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Magnetic circular dichroism in 4*f* photoemission from terbium

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Strong magnetic circular dichroism (MCD) in 4*f* photoemission from remanently magnetized films of Tb(0001) and $TbFe_x$ grown on W(110) is reported. Asymmetries of up to 62% have been observed for the $4f^8$-$^8S_{7/2}$ multiplet component, in agreement with a calculated 93% effect, taking into account the incomplete circular polarization of the photon beam and the experimental geometry. This is a very strong MCD effect and provides interesting perspectives for surface and thin-film magnetism, as well as a versatile sensor for circular polarization of soft x rays over a wide photon-energy range.

Magnetic circular dichroism (MCD) in x-ray absorption (XA) has now been established as a powerful tool for studying magnetically ordered materials in an element-specific way.[1] It is based on the dependence of x-ray absorption at a threshold on the relative orientation between photon spin and sample magnetization. MCD in XA has been applied to high-resolution imaging of magnetic domains,[2] to the study of magnetic multilayers[3] and atomically thin metal films.[4] MCD in photoemission (PE), on the other hand, is more recent and was observed in Fe-2*p* core-level PE,[5,6] and later in valence-band PE from Co.[7] The MCD effects in PE from 3*d* transition metals are relatively small (up to $\cong 10\%$), resulting from the small exchange and/or spin-orbit splitting of the involved states; the highest asymmetries were reported from Fe 2*p* and -3*p* levels in thin films.[8] Very recently, a strong MCD effect has been observed in 4*f* PE from ferromagnetic Gd metal, where an $\cong 18\%$ asymmetry was found using a soft x-ray beam with only $\cong 55\%$ circular polarization.[9] Such strong MCD effects in 4*f* PE from rare-earth (RE) materials are a consequence of a large exchange and spin-orbit interactions of the strongly localized 4*f* electrons.[10] The MCD in 4*f* PE from Gd as well as the MCD effects predicted for other RE elements[11] open new perspectives for surface and interface magnetism studies, also in view of the recently postulated sum rules for $\langle L_z \rangle$ and $\langle S_z \rangle$.[10] A straightforward application of MCD in PE is the measurement of the degree of circular polarization of soft x rays over a wide photon-energy range.

In this paper we report on the observation of a strong MCD effect in 4*f* PE from remanently magnetized films of Tb metal and of $TbFe_x$. Since the Tb $4f^8$ final-state PE multiplet is well resolved in the spectra, Tb provides an ideal case for a quantitative study of MCD in PE. For $TbFe_x$, an experimental asymmetry of 62% was observed for the $^8S_{7/2}$ multiplet component, the strongest MCD effect ever found; it provides a versatile sensor for circular polarization over a wide photon-energy range. A smaller MCD effect was observed for Tb metal, mainly due to the smaller value of T/T_c reached in the experiment. The experimental results are compared with theoretical predictions of Ref. 11. Due to the well-resolved surface splitting of the Tb-$^8S_{7/2}$ multiplet component, a technique for surface magnetism emerges.

The experiments were performed with circularly polarized soft x rays from the plane-grating SX700/III monochromator at the Berliner Elektronenspeicherring für Synchrotronstrahlung (BESSY). By selecting synchrotron radiation at a bending magnet from above and from below the storage-ring plane, respectively, left- or right-circularly polarized soft x rays in the photon-energy range 40–2000 eV are available with up to $\cong 90\%$ circular polarization.[12,13] Monocrystalline Tb(0001) films (150 ± 10 Å thick) were prepared by vapor deposition of Tb metal onto W(110) at $T \cong 110$ K. Tb is expected to grow in a layer-by-layer way for the first few layers, similar to Gd.[9,14] For a film thickness of $\cong 10$ monolayers (ML), the W 4*f* PE doublet vanished completely reflecting the growth of a flat Tb film. The pressure in the experimental chamber was $\leq 3 \times 10^{-11}$ mbar, rising briefly to $\cong 2 \times 10^{-10}$ mbar during evaporation. By subsequent annealing at 900 K for 5 min a well-ordered Tb film was obtained, as confirmed by low-energy electron diffraction (LEED). The film thickness was monitored by a quartz microbalance calibrated via the relative intensities of the 4*f* PE lines from Tb and W (for a film thickness up to a few ML). Chemical cleanliness was checked by O-2*p* PE, which is sensitive to O_2 exposures as low as 0.01 L.

Bulk Tb orders ferromagnetically below $T_c = 220$ K; the easy axis of magnetization is the *b* axis of the hcp lattice.[15] Since MCD is expected to vanish above T_c and to reach a maximum at saturation magnetization, $M(T)/M(0) = 1$, the sample temperature has to be significantly smaller than 220 K to observe large MCD effects; it was 110 K in the present experiments. Because of the large coercive fields in Tb at this temperature, remanent magnetization could be only achieved by cooling the sample in the presence of an external magnetic field of 400 A/cm from above T_c down to 110 K. This relatively small magnetic field was found to be sufficient for reversing the film magnetization at temperatures close to T_c, where the magnetocrystalline anisotropies and the coercivity are small.

In view of these experimental constraints for Tb metal, a $TbFe_x$ intermetallic phase with a much higher T_c ($\cong 700$ K) was also prepared by depositing first $\cong 10$ ML Fe on W(110), followed by $\cong 2$ ML Tb and annealing at 900 K for 10 min. Intermixing of Tb and Fe is assumed since the $^8S_{7/2}$ state is observed at 3.2 eV binding energy (BE) compared to 2.4 eV in pure Tb. Using LEED, a cubic structure was observed, indicating $TbFe_2$ formation at the surface. Coercivities at 110 K were found to be small enough to allow a

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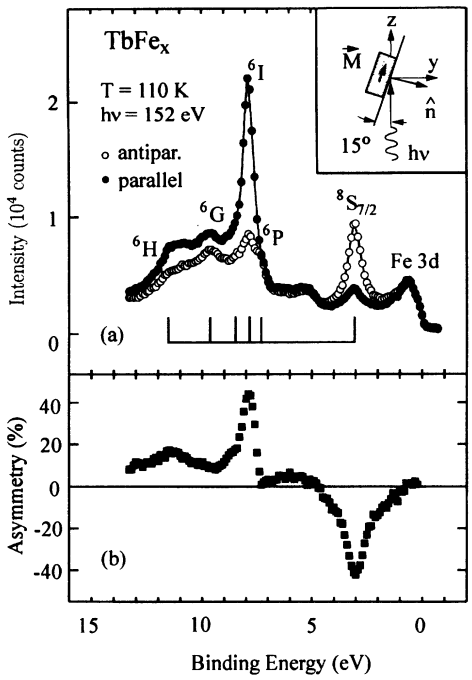

FIG. 1. (a) Tb $4f$ resonant PE spectra ($h\nu=152$ eV) of a remanently magnetized TbFe$_x$ film on W(110) ($\cong 30$ Å thick; $T=110$ K). The filled (open) dots are for approximately parallel (antiparallel) orientation of photon spin and sample magnetization. The inset gives schematically the experimental geometry. (b) Asymmetry calculated from the raw experimental spectra.

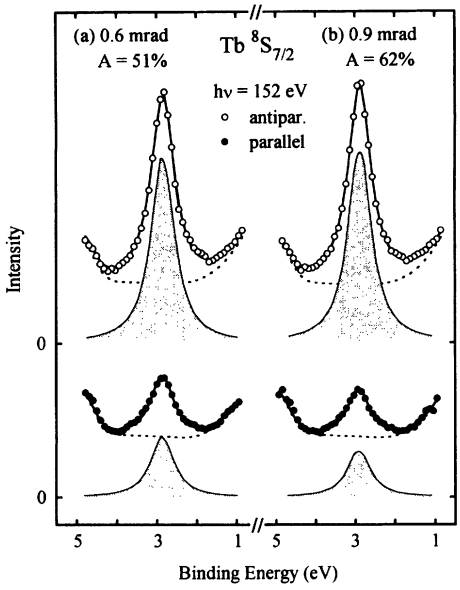

FIG. 2. MCD in PE spectra from TbFe$_x$ in the region of the Tb $^8S_{7/2}$ peak for two different monochromator settings: (a) $\psi=0.6$ mrad and (b) $\psi=0.9$ mrad off the storage ring plane. The results of least-squares fit analyses are given by the solid curves through the data points. The peak asymmetry is obtained from the peak intensities (gray-shaded areas) after background subtraction (dashed lines). All peaks have equal widths and energies.

remanent magnetization of the sample by pulses of 400 A/cm applied through a closeby solenoid. The circularly polarized light was incident at an angle of 15° with respect to the surface plane, and the photoelectrons were collected around surface normal by a hemispherical electron-energy analyzer with moderate angular resolution (±10°); the experimental geometry is given in the inset of Fig. 1.

Figure 1(a) shows $4f$ PE spectra of TbFe$_x$ taken at $h\nu=152$ eV in Tb $4d\rightarrow 4f$ resonance for approximately parallel (filled dots) and antiparallel (open dots) orientation of photon spin and sample magnetization. The two orientations were obtained for a fixed photon helicity (0.9 mrad below the ring plane) by reversing the magnetization point by point. Using this method, spectra for parallel and antiparallel magnetization were taken almost simultaneously for each data point, i.e., the two spectra are automatically normalized to each other. The large MCD effect is clearly visible in Fig. 1(a), particularly for the $^8S_{7/2}$ and ^6I states. Experimental asymmetries, $(I_{\uparrow\uparrow}-I_{\uparrow\downarrow})/(I_{\uparrow\uparrow}+I_{\uparrow\downarrow})$, of -42 and $+44$ %, respectively, are observed. At the Fermi edge, emission from Fe $3d$ states is noticeable, with almost no asymmetry, analogous to recent findings for Fe.[16]

Using the method described in Ref. 9, the intensity ratio expected for $T=0$ K in the Tb $^8S_{7/2}$ state was calculated on the basis of the dipole selection rules, pure LS coupling, and considering only $4f\rightarrow\varepsilon g$ transitions: $I_{\uparrow\uparrow}/I_{\uparrow\downarrow}=1/28$ corresponding to a theoretical peak asymmetry of $A_{\rm th}=93\%$. This value was also reproduced by the intermediate-coupling calculation of Ref. 11. The fact that a smaller asymmetry is experimentally observed follows from the nonideal experimental geometry (i.e., finite angle between in-plane magnetization and photon spin) and, in particular, the incomplete circular polarization of the light; temperature effects are negligible due to the high T_c of TbFe$_x$.

The linear dependence of the MCD effect on the degree of circular polarization suggests its use as a sensor for circular polarization. For a first demonstration, we used two different monochromator settings accepting synchrotron radiation at angles of $\psi=0.6$ mrad and $\psi=0.9$ mrad off the storage ring plane. For these settings PE spectra in the region of the $^8S_{7/2}$ peak in TbFe$_x$ are shown in Fig. 2. For 0.9 mrad [Fig. 2(b)], the peak exhibits a larger intensity difference than for 0.6 mrad [Fig. 2(a)]. In order to extract the experimental peak asymmetry $A_{\rm exp}$, the PE spectra were fitted by using a Lorentzian line on a cubic background, convoluted by a Gaussian to account for finite experimental resolution. Taking the same width and energy position for the $^8S_{7/2}$ peak in all spectra, this procedure yields accurate values for the ^8S-peak intensities and thus for the experimental peak asymmetry $A_{\rm exp}$; the values are $(51\pm5)\%$ for 0.6 mrad and $(62\pm6)\%$ for 0.9 mrad. To relate the experimental peak asymmetry with the degree of circular polarization of the light, described by the Stokes parameter S_3, a correction factor C for the experimental geometry has to be applied. C was obtained by the additional excitation probabilities for $\Delta M_J=0,\pm1$ caused by a magnetization component perpendicular to the photon spin; $C=0.96$ in the present case. With the maximum asymmetry, $A_{\rm th}$, at $T=0$ K and 100% circular polarization, one obtains $A_{\rm exp}=CS_3A_{\rm th}$. The resulting values for S_3 are $(57\pm6)\%$ for 0.6 mrad and $(69\pm7)\%$ for 0.9 mrad. Table I shows that the present data fit well into the

TABLE I. Comparison of experimental values for the degree of circular polarization S_3 of the SX700/III monochromator at BESSY.

$h\nu$ (eV)	0.6 mrad	0.9 mrad	Ref.
70	55		12
152	57±6	69±7	Present work
265	75	90	13

results of optical polarization measurements since S_3 increases with increasing photon energy at the SX700/III beamline.

For a more quantitative understanding of the MCD effect in PE from rare-earth metals, we investigated Tb metal and compared the MCD spectrum with recent theoretical results.[11] Figure 3(a) shows PE spectra of Tb(0001)/W(110) at 110 K for parallel and antiparallel orientation of photon spin and sample magnetization. As expected, the intensity differences of the ^8S and ^6I states are reduced as compared to $TbFe_x$, since T_c for bulk Tb metal amounts to only 220 K. No principal difference between the MCD of Tb metal and that of $TbFe_x$ was observed. The differences refer to the fine structure of the 4f multiplet: in Tb metal, as compared to $TbFe_x$, the spectral lines are narrower (intrinsic widths ≅50 meV), the energies of the multiplet lines are slightly shifted,

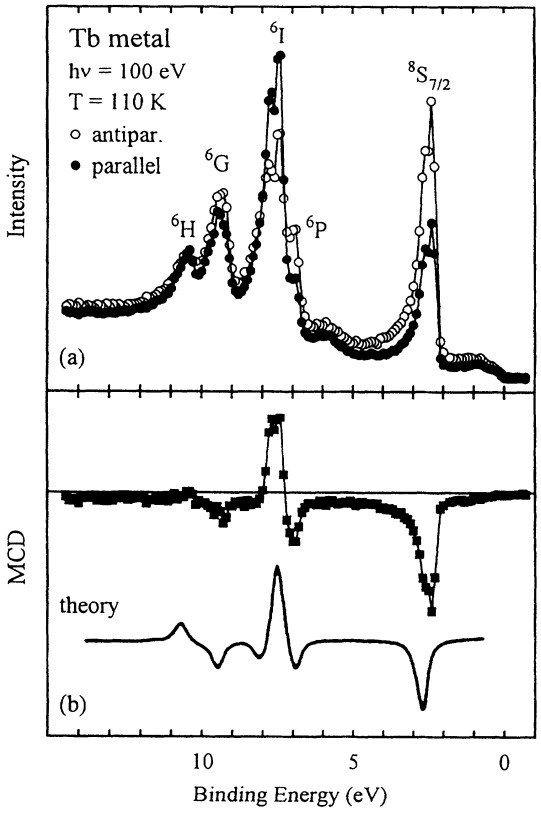

FIG. 3. (a) Tb 4f spectra ($h\nu$=100 eV) of a remanently magnetized Tb(0001)/W(110) film (150 Å thick; T=110 K). The filled (open) dots are for parallel (antiparallel) orientation of photon spin and sample magnetization. (b) Intensity difference (MCD) calculated from the experimental spectra in (a); the bottom curve (solid) represents the theoretical MCD spectrum of Ref. 11.

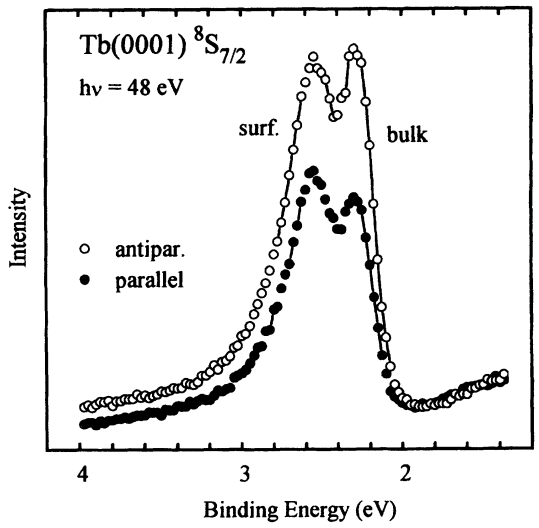

FIG. 4. MCD of the surface/bulk-split $^8S_{7/2}$ multiplet component in 4f PE ($h\nu$=48 eV) from a remanently magnetized Tb(0001)/W(110) film (150 Å thick; T=110 K). The filled (open) dots are for parallel (antiparallel) orientation of photon spin and sample magnetization.

and surface components are clearly resolved, particularly for the ^8S and ^6I lines.[17] Figure 3(b) shows a comparison of the experimental MCD spectrum (filled squares), which is the intensity difference for parallel and antiparallel orientation, with the calculated one from Ref. 11. The latter exhibits all main features observed experimentally; only the calculated relative binding energies and the overall splitting of the $4f^8$ multiplet deviate slightly from experiment.

Finally, we briefly discuss the applicability of the large MCD in 4f PE from RE materials to the study of surface magnetism. Already in the work on MCD in PE from Gd metal,[9] an antiferromagnetic coupling of the topmost surface layer relative to the bulk could be ruled out. In Fig. 4, we present in detail the $^8S_{7/2}$ region of the PE spectrum of Tb(0001) at 110 K taken with 48-eV photons for parallel and antiparallel orientation of photon spin and sample magnetization. The surface component, shifted by 0.26±0.03 eV to higher BE relative to the bulk, reflects a well-ordered Tb(0001) surface.[17] Since the ^8S component is separated from other multiplet lines, the ^8S surface component is well resolved and not mixed with other multiplet lines. It therefore provides, in combination with the strong MCD in 4f PE, an ideal tool for the study of surface magnetism. Differences in the magnetic behavior of surface and bulk are clearly noticeable from a sole inspection of the spectra in Fig. 4: the intensity difference of the surface component is strongly reduced compared to the bulk, reflecting a smaller magnetization of the topmost surface layer at the temperature of measurement (110 K). The application of the strong MCD in 4f PE from Tb to a temperature-dependent study of surface magnetism, including a determination of the surface T_c, is obvious and in progress in our laboratory.

In conclusion, we reported on a study of the MCD in 4f PE from Tb metal and $TbFe_x$. The observed asymmetries (up to 62%) represent the largest MCD effect observed so

far, and the MCD spectrum of Tb metal agrees well with theory.[11] For $TbFe_x$, the applicability of MCD in $4f$ PE as a sensor for the degree of circular polarization of soft x rays was demonstrated for 152 eV photons; until now, no other method was available in this photon-energy region. Note that this polarimeter can be used from $h\nu \cong 40$ eV up to the hard x-ray region, as long as the Tb $4f^8$ multiplet can be resolved. In a preliminary study of Tb(0001), the potential of MCD in $4f$ PE for studies of surface magnetism was demonstrated. Applications to magnetic imaging by PE microscopy will be straightforward.

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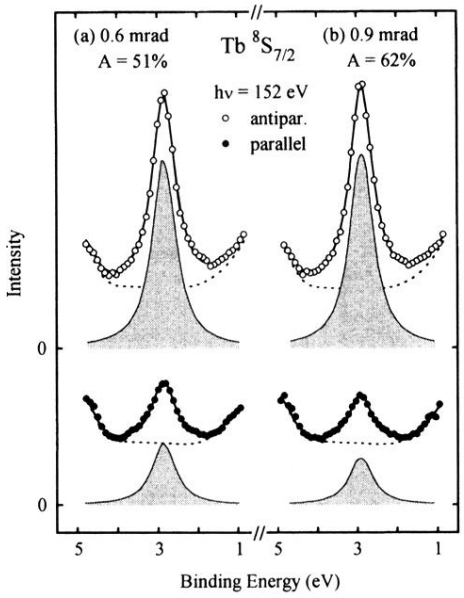

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