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OPTICALLY-INDUCED MAGNETIZATION RESULTING FROM THE INVERSE FARADAY EFFECT*

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The application of high-intensity laser radiation to various materials has produced a number of nonlinear effects which in turn have prompted a reformulation of the laws of optics.[1] One consequence of this reformulation was to establish relations between seemingly different phenomena. The first example of these relations to be experimentally observed has been between the linear electro-optic effect and induction of a dc polarization proportional to the intensity of a light beam (i.e., optical rectification) in crystals which lack inversion centers.[2] General phenomenological considerations, however, do predict other relations,[3] but to date only effects of electric rather than magnetic origin have been observed. The purpose of this Letter is to describe the first observation of an optically-induced magnetization in a nonabsorbing material, and to demonstrate the relationship between this effect, which we will call the inverse Faraday effect (IFE), and the Faraday effect in the same material.

The IFE follows from the existence of a potential function F, which for cubic or isotropic media has the form[3]

$$F=\chi\mathcal{H}\left\{\mathcal{E}_R\mathcal{E}_R^*-\mathcal{E}_L\mathcal{E}_L^*\right\},\qquad(1)$$

where χ is a constant characteristic of the material, and $\mathcal{H}$ is the dc magnetic field whose positive sense is taken in the direction of light propagation. The $\mathcal{E}_R$ and $\mathcal{E}_L$ are amplitudes of right and left circularly polarized light, respectively. For example, the electric field for right circularly polarized light is written

$$E(t)=2\,\mathrm{Re}[(2)^{-1/2}(\tilde{x}+i\tilde{y})\mathcal{E}_R\exp i(\omega t-kz)].$$

In the presence of a magnetic field, Eq. (1) yields a contribution to the optical dielectric constants. For right and left circularly polarized light we have[4]

$$\begin{aligned}\Delta\epsilon_R&=-\partial^2F/\partial\mathcal{E}_R\partial\mathcal{E}_R^*=-\chi\mathcal{H},\\ \Delta\epsilon_L&=-\partial^2F/\partial\mathcal{E}_L\partial\mathcal{E}_L^*=+\chi\mathcal{H},\end{aligned}\qquad(2)$$

which can be shown to lead to a rotation of the plane of polarization of linearly polarized light (Faraday rotation) through an angle $\theta/d=VH$ per unit length. The Verdet constant V is

$$V=-4\pi^2\chi/n_0\lambda_0\ [\mathrm{rad\ Oe^{-1}\ cm^{-1}}],\qquad(3)$$

n_0 is the refraction index in the absence of a dc field, and λ_0 is the vacuum wavelength of the light in cm.

On the other hand, in zero applied magnetic field there is a magnetization

$$M=-\partial F/\partial\mathcal{H}=-\chi(\mathcal{E}_R\mathcal{E}_R^*-\mathcal{E}_L\mathcal{E}_L^*),$$

or in terms of measurable quantities,

$$M=\lambda_0V(2\pi c)^{-1}[I_R-I_L],\qquad(4)$$

where I_R and I_L are the intensities of right and left circularly polarized light inside the medium in cgs units [ergs $\mathrm{sec^{-1}\ cm^{-2}}$] and M is in G $\mathrm{cm^{-3}}$.

The IFE has been observed in $\mathrm{Eu^{+2}{:}CaF_2}$,[5] a number of diamagnetic glasses, and several organic and inorganic liquids. Radiation from a Q-switched ruby laser, 0.1 J with a half-amplitude pulse width of 3×10^{-8} sec, was circularly polarized by a quartz $\lambda/4$ plate. The magnetization was detected by a 30-turn pickup coil wound on a phenolic cylinder and slipped over either the cylindrically ground samples, in the case of $\mathrm{Eu^{+2}{:}CaF_2}$ and the glasses, or the cylindrical sample holder in the case of liquids. All measurements described here were done in zero applied dc magnetic field.

The amplified signal from the pickup coil, (dM/dt), and a monitor of the laser intensity were displayed on a dual-trace oscilloscope. Spurious signals due to pickup, etc., were a problem, but they could essentially be eliminated in most situations. Further experimental details will be published elsewhere.[6]

Figure 1 shows oscilloscope traces of signals observed from a single crystal of 3.1% Eu^{2+}-doped CaF_2 at 4.2°K, for right and left circularly polarized light. As predicted by Eq. (4), the change in sense of polarization reverses the sign of dM/dt.

Further confirmation of the IFE follows from the temperature dependence of the magnetization. Van Vleck and Hebb[7] have shown that in many cases the Verdet constant can be described by $V = C_0 + C_1/T$. This relation is more general than the cases they considered, and we will take it as a general empirical law. The first term arises strictly from changes in both the wave functions and energies of the various eigenstates when the populations of these states are held fixed. This is often called the diamagnetic term.[8] The second term C_1/T results from a population readjustment and is usually associated with paramagnetism. In the IFE, circularly polarized light removes the invariance of a crystal in zero magnetic field to time-reversal symmetry and, in second-order perturbation theory, the degeneracy of the Kramers doublet is lifted. The circularly polarized light thus acts as an effective magnetic field. If relaxation processes are sufficiently rapid, the spins will come to equilibrium in this effective field, and thus give rise to the T^{-1} term in the IFE.[9] Since the experiments were all done in zero magnetic field, and the effective field is small, the cross-relaxation processes in concentrated crystals can establish equilibrium in a time short compared to the laser-pulse duration.[10] The temperature-independent term is not limited by such relaxation effects.

The results of measuring M versus T^{-1} in the crystal of 3.1% Eu^{2+}:CaF_2 are plotted in Fig. 2(a). The Faraday rotation of this crystal was measured at 6940 Å as a function of temperature from 1.3 to 300°K. The Verdet constant was found to be $V = [+2.33\times10^{-6} - 1.7\times10^{-3}T^{-1}]$ rad G^{-1} cm^{-1}, so that even at room temperature the paramagnetic term dominates. Measuring the laser-beam cross section at the sample position, measuring its energy, and assuming a Gaussian time dependence yields a peak I_R or I_L of 10^7 W/cm^2. With the above equation for V and this power, the peak induced magnetization is calculated to be 1.4×10^{-5} G cm^{-3} at 4.21°K. Taking into account the finite

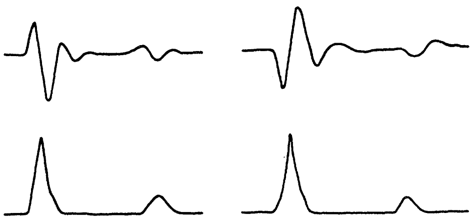

FIG. 1. Top: oscilloscope traces of dM/dt obtained with right and left circularly polarized light for a 3.1% Eu^{2+}-doped CaF_2 crystal at 4.21°K. Bottom: monitor of the laser intensity.

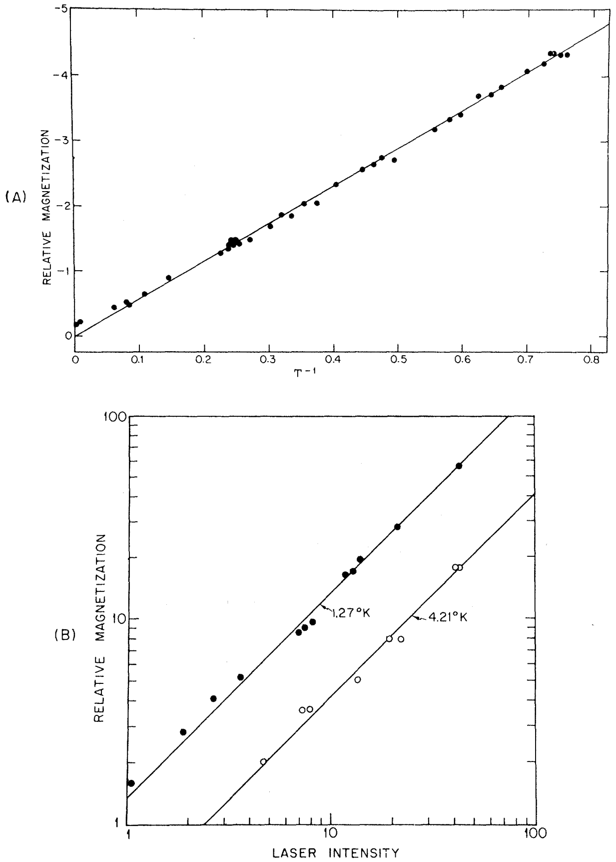

FIG. 2. (a) Temperature dependence of the IFE magnetization in the 3.1% Eu^{2+}:CaF_2 crystals showing the T^{-1} dependence. (b) The linear dependence of the magnetization on the laser intensity measured at 1.27 and 4.21°K. At constant laser power, the ratio of the signals is equal to the inverse ratio of the temperatures.

length of the pickup coil, the fact that the measured cross section of the laser does not fill the coil volume,[11] and using the observed voltage, one calculates a magnetization of 0.94 $\times 10^{-5}$ G, in satisfactory agreement with the above, considering uncertainties in knowing the actual power densities at the sample and the efficiency with which the pickup coil is coupled to the magnetization.

As a further check that we are indeed observing the IFE, Fig. 2(b) shows that the magnetization is a linear function of intensity and that for these two temperatures the T^{-1} behavior is independent of laser power.

A more direct test of Eq. (4) is provided by measuring M/I_R versus V for a variety of materials with different Verdet constants. Table I lists a number of glasses with their measured Verdet constants as well as two Eu^{+2}:CaF_2 crystals. Figure 3(a) shows the plot of measured M/I_R versus V as measured on the same sample. The data are satisfactorily fit by a straight line, even though V changes sign in going from the Eu^{+2}:CaF_2 to the glasses.

Table II lists a variety of liquids with appreciable diamagnetic Verdet constants. The plot of M/I_R versus V in Fig. 3(b) is again a good straight line. The liquids and solids are not compared directly because different coil geometries and filling factors were required, but reasonable estimates of these differences show that the data in Figs. 3(a) and 3(b) are consistent with each other.

A complete quantum-mechanical description of these effects will be reserved for a future publication[6]; however, the main results can

Table I. The measured Verdet constants (in min/Oe cm) of the Schott glasses, "x-ray" glass, and the Eu^{2+}-doped CaF_2 crystals used in the inverse Faraday effect experiment. The wavelength was 6940 Å, and the temperature 295°K. The second column gives the glass type in terms of the Schott catalog number.

No.	Glass type	V
1	SFS 6	0.068
2	SF 6	0.048
3	X ray[a]	0.044
4	SF 4	0.042
5	SF 5	0.032
6	LF 5	0.023
7	BaF 3	0.020
8	BaK 2	0.015
	CaF_2	
9	3.1% Eu^{2+}	−0.012
10	4.9% Eu^{2+}	−0.019

[a] Pittsburgh Plate Glass Company photographic-quality x-ray lead glass.

Table II. The Verdet constants (in min/Oe cm) of several organic and inorganic liquids used in the diamagnetic inverse-Faraday-effect experiment. The wavelength was 6940 Å, and the temperature 295°K.

No.	Liquid	V measured
1	Antimony pentachloride SbCl_5	0.043
2	1-Chloronaphthalene $\mathrm{C}_{10}\mathrm{H}_7\mathrm{Cl}$	0.029
3	Carbon disulfide CS_2	0.027
4	N,N-dimethylanaline $\mathrm{C}_6\mathrm{H}_5\mathrm{N}(\mathrm{CH}_3)_2$	0.026
5	Bromobenzene $\mathrm{C}_6\mathrm{H}_5\mathrm{Br}$	0.0195
6	Benzene $\mathrm{C}_6\mathrm{H}_6$	0.019
7	Nitrobenzene $\mathrm{C}_6\mathrm{H}_5\mathrm{NO}_2$	0.014

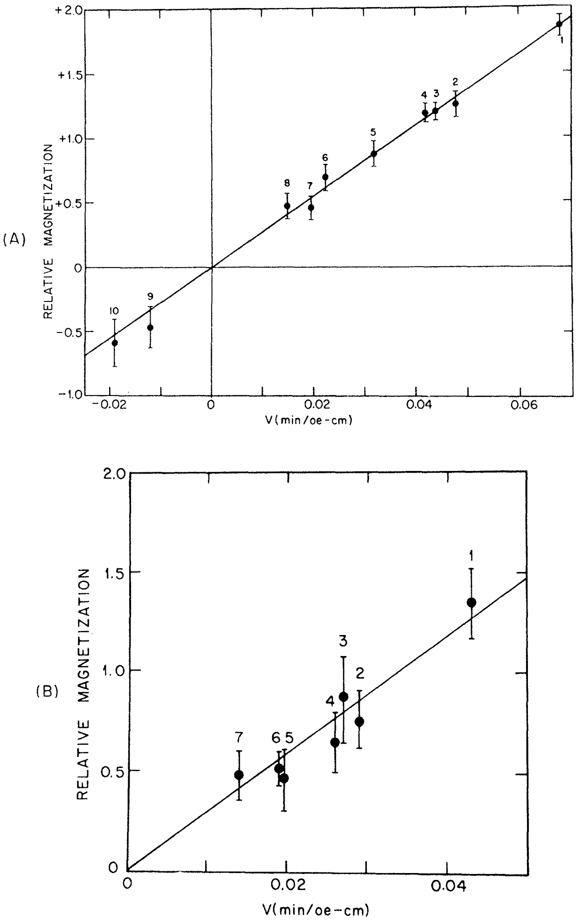

FIG. 3. (a) The measured values of M/I_R for several glasses (1-8) and Eu^{2+}-doped CaF_2 crystals (9-10) (listed in Table I) plotted as a function of the measured Verdet constants at 6940 Å. (b) Similar measurements for the organic and inorganic liquids given in Table II.

be quickly stated. Consider the case of a single atom interacting with an electromagnetic field. Assuming there are no one-photon absorption processes, it is possible to diagonalize the complete Hamiltonian to second order in the interaction with the optical fields. Within the ground-state manifold of the atom, all of the effects of this second-order perturbation treatment are equivalent to an effective Hamiltonian

$$\begin{aligned}\mathcal{H}_{\text{eff}} = {} & a[|\mathcal{E}_R|^2 - |\mathcal{E}_L|^2]J_z \\ & + b[|\mathcal{E}_R|^2 + |\mathcal{E}_L|^2][J_z^{\,2} - \tfrac{1}{3}J(J+1)] \\ & + 2c\,\mathrm{Re}(\mathcal{E}_R\mathcal{E}_L^*)(J_x^{\,2} - J_y^{\,2}) \\ & + 2d\,\mathrm{Im}(\mathcal{E}_R\mathcal{E}_L^*)(J_xJ_y + J_yJ_x). \qquad (5)\end{aligned}$$

In a weak-crystal-field approximation J will be the angular-momentum operator of the ground state and $b = -c = d$. In a strong crystal field J will be the "effective spin" operator and $b \neq -c \neq d$. The paramagnetic term in the IFE can be shown to follow from the first term in Eq. (5). If $\mathcal{E}_R\mathcal{E}_R^* \neq \mathcal{E}_L\mathcal{E}_L^*$ this term is equivalent to an effective magnetic field. For example, for an atom with Kramer's degeneracy, if J_z is diagonal in the eigenstates of the "crystal field," this term lifts the degeneracy and the IFE results from thermal equilibrium in the effective magnetic field. The diagonal parts of the remaining terms in Eq. (5) will cause further splittings of the ground-state manifold, but they will cause no magnetic effects in zero applied dc field.

The off-diagonal terms in Eq. (5) can be shown to induce a Raman effect between states of the ground multiplet.[6] If the linewidth of the laser radiation is greater than the zero-field splitting, $\mathcal{E}\mathcal{E}^*$ has time-dependent terms that will cause transitions which can properly be designated "stimulated Raman transitions." Quantum-mechanical calculations show b, c, and d are generally greater than a. From the present measurements on Eu^{+2}:CaF_2 in an unfocused laser beam, it is possible to obtain an a $\mathcal{E}_R\mathcal{E}_R^*$ term corresponding to an effective field of 0.01 G. With a focused laser, values equivalent to 1-10 G are reasonable, and since b, c, and $d \gtrsim a$, transition rates greater than 10^{10} ($\sec^{-1}$) are obtainable. It is significant that experimental knowledge of the probable size of this unconfirmed effect follows directly from the Verdet constant once the quantum-mechanical basis of the IFE is demonstrated.

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