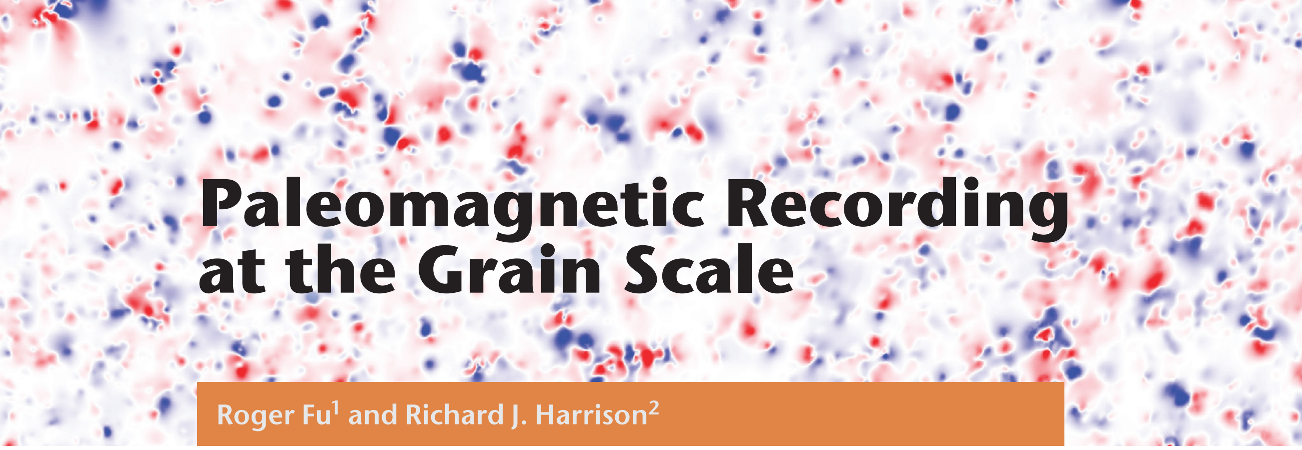

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Paleomagnetic Recording at the Grain Scale

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Natural rocks harbor diverse assemblages of magnetic mineral grains that record information about past dynamo activity and plate motions, among other processes. For much of its history, however, the field of paleomagnetism has counted on a thorough theoretical understanding of only very fine (≤100 nm) grains magnetized during heating. Here we review experimental and computational advances to move beyond this limitation. Magnetic field microscopy allows us to physically identify mineral grains carrying specific paleomagnetic signals, while nanotomography coupled with micromagnetic simulations offers, for the first time, a quantitative picture of how most naturally occurring magnetic grains behave across geologic time. Together, these techniques open the door to retrieving records from less-than-ideal rocks with complex geological histories.

KEYWORDS: magnetism; minerals; geophysics; microscopy

FROM MINERAL GRAINS TO ANCIENT MAGNETIC FIELDS

We are surrounded by constantly changing magnetic fields. Light itself arises from magnetic fields oscillating at hundreds of terahertz. The Earth is immersed in fluctuating solar wind fields. Even the geomagnetic field, steady enough to guide our compasses, wobbles perceptibly over the course of years and reverses polarity completely over geologic time. This is no surprise: a planet's magnetic field is, after all, anchored to a massive sphere of churning metallic fluid deep inside its core.

It is even more amazing to realize that it is possible to reconstruct the magnetic field of the ancient Earth, and beyond, as it existed millions to billions of years ago. The discipline of paleomagnetism uses permanent magnetization carried by ensembles of natural mineral grains to retrieve the strength and direction of past magnetic fields. Not only has this information revealed the behavior of planetary dynamos through time but also, due to the latitudinal dependence on the inclination (angle to horizontal) of the geomagnetic field, the motion of tectonic plates throughout Earth history (Torsvik et al. 2012; Davies and Constable 2014).

Paleomagnetism works because, unlike the ephemeral, large-scale magnetic fields surrounding us, magnetic fields within **microscopic*** mineral grains can remain incredibly stable even over billions of years. If an ancient magnetic field can, under specific conditions, reset the magnetization of magnetic grains, these minerals would encode information about that field long into the future—a permanent record of a fleeting phenomenon.

The pioneering work of Néel in the late 1940s provided a quantitative, physical framework to understand the magnetic recording behavior of small (<100 nm in the case of magnetite), uniformly magnetized "single-domain" (SD) grains (FIG. 1A). SD grains behave in simple and well-understood ways: they adopt one of two or more stable magnetization states with directions defined by crystallography or grain shape. To transition among these states, the grain must overcome an energy barrier via either thermal excitation or applied magnetic fields. If this barrier is much higher than the available thermal or magnetic energy, no transitions occur and the magnetization remains stable over geological time scales (i.e., "blocked"). This unchanging signal is referred to as the "remanent" magnetization.

The height of the energy barrier for SD grains is easy to calculate: it is the product of particle volume (V) and the magnetic anisotropy constant (K). The latter is determined by the mineral type and the grain shape (Harrison and Feinberg 2009). For a grain of a given mineral with a given shape, the transition from unblocked to blocked can be driven either by increasing V or decreasing temperature—both occur in nature, leading to the acquisition of chemical remanent magnetization (CRM) or thermoremanent magnetization (TRM), respectively. Néel's SD theory has been at the heart of paleomagnetism since its emergence in the 1950s and 60s, guiding virtually all methods to measure, isolate, quantify, and interpret the ancient magnetic signals of rocks and meteorites to this day.

In the decades since this first, reassuring theory, it has become increasingly clear that, because most natural rocks are not dominated by SD grains, our understanding needs to be extended to less ideal cases. Rocks that have stayed hundreds of degrees below their Curie temperatures—the temperature necessary for most rocks to completely reset all grains according to single domain theory—have been shown to carry magnetic directions millions of years younger than expected. Explaining these observations is

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* Definitions of terms presented in **blue** are given in the glossary on page 81.

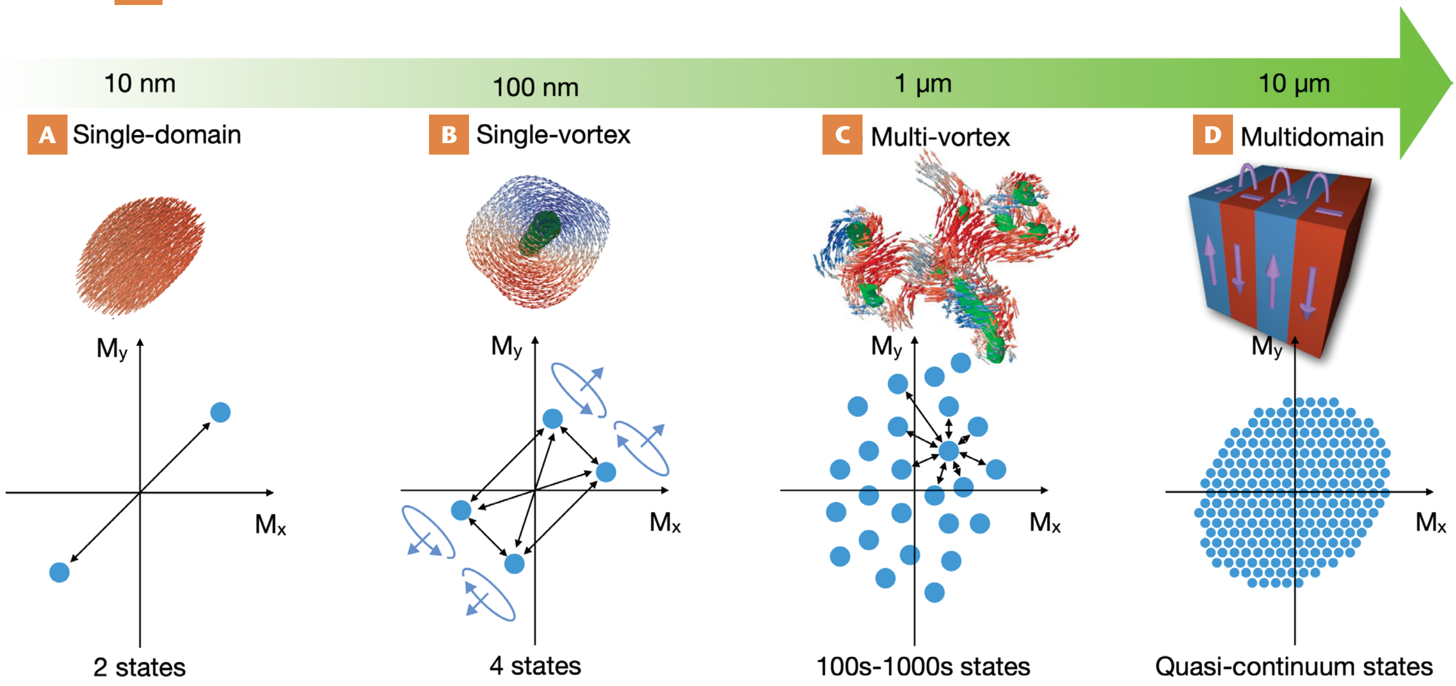


FIGURE 1 (**A–D**) The evolving magnetic structure of natural remanence carriers as a function of increasing grain size assuming a magnetite composition (not to scale). Micromagnetic modelling results in (**A**) and (**B**) from Nikolaisen et al. (2020). Micromagnetic image in (**C**) from Lascu et al. (2018). Blue dots represent the direction and magnitude of stable magnetization states that each grain can adopt. Black arrows represent thermally or applied field activated transitions that can take place between states. Green cylinders represent the core of magnetic vortices.

critical to the credibility of paleomagnetic data and all the tectonic motion, geodynamo behavior, and other implications they hold.

HOW DOES PALEOMAGNETIC RECORDING FAIL?

In broad terms, a paleomagnetic record can be said to "fail" in two ways: remagnetization and non-ideal behavior. During remagnetization, an ancient magnetization is removed and, in more pernicious cases, completely replaced by a much younger magnetization. Total remagnetization has led to dramatic revisions of results. In an extreme example, a paleolatitude of South African rocks originally attributed to 3.4 billion years ago (Ga) was recognized as a likely modern-day (≤700 ka) overprint after more extensive study of surrounding rock units (Biggin et al. 2011).

How does remagnetization occur? In the simplest case, some grains can just "forget" their original magnetizations, even at close to ambient temperatures. Grains larger than a certain, mineral-dependent size threshold (approximately 100 nm in magnetite) no longer behave as uniformly magnetized compass needles. Grains within about 10 times the SD threshold exhibit a rich variety of vortex states with surprising stability (see below). However, the "forgetfulness" of grains rapidly increases past this point. Néel originally described the formation of multidomain (MD) grains in which neighboring volumes of reverse magnetization polarity, or domains, exist in the same grain (FIG. 1D). The energy barriers to shift domain boundaries are generally much lower than those needed to flip SD grains. Therefore, MD grains can have substantial fractions of their magnetizations reset even if the rock is never heated close to the Curie temperature (FIG. 2). This phenomenon has been widely replicated in laboratory experiments (e.g., Shcherbakova et al. 2000).

So far, we have described how existing grains remagnetize over time. A potentially more confounding process is chemical remagnetization, where newly crystallized grains either replace or simply overwhelm a rock's original carriers (FIG. 2). These new grains will typically record the magnetic field during their crystallization and growth, which routinely post-dates original rock formation by millions to billions of years. Chemical remagnetization as a process has been recognized for many decades, famously accounting for puzzlingly uniform magnetization in limestones across vast tracts of the Appalachians (McCabe and Elmore 1989). In general, the pervasive occurrence of chemical remagnetization has become better recognized over time, with large volumes of the seafloor, meteorites, and Archean greenstones exemplifying rock types once thought to carry pristine thermoremanence now reinterpreted to carry chemical remanence (Raymond and LaBrecque 1987; Fu et al. 2014; Brenner et al. 2024).

Finally, we come to "failure" through non-ideal behavior. Even if an ancient magnetization survives unscathed to the present day, can we draw reliable conclusions from it? Paleomagnetic directions are relatively robust in this regard: as long as some grains retain original magnetization, a direction may be recoverable. Studies of the intensity of ancient fields, or paleointensities, are not so lucky. First, if the ancient magnetization is not thermal in origin, its intensity cannot be determined precisely using well-established techniques. Even if a thermoremanence survives, decay in its intensity due to MD grain unpinning or chemical addition of secondary minerals with similar unblocking temperatures (FIG. 2) would bias the inferred paleointensity, usually to lower values. Finally, laboratory alteration can change the grain population carrying the laboratory magnetization used to compute a paleointensity, resulting in potentially severe biases. In these ways, even well-preserved, ancient magnetizations can mislead if they are not carried by a well-identified mineral population with known origin and stability to alteration.

Motivated by these facts, much effort in recent years has focused on the following questions: How can we recognize remagnetized and non-ideal magnetic grains in rocks? And, if present, what is the expected long-term behavior of non-ideal (i.e., non-single domain) grains anyway?

NEW WAYS TO PROBE PALEOMAGNETIC RECORDING AT THE GRAIN SCALE

A key take-away from the complexities discussed above is that natural rocks often host multiple populations of magnetic minerals, each of which may have a different age and different magnetic recording fidelity. The central questions become: which of these population(s) is responsible for carrying observed magnetizations and what does that imply for paleofield direction, intensity, and age?

Although traditional paleomagnetic field tests (i.e., conglomerate, reversal, baked contact, fold) remain powerful and irreplaceable tools for determining the age of magnetization, a field test of appropriate age is often not available. To answer the above questions using a rock's magnetic properties alone, we must overcome two challenges. First, we must spatially resolve and quantify separately the paleomagnetic signals originating from different grain populations within a bulk rock (Fig. 2). Second, we must determine the precise nature of the underlying grain ensemble (i.e., the mineralogical identity, size, shape, position, and orientation of each grain) and on that basis determine their ability to faithfully record paleomagnetic information. Major advances in high-resolution magnetic imaging, nanotomography, and micromagnetic modelling have made substantial progress towards these goals. These three advances are reviewed in turn below.

Magnetic Imaging

Paleomagnetic studies have routinely used optical and electron microscopy to identify ferromagnetic mineral phases; however, these techniques may overlook large portions of grains in the critical 10–1000 nm size range. A more fundamental problem is that the mere detection of remanence carriers cannot be quantitatively related to the bulk magnetic signal. An alternative strategy is to perform detailed rock magnetic characterization on the bulk sample, which can distinguish magnetic mineral populations based on subtle differences in their response to laboratory magnetic fields or temperature. However, such methods miss the spatial context and can be confounded by similar rock magnetic behavior of grain populations that formed in different environments and ages.

Magnetic field microscopy is, in essence, a way to combine these approaches by retrieving spatially resolved magnetic information. The past 15 years have seen rapid growth in the tools available for imaging rock magnetism. Most recently, the quantum diamond microscope (QDM; Box 1) uses a synthetic nitrogen-vacancy doped diamond chip to achieve ~1-µm resolution over a several mm^2 area (Glenn et al. 2017; Fu et al. 2020). In terms of sensitivity, the QDM can detect magnetizations of order 10^{-17} Am^2, corresponding to ~40-nm diameter magnetite grains, therefore providing coverage over some of the SD grain size range and all larger grain types. The detection threshold for magnetization falls off rapidly with depth of the source within the sample due to the $\propto d^{-3}$ relationship between magnetic field and distance (d) to a dipolar source. For example, at 50 µm depth, the detection threshold is close to 10^{-13} Am^2.

How can one associate magnetic microscopy signals, spread across potentially millions of pixels, with a paleomagnetic record, i.e., the net magnetization of a specific grain ensemble? The most straightforward answer is the direct computation of net magnetization from image data. This essentially turns a microscopic ensemble of magnetic minerals into a miniature traditional paleomagnetic core sample. Doing this in practice requires either naturally spatially well-separated grain populations or mechanical isolation of the desired materials. An early application of this concept used the SQUID microscope, a cryogenic instrument with ~200-µm spatial resolution, to measure physically extracted, fine Fe grain–bearing chondrules from a meteorite (Fu et al. 2014a; Fig. 3 rightmost column). More recent use of the QDM on another meteorite managed to retrieve in-situ demagnetization data (Fu et al. 2021). Methods to apply magnetic moment inversions to a large number of resolved grains, potentially combined with 3D

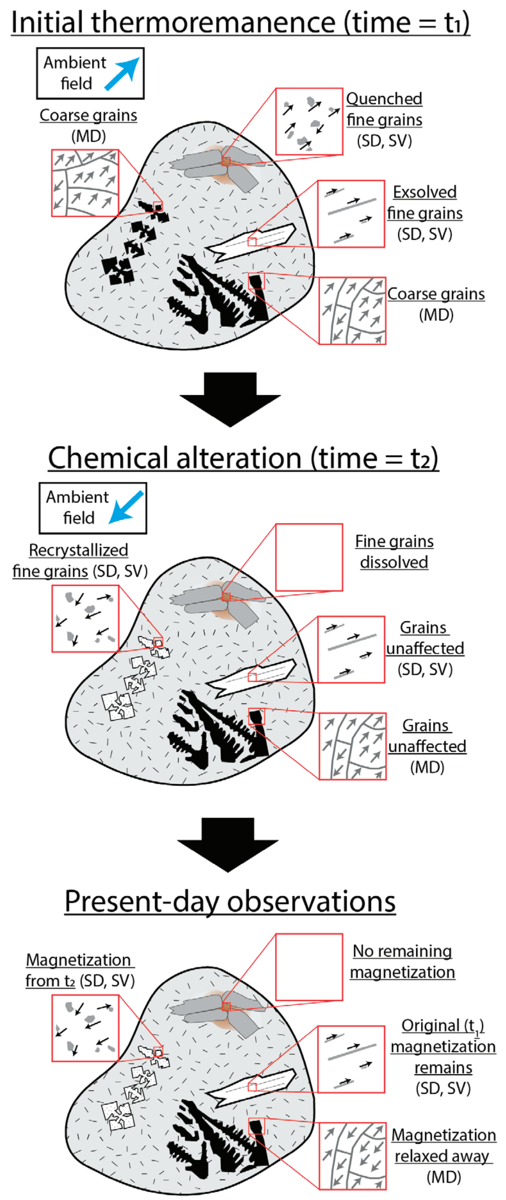
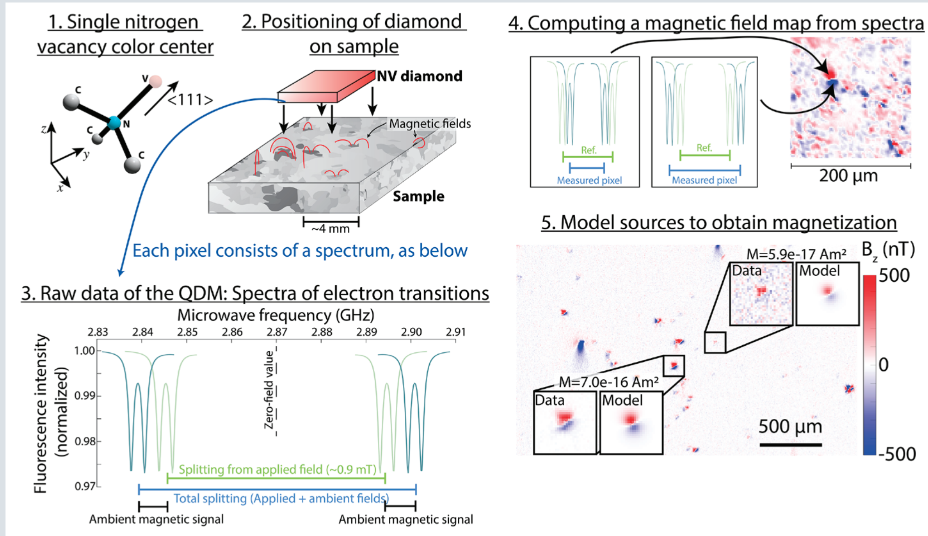


Figure 2 Concept summary of possible fates of magnetizations in complex ancient rocks. Although traditional paleomagnetism has relied on thermoremanent magnetizations, aqueous alteration events can dissolve away and/or precipitate new grains, potentially introducing younger magnetizations. Simultaneously, multi-domain magnetizations may not be stable through geologic time. These grain populations can be localized using magnetic microscopy (Fig. 3). Degree of alignment among grains and domains have been greatly exaggerated: in nature, typically in a single domain or single vortex dominated sample, only <1 to a few percent of grains are biased in the direction of the recorded magnetic field.

Box 1 **HOW DOES THE QUANTUM DIAMOND MICROSCOPE WORK?**

The quantum diamond microscope (QDM) senses magnetic fields using the Zeeman effect, in which the energies of electrons with plus and minus spin states are split (i.e. separated) upon application of a magnetic field. This effect applies to atoms in all materials; what sets the QDM apart is that the Zeeman splitting in nitrogen-vacancy (NV) centers, which occur when a pair of carbon atoms in a diamond lattice are replaced by a nitrogen atom and a vacancy, can be detected optically. Specifically, NV centers reemit light when excited by a higher-energy light source, and the intensity of this reemitted, or fluoresced, light decreases subtly when many electrons reside in specific energy states, denoted |±1⟩. By sweeping across a range of microwave frequencies around 2.9 GHz, we pump electrons into states corresponding to these frequencies. When this frequency resonates with a |±1⟩ state, electrons populate this energy level and the overall fluorescence intensity decreases, typically by a few percent. By plotting the location of resonances as a function of microwave frequency, we can compute the energy level splitting due to the Zeeman effect, resulting in a magnetic field measurement. The result of taking these spectra over a diamond surface is a several-millimeter-sized image with optical (~1 µm) resolution documenting the magnetic field at each pixel. Vector measurements are possible due to the existence of four different crystallographic axes in diamond. Measurements are typically taken while applying a moderate (~0.9 mT) magnetic field. This field is reversed during measurement such that its effects can be removed, resulting in a small residual bias field of several 100 nT. The resulting magnetic field map can be interpreted qualitatively by overlaying with another image (e.g., composition) or quantitatively to obtain the magnetization vectors of single grains or ensembles.



tomographic information, are currently being developed (De Groot et al. 2021). The number of classic SD grains necessary to faithfully record a paleomagnetic field (e.g. with 1 ° accuracy) was thought to be in the millions; however, more recent simulations with more realistic single vortex (see below) states suggest that as few as several thousand may be sufficient (Bellon et al. 2025). In this case, paleomagnetic direction and intensity recovery from grain-resolving maps represents a highly promising direction for studies of complex rocks where it is advantageous to include signal from only a subset of present grains to avoid the effects of, for example, sample alteration and multi-domain recorders.

Currently, however, no technique can reliably quantify net magnetizations in all natural samples based only on magnetic field maps due to the complexity of magnetic field signals from closely spaced grains. For such cases, one simple solution is to observe changes in the sample magnetic field pattern during a demagnetization sequence, thereby quantifying the coercivity or unblocking temperature spectra of each spatially resolved grain population (Volk et al. 2022). Although this analysis does not quantify magnetization direction or intensity, close similarity between a grain population's demagnetization spectrum and the behavior of a certain magnetization component in the bulk sample can associate a paleomagnetic signal with a certain grain population. As an example, this method has been used to show that chemically precipitated fine-grained magnetite is the main carrier in a suite of Archean basalts, demonstrating that the magnetization has a seafloor hydrothermal origin instead of a primary igneous origin (Brenner et al. 2022).

Nanotomography

Having used magnetic microscopy either to directly measure a paleomagnetic signal from a small volume or to associate a bulk magnetization with a certain grain population, nanotomography is the critical next step for characterizing the underlying ensemble of magnetic particles (Fig. 3). Understanding a grain's response to magnetic fields requires resolving the three-dimensional (3D) morphology of grains with resolution of the order ~10 nm. This task is now achievable using either focused-ion beam nanotomography (FIB-nt) or high-resolution, synchrotron-based X-ray tomography methods such as X-ray holotomography and ptychotomography.

Focusing on the FIB-nt technique as an illustration, a FIB instrument combines a scanning electron microscope (SEM) system for high-resolution imaging and a highly focused ion beam (usually Ga ions) for precision milling at an angle of around 50 ° to the electron beam. Deep trenches are milled to the side and in front of the region of interest, exposing the front face (Fig. 4C), which is then imaged using the electron beam. By sequentially milling away 10–30 nm slices from the front face using the Ga ion beam, an automated sequence of many 100s–1000s of backscattered electron (BSE) images is acquired, aligned, stacked and interpolated to reconstruct the volume of interest with a voxel size on the order of a few 10s nm (Fig. 4D). Following segmentation of this data to isolate the bright, dense magnetic minerals from the darker silicate matrix, a 3D model of the remanence carrying ensemble is produced (Fig. 4F).

Micromagnetic Simulations

A step-change in our understanding of magnetic recording has arisen through the development and widespread adoption of powerful-yet-easy-to-use finite-element micromagnetic modelling codes. The powerful combination of nanotomography and micromagnetic modelling means it is—in principle—possible to model all conceivable aspects of the paleomagnetic recording process. In practice, however, computational limitations remain a significant barrier to be overcome.

In essence, a micromagnetic modelling code such as MERRILL (Micromagnetic Earth Related Robust Interpreted Language Laboratory; Ó Conbhuí et al. 2018an open source software package for three-dimensional micromagnetics

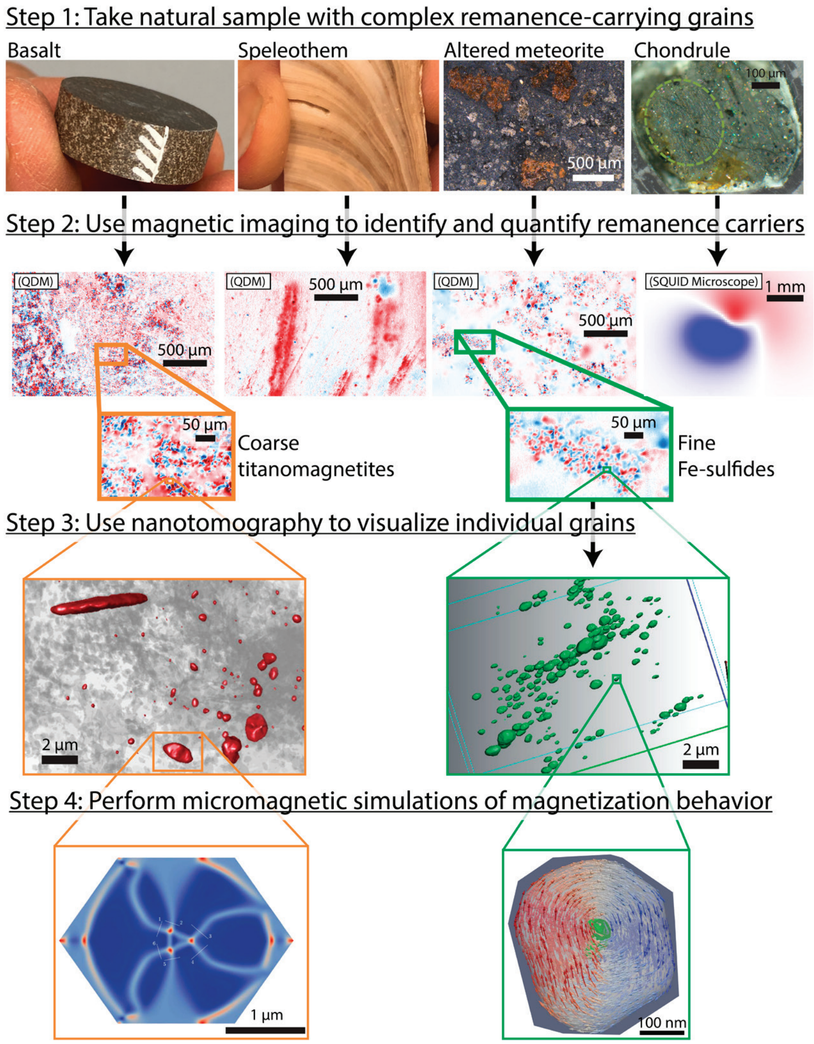


FIGURE 3 Conceptual workflow of using magnetic microscopy to isolate and identify grain populations in complex rocks followed by nanotomography to visualize individual grains and micromagnetic simulations to quantify their paleomagnetic recording potential. Magnetic field maps in step 2 correspond to actual samples shown in step 1 while images in steps 3 and 4 represent similar grain types but are not from the same studies. Chondrule data from Fu et al. (2014a) and Einsle et al. (2016); coarse grain simulations from Nagy et al. (2019) and Donardelli Bellon et al. (2024).

optimized and designed for the calculation of such complex structures. MERRILL has a simple scripting user interface that requires little computational knowledge to use but provides research strength algorithms to model complex, inhomogeneous domain structures in magnetic materials. It uses a finite element/boundary element numerical method, optimally suited for calculating magnetization structures of local energy minima (LEM) computes and minimizes the total magnetic energy of a grain, which is expressed as a function of its internal magnetization state and is the sum of four contributions (see boxes in Harrison and Feinberg 2009):

1. The **exchange energy** derives from spin-spin interactions at the atomic scale. It drives magnetic ordering and can be minimized by keeping neighboring spins either parallel or antiparallel to each other depending on the type of magnetic order.

2. The **anisotropy energy** derives from the spin-orbit coupling at the atomic scale; it is minimized by keeping the magnetization oriented along one of the crystallographically defined easy axes.

3. The **demagnetizing energy** derives by long-range dipole-dipole interactions between spins; it is minimized by the formation of magnetic domains.

4. The **Zeeman energy** represents the effect of an external or neighboring grain fields; it is minimized by aligning the magnetization of the grain parallel to the total field.

Micromagnetic models are a **continuum approximation** that express the total magnetic energy of a grain in terms of its magnetization, represented by a continuous magnetization vector field of fixed amplitude (equal to the saturation magnetization of the material) and spatially varying orientation. A key tenet of micromagnetic theory is that no change in magnetization orientation takes place below a characteristic length scale known as the "exchange length" (l_{ex}, 9.5 nm for magnetite, the most common remanence carrier in Earth rocks and 3.3 nm for kamacite, a metallic Fe phase). On smaller length scales, the exchange interactions dominate and keep neighboring spins strictly aligned with each other. Given the uniformity of magnetization below l_{ex}, it is possible to discretize the grain into a grid or mesh of points with spacing $\leq l_{ex}$. Typically a tetrahedral volume mesh of points is used with the maximum tetrahedral edge length set to the exchange length or finer. Each point in the mesh is assigned a magnetic orientation. The total micromagnetic energy for a given grain and applied field is then minimized using one of a number of algorithms (e.g., conjugate gradient descent). The energy state reached

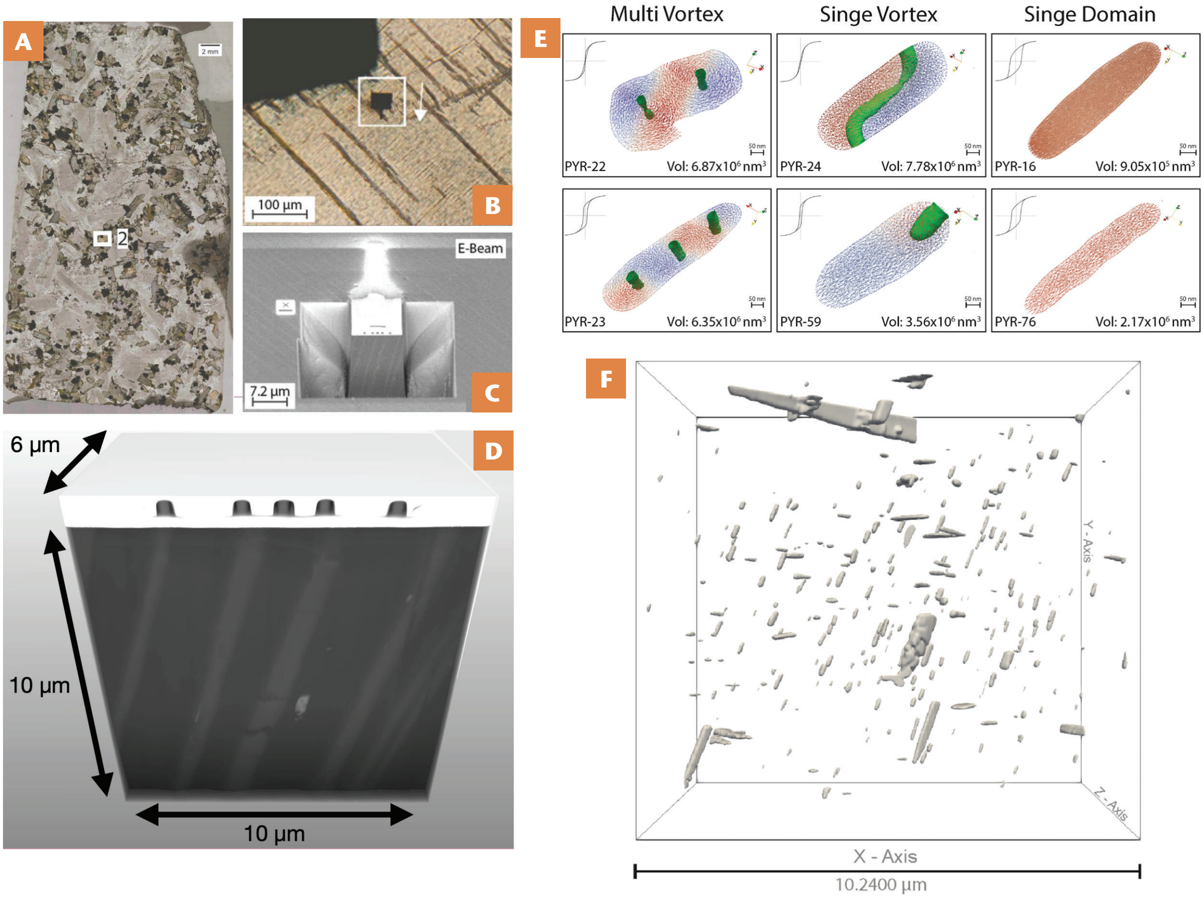


FIGURE 4 Focused-ion beam (FIB) "slice-and-view" nanotomography of a pyroxene grain containing exsolved magnetite needles. MODIFIED FROM NIKOLAISEN ET AL. (2020). (**A**) Thin section image of a gabbroic norite sample. Box marked 2 in (**A**) is the area of panel (**B**). White arrow indicates the direction of FIB sequential milling. (**C**) Electron-beam view of the cross section (front face) of the milling volume. (**D**) Reconstructed 3D volume obtained by stacking and aligning front-face images obtained every 30 nm. Bright feature in the volume is a magnetite particle; dark background is pyroxene. (**E**) Micromagnetic simulations of selected needles, demonstrating SD, SV, and MV states. (**F**) 3D reconstruction of the ensemble of segmented magnetite needles.

at the end of the simulation depends critically on the initial configuration of magnetization assumed at the start of the simulation—such is the nature of magnetic hysteresis!

Under this paradigm, micromagnetic modelling has yielded groundbreaking and at times unexpected insights. It is now known that 100–1000 nm grains adopt complex, non-uniform remanent states, collectively known as "vortex" (V) states (FIG. 1B, 1C; Roberts et al. 2017). Grains at the smaller end of this size range adopt "single-vortex" (SV) states (FIG. 1B): a tornado-like pattern of magnetization twisting around a uniformly magnetized core (green cylinder in FIG. 1B). Grains at the larger end of the vortex size range adopt more complex "multi-vortex" (MV) states (FIG. 1C). In general, as the grain becomes larger, both the number of remanence states and the number of energy barriers and transitions increase dramatically. Potentially non-ideal morphologies of large grains further increase the complexity of the energy landscape, resulting in 100s or 1000s of remanence states and millions of energy barriers. Further complicating the picture is the fact that energy barriers between stable states vary greatly in magnitude, resulting in both slow creep of magnetization over time and different subsets of stable states at different temperatures.

Beyond identifying these stable states, recent simulations aim to simulate TRM acquisition by incorporating the nudged elastic band (NEB) method, which efficiently quantifies the effective energy barrier between stable states (Dittrich et al. 2002). By comparing the magnitude of these energy barriers to thermal fluctuations, Nagy et al. (2017) and successive papers were able to compute the expected stability time, i.e., the average time for a particle to spontaneously remagnetize at a given temperature, for common remanence-carrying minerals (FIG. 1E). Happily for paleomagnetism as an endeavor, these studies have found that most grains in vortex states are stable for billions of years, making them as reliable as the traditional "gold standard" SD grains for carrying paleomagnetic signals.

PROSPECTIVE

The experimental and computational techniques reviewed here, although complex, simply represent recent progress towards a longstanding and obvious goal: to assess and to access the trustworthiness of magnetic signals in natural rocks. In contrast to the preceding decades, the last 15 years have seen rapid progress in detecting and quantifying the long-term behavior of non-single domain, non-thermally magnetized grains, which are more pervasive in nature than previously thought (or hoped for).

As examples, QDM and ptychotomography imaging studies have shown that individual, 100 µm-scale zircon grains can contain sufficient independent magnetite inclusions within to record ambient magnetic fields, opening the possibility of using carefully selected ancient zircons to document the history of the geodynamo when traditional whole rock samples are not available (Fu et al. 2017). Similarly, imaging and micromagnetic models of Fe-bearing chondrules from ancient meteorites have permitted the first reliable inferences of magnetic fields in the Solar System's protoplanetary disk (Fu et al. 2014a), which has been followed up by new studies that paint an increasingly complete picture of protoplanetary disk physics (Borlina et al. 2021). Closer to home in both time and space, magnetic imaging of cave deposits has shown promise in discovering episodes of enhanced soil delivery into caves, thereby providing a new means of identifying past extreme precipitation events with application towards quantifying climate hazards (Piascik et al. 2025).

If the ultimate goal, however, is to squeeze every last morsel of paleomagnetic information from all possible rock types (and to not fall for deviously corrupted records), major tasks remain:

1. The QDM is situated in a "sweet spot" of spatial resolution, sensitivity, and field of view; however, modern instruments are still limited to grains stronger than ~1×10^{-17} Am^2, corresponding to a ~35 nm diameter grain of magnetite. Field sensitivity must be improved by ~1 order of magnitude to capture nearly all SD grains and rock types (Fu et al. 2020).
2. Processing of high-resolution magnetic field images needs to be optimized to retrieve net magnetization from complex maps. Techniques using more sophisticated inversions or automated source detection, which promise to rapidly quantify a statistically insightful ensemble of magnetic sources, are currently being refined.
3. Direct experimental verification of micromagnetic models is still lacking. This should entail directly observing the magnetization states of natural grains and their switching behavior coupled with simulations of the exact same grains.
4. Armed with imaging tools to better identify chemical magnetization, the community can rethink its usage in paleomagnetic questions. For example, what do we know about the acquisition timescales of chemical remanence and what are the implications of this for constraining ancient paleosecular variation?

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