ELSEVIER

Journal of Alloys and Compounds 356–357 (2003) 377–381

Journal of ALLOYS AND COMPOUNDS

www.elsevier.com/locate/jallcom

# μSR experiment on $YMn_2D_{0.5}$ and $YMn_2D_3$ deuterides

M. Latroche[a,*], V. Paul-Boncour[a], P. Mietniowski[b], H. Figiel[b], A. Percheron-Guégan[a]

[a]*Laboratoire de Chimie Métallurgique des Terres Rares, UPR209, IFSCSA, CNRS, 2 Rue H. Dunant, 94320 Thiais Cedex, France*

[b]*Department of Solid State Physics, Faculty of Physics and Nuclear Techniques, University of Mining and Metallurgy, Mickiewicza 30, 30-059 Krakow, Poland*

Received 5 July 2002; accepted 26 October 2002

---

## Abstract

The Laves phase $YMn_2$ exhibits a complex helimagnetic structure, which can be transformed by applying external pressure, chemical substitution or hydrogen absorption. The hydrides $YMn_2H_x$ can be obtained in a cubic structure above $T_{C,N}$ with a cell parameter increase up to $x=4.3$ H/f.u. For $x\leq3.4$ H/f.u., both magnetic and hydrogen orderings appear below $T_c$. These transitions have been studied by μSR experiments in zero field and longitudinal geometry and in a transverse field to study spin fluctuations for $x=0.5$ and 3 D/f.u. down to 20 K. The correlation between the critical parameter $\gamma$ and the deuterium concentration will be shown.

*Keywords:* Intermetallics; Magnetically ordered materials; E-muons spectroscopy

---

## 1. Introduction

The Laves phase $YMn_2$ exhibits a complex helimagnetic structure which has been widely studied [1–3]. Cell parameter modification either by applying pressure [4], by substituting Y or Mn sites by other elements [5] or by hydrogen absorption can easily change the magnetic properties. Single phase hydrides $YMn_2H_x$ exists in the range $0<x\leq4.3$ H/f.u. [6] and the cubic structure is preserved above $T_{C,N}$ for the whole hydrogen concentration range. For $x\leq3.4$ D/f.u., the $YMn_2$–$H_2$ system behaves like a solid solution above $T_c$ [7]. Below $T_c$, hydrogen absorption leads to a ferrimagnetic ground state in which both Curie temperature and Mn moment increase with hydrogen [8]. For $x=0.5$, a temperature-dependent X-ray diffraction (XRD) study [9] has indicated a tetragonal distortion below 250 K followed by an inversion of the $c/a$ ratio at 100 K associated to magnetic transitions observed by magnetization measurements [10]. For $x=1$, correlation exists between deuterium and magnetic orderings around 220 K [11,12]. Coupling between magnetic and hydrogen structures is a general feature for this system and was also observed for $x=4.3$ [13].

Zero and longitudinal field muon spin relaxation (μSR) techniques have been used to study local magnetic properties and spin fluctuations in $YMn_2D$ and $YMn_2D_2$ between 10 and 300 K [14]. The asymmetry parameter indicates a first-order transition to ferrimagnetism at $T_c=$ 220 and 260 K for $YMn_2D$ and $YMn_2D_2$, respectively. In contrast the depolarisation rate $\lambda(T)$ exhibits a critical divergence as $T_c$ is approached from above. These features are consistent with those observed for the parent compound $YMn_2$ [15], and indicate that the spin fluctuations character in the paramagnetic state changes slightly with deuterium loading, despite increase of the magnetic ordering temperature. The $\lambda(T)$ dependence provides evidence of significant spin fluctuations below and above $T_c$, while slight deviations from predicted behaviour indicate further structural or magnetic transitions in the ordered state.

Following these previous μSR measurements, experiments were carried out in zero field and longitudinal geometry and in transverse field to study magnetic behaviour and spin fluctuations in the compounds $YMn_2D_{0.5}$ and $YMn_2D_3$ between 20 and 400 K. The aim of this experiment was to follow the variations of the asymmetry parameter and relaxation rate in the temperature range above and below the magnetic ordering temperatures and to compare their behaviour to that of the previous investigated samples $YMn_2D$ and $YMn_2D_2$.

---

*Corresponding author.

*E-mail address:* latroche@glvt-cnrs.fr (M. Latroche).

0925-8388/02/$ – see front matter © 2002 Elsevier B.V. All rights reserved.
doi:10.1016/S0925-8388(02)01232-X

## 2. Experimental details

Elaboration and characterization of the deuterides has been already described in previous papers [6,7]. The sample preparation and experimental set up were also given in detail in Ref. [14].

Experiments have been carried out on the muon beam line of the ISIS facility on the instrument MuSR. Sample environment consists of Helmotz coils allowing longitudinal and transverse fields from 0 to 200 mT and of a closed cycle refrigerator ranging from 15 to 400 K.

Both samples have been studied in zero field longitudinal geometry (ZF) and in a transverse field (TF) of 25 mT for $x=0.5$ D/f.u. For this latter compound, the temperature range varies from 20 to 300 K, whereas for $x=3.0$ D/f.u., according to the higher transition temperature observed for that composition (around 330 K) the sample was studied from 20 to 400 K.

Time histograms of the positron count rates observed in the backward and forward detectors at each temperature were grouped according to:

$$R(t)=\frac{N_F(t)-N_B(t)}{N_F(t)+N_B(t)}=a_0G_z(t) \tag{1}$$

where $G_z(t)$ is the longitudinal relaxation function. For $x=0.5$ D/f.u., the data were refined by a least-squares procedure using a single exponential relaxation function indicative of temporal fluctuations of the muons in the magnetic environment. For the sample with $x=3.0$ D/f.u., two components were observed in the histograms and $R(t)$ was obtained using two Lorentzian relaxation rates $\lambda_1$ and $\lambda_2$. A constant background **b** representative of the sample environment was also added to the function which was therefore written as:

$$R(t)=\sum_i[a_{oi}\exp(-\lambda_i t)]+b \quad i:[1 \text{ or } 2] \tag{2}$$

For the transverse field geometry, the count rates of the backward detector have been refined using the following function:

$$N(t)=N_o\exp(-t/\tau)[1+a_o\exp(-\lambda t)\cdot\cos(2\pi ft-\phi)] \tag{3}$$

where $N_o$ is the initial count rate at $t=0$, $\tau$ represents the muon lifetime (2.197 μs), $\lambda$ is the relaxation rate, $f$ the Larmor frequency of the muon spin and $\phi$ stands for the phase. From the measurement of the frequency $f$, one can estimate the local field at the muon site from the following formula:

$$B=\frac{2\pi f}{\gamma} \tag{4}$$

where $\gamma$ is the muon gyromagnetic ratio.

## 3. Results and discussion

### 3.1. $YMn_2D_{0.5}$

Typical μSR spectrum at 300 K is shown in Fig. 1. ZF data have been refined according to Eqs. (1) and (2) with $i=1$. The temperature dependence of the asymmetry $a_o$ and of the depolarisation rate $\lambda$ are shown in Fig. 2a,b, respectively. The asymmetry $a_o$ is very small at low temperatures ($<0.05$) and begins to rise when approaching the transition to recover the full asymmetry at 300 K. Concerning $\lambda(T)$, two sharp maxima at ~100 and 240 K are observed.

As already mentioned, magnetic ordering of antiferromagnetic type appears for $YMn_2H_{0.5}$ at 240 K and a magnetic structure change occurs below 100 K. These facts are in good agreement with the observed temperature changes of $a_o$ and $\lambda$. The drop of the asymmetry level at 100 K (Fig. 2a) can be related with the change of magnetic order and a sharp increase of $a_o$ above 250 K reflects the transition to the paramagnetic state. In Fig. 2b, the divergence of $\lambda(T)$ at 240 K is characteristic for the magnetic ordering temperature [16]. However, for lower

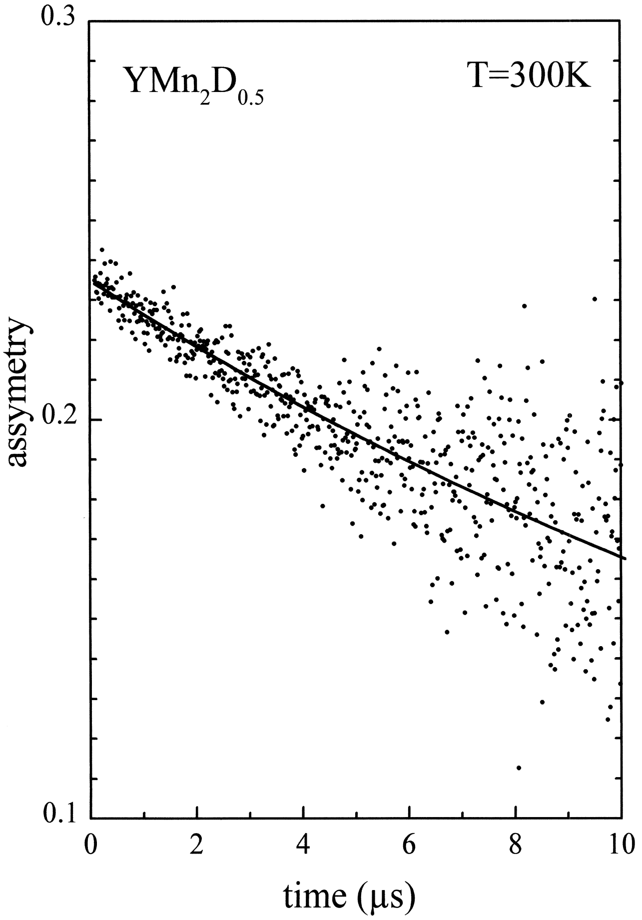

Fig. 1. μSR spectrum at 300 K for $YMn_2D_{0.5}$. The solid line represents a fit of Eq. (2) for $i=1$.

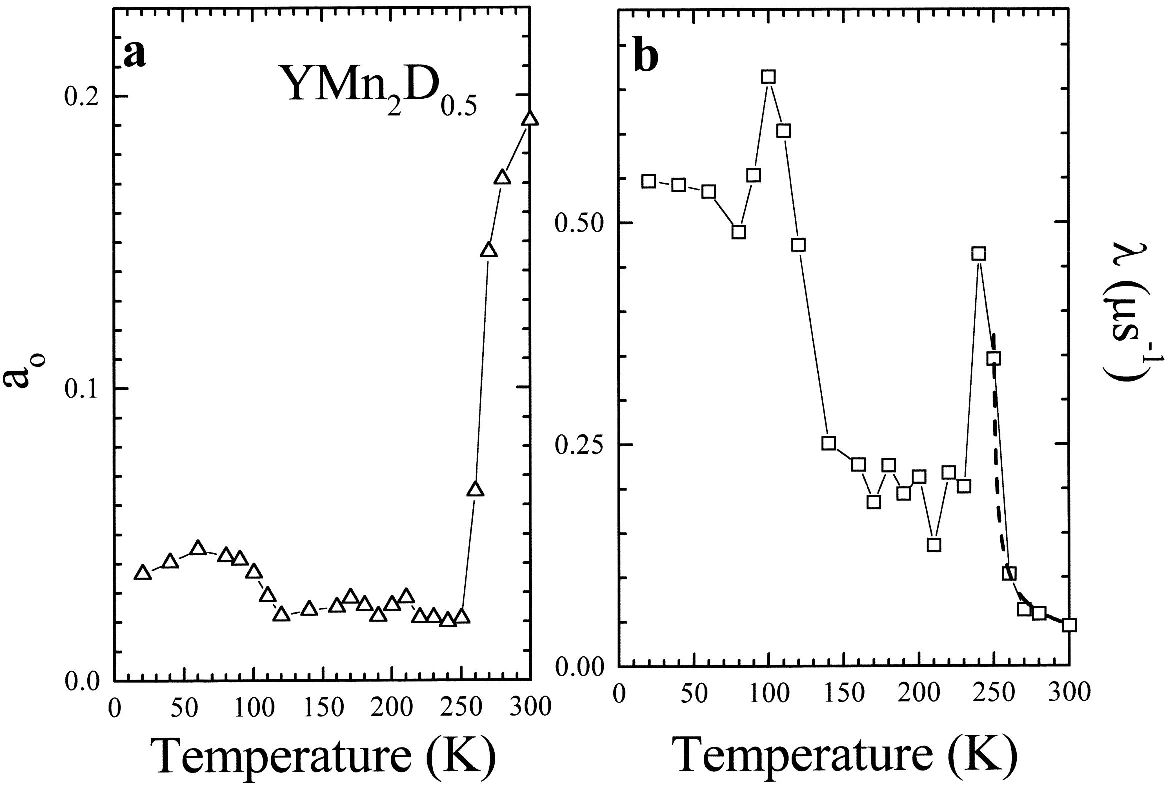

Fig. 2. Temperature dependence of (a) the asymmetry parameter, $a_o$, and (b) the depolarisation rate, $\lambda(T)$, for $YMn_2D_{0.5}$. The broken line represents a fit of Eq. (5).

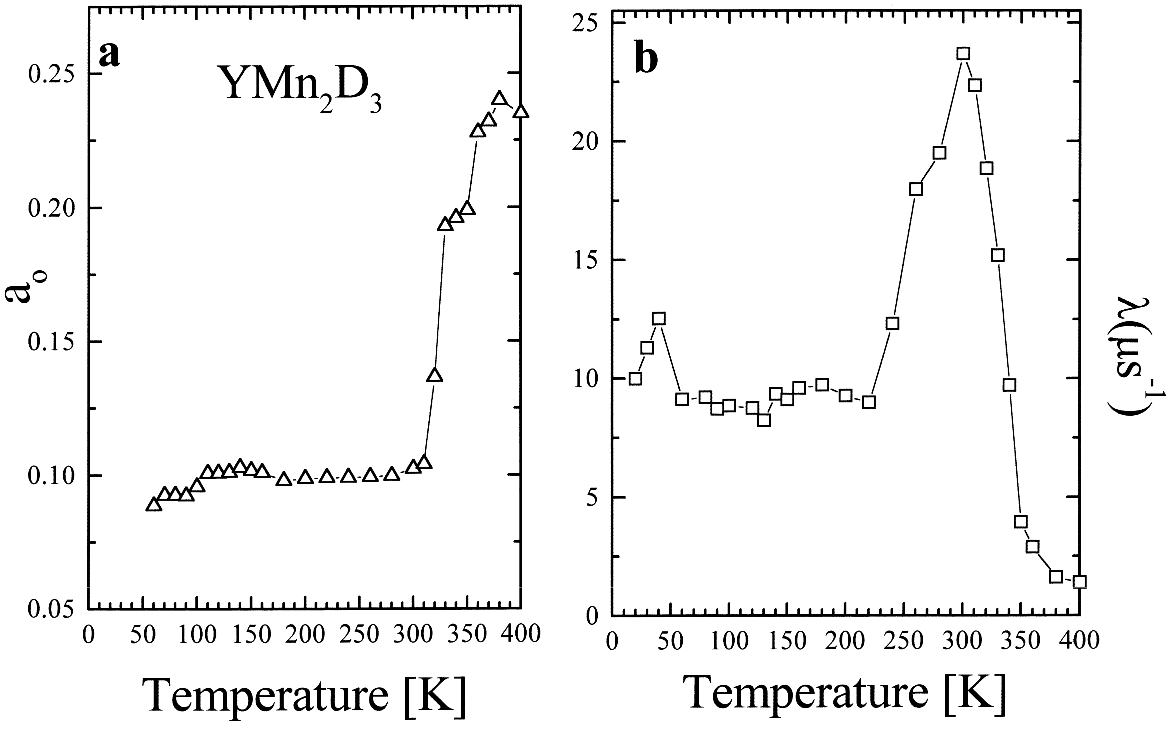

Fig. 3. Temperature dependence of (a) the sum of the two components of the asymmetry parameter, $a_o$, and (b) the second component of the depolarisation rate, $\lambda_2(T)$, for $YMn_2D_3$.

temperatures an increase of relaxation rate as a function of temperature is associated with a clear maximum at 100 K. This was not observed in samples with other concentrations (Fig. 3b for $x=3$ and [14] for $x=1$ and 2) and it corresponds exactly to the transition toward a second crystallographic and magnetic structure. In the paramagnetic range (i.e. above 240 K), the critical divergence of $\lambda$ has been described according to the following equation:

$$\lambda(t) = \lambda_{\mathrm{o}} \left[ \frac{T - T_{\mathrm{c}}}{T_{\mathrm{c}}} \right]^{-\gamma} \tag{5}$$

and the corresponding refined curve is represented in Fig. 2b as a dotted line. Results of the refinement are given in Table 1.

For the TF experiment, the data have been refined according to Eq. (3). In the whole range of temperature, the Larmor frequency fluctuates within the error bar around the constant value both in the magnetically ordered and paramagnetic state. The average value of the refined frequencies (3.402(4) MHz) is in good agreement with the expected one (3.388 MHz) for an external field set to 250 G. This means, that at the paramagnetic range above 240 K the Knight shift is close to zero, and at the magnetically ordered state a mean local magnetic field acting on muons is also zero.

### 3.2. $YMn_2D_3$

All histograms have been refined assuming two components for the exponential function. The asymmetry parameter $a_{\mathrm{o}}$ treated as the sum of these two components (Fig. 3a), drops at the ordering temperature to about one-third of its initial value (0.25) showing behaviour similar to hydrides with $x=1$ and 2 [14]. The relaxation rate of the refined component $\lambda_1$ is in the range 0–0.10 $\mu s^{-1}$ much smaller than the $\lambda_2$ reaching up to 25 $\mu s^{-1}$ as a function of temperature and passes maximum below the magnetic ordering temperature, very similar to those reported for previous samples [14]. The temperature dependence of this second component $\lambda_2$ presented in Fig. 3b is in good correlation with temperature changes of magnetisation [8] and unit cell parameters [9]. The critical parameter $\gamma$ reaches 1.18, a value higher than that for $x=2$ (0.87).

To find out if the nuclear and electronic components in the relaxation process may be decoupled, the measurements in longitudinal fields 5, 50 100 and 200 mT at 60 K (in the magnetically ordered state), 300 K (just below the ordering temperature $T_{\mathrm{c}}$) and 350 K (above $T_{\mathrm{c}}$) were performed. The obtained experimental results for both components gave no evidence of any field dependence which might be interpreted in terms of existing theoretical descriptions.

Table 1
Critical scaling analysis parameters of the muon depolarisation rate, $\lambda(T)$, according to Eq. (3) for $YMn_2D_x$ compounds

| $x$ in $YMn_2D_x$ | Refs. | $\lambda_{\mathrm{o}}$ ($\mu s^{-1}$) | $T_{\mathrm{c}}$ (K) | $\gamma$ |
|---|---|---|---|---|
| 0.0 | [3] | 0.033 | 93–113 | 0.35 |
| 0.5 | | 0.020(4) | 249(3) | 0.53(5) |
| 1.0 | [14] | 0.0307(8) | 219(4) | 0.64(1) |
| 2.0 | [14] | 0.0318(9) | 258.6(1) | 0.87(1) |
| 3.0 | | – | – | 1.18(1) |

Critical parameter $\gamma$ in Table 1 has been collected for $x=0$, 0.5, 1, 2 and 3 (Refs. [3,14] and this work). The increase of $\gamma$ with deuterium content supports our conclusion from [14], that the critical parameter is sensitive to the deuterium concentration due to the wave vector-dependent susceptibility $\chi(q)$ and $q$ dependent width of the spin-fluctuation excitation spectrum. The same dependence of the critical parameter on $x$ has been observed for the NMR relaxation rates of deuterium in $YMn_2D_x$ [16].

## 4. Conclusions

For the compound with low deuterium concentration ($x=0.5$ D/f.u.), the asymmetry parameter and the relaxation rate reflect the magnetic transformations within the magnetically ordered state. No changes in the frequency were observed and this latter parameter remains constant at a value very close to the one expected from a 250-G transverse field. For the deuterium-rich sample ($x=3$ D/f.u.) two relaxation processes have been observed below $T_{\mathrm{c}}$ and only refining of the relaxation rate above $T_{\mathrm{c}}$ was possible. The critical parameter $\gamma$ increases with $x$ consistently with those reported for $x=1$ and 2. The full asymmetry is recovered above $T_{\mathrm{c}}$ for both samples, proving transitions toward purely paramagnetic state.

## References

[1] T. Freltoft, P. Böni, G. Shirane, K. Motoya, Phys. Rev. B 37 (1988) 3454.
[2] A. Ballou, J. Desportes, R. Lemaire, Y. Nakamura, B. Ouladdiaf, J. Magn. Magn. Mater. 70 (1987) 129.
[3] R. Cywinski, S.H. Kilcoyne, C.A. Scott, J. Phys.: Condens. Matter 3 (1991) 6473.
[4] R. Hauser, E. Bauer, E. Gratz, Th. Häufler, G. Hilscher, G. Wiesinger, Phys. Rev. B 50 (1994) 13493.
[5] M. Shiga, H. Wada, H. Nakamura, K. Yoshimura, Y. Nakamura, J. Phys. F 17 (1987) 1781.
[6] J. Przewoznik, V. Paul-Boncour, M. Latroche, A. Percheron-Guégan, J. Alloys Comp. 225 (1995) 436–439.
[7] M. Latroche, V. Paul-Boncour, J. Przewoznik, A. Percheron-Guégan, F. Bourée-Vigneron, J. Alloys Comp. 231 (1995) 99–103.
[8] H. Fuji, M. Saga, T. Okamoto, J. Less Common Met. 130 (1987) 25.
[9] H. Figiel, A. Lindbaum, Cz. Kapusta, E. Gratz, J. Alloys Comp. 217 (1995) 157.
[10] H. Figiel, J. Przewoznik, V. Paul-Boncour, A. Lindbaum, E. Gratz, M. Latroche, M. Escorne, A. Percheron-Guégan, J. Alloys Comp. 274 (1998) 29.

[11] M. Latroche, V. Paul-Boncour, A. Percheron-Guégan, F. Bourée-Vigneron, G. André, Physica B 234–236 (1997) 599–601.
[12] M. Latroche, V. Paul-Boncour, A. Percheron-Guégan, F. Bourée-Vigneron, G. André, J. Solid State Chem. 154 (2000) 398–404.
[13] I.N. Goncharenko, I. Mirebeau, A.V. Irodova, E. Suard, Phys. Rev. B 56 (5) (1997) 2580–2584.
[14] M. Latroche, H. Figiel, G. Wiesinger, Cz. Kapusta, P. Mietniowski, V. Paul-Boncour, A. Percheron-Guegan, R. Cywinski, J. Phys. C (Condens. Matter) 8 (1996) 4603.
[15] S.W. Lovesey, E.B. Karssson, K.N. Trohidu, J. Phys.: Condens. Matter 4 (1992) 2043.
[16] H. Figiel, A. Budziak, P. Mietniowski, M.T. Kelemen, E. Dormann, Phys. Rev. B. 63 (2001) 204403.