SQUID studies of photoinduced magnetization in a γ–Fe_2O_3:Zn single crystal

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A photoinduced change (Δm_l) in the magnetization in a ferromagnetic single crystal γ-Fe_2O_3 doped with zinc ions was discovered with a SQUID magnetometer . The effect is observed at temperatures $T \leqslant 60$ K. Analysis of the characteristic features of the temperature dependences of Δm_l showed that the detected changes caused by the redistribution of the populations of the sublevels of the ground state multiplet of the Fe^{2+} ions in an optical pumping cycle which leads to efficient cooling of the spin subsystem of the photocenters.

The application of SQUID methods to magnetism problems has opened up new possibilities for performing investigations in weak magnetic fields and weakly magnetic substances. The SQUID magnetometer is disinguished from brute-force methods for measuring the magnetic characteristics in investigations of interactions induced by external forces by the important advantage that its sensitivity is independent of the level of the signal against the background of which the induced changes are detected. One direction in which the advantages of this method can be realized is photomagnetism. In this direction we call attention to investigations of the effect of optical radiation on the magnetic properties of semimagnetic semiconductors of the system (MeMn)X, where X = S, Me = Eu, Zn, Cd (Ref. 1) or X = Te and Me = Hg, Cd (Ref. 2).

For magnetically concentrated substances measurements of the magnetic susceptibility by the standard methods with optical irradiation do not yield detailed information about the nature of the photosensitive complexes and the mechanism of the restructuring of their energy structure, since either the methods are not sensitive enough or the effect is masked by a stronger process of magnetic restructuring. For example, a series of experimental studies of the photomagnetic effect was performed on iron spinel samples.[3–6] In these studies the authors explain the photoinduced changes by a restructuring of the domain structure and the mechanism is associated with the charge transfer between heterovalent ions of the same chemical element.

In the present paper we report the observation of the photomagnetic effect in single crystals of the γ phase of iron sesquioxide stabilized by the introduction of zinc ions, and we report the results of experimental studies which illustrate the possibilities of the SQUID technique for studying small changes in a strongly magnetic substance.

The experiments were performed on the apparatus described in Ref. 7 . The sample consisted of a $1 \times 1.5 \times 0.2$ mm^3 plate, whose plane coincided with the (111) crystallographic plane. The optical beam from the laser with $\lambda = 0.63$ μm was directed along the

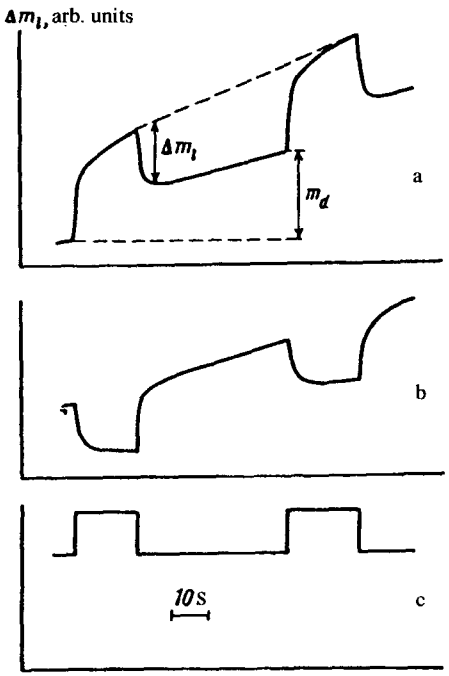

FIG. 1. Time dependence of the magnetization: a — $T = 11$ K, $H = 100$ Oe; b — $T = 17$ K, $H = 300$ Oe; c — light pulse; m_d — domain restructuring, Δm_l — photoinduced change.

$\langle 111 \rangle$ axis and was parallel to the external magnetic field. The maghemite samples were prepared by the technology described in Ref. 8 . The content of zinc ions was ~10 wt. %.

In the experiment we measured the change in the component of the magnetic moment in the direction of the external magnetic field. Figure 1 illustrates the time dependence of the magnetization under optical irradiation. In performing the measurements of the magnetic-field dependences, after a new value of the magnetic field is established, drift of the magnetic moment (m_d) is observed (in a time of the order of several minutes). This drift is associated with the rearrangement of the domain structure and serves as a background with respect to which the photoinduced changes were measured. The quantity m_d and its rate of change depend on the thermomagnetic history. When the light pulse is switched on (see Fig. 1c), a signal Δm_l is observed against the background of m_d. The magnitude of this signal saturates within seconds. After the light is switched off, Δm_l relaxes approximately over the same time (see Fig. 1, a and b). The sign of Δm_l depends on the measurement temperature and the magnitude of Δm_l at fixed T depends on the magnetic field and the intensity of the optical pumping. The quantity Δm_l marked in Fig. 1 is virtually independent of the value and rate of change of m_d, and its value at the moment when m_d saturates is the same as its initial value. Switching the light on and off influences the domain rearrangement process and therefore the rate of change of m_d.

Figure 2 shows the temperature dependences, measured in different magnetic fields

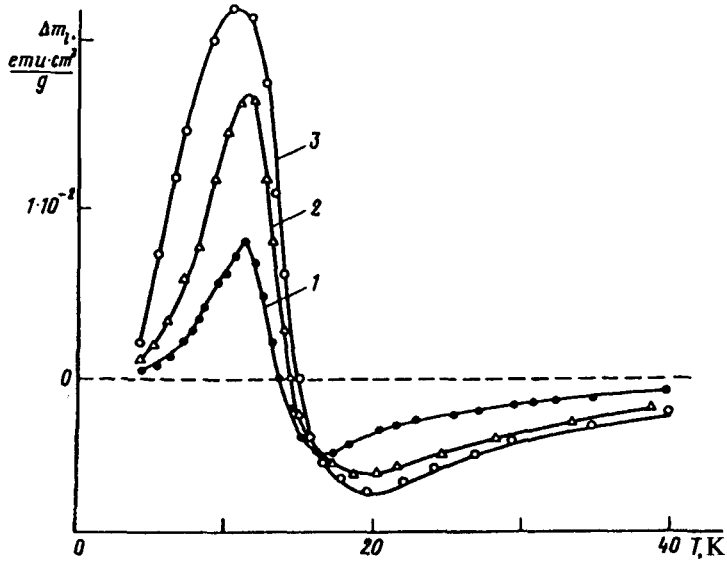

FIG. 2. Temperature dependences of the photoinduced change of the magnetization: *1* — $H=100$ Oe, *2* — $H=300$ Oe, *3* — $H=500$ Oe. Power density of optical radiation $P=0.72$ W/cm^2.

(the accuracy of the measurements falls within the size of the symbol in the plots), of the photoinduced part of the change in the magnetic moment. As one can see from the figure, as the temperature changes, the sign of the effect changes; this is a new feature of photomagnetism in oxide spinels. The temperature at which the sign changes and the temperature at which Δm_l assumes extremal values depend on the magnitude of the magnetic field. The ratio of the quantities Δm_l at temperatures corresponding to the maximum positive value to the maximum negative value is approximately 3:1. The photoinduced changes of the magnetization in magnetic fields $H \leqslant 500$ Oe are observed only at low temperatures, and at $T \geqslant 60$ K the magnitude of the signal is comparable to the noise level.

It is well known[9] that the γ modification of "pure" iron sesquioxide is a metastable phase with tetragonal symmetry and ratio of the axes $c/a=3$. On heating this phase easily transforms into the α phase. In the presence of impurity ions, a rearrangement of the structure characteristic of "pure" γ-Fe_2O_3 is possible. In our case we were able to stabilize the γ phase by introducing zinc ions. The single crystals grown have a spinel structure with $a=8.33$ Å . Taking into account the content of zinc ions in the sample, the formula for the compound obtained can be tentatively written as $Fe_8^{3+}[Fe_{12}^{3+}Fe_{2-x}^{2+}Zn_x^{2+}\square_2]O_{32}$ (Ref. 9), where $x=0.54$, and $\square$ denotes an iron vacancy. This compound with a defective spinel structure contains divalent iron ions, which, as is well known,[10] are responsible for the susceptibility of many nonstoichiometric compounds to illumination.

If it is assumed that the strongly anistropic Fe^{2+} ions occupy octahedral positions

and their local axes of quantization are directed along threefold axes, then for a spinel structure these axes coincide with the four crystallographic directions of the type $\langle 111 \rangle$, which are the easy-magnetization axes of the crystal. In addition, for the given geometry of the experiment there exists a preferred axis and the three other axes are equivalent. The 3:1 ratio of the extremal values of Δm_I can be easily explained by assuming that the Fe^{2+} ions are distributed uniformly over the octahedral positions, which are the positions that respond to irradiation. The high-temperature feature in the region $T \sim 20$ K is due to ions (type I) whose quantization axis coincides with the direction of the external magnetic field and for which optical radiation induces a redistribution of the populations of the levels such that the component of the magnetic moment of a Fe^{2+} ion on the local quantization axis decreases. Because of the exchange coupling with the magnetic environment, the corresponding component of the magnetization of the crystal decreases. The low-temperature feature is related to a group of (type-II) centers, for which the quantization axes are directed at an angle to the direction of the external magnetic field. These axes produce an anisotropy which competes with the anisotropy due to type-I centers. Analysis of the temperature dependences of the form shown in Fig. 2 indicates that as the magnetic field increases, the quantity $|\Delta m_I|$ increases as a result of emptying of the high-lying levels in a cycle of optical absorption and relaxation; i.e., photoinduced cooling of the spin subsystems of the photocenters occurs. It is obvious that the energy splitting between the active levels for the group of type-I centers is larger than for the group of type-II centers. This can serve as an additional argument for a subsequent choice of a model of a photosensitive center.

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