We have observed a quadratic x-ray magneto-optical effect in near-normal incidence reflection at the M edges of iron. The effect appears as the magnetically induced rotation of $\sim 0.1^\circ$ of the polarization plane of linearly polarized x-ray radiation upon reflection. A comparison of the measured rotation spectrum with results from x-ray magnetic linear dichroism data demonstrates that this is the first observation of the Schäfer-Hubert effect in the x-ray regime. Ab initio density-functional theory calculations reveal that hybridization effects of the 3p core states necessarily need to be considered when interpreting experimental data. The discovered magneto-x-ray effect holds promise for future ultrafast and element-selective studies of ferromagnetic as well as antiferromagnetic materials.

X-ray magneto-optical spectroscopy techniques are widespread, sensitive methods for element-selective characterization of magnetic systems. In particular the great sensitivity of resonant magnetic scattering methods has been demonstrated in many experiments exciting the L$_{3,2}$ edges ($2p \rightarrow 3d$ transitions) of 3d transition metals (TM) in magnetic nanostructures with system sizes down to the atomic scale. These experiments have recently been extended from static investigations towards magnetization dynamics. While the temporal structure of synchrotron radiation restricted the time resolution to nanoseconds in the past, studies on the ultrafast magnetization dynamics have become nowadays feasible using femtosecond x-ray pulses provided, e.g., by novel femtosecond facilities at third generation synchrotron radiation sources such as the ALS (Berkeley, USA), BESSY (Berlin, Germany), and the SLS (Villigen, Switzerland). Much higher photon flux and thus improved experimental sensitivity in magneto-optical experiments will become available with the advent of soft x-ray free electron lasers (FEL). However, the existing FEL facility FLASH (Hamburg, Germany) is designed to provide photon energies up to 200 eV, that is, the L$_{3,2}$ edges of 3d TM (in the range of 650 to 950 eV) are currently not accessible.

An alternative is provided by resonant $3p \rightarrow 3d$ transitions, i.e., the M edges (at 50 to 65 eV), where the observable magneto-optical effects may possess almost the same order of magnitude when compared to the L$_{3,2}$ edges (see, e.g., Refs. [5][6]). Moreover, the importance of the M edges for the investigation of TM compounds might reach soon beyond large scale facilities. Berlasso et al. [7] have recently demonstrated the feasibility of performing ultrafast, table-top experiments at the M edges of TM through the higher order harmonic generation (HHG) of fs laser pulses. Consequently, ultrafast, element-selective magneto-optical techniques exciting the 3p core level electrons can become accessible to most laboratories. Despite of these promising properties the M edges are rarely investigated so far and their capabilities for the above described experiments have not yet been explored. To fully profit from current FEL capacities and future HHG possibilities for element-specific static and time-resolved magnetization studies it is then necessary to further explore magneto-optical techniques in this promising energy range.

In this Letter we report the discovery of a novel quadratic x-ray magneto-optical effect at the M edges of TM occurring upon reflection of linearly polarized radiation in near-normal incidence. By comparison with additional x-ray magnetic linear dichroism (XMLD) measurements and ab initio calculations we show that the reported effect is the x-ray analogon to a similar observation made by Schäfer and Hubert in the nineties using visible light which subsequently proved to be a valuable tool for the visualization of magnetic domains.

The Schäfer-Hubert effect results from the symmetry-breaking that occurs due to the preferred magnetization axis in a magnetically ordered material. As a consequence, the indices of refraction are different for linearly polarized light propagating with electric polarization parallel to $\mathbf{M}$ ($n||$) and perpendicular to $\mathbf{M}$ ($n_|$), respectively. Light traversing the material with $\mathbf{E}$ and $\mathbf{M}$ at an angle of 45° contains equal components $E||$ and $E_|$.

In near-normal incidence reflection the magnetic modification embodied in $n||$ and $n_|$ leads to the magnetic Schäfer-Hubert rotation of the polarization plane upon reflection, which, using Fresnel theory, can be expressed as

$$\theta_{SH} \approx \text{Re} \left[ \frac{(n|| - n_|)n_0}{n||n_| - n_0^2} \right] \approx \text{Re} \left[ \frac{(\epsilon|| - \epsilon|)n_0}{(n^2 - n_0^2)n} \right],$$

where $n = (n|| + n_|)/2$, $\epsilon||$, $\epsilon_|$ are the permittivities for $\mathbf{E}||\mathbf{M}$, $\mathbf{E} \perp \mathbf{M}$, respectively, and $n_0$ is the refractive index of the cap layer. The dominating quantity for the effect is $\Delta = \epsilon|| - \epsilon_|$ which also is essential to the XMLD and the x-ray Voigt effect that are both observable in transmission [10]. Earlier investigations proved...
magnetic coils allowed magnetic saturation of \( M \). The investigated sample was deposited to prevent oxidation. The incoming radiation state of the reflected light was analyzed using a two orthogonal directions in the sample plane. The polarization plane of linearly polarized x-rays in the vicinity of the Fe \( M \) edges is depicted in Fig. 1(b). It shows a resonant behavior with \( I(\gamma) = R_0 \cdot [1 + P \cdot \cos 2(\gamma + \theta)] \), where \( R_0 \) denotes the product of the reflectivity of the sample with that of the Au analyzer and \( P \) is the product of the polarizing power of the Au layer and the degree of linear polarization of the reflected radiation. Here we can set \( P = 1 \). Fitting the above equation to the data [cf. the red line in Fig. (c)] we obtain \( \theta \) for each photon energy and magnetization. The Schäfer-Hubert rotation is finally given by 

\[
\theta_{\text{SH}} = \frac{[\theta(m_T) - \theta(m_L)]}{2}
\]

The resulting Schäfer-Hubert rotation spectrum \( \theta_{\text{SH}} \) is given in Fig. 2. It shows a resonant behavior with a twofold sign reversal close to the \( M \) edges and maximum values of about \( 0.1^\circ \). For comparison we have measured the corresponding XMLD effect in transmission geometry \(^{11}\). Figure 3(a) shows \( \text{Im}\Delta \) (open circles) being directly deduced from the transmission data.
and \( \text{Re} \Delta \) (solid circles) obtained from a Kramers-Kronig transformation. Using Eq. (1) and the experimental \( \Delta \) values together with reported data \([7]\) for the permittivity \( \epsilon \), yields the theoretically expected Schäfer-Hubert rotation. As depicted in Fig. 3(b) the calculated (upper white triangles) and measured \( \theta_{\text{SH}} \) (solid circles) spectra agree nicely. It is worth to mention that the XMLD data also allow us to deduce a maximum x-ray Voigt rotation in transmission of \( 8^\circ/\mu\text{m} \) at the \( M \) edges, which is remarkably similar to that measured at the Co \( L \) edges (\( 7.5^\circ/\mu\text{m} \)) \([11]\). This is surprising, since, in the conventional understanding it is the larger spin-orbit splitting of the core \( j_{3/2} \) and \( j_{1/2} \) levels, being nearly a factor ten larger at the \( L \) edges than at the \( M \) edges, that is believed to be responsible for the large \( L \) edge magneto-x-ray effects. As we will show below through \textit{ab initio} calculations, the microscopic mechanism leading to the XMLD effect occurring in reflection \([10]\). The corresponding asymmetry is given by \( A_R = (R_L - R_\parallel)/(R_L + R_\parallel) \) with \( R_L \) and \( R_\parallel \) being the reflectivity for the magnetization perpendicular or parallel to the polarization plane. At near normal incidence it has been shown that \( A_R = 2\theta_{\text{SH}} \) \([15]\). A respective \( \theta_{\text{SH}} \) spectrum computed from the experimentally determined \( A_R \) data is given in Fig. 3(b) (red triangles). The agreement with the measured \( \theta_{\text{SH}} \) rotation is again excellent as it reproduces the experimentally measured rotation spectrum both in shape and magnitude.

\textit{Ab initio} density functional theory calculations have been performed using a full-potential linearized augmented plane wave (FLAPW) method in the WIEN2k implementation \([16]\). We may note that a particular difficulty for the theoretical description of the \( 3p \) semi-core states is related to the relative sizes of the exchange splitting and spin-orbit splitting of the \( 3p \) levels. Whereas at the \( L \) edges the exchange splitting of the \( 2p \) states is quite small and, consequently, can be treated as a perturbation to the spin-orbit split \( j_{3/2} \) and \( j_{1/2} \) levels, this can no longer be done for the \( 3p \) states. In our relativistic calculations exchange and spin-orbit splitting were therefore included on an equal footing. Also, a considerable hybridization of the \( j_{1/2} \) and \( j_{3/2} \) states can be expected at the \( 3p \) level. To allow for this, the \( 3p \) states of iron have been treated as valence states in our calculations. The combined effect of the exchange and spin-orbit interaction as well as of hybridization on the \( 3p \) states is illustrated in Fig. 4(a), where we show the computed \( 3p \) density of states (DOS). Clearly, the \( 3p \) states are not anymore separate \( j_{1/2} \) and \( j_{3/2} \) levels, but are mixtures of all \( jj_z \) components, a situation which is markedly different from that of the \( 2p \) levels. Our relativistically calculated energies of the \( 3p \) levels are in good agreement with a previous calculation \([17]\), which, however, did not consider the hybridization of the \( jj_z \) components.

To obtain the Schäfer-Hubert rotation spectrum we first calculated the complex dielectric tensor of bcc Fe and subsequently applied the four-vector Yeh formalism.
The theoretically derived $\theta_{\text{SH}}$ spectrum shown in Fig. 4(b) agrees well with the experimental one. Respective simulations show that the smaller magnitude of the calculated data with hybridization. Apart from the significant deviations from both the experimental as well the calculated data with hybridization. As shown in Fig. 4(b) this leads to significant deviations from both the experimental as well the calculated data with hybridization. Apart from the different spectral shape the magnitude of $\theta_{\text{SH}}$ is about five times larger than the one with $j,j_z$ mixing included. This demonstrates that for proper description and interpretation of x-ray magneto-optical effects at the $M$ edges it is essential to take the hybridization of the $j,j_z$ levels into account.

In conclusion, we have detected a novel quadratic magneto-x-ray effect occurring upon reflection of linearly polarized x-rays in near-normal incidence. A comparison with x-ray magnetic linear dichroism data and ab initio calculations confirms that this effect is the x-ray analogon of the Schäfer-Hubert in the visible light regime. These calculations also show that the hybridization of the 3p core level states has to be considered for a proper description of magneto-optical effects at the $M$ edges of the 3d transition metals. With the recent advances in the development of short wavelength optics and the increasing availability of ultrashort UV and X-ray pulses the Schäfer-Hubert effect offers promising opportunities for ultrafast and element-specific microscopy of ferromagnetic and antiferromagnetic domains.

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[1] J. Stöhr and H. C. Siegmann, *Magnetism: From Fundamentals to Nanoscale Dynamics* (Springer, Berlin, 2006).
[2] P. Gambardella, S. Rusponi, M. Veronese, S. S. Dhesi, C. Grazioli, A. Dallmeyer, I. Cabria, R. Zeller, P. H. Dederichs, K. Kern, C. Carbone, and H. Brune, *Science* **300**, 1130 (2003).
[3] C. Stamm, T. Kachel, N. Pontius, R. Mitzner, T. Quast, K. Holldack, S. Khan, C. Lupaescu, E. F. Aziz, M. Weietstruk, H. A. Dürr, and W. Eberhardt, *Nature Mater.* **6**, 740 (2007).
[4] R. W. Schoenlein, S. Chattopadhyay, H. H. W. Chong, T. E. Glover, P. A. Heimann, C. V. Shank, A. A. Zholents, and M. S. Zolotorev, *Science* **287**, 2237 (2000); S. Khan, K. Holldack, T. Kachel, R. Mitzner, and T. Quast, *Phys. Rev. Lett.* **97**, 074801 (2006).
[5] M. Hecker, P. M. Oppeneer, S. Valencia, H.-Ch. Mertins, and C. M. Schneider, *J. Electron Spectrosc. Relat. Phenom.* **144**, 881 (2005).
[6] S. Valencia, A. Gaupp, W. Gudat, H.-Ch. Mertins, P. M. Oppeneer, D. Abramsohn, and C. M. Schneider, *New J. Phys.* **8**, 254 (2006).
[7] R. Berlasso, C. Dallora, F. Borgatti, C. Vozi, G. Sansone, S. Stagira, M. Nisoli, G. Ghiringhelli, P. Villoresi, L. Poletto, M. Pascolini, S. Nannarone, S. De Silvestri, and L. Braicovich, *Phys. Rev. B* **73**, 115101 (2006).
[8] R. Schäfer and A. Hubert, *Phys. Status Solidi (a)* **118**, 271 (1990). R. Schäfer, *J. Magn. Magn. Mater.* **148**, 226 (1995).
[9] A. Hubert and R. Schäfer, *Magnetic Domains* (Springer, Berlin, 1998).
[10] P. M. Oppeneer, H.-Ch. Mertins, D. Abramsohn, A. Gaupp, W. Gudat, J. Kunes, and C. M. Schneider, *Phys. Rev. B* **67**, 052401 (2003).
[11] H.-Ch. Mertins, P. M. Oppeneer, J. Kunes, A. Gaupp, D. Abramsohn, and F. Schäfers, *Phys. Rev. Lett.* **87**, 047401 (2001).
[12] R. Follath, F. Senf, and W. Gudat, *J. Synchrotron Radiat.* **5**, 769 (1998).
[13] F. Schäfers, H.-Ch. Mertins, A. Gaupp, W. Gudat, M. Mertin, I. Packe, F. Schmolla, S. Di Fonzo, G. Soullie, W. Jark, R. Walker, X. Le Cann, M. Eriksson, and R. Nyholm, *Appl. Opt.* **38**, 4074 (1994).
[14] J. Kunes and P. M. Oppeneer, *Phys. Rev. B* **67**, 024431 (2003).
[15] P. M. Oppeneer, H.-Ch. Mertins, and O. Zaharko, *J. Phys.: Condens. Matter* **15**, 7803 (2003).
[16] P. Blaha, K. Schwarz, G. K. H. Madsen, D. Kvasnicka, and J. Luitz, *2001 WENK Vienna University of Technology* (ISBN 3-9501031-1-2).
[17] J. Bansmann, L. Lu, K. H. Meives-Broer, T. Schlathölder, and J. Braun, *Phys. Rev. B* **60**, 13860 (1999).
[18] P. Yeh, J. Opt. Soc. Am. **69**, 742 (1979).
[19] M. Osugi, K. Tanaka, N. Sakaya, K. Hamamoto, T. Watanabe, and H. Kinosita, *Jpn. J. Appl. Phys.* **47**, 4872 (2008); J. Gautier, F. Delmotte, M. Roulliat, F. Bridou, M.-F. Ravet, and A. Jerome, *Appl. Opt.* **44**, 384 (2005).
[20] A.-S. Morlens, R. Lopez-Martens, O. Boyko, P. Zeitoun, P. Balcon, K. Vary?, E. Gustafsson, T. Remetter, A. Lhuillier, S. Kazamias, J. Gautier, P. Delmotte, and M.-F. Ravet, *Opt. Lett.* **31**, 1558 (2006).