Magnetization processes in a dot of ferrimagnetic garnet near the compensation temperature

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Abstract. At the compensation temperature of ferrimagnetic materials, magnetization vanishes but domain wall energy does not. Magnetic domains and coercive field become then very large. We are interested in small objects of such materials, in order to study the effect of size, shape, edges... on the domain structure and on the magnetization processes. A magnetic garnet film was patterned by chemical etching, leading to dots 7 µm thick and 10 to 100 µm wide. Domain observations were performed close to compensation (213 K) under magnetic field up to 22 mT. In the case where the dots are connected by a thin continuous magnetic film (partial etching), magnetization reversal occurs via an intermediate spin-flop state, coexisting with perpendicular domains. In order to analyze this process, we performed a mean field computation to characterize the spin-flop state itself, by the amplitude and orientation of the magnetization of each sublattice.

1. Introduction
At the compensation temperature of ferrimagnetic materials, macroscopic magnetization vanishes but domain wall energy does not. Magnetic domains become then very large because dipolar energy vanishes, and also the coercive field because Zeeman energy vanishes. We are interested in observing small objects of such materials, in order to study the effect of size, shape, presence of edges, etc. on the domain structure itself, and on the magnetization processes. Temperature is then a parameter, which allows playing with the characteristic length of the system. We are studying the domain structure and hysteresis loops in a garnet film, using magneto-optical effect and image processing, to characterize the magnetization processes (nucleation, propagation, magnetization reversal).

The magnetic garnet sample, (GdTmPrBi)2(FeGa)O12, provided by CEA-LETI, is an epitaxial film grown on GGG substrate, about 7 µm thick. It has a strong uniaxial magnetic anisotropy perpendicular to the film, leading to up and down domains, separated by Bloch walls. At room temperature the usual maze domain structure is observed. This sample is ferrimagnetic and exhibits a compensation temperature of about 213 K, where the magnetization goes to zero. The resulting domain structure is characterized by very large domains (fig. 1).
We optimized the patterning procedures (lithography, chemical etching…) to obtain lattices of ferrimagnetic dots, squares and disks, between 10 and 100 μm wide and 7 μm thick (fig. 2). The first step of the patterning process is the deposition of a 100 nm-thick layer of SiO₂ on the garnet film, patterned by UV lithography and Reactive-Ion Etching. After lifting off the photo resist, the magnetic film itself is patterned by chemical etching with hot phosphoric acid [1]. Partial etchings were also performed, leaving a thin magnetic layer between the dots.

The present study concerns a partially etched sample, especially four dots of diameter 50 μm, on top of a continuous film 3 μm thick. The domain structure in this thinner films appears like a ‘background’ for the domain structure in the dots (fig. 3). Beside the domain size reduction due to the film thickness, the first observations at 300 K seem to show no interaction between the dots, even through the ‘background’, but the shape and size of the dots seem to affect the domain morphology.

2. Experiment

Our goal is to analyze the magnetization processes close to Tcomp. Observations are realized by means of Faraday effect, using an optical polarizing microscope, under magnetic fields perpendicular to the film up to 22 mT, at temperatures close to Tcomp (213 K).

An example of a field cycle is shown in figure 4. It is not the magnetization as function of the field which is displayed, but the grey levels measured in the images, corresponding to the background and to the dots. The field cycle in term of grey level is quite complex, in particular due to optical effects (not magnetic) resulting from the patterning, so that intensity changes in the background affect the dot, and reciprocally. The interpretation of the field cycle in term of magnetization can be described the following way:

- The initial state, in zero field, is negative dots, negative background (100% remanence).
- When the field increases, there is nucleation of a reverse domain in the background, propagation of this domain (avoiding the dots), leading to a positive background, while the dots are still negative.
Further increase of the field induces nucleation of a spin-flop state at the edge of the dots (ring), propagation in the dots, an intermediate state of coexistence between spin-flop and negative domain inside the dot, then collapse of this negative domain. The final observed state is then spin-flop in the dot, and positive background (fig. 5).

- Decreasing the field, the dots flip to positive domain state, so that in zero field the observed state is positive dots, positive background.
- Finally negative fields lead a similar evolution with a symmetrical situation.

The background is a continuous film, so nucleation of the reverse domain takes place on defects or edges, followed by propagation of the domain wall, leading to a low coercive field. In the dots, the nucleation probability is reduced, leading to a larger coercive field. The dots remain then negative in positive field, surrounded by a positive domain in the background, and with a domain wall in the edge of the dot. This situation is favorable for the nucleation of a spin-flop state when the temperature is close enough to Tcomp, which occurs here at about 20 mT.

The coexistence of negative domain and spin-flop within a dot is somehow analogous to a bubble domain opposite to the field, surrounded by domain in the field direction. Wall and Zeeman energies favor spin-flop. The energy barrier due to dipolar energy leads to a range of stability of such situation, before the collapse of the negative domain under a critical radius.

To go further in such description, it is necessary to better characterize the spin-flop state itself, the amplitude and orientation of the magnetization of each sublattice.

3. Model

The idea is a mean field approach, computing the magnetization of each sublattice as resulting from the local field (resulting from the magnetization of each sublattice).

The real formula for our sample is \((\text{GdTmPrBi})_3(\text{FeGa})_5\text{O}_{12}\) where a small amount of Bi was added to increase the magnetooptical effects, Pr to increase the anisotropy, and Tm to compensate for the lattice parameter mismatch with the substrate. We assume that concerning the magnetic properties, the magnetic elements in the rare-earth site can be described as being Gd, and the non magnetic as being Y, the resulting formula being \((\text{YGd})_3(\text{FeGa})_5\text{O}_{12}\).
The crystallographic cell (8 formula units) can be described as composed by 3 sublattices: Fe-octa, containing 16 Fe$^{3+}$, surrounded by octahedron of oxygen, (referred as Fe1), Fe-tetra, containing 24 Fe$^{3+}$, surrounded by tetrahedron of oxygen, (referred as Fe2), and Gd, containing 24 Gd$^{3+}$, surrounded by dodecahedron of oxygen.

For each sublattice, the magnetization is defined by its amplitude at T= 0, a unit vector $\vec{m}$, the thermal variation being described by a Brillouin function:

$$\vec{M}(T) = M(0)\vec{m}(z)B_s(z)$$

$$B_s(z) = \frac{2S + 1}{2S} \coth \left( \frac{2S + 1}{2S} z \right) - \frac{1}{2S} \coth \left( \frac{z}{2S} \right)$$

with $z = g\mu_B S H_{\text{eff}} / kT$

$$\vec{H}_{\text{eff}}^{Fe1} = \lambda_{11} \vec{M}_{Fe1} + \lambda_{12} \vec{M}_{Fe2} + \lambda_{13} \vec{M}_{Gd} + \vec{H}_0$$

$$\vec{H}_{\text{eff}}^{Fe2} = \lambda_{21} \vec{M}_{Fe1} + \lambda_{22} \vec{M}_{Fe2} + \lambda_{23} \vec{M}_{Gd} + \vec{H}_0$$

$$\vec{H}_{\text{eff}}^{Gd} = \lambda_{31} \vec{M}_{Fe1} + \lambda_{32} \vec{M}_{Fe2} + \lambda_{33} \vec{M}_{Gd} + \vec{H}_0$$

$\vec{H}_{\text{eff}}^n$ is the effective field on each sublattice. The exchange coupling constant $\lambda_{ij}$ are taken from literature, based on study of $(Y)_3(FeGa)_{5}O_{12}$ [2] and $(Gd)_3(Fe)_{5}O_{12}$ [3]. $\vec{H}_0$ is the applied field.

Qualitatively, the stronger coupling is (Fe-tetra)-(Fe-octa) and is negative. The next stronger is (Fe-octa)-(Fe-octa), then (Fe-tetra)-(Fe-tetra). The next one is Gd-(Fe-tetra), also negative, and finally Gd-(Fe-octa). The coupling Gd-Gd is negligible) [2-4].

The values for exchange constant we have taken are the following:

$\lambda_{11} = -721; \lambda_{12} = -1077; \lambda_{13} = -11; \lambda_{22} = -343; \lambda_{23} = -48; \lambda_{33} = -2$.

Adjusting the composition to fit Tcomp (213 K) and Tc (450 K), taking into account the fact that 90% of Ga atoms substitute Fe-tetra [5], lead to the following formula:

$$(Y_{1.4} Gd_{1.6})(Fe_{1.94}Ga_{0.06})(Fe_{2.96}Ga_{0.54})O_{12}$$

The computed thermal variation of spontaneous magnetization for each sublattice is presented in figure 6. As expected it exhibits a compensation temperature close to the experimental value. What is remarkable is that the amplitude of Gd magnetization is very temperature dependent.

![Figure 6. Computed spontaneous magnetizations (total and for each sublattice) as a function of T.](image)
Under a constant magnetic field the thermal variation of the magnetization exhibits a spin-flop transition in a temperature range of about 20 K (fig. 7). The comparison with experiment is quite reasonable (fig. 8), apart from the field range which is very different. This can possibly be adjusted playing finely with the composition and the coupling constant values.

**Figure 7.** Model: thermal variation of Mz under constant field, for each sublattice and the total magnetization: spin-flop between 203 and 223 K, under 10 T.

**Figure 8.** Experiment: thermal variation of Mz under constant field, for 10 and 30 mT. Spin-flop between 210 and 225 K under 30 mT. Far from Tcomp Mz increases, and 10 mT is a field too small to saturate the sample.

The main results from this computation is a description of the spin-flop state, as function of T, around Tcomp. The amplitude and orientation of the magnetization of each sublattice, as function of T, is shown in figure 9, and the angle between the sublattices in figure 10.

**Figure 9.** Rotation, with respect to the normal to the film, of the magnetization of each sublattice, as function of T, during spin-flop.

**Figure 10.** Angle between the two Fe sublattices (left), and between Gd and Fe-octa sublattices (right), as function of T, during spin-flop.
In the temperature range where spin-flop exists, the magnetization of Fe, in each sublattice, is not changing much, while the magnetization of Gd is decreasing a lot while rotating when T increases. Fe-tetra and Fe-octa remain almost antiparallel, and at the compensation they are almost lying in the plane. Gd and Fe-tetra, on the other hand, do not remain antiparallel. The maximum angle between Gd and Fe-tetra is 29°, and one can notice that this maximum deviation does not occur at Tcomp. This is due to the fact that the magnetization of Gd is in the direction of the applied field below Tcomp, but in the direction opposite to the field above Tcomp.

Gd sublattice is able to adjust its amplitude and orientation in order to increase Zeeman energy, which explain the existence of the spin-flop state. This state could occur in smaller field in a dot compared to the case of a continuous film, due to an easier nucleation mechanism.

4. Conclusion
Close to the compensation temperature, in this patterned system with “background”, magnetization reversal occurs in the dot via an intermediate “spin-flop” state, which coexists with the normal domain opposite to the field. A mean field model, related to a film of our sample, lead to a description of the spin-flop state (amplitude and orientation of the magnetization of each sublattice with respect to the film), giving some elements in order to analyze quantitatively the magnetization processes in the dot.

Magnetization of the Gd sublattice is the variable allowing the increase of Zeeman energy. In our model the anisotropy is not yet included, but this will be done and should induce hysteresis. There is still a problem with the field range for spin-flop, which is very different in the model (10 T) and in the experiment (30 mT). We hope this could be solved tuning the composition and the exchange constant values.

The next step of our study will be the description of the domain wall between the spin-flop state and the normal domain (structure, width, energy). It will then be possible to analyze the equilibrium between these two domains, and the collapse of the normal domain when its radius get smaller than a critical value, by analogy with the problem of a magnetic bubble in opposite field.

References
[1] Wet etching of garnet films. Tolksdorf W, Bartels I, Dammann H, Pross E 1987 J. Cryst. Growth, 84 323
[2] Molecular field coefficients of substituted yttrium iron garnets. Röschmann P, Hansen P 1981 J. Appl. Phys 52 6257
[3] Canted spin phase in Gadolinium Iron Garnet. Bernasconi J, Kuse D 1971 Phys. Rev. B 3 811
[4] Modern Magnetooptics and Magnetooptical Materials. Svezdin AK, Kotov VA 1997 Studies in Condensed Matter Physics, IOP Publishing Ltd
[5] Magnetic and magneto-optic properties of lead and Bi substituted YIG films. Hansen P, Witter, Tolksdorf W 1983 Phys. Rev. B, 27