Structural, magnetic and electronic transitions in Ba$_3$NbFe$_3$Si$_2$O$_{14}$ at high pressures up to 70 GPa by synchrotron Mössbauer source spectroscopy

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Iron-containing crystals of the langasite family A$_3$MFe$_3$X$_2$O$_{14}$ (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$_3$NbFe$_3$Si$_2$O$_{14}$ (BNFS) has a layered crystal structure (e.g. gr. P321, 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 $P321 \rightarrow P3$ (or $P321 \rightarrow C2$), 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 Mössbauer Source spectroscopy was used to study several phase transitions under pressurues 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%. Mössbauer 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).

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