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Study of the magnetic disaccommodation in La doped YIG

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The relaxation of the initial magnetic permeability of La doped yttrium iron garnet (YIG) samples with nominal composition $Y_{3-x}La_xFe_5O_{12}$ ($0< x <0.6$) is analysed in this work. The results show a very different behaviour depending on the sintering atmosphere used for the fabrication of the samples. Concretely, for the samples sintered in air, it just have been detected the usual relaxation peak found in YIG at 130 K. However, a new relaxation peak appears around room temperature for the samples sintered in CO_2 atmosphere when the La content is at least of 0.3. These results have been interpreted in terms of the formation of a secondary perovskite phase when the La solubility limit is reached.

1 Introduction

It is well known that Yttrium Iron Garnet (YIG) is a key material for the development of microwave devices due to its electrical and magnetic properties and the possibility of introducing a wide range of dopants for tailoring its properties. It is very usual the substitution of iron by metallic cations in tetrahedral or octahedral sites of the lattice [1–3]. However, the substitution of part of yttrium ions in dodecahedral sites with a rare earth element has been also studied so far [4, 5], showing that the total substitution of Y by lighter 4f rare earth elements La, Ce, Pr and Nd is not possible, and that, above the solubility limit, a secondary perovskite phase forms together with the magnetic garnet. In this sense, it is essential to determine the solubility limit of each extrinsic cation in order to assure that single YIG phase is obtained.

Recently, our group has shown [6] that the magnetic disaccommodation technique is a powerful tool to analyse the garnet-perovskite transition in rare earth element doped YIG. This technique consists in the time variation of the mobility of domain walls after a magnetic shock, and is shown by a temporal evolution of the magnetic permeability after a demagnetization stage. This kind of phenomena, which is strongly temperature-dependent, is undesirable from the technical point of view, as it represents an important source of magnetic losses in these materials, but it is very useful for basic research because such studies yield information about lattice symmetry and dynamics. Its origin has been attributed to either the rearrangement or the diffusion of anisotropic point defects, such as lattice vacancies and interstitials, within the Bloch walls. In this work we have studied the effect of La doping on the magnetic disaccommodation of YIG samples fabricated under different sintering atmospheres.

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2 Experimental

Fe_2O_3, Y_2O_3 and La_2O_3 powders of high purity (over 99%) have been mixed in the appropriate molar ratio in order to obtain samples with nominal compositions $Y_{3-x}La_xFe_5O_{12}$ ($0< x <0.6$). The mixtures were ground and pressed in a cylindrical die. The resulting green samples were sintered in air and CO_2 atmospheres at 1420 ºC for 5 hours (heating rate of 10 ºC/min), and then rapidly quenched to avoid phase annealing.

Magnetic disaccommodation (DA) measurements have been carried out with a computer aided system based on an automatic LCR bridge [7]. During the measuring process, the time variation of the reversible magnetic permeability is recorded in the temperature range 80 K < T < 400 K. Samples were previously demagnetized by using a decreasing ac magnetic field. The results are presented by means of the isochronal curves displaying the relative variation of permeability within different time windows:

$$DA(\%) = \frac{\mu(t_1,T) - \mu(t_2,T)}{\mu(t_1,T)} \times 100 \quad (1)$$

where t_1 = 2 s and t_2 = 4, 8, 16, 32, 64 and 128 s after demagnetization stage. When the time window $(t_2 - t_1)$ is of the same order of magnitude as the relaxation time at a specified temperature, this curve exhibits a maximum. In this way, the isochronal spectra disclose the different relaxation processes in the temperature range tested.

3 Results and discussion

The isochronal disaccommodation spectra obtained for the samples sintered in air atmosphere are shown in Fig. 1. It can be noted the presence of only one relaxation process at low temperatures. The maximum of this peak shifts to lower temperatures as the La content is increased, concretely from 130 K for the sample with x = 0.05 to 120 K for x = 0.6. This process is characteristic of undoped YIG [8] and is usually referred as process II. Its activation energy is around 0.32 eV and is associated with the electronic hopping between ferrous and ferric ions in the inequivalent octahedral sites of the lattice. The presence of these ferrous ions is due to the nonstoichiometry of the samples. Indeed, a slightly oxygen deficient structure appears when preparing high temperature sintered YIG [9]. These oxygen vacancies are compensated by a corresponding valency change of Fe^{3+} into Fe^{2+} ions, which are expected to occupy preferentially the wider octahedral sites. The detected shift of this peak to lower temperatures, and thus to lower activation energies, as the La doping increases must be interpreted in terms of the larger size of the unit cell caused by the introduction of a La ion in a lattice site otherwise occupied by a smaller Y^{3+} ion. In our case, the lattice parameter changes from 12.376 Å for pure YIG to 12.423 Å for the sample with x = 0.5, a similar value to that previously obtained [4].

Nevertheless, very important changes occur for the samples sintered in CO_2, as it can be observed in the isochronal spectra plotted in Fig. 2. Process II is detected again but now fades away when the amount of La doping is greater than x = 0.2. The peak maximum shifts to lower temperatures in the same way described above for the samples sintered in air. Moreover, the accommodation phenomena (i.e. negative disaccommodation) previously found in YIG samples sintered under reducing atmospheres [8] can be only lightly distinguished in this case. According to our theoretical model [8], these processes have their origin in a resonance phenomenon of the domain walls whose features depend on time due to the presence of a relaxation process of the induced anisotropy. Such processes are detected when the resonance frequency is similar to that utilized in the measuring system. Thus, we can conclude that the addition of La impedes the resonance mechanism of the domain walls or it takes place at frequencies higher than our measuring frequency (1 kHz), on the contrary to the case of YIG samples doped with tetravalent cations, as we have previously reported [2, 3].

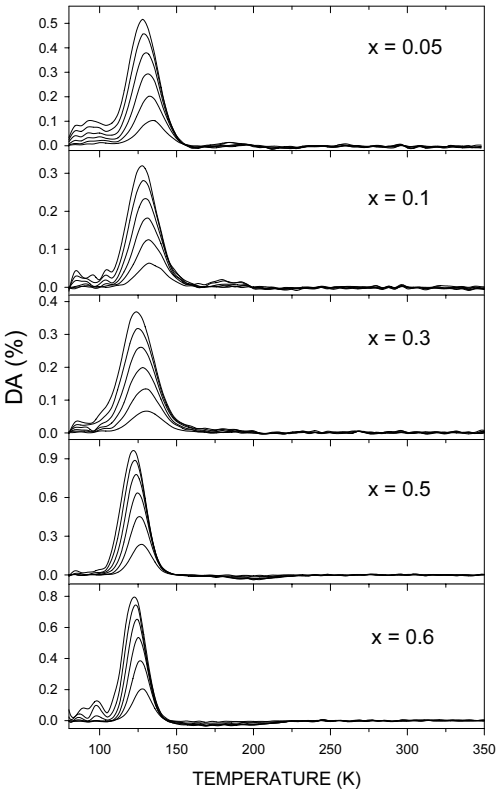

Fig. 1 Isochronal spectra of $Y_{3-x}La_xFe_5O_{12}$ samples sintered in air.

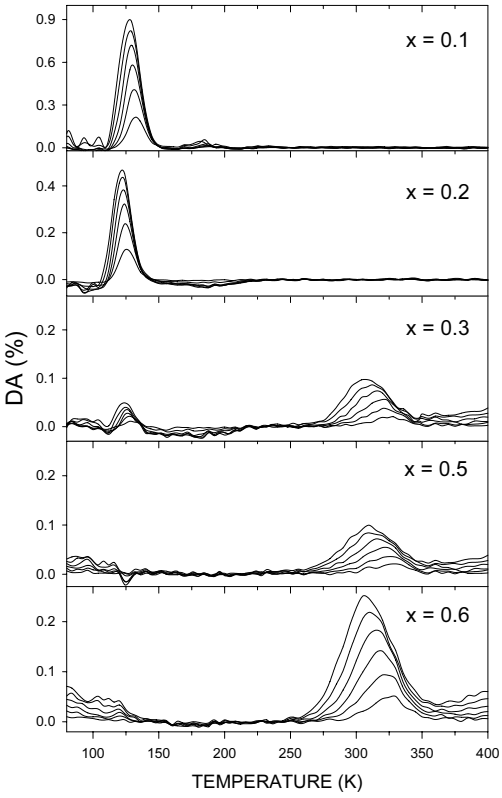

Fig. 2 Isochronal spectra of $Y_{3-x}La_xFe_5O_{12}$ samples sintered in CO_2.

However, the most significant changes take place in the isochronal spectra obtained for the sample with x = 0.3, where it can be noticed the appearance of a new peak around 300 K. For higher substitution rates, the low temperature garnet-related process vanishes and only the room temperature peak can be observed. This process is characteristic of the presence of magnetite in the samples, and is related to jumps of Fe^{2+} ions into crystal vacancies in octahedral positions of the lattice [10]. This result can be interpreted in terms of the phase formation: the Fe/La ratio is greater in the garnet lattice than in the perovskite phase and therefore, once the solubility limit has been reached, there is an excess of iron in the samples, involving the formation of magnetite due to the high-temperature sintering conditions. This assumption has been confirmed previously by using X-ray diffraction techniques [11] in an analogous system (Nd doped YIG) showing a similar behaviour in the DA spectra. Finally, process II disappears as the La content is increased due to the great amount of magnetite and perovskite formed. This conclusion is also supported by the thermal dependence of magnetic permeability shown in Fig. 3. It can be observed a progressive decrease up to x=0.3 and finally a strong reduction above this doping rate due to the formation of substantial amounts of antiferromagnetic perovskite. Therefore, the value obtained in this work for the solubility limit is lower than that previously reported [4] for a sintering atmosphere not as reducing as CO_2.

Hence, it can be stated that the solubility limit in this system decreases with reducing atmospheres, as our group has also previously shown in other systems such as Nd doped YIG [6, 11]. This result must be related to a higher amount of oxygen vacancies as the oxygen partial pressure is reduced [9], that may

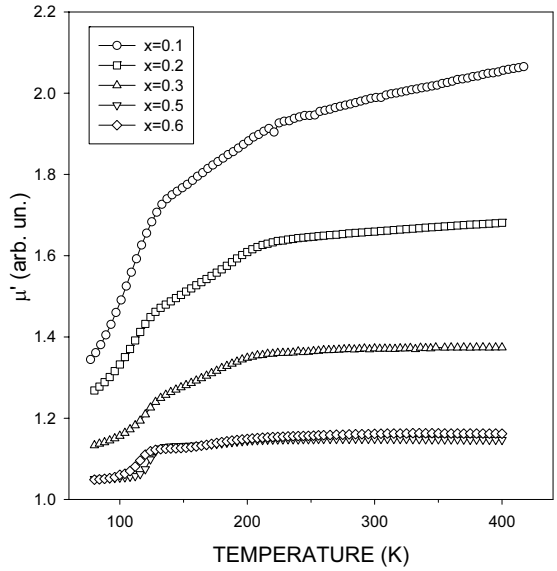

Fig. 3 Thermal evolution of the relative initial magnetic permeability measured 2 s after demagnetization stage for samples with nominal composition $Y_{3-x}La_xFe_5O_{12}$ sintered in CO_2.

facilitate the formation of secondary phases. In this sense, the La solubility limit in YIG should be higher if the oxygen partial pressure is increased, for which further research is required.

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