Magnetic excitations in multiferroic $NdFe_3(BO)_4$:inelastic neutron scattering investigation

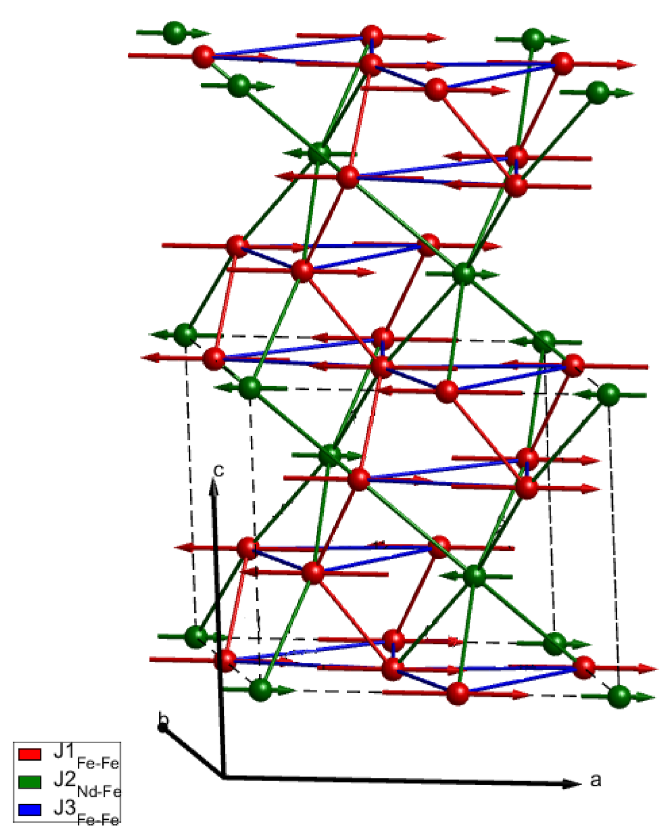
P.G.Matveeva[1,2] , A.K. Ovsyanikov[1,3] , M. Boehm [4] , I.V. Golosovskyi [1] , D.N. Aristov[1,2]
[1] PNPI, Orlova Roscha, Gatchina, [2] Saint Petersburg State University, Saint Petersburg, [3] Peter the Great Saint-Petersburg Polytechnic University, Saint Petersburg, [4] Institut Laue-Langevin, Grenoble, France

Motivation of study

Physical properties of rare-earth ferroborates ($RFe_3(BO_3)_4$), where a large magnetoelectricity is observed, is a topic of large interest today. The knowledge of magnon spectrum in combination with theoretical calculations allow us to obtain the value of exchange interaction parameters, what is important when we investigate the magnetic structure of multifferoic and the origin and value of its electrical polarisation.

We investigate the spectrum of magnetic excitations in neodymium ferroborate $NdFe_3(BO)_4$. We obtain values of exchange parameters and study their anisotropy.

Spin waves in $NdFe_3(BO)_4$



Pic.1. Magnetic structure of $NdFe_3(BO)_4$

The chemical unit cell of $NdFe_3(BO)_4$ (space group $R32$) contains (besides nonmagnetic ions O, B) 3 iron atoms and one atom of neodymium. Since $NdFe_3(BO)_4$ is an antiferromagnet, we consider 8 atoms in the unit cell. The Hamiltonian of our model is (various interactions are shown in Pic.1):

$$\hat{H} = J_1 \sum_{nn} S_i^{Fe} S_j^{Fe} + \\ + J_2 \sum_{nnn} S_i^{Fe} S_j^{Fe} + J_3 \sum_{nn} S_i^{Nd} S_j^{Fe} \tag{1}$$

We imply the parameter J_3 to be anisotropic (easy plane YZ, $J_3^z = J_3^y > J_3^x$). And $S_{Fe} = 2, S_{Nd} = 1/2$. The theoretical analysis has been performed in frames of linear spin-wave theory by using the Maleev-Dyson representation in the form:

$$\begin{cases} S_i^+ = \sqrt{2S}a_i = \sqrt{2S}b_i^\dagger \\ S_i^- = \sqrt{2S}a_i^\dagger = \sqrt{2S}b_i \\ S_i^z = S - a_i a_i^\dagger = -S + b_i b_i^\dagger \end{cases}$$

Where $a, b(h.c.)$– bosonic operators of creation (annihilation) of magnons for sublattices with the opposite spin directions. We substitute them into (1) and obtain (after the Fourier transformation):

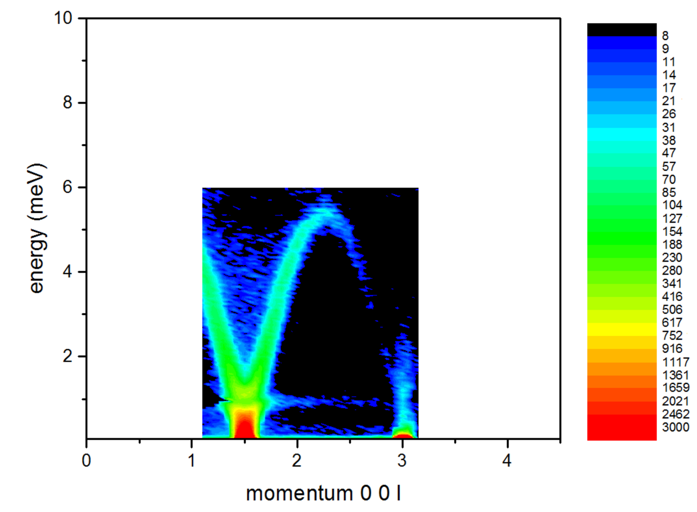
$$\hat{H} = \sum_k \sum_{ij} A_{ij}(k) c^\dagger(k) c_j(k) + B_{ij}(k) c_i c_j + h.c. \tag{2}$$

Where $c_i = (a_i, b_i), c_i^\dagger = (a_i^\dagger, b_i^\dagger)$. To obtain spectrum we use equations of motions for operators $c_i, c_i^\dagger$ for each of sublattices. We obtain the spectrum from the requirement of non-trivial solution of the system of linear equations:

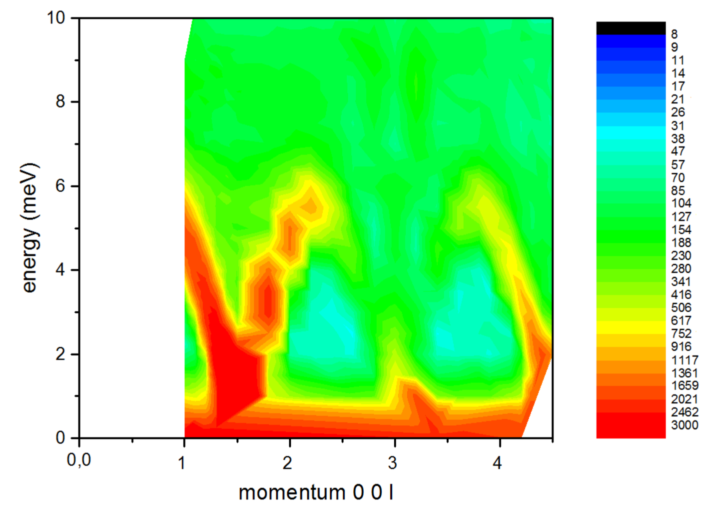
$$(\mathbf{A}+\mathbf{B})(\mathbf{A}-\mathbf{B})\vec{C} = \omega^2(q)\vec{C} \tag{3}$$

where $\vec{C} = (\vec{c}, \vec{c}^\dagger)^T$ and matrices $\mathbf{A}, \mathbf{B}$ (both 16×16) depend on exchange parameters, values of spins and mutual positions of interacting atoms.

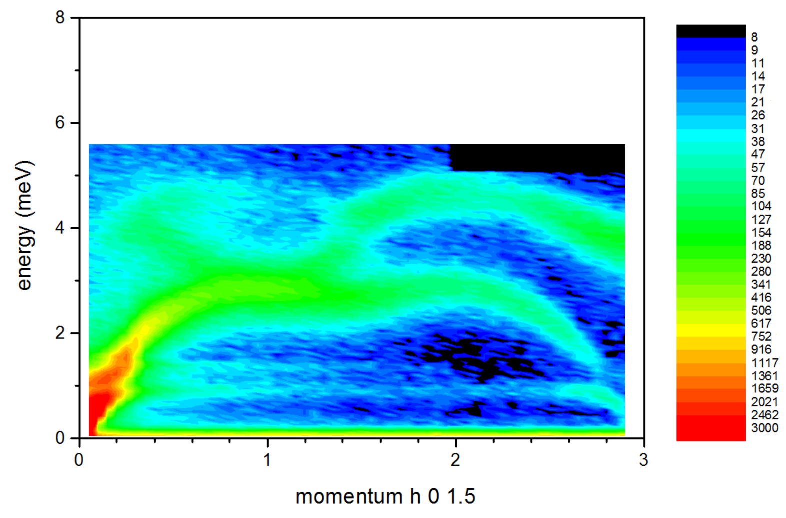
Experimental results



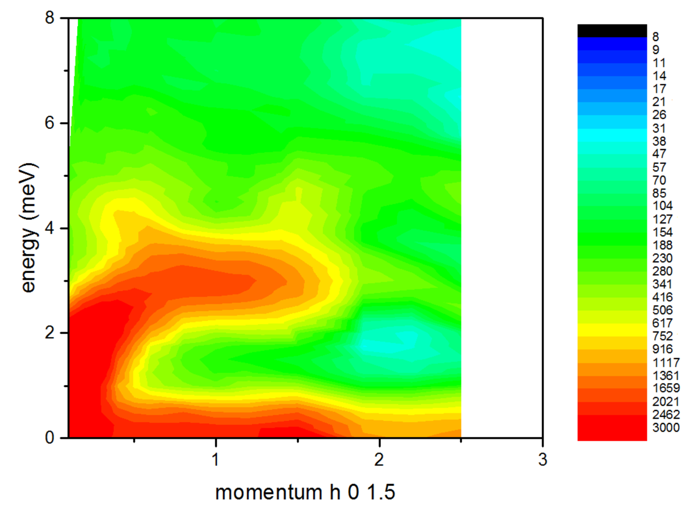
Pic. 2 Map of intensity and spectral branches in $(00l)$ direction from Thales



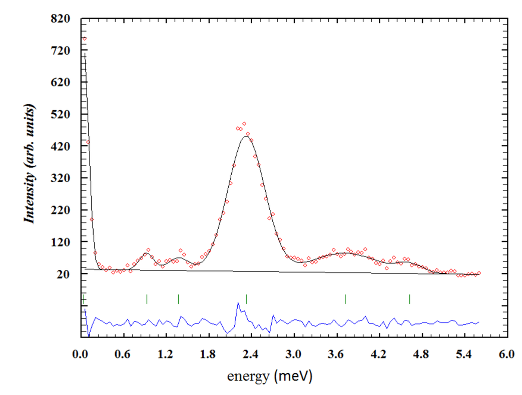
Pic. 3 $(00l)$ direction from IN22



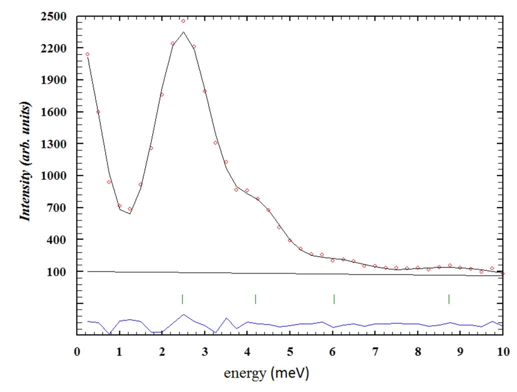
Pic. 4 $(h01.5)$ direction from Thales



Pic. 5 $(h01.5)$ direction from IN22



Pic. 6 Intensity scan from Thales

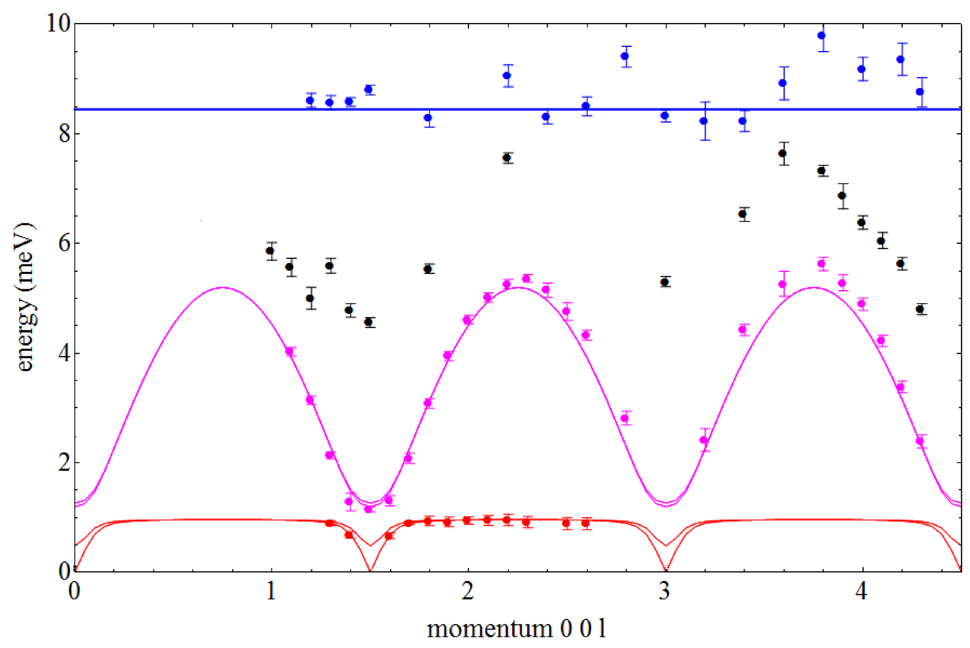


Pic. 7 Intensity scan from IN22

Numerical calculations

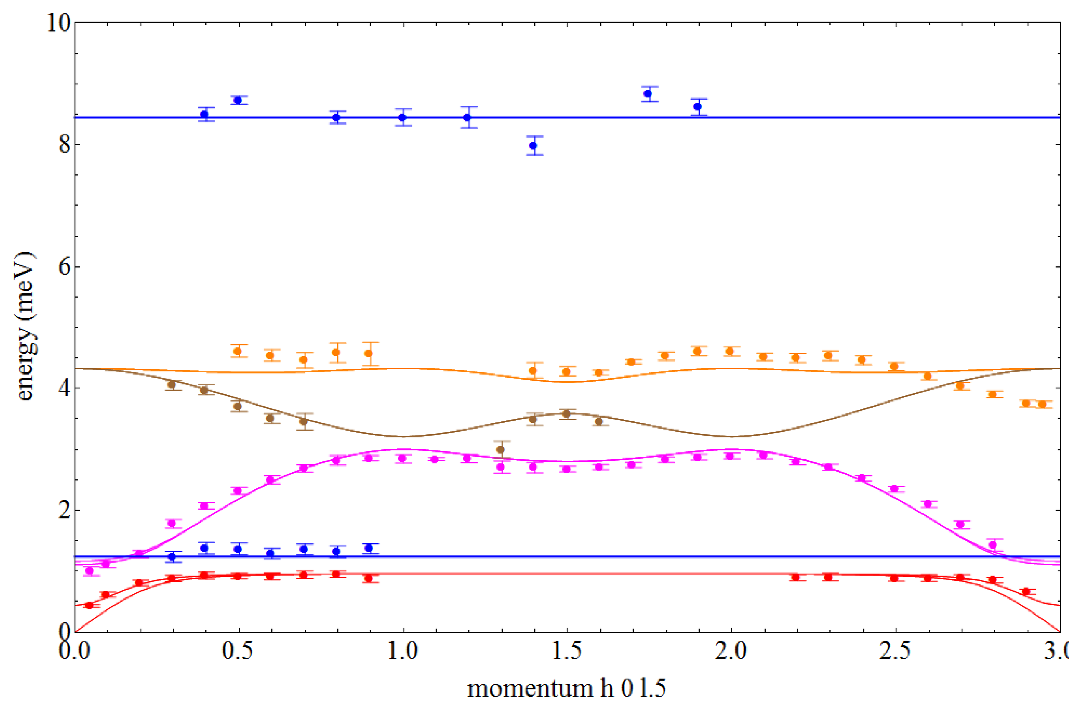
We connect observable splitting of low-energy branch with the existence of easy-plane anisotropy of J_3 and excitation of mode with $J = 15/2$ of Nd GS Kramers doublet.

The ratio of magnon branch splitting at $q = 0$, caused by anisotropy, and Kramers doublet splitting were also measured by optical methods,corresponding energies are $\delta E = 0.46meV$[1] for splitting in magnon spectrum at $q = 0$ and $\Delta = 1,1meV$[2] for Kramers doublet mode.



Pic. 8 Calculated spectral branches in $(00l)$ direction

Blue lines–crystal field modes, Black dots–excitations in two-magnon continuum [3]



Pic. 9 Calculated spectral branches in $(h01.5)$ direction

References

1)A. M. Kuz'menko, A. A. Mukhin et al., JETP Letters, Vol. 94, No. 4 (2011)
2) M. N. Popova et al., Phys. Rev. B 75, 224435 (2007)
3) D. Fausti et al., Phys. Rev. B 74,024403 (2006)

Results

The obtained values of exchange parameters are: $J_1 = (0.55 \pm 0.07)meV$, $J_2 = -(0.04 \pm 0.01)meV$
$J_3^z = J_3^y = (0.042 \pm 0.01)meV$,
$J_3^x = (0.035 \pm 0.01)meV$