Deformation of a sphere made of magnetoactive elastomer under a strong uniform magnetic field

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Abstract. Magnetostriction effect of a spherical sample of a magnetoactive elastomer (MAE) is analyzed. In comparison with the preceding study, the consideration is done on a more realistic basis: taking into account saturation of the MAE magnetization in contrast to the former model where the magnetization was supposed to be linear whatever the field strength. This more thorough investigation has revealed that the striction-induced elongation effect, depending on the material parameters, may occur in two forms. One scenario manifests itself as tapering of the polar zones of the former sphere, where ‘beaks’ are formed, so that the shape of the object drastically deviates from a spheroidal one. The mechanism the underlies the occurrence of beaks is the surface instability of a magnetizable elastic continuum, and the beak nucleation follows the second-order transition pattern; the resulting overall elongation of the body does not display any hysteresis. Another scenario—it is related to MAEs with higher magnetic properties and softer matrices—implies that the beak formation happens simultaneously with a jump-like overall elongation of the former sphere, and this transformation resembles the first-order transition pattern. Upon assessing the chances to observe the predicted effects on the samples of now existing MAEs, one comes to a conclusion that the second scenario is hardly possible, whereas the first one, i.e., beak formation without hysteretic stretching, is much more realizable.

1. Introduction
Magnetoactive elastomers (MAEs) is a term for a family of composite materials which are weakly-linked polymer matrices filled with micron-size ferromagnet/ferrite particles. MAEs demonstrate macroscopically significant shape and force response to applied magnetic fields and, reciprocally, considerable changes of magnetic properties under field stimulation and mechanical loads. In the literature, one can find a number of examples of MAEs employed as active elements of micro-technique devices and medical appliances: field-controlled metamaterials for acoustics [1], microfluidic transportation systems [2, 3], remotely controlled grippers and a variety of microrobots [4, 5, 6, 7]. All that points out the importance of fundamental understanding of physics and mechanics which underlay the functional properties of these composites.

An essential and well known feature of MAEs is magnetostriction, i.e., the ability of a sample to change its shape when subjected to a uniform magnetic field. In terms of continuum media electrodynamics, the magnetostriction of MAEs is due to the tendency to reduce the internal demagnetizing field by way of reshaping a deformable sample, in the course of which substantial portions of the material move with respect to each other. Provided that spatial distribution of the filler particles inside each macroscopically small (but containing many micron-size grains)
element remains the same, this type of magnetostriction may be adequately modelled in the framework of a continuum theory.

The problem that we study here, is a large-scale deformation of a spherical sample of MAE originating from its magnetostriction. Initially, the problem has been inspired by its analogue in the hydrostatics of dense ferrofluids. Since 1980s, it is known [8, 9, 10, 11] that a spherical droplet of a dense ferrofluid subjected to a uniform magnetic field \( H_0 \) stretches along the direction of the latter. As the field strength grows, this elongation, albeit monotonic, is not smooth. Until the field attains some threshold \( H_0^* \), the drop deforms just slightly but at \( H_0^* \) its elongation \( \varepsilon \) enhances stepwise and after that goes on with much greater rate \( d\varepsilon/dH_0 \). Another specific feature of that effect is that below \( H_0^* \) the droplet shape does not differ much from an ellipsoid of revolution (spheroid) whereas above the threshold the shape becomes spindle-like, i.e., resembles a rounded body with tapered tips. It is now well understood that the transition undergone by the droplet results from instability caused by competing magnetostatic and surface tension forces.

It is easy to see that there exist remarkable magnetomechanical similarities between MAEs and ferrofluids: both substances are able to magnetize unhysteretically to quite high levels and are easily deformable. If for ferrofluids the deformability is virtually infinite, sufficiently soft MAEs are as well very labile and able to stretch by hundreds of percent. This suggests an idea that the field-induced striction of an MAE sphere should be and large follow the same hysteretic deformation scenario as a ferrofluid droplet. On the other hand, the particular differences between ferrofluid and MAEs are clear as well. First, the magnetism of a ferrofluid is due to ferrite (e.g., magnetite) nanoparticles whereas the magnetic sensitivity of a standard MAE is ensured by the presence of micron size particles of a low-coercive ferromagnet (carbonyl iron). This implies that the magnetic susceptibility of a typical MAEs is much greater. Another fundamental difference is that the integrity of a ferrofluid droplet is due to the interface tension that acts at the border of the droplet with the liquid it is floating in. In case of an MAE, the sample integrity is ensured by the fact that its body consists of chemically linked macromolecules whereas surface tension plays a minor role. Therefore, the forces, which restore the initial shape of the object after the field turn-off, have different origin: surficial for droplets and bulk for MAE species. Accordingly, the internal mechanics of the objects is substantially different: whereas inside a ferrofluid droplet only isotropic stresses might exist, the deformation state of an MAE body establishes as a result of joint action of isotropic (pressure) and shear stresses. The above-mentioned essential structure differences give to MAEs a big advantage from the viewpoint of observability of the effect. If assess the prospects of obtaining samples with the size convenient for measurements or applications, one finds out that MAE spheres may be manufactured with any diameter ranging from millimeters to centimeters, meanwhile the size of ferrofluid droplets prone to instability cannot exceed 20 \( \mu \)m [10, 11].

The story of studies of the magnetostriction-induced shape transformations of an MAE sphere is not that long and, hopefully, have good prospects. First, we remark the experimental aspect of the problem. Until nowadays, no observations of an MAE sphere stretching, apart from those where this deformation was small and well described by the spheroid approximation, have been reported. In our view, this lack of effort was entailed by the fact that the formerly existing theoretical estimates were completely disappointing: the required levels of magnetic susceptibility were far beyond any reasonable values for any conceivable MAEs. The cause of that is now clear: all those estimates had been obtained on the basis of the spheroidal model, which assumes that the stress and strain fields inside a deformed MAE sphere are uniform. Meanwhile, yet in Ref. [12] it had been shown that even an infinitesimal applied field induces in a sphere made of a magnetizable continuum a non-uniform distribution of stresses and, thus, non-uniform deformations. In Ref. [13] we have discarded the uniformity condition and, upon performing finite-element numerical modelling, came to conclusion that the real requirement on the magnetic susceptibility \( \chi \) of an MAE is, in fact, about two orders of magnitude lower. In other
words, our prediction implied that the hysteretic striction effect, very similar to that observed in ferrofluids, should be observable with the spheres of typical MAEs, e.g. polydimethylsiloxane matrices filled with carbonyl iron micropowders with the volume content of the latter above 25% [14, 15, 16].

The present paper advances Ref. [13] in one important aspect. Namely, in the preceding work, to make the explanations more straight and comprehensible, we have assumed that the model MAE is a linearly magnetizable medium, i.e., the acquired magnetization $M$ is proportional to the internal field $H$ as $M = \chi H$, where susceptibility $\chi$ is a scalar constant. It is evident, however, that the magnetization of any real MAE saturates at sufficiently high fields. In turn, this means that a model that uses an unlimited linear law overestimates the magnetic forces at enhanced fields, which makes it necessary to revise the consideration of Ref. [13] in a more accurate way. Therefore, the subject of our study is to find out whether the magnetomechanical hysteresis of an MAE sphere is expectable in a real situation and, if the saturation prevents this scenario in full, what specific ‘traces’ of it could be discerned in the measurements.

2. Deformation of MAE sphere under a uniform field
2.1. Finite strain approach

The object under consideration is a free-standing sphere of radius $R$ made of an MAE that is taken to be made of a magnetizable deformable continuum. Anticipating large shape changes, the mechanical part of the problem is formulated in terms of finite strains. For that, two configurations are introduced: the initial and actual ones, so that to the radius-vector $r$ defined in the initial configuration corresponds the radius-vector $R = r + u$ in the actual configuration, here $u$ is displacement vector. The basis vectors are defined as $\epsilon_i = \partial r/\partial q_i$ and $\hat{\epsilon}_i = \partial R/\partial q_i$ in the initial and actual configurations, respectively; here $q_i$ are generalized coordinates. Hamilton operators in the initial and actual configurations are introduced as

$$\hat{\nabla} = \hat{\epsilon}_i \partial/\partial q_i$$

where $\epsilon_i$ and $\hat{\epsilon}_i$ are the reciprocal basis vectors. Introducing fundamental kinematic function—deformation gradient—as

$$F = (\nabla R)^T = \hat{\epsilon}_i \epsilon^i = g + \nabla u^T,$$

where $g$ is metric (unit) tensor and index $T$ denotes transposition, for the inverse function one has

$$F^{-1} = (\hat{\nabla} r)^T = \epsilon_i \hat{\epsilon}^i = g - \hat{\nabla} u^T.$$  \hspace{1cm} (2)

In these terms, the Hamilton operators in initial and actual configurations are related to each other as

$$\hat{\nabla} = F^{-T} \cdot \nabla,$$  \hspace{1cm} (3)

where $F^{-T} = (F^T)^{-1}$.

We present the magnetic field as $H = H_0 - \hat{\nabla} \psi$, where $\psi$ is scalar magnetic potential, $H_0$ the external field. Following Ref. [17], the total energy of a magnetoelastic medium in the absence of currents we cast in the form

$$U = \int_{\Omega_{\text{sm}}} \rho \phi(F, H) dV - \frac{1}{8\pi} \int_{\Omega} H^2 dV$$  \hspace{1cm} (4)

with $\rho$ being the density of the medium in actual configuration; here $\Omega$ is the whole calculation region in actual configuration and $\Omega_{\text{sm}}$ the region occupied by the sample in actual configuration. In equation (4) $\phi(F, H)$ is the density of magnetoelastic energy, via which the magnetization and Cauchy stress tensor are expressed as

$$M = -\rho \frac{\partial \phi}{\partial H}, \hspace{0.5cm} T = \rho F \frac{\partial \phi}{\partial F}.$$  \hspace{1cm} (5)
Using the Jacobian of deformation gradient tensor $J = \det(F)$, the density and the element of volume may be transformed from the initial to actual configuration as $\rho = J^{-1}\rho_0$ and $dV = JdV_0$; here subscript 0 denotes the initial state. In these terms, energy (4) takes the form

$$U = \int_{\Omega_{sm}} \rho_0 \phi(F, H) dV_0 - \frac{1}{8\pi} \int_{\Omega'} J H^2 dV_0,$$

(6)

where $\Omega'$ and $\Omega_{sm}$ stand, respectively, for the full calculation region and the region occupied by the sample in the initial configuration. The Hamilton operator enables one as well to transform the magnetic field to the initial configuration as $H = H_0 - F^{-1} \cdot \nabla \psi$.

Upon splitting the magnetoelastic energy density in a sum of magnetic and elastic parts as $\phi = W_{el}/\rho_0 + W_{mag}/\rho$, energy (6) takes the form

$$U = \int_{\Omega_{sm}} (W_{el} + JW_{mag}) dV_0 - \frac{1}{8\pi} \int_{\Omega'} J H^2 dV_0,$$

(8)

To describe the mechanical behavior of an MAE in a realistic way, we use the Peng-Landel elastic potential [18] that is known to be well appropriate for slightly compressible elastomers at large strains:

$$W_{el} = \frac{1}{2} G \left[ J^{-2/3} \text{Tr}(F^T \cdot F) - 3 \right] + \frac{1}{2} K (J - 1)^2,$$

(9)

where $G$ is the initial shear modulus, and we set $K = 500G$.

The magnetic part of the energy density of a magnetizable continuum is rendered by the integral

$$W_{mag} = - \int_0^H M(H') dH',$$

(10)

where we assume that magnetization of the model MAE obeys the Fröhlich-Kennelly relation [19]:

$$M(H) = \frac{\chi_0 M_s}{M_s + \chi_0 H},$$

(11)

with a constant isotropic susceptibility $\chi_0$ and saturation magnetization $M_s$. Given that, the magnetic energy density writes

$$W_{mag} = - \frac{M_s}{\chi_0} \left[ H \chi_0 + M_s \ln(M_s) - M_s \ln(H \chi_0 + M_s) \right].$$

(12)

To maintain continuity of the deformation gradient $F$ everywhere in $\Omega$, we ascribe to the space around the sample an elastic potential in the same form as (9) but with modulus $G_s$ that is several orders of magnitude lower than $G$ to make a particular value of $G_s$ irrelevant for final results. Under those conditions, the elastic energy of the system acquires an additional contribution

$$U'_{el} = k_s \int_{\Omega'_{sp}} W_{el} dV_0,$$

(13)

where $\Omega'_{sp} = \Omega' \setminus \Omega_{sm}$ is the space region around the sample, and $k_s = G_s/G = 10^{-5}$. Combining equation (13) with (8), one arrives at the expression for the magnetoelastic energy of the system:

$$U(\nabla u, \nabla \psi) = - \frac{1}{8\pi} \int_{\Omega'} H^2 \cdot J dV_0 + k_s \int_{\Omega'_{sp}} W_{el} dV_0 + \int_{\Omega_{sm}} (W_{el} + JW_{mag}) dV_0,$$

(14)

whose minimum determines the equilibrium shape of the MAE sample under a given field.

For the problem under study, the general form of variational equation to be solved is

$$\delta U = \frac{\partial U}{\partial (\nabla u)} \cdot (\nabla \delta u)^T + \frac{\partial U}{\nabla \psi} \nabla \delta \psi = 0.$$

(15)
3. Numerical modelling
The problem under study has evident axial symmetry around the direction of applied field. Accordingly, a cylindrical coordinate frame \((\rho, z, \varphi)\) is introduced with the polar axis along \(H_0\). Making use of the symmetry, we consider only the region of space that abuts on the quarter (1st quadrant) of the circle of radius \(10R\) whose plane is perpendicular to \(Oz\) and is centered at the coordinate origin, the outer border of this region is denoted as \(\Gamma\).

Equation (15) is solved numerically by the finite-element method realized in the algorithms of \textsc{FEniCS} computing platform [20, 21]. The employed mesh is nonuniform, it is most dense at the central part of \(\Omega\) gradually becoming more sparse when approaching \(\Gamma\). Two functions: \(u(\rho, z)\) and \(\psi(\rho, z)\) defined in a mixed finite-element space \((P_3, P_1)\) inside \(\Omega\), are evaluated under boundary conditions
\[
 u_{\rho}|_{\rho=0} = 0, \quad u_z|_{z=0} = 0; \quad \psi = 0 \text{ on } \Gamma.
\] (16)

The strength of applied field \(H_0\) is varied gradually in small steps. Equation (15) is solved anew for each value of \(H_0\) with boundary conditions (16) on the adopted mesh by the Newton method (implemented in \textsc{FEniCS} platform); the values of \(u(\rho, z)\) and \(\psi(\rho, z)\) obtained in result of an actual calculation step are taken as initial ones when commencing the next step.

4. Shape transformations of a magnetized MAE sphere
As it is shown in Ref. [13], under enhanced fields, the rounded (quasi-spheroidal) shape of a deformed sphere changes to a spindle-like one. That means that in the vicinity of the poles—locations where the field \(H_0\) is normal to the body surface—the MAE object grows tapered bulges (‘beaks’). In these parts the sign of the curvature of the meridional cross-section is inverted in comparison with that of the middle section of the body. The transition between the curvatures of opposite signs, i.e., the turn-up of a beak is signalled by the change of sign of the second derivative \(z''(\rho)\) where \(Oz\) axis points along the field vector, see Fig. 1.

![Figure 1. Meridional cross-section of a spindle-like MAE object; points show the calculated contour; solid line renders the second derivative of the contour, the value of \(\rho\) at the point where this line crosses the abscissa axis defines the radial size of the beak of length \(h\).](image-url)
We define the geometry characteristics of the beak as follows. The numerical calculation performed according the procedures described in Section 3 yields the contour $\rho(z)$ that describes the meridional cross-section of the deformed MAE. After inverting this function, i.e., transforming it to $z(\rho)$ and using numerical differentiation, its second derivative is evaluated and then the root of equation $z''(\rho_b) = 0$ is found. The value of $\rho_b$ renders the radial size of the beak whereas its length is defined as $h = z(0) - z(\rho_b)$. The overall field-induced elongation of the MAE body under field $H_0$ is rendered by function $u_z(0)\mid_{h_0}$, that is the $z$-component of displacement vector $u$; below, for graphic representation, we use these geometry parameters in normalized forms: $h/R$ and $u_z(0)/R$. In Fig. 2 these characteristics are shown in pairs for three sequentially increasing values of the initial susceptibility of an MAE, see Eq. (11) where $\chi_0$ is defined. The abscissa axes are scaled in nondimensional field $H_0 = H_0/\sqrt{G}$, the saturation magnetization is expressed in $M_s = M_s/\sqrt{G}$ units. To make Fig. 2 more comprehensible, the curves corresponding to quite a large value of the magnetic saturation: $M_s = 6$ are chosen.

As seen, the beak emerges at a finite value of the applied field strength. At low $\chi_0$—the case illustrated by Fig. 2a—the bifurcation of $h(H_0)$ is ‘soft’, i.e., tapering of the MAE body ends occurs following the second-order transition scenario. In the overall deformation it is reflected as some quite moderate enhancement of the $\varepsilon$. The case presented in Fig. 2b corresponds to a higher $\chi_0$, its value is just above the tricritical point. The plot shows that the stretching transition changes its type: from the second to first, i.e., from ‘soft’ to ‘rigid’ (stepwise) onset of the beak; and this regime to a greater extent manifests itself in the overall elongation.

5. Discussion
In Ref. [13] dimensional estimates are given which establish that the anticipated values of the susceptibility—if to consider typical now existing MAEs—are not higher than $\chi_0 \approx 0.4$ [CGS units]. It is instructive, however, to add to Fig. 2 a case where the above-mentioned limiting value is exceeded, see Fig. 2c. This illustration makes the sequence full showing the developed first-type transition: at $\chi_0 = 1$ the bistability region is clearly resolved: the field strength, at which the transition occurs under an increase of the field, is substantially larger than that, at which the MAE object shrinks back under the field diminution. Note that the boundaries of the bistability regions for the beak and for overall elongation virtually coincide that means that, provided the jump-like stretching takes place, it is inseparable from formation of a beak.

Let us compare the here-obtained results with those of Ref. [13]. There the prime issue of investigation was the first-order magnetodeformational transition, i.e., the elongation jump. Concerning the beak, it has been just mentioned that at low values of $\chi$ (in the former treatment the magnetization law was linear) the beak appeared before the elongation jump, if any. Moreover, the threshold scenario of the beak formation has not been revealed.

Now one sees that in a sphere whose magnetization saturates under field, depending on the material parameters of the MAE either one or two transitions might occur. The first one is manifested as a beak formation, is indicated by inversion of sign of the meridional cross-section curvature. At sufficiently low $\chi_0$ the beak emergence and growth evolve as a second-order transition, and is not accompanied by a deformation jump, see Fig. 2a.

Around $\chi_0 = 0.4$ that is, according to our estimates, the highest conceivable value for real MAEs, the peak formation scenario changes from the second-order to the first-order one. In this situation, the transition occurs just above the tricritical point, and because of that the interval of bistability of the peak length $h$ is so narrow that it does not resolve in Fig. 2b. An important conclusion follows, however, from comparison of the numerical evidence rendered by the plots of Fig. 2b. It is seen that as soon as the peak onset becomes first-order, the same applies to the sample elongation as a whole. Furthermore, this inference is supported by the plots of Fig. 2c, which prove that the bistability intervals for the peak length and the overall elongation coincide within the accuracy of numerical results.
As the initial magnetic susceptibility $\chi_0$ of an MAE is directly proportional to the amount of ferromagnetic filler it contains, one arrives at the following conclusions. A sphere of extremely low concentrated MAE ($\chi_0 \ll 0.2$) is expected to demonstrate but tiny elongation $a$ in that smoothly depends on the applied field strength, and its surface always remains entirely convex.
A sphere of an MAE with a moderate amount of filler ($\chi_0 \sim 0.2$) displays peak formation that evolves along the second-order transition scenario: with respect to overall elongation, the effect is notable but not the governing one. The jump-like elongation regime begins with ($\chi_0 \sim 0.4$), where both the peak length and overall elongation enhance coherently and in the first-order transition way.

Figure 3. Dependence of the dimensionless beak length on the applied field strength for $\chi_0 = 0.4$; saturation magnetization of an MAE is: $\tilde{M}_s = 6$ (1), 3 (2), 2 (3), 1.5 (4).

Fig. 3 shows the dependence of the peak length on the applied field strength. All the plots correspond to $\chi_0 = 0.4$ as this is the largest value that is reasonable to assume for now existing MAEs. As seen, in the considered field range, the peak length grows monotonically with the field strength whatever $\tilde{M}_s$. The numerical evidence of Fig. 3 enables one to draw two important easily understandable conclusions. First, the stronger the field the lower is the beak growth rate $dh/d\tilde{H}_2^0$ that is a direct consequence of magnetic saturation: at yet higher fields the beak would finally attain a maximal constant value. Second, at any given field, the beak emerges the earlier and its length the greater, the larger saturation magnetization $M_s$. Indeed, apart from very low field range, it is $M_s$ that defines the intensity of the magnetostatic forces which induce the instability of a formerly convex MAE surface. The dependence of the beak nucleation field on $M_s$ is shown in Fig. 4.

Finally, let us estimate what are the dimensional material parameters of a generic MAE on which the above-described hysteretic magnetodeformational effects may be observed. As mentioned, the initial magnetic susceptibility $\chi_0$ of existing MAEs does not exceed 0.4. If to set the volume fraction of carbonyl iron to $\phi \approx 30$ vol.%, with the saturation magnetization of this ferromagnet $M_S \approx 1.5 \cdot 10^4$ emu/cm$^3$, that gives for the test MAE $M_s \approx \phi M_S \approx 450$ emu/cm$^3$. To be real, this value should be reduced to not greater than $M_s \approx 300$ emu/cm$^3$. If to anticipate a very strong manifestation of the effect (full-scale first-order transition), one needs, for example, $\tilde{M}_s \approx 3$, see Fig. 3. From that, the estimate for shear modulus of the composite is $G \approx (M_s/\tilde{M}_s)^2 \approx 10^6$ dyn/cm$^2 = 1$ kPa that is a very soft material. On the other hand, the critical field of onset of this magnetomechanical transition is—see Fig. 4—$H_0 \approx \sqrt{17} \cdot 10^2 \approx 400$ Oe, i.e., quite low.

The above-obtained estimate requires a shear modulus about 1 kPa, and thus makes it clear that the conclusions of Ref. [13] were excessively optimistic. In other words, one may not anticipate that in the samples of customary MAEs ($G \sim 10/100$ kPa) the hysteretic magnetostriction could be made fully observable. Apparently, due to magnetization saturation,
Figure 4. Critical field of the beak onset as a function of saturation magnetization for MAEs with $\chi_0 = 0.4$.

the magnetostatic forces cannot grow enough to compete with the elastic ones (to generate a jump-like deformation) whatever the field.

This, however, does not entirely forbid the possibility to observe the ‘precursor’ of the hysteretic elongation—the beak onset according to the second-order transition scenario—on much stiffer MAEs. Indeed, setting $M_s \simeq 1$, one arrives at the estimate $G \simeq 10$ kPa that brings the effect within the range of available MAEs. The field strength that ensures this onset under these condition is $H_0 \simeq 1.5$ kOe.

Acknowledgments
The study was performed with the use of the ICMM UB RAS supercomputer Triton.

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