Effect of single-particle magnetostriction on the shear modulus of compliant magnetoactive elastomers

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

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

[2]*Institute of Physics NAS of Ukraine, Prospekt Nauky 46, 03028 Kiev, 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 November 2016; revised manuscript received 23 January 2017; published 30 March 2017)

The influence of an external magnetic field on the static shear strain and the effective shear modulus of a magnetoactive elastomer (MAE) is studied theoretically in the framework of a recently introduced approach to the single-particle magnetostriction mechanism [V. M. Kalita *et al.*, Phys. Rev. E **93**, 062503 (2016)]. The planar problem of magnetostriction in an MAE with magnetically soft inclusions in the form of a thin disk (platelet) having the magnetic anisotropy in the plane of this disk is solved analytically. An external magnetic field acts with torques on magnetic filler particles, creates mechanical stresses in the vicinity of inclusions, induces shear strain, and increases the effective shear modulus of these composite materials. It is shown that the largest effect of the magnetic field on the effective shear modulus should be expected in MAEs with soft elastomer matrices, where the shear modulus of the matrix is less than the magnetic anisotropy constant of inclusions. It is derived that the effective shear modulus is nonlinearly dependent on the external magnetic field and approaches the saturation value in magnetic fields exceeding the field of particle anisotropy. It is shown that model calculations of the effective shear modulus correspond to a phenomenological definition of effective elastic moduli and magnetoelastic coupling constants. The obtained theoretical results compare well with known experimental data. Determination of effective elastic coefficients in MAEs and their dependence on magnetic field is discussed. The concentration dependence of the effective shear modulus at higher filler concentrations has been estimated using the method of Padé approximants, which predicts that both the absolute and relative changes of the magnetic-field-dependent effective shear modulus will significantly increase with the growing concentration of filler particles.

DOI: 10.1103/PhysRevE.95.032503

I. INTRODUCTION

Investigation of magnetoactive elastomers (MAEs) with μm-sized magnetic inclusions (filler particles) is of considerable interest both for industrial applications and fundamental physics of soft matter [1–6]. Under the influence of a magnetic field, MAEs exhibit large magnetostriction (MS) and magnetodeformation which may be several orders of magnitude larger than the magnetostriction of conventional magnetic materials [7–12]. The origin of MS in compliant MAEs is different from MS of solid ferromagnets.

In conventional ferromagnetic materials, MS is a consequence of the occurrence of magnetoelastic stresses upon magnetization [13–17]. In MAEs, the inherent MS of magnetic inclusions can be neglected, because the MS of these composite materials is much larger than the MS of individual inclusions [18]. MS in MAEs is caused by the action of the magnetized particles on the nonmagnetic matrix. Usually, an elongation of MAE in magnetic fields, which is associated with interparticle interactions, is investigated [19,20]. For example, in an MAE, MS can be caused by dipole interactions between the magnetic particles, which either attract or repel each other [21–23]. Driven by these forces, filler particles bound to the polymer network can change their spatial positions; the compliant polymer matrix follows them and the sample becomes deformed [24–26]. However, when averaged over the sample, the appearance of MS in MAEs is similar to the MS of conventional ferromagnetic materials.

In this paper, we shall study shear deformations in an MAE in the presence of a magnetic field. This type of deformation is associated with the rotation of magnetic particles by a magnetic field, as shown in [18]. Rotation of a particle is a consequence of the action of the torque applied to this particle by a magnetic field. Figuratively speaking, the magnetic field acts as a "screwdriver" on the particle winding up the adjacent elastomer matrix. Investigation of this type of mechanical influence on the MAE by a magnetic field and the description of the impact of local moments of force on the values of shear strain and stress is an emerging statement of the problem in mechanics of composite materials. Conveyance of torques from magnetic particles to the polymer matrix and the resulting deformation of a composite material has been previously considered theoretically for MAEs [10,11] and ferrogels [27–29] using alternative material models.

Magnetorheological (MR), or field-stiffening effect, is the most notable property of MAEs. It can be defined as the large increase of static or dynamic elastic moduli in externally applied dc magnetic fields. It has been pointed out by several authors that MS and MR effects are interrelated, since their physical origin is the magnetomechanical coupling

*mikhail.chamonine@oth-regensburg.de

between the constituents of the composite materials (see, e.g., [23,30,31] for recent argumentation). Significant theoretical efforts have been made for disclosing physical mechanisms behind the magnetomechanical coupling phenomena in MAEs and gels. Macroscopic theories are based on the continuum-mechanical representation of both the elastomer matrix and magnetic inclusions; see, e.g., [9,10,32–36]. Mesoscopic approaches can account for the granularity and the constitution of magnetic filler particles as separate things; see, e.g., [37–44]. An overview of currently available theoretical approaches can be found in Ref. [30]. Alternatively, dynamic properties of MAEs can be phenomenologically modeled using equivalent circuits comprising conventional or generalized springs and dashpots [45–51]. Although significant progress has been achieved in recent years, a unified and consistent theoretical description of mechanical behavior of MAEs in magnetic fields is still not complete. Since the underlying physical phenomena are rather complex and different physical effects come simultaneously into play, finding the main physical reason from the results of a numerical experiment could be a daunting task leaving room for the development of simplified physical models.

The elastic properties of MAEs, as well as of any other composite materials, are characterized by their effective moduli, whose values depend on the elastic characteristics of the matrix and inclusions, the shape of the inclusions, and their volume concentration [52]. The calculation of the elastic fields in the vicinity of an isolated nonmagnetic inclusion of a regular shape (cylinder, sphere) is a well-known problem [53]. For the pure shear, local deformations of such inclusions are anisotropic and have angular dependence with a period of π rad [53].

Recently, a single-particle mechanism of MS in MAEs has been considered by us in two dimensions [18]. It was shown that, in an external magnetic field, a magnetically soft but mechanically rigid cylindrical inclusion creates inhomogeneous isotropic local displacements of the elastomer matrix. Deformation around such a single particle, positioned in the center of a polar coordinate system, depends on the distance from the particle and is independent of the polar angle in the plane of the matrix. Therefore, the resulting deformation is inhomogeneous but isotropic. For the single-particle mechanism of MS [18], local deformations of the elastomer matrix in the vicinity of a magnetic particle differ from local deformations observed under shear strain in the vicinity of a nonmagnetic inclusion of the same shape. In the following it will be considered how a particular type of local displacement of the elastomer matrix associated with the single-particle mechanism of MS influences the effective shear modulus of MAEs [53].

The peculiarity of the description of mechanical deformations in MAEs, as compared to conventional elastomers, is that the magnetic inclusion has additional degrees of freedom, which must be taken into account when calculating the effective characteristics of MAE in an external magnetic field. As shown in [18], these degrees of freedom are the directions of the magnetic moment vectors of filler particles and the orientations of the easy magnetization axes of magnetic inclusions. A crucial effect of the magnetic anisotropy of filler particles on the equilibrium structure and magnetization of ferrogels has been recently investigated in [54] by coarse-grained molecular dynamics simulations. It has been also shown that the elastic response of ferrogel systems where the particles can be chemically cross-linked into the polymer matrix and the magnetic moments can be fixed to the particles' axes is strongly influenced by the type of magnetoelastic coupling [55].

In this paper, the effect of magnetic field on the magnitude of the shear modulus of MAEs with a low concentration of magnetically anisotropic particles is studied theoretically. An approach for taking into account the additional degrees of freedom of magnetic filler particles of the composite and calculating the effect of the magnetic field on the elastic moduli is proposed. This procedure allows one to determine the effect of rotation of magnetic particles and their magnetic anisotropy axes on the value of the effective shear modulus of an MAE.

Unlike most other composite engineering materials, magnetic and elastic systems in compliant MAEs are strongly interconnected [56]. Magnetization of an MAE depends on its deformation, and the deformation depends on the magnetic state. Calculation of effective elastic characteristics of an MAE should be performed in conjunction with the calculation of its effective magnetic characteristics. This requirement complicates the statement of the problem. The theoretical solution to the problem requires averaging and is only attainable with model simplifications. In the present paper, we will not consider the effect of the demagnetization factor of inclusions. For this purpose, we have solved a two-dimensional (plane) problem with particles in the form of thin disks, whose demagnetization factor vanishes for all magnetic-field directions in the disk plane. In the case of planar MAE, the external magnetic field is not distorted and is equal to the internal field magnetizing the particles.

In the following considerations, the matrix undergoes plane deformation [57]. This assumption is satisfied if the requirement of a constant thickness of the sample is imposed as a boundary condition. The mathematical task of finding the relationship between the shear deformation and stress in magnetic fields is simplified and corresponding analytical solutions can be easily obtained.

The paper is organized as follows. In Sec. II our theoretical model is presented and the general expression for the energy of the composite material in a magnetic field is formulated. The linearized case is solved in Sec. III. Section IV analyzes the results for the specific case of a magnetomechanically soft elastomer matrix. Under a magnetomechanically soft matrix we understand such a matrix where the following condition is fulfilled: $\mu/K \ll 1$, where μ is the shear modulus of the elastomer matrix and K is the magnetic anisotropy constant of particles. From the point of view of physics the latter condition means that reorientation of the particle in an external magnetic field can easily deform the matrix. The condition $\mu/K \ll 1$ is feasible and the corresponding estimates have been made in Ref. [18]. The concept of effective coefficients for composite magnetoactive materials is addressed in Sec. V, where correct experimental determination of the effective shear modulus is substantiated. The results are extensively discussed and compared with experimental data in Sec. VI. Conclusions are drawn in the final section.

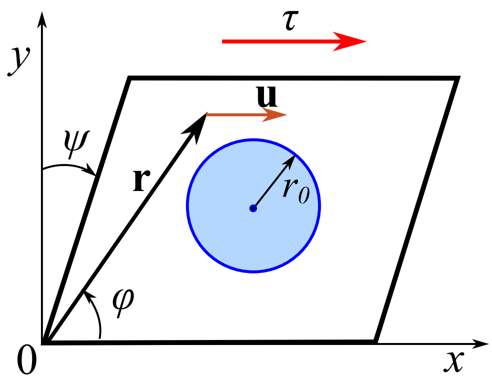

FIG. 1. Simple shear test. The engineering shear strain $\psi \ll 1$ is measured from the y axis and considered to be positive in the clockwise direction. The position of a point in the medium is determined by the radius vector **r**. A positive shear stress $\tau > 0$ is shown. The material thickness D_0 in the direction perpendicular to the plane of the paper is much smaller than the radius of the inclusion r_0: $D_0 \ll r_0$.

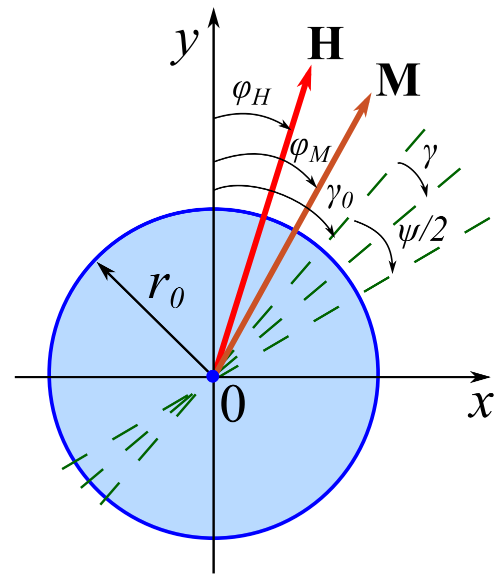

FIG. 2. Orientation of the particle's anisotropy axis (dashed line), the magnetic-field vector **H**, and the magnetic moment vector **M** in the coordinate system shown in Fig. 1. The angles are measured from the y axis and given positive in the clockwise direction. The angles φ_H, φ_M denote the direction of vectors **H** and **M**, respectively. The angle γ_0 specifies the initial direction of the magnetic anisotropy axis of the particle; γ shows the change in the direction of the particle's anisotropy axis after the shear deformation, in which the particle rotates by the angle $\psi/2$.

II. ENERGY OF A COMPOSITE MATERIAL UNDER SHEAR DEFORMATION IN A MAGNETIC FIELD

We shall study the static simple shearing of an MAE sample (see Fig. 1). This is the type of shear, as shown in Fig. 1, for which the dynamic shear modulus of MAE samples is determined in low-frequency (oscillation frequency $f \approx 1 - 10\,\mathrm{Hz}$) oscillatory shear experiments in the plate-plate configuration with the fixed gap between the plates; see, for example, [47,58–61]. In this shear deformation, the displacement vector $\boldsymbol{u}$ depends only on the y coordinate as $\mathbf{u} = y\psi\mathbf{e}_x$, where $\psi \ll 1$, and $\mathbf{e}_x$ is the unit vector along the x axis. In the polar coordinate system, this vector has two components: $\mathbf{u} = (r\psi \sin 2\varphi/2)\mathbf{e}_r + r[(-\psi + \psi \cos 2\varphi)/2]\mathbf{e}_\varphi$. The first term in the second bracket is independent of the angle φ. Because of this term, all points of the sample and, therefore, the entire sample, are rotated at an angle $\psi/2$.

Now, if the sample contains a rigid inclusion (i.e., the Young's and shear moduli are much greater than those of the elastomer matrix and can be considered to be infinitely large) with circular shape of a radius r_0, then, provided that the sample size is much larger than the inclusion's radius, it will also rotate through an angle $\psi/2$ without changing its shape and size. If the inclusion has the anisotropy of magnetic properties, then, in a magnetic field, there will be a torque, acting on the particle as it rotates during the shear deformation. This moment of force will lead to a rotation of the particle and create an additional isotropic [18] deformation around the particle.

The elastomer matrix is considered to be incompressible; this means that the Poisson's ratio ν_M is equal to 0.5. The thickness of the elastomer matrix and the inclusions D_0 is constant everywhere.

Furthermore, we assume that the inclusion consists of an ideal, magnetically soft material (this means that its magnetization m_I depends on magnetic-field strength H as $m_I(H) = m(\Theta(H) - \Theta(-H))$, where $\Theta(H)$ is the Heaviside function), has a uniaxial magnetic anisotropy in the xy plane, and that the anisotropy axis is preferable for the direction of the magnetic moment of the particle. However, there is no loss of generality in the following calculations, since any dependence $m_I(H)$ can be easily introduced into the model.

Denote the initial orientation of the particle's easy axis by the angle γ_0 (see Fig. 2). Under the shear in the zero magnetic field $\mathbf{H} = 0$, the direction of the particle's easy axis is equal to the angle $\gamma_0 + \psi/2$. If $\mathbf{H} \neq 0$, the angle giving the direction of the particle's easy axis will be equal to $\gamma_0 + \gamma$. Thus the rotation angle of the particle due to the shear deformation in the magnetic field is equal to the difference of the above said angles: $\gamma_0 + \psi/2 - (\gamma_0 + \gamma) = \psi/2 - \gamma$.

Magnetic energy of the particle and elastic energy of the surrounding matrix taking into account rotation of the particle in a magnetic field caused by the single-particle mechanism of magnetostriction [18] can be written as the following sum:

$$E = \big[-\tfrac{1}{2}K\cos^2(\gamma_0 + \gamma - \varphi_M) - Hm\cos(\varphi_H - \varphi_M) + 2\mu(\psi/2 - \gamma)^2\big]V_0, \qquad (1)$$

where V_0 is the volume of a single particle, $m = M/V_0$ is the particle's magnetization, K is the constant of magnetic anisotropy, $H = |\mathbf{H}|$, μ is the shear modulus of the matrix, φ_H is the angle determining the direction of the magnetic field (cf. Fig. 2), and φ_M is the angle determining the direction of the magnetic moment ($M = |\mathbf{M}|$) of the inclusion. The first term in (1) describes the energy of the magnetic anisotropy, the second term stands for the Zeeman energy, and the third term represents the elastic energy of the matrix taking into account an additional rotation of the particle due to the influence of the magnetic field [18].

If all particles are identical and have the same orientation of the anisotropy axis, in the case of low concentration, when the mutual influence of the elastic fields of MAE particles can be neglected, the volume density of single-particle energy in a magnetic field can be written as

$$\boldsymbol{\varepsilon}_1 = \frac{NE}{V} = p\left[-\frac{1}{2}K\cos^2(\gamma_0+\gamma-\varphi_M) - Hm\cos(\varphi_H-\varphi_M) + 2\mu(\psi/2-\gamma)^2\right], \quad (2)$$

where N is the number of particles in the volume V, and $NV_0/V = p$ is the volume fraction of magnetic particles in the composite material; in short, it is their concentration.

To write down the expression for the full energy of an MAE under shear deformation in a magnetic field, the expression (2) should be supplemented by the energy contribution from the shear deformation, $[G^{\text{eff}}(0)\psi^2]/2$, and the term $\tau\psi$ that takes into account the shear stress. Thus the final expression for the energy density of a composite with low particle concentration, which includes the magnetic energy of particles, the elastic energy of the matrix caused by particles' rotations in a magnetic field, and the elastic energy of the sheared sample under shear stress, takes the form

$$\boldsymbol{\varepsilon} = p\left[-\tfrac{1}{2}K\cos^2(\gamma_0+\gamma-\varphi_M) - Hm\cos(\varphi_H-\varphi_M) + 2\mu(\psi/2-\gamma^2)\right] + \tfrac{1}{2}G^{\text{eff}}(0)\psi^2 - \tau\psi, \quad (3)$$

where $G^{\text{eff}}(0)$ is the effective shear modulus in the absence of external magnetic field $H = 0$ [53,62,63]; $\tau\psi$ is the work per unit volume, done by an external force to create the shear strain ψ. The term $\frac{1}{2}G^{\text{eff}}(0)\psi^2$ is the shear strain energy density of the sample with inclusions and τ is the external shear stress (see Sec. V below).

Note that the direction of the magnetic field with respect to the anisotropy axis of the particles and the y axis can be arbitrary. Therefore, expression (3) contains finite values for the angles of the magnetic-field direction and for the angular orientation of the anisotropy axes. The inclination of the magnetic moments of the particles, caused by the magnetic field, can be arbitrarily large and its value is not limited. However, the deformation induced by magnetic particles should not be large. This is a typical situation for conventional magnetic materials, where magnetoelasticity causes very small deformations in comparison to those observable in MAEs. In contrast to the crystalline magnetic materials, the elastomer matrix surrounding the magnetic particles can be highly elastic. The resulting rotation of particles in a magnetic field and the induced sample shearing can be large as well. Therefore, in calculations for MAEs with the highly elastic matrix, we controlled the field values and the parameters of the problem in such a way that the resulting deformation remains small.

Thus, for a system of identical particles with the same direction of anisotropy axes, the values of deformation and the angles can be found by minimizing the energy density (3):

$$\frac{\partial\boldsymbol{\varepsilon}}{\partial\varphi_M} = p[-K\cos(\gamma_0+\gamma-\varphi_M)\sin(\gamma_0+\gamma-\varphi_M) - Hm\sin(\varphi_H-\varphi_M)] = 0, \quad (4)$$

$$\frac{\partial\boldsymbol{\varepsilon}}{\partial\gamma} = p[K\cos(\gamma_0+\gamma-\varphi_M)\sin(\gamma_0+\gamma-\varphi_M) - 4\mu(\psi/2-\gamma)] = 0, \quad (5)$$

$$\frac{\partial\boldsymbol{\varepsilon}}{\partial\psi} = p2\mu(\psi/2-\gamma) + G^{\text{eff}}(0)\psi - \tau = 0. \quad (6)$$

Equation (4) is obtained by differentiating the energy over the deviation angle of the magnetic moment vector. In (4), the torque caused by the external field on the magnetic moment of the particle is equal to the torque acting on the magnetic moment vector by the anisotropy field $H_A = K/m$, which in the case of the easy-axis anisotropy is directed along the easy magnetization axis of the particle. Equation (5) corresponds to the mechanical equilibrium condition of the particle. It is obtained by differentiation over the rotation angle of the particle. In (5), the torque acting on the particle due to its magnetic moment is equal to the torque caused by the matrix surrounding the particle. Equation (6) is obtained by differentiation over the shear strain. From Equation (6) it follows that the magnitude of the shear stress is equal to the sum of the contributions caused by the shear deformation and an additional contribution to the strain created in the matrix by the rotation of particles in a magnetic field.

The following expression can be derived from (4)–(6):

$$\tau = G^{\text{eff}}(0)\psi - \tfrac{1}{2}pHm\sin(\varphi_H-\varphi_M). \quad (7)$$

From (7) it can be concluded that the torque acting on the magnetic moment vector of a particle results in an additional contribution to the stress.

III. LINEARIZED PROBLEM

A. Linearization

Because the system of equations (4)–(6) is nonlinear, it is interesting to consider its solution when all the angles in the expression for the energy (3) are small. We also assume in this section that in the initial state $\tau = 0$, $H = 0$, and the angle $\gamma_0 \neq 0$. This means that we consider an MAE filled with particles which have the same direction of the anisotropy axis and this axis is not perpendicular to the direction of the applied shear stress (i.e., the x axis; see Fig. 1). For generality, we assume that the magnetic field can be tilted with respect to the y axis. In this case, the expression for the energy density is simplified and can be written as

$$e = p\left\{-\tfrac{1}{2}K[1-(\varphi_M-\gamma-\gamma_0)^2] - Hm\left[1-\tfrac{1}{2}(\varphi_H-\varphi_M)^2\right] + 2\mu(\gamma-\psi/2)^2\right\} + \tfrac{1}{2}G^{\text{eff}}(0)\psi^2 - \tau\psi. \quad (8)$$

Equations of state for the MAE are now linearized:

$$K(\varphi_M-\gamma-\gamma_0) - Hm(\varphi_H-\varphi_M) = 0, \quad (9)$$

$$-K(\varphi_M-\gamma-\gamma_0) + 4\mu\left(\gamma-\tfrac{1}{2}\psi\right) = 0, \quad (10)$$

$$p\left[-2\mu\left(\gamma-\tfrac{1}{2}\psi\right)\right] + G^{\text{eff}}(0)\psi - \tau = 0. \quad (11)$$

From the equations of state we get an expression for the shear strain ψ, which can be written as

$$\tau = \left[G^{\mathrm{eff}}(0) + \mu p \frac{Hm/K}{(1+Hm/K)(1+4\mu/K)-1}\right]\psi - 2p\mu \frac{\varphi_H - \gamma_0}{(1+Hm/K)(1+4\mu/K)-1}\frac{Hm}{K}. \quad (12)$$

When $H = 0$, Eq. (12) meets the definition of the effective modulus of the composite material in the absence of a magnetic field: $G^{\mathrm{eff}}(0) = \tau/\psi$.

The product $(\varphi_H - \gamma_0)Hm$ in (12) is equal to the magnitude of the vector product of the magnetization and the magnetic-field strength, where $(\varphi_H - \gamma_0)m$ is the component of the magnetization vector in the direction orthogonal to the magnetic-field vector. The right side of Eq. (12) has two contributions. The first contribution is proportional to the strain ψ and independent of the direction of the magnetic field $\mathbf{H}$. The second term on the right side of (12) includes angle φ_H; i.e., this term depends on the direction of the magnetic field. At the same time, there are situations when the signs of τ and φ_H may be either the same (e.g.,$\tau,\varphi_H > 0$) or different (e.g., $\tau > 0, \varphi_H < 0$). This, in turn, leads to asymmetry. In the first case, the shear stress τ and the additional stress created by the action of the magnetic field add to and reinforce each other, contributing to the shear strain. In the second case, these stresses counteract each other, reducing the shear strain. This asymmetry of the influence of the oblique magnetic field and the shear is related to the fact that the magnetic field itself is capable of producing shear deformation.

B. Shear deformation induced by a tilted field

For a system of particles with anisotropy axes perpendicular to the direction of shear stress $\gamma_0 = 0$ and being in a tilted magnetic field ($\varphi_H \neq 0$), the value of shear strain in the absence of stress $\tau = 0$ is given by

$$\psi = \frac{2\mu K p H m \varphi_H}{4\mu K G^{\mathrm{eff}}(0) + Hm[G^{\mathrm{eff}}(0)(K+4\mu) + Kp\mu]}. \quad (13)$$

From (13) it is seen that in weak ($H\to 0$) fields the shear value linearly depends on the field magnitude and it reaches saturation in strong magnetic fields $H\to\infty$. Note that Eq. (13) is nonlinear with respect to p. However, since it is a consequence of Eq. (12) linear dependence of τ on p is preserved.

According to (13), the maximum shear strain induced by a tilted field is achieved in an MAE if $\mu \ll K$. In this case, the easy magnetization axes of the particles are oriented along the magnetic field $\gamma = \varphi_H$ and the limiting value of the field-induced shear deformation is equal to

$$\psi(H\to\infty) = \frac{2\mu}{G^{\mathrm{eff}}(0) + p\mu} p\varphi_H \approx 2p\varphi_H. \quad (14)$$

Therefore the limiting value of the shear deformation induced by a magnetic field in the MAE with $K \gg \mu$ is comparable with the magnitude of the inclination angle of the magnetic field. The magnitude of such a shear deformation is many times greater than the amount of shear induced in the conventional ferromagnetic materials [18].

For MAEs with the small anisotropy constant of particles $K \ll G^{\mathrm{eff}}(0)$, the magnitude of shear strain in the limit of a strong tilted field $H \to \infty$ is significantly weakened in comparison to (14):

$$\psi_H(H\to\infty) = \frac{K}{2G^{\mathrm{eff}}(0)} p\varphi_H. \quad (15)$$

Thus the torque created by the magnetic field induces a shear strain in the absence of an external shear stress. The sign of this shear deformation depends on the direction of the applied field.

C. Shearing in the field orthogonal to the direction of stress ($\varphi_H = \gamma_0 = 0$)

A magnetic field, which is perpendicular to the direction of the external stress (i.e., parallel to the y axis in Fig. 1), is not capable of inducing the shear strain. Thus, when $\varphi_H = 0$, $\gamma_0 = 0$, and the strain vanishes, $\tau = 0$, the vectors of the particle's magnetic moment and the magnetic field are collinear. Therefore, when $\varphi_H = \gamma_0 = 0$ and $\tau = 0$, a perpendicular magnetic field does not generate torque, does not rotate the particle, and does not deform the sample.

However, with $\varphi_H = \gamma_0 = 0$, the magnetic field will have significance, if stress $\tau \neq 0$ is applied to the sample. Under the influence of stress a shear deformation of the sample $\psi \neq 0$ must occur. In this case, the easy magnetization axes will not be directed along the y axis (see Figs. 1 and 2) and there will be torques acting on the particles by virtue of the magnetic field. This torque, creating its additional contribution to the stress τ, will affect the amount of the sample's shear by reducing its value and, respectively, increasing the value of the shear modulus.

Indeed, from (12) we obtain the expression

$$\tau = G^{\mathrm{eff}}(0)\left\{1 + \frac{HmK\mu}{G^{\mathrm{eff}}(0)[4\mu K + Hm(K+4\mu)]} p\right\}\psi. \quad (16)$$

The effective shear modulus of an MAE in an external magnetic field $H \neq 0$ can be defined as a proportionality factor between the shear strain and stress, so that $G_e(H) = \tau/\psi$:

$$G^{\mathrm{eff}}(H) = G^{\mathrm{eff}}(0)\left\{1 + \frac{HmK\mu}{G^{\mathrm{eff}}(0)[4\mu K + Hm(K+4\mu)]} p\right\}. \quad (17)$$

Equation (17) describes the change in the effective shear modulus of the MAE under the influence of a magnetic field directed along the anisotropy axis of the particles, and the applied shear stress, which is perpendicular to the magnetic field.

From (12) it is seen that in weak ($H\to 0$) fields, the magnetic-field contribution to the effective shear modulus is proportional to H. The magnitude of this additional contribution saturates in large ($H \to \infty$) fields. For the case $K \gg \mu$, we obtain in the large-field limit $H \to \infty$ that

$$G^{\mathrm{eff}}(H\to\infty) = G^{\mathrm{eff}}(0) + p\mu. \quad (18)$$

From (18) it follows that in the saturation limit the magnetic-field contribution to the effective modulus depends on the concentration of the particles and the elasticity of the matrix.

IV. LIMITING CASES OF MAEs WITH A MAGNETOMECHANICALLY SOFT MATRIX

Consider some specific cases for the magnetization of the particles possessing strong magnetic anisotropy and located in a compliant matrix. In cases where there is a limiting magnetization (this means that the magnetic moment is directed either along the anisotropy axis or along the external magnetic field), the description of shearing behavior is simplified due to a reduction of the number of degrees of freedom [the number of varying variables in the expression for the energy density (3)].

A. Highly anisotropic particles

Let, as in Sec. II, the axis of the magnetic anisotropy of the particles deviate from the direction of the y axis by an angle γ_0 (cf. Figs. 1 and 2). Furthermore, let the magnetic anisotropy of the particles be so large and the matrix mechanically so soft that the condition $K/\mu \gg 1$ is fulfilled. Estimates of the ratio K/μ have been made in [18]. In such an MAE, the magnetic moment vector of the particle can be with difficulty deflected from the anisotropy axis and it is much easier to rotate the particle. Therefore, the vector of the particle's magnetic moment is always directed along the anisotropy axis of the particle: $\varphi_M = \gamma_0 + \gamma$. When this equality of the angles is fulfilled, the expression for the energy density (3) is simplified and takes the form

$$\begin{aligned}\boldsymbol{\varepsilon} = p\{-Hm\cos[\varphi_H - (\gamma_0+\gamma)] + 2\mu(\psi/2-\gamma)^2\}\\ +\tfrac{1}{2}G^{\mathrm{eff}}(0)\psi^2 - \tau\psi.\end{aligned} \tag{19}$$

For particles oriented perpendicular to the direction of the shear stress $\gamma_0 = 0$, we obtain from (19) in the linear case that

$$\psi = \frac{\tau(4\mu + Hm) + 2\mu p H m \varphi_H}{4\mu G^{\mathrm{eff}}(0) + Hm[G^{\mathrm{eff}}(0) + p\mu]}. \tag{20}$$

It can be easily seen that the expression (20) coincides with (12) if $K \to \infty$. Therefore, the limiting values for the shear magnitude and the shear modulus obtained with the help of (20) will coincide with the values (15) and (18) in saturating magnetic field.

B. Highly anisotropic particles in saturating magnetic fields

Consider the behavior of the MEA comprising the particles with a large magnetic anisotropy and a soft matrix $\mu \ll K$ in a saturating magnetic field. In a strong magnetic field $H \gg \mu/m$, which in this case may be less than the anisotropy field $H < H_A = K/m$, magnetic moments of particles and anisotropy axes of particles will be oriented along the field: $\varphi_H = \varphi_M$ and $\varphi_H = \gamma_0 + \gamma$. Therefore, the rotation angle of the particle's magnetization axis will be equal to the difference $\gamma = \varphi_H - \gamma_0$. In this case, the energy density is of the form

$$\boldsymbol{\varepsilon} = p\{2\mu[\psi/2 - (\varphi_H - \gamma_0)]^2\} + \tfrac{1}{2}G^{\mathrm{eff}}(0)\psi^2 - \tau\psi. \tag{21}$$

Minimizing (21) with respect to ψ, we obtain

$$\psi(H\to\infty) = \frac{\tau + 2\mu p(\varphi_H - \gamma_0)}{[G^{\mathrm{eff}}(0) + p\mu]}. \tag{22}$$

Equation (22) coincides with the formula (14) for the case $\gamma_0 = 0$ and $\tau = 0$. Thus the energy density (21) with a smaller number of degrees of freedom gives the result corresponding to the exact solution for the assumed conditions of large anisotropy, softness of the matrix, and the saturating magnetic field.

V. EFFECTIVE COEFFICIENTS IN MAGNETOACTIVE COMPOSITE MATERIALS

The relationship between the mechanical stresses on the specimen (components of stress tensor $\langle\boldsymbol{\sigma}\rangle$) and the deformations of the MAE sample (components of strain tensor $\langle\boldsymbol{\varepsilon}\rangle$) can be written phenomenologically. $\langle\cdots\rangle$ denotes averaging over the entire volume [53,64,65]. Averaged characteristics correspond to the certain effective medium that is considered to be homogeneous with the averaged properties the same as those of the composite material. If a composite material is replaced by such an effective medium, it will behave in the same way as the original material with respect to external influences.

The phenomenological approach was used in [66] for crystalline ferromagnetic materials. Besides the terms being symmetric with respect to the indices, the elastic stresses in the MAE will also include magnetoelastic coupling caused by the effect of rotation of inclusions in the matrix caused by the torque from an external magnetic field. Given that the torque is proportional to the vector product of the magnetization and the magnetic-field strength, the magnetoelastic additional term in the expression for the stress will contain components of an antisymmetric tensor. As a result, we arrive at the following expression for the stress:

$$\begin{aligned}\langle\sigma_{ik}\rangle = C^{\mathrm{eff}}_{iklm}\langle\boldsymbol{\varepsilon}_{lm}\rangle + A^{\mathrm{eff}}_{iklm}\langle m_l\rangle\langle m_k\rangle\\ + B^{\mathrm{eff}}_{iklm}(\langle m_l\rangle\langle H_m\rangle - \langle m_m\rangle\langle H_l\rangle),\end{aligned} \tag{23}$$

where C^{eff}_{iklm} are effective elastic constants, A^{eff}_{iklm} and B^{eff}_{iklm} are effective magnetoelastic coupling coefficients, $\langle m_l\rangle$ and $\langle m_k\rangle$ are projections of the average magnetization vectors, and $\langle H_l\rangle$ and $\langle H_m\rangle$ are projections of the magnetic-field vector $\mathbf{H} = \langle\mathbf{H}\rangle$, in which the sample is placed. The first magnetoelastic contribution to the stresses (23) is symmetric with respect to the components of the average magnetization, and the second contribution is expressed through the antisymmetric tensor comprising the projections of the magnetic-field strength. Expression (23) is written in the approximation of the smallest exponents with respect to $\langle H_l\rangle$, $\langle m_l\rangle$, and $\langle\boldsymbol{\varepsilon}_{lm}\rangle$. Note that in the linear approximation with respect to $\langle\boldsymbol{\varepsilon}\rangle$, the coefficients C^{eff}_{iklm} may depend on magnetic field $\mathbf{H}$. In the case of a strongly nonlinear MAE, expansion (23) must contain higher-order terms in the power series and take into account the possibility of the formation in MAE structures, such as particle chain aggregates. General considerations of the interactions between electric, magnetic, and elastic subsystems in nonlinear disordered micropolar media in the framework of phenomenological elastomagnetoelectrostatics can be found, e.g., in Ref. [67].

Obviously, the physical approximations made in (23) are satisfied for MAEs with the low particle concentration $p \ll 1$. The approximation made in the formulation of the third term in (23) is also fulfilled if the local additional field is proportional to the average magnetization, what may occur if the particles obey a spatial order in their arrangement [68].

If MAE filler particles have magnetic anisotropy and the particle concentration is low, the third term in (23) may exceed the second term. In this case, for an MAE with the low concentration of ferromagnetic inclusions, the following relation is valid for the shearing experiment of Fig. 1:

$$\langle\tau(\psi \neq 0,\mathbf{H})\rangle = G^{\mathrm{eff}}(\mathbf{H})\psi + B^{\mathrm{eff}}(\langle m_n\rangle\langle H_t\rangle - \langle m_t\rangle\langle H_n\rangle), \tag{24}$$

where G^{eff} is the effective shear modulus and B^{eff} is the effective magnetoelastic coupling constant, both depending on $\mathbf{H}$. The index t indicates projection of the vector along the shear stress, and n denotes projection of the vector in the perpendicular direction. In (24), it is taken into account that both components of the magnetic field $\langle H_t\rangle$ and $\langle H_n\rangle$ create a torque on the inclusions, if the magnetization vector is not collinear to $\mathbf{H}$. The expression (24) completely corresponds to the relationship between shear stress and strain in MAEs at small ($H\to 0$) values of the field, obtained from our model.

From (24) we have that for an MAE with a linearly deformable matrix and single-particle mechanism of MS, the magnetic field in the absence of shear strain $\psi = 0$ and $\langle\mathbf{H}\rangle \neq 0$ induces the shear stress

$$\langle\tau(\psi = 0,\mathbf{H})\rangle = B^{\mathrm{eff}}(\langle m_n\rangle\langle H_t\rangle - \langle m_t\rangle\langle H_n\rangle). \tag{25}$$

From Eqs. (24) and (25) we obtain that the effective shear modulus is determined from the following relationship:

$$G^{\mathrm{eff}}(\mathbf{H}) = \frac{\tau(\psi \neq 0,\mathbf{H}) - \tau(\psi = 0,\mathbf{H})}{\psi}. \tag{26}$$

Definition of the effective shear modulus (26) is rather general and does not contain the restrictions of the phenomenological description (23), which is confirmed by the calculations of our model. This definition is important for the most common deformation experiment of MAEs, as shown in Fig. 1. It allows one to properly take into account the resulting additional magnetoelastic stresses. Otherwise, the result of the experimental data processing may be corrupted. Furthermore, the relation (26) shows how to calculate the effective modulus in the above described model in a tilted ($\varphi_H \neq 0$) magnetic field or in the case $\gamma_0 \neq 0$ (see Sec. VI B below).

In MAEs, all effective coefficients may have significant dependences both on the applied magnetic field (see, e.g., Figs. 5 and 6) and on the strain amplitude [69,70]. The definition of (26) can be generalized to other elastic effective coefficients of MAEs by writing the following matrix equation:

$$\mathbf{C}^{\mathrm{eff}}(\langle\boldsymbol{\varepsilon}\rangle,\langle\mathbf{H}\rangle)\langle\boldsymbol{\varepsilon}\rangle = \langle\boldsymbol{\sigma}\rangle(\langle\boldsymbol{\varepsilon}\rangle \neq 0,\langle\mathbf{H}\rangle) - \langle\boldsymbol{\sigma}\rangle(\langle\boldsymbol{\varepsilon}\rangle = 0,\langle\mathbf{H}\rangle). \tag{27}$$

Expression (27) allows one to take into account the influence of $\langle\mathbf{H}\rangle$ and $\langle\boldsymbol{\varepsilon}\rangle$ on the value of the effective elastic moduli in MAEs.

VI. RESULTS AND DISCUSSION

A. Shearing and effective shear modulus for $\varphi_H = 0$ and $\gamma_0 = 0$

Field dependencies of the shear strain ψ obtained by solving Eqs. (4)–(6) are shown in Fig. 3. They are plotted for $\varphi_H = 0$ and $\gamma_0 = 0$. In this and the following figures of Sec. VI, the field is normalized as $h = H/H_A$, where the ratio $H_A = K/m$ is the magnetic anisotropy field of the inclusion. In the calculations, the concentration $p = 0.1$ is considered to be much less than the percolation threshold of composites $p_c \approx 0.5$. If $p = 0.1$, it can be assumed that the effects of elastic interaction between the particles are negligible and the linear approximation is valid. In the absence of a magnetic field at low concentrations, the effective modulus of the composite for a planar elasticity problem with the shear deformation in the direction transverse to the axis of the particles and inclusions of a cylindrical shape, as shown in Fig. 1, is described by the approximate expression $G^{\mathrm{eff}}(0) = \mu(1 + 2\frac{\lambda+2\mu}{\lambda+3\mu}p)$, where λ and μ are Lamé constants of the matrix [53,63]. It should be noted that the Poisson's ratio of the elastomer matrix ν_M is equal to 0.5, from which it follows that $G^{\mathrm{eff}}(0) = \mu(1 + 2p)$. The curves in Fig. 3(a) are obtained for the shear stress $\tau = 0.001\mu$ and different ratios of the anisotropy constant and the shear modulus of the matrix. The graphs show that the increasing magnetic field counteracts shearing and all the curves decline with the increasing magnetic field. It turns out that the effect of the magnetic field on the magnitude of the shear strain is linear in weak fields and saturates in strong fields. The greatest manifestation of the magnetic field on the magnitude of the shear strain is observed for the magnetomechanically soft matrix.

Figure 3(b) shows dependences of the shear strain $\psi(\varphi_H)$ obtained for different tilt angles φ_H of the magnetic field $\mathbf{H}$. The graphs are calculated in the absence of shear stress $\tau = 0$ at different values of the field and the shear modulus of the matrix. Straight lines on Fig. 4 were obtained by solving the system of equations (9)–(11) for the linear problem, while the curved lines (dash-dotted curves) correspond to the exact solutions of Eqs. (4)–(6). It is seen that the linear approximation provides a reasonable solution for a sufficiently broad range of tilt angles. Notable deviations between the solutions occur at larger tilt angles of the magnetic field, where the approximate solution overestimates the magnitude of induced shear strain.

In particular, it is interesting to consider the case of a rigid matrix [the lowest dash-dotted curve in Fig. 3(b)]. From this curve, it is seen that for $\varphi_H \to \pi/2$ the shear strain vanishes. This is due to the fact that in a rigid matrix the particle practically does not rotate and only the particle's magnetization vector undergoes rotation under the influence of the magnetic field. When there is no shearing and rotation of particles in a rigid matrix, then for $\varphi_H = \pi/2$ in the field which is equal to or greater than the anisotropy field, the angle $\varphi_M \to \pi/2$ (cf. [66]).

Now let us analyze the influence of magnetic field on the shear modulus for the case $\varphi_H = 0$ and $\gamma_0 = 0$. In such an orientation of the magnetic field and the anisotropy axis of the inclusion, there is no shear strain induced by the magnetic field and the value of the shear modulus can be determined from the ratio $\tau/\psi = G^{\mathrm{eff}}(H,\varphi_H = 0,\gamma_0 = 0)$. Using Eqs. (4)–(6), we obtained the shear strains for $\tau = 0.001\mu$, and consequently calculated the field dependence of the shear modulus for $H \neq 0$. Figure 4 shows the dependence of the normalized shear modulus $g^{\mathrm{eff}}(H) = G^{\mathrm{eff}}(H)/G^{\mathrm{eff}}(0)$. In Fig. 4(a), the graphs for the normalized shear modulus are obtained for different ratios of the shear modulus of the matrix and the

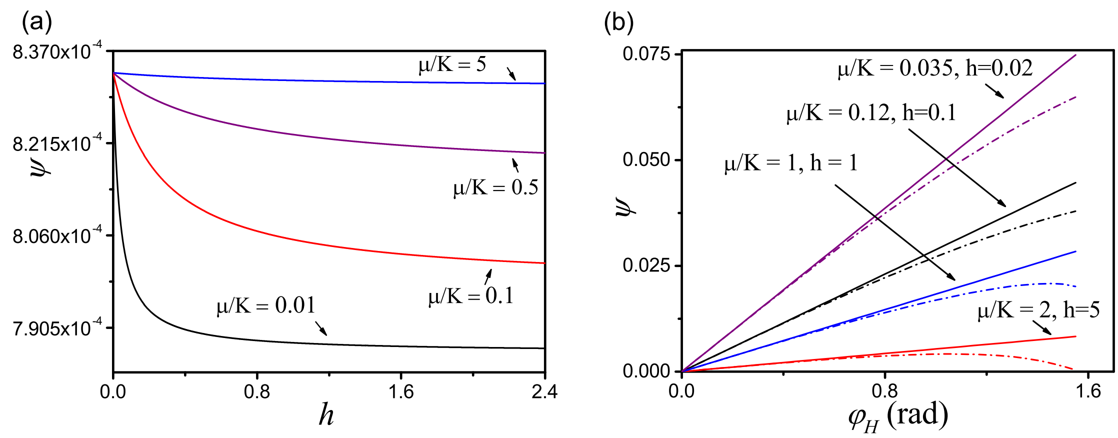

FIG. 3. (a) Dependence of the induced shear strain on the magnitude of the normalized magnetic field h in the presence of the shear strain $\tau = 0.001\mu$ with $\varphi_H = 0, \gamma_0 = 0$ and different values of μ/K. (b) Dependences of the induced shear strain on the inclination angle φ_H of vector $\boldsymbol{H}$ in the absence of the shear stress $\tau = 0$ for different values of the external magnetic field and the elastic shear modulus of the matrix: Straight lines were obtained by solving the system of equations (9)–(11) for the linear problem, while the dash-dotted curves correspond to the exact solutions of Eqs. (4)–(6).

magnetic anisotropy constant of the inclusion. It is seen that the saturation effect of the shear strain in the magnetic field is accompanied by the saturation of the shear modulus. The dependencies show that the influence of the magnetic field on the effective shear modulus is more pronounced for the magnetomechanically soft matrix when $\mu/K \ll 1$.

To illustrate the effect of the elastic properties of the matrix on the magnetic-field enhancement of the shear modulus, the dependencies of the effective shear modulus on the ratio μ/K of the shear modulus of the matrix and the anisotropy constant have been plotted in Fig. 4(b) for different constant magnetic fields at $\varphi_H = 0$, $\gamma_0 = 0$, and $\tau = 0.001\mu$. It can be observed that the value of the normalized shear modulus with the increasing matrix rigidity $\mu/K \gg 1$ tends to unity. For small $\mu/K \ll 1$, that is, for the magnetomechanically soft matrix, the effect of magnetic field on the effective shear modulus is the largest and g^{eff} is equal to its limiting (saturated) value of 1.083. Recall that in Figs. 3 and 4, p is equal to 0.1.

In Figs. 3 and 4, the effect of the magnetic field on the effective shear modulus has been analyzed for $\varphi_H = 0$ and $\gamma_0 = 0$. However, the tilted field induces a shear strain even in the absence of shear stress. Therefore, in a tilted magnetic field, determination of the effective shear modulus requires further discussion.

B. Determination of the effective shear modulus for $\varphi_H \neq 0$ and $\gamma_0 \neq 0$

To determine the effective shear modulus in a tilted magnetic field, we first calculate $\tau(\psi \neq 0, \mathbf{H} \neq 0)$ and $\tau(\psi = 0, \mathbf{H} \neq 0)$ using Eqs. (4)–(6) and then apply the relationship (26). Figure 5 shows the dependences of the normalized effective shear modulus $g^{\text{eff}}(\varphi_H)$ on the tilt angle φ_H of the magnetic field, whose magnitude is kept constant $|\mathbf{H}| = \text{const}$. All dependences are plotted for $\mu/K = 1$ and $\gamma_0 = 0$.

From Fig. 5 it is seen that the effective shear modulus depends nonlinearly on the magnetic-field direction. The

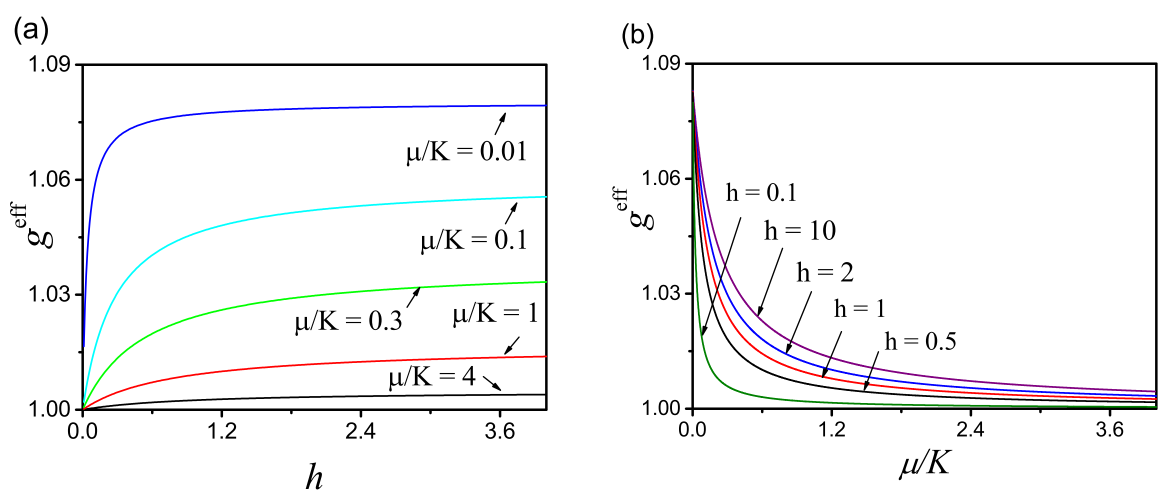

FIG. 4. Dependences of the normalized shear modulus g^{eff} obtained for $\tau = 0.001\mu$ at $\varphi_H = 0$ and $\gamma_0 = 0$. (a) Variation of the normalized magnetic field h with different values of μ/K. (b) Variation of the ratio μ/K with different constant magnetic fields.

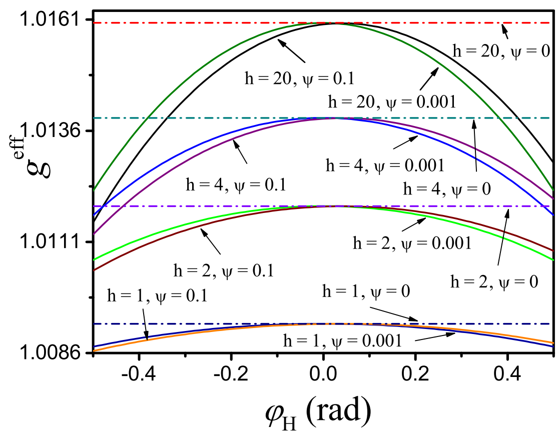

FIG. 5. Dependences of the normalized effective shear modulus $g^{\mathrm{eff}}(\varphi_H)$ on the tilt angle φ_H of the external magnetic field for $\mu/K = 1$, $\gamma_o = 0$ and different values of the normalized magnetic field h. The straight horizontal lines are received from the solution to the linearized problem using Eqs. (9)–(11). Notice that several curves are obtained for the low shear strain $\psi = 0.001$ and compared with curves for the large shear strain $\psi = 0.1$. Notice that curves for the low shear strain for $\psi = 0.001$ appear symmetric with respect to φ_H and the maximum of curves for $\psi = 0.1$ is shifted towards positive φ_H.

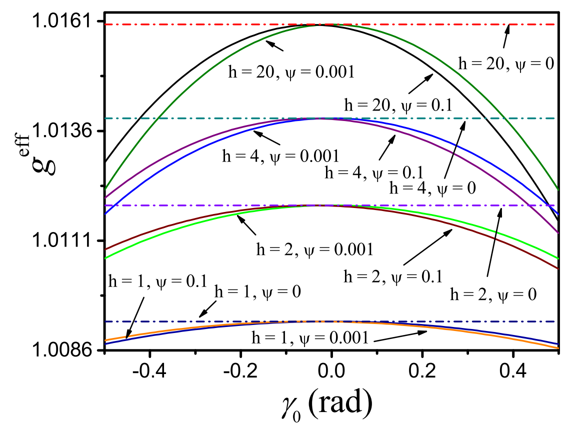

FIG. 6. Dependences of the normalized effective shear modulus $g^{\mathrm{eff}}(\gamma_0)$ on the angle γ_0 for the fixed ratio $\mu/K = 1$, $\varphi_H = 0$, and different values of the normalized magnetic field h. The straight horizontal lines are received from the solution to the linearized problem using Eqs. (9)–(11). Notice that several curves are obtained for the low shear strain $\psi = 0.001$ and compared with curves for the large shear strain $\psi = 0.1$. Notice that curves for the low shear strain $\psi = 0.001$ appear symmetric with respect to $\gamma_0 = 0$ and the maximum of curves for $\psi = 0.1$ is shifted towards negative γ_0.

magnitude of the nonlinearity $g^{\mathrm{eff}}(\varphi_H)$ (deviations from straight lines in Fig. 5), was negligible. Note that, in the calculation of $g^{\mathrm{eff}}(\varphi_H)$, the matrix deforms in a linear fashion, as is implied in (3) and (6). This nonlinearity arises from the influence of the magnetic anisotropy. In real MAEs, an alternative to such nonlinearity can be a loss of coupling between the matrix and the particles at significant particle rotations, which are inevitable in soft matrices for large inclination angles of the external field.

Several curves are obtained for the low shear strain value $\psi = 0.001$. If the shear strain is finite (i.e., the larger value $\psi = 0.1$ is assumed), an asymmetry is observed in the dependence $g^{\mathrm{eff}}(\varphi_H)$. The maximum of the dependence $g^{\mathrm{eff}}(\varphi_H)$ shifts away from the point $\varphi_H = 0$. This shift will be positive if $\psi > 0$ and it will be negative if $\psi < 0$. This leads to the conclusion that when $\psi > 0$ the MAE system in a magnetic field is less rigid for inclinations of the magnetic field in the opposite direction $\varphi_H < 0$ and vice versa. In MAEs, the transition from elastic to plastic deformation is usually observed at strain ψ well above 0.1. For comparison, the elastic deformation range of the tensile strain from 0 to 0.47 has been reported in Ref. [71].

The magnitude of the normalized effective shear modulus $g^{\mathrm{eff}}(\gamma_0)$ depends nonlinearly on the initial orientation angle γ_0 of the axes of anisotropy inclusion particles. Figure 6 shows the dependences $g^{\mathrm{eff}}(\gamma_0)$ obtained for different constant magnetic fields and $\varphi_H = 0$.

As can be seen from Fig. 6, there is a weakly expressed nonlinear dependence of the effective shear modulus of the initial orientation of the anisotropy axes of filler particles. For finite values of shear strain dependence, $g^{\mathrm{eff}}(\gamma_0)$ is not symmetrical with respect to the sign change of γ_0.

The observed asymmetry (see Figs. 5 and 6) was not expected by us. However, we identified it because of the adequate definition of the effective shear modulus (27). This asymmetry has a physical cause. For a tilted field ($\varphi_H \neq 0$), the value of the additional stress created by the magnetic field is different, if the magnetic field has a component directed towards the external shear stress, or against it.

This is a manifestation of a nonlinear behavior of the MAE in a magnetic field. This peculiar effect and variation of g^{eff} with γ_0 or φ_H can be observed only for relatively hard matrices with $\mu/K \geq 1$.

C. Comparison with experiment and critical behavior

Our simplified model described in the preceding sections refers to a particular realization of the composite material which has not been realized in the experiment yet and might be challenging to achieve in practice. However, it should be noted that necessary technological prerequisites for realization of the thin-film MAEs with embedded cylindrical platelets already exist [72,73]. The simple shear testing of a thin-film sample can probably be replaced by a pure shear test method [74]. In the case of tangential stress of incompressible materials, the divergence between simple and pure shear is to be expected at deformations exceeding 30% [75].

In the experiment, the thickness of the MAE film can be controlled by sandwiching it between two parallel plates at the fixed distance. One could suggest first to shear the sample and then constrain it from both sides by two parallel plates. These two parallel plates should not be glued onto the rigid surface because this will destroy boundary conditions on the xy surface of the sample. The rigid surface should be lubricated (e.g., by a silicone oil) to allow slipping of the sample over it. Realization of such an experimental setup would allow one to establish the accuracy of the approximation of plane deformation.

The primary goal of investigating such a theoretical model is that we can identify physical parameters influencing physical effects in more realistic materials. Let us consider as an example the experimental data of Ref. [76] for the shear storage modulus measured as a function of the magnetic flux density B in the range between 0 and 700 mT at a fixed oscillation frequency f of 10 Hz and for different volume fractions of ferromagnetic (iron) filler particles p. This experimental set of data was also taken as the reference in previous theoretical works, e.g., [44]. We are interested in the data for the low concentration of filler particles $p = 0.1$ and isotropic samples. Similarly to our model, the shear storages modulus grows with the increasing magnetic field and indicates saturation in large magnetic fields $B > 400$ mT (cf. Figs. 1 and 3 of [76]). This behavior is well known as the MR effect. It has been shown previously in numerous works that the MR effect can be explained by dipolar interactions between ferromagnetic filler particles. May there be a contribution from the single-particle magnetostriction to the observed MR effect? Our model predicts that the maximum contribution from the single-particle magnetostriction to the shear modulus is of the order of magnitude $\Delta G_{\max} = \Delta G(B \to \infty) \sim pG(B = 0)$, where $\Delta G(B) = G(B) - G(0)$. It is seen from Fig. 1 of [76] that, for small iron particles (mean particle size about 5 μm), $\Delta G_{\max}/G(0) \approx 1.6-1.7$, while for large iron particles (mean particle size about 40 μm), $\Delta G_{\max}/G(0) \approx 1.3$ (cf. Fig. 3 [76]). It can be concluded that the experimentally observed magnetic-field-induced changes of the shear modulus can have a contribution from the single-particle mechanism due to the following two reasons:

(1) Maximum relative magnetic-field-induced changes of the shear modulus in Ref. [76] and in the present paper are of the same order of magnitude;

(2) Dependences of the field-induced shear modulus on the applied magnetic field in Ref. [76] and in the present paper are qualitatively similar.

The origin of this contribution can be shape deviations of filler particles from the perfect sphere (e.g., ellipsoidal or irregular shape). Indeed, the scanning electron microscope photographs of MAE samples presented in Fig. 4 of Ref. [76] do leave such an impression. The MR effect observed at high concentrations ($p \approx 0.3$) of iron particles was of several orders of magnitude higher $\Delta G_{\max}/G(0) \approx 10^2$. In analogy to MR fluids [77], this effect is commonly attributed to rearrangement of filler particles into chainlike aggregates along the magnetic-field lines due to magnetic forces acting between them [1,24,78,79]. This simplified physical picture for high concentrations of magnetizable particles has been recently questioned in Ref. [80], where numerical simulations showed that formation of elongated structures becomes impossible due to purely geometrical constraints.

Calculation of the influence of magnetic field on the effective shear modulus of the MAE was performed in the single-particle approximation, where both elastic and magnetic interactions between the particles can be neglected. Note that in spite of the low concentration of particles, they should not be regarded as solitary because the mechanical deformation is self-consistent. The term $2p\mu(\psi/2 - \gamma)^2$ in the energy density (3) is responsible for the interparticle interaction. It yields that the relationship between the parameters of the problem (particle's rotation γ and shear strain ψ) is described by their product $-p\mu\psi\gamma$. Due to this term, rotation of particles depends on the shearing, and the shearing depends on the rotation of the particles. Indeed, in Eq. (5) for the torques, the angle of the particles' rotation depends on the shearing. In Equation (6) for the stresses, we have that the shear strain depends on the rotation of the particles. Thus, when particles are rotated, there is an interaction between them, which is transmitted via the elastic subsystem.

Accordingly, the expression for the effective modulus (12) includes only the first degree of concentration. It is well known, both experimentally and theoretically, that the effective properties (for a variety of physical situations), when approaching the concentration of the percolation threshold, are strongly increasing functions and behave like the order parameter in the theory of phase transitions [81,82].

Despite the simplicity of the approach used, it is possible to estimate how the concentration dependence of the effective elastic modulus of MAE will behave at higher concentrations. To do this, we resort to the method of Padé approximants [65,82]. We write the concentration dependence of the effective modulus, which is a first-order polynomial, as the ratio of two polynomials.

Already in one of the pioneering papers on MAEs it has been noted that "absolute MR effect in isotropic MR rubbers increases exponentially with increasing iron concentration" [83]. If the concentration of inclusions is increasing, the effective shear modulus goes theoretically to infinity when concentration p approaches particular value p_c. The latter statement is valid even in the absence of an external magnetic field. The method of Padé approximants allows one to estimate the growth rate of effective modulus even if the linear approximation with respect to concentration p is only available. In the simplest case, the Padé approximant (the ratio of two polynomials) can be represented as const $\times (p_c - p)^{-1}$. Refinement of such a choice would be possible in the framework of a theory taking into account higher-order terms with respect to concentration p (p^2, p^3, etc.). Then, according to (17), taking into account that $G^{\text{eff}}(0) = \mu(1 + 2p)$, we have

$$G^{\text{eff}}_{\text{Pade}}(H) = \mu\left[\frac{4\mu + h(K + 4\mu)}{8\mu + h(3K + 8\mu)}\right](p_c - p)^{-1}, \qquad (28)$$

where

$$p_c = \frac{4\mu + h(K + 4\mu)}{8\mu + h(3K + 8\mu)}. \qquad (29)$$

For $p \ll 1$, the expression $G^{\text{eff}}_{\text{Pade}}(H)$ is simplified, as it should be, into (17).

As follows from (29), in high magnetic fields ($H \gg H_A$), p_c is reduced from $\frac{1}{2}$ for $K \ll \mu$ to $\frac{1}{3}$ for $\mu \ll K$. Formula (28) predicts that the magnitude of the magnetic-field-dependent effective shear modulus will grow strongly and nonlinearly with the increasing particle concentration p, which is indeed observed in the experiment. For $H \to 0$, parameter p_c is the well-known percolation threshold, which is somewhat modified in an external magnetic field. Equation (28) should be considered only for $p < p_c$.

Figure 7 shows the dependences of the relative magnetic-field-induced change of the shear modulus, MRE $= [G^{\text{eff}}(H) - G^{\text{eff}}(0)]/G^{\text{eff}}(0)$, on the concentration of filler particles p calculated using Eqs. (28) and (29). It is seen that this relative MR effect (MRE) grows strongly and with

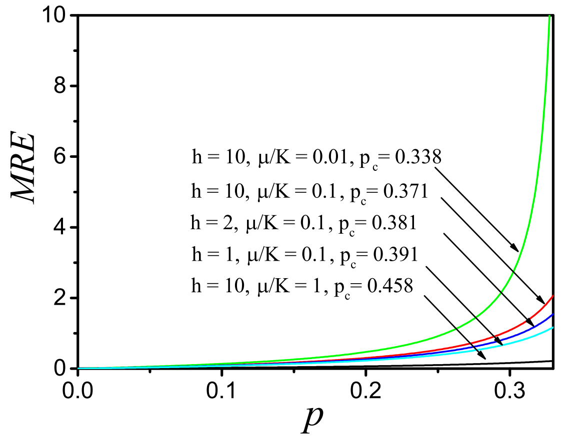

FIG. 7. Dependences of the relative magnetic-field-induced change of the shear modulus, $\mathrm{MRE} = [G^{\mathrm{eff}}(H) - G^{\mathrm{eff}}(0)]/G^{\mathrm{eff}}(0)$, on the concentration of filler particles p for different ratios of μ/K and different normalized magnetic fields h.

increasing concentration p. This result qualitatively agrees with previous experiments on MAEs [84].

In a large magnetic field $H = 10H_A$, MRE is more pronounced for magnetomechanically softer matrices (compare curves in Fig. 7 with different values of μ/K). At a fixed value of μ/K, the relative change of the shear modulus is increasing with the increasing magnetic-field strength H (compare curves in Fig. 7 with different values of H/H_A). Notice the scale on the vertical axis of Fig. 7. The maximum numbers are about two orders larger than the maximum concentration $p \approx 0.3$. This is the result of using the method of Padé approximants. There will be no such strong growth of MRE within the linear approximation with respect to p (17).

VII. CONCLUSION

In this paper, we have studied the effect of the single-particle magnetostriction mechanism on the effective shear modulus of an MAE with a low concentration of ferromagnetic inclusions. The planar (two-dimensional) problem with the inclusions in the form of a disk (platelet) with the axial magnetic anisotropy in the plane of the disk has been solved. It has been shown that any deviation of the magnetic moment of the particle from the easy magnetization axis is accompanied by an enhancement of the effective shear modulus. In particular, the effect of a magnetic field on the effective shear modulus is strongly pronounced in MAEs with a magnetomechanically soft matrix, in which the shear modulus of the matrix is much smaller than the magnetic anisotropy constant. In our model, we were able to demonstrate how the torque of the magnetic field acting on the noncollinear magnetic moment of the particle creates a shear stress. In the phenomenological description, the total torque acting on the particles is determined by the vector product of the particle magnetization in the magnetic-field strength and it corresponds to the antisymmetric tensor of the components of these vectors. It is also shown that the effective shear modulus has a strong nonlinear dependence of the external magnetic field characterized by a saturation effect in high magnetic fields much larger than the magnetic anisotropy field. The experimentally observed magnetic-field-induced increase of the shear modulus at low filler concentrations $p < 0.1$ can have a significant contribution from the proposed single-particle mechanism. The concentration dependence of the effective shear modulus of MAE at higher filler concentrations has been estimated using the method of Padé approximants, which predicts that the magnitude of the magnetic-field-dependent effective shear modulus will significantly grow both absolutely and relatively with the increasing particle concentration p.

[1] G. Filipcsei, I. Csetneki, A. Szilagyi, and M. Zrinyi, Adv. Polym. Sci. **206**, 137 (2007).
[2] Y. Li, J. Li, W. Li, and H. Du, Smart Mater. Struct. **23**, 123001 (2014).
[3] Ubaidillah, J. Sutrisno, A. Purwanto, and S. A. Mazlan, Adv. Eng. Mater. **17**, 563 (2015).
[4] A. M. Menzel, Phys. Rep. **554**, 1 (2015).
[5] S. Odenbach, Arch. Appl. Mech. **86**, 269 (2016).
[6] M. T. Lopez-Lopez, J. D. G. Durán, L. Yu. Iskakova, and A. Yu. Zubarev, J. Nanofluids **5**, 479 (2016).
[7] S. Bednarek, J. Magn. Magn. Mater. **301**, 200 (2006).
[8] X. Guan, X. Dong, and J. Ou, J. Magn. Magn. Mater. **320**, 158 (2008).
[9] K. Danas, S. V. Kankanala, and N. Triantafyllidis, J. Mech. Phys. Solids **60**, 120 (2012).
[10] E. Galipeau and P. Ponte Castañeda, Int. J. Solid Struct. **49**, 1 (2012).
[11] E. Galipeau and P. Ponte Castañeda, Proc. R. Soc. A **469**, 20130385 (2013).
[12] G. Stepanov, E. Y. Kramarenko, and D. Semerenko, J. Phys.: Conf. Ser. **412**, 012031 (2013).
[13] E. Callen and H. Callen, Phys. Rev. **139**, A455 (1965).
[14] P. Morin, D. Schmitt, and E. du Tremolet de Lacheisserie, Phys. Rev. B **21**, 1742 (1980).
[15] V. M. Kalita, A. F. Lozenko, and S. M. Ryabchenko, Low Temp. Phys. **26**, 489 (2000).
[16] V. M. Kalita, A. F. Lozenko, S. M. Ryabchenko, and P. A. Trotsenko, Low Temp. Phys. **31**, 794 (2005).
[17] V. M. Kalita, I. Ivanova, and V. M. Loktev, Phys. Rev. B **78**, 104415 (2008).
[18] V. M. Kalita, A. A. Snarskii, D. Zorinets, and M. Shamonin, Phys. Rev. E **93**, 062503 (2016).
[19] A. Zubarev, Phys. A (Amsterdam, Neth.) **392**, 4824 (2013).
[20] A. Zubarev and D. Yu. Borin. J. Magn. Magn. Mater. **377**, 373 (2015).
[21] G. Diguet, E. Beaugnon, and J. Y. Cavaillé, J. Magn. Magn. Mater. **321**, 396 (2009).
[22] O. V. Stolbov, Y. L. Raikher, and M. Balasoiu, Soft Matter **7**, 8484 (2011).
[23] Y. Han, A. Mohla, X. Huang, W. Hong, and L. E. Faidley, Int. J. Appl. Mech. **07**, 1550001 (2015).
[24] H.-N. An, S. J. Picken, and E. Mendes, Soft Matter **8**, 11995 (2012).
[25] G. V. Stepanov, D. Yu. Borin, Yu. L. Raikher, P. V. Melenev, and N. S. Perov, J. Phys.: Condens. Matter **20**, 204121 (2008).
[26] J. M. Linke, D. Yu. Borin, and S. Odenbach, RSC Adv. **6**, 100407 (2016).

[27] R. Weeber, S. Kantorovich, and C. Holm, Soft Matter **8**, 9923 (2012).
[28] R. Weeber, S. Kantorovich, and C. Holm, J. Magn. Magn. Mater. **383**, 262 (2015).
[29] R. Weeber, S. Kantorovich, and C. Holm, J. Chem. Phys. **143**, 154901 (2015).
[30] G. Pessot, H. Löwen, and A. M. Menzel, J. Chem. Phys. **145**, 104904 (2016).
[31] P. Metsch, K. A. Kalina, C. Spieler, and M. Kästner, Comput. Mater. Sci. **124**, 364 (2016).
[32] E. Jarkova, H. Pleiner, H.-W. Müller, and H. R. Brand, Phys. Rev. E **68**, 041706 (2003).
[33] S. Bohlius, H. R. Brand, and H. Pleiner, Phys. Rev. E **70**, 061411 (2004).
[34] A. Dorfmann and R. Ogden, Eur. J. Mech. A: Solids **22**, 497 (2003).
[35] I. Brigadnov and A. Dorfmann, Int. J. Solids Struct. **40**, 4659 (2003).
[36] D. Ivaneyko, V. Toshchevikov, M. Saphiannikova, and G. Heinrich, Soft Matter **10**, 2213 (2014).
[37] M. R. Dudek, B. Grabiec, and K.W. Wojciechowski, Rev. Adv. Mater. Sci. **14**, 167 (2007).
[38] D. S. Wood and P. J. Camp, Phys. Rev. E **83**, 011402 (2011).
[39] M. A. Annunziata, A. M. Menzel, and H. Löwen, J. Chem. Phys. **138**, 204906 (2013).
[40] G. Pessot, P. Cremer, D. Y. Borin, S. Odenbach, H. Löwen, and A. M. Menzel, J. Chem. Phys. **141**, 124904 (2014).
[41] M. Tarama, P. Cremer, D. Y. Borin, S. Odenbach, H. Löwen, and A. M. Menzel, Phys. Rev. E **90**, 042311 (2014).
[42] P. A. Sánchez, J. J. Cerdá, T. Sintes, and C. Holm, J. Chem. Phys. **139**, 044904 (2013).
[43] J. J. Cerdá, P. A. Sánchez, C. Holm, and T. Sintes, Soft Matter **9**, 7185 (2013).
[44] D. Ivaneyko, V. P. Toshchevikov, and M. Saphiannikova, Soft Matter **11**, 7627 (2015).
[45] B. F. Spencer, S. J. Dyke, M. K. Sain, and J. D. Carlson, J. Eng. Mech. **123**, 230 (1997).
[46] O. Stolbov, Y. L. Raikher, G. Stepanov, A. Chertovich, E. Y. Kramarenko, and A. Khokhlov, Polym. Sci., Ser. A **52**, 1344 (2010).
[47] W. Li, Y. Zhou, and T. Tian, Rheol. Acta **49**, 733 (2010).
[48] L. Chen and S. Jerrams, J. Appl. Phys. **110**, 013513 (2011).
[49] J.-T. Zhu, Z.-D. Xu, and Y.-Q. Guo, Smart Mater. Struct. **21**, 075034 (2011).
[50] F. Guo, C.-B. Du, and R.-P. Li, Adv. Mech. Eng. **6**, 629386 (2014).
[51] T. A. Nadzharyan, V. V. Sorokin, G. V. Stepanov, A. N. Bogolyubov, and E. Y. Kramarenko, Polymer **92**, 179 (2016).
[52] S.-Y. Fu, X.-Q. Feng, B. Lauke, and Y.-W. Mai, Composites, Part B **39**, 933 (2008).
[53] S. Torquato, *Random Heterogeneous Materials. Microstructure and Macroscopic Properties* (Springer Verlag, New York, 2002).
[54] A. V. Ryzhkov, P. V. Melenev, M. Balasoiu, and Y. L. Raikher, J. Chem. Phys. **145**, 074905 (2016).
[55] P. Creemer, H. Löwen, and A. M. Menzel, Phys. Chem. Chem. Phys. **18**, 26670 (2016).
[56] R.W. Ogden and A. Dorfmann, in *Constitutive Models for Rubber IV*, edited by P.-E. Austrell and L. Kari (A. A. Balkema Publishers, Leiden, 2005), p. 531.
[57] L. D. Landau, L. P. Pitaevskii, A. M. Kosevich, and E. M. Lifshitz, *Theory of Elasticity*, 3rd ed., Course of Theoretical Physics Vol. 7 (Elsevier, Oxford, UK, 1986).
[58] H. Sahin, X. Wang, and F. Gordaninejad, J. Intell. Mater. Syst. Struct. **20**, 2215 (2009).
[59] J. Yang, X. Gong, H. Deng, L. Qin, and S. Xuan, Smart Mater. Struct. **21**, 125015 (2012).
[60] X. Dong, N. Ma, M. Qi, J. Li, R. Chen, and J. Ou, Smart Mater. Struct. **21**, 075014 (2012).
[61] I. A. Belyaeva, E. Yu. Kramarenko, G. V. Stepanov, V. V. Sorokin, D. Stadler, and M. Shamonin, Soft Matter **12**, 2901 (2016).
[62] Z. Hashin, J. Appl. Mech. **50**, 481 (1983).
[63] R. M. Christensen, *Mechanics of Composite Materials* (Wiley, New York, 1979).
[64] V. A. Buryachenko, *Micromechanics of Heterogeneous Materials* (Springer Verlag, New York, 2007).
[65] A. A. Snarskii, I. V. Bezsudnov, V. A. Sevryukov, A. Morozovskiy, and J. Malinsky, *Transport Processes in Macroscopically Disordered Media. From Mean Field Theory to Percolation* (Springer Verlag, New York, 2016).
[66] L. D. Landau, L. P. Pitaevskii, and E. M. Lifshitz, *Electrodynamics of Continuous Media*, 2nd ed., Course of Theoretical Physics Vol. 8 (Elsevier, Oxford, UK, 1984).
[67] A. F. Kabychenkov and F. V. Lisiovskii, J. Exp. Theor. Phys. **123**, 254 (2016).
[68] E. Z. Melikhov and R. M. Farzetdinova, J. Exp. Theor. Phys. **122**, 1038 (2016).
[69] H. An, S. J. Picken, and E. Mendes, Polymer **53**, 4164 (2012).
[70] V. V. Sorokin, E. Ecker, G. V. Stepanov, M. Shamonin, G. J. Monkman, E. Y. Kramarenko, and A. R. Khokhlov, Soft Matter **10**, 8765 (2014).
[71] L. Ge, X. Gong, Y. Wang, and S. Xuan, Compos. Science Technol. **135**, 92 (2016).
[72] M. M. Ruiz, M. C. Marchi, O. E. Perez, G. E. Jorge, M. Fascio, N. D'Accorso, and R. M. Negri, J. Polym. Sci., Part B: Polym. Phys. **53**, 574 (2015).
[73] V. Iannotti, G. Ausanio, L. Lanotte, and L. Lanotte, eXPRESS Polym. Lett. **10**, 65 (2016).
[74] G. Schubert and P. Harrison, Polym. Test. **42**, 122 (2015).
[75] D. C. Moreira and L. C. S. Nunes, Polym. Test. **32**, 240 (2013).
[76] H. Böse and R. Röder, J. Phys.: Conf. Ser. **149**, 012090 (2009).
[77] H. See and R. Tanner, Rheol. Acta **42**, 166 (2003).
[78] S. Abramchuk, E. Kramarenko, G. Stepanov, L. V. Nikitin, G. Filipcsei, A. R. Khokhlov, and M. Zrinyi, Polym. Adv. Technol. **18**, 883 (2007).
[79] B. T. Borbath, S. Günther, D. Yu. Borin, Th. Gundermann, and S. Odenbach, Smart Mater. Struct. **21**, 105018 (2012).
[80] D. Romeis, V. P. Toshchevikov, and M. Saphiannikova, Soft Matter **12**, 9364 (2016).
[81] D. Stauffer and A. Aharoni, *Introduction To Percolation Theory*, Rev. 2nd ed. (Taylor & Francis, London, 1994).
[82] H. E. Stanley, *Introduction to Phase Transitions and Critical Phenomena*, International Series of Monographs on Physics, Rev. Ed. (Oxford University Press USA, New York, 1971).
[83] B. Stenberg, M. Lokander, and T. Reitberger, Ann. Trans. Nordic Rheol. Soc. **12**, 163 (2004).
[84] A. Stoll, M. Mayer, G. J. Monkman, and M. Shamonin, J. Appl. Polym. Sci. **131**, 39793 (2014).