Benchmark the performance of the spin-resolved time-of-flight electron analyzer in Artemis

Contact *yu.zhang@stfc.ac.uk*

Y. Zhang, P. Pearcy, G. Karras, R.T. Chapman, A.S. Wyatt, C. Sanders, E. Springate
Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot OX11 0QX, UK

Introduction

The spin-resolved time-of-flight (Spin-TOF) system in Artemis[1] is a time-of-flight electron energy analyzer incorporated with a Mott spin polarimeter. By working with a pulsed photon source, the Spin-TOF can be used for the photoelectron spectroscopy (PES) measurements with the spin analysis of electrons. The spin-resolved band structure measurement offers a great advantage in the study of fundamental properties, such as magnetism and spin-orbital couplings.

However, limited by the low efficiency of the Mott spin polarimeter, the Spin-TOF setup usually requires a very high data collection rate for the spin-resolved measurements. This is rather difficult to achieve by using the current 1 kHz extreme ultraviolet (XUV) source in Artemis, especially in the study of electron dynamics, where the signal is usually an order of magnitude smaller than that of electron bands. The planned upgrade of Artemis with the 100 kHz repetition rate light source can thus benefit the Spin-TOF and improve its performance significantly. To be prepared for the upcoming upgrade and as a comparison to the new light source, we carried out this work to benchmark the performance of the Spin-TOF system on Au(111) with the current 1 kHz XUV source.

The Spin-TOF setup

The Spin-TOF setup is an ultrahigh vacuum (UHV) system including the Spin-TOF analyzer, an analysis chamber and a preparation chamber (see Fig.1). The Spin-TOF analyzer contains a movable multichannel plate (MCP). When it is lowered down and positioned in front of the Mott polarimeter, the analyzer works as a normal TOF analyzer providing spin-integrated signal measurements. When the MCP is lifted up and replaced by a tube, electrons are then ported into the Mott polarimeter for spin-resolved measurements.

The Spin-TOF analyzer is mounted on the analysis chamber, in which a sample is placed on a 4-axis manipulator, enabling liquid He cooling and e-beam heating. An Omicron SPECTALEED has been mounted on the analysis chamber to check the surface quality of samples. The preparation chamber is also connected to the analysis chamber: here samples can be temporarily stored, or be pretreated by using Ar sputtering and e-beam heating.

Spin-polarized surface states on Au(111)

We tested the Spin-TOF by using an Au(111) sample. A Shockley surface state exists in the band gap of the Au(111) surface. Due to the strong spin-orbit interaction of gold atoms, this surface state shows a well-known split in dispersion depending on the spin of electrons (Fig. 2 A, B). This split is also referred to Rashba spin splitting. For electrons with the same wave vector towards the right, the spin up (red) electrons show slightly higher energy (about 200 meV) than the spin down (blue) electrons (Fig. 2 A). For the electrons travelling towards the left, the situation is just reversed [2]. In a 2D top view (Fig. 2 B), the spin of the surface states forms two circles: the outer one from electrons with spins pointing clockwise, and the inner one just reversed.

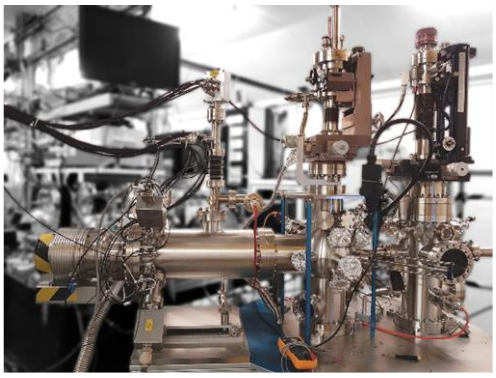

Figure 1 *Photo of the Spin-TOF setup. The time-of-flight analyzer with an integrated Mott spin polarimeter is in the horizontal cylindrical chamber on the right. The main analysis chamber, located in the middle, is equipped with a vertical sample manipulator (brown). Samples can be cooled down to about 30K in a measurement. The preparation chamber is on the right, where samples are loaded and prepared, such as Ar sputtering and annealing. Five samples can be temporarily stored on its manipulator (black).*

In a PES measurement, the sample is rotated with respect to the analyzer direction for the collection of electrons with different in-plane wave vectors. This is illustrated in Fig. 2 C. Photoelectrons generated from the sample fly through the drift tube and are accelerated up to 30 keV towards the gold thin film in the Mott polarimeter. Due to the strong spin orbit interaction, electrons are scattered into different directions depending on their spins. The spin-resolved spectra are obtained from the MCP detectors at different collection positions.

Experiments

The Au(111) sample surface was prepared by a few cycles of sputtering (2 keV, 5 μA/cm^2, 1.5 hours) and annealing (400 V, 16 mA, 1 hour) in the preparation chamber. After the final annealing, the sample was checked by the LEED in the analysis chamber. The LEED pattern shows sharp spots together with the satellite spots due to reconstruction on the Au(111) surface, which indicates a clean sample surface.

The XUV beam was prepared from high harmonic generation of a 400 nm fundamental beam of about 260 mW. The XUV flux was about 2×10^6 photons/pulse for the 7th harmonic (21.7 eV) with pulse duration of about 30 fs and a repletion rate of 1 kHz. A strong space charge effect was, however, observed due to the high density of photoelectrons generated by the XUV pulses. To reduce the space charge effect and also improve angular resolution, the 5th harmonic (15.5 eV) was used instead. The flux was about 7×10^5 photons/pulse. The beam size on the sample is about 1×0.5 mm.

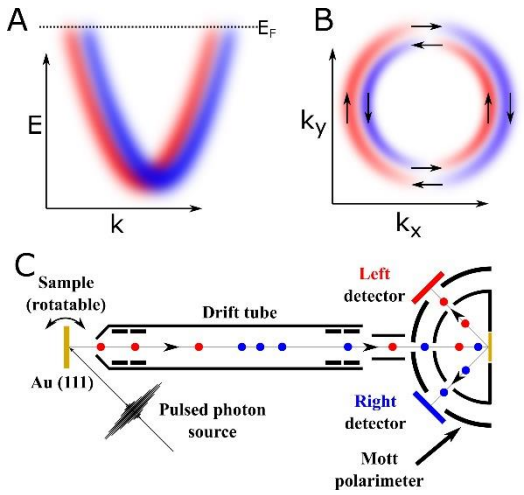

Figure 2 *Schematics of the Rashba spin splitting of the surface state on Au(111) and the experimental geometry.* ***A.*** *The spin-split dispersion of the surface state in the energy-momentum space. The colours indicate the spin orientations.* ***B.*** *The spin-split dispersion of the surface state in k-space. The direction of electron spins are indicated by arrows.* ***C.*** *Schematic of the Spin-TOF working geometry (top view).*

The dispersion of the surface states was first measured with the spin-integrated detector by rotating the sample by ±8 degrees with respect to the direction of the analyzer. The parabolic dispersion observed is shown in Fig. 3 A. To improve the energy resolution, the retarded potential was set to 9.5 V to make the kinetic energy less than 1 eV for electrons in the drift tube. The energy resolution was estimated from the Fermi edge, which is about 130 meV due to the bandwidth of the XUV pulses and instrumental broadening of the analyzer.

After optimization of the spectral intensity, the spin-integrated MCP detector was lifted up. Spin-resolved measurements were then performed with the Mott polarimeter. Compared to the spin-integrated signal, the spin-resolved signal was about three orders of magnitude smaller. For example, the spin-integrated signal was measured at about 600 electrons/second with the sample angle at 4 degrees; when using the Mott polarimeter, the count rate was around 0.6 electrons/second.

Results

Despite the low efficiency of the Mott polarimeter, a spin-resolved measurement was successfully completed after about 48-hours of data collection. Fig. 3 B shows the spin-up and spin-down spectra measured with the sample angle at 4 degrees, which is marked by the dashed line in Fig. 3 A. A small spin split can be distinguished between the two spectra.

The small spin asymmetry mainly came from the large collection angle of the spectrometer, which is about 5.7 degrees derived from the 20 mm sample distance and 2 mm entrance size. When an XUV energy of about 15.5 eV is employed, the dispersion of the surface state distribute between ±7 degrees with respect to the analyzer direction. A collection angle of more than 5 degrees thus largely increases the collection range in terms of wave vector and broadens the peak width in energy. This in turn reduces the difference between the two spin bands shown in Fig. 2 A.

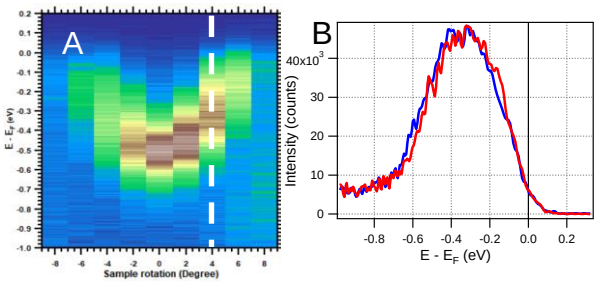

Figure 3 ***A.*** *The spin-integrated band map of the surface state on an Au(111) crystal. The dispersion of the surface state is observed.* ***B.*** *The spin resolved spectra obtained from the Mott polarimeter. The sample angle was at 4 degrees, as marked by the dashed line in the graph A. It corresponds to a wave vector of 0.11 A^{-1}. Red and blue curves are the spectra from spin-up and -down electrons as defined in Fig. 2. Spin-split can be observed from the mismatch between the two spectra.*

Conclusions

The performance of the Spin-TOF analyzer was evaluated by measuring the spin-polarized surface state on an Au(111) crystal with the current 1 kHz XUV source. The parabolic dispersion of the Au(111) surface state was observed, with an energy resolution of about 130 meV. By using a Mott polarimeter, the spin-polarized states were successfully resolved at about 0.11 A^{-1}. However, the measurement took about 48 hours to reach a satisfactory signal-to-noise ratio. Moreover, the test experiments were static measurements of occupied electron bands. For the study of electron dynamics, at least an order of magnitude higher data rate is needed due to the low signal in a pump-probe measurement. The Spin-TOF analyzer with 1 kHz XUV source might be suitable for a pump-probe measurement in a time scale of several days, but this is not a realistic experimental scheme in beam times.

A high repetition rate photon source is therefore of key importance for the Spin-TOF system. It is highly expected that the new 100 kHz light source in Artemis would be able to drive an efficient XUV source and thereafter improve the performance of the Spin-TOF analyzer.

To increase the angular resolution of the Spin-TOF system, a small entrance pinhole may be used to reduce the collection angle. Meanwhile, small XUV energy can be employed to reduce the kinetic energy of the photoelectrons, which will result in a large angular distribution. This will also help to improve the angular resolution of the Spin-TOF system. The development of HHG at low energies, between 6 and 11 eV, is thus of great interest for the application of the Spin-TOF system in the future.

Acknowledgements

Technical support from Phil Rice, Alistair Cox and Dave Rose is kindly acknowledged.

References

1. C.M. Cacho, S. Vlaic, M. Malvestuto, B. Ressel, E.A. Seddon, and F. Parmigiani, Rev. Sci. Instrum. **80**, 043904 (2009)

2. C. Tusche, A. Krasyuk, and J. Kirschner, Ultramicroscopy **159**, 520 (2015)