

Experimental report

05/09/2022

Proposal: 4-05-779

Council: 10/2019

Title: Magnetic moment fragmentation in (NdLa)₂Zr₂O₇ pyrochlore magnets

Research area: Physics

This proposal is a new proposal

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Samples: Nd_{1.6}La_{0.4}Zr₂O₇
Nd_{1.8}La_{0.2}Zr₂O₇

Instrument	Requested days	Allocated days	From	To
IN6-SHARP	3	0		
PANTHER	3	3	25/02/2021	01/03/2021

Abstract:

In the pyrochlore compound Nd₂Zr₂O₇, a peculiar magnetic state has been observed through elastic and inelastic neutron scattering measurements : a dynamic fragmented phase where spin-ice features (pinch point pattern) and a partial all-in all-out ordering (Bragg peaks) coexist.

In order to understand this phenomenon, we study doped Nd₂Zr₂O₇ compounds by substituting non-magnetic ion La³⁺ on the Nd³⁺ site. We expect this substitution to affect directly the all-in all-out order. Their magnetic structure has been determined by experiments at the LLB on the G4.1 powder diffractometer. Despite a doping of x=10% and 20% of the Nd³⁺ magnetic ions, the all-in all-out ordering persists and, in addition, the ordered magnetic moment increases with the doping rate.

To probe the role of disorder on the magnetic state of the Nd³⁺ ions and to determine the associated crystal electric field (CEF) Hamiltonian, we propose to measure the CEF excitations on PANTHER and IN6 on two powder compounds, [Nd(1-x)Lax]₂Zr₂O₇ with x=0.1 and 0.2.

Experimental report 4-05-779: magnetic moment fragmentation in $(\text{NdLa})_2\text{Zr}_2\text{O}_7$ pyrochlore magnets

In the pyrochlore compound $\text{Nd}_2\text{Zr}_2\text{O}_7$, a peculiar magnetic state has been observed through elastic and inelastic neutron scattering measurements: a partial all-in all-out ordering (Bragg peaks) is stabilized while the excitations feature a dynamic fragmented phase including a flat mode characterized by a pinch point pattern. In order to test the robustness of this peculiar state, we have studied doped $\text{Nd}_2\text{Zr}_2\text{O}_7$ compounds by substituting the non-magnetic ion La^{3+} on the Nd^{3+} site. We expect this substitution to affect directly the magnetism. Their magnetic structure has been determined by experiments at LLB on the G4.1 powder diffractometer. Despite a substitution rate of $x=10\%$ and 20% of the Nd^{3+} magnetic ions, the all-in all-out ordering persists and, in addition, the ordered magnetic moment increases with the doping rate. To probe the role of disorder on the magnetic state of the Nd^{3+} ions and to determine the associated crystal electric field (CEF) Hamiltonian, we proposed to measure the CEF excitations on PANTHER on two powder compounds, $[\text{Nd}_{(1-x)}\text{La}_x]_2\text{Zr}_2\text{O}_7$ with $x=0.1$ and 0.2 .

The maps shown in Figure 1 represent the different energy levels of the crystal field and are realized at several temperatures, ranging from 1.5K to 250K . We have suppressed some of the experimental noise by using empty cell measurements. These measurements are performed using an empty sample holder.

For the two substituted samples, we measured the energy levels with an incident energy of 50 meV to confirm or not the presence of the 24meV and $33\text{-}35\text{meV}$ levels observed on the pure compound [1-3]. The figures below show the presence of these levels for both samples at 1.5K . These measurements were carried out for several temperature values up to 250K . They highlight the transitions between the different CEF energy states with the appearance of an energy level at 10meV with the increase of the temperature, when the 24meV level starts to be populated. These transitions become more important at about 100K where we can observe a continuous line of intensity, partly hidden by the presence of phonons.

The measurements also show the presence of phonons around 15 meV , characterised by their very high intensity at high Q , (visible in both samples) and which increases with increasing temperature. To investigate the nature of the levels at 10 and 15 meV , we also made a measurement at an incident energy of 19 meV for the 10% -doped sample. These measurements tend to confirm the presence of a phonon, without it being possible to learn more significantly.

To visualise the level present at 100 meV in the pure compound [3], we also made measurements with an incident energy of 130 meV , but the likely energy level was not visible due to the difficulty of measuring at low and high Q values at energies above 70meV .

All of these measurements showed that the same energy levels (around 24meV and 34meV) are present as in the pure compound, indicating that despite substitution rates of 10% and 20% , the lanthanum substitution does not seem to alter the

magnetic properties of the Nd^{3+} ion. The resolution of the instrument is not sufficient to be able to separate the two levels at 34 and 35 meV and thus confirm that there are two neighbouring levels at this energy.

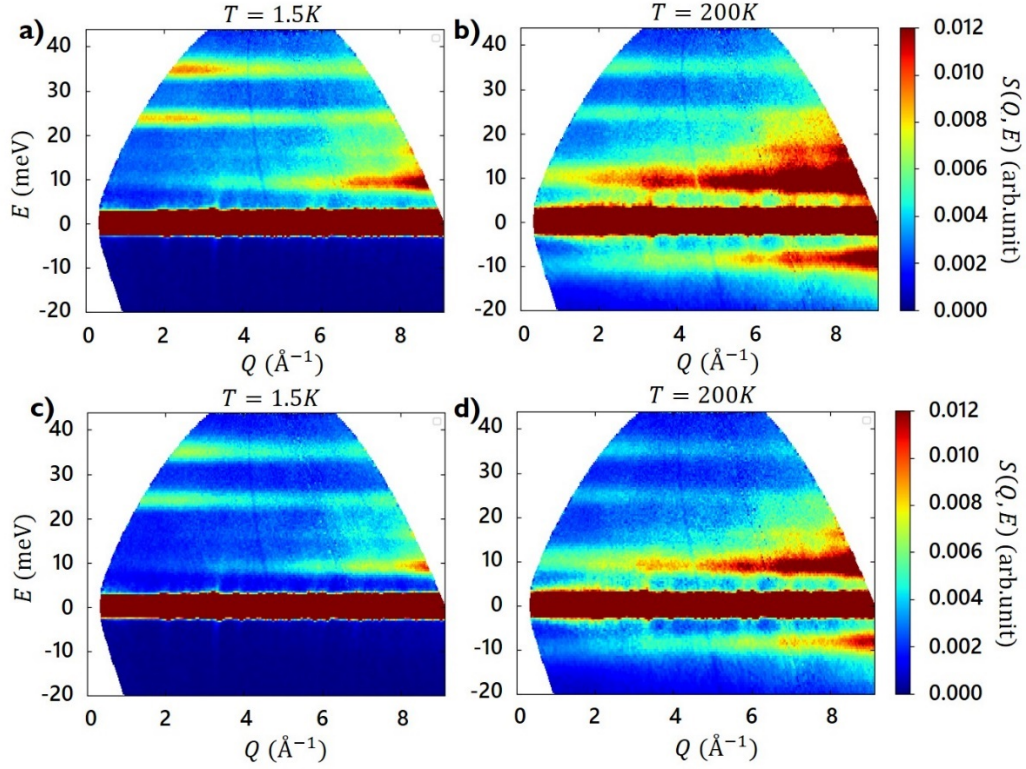


Figure 1: Energy as a function of Q at two temperatures, 1.5K and 200K for the two La-substituted samples: (a-b) 10% substitution and (c-d) 20% substitution.

Bibliography:

- [1] E. Lhotel *et al.*, Phys. Rev. Lett. 115, 97202 (2015)
- [2] M. Ciomaga Hatnean *et al.*, Phys. Rev. B 91, 174416 (2015)
- [3] J. Xu *et al.*, Phys. Rev. B 92, 224430 (2015)