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Microwave absorption in Ni–Zn ferrites through the Curie transition

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Abstract

Microwave absorption measurements were carried out on $Ni_{0.35}Zn_{0.65}Fe_2O_4$ polycrystalline ferrites, at a constant frequency of 9.4 GHz (X-Band) and dc magnetic fields (H_{dc}) in the 0–0.8 T range. The measuring temperature was varied from 300 to 500 K. A clear evolution from ferromagnetic resonance (FMR) to paramagnetic resonance (EPR) was observed as a function of temperature which is related with the passage through the Curie point (~430 K), as observed by thermal variations of magnetic permeability. In addition, a low field ($H_{dc} < 0.06$ T) absorption signal was observed with the same phase as the FMR absorption. However, in contrast with FMR, this signal exhibited hysteresis by cycling the dc field and disappears with temperature (~350 K) before the Curie point is reached. The assignment of this signal as due to non-resonant microwave absorption processes by the partially magnetized sample is discussed. © 2003 Elsevier B.V. All rights reserved.

Keywords: Magnetic measurements; Ferromagnetic resonance; Ferrites

1. Introduction

Polycrystalline ferrites are widely used in high-frequency devices, because of their high permeability in the radio frequency region, high electrical resistance, mechanical hardness and chemical stability.

Ferromagnetic resonance (FMR) spectroscopy has provided important information concerning the homogeneity of ferrite samples [1] and the dynamics of the ferrimagnetic transition. The peak-to-peak linewidths (ΔH_{pp}) are usually large and decrease just below the ferromagnetic transition temperature (T_c).

In this work we have investigated the Curie transition with FMR and have followed the evolution of resonance field (H_{res}) and linewidth (ΔH_{pp}) as function of increasing temperature. Additionally we investigated the temperature dependence of a low field absorption signal (LFS).

2. Experimental procedure

 $Ni_{0.35}Zn_{0.65}Fe_2O_4$ polycrystalline ferrites were prepared by coprecipitation from the nitrates followed by a 1000 °C 8 h thermal treatment [1]. FMR measurements were made using a Jeol JES-RES3X spectrometer operating at 9.4 GHz (X-band). The power of the ac signal was 1 mW. The FMR curves were obtained by the dc magnetic field modulation technique, with a modulation frequency of 100 kHz. The temperature of the sample was controlled by flowing cold or hot N₂ gas through a double walled quartz tube, which through the center of the microwave cavity. A Jeol ES-ZCS2 zero-cross sweep unit compensates digitally for any remanence in the electromagnet, thus allowing measurements to

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be carried out by cycling the dc magnetic field about its zero value, continuously from -0.2 to 0.2 T, with a standard deviation of the measured field of less than 2×10^{-5} T.

The Curie temperature was also measured by an independent technique based on the thermal variations of magnetic permeability [2].

3. Results and discussion

The FMR spectra of nickel zinc ferrites obtained at various temperatures from 300 to 500 K are shown in Fig. 1. It is observed a shift in resonant field and lineshape when temperature increases. The inset in Fig. 1 shows a low field signal (LFS) that is clearly temperature dependent and has the same phase as the main FM resonance.

The temperature dependence of the resonant field is plotted in Fig. 2(a) and the temperature dependence of the total linewidth ΔH_{pp} , measured from the left maximum to the rightmost minimum, are shown in Fig. 2(b). In this work we focus on the linewidth and the zero field absorption analysis and interpretation.

 $\Delta H_{\rm pp}$ decreases about 200% in a quasi-linear fashion when temperature is increased, from room temperature 300 to 412 K; this temperature is 18 K below the $T_{\rm c} = 430$ K, as already reported [3] and measured here by the thermal variations of initial magnetic permeability technique [2] shown in Fig. 3.

The decrease in ΔH_{pp} as temperature increases and the lost long range ferrimagnetic order are explained as due to the weakening of the magneto-crystalline anisotropy as *T* approaches T_c [4].

In the $(T_c - 18) \text{ K} < T < T_c$ range, short-range order fluctuations of both sublattices uncouple sets of magnetic moments that were exchange–coupled at lower temperatures; only dipole–dipole interactions remain. As *T* ap-



Fig. 1. FMR spectra of nickel zinc ferrite in the temperature range 300–471 K. Note the zero field absorption.



Fig. 2. (a) Temperature dependence of resonant field and (b) temperature dependence of linewidth.



Fig. 3. Curie point measured with the thermal variation of magnetic permeability technique.



Fig. 4. Temperature dependence of low field signal (LFS).

proaches T_c , the number of regions that become uncoupled increases, leading to an increase in the number of dipole–dipole interactions. These dipole–dipole interactions have the effect of increasing the linewidth until T_c is reached.

Above T_c the long range magnetic order is, of course, completely lost except for some short-range order islands in the material that contribute heavily to the dipole-dipole broadened line. As temperature is increased further into the paramagnetic regime, these short-range order islands rapidly decrease in number and size, and so does the width of the line, until a relatively narrow, symmetric line ($\Gamma = 0.0204$ T) is obtained at a temperature of 471 K.

The LFS is presented at $300 \le T \le T_c - 18$ K. Its intensity decreases as *T* increases, disappearing completely at *T* = $T_c = -18$ K when the short-range order fluctuations set in (as shown in Fig. 4). It shows hysteresis and it does not obey the FMR resonance condition. LFS seems to follow the thermal behavior of magnetization.

We believe LFS is a non-resonant microwave absorption closely related to the low-field magnetization, $\vec{M}(\vec{r}, T)$, of the sample in the long range order regime. Similar results have been observed in thin films (albeit no hysteresis was detected due to lack of sweeping through zero field), and attributed to spin rotation process [5].

4. Conclusions

Measurements of microwave absorption in polycrystalline ferrites of $Ni_{0.35}Zn_{0.65}Fe_2O_4$ exhibited the evolution of the resonant absorption from FMR to EPR as temperature went through Curie point. Additionally, a microwave absorption was observed at low fields, which can be associated with non-resonant spin rotation in the partially saturated sample.

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