Polarization-dependent photoinduced change in antiferromagnetic resonance of α-Fe_2O_3:Eu,Ga

G. S. Patrin, N. V. Volkov, and G. A. Petrakovskiĭ
L. V. Kirenskiĭ Institute of Physics, Siberian Branch of the Academy of Sciences of the USSR

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A photoinduced change in magnetic properties, which depends on the polarization of the exciting light, has been observed in hematite single crystals containing europium ions in the weakly ferromagnetic phase. This effect is observed at temperatures $T \leqslant 70$ K. A possible mechanism for the photomagnetic interactions is discussed.

Exposure of magnetic materials to optical radiation results, as we know, in a variety of changes in magnetic properties. Photomagnetic materials in which the photosensitive centers are rare-earth ions exhibit some extremely interesting and diverse effects: the formation of a new magnetic state in $EuCrO_3$ (Ref. 1), a photoinduced spin-reorientation transition in $ErCrO_3$ (Ref. 2), and an influence of photoexcited states of holmium ions, at dynamic equilibrium with the optical radiation field, on the magnetic state of the $Y_3Fe_5O_{12}$ crystal.[3] All these effects occur at low temperatures, because of the particular electronic structure of the rare-earth ions.

Hematite single crystals and the effects of various impurities on their properties have been studied in detail and are described at length in the literature. There is, on the other hand, essentially no information on the properties of these crystals when doped with rare-earth ions. In this letter we are reporting a study of weakly ferromagnetic hematite single crystals containing rare-earth ions. In addition to the rare-earth ions, we doped the crystals with gallium ions, which are known[4] to lower the Morin transition temperature, thereby expanding the region in which the weakly ferromagnetic phase exists.

For the measurements we used single crystals of α-Fe_2O_3:Eu (0.035% at.), Ga (5% at.), grown from molten solution. The state of the magnetic system was monitored by measuring the parameters of the antiferromagnetic resonance at a microwave frequency $f = 35$ GHz. The optical pumping was carried out with white light from an

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incandescent lamp. The light propagated along the direction perpendicular to the external magnetic field.

It was found that the α-Fe_2O_3:Eu,Ga crystals undergo a change in magnetic state during exposure to polarized light, different in this regard from doped hematite crystals which have been studied previously.[5,6] Figure 1 shows the temperature dependence of the resonant field [illegible] before and after exposure to the light. Curve *2* was recorded under conditions such that the static magnetic field $\mathbf{H}_0$ and the electric field of the light wave, $\mathbf{E}$, were parallel. We see that exposure to light in this manner results in an increase in the resonant field. The curves of the resonant field before and after exposure to light merge at temperatures $\leqslant$65–70 K. The shape of the microwave absorption line is not changed by the exposure to the light. The maximum photoinduced change does not depend on the intensity of the optical radiation; only the time required to reach the saturation level depends on this intensity. The photoinduced state which arises is also independent of the history of the sample. It is determined exclusively by the direction of the polarization plane of the optical radiation with respect to the direction of the magnetic field. After the light is turned off, however, the magnetic state undergoes a relaxation, as can be seen from the 15–20% decrease in the shift of the resonant field. This value then remains the same for an arbitrarily long time. Exposure to light in the configuration $\mathbf{H}_0 \perp \mathbf{E}$ restores everything to its original state.

Figure 2 shows the resonant field as a function of the angle (θ) between the magnetic field and the polarization plane of the optical radiation. At a fixed temperature T, this behavior is described well by the expression

$$H_{res}(T, \rho) = H^0_{res}(T) + h(T, \rho)\cos^2\theta, \qquad (1)$$

where $H^0_{res}(T)$ is the value of the resonant field in the absence of the optical pump, $h(T,\rho)$ is the maximum shift of the resonant field, and ρ is the density of the optical

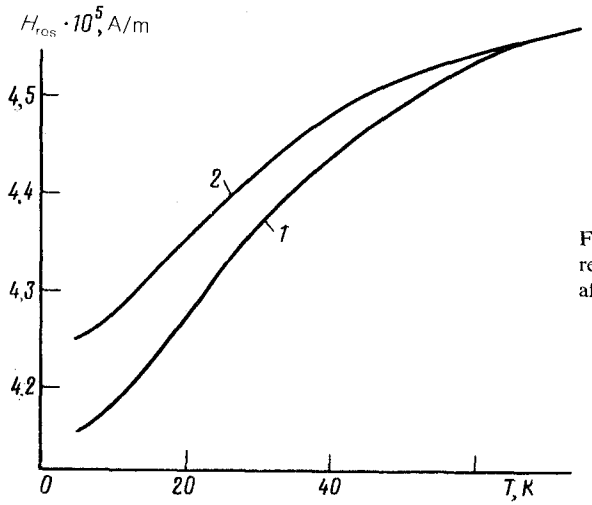

FIG. 1. Temperature dependence of the resonant field. *1*—Original sample. *2*—after exposure to light with $\mathbf{E}\|\mathbf{H}_0$.

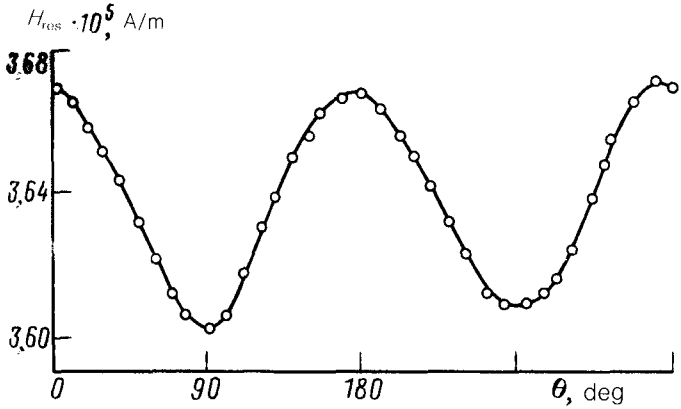

FIG. 2. Resonant field as a function of the angle (θ) between the polarization plane of the light and the direction of the magnetic field.

radiation. This is the behavior, regardless of the direction of the external magnetic field with respect to the crystallographic axes in the basal plane. The probability for an optical transition of a center between states $|m\rangle$ and $|n\rangle$ is given by the following expression in the electric-dipole approximation:

$$W_{nm} \sim |\langle m|\mathbf{dE}|n\rangle|^2, \tag{2}$$

where $\mathbf{d}$ is the electric dipole moment of the absorbing center. If, in the simplest case, of centers which do not interact with each other, the photoinduced shift is proportional to the number of photoexcited centers, then it follows from a comparison of expressions (1) and (2) that the electric dipole moment of the photocenter is along the direction of the external magnetic field. The increase in the resonant field during exposure to the light and its relaxation after the light is turned off indicate that the light puts the magnetic system in an metastable excited state. The ground state is reached only after exposure to light in the configuration $\mathbf{E}\perp\mathbf{H}_0$.

We attribute this behavior of the photoinduced changes to photosensitive complexes which arise because of the presence of europium ions. Hematite crystals doped with gallium ions alone exhibit a completely different response to optical radiation.[6] In the crystals containing europium ions studied in the present experiments, there are no low-temperature anomalies on the temperature dependence of the linewidth or the resonant field which would be linked with the presence of divalent iron ions. One might suggest that the deficiency of oxygen is offset in this case by the appearance of divalent europium (and that this effect reduces the probability for the appearance of Fe^{2+} ions).

Note that the temperature T_f for α-Fe_2O_3:Eu,Ga falls in a temperature region in which anomalies are observed in the electrical properties of nonstoichiometric EuO_x crystals. These anomalies are explained in terms of the presence of impurity magnetic centers consisting of a trivalent europium ion and an electron, captured in the field of

an oxygen vacancy.[7] In the latter case, the changes in the electrical properties are associated with a restructuring of the impurity center and involve an activation energy.

We suggest the following model for interpreting these experimental results. We take the photosensitive center to be a complex consisting of an oxygen vacancy and a divalent europium ion, exchange-coupled with the matrix. Because of the very slight anisotropy of the Eu^{2+} ion, the direction of the external magnetic field is a special direction for such a complex. The existence of this special direction in turn imposes selection rules on optical transitions. The optical transition during the absorption of the light is apparently an intracenter transition. Our reasoning here is that if such a center were excited by a radiationless energy transfer after absorption of the light by the system of iron ions, then all the information about the polarization of the exciting radiation would be lost.

It is assumed that the photoelectron in the excited state is captured by the field of an oxygen vacancy, with the result that an Eu^{3+}-e center forms.

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