Experimental report

15/09/2022

Proposal:	**5-51-578**	**Council:**	10/2020
Title:	Dipoles, octupoles and Nd2Zr2O7		
Research area:	Physics		

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Samples: Nd2Zr2O7

Instrument	Requested days	Allocated days	From	To
D3	7	7	19/05/2021	27/05/2021

Abstract:

We propose to determine whether or not the All in All Out state of the frustrated magnet Nd2Zr2O7 carries an octupolar moment. This challenging experiment would allow to test general theoretical predictions about pyrochlore magnets where dipole-dipole and multipolemultipole conspire to build unconventional ground states.

Exp report on 5-51-578@D3 : Dipoles, octupoles and $Nd_2Zr_2O_7$

Scientific context: In the two last decades, the study of pyrochlore frustrated magnets has led to a flurry of novel concepts and unconventional phases [1,2,3]. Using a combination of neutron scattering measurements, we have especially focused on $Nd_2Zr_2O_7$ [4,5,6] and shown that this compound hosts an antiferromagnetic "all in all out" state (AIAO), where all spins point in or out of a given tetrahedron of the structure. The ordering occurs below T_N=300 mK and is characterized by a reduced ordered moment (about 0.8 μ_B) compared to the effective moment deduced from susceptibility and CEF investigations (2.3 μ_B). Owen Benton has proposed an interesting interpretation for this strong reduction [7]. Using an appropriate set of interactions, he showed that the AIAO can be stabilized, yet, owing to the dipole octupole nature of the Nd^{3+} ion [4,5,6], the magnetization not only involves dipoles but also octupoles moments. To test the theory, it remains to observe this octupolar contribution. In principle, the latter should be visible by means of elastic neutron scattering, taking the form of anomalous intensities of the magnetic Bragg peaks at large Q vectors ($Q > 6\text{Å}^{-1}$). We thus proposed to perform a polarization analysis at D3 to test these ideas.

To this end, we have written a code to compute the neutron cross section from a pyrochlore lattice of ions carrying not only a magnetic moment but more generally described by a wavefunction written in the $|-J, ., , J\rangle$ basis ($J = 9/2$ in the Nd case). We used the articles of S. Lovesey to have an explicit expression of the magnetization $\mathcal{M}$ which incorporates these high order multipoles. The Hamiltonian writes:

$$\mathcal{H} = \sum_{n,m} B_{n,m} O_{n,m} + g_J \mu_B J.h + q\ O_{33}$$

The $O_{n,m}$ are the Stevens equivalent operators, h and q are respectively magnetic and octupolar internal fields. Both are introduced to mimic the AIAO field, which in principle should be solved in a self-consistent manner in a mean field approximation. O_{33} is one of the octupolar operators. The ground state wavefunction ψ is obtained by diagonalizing $\mathcal{H}$. Those calculations show that q/h should be close to about 0.045 to obtain a magnetic moment of 0.8 μ_B (see figure 1 left).

Following classical definitions, the nuclear and magnetic structure factors are defined as:

$$F_N = \sum_i b_i\ e^{iQr_i}, \quad F_y = \gamma r_o \sum_i \mathcal{M}_i.y\ e^{iQr_i}, \quad F_z = \gamma r_o \sum_i \mathcal{M}_i.z\ e^{iQr_i}$$

$\mathcal{M}_i$ is the magnetization at site i, which, in the dipolar approximation, writes: $\mathcal{M}_i = f(Q)\frac{g_J}{2} J_i$. $f(Q)$ is the form factor. The y and z axes are defined as being respectively unit vectors perpendicular to Q in the scattering plane and perpendicular to Q out of the scattering plane, hence along the vertical axis ($(1, -1, 0)$ in the experiment). We compute both $\mathcal{M}$ in the dipolar approximation and in the general case using the wavefunction ψ determined above. We also define :

$$\begin{aligned}
\sigma_N &= |F_N|^2 \\
M_y &= |F_y|^2 \\
M_z &= |F_z|^2 \\
\chi &= 2\left(\mathrm{Re}F_y \mathrm{Im}F_z - \mathrm{Im}F_y \mathrm{Re}F_z\right) \\
I_y &= 2\left(\mathrm{Re}F_N \mathrm{Im}F_y - \mathrm{Im}F_N \mathrm{Re}F_y\right) \\
I_z &= 2\left(\mathrm{Re}F_N \mathrm{Im}F_z - \mathrm{Im}F_N \mathrm{Re}F_z\right) \\
\xi &= 2\left(\mathrm{Re}F_y \mathrm{Re}F_z + \mathrm{Im}F_y \mathrm{Im}F_z\right) \\
R_y &= 2\left(\mathrm{Re}F_N \mathrm{Re}F_y + \mathrm{Im}F_N \mathrm{Im}F_y\right) \\
R_z &= 2\left(\mathrm{Re}F_N \mathrm{Im}F_z + \mathrm{Im}F_N \mathrm{Im}F_z\right)
\end{aligned}$$

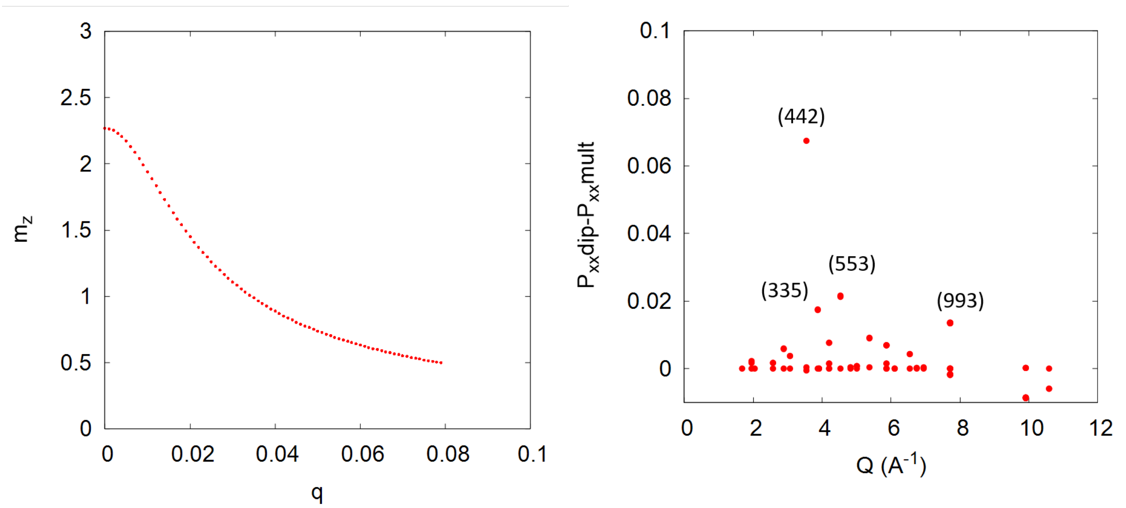

FIG. 1. (left) : calculated magnetic moment as a function of the octupolar molecular field. (right) : Difference of polarization for an AIAO structure where each site is described by a wavefunction ψ whose magnetization contains both a dipolar and an octupolar component. Here, we show only the diagonal elements but do not give the component explicitely. The difference is performed between polarizations calculated in the dipole approximation and in the general case.

and finally the polarization matrix:

$$\sigma_x = \sigma_N + M_y + M_z - \chi$$
$$\sigma_y = \sigma_N + M_y + M_z + R_y$$
$$\sigma_z = \sigma_N + M_y + M_z + R_z$$
$$P = \begin{pmatrix} \frac{\sigma_N - M_y - M_z - \chi}{\sigma_x} & \frac{I_z - \chi}{\sigma_x} & \frac{I_y - \chi}{\sigma_x} \\ \frac{-I_z + R_y}{\sigma_y} & \frac{\sigma_N + M_y - M_z + R_y}{\sigma_y} & \frac{\xi + R_y}{\sigma_y} \\ \frac{-I_y + R_z}{\sigma_z} & \frac{\xi + R_z}{\sigma_z} & \frac{\sigma_N - M_y + M_z + R_z}{\sigma_z} \end{pmatrix}$$

Since the AIAO has two opposite S domains, we further set $I_y = I_z = R_y = R_z = 0$. Based on those results, we conclude that P_{xx} and P_{zz} strong depend on the magnetic moment. The other components of the polarization tensor are close to 0 (except $P_{yy} \approx 1$). Figure 1 right shows the difference between polarizations calculated either in the dipole approximation and in the most general case. The largest difference between the two methods arises from a few peaks : (442), (335), (553) and (993), but remains very small, about 2 % (6% for the (442) family).

Experiment: Sample orientation was carried out at ORIENT-EXPRESS. Installation of cryopad, of the dilution fridge and waiting for thermalization last about one full day. A significant amount of time was also devoted to calibrations carried out by the local contact. For the sake of illustration, we give below the polarization matrix for the purely nuclear $(10, 10, 10)$ reflection.

$$P = \begin{pmatrix} 1.0142 & 0.0283 & 0.0607 \\ 0.0597 & 1.0117 & 0.0282 \\ 0.0195 & 0.0322 & 1.0155 \end{pmatrix}$$

We collected integrated intensities for different series ranging from $(1, 1, \ell)$ up to $(11, 11, \ell)$ at the base temperature of the dilution fridge as well as at 10K. The $+x+x$, $+y+y$ and $+z+z$ channels were measured systematically, and in more specific cases $+x+y$, $+x+z$ and $+y+z$. For some reflections, centering failed and we rejected the data.

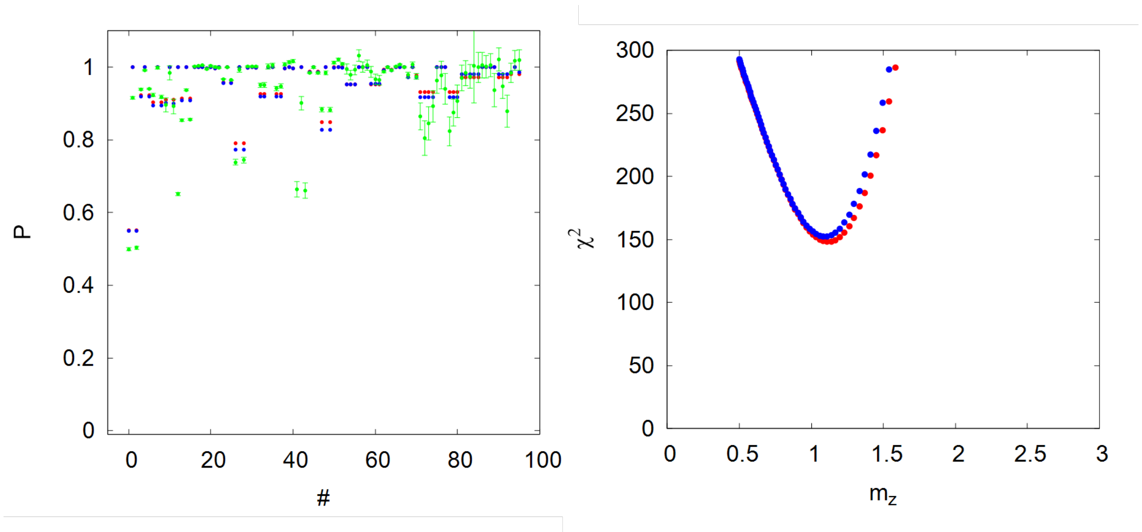

FIG. 2. (left) : Elements of the polarization matrix P (P_{xx}, P_{yy}, P_{zz}). Measured (green), calculated in the dipole approximation (red) and in the general case (blue). (right) the two curves shows the normalized squared difference between measurement and calculations as a function of the magnetic moment calculated according to the models (red,dipole; blue, general case).

The $(2, 2, 2)$ (purely nuclear) was periodicaly monitored to check the polarization efficiency. Special thanks must be given to the ^{3}He cell team who did an exceptional job. However, this constant monitoring of the polarization as well as the constant worry that the next cell might not be ready makes such an experiment on D3 really stressful (the cell has to be changed almost every day).

The left panel of figure 2 shows elements of the polarization matrix for the different Bragg peaks mentionned above P (P_{xx}, P_{yy}, P_{zz}) as measured (green), calculated in the dipole approximation (red) and in the general case (blue) for a magnetic moment of 1.1 μ_B $(h = 0.1, q = 0.031)$. Unfortunately, we suspect a strong depolarization for the (442) peak, which remains to be understood. This is a pity since this is also the one which is the most descriminating between the two models. The right panel shows the normalized squared difference between measured and calculated polarizations as a function of the magnetic moment (red, dipole; blue, general case). We conclude from these calculations (and from both methods) that the ordered magnetic moment is closer to 1.1 μ_B rather than 0.8. However, we could not obtain a direct proof for the existence of an octupolar component.

References: [1] Fennell, T. et al. Science 326, 415 (2009). [2] Balents Nature 464, 199 (2010). [3] Gingras et al., Rep. Prog. Phys. 77, 056501 (2014). [4] E. Lhotel et al, Phys. Rev. Lett. 115, 197202 (2015). [5] S. Petit, E. Lhotel et al, Nature Physics, 12, 746750 (2016) [6] E. Lhotel et al, Nature Comm. vol 9, 3786 (2018) 74] Benton, Phys. Rev. Lett. 121, 037203 (2018).