Journal of Electron Spectroscopy and Related Phenomena 169 (2009) 62–66

Contents lists available at ScienceDirect

Journal of Electron Spectroscopy and Related Phenomena

journal homepage: www.elsevier.com/locate/elspec

Spin-resolved two-photon photoemission on $Fe_{77}B_{16}Si_5$ alloy

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ARTICLE INFO

Article history:
Received 9 February 2007
Received in revised form 22 August 2008
Accepted 5 November 2008
Available online 17 November 2008

Keywords:
2PPE
Laser
Spin
Ferromagnetic

ABSTRACT

We have explored the spin polarisation of excited electrons in amorphous ferromagnetic iron–boron alloy by spin-resolved two-photon photoemission. These measurements were carried out with a spin resolved electron Time-of-Flight (ToF) analyser combined to a femtosecond laser source. The low kinetic energy was probed by caesium preparation of the sample surface. A constant spin polarisation of 18% was measured in the energy region between 1.3 and 3.2 eV above the Fermi level. The magnetic properties of the sample were characterised by a spin-resolved square hysteresis loop.

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1. Introduction

In ferromagnetic metals, the transport of hot electrons depends on the electron spin alignment (parallel or antiparallel) with the majority-spin direction: the so-called *spin-filtering* (SF) effect [1,2]. Various experimental techniques have been developed to measure the electron transmission versus the spin filter thickness in order to extract the mean free path (MFP) for both spin orientations. Spin-resolved photoemission [3–5] and free-electron ballistic transmission [6,7] have been carried out on thin ferromagnetic metal layers to probe the electron transport in the kinetic energy region 5–100 eV above the Fermi level. The very low kinetic energy region (1–2 eV) has been studied by ballistic-electron emission microscopy [8,9], spin valve transistor [10] and tunnelling magnetic transistor [11,12]. This energy region has been also characterised by time- and spin-resolved two-photon photoemission (2PEE) measurements [13–15]. In ferromagnetic metals, it was shown that the lifetime of the majority-spin state electrons is longer than that of the minority-spin state. Here, we investigate the spin polarisation of electrons excited into empty states localised between the Fermi and the vacuum levels by spin-resolved 2PEE. Previously, such measurements were performed with electrostatic electron analyser combined to fast laser source [15,16], for the first time we implement a spin-resolved electron Time-of-Flight (ToF) analyser. The efficiency of the measurement is improved by the ToF analyser where all the photoelectrons are energy and spin resolved simultaneously.

Half-metallic Heusler compounds have attracted great scientific interest in particular for their predicted magnetic properties at the Fermi energy. Wurmehl et al. [17] showed that in $Co_2Cr_{1-x}Fe_xAl$ alloy the Co–Cr disorder reduces the total magnetic moment by ferrimagnetic coupling and the high-energy photoemission reveals a clear difference between the bulk and the surface electronic structure. Furthermore, the surface spin polarisation of Co_2MnSi alloy at the Fermi energy does not exceed 12% at room temperature [18]. The band structure calculations predict a loss of the alloy half-metallic character as the Co–Mn disorder increases resulting in a lower degree of spin polarisation at the Fermi energy. Recently, the micro-magnetic domain structure of Co_2FeSi was resolved by X-ray magnetic circular dichroism and a Fermi energy single domain spin polarisation of 16% was reported [19]. The compound studied here is a ferromagnetic $Fe_{77}B_{16}Si_5$ alloy produced in thin flexible ribbon. The low kinetic energy region (0–20 eV above the vacuum level) was studied by spin-resolved electron scattering, a clear increase of the spin polarisation was observed at the vacuum level due to the electron cascade in the ferromagnetic metal [20]. Low energy (40 eV) spin-resolved photoemission revealed a strong coupling between the Fe-*d* states and the B-*p* states in the valence band [21].

2. Experimental

The light source utilised was a commercial regenerative Ti:sapphire laser system (Libra, Coherent), delivering up to 1 mJ photon pulses at 800 nm with a repetition rate of 1 kHz. This

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doi:10.1016/j.elspec.2008.11.002

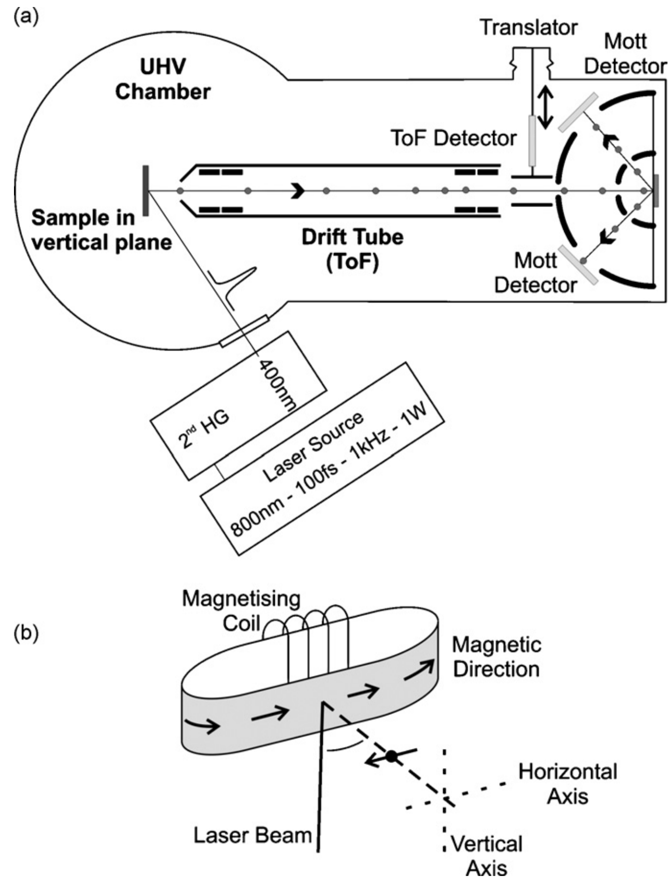

Fig. 1. (a) Diagram showing the experimental setup including the laser source, the Time-of-Flight and the Mott polarimeter. The spin-resolved/integrated mode of operation for the analyser is selected externally with the translator. (b) Diagram showing the ferromagnetic $Fe_{77}B_{16}Si_5$ amorphous alloy ribbon formed into a closed-loop with the magnetising coil positioned at the back. Normal emission is the optimal experimental geometry, permitting the measurement of both transverse components of spin polarisation of the photoelectrons.

fundamental beam was frequency-doubled in a 1-mm thick β-barium-borate crystal producing a 3.1-eV photon energy beam with an estimated pulse width of 140 fs. The beam was then focalised with a 50-cm focal lens onto the sample with a spot size of 1.4 mm diameter and an incident angle of 40° with respect to the surface normal. The spin-resolved measurement was performed with a maximal fluence of 0.2 GW/cm^2 on the sample.

Photoelectron spectroscopy was carried out using an electron Time-of-Flight Spin (ToF-Spin) analyser (Fig. 1a) in a UHV camber with a typical base pressure of 5×10^{-10} mbar. The ToF [22] analyser incorporates electron optics to decelerate electrons into a drift tube of 25 cm length, followed by another electron optic assembly to refocus the electron beam. A 25-mm diameter channel plate detector mounted on a translator (Fig. 1a) can be positioned at the focal point to perform spin-integrated measurements. Alternately, the translator can be raised to its high position, thus positioning a cylinder at the end of the ToF which permits the electron beam to enter the Mott polarimeter for spin-resolved measurements. The horizontal and vertical transverse spin polarisations of the electron beam are detected by a four-channel retarding field Mott polarimeter [23]. The spin selectivity of the Mott polarimeter is estimated at 15%. The experimental asymmetry is removed by reversing the magnetic state of the sample and repeating the measurement. The results presented in this paper are measured with a high voltage (200 V) between the sample and the (grounded) first electrode of the ToF input optics. This increases angular acceptance and collects most of the photoelectrons generated, and so perform an angle-integrated measurement. Following deceleration in the input electron optics, the electrons travel with 6 eV kinetic energy in the drift tube. The time taken for an electron to travel from the sample surface to any of the detectors is measured with a time digitisation of 120 ps, and then converted to kinetic energy. In the long time of flight region, consecutive data points are merged together to ensure a minimum kinetic energy variation of 20 meV between each point after conversion. The time-to-energy calibration is determined through a fit of the Fermi edge time position versus the sample voltages.

The amorphous $Fe_{77}B_{16}Si_5$ ribbon was manufactured using the melt-spinning process in a helium atmosphere. The surface was cleaned by argon ion bombardment without annealing. After few hours of sputtering cycles at 500 eV kinetic energy, the Fermi edge became sharper and clear spin polarisation was detected after each further cycles with good reproducibility. The sample was formed into a closed-loop, and an insulated wire used to form a coil around the ribbon at the rear of the sample holder to permit magnetisation of the sample (Fig. 1b).

3. Results and discussion

The spin integrated spectra measured in the spin-integrated mode (solid line) and spin-resolved mode (open circles) are presented in Fig. 2(a). Given the photon energy of $h\nu = 3.1$ eV, the portion of the spectra in the energy range below 1.47 eV is attributed to the 2PPE process where valence band electrons which are very close to the Fermi level are excited in the empty states. The Fermi edge is clearly visible at the sharp transition (1.47 eV). At low fluence (spin-integrated mode), with less than 0.2 photoelectrons per laser pulse detected within a very large acceptance angle, no space charge effects are expected. At higher fluence (spin-resolved mode) the total photoemission is estimated at 100 electrons per laser pulse. The fact that the Fermi edges for both spectra of Fig. 2(a) coincide is evidence that space charge effects are negligible in the spin-resolved measurements presented here, despite the low repetition rate of the laser source. At the high beam power used in the spin-resolved mode, a small distribution of hot electrons is observed between 2 and 4 eV kinetic energy. At the surface defects, enhancement of the optical field is expected, leading to hot spot-like of emission [24]. The high kinetic energy photoelectrons observed result of the coupling between the laser field and the localised surface plasmons. However, the relative contribution of these electrons is two order of magnitude smaller compared to the photoemission from Ag(1 0 0) surface [25]. The low level of high kinetic energy electrons suggests that emission from hot spot centres is presumably small here. The Fermi energy (1.47 eV kinetic energy) is deduced from the fit of the Fermi edge (inset Fig. 2(a)) with a room-temperature Fermi–Dirac distribution convoluted with a Gaussian profile. Using the relation $\Phi = 2h\nu - E_F$, the vacuum level of the alloy is evaluated at 4.7 eV. This is in good agreement with the 4.4-eV vacuum level for the clean iron surface [26]. The standard deviation (σ) of 98 meV for the Gaussian characterises the energy resolution which is limited by the large acceptance angle of the measurement. At 0 eV kinetic energy, a vacuum cut-off is observed and a minor contribution from scattered electrons with a longer electron flight path is plotted at negative kinetic energy. In spin-resolved mode, fewer scattered electrons are detected due to the small entrance aperture of the Mott polarimeter (3-mm diameter), thus reducing this undesirable dilution effect on the spin polarisation by unpolarised scattered electrons. Fig. 2(b) shows the variation of the electron yield with respect to beam power, each data point is the integral of the electron spectrum measured at that power. The measurements agree well with a

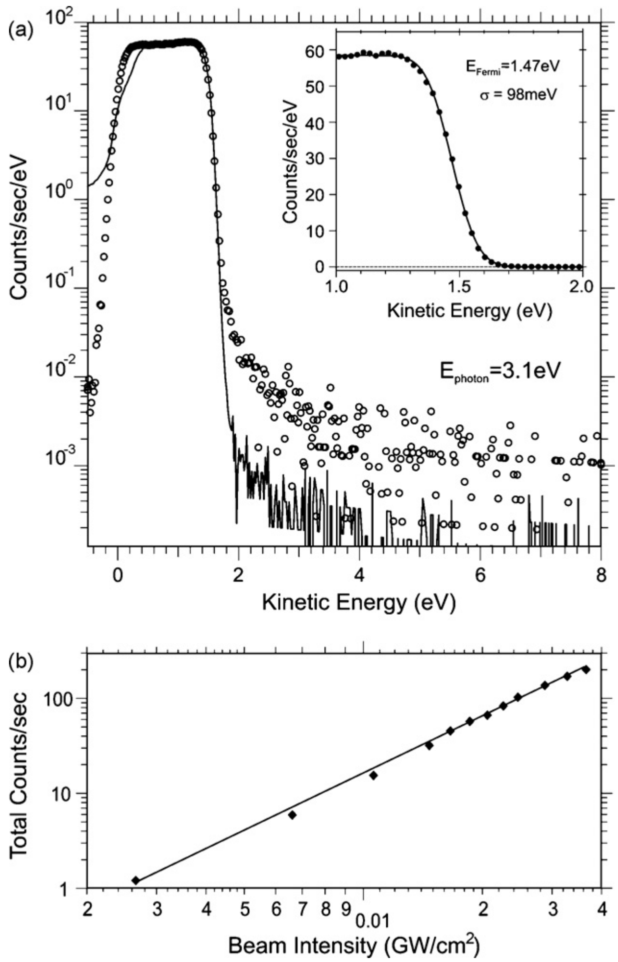

Fig. 2. (a) Two-photon photoemission electron distribution curves measured in spin-integrated mode (solid line), and spin-resolved mode (open circles). To compensate for the intrinsic loss in the Mott polarimeter, the beam power is increased up to 0.19 GW/cm^2 (spin-resolved mode). A small contribution of hot electrons is observed above 2 eV kinetic energy at high beam power (open circles). (b) Variation of the total statistic with respect to probe beam power, measured in spin-integrated mode. The solid line is the power square fit of the measurements.

power-square function (solid line), this being the signature of the 2PPE process.

Electron spin polarisation is defined by the relation $P=(N_\uparrow-N_\downarrow)/(N_\uparrow+N_\downarrow)$, where $N_\uparrow(N_\downarrow)$ is the number of photoelectrons with a spin up(down) orientated along a given direction. The spin-integrated spectrum is obtained by averaging the four detector signals of the Mott polarimeter. In normal-emission geometry, the complete transverse spin polarisation is resolved by the Mott polarimeter. Fig. 3(a) shows the horizontal and vertical components of spin polarisation measured over the valence band, and the spin-integrated spectrum with respect to the electron excitation energy after absorption of the first photon involved in the 2PPE process. The horizontal spin polarisation is relatively constant (18%) across the spectrum and decreases slightly at the Fermi edge. The degree of spin polarisation obtained is in very good agreement with the Co_2FeSi 2PPE spin polarisation reported by Wüstenberg et al. [27]. With a temporal width of the laser pulse longer than the electron lifetime at 3.1 eV excitation, the electron escape depth is expected to be of the order of the mean free path (MFP), i.e. few nm [9]. Consequently the spin polarisation measured has an important bulk contribution. However the spin polarisation measured here remained small compare to the surface spin polarisation obtained at higher photon energy on Co_2FeSi [19].

A reduced spin polarisation is expected from hot spot-like centres due to the higher effective temperature of the electrons. Large contribution of such centres would lower of the total spin polarisation measured. Band structure calculations and experimental studies [17,28] reveal that the minority-spin states conduction gap is strongly reduced by atomic disorder in alloy which leads to a lower spin polarisation at the Fermi energy. Indeed, we attribute the reduced spin polarisation observed here to the amorphous character of the sample. The highest spin polarisation reported so far was obtained on highly ordered $Co_2Cr_{0.6}Fe_{0.4}Al$ surface [28]. The small vertical spin component measured is due to macroscopic defects present in the sample producing magnetic domains tilted with respect to the horizontal axis. The magnitude of this spin component is strongly dependent on the sample area excited.

The variation of the total spin polarisation (obtained by integrating the electron spectra) with respect to the pulsed magnetic field amplitude is presented in Fig. 3(b). The horizontal spin polarisation describes an almost-square hysteresis loop, characterised by a very low coercive field of around 0.5 Gauss with a 100% remanence ratio. Similar variation is observed on the vertical spin component illustrating the small tilt of the total spin polarisation from

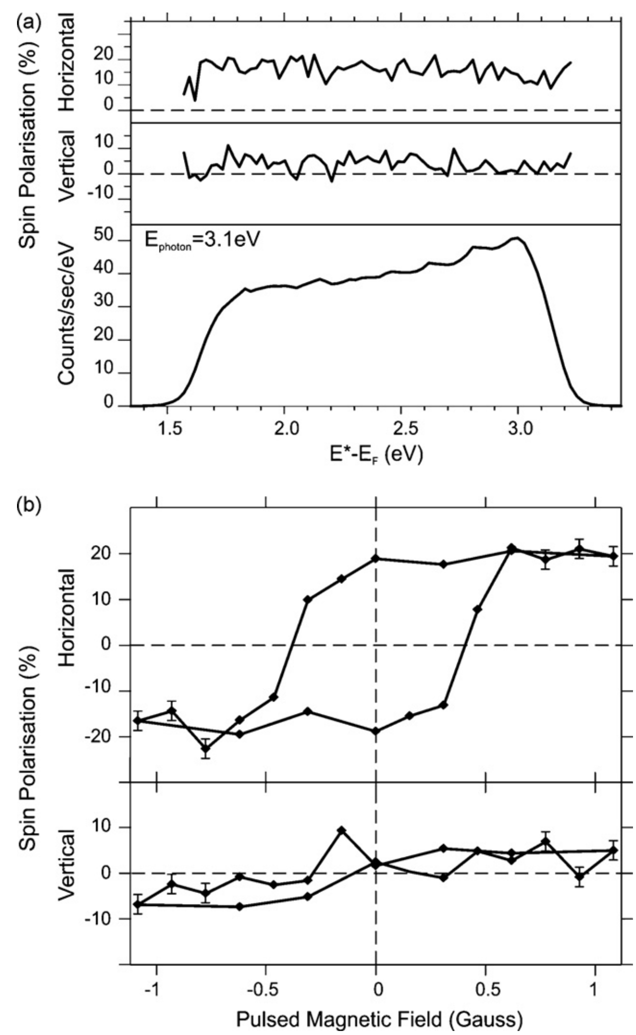

Fig. 3. (a) Horizontal and vertical spin polarisations with the source electron spectrum measured during two-photon photoemission excitation (E_{photon} = 3.1 eV). A clear horizontal spin polarisation is observed coincident with the magnetisation axis of the sample. (b) Variation of the average spin polarisation of the electron spectrum with respect to the amplitude of the pulsed sample magnetising field. A clear hysteresis loop is observed in the horizontal spin polarisation. The measurement was performed with the sample in its remnant state.

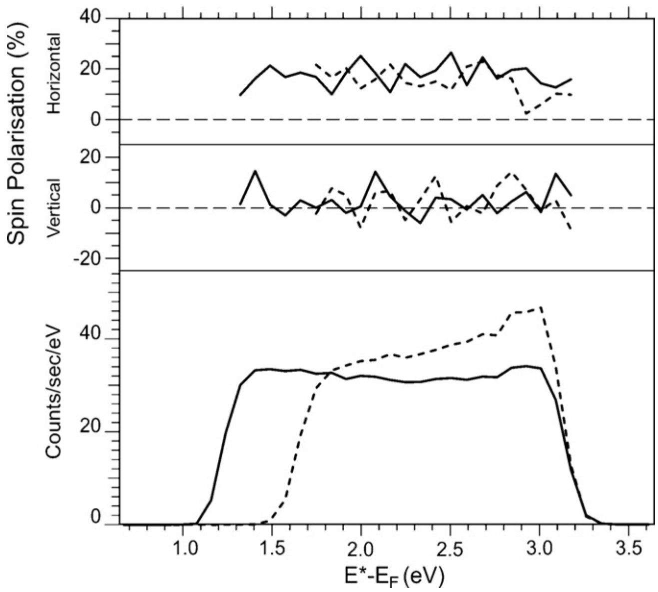

Fig. 4. Horizontal and vertical spin polarisation and corresponding electron spectra measured before (dashed line) and after (solid line) Cs evaporation. The work function of the sample is seen to fall from 4.7 to 4.2 eV. The spin polarisation remains constant between, before and after Cs evaporation, whilst the electron distribution curve clearly falls at the Fermi edge.

the horizontal axis. As the amplitude of the pulsed magnetic field approaches the coercive field threshold, the magnetisation state is reversed by domain nucleation and growth. The high remanence exhibited by this sample is crucial for photoemission measurements involving very low kinetic energy electrons, as no external magnetic field can be applied in the interaction region during the measurements.

It is well known that for clean transition metal or semiconductor surfaces, the vacuum level of the sample can be lowered by the evaporation of caesium combined with fine oxygen dosing, but this has not been tested on these types of alloys. The spin-resolved and spin-integrated spectra measured before (dashed line) and after (solid line) the Cs preparation are presented in Fig. 4 with an energy discrimination of 80 meV. The clear shift at the left-hand edge of the spectrum corresponds to the variation of the vacuum level from 4.7 to 4.2 eV, thus permitting the detection of electrons with lower kinetic energy. After Cs preparation the spectrum is more uniform and the Fermi edge is less pronounced. In the kinetic energy region 1.3–2 eV above the Fermi level the spin polarisation measured remains constant where a negative spin polarisation would be expected due to the minority-spin *d*-band populated by the first photon. The spin polarisation measured differs from the density of state polarisation of the intermediate state. Within the laser pulse duration the intermediate state spin polarisation is altered by the spin dependant relaxation time of the photoexcited electron population. Knorren et al. [29] report a ratio up to 2 between the majority and minority spin lifetime in 3*d* transition metals, measured by spin and time resolved 2PPE. The comparison with calculation of hot electron relaxation time [29] reveals that the electron lifetime depends on the electron–electron scattering and the magnitude of the Coulomb matrix elements. The spin polarisation measured is characteristic of the spin filtering effects occurring in the intermediate state of the 2PPE process. Within the limit of the noise level, no depolarisation of the photoelectron from the alloy is observed between both measurements. The Cs sub-layer has a filtering effect on the photoelectrons, regardless of their spin orientation (i.e., no spin filtering effect), whilst for higher Cs overlayer coverage, magnetic coupling of the Cs with the Co substrate has been observed [30]. As the Cs evaporation time is increased, space charge effects become dominant, and a clear energy shift in the Fermi edge of hundreds of meV is apparent, of with a corresponding reduction of the spin polarisation.

4. Conclusions

The spin-resolved 2PPE technique was applied by combining an electron ToF-Spin analyser with a fs-laser source to measure the spin polarisation of hot electrons in a ferromagnetic amorphous $Fe_{77}B_{16}Si_5$ alloy. The vacuum level of the sample was lowered by Cs preparation of the surface. In the energy range 1.3–3.2 eV above the Fermi level, a constant spin polarisation of 18% was detected. The spin-resolved hysteresis loop shows a low coercive field and a 100% remanence ratio. The low degree of polarisation was attributed to the amorphous character of the sample. At 3.1 eV photon energy, the 2PPE measurements are particularly bulk sensitive and the photoelectron spin polarisation is mainly governed by the spin dependant recombination channels of the electron in the intermediate state. Further time- and spin-resolved measurements should be carried out to explore the lifetime of both spin states of the excited electrons.

Acknowledgements

We gratefully acknowledge support from UK EPSRC (grant GR/M50447), CCLRC and the EUFEL project (ELETTRA). The assistance of G. Miller from Daresbury Laboratory is fully appreciated.

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