kinematics on spin effect observed in SPEELS and (SPe,2e).

2. Experimental details

In our experimental setup we used a spin-polarized electron source based on the photoemission from strained GaAs crystal [13] activated by successive Cs deposition and oxygen adsorption to obtain a negative electron affinity surface. Electrons emitted from this surface under properly tuned laser light are longitudinally polarized with polarization vector either parallel or anti-parallel to the electron momentum depending on the helicity of the laser light. Bending electron trajectories by 90° using a quasi-spherical deflector transforms the polarization of the beam from longitudinal to transverse. The sense of electron beam polarization (“up” or “down”) can be changed by changing the helicity of the laser light used for extracting electrons from the photocathode.

We used a time-of-flight (TOF) technique for the electron energy measurements [14] for which the electron beam was pulsed with a pulse width of about 1 ns and the repetition rate of 2.5 MHz. The pulsing provides reference point on the time scale. The energy resolution of the TOF method varies with the electron energy and is usually better for low-energy electrons. We estimate the energy resolution of the TOF spectrometer is on average better than 0.5 eV in the energy range of the measurements.

A particular feature of our measurements is the possibility of using various kinematics in a wide range of angles of incidence and detection. Figure 1 represents a sketch of the geometrical arrangement with two detectors situated at 50° and 140° with respect to the axis of the spin-polarized electron gun. The sample was mounted on a rotatable manipulator with axis perpendicular to the scattering plane enabling a polar angle change. In addition, the sample can be rotated around the axis perpendicular to its surface for azimuthal angle changes. Three possible polar orientations of the sample are shown in figure 1. Position a) and position b) correspond to a specular and normal incidence geometry, respectively, for detection of scattered electrons by Detector 1. Position c) allows the detection of scattered electrons by detector 1 and detector 2 simultaneously. They can be used independently (two SPEELS spectra) or in coincidence (spin-polarized (e,2e) spectroscopy [15, 16, 17]).

As samples we used a single crystal of W(110) and thin ferromagnetic films (Fe, and Co) deposited on the W(110) substrate. Prior to the measurements on W(110) or a deposition of the film, the W(110) crystal surface was cleaned using well established procedure [18, 19] including oxygen adsorption at the sample temperature about 1600 K with subsequent high temperature flashes up to 2500 K.

Figure 1. Three possible geometrical configurations of experiment.
3. Results and discussion

3.1. Single crystal of tungsten W(110)

The low index faces of a tungsten single crystal, (100), (110) and (111) are very well studied by various electron spectroscopies including Electron Energy Loss Spectroscopy [20, 21], photoelectron spectroscopy [22], secondary emission spectroscopy [23], and two-electron spectroscopy [24]. The W(110) surface is non-reconstructing and very often is used as a substrate for metal films deposition. The large atomic number metal tungsten crystal is a suitable sample for the observation of spin-orbit effects as observed in W(110) using spin-polarized secondary emission spectroscopy [23] and spin-polarized low-energy two-electron spectroscopy [24]. Our aim here is to show how the kinematics of the measurement influences the manifestation of spin-orbit interaction.

Since the spin-orbit effect is proportional to the scalar product of the electron orbital momentum and the electron spin ($L \cdot S$), it is a maximum when the polarization vector of the incident electron is perpendicular to the scattering plane (because the orbital angular momentum of the electron is perpendicular to the scattering plane). In all cases the polarization vector was perpendicular to the scattering plane. We used various geometric arrangements to measure the asymmetry $A$ of the scattered and emitted electrons with the asymmetry defined as $A = (I^+ - I^-)/(I^+ + I^-)$, where $I^+$ and $I^-$ are the energy distributions of scattered electrons for two opposite orientations of the polarization vector of the incident beam.

The presence of the surface breaks the translational symmetry in one direction, perpendicular to the surface, and, as a consequence, only the parallel-to-the-surface component of the electron momentum is conserved (modulo a reciprocal lattice vector). The crystal structure of the sample implies that a reciprocal lattice vector $\mathbf{K}$ can be added or subtracted in the momentum balance of the electron-electron collision. Figure 2 shows the balance of energy and momenta in simple electron-electron scattering.

![Figure 2](image)

**Figure 2.** Geometrical representation of the kinematics of electron-electron scattering: $\mathbf{k}_0$ and $\mathbf{k}_1$ are the wave vectors of incident and scattered electrons; $\mathbf{k}_{0x}$ and $\mathbf{k}_{1x}$ are projections of the incident and scattered electrons on the sample surface; $z$ is the axis perpendicular to the sample surface.

In figure 2, sections a), b) and c) correspond to the geometrical arrangements a), b) and c) of figure 1, respectively. Wave vectors of the incident and scattered electrons are denoted by $\mathbf{k}_0$ and $\mathbf{k}_1$, respectively. The $\mathbf{k}_{0x}$ and $\mathbf{k}_{1x}$ are the projections of the incident and scattered electrons. Given the periodicity along the sample surface, the parallel to the surface component of the electron wave vector should satisfy the following equation: $\mathbf{k}_{0x} - \mathbf{k}_{1x} = q_x = n \mathbf{q}_{x}$, where $h \mathbf{q}$ is the momentum transfer, $n$ is integer, $\mathbf{K}$ is the two-dimensional reciprocal lattice vector. In the case of a creation of an electron-hole pair the momentum of the electron-hole pair is $h \mathbf{q}$. The kinematics presented in figure 2 do not
account for the second electron that remains inside the solid. In some cases the electron-electron scattering may be assisted by the reciprocal lattice vector transfer (\textit{umklapp} process).

An example of a SPEELS measurement on W(110) crystal at three different kinematics is shown in figure 3. Low energy electrons (13 eV) impinge onto the surface at normal incidence and secondary electrons (including true secondaries, elastically and inelastically scattered primary electrons) are detected by the right (Det. 2) or the left (Det. 1) detectors as shown on the section a) of figure 3. The corresponding asymmetries $A$, red circles (Det. 2) and black squares (Det. 1) are mirror symmetric with respect to the $x$-axis. This symmetry is consistent with the general symmetry of the experimental set-up; specifically the sample crystal is symmetric with respect to the plane perpendicular to the sample surface and to the scattering plane. Mirror reflection in this plane of the whole experimental set-up will exchange detectors Det. 1 and Det. 2 and flip the polarization vector of the incident beam (because it is an axial vector) with everything else remaining unchanged. Therefore the asymmetries measured in Det. 1 and Det. 2 are the same but with opposite signs.

For specular geometry and grazing incidence the asymmetry of the secondary emission spectrum (including true secondaries, elastically and inelastically scattered primaries) generated by the 13 eV spin-polarized electron beam is very different from the normal incidence that is shown in figure 3 b).

The origin of the intensity asymmetry of secondary emission spectra excited by spin-polarized electrons in this case is the so-called “spin-filter effect” [25, 26]. The qualitative explanation of this effect is the following. Incident spin-polarized electrons while losing their energy via electron-electron scattering still keep a certain degree of polarization. On the way out of the solid to the vacuum these electrons undergo spin-dependent (due to spin-orbit interaction) scattering at the surface, such that their transmission through the surface potential barrier becomes spin-dependent. Presumably incident electrons also undergo spin-dependent transmission through the surface penetrating into the surface region of the crystal. It is seen from figure 3 that the intensity asymmetry for grazing incidence is even larger than for normal incidence. These features in the intensity asymmetry spectrum are related to the scattering events with large energy and momentum transfer. For the specular geometry the number of such event is relatively small since the elastic maximum is dominating in the spectrum at this geometry. On the other hand these features in the intensity asymmetry highlight the contribution of electron-electron collisions in contrast to the dipole mechanism of the electron-surface interaction.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure3.png}
\caption{a) Asymmetry of the secondary emission spectrum from W(110) for $E_p = 13$ eV, normal incidence; b) asymmetry for $E_p = 13$ eV, grazing incidence ($\theta = 70^\circ$).}
\end{figure}
3.2. Ferromagnetic film of Fe on W(110)
In contrast to the spin-orbit interaction the exchange effect depends on the relative orientation of the polarization vector of the incident beam and the magnetic moment of the magnetized sample. For example, figure 4 shows an asymmetry of the Stoner excitations in a 3ML Fe film on W(110) excited by normal incidence spin-polarized electrons for two opposite orientations of magnetic moment of the sample (solid circles and open circles). These two asymmetries are mirror symmetric with respect to the X- axis. In addition, the asymmetry of SPEELS was measured for the magnetic moment of the sample perpendicular to the polarization vector of the incident beam (triangles).

This asymmetry apparently is zero. Systematic measurements of the asymmetry of Stoner excitations on the Fe film as a function of the azimuthal angle of the sample (i.e. the angle between the magnetic moment of the sample and polarization vector of the incident beam) shows an almost perfect cosine dependence (figure 4b). These measurements confirm that the magnitude of the Stoner excitation asymmetry in case of a single domain ferromagnetic sample is proportional to the scalar product (M·P) of the magnetization (M) of the sample and the polarization vector (P) of the incident beam.

The origin of the asymmetry in the above spectra is the different probability of spin flip exchange process for spin-up and spin-down incident electrons. Each of these scattering processes leads to the creation of an electron-hole pair with opposite spin characters, which is called Stoner excitation. A probability of an energy loss (Stoner excitation) with a corresponding momentum transfer in SPEELS depends on the number of available combinations (pairs) of states in occupied and unoccupied bands of opposite spin character (Density of Stoner states) and a matrix element of a corresponding transitions. The density of Stoner states is given by [27]:

\[ \rho_{\sigma}(E) = \frac{1}{N_0} \sum_{\mathbf{k}} (f_{n\mathbf{k}+\sigma} - f_{n\mathbf{k}-\sigma}) \cdot \delta(E + \varepsilon_{n\mathbf{k}+\sigma}, (\mathbf{k} - \mathbf{q}) - \varepsilon_{n\mathbf{k}-\sigma} (\mathbf{k})) . \]

where \( n \) and \( n' \) are the indices of majority and minority bands, \( \mathbf{k} \) is the Bloch wave vector, \( f_{n\mathbf{k}\sigma} \) is the occupation probability of the state specified by \( (n, \mathbf{k}, \sigma) \), \( \mathbf{q} \) is the momentum transfer. The \( \delta \)-function ensures the energy and momentum balance in the density of Stoner states.
The matrix element of the interband transition (between a majority and a minority bands) in given by [27]:

\[ W_{\alpha \nu'}(p, q, k) = \frac{N_0}{V} \sum_k \hat{v}(k - p - K) \cdot \hat{\phi}_{\alpha k}^*(k - K) \cdot \hat{\phi}_{\nu' k-q}^*(k - q - K), \]  

where \( p, q \) and \( k \) are the incident electron momentum, momentum transfer and Bloch wave vector of the valence state, respectively; \( K \) is the reciprocal lattice vector; \( V \) is the volume of the crystal; \( N_0 \) is the number of sites; functions \( v \) and \( \phi \) with caret signs are the Fourier transforms of the interaction potential and the Wannier functions, respectively. As was pointed out in [28], for a fixed scattering angle, i.e., for given incoming and outgoing momenta, there are three possible scattering processes corresponding to different momentum transfer: i) in the first one a specular scattering follows a small momentum transfer inelastic scattering giving momentum transfer \( q \); ii) in the second, an elastic (or specular) scattering precedes a relatively small-angle inelastic scattering event, giving momentum transfers \( q' \); iii) in the third process, there is a single large-angle inelastic scattering event with a momentum transfer \( q'' \) absorbed by the electron-hole pair excitations. These types of scattering may occur in all three geometrical arrangements shown in figure 2. The processes i) and ii) are explicitly shown only in figure 2a) and c), respectively. Which process with which momentum transfer \( q, q', q'' \) contributes mostly to the observed SPEELS of Stoner excitations depends on the kinematics of the scattering. The partial cross sections of Stoner excitations corresponding to these three possible scenarios of momentum transfer were calculated in [28]. It was found for a specular geometry that the cross section corresponding to \( q'' \) momentum transfer (direct electron-electron encounter) makes a dominant contribution to the total cross section. At off-specular geometry the cross sections corresponding to \( q' \) and \( q'' \) momentum transfer are almost equal and much larger than the cross section corresponding to \( q \) momentum transfer. The overall shape of Stoner excitations in SPEELS is determined by the combined effect of the density of Stoner states (1) and the matrix element (2).

Figure 5 represents SPEELS asymmetry spectra of 5 ML iron film on W(110) excited by 24 eV spin-polarized electrons at three different kinematics: a) specular geometry with the incidence angle of 25°; b) normal incidence and 50° detection angle; c) specular geometry with 70° incident angle. First two spectra show a positive asymmetry with a maximum at about 2.5 eV of energy loss. This is consistent with the Stoner excitation maximum position. The sign of asymmetry corresponds to the magnetization M2 in our nomenclature. The shapes of these two asymmetry spectra are quite different: for specular geometry the asymmetry of elastic scattering has almost the same value as the Stoner excitation asymmetry and there are two kinks at about 5 eV and 9 eV of energy loss. For normal incidence the asymmetry of elastic scattering is almost zero and the Stoner excitation asymmetry is well localised at 2.5 eV. A very different asymmetry is observed for the 70° of incidence: first of all the sign of the asymmetry is changing from negative, around the Stoner excitation energy loss, to a positive sign at larger energy losses. We note here that the
magnetization of the film is the same (M2). It appears that there is an overlap (sum) of two contributions to the asymmetry spectrum at the grazing incidence: the Stoner excitation asymmetry with the positive sign, similar to geometries a) and b), and a negative maximum of asymmetry located at about 4 eV energy losses, which is due to a different from Stoner excitation mechanism. Our preliminary investigation shows that this maximum in the asymmetry spectrum is related rather to an emission feature in the secondary electron emission spectrum. The origin of this maximum needs further investigation.

3.3. Spin-polarized two-electron spectra of Co film on W(110)

In the two-electron spectroscopy in reflection geometry, two low-energy electrons resulting from the interaction of a single incident electron with a surface are detected in coincidence and their energy and momenta are measured. The energy distribution of correlated electron pairs is called the (e,2e) spectrum. Using a spin-polarized incident beam the (e,2e) spectra are measured for spin-up and spin-down incident electrons. Figure 6 illustrates the influence of the geometry of the experiment on the shape of (e,2e) energy distributions and on spin effect in the (e,2e) reaction. The left panel of figure 6 shows binding energy spectra and energy sharing distributions recorded at normal incidence of 24 eV spin-polarized electron beam for 3 ML cobalt film on W(110). The binding energy according to the energy conservation in the (e,2e) reaction is determined by the relation: \( E_b = E_0 - E_1 - E_2 \), where \( E_0, E_1, E_2 \) are the energies of incident and two scattered electrons, respectively. The angle between detectors is 100°. Spectra measured for spin up and spin down polarization of the incident beam are shown together with the difference spectrum (triangles in the figure). The right panel shows binding energy spectra and energy sharing distributions measured on 3 ML cobalt film on W(110) at 57° angle of
incidence. The angle between detectors is 90° and the angle between incident beam and detector D1 is 50°. The shapes of binding energy spectra for both geometrical arrangements are similar (see figure 6 a) and b)) whereas the energy sharing distributions (figure 6 c) and d)) are very different. The difference in the energy sharing distributions for normal and 57° of incidence is explained by the fact that the tangential component of the electron momentum is conserved. Indeed, for the off-normal incidence the total momentum of the pair of electrons is carrying parallel to the surface momentum of the incident electron: $K_{b||} + K_{0||} = K_{1||} + K_{2||}$, where $K_b$, $K_0$, $K_1$, $K_2$ are momentum of the bound (valence) electron, incident electron and two outgoing electrons, respectively. The spin effects (difference spectra), for both binding energy spectra and energy sharing distributions, at two geometries are different. The asymmetry of the binding energy spectrum to a large extent depends on the spin asymmetry of the Bloch spectral density function of the sample [29]. It is evident from figure a) and b) that for the off-normal incidence the asymmetry of the binding energy spectrum (even the sign) depends also on the scattering dynamics. The difference spectrum of the energy sharing distribution for the normal incidence and symmetric detection of pairs shows a maximum in the middle of the spectrum (energies of electrons are almost equal), whereas for off-normal incidence the difference spectrum has complicated structure and asymmetric with respect to the zero point ($E_1 – E_2 = 0$).

4. Conclusions
The experimental results indicate that spin effects in inelastic (and elastic) scattering of spin-polarized electrons from magnetic and nonmagnetic surfaces depend very much on the geometrical arrangement of the experiment, i.e. on angle of incidence and angle of detection (in SPEELS), as well as on angle between detectors (in spin-polarized two-electron spectroscopy). We note that for grazing incidence of spin polarized electrons on 5 ML Fe film on W(110) a new mechanism responsible for intensity asymmetry in the secondary emission spectrum is confirmed. In (e,2e) the intensity asymmetry in binding energy spectra and in energy sharing distributions is determined not only by an imbalance of spin-up and spin-down states in the valence band but also by kinematics.

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