Scenarios for the ordering and structure of self-organized two-dimensional domain blocks in thin magnetic films

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We present new data on the dynamic self-organization of the magnetic moment distribution in thin films induced by a pulsed magnetic field. We discuss possible scenarios for the transition from chaos to self-ordering: the excitation of a nonlinear pulsed mode of domain oscillations, dynamic clustering, and the drift of two-dimensional domain blocks. We study theoretically and experimentally the phenomenon of magneto-optic light diffraction by the regular domain structures arising during the self-organization with various motif-forming elements of the Bravais cells and symmetries. © 1995 American Institute of Physics.

1. INTRODUCTION

References 1 to 4 discussed the observation and study of new kinds of magnetic moment self-organization in films of ferrite-garnets with a large constant uniaxial anisotropy $\beta_{\mu} \gg 4\pi$ under the action of unipolar pulses of a magnetizing field, with the formation of lattices of dumbbell-shaped (or dumbbell-shaped and circular) cylindrical magnetic domains (CMD) with a symmetry which under static conditions corresponds to the two-dimensional space groups Pab2 and P6 and in the dynamic regime to the groups Cmm2 and P6mm. A study of the change in shape of the domains during selforganization via high-speed photography has shown $^{2-4}$ that in this case large-amplitude pulsating domain oscillations are excited, with compression and extension in two mutually perpendicular directions. It was suggested³ that a change in the parameters of the external action as well as the use of films with non-uniaxial anisotropy, might lead to the formation of variously shaped two-dimensional domain blocks with any symmetry allowed by the theory of twodimensional space groups. This hypothesis was confirmed in more recent experiments in which it was possible to observe self-ordered domain structures with symmetries which in the static regime are described by the space groups P2, Pmm2, P6, or Cmm2, and with elements of blocks of, respectively, L- or S-shaped, dumbbell, ring, and twin-ring forms.^{5,6}

The variety of shapes arising in the self-organization of domain structures generates an extensive set of optical patterns observed in the diffraction of light by films with regular inhomogeneous magnetic moment distributions. Since the directions of the diffraction maxima are uniquely determined by the reciprocal lattice vectors, that is, by the symmetry of the domain structures, and the intensity distribution of the diffracted light in magneto-uniaxial films in Faraday effect observations is the spectral density of the correlation function of the magnetization vector \mathbf{M} (more precisely, the component of \mathbf{M} along the normal \mathbf{n} to the film surface), an analysis of the results of experiments on the observation of magneto-optic Fraunhofer diffraction by inhomogeneous magnetic moment distributions arising in the selforganization yields information both on the symmetry of a given two-dimensional block and the degree of ordering of the domain arrangement in it. In other words, light diffraction plays the same role for an analysis of domain structures as X-ray diffraction for the determination of the structure of crystals and other condensed media with long-range and short-range order.⁷ Thus far, experiments and theoretical calculations of the magneto-optic diffraction of light by regular domain blocks apply only to the simplest situations: a strip domain structure, or hexagonal and rectangular arrays of CMD (see Refs. 8 and 9 and the bibliography given there).

In the present paper, we provide new data on dynamic self-organization of the magnetic moment distribution in thin quasiuniaxial ferrite-garnet films subject to a harmonic or pulsed unipolar magnetic field. We discuss possible scenarios for self-ordering: excitation of a nonlinear pulsation mode of dumbbell-shaped (quasielliptical) domains, dynamic clustering of heteropolar hexagonal lattices of circular CMD, and drift of two-dimensional domain blocks.¹⁾

We show that for self-organization with participation of a nonlinear pulsating oscillation mode, there exists a welldefined dependence of the angle to which the axis of the dumbbell-shaped domains is reoriented by each pulse of the magnetizing field on the domain density in the block, that is, on the average magnetization in the cell. If the domain density exceeds some critical value, no reorientation of the axis is observed, in general, and during self-ordering, alternating series of short and long dumbbell-shaped domains are formed, with each pulse of the magnetizing field changing the long domains into short ones and the short domains into long ones.

We observed that for nonlinear oscillations of a onedimensionally periodic system of strip domains, the symmetry of which is described by the so-called border group G_1^3 (Ref. 11), two-dimensionally periodic ordered configurations are formed that are similar to the bimodal structures in liquids in Rayleigh–Bénard convection with a large Prandtl number.¹² It was established that under certain conditions, a strong sinusoidal magnetic field generates a peculiar domain structure in uniaxial films, consisting of blocks (clusters) of heteropolar CMD lattices that can be transformed into ordered two-dimensional blocks of ring domains or twin-ring (figure-eight) domains. Drift (translational motion) of domain structures observed in some films under the action of a pulsed magnetic field is accompanied by the emergence of additional gyroscopic forces, which leads to the selforganization of these structures into blocks of chiral S-shaped elements or (L-shaped) elements shaped like a boomerang.

The conditions for performing the experiments described in what follows were similar to those used in Refs. 1 to 4. The objects of our study were epitaxial films of ferritegarnets of differing composition, possessing a strong induced uniaxial anisotropy with easy magnetization axis (EMA) along the normal **n** to the surface, which were subjected to the action of a (unipolar pulsed or sinusoidal) magnetic field **H** n created by a plane 10-loop coil with internal diameter $\simeq 1$ mm. As necessary, we also used a constant magnetization field $H_0||n$. The domain structures which then appeared can be visually observed and photographed by means of a polarization microscope. The polygraphic processing of the photographs of the domain structures was carried out on a PC using standard photoretouching programs; the same procedure was applied also for the photographs of the Fraunhofer diffraction patterns arising when a laser beam of wavelength $\lambda = 0.6328 \ \mu m$ passed through the film. Photographs were also obtained both for "frozen" (static) domain structures observed after the pulsed magnetic field was turned off, and for the dynamic domain structures existing when there was no \hat{H} field. In the latter case the exposure time was significantly longer than the repetition period of the pulsed magnetic field so that we recorded a picture averaged over many pulses.

Some of the results reported here were published before in Refs. 5 and 6.

2. SELF-ORGANIZATION OF NONLINEAR PULSATING OSCILLATIONS OF DUMBBELL-SHAPED (ELLIPTICAL) DOMAINS

The present section examines domain structure selforganization under the action of a unipolar pulsed field such that in the ordering process, regular blocks of elongated (dumbbell-shaped or elliptical) domains or blocks of elongated and circular domains are formed. The latter are characterized by a change in the position of the major axis of the elongated domains by a well defined angle, or a change in the ellipticity (the ratio of the major and minor symmetry axes) of such domains under the action of each magnetic field pulse. These changes are cyclic in nature; in particular, the structure is exactly repeated after two consecutive pulses. Ordered blocks are formed from the usual labyrinth of domain structures in two stages. Initially a set of magnetic dislocations (i.e., discontinuities of strip domains) is generated, and a simple hexagonal lattice containing only circular CMD is formed; afterwards there is a transformation to more complex lattices, fully or partly consisting of elongated domains.²⁾ The regular domain structures formed in the selforganization process are statically stable, i.e., they are not destroyed after the pulsed magnetic field is switched off. Optimal for the creation of structures of the kind considered is the use of periodic sequences of roughly triangular pulses $(\tau_r \approx \tau_f \approx 1/2 \tau_p$ where τ_r and τ_f are, respectively, the length of the rising and falling edge of a pulse of duration τ_p) with a repetition period T_p which is much longer than the length of the pulse. The strength of the pulsed magnetic field $\tilde{\mathbf{H}}$ was considerably (several times) larger than the strength of the static collapse field H_c of the CMD, and the direction of $\tilde{\mathbf{H}}$ was chosen antiparallel to the direction of the magnetization \mathbf{M} inside the domains, so that the latter were compressed during the pulse.

By slightly varying the shape of the pulse and using a constant magnetization field H_0 which was weak (compared to the collapse field), one can realize structures belonging to two different crystal systems, hexagonal and rectangular. For a static regime ("frozen" configurations), the observation of the symmetry of the domain structures is in the first case (Fig. 1a) described by the two-dimensional space group P6 and in the second case either by the Pab2 (Figs. 2a, b) or the *Pmm2* group (Fig. 2c). In the dynamic regime (photographed when there is no pulsed field with an exposure considerably longer than the repetition period of the pulses), the symmetry of the domain structures is increased for the hexagonal system to P6mm (Fig. 1c), and for the rectangular system to Cmm2 (Figs. 2e, f). The photographs of Fig. 1 and Figs. 2a, d, g were obtained for film No. 1, with composition $(YBi)_3(FeGa)_5O_{12}$, thickness $t=20 \ \mu m$, a substrate of $Gd_3Ga_5O_{12}$ with (111) orientation, and period $d=60 \ \mu m$ of the domain structure labyrinth, uniaxial anisotropy constant $\beta_{\mu} \simeq 60$, magnetization $M \simeq 6.7$ G, and collapse field of the CMD $H_c \approx 14$ Oe. We used sequences of triangular field pulses $(\tau_r \simeq \tau_f \simeq 1/2 \tau_p \simeq 2\mu s)$ with repetition frequency $f_p = T_p^{-1} \simeq 1$ kHz and amplitude $\tilde{H} = 77$ and 53 Oe, respectively. The structures shown in Figs. 2b, c, e, f were realized in film No. 2, with composition (YGdYbBi)₃(FeAl)₅O₁₂ (thickness $t = 20 \ \mu m$, substrate Gd₃Ga₅O₁₂, (111) orientation, period of the domain structure labyrinth $d=58 \ \mu m$, anisotropy constant $\beta_{\mu} \approx 20$, magnetization $M \approx 7.7$ G, collapse field $H_c \approx 39$ Oe) under the action of a periodic sequence of triangular pulses with amplitude $\tilde{H} \simeq 75$ Oe, width $\tau_p \simeq 2 \ \mu s$, and repetition frequency $f_p \approx 1$ kHz. The transition from a regime of oscillations with a reorientation of the major axis of the dumbbell-shaped domains over a nonvanishing angle ψ (Figs. 2a, b, d, e) to a regime without reorientation but with a periodic change in the length of the major axis of the domains in the series (Figs. 2c, f) is realized through a change in the domain density in the initial hexagonal lattice of circular CMD (see below).

Direct observations of domain shape in structures like those in Fig. 1a and Fig. 2d by the high-velocity photography method showed^{2,3} that a change in the position of their axes occurs not through rotation, but due to shock excitation (by each magnetic field pulse) of a single half-period of the pulsed oscillations of the domains with a large amplitude, and with compression and extension in two mutually perpendicular directions. The evolution of the shape of the domains (e.g., in a structure of the type shown in Fig. 1c) for such oscillations is illustrated by the series of photographs in Fig. 3 referring to different times τ after the start of the pulse (τ =0). For the sake of clarity, the centers of the domains were shifted relative to one another. The absence of a simi-

b

а



FIG. 1. Photographs of domain structures of the hexagonal system in film 1 and the corresponding diffraction patterns in static (a,b)and dynamic (c,d) regimes.

larity of the shape of the domains in different stages of the motion indicates the impossibility of describing the observed effects in a model with a "geometric" domain wall with a constant surface-energy density ($\sigma_w = \text{const}$). The oscillations are notably mass-dominated: the domains reach a new equilibrium position after a time three times longer than the magnetic field pulse width.

Although we did not succeed in realizing all types of domain structures shown in Figs. 1 and 2 in a single film, and self-ordered domain blocks with differing symmetry coexisted rather often (see, e.g., Fig. 10 in Ref. 3), one can, nonetheless, state that the critical parameter determining the choice of one of the possible variants is the average value of the z component of the magnetization of the film in the absence of a magnetic field, i.e., the remanent magnetization M_r . Since all films used in the experiments are quasiuniaxial and possess a large uniaxial anisotropy constant ($\beta_u \gg 4\pi$), the magnetization vector is collinear with the normal to the surface, and has two possible antiparallel orientations, so that

$$\langle M_z \rangle = M_r = M_0 (1 - 2p_f), \qquad (1)$$

where $p_f = S_d/S_c$ is the filling factor of the two-dimensional domain block, which is just the ratio of the sum S_d of the areas of the cylindrical domains (or their parts) inside the elementary cell to the area S_c of the latter. The value of p_f

can for various domain lattices be determined by computer methods from the distribution of the optical density in the photographs of Fig. 1a and Fig. 2c using standard image processing software. A computer analysis shows that the values of the filling factor for the structures shown in Fig. 1a and Figs. 2a to c are, respectively, 0.33, 0.4, 0.56, and 0.585.³⁾ Hence it follows that for $p_f < 0.5$, i.e., in sufficiently rarefied two-dimensional CMD blocks, where the effect of neighbors on the behavior of each domain is small, the orientation of the major axis of elliptical (dumbbell-shaped) domains changes under the action of a magnetization field pulse by $\pi/2$. It was noted in Ref. 3, incidentally, that for parameters of the pulsed magnetic field close to those necessary for the formation of domain structures like those in Figs. 1a, c and Figs. 2a, d, the ends of the isolated strip domains also change orientation periodically by $\pi/2$ under the action of each pulse of the magnetization field. Hence, this tendency is not a collective, but an individual effect. In more densely packed domain lattices ($p_f > 0.5$), the effect of the surroundings is no longer negligible and the "rotation" angle of the major axes of the elliptical domains does not reach $\pi/2$; it tends to zero for $p_f \ge 0.58$. We note that in the latter case (ψ =0), there is a change in the symmetry of the twodimensional block from Pab2 to Pmm2.



FIG. 2. Photographs of domain structures of the rectangular system in films No. 1 (a,d) and No. 2 (b,c,e,f) in static (a to c) and dynamic (d to f) regimes, and the corresponding diffraction patterns (g to i). The reorientation angle ψ of the major axis of the elliptical (dumbbell-shaped) domains is $\pi/2(a,d)$; $\simeq \pi/6(b,e)$; and 0(c,f). The diffraction patterns for the corresponding static and dynamic structures have the same form.

Self-organization in the case of nonlinear domain oscillations under the action of a sequence of unipolar magnetization field pulses can proceed not just through a stage of formation of a hexagonal lattice of circular CMD. For instance, if in film No. 1 one creates a disordered labyrinth of domain structures and subjects it to a sequence of pulses with an amplitude and (or) width insufficient for the formation of a lattice of circular CMD, for some critical value of the amplitude $\tilde{H} \simeq H_c$ one observes the appearance of blocks inside each of which there exists a regular one-dimensionally periodic system of strip domains (the symmetry of which is described by the so-called border group $G_1^3[11]$), but the orientation of the domain boundaries (DB) changes randomly from block to block. For a smooth increase in the amplitude of the pulses, nonlinear domain boundary oscillations are excited with an amplitude comparable to the period of the initial strip domain structures, which are "frozen" when the pulsed magnetic field is turned off (Fig. 4a; in the dynamic regime, one observes the configuration shown in Fig. 4b). As the pulse width increases, the domains (at the maximum deviation from their average position) acquire a meandering shape, and we see structures like those in Figs. 4c, d. The resulting two-dimensional ordered configurations bear a remarkable external resemblance to the bimodal structures in liquids for Rayleigh-Bénard convection with a large Prandtl number (cf. Fig. 5 and Fig. 6 in Ref. 12).

3. SELF-ORGANIZATION IN THE CASE OF DYNAMICAL CLUSTERING OF HETEROPOLAR HEXAGONAL LATTICES OF CIRCULAR CMD

It is well known that in magneto-optic films with strong "perpendicular" anisotropy ($\beta_u \ge 4\pi$), an initial labyrinth of domain structures can be transformed into a hexagonal lattice of circular CMD not only by means of a unipolar pulsed magnetization field, but also by some harmonic action.¹³ In the latter case, however, the direction of the magnetization vector inside the CMD (parallel or antiparallel to the normal to one of the film surfaces) is not uniquely determined by the external influence, the steady component of which is zero, so that the emergence of a monopolar CMD lattice suggests spontaneous symmetry breaking.⁴

We have studied the influence of the amplitude of a har-



FIG. 3. Change in the shape of a dumbbell-shaped domain in the structure of Fig. 1c, engendered by a single magnetic field pulse. The delay τ between the start of the pulse and the time of exposure is (from left to right) 0, 2.8, 3.2, 5.2, 5.5, 7.2, 8.8, and 12.0 μ s.





b

С



FIG. 4. Self-organization in the case of nonlinear oscillations of strip domains in film No. 1 induced by unipolar rectangular magnetization field pulses with an amplitude of \approx 15 Oe and width 3 μ s (a,b) and 10 μ s (c,d). Shown are photographs of static (a,c) and dynamic (b,d) configurations.

monic magnetization field \tilde{H} on the formation of CMD lattices in ferrite-gamet film No. 3, with the composition (YBi)₃(FeGa)₅O₁₂ ((111) orientation, $t=7 \mu m$, $d=60 \mu m$, $\beta_{\mu} \approx 60$, $M \approx 6.7$ G, $H_c \approx 14$ Oe). We used a harmonic magnetic field with amplitude $\tilde{H} > H_c$ and frequency $f \sim 10$ Hz. Within a range of magnetic field amplitude, a monopolar hexagonal lattice of circular CMD with the same direction of the vector **M** inside all the CMD formed in the film, starting



b

FIG. 5. Photographs of domain structures in film No. 3, self-organizing due to dynamic clustering of heteropolar CMD lattices.



FIG. 6. Self-organization of domain structures in film No. 4 in the case of drift of two-dimensional blocks as a whole.

out with the initial labyrinth domain structure, i.e., spontaneous symmetry breaking occurred. However, with increasing amplitude H, we observed a restoration of the broken symmetry element through the random generation and growth of clusters that are part of a hexagonal CMD lattice with an antiparallel direction (with respect to the original lattice) of the vector **M** within the CMD.⁵⁾ The number n_d of CMD in such clusters when these are formed can be rather large. If in such a situation one applies a weak constant magnetic field $H_0 \ll H_c$ to the film and gradually increases it, rapid dissociation of the clusters starts (the symmetry is again broken in the system), and n_d decreases monotonically. Simultaneously, the fragments of the initial CMD lattice get displaced. A tendency to an ordering of the clusters starts to manifest itself for $n_d \leq 6$, but a complete self-organization of the cluster lattice expressing itself in the formation of an ordered block of twin-ring domains is observed for $n_d=2$ (see Fig. 5a). The symmetry of such a domain structure is described by the two-dimensional space group Cmm2. For a slightly stronger magnetization field H_0 , the clusters with $n_d=2$ dissociate and form a hexagonal CMD lattice with symmetry P6mm (Fig. 5b).

We note that the self-organization of domain structures in the series of experiments described here was observed only when a small gap existed between the film and the coil producing the alternating magnetic field, i.e., the alternating field possessed a spatially inhomogeneous radial component. The magnitude of the gap was a rather critical parameter: when the gap was too narrow, the formation of clusters of heteropolar CMD lattices was cut off, and when the gap was large, cluster ordering was hindered by the strong inhomogeneity of the alternating magnetic field.

4. SELF-ORGANIZATION DURING DRIFT OF TWO-DIMENSIONAL DOMAIN BLOCKS AS A WHOLE

One quite often observes during self-organization the socalled self-organizing processes which are characterized by the appearance, in a medium which is in a nonequilibrium state, of traveling or standing space-time inhomogeneities which have a well defined lifetime, symmetry, and some degree of order. In magnetics there also exist various self-

organizing effects (spiral waves, "guiding center" oscillations, periodic modulation of the phase front, and so on^{15-19}). From the multiplicity of self-organizing effects which up to the present have been observed in magnetics, only one is of interest for what follows, namely, the drift (translational motion) of two-dimensional domain lattices (circular CMD or strip domains) induced by a uniform external field-a pulsating magnetic field, light, and so on.²⁰⁻²² The reasons for drift have yet to be fully clarified; the existing hypotheses²³⁻²⁵ are not able fully to describe all features of the observed effects. In magneto-optical ferrite-garnet films, the translation of domain structures engendered by a pulsating magnetization field is probably related to a large group of effects called CMD self-motion when there is no magnetization field gradient (see Malozemoff and Slonczewski's monograph¹⁹ and the references in it).

In our experiments, magnetic film No. 4 with composition (LuBi)₃(FeGa)₅O₁₂ was used; it was grown on a substrate of Gd₃Ga₅O₁₂ with (210) orientation, which ensured the presence of a strong rhombic component of induced anisotropy $(\beta_p \sim \beta_u)$, where β_p is the rhombic anisotropy constant). The film had the following parameters: $t=14 \mu m$, $d=14\mu$ m, $\beta_{\mu}\simeq 3.3$, $M\simeq 7.3$ G, $H_c\simeq 66$ Oe. When one acts on the film with unipolar rectangular magnetic field pulses $\tau_p \sim 10 \mu s$ wide, with repetition period $T_r = 10$ ms and amplitude H>90 Oe, the initial strip domain structure broke down, forming a lattice of elliptical domains, and the block drifted as a whole. In the course of this drift, self-organization occurred with the formation of ordered lattices of S- or L-shaped domains (Figs. 6a and 6b) with symmetry described by the group P2. The conditions for the formation of the structures shown in Figs. 6a, b are essentially identical; quite often they simply coexist.

The appearance of chirality ("twisting" of the domain ends) in the elements of the block of Fig. 6a can be explained as follows. It is well known^{19,26–28} that a sequence of unipolar magnetization field pulses will make isolated dumbbellshaped domains start to rotate in a direction determined by the polarity of the pulses. This is related to the fact that in the case of compression (expansion) of a domain with a velocity **v**, distributed gyrotropic forces with a density¹⁹

$$\mathbf{f_g} = \frac{M \sin \vartheta}{\gamma} \left[(\nabla \vartheta \nabla \varphi) \mathbf{v} \right], \tag{2}$$

act on its ends, where γ is the gyromagnetic ratio, $\vartheta = \arccos(M_z/M)$, and $\varphi = \arctan(M_y/M_x)$. Since the vectors v at opposite ends of the domain are antiparallel, it follows from Eq. (2) that a pair of forces appears with a moment which tends to turn the domain. The resulting rotation angle in each pulse of the field must, however, theoretically be equal to zero, since the rotation of the domain in one direction in the compression stage (during the pulse) is cancelled by the opposite rotation in the expansion stage after the pulse ends. Nonetheless, in the experiments, the resulting rotation angle in each pulse can be several tens of degrees, which in the opinion of the authors of Refs. 19 and 26–28 may be due to the coercive force.

In our experiments, however, a unidirectional drift⁶⁾ is superimposed on the domain oscillation process. This leads to a breaking of the collinearity of the velocity vectors of each individual section of the domain boundary of an element of the block in the compression and expansion stages. This can produce a nonvanishing resultant rotation angle of the domain, even without invoking any mechanism involving the coercive force. Moreover, in the lattice, the rotation of each domain as a whole is hindered by the existence of neighbors, so that the effect of the gyrotropic forces reduces to a small twisting of the ends of the domains, i.e., to the emergence of chirality in the elements of the domain block (see Fig. 6a). When the polarity of the pulsed magnetic field changes, so does the direction of the twist. We note that the existence of drift does not always lead to chirality in blockforming elements (see Fig. 6b).

5. DIFFRACTION OF LIGHT BY ORDERED DOMAIN STRUCTURES FORMED IN THE CASE OF SELF-ORGANIZATION

In the present section, we give the results of a theoretical and experimental study of the diffraction of light by selforganized two-dimensional domain blocks with various symmetries and various motif-forming elements of the Bravais cell. We mentioned already in the introduction that experiments carried out so far and calculations of the magnetooptic diffraction of light by domain structures are related only to the simplest situations: a strip domain structure, hexagonal and rectangular CMD lattices.

In the optical images produced by the diffraction of light in films with regular inhomogeneous magnetic moment distribution, the location of the diffraction maxima is uniquely determined by the reciprocal lattice vectors, i.e., by the symmetries of the domain structures, and the distribution of the intensity of the diffracted light is the spectral density of the correlation function of magnetization vector. An analysis of experimental observations of magneto-optical Fraunhofer diffraction by inhomogeneous magnetic moment distributions that result from self-organization therefore yields information not only about the symmetry of a given twodimensional block, but also about the degree of order of the domains within it. Part of the information is then unfortunately lost, for the following reasons. Firstly, only point symmetry groups are determined in diffraction experiments. For two-dimensional magnetic moment distributions, the number of space symmetry groups is 17, and the number of point groups is 10. The relationship between them is given by (see, e.g., Ref. 29):

$$P1 \rightarrow 1; P2 \rightarrow 2; \{Pm, Pa, Cm\} \rightarrow m;$$

$$\{Pmm2, Pam2, Pab2, Cmm2\} \rightarrow mm2;$$

$$P4 \rightarrow 4; \{P4mm, P4am\} \rightarrow 4mm; P3 \rightarrow 3;$$

$$\{P31m, P3m1\} \rightarrow 3m; P6 \rightarrow 6; P6mm \rightarrow 6mm.$$

Secondly, possible types of optical diffraction patterns⁷⁾ in the case of normal incidence of light on sufficiently thin films can only belong to the six two-dimensional point groups which contain an inversion center as symmetry element (the latter is equivalent in the two-dimensional case to a second-order rotation axis). A one-to-one correspondence between the point symmetry group and the diffraction pattern occurs only for the groups 4 and 4mm; scattering by objects characterized by the point groups 1 and 2 is described by the same point group 2, for groups 3 and 6 by group 6, for groups m and mm^2 by group mm^2 , and for groups 3m and 6mm by group 6mm. There is a fairly complete analogy between the use of light diffraction for the analysis of the magnetization distribution in domain structures and the application of x-ray diffraction to determine the structure of crystals and other condensed media with long- and shortrange order.

The experimental observations of light diffraction by ordered domain structures were made with a helium-neon laser operating at $\lambda = 0.6328 \ \mu m$. The diameter of the laser beam incident on the film surface along the normal **n** to the surface was ≈ 1 mm (the beam was necessarily additionally spatially filtered), and the divergence of the beam was at most 10 mrad. The diffraction patterns could be observed visually, photographed, and recorded on the hard disk of a PC using a videocamera-videoadapter system to input the images. The computer "photographs" of the diffraction patterns for some of the observed ordered static and dynamic domain structures, obtained when the transmission planes of the optical analyzer and polarizer were orthogonal, are given in Figs. 1b, d and Figs. 2g-2i. The symmetry of the first of the listed diffraction patterns is described by the point group 6, the second by the group 6mm, and the remaining ones by the point group mm2. For computer processing ("photoretouching") the characteristic Hurter-Drifflield curve (the dependence of the optical density on the exposure on a semilogarithmic scale) was chosen in the form of a step function with a threshold value determined by the minimum level of clearly observed diffraction maxima. All maxima then corresponded to the minimum level of black on a negative (or the maximum level of white on a positive) while the area of the diffraction spots was roughly proportional to their intensity.

A theoretical calculation of the intensity and location of diffraction maxima in the Fraunhofer approximation was carried out using well known methods (Refs. 33 and 34; see also Refs. 8 and 9). In the case of scattering of light incident on the film along the z axis ($\mathbf{e}_z || \mathbf{n}$), maxima will be observed in the directions

$$\mathbf{k}_{(p,q)} = \mathbf{k} + p \mathbf{b}_1 + q \mathbf{b}_2, \tag{3}$$

where **k** is the wave vector of the incident light, \mathbf{b}_1 and \mathbf{b}_2 are the basis vectors of the reciprocal lattice, p and q are integers, and the components E_j (j=1,2) of the diffracted radiation are given by

$$E_{j}(x_{d}, y_{d}) = \frac{\exp(ikz_{0})}{2\pi i} \int_{S} \int E_{j}(x, y)$$
$$\times \exp\left[-\frac{ik}{z_{0}}(xx_{d}+yy_{d})\right] dxdy, \qquad (4)$$

where (x,y) and (x_d,y_d) are systems of coordinates (with collinear unit vectors) in the plane of the object and the plane of diffraction, respectively; z_0 is the distance from the object to the diffraction plane, S is the illuminated area of the object (aperture), and $E_j(x,y)$ is the aperture function, which determines the distribution of the field components of the incident radiation immediately after passing through the film. Introducing the notation $\xi = kx_d/z_0$ and $\eta = ky_d/z_0$ and omitting unimportant factors and indices, we can write Eq. (4) in the form

$$E(\xi,\eta) = \iint_{S} E(x,y) \exp[-i(\xi x + \eta y)] dx dy, \qquad (5)$$

from which it follows that the field components of the diffracted radiation are proportional to the Fourier transform of the aperture function.^{32,33}

We noted in Sec. 3 that in magneto-optic films with $\beta_{\mu} \gg 4\pi$, the magnetization vector in the uniformly magnetized state must for H=0 be collinear with the normal to the surface. In films with domain structures this is, strictly speaking, not the case, since the distribution of the vector M is nonuniform throughout. However, if the conditions for carrying out the experiments are chosen such that the film is far from second order phase transition points or lines, the period of the domain structures is much larger than the width of the domain boundaries, inside which the vector M is not collinear with the normal n. One can then assume that for normal incidence, $\mathbf{k} \uparrow \uparrow \mathbf{M}$ or $\mathbf{k} \uparrow \downarrow \mathbf{M}$ inside the meduim in different domains. In such a geometry, the Faraday effect shows up. In the absence of circular magnetic dichroism, this simply leads to rotation of the polarization plane of linearly polarized light by an angle $\pm \Phi_F = \vartheta_F d$, where ϑ_F is the specific Faraday rotation. The domain structure in the present case is purely a phase diffraction lattice⁸⁾ and the distribution of the angle of rotation of the plane of polarization of the light after passing through the film is identical with the distribution of the zcomponent of the M vector. This remains true when the direction of the vector M in the domains differs from the normal by an angle which is not too close to $\pi/2$; the effect of the second-order magneto-optic constants on the diagonal components of the dielectric tensor is then negligible and the characteristic waves remain circularly polarized.³⁴

If diffraction is produced by an ordered two-dimensional lattice with basis vectors $\mathbf{a}_1 = a_{1x}\mathbf{e}_x + a_{1y}\mathbf{e}_y$ and $\mathbf{a}_2 = a_{2x}\mathbf{e}_x + a_{2y}\mathbf{e}_y$ containing $P \times Q$ elementary cells, we can write Eq. (5) in the form^{32,33}

$$E(\xi,\eta) = L(\xi,\eta)S(\xi,\eta), \tag{6}$$

where $L(\xi, \eta)$ is a lattice function defined by the equation

$$L(\xi,\eta) = \sum_{p=0}^{P-1} \exp[-ip(\xi a_{1x} + \eta a_{1y})] \\ \times \sum_{q=0}^{Q-1} \exp[-iq(\xi a_{2x} + \eta a_{2y})],$$
(7)

while $S(\xi, \eta)$ is the structure factor equal to

$$S(\xi,\eta) = A_c \iint_{S_c} \exp[-i(\xi x + \eta y)] dx dy$$
$$+ (A_d - A_c) \iint_{S_d} \exp[-i(\xi x + \eta y)] dx dy.$$
(8)

Here S_c is the area of an elementary cell, S_d is the sum of the areas of CMD (or their parts) inside an elementary cell, and A_c and A_d are the values of the aperture function for regions outside and inside the CMD, respectively. If without restricting generality we assume that the vector **E** of the incident radiation is polarized along the x axis, i.e., $\mathbf{E} = E_0 \mathbf{e}_x$, and that the angles of rotation of the polarization plane due to the Faraday effect outside the CMD and inside them are, respectively, equal to $+\Phi_F$ and $-\Phi_F$, one easily checks that $\mathbf{A}_c = TE_0(\mathbf{e}_x \cos \Phi_F + \mathbf{e}_y \sin \Phi_F)$ and $\mathbf{A}_d = TE_0(\mathbf{e}_x \cos \Phi_F - \mathbf{e}_y \sin \Phi_F)$, where T is the attenuation coefficient for optical radiation when passing through the film due to reflection from the boundaries and absorption. The expression for the modulus of the lattice function (7) can be transformed to the form

$$|L(\xi,\eta)| = \left| \frac{\sin[\frac{1}{2} P(\xi a_{1x} + \eta a_{1y})]}{\sin[\frac{1}{2} (\xi a_{1x} + \eta a_{1y})]} \right| \\ \times \left| \frac{\sin[\frac{1}{2} Q(\xi a_{2x} + \eta a_{2y})]}{\sin[\frac{1}{2} (\xi a_{2x} + \eta a_{2y})]} \right|,$$
(9)

whence it follows that the extrema of $|L(\xi, \eta)|$ exist when the conditions

$$\xi a_{1x} + \eta a_{1y} = 2p \,\pi, \tag{10}$$

$$\xi a_{2x} + \eta a_{2y} = 2q\,\pi\tag{11}$$

are satisfied. Using the well known expressions for the reciprocal lattice vectors of two-dimensional periodic structures,

$$\mathbf{b}_{1} = \frac{2\pi[\mathbf{a}_{2}\mathbf{e}_{z}]}{([\mathbf{a}_{1}\mathbf{a}_{2}]\mathbf{e}_{z})} = \frac{2\pi(a_{2y}\mathbf{e}_{x} - a_{2x}\mathbf{e}_{y})}{a_{1x}a_{2y} - a_{1y}a_{2x}},$$
(12)

$$\mathbf{b}_{2} = \frac{2\pi[\mathbf{e}_{z}\mathbf{a}_{1}]}{([\mathbf{a}_{1}\mathbf{a}_{2}]\mathbf{e}_{z})} = \frac{2\pi(-a_{1y}\mathbf{e}_{x} + a_{1x}\mathbf{e}_{y})}{a_{1x}a_{2y} - a_{1y}a_{2x}},$$
(13)



FIG. 7. Theoretical diffraction patterns for the static (a) and dynamic (b) domain structures shown, respectively, in Figs. 1a, c.

one checks easily that conditions (10) and (11) are completely equivalent to those described by Eq. (3). The centers of the diffraction spots with indices (p,q) in the (ξ,η) plane are thus positioned at the reciprocal lattice sites, i.e.,

$$\xi_{(p,q)}\mathbf{e}_x + \eta_{(p,q)}\mathbf{e}_y = p\mathbf{b}_1 + q\mathbf{b}_2.$$

For the central diffraction peak $(p=q=0, \text{ i.e.}, \xi=\eta=0)$, the structure factor is

$$\mathbf{S}(0,0) = TE_0 S_c[\mathbf{e}_x \cos \Phi_F + \mathbf{e}_y (1 - 2p_f) \sin \Phi_F], \quad (14)$$

whence it follows that the polarization of the diffracted radiation in this case is the same as the initial polarization only for a filling factor $p_f=0.5$. For the other diffraction peaks $(p \neq 0 \text{ and/or } q \neq 0)$, the first integral in Eq. (8) vanishes due to the periodicity of the boundary conditions, and the structure factor is

$$\mathbf{S}(\xi,\eta) = -2TE_0 \mathbf{e}_{\nu} K(\xi,\eta) \sin \Phi_F, \qquad (15)$$

where

$$K(\xi,\eta) = \iint_{S_d} \exp[-i(\xi x + \eta y)] dx dy.$$
(16)

The plane of polarization of the light in the additional diffraction peaks is always rotated by $\pi/2$ with respect to the original one.

It follows from (4), (6), (8), (14), and (15) that the light intensity in the various diffractions orders is

$$I(0,0) = \frac{1}{8\pi^2} T^2 E_0^2 S_d^2 [\cos^2 \Phi_F + (1-2p_f)^2 \sin^2 \Phi_F],$$
(17)

$$I(\xi,\eta) = \frac{1}{2\pi^2} T^2 E_0^2 |K(\xi,\eta)|^2 \sin^2 \Phi_F.$$
 (18)

The determination of the shape of the diffraction patterns from the observed ordered domain structures is thus reduced to calculating the filling factor p_f (i.e., the areas of the elementary cell and of the domains of various shapes) and evaluating integrals like (16) for actual domain configurations. Although this procedure was carried out for all the kinds of domain structures observed in our experiments, we restrict ourselves here to considering the cases illustrated in Fig. 1 and Fig. 2. It was shown in Ref. 4 that the shape of the boundaries of elongated domains in such structures is best described by the so-called Cassini curves (ovals),³⁵ but we approximate them with ellipses. We assume the aperture of the initial light beam to be infinite; the relative intensity of the diffraction peaks is then completely determined by the structure factor $S(\xi, \eta)$.

Let the center of an elliptical domain with semiaxes r_1 and r_2 $(r_1 > r_2)$ be situated at the point (x_e, y_e) and let its major axis be rotated relative to \mathbf{e}_x by an angle θ_e . By direct calculation,

$$K(\xi,\eta) = 2 \pi r_1 k_r \frac{J_1(r_1 \sqrt{\xi'^2 + k_r^2 \eta'^2})}{\sqrt{\xi'^2 + k_r^2 \eta'^2}} e^{-i(\xi x_e + \eta y_e)},$$
(19)

where $k_r = r_2/r_1$; $J_1(x)$ is the Bessel function, and

$$\xi' = \xi \cos \theta_e + \eta \sin \theta_e,$$

$$\eta' = -\xi \sin \theta_e + \eta \cos \theta_e.$$

For the structure shown in Fig. 1a we can, in a system of coordinates with the x axis horizontal and the y axis vertical, choose the primitive translation vectors in the form $a_1 = \{a, 0\}$ and $a_2 = \{a/2, a\sqrt{3}/2\}$, where a is the distance between the centers of the circular domains. Each elementary cell contains one circular domain of radius R with its center, for instance, at (0,0), and three elongated domains (with semi-axes $r_1 \neq R$ and $r_2 \neq R$) with centers positioned at the points $(a/2, 0), (a/4, a\sqrt{3}/4), and (3a/4, a\sqrt{3}/4), and with major axes rotated with respect to the x axis, respectively, by angles <math>-\pi/4, \pi/12$, and $5\pi/12$ (see Fig. 2 in Ref. 1 or Fig. 9 in Ref. 3). The calculation of the diffraction pattern for such a structure is carried out using the procedure described above, taking into account that in this case

$$K(\xi,\pi) = \sum_{l=1}^{4} K_{l}(\xi,\eta,r_{1}^{(l)},r_{2}^{(l)},\theta_{e}^{(l)}).$$

The results of the theoretical calculation in which the actual values of the parameters for film No. 1 were used (a=160 μ m, R=24.7 μ m, $r_1=46.6$ μ m, $k_r=0.37$) are shown in



FIG. 8. Theoretical diffraction patterns (a,b,c) for the static and dynamic domain structures shown, respectively, in Figs. 2a, d, Figs. 2b, e, and Figs. 2c, f.

in Fig. 7a. The diffraction spots are shown as black circles whose area is proportional to the intensity, while the centers are positioned at the sites of the reciprocal lattice formed by translations of the basis vectors

$$\mathbf{b}_1 = \frac{2\pi}{a\sqrt{3}} \{ \sqrt{3}; -1 \}; \quad \mathbf{b}_2 = \frac{2\pi}{a\sqrt{3}} \{ 0; 2 \}.$$

We show all diffraction peaks with intensity exceeding 1% of the intensity of the largest spot. We assumed that the transmission planes of the analyzer and polarizer were orthogonal; in that case the relative intensity of the zeroth and higher diffraction orders are independent of the angle of rotation of the plane of polarization of the light at the film.

One can obtain the dual of this domain structures, which is produced from the original one by a single magnetization field pulse (as the result of which the major axes of all elliptical domains are reoriented by $\pi/2$) via mirror reflection in a vertical or horizontal plane that passes through the center of the circular CMD perpendicular to the plane of the film. Using the same operation, one can also produce the diffraction pattern for the dual domain structure. The dynamic domain structure (Fig. 1b) is a superposition of the original and dual structures, which exist in nonoverlapping time intervals (in the breaks between neighboring magnetization field pulses), so that one can obtain the diffraction pattern for dynamic domain structures by superimposing the pattern of Fig. 7a and its mirror reflection in a vertical (or horizontal) plane passing through the center of the zeroth diffraction order (see Fig. 7b).

To facilitate an understanding of the diffraction patterns in Fig. 7, we have drawn on them a set of dashed circles with radii given by $(r/b_1)^2 = p^2 - pq + q^2$, where p and q are the indices of the corresponding reflections. A comparison of the diffraction patterns observed experimentally (Figs. 1c, d) and calculated (Figs. 7a, b) shows that there is rather good agreement between them; the small discrepancies in the intensity of the peaks are connected with the rough nature of the approximations of the shape of the dumbbell-shaped domains. A similar method for calculating the diffraction pattern was also used in the case of the domain structures shown in Figs. 2a to c for which in the system of coordinates with the x axis horizontal and the y axis vertical the vectors of the primitive translations were equal to $\mathbf{a}_1 = \{a_1, 0\}$ and $\mathbf{a}_2 = \{0, a_2\}$, where a_1 is the distance between the centers of the domains in the rows and a_2 is twice the distance between the rows. Two quasielliptical domains (with centers at (0,0)and $(a_1/2, a_2/2)$) enter into the elementary cell, with major axes which can be either the same (as in Figs. 2a, b) or different (as in Fig. 2c). In the first case, these axes make angles $-\theta_e$ and $+\theta_e$ with the x axis, with $\pi/4 \ge |\theta_e| \ge 0$ (see also Fig. 2 in Ref. 1 or Fig. 9 in Ref. 3).

The calculated diffraction patterns for the static domain configurations depicted in Figs. 2a-2c are shown in Fig. 8 (going over to dynamic domain structures does not change the form of the diffraction patterns). In the calculations, we used corresponding actual values of the parameters. For Fig. 2a, $|\mathbf{a}_1|=90 \ \mu\text{m}$, $|\mathbf{a}_2|=173 \ \mu\text{m}$, $r_1=82 \ \mu\text{m}$, $k_r=0.34$; for Fig. 2b, $|\mathbf{a}_1|=50 \ \mu\text{m}$, $|\mathbf{a}_2|=150 \ \mu\text{m}$, $r_1=71 \ \mu\text{m}$, $k_r=0.39$; for Fig. 2c, $|\mathbf{a}_1|=50 \ \mu\text{m}$, $|\mathbf{a}_2|=150 \ \mu\text{m}$, $r_1^{(l)}=85 \ \mu\text{m}$, $k_r^{(l)}=0.33$, $r_1^{(s)}=50 \ \mu\text{m}$, $k_r^{(s)}=0.64$. The indices (l) and (s) refer, respectively, to the long and the short domains. As before, the intensity of the diffraction spots is represented by the area of the corresponding circular spots, with centers at the sites of the reciprocal lattice with basis vectors

$$\mathbf{b}_1 = \frac{2\pi}{a_1} \{1;0\}; \quad \mathbf{b}_2 = \frac{2\pi}{a_2} \{0;1\}.$$

We show diffraction peaks with an intensity exceeding 2% of the intensity of the largest spot for crossed transmission planes of the analyzer and the polarizer. Since for the given domain structures the dual modifications differ from one another only by translations of half a period, the shape of the diffraction patterns for them (and thus for the dynamic configurations as well) are identical. The experimental and calculated diffraction patterns are in good agreement. It is clear that when the angle ψ between the major axes of the domains in neighboring rows decreases, the fraction of light transferred into diffraction peaks positioned to the right and to the left of the vertical symmetry plane of the diffraction pattern increases.

We also carried out theoretical calculations of the diffraction of optical radiation by ordered domain structures formed in the case of dynamic clustering of heteropolar CMD lattices (Fig. 5), and in the case of drift of twodimensional domain blocks (Fig. 6). In the latter, the domains was approximated by polygons. The results were found to be in good agreement with the experimentally observed diffraction patterns.

The present theory addresses the diffraction of light by fully ordered domain structures, i.e., by two-dimensional lattices with long-range order. Since the Fourier transform of the intensity of the diffracted radiation in the present case is proportional to the correlation function of the z component of the magnetization vector **M**, an analysis of experiments on the diffraction of light by quasiregular domain structures can yield certain information about the degree of ordering of the domains in actually observed structures. The experimental results (see also Refs. 36 and 37) show that most often the following types of breaking of the order in domain structures are met with.

1. Deviation of the position of the centers of symmetry of the domains from the sites of a geometrically regular equidistant two-dimensional lattice (positional disorder).

2. Noncoincidence of the orientation of the symmetry axes of positionally equivalent domains in different cells of the two-dimensional lattice (orientational disorder).

3. Different shapes of single-type (but not necessarily positionally equivalent) domains in the same cell or different cells (morphological disorder).

4. Magnetic point or line defects such as vacancies, excess domains, dislocations, or disclinations (topological disorder).

Such deviations from regularity in the arrangement, orientation, and shape of the domains leads to a blurring of the diffraction reflections, a diminution of their intensity, and an additional attenuation of the higher-order diffraction peaks. From the nature of the change in the diffraction pattern, one can reach some conclusion about the dominant kind of disordering in a given domain structure. If, for instance, the diffraction peaks have a tendency to get blurred in an arc, this indicates that the orientational disorder dominates, and so on.

6. CONCLUSION

The studies we carried out show that in the case of dynamic self-organization of domain structures in uniaxial films under the action of a pulsating magnetic field, one can realize several qualitatively different scenarios for the transition from chaos to order. A characteristic feature of the scenarios which occur in the experiments described in the present paper is the occurrence of two stages where the first stage—the formation of a hexagonal lattice of circular CMD from the domain structure labyrinth—is common to all scenarios and characterized by a change in the number of domains in the film, i.e., by a modification in the degree of connectivity of the space. It is well known¹⁹ that such nonequilibrium phase transitions in magnetic films are connected with irreversible changes in the topological structure of the domain boundaries, namely with the multiple generation of vertical Bloch lines (VBL). The second stage of the selforganization processes considered by us, which is highly individual and attributes the scenario to one type or another, conserves the number of domains, i.e., there is no change in the degree of connectivity of the space. The transformation of the structure of the domain boundaries (even if it occurs) probably does not play an important role in this case.

The experiments on the magneto-optic diffraction of light by domain structures appearing in the self-organization process show clearly that an analysis of the intensity distribution of the diffracted radiation can be used both for a symmetry analysis of the quasi-two-dimensional domain blocks appearing in the self-organization process and for a determination of its degree of order.

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- ⁴⁾In the initial labyrinth structure, the two possible directions of the magnetization vector $(\mathbf{M}\uparrow\uparrow\mathbf{n} \text{ and } \mathbf{M}\uparrow\downarrow\mathbf{n})$ are equivalent.
- ⁵⁾In the case of pulsed remagnetization of films, a similar effect, called a topological switching of the polarity of the CMD lattice, was observed and described by O'Dell.¹⁴
- ⁶⁾It is unclear whether the strong rhombic anisotropy in film No. 4 plays any role in the appearance of the drift. Our studies show, nonetheless, that in films with $\beta_u \ge \beta_p$ we could not achieve a stable translational motion of two-dimensional domain blocks in the absence of an appreciable inhomogeneity of the film properties in the extended plane.
- $^{7)}$ In the case of x-ray diffraction, the various kinds of symmetry of the diffracted radiation are called Laue classes.^{30,31}
- ⁸⁾If one takes into account circular magnetic dichroism (the difference between the optical absorption coefficients for characteristic waves with right- and left-hand circular polarizations), the lattice becomes an amplitude-phase diffraction lattice. This, however, does not lead to a qualitative modification of the diffraction: one has only intensities of the diffraction peaks and light polarization in the zeroth diffraction order (see, e.g., Refs. 8 and 9).

¹⁾In contrast to scenarios for the transition of dynamical systems from deterministic behavior to chaos (Ruelle-Takens-Newhouse, Pomeau-Manneville, and Feigenbaum), for which a satisfactory mathematical description and physical interpretation has been worked out (see, e.g., Ref. 10), there exists no unified approach to the classification of possible scenarios for the emergence of ordered structures from chaos.

²⁾The transition from a simple to a complex lattice is realized by means of the motion of the phase front from new phase nuclei; their role in this case is played by the elongated domains (see the photographs of Fig. 1 in Ref. 3).

³⁾We note that the filling factor for domain structures like those in Fig. 1 and Fig. 2 are essentially completely predetermined by the values of p_f for the initial simple hexagonal lattices of circular CMD, from which the more complicated configurations of Fig. 1 and Fig. 2 are formed in the self-organization process.

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