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Published in:
Journal of Applied Physics

DOI:
10.1063/1.342292

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
1988

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):
Goedkoop, J. B., Fuggle, J. C., Thole, B. T., & Sawatzky, G. A. (1988). Magnetic x-ray dichroism of rare-earth materials. Journal of Applied Physics, 64(10), 5595-5597. https://doi.org/10.1063/1.342292

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Citation: Journal of Applied Physics **64**, 5595 (1988); doi: 10.1063/1.342292
View online: https://doi.org/10.1063/1.342292
View Table of Contents: http://aip.scitation.org/toc/jap/64/10
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Magnetic x-ray dichroism of rare-earth materials

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We discuss recent developments in the magnetic x-ray dichroism of rare-earth materials. The application of this technique to the study of magnetic materials is discussed. Also, other work on magneto-optical effects in the x-ray range is reviewed.

I. INTRODUCTION

Although magnetic dichroism (MD) is a long known technique in the visible region, until very recently[1-3] its equivalent in the x-ray region (MXD) was unknown. This situation was due to the difficulty of generating and manipulating polarized x-ray beams. Since the advent of electron storage rings dedicated to the production of synchrotron radiation, however, there has been a surge of research employing the unique advantages of x rays over visible light. With low-energy photons the optical transitions of solids are always within the bandstructure and the information obtained is influenced by both initial and final state densities. On the other hand, with x rays it is possible to choose a well-defined energy level for either the initial or the final state. Thus, such spectra are generally much easier to interpret, and it can be expected that this also holds true for the magneto-optical effects.

The technical advances in x-ray optics have made possible the extention of the magneto-optical techniques well known in the visible region (Kerr and Faraday effects, magnetic dichroism). Very strong dichroism effects (up to 20% of the absorption cross section) have been found experimentally in the $3d \rightarrow 4f$ absorption spectra of rare-earth materials.[2] Similar effects have been observed by several other groups in different materials and at other photon energies.

In this paper we discuss our work on MXD in the soft x-ray range. An attempt will be made to show the potential of MXD for investigations of magnetic materials and the use of circularly polarized x rays will be discussed. Also, we will discuss other work on magneto-optical effects at x-ray energies.

II. MAGNETIC X-RAY DICHROISM OF RARE EARTHS

It is well established that the $3d \rightarrow 4f$ spectra of rare-earth materials are predicted to a high degree of accuracy by atomic calculations.[4,5] In such calculations one obtains the line spectrum of all the transitions from the Hund's rule ground state of the initial $3d^{10}4f^n$ configuration, denoted by $|\alpha JM\rangle$, to the many possible states $|\alpha' J' M'\rangle$ of the $3d^9 4f^{n+1}$ final configuration (α denotes all other quantum numbers necessary to specify the state). As a result of the large $3d$-hole spin-orbit interaction, this final state multiplet can be divided to a first approximation in a $3d_{5/2}$ and a $3d_{3/2}$ part. As a typical example we reproduce in Fig. 1, top panel, the calculated spectrum of Tb^{3+}, obtained from the line spectrum by broadening with core hole lifetime and experimental resolution contributions.[6] As was shown in Ref. 4, modern calculations match the experimental spectrum nearly perfectly.

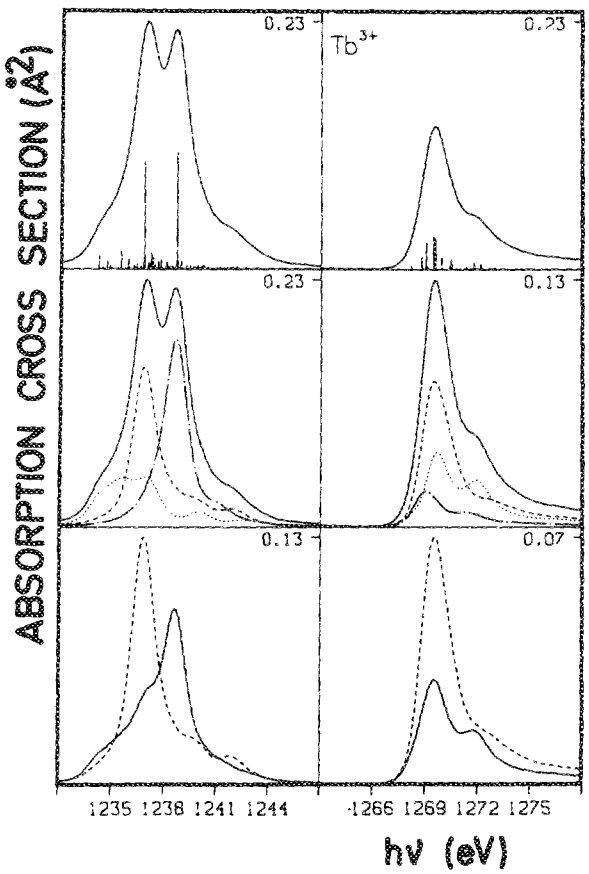

FIG. 1. Upper panel: Calculated $3d \rightarrow 4f$ x-ray absorption spectrum of Tb^{3+}. The full curves denote the absorption cross section of the $3d_{5/2}$ (left) and $3d_{3/2}$ (right) groups in units of Å^2. They were derived from the oscillator strengths (vertical bars) by convolution with Lorentzian and Gaussian curves representing the lifetime and instrumental broadening (see Ref. 4). Middle panel: The same curves with the vertical scale of the $3d_{3/2}$ part expanded, showing the contributions of each ΔJ group to the total spectrum. -·-·-: $\Delta J = -1$, ---: $\Delta J = 0$, ...: $\Delta J = +1$. Lower panel: calculated cross section at $T = 0$ K with the polarization vector parallel (- - -) and normal (—) to the magnetic field.

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It is important to note that, due to the dipole selection rules governing the absorption transition, the spectrum consists of three groups of lines with $\Delta J = -1, 0$, or 1. In the middle panel of Fig. 1 the individual contributions are presented, and it is clear that each ΔJ group has an identifiable contribution to the total spectrum.

In order to explain the MXD effect a second selection rule, limiting $\Delta M = M' - M$ to 0 or ± 1, also has to be taken into account. The former transitions can be excited only by radiation that is linearly polarized parallel to the magnetic moment of the ion, the latter two only by circularly polarized radiation incident along the axis of magnetization.

With the purpose of indicating the mechanism by which the MD effect arises, we give in Fig. 2 the level scheme for the very simple case of the Yb^{3+} ($4f^{13}$) ion. Here we have just one allowed final state and the spectrum consists of just one $\Delta J = -1$ line. In the presence of a magnetic field this line is split into 18 lines divided over three groups with different ΔM. Lower in the figure the division of the linestrength over these lines (given by the squared $3j$ symbol[5]) is indicated and, at the bottom, the polarization state pertinent to the different ΔM transitions.

At room temperature the magnetic splitting goes unnoticed as all the levels are equally populated and as it is much smaller than the experimental resolution. However, the occupancy of the levels is governed by Boltzmann statistics so that when the field splitting $g\mu_B H$ is sufficiently large relative to the thermal energy $k_B T$, the upper levels are less occupied. From the figure it can be seen that in the limit of 0 K, only the lowest level is occupied so that absorption will take place only if the light has a left-hand circularly polarized component.

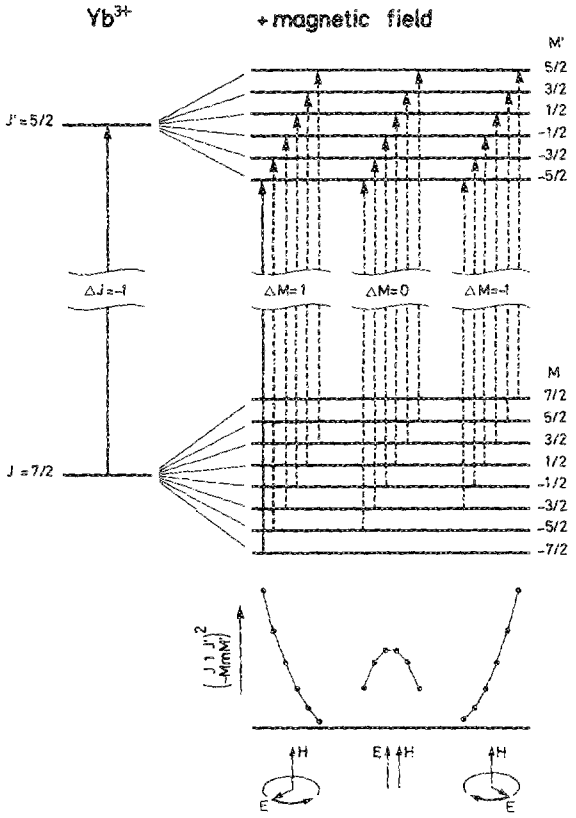

FIG. 2. Energy diagram of the $3d^{10}4f^{13} \rightarrow 3d^9 4f^{14}$ transition of Yb^{3+} without (left) and with (right) a magnetic field. The vertical arrows indicate the dipole selection rule allowed transitions $|\alpha JM\rangle \rightarrow |\alpha' J' M'\rangle$. Their relative intensities are given by the dots. The required polarization is indicated at the bottom.

In other rare-earth ions there are also $\Delta J = 0$ and $+1$ transitions whose intensities, although subject to the same mechanism, have strongly differing dependencies on field, temperature, and polarization. Their combined effect for Tb is shown in Fig. 1, bottom panel, for the two possible linear polarization states for a ferromagnetically ordered Tb material at 0 K. Circular polarization would give, in general, larger line shape variations[6,7] but even with linear polarization the effect is very large and can reach 100% of the nonmagnetic absorption coefficient.

The validity of this model has been confirmed[2] by experiments on magnetically ordered terbium iron garnet ($Tb_3Fe_5O_{12}$) using linearly polarized synchrotron radiation from the ACO storage ring of LURE, France. The size of the measured effect could be reproduced from the known local moments and magnetic structure of this material.

This experiment demonstrates that with linearly polarized x rays MXD can be used to quantitatively determine the absolute value of the magnetization of localized magnetic materials. Also, since the experiment is carried out using photoelectron yield detection under UHV conditions, it is possible to apply MXD to the study of thin-film and surface magnetization. The experimental conditions are somewhat severe, including low temperatures and high magnetic fields in UHV. On the other hand, spectra as shown in Fig. 3 require only a couple of minutes.

At present, there have been only a few experiments with circularly polarized synchrotron radiation (CPSR), and none in the soft x-ray range. Currently a number of alternative sources for CPSR are under development, most notably the crossed undulator. With CPSR the MXD effect will also be sensitive to the direction of the magnetic moment with MXD. Conversely, as is described in Ref. 7, the MXD effect can be used in a narrow bandwidth polarization filter that converts linearly polarized light in circularly polarized light.

III. X-RAY MAGNETO-OPTICAL EFFECTS

The first work in this field was the calculation of the magneto-optical Kerr effect (MOKE) of the M_{23} ($3p \rightarrow 3d$) absorption edge of Ni ($h_\nu = 64$ eV).[8] A substantial change ($\sim 10\%$) in the reflection of p polarized light in the transverse MOKE configuration was predicted. The authors argued that the experiment would be sensitive to the temperature dependence of the exchange splitting of the $3d$ bands, and thus would provide a test for the Stoner model. Also, they noted that although the magneto-optical effects involving transitions from s-core states to p-conduction band states would be much smaller, e.g., 10^{-5}, it would allow one to study the exchange splitting of the unoccupied p states.

Subsequently, a search was made for magneto-optical effects of the Gd L_3 edge ($2p \rightarrow 5d$, $h_\nu = 7243$ eV) of an amorphous GdFe alloy using the out-of-plane CPSR from electron storage rings. An upper limit of 0.02% was established for the dichroism of this edge.[9] Very recently the same experiment has been performed on Gd metal by Schütz *et*

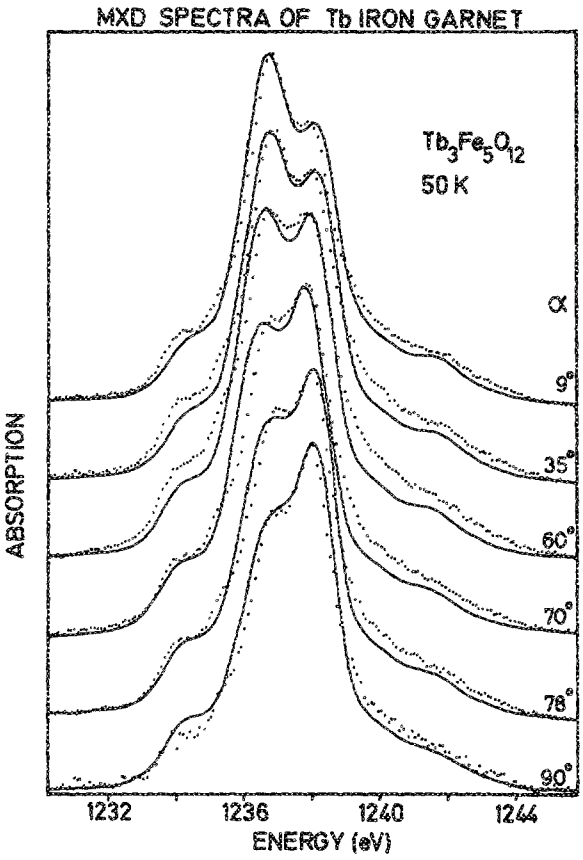

FIG. 3. Experimenal M_5 absorption spectra of $Tb_3Fe_5O_{12}$ at 50 K for various values for the angle between the x-ray polarization vector and the [111] direction of magnetization. The solid curves are theoretical fits with a 0.4-eV FWHM Gaussian and a 0.3-eV FWHM Lorentzian broadening.

al.,[10] who reported a polarization-dependent contribution to the absorption coefficient of 1%. These same authors also performed experiments[3] on the K-edge absorption of a magnetized Fe foil ($1s \rightarrow 4p$, $h_\upsilon = 7112$ eV). From the differences in transmission of left and right CPSR up to 0.1%, they deduced an energy-dependent p-state spin density that corresponds very closely with that obtained from KKR band structure calculations.

In such experiments the polarization dependence arises from spin orbit interaction induced differences in the matrix elements from the unpolarized $1s$ to the $p_{1/2}$ and $p_{3/2}$ final states. This mechanism acts effectively as an intra-atomic source for spin polarized electrons which then couple to the exchange-split final p states. The effects are rather small because the net electron polarization and the p-state spin density are both small. Much larger effects are probable at the $2p$ edges where excitation to the unnoccupied d states dominates the spectra.

IV. CONCLUSION

It is clear that the application of x rays to magneto-optical effects has a large potential. It allows direct probing of the spin-resolved band structure of itinerant magnetic materials, as well as the study of magnetic moment orientation of localized systems, thus offering direct tests for theories of magnetism.

As polarized synchrotron radiation sources are being developed, the possibilities for such research will further increase. Currently we are developing a system with which it will be possible to study MXD effects in paramagnetic materials and magnetic domains. This research will be extended to include $3d$ transition element systems. In this case the effects may be smaller, depending on the quenching of the orbital angular momentum and the effective g factor, but the signal strength is sufficiently high to measure even small effects.

ACKNOWLEDGMENTS

This work was supported in part by the Netherlands Foundation for Chemical Research (Stichting Scheikundig Onderzoek Nederland, SON) with financial support from the Netherlands Organization for Scientific Research (Nederlandse Organisatie voor Wetenschappelijk Onderzoek, NWO) and by the Committee for the European Development of Science and Technology (CODEST) program. We are grateful to the Laboratoire pour l'Utilisation du Rayonnement Electromagnetique (LURE) staff for their valuable aid. One of us (J.B.G.) thanks Shell Netherlands B.V. for a travel grant for this conference.

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