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Photoinduced Magnetic Effects in YIG(Si)

Abstract: This paper summarizes the recent investigations of photoinduced effects in magnetic materials. To date it has been shown that the uniaxial anisotropy, strain, linear dichroism, coercive force and initial permeability can be modified by infrared radiation. The theory interpreting the coercive force and permeability experiments differs from the model used to describe the first three effects and is not discussed here. The first three effects can be interpreted fairly well in terms of a model which has single Fe^{2+} ions preferentially occupying inequivalent octahedral sites. It is shown that with polarized light more than half of the available Fe^{2+} ions can be selectively moved among specific types of sites. Irradiation with unpolarized light essentially leads to a distribution appropriate for thermal equilibrium at the temperature of the sample.

Introduction

This paper is concerned with a new class of magneto-optical effects in which irradiation leads to a change in an intensive property of a magnetic crystal. The primary interest here is on the modification of the magnetic properties. The changes that are induced by the irradiation may be permanent at sufficiently low temperatures or they may be only temporary at higher temperatures. The groups at Philips Research Laboratory, Mullard and the University of Sheffield have come to refer to this field as the study of photomagnetic effects. Since the photomagnetic effect has already been defined as "the direct effect of light on the magnetic susceptibility of certain substances,"[1] we prefer the term *photoinduced* to describe the effects considered here. This concern about nomenclature can be justified by the ever-increasing use of permuted title indexes. To date, the properties that have been influenced by light in this context are the uniaxial anistropy, the strain, the linear optical dichroism, the coercive force and the magnetic permeability. The materials that have shown some or all these effects are $Y_3Fe_{5-\delta}Si_\delta O_{12}$; YIG(Si) and $Cd_{1-\delta}Ga_\delta Cr_2Se_4$.

The history of the photoinduced effects dates from the observation by Teale and Temple[2] that the field for ferromagnetic resonance in YIG(Si) could be influenced by irradiation. Soon afterwards Enz and van der Heide[3] showed that infrared radiation produces substantial changes in the permeability and coercive force in YIG(Si). Similar observations were reported by Lems et al.[4] for the chalcogenide spinel $Cd_{1-\delta}Ga_\delta Cr_2Se_4$. Subsequently, by measuring the static torque at 4.2°K, Pearson et al.[5] showed the importance of the plane of polarization of the irradiating light. This work was reviewed by Teale, Temple, Enz and Pearson at the 1968 Conference of Magnetism and Magnetic Materials[6] and by Enz, Lems, Metselaar, Rijnierse and Teale[7] at the 1969 Intermag. Meeting in Amsterdam. Later Dillon et al.[8] clearly showed that in a (001) plate of YIG(Si) the induced anisotropy axis could be changed from [110] to $[\bar{1}10]$ by changing the plane of polarization of the incident light. Furthermore, it was demonstrated that a linear dichroism is associated with the induced anistropy. The existence of a photoinduced strain in YIG(Si) was reported recently.[9]

All the work on photoinduced effects in YIG(Si) rests in large part on the studies of the thermally induced anistropy in this material.[10,11] The model of Fe^{2+} ions occupying magnetically inequivalent sites used to explain the thermally induced effects also applies to the optical effects. Consequently it is convenient to start this paper with a discussion of thermally induced effects (magnetic annealing) in YIG(Si). Also it appears as if the explanation of the optically induced effects on the permeability and the coercive force differs substantially from the theory used to account for the other optically induced changes. The theory of these photoinduced changes in the initial permeability are very well presented in Ref. 4 and, therefore, we will not discuss these effects here.

Magnetic annealing

We start with a quite general discussion of magnetic anneal. The theory was developed by Néel,[12] Taniguchi and Yamamoto[13] and in a more general form by Slonc-

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zewski.[14] The following development of the theory, limited to cubic crystals, is taken essentially from their work. To proceed we let the induced anisotropy arise from n_0 unspecified impurities or defects per unit volume. We further assume that the ith impurity or defect can be characterized by its energy E_i and its axis of local symmetry $\mathbf{L}^i$. We let

$$E_i = -\epsilon(\cos^2\theta_i - 1/3),$$

where θ_i is the angle between the magnetization $\mathbf{M}$ and $\mathbf{L}^i$.

To continue, we let the material be annealed at an elevated temperature T_a with $\mathbf{M}$ parallel to the unit vector $\boldsymbol{\beta}$. It is assumed that enough time is spent at T_a so that the material will reach equilibrium. The material is then quenched to a lower temperature T. For an arbitrary orientation of $\mathbf{M}$, specified by the unit vector $\boldsymbol{\alpha}$, it can be shown that

$$E = -F(\alpha_1^2\beta_1^2 + \alpha_2^2\beta_2^2 + \alpha_3^2\beta_3^2) - G(\alpha_1\alpha_2\beta_1\beta_2 + \alpha_1\alpha_3\beta_1\beta_3 + \alpha_2\alpha_3\beta_2\beta_3) \qquad (1)$$

where α_p and β_p are the components of $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}$ respectively with respect to the cubic axis. Also we have

$$F = n_0(kT_a)^{-1}\epsilon(T)\epsilon(T_a)\langle L_1^{i4} - L_1^{i2}L_2^{i2}\rangle, \qquad (2)$$

$$G = 4n_0(kT_a)^{-1}\epsilon(T)\epsilon(T_a)\langle L_1^{i2}L_2^{i2}\rangle. \qquad (3)$$

Here L_p^i are the components of $\mathbf{L}^i$ and the last brackets in each of the above two equations mean the average over the allowed orientations of $\mathbf{L}^i$. To obtain Eqs. (2) and (3), use was made of the assumption that $\epsilon/kT_a \ll 1$, which is true of most cases of interest. For $\mathbf{L}^i$ along $\langle 100\rangle$, $G = 0$. For $\mathbf{L}^i$ along $\langle 111\rangle$, $F = 0$ and for $\mathbf{L}^i$ along $\langle 110\rangle$, $G/F = 4$.

For a (100) plane Eq. (1) reduces to

$$E = (F\cos 2\phi_a)\sin^2\theta - (1/4\, G\sin 2\phi_a)\sin 2\theta, \qquad (4)$$

where $\alpha_1 = \cos\theta$ and $\beta_1 = \cos\phi_a$. The results of the formal analysis are easily understood in this simple plane. If $\mathbf{L}^i$ is along $\langle 111\rangle$ and $\boldsymbol{\beta}$ along $\langle 110\rangle$ ($\phi_a = 0°$), we have $E = 0$ and no anisotropy is induced. This fact is evident from the symmetry since in this case $\mathbf{M}$ during the anneal makes equal angles with $\mathbf{L}^i$ (all the θ_i are equal) and no preferred state can occur. Similarly, there is no induced anisotropy if $\mathbf{L}^i$ is along $\langle 100\rangle$ and β along $\langle 110\rangle$.

Now for simplicity we will limit the discussion of thermally induced effects to YIG(Si). The formula for YIG(Si) can be written as

$$\{\mathrm{Y}_3^{3+}\}[\mathrm{Fe}_{2-\delta}^{3+}\,\mathrm{Fe}_\delta^{2+}](\mathrm{Fe}_{3-\delta}^{3+}\,\mathrm{Si}_\delta^{4+})\mathrm{O}_{12},$$

where the first bracket denotes the dodecahedral sites, the square bracket the octahedral sites and the (rounded) bracket the tetrahedral sites.[15] The active impurity then is the Fe^{2+} ions residing on the octahedral sites. Each of these octahedral sites (point symmetry $\bar{3}$) has a trigonal distortion along one of the $\langle 111\rangle$ axes. If we assume that the observed effects can be attributed to single octahedral sites occupied by the Fe^{2+} ion and if we assume that the nearby compensating Si^{4+} does not disturb the local symmetry, we find that $\mathbf{L}^i$ is along one of the $\langle 111\rangle$ directions. Then, according to Eqs. (2) and (3), $F = 0$. Also $n_0 = 4.24 \times 10^{21}\delta$. Note that the occupation by an Fe^{2+} in an Fe^{3+} lattice merely means that there is an extra electron on a particular site. Thus an Fe^{2+} ion can move merely by the transfer of an electron. We let the barrier height for this electron transfer be V_0. The activation energy for electron "hopping" electrical conduction is then V_0. The parameter ϵ, in this case, is related to the spin orbit coupling constant. Along with Refs. 10 and 11 we take ϵ to be positive, although we have no experimental justification for this choice.

Now, if we set $\epsilon(T) = \epsilon(T_a)$, the torque in the (100) plane is given by

$$\tau = \frac{2n_0\epsilon^2}{9kT_a}\sin 2\phi_a \cos 2\theta. \qquad (5)$$

Hunt,[10] using crushed single crystals with $0 < \delta < 0.16$, verified the linear dependence of τ on n_0 (at 50°K). The dependence of τ on the angle θ was investigated by cooling a single crystal (100) disc ($\delta = 0.05$) from room temperature to 15°K for $\phi_a = 45°$ and $\phi_a = 0$ and then measuring τ as a function of θ.[10] For $\phi_a = 45°$ the induced torque is well represented by the $\cos 2\theta$ term. For $\phi_a = 0$ the induced torque is not zero as given by Eq. (5) but rather is approximately a $\cos 2\theta$ curve about 0.3 times as large as the one obtained for $\phi_a = 45°$. So we have $F/G \approx 0.1$ rather than the predicted value of zero.

The experiments such as the one described here, which imply an anneal at T_a and a measurement at T, are sometimes somewhat ambiguous since some annealing can take place as the sample is cooled from T_a to T. Consequently the correct value of T_a for Eq. (5) may not be the annealing temperature but rather some lower temperature. Hunt[10] chose T_a equal to T, the measuring temperature. With this assumption and the value of δ from spectrographic analysis he found that $\epsilon = 5\ \mathrm{cm}^{-1}$.

We digress here to discuss the difficulties of accurately determining δ. The experience of the group at Bell Telephone Laboratories has been that in crystals of the same batch, δ may vary by a factor of two from one crystal to another. Therefore, if δ is not determined for the actual samples used, the magnetic measurements will be somewhat in doubt. Thus the exact value of ϵ and the linear dependence of τ on δ discussed in connection with Eq. (5) cannot be taken as completely determined at present. We have adopted the measurement of the optical absorption at some wavelength (such as 1.06μm) as our analysis for

Fe^{2+}. Use of a curve of optical absorption as a function of Si concentration, such as given by Wood,[16] allows us to determine the Fe^{2+} concentration on the actual discs used for the magnetic measurements. This determination is, of course, nondestructive.

We have measured the torque from a (001) plate ($\delta = 0.03$) at $\theta = 0$ obtained by cooling from room temperature to 4.2°K at various angles ϕ_a. The results are shown in Fig. 1. The solid curve

$$\tau = \frac{2\epsilon n_0}{3} \tanh\left(\frac{\epsilon}{3kT_a}\sin 2\phi_a\right)\cos 2\theta$$

is fitted to the experimental points. The torque is given by this relation rather than Eq. (5) if the assumption $\epsilon(T_a)/kT_a \ll 1$ is not valid. Equation (5) is, of course, the leading term of the preceding expression for the torque. From Fig. 1 we find that $\epsilon(T) = 2.0$ cm^{-1} and that $\epsilon(T_a)/kT_a = 4.1$. If we arbitrarily take T_a as 10°K, $\epsilon(T_a) = 30$ cm^{-1}, an order of magnitude greater than $\epsilon(T)$. This difference between $\epsilon(T_a)$ and $\epsilon(T)$ is probably not significant but reflects only the difficulty previously discussed that arises when the annealing experiments are interpreted with the very simple concepts used to derive Eq. (5). The study of rotational hysteresis as a function of temperature has shown that there are at least two mechanisms contributing to the annealing effect.[10] Furthermore, these mechanisms seem to be governed by quite a large range of activation energies. As a result, it is not surprising that in a given annealing experiment T_a cannot be given with any certainty.

Some of the difficulties inherent in the annealing experiments just described have been eliminated in the study by Van Groenou et al.[11] They used a similar model to describe YIG(Si) but did not assume the existence of thermal equilibrium as was done to derive Eqs. (2), (3) and (5). Rather they calculated the time effects in an explicit systematic way. By performing the rotational hysteresis experiments at a fixed temperature the difficulty of determining the actual annealing temperature (T_a) is eliminated. Their interpretation of the experiments with a crystal ($\delta = 0.03$) at 4.2°K yields $\epsilon = 1.0$ cm^{-1}. The value of F/G obtained from these rotational hysteresis experiments varies from 0.2 at 1.5°K to 0.07 at 20°K.

We may summarize the experiments on magnetic annealing of YIG(Si) by noting that the model of preferential occupation of the octahedral sites by Fe^{2+} at least semiquantitatively explains most of the experimental observations. A major difficulty of the model is that V_0, the height of the barrier for electron hopping from one site to another, does not appear to be well defined. In fact, this model does not account for the presence of at least two relaxation processes in a straightforward way. Furthermore, the model does not explain the slight deviation from the expected value of F/G. In spite of these difficulties the model is a very convenient and relatively accurate guide to the discussion and interpretation of photoinduced effects.

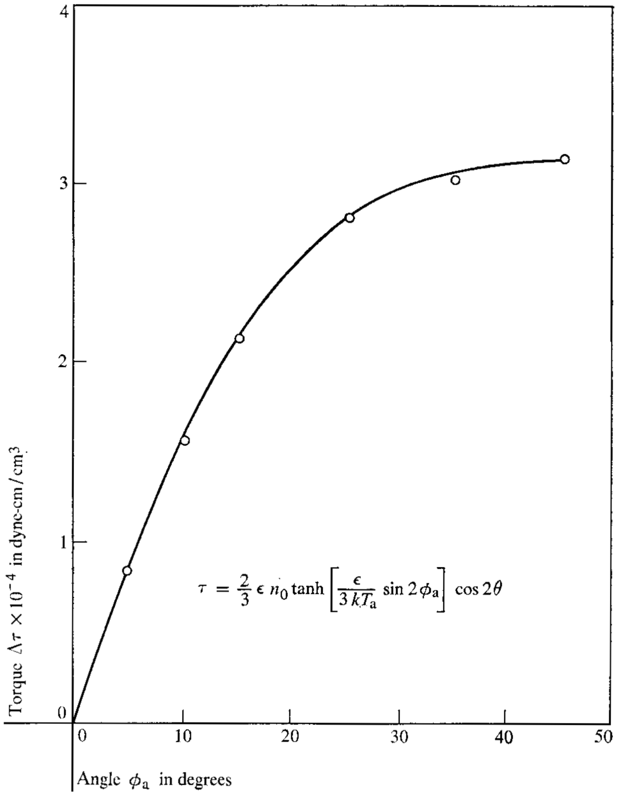


Figure 1 The induced torque measured with $\mathbf{H}_0$ along [010] for a (001) plate of YIG(Si) ($\delta = 0.03$) cooled from room temperature to 4.2°K with $\mathbf{H}_0$ at the angles ϕ_a. The solid line is a theoretical curve fitted to the experimental points.

Photoinduced anisotropy

- *Unpolarized light*

The first observation of photoinduced effects in YIG(Si) was made by Teale and Temple,[2] who showed that the field for rf resonance could be influenced by infrared irradiation. The field for microwave resonance is given by $H_r = H_0 + H_k + \delta H$, where H_0 is the applied field, H_k the effective anisotropy field and δH the dynamic shift due to the presence of Fe^{2+}. The effective anisotropy field (H_k) includes the anisotropy induced by cooling the sample in a magnetic field in addition to the crystalline anisotropy. Teale and Temple cooled a YIG(Si) sphere ($\delta = 0.1$) from 300°K to 20°K with H_0 along [111]. Then they rotated the field to $[11\bar{1}]$ and measured the field for rf resonance. Since [111] and $[11\bar{1}]$ are crystallographically equivalent, any difference in the field for resonance must be attributed to the uniaxial induced ani-

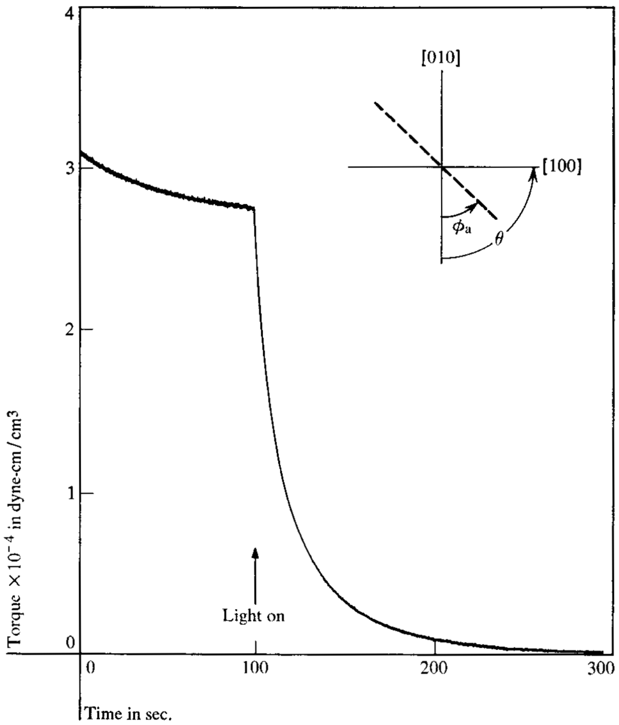


Figure 2 A recorder plot of the relaxation of the thermally induced torque. The sample ($\delta = 0.03$) was cooled to 4.2°K with $\mathbf{H}_0$ along $[\bar{1}10]$ ($\phi_a = 45°$). The torque was measured with $\mathbf{H}_0$ along [100] ($\theta = 90°$). A light (480 mW/cm^2) was turned on at $t = 100$ sec.

sotropy. Here we have tacitly assumed that δH is unchanged. After rotation H_r falls logarithmically by about 50 Oe. The relaxation time for this process is approximately 10 min. However, if the sample is illuminated with unpolarized infrared radiation, H_r falls rapidly by about 200 Oe to approximately the value it would have had if the sample had been cooled along $[11\bar{1}]$ instead of [111]. These experiments, then, show that part of the anisotropy induced by cooling to 20°K spontaneously relaxes by thermal excitation and that infrared irradiation in the presence of a magnetic field is essentially equivalent to a slow magnetic anneal from room temperature. The active radiation for this experiment was in the range 2.2μm $> \lambda > 0.8\mu$m and the light power reaching the sample was approximately 10μW.

These results are readily explained by the rearrangement of Fe^{2+} ions over the various octahedral sites. The rearrangement results in part from thermal activation over the barrier V_0 by some of the Fe^{2+} ions and in larger part from the absorption of a photon which gives an electron on an Fe^{2+} ion sufficient energy to move to another site. However, due to the complications inherent in the interpretation of microwave resonance experiments we delay further detailed explanations until we have considered essentially static experiments. In this connection it should be mentioned that the presence of both thermally induced effects and δH in the expression for the field for resonance has made interpretation of each individually quite difficult in the past.[17] Therefore, Epstein and Bullock[18] plan to use the optically induced effects just discussed to carefully eliminate the thermally induced effects and to study only δH.

We now consider experiments which show the effects of radiation on the thermally induced torque. In these experiments the sample is slowly cooled to helium temperatures with $\mathbf{H}_0$ along some crystallographic direction and then the torque is measured as a function of time with a recording torque magnetometer. Because the projections of the various $\langle 111 \rangle$ directions are orthogonal in the (001) plane, a further simplification results if we limit our discussion of the static effects to this plane. To derive the expression for the torque in this plane we let n_1 be the number of Fe^{2+} ions per unit volume occupying sites whose $\bar{3}$ axis project on the [110]. Let n_2 be the corresponding number for the sites that project on the $[\bar{1}10]$ axis. Then the high field torque is given by

$$\tau = \frac{2\epsilon}{3}(n_1 - n_2)\cos 2\theta. \tag{6}$$

For a sample equilibrated at $T = T_a$ with $\mathbf{H}_0$ at the angle ϕ_a and then quenched at time $t = 0$ to some lower temperature we have that

$$n_1 - n_2 = n_0 \tanh\left[\frac{\epsilon}{3kT_a}\sin 2\phi_a\right].$$

If $\epsilon/kT_a \ll 1$ we can expand and, as expected, obtain Eq. (5). However, to discuss the optical effects the form of Eq. (6) is somewhat more convenient.

Typical experimental results are shown in Fig. 2 where we show a recorder plot of τ as a function of time for a sample cooled to 4.2°K with $\mathbf{H}_0$ parallel to $[\bar{1}10]$ and then rotated so that $\mathbf{H}_0$ is along [100]. We take time $t = 0$ after this rotation, which takes about 5 sec to complete. For this sample $\delta = 0.03$ and the initial induced torque is 3.2×10^4 dyne-cm/cm^3. After 100 sec the torque has relaxed by about 12 percent of its original value. Then unpolarized light is turned on and in the next 100 sec the torque falls rapidly to a value close to zero. The light source was a microscope lamp focused on the sample to give a light intensity of approximately 480 mW/cm^2. Figure 2 is, of course, similar to the plot of the field for resonance as a function of time given in Ref. 2, since both result from the relaxation of the thermally induced torque. The relaxation time with the light on, given in Ref. 2, is of the order of 10^3 sec rather than the 10^2 sec observed in Fig. 2. This difference in the observed relaxation time is probably due to the difference between the irradiating energies used in the two cases.

Another way to observe the induced anisotropy employs a rotating sample magnetometer (RSM) and lock-in amplifier.[19] Some results of this method obtained at 77°K with the same crystal used for Fig. 2 are shown in Fig. 3[20] where we plot the output ($V_\perp$) of the RSM at 2ω and 4ω as a function of time for a sample cooled along $\langle 110 \rangle$. The output $V_\perp$ is given by

$$V_\perp = C\omega M_s \left[\frac{K_1}{HM_s} \sin 4\omega t + \frac{K_u}{HM_s} \sin 2\omega t - \left(\frac{K_u}{M_s H} \right)^2 \sin 4\omega t \right], \qquad (7)$$

where C is the coupling constant of the pick-up coil. From Eq. (7) we see that for sufficiently large fields, $V_\perp(2\omega)$ measures the induced uniaxial component K_u and $V_\perp(4\omega)$ measures the usual four-fold crystalline anisotropy constant K_1. Thus, we see from Fig. 3 that K_u has the same behavior as that given by the rf and torque measurements just discussed. We also see that K_1 does not relax with time nor is it affected by the radiation. (The small change seen in $V_\perp(4\omega)$ can be shown to be due to the last term in Eq. (7) and thus does not reflect a change in K_1.)

So far we have seen that the effects resulting from magnetic anneals can be removed by irradiating with unpolarized light if $\mathbf{H}_0$ is along $\langle 100 \rangle$ as it is in Fig. 2, or if the direction of $\mathbf{H}_0$ time-averages to zero as it does for the RSM measurements of Fig. 3. From Eq. (6) this shows that illumination makes the population difference, $n_1 - n_2$, tend to zero. In the framework of our model we say that the radiation excites the electron trapped on an Fe^{2+} ion to an excited state about $10^4 \mathrm{cm}^{-1}$ above the ground state. The overlap of this excited state is sufficiently large that the electron is free to move to other octahedral sites. The energy of the excited state is so much larger than any energy difference in the ground state caused by the presence of the magnetic field that it is tempting to assume that the excited electrons fall back at random into the four possible types of octahedral sites. This is the assumption made by Pearson et al.[5] For $\mathbf{H}_0$ along $\langle 100 \rangle$ or for a zero-time average of the applied field this certainly appears to be the case and irradiation leads to a random distribution over the octahedral sites. This new distribution removes the effects of any previous thermal anneals. However, if $\mathbf{H}_0$ is along $\langle 111 \rangle$ the excited electrons may not fall back at random. Rather the electrons seem to fall back such as to establish thermal equilibrium appropriate for the temperature of the sample being irradiated. That this may be the case can be seen by again considering the results given in Ref. 2 for the behavior of the field for resonance, H_r. As we stated earlier these results showed that the infrared irradiation was essentially equivalent to a slow anneal from room temperature. In fact, irradiation is slightly more effective than anneal since H_r after illumination is somewhat lower than H_r after slow cooling. Of course, this new distribution could also be obtained by preferential excitation of the Fe^{2+} ions on the various octahedral sites and a random fall back. At present we cannot distinguish between preferential excitation and nonrandom fall back.

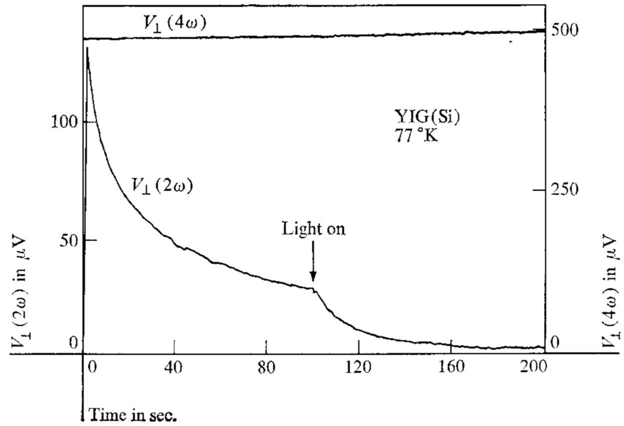


Figure 3 The output of a rotating sample magnetometer plotted as a function of time for the sample used in Fig. 2 cooled to 77°K with $\mathbf{H}_0$ along [110]. The term $V_\perp(2\omega)$ is a measure of the induced uniaxial anisotropy and the term $V_\perp(4\omega)$ measures the crystalline four-fold anisotropy. The data were taken by P. J. Flanders, Ref. 20.

The changes with time of the induced anisotropy in the absence of light indicate the presence of thermal redistribution of electrons at 4.2°K (Fig. 2) and at 77°K (Fig. 3). A single value for the height of the barrier to electron hopping (V_0) obviously cannot account for the experimental observations at these two temperatures. Therefore we have further evidence for the existence of at least two types of mechanism that can lead to induced magnetic effects.

• *Polarized light*

So far we have seen that irradiation with unpolarized light tends to distribute the Fe^{2+} ions in a thermal distribution and thereby eliminate the influence of any preceding thermal treatments. Pearson, Annis and Kompfner[5] have shown that the distribution of the Fe^{2+} ions can be selectively changed by control of the plane of polarization of the incident light. Their experiments consisted of measuring the torque of a YIG(Si) sphere in the (110) plane at 4.2°K under various conditions of illumination. The depolarization effects present in a spherical sample and the nonorthogonality of the $\langle 111 \rangle$ directions in the (110) plane make it more convenient to discuss the same experiment performed with a (001) disc.

A (001) disc of YIG(Si) cooled to 4.2°K with $\mathbf{H}_0$ along [010] will exhibit zero torque if $\mathbf{H}_0$ is kept along [010].

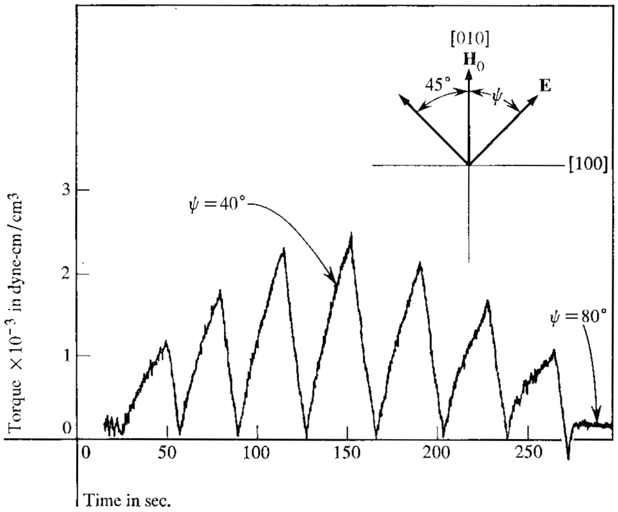


Figure 4 The induced torque as a function of time for a (001) disc ($\delta = 0.03$) cooled to 4.2°K with $\mathbf{H}_0$ along [010]. The angle ψ between **E** and $\mathbf{H}_0$ is zero for the first 25 sec. Then ψ is set at 10° for the next 25 sec (25-50 sec). After this time ψ is rotated to —45° until the torque is reduced to zero. This cycle is repeated with ψ increased in 10° steps.

We have shown[8] that if the sample is now illuminated with polarized light so that the E vector of the light is along [010], the torque remains unchanged. On changing the polarization so that **E** is parallel to [110] the torque increases to about 1.6×10^4 dyne-cm/cm^3 in approximately 200 sec. If the polarization is then rotated parallel to the $[\bar{1}10]$ axis, the torque decreases to about -1.6×10^4 dyne-cm/cm^3. (In Ref. 8 the negative torque is plotted.) With Eq. (6) we find that this photoinduced torque corresponds to $(n_1 - n_2)/n_0 = 0.5$, where we have used $\epsilon = 1$ cm^{-1} from Ref. 11 as the most reliable value of ϵ. These results were obtained with a light intensity of 310 mW/cm^2 and $\delta = 0.03$. We chose to use the value of $\epsilon = 1$ cm^{-1} from Ref. 11 rather than the value $\epsilon = 71$ cm^{-1} obtained from microwave measurements[21] because the experiments performed that obtained the lower value are more closely related to the measurements discussed here than are the rf measurements. It should be noted that the large discrepancy observed in the value of ϵ obtained by the two methods is another indication that the model used to explain the behavior of Fe^{2+} in YIG is not yet complete.

Figure 4 shows similar results for the same sample. Here the sample was cooled to 4.2°K with $\mathbf{H}_0$ along [010]. Then the sample was illuminated with the E vector at 10° to [010]. After 25 sec the plane of polarization of the light was changed to −45° and kept in that direction until the torque was reduced to zero. Subsequently the induced torque was recorded with the angle between **E** and $\mathbf{H}_0$ increased in 10° increments. After 25 sec at each angle the polarization was changed to −45° to return the torque to zero. From Fig. 4 we see that at least for small angles the torque induced in 25 sec is fairly well described by the relationship $\Delta\tau = C \sin 2\psi$ where ψ is the angle between [010] and E and C is a constant of proportionality. As ψ approaches 90° the agreement becomes poorer. In fact, from Fig. 4 we see that the behavior for $\psi = 80°$ is what is expected for $\psi = 90°$.

The observed functional relationship of $\Delta\tau$ on ψ is the same as that found by Teale et al.[22] Using an argument almost identical to the one used by Teale et al.,[6,22] we let the probability of exciting an Fe^{2+} ion on the ith octahedral site be proportional to $\cos^2 \beta_i$, where β_i is the angle between the $\bar{3}$ axis of the ith site and the E vector of the incident light. For the (001) plate with $\mathbf{H}_0$ along [010], from symmetry we assume that the fallback from the excited state is equally divided between the two types of sites. With these assumptions we find that $d(n_1 - n_2)/dt$, as defined for Eq. (6), is given by

$$d(n_1 - n_2)/dt = \frac{3A}{2}\left[\cos^2 \beta_1 - \cos^2 \beta_2\right],$$

where β_1 is the angle between E and the $\bar{3}$ axes that project on [110] and β_2 the analogous angle for the sites that project on $[\bar{1}10]$. A is a constant of proportionality. Writing β_1 and β_2 in terms of ψ leads to

$$d(n_1 - n_2)/dt = A \sin 2\psi, \tag{8}$$

which is the experimentally observed result.

Equation (8) is, however, not valid if $\mathbf{H}_0$ is not along a major crystallographic direction. In a series of experiments we cooled a (001) disc ($\delta = 0.03$) to 4.2°K with $\mathbf{H}_0$ at an angle ϕ_a to the [010] axis. Then we illuminated the sample with E at angle ψ to the [010] direction. For any value of ϕ_a the photoinduced torque was positive for $\psi > \phi_a$ and negative for $\psi < \phi_a$. This unexpected result shows that the transition probability is not determined by the orientation of E with respect to the crystal axes but rather by the orientation of E with respect to $\mathbf{H}_0$. Typical experimental results are shown in Fig. 5, where we plot the torque induced in 50 sec with $\psi = \phi_a + 20°$ and with $\psi = \phi_a - 20°$. The data, as can be seen from Fig. 5, are fairly well described by

$$\Delta\tau = C \sin 2\alpha \cos^2 2\phi_a\,, \tag{9}$$

where $\alpha = \psi - \phi_a$ is the angle between E and $\mathbf{H}_0$. For $\phi_a = 0$, (anneal along [010]), Eq. (9) reduces to Eq. (8) and to the case just discussed. The validity of Eq. (9) has been verified for α as small as 5° for a number of values of ϕ_a. For $\alpha = +5°$ the torque increases and for $\alpha = -5°$ the torque decreases.

The value of the constant C in Eq. (9) has been shown to increase with δ, the Fe^{2+} content. However, with the range of δ available and with the experimental uncertainty illustrated in Fig. 5, it is at present not possible to ascertain if the value of C is linear with δ. In Fig. 6 we show the results for the same experiment, but with the sample cooled to 77°K rather than 4.2°K. The induced torque is a factor of four smaller but the general features observed in Figs. 5 and 6 are very similar.

We conclude our discussion of the effects of polarized light on the induced torque by noting that the earlier model describing the behavior of Fe^{2+} ions must be somewhat modified. The modification only involves making the probability of exciting an Fe^{2+} ion to the higher level dependent on the orientation of **E**. The fact that the pertinent orientation is described by the angle between **E** and $\mathbf{H}_0$ rather than by the angle between **E** and the $\bar{3}$ axes is unexpected but it does not in any sense invalidate the model.

Photoinduced strain

The previous discussion has shown that with polarized light it is possible to preferentially pump Fe^{2+} ions from one type of octahedral site to another. Since the octahedral sites are distorted and since the Fe^{2+} ion is larger than the Fe^{3+} ion, this preferential occupation might be expected to be reflected in the physical size of the crystal. This is indeed the case. As was reported earlier[9], if the strain in a $(1\bar{1}0)$ disc of YIG(Si) is measured as the population difference of the various sites is modified by polarized radiation, the sample elongates or contracts. For $\delta = 0.04$ the total change in length observed $\Delta\ell/\ell = 4 \times 10^{-6}$ at 4.2°K. The photoinduced strain is measured with strain gauges mounted directly on the sample. The size of the gauge requires relatively large samples so that experiments have to be performed with the crystal growth face $(1\bar{1}0)$ rather than the (001) disc that is more convenient from an analytic viewpoint. Specifically, for a gauge on the $(1\bar{1}0)$ plane mounted so as to respond to strains parallel to [111], the crystal expands along this direction when **E** is parallel to [111] and contracts when **E** is parallel to $[11\bar{2}]$ (perpendicular to [111]). The field $\mathbf{H}_0$ is kept along [111]. This implies, if our assignment of the sign of ϵ is correct, that the occupation of an octahedral site by an Fe^{2+} ion expands the site along the $\bar{3}$ direction of that site. It is tempting to interpret this expansion in terms of the shape of the Fe^{2+} wave function. The complications inherent in this interpretation are such that a convincing argument cannot be given at present.

Note that in the experiments described the applied field is kept in a fixed direction. Thus the strains are not a magnetostriction. Rather they represent a lowering of the crystal symmetry by radiation having a specific polarization.

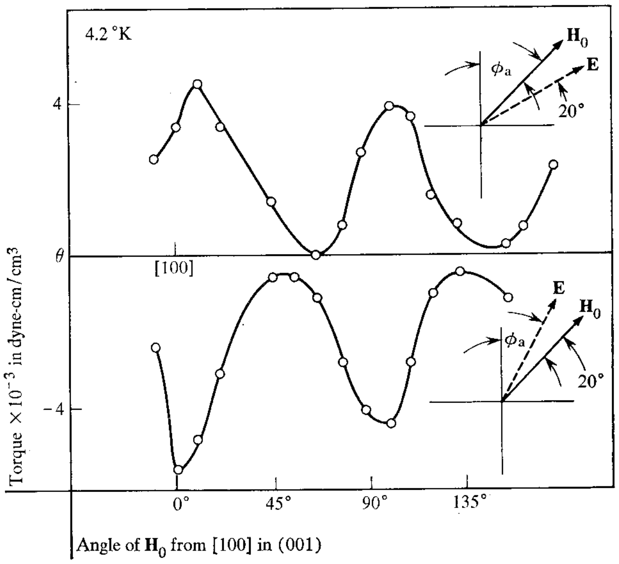
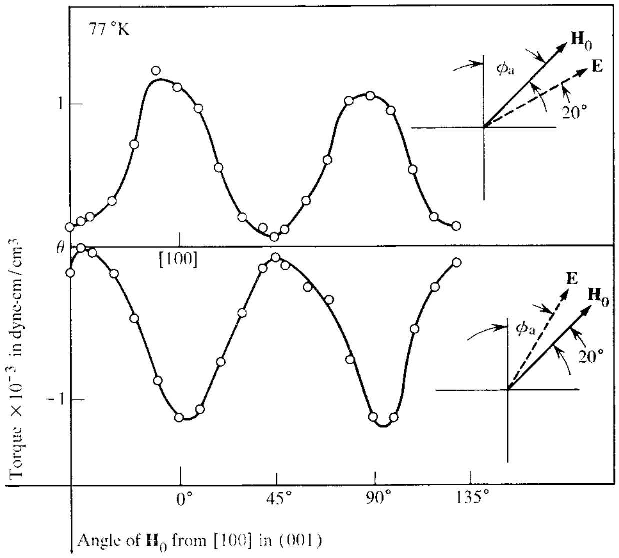


Figure 5 The torque induced in 50 sec by illumination with **E** at ±20° to $\mathbf{H}_0$. The light is turned on after the (001) disc has been cooled to 4.2°K with $\mathbf{H}_0$ at some angle ϕ_a to the [010] axis. The sample is the same as that used for Fig. 4.

Figure 6 Same as Fig. 5 except that the sample was cooled to 77°K rather than 4.2°K.



Photoinduced dichroism

The observed plotoinduced torque implies the presence of a photoinduced dichroism. Again we limit ourselves to the (001) plane with $\mathbf{H}_0$ along [010] and we call the octahedral sites whose $\bar{3}$ axes project on [110] type 1 and those whose axes project on $[\bar{1}10]$ type 2. As before the number of Fe^{2+} ions on type 1 and type 2 sites will be n_1 and n_2 respectively. With **E** along [110] the torque experiments have shown that n_1 increases as n_2 decreases. This observed change in population can occur only if sites of type 2 are preferentially excited with respect to

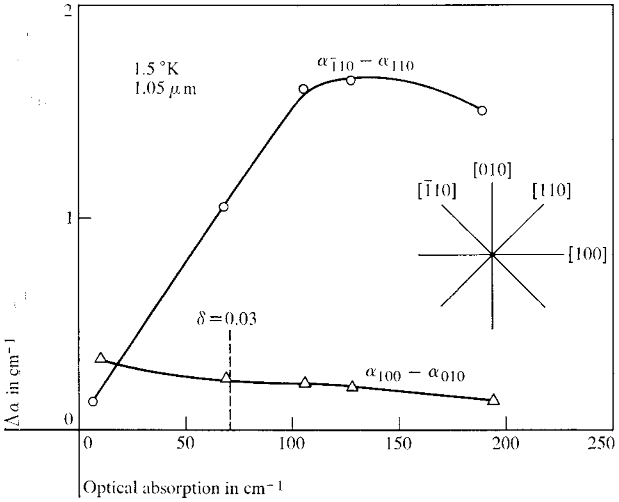


Figure 7 The top curve (circles) is the measured dichroism for **E** along [$\bar{1}$10] and [110] after irradiation with **E** along [110]. The lower curve (triangles) is the dichroism for **E** along [100] and [010] after irradiation with **E** parallel [010]. The data are plotted as a function of the total optical absorption at 1.05μm. The applied field is along [010].

sites of type 1. Here we assume random fallback of the electrons from the excited state, which is certainly valid if $\mathbf{H}_0$ is along [010]. The preceding statements can be rephrased to say that the probability of exciting an Fe^{2+} ion whose $\bar{3}$ axis projection on the (001) plane is perpendicular to E ($P_\perp$) is greater than the probability of exciting an Fe^{2+} ion whose $\bar{3}$ axis projection on the (001) plane is parallel to E ($P_\parallel$).

After the population difference has been established by an intense white light (E parallel to [110]), the optical attenuation constant for E along [110], (α_{110}), and for E along [$\bar{1}$10], ($\alpha_{\bar{1}10}$), can be obtained with weak monochromatic radiation. The constants α refer to the usual relation $I = I_0 e^{-\alpha z}$ where z is the thickness in cm. From our previous definitions we may write

$$\alpha_{\bar{1}10}/\alpha_{110} = \frac{n_1 P_\perp + n_2 P_\parallel}{n_2 P_\perp + n_1 P_\parallel} > 0, \qquad (10)$$

where we have used the relations $n_1 > n_2$ and $P_\perp > P_\parallel$. Equation (10) is, of course, the analytic definition of linear dichroism. To derive Eq. (10) we have tacitly assumed that the only absorption arises from the excitation of the Fe^{2+} ions. To account for losses from other sources a constant should be added to the numerator and denominator of Eq. (10).

Similar experiments have been carried out with the intense light polarized along [010] and the attenuation constant measured for E parallel to [100], (α_{100}) and for E parallel to [010], (α_{010}). If $\mathbf{L}^i$ were along $\langle 111 \rangle$, symmetry would require that $\alpha_{010} = \alpha_{100}$. The observed dichroism for this case indicates that $\mathbf{L}^i$ is not completely along $\langle 111 \rangle$. This result is not unexpected since it has already been shown that the observed F/G ratio obtained from torque[10] and rotational hysteresis measurements[11] are also not completely consistent with pure $\langle 111 \rangle$ symmetry.

Experimental results are shown in Fig. 7 where we plot $\alpha_{\bar{1}10} - \alpha_{110}$ (measured after irradiation with E along [110]) and $\alpha_{100} - \alpha_{010}$ (measured after irradiation with E along [010]) as a function of the total absorption at 1.05 μm, $\alpha_{1.05}$. The Fe^{2+} content, as discussed earlier, is proportional to $\alpha_{1.05}$; $\alpha_{1.05} = 70$ cm^{-1} corresponds to $\delta = 0.03$. The measurements of the dichroism were made at 1.5°K with 1.05 μm radiation. The measured dichroism peaks quite sharply at about 1 μm. The predominant term, $\alpha_{\bar{1}10} - \alpha_{110}$ is seen to increase linearly with δ for δ less than about 0.05 and then appears to be almost independent of concentration. The smaller dichroism ($\alpha_{100} - \alpha_{010}$) decreases slowly with Si content.

From the torque experiments we know that the population difference ($n_1 - n_2$) is fairly large. This large population difference requires that $P_\perp$ be substantially larger than $P_\parallel$. (For **E** along [110] and by neglecting the small thermal redistribution, the steady state during irradiation is given by $n_1 P_\parallel = n_2 P_\perp$). The one-to-two percent dichroism experimentally observed then shows that Eq. (10) does not account for all the losses and that the constant required in both the numerator and demoninator is actually the predominant term.

The various measurements of dichroism just discussed show that our model of YIG(Si) can explain the main features but that many important details are not accounted for. For example, the nonlinear behavior of $\alpha_{\bar{1}10} - \alpha_{110}$ at high concentrations and the strange behavior of $\alpha_{100} - \alpha_{010}$ as a function of Fe^{2+} content are not at all explained by the model. In addition the model cannot predict the relative size of $P_\perp$ and $P_\parallel$.

Conclusion

In this paper we have demonstrated that irradiation of YIG(Si) by polarized light results in a new class of magneto-optic effects in which the magnetic symmetry of the material can be changed in a controlled way. The explanation of these effects is based on a model of Fe^{2+} ions preferentially occupying some of the octahedral sites of the crystal. This model was previously used to explain thermal annealing effects and microwave losses. The study of the photoinduced effects has led to substantially better understanding of the details and limitations of this model. A number of major points are still not understood. The most important defect of the model is that it does not explain the modifications of the coercive force and of the permeability upon irradiation. The theory[4] that explains these observations does not readily fit the data presented here for the effects on the uniaxial anisotropy, strain and linear dichroism. Perhaps the next most important question remaining is the question of the existence of two mechanisms that appear to lead to an induced anisotropy. It is hoped that further experiments on photoinduced effects will help clarify these points.

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