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Dynamics of a flexible magnetic chain in a rotating magnetic field

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The model of an elastic magnetic rod is applied for a study of a behavior of the flexible magnetic particle chain in a rotating magnetic field. By numerical simulation it is shown that behavior of a flexible magnetic chain is characterized by the existence of a critical frequency beyond which the dynamics of the rod is periodic with subsequent stages of bending and straightening. The value of the critical frequency found is explained by a simple model. Below the critical frequency the chain is bent and rotates synchronously with a field. It is illustrated that in particular cases the considered model reproduces phenomena observed experimentally and numerically for the magnetic particle chains in magnetorheological suspensions. It is emphasized that the present approach gives the general framework for the description of different phenomena in magnetorheological suspensions.

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I. INTRODUCTION

The dynamics of extended objects like elastic rods has recently obtained a considerable interest [1–5]. This is caused by the understanding of the behavior of different filaments in biological systems [6–9] and macromolecules [10,11]. The objects behaving like flexible rods appear also in the magnetorheological suspensions where the chainlike aggregates form due to magnetic interactions [12–15]. The chains of magnetic particles have interesting biological applications [16]. We will illustrate in this paper that behavior of these objects under the action of the rotating magnetic field may be understood by the model of an inextensible magnetic rod proposed in Ref. [17]. The filamentary elastic magnetic objects of other kind are formed linking microsize functionalized paramagnetic particles with some polymers [18,19]. In this way a new system possessing the features of flexible polymers with strong magnetic properties is created. Elastic properties of such objects, using optical trapping technique, were determined recently [19]. It was found that the curvature elasticity constant C of the linked magnetic chain determined by the bending, compression, and relaxation experiments depending on the linker molecules is about $10^{-12}-10^{-14}$ erg cm. It should be mentioned that the curvature elasticity constant of the magnetic chain has contribution due to the magnetic interaction forces which can be estimated according to the relation [20]

$$C_m=\frac{(\mu_{ch}-1)^2}{2\pi}H_0^2\ln\left(\frac{2L}{d}\right)I, \tag{1}$$

where $I=\pi a^4/4$ is the inertia moment of the cross section of the chain approximated by the cylinder with the radius a. Estimating the magnetic permeability of the chain μ_{ch} as $1+3(\mu-1)/\mu+2$, where μ is the magnetic permeability of the paramagnetic particles, the relation (1) for the curvature elasticity constant of the chain of the particles with the diameter $d=0.78\ \mu$m and the length 30 μm aligned along the magnetic field lines at the field strength $H_0=310$ Oe [19] gives the value about 10^{-12} erg cm which has the order of magnitude of the curvature elasticity found in the experiment [19]. The curvature elasticity of the chain of magnetic particles connected with the another linker (glutaraldehyde) are stiffer [18] and for those chains the contribution of the magnetic interactions could be neglected. It should be also remarked that the curvature elasticity depends on the angle which the magnetic field makes with the chain causing quite interesting peculiarities in the behavior of the magnetic chains which will be considered elsewhere [20]. Another peculiarity of the magnetic chains is connected with the magnetic torque arising when the magnetic field is not parallel to the chain. The magnetic field torque is responsible for the rotation of the magnetic chains under the action of the rotating magnetic field [12–15]. For the dynamics of the magnetic chain in the rotating field several interesting features are observed as the formation of bent configuration and breaking at increase of the angular velocity of the field rotation. Balance of the chain breaking and growth due to axial coalescence leads to the characteristic chain length dependence on a rotating field frequency $\omega^{-1/2}$ observed in different experiments with magnetorheological suspensions. In the first section of the present work following Ref. [17] we formulate the model for the elastic magnetic chain in a rotating magnetic field. The numerical simulation results, illustrating its behavior, are given in the following section. Further the obtained results are considered from perspective of their application to the description of the behavior of the chains of magnetic particles in magnetorheological suspensions and good quantitative agreement with available experimental data is illustrated.

II. MODEL

According to the Kirchhoff model the energy of an elastic rod, including the magnetic term, has the form [21]

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$$E=\frac{1}{2}C\int\frac{1}{R^2}dl-\frac{2\pi^2a^2\chi^2H_0^2}{\mu+1}\int(\vec{h}\vec{t})^2dl-\int\Lambda dl, \tag{2}$$

where the term with a Lagrange multiplier Λ accounting for the local inextensibility is introduced. Here R is the radius of the curvature of the centerline; $\mu=1+4\pi\chi$, χ is the magnetic susceptibility of the rod, $\vec{t}$ is the tangent to the centerline of the rod. Considering the variation Eq. (2) at the changing position of the centerline $\vec{r}'=\vec{r}+\vec{\xi}$ we have

$$\delta E=[M\delta\varphi]+[F_t\xi_t]+[F_n\xi_n]-\int K_n\xi_n dl-\int K_t\xi_t dl. \tag{3}$$

Here [ ] denotes the difference of the values at the ends of the rod, ξ_n and ξ_t are components of the Lagrange displacement in the directions of the normal and tangent to the centerline, respectively, but $\delta\varphi=\partial\xi_n/\partial l-\xi_t/R$ is the angle of the tangent angle rotation at the Lagrange displacements $\vec{\xi}$. The tangent and normal vectors are connected according to the Frenet equation $d\vec{t}/dl=-1/R\vec{n}$, where l is the arc length of the rod's centerline. The binormal $\vec{b}$ to the centerline is defined by $\vec{b}=[\vec{t}\times\vec{n}]$. Only motions of the rod in the plane are considered and $\vec{t}=(\cos\theta,\sin\theta)$, where θ is the angle which the tangent makes with the magnetic field direction. According to the relation (3) the following expressions for the components of the body force $\vec{K}$, stresses $\vec{F}$ and momentum stresses $\vec{M}=M\vec{b}$ are valid,

$$F_n=C\left(\frac{1}{R}\right)_l+\frac{2\pi^2a^2\chi^2H_0^2}{\mu+1}\sin(2\theta), \tag{4}$$

$$F_t=-\left(\frac{C}{2R^2}+\Lambda\right), \tag{5}$$

$$K_n=\frac{dF_n}{dl}-\frac{F_t}{R}=C\left(\left(\frac{1}{R}\right)_{ll}+\frac{1}{2}\frac{1}{R^3}\right)+\Lambda\frac{1}{R}+\frac{2\pi^2a^2\chi^2H_0^2}{\mu+1}\frac{d(\sin(2\theta))}{dl}, \tag{6}$$

$$K_t=\frac{dF_t}{dl}+\frac{F_n}{R}=-\Lambda_l+\frac{1}{R}\frac{2\pi^2a^2\chi^2H_0^2}{\mu+1}\sin(2\theta), \tag{7}$$

$$M=-\frac{C}{R}. \tag{8}$$

The simplest case of Rouse dynamics for the rod $\zeta\vec{v}=\vec{K}$ [6] is taken, where ζ is the friction coefficient of the chain per length unit. The equation for the tangent angle reads

$$\frac{d\theta}{dt}=\frac{\partial v_n}{\partial l}-\frac{v_t}{R} \tag{9}$$

but the condition of a local inextensibility has the form

$$\frac{\partial v_t}{\partial l}+v_n\frac{1}{R}=0. \tag{10}$$

In the case of the rotating field in relations (4), (6), and (8) for the angle θ we must take $\tilde{\theta}-\omega t$, where $\tilde{\theta}$ is the angle which the tangent makes with x axis of the laboratory set of coordinates. Then introducing the phase lag $\beta=\omega t-\tilde{\theta}$ of the tangent from the magnetic field direction and introducing the characteristic time scale $\tau=\zeta L^4/C$, the length scale L, where $2L$ is the length of the rod, the following dimensionless equations for the phase lag and the tension of the rod are obtained:

$$\omega\tau=\beta_t+(\beta_{llll}+\tfrac{1}{2}(\beta_l^3)_l)+(\beta_l\Lambda)_l+\Lambda_l\beta_l-\mathrm{Cm}(\sin 2\beta)_{ll}+\mathrm{Cm}(\beta_l)^2\sin(2\beta) \tag{11}$$

and

$$\beta_l^2\Lambda-\Lambda_{ll}=-\beta_l(\beta_{lll}+\tfrac{1}{2}\beta_l^3)+\mathrm{Cm}\beta_l^2 2\cos 2\beta+\mathrm{Cm}(\beta_l\sin(2\beta))_l. \tag{12}$$

Here $\mathrm{Cm}=2\pi\chi^2H_0^2\pi a^2L^2/(\mu+1)C$ is the magnetoelastic number characterizing the ratio of the magnetic and elastic forces. Taking for the magnetic permeability, the radius of chain and the curvature elasticity constant the values given above for the parameter Cm at the chain length 60 μm in the magnetic field $H_0=10^2$ Oe we have the value about 30. The set of boundary conditions necessary for the resolution of the Eqs. (11) and (12) follow from the absence of the stresses and momentum stresses on the free ends of the rod which gives

$$-\beta_{ll}+\mathrm{Cm}\sin 2\beta=0, \tag{13}$$

$$\beta_l=0, \tag{14}$$

and

$$\Lambda=0, \tag{15}$$

at $l=\pm 1$.

The set of equations (11) and (12) with the boundary conditions Eqs. (13)–(15) is solved numerically by the implicit scheme approximating derivatives by the finite differences. The nonlinear equations for the values of the tangent angle at new time step are solved by the Newton method. Tension Λ for the current time step is found by tridiagonal solver.

III. NUMERICAL SIMULATION RESULTS

In Ref. [17] it was found that besides the trivial straight configuration of the rod in the static magnetic field "U" like turns are also possible as stable configurations. Such configurations do not exist under the action of the rotating field. The process of the transformation of U like turn in the rotating field is shown in Fig. 1. We see that the process of the hairpin transformation occurs with the constant velocity of

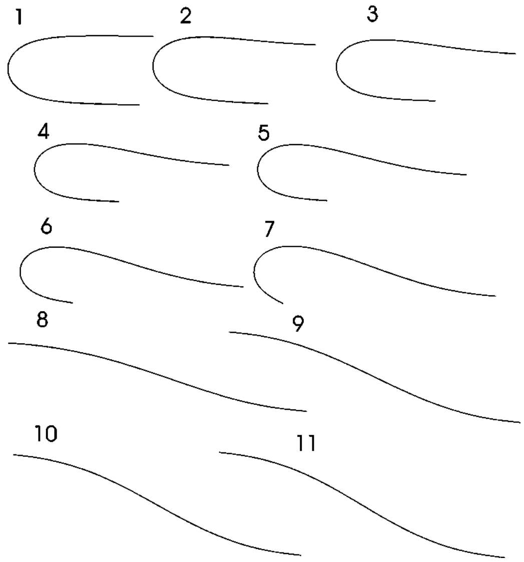

FIG. 1. Relaxation of U turn in rotating field from point of view of the set of coordinates connected with the field (magnetic field strength is along x axis). Cm=25; $\omega\tau=50$. $t=0.0087(1)$, 0.0174(2), 0.0260(4), 0.0347(5), 0.0434(6), 0.0521(7), 0.0608(8), 0.0694(9), 0.0781(10), 0.0868(11).

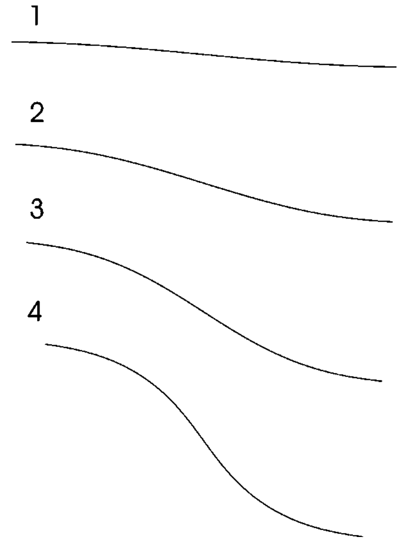

FIG. 3. Stationary rod configurations in rotating field from point of view of the set of coordinates connected with the field (magnetic field strength is along x axis). Cm=25, $\omega\tau=10(1)$, 30(2), 50(3), 60.6(4).

the U turn tip propagation. This finally ends with the fast process of the straightening of the rod. These features of the hairpin relaxation in the rotating field are shown in Fig. 2. From Fig. 1 we see that after relaxation process a stationary state of the elastic rod rotating with the angular velocity of the field is established. The extent of the bending of the rod increases with the frequency of the rotating field. This is illustrated by Fig. 3 where stationary configurations of the elastic rod for the different values of the rotating field frequency are shown. We see that the phase lag in the center of the rod increases with the frequency of the rotating field. The last configuration in Fig. 3 corresponds to the frequency close to the critical for given Cm when no stationary state of the rod motion arises. The distribution of the tension along the rod for one stationary configuration is shown in Fig. 4. Reaching the critical frequency the tangent angle in the center of droplet abruptly switches and "S" like configuration arises similarly to what has been observed in numerical simulation of viscous magnetic drop [22]. After process of the propagation of U turns the abrupt rotation of the ends of the rod takes place. As a result rather interesting periodic regime of the rod rotation arises. This sequence of the events is illustrated in Fig. 5 from the point of view of the laboratory set of coordinates. At high frequencies of the field rotation the mean angular velocity $\overline{\omega}$ of the chain rotation in the laboratory set of coordinates is small. The dependence of the angular velocity in the center of the rod on time for the periodic regime at $Cm=25, \omega=500$ illustrates this in Fig. 6. For the mean angular velocity

$$\overline{\omega}\tau=\omega\tau\left(1-\frac{1}{\pi}\int_0^{T/2}\frac{d\beta}{dt}dt\right), \tag{16}$$

in this case we have $\overline{\omega}/\omega=0.0022$. Similarly to the asynchronous regime of the magnetic dipole under the action of

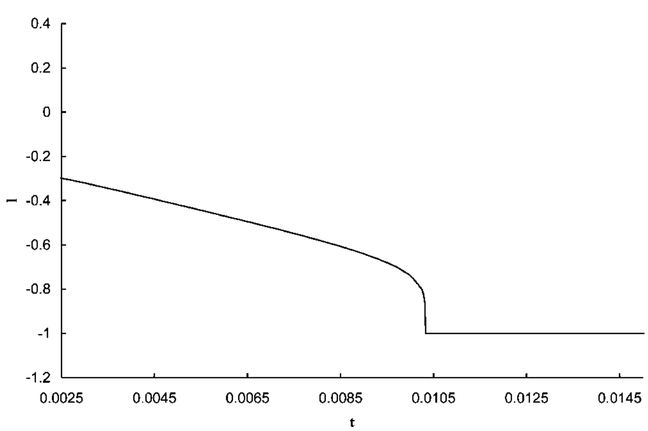

FIG. 2. Position of the U turn tip along the chain in dependence on time in the rotating field. Cm=25, $\omega\tau=50$.

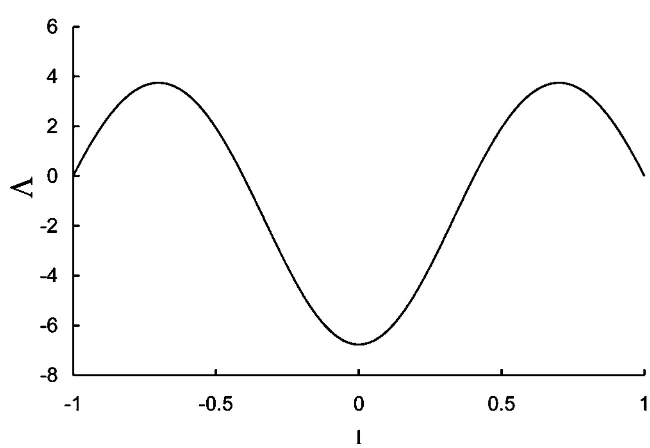

FIG. 4. Distribution of tension along the rod. Cm=25, $\omega\tau=50$.

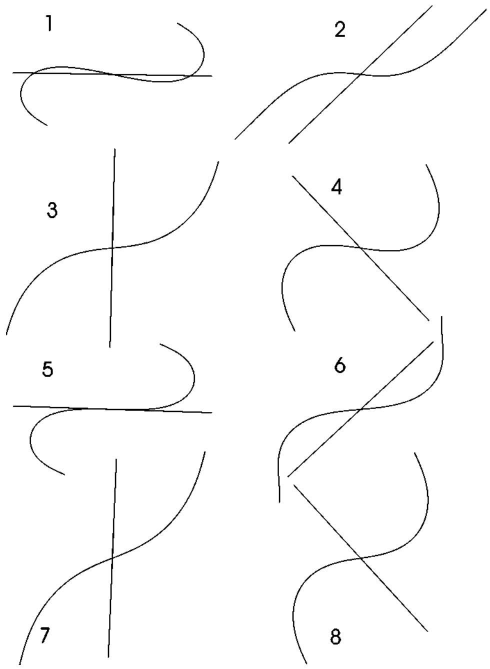

FIG. 5. Configurations of flexible magnetic chain from point of view of the laboratory set of coordinates. Straight line shows the magnetic field direction. Cm=25, $\omega\tau=300$. $t=0.0104(1)$, 0.01302(2), 0.0156(3), 0.0182(4), 0.0208(5), 0.0234(6), 0.0260(7), 0.0286(8).

the rotating field [23] forward and backward rotations of the flexible chain occur leading at high angular velocities of the field to observed small overall rotation.

The value of the critical frequency at which the switching of the tangent direction takes place depends linearly, as shown in Fig. 7, on the magnetoelastic number Cm. This means that similarly to what is taking place in the case of the magnetic droplet with finite surface tension the critical frequency is determined by Mason number [22]. Thus results in Fig. 7 can be explained by simple model. Assuming that tangential motion is negligible we can take $K_t=0$. Then Eq. (7) introducing dimensionless quantities and angle β can be

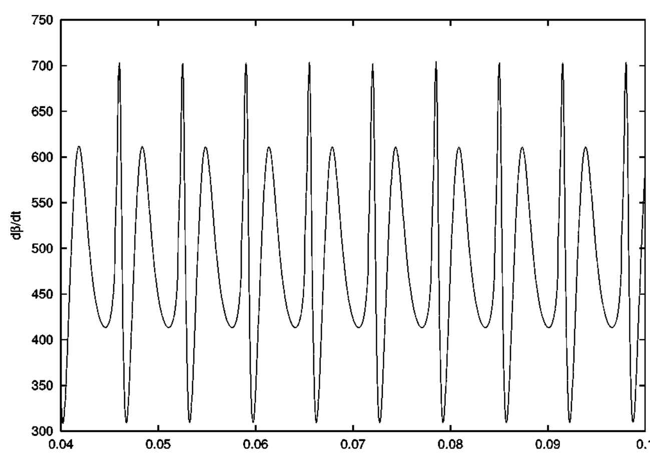

FIG. 6. Angular velocity in the center of the rod in dependence on dimensionless time in rotating frame. Cm=25, $\omega\tau=500$.

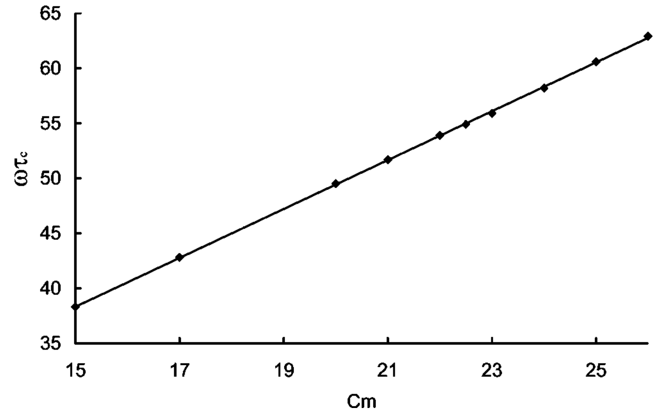

FIG. 7. Critical frequency in dependence on the magnetoelastic parameter.

integrated and accounted for the boundary condition on the free ends that results in

$$\Lambda=\frac{\mathrm{Cm}}{2}(\cos(2\beta)-\cos(2\beta_0)), \tag{17}$$

here $\beta_0=\beta(\pm 1)$. With that simple expression for Λ the last terms in Eq. (11) can be simplified what gives

$$\omega\tau=\beta_t+\beta_{llll}+\frac{1}{2}(\beta_l^3)_l-\left(\frac{\mathrm{Cm}}{2}\cos(2\beta_0)\beta+\frac{3\mathrm{Cm}}{4}\sin(2\beta)\right)_{ll}. \tag{18}$$

In this form the equation for the tangent angle is close to considered in Ref. [22] for the dynamics of the viscous drop in a rotating field. The bending elasticity term in Eq. (18) similarly to the viscous momentum stresses in Ref. [22] for large Cm is playing the regularizing role. In steady case when Cm is large the contribution of the elasticity terms can be neglected and Eq. (18) has the analytical solution [22]

$$\frac{3\mathrm{Cm}}{4}\left(\tfrac{2}{3}\cos(2\beta_0)\beta+\sin(2\beta)\right)=\tfrac{1}{2}\omega\tau(1-l^2). \tag{19}$$

The critical value of $\omega\tau$ for the formation of the jump of the tangent angle can be found from the value of the first maximum of the function $F_1(\beta)=\frac{2}{3}\cos(2\beta_0)+\sin(2\beta)$. This for the critical frequency [taking $\cos(2\beta_0)=1$ for large Cm] gives

$$(\omega\tau)_c=\mathrm{Cm}\left(\frac{\pi}{2}-\frac{1}{2}\arccos\left(\frac{1}{3}\right)+\frac{2\sqrt{2}}{3}\right), \tag{20}$$

which shows that for enough large Cm $(\omega\tau)_c=2.37$ Cm in good agreement with data given in Fig. 7. The dependence of the angle in the center of the droplet on Cm at the critical frequency is less trivial and the curve with minima has been obtained in numerical calculations as shown in Fig. 8. Nevertheless the values shown in Fig. 8 are very close to the value obtained from the first maximum of the function

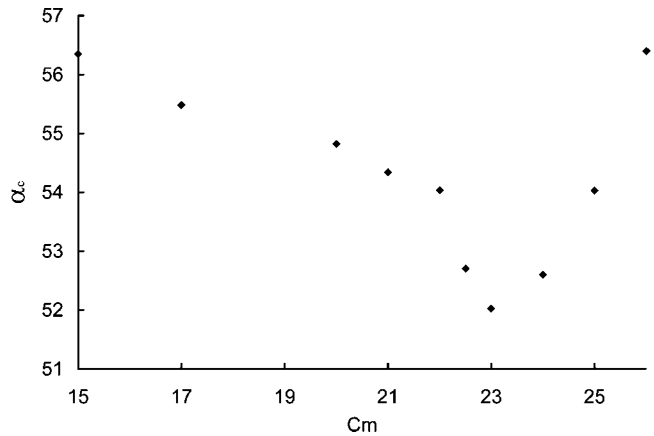

FIG. 8. Tangent angle in the center of the rod at critical frequency.

$F_1(\beta)$, $\beta_c=\frac{1}{2}(\pi-\arccos(\frac{1}{3}))$, which corresponds to the angle $\beta_c\approx 54.7$ deg. More complex dependence shown in Fig. 8 may be connected with the fact of more complicated behavior of the tension in the rod than given by Eq. (17). Nevertheless the main peculiarities of the flexible chain dynamics may be understood by the simple model given above.

IV. DISCUSSION OF THE EXPERIMENTAL RESULTS OF THE MAGNETIC CHAIN DYNAMICS IN MAGNETORHEOLOGICAL SUSPENSIONS

The model of an inextensible magnetic rod with some curvature elasticity, arising from the magnetic interactions, is also applicable to a chain of magnetic particles in a rotating field of a low frequency when a chain moves synchronously with a field. In this case it turns out that the tension in the chain arising due to the shearing normal forces is less than magnetic force holding the particles together and the chain behaves as an inextensible rod. In the region of the compression stresses they are balanced by the reaction forces due to the hard cores of the magnetic particles. Let us consider the chain of magnetic dipoles with hard core radius a. Let the angle of the chain axis with x axis be θ but the angle of the field φ. If the external magnetic field strength is larger than magnetic field strength due to neighboring particles in the chain then the angle α which the magnetization makes with x axis is close to θ. The energy of the particle interacting with its two neighbors in the chain is

$$E=-mH_0\cos(\varphi-\alpha)+2U,$$

where

$$U=\frac{m^2}{(2a)^3}(1-3\cos^2(\alpha-\theta)).$$

The condition $\partial E/\partial\alpha=0$ for the magnetic torque on the particle K gives

$$K=mH_0\sin(\varphi-\alpha)=\frac{6m^2}{(2a)^3}\sin 2(\varphi-\theta).$$

As a result the torque per length unit $k=K/2a$ is

$$k=\frac{6m^2}{(2a)^4}\sin 2(\varphi-\theta).$$

It is easy to see that the normal shearing force $F_n=-2F_\theta=2/2a\,\partial U/\partial\theta=-6m^2/(2a)^4\sin 2(\varphi-\theta)$ arising due to the magnetic interactions between neighboring particles is equal to its value $F_n=-k$ in the model of an inextensible magnetic rod without momentum stresses. In the case when the curvature radius of the rod is large enough in comparison with its length the tension in the rod Λ is determined by the shearing normal force

$$\Lambda_l=\frac{1}{R}\frac{6m^2}{(2a)^4}\sin 2(\theta-\varphi), \tag{21}$$

which corresponds to that obtained from the relation Eq. (7) when the following substitution

$$\frac{2\pi^2a^2\chi^2H_0^2}{\mu+1}\to\frac{6m^2}{(2a)^4} \tag{22}$$

is done. Introducing the phase lag $\beta=\varphi-\theta=\omega t-\theta$ Eq. (21) can be integrated as

$$\Lambda=\frac{6m^2}{(2a)^4}\frac{1}{2}(\cos 2\beta-1) \tag{23}$$

[$\beta(\pm 1)=0$ since the normal shearing force at the ends of the chain vanishes]. In a steady case when the rod rotates synchronously with the applied field

$$\frac{dv_n}{dl}=\omega.$$

This relation, accounting for the expression of the normal force Eq. (6), when the force due to the curvature elasticity is neglected, gives

$$\frac{d}{dl}\left(\Lambda\beta_l-\frac{6m^2}{(2a)^4}\frac{d}{dl}\sin 2\beta\right)=\zeta\omega. \tag{24}$$

Substituting the expression (23) into the relation (24) we obtain

$$\frac{d^2}{dl^2}\left(-\frac{6m^2}{(2a)^4}\frac{1}{2}\beta-\frac{3}{4}\frac{6m^2}{(2a)^4}\sin 2\beta\right)=\zeta\omega.$$

As a result the equation for the tangent angle reads

$$\frac{6m^2}{(2a)^4}\left(\tfrac{3}{2}\sin 2\beta+\beta\right)=\zeta\omega(L^2-l^2). \tag{25}$$

This is the relation obtained in a model of an inextensible magnetic rod when the curvature elasticity of the rod is neglected. Since the function $F(\beta)=\frac{3}{2}\sin 2\beta+\beta$ on the left-hand side of Eq. (25) is not monotonous there is the critical frequency at which the jump of tangent angle takes place. At this condition disintegration of chain occurs. The value of the critical frequency can be found from the maximal value of function $F(\beta)$ which takes place at $\beta_c = \pi/2 - \frac{1}{2}\arccos 1/3$. Accounting for relation $\zeta = 4\pi\eta/\ln L/a + c$ [6] and expressing the magnetic moment of particle as $m = M 4\pi/3a^3$, where $M = \chi_p H_0$ [15] we have for the critical frequency of the chain of $N = 2L/2a$ particles the relation

$$MaN^2 = 2\pi\left(\tfrac{3}{2}\sin 2\beta_c + \beta_c\right)\left(\ln\frac{L}{a} + c\right). \tag{26}$$

Here the Mason number $Ma = 12\eta\omega/M^2$ according to the notation of the paper [14] is introduced. Relation (23) shows that with the increase of the Mason number the number of the particles N which can be held together by attractive magnetic force diminishes as

$$N = \sqrt{\frac{const}{Ma}},$$

where *const* according to the relation (23) has order of magnitude 6. This is what is observed in experiments (see, for example, Fig. 3 in Ref. [14]) where diminishing of the number of aggregated magnetic particles starts at the Mason number value about 1. It should be noted that at critical value of the Mason number the condition for existence of the chain of the magnetic particles

$$\Lambda + F_r \leqslant 0 \tag{27}$$

is satisfied, where F_r is the magnetic interaction force

$$F_r = -\frac{\partial U}{\partial r} = \frac{3m^2}{(2a)^4}\left(1 - 3\cos^2(\varphi - \theta)\right)$$

$$= \frac{3m^2}{(2a)^4}\left(-\tfrac{1}{2} - \tfrac{3}{2}\cos 2\beta\right),$$

holding particles together. Indeed according to the relation (19) the left-hand side of the condition (27) is

$$\frac{3m^2}{(2a)^4}\left(-\frac{3}{2} - \frac{\cos 2\beta}{2}\right) < 0,$$

which justifies the applicability of the model of an inextensible magnetic rod for the description of the chain of magnetic particles in a rotating field. For the Mason number larger than critical the chain breaks. We believe that the full model for description of this phenomenon should account for the anisotropy of the bending elasticity of the chain of the magnetic particles which will be considered elsewhere [20]. It should be remarked that breaking of the chains, which are long enough, sustains some kind of the self-organized critical state [24] in which the chains are growing due to axial coalescence on one hand and are breaking if the size is larger than critical on the other hand. Since the characteristic size distribution of the chains arising due to the competition of these processes remains unknown at the present moment it seems impossible to make quantitative comparisons with existing experimental data on birefringence and dichroism of the magnetorheological suspensions in a rotating field [12,14].

Concerning the relation (22) it should also be remarked that, apart from term β on the left side, it is close to the relation obtained in Ref. [15] in the frame of the discrete model of the magnetic particle chain. This relation written in terms of the notations of Ref. [15] is ($Ma' = 3/32\pi Ma$)

$$\sin 2\beta = \frac{32}{9(\ln L/a + c)} Ma' N^2\left(1 - \left(\frac{l}{L}\right)^2\right),$$

where the right-hand side differs by multiplier $2/9(\ln L/a + c)$ from the relation given in Ref. [15]. It should be also remarked that since the function $F(\beta)$ on the left-hand side of the relation (22) is no monotonous the hysteretic phenomena in the range of the angles in the center of a chain [11°,54°] are possible. This behavior possibly has been remarked in the numerical simulations of the chain of the magnetic particles in Ref. [15] and the terms of brittle and ductile failure for two kinds of the chain breaking connected with this hysteretic behavior were introduced. Thus we believe that the model of an inextensible magnetic rod can also be applied to the description of the chains of unlinked magnetic particles held together by the magnetic attraction forces. This model, extended to account for different details, may play the same role in understanding of the behavior of the magnetorheological suspensions which Kirchhoff's model of the elastic rod plays in the study of DNA and other macromolecules [25].

V. CONCLUSIONS

We have shown that the flexible magnetic chains have quite a rich behavior under the action of the rotating magnetic field. At low frequency of the magnetic field the rod has a bent shape rotating synchronously with the applied field. The phase lag of the center of the rod increases with the frequency of a rotating field. At the critical frequency which—for the considered range of parameters—is proportional to the square of the field strength asynchronous regime of the rod motion arises. This may be described by a simple model neglecting tangential motion of the rod. The parameters of the considered model correspond to the real existing systems and the results of our simulations can be checked experimentally. It is illustrated in particular cases that the considered model reproduces the phenomena observed experimentally for the magnetic particle chains in magnetorheological suspensions. The model gives a general framework for the description of various phenomena in magnetorheological suspensions.

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