Magnetic excitations in a new anisotropic Kagomé antiferromagnet

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Abstract

The Nd-langasite compound contains planes of magnetic Nd^{3+} 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 antiferromagets, 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 $Nd_3Ga_5SiO_{14}$ compound, these sites are all occupied by the magnetic Nd^{3+} ions, antiferromagnetically coupled to each other by superexchange. Nd^{3+}, with electronic configuration $4f^3$, 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

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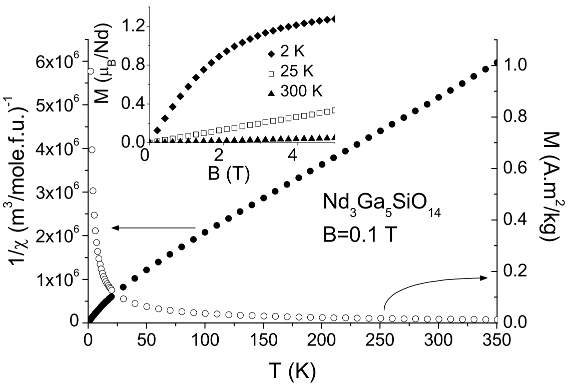

Fig. 1. Magnetization and inverse linear susceptibility measured from 2 to 350 K in a 0.1 T magnetic field on Nd-langasite powder. Inset : magnetization versus magnetic field at different temperatures.

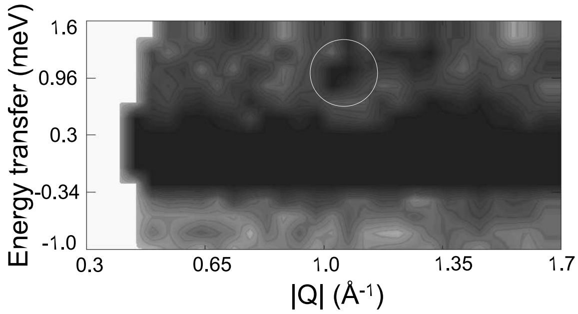

Fig. 2. Iso-intensity cut of the time-of-flight spectrum at 2 K in Nd-langasite powder. Positive energy transfers are on the neutron energy loss side. The darkest areas correspond to maximum intensity : an horizontal stripe around the elastic position and a localized one, spotted by the white circle.

on single-crystal. Large single-crystals of Nd-langasite were indeed successfully grown by a floating zone method using an image furnace, starting from a powder obtained through a solid state reaction at 1420°C in air [8]. The powder neutron scattering experiments were performed at the Institut Laue-Langevin on the time-of-flight spectrometer IN5. The results presented here were obtained with an incident wavelength of 4.5 Å with a chopper speed of 12000 rpm and an energy resolution (FWHM) of 100 μeV. Neutron scattering spectra were recorded at 2 K in the wave-vector Q range [0.46, 2.48 Å^{-1}] and energy range [-197, +2.8 meV]. The dynamical magnetic correlations and their localization in reciprocal space were more precisely determined on a single-crystal at 2 K using the cold-neutron three-axis spectrometer IN14 with fixed final energy of 4.66 meV and energy resolution of 165 μeV.

As shown in Fig. 1, the thermal variation of the magnetization measured on a powder sample under an applied magnetic field of 0.1 T down to 2 K shows no anomaly nor any thermomagnetic hysteresis that would indicate a transition towards a long range order or a spin glass state. The inverse susceptibility is linear down to 70 K before diving towards zero, the linear part extrapolating to a negative intercept of the temperature axis. The shape of the susceptibility is modulated by the anisotropy. Its analysis, reported elsewhere [7], is based on single-crystal measurements with magnetic field applied parallel and perpendicular to the kagomé planes. The high temperature analysis yields an effective moment $\mu_{\rm eff} \approx 3.77\ \mu_{\rm B}$ close to the value of the Nd^{3+} free ion and a paramagnetic Néel temperature θ of -52 K, which confirms the existence of significant antiferromagnetic interactions between the Nd^{3+} ions. The fact that no long range order is detected down to 2 K, a temperature well below the θ value, shows that 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], [4$\bar{1}$0] and [2$\bar{1}$0] directions, are reported. The spectra for the 3 directions are very similar yielding a ring-shaped maximum of intensity at around 1.15 Å^{-1}. Then, there is a minimum at 2.2 Å^{-1} and a second weak maximum rising at larger Q values (at least for the [2$\bar{1}$0] direction). This underlines that the magnetic intensity pattern is not equally distributed in all the BZ.

Quantitatively, the main peak was fitted with a

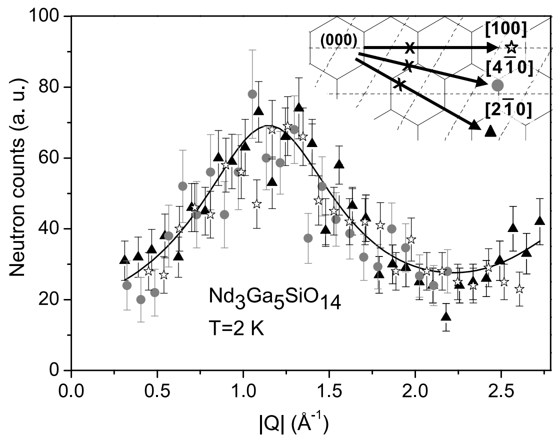

Fig. 3. Three-axis measurements on a Nd-langasite single-crystal: Q-scans in the [100], [4$\bar{1}$0] and [2$\bar{1}$0] directions at 0.85 meV drawn as a function of $|Q|$. The line is a lorentzian based fit (see text). Right up corner : the spanning of the reciprocal space, with $|a^*|$=$|b^*|$=0.899 Å^{-1}, is sketched and the neutron intensity maximum for the 3 directions is indicated by a cross.

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 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 $ZnCr_2O_4$ [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.

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