This article was published as: Physica A: Statistical Mechanics and its Applications 486, 908-914, 2017

DOI: https://doi.org/10.1016/j.jmmm.2019.01.112

To the theory of shear elastic properties of magnetic gels.

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Abstract

We present results of theoretical study of elastic shear modulus of magnetic gels, consisting of single non Brownian magnetic particles, homogeneously (gas-like) distributed in an elastic matrix. The composite is placed in magnetic field, perpendicular to the direction of the sample shear. Effect of both, magnetically hard and soft particles is studied. In order to get mathematically rigorous results, we have restricted ourselves by the analysis of the composites with low concentration of the particles and neglected any interactions between them. Only small deformations of the system were considered. Analysis shows that effect of magnetic field on the macroscopic (measurable) shear modulus of the composite can be comparable with that, provided by the presence of the rigid inclusions in the elastic matrix. The suggested asymptotic model can be a robust background for the study of the systems with moderate or high concentration of the particles.

Keywords: Magnetic gels; shear deformations; elastic modulus

I. Introduction.

Magnetic gels and elastomers are composites of fine magnetic particles in soft polymer matrixes. Coupling of rich set of physical properties of polymer and magnetic materials is very promising for many modern and perspective technologies. Discussions of technical and biomedical application of these systems can be found, for example, in [1-12]. A short overview of works on mechanical properties and behavior of magnetic polymers is given in [13].

Uniaxial elongation and magnetostriction effects in magnetic gels have been studied in many works (see, for example [13-19]). The shear deformations of these systems also present significant interest both from scientific and practical points of view. Theoretical studies of the shear effects in the composites with the particles, united in linear chain-like aggregates, have been done in [20-22]. The general conclusion of these works is that an external magnetic field can significantly increase the shear modulus of these composites.

As a rule, the chain-like aggregates appear in magnetic polymers on the stage preceding the composite curing due to the action of an external magnetic field (field of polymerization). On the other hand, very often magnetic gels are prepared without this field. The spatial distribution of particles in these systems is rather random and isotropic (see, for example, [15,17,23]). The aim of this work is theoretical study of effect of an external magnetic field on the shear elastic modulus of magnetic gels with homogeneous and isotropic distribution of non Brownian particles in a continuous matrix. It should be noted that usually the Brownian effects are negligible for the magnetic particles with the diameter 100nm and more. Composites with the particles of these sizes present the main interest from the point of view of the magnetomechanic effects, since these effects, in the systems with the smaller particles, as a rule, are very weak.

The matrix is supposed elastic with the linear law of deformation and incompressible. It should be noted that the last condition is fulfilled not for all gels; however it allows us to restrict calculations and to get the final results in transparent forms. Analysis of effects of the composite compressibility can be considered as a natural generalization of this model.

The principal and not overcome problem of the theory of composite materials is account of multiparticle interactions, both the direct ones and interactions through the perturbations of the current matrix Usually these effects are taken into account by using various empirical and semi empirical approaches, which accuracy a priory is unknown [24].

In order to achieve mathematically rigorous results, here we will consider the systems with low concentration of the particles and neglect any interactions between them. One needs to admit that the low-concentrated systems are not very interesting from the practical point of view. However this limiting model allows us to avoid intuitive and heuristic constructions. That is why the strict results can be considered as a robust asymptotic background for the analysis of the concentrated system with the interacting particles.

The structure of the paper is the following. In the part II we study the composite with identical spherical magnetically hard particles; each particle has a permanent magnetic moment bounded with the particle body. Part III deals with the systems of magnetically soft ellipsoidal particles with random orientations of the ellipsoids axes.

II. Magnetically hard spherical particles.

We consider a system of identical spherical non Brownian particles embedded in an elastic continuous medium. All particles have the permanent magnetic moment $\boldsymbol{m}$ “frozen” in the particle body. This means that the moment can turn round only with the particle. We suppose that the volume concentration φ of the particles is low and will neglect any interactions between them. Additionally, for the maximal simplification of calculations, we suppose the strong coupling between the particles and the host gel, i.e. the no-slipping condition on the particles surface.

Let us suppose that the composite is placed in a uniform magnetic field **H** and experiences the small shear deformation in the plane, perpendicular to the field. Since the concentration of the particles in the composite is supposed small, we will not take into account the difference between the external field **H** and the field inside the sample.

It is convenient to introduce a Cartesian coordinate system with the axis *Oz* in the field direction and the axis *Ox* in the direction of the shear. By using the mathematical similarity between the stationary Navier – Stokes equation of Newtonian incompressible fluid at the low Reynolds number and the Lame equation of deformation of an elastic poorly compressible medium [24], as well as the results of theory of dilute magnetic fluids (see, for example, [25]), one can present the needed component of the macroscopic (measurable) stress $\boldsymbol{\sigma}$ in the composite as:

$$\sigma = G_0(1 + 2.5\varphi)\gamma + \mu_0 \frac{m}{2v} < e_x > H\ , \quad \sigma = \sigma_{xz} \qquad (1)$$

Here G_0 is the shear modulus of the pure polymer matrix, $= \frac{\partial u_x}{\partial z}$, u_x is the component of the macroscopic (measurable) vector $\boldsymbol{u}$ of the composite displacement, μ_0 is the magnetic permeability of vacuum, v is volume of the particle, e_x is the component of the unit vector $\boldsymbol{e}$, directed along the magnetic moment $\boldsymbol{m}$ of a particle, the angle brackets <…> mean averaging over the orientations of all particles. Our aim now is to determine the mean component $< e_x >$.

To this end we will consider an arbitrary particle situated in the field **H** and denote by $\boldsymbol{e}_0$its initial (before the composite deformation) vector $\boldsymbol{e}$. Equation for this vector can be obtained by using equations [25-27] of magnetic particle rotation in a viscous fluid and the mathematical identity of the Navier-Stokes and Lame equations.

In the chosen Cartesian coordinate system equations [25-27] for the components of the vector $\boldsymbol{e}$ can be presented as:

$$\frac{de_x}{dt} = \frac{1}{2}\dot{\gamma}e_z - se_xe_z, \qquad (2)$$

$$\frac{de_z}{dt} = -\frac{1}{2}\dot{\gamma}e_x + s(1 - e_z^2)$$

$$s = \frac{\mu_0 mH}{6\eta_0 v}$$

Here $\dot{\gamma}$ is the shear rate of the suspension flow, η_0 is viscosity of the current fluid. 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 shear rate $\dot{\gamma}$ to the shear strain γ; the viscosity η_0 to the matrix shear modulus G_0; the derivates $\frac{de_i}{dt}$ to the deviations $\delta e_i = e_i - e_{i0}$ of the vector $\boldsymbol{e}$ components from their initial (before the macroscopic deformation and the field application) magnitudes e_{i0} [24]. As a result, the needed equations read:

$$\delta e_x = \frac{1}{2}\gamma e_z - \kappa e_x e_z, \tag{2}$$

$$\delta e_z = -\frac{1}{2}\gamma e_x + \kappa(1 - e_z^2)$$

$$\kappa = \frac{\mu_0 mH}{6G_0 v}$$

The classical Lame equations correspond to the linear Hook approximation with respect to the matrix deformation. Keeping it in mind, in the linear approximation with respect to γ after transformations we get:

$$\delta e_x = \frac{1}{2}\gamma\left(\frac{e_{z0}}{1+\kappa e_{z0}} + \frac{1-e_{z0}^2+e_{x0}^2}{(1+\kappa e_{z0})(1+2\kappa e_{z0})}\right) - \kappa^2 \frac{e_{x0}(1-e_{z0}^2)}{(1+\kappa e_{z0})(1+2\kappa e_{z0})} \tag{3}$$

We suppose that initially the particles had random orientation of their magnetic moments, i.e. $< e_{x0} > = 0$. Therefore

$$< e_x > = < e_{x0} > + < \delta e_{x0} > = < \delta e_{x0} >. \tag{4}$$

This is convenient to introduce the spherical coordinate system with the polar θ and azimuthal ϕ angles, so that:

$$e_{z0} = \cos\theta\,; \quad e_{x0} = \sin\theta\cos\phi \tag{5}$$

By using (5), one can get:

$$< \delta e_x > = \frac{1}{4\pi}\int_0^{\pi} \sin\theta d\theta \int_0^{2\pi} \delta e_x d\phi \tag{6}$$

Substituting (3) and (5) into (6), we obtain:

$$< \delta e_x >= \frac{1}{4}\gamma\left(\int_{-1}^{1}\frac{x}{1+\kappa x}dx + 1.5\int_{-1}^{1}\frac{1-x^2}{(1+\kappa x)(1+2\kappa x)}dx\right) \quad (7)$$

The integrals (7) can be calculated analytically; however they have cumbersome forms, that is why we omit these forms here.

Parameter κ presents the ratio of the magnetic and elastic torques, acting on the particle. The Lame equations of the elastic deformation of a continuum are valid only in the case of small deformations of this medium. In part, this means that the angle of the particle turn, under the action of the magnetic and elastic torques, must be small for these equations applicability. This leads to the condition $\kappa < 1$ of restriction of the linear approximation.

Combining eqs. (7) and (1), we come to the following relations:

$$\sigma = G_1\gamma; \quad (8)$$

$$G_1 = G_0(1 + \frac{5}{2}\varphi + q(\kappa)\varphi)$$

$$q(\kappa) = \frac{3}{4}\kappa\left(\int_{-1}^{1}\frac{x}{1+\kappa x}dx + 1.5\int_{-1}^{1}\frac{1-x^2}{(1+\kappa x)(1+2\kappa x)}dx\right)$$

Parameter G_1 is the effective shear modulus of the composite with the magnetically hard spheres, q reflects the addition to G_1 due to the magnetic field effect. Results of this parameter calculation are shown in Fig.1.

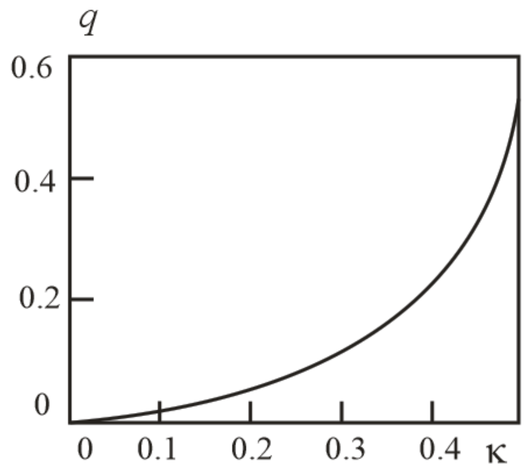

Fig.1 Parameter q in (8) vs. the dimensionless magnetic field κ

In the frame of the used approximation, effect of magnetic field on the effective shear modulus of the composite can achieve about one fourth of the effect of the solid inclusions, described by the Einstein term 2.5φ. It should be noted that the situation when $\kappa \sim 1$ or more is beyond of the approximation of small deformation inside the elastic matrix. The linear Lame equations cannot be used for description of the particle rotation under the magnetic and mechanic torques for the large values of κ. Analysis of this case requires numerical solution of non linear equations of the polymer matrix deformation.

III. Ellipsoidal magnetically soft particles.

In this part we consider a system of ellipsoidal magnetically soft particles randomly distributed in an elastic matrix. For the maximal simplification of calculations and to get transparent physical results, we will restrict ourselves by the approximation of the linearly magnetizable particles. We suppose again the strong coupling of the particles with the polymer matrix and the non slipping condition on the particles surface. The generalization to the non linear magnetization as well as to the finite coupling is not difficult, but leads to cumbersome calculations and final results.

We again suppose that the composite experiences the deformation of simple shear with the mean displacement $\boldsymbol{u}$ in the direction *Ox* and the gradient of the displacement along the axis *Oz* . Magnetic field **H** is aligned along the axis *Oz*.

The analytical form for the components of the stress tensor in suspension of ellipsoidal particles is given in [26,27]. In order to get the expression for the stress in the elastic composite, we replace, in the relation [26,27], the shear rate $\dot{\gamma}$ to the shear strain γ and coefficient of the current fluid viscosity η_0 to the shear modulus G_0 of the elastic matrix. As a result, instead of eq. (1), we get

$$\sigma = G_0\Big\{1+\varphi\Big[\alpha+\frac{1}{2}[(\xi+\beta\lambda)(<e_x^2>+<e_z^2>)+\beta(<e_z^2>-<e_x^2>)+2(\chi-2\lambda\beta)-<e_x^2e_z^2>]\gamma+\varphi g h^2<e_xe_z>\Big]\Big\}, \tag{9}$$

$$\sigma=\sigma_{xz}\,;\gamma=\frac{\partial u_x}{\partial z}, \qquad g(r)=\frac{(\mu_p-1)^2(N_\perp-N_\parallel)}{(1+(\mu_p-1)N_\perp)(1+(\mu_p-1)N_\parallel)}, \qquad h=\sqrt{\frac{\mu_0H^2}{2G_0}}$$

Here $\boldsymbol{e}$ is the unit vector aligned along the particle axis of symmetry; α,β,λ,ξ and χ are functions on the aspect ratio *r* of the ellipsoidal particle (the ratio of the particle axis of symmetry to its diameter), μ_p is the particle relative magnetic permeability, $N_\parallel$ and $N_\perp$ are the demagnetizing factors of the particle along and perpendicularly its axis of symmetry respectively. The explicit forms of the shape-functions $\alpha,\beta,\lambda,\xi,\chi$ as well as of the factors $N_\parallel$ and $N_\perp$ are given in the Appendix.

We will present again the components of the vector $\boldsymbol{e}$ of an arbitrary particle as

$e_i = e_{i0} + \delta e_i$, i=x,z. Because of the initial random orientation of the particles, we get: $< e_{i0}^2 >= \frac{1}{3}$; $< e_{x0}e_{z0} >= 0$; $< e_{x0}^2 e_{z0}^2 >= \frac{1}{15}$. By using these relations in (9), in the linear approximation with respect to γ one can obtain:

$$\sigma = G_0\left\{1 + \varphi[\alpha + \frac{1}{3}(\xi + \beta\lambda) + \frac{1}{15}(\chi - 2\lambda\beta)]\gamma + \varphi g h^2(< e_{x0}\delta e_z > + < e_{z0}\delta e_x >)\right\} \quad (10)$$

Equations [27] for the vector $\boldsymbol{e}$ of a magnetizable ellipsoid in a viscous fluid can be written down as:

$$\frac{de_x}{dt} = \frac{1}{2}\dot{\gamma}[\lambda(1 - 2e_x^2) + 1]e_z - \Psi e_x e_z^2,$$

$$\frac{de_z}{dt} = \frac{1}{2}\dot{\gamma}[\lambda(1 - 2e_z^2) - 1]e_x + \Psi(e_z - e_z^3)$$

$$\Psi(r) = \frac{\mu_0 H^2}{2\eta_0}\frac{g(r)}{3\delta(r)}$$

Here $\dot{\gamma}$ and η_0 are the shear rate and viscosity of the fluid; $\delta(r)$ is a function of the particle aspect ratio r (don't miss with the Dirac function). The explicit form of this function is given in the Appendix.

Replacing again $\dot{\gamma}$ to γ; η_0 to G_0 and $\frac{de_i}{dt}$ to δe_i, we get:

$$\delta e_x = \frac{1}{2}\gamma[\lambda(1 - 2e_x^2) + 1]e_z - \psi e_x e_z^2, \quad (11)$$

$$\delta e_z = \frac{1}{2}\gamma[\lambda(1 - 2e_z^2) - 1]e_x + \psi(e_z - e_z^3)$$

$$\psi(r) = h^2\,\frac{g(r)}{3\delta(r)} \quad (12)$$

Parameter h presents the ratio of the magnetic and elastic torques, acting on the particle in the non deformed composite. Similar to the previous case of the magnetically hard particles, the linear Lame equations of the small deformations of the elastic matrix are applicable only when the inequality h<1 is held.

Substituting the form $e_i = e_{i0} + \delta e_i$ into eq. (11), after simple transformations, in the linear approximation in , we come to the relations:

$$< e_{z0}\delta e_x >= \frac{1}{2}\gamma\left[< \frac{Ae_{z0}}{1-\psi e_{z0}^3} > -\psi < \frac{2Be_{x0}e_{z0}}{(1-\psi e_{z0}^3)(1-\psi(1-3e_{z0}^2))} >\right] \quad (13)$$

$$< e_{x0}\delta e_z >= \frac{1}{2}\gamma < \frac{e_{x0}B}{1-\psi(1-3e_{z0}^2)} >,$$

Here

$$A = [\lambda(1 - 2e_{x0}^2) + 1]e_{z0}\,, B = [\lambda(1 - 2e_{z0}^2) - 1]e_{x0}$$

Combining (10) and (13), we get:

$$\sigma = G_2\gamma, \tag{14}$$

$$G_2 = G_0\{1+\varphi f(r,h)\},$$

$$f(r,h) = [f_1(r) + f_2(r,h)]$$

$$f_1 = \alpha + \frac{1}{3}(\xi + \beta\lambda\,) + \frac{1}{15}(\chi - 2\lambda\beta)$$

$$f_2 = \frac{3}{2}\psi\delta(r)\left[< \frac{Ae_{z0}}{1-\psi e_{z0}^3} > -h^2 < \frac{2Be_{x0}e_{z0}}{(1-\psi e_{z0}^3)\big(1-\psi(1-3e_{z0}^2)\big)} + < \frac{e_{x0}B}{1-\psi(1-3e_{z0}^2)} >\right]$$

Here G_2 is the effective shear modulus for the composite with the magnetically soft ellipsoidal particles. The terms f_1 describes effect of the rigid randomly oriented particles on this modulus; the term f_2 reflects the influence of the magnetic field H on G_2, f indicates the total effect of the particles on the elastic modulus G_2 . For the spherical particles ($r = 1$) the relations, given in the Appendix, read: $\alpha = \frac{5}{2}$; $\beta, \lambda, \xi, \chi, \psi$ =0. Therefore, the Einstein formula $G_2 = G_0\left(1+\frac{5}{2}\varphi\right)$ for these particles is fulfilled.

Some results of calculations of the terms f, f_1 and f_2 , vs. the dimensionless magnetic field h as well as vs. the particle aspect ratio r , are shown in Figs. 2 and 3 respectively.

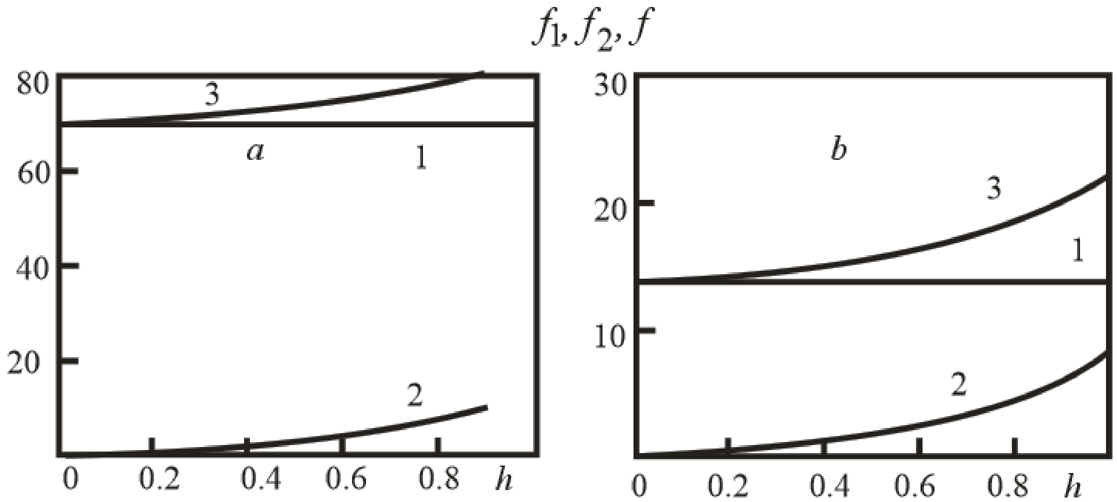

Fig.2. Parameters f_1, f_2 and f, which determine the effective shear modulus G_2 in eq. (14) of the composite with the magnetically soft ellipsoids vs. the dimensionless magnetic field h. (a) and (b) – the particle aspect ratio r = 0.01 and 10 respectively. Figures near the curves: 1 – f_1; 2 - f_2; 3 – $f = f_1 + f_2$

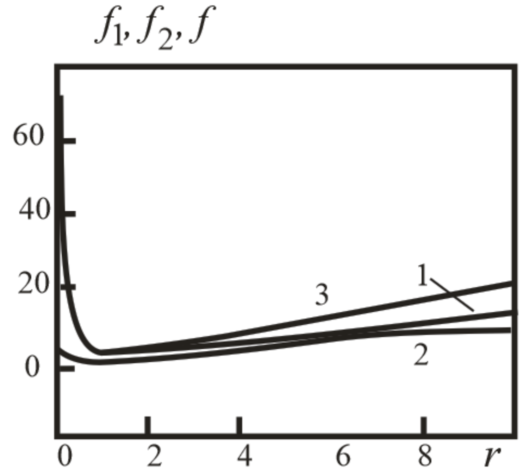

Fig.3. Parameters $f_1, f_{2,} f$ vs. the particle aspect ratio *r*. The dimensionless magnetic field *h*=1. Figures near curves mean the same as in Fig.2.

These results demonstrate that the "magnetic" term f_2 can give contribution to G_2 very close to the "rigid particles" term f_1. This contribution is especially significant for the highly elongated particles (*r*>>1). For the oblate particles (*r*<1) and the relatively weak fields (*h*<1) the term f_2 is much less than f_1.

Conclusion.

We present results of theoretical study of effect of uniform magnetic field on the shear modulus of a ferrogels, consisting of magnetic particles randomly distributed in a polymer matrix. In order to achieve mathematically strict results, we have restricted ourselves by analysis of the dilute systems and neglected any interactions between the particles. The results show that magnetic field increases the modulus and this effect can be quite comparable with that, provided by the particles as rigid inclusions in the composite. We believe that the results, obtained in the limiting case of the low concentrated systems, can be a robust background for the development of theory of the moderately and highly concentrated soft magnetic composites. It should be noted, that the we restricted ourselves by the spherical shape of the magnetically hard particles just for maximal simplification of mathematical part of the work. Combining the approaches, considered in the parts II and III, one can easily generalize this analysis for the magnetically hard ellipsoids.

Acknowledgements

A.Z. and L.I. are grateful for the financial support to the Russian Fund for Basic Research, 16-58-12003 and the Program of Russian Federation Ministry of Science and Education, project 3.1438.2017/PCh.

M.L-L was supported by the project FIS2013-41821-R (Plan Nacional de Investigación Científica, Desarrollo e Innovación Tecnológica, Ministerio de Economía y Competitividad, Spain, co-funded by ERDF, European Union).

APPENDIX

The shape-coefficients $\alpha,\ldots\chi$, as functions of the particle aspect ratio r, have the following forms [25]:

$$\alpha(r) = \frac{1}{r\alpha_0'}, \ \beta(r) = \frac{2\left(r^2-1\right)}{r\left(r^2\alpha_0+\beta_0\right)}, \ \zeta(r) = \frac{4}{r\left(r^2+1\right)\beta'_0} - \frac{2}{r\alpha_0'},$$

$$\chi(r) = \frac{2\alpha_0''}{r\alpha_0'\beta_0''} - \frac{8}{r\left(r^2+1\right)\beta'_0} + \frac{2}{r\alpha_0'}, \ \lambda(r) = \frac{r^2-1}{r^2+1}, \delta(r) = \frac{\beta(r)}{3\lambda(r)}$$

Here

$$\alpha_0 = -\frac{1}{r^2-1}\left[\frac{2}{r} + \frac{1}{\sqrt{r^2-1}}\ln\left(2r^2-1-2r\sqrt{r^2-1}\right)\right],$$

$$\beta_0 = \frac{1}{r^2-1}\left[r - \frac{1}{2\sqrt{r^2-1}}\ln\left(2r^2-1+2r\sqrt{r^2-1}\right)\right],$$

$$\alpha_0' = \frac{1}{4\left(r^2-1\right)^2}\left[r\left(2r^2-5\right) - \frac{3}{2\sqrt{r^2-1}}\ln\left(2r^2-1-2r\sqrt{r^2-1}\right)\right],$$

$$\beta_0' = \frac{1}{\left(r^2-1\right)^2}\left[\frac{r^2+2}{r} - \frac{3}{2\sqrt{r^2-1}}\ln\left(2r^2-1+2r\sqrt{r^2-1}\right)\right],$$

$$\alpha_0'' = \frac{1}{4\left(r^2-1\right)^2}\left[r\left(2r^2+1\right) - \frac{4r^2-1}{2\sqrt{r^2-1}}\ln\left(2r^2-1+2r\sqrt{r^2-1}\right)\right],$$

$$\beta_0'' = -\frac{1}{\left(r^2-1\right)^2}\left[3r + \frac{2r^2+1}{2\sqrt{r^2-1}}\ln\left(2r^2-1-2r\sqrt{r^2-1}\right)\right].$$

The demagnetizing shape-factors $N_{\|}$ and $N_{\perp}$ are [28]

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

$$N_{\perp} = \frac{1-N_{\|}}{2}$$

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