MRS Communications (2024) 14:835–845

https://doi.org/10.1557/s43579-024-00624-6

MRS Communications

Early Career Materials Researcher Prospective

Rethinking hysteresis in magnetic materials

Ananya Renuka Balakrishna, Materials Department, University of California, Santa Barbara, CA 93106, USA

Address all correspondence to Ananya Renuka Balakrishna at **ananyarb@ucsb.edu**

(Received 4 April 2024; accepted 23 July 2024; published online: 8 August 2024)

Abstract

Magnetic materials with zero hysteresis are posed to have a significant impact on sustainable energy conversion, electronics, and communication technologies. As the global market for soft magnetic materials continues to expand, driven by increasing demand in renewable energy, automotive, and power transmission sectors, it is important to design magnets that can be cycled under an external field reversibly and rapidly multiple times without a decay in magnetic response. However, designing magnets with small hysteresis has been a challenge because we do not fully understand its origins. In this article, I outline key research efforts investigating the fundamental mechanisms underpinning hysteresis in soft magnets and, highlight recent developments in the use of nonlinear analysis and nucleation barrier methods to predict coercivity in these materials.

Magnetic hysteresis

Most of the interest in magnetic materials derives from the fact that the intrinsic magnetic dipole can be reoriented, reversibly and rapidly, under an external field. However, this reorienting of the magnetic dipole commonly exhibits a lag in response to the changing external magnetic field. This characteristic lag in the material response is called the *hysteresis effect* and is a widespread phenomenon observed in ferromagnets,[1, 2] see [Fig. 1(a)]. This hysteresis is not only important to the design of memory-based devices but also signifies the energy lost during the repeated and reversible cycling of magnets.[3, 4] Despite the common occurrence of hysteresis across magnets, we lack a clear understanding of its origins and cannot accurately predict its nonlinear manifestation under an applied field.[5–7] This gap in knowledge limits our ability to design and discover new magnetic materials with zero hysteresis.

Let us consider the example of the permalloy problem: In 1953, a magnetic alloy ($Fe_{21.5}Ni_{78.5}$) with unusually low hysteresis was discovered at the Bell Laboratories.[12] This alloy, now known as the permalloy, resulted from a series of investigations on the iron-nickel alloy system. Experimentalists systematically varied the percent of nickel content in the Fe–Ni alloy and investigated the effect of composition on coercivity. The results showed a drastic decrease in coercivity at the 78.5% Ni-content (i.e., the permalloy). Several researchers have attributed this decrease in coercivity to the permalloy's small anisotropy constant—a material constant that quantifies the difficulty of rotating magnetization away from a preferred crystallographic axis—however, a closer examination of the Fe–Ni alloy composition shows several peculiarities in behavior that contradict our current understanding of the origins of magnetic hysteresis.[13] For example, the anisotropy constant of the $Fe_{25}Ni_{75}$ alloy is zero, however, there is not even a local minimum of the coercivity at this composition.[13] By contrast, the coercivity is relatively small in the $Fe_{55}Ni_{45}$ alloy, although its anisotropy constant is relatively large (e.g., $\kappa_1 = 1000 \mathrm{J/m^3}$). Besides the anisotropy constant, researchers have suspected that magnetostriction constants (λ_{100}, λ_{111}) play a role in lowering hysteresis.[10, 14, 15] These discrepancies have been observed in other magnetic alloys, including the Fe–Si system and Fe–Co system;[16, 17] however, there is no unified theory that explains the precise role of magnetic material constants on hysteresis.[13, 16]

Kersten and Becker[15] explain magnetic hysteresis using the domain theory.[10] In this framework, a delay in magnetization reversal is attributed to domain wall pinning and the energy dissipated by phase boundary propagation, see [Fig. 1(b)]. Kersten's theory of inclusions suggests that domain walls minimize their surface area when pinned to a defect and are in equilibrium, and therefore, one has to supply additional energy to induce magnetization reversal.[8] Similarly, Becker postulates that the local variation in surface energy of the domain walls (e.g., arising from magnetostriction and elastic deformation) plays an important role in governing magnetization reversal.[9] These analyses show a correlation between non-magnetic inclusions (defects) and hysteresis in annealed carbon steels,[8] however, the calculations do not account for the significant effect of the demagnetization field in the proximity of inclusions and assume a simple planar geometry for domain walls, see [Fig. 1(b)].

Besides domain wall pinning effects, magnetic constants such as magnetocrystalline anisotropy and magnetostriction[12] are known to affect the hysteresis width in soft magnetic alloys. For example, the magnetocrystalline anisotropy constant, which represents the barrier for magnetization switching, is known to directly impact magnetization reversal.[5, 25] Additionally, the zero magnetostriction constants, e.g., $\lambda_{111} = 0$ in the $Fe_{20}Ni_{80}$ alloy and $\lambda_{100} = 0$ in the $Fe_{55}Ni_{45}$ alloy, are proposed to offer lower resistance to magnetization rotation and are correlated with small coercivities,[12, 13] see [Fig. 2(a)].[18]

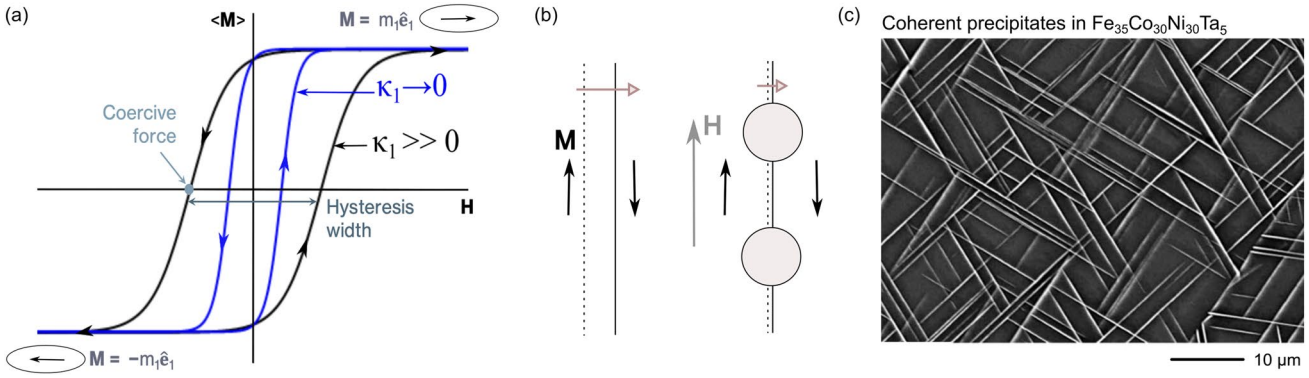

Figure 1. (a) A schematic illustration of magnetic hysteresis loops. These loops show a relation between average magnetization in the ellipsoid $< \mathbf{M} >$ under an applied field **H**. The value of the external field at which the average magnetization in the ellipsoid reduces to zero is referred to as the coercive force, and the width of the loop is referred to as the hysteresis width. Commonly, soft magnetic materials with small anisotropy constants $\kappa_1 \to 0$ are characterized to having small hysteresis. (b) The domain wall pinning theory[8–10] suggests that defects such as non-magnetic inclusions or cavities stabilize domain walls by reducing their surface areas. This pinning, consequently, requires a large driving force for magnetization reversal and contributes to an increase in the hysteresis width. However, this explanation overlooks the large demagnetization fields that are generated in the vicinity of defects and affects magnetization reversal.[6] (c) Magnetic alloys with high aspect ratio precipitates have reduced domain wall pinning and are characterized by smaller hysteresis.[11] Subfigure (c) is adapted from Ref. 11 and is licensed under CC BY 4.0.

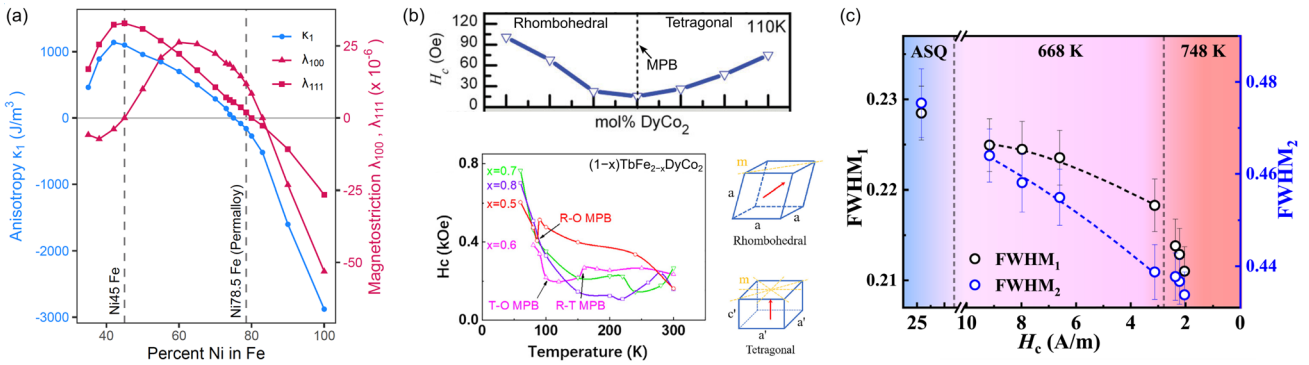

Figure 2. (a) Fundamental magnetic material constants are known to affect magnetic hysteresis. For example, the researchers at the Bell laboratories systematically synthesized Fe–Ni alloys (as a function of Ni-composition) and show that at 78.5% Ni in Fe (Permalloy) magnetic hysteresis is the smallest. A puzzling feature about this observation is that at 78.5% Ni, neither anisotropy κ_1 nor magnetostriction constants λ_{100}, λ_{111}, are zero, however, magnetic hysteresis is minimum.[12, 18] (b) The presence of morphotropic phase boundaries, e.g., between the rhombohedral and tetragonal phases in $DyCo_2$, is correlated with reduced hysteresis.[19–21] These phase boundaries are postulated to increase the magnetoelastic response in the alloy and are a factor in lowering hysteresis widths. Subfigure (b) is reprinted with permission from Ref. 19 (Copyright 2010 by the American Physical Society) and Ref. 20 (Copyright 2021 with permission from Elsevier). (c) Magnetic alloys with nanocrystalline and/or amorphous microstructural features are a special class of soft magnetic alloys with small collective anisotropies and are known to have small hysteresis.[22–24] Subfigure (c) is reprinted with permission from Ref. 23 (Copyright 2022 with permission from Elsevier).

Microstructural features, such as the morphotropic phase boundaries, are also correlated with reducing coercivity in magnetic materials.[19, 21] These boundaries separating magnetic domains with different lattice geometries (e.g., rhombohedral and tetragonal phases in $TbCo_2$–$DyCo_2$[19]) generate significant magnetoelastic interactions that are postulated to reduce hysteresis in magnetic materials,[21] see [Fig. 2(b)]. More recently, nanocrystalline and amorphous alloys, which are a class of magnetic alloys with small collective anisotropy constants, have been shown to have small magnetic hysteresis,[22–24] see [Fig. 2(c)]. Each of the theories provides key insights into the individual roles of the anisotropy constant, microstructural evolution pathways, and elastic interactions affecting the switching of magnetization under an external field. However, the overall mechanism underpinning magnetic hysteresis is still unclear.

Attributing magnetic hysteresis to domain wall pinning, anisotropy constant, or magnetoelastic responses alone does not paint a complete picture of the origins of magnetic hysteresis. Néel highlighted similar shortcomings in his early studies of coercive force in magnets and called for a rigorous mathematical description to study the interplay between different mechanisms

governing magnetization reversal.[6] Furthermore, the role of 'internal magnetic charges' or the corresponding demagnetization fields generated in the vicinity of non-magnetic inclusions or defects offers significant energy barriers. It is important to analyze the balance between these different energy contributions in a unified theoretical framework that takes into account various magnetic constants and microstructural features, and describe their interplay on magnetization reversal.

Micromagnetics

The micromagnetics framework, proposed by Brown,[5, 25] is a continuum theory based on a concept of continuous magnetization. In this framework, the different energy contributions, such as magnetocrystalline anisotropy and magnetostatic energy terms, are accounted for in a single model. This framework has been successfully applied to find energy-minimizing microstructures that arise naturally as a consequence of the theory[28] and to predict the magnetic response of materials used in recording devices and tapes.[29] A distinguishing feature of micromagnetics is that the total energy is expressed in terms of conventional material constants that are measured in specific experiments on bulk magnetic materials (and are not fitted parameters to a measured macroscopic response of the material):

$$\begin{aligned}\psi = &\int_{\mathcal{E}} \{\nabla \mathbf{m} \cdot \mathrm{A} \nabla \mathbf{m} + \kappa_1(\mathrm{m}_1^2\mathrm{m}_2^2 + \mathrm{m}_2^2\mathrm{m}_3^2 + \mathrm{m}_3^2\mathrm{m}_1^2) \\ &+ \frac{1}{2}[\mathbf{E} - \mathbf{E}_0(\mathbf{m})] \cdot \mathbb{C}[\mathbf{E} - \mathbf{E}_0(\mathbf{m})]\}\mathrm{d}\mathbf{x} \\ &\int_{\mathcal{E}} -\mu_0 m_s \mathbf{H}_\mathrm{e} \cdot \mathbf{m}\mathrm{d}\mathbf{x} + \frac{\mu_0}{2}\int_{\mathbb{R}^3} |\nabla \zeta_\mathbf{m}|^2 \mathrm{d}\mathbf{x}.\end{aligned} \tag{1}$$

The micromagnetics energy in Eq. 1 is a function of magnetization $\mathbf{m} = \frac{\mathbf{M}}{m_s} = \mathrm{m}_1\hat{\mathbf{e}}_1 + \mathrm{m}_2\hat{\mathbf{e}}_2 + \mathrm{m}_3\hat{\mathbf{e}}_3$ and strain $\mathbf{E}$ tensors and is described for a magnetic ellipsoid body $\mathcal{E}$ in a three-dimensional Euclidean space $\mathbb{R}^3$. The gradient energy term $\int_{\mathcal{E}} \nabla \mathbf{m} \cdot \mathrm{A} \nabla \mathbf{m} \mathrm{d}\mathbf{x}$ penalizes changes in the magnetization and the magnetocrystalline anisotropy energy term $\int_{\mathcal{E}} \kappa_1(\mathrm{m}_1^2\mathrm{m}_2^2 + \mathrm{m}_2^2\mathrm{m}_3^2 + \mathrm{m}_3^2\mathrm{m}_1^2)\mathrm{d}\mathbf{x}$ penalizes magnetization rotation away from the easy axes. The elastic energy term $\int_{\mathcal{E}} \frac{1}{2}[\mathbf{E} - \mathbf{E}_0(\mathbf{m})] \cdot \mathbb{C}[\mathbf{E} - \mathbf{E}_0(\mathbf{m})]\mathrm{d}\mathbf{x}$ is related to the magnetostrictive response of the material. We consider geometrically linear strains $\mathbf{E}$, which deviate from the spontaneous strain values $\mathbf{E}_0(\mathbf{m})$, and are calculated by solving the mechanical equilibrium condition $\nabla \cdot \sigma = \nabla \cdot \mathbb{C}[\mathbf{E} - \mathbf{E}_0(\mathbf{m})] = 0$. The spontaneous strain tensor $\mathbf{E}_0(\mathbf{m})$ is a function of the magnetostriction constants $\lambda_{100}, \lambda_{111}$ that describes lattice deformation accompanying magnetization switching:

$$\mathbf{E}_0(\mathbf{m}) = \frac{3}{2}\begin{bmatrix} \lambda_{100}(\mathrm{m}_1^2 - \frac{1}{3}) & \lambda_{111}\mathrm{m}_1\mathrm{m}_2 & \lambda_{111}\mathrm{m}_1\mathrm{m}_3 \\ & \lambda_{100}(\mathrm{m}_2^2 - \frac{1}{3}) & \lambda_{111}\mathrm{m}_2\mathrm{m}_3 \\ \textit{symm.} & & \lambda_{100}(\mathrm{m}_3^2 - \frac{1}{3}) \end{bmatrix} \tag{2}$$

The magnetostriction constants for common soft magnetic alloys are small ($\lambda_{100}, \lambda_{111} \approx 10^{-6}$), and the magnetoelastic energy term is often neglected in the coercivity calculations. The external energy $\int_{\mathcal{E}} -\mu_0 m_s \mathbf{H}_\mathrm{e} \cdot \mathbf{m}\mathrm{d}\mathbf{x}$ accounts for mutual interaction between the magnetization vector and the applied external field. Finally, the magnetostatic energy term $\frac{\mu_0}{2}\int_{\mathbb{R}^3} |\nabla \zeta_\mathbf{m}|^2 \mathrm{d}\mathbf{x}$ is related to the work required to arrange magnetic dipoles into a specific geometric configuration. This energy term scales quadratically with the demagnetization field, $\mathbf{H}_d = -\nabla \zeta_\mathbf{m}$, which we compute by solving magnetostatic equilibrium $\nabla \cdot (\mathbf{H}_d + \mathbf{M}) = 0$ on all of space. This magnetostatic energy can be significant in the vicinity of defects and plays an important role in magnetization switching.

Micromagnetics serves as an ideal framework to investigate the links between material constants, microstructural patterns, and applied magnetic fields.[28–30] However, a well-known difficulty with the theory, popularized as the "coercivity paradox" by Brown,[5, 25] is that a linear approximation of a uniformly magnetized domain in the micromagnetics framework fails to predict coercive forces quantitatively. For example, to estimate the coercivity of a magnetic material, researchers study the breakdown of a uniformly magnetized single domain under a constant external field, see [Fig. 3(a)]. In this approach, a large external field is applied across a magnetized ellipsoid, and its value is reduced until the magnetization reorients with the applied field (i.e., a linear stability analysis of a uniformly magnetized domain). This linear approximation predicts the coercive force for a magnet to linearly depend on its anisotropy constant and inversely on its saturation magnetization, $\mathbf{H}_e \approx \frac{2\kappa_1}{m_s}$. However, substituting the anisotropy constant κ_1 and saturation magnetization m_s of well-characterized materials such as Fe–Ni alloys in the $\mathbf{H}_e \approx \frac{2\kappa_1}{m_s}$ expression, the coercive force predictions are often 2–3 orders of magnitude larger than the corresponding experimental measurements. This discrepancy between theory and experiments is known as the "coercivity paradox,"[5, 25] and several continuum models developed today use linear approximations of micromagnetics. This approach often overestimates the coercivity on the shoulder of hysteresis loops, see [Fig. 3(b, c)].[26, 27]

In recent times, researchers have been developing modeling tools based on machine learning methods to predict coercivity in magnetic materials.[31–34] For example, Foggiatto et al.[34] analyze morphological defects in magnetization images and use techniques such as principal component analysis to construct a realistic free energy landscape.[34] In these studies, researchers predict coercive forces as an interplay between magnetization and microstructural features in the material[27, 32, 34] and emphasize the role of morphological and structural defects in computing magnetic hysteresis.[34] These works build on prior efforts in which topological defects such as dislocations and grain boundaries are shown to affect the driving forces for magnetization reversal.[35] Similarly, the role of mechanics such as elastic strains at morphotropic phase boundaries and residual stresses arising from heat treatment of alloys have all been identified to contribute to magnetic hysteresis,[13] see [Fig. 3(c)]. The multitude of factors that contribute to magnetic hysteresis is

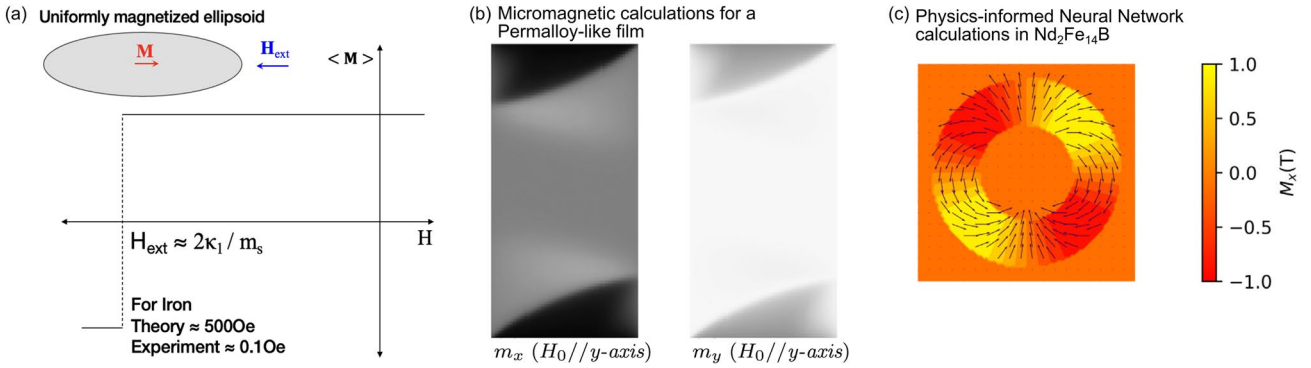

Figure 3. (a) In a micromagnetics framework, the linear approximation of a uniformly magnetized ellipsoid predicts a coercive force $H_c \approx 2\kappa_1/m_s$ (with m_s representing the saturation magnetization of the material). Using this analytical relation, the coercive force for iron is predicted to be 500 Oe, which is three orders of magnitude larger than the experimentally measured coercivity for iron. This discrepancy between theory and experiments is referred to as the "coercivity paradox."[5] (b, c) Today, numerical micromagnetic calculations commonly use the linear stability analysis approach to predict magnetization switching.[26, 27] Subfigures (b) and (c) are reprinted with permission from Ref. 26 (Copyright 2010, with permission from Elsevier) and Ref. 27 (Creative Commons CC-BY 4.0 license), respectively.

enormous; however, there is a need for a general or unifying theory to interpret magnetic hysteresis that is applicable to all magnetic materials, irrespective of their defect types, defect distributions, and/or magnetoelastic interactions.

Nucleation barriers

Recent progress in the use of nonlinear analysis to interpret hysteresis in shape-memory alloys—another class of phase transformation materials[1, 36, 37]—motivates us to rethink our approach of computing hysteresis in magnetic alloys. In this nonlinear analysis methodology, researchers analyze a pre-existing nucleus of a transformed phase and estimate the energy barriers limiting the growth of this nucleus. For example, in shape-memory alloys Zhang et al.,[1] assume a small lenticular-shaped nucleus of martensitic phase, see [Fig. 4(a)]. This nucleus, often stabilized at a grain boundary or a triple junction, exists at temperatures above which the martensitic phase is unfavorable. During phase transformation, this nucleus grows and the energy barrier limiting this growth primarily governs hysteresis widths. This energy barrier is postulated to arise from the elastic energy stored at the austenite/martensite interface. Similarly, Knüpfer et al.,[36] have analyzed the nucleation barriers in shape-memory alloys undergoing a cubic to tetragonal transformation and correlated the energy dissipated during transformation with the width of the hysteresis loop.

By minimizing the energy barriers, a mathematical criterion emerges for which the energy barriers and thermal hysteresis can be reduced. For shape-memory alloys, these criteria correspond to a special combination of lattice parameters that minimize lattice misfit strains and are commonly referred to as the $\lambda_2 = 1$ design rule.[1, 37] This $\lambda_2 = 1$ criterion corresponds to a lattice deformation in which one of the sides is relatively unstretched with respect to its reference configuration and serves as a simple design rule to discover shape-memory alloys with small hysteresis and improved reversibility. Researchers have systematically tuned alloy compositions to satisfy specific lattice geometries, and the resulting alloys show incredible improvements in reversible cycling and a dramatic drop in thermal hysteresis,[37, 40, 41] see [Fig. 4(b, d)]. For example, a Ti-Ni-Cu alloy, which typically develops fatigue after 200 stress–strain cycles, was systematically doped to form low-elastic-energy microstructures.[40] The resulting alloy $Ti_{54}Ni_{34}Cu_{12}$ was reversibly cycled for over ten million cycles and showed negligible fatigue despite its large transformation strain ($\sim 8\%$).[40] Beyond shape-memory alloys, researchers have stabilized these highly reversible microstructures in ferroelectric energy harvesters,[42, 43] in magnetocaloric Heusler alloys,[44] and to enhance the reversible metal-insulator phase transformations.[45]

Recently, a stronger set of criteria, known as the 'cofactor conditions', has been proposed by James and co-workers[37, 41] that greatly improves material reversibility. Following these conditions, James and co-workers[41] identified a new alloy composition, $Zn_{45}Au_{30}Cu_{25}$, that shows remarkable reversibility. This alloy undergoes a cubic-to-monoclinic transformation and is accompanied by $\sim 8\%$ lattice strains; however, it shows only a migration of 0.5°C despite cycling the material over 16,000 times. This reversibility is because the $Zn_{45}Au_{30}Cu_{25}$ alloy can form zero-elastic-energy interfaces and has multiple twin solutions that collectively generate microstructures that accommodate around defects with minimum stresses during phase transformation.

These recent developments in using nonlinear approximations and nucleation barriers have had a transformative impact on designing shape-memory alloys with small hysteresis and enhanced reversibility. The success of these methods has inspired our research to seek whether a mathematical criterion, analogous to the $\lambda_2 = 1$ criterion, exists for soft magnetic materials with small magnetic hysteresis.

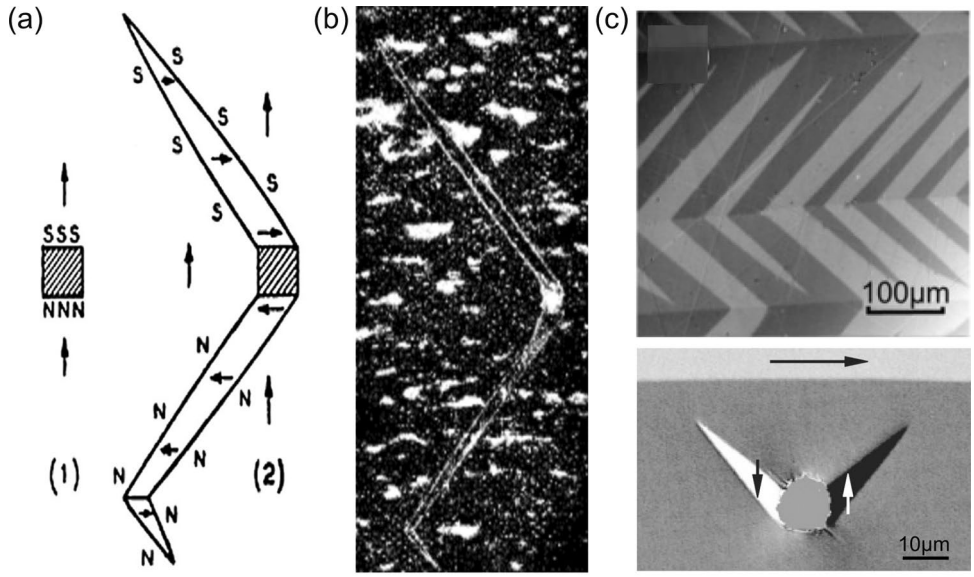

Figure 4. (a) In shape-memory alloys, another type of phase transformation material, a self-accommodating martensite microstructure is assumed to serve as a potent nucleus. This nucleus grows during phase transformation, and the energy barrier limiting the growth of this nucleus is theorized to contribute to the finite width of the thermal hysteresis loop.[36, 38] (b) For a specific combination of lattice geometries, now referred to as the cofactor conditions with the middle eigenvalue $\lambda_2 = 1$ being a key criteria, the energy barrier limiting the growth of the martensitic nucleus is minimized. Shape-memory alloys satisfying this specific lattice compatibility (i.e., $\lambda_2 = 1$) are shown to have a dramatic drop in thermal hysteresis.[39] (c) A riverine-like microstructure is observed in the $Zn_{45}Au_{30}Cu_{25}$ alloy that is highly reversible. Similar microstructures in (d) Ti-Ni-Cu alloys have contributed to ultra-low fatigue materials.[40] Subfigure (a) is adapted from Ref. 36, 38 with permission from John Wiley and Sons and the Copyright Clearance Center. Subfigure (b) is adapted from Ref. 39 with permission from Copyright 2006 Springer Nature. Subfigure (c) is adapted from Ref. 41 with permission from Copyright 2013 Springer Nature. Subfigure (d) is adapted from Ref. 40 with permission from AAAS and CCC.

Localized disturbance and hysteresis

In our recent and ongoing works,[2, 18, 46] we are using nonlinear approximations and nucleation barrier analysis to investigate magnetization reversal. A first step in this approach is to identify a potent nucleus that is pre-existing in a soft magnetic material and which serves as a large localized disturbance that grows under an external magnetic field. To this end, we choose a needle domain as a potent nucleus, and note two seminal works that motivated our choice of this needle domain.[47, 48]

First is the early study on coercive forces by Néel, in which he predicted that a spike-like (or needle-like) domain forms around defects such as cavities or non-magnetic inclusions.[6, 47] He argued that the simple domain construction comprising uniformly magnetized domains in the vicinity of defects assumed in previous works is energetically unfavorable, see [Fig. 1(b)]. This is because defects generate a large demagnetization field that is not accounted for in the earlier calculations.[8, 15] Using analytical constructions, Néel showed that a spike-like domain forms around these defects to reduce the demagnetization energy, see [Fig. 5(a)]. These spike domains were later confirmed experimentally by Williams,[49] see [Fig. 5(b)], and today, they have been widely imaged in a variety of soft magnetic materials, see [Fig. 5(c)].[50] For example, these domains commonly occur at grain boundaries, cavity/pore defects, and sharp edges/corners in magnetic materials and do not disappear when the applied field is switched off. The needle domains serve as potent nuclei in our energy barrier calculations.[2, 46]

Second is a doctoral dissertation work by Pilet,[48] in which he observed the recurring appearance of reverse domains under an external magnetic field. The nuclei of reverse domains were observed precisely at the same defect location in the material on each magnetization reversal cycle and appeared on the shoulder of the hysteresis loop. Pilet reported a correlation between the formation of these nuclei and the coercive force measured for Ni films. Similarly, Schäfer and co-workers imaged needle-like domains in the $Fe_{83}Ga_{17}$ alloy that tend to grow indefinitely (first in length and then in width) under critical values of an externally applied field.[51] These nuclei serve as localized disturbances to the uniformly magnetized domain, and the growth of these nuclei under an external field is not likely captured by linear stability analysis. These observations, combined with the nucleation barrier concept, suggest a way forward for interpreting hysteresis in magnetic materials and form the thesis of our research on the origins of hysteresis in soft magnetic materials.[2, 18, 46, 52]

We are developing a coercivity tool in which we model the growth of a needle domain (pre-existing nucleus) under an external field,[46] see [Fig. 6(a)]. We hypothesize that the energy barriers limiting the growth of this needle domain play an important role in the hysteresis phenomena in soft magnets. Overcoming these energy barriers, shaped by fundamental magnetic material

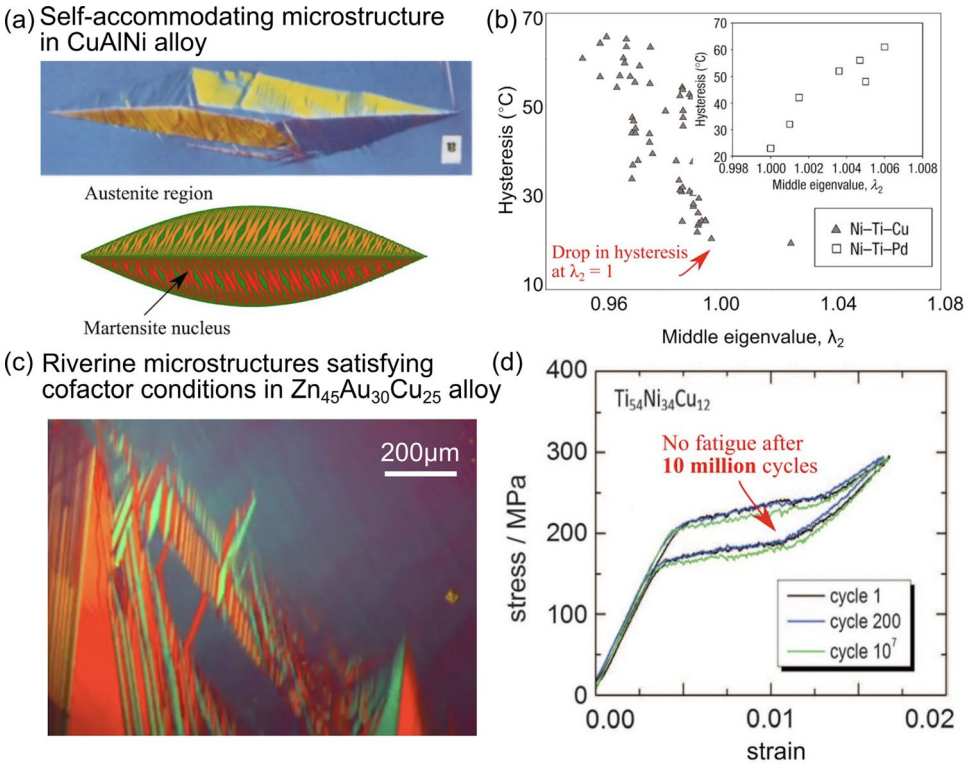

Figure 5. (a) Néel hypothesized that sharp needle-like domains form around defects, such as non-magnetic inclusions or cavities, to minimize the demagnetization field energy in soft magnets.[6, 47] (b) These needle-like domains were imaged by Williams in the Fe–Si alloy,[49] and (c) have since been commonly observed in soft magnetic materials at grain boundaries and/or pore-like defects.[50] Subfigures (a, b) and (c) are reprinted from Ref. 49 (Copyright 1947 by the American Physical Society) and Ref. 50 (with permission from Springer Nature), respectively.

constants, requires an additional driving force that we show to contribute to the finite width of hysteresis loops, see [Fig. 6(b, c)].[2, 18, 46] Our coercivity tool is rooted in micromagnetics with magnetostriction, and unlike previous research, we compute a nonlinear stability analysis (i.e., model the growth of a needle domain) of a magnetized domain.[46] That is, in our calculations, we assume the magnetic body to be an ellipsoid $\mathcal{E}$ that supports a magnetization $\mathbf{m}(\mathbf{x})$ and comprises a non-magnetic inclusion defect at its geometric center, see [Fig. 6(a)].

The presence of a defect introduces a local perturbation to the uniform magnetization on the ellipsoid and generates needle-like domains in its vicinity. These needle domains are similar to those observed in experiments[49, 50] and form to reduce the demagnetization energy and eliminate magnetic poles on the (non-magnetic) defect. Under an external field, these needle domains grow first in length and then in width.[46] This growth is modest at first and lowers the contribution of the mutual energy (or Zeeman energy) and the demagnetization energy. At a coercive field value, the needle domains abruptly grow into a stripe-like pattern and induce a complete reversal of magnetization in the ellipsoid, see [Fig. 6(b)]. Figure 6(c) shows the corresponding microstructural states on the hysteresis loop.[46]

Using our modeling framework, we predict the coercive force of iron to be ~ 5 Oe. This coercive force value is an order of magnitude larger than those reported in experiments for iron,[5, 12, 13] however, is more accurate than the coercivity predictions from linear stability analysis.[5, 25] We attribute this discrepancy between our modeling predictions and experiments to several factors. First is our simplified assumption of a perfectly smooth ellipsoid body and the presence of a single localized disturbance (needle domains) at the ellipsoid center. Second, features such as the geometry and orientation of the needle domains could also affect the coercivity calculations. We provide details on these assumptions and describe the ellipsoid theorem, reciprocal theorem, and Gauss-Siedel projection methods that were used to develop our modeling framework, which are discussed in detail in Ref. 46.

We have applied this model to understand the permalloy problem in soft magnets-that is, why the specific composition of the $Fe_{21.5}Ni_{78.5}$ alloy shows the smallest hysteresis when there is no obvious signal of this behavior,[18] see [Fig. 7(a)]. By introducing the localized disturbance, we capture the delicate balance between magnetic material constants that govern hysteresis and predict coercivity with greater accuracy than other methods based on linear stability analysis. Our work reveals insights into the magnetoelastic energy barriers that dominate the width of the hysteresis loop in soft magnets.[2, 25] The key feature of our work is that we show a delicate balance

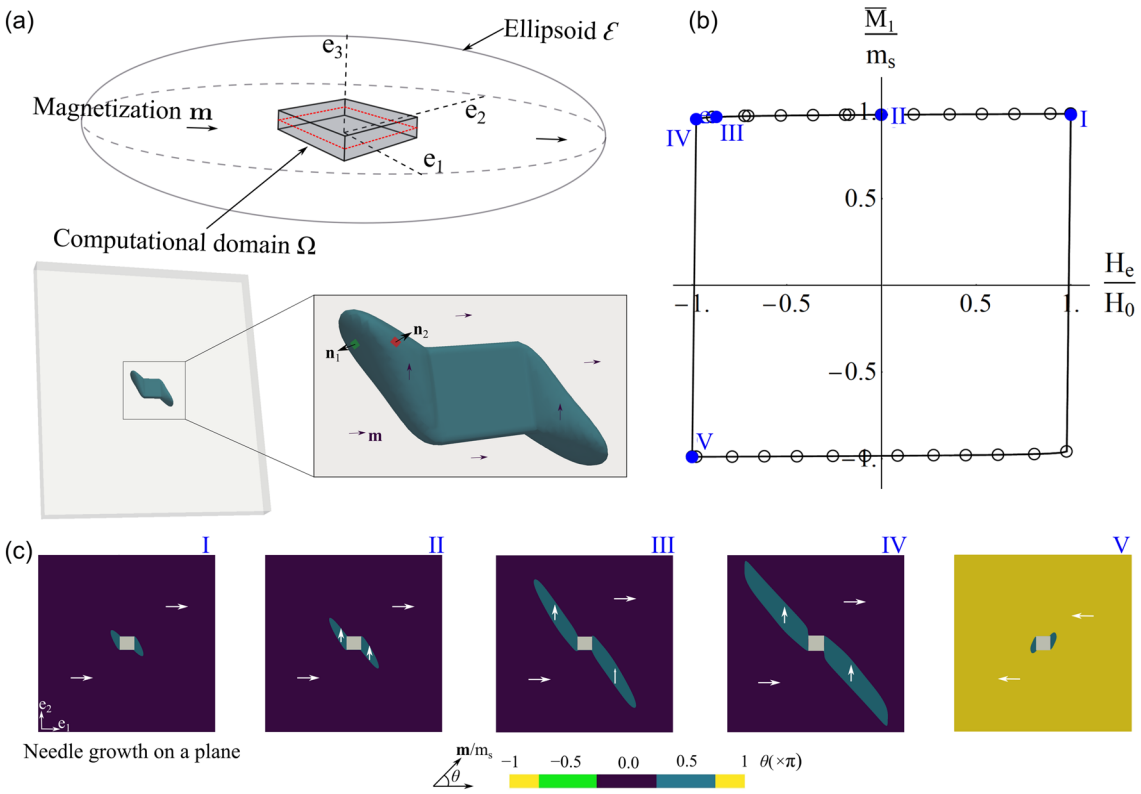

Figure 6. (a) We compute the hysteresis of a uniformly magnetized ellipsoid body with a large localized disturbance (e.g., a needle-like domain) at its center.[2, 46] In our computations, we model a domain Ω and analyze the growth of the needle domain under an external field. Subfigure (a) is adapted from Ref. 2 and is licensed under CC BY 4.0. (b) Our calculations predict a square magnetic hysteresis loop, and the corresponding microstructural features (I–V) are mapped. (c) The needle domain grows rapidly under an applied field, and at the shoulder of the hysteresis curve (IV–V), the average magnetization in the domain reverses rapidly. Subfigures (b, c) are adapted from Ref. 46, Copyright 2021, with permission from Elsevier.

between material constants, $\kappa_1, \lambda_{100}, \lambda_{111}$, is necessary for the low coercivity at 78.5% Ni-content, see [Fig. 7(a)]. This finding contrasts with some reports in the literature in which the small anisotropy constant $\kappa_1 \to 0$ is considered to be the *only* factor responsible for lowering hysteresis.[13, 53] Although $\kappa_1 = 0$ lowers the coercivity (e.g., at 75% Ni-content), we find that magnetostriction also plays an important role in governing magnetic hysteresis.

In our ongoing work, we are systematically investigating the delicate interplay between fundamental material constants on the coercive force and deriving a mathematical criterion for soft magnets (analogous to the $\lambda_2 = 1$ criterion for shape-memory alloys) for which magnetic hysteresis is minimized. Our initial results demonstrate a relation $\kappa_1 \propto \lambda_{100}^2$ for which magnetic hysteresis is reduced (κ_1, λ_{100} are magnetic material constants),[2, 46] see [Fig. 7(b, c)]. These analyses are based on minimizing the energy barrier corresponding to the growth of a localized disturbance (or a potent nucleus), such as the needle-like domain. A key feature of our work is that we demonstrate that the magnetostriction constants play an important role in governing magnetic coercivity.[2, 18] These findings contrast with prior research in which the anisotropy constant has been regarded as the *only* material parameter that governs magnetic hysteresis, and the contribution from magnetostriction constants has been largely neglected. However, by accounting for both magnetocrystalline anisotropy and magnetostriction constants in a nonlinear micromagnetics framework, we show that magnetic materials, despite their large κ_1 values, can have small coercivities for suitable values of the magnetostriction constants. Based on these results, we propose a relationship $\frac{(c_{11}-c_{12})\lambda_{100}^2}{2\kappa_1} \approx 81$ between magnetic material constants at which the coercivity is small,[2] see [Fig. 7(b, c)]. This generic formula would serve as a theoretical guide to the magnetic materials development program by suggesting alternative combinations of material constants—beyond $\kappa_1 = 0$—to develop novel soft magnets.

A general theory of magnetic hysteresis

Moving forward, we are working to expand the material parameter space to search for new combinations of magnetic constants, including $\lambda_{100}, \lambda_{111}, \mathbb{C}$, with small hysteresis.[54] Our recent work shows that in addition to designing fundamental material constants, one can introduce mechanical

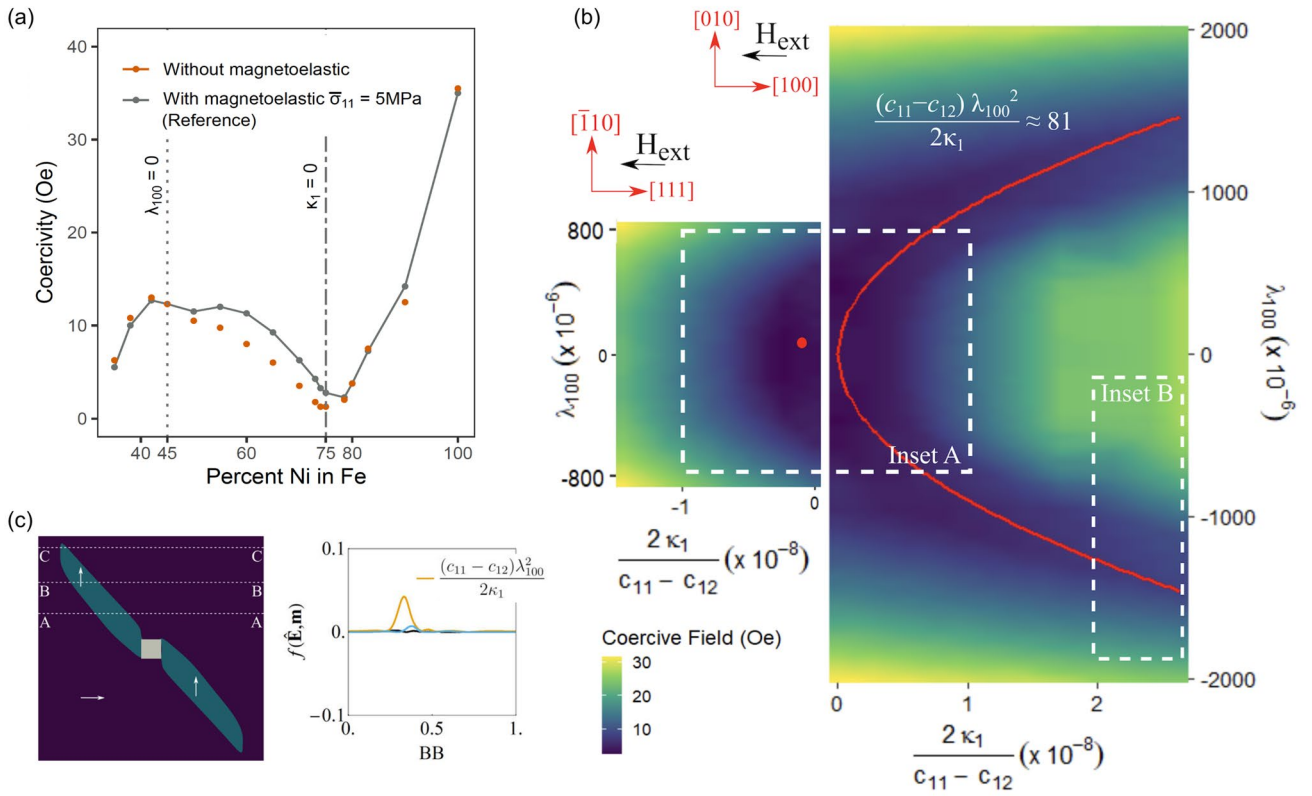

Figure 7. (a) Our approach to interpreting hysteresis in terms of localized disturbances and nucleation barriers sheds fundamental insights into the permalloy problem.[18] Our calculations show that magnetoelastic interactions, however small, have an important effect on lowering hysteresis at the permalloy composition. Subfigure (a) is adapted from Ref. 18 with the permission of AIP Publishing. (b) We systematically investigated the interplay between fundamental magnetic constants and identified a parabolic relationship $\kappa_1 \propto \lambda_{100}^2$ for which magnetic hysteresis is minimum.[2] This relationship serves as a material design rule that could guide the synthesis of new soft magnetic materials with small hysteresis. (c) Our energy barrier analysis reveals that the magnetoelastic energy term (e.g., with coefficient $\frac{(c_{11}-c_{12})\lambda_{100}^2}{2\kappa_1}$) serves as a primary energy barrier limiting magnetization reversal.[2] Subfigures (b, c) are adapted from Ref. 2 and are licensed under CC BY 4.0.

constraints, such as epitaxial strains or volumetric strains, to reduce magnetic hysteresis width.[54] For example, we show that compressive residual stresses reduce magnetic hysteresis and affect the parabolic locus for minimum coercivity. Furthermore, we show how residual stresses affect the shape and size of hysteresis loops, which would be relevant to the application of magnetic materials in memory and actuator devices.

Beyond magnetic hysteresis, we are analyzing the relation between material constants and microstructures that are conducive to low material fatigue and high reversibility in magnetocaloric materials.[52] For example, our microstructural analysis shows that magnetic materials with $\kappa_1 \leq 0, \kappa_2 \geq \frac{9|\kappa_1|}{4}$ and $\lambda_{111} = -\frac{\lambda_{100}}{3}$ form exactly-compatible and divergence-free phase boundaries[52] during paramagnetic to ferromagnetic transformations, see [Fig. 8(a)]. These relations are consistent with experimental observation, and magnetic alloys that undergo cubic-to-monoclinic (II) transformation have multiple solutions satisfying the high-reversibility condition and are suitable candidates for alloy development. Broadly, the results could guide the design of magnetic materials with low fatigue and hysteresis for energy applications.

At present, the design of soft magnetic materials focuses on reducing only the anisotropy constant to zero. While this approach has led to the development of technologically important magnets, such as Sendust and Permalloy, this material development strategy is limited to a small parameter space. Consequently, material compositions that lie beyond the $\kappa_1 = 0$ region with large magnetostriction constants (that we hypothesize to play a key role in reducing hysteresis) are unknown. Using the mathematical criterion between material constants (e.g., the parabolic relation $\kappa_1 \propto \lambda_{100}^2$), we can guide a systematic synthesis of soft magnetic materials, which, despite their large anisotropy constants (with suitable magnetostriction constants), will have small hysteresis.[2, 52] These efforts will not

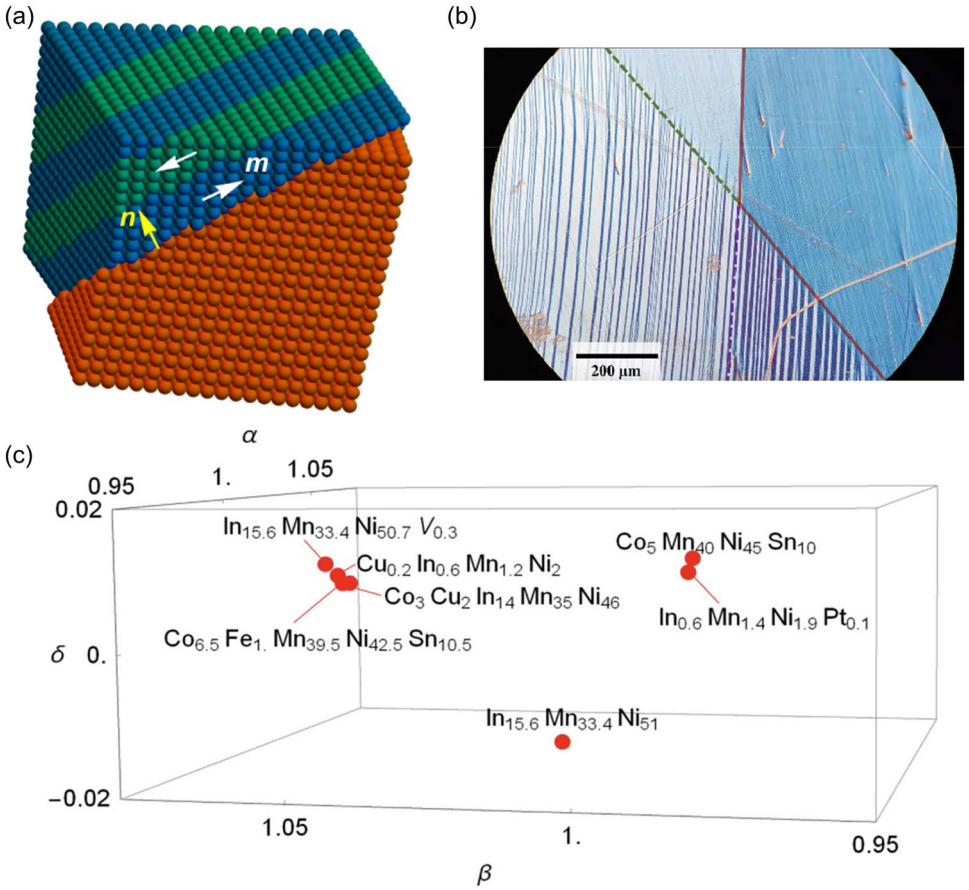

Figure 8. (a) Microstructures in magnetic materials not only form to minimize lattice misfit strains between paramagnetic and ferromagnetic phases but also need to satisfy magnetization compatibility.[52] (b) Magnetic laminate domains are commonly observed to form at the phase boundary during paramagnetic to ferromagnetic phase transformation.[55] Subfigure (b) is adapted from Ref. 55, Copyright 2024, with permission from Elsevier. (c) Several magnetocaloric alloys that satisfy the high-reversibility $\lambda_2 = 1$ compatibility condition often undergo cubic-to-monoclinic transformation.[56] Subfigures (a, c) are adapted from Ref. 52, Copyright 2023 by the American Physical Society.

only enable us to discover a new material but also establish a theory-guided strategy to discover a new generation of magnets with small hysteresis.

Several questions, however, remain unresolved in this effort. For example, do the parabolic relations between magnetocrystalline anisotropy and magnetostriction $\kappa_1 \propto \lambda_{100}^2$ depend on the nature of the localized disturbance? Do the differences in the microstructural growth of needle domains observed at different defect sites, e.g., at the pore defect, grain boundary, or surface/edges of a magnetic particle, affect the energy barrier calculations and thereby influence hysteresis loops? Our initial analysis shows that defect density and defect geometries can affect the coercive force for a given magnetic material; however, this effect is small when compared to the role of fundamental magnetic constants.[46] Future work would require rigorous and in situ characterization of these localized disturbances and contribute to generalizing the coercivity tool for all defect geometries and types.

Acknowledgments

I gratefully acknowledge research funding by the Air Force Fiscal Year 2023 Young Investigator Research Program, United States, under Grant No. FOA-AFRL-AFOSR-2022-0005, and by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, under Award DE-SC0024227. I would like to thank Hongyi Guan and Carlos Garcia-Cervera from the University of California, Santa Barbara, and Richard James from the University of Minnesota for their wonderful discussions on this topic.

Author contribution

A. Renuka Balakrishna drafted the article.

Funding

A. Renuka Balakrishna gratefully acknowledges research funding by the Air Force Fiscal Year 2023 Young Investigator Research Program, United States, under Grant No.

FOA-AFRL-AFOSR-2022-0005, and by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, under Award DE-SC0024227.

Data availability

Not applicable.

Declarations

Conflict of interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

Open Access

References

1. Z. Zhang, R.D. James, S. Müller, Energy barriers and hysteresis in martensitic phase transformations. Acta Mater. **57**(15), 4332–4352 (2009)
2. A.R. Balakrishna, R.D. James, Design of soft magnetic materials. npj Comput. Mater. **8**(1), 1–10 (2022)
3. A. Fert, Nobel lecture: origin, development, and future of spintronics. Rev. Mod. Phys. **80**(4), 1517 (2008)
4. J.M. Silveyra, E. Ferrara, D.L. Huber, T.C. Monson, Soft magnetic materials for a sustainable and electrified world. Science **362**(6413), eaao0195 (2018)
5. W.F. Brown Jr., Micromagnetics, domains, and resonance. J. Appl. Phys. **30**(4), S62–S69 (1959)
6. L. Néel, Some unresolved problems in magnetism. IEEE Trans. Magn. **17**(6), 2516–2519 (1981)
7. R.D. James, Hysteresis in phase transformations. *IMA Preprint Series* (1995)
8. M. Kersten, Grundlagen theorie der ferromagnetischer hysterese und der koerzitivkraft (1943)
9. R. Becker, W. Doring, *Ferromagnetismus* (Springer, Berlin, 1939)
10. C. Kittel, J.K. Galt, Ferromagnetic domain theory. in *Solid State Physics*, vol. 3 (Elsevier, Amsterdam, 1956), pp. 437–564
11. L. Han, F. Maccari, I. Soldatov, N.J. Peter, I.R. Souza, R.S. Filho, O. Gutfleisch, Z. Li, D. Raabe, Strong and ductile high temperature soft magnets through widmanstätten precipitates. Nat. Commun. **14**(1), 8176 (2023)
12. R.M. Bozorth, The permalloy problem. Rev. Mod. Phys. **25**(1), 42 (1953)
13. R.M. Bozorth, *Ferromagnetism* (Springer, Berlin, 1993)
14. B. Lewis, The permalloy problem and anisotropy in nickeliron magnetic films. Br. J. Appl. Phys. **15**(5), 531 (1964)
15. R. Becker, W. Döring, *Ferromagnetismus* (Springer, New York, 2013)
16. R.C. Hall, Single crystal anisotropy and magnetostriction constants of several ferromagnetic materials including alloys of NiFe, SiFe, AlFe, CoNi, and CoFe. J. Appl. Phys. **30**(6), 816–819 (1959)
17. J.B. Restorff, M. Wun-Fogle, K.B. Hathaway, A.E. Clark, T.A. Lograsso, G. Petculescu, Tetragonal magnetostriction and magnetoelastic coupling in Fe – Al, *Fe – Ga*, Fe – Ge, Fe – Si, *Fe – Ga – Al*, and Fe – Ga – Ge alloys. J. Appl. Phys. **111**(2), 07A921 (2012)
18. A.R. Balakrishna, R.D. James, A solution to the permalloy problem-a micromagnetic analysis with magnetostriction. Appl. Phys. Lett. **118**(21), 212404 (2021)
19. S. Yang, H. Bao, Yu. Chao Zhou, X.R. Wang, Y. Matsushita, Y. Katsuya, M. Tanaka, K. Kobayashi, X. Song et al., Large magnetostriction from morphotropic phase boundary in ferromagnets. Phys. Rev. Lett. **104**(19), 197201 (2010)
20. H. Zhao, Y. Ji, T. Ma, M. Fang, Y. Hao, T. Yang, C. Zhou, S. Yang, X. Ren, Exceptional combination of large magnetostriction, low hysteresis and wide working temperature range in $(1 - -x)TbFe_{(2--x)}DyCo_2$ alloys. Acta Mater. **220**, 117308 (2021)
21. Y.X. Xu, Y.Y. Wu, C.C. Hu, T.T. Cai, Z. Zhang, H.H. Huang, P. Fu, H.B. Huang, L.Q. Chen, Domain engineering in ferromagnetic morphotropic phase boundary with enhanced and non-hysteretic magnetostriction: a phase-field simulation. Scripta Mater. **242**, 115916 (2024)
22. M.E. McHenry, M.A. Willard, D.E. Laughlin, Amorphous and nanocrystalline materials for applications as soft magnets. Prog. Mater Sci. **44**(4), 291–433 (1999)
23. X. Tong, Y. Zhang, Y. Wang, X. Liang, K. Zhang, F. Zhang, Y. Cai, H. Ke, G. Wang, J. Shen et al., Structural origin of magnetic softening in a fe-based amorphous alloy upon annealing. J. Mater. Sci. Technol. **96**, 233–240 (2022)
24. F. Unai Urdiroz, A. Javier Palomares, I.V.S. Mayoral, R. Schäfer, J.M. Gonzalez, M. Sanchez-Agudo, A. Enrique Navarro, M.A. Ruiz et al., Role of the interfaces in the crystallization and hysteresis mechanisms of amorphous fe-b thin films. J. Alloy Compd. **869**, 159276 (2021)
25. W.F. Brwon Jr., Theory of magnetoelastic effects in ferromagnetism. J. Appl. Phys. **36**(3), 994–1000 (1965)
26. C. Xie, C.J. García-Cervera, C. Wang, Z. Zhou, J. Chen, Second-order semi-implicit projection methods for micromagnetics simulations. J. Comput. Phys. **404**, 109104 (2020)
27. A. Kovacs, L. Exl, A. Kornell, J. Fischbacher, M. Hovorka, M. Gusenbauer, L. Breth, H. Oezelt, D. Praetorius, D. Suess et al., Magnetostatics and micromagnetics with physics informed neural networks. J. Magn. Magn. Mater. **548**, 168951 (2022)
28. A. De Simone, Energy minimizers for large ferromagnetic bodies. Arch. Ration. Mech. Anal. **125**, 99–143 (1993)
29. C.J. García Cervera, Numerical micromagnetics: a review. SeMA J. **39**, 103–136 (2007)
30. A. DeSimone, R.D. James, A constrained theory of magnetoelasticity. J. Mech. Phys. Solids **50**(2), 283–320 (2002)
31. A. Kovacs, L. Exl, A. Kornell, J. Fischbacher, M. Hovorka, M. Gusenbauer, L. Breth, H. Oezelt, M. Yano, N. Sakuma et al., Image-based prediction and optimization of hysteresis properties of nanocrystalline permanent magnets using deep learning. J. Magn. Magn. Mater. **596**, 171937 (2024)
32. Y. Wang, Y. Tian, T. Kirk, O. Laris, J.H. Ross Jr, R.D. Noebe, V. Keylin, R. Arróyave, Accelerated design of Fe-based soft magnetic materials using machine learning and stochastic optimization. Acta Mater. **194**, 144–155 (2020)
33. N. Vuokila, C. Cunning, J. Zhang, N. Akel, A. Khan, D.A. Lowther, The application of neural networks to the modeling of magnetic hysteresis. IEEE Trans. Magn. (2023)
34. A.L. Foggiatto, S. Kunii, C. Mitsumata, M. Kotsugi, Feature extended energy landscape model for interpreting coercivity mechanism. Commun. Phys. **5**(1), 277 (2022)
35. E.C. Stoner, E.P. Wohlfarth, A mechanism of magnetic hysteresis in heterogeneous alloys. Philos. Trans. R. Soc. Lond. Ser. A **240**(826), 599–642 (1948)
36. H. Knüpfer, R.V. Kohn, F. Otto, Nucleation barriers for the cubic-to-tetragonal phase transformation. Commun. Pure Appl. Math. **66**(6), 867–904 (2013)

37. X. Chen, V. Srivastava, V. Dabade, R.D. James, Study of the cofactor conditions: conditions of supercompatibility between phases. J. Mech. Phys. Solids **61**(12), 2566–2587 (2013)
38. S. Tan, X. Huibin, Observations on a CuAlNi single crystal. Continuum Mech. Thermodyn. **2**, 241–244 (1990)
39. J. Cui, Y.S. Chu, O.O. Famodu, Y. Furuya, J. Hattrick-Simpers, R.D. James, A. Ludwig, S. Thienhaus, M. Wuttig, Z. Zhang et al., Combinatorial search of thermoelastic shape-memory alloys with extremely small hysteresis width. Nat. Mater. **5**(4), 286–290 (2006)
40. C. Chluba, W. Ge, R. Lima de Miranda, J. Strobel, L. Kienle, E. Quandt, M. Wuttig, Ultralow-fatigue shape memory alloy films. Science **348**(6238), 1004–1007 (2015)
41. Y. Song, X. Chen, V. Dabade, T.W. Shield, R.D. James, Enhanced reversibility and unusual microstructure of a phase-transforming material. Nature **502**(7469), 85–88 (2013)
42. C. Zhang, Z. Zeng, Z. Zhu, N. Tamura, X. Chen, Energy conversion from heat to electricity by highly reversible phase-transforming ferroelectrics. Phys. Rev. Appl. **16**(2), 024064 (2021)
43. M. Wegner, G. Hanlin, R.D. James, E. Quandt, Correlation between phase compatibility and efficient energy conversion in Zr-doped barium titanate. Sci. Rep. **10**(1), 3496 (2020)
44. V. Srivastava, X. Chen, R.D. James, Hysteresis and unusual magnetic properties in the singular heusler alloy $Ni_{45}Co_5Mn_{40}Sn_{10}$. Appl. Phys. Lett. **97**(1), 014101 (2010)
45. Y.G. Liang, S. Lee, H.S. Yu, H.R. Zhang, Y.J. Liang, P.Y. Zavalij, X. Chen, R.D. James, L.A. Bendersky, A.V. Davydov et al., Tuning the hysteresis of a metal-insulator transition via lattice compatibility. Nat. Commun. **11**(1), 3539 (2020)
46. A.R. Balakrishna, R.D. James, A tool to predict coercivity in magnetic materials. Acta Mater. **208**, 116697 (2021)
47. N. Kurti, *Selected Works of Louis Neel* (CRC Press, Boca Raton, 1988)
48. N. Pilet, The relation between magnetic hysteresis and the micromagnetic state explored by quantitative magnetic force microscopy. PhD thesis, University of Basel (2006)
49. H.J. Williams, Direction of domain magnetization in powder patterns. Phys. Rev. **71**(9), 646 (1947)
50. A. Hubert, R. Schäfer, *Magnetic Domains: The Analysis of Magnetic Microstructures* (Springer, New York, 2008)
51. J.M.D. Yangkun He, R.S. Coey, C. Jiang, Determination of bulk domain structure and magnetization processes in BCC ferromagnetic alloys: Analysis of magnetostriction in $Fe_{83}Ga_{17}$. Phys. Rev. Mater. **2**(1), 014412 (2018)
52. A.R. Balakrishna, Compatible microstructures in magnetic materials. Phys. Rev. Mater. **6**(7), 074402 (2022)
53. G. Herzer, Anisotropies in soft magnetic nanocrystalline alloys. J. Magn. Magn. Mater. **294**(2), 99–106 (2005)
54. H. Guan, N. Ahani, A.R. Balakrishna, Magnetoelastic interactions reduce hysteresis in soft magnets. Manuscript under review (2024)
55. C. Zhang, X. Balandraud, Y. He, Coexistence of five domains at single propagating interface in single-crystal NiMnGa shape memory alloy. J. Mech. Phys. Solids **183**, 105481 (2024)
56. A.R. Balakrishna, W.C. Carter, Combining phase-field crystal methods with a Cahn-Hilliard model for binary alloys. Phys. Rev. E **97**(4), 043304 (2018)