

5-15-2002

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Recommended Citation

Wende, H.; Li, Z.; Scherz, A.; Ceballos, G.; Baberschke, K.; Ankudinov, A.; Rehr, J. J.; Wilhelm, F.; Rogalev, A.; Schlagel, Deborah L.; and Lograsso, Thomas A., "Quadrupolar and dipolar contributions to x-ray magnetic circular dichroism at the Tb L_{3,2} edges: Experiment versus theory" (2002). *Ames Laboratory Conference Papers, Posters, and Presentations*. 27.
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Abstract

We investigate the x-ray magnetic circular dichroism (XMCD) at the L_{3,2} edges using a single crystal of Tb as a prototype system for a one-element magnet in order to ascertain the multipolar nature of the features in the dichroic spectra. The high resolution of the experimental data allows for a clear identification of the dipolar (E1: 2p→5d) and quadrupolar (E2: 2p→4f) transitions. On the basis of *ab initio* calculations we developed a simple procedure to extract the quadrupolar part by subtracting the derivative of the spin-averaged absorption spectra from the experimental XMCD data. The deconvolution has to be carried out before applying sum rules to determine 4f and 5d moments.

Keywords

terbium, magnetic circular dichroism, X-ray absorption spectra, quadrupole interactions, ab initio calculations, magnetic moments

Disciplines

Condensed Matter Physics | Metallurgy

Comments

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The following article appeared in *Journal of Applied Physics* 91 (2002): 7361 and may be found at <http://dx.doi.org/10.1063/1.1450792>.

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Quadrupolar and dipolar contributions to x-ray magnetic circular dichroism at the Tb $L_{3,2}$ edges: Experiment versus theory

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We investigate the x-ray magnetic circular dichroism (XMCD) at the $L_{3,2}$ edges using a single crystal of Tb as a prototype system for a one-element magnet in order to ascertain the multipolar nature of the features in the dichroic spectra. The high resolution of the experimental data allows for a clear identification of the dipolar ($E1: 2p \rightarrow 5d$) and quadrupolar ($E2: 2p \rightarrow 4f$) transitions. On the basis of *ab initio* calculations we developed a simple procedure to extract the quadrupolar part by subtracting the derivative of the spin-averaged absorption spectra from the experimental XMCD data. The deconvolution has to be carried out before applying sum rules to determine $4f$ and $5d$ moments. © 2002 American Institute of Physics. [DOI: 10.1063/1.1450792]

Since the first observation of x-ray magnetic circular dichroism (XMCD) in the $L_{3,2}$ edges of rare-earth (RE) compounds,¹ an extensive series of experiments have been performed, due to their great technological application such as high-performance permanent magnets² and information storage.³ The dipolar transition ($E1: 2p \rightarrow 5d$) seemed to make these spectra promising for the study of the role of $5d$ electrons in the complex magnetic phenomena of the RE materials. Unfortunately, the appearance of quadrupolar transitions ($E2: 2p \rightarrow 4f$)⁴⁻⁶ makes the interpretation of the spectra very difficult because they did not follow simple rules. It was claimed for Gd⁴ and Nd⁷ systems that the $E1$ and $E2$ channels could be separated by studying the resonant inelastic x-ray scattering (RIXS) behavior. Other studies did not show any evidence of $E2$ character for the L_2 edge XMCD in several RE ions.⁸ Even more confusing, a dichroic peak lying at the Yb L_3 absorption energy has been interpreted as an $E2$ signal in YbFe₂, while the lower energy feature was assigned to a $E1$ excitation.⁹ So far, no simple method has been found to correctly separate the quadrupolar contributions from the dichroic spectra. The identification of the quadrupolar contributions in the dichroic spectra is essential for the proper application of the sum rules because dipolar and quadrupolar transitions obey distinct sum rules.⁶

Most of the data were obtained from RE compounds, which are complicated systems including various interactions. Here we used a single element crystal with small static disorder which is very helpful to achieve a quantitative analysis of the XMCD signal. By taking advantage of the intense and tunable third generation radiation source, we

present high quality XMCD data which facilitate the clear identification of various fine structures in the XMCD. On the basis of *ab initio* calculations we show that the quadrupolar part can be extracted from XMCD data with the help of the derivative of the spin-averaged absorption spectra.

The $L_{3,2}$ XMCD spectra were recorded on a single crystal Tb specimen at the European Synchrotron Radiation Facility beamline ID12A at 10 K in fluorescence yield detection. The circular polarization rate of the undulator radiation was 84%. The Tb(0001) crystal was mounted at normal incidence to the x rays with the c axis parallel to the k -vector of the photons. The XMCD signals were obtained through the difference of the x-ray absorption spectra recorded consecutively either by reversing the helicity of the incident beam or by flipping the magnetic field of 7 T which was applied along the beam direction. To ensure that the sample was completely saturated, the magnetization $M(H)$ versus the magnetic field was measured.

The spin-dependent absorption coefficient was obtained from the difference of the x-ray absorption coefficients $\Delta\mu(E) = \mu^+(E) - \mu^-(E)$ for parallel (μ^+) and antiparallel (μ^-) orientation of the photon helicity and the magnetic field applied to the sample. The spectra were normalized by setting the total L_3 and L_2 edge jump to unity. The helicity-dependent absorption spectra at 10 K are presented in the top of Fig. 1 at both the Tb L_3 and L_2 edges and the corresponding XMCD spectra are shown at the bottom. A shoulder peak at the L_2 edge XMCD centered at 5 eV above the absorption edge was observed. This indicates a clear improvement of the quality of XMCD compared to spectra published previously where this feature could not be detected.¹⁰ The main peak in the XMCD corresponds to the dipolar part, presenting opposite signs at the L_3 and L_2 edges. The arrows indi-

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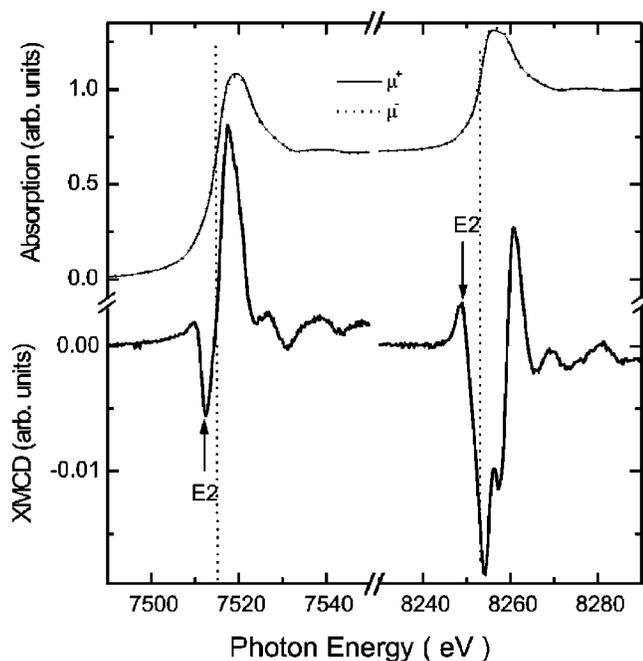


FIG. 1. Normalized Tb absorption (top) and XMCD (bottom) spectra. Possible quadrupolar transitions ($2p \rightarrow 4f$) are marked with arrows (E2).

cate the position of the quadrupolar contributions, where for each edge the sign is opposite to that of the dipolar part. One can also understand why the quadrupolar peak is negative: at L_3 edge the “+” polarization produces more spin-up electrons, while “-” polarization produces more spin-down electrons. The quadrupolar signal is proportional to the difference between spin up and spin down. Since all spin-up $4f$ states are occupied, one expects negative quadrupolar XMCD. The $E2$ contribution to the spectra appears below the absorption edge because of stronger Coulomb interaction of the $4f$ states with the $2p$ core hole.¹¹

In order to determine the pre-edge structure of the measured spectra, we performed theoretical calculations within the local spin density functional approximation by using the most recent version of the FEFF8 code.¹² Here we make use of the possibility of switching “on/off” the quadrupolar contributions in the theory in addition to the dipolar ones. Therefore, we are able to separate the $E1$ from the $E2$ contributions using the *ab initio* theory. Due to the very narrow $4f$ bands in Tb, the default atomic electron configurations was redefined to “solid state” configurations instead of atomic configurations to help convergence. Dirac–Hara self-energy was used to reproduce peak separation for both XMCD and x-ray absorption spectra (XAS). Experimental spectra along with theoretical spectra are shown in Fig 2. The calculated curves have been scaled down to match the experimental data. It is very interesting to see that the agreement between theoretical calculation and experiment is very good. Furthermore, our calculations show that the dipolar contribution displays a derivative shape¹⁰ of the spin-averaged spectra, especially in the low energy side (see Fig. 3). This fact can now be used to separate the $E1$ from the $E2$ contribution as it is given in Fig. 3.

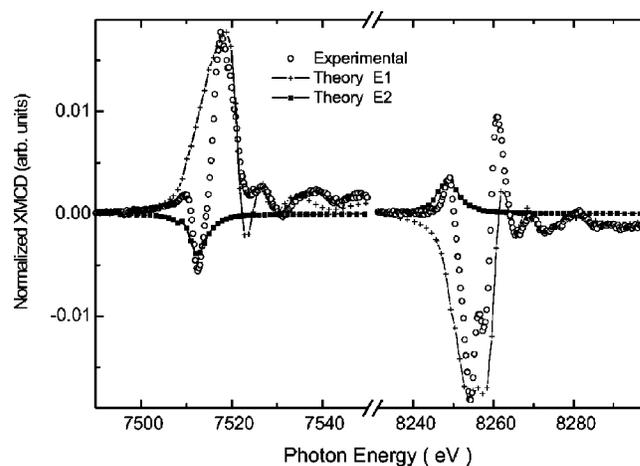


FIG. 2. Comparison of experimental (open circles) and theoretical XMCD spectra (FEFF) at Tb L_3 (left) and L_2 (right) edges: dipolar (+) and quadrupolar (squares) contributions. The theory curves have been scaled down to match the experimental data.

In Fig. 3 the XMCD spectra are compared to the derivatives of the spin-averaged XAS spectra. The latter curves are scaled by an arbitrary factor to the main peaks of the XMCD and shifted in energy (L_3 : 2.4 eV, L_2 : 0.2 eV) to match the experimental data. On the basis of the FEFF calculations mentioned above, we deconvolute the XMCD spectra of both $L_{3,2}$ edges into their dipolar (D) and quadrupolar (Q) contributions: The quadrupolar contributions were obtained by subtracting the derivative of the spin-averaged spectra from XMCD data below the main peak, because the dominant contribution in the isotropic spectrum comes from dipolar transitions into the nearly empty $5d$ shell, and in the whole RE series $E2$ s are usually located more than 5 eV below the maximum of the white line.⁵ For other light REs with multiple $E2$ peaks such as Nd and Sm,⁵ the decomposition by our method should also be possible, although the advantage of Tb is that the $E2$ contribution is a unique line; while the remained parts definitely belong to the dipolar contributions.

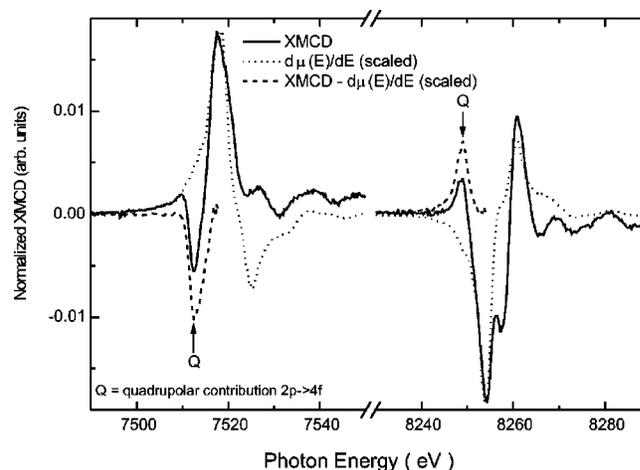


FIG. 3. The difference (dotted line) between XMCD spectra (solid line) and the derivatives $d\mu(E)/dE$ (dashed line) of the spin-averaged XAS spectra. The quadrupolar contribution is calculated by subtracting the derivative from the experimental XMCD.

Both shape and intensity are in fair agreement with the results from the FEFF calculations as shown in Fig. 2.

The procedure described above now opens the possibility of applying sum rules to determine the $5d$ and $4f$ moments from the separated $E1$ and $E2$ contribution, respectively (see, e.g., Ref. 13). But a careful analysis reveals various problems in this application. Concerning the $5d$ moments it was suggested by Wang *et al.*¹¹ that the transition matrix elements can be spin dependent. If this dependency is not taken into account the $5d$ moment determination is questionable. Furthermore, a nonvanishing magnetic background in our spectra, especially at the L_3 edge, can be detected which extends far above the Fermi level. Therefore the range for integration cannot be determined easily. The $4f$ moment determination can be done in principle using sum rules of Carra *et al.*¹⁴ for the quadrupolar contribution ($2p \rightarrow 4f$). For the exact determination the quadrupolar contribution in the XMCD spectra has to be normalized by the isotropic (spin-averaged) quadrupolar spectrum. Since those features cannot be detected easily in the isotropic absorption spectra, a precise determination of the $4f$ moment is also questionable. The difficulties mentioned above will be discussed together with theoretical calculations trying to solve them in a forthcoming article.

In summary, by taking advantage of a simple model where the dipolar contribution in XMCD displays a derivative shape, we successfully separated the quadrupolar contributions from the total XMCD signal. Compared with the RIXS method,⁷ our approach is much simpler and easier to handle. Since the spin-averaged XAS can be obtained at the

same time with XMCD data, no other measurements are needed. In contrast, RIXS does not allow a direct multipolar assignment of the observed absorption channels.⁵ Our procedure opens the way to an accurate determination of $E1$ contribution and to the study of $5d$ magnetism in rare earths.

This work is supported by BMBF (05KS1 KEB4) and DFG (Sfb 290).

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