I. INTRODUCTION

The rare-earth–transition-metal intermetallic compounds offer the interesting characteristic of combining the itinerant magnetism of transition metals (TM) with the localized and anisotropic magnetism of the rare earths (RE's). Among these materials, the RNi$_5$ and RCo$_5$ compounds, which crystallize in the hexagonal CaCu$_5$-type structure, have been widely investigated. The Curie temperatures of RCo$_5$ compounds are very high and the Co moment is close to its maximum value. These compounds order ferrimagnetically with the spins on the TM and RE sites coupled antiparallel. In contrast, in the RNi$_5$ series the Ni ions do not carry a magnetic moment. Magnetism comes from the RE ions only and these compounds exhibit very low Curie temperatures. The magnetic properties of these compounds are understood in terms of crystalline electric field (CEF) interactions and RE-RE, RE-TM, and TM-TM exchange interactions. The 5d-3d coupling plays an important role in RE-TM compounds, since it mediates the interactions between RE 4f and TM 3d spins.

In general, the polarization of the 5d bands in RE-TM compounds is tuned (i) by the hybridization of the 3d and 5d bands, (ii) by the positive 4f-5d direct exchange. At the formation of the RE-TM alloys the screening of the nuclear potentials are modified by the transfer of 5d (or 4d) electrons to the unfilled 3d band. The two d bands draw closer and hybridize to form bonding and antibonding bands. Using the molecular orbital formalism Brooks et al. showed that in the case of LuFe$_5$ the entire content of the 5d band is due to hybridization. Hybridization also originates the antiparallel polarization of the local 3d and 5d moments. When the RE is magnetic, the 4f-5d exchange splits the 5d band further and increases both the antiparallel 3d and 5d moments. Quite similar effects occur in the RCo$_5$ and RNi$_5$ compounds. Earlier calculations of the electronic structure of the Y-Ni compounds show that a large hybridization exists between d bands of Y and Ni atoms. This hybridization transfers 0.1–0.2 Ni 3d electrons to the Y 5d band. In RNi$_5$ compounds, the polarization of the RE 5d band is mainly due to the local 4f-5d positive exchange. Because of the 5d-3d hybridization, a net moment is then induced on Ni.

It is well known that with the use of circular polarized light of synchrotron sources, x-ray-absorption spectroscopy has become a unique probe of local magnetism. Magnetic circular x-ray dichroism (MCXD) measures the dependence of the absorption cross section on the helicity of the incident radiation. This phenomenon is governed by the selection rules for dipolar electric transitions (\(\Delta l = \pm 1\), \(\Delta J = 0, \pm 1\), and \(\Delta m_J = \pm 1\) for left and right polarization). The tunability of synchrotron radiation and the selectivity of x-ray-absorption spectroscopy allow the unoccupied electronic levels of a selected symmetry to be probed independently for each atomic species. With the use of hard x rays the magnetic polarization of 4p bands of TM and 5d bands of RE can be studied. Although the 5d electronic states are not directly responsible for the magnetism of RE compounds, they are of great interest since they govern the magnetic order through the indirect exchange with the 3d bands.

The MCXD spectra are well understood for the case of 4f magnetism, where the atomic multiplets approach can be applied (\(M_{4,5}\) edges of RE). The interpretation is still a matter of discussion for \(L_{2,3}\) edges of itinerant ferromagnetic materials Co, Ni, and Fe. Important progress has been achieved with the sum rules proposed for MCXD. It has been shown that the ground-state orbital and spin moments of the 3d band can be deduced from MCXD signal of the 2p core level split by a spin-orbit interaction. Some discussion is still open to precise the
limits of applicability of the second or "spin" sum rule.

The interpretation of the L_{2.3}-edge signals of rare-earth elements is far more complex than that of 3d metals, essentially because of the 4f-5d exchange which is at the origin of a spin-dependent spectral weight. The shape and the magnitude of the spectra strongly depend on the rare-earth atom and, for the same element, the profiles can be very different for L2 and L3 edges. For heavy rare earths like Gd, Tb, or Dy, the L2 (L3) edge spectra have a main negative peak due to dipolar transitions to the 5d band. The weak feature below the edge observed at the L3 edge has been attributed to quadrupolar transitions (2p → 4f) but there is no definite experimental proof for this interpretation.10

Using a simple one-particle model, the dipolar part of the L_{2.3}-edge MCXD spectra (μ_c) has been related to the spin-dependent final-state density by the expression11

\[ μ_c/μ_0 = (μ^+ - μ^-)/μ_0 = -P_x Δρ(E)/ρ, \]

where \( P_x = (n_1 - n_{-1})/(n_1 + n_{-1}) \) is the photoelectron polarization, where \( n^+ \) and \( n^- \) are the probabilities to create a photoelectron of a given spin. \( Δρ(E) = ρ'(E) - ρ''(E) \) is the difference between the unoccupied 5d states with opposite spins. In this approach, the radial part of the matrix element is assumed to be energy and spin independent.

Extensive studies of RE L_{2.3}-edge MCXD spectra in RE-TM compounds have shown that this model is adequate only for 5d impurities in TM (Ref. 11) and for non-magnetic rare-earth elements, when the 5d moment is induced by the hybridization with the 3d band of the TM.9,11,12 The MCXD signal has been shown to be proportional to the 5d magnetic moment in Hf, Lu, La, and intermediate valence Ce compounds. As soon as the rare earth has an incomplete 4f shell, the one-particle model fails to explain the sign and the magnitude of the MCXD signal. For example, the 5d moment deduced by scaling the MCXD signal of GdFe_2 to that of LuFe_2 is much larger than the one deduced by neutron measurements.9 Furthermore the direction of the 5d polarization deduced from the sign of the Gd L3-edge MCXD is wrong when Eq. (1) is used. The correct sign is obtained only for RE "without 4f electrons." To explain this discrepancy, Wang et al.13 argue that when the 4f shell bears a magnetic moment the spin dependence of the matrix elements of the dipolar transitions should be taken into account. The dichroic signal μ_c is better approximated by the expression

\[ μ_c/(μ^+ + μ^-) = (μ^+ - μ^-)/(μ^+ + μ^-) = -P_x(ρΔM/M_ρ), \]

where \( ΔM = M^+ - M^- \) is the difference between the spin-independent radial parts of the matrix elements. The work by Harmon and Freeman14 has shown that the spin-up 5d radial functions are pulled in towards the core of the atom with respect to the spin-down 5d orbitals. The 2p-5d overlap is therefore spin dependent. This increases the magnitude of the radial part of the matrix element of spin-up electrons with respect in this case to the quantization axis associated with the 4f spin) and causes \( M^+ \) to be larger than \( M^- \). According to Lang et al.15 the difference between \( M^+ \) and \( M^- \) can be as large as 30% in rare-earth compounds. The larger transition probability to spin-up states explains the positive sign of the L_{3}-edge dichroic spectra obtained for Gd and Tb, in spite of the fact that the number of unoccupied 5d\(^+\) states is smaller than that of 5d\(^-\) states. In other words, the term \( ρΔM \) dominates with respect to \( MΔρ \).

Since the second term in Eq. (2) is negative, it also follows that the MCXD signal decreases when the 5d polarization increases. It has been found that the L-edge signal measured for amorphous GdFe_2 is larger than that measured for crystalline GdFe_2 in spite of the larger polarization of the 5d band in the crystalline phase.15

In this paper we present the results of magnetic circular x-ray dichroism measurements performed at the L_{2,3} edges of the rare earths in La(Ni\(_x\)Co\(_{1-x}\))\(_2\), GdNi\(_5\), and TbNi\(_5\), and in the ferrimagnetic solid solutions Tb(Ni\(_x\)Co\(_{1-x}\))\(_5\). In the La(Ni\(_x\)Co\(_{1-x}\))\(_5\) solutions, as in LuFe\(_2\), the polarization of the 5d band comes exclusively from the hybridization with the 3d band. Most of the 3d moment is carried by Co and the La 5d polarization is expected to increase as the Co content increases. In GdNi\(_5\) and TbNi\(_5\) the 5d polarization is induced exclusively by the 4f-5d local exchange while in the Tb(Ni\(_x\)Co\(_{1-x}\))\(_5\) solid solutions the two aspects are combined. In these solutions the 5d polarization is expected to increase when the number of Co neighbors increases.

II. SAMPLES AND EXPERIMENTAL DETAILS

Polycrystalline GdNi\(_5\), TbNi\(_5\), and solid solutions of Tb(Ni\(_x\)Co\(_{1-x}\))\(_5\) (\( x = 0.2, 0.4, \) and \( 0.6 \)) and La(Ni\(_x\)Co\(_{1-x}\))\(_5\) (\( x = 0.4, 0.2, \) and \( 0.0 \)) were prepared at the Laboratoire Louis Neel by rf melting of the pure elements in a water-cooled crucible under argon pressure. The samples were finely powdered and deposited uniformly on a kapton foil in a layer less than 10 \( μm \) thick. The magnetic measurements on the same powders were carried out using an extraction magnetometer in magnetic fields up to 10 T. The ordering and compensation temperatures are reported in Table I as well as the ratios between the magnetization

<table>
<thead>
<tr>
<th>Sample</th>
<th>( T_C (K) )</th>
<th>( T_{comp} (K) )</th>
<th>( T_{magn}/T_C )</th>
<th>( M/M_s )</th>
</tr>
</thead>
<tbody>
<tr>
<td>GdNi(_5)</td>
<td>32</td>
<td>0.3</td>
<td>0.94</td>
<td></td>
</tr>
<tr>
<td>TbNi(_5)</td>
<td>23</td>
<td>0.4</td>
<td>0.94</td>
<td></td>
</tr>
<tr>
<td>Tb(Ni(<em>{0.2})Co(</em>{0.8}))(_5)</td>
<td>( \approx 1000 )</td>
<td>200</td>
<td>( \approx 0.01 )</td>
<td>0.98</td>
</tr>
<tr>
<td>Tb(Ni(<em>{0.4})Co(</em>{0.6}))(_5)</td>
<td>650</td>
<td>270</td>
<td>( \approx 0.01 )</td>
<td>0.98</td>
</tr>
<tr>
<td>Tb(Ni(<em>{0.6})Co(</em>{0.4}))(_5)</td>
<td>300</td>
<td>( \approx 0.03 )</td>
<td>( \approx 1 )</td>
<td></td>
</tr>
<tr>
<td>LaCo(_5)</td>
<td>800</td>
<td>0.94</td>
<td></td>
<td></td>
</tr>
<tr>
<td>La(Ni(<em>{0.4})Co(</em>{0.6}))(_5)</td>
<td>668</td>
<td>0.86</td>
<td></td>
<td></td>
</tr>
<tr>
<td>La(Ni(<em>{0.6})Co(</em>{0.4}))(_5)</td>
<td>521</td>
<td>0.98</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
under a magnetic field of 0.6 T at the temperature of the MCXD measurements and the spontaneous magnetization at the same temperature.

Except for GdNi$_5$, all the studied samples present a very strong magnetocrystalline anisotropy. In TbNi$_5$ and Tb(Ni$_{5-x}$Co$_x$)$_5$ the easy magnetization direction is along the a axis of the hexagonal cell.$^{2,16,17}$ In TbNi$_5$ the anisotropy within the basal plane is negligible while for the Tb solid solutions some anisotropy is still present within the basal plane. In single-crystal measurements, the magnetization measured in a magnetic field applied within the basal plane perpendicular to the a axis reaches only 90% of the magnetization along the a axis.$^{16,17}$

The good agreement between the magnetization values determined for our powdered samples and those found for single crystals is an indication of the good homogeneity of our powders. For GdNi$_5$ where no anisotropy is present, the magnetization of monocrystalline$^5$ and powdered samples are equivalent. In the La solid solutions the anisotropy imposed by the cobalt atoms is uniaxial along the c axis.$^{18}$ Since measurements on single crystals are not reported in the literature, our magnetization measurements on powdered samples could not be quantitative.

The MCXD measurements were carried out at the Laboratoire pour l'Utilisation du Rayonnement Electromagnétique (Orsay) on the energy-dispersion spectrometer of the 1.85-GeV DCl storage ring. The energy-dispersive setup combines an x-ray optics made of a curved Si(111) monochromator with a position-sensitive detector able to work under very high photon flux. Right circularly polarized x rays were selected by positioning a 1-mm-wide slit 3 mrad below the orbit plane of the storage ring. The MCXD signals were obtained from the XAS spectra recorded in transmission mode in a magnetic field of 0.6 T, applied alternatively in the direction parallel and antiparallel to the photon propagation vector k. The magnetization is therefore alternatively parallel and antiparallel to the x-ray beam direction and this is equivalent to the helicity reversal.

The thermal evolution of the Tb L$_3$-edge signal in TbNi$_5$ and Tb(Ni$_{5-x}$Co$_x$)$_5$ was observed down to 4.2 K using a He cryostat. For all the other compounds the low-temperature measurements were carried out down to 10 K using a commercial closed-cycle refrigerator.

The MCXD signals are presented in the figures as the difference $\mu(B)-\mu(-B)$. The spectra were normalized to the height of the absorption edge step. In all the figures the origin of the energy scale has been chosen as the inflection point of the absorption edge. In Table II (column 1) the experimental MCXD values are corrected to take into account the variation with energy of the circular polarization rate.

### III. RESULTS AND DISCUSSION

#### A. The role of the 3d-5d hybridization:

La(Ni$_{5-x}$Co$_x$)$_5$ solid solutions

In RE-TM compounds the polarization of the RE 5d band is tuned by the 4f-5d exchange and by the 3d-5d hybridization. The two mechanisms have different effects on the MCXD signals at the RE $L_{3,2}$ edges. When only the 3d-5d hybridization plays a role, the MCXD signal can be related directly to the 5d polarization, as already seen for La, Lu, and Hf compounds.$^9$ When the rare earth has an incomplete 4f shell, the presence of spin-dependent radial matrix elements makes it difficult to correlate the MCXD signal to the 5d polarization.

In La(Ni$_{5-x}$Co$_x$)$_5$ solid solutions the amplitudes of the La $L_{2,3}$-edge MCXD signals are expected to vary according to the polarization of the 5d band. The La $L_3$-edge MCXD spectra measured at 300 K for La(Ni$_{5-x}$Co$_x$)$_5$ with $x=0, 0.2$, and 0.4 are shown in Fig. 1. The signals consist of a weak single-peaked structure centered at the absorption edge, associated with $2p \rightarrow 5d$ dipolar transitions. The sign of the spectra is positive in agreement with the previous results on other La, Lu, or Hf compounds. The MCXD amplitudes are of the same order of magnitude of that measured for LuFe$_2$.$^9$

#### TABLE II. Column 1 gives the amplitude of the MCXD signal at the rare-earth $L_3$ edges for the Tb and La solid solutions. In column 2 the MCXD signal has been corrected for the $M/M_r$ ratio.

<table>
<thead>
<tr>
<th>Element</th>
<th>$L_3$ (%)</th>
<th>$L_{3,2}$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>TbNi$_5$</td>
<td>2.4±0.1</td>
<td>2.55</td>
</tr>
<tr>
<td>Tb(Ni$_{5-x}$Co$_x$)$_5$</td>
<td>2.2±0.1</td>
<td>2.2</td>
</tr>
<tr>
<td>Tb(Ni$_{5-x}$Co$_x$)$_5$</td>
<td>2.1±0.1</td>
<td>2.14</td>
</tr>
<tr>
<td>Tb(Ni$_{5-x}$Co$_x$)$_5$</td>
<td>1.8±0.1</td>
<td>1.84</td>
</tr>
<tr>
<td>La(Ni$_{5-x}$Co$_x$)$_5$</td>
<td>0.16±0.02</td>
<td>≈0.16</td>
</tr>
<tr>
<td>La(Ni$_{5-x}$Co$_x$)$_5$</td>
<td>0.25±0.02</td>
<td>0.29</td>
</tr>
<tr>
<td>LaCo$_5$</td>
<td>0.32±0.02</td>
<td>0.34</td>
</tr>
</tbody>
</table>

#### FIG. 1. MCXD spectra observed at the La $L_3$ edge in (a) La(Ni$_{5-x}$Co$_x$)$_5$, (b) La(Ni$_{5-x}$Co$_x$)$_5$, and (c) LaCo$_5$ at $T=300$ K. For clarity, the spectra have been offset by 0.4.
The MCXD amplitude decreases as Co content decreases, in agreement with the expected behavior of the 5d polarization. In Table II are reported the MCXD amplitudes corrected by the ratio $M(0.6\,\text{T},\,300\,\text{K})/M_0(300\,\text{K})$. A comparison between the three compounds shows that the MCXD amplitude is, within the experimental accuracy, proportional to the Co concentration (MCXD amplitude $\approx 0.06\%$ per Co atom). In earlier studies Brouha et al.\textsuperscript{18} have shown that for high Co concentrations ($x \leq 0.4$) the La(Ni$_{1-x}$Co$_{1+x}$)$_5$ magnetic behavior can be described in terms of a rigid band model. In this model the saturation moment per Co/Ni atom decreases by $1.1\mu_B$ replacing a Co by a Ni atom. Present results appear in good agreement with this description and therefore confirm experimentally that the La 5d band polarization is proportional to the 3d moment. It also shows that the MCXD signal can be directly correlated to the polarization $\Delta\rho$ of the La 5d band according to Eq. (2) since $M$ is spin independent.

B. The role of the 4f-5d exchange: GdNi$_5$ and TbNi$_5$

In GdNi$_5$ and TbNi$_5$, the Stoner criterion is not satisfied for Ni and the polarization of the 5d band derives exclusively from the direct 4f-5d exchange. The MCXD signal at the Gd and Tb $L_{2,3}$ edges is related to the presence of a 5d moment ($\Delta\rho$) and to the presence of a spin-dependent spectral weight [$\Delta M$ in Eq. (2)].

The MCXD spectra of GdNi$_5$ and TbNi$_5$ recorded at the $L_{2,3}$ edges of Gd and Tb at 10 K are shown in Figs. 2 and 3. The $L_3$ ($L_2$) signal consists of strong positive (negative) peak due to dipolar transitions to the 5d states. The peak maxima are observed at around $E-E_0 \approx 1$ eV and the width at half maximum is 6 eV. The features at around 30 eV in the MCXD spectra reflect the arrangement of the magnetic atoms around the probed RE site.

[Fig. 3. MCXD signal measured at $T = 10$ K at the $L_{2,3}$ edges of Tb in TbNi$_5$.]

[It is the beginning of the so-called “magnetic” extended x-ray-absorption fine structure (EXAFS)].

The shape and the sign of the MCXD spectra are coherent with those previously found for other Gd and Tb intermetallics.\textsuperscript{9,11,12} A peculiarity of GdNi$_5$ with respect to other Gd intermetallic compounds is the absence in the $L_{2,3}$-edge MCXD spectrum of the negative preedge structure, assigned in Ref. 13, to quadrupolar ($E2$) transitions to 4f states. The spectrum is however asymmetric, suggesting that a weak negative contribution may be overlapped on the low-energy side with the larger positive structure due to dipolar ($E1$) transitions. This point will be discussed in the next subsection.

In GdNi$_5$ and TbNi$_5$, the $L_{2,3}$-edge MCXD amplitudes have the same thermal variation as the bulk magnetization. In Fig. 4, we show the thermal evolution of the $L_{2,3}$-edge MCXD signal and the magnetization in TbNi$_5$. The MCXD signal is normalized to the magnetization at 10 K. Such a behavior is consistent with a 5p polarization driven only by the local 4f-5d exchange.

In order to compare the Gd and Tb signals it is necessary to take into account the effects of the magnetocrystalline anisotropy in TbNi$_5$. As the dipolar circular dichroism is proportional to the projection of the local magnetization on the photon momentum, in a powdered sample the correction can be roughly made multiplying the TbNi$_5$ amplitudes by $\frac{1}{2}$. We obtain that in GdNi$_5$ and TbNi$_5$, the $L_{2,3}$ MCXD signals have the same amplitudes $\approx 3.5\%$.

According to Eq. (2) the dichroic signal for dipolar transitions can be reduced to $\mu_\perp = \Delta M/M + \Delta\rho/\rho$. The first term measures the difference between radial matrix elements and can be of the order of 0.2–0.3 for Gd or Tb.\textsuperscript{15} The second term is negative and can be of the order of $0.6/9 \approx 0.07$ (according to band-structure calculations),\textsuperscript{3} the 5d moment of Gd and Tb in $R$Fe$_2$ compounds...
is expected to be of the order of $0.5\mu_B$ and the number of holes in the 5d band of the order of 9). It is clear that the term induced by the spin dependence of $M$ dominates the signal. In agreement with this estimation, a study of Gd-based compounds by Giorgi et al.\textsuperscript{9} and the present work show that the $L_1$-edge MCXD amplitude is of the same order of magnitude ($\approx 3\%$) for compounds as different as GdFe$_2$, Gd$_2$Fe$_{17}$, GdFe$_3$, GdCo$_2$, and GdNi$_5$.

The similar amplitudes of the MCXD signals in GdNi$_5$ and TbNi$_5$ can be explained using the same arguments. In the RF$_2$ series, Brooks et al.\textsuperscript{3} calculate a 5d moment at the RE site decreasing from Gd to Yb. This may also be understood as a decrease of the local 4f-5d exchange interaction going from Gd to heavier rare earths. The same behavior is expected in the RNi$_5$ series. Then both $\rho\Delta M/M$ and $M\Delta\rho/\rho$ scale with the 4f filling and are expected to be larger in GdNi$_5$ with respect to TbNi$_5$. The two effects having opposite signs they apparently compensate to give similar MCXD amplitudes for the two compounds. All these results point out the delicate balance between the radial matrix $\Delta M/M$ and the polarization $\Delta\rho/\rho$ terms in the MCXD amplitude.

The $L_2$ to $L_3$ amplitude ratio, $R(L_2/L_3)$, is close to $-2$ in GdNi$_5$ but largely deviates from $-2$ for TbNi$_5$ [$R(L_2/L_3) = -1.4$]. The qualitative application of the first sum rule would indicate a larger 5d orbital moment for the Tb compound. We found $5 \times 10^{-4}\mu_B$ for GdNi$_5$ and about $1 \times 10^{-3}\mu_B$ for TbNi$_5$.

Despite the fact that Ni does not carry a magnetic moment, the significant MCXD signal measured at the Ni K edge on both GdNi$_5$ and TbNi$_5$ indicates that the 4p bands of Ni are polarized. The signals show a double-peaked structure similar in magnitude and shape to the Fe K edge observed in GdFe$_2$\textsuperscript{9,15} but opposite in sign.

The double-peaked $K$-edge MCXD spectra of pure 3d metals are currently related to the weak ferromagnetic behavior of the 3d band with which the 4p band is hybridized. In the present case the polarization of the 4p band comes exclusively from the RE 5d band, since the 3d band does not carry a magnetic moment. The shape of Ni $K$-edge spectra is consistent with the weak ferromagnetic behavior of the 5d band and the sign with an anti-parallel alignment between 4p and 5d moments.

C. The relative roles of 3d-5d hybridization and 4f-5d exchange: Tb(Ni$_{1-x}$Co$_x$)$_5$ compounds

The MCXD signals of Tb(Ni$_{1-x}$Co$_x$)$_5$ solid solutions are the most complex among the compounds examined in this work, since the polarization of the Tb 5d band is related to both 3d-5d hybridization and 4f-5d direct exchange.

The $L_3$-edge MCXD spectra measured at 10 K for Tb(Ni$_{1-x}$Co$_x$)$_5$ solutions with $x = 0.6$, 0.4, and 0.2 are presented in Fig. 5 and compared with that of TbNi$_5$ at the same temperature. All the spectra present a main positive peak, asymmetric on the low-energy side due to a weak negative structure, attributed to $2p \rightarrow 4f$ quadrupolar transitions. This structure becomes more well marked when the Co concentration increases.

For Tb(Ni$_{1-x}$Co$_x$)$_5$ solutions, the observed main peak amplitudes are 1 order of magnitude larger than those observed in the La(Ni$_{1-x}$Co$_x$)$_5$ solutions. Their evolution as a function of Co concentration is opposite in the two series of compounds. In Table II are reported the amplitudes observed at the $L_3$ edge in Tb and La solutions (column 1). In column 2, the amplitudes have been corrected with the ratio $M(T;0.6 T)/M_s(T)$, where $M_s(T)$ is the spontaneous magnetic moment deduced from magnetization measurements. As the anisotropy is not very different in TbNi$_5$ and in the Tb solutions, the

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**Figure 4.** The thermal variation of the Tb $L_1$-edge MCXD signal in TbNi$_5$ is compared with the macroscopic magnetization. The MCXD signal is normalized to the magnetization at 10 K.

**Figure 5.** MCXD spectra observed at the Tb $L_3$ edge in (a) Tb(Ni$_{0.7}$Co$_{0.3}$)$_5$, (b) Tb(Ni$_{0.5}$Co$_{0.5}$)$_5$, (c) Tb(Ni$_{0.3}$Co$_{0.7}$)$_5$, and (d) TbNi$_5$ at $T = 10$ K. The MCXD spectra have been offset by 2.
MCXD amplitudes could be directly compared. However, no direct comparison is possible between the amplitudes in La and Tb solutions. Table II points out the opposite evolutions with Co concentration. Going from TbNi₃ to Co-rich solutions, we expect a progressive increase of the 5d polarization. Experimentally, the L₃-edge signal is maximum for TbNi₃ (2.4%) and decreases down to 1.8% for Tb(Ni₀.₅Co₀.₅)₃. This behavior can be explained according to Eq. (2). In Tb solutions ΔM/M dominates and should not change much with Co content. On the other hand, Δρ/ρ increases with Co content and since it is negative it leads to a global decrease of the MCXD signal.

In summary, the different behavior of the MCXD spectra for La and Tb solid solutions as a function of Co content is related to the 4f-5d exchange coupling which is responsible for a spin-dependent spectral weight. This interaction is responsible for the opposite signs of the L₃-edge MCXD spectra for magnetic and nonmagnetic RE (i.e., Tb vs La) and causes the amplitude of the spectra for magnetic RE to become larger for less polarized 5d bands.

For all the Tb compounds, a negative preedge feature, normally attributed to quadrupolar (E₂) transitions, is clearly observed in all the MCXD spectra. The fact that this feature becomes more well marked as the Co concentration increases may be interpreted in terms of the decrease of the positive dipolar contribution to the MCXD signal. The unexpected absence of a preedge feature in the spectrum of GdNi₃ (note that all the Gd compounds studied so far present a negative preedge feature) may also be explained similarly, by the fact that the negative E₂ contribution has collapsed into the much stronger E₁ structure, leaving however an asymmetry at the low-energy side of the spectrum. According to Wang et al., the position of the E₂ peak relative to the Fermi level depends only on the rare earth and ranges almost linearly from ~3.4 to ~5.35 eV going from Gd to Tm. The larger separation between E₂ and E₁ in Tb compounds can explain the fact that the preedge feature becomes more pronounced in TbNi₃ with respect to GdNi₃. We attribute the difference in balance of quadrupolar and dipolar contributions in GdNi₃ and GdF₂ spectra to the variation of Δρ/ρ due to the different 3d-5d hybridization in the two atomic environments.

Before concluding let us discuss the thermal variation of the Tb L₃-edge MCXD signal in Tb(Ni₀.₅Co₀.₅)₃. The L₃-edge MCXD spectra are shown in Fig. 6. The amplitude of the signal decreases slowly as the temperature increases. It vanishes at about 270 K, the compensation temperature, and changes its sign for higher temperatures. Figure 7 shows that the MCXD signal decreases with temperature much more slowly than the net magnetization. This difference is related to the selectivity of MCXD which probes only the RE 5d band.

At low temperature the Tb moment exceeds that of the five Co atoms. The localized 4f moment follows a Boltzmann statistics and decreases with temperature faster than the 3d moment. At the compensation temperature, the magnetization of the Tb sublattice equals that of the TM sublattice. The net magnetization and therefore the MCXD signal vanish. Beyond this temperature the magnetization of the TM sublattice becomes larger. In the presence of an external magnetic field B, the net magnetization, M, tends to align parallel to it. The Tb moments are therefore parallel to B for T < Tₐₐₐₐ and anti-parallel for T > Tₐₐₐₐ. This explains the opposite signs of the Tb L₃-edge MCXD signal on either sides of Tₐₐₐₐ. The main difference between the thermal dependence of M and the MCXD spectra is the fact that the latter mainly reflects the temperature dependence of the 4f moment via the spin-dependent matrix elements. On the other hand, the net magnetization and its thermal evolution result from the contributions of both Tb and Co moments.

**FIG. 6.** Thermal evolution of the MCXD spectra in Tb(Ni₀.₅Co₀.₅)₃ at the Tb L₃ edge. For clarity the spectra have been offset by 2.

**FIG. 7.** Comparison between the thermal variation of the Tb L₃-edge MCXD amplitude and the bulk magnetization in Tb(Ni₀.₅Co₀.₅)₃.
IV. CONCLUSIONS

This study of $R(Ni_{1/3}Co_{2/3})_5$ compounds ($R = La$, Gd, and Tb) has allowed us to investigate the effects of 3$d$-5$d$ and 4$f$-5$d$ exchanges on the RE $L_{2,3}$-edge MCXD spectra. In La compounds, where the 5$p$ polarization is induced by the hybridization with the 3$d$ band of magnetic TM, the signal can be explained using a one-particle model for the absorption process. When the 4$f$-5$d$ exchange intervenes, like in the Gd or Tb compounds, its effects on the MCXD sign and amplitude are very important. The local 4$f$-5$d$ exchange gives rise to the 5$d$ polarization but also to the difference in the radial matrix elements for spin $\uparrow$ and spin $\downarrow$ transitions. A description of $L_{2,3}$-edge MCXD taking into account the spin dependence of the radial matrix elements appears to explain qualitatively our results and let suppose that the main contribution to the MCXD amplitude comes from the spin dependence of these radial matrix elements.

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