doi:10.1088/1742-6596/100/7/072033

Low-energy spin-polarized two-electron spectroscopy: a powerful tool for studying exchange correlation and spin-orbit interaction on surfaces

S Samarin¹, OM Artamonov², AD Sergeant¹, J Kirschner³, and JF Williams¹

¹Centre for Atomic, Molecular and Surface Physics, University of Western Australia, Perth, WA 6009, Australia,

²Research Institute of Physics, St. Petersburg University, St. Petersburg, Russia.

³Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany.

samar@physics.uwa.edu.au

Abstract. Spin-polarized two-electron spectroscopy-in-reflection has been proven to be a very efficient technique for studying the exchange correlation and spin-orbit interaction on magnetic and non-magnetic surfaces and thin ferromagnetic films. The essence of the technique is the detection of two time-correlated electrons emitted from the sample surface upon an impact of a single incident electron and measurements of their momenta. The coincidence technique has been combined with a time-of-flight electron energy analysis to measure momentum distributions of correlated electron pairs for various orientations of the electron beam polarization relative to the scattering plane and magnetization of the sample. This set of measurements allows to extract the information on exchange correlation and spin-orbit interaction from measured spectra. Energy- and momentum-conservation in the electron-electron scattering allows the valence electron involved into collision to be located in the energy-momentum space of the valence band of the sample. In this way the energy and momentum location of the exchange and SOI can be established. A few examples of the application of this technique for studying ferromagnetic and non-magnetic surfaces are presented.

1. Introduction

In conventional Electron Energy Loss Spectroscopy (EELS) one electron resulting from an interaction of the incident electron with a surface is detected and its energy is measured. It is essentially a single-electron spectroscopy. In contrast, in two-electron spectroscopy (hereafter referred to as (e,2e) spectroscopy) two time-correlated electrons resulting from an interaction of an incident electron with a surface are detected and their momenta are measured [1]. Spin-polarized version of the (e,2e) spectroscopy, where the incident electron beam is spin-polarized is inherently suitable for studying exchange correlation (EC) [2] and spin-orbit interaction (SOI) [3] on surfaces. It will be demonstrated below that the spin-polarized (e,2e) spectroscopy allows to unravel spin correlations, which do not show up in a single electron spectroscopy.

Two spin-dependent interactions: the exchange interaction and the spin-orbit interaction play a fundamental role in itinerant ferromagnetism. The exchange interaction is a result of the fermionic (spin = $\frac{1}{2}$) nature of the electrons. It is essentially a many-particle effect. To observe the exchange interaction one needs to monitor at least two interacting electrons with well-defined spin states.

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Journal of Physics: Conference Series 100 (2008) 072033

doi:10.1088/1742-6596/100/7/072033

The SOI is an interaction of the electron spin with its orbital momentum. It shows up, for example, in elastic electron scattering by the ion core potential and has a maximum value when the polarization vector of the incident beam is perpendicular to the scattering plane. It can also be seen in an inelastic scattering channel using spin-polarized two-electron spectroscopy [3].

In general, both of these effects, exchange and SOI, are present when a spin-polarized electron is scattered from a ferromagnetic surface, but the strength and relative contribution of each of them depend on the surface properties and scattering geometry.

2. Spin-polarized two-electron spectroscopy of surfaces

The essence of the two-electron spectroscopy is the detection of two time-correlated electrons resulting from an interaction of a single incident electron with a surface. It was demonstrated that in

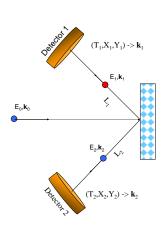


Figure 1. Experimental set-up

particular experimental conditions these two electrons result from a single electron-electron collision where energy and momentum are conserved [4]. 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.

The momentum conservation for the component parallel to the surface implies that $k_{b\parallel}=k_{1\parallel}+ng_{\parallel}+k_{2\parallel}$ - $k_{0\parallel}$ (indices $_{b}$,1,2,0 have the same meaning as for energies, g_{\parallel} - surface reciprocal lattice vector and n=0 for energies and geometry used in our experiments). Thus, energy and momentum conservation laws 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 unresolved second electron state.

The experimental set-up of the (e,2e) spectrometer is described in details elsewhere [5] and geometry is depicted in Fig.1.

In the spin-polarized (e,2e) experiment the polarization of the incident beam is chosen to be perpendicular to the scattering plane and spectra are measured for two orientations of the spin.

3. Overview of spin effects that can be observed on surfaces using spin-polarized (e.2e)

We present two examples of application of spin-polarized two-electron spectroscopy for studying spin-orbit interaction (SOI) and exchange correlation on surfaces. One concerns a non-magnetic material, tungsten crystal W(110), and the second – thin ferromagnetic layer of cobalt on W(110). The advantageous features of the two-electron spectroscopy are outlined by comparison with a single-electron spectroscopy.

3.1. Spin-orbit interaction on W(110) surface

Tungsten is the ideal model target for studying SOI. It is a large Z-metal that implies a strong SOI. The tungsten surface can be prepared and cleaned in a reproducible way that allows a comparison of results obtained by different methods. It has been studied by most of the surface science techniques providing rather complete information on the electronic and geometric structure of the surface. The spin-polarized low energy electron diffraction (SPLEED) from W(001) has been studied extensively both experimentally and theoretically [6,7]. Much less is known about inelastic scattering of spin-polarized electrons from tungsten. It was reported recently [8] that a secondary emission spectrum excited by spin-polarized electrons from W(110) exhibits intensity asymmetry as a function of electron energy. Spin-polarized two-electron spectroscopy unraveled the anisotropy of SOI in W(110) [9].

The overview of the spin effects in the spin-polarized (e,2e) spectrum is represented in Fig. 2. The left panel shows 2D difference spectrum, which is obtained by subtracting the (e,2e) spectrum

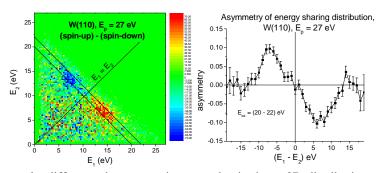


Figure 2. Left panel - difference between spin -up and spin-down 2D distributions of correlated electron pairs. Right panel - spin asymmetry of energy sharing distributin for total energy of pairs insid the band between two black lines on left panel.

measured with spin-down incident electrons from the spectrum measured with spin-up incident electrons. The main contrast is observed in the total energy band of 2 eV width (marked by two black lines) just below the Fermi edge. The energy-sharing distribution corresponding to this total energy exhibits a strong asymmetry, which is shown in the right panel. The shape of the curve is compatible with the symmetry property of the SOI. Indeed, mirror reflection in the symmetry plane perpendicular to the scattering plane reverses the sign of polarization and interchanges the detectors (E_1 and E_2).

3.2. Exchange and spin-orbit interaction in a ferromagnetic layer An overview of the exchange correlation in 3 Ml Co film deposited on W(110) substrate is presented in Fig. 3.

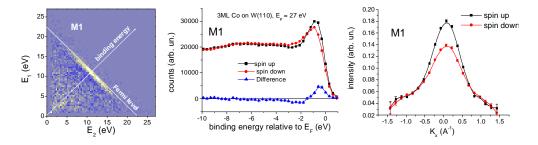


Figure 3. Overview of exchange correlation in 3 ML Co film on W(110). Left panel - difference between spin-up and spin-down 2D distributions of correlated electron pairs. Middle panel - binding energy spectra for spin-up and spin-down primary electrons. Right panel $-k_x$ – distributions.

The left panel shows the 2D difference spectrum, which is obtained by subtracting the (e,2e) spectrum measured with 22 eV primary energy spin-down incident electrons from the spectrum measured with spin-up incident electrons. In contrast to the difference spectrum of tungsten (Fig. 2) this spectrum exhibits the contrast, which is symmetric relative to the diagonal white line (marked as binding energy axis). The binding energy spectra shown in the middle panel are different for spin-up and spin-down primary electrons. The main difference occurs within 1 eV binding energy just below the Fermi level. This difference mainly reflects the difference in the density of states of spin-up and spin-down electrons in the valence band of cobalt. The extensive theoretical work by J. Berakdar [10] pointed out that the selection of correlated electron pairs, which are detected symmetrically (equal energies and symmetric angles) would allow to extract the spin asymmetry of the spectral density function in the centre of the surface Brillouin zone [5]. Moreover, the analysis of k_x – distributions (k_x – parallel to the surface and to the scattering plane component of the bound electron wave vector) recorded for two

Journal of Physics: Conference Series 100 (2008) 072033

doi:10.1088/1742-6596/100/7/072033

opposite magnetizations of the sample (Fig. 3, right panel), provides the information on energy- and momentum - resolved exchange and spin-orbit contribution to the measured asymmetry [11].

3. Comparison of spin-polarized single- and two-electron spectroscopies

We would like to emphasise here that the spin-polarized (e,2e) spectroscopy provides observations of

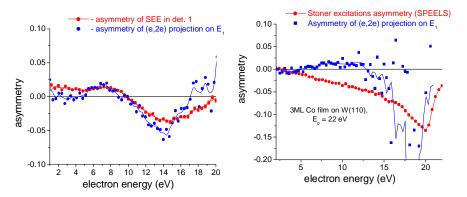


Figure 4. Comparison of spin effects observed in single-electron spectroscopy (SEE) and in two-electron spectroscopy. Left panel - SOI observed in (e,2e) spectrum of W(110) is also visible in a single-electron spectrum (SEE). Right panel - spin correlations observed in (e,2e) spectrum of Co film are not visible in a single electron spectrum (SPEELS).

spin-dependent features in electron scattering from surfaces that are not accessible with a single-electron spectroscopy, SPEELS, for example. First of all it identifies the location in energy-momentum space of the valence electron involved in the collision. In a nonmagnetic material (tungsten) the spin asymmetry projected on one of the energy axis is also visible in the secondary emission spectrum (Fig. 4, left panel) although the effect in the (e,2e) spectrum is much larger (see Fig. 2, right panel). In contrast, exchange correlation, which is essentially a many-electron effect, observed in the (e,2e) spectrum (Fig. 3), is not seen in the single-electron spectrum (SPEELS) (Fig. 4, right panel). Indeed, the large positive contrast in 2D difference spectrum of Co (Fig. 3, left panel) at $E_1 = E_2$ would correspond to about 10 eV energy of both electrons of a pair. This positive asymmetry is still visible in asymmetry of the 2D projection on E_1 axis (blue line in Fig. 4, right panel), while the asymmetry of the SPEELS is dominated by the Stoner excitation asymmetry.

Acknowledgement. This work was supported by The Australian Research Council and The University of Western Australia.

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