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Epitaxial Garnet Investigation; Technical Report, Foreign Travel

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Applied Optics Branch
Optical Sciences Division

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Report of European trip to investigate techniques for growing epitaxial garnets with high Faraday rotation and low optical and magnetostatic wave attenuation. Bismuth substituted YIG is indicated.

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CONTENTS

EPITAXIAL GARNET INVESTIGATION; TECHNICAL REPORT, FOREIGN TRAVEL

Three foreign laboratories were visited. Those laboratories, and the names of people with whom technical discussions were held, are listed here.

3-2-87 Crismatec Corporation, Grenoble, France
Jean Mareschal, Director General
Herve Le-Gal, Director of Production
Gilles Le Blevennec, Engineer: responsible for epitaxial materials production.

Also: Henri Le Gall from CNRS (French national scientific research center) at Meudon-Bellevue near Paris, France - a scientific advisor to Crismatec, also currently growing epitaxial garnets in his laboratory, present by request of Jean Mareschal.

3-4-87 Philips Research Laboratories, Hamburg, W. Germany
Dr. Peter Hansen
Dr. W. Tolksdorf - all research physicists who grow, characterize,
Dr. Damman or use epitaxial garnet films for Philips
Dr. Krumme
Dr. Doormann

3-6-87 Thomson - CSF Research Center, Orsay, France
Dr. J. P. Castera
Dr. P. L. Meunier all research physicists who grow, characterize,
Dr. J. Y. Beguin or use epitaxial garnet films for Thomson
CSF.
Dr. J. L. Rolland
Dr. P. Friez

The technical purpose for these visits was to ascertain the prospects for obtaining epitaxially grown ferrimagnetic garnets which incorporate bismuth to increase the magnitude of the Faraday rotation constant. Subsidiary materials issues were concurrently to retain low optical and magnetostatic wave absorption and the control, or reproducibility, of the value of the saturation magnetization and the optical and magnetic anisotropy.

To stimulate interest, at all three laboratories I explained the basic principles of the waveguide magnetostatic wave - optical diffraction Bragg cell being developed in our laboratory at NRL. Higher Faraday rotation in the garnet materials will lead to higher diffraction efficiency.

The Crismatic facility at Gieres, near Grenoble, is primarily a manufacturing facility. Boules of various materials, e.g. gadolinium gallium garnet, GGG, are grown by the Czochralski method. Pure yttrium iron garnet (YIG) films are grown on GGG substrates by liquid phase epitaxy (LPE) in production lots. In addition, one or two furnaces (of perhaps twenty) have been set aside for developing growth procedures for new materials. This work is being done by M. Le Blevennec.

The current experimental material is BiGdIG. A 7 μm thick film was grown on both sides of a GGG substrate of approximately 620 μm thickness. At an optical wavelength of 1.3 um the sample transmission was about 80%. Most of the loss is caused by surface reflection.

Manuscript approved August 17, 1988.

Optical resonances in the multiple layer sample were apparent in the spectrophotometer transmission curves. The optical absorption edge was for wavelengths shorter than about 1.3 µm. Faraday rotation values were intriguing - $\Phi_F = 1550^{\circ}/cm$ at 1.3 µm wavelength and $\Phi_F = 1000^{\circ}/cm$ at 1.5 µm - and optical absorption was figured to be about $\alpha = 2.5/cm$ at 1.3 µm, but the consensus opinion was that ferrimagnetic resonance (FMR) linewidth would be large, say $\Delta H > 5$ Oe, because of the inclusion of gadolinium.

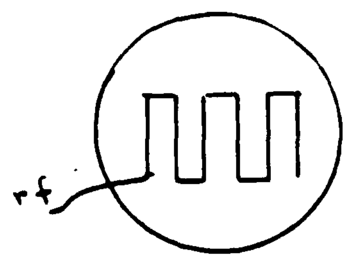
Crismatic also has a small facility at Paris, apparently, where some new materials development is taking place. M. Desvignes there has grown bismuth substituted YIG films with up to .4 formula units Bi on GGG substrates; and on a large lattice constant (LLC) modification of GGG incorporating calcium, magnesium, and zirconium according to a technique pioneered in Europe (by Philips, I think) films of composition $Bi_{1.4}Y_{1.6}IG$ have been grown 3 um thick. These films are a good theoretical lattice match to the substrate and have high Faraday rotation: $\Phi_F = -2700^{\circ}/cm$ at 1.15 um wavelength and $\Phi_F = 12500/cm$ at .6328 um. $4\pi M_s = 1750$ Oe and the ferrimagnetic resonance linewidth, although high, is not unreasonable for an early effort: $\Delta H = 4\text{-}5$ Oe at 9 GHz over the full wafer. The magnetic anisotropy is quite high however; $K_u = 65000\ erg/cm^3$. The details of this anisotropy were elucidated later on my trip by the scientists at Philips - see the discussion below.

Crismatic has a connection with another scientist in or near Grenoble who has equipment to do some optical characterization, M. Oliver at CEA-CENG-LETI/ CRM 85X-38041/Grenoble Cedex/France (Centre d'Energie Atomique - Centre d'Enginieure - Laboratoire d'Electronique Technologie Information is approximately the decoding of this acronymn).

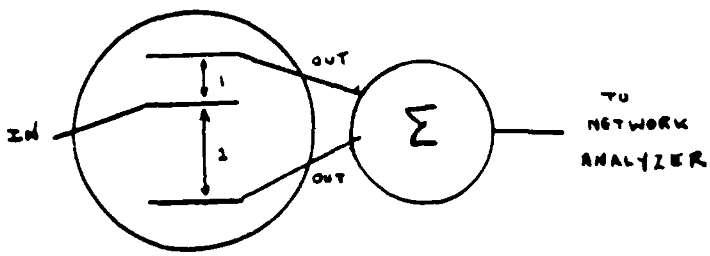
Henri Le Gall / CNRS /Place A. Briand / 92195 Meudon-Bellevue/France was also at Crismatec for the day. (CNRS = Centre Nationale de Research Scientifique). His Laboratory near Paris is experimenting with growing and characterizing bismuth substituted garnets, and he expressed interest in trying to grow the material we need on an experimental basis, perhaps with a follow-on contractual relationship through Crismatec. He may send me a sample, for assistance in characterization.

To pursue this contact: (1) Send Crismatec a copy of the specifications for the material as listed in the NRL RFQ. (2) Correspond with Herve Le-Gal at Crismatic and Henri LeGall at CNRS regarding the importance and apparent difficulty of controlling magnetic and optical anisotropy in bismuth substituted films. (3) Add Crismatec to the list of potential sources for the RFQ.

Other Notes: (1) We should characterize the 1" and 2" diameter films recently purchased from Crismatec, particular their ferrimagnetic linewidths, as soon as possible. The 2" film was suspected to have larger H. (2) Crismatec's ownership is changing, and it is anticipated that it will soon be part of a holding company having a US affiliate, Balkowsky Corporation of Charlotte, North Carolina, through which purchases can be made. This reorganization may affect the relationship of Crismatec to LETI (Mr. Olivier's facility) (3) Two new characterization techniques were discussed: (a) to measure ΔH, follow procedure devised by Artmann at Carnegie-Mellon University - see sketch.



(b) To measure insertion losses, set up a network analyzer as shown in the sketch, sum the outputs, and analyze the output interference to correct for different insertion losses.



The work on garnets at Philips Research Laboratories at Hamburg is sporadic, but it is being pursued from several perspectives by men who are, seemingly, experts. Tolksdorf is a physical chemist; Hansen understands garnet LPE; Dammann knows the optical characteristics; Krumme is exploring a new growth technology, sputtered garnets.

Rather than explicit notes, I brought from Philips a sheaf of reprints, not yet read. I will address briefly here the salient points of our discussions.

Garnets are highly elastic materials, and quite stiff. Consequently, internal strain is rarely compensated by either point or structural dislocations. This indicates that an epitaxial garnet film grown on a substrate whose lattice constant is not identical will incorporate residual strain at the interface which will be gradually eased with increasing distance from the interface. Consequently, a material anisotropy will be inherent, oriented perpendicular to the growth plane. For example, the lattice constant of bismuth substituted YIG exceeds that of GGG, so growth on GGG will compress the BiYIG in the film plane. Because of its elasticity, the BiYIG atomic spacing will be dilated perpendicular to the surface to compensate. This strain becomes less pronounced for regions of the BiYIG farther from the interface - hence an anisotropy. This anisotropy has been assigned two different descriptions: growth-induced and stress-induced anisotropy. It is evidenced both in the magnetic realm, via the alignment of magnetic dipoles in the film perpendicular to its surface, and optically, producing a uniaxially birefringent material. I think growth-induced anisotropy may be ameliorated by means of annealing (annealing, however can permit some widening of the interface layer, where BiYIG and GGG to some extent are intermixed over a thickness of maybe 10 nm). Stress-induced anisotropy can be relieved by piezoelectric bending of the sample (this technique might provide a means for tuning the optical birefringence).

Films grown by LPE are generally drawn from lead (Pb) fluxes (although I think I have heard, previously, that it may be possible to use bismuth fluxes), and from platinum (Pt) crucibles. Hence Pb and Pt are often incorporated in small amounts into YIG films (and their variants). Pb increases the Faraday rotation in YIG but at a high cost - it increases the material's optical absorptivity dramatically.

The amount of bismuth incorporated is dependent on the melt composition, on the growth temperature (how supercooled the flux is - how much the melt is maintained below the temperature at which crystal growth initiates during growth), and on the spin rate of the substrate, which affects melt mixing. These parameters also influence, dramatically, the anisotropy coefficients.

The Philips people concur that although praseodymium and neodymium contribute strongly to increased Faraday rotation in garnets, their natural magnetic dipole moments cause high damping of magnetostatic waves (i.e., high ferrimagnetic resonance linewidths, ΔH). Bismuth enhances Faraday rotation by affecting the extent that the d-shell electrons of iron, those that provide its dipole moment, are shielded in the garnet crystal through a complicated interaction via the oxygen lattice; this mechanism may be described in one of the papers I was given. Bismuth does not have an intrinsic dipole moment.

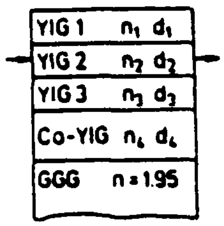
Krumme is growing BiYIG by means of argon ion sputtering from targets. The targets need not contain Pb (or Pt), so that annoyance is alleviated. But other processing difficulties arise. The various elements in the target do not sputter out with the same probability, or with the same velocities. So target compositions must be designed with sputtering-ion energies in mind, or vice versa. Sputtered elements which impact the substrate will stick where they make contact (unless the velocity is too high) - these may not be crystallographically correct positions. An auxiliary ion-glow discharge is often incorporated in the vicinity of the substrate to keep deposited atoms hot enough on the surface that they may find their lowest energy and crystallographically correct positions. Of course, some will, statistically, be re-liberated, another problem for controlled crystal growth. Incidental charge transfer may also influence atomic alignment. Promising results are being achieved with these sputtered garnets. However, I seem to recall that Philips' proposed uses for them do not require single crystal films.

Although we did not discuss the topic thoroughly, I did not get support for the idea, conceived elsewhere, that non-uniform (statistical) population of the appropriately coordinated lattice sites by Bi atoms in YIG (which might occur even in a film where the average bismuth content per unit cell was one and must perforce occur otherwise) could contribute to increased FMR linewidth, ΔH. In fact, even if each unit cell had exactly one Bi atom, there are three appropriately coordinated sites per unit cell, and variation from cell to cell undoubtedly distorts the lattice. Either this possible effect has not been considered, is too difficult to estimate, or has been estimated to be insignificant and/or averaged out in regions of the crystal that are large enough to be important for magnetostatic wave propagation.

Philips scientists felt that BiYIG film growth processes could probably be engineered and controlled to produce films that were consistent in bismuth content from batch to batch within a few percent, and that the lattice constant of the substrate could be adjusted by means of Zr, Mg, and Ca concentrations to match most of the range of desireable Bi substitution. However, the growth- and stress-induced magnetic and optical anisotropies are very critically dependent on all of the various growth parameters. These are almost impossible to replicate from one run to the next, and, in fact, vary during any particular run as the film-producing ingredients in the flux are depleted. They suggested that I seriously study whether useful devices could still result from materials in which the anisotropy might be controlled only within a rather broad range before embarking on a costly materials procurement project.

If I feel the materials anisotropy problem may be resolved positively, Philips would be willing to send me an occasional sample, or perhaps to initiate internally an effort to grow the type of film I need. (They cannot stir management interest in a sale of garnets whose value is less than, say, \$100M!). One further possible impediment is an existing gentleman's agreement to provide BiYIG samples to an ex-colleague who has moved from Philips to a nearby university. Although his interest does not precisely overlap mine, he apparently would be consulted before offering me samples. Publication and scientific interchange, not proprietary interest, are the motivations at Philips.

At Philips, perhaps under Dammann, a multiple layer ridge waveguide structure has been fabricated in garnet materials for use as a polarization rotator/isolator. The geometry, as I remember, is in the sketch below. The second GGG layer provides for a symmetric waveguide structure, while the metal layer absorbs higher order optical modes.

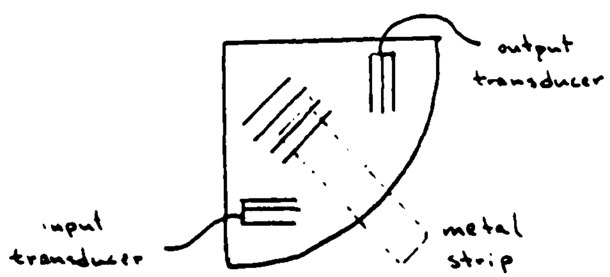


$\Delta n = n_2 - n_3 = 3 \cdot 10^{-3}$
$n_1 = n_3$
$n_4 = n_3$
$d_1, d_3, d_4 = 3-4 \mu m$
$d_2 = 5 \mu m$
YIG 2 waveguide core
YIG 1, YIG 3 cladding
Co-YIG highly absorbing layer

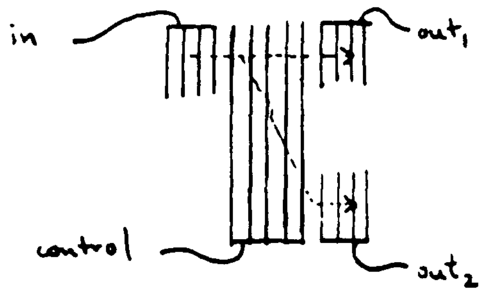
I think I have a paper describing this device in detail.

At Thomson - CSF, Jean-Paul Castera heads a group of six to eight scientists in magneto-optics. Castera himself has done some work with magnetostatic waves in the past, but the current efforts at Thomson are directed primarily toward magnetooptic readout of various memory media: digital magnetic tape, magnetic floppy disks, and audio and video disks where information is stored by optical ablation. Garnets having large values of Faraday rotation are of interest to them, but low FMR linewidths are not important. BiYIG is being used, and I think there is a research-scale effort at Thomson to grow garnets. BiYIG films at Thomson have parameter values in line with those observed elsewhere. In particular, the magnetic anisotropy can be nearly as large as the saturation magnetization in BiYIG; for a particular sample, $Bi_xY_{3-x}IG$ with x=0.3, K_u=1660 Oe, approximately. The magnetooptic readout devices place thin BiYIG films in contact with the recording medium. The magnetic state of the medium determines the alignment, locally, of magnetic dipole domains in the garnet. These domains are read by double-pass transmission, optically; read-out trades could be accessed in parallel with a density as high as 20 μm track separation.

Castera described two devices which use only magnetostatic waves (MSWs). One is a resonator with an amplifier, which incorporates a surface grating structure to reflect the MSWs at right angles. This structure configures the input and output antennas perpendicular to each other, reducing direct rf pickup. In addition, a metal strip has been placed over part of the device to further suppress direct rf transmission between transducers.



The second device is a form of resonant coupler, similar to TRW's MSWFAST concept.



Dr. Friez has built a ridge waveguide on some type of YIG for use as a polarization rotator/isolator. The waveguide structure is achieved by ion beam etching. Up to 99.4% TM to TE conversion has been achieved at 87°C.