Investigation of magnetic emulsions in magnetic field by rotating test-tube method

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Abstract. The effect of diffraction scattering of light in a magnetic emulsion in a magnetic field in a rotating test-tube has been investigated. The non-linear dependence of the angle of deviation of the diffraction band on the rotation speed is established. An interpretation of the effect is proposed based on the balance of torques from the magnetic field and the drag force of the fluid. Agreement with the experiment is possible on the assumption of the destruction of the chains of magnetic emulsion microdroplets during rotation.

1. Introduction
Among the new functional nanomaterials, products based on magnetic fluids of composite nanocolloids are of considerable interest. Such media are complex dispersed systems in which there is a fraction of single-domain magnetic nanoparticles and larger ones, some microns in size, with magnetic or nonmagnetic included forms. A special class of composite nanomaterials are magnetic or magnetodielectric emulsions. Magnetic emulsions are a liquid medium in which microdroplets of a magnetic fluid of micron or submicron size are suspended. Drops of magnetic emulsions actively interact with an external magnetic field, which leads to a significant anisotropy of the magnetic, rheological and optical properties of such systems. The dipole-dipole interaction of droplets in magnetic emulsions leads to the formation of complex aggregative structures of various types [1,2]. In magnetic emulsions with high interfacial tension (water-oil type) in a magnetic field, emulsion droplets can line up in chains, while maintaining a spherical shape [1]. When exposed to a chain of external forces, it can turn or collapse. The presence of chain aggregates in the emulsion significantly changes its optical properties [3].

2. Experiments
We investigated an emulsion of droplets of magnetic fluid based on kerosene suspended in water. The concentration of magnetite in the magnetic fluid was about 10%. For the synthesis of stable magnetic emulsion, an ultrasonic method was used with a small amount of surfactant in water to improve aggregative stability. The concentration of droplets in water, after magnetic separation, was less than 1%, and their average size was 100 ÷ 200 nm with a small number of large micron droplets (less than 5% by volume concentration). In the experiment, the sample was placed in a test-tube located in the uniform field region of a Helmholtz coils (figure 2). The test-tube was rotated using an electromotor with a PWM regulator (rotation speed from 20 to 200 rpm (min−1)). The light of a helium-neon laser with a wavelength of 633 nm was passed through the cell and was scattered. The light scattering
pattern on the screen was recorded by the camera. Scattered light image analysis was performed using the ImageJ application [4].

![Figure 1](image1.png)

**Figure 1.** The photo of chain aggregates of microdroplets of a magnetic emulsion in a magnetic field.

When exposed to a magnetic field in a magnetic emulsion, the effect of diffraction scattering of light is observed [5]. This effect represents a bright narrow band of scattered light perpendicular to the direction of the field (figure 3a). The emulsion was also investigated using the rotating test-tube method, which is used to study magnetic fluids [6]. When the test-tube is rotated in a magnetic field, a deviation of the diffraction scattering band of light from the initially vertical position is observed (figure 3b). The dependencies of the deviation angle of the diffraction band on the rotation speed and field strength are shown in figure 4 and figure 5.

![Figure 2](image2.png)

**Figure 2.** The scheme of the experimental setup for the study of diffraction scattering by rotating test-tube method.
**Figure 3.** Diffraction light scattering in magnetic field: a – without rotation of the test-tube; b – when the test-tube is rotate.

**Figure 4.** Dependencies of the deviation angle of the diffraction band on the rotation speed of the test-tube at constant values of magnetic field strength.
3. Results and Discussion

To describe the dependence of the deviation angle of the diffraction pattern on the rotation speed, we consider the following model. Let us imagine a magnetic emulsion as a system identical spherical droplets of radius $R$ randomly arranged in a fluid with viscosity $\eta$. Magnetic fluid droplets have a magnetic susceptibility. In the absence of external influences, such a system can be considered as isotropic. Under the action of a uniform magnetic field $H$, emulsion droplets form chains oriented along the field direction. Suppose all chains are the same size and consist of $n$ separate droplets. The emulsions studied by us have a high interfacial tension. Therefore, the deformation of individual drops under the action of the field can be neglected. To calculate the energy of the chain in a field, we will consider the chain as a uniformly magnetized long ellipsoid (analysis of photographs (see figure 1) of large droplets of the emulsion, it is clear that small droplets tend to take place in the spaces between large ones, thereby filling the space better). We will also assume that the weak field condition is satisfied and the magnetic moment of the droplet chain is proportional to the strength of the external field. In this case the magnetic energy of the chain is defined as:

$$ W = -\frac{\Delta \chi V}{2} H^2 \cos^2 \theta, $$

where $H$ is magnetic field strength, $\Delta \chi = \chi_\parallel - \chi_\perp = \frac{4\pi \chi^2 (N_\perp - N_\parallel)}{(1 + 4\pi N_\parallel \chi)(1 + 4\pi N_\perp \chi)}$ – anisotropy of magnetic susceptibility and $N_\perp$ и $N_\parallel$ - demagnetizing factors across and along the axis of the ellipsoid, respectively. Chain torque under the action of external field can be written as:

$$ \frac{\partial W}{\partial \theta} = \frac{\Delta \chi V}{2} H^2 \sin 2\theta. $$

This torque is balanced by the torque of drag force of the fluid $L\omega$, which occurs when a fluid flows around a chain rotating with an angular velocity $\omega$. $L$ is the drag coefficient for rotating chain. For the long ellipsoid with length $2nR$ and the ratio of the long and short axes equal to $n$, the rotational friction coefficient in a viscous liquid is [7]:

![Figure 5. Dependencies of the deviation angle of the diffraction band on the field strength at constant rotation speed of the test-tube.](image)
where \( D_r \) – rotational diffusion coefficient for long ellipsoid. Equating these torques, we get:

\[
\frac{\Delta \chi V}{2} H^2 \sin 2\theta = \frac{8\pi \eta (nR)^3 \omega}{3(\ln n + 0.57)} .
\]

(4)

The chain volume is defined as the sum of the volumes of the droplets \( V = \frac{4}{3} \pi R^3 \). Thus we get the expression for the dependence of the deviation angle of the chain from the direction of the field:

\[
\theta = \frac{1}{2} \arcsin \left( \frac{4\pi n^2 \omega}{\Delta \chi H^2 (\ln n + 0.57)} \right) .
\]

(5)

From formula (5) follows an approximately linear relationship between the deviation angle of the chain and the angular velocity of rotation. However, as can be seen from figure 4 this dependence is non-linear. The explanation of this fact can be the consideration of breaking the chains with increasing speed of rotation of the cuvette. This reduces the size of the chains and decreases the angle of inclination of the angle of rotation from the field. If we take into account the possibility of gradual destruction of the chains with an increase in the rotation speed of the cell the expression (5) will qualitatively agree with the experimental data (figure 6). This leads to the fact that the dependence of the angle of deviation on speed smoothly changes the angle of inclination (in figure 4 there are no pronounced kinks). At high rotation speed, the number of droplets in the chains stabilizes and the angle of deviation becomes constant. It was shown in [8, 9] that a break in the chains of magnetic particles is preceded by a loss of straightness of the chain and bending of its ends. In our experiments, this should lead to a broadening of the diffraction scattering band due to bending of the ends of the chains. However, we were not able to detect such an effect experimentally.

![Figure 6](image_url)

**Figure 6.** Experimental and theoretical dependences of the angle of deviation of a chain of droplets on the speed of rotation of the test-tube.

4. Conclusion

Magnetic emulsion droplets form chains when exposed to a magnetic field. The irregular structure of the chains causes diffraction scattering of light. Such scattering has the form of a bright diffraction
band, which perpendicular to the magnetic field direction without pronounced maxima and minima of intensity.

The study of diffraction scattering of light in magnetic emulsions has shown that during rotation of the test-tube, the diffraction band rotates through a certain angle. The nonlinear dependence of the angle of deviation and the rotation speed of the test-tube is established. The agreement between the theoretical model and the experiment turns out to be satisfactory under the assumption of the gradual destruction of the chains of emulsion droplets as the angular velocity of the test-tube increases. The interpretation of experimental results for arbitrary values of the field strength requires taking into account the features of magnetization of magnetic emulsions in both the monodisperse [10] and polydisperse approximations [11]. The nonlinear dependence of the magnetic susceptibility of the emulsion droplets on the field strength will also play a significant role in the optical effects in magnetic emulsions in strong fields.

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