# Single-particle mechanism of magnetostriction in magnetoactive elastomers

Viktor M. Kalita,[1,2] Andrei A. Snarskii,[1,3] Denis Zorinets,[1] and Mikhail Shamonin[4,*]

[1]*National Technical University of Ukraine "Kyiv Polytechnic Institute," Prospekt Peremohy 37, Kiev 03056, Ukraine*

[2]*Institute of Physics NAS of Ukraine, Prospekt Nauky 46, Kiev 03028, Ukraine*

[3]*Institute for Information Recording NAS of Ukraine, Shpaka Street 2, 03113 Kiev, Ukraine*

[4]*East Bavarian Centre for Intelligent Materials (EBACIM), Ostbayerische Technische Hochschule Regensburg, Prüfeninger Strasse 58, 93049 Regensburg, Germany*

(Received 24 March 2016; revised manuscript received 27 May 2016; published 28 June 2016)

Magnetoactive elastomers (MAEs) are composite materials comprised of micrometer-sized ferromagnetic particles in a nonmagnetic elastomer matrix. A single-particle mechanism of magnetostriction in MAEs, assuming the rotation of a soft magnetic, mechanically rigid particle with uniaxial magnetic anisotropy in magnetic fields is identified and considered theoretically within the framework of an alternative model. In this mechanism, the total magnetic anisotropy energy of the filling particles in the matrix is the sum over single particles. Matrix displacements in the vicinity of the particle and the resulting direction of the magnetization vector are calculated. The effect of matrix deformation is pronounced well if the magnetic anisotropy coefficient $K$ is much larger than the shear modulus $\mu$ of the elastic matrix. The feasibility of the proposed magnetostriction mechanism in soft magnetoactive elastomers and gels is elucidated. The magnetic-field-induced internal stresses in the matrix lead to effects of magnetodeformation and may increase the elastic moduli of these composite materials.

DOI: 10.1103/PhysRevE.93.062503

## I. INTRODUCTION

Magnetoactive elastomers (MAEs) consist of micrometer-sized ferromagnetic particles embedded into a nonmagnetic soft elastomer matrix [1]. Since the resulting composite material is ferromagnetic, an associated physical phenomenon, the so-called magnetostriction (MS) is observed in MAEs [2–6]. Obviously, an externally applied magnetic field can cause MS of filling particles, but this effect is very small (field-induced strain $\sim 10^{-6} - 10^{-5}$) and can be neglected in comparison with the observed deformations ($\sim 10^{-2} - 10^{-1}$). It should be noted that in conventional magnetic materials there are two main mechanisms of MS, namely, the interparticle MS and the single-ion MS [7–15]. The interparticle MS is associated with the dependence of the spin-interaction parameters on the mutual arrangement of the atoms, and the single-ion MS is related to the field distortion of the ligands in the spin state of the ions. In MAEs there also exists a possible MS mechanism associated with the interparticle interactions. For example, elongation of the MAE sample along the magnetic field decreases the magnetostatic energy related to the interparticle magnetic dipole-dipole interactions [16–20]. Such a MS can reach tens of percent [16–20].

Alternatively, the change of the sample magnetic susceptibility due to the rearrangement of ferromagnetic particles in external magnetic fields can stimulate either contraction or elongation of the sample [21,22]. It has been theoretically shown that samples with a high concentration of particles should elongate [22]. A comprehensive review of MS in MAEs and its relation to the field-stiffening (magnetorheological) effect can be found in Ref. [23]. The physical phenomenon of a distortion of the polymer matrix due to the transmission of torques from magnetic particles to the polymer matrix has been previously analyzed using microscopic models, corresponding to different experimental situations. Galipeau and Ponte Castañeda [5] considered MS of composites consisting of aligned, ellipsoidal magnetic particles distributed randomly with "ellipsoidal" symmetry under combined magnetic and mechanical loading. This model captures the coupling between the magnetic and mechanical fields, including the effects of magnetic saturation. Based on these considerations, a new class of MAEs with doubly layered, herringbone-type microstructures capable of generating large field-induced strains of up to 100% has been proposed [6]. Weeber *et al.* [24–26] considered ferrogels comprising embedded magnetic nanoparticles with the size of about 10 nm. These particles had a permanent magnetic moment and reacted superparamagnetically to an external magnetic field. These works did not treat the polymer matrix as a continuous medium but explicitly included polymer chains bound to specific spots on the surface of nanoparticles.

In the present work, a single-particle mechanism of MS in MAEs, assuming the rotation of a magnetized soft magnetic particle in a magnetic field, will be considered using an alternative theoretical model, which can be solved analytically for the experimental situation of particular interest. In the proposed single-particle mechanism of MS, the mechanical stress, generated in the matrix by the ferromagnetic particle under an external magnetic force, is localized and related only to the surroundings of this particle. If the dipole-dipole interactions are taken into account, the resulting mechanical stresses are coherently generated in the sample as a result of particle interactions; see, e.g., Ref. [27].

By scrutinizing the single-particle mechanism of MS, we will show that in an external magnetic field two processes minimizing the magnetic energy occur: (i) rotation of the particle's magnetic moment away from its easy axis and (ii) rotation of the particle's easy axis. It will be demonstrated that, if the matrix is sufficiently soft, then the magnetic moment of the particle is reoriented due to the rotation of the particle, rather than rotation of the magnetic moment vector. Rotation of the particle is accompanied by deformation of the surrounding

*Corresponding author: mikhail.chamonine@oth-regensburg.de

 

matrix. However, the theoretical problem of finding the equilibrium orientation of the particle's magnetic moment and the direction of its easy magnetization axis is not limited to the calculation of the elastic deformation of the matrix due to the rotation of the particle. The formulated physical problem is thermodynamic by nature. The calculation of the magnetic state of the particle and the elastic deformation of the matrix due to the interaction of the magnetic moment with the external magnetic field involves determination of the equilibrium orientation of the particle's magnetic moment consistent with the matrix deformation. Therefore, we will study the phenomenon of single-particle MS in MAEs employing an alternative theoretical approach. It is emphasized that in our model the total magnetic anisotropy energy of the filling particles in the matrix is the sum over single particles. The interparticle MS is associated with dipole magnetic interactions. Therefore, it is generated by interactions described by a sum of dipole-dipole interactions over all pairs of particles. Dipole-dipole interactions are anisotropic, since they are dependent on the positions of the filling magnetic particles and spatial orientation of induced magnetic moments, for example, if the magnetic particles are aligned in chain aggregates. Obviously, when considering such an interparticle MS it is not possible to separate one particle from the totality of particle pairs. In the case of single-particle MS, consideration of a single particle is justified approximation. Therefore, for the physical description of a single-particle MS mechanism a soft magnetic particle embedded into the elastomer matrix will be considered. The shape of the particle and its arrangement in the matrix will be chosen in such a way that the underlying physics will be not unnecessarily complicated by mathematical transformations. The calculation of deformations should remain simple. The magnetic interactions should not be complicated by magnetostatic corrections associated with the shape of particles. Such conditions are well fulfilled with magnetic particles in the form of a disk (platelet).

## II. MODEL

In most experiments, the micrometer-sized magnetic particles are of nearly spherical shape and the particle diameter is much larger than the single-domain critical size. Therefore, the demagnetizing field must be considered for the magnetization of the particle. In general, the demagnetization field generates an inhomogeneous magnetic state in the particle. This complicates the calculations of the anisotropy energy and the magnetic moment of the particle. To avoid the influence of demagnetization, let us consider a disk-shaped ferromagnetic particle with the radius $r_0$ being much larger than the disk thickness $D_0 \ll r_0$. The particle is placed into the magnetic field parallel to the plane of the disk. The particle is embedded into the elastic film of the same thickness $D_0$ (Fig. 1). Models with disk-shaped particles are often considered in statistical mechanics [28–32].

Let the easy magnetization axis $L$ be in the plane of the particle. $\varphi_H$ denotes the angle between the easy magnetization axis and the external magnetic field (see Fig. 2).

Since the particle is magnetically anisotropic (there exists the easy magnetization axis), in the external magnetic field $\boldsymbol{H}$ not parallel to $L$, the direction of the magnetic moment $\boldsymbol{M}$ will not coincide with the direction of $\boldsymbol{H}$. Denote the resulting angle between $\boldsymbol{M}$ and $L$ as $\varphi_M$.

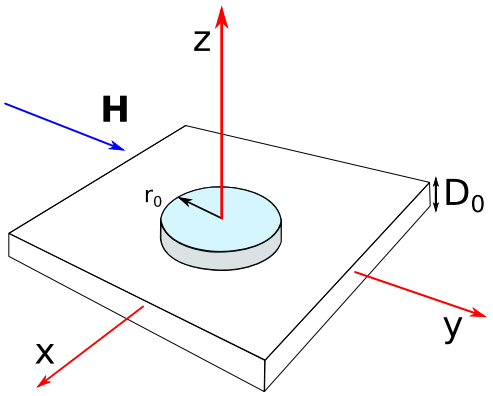


FIG. 1. Soft magnetic disk-shaped particle in an elastic matrix. $\boldsymbol{H}$ denotes the external magnetic field strength. The thickness of the disk is equal to the thickness of the elastic film.

Hence there will be the nonvanishing torque acting on the magnetic moment $\boldsymbol{M}$ and proportional to $\boldsymbol{M} \times \boldsymbol{H}$. In the static case, this torque is compensated by elastic stresses arising in the matrix due to rotation of the particle.

Since the particle is considered to be rigid (the elastic modulus of the particle is much larger than that of the elastomer matrix), the rotation of the matrix will be determined by the rotation angle $\gamma$ of the easy magnetization axis (cf. Fig. 2).

The directions of the external magnetic field $\boldsymbol{H}$, the easy magnetization axis $L$ in the absence of the field ($\boldsymbol{H} = 0$), the easy magnetization axis $L'$ in the presence of the field ($\boldsymbol{H} \neq 0$), and the magnetic moment $\boldsymbol{M}$ of the particle are shown.

In the field $\boldsymbol{H}$, the energy of the "magnetic particle–deformed matrix" system can be written as the sum of three terms,

$$E = E_A + E_H + E_{\mathrm{el}}, \tag{1}$$

where $E_A$ is the magnetic anisotropy energy of the easy-axis type, $E_H$ is the energy of the magnetic moment in the field $\boldsymbol{H}$, and $E_{\mathrm{el}}$ is the energy of elastic deformation of the matrix. The magnetic anisotropy energy is written similarly to [33], and in

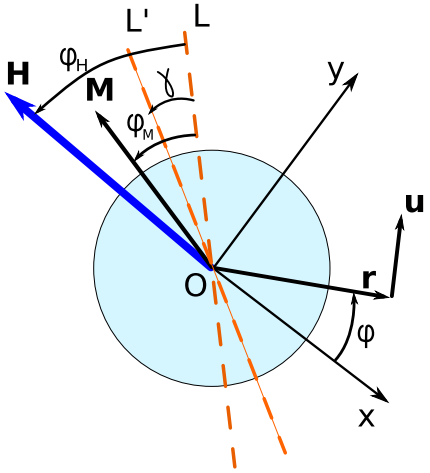


FIG. 2. Geometry of the problem.

our case it will have the following form:

$$E_A = -\tfrac{1}{2}K\cos^2(\varphi_M - \gamma)\pi r_0^2 D_0, \tag{2}$$

where $K$ is the magnetic anisotropy constant, whose value is given by the product of the anisotropy field $H_A$ and the magnetization $m$: $K = mH_A$ and $M = m\pi r_0^2 D_0$.

The energy $E_H$ can be written as

$$E_H = -Hm\cos(\varphi_H - \varphi_M)\pi r_0^2 D_0, \tag{3}$$

where $H = |\boldsymbol{H}|$.

Since the rotation of the particle causes the inhomogeneous deformation of the matrix, the determination of $E_{\rm el}$ requires the solution to the elasticity problem of distribution of deformation in the matrix with the given rotation angle of the particle $\gamma$.

The proposed microscopic model explicitly includes the influence of uniaxial magnetic anisotropy and its interplay with the linear elasticity.

## III. CALCULATION OF ELASTIC DEFORMATIONS

For linear and isotropic elastic medium, the equilibrium equations have the following form [34]:

$$(\lambda + 2\mu)\,\mathbf{grad}\ \mathrm{div}\mathbf{u} - \mu\,\mathbf{curl}\,\mathbf{curl}\,\mathbf{u} = 0, \tag{4}$$

where $\lambda$ and $\mu$ are the Lamé parameters and $\mathbf{u}$ is the displacement vector. In the context of elasticity, $\mu$ is called the shear modulus.

The boundary conditions for the displacement vector $\mathbf{u}$ are

$$u_\varphi|_{r=r_0} = r_0\gamma, \quad u_r|_{r=r_0} = 0, \tag{5}$$

$$u_\varphi|_{r=\infty} = 0, \quad u_r|_{r=\infty} = 0. \tag{6}$$

The condition (6) means that the matrix as a whole does not rotate, except in the vicinity of the particle.

According to the symmetry expressed in the boundary conditions, the partial derivative of the displacement component $u_\varphi$ on the angular coordinate $\varphi$ vanishes: $\partial u_\varphi/\partial\varphi = 0$. The radial projection of displacement is also zero: $u_r = 0$. This condition corresponds to the volume conservation of the elastomer surrounding the particle under deformations given by boundary conditions (5) and (6). Now Eq. (4) can be written in polar coordinates as follows:

$$\frac{1}{r}\frac{\partial(ru_\varphi)}{\partial r} = a, \tag{7}$$

where $a$ is a constant value.

After integration of (7), the constants of integration can be found from the boundary conditions. It turns out that $a = 0$.

Solution of Eq. (4) with conditions (5) and (6) in the polar coordinate system has the following form:

$$u_\varphi = u_0\frac{r_0}{r}, \quad u_r = 0, \quad u_0 = r_0\gamma, \tag{8}$$

where $u_\varphi$ is the component of the displacement vector along the angular coordinate $\varphi$ in the point $\mathbf{r}$ (see Fig. 2).

The corresponding (nonvanishing) component of the strain tensor is

$$u_{r\varphi} = \frac{1}{2}\left(\frac{\partial u_\varphi}{\partial r} - \frac{u_\varphi}{r}\right) = -u_0\frac{r_0}{r^2}. \tag{9}$$

Taking into account (9), the expression for the elastic energy density of the matrix can be written in the following form:

$$e_{\rm el} = 2\mu u_{r\varphi}^2 = 2\mu u_0^2\frac{r_0^2}{r^4}. \tag{10}$$

The missing expression for the elastic energy [cf. Eq. (1)] is found by integration of Eq. (10):

$$E_{\rm el} = \int_V e_{\rm el}dV = 2\pi D_0\int_{r_0}^{\infty} 2\mu u_0^2\frac{r_0^2}{r^3}dr = 2\pi r_0^2 D_0\mu\gamma^2. \tag{11}$$

## IV. CALCULATION OF PHYSICAL QUANTITIES FOR SINGLE-PARTICLE MAGNETOSTRICTION

Now it is possible to determine how the magnitude of deformation depends on the magnetic field direction and its magnitude. We use the equilibrium conditions for the particle corresponding to the minimum of the total energy of the system,

$$\frac{\partial E}{\partial\varphi_M} = 0, \quad \frac{\partial E}{\partial\gamma} = 0. \tag{12}$$

By substituting the expressions for $E_A$ [Eq. (2)], $E_H$ [Eq. (3)], and $E_{\rm el}$ [Eq. (11)] into Eq. (12), we arrive at the following system of equations for $\varphi_M$ and $\gamma$:

$$\tfrac{1}{2}K\sin 2(\varphi_M - \gamma) - Hm\sin(\varphi_H - \varphi_M) = 0, \tag{13}$$

$$\tfrac{1}{2}K\sin 2(\varphi_M - \gamma) - 4\mu\gamma = 0. \tag{14}$$

The system of equations (13) and (14) can be solved numerically for $\varphi_M$ and $\gamma$ with given $\varphi_H$, $\mu$, and $K$.

Figure 3 shows the dependencies of angles $\varphi_M$ and $\gamma$ on the ratio $\mu/K$. The graphs are obtained for the following external magnetic fields: $H = 0.1H_A$, $0.5H_A$, $H_A$, $2H_A$, comparable with the anisotropy field and inclined at angle $\varphi_H = 0.01$ rad (i.e., slightly more than half of a degree). Recall that $H_A = K/m$, $\varphi_M$ is the rotation angle of magnetization vector and $\gamma$ is the rotation angle of the particle.

It is seen that if the matrix elasticity is predominant ($\mu/K \gg 1$), rotation of the particle is small and rotation angle $\gamma$ tends to zero. For large values of $\mu/K$, the vector of the magnetic moment just inclines in the direction of the external magnetic field. The corresponding graphs for the angle $\varphi_M$ in Fig. 3(b) asymptotically approach constant values:

$$\varphi_M\left(\frac{\mu}{K} \to \infty\right) = \frac{\varphi_H}{1 + (H_A/H)}. \tag{15}$$

If the magnetic anisotropy is dominating ($\mu/K \ll 1$), the rotation angle of the particle almost coincides with the angle $\gamma \to \varphi_H$. In this case, the single-particle MS is prevailing and the magnetic moment vector of the particle is oriented towards the field due to the rotation of the particle and $\varphi_M \to \varphi_H$.

Consider small deviations of easy magnetization axes from the direction of the external magnetic field $\boldsymbol{H}$: $\varphi_M \ll 1, \gamma \ll 1$, $\varphi_H \ll 1$. For small angles, the following formulas for angles

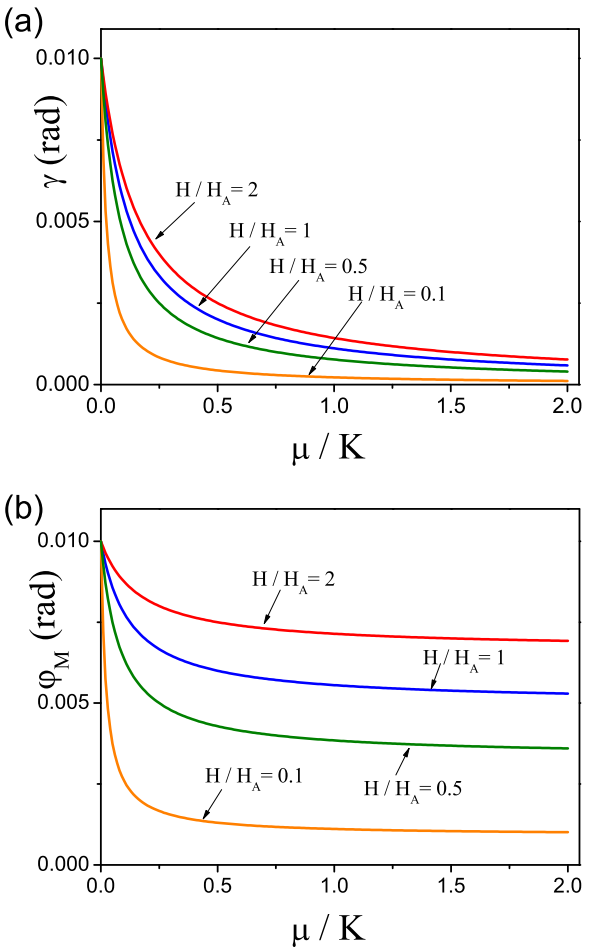


FIG. 3. Dependencies of angles $\gamma$ (a) and $\varphi_M$ (b) on the ratio of the elastic modulus and the anisotropy constant $\mu/K$.

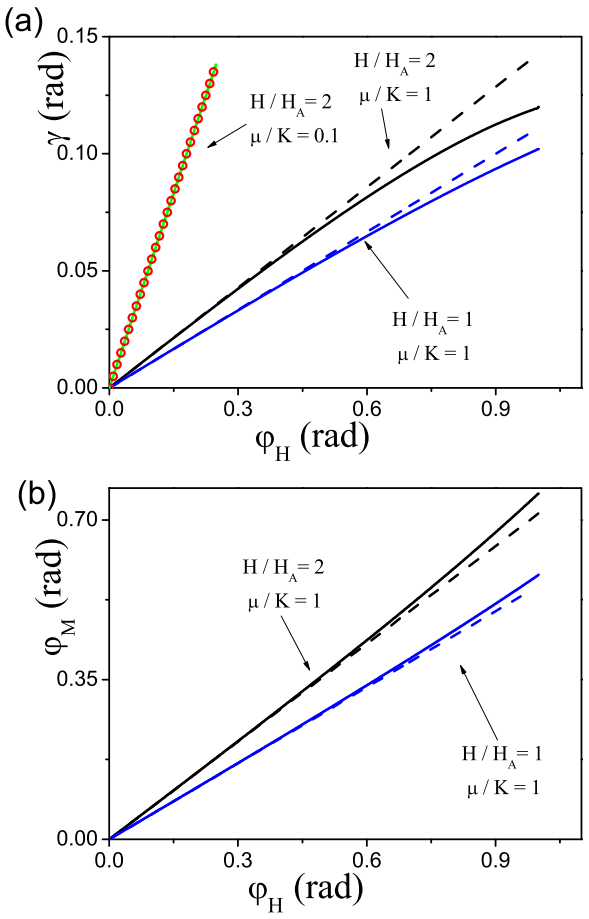


FIG. 4. Solutions for $\gamma$ (a) and $\varphi_M$ (b). Comparison of numerical (solid lines) and analytical (dashed lines or unfilled circles) solutions to the system of equations (13) and (14) in dependence on the angle $\varphi_H$ for different parameter sets.

$\varphi_M$ and $\gamma$ are easily obtained after some algebra:

$$\varphi_M = Hm\frac{K+4\mu}{KHm+4\mu(K+Hm)}\varphi_H, \tag{16}$$

$$\gamma = Hm\frac{K}{KHm+4\mu(K+Hm)}\varphi_H. \tag{17}$$

Accordingly to (17), the equilibrium component of the strain tensor will be given by

$$u_{r\varphi} = -Hm\frac{K}{KHm+4\mu(K+Hm)}\varphi_H\frac{r_0^2}{r^2}. \tag{18}$$

The obtained expressions (16) and (17) agree well with the numerical solutions given in Fig. 3. On the scale of the figure, they are indistinguishable from the curves shown in Fig. 3. Figure 4 compares some numerical solutions with approximate solutions (16) and (17). It can be concluded that approximate solutions work very well for $\varphi_H < 0.4$ rad in the wide range of parameters. Analytical results for $H/H_A = 2, \mu/K = 0.1$ are shown in Fig. 4(a) as unfilled circles, since otherwise they would be indistinguishable from the numerical solution. This set of parameters is not shown in Fig. 4(b) where it would overlap with other curves. From Eq. (18) it is seen that the magnitude of matrix deformation in high magnetic fields is determined by the ratio of the elastic modulus and the anisotropy constant. In high magnetic field, the limiting particle rotation angle is $\gamma = \varphi_H K/(K+4\mu)$. In this case for the soft matrix $\mu/K \ll 1$ the strain will be maximal and equal to

$$u_{r\varphi} = -\varphi_H\frac{r_0^2}{r^2}. \tag{19}$$

It follows from (18) that the magnitude of small matrix deformations in low fields $H \to 0$ is directly proportional to the magnitude of the external magnetic field. This is related to the fact that the magnetization of a magnetically soft disk along its easy magnetization axis, as in Fig. 2, leads to the saturation of magnetization in small magnetic fields. For a spherical particle, such a magnetization is impossible due to the action of the demagnetizing field. It is expected that for the spherical particle there will be no MS directly proportional to the small magnetic field.

## V. DISCUSSION

The condition $\mu/K \ll 1$ is essential for observation of single-particle MS. This condition is feasible in ultrasoft MAEs and gels, where $\mu \sim 10^3 - 10^4$ Pa can be realized

[35–38]. The room temperature magnetocrystalline anisotropy constant of Fe, Co, and Ni is one to two orders of magnitude larger: $K_1 \sim (0.5 - 50) \times 10^4$ J/m$^3$. Most of the experiments with MAEs employ almost spherical carbonyl iron particles. However, it is obvious that any deviation of the particle shape from the perfect sphere should lead to the appearance of the magnetic shape anisotropy since the demagnetizing field will not be equal for all directions, creating one or more easy axes. The value of uniaxial anisotropy $K$ is directly proportional to the difference $\Delta N$ between the largest and the smallest values of the demagnetizing tensor. Assume that the iron particle is magnetized to the saturation ($m = 1.72 \times 10^6$A/m $= 1720$ emu/cm$^3$). For a prolate, almost spherical ellipsoid with the ratio of half axes $a/b = 1.1$, it can be calculated that $\Delta N \approx 0.47$ (the sum of the eigenvalues of the demagnetizing tensor $N_a + 2N_b = 4\pi$) and $K \approx 138$ kJ/m$^3$. This corresponds to the anisotropy field $H_A \approx 63\,660$ A/m $=$ 800 Oe. If $a/b = 1.2$, the values of uniaxial anisotropy $K \approx$ 260 kJ/m$^3$ and anisotropy field $H_A \approx 119$ kA/m $= 1500$ Oe are almost two times larger. Similar considerations also arise in the simplified dipole-spring models of MAEs where the so-called orientation memory terms include possible orientation coupling of magnetic particles to the polymer network [39].

The magnetostriction in MAEs arising from the average effect of internal forces between the induced magnetic dipoles is proportional to the square of the particle concentration $c^2$. The magnetic torques on particles are induced through the direct interaction of them with the applied magnetic field. Therefore, their magnitude is of order $c$ [6]. This means that single-particle mechanisms can dominate for relatively small particle concentrations, but their effect on the magnetodeformation can be significant. Indeed, the numerical simulations in Ref. [24] revealed that magnetodeformation could be large (area shrinkage ~20%) although dipolar interactions were irrelevant for investigated systems.

Moreover, even the thin-film geometry with embedded cylindrical platelets shown in Fig. 1 is feasible, since the necessary technological prerequisites already exist. Ruiz *et al.* [40] fabricated structured elastomer films with thicknesses about 100 μm presenting piezo- and magnetoresistance. The films are composites of magnetite filler particles, which are both electrically conductive and magnetic, dispersed in an elastomeric matrix. Iannotti *et al.* [41] fabricated novel composite MAE samples with iron microparticles in the form of platelets with an average thickness of 7 $\mu$m and an average size of particles' major axis around 37 $\mu$m. The preferential in-plane magnetization of a single particle due to the shape anisotropy has been indeed achieved.

It is well known that MAEs display an extremely large but reversible increase in dynamic shear modulus upon application of external magnetic field [denoted as the field-stiffening or magnetorheological (MR) effect]. Similarly to MR fluids, the change in MR properties is believed to be due to the magnetic polarization induced in each particle by the external field, with the resulting interaction forces between the particles leading to the formation of elongated aggregates in the direction of the field [42–45]. The large increase in shear modulus is attributed to these magnetic-network aggregates [46–50]. However, complete understanding of the physics of this phenomenon in MAEs, including a comprehensive mathematical model, is still missing, in spite of significant theoretical efforts and progress achieved (see, e.g., [51,52]). In the above mentioned experiments, induced magnetization of magnetically soft MAE samples is practically parallel to the external magnetic field. Therefore, the analysis of experimental data leaves the impression that the magnetization of the sample does not create an additional mechanical moment on the particles, and, seemingly, the magnetic field is not involved in shear deformation. Note that such a large increase in the shear modulus has not been observed previously in the homogeneous magnetic crystals, even upon martensitic transitions [53,54].

We hypothesize that the large increase of the shear modulus $\mu$ in MAEs may have a contribution from the single-particle deformations caused by particle rotations in external magnetic fields. Indeed, the particle in a compliant matrix (see Fig. 1) can be easily rotated at an arbitrary angle $\gamma$ in the absence of magnetic field $H = 0$. If $H \neq 0$ and it is directed along the easy axis, the magnetic field will withstand the particle rotation. The deformation field will be set up around the particle, but its calculation will be the same as described above, if we put $\varphi_H = \gamma$. Rotation of the particle and the surrounding matrix in a strong magnetic field will require additional effort, when the shear modulus of the matrix is much less than the anisotropy constant of the particle material. A possible interpretation of the external observer is that the magnetic field leads to the local increase of elastic modulus in the vicinity of the particle, and the particle and the surrounding polymer become locally less sensitive to external influences under applied magnetic fields. In the case of a composite material, such a behavior will occur in the vicinity of each particle, resulting in an increase of "effective" elastic modules of the composite material. Full calculation of the field dependence of the effective shear modulus will be a subject of future publications. However, the above arguments make clear that the effective shear modulus will depend on the ratio of the constant of the particle's magnetic anisotropy and the shear modulus of the matrix, as well as on the concentration of particles.

It should also be noted that the field dependence of the rotation angle of spherical particles, surrounded by an elastomeric matrix in magnetic fields larger than the maximum demagnetizing field, will have the form of Eq. (17), but with slightly different numerical coefficients. This means that, qualitatively, our result for the deformation limit will be valid for spherical particles as well. However, it is expected that for spherical particles in small fields the influence of the demagnetizing field will lead to deviations to Eq. (17) for the dependence of the rotation angle on the field.

The calculation of the rotation angle of the magnetic moment and the particle rotation is based on the minimization of the total energy of the system. Such a minimum corresponds to the conditions of the mechanical equilibrium of the particle. Indeed, Eq. (13) corresponds to the vanishing sum of torques applied to the magnetic moment of the particle by the external magnetic field and the anisotropy field. Equation (14) corresponds to the equality of the total moment of force applied to the particle, both from the side of the magnetic subsystem and the elastically deformed matrix.

## VI. CONCLUSION

In this paper, a mechanism of magnetic-field-induced MS in mechanically soft MAEs is proposed and described mathematically for a simplified model. The mechanism is based on the rotation of a single soft magnetic particle with uniaxial magnetic anisotropy in external magnetic fields. The mechanism is denoted as single-particle MS because the total magnetic anisotropy energy of the filling particles in the matrix is the sum over single particles. It is shown that the particle rotation leads to the mechanical deformations and therefore mechanical stresses in the vicinity of the particle. This effect is pronounced well if the magnetic anisotropy coefficient $K$ is much larger than the shear modulus $\mu$ of the elastic matrix. The magnetic-field-induced internal stresses may lead to effects of magnetodeformation and diminish the mechanical compliance of the composite material, known as the MR effect.


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