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## <sup>170</sup>Yb Mössbauer study of YbPt<sub>3</sub> and Yb<sub>3</sub>Pt<sub>5</sub>

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**Abstract.** The intermetallic compounds YbPt<sub>3</sub> and Yb<sub>3</sub>Pt<sub>5</sub>, which present magnetic ordering below 0.25 K and 0.95 K respectively, have been investigated by <sup>170</sup>Yb Mössbauer spectroscopy at very low temperatures. In YbPt<sub>3</sub>, the electronic ground Kramers doublet of Yb<sup>3+</sup> has been determined, as well as its exchange splitting and spin–spin relaxation rate; the lowest Yb<sup>3+</sup> crystal field excitation has been estimated. The surprising finding that the crystal electric field has strong axial character in YbPt<sub>3</sub>, instead of the expected cubic symmetry, is discussed.

### 1. Introduction

Among the rare earth (RE) intermetallic compounds, materials containing Ce or Yb are known to exhibit various degrees of hybridisation between the 4f electrons and the band electrons. In the case where hybridisation is weak, the valence of the Ce or Yb ion is very close to 3 and some bulk properties, such as magnetic susceptibility or x-ray L<sub>III</sub> edge absorption spectra, present slight anomalies reflecting the non-integral valence of the RE ion. At low temperature, the physical properties of these weakly hybridised systems are dominated by the competition between the exchange coupling between rare earth ions and the local demagnetisation due to hybridisation. Magnetic ordering, when it appears, can be strongly frustrated, as was demonstrated in the Yb monopnictides [1]. It is thus of interest to investigate the low temperature magnetic properties of Ce and Yb intermetallics. As Mössbauer spectroscopy can be carried out on Yb, this provides unique microscopic information in these systems [2].

In the Pt rich Yb–Pt alloys, which have been previously characterised and investigated by a number of bulk measurements [3], the Yb ion is found to be trivalent. In YbPt<sub>3</sub>, one observes a weak lattice constant anomaly with respect to the heavy RE REPt<sub>3</sub> compounds and an effective high temperature magnetic moment reduced from the free ion value [3]. This suggests that the Yb valence is slightly lower than 3. In Yb<sub>3</sub>Pt<sub>5</sub>, x-ray L<sub>III</sub> edge absorption spectra yield a valence of 2.95 for Yb [4]. At low temperature, the behaviours of the magnetic susceptibility and of the electrical resistivity indicate the presence of a magnetic transition near 0.25 K in YbPt<sub>3</sub> and 0.95 K in Yb<sub>3</sub>Pt<sub>5</sub>. These two compounds thus appear to be weakly hybridised magnetic Yb materials.

We report here on an x-ray powder study of YbPt<sub>3</sub> at 10 K and room temperature and on a <sup>170</sup>Yb Mössbauer study of the two compounds in the temperature range

$0.04 \text{ K} \leq T \leq 20 \text{ K}$ . We confirm the onset of magnetic ordering below the above-mentioned temperatures in both compounds. In  $\text{YbPt}_3$  we find that the Yb site symmetry has strong axial character and that the ground Kramers doublet has extreme magnetic anisotropy, despite the fact that the lattice has cubic symmetry (AuCu<sub>3</sub> type). In this compound we could measure the exchange field splitting and the spin–spin relaxation rate of  $\text{Yb}^{3+}$ , and estimate the lowest crystal field excitation. In  $\text{Yb}_3\text{Pt}_5$  a detailed analysis could not be made due to the greater number of parameters introduced by the very low site symmetry of Yb.

## 2. Experimental details

The  $^{170}\text{Yb}$  Mössbauer transition takes place between the nuclear ground state with spin  $I_g = 0$  and the first excited state with spin  $I_e = 2$ . The transition energy is 84.3 keV and the Mössbauer velocity unit of  $1 \text{ mm s}^{-1}$  corresponds to a frequency of 68 MHz. The spectra were recorded using a triangular Doppler velocity signal. The  $\gamma$ -ray source was neutron-activated  $\text{TmB}_{12}$  whose linewidth (FWHM) versus an  $\text{YbAl}_3$  reference absorber was  $2.8 \text{ mm s}^{-1}$ . The spectra below 1.4 K were recorded in a  $^3\text{He}$ – $^4\text{He}$  dilution refrigerator.

A general feature of the Mössbauer experiments in these Pt rich alloys is the relatively low statistics of the spectra. This is due on the one hand to the high electronic absorption of the 84.3 keV  $\gamma$ -rays by Pt, and on the other hand to the relatively small value of the Debye temperature of these alloys:  $\theta_D = 130 \text{ K}$  [3].

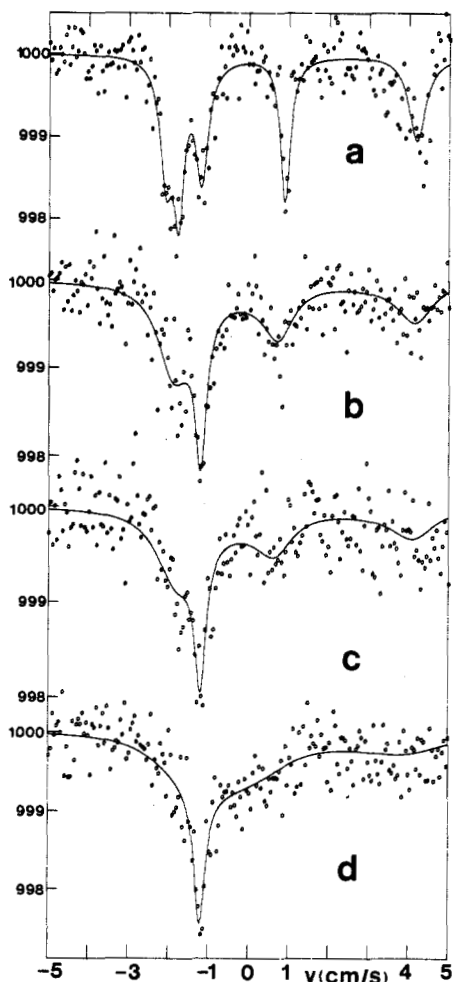
## 3. $\text{YbPt}_3$

### 3.1. X-ray investigation

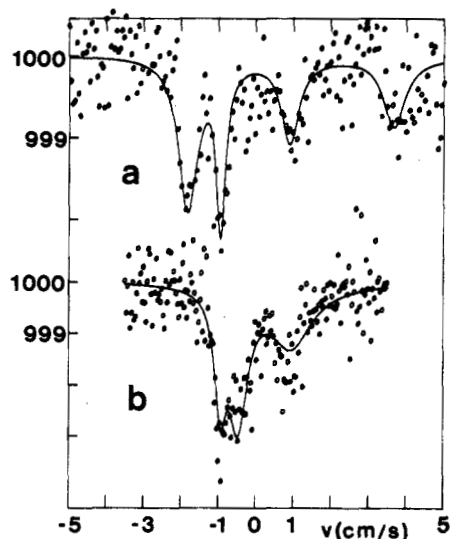
The preparation and characterisation of our polycrystalline  $\text{YbPt}_3$  sample has been described in [3], where it was found to have a chemical formula  $\text{YbPt}_{3.26}$ . In order to investigate more closely the departure from the stoichiometry, the ordering and the symmetry of the lattice, we performed x-ray intensity measurements at 10 K and at room temperature. First of all, we found that our sample contained a small quantity of Pt metal. Intensity calculations assuming an overall 1 : 3.26 atomic ratio between Yb and Pt contained in the sample yielded a chemical formula for the alloy which was quite close to  $\text{YbPt}_3$ . The x-ray powder spectra of  $\text{YbPt}_3$  were characteristic of the AuCu<sub>3</sub> structure with a cubic lattice parameter  $a = 4.0820(5) \text{ \AA}$ ; the (110) superstructure line was detected, which shows that the alloy presents a good degree of order of the Pt and Yb atoms in the AuCu<sub>3</sub> type structure. The spectra at 10 K and at room temperature are very similar, indicating the absence of any structural transition: the lattice remains cubic down to low temperature, at least on average. Our x-ray analysis has thus demonstrated that our sample is a crystallographically ordered, close to stoichiometric  $\text{YbPt}_3$  alloy.

### 3.2. Mössbauer spectrum at $T = 0.040 \text{ K}$

The spectrum obtained at  $T = 0.040 \text{ K}$  is represented in figure 1(a). It is an effective field hyperfine spectrum of  $^{170}\text{Yb}^{3+}$ , which can be accounted for by an axially symmetric



**Figure 1.** <sup>170</sup>Yb Mössbauer absorption spectra in YbPt<sub>3</sub>. (a):  $T = 0.04$  K; (b):  $T = 0.185$  K; (c):  $T = 0.23$  K; (d):  $T = 0.45$  K. The fits (full curves) are explained in the text.



**Figure 2.** <sup>170</sup>Yb Mössbauer absorption spectra in Yb<sub>3</sub>Pt<sub>5</sub>. (a):  $T = 0.04$  K; (b):  $T = 4.2$  K. The fits (full curves) are explained in the text.

Hamiltonian, the  $z$  axis being the common direction of the hyperfine field  $H_{\text{hf}}$  and of the principal axis of the electric field gradient (EFG) at the Yb site

$$\mathcal{H}_{\text{hf}} = -g_n \mu_n \mathbf{I}_c \cdot \mathbf{H}_{\text{hf}} + \alpha_Q (I_z^2 - 2). \quad (1)$$

The fitted values are  $H_{\text{hf}} = (3980 \pm 50)$  kOe for the hyperfine field and  $\alpha_Q = eQV_{zz}/8 = 6 \pm 0.1 \text{ mm s}^{-1}$  for the hyperfine quadrupolar constant ( $Q$  is the nuclear excited state quadrupole moment and  $V_{zz}$  the main component of the EFG tensor). The spontaneous electronic moment value derived from  $H_{\text{hf}}$  (using the conversion factor of  $1043 \text{ kOe } \mu_B^{-1}$ ) is:  $\mu = (3.82 \pm 0.05) \mu_B$ . The magnetic moment and the quadrupole constant measured here are, to our knowledge, the biggest ever observed in an Yb compound. They are very close to the maximum possible values of the magnetic moment and quadrupole constant expected for a  $|J_z = \pm \frac{7}{2}\rangle$  Kramers doublet:  $4\mu_B$  and  $5.8 \text{ mm s}^{-1}$  respectively.

The small excess value ( $0.2 \text{ mm s}^{-1}$ ) of the measured quadrupole constant over  $5.8 \text{ mm s}^{-1}$  must be assigned to a lattice contribution of the EFG.

The spectrum lines have a mean width (FWHM) of  $3.8 \text{ mm s}^{-1}$ . The individual lines have inhomogeneous widths ranging from 3 to  $5 \text{ mm s}^{-1}$ , while the minimal experimental linewidth is  $2.8 \text{ mm s}^{-1}$ . The excess broadening, together with the greater linewidth of the  $\Delta I_z = \pm 2$  line, are the hallmark of a small distribution of hyperfine fields among the Yb sites due to the natural inhomogeneity of the sample.

The ground doublet of  $\text{Yb}^{3+}$  in  $\text{YbPt}_3$  is thus a quasi-pure  $|J_z = \pm \frac{7}{2}\rangle$  state with  $g_z \approx 7.64$ , which is quite surprising in a material with a cubic  $\text{AuCu}_3$ -like structure. Indeed, the  $\text{Yb}^{3+}$  wave functions occurring in a cubic crystal electric field (CEF) are the isotropic  $\Gamma_6$ ,  $\Gamma_7$ , or  $\Gamma_8$  states, whereas the extremely anisotropic ( $g_z = 8$ ,  $g_\perp = 0$ )  $|J_z = \pm \frac{7}{2}\rangle$  state is the ground doublet in the presence of a dominant axial crystal field term  $O_2^0 = B_2^0 J_z^2$  with  $B_2^0 < 0$ . However, a  $|J_z = \pm \frac{7}{2}\rangle$  doublet can be the ground state in the presence of a small axial distortion with respect to cubic symmetry in the particular case of an accidental degeneracy between the cubic eigenstates  $\Gamma_6$  and  $\Gamma_8$  or  $\Gamma_7$  and  $\Gamma_8$ . This point will be further discussed in section 3.5.

We pointed out that the spectrum at  $T = 0.040 \text{ K}$  could also be fitted to a paramagnetic slow relaxation hyperfine Hamiltonian with an extremely anisotropic hyperfine **A** tensor ( $A_z = 3.85 g_z \text{ mm s}^{-1} = 30.8 \text{ mm s}^{-1}$ ,  $A_\perp = 0$ )

$$\mathcal{H}_{\text{hf}} = A_z I_z S_z + \alpha_Q (I_z^2 - 2) \quad (2)$$

where  $S = \frac{1}{2}$  is the effective spin of the doublet state. Hamiltonian (2) is formally identical to Hamiltonian (1), but it describes the fundamentally different physical situation of a paramagnetic degenerate Kramers doublet and it is thus relevant only in the paramagnetic phase.

As temperature increases above  $0.04 \text{ K}$ , the hyperfine splitting remains resolved up to  $0.23 \text{ K}$  with, however, significant line broadening. At  $0.275 \text{ K}$  we observe that the magnetic structure is almost entirely smeared out; between  $0.275 \text{ K}$  and  $15 \text{ K}$ , the spectra remain practically unchanged. We shall first analyse the spectra above  $0.275 \text{ K}$  which, in agreement with [3], correspond to the paramagnetic phase. We shall then come back to the spectra in the magnetic phase.

### 3.3. Mössbauer spectra in the range $0.275 \text{ K} \leq T \leq 20 \text{ K}$

The spectra in this temperature range show practically no thermal variation up to  $15 \text{ K}$ . The spectrum at  $T = 0.45 \text{ K}$  is represented on figure 1(d). It is a paramagnetic hyperfine spectrum where the  $\text{Yb}^{3+}$  electronic relaxation rate is of the same order of magnitude as the hyperfine frequency ('intermediate relaxation' spectrum). As the electronic ground state is very close to the  $|J_z = \pm \frac{7}{2}\rangle$  doublet, the spectra in this temperature range were fitted to a relaxational lineshape based on Hamiltonian (2), which involves only one dynamical parameter: the relaxation rate  $1/T_1$  between the two degenerate states of the ground doublet. This lineshape takes the following very simple form:

$$I(p) = \text{Re} \sum_{m=0, \pm 1, \pm 2} \frac{p + 1/T_1 - iE_Q^m}{[p - i(m(A_z/2) + E_Q^m)][p - i(-m(A_z/2) + E_Q^m)] + (1/T_1)(p - iE_Q^m)} \quad (3)$$

where  $p = \Gamma/2 - i\omega$ ,  $\Gamma$  being the experimental linewidth; the hyperfine parameters are the quadrupolar energy  $E_Q^m = \alpha_Q(m^2 - 2)$  and  $A_z$ , the only non-zero component of the magnetic hyperfine tensor. In the curve fits,  $A_z$  was fixed at the value  $30.80 \text{ mm s}^{-1}$  appropriate for the  $|J_z = \pm \frac{7}{2}\rangle$  state and the linewidth  $\Gamma$  was fixed at  $3.8 \text{ mm s}^{-1}$  which is an estimation of the static experimental linewidth on the  $T = 0.04 \text{ K}$  spectrum. The two parameters  $\alpha_Q$  and  $1/T_1$  were left free. The lineshape (3) accounts very well for the spectra up to  $15 \text{ K}$ , and the fits yield a constant value of  $6 \text{ mm s}^{-1}$  for  $\alpha_Q$  up to  $10 \text{ K}$ . At  $15 \text{ K}$ ,  $\alpha_Q$  has slightly decreased to  $5.65 \text{ mm s}^{-1}$ . The constant  $\alpha_Q$  is very precisely determined by the position of the narrow  $\Delta I_z = 0$  line which is unaffected by relaxation within the ground state doublet. The relaxation rate  $1/T_1$ , which is determined by the overall spectral shape, is found to be temperature independent up to  $15 \text{ K}$ , although its measurement accuracy is reduced due to the relatively large dispersion of the experimental data. We find:  $1/T_1 = (1.45 \pm 0.35) \text{ GHz}$ , which is to be compared with  $A_z = 2.0 \text{ GHz}$ .

The spectrum recorded at  $T = 20 \text{ K}$  is different from the  $15 \text{ K}$  spectrum, and cannot be fitted with a lineshape involving the ground Kramers doublet alone. We think that it contains contributions from excited crystal field levels. Tentative fits using a lineshape involving spin–spin relaxation between the ground state  $|J_z = \pm \frac{7}{2}\rangle$  and an assumed  $|J_z = \pm \frac{5}{2}\rangle$  excited state are relatively satisfactory and suggest that the excited crystal field state lies at near  $30 \text{ K}$  above the ground state. The relaxation rate within the ground doublet at  $20 \text{ K}$  is found to be twice that at  $15 \text{ K}$ ; this increase is possibly due to the onset of phonon driven relaxation.

Two conclusions can be drawn from these results: first, the weak thermal variation of the quadrupolar constant  $\alpha_Q$  between  $0$  and  $15 \text{ K}$  shows that the first excited crystal field state lies higher than  $15 \text{ K}$  from the ground state, which agrees with the results of the lineshape analysis of the  $T = 20 \text{ K}$  spectrum; second, the absence of any thermal variation of the relaxation rate  $1/T_1$  up to  $15 \text{ K}$  implies that relaxation in this temperature range is driven by spin–spin interactions (RKKY exchange or dipolar interactions) rather than by phonons ( $1/T_1 \propto T^9$ ) or by exchange with conduction electrons ( $1/T_1 \propto T$ ).

Indeed, as long as the excited crystal field states are negligibly populated, spin–spin relaxation consists of mutual ‘flip-flops’ within the ground doublet between neighbouring ions and the relaxation rate is temperature independent. However, the ‘flip-flop’ transition probability involves the quantity  $g_{\perp}$ , which should be very small in our case due to the extremely anisotropic character of the ground doublet. The observation of a sizable relaxation rate at very low temperature is thus surprising; this point will be further discussed in section 3.5.

The spectra observed in the magnetic phase ( $T \leq 0.23 \text{ K}$ ) will now be interpreted in terms of the above relaxation analysis.

### 3.4. Mössbauer spectra in the magnetically ordered phase ( $T \leq 0.23 \text{ K}$ )

The spectrum at  $T = 0.15 \text{ K}$  is very similar to that at  $0.04 \text{ K}$ . The spectra at  $T = 0.185 \text{ K}$  and  $0.23 \text{ K}$  are represented in figures 1(b)–(c). The individual lines are still resolved but are broadened, except for the narrow line corresponding to the  $\Delta I_z = 0$  transition. The most straightforward and coherent interpretation of these spectra is the following: each  $\text{Yb}^{3+}$  magnetic moment is fluctuating between the two exchange split levels of the ground Kramers doublet, the fluctuations being due to RKKY or dipole–dipole interactions. The line broadening therefore has a dynamical character. We are dealing here with the rather uncommon situation where the magnetic relaxation rate can be measured in the ordered

phase. This occurs because the relaxation rate is comparable with the hyperfine line spacing.

The fits shown on figures 1(b)–(c) were performed using the relaxation lineshape of [5], which reduces to lineshape (3) when the exchange splitting vanishes. The hyperfine parameters  $A_z$  and  $\alpha_Q$  were kept fixed at the preceding values. Two electronic variables are involved: the relaxation rate  $1/T_1$  and the exchange splitting  $\Delta$  of the ground doublet. This lineshape in fact does not allow  $1/T_1$  and  $\Delta$  to be fitted independently in an unambiguous way. We assume that, as relaxation occurs via spin–spin ‘flip-flops’ both above and below  $T_N$ , the relaxation rate  $1/T_1$  is the same above and below  $T_N$ ; it was thus kept fixed in the curve fits at the value 1.45 GHz. This is the reason why we postponed this discussion until after that concerning the determination of  $1/T_1$  in the paramagnetic phase.

The exchange splitting  $\Delta$  is the only adjustable parameter in this lineshape; at 0.185 K and 0.23 K, we obtain  $\Delta = 0.31$  K and 0.29 K respectively. The spectrum at 0.15 K does not show sufficient dynamical broadening to allow an accurate determination of  $\Delta$ , but it is well accounted for by a  $\Delta$  value of 0.3 K. The spectrum at  $T = 0.275$  K is also well fitted with this lineshape and yields a  $\Delta$  value of 0.12 K. However, this fit is not different from the fit using the paramagnetic lineshape (3) and so the (small)  $\Delta$  value obtained is not really significant and we estimate that  $\Delta$  is zero at 0.275 K. Therefore we think that the magnetic transition temperature  $T_N$  lies between 0.23 K and 0.275 K; our estimation of  $T_N$  is slightly higher than that of [3] which assigns  $T_N \approx 0.2$  K. The measured exchange splitting of 0.3 K is of the same magnitude as  $k_B T_N$ , as expected in a molecular field model. The molecular field  $H_{\text{mol}} = \Delta/g_z \mu_B = 560$  Oe is found to be quite small.

There are two alternative assumptions that could equally well fit the spectra between 0.15 K and 0.275 K, but which are to be rejected on physical grounds: first, one could assign the line broadenings to a static distribution of hyperfine fields in the rapid relaxation limit. In this case, the relaxation rate would show an unphysical drop as the temperature increases from 0.23 K to 0.275 K. Second, the spectra below 0.275 K could be fitted to the paramagnetic relaxation lineshape (3), but then the paramagnetic relaxation rate would grow from 0.5 GHz at 0.23 K to 1.45 GHz at 0.275 K, which cannot be accounted for by any mechanism at such low temperature.

### 3.5. Discussion of the results

All the  $^{170}\text{Yb}$  Mössbauer spectra in  $\text{YbPt}_3$  up to 15 K are consistent with the picture of an isolated quasi-pure  $|J_z = \pm \frac{7}{2}\rangle$  crystal field ground state of the  $\text{Yb}^{3+}$  ions, having a temperature independent relaxation rate of about 1.45 GHz due to spin–spin interactions (RKKY exchange or dipolar interactions) and undergoing a magnetic phase transition at about 0.25 K.

However, two problems arise within this interpretation, concerning the origin of the Ising-type electronic ground state and the magnitude of the spin–spin relaxation rate.

First, the Mössbauer data show that the  $\text{Yb}^{3+}$  ground state is very close to an extremely anisotropic  $|J_z = \pm \frac{7}{2}\rangle$  state. The total CEF interaction at a Yb site thus possesses an axial component along a local  $z$  axis and can be written:

$$\mathcal{H}_{\text{CEF}} = \mathcal{H}_{\text{cub}} + B_2^0 [J_z^2 - J(J+1)/3]$$

where  $\mathcal{H}_{\text{cub}}$  is the cubic term and  $B_2^0$  scales with the strength of the axial component. As our x-ray investigation has shown, the lattice structure of  $\text{YbPt}_3$  remains cubic at all temperatures. The axial term thus cannot be a dominant CEF term and has to be

considered as a distortion with respect to cubic symmetry. In such a case, the perturbed CEF eigenstates are in general close to the cubic ones, except in the particular situation where the cubic level scheme is near a level crossing (accidental degeneracy). The ground wave functions are then strongly altered by the axial component and our calculations show that they can be very close to a  $|J_z = \pm \frac{7}{2}\rangle$  state when  $B_2^0 < 0$  and the system is near the  $\Gamma_7$ - $\Gamma_8$  crossing point (parameter  $x \approx -0.58$  as defined in [6]) or the  $\Gamma_6$ - $\Gamma_8$  crossing point ( $x \approx 0.75$ ). So we propose the following interpretation for the presence of the extremely anisotropic  $|J_z = \pm \frac{7}{2}\rangle$  ground state of Yb<sup>3+</sup> in cubic YbPt<sub>3</sub>: the cubic CEF level scheme of Yb<sup>3+</sup> shows a quasi-degeneracy of  $\Gamma_8$  and  $\Gamma_6$  or  $\Gamma_7$ , and there exist random axial distortions in the sample which preserve the overall cubic symmetry. These distortions have a magnitude of about 20 K, which is a relatively large value compared with that usually accepted for random strains (1–2 K). This suggests that the distortions in YbPt<sub>3</sub> might be coherent over a few tens of lattice cells, i.e. that there exists a kind of micro-twinning in this compound. These distortions could account for the line broadenings observed on the x-ray spectra (poorly resolved  $K\alpha_1$ - $K\alpha_2$  doublets at large angles) in spite of the large size of the crystallites ( $\approx 1 \mu$ ). Such a cooperative distortion could be favoured by the assumed quasi-degeneracy of the cubic CEF levels (Jahn-Teller distortion). The assumption of coherent domains of distortions is also in agreement with the absence of important static broadening of the Mössbauer lines at 0.04 K, which means that the majority of the Yb ions have identical environments.

The second problem concerns the magnitude of the spin-spin relaxation rate  $1/T_1$ , which is expected to be quite small for an extremely anisotropic Kramers doublet at low temperature. Relaxation in this case is indeed driven by the transverse part of the RKKY exchange or dipolar interaction between the effective  $S = \frac{1}{2}$  spins  $S_i$  and  $S_j$  of ions  $i$  and  $j$ , which is proportional to  $g_\perp S_i^+ S_j^-$ , assuming a common  $z$  axis for the interacting ions. Using a model of quasi-Ising spins in equivalent sites, where magnetic ordering is assumed to arise from the diagonal part of the RKKY interaction, one can show that the ratio  $(h/T_1)/k_B T_N$  scales roughly with  $(g_\perp/g_z)^4$  [7]. As  $g_\perp$  is less than  $g_z/10$  for the quasi-pure  $|J_z = \frac{7}{2}\rangle$  observed state, the spin-spin relaxation rate is expected to be orders of magnitude lower than  $k_B T_N/h$  (about 5 GHz) and also much lower than the Mössbauer Larmor frequency (about 1 GHz). Then the spectrum in the magnetic phase should be a slow relaxation spectrum, which actually occurs in many Dy intermetallic compounds with a  $|J_z = \pm \frac{15}{2}\rangle$  ground state [8]. On the contrary, in YbPt<sub>3</sub> we find a sizable Yb<sup>3+</sup> spin-spin relaxation rate of about 1.5 GHz at low temperature, which is of the same order of magnitude as the exchange energy  $k_B T_N$  and as the Mössbauer Larmor frequency. So we have to reconsider one of the assumptions made above. One can first think of a random distribution of easy magnetisation  $z$  axes from site to site in the crystal, caused by some structural disorder or defects (for instance partial disorder between Pt and Yb atoms) which would conserve the cubic symmetry of the crystal. The magnetic ordering would then be of Ising spin-glass type, and  $1/T_1$  and  $k_B T_N$  could be of the same order of magnitude. However, such a situation would very probably lead to an important distribution of ground state wave functions and hyperfine parameters, which is not observed on the spectra.

Alternatively, the origin of the RKKY coupling can be reexamined in view of the fact that Yb intermetallic compounds frequently present 4f-band electron hybridisation. Hybridisation gives rise to an effective RKKY exchange coupling between Yb ions [9] which, contrary to the classical RKKY coupling derived from  $k$ - $f$  exchange, does not involve the ionic  $g$  tensor and so does not forbid 'flip-flop' transitions between the states  $|J_z = \frac{7}{2}\rangle$  and  $|J_z = -\frac{7}{2}\rangle$ . It is possible that a weak hybridisation of the 4f electrons with



band electrons in YbPt<sub>3</sub> could account for the slight valence anomaly of Yb and the large residual resistivity observed in this compound by the authors of [3], as well as for the sizable spin–spin relaxation rate measured in the present work between the states of the  $|J_z = \pm\frac{7}{2}\rangle$  ground doublet.

#### 4. Yb<sub>3</sub>Pt<sub>5</sub>

Yb<sub>3</sub>Pt<sub>5</sub> crystallises into a monoclinic lattice structure [3]; the site symmetry of the Yb atom is thus quite low. We recorded spectra at 0.04 K, 0.8 K, 1 K and 4.2 K. The  $T = 0.04$  K spectrum (see figure 2(a)) is an effective field spectrum, well accounted for by an axial hyperfine Hamiltonian where the principal EFG axis is parallel to the hyperfine field (1). The obtained values are:  $H_{\text{hf}} = 3645$  kOe and  $\alpha_Q = 4.7$  mm s<sup>-1</sup>. This confirms the presence of magnetic ordering of Yb at 0.04 K with a saturated moment of  $3.5 \mu_B$ . The ground Kramers doublet thus contains a dominant  $|J_z = \pm\frac{7}{2}\rangle$  contribution. At 0.8 K the magnetic structure is much broadened and at 1 K it has practically disappeared, which is in accordance with the determination of  $T_N \approx 0.95$  K of [3]. At 4.2 K, we observe a paramagnetic relaxation spectrum (see figure 2(b)) which is satisfactorily reproduced by an axial lineshape (3) with  $\alpha_Q = 4.7$  mm s<sup>-1</sup>, an extremely anisotropic **A** tensor  $A_z = 3.85 \times 2 \mu/\mu_B = 27$  mm s<sup>-1</sup>,  $A_{\perp} = 0$  and a relaxation rate  $1/T_1 = 10$  GHz, equivalent to an energy of 0.5 K. The exchange energy in Yb<sub>3</sub>Pt<sub>5</sub> thus scales with the relaxation rate in a similar way as in YbPt<sub>3</sub>, and the presence of a weak 4f-band electron hybridisation can also be invoked here to explain the finite and rather high value of the Yb<sup>3+</sup> relaxation rate. This is further supported by the moderately high electronic specific heat coefficient  $\gamma = 83$  mJ mol<sup>-1</sup> K<sup>-2</sup> measured in Yb<sub>3</sub>Pt<sub>5</sub> [3].

#### 5. Summary and conclusions

Very low temperature <sup>170</sup>Yb Mössbauer absorption experiments on the two intermetallic compounds YbPt<sub>3</sub> and Yb<sub>3</sub>Pt<sub>5</sub> have confirmed the presence of magnetic ordering of the Yb<sup>3+</sup> magnetic moments below 0.25 K and 0.95 K respectively. The measured saturated moments (near  $4 \mu_B$ ) and the value of the quadrupolar coupling parameter show that the ground Kramers doublet in both compounds is close to a  $|J_z = \pm\frac{7}{2}\rangle$  state. In YbPt<sub>3</sub>, the first crystal field excitation is estimated to be about 20 K. The occurrence of a very anisotropic electronic ground state in this compound, where x-ray spectra do not reveal any departure of the lattice from the cubic AuCu<sub>3</sub> type structure, is tentatively explained in terms of the presence of a micro-twinning of domains with axial distortion.

In both compounds, the paramagnetic Yb<sup>3+</sup> relaxation could be measured and assigned to spin–spin couplings. The relaxation rate was found to scale with the exchange energy  $k_B T_N$ . As the dipole–dipole or the usual RKKY exchange interactions are expected to give zero-transition matrix elements between the  $|J_z = \frac{7}{2}\rangle$  and  $|J_z = -\frac{7}{2}\rangle$  states, and thus a vanishingly small relaxation rate, we suggest the presence of a hybridisation induced RKKY coupling to account for the measured relaxation rates. The presence of weak hybridisation in both compounds is supported by the slightly reduced valence (2.95 instead of 3) assigned to Yb in [3] and [4] and by the relatively high electronic specific heat coefficient  $\gamma = 83$  mJ mol<sup>-1</sup> K<sup>-2</sup> measured in Yb<sub>3</sub>Pt<sub>5</sub> [3].

**References**

- [1] Bonville P, Hodges J A, Hulliger F, Imbert P and Ott H R 1987 *J. Magn. Magn. Mater.* **63+64** 626  
Bonville P, Hodges J A, Hulliger F, Imbert P, Jéhanno G, Marimon da Cunha J B and Ott H R 1988 *Hyperfine Interact.* **40** 381
- [2] Bonville P 1988 *Hyperfine Interact.* **40** 15
- [3] Politt B 1987 *Thesis* Köln unpublished  
Oster F, Politt B, Braun E, Schmidt H, Langen J and Lossau N 1987 *J. Magn. Magn. Mater.* **63+64** 629
- [4] Lossau N, Neumann G, Schlabit W, Wohlleben D 1988 *Phys. Scr.* **37** 809
- [5] Nowik I 1967 *Phys. Lett.* **24A** 487
- [6] Lea K R, Leask M J M and Wolf W P 1962 *J. Phys. Chem. Solids* **23** 1381
- [7] Bonville P, Hodges J A, Imbert P and Hartmann-Boutron F 1978 *Phys. Rev. B* **18** 2196
- [8] Nowik I and Wickmann H H 1965 *Phys. Rev.* **140** 869  
Asch L, Bogé M, Chappert J, Gal J, Gorlich E A and Kalvius G M 1983 *Hyperfine Interact.* **15+16** 713  
Rechenberg H R, Bogé M, Jeandey C, Oddou J L, Sanchez J P and Tenand P 1987 *Solid State Commun.* **64** 277
- [9] Coqblin B and Schrieffer J R 1969 *Phys. Rev.* **185** 847