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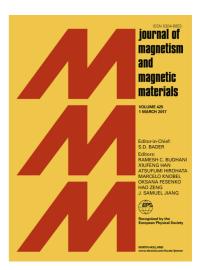
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### ACCEPTED MANUSCRIPT

# Exchange-coupling of hard and soft magnetic sublattices and magnetic anomalies in mixed spinel NiFe<sub>0.75</sub>Cr<sub>1.25</sub>O<sub>4</sub> nanoparticles

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#### **Abstract**

A set of single-crystalline nanoparticles (NPs) of nickel-chromium ferrite NiFe $_{0.75}$ Cr $_{1.25}$ O<sub>4</sub> with a cubic spinel structure were synthesized and investigated. The NPs size can be varied from about 5 to 50 nm by the final annealing of the precursor at different temperatures. The distribution of cations over the tetrahedral (A) and the octahedral [B] sites (Fe $_{0.75}$  Ni $_{0.25}$ ) [Ni $_{0.75}$  Cr $_{1.25}$ ] O<sub>4</sub> was established from the magnetic and Mössbauer measurements. In large NPs, the magnetic structure at low temperatures is close to the collinear antiferromagnetic (AFM) structure of the Neel type; and the total magnetic moment  $M_{tot}$  of the ferrite coincides with the direction of the B-sublattice moment. Several size-dependent magnetic anomalies were revealed. Three types of magnetic ions present in the A- and B- sublattices cause the competition of AFM and FM exchange interactions resulting in the highly frustrated magnetic ordering and the occurrence of canted magnetic structure in the octahedral B-sublattice. The frustrated structure is very flexible and significantly subjected to temperature and applied field. It results in several magnetic anomalies observed, including the occurrence of magnetic compensation, abnormal behavior of ZFC and FC magnetization curves and hysteresis loops. It was shown that magnetic anomalies can be explained in terms of exchange coupling of "soft" and "hard" magnetic B-and A-sublattices. This effect in the (Fe $_{0.75}$  Ni $_{0.25}$ ) [Ni $_{0.75}$ Cr $_{1.25}$ ] O<sub>4</sub> NPs can be considered as an *atomic-scale analog* of a similar effect observed in two-phase exchange-coupled alloys developed for permanent magnets and for the perpendicular recoding media.

**Keywords**: nickel-chromium-ferrites nanoparticles, frustrated magnetic system, magnetic anomaly, compensation temperature, hard - soft magnetic sublattices, Mössbauer spectroscopy

#### 1. Introduction

The perspective of magnetic materials with the spinel type crystal structure originates from their structural, magnetic and electronic features modifiable by introducing different metal ions into the local crystal sites [1,2]. Possibility of tuning of magnetic properties, easy and cheap production, make them widely used in electronic devices, for magnetic record, ferro-liquids, biosensors and other applications [3]. For binary spinels with cubic space group  $Fd\bar{3}m$ , the general formula is  $(N^{2+}_{1-x}M^{3+}_{x})[N^{2+}_{x}M^{3+}_{2-x}]$  O<sub>4</sub>. Round and square brackets denote tetrahedral and octahedral positions, respectively, and x is a parameter of inversion. When x = 0, all bivalent ions are located on tetrahedral sites and such spinel is called "normal". If x = 1, half of the trivalent ions are in tetrahedral sites, while all bivalent and other trivalent ions are in octahedral sites, and this spinel is called "inverse".

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