Shape Memory and Recovery Mechanism in Hard Magnetic Soft Materials (HMSMs)

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Supplemental Information

A1 Magnetic and Mechanical Properties of HMSMs

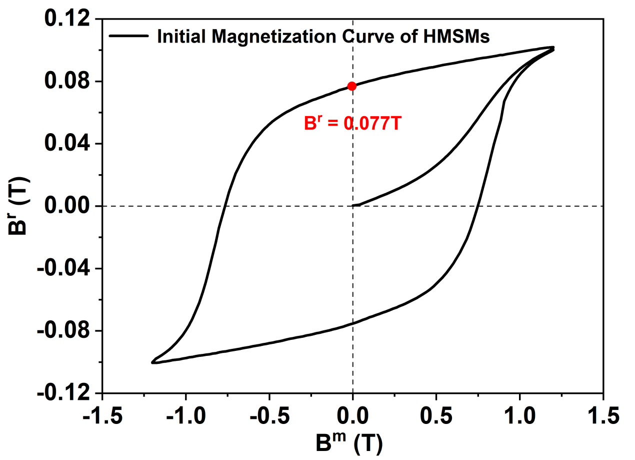

Figure S1: Initial magnetization curve of HMSMs

The magnetic hysteresis loop ($\mathbf{B}^{\mathrm{m}} - \mathbf{M}^{\mathrm{r}}$) was plotted using by a Vibrating Sample Magnetometer (MPMS-squid VSM-094). The intercept on the $\mathbf{M}$ axis represents the residual magnetization density $\mathbf{M}^{\mathrm{r}}$. The value of $\mathbf{B}^{\mathrm{r}}$ is calculated as $\mathbf{B}^{\mathrm{r}} = \frac{10^4}{\pi}\mathbf{M}^{\mathrm{r}}\rho$, where ρ presents the mass density of the material. Experimental results indicate that the residual magnetization density of HMSMs is 0.077 T.

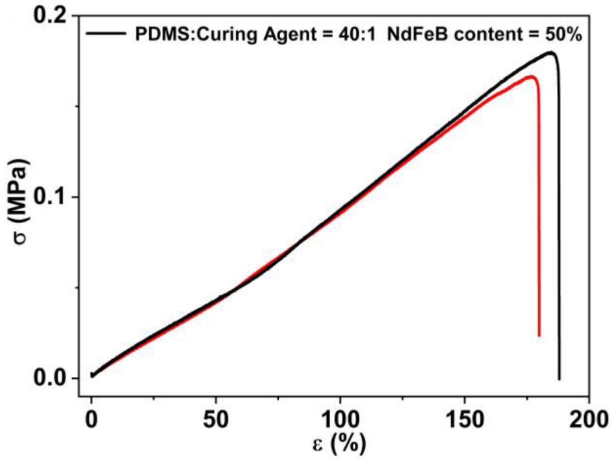

Figure S2: Stress-strain curve of HMSMs (PDMS : curing agent = 40 : 1)

To determine Young's modulus, a mechanical testing machine (Electroforce 3230, TA, US) was used to stretch a dumbbell-shaped sample (length: 50 mm, thickness: 2 mm, width: 4 mm) at a speed of 0.05 mm/s until until the sample broke. Young's modulus was calculated as the slope of the tensile stress-strain curve. Experimental results show that the Young's modulus of HMSMs with a PDMS polymer matrix (PDMS : curing agent = 40 : 1) is 0.1 MPa.

A2 Preparation and experimental testing of HMSMs

Polydimethylsiloxane (PDMS) were used as the soft matrices, with NdFeB micro-particles (average diameter 38 μm) added as the magnetic component for HMSMs. The filler content was kept at 50 wt% in all samples. The mixture was then molded into the required shape and cured at 80C for 3 hours.

An external magnetic field $\mathbf{B}^{\mathrm{m}}$ was produced using a Multi-field Coupling Measurement System for Force-Electric-Magnetic-Thermal Loading S&T. Displacement δ under the actuating magnetic field $\mathbf{B}^{\mathrm{a}}$ was measured with a Laser Displacement Sensor (LK-G500). The actuating magnetic field was generated by a Three-Dimensional Helmholtz Coil (PS-3HM400), capable of producing a maximum magnetic field of 180 Oe (0.018 T).

A3 Remanent magnetic flux density vector and magnetic force of HMSMs

The remanent magnetic flux density vector of the cantilever beam

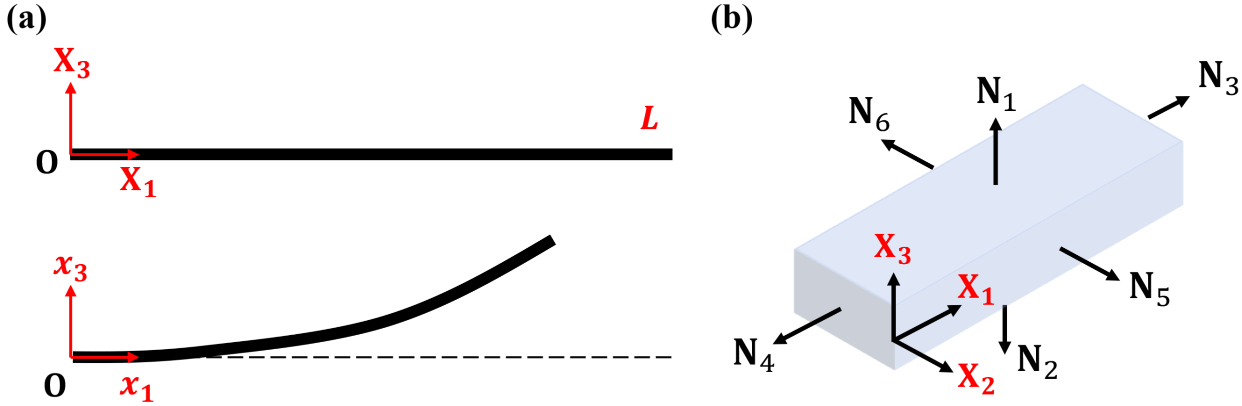

Figure S3: (a) A bending cantilever beam (b) A 3D model of HMSMs

HMSMs, with dimensions of length L, width b and thickness t, transitioned from a horizontal configuration to a cantilever beam in bending. The initial state is defined as:

$$\begin{cases} X_1 = X_1, \quad X_1 \in [0, L] \\ X_3 = 0 \end{cases} \tag{S1}$$

The shape function of the bending beam's centerline is as follows:

$$\begin{cases} \int_0^{x_1} \sqrt{1 + \frac{1}{36}(\frac{P}{EI})^2(6lx_1 - 3x_1^2)} = X_1 \\ x_3 = -\frac{Px_1^2}{6EI}(3l - x_1) \end{cases} \tag{S2}$$

Eq. (S2)$_1$ is a nonlinear integral equation that we approximate with polynomials to establish the relationship between x_1 and X_1, expressed as, $x_1 = f(X_1)$. This allows us to compute the deformation gradient $\mathbf{F}^{\mathrm{E}}$,

$$\mathbf{F}^{\mathrm{E}} = \begin{bmatrix} f' + X_3 \cdot \frac{f'}{|g|}\xi & 0 & \frac{g}{\sqrt{f'^2+g^2}} \\ 0 & 1 & 0 \\ -g - X_3 \cdot \frac{g}{|g|}\xi & 0 & \frac{f'}{\sqrt{f'^2+g^2}} \end{bmatrix} \tag{S3}$$

where $g = -\frac{P}{2EI}(2lf \cdot f' - f^2 f')$ and $\xi = \frac{f''(f'^2+g^2) - f'(2f'f''+2gg')}{f'^2+g^2}$. Then we can calculate the remanent magnetic flux density vector $\mathbf{B}_0^{\mathrm{r}}$,

$$\mathbf{B}_0^{\mathrm{r}} = \begin{bmatrix} g + X_3 \cdot \frac{g}{|g|}\xi \\ 0 \\ f' + X_3 \cdot \frac{f'}{|g|}\xi \end{bmatrix} B_1^{\mathrm{r}} \tag{S4}$$

where $\mathbf{B}_1^{\mathrm{r}} = [0, 0, 1]^{\mathrm{T}} B_1^{\mathrm{r}}$.

The magnetic force of the cantilever beam

Then, we obtain the Cauchy stress of the magnetic part as follows:

$$\mathbf{\Sigma}^{\mathrm{mag}} = -\frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0|} \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ g + X_3 \cdot \frac{g}{|g|}\xi & 0 & f' + X_3 \cdot \frac{f'}{|g|}\xi \end{bmatrix} \tag{S5}$$

The magnetic force as follows:

$$\begin{aligned}
\mathbf{T}_1^{\mathrm{m}} &= -\mathbf{\Sigma}^{\mathrm{mag}}\mathbf{N}_1 = \frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0} \begin{bmatrix} 0 \\ 0 \\ f' + \frac{t}{2} \cdot \frac{f'}{|g|}\xi \end{bmatrix} \\
\mathbf{T}_2^{\mathrm{m}} &= -\mathbf{\Sigma}^{\mathrm{mag}}\mathbf{N}_2 = \frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0} \begin{bmatrix} 0 \\ 0 \\ f' + \frac{t}{2} \cdot \frac{f'}{|g|}\xi \end{bmatrix} \\
\mathbf{T}_3^{\mathrm{m}} &= -\mathbf{\Sigma}^{\mathrm{mag}}\mathbf{N}_3 = \frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0} \begin{bmatrix} 0 \\ 0 \\ g + X_3 \cdot \frac{g}{|g|}\xi \end{bmatrix} \\
\mathbf{T}_4^{\mathrm{m}} &= -\mathbf{\Sigma}^{\mathrm{mag}}\mathbf{N}_4 = \frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0} \begin{bmatrix} 0 \\ 0 \\ -g - X_3 \cdot \frac{g}{|g|}\xi \end{bmatrix} \\
\mathbf{T}_5^{\mathrm{m}} &= \mathbf{T}_6^{\mathrm{m}} = \mathbf{0}
\end{aligned} \tag{S6}$$

In this case, the $\frac{P}{EI} = 0.48\mathrm{mm}^{-2}$, $l = 24\mathrm{mm}$.

The remanent magnetic flux density vector of the arc

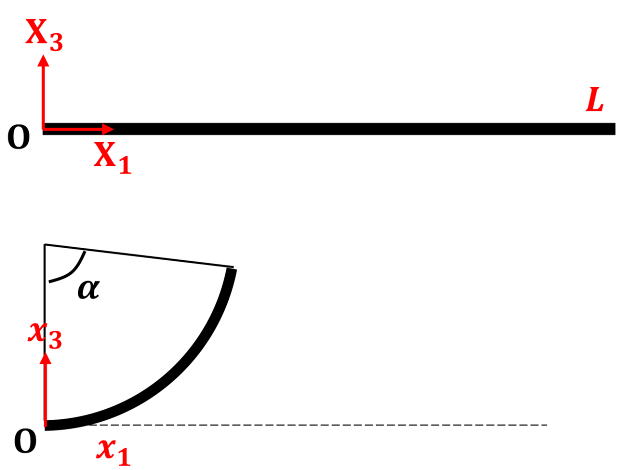

Figure S4: An arc shape

The HMSMs start in a horizontal position with dimensions of length L, width b, and thickness t. When subjected to mechanical constraints, they bend into an arc forming an angle γ. The shape function of the centerline before deformation is given by Eq. (S1)

The length of the centerline remains unchanged before and after deformation. The shape function after deformation is given by:

$$\begin{cases} x_1 = \dfrac{L}{\gamma}\sin\dfrac{\gamma X_1}{L} \\ x_3 = \dfrac{L}{\gamma}(1-\cos\dfrac{\gamma X_1}{L}) \end{cases} \tag{S7}$$

We can calculate the deformation gradient $\mathbf{F}^{\mathrm{E}}$,

$$\mathbf{F}^{\mathrm{E}} = \begin{bmatrix} \cos(\frac{\gamma X_1}{L}) - X_3 \cdot \cos(\frac{\gamma X_1}{L}) \cdot \frac{\gamma}{L} & 0 & -\sin(\frac{\gamma X_1}{L}) \\ 0 & 1 & 0 \\ \sin(\frac{\gamma X_1}{L}) - X_3 \cdot \sin(\frac{\gamma X_1}{L}) \cdot \frac{\gamma}{L} & 0 & \cos(\frac{\gamma X_1}{L}) \end{bmatrix} \tag{S8}$$

and the remanent magnetic flux density vector $\mathbf{B}_0^{\mathrm{r}}$,

$$\mathbf{B}_0^{\mathrm{r}} = J(\mathbf{F}^{\mathrm{E}})^{-1}\mathbf{B}_1^{\mathrm{r}} = \begin{bmatrix} -\sin(\frac{\gamma X_1}{L}) + X_3 \cdot \sin(\frac{\gamma X_1}{L}) \cdot \frac{\gamma}{L} \\ 0 \\ \cos(\frac{\gamma X_1}{L}) - X_3 \cdot \cos(\frac{\gamma X_1}{L}) \cdot \frac{\gamma}{L} \end{bmatrix} B_1^{\mathrm{r}} \tag{S9}$$

where $\mathbf{B}_1^{\mathrm{r}} = B_1^{\mathrm{r}}[0,0,1]^{\mathrm{T}}$, and $J = |\mathbf{F}^{\mathrm{E}}| = 1$ under the assumption of incompressibility. By setting $X_3 = 0$, we can obtain the remanent magnetic flux density vector distribution of the centerline, i.e.,

$$\mathbf{B}_0^{\mathrm{r}} = \begin{bmatrix} -\sin(\frac{\gamma X_1}{L}) \\ 0 \\ \cos(\frac{\gamma X_1}{L}) \end{bmatrix} B_1^{\mathrm{r}} \tag{S10}$$

The magnetic force of the arc

We can calculate the magnetic forces on all surfaces of the HMSMs in reference configuration. First, we obtain the Cauchy stress of the magnetic part as follows,

$$\begin{aligned} \mathbf{\Sigma}^{\mathrm{mag}} &= -\frac{1}{\mu_0}(\mathbf{B}^{\mathrm{a}} \otimes \mathbf{B}_0^{\mathrm{r}}) \\ &= -\frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0} \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ -\sin(\frac{\gamma X_1}{L}) + X_3 \cdot \sin(\frac{\gamma X_1}{L}) \cdot \frac{\gamma}{L} & 0 & \cos(\frac{\gamma X_1}{L}) - X_3 \cdot \cos(\frac{\gamma X_1}{L}) \cdot \frac{\gamma}{L} \end{bmatrix} \end{aligned} \tag{S11}$$

where $\mathbf{B}^{\mathrm{a}} = B^{\mathrm{a}}[0,0,1]^{\mathrm{T}}$, the magnetic force distribution on each surface of the HMSMs is as follows:

$$\begin{aligned}
\mathbf{T}_1^{\mathrm{m}} &= -\mathbf{\Sigma}^{\mathrm{mag}}\mathbf{N}_1 = \frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0}\begin{bmatrix} 0 \\ 0 \\ \cos(\frac{\gamma X_1}{L}) - \frac{t}{2}\cdot\cos(\frac{\gamma X_1}{L})\cdot\frac{\gamma}{L} \end{bmatrix} \\
\mathbf{T}_2^{\mathrm{m}} &= -\mathbf{\Sigma}^{\mathrm{mag}}\mathbf{N}_2 = \frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0}\begin{bmatrix} 0 \\ 0 \\ -\cos(\frac{\gamma X_1}{L}) - \frac{t}{2}\cdot\cos(\frac{\gamma X_1}{L})\cdot\frac{\gamma}{L} \end{bmatrix} \\
\mathbf{T}_3^{\mathrm{m}} &= -\mathbf{\Sigma}^{\mathrm{mag}}\mathbf{N}_3 = \frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0}\begin{bmatrix} 0 \\ 0 \\ -\sin(\gamma) + X_3\cdot\sin(\gamma)\cdot\alpha \end{bmatrix} \\
\mathbf{T}_4^{\mathrm{m}} &= \mathbf{T}_5^{\mathrm{m}} = \mathbf{T}_6^{\mathrm{m}} = \mathbf{0}
\end{aligned} \tag{S12}$$

where unit normal vector $\mathbf{N}_5 = [0,1,0]^{\mathrm{T}}$ and $\mathbf{N}_6 = [0,-1,0]^{\mathrm{T}}$, resulting corresponding magnetic force equal to zero.

The remanent magnetic flux density vector of the cosine function

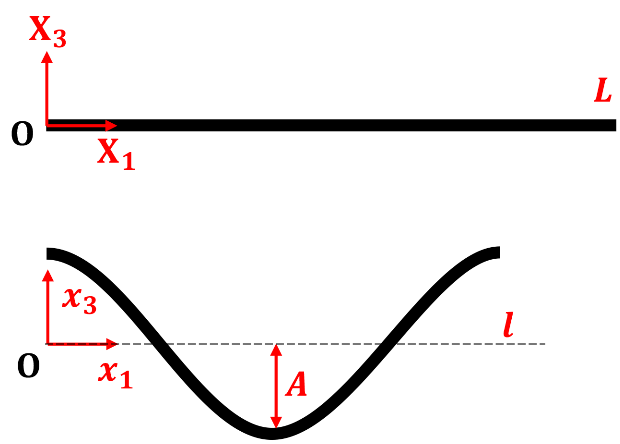

Figure S5: A cosine function shape.

The HMSMs, initially characterized by length L, width b and thickness t, transfer from a straight line into a single-period cosine curve. The initial shape function of the centerline before deformation is defined by Eq. S1. The shape function of the centerline post-deformation is expressed as:

$$\begin{cases} \displaystyle\int_0^{x_1} \sqrt{1 + \frac{4\pi^2 A^2}{l^2}\sin^2\frac{2\pi x}{l}}\,dx = X_1 \\ x_3 = A\cos\dfrac{2\pi x_1}{l} \end{cases} \tag{S13}$$

where A and l are the amplitude and wavelength of the cosine function, respectively. The transcendental function in Eq. $(\text{S13})_1$ is approximated by a high-order polynomial fit, i.e.,

$x_1 = \Phi(X_1)$. We can obtain the deformation gradient $\mathbf{F}^{\mathrm{E}}$,

$$\mathbf{F}^{\mathrm{E}} = \begin{bmatrix} \Phi' + X_3 \cdot \frac{\Phi'}{|\Psi|}\zeta & 0 & \frac{\Psi}{\sqrt{\Phi'^2+\Psi^2}} \\ 0 & 1 & 0 \\ -g - X_3 \cdot \frac{\Psi}{|\Psi|}\zeta & 0 & \frac{\Phi'}{\sqrt{\Phi'^2+\Psi^2}} \end{bmatrix} \tag{S14}$$

where $\Psi = \frac{2\pi A\Phi'}{l}\sin\frac{2\pi\Phi}{l}$ and $\zeta = \frac{\Phi''(\Phi'^2+\Psi^2)-\Phi'(2\Phi'\Phi''+2\Psi\Psi')}{\Phi'^2+\Psi^2}$. Then we can calculate the remanent magnetic flux density vector $\mathbf{B}_0^{\mathrm{r}}$,

$$\mathbf{B}_0^{\mathrm{r}} = \begin{bmatrix} g + X_3 \cdot \frac{\Psi}{|\Psi|}\zeta \\ 0 \\ \Phi' + X_3 \cdot \frac{\Phi'}{|\Psi|}\zeta \end{bmatrix} B_1^{\mathrm{r}} \tag{S15}$$

The magnetic force of the cosine function

Then, we obtain the Cauchy stress of the magnetic part as follows:

$$\mathbf{\Sigma}^{\mathrm{mag}} = -\frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0|} \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ g + X_3 \cdot \frac{\Psi}{|\Psi|}\zeta & 0 & \Phi' + X_3 \cdot \frac{\Phi'}{|\Psi|}\zeta \end{bmatrix} \tag{S16}$$

The magnetic force as follows:

$$\begin{aligned} \mathbf{T}_1^{\mathrm{m}} &= \frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0} \begin{bmatrix} 0 \\ 0 \\ \Phi' + \frac{t}{2} \cdot \frac{\Phi'}{|\Psi|}\zeta \end{bmatrix} \\ \mathbf{T}_2^{\mathrm{m}} &= \frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0} \begin{bmatrix} 0 \\ 0 \\ -\Phi' + \frac{t}{2} \cdot \frac{\Phi'}{|\Psi|}\zeta \end{bmatrix} \\ \mathbf{T}_3^{\mathrm{m}} &= \frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0} \begin{bmatrix} 0 \\ 0 \\ \Psi + X_3 \cdot \frac{\Psi}{|\Psi|}\zeta \end{bmatrix} \\ \mathbf{T}_4^{\mathrm{m}} &= \frac{B^{\mathrm{a}}B_1^{\mathrm{r}}}{\mu_0} \begin{bmatrix} 0 \\ 0 \\ -\Psi - X_3 \cdot \frac{\Psi}{|\Psi|}\zeta \end{bmatrix} \\ \mathbf{T}_5^{\mathrm{m}} &= \mathbf{T}_6^{\mathrm{m}} = \mathbf{0} \end{aligned} \tag{S17}$$

Based on the calculations, we determined the magnetic force distribution of HMSMs under an actuating magnetic field $\mathbf{B}^{\mathrm{a}}$. By converting the complex magneto-elastic coupling problem into a purely elastic one, we applied the magnetic force as a mechanical load in finite element software. This approach enabled us to efficiently predict the recovery behavior of HMSMs under the actuating magnetic field.