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Magnetic phase separation and strong enhancement of the Neel temperature at high pressures in a new multiferroic $Ba_3TaFe_3Si_2O_{14}$

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Langasites ($La_3Ga_5SiO_{14}$) crystals are famous due to their unique piezoelectric, laser and nonlinear optical properties [1, 2]. Iron-containing langasites, such as $Ba_3TaFe_3Si_2O_{14}$, $Ba_3NbFe_3Si_2O_{14}$ are now of great interest as a new type of multiferroics [3–10].

$Ba_3TaFe_3Si_2O_{14}$ has non-centrosymmetric trigonal structure of the $Ca_3Ga_2Ge_4O_{14}$ type with space group $P321$ and $Z = 1$. It has a layered structure where iron Fe^{3+} and silicon Si^{4+} ions occupy the $3f$ and $2d$ tetrahedral sites in the ab plane, respectively. The Fe^{3+} ions form a net of triangle clusters on a hexagonal lattice in the ab planes creating the triangle magnetic structure with frustrated interactions [3, 6] The neutron diffraction studies of the similar crystal $Ba_3NbFe_3Si_2O_{14}$ [3, 5] revealed the 120° – ordering of Fe magnetic moments in the ab plane at low temperatures. In addition, the magnetic vector rotates from layer to layer (in the ab plains), when alternating along the c axis, thus forming a helical magnetic structure [3].

Transmission Mossbauer spectroscopy revealed splitting of crystallographic iron sites into two sublattices at ambient pressures [4, 6] which was associated with the structural phase transition induced by the magnetic ordering below Neel temperature $T_N = 27.2$ K. At high pressures $P > 20$ GPa the giant increasing of the Neel temperature up to 120 K was found by Nuclear Forward Scattering (NFS) [11, 12]. However the information about non-equivalent iron sites and magnetic structure at high pressures is not obtained from NFS spectra.

In the present paper, the polycrystalline samples of langasite $Ba_3TaFe_3Si_2O_{14}$ were studied under high pressures in a diamond-anvil cell (DAC) [13] by a new method of synchrotron Mossbauer spectroscopy (SMS) [14–16] in the temperature range of 4.2–295 K and at pressures from ambient to 30 GPa. At pressure above the structural transition ($P > 20$ GPa), the separation of the sample into two magnetic phases with different T_N values of about 50 and 130 K was revealed (Fig. 1). This effect can be explained by redistribution of Fe ions over $3f$ and $2d$ tetrahedral sites of the langasite structure. In this case, the short Fe–O distances and favorable Fe–O–Fe bond angles create conditions for strong superexchange interactions between iron ions in the (ab) plane providing effective two-dimensional (2D) magnetic ordering. In addition, the significant reduction of the c parameter should lead to a substantial increase in the Fe–O–O–Fe exchange interaction between adjacent ab planes, which further leads to an increase in the Neel point.

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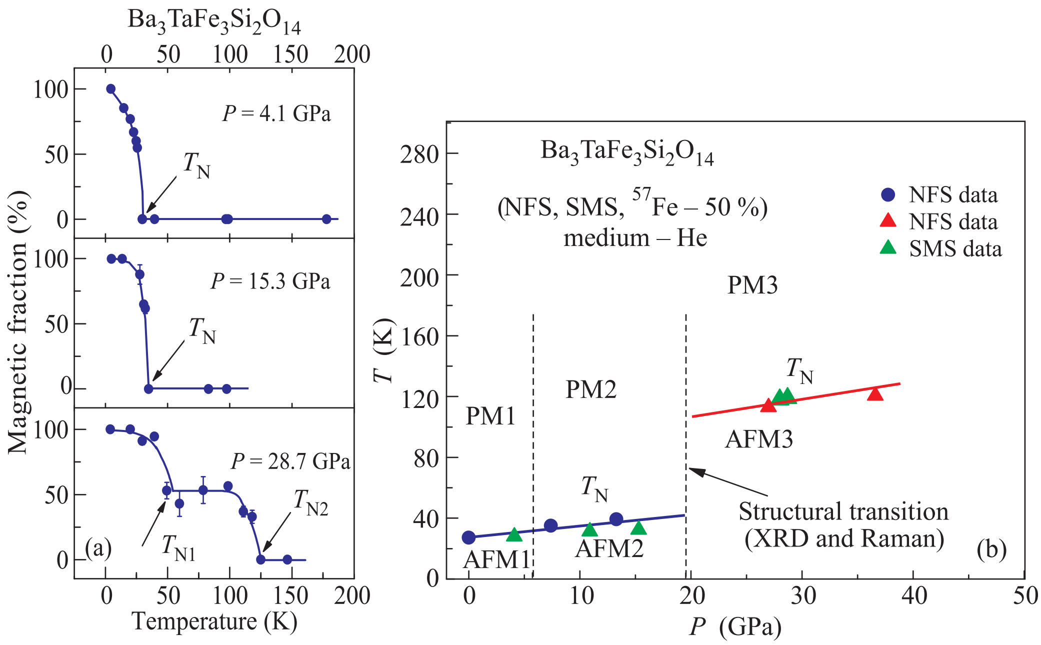

Fig. 1. (Color online) (a) – Temperature dependences of areas of magnetic (blue points) components in the Mössbauer spectra of $Ba_3TaFe_3Si_2O_{14}$ at different pressures below and above the structural transition at $P = 20$ GPa. (b) – Magnetic (P–T) phase diagram of $Ba_3TaFe_3Si_2O_{14}$

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