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# Ferromagnetic resonance study of the non-stoichiometric double perovskite $\text{Sr}_2\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$

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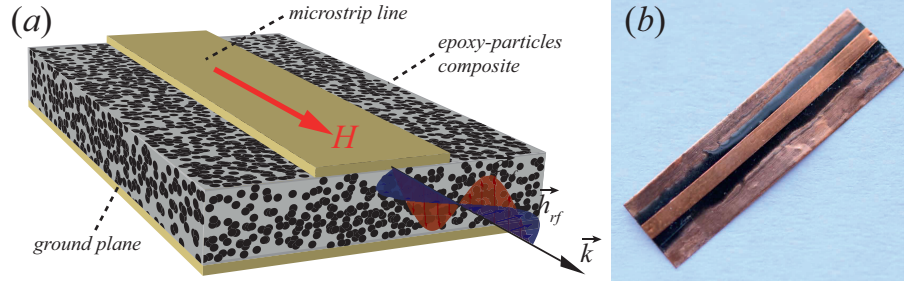
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**Abstract.** In this work we report a ferromagnetic resonance study on the magnetic properties of double perovskite compounds fabricated by solid state reaction. Based on a mean field approach, along with morphological considerations, we accurately determined the saturation magnetization of the non-stoichiometric double perovskite  $\text{Sr}_2\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ . Our approach has revealed a direct influence of composition on the overall magnetic behavior of these materials, providing complementary experimental evidence that corroborates previous theoretical findings. The understanding of the influence of composition is of paramount importance for the design of ferromagnetic oxides with tunable magnetic and magneto-transport behavior.

## INTRODUCTION

Half-metallic ferromagnetic oxides are nowadays interesting materials for their use as sources of spin polarized currents and information storage devices due to its considerable low-field magnetoresistance [1]. The high Curie temperature of the double perovskite  $\text{Sr}_2\text{FeMoO}_6$  makes it a very appealing material for spintronic applications and memory devices [2]. Previous theoretical works have reported on the effect of composition on the magnetic and spin polarization properties of the non-stoichiometric double perovskite  $\text{Sr}_2\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$  [3]. The perfectly ordered lattice of the stoichiometric compound  $\text{Sr}_2\text{FeMoO}_6$ , which consists of alternating Fe and Mo atoms, is modified when mis-sites are created when their positions are exchanged [4]. As a result, Fe (Mo) rich compounds are obtained with expected changes on their magnetic properties. In this work, we provide further experimental understanding on the magnetic properties of double perovskites compounds, which were fabricated by following the solid state reaction procedure reported previously [4]. Ferromagnetic resonance (FMR) and alternating gradient magnetometry (AGM) have been used for the determination of magnetic properties like the saturation magnetization ( $M_s$ ) and the effective anisotropy field ( $H_{eff}$ ). Particularly, FMR has proven to be a very reliable characterization tool for the determination of magnetic and microwave absorption properties of nanomaterials [5, 6], magnetization dynamics [7] and its applications for the development of FMR absorption-based microwave devices [8, 9]. The FMR absorption properties of these materials have revealed a direct influence of the composition on the overall magnetic behavior of these materials. Particularly, we provide simple FMR arguments based on a mean field approach for an accurate determination of the saturation magnetization for different double perovskites. The present investigations provide complementary and quantitative experimental results for the magnetic properties of these materials, which corroborates theoretical findings [3].



**FIGURE 1.** (a) Schematic representation of the microstrip transmission line consisting of a Cu ground plane, an epoxy-perovskite particles composite and the metallic strip line. (b) Photograph of a  $100\ \mu\text{m}$  thick, 1 mm wide and 2 cm long strip line stuck on a real composite.

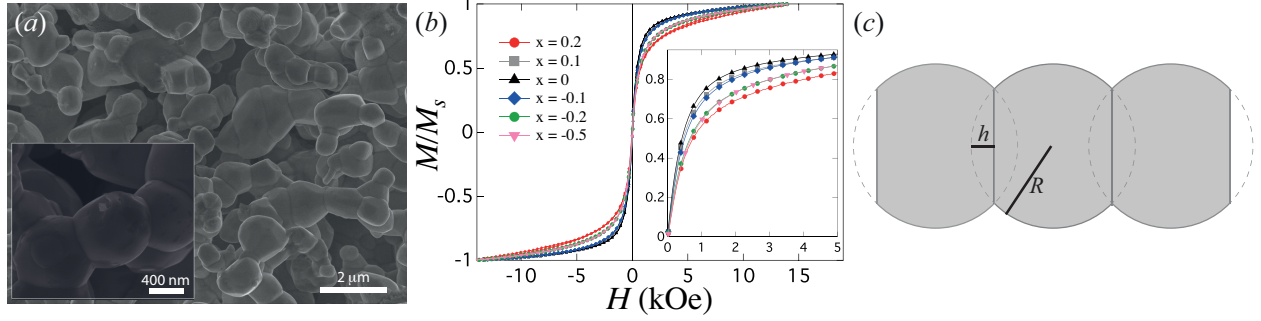
## EXPERIMENTAL DETAILS

Double perovskite compounds  $\text{Sr}_2\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ , with composition in the range  $-0.5 \leq x \leq 0.2$ , have been synthesized as powders by following a stoichiometric reaction procedure [4]. The morphology of the double perovskite compounds was characterized by using a JEOL 7600 field-emission scanning electron microscope (SEM). Each powder sample was subsequently mixed with an epoxy resin in order to obtain resin/particles composites with a ratio of about 10:3. Each mixture was then spread on Cu foils in order to make  $\approx 300 - 400\ \mu\text{m}$  thick supported composite films. The magnetic properties of the composites have been characterized from room temperature AGM and FMR experiments. Magnetization hysteresis loops have been recorded by applying the magnetic field along the plane of the composites in the range  $\pm 14\ \text{kOe}$ . Ferromagnetic resonance experiments were carried out by using a Vector Network Analyzer (VNA) and a strip line technique, which consists of sticking a thin Cu strip line on the free surface of the distinct epoxy-particles composite films, as seen in Fig. 1 (a). A photograph of a  $100\ \mu\text{m}$  thick, 1 mm wide and 2 cm long strip line stuck on a real composite material is shown in Fig. 1 (b). The strip line on the distinct composites are adequate wave-guides to carry out FMR absorption experiments as it permits to explore the microwave absorption characteristics of a large amount of particles. Measurements were done at a constant frequency in the range from 11 – 30 GHz, by sweeping the external DC magnetic field from 10 kOe down to zero field which is applied parallel to the strip line direction. All measurements were performed close to the saturated state in order to ensure the macrospin regime for the absorption of microwave energy.

## RESULTS AND DISCUSSION

Figure 2 (a) shows a scanning electron microscope (SEM) micrograph of the perovskite compound  $\text{Sr}_2\text{Fe}_{0.9}\text{Mo}_{1.1}\text{O}_6$  ( $x = -0.1$ ). As seen from this figure, the compound is composed of quasi-spherical and ellipsoidal particles with flat surfaces at their percolation sites with neighboring particles and have a wide size distribution lying in the range from 100 nm to  $2\ \mu\text{m}$ . The morphology of other compounds with different compositions is similar to that for these two samples. Details of the particular geometry of some particles in the perovskite compound  $\text{Sr}_2\text{Fe}_{0.5}\text{Mo}_{1.5}\text{O}_6$  ( $x = -0.5$ ) is observed from the close view of the SEM micrograph shown as inset of Fig. 2 (a). Normalized hysteresis loops measured at room temperature with the field applied along the plane of the various perovskite compounds are shown in Fig. 2 (b). Although magnetic contributions of different origins may influence the magnetization reversal, the dominating contribution is of magnetostatic origin for all the perovskite compounds. Indeed, as observed from the inset in this figure, the decrease of the squareness or the initial susceptibility of the hysteresis loops is consistent with the decrease of the saturation magnetization as  $x$  is deviated from zero, as suggested by theoretical predictions [3]. That is, as  $x \neq 0$  other Fe (Mo) atoms occupy the sites of Mo (Fe) atoms, leading to changes in the interatomic distance between neighboring Fe atoms, so the larger the difference of  $x$  from zero, the more the non-stoichiometric the compound is. As a result, the ideal ferromagnetic arrangement of the stoichiometric perovskite is perturbed as a result of the appearance of local antiferromagnetic contributions, which in turn decrease the corresponding saturation magnetization and thus the magnetostatic energy.

In order to quantitatively account for the influence of Fe-Mo composition on the magnetic behavior of the different compounds, field swept FMR measurements have been performed at constant frequency in the range from 11 to 30 GHz. Fig. 3 (a) shows FMR spectra recorded at 20 GHz on the different compounds considered in this work,



**FIGURE 2.** (a) SEM micrograph of the double perovskite compound  $\text{Sr}_2\text{Fe}_{0.9}\text{Mo}_{1.1}\text{O}_6$  ( $x = -0.1$ ). The inset shows a close view of a SEM micrograph for the double perovskite compound  $\text{Sr}_2\text{Fe}_{0.5}\text{Mo}_{1.5}\text{O}_6$  ( $x = -0.5$ ). (b) Magnetization hysteresis loops recorded on the different perovskite compounds, where the inset displays the first quadrant of the same magnetization curves. (c) Schematics of the transversal section of a chain of quasi-spherical touching particles without lateral caps, where  $R$  is the particles radius and  $h$  the depth of the missing caps.

where differences between them are appreciable. Particularly, the FMR spectra absorb at different resonance fields ( $H_r$ ) which correspond to the field values at maximum absorption. As seen from the inset, the slight resonance field shift towards higher fields when  $x \neq 0$  further corroborates the observed changes in Fig. 2 (b). Recording absorption spectra at different excitation frequencies in the range 11 to 30 GHz allow to construct the frequency-field dispersion relation, as shown in Fig. 3 (b). Comparing this figure with Fig. 2 (b) we observe that the resonance field of about 4 kOe obtained at 11 GHz corresponds to a minimum magnetization state for all the samples that is about 80% of the saturated state. This means that the FMR experiments of this work have been carried out in the macrospin regime with a fair accuracy. The inset in this figure shows a close view of the extrapolated dispersion relations for the compounds with different  $x$  values. As observed, for  $x \neq 0$  in either case  $x < 0$  or  $x > 0$  the zero field resonance is shifted towards lower values, which means that the overall magnetic anisotropy is decreased as the crystal structure becomes non-stoichiometric. Since the particles embedded in the epoxy resin preserve the randomly oriented chain morphology, as shown in Fig. 2 (b), it was considered the very simple framework of the resonance condition for a chain of touching particles where the external field  $H_r$  is applied along its long axis, that is

$$f_r = \gamma(H_r + H_{eff}), \quad (1)$$

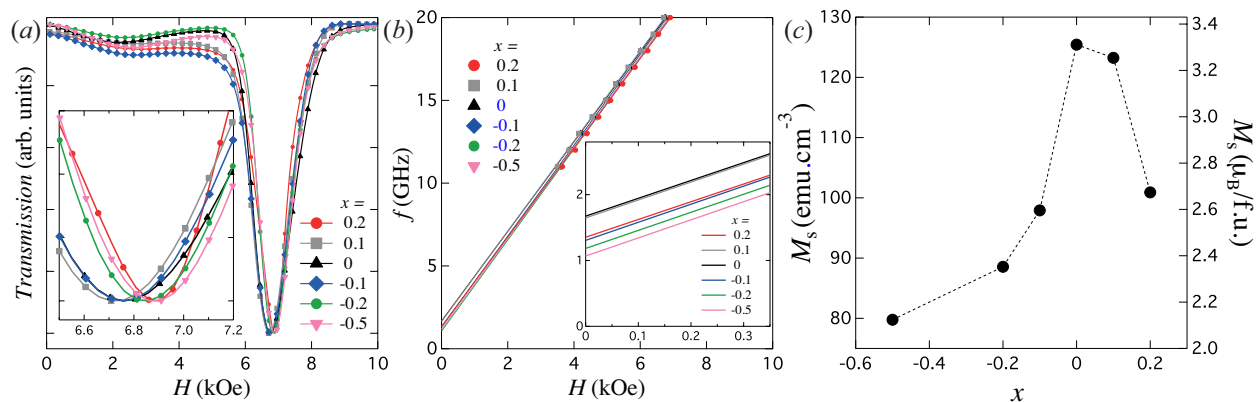
where  $\gamma$  is the gyromagnetic ratio of the double perovskite compounds spanning in the range 2.70 – 2.75 GHz/kOe and  $H_{eff} = 2\pi M_s P$  is the effective field of magnetostatic nature for a chain of touching single domain particles, obtained via a mean field approximation [10]. This magnetostatic field is obtained using the demagnetizing factors of the limiting cases of an isolated single particle and the completely filled particles container, so it accounts for the shape anisotropy contribution and the dipolar interaction between particles. Equation (1) gives the dependence between the resonance frequency ( $f_r$ ) and the resonance field ( $H_r$ ) and is used to fit the experimental data of Fig. 3 (b). From the linear fits the extrapolated zero field resonance frequency corresponds to  $f_0 = \gamma H_{eff}$ , which combined with the expression for  $H_{eff}$  allows determining  $M_s$  as

$$M_s = \frac{f_0}{2\pi\gamma P}. \quad (2)$$

Besides, a wide range of inter-particle distances are appreciated from Fig. 2 (a) which account for dipolar interaction contributions with different strengths. To a first approximation, let us consider the strongest of these interactions which arise from touching particles with flat surfaces at their percolation sites. According to the expression for  $H_{eff}$ , the dipolar interaction contribution is accounted by the packing fraction  $0 \leq P \leq 1$ , so that it yields a decrease of the shape contribution ( $2\pi M_s$ ) for an isolated infinite nanowire. The packing fraction of our system can be determined by considering the schematic representation of the transversal section of a chain of quasi-spherical touching particles without lateral caps, as shown in Fig. 2 (c). For this arrangement the packing fraction is

$$P = \frac{2 - f^2(3 - f)}{3(1 - f)}, \quad (3)$$

where  $f = h/R$  is the particles flatness factor given in terms of the radius ( $R$ ) of the particles and the depth ( $h$ ) of the missing caps, thus  $0 \leq f \leq 1$ . The limiting values of the packing fraction are then  $P = 2/3$  for percolated



**FIGURE 3.** (a) Absorption spectra recorded at an excitation frequency of 20 GHz for the non-stoichiometric double perovskite  $\text{Sr}_2\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$  with  $-0.5 \leq x \leq 0.2$ . (b) Frequency-field dispersion relations for the same compounds of (a). The insets in (a) and (b) display their corresponding close views. (c) Variation of the saturation magnetization ( $M_s$ ) as a function of  $x$ , given in  $\text{emu}/\text{cm}^3$  and  $\mu_B/\text{f.u.}$  units.

spherical particles and  $P = 1$  for completely touching flat discs which reproduce a solid isolated nanowire of radius  $R$ . In our double perovskite compounds the mean flatness factor is about 0.186, which along with the measured  $f_0$  values and Eqs. (2) and (3) allow determining approximate values of  $M_s$ . Figure 3 (c) shows the variation of the saturation magnetization with the Fe-Mo composition represented by  $x$ . As expected,  $M_s$  decreases from the maximum value obtained for  $x = 0$  which corroborates the presence of antiferromagnetic contributions which results in a decrease of the ferromagnetic order with respect to the perfectly ordered stoichiometric compound  $\text{Sr}_2\text{FeMoO}_6$ . The corresponding conversion from  $\text{emu}/\text{cm}^3$  to Bohr magnetons per f.u. is shown by the vertical right axis and has been determined by using previous reported values of the unit cell volume and density for each compound [4]. These results are in good agreement with theoretical predictions [3] and with other experimental values of  $M_s$  [11].

In summary, we have investigated by FMR and AGM experiments the influence of Fe-Mo composition on the magnetic properties of non-stoichiometric double perovskite compounds. We have provided a simple FMR approach based on a mean field approximation, along with geometrical considerations about the morphology of the particles, which allows determining with good accuracy the experimental saturation magnetization of the different compounds considered in this study.

## ACKNOWLEDGMENTS

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