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$4f$ orbital and spin magnetism in cerium intermetallic compounds studied by magnetic circular x-ray dichroism

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Magnetic circular x-ray dichroism experiments at the $M_{4,5}$ absorption edges of cerium in the intermetallic compounds CeCuSi, $CeRh_3B_2$, and $CeFe_2$ are reported. By applying general sum rules, it is shown that these experiments are able to yield both the magnitude and the direction of the $4f$ magnetic moment on Ce. An estimation of the orbital contribution to those $4f$ moments is given. Our experiments demonstrate the existence of a $4f$ magnetic moment on Ce in $CeFe_2$ and confirm the extreme sensitivity of the $4f$ orbital contribution to the degree of localization of the $4f$ electrons. This $4f$ orbital contribution is significantly higher than the one predicted from spin-resolved band-structure calculations.

The existence of magnetic moments of $4f$ origin in strongly correlated ferromagnetic systems, such as, for instance, the intermediate-valence (IV) α-cerium compounds $CeFe_2$ or $CeCo_5$,[1,2] is a controversial subject. In this class of materials we must include also the nearly trivalent cerium compounds for which the $4f$ electrons form a 2–3 eV broad $4f$ band; the best example is probably $CeRh_3B_2$ which orders ferromagnetically at $T_c = 115$ K.[3] The controversy comes from the fact that the itinerant character of the $4f$ electrons in such materials could prevent the formation of $4f$ magnetic moments. Even if spin-polarized energy-band calculations were able to predict the existence of a $4f$ magnetic moment in $CeFe_2$ ($M_{Ce}^{4f} \approx -0.27\mu_B$) (Ref. 4) of almost pure spin origin (the orbital contribution being quenched by the itinerant character of the $4f$ electrons), experimental evidence for the existence of a magnetic moment at the Ce sites was only given in 1990 by neutron-diffraction experiments,[5] which indicated a total moment on Ce of about $-0.5\mu_B$, but were unable to separate the $4f$ and the $5d$ contributions. Our previous magnetic circular x-ray dichroism (MCXD) experiments performed at the $L_{2,3}$ absorption of Ce,[6,7] which probe the $5d$ empty states, led to a $5d$ magnetic moment on Ce of about $-0.35\mu_B$ and it was proposed that the complement ($\approx -0.15\mu_B$) may be due to a $4f$ contribution. The aim of this work is to give unambiguous evidence of the existence of such a $4f$ magnetic moment and to try to separate the orbital and the spin contributions, which should be of prime importance for knowledge of the $4f$ magnetism and justifies the present study on some Ce intermetallic compounds.

MCXD experiments present the huge advantage of being site and orbital selective; moreover, it is now well known that the use of general sum rules[8,9] enables one to measure the ground-state orbital contribution to the magnetic moment of any given shell. It is thus an exciting challenge to try to use such a powerful technique for the determination of $4f$ magnetism in situations where the hybridization between the $4f$ electrons and the conduction electrons is rather different. This can be achieved by performing MCXD at the $M_{4,5}$ absorption edges of Ce where, due to the electric dipole selection rules, only $3d^{10}4f^n \rightarrow 3d^94f^{n+1}$ transitions contribute. Such spectra are dominated by strong multiplet effects due to the Coulomb interaction between the $4f$ electrons and the $\underline{3d}$ core hole.[10] In the general case of the rare earths (RE's), atomic calculations[11,12] reproduce fairly well the shape of such $M_{4,5}$ absorption edges. In the case of cerium-based

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systems the agreement is less satisfactory,[13] and these discrepancies will be discussed.

In order to get a quantitative analysis, we use the following sum rule proposed recently by Carra and coworkers.[8,9] At the spin-orbit-split $M_{4,5}$ absorption edges,

$$\rho = \int_{M_{4,5}} (\mu^+ - \mu^-) dE \Big/ \int_{M_{4,5}} (\mu^+ + \mu^- + \mu^0) dE = \frac{-1}{3(14-n)} \langle L_z \rangle \ , \qquad (1)$$

where μ^+, μ^-, and μ^0 denote the absorption coefficient for left, right, and linearly polarized light, n is the occupancy of the $4f$ shell ($n=1$ for Ce^{3+}), and $\langle L_z \rangle$ is the orbital magnetic moment of the $4f$ shell in the *initial state*. The pertinence of this sum rule has been demonstrated on transition metals.[8] However, it has to be stressed that the use of this general sum rule is much easier in the case of the $M_{4,5}$ edges of rare earths due to the relative weakness of the transitions towards the continuum states.

Let us mention that another sum rule has been proposed:[9] to measure a linear combination of $\langle S_z \rangle$ and $\langle T_z \rangle$ (associated with a magnetic dipole operator). However, the derivation of this sum rule assumes that the M_5 and M_4 edges correspond to pure $3d_{5/2}$ and $3d_{3/2}$ states of the core hole. In the case of Ce, this assumption is not valid since the spin-orbit parameter ξ_{3d} deduced from the energy separation between M_5 and M_4 edges is 7.4 eV, which is not large compared to the Slater integrals (ranging from 1.6 to 10 eV, according to Refs. 10 and 13). Therefore, Coulomb interaction mixes spin-orbit-split states. To test the breakdown of the sum rule of Ref. 9, we have calculated the normal and dichroic spectra of Ce according to the method of Refs. 10, 13, and 14. The areas of the isotropic M_5 and M_4 edges are 0.29 and 0.33, respectively, instead of $\frac{25}{63}$ and $\frac{2}{9}$ expected from the sum rule. The areas of the MCXD at the M_5 and M_4 edges are $+0.045$ and -0.18, respectively, instead of $+\frac{2}{735}$ and $-\frac{2}{15}$ expected from the sum rule. Therefore, the sum rule of Ref. 9 is invalid for Ce and cannot be used to deduce the spin contribution to the $4f$ moment.

On the other hand, the sum rule giving $\langle L_z \rangle$ is much more general, being true even in the presence of hybridization,[8] and $\langle S_z \rangle$ can be estimated indirectly from it. The values of the total magnetic moment on cerium M_{Ce}^T are known from either magnetization or neutron-diffraction experiments. Furthermore, previous MCXD experiments at the $L_{2,3}$ edges provide an estimation of the magnetic moment of $5d$ origin.[7] Thus, the $4f$ magnetic moment of Ce being $M_{Ce}^{4f} = -(\langle L_z \rangle + 2\langle S_z \rangle)$, the deduction of the spin contribution is straightforward. Because of the Wigner-Eckart theorem, within a given J manifold, the ratio $\langle S_z \rangle / \langle L_z \rangle$ is independent of perturbations such as the crystal field or the magnetic field (for $J=\frac{5}{2}$ this ratio is $-\frac{1}{8}$). The discussion about the determination of $\langle S_z \rangle$ will appear at the end of this paper.

The MCXD experiments were performed at the French Synchrotron Facility (LURE) in Orsay using the light emitted from the asymmetric wiggler inserted in the Super-ACO ring.[15] The left ($+\hbar$ helicity) circular polarization rate (typically 25% at the working energies) is obtained by moving all the optics +0.3 mrad above the orbit plane. The relatively small polarization rate is explained by a significant depolarization of the beam consecutive to the two Bragg reflections on a beryl double-crystal monochromator. The dichroic signal is obtained simply by the difference between the absorption spectra recorded for two opposite directions of the external magnetic field (**B** parallel or antiparallel to the photon helicity). Reversing the applied field is fully equivalent to changing the helicity of the photons. The magnetic field ranges up to 7 T and the lowest sample temperature that can be achieved is 15 K. The dichroic spectra presented in this paper are normalized to 100% of circular polarization.

The first example concerns the CeCuSi compound. This γ-type compound orders ferromagnetically at $T_C = 15$ K, the highest Curie temperature for a γ-Ce compound. It crystallizes in the AlB_2-type structure, which corresponds to a hexagonal symmetry (space group $P6/mmm$);[16] the bigger trivalent Ce atom is at the center of the cavity defined by a net of hexagonal Cu-Si rings. From magnetization measurements the total Ce magnetic moment, in $B=6$ T, is $+0.94\mu_B$ at $T=4.2$ K and at $T=15$ K this moment is reduced to $+0.76\mu_B$. Such values are far away from the $gJ=2.14\mu_B$ moment expected from a $(M_J=\frac{5}{2}, J=\frac{5}{2})$ ground state and reflect the crystal-field effects which act on the Ce atoms. In Fig. 1 we report the $M_{4,5}$ absorption edges and the MCXD of Ce in CeCuSi at $T=15$ K under a magnetic field of 6 T. The existence of a $4f$ magnetic moment on Ce, in the direction of the applied magnetic field, is thus clearly established. Moreover, we notice that the shape and intensity of the MCXD effect are well reproduced using atomic calculations in a pure $J=\frac{5}{2}$ ground state (full line in Fig. 1).

As shown in Table I, considering a total magnetic moment $M_{Ce}^T = +0.76\mu_B$ and a partial $M_{Ce}^{5d} \approx 0.1\mu_B$, the magnetism of this compound is essentially of $4f$ origin with $M_{Ce}^{4f} = +0.66\mu_B$. Applying the relation (1), the value obtained from MCXD for $\langle L_z \rangle$ is -0.88. The value of $\langle S_z \rangle$ can then be estimated to $\approx +0.11$. It has to be stressed that, in this case, the ratio between $\langle L_z \rangle$ and $\langle S_z \rangle$ is exactly -8 as expected for a pure $J=\frac{5}{2}$ initial state. This MCXD analysis confirms the high localization of the $4f$ electrons in CeCuSi deduced from the thermodynamic properties.[16]

The second example proposed is the $CeRh_3B_2$ compound. This compound (distorted hexagonal $P6/mmm$ $CeCo_3B_2$-like structure) is of great interest because of its exceptional magnetic properties: It is ferromagnetic with a Curie temperature of 115 K, higher than that of $GdRh_3B_2$ ($T_c \approx 90$ K).[17,18] The rather small magnetic moment at low temperature (≈ 0.37 μ_B/f.u.) (Ref. 19) has been explained by itinerant magnetism consecutive to Ce $4f$-Rh $4d$ hybridization which leads to the formation of a wide (2–3 eV) $4f$ band.[20] From the $L_{2,3}$ x-ray absorption experiments,[21] the Ce ions are found to be almost in a trivalent state (i.e., the occupancy of the $4f$ shell is $n_f \approx 1$ as for CeCuSi). From spin-polarized

TABLE I. Magnetic moments deduced from MCXD analysis compared to those obtained by band-structure calculations (BSC) in the CeCuSi, $CeRh_3B_2$, and $CeFe_2$ compounds. The uncertainties on the moment determination are estimated to be about 15% for CeCuSi and $CeRh_3B_2$, and 50% for $CeFe_2$.

Compounds	$\langle L_z^{4f} \rangle$	M_{Ce}^{T} (μ_B)	M_{Ce}^{5d} (μ_B)	M_{Ce}^{4f} (μ_B)	$\langle S_z^{4f} \rangle$
CeCuSi		+0.76[a]			
MCXD	−0.88		+0.1[b]	+0.66	+0.11
$CeRh_3B_2$		+0.30[c]			
MCXD	−0.3		+0.1[b]	+0.2	+0.05
BSC[d]	+0.04(4*f* +5*d*)	+0.25			−0.14(4*f* +5*d*)
$CeFe_2$		≈ −0.5[e]			
MCXD	+0.2		−0.35[b]	−0.15	−0.02
BSC[f]	−0.15	−0.57	−0.3	−0.27	+0.21

[a] From magnetization experiments at $T=15$ K and $B=6$ T.
[b] From MCXD at Ce $L_{2,3}$ edges (Ref. 7).
[c] From magnetization experiments at $T=30$ K and $B=1$ T.
[d] From Ref. 20.
[e] From Ref. 5.
[f] From Ref. 4.

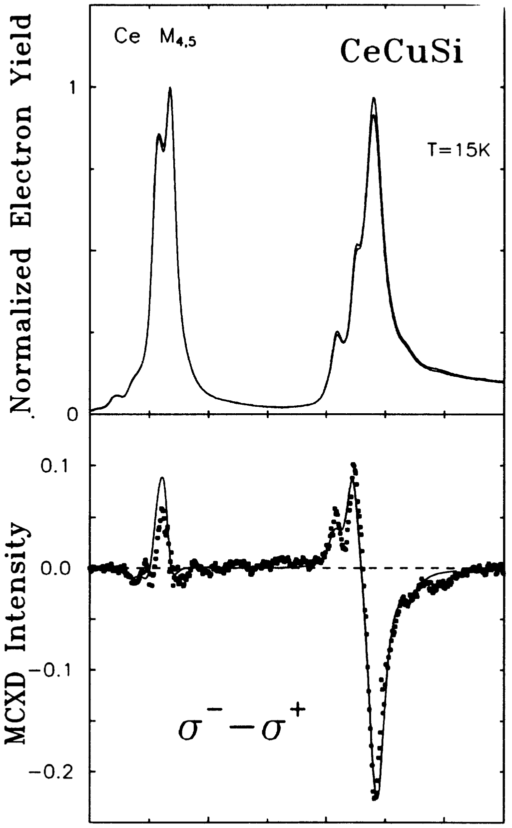

FIG. 1. Upper panel: Ce $M_{4,5}$ normalized absorption spectra in CeCuSi, with left circularly polarized light at $T=15$ K, under B= +6 and −6 T. Lower panel: MCXD experimental (●) and theoretical (full line) spectra. The experimental MCXD spectrum is obtained by the difference between the two absorption spectra, i.e., $(\sigma^- - \sigma^+)$, σ^+ (σ^-) standing for the absorption with the magnetic field parallel (antiparallel) to the photon propagation vector. The experimental MCXD spectrum is normalized to 100% of left circular polarization rate and the calculated $(J=\frac{5}{2})$ MCXD is normalized to the minimum of the experimental spectrum.

energy-band calculations,[20] the total magnetic moment is $+0.27$ μ_B/f.u., decomposed into a Ce moment of $+0.25$ μ_B/atom and a Rh moment of $+0.02$ μ_B/atom. According to these calculations the magnetic moment on Ce appears to be essentially of spin origin, since the calculated $(4f+5d)$ orbital contribution is found to be only $-0.04\mu_B$.

The spectra (edges and MCXD) recorded for $CeRh_3B_2$ ($T=30$ K and $B=\pm 1$ T) are displayed in Fig. 2. At first sight, both the absorption edges and the MCXD look like those of CeCuSi. However, several significant differences can be seen. Weak satellites associated with the Ce $4f^0$ configuration,[22] are located near 887 and 903 eV. Moreover, some changes are detected in the shape of the MCXD spectra, especially at the M_4 edge. These particular features can be quantitatively explained by hybridization between the 4*f* and the conduction electrons.

Once more, as shown in Table I, considering a total magnetic moment $M_{\mathrm{Ce}}^{T}=+0.30$ μ_B/f.u. and $M_{\mathrm{Ce}}^{5d}\approx 0.1\mu_B$, if the moment of rhodium is neglected one gets $M_{\mathrm{Ce}}^{4f}=+0.20\mu_B$. The use of the sum rule leads to $\langle L_z \rangle = -0.3$ and consequently $\langle S_z \rangle = +0.05$. One can notice that the value expected for a $J=\frac{5}{2}$ ground state is $\langle S_z \rangle = +0.04$. The discrepancy with the band-structure calculations is evident, even if the order of magnitude and the sign of the total 4*f* moment are correctly reproduced. The orbital contribution appears to be incompletely quenched in $CeRh_3B_2$ and such a conclusion is in agreement with recent results obtained from Kerr spectroscopy.[23]

The last example deals with the $CeFe_2$ compound which is of particular interest because both the Ce and Fe atoms bear magnetic moments coupled antiferromagnetically[5] ($T_C=230$ K). This magnetic coupling, which is an unusual situation in light-RE intermetallic compounds, is consecutive to the delocalization of the 4*f* electrons; the Ce ions are strongly mixed valent ($V\approx 3.3$, i.e., $n_f\approx 0.7$).[6,7] Spin-resolved band calculations[4] yield a 4*f* magnetic moment on Ce of $-0.27\mu_B$ essentially of spin origin ($-0.42\mu_B$), the orbital contribution being only

$+0.15\mu_B$.

In Fig. 3, we show the absorption and MCXD spectra obtained for $CeFe_2$ at $T=30$ K and $B=\pm1$ T. The IV character of Ce in $CeFe_2$ is clearly evidenced in the absorption edges by the $4f^0$ structures located at 887 and 903 eV, which are much more intense that in the case of $CeRh_3B_2$. The multiplet structures, associated with a $J=\frac{5}{2}$ ground state, are still present but are strongly smoothed, as already observed on other IV compounds.[22] Furthermore, the M_4 edge is more intense than the M_5 one, which is the opposite of the situation encountered for $CeRh_3B_2$ and CeCuSi. The observation of a dichroism response demonstrates the existence of a $4f$ magnetic moment on Ce. As for the edges, the shape of the MCXD signal at the M_4 edge is significantly changed. Indeed, all the MCXD structure characteristic of $J=\frac{5}{2}$ appears in the $CeFe_2$ spectrum, but with huge changes in the relative intensities of the dichroic lines. All these observations confirm the increase of the hybridization strength when going from CeCuSi to the IV compound $CeFe_2$.

From a quantitative point of view, our starting point for the analysis of MCXD for $CeFe_2$ is a neutron-diffraction study[5] yielding the ratio of the magnetic moments on the Ce and Fe atoms: $M_{Ce}/M_{Fe}=-0.3$. The total magnetic moment on the compound being $2.6\mu_B$ (under a 1 T magnetic field and at $T=30$ K), the magnetic moment on the Ce site is about $-0.5\mu_B$. As shown in Table I, the $5d$ magnetic moment has been evaluated[7] to be $-0.35\mu_B$. Consequently, the $4f$ magnetic moment has to be about $-0.15\mu_B$. The decomposition of the $4f$ moment from the MCXD spectrum using relation (1) yields an orbital contribution of $\langle L_z^{4f}\rangle=+0.2\pm0.1$ and a spin part $\langle S_z^{4f}\rangle=-0.02\pm0.05$ (the large error bars are due to the low signal-to-noise ratio). These MCXD results give again rather different conclusions from those obtained from the band-structure calculations. Even if the values of the $4f$ and $5d$ magnetic moments are roughly in agreement with them both in magnitude and direction, the decomposition of the $4f$ magnetic moment in orbital and spin contributions extracted from MCXD analysis and that obtained from the calculations are in disagreement. The $4f$ orbital part in $CeFe_2$ is found to be rather large and its direction is opposite to that predicted

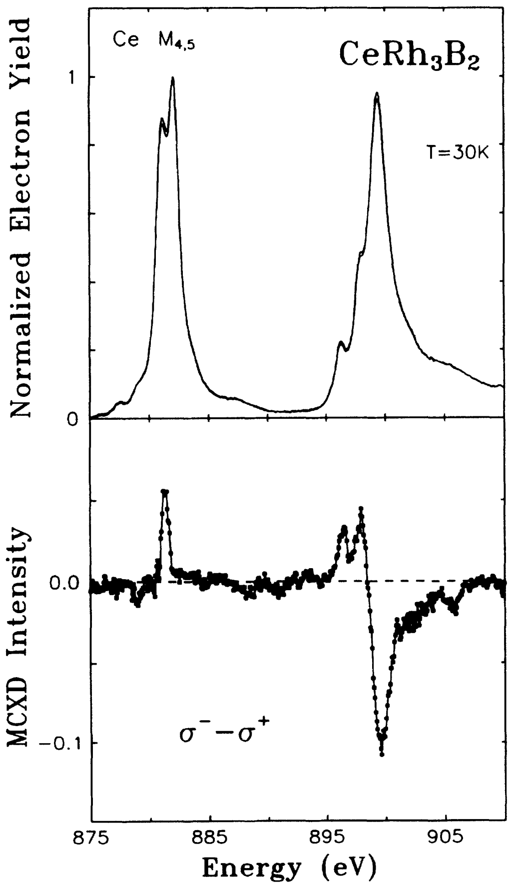

FIG. 2. Upper panel: Ce $M_{4,5}$ normalized absorption spectra in $CeRh_3B_2$, with left circularly polarized light at $T=30$ K, under B = +1 and −1 T. Lower panel: MCXD experimental spectrum, with the same convention as in Fig. 1. Full line is a guide for the eyes.

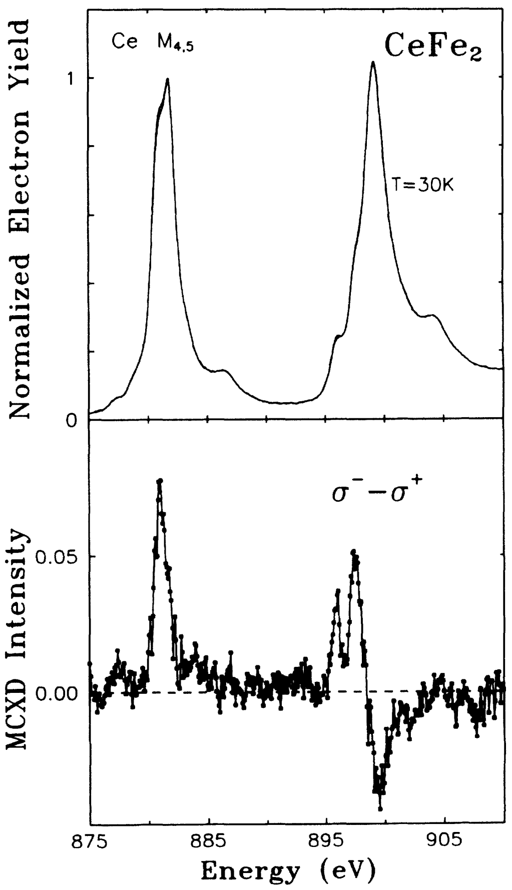

FIG. 3. Upper panel: Ce $M_{4,5}$ normalized absorption spectra in $CeFe_2$, with left circularly polarized light at $T=30$ K, under B = +1 and −1 T. Lower panel: MCXD experimental spectrum, with the same convention as in Fig. 1. Full line is a guide for the eyes.

by the calculations. The large value of the 4f orbital contribution excludes the possibility of obtaining a spin contribution of $-0.42\mu_B$ as expected from the calculations.

Let us now discuss why the experimental results are analyzed using the L_z sum rule only and why we do not try to compare the shape of the experimental spectra to the calculated ones. There are two reasons. First, the M_5 edge is not reproduced well in Ref. 14 for an initial state $J=\frac{5}{2}$. Secondly, the shape of MCXD spectra is always the same when the 4f shell state is in a pure J state split by crystal field, exchange, and external magnetic field.[24] This is contrary to our experiments on Ce compounds. A possible explanation for the changing shape of the MCXD spectra is that hybridization can mix $4f_{7/2}$ initial states with $4f_{5/2}$ ones.[13] To estimate the degree of hybridization it can be noticed that, for a nonmagnetically oriented powder, the absorption spectrum is a weighted sum of the $J=\frac{5}{2}$ and $J=\frac{7}{2}$ spectra (there is no interference term). The M_5 edge is reproduced with 30% of $J=\frac{7}{2}$ in the initial state, even for CeCuSi, which would be in contradiction with magnetic measurements. However, the $4f^0$ contribution, which is a clear indication of hybridization, is not present in CeCuSi. Because of these problems, we preferred to stick to the L_z sum rule, whose range of validity is broader than the calculations made in Ref. 13.

In conclusion, in this paper we have reported the application of the sum rule to Ce intermetallic compounds and shown how MCXD experiments at the $M_{4,5}$ absorption edges of Ce can yield valuable information on the magnetic properties of 4f electrons even in highly correlated materials. Such observations should stimulate further theoretical work on this subject. Finally, we want to emphasize that the MCXD experiments, performed at both the $L_{2,3}$ and $M_{4,5}$ absorption edges of Ce, allow one to solve completely the problem related to the determination of the local magnetic properties of Ce in any given material. In that sense such MCXD experiments are rather unique in the field of magnetism.

Note added. A recent neutron-diffraction study of CeFe$_2$ [S. J. Kennedy, P. J. Brown, and B. R. Coles, J. Phys. Condens. Matter **5**, 5169 (1993)] leads to a total magnetic moment on Ce of $M_{\text{Ce}}^T=-0.15\mu_B$, but this value is obtained taking the 5d/4f magnetization ratio from the band-structure calculations.

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