PHOTOMAGNETIC EFFECT IN A CHALCOGENIDE SPINEL

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Photomagnetic change in the initial permeability has been investigated in Ga-doped $CdCr_2Se_4$. A simple model is shown to account satisfactorily for the experimental data.

Direct influence of electromagnetic (infrared) radiation on magnetic properties was recently discovered in Si-doped yttrium iron garnet (YIG) by Teale and Temple,[1] who observed an appreciable change in the ferromagnetic resonance field. Subsequently, Enz and van der Heide[2] reported large changes in the initial permeability and the coercive force. We wish to present similar results observed in a rather different material, the chalcogenide spinel $CdCr_2Se_4$ which is a semiconductor and ferromagnetic below $T_C = 130°K$.[3] An n-type conducting single-crystal sample, of composition $Cd_{1-x}Ga_xCr_2Se_4$ with $x = 0.015$, was cut into a ring of approximately 1-mm diameter, the axis of the ring being a $\langle 111\rangle$ direction. The initial permeability μ was measured by recording the output in response to a low-field 10-kHz input current.

The behavior of μ with irradiation is shown in Fig. 1(a). After cooling in the dark to 77°K, we observed an initial permeability $\mu_d \approx 320$. Upon illumination with a "white" thermal light source (intensity 10^{-2} W/cm^2 at the surface of the sample for the curve shown) μ decreased steeply to a much lower stationary value $\mu_S \approx 160$. After some time the light was switched off, and μ returned comparatively slowly to its original level μ_d. In the same figure we show a similar plot recorded at 4.2°K: Qualitatively the behavior is similar, but the recovery process is several orders of magnitude slower than at 77°K. If the recovery process is thermally activated, a rough estimate of the activation energy involved gives 0.005 eV.

Both the stationary value μ_S, reached after irradiating for some time, and the rate of change of μ when the light is switched on were found to depend on the light intensity I_w. In Fig. 1(b)—and likewise in the following discussions—we consider the change in the "stiffness" μ^{-1} instead of that in μ, since various properties like anisotropy and magnetostriction contribute additively to the stiffness. The light-induced change $(\Delta\mu^{-1})_S = \mu_S^{-1} - \mu_d^{-1}$ is given as a function of I_w, by the experimental points (dots) in Fig. 1(b). For small intensities it increases sharply with I_w, leveling off for values of I_w larger than 5×10^{-4} W/cm^2. Saturation is reached for I_w roughly 10^{-2} W/cm^2. (The light sensitivity of $CdCr_2Se_4$ is several orders of magnitude higher than that of Si-doped YIG.[2])

A preliminary investigation of the spectral dependence of the effect indicates that only photons of wavelength less than approximately 1.1 μm are active; this roughly coincides with the normal absorption edge of $CdCr_2Se_4$ which at 77°K is found[4] at 1 μm. We also measured the permeability, demagnetizing the sample after the low μ level was reached, or applying a strong

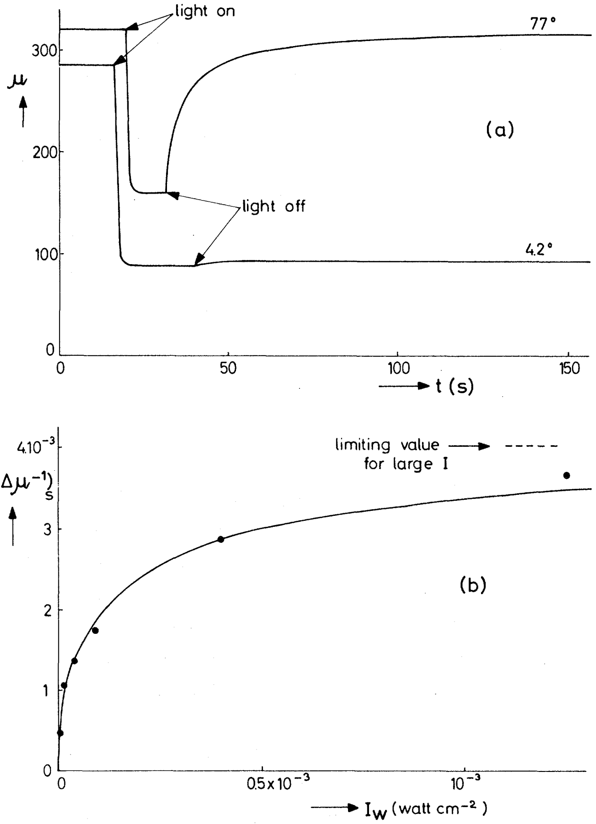

FIG. 1. Experimental results. (a) Initial permeability μ as function of time t for $T = 77$ and 4.2°K. Light intensity 10^{-2} W/cm^2. (b) Stationary change in stiffness $(\Delta\mu^{-1})_S$ versus light intensity I_w at 77°K. Dots: experimental points; drawn curve (calculated): $8\times10^{-3}/[1+(1+8.84\times10^{-4}/I_w)^{1/2}]$.

dc bias field during illumination so that no Bloch walls could be present. Essentially the same change in μ was recorded as in the previous experiments. This shows that the hypothesis of changes local to domain walls must be rejected and strongly suggests that there is a change in permeability throughout the material.

To describe the results obtained we adopt a model similar to that proposed by Enz[2] for Si-doped YIG. There one assumes that a localized Fe^{2+} ion can be in a crystal site adjacent to a Si^{4+} ion (type I) or in a remote position (type II), the contribution of the Fe^{2+} to the magnetic properties being different for the two types of site. Since the energy of electrons in type-I positions is lower, nearly all electrons (and thus Fe^{2+} ions) are at type-I sites after cooling in the dark. The effect is then ascribed to a light-induced electron transfer, causing Fe^{2+} to move from type-I to type-II sites.

For the case of $CdCr_2Se_4$ we will also employ a two-center model: Type I is a more stable center, from which a less stable type-II center can be formed by electron transfer. Centers of type I could be filled Ga donors consisting of a Ga^{3+} ion associated with a more or less localized Cr^{2+} ion. The nature of type-II centers is not known.

In order to calculate the effect of irradiation quantitatively, two assumptions have to be made: (i) The change in stiffness $\Delta\mu^{-1}=\mu^{-1}-\mu_d^{-1}$ is proportional to the density n of type-II centers formed (which is equal to the density of dissociated type-I centers):

$$\Delta\mu^{-1}=Cn. \tag{1}$$

(ii) During and after illumination, electrons will be transferred back from type-II to empty type-I centers by thermal activation, thus diminishing n; the assumption of random recombination leads to a recombination rate proportional to n^2. Both assumptions seem to be justified as long as the density n_0 of Ga atoms is small, which is the case for the crystal under consideration.

As the rate of dissociation of the Ga-donor centers is proportional to the photon intensity and to the density (n_0-n) of remaining type-I centers, the rate equation for n becomes

$$dn/dt=-\alpha n^2+\beta I(n_0-n). \tag{2}$$

Here I is the intensity of photons active in the dissociation process, and α and β are proportionality constants; for a thermally activated recombination with activation energy G, α will be proportional to $\exp(-G/kT)$. If at time $t=0$ the light is switched on and $n=0$ for $t<0$, then n (and thus $\Delta\mu^{-1}$) increases as

$$n=2n_0/[1+(1+4\alpha n_0/\beta I)^{1/2}\times\coth\{\tfrac{1}{2}\beta I(1+4\alpha n_0/\beta I)^{1/2}t\}]. \tag{3}$$

After a long enough time n reaches a stationary value

$$n_s=2n_0[1+(1+4\alpha n_0/\beta I)^{1/2}]^{-1}, \tag{4}$$

showing that $n_s\approx n_0$ if $4\alpha n_0/\beta I\ll 1$, i.e., for high light intensity or low recombination rate (as at very low temperature). Now if the light is switched off at $t'=0$, say, and $n=n_s$ for $t'<0$, we obtain

$$n=n_s/(1+\alpha n_s t'). \tag{5}$$

Let us examine our experimental results to check whether they conform to the above expressions. First consider the stiffness change $(\Delta\mu^{-1})_s$ as a function of I_w as shown in Fig. 1(b). We see from Eqs. (1) and (4) that the constants C and α/β describe this curve completely. Calculating these constants from the saturation level $(I_w\to\infty)$ and from one other experimental point, we obtain the drawn curve in Fig. 1(b), which agrees very well with the experimental points.

Next, let us consider the recovery process after the light is switched off, as in Fig. 1(a). According to Eqs. (1) and (5) a plot of $(\Delta\mu^{-1})^{-1}$ vs t

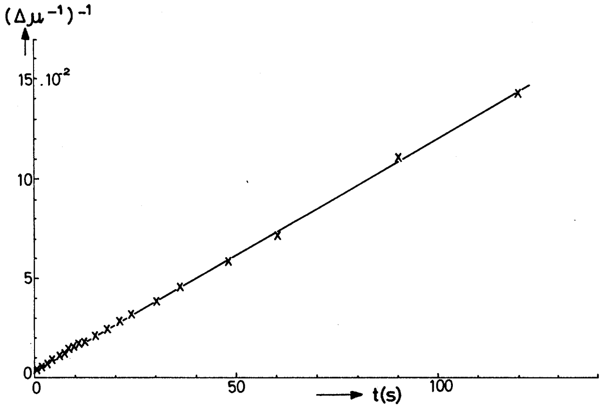

FIG. 2. Inverse of the change in stiffness as a function of time after switching off light of intensity 10^{-2} W/cm² (77°K). Crosses: experimental points; drawn curve (calculated): $10^3\times(0.25+0.12t)$.

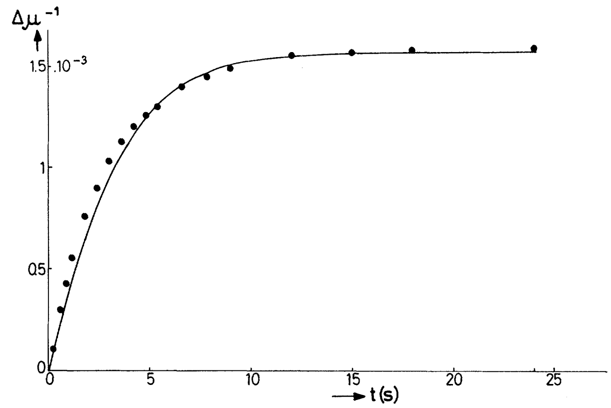

FIG. 3. Change in stiffness as a function of time after switching on light of intensity 9×10^{-5} W/cm^2 (77 °K). Dots: experimental points; drawn curve (calculated): $7.08\times10^{-3}/[1+3.5\coth(0.2t)]$.

should yield a straight line:

$$(\Delta\mu^{-1})^{-1}=1/Cn_s+(\alpha/C)t. \qquad (6)$$

In Fig. 2 just such a plot is shown: The experimental points lie on a straight line to a very good approximation. Equation (6) also shows that the slope of this line should be independent of the stationary level n_S from which the recovery starts. This is indeed the case: We cannot show this in Fig. 2, since most experimental points coincide on the scale of that figure. From the slope α/C of the line and from the value C calculated above we find α; this leads directly to β since α/β was also calculated before.

Finally we examine the decrease in μ after the light is switched on. As a test we have in Fig. 3 plotted $\Delta\mu^{-1}$ vs t for a light intensity I_w not used in the calculations above. The dots are experimental points: The drawn curve represents $\Delta\mu^{-1}$ according to Eqs. (1) and (3) with the values of C, α, and β calculated above. A slight deviation for small t might be due to inaccuracy in determining the point $t=0$. In general we observe a very satisfactory agreement between experiment and calculations.

The proposed model does not specify how the difference in contribution to magnetic properties of centers of type I and II arises, what exactly is the mechanism by which light induces a dissociation of a type-I center, or how electrons, once at a type-II center, move back to Ga donors. Further investigation of each of these points should eventually lead to a quantitative prediction of the constants C, α, and β, in that order. However, the excellent agreement between model calculations and experimental results strongly suggests that Eq. (2) correctly describes the essential features of the process, thus justifying the assumed model.

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ELECTRIC DIPOLE MOMENT OF THE CESIUM ATOM. A NEW UPPER LIMIT TO THE ELECTRIC DIPOLE MOMENT OF THE ELECTRON*

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An atomic-beam magnetic-resonance apparatus has been used in a test of parity and time-reversal invariance in atomic systems. An upper limit to the permanent electric dipole moment of the cesium atom, $|d_{\rm Cs}|<3.7\times10^{-22}e$ cm, has been set. This result leads to an upper limit to the electric dipole moment of the electron, $|d_e|<3\times10^{-24}e$ cm.

An atomic-beam magnetic-resonance technique has been used to set a new upper limit to the electric dipole moment (EDM) of the cesium atom. This upper limit leads in turn to a new upper limit to the EDM of the electron which is a factor of 10 lower than any limit previously reported.

The importance of these experiments lies in the fact that the observation of an EDM in an atomic system of well-defined angular momen-