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Polarization-dependent photoinduced change of magnetic state in α-Fe$_2$O$_3$:Ga, Yb crystals

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Abstract. The experimental investigation of the polarization-dependent photoinduced changes of the magnetic state in the haematite crystals doped with ytterbium and gallium ions is presented. The method of antiferromagnetic resonance was used. A detailed study of the kinetics and temperature changes of the photoinduced shift of the magnetic resonance field is made.

1. Introduction

The creation of nonequilibrium metastable states in magnets by optical influence is a rather tempting way to control the magnetic properties. In addition it is possible with light selectively to change the magnetic state of ions which are responsible for the magnetic properties of the materials. The photoinduced changes can occur due to either the transition of single centres in an optically excited state [1, 2], or a co-operative effect [3], when the reconstruction affects the whole cluster. In a number of cases the change of the magnetic properties depends not only on the energy of an optical quantum, but also on the light polarization. MnF$_2$ [4], Y$_3$Fe$_5$O$_{12}$:Si [5] and Ca$_3$Mn$_2$Ge$_3$O$_{12}$ [6] limit the series of magnets which reveal the polarization-dependent photomagnetic effect, and the search for new photosensitive materials continues. By the antiferromagnetic resonance (AFMR) method we have revealed that the weak ferromagnetic crystals α-Fe$_2$O$_3$:Ga exhibit polarization-dependent changes of the magnetic properties when they are doped with the Eu and Yb rare earth ions. For the haematite crystals containing europium ions, the effect has been investigated in the whole area of its existence ($T \leqslant 70$ K) [7], and for the crystals with the ytterbium ions only in the $80 \leqslant T \leqslant 130$ K temperature range [8]. In the last case distinctive features (in contrast to the crystals with europium) are the dependence of the AFMR line width on the polarization of the optical radiation and a change of sign of the resonance field shift (δH_r) with the change of the polarization.

In the present paper we report the temperature and the temporal behaviour of the photoinduced changes of the magnetic state in α-Fe$_2$O$_3$:Ga, Yb crystals by the AFMR method.

2. Experiment

The α-Fe$_2$O$_3$:Ga, Yb crystals were grown by spontaneous crystallization from a solution in a melt with bismuth–sodium solvent. The content of the gallium ions was 5 at.%, and they were added as before, to extend the temperature range of the weakly ferromagnetic phase.

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The ytterbium ion content was 0.005 at.%. The samples consist of slabs with thickness $t \approx 0.2$ mm and average transverse size $d \approx 2$ mm. The magnetic field lay in the basal plane of the sample and was perpendicular to the optical beam. The light source was an incandescent lamp. The sample was placed inside a resonant cavity for the microwave frequency $f_{UHF} = 35$ GHz and the TE_{011} mode.

3. Results and discussion

First of all we note that the α-Fe_2O_3:Ga, Yb crystals, in contrast to the crystals doped with the europium ions, show a strong anisotropy of the resonance field. It is the hexagonal and has a value of ~1 kOe, when the irradiation is absent (figure 1). As the temperature of the sample decreases from 300 K to 4.2 K, the intensity of the magnetic resonance absorption is decreased approximately by two orders of magnitude. This fact suggests the change of the magnetic moment component in the crystal basal plane.

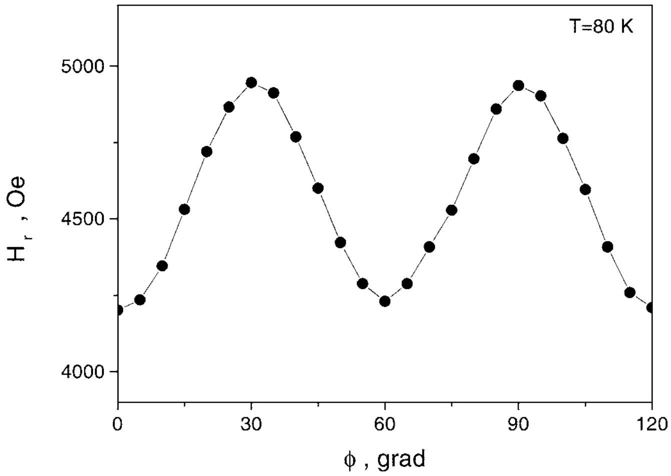

Figure 1. Angular dependence of the resonance field in the basal plane of the α-Fe_2O_3:Ga, Yb crystal. $T = 80$ K.

Figure 2 shows the temporary changes of the resonance field shift (δH_r) as a function of the light polarization at various temperatures. The sections I and III correspond to the cases when the crystal is irradiated with light with electric-field vector $\boldsymbol{E}$ accordingly parallel to the external magnetic field $\boldsymbol{H}_0$ ($\boldsymbol{E} \parallel \boldsymbol{H}_0$) and perpendicular to $\boldsymbol{H}_0$ ($\boldsymbol{E} \perp \boldsymbol{H}_0$). The sections II and IV correspond to a free relaxation of the resonance field value when the light is switched off. It is seen, as the temperature increases from 8 K up to 80 K, that value of the photomagnetic effect ($(\delta H_r)_{max}$) drops approximately by an order of magnitude. Besides, when the light with $\boldsymbol{E} \parallel \boldsymbol{H}_0$ is switched off, the residual quantity $(\delta H_r)_{res}$ decreases proportionally to $(\delta H_r)_{max}$, but it does not reach zero. The irradiation with polarization $\boldsymbol{E} \perp \boldsymbol{H}_0$ changes the sign of the shift of the resonance field, but $(\delta H_r)^{\perp}_{max} \neq (\delta H_r)^{\parallel}_{max}$. The system returns to the initial state ($\delta H_r = 0$) only from the state induced by the radiation with the polarization $\boldsymbol{E} \perp \boldsymbol{H}_0$. In contrast to the crystals doped with the europium ions in this case the free relaxation is not described by a single-exponential dependence and does not obey the Arrhenius law.

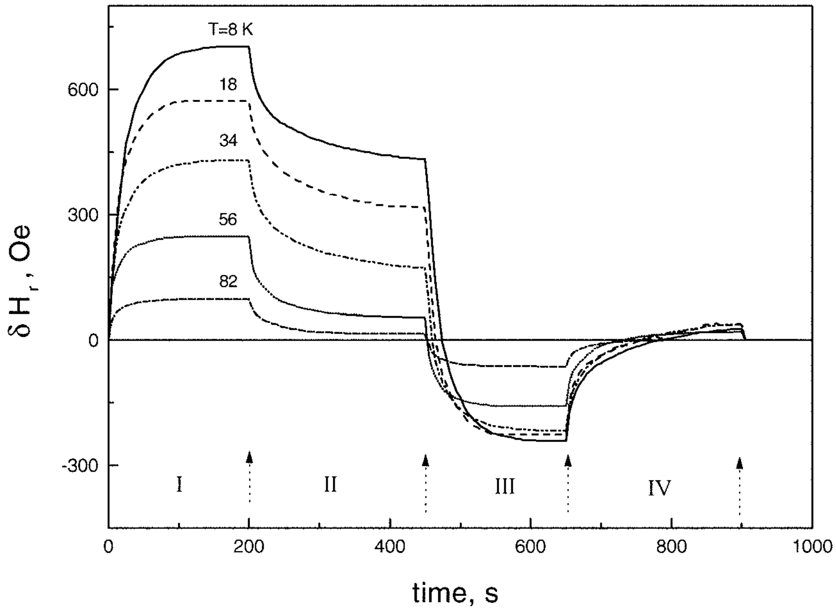

Figure 2. Time dependence of the resonance field shift as a function of the irradiation at various temperatures. $\varphi = 30°$.

The elementary relation, which correctly reproduces the temporary behaviour of (δH_r) in each of the sections in figure 2, is a double-exponential dependence of the form:

$$|\delta H_r| = A_1\{a + b\exp(-t/\tau_1)\} + A_2\{a + b(\exp(-t/\tau_2)\} \tag{1}$$

where $a = -b = 1$ for section I and $a = 0, b = 1$ for all remaining sections. For sections I and III it is possible to understand such behaviour if we assume the generalized three-level model of the photosensitive centre for a cycle 'an optical absorption—optical relaxation'. For sections II and IV (the free relaxation) either the use of a three-minimum potential or supposition about several non-equivalent photocentres is required.

In figure 3 the temperature dependences of the amplitudes $(A_j,\ j = 1, 2)$ and the characteristic times (τ_j) with the double-exponential approximation (1) for each section in figure 2 are shown. We can notice that the dependences of the parameters in sections I and III (parts (a) and (c)) and also in sections II and IV (part (b) and (d)) are qualitatively similar among themselves. Taking into consideration these results we can suppose that the characteristic altitudes of the energy barriers, separating different induced states, are close to one another in value.

As seen from figure 4 the values of the photoinduced shifts of the resonance field (δH_r) for the various optical polarizations depend on the magnetic field direction in the crystal basal plane. For polarization $\boldsymbol{E} \parallel \boldsymbol{H}_0$ the value of the effect is maximum when the magnetic field $\boldsymbol{H}_0$ is along the hard magnetization direction in the crystal, whereas for the magnetic field applied along the axis of the easy magnetization $(\delta H_r)^{\parallel}_{max}$ decreases and $(\delta H_r)^{\perp}_{max}$, in contrast, increases.

To interpret the experimental data the knowledge of the structure of the photosensitive centre is necessary. As established earlier [8], the temperature dependences of the magnetic resonance parameters without the optical irradiation have no features attributed to Fe^{2+}, which appear in the crystals because of the oxygen nonstoichiometry. Evidently, in α-Fe_2O_3:Ga, Yb

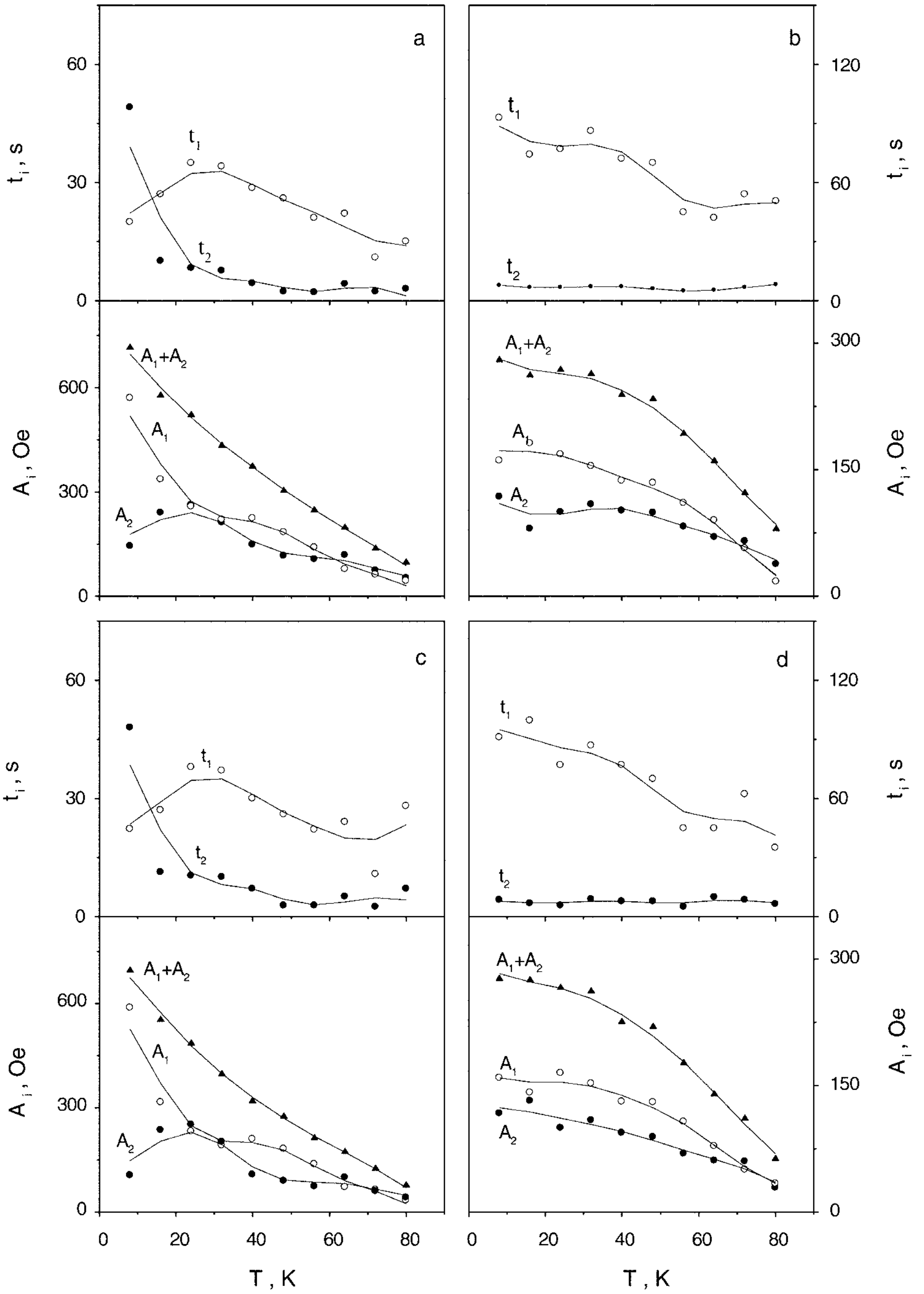

Figure 3. Temperature dependences of the parameters of photoinduced shifts of the resonance field with a double-exponential approximation (see (1)), for each section in figure 2(a) I, (b) II, (c) III, (d) IV.

the oxygen deficit is compensated by the appearance of the two-valent ytterbium ions. In the present case the formation of a defect consisting of an anion vacancy and the ytterbium ions

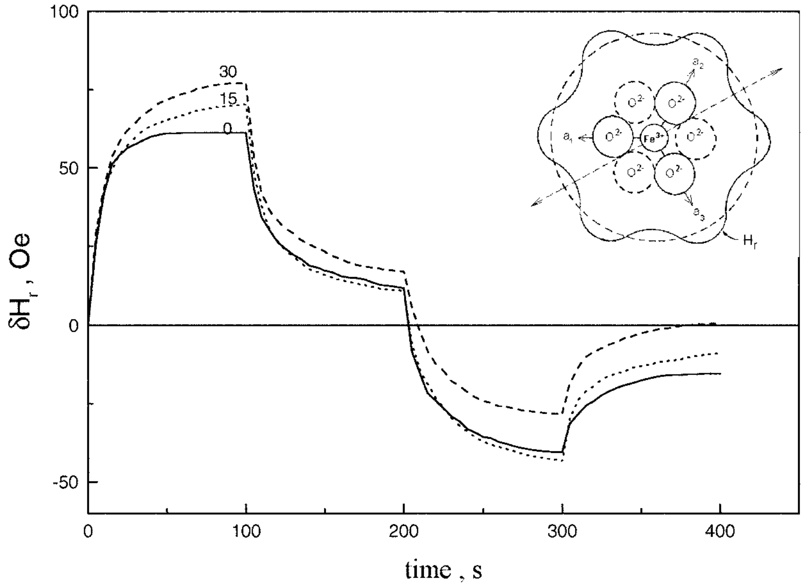

Figure 4. Time dependences of the resonance field as a function of optical polarization for various directions of the magnetic field in the basal plane of the sample. The inset illustrates the dependence of the resonance field with respect to the crystalline structure without light [11]. $T = 80$ K.

located near one is probable. The cation vacancy formation is improbable since the haematite crystals doped only with europium and ytterbium ions have photoinduced properties, though we checked the influence of almost all rare earth ions. In respect of the europium and the ytterbium ions it is well known [9] that they easily exhibit both valency 3 and 2. Thus the initial state of the photocentre, which has not been subject to the optical effect, can be either (1) $2Yb^{2+}+\boxed{O^{2-}}$ or (2) $2Yb^{3+} + F'$, where $\boxed{O^{2-}}$ is the oxygen vacancy and F′ is the centre consisting of two electrons captured in a potential field of the oxygen vacancy. Thus, as a result of the optical radiation the photocentre transformation can be the following:

$$2Yb^{2+}+\boxed{O^{2-}} \;\underset{\longleftarrow}{\overset{\longrightarrow}{h\nu}}\; Yb^{3+}+Yb^{2+}+F' \qquad (2a)$$

or

$$F'(S=0) \;\underset{\longleftarrow}{\overset{\longrightarrow}{h\nu}}\; F'(S=1) \qquad (2b)$$

when the F′ centre passes through the optically excited state from a singlet state into a triplet one. It has been proved [10] that the ground state of the F′ centre is the singlet and in the oxygen compounds the singlet and triplet are closely located. It is easy to see from the inset in figure 4 that the oxygen vacancy appearance results in the local distortion of the structure. The distortion axis coincides with the hard magnetization direction of the α-Fe_2O_3 crystal. Just this axis (Z') will be the local axis of quantization of the photocentre. As a result of the electronic selection rules for the F′ centre the optical transitions are induced by light with the vector of the electrical field either along the centre axis or perpendicular to one [10]. The irradiation by light with the polarization $\boldsymbol{E} \parallel \boldsymbol{H}_0$ always increases the value of the resonance field, and with the polarization $\boldsymbol{E} \perp \boldsymbol{H}_0$ it reduces it. From our experiment it follows that the relation $(\delta H_r)^{\parallel}_{max}/(\delta H_r)^{\perp}_{max}$ for the magnetic field in the directions of the hard ($\varphi = 30°$) and easy ($\varphi = 0°$) magnetization in the basal plane gives values 1.5 and 1.18, accordingly.

Using the inset in figure 4 we have calculated the total projections of the light vector $\boldsymbol{E}$ on the local axes of the centres for the longitudinal and transversal polarization. Their relation gives the comparable values 1.18 and 1.4, but in contrast for the cases when the magnetic field $\boldsymbol{H}_0$ is along the easy and hard magnetization directions, respectively. These values are proportional the number of centres involved in the transitions with $\boldsymbol{E} \parallel \boldsymbol{H}_0$ and $\boldsymbol{E} \perp \boldsymbol{H}_0$. This apparent inconsistency is removed if we assume that the magnetic moment of the photocentre ‘feels’ not the magnetic field itself but the effective field created by one of the magnetic sublattices of the crystal matrix. This effective field is perpendicular to the direction of the magnetic field. From here it follows that the photosensitive centre can be considered as a quasi-Ising ion with the quantization axis directed along the axis of the local distortion. Then under the irradiation the magnetic anisotropy of the dopants either decreases or increases depending on the light polarization. The change of the magnetic interactions occurs for both models of the photocentre above. In fact, there is either origin of the strong anisotropic magnetic centre due to the occurrence of the Yb^{3+} ion (scheme (2a)) or transition of the F' centre in the magnetic state with the spin $S = 1$ (scheme (2b)).

The temperature behaviour of the photoinduced changes (figure 3) is determined by two processes: (1) the light changes the magnetic energy of the centres as a result of the increasing of the population of the high-lying energetic levels and (2) changes of the number of photomagnetic centres occur because of the thermal relaxation. The non-monotonic character of the dependences of A_j in figure 3 suggests a competition of the mechanisms.

For a more detailed description of the observed effects of the spectral investigation of the photosensitivity, study of the photoinduced change of the anisotropic magnetic properties and also their dependences on the optical radiation power are necessary.

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