Low-temperature spin fluctuations in geometrically frustrated $Yb_3Ga_5O_{12}$

2003 J. Phys.: Condens. Matter 15 4631

(http://iopscience.iop.org/0953-8984/15/26/313)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **15** (2003) 4631–4639 PII: S0953-8984(03)59197-4

Low-temperature spin fluctuations in geometrically frustrated $Yb_3Ga_5O_{12}$

J A Hodges[1], P Bonville[1], M Rams[2] and K Królas[2]

[1] Centre d'Etudes de Saclay, Service de Physique de l'Etat Condensé, 91191 Gif-sur-Yvette, France
[2] Institute of Physics, Jagiellonian University, 30-059 Kraków, Poland

E-mail: hodges@drecam.saclay.cea.fr

Received 4 February 2003
Published 20 June 2003
Online at stacks.iop.org/JPhysCM/15/4631

Abstract

In the garnet-structure compound $Yb_3Ga_5O_{12}$, the Yb^{3+} ions (ground-state effective spin $S' = 1/2$) are situated on two interpenetrating corner-sharing triangular sublattices such that frustrated magnetic interactions are possible. Previous specific heat measurements have evidenced the development of short-range magnetic correlations below ∼0.5 K and a λ-transition at 0.054 K (Filippi *et al* 1980 *J. Phys. C: Solid State Physics* **13** 1277). From ^{170}Yb Mössbauer spectroscopy measurements down to 36 mK, we find that there is no static magnetic order at temperatures below that of the λ-transition. Below ∼0.3 K, the fluctuation frequency of the short-range correlated Yb^{3+} moments progressively slows down and, as $T \to 0$, it tends to a quasi-saturated value of 3×10^9 s^{-1}. We also examined the Yb^{3+} paramagnetic relaxation rates up to 300 K using ^{172}Yb perturbed angular correlation measurements: they evidence phonon-driven processes.

1. Introduction

For most crystallographically ordered compounds containing magnetic ions, the limiting low-temperature magnetic ground state involves long-range magnetic order where the spin fluctuations die out as $T \to 0$. For some particular lattice structures, however, the geometric arrangement of the magnetic ions is such that it may not be possible to minimize simultaneously all pairs of interaction energies. The resulting frustration may then lead to a situation where long-range order does not occur [1, 2] and where the presence of a large number of low-energy states leads to the continued presence of spin fluctuations as $T \to 0$.

Systems with frustrated interactions that are of current interest include the Kagomé lattice [3, 4] where the ions are arranged on a motif of corner-sharing triangles, the pyrochlore lattice [5] where the ions are arranged on corner-sharing tetrahedra, and the garnet lattice

($R_3T_5O_{12}$) [6] where the rare earths (R) form two interpenetrating, non-coplanar, corner-sharing triangular sublattices. This geometry does allow frustration to be operative, provided that there is a suitable combination of the nature and the size of the rare-earth anisotropy and of the sign of the interionic interactions. A number of the rare-earth garnets appear to evidence a conventional long-range ordered state [7] suggesting that, in these cases, frustration plays a negligible role. In fact, to date, frustration has been reported to play a major role in only one garnet, $Gd_3Ga_5O_{12}$ [8–13], where the S-state Gd^{3+} ion has a very small intrinsic anisotropy and where the dominant coupling is antiferromagnetic.

Amongst the garnets made with the non-S-state rare earths, $Yb_3Ga_5O_{12}$ is unusual in that the Yb^{3+} ground state shows only a relatively modest crystal field anisotropy (see below). Specific heat data has evidenced a broad peak centred near 0.2 K, attributed to short-range correlations and a sharp λ-peak at 0.054 K (initially attributed to the onset of long-range magnetic order) [14]. We have carried out ^{170}Yb Mössbauer spectroscopy measurements down to 0.036 K in order to examine the behaviour of the Yb^{3+} moments as the temperature is lowered through that of the λ-transition and to examine the low-temperature spin dynamics. We also report ^{172}Yb perturbed angular correlation (PAC) measurements, carried out from 14 to 300 K, which provide information concerning the thermal dependence of the Yb^{3+} fluctuation rates in the paramagnetic region.

2. Background properties

The single-phase polycrystalline sample was prepared by heating the constituent oxides to 1100 °C four times with intermediate grindings.

In the garnet lattice (space group $Ia\bar{3}d$), the rare-earth-site point symmetry is orthorhombic (mmm). The crystal field acts on the Yb^{3+}, $^2F_{7/2}$ state to leave a ground-state Kramers doublet which is very well isolated from the excited Kramers doublets [15]. For Yb^{3+} ions diluted in $Y_3Ga_5O_{12}$, the ground doublet g-values are $g_x = 3.73$, $g_y = 3.60$ and $g_z = 2.85$ [16, 17] and the wavefunction is derived from the cubic Γ_7 state ($g = 3.43$). In $Yb_3Ga_5O_{12}$, the Yb^{3+} g-values should be quite similar.

A magnetic 4f-shell contribution to the specific heat is visible below ∼0.5 K (see figure 1): there is a broad peak centred near 0.2 K, followed by a λ-anomaly at 0.054 K. The total electronic entropy change below 1 K is very close to the value $R \ln 2$ that is expected for an isolated Kramers doublet, and only about 10% of it is released at the λ-transition [14]. If we attribute the broad peak to exchange-driven short-range correlations, then the exchange energy scale is ∼0.2 K. The susceptibility data [14, 18] follow a Curie–Weiss behaviour down to ∼1.0 K, with a small extrapolated paramagnetic Curie–Weiss temperature ($\theta_p = 0.05$ K) corresponding to a net ferromagnetic interaction. Below ∼1.0 K, in the region where the broad specific heat peak occurs and where the magnetic correlations develop, the thermal dependence of the susceptibility falls below that corresponding to the extrapolated Curie–Weiss dependence. This behaviour shows the presence of magnetic correlations that are antiferromagnetic. In $Yb_3Ga_5O_{12}$, there is thus evidence for the existence of both ferromagnetic and antiferromagnetic interactions. The small value for θ_p suggests that the two types of interactions have comparable strengths. The magnetic frustration in $Yb_3Ga_5O_{12}$ that is evidenced in this report thus appears to be linked to the presence of antiferromagnetic correlations within a Heisenberg-like system on triangular sublattices and to the presence of interactions with opposite signs. The isomorphous compound $Gd_3Ga_5O_{12}$, where frustration is also operative, evidences a dominant nearest-neighbour interaction that is antiferromagnetic as well as other interactions with competing signs [8].

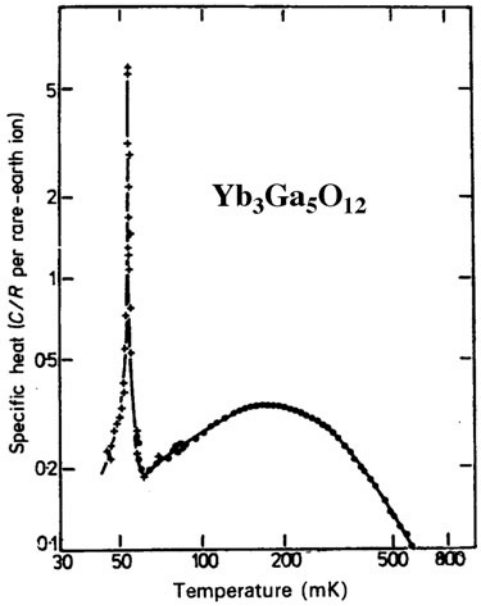

Figure 1. The specific heat in $Yb_3Ga_5O_{12}$, reproduced from [14].

3. ^{170}Yb Mössbauer measurements

3.1. General features

The ^{170}Yb Mössbauer absorption measurements ($I_g = 0$, $I_e = 2$, $E_\gamma = 84$ keV; 1 cm s^{-1} corresponds to 680 MHz) were made down to 0.036 K in a ^{3}He–^{4}He dilution refrigerator using a neutron-activated TmB_{12} source displaced with a triangular velocity sweep. Selected spectra at 4.2, 0.15, 0.075 and 0.036 K are shown in figure 2. The last two temperatures are situated either side of the temperature of the specific heat λ-transition (0.054 K).

At 4.2 K, the absorption takes the form of a broad Lorentzian-shaped line with a half-width at half maximum, $\Gamma = 4$ mm s^{-1}, that is significantly broader than the experimental half-width of the TmB_{12} source, $\Gamma_0 = 1.35$ mm s^{-1}. As the temperature is lowered to ∼0.25 K, the absorption line remains Lorentzian shaped and keeps essentially the same width, but moves slightly towards negative Doppler velocities. This means that the centre of gravity of the absorption no longer corresponds to the value of the isomer shift, which is close to 0.0 mm s^{-1} relative to the TmB_{12} source. Below ∼0.3 K, the absorption line moves markedly towards more negative velocities. It also progressively broadens and becomes slightly asymmetric (see the spectra at 0.075 and 0.036 K in figure 2). No resolved hyperfine structure is visible at any temperature, even at temperatures below that of the λ-transition. This indicates that there is no 'static' long-or short-range magnetic order and that magnetic fluctuations persist down to the lowest temperatures. In relation to the characteristic frequency scale of the present ^{170}Yb Mössbauer measurements, the absence of a well resolved hyperfine structure means that the fluctuation frequency of the Yb^{3+} magnetic moments remains above the threshold value of $\sim 3 \times 10^8$ s^{-1}. The quantitative analysis of the fluctuation rate is presented in the next section.

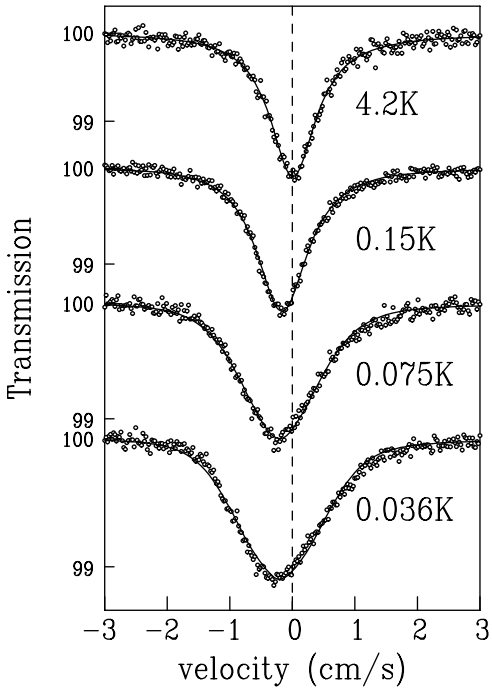

Figure 2. $^{170}Yb^{3+}$ Mössbauer absorption in $Yb_3Ga_5O_{12}$. At 4.2 K, the fitted curve was obtained using a relaxation line shape appropriate for paramagnetic fluctuations. At 0.15, 0.075 and 0.036 K, the data fits were obtained using a line shape appropriate for hyperfine field fluctuations. As the temperature decreases, the absorption broadens and its centre of gravity moves towards velocities that are more negative than the isomer shift value shown by the dashed line.

3.2. Quantitative analysis

The broad, single-line nature of the absorption in the paramagnetic region at 4.2 K arises because the Yb^{3+} magnetic hyperfine splitting is 'motional narrowed' by the fast fluctuations of the Yb^{3+} magnetic moment. It is possible to extract the fluctuation frequency from the measured line shape by using a paramagnetic spin relaxation model based on a perturbative approach [19], provided that the magnetic hyperfine tensor $\mathcal{A}$ is known. With the approximation of local axial symmetry, we obtained the components of this tensor from a ^{170}Yb Mössbauer measurement on Yb^{3+} ions diluted in $Y_3Ga_5O_{12}$ where, because the dilution removes the spin–spin coupling, the fully resolved hyperfine splitting is observable. We obtained the values $A_z/h = 738$ MHz and $A_\perp/h = 952$ MHz, corresponding respectively to g-values $g_z = 2.82$ and $g_\perp = 3.63$. These values are essentially equivalent to those measured previously by electron spin resonance ($g_z = 2.85$ and $g_\perp = (g_x + g_y)/2 = 3.66$ [18]). We note that, with an isotropic $\mathcal{A}$ tensor and in the fast relaxation limit (i.e. when $A/\hbar \ll 1/\tau$, where $1/\tau$ is the Yb^{3+} paramagnetic spin relaxation rate), this line-shape model leads to a single line of Lorentzian shape with a dynamical half broadening given by:

$$\Delta\Gamma_R = \tfrac{3}{2}(A/\hbar)^2\tau. \tag{1}$$

On fitting the data at 4.2 K using the perturbation approach [19] and the axially symmetric $\mathcal{A}$ tensor components, we obtain the Yb^{3+} paramagnetic spin fluctuation rate: $1/\tau \simeq 3.8 \times 10^{10}$ s^{-1}. This rate is constant between ~0.25 K and the highest measurement temperature of 80 K, indicating that the driving mechanism is the temperature-independent

spin–spin coupling between the Yb^{3+} ions. A rough estimate of the strength of this coupling, using the relation $\hbar/\tau \sim k_B T_{ex}$, yields $T_{ex} \sim 0.3$ K—consistent with the temperature of the broad maximum of the exchange-induced specific heat (∼0.2 K).

The shift in the centre of gravity of the absorption away from the temperature-independent isomer shift value evidenced in figure 2 is analogous to that previously observed at low temperatures in $YbAlO_3$ [20] and $YbBe_{13}$ [21]. It seems to be related to the inadequacy, in these cases, of the perturbative relaxation line shape when the driving mechanism is the exchange spin–spin interaction. As discussed in [21], it is likely that the matrix elements of the relaxation operator have non-vanishing imaginary parts [22, 23] and that these generate line shapes whose spectral signature is an anomalous shift of the centre of gravity of the absorption. The fact that the anomalous shift increases as the temperature decreases could be linked to the growing influence of the spin–spin correlations, and hence to the growing inadequacy of the standard perturbative relaxation line shape. The relaxation rates obtained from the perturbation analysis are not influenced by the shift of the centre of gravity of the absorption since the relaxation rate is linked to the real part of the matrix elements, i.e. to the width of the absorption line.

At very low temperatures ($T < 0.1$ K), the shape of the experimental absorption can no longer be reproduced correctly by the paramagnetic relaxation model. Since the specific heat data (figure 1) show that magnetic correlations are present in this temperature region, we fitted the experimental data below 1 K using a relaxation model involving hyperfine field fluctuations that were considered in the random phase approximation (RPA) [24]. A hyperfine field (H_{hf}) is indeed present at the ^{170}Yb nucleus when the Yb^{3+} moments are short (or long) range correlated and the size of the field is proportional to that of the Yb^{3+} moment. The time-dependent Hamiltonian for this relaxation line shape is

$$\mathcal{H} = \mathcal{H}_Q - g_n \mu_n H_{hf} \sum_{j=1,N} I_j f_j(t), \tag{2}$$

where $\mathcal{H}_Q$ is the quadrupolar hyperfine Hamiltonian, g_n is the gyromagnetic factor of the excited nuclear state, μ_n is the nuclear Bohr magneton, the summation is over the N directions among which the hyperfine field fluctuates, and $f_j(t)$ is a random function of time with appropriate values corresponding to the different possible forms of the Hamiltonian. For $\mathcal{H}_Q$ we take the (very small) value that we have determined from ^{170}Yb measurements on Yb^{3+} substituted into $Y_3Ga_5O_{12}$. Then, the line shape depends on a single dynamic parameter—the fluctuation frequency of the hyperfine field $(1/\tau)_{hf}$—and on the choice of the directions between which the fluctuations take place. We obtain very satisfactory fits to the data below 0.2 K (the solid lines in figure 2 at 0.036, 0.075 and 0.15 K) by assuming that the hyperfine field fluctuates between the three principal directions of the local coordinate frame, i.e. between OX, OY and OZ, the principal directions of the electric field gradient tensor. In the Hamiltonian (2), this corresponds to $N = 6$ and $j = \pm X, \pm Y$ and $\pm Z$. If we assume that the hyperfine field fluctuates along only one of these axes, we obtain much poorer data fits. In the rapid relaxation rate limit, this model yields a line of Lorentzian shape with a dynamical half-width:

$$\Delta\Gamma_R = 2\frac{(g_n \mu_n H_{hf}/\hbar)^2}{(1/\tau)_{hf}}. \tag{3}$$

The 0.036 K line shape is well broadened, and this enables us to obtain both the magnitude of the hyperfine field and its fluctuation rate. We find that $H_{hf} \simeq 140(10)$ T, which corresponds to a Yb^{3+} moment of $\simeq 1.4\ \mu_B$ (for $^{170}Yb^{3+}$, 1 μ_B yields a hyperfine field of 102 T) and $(1/\tau)_{hf} = 3 \times 10^9\ s^{-1}$. The value for the Yb^{3+} moment is not far from the mean value expected both from the average g-tensor (3.4) and from the saturated magnetization measured at 0.09 K in [14] (1.7 μ_B). In the fits for the spectra up to 0.2 K, we used an intrinsic half-width,

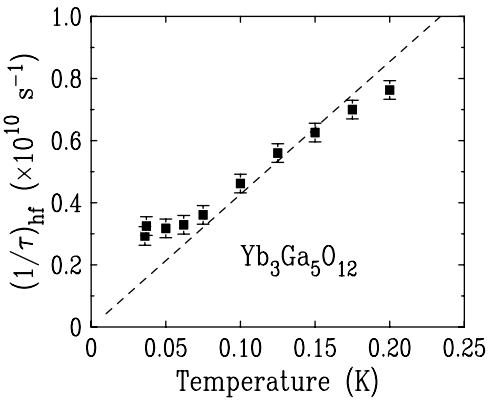

Figure 3. Thermal variations, in $Yb_3Ga_5O_{12}$, of the Yb^{3+} hyperfine field fluctuation frequency extracted from the ^{170}Yb Mössbauer spectra. The dashed line is the law $\hbar(1/\tau)_{hf} = 0.3k_BT$.

$\Gamma_0 = 1.35$ mm s^{-1}, and assumed that the fluctuating hyperfine field has a size that remains constant at the value 140 T derived at 0.036 K. This assumption should hold as long as the correlations are well developed, but it cannot be ascertained that it is completely correct up to 0.2 K.

The thermal variation of $(1/\tau)_{hf}$ is shown in figure 3. As the temperature is lowered over the range 0.2 to 0.1 K, the frequency decreases approximately linearly according to a law $\hbar(1/\tau)_{hf} = 0.3k_BT$. Then, below about 0.1 K, it tends to saturate towards the value 3×10^9 s^{-1}. There is essentially no difference between the rates either side of the specific heat λ-transition (0.054 K). The T-linear dependence of $(1/\tau)_{hf}$ pertains only to a limited temperature range, and it must be kept in mind that it somehow depends on the validity of the assumption about the constant magnitude of the fluctuating hyperfine field. We note that a linear variation has also been encountered theoretically for the case of a frustrated Heisenberg pyrochlore antiferromagnet [5]. The decrease in the relaxation rate, followed by a saturation, is similar to the behaviour observed (by μSR [13]) in the isomorphous compound $Gd_3Ga_5O_{12}$.

4. ^{172}Yb perturbed angular correlation (PAC) measurements

The PAC measurements provide the Yb^{3+} paramagnetic relaxation rates in the temperature range 14–300 K. The measurements were made using the 91–1094 keV γ–γ cascade from the ^{172}Lu → ^{172}Yb β decay. The ^{172}Lu nuclei are obtained by proton irradiating the sample, which is then annealed at 800 °C to remove irradiation defects. The intermediate level of the cascade, namely the 1172 keV nuclear excited level of ^{172}Yb with spin $I = 3$ and half-life 8.3 ns, is used to observe the perturbation of the γ–γ directional correlations due to the hyperfine interactions. In the case of a static quadrupolar or magnetic hyperfine interaction, oscillations are observed in the time evolution of the perturbation factor $R(t)$ [25].

Up to ∼300 K, we observe that the perturbation factor, $R(t)$, does not show oscillations but is instead an exponential function of time, $R(t) = A\exp(-\mu t)$ (see figure 4). This suggests a dynamic hyperfine interaction. Paramagnetic relaxation within the ground Yb^{3+} doublet, with effective spin $\frac{1}{2}$, leads to such an exponential decay in the fast relaxation limit: $A^{172}/\hbar \ll 1/\tau$, where A^{172} is the hyperfine constant of the intermediate $I = 3$ level of ^{172}Yb and $1/\tau$ is the electronic spin fluctuation frequency. Then, in a manner analogous to equation (1), the

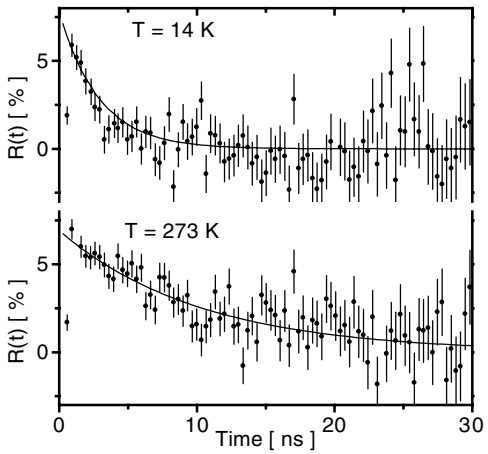

Figure 4. ^{172}Yb PAC spectra at 14 and 300 K, fitted to an exponential decay. The first channel, spoilt due to prompt coincidences, was removed from the fits.

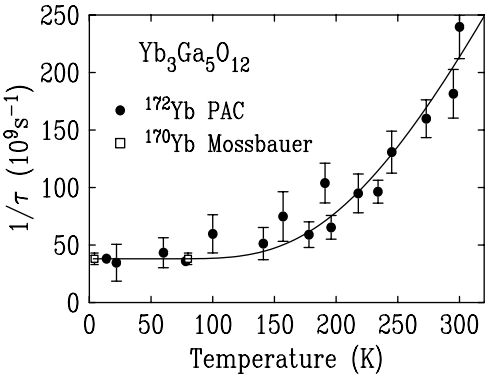

Figure 5. Thermal variation of the Yb^{3+} 4f shell magnetic fluctuation rate in $Yb_3Ga_5O_{12}$ derived from PACs measurements (black dots). Two values obtained from the ^{170}Yb Mössbauer spectra at 4.2 and 80 K are also shown (open squares). The solid curve is a fit to a sum of spin–spin- and spin–phonon-driven relaxation rates (see text).

damping rate μ is given by [26]

$$\mu = \tfrac{3}{2}(A^{172}/\hbar)^2\tau. \tag{4}$$

The value for the hyperfine constant of the $I = 3$ level of ^{172}Yb can be obtained by scaling the mean A^{170} value appropriate for the Yb^{3+} ground state ($A^{170}/h \simeq 884$ MHz) with the nuclear g-factors. We obtain $A^{172}/h \simeq 565$ MHz. The thermal variation of $1/\tau$, derived from that of μ, is shown in figure 5, which also shows the ^{170}Yb Mössbauer-derived values at 4.2 and 80 K. The two sets of values agree quite well. The relaxation rate is constant up to $\sim$150 K, then increases monotonically. The observed thermal dependence can be fitted to the sum of a temperature-independent spin–spin term and an exponential term associated with a two-phonon real process through the excited crystal field states (Orbach process) [27]:

$$\frac{1}{\tau} = \left(\frac{1}{\tau}\right)_{ss} + B\exp\left(-\frac{\Delta}{k_B T}\right), \tag{5}$$

where Δ is the energy of an excited crystal field level. The fit yields $(1/\tau)_{ss} = 3.8 \times 10^{10}$ s^{-1}, $B = 3.3 \times 10^{12}$ s^{-1} and $\Delta = 880(50)$ K. This last value is in very good agreement with the mean energy distance between the Yb^{3+} ground doublet and the three closely spaced excited crystal field doublets (∼850 K) [15]. Additional PAC measurements concerning the thermal variation of the electric field gradient at the ^{172}Yb nucleus above 300 K are reported in [28].

5. Discussion and conclusions

Previous specific heat measurements [14] have shown that, in $Yb_3Ga_5O_{12}$, magnetic correlations develop below ∼0.5 K and that a λ-transition occurs at 0.054 K. Our present investigations, using ^{170}Yb Mössbauer spectroscopy down to 36 mK and ^{172}Yb PACs measurements up to 300 K, provide insight into the correlations and the dynamic behaviour of the Yb^{3+} spins over a wide temperature range.

Above ∼0.5 K, the Yb^{3+} magnetic moments undergo paramagnetic fluctuations. Up to ∼150 K, the fluctuation rate has a temperature-independent value of $\simeq 3.8 \times 10^{10}$ s^{-1} and the driving mechanism is the exchange interaction between the Yb^{3+} spins. Above 150 K, additional temperature-dependent relaxation occurs through coupling to phonons, according to a two-phonon Orbach process involving the excited crystal field states near 850 K.

The low-temperature Yb^{3+} magnetic correlations show up in the ^{170}Yb Mössbauer measurements below ∼0.3 K through changes in the line shape. The fluctuation frequency of the correlated moments decreases as the temperature is lowered and, below 0.1 K, it tends to a quasi-saturated value of 3×10^9 s^{-1}. This is a quite high value and, in fact, $Yb_3Ga_5O_{12}$ is the only known compound where the $T \rightarrow 0$ spin fluctuation rate is rapid enough to fall within the ^{170}Yb Mössbauer spectroscopy frequency window. The $T \rightarrow 0$ state in $Yb_3Ga_5O_{12}$ is therefore a dynamic short-range correlated spin-liquid state. On crossing the temperatures of the specific heat λ-anomaly, there is no significant change in the ^{170}Yb Mössbauer line shape. Usually, a transition to a long-range magnetically ordered phase reveals itself in the Mössbauer spectra by the appearance of a well defined magnetic hyperfine splitting. This does not appear in the present case, showing clearly that there is no long-range order at temperatures below that of the specific heat peak. This peak has the intriguing characteristic that the associated entropy gain is very small, i.e. it amounts to about 10% of the total $R \ln 2$ entropy gain associated with the Yb^{3+} ground-state Kramers doublet.

Some examples are already known of frustrated systems where the specific heat peak has an associated low entropy. In the pyrochlore compound $Gd_2Ti_2O_7$ there is an entropy gain of 50% of $R \ln 8$ at the transition at 1 K [29], which has been shown by neutron diffraction to involve long-range magnetic order [30]. $Yb_2Ti_2O_7$ presents an entropy gain of about 20% of $R \ln 2$ [31] at the transition at 0.25 K, which has been shown to be associated with a first-order change in the fluctuation rate of the correlated Yb^{3+} moments [32]. In $Yb_3Ga_5O_{12}$, the reduced-size specific heat peak is linked neither to the appearance of long-range order nor to a measurable change in the fluctuation rate of the short-range correlated moments. In fact, we observe a slowing down of the fluctuations of the correlated moments below ∼0.3 K, i.e. at temperatures well above that of the specific heat peak, and the fluctuation rate is quasi-temperature-independent in the region where the specific heat peak occurs.

Usually, for frustrated systems presenting a phase transition, the temperature at which this transition occurs is sizeably lower than the temperature associated with the strength of the interionic interaction. The energy scale of this interaction may be estimated in different ways: from the paramagnetic Curie–Weiss temperature; from the temperature where the specific heat evidences a magnetic contribution (in the present case, the temperature of the broad maximum); and from the paramagnetic spin–spin relaxation rate. In $Yb_3Ga_5O_{12}$, the last two methods lead

to an interaction of equivalent strength 0.2–0.3 K, whereas the paramagnetic Curie temperature (θ_p) is smaller (0.05 K). The correspondence between θ_p and the temperature of the specific heat peak appears to be a mere coincidence. The precise origin of this low-entropy specific heat peak within a spin liquid phase remains an unresolved issue.

Acknowledgments

We would like to thank Mrs Anne Forget for preparing the samples and Mrs J Wojtkowska of the Institute for Nuclear Studies, Otwock, Poland for the proton irradiation for the PAC experiments.

References

[1] Diep H T (ed) 1994 *Magnetic Systems with Competing Interactions* (Singapore: World Scientific)
[2] Villain J 1979 *Z. Phys.* B **33** 31
[3] Broholm C, Aeppli G, Espinosa G P and Cooper A S 1990 *Phys. Rev. Lett.* **65** 3173
[4] Watanabe I, Wada N, Yano H, Okuno Ts, Awaga K, Ohira S, Nishiyama K and Nagamine K 1998 *Phys. Rev.* B **58** 2438
[5] Moessner R and Chalker J T 1998 *Phys. Rev. Lett.* **80** 2929
Moessner R and Chalker J T 1998 *Phys. Rev.* B **58** 12049
[6] Schiffer P and Ramirez A P 1996 *Comments Condens. Matter Phys.* **18** 21
[7] Onn D G, Meyer H and Remeika J P 1967 *Phys. Rev.* **156** 663
Herpin A and Meriel P 1964 *C. R. Acad. Sci., Paris* **259** 2416
[8] Kinney W I and Wolf W P 1979 *J. Appl. Phys.* **50** 2115
[9] Hov S, Bratsberg H and Skjeltorp A T 1980 *J. Magn. Magn. Mater.* **15–18** 455
[10] Schiffer P, Ramirez A P, Huse D A and Valentino A J 1994 *Phys. Rev. Lett.* **73** 2500
[11] Petrenko O A, Paul D McK, Ritter C, Zeiske T and Yethiraj M 1999 *Physica* B **266** 41
[12] Dunsiger S R, Gardner J S, Chakhalian J A, Cornelius A L, Jaime M, Kiefl R F, Movshovich R, MacFarlane W A, Miller R I, Sonnier J E and Gaulin B D 2000 *Phys. Rev. Lett.* **85** 3504
[13] Marshall I M, Blundell S J, Pratt F L, Husmann A, Steer C A, Coldea A I, Hayes W and Ward R C C 2002 *J. Phys.: Condens. Matter* **14** L157
[14] Filippi J, Lasjaunias J C, Hebral B, Rossat-Mignod J and Tcheou F 1980 *J. Phys. C: Solid State Phys.* **13** 1277
[15] Buchanan R A, Wickersheim K A, Pearson J J and Hermann G F 1967 *Phys. Rev.* **159** 245
[16] Wolf W P, Ball M, Hutchings M T, Leask M J M and Wyatt A F G 1962 *J. Phys. Soc. Japan Suppl.* B-1 **17** 443
[17] Carson J W and White R L 1960 *J. Appl. Phys.* **31** 53S
[18] Ball M, Garton G, Leask M J M and Wolf W P 1960 *Proc. 7th Int. Conf. on Low Temperature Physics (Toronto 1961)* (Amsterdam: North-Holland) p 128
[19] Gonzalez-Jimenez F, Imbert P and Hartmann-Boutron F 1974 *Phys. Rev.* B **9** 95
[20] Bonville P, Hodges J A and Imbert P 1978 *Phys. Rev.* B **18** 2196
[21] Bonville P, Imbert P and Jéhanno G 1986 *J. Phys. F: Met. Phys.* **16** 1873
[22] Afanas'ev A M and Onishchenko E V 1978 *Zh. Eksp. Teor. Fiz.* **74** 1115 (Engl. transl. 1978 *Sov. Phys.–JEPT* **47** 585)
[23] Afanas'ev A M, Onishchenko E V, Asch L and Kalvius G M 1978 *Phys. Rev. Lett.* **40** 816
[24] Dattagupta S 1977 *Phys. Rev.* B **16** 158
[25] Frauenfelder H and Steffen R M 1965 *Alpha- Beta- Gamma-Ray Spectroscopy* ed K Siegbahn (Amsterdam: North-Holland) ch XIXA
[26] Abragam A and Pound R V 1953 *Phys. Rev.* **92** 943
[27] Orbach R 1961 *Proc. Phys. Soc.* A **264** 458
[28] Królas K, Rams M, Forget A and Wojtkowska J 1999 *Hyperfine Interact.* **120/121** 231
[29] Raju N P, Dion M, Gingras M J P, Mason T E and Greedan J E 1999 *Phys. Rev.* B **59** 14489
[30] Champion J D M, Wills A S, Fennel T, Bramwell S T, Gardner J S and Green M A 2001 *Phys. Rev.* B **64** 140407(R)
[31] Blöte H W J, Wielinga R F and Huiskamp W J 1968 *Physica (Utrecht)* **43** 549
[32] Hodges J A, Bonville P, Forget A, Yaouanc A, Dalmas de Réotier P, André G, Rams M, Królas K, Ritter C, Gubbens P C M, Kaiser C T, King P J C and Baines C 2002 *Phys. Rev. Lett.* **88** 077204