PHOTOMAGNETIC ANNEAL, A NEW MAGNETO-OPTIC EFFECT, IN Si-DOPED YTTRIUM IRON GARNET

R. W. Teale and D. W. Temple
Physics Department, The University, Sheffield, Yorkshire, England
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Experimental evidence is given to show that the magnetocrystalline anistropy of silicon-doped yttrium iron garnet, nominal composition $Y_3Fe_{4.9}Si_{0.1}O_{12}$, can be changed by more than 100 Oe at 20°K through irradiation with near-infrared photons. The change is interpreted as due to the redistribution of electrons between iron ions, that is valence exchange.

The purpose of this Letter is to report that magnetic anneal can be substantially speeded up, in silicon-doped yttrium iron garnet (YIG), if the specimen is exposed to infrared radiation.

If the magnetocrystalline anisotropy of a ferromagnetic or ferrimagnetic sample is influenced by the past crystallographic orientation of the intrinsic magnetization, M_0, this phenomenon is known as magnetic anneal. Usually, but not necessarily, the sample is cooled with M_0 in a given orientation and the term magnetic anneal is normally reserved for cases where the "memory" persists over minutes or hours. Recently interest has revived[1] in this effect for ferrites which contain both Fe^{2+} and Fe^{3+} ions. Si-doped YIG is such a material since the silicon, which is tetravalent, replaces some Fe^{3+} and, for charge neutrality, gives rise to Fe^{2+}.

It is fairly well established[1] that the effects associated with Fe^{2+} and Fe^{3+} result from the migration of the extra electrons between iron ions, the occupancy of some iron ions being preferred, on energetic grounds, for a given crystallographic orientation of M_0. When this orientation is changed the change to the different preferred occupancy takes minutes or longer at low temperatures. The magnetocrystalline anisotropy and associated anisotropy field are influenced by the electron distribution and hence exhibit memory of the past orientation of M_0 and relax over long periods.

Since the applied field for ferromagnetic resonance depends, in part, upon the anisotropy field it is influenced by magnetic anneal. If a small spherical specimen is cooled slowly from room temperature to temperature T with M_0 along the [111] direction the site occupancy approximates to that appropriate to thermal equilibrium; we denote the measured resonance field by H_1. If now M_0 is rotated to $[11\bar{1}]$ the site occupancy no longer corresponds to thermal equilibrium and the resonance field at the same microwave frequency and temperature is different, denoted H_2. Curve A of Fig. 1 shows H_2 as a function of time with $T = 20°K$ for a crystal of nominal composition $Y_3Fe_{4.8}^{3+}-(SiFe^{2+})_{0.1}O_{12}$; the microwave frequency is close to 9400 Mc/sec. The time axis starts when M_0 was rapidly (in ~5 sec) rotated from [111] to $[11\bar{1}]$. The sample was enclosed in a cavity with only the normal coupling holes to the closed waveguide system. H_2 relaxes towards H_1, which was 2531 G, but after falling about one-third of the way becomes steady at a metastable value. Curve B shows H_2 as a function of time after the sample had been prepared by cooling and rotation in the same manner, but in this case the specimen was mounted on the end of a crude light pipe with one end outside the cryostat; the cavity was unchanged. Up to the point marked on the curve only the radiation in the room was incident upon the polythene window covering the room-

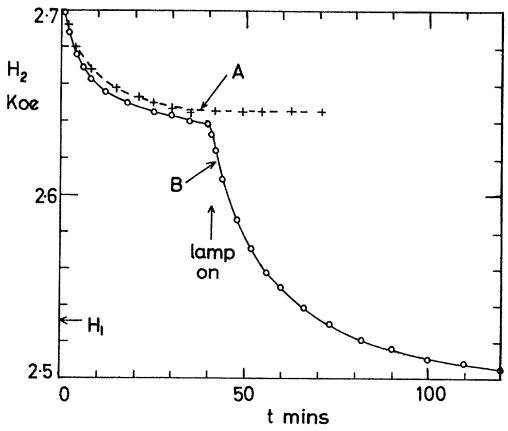

FIG. 1. Field for ferromagnetic resonance, H_2, plotted against time t, after cooling specimen with M_0 along [111] from 300 to 20°K then rotating M_0 to $[11\bar{1}]$. Zero time is when rotation was performed. Frequency 9400 Mc/sec. Specimen $Y_3Fe_{4.9}Si_{0.1}O_{12}$. Curve A, without photon irradiation; Curve B, irradiation switched on after 40.5 min.

temperature end of the light pipe; at the point indicated a 50-W tungsten iodine lamp mounted 10 cm from the end of the light pipe was switched on. H_2 clearly falls in response to the lamp and tends to a value which is somewhat less than H_1. This photomagnetic anneal was also observed at 4.2 and 66°K; the changes of H_2 due to photon irradiation were 200 and 21 G, respectively, at these temperatures.

Our present knowledge of the dependence of the photomagnetic anneal upon the radiation wavelength and power is limited. The lamp envelope transmits radiation in the visible and out to ~5 μ. YIG is opaque in the visible and transparent between about 1 and 5 μ, though silicon doping reduces this transparency.[2] The sample was a sphere of ~0.3 mm diam. Filtration experiments were performed with a glass block (transparent out to ~2.2 μ) and with a 2.5% solution of $CuCl_2$ in water (transmitting the visible and cutting off radiation with $\lambda > 0.8$ μ); the former filter left the photon effect unchanged but the latter eliminated it completely. It seems that the active radiation lies in the range $2.2\ \mu > \lambda > 0.8\ \mu$, where Si-doped YIG shows increased absorption compared with pure YIG. A very rough estimate of the light-pipe efficiency suggests that ~10 μW of the active radiation may reach the sample. Dr. R. F. Pearson and Mr. D. Annis of Mullard Research Laboratories confirmed the photon effect using a torque magnetometer, finding that with very roughly 1 mW incident on the sample the relaxation due to photons occurred in 1 sec or less. The immersion of the sample in liquid helium eliminated the possibility that the effect might arise from heating of the sample by the radiation.

A qualitative interpretation can readily be given. After cooling the sample to 20°K and rotation of M_0, some of the excess electrons redistribute between the available iron ions in response to thermal vibrations, hence causing the initial decay of H_2 in Fig. 1. Other electrons are trapped on sites from which their escape is very slow; hence H_2 remains, in Curve A, at its metastable value. When radiation is applied, the absorption of a photon gives a trapped electron sufficient energy to raise it to a much higher state from which it may fall very rapidly to a different iron site. This process allows thermal equilibrium between the sites and the much higher states, hence the site occupancy thermalizes and H_2 relaxes to its equilibrium value, which is somewhat lower than H_1.

The effect should be of use in the study of magnetic anneal where the ability to thermalize the site occupancy rapidly and at will should simplify measurements. Further irradiation experiments might improve knowledge of the mechanism of electron migration and yield further links with the electron transport properties. Clearly a radiation detector with a long built-in integration time could be constructed on the basis of the effect reported and the integration gives rise to the possible use as a store. Since the time for response of the magnetocrystalline energy to irradiation may be limited by the relaxation time of the electrons from the high excited state to the traps this could be very fast. Attempts to investigate this response time using flash bulbs indicated only that it was less than ~0.02 sec. Since magnetic domain-wall mobility, the B-H loop shape, and magnetic permeability are known to depend upon the relaxation mechanism whose speed the photons can control, it should be possible to modify these phenomena through photon irradiation.

The specimen was grown from a flux by Mr. J. Page of Mullard Research Laboratories and loaned to us; chemical analysis indicates that only about half the silicon concentration implied by the nominal composition is actually found in the crystals.

[1]R. P. Hunt, J. Appl. Phys. 37, 1330 (1966); A. Broes Van Goenou, J. L. Page, and R. F. Pearson, J. Phys. Chem. Solids 28, 1017 (1967); D. I. Tchernev, J. Appl. Phys. 37, 1318 (1966).

[2]R. C. LeCraw, D. L. Wood, J. F. Dillon, Jr., and J. P. Remeika, Appl. Phys. Letters 7, 27 (1965).