Absorption of Circularly Polarized X Rays in Iron

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The transmission of synchrotron radiation through magnetized iron at energies above the K-absorption edge shows relative differences for right and left circular polarization of several times 10^{-4}. The observed spin dependence of the near-edge photoabsorption is proportional to the difference of the spin densities of the unoccupied bands. In the extended absorption region up to 200 eV above the Fermi level a small spin-dependent absorption is observed and thus is expected to give information on the magnetic neighborhood of the absorbing atom.

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For a better understanding of the remaining problems of ferromagnetism, especially for a clarification of the controversy between the "localized"- and "itinerant"-electron models,[1,2] further experiments are needed to determine the spin-density distribution within the bands near the Fermi level. Besides studies of spin polarization of the occupied states with spin-resolved photoemission spectroscopy,[3] it is of equal importance to investigate the unoccupied ones, which play an essential role for spin fluctuations and excitations. The first steps to address this interesting problem were taken by spin-polarized inverse photoelectron spectroscopy.[4,5] However, because of the small range of low-energy electrons this method is sensitive only to surface layers, whose magnetic properties may differ from those of the bulk.[6] Furthermore the method used is restricted by the limited energy range in which low-energy photons can be detected efficiently by Geiger-Müller counters. The magneto-optic Kerr effect, which is sensitive to the electron spin polarization of states both below and above the Fermi level, is difficult to interpret.[7] Recently we have shown,[8,9] using 9.17-keV circularly polarized x rays emitted after internal conversion in the decay of excited oriented nuclear states, that the photoabsorption cross section of iron contains at these relatively high energies a small spin-dependent contribution of $(1.7\pm0.4)\times10^{-3}$. High-energy storage rings are intense, energy-tunable sources of circularly polarized synchrotron radiation (CPSR) in the kiloelectronvolt energy range observable at well-defined small angles above and below the plane of the electron orbits. Thus it seemed to be feasible to measure the spin dependence of photoabsorption above the K edge of $3d$ ferromagnets. Similarly one may study spin-dependent L- and M-edge absorption in magnetic materials containing $4f$ and $5f$ elements, respectively. Recently an upper limit of 2×10^{-3} was observed for the spin-dependent part of the Gd L_3-edge absorption in a $Fe_{82}Gd_{18}$ alloy.[10] In the course of this work, we have observed[11] a remarkably large spin dependence of the photoabsorption in Gd metal at the L_3 and L_2 edges of 10^{-2} with opposite sign. Here we present the first observation of spin-dependent absorption in the K-edge region of iron using CPSR from the storage ring DORIS at the Deutsches Elektron-Synchrotron DESY. Our method is discussed as a new technique to determine spin densities of unoccupied bands and to study the magnetic structure of the neighborhood of the absorbing atom. A comparison of the results on iron with spin densities calculated in the frame work of an "itinerant"-electron model is presented and discussed.

The precondition for the observation of spin-dependent K photoabsorption is that circularly polarized photons create partially polarized photoelectrons from the unpolarized $1s$ state. The $E1$ transition-matrix elements from the $1s$ state to a continuum $p_{1/2}$ and $p_{3/2}$ doublet are different because of their spin-orbit splitting. This leads to a photoelectron polarization of about 10^{-2} for $Z=26$ in direction of the photon spin.[9,12] Thus the K absorption is described in addition to a spin-independent coefficient μ_0 by a spin-dependent part μ_c, if the final states are also spin polarized as in magnetic materials. The latter is proportional to the polarization P_e transferred to the photoelectron and to differences of the spin density $\Delta\rho=\rho_{\uparrow\uparrow}-\rho_{\downarrow\uparrow}$ of the final states with their spins parallel ($\uparrow\uparrow$) or antiparallel ($\downarrow\uparrow$) to the spins of the magnetic d electrons.

With the use of these definitions the total K-shell x-ray absorption coefficient μ_K may be written as

$$\mu_K = \mu_0 + \mu_c \sim |M_{f,1s}(E_x)|^2[\rho(E) + P_e\Delta\rho(E)], \quad (1)$$

where $M_{f,1s}(E_x)$ is the transition-matrix element for the photon with energy E_x from the $1s$ state to the final state of the photoelectron with the kinetic energy E. The total density of the final states is represented by $\rho(E) = \rho_{\uparrow\uparrow} + \rho_{\uparrow\downarrow}$. Thus the spin-independent absorption coefficient measured when P_e or $\Delta\rho$ vanishes is proportional to $|M_{f,1s}(E_x)|^2\rho(E)$. By a measurement of $\mu_c/\mu_0 = P_e \times (\Delta\rho/\rho)$ and with known electron polarization P_e it is possible to determine the relative difference of the spin density $\Delta\rho/\rho$ of the unoccupied states as function of E above the Fermi level.

The measurements were performed at the x-ray spectrometer RÖMO of the Hamburger Synchrotron Radiation Laboratory with the use of partially circularly polarized synchrotron radiation viewed at angles of 0.13 $\pm$0.03 mrad above and below the plane of the electron orbits. The degree of polarization before monochromatization was calculated to be $|P_c| = 0.9 \pm 0.1$ for an electron energy of 5.3 GeV nearly independent of the photon energy between 7 and 9 keV. The x rays were monochromatized by use of a Si(311) double-crystal monochromator with an energy resolution of $\Delta E = 0.8$ eV. The degree of circular polarization after Bragg reflection is decreased by a factor of 0.5 at 7 keV. The iron absorber foil with a thickness of 2 mg/cm^2 was mounted inside a solenoid, which provided a magnetic field of 800 G parallel to the beam. The foil was tilted by 30° relative to the field. The direction of the foil magnetization relative to the photon polarization was reversed every second and monitored with Hall probes. The absorption of the left- and right-CPSR beams viewed above and below the plane of the electron orbits was measured simultaneously with two double ionization chambers, one in front and one behind the absorber foil. The spectra were measured at room temperature.

The spin-dependent transmission through a magnetized foil with thickness d is given by the difference of the transmitted intensities of photons with spins parallel, $I(\uparrow\uparrow)$, or antiparallel, $I(\uparrow\downarrow)$, to the spin direction of the magnetized d electrons in the Fe foil:

$$\Delta I/I = \frac{I(\uparrow\uparrow) - I(\uparrow\downarrow)}{I(\uparrow\uparrow) + I(\uparrow\downarrow)} = -\tanh(P_c'\mu_c d) \cong -P_c'\mu_c d. \quad (2)$$

The tilt of the foil by an angle θ relative to field direction causes a reduced effective polarization $P_c' = P_c\cos\theta$. Several scans of the transmission as a function of E_x were performed with an accumulation time of 2 sec for each energy setting. Up to forty scans were added to enhance statistical accuracy. In Fig. 1(a) the measured absorption I_0/I and in Fig. 1(b) the spin-dependent transmission $\Delta I/I$ are shown as functions of E above the K edge up to $E = 27$ eV with energy steps of 0.2 eV. The kinetic photoelectron energy is set equal to $E = E_x - E_0$, where E_0 represents the Fermi level. Its position at the absorption edge, i.e., the origin of the energy scale, can be determined by comparison of the theoretical and ex-

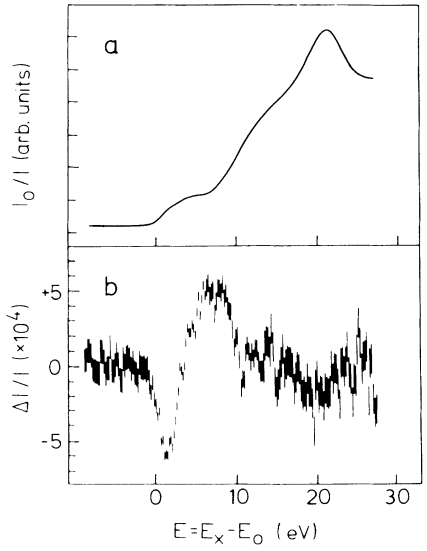

FIG. 1. (a) Absorption I_0/I of x rays as function of the energy E above the K edge of iron and (b) the difference of the transmission $\Delta I/I$ of x rays circularly polarized in and opposite to the direction of the spin of the magnetized d electrons.

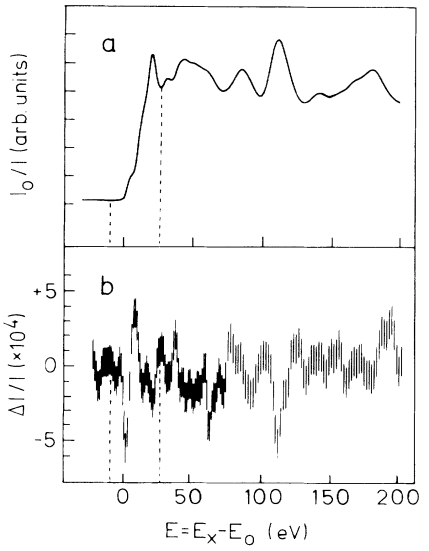

FIG. 2. (a) Extended x-ray absorption fine structure of iron in the energy region up to 200 eV above E_0 and (b) the spin-dependent transmission $\Delta I/I$. The energy region marked by dashed lines corresponds to the energy region shown in Fig. 1.

perimental onset of ρ or $\Delta\rho/\rho$ as shown later in Fig. 3.

Figure 2(a) shows again the absorption and Fig. 2(b) the spin-dependent transmission, measured in steps of 1.2 eV for $-25\ \text{eV} \leq E \leq 75$ eV and 1.8 eV up to $E = 200$ eV. This measurement was carried out to study the spin-dependent absorption mainly in the region of the extended x-ray absorption fine structure (EXAFS). From the spin-dependent transmission $\Delta I/I$ and the absorption I_0/I one can deduce a normalized spin-dependent absorption coefficient $\mu_c/\mu_0 = P_e(\Delta\rho/\rho)$ using the expression

$$\frac{\mu_c}{\mu_0} = \frac{\Delta I/I}{P_c' \ln(I/I_0)}. \tag{3}$$

The measurements were evaluated with values of $P_c' = 0.42 \pm 0.06$ at the K-edge energy of 7.1112 eV.

The normalized spin-dependent absorption coefficient is shown in Fig. 3(a) (left scale) as a function of E. Note that μ_c/μ_0 shows a maximum of $+(5.1 \pm 0.7)\times 10^{-3}$ at $E \cong 2$ eV. The positive sign of μ_c/μ_0 means stronger absorption of photons with spins parallel to the spins of the d electrons. This implies that the empty p-like band at 2 eV is populated preferentially by photoelectrons with spins in the direction of the d electrons, thus belonging to a majority-spin band. For less bound final states, μ_c/μ_0 reverses sign with a minimum value of $\mu_c/\mu_0 = -(1.8 \pm 0.2)\times 10^{-3}$ at $E = 7$ eV revealing a broader, less bound minority-spin band.

The relative differences of the spin density $\Delta\rho/\rho$, Fig. 3(a) (right scale), were determined from $\mu_c/\mu_0 = P_e(\Delta\rho/\rho)$ with the use of a calculated[12] value for $P_e = 8.4 \times 10^{-3}$, which is approximately constant up to 30 eV above the Fermi energy. In the calculation of P_e, residual interactions of the photoelectrons were ignored, which may introduce a systematic error in the absolute experimental values of $\Delta\rho/\rho$. The relative difference of the spin densities of the majority p band centered around 2 eV above E_0 reaches a maximum value of $+0.64 \pm 0.10$, whereas the minority band at 7 eV shows $\Delta\rho/\rho = -0.25 \pm 0.03$.

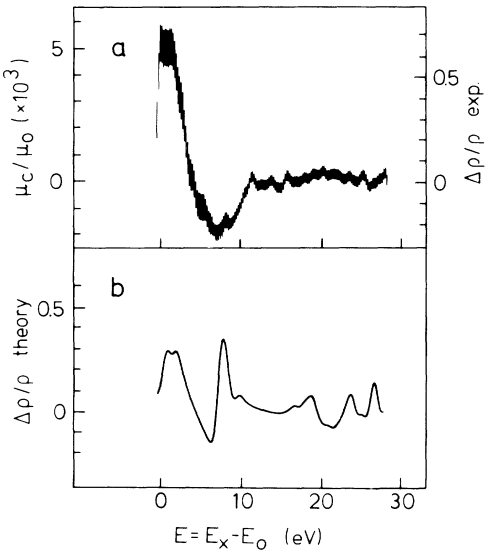

FIG. 3. (a) The normalized spin-dependent x-ray absorption coefficient μ_c/μ_0 in the near K-edge absorption region of iron (left scale) and the normalized differences of the spin density $\Delta\rho/\rho$ as function of the energy E above the Fermi level deduced from μ_c/μ_0 (right scale). (b) Calculated differences of the spin densities $\Delta\rho/\rho$ with use of the Korringa-Kohn-Rostoker band-structure method.

Calculated differences of the spin densities $\Delta\rho/\rho$ of the empty states are shown in Fig. 3(b) as a function of E. They have been obtained by the Korringa-Kohn-Rostoker band-structure method with an angular momentum cutoff at $l = 4$ with use of the self-consistent potentials of Moruzzi, Janak, and Williams[13] generated by the local spin-density approximation of the density-functional theory. The calculated spin density is folded with a spectral function corresponding to the width of the $1s$ hole (0.8 eV). A comparison with the experimental values shown in Fig. 3(a) shows agreement with the result that a majority-spin band is located at 2 eV above the absorption edge. The broad minimum around 7 eV is not reproduced as well and the majority-spin band predicted around 8 eV is not seen. The smaller absolute theoretical value of $\Delta\rho/\rho$ compared with the experimental result may be caused by theoretical values of the total p-projected density of states above the Fermi level which are too high in comparison with the values deduced from the normal Fe near-edge absorption spectra.[14]

The data in the EXAFS region shows, correlated with the absorption maximum at 115 eV [Fig. 2(a)], a statistically significant minimum in the spin-dependent transmission $\Delta I/I$ of $(5 \pm 1)\times 10^{-4}$ [Fig. 2(b)]. Other correlations may be indicated. The absorption fine structure results from interference between outgoing electron waves and those backscattered by neighboring atoms.[15] If the outgoing electron wave is polarized after absorption of CPSR, the backscattering amplitude will be sensitive to the spin polarization of the neighbors resulting from exchange interaction.[16] The spin-dependent modification of the backscattering amplitude may be expected to be in the order of some percent.[16] With a polarization of the outgoing photoelectrons of 10^{-2} taken into account, a resulting spin-dependent part of the absorption in the EXAFS region of 10^{-4} is expected in qualitative agreement with our experimental results. However, theoretical analysis of EXAFS in ferromagnets taking into account spin-dependent effects is required to extract information on magnetic structure from our data.

In summary, the present results may be considered to be important in two aspects: First, we have demonstrated that spin-dependent x-ray absorption measurements in the near K-edge region of ferromagnets are a new experimental method to measure differences of the spin

densities of empty bands near the Fermi level. The experimental procedure is relatively simple and not masked by surface effects, and covers a larger energy range compared to the equivalent inverse photoelectron spectroscopy. Since the initial electron state, the $1s$ level, has no spin density or spin-orbit coupling the spin-dependent absorption depends only on the differences of the spin densities of the final state, which is a simplification for theoretical interpretations. Second, we have observed for the first time spin-dependent EXAFS at the K edge of iron. This effect may be caused by the spin dependence of the electron backscattering from magnetized neighbors. Thus spin-dependent EXAFS may become a new method for study of the magnetic structure of the neighborhood of the absorber atom.[16]

Our experimental method is applicable to all ferromagnetic or ferrimagnetic materials, intermetallic compounds, and alloys which can be produced as foils with a thickness of some mg/cm^2. Furthermore, spin-dependent absorption measurements of impurities in ferromagnetic hosts can yield information on their modified band structures and thus provide progress in the explanation of hyperfine fields.[17]

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