TEMPERATURE-INDUCED METAMAGNETISM IN MgMn FERRITE

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Abstract

The metamagnetic transition (MT) from AFM to the FM state by applying a magnetic field was studied in the Fe-deficient MgMn ferrite. It is presented that MgMn ferrites will show MT, if the sample is in temperature variations under applied field below the critical value of field strength H_t. The critical transition field H_t = 580 A/m was estimated for MT from the antiferromagnetic (AFM) to ferrimagnetic (FM) state from the plot of M_r versus temperature T. It is assumed, that the MT can be induced by changes of the anisotropy. In the studied magnetic system the AFM state changes to FM state under the influence of magnetic field at adequate transition temperature T_t. One class of the MgMn ferrites that can become attractive are the metamagnetic systems.

1. Introduction

Metamagnetism belongs to the family of ferro- and antiferromagnetism, where long-range collinear order exists among spontaneous moments constrained to either parallel or antiparallel to an easy axis. In a metamagnetic system the antiferromagnetic state changes to ferromagnetic one under the influence of magnetic field. Magnetic instability can, however, be initiated with temperature due to a change in internal molecular field that affects the metamagnetic d-electron subsystems. Below its magnetic ordering temperature, a typical metamagnet is an antiferromagnet, but at increasing the applied field strength it can ultimately overcome the crystal anisotropy forces and thus to change abruptly the internal magnetic structure. The resulting field-induced magnetic transition from a state of low magnetization to one of relatively high magnetization is called metamagnetism [1-3]. Let suppose that the applied field has a value H_i inside an antiferromagnet and is parallel to the anisotropy (K) axis. As H_i increases with $T < T_N$ (Neel temperature), a critical value is reached where the force on an unfavourably aligned moments exceeds the constraint from the crystal-field anisotropy. The consequences can be divided into two kinds, depending on whether anisotropy is strong or weak (large or small K). For large K, an unfavourably aligned moment spontaneously reverses its direction at the critical field; its spin flips to become parallel to the direction of the applied field. For small K, however, with H_i along the easy axis, a moment gains magnetic energy by orienting perpendicularly to H_i, rather than parallel or antiparallel to it. Above the critical field, the unfavourably aligned moments therefore tend to rotate away from the K axis, to get closer to the perpendicular orientation. This is called a spin flop. At higher temperatures, the transition is to a saturated paramagnetic state in both cases. Metamagnetic behaviour for the examples has many variations. In particular, in some systems with large K the ferrimagnetic phase is absent and the transition at lower temperatures is directly from antiferromagnetism to saturated paramagnetism. But the variations of the moments in the antiferro-, ferri- and paramagnetic phases are constrained by the crystal field to lie along the easy axis K.

The metamagnetic transitions from AFM to the FM state by applying the magnetic field was discovered in the Fe-deficient MgMn ferrite and early published in [4-6]. In present paper we discuss the results of the investigation of metamagnetic effects of $Mg_{0.347}Mn_{0.894}Fe_{1.758}O_{3.88}$

ferrite with rectangular hysteresis loop under influence of magnetic field and temperature. One important class of MnMg ferrites due to their change of ground-states under magnetization can become very attractive in the metamagnetic systems.

2. Experimental Section

Non-stoichiometric Mn–Mg spinel ferrites were fabricated by a conventional ceramic processing method. The mixtures of Fe_2O_3, MnO and MgO, were sintered at 1200°C. The studied $Mg_{0.347}Mn_{0.894}Fe_{1.758}O_{3.88}$ ferrite samples have rectangular hysteresis loop and relatively low coercivity. Samples had toroidal shape. Inner and outer diameters of ring samples are 3.9 mm and within interval <6.15 – 6.10> mm, respectively. The ratio of inner to outer diameter of the cores was from interval <0.634 – 0.639>. The measured samples here are relative thick walled toroids. In this case the applied field was not effectively uniform over the toroid and measured values of remanent magnetic flux density B_r, coercivity H_C and the hysteresis loops obtained did not closely represent the actual properties of the ferrite. The magnetisation characteristics have been measured by means of conventional experimental set-up. Magnetic properties at low frequencies were evaluated from the set of minor loops measured by means of computer-controlled hysteresis graph built-up from commercially available measuring instruments. We followed the loss of coercivity associated with the phase transition by measuring the width of hysteresis loop (at zero magnetization) as a function of field at different temperatures for different values of applied field as a parameter. The experimental set-up allows usage of analogue (hardware) as well as digital (software) feedback to control the waveform shape of either exciting field $H(t)$ or flux density $B(t)$.

3. Results and Discussion

The observation of different temperature variation of the B_r versus temperature T at which the ac (f = 50 Hz) magnetic field H_a is made. The measuring procedure was magnetizing from room temperature (~22°C) to a high one up to the Curie temperature T_C = 297°C and then from high temperature to low one. At cooling the sample, the magnetizatiom at high field H_a = 580 A/m increases down to -193°C, see Fig.1. The induction increases upon cooling the sample to B_r = 0.257 T at -193°C and it continues increasing slightly up to the lowest temperature. The absence of the transition at this high field in the data also suggests that there may be a metamagnetic transition with critical field laying roughly at 580 A/m. Below this critical field the evolution of the B_r as function of T is fairly sensitive to field strength and to the temperature at which the field is applied, see Fig. 1.

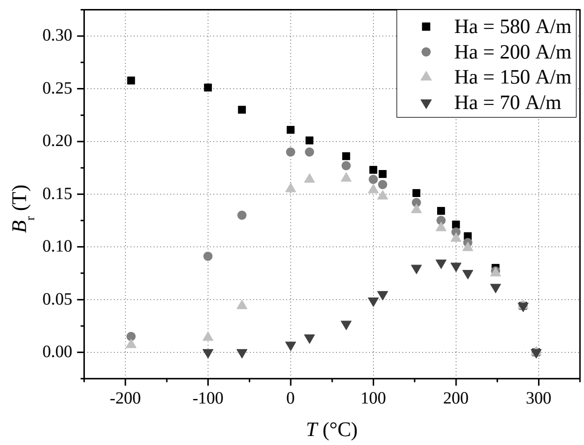

Fig. 1: *The remanent magnetic flux density B_r as function of temperature T for applied fields H_a = 580, 200, 150 and 70 A/m.*

As an example, at cooling the sample from T_C, the B_r at lower applied field H_a = 200 A/m increases up to 15°C and then the B_r decreases significantly down to -198°C, where the curve reaches zero value. On the other hand, on cooling the sample from T_C, the B_r at lower applied field H_a = 150 A/m increases up to 45°C and then the induction decreases significantly down to -100°C and the zero value reaches at 115°C. In another case, at cooling the sample from T_C, the M_r at applied field H_a = 70 A/m increases up to 183°C and then induction decreases significantly down to zero value roughly at -5°C.

In other experiment the coercivity values were measured upon cooling in fields of H_a = 580, 150, 100 and 80 A/m from T_C down to -193°C. At cooling the sample at applied field H_a = 580 A/m, the coercive field increases slightly down to 150°C, before increasing rapidly to H_C = 250 A/m at -193°C, see Fig. 2. The H_C values are continually increasing up to the lowest temperature for the curve measured at high field H_a = 580 A/m. Below this field the evolution of the coercive field as function of temperature T is also sensitive to field strength and to the temperature at which the field is applied. As an example, at cooling the sample from T_C, the H_C at lower applied field H_a = 150 A/m increases up to -20°C and then the coercive field decreases significantly down to -218°C, where the H_C reaches zero value. On the other hand, at cooling the sample from T_C, the H_C at applied field H_a = 100 A/m increases up to 60°C and then H_C decreases significantly down to roughly -100°C and the zero value reaches. In another case, at cooling the sample from T_C, the H_C at applied fields H_a = 80 A/m increases up to 80°C and then the H_C decreases significantly down to zero value roughly at -85°C. As the cores used are not thin-walled, the magnetic field in this case is not uniform across the radial thickness of the sample. As a result, curves depend not only on magnetic properties of the material, but also on geometry of the toroid. It is not feasible to solve the flux reversal problem due to basic principles. It is rather necessary to make some simple assumption. This necessity arises at least in part from the fact that it is not possible to describe completely the magnetic properties of material, including the effect of material inhomogeneities. For example, a polycrystalline nature of the ferrites is very difficult to specify in detail. It is known, that the small imperfections play a vital role in the reversal process. Because of these complications it is necessary to neglect much of properties of the magnetic structure of material in order to approach the problem of flux reversal.

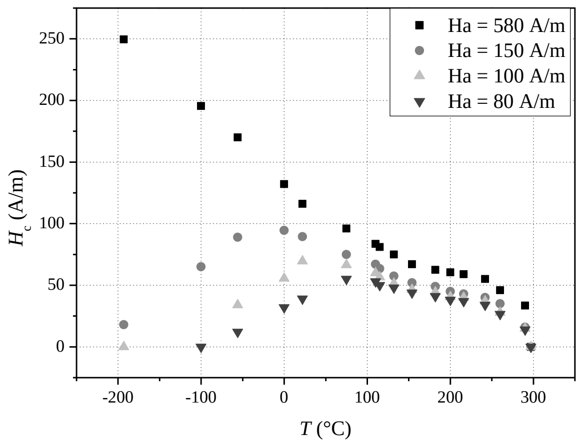

Fig. 2: *The coercivity H_C as function of temperature T for applied fields H_a = 580, 150, 100 and 80 A/m.*

In addition, it is necessary to postulate certain types of reversal process. In a system with multi-domain structure, it is possible to ascribe a reversal mechanism to three regions: domain-wall motion at low drives (~95.5 A/m), non-uniform rotation in intermediate drives (up to ~ 573 A/m), and uniform rotation at high drives (over 573 A/m) [7]. The curves of B_r and H_C as functions of temperature presented in Fig. 1-2 being measured up to external field H_a = 100 A/m, are probably determined by domain-wall motion reversal mechanism. The curves with transition critical field laying roughly at 580 A/m can be governed by uniform rotation mechanism. Then remaining curves of Fig. 1-2 can be corresponding to non-uniform rotation of magnetization.

4. Nature of the Magnetic Ground States

As an example of the nature of phase diagram it may be explained by the evolution of the remanent magnetic flux density B_r as function of temperature T measured at applied fields H_a = 150 A/m, as presented in Fig. 1. This selected phase diagram showing the different ground-states of $Mg_{0.347}Mn_{0.894}Fe_{1.758}O_{3.88}$ is depicted in Fig. 3. The magnetic field temperature phase diagram is found to be composed of three different magnetic phases. Below 0°C, the remnant magnetization decreases significantly down to -100°C and so does B_r, tending to zero value. Please note that magnetization below -100°C up to -273°C is collinear antiferromagnetism measured in H_a = 150 A/m. The data above -100°C up to T_C = 297°C are consistent probably with a canted-ferrimagnetic ground state at these temperatures in contrast to antiferromagnetic ground state. The transition at -100°C is then from a antiferromagnet to a canted- ferrimagnet. The third ground state above T_C is a paramagnet. There is principally one possible mechanism that may be regarded to offer this magnetic behavior. The non-uniform rotation mechanism can explain the observation of temperature dependence of the magnetization in the temperature interval <-100°C, 297°C>. Such a mechanism is, however, expected to produce results that should in principle, be dependent on nature of sample; shape and size of grains. The *T-H* magnetic phase diagram of $Mg_{0.347}Mn_{0.894}Fe_{1.758}O_{3.88}$ is summarized in Fig. 4. The phase boundaries shown in the magnetic phase diagram are determined from characteristic temperatures T_t = -100°C and 297°C. There K is small and one of spinel sublattices (probably B) can be canted below the critical external field at an adequate temperature.

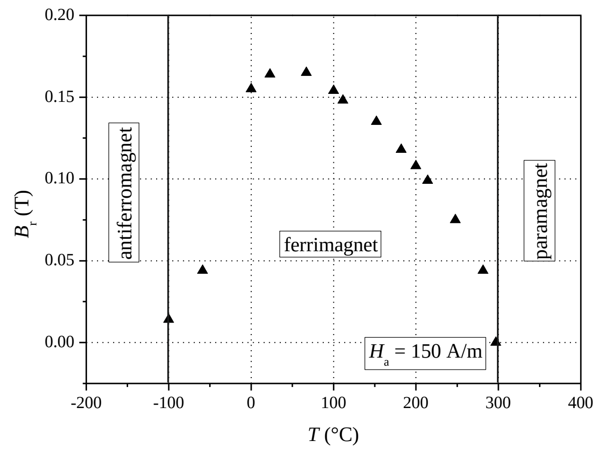

Fig. 3: *Phase diagram of $Mg_{0.347}Mn_{0.894}Fe_{1.758}O_{3.88}$ ferrite showing the three principial ground states for magnetization process at applied field H_a = 150 A/m.*

Upon the observation of different temperature variation of the magnetization depending on the temperature at which 330 A/m field is applied, the measurements of hysteresis loops (isothermal magnetization) were made. The rectangular hysteresis loops for field between 330 A/m and -330A/m are shown in Fig. 4, for selected temperatures. We note first an unexpectedly highest loop at 0°C, in spite of that widest loop is at -193 A/m, although the height of this loop tends to be lower with decreasing temperature. Below -193°C, the height of the loop will be zero approximately at -263°C and the loop decays.

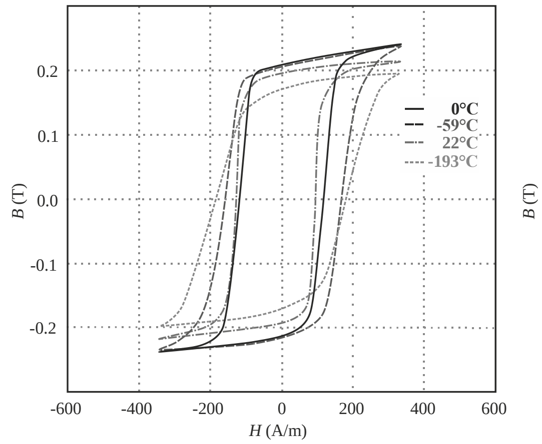

Fig. 4: *Hysteresis loops measured at applied field H_a = 330 A/m for selected temperatures.*

Below the critical field H_a = 580 A/m, the $Mg_{0.347}Mn_{0.894}Fe_{1.758}O_{3.88}$ ferrite under the influence of lower magnetic field is an antiferromagnet at a temperature range from -273°C up to adequate critical transition temperature (T_t); and exhibits a transition to canted ferrimagnet at T_t. The exact characterization of this transition had not been proposed. It is generally known that metamagnetism is induced by strong magnetic anisotropy in majority of cases. That is, however, not valid as for $Mg_{0.347}Mn_{0.894}Fe_{1.758}O_{3.88}$ ferrite. Contrarily, transition from ferrimagnetic to antiferromagnetic ground state may be related to the temperature dependence of the anisotropy field that increases on lowering the temperature. On top of it the small value of magnetostriction coefficient λ_{111} is included so as to minimize the effect of stress anisotropy. Mn-ferrites seem to be the most attractive ones, because of their extremely small value of λ_{111}. To improve the properties of $MnFe_2O_4$ - an effective combination is a MgMn mixture to compensate the magnetostriction coefficient down to zero and simultaneously a mixed ferrite enhances also the squareness of loop. The rectangular shape of loop was observed continuing up to transition temperatures.

4. Conclusion

This paper presents a brief review of existing works and original results on temperature-induced metamagnetism in substituted $Mg_{0.347}Mn_{0.894}Fe_{1.758}O_{3.88}$ ferrite. Below T_C, the evolution of the remanent flux density B_r and coercivity H_C as function of temperature is quite sensitive to field strength and to the history of magnetization of sample as well as the temperature at which the field is applied. Systematic investigations of the $Mg_{0.347}Mn_{0.894}Fe_{1.758}O_{3.88}$ ferrite made it possible to determine some specific features of the metamagnetic phenomenon and to formulate criteria to reveal such transitions in other spinel ferrites as well. We suggest that these observations are intrinsic to the chemical and structural nature of this compound. An example of MgMn ferrites that exhibit a sudden transition from the antiferromagnetic state to a highly magnetized ferrimagnetic state under influence of magnetic field is the compound $Mg_{0.347}Mn_{0.894}Fe_{1.758}O_{3.88}$, in which together with the particular magnetization the switching of magnetic flux from one to other stable state also makes a jump upwards in a process of target utilizing of ferrites for recording, pulse and digital applications.

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