

## Magnetic circular x-ray dichroism measurements of Fe-Co alloys and Fe/Co multilayers

S. Pizzini, A. Fontaine, E. Dartyge, and C. Giorgetti

*Laboratoire pour l'Utilisation du Rayonnement Electromagnétique, Bâtiment 209D, Centre Universitaire Paris-Sud, 91405 Orsay, France*

F. Baudalet

*Laboratoire de Physique des Solides, Université Nancy I, 54506 Vandoeuvre-les-Nancy, France*

J. P. Kappler

*IPCMS-GEMME, Université Louis Pasteur, 67070 Strasbourg, France*

P. Boher and F. Giron

*Laboratoire d'Electronique Philips, 22 Avenue Descartes, 94453 Limeil-Brévannes, France*

(Received 21 December 1993; revised manuscript received 16 March 1994)

In this paper we discuss the specific features of *K*-edge magnetic circular x-ray-dichroism (MCXD) spectra of Fe and Co in bcc Fe-Co alloys and Fe/Co multilayers. In the alloys, the systematic variations of shape and amplitude of the near-edge MCXD signals as a function of concentration can be correlated with the changes in local atomic environment. The difference between the Co *K*-edge and Fe *K*-edge spectra of the same alloy shows that the signal is central atom dependent. Well above the absorption edge ( $E - E_0 > 80$  eV) the extended x-ray-absorption fine structure (EXAFS)-like oscillations in the MCXD spectra clearly reflect the local bcc structure and appear not to depend on the absorbing atom. In both alloys and multilayers, the magnetic EXAFS intensity can be correlated to the average magnetization in the neighborhood of the absorption atom.

### I. INTRODUCTION

#### A. Fe-Co alloys and Fe/Co multilayers

The comparison between alloys and multilayers is of great interest for the understanding of the magnetism of interfaces. It was first pointed out by Néel<sup>1</sup> that the lowered symmetry at a surface may be the cause of magnetic anisotropy energies larger than those found for bulk materials. Co/*M* multilayers with [111]fcc orientation (*M* = Fe, Pd, Au, or Ir) exhibit perpendicular magnetization when Co layers are sufficiently thin. Other multilayer systems such as Co/Cu or Fe/Cu exhibit very large magnetoresistance and magnetic coupling between magnetic layers oscillating as a function of the nonmagnetic spacer.<sup>2</sup>

In this paper we present the results of *K*-edge magnetic circular x-ray dichroism (MCXD) measurements on a series of Fe-Co alloys and Fe/Co multilayers. The measurements on disordered Fe-Co model alloys have been found to be essential to help general understanding of *K*-edge MCXD signals and the interpretation of the measurements on Fe/Co multilayers.

The magnetic properties of Fe-Co alloys have been studied extensively, and are well understood. Because of their similar electronic structure and atomic volumes, Fe and Co mix with each other over a wide range of concentrations to form alloys with bcc structure in the range 0–80% Co and hcp structure for more than 80% Co. The saturation magnetization of Fe-Co alloys measured

by Weiss and Forrer<sup>3</sup> and later by Bardos<sup>4</sup> has a non-linear behavior as a function of Co concentration, with a maximum for about 30% Co. Polarized neutron-diffraction studies<sup>5</sup> indicate that this nonlinearity is due to the increase of the magnetic moment of Fe from  $2.2\mu_B$  for pure Fe to  $\approx 3\mu_B$  beyond 50% Co.

The interpretation of the magnetic behavior of bcc Fe-Co alloys has been the subject of extensive theoretical work.<sup>6–11</sup> The magnetic properties of these materials can be explained in terms of the increase of the magnetic moment on Fe with Co concentration. Calculations show that the enhancement of the Fe magnetic moment is due to the progressive increase of the exchange energy between the two spin subbands which leads to a filling of the majority band (pure Fe is a weak ferromagnet). For 50% Co, Fe acquires a strong ferromagnetic behavior and its moment remains constant at around  $3\mu_B$ . Since in Co the majority band is filled and no charge transfer occurs in the formation of the alloy, the moment of cobalt changes little (from  $1.65\mu_B$  to  $1.83\mu_B$ ) as a function of concentration.

Because of the different magnetic behavior of Co and Fe atoms, Fe-Co alloys are very attractive model materials for a site-selective probe such as MCXD. Fe-Co alloys have recently been studied by magnetic circular dichroism at the  $L_{2,3}$  edge by Idzerda *et al.*<sup>12</sup> The variation of the intensity ratio between  $L_3$  and  $L_2$  edge MCXD signals indicates a variation of the orbital moment as a function of Fe or Co concentration, and suggests that the orbital contribution to the magnetic mo-

ment plays an important role in these transition-metal alloys.

The interest of Fe/Co multilayers resides in their high magnetization, which makes them promising magnetic head materials.<sup>13–15</sup> In this paper, using the selectivity of MCXD and a comparison with Fe-Co alloys, we will show that their high magnetization cannot be attributed only to the alloying effect at Fe-Co interfaces.

### B. Magnetic circular x-ray dichroism

Magnetic circular x-ray dichroism is a probe of the local magnetic properties of ferromagnetic or ferrimagnetic materials. MCXD is the difference between the absorption of circularly polarized x rays by a material magnetized by a magnetic field applied respectively in the direction antiparallel and parallel to the propagation vector of the x-ray beam. The MCXD signal is usually defined as the absorption difference normalized to the total absorption  $I_{\text{MCXD}} = (\mu^- - \mu^+) / (\mu^- + \mu^+)$ .  $\mu^-$  ( $\mu^+$ ) is the absorption coefficient of the sample when the magnetic field is applied antiparallel (parallel) to the propagation direction of an x-ray beam with right circular polarization.

The first magnetic x-ray dichroism measurements in the x-ray domain have been carried out by van der Laan *et al.* with linearly polarized soft x rays,<sup>16</sup> and by Schütz *et al.* with circularly polarized hard x rays.<sup>17</sup> Since then, a larger number of measurements have been carried out at synchrotron radiation sources both with hard and soft x rays. Several systems have been investigated, ranging from rare-earth-transition-metal compounds<sup>18–20</sup> to transition metals and alloys,<sup>21–23</sup> paramagnetic thin films,<sup>24</sup> and magnetic multilayers.<sup>25–30</sup> Magnetic dichroism signals deriving from localized electronic states ( $4f$  states for  $M_{4,5}$  edges of rare-earth elements) can be described fully using the atomic multiplets formalism.<sup>31</sup>

The interpretation of MCXD at the  $K$  edge of transition metals and at the  $L_{2,3}$  edges of rare earths, which probe conduction-band magnetism, is much more complex and is still the subject of theoretical developments.<sup>32–34</sup> The occurrence of MCXD can be simplified and visualized as a two-step model. The absorption of circularly polarized photons creates a photoelectron which is spin polarized due to the spin-orbit interaction either in the initial or final states. Thanks to the electric dipole selection rules, this photoelectron is also strongly angular momentum polarized. The photoelectron is emitted into a band which is polarized by exchange and therefore behaves like a “spin detector.” For a free magnetic atom, the use of left circularly polarized photons induces transitions preferentially to spin-down states for  $L_2$  edges, and to spin-up states of  $L_3$  edges.

MCXD measurements benefit from the double selectivity of x-ray-absorption spectroscopy. They are sensitive to the chemical species of the absorbing atom and, thanks to the electric dipole selection rules, to the symmetry of the unoccupied electronic states probed by the photoelectron.

In some cases, as for  $L_{2,3}$  edges of  $5d$  impurities in Fe,<sup>35</sup> the MCXD signal is proportional to the relative spin polarization of the unoccupied final states

$\Delta\rho/\rho = (\rho^+ - \rho^-) / (\rho^+ + \rho^-)$ . In these cases, the MCXD signal can be used to estimate the value of the symmetry-projected magnetic moment on the absorbing atom. According to Wang *et al.* this single-particle model is not correct when applied to  $L_{2,3}$  edge spectra of rare-earth metals, because it neglects the spin dependence of the transition matrix elements induced by the  $5d$ - $4f$  exchange. Because of their stronger exchange interaction with the  $4f$  orbitals, the spin-up  $5d$  radial functions are “pulled in” relative to the spin-down functions, and cause the  $L_3$  edge spectra to be dominated by spin-up transitions.<sup>33</sup>

Recent calculations<sup>36</sup> have also shown that, assuming that the matrix elements are spin and energy independent, the full  $L_{2,3}$  dichroism spectrum can be related by sum rules to the ground-state expectation value of the orbital and spin moments of the absorbing atom. The sum rules have been applied, for example, to demonstrate an enhancement of the interfacial orbital moment on cobalt in Co/Pd multilayers.<sup>27</sup>

The interpretation of  $K$ -edge ( $1s \rightarrow 4p$ ) MCXD spectra is still far from being resolved. The amplitude of  $K$ -edge MCXD signals is two orders of magnitude smaller than the corresponding  $L_{2,3}$ -edge signals, since the weak spin-orbit interaction in the  $4p$  band is at the origin of the effect. Whether the spin polarization or the orbital polarization dominates the  $K$ -edge signal is still an open question. Calculations<sup>37,38</sup> show that the  $K$ -edge signals cannot easily be related to the density of unoccupied  $4p$  states of opposite spins. Multiple-scattering calculations<sup>39</sup> are able to reproduce reasonably well the experimental spectrum of pure Fe and Co. Fully relativistic band-structure calculations are in good agreement with spin-polarized measurements for Fe but not for Ni.<sup>38</sup> Systematic  $K$ -edge studies<sup>38</sup> show that MCXD  $K$ -edge spectra of  $3d$  transition-metal compounds are not easily correlated to their magnetic moments. Stähler, Schütz, and Ebert suggest that, in the case of Co, a correlation exists between the MCXD signal and the  $p$  moments. As a general rule, variations of MCXD amplitudes are correlated to variations in the local magnetization, and the spectra can therefore be studied as a function of an external parameter such as the intensity of the magnetic field, the temperature, or the applied pressure.

The different shapes of the  $K$ -edge MCXD spectra of Fe on one side and Co and Ni on the other may be correlated qualitatively respectively with the weak or strong ferromagnetic nature of the two types of materials. The MCXD spectrum of pure iron exhibits a double-peaked structure, with positive and negative peaks covering a total energy range of about 17 eV around the absorption edge (see Fig. 3). The spectra of Co and Ni exhibit only a negative peak over an energy width of about 15 eV. Fe is a weak ferromagnetic material, i.e., its Fermi level crosses both the majority- and the minority-spin  $d$  bands, while cobalt and nickel are strong ferromagnets, and their majority-spin band is full. It has been suggested that the positive peak could be related to the dominant spin-up  $d$ -like density of states close to the Fermi level, specific of Fe, while the negative peak observed at higher energies would be related to the spin-down density of states for

Fe, Co, and Ni. Multiple-scattering calculations<sup>39</sup> partly support this qualitative correlation, as they show that the positive peak observed in Fe *K*-edge MCXD spectrum of pure Fe is related to the *d* component of the photoelectron scattered by the spin orbit of the neighboring atoms.

Although the dichroic signal is stronger close to an absorption edge, important oscillations are observed up to several hundred eV above this energy. These modulations, sometimes referred to as the magnetic extended x-ray-absorption fine structure (EXAFS), are the magnetic analogs of the oscillations in the spin-averaged absorption spectra. They are mostly caused by an exchange contribution to the backscattering amplitude of the photoelectron, when the neighboring atoms are magnetic. The periods and amplitudes of the oscillations can be related to the radial distribution of the magnetic atoms around the absorption atoms, i.e., to the local magnetic structure.<sup>40,41</sup>

## II. SAMPLE PREPARATION AND EXPERIMENTS

Disordered alloys of composition Fe<sub>20</sub>Co<sub>80</sub>, Fe<sub>50</sub>Co<sub>50</sub>, and Fe<sub>70</sub>Co<sub>30</sub> were prepared from high-purity Co and Fe using a conventional triarc furnace under Ar atmosphere. The as-cast ingots were crushed and ground for 48 h to obtain a particle size of about 5 μm. The grinding treatment was performed in a solution of *n*-hexane to prevent oxidation and overheating. The bcc structure of the particles was checked by x-ray powder diffraction using a curved INEL detector.

Fe/Co multilayers of nominal composition Fe<sub>11</sub> Å Co<sub>20</sub> Å, Fe<sub>17</sub> Å Co<sub>20</sub> Å, Fe<sub>21</sub> Å Co<sub>10</sub> Å, Fe<sub>21</sub> Å Co<sub>30</sub> Å and Fe<sub>21</sub> Å Co<sub>108</sub> Å were prepared at the Laboratoire d'Electronique Philips by UHV rf sputtering by deposition both on Si(111) and kapton substrates. The samples were characterized by x-ray-diffraction, nuclear-magnetic-resonance (NMR) and x-ray-absorption measurements.<sup>14,15</sup> The samples are polycrystalline, have sharp interfaces [1–2 monolayers (ML)], and their structure is bcc(110) for Co layers thinner than about 25 Å. NMR measurements indicate that in thicker layers Co changes to a mixed hcp/fcc structure before attaining hcp structure. Magnetization measurements on these multilayers<sup>14</sup> indicate that, close to the interface, over a region of about two layers of Co and three layers of Fe larger than the interdiffusion region, the magnetization is considerably enhanced with respect to the bulk values, reaching an average value of 2.7μ<sub>B</sub>/atom compared with 2.2μ<sub>B</sub> for pure Fe. Away from the interface, Fe and Co have the saturation magnetization characteristic of bulk bcc phases. The magnetization measurements also indicate that the magnetic moment on bcc Co is about 1.65μ<sub>B</sub>, in agreement with previous measurements.<sup>42</sup>

Magnetic circular x-ray dichroism measurements on Fe-Co alloys and Fe/Co multilayers were carried out on the energy dispersive x-ray-absorption beamline of the positron-injected storage ring DCI at the French synchrotron radiation facility LURE. The exceptionally long lifetime (≈100 h) and the good spatial stability of the source are beneficial for such low signal measure-

ments. Right circularly polarized photons (polarization rate about 80%) were selected by positioning a 1-mm-wide slit 3 mrad below the synchrotron orbit plane. The x-ray-absorption spectra were measured using a Si(111) curved polychromator, focusing at 80 cm away, in the gap between the poles of an electromagnet. Higher harmonics were rejected by a zerodur mirror positioned before a photodiode array detector. The absorption spectra were measured, in transmission geometry, in a magnetic field applied alternatively parallel (μ<sup>+</sup>) and antiparallel (μ<sup>-</sup>) to the propagation direction of the x-ray beam. The magnetic-field direction was switched typically 500 times between the two positions and the total acquisition time was about 8 h for the most diluted samples. For the measurement of the alloys spectra, the intensity of the applied magnetic field was 1 T. For the multilayers a glancing angle geometry was used to enhance the probed volume. In this geometry the magnetic field was in the plane of the layers. In this case the poles of the electromagnet were kept farther apart, and the magnetic field on the samples was 0.4 T. In both cases the applied magnetic field was sufficient to obtain magnetic saturation.

The absorption spectra of alloys and multilayers were measured at the Fe and Co *K* edges. The difference spectra (μ<sup>+</sup> - μ<sup>-</sup>) were normalized by division by a constant value, the absorption edge step height. In this paper this normalized difference spectrum will be referred to as the MCXD signal. In all samples its amplitude is of the order of 1–2 × 10<sup>-3</sup>. The origin of the energy scale was chosen as the steepest point in the raising edge (7112 eV for the Fe *K* edge, 7709 eV for the Co *K* edge, and 8979 eV for the Cu *K* edge).

In order to obtain accurate information about the local structure around Fe and Co in the Fe/Co multilayers, EXAFS spectra at the Co and Fe *K* edges were measured by electron yield on beamline D41 at LURE, using a Si(331) channel cut monochromator. The EXAFS spectra were simulated in the wave-vector region 3.4–12 Å<sup>-1</sup> using the curved-wave program Excurv90 and *ab initio* phase shifts and backscattering phases.<sup>43</sup>

## III. STRUCTURE OF Fe-Co ALLOYS AND Fe/Co MULTILAYERS

The main structures of a *K*-edge x-ray-absorption near-edge spectrum are a fingerprint of the local crystallographic structure around the absorbing atom. Near-edge spectra allow a bcc structure to be easily distinguished from a hcp/fcc structure.

The most representative near-edge x-ray-absorption spectra measured for Fe-Co alloys and Fe/Co multilayers are shown in Fig. 1, where they are compared with the *K*-edge spectra of pure Fe and Co. The spectra of the pure Fe and Co metals can be used as models for bcc and close-packed structures, respectively. The measurements confirm that for each of the alloys the local structure around Fe and Co is bcc.

X-ray-diffraction and NMR measurements have shown that the structure of Fe/Co multilayers is bcc for thin Co layer thickness, and that it evolves toward hcp/fcc as the Co thickness increases beyond about 25 Å.<sup>15</sup> The evolu-

tion of the local structure of Co from a well-defined bcc (10-Å Co) to a hcp phase (108-Å Co) is confirmed by x-ray-absorption measurements (Figs. 1 and 2). Simulations show that for 25-Å Co the near-edge spectrum can be obtained as a mixed 75% bcc–25% hcp phase.<sup>15</sup> The simulations of the Co *K*-edge EXAFS spectra as a function of Co thickness give a better insight into the local Co structure. The Fourier transforms (FT's) of the Co *K*-edge spectra of  $\text{Fe}_{21\text{Å}}\text{Co}_{10\text{Å}}$ ,  $\text{Fe}_{17\text{Å}}\text{Co}_{20\text{Å}}$ ,  $\text{Fe}_{21\text{Å}}\text{Co}_{30\text{Å}}$ , and  $\text{Fe}_{21\text{Å}}\text{Co}_{108\text{Å}}$  multilayers, measured with x-ray beam polarization in the plane of the layers, are shown in Fig. 2. For 10-Å and 20-Å Co the FT's present features typical of the bcc structure of Fe. For 30-Å Co the first-neighbor peak narrows and the features at higher distances decrease in intensity. Finally for 108-Å Co the FT presents all features typical of the hcp bulk structure. The results of the simulations of the Co *K*-edge EXAFS spectra are shown in Table I. For multilayers with  $t_{\text{Co}} = 10\text{--}20\text{Å}$ , the first peak of the FT could be simulated with two neighbor distances  $R1 = 2.45\text{--}2.46\text{Å}$  and  $R2 = 2.83\text{--}2.85\text{Å}$ , typical of a bcc structure with a nearest-neighbor distance contracted with respect to bulk Fe. For  $t_{\text{Co}} = 30\text{Å}$  the first peak is best fitted with a nearest neighbor at 2.46 Å and a very disordered second-neighbor shell. The large Debye-Waller factor of the second shell and the weaker high-distance peaks probably reflect the larger disorder associated with a mixed bcc/hcp phase. The same trends in the absorption spec-

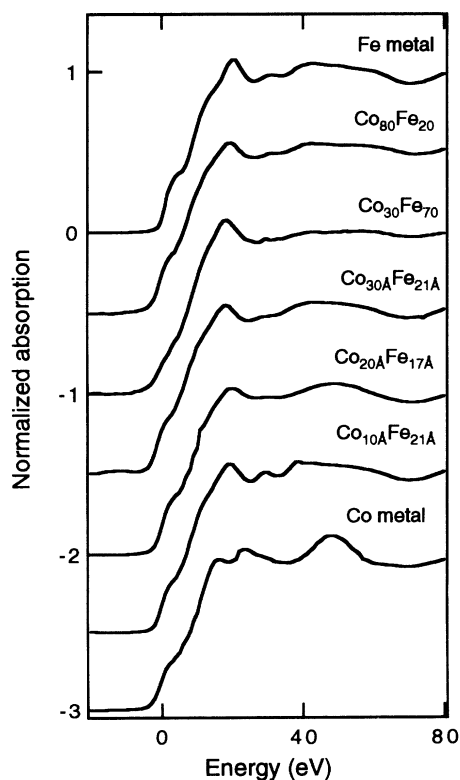


FIG. 1. The Co *K* near-edge x-ray-absorption spectra for some representative Fe-Co alloys and Fe/Co multilayers are compared with the Co *K* edge of pure hcp Co and the Fe *K*-edge spectrum of pure bcc Fe.

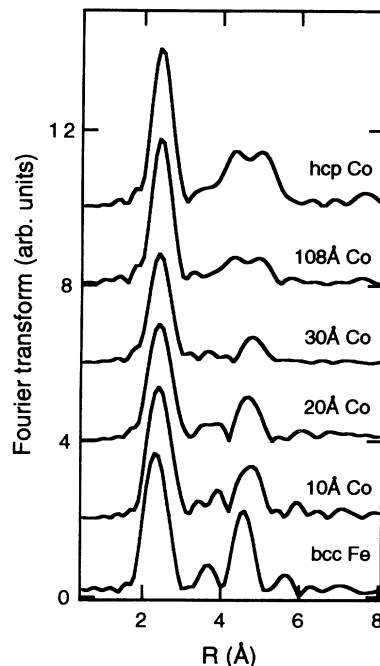


FIG. 2. The Fourier transforms (FT) of the in-plane Co *K*-edge EXAFS spectra of the multilayers  $\text{Fe}_{21\text{Å}}\text{Co}_{10\text{Å}}$ ,  $\text{Fe}_{17\text{Å}}\text{Co}_{20\text{Å}}$ ,  $\text{Fe}_{21\text{Å}}\text{Co}_{30\text{Å}}$ , and  $\text{Fe}_{21\text{Å}}\text{Co}_{108\text{Å}}$  ( $3\text{--}12\text{Å}^{-1}$ ) are compared with the FT of bulk Fe at the Fe *K* edge and of hcp Co at the Co *K* edge. For clarity, the spectra have been shifted with respect to bulk Fe.

tra have been found in other EXAFS studies of Ru/Fe and Co/Fe multilayers.<sup>15,44</sup> For the largest thickness (108-Å Co), Co layers acquire a hexagonal structure which is stable in the bulk material.

The Fe layers present a bcc structure typical of the bulk material for each Fe thickness. The first-neighbor distance ( $R1 = 2.48\text{Å}$ ) is the same as for bulk Fe.

#### IV. NEAR-EDGE MCXD SPECTRA

##### A. Results

The Fe and Co *K*-edge MCXD spectra measured for Fe-Co alloys and Fe/Co multilayers are shown in Figs.

TABLE I. Coordination numbers ( $N_1, N_2$ ), distances ( $R_1, R_2$  in Å) and Debye-Waller factors ( $\sigma_1, \sigma_2$  in Å<sup>2</sup>) for the two nearest-neighbor shells of Co, obtained from the theoretical simulations of the Co *K*-edge EXAFS spectra of hcp Co and Fe/Co multilayers. The spectra of the multilayers were measured with the x-ray polarization direction in the plane of the (110) layers. The structural parameters are compared with those obtained for bulk bcc Fe.

Sample	$N_1$	$R_1$	$\sigma_1$	$N_2$	$R_2$	$\sigma_2$
Fe bcc	8	2.48	0.013	6	2.86	0.018
10-Å Co	8	2.46	0.016	6	2.84	0.020
20-Å Co	8	2.45	0.015	6	2.84	0.027
30-Å Co	8	2.46	0.016	6	2.73	0.047
108-Å Co	12	2.50	0.017			
Co hcp	12	2.50	0.016			

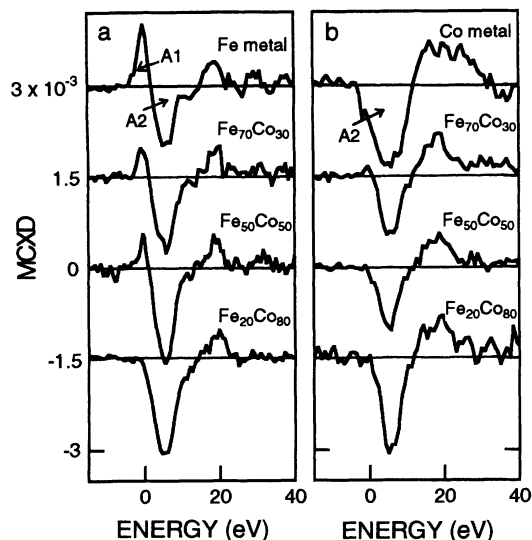


FIG. 3. Fe  $K$  and Co  $K$  near-edge MCXD spectra for bcc Fe and hcp Co and for the  $\text{Fe}_{70}\text{Co}_{30}$ ,  $\text{Fe}_{50}\text{Co}_{50}$  and  $\text{Fe}_{20}\text{Co}_{80}$  alloys. The energy axis is scaled to be absorption edge positions and the MCXD signals are normalized to the absorption edge step height. (a) Fe  $K$  edge; (b) Co  $K$  edge. For convenience, the spectra have been shifted with respect to bcc Fe in (a) and hcp Co in (b).

3–5, where they are compared with the spectra of bcc Fe and hcp Co metals.

The shape and amplitude of the near-edge MCXD signal of the Fe-Co alloys strongly depend on the alloy composition. Up to 50% Co the shape of the Fe  $K$ -edge MCXD signal [Fig. 3(a)] is similar to that of pure Fe, with a positive peak ( $A1$ ) centered at the absorption edge ( $E=0$ ) followed by a negative peak ( $A2$ ) at around 6 eV. As Co concentration increases, the amplitude of peak  $A1$  decreases and the amplitude of peak  $A2$  increases with respect to bulk Fe. In the  $\text{Fe}_{20}\text{Co}_{80}$  alloy, where most Fe atoms are surrounded by Co, the positive peak disappears and the MCXD signal consists of a single and wider neg-

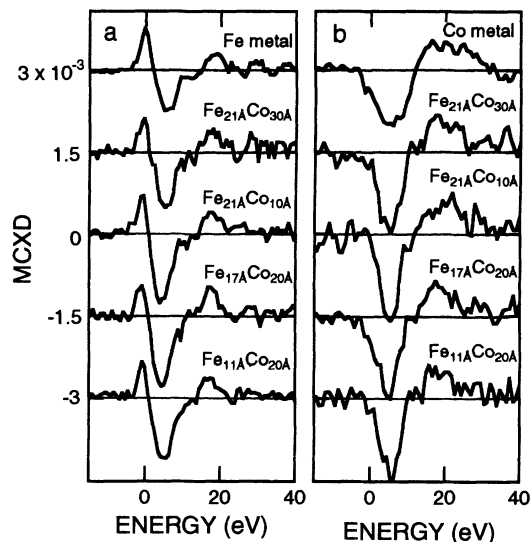


FIG. 4. Near-edge MCXD spectra for Fe/Co multilayers. The energy axis is scaled to the absorption edge positions, and the MCXD signals are normalized to the absorption edge step height. (a) Fe  $K$ -edge spectra compared and shifted with respect to Fe metal. (b) Co  $K$ -edge spectra compared and shifted with respect to hcp Co.

ative peak, twice as intense as the corresponding negative peak in pure Fe. The integrated intensities of peaks  $A1$  and  $A2$  are reported in Table II. The maximum amplitude of peak  $A2$  ( $A2^*$ ) is also shown in the table.

The Fe  $K$ -edge MCXD spectra measured for Fe/Co multilayers exhibit the double-peaked structure typical of Fe metal [Fig. 4(a)]. The negative peak  $A2$  is importantly enhanced with respect to pure metal, and its integrated intensity decreases slightly as the Fe thickness increases (Table III). The integrated intensity  $A2$  measured for the  $\text{Fe}_{11}\text{Å}\text{Co}_{20}\text{Å}$  multilayer is twice as large as that measured for pure Fe. While for Fe-Co alloys the increase of  $A2$  with respect to pure Fe is matched by a decrease of  $A1$ ,

TABLE II. The integrated intensities ( $A1$  and  $A2$ ) and the energy widths ( $\Delta E1$  and  $\Delta E2$ ) of the two near-edge peaks of the Fe  $K$ -edge and Co  $K$ -edge MCXD spectra measured for the Fe-Co alloys (see Fig. 3) are compared with the  $d$  moments measured by Bardos (Ref. 4).  $A2^*$  is the maximum amplitude of the negative peak. Uncertainties in  $A1$  and  $A2$  are of the order of 10–15%.

Sample	Fe $K$ edge					
	$A1$ ( $10^{-3}$ )	$A2$ ( $10^{-3}$ )	$A2^*$ ( $10^{-3}$ )	$\Delta E1$ /eV	$\Delta E2$ /eV	$\mu_{d,\text{Fe}}/\mu_B$
Fe (bcc)	3.1	-4.9	0.95	6	13	2.2
$\text{Fe}_{70}\text{Co}_{30}$	1.8	-6.9	1.30	4.5	13	2.77
$\text{Fe}_{50}\text{Co}_{50}$	1.6	-8.8	1.60	5.2	12.2	3.00
$\text{Fe}_{20}\text{Co}_{80}$		-11.0	1.55		16	3.00
Sample	Co $K$ edge					
	$A1$ ( $10^{-3}$ )	$A2$ ( $10^{-3}$ )	$A2^*$ ( $10^{-3}$ )	$\Delta E1$ /eV	$\Delta E2$ /eV	$\mu_{d,\text{Co}}/\mu_B$
$\text{Fe}_{70}\text{Co}_{30}$	0.3	-5.6	0.95	3	10.7	1.71
$\text{Fe}_{50}\text{Co}_{50}$		-6.0	1.0		13.2	1.77
$\text{Fe}_{20}\text{Co}_{80}$		-9.5	1.5		11.8	1.83
Co (hcp)		-12.4	1.3		15.4	1.7

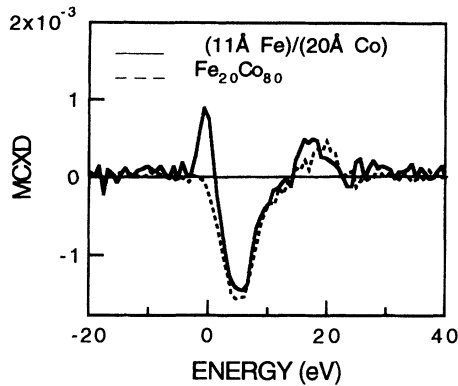


FIG. 5. Comparison between the Fe  $K$ -edge MCXD spectrum of the  $\text{Fe}_{11\text{Å}}\text{Co}_{20\text{Å}}$  multilayer and the  $\text{Fe}_{20}\text{Co}_{80}$  alloy. In both samples the negative peak is enhanced with respect to Fe, but a positive peak is present only for the multilayer.

the amplitude of peak  $A1$  measured for each of the multilayers is nearly identical to that of Fe. In Fig. 5 the Fe  $K$ -edge MCXD spectrum of the  $\text{Fe}_{20}\text{Co}_{80}$  alloy is compared with the spectrum measured for the  $\text{Fe}_{11\text{Å}}\text{Co}_{20\text{Å}}$  multilayer. While the amplitudes of the negative peaks are similar, and both enhanced with respect to pure Fe, a positive peak is observed only for the multilayer sample.

The Co  $K$ -edge MCXD spectra measured for Fe-Co alloys and Fe/Co multilayers consist of a main negative peak ( $A2$ ) as for bulk hcp Co [Figs. 3(b) and 4(b)]. The energy width  $\Delta E$  of this peak (10–12 eV) is smaller than that of pure hcp Co ( $\approx 15$  eV) and similar to that of the negative peak  $A2$  at the Fe  $K$ -edge spectrum.  $\Delta E$  therefore depends on the crystallographic structure around the absorbing atom. The shape and intensity of the Co  $K$ -edge MCXD signal of the Fe-Co alloys depend on the alloy composition. The integrated intensity of the negative peak decreases as the Fe concentration increases. In the  $\text{Fe}_{70}\text{Co}_{30}$  alloy the amplitude of the negative peak  $A2$  is about 50% of that measured for the  $\text{Fe}_{20}\text{Co}_{80}$  alloy, and a weak positive peak is superimposed to the negative peak on its low-energy side. On the other hand, for the Fe/Co multilayers the amplitude of the negative Co  $K$ -edge peak changes slowly with Co thickness and is enhanced with respect to the intensity observed for all Fe-Co alloys.

TABLE III. The integrated intensities ( $A1$  and  $A2$ ) of the near-edge peaks of the Fe  $K$ -edge and Co  $K$ -edge MCXD spectra measured for the Fe/Co multilayers (Fig. 4) are compared with bcc Fe and hcp Co. Uncertainties in  $A1$  and  $A2$  are of the order of 10–15%.

Sample	$A1$ ( $10^{-3}$ )	$A2$ ( $10^{-3}$ )	$A2$ ( $10^{-3}$ )
	Fe $K$	Fe $K$	Co $K$
Fe (bcc)	3.1	−4.9	
$\text{Fe}_{21\text{Å}}\text{Co}_{30\text{Å}}$	3.1	−7.4	−13.4
$\text{Fe}_{21\text{Å}}\text{Co}_{10\text{Å}}$	3.3	−8.8	−12.7
$\text{Fe}_{17\text{Å}}\text{Co}_{20\text{Å}}$	3.2	−8.8	−13.4
$\text{Fe}_{11\text{Å}}\text{Co}_{20\text{Å}}$	3.0	−10.1	−11.3
Co (hcp)			−12.4

## B. Interpretation

### 1. Fe-Co alloys

The MCXD data for Fe-Co alloys may be discussed on the basis of the results of band-structure calculations by Hamada.<sup>10</sup> These calculations show that as Co concentration increases, Fe changes from weak to strong ferromagnetic behavior thanks to the progressive filling of the majority  $d$  band. This occurs essentially at the expense of the minority-band filling, since the Co moment does not show any significant change. We have seen that the difference between  $K$ -edge MCXD signals for strong (Co and Ni) and weak ferromagnetic materials (Fe), single peaked in the first case and double peaked in the second, suggests that the presence of a positive peak in Fe might be related to the presence of unoccupied majority  $d$  states close to the Fermi level. This interpretation is supported by the fact that the structure in the absorption spectrum corresponding to this positive peak is due to transitions to  $4p$  states which are hybridized with the  $3d$  states of the surrounding atoms.<sup>45,46</sup>

Within this interpretation, the decrease of the integrated intensity of the Fe  $K$ -edge positive peak with increasing Co concentration is in agreement with the progressive filling of the majority  $d$  spin band of Fe. The disappearance of the positive peak for the  $\text{Fe}_{20}\text{Co}_{80}$  alloy would then indicate the complete filling of the majority spin band of Fe. At the same time the presence of a single negative peak in the Co  $K$ -edge MCXD signal could be related to the fact that Co has a strong ferromagnetic behavior for all concentrations.

Note, however, that band-structure calculations, in agreement with experimental determinations,<sup>4</sup> predict that Fe should acquire strong ferromagnetic behavior for about 50% Co, while a positive peak is still observed in the MCXD spectrum of the  $\text{Fe}_{50}\text{Co}_{50}$  alloy. A similar behavior was observed for the MCXD data on a series of fcc Ni-Fe alloys measured by Sakurai *et al.*<sup>22</sup> The Fe  $K$ -edge spectrum of  $\text{Fe}_{50}\text{Ni}_{50}$  alloy exhibits a positive peak even if Fe has strong ferromagnetic behavior. In summary, in both Fe-Co and Fe-Ni alloys, magnetically saturated Fe exhibits a two-peaked MCXD spectrum. This result seems to weaken the idea that the double-peaked structure of the MCXD spectrum simply reflects the filling of the two  $d$  spin subbands of the absorbing atom.

Our results support an alternative interpretation which stems from multiple-scattering calculations.<sup>39</sup> The calculation show that close to the absorption edge the MCXD structures arise mainly from the scattering of the  $p$  component of the photoelectron on the absorbing atom, and from the  $p$  and  $d$  components on the neighbors by the spin orbit of the neighboring sites. The positive contribution calculated for Fe arises from the scattering of the  $d$  component on neighboring sites. The absence of this positive contribution in the spectrum of hcp Co probably derives from the different polarization of the  $d$  bands of the two metals.

Our results show that when Fe is surrounded entirely by Fe, as in pure Fe, the Fe  $K$ -edge MCXD signal presents a double-peaked structure, while when Fe is sur-

rounded mainly by Co, as in the  $\text{Fe}_{20}\text{Co}_{80}$  alloy, it presents a single negative peak. The Fe  $K$ -edge MCXD spectra for the two intermediate concentrations  $\text{Fe}_{50}\text{Co}_{50}$  and  $\text{Fe}_{70}\text{Co}_{30}$  can be obtained as linear combinations of the spectra of pure Fe and of the  $\text{Fe}_{20}\text{Co}_{80}$  alloy, using weights proportional to the respective concentrations of Fe and Co in the alloys. The spectra can then be visualized as a superposition of two contributions, one coming from  $\text{Fe}^*\text{-Co}$  pairs of atoms giving a negative signal, and one from  $\text{Fe}^*\text{-Fe}$  pairs giving a double-peaked structure ( $\text{Fe}^*$  being the central atom and Fe and Co the neighboring atoms). The fact that the Fe  $K$ -edge spectra of the alloys can be obtained as linear combinations of contributions coming from  $\text{Fe}^*\text{-Fe}$  and  $\text{Fe}^*\text{-Co}$  pairs of atoms indicates that the changes in the signal are determined by the modification of the local environment of the absorbing atom. On the other hand, the fact that the Co and Fe  $K$ -edge spectra of the same alloy are different (see Figs. 3 and 4) indicates that the contributions to the MCXD signal of  $\text{Fe}^*\text{-Fe}$  and  $\text{Co}^*\text{-Fe}$  pairs are not equivalent. In order to confirm this model, additional multiple-scattering calculations on this specific system should be performed.

Sakurai *et al.*,<sup>22</sup> in their investigation of Ni-Fe alloys, suggest that the amplitude of the MCXD signal close to the edge should be correlated to the magnetic moment of the absorbing atom. The integrated intensities of the measured MCXD peaks, and the magnetic moments on Fe and Co as a function of Co concentration in Fe-Co alloys, are summarized in Fig. 6. The integrated intensity  $A2$  measured at the Fe  $K$  edge increases with concentration but, unlike the magnetic moment, does not saturate for more than 50% Co. The maximum amplitude of peak  $A2$  ( $A2^*$ ) follows the magnetic moment behavior more closely since it saturates for more than 50% Co. For Co, small changes of the magnetic moment provoke very important changes of the Co  $K$ -edge intensity. The Co  $K$ -edge signal is very sensitive to the change in local

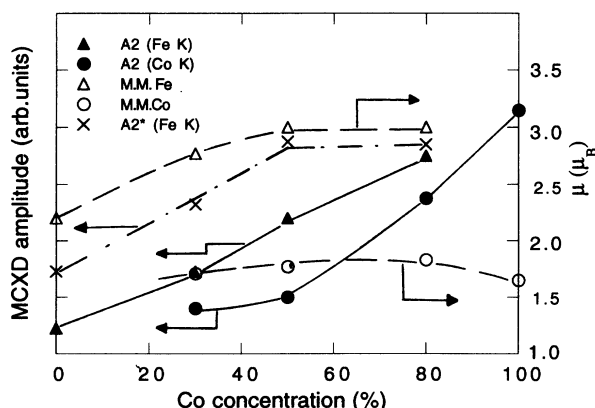


FIG. 6. The integrated intensities of the negative peaks  $A2$  of the MCXD spectra measured for bcc Fe, hcp Co, and Fe-Co alloys (Table II) are compared with local magnetic moments (M.M.) on the Fe and Co sites.  $A2$  are the integrated intensities,  $A2^*$  the maximum amplitudes. The value reported for pure bcc Co is the average signal obtained for the bcc Co films in the Fe/Co multilayers. The dotted lines are guides for the eye.

environment. As the Co concentration increases, the MCXD signal increases, and a discontinuity occurs for 50% Co. For the  $\text{Fe}_{70}\text{Co}_{30}$  alloy a weak positive peak appears at the energy position, characteristic of the positive peak of pure Fe.

## 2. Fe/Co multilayers

The negative peak  $A2$  measured at the Fe  $K$  edge for Fe/Co multilayers is enhanced with respect to Fe metal (Table III and Fig. 4), as already found for Fe-Co alloys. The enhancement of peak  $A2$  is more important for the thinnest Fe films (11 Å), and this indicates that the Fe signal is larger for atoms closer to the Fe/Co interface. The results obtained for Fe-Co alloys suggest that the enhancement of the Fe  $K$ -edge signal may be correlated with an increased magnetization of Fe in these thin films. The similar amplitudes measured for  $A2$  in the  $\text{Fe}_{11}\text{ÅCo}_{20}\text{Å}$  and in the  $\text{Fe}_{20}\text{Co}_{80}$  alloy (Tables II and III) suggest that the magnetic moment of Fe may be enhanced to a value typical of saturated Fe, i.e.,  $3\mu_B$ , over the whole Fe layer. This is in agreement with previous magnetization measurements for these multilayers, which show that the extension of the magnetic interface, where the magnetization is enhanced with respect to Fe, is about 6 Å.<sup>14</sup> The smaller amplitude measured for peak  $A2$  in the thicker Fe layers is consistent with the fact that far away from the interface the magnetic moment of Fe remains close to the bulk value.

The amplitude of the positive peak  $A1$  for the Fe layers is very similar to that of bulk Fe. The comparison between the Fe  $K$ -edge spectra of the  $\text{Fe}_{20}\text{Co}_{80}$  alloy and the  $\text{Fe}_{11}\text{ÅCo}_{20}\text{Å}$  multilayer is shown in Fig. 5. While the two  $A2$  peaks have the same amplitude, the positive peak typical of bcc Fe is observed only for the multilayer. If we admit that the amplitude of the negative peak is characteristic of saturated Fe, we realize that the presence of a positive peak in the  $K$ -edge signal of the multilayers is in disagreement with an interpretation that attributes this peak to a partially filled majority band. These results seem to reinforce the second model proposed for the interpretation of  $K$ -edge spectra, and suggests that the presence of a positive peak depends on the presence of Fe atoms in the neighborhood of the absorbing Fe atom. In the Fe/Co multilayers studied here, Fe atoms have mainly Fe neighbors [ $\approx 90\%$  Fe for the  $(110)\text{Fe}_{11}\text{ÅCo}_{20}\text{Å}$ ] and this would explain the large amplitude of the positive peak.

The Co  $K$ -edge MCXD signal measured for the Fe/Co multilayers, where the local Co structure is bcc, consists of a single negative peak. Its amplitude varies little with Co thickness, and it is sensibly enhanced with respect to the maximum signal observed for the Fe-Co alloys. The average signal measured for the multilayers could be taken as representative of bcc Co surrounded entirely by Co neighbors, as is the case for these relatively thick Co layers. These results strongly indicate that the decrease of the intensity of the negative contribution at the Co  $K$  edge in Fe-Co alloys is due to the change of the local environment of Co.

The results of both Fe and Co  $K$ -edge spectra for the

Fe/Co multilayers seem to rule out the presence of important interfacial interdiffusion.

### V. MAGNETIC EXAFS SPECTRA

The high-energy MCXD spectra of bcc Fe, hcp Co, and the most representative Fe-Co alloys and multilayers are represented in Fig. 7. All spectra exhibit very intense oscillations similar to EXAFS in the total absorption. The Fe and Co *K*-edge MCXD spectra measured for bcc alloys and bcc multilayer samples have features similar to those found in the spectrum of pure bcc Fe. The Co *K*-edge spectrum of bulk hcp Co is clearly different. These results confirm the close dependence of the high-energy spectra on the local magnetic structure.

The comparison between the fine structure in the total absorption and in the difference (MCXD) spectra can be seen in Fig. 8 for bcc Fe, Fe<sub>70</sub>Co<sub>30</sub> (Fe and Co *K* edges), and hcp Co (Co *K* edge). Close to the absorption edge the two sets of spectra are clearly different, and the magnetic spectra exhibit a much more complex structure. For higher energies magnetic and nonmagnetic EXAFS oscillations have the same features and are in phase with each other. Important differences between the two sets of spectra occur up to about 80 eV beyond the absorption edge. In particular, a very strong feature, absent in the total absorption spectra, appears at around 60–70 eV in *all* MCXD spectra. The structure is more intense for bcc Fe and bcc alloys than for Co metal. We suggest that this structure could correspond to the presence of multi-electron excitations involving  $3p \rightarrow 4p$  transitions ( $|1s^2 \dots 3p^6 \dots 4p^0\rangle \rightarrow |1s^1 \dots 3p^5 \dots 4p^2\rangle$ ).<sup>47</sup> The binding energies of  $3p$  electrons for Fe and Co are indeed around 60 eV.

Figures 7 and 8 show that for high energies ( $E - E_0 > 80$  eV) the Co and Fe *K*-edge spectra for the

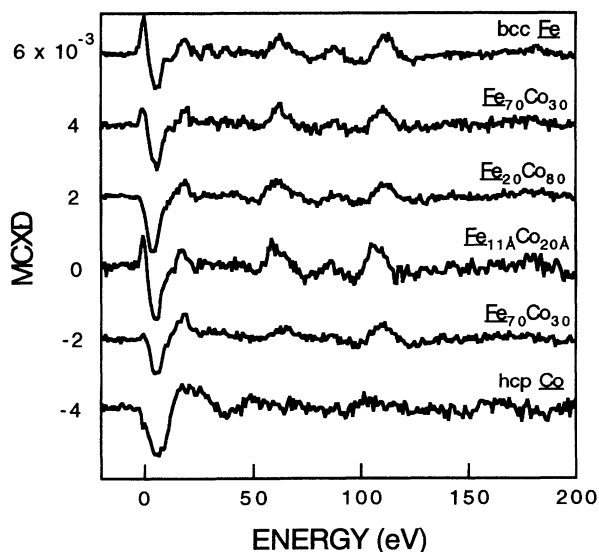


FIG. 7. High-energy MCXD spectra (magnetic EXAFS) of Fe, hcp Co, Fe<sub>70</sub>Co<sub>30</sub> (Fe *K* and Co *K* edge), and Fe<sub>20</sub>Co<sub>80</sub> alloys, and the Fe<sub>11</sub>Å Co<sub>20</sub>Å multilayer. Fe: Fe *K* edge. Co: Co *K* edge.

Fe<sub>70</sub>Co<sub>30</sub> alloy are practically identical. This suggests that the high-energy MCXD spectra are independent of the absorbing atom and essentially determined by the local environment, which is the same for Co and Fe in these disordered alloys.

Multiple-scattering calculations<sup>39</sup> show that for high energies the MCXD spectrum is dominated by the magnetic polarization of the *p* bands of the absorbing atom and those of the neighbors. Since these bands have an important spatial extension, it is not unreasonable that the MCXD signal may be sensitive to magnetization in the neighborhood of the absorbing atom.

The intensities of the high-energy Fe *K*-edge magnetic EXAFS signal for bcc Fe and Fe-Co alloys, Fe<sub>11</sub>Å Co<sub>20</sub>Å and Fe<sub>21</sub>Å Co<sub>30</sub>Å multilayers are shown in Fig. 9. The intensities were obtained by integrating the spectra between  $\approx 103$  and  $\approx 116$  eV (the strongest and better defined magnetic peak) and dividing these values by the amplitude of the corresponding peak in the total absorption spectrum. In the same figure we have reported, for

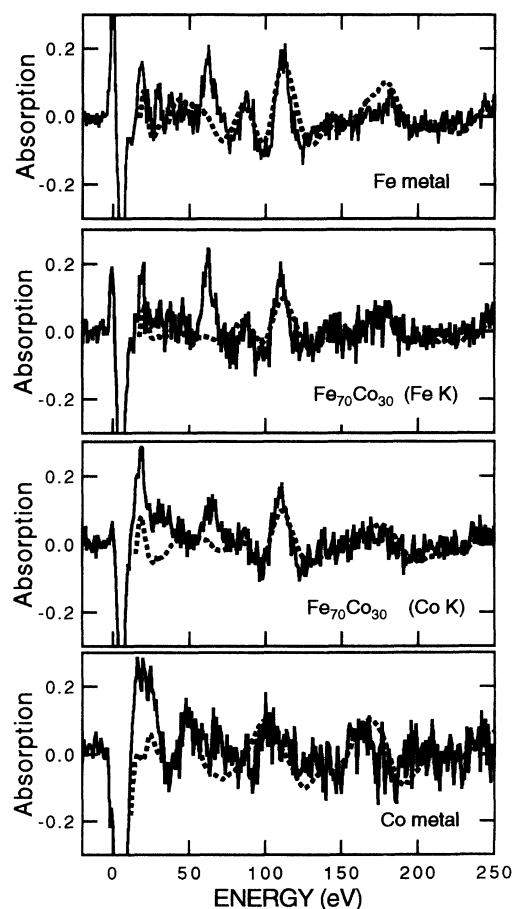


FIG. 8. The magnetic EXAFS spectra for Fe metal (Fe *K*), Fe<sub>70</sub>Co<sub>30</sub> alloy (Fe *K* edge and Co *K* edge), and hcp Co metal (Co *K* edge), multiplied by a factor 400 (full lines) are compared with the EXAFS oscillations in the total absorption (dotted lines). Note the good correspondence between the two spectra above about 80 eV. The intense structure at about 60 eV may be due to multi-electron excitations.



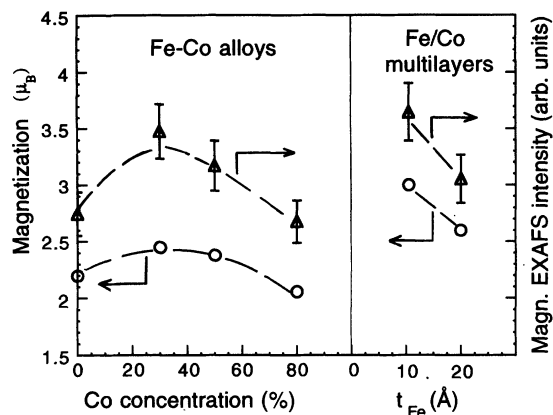


FIG. 9. The normalized intensity of the magnetic EXAFS oscillations (triangles) in the Fe  $K$  edge spectra of Fe, Fe<sub>70</sub>Co<sub>30</sub>, Fe<sub>50</sub>Co<sub>50</sub>, and Fe<sub>20</sub>Co<sub>80</sub> alloys, and in the two Fe<sub>11</sub>  $\text{\AA}$ Co<sub>20</sub>  $\text{\AA}$  and Fe<sub>21</sub>  $\text{\AA}$ Co<sub>30</sub>  $\text{\AA}$  multilayers are compared with the macroscopic magnetization of the same samples (circles). The dotted lines are guides for the eye.

the same samples, the average magnetization around Fe. For Fe-Co alloys we have assumed that this is equivalent to the macroscopic magnetization. For the two multilayers we have assumed, as indicated by magnetic measurements, that the magnetic moment of Fe is  $2.2\mu_B$  away from the interface and  $3\mu_B$  at the interface, over a total thickness of 12  $\text{\AA}$ .

The figure shows that moving from Fe to Fe-Co alloys with increasing Co concentration, the normalized magnetic EXAFS intensity first increases, reaches a maximum for Fe<sub>70</sub>Co<sub>30</sub>, and then decreases again. This behavior is very similar to the well-known trend for the macroscopic magnetization of Fe-Co alloys. Although the variations of the magnetic signals for the intermediate alloys appear to be more important than the variations of the magnetization, in both curves a maximum is reached for 30% Co. For the two Fe/Co multilayers, both the magnetic signal and the average magnetization are enhanced with respect to bulk Fe, and decrease as the Fe thickness increases.

In summary, these results clearly show that the normalized magnetic EXAFS intensity seems to follow the general trends of the local average magnetization. The ratio between the EXAFS signal and the macroscopic magnetization is not constant within the series of samples studied here, and in order to fully understand their relationship a systematic study of a larger number of alloys should be carried out.

## VI. CONCLUSION

In this paper we have discussed the specific features of Fe and Co  $K$ -edge MCXD signals in bcc structures. The

main differences between near-edge MCXD spectra of alloys and multilayers can be explained in terms of the difference of the local environment of Fe and Co in the two systems.

Our discussion of near-edge MCXD spectra for Fe-Co alloys and Fe/Co multilayers is based on the observed correlation between the amplitudes of positive peak  $A1$  and negative peak  $A2$ , and the change of magnetic character of the Fe and Co atoms. The general trends observed for the experimental MCXD spectra indicate that at the Fe  $K$  edge the amplitudes of  $A2$  might be related to the magnetic moment of the absorbing atom. The presence of a positive peak at the Fe  $K$  edge does not appear to be related only to the magnetic weakness of the absorbing atom. A positive peak has in fact been observed both for saturated Fe and Co in Fe-Co alloys and for Fe in Fe/Co multilayers. The MCXD Fe  $K$ -edge spectra can be visualized as combinations of contributions due to Fe\*-Fe and Fe\*Co pairs, giving double-peaked and single peaked structures, respectively. Although the reasons for this difference are not fully understood, they are likely to be related to the dominant spin density close to the Fermi level. The differences between the Co and Fe  $K$ -edge spectra for the same alloy indicate that the signal is strongly dependent on the chemical nature of the absorbing atom.

Extending the idea developed by Wang *et al.*<sup>33</sup> for  $L_{2,3}$  edges of rare earths, it is possible that the  $4p$ - $3d$  exchange may play a role in tuning the amplitude of the positive peak  $A1$  in  $K$ -edge spectra. If the spin-up  $4p$  wave functions are "pulled in" relative to the spin-down, as may be the case for Fe, an amplification of the positive contribution close to the Fermi energy may occur. An opposite effect may be expected for Co. To pursue this idea for the interpretation of  $K$ -edge spectra, further theoretical developments are necessary.

We have shown that for high energies (more than about 80 eV beyond the absorption edge) the MCXD signal depends essentially on the local environment around the absorbing atom. The close similarity between Fe and Co  $K$ -edge spectra of the Fe<sub>70</sub>Co<sub>30</sub> alloys shows that in these systems the spectra do not depend strongly on the nature of the absorbing atom. For alloys and multilayers, the normalized magnetic EXAFS intensity can be correlated with the average magnetization in the neighborhood of the absorbing atom.

## ACKNOWLEDGMENTS

The authors would like to acknowledge G. Schmerber for the preparation of the Fe-Co alloys. We would like to thank Ch. Brouder for many helpful discussions. P. Beauvillain, P. Veillet, and C. Chappert have kindly helped with the magnetic measurements on Fe/Co multilayers and their interpretation.

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