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

I. S. Lyubutin[a 1)], *S. S. Starchikov*[a,b], *A. G. Gavriliuk*[a,b,c], *I. A. Troyan*[a,b,c], *Yu. A. Nikiforova*[a,b], *A. G. Ivanova*[a,b], *A. I. Chumakov*[d,e], *R. Rüffer*[d]

[a] *FSRC "Crystallography and Photonics" of Russian Academy of Sciences, 119333 Moscow, Russia*

[b] *Institute for Nuclear Research, Russian Academy of Sciences, 142190 Moscow, Troitsk, Russia*

[c] *REC "Functional Nanomaterials", Immanuel Kant Baltic Federal University, 236041 Kaliningrad, Russia*

[d] *European Synchrotron Radiation Facility, CS40220, F-38043, Grenoble, France*

[e] *National Research Center "Kurchatov Institute", 123182 Moscow, Russia*

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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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1) e-mail: lyubutinig@mail.ru

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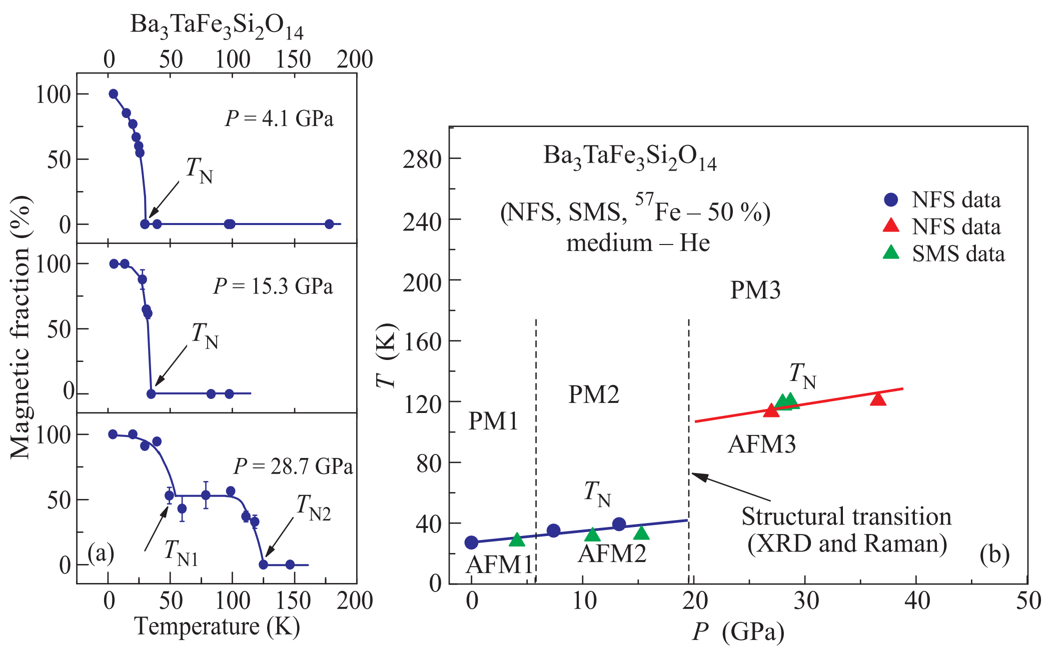

Fig. 1. (Color online) (a) – Temperature dependences of areas of magnetic (blue points) components in the Mössbauer spectra of $Ba_3TaFe_3Si_2O_{14}$ at different pressures below and above the structural transition at $P = 20$ GPa. (b) – Magnetic (P–T) phase diagram of $Ba_3TaFe_3Si_2O_{14}$

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