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Effect of antimony doping on magnetic properties of Ni–Zn ferrites

M. PURNANANDAM, T. BHIMASANKARAM, S. V. SURYANARAYANA
Department of Physics, Osmania University, Hyderabad-500 007, India

The influence of Sb^{5+} doping on magnetic initial permeability (μ') and magnetic loss factor (μ'') in Ni–Zn ferrites over a frequency range from 10 kHz to 1 GHz at room temperature is studied. The Curie temperature (T_c), saturation magnetization (M_s), lattice constant (a) and mean grain diameter (D_m) of the pure and doped ferrites have also been evaluated. Domain wall relaxation has been observed in all the samples. Using the existing theories the magnetocrystalline anisotropy constant (K) and certain domain wall constants like wall damping parameters (β), domain wall energy (σ_w), the wall mobility (μ_∞) and the wall mass (m_w) have been evaluated and the results are compared and discussed with the similar data available on other ferrimagnetic oxides.

1. Introduction

Small amounts of additives to Ni–Zn ferrites which influence the grain growth, magnetic and electrical properties of the ferrites have been the subject of extensive research. Aliovalent impurities such as Ca^{2+}, Si^{4+}, Ti^{4+}, Ge^{4+}, Sn^{4+}, Nb^{5+} [1–6] in Ni–Zn ferrites have been found to affect the magnetic properties. From the studies on exchange interactions in antimony substituted nickel–zinc ferrite Blasse [7] concluded that Sb^{5+} ion has octahedral site preference compared to Ni^{2+} ions. Recently El-Nimr [8] supported the above observations while investigating Ni–Zn–Sb spinel ferrites. The aim of the present work is to study the influence of antimony on magnetic properties of $Ni_{0.35}Zn_{0.65}Sb_x\square_{0.02-x}Fe_{1.98}O_4$, where mole percent of x being 0, 0.5, 1.0, 1.5 and 2.0 are hereafter referred to as S_1, S_2, S_3, S_4 and S_5, respectively.

2. Experimental procedure

The ferrite samples used in this work were prepared by the double sintering technique [9]. The X-ray analysis of the samples confirmed the formation of spinel structure. The bulk densities (d_B) were measured by the Archimedes method, while X-ray densities (d_X) were computed using lattice constant values. The presence of antimony was confirmed by Auger spectroscopy.

The values of T_c of the ferrites were determined using thermal variation of relative susceptibility measurements [10]. The mean grain size was found from microstructures of the samples as suggested by Globus [11].

Saturation magnetization [M_s] was determined using soft magnetic measuring system of Walker Instruments Inc. (USA). The initial permeability (μ') and magnetic loss factor (μ'') were evaluated from inductance and Q measurements in the frequency range from 10 kHz to 10 MHz with HP-4275A impedance analyser and in the frequency range 1 MHz to 1 GHz with HP-4191A impedance analyser.

3. Results and discussion

The measured T_c and M_s values of the ferrite specimens are given in (Table I) along with the calculated values of the Bohr magneton number n_B. The lattice parameters a, T_c, and M_s values of basic ferrite (S_1) are in good agreement with those published earlier [12] taking into consideration that in the present investigation the undoped sample is iron deficient. The value of M_s in the antimony-doped samples is less compared to pure ferrite and decreases with increasing amount of the dopant. This may be due to the fact that the participating A–B interaction is weak resulting in the reduction of magnetization. The same trend is observed in the T_c, n_B and D_m values. The doped samples show a change in the lattice parameters, an increase from S_1 to S_4 and then a decrease in S_5.

It is observed that the initial permeability decreases with increasing antimony content as shown in (Fig. 1) and (Table I). This variation is the same as that of M_s with the dopant content. The initial permeability has been corrected to density and grain size [11]. The corrected initial permeability $(\mu - 1)'_c/D_m$ shows a rise in the 0.5% antimony-doped sample and then a gradual fall with further increase of antimony. The magnetic quality factor μ_Q also shows a similar variation. Das [2] observed a similar change in Si^{4+}-doped Ni–Zn ferrites. In the present case 0.5 mol % antimony doping has increased both μ_Q and the corrected initial permeability values showing that the pores might have been filled by the dopant even if there was a reduction in the grain size suggesting an offset by the small reduction in the permeability. The decrease in

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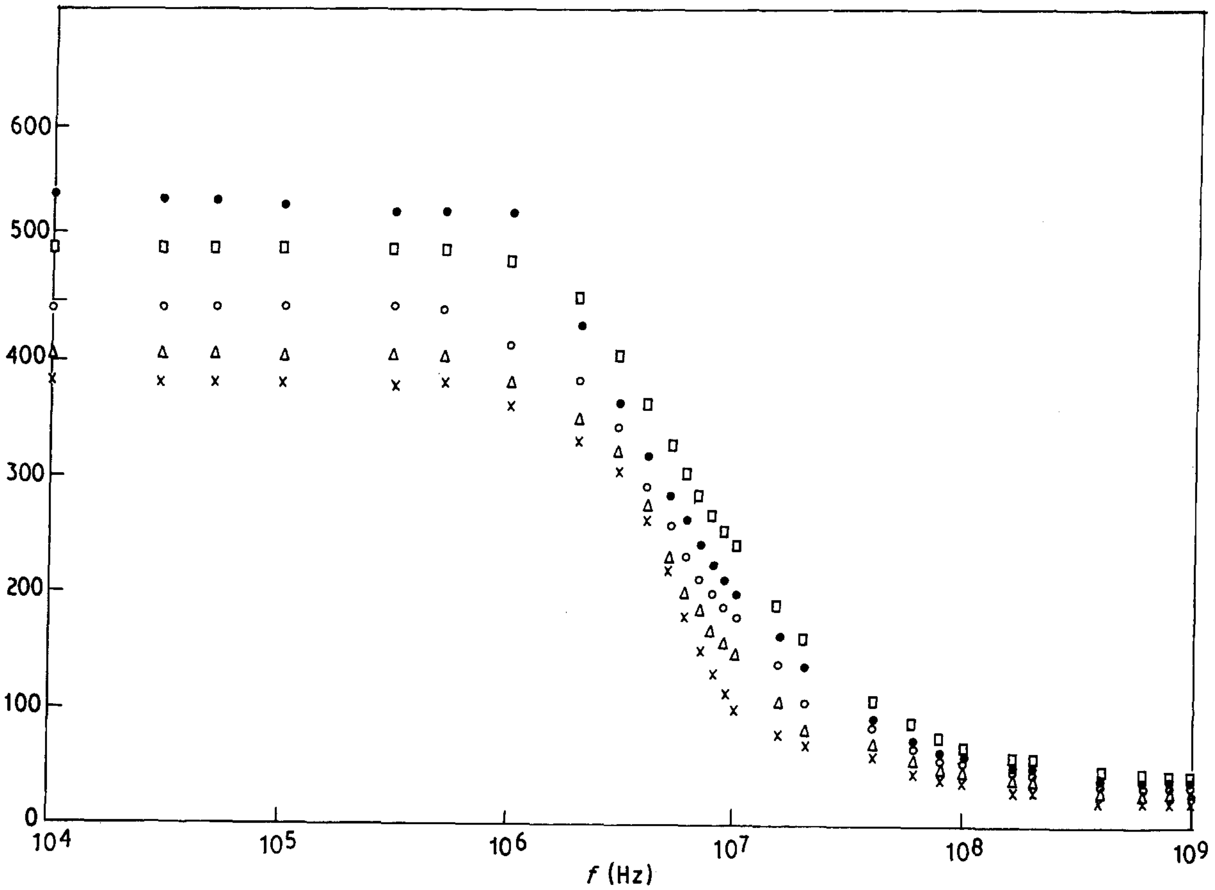

Figure 1 Variation of $(\mu - 1)'_c$ with frequency $Ni_{0.35}Zn_{0.65}Sb_x\,\square_{0.02-x}\,Fe_{1.98}O_4$. (● $x = 0$, □ $x = 0.05$, ○ $x = 0.010$, △ $x = 0.015$, × $x = 0.020$)

TABLE I

Sample	X	a (nm)	T_c (K)	M_s Gauss/cm^3	n_B	$\frac{(\mu - 1)'_c}{D_m}$	$\frac{\lvert K\rvert}{(\mathrm{erg\,cm^{-3}})}$	μ_Q
S_1	0.000	0.8390	393	287.5	2.49	146	12 984	38 425
S_2	0.005	0.8393	389	281.7	2.44	180	11 871	52 625
S_3	0.010	0.8396	382	267.8	2.38	170	11 554	36 200
S_4	0.015	0.8399	376	256.8	2.21	162	11 199	28 910
S_5	0.020	0.8395	371	247.1	2.09	157	10 961	19 175

magnetic properties of this composition due to diamagnetic antimony in the octahedral site is less pronounced compared to the rise in Q factor. Further addition of antimony reduces the magnetic parameters significantly.

Magnetic domain wall relaxations have been observed in all ferrimagnetic materials where the domain size is smaller than the grain size [11]. As the size of the grain becomes smaller the number of domains and their wall movement would be smaller ultimately the material will be of single domain size.

The total permeability of a magnetic material comprises domain wall permeability and spin permeability. With a reduction in the grain size, the total permeability decreases until the limit of spin permeability. A single domain grain specimen has spin permeability only. Globus [11] in his studies on Ni–Zn ferrites mentioned in general, the wall contribution to the permeability is dependent on the grain size of the sample. He also has shown in μ–f curves of a specimen of different grain sizes the wall permeability merges with the spin permeability at a high frequency of about 0.5 to 1 GHz. In the present study it is observed that all samples exhibit the domain wall relaxation (Fig. 1) which is shown by permeability dispersion. From μ'_c values at high frequency, $(\mu - 1)'_{c\,\mathrm{Rot}}$, the corrected rotational permeability component has been evaluated. Using the formula developed by Globus [11] the global magnetocrystalline anisotropy constant, $|K|$ of all the samples has been evaluated.

$$|K| = \frac{2\pi M_s^2}{(\mu - 1)'_{c\,\mathrm{Rot}}}$$

The $|K|$ value of undoped sample is nearly the same as reported by Globus [13] while that of antimony-doped samples gradually decreases with the increase in antimony content.

In a magnetic material, the translation of the spin direction from one Weiss domain to another is a

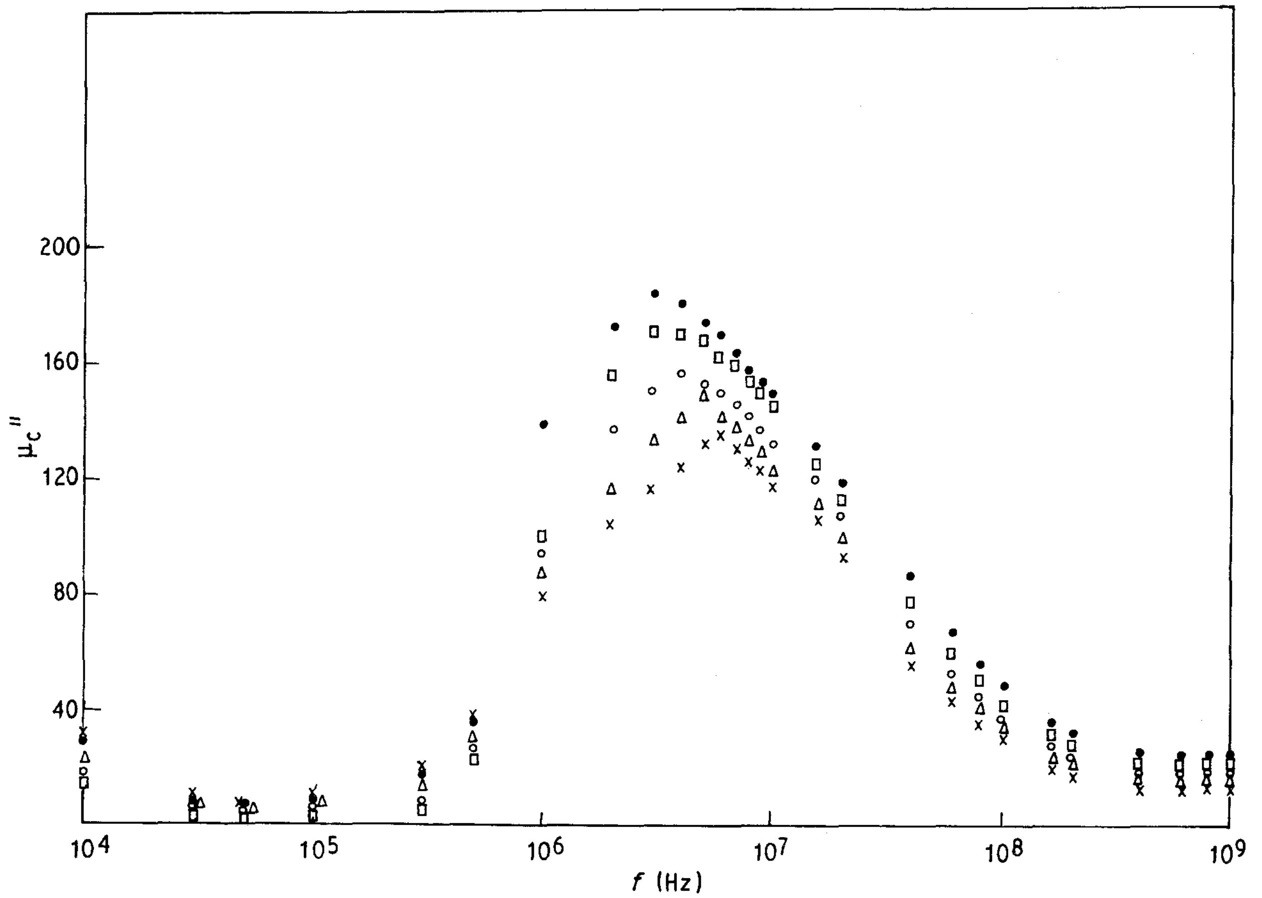

Figure 2 Variation of μ_c'' with frequency for $Ni_{0.35}Zn_{0.65}Sb_x\,\square_{0.02-x}Fe_{1.98}O_4$. (● $x = 0$, □ $x = 0.005$, ○ $x = 0.010$, △ $x = 0.015$, × $x = 0.020$)

TABLE II

Sample	σ_w (erg cm^{-2})	β (c.g.s.)	μ_∞ (cm s^{-1} Oe^{-1})	m_w ($\times 10^{-11}$ g cm^{-2})
S_1	0.130	1.61	714	5.00
S_2	0.123	2.46	458	4.85
S_3	0.120	2.26	474	4.82
S_4	0.117	1.91	538	4.79
S_5	0.115	1.69	585	4.76

gradual rotation of the spin vectors called Bloch wall [12]. The total energy (σ_w) required for the wall to exist is due to exchange energy between spins (σ_{ex}) and crystal energy component (σ_k). An estimate of the value of the total domain wall energy is made using the following expression [12]. $\sigma_w = (2|K|kT_c)/a$ where k is Boltzmann's constant. The values are given in Table II and are in the same order of magnitude as those reported in other ferrimagnetic materials [2, 14].

The doping of ferrite with antimony has reduced the magnetocrystalline anisotropy constant and also the Curie temperature, consequently the domain wall energy gets reduced with increasing antimony doping.

Döring [15] treated the domain wall as a vibrating membrane experiencing a force due to the variable magnetic field. As the applied frequency is varied the domain wall relaxes at a frequency where the imaginary part of the permeability shows a maximum (Fig. 2). In the present case it is observed that the peak value frequency (f_0) shifts to higher values with antimony content confirming that antimony occupies the octahedral site in the spinel lattice.

The domain wall damping factor

$$\beta = \frac{(16\sigma_w)}{(\pi f_0 D_m^2)}$$

and the domain wall mobility

$$\mu_\infty = \frac{4M_s}{\beta}$$

[16] are evaluated and given in Table II. The order of magnitude of β and μ_∞ of undoped Ni–Zn ferrite is comparable with the values reported by Merceron *et al.*, [17]. Sample S_2 has the highest domain wall damping factor which is also the least doped sample. Table II also shows the values of the domain wall mass per unit area (m_w) which are calculated using the expression [12]

$$m_w = \left[2\pi\gamma_e^2 \frac{\sigma_w}{|K|}\left(1 + \frac{|K|}{2\pi M_s^2}\right)\right]^{-1}$$

where γ_e is the gyromagnetic ratio for the electron (1.76×10^7 rad s^{-1} Oe^{-1}). This value decreases gradually with antimony content in Ni–Zn ferrite and is comparable with m_w of other ferrimagnetic materials [18–20].

4. Conclusions

The present work shows that in the case of antimony-doped Ni–Zn ferrites both saturation magnetization value and domain wall mass per unit area value show a decrease with increasing antimony content. The initial permeability decreases with antimony content in the samples under study. The magnetic quality factor (μ_Q) shows an increase in the Ni–Zn ferrite with 0.5% antimony and a decrease with higher impurity content.

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