



Room temperature ferromagnetism in Fe-doped BaSnO₃

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ABSTRACT

Polycrystalline bulk samples of BaSn_{1-x}Fe_xO₃, with $x = 0.03, 0.04$ and 0.05 were prepared by solid-state reaction method. These Fe-doped BaSnO₃ systems exhibit ferromagnetism at room temperature with an average magnetic moment of $0.047, 0.038$ and $0.025 \mu_B/F_c$ and Curie temperature of $510, 462$ and 446 K, respectively. The observed magnetic properties are not attributed to any of the known impurity phases or clusters. The presence of ferromagnetic interaction at room temperature is supported by the ferromagnetic resonance (FMR) signals observed in the electron spin resonance spectra. The ferromagnetic order between Fe-ions is expected to be mediated by an electron trapped at the oxygen anion vacancy, called the *F*-centre exchange (FCE) interaction.

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1. Introduction

The wise idea of utilizing the spin, another entity, of the electrons in addition to the charge used in the present electronics technology gave birth to the spin-based electronics, called spintronics [1]; also called magnetoelectronics [2]. The realization of useful spin-based electronic devices strongly depends on the availability of single-phase semiconducting materials, which are simultaneously ferromagnetic with high Curie temperature (T_C). It is also important that the ferromagnetism (FM) should be intrinsic and not arising from any impurity phase. A new class of semiconducting materials, like 3–5% Mn-doped InAs [3] GaAs [4], etc., called the diluted magnetic semiconductors (DMS) were developed for spintronic applications. Although these materials have been useful in realizing certain prototype spintronic devices [5], they work only at very low temperature [6]. The observed room temperature ferromagnetism in transition metal doped ZnO and in the Co-doped TiO₂ [7] has drawn the attention of the researchers towards oxide-diluted magnetic semiconductors (O-DMS). Following these, several other O-DMS based on SnO₂, Cu₂O and perovskite oxides of BaTiO₃, SrTiO₃, LaTiO₃, and KTaO₃ have been reported [8]. It is noteworthy that for the same Fe-concentrations, Fe-doped ZnO exhibits ferromagnetism at room temperature [9], while enriched ⁵⁷Fe-doped ZnO exhibits a paramagnetic Curie transition at 2.7 K

[10]. Although most of these materials exhibit FM with $T_C > 300$ K, in some cases the observed FM are not intrinsic. For example, oriented Co precipitates in Co-implanted ZnO and the Mn₃O₄ precipitates in the Mn-doped Cu₂O were found to be responsible for the observed FM [11].

BaSnO₃, an interesting wide band gap (3.1 eV) [12] oxide semiconductor, has been left unstudied for magnetoelectronics applications. BaSnO₃ has an ideal cubic perovskite structure belonging to the space group $Pm\bar{3}m$. Band structure calculations show that the BaSnO₃ is satisfying the conditions for a transparent conducting oxide and the charge carriers in the doped BaSnO₃ should have a high mobility [12,13]. Moreover, unusual near-infrared (NIR) luminescence was observed in BaSnO₃ semiconductor [14]. There are reports on transition metal substituted BaSnO₃ [15] and its electrical conductivity properties [16]. Yet, to our knowledge, there are no reports on the magnetic properties of the transition metal doped BaSnO₃. In this paper, we report magnetic properties of Fe-doped BaSnO₃ systems, which are ferromagnetic with high T_C . With reference to the ESR spectra, these systems are identified to be similar to Fe-doped SnO₂, which is an inhomogeneous ferromagnet with high T_C [17].

2. Experimental details

BaSn_{1-x}Fe_xO₃ with $x = 0, 0.03, 0.04$ and 0.05 samples were prepared using the starting materials (all high purity chemicals from Sigma-Aldrich), BaCO₃, SnO₂ and Fe₃O₄ by standard solid-state reaction method. Stoichiometric quantities of the starting materials were mixed well using agate mortar and pestle. These mixtures were then heated once at 1173 K for 12 h by taking them in alumina crucibles. Thereafter these mixtures were reground and heated again at 1173 K for 12 h; followed by heating at 1273 K, 1473 K for 24 h with intermediate grinding. These

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mixtures were further heated at 1573 K for 48 h, with intermediate grinding, to ensure the complete reaction. X-ray diffraction (XRD) measurements of the samples were done using (PANalytical X'Pert Pro) X-ray diffractometer with using Cu K α radiation. Magnetic properties were studied using vibrating sample magnetometer (ADE technologies, model EV7 VSM) of sensitivity below 0.5 $\mu\text{e.m.u.}$ and noise level < 1 $\mu\text{e.m.u.}$ Electron spin resonance studies were performed at X-band, 9.5 GHz (Varian E-112 ESR Spectrometer). Diphenyl-picrylhydrazyl (DPPH) was used as the reference for the calibration of g-factor and the corresponding resonance frequency (9.19 GHz in our case). In order to make better comparative analysis the spectra were recorded by keeping the instrument settings same for all the samples. Electrical resistivity measurements were performed using Keithley 617 Programmable Electrometer.

3. Results and discussion

3.1. X-ray diffraction pattern

Fig. 1(a) shows the powder XRD patterns, of undoped BaSnO₃, Fe 3%, 4% and 5% doped BaSnO₃ samples, with intensity in logarithmic scale. The diffraction peaks of all the samples were matched and indexed with reference to that of a standard XRD pattern of BaSnO₃ (Powder Diffraction File No. 15-0780). All the samples are found to be single-phase materials. Relative shift in the position of the diffraction peaks of all the Fe-doped samples with respect to that of undoped sample was observed as shown in Fig. 1(b). The

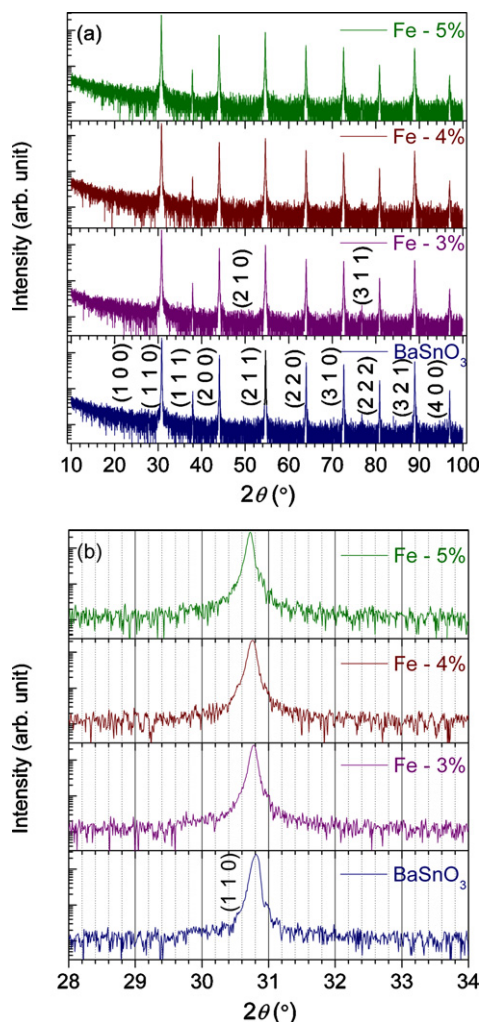


Fig. 1. (a) XRD patterns of BaSnO₃, Fe 3%, 4% and 5% doped BaSnO₃ samples, with intensity in logarithmic scale and (b) expanded view of (110) peaks of all the samples show relative shift in the position.

shift towards the lower angle indicates an increase in the lattice constant with increasing Fe content. This happens as the Sn⁴⁺ ions of ionic radius 0.69 Å are replaced by the Fe²⁺ ions of radius 0.78 Å, corresponding to the high spin (HS) ground state of Fe²⁺ with 3d⁶ electronic configurations. The Fe²⁺ with HS is chosen to be the ionic state of the Fe in our samples, since the ionic radii of all other ionic states of Fe including Fe²⁺ with low spin (LS) state are lesser than that of Sn⁴⁺. The calculated lattice constants of undoped and Fe 3%, 4%, 5% doped BaSnO₃ samples are 4.0867, 4.1047, 4.1073 and 4.1125 Å, respectively.

3.2. Magnetization

Fig. 2 shows the field-dependent magnetization, $M(H)$ curves of Fe 3%, 4% and 5% doped BaSnO₃ samples measured at 300 K; the observed maximum value of magnetization (M_{max}) of these samples, for the maximum applied field of 2 T, are 0.047, 0.038 and 0.025 $\mu\text{B}/\text{Fe}$, respectively. The magnetization plot of undoped BaSnO₃ is shown in the lower inset of Fig. 2, which exhibits a clear diamagnetism. Whereas hysteretic behavior is observed in all Fe-doped samples, consistent with ferromagnetic nature (see upper inset of Fig. 2). A coercivity (H_c) of 8.03, 10.69, and 6.89 mT and a remanence ratio (M_r/M_{max}) of 7.86, 4.83, and 5.04 percentage were observed for the Fe 3%, 4% and 5% doped samples respectively; where M_r is the remanent magnetization. All these values are quite consistent with those values reported by Coey as typical to DMS systems [18]. The hysteretic curves show an unusual behavior, that each of the loops closes before the saturation occurs, in contrast to that of conventional ferromagnets. Temperature dependency of the magnetization, $M(T)$ of these Fe-doped BaSnO₃ samples are shown in Fig. 3. The T_c of each of these samples is found by drawing two tangents (see Fig. 3). This method has been previously used to determine the T_c of Cr-doped In₂O₃ [19] and Fe-doped SnO₂ thin films [17]. The sample with Fe, 3% exhibits a linear to concave upwards shaped $M(T)$ -curve below its $T_c = 510$ K. This is also an unusual behavior in contrast to that of conventional ferromagnets, which generally show convex upward shape. Whereas Fe 4% and 5% doped samples behave like the conventional ferromagnetic samples; but not a typical one. Their T_c is 462 and 446 K, respectively. The unusual magnetic properties of Fe-doped BaSnO₃ are similar to that of Co- and Mn-implanted BaTiO₃, SrTiO₃ and KTaO₃

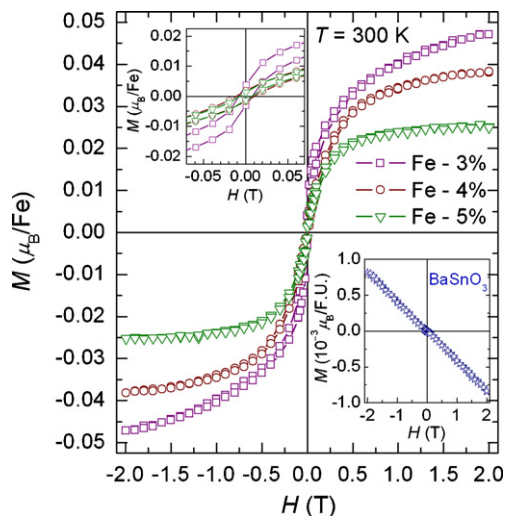


Fig. 2. Field-dependent magnetization of Fe 3%, 4% and 5% doped BaSnO₃ samples. Lower inset: field-dependent magnetization of undoped BaSnO₃. Upper inset: expanded view of field-dependent magnetization of Fe 3%, 4% and 5% doped BaSnO₃ samples.

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