

Phase transition to a modulated state in photoexcited FeBO₃:Ni, an easy-plane magnet

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It is shown experimentally that a phase transition to a modulated state occurs in photoexcited magnetically ordered FeBO₃:Ni crystals as the temperature decreases. The transition temperature T_m decreases with increasing magnetic field strength and depends on the angle of the field relative to the basal plane. The magnitude of the wave vector k_0 in the modulated phase is investigated as a function of the external magnetic field H_0 . It is suggested that the transition to the modulated phase is due to a photoinduced indirect interaction among the impurity oxygen-nickel complexes (ONC) mediated by magnetoacoustic waves. The theoretical forms of $T_m(H_0)$ and $k_0(H_0)$ agree closely with the experimental findings.

1. INTRODUCTION

The macroscopic properties (the magnetization, susceptibility, coercive strength, etc.) of magnetically ordered materials generally change only slightly under illumination.¹ However, in a few magnetic crystals photoexcitation gives rise to new properties not present in the ground state. Such crystals include antiferromagnetic MnF₂, in which illumination produces a ferromagnetic moment,² weakly ferromagnetic FeBO₃ doped with nickel, in which light induces a uniform ordering of the impurity states,³ and EuCrO₃, in which intense optical pumping causes a magnetic phase transition.⁴ A novel effect, in which light induces a space- and time-dependent magnetic structure, has recently been reported in nickel-doped iron borate.⁵ Illumination by unpolarized light at $T = 80$ K was found to excite magnetization waves propagating in the basal plane along directions normal to the C_2 symmetry axes. To explain this experimental finding, one must know something about the mechanism for low-frequency excitation of the magnetic system when the latter is perturbed at a frequency much greater than the natural frequencies of the system. The physical mechanisms proposed in Refs. 7 and 8, in which the dynamic destabilization of the magnetic system is taken to be of the moving-domain-wall type, were motivated by the outward resemblance of this phenomenon to the light-induced autowave phenomena familiar in systems far from equilibrium.⁶ However, this approach gives rise to a square-root dependence of the wall velocity on the light intensity, in conflict with the experimentally observed linear dependence.⁹ In addition, such a treatment fails to account for the observed magnitude of the spatial period of the photoexcited magnetization waves.

Alternatively, light-induced changes in the interaction constants could also be responsible for a thermodynamic transition to a modulated phase. Such a transition would generate a Goldstone mode with no energy gap in the elementary excitation spectrum of the crystal (a phason branch), which would manifest itself as a spatial motion of the modulated structure. The velocity would be proportional to the absorbed light power, which is expended in counteracting the frictional forces.

In this context we carried out new experimental studies of FeBO₃ whose purpose was to detect an additional light-

induced temperature-dependent phase transition to the modulated phase. We found that there is a critical temperature T_m below which the ferromagnetic vector oscillates with a nonzero amplitude that increases in much the same temperature dependence as that of the corresponding order parameter during a singular second-order phase transition. We find that T_m decreases with increasing magnetic field, applied along the axis of crystallographic anisotropy in the basal plane, and is sensitive to the angle of the field vector relative to this plane. We account for the experimental results by postulating that a photoinduced interaction among the oxygen-nickel complexes (ONC) mediated by magnetoelastic waves is responsible for the phase transition to the modulated state. We calculate the transition temperature and the amplitude of the wave vector of the superstructure as functions of the external magnetic field. Comparison with experimental data yields a close agreement.

2. SPECIMENS AND MEASUREMENT TECHNIQUE

We analyzed single-crystal iron borate wafers synthesized from solution in a melt to which 0.1 wt. % nickel oxide was added. The wafers were 50–100 μm thick with linear dimensions 2–3 mm. We selected optically homogeneous wafers, which had been annealed in air to eliminate residual stresses. The extra absorption band, shifting the primary absorption edge toward longer wavelengths, served as a qualitative test for the presence of the impurity.¹⁰ The temperature studies were conducted in an optical cryostat; the crystal rested on a copper finger whose temperature could be varied at a constant rate from 80 to 320 K. The spontaneous onset of the modulated state with cooling was detected magneto-optically by measuring the change in the ellipticity χ of the probe beam from a region of the crystal of diameter less than the characteristic half-period of the superstructure. The simple formula

$$\chi \sim \sin 2(\theta - \varphi) \quad (1)$$

relates the ellipticity χ to the azimuthal angle θ of the polarization plane of the probe beam and to the orientation φ (or $\varphi + \pi/2$) of the ferromagnetic (respectively, antiferromagnetic) vector. This relation is valid for light incident nearly parallel to the optic axis of the crystal if the circular dichroism is small compared to the linear birefringence. Since the

latter condition holds at minimum absorption, we employed a weak monochromatic probe beam ($\lambda \approx 520$ nm) which did not appreciably disturb the magnetic state of the crystal. According to Ref. 11, the space and time dependence of the azimuthal angle of the ferromagnetic vector can be approximated by

$$\varphi(x, t) \approx \varphi_1 \sin(\omega t - kx), \quad (2)$$

with $\varphi_1 \approx 10^\circ$. For $\theta \approx \varphi_1$, the change in the ellipticity is therefore proportional to the oscillation amplitude φ_1 of $\varphi(x, t)$. Relations (1) and (2) also imply that χ begins to oscillate with time at the phase transition temperature. The ellipticity was measured automatically by an ellipsometer and plotted on a recorder, from which T_m was deduced to within a few degrees.

The wafers were illuminated by a separate stabilized source of unpolarized light with a large bandwidth $\lambda > 800$ nm. Under these conditions the excitation beam had no effect on the photoreceiver used in the measurements. The illumination intensity was $I < 0.1$ W/cm². The applied magnetic field was generated by Helmholtz coils equipped with stabilized current sources.

3. EXPERIMENTAL RESULTS AND DISCUSSION

We studied the temperature dependence of the oscillation amplitude of the ferromagnetic vector under the following experimental conditions. A magnetic field was applied to the wafer at room temperature, the excitation beam was turned on, and the specimen was cooled at a constant rate to 80 K. The cooling rate was low enough so that the crystal remained in equilibrium with the cold finger, and its magnetic state was steady at any given temperature within the interval investigated. The cooling rate was $dT/dt \approx 20$ K/h in our experiment. Figure 1 shows the time dependence of the ellipticity of the probe beam (1) and crystal temperature (2) as cooling proceeded from 150 to 100 K. We see that there is a temperature T_m at which $\chi(t)$ oscillates; since we have $\chi \sim \varphi$, these oscillations signal the onset of the inhomogeneous magnetic state. Measurements were made on various parts of the specimen, and in all cases the oscillations began at essentially the same temperature T_m , indicating that the transition was spatially coherent. The oscillations grew with

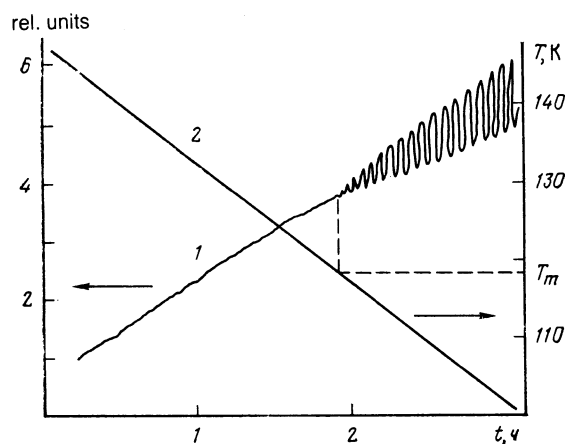


FIG. 1. Time dependence of the ellipticity χ of the probe light (1) and crystal temperature (2). The temperature T_m corresponds to the appearance of modulation in the magnetic birefringence. The χ oscillation period is exaggerated for the sake of clarity.

decreasing temperature, the amplitude rising monotonically from zero when $T \geq T_m$ to finite values below T_m . The oscillation frequency depended only weakly on the temperature but was proportional to the intensity of the excitation beam. If the latter was turned off for $T \leq T_m$, the superstructure stopped moving but the ferromagnetic vector remained modulated for some time thereafter. The relaxation time from the modulated state increased as T decreased and reached ≈ 10 h at $T = 80$ K. The slow relaxation (long "memory") at these temperatures indicates that during illumination the atomic complexes must become ordered, complexes in equivalent positions being separated by rather high potential barriers; moreover, the ordered complexes do not contain optically excited electron states in the matrix material, which have short lifetimes. The nature of the thermal decay of the magnetic superstructure suggests that it proceeds by statistically independent events, i.e., the long-range correlation present in the modulated state becomes vanishingly small after the photoexcitation is discontinued.

Figure 2 shows the temperature dependence of the oscillation amplitude of the ferromagnetic vector in the basal plane. The experimental values (points) are closely approximated by a straight line. According to the phenomenological theory of phase transitions, such a temperature dependence is typical for a secondary order parameter, which interacts with the true order parameter and whose transformation properties completely describe the change in crystal symmetry for $T \leq T_m$. The inhomogeneous magnetic state is thus a consequence of a continuous phase transition, which is accompanied by the formation of a superstructure with a large spatial period. Thus, when a photoexcited FeBO₃:Ni crystal is cooled a magnetic superstructure, in which the oscillations of the sublattice moments have a large spatial period, forms spontaneously against a background of weakly ferromagnetic material. We note that the time required for the order parameter to relax to equilibrium after a change in temperature is very sensitive to the intensity of the excitation beam. When the crystal is cycled from low to high temperature, the temperature dependence $\varphi_1(T)$ exhibits hysteresis if the intensity is decreased; the hysteresis can be suppressed by changing the sample temperature at a lower rate. In addition, the critical temperature T_m drops when the photoexcitation intensity is decreased. These findings indicate that the force restoring the order parameter to its equilibrium value and the temperature T_m are both determined by the photoexcitation intensity.

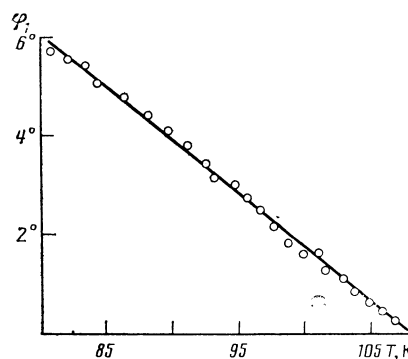


FIG. 2. Amplitude of the oscillations in the ferromagnetism vector versus temperature.

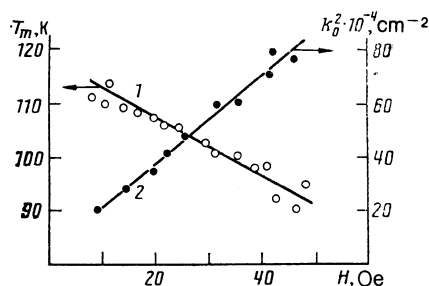


FIG. 3. Critical temperature T_m at which the inhomogeneous state forms (1) and the square of the wave vector k_0 of the state (2) as functions of the magnitude of the external magnetic field applied parallel to the axis of anisotropy in the basal plane.

We investigated the effect of a magnetic field on the formation of the modulated state in an $\text{FeBO}_3\text{:Ni}$ crystal photoexcited by radiation of constant intensity. The magnetic field \mathbf{H}_0 was applied in the basal plane; its intensity was varied, as was its orientation relative to the axes of magnetic anisotropy. Visual observations established that the temperature T_m coincided quite closely with the temperature at which the superstructure began to slip (move). Changing the magnetic field did not appreciably alter the behavior of the temperature curve $\varphi_1(T)$ (Fig. 2). However, the critical temperature T_m and the wave vector k_0 of the superstructure were very sensitive to the field \mathbf{H}_0 . Figure 3 plots the experimental values of T_m (1) and k_0^2 (2) versus magnetic field strength when the field was applied along one of the axes of anisotropy. The figure shows that the magnetic field suppressed the transition to the modulated phase, i.e., T_m decreases with increasing field. The wave vector changes in such a way that k_0^2 depends linearly on the magnetic field.

The behavior of T_m and k_0 was found to be asymmetric when the direction of \mathbf{H}_0 in the basal plane was changed relative to the axis of anisotropy. Figure 4 plots T_m (1) and k_0 (2) as functions of the azimuthal angle α of the magnetic field for the case $|\mathbf{H}| = 30$ Oe. We see that the critical temperature and the spatial period $\lambda = 2\pi/k_0$ increase when \mathbf{H}_0 is rotated toward positive values α , whereas they decrease when the field rotates away from the anisotropy axis in the opposite direction. Recalling the magnetic structure of the modulated state,¹¹ we see that positive angles α correspond to rotations of \mathbf{H}_0 toward the equilibrium direction of the ferromagnetic vector in the superstructure.

Since the uniform magnetic field $\mathbf{H}_0 = (H_x, H_y, 0)$ is unrelated to the order parameter in the modulated phase, it

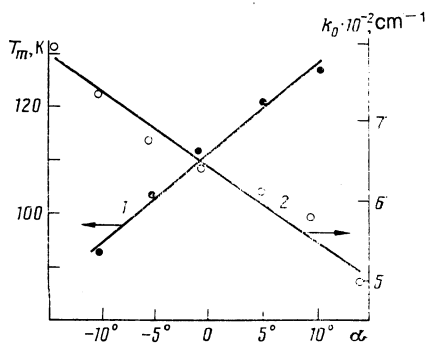


FIG. 4. Critical temperature T_m and wave vector k_0 as functions of the orientation of the magnetic field relative to the axis of anisotropy in the basal plane for $H = 30$ Oe.

should clearly have little influence on the temperature behavior $\varphi_1(T)$. The fact that T_m and k_0 were sensitive to this field thus indicates that the magnetic degrees of freedom play a significant role in the formation of the modulated state.

The experimental results may thus be interpreted as follows. When an $\text{FeBO}_3\text{:Ni}$ crystal is photoexcited, interactions are induced among the oxygen-nickel complexes; these compete with thermal fluctuations and dominate at low temperature, at which the complexes become ordered in a continuous phase transition. The phase transition is marked by the formation of a long-wave magnetic superstructure against a background of weakly ferromagnetic material.

4. MAGNETOELASTIC MECHANISM FOR THE PHASE TRANSITION TO THE MODULATED STATE

Several mechanisms are known that can lead to the formation of magnetic modulated structures in nonmetallic crystals. For instance, the relativistic Dzyaloshinskii-Mori interaction of the type $\mathbf{m} \times (\nabla \times \mathbf{m})$ gives rise to a helicoidal ordering,¹² and experimentally the resulting weak magnetic moment is found to undergo small periodic oscillations. Competitive interactions in the innermost coordination spheres can also result in magnetic modulation. In the long-wave approximation, it is easy to show that the wave vector for the modulated structure satisfies $(ak_0)^2 \approx (J_1 - \gamma J_2)/J_1$, where J_1 and J_2 are the strengths of the competing couplings, the number $\gamma \sim 1$ depends on the crystal lattice structure, and a is the lattice constant. Due to the extraordinarily small period ($\sim 10^{-2}$ cm) of the superstructure in $\text{FeBO}_3\text{:Ni}$, the competing couplings must cancel each other to within one part in 10^9 , which seems implausible. Finally, modulation may also occur when the homogeneous ground state of the system becomes unstable as one of the sample dimensions along the axis of magnetic anisotropy decreases (as in thin magnetic wafers and films); this instability gives rise to a stripe domain structure.¹³⁻¹⁵ However, this mechanism predicts a dependence $k_0^2 \propto H_0^{1/2}$, where H_0 is the external magnetic field; this behavior is not found experimentally.

We propose a new mechanism, in which the superstructure forms due to an indirect oscillating interaction among the oxygen-nickel complexes via the magnetoacoustic modes of the crystal. According to Ref. 10, the Ni ions in an FeBO_3 crystal deform the oxygen tetrahedra locally; owing to the rhombohedral symmetry, these deformations have three equivalent directions. Proceeding in analogy with the pseudospin formalism for particles in a double potential well, we will describe the states of the oxygen-nickel complexes in terms of the Hubbard operators $\hat{X}^{a,b} = |a \rangle \langle b|$, where $a, b = 1, 2, 3$. Experiments¹⁰ show that at $T = 80$ K, the ONC system is paramagnetic in the absence of light. Light excites the iron ions from the state $|S = 5/2, L = 0\rangle$ to the state $|S = 3/2, L = 1\rangle$, which has a nonzero orbital momentum. The ONC's are thus coupled with the spin waves in the matrix material (most likely due to the spin-orbit interaction of excited Fe ions with the ONC's and to the spin-spin interaction between excited and ground-state Fe ions). In addition, we postulate that during photoexcitation the ONC's become coupled to elastic crystal deformations, and the magnitude of the coupling may vary. In the simplest case, these photoinduced interactions lead to pro-

cesses in which spin waves and phonons are absorbed and emitted; the spin waves are described by the operators α_k^+ , α_k with coupling constant g_1 , while the phonons are described by the Bose operators β_k^+ , β_k with magnetoelastic coupling constant g_2 ; these operators correspond to transitions of an ONC from one equivalent position to another, where the transitions are described by the operators $\hat{X}^{P,P\pm 1}$ (here $P = 1, 2, 3$ labels the three possible state of the ONC). We assume a Hamiltonian of the form

$$\begin{aligned} \mathcal{H} = & \sum_{\mathbf{k}} \omega_s(\mathbf{k}) \alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} + \sum_{\mathbf{k}} s k \beta_{\mathbf{k}}^+ \beta_{\mathbf{k}} + b \sum_{\mathbf{k}} k^{1/2} (\alpha_{\mathbf{k}} \beta_{\mathbf{k}}^+ + \text{c.c.}) \\ & + (n/N)^{1/2} \sum_{\mathbf{k}} \sum_{f=1}^N \sum_{p=1}^3 (\hat{X}_f^{P,P+1} + \hat{X}_f^{P,P-1}) [g_1 (\alpha_{\mathbf{k}} + \alpha_{-\mathbf{k}}^+) \\ & + g_2 k^{1/2} (\beta_{\mathbf{k}} + \beta_{-\mathbf{k}}^+)] \\ & \cdot \exp(i\mathbf{k}\mathbf{x}_f) + \sum_{f=1}^N \sum_{p=1}^3 \varepsilon_p \hat{X}_f^{P,P}, \end{aligned} \quad (3)$$

where

$$\begin{aligned} \omega_s(k) &= \omega_0 a (k_d^2 + k^2)^{1/2}, \quad k_d^2 = (\omega_0 a)^{-2} \mu_B^2 (2H_\alpha H_E + H_0 H_D), \\ \omega_0 &= 4\pi S A, \end{aligned} \quad (4)$$

$\omega_s(k)$ is the spin-wave spectrum for iron borate, s is the speed of sound, H_α is the anisotropy field in the basal plane, H_E is the exchange field, A is the exchange interaction, $S = 5/2$, H_D is the Dzyaloshinskii field, b is the magnetoelastic coupling constant, N is the number of oxygen-nickel complexes (of concentration n), and ε_p denotes the tunnel energy levels of the ONC. For simplicity we consider only a single transverse sound wave propagating in the basal plane, since only this wave interacts with the spin waves.¹⁶ After a canonical transformation

$$\alpha_{\mathbf{k}} = \cos \theta_{\mathbf{k}} a_{\mathbf{k}} + \sin \theta_{\mathbf{k}} b_{\mathbf{k}}, \quad \beta_{\mathbf{k}} = -\sin \theta_{\mathbf{k}} a_{\mathbf{k}} + \cos \theta_{\mathbf{k}} b_{\mathbf{k}}, \quad (5)$$

where

$$\operatorname{tg} \theta_{\mathbf{k}} = \frac{s k - \Omega(\mathbf{k})}{b k^{1/2}} \approx \frac{b k^{1/2}}{\omega_s(0)}, \quad (6)$$

the Hamiltonian (1) becomes

$$\begin{aligned} \mathcal{H} = & \sum_{\mathbf{k}} \Omega(\mathbf{k}) a_{\mathbf{k}}^+ a_{\mathbf{k}} + \sum_{f=1}^N \sum_{\alpha=1}^3 \varepsilon_\alpha \hat{X}_f^{\alpha,\alpha} \\ & + \left(\frac{n}{N}\right)^{1/2} \sum_{\mathbf{k}} \sum_{f=1}^N \sum_{\alpha=1}^3 \Lambda(\mathbf{k}) (a_{\mathbf{k}} + a_{-\mathbf{k}}^+) \\ & \cdot (\hat{X}_f^{\alpha,\alpha+1} + \hat{X}_f^{\alpha,\alpha-1}) \exp(i\mathbf{k}\mathbf{x}_f); \end{aligned} \quad (7)$$

here

$$\Omega(\mathbf{k}) = \frac{1}{2} \{ s k + \omega_s(\mathbf{k}) - [(\omega_s(\mathbf{k}) - s k)^2 + 4 b^2 k^2]^{1/2} \} \quad (8)$$

is the frequency of the lower acoustic branch of the magnetoacoustic mode described by the Bose operators $a_{\mathbf{k}}$, $a_{\mathbf{k}}^+$. For small k , (5) and (6) give

$$\Lambda(\mathbf{k}) = [g_1 + g_2 b / \omega_s(0)] k^{1/2}. \quad (9)$$

We have omitted the second branch of the magnetoacoustic modes in Eq. (7), because this branch decays rapidly for

small k owing to its close proximity to the spin-wave modes.¹⁷

The coupling between the oxygen-nickel complexes and the magnetoacoustic waves gives rise to an indirect interaction $V(\mathbf{x})$ among the ONC's, which can be expressed explicitly in terms of the magnetoacoustic Green's function:

$$\begin{aligned} V(\mathbf{x}) &= \frac{1}{2} \sum_{\mathbf{k}} \Lambda^2(\mathbf{k}) \langle (a_{\mathbf{k}} + a_{-\mathbf{k}}^+) (a_{\mathbf{k}} + a_{-\mathbf{k}}^+) \rangle e^{i\mathbf{k}\mathbf{x}} \\ &= \sum_{\mathbf{k}} \frac{\Lambda^2(\mathbf{k})}{\Omega(\mathbf{k})} e^{i\mathbf{k}\mathbf{x}}. \end{aligned} \quad (10)$$

According to (9) the ratio $\Lambda^2(k)/\Omega(k)$ is proportional to $k/\Omega(k)$, and it has a minimum at $k = k_0 \neq 0$ (see Fig. 5). We also note that the speed of sound (equal to $\partial\Omega(k)/\partial k$) has the typical z-shaped dependence for k near k_0 . Although this behavior was observed experimentally¹⁸ in iron borate for $k \approx k_0$, it has remained unexplained until now.

Most of the contribution to (10) comes from the term $\Lambda^2(k)/\Omega(k)$ with $k = k_0$; this implies that the indirect interaction (10) oscillates, and consequently the spontaneous ordering in the ONC system has a spatial period equal to $2\pi/k_0$ (it is not spatially homogeneous). This can be seen by considering the static susceptibility of the system (7) (Ref. 19):

$$\chi^{-1}(\mathbf{k}) = \chi_0^{-1} - \Lambda^2(\mathbf{k}) D_0(\mathbf{k}). \quad (11)$$

Here $\chi_0 = \beta = T^{-1}$ is the static susceptibility of the paramagnetic ONC system, and $D_0(k) = \Omega^{-1}(k)$ is the magnetoacoustic Green's function at zero frequency. Inserting (9) into (11), we obtain

$$T_m = \frac{n [g_1 + g_2 b / \omega_s(0)]^2 k_0}{\Omega(k_0)}, \quad (12)$$

where $\Omega(k)/k$ is a minimum at k_0 , and k_0 determines the period of the modulated structure for $T < T_m$. The frequency of the magnetoacoustic mode in the collective excitation spectrum of the system vanishes at T_m ; it is given by the equation¹⁹

$$\omega - \Omega(\mathbf{q}) - n \Lambda^2(\mathbf{q}) \Pi(\omega) = 0, \quad (13)$$

where $\Pi(\omega)$ is the analytic continuation

$$\Pi(\omega_m) = \frac{1}{\beta} \sum_{\alpha=1}^3 \int_0^\beta \exp(i\omega_m \tau) \langle \hat{X}^{\alpha,\alpha+1}(\tau) \hat{X}^{\alpha+1,\alpha}(0) \rangle d\tau,$$

$$\omega_m = 2\pi m T, \quad m = 0, \pm 1, \pm 2, \dots$$

Expressing the Hubbard operators in terms of Fermi operators and making use of projection operators,²⁰ one can show that

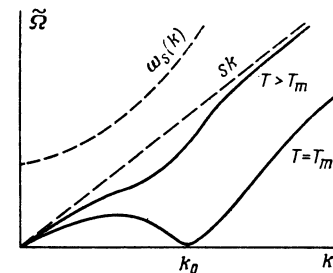


FIG. 5. Temperature behavior of the collective mode, $\tilde{\Omega}(k)$ (solid traces); of the spin-wave branch, $\omega_s(k)$; and of the acoustic branch, sk . Here T_m is the temperature at which the transition occurs to the modulated state with wave vector k_0 .

$$\Pi(\omega) = \beta\Delta/(\omega - \Delta), \quad (14)$$

where Δ is the size of the tunnel splitting of the ONC energy level. Substituting (14) in (13), we get the following formula for the frequency of the soft mode in the collective excitation spectrum for a system of oxygen-nickel complexes coupled by magnetoelastic interactions:

$$\bar{\omega} = \frac{\Omega(\mathbf{k}) + \Delta}{2} - \left[\left(\frac{\Omega(\mathbf{k}) - \Delta}{2} \right)^2 + \beta n \Delta \Lambda^2(\mathbf{k}) \right]^{1/2}. \quad (15)$$

The temperature dependence of the soft mode is shown in Fig. 5. Using the smallness of the magnetoelastic constant b , we can rewrite (12) as

$$T_m \approx \frac{n[g_1 + g_2 b/\omega_s(0)]^2}{2\omega_0} \quad (16)$$

$$\left[\frac{1}{2} + \frac{b^2}{k_0 \omega_0 (\alpha^{-1} - \alpha)} \right], \quad (17)$$

$$k_0 = k_d \alpha / (1 - \alpha^2)^{1/2}, \quad \alpha = \hbar s / a \omega_0.$$

It is plain from (17) that k_0^2 increases linearly with the magnetic field, as found experimentally. Substituting the numerical values $H_E \approx 3 \cdot 10^6$ Oe, $a = 5 \cdot 10^{-8}$ cm, $H_a \approx 0.5$ Oe, $\omega_0 \approx 1.1 \cdot 10^{-13}$ erg, $s = 4.3 \cdot 10^5$ cm/s, and $H_D \approx 10^5$ Oe (Ref. 21), we obtain $\alpha = 0.08$ for $H_0 = 0$ and $k_d = 2.8 \cdot 10^3$ cm $^{-1}$. This gives $\lambda = 2\pi k_0^{-1} = 2.7 \cdot 10^{-2}$ for the period of the superstructure, in very close agreement with the experimental value (Fig. 3). We note that due to the smallness of the coefficient α , the period of the superstructure in the ONC system is an order of magnitude larger than the characteristic magnetic scale, equal to the width of a domain wall. The proportionality factor η in the formula $k_0^2 = \eta H_0$ can also be evaluated and compared with the experimental value. We find

$$\eta_{th} = \alpha^2 \mu_B^2 H_D / (\omega_0 a)^2 = 2.6 \cdot 10^3, \quad \eta_{exp} = 16.4 \cdot 10^3,$$

which have the same order of magnitude, again demonstrating the agreement between theory and experiment.

Formula (16) predicts that the phase transition temperature T_m depends on the gap $\omega_s(0)$ in the spin-wave spectrum, i.e., on the frequency of the homogeneous antiferromagnetic resonance. The size of the gap can be varied by changing the magnitude or the direction of the external magnetic field \mathbf{H}_0 . In the first case, we see from (16) that $\Delta T_m \sim -H_0$ holds to first order in H_0 , i.e., T_m should decrease linearly with increasing magnetic field (applied in the basal plane along the axis of crystallographic anisotropy), as was observed experimentally (Fig. 3). Regarding the second situation, one must bear in mind that the hexagonal crystallographic anisotropy is not the only factor contributing to the magnetic anisotropy in the easy plane in FeBO $_3$:Ni—the light-induced uniaxial magnetic anisotropy also contributes, and the axis of the latter makes an angle with the C_2 symmetry axis of the crystal. The competition between the Zeeman and the magnetically anisotropic interactions will determine the equilibrium direction of the weak ferromagnetic moment, which turns out to be aligned at an angle $\approx 10^\circ$ relative to the direction of the magnetic field when $H_0 \perp C_2$. The gap in the spin-wave spectrum will obviously decrease when the magnetic field rotates toward the equilibrium position \mathbf{m}

(i.e., toward the easy magnetization axis for the induced anisotropy), while it will increase when the rotation is in the opposite direction. Similarly, changes in the direction of the magnetic field should also affect the period of the superstructure, and a dependence of T_m and k_0 on the magnetic field was in fact observed experimentally (Fig. 4).

5. CONCLUSIONS

In summary, the phase transition mechanism proposed above yields results in satisfactory agreement with experiment, which suggests that the light-induced transition to the modulated phase in FeBO $_3$:Ni is thermodynamic in nature. The spatial period depends on the magnetic and elastic properties of the FeBO $_3$ matrix, while the transition temperature depends on the coupling strengths characterizing the photoinduced interaction among the impurity complexes. All the features typical of phase transition are therefore present in the ONC system. The magnetic system, in which a modulated structure with a large spatial period forms at temperatures below T_m , is a sensitive indicator of the presence of inhomogeneous ordering of the lattice impurities, and the changes in magnetic structure are accompanied by changes in crystal symmetry. On the other hand, because the ONC's are coupled by the magnetoacoustic modes, the phase transition can be regulated by means of an external magnetic field.

When $T < T_m$, the dynamic properties of the FeBO $_3$:Ni system give rise to a movement (slipping) of the modulated structure due to the excitation of a light-induced phason mode; this mode results from a spontaneous breaking of translational invariance which is similar to that found in the dynamics of charge density waves in quasi-one-dimensional metals. This is observed experimentally⁵ as a moving grid of stripes which have contrasting magneto-optical properties and correspond to oscillations of the ferromagnetic vector. A more detailed description of the dynamic behavior will be given elsewhere.

In closing, we note the specific requirements for a light-induced superstructure to form in magnetic materials: the spin-wave spectrum must lie above the acoustic branch; the ground state of the impurity must be degenerate (or nearly so); and finally, the coupling between the impurity and the magnetic matrix must be weak in the absence of illumination.

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