

"Spatially modulated spin ordering in  
compounds with a langasite type crystal structure:  $^{57}\text{Fe}$  Mössbauer reinvestigation"

Langasite family compounds based on the  $\text{La}_3\text{Ga}_5\text{SiO}_{14}$  mineral attract high interest that mainly caused by their multiferroic properties that are closely related to the unusual magnetic structure of these compounds. The Type-II multiferroic langasite-like  $\text{Ba}_3\text{SbFe}_3\text{Si}_2\text{O}_{14}$  oxide has a noncollinear magnetic structure with a magnetic ordering temperature  $T_N \sim 34\text{K}$ . The magnetic helix structure of  $\text{Ba}_3\text{SbFe}_3\text{Si}_2\text{O}_{14}$  below  $T_N$  is a result of competing antiferromagnetic intralayer and interlayer interactions.

The present work reports new analysis and reinterpretation of the  $^{57}\text{Fe}$  Mössbauer data [1] for the ferrite  $\text{Ba}_3\text{SbFe}_3\text{Si}_2\text{O}_{14}$  with langasite-like crystal structure. The sample enriched with  $^{57}\text{Fe}$  isotopes was obtained by the solid-phase synthesis method. The stoichiometric amounts of pre-dried  $\text{BaCO}_3$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{Sb}_2\text{O}_5$ ,  $\text{SiO}_2$  were used as precursors. The annealing was carried out at two stages: at  $1000^\circ\text{C}$  and  $1225^\circ\text{C}$  for 24 hours.

$^{57}\text{Fe}$  Mössbauer spectra recorded above  $T_N \approx 34\text{ K}$  consist of a quadrupole doublet with a high quadrupole splitting ( $\Delta_{35\text{K}} \approx 1.32\text{ mm/s}$ ) underlying distorted local surrounding of the unique  $\text{Fe}^{3+}$  sites. The self-consistent calculations of the lattice contribution to the electric field gradient (EFG) at  $^{57}\text{Fe}$  nuclei have show a significant dipole contribution to the EFG with the oxygen polarizability of  $\alpha_O \approx 0.4\text{ \AA}^3$ . In the temperature range  $T < T_N$ , Mössbauer spectra are consistent with a modulated helical magnetic order, where iron magnetic moments rotate within the  $(ab)$  plane with propagation vector along  $c$  axis. All experimental spectra at  $T < T_N$  were analyzed assuming the modulation of the electric hyperfine interactions and the anisotropy of the magnetic hyperfine field  $H_{hf}$  at the  $^{57}\text{Fe}$  nuclei when the  $\text{Fe}^{3+}$  magnetic moment rotates with respect to the principal axis of the EFG tensor. The anisotropic part of hyperfine field have been given in terms of the hyperfine coupling tensor ( $A_{xx} > A_{zz} > A_{yy}$ ) and the dipolar fields ( $A_{\text{dip}}$ ) arising from external iron moments  $\mu_{\text{Fe}}$ . The obtained from the fitting a small bunching parameter ( $\xi \approx 0.05$ ) of the spiral spin structure results from easy-axis anisotropy in the plane of the iron spin rotation. The Bean-Rodbell magnetostriction exchange model was used to describe the temperature evolution of the reduced hyperfine field.

## References

- 1) I. S. Lyubutin et al., JETP Letters, 12, 798 (2014).