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# DISSIPATIVE SELF-ORGANIZATION OF A SINGLE LAYER OF FERROSUSPENSION PARTICLES IN A HIGH-FREQUENCY MAGNETIC FIELD OF DIFFERENT POLARIZATION

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*Structural transformations in a plane layer of spherical soft-magnetic particles on exposure to a rotating elliptically polarized field whose plane of polarization is perpendicular to the layer's plane have been studied by the molecular-dynamics method. The influence of the concentration of the particles and the polarization of the field on the kinetics of aggregation and the equilibrium form of disperse structures has been studied in the high-frequency limit.*

**Introduction.** Aggregation processes occurring in liquid-disperse magnetic systems under the action of electric and magnetic fields have been studied for several recent decades because of the possibility of controlling electromagnetically the physical properties of these systems, primarily their fluidity [1]. When the external field is applied, the particles become magnetized (polarized) and form chains or bundles oriented fieldwise, which produces a sharp change in the rheological properties of the system (e.g., gives rise to the yield stress) [1–3]. Interest in structural transformations caused by the exposure to a rotating magnetic field is comparatively recent [4–6]. The stationary dynamic state of a system in this case is dependent on two dimensionless parameters: the concentration of particles $c$ and the relative rotational frequency of the field $\nu$, which is determined as the ratio of the frequency $\omega$ to the critical frequency $\omega^* = \pi M^2/(18\eta)$ of rotation (synchronous with the field) of a pair of contacting magnetic particles. The behavior of ensembles of magnetic particles in the field of circular polarization is determined by the distinctive feature of pair interparticle interaction that the plane of rotation of the field is attracting for the pair of particles [7]. As a consequence, a bulk disperse structure consisting of plane granules oriented in the plane of rotation of the field is formed in the high-frequency limit. The possibility of forming a dissipative structure in a liquid polymer matrix with its subsequent fixation due to the polymer cross-linking can be used for the production of composite materials with special structure and properties [8]. The dynamic structure of the suspension can be controlled additionally by changing the configuration of a rotating field. The behavior of a two-dimensional system of particles in a liquid matrix on exposure to an elliptically polarized field whose polarization ellipse lies in a plane which is perpendicular to the layer and whose long semiaxis is parallel to the particle plane has been considered in [9] for the particular case $c = 0.3$. Below, we consider the influence of the concentration of particles and the polarization of the field and its orientation relative to the particle layer on the kinetics of structurization and on the character of final structures.

**Numerical Model.** We model the behavior of the system of interacting particles in the elliptically polarized field in a dipole-dipole approximation, disregarding inertial and hydrodynamic-interaction forces. The calculation is made in the high-frequency limit, when the displacement of the particles over the period of rotation of the field is negligible and the behavior of the system on times longer than the period of variation in the field can be described by the stationary dipole-dipole-interaction pseudopotential obtained by averaging the true potential over orientation variables over the period of variation in the field. In the case of the elliptically polarized field the expression for the pseudopotential has the form [9]

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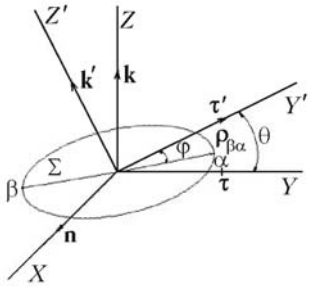



Fig. 1. Position of a pair of particles in space.

$$\langle U_{\beta\alpha}\rangle = \frac{a^6\langle H^2\rangle}{R_{\beta\alpha}^3} - \frac{3a^6\langle H^2\rangle}{R_{\beta\alpha}^3}\frac{1}{2-\varepsilon^2}\left[(1-\varepsilon^2)\,(1-(\boldsymbol{\rho}_{\beta\alpha}\mathbf{n})^2) + \varepsilon^2\,(\boldsymbol{\rho}_{\beta\alpha}\boldsymbol{\tau})^2\right]. \tag{1}$$

The mutual position of particles and the field is shown in Fig. 1. The polarization ellipse is tied to the Cartesian coordinate system $X$, $Y$, $Z$ so that the plane of polarization lies in the $Y$, $Z$ plane; the $Y$ axis is coincident with the long axis of the polarization ellipse. The positive directions of the $X$, $Y$, and $Z$ axes are denoted by the unit vectors $\mathbf{n}$, $\boldsymbol{\tau}$, and $\mathbf{k}$. The particles lie in the $\Sigma$ plane containing the $X$ axis. This plane is tied to the dashed coordinate system $X'$, $Y'$, $Z'$. The position of the $\Sigma$ plane relative to the long axis of the polarization ellipse is prescribed by the angle $\theta$. Therefore, the position of a pair of particles in space is determined by the distance between their centers, the angle of orientation of the particle-containing plane $\theta$, and the angle of deflection of the straight line connecting the centers of the particles from the plane of polarization $\varphi$.

The magnetic force with which a particle $\beta$ acts on $\alpha$ will be written in the form

$$\mathbf{F}_{\mathrm{m}}^{\beta\alpha} = -\frac{\partial\langle U_{\beta\alpha}\rangle}{\partial\mathbf{R}_\alpha} = \frac{3a^6\langle H^2\rangle}{R_{\beta\alpha}^4}\frac{1}{2-\varepsilon^2}\left[(2\varepsilon^2-1)\,\boldsymbol{\rho}_{\beta\alpha} + \varepsilon^2\left[2\boldsymbol{\tau}\,(\boldsymbol{\rho}\boldsymbol{\tau}) - 5\boldsymbol{\rho}\,(\boldsymbol{\rho}\boldsymbol{\tau})^2\right] - (1-\varepsilon^2)\left[2\mathbf{n}\,(\boldsymbol{\rho}\mathbf{n}) - 5\boldsymbol{\rho}\,(\boldsymbol{\rho}\mathbf{n})^2\right]\right].$$

To rule out the interpenetration of particles we introduce the model force $\mathbf{F}'_{\beta\alpha}$ equal to zero for $R_{\beta\alpha} \ge 2a + \delta$ and $\mathbf{F}'_{\beta\alpha}(\psi) = k[P\psi^2 + Q\psi + S]\boldsymbol{\rho}_{\beta\alpha}$ for $R_{\beta\alpha} < 2a + \delta$, where $\psi = R_{\beta\alpha} - 2a$. The constants $P$, $Q$, and $S$ are found to be equal to $1/\delta^2$, $-2/\delta^2$, and 1 respectively from the conditions $F'(\delta) = 0$, $F'(0) = 1$, and $\left.\dfrac{\partial F'}{\partial x}\right|_{x=\delta} = 0$. In numerical experiments, we select the values $k = 0.6$ and $\delta = 0.1$. Boundary conditions will be specified below.

The force acting on the particle $\alpha$ on the source side of all the remaining particles, with allowance for the impermeability of the particles, will be found by summation of the pair-interaction forces

$$\mathbf{F}_\alpha = \sum_{\beta\ne\alpha}\mathbf{F}_{\beta\alpha}, \quad \mathbf{F}_{\beta\alpha} = \mathbf{F}_{\beta\alpha}^{\mathrm{m}} + \mathbf{F}'_{\beta\alpha}.$$

The system of equations of particle motion in the Stokes approximation has the form

$$\frac{d\mathbf{R}_\alpha}{dt} = \frac{\mathbf{F}_\alpha}{6\pi\eta a}, \quad \alpha = 1, 2, ..., N. \tag{2}$$

Using the distance $2a$ and time scales $t^* = 18\eta/(\pi M^2)$ and the notation $\tau = t/t^*$ and $\mathbf{r} = \mathbf{R}/(2a)$, we reduce (2) to the dimensionless form

$$\frac{d\mathbf{r}_\alpha}{d\tau} = \mathbf{f}_\alpha, \quad \mathbf{f}_\alpha = \sum_{\beta\ne\alpha}\mathbf{f}_{\beta\alpha}^{\mathrm{m}} + \sum_{\beta\ne\alpha}\mathbf{f}'_{\beta\alpha}, \quad \alpha = 1, 2, ..., N;$$

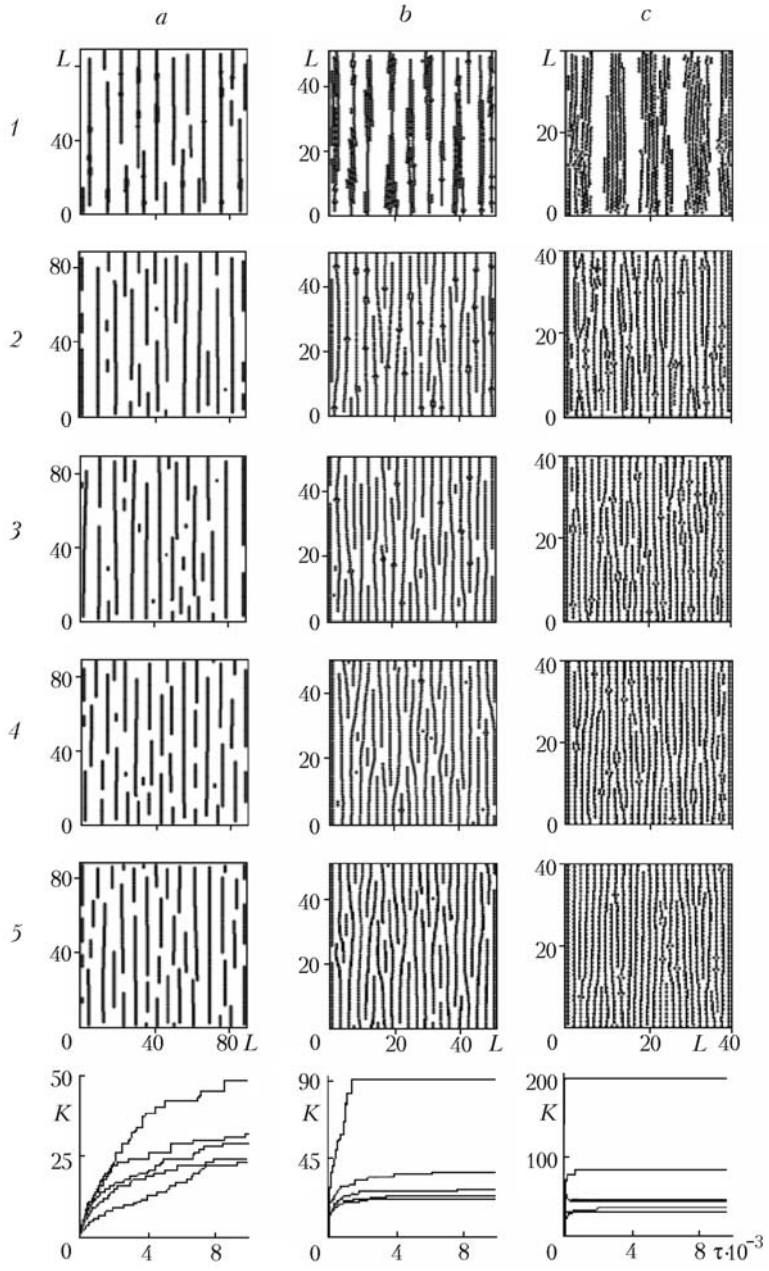
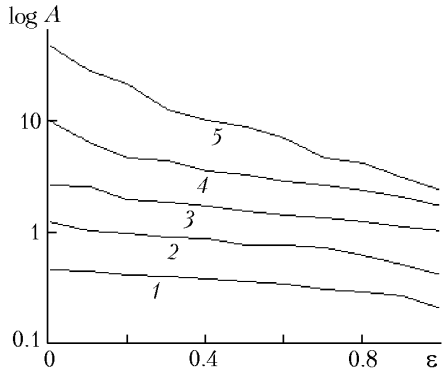



Fig. 2. Structures formed in the fields of different polarization for certain concentrations of particles and the corresponding changes in the average cluster size (a) $c$ = 0.1, b) 0.3, and c) 0.5): 1) linear polarization; 2) ε = 0.3, 3) 0.5, and 4) 0.7; 5) circular polarization.

$$\mathbf{f}_{\beta\alpha} = \frac{1}{2r_{\beta\alpha}^4}\frac{1}{2-\varepsilon^2}\left[(2\varepsilon^2-1)\,\boldsymbol{\rho}_{\beta\alpha} + \varepsilon^2\left[2\boldsymbol{\tau}\,(\boldsymbol{\rho}\boldsymbol{\tau}) - 5\boldsymbol{\rho}\,(\boldsymbol{\rho}\boldsymbol{\tau})^2\right] - (1-\varepsilon^2)\left[2\mathbf{n}\,(\boldsymbol{\rho}\mathbf{n}) - 5\boldsymbol{\rho}\,(\boldsymbol{\rho}\mathbf{n})^2\right]\right], \tag{3}$$

$$\mathbf{f}'_{\beta\alpha} = k\left[1 - 2\,\frac{r_{\beta\alpha}-1}{\delta} + \left(\frac{r_{\beta\alpha}-1}{\delta}\right)^2\right]\boldsymbol{\rho}_{\beta\alpha}\,.$$

**Numerical Experiment.** We consider an ensemble of $N$ particles placed in a square domain with side $L$ by the random-number generator. The surface concentration of the particles $c$ is determined as the ratio of the area occupied by the particles to the area of the particle-containing box $c = \pi N/(4L^2)$. The polarization of the field and its ori-



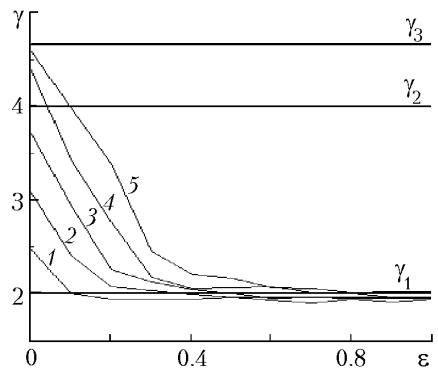




Fig. 3. Rate of growth in the average cluster size vs. polarization of the field for different concentrations: 1) $c$ = 0.1, 2) 0.2, 3) 0.3, 4) 0.4, and 5) 0.5.

Fig. 4. Average number of neighbors vs. polarization of the field for different concentrations. Notation 1–5 is the same as in Fig. 3.

entation relative to the particles' plane are determined by prescription of the parameters ε and θ. Let us dwell on two θ values: the first value (θ = 0) corresponds to the orientation of the larger semiaxis of the polarization ellipse in parallel to the layer, whereas the second (θ = π/2) corresponds to that perpendicular to the layer. The parameter ε in both cases is variable from 0 (linearly polarized field) to 1 (field of circular polarization) with a step of 1/10.

Let the large semiaxis of the polarization ellipse lie in the particles' plane (θ = 0). At the boundary of the domain, we specify the impermeability conditions: if the distance to the boundary Δ is smaller than $2a + \delta$, the particle is artificially "shifted" by the distance $\Delta = 2a + \delta$. The dimensionless computation time is bounded by the value $\tau = 10^4$. Computations are performed for particle concentrations of 0.1 to 0.5.

We will characterize the structures quantitatively by the average cluster size $K$ determined as the ratio of the number of particles $N$ to the total number of clusters of all sizes and the average number of neighbors γ according to the relation $\gamma = \sum_{\alpha=1}^{N} \vartheta_\alpha / (N - \vartheta_w)$. The algorithms of computing $K$ and γ have been given in [9]. Qualitatively, the structures will be characterized by the presence of defects classified as a "composite" one, a "square," and a "cross" [9].

A number of structures formed under different conditions are shown in Fig. 2. The corresponding growth in the average cluster size with time is given in the same figure (the upper branch corresponds to the field of linear polarization, whereas the lower one corresponds to the field of circular polarization). As we can see, the growth in the clusters is the most intense in the initial step of formation of the structure and can be approximated by a dependence of the form $K = K_0 + A(c, \varepsilon)\tau^{1/2}$, where $A$ is a function dependent on the concentration of particles and the polarization of the field. Figure 3 gives the change in the quantity $A$ as a function of the field's polarization for the concentrations considered. In the case of diluted suspensions ($c \le 0.3$) this dependence can be approximated by a function of the form $A \sim c^{0.35}(1 - \varepsilon)^{0.25}$. It is noteworthy that the process of aggregation is the faster the higher the concentration of the dispersed phase and the closer the field's polarization to the linear one.

The result of computations of the average number of neighbors in the structures formed as a function of the polarization of the field for different concentrations is shown in Fig. 4. Also, this figure gives the basic values: $\gamma_1$ = 2, the average number of neighbors for an isolated chain of $N$ particles, $\gamma_2$ = 4, for a bundle of two chains, and $\gamma_3$ = 4.67, for a bundle of three chains. It is noteworthy that isolated defect-free chains are formed in the field of circular polarization for all the concentrations studied. Defects appear in the chain structure as the polarization approaches linear; the higher the concentration of the dispersed phase, the larger the ε value for which this occurs. Defects formed in the process of structure formation under different conditions are shown in Fig. 5. As we can see, increase in the concentration of the filler and decrease in ε contribute to their formation.

Let the larger semiaxis of the polarization ellipse be perpendicular to the particles' plane, θ = π/2. Preliminary numerical experiments have shown that, for a certain ε, we have a qualitative change in the character of the

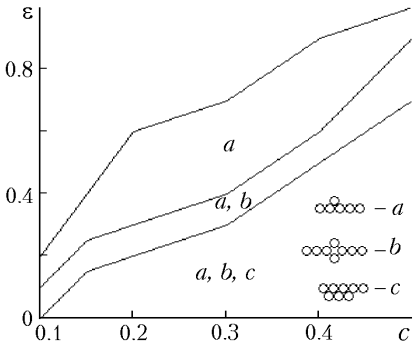



Fig. 5. Forms of defects and their relation to the polarization of the field and the concentration of the dispersed phase: a) defect of the "cross" type; b) "square"; c) "composite" defect.

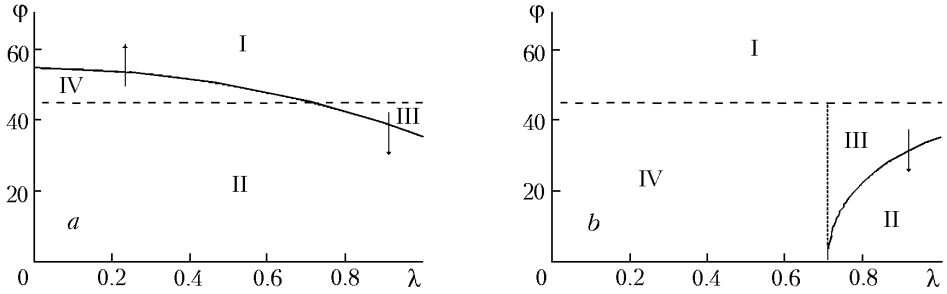



Fig. 6. On consideration of the behavior of a pair of particles as a function of the relation of critical angles: a) θ = 0; b) π/2.

structures formed: particles in the chains cease to be in contact. Since the latter is due to the character of the interaction of particles, which is determined by the shape and orientation of the polarization ellipse, we will consider this in greater detail. Expression (1) for pseudopotential is factorized and, allowing for the relations $(\boldsymbol{\rho}\cdot\boldsymbol{\tau}) = (\boldsymbol{\rho}\cdot\boldsymbol{\tau}')\cos\theta = \cos\varphi\cos\theta$ and $(\boldsymbol{\rho}\cdot\mathbf{n}) = \sin\varphi$, we have

$$\langle U_{\beta\alpha}\rangle = \Psi(R_{\beta\alpha})\,\Omega(\lambda,\theta,\varphi), \quad \Psi(R_{\beta\alpha}) = \frac{a^6\langle H^2\rangle}{R_{\beta\alpha}^3}, \quad \Omega(\lambda,\theta,\varphi) = 1 - 3\cos^2\varphi\,\frac{1}{1+\lambda^2}\left[\lambda^2 + (1-\lambda^2)\cos^2\theta\right].$$

The component Ψ determines the radial component of the interparticle-interaction force, and Ω determines the azimuthal component. The direction of the radial force is determined by the sign of the derivative $-\dfrac{\partial\langle U_{\beta\alpha}\rangle}{\partial R_{\beta\alpha}}$, i.e., by the sign of the function Ω. Particles are attracted when $\Omega > 0$ and are repelled when $\Omega < 0$. The equation $\Omega(\lambda, \theta, \varphi) = 0$ or

$$\cos^2\varphi = \frac{1}{3}\,\frac{1+\lambda^2}{\lambda^2 + (1-\lambda^2)\cos^2\theta}$$

for prescribed orientation and eccentricity of the polarization ellipse determines, in the plane of motion Σ, the boundary $\varphi_r^*$ between sectors in which particles are mutually attracted ($\varphi < \varphi_r^*$) or repelled ($\varphi > \varphi_r^*$).

The orientation attraction or repulsion of a pair of particles from the plane of polarization of the field is determined by the sign of the partial derivative

$$-\frac{\partial\Omega}{\partial\varphi} = -\frac{3}{1+\lambda^2}\left[\lambda^2 + (1-\lambda^2)\cos^2\theta\right]\cos 2\varphi\,.$$

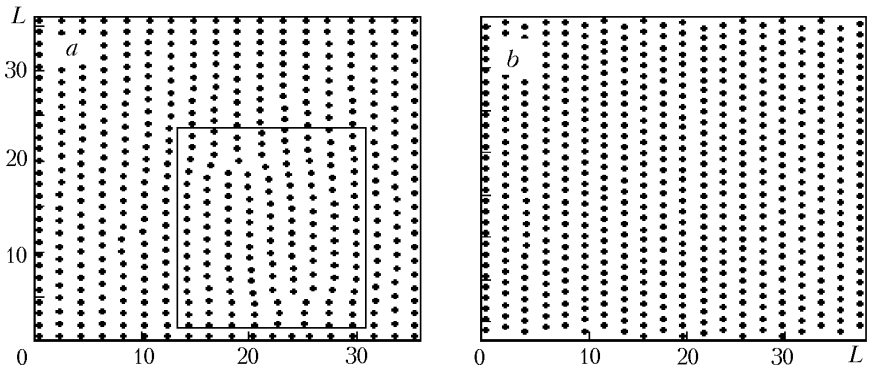



Fig. 7. Result of the use of periodic boundary conditions.

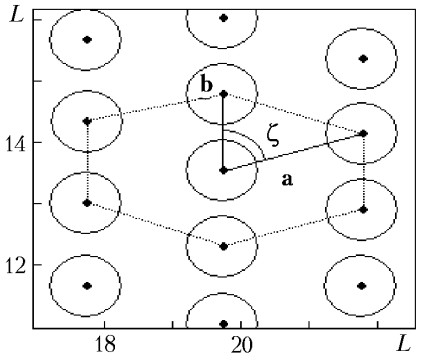



Fig. 8. Unit cell and fundamental periods for analysis of the structure formed in elliptically polarized fields with $\theta = \pi/2$.

The expression in square brackets here is positive and vanishes only when the conditions $\lambda = 0$ and $\theta = \pi/2$ are simultaneously observed, which corresponds to the field of linear polarization acting perpendicularly to the particles' plane Σ. Consequently, the plane of polarization is attracting for a pair of particles for all values of the parameters of the polarization ellipse on condition that the angle φ of orientation of the pair lies in the sector from $-\varphi_a^*$ to $\varphi_a^*$, where the value $\varphi_a^* = \pi/4$ is determined from the condition $\cos 2\varphi = 0$.

The critical angles $\varphi_a^*$ and $\varphi_r^*$ divide the plane Σ into four parts shown in Fig. 6a for the case $\theta = 0$ and in Fig. 6b for $\theta = \pi/2$. When $\theta = 0$, in region I, particles are repelled from both the plane of polarization and each other; in zone II, we have both the radial and orientation attraction. In zone III, particles are repelled from each other but are attracted in the plane of polarization; consequently, they will arrive at zone I and will begin to be attracted. In zone IV, the opposite situation occurs: particles, being attracted to each other at first, will arrive at zone II and will begin to be repelled.

A different situation is observed for $\theta = \pi/2$. In the first, second, and third regions, the behavior of particles is analogous to the previous case, whereas in the fourth zone, particles are attracted to the plane of polarization but are always repelled from each other. The threshold λ value is $1/\sqrt{2}$.

An example of such a structure for $c = 0.3$ and $\varepsilon = 0.5$ is shown in Fig. 7a. To eliminate the influence of the boundaries which gives rise to defects (it is marked in Fig. 7a) we specify periodicity conditions and increase the dimensionless computation time to $2 \cdot 10^5$. The result is shown in Fig. 7b. Noticing that the arrangement of particles is similar to the arrangement of atoms in the two-dimensional model of a crystal, we will speak of the structure formed as of a crystal lattice and will characterize it by the fundamental periods. The unit cell and the fundamental periods are shown in Fig. 8 (part of the square region for the case $c = 0.3$, $\varepsilon = 0.5$, and $\tau = 2 \cdot 10^5$ is shown). The change in the periods with ε for different $c$ values is illustrated by Fig. 9a. Decrease in both **a** and **b** for all ε values is observed with growth in the concentration. The same square (Fig. 9b) shows, on a log-log scale, the linear change in the periods with growth in the concentration for two boundary values of ε. Intermediate polarizations lie between these two values and are not shown so as not to clutter the figure. Polarization influences these periods in different manners:

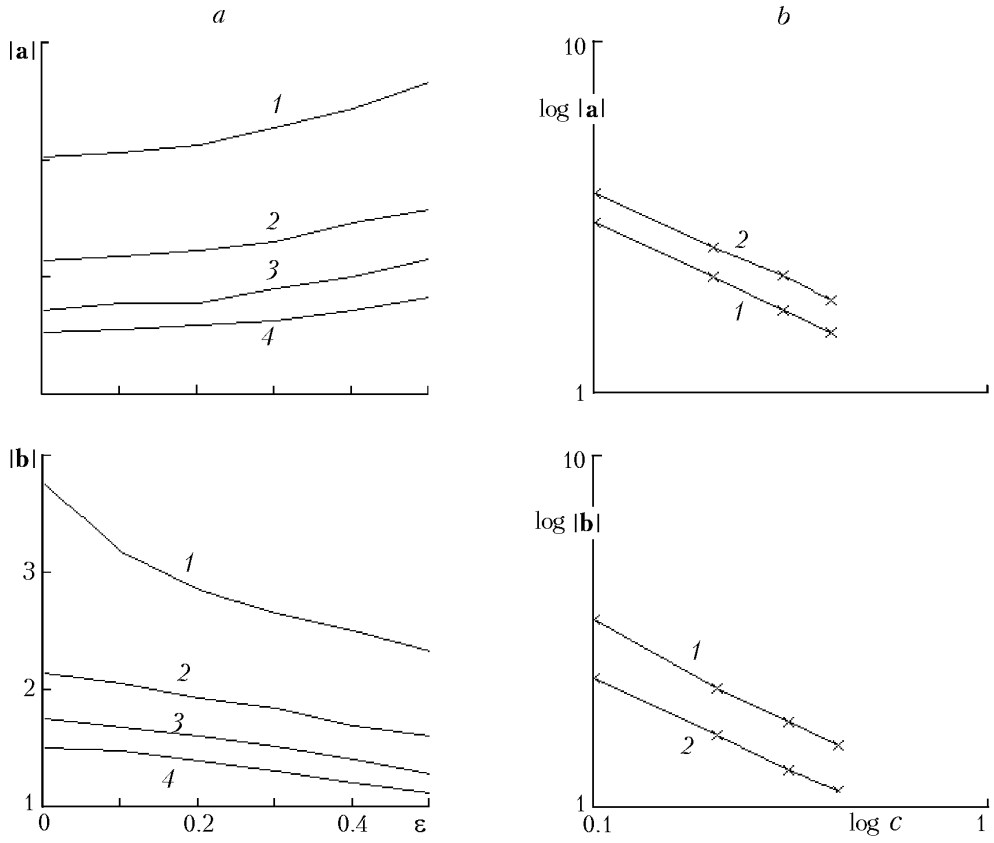



Fig. 9. Fundamental periods vs. polarization of the field (a) (1) $c$ = 0.1, 2) 0.2, 3) 0.3, and 4) 0.4) and vs. concentration of the filler (b) (1) $\varepsilon = 0$ and 2) 0.5).

thus, **a** decreases with ε, whereas **b** increases. For the angle φ we note that it is virtually independent of concentration up to the polarization $\varepsilon = 0.2$ and decreases with ε. As the polarization of the field approaches linear, $\varepsilon \leq 0.2$, the angle changes in an unpredictable manner, which is due to the loss of the separated direction in the layers' plane; this direction in the case in question is created by the smaller semiaxis of the polarization ellipse, which lies in the particles' plane.

**Conclusions.** The above results demonstrate the possibility of controlling the dissipative structure of ferroparticles dispersed in the liquid by variation of the polarization of a rotating field and the concentration of a dispersed phase, which is of interest for problems of electromagnetic formation of filled polymer materials with special properties.

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## NOTATION

$a$, particle radius; $A$, rate of growth in the average cluster size; **a** and **b**, fundamental periods of a unit cell; $c$, surface concentration of particles; **f**, dimensionless force; **F**, force; **F′**, model force; **H**, magnetic field; $\langle H^2 \rangle$, average value of the square of the field; $H_{0z}$ and $H_{0y}$, vector components; $k$, constant; **k**, unit vector; $K$, average cluster size; $L$, dimension of the square region; $M$, magnetization; **n**, unit vector; $N$, number of particles; $P$ and $Q$, constants; **r**, dimensionless distance; **R**, radius vector; $S$, constant; $t$, time; $t^*$, time scale; $\langle U \rangle$, pseudopotential of dipole-dipole interaction; $X$, $Y$, $Z$, axes of the coordinate system; $Y'$, $Z'$, axes of the coordinate system tied to the Σ plane; ν, relative rotational frequency of the field; λ, ratio of the semiminor axis of the polarization ellipse to the semimajor axis, $\lambda = H_{0z}/H_{0y}$; η, viscosity; ε, eccentricity of the polarization ellipse, $\varepsilon = (1 - \lambda^2)^{1/2}$; **ρ**, unit radius vector, $\boldsymbol{\rho} = \mathbf{R}/R$; γ, average number of neighbors; $\gamma_1$, $\gamma_2$, and $\gamma_3$, basic values of the average number of neighbors; τ, dimensionless time; **τ**, unit vector; **τ′**, unit vector in the coordinate system $X$, $Y'$, $Z'$; $\vartheta_\alpha$, number of neighbors of the particle α; $\vartheta_w$, number of particles located at a distance smaller than unity from the boundaries of the domain; Δ, distance to the

boundary; Σ, particle-containing plane; θ, angle of orientation of the particle-containing plane; δ, constant; φ, angle of deflection of the straight line connecting the centers of particles from the plane of polarization; $\varphi^*$, critical value of the angle φ; ψ, distance between particle walls, $\psi = R_{\beta\alpha} - 2a$; Ψ, radial component of the force; Ω, azimuthal component of the force; ω, rotational frequency of the field; $\omega^*$, critical frequency. Subscripts: 0 corresponds to the initial instant of time; a, azimuthal; m, magnetic; r, radial; α and β, particle Nos.

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