Resonant magnetic scattering with soft x-ray pulses from a free-electron laser operating at 1.59 nm

C. Gutt,[1] L.-M. Stadler,[1] S. Streit-Nierobisch,[1] A. P. Mancuso,[1] A. Schropp,[1] B. Pfau,[2] C. M. Günther,[2] R. Könnecke,[2] J. Gulden,[1] B. Reime,[1] J. Feldhaus,[1] E. Weckert,[1] I. A. Vartanyants,[1] O. Hellwig,[3] F. Staier,[4] R. Barth,[4] M. Grunze,[4] A. Rosenhahn,[4] D. Stickler,[5] H. Stillrich,[5] R. Frömter,[5] H. P. Oepen,[5] M. Martins,[6] T. Nisius,[7] T. Wilhein,[7] B. Faatz,[1] N. Guerassimova,[1] K. Honkavaara,[1] V. Kocharyan,[1] R. Treusch,[1] E. Saldin,[1] S. Schreiber,[1] E. A. Schneidmiller,[1] M. V. Yurkov,[1] S. Eisebitt,[2] and G. Grübel[1]

[1]*DESY, Notkestraße 85, 22607 Hamburg, Germany*

[2]*Berliner Elektronenspeicherring-Gesellschaft für Synchrotronstrahlung m.b.H. (BESSY), Albert-Einstein-Straße 15, D-12489 Berlin, Germany*

[3]*Hitachi Global Storage Technology, 3403 Yerba Buena Road, San Jose, California 95135, USA*

[4]*Angewandte Physikalische Chemie, Universität Heidelberg, Im Neuenheimer Feld 253, D-69120 Heidelberg, Germany*

[5]*Institut für Angewandte Physik, Universität Hamburg, Jungiusstraße 11, D-20355 Hamburg, Germany*

[6]*Institut für Experimentalphysik, Universität Hamburg, Luruper Chaussee 149, D-22761 Hamburg, Germany*

[7]*Institute for X-ray-Optics, RheinAhr-Campus Remagen, FH Koblenz, Südallee 2, D-53424 Remagen, Germany*

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We report on a resonant magnetic scattering experiment using soft x-ray pulses generated from a free-electron laser (FEL). The free-electron laser was operated at a fundamental wavelength of 7.97 nm and radiation at the fifth harmonic originating from self-amplified stimulated emission at 1.59 nm with an average energy of 4 nJ per pulse was detected. We demonstrate the feasibility of resonant magnetic scattering at FEL sources by using a Co/Pd multilayer as prototype sample that was illuminated with 20-fs-long soft x-ray pulses tuned to the Co L_3 absorption edge at 778.1 eV (1.59 nm).

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The quest for smaller and faster magnetic storage devices is a formidable challenge in modern magnetism. Ideally, one would like to probe elementary magnetization dynamics such as spin-flip processes and their coupling to the electronic system on their intrinsic time scales in the femtosecond regime.[1–3] At the same time nanometer spatial resolution and element-specific information is required in order to account for the complex composition of technologically relevant magnetic media and devices. Simultaneous fulfillment of these requirements mandates ultrafast magnetic scattering experiments using flashes of resonantly[4–8] tuned soft x rays, in particular, for the technologically relevant transition metals Cr, Mn, Fe, Co, and Ni with $2p$ electron binding energies between approximately 550 and 900 eV. Such experiments can be anticipated in the near future given the current construction of x-ray free-electron lasers (FELs) in the USA, Japan, and Germany.[9–11] At present the world's most powerful FEL—FLASH in Hamburg, Germany—provides uniquely intense coherent short pulses in the extreme ultraviolet (EUV) energy range with the shortest fundamental wavelength of 6.5 nm. Evidence for lasing at higher harmonics of the fundamental mode has been reported recently[12] and the use of the fifth harmonic of the fundamental at 7.97 nm brings the FEL wavelength down to 1.59 nm which has enabled us to demonstrate the feasibility of resonant magnetic scattering experiments at FEL sources.

State-of-the-art free-electron lasers are based on the principle of self-amplified spontaneous emission (SASE) where laser radiation builds up during a single pass of an electron bunch through a long periodic magnetic undulator. The FLASH facility in Hamburg uses this concept to produce ultrashort pulses of EUV radiation. Bunches of electrons are accelerated to a maximum electron energy of 1 GeV and compressed to a high peak current in magnetic chicane bunch compressors.[13] This nonlinear longitudinal magnetic compression yields a non-Gaussian electron density distribution within each bunch, leading to a high value of local charge concentration. While the electron beam is passing through the undulator, the self-amplification process starts from the shot noise of the beam resulting in a microbunching of the electrons at the scale of the radiation wavelength. In each microbunch the electrons emit in phase, thereby producing powerful radiation pulses of 10–50 fs duration. In this way FLASH covers the wavelength range from 6.5 to 50 nm, i.e., it reaches a maximum photon energy of 191 eV. With these wavelengths the energies of the L edges of the $3d$ transition metals, between 550 and 900 eV, are out of reach.

However, the FEL radiation contains odd harmonics in addition to the fundamental mode. This refers to both incoherent and coherent radiations.[14] At 13.7 nm fundamental x-ray wavelength, the relative contributions of the third and the fifth harmonics to the total radiation power have been measured as 0.5% and 0.03%, respectively.[12] We demonstrate the generation of fifth harmonic FEL radiation at 1.59 nm and perform a resonant magnetic scattering experiment at the Co L_3 edge. To this end FLASH was fine tuned to a fundamental wavelength of 7.97 nm with an average energy per pulse of 15 μJ and a 10 fs pulse duration. SASE soft x-ray radiation at the fifth harmonic (778.1 eV photon energy) was thus in resonance with the magnetically dichroic transitions of Co $2p_{3/2}$ electrons to the first unoccupied electronic states in the magnetic material and thus provided magnetic contrast for a scattering experiment. At the resonance energy, the fifth harmonic was detected with an average en-

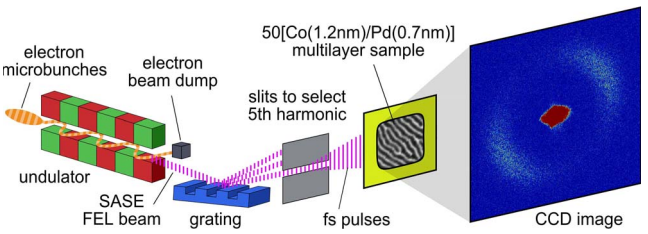

FIG. 1. (Color online) Scheme of the experimental setup.

ergy of about 4 nJ per pulse and a relative spectral bandwidth of 0.7–0.9 %. We estimate that the highly nonlinear amplification process at the fifth harmonic leads to a pulse duration of the fifth harmonic radiation as short as 1 fs and energy bandwidth of 5–7 eV.

The experiment has been performed at beamline PG2 (Ref. 15) (Fig. 1). A plane grating monochromator separates the fifth harmonic from the other wavelengths contained in the FEL beam. The use of a grating monochromator leads to a slight temporal broadening of the soft x-ray laser pulse. Based on ray tracing calculations we estimate that the pulse length was increased to 20–30 fs after the monochromator.

FLASH can provide almost fully coherent, linearly polarized, femtosecond long soft x-ray pulses at a transition metal L edge with an intensity of 3×10^7 photons per pulse. This source flux is several orders of magnitude larger than available by the technique of femtoslicing which is used to produce x-ray pulses of several 100 fs duration at third generation synchrotron sources.[16] The x-ray beam is linearly polarized in the horizontal plane. The carbon-coated optical elements (optimized for FLASH fundamental wavelengths) lead to a low beamline transmission of 2.3×10^{-4} at 1.59 nm. Due to this fact, only 7×10^3 photons per pulse were available for the experiment here and have been detected using an in-vacuum charge-coupled device (CCD) camera.

In order to quantify the ratio between higher harmonic laserlike SASE and the spontaneous radiation we measured the incident intensity with the SASE process switched on and off. The switching-off procedure consisted of a rapid change in the phases of the radio frequency system of the driving accelerator. This leads to a change in the longitudinal profile of the electron bunch, lowering the peak current, such that the SASE process is suppressed. Figure 2 displays line profiles through the direct beam with SASE switched on and off, showing that the average intensity of the ultrashort laser pulses exceeds the spontaneous radiation at the fifth harmonic by 1 order of magnitude.

Using the soft x-ray SASE photons we demonstrate resonant magnetic diffraction pattern of a Co/Pd multilayer sample in transmission geometry with the x-ray energy tuned to the Co L_3 edge at 778.1 eV. The [Co(1.2 nm)/Pd(0.7 nm)]50 multilayer was a sample similar to the one used in Ref. 17. The sample was grown via magnetron sputtering on a 20 nm Pd seed layer on a 150-nm-thick Si_3N_4 membrane. The multilayer was capped by 1.2 nm Pd to prevent corrosion.[18,19] In multilayer samples of this composition magnetic domains with alternating up and down magnetization form with a typical spatial correlation length on the order of 200–300 nm, which generate the magnetic scattering

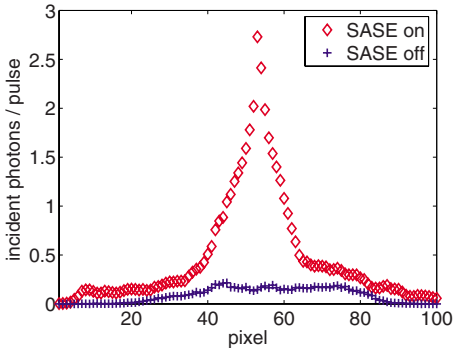

FIG. 2. (Color online) The curves show profiles of the direct beam of FLASH at the fifth harmonic with the SASE process switched on and off (see text for details).

contrast. The resonant (electrical dipole) scattering amplitude f_n at each lattice site n may be written as[6]

$$f_n = \mathbf{e}' \cdot \mathbf{e} F_n^c - i(\mathbf{e}' \times \mathbf{e}) \cdot \mathbf{M}_n F_n^{m1} + (\mathbf{e}' \cdot \mathbf{M}_n)(\mathbf{e} \cdot \mathbf{M}_n) F_n^{m2}, \tag{1}$$

where $\mathbf{e}$ and $\mathbf{e}'$ denote the polarization vectors of the incident and scattered radiations, respectively, F_n^c is the anomalous charge scattering factor, $\mathbf{M}$ is the unit vector of the magnetization, and the complex factors $F_n^{m1,2}$ contain transition matrix elements describing the resonant atomic excitation and decay process. The first term in Eq. (1) is irrelevant for our sample since no charge heterogeneities exist on the addressed length scales.[17,19,20] The second term shows a nonvanishing amplitude for scattering from linear polarization $\mathbf{e}$ to the perpendicular polarization $\mathbf{e}'$ with varying contrast according to whether $\mathbf{e}\times\mathbf{e}'$ is parallel or antiparallel to a magnetic domain orientation. The third term does not contribute to the scattering intensity because the magnetization is perpendicular to the polarization of the x-ray beam. The scattering intensity $I(q)$ is then given as a sum over the lattice sites n located at r_n: $I(q)=|\Sigma_n f_n \exp(iqr_n)|^2$ with the wave vector transfer q being related to the inverse of the correlation length probed. Within a magnetic domain all lattice sites provide the same scattering amplitude; thus the scattering intensity is reflecting the structure factor of the magnetic domains, i.e., $I(q) \propto |M(q)|^2$, with $M(q)$ being the Fourier transform of the magnetization.[21]

The scattering cross section of Eq. (1) may be affected by the ultrashort pulse duration as temporal coherence τ_c and bandwidth $\Delta\omega$ are conjugated quantities.[22] The integral of the transition matrix elements over our bandwidth is relevant for the scattered intensity. Any mismatch between the energy width of the resonance (typically about 1 eV) and the bandwidth of the beam will affect thus the scattering signal only.

Figure 3 (top) shows the measured magnetic scattering pattern with a pronounced small-angle signal. For the CCD image we employed a pixel binning of 2×2 pixels. The observed intensity maximum at a wave vector transfer of $|q_{\max}|=0.033$ nm^{-1} reflects the mean distance of 190 nm between two domains with the same orientation of the magnetic moment which implies a domain size of about 95 nm.

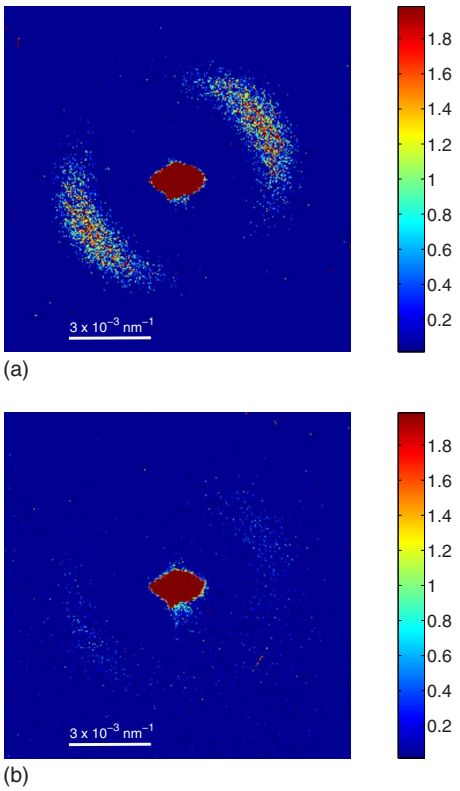

FIG. 3. (Color online) Top: CCD image of the magnetic diffraction pattern recorded with soft x-ray SASE laser radiation at a wavelength of 1.59 nm (Co L_3 edge, photon energy of 778.1 eV). The color bar indicates the number of photons recorded per pixel. Bottom: CCD image of the magnetic diffraction pattern recorded with a photon energy of 783.5 eV, slightly off resonance.

The specific lateral domain arrangement depends on the magnetic history of the sample. An in-plane component of a previously applied magnetic field gives rise to a partial alignment of the magnetic stripe pattern[18] thus causing an azimuthal oscillation of the magnetic diffraction pattern (Fig. 4). The diffraction pattern has been recorded within 1000 s and contains 6.7×10^4 scattered photons.

Detuning the photon energy away from the resonance is expected to reduce the magnetic scattering described by the

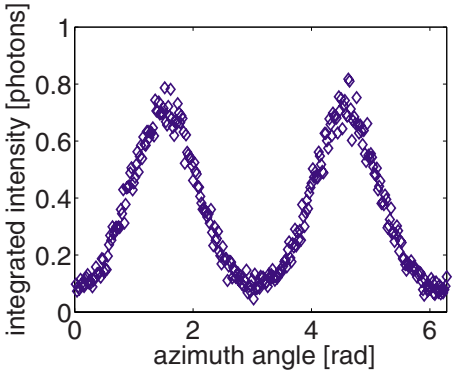

FIG. 4. (Color online) Azimuthal dependence of the scattering intensity. The asymmetry shows a preferred orientation of the domains—a so-called stripe phase.

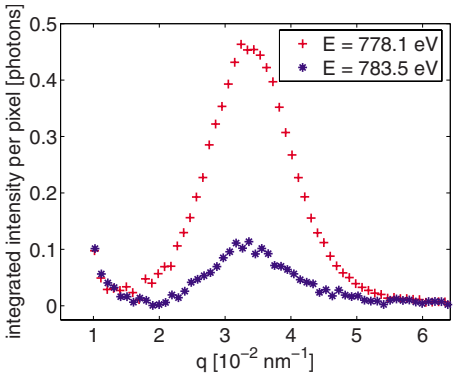

FIG. 5. (Color online) The curves show the azimuthally integrated magnetic scattering intensity. Red (+): photon energy of 778.1 eV, on resonance at the Co L_3 edge. Blue (∗): photon energy of 783.5 eV, slightly off resonance.

second term in Eq. (1). As FLASH uses a fixed gap undulator the photon energy was modified by changing the energy of the electrons entering the undulator while preserving the microbunching necessary for SASE. Figure 3 (bottom) shows the CCD image measured with a photon energy of 783.5 eV, i.e., slightly off-resonance. The scattered intensity has decreased considerably with the small-angle scattering ring barely visible. The azimuthally integrated scattering signals measured at 778.1 eV (on resonance) and at 783.5 eV (slightly off resonance) are shown in Fig. 5. Both signals clearly exhibit a peak at the same position in reciprocal space with a much reduced intensity in the off-resonance case. Taking the relatively large bandwidth of 0.7–0.9 % into account we estimate with the help of the energy-dependent magnetic scattering factors[20] a signal reduction of a factor 4–6 which is in good agreement with the observation. We would like to point out that this bandwidth is sufficient to generate contrast for resonant magnetic scattering. This implies that there is no narrow bandwidth monochromator required. As a result, the beamline transmission losses could be easily reduced by 2–3 orders of magnitude with suitably designed optical elements.

In conclusion, we have demonstrated that the higher harmonics of the EUV laser FLASH reach well into the soft x-ray regime and provide access to the absorption edges of the 3*d* transition metals. In particular, we have demonstrated the feasibility of resonant magnetic scattering using soft x-ray laser pulses via the fifth harmonic thus paving the way for measuring element-specific magnetization dynamics on femtosecond and picosecond time scales and on nanometer spatial length scales. Optimized setups for soft x-ray wavelengths will lead to intensity gains of 2–3 orders of magnitude which will make optical pump and x-ray diffraction probe measurements possible at FLASH using a single FEL pulse. The high intensities delivered on ultrafast time scales in combination with the high degree of spatial coherence of the FEL radiation will allow the measurement of element-specific spatial correlation functions in magnetic systems.

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