Observation of inverse Cotton-Mouton effect in the magnetically ordered crystal (Lu, Bi)$_3$(Fe, Ga)$_5$O$_{12}$

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A nonthermal increase in the magnetization of a magnetic crystal illuminated by linearly polarized optical radiation has been observed for the first time. A first experimental estimate of the Cotton-Mouton constant has been found for a film of a rare-earth iron garnet containing bismuth.

Several studies of nonlinear-optics phenomena in magnetically ordered media have now been carried out.[1] In particular, there have been studies of changes in the optical characteristics[2] and the magnetization of crystals caused by intense laser light which is propagating through them.[3–6]

An inverse Cotton-Mouton effect was described in Ref. 7: a magnetization of a medium by linearly polarized laser light. It was shown that the ratio of the magnetizations induced by the light in the inverse Faraday effect and in the inverse Cotton-Mouton effect is equal to $\Delta n_c/\Delta n_e$ in order of magnitude, where $\Delta n_{c,e}$ are the changes in the refractive indices which lead to a magnetic circular and a magnetic linear birefringence, respectively. Theoretical and experimental data[8] on a long list of magnetic crystals are known to indicate a relation $\Delta n_c \simeq \Delta n_e$.

In the present letter we report an experimental observation of the inverse Cotton-Mouton effect. The $(\mathrm{R,Bi})_3(\mathrm{Fe,Ga})_5\mathrm{O}_{12}$ ($\mathrm{R} = \mathrm{Lu}$) films, grown by epitaxy on a gallium-gadolinium garnet substrate in the [111] orientation, have a thickness $\simeq 10\ \mu$m. The linearly polarized beam from a neodymium laser with a pulse length $\tau \simeq 20$ ns and a beam diameter of 1.3 mm is incident normally on the surface of the sample, which is immersed in an external magentic field directed parallel to the laser beam. The polarization of the light can be varied with a $\lambda/4$ plate. A change in the magnetization of the sample is detected by a planar three-turn coil on the surface of the sample. The signal from this coil is fed to the input of a broad-band, low-noise amplifier and then to an oscilloscope. The rise time of the transient characteristic of the signal transmission line is ~ 3.5 ns.

All the measurements are taken at room temperature. We made a special study of the contribution of clean substrates to the observed effect. We found no signal from them.

Figure 1 shows a typical oscilloscope trace of the laser pulse (a) and the detected signal (b). Changing the direction of the external magnetic field changes the sign of the emf. Within the measurement error ($\sim 15\%$), we observe no dependence of the detected signal on the polarization of the laser light.

There is a fundamental distinction between the shape and the sign of the signal (the measured emf), on the one hand, and those of signals associated with thermal and

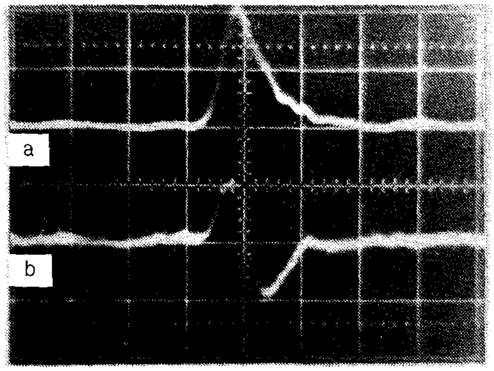

FIG. 1. a—Shape of the laser pulse; b—shape of the emf signal detected, which is proportional to the rate of change of the magnetization of the crystal. The sweep is 20 ns/div.

thermoelastic effects, which have been observed in ferromagnetic metals and ferrites.[9–11] In our case the change in the magnetization corresponds to an *increase* in the magnetization in comparison with the steady-state value in the absence of the laser light, and the shape of the detected signal is approximately the derivative of the intensity envelope. In Refs. 10 and 11, on the other hand, the pulse of variable emf was similar in shape to the intensity envelope, and its sign corresponded to a *decrease* in the steady-state magnetization.

An increase in the light intensity to ~ 70 MW/cm^2 causes damage to the surface of the film, an increase in the absorption, and a distortion of the shape of the signal, apparently due to thermal effects. Figures 2 and 3 show the peak-to-peak amplitude of the signal per turn of the coil versus the external magnetic field and the energy of the laser pulses for saturating magnetic fields at nondestructive intensities.

To find a theoretical description of the inverse Cotton-Mouton effect, we used a single-sublattice model which combines the tetrahedral and octahedral sublattices of Fe^{3+} ions into a common sublattice (the Lu^{3+} ion is nonmagnetic). A model of this sort gives a satisfactory description of the magnetic linear birefringence in iron garnets.[8,12] We can write the change in the free energy of a single domain of a cubic crystal in the presence of an electromagnetic field $\mathbf{E}(t)$ in the transparency region as

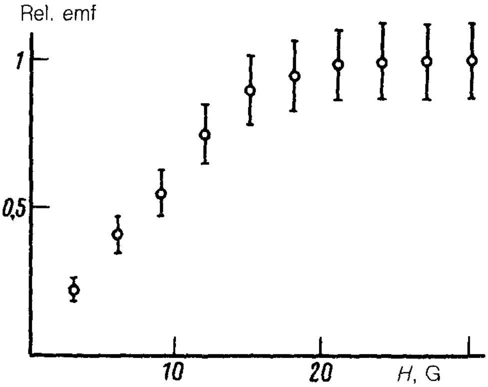

FIG. 2. Amplitude of the emf signal versus the magnetic field H.

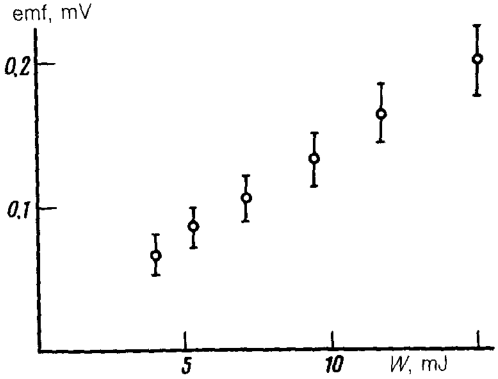

FIG. 3. Peak-to-peak amplitude of the emf signal versus the energy of the laser pulses, W.

$$-16\pi\Delta F = g_{12}M^2(\mathbf{EE}^*) + 2g_{44}(\mathbf{ME})(\mathbf{ME}^*) + (g_{11} - g_{12} - 2g_{44})M_i^2|E_i|^2 , \qquad (1)$$

where g_{ij} are the components of the dielectric tensor, $\mathbf{M} = \mathbf{M}_0 + \chi\mathbf{H}$, $\mathbf{M}_0$ is the magnetization of the domain, $\mathbf{H}$ is the magnetic field, and χ is the susceptibility of the paramagnetic process. In writing (1) we have ignored terms which stem from the nonuniform interaction of the domain walls with the radiation field. The constants g_{44} and $(g_{11} - g_{12})$ describe the magnetic linear birefringence, while the isotropic term $\sim g_{12}$ drops out of the magnetic linear birefringence.

For the geometry corresponding to our experiment, and for a magnetization induced by a light wave in the [111] direction, we find, after taking an average over all domains,[7]

$$\Delta M_{[111]} = \frac{\chi M(\mathbf{H}) c n_0 \gamma}{12\pi\omega}\, \mathbf{E}(t)\mathbf{E}^*(t) C_{[111]}(\omega)\left(1 + \frac{3}{2}\,\frac{g_{12}}{g_{11} - g_{12} - 2g_{44}}\right). \qquad (2)$$

Here n_0 is the refractive index, ω is the light frequency, $\gamma = (1 - a)/a$, a is the magnetooptic anisotropy, $C(\omega)$ is the Cotton-Mouton constant, and $M(\mathbf{H})$ is the magnetization of the crystal. It follows from (2) that the inverse Cotton-Mouton effect is determined both by the constants of the magnetic linear birefringence $[C(\omega),\gamma]$ and by the constant $\sim g_{12}$, which describes the isotropic magnetic refraction.

We see that the dependence of the single amplitude on the magnetic field should be the same as the $M(\mathbf{H})$ dependence and should depend linearly on the light intensity. Experimentally, we see both of these dependences. The shape of the emf signal under quasisteady conditions reproduces the derivative of the shape of the laser pulse and does not depend on the polarization of the light, again in agreement with experimental data.

Let us estimate $\Delta n = (c/\omega)CM_0^2$, assuming[8,12] $g_{12}/(g_{11} - g_{12} - 2g_{44}) \sim 1$. Using $\Delta M_{[111]} = 6.4\times 10^{-4}$ G at $E = 1.4\times 10^5$ V/cm, and also assuming $n_0 = 2.3$, $\chi = 2\times 10^{-3}$, $a = 1.7$, and $\lambda = 1.06\ \mu$m, we find $\Delta n = 5\times 10^{-5}$.

We also studied films with R = Yb and Tm of similar composition. For the films with R = Yb, we again observe an inverse Cotton-Mouton effect, while for the films with Tm, which—in contrast with the samples with Lu—have a significant absorption coefficient at frequencies corresponding to the output of a neodymium laser, we observe only a thermal signal of significantly greater amplitude, similar to that which has been observed previously[11] in $YFeO_3$.

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