Experimental report

21/10/2019

Proposal: 7-02-181 **Council:** 10/2018

Title: Soft phonon modes in RFe3(BO3)4 crystals

Research area: Physics

This proposal is a new proposal

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Samples: TbFe3(BO3)4
NdFe3(BO3)4
PrFe3(BO3)4

Instrument	Requested days	Allocated days	From	To
THALES	6	0		
IN8	6	2	25/06/2019	27/06/2019

Abstract:

We are planning to study phonon excitations in single crystals of TbFe3(B^11O3)4 and NdFe3(B^11O3)4 undergoing the structural phase transition (R32 -> P3_121) and without it for understanding the mechanism of the structural instabilities.

Introduction

In crystals of $RFe_3(BO_3)_4$ at R = Eu - Er, Y, a structural phase transition of the displacement type from a phase with space group R32 to a phase with space group $P3_121$ is observed with decreasing temperature [1,2]. In this transition, the point symmetry (group D3) of the crystal does not change, only translational symmetry changes, the cell volume increases three times. Moreover, depending on the rare-earth ion, the transition temperature varies in a very wide range from 88 K to 450 K [1]. In crystals where R = La - Sm, a structural transition was not experimentally observed.

In [3], we showed that the phase transition R32 → $P3_121$ is associated with the boundary point of the Brillouin zone Λ ($\mathbf{q}_\Lambda$ = 1/3 ($\mathbf{b}_1$ - 2$\mathbf{b}_2$ + $\mathbf{b}_3$); $\mathbf{b}_1$, $\mathbf{b}_2$, $\mathbf{b}_3$ are reciprocal lattice vectors). In [4], we calculated the lattice dynamics of $RFe_3(BO_3)_4$ crystals (R = Pr, Nd, Sm, Gd, Tb, Dy, Ho) in the highly symmetric phase with the space group R32. Significant changes in the spectra of crystals with various rare-earth ions were obtained only in the vicinity of the boundary point Λ of the Brillouin zone for acoustic vibration branches. A softening of frequency of the transverse acoustic mode at the Λ point was found in all the studied compounds (even in compounds in which no structural phase transition is observed). It was found that the magnitude of this frequency depends on the type of the rare-earth ion and decreases from the compound with praseodymium to the compound with holmium down to imaginary values.

In the work [5] the phonon dispersion in terbium iron borate $TbFe_3(BO_3)_4$ has been measured by inelastic neutron scattering in a temperature range 180 < T < 350 K through the displacive structural transition at T_S = 192 K. Measurements were performed at the Cold Neutron Chopper Spectrometer at Oak Ridge National Laboratory. Significant, but not complete, softening of the transverse acoustic (TA) branch has been observed at the corner of the Brillouin zone (Λ point) at a temperature near T_S. The transverse acoustic soft mode undergoes considerable broadening at the Λ point near the transition temperature that can be attributed to the anharmonic interference between transverse acoustic and optical modes. Only three symmetry types of the vibrations are possible at the boundary point Λ of the Brillouin zone in the R32 phase of TbFe3(BO3)4, and all three acoustic modes at this point have different symmetries. Therefore, if there is an unstable optical mode close to the acoustic ones, then its symmetry will necessarily coincide with the symmetry of the vibration of one of the acoustic modes at this point. When the temperature is lowered, the frequency of the soft optical mode gradually decreases. The TO and TA modes of the same symmetry cannot cross each other, implying the anharmonic interaction between two modes.

In this experiment we investigated the existence of instability near Λ point in oxyborates without the structural phase transition. The soft mode can be exist but not induce the phase transition due to the same mechanism of damping the unstable optical mode by stable acoustic mode. For this purpose we've grown the single crystals of $PrFe_3(B^{11}O_3)_4$ and $NdFe_3(B^{11}O_3)_4$ without the phase transition.

The single crystals of $PrFe_3(B^{11}O_3)_4$ and $NdFe_3(B^{11}O_3)_4$ with a mass of ~ 3 g was grown from flux. The samples was prepared with ^{11}B enriched to 99% to minimize the strong neutron absorption of natural boron. Both single crystals was oriented in the (H0L) scattering plane (hexagonal lattice settings are being used).

Results

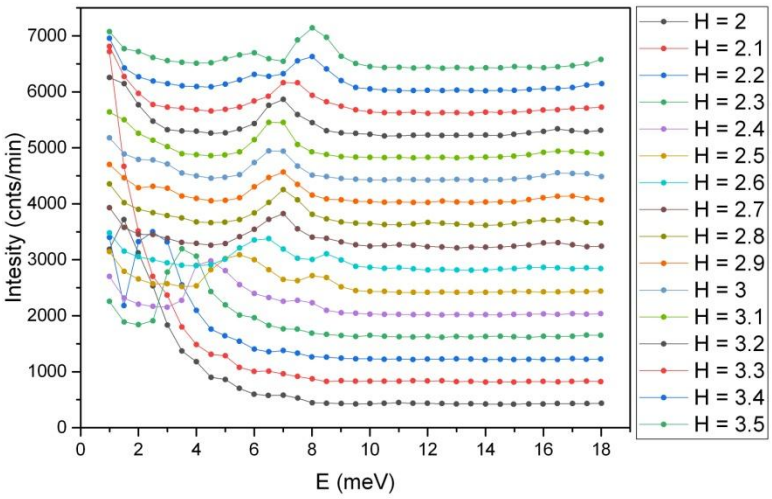

Figure 1. Constant-Q scans at Q = (H 0 4) at T = 50 K for $PrFe_3(B^{11}O_3)_4$

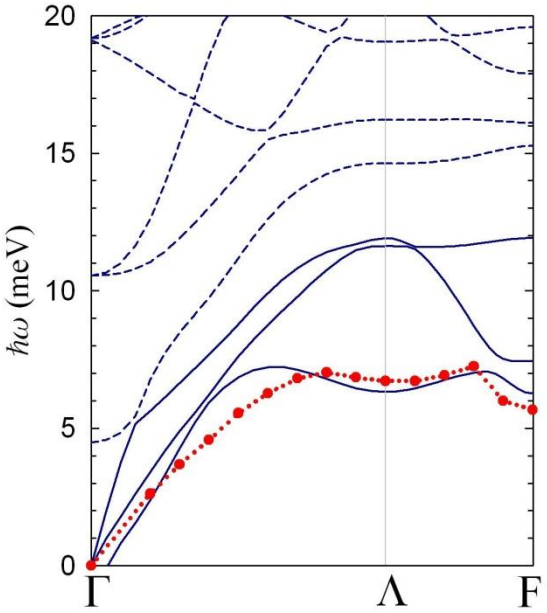

Figure 2. Comparison between experimentally observed TA phonon branch (red line, T = 50K) and calculated (blue line, solid lines – dispersion curves of acoustic modes, dashed lines - dispersion curves of optical modes) TA phonon dispersion along the Γ→Λ direction.

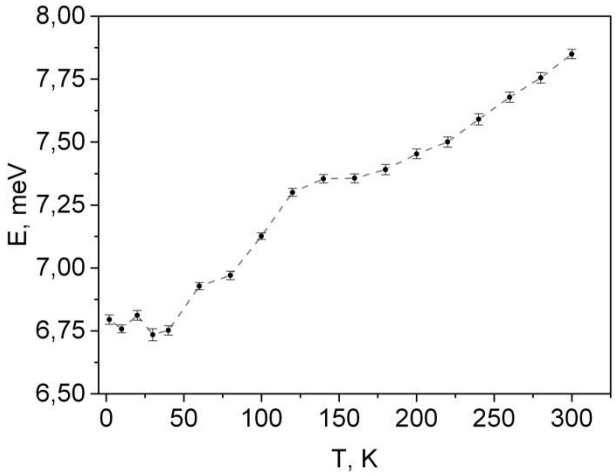

Figure 3. Temperature dependence of TA mode energy at Q = (3 0 4) corresponding to Λ point

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[2] S.A. Klimin, D. Fausti, A. Meetsma, L.N. Bezmaternykh, P.H.M. van Loosdrecht, T.T.M. Palstra. Acta Crystallogr. B 61, 481 (2005).
[3] V.I. Zinenko, M.S. Pavlovskii, A.S. Krylov, I.A. Gudim, and E.V. Eremin. JETP Vol. 117, 1032 (2013).
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[5] M.S. Pavlovskiy, K.A. Shaykhutdinov, L.S. Wu, G. Ehlers, V.L. Temerov, I.A. Gudim, A.S. Shinkorenko, A. Podlesnyak. Phys. Rev. B 97, 054313 (2018).