Ultrafast Spin-Dependent Electron Dynamics in fcc Co

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The spin dependence of the lifetime of electrons excited in ferromagnetic cobalt is measured directly in a femtosecond real-time experiment. Using time- and spin-resolved two photon photoemission, we show that the lifetime of majority-spin electrons at 1 eV above the Fermi energy is twice as long as that of minority-spin electrons. The results demonstrate the feasibility of studying spin-dependent electron relaxation in ferromagnetic solids directly in the time domain and provide a basis for understanding the dynamics of electron transport in ferromagnetic solids and thin films. [S0031-9007(97)04853-9]

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There is much indirect evidence showing that the inelastic lifetime τ of electrons excited in ferromagnetic materials is different for majority-spin ($\uparrow$) compared to minority-spin ($\downarrow$) electrons. This spin-dependent lifetime has important consequences such as the occurrence of giant magnetoresistance in coupled multilayers [1]. Initial evidence that $\tau^{\uparrow} \neq \tau^{\downarrow}$ stems from measurements such as the enhancement of secondary electron spin polarization at low kinetic energies [2–4]. More recent experimental information is based on spin-resolved overlayer experiments which investigate the transmission of electrons through ultrathin ferromagnetic metal films as it depends on the spin state [5–13].

On the theoretical side, efforts have been made to explain the enhancement of the secondary electron spin polarization by spin-dependent electron dynamics. Some of these theories assume that spin flip collisions of the type of Stoner excitations are the dominant contribution to the enhancement of the spin polarization of the low energy cascade [13], while others consider the imbalance between unfilled majority- and minority-spin electronic states as the only crucial factor [14], producing Stoner excitations as well as other spin-polarized scattering events. Still others believe that the elastic scattering of electrons is responsible for the enhancement [15]. Compiling the data of many attenuation experiments on a number of materials reveals an empirical rule for inelastic scattering of excited electrons: namely, that the dominant electron-electron scattering rate is proportional to the number of unoccupied electronic states into which excited electrons can scatter [16]. Because of the exchange splitting in ferromagnetic materials the number of holes is different for the two spin states, and the scattering becomes dependent on the spin state of the electrons. Hence, the origin of the scattering asymmetry is the imbalance between majority- and minority-spin electrons, so that in strong ferromagnets majority-spin electrons are less scattered than minority-spin electrons. This must result in a longer lifetime for majority-spin electrons as compared to the minority-spin electrons. Consequently, minority-spin electrons are preferentially filtered out if an electron beam passes through a ferromagnet.

While evidence for such a spin filter effect is mostly based on measurements of the spin-dependent mean free path of electrons, the spin-dependent lifetimes have never been measured directly. The measurement of lifetimes instead of mean free paths may be more appropriate for investigating the spin-dependent dynamics of electron relaxation mechanisms. This is especially true when ultrathin metal films are considered. Here, the decomposition of the process of electron emission into single steps is not really possible. Then, the term "mean free path" which is defined by the step of transport to the surface, becomes questionable. We note that besides the direct time-dependent experiment, the spectroscopic linewidth is another feature containing information about lifetimes [17].

Here, we apply a new time- and spin-resolved technique in order to measure spin-dependent lifetimes in ferromagnets. It involves excitation of electrons by a first ultrashort pump laser pulse, followed by a second, suitably delayed, probe laser pulse which leads to photoemission of the electrons. By measuring the number of photoelectrons in the two spin states as a function of the delay between the pump and the probe pulse, we are able to investigate the spin-dependent lifetime of excited electrons. The investigation of fcc Co(001) films shows clear evidence of a spin-dependent lifetime: a ratio $\tau^{\uparrow}/\tau^{\downarrow}$ of up to 2 is obtained for states at $\approx$1 eV above the Fermi energy E_F.

The experimental setup used in this investigation is similar to that reported in Ref. [18], with the extension that a spin analyzer [19] is added to the energy analyzer, making possible the separate but simultaneous measurement of both spin states. A detailed report on this new setup, optimized to meet the specific requirements for

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spin, time, and energy analysis, will be presented elsewhere [20]. In brief, two equally intense, collinear, orthogonally polarized light pulses with an adjustable delay interact with the metal surface. The mutual orthogonal linear polarization of the two pulses suppresses coherent excitation to a large extent [18]. While the first pulse excites electrons from their ground state into an intermediate state below the vacuum level, the second pulse—after a delay—interacts with the still excited electrons. In this second step the electrons are excited above the vacuum level, so that they can be detected as photoelectrons. By measuring the photocurrent as a function of the delay between the two light pulses, the lifetime of the excited electrons at a fixed intermediate state energy is determined. We emphasize that this two pulse correlation experiment allows one to determine lifetimes which are considerably shorter than the duration of the laser pulse [18]. A Ti:sapphire laser operating at a repetition rate of 82 MHz and a pulse width of about 40 fs is used as a pulsed light source. The linearly polarized laser output is frequency doubled in a 0.2 mm thick beta barium borate crystal to produce UV pulses between 3 and 3.4 eV photon energy. We used laser pulses of low fluence and peak power in order to avoid space charge effects on the energy distribution of the electrons. In this way, the relaxation of individual excited electrons rather than the collective behavior of a transiently heated nonequilibrium distribution is measured. Note that the count rate is much lower than one electron per pulse.

The two photon-photoemission (2PPE) experiments are performed in an ultrahigh vacuum system equipped with a cylindrical sector energy analyzer. The orientation of the sample is 45° with respect to the laser beam, and the electrons are detected in normal emission geometry. Epitaxial fcc Co films, about 10 nm thick, are grown onto a Cu(001) single crystal at room temperature [21]. The growth rate is 0.2 nm/min. The thickness of the film is measured by a calibrated quartz microbalance. Remanent magnetization of the Co(001) films is achieved by magnetizing them by a magnetic field pulse from a coil. The easy axes of the magnetization in Co/Cu(001) are shown to be the in-plane [110] directions [22]. The geometry of the system allows the measurement of the projection of the spin polarization vector along the in-plane Co [110] direction: $P = (N^{\uparrow} - N^{\downarrow})/(N^{\uparrow} + N^{\downarrow})$ with $N^{\uparrow}$ ($N^{\downarrow}$) the number of photoemitted electrons with their magnetic moment parallel (antiparallel) to the sample magnetization. The lowest available intermediate state energy that can be probed is given by $\Phi - h\nu$, with Φ being the work function of clean Co(001) ($\approx$5 eV) and $h\nu$ the photon energy. In order to extend the energy range to lower energies, we can reduce the work function by depositing small amounts ($\leq$0.1 ML) of Cs onto the Co surface. The effect of Cs on the electron scattering is negligible: within the present time resolution we do not observe a difference in the spin-integrated lifetime measurements between a clean Co surface and a Cs/Co surface in the overlapping energy region between 1.7 and 3.3 eV.

A normal emission 2PPE spectrum of a cesiated 10 nm thick Co(001) film with one laser beam blocked (i.e., without time resolution) at a photon energy of 3 eV is shown in Fig. 1. The electron energy values correspond to the actual kinetic energy of the free photoelectron: $E_{\mathrm{kin}} = 2h\nu - \Phi - E_{\mathrm{i}}$ with E_{i} being the initial state energy below E_{F}; the shift due to the difference in the work function between the sample and the detector has been compensated and the applied bias subtracted. The peak at the largest kinetic energies is built up by electrons coming from the vicinity of E_{F}, where Co 3d emission is strongest. That this structure is actually built up by Co 3d emission can be seen easily by the strong increase in the 2PPE intensity upon the subsequent evaporation of Co on the Cu substrate. The enhancement of the 2PPE yield at lower energies, on the other hand, is not due to an intrinsic feature of the Co band structure. Instead, this increase is due to the increase in the lifetime of the intermediate states at lower energies (see Fig. 2) and also to the buildup of the secondary electron cascade. The population of a state may be refilled through inelastic collisions of excited electrons in energetically higher lying states. The open squares in Fig. 1 represent the spin polarization of the photoemitted electrons. In spite of the quite structured spin-dependent density of states of Co (see inset in Fig. 3), the spin polarization does not show very much structure. Even more remarkable, the maximum spin polarization value is about 65%, which is 1.5 times higher than values obtained in single photon threshold photoemission experiments [23]. The occurrence of this

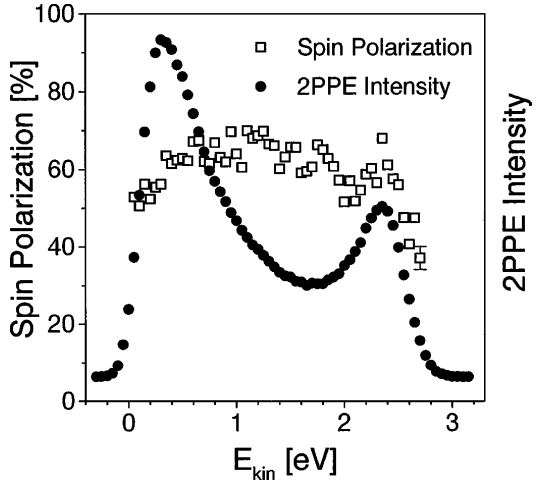

FIG. 1. Intensity (filled circles) and spin polarization (open squares) of a cesiated 10 nm thick Co(001) film as a function of the kinetic energy, obtained in a 2PPE experiment with one laser beam blocked. The photon energy of the laser light is 3 eV. The work function of this particular cesiated sample is 3.4 eV.

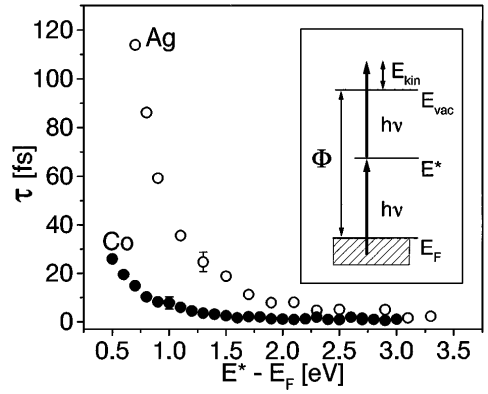

FIG. 2. The spin-integrated inelastic lifetime of cesiated Co(001) ($h\nu = 3$ eV, $\Phi = 3.5$ eV) and Ag(111) ($h\nu = 3.3$ eV, $\Phi = 4.1$ eV) as a function of the intermediate state energy above E_F. The inset shows a scheme of the energy levels involved in the 2PPE process.

enhanced spin polarization is consistent with the existence of a spin filter effect in Co, as it preferentially depletes the population of the excited minority-spin electrons during the first and the second excitation process. However, the large spin polarization of the 2PPE yield is not a conclusive proof of the spin filter effect, since different excitation processes and, hence, transition matrix elements have to be considered in 2PPE compared to regular photoemission induced by absorption of a single photon.

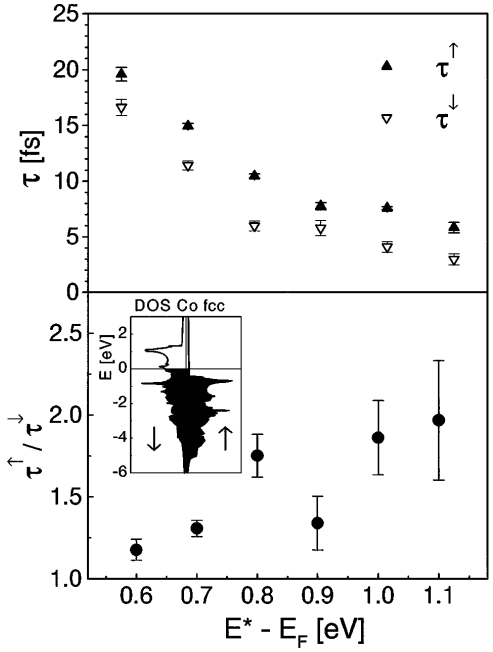

FIG. 3. The spin-resolved inelastic lifetime (top) and the ratio $\tau^{\uparrow}/\tau^{\downarrow}$ (bottom) of a cesiated 10 nm thick Co(001) film as a function of the intermediate state energy above E_F. In the top panel filled symbols correspond to the majority-spin direction and open symbols to the minority-spin direction. The photon energy is 3 eV. The inset in the bottom panel shows the spin-resolved density of states of fcc Co [26].

The marked drop in P at the highest kinetic energies (corresponding to initial state energies around E_F) is consistent with single photon threshold photoemission experiments on thick Co(001) films, where P decreases from 40% to 20% on approaching the photothreshold [23]. This decrease in P has been attributed to the fact that Co is a strong ferromagnet and, hence, has no majority-spin d density of states at E_F. That P is positive even at the photothreshold has been one of the main arguments for the spin filter process [23]. At the lowest kinetic energies in the 2PPE spectrum, however, the drop in P might be generated by the growing importance of secondary electrons.

Figure 2 shows the spin-integrated lifetime τ of both cesiated Co and the noble metal Ag for comparison as a function of the intermediate state energy $E^{\star}$. The data are extracted from the experimentally obtained cross-correlation traces using a rate equation model for the population of the intermediate state [20]. This model is equivalent to the Bloch equations for a three level system in the limit of rapid dephasing [24]. In this case, the evolution of the transient population $N^{\star}(t)$ of the intermediate level is given by $dN^{\star}(t)/dt = A(t) - N^{\star}(t)/\tau$, where $A(t)$ is the excitation induced by the first (pump) laser pulse. It should be noted, however, that the depletion of a photoexcited population at $E^{\star}$ is governed not only by an energy-dependent lifetime. At lower energies, the cross-correlation signal is affected both by secondary electrons (cascade and Auger electrons) and transport processes. Secondary electrons repopulate the probed state, whereas the diffusion of the electrons out of the probed region (transport) depopulate the state. These two opposed contributions are not involved in the rate equation model and therefore neglected in this first interpretation. The lifetime curves for both Co and Ag show a clear increase in lifetime on approaching E_F. The cause for this increase in lifetime is the decreasing phase space that is available for scattering processes. Within the framework of a conventional Fermi liquid theory, a $1/(E^{\star} - E_F)^2$ behavior is expected [25], which is in fair agreement with the observations displayed in Fig. 2. On the other hand, the lifetimes in Co are much shorter than in Ag. In the noble metal Ag, the d band is completely filled, while the d band of the transition metal Co is only partially filled, and relaxation is dominated by the d electrons. The higher density of occupied and unoccupied states near E_F in Co is thus seen to lead to a faster relaxation and hence to shorter inelastic lifetimes.

In the top panel of Fig. 3, the spin-dependent lifetime of Co as a function of the intermediate state energy between 0.6 and 1.1 eV above E_F is shown. Each pair of data points is the result of $\approx$10 h of measuring time. Clear lifetime differences between the two spin states are found, resulting in a longer lifetime of majority-spin electrons. The error bars in the plot represent the statistical scatter as described in Ref. [20]. Besides the pure statistical

error, one important additional source of error is the determination of the zero point of the lifetime scale. Thus the absolute values of the lifetimes are correct only within a few femtoseconds [18]. However, this does not influence the lifetime difference found for the two spin channels, because both were measured simultaneously. The qualitative behavior of the spin-dependent lifetime, namely, the longer lifetime of majority-spin electrons, can readily be explained by the excess of unfilled minority-spin states compared to unfilled majority-spin states, as discussed above. Because of the larger number of minority-spin holes, the minority-spin electrons are scattered out of a given energy at a faster rate than majority-spin electrons. Explaining the details of the curve is obviously more complicated and requires as the next step that the actual electronic band structure of Co(001) be taken into consideration. In the bottom panel of Fig. 3 the lifetime ratio $\tau^{\uparrow}/\tau^{\downarrow}$ is plotted, exhibiting a value of roughly 2 at 1 eV above E_F. A decreasing ratio is found towards lower energies. We note that the spin-resolved measurements are stopped at 1.1 eV, because the errors bars are getting too large due to the short lifetimes, which are at the limit of our present time resolution. A look at the Co density of states [26] (see inset) might give a clue about the behavior of the lifetime ratio. For intermediate state energies above $\approx$1.3 eV, all empty d states, in particular those which build up the strong density of states peak at around 1 eV, are available for minority-spin electrons to scatter into. For intermediate state energies clearly below 1 eV, however, the strong density of states peak is no longer available for the scattering of minority-spin electrons. Since the phase space for scattering of majority-spin electrons, on the other hand, does not change as much as in the case of minority-spin electrons—due to the constant majority-spin density of states in the energy range of interest—the lifetime ratio should change in favor of the minority-spin electron lifetime at lower energies.

In conclusion, by combining time- and spin-resolved photoemission techniques, we have demonstrated that the spin-dependent dynamics of optically excited electrons can be studied in real time. The spin-dependent lifetime of electrons in Co(001) is determined directly from the experiment and is found to be larger for majority-spin electrons than for minority-spin electrons. It is hoped that these experiments will contribute to a better understanding of spin-dependent scattering, which is of eminent importance for understanding transport properties in magnetic materials as well as the process of photoemission at low photon energies. There are also a number of applications of the spin filter effect in ferromagnets such as, for instance, the possibility to build very efficient detectors for electron spin polarization opening up the possibility to apply spin-polarized electrons more widely. At any rate, this experiment provides the first information on the spin-dependent lifetimes in the energy range between the Fermi energy and the vacuum level, thereby closing the gap between information from electric transport and mean free path experiments.

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