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Research articles

Glassy behavior of diluted Cu-Zn ferrites

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ABSTRACT

The magnetic behavior of Zn substituted Cu-Zn spinel ferrites having chemical formula $Cu_{1-x}Zn_xFe_2O_4$ (x = 0.7, 0.8, 0.9 and 1.0) has been studied by SQUID magnetometry, by means of magnetic hysteresis, field-cooled (FC) and zero-field-cooled (ZFC) magnetization, memory effect and low field ac susceptibility measurements. These measurements suggest that the ferrimagnetic phase of the $x \leq 0.8$ samples is gradually turned into a spin glass ($x \geq 0.9$). The compound with x = 0.9 exhibits the typical dynamical behavior of spin glasses, with indication of aging, rejuvenation and memory effects. The evolution of the magnetic properties of Cu-Zn spinel ferrites with substitution of Zn for Cu is discussed.

1. Introduction

Ferrites having the combined properties of magnetic and dielectric materials have been intensively used in many electronic and electrical devices. Their structural and magnetic properties strongly depend on cation distribution among tetrahedral (A) and octahedral (B) sites and relative strength of various superexchange interaction among the magnetic moments on A and B sites [1]. When the superexchange interaction between A-B (J_{AB}) is stronger than B-B (J_{BB}) and A-A (J_{AA}), it leads a collinear spin structure but when J_{AA} or J_{BB} becomes comparable with J_{AB}, it leads a non-collinear spin structure [2]. When the ferrites are sufficiently doped with non-magnetic ions, they show a wide spectrum of magnetic structures such as ferrimagnetic order, local spin canting (LSC), anti-ferromagnetism, reentrant spin glass and spin glass system [3]. Spin glasses (SG) display complex magnetic order as the result of the coexistence of ferromagnetic (FM) and anti-ferromagnetic (AFM) interactions yielding magnetic frustration (randomness). In reentrant spin glass (RSG) systems, a transition first occurs from a paramagnetic (PM) to a ferromagnetic (FM) or anti-ferromagnetic (AFM) state. At lower temperature, this ordered state is turned into a (re-entrant) spin glass phase [4]. These types of transition show irreversibility indicating by the difference between field-cooled (FC) and zero-field-cooled (ZFC) magnetizations which is one of the characteristic features of a spin glass and other disordered systems [5,6].

Copper ferrite ($CuFe_2O_4$) is an inverse ferrite in which 6–24% of Cu^{2+}ions occupy the A-site and is ferrimagnetic at room temperature with Curie temperature T_c of 738 K [7]. The exchange coupling between Cu^{2+} on A-site and Fe^{3+} on B-site is weak which may result in spin canting. As Zn is doped in $CuFe_2O_4$, Tc decreases monotonously with the Zn content and $T_c \sim 450$ K for x = 0.5 [7]. $ZnFe_2O_4$ possesses a normal spinel structure which is reported to be antiferromagnetic below a Neel temperature of ~10 K and paramagnetic at room temperature. Zn^{2+} cations prefer to occupy the A-site of the structure, while Fe^{3+} cations occupy the B-site, and the negative superexchange interaction (J_{BB}) dominates the magnetic properties of $ZnFe_2O_4$ [1]. Significant geometric frustration in the magnetic interaction has been reported [8], as well as spin glass behavior [9]. In the present article, we have investigated the evolution of the magnetic properties of the $Cu_{1-x}Zn_xFe_2O_4$ (x = 0.7, 0.8, 0.9 and 1.0) ferrites upon substitution of Cu for Zn.

2. Materials and methods

Cu-Zn ferrite samples having chemical formula $Cu_{1-x}Zn_xFe_2O_4$ (x = 0.7, 0.8, 0.9 and 1.0) were synthesized in polycrystalline form by conventional ceramic method. The powder oxides of CuO, ZnO, and Fe_2O_3 were mixed in stoichiometric ratio and then grounded thoroughly for 6 h by using agate mortar and pestle. The mixture was ball milled for 4 h and then pelletized and the pellets were

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pre-sintered at 850 °C for 4 h to form ferrite through chemical reaction. The pre-sintered pellets were grounded again and passed through the same process of final sintering at 1050 °C for 2 h. Sintered samples were cut into small pieces with weight 5–8 mg for magnetization measurements. All measurements have been done by a superconducting quantum interference device (SQUID) magnetometer from Quantum Design Inc (MPMS). The samples were cooled from 300 K to 5 K in absence of dc magnetic field in ZFC mode and in the presence of magnetic field for FC mode. The data for magnetic field dependence of magnetizations (M vs H) at 5 K up to 2 Tesla were recorded. The ac susceptibility at frequencies of 1.7, 17 and 170 Hz with an ac field of 2 Oe was measured both in phase and out of phase of susceptibility.

3. Results and discussion

3.1. Structural analysis

Typical XRD pattern of the sample x = 0.9 of $Cu_{1-x}Zn_xFe_2O_4$ ferrite was done by X-ray diffraction using Philips X'part PRO X-ray diffractometer and illustrated in Fig. 1. The XRD pattern of the sample, showing well-defined reflection without any ambiguity, exhibits the formation of single phase cubic spinel structure. No other crystalline phase was observed indicating good stability of the ferrite compositions. The line profile shown in Fig. was fitted for 7 peaks (220), (311), (222), (400), (422), (511) and (440). The existence of (311) peak confirms the formation of cubic spinel structure. Details structural and microstructural features of $Cu_{1-x}Zn_xFe_2O_4$ ferrite has been explained in our earlier publication by Shahida et al. [10].

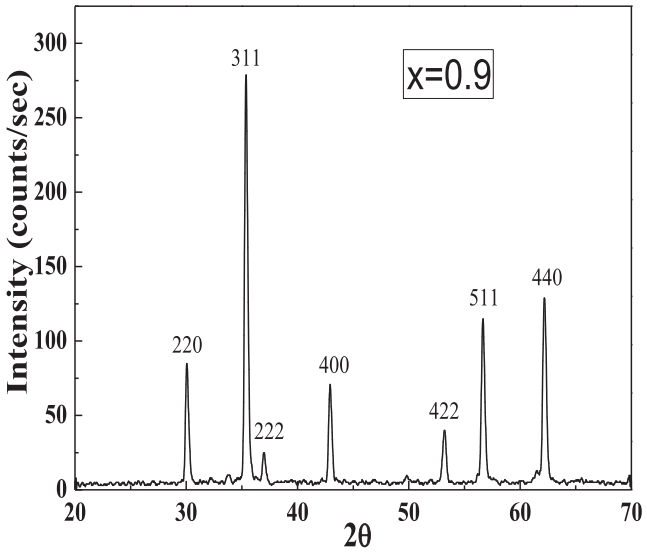

Fig. 1. XRD pattern of sample, x = 0.9 of Cu-Zn ferrites.

3.2. Field dependent dc magnetization

Fig. 2 (a) represents the magnetic hysteresis loops of samples x = 0.7, 0.8, 0.9 and 1.0 at 5 K up to field 20 kOe. The hysteresis curves of the compounds with x = 0.7 and 0.8 are typical for ferrimagnets, with a significant remanent magnetization. The magnetization vs field curves of ferrimagnets inherently have a finite positive slope above the apparent saturation; frustrated spins contribute as well to the increase of the magnetization at larger fields. The value of magnetization sharply increases for all samples with the external magnetic field strength at low field region. It cannot reach a saturation state with strong magnetic field of even 20 kOe. The magnetic hysteresis loop for x = 0.8 in the low field region is shown in Fig. 2(b). The coercivity (H_c) of samples with x = 0.7, 0.8, 0.9 and 1.0 are 200 Oe, 650 Oe, 500 Oe and 390 Oe, respectively. The magnetization decreases with increasing Zn content and the ferromagnetic-like hysteresis curves of the low-doped compounds are replaced by 'S-shape' ones for the larger doping levels. Such an evolution has been observed by Satter et al. in Cu-Zn ferrite [11] and by Serthol et al. in Ni-Zn ferrite [12].

3.3. Temperature dependence dc magnetization

Fig. 3(a, b) shows the temperature dependence of magnetization in the ZFC and FC mode for samples x = 0.7 and x = 0.8. The main observations are noticeable from Fig. 3(a): upon decreasing temperature from 400 K, the ZFC magnetization increases very sharply to a high value of magnetization exhibiting paramagnetic to ferrimagnetic transition at the Curie temperature $T_c \approx 272$ K. At lower temperatures, the ZFC curve shows a distinct plateau in the temperature range from 268 K to 57 K. Below that a sharp fall of the magnetization is observed. As seen in Fig. 3(b), a similar behavior is observed for the compound with x = 0.8; the temperature dependent magnetization data suggests a ferrimagnetic-like phase transition temperature at about 140 K. Below 60 K the ZFC curve decreases sharply with further decrease of temperature.

ZFC and FC magnetization curves in applied fields of 50 Oe for samples x = 0.9 were measured in temperature range of 5–150 K and are depicted in Fig. 4(a). ZFC and FC magnetization curves exhibit a cusp near 25 K. Some magnetic irreversibility is observed above the cusp temperature. As shown in Fig. 4(b), this irreversibil-

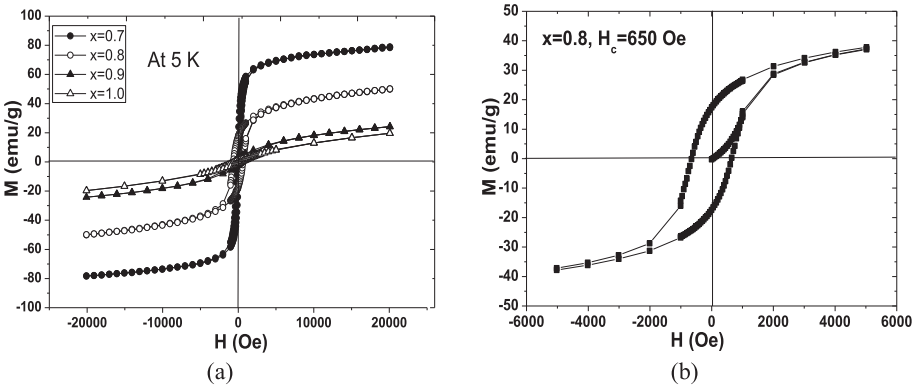

Fig. 2. (a) M–H hysteresis curves (x = 0.7, 0.8, 0.9 and 1.0) and (b) showing coercivity of sample x = 0.8.