Diversifying temporal responses of magnetoactive elastomers

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Abstract

Magneactive elastomers (MAEs) are able to deform significantly in response to the application of magnetic fields. Usually, a magnetic field which is harmonic in time usually results in a harmonic mechanical response of the MAEs. To render MAEs with the ability of responding or deforming diversely or anharmonically in time, in this work, we propose a hybrid MAE which is based on a rubber matrix embedded with both soft iron particles and hard NdFeB-alloy particles. Firstly, based on the principle of minimum free energy, we establish a theoretical model to study magnetomechanical behaviors of the proposed hybrid MAEs. Then, through both theoretical and experimental studies, we show that the response of a hybrid MAE sample to the applied magnetic field is usually complex, i.e. the deformation induced by a harmonic magnetic field in time is anharmonic. At last, the effect of two main factors, the state of magnetization and the amplitude of the applied magnetic field, is studied both experimentally and theoretically. This work provides a new idea of diversifying the temporal response of MAEs to the application of harmonic magnetic fields (harmonic in time). The hybrid MAEs may serve as a complement to the recently proposed 3D-printed hard MAEs which are able to deform inhomogeneously in space in response to a uniform magnetic field.

1. Introduction

In recent years, using the magnetic field to actuate or control the motion of materials is a new focus in soft active materials or soft robotics (Hu et al 2018, Kim et al 2018, Yoonho et al 2019, Liu et al 2019, Ren et al 2019). An interesting and well known example of magnetic active materials is MAE, an elastomer embedded with magnetic particles, owing to its large deformation in response to the applied magnetic field (Hu et al 2018, Kim et al 2018, Zhao et al 2011). Due to such strong magneto-elastic coupling, MAEs have found a wide range of applications in controllable actuators, rapid response regulators for mechanical systems, adaptively tuned vibration absorbers, and stiffness tunable mounts (Ivaneyko et al 2012, Filipcei et al 2007, Yang and Sun 2014, Opie and Yim 2009). Early efforts in MAEs are mainly focused on producing larger deformation of materials using a relatively small magnetic field $\mathbf{H}$. For example, under a magnetic field of 120 kA m$^{-1}$, 1.5% magnetostriction strain was obtained in a MAE bar embedded with iron particles at a volume ratio of 10% (Coquelle and Bossis 2005). Theoretical studies indicate that the magnetic force that deforms the material stems from the Maxwell stress $\sigma_{MW}$ which is usually on the order of kPa (Liping and Pradeep 2013). Thus, the material should be soft enough to exhibit significant deformation. Also, since the magnetic field induced Maxwell stress increases with the permeability of the material, particles with high permeability, such as iron and iron oxides, were usually used to fabricate MAEs (Guan et al 2008, Riebi and Jilken 1983, Zrinyi et al 1996). However, for a free standing MAE sample in a uniform magnetic field, the deformation is always uniform. To deform the sample inhomogeneously, a common way is to apply inhomogeneous mechanical constraints (mostly through boundary conditions), which is both inefficient and inconvenient (Feng et al 2017, 2018, Qi et al 2018).
More recently, a new thought of producing controllable or programmable inhomogeneous deformation in MAEs has been proposed based on the idea of substituting soft-magnetic particles (iron or iron oxides materials) with hard-magnetic particles (neodymium-iron-boron (NdFeB)) (Kim et al. 2018, Lum et al. 2016). Owing to the ability of retaining high residual magnetic flux density \( \mathbf{b}' \), hard magnetic particles were doped in elastomers to produce complex deformation of a free standing sample in a uniform magnetic field (Hu et al. 2018). To accurately control the complex deformation of such hard magnetic particles embedded MAEs (H-MAEs), a 3D printing technique for programming ferromagnetic domains were reported (Kim et al. 2018). Theoretically, Zhao et al. reformulate the mechanics of H-MAEs based on the framework of continuum mechanics (Zhao et al. 2018). In their work, since the permeability of the material is very close to 1, the magnetic Maxwell stress was ignored. While another kind of magnetic stress caused by the interaction between hard-magnetic particles and the magnetic field was proposed in Zhao et al. (2018). It indicated that the deformation of an H-MAE sample was mainly caused by such magnetic stress. To distinguish from the magnetic Maxwell stress, in this work, we call the magnetic stress proposed by Zhao et al.’ remanence stress’ since it is related to the residual magnetization of the material.

Motions in the real world are always complex. To meet the requirement for different potential applications, it may be necessary to diversify MAEs’ responses, both in space and in time, without manipulating the magnetic stimulation too much. Thus, other than producing inhomogeneous, complex deformation of free standing MAE samples, it’s also desirable to render materials with the ability of responding or deforming diversely in time. In previous works, a most famous and straightforward way to diversify the temporal responses of a system is to introduce nonlinear dynamics (Qian et al. 2019, Kim and Seok 2014, Zhou and Zuo 2018, Zhou et al. 2017). But, it often requires a complex structural design and usually works for certain frequencies. In this paper, we want to provide a new idea to diversify the temporal responses of a system at any frequency. For the case of H-MAEs, the remanence stress \( \sigma_{RE} \) is proportional to the magnetic stimulation \( \mathbf{h}' \). If \( \mathbf{h}' \) is small and the geometrical nonlinearity is negligible, the system would run almost linearly with respect to the applied field. While for the case of soft MAEs (an elastomer embedded with soft magnetic particles), since the Maxwell stress \( \sigma_{MW} \) is always proportional to \( (\mathbf{h}')^2 \), the system’s response should always be nonlinear no matter how small \( \mathbf{h}' \) is. As can be seen in figure 1, in a harmonic magnetic field \( \mathbf{h}' \), the deformation for the H-MAE disc can be positive or negative depending on the direction of \( \mathbf{h}' \), while the deformation for the soft MAE disc is always positive regardless of the direction of \( \mathbf{h}' \). Consequently, the H-MAE disc deforms synchronously with \( \mathbf{h}' \), while the soft MAE disc deforms at a frequency twice that of \( \mathbf{h}' \).

To take advantages of both hard and soft MAEs, in this work, we mixed hard and soft magnetic particles into the same elastomer(named as a hybrid MAE) and measured its responses to a harmonic magnetic field. Currently, although theories for both soft MAEs and H-MAEs have been proposed separately (Kankanala and Triantafyllidis 2004, Liu 2014, Zhao et al. 2018), a general theory considering both of these two materials is still missing. Thus, we firstly formulate the problem in the frame of continuum mechanics and follow the free energy minimization approach in Liu’s work (Liu 2014) to derive the governing equations. Subsequently, the deformation of a transverse isotropic material is predicted under an 1 Hz sinusoidal magnetic field. Compared with the soft MAEs and the H-MAEs, the deformation of the hybrid is more diverse. Finally, we design an experiment to verify our theoretical prediction.

Figure 1. (a) The schematic of the H-MAE and the soft MAE. (b) The deformation of MAEs under a sinusoidal magnetic field. The black line, the black marked line and the red marked line represent the applied magnetic field(\( h' \)), the deformation of the H-MAE and the deformation of the soft MAE, respectively.
2. Energy formulation and Euler–Lagrange equations of a continuum body in magnetic fields

In this section, we establish the Euler–Lagrange equations and boundary conditions for a hybrid MAE through the principle of free energy minimization. Consider a deformable continuum body as shown in Figure 2. Let the three-dimensional region occupied by the undeformed body be denoted $V$, with the boundary $\partial V$. We refer to the undeformed body as the reference configuration and the deformed body as the current configuration. The position of a material point $A$ in $V$ can be described by the coordinate system $X_K$ ($K = 1, 2, 3$). After deformation $\chi$, the material point $A$ moves to a new position which is described by $x_k$ ($k = 1, 2, 3$), a new coordinate system associated with the deformed body that occupies the region $v$. In the traditional continuum mechanics, we call $X_K$ the material or Lagrangian coordinates and $x_k$ the spatial or Eulerian coordinates. The base vectors for the material coordinate system and the spatial coordinate system are respectively denoted by $I_K$ and $i_k$. The deformation $\chi$ may be interpreted as the mapping from the material coordinates to the spatial coordinates, $\chi = x(X)$. Based on the above setup, the deformation gradient is defined as $F = \frac{\partial x}{\partial X}$.

Throughout the whole work, we use, 'K' and, 'k' to represent the partial derivatives with respect to the coordinates $X_K$ and $x_k$, respectively. The determinant of the deformation gradients is the so-called Jacobian which is denoted by $\det F = J$. The mass density field $\rho_0$ of the undeformed body is related to its counterpart $\rho$ in the deformed body by $\rho_0 = J \rho$ due to the assumption of local mass conservation.

In the current configuration, the Maxwell equations are

$$\varepsilon_{kln} h_{lk} = 0, \quad \text{and} \quad b_{lk} = (\mu_0 h_k + \mu_0 m_k + b'_k)_{lk} = 0,$$

where $\varepsilon_{kln}$ is the alternating symbol, $b_k$ is the magnetic flux density, $\mu_0$ denotes the permeability of the vacuum, $h_k$ is the magnetic field, $m_k$ is the magnetization, and $b'_k$ is the residual magnetic flux density. For convenience, let $h_k = -\xi_{k}$, in which $\xi$ is the magnetic potential.

One may define the following Lagrangian counterparts of $h_k$, $m_k$, $b_k$ and $b'_k$ as

$$H_k = h_k x_{k,K}, \quad M_k = (\mu_0 + \mu_0 m_k) b'_k, \quad B_k = \mu_0 H_k X_{K,k}, \quad B'_k = \mu_0 H_k X_{K,k} b'_k.$$  

So we have

$$B_k = \mu_0 J X_{K,l} H_k X_{K,k} + \mu_0 X_{K,l} M_k \alpha_{k,k} + B'_k,$$

where $B_k$ denotes the nominal magnetic flux density, $H_k$ denotes the nominal magnetic field, $M_k$ is the nominal magnetization, $\alpha_{k,k}$ is the shifter which may be viewed as the rotation from the coordinate frame $I_k$ to $i_k$, and $B'_k$ is the nominal residual magnetic flux density. Then, the Lagrange form of Maxwell equations can be written as

$$\varepsilon_{kln} H_{L,k} = 0, \quad \text{and} \quad B_{K,K} = 0.$$

2.1. Free energy of the system

The total free energy of a magnetomechanical system can be expressed as (Liu 2014)

$$\mathcal{F}[x_k, M_k] = \mathcal{W}[x_k, M_k] + \mathcal{E}^{\text{mag}}[x_k, M_k] + \mathcal{E}^{\text{mech}}[x_k].$$

Figure 2. (a) The reference configuration. (b) The current configuration.
Here, $\mathcal{H}[x_k, M_k]$ is the internal energy given by the following form:

$$\mathcal{H}[x_k, M_k] = \int_V \Phi(x_k, M_k) dV,$$

where $\Phi(x_k, M_k)$ is the internal energy function. Since a magnetized solid changes the magnetic field of the free space around it, the magnetic energy ($\mathcal{E}^{\text{mag}}$ in equation (5)) can be written as

$$\mathcal{E}^{\text{mag}} = \frac{\mu_0}{2} \int_{\mathbb{R}^3} h_k h_k dv - \int_{s} (\mu_0 h_k^c m_k + h_k^c b_k^r) dV,$$

where $\mathbb{R}^3$ is the total space which is the sum of the domain of the deformed body plus the free space around it, and $h_k^c$ is the externally applied magnetic field. Note that the first term on the right hand side of equation (7) corresponds to the energy associated with the change of the magnetic field by the magnetized body, and the second term corresponds to the energy of the external magnetic field. For the convenience of calculation, it is helpful to decompose the magnetic field $h_k$ into the externally applied field $h^e$ and the perturbation field $h^{\text{eff}}$. Finally, $\mathcal{G}^{\text{mech}}$ in equation (5) is defined by

$$\mathcal{G}^{\text{mech}}[x_k] = -\int_{V} \frac{1}{2} \rho \dot{x}_k \dot{x}_k dv - \int_{s} \rho f_k x_k dv - \int_{s} t_k x_k n_k da,$$

where $\dot{x}_k = \frac{\partial x_k}{\partial t}$ denotes the velocity, $f_k$ is the body force and $t_k$ is the surface traction.

### 2.2 Euler–Lagrange equations and boundary conditions

By the principle of minimum free energy, the equilibrium state of the system is determined by

$$\text{Min}_{(x_k, M_k) \in \mathcal{S}} \mathcal{F}[x_k, M_k]$$

with the constraint of Maxwell equations (1) or (4), where $\mathcal{S}$ is the admissible space for the state variables.

To obtain the Euler–Lagrange equations associated with a minimizer of equation (9), we consider the variations of

1. magnetization
   $$x_k \rightarrow x_k, \quad M_k \rightarrow M_k + \delta M_k,$$
   (10)
2. deformation
   $$x_k \rightarrow x_k + \delta u_k, \quad M_k \rightarrow M_k,$$
   (11)

where $\delta$ is a small number which controls the magnitude of the variations, $\delta M_k$ and $\delta u_k$ are the admissible variations of the field variables $M_k$ and $x_k$, respectively. By applying variations (10) and (11) to (5) respectively, the Euler–Lagrange equations and associated boundary conditions are given by

$$\begin{align*}
\frac{\partial \mathcal{L}}{\partial M_k} &= \mu_0 H_L X_{LM} \delta M_k & \text{in } V \\
\left(\frac{\partial \mathcal{L}}{\partial u_k} + \Sigma_{\text{ME}} + \Sigma_{\text{RE}}\right)_R + \rho_0 f_k &= \rho_0 \frac{\partial^2 x_k}{\partial t^2} & \text{in } \mathbb{R}^3 - V \\
\left(\frac{\partial \mathcal{L}}{\partial r_R} + \Sigma_{\text{ME}} + \Sigma_{\text{RE}}\right)_R &= 0 & \text{on } \partial V
\end{align*}$$

(12)

where $\Sigma_{\text{ME}}$ is the first Piola-Maxwell stress, $\Sigma_{\text{RE}}$ is the first Piola-remanence stress, $[[I]] \equiv I_r - L_i$ is the jump of the quantity $I$ evaluated at either side of the discontinuity surface, $T_{\partial R}$ is the reference traction on the boundary (force per unit reference area), $N_R$ is the outward unit normal vector to the surface in the reference configuration, and $\mathbb{R}^3 - V$ is the free space around the deformed body. The detailed derivation can be found in appendices A and B. $\Sigma_{\text{ME}}$ and $\Sigma_{\text{RE}}$ can be written as

$$\Sigma_{\text{ME}} = -\frac{\mu_0}{2} J_{X_{LM}}(H_L^{\text{eff}} X_{LM} + H_k^{\text{eff}} X_{KL} + H_k^{\text{eff}} X_{KM} + H_k^{\text{eff}} X_{KLM}) + \mu_0 J_{H_L^{\text{eff}} X_{LM} + H_k^{\text{eff}} X_{KL} + H_k^{\text{eff}} X_{KM} + H_k^{\text{eff}} X_{KLM}},$$

and

$$\Sigma_{\text{RE}} = -H_k^{\text{eff}} B_k^R.$$

The corresponding Euler form can be written as

$$\sigma_{\text{ME}} = h_k b_k^l - \frac{\mu_0}{2} (h_k h_l) b_{kl},$$

$$\sigma_{\text{RE}} = -h_k^{\text{eff}} b_k^l,$$

(15)
where \( b_k^l = b_k - b^l \), and \( \delta_{kl} = \mathbf{i}_k \cdot \mathbf{i}_l \) is the Kronecker delta which is non-zero only if \( k = l \). The same form of the Maxwell stresses can be found in Ball (1976)'s works. Using the relationship between the Cauchy stress and the first Piola-Kirchhoff stress, the Euler–Lagrange equations (12) can be further expressed in its Euler form as

\[
\begin{align*}
\frac{\partial \phi}{\partial m_k} &= \mu_0 h_k, & \text{in } V \\
\left( J^{-1} \frac{\partial \phi}{\partial x_{kR}} \mathbf{x}_{lR} + \sigma_{kl}^{MW} + \sigma_{kl}^{RE} \right)_k + Jp_f = J\mu_0 \frac{\partial^2 \mathbf{x}_k}{\partial t^2}, & \text{in } \mathbb{R}^3 - V, \\
(\sigma_{kl}^{MW})_k &= 0, & \text{in } \mathbb{R}^3 - V, \\
\left[ J^{-1} \frac{\partial \phi}{\partial x_{kR}} \mathbf{x}_{lR} + \sigma_{kl}^{MW} + \sigma_{kl}^{RE} \right] n_l &= \mathbf{t}_{kl} n_l, & \text{on } \partial V
\end{align*}
\]

where \( n_l \) is the outward unit vector normal to the surface in the current configuration.

### 2.3. Transverse isotropic nonlinear materials

Now we study a transverse isotropic hybrid MAE plate subjected to a uniform magnetic field \((h^*)\) parallel to \(X_1\) direction, in which the residual flux density \(B_r\) is along the thickness direction (or \(X_1\) direction), as presented in figure 3. The domain occupied by the hybrid MAE plate, in the reference configuration, is given by

\[
V = \{ X \in \mathbb{R}^3; 0 \leq X_1 \leq L, X_2^2 + X_3^2 \leq \pi R^2 \},
\]

where \(L\) is the thickness of the hybrid MAE plate and \(R\) is the radius of the hybrid MAE plate. Under an applied uniform magnetic field, \(L\) and \(R\) become \(l\) and \(r\). Here, we presume that the magnetic field changes slowly with time in order to separate the nonlinear dynamics from the phenomenon. So, the solution can be regarded as quasi-static. In addition, since the transverse isotropic film is thin \((L\) is much smaller than \(R\)), we assume its deformation is uniform and the deformation gradient is given by

\[
\mathbf{F} = \begin{bmatrix} \lambda_1 & 0 & 0 \\ 0 & \lambda_2 & 0 \\ 0 & 0 & \lambda_3 \end{bmatrix},
\]

where \(\lambda_i (i = 1, 2, 3)\) is the stretch in the \(X_i\) direction and \(\lambda_2 = \lambda_3\). We assume that the material is magnetostatically linear. The constitutive relation of the films is given by

\[
m = (\mu_r - 1) h.
\]

where \(m\) and \(h\) are the magnetization and the magnetic field in the \(X_1\) direction, respectively. The stored or internal energy density function of the material is given by the following form (Alameh et al 2018, Liping and Pradeep 2013, Krichen et al 2017):

\[
\Phi(\lambda_i, M) = W_{eh}(\lambda_i) + \frac{\mu_0 M^2}{2(\mu_r - 1)} J,
\]

where \(M\) is the counterpart of \(m\) defined in the reference configuration. We use the following Neo-Hookean hyperelasticity model to describe the mechanical property of the material (Treloar 1949, Deng et al 2014):
where \( c_1 \) is the shear modulus, and \( k \) is the bulk modulus. For the 1D magneto-mechanical system, the total free energy equation (5) can be expressed as

\[
\mathcal{F} = \pi R^2 \int_0^L \left( W_{\text{clast}} + \frac{\mu_0 M^2}{2(\mu_r - 1)} \right) dX_i + \pi R^2 \int_0^L \frac{\mu_0 J}{2 \lambda_1} (\xi_{\text{self}})^2 dX_i \\
- \pi R^2 \int_0^L \mu_0 h^2 M dX_i - \pi R^2 \int_0^L h^2 B' \lambda_1 dX_i + \pi R^2 \int_0^L q(J - 1) dX_i,
\]

where the last term is a constraint to the incompressible materials (Deng et al 2014), \( q \) is the Lagrangian multiplier which can be interpreted as the hydrostatic pressure. We assume that the relative permeability of the free space around the deformed body is equal to 1 and the perturbation field outside \( V \) vanishes because of the thin-film geometry (Coey 2011, Alameh et al 2015). The magnetic field and the perturbation field in the current configuration are given by

\[
\begin{align*}
    h &= \frac{h'}{\mu_r}, \quad 0 < x_i < l, \\
    h &= h', \quad \text{otherwise,}
\end{align*}
\]

and

\[
\begin{align*}
    -\xi_{\text{self}} &= -\frac{\mu_r - 1}{\mu_r} h', \quad 0 < x_i < l, \\
    -\xi_{\text{self}} &= 0, \quad \text{otherwise.}
\end{align*}
\]

Minimizing equation (22), carrying out the appropriate variational calculations with respect to \( \lambda_1, \lambda_2, \) and \( M \) yields the governing equations (the detailed derivations can be found in appendix C):

\[
m = (\mu_r - 1) h,
\]

and

\[
\xi \lambda_1 \left( \lambda_1 - \frac{1}{\lambda_1^2} \right) - \mu_0 \left( \frac{\mu_r - 1}{\mu_r} h' \right)^2 - h' \lambda_1 B'_r = 0.
\]

To explore the response of a hybrid MAE to a harmonic magnetic field, as shown in figure 4(a), we assume that the frequency of the magnetic field is 1 Hz. As an example, the material properties used as in the calculation are listed below: \( c_1 = 93 \) kPa, \( B' = 1.2 \) mT, \( \mu_r = 1.2 \). The amplitude of the applied magnetic field is 400 Oe. By numerically solving equation (26), the strain is presented in figure 4(b). The result indicates that the deformation is non-sinusoidal under the sinusoidal magnetic field. Compared with the pure soft MAEs or H-MAEs (shown by figure 1(b)), we conclude that the hybrid MAEs may exhibit more diverse responses to a harmonic magnetic field, which provides us more freedom in designing magnetic actuating devices.

Figure 4. The deformation diagram of a hybrid MAE under a sinusoidal magnetic field. (a) The applied magnetic field. (b) The strain of the hybrid MAE in \( X_1 \) direction.

\[ W^{\text{clast}} = \frac{c_1}{2} \left[ J^{-2} (\lambda_1^2 + \lambda_2^2 + \lambda_3^2) - 3 \right] + \frac{k}{2} (J - 1)^2, \]
To verify our theoretical predictions for the performance of hybrid MAEs, in this section, we design the following experiments: (1) the sample is loaded by the magnetic field with different amplitudes (section 3.1), 2 samples with different magnetization states are loaded by the same magnetic field (section 3.2).

In the experiment, the Ecoflex-0010 from Smooth-on Inc. was used as a matrix material. Iron powders with the mean diameter of ~0.5 μm were bought from NANGONG XINDUN ALLOY WELDING MATERIAL SPRAYING CO., China and used as the magnetically soft component. Neodymium-iron-boron (NdFeB) alloy powders with the mean diameter of ~38 μm were bought from GUANZHOU XINNUODE TRANSMISSION PART CO., China and used as the magnetically hard component. The overall concentration of the filler in all samples was 50wt% with the ratio between the magnetically soft and hard components being 1:1. Hybrid MAEs were prepared by mixing neodymium-iron-boron (NdFeB) and iron micro-particles with silicone elastomer (Ecoflex 00-10) at the aforementioned weight fraction. The mixture was poured into a mold to obtain desired geometry and then cured at 80 °C for 10 min, in which the thickness (L) of the sample is 5 mm and the radius (R) is 20 mm. The cured material was uniformly magnetized along the thickness direction by applying an impulse magnetic field to produce $B^r$ in materials. The magnitude of $B^r$ can be tuned by changing the peak value of the pulsed magnetic field. The relationship between $B^r$ and the peak value of the pulsed magnetic field is presented in table 1. As shown in figure 5, the sample was loaded by using a customized solenoid that generated the AC magnetic field $h^e$ and deformations of these materials were detected by using a Laser Scanning Vibrometer (Polytec OFV-5000, Germany). The experiments were carried out on the vibration isolation workstation (M-VIS3048-IG2-125A, Newport, American). Throughout the whole work, the frequency of the magnetic field is fixed to 1 Hz.

### Table 1. The residual magnetic flux density as a function of the peak value of the pulsed magnetic field

| peak value of the pulsed magnetic field (Oe) | 0 | 3000 | 6000 | 9000 |
|-------------------------------------------|---|------|------|------|
| Residual magnetic flux density (mT)       | 0 | -0.362 | -2.194 | -11.486 |

### 3. Experimental observations of the behavior of hybrid MAEs

To verify our theoretical predictions for the performance of hybrid MAEs, in this section, we design the following experiments: (1) the sample is loaded by the magnetic field with different amplitudes (section 3.1), 2 samples with different magnetization states are loaded by the same magnetic field (section 3.2).

In the experiment, the Ecoflex-0010 from Smooth-on Inc. was used as a matrix material. Iron powders with the mean diameter of ~0.5 μm were bought from NANGONG XINDUN ALLOY WELDING MATERIAL SPRAYING CO., China and used as the magnetically soft component. Neodymium-iron-boron (NdFeB) alloy powders with the mean diameter of ~38 μm were bought from GUANZHOU XINNUODE TRANSMISSION PART CO., China and used as the magnetically hard component. The overall concentration of the filler in all samples was 50wt% with the ratio between the magnetically soft and hard components being 1:1. Hybrid MAEs were prepared by mixing neodymium-iron-boron (NdFeB) and iron micro-particles with silicone elastomer (Ecoflex 00-10) at the aforementioned weight fraction. The mixture was poured into a mold to obtain desired geometry and then cured at 80 °C for 10 min, in which the thickness (L) of the sample is 5 mm and the radius (R) is 20 mm. The cured material was uniformly magnetized along the thickness direction by applying an impulse magnetic field to produce $B^r$ in materials. The magnitude of $B^r$ can be tuned by changing the peak value of the pulsed magnetic field. The relationship between $B^r$ and the peak value of the pulsed magnetic field is presented in table 1. As shown in figure 5, the sample was loaded by using a customized solenoid that generated the AC magnetic field $h^e$ and deformations of these materials were detected by using a Laser Scanning Vibrometer (Polytec OFV-5000, Germany). The experiments were carried out on the vibration isolation workstation (M-VIS3048-IG2-125A, Newport, American). Throughout the whole work, the frequency of the magnetic field is fixed to 1 Hz.

### 3.1. The responses of the hybrid MAE thin film under different magnetic fields

Here, the sample is loaded by the magnetic field with different amplitudes (89 Oe, 300 Oe, and 600 Oe). The experimental results are presented in figures 6(a)–(c). As one may see in figure 6(a), the solid blue line represents the applied magnetic field $h^e$ which is harmonic in time. While the induced a change in thickness $\Delta L$, shown experimentally by the black marked line and theoretically by the red marked line, is anharmonic. Our theoretical formulation (26) indicates that the driving force comes from two parts: the Maxwell stress part $\sigma^{MW} = \mu_0 \left( \frac{\mu_h}{\mu_s} - 1 \right) h^e$ and the remanence stress part $\sigma^{RE} = h^e \lambda_1 B^r$. So, the frequency of the deformation

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**Figure 5.** (a) Photographs of experimental apparatus. One side of the sample is fixed on a non-magnetic fixed frame at the center of the spiral tube. In this work, the shear modulus is 40.1 kPa and the relative permeability is 1.31 for the sample. (b) The schematic of the deformation of the hybrid MAE. One side of the sample is fixed. The initial thickness of the hybrid MAE is $L$. Under an applied uniform magnetic field, $L$ becomes $L'$. **Table 1.** The residual magnetic flux density as a function of the peak value of the pulsed magnetic field.

| peak value of the pulsed magnetic field (Oe) | 0 | 3000 | 6000 | 9000 |
|-------------------------------------------|---|------|------|------|
| Residual magnetic flux density (mT)       | 0 | -0.362 | -2.194 | -11.486 |
induced by $\sigma_{MW}$ is 2 Hz, double of that of the magnetic field, and the frequency of the deformation induced by $\sigma_{RE}$ is 1 Hz. To identify the contribution from $\sigma_{MW}$ and $\sigma_{RE}$, in figures 6(a)–(i), we decompose the total displacement $\Delta L$ measured in the experiment into two parts: $\Delta L_{RE}$ (1 Hz) and $\Delta L_{MW}$ (2 Hz). As described in figures 6(a)–(ii), the ratio of the peak-peak value (PPV) of $\Delta L_{MW}$ to the PPV of $\Delta L_{RE}$ is equal to 7%, when the amplitude ($h_A$) of $h^t$ being 89Oe. When $h_A$ increases to 300Oe, the trough of the deformation flattened as present in figures 6(b)–(i) and the ratio of the PPV of $\Delta L_{MW}$ to the PPV of $\Delta L_{RE}$ value becomes 44% as shown in figures 6(b)–(ii). With $h_A$ further increased to 600Oe, as shown in figures 6(c)–(i), the trough, which was originally negative (figures 6(a)–(i)), became positive. The ratio of the PPV of $\Delta L_{MW}$ to the PPV of $\Delta L_{RE}$ is equal to 88% as presented in figures 6(c)–(ii).

As we know, the consequence of the magnetic Maxwell stress is always stretching (the value of deformation is always positive no matter which direction the magnetic field is pointing at (upward or downward)). Meanwhile the body will be either stretched or compressed under a remanence stress depending on the direction of the applied magnetic field. As presented in figure 7, when the applied magnetic field is small, the Maxwell stress is smaller than the remanence stress in magnitude. However, with the increase of the magnitude of the magnetic field, the Maxwell stress would eventually exceed the remanence stress. Therefore, with the increase of the external magnetic field, the 2 Hz deformation increase faster than the 1 Hz deformation. It is possible to

Figure 6. The deformation ($\Delta L$) of the hybrid MAE in a time variant uniform magnetic field ($h^t$) with different amplitudes ($h_A$). (a) $h_A = 89$Oe. (b) $h_A = 300$Oe. (c) $h_A = 600$Oe. The total displacement comes from the contribution of two parts: 1 Hz ($\Delta L_{RE}$) and 2 Hz ($\Delta L_{MW}$). The ratio of the peak-peak value (PPV) of $\Delta L_{MW}$ to PPV of $\Delta L_{RE}$ increases with the increase of $h_A$. 
manipulate the waveform of $\Delta L$ by adjusting $h_e$. Note that the deformation lags behind the external magnetic field due to the viscoelastic effect.

### 3.2. Hybrid MAE’s responses to different residual magnetic flux densities

From equation (26), the remanence stress $\sigma^{RE}$ is closely correlated to the residual magnetic flux density $B'$. Here, samples with different $B'$ are prepared and results are presented in figures 8(a)–(d). As presented in figure 8(a), when $B'$ equals to 0 mT, since there is only $\sigma^{MW}$ in the sample, the ratio of the PPV of $\Delta L^{RE}$ to the PPV of $\Delta L^{MW}$ equals to 0 and the frequency of the deformation is 2 Hz, which is twice the frequency of the external magnetic field. As shown in figure 8(b), when the absolute value of $B''$ is equal to 0.362 mT, $\sigma^{RE}$ appears. The ratio of the PPV of $\Delta L^{RE}$ to the PPV of $\Delta L^{MW}$ is equal to 21% and the waveforms begin to appear two positive peaks with different amplitudes within a period. With the increase of $B''$, the effect of $\sigma^{RE}$ is enhanced. When $B''$ is equal to 2.194 mT, the ratio of the PPV of $\Delta L^{RE}$ to the PPV of $\Delta L^{MW}$ is equal to 113% and the positive peaks with different amplitudes becomes more obvious as shown in figure 8(c). Finally, when $B''$ increases to 11.486 mT, the ratio of the PPV of $\Delta L^{RE}$ to the PPV of $\Delta L^{MW}$ is equal to 755% and the waveform becomes almost sinusoidal with a frequency of 1 Hz, as presented in figure 8(d). This conclusion also indicates that a good way, to manipulate the waveform of $\Delta L$, is to adjust $B'$.

### 4. Conclusions

In this work, we aim to add more degrees of freedom in designing magnetic active materials by rendering them with the ability of responding or deforming diversely in time. Particularly, a hybrid MAE consisting of rubber matrix embedded with both iron particles and neodymium-iron-boron (NdFeB) particles is proposed. In order to better predict its response in external magnetic fields, we have developed an energy formulation for the magnetomechanical behaviors of general magnetoactive bodies based on the principle of minimum free energy. The theoretical and experimental studies both indicate that the waveforms for the responses of hybrid MAE samples largely depend on the magnetization of the sample and the amplitude of the applied magnetic field. We find that the total deformation measured in the experiment can be decomposed into two parts: the mechanical responses with the same frequency as the external magnetic field ($\Delta L_1$) and the mechanical responses with a frequency twice of that for the external magnetic field ($\Delta L_2$). With the increase of the external magnetic field, $\Delta L_2$ increase faster than $\Delta L_1$. In addition, $\Delta L_1$ is tuned by variation of the magnetization of the sample. These conclusions may provide us effective ways of further diversifying the responses of magnetoactive materials. It is undoubtedly useful to have a clear exploration of the magneto-elastic coupling. Besides, since motions in the real world are always complex in time and space, this work provides a new idea of diversifying the temporal response of MAEs to the application of harmonic magnetic fields (harmonic in time). The hybrid MAEs may serve as a complement to the recently proposed 3D-printed hard MAEs and is expected to be used in the soft robots to imitate more complex real motions, without manipulating the magnetic stimulation too much.
Figure 8. The deformation ($\Delta L$) of hybrid MAEs with different residual magnetic flux densities ($B_r$) in a time variant uniform magnetic field. (a) $B_r = 0$ mT, (b) $B_r = -0.362$ mT, (c) $B_r = -2.194$ mT, (d) $B_r = -11.486$ mT.

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Appendix A. Variations of magnetization

Here, we will give a detail derivation of the Euler–Lagrange equations and boundary conditions. Firstly, we rewrite the free energy into the following form

\[
\mathcal{F} = \int_V \Phi(x_k, M_k) dV + \frac{\mu_0}{2} \int_{\mathbb{R}^3} JH_k X_{k,k} H_k x_{k,k} dV - \int_V H^4_k (\alpha_{1k} M_k + B^\prime_k x_{k,k}) dV
- \int_V J_{0f} u_r dV - \int_{\partial V} T_{\rho_0} u_r N_{k} dA - \int_V \rho_0 x_{k,k} dV.
\] (A.1)

A.1. Variations of magnetization in the magnetic energy

We first consider the variation of the magnetization. Physically, the variation of the magnetization \(M_k\) should result in the change of \(H^4_k\), \(H^\prime_k\), and \(B_k\) because of the Maxwell equation (4). We assume that the variation \(\delta M_k\) results in

\[
\hat{h}_k^\prime \to \hat{h}_k^\prime + \delta \hat{h}_k^\prime, \quad H^\prime_k \to H^\prime_k + \delta H^\prime_k, \quad B_k \to B_k + \delta B_k
\] (A.2)

It is noted that the perturbation field \((H^{\prime \prime \prime})\) far away from the solid must vanish, which can be described as

\[
H^{\prime \prime \prime} \to 0 \quad \text{as} \quad ||X|| \to \infty.
\] (A.3)

By equation (10), the variational calculations of \(\delta^{\text{mag}}\) can be written as

\[
\frac{d\delta^{\text{mag}}[M_k + \delta M_k, x_k]}{d\delta} \bigg|_{0} = \int_{\mathbb{R}^3} \mu_0 J(H^\prime_k X_{k,k} + \hat{h}_k^\prime) (\hat{H}^\prime_k X_{k,k} + \hat{h}_k^\prime) dV - \int_{\mathbb{R}^3} \mu_0 H^\prime_k \delta \hat{M}_k \alpha_{1k} dV
- \int_{\mathbb{R}^3} \mu_0 \hat{H}^\prime_k \delta \tilde{X}_{k,k} dV
+ \int_{\mathbb{R}^3} \mu_0 \hat{h}_k^\prime \delta \hat{h}_k^\prime dV
- \int_{\mathbb{R}^3} \mu_0 \hat{H}^\prime_k \delta \hat{M}_k dV + \int_{\mathbb{R}^3} \mu_0 \hat{h}_k^\prime \delta \hat{H}^\prime_k X_{k,k} dV.
\] (A.4)

Using the divergence theorem, we can rewrite the last term in the following form:

\[
\int_{\mathbb{R}^3} \mu_0 \hat{H}^\prime_k \delta \hat{H}^\prime_k X_{k,k} dV = \int_{\partial \mathbb{R}^3} \mu_0 \hat{h}_k^\prime \delta \hat{h}_k^\prime n_k dA + \int_{\mathbb{R}^3} \mu_0 \hat{h}_k^\prime \delta \hat{h}_k^\prime dV.
\] (A.5)

Since the magnetic field exists in the absence of the solid \((m = 0, \mathbf{b}^t = 0)\), we have \(\hat{h}_k^\prime = 0\). Then, equation (A.4) can be rewritten as

\[
\frac{d\delta^{\text{mag}}[M_k + \delta M_k, x_k]}{d\delta} \bigg|_{0} = \int_{\mathbb{R}^3} \mu_0 JH^\prime_k \hat{H}^\prime_k X_{k,k} dV - \int_{\mathbb{R}^3} \mu_0 H^\prime_k \alpha_{1k} \hat{M}_k dV.
\] (A.6)

Using equation (3), \(\hat{B}_L\) satisfies

\[
\hat{B}_L = \mu_0 J_{k,k} \hat{H}^\prime_k X_{k,k} + \mu_0 X_{k,k} \hat{M}_k \alpha_{1k}.
\] (A.7)

By the divergence theorem and the Maxwell equations, we have

\[
\int_{\partial \mathbb{R}^3} \xi^{\text{mag}} \hat{B} \cdot dA = \int_{\mathbb{R}^3} \xi^{\text{mag}} \hat{B} \cdot dV,
\] (A.8)

Substituting equation (A.7) into equation (A.8), we can obtain

\[
\int_{\partial \mathbb{R}^3} \xi^{\text{mag}} \hat{B} \cdot dA = -\int_{\partial \mathbb{R}^3} \mu_0 J_{k,k} H^\prime_k \hat{H}^\prime_k X_{k,k} dV - \int_{\mathbb{R}^3} \mu_0 H^\prime_k \alpha_{1k} dV.
\] (A.9)

Substituting equation (A.9) into equation (A.1), we have

\[
\frac{d\delta^{\text{mag}}[M_k + \delta M_k, x_k]}{d\delta} \bigg|_{0} = -\int_{\partial \mathbb{R}^3} \mu_0 J_{k,k} H^\prime_k \hat{M}_k dV - \int_{\mathbb{R}^3} \mu_0 H^\prime_k \tilde{M}_k dV - \int_{\partial \mathbb{R}^3} \xi^{\text{mag}} \hat{B} \cdot dA,
\] (A.10)

The value \(\tilde{M}_k\) is equal to 0 outside the deformed body. According to the Maxwell equations, the third term is equal to 0.
A.2. Variations of magnetization in the internal energy

For the internal energy, the variation of the magnetization can be written as

\[
\frac{d \mathcal{U}[M_e + \delta M_K, M_0]}{d \delta} \bigg|_{\delta = 0} = \int_V \left( \frac{\partial \Phi}{\partial M_K} \right) M_K dV.
\]

(A.11)

Since \(M_K\) is arbitrary, the first Euler–Lagrange equation can be obtained as

\[
\frac{\partial \Phi}{\partial M_K} = \mu (H^\text{eff}_L X_{L,k} \alpha_k + H^\text{eff}_K) = \mu_B h_k \alpha_k.
\]

(A.12)

Appendix B. Variations of deformation

In association with the variation (11), there will also be changes of \(F_{KK}, F_{KK}^{-1}, J\) and \(H^\text{eff}_K\)

\[
F_{KK} \rightarrow F_{KK} + \delta u_{k,K}, \quad J \rightarrow J + \delta X_{r,K} u_{r,K},
\]

\[
F_{KK}^{-1} \rightarrow F_{KK}^{-1} - \delta X_{r,K} X_{r,L} u_{r,L,K} H^\text{eff}_K \rightarrow H^\text{eff}_K + \delta \hat{H}^\text{eff}_K.
\]

(B.1)

We noted that the reference quantities \(M_K, H^\text{eff}_K,\) and \(B_k\) do not change with the variation \(u_k\). However, the current quantities \(m_k, h_k\) and \(b_k\) are affected by the variation of \(u_k\) because of equation (2).

B.1. Variations of deformation in internal energy

Using the divergence theorem, we first rewrite the variation of deformation in the internal energy into the following form

\[
\frac{d \mathcal{U}[x_r + \delta u_r, M_0]}{d \delta} \bigg|_{\delta = 0} = \int_V \left( \frac{\partial \Phi}{\partial F_{r,K}} \right) u_{r,K} dV = \int_V \left( \frac{\partial \Phi}{\partial F_{r,K}} \right) u_{r,K} N_R dA.
\]

(B.2)

B.2. Variations of deformation in magnetic energy

Using equation (B.1), the variational calculations of \(\varepsilon^{\text{mag}}\) can be written as

\[
\frac{d \varepsilon^{\text{mag}}[x_r + \delta x_r, M_0]}{d \delta} \bigg|_{\delta = 0} = \int_{\mathbb{R}^3} \mu_0 J X_{r,K} (H^\text{eff}_L X_{L,k} + H^\text{eff}_K X_{K,k} + H^\text{eff}_K) u_{r,K} dV
\]

\[
- \int_{\mathbb{R}^3} \mu_B H^\text{eff}_L X_{r,K} X_{r,L} (H^\text{eff}_K X_{K,k} + H^\text{eff}_K) u_{r,L} dV
\]

\[
+ \int_{\mathbb{R}^3} \mu_0 J (H^\text{eff}_L X_{L,k} + H^\text{eff}_K) \hat{B}_K X_{K,k} dV
\]

\[
- \int_{V_{\mathbb{R}^3}} H^\text{eff}_K B^\text{eff}_K u_{r,K} dV.
\]

(B.3)

To acquire the change of the magnetic field, we again assume (A.2) and have

\[
\hat{B}_L = \mu_0 J X_{r,K} X_{L,k} (H^\text{eff}_L X_{L,k} + H^\text{eff}_K) u_{r,K} - \mu_0 J X_{r,K} X_{L,r} (H^\text{eff}_L X_{K,k} + H^\text{eff}_K) u_{r,r} dV
\]

\[
- \mu_0 J X_{r,L} X_{K,k} X_{K,r} u_{r,r,K} + \mu_0 J X_{L,K} \hat{B}^\text{eff}_K X_{K,k} - \mu_0 J X_{r,K} X_{L,K} M_K \alpha_{KK} u_{r,K}.
\]

(B.4)

Using the Maxwell equations and the divergence theorem, we have

\[
\int_{\mathbb{R}^3} \xi^\text{int}_L \hat{B}_L dV = \int_{\mathbb{R}^3} \xi^\text{int}_L \hat{B}_L dV = - \int_{\mathbb{R}^3} H^\text{eff}_L \hat{B}_L dV,
\]

(B.5)

where \(h_k = H^\text{eff}_L X_{L,k} + H^\text{eff}_K\). Inserting equation (B.4) into the above equation, we obtain

\[
\int_{\mathbb{R}^3} \mu_0 J X_{r,K} X_{L,k} \hat{B}^\text{eff}_K X_{K,k} dV = - \int_{\mathbb{R}^3} \mu_0 J X_{r,K} (H^\text{eff}_L X_{L,k} + H^\text{eff}_K X_{K,k} + H^\text{eff}_K) u_{r,K} dV
\]

\[
+ \int_{\mathbb{R}^3} \mu_0 J X_{r,K} (H^\text{eff}_L X_{L,k} + H^\text{eff}_K) (H^\text{eff}_L X_{K,k} + H^\text{eff}_K) u_{r,K} dV
\]

\[
+ \int_{\mathbb{R}^3} \mu_0 J (H^\text{eff}_L X_{L,k} + H^\text{eff}_K) H^\text{eff}_K X_{K,k} u_{r,K} dV
\]

\[
+ \int_{\mathbb{R}^3} \mu_0 J (H^\text{eff}_L X_{L,k} + H^\text{eff}_K) X_{K,k} M_K \alpha_{KK} u_{r,K} = \int_{\mathbb{R}^3} \xi^\text{int}_L \hat{B}_L dA.
\]

(B.6)

Because of \(B_L = \mu_0 H^\text{eff}_L\) on the boundary of \(\mathbb{R}^3\), it is clear that \(\hat{B}_L = \mu_0 \hat{H}^\text{eff}_L = 0\). Using the Maxwell equations, we obtain the following form for the variation of the deformation in the magnetic energy:
\[
\frac{d \gamma_{\text{mech}}[x_r + \delta x_r, M_k]}{d \delta} \bigg|_{\delta = 0} = -\int_{\mathbb{R}^3} \frac{\mu_0}{2} J X_{R, r}(H_{L, \text{eff}} X_{L, k} + H_{K, \text{eff}} X_{K, k} + H_{H, \text{eff}} X_{H, k}) u_{r, k} dV
+ \int_{\mathbb{R}^3} \mu_0 J (H_{L, \text{eff}} X_{L, r} + H_{K, \text{eff}} X_{K, r} + H_{H, \text{eff}} X_{H, r}) X_{R, r, k} u_{r, r, k} dV
+ \int_{\mathbb{R}^3} \mu_0 (H_{L, \text{eff}} X_{L, r} + H_{K, \text{eff}} X_{K, r} + H_{H, \text{eff}} X_{H, r}) X_{R, r, k} \alpha_{k} \delta x_{r} dV - \int_{V} H_{R} B_{R} B_{R}^T u_{r, r} dV.
\] (B.7)

Let
\[
\Sigma_{\text{MW}}^{R} = -\frac{\mu_0}{2} J X_{R, r}(H_{L, \text{eff}} X_{L, k} + H_{K, \text{eff}} X_{K, k} + H_{H, \text{eff}} X_{H, k})
+ \mu_0 J (H_{L, \text{eff}} X_{L, r} + H_{K, \text{eff}} X_{K, r} + H_{H, \text{eff}} X_{H, r}) X_{R, r, k}
+ \mu_0 (H_{L, \text{eff}} X_{L, r} + H_{K, \text{eff}} X_{K, r} + H_{H, \text{eff}} X_{H, r}) X_{R, r, k} \alpha_{k},
\] (B.8)

and
\[
\Sigma_{\text{RE}}^{R} = -H_{R} B_{R}.
\] (B.9)

We refer to \(\Sigma_{\text{MW}}^{R}\) and \(\Sigma_{\text{RE}}^{R}\) as the first Piola-Maxwell stress and the first Piola-remanence stress respectively, which are related to the Cauchy stresses defined in the current configuration
\[
\sigma_{\text{MW}}^{R} = h_{1} h_{r} - \frac{\mu_0}{2} J h_{k} h_{k} \delta_{r} + \mu_0 h_{1} m_{r} = h_{1} h_{r} - \frac{\mu_0}{2} J h_{k} h_{k} \delta_{r},
\] (B.10)

and
\[
\sigma_{\text{RE}}^{R} = -h_{r} h_{r}.
\] (B.11)

by
\[
\Sigma_{\text{RE}}^{R} = J X_{R, r} \sigma_{r}.
\] (B.12)

Note that \(h_{r} = b - b^\prime\). Then, equation (B.3) can be rewritten as
\[
\frac{d \gamma_{\text{mech}}[x_r + \delta x_r, M_k]}{d \delta} \bigg|_{\delta = 0} = \int_{\partial V} [J(\sigma_{\text{MW}}^{R} + \sigma_{\text{RE}}^{R}) X_{R, k} N_{R} u_{r} dA - \int_{V} J(\sigma_{\text{MW}}^{R} + \sigma_{\text{RE}}^{R}) X_{R, k} N_{R} u_{r} dV],
\] (B.13)

where \([\partial V]\) is a jump across the boundary.

**B.3. Variations of deformation in the potential energy of mechanical loadings and the kinetic energy**

The variation of \(\gamma_{\text{mech}}\) is given by
\[
\frac{d \gamma_{\text{mech}}[x_r + \delta x_r, M_k]}{d \delta} \bigg|_{\delta = 0} = -\int_{V} J \rho_{0} \delta x_{r} u_{r} dV - \int_{\partial V} T_{R} N_{R} \rho_{0} \delta x_{r} u_{r} dA - \left(\int_{V} \frac{1}{2} \rho_{0} \dot{x}_{r} u_{r} dV\right)_{t} + \int_{V} \frac{1}{2} \rho_{0} \dot{x}_{r} u_{r} dV.
\] (B.14)

By equations (B.2), (B.13) and (B.14), we have the second Euler–Lagrange equations and the associated boundary conditions
\[
\left[\left[\frac{\partial}{\partial x_{r}} \frac{\partial}{\partial x_{r}} + \Sigma_{\text{MW}}^{R} + \Sigma_{\text{RE}}^{R}\right]_{R} + \rho_{0} \dot{x}_{r} = \rho_{0} \dot{x}_{r}\right]_{V} ~ \text{in} ~ V
\]
\[
\left[\Sigma_{\text{MW}}^{R} + \Sigma_{\text{RE}}^{R} \right]_{R} = 0 ~ \text{in} ~ \mathbb{R}^3 - V.
\] (B.15)

In summary, the magnetoelastic system should satisfy the following governing equations and boundary conditions
\[
\left[\left[\frac{\partial}{\partial x_{r}} \frac{\partial}{\partial x_{r}} + \Sigma_{\text{MW}}^{R} + \Sigma_{\text{RE}}^{R}\right]_{R} + \rho_{0} \dot{x}_{r} = \rho_{0} \dot{x}_{r}\right]_{V} ~ \text{in} ~ V
\]
\[
\left[\Sigma_{\text{MW}}^{R} + \Sigma_{\text{RE}}^{R} \right]_{R} = 0 ~ \text{in} ~ \mathbb{R}^3 - V.'
\] (B.16)

In summary, the magnetoelastic system should satisfy the following governing equations and boundary conditions
\[
\left[\left[\frac{\partial}{\partial x_{r}} \frac{\partial}{\partial x_{r}} + \Sigma_{\text{MW}}^{R} + \Sigma_{\text{RE}}^{R}\right]_{R} + \rho_{0} \dot{x}_{r} = \rho_{0} \dot{x}_{r}\right]_{V} ~ \text{in} ~ V
\]
\[
\left[\Sigma_{\text{MW}}^{R} + \Sigma_{\text{RE}}^{R} \right]_{R} = 0 ~ \text{in} ~ \mathbb{R}^3 - V.
\] (B.16)
Appendix C. The derivation of the governing equations for the 1-D magneto-mechanical system

There are three generalized coordinates (independent variables) $\lambda_1$, $\lambda_2$ and $M$ included in the expression of the free energy (22). The first variation condition can be written as:

$$\delta \mathcal{F} = \frac{\partial \mathcal{F}}{\partial M} \delta M + \frac{\partial \mathcal{F}}{\partial \lambda_1} \delta \lambda_1 + \frac{\partial \mathcal{F}}{\partial \lambda_2} \delta \lambda_2 = 0 \quad (C.1)$$

Substituting the free energy (22) into (C.1), we can obtain

$$\begin{align*}
\frac{\partial \mathcal{F}}{\partial M} &= \pi R^2 \int_0^L \left( \frac{\mu_0 M}{(\mu_0 - 1)} - \mu_0 h^2 + \mu_0 \frac{f}{2} (\xi_1 \lambda_1 M) \right) dX_1, \\
\frac{\partial \mathcal{F}}{\partial \lambda_1} &= \pi R^2 \int_0^L \left( \frac{\partial \mathcal{F}_{\text{elas}}}{{\partial \lambda_1}_{\partial M}} \lambda_1 = \frac{\mu_0 m^2}{2(\mu_0 - 1)} - \frac{\mu_0}{2} (h^2 h^2) + \lambda_2 h^2 + q \right) dX_1, \\
\frac{\partial \mathcal{F}}{\partial \lambda_2} &= \pi R^2 \int_0^L \left( \frac{\partial \mathcal{F}_{\text{elas}}}{{\partial \lambda_2}_{\partial M}} \lambda_2 = \frac{\mu_0 m^2}{2(\mu_0 - 1)} + \frac{\mu_0}{2} (h^2 h^2) + q \right) dX_1.
\end{align*} \quad (C.2)$$

Because $\delta M$, $\delta \lambda_1$ and $\delta \lambda_2$ are arbitrary, we can obtain the following governing equations:

$$\begin{align*}
\frac{\partial \mathcal{F}_{\text{elas}}}{{\partial \lambda_1}_{\partial M}} \lambda_1 - \frac{\mu_0 m^2}{2(\mu_0 - 1)} - \frac{\mu_0}{2} (h^2 h^2) + \lambda_2 h^2 + q &= 0, \\
\frac{\partial \mathcal{F}_{\text{elas}}}{{\partial \lambda_2}_{\partial M}} \lambda_2 - \frac{\mu_0 m^2}{2(\mu_0 - 1)} + \frac{\mu_0}{2} (h^2 h^2) + q &= 0.
\end{align*} \quad (C.3)$$

Eliminating $q$ in (C.3) and substituting (24) and (20) into (C.3), we have

$$\begin{align*}
m &= (\mu_0 - 1) h \\
\lambda_1 \left( \lambda_1 - \frac{1}{M} \right) &= \mu_0 \left( \frac{\mu_0 - 1}{\mu_0} \right) h c \lambda_1 B_i^2 = 0.
\end{align*} \quad (C.4)$$

which are the governing equations for the 1-D magneto-mechanical system.

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References

Alameh Z, Qian D, Liu L and Sharma P 2015 Using electrets to design concurrent magnetoelectricity and piezoelectricity in soft materials J. Mater. Res. 30 93–100
Alameh Z, Yang S, Qian D and Sharma P 2018 Emergent magnetoelectricity in soft materials, insta- bility, and wireless energy harvesting Soft Matter 14 5856–68
Ball J M 1976 Convexity conditions and existence theorems in nonlinear elasticity Arch. Ration. Mech. Anal. 63 337–403
Coey J M D 2011 Hard magnetic materials: a perspective IEEE Trans. Magn. 47 6671–81
Coquille E and Bossis G 2005 Magnetostriiction and piezoresistivity in elastomers filled with magnetic particles Journal of Advanced Science 17 132–8
Deng Q, Ahmadpoor W E, Brownell P and Sharma P 2019 The collision of flexoelectricity and hopf bifurcation in the hearing mechanism J. Mech. Phys. Solids 130 245–61
Deng Q, Liu L and Sharma P 2014 Flexoelectricity in soft materials and biological membranes J. Mech. Phys. Solids 62 209–27
Feng J, Xuexi S, Li D and Gong X 2017 Magnetoactive elastomer/pvdf composite film based magnetically controllable actuator with real-time deformation feedback property Composites Part A 103 25–34
Feng J, Xuexi S, Li D and Gong X 2018 Magnetic-field-induced deformation analysis of magnetoactive elastomer film by means of bics, kdv, and ferm Ind. Eng. Chem. Res. 57 1246–54
Filipcesei G, Getzinski I, Sirlagi A and Zrimyi M 2007 Magnetic-field-responsive smart polymer composites Molecular Imprinting 206 137–89
Guan X, Dong X and Jinping O U 2008 Magnetostriuctive effect of magnetorheological elastomer J. Magn. Magn. Mater. 320 158–63
Hu W, Lem G Z, Mastrangelo M and Sitti M 2018 Small scale soft-bodied robot with multimodal locomotion Nature 554 81–5
Ivaneyko D, Toshchevikov V, Saphiannikova M and Heinrich G 2012 Effects of particle distribution on mechanical properties of magneto-sensitive elastomers in a homogeneous magnetic field Condens. Matter Phys. 15 33601
Kankanal S V and Triantafyllidis N 2004 On finitely strained magnetorheological elastomers J. Mech. Phys. Solids 52 2869–908
Kim F and Seok J 2014 A multi–stable energy harvester: dynamic modeling and bifurcation analysis J. Sound Vib. 333 5525–47
Kim Y, Yuk H, Zhao R, Chester S A and Zhao X 2018 Printing ferromagnetic domains for untethered fast-transforming soft materials Nature 558 274–9
Krichen S, Liu L, Sharma P, Krichen S, Liu L, Sharma P, Krichen S, Liu L and Sharma P 2017 Biological cell as a soft magnetoelectric material: Elucidating the physical mechanisms underpinning the detection of magnetic fields by animals Phys. Rev. E 96 042404
Liping L and Pradeep S 2013 Giant and universal magnetoelectric coupling in soft materials and concomitant ramifications for materials science and biology Phys. Rev. E 88 040601
Liu J, Gillen J, Mishra S, Evans B and Tracy J 2019 Photothermally and magnetically controlled reconfiguration of polymer composites for soft robotics Science Advances 5 eaaw2897
Liu L 2014 An energy formulation of continuum magneto-electro-elasticity with applications J. Mech. Phys. Solids 63 451–80
Lum G Z, Ye Z, Dong X, Marvi H, Erin O, Hu W and Sitti M 2016 Shape-programmable magnetic soft matter Proc. of the National Academy of Sciences of the United States of America 113 E6007–15
Opie S and Yim W 2009 Design and control of a real-time variable stiffness vibration isolator IEEE/ASME International Conf. on Advanced Intelligent Mechatronics 380–5
Qi S, Guo H, Chen J, Fu J, Hu C, Yu M and Wang Z 2018 Magnetorheological elastomers enabled high sensitive self-powered tribo-sensor for magnetic field detecting Nanoscale 10 4745–52
Ren Z, Hu W, Dong X and Sitti M 2019 Multi-functional soft-bodied jellyfish-like swimming Nat. Commun. 10 2703
Rigbi Z and Jilken L 1983 The response of an elastomer filled with soft ferrite to mechanical and magnetic influences J. Magn. Magn. Mater. 37 267–76
Treloar L R G 1949 Stresses and birefringence in rubber subjected to general homogeneous strain Proc. of the Physical Society 60 135–44
Yang J, Sun S S, Du H, Li W H, Alici G and Deng H X 2014 A novel magnetorheological elastomer isolator with negative changing stiffness for vibration reduction Smart Mater. Struct. 23 105023
Yoonho K, German P, Shengduo L and Xuanhe Z 2019 Ferromagnetic soft continuum robots Science Robotics 4 eaaax7329
Zhao R, Kim Y, Chester S A, Sharma P and Zhao X 2018 Mechanics of hard-magnetic soft materials J. Mech. Phys. Solids 124 244–63
Zhao X, Kim J, Cezar C A, Huebsch N, Lee K, Bouhadir K, Mooney D J and Klibanov A M 2011 Active scaffolds for on-demand drug and cell delivery Proc. of the National Academy of Sciences of the United States of America 108 67–72
Zhou S and Zuo L 2018 Nonlinear dynamic analysis of asymmetric tristable energy harvesters for enhanced energy harvesting Commun. Nonlinear Sci. Numer. Simul. 61 271–84
Zhou Z, Qin W and Zhu P 2017 Harvesting acoustic energy by coherence resonance of a bi-stable piezoelectric harvester Energy 126 527–34
Zrinyi M, Barsi L and Buki A 1996 Deformation of ferrogels induced by nonuniform magnetic fields J. Chem. Phys. 104 8750–6