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PHOTOMAGNETIC ANNEAL PROPERTIES OF SILICON-DOPED YTTRIUM IRON GARNET

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Torque measurements have been made at 4.2°K on crystals of silicon-doped yttrium iron garnet which were irradiated with infrared light. The radiation caused large photomagnetic anneal effects which were found to depend on the plane of polarization of the light.

The effect of infrared radiation on the magnetic properties of silicon-doped yttrium iron garnet has been reported by Teale and Temple,[1] and Enz and van der Heide.[2] Measurements of ferrimagnetic resonance, initial permeability, and hysteresis loop shape were made. The large effects that they observed were termed photomagnetic anneal and interpreted as a light-induced redistribution of Fe^{2+} ions in the crystals. From torque curves on single crystals we have directly measured the induced anisotropy in these samples and confirmed that it can be altered considerably by photon irradiation. Changes in anisotropy up to 100×10^3 erg/cm^{-3} were observed in a sample of nominal composition $Y_3Si_{0.3}Fe_{4.4}{}^{3+}Fe_{0.3}{}^{2+}O_{12}$.

The experimental arrangement is shown in Fig 1. The sample, a sphere about 1.5 mm diam, was mounted in a conventional torque magnetometer[3] such that the applied magnetic field could be rotated in a (110) plane. Infrared radiation from a 2.5-V, $\frac{1}{2}$-W tungsten lamp inside the cryostat was polarized by a sheet of infrared Polaroid and produced approximately 10^{-3} W at the sample. The plane of polarization could be rotated through all directions in the (110) plane.

Measurements were made on crystals of nominal composition $Y_3Si_{0.1}Fe_{4.9}O_{12}$ and $Y_3Si_{0.3}Fe_{4.7}O_{12}$. At 4.2°K rotational hysteresis and time-dependent torque curves were obtained. The mean torque curves in applied fields of 15 kG were analyzed into harmonic components $\sin2\varphi$, $\cos2\varphi$, $\sin4\varphi$, $\cos4\varphi$, etc., where φ is the angle between the ap-

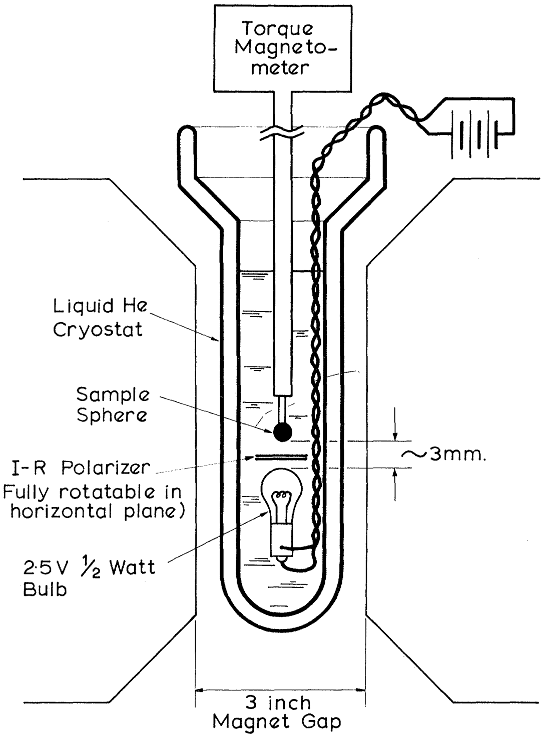

FIG. 1. Experimental arrangement.

plied field and the [100] direction in the crystal. These materials show induced anisotropy effects at low temperatures.[4,5] This is shown, for example, by the appearance of a large $\cos 2\varphi$ component in the torque curve on cooling with a field applied along a [111] direction. When the sample was illuminated for a period of 4 min with the magnetic field applied along the [111] direction, the subsequent torque curve showed a change in the $\cos 2\varphi$ component. Further irradiation in each case for 4 min with the field at an angle Φ to the [100] produced further changes in this component. The amplitude of the $\cos 2\varphi$ component is plotted as a function of Φ in Fig. 2. When the incident light was polarized, the results were found to depend on the direction of polarization relative to the crystal axes, and values of the $\cos 2\varphi$ and $\sin 2\varphi$ amplitudes as a function of Φ are shown in Fig. 2 for the light polarized parallel and perpendicular to a [111] direction. Similar, but larger, effects were observed in the $Si_{0.3}$-substituted crystal.

Teale[6] has pointed out that it is possible to explain these results by assuming that the probability of photon-induced excitation of an electron from an Fe^{2+} ion in an octahedral site will depend on orientation of the light polarization and the applied field direction during irradiation. Neglect-

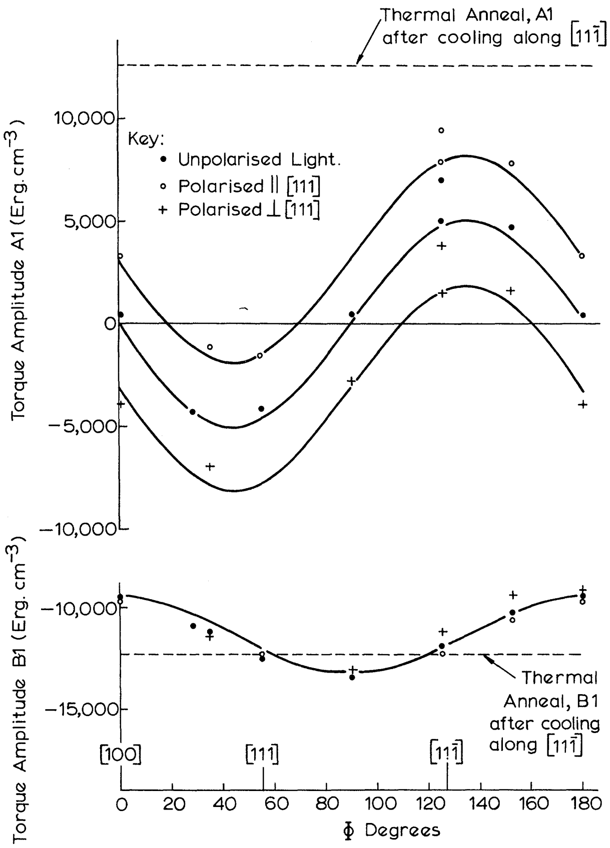

FIG. 2. Experimental values of the coefficients A_1 and B_1 of the $\sin 2\varphi$ and $\cos 2\varphi$ components of the torque curves measured in the (110) plane at 4.2°K on a crystal of $Si_{0.034}Y_3Fe_{4.966}O_{12}$ after irradiation at this temperature. The Fourier coefficients A_1 and B_1 are plotted against the angle Φ at which a saturating field was applied during irradiation. The three curves of A_1 represent separate experiments in which the radiation was unpolarized, polarized parallel, and polarized perpendicular to [111] direction.

ing any thermal redistribution effects and assuming that the excited electrons fall back randomly into the four possible types of octahedral site, then the equilibrium number of Fe^{2+} after exposure to radiation is given by

$$N_j = N\left\{W_j\left(\sum \frac{1}{W_j}\right)\right\}^{-1},$$

where N is the total number of Fe^{2+} ions and W_j is the probability of excitation from site j. Assuming a very simple form of W_j, such as

$$\frac{1}{W_j} \propto 1 + a\cos^2\alpha_j + b\cos^2\beta_j,$$

where α_j, β_j are the angles made by the polarization direction of the incident light and magnetization direction, respectively, with the local trigonal axis of the site j, and assuming that the anisotropy energy for an Fe^{2+} ion can be expressed as

$$\epsilon_j = W \cos^2\beta_j,$$

then the torque (that is, the derivative of the anisotropy energy) is given by

$$L = A_1 \sin 2\varphi + B_1 \cos 2\varphi + \text{higher terms},$$

where φ is the angle between the magnetization and the [100] direction during the torque measurement, and

$$A_1 = \frac{2WN}{9C}(b \sin 2\theta + c \sin 2\Phi),$$

$$B_1 = \frac{WN}{18C}(b + c - b \cos 2\theta - c \cos 2\Phi),$$

where θ, Φ are angles between the polarization direction and the applied field during irradiation. Thus, if we keep the polarization direction θ fixed, then A_1 will be proportional to $\sin 2\Phi$, while B_1 is proportional to $\cos 2\Phi$. This is confirmed by the results in Fig. 2.

Further experimental results not indicated in the graph showed a slight dependence of the photomagnetic effect on the direction of the applied field during cooling to 4.2°K. This would indicate that some thermally induced anisotropy remains in the crystal even after prolonged irradiation.

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SPIN-OPTICAL-PHONON INTERACTION IN ANTIFERROMAGNETIC CoF_2

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Spin-optical-phonon interaction is observed in CoF_2 by the transfer of magnetic dipole intensity to the otherwise optically inactive E_g phonons. Theory predicts that the temperature dependence of the intensity is caused by the change in longitudinal spin correlation, and the change in energy and thermal population of the magnetic excitations. Experimentally, only the latter are important in CoF_2.

Unquenched orbital motion in the single-ion ground state can significantly alter the magnetic properties of the concentrated salt. Off-diagonal exchange coupled with the spin-orbit interaction produces multipole spin-spin interactions.[1-3] Purely electrostatic interactions between charge distributions can also lead to complex effective spin interactions.[4,5] Equally important is the presence of spin-lattice coupling which produces a large single-ion anisotropy field and is expected to couple magnetic excitations to phonons. The virtual emission and reabsorption of phonons will also generate multipole spin-spin interactions in suitable systems.[6-9] Since the above spin-spin interactions often have the same phenomenological form, it is difficult to extract unambiguously from measured spin-wave energies or ion-pair energy levels the various contributions to the ion-pair interactions and it is desirable to perform other measurements which are more sensitive to a particular interaction.

In the present Letter we discuss an experiment which directly measures the interaction between magnetic excitations and the phonons in the antiferromagnet, CoF_2, by observing the transfer of magnetic-dipole intensity to the otherwise optically inactive, $q = 0$, E_g phonon.[10] Since the experiment measures changes in the phonon state, it is sensitive to spin-phonon coupling but relatively insensitive to the complicated exchange interactions between Co^{2+} ions. The observed spin-phonon interaction is sensitive, however, to the gross changes in magnetic state produced by ordering and thermal population of the magnetic excitations, and it is the manifestation of these effects in the spin-phonon coupling that is discussed in the following.

Extrapolation of the E_g phonon frequency in