Accepted Manuscript

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S1293-2558(16)30546-5 DOI: 10.1016/j.solidstatesciences.2016.10.012

Reference: SSSCIE 5404

PII:

To appear in: Solid State Sciences

Received Date: 8 August 2016

Accepted Date: 18 October 2016

Please cite this article as: E.S. Zhukova, A.S. Mikheykin, V.I. Torgashev, A.A. Bush, Y.I. Yuzyuk, A.E. Sashin, A.S. Prokhorov, M. Dressel, B.P. Gorshunov, Crucial influence of crystal site disorder on dynamical spectral response in artificial magnetoplumbites, Solid State Sciences (2016), doi: 10.1016/ i.solidstatesciences.2016.10.012.

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Crucial influence of crystal site disorder on dynamical spectral response in artificial magnetoplumbites

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1. Abstract.

High quality PbFe₁₂O₁₉ single crystals were grown by a solution-melt crystallization technique and characterized by synchrotron radiation diffraction structural studies, Mössbauer, Raman and infrared spectral measurements. In the terahertz and sub-terahertz range two absorption bands are observed, which are associated with the dynamics of structurally disordered lead and iron ions. We develop a structural model that comprises Fe^{2+} ions at the $(1/2 \times 4e)$ (0, 0, z) position with bipyramidal oxygen environment, and Pb ions at the (24l) (x, y, z) position with generic strongly distorted 12 vertex polyhedron of oxygen. The atomic displacement ellipsoids of the oxygen surrounding the lead ion are stretched towards the Pb ion within the plane perpendicular to the hexagonal axis. The suggested model is in full agreement with the local symmetry of the Fe2 and Pb ions and with the chemical composition of the sample determined by X-ray analysis.

2. Introduction and motivation.

Compounds with chemical formula AFe₁₂O₁₉ (A=Ba, Sr, Pb) comprise a large class of ferromagnetic oxides called M-type hexaferrites that are structurally isomorphic to magnetoplumbite mineral. A member of the family, the natural magnetoplumbite PbFe₁₂O₁₉ has a complex chemical composition due to the presence of various oxides like MnO, Mn₂O₃, Al₂O, TiO, and its actual formula is Pb₂Fe₁₅Mn₇(A1Ti)O₃₈. Although the natural form of the mineral does not contain any Fe²⁺ ions, it exhibits a considerable residual magnetization that makes it particularly attractive for industrial applications. Indeed, one of the various artificial magnetoplumbite phases, Pb²⁺Fe₁₂³⁺O₁₉, exhibits excellent magnetic properties and it is used for manufacturing permanent magnets [1]. Some of the hexaferrite compounds not only possess outstanding magnetic characteristics, they also show signs of multiferroicity. For example, the diluted compound BaFe_{1-x- δ}Sc_xMg $_{\delta}$ O₁₉ (δ =0.05) with x = 1.6 and x=1.75 reveals the formation of multiferroic phase with the robust magnetoelectric effect [2]. Recent measurements of the artificial magnetoplumbite PbFe₁₂O₁₉ suggest a large residual polarization of P=104 µC/cm² [3], however, the results should be verified since they could be influenced by a rather high conductivity of the studied samples.

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