

## Structural Organization in the System of Magnetized Aggregates in Elliptically Polarized Rotating Magnetic Field

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(Received 19 May 2014; published online 15 July 2014)

Observed data supporting the possibility of magnetic ordering in magnetic colloids are reported. A possibility of aggregate spatial ordering in a system of magnetized aggregates was found under the influence of rotating and crossed constant and alternating magnetic fields.

**Keywords:** Magnetic colloid, Magnetic ordering, Rotating magnetic field, Structural ordering.

PACS numbers: 47.57.J, 75.50.Mm, 82.70.Dd

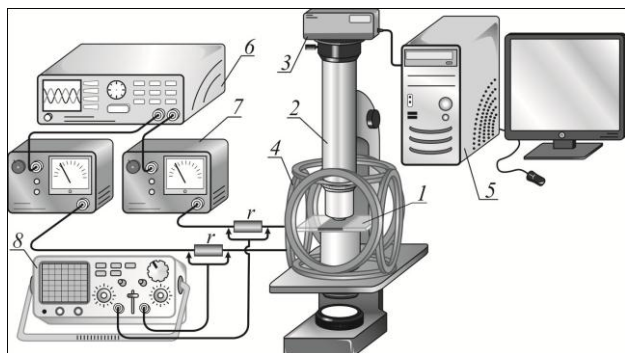
### 1. INTRODUCTION

Magnetic colloid nanosystems (magnetic fluids) remain a field of interest attracting the attention of researchers, which is due not only to the possibilities of their practical applications, but also to the emergence of a range of physical problems such as the possibility formation of magnetic orderings within them.

Previously [1-3] we have also identified a well-developed system of magnetized aggregates in kerosene-based magnetic liquids in certain conditions and carried out investigations of magnetic properties of magnetic colloids, containing such aggregates. This paper presents the results obtained by investigating structures, emerging within the magnetic colloid with the magnetized aggregates under the action of elliptically polarized rotating magnetic field.

### 2. EXPERIMENTAL METHOD AND EQUIPMENT. SAMPLES

Structural aggregate formations in magnetic fluids were studied via observations using an optical microscope equipped with three pairs of Helmholtz coils to induce a magnetic field. In that case, the design of the magnetizing system made it possible to induce as a direct as an alternating and a rotating magnetic field in vertical and horizontal plane (Figure 1).



**Fig. 1** – Schematic presentation of a setup for optical observations: 1 – sample of magnetic colloid; 2 – microscope; 3 – digital camera; 4 – Helmholtz coils; 5 – PC; 6 – generator; 7 – amplifier; 8 – oscillograph

A thin layer with the examined sample was placed horizontally. Elliptically polarized rotating magnetic field was created in vertical plane, perpendicularly to the layer containing sample of the magnetic colloid. Effect of elliptical form of polarization on the behavior of magnetized aggregates was investigated. The resulting magnetic field consists of the perpendicular  $H_{\perp}$  and the longitudinal  $H_{\parallel}$  to the constituting layer. In the experiments the relation of their peak value varied in the range from  $\infty$  (linear polarization perpendicular to the layer) to 0 (linear polarization along the layer) as follows: 5, 4, 3, 2, 1, 1/2, 1/3, 1/4, 1/5. The maximal peak values of each component of the fields was equal to 3.2 kA/m.

Additionally, to gain information about peculiarities of the aggregate motion, investigations on light scattering by thin layers of magnetic fluids containing such aggregates were performed. A laser beam (wavelength 630 nm) going perpendicularly to a MF layer was used as a light source.

### 3. RESULTS AND DISCUSSION

The conditions of aggregation in magnetic colloids are determined by the ratio of forces of repulsion and attraction of colloidal particles. The most widespread kerosene-based colloids are stabilized by the repulsion of sorption-solvate shells formed by long-chained molecules of oleic acid. The attraction of the particles is determined by magneto-dipole forces and van der Waals forces. If the latter are of isotropic nature, the effectiveness of the magneto-dipole interaction depends on the mutual location of the particle moments. Herewith, the location is characterized as by the forces directly as by their rotary moments. The direction of the dipole moment of a single-domain particle can be changed due to thermal fluctuations of the magnetic moment direction inside the particle or by particle rotation due to rotational diffusion. Obviously, the domination of one or the other of the above situations must affect the processes of structure formation, however, with theoretical justification for the aggregation processes in magnetic colloids, it is usually ignored.

Real magnetic colloids are polydisperse, but they might contain as small particles, which are characterized by the Neel mechanism of relaxation, as larger particles,

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the magnetic moment of which can be assumed to be rigidly connected to the solid matrix at room temperatures.

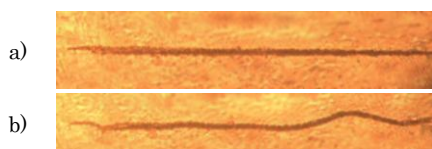
Presently, with the intention to produce stable magnetic colloids, technologies have been developed providing synthesis of such systems with particles of a limiting small size (less than 8-9 nm), in which the magnetic moment direction changes due to thermal fluctuations. Most of the colloids with larger particles (of the mean diameter more than 12-15 nm) may be rigid magnetic dipoles. With reference to the results of our studies [2], aggregates, which maintain the magnetic moment, can arise in such colloids under certain conditions. Such aggregates, as a rule, exhibit a well-pronounced shape anisotropy, respond quickly enough to the switch on of an external magnetic field and, in its absence, are aligned along the force lines of the earth magnetic field. In some case, these aggregates demonstrate a filament shape, and, in the absence of magnetic field, the minimum of magnetic energy is obtained by branching and weaving of the aggregates.

The emergence of magnetized aggregates is possible at long-term (up to several dozen years) storage of magnetic colloids with a relatively large mean particle size, when the Brownian mechanism of magnetic relaxation is expected. Furthermore, such aggregates can be produced by artificial violation of the aggregative stability of such, originally homogeneous, colloids, when coagulants, for example, an excess of oleic acid, are added.

Note that a typical feature of the magnetized aggregates is that, being exposed to the external magnetic field, they trigger the formation of long enough filamentary structures. Previously it was reported in [3], that in some cases, they may be flexible that leads to interesting features of their motion when the direction of the field changes. As an example, Fig. 2 illustrates the behaviour (motion) of a worm-like magnetized aggregate caused by the change of the external magnetic field direction: (a) the field is directed from right to left along the figure plane; (b)-(e) the position of the aggregate in equal time intervals (about 1 s) after the field changes its direction to opposite.

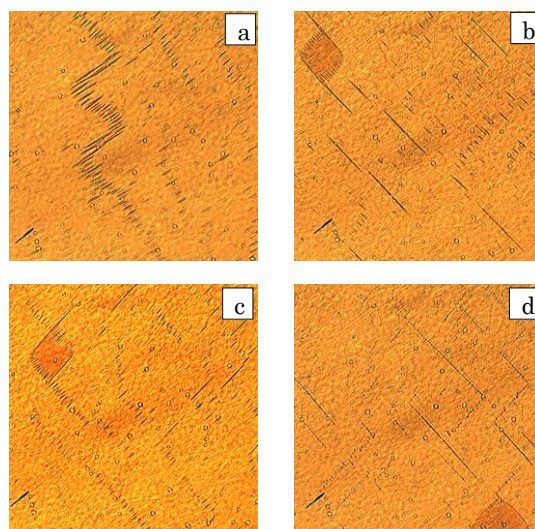
As one can see from the figure, when the direction of the magnetic field changes, such aggregate behaves like a liquid magnetized object. The behavior of the flexible ferromagnetic aggregate at field reversal quite similar to that is considered in [4].

It has turned out that under the concurrent action of crossed steady and alternating magnetic fields, a system of mutually perpendicular filament structures is formed, which periodically split up in a number of tiny fluctuating needle-like aggregates. Fig. 3 illustrates a gradual transformation of a structure formed in a steady magnetic field ( $H = 2$  kA/m) when the frequency of the alternating magnetic field, additionally imposed perpendicularly to the direct field one, increases from 5 Hz to 30 Hz.



**Fig. 2** – 180° rotation of the magnetized aggregate when the external magnetic field changes its direction to opposite

Note, that structures similar to those shown in Fig. 3, have been observed earlier in the paper [5] for the system of spherical paramagnetic micro-particles, weighted in water. A cell containing such particles was under the action of mutually perpendicular direct and alternating magnetic fields, acting along the layer with sample.



**Fig. 3** – Gradual transformation of a structure formed in the steady magnetic field ( $H = 2$  kA/m) when the frequency of the additionally imposed alternating field increases: (a) 5 Hz, (b) 10 Hz, (c) 15 Hz, (d) 30 Hz.

We have identified, that the action on the sample of magnetic colloid with a well-developed magnetized aggregates rotating magnetic field leads to an emergence of principally different structures. In such case the characteristics of such structures depends on the frequency of rotating magnetic field, degree of its polarization and the orientation of ellipsis of polarization relative to the layer MFs. Figure 4 shows structures, emerging in magnetic colloid in a rotating magnetic field of varying frequency and under varying relation of perpendicular  $H_{\perp}$  and the longitudinal  $H_{\parallel}$  relative to the layer of field components. Fig. 5 shows the diffraction pictures corresponding to the obtained structures. As seen in the picture, under the lower frequencies of the field and the presence of only aggregates line up along the field perpendicularly to the layer, while the larger ones, emerged under the alignment of small, form complex winding structures. Increase in frequency of alternating field in this case leads to a decrease in the typi-

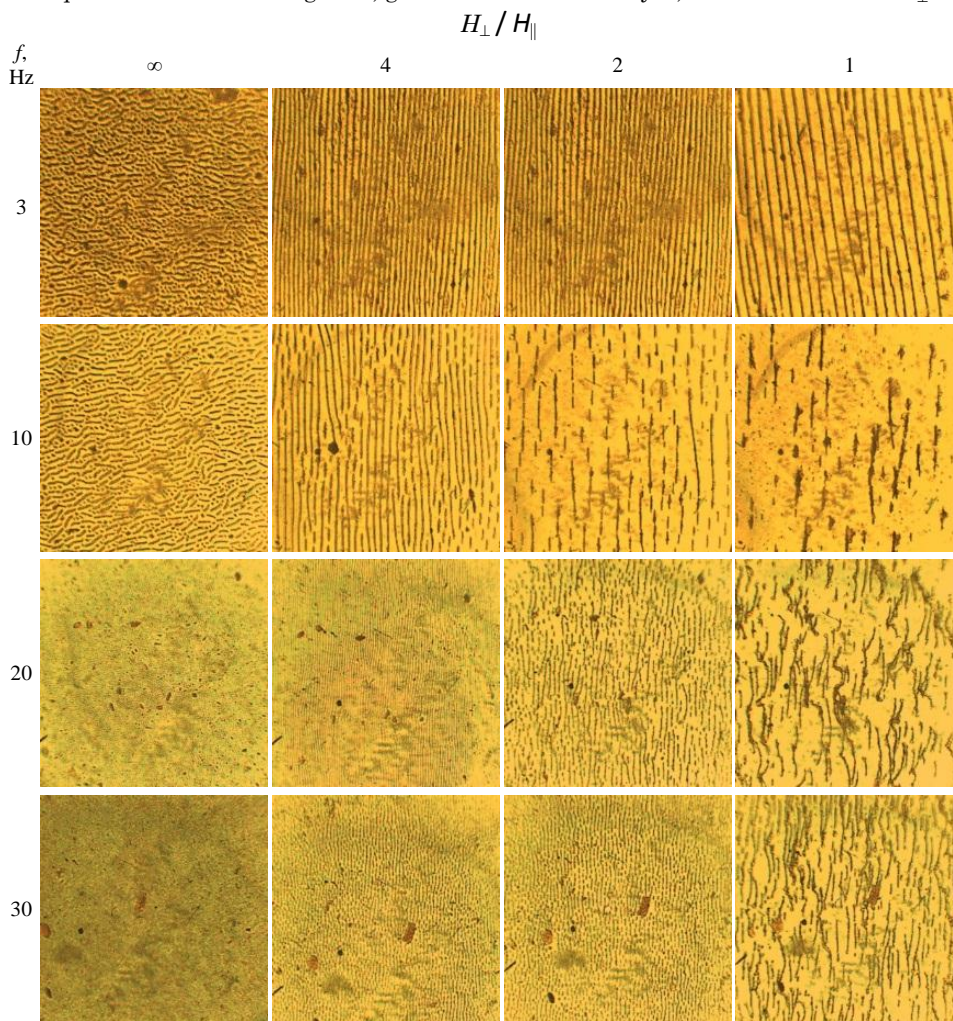
cal size of such structures. Under the frequencies higher than 10 Hz aggregates stop aligning, yet remaining distinct. Further increase in the frequency caused aggregates to breakdown into smaller and directed in the direction of the field, which is evident in the following diffraction picture. With an increase in the frequency of linearly polarized magnetic field the diffraction ring becomes more pronounced, furthermore, an expansion of its diameter also takes place (see Fig. 5).

Emergence of horizontal component of the field  $H_{\parallel}$  leads to a dramatic structural transformation. In this case, the aggregates rotating perpendicularly to the layer, form long filaments, along which may migrate larger formations. (Fig. 4 –  $H_{\perp} / H_{\parallel} = 4, 2$ ) Sufficient regularity of the emerged structure allows the diffraction picture to be observed while sending a beam of laser light through it. It is also worth noting, that an increase in frequency of the field also causes the long filaments to breakdown into separate elongated ensembles of magnetized aggregates, which remain orientated along the direction of the longitudinal component of the field. Similar picture is observed if the amplitudes of the longitudinal and perpendicular components of the field are equal for all frequencies of the rotating field, greater

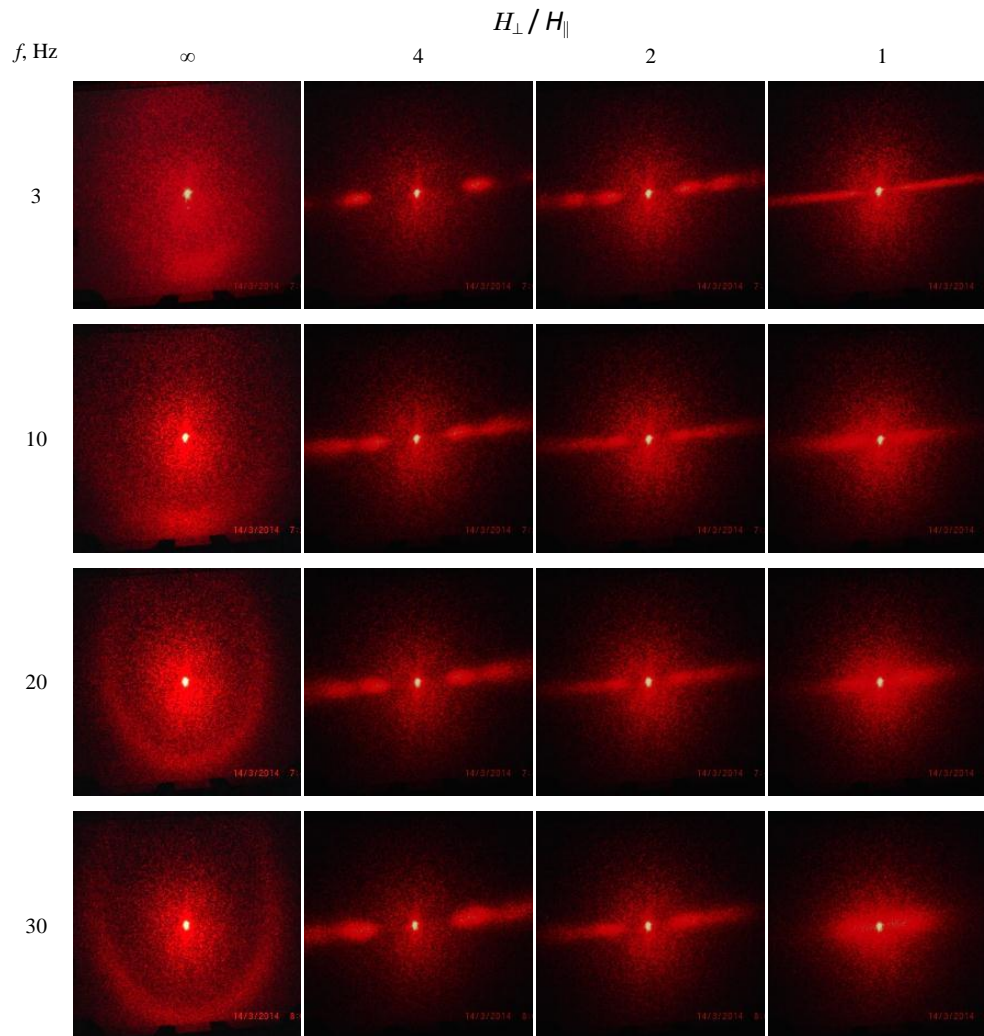
than 5 Hz. At the frequencies less than 5 Hz long filaments of aggregates don't breakdown, however the previously observed transition of long formations along them, ceases. Further increase in the amplitude of the longitudinal component of the field (at  $H_{\perp} / H_{\parallel} = 1/2, 1/4, 0$ ) leads to formation of threads, stretched along the field, which emerge due to the alignment of separate aggregates and their ensembles, as well as due to the fusion of adjacent threads.

It is worth noting, that previously structural organization in rotating magnetic fields was studied in suspensions of soft-magnetic spherical particles [6, 7]. In [6] the modeling was carried out using the method of molecular dynamic of structural transformations, occurring the thin layer of such suspension in elliptically polarized magnetic field. In [7] was made a comparison of results obtained in experimental and quantitative investigations of structural formations in such systems.

However, the investigations we have carried out, have shown, that for a system of aggregates, whose magnetic moment does not equal zero, the most ordered structures emerge in elliptically polarized magnetic field within the orientation of ellipsis being perpendicular to the layer, i.e. for cases when  $H_{\perp} / H_{\parallel} > 1$ .



**Fig. 4** – Structures, emerging in system of magnetized aggregates under the action of rotating in the vertical plane magnetic field of varying frequencies and varying relation of vertical and horizontal components of the field



**Fig. 5** – Diffraction pictures obtained under the action of rotating in the vertical plane of magnetic field of varying frequency and under varying relation between the vertical and horizontal components of the field

#### 4. CONCLUSION

The conducted studies show, that under the action of external magnetic fields of varying configuration on a magnetic colloid with a well-developed system of magnetized aggregates, the processes of structural organization have been observed. The emerging structural ordering in this case depends on the frequency and orientation of ellipsis of polarization of the rotating magnetic field, the value of additionally applied, perpendicular to the rotating, constant magnetic field and the volume concentration of magnetized aggregates. In

this way, magnetic colloid with magnetized aggregates can be seen as a molecular environment for modelling and design of new composite materials, the structure and properties of which can be manipulated using external fields.

#### ACKNOWLEDGEMENTS

This work was supported by The Ministry of Education and Science of the Russian Federation and Russian Foundation for Basic Research (RFBR) (project No.14-03-00312).

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