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Spin-dependent electron dynamics investigated by means of time- and spin-resolved photoemission

M. Aeschlimann[a,*], R. Burgermeister[b], S. Pawlik[a], M. Bauer[a], D. Oberli[b], W. Weber[b]

[a]*Laboratory for Technical Chemistry, ETH Zürich, CH-8092 Zürich, Switzerland*
[b]*Laboratory for Solid State Physics, ETH Zürich, CH-8092 Zürich, Switzerland*

Abstract

The power of time- and spin-resolved two photon photoemission is demonstrated. This method allows the determination of the spin-dependent decay of an excited electron population in ferromagnetic materials by means of a real time experiment. The lifetime of majority-spin electrons is found to be longer compared with that of minority-spin electrons for both cobalt and iron. This study shows that the relaxation dynamics of excited electrons is strongly influenced by the imbalance between majority- and minority-spin electrons in a ferromagnet. © 1998 Elsevier Science B.V.

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1. Introduction

Combined with ultra-fast laser techniques, time-resolved two-photon photoemission (TR-2PPE) is one of very few techniques that enables us to study the state of the electron system during the course of its transformation from a nascent (as photoexcited) non-thermal electron distribution to an *excited* Fermi Dirac distribution [1]. The data are obtained by means of a pump–probe experiment in which the first laser pulse pumps electrons into an unoccupied intermediate state, followed by a second pulse which subsequently photoemits the excited electrons (see Fig. 1). By varying an optical delay line between the two pulses, the population decay of the intermediate (unoccupied) states can be determined. A stringent condition is that the photon energy is smaller than the work function in order to avoid one-photon photoemission, which would completely obscure the 2PPE spectrum. TR-2PPE measurements of metals are challenging because they have to be performed in a time domain of only a few femtoseconds (fs). Only recently, Schmuttenmaer et al. demonstrated the feasibility of this technique by studying the relaxation of single excited electrons at the Cu(100) surface [2]. By using state-of-the-art laser technology, it is possible to investigate electron dynamical processes at surfaces with a time-resolution below 5 fs [3–5].

The relaxation time of electronic excitations in solids is controlled by the available decay channels including collisions with other electrons, holes, phonons, plasmons, defects and impurities. In particular, the inelastic relaxation depends critically on the electronic band structure. As a consequence, in ferromagnetic materials, where the band structure separates into a majority- and a minority-spin part, electrons scatter differently depending on their spin. A spin-dependent relaxation has important implications: e.g. the interpretation of spin-polarized spectroscopy of ferromagnetic materials must take

* Corresponding author.

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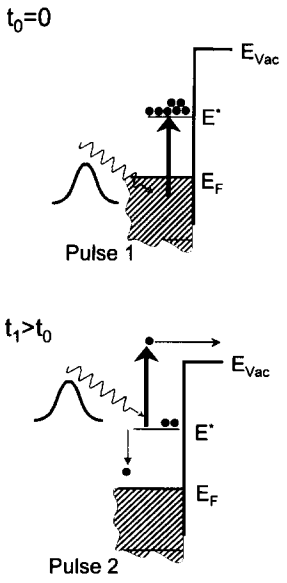

Fig. 1. A schematic figure of the time-resolved two-photon photoemission process based on a pump probe approach, where the energy level E^* represents the intermediate state, E_{vac} the vacuum energy and E_F the Fermi energy.

into account a spin-dependent transport of the excited electrons to the surface, known as the spin filter effect [6–10].

We attached a spin analyzer to a conventional time-resolved 2PPE set-up in order to investigate the spin-dependent electron dynamics in ferromagnetic metals. We report our investigation of bcc Fe and fcc Co grown on a Cu(001) single crystal. A faster relaxation of minority-spin electrons compared with majority-spin electrons is found for both ferromagnets.

2. Experimental set-up

The light source is a femtosecond Ti:sapphire laser system (Tsunami, Spectra Physics), which delivers transform-limited and $sech^2$ temporally shaped pulses of up to 15 nJ/pulse with a duration of $\leq$ 40 fs at a repetition rate of 82 MHz. The linearly polarized output of the Ti:sapphire laser is frequency doubled in a beta barium borate (BBO) crystal, 0.2 mm thick, to produce UV pulses between 3 and 3.4 eV. In order to obtain a 2PPE spectra, the beam is focused directly on the sample surface and the number of photoelectrons, as a function of their kinetic energy, is determined. The polarized laser beam is incident on the sample at an angle of 45° with respect to the surface normal and the electrons are detected in normal emission geometry. In order to compare the 2PPE spectra, taken with $h\nu$ = 3 eV, with the equivalent (regarding the final state energy) one-photon photoemission (1PPE) spectra, the fundamental light frequency is quadrupled using a 0.5 mm LBO crystal in a first stage and a 1 mm thick BBO crystal in a second stage to generate a photon energy of 6 eV.

For the time-resolved experiment, the pulses are split by a beam splitter into two beams of equal intensity (pump and probe pulses) and one path is delayed with respect to the other by a computer-controlled delay stage. Both beams are combined colinearly—but cross-polarized—by a second beamsplitter and are focused on the sample surface. The time-averaged photocurrent at a fixed kinetic energy is measured as a function of the delay between the two beams (two pulse correlation technique). The nonlinearity of the two-photon process leads to an increase in the 2PPE yield when the pulses are spatially and temporarily superimposed. As long as the two laser pulses temporarily overlap it is obvious that an electron can be emitted by absorbing just one photon from each pulse. However, if the pulses are temporarily separated, then an excited electron from the first pulse is able to absorb a photon from the second pulse only as long as the inelastic lifetime of the intermediate state exceeds the delay. Due to a precise measurement of the delay between the two pulses, this technique allows us to analyze lifetimes which are considerably shorter than the laser pulse duration.

Laser pulses at low fluence and peak power are used to avoid space charge effects or highly excited electron distributions. We emphasize that the count rate is much lower than one electron per pulse. Therefore, the relaxation of individual excited electronic states rather than the collective behavior of transiently heated nonequilibrium distribution is measured.

The samples are mounted in a UHV chamber equipped with a cylindrical sector electron energy analyzer. To investigate the electron dynamics separately for both spin directions, we mounted a spin analyzer (SPLEED, Focus, Germany) on top of the electron energy analyzer. This enables the measurement of one in-plane component of the spin-polarization vector. A bias voltage of −14 V is applied

to the sample to eliminate the effects of any stray electric and magnetic fields.

Iron and cobalt films are grown on a Cu(100) crystal at room temperature. The cleanliness of the Cu surface is examined by Auger spectroscopy and its crystal structure by low-energy electron diffraction (LEED). During evaporation the pressure is kept in the 10^{-10} mbar region. The thickness of the film was measured by a calibrated quartz thickness monitor. The growth rate is 2 Å/min for both iron and cobalt. Note that both iron and cobalt films have an easy magnetization axis in the plane. After deposition, the film is magnetized remanently by applying a current pulse through a coil mounted close to the sample in order to establish a single-domain magnetization. The presented technique is not restricted to a magnetic film structure but can also be applied to bulk magnetic systems. In this case, however, a magnetic field may be required to maintain the single-domain magnetization. The use of a magnetic frame crystal may overcome this problem.

The clean metal surfaces are first dosed with Cs to lower the surface work function, a well-known technique. This enables lifetime measurements of lower excited states, increasing the useful energy range of the spectra closer to the Fermi energy. Cs is evaporated from a thoroughly outgassed getter source (SAES, Milano, Italy). No differences in spin polarization and lifetime measurements using cross-polarized pulses are found between the clean and the cesiated surfaces in the overlapping energy region.

3. Results and discussion

Fig. 2 shows the spin polarization P of a bcc iron film, 100 Å thick, obtained in a 2PPE experiment ($h\nu$ = 3eV) as compared with the equivalent 1PPE experiment ($h\nu$ = 6 eV). All data were recorded at room temperature. The spin polarization P is defined as $P = (N\uparrow - N\downarrow)/(N\uparrow + N\downarrow)$ with $N\uparrow$ ($N\downarrow$) as the number of photoemitted electrons with their magnetic moment parallel (antiparallel) to the sample magnetization. It is remarkable that the spin polarization values for 2PPE at lower energies are substantially higher than values obtained in the equivalent 1PPE experiments. Such an enhancement was also found in fcc cobalt films [11]. In spite of the fact that different

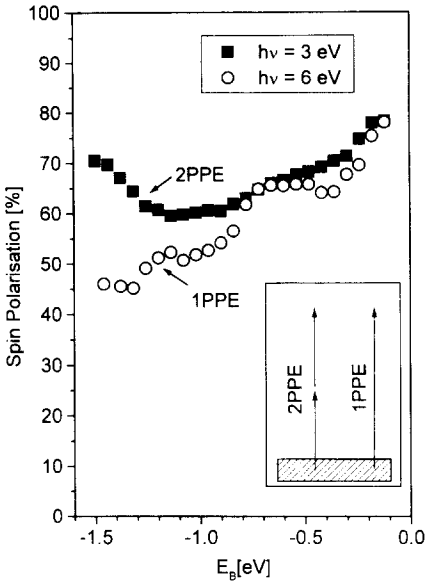

Fig. 2. Spin polarization of a bcc iron film versus binding energy E_B obtained by means of a 2PPE ($h\nu$ = 3 eV) and a 1PPE ($h\nu$ = 6 eV) measurement.

excitation processes and, hence, different transition matrix elements have to be considered in 2PPE compared with the normal photoemission, this substantially enhanced spin polarization for 2PPE at lower energies might be attributed to the existence of a spin filter effect in ferromagnetic metals, resulting in a change in the ratio $N\uparrow/N\downarrow(E)$ of the excited electrons during the first and the second excitation process. The increase in P for iron on approaching the Fermi energy E_F is consistent with normal photoemission experiments on bcc iron and is attributed to the strong spin-dependent asymmetry of the density

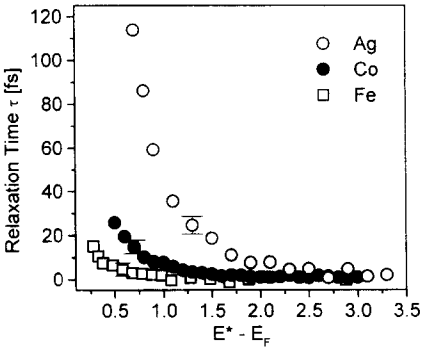

Fig. 3. Spin-integrated relaxation time τ of Ag, Co and Fe as a function of the intermediate state energy E^* above E_F.

of states around E_F [12]. This is contrary to fcc cobalt, where a strong drop in P towards E_F is found [11]. Compared with iron, however, cobalt is a strong ferromagnet and, hence, has no majority-spin density of states at E_F.

Fig. 3 shows the spin-integrated depopulation time τ of both cesiated iron and cobalt as a function of the intermediate state energy E^*. For comparison, the values for the noble metal silver are also plotted. The data are extracted from the experimentally obtained cross-correlation traces using a rate equation model for the population of the intermediate state. In this case, the evolution of the transient population $N^*(t)$ of the intermediate state is given by $dN^*(t)/dt = A(t) - N^*(t)/\tau$ where $A(t)$ is the excitation induced by the first (pump) laser pulse. All curves show a clear increase in the relaxation time τ on approaching E_F. This trend can be explained by the fact that according to the conventional Landau's Fermi liquid theory, the available phase space for scattering of an excited electron with electrons from the 'cold' Fermi sea changes proportional to $(E^* - E_F)^2$ [13]. As the energy of the intermediate state approaches E_F, less phase space is available and hence the relaxation slows down.

The data show that the lifetimes in iron and cobalt are substantially shorter than in silver. This can also be explained by the phase space argument. Compared with the noble metal silver, in which the d-band is completely filled and localized well below the Fermi energy, the d-band of the transition metals iron and cobalt are only partially filled and thus increase the available phase space for electron–electron scattering [10]. This shows that the relaxation is dominated to a considerable degree by the density of the occupied and unoccupied states near E_F. Fe ($3d^6$) has one additional unoccupied d-orbital compared with Co ($3d^7$) and thus a slightly faster relaxation is expected for iron. The data, however, indicate a difference in the relaxation of iron and cobalt by a factor of almost 2, which is much larger than expected. It is even more striking that the relaxation of iron is faster than that of Ta ($5d^3$) [3], which has nearly twice as many unoccupied d-states as iron. Our simple model, however, neglects screening as well as differences in cross-sections between singlet ($\uparrow\downarrow$) and triplet ($\uparrow\uparrow$) electron–electron scattering processes.

Fig. 4 displays the spin-dependent relaxation decay

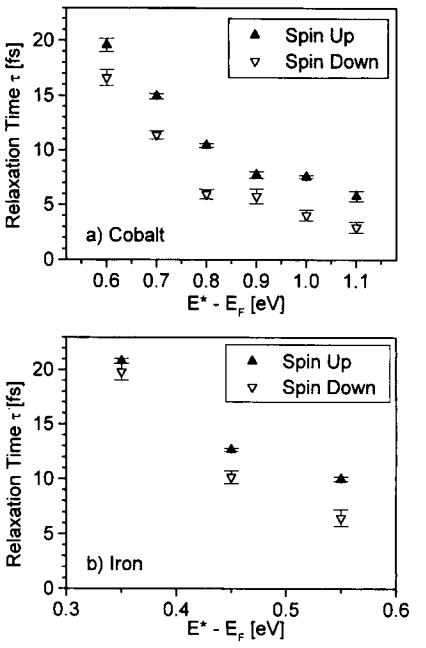

Fig. 4. Spin-resolved relaxation-time τ for (a) fcc Co and (b) bcc Fe. For both ferromagnets, the population of excited minority-spin electrons decays faster than that of majority-spin electrons within the investigated energy range.

for (a) cobalt and (b) iron. The data show a clear difference in relaxation time between the minority-spin and the majority-spin electrons. The error bars in the plot represent the statistical scatter. It is expected that the spin-dependence of the relaxation rates is stronger for cobalt than for iron in the investigated energy range. Cobalt is a strong ferromagnet and has no majority-spin density of states above E_F. In contrast, the majority-spin d-band of bcc iron is not fully occupied. In fact, the band structure data for iron even predict a change in the sign of $R = (\tau^{\uparrow} - \tau^{\downarrow})/(\tau^{\uparrow} + \tau^{\downarrow})$ around $(E^* - E_F) = 1$ eV (W. Wolf and J. Noffke, private communication). Below this energy, the integrated density of unoccupied states in the majority-spin band becomes larger than that of the minority-spin band. Indeed, giant magnetoresistance (GMR) studies show a longer mean free path for the majority-spin electrons in cobalt, whereas in iron, the mean free path of minority-spin electrons is larger [14]. Approaching the Fermi energy, the data for iron indicate that the ratio R becomes smaller. A change in the sign of R is, however, not observed below $E^* - E_F = 1$ eV, i.e. $\tau^{\downarrow} < \tau^{\uparrow}$. It should be

noted that the relaxation time τ of photoexcited electrons is governed not only by the inelastic lifetime of the excited electrons, but also by secondary electrons (Auger and cascade electrons) and the transport of electrons out of the probed region. Further work and simulation of time evolutions of hot electron distribution on surfaces is in progress in order to investigate the influence of these two effects on the dynamical properties.

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