PHOTOINDUCED MAGNETIC ANISOTROPY AND OPTICAL DICHROISM IN SILICON-DOPED YTTRIUM IRON GARNET

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It has been found that an intense beam of linearly polarized light can induce a magnetic anisotropy in silicon-doped yttrium iron garnet at liquid-helium temperatures. Furthermore, there is associated with this anisotropy a substantial linear dichroism.

It has been known for some time that the addition of silicon to yttrium iron garnet (YIG) makes it possible to induce a low-temperature magnetic anisotropy by cooling in a field.[1,2] A low-temperature rotational hysteresis is also observed.[3,4] Recently the effect of infrared radiation on this induced anisotropy has been investigated. Teale and Temple[5] have studied the field for ferromagnetic resonance and have shown that the decay of the induced anisotropy is greatly enhanced by infrared radiation. Enz and van der Heide reported that similar radiation markedly reduced the permeability of single-crystal and polycrystal cores of YIG(Si).[6] Comparable results were reported by Lems et al.[7] for $CdCr_2Se_4$(Ga). Using a torque magnetometer, Pearson, Annis, and Kompfner[8] have seen that the details of the decay of the induced anisotropy depend on the polarization of the infrared radiation. In this Letter we report experiments which demonstrate that a magnetic anisotropy may be isothermally induced in YIG(Si) by the influence of polarized light alone. In addition we have observed that polarized light induces a substantial linear dichroism as well as the anisotropy.

Since it has previously been shown that the anisotropy induced on cooling in a field is primarily along the ⟨111⟩ directions,[4] we have chosen the (001) plane for our experiments. In this plane the projection of the ⟨111⟩ directions are orthogonal and lie along [110] and [$1\bar{1}0$]. For both measurements of the torque and of the dichroism the sample was a polished (001) plate 0.05 cm thick. The crystal was grown from a PbO-B_2O_3 flux using ultrahigh-purity starting materials. A measure of the silicon content [0.028 atoms per $Y_3Fe_5O_{12}$ formula] was obtained from the value of the optical absorption at 1.20 μ.[9]

To investigate the induced anisotropy the sample was mounted in a recording torque magnetometer. The Dewar containing the sample had flat-bottom windows so that the sample could be illuminated from below. The use of a flat sample and of flat-bottom windows in the Dewar allowed uniform illumination of the sample and minimized depolarization of the light. The light source was a tungsten lamp whose radiation was focused on the plane of the sample after passing through a Glan-Thompson polarizer. The axis of polarization could be rotated in the (001) plane of the crystal. At the sample, the energy density of light was 310 mV/cm².

To observe the isothermally photoinduced anisotropy the sample was cooled to 4.2°K with an applied field of 4 kOe along the [100] direction. At 4.2°K the observed torque with $\vec{H}$ along [100] was zero since there is a maximum in the magnetocrystalline anisotropy energy surface for the field parallel to [100]. The sample was then illuminated for 50 sec with light whose $\vec{E}$ vector lay along [100] (Fig. 1). During this period the torque remained zero. On changing the polarization so that the $\vec{E}$ vector was parallel to [110], the torque increased exponentially to a value of about 1.44 $\times 10^4$ dyn cm/cm³ after 100 sec. At this time, as shown in Fig. 1, the polarization was rotated to

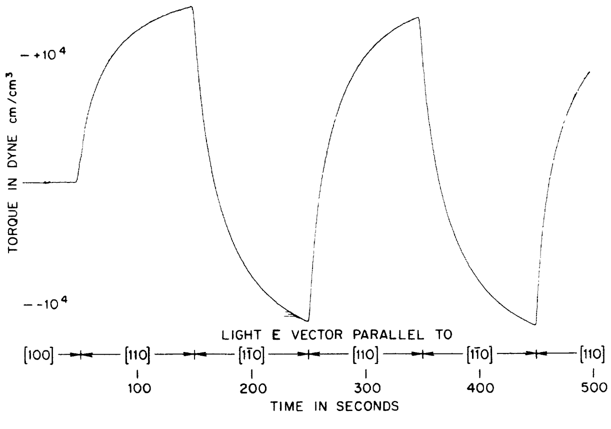

FIG. 1. The photoinduced torque at 4.2°K for a (001) plate of YIG (0.028 Si). The applied field is along the [100] direction. For time t, $0 \leqslant t \leqslant 50$ sec, the polarization of the light vector $\vec{E}$ is along [100], and the torque remains at zero. For $50 \leqslant t \leqslant 150$ sec $\vec{E}$ is parallel to [110], and for $150 \leqslant t \leqslant 250$ sec $\vec{E}$ is parallel to [$1\bar{1}0$]. This cyclic variation of the $\vec{E}$ vector is then continued.

make it parallel to $[1\bar{1}0]$. The torque then decreased until after 100 sec it reached -1.13×10^4 dyn cm/cm^3. As is shown in Fig. 1, cyclic rotations of the axis of polarization between [110] and $[1\bar{1}0]$ lead to additional cyclic changes in the torque. It should be emphasized that during the whole time the torque was being measured, the applied field was constant along the [100] direction.

If the polarizer is not rotated and $\vec{E}$ is left along [110], the torque asymptotically approaches 1.6×10^4 dyn cm/cm^3. This value corresponds to an effective field of 80 Oe.

The same YIG(Si) sample was used for the dichroism measurements. The sample, immersed in liquid helium at 1.5°K and mounted in a transmission optical system with a field of 8.0 kOe along [100], was illuminated with substantially unpolarized infrared light from a monochromator. If the sample shows linear dichroism, one linear polarization is absorbed more strongly than the orthogonal polarization, and the transmitted light is in part linearly polarized. The light transmitted by the crystal passes through a Glan-Thompson prism rotating at 53.3 Hz and is then focused on a PbS detector. With this arrangement the linearly polarized component of the transmitted light produces a sinusoidal variation at 106.6 Hz in the detected light intensity. Using a lock-in detector, this signal was plotted on the y axis of a y-time recorder. Comparison with the dc signal made it possible to scale the plot of $(I_{110}-I_{1\bar{1}0})/I$ in percent.

In addition to the transmission optical system just described, the sample could be irradiated with an intense beam of linearly polarized tungsten light (energy density 130 mW/cm^2). Because it was necessary to mask the detector during this irradiation the signal on the recorder was zero.

The results of a typical experiment are shown in Fig. 2. The sample was first irradiated with the $\vec{E}$ vector along [110].

After about 90 sec of this intense polarized illumination, the fractional polarization at 1.20 μ approached 2%. After irradiation with light polarized parallel to $[1\bar{1}0]$, the polarization exceeded -2%. Irradiation with light polarized parallel to either [100] or [010] leads to a fractional polarization of -0.45%. This corresponds to the slight residual polarization of the original beam. It is seen in Fig. 2 that irradiation by linearly polarized light gives rise to a substantial dichroism. On comparison with the torque experiment above, it is clear that this dichroism is to be associated with the magnetic anisotropy. This contrasts to the usual magneto-optical effects which are associated with the magnetization.

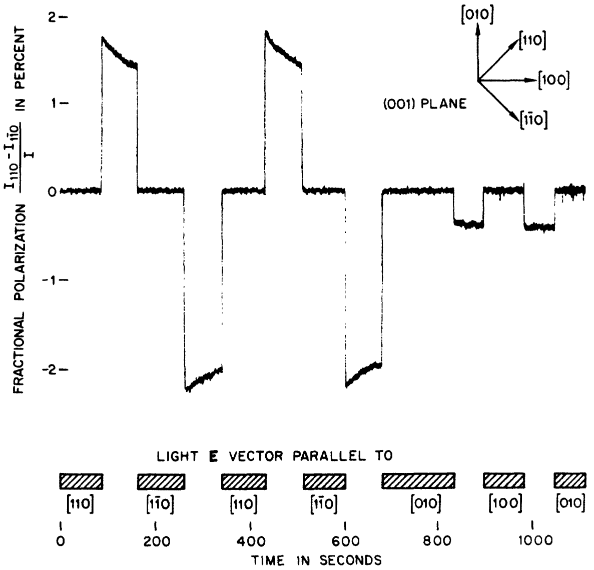

FIG. 2. Dichroism of YIG(Si) after pumping by polarized light. The plot shows the partial polarization for 1.20-μ light transmitted by a 0.05-cm thick (001) plate of YIG (0.028 Si) at 1.5°K after irradiation by an intense white light of the specified linear polarizations. An applied field of 8.0 kOe was along [100]. The PbS detector was masked during irradiation and thus the signal was zero. At 1.2 μ, $\alpha=43$ cm^{-1}, and the dichroism $\alpha_{110}-\alpha_{1\bar{1}0}$ corresponding to a 4% change in the fractional polarization is $\Delta\alpha=0.8$ cm^{-1} [where $I=I_0\times\exp(-\alpha t)$].

The preceding observations of the photoinduced anisotropy and dichroism can be interpreted in terms of the model used to explain the anisotropy induced by cooling in an applied field.[4] In this model, the Fe^{++} ions preferentially occupy the octahedral sites whose trigonal axes are nearest to the magnetization vector $\vec{M}$. The trigonal axes of the four types of octahedral sites are coincident with the $\langle111\rangle$ axes. The anisotropy energy of an Fe^{++} ion may be written $W_j=-\epsilon\cos^2\theta_j$, where θ_j is the angle between the appropriate $\langle111\rangle$ axis and $\vec{M}$. The constant ϵ is positive, so the trigonal axis of a site is the easy axis of the Fe^{++} ion on that site. We follow Pearson et al.[7] and assume that the probability of ionizing an Fe^{++} ion is a function of the angle between $\vec{E}$ and the local trigonal axis. The excited electrons are assumed to fall back at random into the four types of sites.

For the (001) plane, only two sites need be considered: those whose trigonal axes project on [110] (type 1) and those whose trigonal axes project on [1$\bar{1}$0] (type 2). We let n_1 and n_2 be the number of Fe^{++} ions per unit volume on sites of types 1 and 2, respectively. After cooling with the applied field $\vec{H}$ along [100], $n_1 = n_2$ and the induced torque is zero. Radiation with $\vec{E}$ parallel to [110] increases n_2 at the expense of n_1, and [1$\bar{1}$0] becomes the energetically preferred axis. As a result, for $\vec{H}$ along [100], the torque increases. In addition, after this irradiation $n_2 > n_1$; so the optical absorption for $\vec{E}$ parallel to [1$\bar{1}$0] is greater than for $\vec{E}$ parallel to [110].

Irradiation with $\vec{E}$ along [1$\bar{1}$0] reverses the above situation both with respect to torque and with respect to dichroism.

The decay of the dichroism seen in Fig. 2 may be ascribed partly to photoinduced and partly to a thermally activated rearrangement of Fe^{++} ions on octahedral sites. In this connection it may also be remarked that in the absence of light there is a slow spontaneous decay of the induced anisotropy.

From the maximum torque induced (1.60×10^4 dyn cm/cm^3) we calculate that $(n_2-n_1)/n_0 = 0.1$ or 0.5, where n_0 is the total number of Fe^{++} ions per cm^3. The number we obtain depends on whether we use $\epsilon = 5$ cm^{-1} from Ref. 4 or $\epsilon = 1$ cm^{-1} from Ref. 3.

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DIELECTRIC THEORY OF COHESIVE ENERGIES OF TETRAHEDRALLY COORDINATED CRYSTALS

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Pauling's resonating-bond theory is reformulated in terms of an itinerant dielectric model. In extreme cases discrepancies of more than 200 kcal/mole between the observed cohesive energy and Pauling's value are reduced to 1 kcal/mole. The results completely justify the concept of resonating covalent and ionic contributions to cohesive energies of tetrahedrally coordinated crystals.

The most general theory of the cohesive energies of covalently bonded crystals and molecules is that of Pauling.[1] His basic idea is that the energy of the covalent bond contains two resonating components, a homopolar part and a heteropolar, or ionic, part. The basic parameters in Pauling's theory are derived from his table of elemental electronegativities. In the early work in the 1930's Pauling found it necessary to obtain electronegativities from heterogeneous sources. Bellugne and Daudel[2] obtained more consistent results using saturated molecules, while retaining Pauling's key assumptions (based on the now defunct Heitler-London model) of additivity for both electronegativity differences and bond energies. In this note, however, a basic restructuring of Pauling's theory is proposed in terms of itinerant dielectric theory. Dramatic improvements in the prediction of cohesive energies of covalent crystals are obtained. In extreme cases the error of more than 200 kcal/mole resulting from the Pauling formulation is reduced to 1 kcal/mole by the new theory.

The resonating character of covalent and ionic components of bond energy, an ad hoc element of Pauling's theory, appears naturally in the microscopic dielectric formulation.[3] For each covalent crystal one defines an average energy gap E_g which is related to the electronic dielectric constant ϵ_0 by[3]

$$\epsilon_0 = 1 + (\hbar\omega_p/E_g)^2 A, \tag{1}$$