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Magnetic properties of $YbNi_5$ from ^{170}Yb Mössbauer and magnetisation measurements

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Abstract. From Yb^{170} Mössbauer measurements on $YbNi_5$, we establish: the Yb^{3+} ground state Kramers doublet is well isolated and has uniaxial anisotropy, the compound magnetically orders at 0.55 K, the Yb^{3+} moments are aligned along the hexagonal axis, they have a saturated value of $3.9\mu_B$ and a thermal variation close to a mean field $S = 1/2$ law. We obtain the strength of the Yb^{3+}–Yb^{3+} coupling which we find is considerably smaller than that between the rare earths (R^{3+}) in the other RNi_5. From a study of the spin dynamics, we find that dynamic short range order is present just above the ordering temperature and that above 2 K, the relaxation rate of the paramagnetic Yb^{3+} spins follows a T-linear Korringa law with a relatively pronounced slope which we link to a high density of states at the Fermi level. Magnetic susceptibility and magnetisation measurements establish the ferromagnetic nature of the Yb^{3+}–Yb^{3+} coupling. We comment on the Yb^{3+} crystal field properties.

PACS. 75.50.Cc Other ferromagnetic metals and alloys – 76.80.+y Mössbauer effect

1 Introduction

In the hexagonal compounds RNi_5 (R is yttrium or one of the rare earths), the nickel atoms carry essentially no intrinsic magnetic moment because the holes in the nickel $3d$ band are filled by electrons coming from the rare earth atom [1,2]. In fact, the compounds are exchange enhanced Pauli paramagnets with (at least in $LaNi_5$ [2,3]) a high density of states at the Fermi level. Small Ni moments may be induced by an external magnetic field and/or by the molecular field coming from the ordered rare earth moments [2,4,5]. The magnetic properties of RNi_5 are dominated by those of the magnetic rare earth, i.e. by R-R exchange. Most of the RNi_5 have been examined and the general features of the series have been established. They are ferromagnetic [6] and for the heavy rare earths (that is from $DyNi_5$ to $TmNi_5$), the rare earth exchange interaction was reported to be independent of the rare earth, whereas for the light rare earths the interaction is both more important and rare earth dependent [7]. The size of the rare earth magnetic moment is influenced by the crystal field which also governs its direction in the ferromagnetic phase [8]: if the second order crystal field parameter B_2^0 is negative, the moments are parallel to the hexagonal axis, whereas if it is positive, they lie in the basal plane.

To our knowledge, little information has been published concerning the low temperature magnetic properties of $YbNi_5$, the end magnetic member of the series. For example, there does not seem to be any published evidence concerning its magnetic ordering temperature. We present here ^{170}Yb Mössbauer measurements down to 0.05 K which establish the magnetic ordering temperature and provide the size and direction of the magnetically ordered moments and the strength of the Yb^{3+}–Yb^{3+} coupling. These measurements also provide information concerning the dynamics of the Yb^{3+} magnetic moments both in a small temperature range below the magnetic ordering temperature and in a wide temperature range above. We also present susceptibility and magnetisation measurements which establish the ferromagnetic nature of the magnetic transition, the magnetisation measurements also providing a second estimate of the strength of the Yb^{3+}–Yb^{3+} coupling. We also discuss the crystal field properties of the Yb^{3+} ion and compare them with those for the rare earths in the neighbouring RNi_5.

2 Sample and structure

The sample was made by melting the constituent elements in an arc furnace. Three further remeltings optimised the homogeneity. The composition of the starting mixture was adjusted to allow for the preferential evaporation of the ytterbium. The room temperature X-ray analysis showed the sample formed in the expected hexagonal structure ($CaCu_5$-type, space group P6/mmm) and has lattice parameters $a = 0.4847$ and $c = 0.3957$ nm. It contains

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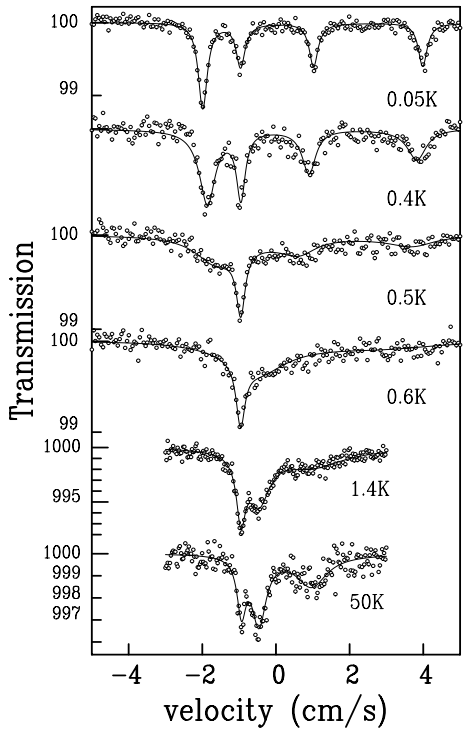

Fig. 1. ^{170}Yb Mössbauer absorption spectra in $YbNi_5$ below and above the magnetic ordering temperature, $T_C = 0.55\,\mathrm{K}$. The lines are fits as explained in the text.

a small amount (<5%) of unidentified impurity phases. Each Yb^{3+} has 6 Ni atoms as nearest neighbours, with the two nearest Yb^{3+} neighbours lying along the **c** axis (distance 0.3957 nm) and the six next nearest Yb^{3+} neighbours lying in the basal plane (distance 0.4847 nm).

3 ^{170}Yb Mössbauer measurements

The ^{170}Yb Mössbauer absorption measurements were made over the temperature range 60 to 0.05 K using a source of Tm^*B_{12} and a triangular velocity sweep. For ^{170}Yb, the ground nuclear state has a spin $I_g = 0$, the excited nuclear state has a spin $I_e = 2$ and a quadrupole moment $Q = -2.11\,b$. The transition energy is $E_\gamma = 84.3\,\mathrm{keV}$ so that 1 cm/s corresponds to 680 MHz. Figure 1 presents spectra for selected temperatures.

3.1 Magnetically ordered region

Anticipating the results of Section 4, we note that the order is ferromagnetic. In the following, we thus refer to the magnetic ordering temperature as a Curie temperature (T_C).

At 0.05 K, in the magnetically saturated state, an effective hyperfine field line shape is observed (Fig. 1 top) and this provides the saturated values of the hyperfine magnetic field (399(2) T) and of the hyperfine quadrupole interaction ($eQV_{zz}/8 = 4.90(0.05)\,\mathrm{mm/s}$). The lineshape analysis also establishes that the hyperfine field (and the Yb^{3+} magnetic moment) lie parallel to the principal axis of the electric field gradient tensor, with component V_{zz}. The magnetic moments thus lie along the hexagonal **c** axis.

For Yb^{3+} ($4f^{13}$, $^2F_{7/2}$) in a metallic material, the dominant contribution to the magnetic hyperfine field comes from the self ion $4f$ shell contribution and very much smaller contributions may arise from the polarisation of the conduction electrons. Assuming the latter may be neglected and using the hyperfine constant for ^{170}Yb: $C \simeq 102\,\mathrm{T}/\mu_B$ [9], we obtain a value $m(0) = 3.9(1)\,\mu_B$ for the saturated Yb^{3+} magnetic moment. The quoted uncertainty is bigger than that in the measurement of the hyperfine field as it allows for a possible contribution of the conduction electrons to the hyperfine field. The z-component of the g-tensor of the ground state Kramers doublet is thus: $g_z = 7.8(2)$. The magnetic moment and the value for g_z are close to those for a fully stretched $|J = 7/2, J_z = \pm 7/2\rangle$ state ($m(0)$=4.0μ_B, $g_z = 8.0$).

The quadrupole hyperfine interaction is also dominated by the contribution coming from the self ion $4f$ shell with a smaller contribution coming from the asymmetric distribution of lattice charges. The value of the lattice contribution ($eQV_{zz}^{latt}/8 \simeq -0.3\,\mathrm{mm/s}$) may be estimated by scaling from the directly measured lattice contribution in $GdNi_5$ [10,11]. The self ion $4f$ shell contribution to the quadrupole hyperfine interaction of Yb^{3+} is thus 5.2(0.1) mm/s which approaches the maximum possible value of 5.8 mm/s linked with the fully stretched $|J = 7/2, J_z = \pm 7/2\rangle$ state. From the values of g_z and of the $4f$ contribution to the quadrupolar hyperfine interaction, the wave-function of the ground doublet can be derived with a good accuracy:

$$|\psi_g\rangle = 0.985\left|\pm\frac{7}{2}\right\rangle + 0.173\left|\mp\frac{5}{2}\right\rangle. \qquad (1)$$

From this we obtain that the value of $g_\perp$ the transverse component of the g-tensor falls in the range 0.5 to 1.0. The Yb^{3+} ground doublet has strong Ising character since $g_z/g_\perp$ lies in the range 8 to 16.

In the small temperature range between ∼0.3 K and T_C, we observe that the widths of some of the lines change whereas there is no clear shift in their positions (see Fig. 1 at 0.4 K). This behaviour indicates that a magnetic hyperfine field (and a spontaneous Yb^{3+} moment) is still present (the compound remains magnetically ordered), but that the correlated moments fluctuate at frequencies which lie within the ^{170}Yb Mössbauer window (from $\sim 2 \times 10^9$ to $\sim 5 \times 10^{11}\,\mathrm{s}^{-1}$).

To obtain more quantitative information from the line shapes, we first note that the crystal field interaction leaves a well isolated Kramers doublet as the ground state (Sect. 5). At temperatures where magnetic order is present, only the two conjugate states of this doublet are thermally populated. Since these two states have strong Ising character, the magnetic fluctuations are longitudinal in nature. We thus analyse the lineshapes in terms of the longitudinal fluctuation model of Nowik et al. [12]. The sizes of the saturated hyperfine field and the quadrupole

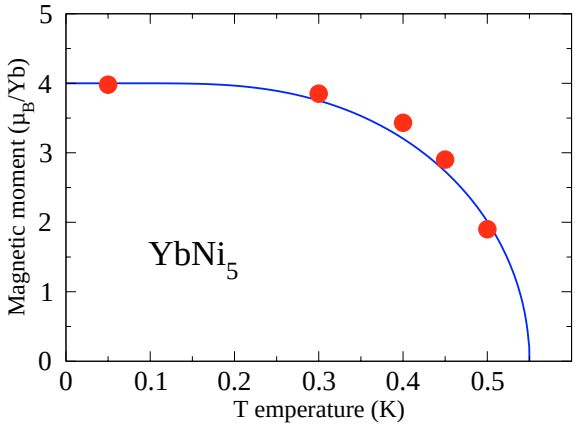

Fig. 2. Thermal variation of the Yb^{3+} spontaneous magnetic moment in $YbNi_5$ obtained from ^{170}Yb Mössbauer measurements. The solid line is a mean field $S = 1/2$ variation with an ordering temperature of $T_C = 0.55$ K.

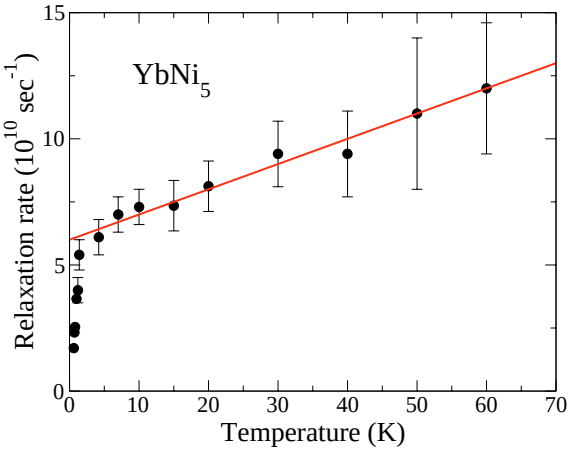

Fig. 3. Thermal dependence of the Yb^{3+} longitudinal spin fluctuation rate in $YbNi_5$ in the paramagnetic region obtained from ^{170}Yb Mössbauer measurements. The line is a fit to the sum of a constant spin-spin term and a T-linear Korringa law term (see text).

interaction are given by the measurement at 0.05 K and the linefits provide values for the two parameters associated with the model: the longitudinal fluctuation rate ($\nu(T)$) and the separation between the two conjugate substates ($\Delta(T)$).

We find that between 0.3 K and T_C, the Yb^{3+} fluctuation rates are of the order of $\nu = 2\times10^{10}\,s^{-1}$. That is, they are close to the rate observed just above T_C (Sect. 3.2).

As the temperature decreases from T_C to 0.3 K, we find that $\Delta(T)$ increases progressively. The saturated value $\Delta(0) = 1.3(10)$ K is obtained by extrapolation. The spontaneous Yb^{3+} moment at each measurement temperature is obtained from the value of $\Delta(T)$ using the relation: $m(T) = m(0)\tanh(-\frac{\Delta(T)}{2k_BT})$, and the exchange field is given by: $H_{ex}(T) = \frac{1}{2}\Delta(T)/m(T)$. The values of the Yb^{3+} magnetic moment so obtained in the range 0.5 to 0.3 K, together with the value obtained directly at 0.05 K, are shown in Figure 2. The data are fitted in terms of a $S = 1/2$ mean field model with a T_C of 0.55 K. Although this model provides a reasonable description of the data, we note that the experimental points at 0.4 and 0.45 K lie slightly above the mean field curve. This raises the possibility that the thermal dependence near T_C is slightly steeper than that given by the mean field model. Such behaviour may be attributed to the influence of the Ising character of the system. However, we also note that dimensional effects do not play a major role since the magnetic ordering temperature is not depressed relative to the energy of the Yb^{3+}–Yb^{3+} coupling (see below). More accurate values for the thermal dependence of the Yb^{3+} moment could probably be obtained by neutron diffraction. These could confirm/infirm the slight difference observed relative to the mean field behaviour.

From the value of $\Delta(0)$, we derive the size of field acting on the Yb^{3+} spin at zero temperature: $H(0) = 0.25$ T. Within the molecular field model, the size of this field is equivalent to a magnetic ordering temperature of $\frac{1}{k_B}m(0)\cdot H(0) = 0.65$ K. This is close to the directly measured value $T_C = 0.55$ K.

3.2 Paramagnetic region

Above T_C, the lineshapes have the characteristic form associated with the combined presence of an axially symmetric quadrupole interaction and paramagnetic fluctuations. These two features were previously established by Nowik et al. who reported a measurement carried out at 4.2 K [13]. At this temperature our results agree with theirs. We observe that the size of the quadrupole interaction remains essentially independent of temperature up to the highest measurement temperature of 60 K. This indicates that up to this temperature, only the lowest crystal field Kramers doublet is thermally populated. (We estimate the lowest excited doublet lies at least 150 K above the ground doublet.) For all the measurement temperatures, the magnetic fluctuations thus only concern the two conjugate, highly anisotropic sublevels of the ground doublet. In the paramagnetic region where there is no spontaneous magnetic moment, the two sublevels remain degenerate. The thermal dependence of the longitudinal fluctuation rate is shown in Figure 3. The relaxation rate at 0.6 K, just above the magnetic ordering temperature, is $1.7\times10^{10}\,s^{-1}$. As the temperature is increased, two different regimes are visible. Up to ∼2 K there is a rapid quasi-linear increase of the relaxation rate with $\nu/T \simeq 1.7\times10^{10}\,s^{-1}/K$, whereas above ∼2 K the increase of the relaxation rate is quasi-linear with $\nu/T \simeq 10^9\,s^{-1}/K$.

We attribute the rapid thermal decrease as the temperature is lowered below ∼2 K to critical slowing down of spin-spin exchange driven fluctuations. It is to be noted that this critical regime extends over a range $\simeq 3T_C$ above the magnetic ordering temperature. The relatively large range of the critical region may be linked with the Ising character of the Yb^{3+} spins and the specific interactions with the nearest and next nearest neighbours. We attribute the quasi-linear increase above 2 K to a Korringa mechanism, where the Yb^{3+} spin relaxation is driven by

coupling to the conduction electrons through $k-f$ exchange.

The line in Figure 3 is a fit to a the law:

$$\nu(T) = \nu_{ex} + C_K T, \tag{2}$$

where the two terms correspond respectively to a temperature independent relaxation rate linked with the exchange interaction and to a Korringa relaxation mechanism.

The fitted value of the exchange rate ν_{ex} is $6\times10^{10}\,\mathrm{s}^{-1}$, and it corresponds to an equivalent temperature of 0.5 K, very close to the value of T_C. For system with an isotropic g-tensor, the correspondence $h\nu_{ex} \sim k_B T_C$, is expected since both quantities should vary as $g^2\mathcal{J}$, where $\mathcal{J}$ is the interionic exchange integral. However, for a very anisotropic doublet, $h\nu_{ex}$ is related to $g_\perp^2$ whereas $k_B T_C$ is related to g_z^2. In the present case where $g_z/g_\perp \sim 10$, the observed equivalence between the values of $k_B T_C$ and $h\nu_{ex}$ should thus be considered as coincidental.

The Korringa constant C_K is:

$$C_K = \frac{4\pi}{\hbar}\left[g_\perp \cdot \frac{g_J - 1}{g_J}\right]^2 \cdot \left[J_{kf} n(E_F)\right]^2 k_B, \tag{3}$$

where J_{kf} is the conduction electron — $4f$ exchange integral, $n(E_F)$ the conduction electron density of states at the Fermi level, $g_\perp$ the transverse component of the ground state g-tensor and $g_J = 8/7$ (for Yb^{3+}). The measured value for C_K then yields: $g_\perp |J_{kf} n(E_F)| \simeq 0.19$. Using the value $g_\perp = 0.5$ to 1.0 (Sect. 3.1), we obtain an $k-f$ exchange coupling constant: $|J_{kf} n(E_F)| \simeq 0.30(10)$. This is an order of magnitude higher than the value ($\sim$0.02) observed for rare earths in metals [14] where the exchange is dominated by the conventional atomic exchange interaction. It is also noticeably higher than the value observed for Yb^{3+} in Pd [15] where the coupling is enhanced by the strong interactions within the Pd d-band.

The large value observed for the product $|J_{kf} n(E_F)|$ in $YbNi_5$ can be linked to the observation of an unusually large Pauli susceptibility and electronic specific heat in isomorphous $LaNi_5$ and YNi_5 [2], which were attributed to a high density of $3d$ states at the Fermi level.

4 Susceptibility and magnetisation measurements

dc susceptibility measurements, made to below the magnetic ordering temperature in a field of 2.6 mT, are shown in Figure 4. The upturn of the susceptibility at the ordering temperature (0.55 K) establishes the ferromagnetic nature of the transition.

Magnetisation measurements in the paramagnetic region, made on the crushed polycrystalline sample set in a resin are shown in Figure 5. These values are consistent with those expected for a randomly oriented polycrystalline sample and the wave functions of the Yb^{3+} ion as established above but they are approximately 50% lower than the values reported in reference [6]. We suggest that the reported values are erroneous. We have checked

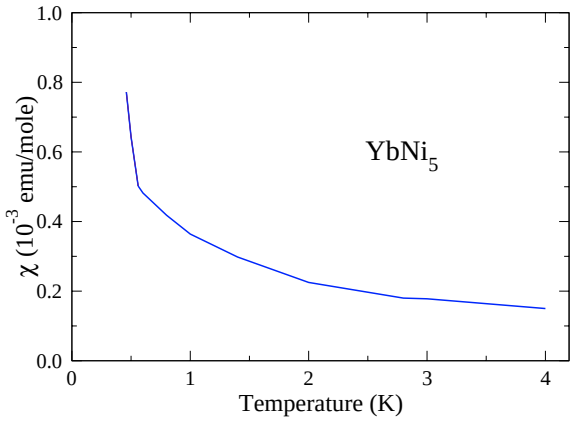

Fig. 4. Magnetic susceptibility for $YbNi_5$ evidencing the ferromagnetic nature of the magnetic transition near 0.5 K. The line links the data points which have been omitted.

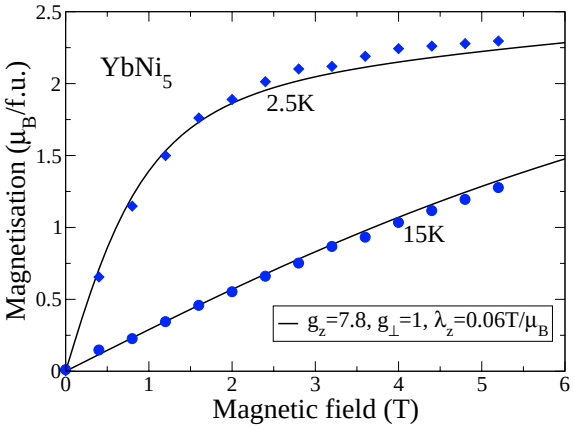

Fig. 5. Isothermal magnetisation measurements for polycrystalline $YbNi_5$ at 2.5 and 15 K. The solid lines are calculated as described in the text.

however that the values of reference [6] are obtained if the crushed polycrystalline sample is not set in a resin. In this case it appears that the individual grains are largely single crystal and under the influence of the very large intrinsic uniaxial anisotropy, the grains rotate so that their c-axes are parallel to the direction of the applied field. The same situation pertains to $ErNi_5$: the reported magnetisation [6] for a putative randomly oriented sample is not coherent with the values that were subsequently obtained for a single crystal sample [5]. We have further checked that the reported values [6] are obtained if the crushed polycrystalline sample is not set in a resin.

The calculated magnetisation curves in Figure 5 were obtained assuming that the magnetisation comes only from the Yb^{3+} ions. We consider a unique Kramers doublet with an axially symmetric g-tensor with $g_z = 7.8$ and $g_\perp = 1$ (Sect. 3.1), and allow for a small Van-Vleck term ϵH, proportional to the field, which accounts for the small mixing, due to the magnetic field, of the excited CEF states into the ground doublet ($\epsilon = 0.0035\,\mu_B/\mathrm{T}$). A self-consistent calculation was performed by introducing a molecular field constant λ, describing Yb-Yb

exchange. The curves shown, which match quite well the data, were obtained with a positive (ferromagnetic) value $\lambda = 0.06\,\mathrm{T}/\mu_B$. This corresponds to a saturated exchange field $H_{ex}(0) = 0.23\,\mathrm{T}$, in good agreement with the value (0.25 T) obtained in Section 3 from the Mössbauer data. The good agreement between the measured and calculated magnetisation data also indicates that any Ni based magnetic moment is quite small.

Belorizky et al. [7] report that in the RNi_5 where R is one of the heavy rare earths (Gd to Tm), the exchange interaction is essentially independent of R and the volumic molecular field constant n_{RR} is roughly equal to 150 cgs. We estimate n_{RR} for $YbNi_5$ from the ionic molecular field constant λ derived above, from the relationship:

$$n_{RR} = \left(\frac{g_J}{g_J - 1}\right)^2 \frac{\lambda}{4N}, \tag{4}$$

where N is the density of rare earth ions. We obtain: $n_{RR} \simeq 85$ cgs, which amounts to only about half of the value established in the neighbouring RNi_5. For the heavy rare earths, the exchange interaction in the RNi_5 is thus not totally independent of the rare earth.

A confirmation of the relatively small value for the Yb^{3+}–Yb^{3+} coupling is provided by comparing the magnetic ordering temperature and the size of the saturated moment in $YbNi_5$ with those in $ErNi_5$. In both cases the ground doublet has a fully stretched wave function. The standard relation linking the ordering temperature ($T_C^{(R)}$), the exchange coupling ($\mathcal{J}^{(R)}$) and the rare earth g_J and J values is written:

$$T_C^{(R)} = \mathcal{J}^{(R)}(g_J - 1)^2 J(J+1). \tag{5}$$

Using the known values (Yb: $T_C = 0.55\,\mathrm{K}$ $g_J = 8/7$, $J = 7/2$, Er: $T_C = 9\,\mathrm{K}$ $g_J = 6/5$, $J = 15/2$), we again find $\mathcal{J}^{(\mathrm{Yb})}$ amounts to only about a half of $\mathcal{J}^{(\mathrm{Er})}$.

5 Crystal electric field

Using the standard equivalent operator notation, the crystal field Hamiltonian is written:

$$\mathcal{H}_{CEF} = \sum_{n,m} B_n^m \theta_n \langle r_{4f}^n \rangle O_n^m \tag{6}$$

where for the hexagonal symmetry of the rare earth site, the non-zero parameters are B_2^0, B_4^0, B_6^0 and B_6^6. Sets of crystal field parameters have been established for a number of the rare earths in the RNi_5 series (a list is given in Radwanski et al. [16]), but none have been proposed for $YbNi_5$. We consider the sets of parameters that have been established for the two adjacent rare earth ions: Er^{3+} ($B_2^0 = -251.7$, $B_4^0 = -51.2$, $B_6^0 = 11.2$ and $B_6^6 = 63.8\,\mathrm{K}$) [17]; Tm^{3+} ($B_2^0 = -376.2$, $B_4^0 = -77.2$, $B_6^0 = -32.6$ and $B_6^6 = 135.6\,\mathrm{K}$) [18] and examine their compatibility with the experimental data for Yb^{3+}. Both sets lead for Yb^{3+} to a situation where each of the four crystal field Kramers doublets is strongly dominated in turn by one of the four possible $|J_z = m\rangle$ components. In both cases the ground doublet is dominated by $|7/2\rangle$ with some admixture with $|-5/2\rangle$, the first excited doublet is dominated by $|5/2\rangle$ with some admixture with $|-7/2\rangle$, and the second and third excited doublets are pure $|\pm 3/2\rangle$ and $|\pm 1/2\rangle$ respectively (these two states remain unmixed for hexagonal symmetry). The set for Er^{3+} when applied to Yb^{3+} leads to a ground state magnetic moment and a low temperature quadrupole interaction which are respectively ∼15% and ∼10% lower than experiment. It predicts a thermal variation of the Yb^{3+} quadrupole interaction which is much more pronounced than experiment (we estimate the energy of the first excited Kramers doublet is ∼150 K whereas the value calculated from the CEF parameters for $ErNi_5$ is only 30 K). The set for Tm^{3+} when applied to Yb^{3+} leads to a ground state magnetic moment and a low temperature quadrupole interaction which are respectively ∼5% and 3% lower than experiment and it predicts a thermal variation of the quadrupole interaction which is only slightly more pronounced than experiment (the first excited doublet is predicted to lie at 100 K, somewhat lower than the estimated value of ∼150 K).

The crystal field parameters relevant for Yb^{3+} thus appear to be closer to those proposed for Tm^{3+} than to those proposed for Er^{3+}. To more fully account for the properties of Yb^{3+}, the parameters for Tm^{3+} need to be modified in two ways. First, to ensure the crystal field states of Yb^{3+} are almost pure $|J_z = m\rangle$ states, the mixing parameter B_6^6 must play a less important role than in $TmNi_5$. Second, to increase the separation between the lowest and the first excited crystal field states for Yb^{3+} to at least 150 K, some changes must also occur in the size of some of the other parameters. Inelastic neutron scattering measurements of the energies of the excited levels would enable a more extended analysis to be made.

6 Summary

We have established the basic magnetic properties (both static and dynamic) of $YbNi_5$, the last member of the RNi_5 series to be reported. The properties are dominated by those of the Yb^{3+} ions and any contribution coming from the Ni atoms is, at most, quite small. The Yb^{3+} magnetic moments order ferromagnetically at 0.55 K. They are aligned along the crystal hexagonal axis and have a saturated value of $3.9\mu_B$. The ordered moment shows a thermal variation which is close to a mean field $S = 1/2$ dependence. The ground state Kramers doublet which is well isolated, is close to a fully stretched $|J = 7/2, J_z = \pm 7/2\rangle$ state. From the measurement of the spin fluctuation rate within this doublet, we identify Korringa processes due the coupling of the rare earth spin with conduction electron spins. The high value obtained for the Korringa constant suggests the density of states at the Fermi level is high, as in the non-magnetic counterparts $LaNi_5$ and YNi_5. The Yb^{3+}–Yb^{3+} exchange obtained from the Mössbauer measurement, which is consistent with that obtained from the magnetisation data, is noticeably smaller (∼50%) than that between the rare earths in the neighbouring RNi_5.

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