Dynamics of an elongated magnetic droplet in a rotating field

A. Cēbers*
Institute of Physics, University of Latvia, Salaspils-1, LV-2169, Latvia
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A model is proposed for the dynamics of an elongated droplet under the action of a low frequency rotating magnetic field. This model determines a set of critical frequencies at which the transitions to more complex bent shapes take place. These transitions occur through propagation of jumps of the droplet's axial tangent angle described by a nonlinear singularly perturbed partial differential equation with the intrinsic viscosity of the droplet playing the regularizing role.

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I. INTRODUCTION

The behavior of liquid magnetic droplets under a rotating magnetic field exhibits several fascinating features. One of them, regarding the reentrant transition oblate-prolate-oblate in which the droplet changes its shape upon the increase of magnetic field from oblate to prolate and then back to oblate, has been analyzed in a number of studies [1–4]. At the same time the very rich dynamics of the magnetic droplet in the intermediate range of magnetic field strength [5,6] still remains unexplored. The synchronous mode of magnetic droplet dynamics is typified by a "forklike" light diffraction pattern [5]. The formation of such diffraction patterns is attributed to elongated slightly bent droplets ("magnetic worms") synchronously rotating with the magnetic field. Formation of such shapes has also been observed by numerical simulation of the dynamics of two-dimensional (2D) magnetic fluid droplets under the action of a rotating magnetic field [7]. At higher rotation frequencies transitions occur to more complex, typically S and 8, shapes [5,6]. Transitions between those shapes are not yet understood. In order to elucidate these phenomena, a proposed model is based on the magnetic droplet dynamics as determined by the magnetic torques and capillary and viscous forces. Different regimes of droplet dynamics are found depending on the frequency of the rotating field. At small frequencies bent shapes with less than $\pi/4$ angle between the tangent and the field direction occur. At a critical frequency the S shape appears, which transforms to the 8 shape as the second critical frequency is reached. Transitions to a different family of shapes occur through formation of jumps of the tangent and their propagation. For a certain range of tangent angles the model is reduced to a backward parabolic equation, which is inherently unstable and requires regularization. The latter is provided by taking into account the torque stresses due to the intrinsic viscosity of the droplet. The derivation of the equation for droplet dynamics is carried out in terms of the slender body approach, similar to the Kirchhoff model of rods in the theory of elasticity [8]. Models of this type have been used for description of complex dynamics of various filamentary biological objects [9,10].

*Electronic address: aceb@tesla.sal.lv

II. THE MODEL

The equations for an elongated magnetic droplet under the action of capillary, magnetic, and viscous forces are derived in the framework of slender body theory. This approximation has been successfully applied to the bending of a viscous fluid jet under the action of gravity [11] and the electrospinning of a jet in an axial electric field [12]. According to this approach the cross section of the droplet is assumed to be circular and constant. Thus tangential forces, which can lead to a change of the droplet length, are neglected [12]. To justify this approach, we point out that the viscosity of the surrounding fluid, which controls the droplet's shape change, and that of the droplet fluid, controlling the rate of its length variation, differ considerably. For example, the viscosity of the concentrated phase of the magnetic liquid used in the experiments [5,6] that we refer to is more than two orders of magnitude higher than the viscosity of the surrounding liquid. The shape of the droplet is described by the position of its centerline. Let its tangent be $\vec{t}$, the normal $\vec{n}$, and the binormal $[\vec{t}\times\vec{n}]$. The relevant forces and torques are the following: the normal force F_n acting on the cross section of the droplet, arising due to the action of the magnetic torque; K_n, the surface tension force per unit length of the droplet; the torque of the viscous stresses M_b, arising upon the bending of the droplet; and the external torque due to the applied rotating field equal to T_0 per unit of the droplet's length. Magnetic interactions between the distant parts of the droplet are neglected. Only the motion in the plane of the field rotation is considered. The latter assumption is in accord with the experimental observations [1,2,4–6].

For the tangent and the normal vectors, Frenet equations are valid, $d\vec{t}/dl=(1/R)\vec{n}$, $d\vec{n}/dl=-(1/R)\vec{t}$, where R is the radius of curvature of the centerline and l its arc length. The balance of forces in the direction normal to the centerline gives

$$-\delta v_n+\frac{dF_n}{dl}+K_n=0, \tag{1}$$

where the coefficient δ in the friction force $-\delta v_n$ exerted by the surrounding fluid may be expressed, neglecting the hydrodynamic interactions, as [13,14]

$$\delta = \frac{4\pi\eta}{\ln(L/a)+c}.$$

Here $2L$ is the length of the droplet, a the radius of its cross section, c a constant of the order of unity, and η the viscosity of the surrounding fluid. Since the droplet is in rotational equilibrium at every instant, the torque balance yields

$$\frac{dM_b}{dl} + F_n + T_0 = 0. \tag{2}$$

The torque M_b due to the intrinsic viscosity of the droplet is calculated by referring to an analogy between the elastic and viscous phenomena [11] as

$$M_b = \frac{3\pi}{4} a^4 \eta_i \frac{\partial}{\partial t}\left(\frac{1}{R}\right). \tag{3}$$

Here η_i is the intrinsic viscosity of a droplet. The external torque T_0 acting per unit length of the droplet is given by the expression $T_0 = [\vec{M} \times \vec{H_0}]_b \pi a^2$. The magnetization and the external field vectors are noncollinear because the demagnetizing field coefficients along the axis of the locally cylindrical droplet and in the direction perpendicular to it are different. According to our model, the demagnetizing field coefficient $N_{||}$ for the magnetic field component along the centerline of the droplet is assumed to be equal to zero, whereas the one in the perpendicular direction $N_\perp = 2\pi$. The magnetization components then are as follows:

$$M_t = \chi H_{0t}, \; M_n = \frac{\chi H_{0n}}{1+2\pi\chi}.$$

As a result, the magnetic torque is

$$T_0 = \frac{4\pi\chi^2 H_{0n} H_{0t} \pi a^2}{\mu+1}. \tag{4}$$

Some comments concerning the last relation are appropriate. The magnetic torque on a volume element with ellipsoidal shape is generally determined by two contributions: the torque due to the local magnetic field $[\vec{M} \times \vec{H}]V$ and the torque due to the surface magnetic forces $2\pi(\vec{M}\vec{n})^2\vec{n}$. It is possible to show by a direct calculation that the sum of these torques is equal to $[\vec{M} \times \vec{H_0}]V$ [15]. Usually the volume magnetic torque $[\vec{M} \times \vec{H}]V$ can be neglected due to the small magnetic relaxation time. Whenever this is not the case, more complex phenomena with internal circulation in the droplet take place [15]. These phenomena are disregarded in the present model.

Another complication connected with the calculation of the magnetic torque on the droplet may be caused by the nonlinearity of the magnetization dependence on the internal magnetic field. In this case, it is possible to calculate the magnetic torque numerically from the expression for the magnetic energy of an ellipsoidal droplet with the main axis at the angle θ with respect to the external field [16]:

$$E = V\left(-\cos^2\theta \frac{H_0^2}{2}\frac{M}{H_i + N_{||}M} - \sin^2\theta \frac{H_0^2}{2}\frac{M}{H_i + N_\perp M} + \frac{1}{2} M H_i - \int_0^{H_i} M \, dH_i\right). \tag{5}$$

Here the internal magnetic field strength H_i in the droplet is to be found from the solution of the equation

$$\cos^2\theta \frac{H_0^2}{(H_i + N_{||}M)^2} + \sin^2\theta \frac{H_0^2}{(H_i + N_\perp M)^2} = 1. \tag{6}$$

For the magnetic liquid the Langevin law $M = M_S L(H/H_*)$ may be used for the magnetization where H_* is expressed through the magnetic moment of the colloidal particle m via the relation $H_* = k_B T/m$ and M_S is the saturation magnetization of the liquid. Since the magnetic field strength employed in the experiments with the droplets of the concentrated phase of a magnetic colloid is rather low [1,2,5,6], the aforementioned nonlinearity of the magnetization field dependence is not taken into account in the present model. For a linear magnetization law, the last two terms in the relation (5) cancel, which yields for the magnetic torque $K_0 = -\partial E/\partial\theta$ the expression

$$K_0 = V\frac{H_0^2}{2}\sin 2\theta\left(-\frac{\chi}{1+N_\perp\chi} + \frac{\chi}{1+N_{||}\chi}\right). \tag{7}$$

For a cylinder with demagnetizing field coefficients $N_{||} = 0$ and $N_\perp = 2\pi$ for the torque per unit length the latter relation yields the expression (4).

Introducing the angle θ between the local tangent to the centerline of the droplet and the abcissa axis $[\vec{t} = \partial\vec{r}/\partial l = (\cos\theta, \sin\theta)]$ and using the commutator

$$\frac{d}{dt}\frac{\partial}{\partial l} - \frac{\partial}{\partial l}\frac{d}{dt} = -\left(\frac{\partial v_t}{\partial l} - \frac{1}{R}v_n\right)\frac{\partial}{\partial l},$$

where the velocity of the points on the curve is $\vec{v} = v_n\vec{n} + v_t\vec{t}$, we derive the following expression for the time derivative of the tangent angle:

$$\frac{d\theta}{dt} = \frac{\partial v_n}{\partial l} + v_t\frac{1}{R}. \tag{8}$$

Since the total length of the droplet is assumed constant, the tangential velocity is zero. The normal force K_n due to the surface tension is deduced from the variation of the surface energy at constant volume of a droplet element of length dl. The conservation of volume implies

$$\frac{\delta a}{\delta dl} = -\frac{a}{2dl}.$$

Thus, the variation of surface energy for a displacement $\delta\vec{r}$ is

$$\delta E = \gamma\pi a \int \delta dl = -\gamma\pi a \int \frac{1}{R}(\delta\vec{r}\vec{n})dl.$$

As a result, the normal force K_n due to the surface tension reads

$$K_n = \frac{\pi \gamma a}{R}. \tag{9}$$

The relations (1)–(4) and (8), (9) yield, taking into account that $\partial \theta / \partial l = 1/R$, the following equation for the tangent angle:

$$\frac{\partial \theta}{\partial t} = \frac{\pi \gamma a}{\delta} \frac{\partial^2 \theta}{\partial l^2} - \frac{1}{\delta} \frac{\partial^2 T_0}{\partial l^2} - \frac{3 \pi a^4}{4} \frac{\eta_i}{\delta} \frac{\partial^5 \theta}{\partial l^4 \partial t}.$$

For a rotating field $H_0 = (\cos \omega t, \sin \omega t)$ the relation (4) yields

$$T_0 = \frac{4 \pi \chi^2 H_0^2 \cos(\omega t - \theta) \sin(\omega t - \theta) \pi a^2}{\mu + 1},$$

and, introducing the phase lag $\beta = \omega t - \theta$ between the local tangent direction and the magnetic field, the equation for the angle β assumes the form

$$\omega = \frac{\partial \beta}{\partial t} - \frac{\partial^2}{\partial l^2} \left(\frac{\pi \gamma a}{\delta} \beta + \frac{2 \pi^2 \chi^2 H_0^2 a^2}{\delta(\mu + 1)} \sin 2\beta \right) + \frac{3 \pi a^4}{4} \frac{\eta_i}{\delta} \frac{\partial^5 \beta}{\partial^4 l \partial t}. \tag{10}$$

In order to present Eq. (10) in dimensionless form, we choose the characteristic time $\tau = \delta L^2 / M$ (the time of orientation of a droplet of length $2L$ under the action of the torque M). In the present case this gives

$$\tau = \frac{(\mu + 1) \delta L^2}{2 \pi^2 \chi^2 H_0^2 a^2}.$$

As a result, Eq. (10) in the dimensionless form reads

$$\omega \tau = \frac{\partial \beta}{\partial t} - \frac{\partial^2}{\partial l^2} \left(\frac{1}{\mathrm{Bm}} \beta + \sin 2\beta \right) + \epsilon \frac{\partial^5 \beta}{\partial l^4 \partial t}. \tag{11}$$

Here

$$\epsilon = \frac{3\pi}{4} \left(\frac{a}{L} \right)^4 \frac{\eta_i}{\delta}$$

is a small parameter depending on the ratio of the cross section radius to the length of the droplet $2L$. Thus, for droplets of the concentrated phase of a magnetic colloid with $\eta_i / \eta = 3.10^2$ [5] and the axis ratio $a/L = 1/40$, ϵ is about 10^{-4}. Bm is the magnetic Bond number given by the ratio of the magnetic and capillary forces,

$$\mathrm{Bm} = \frac{2 \pi \chi^2 H_0^2 a}{(\mu + 1) \gamma}.$$

Equation (11) must be supplemented by boundary conditions. For a free magnetic droplet in the rotating field, those

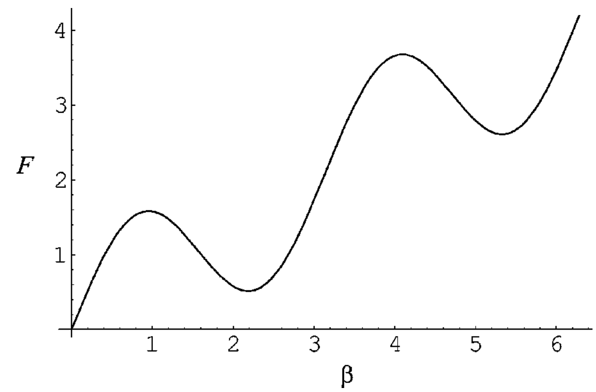

FIG. 1. Effective torque in dependence on tangent angle. Bm $= 1.5$.

are the conditions for the absence of normal forces and torques at the tips of a worm $F_n(\pm 1) = 0$; $M_b(\pm 1) = 0$. This, according to Eqs. (2) and (3), gives

$$\left. \frac{\partial^2 \beta}{\partial l \partial t} \right|_{l = \pm 1} = 0, \tag{12}$$

$$\left. \sin 2\beta - \epsilon \frac{\partial^3 \beta}{\partial l^2 \partial t} \right|_{l = \pm 1} = 0. \tag{13}$$

III. THE SHAPES OF THE DROPLETS IN A ROTATING FIELD: DISCUSSION OF THE EXPERIMENTAL RESULTS

Equation (11) with the boundary conditions (12), (13) determines the dynamics of the magnetic droplet under the action of a rotating field. Equation (11) without the last term can be written in the form

$$\omega \tau = \frac{\partial \beta}{\partial t} - \frac{\partial^2}{\partial l^2} F(\beta). \tag{14}$$

Without this last term of Eq. (11) the problem is ill posed. Indeed, the function $F(\beta) = (1/\mathrm{Bm})\beta + \sin 2\beta$ is not monotonic for magnetic Bond number values larger than $\frac{1}{2}$ (Fig. 1). This corresponds to the backward diffusion equation [17]. Numerical solution of the reduced equation (14) shows instabilities. The last term of Eq. (11), which describes the action of the viscous torques due to the intrinsic viscosity of the worm, plays a regularizing role. For steady state, Eq. (14) with boundary conditions $\beta|_{l = \pm 1} = 0$ possesses a simple solution

$$F(\beta) = \frac{1}{2} \omega \tau (1 - l^2). \tag{15}$$

The tangent angle β dependence on the arc length variable l of the curve is discontinuous for field rotation frequencies larger than the critical value. In fact there is a set of critical frequencies determined by the equations

$$(\omega \tau)_{c_n} = 2F(\beta_n),$$

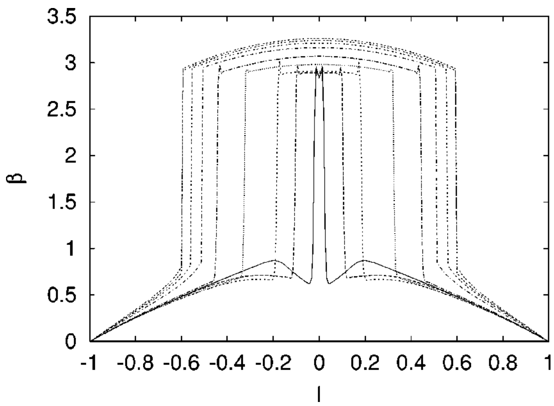

FIG. 2. Development of S-like shape at shock wave propagation. Bm=1.5; $\omega\tau=5.0$; $\epsilon=10^{-7}$. Dimensionless time starting from center 0.28, 0.42, 0.56, 0.84, 1.12, 1.39, 1.68, 2.08, 2.78.

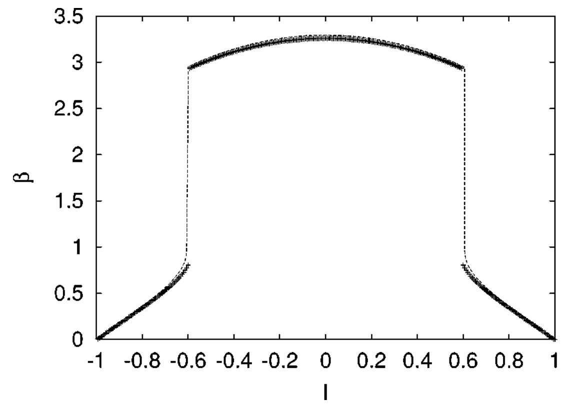

FIG. 3. Tangent angle of S-like shape. Numerical data, crosses; theoretical solution determined by Eq. (15) and β_1, dotted line. Bm=1.5, $\omega\tau=5.0$, and $\epsilon=10^{-7}$.

where $n=1,2,\ldots$ and β_n $[F'(\beta_n)=0]$ correspond to the local maxima of the function $F(\beta)$. The dependence of the critical frequency $(\omega\tau)_{c1}$ on the magnetic Bond number is determined by the relation

$$(\omega\tau)_{c1}=2\left\{\frac{1}{\mathrm{Bm}}\left[\frac{\pi}{2}-\frac{1}{2}\arccos\left(\frac{1}{2\,\mathrm{Bm}}\right)\right]+\sin\left(\arccos\frac{1}{2\,\mathrm{Bm}}\right)\right\}. \tag{16}$$

$(\omega\tau)_{c1}$ diminishes with increase of the magnetic Bond number, reaching the limiting value $(\omega\tau)_{c1}=2$ at large magnetic Bond numbers. At the critical value of the rotating field frequency a jump of the tangent angle appears near the droplet's center and propagates until a new steady shape of the droplet is established. The transition to a new shape obtained by a numerical solution of Eq. (11) with boundary conditions (12), (13) is shown in Fig. 2. The numerical solution is obtained by an implicit scheme with the spatial derivatives approximated by central differences. The nonlinear equations for the values of the tangent angle at each time step are solved by Newton iterations. To approximate the boundary conditions (12), (13), the Taylor expansion of the function $\beta(l)$ near the end points is used up to the third order terms, yielding the following expressions for the first and second order derivatives at the left end point:

$$\beta'(0)=\frac{1}{2h}[4\beta(1)-\beta(2)-3\beta(0)],$$

$$\beta''(0)=\frac{1}{h^2}[\beta(2)+\beta(0)-2\beta(1)].$$

Here h is the mesh size. Similar relations are valid at the right end point.

The values of the physical parameters for the process shown in Fig. 2 are as follows: Bm=1.5, $\omega\tau=5$, and $\epsilon=10^{-7}$. The number of mesh points is 300. The initial condition $\beta=0$ is used. The value of the critical frequency of the transition to the S shape for the given magnetic Bond number is about 3.16. Ultimately the stationary droplet configuration (15) is established [see Fig. 3 for a comparison of the theoretical steady state solution (15) with that found numerically by establishment from the initial state $\beta=0$]. It should be remarked that for a different initial condition another steady state configuration may develop. It is characterized by positions of the jumps closer to the ends of the droplet, which correspond to the local minima of the function $F(\beta)$. Experimentally such configurations could be realized by decreasing the field rotation frequency from a higher initial value. Such experiments are not currently available. The shapes of a droplet under the action of a rotating field may be classified by the number of jumps. The actual shape of a droplet corresponding to the solutions found for the tangent angle may be found by numerical integration of the equations

$$\frac{dx}{dl}=\cos\beta$$

and

$$\frac{dy}{dl}=-\sin\beta.$$

The shapes found for several values of the field rotation frequency are shown in Fig. 4. This figure illustrates how the shapes of the droplets observed in experiments [5,6] may be classified according to the number of jumps of the tangent angle. Thus, the transition from the bent shape to the S shape ($1\rightarrow2$ in Fig. 4) is characterized by two jumps of the tangent angle; the transition to the 8 shape ($3\rightarrow4$ in Fig. 4) taking place at higher rotation frequencies is characterized by four jumps of the tangent angle.

Currently, there are no quantitative experimental data concerning the critical frequencies of the rotating magnetic field at which the transition occurs to the S- or more complex 8-like shapes. Nevertheless, some comparison is possible using the available experimental data [5] for the dynamics of elongated magnetic droplets of the concentrated magnetic phase under the action of a low frequency rotating magnetic field. The droplets of the concentrated phase [5] have high

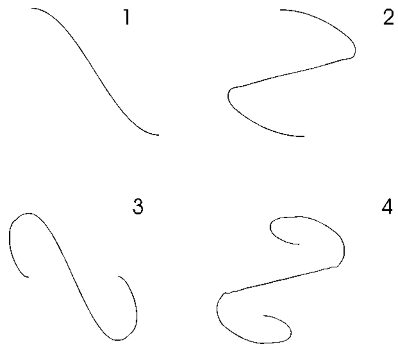

FIG. 4. Transition to S and 8 shapes. $\mathrm{Bm}=1.5, \epsilon=10^{-3}$, $\omega\tau=2.0$ (1), 3.5 (2), 6.0 (3), and 8.0 (4).

magnetic permeability with values up to about 86 and surface tension 1.1×10^{-3} erg/cm^2. The magnetic Bond number $\mathrm{Bm}=\frac{1}{2}(\mu-1)^2/(\mu+1)(a/L)^{1/3}H_0^2R_0/4\pi\sigma$ for such droplets in a magnetic field with $H_0^2R_0/4\pi\sigma=2.5$ [5] is equal to 30. Here the radius of the central part of the elongated droplet has been estimated from the radius of the droplet R_0 according to the relation $R_0=a(L/a)^{1/3}$. For such rather high values of the magnetic Bond number the critical frequency of the transition to the S shape is equal to its limiting value and thus may be estimated from the relation

$$\omega_c=\frac{2}{\tau}=\frac{4\pi^2\chi^2H_0^2a^2}{\delta L^2(\mu+1)}, \tag{17}$$

which for the above parameter values gives the critical frequency 25.5 Hz. This value is exactly in the range of frequencies of the rotating field ($\omega/2\pi<10$ Hz) where the transition to S-like shapes is observed [5]. It should be remarked, however, that in contrast to those considered in the present model, the droplets observed in [5] do not possess a steady state S shape rotating synchronously with the field. Rather, after the transition to the S shape they break up into three droplets. This is in accord with the S shape of the "mother" droplet and may be explained by the repulsion of its distant parts due to long-range magnetic interactions not taken into account in the present model. These effects will be taken into account by a more complex model, currently under development, which in addition to the long-range magnetic interactions will also consider the variation of the magnetic thread radius due to nonhomogeneity of the magnetic forces. Rather convincing support for the present model of the elongated droplet dynamics in a rotating magnetic field is also given by the experimental data of [18], which show that the stationary length of the chain of magnetic particles scales with frequency as $\omega^{-1/2}$. This corresponds to what is expected from relation (17) according to which jumps of the tangent are formed (they are actually observed in [18]) for chains with length larger than critical, and lead to its breakup. Thus, the observation of scaling is just what follows from relation (17) (apart from the weak logarithmic dependence of the friction coefficient δ on the droplet's length).

The jump dynamics is similar to that of a shock wave. Jump propagation provides the mechanism by which the transition to a new steady state synchronously rotating with the applied field is achieved. Formation of these jumps also bears a resemblance to the backward motions arising, for example, in a system of two magnetic holes [19] when the critical lag between the direction of the external field and the axis of the doublet is reached. Whenever the lag in the central region of the droplet reaches a critical value jumps of the tangent angle are formed.

IV. CONCLUSION

A simple model of the elongated magnetic liquid droplet rotating synchronously with an applied magnetic field has been proposed. This model allows us to identify the set of critical frequencies at which the droplet's shape transitions take place. The determined values of the critical frequencies coincide reasonably well with the available experimental data. For a numerical simulation of the transition to more complex droplet shapes occurring by propagation of a jump of the tangent angle, a regularization of the corresponding partial differential equation is necessary. This is achieved by taking into account the small effect of the intrinsic viscosity of the droplet. Further extension of this work will concern the study of possible nonsteady-state regimes in the system. It will also be interesting to take into account the extensional dynamics of the droplet due to the long-range magnetic interactions and the intrinsic stresses due to internal rotations in the magnetic fluid [20].

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