

Calculation of magnetic circular dichroism in core level photoemission spectroscopy

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1991 J. Phys.: Condens. Matter 3 1015

(<http://iopscience.iop.org/0953-8984/3/8/015>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.151

The article was downloaded on 11/05/2010 at 07:07

Please note that [terms and conditions apply](#).

LETTER TO THE EDITOR

Calculation of magnetic circular dichroism in core level photoemission spectroscopy

Gerrit van der Laan

SERC Daresbury Laboratory, Warrington WA4 4AD, UK

Received 6 December 1990

Abstract. Magnetic x-ray dichroism in core level photoemission is calculated using a localized model. It is shown that the angular dependent part of the electrostatic interaction between the core hole and the 3d electrons determines the magnetic dichroism in the 2p photoemission of 3d transition metal compounds. The circular dichroism is expected to be very strong in compounds with localized 3d electrons.

Recently, Baumgarten *et al* [1] observed magnetic x-ray dichroism in the core level photoemission of ferromagnetic iron using circularly polarized radiation. The dependence of the photoemission on the circular polarization of the x-rays was explained in terms of spin-selective dipole transitions in the presence of spin-orbit coupling. Here we will give results of a calculation of the circular dichroism using localized wavefunctions, and show that the size of the effect in the 2p photoemission spectrum is determined by the angular dependent part of the Coulomb and exchange interaction between the core hole and 3d valence electrons. This intra-atomic interaction decreases when the wavefunction of the 3d valence electrons becomes more delocalized. Therefore, dichroism will be more pronounced in materials with localized valence electrons. It is the purpose of this letter to demonstrate the large magnetic dichroism in a simple case. We have chosen for the 2p photoemission satellite peaks of divalent Cu, which are well-known from the literature [2]. At high photon energies the emission is given by the transition, $\text{Cu } 3d^9 \rightarrow 2p^5 3d^9 \epsilon d$, where ϵd is a non-interacting continuum level of d symmetry. We have neglected the configuration interaction with the closed-shell $3d^{10}$ level in the initial and final state. The configuration interaction can be taken into account using a cluster [3] or impurity model [4, 5], but this introduces several parameters that are not important for the understanding of the mechanism.

In the spherically symmetric potential of the atom the spin-orbit interaction splits the Cu $3d^9$ initial state into two levels with total angular momentum $J = \frac{5}{2}$ and $\frac{7}{2}$. These levels are $(2J + 1)$ -fold degenerate, corresponding to the number of distinct eigenvalues $M = -J, -J + 1, \dots, J$. A magnetic field lifts this degeneracy, making the energy of the M sublevels equal to $-g\mu_B H M$ with only the level $M = -J$ populated at $T = 0$ K. The energies of the $2p^5 3d^9 \epsilon d$ final state levels are determined by the angular dependent part of the 2p-3d Coulomb and exchange interactions. The interaction between the ϵd and the other states is zero. The core hole spin-orbit interaction splits the levels into a $2p_{3/2}$ and a $2p_{1/2}$ manifold with an energy separation of 19.9 eV. The transition probability

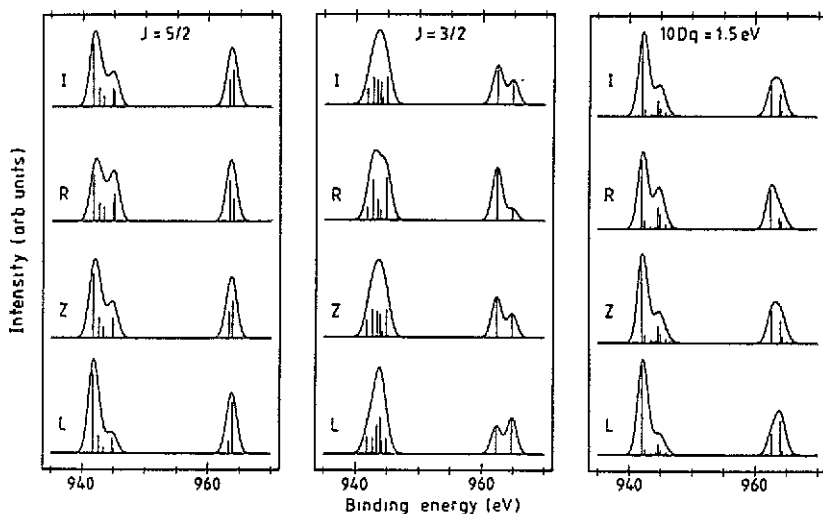


Figure 1. The vertical lines give the calculated Cu 2p photoemission spectrum with a magnetic field in the z -direction excited with (I) isotropic, (R) right-circularly, (Z) z - and (L) left-circularly polarized x-rays from a Cu $3d^9$ ground state (a) $J = \frac{5}{2}$, (b) $J = \frac{3}{2}$ and (c) $10Dq = 1.5 \text{ eV}$. The angular dependent Slater integrals used in the calculation are $F^2(2p, 3d) = 7.02$, $G^1(2p, 3d) = 5.36$, $G^3(2p, 3d) = 3.05 \text{ eV}$, and the spin-orbit parameters are $\zeta_{2p} = 13.5$ and $\zeta_{3d} = 0.1 \text{ eV}$. The curves result from a convolution of the lines with a Gaussian of $\sigma = 0.8 \text{ eV}$.

from the initial to the final state was calculated with Cowan's relativistic Hartree-Fock code [6] using atomic Slater integrals reduced to 80% to account for relaxation effects.

The spectra of the $J = \frac{5}{2}$ and $\frac{3}{2}$ initial state in a magnetic field are shown in figure 1(a) and (b), respectively, for isotropic, right-circularly, z and left-circularly polarized radiation. The isotropic spectra are identical to the non-magnetic spectra that have been published earlier [3-6]. The z -polarized spectra are not notably different from the isotropic spectra: there is no linear dichroism. However, the spectra for right- and left-circular polarization are quite different. When the spin direction of the circular polarization (or the direction of the magnetic field) is reversed, a large change is observed in the $J = \frac{5}{2}$ spectrum mainly for the $2p_{3/2}$ peak and in the $J = \frac{3}{2}$ spectrum mainly for the $2p_{1/2}$ peak. Even in the presence of a crystal field that mixes the two J levels, there is an effect. This is shown in figure 1(c) for an octahedral crystal field of $10Dq = 1.5 \text{ eV}$.

The circular dichroism in the Cu 2p photoemission is governed by the optical selection rules. The energy distribution of the levels with different total angular momentum J' in the final state configuration $2p^5 3d^9 ed$ depends on the electrostatic interaction between the 2p core hole and 3d valence electrons. This is seen in figure 2 which shows the separate ΔJ contributions in the $J = \frac{5}{2}$ spectrum. The dipole selectivity allows transitions $\Delta J = +1, 0$ and -1 to the final state levels $J' = \frac{7}{2}, \frac{5}{2}$ and $\frac{3}{2}$, respectively. Excitation with left-circularly polarized radiation ($\Delta M = -1$) from the $M = -\frac{5}{2}$ ground state is only allowed to the $J' = \frac{7}{2}$ levels; the only levels which contain an $M' = -\frac{7}{2}$ sublevel. Excitation with right-circularly polarized radiation ($\Delta M = +1$) is allowed to all final state levels; they all contain an $M' = -\frac{5}{2}$ sublevel. The transition probability is obtained with the Wigner-Eckart theorem, which can also be used to describe the temperature dependence [7]. The dichroic effect is strongest at $T = 0 \text{ K}$ and gradually reduces when higher

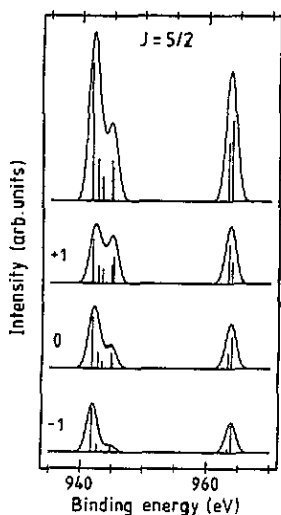


Figure 2. The Cu 2p photoemission spectrum for the Cu $3d^9$ state of total angular momentum $J = \frac{5}{2}$, together with the three separate contributions $\Delta J = J' - J = +1, 0, -1$ to this spectrum.

magnetic sublevels are populated. If there is a crystal field interaction which mixes the initial state J levels, magnetic dichroism will be present in the case that the core hole interaction is larger than the crystal field interaction. Without core hole interaction, the multiplet structure reduces to a single line and the polarization dependence disappears.

In conclusion, magnetic dichroism in photoemission using circularly polarized x-rays can be explored to measure the magnetic properties of bulk materials as well as magnetic overlayers and thin films. It is complementary to magnetic techniques such as neutron diffraction, paramagnetic resonance and Mössbauer spectroscopy, because it provides information about the *local* magnetic structure. The difference with magneto-optical effects in optical spectroscopy is the role of the core hole. With the excited electron in a free-electron state, the photoemission can only be polarization dependent if there are electrostatic interactions between the core hole and the valence electrons. The magnetic dichroism can be analysed using an atomic calculation including crystal field, hybridization (mixing) and configuration interaction. Although, we have only given a simple example, the analysis can be generally applied to transition metal and rare earth compounds where the localized character of the valence electrons gives rise to a strong variety of magnetic structures.

Acknowledgements. I thank B T Thole for making the code available, and I W Kirkman for assistance with the computer calculations.

References

- [1] Baumgarten L, Schneider C M, Petersen H, Schäfers F and Kirschner J 1990 *Phys. Rev. Lett.* **65** 492
- [2] Davis L C 1986 *J. Appl. Phys.* **59** 25
- [3] van der Laan G, Westra C, Haas C and Sawatzky G A 1981 *Phys. Rev. B* **23** 4369
- [4] Okada K and Kotani A 1989 *J. Phys. Soc. Japan* **58** 2578
- [5] Okada K and Kotani A 1990 *J. Elect. Spectrosc.* **52** 313
- [6] Cowan R D 1981 *The Theory of Atomic Structure and Spectra* (Berkeley, CA: University of California)
- [7] Thole B T, van der Laan G and Sawatzky G A 1985 *Phys. Rev. Lett.* **55** 2086