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XMCD at the Co K-edge in RCo₂ intermetallics: influence of the rare earths

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X-ray magnetic circular dichroism (XMCD) studies have been performed at the Co K edge in RCo₂ intermetallics (R = Nd, Tb and Ho). A drastic evolution in the structure of XMCD spectra is observed between Co metal and alloys. The RCo₂ dichroic spectra exhibit a three-peaks structure: one negative in the middle of the positive ones. Sizeable changes in the intensities are observed when switching the rare earth. A huge signal, ≈ 0.6%, is measured on HoCo₂ single crystal. Results show up the rare earth influence on the XMCD structure at the transition metal K edge in these alloys.

Keywords: XMCD; Hard X-rays; XANES.

1. Introduction

The interpretation of the XMCD signal at the K-edge of 3d metals is far from being achieved. The XMCD spectra at the K-edge of Fe, Co and Ni have been rather well reproduced in a fully relativistic multiple-scattering formalism (Ebert 1988, Stähler 1993). These calculations are however difficult to interpret physically and the correlation between XMCD and magnetic quantities is not easy. Later on Brouder et al. (Brouder 1996) have calculated the K-edge XMCD spectrum of Fe using a semi-relativistic approach of the multiple-scattering which allows to analyze the different contribution to the XMCD spectrum. The Fe dichroic signal was well reproduced and it was shown that the spin-orbit potential of the *d* states of the neighboring ions originates the first positive peak. The role of the spin-orbit on the 3 *d* states of neighbors was also reported by Igarashi and Hirai (Igarashi & Hirai 1994, Igarashi & Hirai 1996) for Fe, Co and Ni. On the same time Guo (Guo 1996) has derived two rules relating the XMCD at the K-edge to the *p*-projected spin and orbital magnetization density of the unoccupied states and came to the conclusion that XMCD at the K-edge probes the *p*-projected orbital magnetization density of these empty states. The XMCD has been also measured at the K-edge of Fe and Co in alloys (Stähler 1993) and it was observed that the structure of the signal may be different of that of pure 3d metal in some of

them. Our recent studies in the hexagonal LaCo₅ and TbCo₅ compounds have shown that the structure of the dichroic signal at the Co K-edge is very sensitive to the magnetic environment of the Co (Rueff 1998). We present here further experiments performed at the Co K-edge in the cubic compounds RCo₂, with R=Nd, Tb and Ho.

2. Experimental

XMCD measurements on NdCo₂ and TbCo₂ have been performed at LURE on the energy dispersive D11 beamline (Baudeflet 1991). Right circularly polarized beam is selected by 1 mm-wide slits positioned 3 mrad below the orbit plane, the circular polarization of the beam is $P_c \approx 0.65$. As the XAFS spectra are recorded in transmission mode, alloys were crushed into fine powder and layered on a Kapton foil. They were mounted in a helium cryogenerator inserted between the electromagnet poles and cooled down to 10 K. The intensity of the applied magnetic field is $B = 0.4$ T. The XMCD signal was obtained as the difference between two consecutive XAFS spectra with opposite direction of the field: $\mu_{\text{XMCD}} = \mu_-(B) - \mu_-(-B)$. Measurements on HoCo₂ have been performed at the ESRF ID12A beamline optimized for polarization dependent XAFS studies (Goulon, 1998). The source of circularly polarized radiation is the Helios II undulator which allows one to flip the helicity of the X-rays after each energy scan. Double crystal monochromator was equipped with a pair of Si<111> crystals cooled down to -140°C. The circular polarization rate is $P_c \approx 0.88$ in the energy range used for the experiment (Varga, 1997). The HoCo₂ sample is a monocrystalline disk ($\phi = 6$ mm, $h = 2$ mm) optically polished. It was mounted in a liquid helium cryostat inserted between the poles of a superconducting electromagnet and oriented so that its [001] easy axis coincides with the direction of the magnetic field. XAFS spectra were recorded in the total fluorescence detection mode at 20 K with a constant applied field of 3 T (under these conditions, saturation of the magnetization is completed). The detector was a Si photodiode mounted inside the cryomagnet at 90° with respect to the incident beam. The XMCD signal was obtained as a direct difference between two consecutive XAFS spectra with opposite helicities: $\mu_{\text{XMCD}} = \mu_-(B) - \mu_+(B)$. The resulting XMCD spectra were checked to be free of any artefacts by repeating the measurements with opposite direction of the magnetization.

3. Results and discussion

Figure 1 compares the XMCD spectra at the Co K edge in pure Co metal and in the RCo₂ alloys. Instead of the one negative peak structure characteristic of Co, all the RCo₂ spectra show a three-peaks structure: one negative in the middle of the positive ones. In the powdered samples the first positive peak has roughly the same intensity, while the second (negative) and third (positive) ones strongly change with the rare earth. In NdCo₂ the central negative peak is twice as intense as the negative peak in TbCo₂. The intensity of the third (positive)

peak decreases between NdCo_2 and TbCo_2 . The dichroic spectrum of HoCo_2 presents the same three-peaks structure, with in addition a small positive structure in the middle of the negative one. Such a structure is guessed in powdered samples but not resolved. This points out that the use of magnetically saturated single crystals allows a considerable gain in resolution. In powdered samples the peak intensities never exceed 0.2% but are comparable to that in Co metal while in RCo_2 the Co moment is only of $1\mu_B$. The fact that in HoCo_2 the peak intensity reaches values as large as 0.6% cannot be explained by only the Co contribution and emphasize the influence of the neighboring R ions on the XMCD spectra at

the Co K-edge. The three-peaks structure of the spectra in the RCo_2 series is very reminiscent of the XMCD structure observed in TbCo_5 (Rueff 1998). In the RCo_5 compounds the Co is a strong ferromagnet with a moment, $\mu \approx 1.6\mu_B$, close to the value in pure Co ($\mu \approx 1.7\mu_B$). The XMCD study at the Co K-edge in LaCo_5 and TbCo_5 has shown that the structure of the XMCD spectrum changes between the La and the Tb compounds. LaCo_5 presents a one-peak structure, similar to that of pure Co, while in TbCo_5 a three-peaks structure is observed. The XMCD spectra of both compounds have been calculated using the semi-relativistic approach proposed by Brouder *et al.* (Brouder 1996). In this approach the

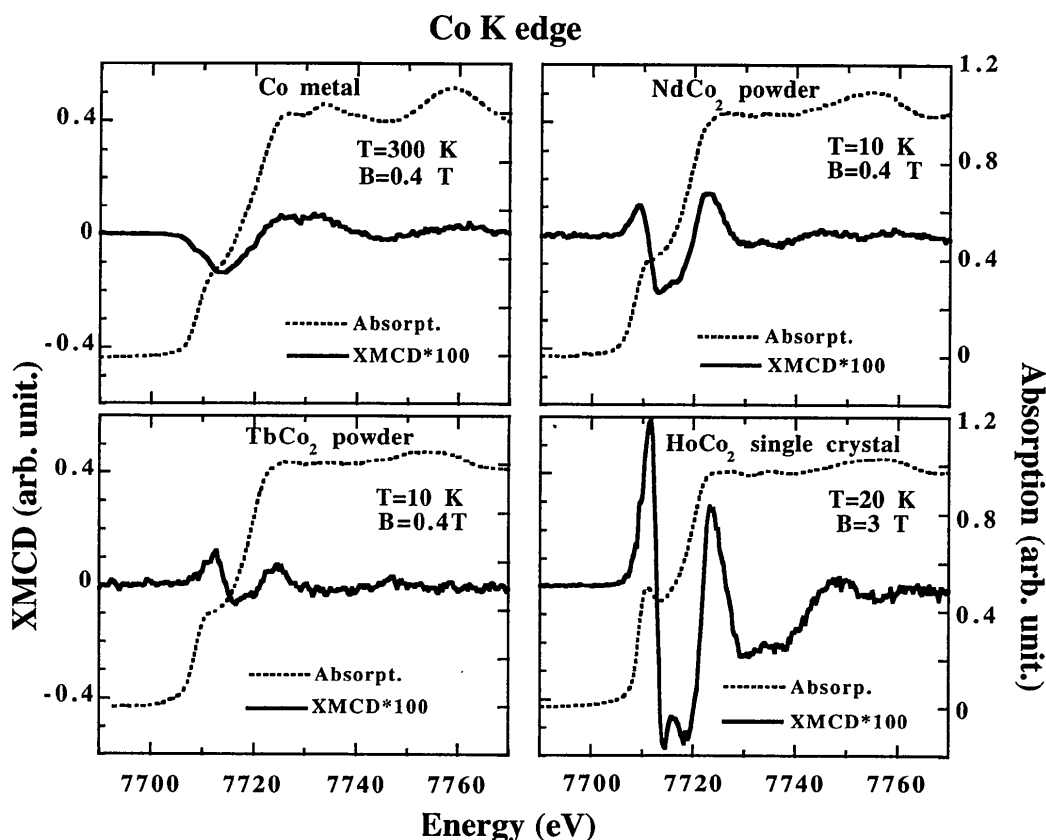


Figure 1

XMCD (left scale) and absorption (right scale) spectra at the Co K-edge in pure metal Co and in the NdCo_2 , TbCo_2 and HoCo_2 compounds. Measurements on NdCo_2 and TbCo_2 have been performed on powdered samples at 10 K and under an applied magnetic field of 0.4 T. The measurements on HoCo_2 have been performed on a single crystal at 20 K with a field of 3 T applied along the easy magnetization axis in order to reach a complete saturation of the magnetization.

XMCD spectrum is the sum of three contributions: a purely atomic contribution, related to the Fano effect, a local contribution coming from the spin polarization of the p -states and a contribution due to the scattering of the photoelectron by the spin-orbit potential of the absorber and the neighboring atoms. The expansion of this last contribution into orbitals and sites allows to analyse the physical origin of some structures in the XMCD spectra. The calculated spectra

reproduce quite well the near-edge structures (Rueff 1998). The analysis of the different contributions has shown that in Co metal the near-edge structures of the XMCD spectrum mainly result from the spin-orbit scattering by the p and d shells of the neighboring atoms. In the intermetallic compounds the scattering by the spin-orbit of the rare earth shells becomes important. In LaCo_5 , where La is non magnetic, the spin-orbit contributions from Co and La shells have comparable

amplitudes. In TbCo_5 , where Tb is magnetic, it was shown that the scattering of the photoelectron by the spin-orbit of the Tb d shells largely dominates all the other contributions and originates the three-peaks structure of the XMCD spectrum. This is consistent with a larger spin-orbit coupling in the rare earth $5d$ shell than in the transition metal $3d$ shell. Present results on the RCO_2 strongly reinforce this interpretation. Moreover they show that the influence of the rare earth is independent of the magnetic moment of the $3d$ metal. Indeed in the RCO_2 series the Co moment, induced by the rare earth reaches only $1\mu_B$, while in the RCO_5 series Co behaves as a strong ferromagnet with a moment of $1.6\mu_B$.

4. Conclusion

The comparative study in the RCO_2 compounds has confirmed the strong influence of the rare earth on the Co K-edge XMCD signal. The results can be understood, within the multiple-scattering approach, as the consequence of the diffusion of the photoelectron by the spin-orbit potential of the neighboring rare earth electronic shells. It is also shown that the use of single crystals, magnetically saturated, leads to huge and well resolved signal.

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