



## Research articles

Observation of extraordinarily large magnetization in CoFe<sub>2</sub>O<sub>4</sub>/ZnFe<sub>2</sub>O<sub>4</sub> bilayersB.N. Sahu<sup>a</sup>, S.C. Sahoo<sup>b</sup>, N. Venkataramani<sup>c</sup>, Shiva Prasad<sup>a,d,\*</sup>, R. Krishnan<sup>e</sup><sup>a</sup> Department of Physics, Indian Institute of Technology, Bombay, Powai, Mumbai 400076, India<sup>b</sup> Department of Physics, Central University of Kerala, Tejaswini Hills, Periya, Kasaragod, Kerala 671320, India<sup>c</sup> Department of Metallurgical Engineering and Materials Science, Indian Institute of Technology Bombay, Mumbai 400076, India<sup>d</sup> Institute of Infrastructure Technology, Research and Management, Maninagar (E), Ahmedabad 380 026, India<sup>e</sup> Retired Scientist, Groupe d'Etude de la Matière Condensée, CNRS/Université de Versailles-St-Quentin Versailles Cedex, 78035 France

## A B S T R A C T

In this paper, we report our observation of an extraordinarily large value of magnetization in an annealed CoFe<sub>2</sub>O<sub>4</sub>/ZnFe<sub>2</sub>O<sub>4</sub> bilayer. Sets of bilayers of CoFe<sub>2</sub>O<sub>4</sub>/ZnFe<sub>2</sub>O<sub>4</sub> were deposited at ambient temperature, using pulsed laser deposition setup equipped with Nd: YAG laser, and subsequently annealed in air at different temperatures for 2 h. The highest attained value of magnetization was  $\approx 7200$  G at 300 K and  $\approx 15000$  G at 10 K for a bilayer with a 60 nm thickness  $t_{\text{Co}}$  of CoFe<sub>2</sub>O<sub>4</sub> and 40 nm thickness  $t_{\text{Zn}}$  of ZnFe<sub>2</sub>O<sub>4</sub>, annealed at 650 °C. This measured magnetization value is the highest for any oxide spinel reported thus far in the literature. The observed value is close to that of metallic Ni and cannot be explained by current theories of magnetism in oxides.

Ferrites with chemical formula MFe<sub>2</sub>O<sub>4</sub>, are composed of O<sup>2-</sup>, Fe<sup>3+</sup> and M<sup>2+</sup> ions (M is a single or a combination of several divalent 3d-transition metals), and crystallize with the spinel structure. Its lattice type is face centered cubic, formed by O<sup>2-</sup> ions. There are two sites, namely tetrahedral (A) and octahedral (B), in this structure that are occupied by M<sup>2+</sup> and Fe<sup>3+</sup> ions. Spinel ferrites are ferrimagnetically ordered. According to the Néel model, net magnetic moment of any spinel ferrite is given by  $(\mu_B - \mu_A)$ , where  $\mu_B$  and  $\mu_A$  are the magnetic moments on the 'B' and 'A' sites respectively, considering collinear moments in both the sites. The ferrimagnetic order results from a dominant antiferromagnetic super exchange interaction between the magnetic moments on the 'A' and 'B' sites dictating the resultant spin order in a collinear sense. The cation distribution in bulk spinel ferrites depends on the site preference of the involved divalent metal ions. A wide range of spinel ferrites with varying magnetic properties have been synthesized in the bulk over the years resulting in very useful applications [1–6].

Spinel ferrites are used in high frequency devices due to their high electrical resistivity. A major drawback is their significantly lower magnetization, when compared with many useful metallic magnetic materials. Early workers have shown an increase in magnetic moment with substitution of a non-magnetic ion like Zn<sup>2+</sup> ion on the 'A' site. Substitution of Zn<sup>2+</sup> ion results in an overall increase in the magnetic moment, because of reduction of  $\mu_A$ . However, such a substitution can increase the magnetic moment only up to about 50% substitution on 'A'

sites. Larger substitution of the non-magnetic Zn<sup>2+</sup> ion on the 'A' sites results in the weakening of the super-exchange interaction between the cations on the A and B sites. The weakening of the dominant antiferromagnetic A-B interaction causes a departure from the parallel alignment of moments on the 'B' sites, resulting in a phenomenon called canting [7–10]. Pure Zn ferrite (ZnFe<sub>2</sub>O<sub>4</sub>), which is devoid of magnetic ion on 'A' sites, is thus a paramagnet at room temperature and orders as a weak antiferromagnet  $\approx 10$  K, owing to a weak B-B interaction [11,12].

It has been observed through several experiments that the cation distribution in a nano-crystalline ferrite thin film could differ significantly from their bulk counterpart [13,14]. The cation distribution is observed to be process dependent and often results in magnetization values that are larger than its corresponding compositional bulk value. For example, in Co ferrite (CoFe<sub>2</sub>O<sub>4</sub>), magnetization values higher than bulk have been observed, when the films are deposited at room temperature (RT) and then subsequently annealed at temperatures  $T_A \geq 650$  °C [15]. Similarly, nano-crystalline ZnFe<sub>2</sub>O<sub>4</sub> thin films show magnetic ordering at room temperature, even though the bulk is a paramagnet [13,14]. Hence, a way to increase the magnetic moment of the spinel ferrites is to prepare the materials in nano-crystalline form and look for an enhancement of the magnetic moment. We have been studying such possibilities in double layer spinel ferrite thin films. In the present work, we report an extraordinarily large magnetization value (the highest reported value) obtained in nanocrystalline CoFe<sub>2</sub>O<sub>4</sub>/

\* Corresponding author.

E-mail address: [shiva.pd@gmail.com](mailto:shiva.pd@gmail.com) (S. Prasad).<https://doi.org/10.1016/j.jmmm.2020.167629>

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