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To the theory of Mechano-Magnetic effects in ferrogels.

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Abstract

The paper deals with theoretical study of effect of ferrogels uniaxial elongation on magnetic susceptibilities of these composite materials. We have considered the systems with magnetically soft ellipsoidal and spherical particles. The results show that elongation of the composites with the ellipsoidal particles enhances the susceptibility in the direction of the elongation, whereas the deformation of ferrogels with the spherical particles decreases the susceptibility when the particles concentration is small enough and increases it when the concentration exceeds some threshold magnitude.

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1. Introduction.

Magnetic gels and elastomers present new kind of composite materials, consisting of fine (nano- or micronsized) magnetic particles embedded into polymer matrix. Combination of rich set of physical and mechanical properties of polymer and magnetic materials attracts considerable interest to these systems and to their usage in various industrial and bio-medical applications [1-7]. In part, magnetic gels are used for address drug delivery; for industrial and biological sensors [8-14]; for construction of soft actuators and artificial muscles [2,15]; for cancer therapy, regenerative medicine and tissue engineering [16-24].

From the viewpoint of biomedical applications, magnetic hydrogels are very promising materials due to their biocompatibility and ability to mimic some cellular functions [25]. One of the remarkable properties of these systems is an opportunity to change, under the action of an external magnetic field, their microstructure, magnetic, mechanical and other macroscopic properties, size and shape. This gives a possibility to control, with the help of the field, mechanic behavior, transport and electrical processes in these systems. In its turn, this ability presents significant advantage for biosensoric and other high-tech applications [8,9,20,23,25-27].

On the other hand, one can expect the inverse effect of macroscopic deformation of the composites on their magnetic properties. This effect is interesting from the viewpoint of development of technologies of artificial muscles and actuators, sensors, magnetocontrolled scaffolds for growth and engineering of biological tissues. It should be noted that the similar effect of inverse relationship between gel longitudinal deformation and its electrical potential has been recently discovered and studied in refs. [28,29].

In this work we present results of theoretical study of effect of a ferrogel uniaxial elongation on its magnetic susceptibility. The systems with magnetically soft ellipsoidal and spherical particles are considered. In order to avoid intuitive and heuristic theoretical construction, we consider the systems with small concentrations of the particles. This allows us to develop mathematically regular approaches, which can be used as a background for theoretical study of more concentrated materials.

We believe that the discussed mechanomagnetic effect have a high potential for the development of technologies in the area of artificial muscles, actuators, sensors, magnetocontrolled scaffolds and other high-tech applications of magnetic gel.

2. Ellipsoidal magnetically soft particles.

Let us consider a ferrogel sample, consisting of non Brownian magnetic ellipsoidal particles chaotically (gas-like) distributed in a polymer matrix. Physically this means that the gel was curried without action of an external field. To avoid problems connected with demagnetizing field, we suppose that the sample is highly elongated, magnetic field **H** is aligned along its major axis; the elongation takes place in the same direction. This situation is illustrated in Fig.1.

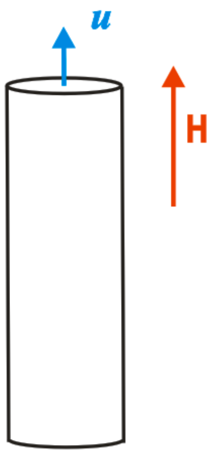

Fig.1. Illustration of the sample; *u* is the mean (measurable) displacement.

Our goal is to estimate effect of the small elongation on the initial magnetic susceptibility χ of the gel. The generalization to the non linear magnetization of the particles is not difficult, but leads to cumbersome calculations and final results. We suppose the strong coupling of the particles with the polymer matrix and the non slipping condition on the particles surface.

The sketch of the particle is presented in Fig.2.

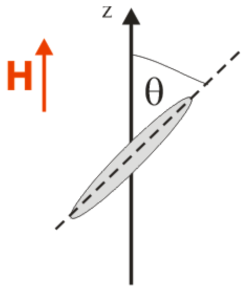

Fig.2. Sketch of the ellipsoidal particle deviated from the field **H**.

By definition, the macroscopic magnetization M_c of the composite, containing the particles, can be presented as:

$$M_c = \varphi < M_{in,z} > \qquad (1)$$

Here φ is volume concentration of the particles, $\boldsymbol{M}_{in}$ is magnetization inside the particle, $M_{in,z}$ is component of $\boldsymbol{M}_{in}$ in *z* direction, i.e. along the applied field **H;** brackets mean statistical averaging over all orientations of the particle axis.

The linear, with respect to the field **H**, magnetization $\boldsymbol{M}_{in}$ in the particle can be determined by using the classical results of the theory of polarization of dielectric ellipsoids [30]. After simple transformations, the component $M_{in,z}$ can be calculated as:

$$M_{in,z} = \chi_p H \left(\frac{\cos^2\theta}{1+\chi_p N_\parallel} + \frac{\sin^2\theta}{1+\chi_p N_\perp} \right) \quad , N_\perp = \frac{1-N_\parallel}{2} \tag{2}$$

Here χ_p is initial susceptibility of the particle material, angle θ is shown in Fig.2, $N_\parallel$ and $N_\perp$ are the components of the particle demagnetizing factor along and perpendicular to the main axis of the ellipsoid respectively.

The explicit form of the demagnetizing factor is [30]:

$$N_\parallel = \begin{cases} \dfrac{\xi}{2\left(\xi^2-1\right)^{3/2}} \left[\ln \dfrac{\xi+\sqrt{\xi^2-1}}{\xi-\sqrt{\xi^2-1}} - 2\dfrac{\sqrt{\xi^2-1}}{\xi} \right], \xi > 1 \\ \dfrac{\xi}{\left(1-\xi^2\right)^{3/2}} \left[\dfrac{\sqrt{1-\xi^2}}{\xi} - \mathrm{a}\tan \dfrac{\sqrt{1-\xi^2}}{\xi} \right], \xi < 1 \end{cases}$$

Here ξ is aspect ratio of the ellipsoid (ratio of the ellipsoid axis of symmetry to its diameter).

Let $\boldsymbol{u}$ be a vector of the macroscopic displacement in the composite. The uniaxial elongation of an incompressible sample corresponds to the following relations:

$$\frac{\partial u_z}{\partial z} = \varepsilon, \frac{\partial u_x}{\partial x} = -\frac{\varepsilon}{2}, \ \frac{\partial u_y}{\partial y} = -\frac{\varepsilon}{2} \tag{3}$$

Here ε is relative elongation of the sample, which is supposed small ($\varepsilon \ll 1$).

We will denote initial, before the sample elongation, value of the angle θ of a given particle as θ_0. Since the elongation ε is supposed small, the difference $\delta\theta = \theta - \theta_0$ also must be small. By using $\theta = \theta_0 + \delta\theta$ in eq.(2), in the linear approximation with respect to $\delta\theta$ we get:

$$M_{in,z} = M_{in,z0} + \delta M_{in},$$

$$M_{in,z0} = M_{in,z}(\theta_0), \quad \delta M_{in} = -\chi_p H (\kappa_\parallel - \kappa_\perp) \sin 2\theta_0 \, \delta\theta \tag{4}$$

$$\kappa_{\parallel,\perp} = \frac{1}{1+\chi_p N_{\parallel,\perp}}$$

In order to determine the deviation angle , we will use the results [31,32] of hydromechanics of suspension of elongated particles, as well as the mathematical identity of the linear Navier - Stokes equation and the Lame equation of the small deformations of elastic media.

Equations [31,32] of dynamics of a non Brownian magnetizable particle, suspended in a Newtonian liquid, can be presented as:

$$\frac{d\theta}{dt} = -\frac{3}{2}\lambda\dot{\varepsilon}\sin\theta\cos\theta + \mu_0 H^2 \frac{1}{6\eta\varrho}\chi_p^2 (N_\perp - N_\parallel)\,\kappa_\parallel\kappa_\perp \sin\theta\cos\theta$$

$$\lambda = \frac{\xi^2-1}{\xi^2+1}$$

$$\varrho = \frac{\xi^2 + 1}{4(N_{\parallel}(2\xi^2 - 1) \quad + 1)}$$

Here $\dot{\varepsilon}$ is the rate of elongation of the suspension flow, η is viscosity of the current fluid, μ_0 is the vacuum magnetic permeability. These equations, in the inertialess approximation, correspond to the balance between the hydrodynamic and magnetic torques, acting on the particle.

To get the equations for the particle turn in an elastic medium, we must replace the elongation rate $\dot{\varepsilon}$ to the static elongation ε; the viscosity η to the matrix shear modulus *G*; the derivates $\frac{d\theta}{dt}$ to the deviation angle $\delta\theta$. In the linear approximation with respect to ε , one can get:

$$\delta\theta = -\frac{3}{4}\varepsilon\lambda \sin 2\theta_0 + insignificant\ terms \tag{5}$$

The mean magnetization of a particle can be calculated as:

$$< M_{in,z} > = < M_{in,z0} > + < \delta M_{in,z} >, \tag{6}$$

$$< \cdots > = \frac{1}{2}\int_0^{\pi} \ldots \sin\theta_0 d\theta_0$$

Here brackets mean averaging over all initial orientations of the particles.

Substituting eqs. (2), (4) and (5) into (6), we calculate the average magnetic moment of the particle:

$$< M_{in,z0} > = \frac{1}{3}\chi_p H(\kappa_{\parallel} + \kappa_{\perp}) \tag{7}$$

$$< \delta M_{in,z} > = \frac{2}{5}\lambda\chi_p H(\kappa_{\parallel} - \kappa_{\perp})\varepsilon$$

Combining eqs.(1) and (7), one gets:

$$\chi = \chi_0 + \delta\chi,$$

$$\chi_0 = \frac{1}{3}\chi_p(\kappa_{\parallel} + \kappa_{\perp})\varphi \tag{8}$$

$$\delta\chi = Q\chi_p\varphi\varepsilon; \quad Q = \frac{2}{5}\lambda(\kappa_{\parallel} - \kappa_{\perp})$$

Some results of calculations of the coefficient Q as a function of the aspect ratio ξ are shown in Fig.3.

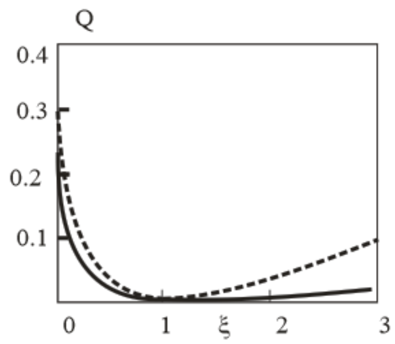

Fig.3 The factor Q vs. the particle aspect ratio ξ. Dashed and solid lines $\chi_p = 10$ and 100 respectively.

The coefficient Q is positive for all magnitudes of the aspect ratio ξ. Therefore elongation of the sample leads to increase of the composite magnetic susceptibility.

3. **Spherical magnetizable particles**.

In the case of the spherical particles (ξ=1, $\kappa_{\parallel} = \kappa_{\perp}$) the approximation (8) of the non interacting inclusions gives $\delta\chi = 0$. Thus, to determine the mechanomagentic effect in the composite with magnetizable spheres, the interparticle interactions must be taken into account.

Magnetic susceptibility χ of a composite with the spherical inclusions can be calculated from the general relation [30,33]

$$\chi H = \chi_p \varphi < H_{in} > \qquad (9)$$

Here *H* again is the mean (Maxwell) field inside the sample, H_{in} is the field inside an arbitrary particle, the brackets <…> mean averaging over all physically possible positions of other particles.

The main problem of a theory of composite materials is account of the cooperative interaction between many particles. No general results have been obtained here. However in the case of single non interacting particles this problem can be solved strictly [30,33,34]. For the spherical particles with high susceptibility $\chi \gg 1$ the result reads:

$$H_{in}^0 \approx 3H \qquad (10)$$

The superscript 0 denotes the approximation of the non interacting particles.

In order to take the interparticle interaction by using as mathematically strict results as possible, we will restrict ourselves by the well-known pair approximation, i.e. will take into account interaction only between two particles, ignoring effects of any third one. The particles are illustrated in Fig.4.

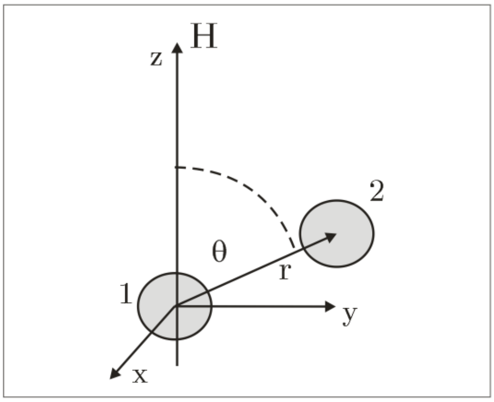

Fig.4. Cluster of two interacting particles. Explanations are in the text.

Magnetic field, induced by a particle inside the other one (say, by the particle number 2 inside the particle number 1 in Fig.4) can be estimated in the simplest dipole-dipole approximation. This approximation is quite accurate when the distance r between the particles centers significantly exceeds diameter of the particle, however it leads to serious mistakes at the particles close disposition [35,36].

To determine the induced field, we will use here the results of [36], where energy of the multidipole magnetic interaction between two particles has been estimated on the basis of analytical extrapolation of results of numerical calculations. In the case $\chi_p \gg 1$, the result of [36] reads:

$$W = -\mu_0 v \left[3 + 3\sum_{k=3}^{7}\left(\frac{a_k}{(\rho-b_k)^k} + \frac{c_k}{(q-d_k)^k}\cos^2\theta\right)\right] H^2 \qquad (11)$$

Here W is total energy of two linearly magnetizable particles placed in the field H; $\rho=r/a$; a is the particle radius, parameters $a_k \ldots d_k$ are tabulated in [36]; θ is angle between the radius vector $\boldsymbol{r}$, linking the particles, and the field **H** (see Fig.4). The first term in the square brackets of (11) presents the energy of interaction of two isolated particles with the field H; the second one, in the extrapolation of [36], corresponds to the multipole interaction between these particles. For ρ>>1 the second term in (11) coincides with the energy of the dipole-dipole interaction.

The needed z-component of magnetization $\boldsymbol{M}_{in}$ inside each of the particles can be calculated from the general thermodynamic relation between a material magnetization and its energy in an external field (see, for example, [30]). Taking into account that *W* is energy of the *two* particle cluster, by using eq. (11), one gets:

$$M_{in,z} = -\frac{1}{2v\mu_0}\frac{\partial W}{\partial H} = \left[3 + 3\sum_{k=3}^{7}\left(\frac{a_k}{(\rho - b_k)^k} + \frac{c_k}{(q-d_k)^k}\cos^2\theta\right)\right]H \quad (12)$$

Taking into account that $M_{in,z} = \chi_p H_{in}$, by using eq.(9) one comes to the relation:

$$\chi = \varphi\frac{< M_{in,z} >}{H} \quad (13)$$

In order to determine the magnetization $< M_{in,z} >$ in (13), one must average eq. (12) over radius-vector $\boldsymbol{r}$ of all physically possible positions of the second particle, shown in Fig.4.

For simplification of the further consideration, we will present the magnetization as:

$$M_{in,z} = M_{in,z}^0 + M'_{in,z},$$

$$M_{in,z}^0 = 3H, \quad M'_{in,z} = 3H\sum_{k=3}^{7}\left(\frac{a_k}{(\rho - b_k)^k} + \frac{c_k}{(q-d_k)^k}\cos^2\theta\right) \quad (14)$$

Here $M_{in,z}^0$ is magnetization of the single particle, M'_{inz} is a part o the magnetization induced by the second particle inside the first one.

Let $p(\mathbf{r})$ be the density of probability of the given relative disposition of the particle, normalized as:

$p \to 1$, at $r \to \infty$.

By using standard considerations of statistical mechanics (see, for example, [37]) one can present the mean magnetization of the particle as:

$$< M_{in,z} >= M_{in,z}^0 + \frac{\varphi}{v}\int M'_{in,z}(\boldsymbol{r})\, p(\boldsymbol{r}) d\boldsymbol{r} \quad (15)$$

Combining eqs.(15) and (13), one determines the susceptibility χ of the composite.

Let us write down the distribution function *p* as:

$$p = p_0 + \delta p \quad (16)$$

The function p_0 corresponds to the initial, non elongated state of the composite, δp reflects effect of the sample elongation on the particles relative disposition.

By using the results of [38], we come to the following equation with respect to δp

$$\delta p = -div(p_0\mathbf{w}) = -\left(p_0 div\mathbf{w} + (\mathbf{w}\cdot\nabla)p_0\right) \tag{17}$$

Here **w** is the vector of displacement of the second particle with respect to the first one.

We will write down p_0 in the form of the pair distribution function the gas of hard spheres [37]:

$$p_0 = \begin{cases} 0, r < 2a \\ 1 + 8\varphi\left(1 - \frac{3r}{8a} + \frac{r^3}{128a^3}\right), 2a < r < 4a \\ 1, r > 4a \end{cases} \tag{18}$$

In order to determine the vector **w,** we will use the results of [39] of hydromechanics of suspensions of hard spheres in Newtonian liquids as well as the similarity between equation of the liquid flow and equation of the Hook deformations of elastic media. By using the spherical coordinate system, with the radius-vector *r* and the polar angle θ (see Fig.4) , and the results of [39] for the uniaxial elongation flow, we come to the following relation

$$div\mathbf{w} = -\frac{\varepsilon}{2}\left[\frac{3(A(r) - B(r))}{r^2} + \frac{1}{r}\frac{dA(r)}{dr}\right](3\cos^2\theta - 1)r^2 \tag{19}$$

$$(\mathbf{w}\nabla)p_0 = w_r\frac{\partial}{\partial r}p_o$$

$$w_r = \frac{\varepsilon}{2}(1 - A(r))(3\cos^2\theta - 1)r$$

Here w_r is the radial component of the vector **w**; *A* and *B* are functions of the distances *r* between the particles centers, introduced and tabulated in [39].

By using (13) and (15,16), after simple transformations, one gets:

$$H\delta\chi = \varphi\delta < M'_{in,z} > \tag{20}$$

$$\delta < M'_{in,z} > = \frac{\varphi}{\mathrm{v}}\int M'_{in,z}(\mathbf{r})\delta p(\mathbf{r})d\mathbf{r} = 2\pi\frac{\varphi}{\mathrm{v}}\int_{2a}^{\infty}\left(\int_0^{\pi} M'_{in,z}(\mathbf{r})\delta p(\mathbf{r})\sin(\theta)d\theta\right)r^2 dr$$

Combining eqs.(13) and (17- 20) we calculate the term $\delta < M'_{in,z} >$ and, therefore, the change $\delta\chi$ of the sample susceptibility .

By using the table of numerical values of the functions *A* and *B*, given in [39], we have estimated the integral over *r* in eq. (20) in the form of the trapeze approximation of the Riemann sum. The integral over the angle θ in (20) is calculated analytically.

After these transformations, the following relation for the composite susceptibility has been obtained:

$$\chi \approx 3\varphi + \delta\chi \qquad (21).$$

$$\delta\chi = -\varepsilon\varphi^2 \frac{12}{5}(1.3 - 2.8\varphi)$$

The term 3φ in (21) is the susceptibility of the non deformed composite. For simplicity we have omitted here the term, proportional to φ^2. That is acceptable for the practical use when the concentration φ is in the frames of 10-15%. The term $\delta\chi$ is the change of the composite susceptibility because of its elongation. This result demonstrates that the term $\delta\chi$ is negative (i.e. effective susceptibility χ decreases while the sample elongation) if the particles concentration φ is small enough; in contrast, it increases when the concentration exceeds some threshold magnitude φ_c , estimated as $\varphi_c \approx 0.45$. The physical reason of this change of the sign of $\delta\chi$ is appearance of the short ranged order of the particles spatial disposition. One needs to note that the term $1.3 - 2.8\varphi$ in (21) is determined by the used method of numerical calculation of the integral over *r* in (20). It can be précised if the explicit forms of the functions $A(r)$and $B(r)$ are known.

Conclusion.

We present results of theoretical study of effect of ferrogels elongation on their effective magnetic susceptibility. In order to get mathematically regular results, free from any intuitive and heuristic constructions, we have considered the systems with low concentration of the particles. Our results show that susceptibility of the composites with non spherical (ellipsoidal) inclusions enhances at the sample elongation. In the case of the systems with spherical particles, the mechanomagnetic effect appears because of change, at the macroscopic deformation, of the mutual disposition of the magnetically interacting particles. The susceptibility decreases, after the sample elongation, if the concentration of the particles is low enough and increases when the concentration exceeds some threshold magnitude.

For mathematical simplicity and transparence of the physical results, we have considered the situation, when the applied field **H** is parallel to the axis of the sample elongation. The developed approach, without serious modifications, allows studying of the mechanomagnetic effect at the arbitrary orientations of the field.

We believe that the present mathematically regular results, obtained in the asymptotic of low concentration of the particles, can be a robust background for development of models of this effect in the more concentrated magnetic gels. The studied mechanomagnetic effect can be promising for development of technologies of artificial muscles, actuators, sensors, magnetocontrolled scaffolds and tissue engineering

Coming back to possible biomedical applications, we would like to mention recent strong request for multifunctional biosensor systems where soft matters play different roles. For example, it was shown that ferrogel thin layer can be a good basis for enhanced cells adhesion [25]. We can therefore propose hypothetical situation for low invasive surgery with location of ferrogel implant sample carrying certain amount of cells for in-situ tissue regeneration. Application of external magnetic field of controlled strength can be useful for the implant size adjustment and this adjustment can be controlled by highly sensitive to the effective magnetic susceptibility magnetic field detector like giant magnetoimpedance based sensor [40].

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