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Ultrafast reduction of the total magnetization in iron

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Surprisingly, if a ferromagnet is exposed to an ultrafast laser pulse, its apparent magnetization is reduced within less than a picosecond. Up to now, the total magnetization, i.e., the average spin polarization of the whole valence band, was not detectable on a sub-picosecond time scale. Here, we present experimental data, confirming the ultrafast reduction of the total magnetization. Soft x-ray pulses from the free electron laser in Hamburg (FLASH) extract polarized cascade photoelectrons from an iron layer excited by a femtosecond laser pulse. The spin polarization of the emitted electrons is detected by a Mott spin polarimeter. © *2014 AIP Publishing LLC*.
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The most commonly used method to study ultrafast magnetism is the magneto-optical Kerr effect with visible or near infrared light.[1–3] It is based on spin-orbit coupling, as light does not directly interact with the spin of the electron, and probes, furthermore, only electrons close to the Fermi energy. This method detects the magnetization only indirectly and it is controversial to which extent it is applicable for laser excited hot electrons.[4–7] Time-resolved magneto-optics can be extended to the vacuum ultraviolet by employing high harmonic generation[8,9] and to the x-ray range using x-ray magnetic circular dichroism at slicing sources.[10] But all these methods still depend on spin-orbit coupling. Recent time resolved experiments have shown that spin-orbit coupling can be altered by an infrared laser pulse.[11] A spin-orbit coupling independent way of probing ultrafast magnetodynamics can be achieved by analyzing the generation of terahertz radiation.[12,13] However, this technique only gives the time derivative of the magnetization filtered with the bandwidth of the detector.

Another, more direct way of detecting the magnetization is photoemission with spin analysis. Ultraviolet or x-ray photons cause electrons to be emitted from the sample into vacuum. Here, the spin polarization is detected by a spin polarimeter. Photoemission is more direct than all-optical detection methods as it does not rely on spin-orbit coupling inside the laser excited sample. So far, spin resolved ultrafast photoemission experiments have been carried out with laser based light sources reaching a photon energy of up to 6 eV.[14,15] If we subtract the work function of $E_{vac} \approx 5$ eV, these experiments probe only the top 1 eV of the band structure.

Our approach is to use soft x-ray pulses of 182 eV from a free electron laser (FEL) to detect the spin polarization of the whole valence band of a 3d ferromagnet. This way we can detect the total magnetization on the femtosecond time scale.

The principle of the experiment is shown in Fig. 1. An iron film on tungsten (110) serves as the magnetic sample.[16] The resulting clean, single-crystalline iron film has a magnetic in-plane easy axis along the [1$\bar{1}$0] direction and yields, after brought to magnetic saturation, a magnetic single domain state. The change of the magnetization is measured by a pump-probe technique where the sample is demagnetized by an 800 nm laser pulse and probed by the 182 eV FEL pulse. Photoelectrons are created by excitation with the FEL and their spin is measured by a Mott spin detector.

Here, we study the spin polarization of the cascade electrons. They originate from everywhere in the valence band. Hence, their spin polarization P is an average measure of the actual spin polarization of the whole valence band. An increase of the detected spin polarization is observed, due to the spin-filtering effect. On their way to the surface, minority electrons scatter more efficiently into unoccupied states than majority electrons. This increases the share of majority

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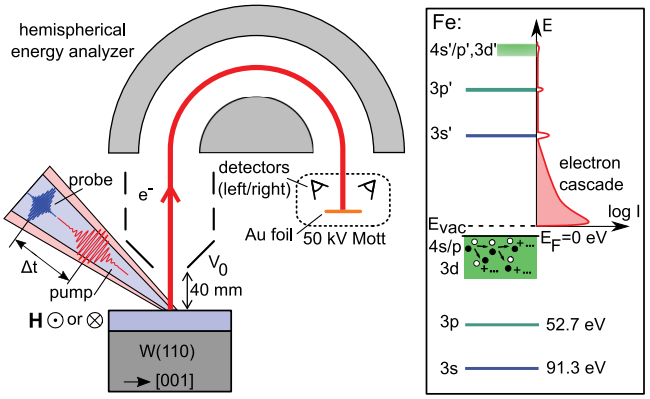

FIG. 1. Experimental overview: The iron film is excited by an 800 nm pump laser. The subsequent FEL probe pulse generates an electron cascade which passes through a hemispherical energy analyzer. The spin polarization is measured with a Mott detector. An electric field of 2.5 kV/m is applied between the sample and the first lens element to accelerate the electrons towards the electron optics. The inset shows the energy levels involved and the generation of the electron cascade with the electron yield denoted as I.

electrons leaving the sample compared to the minority electrons.[17] Nevertheless, excitation of the electron gas by the pump laser will redistribute electrons in such a way that occupied and empty states change place within the pump laser photon energy of 1.5 eV around the Fermi energy.[18] Therefore, the *total number* of empty states is not affected by the pump beam. The electrons, which can leave the sample, need an energy exceeding the work function of iron (110) of 5.1 eV.[19] The spin filter effect is caused by scattering into the *total number* of unoccupied states. The spin filtering is, therefore, not expected to get altered by pumping the sample with near infrared radiation of 1.5 eV. Thus, the spin polarization of the cascade electrons represents the "total" magnetization of the sample, even in the pumped state.

The cascade not only provides a gain mechanism by increasing the polarization of the low energetic electrons but also the cascade formation enhances the yield of the emitted electrons.

The x-ray probe and infrared pump experiment has been performed at the PG2 beamline[20,21] of the free electron laser in Hamburg (FLASH).[22] The sample is excited by <120 fs (Ref. 23) full width half maximum (FWHM) pump pulses from a Ti:sapphire laser, and subsequently measured by FEL probe pulses of ≈50 fs FWHM average length at a photon energy of 182 eV. The pulse length is derived from the shape of the photon spectra and the dispersion properties of the PG2 beamline. The FEL pulse energy on the sample is fluctuating in the range of 6–300 nJ per pulse. Both lasers are p-polarized and impinge collinearly, under 45° onto the sample. The spot-size of the pump beam ($310 \times 220\,\mu m^2$) is larger than the FEL spot-size ($130 \times 150\,\mu m^2$) in order to measure a homogeneously pumped sample. The resulting incident pump energy density is 12 mJ/cm^2. The time delay of the optical laser is varied in respect to the FEL with an optical delay line. The relative arrival time of pump and probe pulses is monitored with a streak camera[23] and an electro optical beam arrival time monitor for the electrons in the FEL.[24] This way the measurement can be corrected for drifts and the inherent time jitter of the FEL. The FEL is operated in burst mode where the machine delivers bursts of pulses at a rate of 10 Hz. Each burst consists of 300 pulses separated by 1 μs. On every other FEL pulse, the film is excited by the pump laser. The un-pumped events provide a measurement of the full magnetization. For normalization, the sample's magnetic orientation is reversed between bursts by magnetic field pulses.

The cascade electrons are collected by an electrostatic lens system which also provides energy analysis. A voltage of −98 V is applied to the sample while the first lens element (V_0) is on ground potential. This makes the electrons trajectories less sensitive to magnetic stray fields and achieves a higher transmission of electrons into the Mott detector. The energy analyzer is set to a center kinetic energy of 100 eV with an energy passband of 4.8 eV, corresponding to the peak of the electron cascade.

A Mott spin polarimeter is used to detect the spin polarization along the magnetization direction of the sample. As the FEL causes more than one detected electron per pulse, the Mott detector uses calorimetric electron detectors, providing an electrical pulse height $I^{\uparrow,\downarrow}_{\text{left,right}}$ proportional to the number of electrons on the left and right detector channels and for the magnetization directions $\uparrow, \downarrow$. The pulse heights from FEL pulses within the same pump-probe delay interval of 25 fs are averaged resulting in $\hat{I}^{\uparrow,\downarrow}_{\text{left,right}}$. From the averaged pulse heights, the electron polarization P along the magnetization direction $M_{\uparrow,\downarrow}$ is calculated as in Ref. 25

$$P = \frac{1}{S}\frac{\sqrt{\hat{I}^{\uparrow}_{\text{left}}\hat{I}^{\downarrow}_{\text{right}}} - \sqrt{\hat{I}^{\downarrow}_{\text{left}}\hat{I}^{\uparrow}_{\text{right}}}}{\sqrt{\hat{I}^{\uparrow}_{\text{left}}\hat{I}^{\downarrow}_{\text{right}}} + \sqrt{\hat{I}^{\downarrow}_{\text{left}}\hat{I}^{\uparrow}_{\text{right}}}}, \tag{1}$$

using a Sherman factor of $S = 0.17$. The ultrafast change of the relative magnetic polarization is measured on an 8 monolayer thick iron film. Fig. 2(a) shows the polarization of the pumped (red) $P(t)$ and un-pumped sample $P_0(t)$ (black). The data have been binned to a temporal resolution of 200 fs. Fig. 2(b) shows the laser induced magnetization loss $\Delta P/P_0 = (P - P_0)/P_0$ as a function of the pump-probe delay. The time dependent data are smoothed by a Savitzky Golay filter with polynomial of degree 3 and a window length of 11 data points. Before smoothing, the data are fitted with an exponential decay of time constant τ together with an exponential recovery of time constant ρ convoluted with a Gaussian excitation $G(t)$

$$\frac{\Delta P}{P_0} = A(1 - e^{-t/\tau})e^{-t/\rho}\Theta(t) \otimes G(t), \tag{2}$$

where P_0 is the polarization of the un-pumped sample, A represents the amplitude of the demagnetization, and $\Theta(t)$ denotes the Heaviside function.

The results of the fit lead to an exponential decay constant of $\tau = 45 \pm 50$ fs, a recovery constant of $\rho = 5.2 \pm 2.7$ ps, and a quenching of $A = -22 \pm 3\%$. The minimum of the demagnetization lies at 180 ± 100 fs. The given uncertainties correspond to $\pm\sigma$. Notice that the apparent oscillations are a consequence of noise: they also partially appear in the un-pumped data P_0 (Fig. 2(a)).

Since our method measures the spin polarization averaged over the whole valence band, we conclude that the total

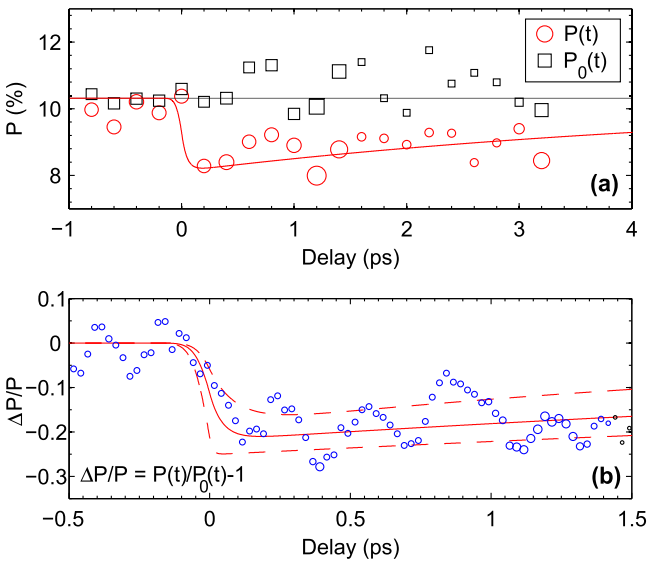

FIG. 2. (a) The polarization as a function of pump-probe delay is shown for the pumped (red) and un-pumped (black) data points. The data have been binned to 200 fs. The average polarization is shown for the un-pumped sample (black line), and the fit from (b) is shown for the pumped sample (red line). The diameter of the data points represents their weight being proportional to the number of recorded FEL pulses. Notice that more FEL pulses have been recorded for the range of −0.5 to 1.5 ps resulting in less noise in this region. (b) The relative magnetization loss $\Delta P/P_0$ is smoothed by a Savitzky Golay filter with polynomial of degree 3 and a window length of 11 for display. The apparent oscillations are caused by noise and not by the filtering process. The red curve is a fit to the measured values and the red dashed lines represent $\pm\sigma$ fitting error boundaries.

magnetization of the sample can be quenched within a 1/e-decay time of 100 fs. Hence, our experiment confirms the time scale of ultrafast demagnetization. We can therefore rule out that angular momentum of the spins is "hidden" deeper in the valence band, where it would be inaccessible to other measurement techniques.

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[1]E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, Phys. Rev. Lett. **76**, 4250 (1996).
[2]E. Carpene, E. Mancini, C. Dallera, M. Brenna, E. Puppin, and S. De Silvestri, Phys. Rev. B **78**, 174422 (2008).
[3]A. Kirilyuk, A. V. Kimel, and T. Rasing, "Ultrafast optical manipulation of magnetic order," Rev. Mod. Phys. **82**, 2731–2784 (2010).
[4]B. Koopmans, M. van Kampen, J. T. Kohlhepp, and W. J. M. de Jonge, Phys. Rev. Lett. **85**, 844 (2000).
[5]H. Regensburger, R. Vollmer, and J. Kirschner, Phys. Rev. B **61**, 14716 (2000).
[6]G. P. Zhang, W. Hubner, G. Lefkidis, Y. Bai, and T. F. George, Nat. Phys. **5**, 499 (2009).
[7]K. Carva, M. Battiato, and P. M. Oppeneer, Nat. Phys. **7**, 665 (2011).
[8]C. La-O-Vorakiat, M. Siemens, M. M. Murnane, H. C. Kapteyn, S. Mathias, M. Aeschlimann, P. Grychtol, R. Adam, C. M. Schneider, J. M. Shaw, H. Nembach, and T. J. Silva, Phys. Rev. Lett. **103**, 257402 (2009).
[9]S. Mathias, C. La-O-Vorakiat, P. Grychtol, P. Granitzka, E. Turgut, J. M. Shaw, R. Adam, H. T. Nembach, M. E. Siemens, S. Eich, C. M. Schneider, T. J. Silva, M. Aeschlimann, M. M. Murnane, and H. C. Kapteyn, Proc. Natl. Acad. Sci. U.S.A. **109**, 4792 (2012).
[10]C. Stamm, T. Kachel, N. Pontius, R. Mitzner, T. Quast, K. Holldack, S. Khan, C. Lupulescu, E. F. Aziz, M. Wietstruk, H. A. Dürr, and W. Eberhardt, Nature Mater. **6**, 740 (2007).
[11]C. Stamm, N. Pontius, T. Kachel, M. Wietstruk, and H. A. Dürr, Phys. Rev. B **81**, 104425 (2010).
[12]D. J. Hilton, R. D. Averitt, C. A. Meserole, G. L. Fisher, D. J. Funk, J. D. Thompson, and A. J. Taylor, Opt. Lett. **29**, 1805–1807 (2004).
[13]E. Beaurepaire, G. M. Turner, S. M. Harrel, M. C. Beard, J.-Y. Bigot, and C. A. Schmuttenmaer, "Coherent terahertz emission from ferromagnetic films excited by femtosecond laser pulses," Appl. Phys. Lett. **84**, 3465–3467 (2004).
[14]A. Scholl, L. Baumgarten, R. Jacquemin, and W. Eberhardt, Phys. Rev. Lett. **79**, 5146 (1997).
[15]A. Weber, F. Pressacco, S. Günther, E. Mancini, P. M. Oppeneer, and C. H. Back, Phys. Rev. B **84**, 132412 (2011).
[16]S. Miesch, A. Fognini, Y. Acremann, A. Vaterlaus, and T. U. Michlmayr, J. Appl. Phys. **109**, 013905 (2011).
[17]D. R. Penn, S. P. Apell, and S. M. Girvin, Phys. Rev. B **32**, 7753 (1985).
[18]H.-S. Rhie, H. A. Dürr, and W. Eberhardt, Phys. Rev. Lett. **90**, 247201 (2003).
[19]W. Gudat and O. Rader, *Magnetic Transition Metals, Landolt-Börnstein, New Series, Group III Condensed Matter, Vol. 23c2*, edited by A. Goldmann (Springer-Verlag, 1999).
[20]M. Martins, M. Wellhoefer, J. T. Hoeft, W. Wurth, J. Feldhaus, and R. Follath, Rev. Sci. Instrum. **77**, 115108 (2006).
[21]N. Gerasimova, S. Dziarzhytski, and J. Feldhaus, J. Mod. Opt. **58**, 1480 (2011).
[22]W. Ackermann, G. Asova, V. Ayvazyan, A. Azima, N. Baboi, J. Bahr, V. Balandin, B. Beutner, A. Brandt, A. Bolzmann *et al.*, Nature Photon. **1**, 336 (2007).
[23]H. Redlin, A. Al-Shemmary, A. Azima, N. Stojanovic, F. Tavella, I. Will, and S. Düsterer, Nucl. Instrum. Methods Phys. Res. Sect. A **635**, S88 (2011).
[24]F. Löhl, V. Arsov, M. Felber, K. Hacker, W. Jalmuzna, B. Lorbeer, F. Ludwig, K.-H. Matthiesen, H. Schlarb, B. Schmidt, P. Schmüser, S. Schulz, J. Szewinski, A. Winter, and J. Zemella, Phys. Rev. Lett. **104**, 144801 (2010).
[25]P. D. Johnson, Rep. Prog. Phys. **60**, 1217 (1997).