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Magnetic excitations in a new anisotropic Kagomé antiferromagnet

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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 characteristic 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 4f⁶, 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...
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 μ_eff ≈ 3.77 μB close to the value of the Nd³⁺ free ion and a paramagnetic Néel temperature θ of -52 K, which confirms the existence of significant antiferromagnetic interactions between the Nd³⁺ 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| (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
The distribution of magnetic intensity is therefore in best 
Fig. 3. Three-axis measurements on a Nd-langasite sin-
-interpret the Q pattern of the dynamical correlations ob-
-structured pyrochlore lattice of magnetic atoms : the itiner-
-2 K and [210] directions at 0.85 meV drawn as a function of [Q]. The line is a lorentzian 
lorentzian, multiplied by the square of the Nd$^{3+}$ mag-
|Q (Å$^{-1}$)|

\[
\begin{align*}
\text{Neutron counts (a. u.)} & \\
0 & 20 \\
0.5 & 40 \\
1 & 60 \\
1.5 & 80 \\
2 & 100 \\
2.5 &
\end{align*}
\]

\[\text{Nd}_2\text{Ga}_5\text{SiO}_{14}\]

T=2 K

\[\text{[100]} \quad \text{[410]} \quad \text{[210]}\]

\[\begin{align*}
0.5 & 1 \\
1 & 2 \\
2.5 &
\end{align*}\]

References


