Magnetic excitations in a new anisotropic Kagomé antiferromagnet

Julien Robert, Virginie Simonet, Benjamin Canals, Rafik Ballou, Pierre Bordet, Isabelle Gelard, Alain Ibanez, Pascal Lejay, Jacques Ollivier, Anne Stunault

To cite this version:
Julien Robert, Virginie Simonet, Benjamin Canals, Rafik Ballou, Pierre Bordet, et al.. Magnetic excitations in a new anisotropic Kagomé antiferromagnet. 2006. hal-00068174

HAL Id: hal-00068174
https://hal.science/hal-00068174
Preprint submitted on 10 May 2006

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
Magnetic excitations in a new anisotropic Kagomé antiferromagnet

J. Robert a V. Simonet a,∗ B. Canals a R. Ballou a P. Bordet b I. Gelard b A. Ibanez b P. Lejay c J. Ollivier d A. Stunault d

a Laboratoire Louis Néel, CNRS, B.P. 166, 38 042 Grenoble Cedex 9, France
b Laboratoire de Cristallographie, CNRS, B.P. 166, 38 042 Grenoble Cedex 9, France
c Centre de Recherches des Très Basses Températures, CNRS, B.P. 166, 38 042 Grenoble Cedex 9, France
d Institut Laue-Langevin, BP 154, 38042 Grenoble Cedex, France.

Abstract

The Nd-langasite compound contains planes of magnetic Nd³⁺ ions on a lattice topologically equivalent to a kagomé net. The magnetic susceptibility does not reveal any signature of long-range ordering down to 2 K but rather a correlated paramagnetism with significant antiferromagnetic interactions between the Nd and a single-ion anisotropy due to crystal field effect. Inelastic neutron scattering on Nd-langasite powder and single-crystal allowed to probe its very peculiar low temperature dynamical magnetic correlations. They present unusual dispersive features and are broadly localized in wave-vector Q revealing a structure factor associated to characteristics short range-correlations between the magnetic atoms. From comparison with theoretical calculations, these results are interpreted as a possible experimental observation of a spin liquid state in an anisotropic kagomé antiferromagnet.

Key words: kagomé, spin liquid, inelastic neutron scattering, magnetic anisotropy

The Heisenberg kagomé antiferromagnet is the archetypal example of a highly frustrated magnetic 2-dimensional lattice, capable of stabilizing a spin-liquid state. Extensive theoretical work was devoted to the study of the peculiar nature of this spin liquid, classically described by a non-magnetic highly degenerate fluctuating ground state. Unfortunately, it is usually destabilized by second-order perturbations, as well as by entropic selection of soft modes via the "order by disorder" mechanism [1]. From the experimental side, very few examples of ideal kagomé magnetic lattice were found in real systems which were, moreover, often prone to non-stoichiometry. Among these, we find the kagomé bilayers SCGO and BSZCGO [2], the jarosites [3], and the natural volborthite [4], which stabilize non-conventional spin glasses, exotic ordered phases, and show signatures of correlated paramagnetism below the paramagnetic Néel temperature. All these are examples of Heisenberg kagomé antiferromagnets. The case of anisotropic kagomé antiferromagnets, in which interesting new magnetic behaviours are expected, has been much less studied theoretically and was, up to now, still waiting for physical realizations.

The present study is devoted to a langasite compound, a family better known for their application in the domain of piezoelectricity [5]. However, a thorough analysis of their structure [6] (space group P321) indicates that the 3e sites belonging to planes stacked perpendicular to the 3-fold c-axis, form lattices with the same overall topology as the kagomé one [7]. In the studied Nd₃Ga₅SiO₁₄ compound, these sites are all occupied by the magnetic Nd³⁺ ions, antiferromagnetically coupled to each other by superexchange. Nd³⁺, with electronic configuration 4f³, is expected to present strong anisotropy due to the crystal field splitting of the fundamental multiplet J=9/2.

In the following, we report results of magnetization measurements performed on a Quantum Design MPSMS SQUID magnetometer and of inelastic neutron scattering measurements on powder sample and

∗ Corresponding Author: Email: simonet@grenoble.cnrs.fr

Preprint submitted to Elsevier Science

10 May 2006
The compound is indeed frustrated and a good candidate for a spin liquid phase. At high temperature, the anisotropy of Nd-langasite is most probably described by coplanar rotators lying in the kagomé planes. A change of the anisotropy occurs at 33 K, the c axis becoming the magnetization one at lower temperature, due to higher order anisotropy terms in the crystalline electric field potential [7].

To characterize the magnetic excitations in the system, inelastic neutron scattering measurements were carried out. The main features revealed by the time-of-flight experiment are the low levels of the $J=9/2$ multiplet splitted by the crystal field, with a first intense one detected around 8.5 meV. In addition, a much weaker signal could be detected, localized in modulus of $Q$ in contrast to the crystal field levels that are constant in $|Q|$ (neglecting the magnetic form factor). This signal is observed between 0.8 and 1.2 meV and around 1.1 Å$^{-1}$ (cf. Fig. 2) [9].

A three-axis inelastic neutron scattering experiment on single crystal was necessary in order to confidently measure and characterize this small signal. The measurements were done in the horizontal scattering plane containing the [100] and [010] axes. Energy scans, performed at different points of the reciprocal space, confirmed the presence of a small signal around 1 meV and $Q=1.1$ Å$^{-1}$. This signal mimics those of the calculated static magnetic structure factors which suggests its magnetic origin [9]. $Q$-scans were then performed at several energies around 1 meV in different directions of the reciprocal space. In Fig. 3, the results at an energy of 0.85 meV, spanning the 15° rotated [100], [410] and [210] directions, are reported. The spectra for the 3 directions are very similar yielding a ring-shaped maximum of intensity at around 1.15 Å$^{-1}$. Then, there is a minimum at 2.2 Å$^{-1}$ and a second weak maximum rising at larger $Q$ values (at least for the [210] direction). This underlines that the magnetic intensity pattern is not equally distributed in all the BZ.

Quantitatively, the main peak was fitted with a

Fig. 2. Iso-intensity cut of the time-of-flight spectrum at 2 K in Nd-langasite powder. Positive energy transfers are on the neutron energy loss side. The darkest areas correspond to maximum intensity: an horizontal stripe around the elastic position and a localized one, spotted by the white circle.
lorentzian, multiplied by the square of the Nd$^{3+}$ magnetic form factor. This analysis yielded a HWHM of 0.49 Å$^{-1}$, i.e. a very short correlation length of 2 Å, smaller than the distance between two Nd (4.2 Å).

The full analysis of the magnetic $Q$ distribution as a function of energy shows a complex behavior, which is detailed elsewhere [9]. A second peak, whose position varies, is found at certain energies. The first peak position also changes slightly, which indicates a significant dispersion of the magnetic scattering with $Q$ in the range 0.5 to 1 meV. The life time of these magnetic fluctuations, estimated from the half width of the energy response at constant $Q$, is of the order of $1.4 \times 10^{-12}$ s.

An interpretation of these results can be obtained in the light of both experimental and theoretical works. Calculations were performed using several models of spin-spin correlation functions on the antiferromagnetic kagomé lattice obtained with the real crystallographic positions of the Nd$^{3+}$ ions [9]. The calculations give the static magnetic structure factor for the spin liquid kagomé phase, which is characteristic of the low energy spin fluctuations. As expected, the $Q$-distribution of magnetic intensity is therefore in best agreement with the experimental results at the lowest investigated energy of 0.5 meV [9]. Actually, when first neighbors only are considered whatever the spin dimension, an empty first BZ and a ring of maximum intensity around 0.9 Å$^{-1}$ are found. This magnetic structure factor, based on a disordered state, is for instance compatible with short-range magnetic correlations between 3 Nd$^{3+}$ first neighbors on a triangle forming a non magnetic state in the XY and Heisenberg cases.

These non magnetic states were already invoked to interpret the $Q$ pattern of the dynamical correlations obtained in previous inelastic neutron scattering experiments on single-crystal compounds containing frustrated pyrochlore lattice of magnetic atoms: the itinerant Y$_{0.97}$Sc$_{0.03}$Mn$_2$ [10] and the insulating ZnCr$_2$O$_4$ [11]. The $Q$ patterns, measured in these compounds and interpreted as originating from spin liquid correlations, are indeed close to the present one. The similarity can be related to the fact that a cut perpendicular to the cube diagonal of the pyrochlore lattice leads to the kagomé one. In addition, the very short correlation lengths found in Nd-langasite and in Y$_{0.97}$Sc$_{0.03}$Mn$_2$ [10] are thought to come from the high degeneracy of the magnetic modes which is another signature of a spin–liquid state in a frustrated antiferromagnet. However, for the Y$_{0.97}$Sc$_{0.03}$Mn$_2$ compound, the life time of the spin correlations was found much shorter than in the present case and no dispersion was observed. The understanding of these differences and the thorough characterization of the $Q$-distribution of the magnetic intensity must now be tackled with realistic calculations taking into account the anisotropy of the system.

In conclusion, the analysis of the static magnetic properties and of the dynamical magnetic correlations of Nd-langasite suggests that this compound could be the first example of a spin liquid state stabilized in an anisotropic kagomé antiferromagnet.

References

[1] D. A. Huse et al., Phys. Rev. B 45, 7536 (1992); A.B. Harris et al., Phys. Rev. B 45, 2899 (1992); J. N. Reimers et al., Phys. Rev. B 48, 9539 (1993); P. W. Leung et al., Phys. Rev. B 47, 5459 (1993); Ch. Waldtmann et al., Eur. Phys. J. B 2, 501 (1998); D. A. Garanin, and B. Canals, Phys. Rev. B 59, 443 (1999).
[2] L. Limot et al., Phys. Rev. B 65, 144447 (2002); D. Bono et al., Phys. Rev. Lett. 92, 217202 (2004).
[3] A. S. Wills, Can. J. Phys. 79, 1563 (2001); D. Grohol et al., Nature Materials 4, 323 (2005).
[4] A. Fukaya et al., Phys. Rev. Lett. 91, 207603 (2003); F. Bert et al., cond-mat/0507250.
[5] T. Iwataki, H. Ohsato, K. Tanaka, H. Morokoshi, J. Sato, and K. Kawasaki, J. Eur. Cer. Soc. 21, 1409 (2001).
[6] B. V. Mill, A. V. Butashin, G. G. Kudrashbayan, E. L. Belokonova, and N. V. Belov, Dokl. Akad. Nauk SSSR 264 (6) 1395 (1982).
[7] P. Bordet et al., submitted to J. Phys.: Condens. Matter (2005).
[8] P. Lejay et al., in preparation.
[9] J. Robert et al., in preparation.
[10] R. Ballou et al., Phys. Rev. Lett. 76, 2125 (1996); R. Ballou, Can. J. Phys. 79, 1475 (2001).
[11] S. -H. Lee et al., Nature 418, 856 (2002).