

Supplementary Information: Encoding reprogrammable properties into magneto-mechanical materials via topology optimization

Zhi Zhao¹ and Xiaojia Shelly Zhang^{1,2,*}

¹Department of Civil and Environmental Engineering, University of Illinois
Urbana-Champaign, Urbana, IL 61801

²Department of Mechanical Science and Engineering, University of Illinois
Urbana-Champaign, Urbana, IL 61801

*Email: zhangxs@illinois.edu

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Supplementary Methods

Constitutive model for hard-magnetic soft materials

Consider a deformable solid that occupies a domain Ω in its undeformed configuration with $\mathbf{X}$ denoting the position vector. On the boundary, the solid is subjected to an applied displacement field $\tilde{\mathbf{u}}$ on Γ_u (Dirichlet boundary condition) and an applied traction on Γ_t (Neumann boundary condition), where $\Gamma_u \cup \Gamma_t = \partial\Omega$ and $\Gamma_u \cap \Gamma_t = \emptyset$. The solid undergoes a deformation map χ , which deforms a material point $\mathbf{X}$ to the position $\mathbf{x} = \chi(\mathbf{X})$ in the deformed configuration Ω_t . The corresponding deformation gradient tensor $\mathbf{F}$ is defined by $\mathbf{F} = \nabla\chi$, where ∇ stands for the gradient operator with respect to the undeformed configuration. This study adopts the constitutive model for ideal hard-magnetic soft materials developed by [1]. This model has been validated to possess prediction in great agreement with experimental results [1]. We denote $\mathbf{B}_r$ and $\mathbf{B}_a$ as residual magnetic flux density distribution for hard-magnetic soft material (i.e., magnetically saturated pre-magnetization) and externally applied magnetic flux density in the undeformed configuration.

To construct the constitutive model, several rational assumptions are introduced to simplify what would otherwise be a complex and coupled nonlinear magneto-mechanical theory [1]. These assumptions include: (1) The magnetic material is magnetically saturated and retains a residual magnetic flux density $\mathbf{B}_r$ independent of the external magnetic field $\mathbf{H}$, whose strength is much lower than the magnetic coercivity $\mathbf{H}_c$. (2) The magnetic permeability (μ) of pre-magnetized hard-magnetic soft material is assumed to be the vacuum (or air) permeability, i.e., $\mu = \mu_0 = 1.257 \times 10^{-6}$ H/m. (3) In this study, the applied magnetic flux density $\mathbf{B}_a$ is assumed to be a uniform vector.

According to the theory, the constitutive relation of the ideal hard-magnetic soft materials is described by the following nominal Helmholtz free energy function (per unit volume in the undeformed configuration) [1]:

$$W(\mathbf{F}) = W_E(\mathbf{F}) + W_M(\mathbf{F}) = W_E(\mathbf{F}) - \frac{1}{\mu_0} \mathbf{F} \mathbf{B}_r \cdot \mathbf{B}_a, \quad (1)$$

where $W_E(\mathbf{F})$ is hyperelastic stored-energy of compressible soft matrix materials (e.g., silicone rubber); $W_M(\mathbf{F}, \mathbf{B}_r, \mathbf{B}_a) = -\frac{1}{\mu_0} \mathbf{F} \mathbf{B}_r \cdot \mathbf{B}_a$ is magnetic free energy. By taking derivative of Eq. (1) with respect to $\mathbf{F}$,

we can obtain the first Piola-Kirchoff stress $\mathbf{P}$ [1, 2]:

$$\mathbf{P}(\mathbf{F}) = \frac{\partial W_E(\mathbf{F})}{\partial \mathbf{F}} - \frac{1}{\mu_0} \mathbf{B}_a \otimes \mathbf{B}_r. \quad (2)$$

The elastic part of the free energy function $W_E(\mathbf{F})$ can be any commonly-used compressible hyperelastic models. In this study, we adopt the compressible Ogden model [3, 4] for $W_E(\mathbf{F})$, which is of the form:

$$W_E(\mathbf{F}) = \sum_{a=1}^{N_a} \frac{\mu_a}{\alpha_a} (\lambda_1^{\alpha_a} + \lambda_2^{\alpha_a} + \lambda_3^{\alpha_a} - 3) + \sum_{a=1}^{N_a} \frac{\mu_a}{\alpha_a \beta_a} (J^{-\alpha_a \beta_a} - 1), \quad (3)$$

where $\lambda_1, \lambda_2, \lambda_3$ are the principal stretches associated with the deformation gradient $\mathbf{F}$; $J \doteq \det(\mathbf{F})$; and μ_a , α_a , N_a , and β_a are material parameters such that the initial shear modulus $G = \frac{1}{2} \sum_{a=1}^{N_a} \mu_a \alpha_a$ and initial bulk modulus $\kappa = \sum_{a=1}^{N_a} \mu_a \alpha_a (\frac{1}{3} + \beta_a)$. In this work, the one-term Ogden model, i.e., $N_a = 1$, is adopted.

In the undeformed configuration, the equilibrium of the solid is governed by the following partial differential equations (body force is neglected):

$$\begin{aligned} \nabla \cdot \mathbf{P} &= \mathbf{0} \quad \text{in } \Omega, \\ \mathbf{u} &= \tilde{\mathbf{u}} \quad \text{on } \Gamma_u, \\ \mathbf{P}\mathbf{N} &= \tilde{\mathbf{t}} \quad \text{on } \Gamma_t, \end{aligned} \quad (4)$$

where $\nabla \cdot$ stands for the divergence operator in the undeformed configuration, $\mathbf{N}$ is the outward unit vector normal to the undeformed boundary of the solid, and $\tilde{\mathbf{t}}$ is the applied traction. We adopt the standard displacement-based formulation. Given a mesh (Ω_e refers to the e th element) of the domain discretized by quadrilateral elements, the total potential energy, $\Pi(\mathbf{u})$, and its stationary condition (equilibrium), $\mathbf{r}(\mathbf{u})$, are given by

$$\Pi(\mathbf{u}) = \sum_e \int_{\Omega_e} W(\mathbf{u}_e) d\mathbf{x} - (\mathbf{f}_{\text{ext}})^T \mathbf{u}, \quad (5)$$

and

$$\mathbf{r}(\mathbf{u}) = \frac{\partial \Pi}{\partial \mathbf{u}}(\mathbf{u}) = \mathbf{f}_{\text{int}}(\mathbf{u}) - \mathbf{f}_{\text{ext}}(\mathbf{u}) = \mathbf{0}, \quad (6)$$

respectively. In the formulas, $\mathbf{u}$ is the displacement vector; and $\mathbf{r}$, $\mathbf{f}_{\text{int}}$ and $\mathbf{f}_{\text{ext}}$ are the global residual, internal force, and external force vectors, respectively. The above nonlinear system of equations is solved iteratively using the Newton-Raphson procedure [5] with an inexact line search method [6, 7]. In this study, we assume the plane stress condition in 2D, making the displacement-based finite elements free of volumetric locking even with soft materials possessing near-incompressible behaviors [8].

Parameterization of matrix distribution (i.e., topology)

The distribution of the matrix characterizes the spatial occupancy of material. Here, a density-based approach [9] is adopted. The matrix distribution is associated with the density variable ρ with ρ_e for the e th element. We apply the Heaviside projection operator [10] (with 1/2 being its threshold) to the density variable, obtaining the physical density variables $\bar{\rho}$ with $\bar{\rho}_e$ given by

$$\bar{\rho}_e = \frac{\tanh(\frac{\beta_\rho}{2}) + \tanh(\beta_\rho(\tilde{\rho}_e - \frac{1}{2}))}{2 \tanh(\frac{\beta_\rho}{2})}, \quad (7)$$

with β_ρ being the parameter controlling the discreteness of the projection, and $\tilde{\rho}_e$ being the intermediate design variable regularized via the density filter [11, 12] as follows:

$$\tilde{\rho}_e = \frac{\sum_{i \in \mathcal{I}_e(R_\rho)} w_\rho^{(i,e)} v_i \rho_i}{\sum_{i \in \mathcal{I}_e(R_\rho)} w_\rho^{(i,e)} v_i}, \quad (8)$$

where $\mathcal{I}_e(R_\rho)$ is the e th element set within a prescribed region defined by a circle with a radius of R_ρ at the centroid of e th element; and v_i is the i th element volume. The weighting factor $w_\rho^{(i,e)}(R_\rho, r_\rho)$ depends on

the distance between the centroids of i th and e th elements (denoted as $\mathbf{X}_i$ and $\mathbf{X}_e$, respectively), namely, $w_{\rho}^{(i,e)} = 1 - (\|\mathbf{X}_i - \mathbf{X}_e\|/R_{\rho})^{r_{\rho}}$, with r_{ρ} being the power of the filter. The physical design variable $\bar{\rho}_e$ serves as an indicator of whether a given location in space is solid or void: $\bar{\rho}_e = 1$ represents solid and $\bar{\rho}_e = 0$ represents void.

Parameterization of magnetization distribution

The residual magnetic flux density at each location of the design is selected from a set of N_m pre-selected candidate residual magnetic flux densities, $\mathbf{B}_r^{(1)}, \dots, \mathbf{B}_r^{(N_m)}$, each pointing at one direction. Formally, we define the residual magnetic flux density in element e as

$$\mathbf{B}_{r,e} = \sum_{j=1}^{N_m} \left(\bar{m}_e^{(j)} \right)^{p_m} \mathbf{B}_r^{(j)}. \quad (9)$$

In the above interpolation, $\bar{m}_e^{(j)}$ is the physical magnetization variable which serves as an indicator of the magnetization of element e : $\bar{m}_e^{(j)} = 1$ means the j th candidate residual magnetic flux density $\mathbf{B}_r^{(j)}$ is selected, and $\bar{m}_e^{(j)} = 0$ means the j th candidate residual magnetic flux density $\mathbf{B}_r^{(j)}$ is not selected. A Solid Isotropic Material with Penalization (SIMP)-type [13, 14] penalization power p_m is introduced to penalize the mixture of candidate magnetizations and to promote the convergence of the physical magnetization variables $\bar{m}_e^{(j)}$ to either 1 or 0.

In this work, we aim to promote discrete magnetization distribution and allow non-magnetized regions to appear in the design. Therefore, the physical magnetization variables need to satisfy the following two constraints: 1) $\sum_{j=1}^{N_m} \bar{m}_e^{(j)} \leq 1$ and 2) $\bar{m}_e^{(j)} \geq 0, \forall j$. To this end, we adopt the Hypercube-to-Simplex Projection (HSP) approach [15, 16] to define $\bar{m}_e^{(j)}$. The main benefit of using HSP is both above-mentioned constraints 1) and 2) are automatically satisfied by construction. As a result, no extra constraint of $\bar{m}_e^{(j)}$ is needed in the optimization formulation. The HSP approach defines the physical magnetization variables $\bar{m}_e^{(j)}$ as

$$\bar{m}_e^{(j)} = \sum_{i=1}^{2^{N_m}} s_i^{(j)} \left((-1)^{(N_m + \sum_{j=1}^{N_m} c_i^{(j)})} \prod_{k=1}^{N_m} \left(\bar{\xi}_e^{(k)} + c_i^{(k)} - 1 \right) \right), \quad (10)$$

where $\bar{\xi}_e^{(j)}$ is the magnetization variable subject to filtering and projection through the same expressions in Eqs. (7) and (8). The parameter $c_i^{(j)} = \{0, 1\}$ is the i th vertex of a N_m -dimensional unit hypercube for the j th candidate remnant magnetization vector, and $s_i^{(j)}$ is the mapped vertex of a N_m -dimensional standard simplex domain:

$$s_i^{(j)} = \begin{cases} \frac{c_i^{(j)}}{\sum_{j=1}^{N_m} c_i^{(j)}} & \text{if } \sum_{j=1}^{N_m} c_i^{(j)} \geq 1, \\ 0 & \text{otherwise.} \end{cases} \quad (11)$$

Interpolation of the Helmholtz free energy function

To describe the nonlinear mechanical behavior of the hard-magnetic soft material designs, we introduce the following interpolation of the Helmholtz free energy function from the physical variables $\bar{\rho}$ and $\bar{\mathbf{m}}^{(j)}$, $j = 1, \dots, N_m$. The interpolated free energy W_e of element e is given by

$$W_e(\bar{\rho}_e, \bar{m}_e^{(1)}, \dots, \bar{m}_e^{(N_m)}, \mathbf{u}_e^L) = [\epsilon + (1 - \epsilon)(\bar{\rho}_e)^{p_{\rho}}] W_E(\bar{\rho}_e, \mathbf{u}_e^L) + (\bar{\rho}_e)^{p_{\rho}} W_M(\mathbf{u}_e^L, \mathbf{B}_{r,e}(\bar{m}_e^{(1)}, \dots, \bar{m}_e^{(N_m)})), \quad (12)$$

where $\mathbf{u}_e^L$ is the displacement vector in element e under mechanical load ($L = (F)$) or magneto-mechanical load ($L = (F, B_a)$); and $\epsilon = 10^{-5}$ is a small value to avoid singular stiffness. In the above interpolation formula, the SIMP approach [9, 14] is used to penalize both elastic-stored energy and magnetic free energy to drive the variable $\bar{\rho}_e$ toward either 0 or 1 and generate a nearly discrete design. The penalization parameters associated with both energies are taken to be the same. In addition, based on our numerical experience, excessive deformations of low-stiffness regions can lead to numerical instabilities in FE analysis

during optimization. Thus, the energy interpolation scheme [17] depending on design variable $\bar{\rho}_e$ is applied to the stored-energy W_E to address the numerical instabilities of low stiffness regions (defined to be regions with $(\bar{\rho}_e)^{p_\rho} \leq 0.01$). We also apply the same concept to the magnetic free energy W_M so that the magnetic actuation in low stiffness regions is negligible. The readers are referred to [17, 18] for details.

Optimization formulation

With the introduced design space parameterization and free-energy interpolation schemes, we now present the topology optimization formulation to generate multi-functional metamaterial and structural designs under mechanical and magneto-mechanical loads, respectively. The mesh Ω_h is composed of N_e elements and N_n nodes. The goal of the proposed topology optimization is to minimize a certain performance objective function $J(\cdot)$ with the constraints and the nested equilibrium satisfied.

Formally, we formulate the topology optimization problem as:

$$\begin{aligned}
& \min_{\boldsymbol{\rho}, \boldsymbol{\xi}^{(1)}, \dots, \boldsymbol{\xi}^{(N_m)}} J\left(\mathbf{u}^{(F)}, \mathbf{u}^{(F, B_a)}\right), \\
& \text{s.t. :} \quad \frac{\mathbf{v}^T \bar{\boldsymbol{\rho}}}{|\Omega_h|} \leq v_{\max}, \\
& \quad F(\mathbf{u}^L) \geq F_{\min}^L, \quad L \in [(F), (F, B_a)], \\
& \quad \left\{ \sum_{e=1}^{N_e} \left[\frac{w_\sigma(\bar{\rho}_e)}{v_e} \int_{\Omega_{h,e}} \sigma_{VM}(\sigma_E(\mathbf{u}^L)) d\mathbf{X} \right]^{p_n} \right\}^{1/p_n} \leq \sigma_{\max}^L, \quad L \in [(F), (F, B_a)], \\
& \quad \mathbf{r}(\bar{\boldsymbol{\rho}}, \bar{\mathbf{m}}^{(1)}, \dots, \bar{\mathbf{m}}^{(N_m)}, \mathbf{u}^L) = \mathbf{0}, \quad L \in [(F), (F, B_a)], \\
& \quad \mathbf{0} \leq \boldsymbol{\rho} \leq \mathbf{1}, \\
& \quad \mathbf{0} \leq \boldsymbol{\xi}^{(j)} \leq \mathbf{1}, \quad j = 1, \dots, N_m,
\end{aligned} \tag{13}$$

where $\mathbf{u}^L$ is the converged displacement vector under mechanical load ($L = (F)$) or magneto-mechanical load ($L = (F, B_a)$); $\mathbf{v}$ is the element volume vector with v_e being the volume of element e ; $v_{\max}$ is the assigned maximum volume for matrix materials. We note that the force constraint [19, 20] is used for designing magneto-active structures with adaptable snap buckling, with F being the summation of force at target degree of freedoms (DOFs) and $F_{\min}^L$ being the prescribed minimum force. In addition, the above optimization formulation incorporates stress constraints in which the von-Mise stress [21] $\sigma_{VM}(\cdot)$ associated with the Cauchy stress tensor of elastic stored-energy $\sigma_E = 1/J(\partial W_E(\mathbf{F})/\partial \mathbf{F})\mathbf{F}^T$ (i.e., mechanical Cauchy stress) is constrained to be below a prescribed upper bound. The stress constraint is imposed individually on the deformation state $\mathbf{u}^L$ with the upper bound $\sigma_{\max}^L$. To prevent the singularity issue in the stress-constrained topology optimization [22, 21, 23], we adopt the relaxation approach and define $w_\sigma(\bar{\rho}_e) \doteq \epsilon + (1 - \epsilon)\bar{\rho}_e^{q_\rho}$ with $q_\rho < 1$. The stress constraint is handled by the p-norm approach [21] with the power of p_n . We emphasize that the stress constraints, as a numerical technique, are effective to prevent thin members and hinge-like connections from appearing in the optimized design as well as to restrict the level of local deformations [18, 8].

The proposed formulation (13) is solved by a gradient-based update algorithm. In this work, we adopt the method of moving asymptotes (MMA) [24]. The sensitivities information of objective and constraint functions with respect to the design variables are derived by the adjoint method [9].

Fabrication approach

The overall fabrication procedure is presented in Fig. 5(a) in the main text. For the hard-magnetic soft material, Dow Sylgard 184 Kit (Polydimethylsiloxane, PDMS) (Fisher Scientific) and NdFeB particles (the average particle size of 25 μm , Magnequench) are used. The base-agent weight ratio of PDMS is 25 : 1, and the NdFeB particles of 15% volume ratio are added. The base, agent, and NdFeB particles are mixed together in a tray for 20 minutes, followed by degassing the mixture under -0.8 atmosphere for 1 hour to avoid trapped air bubbles. We then inject the degassed mixture into polyvinyl alcohol (PVA) molds (possessing the geometries of different parts of the specimen) printed by a Fused Deposition Modeling (FDM) printer

(Prusa Research). The mixture with the mold is cured at 80 °C for 2 hours in an oven before demolding. Notably, the PVA mold is soft and fragile at high temperatures, significantly easing the demolding process. According to the optimized magnetization directions, we individually magnetize different parts under a 2 T magnetizing field generated by an impulse magnetizer (IM-10-30, ASC Scientific). The complete sample is assembled by injecting the uncured mixture onto the interfaces of different magnetized parts and curing at 80 °C for 2 hours in an oven.

Material characterization

For the mechanical property, the parameters in the constitutive model are characterized by measuring the stress-strain relations of dogbone specimens of the fabricated hard-magnetic soft material according to the standard [25]. The fitted parameters are: $\alpha = 2.543$ MPa and $\mu = 0.1183$ MPa. The Poisson's ratio is assumed to be $\nu = 0.49$. The residual magnetic flux density is measured to be 100 mT using a vibrating-sample magnetometer (Quantum Design MPMS3).

Experimental setup

The loading frame to perform mechanical compression is printed by an FDM printer using Polylactic acid (PLA). The specimen is glued onto the interior loading blocks of the frame and suspended above the ground to avoid in-plane frictions (the self-gravity is negligible compared to the applied loading). The frame is glued onto the ground to eliminate rigid body motion. The magnetic field (with a magnitude of 25 mT) is generated through a pair of electromagnets. The magnetic field is measured by a Gauss meter (PCE-MFM 4000). A camera (SONY α 7R) is placed on top of the loading frame to film the loading process.

Supplementary Note 1

Summary of discovered magneto-mechanical metamaterials and structures

We adopt the proposed optimization-guided design approach to discover three promising multi-functional magneto-mechanical metamaterials and structures. As shown in Supplementary Table 1, each of them can achieve two different functionalities under mechanical and magneto-mechanical loads, respectively.

Supplementary Table 1: Brief descriptions of discovered multi-functional magneto-mechanical metamaterials and structures with tunable properties.

Name/Feature	Property 1 under purely mechanical load	Property 2 under magneto-mechanical load
Magnetic actuators with sufficiently high stiffness	Sufficient structural stiffness	Maximized magnetic actuation
Magneto-active metastructures with adaptable snap buckling	Delayed buckling	Ensured snap buckling
Magneto-active metamaterials with distinct deformation modes	Mimicking material behavior of positive Poisson's ratio	Mimicking material behavior of negative Poisson's ratio

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