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New Insights on the Spin Glass Behavior in Ferrites Nanoparticles

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Abstract: The magnetic properties of nanocrystalline $M_xFe_{3-x}O_4$ ferrites with M=Fe, Co, and Zn were investigated. The data support a core–shell model, where the core is ferrimagnetically ordered, and the shell shows a spin glass type behavior. The reduced magnetizations of spin glass components follow an $m_g = (1 - b/H^{-1/2})$ field dependence. The b values are strongly correlated with the intensities of exchange interactions. The field dependences of the magnetoresistances of Fe_3O_4 and $Zn_xFe_{3-x}O_4$ nanoparticles pellets, experimentally determined, are well described if instead of the core reduced magnetization, commonly used, that of the shell is taken into account. For similar compositions of the nanoparticles, identical *b* values are obtained both from magnetization isotherms and magnetoresistances studies. The half-metallic behavior of spinel Fe_3O_4 based nanoparticles is discussed comparatively with those of double perovskites.

Keywords: ferrite nanoparticles; magnetic properties; spin-glass; exchange field

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Citation: Burzo, E.; Tetean, R. New Insights on the Spin Glass Behavior in Ferrites Nanoparticles. *Nanomaterials* **2022**, *12*, 1782. https://doi.org/10.3390/nano12101782

Academic Editor: Detlef W. Bahnemann

Received: 14 April 2022
Accepted: 21 May 2022
Published: 23 May 2022

1. Introduction

Ferrite nanoparticles with a spinel-type structure have garnered a great deal of attention due to their basic properties and applications in various fields such as medicine [1], adsorption potential to abate heavy metals and dyes from aqueous solutions [2], catalytic properties [3], magnetoresistive devices [4], etc. The Fe_3O_4 based nanoparticles with spinel-type structure can be described as core–shell systems, where the structure and magnetic properties of the shell are different from that of the core [5].

The bulk magnetite $(Fe^{3+})_A[Fe^{3+}Fe^{2+}]_B$, at ambient temperature, has a cubic inverse spinel-type structure. In this lattice, the O^{2-} anions form an *fcc* type lattice, the Fe^{3+} ions being located in tetrahedral interstices (A) and the Fe^{3+} and Fe^{2+} in the octahedral interstices (B). The nature of spinel structures, such as normal, inverse, or mixed in substitutional ferrites $Fe_{3-x}M_xO_4$ with M=Co or Zn, relies on lattice occupancy by these ions. The Zn^{2+} ions are mainly located in tetrahedral sites [6,7]. The cobalt ferrites exist as partially inverse spinel structures in which both A and B sites contain a fraction of Co^{2+} ions, the largest being located at B sites [5,8].

Upon cooling, bulk Fe_3O_4 displays a sharp Verwey transition at the temperature T_V = 122 K, characterized by a structural transition from a cubic to monoclinic lattice together with an abrupt drop in the electrical conductivity, associated with a "freezing out" of the electron hopping between the Fe^{2+} and Fe^{3+} ions in B sublattice, which is the primary conduction mechanism at temperatures $T > T_V$ [9].

The surface structure of magnetite differs from that of bulk material. A large number of studies were performed in order to analyze the Fe_3O_4 surfaces. There are two possible truncations [10]. At the Fe_3O_4 (111) surface, three distinct terminations are observed, exposing either a close-packed oxygen plane, Fe_A, or Fe_B atoms [11]. The stable Fe_3O_4 (111) termination might have oxygen [12,13] or contain a fraction of iron and oxygen monolayers exposed over a closely packed oxygen layer [14,15]. In the case of Fe_3O_4 (100), surface terminations with ordered oxygen vacancies or Fe adatoms were proposed [16].