Colossal Magnetoelastic Effects in Magnetoactive Elastomers
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Abstract

Composite materials where soft-magnetic micrometer-sized particles are embedded into a compliant polymer matrix are known as magnetoactive or magnetorheological elastomers. They are distinguished by huge variations of their physical properties in a magnetic field, which is commonly attributed to the restructuring of the filler. The process of the magnetic-field-induced restructuring in a magnetorheological elastomer is interpreted as a progression towards percolation. The theoretical approach is built upon a self-consistent effective-medium theory for the elastic properties, extended to the variable (field dependent) percolation threshold. The proposed model allows one to describe the large variations (over several orders of magnitude) of the effective elastic moduli of these composite materials, known as the giant magnetorheological and field-stiffening effects. An existence of a giant magnetic Poisson effect is predicted.

Magnetoactive elastomers (MAEs) are a class of composite materials where significant changes of physical properties are observed in moderate (several hundred mT) dc magnetic fields [1-7]. The most prominent phenomenon is the magnetorheological (MR) or field-stiffening effect, when the elastic moduli of these materials grow over several (up to four) orders of magnitude, respectively. In this context, these materials are often referred to as magnetorheological elastomers. MAEs consist of rigid, ferromagnetic magnetic particles (usually of a spherical shape)
dispersed in a compliant (soft) polymer matrix. It is essential that the elastic moduli of the inclusions (e.g. iron, Young’s modulus $Y_{Fe} \approx 2 \cdot 10^{11}$ Pa) are many orders of magnitude larger than those of the matrix (e.g. polydimethylsiloxane (PDMS) $Y_{PDMS} \sim 10^3 – 10^5$ Pa).

Large theoretical efforts have been made for explaining the giant [8] (or even colossal [9]) MR effect in MAEs [10, 11]. The general approaches can be roughly divided into two different groups: i) the cause is in the interaction between induced magnetic moments between the particles [12] (here the simplest model is a system of point dipoles connected by elastic springs [13]); ii) the cause is the rotation of individual particles [14-17]. Although these mechanisms are undoubtedly contribute to the MR effect in MAEs, the existing approaches are not satisfactory when it comes to the description of the MR effect over several (say, three) orders of magnitude. As an example, a 20-fold field-induced increase of the moduli has been calculated in [18].

The purpose of this paper is to propose an alternative physical model based on the idea that the restructuring of the filler [19-21] means the progression towards the percolation structure. Figure 1 illustrates the proposed physical picture [22]. When a magnetic field (purple arrow) is applied to the composite, a significant part of the magnetic flux passes through the pre-cluster (shaded purple region). The ferromagnetic particles in a vicinity of the pre-cluster are “sucked” towards pre-cluster by magnetic forces.

**Figure 1.** Artist’s impression of the restructuring in an applied magnetic field.
Such a reconfiguration of the microstructure is quantitatively characterized by the difference between the total particle concentration $p$ and the field-dependent percolation threshold (PT) $p_c$. Although we do not use the methods of percolation theory [23], the modified effective medium theory (EMT [24]) comprehends the existence of the PT (i.e. a particular concentration where there is a steep rise of elastic modulus) and its dependence of an external magnetic field. It will be shown below that our model is capable of predicting the correct order of magnitude of the MR effect, while keeping the qualitative behavior unaltered. As a mathematical instrument, we will employ the well known self-consistent EMT for the elastic properties, modified in such a way that a steep change in an elastic modulus occurs at a given concentration of rigid inclusions. We utilize the previously introduced concept of the moveable (field dependent) PT [22] and use the empirical dependence of the PT on a magnetic field found in [25].

$G_e, \nu_e$ are the effective shear modulus and the Poisson’s ratio, respectively, while $G_1, G_2, \nu_1, \nu_2$ are the values of these moduli in the first and second phases.

We assume iron ($G_1 = 0.7 \times 10^{11}$Pa, $\nu_1 = 0.29$) as a ferromagnetic filler embedded in a very soft polydimethylsiloxane matrix ($G_2 = 1.3 \times 10^3$Pa, $\nu_2 = 0.49$) [9]. With these materials, the volume concentration $p = 0.23$ corresponds to the typical concentration of iron particles about 70 mass%.

The system of equations of the classical self-consistent EMT [26, 27] can be written in the following form:

$$
\begin{align*}
\Omega_1 p + \Omega_2 (1-p) &= 0 \\
\Theta_1 p + \Theta_2 (1-p) &= 0
\end{align*}
$$

where

$$
\Omega_3 = \frac{G_e \left(1+\frac{1+\nu_e}{G_e} \cdot \frac{1-2\nu_e}{1-2\nu_i} \right)}{1+\alpha_e \left(\left(\frac{G_i}{G_e}\right) \cdot \left(1+\frac{1+\nu_i}{G_e} \cdot \frac{1-2\nu_i}{1-2\nu_e} \right) - 1\right)},
\Theta_3 = \frac{G_e - 1}{1+\beta_e \left(\left(\frac{G_i}{G_e}\right) - 1\right)},
\alpha_e = \frac{1}{3} \cdot \frac{1+\nu_e}{1-\nu_e},
\beta_e = \frac{2}{15} \cdot \frac{4-5\nu_e}{1-\nu_e}.
$$
Very recently, it was proposed to modify the equations of EMT for the elasticity problem in the following way [28]:

\[
\begin{align*}
\frac{\Omega_1}{1 + s(p, \tilde{p}_c) \Omega_1} p + \frac{\Omega_2}{1 + s(p, \tilde{p}_c) \Omega_2} (1 - p) &= 0, \\
\frac{\Theta_1}{1 + s(p, \tilde{p}_c) \Theta_1} p + \frac{\Theta_2}{1 + s(p, \tilde{p}_c) \Theta_2} (1 - p) &= 0.
\end{align*}
\] (3)

Similar modification was established for galvanomagnetic phenomena in [29]. The term \( s(p, \tilde{p}_c) \) is

\[
s(p, \tilde{p}_c) = (1 - 2 \tilde{p}_c) \left( \frac{p}{\tilde{p}_c} \right) \tilde{p}_c \left( \frac{1 - p}{1 - \tilde{p}_c} \right)^{-1}. \] (4)

It allows one to set the PT of the first phase \( p_c \) to \( \tilde{p}_c \).

In [25], the following empirical relationship was proposed:

\[
\tilde{p}_c \left( \langle H \rangle \right) = \tilde{p}_c (0) e^{-\frac{\langle H \rangle}{H_c}},
\] (5)

where \( H \) is the magnetic field inside the composite material, \( H_c \) is the characteristic magnetic field strength, \( \langle ... \rangle = 1/V \int ... dV \), \( V \) is the averaging volume, wherein the characteristic dimensions of the averaging region should be much larger than the correlation length. In the following, we denote \( \langle \langle H \rangle \rangle \) as \( H \). The order of magnitude of \( H_c \) was found to be \( 10^5 - 10^6 \) A/m [22, 25]. To the best of our knowledge, the hypothesis of the field dependence of the percolation threshold, for the case of magnetorheological fluids, was introduced in [30].

In [25], equation (5) was used in the percolation formula \( G \sim (p - \tilde{p}_c)^\gamma \), \( p > \tilde{p}_c \). However, percolation formulas work only in the very narrow region (critical region) in a vicinity of the PT. In [22], a modified EMT allowed one to consider the entire concentration range up to the PT and provided a good agreement with measurements of dielectric and magnetic properties of MAEs.
The system of transcendental equations (1) is solved numerically, for given values of parameters $p, h, \tilde{p}_c(0), G_1, G_2, \nu_1, \nu_2$ and $H_c$.

Figure 2. Field dependence of the percolation threshold.

Figure 2 shows the dependence of the field-dependent PT $\tilde{p}_c(H)$ on the magnetic-field strength $H$ for a number of various values of the critical field $H_c$. In the region $p < \tilde{p}_c(H)$, the model (1) – (5) describes a composite material with inclusions of the phase 1 embedded into a matrix (phase 2). Above the PT, the equations describe inclusions of the first phase in the second phase, which does not correspond to the MAE structure. In the following, the solutions above the PT are shown by dashed lines. Vertical dashed lines in Figure 1 indicate the maximum value of the magnetic field $H_{\text{max}}$, where the solution of (4) correspond to the microstructure of interest. It can be easily obtained that $H < H_{\text{max}} = H_c \ln(\tilde{p}_c(0)/p)$.

Figure 3 shows the calculated MR effect defined as

$$MRE(H) = \frac{G_e(H) - G_e(H = 0)}{G_e(H = 0)}.$$  \hspace{1cm} (7)
The parameters, displayed in the Figure, are taken from [22], where they have been used to describe the magnetic properties of a real MAE and provided an excellent agreement between numerical and experimental results.

It is observed that the MR effect grows with an increasing magnetic field and tends to saturate in large magnetic fields. The effect is bigger for the larger concentration of inclusions. Within the applicability region of the model with respect to the microstructure of the material, the effect reaches the order of magnitude of $10^3$.

![Figure 3](image)

**Figure 3.** Field dependence of the magnetorheological effect.

Moreover, the model predicts a giant variation of the Poisson’s ratio with the applied magnetic field (Figure 4). We take courage to name this phenomenon – magnetic Poisson effect (MPE) and encourage experimentalists to verify our prediction. It is quantified as

$$
MPE(H) = \frac{v_e(H) - v_e(H = 0)}{v_e(H = 0)}.
$$

(8)
We excluded other possible mechanisms (magnetic forces between magnetized particles, rotations of particles due to an applied magnetic field) from our consideration. The calculated effect is solely due to restructuring of the filler. The order of magnitude of the MRE effect in ultra-soft MAEs is explained. For the refinement of the results, one has to take into account the field-induced anisotropy of effective properties and the alternative mechanisms, which should also contribute to the field-stiffening or magnetorheological effects (by further enhancement) when a significant amount of particles will be in the pre-cluster region in a vicinity of each other.

Our model allows one to describe in a unified manner magnetic properties, magnetodielectric effect [22] and, henceforth, to explain the mechanism of the giant increase in elastic properties in MAEs.

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