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Hard X-ray MCD in $GdNi_5$ and $TbNi_5$ single crystals

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XMCD experiments have been performed at the R $L_{2,3}$ and Ni K-edges on magnetically saturated single crystals of $GdNi_5$ and $TbNi_5$ ferromagnetic compounds. The spectra present huge and well structured dichroic signals at both the R $L_{2,3}$ and the Ni K-edges. Structures from the quadrupolar ($2p$ -> $4f$) transitions are clearly observed at the R $L_{2,3}$-edges. Though Ni is not magnetic, large intensities, up to 0.4%, are measured at the Ni K-edge. The Ni K-edge XMCD shows a three-peak structure which intensities dependent on the rare earth.

Keywords: XMCD, Hard X-rays, XANES.

1. Introduction

X-ray magnetic circular dichroism (XMCD), being element and orbital selective, offers the opportunity to probe the R d and TM p band states by scanning through the R L and TM K-edge. As they mediate the exchange mechanisms in the R-TM intermetallics, these band states play an important role in the magnetic properties. Unfortunately, for these edges the interpretation of the dichroic signal is still matter of debate and the validity of the magneto-optical sum rules which relate the spin and orbital magnetic moments to XMCD spectra, is not yet proved. Most of the data were obtained from polycrystalline or powdered samples, where i) the single domain phase is never reached in anisotropic systems, ii) high fields are necessary to reach the magnetic saturation in the isotropic ones. This strongly reduces the dichroic intensity and smears the small structures. The use of single crystals in the single domain phase would be very helpful to achieve a more quantitative analysis of the XMCD signal. The RNi_5 crystallize in the simple hexagonal $CaCu_5$-type structure. The $3d$ shell of Ni is almost full and its contribution to magnetism is very small. The magnetic properties are driven by the indirect bilinear exchange interactions between R ions and the crystalline electric field, responsible for the magnetocrystalline anisotropy. They appear as well suited compounds to improve our knowledge of the XMCD at the R $L_{2,3}$-edges. We present here the results of XMCD at the $L_{2,3}$-edges of the rare earth and K-edge of Ni in single crystals of $GdNi_5$ and $TbNi_5$.

2. Experimental

$GdNi_5$ and $TbNi_5$ order at $T_C = 34$ K and 23 K respectively. $GdNi_5$ is isotropic, while the strong magnetocrystalline anisotropy of the Tb leads to an easy magnetization plane perpendicular to the c axis in $TbNi_5$. The experiments have been performed at the E.S.R.F. beamline ID12A 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. Polarimetry experiments have shown that the circular polarization rate is 88% in the energy range used for these experiments (Varga 1997). The samples of parallelepipedic shape, 5 mm × 3 mm × 3 mm for $GdNi_5$ and 5 mm × 4 mm × 1.5 mm for $TbNi_5$, were optically polished. They were mounted on the cold finger of the liquid helium cryostat inserted between the poles of superconducting electro-magnet. Experiments were performed at 12 K under a magnetic field of 3 T. For the sake of comparison both samples were oriented so that the direction of the magnetic field coincides with an axis of the basal plane, easy magnetization plane for $TbNi_5$. They were magnetically saturated under these experimental conditions. A complete saturation is important to be sure that the sample is single domain.

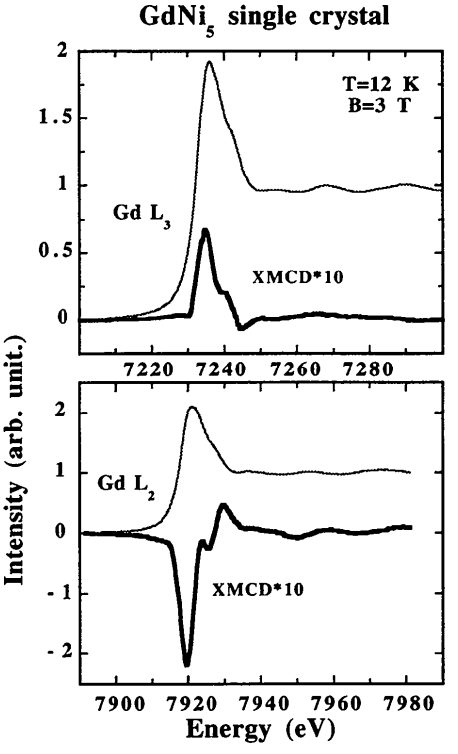

Figure 1
Absorption and XMCD spectra at the Gd $L_{2,3}$-edges measured on $GdNi_5$.

XAFS spectra were recorded in the total fluorescence detection mode. The detector was a Si photodiode mounted inside the cryomagnet at 90° with respect to the incident beam. XAFS spectra were systematically corrected for the self-absorption effects after a proper normalization to the edge jump. The XMCD signal was obtained as a direct difference between two consecutive XAFS spectra with opposite helicities: $\mu_{XMCD} = \mu_-(B) - \mu_+(B)$. 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 shows the absorption and dichroic spectra at the Gd $L_{2,3}$-edges in $GdNi_5$. In the absorption spectra the white line is followed by an other structure located at about 8 eV above the edge. The Gd L_3-edge XMCD spectrum presents a main positive peak which reaches up to 6.7%. In addition two other features are resolved: a small negative peak about 2.6 eV below the edge and a positive structure at approximately 8 eV above the edge. These features were not observed in previous measurements on powdered samples (Galera 1995). At the L_2-edge the structure above the edge has evolved in a well resolved negative peak, while no clear structure is observed below the edge. The main negative peak reaches about -21%.

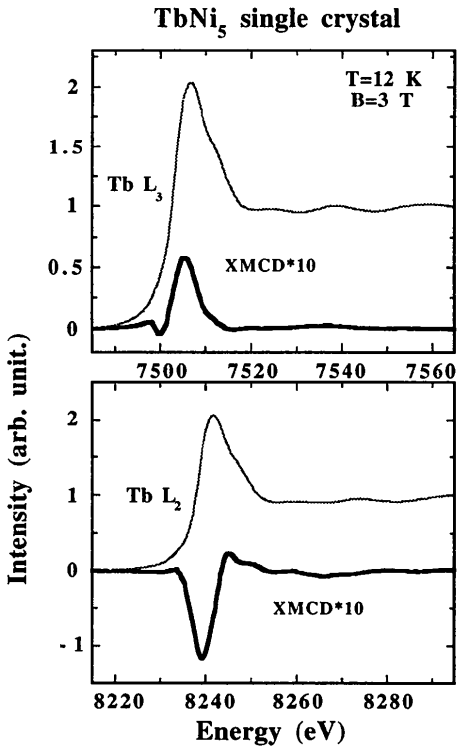

Figure 2
Absorption and XMCD spectra at the Tb $L_{2,3}$-edges measured on $TbNi_5$.

In $TbNi_5$ the absorption spectra present the same structure than in $GdNi_5$, 8 eV above the edge (Fig. 2). The Tb L_3 XMCD reveals a well resolved negative structure, 3 eV below the edge, followed by a large positive peak (up to 5.8%). At the Tb L_2 a positive structure is also observed below the main negative peak. It is now established that the quadrupolar (E_2) $2p$ -> $4f$ transitions have an important contribution to the XMCD spectrum at the R $L_{2,3}$-edges (Carra & Altarelli 1990). Experimentally E_2 structures have been evidenced at the L_3 edge in some rare earth compounds (Lang 1995, Giorgetti 1995) but never clearly observed at the L_2. In $TbNi_5$ the negative structure at the Tb L_3-edge, is very likely of quadrupolar origin. Therefore the positive structure observed at the L_2-edge has certainly the same E_2 origin, but with a smaller intensity. This is certainly why quadrupolar structures are less resolved at the R L_2-edge. This is confirmed by the results on $GdNi_5$. At the Gd L_3-edge the small negative structure 2.6 eV below the edge can be attributed to the E_2 contribution. A similar structure is no more resolved at the L_2, but the asymmetry at the bottom of the main negative peak reveals that this contribution is still present. The comparison of the R $L_{2,3}$-edges absorption spectra shows that the electronic band structure is the same for $GdNi_5$ and $TbNi_5$. The origin of the structure observed 8 eV above the edge in all the absorption spectra is very surprising. In $GdNi_5$ they are seemingly correlated with the structures observed in the XMCD spectra. Figure 3 compares the spectra measured at the Ni K-edge. The XMCD spectra show a three-peaks structure, very reminiscent of the one observed at the Co K-edge in the RCo_5 alloys (Rueff 1998). The first positive peak has roughly the same intensity in both compounds but the negative one is strongly reduced in $GdNi_5$. While Ni is not magnetic, surprisingly the XMCD presents a huge intensity compared to the signals observed so far at the K-edge of pure Ni and even at K-edge of Fe or Co metal. The peak intensities and their evolution between Gd and Tb confirm the influence of the rare earth in the structure of the XMCD spectrum at the TM K-edge in R-TM alloys.

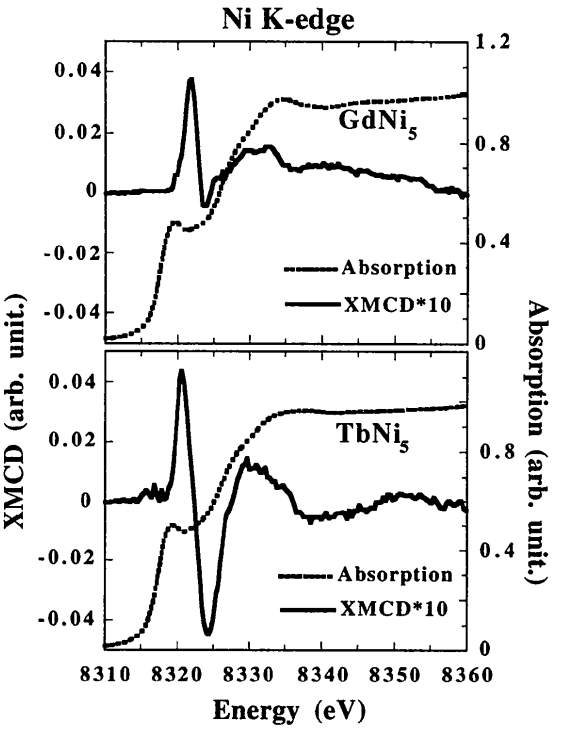

Figure 3
Ni K-edge absorption and XMCD spectra measured in $GdNi_5$ and $TbNi_5$.

4. Conclusion

XMCD experiments performed on $GdNi_5$ and $TbNi_5$ at the R $L_{2,3}$ edges and Ni K-edge have shown that the use of magnetically saturated single crystals strongly enhance the resolution of the absorption and dichroic spectra. In the XMCD spectra the structures arising from the quadrupolar transitions have been clearly observed at the R L_3 and L_2-edges. New structures above the edge have been also evidenced in the absorption spectra. In these compounds where Ni ions do not satisfy the Stoner criterion the large XMCD signals

observed at the Ni K-edge are ascribed to the influence of the rare earth. This influence is apparent from the comparison of $GdNi_5$ and $TbNi_5$ spectra.

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