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DEVICE APPLICATIONS OF GARNET THIN FILMS

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Résumé - Les 15 dernières années ont été très fructueuses pour le développement de dispositifs réalisés avec des films minces de grenats. Les principaux domaines d'application sont les mémoires à bulles magnétiques, les mémoires optiques, les dispositifs d'affichage, les dispositifs hyperfréquences à ondes magnétostatiques, mais également d'autres domaines tels que les composants optiques intégrés pour les télécommunications sur fibre optique. L'objectif de cette revue est d'examiner les développements récents des composants en relation avec les propriétés souhaitées pour les films de grenat : anisotropie magnétique, aimantation, rotation Faraday, largeur de raie de résonance ferromagnétique, etc... Les composants seront décrits et les problèmes techniques restant à résoudre seront discutés.

Abstract - The past 15 years have been very fruitful in the field of development of devices making use of garnet thin films. The major device areas are the magnetic bubble memories, the magneto-optical displays and printers, the optical memories, the magnetostatic surface wave filters, but, also, other devices such as integrated optics components are of current interest. The purpose of this review is to examine the recent developments of components in relation to the suitable properties of the garnet films : magnetic anisotropy, magnetization, Faraday rotation, FMR linewidth, etc... The components will be described, as well as technical problems still to be solved.

I - INTRODUCTION

The garnet system has been known since 1958 /1/ the best-known material is yttrium-iron garnet, Y₃Fe₅O₁₂, or YIG, which is used for microwave applications. Up to 1970, the garnet single crystals where obtained in the form of bulk materials used to make small spheres for microwave purpose. At that time, the enthusiasm for the large scale manufacture of magnetic bubble memories made it possible to develop thin garnet films from liquid phase epitaxy technique. This powerful technique allowed the fabrication of many garnet materials for new applications. To date, the major device areas are magnetic bubble memories, magneto-optical displays and printers, optical storage, microwave filters and integrated optics components.

The purpose of this paper is to review the recent developments of components in relation with the suitable properties of garnet films.

II - THE GARNET MATERIALS

The rare earth (R.E.) iron garnets have the following formula : \( \{ \text{R.E}_3 \text{Fe}_2 \text{Fe}_3 \} \text{O}_{12} \) where \{ , [ ], and ( ) stand for dodecahedral (c), octahedral (a) and tetrahedral (d) sites respectively /2/. The coupling between the sublattices is antiferromagnetic, as shown below for the "heavy" rare-earths."
The most important characteristic of the garnets is the possibility to adjust their composition and therefore their magnetic properties thanks to the substitutions of desired ions on sites (c), (a), or (d).

Table 1

<table>
<thead>
<tr>
<th>Site d</th>
<th>Site a</th>
<th>Site c</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe^{3+}</td>
<td>Fe^{3+}</td>
<td>V^{3+}</td>
</tr>
<tr>
<td>Si^{4+}</td>
<td>Ti^{4+}</td>
<td>Ca^{2+}</td>
</tr>
<tr>
<td>Ge^{4+}</td>
<td>Sn^{4+}</td>
<td>Pb^{2+}</td>
</tr>
<tr>
<td>Ga^{3+}</td>
<td>Zr^{4+}</td>
<td>Bi^{3+}</td>
</tr>
<tr>
<td>Al^{3+}</td>
<td>In^{3+}</td>
<td>Pr^{3+}</td>
</tr>
<tr>
<td>V^{5+}</td>
<td>Co^{2+}</td>
<td>Nd^{3+}</td>
</tr>
<tr>
<td>Sc^{3+}</td>
<td>Sb^{5+}</td>
<td>Tb^{3+}</td>
</tr>
<tr>
<td>Tb^{3+}</td>
<td>Sm^{3+}</td>
<td></td>
</tr>
</tbody>
</table>

The magnetization is changed by placing non-magnetic ions on the tetrahedral site: increasing the amount of Ga^{3+}, Al^{3+}, Ge^{4+}, or Si^{4+} ions will decrease the magnetization. Alternatively, increasing the amount of Sc^{3+} or In^{3+} on the octahedral sites will raise the magnetization. The incorporation of Al^{3+}, Ga^{3+}, Sc^{3+} is straightforward while the use of Si^{4+} or Ge^{4+} requires the simultaneous addition of a divalent cation like Ca^{2+} for charge compensation.

Ion substitutions are also used to adjust other magnetic properties (anisotropy field, coercivity, magnetostriction, etc... and to optimize optical properties (Faraday rotation and light absorption).

The material requirements are also met thanks to the versatility of the fabrication techniques: essentially all magnetic garnet films reported to date have been grown by the Liquid Phase Epitaxy (LPE) technique /3/.

This method is very powerful because it leads to high quality, very reproducible thin films. The LPE growth process is now well standardized among the various workers in the field. The LETI procedure which is fairly typical is as follows: the films are grown on [111] surfaces of gadolinium gallium garnet (GGG) polished substrates under isothermal conditions. A four-pronged platinum holder is used to support five wafers.

![Fig. 2 - Bubble memory cost](image-url)
The substrates are introduced into the furnace chamber and lowered to just above the solution surface for preheating. Baffles and reflectors are positioned above the platinum crucible to obtain the optimum temperature profile. A horizontal dipping technique at 60 rpm is used during growth. The composition suitable for the application is obtained from supersaturated solutions. The saturation temperature is in the range 750-950°C and the films are grown isothermally under a supercooling of ~25°C. The growth of a 1-micron thick film takes about 1 minute. The GGG substrate is now available commercially in the form of polished wafers as large as 10 cm in diameter and with a defect density lower than 1 per cm².

III - MATERIAL FOR HIGH DENSITY BUBBLE MEMORIES

Since the introduction of magnetic bubble memories (MBM) in 1967 /4/ rather remarkable developments have been achieved. Several companies (Intel, Motorola, Fujitsu, Sagem...) are selling one megabit bubble memory devices made from (YSm)(FeCaGe) garnet thin films supporting 2 µm-diameter bubbles. MBM systems offer several advantages over other kind of memories. These include the absence of moving parts, high reliability, non volatility of data, and easy maintenance. In bubble memories the information is stored in the form of cylindrical (bubble) domains whose magnetization is the reverse of that of surrounding area. Their presence ("1") or absence ("0") in specific places corresponds to binary digits stored at those locations /5/. MBM are still quite expensive, so, in order to decrease the price of devices to an attractive commercial level, say 3 mc per bit (fig. 2), industrial companies are fabricating 4 Mbits memory chips with garnet films supporting 1 micron bubbles /6/. Epitaxial growth on the (111) plane of the GGG substrate produces the so-called "growth induced anisotropy" necessary to stabilize the magnetic bubbles. This anisotropy is attributable to the mixing of large and small ions on the dodecahedra1 sites: Sm is the large ion commonly used while Tm and Lu serve as the small ions. Bubbles can be obtained only when:

$$H_k = 2K/uM_s > 4\pi M_s$$

where $K_u$ is the uniaxial anisotropy constant, $H_k$, the anisotropy field and $4\pi M_s$, the saturation magnetization. The bubble diameter is approximately given by

$$D = 9(AK_u)^{1/2}/\pi M_s^2$$

Submicron bubbles are well within the capabilities of the garnet systems. Table II gives examples of garnets which are already able to support bubbles down to 0.5 micron.

<table>
<thead>
<tr>
<th>Sample name</th>
<th>Strip width w(µm)</th>
<th>Film thickness h (µm)</th>
<th>Bubble collapse field $H_o$ (Oe)</th>
<th>Saturation induction $4\pi M_s$ (G)</th>
<th>Anisotropy field $H_k$ (Oe)</th>
<th>Curie temperature $T_c$ (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>1.0</td>
<td>1.1</td>
<td>450</td>
<td>870</td>
<td>2100</td>
<td>220</td>
</tr>
<tr>
<td>B</td>
<td>0.7</td>
<td>0.8</td>
<td>540</td>
<td>1050</td>
<td>2500</td>
<td>233</td>
</tr>
<tr>
<td>C</td>
<td>0.5</td>
<td>0.6</td>
<td>730</td>
<td>1400</td>
<td>2800</td>
<td>253</td>
</tr>
</tbody>
</table>

Table II - Submicron Bubble Garnets

The limit of present technology seems to be in the order of 32 Mbits/cm² where 0.35 micron bubbles are needed. This can be obtained by the use of (Sm, Lu)₃(Fe₃Sc)₉ garnets. Scandium ions occupy octahedral sites and would bring both a decrease in the exchange stiffness $A$ and an increase in magnetization $M_s$. To overcome the
fundamental limitation due to the concept of bubble domain memories one has recently proposed a new solid state magnetic memory named Bloch line memory /6/. The "1" and "0" are stored in the stripe domain walls (fig. 3) instead of using the well-known presence or absence of cylindrical "bubble" domains. This new technology is expected to facilitate the production of memories with capacities of the order of one gigabit on a single chip measuring about one square centimeter /7/.

![Bloch-line memory](image)

**Fig. 3 - Bloch-line memory**

**IV - GARNETS AS MATERIALS FOR OPTICAL DEVICES**

Magneto-optical effects in garnet crystals have been the subject of large interest since the mid-sixties. Possible applications include optical storage, displays, printers and fiber-optics telecommunications. Before any description of potential applications we shall present the essential features of the magneto-optical materials.

**IV-1 - Optimum composition of a magneto-optical garnet film**

The primary requirement is that there must be the greatest contrast between light beams that have passed through areas magnetically polarised in opposite directions (fig. 4). The difference $\Delta I$ in intensity is large if the ratio $Q = 2 \theta F/\alpha$ is large:

- $\theta F$ is the Faraday rotation per metre,
- $\alpha$ is the absorption coefficient per metre,
- $Q$ is the figure of merit of the material.

Many studies /8,9/ have shown that the optical contrast is greatly enhanced by substituting bismuth at dodecahedral sites in the lattice. The general formula of a garnet used for an optical application is

$$\text{RE}_{3-x} \text{Bi}_x \text{FeGa}_5 \text{O}_{12}$$

where the bismuth content is between $x = 0.5$ and $x = 1.5$. Gallium is used to adjust the magnetization. A large bismuth concentration changes magnetic properties like anisotropy field, magnetization, compensation temperature, and Curie point. The consequence is that the optimum composition depends on the application and can be relatively complex, as it is in the case of bubble memories.

**IV-2 - Light switching displays and arrays**

The switching cells shown on fig. 4 can be configured as a linear array that can be used for electrophotographic printing /10/. When arranged in a matrix the cells could form the basis for data displays /11/. The principle of recording is as follows: the garnet composition is chosen so as to obtain an anisotropy field $H_a$ which is high at room temperature and decreases rapidly when the material is heated (fig. 5). So, the direction of magnetic polarization can be reversed if an external magnetic field and a heating pulse are applied to the material at the same time. Called LISA for light switching array the device proposed by Philips Research Laboratories in Hamburg /11/ has very attractive features: typical switching time is 20 $\mu$s, up to 2000 optical patterns can be generated per second, and the resolution is typically 300 dots/inch. An A4 page can be exposed and printed within only 2 $\mu$s. The design of a switching cell is shown on fig. 6. The temperature is raised by an electronic current pulse applied to the resistor and the applied magnetic field is in the region of 0.03 Tesla. Besides printing this kind of device could lead to display application. A two-dimensional switching device of 256 x 256 pixels which operates in the same
way as the LISA array has been achieved by Philips researchers. A 16 K-pixels device was recently tested by Pulliam et al /12/ for use as a spatial light modulator in optical processing applications.

For these applications it could be necessary to grow garnet films with a figure of merit as large as 4 degree/db. This can be obtained with a \((\text{GdBi})_3(\text{FeGaAl})_{50}12\) garnet as shown by Ferrand et al /13/.

Optical disk storage units are generally classified in two types: read only and rewritable types (fig. 7). Read only optical disk storage corresponds to digital audio disk and archival media on which records are not erasable. On the other hand, in the case of rewritable disks it is possible to write, read, erase, and write again data, using a laser beam. Several read-only-material systems are already available in the market place. Until recently, many people have argued that erasable optical storage was neither needed or desired. However, recent developments in both magneto-optical /14/ and phase change /15/ materials have resulted in renewed enthusiasm. This interest can be attributed to:

1. The technology base (including GaAlAs semiconductor lasers) which were provided by read-only optical disk storage.
2. The development of amorphous materials with large magneto-optic Kerr rotation leading to greatly improved signal to noise ratio, in the order of 50 db in a 30 KHz bandwidth.

The principle of recording is similar to that used for displays and arrays. A weak magnetic field (~ 0.02 T) is produced and, at the same time, the temperature of the material is increased by the laser beam focused on the disk. As a result the coercive field of the magnetic layer decreases and the direction of magnetization is reversed. The diameter of the area to which the laser beam is applied can be reduced to about 1 micron. Hence, the recording density is ~4x10^7 bits/cm², that is an order of magnitude higher than those of magnetic storage. Read out is generally based on magneto-optical Kerr rotation since the light is reflected from the metallic storage. Some 40 000 A4 documents can be recorded on a 30 cm diameter disk.

The magnetic film is usually made from amorphous alloys. Examples are TbFeCo alloys which are already in development. Among the materials which have potentialities to be a good medium, the garnets are very attractive because they exhibit large magneto-optical rotation, and, in contrary to amorphous metals are chemically stable.

In order to obtain uniform magnetic thin layer on amorphous substrates larger than 5 inch, it is necessary to choose a suitable deposition technique. A drawback of LPE is a need for lattice-matched single crystalline substrates. Furthermore, GGG substrates are limited in size and very expensive (150 $ a 4 inch-wafer in 1985).

For these reasons other techniques have to be investigated. RF diode sputtering seems to be suitable since (GdBi)₃(FeAl)₅O₁₂ garnets have been obtained with properties similar to those of LPE films with the same compositions [16].

IV-4 - Integrated optical devices

It is known since the work of Tien et al [17] that LPE iron garnet films are suitable for optical waveguides between 1.1 μm and 4-5 μm wavelength. In this magneto-optical waveguide Faraday and Cotton-Mouton effects cause mode conversion between TE and TM guided modes. These properties can be used to build modulators as well as non reciprocal devices such as isolators and circulators which could be very useful for fiber-optic communication systems [18,19]. However, the efficiency of the mode conversion and therefore the performance of devices are strongly affected by the optical birefringence in the film. It was recently demonstrated that this birefringence is composed of a stress-induced part and of a growth-induced part [20]. It means that a high efficiency mode conversion remains a problem to be solved, specially in bismuth garnet films for which the origine of the growth induced anisotropy is not yet clearly understood.

V - Magnetostatic-wave devices for microwave systems

For years, engineers have been trying to find a planar technology to make microwave devices easier to be built than Yttrium Iron Garnet (YIG) spheres used as resonators...
in microwave systems. Since 1 to 50 micron-thick YIG films can be grown with low defect density, impressive good experimental results have been obtained in this field /21-25/. These devices depend on the propagation of slow, dispersive wave at microwave frequencies in a low loss ferrimagnetic material. Presently, YIG films grown on GGG substrates by the liquid-phase epitaxy technique are characterized by a ferrimagnetic resonance linewidth $\Delta H$ comparable to those of YIG spheres, say 0.5 oersted at 10 GHz. A basic configuration of a delay line based on forward volume waves propagation is shown on fig. 9.

![Diagram](image)

**Fig. 9 - Basic configuration of a magnetostatic wave delay /25/**

When a RF field is applied to the input transducer a local perturbation of the spins occurs, which then propagates through the YIG film via the coupling of adjacent magnetic moments. This wave reaches the output transducer some time later and induces a current in the microstrip. A typical result is shown on Table 3 /25/.

| Thickness of the GaYIG film : 18.7 microns |  
| Center frequency : 840 MHz | Table III  
| Bias field $H_0$ : 85 oersteds |  
| Delay : 150 ns |  
| Bandwidth : 120 MHz |  

Without changing the bias field $H_0$, the same devices can be used as dispersive delay lines which are attractive for pulse compression applications. Multielement grating transducers are used to make narrow band filters: bandwidth as narrow as 15 MHz were obtained for the S (2 to 4 GHz), C (4 to 8 GHz), X (8 to 12 GHz) and K (18 to 27 GHz) frequency bands. One-part and two-part tunable resonators can be fabricated using periodic etched-groove gratings as selective reflectors /22/.

**VI - CONCLUSION**

This rapid survey of the applications of garnet thin films shows that a large number of devices making use of these materials are still in development. Among them, the most promising are magneto-optical displays, optical memories and integrated optical devices. The magnetostatic wave devices makes them very competitive with other microwave device technology such as YIG spheres in filters and oscillators. The market of these components is just taking off. On the contrary, the production and demand for magnetic bubble memories cannot be said to be low. In addition to a constant rate of increase in demand for NC machine tools, robotics, military equipments, their use is rapidly increasing in POS terminals and portable computers. The worldwide market for MBM was approximately 200 millions dollars in 1984 and will become a 300 millions dollars market during 1987. In conclusion, one can say that garnet thin films are very good examples of new materials successfully developed to meet the needs of electronic industry and have opened the way to large markets in the field.
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