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Spin- and time-resolved photoemission studies of thin Co_2FeSi Heusler alloy films

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Abstract

We have studied the possibly half-metallic Co_2FeSi full Heusler alloy by means of spin- and time-resolved photoemission spectroscopy. For excitation, the second and fourth harmonic of femtosecond Ti:sapphire lasers were used, with photon energies of 3.1 and 5.9 eV, respectively. We compare the dependence of the measured surface spin polarization (SSP) on the particular photoemission mechanism, i.e. 1-photon photoemission (1PPE) or 2-photon photoemission (2PPE). The observed differences in the spin polarization can be explained by a spin-dependent lifetime effect occurring in the 2-photon absorption process. The difference in escape depth of the two methods in this context suggests that the observed reduction of spin polarization (compared to the bulk) cannot be attributed just to the outermost surface layer but takes place at least 4–6 nm away from the surface.

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The success of modern spintronics devices, i.e. devices relying on the electron spin as carrier of information, depends crucially on the ability to store, transport and manipulate the spin state of an electron within a properly chosen material system. While storage is traditionally accomplished by the use of magnetoresistive effects in magnetic multilayer systems, the latter problems seem to be trackable by the use of (partly) semiconductive materials, where typical spin diffusion lengths are much larger and material properties can be tuned precisely to establish a coupling to external fields for coherent spin manipulation purposes [1]. However, the conductivity mismatch [2,3] between ferromagnetic metals and semiconductors, which hinders efficient spin injection, is still an unsolved issue. A promising class of materials to overcome this issue consists of half-metallic Heusler alloys, exhibiting metallic behavior for one-spin direction and a band gap for the other. This leads to a full-spin polarization P at the Fermi level E_F, which is commonly defined as the normalized difference of spin up and spin down electron occupation numbers with respect to a certain energy and given quantization axis

$$P(E) = \frac{n_\uparrow(E) - n_\downarrow(E)}{n_\uparrow(E) + n_\downarrow(E)}. \quad (1)$$

Fabrication of such alloys with predictable bulk properties is nowadays routinely possible. This is shown by the increasing coherence between measured and predicted values for the element-specific number of magnetic moments μB per unit cell [4,5]. However, the connection to the spin polarization as the relevant parameter is not straightforward and relies heavily on the chosen model potential in calculations. In the case of spin transport between different materials the interface region is of extreme importance, since various mechanisms may drastically reduce the spin polarization of electrons in the surface region. To our knowledge, the predicted value of $P = 100\%$ has not yet been observed in Heusler compounds. Moreover, the value of the surface spin polarization (SSP) depends on temperature. Surface-sensitive measurements performed on Co_2MnSi yield values no more than 12% at room temperature, and only at low temperatures (<20 K) values up to 60% have been

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reported ([6], and references therein). For the half-Heusler compound NiMnSb, Kolev and coworkers [7] excluded surface states, chemical disorder as well as structural defects as sources of reduced SSP. Our experiments with spin-resolved photoemission techniques indicate that also for thin films of the full-Heusler compound Co_2FeSi (CFS), the spin polarization of electrons emitted from the surface at E_F is comparable to the results above. We observe differences in the absolute SSP value, depending on the employed photo-excitation scheme (1-photon photoemission (1PPE) and 2-photon photoemission (2PPE)). With a simple theoretical model, we show that these differences can be explained by the so-called spin-filter effect [8,9]. We conclude that the SSP loss is not a single-layer effect but extends over a distance of at least four times the escape depth of standard photoemission, i.e. 4–6 nm.

For our measurements, we used both conventional spin-polarized photoemission spectroscopy (SP-1PPE) as well as spin-polarized two-photon photoemission spectroscopy (SP-2PPE). The two excitation schemes are displayed in Fig. 1. In a conventional SP-1PPE experiment (right panel in Fig. 1), an electron is excited directly from its ground state $|g\rangle$ below the Fermi energy E_F into an evanescent state $|f\rangle$ slightly above the vacuum level E_{vac} ($\hbar\omega \gtrsim \Phi$, Φ = work function). After passing a kinetic energy filter (Focus CSA 300), the longitudinal in-plane SSP component is analyzed by means of spin-polarized low-energy electron diffraction at a tungsten crystal (Focus SPLEED). The spectra were recorded at room temperature using the fourth harmonic of a pulsed-Ti:sapphire oscillator (Spectra Physics Tsunami), created by two-fold frequency doubling within two thin β-barium-borate (BBO) crystals. The average energy per pulse is 37 nJ at $\hbar\omega = 5.9$ eV. In SP-2PPE, the excitation is accomplished via two-photon absorption within a single-laser pulse. In this process, the first photon excites an electron from its ground state $|g\rangle$ below E_F into an intermediate state $|i\rangle$ below E_{vac}. In this state, dephasing as well as decay processes take place, both being potentially spin dependent [8]. The second photon is used to excite the electron into the final state $|f\rangle$ known from the 1PPE process. In this experiment, we used the frequency-doubled output of a Ti:sapphire oscillator (Femtosource) with photon energy $h\nu = 3.1$ eV, pulse width of 40 fs and 10 μJ average energy per pulse. Since the electron in state $|i\rangle$ may propagate prior to photoemission, the escape depth of 2PPE is larger than for 1PPE. Thus, we can conclude on the extension of the spin-depolarizing surface layers by comparing the spectra obtained by the two methods.

We used a CFS Heusler alloy for our investigations. CFS alloys possess a high magnetic moment of almost 6 μB per unit cell, as well as a high Curie temperature of 1100 K [4,5]. A 70 nm thin film was grown epitaxially on a MgO(1 0 0) substrate. The 4 nm Al cap layer was removed under UHV conditions in situ by sputtering with 500 eV Ar^+ ions. Reproducible results were obtained, if the sample was sputtered and annealed (at 570 K) several times prior to the measurements. The details of the preparation process and a full characterization of the sample are described elsewhere [10]. Here, we just note that both Auger as well as LEED measurements indicate a clean and well-ordered surface. For the experiments the sample was magnetized remanently along the longitudinal axis by an external in-plane magnetic field.

The SP-1PPE spectra obtained in normal emission geometry show the longitudinal in-plane SSP component and are displayed in Fig. 2, together with the corresponding electron yield. The energy values in both spectra refer to the same ground state $|g\rangle$ of the electron. Intermediate state energies can be obtained by adding the photon energy (3.1 eV) to the ground-state energy. The spin-integrated spectra do not show any prominent features, over an energy range down to 1.4 eV below E_F. In contrast to the electron yield data, the SSP data show a more structured behavior, indicating the spin dependence of the band

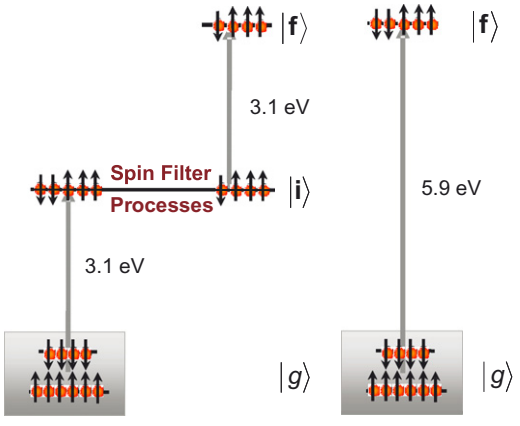

Fig. 1. Photoemission schemes used in this work. Left panel: 2-photon absorption (2PPE) within one single pulse. Right panel: conventional photoemission (1PPE).

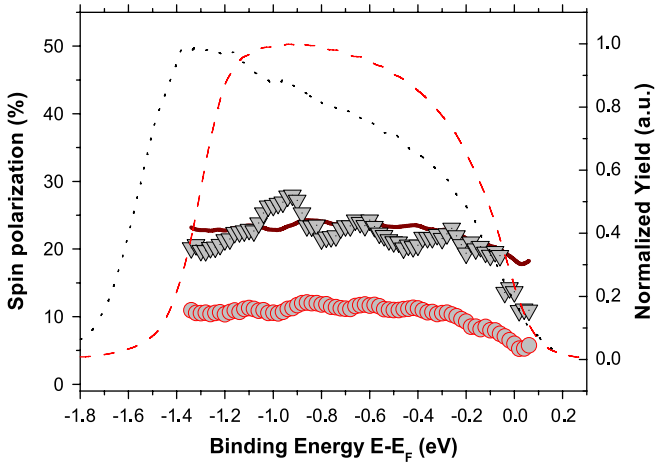

Fig. 2. SP-1PPE (red circles), SP-2PPE (black triangles) and simulated SP-2PPE data (blue full line). The dashed and dotted lines show the corresponding spin integrated 1PPE and 2PPE spectra, respectively.

structure in the magnetic compound. The spin polarization values for both methods are almost constant for lower energies and decay both to approximately half of their low-energy value when approaching the Fermi level.

Comparing the SSP spectra for SP-1PPE and SP-2PPE, we observe a general similarity. However, the spin polarization for SP-1PPE is lowered by a factor of 2 compared to the SP-2PPE data. Similar effects were obtained for the itinerant ferromagnet iron and could be explained by a spin filter effect for hot electrons [8,9]. This effect is caused by spin-dependent electron relaxation processes in the intermediate state $|i\rangle$, which can take place at timescales comparable or even smaller than the temporal width of the probing laser pulse. Since, in ferromagnetic materials, the minority (spin down) electrons decay faster than the majority ones, the resulting intermediate-state polarization increases even within a single-laser pulse.

In order to quantify the impact of the spin-filter effect on the increase of polarization in the SP-2PPE data, we developed a simple numerical model containing the basic ingredients of our experiment. First, we calculate the polarization in an intermediate state by evaluating the time-dependent numbers of up and down electrons in a rate equation model

$$\dot{n}_{\uparrow,\downarrow} = D_{\uparrow,\downarrow} p(t) - n_{\uparrow,\downarrow}/\tau_{\uparrow,\downarrow}. \quad (2)$$

Here, $n_{\uparrow,\downarrow}$ is the occupation number for majority and minority electrons, $\tau_{\uparrow,\downarrow}$ the spin-dependent electron lifetimes in the intermediate state, $p(t)$ a Gaussian-shaped pulse with given full width at half maximum (FWHM) and $D_{\uparrow,\downarrow}$ a density of states-type factor taking care for the different spin up and spin down density of states in the ground state. It can be obtained from the measured SP-1PPE data points by setting the ratio of $D_\uparrow$ and $D_\downarrow$ such that the polarization value of the measurement is reproduced. The spin-integrated lifetimes $\tau_{\uparrow,\downarrow}$ were measured with time-resolved 2PPE, an experimental technique described in detail in Ref. [8]. The lifetimes are shown in Fig. 3. The ratio of $\tau_\uparrow/\tau_\downarrow$ was determined in a bichromatic spin- and time-resolved experiment (STR-2PPE), revealing an almost energy-independent ratio of 1.3. Since in our case the photons of a single pulse are used, the final SP-2PPE spectra can be calculated from the solution of (2) by simply integrating over a second, identical pulse profile. The resulting simulated SP-2PPE spectrum is displayed in Fig. 2 (blue full line). One can state that the measured SP-2PPE polarization is well reproduced, in spite of local effects caused by the particular density of states in the intermediate state in the 2PPE process. This is an interesting result for general interpretation of SP-1PPE data. To draw conclusions regarding the possible use in spintronics we want to take a closer look on the origin of the observed electrons within the thin film.

The lifetime of an excited electron in the final state $|i\rangle$ within the crystal is very short (less than 3 fs), causing the strong-surface sensitivity of 1PPE data. This timescale is

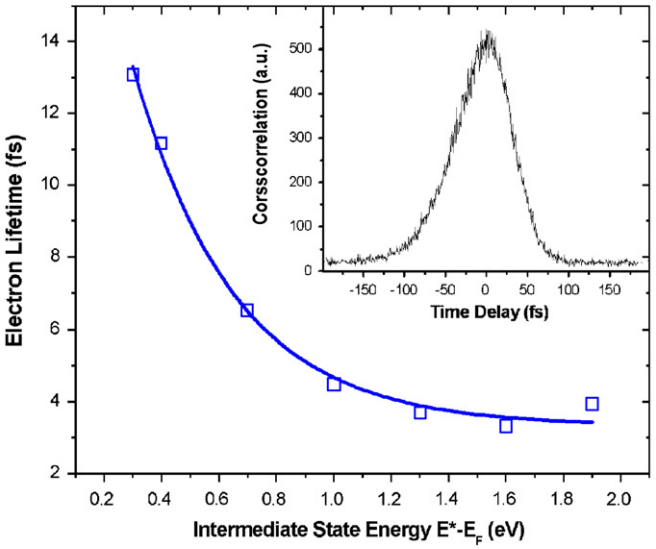

Fig. 3. Spin-integrated electron lifetimes (blue squares) and exponential fit (blue line) for intermediate states at E^* ($E_g = E^* - 3.1$ eV). Inset: example bichromatic STR-2PPE cross correlation data used to extract $n_{\uparrow,\downarrow}$.

short even compared to the laser FWHM. For a corresponding 2PPE process, we have to study the influence of the intermediate state at $E^* - E_F \sim (E(|f\rangle) - E_F)/2$. From standard Fermi liquid theory, the lifetime of this state can be estimated to be four times the one at the final-state energy. Assuming that the lifetime is proportional to the distance the electron can travel (within the laser width), we conclude that the mean depth of origin of electrons emitted in a 2PPE process must be approximately four times larger than the one for 1PPE. In other words, the possible loss in spin polarization is not likely to occur exclusively at the very surface but must take place deeper than roughly four times the information depth of 1PPE, the latter corresponding to a layer of 1–2 nm. Since the measured differences in SSP magnitude for the full-Heusler Co_2FeSi can be explained solely by lifetime effects we conclude that the experimentally observed loss in SSP must extend over a range of up to 4–6 nm, i.e. the escape depth for 2PPE.

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