Photoinduced structural and orientational magnetic phase transitions in $FeBO_3$:Ni

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The temperature dependence of the ellipticity ψ and the angle of Faraday rotation ϕ have been studied for the case where the measuring beam ($\lambda = 0.525\,\mu$m) propagates almost along the threefold symmetry axis and the magnetic field is applied in the basal plane of the sample. Two types of curves were measured: 1) $\psi^*(T)$ and $\phi^*(T)$ for a cooled sample irradiated with an additional source with $\lambda > 0.75\,\mu$m; 2) $\psi(T)$ and $\phi(T)$ without the additional irradiation. The differences between the two types of curves, $\Delta\psi(T) = \psi^* - \psi$ and $\Delta\phi = \phi^* - \phi$, exhibit critical behavior, leading to the conclusion that structural and orientational magnetic phase transitions occur simultaneously. It was found that the transition temperature depends on the orientation of the magnetic field and the intensity of the irradiation. The dependence on the intensity is well approximated by the function $T = 158\exp(-0.05/I^{1/3})$, where I is the intensity of the irradiation and the numbers 158 and 0.05 are fitting parameters. The results are interpreted by the phenomenological theory of phase transitions with the use of a two-component order parameter and a model of photoinduced interacting centers, with a long-range interaction potential that falls off exponentially with distance.

The photoexcitation of solids often induces a variety of new physical properties that are due to the generation of exciton states whose influence on the spectrum of a solid is comparable to that of impurity states. The phenomena that are of the greatest scientific and technological interest are those of a cooperative nature, since in these cases the response of the systems to optical excitation is large. These collective effects are produced when the external variable parameters (intensity, temperature, or both together) take on critical values, and, by analogy with equilibrium thermodynamics, have been called optically-induced phase transitions. Such phase transitions occur in systems far from equilibrium[1] and in systems in quasiequilibrium, where the number of optically excited atoms is small and all the states can be described approximately by a Boltzmann distribution of the order parameter.[2]

In magnetically ordered media the phenomena of the first type are represented by the laser excitation of new magnetic states in MnF_2, $EuCrO_3$ (Refs. 3,4), which have been observed at high intensities of the incident light. Phenomena of the second type include effects that are due to the thermalization of the excitation through specially synthesized impurity states, and have so far been studied in detail only for the case of $Y_3Fe_5O_{12}$:Si, where the photoexcitation reorients the magnetic moment to a new axis that is induced by the magnetic anisotropy through redistribution of anisotropic electron states that have spatial degrees of freedom.[5]

Theoretical treatments of the photoinduced anisotropic phenomena in magnets are based on the idea of noninteracting photosensitive anisotropic centers.[6,7] The centers that have been used have a very wide range of composition, symmetry, etc, and their common property is that they do not interact with each other. A large amount of experimental data has been collected, which fit well into the framework developed on the basis of these ideas, at least in the cases of yttrium iron garnet (YIG) and $CdCr_2Se_4$. By analogy, these ideas were used to interpret the experimental results obtained during irradiation of iron borate samples, prepared as nominally pure single crystals or single crystals doped with various elements.[8]

However, the spectral dependences of the effects, which for YIG were impurity related, for the iron borates were due to excitation of the matrix ions. Therefore a model was proposed[9] for the induced anisotropic properties, based on the interaction of impurity or defect states with the photoexcited states, resulting in ordering of the impurity or defect states and the appearance of uniform anisotropy. The experimental results obtained for samples $FeBO_3$:Ni, which exhibit the highest photosensitivity, are in quite good agreement with these ideas; hence it follows that the magnitude of the anisotropic effects resulting from the competition between the entropy and the ordering terms is proportional to the Boltzmann factor, or, in the high-temperature approximation, proportional to $1/T$.

The first signs that the phenomenon were more complicated were found in the work by Chzhan *et al.*,[10] where the temperature dependence of the initial susceptibility below the Néel temperature T_N was discovered to be nonmonotonic, a result due to the presence of impurity or defect centers and their effect on the magnetic structure. Moreover, more recent data indicate the possibility of photoinduced enhancement of the interaction of the impurity states through magnetoelastic waves,[11] which leads to the formation of a modulated state in this crystal. It is therefore an important matter to undertake temperature-dependent investigations of effects that indicate reliably the presence of structural or magnetic transformations in a medium, one such effect being optical anisotropy, which undergoes a transformation in both cases.[12] The additional contribution to the birefringence that appears at low temperatures can be due either to changes in the magnetic system, which shows up in the Cotton–Mouton effect, or to structural distortions that lower the symmetry of the crystal. These two effects can also appear simultaneously.

Since this problem is of a fundamental nature, to solve it we have developed a special method by which one can sepa-

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rate the lattice and the magnetic contributions and analyze them separately. The experimental facts obtained in this approach indicate the presence of both a structural and a magnetic phase transition, which are described from a common thermodynamic point of view, so that the calculated and experimental temperature dependences are found to be in satisfactory agreement.

METHOD

It is well known[13] that when rhombohedral crystals undergo magnetic ordering with weak ferromagnetism they become optically biaxial, but because the angles between the optical axes are very small there is virtually no anisotropy in the cross section of the optical ellipsoid cut by a plane perpendicular to the C_3 symmetry axis. Consequently, we can write

$$\Delta n_{x,y}=\Delta n^m, \quad Z\|C_3, \tag{1}$$

where Δn^m is the magnetic contribution to the birefringence. However, a change in the birefringence can be due both to changes in the lattice contribution Δn^s and the magnetic contribution, so that

$$\delta\Delta n=\delta\Delta n^s+\delta\Delta n^m. \tag{2}$$

According to Ref. 14, the rotation ϕ of the plane of polarization and the ellipticity ψ induced by magnetooptical effects in light travelling in a direction close to the optic axis of a rhombohedral crystal can be written

$$\phi=al^2\sin 2(\theta-\varphi)+m[b\beta\sin(\gamma-\varphi)+c\sin 3(\varphi+\varphi_0)], \tag{3}$$

$$\psi=a'l^2\sin 2(\theta-\varphi)+m[b'\beta\sin(\gamma-\varphi)+c'\sin 3(\varphi+\varphi_0)]. \tag{4}$$

respectively. Here m and l are the moduli of the ferromagnetic and antiferromagnetic moments, a,a' b,b' and c,c' are the magnetooptical constants, β is a coefficient that depends on the angle between the direction of propagation of light and the optic axis, θ is the azimuth of the initial polarization of the light, γ defines the orientation of the axis about which the crystal is tilted away from normal propagation of the light, φ_0 defines the orientation of the twofold symmetry axis, and φ defines the orientation of the ferromagnetic moment in the basal plane. It has been established experimentally that for $\lambda=525$ nm, $a\gg a'$, and if the crystal is also positioned normal to the measuring beam, i.e., $\beta=0$ we have from Eq. (4)

$$\psi=a'l^2\sin 2(\theta-\varphi). \tag{5}$$

The small changes $\Delta\psi$ in the ellipticity under these conditions can be attributed to changes in the following parameters:

$$\Delta\psi=2a'l^2\cos 2(\theta-\varphi)\Delta\varphi+2a'l\Delta l\sin 2(\theta-\varphi)+\Delta a'l^2\sin 2(\theta-\varphi). \tag{6}$$

According to this expression the magnetic contribution to the change in the ellipticity may be due to a change Δl in the modulus of the antiferromagnetism vector, a change $\Delta a'$ in the magnetooptical constant, or a change $\Delta\varphi$ in the orientation of the antiferromagnetism vector. By means of specially designed experiments we were able to determine that within experimental error the intensities of the optical excitation used in subsequent experiments did not change l or a'. This result is evidence that variations in the orientation of the antiferromagnetism vector and structural transformations of the lattice play the primary roles in the externally induced changes in the ellipticity of transmitted light. For $\theta-\varphi=45°$ the magnetic contribution is insignificant, and hence in this geometry of light propagation the changes in $\Delta\psi$ allow one to follow just the response of the crystal structure to the external agent.

Returning now to Eq. (3), if $\theta-\varphi=0$ and $b\beta\gg c$, then the latter condition is satisfied even for small inclinations of the sample, the rotation of the plane of polarization is

$$\phi=b\beta m\sin(\gamma-\varphi), \tag{7}$$

and its variation will be described by the following expression

$$\Delta\phi=b\beta\sin(\gamma-\varphi)\Delta m+b\beta m\cos(\gamma-\varphi)\Delta\varphi+\beta m\sin(\gamma-\varphi)\Delta b. \tag{8}$$

The most important cause of a change in ϕ for $\gamma\approx\varphi$ is a change in the orientation of the ferromagnetism vector; that is, if the crystal is tilted about an axis that coincides with the azimuth of the ferromagnetism vector, then the variation in the Faraday effect will mainly reflect only the variation in this azimuth.

Therefore, by means of polarization-dependent measurements of the parameters of the light transmitted through the sample one can independently follow small externally-induced changes in the structural and the magnetic parameters of the samples.

EXPERIMENTAL RESULTS AND DISCUSSION

We have used the experimental method and geometry of light propagation described above to determine the temperature dependence of the ellipticity $\psi(T)$ and rotation $\phi(T)$ of the plane of polarization of light of wavelength $\lambda=525$ nm transmitted though $FeBO_3$:Ni samples in the absence of "active" irradiation and have also determined the analogous quantities $\psi^*(T)$ and $\phi^*(T)$ with supplemental irradiation of the sample in the wavelength range 800 to 1000 nm with an intensity $I\approx 0.1$ W/cm^2. The difference curves $\Delta\psi(T)=\psi^*-\psi$ and $\Delta\phi(T)=\phi^*-\phi$ are shown in Figs. 1 and 2, respectively. According to the above discus-

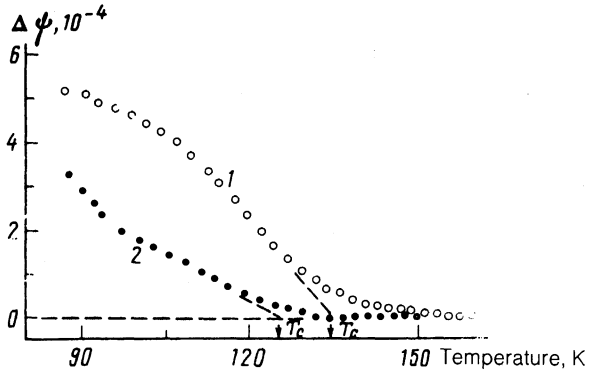

FIG. 1. Temperature dependence of the photoinduced ellipticity of a measuring beam transmitted through a $FeBO_3$:Ni sample along the optical axis (the C_3 axis). ■) Magnetic field $H=100$ Oe applied in the direction of the twofold symmetry axis. Scale of the curve reduced by a factor of two. +) magnetic field $H=100$ Oe applied in the basal plane perpendicular to the C_2 axis.

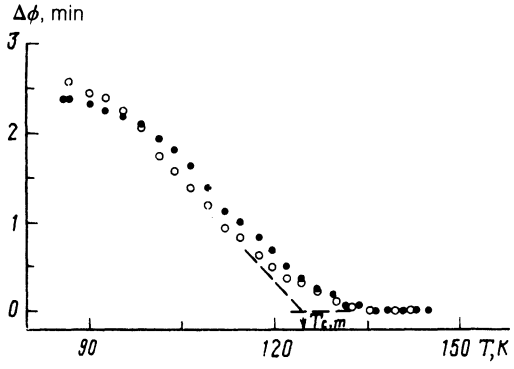

FIG. 2. Temperature dependence of photoinduced rotation of the plane of polarization of the measuring beam transmitted through the $FeBO_3$:Ni sample at a small angle to the optical axis. ■ and +) Magnetic field $H = 100$ Oe and 40 Oe, respectively, applied to the basal plane perpendicular to the C_2 axis. $\Delta\phi = 2.5'$ corresponds approximately to 3 deg of tilt of the ferromagnetic moment from the direction given by the external magnetic field.

sion, the photoinduced changes in the ellipticity and rotation of the plane of polarization reflect the temperature-related behavior of the lattice distortions and the temperature-induced variation in the azimuth of the ferromagnetism vector.

Let us consider in more detail the characteristic features of Fig. 1. The variation of the ellipticity, which is proportional to the variation of the linear optical anisotropy of the crystal, becomes appreciable when the temperature of the sample is lowered a certain amount below a critical temperature T_c, and it exhibits a threshold dependence $\propto (T - T_c)$. Over a considerable temperature range below T_c the dependence can be well approximated by a linear function of the temperature. The threshold temperature and the slope of the curves relative to the temperature axis depend on the direction of the external magnetic field, which orients the ferromagnetism vector in the basal plane relative to the C_2 symmetry axis. When the magnetic field is along the twofold symmetry axis the temperature T_c and the rate that the optical anisotropy increases with the temperature are higher than when the magnetic field lies in the basal plane orthogonal to the C_2 symmetry axis.

Let us consider the reasons for the appearance of optical anisotropy below T_c when the sample is given supplementary irradiation. Because of the experimental conditions that were chosen, the measured ellipticity can be due only to structural changes. Since Δn was vanishing small, $n_x = n_y$, before the irradiation, a proportional change in the refractive index resulting from the lowering of the symmetry with no change in the crystal system could not produce any appreciable birefringence. We must therefore conclude that the optical anisotropy is due to the tilting of the optical axis relative to the C_3 symmetry axis. This situation can be realized only if the symmetry of the crystal is reduced from trigonal (optically uniaxial) to a lower symmetry (optically biaxial). From general arguments, the resulting symmetry must be the intersection of the initial symmetry group with the symmetry of the external influence. However, in the present case, the external influence is isotropic (unpolarized photoexcitation). Therefore, the change in the point group that occurs can be considered as spontaneous—a result of internal degrees of freedom. This cause of the change Δn, as well as the critical nature of its temperature dependence, is evidence for a photoinduced structural transition with temperature in samples of $FeBO_3$ with Ni impurities. These latter factors must also be involved in the mechanism of the crystal lattice transformation, in which prior to the photoexcitation the impurity states are disordered, while with the photoexcitation they become ordered at temperatures below the critical temperature. According to previous ideas,[9,11] the impurity ions form anisotropic oxygen-nickel complexes, with a degree of ordering that depends on the direction of the ferromagnetism vector relative to the twofold symmetry axis. During the photoexcitation an interaction, which is somewhat greater when the ferromagnetism vector is aligned along the C_2 axis, arises between these complexes, which have orientational degeneracy, and in this case both the slope of the temperature dependence relative to the temperature axis and the critical temperature will be greater when the magnetic field is parallel to the C_2 axis: $H \| C_2$.

Let us turn now to Fig. 2, which shows the temperature variations of the Faraday rotation that results from photoexcitation of the sample, and which, according to Eq. (8), is due to a change in the orientation of the ferromagnetism vector in the basal plane relative to the direction of the applied field. Over a wide temperature range these curves can be approximated by a linear function of the temperature with a threshold temperature $T_{c,m}$ that does not depend on the external magnetic field and is close to the structural transition temperature T_c. A characteristic fact that bears on these experiments is that there is no change at all in the orientation of the ferromagnetism vector when this vector is aligned with the C_2 symmetry axis. Hence this vector is reoriented to a new direction when the temperature of a photoexcited $FeBO_3$:Ni crystal with the direction of the ferromagnetic moment normal to the C_2 axis is lowered below the threshold temperature $T_{c,m}$. When the ferromagnetism vector is aligned with the C_2 axis the equilibrium direction remains unchanged.

It follows from the general theory of orientational transitions[15,16] that the process described above can occur when there is a competing anisotropic interaction, and hence in the simplest case this result can be considered, in addition to the crystallographic anisotropy, as another form of light-induced magnetic anisotropy, whose easy magnetization axis does not coincide with the crystallographic axis in the first case and either coincides with it or is normal to it in the second case. By means of visual observations it was determined that in the second case, when the temperature is below the transition temperature a decrease in the magnetic field is accompanied by a symmetric reorganization of the magnetic moment with the formation of a stripe magnetic structure. This observation is evidence that when the ferromagnetic moment is oriented along the C_2 axis, the easy axis of the induced anisotropy is normal to that axis. By use of the first case ($H \perp C_2$) one can estimate the value of the induced anisotropy field H_k as being > 100 Oe and the deviation of the direction of this anisotropy relative to the C_2 axis, which is of the order of 10°. For this estimate we used very simple arguments involving the energy extremum of the sample with a uniform magnetic moment in an external field and the corre-

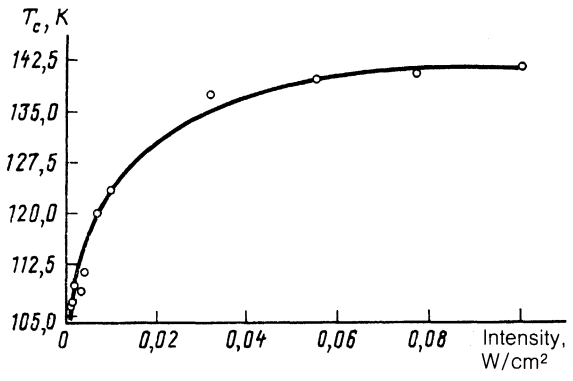

FIG. 3. Dependence of the transition temperature on the intensity of the photoexcitation.

sponding anisotropy fields, as well as the fact that the rotation of the magnetic moment was about 3°, as calculated from experimental data on the Faraday effect.

If, therefore, it is assumed that the photoexcitation causes an interaction between impurity complexes, then the observed phenomenon can be interpreted as a symmetry-lowering phase transition, which, for $\mathbf{M}\perp\mathbf{C}_2$ is accompanied by magnetic-orientation phase transition. The orientation of the magnetic field has an influence on the transition temperature ($T_c = 125$ K for $\mathbf{H}\perp\mathbf{C}_2$ and $T_c = 135$ K for $\mathbf{H}\|\mathbf{C}_2$), but its magnitude does not. Moreover, it has been determined that the intensity of the active photoexcitation influences the critical temperature. This latter dependence is shown in Fig. 3, and is readily approximated by an exponential. The fact that the intensity of the irradiation influences the transition temperature is evidence that the magnitude of the interaction between impurity complexes depends on the number of photoexcited ions. According to Rudenko,[17] the critical temperature in disordered systems can be related to the interaction potential between randomly arranged effective spins by the expression

$$T_c = T_0 \exp\left(-\frac{0{,}87}{Rn^{1/3}}\right). \tag{9}$$

Comparing this expression to the calculated curve $T_c = 158\exp(-0.05/I^{1/3})$, which gives a minimum in the rms deviation of T_c from the experimental points and is shown in Fig. 3 by the solid line, it is easy to determine that for the photoexcitation intensities used in the experiment the correlation length of the interaction potential is $R = 365$ Å, and the maximum value of the critical temperature is $T_c = 158$ K. In these estimates it is assumed that the correlation length depends on the intensity according to the expression $R = 17.4(I/n)^{1/3}$ with the concentration of impurity atoms taken to be $n = 10^{19}$ cm^{-3}, as estimated from the amount of nickel oxide in the charge.

Temperature-dependent studies have thus shown that the optical anisotropy of $FeBO_3$ crystals containing impurities subjected to continuous photoexcitation has a critical nature as its temperature is reduced, which is evidence of a structural transformation from D_{3d} to C_2 or C_s, which are permitted by symmetry. This transformation is accompanied by a reorientation of the sublattice moments when the external magnetic field is perpendicular of the the C_2 symmetry axis, while the sublattice moments remain unchanged for $H\|C_2$. The parameters have been determined for the potential that describes the interaction between impurity complexes that arises in the photoexcitation of the matrix ions.

PHENOMENOLOGICAL THEORY

To describe adequately the results obtained we use the thermodynamic theory of phase transitions, under the assumption that the photoexcitation of the crystal results in an increase of the critical temperature, as is the case for the transition into the modulated state.[11] Then the lowering of the initial symmetry from D_{3d} to C_2 or C_s, the specific occurrence of which will be established later, can be described by the two-component order parameter $\eta = (\eta_1, \eta_2)$, whose contribution to the thermodynamic potential is

$$\Phi_\eta = r(\eta_1^2+\eta_2^2)+u_1(\eta_1^2+\eta_2^2)^2+u_2\eta_1^2\eta_2^2. \tag{10}$$

We take into account the magnetic part of the thermodynamic potential by the presence of anisotropy of the antiferromagnetism vector $\mathbf{l} = l(\cos\varphi,\sin\varphi,0)$ with an energy density K and an easy axis along the unit vector $\mathbf{n} = (\cos\varphi,\sin\varphi,0)$, thus giving a magnetic contribution in the form $K(\mathbf{n}\cdot\mathbf{l})$, plus a Zeeman interaction $\mathbf{m}\cdot\mathbf{h}$, where $\mathbf{m}$ is the ferromagnetism vector, $\mathbf{m} = (\mathbf{M}_1 + M_2)/M$ $(\sin\varphi,\cos\varphi,0)$, and $\mathbf{h} = \mathbf{H}\cdot\mathbf{M} = h(\sin\varphi,\cos\varphi,0)$ is the magnetic field. Then, with φ a small quantity, the magnetic contribution is

$$\Phi_\varphi = \Phi_0 + K\varphi^2 + mh\varphi^2. \tag{11}$$

It is easy to show that the interaction potential of the microscopic order parameter η and the macroscopic order parameter φ in the linear approximation in the small parameter φ can be written as

$$\Phi_{int} = \alpha l^2\eta_1\eta_2\varphi. \tag{12}$$

From the condition that the total potential be an extremum and stable, one can find that in addition to the initial phase $\eta_1 = \eta_2 = 0$ and $\varphi = 0$, two asymmetric phases are possible for $T < T_c$

1) $\eta = (\eta_1,0)$ or $(0,\eta_2)$, $\varphi = 0$, $\eta^2 = -r/2u_1$,

(13)

for

$r\leqslant 0$, $u_1 \geqslant 0$, $u_2^* \geqslant 0$, where $u_2^* = u_2 - \alpha^2/4(k+mh)$,

2) $\eta = (\eta_1,\eta_2)$, $\varphi = \alpha r/[2(4u_1+u_2)(k+\mathbf{mh}) - \alpha^2/2]$,

(14)

for

$\eta_1^2 = \eta_2^2 = \eta^2 = -r/(4u_1 + u_2^*)$ and $r < 0$,

$u_1 > 0$, $0 \leqslant u_2^* \leqslant 4u_1$.

The phases that appear below T_c can be compared with those obtained experimentally. The first, with $\eta = \eta_{1,2}$ and $\varphi = 0$, corresponds to the state that is obtained as the temperature is lowered during photoexcitation, when the magnetic field is oriented in the basal plane along the twofold symmetry axis. In this case it is found both experimentally and by calculation that $\Delta n \neq 0$ and $\varphi = 0$. The second phase, where there are structural transformations and a change in the equilibrium orientation of the sublattice moments, corresponds experimentally to the situation where the magnetic field is in the basal plane and normal to the twofold symmetry axis.

Since Δn is usually proportional to the square of the order parameter,[12] the expressions that we have derived imply that the experimentally measured quantities $\Delta\psi$ and $\Delta\phi$ must be linear functions of the temperature, as is quite satisfactorily confirmed by the curves in Figs. 1 and 2. Furthermore, since the constant of proportionality between the order parameter and the temperature is larger in the first case than in the second, this means that the experimentally measured quantities that are proportional to the birefringence (the ellipticity) must be larger over the entire temperature range when the new phase appears in a magnetic field that is oriented parallel to the twofold axis. This conclusion corresponds entirely to the experimental results.

This phenomenological description, which is based on two interacting order parameters, is in good agreement with the majority of the experimental facts. Consequently, we can, on the basis of the experimental results, make an estimate of the constants that figure in the potential that is constructed. Unfortunately, since the measurements of the microscopic order parameter are indirect, this approach does not permit one to determine all the external parameters, but nevertheless the subject is of interest for the purpose of determining the physical meaning of these parameters. From a comparison of the theoretical and experimental curves, it is possible to estimate the ratio of the coefficients $u_1/u_2 = 1$–10, and the anisotropy energy density $K = 5\cdot 10^3$ erg/cm^{-3}, which in order of magnitude corresponds to that previously measured.

CONCLUSIONS

It can thus be concluded that with reduced temperature a structural phase transition takes place in photoexcited iron borate crystals doped with nickel, and this phase transition is due to the photoinduced interaction between oxygen-nickel complexes. This lattice transformation also involves the magnetic subsystem, which undergoes an orientational phase transition, which is related to the appearance of magnetic anisotropy through a coupling with the lattice subsystem. The transition temperature depends on the intensity of the excitation radiation and the orientation of the external magnetic field in the basal plane of the crystal.

We have made an estimate of the correlation length and the maximum critical temperature. We have proposed a phenomenological theory, whose results are in qualitative agreement with experimental results. The components of the microscopic order parameter can be taken to be those introduced in Ref. 9—inequalities in the equally probable concentrations of defects orientationally degenerate in the three equivalent directions and weakly coupled with the magnetic subsystem. The photoexcited ions, interacting with these impurity complexes, produce ordering in them, depending on the direction of the ferromagnetism vector in the basal plane, thereby favoring anisotropy of the populations of these centers. As a whole, the physical picture is analogous to the behavior of effective spins in a field that couples them, and gives a good description of the field-, orientational- and relaxation-dependent experiments. The introduction of a photoinduced interaction between these centers has permitted a description of the experimental data and the temperature behavior of the photoexcited crystals that is adequate to the experimental data, and thus explains completely the phenomenon of photoinduced anisotropic properties in iron borates.

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