Photoinduced Magnetic Effects in an Insulating Spin-Glass: Cobalt Aluminosilicate Glass

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From Faraday rotation measurements, evidence is given that irradiation, with near-infrared light, of the cobalt aluminosilicate glass in its spin-glass state hastens the relaxation of the thermoremanent magnetization. Experimental data on the dependence of this photomagnetic anneal with radiation power, light wavelength, and polarization are discussed within a simple phenomenological model.

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The cobalt aluminosilicate glass exhibits similar magnetic properties as the Cu-Mn metallic alloy and so may be considered as an insulating spin-glass at low temperature.[1-3] We report here on the discovery of photoinduced magnetic effects below its freezing temperature T_f.

In this Letter we shall demonstrate how the relaxation of the thermoremanent magnetization (TRM) or the in-field magnetic aftereffect can be speeded up during exposure of the sample to near-infrared radiation even under a low light intensity ($I_0 \simeq 3$ μW/mm^2). This effect allows us for the first time to manipulate the TRM of a spin-glass without use of an external field.

The evidence of photomagnetism in insulating magnetic crystals was first pointed out by Teale and Temple[4] in Si^{4+}-doped yttrium iron garnet (YIG). It was interpreted as a light-induced change of the anisotropy via electron charge-transfer transitions between Fe^{2+} and Fe^{3+} ions.

In the present case the photomagnetism could not be explained within a similar model and we shall give a phenomenological interpretation of this phenomenon in the framework of two-level systems. The experimental data on the dependence of the photomagnetic anneal upon the radiation power, the wavelength, and the state of polarization of the light agree with our proposed model.

The 40% CoO, 20% Al_2O_3, 40% SiO_2 (13.3 at.% Co) sample investigated was cut and optically polished as a thin plate ($0.16 \times 2 \times 2$ mm^3). Similarly prepared glasses showed no crystallinity when examined by powder x-ray diffraction and electron microscopy.[1] The cobalt ions are presumably in their divalent state but no information is available on their distribution between tetrahedral and octahedral sites.

The measurements of the time dependence of the TRM after switching off the magnetic field or of that of the magnetic aftereffect after switching on the field were performed by Faraday rotation (FR).[2,5] The light beam was always propagating along the magnetic field direction and normally to the plate. As expected, the FR in our sample was found to be proportional to the magnetization measured directly by a Foner magnetometer. For the considered specimen, the sensitivity of the FR apparatus was good: $\delta M = 10^{-6}$ emu. With a cw AsGa laser diode ($\lambda = 8330$ Å) as a light source, the calibration was $\theta/M = 435$ deg/emu. The use of a small superconducting solenoid to generate the magnetic field allowed us to switch fields, as high as 5 kOe, in less than 0.1 s.

Two types of experimental setup were used to prove that the measured FR is only due to the change in magnetization: In setup A the state of polarization of the laser beam crossing the sample was modulated at 100 kHz to measure the FR. Its intensity was controlled by neutral-density filters. In setup B the intensity of the light beam used for the FR measurement was kept as low as possible (~1.5 μW/mm^2). The sample illumination may be monitored independently in intensity, polarization, and wavelength by a second beam coming from another cw AsGa laser diode or from a XBO xenon lamp coupled to a continuously variable spectral filter. A monochromatic filter was placed just in front of the photomultiplier to eliminate only the pumping radiation.

The most direct evidence for the photoinduced magnetic effect is shown in Fig. 1 from the acceleration of the relaxation of the thermoremanent FR (TRFR), measured well below T_f at 1.29 K, just after a sudden illumination (100 μW/mm^2) of the sample. This effect turned out to be irreversible since the FR lowered by illumination cannot be raised again by switching off the light or by decreasing the temperature. We recall that the thermoremanent magnetization M_R is obtained after cooling the sample in a magnetic field through T_f and switching off the field for a given temperature $T < T_f$.

We have excluded sample heating as the source

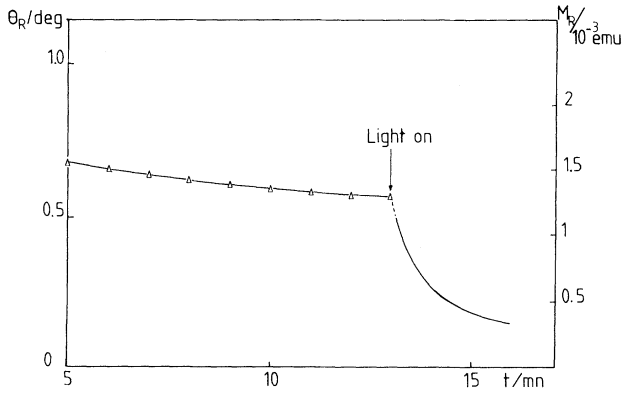

FIG. 1. Time dependence of the thermoremanent Faraday rotation of the sample at 1.29 K and of the corresponding TRM, after it is magnetized under 4 kOe. In order to discern only the thermal relaxation we have rapidly measured the FR for discrete values of the time (Δ) under a weak light intensity of 1.5 μ W/mm^2. A drastic change in the relaxation appears after lighting the sample (t > 13 min) with 100 μ W/mm^2.

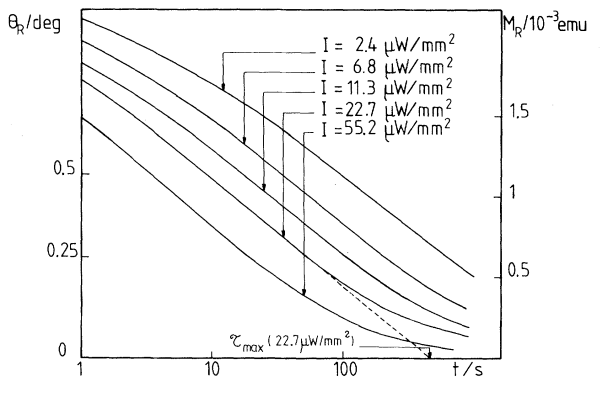

FIG. 2. Time dependence of the thermoremanent Faraday rotation (or of the corresponding TRM) of the sample under illumination for several values of the light intensity at 1.29 K, after it is magnetized under 7.5 kOe.

of magnetic changes since an estimation of the increase of the lattice temperature, based on the known values of the thermal conductivity and absorption coefficient, is only $\Delta T = 7\times10^{-5}$ K per μW/mm^2. This leads to a decrease in remanent magnetization which cannot exceed $10^{-2}M_R$ for the highest radiation power used, i.e., a small variation in comparison with experimental data.

To interpret the photomagnetic anneal of the TRM we shall assume that the spin-glass phase may be described as an assembly of weakly interacting magnetic clusters which are randomly distributed inside the sample. We also suppose that the optical excitation of any ion belonging to one cluster induces quite instantaneously the relaxation of its metastable spin equilibrium. The demagnetization time of any optically excited cluster always becomes shorter than the time of measurement. The thermal relaxation of the TRM in darkness will be neglected in this treatment, which is reasonable considering the data given in Fig. 1.

Let us define by $n(t,x)$ the number of unexcited clusters per unit length after a time t following the switching of the field, at a depth x from the illuminated crystal face. The number of excited clusters per unit time is

$$n_{ex}(t,x) = n(t,x)wI_0e^{-\mu x}. \quad (1)$$

I_0 stands for the radiation power per unit area and w is the absorption cross section per unit of energy. The absorption coefficient for an optically excited cluster is called μ.

Since $dn(t,x) = -n_{ex}(t,x)dt$, one gets

$$n(t,x) = n(0,x)e^{-t/\tau}, \quad \tau = e^{\mu x}/wI_0. \quad (2)$$

The integration of expression (2) over the thickness d allows us to determine the TRM of the illuminated sample:

$$M_R(t) = \frac{M_R(0)}{\mu d}\int_{\tau_{min}}^{\tau_{max}} e^{-t/\tau}\frac{d\tau}{\tau}, \quad (3)$$

where $\tau_{max} = e^{\mu d}/wI_0$ and $\tau_{min} = 1/wI_0$.

This expression (3) of M_R is similar to that used by Richter[6] to interpret the magnetic viscosity of rocks and may be analyzed in a similar manner. However, the origin of such an expression differs significantly in the two cases. For Richter it results from the distribution of the relaxation times while in our case it arises from the progressive light absorption inside the sample. The photoinduced change of the TRM is governed by Eq. (3), which yields the particular solutions

$$\begin{aligned} &M_R(t \ll \tau_{min}) = M_R(0)[1-(wI_0/\mu d)(1-e^{-\mu d})t], \\ &M_R(\tau_{min} \ll t \ll \tau_{max}) \\ &\quad = M_R(0)[0.423-(\mu d)^{-1}(\ln wI_0 + \ln t)], \\ &M_R(t \gg \tau_{max}) = M_R(0)(e^{\mu d}/\mu d wI_0)t^{-1}\exp(-t/\tau_{max}). \end{aligned} \quad (4)$$

In Fig. 2, we report the time evolution of the TRFR (or of the corresponding TRM) measured at 8330 Å with setup A for different values of the light intensity. The relaxation of the TRFR, θ_R, looks similar to that predicted theoretically by Richter.[6] As expected from expression (4), three

regimes may be distinguished for all $\theta_R(\ln t)$ curves, depending on the t value. In the intermediate time range (for example, $3.5 < t < 60$ s for the 22.7-μW/mm^2 curve) the TRFR is found to be proportional to $-\ln t$ and its slope $\partial\theta_R/\partial\ln t$ stays nearly constant whatever the light power is. This agrees well with expression (4) for $\tau_{min} \ll t \ll \tau_{max}$. In this time range, all curves shown in Fig. 2 may be deduced from one to another just by a translation in the $\ln t$ scale.

For long times the TRFR fails to vary like $-\ln t$ and is probably better fitted by the predicted $t^{-1}\exp(-t/\tau_{max})$ law, but the lack of measurements for $t \gg \tau_{max}$ does not permit us to verify such a behavior. A crude estimation of τ_{max} may be drawn from the value of t obtained by extrapolation of the intermediate part of the relaxation curve to zero (Fig. 2). τ_{max} decreases when the light intensity I_0 is raised and within the above approximation one may consider that it varies closely to I_0^{-1}, in agreement with Eq. (4). A similar variation is exhibited by τ_{min} since the length of the $\ln t$ linear part of the relaxation curves remains nearly constant, whatever the intensity. This is still expected because the quantity $\ln(\tau_{max}/\tau_{min}) = \mu d$ does not depend on the intensity of the incident radiation. Since $d = 4.02$ at 8330 Å, one finds $\tau_{max}/\tau_{min} = 56$, a value slightly lower than that deduced from the curves shown in Fig. 2.

From this data we deduced the dependence of the TRFR, θ_R, with the light intensity for several values of t. The $\theta_R(\ln I_0)$ curves look similar to those reported in Fig. 2. This behavior is well explained from the same role played by t and I_0 in expressions (4) when $t \ll \tau_{max}$. In particular θ_R is proportional to $-\ln I_0$ in the intermediate power range.

Similar results have been obtained when the illumination is monitored with a second laser beam (setup B), but the photomagnetic efficiency was lower than that obtained in the previous case (setup A). This may be understood if one notes that the cross section and homogeneity of the two light beams on the sample can differ significantly.

Since the slope $\partial\theta_R/\partial\ln t$ is strongly temperature dependent, our data still confirm that the sample heating is negligible. The spin temperature is obviously increased during the photomagnetic anneal but the lattice temperature is practically unaffected. The light only destroys the magnetic metastability in the system.

Using setup B we have checked that the state of polarization of the light does not significantly affect the relaxation of the TRFR. In particular, one cannot invoke a change in magnetization induced by optical pumping since right- or left-circularly-polarized light equally demagnetizes the sample in spite of the axial symmetry of the TRM which would favor one sense of polarization (inverse Faraday effect). We also checked that the illumination of the sample with polarized or unpolarized light does not modify its absorption coefficient μ_s at 8330 Å ($\Delta\mu_s/\mu_s < 10^{-4}$).

The photomagnetism in such an amorphous insulating spin-glass differs significantly from that found in Si^{4+}-YIG, for which optical pumping by polarized light induces a large dichroism.[7]

The radiation is active for wavelengths longer than 6800 Å, for which the sample becomes patially transparent. Below 6800 Å the too-strong absorption of the sample used does not allow one to perform FR measurements. Below 8200 Å the sample absorption coefficient μ_s increases. If one assumes that $\mu = \mu_s$, τ_{max} is enhanced and the photoinduced magnetic anneal efficiency is reduced, as predicted from Eq. (4).

Our experimental data on the in-field magnetic aftereffect are well described by equations conjugate to expressions (4), in particular for the photoinduced speediness in the time of acquisition of the magnetization.

To go further one needs a better understanding of the microscopic origin of this effect. A preliminary experiment shows that the illumination of the sample on a small area does not perturb the relaxation of the TRM measured on another part of the sample close to the former (~0.5 mm). This demonstrates that the spin excitation does not extend over the whole sample but remains rather localized, at least under the present experimental conditions. Thus one can imagine that spin clusters are excited by the light quite independently. This does not exclude a weak coupling between clusters. Instantaneously, the optical excitation of a given ion may modify the local anisotropy or exchange interaction, thus releasing the metastable magnetic state of the whole cluster because of the large antiferromagnetic exchange interaction between Co^{2+} ions.[1]

A crude calculation shows that each photon must change the spin state on an average of at least 100 Co^{2+} ions to explain the experimental results. Then, one may suppose that the light induces spin defrustration of some parts of the system. Another possibility is to suppose that metastable small domain structure exists in the

magnetized sample and that the illumination favors domain-wall movement by releasing the local anisotropy of Co^{2+} ions or defects.

In summary, we have demonstrated that light irradiation changes the magnetization of such an amorphous insulating spin-glass. This photoinduced magnetic effect is not a thermomagnetic effect and may be used to obtain a better knowledge of the spin-glass state. As we have mentioned earlier,[2] starting from a zero-field–cooled process, it is easy to achieve rapidly the in-field magnetic state at equilibrium under illumination, allowing us to study the behavior of the TRM without heating the sample above T_f. Further investigations are under way to improve our knowledge of the origin of photomagnetism.

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Magnetoresistance Measurement of the Electron Inelastic Scattering Time in Two-Dimensional Al Films in the Presence of Superconducting Fluctuations

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Magnetoresistance measurements of thin Al films ($1\ \Omega/\square < R_\square < 60\ \Omega/\square$) between 2.5 and 30 K are reported and analyzed within the framework of localization theories. The influence of superconducting fluctuations is felt far above the critical temperature, in agreement with the theory of Larkin. The electron inelastic scattering is inversely proportional to the temperature in accordance with Abrahams *et al.* Its absolute value, which agrees with phase-slip-center measurements, is 1 order of magnitude larger than predicted.

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A fairly complete theoretical picture of electron localization effects in two dimensions[1] has emerged in the last few years, and most of the predicted effects[2] have been observed experimentally in thin metallic films and in metal-oxide-semiconductor field-effect transistor devices.[3] However there seems to be at this point one serious disagreement between theory and experiment, related to the dependence of the inelastic scattering time τ_i on the temperature T and on the coefficient of diffusion D. Magnetoresistance (MR) measurements which lead to a determination of τ_i (Ref. 4) in thin films[5-13] have failed until now to verify the prediction by Abrahams *et al.*[14] that τ_i is dominated by electron-electron scattering, and is inversely proportional to $T \ln T$ and proportional to D. In particular, Bergmann[15] has found in quench-condensed metal films $(\tau_i)^{-1} \propto T^{1.65}$,