Structural, magnetic and electronic transtions in $Ba_3NbFe_3Si_2O_{14}$ at high pressures up to 70 GPa by synchrotron Mössbauer source spectroscopy

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Iron-containing crystals of the langasite family $A_3MFe_3X_2O_{14}$ (A = Ba, Sr; M = Sb, Nb, Ta; X = Si, Ge) are a new class of multiferroics in which ferroelectricity can be induced by magnetic ordering. [1-4]

$Ba_3NbFe_3Si_2O_{14}$ (BNFS) has a layered crystal structure (eg gr. *P*321, Z = 1), in which the Fe^{3+} and Si^{4+} ions occupy 3*f* and 2*d* tetrahedral sites in the *ab* plane, respectively.[5] At ambient pressure, BNFS is an antiferromagnetic with T_N = 27.1 K. The unit cell parameters obtained recently from single-crystal refinement are *a* = 8.52422(8) and *c* = 5.23372(5) Å.[5] Below T_N, Fe ions form a net of triangle clusters on a hexagonal lattice in the *ab* planes creating the triangle magnetic structure with frustrated interactions (Fig. 1a). In addition, the helical arrangement of iron magnetic moments was observed from plane to plane with a period of about 7 unit cells [6] (Fig. 1b).The low-temperature magnetic transition induces a structural transition *P*321 → *P*3 (or *P*321 → C2), which favors the ferroelectric state.

In this work, the structural and magnetic properties of BNFS crystals at high-hydrostatic pressures (up to 70 GPa) created in diamond anvil cells were investigated at temperatures from 4.2 to 300 K. Synchrotron Mossbauer Source spectroscopy was used to study several phase transitions under pressueres and low temperatures. A cascade of structural phase transitions at *P* = 3, 18, and 45 GPa was detected.[7,8] The most drastic changes in the structure and magnetic properties are observed during the second transition at about 18 GPa, where the unit cell parameter *c* strongly decreases, and the cell volume drops abruptly by 7%. Mossbauer studies at low temperatures showed that this transition significantly changes the magnetic properties of the crystal. In particular, we found a colossal increase in the Neel point up to 100 K, which is almost four times higher of the corresponding value at ambient pressure (27 K).

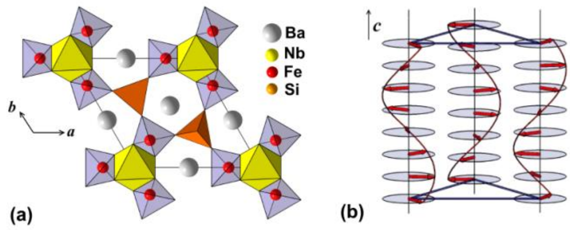

Figure 1.(a) *ab* projection of $Ba_3NbFe_3Si_2O_{14}$ crystal structure. (b) Helical spin ordering of Fe magnetic moments along the *c* axis.

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