We present a discussion of published x-ray magnetic circular dichroism (XMCD) measurements performed at the uranium M\(_{4,5}\) edges of metallic uranium compounds, focusing on the shape of the dichroic signal at the M\(_5\) edge. A well resolved double lobe structure, comprised of a positive and negative peak, is sometimes observed. Out of the twelve metallic uranium compounds so far investigated by XMCD, six exhibit an intense double-lobe structure at the M\(_5\) edge. This line shape gives information on the empty 5\(f\) magnetic density of states with angular quantum number \(j = 7/2\). Conclusions about the difference between these two families of compounds are given regarding the splitting of the \(j = 7/2\) band and the occupation among the different \(m_{7/2}\) sublevels.

### I. INTRODUCTION

In recent years uranium compounds have been the subject of increasing interest because their ground state exhibits a variety of physical properties. They can be Pauli paramagnets or display an ordered magnetic state. The electronic correlations can be very large as revealed by a strong heavy fermion character or Kondo effect [1]. Even more surprising, at low temperature four uranium compounds are both superconductors and magnetic at ambient pressure [2].

Although the 5\(f\) electrons of metallic uranium compounds are more easily treated in a localized magnetism framework, their hybridization with the conduction and ligand electrons can not be neglected [3]. Their ground state properties reflect the competition between at least four types of interactions of about the same strength: Coulomb and exchange, crystal field, hybridization and spin-orbit coupling. Due to the complexity of the physics involved, a complete understanding of an uranium compound has not yet been achieved.

In comparison to the vast theoretical literature on the 5\(f\) electronic properties, experimental microscopic information is scarce. It is usually obtained from photoemission, de Haas-Van Alphen measurements, neutron scattering and muon spin spectroscopy. With the advent of third generation synchrotron radiation sources, new experimental techniques have become available, such as x-ray magnetic circular dichroism (XMCD) [4]. This paper presents a discussion of published XMCD spectra recorded at the M\(_{4,5}\) edges of uranium atoms in metallics uranium compounds. Our interest here is to compare the XMCD spectra with the purpose to find relations between their characteristics and the electronic structure of the compounds.

### II. THE X-RAY MAGNETIC CIRCULAR DICHROISM DATA

We recall that the XMCD technique consists of recording two spectra at the absorption edges of a spin-orbit split core state chosen to probe the electronic state of interest. The two spectra differ by the handedness of the circularly polarized light used to record them. Information about the 5\(f\) uranium states can be obtained by performing measurements at the uranium M\(_{4,5}\) edges. The XMCD technique is known to provide information on the orbital and spin magnetic moments through the use of sum rules which only involve the integrated intensity of the absorption and dichroic spectra, i.e. they are independent of the shape of the XMCD spectra. However, measurements of twelve uranium compounds show that in fact the shape at the M\(_5\) edge depends strongly on the compound. This is illustrated by two examples in Fig. 1. The dichroism at the M\(_4\) edge consists always of a single negative lobe that has no distinct structure. On the other hand, two strong lobes, a positive and a negative one, or a single lobe, can be observed at the M\(_5\) edge. In fact, a double-lobe structure is detected for UPd\(_2\)Al\(_3\) [5], UBe\(_{13}\) and UPt\(_3\) [6], U\(_{0.3}\)La\(_{0.7}\)S and U\(_{0.4}\)La\(_{0.6}\)S [7] and UGe\(_2\) [8]. It does not exist (or the second lobe is very small compared to the first one) for the following six compounds: US [9,6,10], USb\(_{0.5}\)Te\(_{0.5}\) [11], UFe\(_2\) [12], URh\(_2\)Si\(_2\) [5], URhAl [13] and U\(_{0.6}\)La\(_{0.4}\)S [7]. XMCD measurements on UNi\(_2\)Al\(_3\) were performed but the signal intensity at the M\(_5\) was too small to determine its shape [14].

We quantify the shape of the XMCD spectra at the M\(_5\) edge in Table I by the ratio \(R_a\) of the algebraic area of the two lobes and the relative energy \(\Delta E\) at which the dichroism signal vanishes between them. The parameters introduced to characterize the XMCD response are...
III. IMPLICATIONS ON THE ELECTRONIC DENSITY OF STATES

Due to the selection rules for dipolar electronic transitions induced by light, the structure at the $M_5$ edge of the dichroic resonance provides information on the magnetic character of the density of states (DOS) above the Fermi level \[6,19\].

We recall that the $M_4$ ($M_5$) edge corresponds to $3d_{3/2}/3d_{5/2} \rightarrow 5f$ transitions. The $M_4$ absorption signal is proportional to the number of $f_{5/2}$ holes, while the $M_5$ absorption signal depends primarily on the number of $f_{7/2}$ holes. Since the XMCD technique uses circular polarized light, the dichroism contains information about the magnetic character of the sublevels in the DOS.

We first use the atomic picture with the $jj$-coupling. For both $M_4$ and $M_5$ edges, within the conventions adopted, the unoccupied sublevels above the Fermi level with negative magnetic quantum number $m_j$ give a positive dichrois signal and those with positive $m_j$ values a negative one. Qualitatively, it is expected that the two or three $f$ electrons (the valency of the compounds under interest is expected to be between $+4$ and $+3$) mainly occupy the sublevels with negative $m_{5/2}$, i.e. $-5/2$, $-3/2$ and $-1/2$, so that most of the hole density is in sublevels with positive $m_{5/2}$ values.

With this background, we expect to observe an essentially negative dichroic signal at the $M_4$ edge. Such a feature has been systematically observed for all uranium compounds.

We now consider the $M_5$ edge. The energy sequence of the $m_{7/2}$ sublevels is opposite to the $m_{5/2}$ one: the negative $m_{7/2}$ sublevels are located at higher energy relative to the positive $m_{7/2}$ sublevels. This reflects the gain in energy due to the alignment of the spin with the exchange field. Perturbations such as hybridization mix levels with same $m_j$ values. Crystal-field, Coulomb and exchange interactions mix levels of similar $m_s$ and $m_l$ values. Since, within the $j = 5/2$ levels, the negative $m_{5/2}$ sublevels are mostly occupied, it results that the negative $m_{3/2}$ sublevels are also preferably electron occupied. Therefore, a relatively strong negative lobe at low energy is expected and an eventual weak positive lobe at high energy is possible. These predictions of the atomic model provide a qualitative understanding of the results for the last six compounds of Table I. In the case where the energy splitting of the $m_{7/2}$ sublevels is small compared to the intrinsic width of the electronic transition (given by the core hole lifetime), the observation of a single lobe remains, of course, expected.

Now, for the first six compounds of Table I, we observe a redistribution of weight between the two lobes. This remarkable feature first implies that the energy splitting within the $m_{7/2}$ sublevels is large. The high-energy lobe is even the more intense lobe for the first three compounds. This means that the negative $m_{7/2}$ sublevels are less electron occupied than expected from the atomic $jj$-coupling point of view, i.e. the density of empty negative $m_{7/2}$ sublevels is larger than expected.

We have already pointed out that the compounds with a large $R_a$ ratio, i.e. with two lobes, have their $E_1$ shifted to an energy smaller than $E_0$. We do not have yet an understanding of this effect. Interestingly, we note that the majority of the spectral weight is at an energy smaller than $E_0$ for all the compounds, i.e. even for those exhibiting only one lobe. Probably, the observation of the two lobes and of the shift of $E_1$ reflect the same physics.

To understand the origin of the double-lobe, we may leave the $jj$-coupling scheme and work in the intermediate coupling scheme which allows a mixing of negative $m_{5/2}$ with positive $m_{7/2}$ sublevels. The breakdown of the $jj$-coupling scheme for the double-lobe compounds means that the hybridization, Coulomb and exchange and crystal field energies can no longer be taken as a perturbation relative to the $5f$ spin-orbit interaction for these compounds. Indeed, it has been shown that, for example, a crystal field of 1 eV or larger can lead to a double-lobe structure \[5\]. Although such a strong crystal field is not realistic, it shows that the breaking of the $jj$-coupling approximation leads to the double-lobe structure.

Instead of starting from the atomic picture, one may use a band-like approach as done by Shishidou and co-workers \[19\]. In fact, their computation for US yields a weak high-energy lobe as found experimentally. The band-like picture seems to be appropriate for the compounds with a double-lobe structure because of their large Sommerfeld coefficient which means that their density at the Fermi level is large. However, no matter the starting point of the description, i.e. the atomic or the band limit, our previous conclusion that the double-lobe structure is a signature of a relative large density of empty negative $m_{7/2}$ sublevels is a robust result since it arises basically from the selection rules for x-ray induced atomic transitions.
IV. CONCLUSION

Many experimental methods such as bulk techniques, neutron scattering and μSR spectroscopy have suggested that strongly electronically correlated uranium compounds can be thought of as systems with two components: conduction electrons and local moments. UPd$_2$Al$_3$ [20–24] seems to be the cleanest example with two localized 5f electrons per uranium in the U$^{4+}$ state responsible for the measured magnetic moment, while the remaining 5f electron density is itinerant. UBe$_{13}$ [25] and UPt$_3$ [26] are other two examples of the localized-itinerant duality, although for these two compounds there is no magnetic moment (UBe$_3$) or it is very small (UPt$_3$). Recently, UGe$_2$ has been found to be similar to UPd$_2$Al$_3$ [27]. The XMCD results suggest that the low-energy lobe arises from the U$^{4+}$ state under the influence of the crystal field [5] and the high-energy lobe is a signature of the itinerant component. Within this picture, the latter component has a strong $m_{7/2}$ character, in particular for UPd$_2$Al$_3$. Clearly, band structure computations are needed to test our suggestion.

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TABLE I. Specific heat and XMCD data for twelve uranium-based compounds. The values of the Sommerfeld parameter $\gamma$ are taken from the literature and the algebraic area ratio $R_a \equiv B/A$ and energy difference $\Delta E \equiv E_1 - E_0$ are estimated from published x-ray M$_5$ absorption and XMCD spectra. The parameters $A$, $B$, $E_0$ and $E_1$ are defined in Fig. 2. The compounds are classified by increasing values of $\Delta E$ and an horizontal line is used to distinguish the compounds with negative or zero $\Delta E$ value from the other compounds. We note that this order is also compatible, within the error bars, to increasing values of $R_a$. A question mark in the $\gamma$ column means that for the given compound we are not aware of any published value for this parameter. An hyphen in the $\Delta E$ column means that the value is irrelevant: when only one lobe is identified ($R_a = 0$) neither $E_1$ nor $\Delta E$ is defined.

| Compound     | Specific heat data | References | XMCD data | References |
|--------------|--------------------|------------|-----------|------------|
|              | $\gamma$(mJ.mol$^{-1}$.K$^{-2}$) | References | $R_a$ | $\Delta E$ (eV) | References |
| UPd$_2$Al$_3$ | 145                | [2]        | $-24$ (10) | $-0.8$ (2) | [5]        |
| UBe$_{13}$   | 1100               | [2]        | $-3.0$ (2) | $-0.5$ (2) | [6]        |
| UPt$_3$      | 450                | [2]        | $-2.2$ (5) | $-0.2$ (2) | [6]        |
| U$_{0.3}$La$_{0.7}$S | ?                 | -         | $-0.5$ (2) | $-0.2$ (1) | [7]        |
| U$_{0.4}$La$_{0.6}$S | ?                 | -         | $-0.4$ (2) | $-0.2$ (1) | [7]        |
| UGe$_2$      | 32                 | [15]       | $-0.54$ (4) | $0.1$ (1) | [8]        |
| U$_{0.6}$La$_{0.4}$S | 30                | [16]       | $-0.06$ (1) | $0.6$ (2) | [7]        |
| US           | 23                 | [17]       | $-0.04$ (1) | $0.7$ (2) | [6, 10]    |
| UFe$_2$      | 45                 | [18]       | $-0.06$ (3) | $0.8$ (2) | [12]       |
| USb$_{0.5}$Te$_{0.5}$ | ?                | -         | $0.0$ (1)  | -           | [11]       |
| URu$_2$Si$_2$ | 65                 | [2]        | $0.00$ (1) | -           | [5]        |
| URhAl        | 76                 | [13]       | $0.00$ (2) | -           | [13]       |
FIG. 1. Examples of $M_{4,5}$ absorption spectra and corresponding dichroism [6]. The dichroic spectra are obtained by simple difference of the absorption spectra without any further data manipulation.
FIG. 2. Drawing used to define the physical quantities needed to quantify the XMCD spectra at the M₅ edge. $E₀$ is the energy at which the absorption is maximum, $E₁$ the energy where the XMCD response vanishes between the two lobes, A the algebraic area of the lobe located below $E₁$ and B the algebraic area of the lobe appearing above $E₁$. 