

## Interfaces of Ce/Fe and La/Fe multilayers probed by magnetic circular x-ray dichroism

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An experimental study of conventional x-ray absorption and of magnetic circular x-ray dichroism has been performed on La/Fe and on Ce/Fe multilayers at the  $L_2$  and  $L_3$  edges of La and Ce and at the  $K$  edge of Fe to characterize the impact of the interaction of La or Ce and Fe on the local electronic and magnetic structure of the interfaces. The spectra reveal that the interaction is particularly unusual for Ce/Fe, where two effects have been observed. First, on a depth scale of at least 15 Å near the interface, the Ce atoms adopt the electronic structure of the  $\alpha$  phase, with itinerant  $4f$  states. Second, these  $\alpha$ -phase-like Ce atoms are magnetically polarized and carry an ordered magnetic  $5d$  spin moment. This moment is antiferromagnetically coupled to the Fe  $3d$  moment. Interface mixing on a length scale of the magnetic polarization can be excluded. For the La/Fe system, a similar magnetic polarization is effective, but in contrast to Ce/Fe it is restricted to the immediate interface with Fe. The results are discussed with reference to recent measurements of the macroscopic magnetization and Mössbauer spectra. Comparison with intermetallic compounds of rare-earth and late transition-metal elements points to the importance of hybridization effects for the Fe and Ce or La electronic states near the interfaces in the multilayers.

### I. INTRODUCTION

In cerium metal the electronic  $4f$  states are at the borderline between localized and itinerant behavior. This is reflected in the remarkable transition from the fcc  $\gamma$  phase with localized  $4f$  configuration into the isostructural  $\alpha$  phase with delocalized  $4f$  configuration. The transition may be driven thermally, by the application of pressure or by alloying and formation of compounds with  $d$ -band transition metals, and is intimately related to a varying degree of mixing of the  $4f$  and conduction-electron states, i.e., to the formation of a highly correlated electronic structure. This interaction has fascinating consequences, as is apparent for instance in the particularly unusual properties of the ferromagnet  $\text{CeFe}_2$ . In this compound, which is a prototype of a mixed-valent system, the Ce  $4f$  states are strongly hybridized with the Fe  $3d$  states<sup>1</sup> and hence delocalized, which leads to a  $4f$ -occupancy number well below unity ( $n_f \approx 0.7$ ). The result is an anomalously low lattice parameter and Curie temperature ( $T_c = 230$  K) compared to the corresponding homologous compounds of the other rare earths, a small magnetic moment of the Fe ions ( $\mu_{\text{Fe}} \approx 1.4\mu_B$ ), and a magnetic moment of the Ce ions ( $\mu_{\text{Ce}} \approx 0.7\mu_B$ ) coupled antiferromagnetically to the Fe moment. Comparison with the "non- $4f$ " intermetallic  $\text{LuFe}_2$  ( $T_c = 610$  K,  $\mu_{\text{Fe}} \approx 1.7\mu_B$ , La-Fe compounds do not exist) clearly reveals that the anomalous properties of  $\text{CeFe}_2$  originate from the  $4f$  contribution to the conduction band.<sup>2</sup>

In view of these properties, the recent developments in the synthesis of layered structures offer an attractive opportunity to probe the interaction between the electronic

states of a rare earth (RE) and a transition metal (TM) at the interfaces of artificially grown multilayers of these elements. The present paper deals with such heterostructures of Ce and Fe, and of La and Fe as a non- $4f$  reference system. These systems are well suited for the study of interface phenomena since they can be prepared with sharp interfaces<sup>3,4</sup> and have been thoroughly characterized previously by different techniques.<sup>3-6</sup> The saturation magnetization of these multilayers when referred to Fe is reduced below the bulk Fe value. This points to strong interaction effects between the Ce or La and Fe electronic states. The larger reduction for Ce/Fe reveals the involvement of the Ce  $4f$  states in this interaction. It is the goal of this investigation to provide a microscopic characterization of the local electronic and magnetic structure at the Ce-Fe and La-Fe interfaces. This information is derived by absorption spectroscopy at the  $L_2$  and  $L_3$  edges of Ce and La and at the  $K$  edge of Fe with circularly polarized hard x rays, taking benefit from the element and site selectivity of the method and the appearance of magnetic dichroism near the absorption edges involved. The most important results are that (i) Ce adopts, on a considerable length scale near the interface, the electronic structure of the  $\alpha$  phase, and (ii) even at room temperature a considerable portion of these  $\alpha$ -phase-like Ce atoms carries an ordered magnetic moment on their  $5d$  states which is in antiparallel configuration with the Fe moment. A part of the La atoms is equally polarized magnetically, but surprisingly the polarization is confined to the immediate interface. The effects observed present evidence of a significant hybridization of the Ce or La  $5d$  and the Fe  $3d$  states near the interfaces in the multilayers.

## II. MAGNETIC X-RAY ABSORPTION

Conventional x-ray-absorption spectroscopy at the  $L_{2,3}$  edges of rare-earth elements have been widely used to study their electronic states in intermetallic compounds. The method is of particular importance for Ce systems which are exceptional among the lanthanides with respect to their unusual physical properties,<sup>7</sup> the aforementioned CeFe<sub>2</sub> being a prominent example. As the inner-shell photoabsorption process involves the creation of a core hole, the interpretation of the measured spectra is not straightforward because of the presence of final-state effects. But it has been shown,<sup>8,9</sup> invoking the model of Gunnarsson and Schönhammer,<sup>10</sup> that the  $L_{2,3}$  edges of Ce yield a reliable image of its ground-state configuration if the relevant Coulomb interactions between the photoelectron and the remaining electronic system are properly taken into account. In particular, the degree of delocalization of the Ce 4*f* states, which varies with the local structural and chemical environment in the different systems, is sensitively reflected in the  $L_{2,3}$  absorption-edge profiles.

In the present context it is important that x-ray-absorption spectroscopy provides access to the magnetic properties of solids when performed with circularly polarized x rays. In this case electrons excited from a core level [here  $2p_{1/2}$  and  $2p_{3/2}$  for the  $L_2$  and  $L_3$  edges of a rare earth (RE),  $1s$  for the  $K$  edge of a transition metal (TM)] are spin polarized.<sup>11</sup> The transitions of such electrons into exchange-split unoccupied states at the same site then allow the study of local magnetic properties of the absorbing atomic species in a ferromagnetic or ferrimagnetic material, because the photoabsorption cross section depends on the helicity of the incident light (left or right circularly polarized) with respect to the direction of the magnetization. This magnetic circular x-ray dichroism (MCXD) is governed by dipole transitions driven by the electric field of the radiation. Since the electric dipole operator does not act on the electron spin, spin selectivity of the absorption arises via spin-orbit coupling in the final state, either of the excited photoelectron or of the core hole.<sup>12</sup> Studies of MCXD in the hard x-ray range (5–10 keV) have addressed RE/TM systems at the  $L_{2,3}$  and  $K$  edges of the RE and TM, respectively. These experiments have established by now magnetic x-ray absorption as a powerful spectroscopy for probing element- and site-specific local magnetism in solids. For example, the MCXD studies performed on multilayers of Nd/Fe,<sup>13</sup> Tb/Fe,<sup>14</sup> Co/Pt,<sup>15,16</sup> Co/Pd,<sup>17</sup> and Co/Cu,<sup>18</sup> or on ultrathin Fe layers,<sup>19</sup> have illustrated the importance of the method as a tool for getting access to interfacial magnetism, magnetic polarization or magnetic anisotropy. Recent MCXD studies<sup>20,21</sup> performed at the  $L_{2,3}$  edges of Ce in several different intermetallic compounds with TM elements, which probe the exchange-polarized  $5d$  conduction-band states, have elucidated the key role of the nature of the 4*f* states, which are coupled to the  $5d$  states by local exchange, in the absorption process: the spectra are very different for  $\gamma$ -phase-like Ce compounds (localized 4*f* states) and  $\alpha$ -phase-like Ce compounds (delocalized 4*f* states). There is evidence<sup>21</sup> that only in

the former ones the 4*f* shell contributes to the  $L_{2,3}$  MCXD spectra. Obviously these results are important in the context of the present study.

In spite of large efforts the present level of understanding of the MCXD process is rather limited. In the one-electron approach,<sup>22</sup> at the  $L_{2,3}$  edges of a RE and the  $K$  edge of a  $3d$  TM, only the  $2p$  to  $5d$  and the  $1s$  to  $4p$  transitions permitted by the dipole selection rules contribute to the absorption process. The significance of many-body effects and the core-hole-induced relaxation await to be elucidated in a more rigorous treatment. Furthermore, the question concerning the contribution of quadrupolar transitions<sup>23</sup> to the magnetic cross section remains to be answered. In the case of RE-TM compounds, the systematics of MCXD revealed by extensive experimental data has provided an important insight into the local magnetic structure and the coupling between the magnetic moments<sup>20,21</sup> although a thorough theoretical treatment is missing. For the RE  $L_{2,3}$  edges and the  $3d$  TM  $K$  edge the dichroic signal essentially results from the spin polarization of the unoccupied  $5d$  and  $4p$  band states, respectively, reached in the absorption process. Since the conduction electrons play a key role in the interaction between the RE and the TM which tunes the magnetic properties,<sup>2</sup> the information about  $5d$  magnetism in such systems, in particular, is of fundamental interest. But due to the spin coupling of the RE  $5d$  and  $4f$  states and the strong hybridization between the  $5d$  and TM  $3d$  states the interpretation of the MCXD spectra is generally a complicated task. Jo and Imada<sup>24</sup> recently succeeded to interpret the observed systematic trend of MCXD in the  $L_{2,3}$  photoabsorption of trivalent RE elements (localized 4*f* states) in compounds (RE)<sub>2</sub>Co<sub>17</sub>. Their calculation is based on an one-electron model and invokes the  $5d$  spin and orbital moments induced through the intraatomic 4*f*- $5d$  exchange interaction. Quite generally, the relation between the dichroic signal and the total magnetic moment of the absorbing atom in TM or RE-TM systems is a matter of a current debate.<sup>17,20,21,25</sup> In general the integrated signals at the individual edges are not proportional to this moment. But remarkably, it has been established<sup>20,21</sup> for a certain class of RE-TM compounds, that the  $L_{2,3}$  MCXD intensity on the RE and the  $5d$  magnetic moment are simply proportional provided the 4*f* states are itinerant and hence their orbital moment is quenched. The ferromagnet CeFe<sub>2</sub> is a prominent example. For RE systems with localized 4*f* states this simple correlation breaks down.

## III. EXPERIMENTAL DETAILS AND SAMPLE CHARACTERISTICS

The Ce/Fe and La/Fe multilayers were grown by ion-beam sputtering using Ar gas in an UHV chamber (base pressure below  $5 \times 10^{-10}$  mbar). Partial pressures of residual gases (e.g. O<sub>2</sub>, N<sub>2</sub>, H<sub>2</sub>O) were below  $10^{-10}$  mbar during deposition. For the present experiments, Kapton foil coated with a 40-Å-thick Cr buffer layer was used as substrate, while most of the previous structural and magnetic measurements were performed on samples deposited on equally precoated single-crystalline Si(100). Care-

ful comparative studies testify that, because of the same buffer layer, the properties do not depend on the substrate. The substrate holder was cooled to liquid-nitrogen temperature to minimize diffusion. The samples were protected from oxidation by a 60-Å-thick Cr top layer. The deposition process was controlled by a quartz rate monitor and a computer. Deposition rates were typically 0.5 Å/s, the total thickness of the multilayers was near 4000 Å.

The multilayers had been well characterized previously.<sup>3-6</sup> For the samples studied by x-ray-absorption (individual layer thicknesses  $t_{\text{Fe}} > 25$  Å,  $10$  Å  $\leq t_{\text{Ce,La}} \leq 60$  Å), Fe is crystalline with bcc structure, Ce is amorphous, La is crystalline with fcc structure above 20 Å and amorphous below. In Ce/Fe, the Fe layers are strongly textured, with their densest plane (110) in the film plane, while the crystallites are randomly oriented in La/Fe. In both cases, the Fe lattice is expanded in the growth direction of the films by about 1%. The lateral extension of the crystallites is between 80 and 100 Å. Special efforts were devoted to the structural characterization of the interfaces. Small-angle x-ray diffraction and subsequent Monte Carlo simulations of the diagrams<sup>3,4</sup> revealed that diffusion is negligible, and local fluctuations of the individual layer thicknesses, representing "roughness" at the interfaces, are limited to an average value of  $\pm 0.25$  atomic layers in Ce/Fe and of  $\pm 0.1$  atomic layers in La/Fe. Hence, the interfaces are somewhat rougher in Ce/Fe, but the actual numbers found permit to conclude that in both multilayer systems they are rather sharp. In spite of the large atomic-size mismatch between Fe and Ce (26%,  $\alpha$  phase) or La (32%) and the amorphous structure of Ce or La (for  $t_{\text{La}} < 20$  Å only) which prevent coherence, disorder at the interfaces appears to be rather limited. Cross section transmission electron microscopy images of the Ce/Fe multilayers taken recently<sup>26</sup> clearly show that the periodic array of the component films is very regular and that the interfaces are flat and parallel. Finally, it is interesting to note in the present context that a recent study of the growth mode of Ce on Fe (100) has shown that interdiffusion does not occur even for deposition at room temperature.<sup>27</sup>

Complementary information about the interfaces may be derived from <sup>57</sup>Fe Mössbauer spectra of the multilayers. These results are only briefly addressed here, a more detailed discussion can be found in Refs. 4 and 6. The spectra were decomposed into a sextet component representing the normal bcc part in the core of the Fe layers and a residual component representing the interface part. The latter subspectrum was analyzed then in terms of a probability distribution of magnetic hyperfine fields. The contribution of the interface component to the total intensity of the spectrum corresponds to a nominal Fe-layer thickness of about 9 Å per interface both in Ce/Fe and in La/Fe. This signifies that the formation of the interface affects the same length scale in the Fe layers of the two multilayer systems. The value exceeds the structural extension of the interfaces resulting from local thickness fluctuations, as derived from small-angle x-ray diffraction, and points to a magnetic "proximity effect" which must be connected to a modification of the elec-

tronic structure in the interfacial zone induced by the Ce-Fe and La-Fe intercalation. This modification of the electronic interaction is different in the Ce/Fe and La/Fe multilayers, which is reflected in different distributions of magnetic hyperfine fields underlying the residual subspectra. We only mention that for Ce/Fe, there is a well-defined maximum at 45 kOe (at 300 K) which is absent in La/Fe. The maximum in Ce/Fe may be attributed to an "interface alloy" which is weakly ferromagnetic at room temperature and involves nominally about 2.5 Å of Fe, corresponding to one monolayer. This is on the length scale of the local thickness fluctuations determined by x-ray diffraction. In contrast, the corresponding distribution of hyperfine fields in La/Fe shows only a broad feature with small probability amplitudes at low fields. This difference must be connected to the distinctly smaller degree of interfacial roughness in this multilayer system.

The interpretation of the small-angle x-ray diagrams and of the Mössbauer spectra is based on the assumption of structurally and electronically symmetric interfaces in both systems. This may be oversimplified since the alternate stacking of the amorphous and crystalline layers investigated here might well result in asymmetric interfaces. But at present there is no experimental evidence for such a structure.

The average magnetic hyperfine field  $\langle H_{\text{hf}} \rangle$  resulting from the total <sup>57</sup>Fe Mössbauer spectra is considerably smaller in the Ce/Fe multilayers as compared to La/Fe, which presents additional evidence for a stronger RE-Fe interaction in the former system. For the multilayer Ce(30Å)/Fe(29 Å), for example, we have  $\langle H_{\text{hf}} \rangle = 260$  kOe at room temperature, which compares to  $\langle H_{\text{hf}} \rangle = 307$  kOe for (30 Å)La/(29 Å)Fe (the bulk bcc  $\alpha$ -Fe value is 334 kOe at 300 K). Bearing in mind the strong electronic interaction at the interfaces, it is clear that the experimental hyperfine fields of these layered samples cannot be related in a simple way to their macroscopic magnetization or to the Fe part of this quantity. Figure 1 presents the macroscopic spontaneous magnetization  $M_s$  referred to Fe at room temperature of several La/Fe and Ce/Fe multilayers with a constant Fe-layer thickness of 29 Å and varying thicknesses of the La and Ce layers. These magnetization measurements were performed between 4.2 and 300 K, in magnetic fields up to 50 kOe applied in the film planes, using a vibrating-sample magnetometer.  $M_s$  is reduced below the value of bulk iron, about 8% for La/Fe and 30% for Ce/Fe. This compares to a reduction of the average hyperfine fields at the Fe sites of 8% in La/Fe and of 22% in Ce/Fe. Qualitatively, this is an indication that the macroscopic magnetization mainly reflects the contribution of the Fe layers, which obviously is of different magnitude in the two multilayer systems. The magnetization varies with temperature according to  $M_s(T)/M_s(0) = (1 - bT^{3/2})$  where  $b$  is smaller for La/Fe than for Ce/Fe (for example,  $b = 1.4 \times 10^{-5} \text{ K}^{-3/2}$  for the 30-Å La and  $b = 3.0 \times 10^{-5} \text{ K}^{-3/2}$  for the 30-Å Ce layer combined with 29 Å of Fe).<sup>4</sup> This indicates that the Curie temperatures are far above room temperature in both cases, but that the absolute values must be considerably lower for the Ce/Fe multi-

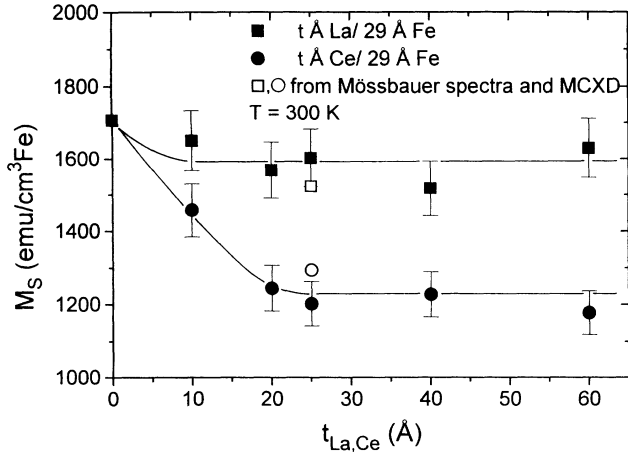


FIG. 1. Total spontaneous magnetization of La/Fe and Ce/Fe multilayers (Fe-layer thickness 29 Å, total thickness  $\sim 4000$  Å) at 300 K as a function of the La- and Ce-layer thickness. The open symbols at  $t_{\text{La,Ce}} = 25$  Å represent the values estimated from the MCXD and Mössbauer spectra (see text).

layers. (With the usual relation between the spin-wave parameter  $b$  and the exchange constant  $J$ ,  $b \sim J^{-3/2}$ , and the mean-field relation  $T_C \sim J$  we have  $T_{C,\text{La}}/T_{C,\text{Ce}} = 1.7$ ).

The x-ray-absorption spectra were measured in transmission mode at room temperature mainly, on the energy-dispersive spectrometer of the DCI storage ring at LURE (Orsay), using a position-sensitive detector. The appropriate sample thickness for the experiments was achieved by stacking 4–5 kapton foils carrying the multilayers. The MCXD spectra were recorded at 0.3 mrad below the positron orbit plane where the light is right circularly polarized to 80%, for two opposite directions of an external magnetic field  $B_+$  and  $B_-$  along the wave vector of the light. It is applied to suppress magnetic domains and to reverse the magnetization, it is directed at grazing incidence to the multilayer planes and amounts to 5 kOe, which is strong enough to saturate the magnetization (the samples show in-plane anisotropy). Details of data collection and the experimental setup are described elsewhere.<sup>13,28</sup>

Special attention was paid to possible oxygen contaminations of Ce and La. Separate measurements on thick crystalline Ce and La films prepared in the same way as the other samples testify that RE-oxide-related near-edge structures in the  $L_{2,3}$  spectra were absent. These films were used for the calibration of the energy scale near the absorption edges.

The MCXD spectra are presented by the difference  $\mu^-(B_+) - \mu^-(B_-)$  where  $\mu^-(B_+)$  and  $\mu^-(B_-)$  are the absorption coefficients for right-circularly polarized x rays (helicity  $-\hbar$ ) and the two field directions  $B_+$  parallel and  $B_-$  antiparallel to the wave vector of the light. Let us mention that in the electric dipole approximation the change of the field directions (hence of the magnetization) is equivalent to the change of the helicity of the photons. The absorption coefficients  $\mu^-(B_{+,-})$  are normalized to one far beyond the edge. This procedure makes the spectra independent of the number of atoms

probed in a measurement. The origin of the energy scale is chosen at the inflection point of the absorption-edge profile and taken as the position of the Fermi level.

#### IV. RESULTS AND DISCUSSION

In Fig. 2, the conventional x-ray-absorption spectra at the Fe  $K$  edge of two representative Ce/Fe and La/Fe multilayers, of Fe metal and of the compound  $\text{CeFe}_2$  are compared. Obviously, all features of the bcc-Fe curve are reproduced for the multilayers. However, in the case of  $\text{CeFe}_2$ , the structure of the spectrum is noticeably different. This is an indication of the different environment of the Fe atoms in the compound. As can be seen in Fig. 3, the MCXD signal at the Fe  $K$  edge of the multilayers is a close image of the corresponding signal of Fe metal, as for the total absorption spectrum: the most prominent features are a maximum at 0 eV followed by a minimum at about 5 eV. Unfortunately, although this dichroic signal is the result of a transition into the spin-polarized  $4p$  final states, its correlation to local magnetism is not clear.<sup>22</sup> A recent MCXD study<sup>25</sup> at the  $K$  edge of several  $3d$  elements in various alloys and compounds has demonstrated that there is no proportionality of the dichroic absorption signal to the local  $p$  or  $d$  magnetic moment on Fe. There is an indication in Fig. 3 that for the Ce/Fe multilayer the signal is somewhat reduced with respect to Fe metal, as might be expected from the observation of a significantly reduced average magnetic hyperfine field, as we have indicated above. But the signals are very small (about one order of magnitude below the MCXD amplitude at the  $L_{2,3}$  edges of Ce and La, see below), and in view of the appreciable noise a reliable value of the amplitudes cannot be determined.

The experimental results of x-ray absorption in the multilayers at the  $L_2$  and  $L_3$  edges of Ce and La are displayed in Figs. 4–7. The most interesting feature is the observation of MCXD for both metals in the artificial heterostructure even at room temperature (Figs. 5–7), where in bulk form even Ce is paramagnetic. In addition,

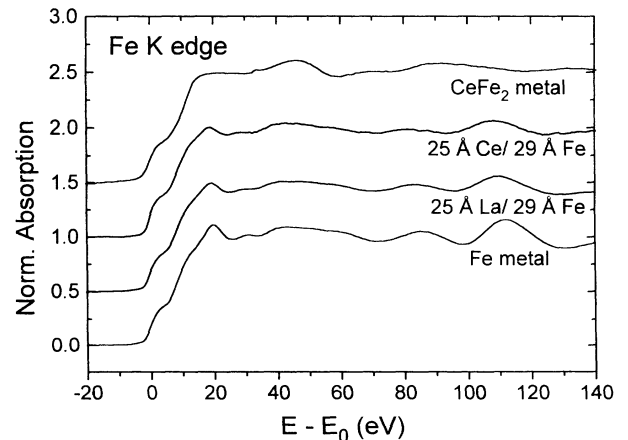


FIG. 2. Normalized x-ray-absorption spectra at the Fe- $K$  edge of a La/Fe and a Ce/Fe multilayer (thickness  $\sim 4000$  Å), of bcc Fe metal and the intermetallic compound  $\text{CeFe}_2$ . The curves have been displaced vertically.  $E_0 = 7111$  eV.

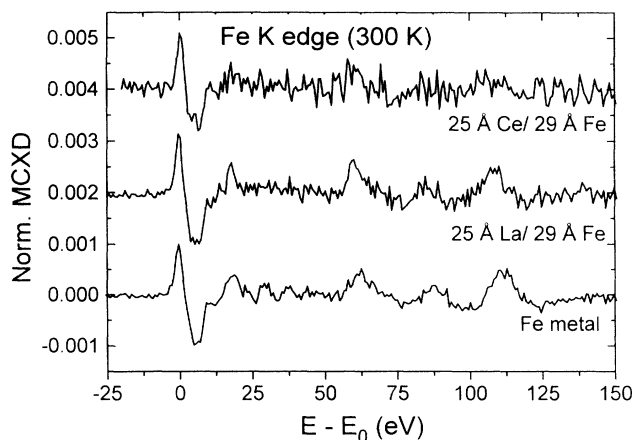


FIG. 3. Normalized MCXD spectra of the Fe-K edge of a La/Fe and a Ce/Fe multilayer (thickness  $\sim 4000$  Å), and of bcc Fe metal in an external magnetic saturation field (5 kOe) at room temperature. The curves have been displaced vertically.

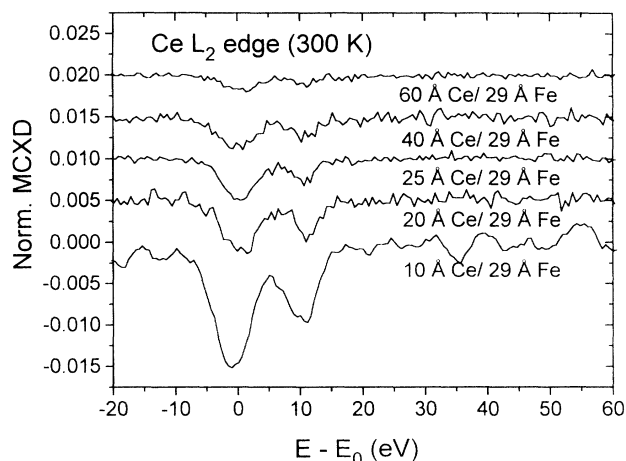


FIG. 5. Normalized MCXD spectra at the Ce- $L_2$  edge of Ce/Fe multilayers (Fe-layer thickness 29 Å, total thickness  $\sim 4000$  Å) in an external magnetic saturation field (5 kOe) at room temperature. The curves have been displaced vertically.

the near-edge structure in the absorption spectra of the Ce/Fe multilayers is remarkable. This is immediately apparent in Fig. 4, which shows the results obtained at the  $L_2$  edge for five Ce/Fe multilayers with constant thickness of the individual Fe layers ( $t_{\text{Fe}} = 29$  Å), as compared to Ce metal, the intermetallic compound  $\text{CeFe}_2$ , and a La/Fe multilayer with 25-Å-thick La layers. With decreasing thickness of the Ce layers, the absorption profile changes noticeably. This reflects a progressive modification in the Ce 4*f* configuration. In contradiction to the  $L_2$  spectrum of Ce metal which shows the “white-line” profile of the  $\gamma$  phase with its well-localized 4*f* states, those of the Ce/Fe multilayers exhibit a second peak located at  $\sim 10$  eV above the first one and gradually approach, with decreasing Ce-layer thickness, the spec-

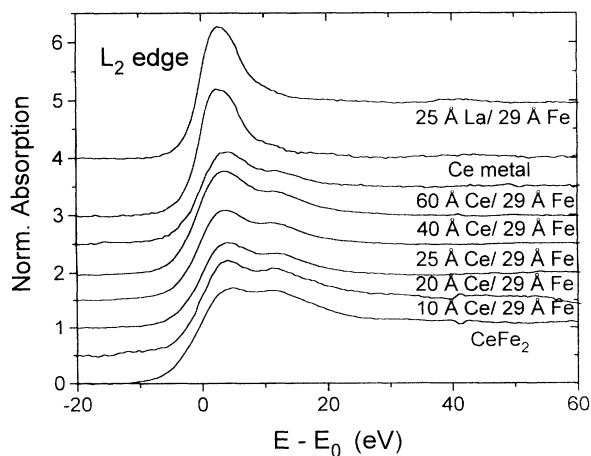


FIG. 4. Normalized x-ray-absorption spectra at the La- or Ce- $L_2$  edge of a La/Fe multilayer and of several Ce/Fe multilayers (Fe-layer thickness 29 Å, total thickness  $\sim 4000$  Å), of  $\gamma$ -Ce metal and the intermetallic compound  $\text{CeFe}_2$ . The curves have been displaced vertically.  $E_0 = 5891$  eV (La) and 6164 eV (Ce).

trum of  $\text{CeFe}_2$ . As we have emphasized above, this compound is a well-known  $\alpha$ -Ce-like (or mixed-valent) system, with delocalized 4*f* states owing to a strong hybridization of the 4*f* and conduction-band states.<sup>1</sup> Exactly the same behavior was observed for the  $L_3$  edge. The characteristic double-peak structure in the  $L_{2,3}$  absorption edges of Ce is a signature of final-state effects associated with 4*f*<sup>1</sup> and 4*f*<sup>0</sup> channels. It reflects the two possible screening mechanisms of the 2*p* core hole, via 4*f* electron or 5*d* band electrons.<sup>29</sup> The continuous evolution in relative intensities of the two maxima in the  $L_{2,3}$  spectra as the Ce-layer thickness in the multilayers decreases indicates the presence of Ce with  $\alpha$ -phase-like 4*f* configuration, in addition to  $\gamma$ -phase-like Ce with 4*f*<sup>1</sup> configuration. The most obvious interpretation is to assume that the  $\alpha$ -phase-like Ce atoms are located in the in-

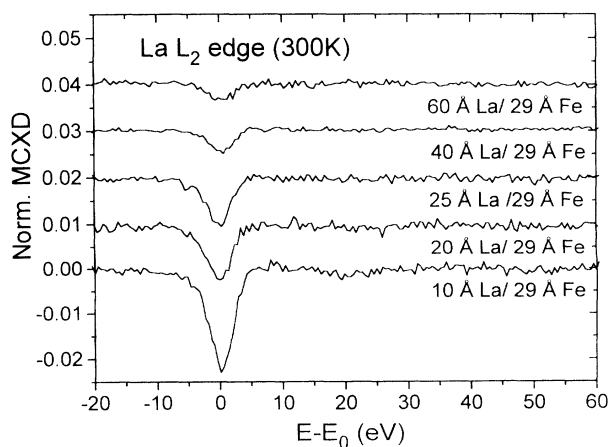


FIG. 6. Normalized MCXD spectra at the La- $L_2$  edge (Fe-layer thickness 29 Å, total thickness  $\sim 4000$  Å) in an external magnetic saturation field (5 kOe) at room temperature. The curves have been displaced vertically.

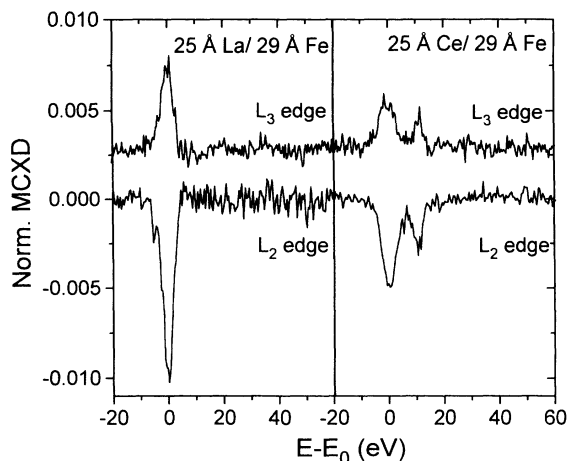


FIG. 7. Normalized MCXD spectra at the  $L_2$  and  $L_3$  edges of a La/Fe and a Ce/Fe multilayer (total thickness  $\sim 4000$  Å) in an external saturation field at room temperature.  $L_3$  edge:  $E_0 = 5483$  eV (La) and  $5723$  eV (Ce).

terface region and the  $\gamma$ -phase-like-ones in the core of the Ce layers. We recall that Ce is amorphous. It has been already shown by Malterre *et al.*<sup>30</sup> that crystallinity is not a prerequisite for the appearance of mixed valency, in fact it is of minor importance.

The presence of a MCXD signal at the  $L_{2,3}$  edges of both Ce and La in their multilayers with Fe even at room temperature (Figs. 5–7) indicates that a considerable portion of the RE atoms carries an ordered  $5d$  magnetic moment (note that the MCXD intensity is only a small fraction of the total edge jump). For the Ce/Fe multilayers, the MCXD signal consists of two contributions (Fig. 5), as the total absorption signal, with roughly the same splitting in energy in both cases. This reveals that in the two underlying channels a  $5d$  magnetic moment must be present. In contrast to the total spectrum, in the dichroic spectrum the amplitude ratio of the two lines does not vary with the Ce-layer thickness  $t_{\text{Ce}}$ , in spite of a considerable increase of the amplitudes themselves with decreasing  $t_{\text{Ce}}$  (compare Figs. 4 and 5). Amplitude ratio, splitting, relative linewidths and sign of the signals [negative for  $L_2$ , positive for  $L_3$  (Fig. 7)], i.e. the overall structure, are almost equal to the MCXD spectrum of the  $\alpha$ -Ce-like compound  $\text{CeFe}_2$  well below its Curie temperature ( $T_C = 230$  K).<sup>28,21</sup> It can be concluded then that only  $\alpha$ -phase-like Ce atoms carry the ordered magnetic  $5d$  moment reflected in the MCXD signal.

To obtain an estimate of the portion of  $\alpha$ -phase-like Ce in the multilayers, we apply a phenomenological analysis: we model the measured absorption spectra in Fig. 4 by superposing those of Ce metal and  $\text{CeFe}_2$ . It should be noted that this approximation neglects the possibility of a gradient in the Ce  $4f$ -state occupancy near the Ce/Fe interface, extending from the saturated mixed-valent value<sup>29</sup>  $n_f \approx 0.7$  as in  $\text{CeFe}_2$  to somewhat higher values. The present estimate therefore yields the lower limit of the  $\alpha$ -phase-like Ce part in the multilayers. The result, expressed in partial thickness of  $\alpha$ -phase-like Ce in the individual Ce layers, is shown in Fig. 8(a). The data ap-

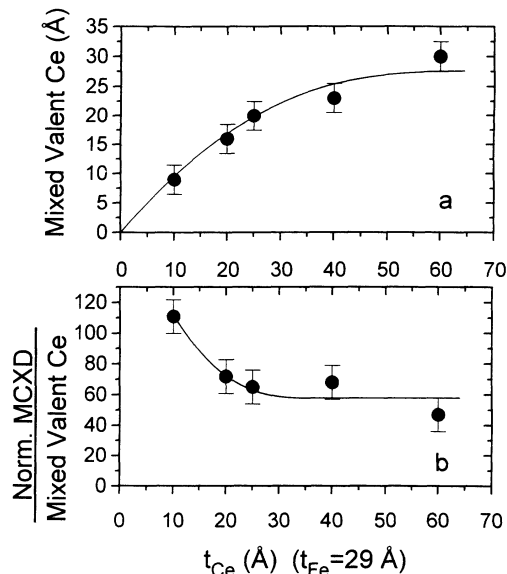


FIG. 8. (a) Partial thickness of  $\alpha$ -phase-like (mixed valent) Ce near the interfaces of Ce/Fe multilayers (Fe-layer thickness  $t_{\text{Fe}} = 29$  Å, total thickness  $\sim 4000$  Å) versus the total thickness of the Ce layers,  $t_{\text{Ce}}$ . (b) Integrated normalized MCXD intensity (in arbitrary units) referred to the contribution of  $\alpha$ -phase-like (mixed valent) Ce of Ce/Fe multilayers as a function of the total Ce-layer thickness. The solid lines in (a) and (b) are guides to the eye.

proach a saturation value for the  $\alpha$ -phase-like Ce part at large Ce-layer thicknesses  $t_{\text{Ce}}$ . This saturation value could not be determined here, but clearly it surpasses  $30$  Å. According to the results of the structural investigations reported above which exclude diffusion,<sup>3</sup> this value is much too large to be attributed to a chemical effect which might result from the close proximity of Ce and Fe at the interface. Moreover, this is ruled out by the profile of the Fe  $K$  absorption edge. Therefore, we suggest that Ce near the interface adopts, on a considerable length scale, the  $\alpha$ -phase-like electronic configuration and hence a smaller atomic volume to reduce the misfit and strain at the interfaces. (The  $\gamma$ -to- $\alpha$  phase transition in Ce metal is connected with a 17% decrease of the atomic volume<sup>31</sup>). Indeed, the thicknesses of the individual Ce layers in the multilayers under study determined from small-angle x-ray diffraction are significantly smaller than the nominal ones calculated from the mass coverage of the quartz crystal of the thickness monitor and the density of crystalline  $\gamma$ -Ce.<sup>3</sup> For the multilayer Ce(20 Å)/Fe(29 Å), for example, the deficit in Ce-layer thickness is 12%. Apparently the decrease in thickness due to the adoption of the  $\alpha$ -phase-like state outweighs, to a large extent, the increase which should result from the lower density of the amorphous structure.

An important aspect is the driving mechanism for magnetic order both in the Ce and La layers near the interface with Fe revealed by the presence of MCXD at the  $L_{2,3}$  edges of these metals. It is essential to point out that the dichroic signals do not originate from a possible Ce-Fe or La-Fe compound or alloy at the interface as, for the

Ce/Fe system, might naively be conjectured on account of the similarities to CeFe<sub>2</sub> just mentioned. This can be ruled out since the magnetic ordering temperatures of all existing Ce-Fe or La-Fe intermetallic compounds and alloys are below room temperature, while MCXD at the  $L_{2,3}$  edges Ce and La is observed at room temperature. [The only compounds are CeFe<sub>2</sub> and Ce<sub>2</sub>Fe<sub>17</sub>, no compounds are known between La and Fe. For both RE-Fe systems, only amorphous alloys exist, with maximum magnetic ordering temperatures of 220 K (30 at. % Ce)<sup>32</sup> and 290 K (25 at. % La)<sup>33</sup>]. The overall intensity of the signal decreases with the thickness of the Ce and La layers (Figs. 5 and 6). This reveals clearly that it is induced by the Fe layers. Indeed, measurements performed down to 4.2 K (for the Ce/Fe layers) demonstrate that the integrated MCXD intensity follows rather closely the temperature variation of the macroscopic magnetization of the multilayers. This implies that the dichroic signal observed reflects ground-state properties of the probed RE ions. It may be assumed that the underlying magnetic polarization of the RE  $5d$  states is generated by their hybridization with the Fe  $3d$  states near the interface, in a similar way as in RE-Fe intermetallics. Theoretical work<sup>2</sup> has shown that this mechanism produces a significant  $5d$  spin density at the RE sites of such compounds, even if the  $4f$  moments are zero. We shall address this important effect again below in the course of this discussion.

Figure 9 shows that for the La/Fe multilayers the MCXD intensity varies linearly with the inverse of the La-layer thickness,  $1/t_{\text{La}}$ , in good approximation, down to the lowest thickness investigated ( $t_{\text{La}} = 10 \text{ \AA}$ ). This indicates that magnetic order in the La  $5d$  states is confined to the immediate interface with Fe, i.e. to  $t_{\text{La}} < 5 \text{ \AA}$  at each interface. The situation is different for the Ce/Fe multilayers. As we have pointed out, comparison of the data in Figs. 4 and 5 reveals that only  $\alpha$ -phase-like Ce atoms near the interfaces are magnetically polarized. Therefore, we present in Fig. 8(b) the integrated Ce- $L_2$  MCXD signal referred to the  $\alpha$ -phase-like part of the individual Ce layers as a function of their thickness  $t_{\text{Ce}}$ , for

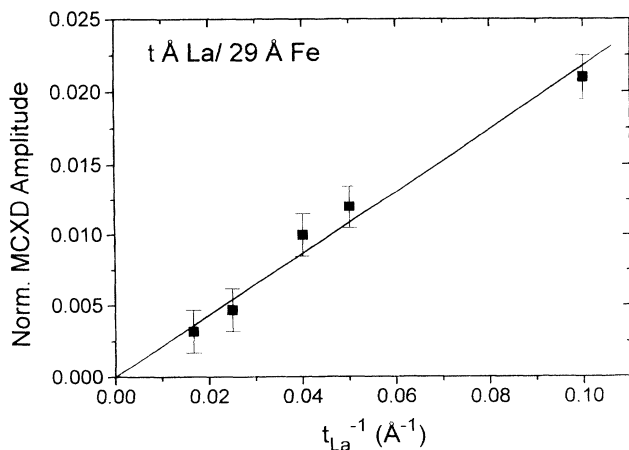


FIG. 9. Normalized La- $L_2$  MCXD amplitude of the La/Fe spectra in Fig. 6 as a function of the inverse La-layer thickness.

$t_{\text{Fe}} = 29 \text{ \AA}$ . A constant value is approached for  $t_{\text{Ce}}$  somewhat above  $30 \text{ \AA}$ . The most obvious way to understand this result is to interpret the Ce-layer thickness of  $\sim 30 \text{ \AA}$ , where the normalized MCXD data merge into a constant value, as the effective thickness of the magnetically polarized Ce atoms in these multilayers. This value for the “magnetic extension” of the interface ( $\sim 15 \text{ \AA}$ ) by far surpasses the above-mentioned structural irregularities due to roughness, which were determined by small-angle x-ray diffraction.<sup>3</sup> (We recall that there is no diffusion). The results imply that the magnetic polarization of the  $5d$  states of the  $\alpha$ -phase-like Ce atoms extends to nominally about four atomic layers into the Ce layers on each side of the interface, while in the case of La only about one atomic layer is affected. The pronounced upturn of the normalized MCXD intensity for the multilayer Ce ( $10 \text{ \AA}$ )/Fe( $29 \text{ \AA}$ ) displayed in Fig. 8(b) finds a natural explanation in that for this sample the effective magnetic thickness surpasses the thickness of the Ce layers and hence both Fe layers neighboring a Ce layer contribute to its magnetic polarization.

The different extension of the magnetically polarized region in the individual La and Ce layers near the interface with Fe is surprising. We recall that the magnetic hyperfine fields in the Fe layers near the interfaces of the La/Fe and Ce/Fe multilayers are perturbed in a different way. Both effects provide evidence for a different hybridization between the electronic states of Fe with those of La or Ce. It is reasonable to normalize the MCXD signals observed, which in Figs. 5 and 6 are referred to the total number of RE atoms, with the magnetically polarized La and Ce atoms only (we recall that at most only  $5 \text{ \AA}$  of La but about  $15 \text{ \AA}$  of Ce are concerned at each interface of the multilayers). The resulting average dichroic intensity per magnetic RE atom is considerably higher in the La/Fe system. This is surprising in view of the recent observations<sup>21</sup> in related intermetallic compounds with  $\alpha$ -phase-like Ce and their comparable non- $4f$  counterparts, for example CeFe<sub>2</sub> and LuFe<sub>2</sub>, or CeCo<sub>5</sub> and LaCo<sub>5</sub>, where the dichroic signals on the  $L_{2,3}$  edges of Ce and La or Lu appear to be very similar in magnitude. For these compounds, it was clearly established that the RE  $L_{2,3}$  MCXD intensity is proportional to the magnitude of the  $5d$  magnetic moment of the probed atoms. For the Ce compounds, this appears to be related to the delocalization of the  $4f$  states. This rule should equally apply to the present RE/Fe multilayers, since La is a non- $4f$  element and only the  $\alpha$ -phase-like Ce atoms have their  $5d$  states magnetically polarized.

Adopting the relation between the  $L_{2,3}$  MCXD amplitude and the  $5d$  magnetic moment of the RE given in Ref. 21 (Table II) for the compounds LuFe<sub>2</sub> and LaCo<sub>5</sub> and bearing in mind that at each interface at most  $5 \text{ \AA}$  of La is magnetically polarized according to Fig. 9, we estimate a magnetic  $5d$  moment of  $0.6\mu_B$  for a La atom at the interface with Fe in the multilayers. For the Ce/Fe interface, the situation is more complex. Using the relation between the MCXD amplitudes and the  $5d$  magnetic moment specified in Ref. 21, we estimate an average  $5d$  moment of  $0.2\mu_B$  per Ce atom, if the signal is referred equally to the entire polarized part in the Ce layers. But



in view of its relatively large extension and of the special hyperfine-field distribution discussed above, and owing to the fact that the dichroic signal has been found to be proportional to the magnetization, this is not justified. We recall that according to the Mössbauer spectra<sup>6</sup> the Fe atoms immediately at the interface with Ce, on a length scale defined by the local thickness fluctuations (roughness) of less than two atomic layers, display a very small magnetic hyperfine field (centered at 45 kOe compared to 334 kOe for bulk Fe). Hence this region is only weakly magnetic and contributes only little to the magnetic polarization of the Ce atoms it encloses. Furthermore, for the remaining Ce atoms the strength of ferromagnetic ordering induced by the Fe layers will decay with the distance from the interfaces. As a result of these two effects, the local  $5d$  magnetic moments generating the MCXD signal are not uniformly distributed over the polarized part of the Ce layers. There will be a maximum value at some distance beyond the first atomic layer. The overall average magnetic hyperfine field of the Fe layers is about 15% smaller in the Ce/Fe than in the La/Fe heterostructures (see above), and qualitatively the same trend is to be expected for the Fe-layer magnetization. Therefore, magnetic polarization by Fe will be less intense in the Ce/Fe than in the La/Fe system. Taken together, these characteristics provide a satisfactory explanation for the different magnitude of the  $5d$  magnetic moment induced on the Ce and La atoms near the interfaces in the multilayers, but they do not explain the difference in the length scale of the magnetic polarization in the two systems. It should be noted that, compared to the  $5d$  moments of the related compounds<sup>21</sup> (we have  $0.3\mu_B$  for Lu in LuFe,  $0.2\mu_B$  for La in LaCo<sub>5</sub>, and  $0.3\mu_B$  for Ce in CeFe<sub>2</sub>), the  $5d$  moment of La at the La/Fe interface is exceptionally high.

Figure 7 shows that the branching ratio of the MCXD intensities at the  $L_2$  and  $L_3$  edges,  $I(L_2)/I(L_3)$  is close to  $-2$  in both RE/Fe multilayer systems. According to recent theoretical work this implies that the orbital contribution to the  $5d$  magnetic moment almost vanishes.<sup>34</sup> This is a manifestation of crystal-field effects which are commonly expected to quench the orbital moments of  $d$ -band electrons. Hence the  $5d$  moments observed on La and Ce at the interfaces of the multilayers are spin moments.

An important feature of MCXD is that the experimentally determined sign of the signal permits to deduce the orientation of the spin polarization of the absorbing atomic species relative to the net macroscopic magnetization. For the Ce/Fe multilayers (Figs. 3, and 5–7), the signs are identical to those in CeFe<sub>2</sub>:<sup>21,28</sup> positive just above the  $K$  edge of Fe, and negative (positive) for the double-peak structure at the  $L_2$  ( $L_3$ ) edge of Ce. For the compound, this is correlated with an orientation of the Fe  $3d$  magnetic moment parallel and of the Ce  $5d$  moment antiparallel to the net magnetization pointing in the direction of the external magnetic field,<sup>21</sup> in agreement with the one-electron approach of MCXD.<sup>22</sup> Note also that the sign and shape of the dichroic signal at the  $K$  edge of Fe are the same in the multilayers and in pure Fe (Fig. 3). Hence the Fe  $3d$  magnetic moment has the same

orientation in both cases, it is parallel to the applied magnetic field. We can conclude then that we have an antiferromagnetic coupling between the  $3d$  moments of Fe and the  $5d$  moments of  $\alpha$ -phase-like Ce near the interfaces in the multilayers. As the  $L_{2,3}$  MCXD signals of the La/Fe multilayers (Figs. 6 and 7) have the same sign as for Ce/Fe, we have the same relative orientation at the magnetically polarized La/Fe interfaces, i.e., the La  $5d$  moment is antiparallel to the Fe  $3d$  moment. This corroborates the usual coupling scheme<sup>35</sup> known for compounds of RE and ferromagnetic  $3d$  TM elements, where the  $5d$  and  $3d$  electrons strongly hybridize.

The  $5d$  moments of La and Ce derived from MCXD can be used to estimate their contribution to the macroscopic global magnetization of these ferrimagnetic multilayers. To make a comparison with the directly measured data in Fig. 1, we apply the relative decrease of the average magnetic hyperfine field with respect to bulk bcc Fe (8% for La/Fe, 22% for Ce/Fe, see above) to estimate the contribution of the Fe part to the magnetization of the multilayers, even though this is not strictly justified. It results that the  $5d$  magnetization induced on La and Ce near the interfaces amounts to only 3% of the respective Fe-layer contribution, which is quite small. For the multilayers with 25 Å of La and Ce we have included the resulting total magnetization values in Fig. 1. They are within the error bars of the directly determined data. Obviously, the main contribution to the macroscopic magnetization of the multilayers is the part provided by the Fe layers. It is remarkable that this Fe part of the magnetization is very different in the two systems. We suggest that hybridization effects of Fe and RE electron states in the vicinity of the interfaces are at the origin of this behavior, by a similar mechanism as in intermetallic compounds of RE and late TM elements (TM: e.g., Fe, Co). It has been shown<sup>2</sup> that in such systems hybridization of the TM  $3d$  states with the RE  $5d$  states induces  $5d$  magnetic spin moment in opposite direction to the  $3d$  magnetic moment. In compounds with itinerant  $4f$  states like in CeFe<sub>2</sub>, additional  $4f$ - $3d$  hybridization is important which has two effects: first, it induces a  $4f$  spin moment opposite to the  $3d$  moment in the same way as for the  $5d$  electrons, second, it reduces the  $3d$  moment owing to a diminished occupation of the TM  $3d$  majority spin band. Obviously, if the interaction between Fe and La or Ce near the interfaces in the multilayers is governed by a similar mechanism as in the intermetallic compounds, this would provide an explanation for the different Fe-layer magnetization in the two systems.

Schillé *et al.*<sup>36</sup> have recently performed a MCXD experiment at the  $M_{4,5}$  edge of Ce in CeFe<sub>2</sub> which directly probes  $4f$  magnetism. They determined a  $4f$  magnetic moment of  $0.2\mu_B$  per Ce atom. Let us mention that an extra  $4f$  contribution of similar magnitude would be compatible with the directly measured magnetization of the Ce/Fe multilayers in Fig. 1.

Finally it is important to point out that the ground-state configuration with antiferromagnetic coupling of the Fe  $3d$  and the RE  $5d$  or  $4f$  moments is very stable in the multilayers. Measurements of the magnetization do not provide any indication that the magnetic polarization



of the RE atoms can be reversed by the available field strength of 50 kOe.

## V. CONCLUSION

Absorption spectroscopy with hard x rays performed on Ce/Fe and La/Fe multilayers with sharp interfaces provides information on the interaction between Ce or La and Fe at their common interface. Ce adopts, on a considerable length scale near the interface, the electronic structure of the  $\alpha$ -phase which is characterized by delocalized  $4f$  states. It can be derived from the spin-resolved absorption spectra that Fe induces an ordered magnetic spin moment on the  $5d$  states of the  $\alpha$ -phase-like Ce atoms, which is antiferromagnetically coupled to the Fe moment. The La atoms are similarly magnetically polarized, but only immediately at the interface. It is suggested that the ordered magnetism on the interfacial RE atoms is induced by hybridization of the Fe  $3d$  and RE  $5d$  electron states, with different strength in the two multilayer systems. The contribution of the RE  $5d$  moment to the total magnetization of the multilayers for

both systems amounts to about 3% of the Fe-layer part estimated from the magnetic hyperfine fields. A magnetic polarization of a paramagnetic RE metal near the interface with bcc Fe in a layered structure has been observed before, on a comparable length scale as in the Ce/Fe structures, for Nd/Fe multilayers by MCXD at the Nd  $L_2$  edge<sup>13</sup> and for thin Sm overlayers on Fe(100) by spin-resolved  $4f$  photoemission.<sup>37</sup> In the latter case, this is attributed to a Sm  $4f$ -Fe  $3d$  hybridization. The Ce/Fe system discussed here is unique in the sense that Ce atoms with localized as well as itinerant  $4f$  states are present in the layered structure and that only the latter ones have their  $5d$  states magnetically polarized.

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