Toroid polarization of aggregated magnetic suspensions and composites and its use for information storage

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The importance of studying a new magnetic order parameter of aggregated suspensions—the toroid polarization—is demonstrated. The multipole parametrization is used to develop several methods of introducing the toroid dipole moment of an aggregate of magnetic particles. Methods for calculating the toroid dipoles of aggregates and the toroid polarization of suspensions as functions of the applied external fields and thermodynamic conditions are presented. This shows how one can control the toroid dipole of an individual aggregate of nanometer size. These theoretical developments open up prospects of new approaches to the recording and storage of information in magnetic media.

INTRODUCTION

Ferromagnetic suspensions (or, as they are also called, magnetic fluids)[1–3] are colloidal solutions of complicated composition including a carrier liquid, magnetic particles, and a surface-active substance (surfactant). The magnetic particles, almost spherical in shape, have sizes close to the single-domain limit (10–15 nm for most ferromagnets), and their magnetic moments are of the order of 10^4–10^5 Bohr magnetons. A typical magnetic fluid consists of magnetite particles suspended in kerosene with oleic acid as a surfactant. The surfactant covers the particles with a film of thickness about 2–4 nm, and this makes it possible to prevent coagulation completely or partly. Magnetic fluids can change their properties significantly, depending on the chosen surfactant, the thickness of the film that it forms on the particle surface, the size and shape of the particles themselves, and the magnitude and type of their magnetization. By choosing appropriate surfactants, one can in practice achieve rather high concentrations of magnetic particles (up to 10^{18} cm^{-3}). By virtue of their unusual magnetic properties ("fluid magnetic materials"), magnetic fluids have found wide application in technology (for more details, see Refs. 1 and 2).

At sufficiently low concentrations of the particles, many of the observed properties of magnetic suspensions can be explained by a model of noninteracting Brownian particles that possess fixed magnetic moments.[1–3] In this case, the dependence of the suspension magnetization **M** on the magnetic field **H** and on the temperature T can be expressed by Langevin's formula:

$$\mathbf{M}=\mu_0 N L(\xi)\frac{\mathbf{H}}{H};\quad L(\xi)=\coth\xi-\frac{1}{\xi}, \tag{1}$$

where N is the particle-number density, μ_0 is the magnetic moment of one particle, and the argument of the Langevin function $L(\xi)$ is a dimensionless parameter equal to the ratio of the magnetic and thermal energies of the particle: $\xi=\mu_0 H/kT$. However, with increasing concentration of the particles, the interaction between them becomes important, and one therefore observes a significant deviation of the behavior of the suspension from the model of noninteracting point Brownian dipoles. In particular, one observes a deviation from Langevin behavior of not only the magnetization but also the optical and rheological properties of the suspension (see, for example, Refs. 4–9). The most important effect that results from the interaction between the particles is the formation of aggregates of magnetic particles.[1–13] The theory of the formation of aggregates was originally developed by De Gennes and Pincus[14] and then by other authors (see the reviews of Refs. 10 and 11, and also Refs. 15–17). Aggregates of magnetic particles were directly observed in a recent study[17] using small-angle neutron scattering. These investigations show that at low particle concentrations, small aggregates consisting of 2–10 particles are formed. At higher concentrations, the formation of fractal clusters, network structures, "domains," layers of moving concentration, etc., becomes advantageous. In this study, we shall consider suspensions at fairly low concentrations, at which only small aggregates are formed, as is usually the case in stable suspensions. One can then describe an aggregated suspension by a model of noninteracting particles that have a complicated structure which changes under the influence of an external magnetic field and temperature. It should be noted that despite the significant number of publications devoted to aggregated suspensions most have been made without detailed allowance for the shape of the aggregates and the mutual orientation of the magnetic moments of the particles forming an aggregate. The main aim of this paper is to take into account systematically the spatial and magnetic configurations of the aggregates and their variation under the influence of an external magnetic field and temperature, and also to establish their macroscopic manifestations.

If it is assumed that the particles of a suspension before aggregation were magnetically homogeneous (in what follows, we shall for brevity refer to these particles as *embryons*; the concept of embryonic forms for atomic clusters was introduced in Ref. 18), then an aggregate of them is a

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new particle that is characterized by an inhomogeneous distribution of the magnetization over its volume. In particular, in the absence of a magnetic field it is energetically advantageous for the aggregates to form with closed magnetic flux and zero total magnetic moment (just as in a bulk ferromagnet a domain structure with closing domains is formed[19]). The absence of a total magnetic moment means that to describe the magnetic properties of an aggregate it is necessary to introduce multipole moments of higher order, including the quadrupole moments and also the toroid moments[20–22] and a magnetic "charge." It is important that the closed (vortex) structure of the distribution of embryon magnetic moments is most naturally described by the toroid moment. Thus, independent measurement of the toroid moment of a suspension, and also of its susceptibility to an inhomogeneous magnetic field (toroid susceptibility) together with measurements of the magnetization in a homogeneous magnetic field can give valuable information about the structure of a magnetic fluid and the aggregates that are forming (or exist) in it.

The review is arranged as follows. In Sec. 1, we consider the classification of aggregates of the magnetic particles of a suspension. We show that aggregates of two types can exist: magnetic aggregates, for which the shape-determining factor is the dipole–dipole interaction of the embryons, and nonmagnetic aggregates, which form by the coalescence of the surfactant envelopes of the embryons. These aggregates differ in shape and magnetic properties. In Sec. 2, we introduce the multipole characteristics of aggregates. We show that an aggregate as a system consisting of a finite number of dipoles has a finite number of magnetic degrees of freedom and therefore can be described by a finite number of independent multipole moments that describe its magnetic state. We introduce other "representations" of the magnetic configuration of aggregates and analyze in detail their significance and relationship to each other. In Sec. 3, we consider the interaction of aggregates with an inhomogeneous magnetic field and with one another. We show that the parameters of the effective field may differ from those of the original one. In particular, depending on the aggregate shape, the effective field may have a solenoidal configuration even though the original field does not. This makes it possible to measure the toroid polarization of a suspension in a quasistatic regime without using high-frequency sources of a solenoidal magnetic field (produced, for example, by displacement currents). In the same section, we analyze methods of forming an external field of a given configuration by means of a system of macroscopic dipoles. We consider the interaction of two "finite toroids" in different positions.

In Sec. 4, we describe algorithms for calculating the magnetic configuration of aggregates and their shape on the basis of the methods of micromagnetism and molecular dynamics. In the concluding, fifth section we give the results of calculations,[50] which are used to investigate the magnetization of aggregates by a solenoidal magnetic field. We note possibilities for controlling the toroidal properties of a magnetic medium in order to record and store information.

1. TYPES OF AGGREGATES OF MAGNETIC PARTICLES

Many publications (see the reviews and monographs of Refs. 1–2 and 10–13 and the references given there) have been devoted to the processes of aggregation of the particles of magnetic suspensions. Initially, aggregates were observed using the methods of electron microscopy on magnetic-fluid samples suspended on special substrates or microtomed polymerized fluids.[10] However, recently fluids have been investigated by small-angle neutron scattering,[17] which makes it possible to observe the aggregates *in situ*. Important evidence for the ideas presented below is provided by observations of fluids by the method of optical-mixing spectroscopy,[8] and also modeling of a suspension by means of suspended magnetic microparticles observed in an optical microscope.[23] Macroscopic manifestations of the formation of aggregates were observed in investigations of the susceptibility of suspensions,[4–5] optical effects,[4–9] and also in rheological experiments.[10–11,13] Theoretically, the formation of aggregates in weakly concentrated suspensions in the presence (and absence) of a magnetic field was considered by De Gennes and Pincus,[14] who obtained an estimate of the length of a chain as a function of the interaction parameter $\lambda=\mu^2/a^3kT$ of the particles (here, a is the particle diameter) and their volume concentration.

In Ref. 24, Jordan attempted to investigate more complicated structure of aggregates, regarded as polymer molecules. Pincus[25] considered qualitatively the magnetic configuration of aggregates as a function of their shape; he predicted that in the absence of a field there could be formation of annular structures with closed magnetic flux possessing zero total magnetic moment. Similar predictions were made by Scholten[26,27] on the basis of an analysis of the interaction between dispersed particles and also using the data of measurement of birefringence and dichroism of magnetic fluids. In addition, ensembles of interacting dipoles have been modeled numerically by the Monte Carlo method[16] and other methods,[15,28] which also indicate the formation of aggregates of magnetic particles of different types.

For a qualitative understanding of the aggregation process in a magnetic suspension, it is convenient to start with the dependence of the potential of the binary interaction of the particles of the suspension on the distance between them. This is shown schematically in Fig. 1a in accordance with the data of Refs. 12, 26, and 29. The potential curve has two minima separated by a potential barrier of height U_0. The height of the barrier is a measure of the repulsion of the particles of the suspension covered by the surfactant films (steric or entropy repulsion; note that here we do not consider the nature of this interaction). The right-hand minimum of the potential (A) corresponds to a magnetic dipole attraction between the particles that does not disturb the integrity of the surfactant films on the surface of the particles (Fig. 1b). In what follows, we shall call such aggregates formed as a result of magnetic attraction *magnetic* or *dipole* aggregates. It is the process of dipole aggregation of the particles of a magnetic suspension (the most

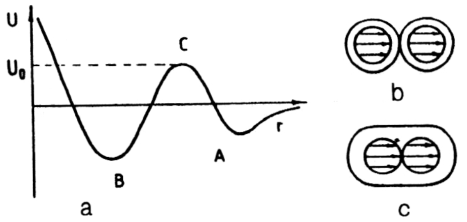


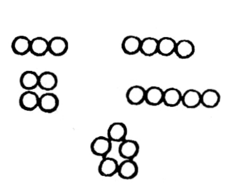
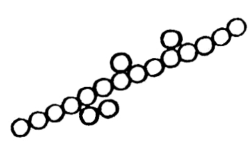
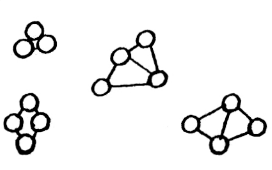
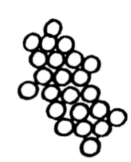
FIG. 1. Schematic representation of the potential of the two-body interaction between the particles of a magnetic suspension (a). The minimum A corresponds to magnetic binding of particles in an aggregate (b), and the minimum B corresponds to nonmagnetic binding (c).

characteristic for magnetic fluids) that has been investigated in most of the studies cited above. In stable suspensions, the depth of minimum A in the curve of the potential may be assumed to be small compared with the energy kT of the thermal motion. Therefore, the process of dipole aggregation occurs, if we use modern terminology, in accordance with the scenario of diffusion-limited aggregation.[28,30] In the absence of a magnetic field, the mean number of particles in an aggregate depends on their total concentration in the suspension: At a low concentration not exceeding a certain critical value, aggregates of a small number of particles (less than 10) are present in the suspension. This is indicated not only by direct observations[8,17] but also by numerical modeling.[15–17] Small-angle neutron scattering makes it possible to observe independently both the surfactant shell and the magnetic particles themselves. In the observations described in Ref. 17, the mean radius of one suspension particle was 6 nm, and the mean thickness of the surfactant film on a particle was 4 nm. At low concentration, it was mainly dimers with mean distance 21 nm between the centers of the particles that were observed in the fluid; this indicated a dipole origin of these aggregates. With increasing concentration of particles in the fluid, chain and three-dimensional aggregates consisting of a large number of particles were also observed.[17] When the critical concentration was reached, the formation of infinite fractal dipole aggregates becomes possible; as a rule, the fractals have a low fractal dimension D ($D \approx 2$ in the absence of a field or $D \approx 1$ in the presence of a field).[23,28,31] Dipole aggregates can change their shape in sufficiently strong magnetic fields, this being manifested in the so-called Curie–Weiss behavior of the suspension susceptibility and the birefringence.[4–7,9]

Besides dipole aggregates, which are characteristic of magnetic suspensions, there may also be ordinary (nonmagnetic) binding of particles into aggregates in a magnetic fluid. The second minimum in the potential curve of Fig. 1a (minimum B) corresponds to this possibility. In the case of nonmagnetic aggregation, the surfactant films coalesce, and the particles are held together by van der Waals forces or by the surface-tension forces of the film (Fig. 1c). Since the barrier U_0 is fairly high, aggregates form in this manner comparatively slowly; under the condition $U_0 \gg kT$, this process is called reaction-limited aggregation.[30] In the initial stage, it can be assumed that only small aggregates of such type are present in the fluid.[8] For very long times, this aggregation process leads ultimately to the formation of fractal aggregates with a relatively large fractal dimension $D \approx 2.5$, indicating practically complete irreversible coagulation of the suspension.[32]

Thus, the existing data indicate that in a magnetic suspension over short time intervals and for sufficiently low particle concentrations (less than the critical value; see above) there exist aggregates of small numbers (2–10) of particles of two kinds—dipole (magnetic) and nonmagnetic aggregates (see Table I). These aggregates differ both in shape and in their magnetic properties. Dipole aggregates may also change their shape under the influence of external factors, while nonmagnetic aggregates can be regarded to a high degree of accuracy as having unchanged shape (rigid). In the literature, there is no detailed description of small aggregates, i.e., their shapes, distributions of magnetic moments, and the particles (embryons) that

TABLE I. Types of aggregates in magnetic suspensions.

Type of binding of particles	Dimension (number of particles)	
	Finite number of particles (small aggregates)	Fractal aggregates containing many particles
Dipole (magnetic)		 $D \approx 1$
Nonmagnetic		 $D \approx 2,5$

form them and the related behavior of the magnetic suspension. The main aim of the present paper is to consider these questions.

As we noted in the Introduction, to describe stable magnetic suspensions the model of noninteracting Brownian magnetic dipoles is used in many cases. The use of this model is justified by the fact that despite the long-range nature of the magnetic dipole interaction ($U \sim 1/r^3$) it also depends strongly on the mutual orientation of the interacting dipoles. After averaging over the orientation of the Brownian particles, the effective interaction between them decreases with the distance already as $1/r^6$ (in accordance with van der Waals's classical result). Therefore, for a sufficient thickness of the surfactant film covering the particles, the dipoles can be assumed to interact weakly with each other. These considerations show that a single-particle model is a good first approximation for describing a magnetic fluid. In the framework of this model, many observed magnetic properties of suspensions can be explained. An important modification of the model is to take into account the Néel mechanism of relaxation of the magnetic moment of an individual particle with respect to its anisotropy[3,33,34] (model of "unfrozen" dipoles).

A further development of the single-particle model indicated by the experimental data (Refs. 4–9, 27, and 34) is the assumption that the magnetic suspension contains not only isolated particles but also small aggregates, which can significantly change its properties, as we have already noted. For the same reasons as in the case of isolated magnetic dipoles, one can ignore the interaction of the aggregates with one another, especially because on aggregation the total magnetic moment of the aggregate that is formed may be less than the sum of the moments of the individual embryons, as occurs, for example, in the formation of annular aggregates. The interaction between aggregates can become important only if there is a sufficiently high concentration of embryons, when an unrestricted growth of aggregates begins. Note also that the critical concentration at which this process begins depends on the magnetic field—in the presence of a field, the dipole aggregates are drawn out in linear chains and can form a spatial network. An alternative process is stratification of the fluid into weakly and strongly concentrated fractions or the formation of ordered structures (domains, etc.) (Refs. 1, 2, 35, and 36).

In accordance with these arguments, we can at the least assert that in the space with the coordinates N (the particle concentration), H (magnetic field), and t (time) it is possible to identify a region, not too far from the origin, in which it is valid to use the model of an aggregated suspension, which consists of small aggregates of dipole particles that interact weakly with one another (single-particle model of aggregates). The study of magnetic suspensions in the framework of this model is of independent interest, since this medium possesses new magnetic properties—besides a magnetic moment, its particles can possess multipole moments of higher rank (toroid, quadrupole, etc.), and the properties of the particles themselves depend on external factors. Such aspects of the behavior of the particles are manifested macroscopically as a non-Langevin behavior of the suspension. Besides its independent interest, study of this model of a suspension also has practical interest. If it is assumed that the solution of most applied problems requires magnetic suspensions with well-separated particles, then the formation of aggregates, i.e., the transition of the suspension to the aggregated state, is an undesirable process that must be controlled. Investigation of suspension behavior in the framework of the model will also help to solve this problem.

2. MAGNETIC PARAMETERS OF AGGREGATES

In the following exposition, we shall assume that the original particles of the suspension (the embryons) that form aggregates are spherical in shape and have "frozen" dipole moments ("rigid dipoles"). The condition of "freezing" is discussed in detail in Refs. 3 and 33–35. For given density of the anisotropy energy of the ferromagnet of which the particles consist, the condition is well satisfied for the particles when their diameter exceeds a certain critical value. In this paper, we shall consider only the equilibrium properties of suspensions, and, therefore, strictly speaking, the assumption of rigidity of the dipoles at this stage is redundant, since a rigid connection between a particle and its dipole moment is manifested only when one considers relaxation phenomena or other unsteady phenomena such as transient processes accompanying changes in the shape of aggregates. During the exposition, we shall see the extent to which this assumption is important in the consideration of the various problems.

In real suspensions, the particles have a fairly broad distribution of sizes. Since the shape of an aggregate and its magnetic properties depend on the relative size of the embryons, in a consistent treatment it would be necessary to investigate the properties of aggregates for arbitrary sizes of the particles forming them, and then average over the known (usually log-normal) distribution of the particles. Naturally, this would greatly complicate the problem. However, experimental data[17] obtained by observing particles through the scattering of slow neutrons show that even in a stable (standard) magnetic suspension up to 70% of the particles are in the aggregated state. At the same time, it is well known that the formation of aggregates of particles differing strongly in size is most probable, with very large particles serving as centers of attraction for smaller particles. Since the relative fraction of both the large and the small particles is small, and particles of average size form the main bulk, the fraction of aggregates formed from particles differing strongly in size is also relatively small. Therefore, in a first approximation one can ignore the contribution of such aggregates and assume that all the embryons in the aggregates have approximately the same (average) size. In other words, we here, first, ignore the correlation between the sizes of particles that form the aggregates, i.e., we assume that embryons of all sizes are found in aggregates with equal probability and independently of each other, and, second, we replace the volume V of a particle by its mean value $\bar{V}$. This replacement on averaging leads to an exact result if the averaged function

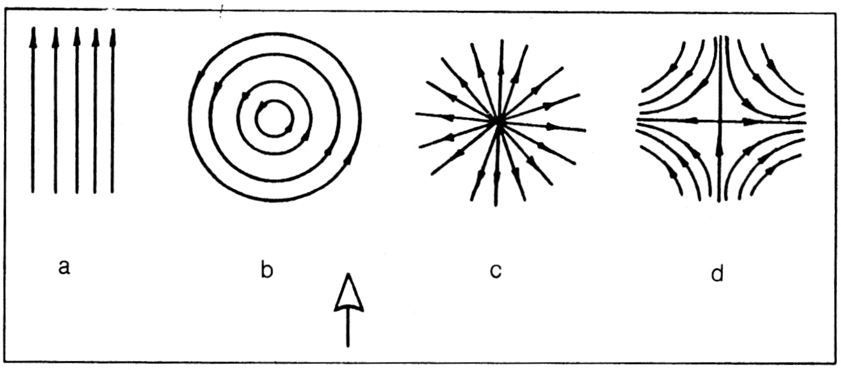


FIG. 2. Homogeneous and inhomogeneous distribution of dipole orientations in aggregates described by the following moments: a) magnetic; b) toroid; c) scalar; d) quadrupole.

$f(V)$ is linear in V, and in the remaining cases it is equivalent to the approximate equality $\overline{f(V)} \approx f(\overline{V})$.

2.1. Multipole moments of a medium of point dipoles

In what follows, we shall describe each aggregate by the coordinates $\mathbf{r}_a$ of its constituent embryons and indicate the orientation of their magnetic moments by $\mathbf{m}_a$, where the index a labels the embryons and takes values from 1 to n (n is the number of particles in the aggregate). In accordance with the assumption made above, the moments of all the embryons have the same magnitude: $|\mathbf{m}_a| = \mu_0$. If the rigid-dipole model is valid, each vector $\mathbf{m}_a$ can be expressed in the form $\mathbf{m}_a = \mu_0 \mathbf{e}_a$, where $\mathbf{e}_a$ is a unit vector rigidly attached to the particle itself. The set of coordinates $\mathbf{r}_a$ determines the shape of the aggregate (the spatial configuration), and the set of moments $\mathbf{m}_a$ determines its magnetic configuration. Algorithms for calculating these quantities for given H will be discussed below; here, we consider some integrated properties of the aggregates. In what follows, it is assumed that the origin is taken at the geometrical center of the aggregate, i.e.,

$$\sum_a \mathbf{r}_a = 0. \tag{2}$$

The most important quantity that describes the magnetic properties of the aggregate as a whole is its total magnetic moment

$$\boldsymbol{\mu} := \sum_a \mathbf{m}_a, \tag{3}$$

which determines the energy of the interaction of the aggregate with the homogeneous magnetic field $\mathbf{H}_0$:

$$E_H = -(\boldsymbol{\mu}\mathbf{H}_0). \tag{4}$$

In addition, we can introduce multipole moments of higher order that describe the spatially inhomogeneous distribution of the moments $\mathbf{m}_a$ and the interaction of the aggregates with the inhomogeneous fields. These include the quadrupole moment

$$\kappa_{ik} := \tfrac{1}{2} \sum_a [m_{ai} x_{ak} + m_{ak} x_{ai} - \tfrac{2}{3} \delta_{ik} (\mathbf{m}_a \mathbf{r}_a)], \tag{5}$$

the toroid moment[20–22]

$$\boldsymbol{\tau} := \tfrac{1}{2} \sum_a [\mathbf{r}_a \mathbf{m}_a], \tag{6}$$

the scalar moment

$$\sigma := \tfrac{1}{3} \sum_a (\mathbf{r}_a \mathbf{m}_a), \tag{7}$$

and also other moments containing higher powers of $\mathbf{r}_a$. The characteristic spatial distributions of the dipoles that are described by the moments $\boldsymbol{\mu}$, $\boldsymbol{\tau}$, σ, and κ_{ik} are shown in Fig. 2. A distribution of dipoles with closed magnetic flux (Fig. 2b) has a toroid moment, while the scalar σ characterizes fields that have "sources" or "sinks" (Fig. 2c).

In Refs. 20–22, toroid multipole moments were introduced into electrodynamics in connection with a system of currents. To establish the connection between the "current" moments and the "dipole" moments considered here, it is convenient to go over to a description of the magnetic configuration of the aggregate by means of a continuous magnetization distribution $\mathbf{m}(\mathbf{r})$. Then one can go over from the expressions (3) and (5)–(7) to expressions in which the substitution $\Sigma_a \to \int_V$, where V is the volume occupied by the aggregate, has been made:

$$\boldsymbol{\mu} := \int_V \mathbf{m}(\mathbf{r}) dV, \tag{3'}$$

$$\kappa_{ik} := \tfrac{1}{2} \int_V [m_i x_k + m_k x_i - \tfrac{2}{3} \delta_{ik} (\mathbf{mr})] dV, \tag{5'}$$

$$\boldsymbol{\tau} := \tfrac{1}{2} \int_V [\mathbf{rm}] dV, \tag{6'}$$

$$\sigma := \int_V (\mathbf{rm}) dV. \tag{7'}$$

It is important to note that for the decomposition of the electric-current density analyzed in detail in Ref. 22, the following conditions hold in the (quasi)static case:

$$\operatorname{div} \mathbf{j} = 0; \quad \int \mathbf{j} dV = 0. \tag{8}$$

The first of these conditions follows from the charge conservation law, and the second is the requirement that the total current flowing in the system be zero. By virtue of these conditions, the decomposition of the total current cannot contain moments analogous to the total dipole moment (3′) and the scalar σ (5′), which appear when the magnetization distribution $\mathbf{m}(\mathbf{r})$ is decomposed.

2.2. Dipole charges and currents

The scalars, vectors, and tensors that characterize the continuous vector field $\mathbf{m}(\mathbf{r})$ introduced above can also be obtained in a different manner, namely, as the mean values of the derivatives of the function $\mathbf{m}(\mathbf{r})$ with respect to the coordinates:

$$\rho_m := \operatorname{div} \mathbf{m}(\mathbf{r});$$
$$\mathbf{j}_m := c \operatorname{curl} \mathbf{m}(\mathbf{r}); \tag{9}$$
$$q_{ik} := \tfrac{1}{2}(\nabla_i m_k + \nabla_k m_i - \tfrac{2}{3}\delta_{ik} \operatorname{div} \mathbf{m}),$$

where c is the speed of light. The integrals of these quantities over the volume of the aggregate can be used for a generalized description of the magnetic configuration of aggregates together with the multipole moments σ, τ_i, κ_{ik} introduced above. Thus, we can introduce the following quantities:
the "magnetic charge"

$$e_m := \int \rho_m(\mathbf{r}) dV, \tag{10}$$

the total "dipole current"

$$\mathbf{i} := \int \mathbf{j}_m(\mathbf{r}) dV, \tag{11}$$

and the tensor of the magnetization gradient of the aggregate

$$p_{ik} := \int q_{ik}(\mathbf{r}) dV. \tag{12}$$

It should be emphasized especially that in the expressions (10)–(12) the integration is only over the volume of the ferromagnet, i.e., we exclude the transition region between the ferromagnetic medium and the vacuum, where the function $\mathbf{m}(\mathbf{r})$ has a discontinuity and its derivatives with respect to the coordinates are singular. In particular, the region of integration cannot be arbitrarily extended into the region where there is no ferromagnet and where $\mathbf{m}(\mathbf{r}) = 0$. Before we turn to a detailed discussion of the quantities that we have introduced, we establish connections between their "densities" (9) and the multipole moments σ, $\boldsymbol{\tau}$, k_{ik}. To clarify the relationship between the toroid moment $\boldsymbol{\tau}$ and the corresponding properties of space–time symmetry of the vector $\mathbf{j}_m$ of the dipole-current density, we transform the integrand in the expression (6′) identically:

$$\tfrac{1}{2}[\mathbf{r}\mathbf{m}(\mathbf{r})]_i = \tfrac{1}{10}[\mathbf{m}(\mathbf{r})\nabla]_k (x_i x_k - 2r^2 \delta_{ik}). \tag{13}$$

After this, we can integrate in the expression (6) by parts and represent the integrand in the form

$$\tau_i = \frac{1}{10c} \int (x_i x_k - 2r^2 \delta_{ik}) j_{mk} dV - \tfrac{1}{10} \oint [\mathbf{m}(\mathbf{r})\mathbf{n}]_k \times (x_i x_k - 2r^2 \delta_{ik}) dS, \tag{14}$$

where the first term is identical to the definition of the "current" toroid moment given in Refs. 20–22. Similarly, we can establish a relationship between the moments σ and κ_{ik}, on the one hand, and the densities ρ_m and q_{ik} on the other. Representing the integrands in (7′) in the form

$$(\mathbf{r}\mathbf{m}) = \tfrac{1}{2}(\mathbf{m}(\mathbf{r})\nabla) r^2,$$
$$x_i m_k + m_i x_k - \tfrac{2}{3}\delta_{ik} m_j x_j \tag{15}$$
$$= (\nabla_i m_k + m_i \nabla_k - \tfrac{2}{3}\delta_{ik} m_j \nabla_j) \tfrac{1}{2} r^2$$

and transforming the integrals in (7) by parts, we can find that the moments σ and κ_{ik} are quadratic "radii" (cf. Refs. 20–22) of the charge density ρ_m and the local gradient of the magnetization q_{ik}:

$$\sigma = \tfrac{1}{2} \int r^2 \rho_m dV + \dots; \quad \kappa_{ik} = \tfrac{1}{2} r^2 q_{ik} dV + \dots. \tag{16}$$

In these expressions, the surface integrals have been omitted for brevity. Comparing the two forms of integrated characteristics—the quantities e_m, $\mathbf{j}$, and p_{ik} given by (10)–(12) and the expressions for σ, $\boldsymbol{\tau}$, and κ_{ik} represented in the form (14), (16)—we can see that, for example, the dipole current $\mathbf{i}$ is the moment of zeroth order in powers of the radius vector of the dipole-current density $\mathbf{j}_m$, whereas the toroid moment $\boldsymbol{\tau}$ of the aggregate is the moment of second order of the same current, as follows from comparison of the expressions (10) and (14). Similarly, the mean values e_m and p_{ik} are the moments of zeroth order of the densities ρ_m and q_{ik}, and σ and κ_{ik} are the moments of second order. In this connection we note that of the two first-order moments of the current density $\mathbf{j}_m$—the scalar and vector, which have the form of the integrals

$$\int (\mathbf{r}\mathbf{j}_m) dV; \quad \int [\mathbf{r} \times \mathbf{j}_m] dV,$$

the scalar moment has zero volume density, as one can see by using the definition (9) of the current density $\mathbf{j}_m$, while the vector moment can be reduced to the magnetic moment $\boldsymbol{\mu}$ of the aggregate as a whole.

To obtain a deeper understanding of the connection between the toroid moment $\boldsymbol{\tau}$ of the aggregate and the dipole current $\mathbf{i}$, we use Helmholtz's theorem. We introduce the vector and scalar "potentials" of the field $\mathbf{m}(\mathbf{r})$:

$$\mathbf{m}(\mathbf{r}) = -\nabla \varphi_m + \operatorname{curl} \mathbf{a}_m. \tag{17}$$

On the one hand, using the Neumann–Debye theorem,[22] we can readily prove that the "vector potential" $\mathbf{a}_m$ of the field $\mathbf{m}(\mathbf{r})$ is the volume density of the toroid moment: $\boldsymbol{\tau} \sim \int \mathbf{a}_m dV$. On the other hand, it follows from Helmholtz's theorem (see, for example, Ref. 37) that there exists a connection of the following form between this potential $\mathbf{a}_m$ and the curl of the field $\mathbf{m}$ [in accordance with (9), the curl of the field $\mathbf{m}$ is $\mathbf{j}_m/c$]:

$$\mathbf{a}_m(\mathbf{r}) = \frac{1}{c} \int \frac{\mathbf{j}_m(\mathbf{r})}{4\pi |\mathbf{r} - \mathbf{r}'|} dV'. \tag{18}$$

Thus, the current density $\mathbf{j}_m$ is a "source" for the field $\mathbf{a}_m(\mathbf{r})$, i.e., a toroid moment of the aggregate exists only when the dipole-current density $\mathbf{j}_m$ exists. At the same time, the total current $\mathbf{i}$ can be zero, just as the opposite is possible, i.e., the toroid moment $\boldsymbol{\tau}$ can be zero and the current $\mathbf{j}_m$ nonzero. Using the definition (7) of the scalar moment σ and the representation of the magnetization distribution in the form (17), we can show that the scalar potential φ_m is the density of the scalar σ, i.e., $\sigma \sim \int \varphi_m dV$.

On the other hand, in accordance with Helmholtz's theorem, the potential φ_m is related to the divergence of the field $\mathbf{m}$, this divergence being in accordance with the definition (9) equal to the "charge" density ρ_m:

$$\varphi_m(\mathbf{r}) = \int \frac{\rho_m(\mathbf{r}')}{4\pi|\mathbf{r}-\mathbf{r}'|}\, dV. \tag{19}$$

For what follows, it is very important to answer this question: Can the dipole current $\mathbf{i}$ defined by the expression (11) exist as an integral of the current density $\mathbf{j}_m$? In accordance with (8), the analogous integral of the "charge" density of the current is zero. Although the transversality condition $\operatorname{div}\mathbf{j}=0$ is obviously also satisfied for the dipole current, $\operatorname{div}\mathbf{j}_m=0$, this does not yet mean that the total current $\mathbf{i}$ is also zero; in general, it does not even mean that the lines of the current $\mathbf{j}_m$ are closed (see the interesting discussion of this question in Ref. 40 in connection with the force lines of the magnetic field, which, as is well known, also satisfies the equation $\operatorname{div}\mathbf{H}=0$). Taking into account the connection $\mathbf{j}_m = c\operatorname{curl}\mathbf{m}$, we can transform the expression for the current (11) into the surface integral

$$\mathbf{i} = c\int [\mathbf{n}\times\mathbf{m}]\, dS. \tag{20}$$

However, as we have already noted, in this integral we cannot transfer the surface of integration to a region in which $\mathbf{m}(\mathbf{r})=0$; for in accordance with the definition, the integration in Eqs. (10)–(12) is over the volume occupied by the ferromagnet. For aggregates of magnetic particles, the introduction of the function $\mathbf{m}(\mathbf{r})$, which is continuous over the entire volume occupied by such a composite particle, is an approximate operation, and therefore one can speak of a dipole current only approximately. However, the basic possibility of describing the magnetization distribution by means of a total dipole current $\mathbf{i}$ can be considered for the example of relatively large particles of a ferromagnet with diameters greater than the single-domain radius.

The experimental data show[41] that fine iron particles preserve the single-domain state up to diameters of order 40 nm, after which they go over to a state with single-vortex magnetization and preserve it in the range of diameters from 40 to 150 nm. At larger dimensions, the single-vortex state is replaced by a multivortex state, and, finally, at diameters greater than 1000 nm a multidomain state arises. Similar results have been obtained by the methods of micromagnetics,[42–45] which have shown that for definite sizes of the particles of a ferromagnet the distribution of the magnetization in such particles can be characterized as on the average a solenoidal distribution, i.e., as definitely having a mean value of the curl of the vector field $\mathbf{m}(\mathbf{r})$ over the volume that does not vanish; this is equivalent to the presence of the dipole current $\mathbf{i}$ introduced here. It must be emphasized that a mean "molecular current," which leads to the very existence of magnetic moments, is of course absent, i.e., it satisfies, as it must, the relations (8). Note that in large magnetic particles one also encounters a magnetization distribution described on the average by a magnetic charge e_m and a magnetization gradient p_{ik}. In the literature, such distributions are called "flower configurations."

2.3. Multipole approximation of the magnetization of an individual aggregate

We introduced the magnetization of an aggregate initially as a collection of point dipoles $\{\mathbf{m}_a\}$ distributed discretely over space. The most economic and numerically most readily realized method of going over from the collection of $\{\mathbf{m}_a\}$ values to a continuous differentiable distribution $\mathbf{m}(\mathbf{r})$ consists, as is well known, of using spectral methods.[38] In the framework of this approach, we shall assume that the dipole moments $\mathbf{m}_a$ of embryons specified at the points $\mathbf{r}_a$ are the values of a function $\mathbf{m}(\mathbf{r})$, i.e., $\mathbf{m}(\mathbf{r}_a)=\mathbf{m}_a$. We shall seek the unknown function $\mathbf{m}(\mathbf{r})$ as an expansion with respect to a finite set of given functions $\Phi_\alpha(\mathbf{r})$ ($\alpha=0,1,2,\ldots,K$):

$$\mathbf{m}(\mathbf{r}) = \sum_\alpha \mathbf{b}_\alpha \Phi_\alpha(\mathbf{r}). \tag{21}$$

In what follows, it will be assumed that the number $(K+1)$ of these functions is smaller than the number n of embryons, $K+1\leqslant n$, and, in addition, that the basis functions are chosen in such a way that the system of n-dimensional vectors $\{|\Phi_\alpha\rangle\}$ with components $\Phi_\alpha(\mathbf{r}_a)$ ($a=1,2,\ldots,n$) is linearly independent.

To calculate the expansion coefficients $\mathbf{b}_\alpha$, it is necessary to find the values of the right- and left-hand sides of Eq. (21) at the points $\mathbf{r}_a$, multiply the equation $\mathbf{m}_a=\Sigma_\alpha \mathbf{b}_\alpha\Phi_\alpha(\mathbf{r}_a)$ then obtained by $\Phi_\beta(\mathbf{r}_a)$, and then sum all these relations over a. We shall then obtain the system of linear equations

$$\sum_\beta L_{\alpha\beta}\mathbf{b}_\beta = \mathbf{N}_\alpha,$$
$$L_{\alpha\beta} = \sum_a \Phi_\alpha(\mathbf{r}_a)\Phi_\beta(\mathbf{r}_a), \tag{22}$$
$$\mathbf{N}_\alpha = \sum_a \mathbf{m}_a\Phi_\alpha(\mathbf{r}_a).$$

This system has unique solutions, since by virtue of the linear independence of the basis functions on the set of points $\mathbf{r}_a$, the determinant of the matrix $L_{\alpha\beta}$ is nonzero. If we redefine the basis functions in such a way that the n-dimensional vectors $|\Phi_\alpha\rangle$ are mutually orthogonal and normalized, $\langle\Phi_\alpha|\Phi_\beta\rangle=\delta_{\alpha\beta}$, then $L_{\alpha\beta}$ will be the unit matrix, and the system (22) will have the simple solution $\mathbf{b}_\alpha=\mathbf{N}_\alpha$.

Knowing the expansion coefficients $\mathbf{b}_\alpha$, and using the relations (21), (9)–(12), we can calculate the integrated characteristics of the aggregates that we introduced above—the magnetic charge, the dipole current, and the magnetization gradient:

$$e_m=\sum_\alpha (\mathbf{b}_\alpha \boldsymbol{\lambda}_\alpha), \quad \mathbf{i}=\sum_\alpha [\boldsymbol{\lambda}_\alpha \mathbf{b}_\alpha],$$
$$p_{ik}=\tfrac{1}{2}\sum_\alpha (b_{\alpha i}\lambda_{\alpha k}+b_{\alpha k}\lambda_{\alpha i}-\tfrac{2}{3}b_{\alpha j}\lambda_{\alpha j}\delta_{ik}), \tag{23}$$

where we have written

$$\boldsymbol{\lambda}_\alpha=\sum_a [\nabla\Phi_\alpha(\mathbf{r})]_{\mathbf{r}=\mathbf{r}_a}. \tag{24}$$

As an example, we consider the simplest and most natural choice of the basis functions, assuming them to be $\Phi_0(\mathbf{r})=1$, $\Phi_i(\mathbf{r})=x_i$ $(i=1,2,3)$, where x_i are the components of the radius vector $\mathbf{r}$. Note that if the dipoles are in a single plane (two-dimensional aggregate), then instead of the four functions 1, x, y, z we must restrict ourselves to the three functions 1, x, y, where it is assumed that the particles lie in the (x,y) plane. If we do not, the condition of linear independence of the vectors $|\Phi_\alpha\rangle$ will be violated. In the considered case, the expansion (21) can be expressed in the form

$$m_i(\mathbf{r})=b_{0i}+b_{ik}x_k. \tag{25}$$

In the system of equations (22), which determines the coefficients $b_{\alpha i}$, the matrix $L_{\alpha\beta}$ has, with allowance for the condition (2) of the choice of the origin, the form

$$L_{00}=n; \quad L_{0i}=L_{i0}=0, \quad L_{ik}=\sum_a x_{ai}x_{ak}, \tag{26}$$

and the inhomogeneities $\mathbf{N}_\alpha$ can be uniquely expressed in terms of the multipole moments σ, $\boldsymbol{\tau}$, κ_{ik}. Indeed, as follows from (22) and the definition $\Phi_0(\mathbf{r})=1$, the vector $\mathbf{N}_0$ is equal to the magnetic moment of the aggregate:

$$\mathbf{N}_0=\sum_a \mathbf{m}_a=\boldsymbol{\mu}. \tag{27}$$

With regard to the vectors $\mathbf{N}_\alpha$, $\alpha=1, 2, 3$, these three vectors form a second-rank tensor with components

$$N_{ik}=\sum_a x_{ai}m_{ak}. \tag{28}$$

Decomposing this tensor into irreducible parts,

$$N_{ik}=N^{(0)}\delta_{ik}+e_{ikl}N_l^{(1)}+N_{ik}^{(2)}, \tag{29}$$

where we have introduced the scalar $N^{(0)}$, the vector $N^{(1)}$, and the irreducible second-rank tensor $N^{(2)}$, which are related to the components of the original tensor by

$$N^{(0)}=\tfrac{1}{3}N_{jj}; \quad N_i^{(1)}=\tfrac{1}{2}e_{ikl}N_{kl}$$
$$N_{ik}^{(2)}=\tfrac{1}{2}(N_{ik}+N_{ki}-\tfrac{2}{3}N_{jj}\delta_{ik}), \tag{30}$$

we can readily show that these quantities are identical to the multipole moments σ, $\boldsymbol{\tau}$, κ_{ik} defined by Eqs. (5)–(7):

$$\sigma=N^{(0)}; \quad \boldsymbol{\tau}=\mathbf{N}^{(1)}; \quad \kappa_{ik}=N_{ik}^{(2)}. \tag{31}$$

In the given case, the system of equation for the unknown coefficients $\mathbf{b}_\alpha$ decomposes into two subsystems:

$$nb_{0i}=\mu_i, \quad L_{ik}b_{kj}=N_{ij},$$

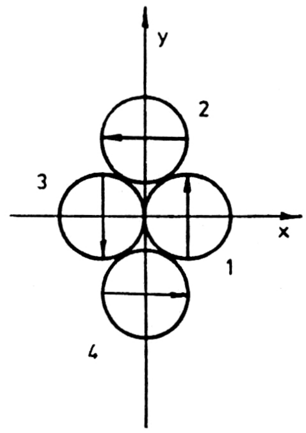


FIG. 3. Aggregate in the form of a rhombus. The arrows show the orientation of the magnetic moments of the particles in the ground state.

from which we find the solution

$$b_{0i}=\frac{\mu_i}{n}, \quad b_{ik}=L_{ij}^{-1}N_{jk}, \tag{32}$$

where the matrix L_{ij}^{-1} is the inverse of L_{ij} (the nondegeneracy of this matrix is due to the choice of the basis). Knowing the coefficients $\mathbf{b}_\alpha$ in (25), we can find all the dipole charges and currents in which we are interested. In accordance with the definition (24), the vector $\boldsymbol{\lambda}_\alpha$ for $\alpha=0$ vanishes, and the remaining vectors for $\alpha=1, 2, 3$ form, up to a factor, the unit second-rank tensor: $\lambda_{ik}\sim\delta_{ik}$. For this reason, as can be seen from the relation (23), the dipole charges and currents are equal to the irreducible parts of the tensor b_{ik}:

$$e_m=b^{(0)}, \quad \mathbf{i}=\mathbf{b}^{(1)}, \quad p_{ik}=b_{ik}^{(2)}, \tag{33}$$

where the tensor b_{ik} is decomposed into irreducible parts in the same way as the tensor N_{ik}, for which this is done by means of the relations (29) and (30).

In the general case, the basis functions $\Phi_\alpha(\mathbf{r})$ of the expansion (21) can be chosen as part of a complete set of functions. For example, the functions 1, x_i considered here are the first four functions of the complete set formed from all possible powers of the components of the radius vector: 1, x_i, x_ix_k, $x_ix_kx_j$,.... It is more convenient to express these functions in terms of the spherical harmonics $Y_{lm}(\theta,\varphi)$, where θ and φ are the spherical angles of the radius vector. As was shown in Refs. 22 and 39, in the long-wavelength approximation functions of the form $r^sY_{lm}(\theta,\varphi)$ can be chosen.

As a simple illustration, we consider a two-dimensional aggregate consisting of four particles in the shape of a rhombus with diagonals of length $2l_1$ (along the X axis) and $2l_2$ (along the Y axis) and orientation of the magnetic moments of the embryons as shown in Fig. 3 (the ground state, in which all the moments are perpendicular to each other and to the diagonals of the rhombus and lie in the plane of the aggregate). Simple calculations (see below) show that in this case the magnetic moment $\boldsymbol{\mu}$ of the aggregate, and also the scalar moment σ are zero, while the toroid and quadrupole moments are

$$\boldsymbol{\tau}=\mu_0(l_1+l_2)\mathbf{k};\quad \kappa_{12}=\kappa_{21}=\mu_0(l_1-l_2), \tag{34}$$

where $\mathbf{k}$ is the unit vector along the z axis perpendicular to the plane of the aggregate; the remaining components of the quadrupole moment, not given here, are zero. Using the solution (32) and the expressions (33), we can show that in the given case there is no dipole charge e_m, the dipole current is

$$\mathbf{i}=\frac{4\mu_0}{c}\left(\frac{1}{l_1}+\frac{1}{l_2}\right)\mathbf{k}, \tag{34$'$}$$

and the tensor of the magnetization gradient has only the two components

$$p_{12}=p_{21}=4\mu_0\left(\frac{1}{l_1}-\frac{1}{l_2}\right).$$

Note that for an aggregate in the shape of a square ($l_1=l_2$) our expressions show that only the toroid moment in (34) and the dipole current (34′) are nonzero, while $\kappa_{ik}=0$ and $p_{ik}=0$.

2.4. Parameters of the magnetic state of an aggregate as a finite discrete system

We consider a somewhat different interpretation of the multipole moments (and also of the dipole charges and currents) not associated with any approximate approach. Namely, we shall show that they can be regarded as new (and exact) parameters that specify the magnetic state of the aggregate. Indeed, the magnetic configuration of n dipoles is specified by $3n$ linearly independent variables—the vectors of the magnetic moments $\{\mathbf{m}_a\}$ of the embryons. We introduce for these variables the generalized notation M_J, where the index J takes values from 1 to $3n$. It is obvious that in place of the coordinates M_J we can use any linear combinations of them:

$$X_K=\sum_J U_{KJ}M_J, \tag{35}$$

where U_{KJ} is a nondegenerate matrix. The actual choice of the new variables X_K is dictated by considerations of convenience in solving particular problems.

We show that the two sets of characteristics—the multipole moments $\boldsymbol{\mu}$, $\boldsymbol{\tau}$, σ,... and the dipole charges and currents [more precisely, the values of their densities taken at the origin: $\rho_m(0)$, $\mathbf{j}_m(0)$,...]—can be regarded as two different sets of variables that describe the magnetic state of the aggregate and are linearly related to the old variables $\{\mathbf{m}_a\}$. We consider first the system of multipole moments. By definition, they are all possible irreducible tensors that can be separated from the Cartesian tensors of rank $S+1$ having the form

$$T_{i_1i_2\ldots i_sj}=\sum_a x_{ai_1}x_{ai_2}\ldots x_{ai_s}m_{aj}. \tag{36}$$

This relation can be regarded as a linear transformation from the old variables $\{\mathbf{m}_a\}$ to the new variables $T_{i_1i_2\ldots i_sj}$. Since the aggregate consists of a finite number of particles, it can be described by only a finite number of independent coordinates, and therefore we can choose from the set of all possible components of the tensors $T_{i_1i_2\ldots i_sj}$ a set that contains $3n$ quantities and use it to describe the states of the aggregate. In what follows, we shall denote these quantities by the symbols T_K, where $K=1, 2,\ldots,3n$. Between the variables of state T_K and M_J there exists in accordance with (36) a linear relationship of the type (35).

To obtain more compact and transparent relations, we go over from Cartesian to spherical coordinates. To this end, we introduce in place of the products $x_{ai_1}x_{ai_2}\ldots x_{ai_s}$ under the summation sign in Eq. (36) the spherical harmonics $r_a^sY_{lm}(\theta_a,\varphi_a)$, where the index l for given s takes values equal to s, $s-2$,... to 1 for odd s and 0 for even s. It is readily verified that the number of independent components of the symmetric tensor $x_{ai_1}x_{ai_2}\ldots x_{ai_s}$, which is equal to $(s+1)(s+2)/2$, and the number of functions $r_a^sY_{lm}(\theta_a,\varphi_a)$ with the given l, s, and m are exactly equal. Making this substitution, we can then form the irreducible L-rank tensors $T_{LM}^{(sl)}$ with given s and l, using the well-known rules for transforming tensors in spherical coordinates. Thus, we obtain

$$T_{LM}^{(sl)}=\sum_a\sum_m(\mathbf{m}_a\mathbf{C}_{LMlm})r_a^sY_{lm}(\theta_a,\varphi_a), \tag{37}$$

where the symbol $\mathbf{C}_{LMlm}$ denotes the Clebsch–Gordan coefficient C_{LMlm}^{1q}, which for convenience we have translated into Cartesian form with respect to the upper (vector) index. In accordance with the well-known "triangle" rule satisfied by the indices of a Clebsch–Gordan coefficient, the index L can take the values l, $l\pm1$ if $l\neq0$ and $L=1$ if $l=0$. It follows from this definition, in particular, that for $s=0$ or $s=1$ the index l takes the values $l=0$ or $l=1$, respectively. For $l=0$, the rank L of the tensor $T_{LM}^{(sl)}$ is equal to unity, while for $l=1$ we have $L=0$, 1, 2. Thus, for $s=0$, 1 there exist four irreducible tensors $T_{LM}^{(sl)}$: $T_{1M}^{(00)}$, $T_{00}^{(11)}$, $T_{1M}^{(11)}$, $T_{2M}^{(11)}$, the Cartesian analogs of which are, respectively, $\boldsymbol{\mu}$, σ, $\boldsymbol{\tau}$, κ_{ik}. For $s=2$, the index l can have two values: $l=0$ and $l=2$. If $l=0$, then, as before, $L=1$, while if $l=2$, then $L=0$, 1, 2, 3. Thus, in this case there exist the following tensors $T_{LM}^{(sl)}$: $T_{1M}^{(20)}$, $T_{1M}^{(22)}$, $T_{2M}^{(22)}$, $T_{3M}^{(22)}$. At the same time, the second-rank tensor $T_{2M}^{(22)}$ is the moment of the toroid series. In Cartesian form, it is a sum of symmetrized direct products of the vectors $\mathbf{r}_a$ and $[\mathbf{r}_a\mathbf{m}_a]$. Note that in the general case the multipoles of the toroid series for given s have ranks L equal to s, $s-2$... .

From all possible tensors $T_{LM}^{(sl)}$ we choose $3n$ and introduce for them the notation T_K given above. In accordance with (37), the relation between T_K and M_J can be written in the form

$$T_K=\sum_J U_{KJ}M_J, \tag{38}$$

where the matrix U_{KJ} can be expressed in terms of the coordinates of the dipoles by means of the function $r_a^s Y_{lm}(\theta_a,\varphi_a)$. The remaining multipole moments that have not been included in the set of variables T_K can be expressed in terms of T_K. Indeed, since by hypothesis the matrix U_{KJ} is nondegenerate, we can use its inverse U_{KJ}^{-1} to express M_J in terms of T_K:

$$M_J=\sum_K U_{JK}^{-1}T_K,$$

and then, substituting M_J in (37) in place of $\mathbf{m}_a$, we obtain the required relation.

Thus, we have shown that the multipole moments T_K can be regarded as independent parameters that describe the magnetic configuration of aggregates. As an example, for an aggregate consisting of four particles ($3n=12$) we can take the T_K to be the 12 variables $T_{1M}^{(00)}$, $T_{00}^{(11)}$, $T_{1M}^{(11)}$, and $T_{2M}^{(11)}$ (in counting the number of variables, one must bear in mind that for given L the index M takes $2L+1$ values, which are equal to $0, \pm 1,..., \pm L$). For an aggregate of five particles ($3n=15$), we can add to these variables, for example, the three components of the tensor $T_{1M}^{(20)}$, and for six particles ($3n=18$) we can also include $T_{1M}^{(22)}$. A simple count shows that the complete set of $T_{LM}^{(sl)}$ with values of s equal to 0, 1, 2 and, at the same time, with all possible values of the indices l, L, and M (as these tensors were listed above) constitutes just 30 variables, which are sufficient to describe the states of an aggregate of ten particles. It should be noted that if all dipoles lie on a straight line or in one plane (in what follows, we shall call these aggregates linear and planar, respectively), then their coordinates $\mathbf{r}_a$ are related by the linear dependence of belonging to a line or plane. For this reason, some components of the tensors $T_{LM}^{(sl)}$ are linearly dependent on each other, and therefore one must take care when interpreting them as independent coordinates.

We now show how the dipole charges and currents can be considered from such a point of view, as the introduction of new variables for the description of magnetic states. We shall proceed from an expansion analogous to (21), in which as basis functions $\Phi_s(\mathbf{r})$ we chose the tensors 1, x_{ai_1}, $x_{ai_1}x_{ai_2}$,... . We substitute in place of the current coordinate $\mathbf{r}$ the dipole coordinates $\mathbf{r}_a$ and obtain as a result the relation

$$m_{aj}=Q_j+Q_{ji_1}x_{ai_1}+Q_{ji_1i_2}x_{ai_1}x_{ai_2}+\dots . \quad (39)$$

We have here denoted the expansion coefficients by the symbols $Q_{ji_1\dots i_s}$ instead of those used in (21), since, in contrast to the previously described approximate method of their calculation, they will be found here exactly. To this end, we shall interpret Eq. (39) as a linear relationship between the old and new variables $\mathbf{m}_a$ and $Q_{i_1\dots j}$, respectively. If, as in (37), we go over from Cartesian to spherical coordinates, then Eq. (39) can be expressed in the more convenient form

$$\mathbf{m}_a=\sum_{slLM} Q_{LM}^{(sl)}\mathbf{C}_{LMlm}r_a^s Y_{lm}(\theta_a,\varphi_a), \quad (40)$$

where the indices s, l, L, and M are connected by the same relations as in (37). Choosing among the tensors $Q_{LM}^{(sl)}$ precisely $3n$ independent variables (in what follows, we shall denote these variables by the generalized symbol Q_K) and setting the remaining $Q_{LM}^{(sl)}$ equal to zero, we can regard Eq. (39) as a linear transformation from the variables M_J to the variables Q_K. In generalized notation, the expression (40) can be rewritten in the form

$$M_J=\sum_K V_{JK}^{-1}Q_K. \quad (41)$$

The exact form of the transformation matrix V_{JK}^{-1} in this expression can be readily established by comparing Eqs. (41) and (40) for definitely chosen values of the indices s, l and L, M. Since by hypothesis the matrix Q_J is nondegenerate, the transformation (41) can be inverted:

$$Q_K=\sum_J V_{JK}M_J. \quad (42)$$

Thus, we have shown here that the coefficients Q_J of the finite sum (39) or (40) can be used to describe the magnetic configuration of the aggregate along with other variables. On the other hand, as will be shown below, the Q_J are related in a definite manner to the dipole charges and currents, and, therefore, the use of the latter can be interpreted as a transition to new variables.

Note that the expressions obtained here not only enable us to find the new variables (T_j and Q_j) in terms of the old variables M_j but also enable us to relate T_j and Q_j to each other; this is of interest in view of the connection considered above between the multipole moments, on the one hand, and the dipole charges and currents, on the other. Assuming that in Eq. (40) on the right-hand side we retain only the variables that are chosen to describe the state, and substituting (40) in (37), we obtain the required relation in the form

$$T_{LM}^{(sl)}=\sum_{L'M'}\sum_{s'l'} U_{LML'M'}^{(sls'l')}Q_{L'M'}^{(s'l')}, \quad (43)$$

where we have written

$$U_{LML'M'}^{(sls'l')}=\sum_a (\mathbf{Y}_{LM}^{(sl)}(\mathbf{r}_a)\mathbf{Y}_{L'M'}^{(s'l')}(\mathbf{r}_a)).$$

For brevity, we have here introduced vector spherical harmonics

$$\mathbf{Y}_{LM}^{(sl)}(\mathbf{r}_a)=\sum_m \mathbf{C}_{LMlm}Y_{lm}(\theta_a,\varphi_a)r_a^s.$$

As can be seen from (43), the transformation matrix U that relates the moments T_{LM} and the coefficients Q_{LM} depends on the spatial disposition of the dipoles, i.e., on the geometrical shape of the aggregate. We note in this connection that neither the matrix U_{JK} in (38) nor V_{JK} in (42) is unitary. The fact is that the choice of the basis functions in the form $r_a^s Y_{lm}(\theta_a,\varphi_a)$ is "tied" to the definition of the multipole moments, but, in general, these functions are not adequate to describe the geometrical shape of the aggregate. For this reason, the moments T_{LM} and the coefficients Q_{LM} are not related, and the matrix U in (43) is in the general case nondiagonal and depends on the coordinates. If in place of the functions $r_a^s Y_{lm}(\theta_a,\varphi_a)$ we were to use basis functions $\Phi_s(\mathbf{r})$ that were orthogonal and normalized, as was described above after Eq. (22), then the corresponding moments and coefficients would be equal, but these new variables would no longer have the significance of multipole moments and dipole charges and currents.

We now return to the relation (39) and establish the (approximate) significance of the tensor coefficients $Q_{ji_1i_2...i_s}$ and, at the same time, of the irreducible tensors $Q_{LM}^{(sl)}$ contained in these tensors. To this end, we shall again consider the sum (39) as an approximate series expansion in powers of the radius vector $\mathbf{r}$ of some function $\mathbf{m}(\mathbf{r})$ that has the values $\mathbf{m}_a$ at the points $\mathbf{r}=\mathbf{r}_a$. Then the expansion coefficients in the sum (39) are the derivatives of this function taken at the point $\mathbf{r}=0$ [we recall that the origin is taken at the geometric center of the aggregate in accordance with the condition (2)]:

$$Q_{ji_1i_2...i_s}=\frac{1}{s!}\left[\frac{\partial}{\partial x_{i_1}}\frac{\partial}{\partial x_{i_2}}\cdots\frac{\partial}{\partial x_{i_s}}m_j(\mathbf{r})\right]_{\mathbf{r}=0}. \qquad (44)$$

Comparing this relation with Eq. (9), we can see that for $s=1$ the irreducible parts of the second-rank tensor Q_{ji_1} are equal to the values of the charge density ρ_m, the current density $\mathbf{j}_m$, and the magnetization gradient q_{ik} taken at the point $\mathbf{r}=0$:

$$Q_{ji_1}=q_{ji_1}(0)+\frac{1}{2c}e_{ji_1k}j_{mk}(0)+\frac{1}{3}\delta_{ji_1}\rho_m(0).$$

It follows from (44), as one can show, that the tensors $Q_{ji_1i_2...i_s}$ can be regarded as multipole moments in the Fourier space. Indeed, if we replace the function $\mathbf{m}(\mathbf{r})$ by its Fourier transform $\mathbf{m}(\mathbf{k})$, related to the original function by

$$\mathbf{m}(\mathbf{r})=\int e^{i\mathbf{kr}}\mathbf{m}(\mathbf{k})d\mathbf{k}, \qquad (45)$$

substitute this expression in (44), differentiate under the integral sign with respect to $\mathbf{r}$, and then set $\mathbf{r}=0$, then we obtain in place of (44) the integral

$$Q_{ji_1i_2...i_s}=\frac{(i)^s}{s!}\int k_{i_1}k_{i_2}\ldots k_{i_s}m_j(\mathbf{k})d\mathbf{k}. \qquad (46)$$

Thus, the tensors $Q_{ji_1i_2...i_s}$ play in the k space the same role as the tensors $T_{i_1i_2...i_sj}$ in ordinary space. To demonstrate this analogy more fully, we write the relation (36) in the integral form

$$T_{i_1i_2...i_sj}=\int x_{i_1}x_{i_2}\ldots x_{i_s}\mathbf{m}_j(\mathbf{r})dV, \qquad (47)$$

so that the analogy (46) becomes obvious. In addition, it can be shown that for the tensors $T_{i_1i_2...i_sj}$ in k space there exists a relation analogous to (44) for $Q_{ji_1i_2...i_s}$. Indeed, using the inverse Fourier transformation

$$\mathbf{m}(\mathbf{k})=\frac{1}{(2\pi)^3}\int e^{-i\mathbf{kr}}\mathbf{m}(\mathbf{r})dV \qquad (48)$$

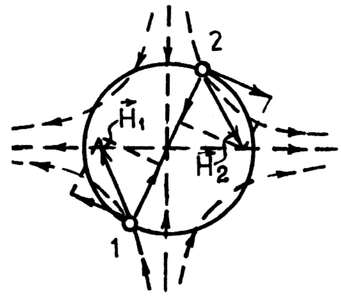


FIG. 4. The two-particle aggregate 1–2 is in an inhomogeneous magnetic field $\mathbf{H}(\mathbf{r})$ (potential and solenoidal); the force lines of the field are shown by the dashes. The vectors $\mathbf{H}_1$ and $\mathbf{H}_2$ show the values of the field $\mathbf{H}(\mathbf{r})$ at the positions of the particles (acting fields). The circle shows a force line of the acting field $\mathbf{H}'(\mathbf{r})$.

and differentiating the right- and left-hand sides of this expression with respect to $\mathbf{k}$, and then setting $\mathbf{k}=0$, we obtain on the right the integral (47), from which it follows that there is a relation of the following form between the derivatives of the Fourier transform $\mathbf{m}(k)$ and the multipole moments $T_{i_1i_2...i_sj}$:

$$T_{i_1i_2...i_sj}=\frac{(2\pi)^3}{(-i)^s}\left[\frac{\partial}{\partial k_{i_1}}\frac{\partial}{\partial k_{i_2}}\cdots\frac{\partial}{\partial k_{i_s}}m_j(\mathbf{k})\right]_{\mathbf{k}=0}. \qquad (49)$$

Thus, these quantities play the role of the "coefficients of an expansion" with respect to $\mathbf{k}$ in a relation analogous to (39), but the expansion is made, not in powers of $\mathbf{r}$, but in powers of $\mathbf{k}$. In this sense, the multipole moments correspond to the "long-wavelength" approximation in the expansion of the magnetization field $\mathbf{m}(\mathbf{r})$, while the coefficients $Q_{ji_1i_2...i_s}$, in contrast, correspond to a "short-wavelength" approximation of the same field (cf. Ref. 22). If by the symbol l_0 we denote the order of magnitude of the linear dimension of an aggregate, then $T_{i_1i_2...i_sj}$ can be estimated as $l_0^s\mu_0$ and $Q_{ji_1i_2...i_s}$ as μ_0/l_0^s, and this confirms once more the significance of these quantities that we noted above.

2.5. Example: rhombic aggregate

As a simple example, we again consider a planar aggregate consisting of four particles and having the shape of a rhombus with diagonals of length $2l_1$ (along the x axis) and $2l_2$ (Fig. 3). The original magnetic configuration of the aggregate is assumed to be known, i.e., we assume that we know the orientation of the particle magnetic moments $\mathbf{m}_a$ ($a=1,...,4$), which constitute 12 variables. As an example of a configuration, we show the vortex orientation of the dipoles (Fig. 4), which corresponds to the ground state of the aggregate, i.e., to the global minimum of the energy of the dipole interaction in the absence of a magnetic field. To go over to a description of the states by means of the multipole moments, it must be borne in mind that the radius vectors $\mathbf{r}_a$ of the particles are linearly independent. Therefore, although the moments of zeroth and first order in $\mathbf{r}$ do contain 12 variables, some of them are linearly dependent, and it is therefore necessary to use moments of second order (but not higher, since we restrict ourselves to the moments of lowest order).

Thus, in what follows we shall consider the tensors T_j, $T_{i_1 j}$, $T_{i_1 i_2 j}$, which are defined by the general relation (36). To simplify the notation, we shall use Greek indices to denote the Cartesian components of tensors in the plane of the aggregate. For the choice of the axes shown in Fig. 3, only some of the components of the tensors $T_{i...j}$, which can be expressed in the adopted notation as T_j, $T_{\alpha j}$, and $T_{\alpha\beta j}$, will be nonzero. In what follows, it will also be convenient to use for these quantities a succinct vector form of expression with respect to the index j, i.e., to denote them as $\mathbf{T}$, $\mathbf{T}_\alpha$, and $\mathbf{T}_{\alpha\beta}$, respectively. Using the expressions (36), we readily find that in the considered example they have the values

$$\mathbf{T}=\boldsymbol{\mu}=\mathbf{m}_1+\mathbf{m}_2+\mathbf{m}_3+\mathbf{m}_4,$$
$$\mathbf{T}_1=l_1(\mathbf{m}_1-\mathbf{m}_3),\quad \mathbf{T}_2=l_2(\mathbf{m}_2-\mathbf{m}_4), \tag{50}$$
$$\mathbf{T}_{11}=l_1^2(\mathbf{m}_1+\mathbf{m}_3),\quad \mathbf{T}_{22}=l_2^2(\mathbf{m}_2+\mathbf{m}_4),\quad \mathbf{T}_{12}=\mathbf{T}_{21}=0.$$

One can show that these vectors satisfy a linear dependence of the form

$$l_2^2\mathbf{T}_{11}+l_1^2\mathbf{T}_{22}-l_1^2l_2^2\mathbf{T}=0.$$

In the given case, the multipole moments σ, $\boldsymbol{\tau}$, and $\boldsymbol{\kappa}$, which are the irreducible parts of the tensor $T_{i_1 j}$, are linearly dependent, since only six of the nine components of this tensor are nonzero (and independent). To obtain a set of independent multipoles, we form from the components of the tensor $T_{\alpha j}$ a two-dimensional tensor (abbreviated: 2-tensor) of second rank $T_{\alpha\beta}$ and the 2-vector $T_{\alpha z}$. From the 2-tensor $T_{\alpha\beta}$ we can separate two 2-scalars:

$$\sigma=\tfrac{1}{2}(T_{11}+T_{22}),\quad \tau_z=\tfrac{1}{2}(T_{12}+T_{21}). \tag{51}$$

Bearing in mind also that the x and y components of the toroid moment are proportional to the components of the 2-vector $T_{\alpha z}$, as independent moments we can choose the following six quantities: 1) the scalar σ; 2) the toroid moment $\boldsymbol{\tau}$ of the aggregate with components $(\tfrac{1}{2}T_{23}, -\tfrac{1}{2}T_{13}, \tfrac{1}{2}(T_{12}-T_{21}))$; 3) the irreducible (i.e., symmetric and traceless) second-rank 2-tensor $\kappa_{\alpha\beta}$ ("two-dimensional" quadrupole moment), which has components satisfying

$$\kappa_{11}=-\kappa_{22}=\tfrac{1}{2}(T_{11}-T_{22}),\quad \kappa_{12}=\kappa_{21}=\tfrac{1}{2}(T_{12}+T_{21}).$$

In the second order in $\mathbf{r}$, only three of the 27 components of the tensor $T_{i_1 i_2 j}$ are independent. In fact, the three components have dipole radius

$$\boldsymbol{\mu}_2=\sum_a r_a^2\mathbf{m}_a. \tag{52}$$

However, it is easy to see that when the rhombus degenerates into a square the moment $\boldsymbol{\mu}_2$ is proportional to the total dipole moment $\boldsymbol{\mu}$ of the aggregate, and, thus, this quantity ceases to be an independent variable. The other possible vector containing expressions of the form $\mathbf{r}_a(\mathbf{r}_a\mathbf{m}_a)$ under the sign of the sum over a has only two components instead of the necessary three and therefore also cannot satisfy us. All the other moments $T_{LM}^{(sl)}$ with $s=2$ can be analyzed similarly. It can be shown that the necessary conditions are satisfied only by the second-rank tensor $T_{2M}^{(22)}$ (toroid quadrupole), which in the given case has three independent components. In Cartesian form, this tensor can be represented as the sum

$$\gamma_{ik}=\tfrac{1}{2}\sum_a\{x_{ai}[\mathbf{r}_a\mathbf{m}_a]_k\}, \tag{53}$$

where the braces denote symmetrization with respect to the indices i and k: $\{a_{ik}\}=\tfrac{1}{2}(a_{ik}+a_{ki})$. It is easy to show that for a rhombic aggregate all three diagonal components of this tensor vanish, while the nondiagonal components can be expressed in terms of $T_{\alpha\beta j}$ as follows:

$$\gamma_{12}=\gamma_{21}=\tfrac{1}{2}(T_{223}-T_{113}),$$
$$\gamma_{13}=\gamma_{31}=\tfrac{1}{4}T_{112},\quad \gamma_{23}=\gamma_{32}=-\tfrac{1}{4}T_{221}. \tag{54}$$

Thus, the magnetic state of the rhombic aggregate is described by the following independent multipole moments (multipole coordinates of the state): $\boldsymbol{\mu}$, σ, $\boldsymbol{\tau}$, $\boldsymbol{\kappa}_{\alpha\beta}$, γ_{ik}.

As an example, we calculate the multipole coordinates for two states—the ground state (vortex magnetization) and a uniformly magnetized aggregate (which arises in the presence of a very strong field). In the ground state (Fig. 3), the magnetic moments have the components $\mathbf{m}_1=-\mathbf{m}_3=(0;\mu_0;0)$, $\mathbf{m}_2=-\mathbf{m}_4=(-\mu_0;0;0)$. As can be seen from (50), in this case the vectors $\mathbf{T}$ and $\mathbf{T}_{\alpha\beta}$ vanish, while the vectors $\mathbf{T}_\alpha$ have the components $\mathbf{T}_1=(0;2l_1\mu_0;0)$, $\mathbf{T}_2=(-2l_2\mu_0;0;0)$. Therefore, in accordance with the expressions given above, the multipole coordinates of this state have the values $\boldsymbol{\mu}=0$, $\sigma=0$, $\tau=\mu_0(l_1+l_2)$; $\boldsymbol{\kappa}_{11}=\boldsymbol{\kappa}_{22}=0$; $\boldsymbol{\kappa}_{12}=\mu_0(l_1-l_2)$, $\gamma_{ik}=0$. Note that for a symmetric aggregate, when $l_1=l_2$, the quadrupole moment vanishes, and the vector $\boldsymbol{\tau}$ becomes the only quantity that describes this state.

In a strong magnetic field oriented along a certain unit vector $\mathbf{h}$, the magnetic moments of the particles are directed along the field and can be written in the form $\mathbf{m}_a=\mu_0\mathbf{h}$ (state of homogeneous magnetization). In this state, the vectors $\mathbf{T}$, $\mathbf{T}_\alpha$, and $\mathbf{T}_{\alpha\beta}$ have in accordance with (50) the values $\mathbf{T}=4\mu_0\mathbf{h}$, $\mathbf{T}_\alpha=0$, $\mathbf{T}_{11}=2l_1^2\mu_0 h$, $\mathbf{T}_{22}=2l_2^2\mathbf{h}$, at the same time $\boldsymbol{\mu}=4\mu_0\mathbf{h}$, $\sigma=0$, $\boldsymbol{\tau}=0$, $\kappa_{\alpha\beta}=0$, and the tensor of the toroid quadrupole moment has in accordance with (54) the components

$$\gamma_{12}=(l_2^2-l_1^2)\mu_0h_z,\quad \gamma_{13}=\tfrac{1}{2}l_1^2\mu_0h_y,\quad \gamma_{23}=-\tfrac{1}{2}l_2^2\mu_0h_x.$$

To describe the aggregate in terms of "current" magnetic coordinates given by the tensors $Q_{ji_1...i_s}$, we proceed from Eq. (39), which relates the magnetic moments $\mathbf{m}_a$ and the tensors $Q_{ji_1...i_s}$. As in the case of the multipole description, we shall use Greek indices to denote the coordinates of the particles in the plane of the aggregate. We shall assume that the independent variables are the vector Q_j (three variables), the tensor Q_{ji_1}, which has only six independent components $Q_{j\alpha}$ or, in "vector" form $\mathbf{Q}_\alpha$ (cf.

the notation used above: $T_{i_1 j} \to T_{\alpha j} \to \mathbf{T}_\alpha$), and, finally, the irreducible second-rank tensor f_{ik}, which is contained in the third-rank tensor $Q_{ji_1i_2}$ as follows:

$$Q_{jik}=\tfrac{1}{4}(e_{jil}f_{kl}+e_{jkl}f_{il}). \qquad (55)$$

At the same time, it is assumed that the diagonal elements of the tensor f_{ik} are zero, i.e., it has only three independent components (note that the relation between the components of the tensors $Q_{ji_1i_2}$ and f_{ik} is completely analogous to the relation between the tensors $T_{i_1i_2j}$ and γ_{ik} considered above). Thus, we have determined 12 new variables that specify the magnetic state of the aggregate.

The relationship between the new and old variables can be expressed in the form

$$\mathbf{m}_a=\mathbf{Q}+\mathbf{Q}_\beta x_{a\beta}+\tfrac{1}{2}[\mathbf{r}_a\mathbf{f}_\beta]x_{a\beta}. \qquad (56)$$

The inverse relations—between the new and the old variables—can be found by substituting in (56) the explicit values of the coordinates of the particles of the aggregate and solving the resulting system of equations for $\mathbf{Q}$, $\mathbf{Q}_\beta$, and $\mathbf{f}_\beta$. As a result, we obtain

$$\mathbf{Q}=\hat{a}(\mathbf{m}_1+\mathbf{m}_3)+\hat{b}(\mathbf{m}_2+\mathbf{m}_4),$$
$$\mathbf{Q}_1=\frac{1}{2l_1}(\mathbf{m}_1-\mathbf{m}_3), \quad \mathbf{Q}_2=\frac{1}{2l_2}(\mathbf{m}_2-\mathbf{m}_4), \qquad (57)$$
$$f_{ik}=C_{ik}(m_{1j}+m_{3j}-m_{2j}-m_{4j}), \quad i\neq k,$$

where $\hat{a}$ and $\hat{b}$ are diagonal matrices that have the form $\operatorname{diag}\hat{a}=(\tfrac{1}{2};0;l_2^2/2(l_1^2+l_2^2))$ and $\operatorname{diag}\hat{b}=(0;\tfrac{1}{2};l_1^2/2(l_1^2+l_2^2))$. In the last of the equations in (57) the index j on the right-hand side is complementary to the indices i and k, i.e., if, for example, $i=1$, $k=2$, then $j=3$, etc.; the coefficients C_{ik} have the values $C_{12}=-1/(l_1^2+l_2^2)$, $C_{13}=1/l_1^2$, $C_{23}=1/l_2^2$. By means of Eqs. (57), we can readily find the values of the magnetic charge and current, which are irreducible parts (scalar and vector) of the tensor $Q_{j\alpha}$:

$$e_m=\tfrac{1}{3}(Q_{11}+Q_{22}),$$
$$\mathbf{i}=\frac{1}{2c}\left[-Q_{32};\tfrac{1}{2}Q_{31};\tfrac{1}{2}(Q_{12}-Q_{21})\right]. \qquad (58)$$

As an example, we give the values of these parameters for the ground state of the aggregate (vortex distribution of magnetic moments): $\mathbf{Q}_1=\mathbf{m}_1/l_1$; $\mathbf{Q}_2=\mathbf{m}_2/l_2$; $\mathbf{Q}=0$; $\mathbf{f}_\alpha=0$, and for the state of uniform magnetization: $\mathbf{Q}=\mu_0\mathbf{h}$; $\mathbf{Q}_1=\mathbf{Q}_2=0$; $\mathbf{f}_\alpha=0$.

Finally, we consider approximate values of the dipole charge and current, which can be calculated by means of the procedure described in Sec. 3.3. Restricting ourselves to the simplest approximation, we represent the magnetization distribution in the form (25); moreover, by virtue of the linear dependence of the coordinates of the particles of a planar aggregate, we retain only the two components x and y of the radius vector and obtain as a result

$$\mathbf{m}(\mathbf{r})=\mathbf{b}_0+\mathbf{b}_\alpha x_\alpha \qquad (59)$$

with the same meaning of the index α as above in this section. To determine the coefficients $\mathbf{b}_0$ and $\mathbf{b}_\alpha$ as described in Sec. 3.3, we multiply Eq. (59) from the right and left by x_β, then set $\mathbf{r}=\mathbf{r}_a$, and sum over a. The tensor $N_{\alpha j}$ that we obtain in accordance with (38) on the left-hand side is identical to the tensor $T_{\alpha j}$ given above in Eq. (51). The matrix L_{ik} is transformed in this case into the second-rank matrix $L_{\alpha\beta}$ and in the chosen coordinate system (Fig. 3) is diagonal, as a result of which the general solution (32) can be written in the form

$$\mathbf{b}_0=\boldsymbol{\mu}/4; \quad \mathbf{b}_\alpha=L^{-1}_{\alpha\beta}\mathbf{T}_\beta=\mathbf{Q}_\alpha, \qquad (60)$$

where we have used the fact that $L^{-1}_{11}=1/2l_1^2$, $L^{-1}_{22}=1/2l_2^2$, $L^{-1}_{12}=L^{-1}_{21}=0$, and also the explicit forms (51) and (56) of the tensors T_α and Q_α. The values of the dipole charge and current in the given case are equal to the exact expressions (58) given above; for the ground state, they are given in Sec. 2.3.

3. INTERACTION OF AGGREGATES WITH A MAGNETIC FIELD

The parameters that we introduced in the previous section to describe the magnetic state of aggregates consisting of magnetic dipoles can be used to describe their interaction with an external magnetic field and with each other. As before, we shall assume that the magnetic and spatial configurations of the aggregate are given, i.e., we assume that we know the position $\mathbf{r}_a$ and orientation of the magnetic moment $\mathbf{m}_a$ of each of the dipoles. The interaction energy of a system of dipoles with an inhomogeneous external field $\mathbf{H}(\mathbf{r})$ can be written in the form

$$\mathscr{E}=-\sum_a(\mathbf{m}_a\mathbf{H}_a), \qquad (61)$$

where $\mathbf{H}_a$ denotes the field $\mathbf{H}(\mathbf{r})$ at the point $\mathbf{r}_a$, i.e., $\mathbf{H}_a=\mathbf{H}(\mathbf{r}_a)$. In what follows, we shall call the set of vectors $\{\mathbf{H}_a\}$ the configuration of the magnetic field. It is obvious that the field configuration depends not only on the original function $\mathbf{H}(\mathbf{r})$ but also on the positions of the dipoles. It can also be said that $\{\mathbf{H}_a\}$ are the fields that actually act on the particles of the aggregate (the "acting fields").

3.1. Configuration parameters of the applied field

For an integral description of the set of vectors $\{\mathbf{H}_a\}$, we can use variables analogous to those introduced in the previous section to describe the magnetic configuration $\{\mathbf{m}_a\}$. These variables include: 1) moments of the inhomogeneous field, which are analogous to multipole moments; 2) the derivatives of the field with respect to the coordinates, which are analogous to the dipole charges and currents. The general expression for a moment of order s has the form [cf. (36)]

$$A_{i_1i_2\ldots j}=\sum_a x_{ai_1}x_{ai_2}\ldots x_{ai_s}H_{aj}. \qquad (62)$$

The simplest moments—the moments of first order—are the scalar moment ψ [analogous to the moment σ defined by the expression (7)],

$$\psi=\sum_a(\mathbf{r}_a\mathbf{H}_a), \qquad (63)$$

the vector moment [analogous to the toroid moment $\boldsymbol{\tau}$ (6)],

$$\boldsymbol{\alpha}=\frac{1}{2}\sum_a [\mathbf{r}_a\mathbf{H}_a], \tag{64}$$

and the tensor moment [the analog of the quadrupole moment κ_{ik} (5)]:

$$\beta_{ik}=\frac{1}{2}\sum (x_{ai}H_{ak}+x_{ak}H_{ai}-\tfrac{2}{3}(\mathbf{r}_a\mathbf{H}_a)\delta_{ik}). \tag{65}$$

Note that the vector potential of the magnetic field is the volume density of the vector moment $\boldsymbol{\alpha}$, while the scalar potential is that of the scalar ψ; this is analogous to the relationships between the moments of the magnetization field and its potentials [see the text after Eqs. (17) and (18)].

As in the case of the multipole moments $T_{i_1...i_s j}$, not all moments of the field $A_{i_1...i_s j}$ are independent. Since the coordinates $\{\mathbf{H}_a\}$ of the "state of the field" include a finite number of variables (their number is $3n$), among all the components of the tensors $A_{i_1...i_s j}$ only $3n$ are independent, and all the remainder can be expressed in terms of them. Using, as in Sec. 3.4, generalized notation for the field coordinates H_K, $K=1,..., 3n$, and for the selected independent moments the notation A_J, we can write the relation between them in the form

$$A_J=\sum_K U_{JK}H_K, \tag{66}$$

where the form of the matrix U_{JK} can be established by comparing the expressions (62) and (66) for the given values of A_J. As A_J, moments of low order are chosen.

The values of the field derivatives of order s can also be used to parametrize the field configuration $\{\mathbf{H}_a\}$. Like the coefficients $Q_{ji_1...i_s}$, which describe the magnetic configuration of the aggregate, these are defined as the coefficients in the expression [cf. (39) and (40)]

$$H_{aj}=G_j+G_{ji_1}x_{ai_1}+G_{ji_1i_2}x_{ai_1}x_{ai_2} +...+G_{ji_1...i_s}x_{ai_1}...x_{ai_s}. \tag{67}$$

As in the case considered above of the expansion of the magnetization field $\{\mathbf{m}_a\}$, it is assumed that the sum (67) contains a finite number of terms. If as is done in (40) we separate from each tensor $G_{ji_1...}$ its irreducible parts—the tensors $G^{(sl)}_{LM}$—then the first terms of the sum (67) can be written in the form

$$\mathbf{H}_a=\mathbf{G}^{(0)}+G^{(1)}\mathbf{r}_a+[\mathbf{G}^{(1)}\mathbf{r}_a]+..., \tag{68}$$

where we have used the Cartesian components of the irreducible tensors and have retained only one superscript, which shows the order s.

Note that although the original field $\mathbf{H}(\mathbf{r})$ satisfies the Maxwell equations

$$\text{div}\,\mathbf{H}=0;\quad \text{curl}\,\mathbf{H}=0, \tag{69}$$

this does not mean that the scalar $G^{(1)}$ and vector $\mathbf{G}^{(1)}$ in Eq. (68), which are the first derivatives of the field with respect to the coordinates [cf. (44)], are zero. The fact is that the acting field $\mathbf{H}'(\mathbf{r})$, which is determined from the values $\{\mathbf{H}_a\}$ of the original field at the points $\mathbf{r}_a$, may be very different from this field $\mathbf{H}(\mathbf{r})$ itself. For example, in Fig. 4 the broken curves show the force lines of the original magnetic field $\mathbf{H}(\mathbf{r})$, which in accordance with the Maxwell equations (69) is simultaneously solenoidal and potential. But if in this field there is a two-particle aggregate arranged as is shown in Fig. 4, then the field configuration, which in the given case is specified by the two vectors $\mathbf{H}_1$ and $\mathbf{H}_2$, is such that it is natural to ascribe a solenoidal (vortex) structure to the acting field $\mathbf{H}'(\mathbf{r})$, which can be found from the values of $\mathbf{H}_1$ and $\mathbf{H}_2$. The force lines of this field are concentric circles. The method of calculating the acting field $\mathbf{H}'(\mathbf{r})$ is completely analogous to the method described in Sec. 2.3 for calculating the magnetization field $\mathbf{m}(\mathbf{r})$. This field can also be obtained by assuming that the exact expression (67) is valid not only at the points $\mathbf{r}_a$ but also in the complete volume occupied by the particles of the aggregate.

3.2. Energy of the aggregate in an external field

Returning to the expression (61) for the energy of the aggregate in an external field, we substitute for $\mathbf{H}_a$ the expansion (68). If we take into account the definition (5)–(7) of the multipole moments, then the expression (61) can be rewritten in this case in the form

$$\mathscr{E}=-\boldsymbol{\mu}\mathbf{G}^{(0)}-\sigma G^{(1)}-(\boldsymbol{\tau}\mathbf{G}^{(1)})-\kappa_{ik}G^{(1)}_{ik}-.... \tag{70}$$

Similarly, using the representation (40) for the magnetic moments $\mathbf{m}_a$, which we represent here in Cartesian form, we can write the expression for the energy (61) in the equivalent form

$$\mathscr{E}=-\boldsymbol{\mu}\langle\mathbf{H}\rangle-e_m\psi-\frac{1}{c}\mathbf{j}_m\boldsymbol{\alpha}-p_{ik}\beta_{ik}-..., \tag{71}$$

where

$$\langle\mathbf{H}\rangle=\frac{1}{n}\sum_a \mathbf{H}_a.$$

Thus, we have used the introduced parameters to calculate the energy of the particles. Moreover, as can be seen from comparison of the expressions that we have obtained, there is a kind of complementarity between the parameters—if integral parameters are used to describe the magnetic configuration, it is natural to use differential parameters for the field configuration, and vice versa.

3.3. Expansion of the field with respect to the parameter 1/L

Hitherto, we have not used any approximations in our exposition. We shall now assume that the field $\mathbf{H}(\mathbf{r})$ has inhomogeneity scale L much greater than the aggregate scale l, i.e.,

$$l\ll L. \tag{72}$$

If this condition is satisfied, the original inhomogeneous field can be expanded in the power series

$$H(\mathbf{r})=\mathbf{H}(0)-(\mathbf{r}\nabla)\mathbf{H}(0)$$
$$+...+\frac{(-1)^s}{s!}(\mathbf{r}\nabla)^s\mathbf{H}(0)+..., \quad (73)$$

where the derivatives are calculated at the point $\mathbf{r}=0$ (i.e., at the center of the aggregate). In what follows, we shall use for these quantities the abbreviated notation

$$H^{(s)}_{i_1i_2...i_sj}=\frac{1}{s!}[\nabla_{i_1}\nabla_{i_2}...\nabla_{i_s}H_j(\mathbf{r})]_{\mathbf{r}=0}. \quad (74)$$

Since the field $\mathbf{H}(\mathbf{r})$ satisfies the Maxwell equations (69), it is readily verified that the tensors $H_{i_1...i_sj}$ are, first, symmetric with respect to all their indices and not only with respect to the indices $i_1...i_s$, as follows from their definition (74). Second, the contraction on the index j and any of the indices i_k is zero, i.e., we have

$$H^{(s)}_{i_1i_2...i_sj}=H^{(s)}_{ji_2...i_si_1};\quad H^{(s)}_{ji_2...i_sj}=0. \quad (75)$$

It follows from this that all the second-rank tensors $H^{(s)}_{ji_1...i_s}$ are irreducible. Using the notation (74), we can rewrite the expansion for the field (63) in the form

$$H_j(\mathbf{r})=H^{(0)}_j-H^{(1)}_{ji_1}x_{i_1}+H^{(2)}_{ji_1i_2}x_{i_1}x_{i_2}+... . \quad (76)$$

Substituting the expression (76) in the expression (61) for the energy, we can transform it to

$$\mathscr{E}=-\mu_jH^{(0)}_j-\kappa_{jk}H^{(1)}_{jk}-\lambda_{jkl}H^{(2)}_{jkl}-\nu_{ijkl}H^{(3)}_{ijkl}-..., \quad (77)$$

where κ_{jk} is the tensor of the quadrupole moment of the aggregate, and λ_{jkl}, ν_{ijkl},... are the irreducible tensors of third, fourth,... ranks corresponding to the octupole, hexadecapole, and other multipole moments of higher rank. Thus, the expansion (77) does not contain moments of the toroid series. It would appear from this that the odd moments do not play any role in the description of the interaction of aggregates of magnetic particles with an external field. However, as we shall see, in reality this is not the case.

The fact is that aggregates of magnetic particles contain a finite number of dipoles, and therefore the number of their magnetic degrees of freedom is bounded (in contrast, for example, to the idealized toroid moment considered in Refs. 20–22, which in this sense has an infinite number of degrees of freedom, since it can represent, for example, a continuous distribution of dipoles around a ring). As was described in Sec. 2.4, in this case the multipole moments are linearly dependent, i.e., if we choose a certain system of independent moments, then it will be possible to express all the remainder linearly in terms of them. If we include among the independent moments the toroid moment (this is natural, since the ground state of the aggregate can be most simply described by means of this moment), then the dependent moments will be expressed in terms of the toroid moment, and, thus, it will occur in the expression for the energy of the aggregate in the external field.

3.4. Example: square aggregate in an external field

As an example, we consider an aggregate of four particles in the form of a square that is in the magnetic ground state shown in Fig. 3. In accordance with the results of Sec. 2.5, we choose as the variables of state of this aggregate $\boldsymbol{\mu}$, σ, $\boldsymbol{\tau}$, $\kappa_{\alpha\beta}$ (12 parameters in total). However, in the ground state only the toroid moment $\boldsymbol{\tau}$ is nonvanishing, and inversion of the equation (38) that connects the old ($\mathbf{m}_a$) and new ($\boldsymbol{\mu}$, σ, $\boldsymbol{\tau}$, $\kappa_{\alpha\beta}$) variables in this case leads to the relation

$$\mathbf{m}_a=\frac{1}{2l^2}[\boldsymbol{\tau}\mathbf{r}_a], \quad (78)$$

where $2l$ is the diagonal of the square and, in accordance with the results of Sec. 3.5, $\boldsymbol{\tau}=2l\mu_0\mathbf{e}_z$ (here, $\mathbf{e}_z$ is the unit vector along the z axis for the choice of the coordinate axes shown in Fig. 3). On the other hand, as is readily verified, in this magnetic state the moments of the dipole series include the nonvanishing fourth-order moment ν_{ijkl}, which is symmetric in all its indices and is defined by

$$\nu_{ijkl}=\sum_a\{x_{ai}x_{aj}x_{ak}m_{al}\}, \quad (79)$$

where the braces denote symmetrization of the tensor with respect to all indices. Since the moment ν_{ijkl} is dependent, it can be expressed in a definite manner in terms of the chosen independent moments (we do not give here the general expressions for the connection), and in the considered state it can be expressed solely in terms of the toroid moment as the unique nonvanishing moment. Substituting in place of the vectors $\mathbf{m}_a$ in (79) the expressions for them in terms of the toroid moment (78) and summing over the coordinates of the dipoles shown in Fig. 3, we obtain

$$\nu_{ijkl}=\tau l^2(\{e_{xi}e_{xj}e_{xk}e_{yl}\}-\{e_{yi}e_{yj}e_{yk}e_{xl}\}), \quad (80)$$

where $\mathbf{e}_x$ and $\mathbf{e}_y$ are unit vectors directed along the axes of the coordinate system (Fig. 3). Substituting (80) in (76), we finally obtain

$$\mathscr{E}=-(\boldsymbol{\tau}\mathbf{G}^{(1)}), \quad (81)$$

where $\mathbf{G}^{(1)}$ is the effective curl of the magnetic field, which in the considered approximation is

$$G^{(1)}_x=G^{(1)}_y=0;\quad G^{(1)}_z=l^2(H^{(3)}_{xxxy}-H^{(3)}_{xyyy}), \quad (82)$$

and the components of the tensor $H^{(3)}_{ijkl}$ are given in the system of axes $\mathbf{e}_x$, $\mathbf{e}_y$, $\mathbf{e}_z$.

Note that a similar result can be obtained if instead of the integrated moment $\boldsymbol{\tau}$ we use the dipole current density $\mathbf{j}_m$. In this case, the interaction energy can be written in the form

$$\mathscr{E}=-\frac{1}{c}(\mathbf{j}_m\boldsymbol{\alpha}), \quad (83)$$

where $\mathbf{j}_m=c\tau/l^2$ and $\boldsymbol{\alpha}=\mathbf{G}^{(1)}/l^2$. The appearance of the derivatives of higher order in the expression (82) for the curl of the acting magnetic field is here due to the high symmetry of the aggregate. For example, for a less symmetric aggregate in the form of a rhombus, the effective curl can be expressed in terms of the first-order derivatives $H_{ji}^{(1)}$. In this way one can consider, in particular, not only the effective curl of the field but also the effective divergence (the scalar ψ).

3.5. Expansion of the field-configuration parameters

In the general case, the derivatives of the acting field $G_{ji_1...i_s}$ can be represented in the form of expansions in powers of the parameter l/L. The general procedure of such expansions consists of the following sequence of operations. First, using the expansion (76), it is necessary to find the values of the acting field at the points $\mathbf{r}_a$. Then, using (62) and (67), it is necessary to calculate the required parameters. In this way one can readily find the integrated field moments $A_{i_1...i_s j}$. Indeed, in accordance with (62) and (76) we obtain

$$A_{i_1...i_s j}=H_j^{(0)}\sum ax_{ai_1}...x_{ai_s}+H_{jk_1}^{(1)}\times\sum ax_{ai_1}...x_{ai_s}x_{ak_1}+... . \quad (84)$$

To calculate the "differential" parameters $G_{ji_1...i_k}$, it is necessary to solve the system of equations (67) with the $3n$ unknowns $G_{ji_1...i_k}$, which in the given case can be written in the form

$$G_j+G_{ji_1}x_{ai_1}+...+G_{ji_1...i_s}x_{ai_1}...x_{ai_s}=H_j^{(0)}+H_{jk_1}^{(1)}x_{ak_1}+...+H_{jk_1...k_p}^{(p)}x_{ak_1}...x_{ak_p}. \quad (85)$$

Although at the first glance the right- and left-hand sides of this system are identical (apart from the notation for the field gradient), in reality the number of terms on the left-hand side is determined by the number of independent parameters chosen to describe the configuration of the magnetic field, while the number of terms on the right-hand side is determined by the chosen degree of approximation with respect to the expansion parameter l/L. Therefore, although the tensors of the field gradients $H_{jk_1...k_p}^{(p)}$ are symmetric in all indices, the tensors of the effective gradient $G_{ji_1...i_k}$ are in the general case symmetric only with respect to the indices $i_1,...,i_k$, as follows from their definition. The same also applies to the contractions of the tensors $G_{ji_1...i_k}$ on the index j and any of the indices i—in the general case, they are nonzero. It follows from this, in particular, that the second-rank tensor G_{ji_1} can contain an asymmetric part and may have nonzero contraction. This means that such a configuration of the magnetic field possesses an effective curl $\mathbf{G}^{(1)}$ and an effective divergence $G^{(1)}$, although, as follows from Maxwell's equations, the original static magnetic field is potential and solenoidal. The possibility of such a solution is shown qualitatively in Fig. 4 and can be demonstrated by direct calculations for a square aggregate in the ground state.

If for any reason the solution of the system of equations (85) presents difficulties, then to calculate the acting field $H'(\mathbf{r})$ we can also use an approximate method, as was described in Sec. 2.3 for the magnetic configuration $\{\mathbf{m}_a\}$.

3.6. Choice of representation of the magnetic configuration

For a more complete understanding, it is helpful to formulate the results obtained in this section by means of Dirac's notation.[46] We shall describe the magnetic state of the aggregate by a $3n$-dimensional vector (magnetic row) $\langle M|$ with components M_J, and the field configuration by a vector (magnetic column) $|H\rangle$ with components H_J. The "observable" quantity—the energy of the interaction of the aggregate with the field—can be expressed in the form of the so-called "complete bracket expression"

$$\mathscr{E}=-\langle M|H\rangle. \quad (86)$$

For the state vector of the magnetic dipoles $\langle M|$, we can use the multipole "representation" $\langle T|$, which is related to $\langle M|$ by a nondegenerate linear transformation:

$$\langle M|=\langle T|\hat{U}. \quad (87)$$

Substitution of (87) in the expression (86) makes it possible to represent the energy of the interaction of the aggregate with the magnetic field in the form

$$\mathscr{E}=-\langle T|G\rangle, \quad (88)$$

where we have introduced the vector of the magnetic-field configuration in the new representation (representation of the field by means of effective derivatives)

$$|G\rangle=\hat{U}|H\rangle. \quad (89)$$

As we have already shown, in place of the system of moments $\langle T|$ we can choose a different system of multipoles $\langle T'|$. In this language, this will mean the choice of a different representation of the state:

$$\langle T|=\langle T'|\hat{U}', \quad (90)$$

where $\hat{U}'$ is some nondegenerate matrix. From the formal point of view, the actual choice of the system of moments for describing the state ($\langle T|$ or $\langle T'|$) can be made quite arbitrarily. In particular, this system need not include the moments of the toroid series. However, as will be shown later, the actual magnetic state of the aggregate cannot be arbitrary—it is determined by the minimum of the interaction energy of the magnetic dipoles. At the same time, as we have already said, to the ground state there corresponds a global minimum of the energy, and to the remaining ("stationary") states local minima. It is clear that the description of the magnetic configuration in the representation of stationary states (in the "energy representation") is the most natural. Therefore, on the basis of these considerations, among all possible multipole representations of the magnetic state, $\langle M|$, $\langle T|$, $\langle T'|$..., the one closest to the ground state is preferable. From this point of view, the choice of the "toroid representation" (i.e., the inclusion in the system of multipoles of the toroid moment) is justified by the fact that in the ground state the interacting magnetic moments tend to form structures with closed magnetic flux, corresponding precisely to the presence of a toroid moment in this state.

The expansion of the vector of the acting field $|H\rangle$ with respect to the small parameter l/L, expressed in the form (76), can also be interpreted as a certain linear transformation of variables. Indeed, setting $\mathbf{r}=\mathbf{r}_a$ in (76), we obtain on the left the vectors $\mathbf{H}_a$, and the complete equation can be interpreted as a transformation from the variables $\mathbf{H}_a$ to the set of variables $H_j^{(0)}, H_{ji_1}^{(1)},...,H_{ji_1i_2...i_p}^{(p)}$. An easy calculation shows that the number of these variables is

$$\nu=\sum_{k=0}^{p}(2k+3)=(p+1)(p+3).$$

Thus, taking into account the terms of the expansion up to order p, we can parametrize the field by a ν-dimensional vector of gradients $|H^{(p)}\rangle$. The relation (76) (with $\mathbf{r}=\mathbf{r}_a$) is to be regarded as a linear relationship between the $3n$-dimensional vector $|H\rangle$ and the ν-dimensional vector $|H^{(p)}\rangle$, which can be written in the form

$$|H\rangle=\hat{W}|H^{(p)}\rangle, \tag{91}$$

where the matrix W is nonunitary and, in general, is not quadratic and may even be degenerate. Substituting this relation in Eq. (89), we obtain

$$|G\rangle=\hat{U}\hat{W}|H^{(p)}\rangle. \tag{92}$$

This relation represents in symbolic form Eq. (85). Taking into account the properties of the matrices $\hat{W}$ and $\hat{U}$ listed above, it is difficult to expect a correspondence between the gradients of the acting field $|G\rangle$ and the gradients $|H^{(p)}\rangle$. In particular, the presence of some components of the one does not mean that there will be similar components of the other, and vice versa.

Exactly similar arguments can be made in the case when we use as variables of state the "derivatives" of the magnetization field—the tensors $Q_{ji_1...i_s}$. We denote the state vector in this representation by the symbol $\langle Q|$. The relation between the representations $\langle M|$ and $\langle Q|$ is also given by some nondegenerate matrix $\hat{V}$:

$$\langle M|=\langle Q|\hat{V}. \tag{93}$$

The role of the field parameters in this case will be played by its integrated moments, and the energy $\mathscr{E}$ can be expressed in the form

$$\mathscr{E}=-\langle Q|A\rangle, \tag{94}$$

where we have written

$$|A\rangle=\hat{V}|H\rangle. \tag{95}$$

As in the case of multipoles, the choice of the state parameters is not unique, and the most natural parameters are determined by the same arguments as in the case of the multipole representation.

3.7. Field of a "finite" toroid at large distances

Besides the problem of expressing the energy of an aggregate in an external field in terms of the parameters that we have introduced, two more problems are of interest—the calculation of the field of the aggregate at large distances and the mutual interaction of aggregates. To solve the first problem, we shall take as our point of departure the fact that the magnetic field of the system of point dipoles can be represented in the form of the sum

$$\mathbf{H}(\mathbf{r})=\sum_a\frac{[3\mathbf{R}_a(\mathbf{m}_a\mathbf{R}_a)-\mathbf{m}_a\mathbf{R}_a^2]}{R_a^5}, \tag{96}$$

where the vector $\mathbf{R}_a$ is directed from the position of dipole a to the point of observation, and the vector $\mathbf{r}$ from the center of the aggregate to the point of observation; we have $\mathbf{R}_a=\mathbf{r}-\mathbf{r}_a$, where, as before, $\mathbf{r}_a$ is the vector of dipole a relative to the center of the aggregate. We can also rewrite (96) in the equivalent form

$$\mathbf{H}(\mathbf{r})=\sum_a(\mathbf{m}_a\nabla_a)\nabla_a\frac{1}{R_a}, \tag{97}$$

where ∇_a is the operator of the derivative with respect to the components of the vector $\mathbf{R}_a$.

From the formal point of view, the expressions (96), (97) can be regarded as some linear function of the state vector $|M\rangle$, which we can write in the form

$$\mathbf{H}(\mathbf{r})=\langle M|\mathbf{X}(\mathbf{r})\rangle, \tag{98}$$

where $|\mathbf{X}(\mathbf{r})\rangle$ are certain $3n$-dimensional vectors. Going over in this expression to a different representation of the state $\langle M|$, we can obtain the exact value of the field at the point $\mathbf{r}$ produced by the independent multipoles, among which we have included the toroid moments.

We shall now assume that the distance from the point of observation to the aggregate is so large that $r\gg r_a$, and therefore the expression for the field can be written approximately in the form of a series in powers of r_a/r:

$$\mathbf{H}(\mathbf{r})=\Big\{(\mu_k\nabla_k-\kappa_{ik}\nabla_i\nabla_k+\lambda_{ikl}\nabla_i\nabla_k\nabla_l-\nu_{ijkl}\nabla_i\nabla_j\nabla_k\nabla_l\Big\}\nabla\frac{1}{r}, \tag{99}$$

where, as in (77), κ, λ, ν,... are symmetric tensors; the operator ∇ denotes differentiation with respect to the coordinates of the vector $\mathbf{r}$. Expressing these tensors in terms of the chosen independent multipole parameters, we can obtain the expression for the magnetic field at large distances.

As a specific example, we again consider a four-particle square aggregate in the ground state. Using the expression (80) for the components of the symmetric fourth-rank tensor ν_{ijkl} in terms of the components of the toroid moment (in this case, the remaining tensors, μ, κ, λ, are zero), and calculating the derivatives with respect to the coordinates in (99), we obtain in the first nonvanishing approximation

$$\mathbf{H}(\mathbf{r})=-\frac{105}{r^6}\tau l^2((3n_x^2n_y-n_y^3)\mathbf{e}_x-(3n_xn_y^2-n_x^3)\mathbf{e}_y+9n_xn_y(n_x^2-n_y^2)\mathbf{n}), \tag{100}$$

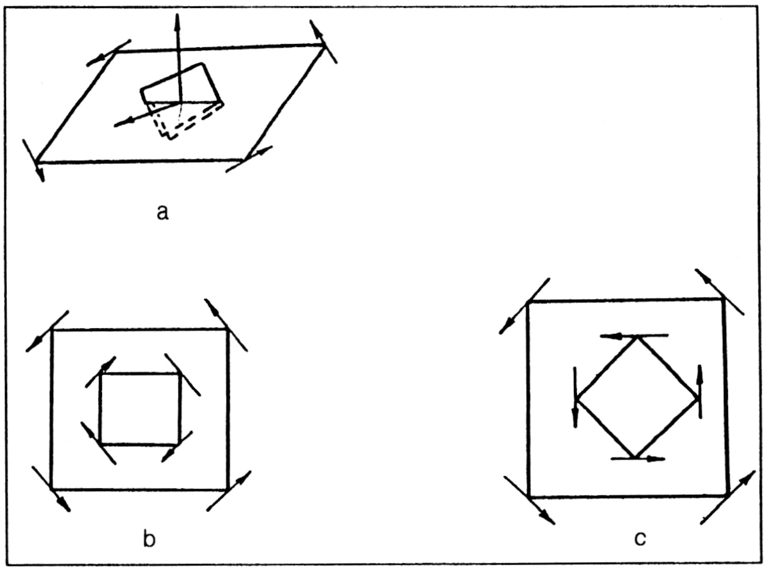


FIG. 5. Mutual disposition of the axes of the coordinate system of the coils (x,y) and of the aggregate of magnetic particles (x',y') in a general position (a) and in the position corresponding to the minimum of the energy for parallel (b) and antiparallel (c) orientation of the toroid moments.

where $\mathbf{n}$ is the unit vector directed along $\mathbf{r}$. Thus, in the given case the field of the toroid moment decreases as $1/r^6$. In the general case, the power with which the field decreases depends on the shape of the aggregate.

3.8. Toroidal aggregate at the center of a toroidal system of field sources

Another asymptotic limit of interest is to calculate the field near the center of a system of dipoles. This problem is interesting in connection with the need to choose sources that create a magnetic field of a given configuration. We shall represent a system of coils with current or a system of permanent magnets as a certain distribution of magnetic dipoles $\{\mathbf{m}_a\}$ and assume that the aggregate of magnetic particles is at the center of this macroscopic "aggregate."

In the considered case, a small expansion parameter is the ratio r/r_a, since here r is of the order of the scale of the microscopic aggregate of magnetic particles of the ferromagnetic suspension, i.e., $r\sim l$, while $\mathbf{r}_a$ is the scale of the system of coils with currents $(r_a\sim L)$. Expanding the right-hand side of the expression (96) in a series in powers of the small parameter l/L, we obtain the value of the magnetic field near the center of the system in the form

$$\mathbf{H}(\mathbf{r})=\sum_a\left(1-(\mathbf{r}\nabla'_a)+\frac{1}{2!}(\mathbf{r}\nabla'_a)^2+\ldots\right)\times(\mathbf{m}_a\nabla'_a)\nabla'_a\frac{1}{r_a}, \qquad (101)$$

where the symbol ∇'_a denotes differentiation with respect to the components of the radius vector $\mathbf{r}_a$. Comparing this expansion with the general expression (76), we can see that in the given case the gradient tensor of the magnetic field at the center of the system has the form

$$H^{(p)}_{ji_1\ldots i_p}=\frac{(-1)^p}{p!}\sum_a(\mathbf{m}_a\nabla'_a)\nabla'_{aj}\nabla'_{ai_1}\ldots\nabla'_{ai_p}\frac{1}{r_a}. \qquad (102)$$

This expression can be used to calculate the gradients for particular experimental devices that produce an inhomogeneous magnetic field at a sample.

As an example, we give calculations for a system of four identical magnetic dipoles (for example, coils with current) at the corners of a square with diagonal L and oriented in such a way that this distribution of magnetic moments possesses only a toroid moment $\boldsymbol{\tau}$ (Fig. 5a), all the remaining multipole moments being either zero or expressible in terms of $\boldsymbol{\tau}$. It is readily seen that in this case the constant part $H^{(0)}$ of the magnetic field and the gradients of first and second order, $H^{(1)}$ and $H^{(2)}$, vanish, while the first nonvanishing term appears in Eq. (101) only in the third order. Using (77), which relates the magnetic moments of the coils to the toroid moment of the system, and summing in the expression for the third-order gradient $H^{(3)}_{ji_1i_2i_3}$, given by the general expression (18), we obtain after simple calculations

$$\mathbf{H}(\mathbf{r})=-\frac{32\tau_z}{2L^7}\left(y(3x^2-y^2)\mathbf{e}_x+x(x^2-3y^2)\mathbf{e}_y\right), \qquad (103)$$

where x and y are the projections of the radius vector $\mathbf{r}$ onto the axes $\mathbf{e}_x$ and $\mathbf{e}_y$ shown in Fig. 5a.

We use the expression obtained for the field to calculate the energy of the square aggregate of magnetic particles of the suspension in the field of the considered system of coils. At the same time, we assume that the aggregate has only a toroid moment $\boldsymbol{\tau}'$ and that the remaining independent moments which describe its magnetic state are zero. To calculate the energy, we can use either the general expression (61) for the energy of the aggregate in an external field and the value (103) of the field, taken at the positions of the particles that form the aggregate, or the expression (81), in accordance with which the effective curl of the field acting on the square aggregate can be expressed in terms of the gradient of third order, and the energy can then be found in accordance with (80) as the product of the toroid moment $\boldsymbol{\tau}'$ and the effective curl

$\mathbf{G}^{(1)}$. As a result of these calculations, the interaction energy of the two toroid moments $\boldsymbol{\tau}'$ and $\boldsymbol{\tau}$—of the system of coils and of the microscopic aggregate at its center—can be found in the form

$$\mathscr{E}_\tau = \frac{35 l^2}{2L^7} (\boldsymbol{\tau}\mathbf{e}_z)(\boldsymbol{\tau}'\mathbf{e}_z') [c_{12}c_{21}(3c_{11}^2 - c_{21}^2 - c_{12}^2 + 3c_{22}^2) + c_{11}c_{22}(c_{11}^2 - 3c_{21}^2 - 3c_{12}^2 + c_{22}^2)], \quad (104)$$

where $c_{ik} = (\mathbf{e}_i \mathbf{e}_k')$ are the elements of the matrix of the transformation from the one system of coordinates ($\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z$) associated with the system of coils to the other system ($\mathbf{e}_x', \mathbf{e}_y', \mathbf{e}_z'$) associated with the microscopic aggregate.

The expression (104) for the energy depends on the mutual orientation of the aggregates and their toroid moments. We assume that the coordinate axes $\mathbf{e}_z$ and $\mathbf{e}_z'$ are parallel. In this case, the transformation matrix has elements $c_{11} = c_{22} \cos\varphi$, $c_{12} = -c_{21} = \sin\varphi$, where φ is the angle between the axes $\mathbf{e}_x$ and $\mathbf{e}_x'$ (Fig. 5a). With allowance for this, the expression (104) can be transformed to

$$\mathscr{E}_\tau = 280 \frac{l^2}{L^7} (\tau_z \tau_z') \left(\cos^4\varphi - \cos^2\varphi + \frac{1}{8} \right). \quad (105)$$

It is easy to show that the function of the angle φ in this expression in the round brackets has three maxima on the interval of angles $-\pi/2 \leqslant \varphi \leqslant \pi/2$, which are at the points $\varphi = 0$ and $\pm\pi/2$, and two minima at $\varphi = \pm\pi/4$. It follows from this that for parallel toroid moments ($\boldsymbol{\tau} \| \boldsymbol{\tau}'$) the advantageous orientation of the aggregate is one for which $\varphi = \pm\pi/4$ (Fig. 5b), while for antiparallel orientation of $\boldsymbol{\tau}$ and $\boldsymbol{\tau}'$ the arrangement with $\varphi = 0$, $\pm\pi/2$ (Fig. 5c) is advantageous. This result is in agreement with simple physical arguments. In the case of parallel toroid moments, it is advantageous to have a mutual orientation of the dipoles for which the moments of one system close the magnetic fluxes in the spaces between the dipole moments of the other. For an antiparallel arrangement, the dipole moments of the particles close each other pairwise. It is curious that the value of the energy at the point of the minimum in the two cases is the same, and therefore parallel and antiparallel orientations of the toroid moments of the systems are equally advantageous. The difference between them is solely in the different orientation of the aggregates themselves.

The order of magnitude of the energy $\mathscr{E}$ of the toroid interaction is appreciably less than the interaction energy of two dipoles μ and μ' at distance $L(\mathscr{E}_\mu \sim \mu\mu'/L^3)$; an isolated particle of a suspension (embryon) with moment μ' at distance L from a coil with current has, for example, such an order of magnitude. Indeed, bearing in mind that $\tau \sim L\mu$, $\tau' \sim l\mu'$, we conclude from (105) that at the point of the minimum of the energy the toroid interaction has the form

$$\mathscr{E}_\tau \sim 30 \mathscr{E}_\mu (l/L)^3, \quad (106)$$

i.e., the ratio $\mathscr{E}_\tau / \mathscr{E}_\mu$ is an order of magnitude greater than $(l/L)^3$. For an estimate, we note that if, for example, as a source of an inhomogeneous magnetic field we use thin magnetic wires with transverse magnetization, as has been done recently to study the magnetic moments of molecular and supermolecular structures of cells, cell fragments, and organelles,[47] then the scale of the external device L is a few micrometers ($L \sim 10^{-4}$ cm). If the aggregate measures $l \sim 10^{-5}$ cm, the ratio $\mathscr{E}_\tau / \mathscr{E}_\mu$ of the energies is 10^{-2}.

3.9. Interactions of toroidal aggregates at large distances

Finally, we consider the interaction of aggregates with one another. Let $\{\mathbf{m}_a\}$ and $\{\mathbf{m}_b'\}$ be the given dipole moments of two aggregates, which we shall here regard as rigid formations. The energy of the dipole interaction between the particles of the aggregates has the form

$$\mathscr{E} = -\sum_a \sum_b (\mathbf{m}_a \nabla_{ab})(\mathbf{m}_b \nabla_{ab}) \frac{1}{r_{ab}}. \quad (107)$$

In this expression, we have omitted the internal energy of the aggregates, since we are only interested in the interaction of the aggregates with one another. By means of the generalized notation introduced earlier, we can rewrite the expression (107) in the form

$$\mathscr{E} = -\langle M | \hat{Z} | M' \rangle, \quad (108)$$

where $\langle M|$ and $|M'\rangle$ are the state vectors of the first and second system of dipoles, and $\hat{Z}$ is a matrix that depends on the mutual disposition of the particles. The interaction energy can also be expressed in the multipole representation

$$\mathscr{E} = -\langle T | \hat{U} \hat{Z} \hat{U}' | T' \rangle, \quad (109)$$

where $\hat{U}$ and $\hat{U}'$ are the matrices of the transition to the new representation. Thus, the energy can be expressed in terms of a system of independent multipole moments that describe the magnetic state of each of the aggregates. In particular, this makes it possible to estimate the energy of the aggregates at large distances from each other.

As an example, we estimate the asymptotic dependence of the energy on the distance for two four-particle aggregates, each of which is in the ground state. We shall assume that the distance L between the centers of the aggregates is much greater than the scale of the aggregates themselves: $l \sim l' \ll L$. When the energy (107) is expanded in a series in the parameter l/L, there will appear only multipole moments of the magnetic-dipole series [see the expression (99) introduced above], which we have denoted by the symbols μ, κ, λ, ν,... . For the two considered aggregates, only the hexadecapole moments ν and ν' are nonvanishing, and they are uniquely related to the toroid moments τ and τ' in accordance with the expression (80). Therefore, in the first nonvanishing approximation the interaction between these aggregates is manifested only as a hexadecapole–hexadecapole interaction:

$$\mathscr{E} \sim \frac{\nu\nu'}{L^9} \sim \frac{\tau\tau' l^2 l'^2}{L^9}. \quad (110)$$

Thus, this interaction decreases very rapidly (as $1/L^9$) with the distance between the aggregates, and therefore with good accuracy it can be ignored. In particular, this

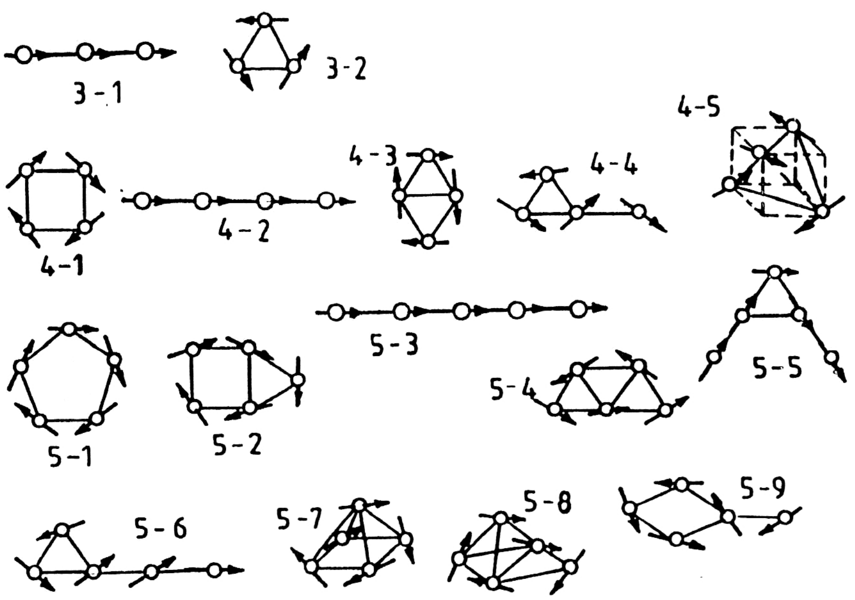


FIG. 6. Possible shapes of dipole aggregates of magnetic particles with a number of particles $N=3, 4, 5$. The aggregates are labeled by the symbols $N-i$ in order of the decrease of their binding energy for given N.

justifies the validity of a model of a magnetic suspension treated as a "gas" of noninteracting aggregates of magnetic particles.

4. TOROID POLARIZABILITY OF SUSPENSIONS

4.1. Model of rigid aggregates

As we have already noted in the Introduction, aggregation of magnetic particles greatly influences the properties of suspensions. We shall assume that the number density N of the particles (embryons) is given, and we shall consider in a simple example how the susceptibility changes when aggregates are formed. In a weak magnetic field, the magnetization of a suspension without aggregation has in accordance with (111) the form

$$\mathbf{M}=\chi_0\mathbf{H};\quad \chi_0=\mu_0^2N/3kT, \tag{111}$$

where χ_0 is the magnetic susceptibility of the suspension, μ_0 is the absolute magnetic moment of the embryon, k is Boltzmann's constant, and T is the temperature. We now suppose that three-particle aggregates have formed in the suspension. Then the effective number density of the particles is reduced by a factor 3, while the magnetic moment of each aggregate depends on its type. If the aggregates form as a result of magnetic forces, then, as is shown in Fig. 6, *3–1*, the magnetic moment of an individual aggregate will be $3\mu_0$, and the susceptibility of the aggregated suspension will in accordance with (111) be

$$\chi=(3\mu_0)^2(N/3)/3kT=\mu_0^2N/kT, \tag{112}$$

i.e., it will be three times greater than in the suspension without aggregation. But if the aggregates form as a result of nonmagnetic forces, then, as can be seen from Fig. 7, *3–1*, the magnetic moment of an aggregate will in fact be zero, and, accordingly, so will the magnetic susceptibility. However, in this case the suspension may be polarized by a solenoidal magnetic field, and this can be described by a new characteristic—the toroid susceptibility.

In the general case, as was shown in the previous sections, aggregates possess both magnetic moment and toroid moment. Therefore, they can acquire toroid polarization when a homogeneous magnetic field is imposed or, conversely, a magnetic polarization in a solenoidal magnetic field. The connection between the magnetic and toroid moments of the suspension, which we shall denote by $\mathbf{M}$ and $\mathbf{T}$, on the one hand, and the applied fields $\mathbf{H}$ and $\mathbf{G}$, on the other, can be expressed in the linear approximation in the form

$$\begin{aligned} M_i&=\chi_{ik}^{(M)}H_k+\chi_{ik}^{(MT)}G_k,\\ T_i&=\chi_{ik}^{(TM)}H_k+\chi_{ik}^{(T)}G_k, \end{aligned} \tag{113}$$

where we have introduced the magnetic, $\chi^{(M)}$, and toroid, $\chi^{(T)}$, susceptibilities, and also "crossed" susceptibilities. In the general case, all these quantities are functions of the temperature and of the amplitude of the applied fields, and they also depend on the number of embryons and on the shape of the aggregate. The calculation of the suspension susceptibilities is in the general case a complicated problem, and we therefore restrict ourselves to the most important but fairly simple models.

The choice of a particular model of the aggregate depends on the nature of the interaction between the particles that constitute it. In what follows, we shall use a model of an aggregate with a fixed shape (or with fixed relative disposition of the particles within the aggregate). This model is valid if the mean energy $\bar{U}$ of the interaction between the particles is greater than the thermal energy and greater than the energy of the interaction with the external field:

$$\bar{U}\gg kT,\quad \bar{U}\gg\mu_0H. \tag{114}$$

We now separate from the energy $\bar{U}$ the mean energy $\bar{U}_m$ of the magnetic interaction between the particles. If the inequalities (114) also hold for the magnetic energy, i.e.,

$$\bar{U}_m\gg kT,\quad \bar{U}_m\gg\mu_0H, \tag{115}$$

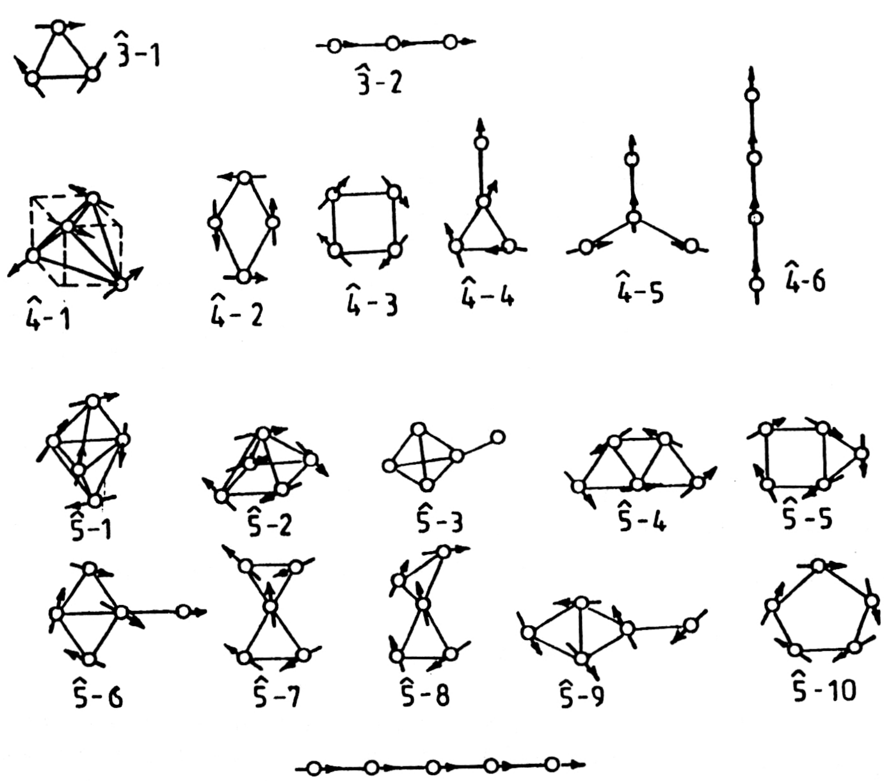


FIG. 7. Possible shapes of "nonmagnetic" aggregates with a number of particles $N \leqslant 5$. The aggregates are labeled by the symbols $\hat{N}-i$ in the order of decrease of their binding energy for given N.

then it can also be assumed that the magnetic moments of the particles have fixed values, i.e., they are frozen in the "body" of the aggregate.

Another model that can be considered here is one in which the aggregate has a fixed spatial structure but the magnetic moments of the embryons are not fixed. If this model is to be valid, the inequalities (114) must still hold, while the inequalities (115) must be replaced by approximate equalities. It is obvious that in this case we assume that the aggregate is formed by nonmagnetic forces, which are large compared with the dipole–dipole interaction.

If the second of the inequalities in (114) is replaced by an approximate equality, then in this case thermal motion does not yet disrupt the aggregates themselves, but under the influence of the applied field both the magnetic and the spatial degrees of freedom are "softened," i.e., the aggregates can change their shape. In such a strong field, the shape of the aggregates ceases to be compact and becomes elongated, i.e., in this case so-called chain aggregates are formed. This model is fairly well described in the literature (see the reviews and monographs of Refs. 1–11), and therefore we shall not consider it. In addition, chain aggregates do not possess toroid moments, and therefore they are of no interest for us from this point of view. Finally, we note that if the opposite inequalities hold in the relations (114), the thermal motion disrupts the aggregates, and we return to the case of a suspension without aggregation, which has also been considered in detail in the literature.

4.2. Polarization of a suspension in an external field (model of frozen dipoles)

The suspended particles of a magnetic suspension execute a Brownian motion, and therefore to calculate the observable quantities it is necessary to average over the thermal fluctuations. For rigid aggregates with frozen magnetic moments of the embryons, the rotational Brownian motion plays the main role. The aggregate rotates as a whole, remaining unchanged in shape and without changing its internal magnetic structure. In this case, the energy of the internal interaction of the aggregate can be assumed to be constant, and, therefore, it need not be taken into account in the distribution function with respect to the orientation angles. Thus, this function can be expressed in the form

$$W(\Omega) = C \exp[(\mathbf{e}\cdot\boldsymbol{\xi}) + (\mathbf{n}\cdot\boldsymbol{\zeta})], \tag{116}$$

where for the energy in the argument of the exponential we have used the expression $U_f = -\boldsymbol{\mu}\mathbf{H} - \boldsymbol{\tau}\mathbf{G}$, and for the dimensionless fields we have introduced the notation

$$\boldsymbol{\xi} = \mu\mathbf{H}/kT, \quad \boldsymbol{\zeta} = \tau\mathbf{G}/kT. \tag{117}$$

Here, $\boldsymbol{\mu}$ and $\boldsymbol{\tau}$ denote, respectively, the magnetic and toroid moments of the aggregate, and $\mathbf{e}$ and $\mathbf{n}$ are unit vectors parallel to them. The constant in Eq. (116) must be determined from the normalization condition

$$\int W(\Omega) d\Omega = 1. \tag{118}$$

By Ω, we understand the set of Euler angles θ, φ, and ψ (Fig. 8).

The field vectors $\mathbf{H}$ and $\mathbf{G}$ are fixed in the laboratory coordinate system S, while the unit vectors $\mathbf{e}$ and $\mathbf{n}$ are fixed relative to the coordinate system S_0 rigidly attached to the aggregate (Fig. 8). Thus, the components of the vectors $\mathbf{e}$ and $\mathbf{n}$ depend on the Euler angles that determine the orientation of the one system of coordinates relative to the other.

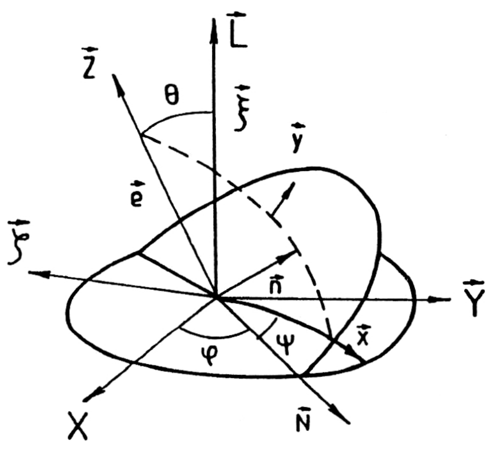


FIG. 8. Positions of coordinate axes of moving and fixed systems. The magnetic and toroid moments of the aggregate are rigidly attached to the moving system of coordinates, and therefore their components with respect to the fixed system of coordinates (with respect to which the homogeneous and solenoidal magnetic fields are fixed) depend on the Euler angles.

The mean magnetic and toroid moments of the suspension (per unit volume) can be calculated by means of the distribution function as follows:

$$\mathbf{M}=N\langle\boldsymbol{\mu}\rangle=\mu N\int \mathbf{e}W(\Omega)d\Omega,$$
$$\mathbf{T}=N\langle\boldsymbol{\tau}\rangle=\tau N\int \mathbf{n}W(\Omega)d\Omega. \qquad (119)$$

We find these quantities for the case of weak fields (or, which is the same thing, for the case of high temperatures), when the conditions $\xi\ll1$, $\zeta\ll1$ hold. In this approximation, the distribution function (116) can be expressed approximately in the form

$$W(\Omega)\approx\frac{1}{8\pi^2}[1+(\mathbf{e}\cdot\boldsymbol{\xi})+(\mathbf{n}\cdot\boldsymbol{\zeta})]. \qquad (120)$$

Taking into account the values of the integrals

$$\int \mathbf{e}d\Omega=\int \mathbf{n}d\Omega=0,\quad \frac{1}{8\pi^2}\int e_in_kd\Omega=\frac{1}{3}(\mathbf{en})\delta_{ik},$$
$$\frac{1}{8\pi^2}\int e_ie_kd\Omega=\frac{1}{8\pi^2}\int n_in_kd\Omega=\frac{1}{3}\delta_{ik}, \qquad (121)$$

we obtain

$$\mathbf{M}=\tfrac{1}{3}\mu N(\boldsymbol{\xi}+(\mathbf{en})\boldsymbol{\zeta}),\quad \mathbf{T}=\tfrac{1}{3}\tau N((\mathbf{en})\boldsymbol{\xi}+\boldsymbol{\zeta}). \qquad (122)$$

From these expressions, on the basis of the definitions (113), we can find the susceptibilities of the suspension [for the transformations, it is necessary to use the definition (113) of the vectors $\boldsymbol{\xi}$ and $\boldsymbol{\zeta}$]:

$$\chi_{ik}^{(M)}=(\mu^2N/3kT)\delta_{ik},\quad \chi_{ik}^{(T)}=(\tau^2N/3kT)\delta_{ik},$$
$$\chi_{ik}^{(MT)}=\chi_{ik}^{(TM)}=(\mu\tau N(\mathbf{en})/3kT)\delta_{ik}. \qquad (123)$$

Note that in the given approximation the crossed susceptibilities vanish for mutually perpendicular magnetic and toroid moments of the aggregate.

In the general case of arbitrarily oriented and not small fields **H** and **G**, the integrals (119) can be expressed in terms of the so-called generalized Bessel functions.[53] We shall not give here the corresponding cumbersome general expressions but limit ourselves to some special cases. For parallel fields **H** and **G**, the distribution function can be represented in the form

$$W(\Omega)=\frac{1}{\sinh\xi'}\exp(\mathbf{e}'\boldsymbol{\xi}'), \qquad (124)$$

where the unit vector $\mathbf{e}'$ and the field $\boldsymbol{\xi}'$ are defined by the equations

$$\mathbf{e}'=(\mathbf{e}\xi+\mathbf{n}\zeta)/(\xi^2+2\xi\zeta(\mathbf{en})+\zeta^2)^{1/2},$$
$$\xi'=\xi(1+2(\zeta/\xi)(\mathbf{en})+(\zeta/\xi)^2)^{1/2}. \qquad (125)$$

After simple calculations (for more details, see Ref. 54) the magnetic and toroid moments of the suspension are obtained in the form

$$\mathbf{M}=\mu N(\mathbf{ee}')L_1(\xi')\mathbf{h}',\quad \mathbf{T}=\tau N(\mathbf{ne}')L_1(\xi')\mathbf{h}', \qquad (126)$$

where $L_1(\xi)$ is the Langevin function defined by the relation (1), and $\mathbf{h}'$ is the unit vector directed along the field $\boldsymbol{\xi}'$.

The case in which the magnetic and toroid moments of the aggregate are parallel ($\mathbf{n}=\mathbf{e}$) is just as simple. The magnetic and toroid moments of the suspension have the same form (126), in which, however, it must be borne in mind that

$$\mathbf{e}'=\mathbf{n}=\mathbf{e},\quad \boldsymbol{\xi}'=\boldsymbol{\xi}+\boldsymbol{\zeta}. \qquad (127)$$

The case when one of the fields, ξ or ζ, is weak can also be treated relatively easily. We shall assume that $\zeta\ll1$. This enables us to express the distribution function in the form

$$W(\Omega)=\frac{1}{\sinh\xi}e^{(\mathbf{e}\boldsymbol{\xi})}[1+(\mathbf{n}\boldsymbol{\zeta})]. \qquad (128)$$

To calculate the moments, we use the expressions for the mean values of an arbitrary vector p_i and an irreducible second-rank tensor q_{ik} with respect to the distribution $\exp(\mathbf{e}\boldsymbol{\xi})/\sinh\xi$ obtained in Ref. 54:

$$\langle p_i\rangle_0=L_1(\xi)(\mathbf{ep})\mathbf{h},\quad \mathbf{h}=\boldsymbol{\xi}/\xi,$$
$$\langle q_{ik}\rangle_0=L_2(\xi)(q_{jl}e_je_l)(h_ih_k-\tfrac{1}{3}\delta_{ik}), \qquad (129)$$

where we have written $\langle\ldots\rangle_0=\int\ldots\exp(\mathbf{e}\boldsymbol{\xi})d\Omega/\sinh\xi$, $L_2(\xi)=1-3L_1(\xi)/\xi$.

After simple manipulations, we obtain

$$\mathbf{M}=\mu N\{L_1\mathbf{h}+\tfrac{1}{3}(\mathbf{en})\boldsymbol{\zeta}+\tfrac{2}{3}(\mathbf{en})L_2[(\mathbf{h}\boldsymbol{\zeta})\mathbf{h}-\tfrac{1}{3}\boldsymbol{\zeta}]\},$$
$$\mathbf{T}=\tau N\{(\mathbf{en})L_1\mathbf{h}+\tfrac{1}{3}\boldsymbol{\zeta}+[(\mathbf{en})^2-\tfrac{1}{3}]\times L_2[(\mathbf{h}\boldsymbol{\zeta})\mathbf{h}-\tfrac{1}{3}\boldsymbol{\zeta}]\}, \qquad (130)$$

where for brevity we have omitted the arguments of the functions L_1 and L_2 and introduced the unit vector $\mathbf{h}=\boldsymbol{\xi}/\xi$. The expressions (122) can be found from (130) in the limit $\xi\to0$ with allowance for the fact that for small ξ we have approximately $L_1\approx\xi/3$, $L_2\approx\xi^2/15$. In the opposite limiting case of strong fields ($\xi \gg 1$), we have $L_1 \approx 1$, $L_2 \approx 1$, and this enables us to find the values of the moments readily in this limit from (130). Note also that in the absence of a solenoidal field ($\zeta = 0$) it follows from (130) that

$$\mathbf{M} = \mu N L_1 \mathbf{h}, \quad \mathbf{T} = \tau N(\mathbf{en}) L_1 \mathbf{h}. \tag{131}$$

All the expressions that we have obtained apply to aggregates of only one type. In reality, in a suspension there is an entire range of aggregates of different kinds, and the distribution function with respect to the number of particles in an aggregate varies with time (the magnetic suspension "ages"). For a given number of particles, there exist several equilibrium shapes differing both in binding energy and in magnetic parameters (see Figs. 6 and 7). We shall assume that the binding energy of an aggregate of species a consisting of n particles is equal to E_{na}, and that its magnetic and toroid moments are, respectively, μ_{na} and τ_{na}. It follows from the previous consideration that the contributions to the magnetization and toroid moment of the suspension from these aggregates are

$$\begin{aligned} \mathbf{M}_{na} &= N_{na} \mathbf{m}(\xi_{na}, \zeta_{na}, \theta_{na}), \\ \mathbf{T}_{na} &= N_{na} \mathbf{t}(\xi_{na}, \zeta_{na}, \theta_{na}), \end{aligned} \tag{132}$$

where N_{na} is the number of considered aggregates per unit volume, and θ_{na} is the angle between the vectors $\mathbf{e}_{na}$ and $\mathbf{n}_{na}$. For the averaging over the distribution of aggregates of different species but with a given number n of particles, we use the distribution function

$$N_{na} = N_n e^{-E_{na}/kT} / \sum_a e^{-E_{na}/kT}, \tag{133}$$

where N_n is the number of aggregates with the given n.

To calculate N_n, it is necessary to solve the problem of aggregation kinetics. The best known model was constructed by Smoluchowski. Assuming that the aggregation of the particles takes place in accordance with the type of the chemical reaction of polymerization $x_1 + x_n = x_{n+1}$ with a certain given time constant t_0, we can find the time dependence of N_n in the form (Smoluchowski's formula)

$$N_n(t) = N_n(0) \frac{(t/t_0)^{n-1}}{1 + (t/t_0)^{n+1}}, \tag{134}$$

where $N_n(0)$ is the initial number of embryons, and t_0 is the time during which $N_n(0)/2$ aggregates.

As a result, the moments in which we are interested will be found with allowance for the aggregate distribution:

$$\begin{aligned} \mathbf{M} &= \sum_{n,a} N_{na} \mathbf{m}(\xi_{na}, \zeta_{na}, \theta_{na}), \\ \mathbf{T} &= \sum_{n,a} N_{na} \mathbf{t}(\xi_{na}, \zeta_{na}, \theta_{na}). \end{aligned} \tag{135}$$

These calculations were made numerically. Figure 9 shows the magnetization of the suspension and its toroid moment as functions of the time for different temperatures and fields.

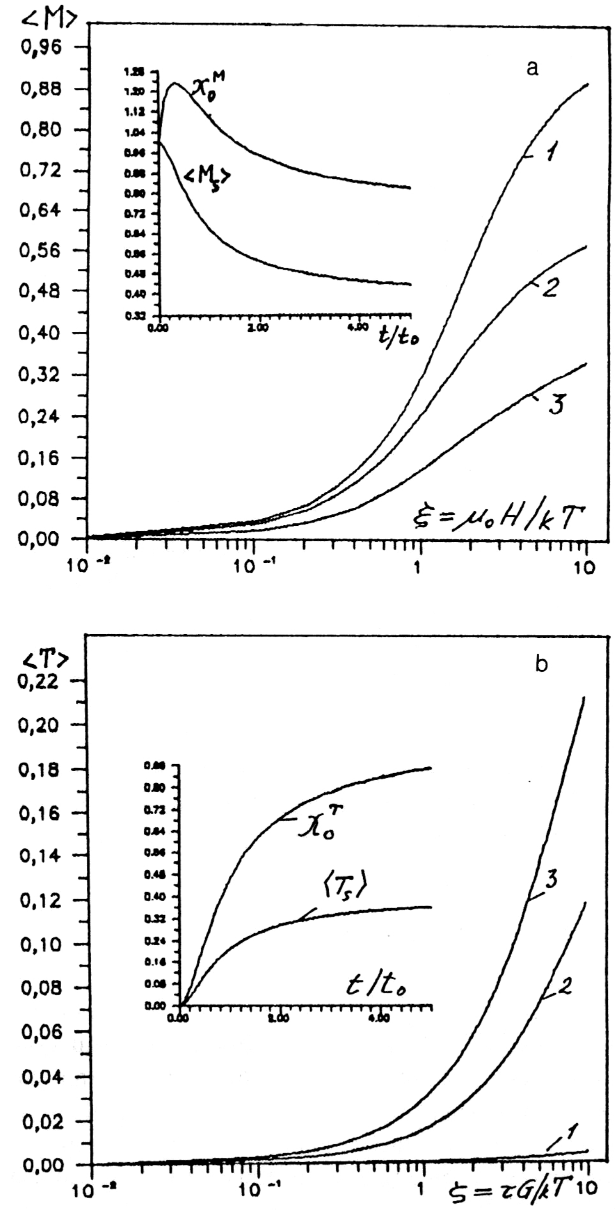


FIG. 9. Dependence of magnetic (a) and toroid (b) moments on the dimensionless fields ξ and ζ with allowance for coagulation of particles. Curves *1–3* correspond to values of the dimensionless time t/t_0 equal to 0.1, 1, and 5, respectively. The insets on the left in Figs. 9a and 9b show the time dependence of the magnetic and toroid initial susceptibilities χ_0^M and χ_0^T, which are proportional to the slopes of curves *1, 2*, and *3* at small values of the fields ξ and ζ. Also shown is the time dependence of the limiting magnetization of the suspension and its toroid moment (as $\xi \to \infty$ and $\zeta \to \infty$, respectively). As can be seen from the figure, if the particles coagulate, the magnetic parameters of the suspension decrease with time, while the toroid parameters increase.

4.3. Model of mobile dipoles

If it is assumed that the magnetic moments of the particles are rigidly attached to the spatial configuration of the aggregate, as in the previous section, then the influence of external fields on the orientation diffusion of the particles will be maximal. In the other limiting case, when the magnetic moments of the embryons are weakly coupled and can be freely oriented under the influence of an external field (limit of superparamagnetic aggregates), external fields do not influence the Brownian diffusion of the particles, and by magnetic measurements one cannot learn about the aggregation structure of the suspension. This case is of no interest for us, since it reduces to the consideration of a system of independent particles.

In the general case, to find the mean values of the moments in which we are interested it is necessary to use a distribution function of the form

$$dW(\mathbf{m}_1,...,\mathbf{m}_n,\Omega)$$
$$=Ce^{-U(\mathbf{m}_1,...,\mathbf{m}_n,\Omega)/kT}d\mathbf{m}_1 d\mathbf{m}_2...d\mathbf{m}_n d\Omega, \quad (136)$$

where $U(\mathbf{m}_1,...,\mathbf{m}_n,\Omega)$ is the energy of the magnetic interaction of the embryons with one another and with the external fields. At the same time, the integration over the moments $\mathbf{m}_a$ of the individual particles (embryons) must be made with allowance for the constancy of the lengths of these vectors. The calculation by means of such a distribution function encounters great difficulties in both analytic and numerical calculation, and therefore approximate methods of calculation are usually employed.

If the internal fields at the particles of the aggregate are large compared with the external fields, then measurements of the moments of the aggregate can be taken into account by introducing its magnetic and toroid polarizabilities in accordance with the definition

$$\mu_i=\mu_i^{(0)}+\kappa_{ik}^{(m)}H_k+\kappa_{ik}^{(m\tau)}G_k,$$
$$\tau_i=\tau_i^{(0)}+\kappa_{ik}^{(\tau m)}H_k+\kappa_{ik}^{(\tau)}G_k, \quad (137)$$

where $\mu_i^{(0)}$ and $\tau_i^{(0)}$ are the values of $\boldsymbol{\mu}$ and $\boldsymbol{\tau}$ in the absence of a field, and the polarizability tensors depend on the shape of the aggregate and have principal axes rigidly fixed relative to the spatial disposition of the particles. Expressing the energy of the aggregate in the external field in the form $U=-(\boldsymbol{\mu}\mathbf{H})-(\boldsymbol{\tau}\mathbf{G})$ and substituting in this expression the values of $\boldsymbol{\mu}$ and $\boldsymbol{\tau}$ from (137), we obtain

$$U=-\boldsymbol{\mu}^{(0)}\mathbf{H}-\boldsymbol{\tau}^{(0)}\mathbf{G}-\kappa_{ik}^{(m)}H_iH_k-\kappa_{ik}^{(\tau)}G_iG_k$$
$$-(\kappa_{ik}^{(m\tau)}+\kappa_{ki}^{(\tau m)})G_kH_i. \quad (138)$$

Note that in this expression it is possible to omit the contractions of the tensors in front of H_iH_k, G_iG_k, and G_iH_k, since these quantities do not depend on the angles of the orientation of the aggregate.

Restricting ourselves to the case of weak fields, we can write the distribution function $W(\Omega)$ in the form $W(\Omega)\approx C(1-U/kT)$. Using the value of the energy (138) and the values of the moments (137), we can calculate the mean magnetic and toroid moments of the suspension (for aggregates of a given species):

$$\mathbf{M}=\mu N\left\{\left[\frac{1}{3}+\frac{2}{15}\kappa_{ik}^{(m)}\kappa_{ik}^{(m)}\frac{H^2}{\mu^2}\right]\boldsymbol{\xi}+\left[\frac{1}{3}\,\mathbf{en}+\frac{1}{30}\kappa_{ik}^{(m)}(\kappa_{ik}^{(\tau m)}+3\kappa_{ik}^{(m\tau)})\frac{H^2}{\mu\tau}\right]\boldsymbol{\zeta}\right\},$$

$$\mathbf{T}=\tau N\left\{\left[\frac{1}{3}\,\mathbf{en}+\frac{2}{15}\kappa_{ik}^{(\tau m)}\kappa_{ik}^{(m\tau)}\frac{H^2}{\mu\tau}\right]+\left[\left[\frac{1}{3}+\frac{1}{10}\kappa_{ik}^{(\tau m)}(\kappa_{ik}^{(m)}+\kappa_{ik}^{(m\tau)})\frac{H^2}{\tau^2}\right]-\frac{1}{15}\kappa_{ik}^{(m)}\kappa_{ik}^{(\tau)}\frac{H^2}{\tau^2}\right]\boldsymbol{\zeta}\right\}. \quad (139)$$

These expressions were obtained under the assumptions that $\kappa_{ik}^{(m\tau)}$ and $\kappa_{ik}^{(\tau m)}$ are symmetric in the indices i and k, an assumption that holds for most types of aggregate, that $\mathbf{G}\perp\mathbf{H}$, and that $\mathbf{G}$ can be taken into account in the first order. The most important qualitative result that follows from these expressions is that, in contrast to the case of a nonaggregated suspension, the magnetic moments of the aggregates can change under the influence of the external field, and therefore the dependence of the mean values (139) on the Langevin argument, which is usually used to interpret the results, has a more complicated nature than would be the case for nonaggregated suspensions or for a suspension of aggregates with "frozen" moments. This can be most clearly seen for the case when in the absence of a field the aggregates did not have magnetic moments at all ($\boldsymbol{\mu}^{(0)}=0$). Then in the initial section of the curve $\mathbf{M}(\mathbf{H})$, a nonlinear dependence on the field will be observed. It is a dependence of this kind that is most often obtained when measurements are made on an aggregated suspension.[1-4] To interpret the results, some authors (see, for example, Ref. 34) introduce effective anisotropy constants of the suspension particles, etc. For a complete solution to the problem of which precise mechanism of occurrence of the quadratic dependence of the magnetization on the field in the initial section of the curve plays the main role—the spread of particles of the nonaggregated suspension in size or the aggregation of particles and "difficult" polarization of the aggregates—it is necessary to make measurements of the toroid susceptibility of the suspension; for it is obvious that isolated particles cannot contribute to such a susceptibility.

For nonsmall values of the fields, the magnetic and toroidal moments can be found by numerical integration. However, the use of the distribution function (136) makes it necessary to calculate multidimensional integrals, and this presents considerable computational difficulties. Instead of this, to calculate the mean values, we can use the following approximate procedure. We assume that the aggregates are at rest and that the fields $\mathbf{H}$ and $\mathbf{G}$ rotate at random around them. We shall here assume that the fields and also their mutual orientation are given. It is obvious that in this case the distribution function will depend on the three Euler angles Ω that determine the orientation of the frame that can be constructed using the vectors $\mathbf{H}$ and $\mathbf{G}$ with respect to the fixed aggregate (in the case when the vectors $\mathbf{H}$ and $\mathbf{G}$ are parallel, the problem simplifies, since it is sufficient to specify only two Euler angles instead of three). For each given orientation of the fields, we can find the distribution of the magnetic dipoles of the aggregate numerically by calculating the minimum of the interaction energy. As a result, for each Ω we shall find $E(\Omega,G,H)$, $\boldsymbol{\mu}(\Omega,G,H)$, and $\boldsymbol{\tau}(\Omega,G,H)$. After this, averaging over Ω can be done by using the distribution function

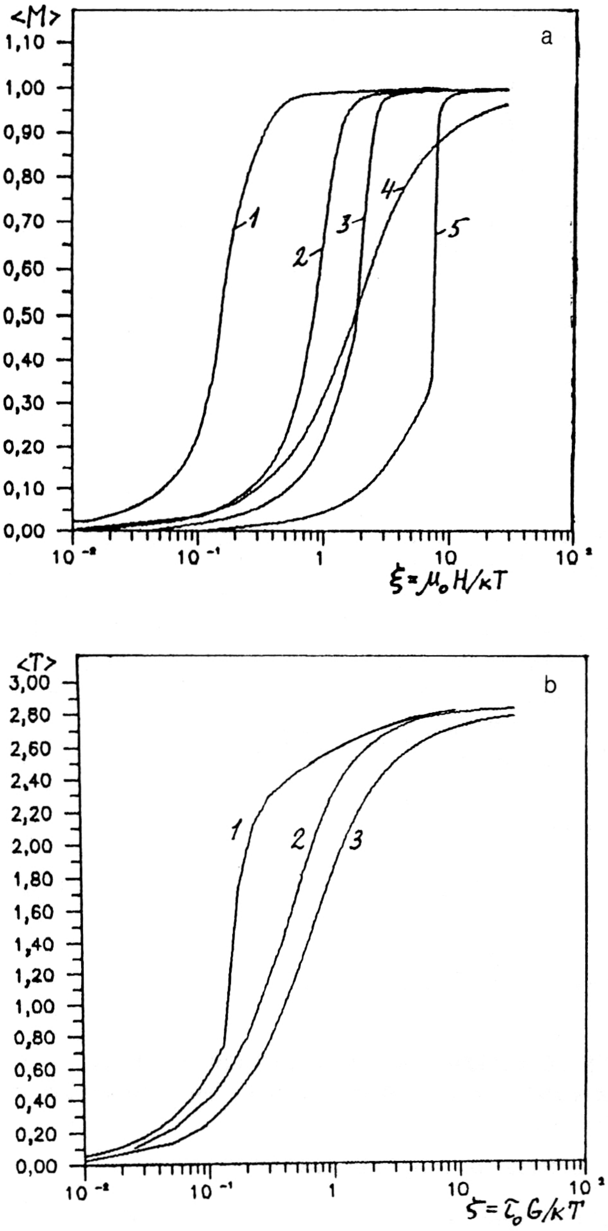


FIG. 10. Dependence of the magnetization (a) and toroid moment (b) of a suspension consisting of four-particle aggregates in the shape of a square with mobile dipoles on the dimensionless fields ξ and ζ for different values of the reduced temperature $\beta=\mu_0^2/a^3kT$ (curves *1–3* and *5* in Fig. 10a correspond to $\beta=0.1$, 0.5, 1, and 5, and curves *1* and *2* in Fig. 10b correspond to $\beta=0.1$ and 4). For comparison, we show in Figs. 10a and 10b the Langevin functions (curves *4* and *3*, respectively) that describe the limiting cases of superparamagnetic aggregates (a) and rigid aggregates (b).

$$dW(\Omega)=C\exp\{-E(\Omega,G,H)/kT\}d\Omega. \qquad (140)$$

Numerical calculations of this kind were made for aggregates consisting of four particles in the form of a square.[52] The dependence of the mean magnetic and toroid moments is shown in Fig. 10. For comparison, in the same figure the same dependences are shown for a rigid aggregate with frozen dipoles and for a superparamagnetic aggregate. As can be seen from these data, a particularly strong difference is manifested in the initial section of the curve.

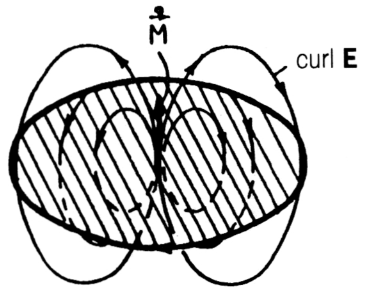


FIG. 11. Force lines of a solenoidal electric field around a vibrating magnetic dipole. The heavy curve shows the winding of the detecting coil.

4.4. Measurement of toroid susceptibility of a suspension

Toroid moments are comparatively new physical quantities, and therefore we consider here the basic arrangement of a measurement of the toroid susceptibility of samples. We first recall that the measurement of a magnetic moment is done as follows. In the measurements, the magnetic moment of a sample is set in motion in some way or other (by means of a "measuring" field, by rotation or vibration of the sample itself, etc.) relative to the measuring coil, in which an emf is induced as a result. The magnetization of the sample can be deduced from the emf. The type of coil employed and the magnitude of the emf can be estimated as follows.

For simplicity, we suppose that the investigated system is a point magnetic dipole **M**. As is well known, a fixed dipole creates around it a magnetic field equal to

$$\mathbf{H}=\frac{3(\mathbf{M}\mathbf{r})\mathbf{r}-\mathbf{M}r^2}{r^5}. \qquad (141)$$

The lines of force of the field are shown in Fig. 11. If the moment **M** varies with time, then so will the field **H**, and in the surrounding space there arises a solenoidal electric field, which in accordance with Maxwell's equation is determined by

$$\mathrm{curl}\,\mathbf{E}=-\frac{1}{c}\dot{\mathbf{H}}=-\frac{3(\dot{\mathbf{M}}\mathbf{r})\mathbf{r}-\dot{\mathbf{M}}r^2}{cr^5}. \qquad (142)$$

Thus, the force lines of the vector $\mathbf{F}=\mathrm{curl}\,\mathbf{E}$ are distributed around the source $\dot{\mathbf{M}}$ in exactly the same way as the lines of the magnetic field around its source $\dot{\mathbf{M}}$ (Fig. 11). The emf induced in the measuring circuit can be found by integrating the field curl **E** over a surface spanned by this circuit. The structure of the force lines shown in Fig. 11 indicates that the circuit must have a position relative to $\dot{\mathbf{M}}$ as is shown in the same figure. For such an arrangement, the flux of the vector curl **E** through the circuit will be maximal. In practice, the detecting coil, which consists of several windings, may have the form of a cylindrical solenoid.

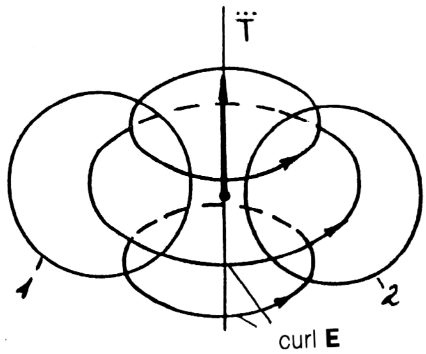


FIG. 12. Force lines of the solenoidal electric field around a vibrating toroid dipole. The windings of the detecting coil are shown by the heavy curve.

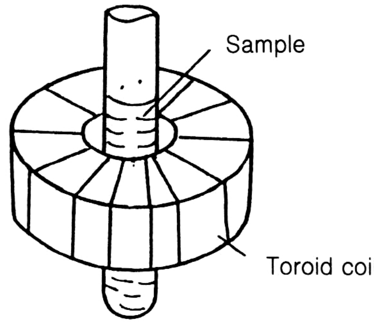


FIG. 13. Arrangement of a sample and detecting coils for measurement of toroid susceptibility.

We consider in the same way a point toroid moment **T**. A toroid at rest does not create around itself either an electric or a magnetic field, but creates only a field of the vector potential:[55]

$$\mathbf{A}=\left.\frac{3(\mathbf{Tr})\mathbf{r}-\mathbf{T}r^2}{r^5}\right|_{r\neq 0}+\frac{8\pi}{3}\mathbf{T}\delta(\mathbf{r}). \tag{143}$$

Now suppose that the moment **T** changes with time. As is well known, a vector potential that changes in time leads to the appearance of an electric field that in accordance with (143) is

$$\mathbf{E}=-\frac{1}{c}\dot{\mathbf{A}}=-\frac{1}{c}\frac{3(\dot{\mathbf{T}}\mathbf{r})\mathbf{r}-\dot{\mathbf{T}}r^2}{r^5}. \tag{144}$$

However, it is readily seen that this is an irrotational field, and therefore it cannot induce an emf in any circuit. If, however, the field **E** itself changes in time, then it can be a source for a solenoidal magnetic field:

$$\text{curl}\,\mathbf{H}=\frac{1}{c}\dot{\mathbf{E}}=-\frac{1}{c^2}\ddot{\mathbf{A}}. \tag{145}$$

Representing the vector potential (143) in the form

$$\mathbf{A}=\text{curl}\,\frac{[\mathbf{Tr}]}{r^3}, \tag{146}$$

we find from (145) that the magnetic field itself is in this case

$$\mathbf{H}=-\frac{1}{c^2}\frac{[\ddot{\mathbf{T}}\mathbf{r}]}{r^3}, \tag{147}$$

while the solenoidal electric field that it induces is determined by

$$\text{curl}\,\mathbf{E}=-\frac{1}{c^3}\frac{[\dddot{\mathbf{T}}\mathbf{r}]}{r^3}. \tag{148}$$

The force lines of this field are shown in Fig. 12. They have the form of circles whose centers lie on an axis that passes through the vector $\ddot{\mathbf{T}}$. It follows from this that the receiving coil must have the form of a toroidal solenoid. Two windings of this coil (*1* and *2*) are shown in Fig. 12.

Note that for excitation of a solenoidal magnetic field on the sample a toroidal coil can also be used (Fig. 13). It is easy to show that when an alternating current is passed through it an alternating electric field is generated in the region of the position of the sample, and as a result of this a solenoidal magnetic field will be generated at the sample, as follows from the Maxwell equation (145).

To estimate the current induced in a conductor placed at the center of a small coil, we can use the approximate expressions

$$\mathbf{A}(0)=\frac{8\pi}{3}\mathbf{T}\delta(\mathbf{r}), \tag{143a}$$

$$\mathbf{E}(0)=-\frac{8\pi}{3c}\dot{\mathbf{T}}\delta(\mathbf{r}),\quad \mathbf{j}=\sigma\mathbf{E}. \tag{144a}$$

5. REMAGNETIZATION OF AGGREGATES BY A SOLENOIDAL FIELD AND TOROID INFORMATION STORAGE

5.1. Particles for magnetic storage

Small magnetic particles are the main units for the storage of information in magnetic storage media. Because of the high anisotropy energy in such particles, there exist two equivalent directions of the magnetization separated by a rather high potential barrier. The height of the barrier is usually characterized by the strength of the remagnetization field, which is called the coercive field H_c. In the storage process, the particle is magnetized in a particular direction and can then remain in this state for a long time, thereby storing information. To increase the density of stored information, it is natural to try to reduce the size of the particles. However, as the detailed analysis made in Ref. 48 shows, the coercive field H_c is greatly reduced when the particles are smaller, and this reduces the stability of magnetic storage. As was shown in Ref. 48, the minimum volume of an iron oxide particle is $V\sim 3\cdot 10^{-17}$ cm^3. At the same time, to ensure sufficient anisotropy energy (with $H_c\sim 900$ Oe), it is necessary to use strongly elongated particles with length-to-diameter ratio 10:1. For these dimensions, the area occupied by a dipole particle on the support is approximately $S_d\sim 10^{-11}$ cm^2, but this does not mean that the same area is required for one bit of

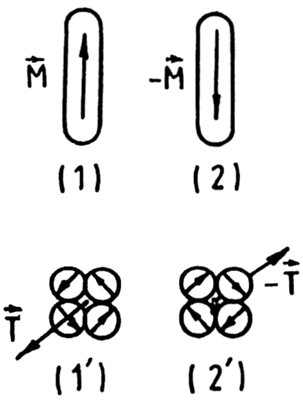


FIG. 14. When the dipole particle is remagnetized from state (1) to state (2), its magnetic moment is reversed. Aggregates of magnetic particles with closed flux do not possess total magnetic moments, but they can have opposite directions of the solenoidal magnetization as in (1′) and (2′). On remagnetization of the aggregate from state (1′) to state (2′) its toroid moment is reversed.

information. The maximum storage density achieved in practice is 1 Gbit per square inch,[49] which corresponds approximately to an area of $S_{db}\sim 10^{-8}$ cm^3 for 1 bit of information. This is very different from the area occupied by one particle ($S_{db}\gg S_d$), the reason for which is that a magnetized particle creates around it a scattering field that distorts the magnetic structure of the medium, and the true storage density is restricted by the region in which this field is essentially nonzero.

In this section, we consider an essentially new possibility for realizing magnetic storage,[50] for which the basic storage units of the medium are aggregates of small magnetic particles that have no total magnetic moment. Such aggregates can be formed during the preparation of the storage medium before polymerization of the carrier matrix, when the magnetic particles are suspended in the carrier fluid. The size and shape of the aggregates can be regulated by choosing the amount of surfactants that are usually added to the suspension to prevent the coalescence of the magnetic particles. Because of the strong dipole–dipole interaction, the aggregates form magnetic structures with closed magnetic flux. Such a structure is characterized by a definite direction in which the magnetic moments of the particles "turn" (Fig. 14) and is described by the magnetic toroid moment[20–22]

$$\mathbf{T}=\tfrac{1}{2}\sum_a [\mathbf{r}_a\mathbf{m}_a],$$

where $\mathbf{r}_a$ and $\mathbf{m}_a$ are the radius vector and dipole moment of one particle within the aggregate. In the presence of a solenoidal magnetic field, the aggregate can reverse the orientation of the toroid moment (Fig. 14), making possible magnetic storage in this case.

We estimate the field H_r needed for the remagnetization of an individual particle within an aggregate. This field will play a role analogous to that of the coercive field for dipole particles and will give an estimate of the length of time for which information can be stored. We shall assume that the aggregate consists of small magnetic particles of iron oxide obtained by nanotechnology. According to the experimental data of Ref. 51, in this case the particles have the shape of weakly prolate spheroids measuring on the average 8×12 nm and a saturation magnetization M_s equal to 300 emu/cm^3 and a coercive field H_c of order 20–60 Oe. Such particles are certainly unsuitable for "dipole" storage, since they have a too low coercive field. The field H_r that we need can be assumed equal to the sum of the fields H_c and H_d, where H_d is the total field exerted on a considered particle by the dipole particles that surround it. Ignoring the small field H_c, we can estimate the strength of this field as

$$H_r\sim H_d\sim Nm/r^3,$$

where N is the number of particles in the aggregate, r is the mean distance between the particles, which for the estimate we take equal to the particle diameter d, and $m=M_sV$ is the dipole moment of the particle, with $V\sim d^3$. Using these estimates, we find, in accordance with (162),

$$H_r\sim NM_s.$$

Taking $N\approx 4$ and $M_s=300$ emu/cm^3, we obtain the estimate $H_r\sim 1200$ Oe. Thus, the particle remagnetization field within an aggregate in this case has a value that is quite sufficient to ensure stability of information storage even when smaller particles than in the case of ordinary storage are used. In addition, remagnetization of the aggregate as a whole requires an inhomogeneous field of a definite (solenoidal) configuration, and this provides additional protection of the stored information from random disturbances.

Aggregates with a closed magnetic flux create a weak scattering field that decreases with distance much more rapidly than the field of dipole particles, and therefore they interact with each other very weakly. As an example, we note that whereas the interaction energy of two dipoles depends on the distance between them as $1/r^3$, the interaction energy of two aggregates in the form of a square decreases as $1/r^7$. Because of this, one can considerably increase the specific density of magnetic storage, the estimate of which is based, as above, on the size of the scattering field. For comparison, Fig. 15a shows a dipole particle measuring 10×100 nm (such particles are used for high-density magnetic storage), while Fig. 15b shows, on the same scale, an aggregate of four single-domain particles having a diameter of about 10 nm. The aggregate and particle occupy approximately the same areas (about 10^3 nm^2), but the regions in which the scattering fields are nonzero differ strongly. Therefore, the area per bit of information, estimated from the area of the scattering field created by the storage unit (enclosed by the broken circles in the figure) is in the first case three orders of magnitude greater than the area of the particle itself, whereas in the second case it is approximately equal to the area of the aggregate. Thus, the use of "toroid" information carriers makes it possible in principle to increase considerably (by two or three orders) the density of magnetic storage and simultaneously raise the reliability of its storage.

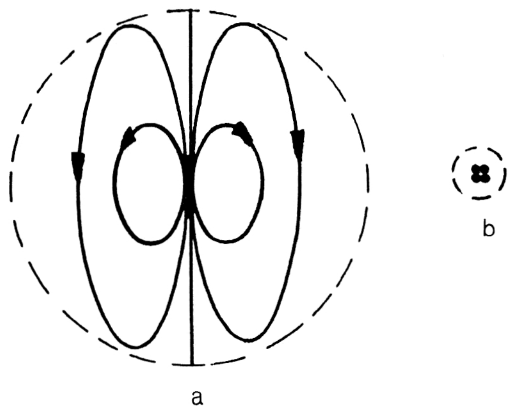


FIG. 15. Relative sizes and scattering fields for an elongated dipole particle (a) and aggregate of single-domain magnetic particles (b). The broken circles show the regions in which the scattering fields of the two particles are nonzero. These regions correspond to the area for 1 bit of information.

However, these advantages can be realized only if there is an appropriate modification of the method of storage and reproducing information. This question is also discussed below. We first consider the problem of the remagnetization of an aggregate of magnetic particles by a solenoidal magnetic field. For this purpose, we use an approximate method based on the introduction of a toroid order parameter. The same problem was solved numerically by means of the recently developed methods of micromagnetics,[42] which were somewhat modified for our purposes. The solenoidal field that remagnetizes an annular aggregate (more precisely, the "coercive curl" G_c) was found as a function of the magnetic properties of the particles and the geometrical shape of the aggregate. Also found was the dependence of G_c on an applied homogeneous magnetic field H, and it was shown that the value of G_c can be appreciably reduced if a homogeneous field is applied to the aggregate during the remagnetization process. It follows from this that for "toroid" storage one can use weak sources of a solenoidal magnetic field.

5.2. Remagnetization of an aggregate by the "fundamental harmonic" of an external field

To analyze the possibilities of remagnetizing an aggregate, we shall describe its magnetic state approximately by two vectors—the magnetic and toroid moments $\boldsymbol{\mu}$ and $\boldsymbol{\tau}$. In this case, the energy of the aggregate in an external field U can be expressed approximately as a sum of terms that take into account all forms of its internal interactions and the interactions of its magnetization and toroid moment with the field: $a_{ik}\mu_i\mu_k$, $b_{ik}\tau_i\mu_k$, $c_{ik}\tau_i\tau_k$, $-\mu_iH_i$, and $-\tau_iG_i$. If we replace the two three-dimensional vectors $\boldsymbol{\tau}$ and $\boldsymbol{\mu}$ by the six-dimensional vector $|\psi\rangle$, which can be assumed normalized to unity, $\langle\psi|\psi\rangle=1$, because the absolute magnitudes of the magnetic moments of the embryons are bounded, then the energy U can be written in the form $U=\langle\psi|\hat{\mathscr{H}}|\psi\rangle+\langle f|\psi\rangle$, where the "Hamilton operator" $\hat{\mathscr{H}}$ can be expressed in a definite manner in terms of the 3-tensors a_{ik}, b_{ik}, and c_{ik}, and the 6-vector $\langle f|$ can be expressed in terms of the components of the field vectors $\mathbf{H}$ and $\mathbf{G}$. We expand the state vector $|\psi\rangle$ with respect to eigenstates $|\psi_n\rangle$ $(n=0,1,\ldots,5)$ of the Hamiltonian $\hat{\mathscr{H}}$:

$$|\psi\rangle=\sum_n C_n|\psi_n\rangle. \tag{149}$$

Because the Hamilton operator $\hat{\mathscr{H}}$ is Hermitian, the basis vectors $|\psi_n\rangle$ form an orthonormal set, $\langle\psi_n|\psi_m\rangle=\delta_{nm}$, and this makes it possible to express the coefficients C_n in (149) in terms of the projection of the state $|\psi\rangle$ onto $|\psi_n\rangle$:

$$C_n=\langle\psi_n|\psi\rangle. \tag{150}$$

Using the expansion (149), we can write the expression $\langle\psi|\hat{\mathscr{H}}|\psi\rangle$ in the form $\Sigma C_n^2E_n$ and the product $\langle f|\psi\rangle$ as Σf_nC_n, where

$$f_n=\langle\psi_n|f\rangle, \tag{151}$$

and the length of the vector $|\psi\rangle$, the value of $\langle\psi|\psi\rangle$, in the form ΣC_n^2. To simplify the expressions, we shall in what follows denote the length of the vector $|\psi\rangle$ by the symbol C, i.e., for example, $\langle\psi|\psi\rangle=C^2$. With these manipulations, the energy of the aggregate can now be expressed in the form[50]

$$U=\sum_n\left(E_n\frac{C_n^2}{C^2}-f_n\frac{C_n}{C}\right). \tag{152}$$

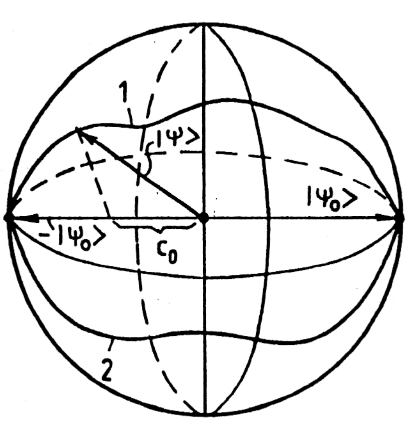
In what follows, we shall call the coefficients f_n determined by the relation (151) the amplitudes of the field "harmonics." This term is justified because f_n determine the contributions of the different "harmonics" $|\psi_n\rangle$ to the expansion of an arbitrary field configuration:

$$|f\rangle=\sum f_n|\psi_n\rangle. \tag{153}$$

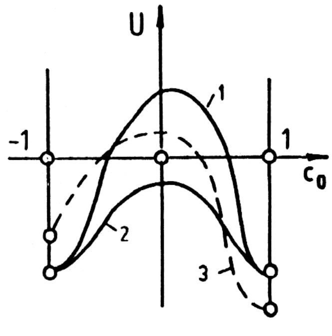
To "switch on" a particular field "harmonic," it is necessary to create the configuration of the fields $\mathbf{H}$ and $\mathbf{G}$ corresponding to it, i.e., to specify definite magnitudes and orientations of these vectors in accordance with the known shape of the aggregate.

We now consider the conditions of remagnetization of the aggregate by applying the fundamental harmonic of the field, i.e., we shall assume that the configuration of the fields $\mathbf{H}$ and $\mathbf{G}$ is chosen in such a way that all the coefficients f_n except f_0 are zero. We assume that the aggregate in the absence of the field was in the state $-|\psi_0\rangle$, i.e., we have the coefficient $C_0=-1$, and all the remaining coefficients C_n with $n\neq0$ were zero. If the external field $|f\rangle=f_0|\psi_0\rangle$ has orientation directly opposite to the initial state of the aggregate, i.e., the coefficient f_0 is positive ($f_0\geqslant0$), then for a definite field amplitude f_0 the aggregate goes over from the state $-|\psi_0\rangle$ to the state $|\psi_0\rangle$. At the same time, the magnetic particles reverse their orientation, i.e., the aggregate is remagnetized.

The states $-|\psi_0\rangle$ and $|\psi_0\rangle$ are two opposite poles of the six-dimensional sphere defined by the equation $\langle\psi|\psi\rangle=1$ in the $|\psi\rangle$ space. We can regard the process of aggregate remagnetization in the external field as displacement of a point on the surface of this sphere from one pole to another along a certain trajectory (Fig. 16a). For each



a



b

FIG. 16. As the end of the state vector $|\psi\rangle$ moves over the surface of the sphere of unit radius in the 6-dimensional space from the point $-|\psi_0\rangle$ to the point $|\psi_0\rangle$ (Fig. 16a, the 6-dimensional sphere is replaced by a three-dimensional one) the potential U and the projection C_0 of the vector $|\psi\rangle$ on the axis $|\psi_0\rangle$ change at the same time. The dependence of U on C_0 in the absence of an external field for two trajectories is shown schematically in Fig. 16b (curves *1* and *2*). When an inhomogeneous magnetic field is applied to the aggregate along the "direction" $|\psi_0\rangle$, the energy at one of the minima in the curve, the one at the point $C_0=-1$, is raised, and at the other it is lowered (curve *3*). The state $-|\psi_0\rangle$ is metastable.

value of the vector $|\psi\rangle$ corresponding to points of the trajectory on the sphere, we can calculate the aggregate energy $U(|\psi\rangle)$,[50] and also find the projection of this vector $|\psi\rangle$ onto the state $|\psi_0\rangle$. Thus, with each trajectory we can associate a certain function $U=U(C_0)$. In Fig. 16b, this function is shown schematically for two different trajectories (curves *1* and *2*) for the case when there is no external field. To the points $C_0=\pm 1$ there corresponds the minimum energy E_0, since by definition the vectors $|\psi_0\rangle$ and $-|\psi_0\rangle$ describe the two ground states of the system. For a field applied along the direction $+|\psi_0\rangle$, the minimum at the point $C_0=-1$ becomes shallower and the minimum at the point $C_0=+1$ becomes deeper (broken curve *3* in Fig. 16b). Thus, if the system before the field was applied was in the state $-|\psi_0\rangle$, corresponding to the global minimum of the potential, then in the presence of a field this state becomes metastable. At a certain (critical) value of the field, the system undergoes a transition (of the first type) to the state $|\psi_0\rangle$. Thus, the system possesses hysteresis behavior, and this makes it possible to use aggregates of magnetic particles for magnetic storage. In general, the critical transition field depends on the path of the transition. We shall now find the minimum value of the critical field, which in the given case will determine the stability of information stored on toroid carriers.

To determine the amplitude of the remagnetization field, we must calculate the first and second derivatives of the potential (152) with respect to the parameters C_n, equate them to zero, and solve the resulting system of equations. From the vanishing of the first derivative

$$\frac{\partial U}{\partial C_n}=0$$

we can find stationary states of the system in the presence of an external field, and from the vanishing of the second derivative

$$\frac{\partial^2 U}{\partial C_n \partial C_m}=0$$

we can find the critical field f_0^*. After calculation of the derivatives, we obtain the relations

$$\left[2(E_n-U)-\left(\sum_m f_m C_m\right)\right]C_n=f_n, \qquad (154)$$

$$\left[2(E_n-U)-\left(\sum_l f_l C_l\right)\right]\delta_{nm}-(f_n C_m+f_m C_n)+\left(\sum_l f_l C_l\right)C_n C_m=0. \qquad (155)$$

Since in the considered case $f_n=f_0\delta_{n0}$, Eq. (154) has solutions $C_0=\pm 1$, $C_n=0$. At the same time, as is readily seen from (154), the states $\pm|\psi_0\rangle$ have energies $E_\pm=E_0\pm f_0$, respectively. Thus, the external field lifts the energy degeneracy between the states $|\psi_0\rangle$ and $-|\psi_0\rangle$.

We now find the value of the field at which the transition from the metastable state $-|\psi_0\rangle$ to the state $|\psi_0\rangle$ occurs. For this, we substitute in the expression (155) $f_n=f_0\delta_{n0}$ $(f_0>0)$, $C_0=-1$, $C_n=0$ $(n\neq 0)$, i.e., we seek the stability of the state $-|\psi_0\rangle$. After simple manipulations, we find that Eq. (154) is satisfied if the following condition holds:

$$f_0^*=2(E_n-E_0)\quad (n\neq 0). \qquad (156)$$

Thus, we have obtained a complete spectrum of critical fields. The minimum value of the critical field at which the transition can occur is

$$f_{0\min}^*=2(E_1-E_0). \qquad (157)$$

This determines the remagnetization field for an aggregate of given shape and given orientation of its anisotropy axes. The field is twice the difference of the eigenvalues of the Hamiltonian $\hat{\mathscr{H}}$ in the ground and first "excited" states. This field plays for aggregates the same role as the coercive field H_c for dipole particles. Therefore, from the magnitude of this field we can judge the stability of magnetic storage with respect to random external disturbances.

5.3. Remagnetization of an aggregate by two field harmonics

We now assume that the external field has a configuration described by two harmonics, i.e., among the com-

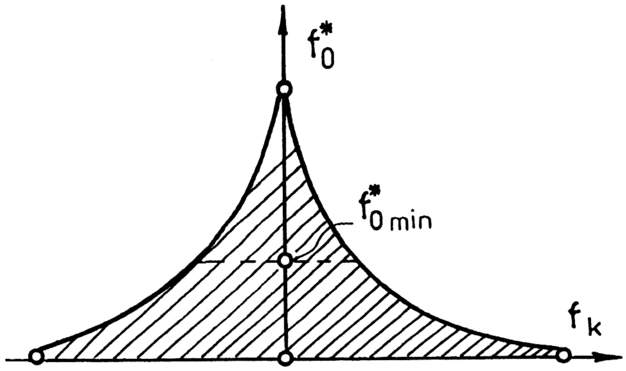


FIG. 17. Dependence of the critical remagnetization field f_0^* of the aggregate on the amplitude of the second (additional) harmonic f_k. The region in which metastable states can exist is hatched. In the upper part (down to the broken horizontal line drawn at the level corresponding to the minimum critical remagnetization field in the absence of the additional harmonic $f_{0\min}^*$), the shape of the curve can change, depending on the trajectory followed by the phase point on the transition from the metastable state to the position of global minimum of the energy.

plete set of coefficients f_n in (153) only f_0 and f_k are nonzero, and all the remaining f_n with $n\neq 0$, $n\neq k$ are zero. It is obvious that we must now seek a solution of Eqs. (154) and (155) on the two-dimensional subspace of the phase space of the system spanned by the vectors $|\psi_0\rangle$ and $|\psi_k\rangle$. Indeed, if the condition

$$[2(E_n-U)-f_0C_0-f_kC_k]\neq 0 \qquad (158)$$

is satisfied, the homogeneous equations (154) for the coefficients C_n with $n\neq 0$, $n\neq k$ have only zero solutions ($C_n=0$). Since, on the other hand, any state vector is normalized, the coefficients C_0 and C_k are related by the condition $C_0^2+C_k^2=1$. If we introduce a parameter θ such that $C_0=\cos\theta$, $C_k=\sin\theta$, then the normalization condition will be satisfied automatically.

To calculate the critical field in the given case, it is convenient to proceed directly from the expression (152) for the energy. Retaining in the sum over n only two terms, with $n=0$ and $n=k$, and expressing the coefficients C_0 and C_k in terms of θ, we obtain

$$U=E_0+(E_k-E_0)\sin^2\theta-f_0\cos\theta-f_k\sin\theta. \qquad (159)$$

It is readily seen that this expression is formally identical to the energy of a uniaxial ferromagnet in a magnetic field with E_k-E_0 playing the role of the anisotropy energy and f_0 and f_k as the components of the external field along the x and z axes (see Sec. 41 in Ref. 53). It is important to note that our case corresponds to anisotropy of easy-axis type, since E_k-E_0, which plays the role of an anisotropy constant, is positive. Indeed, since E_0 is the ground-state energy, the condition $E_0<E_k$ is satisfied.

In Ref. 53, an equation was obtained for the neutral curve, which in the f_0, f_k plane separates the region of possible metastable states. In our notation, the equation of this curve has the form

$$f_0^{2/3}+f_k^{2/3}=[2(E_k-E_0)]^{2/3}. \qquad (160)$$

In Fig. 17, the neutral curve is shown only for positive values of f_0 (in the negative region, it has the same form). Metastable states are possible in the hatched region of magnetic fields.

In what follows, we shall interpret this curve as the dependence of the remagnetization field f_0^* on the amplitude of the "additional" field f_k. In accordance with the results that we have obtained, we can say that when the additional field f_k is imposed the remagnetization field f_0^* of an aggregate that was initially in the state $-|\psi_0\rangle$ is lowered in accordance with the curve in Fig. 17.

We must here make a remark concerning the values of the remagnetization field f_0^* for $f_k=0$. It follows from the expression (160) that this field is equal to $f_0^*|_{f_k=0} = 2(E_k - E_0)$. However, in the previous section it was shown that in the case of excitation of a transition by the fundamental harmonic of the field the critical field depends on the trajectory of the transition and that there exists an entire spectrum of critical fields. If it is assumed that the remagnetization occurs for the minimum field value $f_{0\min}^*$, which is determined by (157), then the curve in Fig. 17 will have in its upper part a "plateau," as shown in the same figure by the broken line. In the general case, it can only be asserted that in its upper part the critical curve lies in the region bounded by the curvilinear triangle between the apex and the plateau.

Our result has important practical significance. Indeed, for the remagnetization of aggregates by a solenoidal magnetic field in the absence of a "magnetizing" homogeneous field one would require a source that generates an inhomogeneous (solenoidal) field of very great strength. As was shown in Sec. 5.1, the remagnetization field H_r of one particle within an aggregate is in order of magnitude 10^3 Oe. For an aggregate having diameter $d\sim 10^{-6}$ cm, the critical G_c, which is estimated at H_r/d, has a very large value: $G_c\sim 10^9$ Oe/cm. However, if at the time of recording a homogeneous magnetic field is applied, then, as was shown above, the critical G_c can be appreciably reduced down to reasonable values, namely, $G_c\sim 10$–10^2 Oe/cm.

Note also that the qualitative result—the lowering of the critical field f_0^* when an additional (not fundamental) harmonic is applied—also remains valid if the aggregate is remagnetized by some more complicated combination of harmonics that includes not just one but several additional fields f_k. This result is also important from the practical point of view. The point is that on a support there may exist aggregates of different shapes, and therefore a given configuration of external fields **H** and **G** will be "felt" by each of them in the form of a corresponding combination of harmonics (we recall that the harmonics are determined by the shape of the aggregate). Nevertheless, one can always choose external fields in such a way that the remagnetization of the aggregates will occur in a desired regime.

The recently developed methods of micromagnetics (see, for example, Ref. 42) also make it possible to model the process of remagnetization of an aggregate numerically.[50] Under the assumption that the ground state is known, one can turn to the solution of the problem of aggregate remagnetization by an external magnetic field. In the general case, this problem is solved as follows. Having

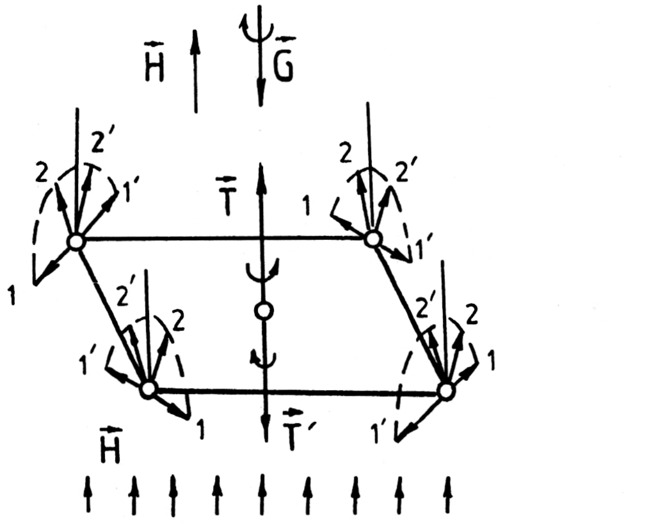


FIG. 18. Toroid remagnetization of an aggregate of magnetic particles in the shape of a square. In the ground state, the magnetic moments of the particles have the orientation indicated by the number *1*, and the aggregate as a whole possesses a toroid moment T_0. In the remagnetized state, the moments of the particles are oriented in the direction *1′*, and the toroid moment T_0' is in exactly the opposite direction to the initial one. To remagnetize the aggregate, it is necessary to apply a solenoidal field with curl **G** having opposite orientation to that of the initial toroid moment and magnitude greater than the critical value G_c. When a homogeneous field **H** is applied, the particles will have the orientation indicated by the number *2*, and in the remagnetized state the orientation *2′*. To remagnetize the aggregate from state *2* to state *2′* one needs a much weaker solenoidal field than for the transition from state *1* to *1′*.

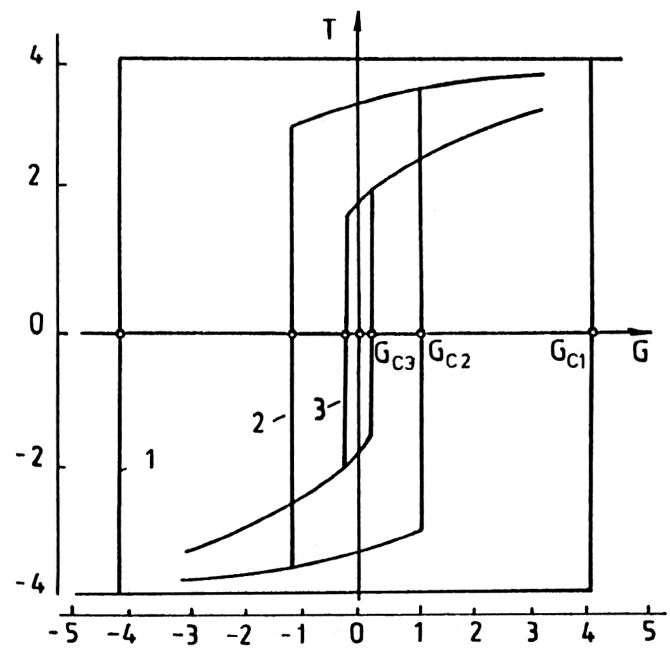


FIG. 19. Hysteresis curves (*1–3*) of toroid remagnetization of an aggregate of magnetic particles in the shape of a square (see Fig. 18), obtained by numerical modeling. Curves *1–3* were obtained for the field values $H_1=0$, $H_2=3$, and $H_3=5$, respectively. In the absence of a field in the ground state, the toroid moment of the aggregate can have two values corresponding to opposite directions. Under the influence of the curl **G** having opposite direction to the initial orientation of the toroid moment **T** (Fig. 18) the aggregate is remagnetized when the curl reaches the critical value G_c.

specified a certain direction of the magnetic field and the curl of the field, we gradually increase these fields with a certain step, which specifies the accuracy of determination of the remagnetization field. Knowing the initial state (which we always take to be the ground state), we can find the effective field that acts on each particle, determine the vector increment $\Delta \mathbf{m}_a$, and repeat this procedure until we find the position of the minimum of the potential U that corresponds to the distribution of the magnetic moments of the particles in the presence of the external magnetic field. Simultaneously, we can calculate the magnetic and toroid moments of the aggregate. Remagnetization occurs when these moments change their initial direction abruptly.

As a specific example, there was considered in Ref. 50 the problem of remagnetization of an aggregate having the shape of a square under the influence of a solenoidal magnetic field. In the ground state, this aggregate has the configuration shown in Fig. 18. This configuration is described by a toroid moment **T** directed perpendicular to the plane of the aggregate. If the curl **G** of the magnetic field is in the direction opposite to the initial direction of the toroid moment, then when it is gradually increased to the value G_c the toroid moment abruptly reverses its orientation, i.e., becomes parallel to the vector **G** (Fig. 18). The magnetization curves of this aggregate are shown in Fig. 19. As follows from the general arguments given in Sec. 2, in this case hysteresis is observed.

If a homogeneous field **H** is applied in the direction parallel to the vector **G**, the value of the critical field is reduced. The curves of toroid remagnetization in the presence of an external field are shown in Fig. 19, and the dependence $G_c=G_c(H)$ is shown in Fig. 20. The decrease of the critical field G_c when a homogeneous magnetic field is applied can be understood on the basis of the following transparent picture. Under the influence of the homogeneous field, the moments of the particles are rotated from position *1* to position *2* in the direction of the field **H** (Fig. 18). In this position, the solenoidal remagnetization of the

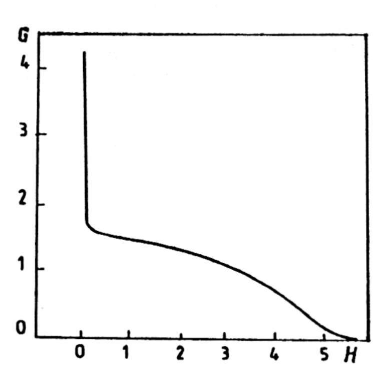


FIG. 20. Dependence of the critical field curl G_c on the strength of the homogeneous field **H** parallel to **G** for the aggregate in the shape of a square (Fig. 18), found by numerical modeling. The curve is in qualitative agreement with the curve shown in Fig. 17.

particles is made easier, since it reduces to rotation of the moments through a small angle around axes that lie in the plane of the aggregate.

It should be noted that the overall dependence of the critical field G_c on H corresponds to the dependence that was obtained by means of the approximate treatment (cf. the "theoretical" curve in Fig. 17 with the "experimental" curve in Fig. 20). Thus, the numerical calculations confirm the general conclusions drawn in the previous section.

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