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Citation for published version (APA):

Metselaar, R., Huyberts, M. A. H., & Logmans, H. (1975). Light-induced changes in permeability of n- and p-type YIG [yttrium-iron garnet] films. *Journal of Applied Physics*, 46(7), 3171-3174. <https://doi.org/10.1063/1.321966>

DOI:

[10.1063/1.321966](https://doi.org/10.1063/1.321966)

Document status and date:

Published: 01/01/1975

Document Version:

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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Light-induced changes in permeability of *n*- and *p*-type YIG films

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(Received 24 January 1975; in final form 24 March 1975)

The influence of irradiation with white light on the permeability at 77 K on *n*- and *p*-type yttrium iron garnet films has been investigated. The investigations were performed on epitaxially grown films with varying concentrations of Pb and Si. The existence of an irreversible photomagnetic effect in both *n*- and *p*-type YIG is demonstrated. Measurements of the spectral dependence of the photomagnetic effect in the wavelength region 1000–300 nm show a rapid increase of the sensitivity towards shorter wavelengths.

PACS numbers: 75.90., 78.20.L

I. INTRODUCTION

Light-induced changes of the permeability μ in both single-crystalline and polycrystalline^{1,2} *n*-type $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (YIG) have been reported extensively. In Si- or Ge-substituted YIG an irreversible decrease of the permeability is observed after irradiation with white light at 77 K. This has been called a type-II photomagnetic effect.² In samples with silicon (or germanium) concentrations above about 0.01 atoms per formula unit the photomagnetic changes are of a reversible nature (so called type-I effects)². For these high doping levels the behavior of the permeability is difficult to observe since the absolute magnitude of μ becomes very low due to the occurrence of magnetic losses. In the case of *n*-type YIG both type-I and type-II behavior is explained in terms of light-induced charge transfer between Fe^{2+} and Fe^{3+} ions.

When trivalent ions in YIG are replaced by divalent ions, e.g., Ca^{2+} or Pb^{2+} , charge-compensating holes are created which give rise to a *p*-type thermoelectric power. It is usually assumed that the holes are localized on Fe^{3+} ions and it is of interest to see whether the $\text{Fe}^{4+} \leftrightarrow \text{Fe}^{3+}$ system shows photomagnetic effects similar to those due to the $\text{Fe}^{2+} \leftrightarrow \text{Fe}^{3+}$ transitions in *n*-type YIG. Hisatake and Ohta³ found two disaccommodation peaks in polycrystalline Sr-doped YIG, situated at -170 and $+50^\circ\text{C}$, with activation energies of 0.1 and 1.0 eV, respectively. At about 65°C the disaccommodation rate increased considerably upon irradiation. Since the permeability was brought back to its original value by demagnetization we can classify this result as a type-I effect. Though not of a magnetic nature we may also mention experiments of Gyorgy, Dillon, and Remeika,⁴ who investigated the influence of light on the optical absorption constant near 1000 nm in single crystals of Si- and Ca- substituted YIG. In YIG-Si, illumination at temperatures below about 100 K causes an increase in the absorption constant, while the same procedure causes a decrease of the absorption in YIG-Ca.

We have tried to measure photomagnetic effects in polycrystalline Mg-, Ca-, or Sr-substituted YIG, but the results were not convincing. Recently, the technique of liquid phase epitaxy (LPE) growth offered the possibility to obtain single-crystalline YIG films. In these films Pb^{2+} ions are incorporated from the $\text{PbO-B}_2\text{O}_3$ flux, leading to *p*-type behavior.⁵ The amount of lead decreases with increasing growth temperature. By

adding SiO_2 to the melt and varying the growth temperature we can vary the degree of compensation of Si^{4+} by Pb^{2+} . In this way both *n*- and *p*-type films can be produced. Details of the experiments are given in Sec. II, and the light-induced changes in permeability measured on these films are discussed in Sec. III.

Since the absorbance of the approximately $5\text{-}\mu\text{m}$ thin films is so much lower than that of the 1-mm-thick cores used for the earlier experiments, we are now able to investigate the dependence of the photomagnetic effect on the wavelength of the light in the visible region as well. Results of these measurements are given in Sec. IV.

II. EXPERIMENTAL

The garnet films were prepared by the technique of liquid phase epitaxy by Robertson of this laboratory. Details on the growth conditions of the films used for our investigations are given in Refs. 5 and 6. These two papers also describe the measurements of thickness, electrical conduction, Seebeck coefficient, and optical absorption constant. The films were grown by dipping $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ substrates in a $\text{PbO-B}_2\text{O}_3$ melt containing the appropriate amounts of Y_2O_3 and Fe_2O_3 . Two series of films were investigated. In series A 0.044 mole% of SiO_2 was added to the melt, in series B 0.44 mole% of SiO_2 was added. It has been shown in Ref. 5 that the Pb and Si concentrations in the films can be varied by varying the growth temperature T_g . From an electron microprobe analysis it was found that the lead concentration $[\text{Pb}]$ expressed in at. % per formula unit can be represented by

$$[\text{Pb}] = K_0 \exp(-T_g/T_0). \quad (1)$$

For our films, with T_g between 790 and 860°C , the constant K_0 was found to be 5.2×10^7 and 7.0×10^7 for series A and B, respectively. For both series the constant $T_0 = 55^\circ\text{C}$. According to the microprobe analysis the silicon concentrations can be represented by a similar equation, where for series A $K_0 = 800\text{--}1800$, $T_0 = 124^\circ\text{C}$, while for series B $K_0 = 7500$ and $T_0 = 124^\circ\text{C}$. The inaccuracy in the silicon analysis is largest for series A with a low dopant concentration. Referring to the data shown in Table I we estimate for the extreme cases of film 1A $[\text{Pb}] - [\text{Si}] = 0.25 \pm 0.02$ at. %, and for film 17A 0.011 ± 0.005 at. %.

For measuring the permeability of the thin platelets

we used a specially designed apparatus. In this configuration the platelet to be measured forms a high-reluctance part of an inductor, which in turn determines the frequency of a low-amplitude LC oscillator. The inductance consists of a half-potcore with polished faces and with a wire-wound solenoid; the sample is placed on top of this and pressed against the potcore by a spring-loaded holder with a flat quartz window through which radiation can be applied. The absolute accuracy of a permeability determination is estimated at about 30% due to a lack of knowledge of the precise flux configuration, air gap, etc. However, the sensitivity to small changes in permeability is such that changes of $\Delta\mu h \approx 1$ are detectable (h is the platelet thickness in μm).

For measuring the light-induced change in permeability we used the following procedure. After cooling in the dark by immersion of the sample holder in liquid nitrogen, the sample is ac demagnetized, and the permeability value obtained 5 sec after demagnetization is called μ_{dark} . The sample is kept in the dark for a sufficiently long time to enable the observation of possible disaccommodation effects. Thereafter, the sample is irradiated with the unfiltered light of a xenon lamp (Philips CSX 150 W) until a saturation value μ_{irr} is reached. Finally demagnetization leads to a permeability value μ_{dem} .

For the investigation of the spectral dependence of the photomagnetic effect a set of interference filters (Balzers, half-width 40 nm) in combination with the appropriate glass filters was used. A part of the light beam was directed towards a calibrated phototube (Philips 150 CV or 150 UV) to allow a continuous read-

ing of the photon flux. For measurements in the ultraviolet region a mercury lamp (Philips CS 200 W) was used as a light source.

III. PHOTOINDUCED PERMEABILITY CHANGE IN n - AND p -TYPE YIG

In Table I a survey is given of a number of characteristic properties of the YIG films investigated. The films are listed in the sequence of increasing growth temperature and as a consequence the lead excess decreases in this order. Films 1–10 are all p type, while films 11–21 are n type. The p -type behavior is due to the excess of lead ions which act as acceptor centers. Due to the higher value of T_0 in Eq. (1) for Si in comparison with Pb, the silicon donor concentration exceeds the lead acceptor concentration at the highest growth temperatures. This explains the n -type behavior of films 20 and 21. The n -type behavior observed for films 11–19 in spite of the small excess of Pb over Si is attributed to the presence of oxygen vacancies acting as donor centers.⁵ It has been shown earlier that the type-II photomagnetic effect in undoped polycrystalline YIG is indeed due to the presence of oxygen vacancies.² The specific resistance R_0 is given in column 3. It can be seen that R_0 reaches a maximum near the transition from n to p type. Values of the initial permeability measured at 50 kHz are given in columns 4–7 of Table I. The room-temperature values $\mu(300)$ were measured 5 sec after ac demagnetization. A comparison with the permeability obtained at 77 K, after cooling in the dark shows that nearly all of the p -type films have $\mu(300) < \mu(77)$, while n -type films show a normal behavior, i. e., $\mu(300) > \mu(77)$. While none of the films show dis-

TABLE I. Impurity content, specific resistance, and permeability at 300 and 77 K of two series of YIG films. For films of series A 0.044 mole% of SiO_2 was added to the melt, for series B the SiO_2 content was about 0.4 mole%. Films 1–10 are p type, films 11–21 are n type.

Sample	[Pb] – [Si] (at.%)	R_0 ($\Omega\text{ cm}$)	$\mu(300)^a$	$\mu_{\text{dark}}(77)^a$	$\mu_{\text{irr}}(77)^b$	$\mu_{\text{dem}}(77)^b$
1A	0.25	3.0×10^6	24	70	33	34
2A	0.33	3.7×10^6	15	75	50	53
3A	0.17	6.2×10^6	20	84	40	55
4B	0.21	6.8×10^6	13	17.0	14.5	15.7
5A	0.15	7.8×10^6	19	42	30	33
6B	0.27	8.9×10^6	7	29.2	27.7	28.5
7A	0.09	1.1×10^7	22	27	22.6	24.6
8B	0.12	2.1×10^7	10	37	36	36
9B	0.076	5.6×10^7	13	10.0	9.5	9.7
10A	0.073	2.3×10^8	24	22.4	19.9	21.4
11A	0.027	6.0×10^8	75	32	29.7	30.6
12A	0.040	2.6×10^8	67	27.2	24.7	25.7
13A	0.058	1.3×10^8	38	27.0	23.8	25.3
14B	0.047	1.1×10^8	9	27.3	26.7	26.4
15A	0.015	5.9×10^7	26	26.3	24.6	25.3
16B	0.035	1.2×10^7	25	17.3	16.4	16.4
17A	0.011	2.9×10^6	49	25.3	22.7	23.6
18B	0.011	6.4×10^4	50	7.4	6.5	6.9
19B	0	4.6×10^4	124	7.1	6.6	6.7
20B	–0.016	1.8×10^4	115	23	23	23
21B	–0.011	1.6×10^4	125	4.1	4.1	4.1

^a Measured 5 sec after demagnetization.

^b Measured after illumination until a saturation value is reached.

FIG. 1. Permeability μ_t , measured t sec after demagnetization. Solid line: μ_5 as a function of temperature. Dashed line: Disaccommodation $(\mu_{60} - \mu_5)/\mu_5$, as a function of temperature.

accommodation (DA) at room temperature, films 1–4 exhibit DA at 77 K. This effect was most pronounced in film 3A. For this film the permeability as a function of temperature is shown in Fig. 1. The solid line gives μ values 5 sec after demagnetization, while the dotted line shows the DA $(\mu_{60} - \mu_5)/\mu_5$. Here μ_t is the permeability value obtained t sec after demagnetization. The activation energy for the low-temperature DA process is about 0.2 eV. This value gives an estimated relaxation time at room temperature of $\tau \sim 10^{-7}$ sec. The occurrence of this rapid DA could be an explanation for the low permeability values $\mu(300)$ observed for the p -type films. We could not verify this assumption further, however. Both at 77 and 300 K the permeability of film 3A is nearly constant in the frequency range 500–50 000 Hz. Films of series B having a higher absolute Pb and Si content than films of series A also tend to have lower values of $\mu(300)$.

The data in column 6 of Table I show the saturation value of the permeability after irradiation at 77 K with white light. It can be seen that nearly all compositions show photomagnetic effects. The decrease of the permeability can only be removed partly by demagnetization (cf. column 7), which proves that we have both type-I and type-II photomagnetic effects. This is the first time that type-II effects have been observed in p -type YIG. In n -type YIG we assume that the light induces an electron transfer from an Fe^{2+} -donor pair to an Fe^{3+} ion which is far away from the donor.¹ Since the site symmetry is higher for the isolated ferrous ion than for the ferrous ion near a donor, the orbital momentum contribution to the anisotropy of the Fe^{2+} ion increases after irradiation. As a result of the increased anisotropy of the Fe^{2+} centers the domain walls are pinned more strongly, hence the permeability decreases.

The p -type behavior of acceptor-doped YIG is explained by the presence of Fe^{4+} ions.⁷ Very little is known about the properties of these ions in the garnet structure. According to Tchernev⁸ Fe^{4+} ions are located on the tetrahedral sites. In view of the ionic radius such a site preference also has to be expected. The magnetic properties of Fe^{4+} in a tetrahedral surrounding will be comparable to the properties of Fe^{2+} on octahedral sites

since both ions will have threefold degenerate ground states. Indeed, according to Sturge *et al.*⁹ values of the cubic anisotropy contribution of Fe^{4+} on d sites and Fe^{2+} on a sites in YIG are of the same order of magnitude. Analogous to the explanation of the photomagnetic effect in n -type garnet we can therefore assume that irradiation of the p -type samples induces a hole transfer from an Fe^{4+} site near an acceptor to a site far away from an acceptor.

IV. THE WAVELENGTH DEPENDENCE OF THE PHOTOMAGNETIC EFFECT

The spectral dependence of the class-II photomagnetic effect of an n -type sample was investigated by measuring at 77 K the rate of change of the permeability as a function of the wavelength of the irradiating light. To characterize the photomagnetic effect we use the change in the inverse susceptibility $\chi = (4\pi\chi)^{-1}$, since this quantity is proportional to the wall stiffness. The sensitivity β is defined by $\beta = (1/I)(d\chi/dt)_{t=0}$, where I is the photon current density, and the derivative is taken at the moment of switching on the light. In an earlier investigation¹ we measured the sensitivity of a polycrystalline core of about 1 mm thickness. In that study the investigations were confined to the wavelength region 1400–700 nm. Above 1400 nm the sensitivity was too low to produce measurable changes in μ , below 700 nm the optical density (absorption constant times thickness) of the sample was too high. The general conclusion from these experiments was that the sensitivity increases continuously over the wavelength region investigated. Using the thin films available now we could extend the

FIG. 2. Solid lines: Absorption constant α as a function of wavelength, and spectral dependence β of the photomagnetic effect at 77 K. $\beta = (1/I)[d(4\pi\chi)^{-1}/dt]$. Dashed line: Spectral dependence of the photomagnetic effect after correction for absorption. $\beta' = \{\alpha h/[1 - \exp(-\alpha h)]\}\beta$, where h is the sample thickness.

measurements to 300 nm. First, we verified that at a fixed wavelength, using incident powers between 4 and 60 μW , the rate of change $(dr/dt)_{t=0}$ was proportional to I . During the investigation of the spectral dependence the total incident light power was kept approximately constant at 30 μW . The results are shown in Fig. 2. Together with the sensitivity β we have plotted the absorption constant α of the same film. It is seen that α increases towards higher energies. The structure near 600, 670, and 900 nm is due to crystal field transitions of the ferric ions. With our planar geometry we can easily correct β for the influence of the light absorption. The corrected sensitivity value β' is obtained from β by

$$\beta' = \frac{\alpha h}{1 - \exp(-\alpha h)} \beta,$$

where h is the film thickness.

The curve for β' (dashed line in Fig. 2) rises smoothly, without any structure near 600 nm. Using literature data¹⁰ for the absorption constant below 500 nm we estimate that the corrected sensitivity at 320 nm is about $3 \times 10^{-15} \text{ cm}^2$. Due to the low optical density of our film above 1000 nm we were not able to investigate the behavior of β in this region. Comparison with the results obtained on n -type YIG, as reported in Ref. 1, suggests that β' increases at least five orders in magnitude from 1400 to 300 nm. The small polaron model as developed by Reik and Schirmer¹¹ to explain type-I photoinduced effects does not seem applicable to our results for type-II effects. According to this model the sensitivity function is described by a Poisson distribution curve centered around about 1000 nm. Obviously this does not correspond with the β' curve shown in Fig. 2. The wavelength dependence of the sensitivity is, however, related to that of the photoconductivity as reported for YIG.¹² The onset of photoconductivity was found at 1100 nm, with a rapid increase towards shorter wavelengths. According to measurements of the electrical conductivity the band-gap energy is about 2.9 eV. This suggests

that both in photoconductivity and in the photomagnetic effect transitions across the band gap are involved. Since both effects are also strongly influenced by impurities¹³ no further conclusions can be drawn as yet. Measurements of the photoconductivity of our samples are in progress.

ACKNOWLEDGMENTS

The authors are grateful to P.J. Rijnierse for his contributions to the design of the setup for permeability measurements on thin films, and to R.W. Teale and P.K. Larsen for valuable discussions. They are indebted to J.M. Robertson for providing the garnet films.

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