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Magnetism in rocks

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Abstract. Rock magnetism is the study of induced and remanent magnetization of ferrimagnetic mineral grains in rocks, sediments, soils, and organisms. Its applications include environmental magnetism, magnetic anisotropy, sources of continental and oceanic magnetic anomalies, records of geomagnetic field variations and polarity reversals, and the paleomagnetic record of plate motions and the Wilson cycle. This paper reviews the beginnings of rock magnetism and then traces the development of six particularly interesting areas: pseudo-single-domain behavior; magnetic domains and micromagnetic structures; diagnostic tests of the type and stability of remanent magnetization; magnetic microanalysis; thermoviscous remagnetization; and chemical remanent magnetization. Other areas, including sediment and soil magnetism, are covered in a companion paper by Verosub and Roberts on environmental magnetism.

Introduction

Magnetism has fascinated mankind since the invention of compasses that could track invisible magnetic field lines over Earth's surface. Much later came the discovery that magnetic grains in rocks act as microscopic compasses that fossilize a record of ancient magnetic fields. Rock magnetism is the study of these magnetic grains, both in the laboratory and in nature.

The earliest compasses were lodestones, naturally occurring ores of magnetite (Fe_3O_4), which rotate in order to align preexisting remanent magnetization $\mathbf{M_r}$ with the geomagnetic field **H**. As well as their irreversible magnetic memory or remanence, ferrimagnetic mineral grains also acquire a reversible induced magnetization **M** in the direction of **H**. (Definitions and abbreviations are summarized in Table 1.)

Both induced and remanent magnetizations originate in magnetic domains, regions of the crystal in which atomic magnetic moments are exchange coupled parallel, creating a powerful spontaneous magnetization $\mathbf{M_s}$. In transition regions between domains, called walls, the atomic moments rotate gradually against exchange forces over several hundred lattice spacings. If each moment in a wall rotates through a small angle in response to **H**, the wall effectively moves, enlarging one set of domains (those in which $\mathbf{M_s}$ is parallel or nearly parallel to **H**) at the expense of less favorably oriented domains. Domain wall motion is the usual mechanism of induced magnetization. Crystal defects, such as dislocations, inhibit the rotation of atomic moments and tend to pin the wall. Wall pinning is one mechanism of remanence.

Because of the mobility of domain walls, the susceptibility k = M/H would be very high if it were not for the internal self-demagnetizing field −NM which opposes **H**. N is called the demagnetizing factor and is determined principally by the shape of the ferrimagnetic grain (for equidimensional grains, N ≈ 1/3). Self-demagnetization is so powerful that k is limited to relatively small values: ≤ 1/N ≈ 3. Because N is almost independent of physical factors like grain size, number of domains, or temperature, the amplitudes of magnetic anomalies due to induced magnetization tend to be determined simply by the composition and amount of ferrimagnetic material a rock contains.

Linear magnetic anomalies recording seafloor spreading have a different origin: natural remanent magnetization (NRM) $\mathbf{M_r}$ dating from the time the oceanic crust formed. Induced magnetization in the present geomagnetic field is also present but the Koenigsberger ratio, Q = M_r/kH, of remanent to induced magnetization tends to be large, particularly for oceanic basalts near spreading centers. The fine grain size and high stress state of titanomagnetites in these rocks, resulting from initial quenching and later surface oxidation, cause pinning of domain walls. The state of magnetization produced by the paleomagnetic field at the time of cooling or shortly thereafter (the primary NRM) is preserved in metastable equilibrium and the response of walls to the present field is muted.

Sufficiently small grains have very few walls, or no walls at all. These latter grains, containing a single magnetic domain, have an intense magnetization equal to the spontaneous magnetization $\mathbf{M_s}$. Since the magnitude of $\mathbf{M_s}$ is fixed and its orientation can only be changed by rotating all atomic magnetic moments against crystalline or magnetostatic coupling forces, single-domain (SD) grains provide the ultimate in strong and stable remanence $\mathbf{M_r}$.

Applications of Rock Magnetism

Pioneering textbooks by *Nagata* [1961], *Irving* [1964], and *McElhinny* [1973] had a formative influence on the generation of geoscientists who came of age during the plate tectonic revolution because they placed rock magnetism in a geological and paleomagnetic context. Today rock magnetism is applied even more widely than those authors could have imagined.

Environmental Magnetism

Magnetic minerals in soils, paleosols and loess record past changes in climate and rainfall, through their composition/oxidation state and susceptibility, and also the timing of climatic episodes, through matching the direction and intensity of $\mathbf{M_r}$ to the geomagnetic paleosecular variation record [*Singer and Fine*, 1989; *Banerjee et al.*, 1993]. Magnetic grains are

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Table 1. Abbreviations, Acronyms, and Definitions

Abbreviation/Acronym	Definition
AF demagnetization	alternating field demagnetization; progressive demagnetization effected by a field of alternating polarity and smoothly decreasing amplitude; the peak amplitude increases with each step of the demagnetization procedure
Blocking temperature T_B	temperature at which the relaxation time for magnetization changes becomes equal to the time of an experiment or of a magnetization process in nature
Coercive force H_c	field required to move pinned domain walls or to rotate single-domain magnetization
CRM	chemical remanent magnetization; remanence acquired when a magnetic mineral either nucleates and grows through a critical size or grows at the expense of a parent magnetic phase in a magnetic field
Domain	region of a crystal (usually platelike) in which the magnetization is uniform and equal to $\mathbf{M_s}$, or nearly so
DRM	depositional remanent magnetization; remanence acquired during or shortly after deposition of magnetic grains in water in the presence of a field
Induced magnetization $\mathbf{M}$	magnetization produced by an applied field $\mathbf{H}$; part or all of $\mathbf{M}$ is lost when $\mathbf{H}$ is removed
LEM	local energy minimum; one of several minimum-energy states of a system but not necessarily the equilibrium or lowest energy state
Micromagnetic	magnetization structure viewed at a scale intermediate between the atomic and the macroscopic; also the method of unconstrained energy minimization used to calculate such structures
MD	multidomain; containing more than one magnetic domain; magnetization changes occur mainly by moving walls or by nucleating/denucleating domains
N	demagnetizing factor, the ratio between internal demagnetizing field and magnetization M; determined by geometry and to a lesser extent by domain structure
NRM	natural remanent magnetization; any remanence acquired by a rock, sediment or soil under natural as opposed to laboratory conditions; the exact acquisition process is often unknown
pTRM	partial thermoremanent magnetization; remanence acquired by cooling in a field over a specified temperature interval and in zero field over all other temperature intervals
PSD	pseudo-single-domain; a remanence or a magnetization structure which is either non-uniform or occurs in grains larger than equilibrium SD size, yet has some aspects of SD behavior, e.g. high $\mathbf{M_{rs}}$ or $\mathbf{H_c}$
Remanent magnetization or remanence $\mathbf{M_r}$	net magnetization remaining in zero applied field
Saturation remanence $\mathbf{M_{rs}}$	remanence remaining after removal of a field large enough to saturate $\mathbf{M}$
Saturation or spontaneous magnetization $\mathbf{M_s}$	magnetization within a domain, produced spontaneously by exchange coupling of atomic spins; revealed macroscopically by applying a saturating field
Susceptibility k	ratio of induced magnetization $\mathbf{M}$ to applied field $\mathbf{H}$; numerically close to 1/N in MD ferromagnetics
SD	single-domain; containing only one domain; magnetization changes occur by rotating all or most atomic spins simultaneously
SP	superparamagnetic; a condition in which magnetization changes are thermally activated in times much shorter than observation times, usually at high temperatures or small grain sizes; an ensemble of such grains has an equilibrium magnetization
Thermal demagnetization	progressive demagnetization effected by zero-field heating, the peak temperature increasing with each step of the procedure; in continuous thermal demagnetization, magnetization is measured at each peak temperature; in stepwise thermal demagnetization, commonly used by paleomagnetists, the sample is zero-field cooled to room temperature for measurement at each step
Thermoviscous remanence	remanence acquired during very slow cooling in a field, in which time t and temperature T both play a role; sometimes called viscous pTRM
Transdomain	a remanence, a structure, or a magnetization process involving changes between LEM states; in grains large enough to contain recognizable domains, this requires nucleation/denucleation of domains
TRM	thermoremanent magnetization; remanence produced by cooling in a field from the Curie point to room temperature
VRM	viscous remanent magnetization; remanence acquired in a field applied at constant temperature T for a long time t
Wall	the boundary region between domains, in which atomic spins change direction over several hundred lattice spacings and $\mathbf{M}$ is locally nonuniform; walls move to enlarge some domains and shrink others, thus changing the net $\mathbf{M}$ of a grain

also sensitive tracers of the provenance and dispersal of sediments, soils, and pollutants [*Thompson and Oldfield*, 1986]. For a detailed discussion, including catchment studies, diagenesis, core correlation, and paleoseismicity, see the accompanying paper by *Verosub and Roberts* [1995].

Magnetic Anisotropy, Rock Fabrics, and Deformational Histories

Anisotropies of susceptibility and of laboratory produced remanence are sensitive indicators of igneous, sedimentary and metamorphic petrofabrics [*Jackson and Tauxe*, 1991; *Rochette et al.*, 1992]. They also measure strain in deformed rocks and so are directly relevant to tectonic interpretations [*Richter et al.*, 1993].

Magnetic Anomalies and Lithospheric Magnetic Properties

Regional to global scale magnetic anomalies provide information about the chemical and hydrothermal environment of the present and ancient lithosphere and about the depth extent of tectonic processes in orogenic belts. These anomalies reveal high magnetizations in the continental lower crust and oceanic upper mantle and a contrast between generally magnetic Archean cratons and essentially demagnetized mobile belts bounding them [*Arkani-Hamed*, 1992]. Direct information about these deep rocks comes from sampling the upper 2 km of oceanic crust in situ, up-faulted middle to lower crustal and upper mantle sections [*Percival et al.*, 1989; *Shipboard Scientific Party*, 1993], and deeply exhumed orogens like the Grenville Province of North America. Systematic studies of k and M_r for such lithospheric sections are in their infancy [*Frost and Shive*, 1986; *Smith and Banerjee*, 1986; *Hall et al.*, 1991].

Records of Polarity Reversal of the Geomagnetic Field

Gauss and his contemporaries in the mid-nineteenth century observed that Earth's field is not static. The most striking change in the field between Gauss's time and our own is a >5% decrease in Earth's dipole moment. Similar changes in the paleofield intensity are well documented in rocks. Sometimes they presage a reversal of the field from one polarity state to another.

Earlier this century it was not clear whether the field had actually reversed or whether some rocks could become magnetized antiparallel to the field ("self-reversal"). *Brunhes* [1906] and *David* [1904] were the first to measure NRMs that were antiparallel to the local geomagnetic field. Both lavas and the clays they had baked were reversely magnetized. Since the two materials have very different mineralogies, it is unlikely that both would exhibit self-reversal.

Matuyama [1929] showed that younger Quaternary lavas, now known to be <0.78 Ma in age, were all magnetized normally (parallel to the present geomagnetic field), whereas older lavas were reversely magnetized. The association of normal and reverse polarities with different geological times confirms that Earth's field has reversed polarity. It also forms the basis for the geomagnetic polarity time scale [*Cox et al.*, 1963] and magnetostratigraphy [*Harland et al.*, 1990].

An exciting area of recent research is tracking the geomagnetic field during a reversal [*Prévot et al.*, 1985; *Tric et al.*, 1991]. These data constrain models of the geodynamo, and by inference tell us about the nature of the core and the core-mantle boundary region. Field behavior is deduced from the vector record of NRM $\mathbf{M_r}$ in successive lava flows or rapidly deposited sediments. The fidelity of the recording at a microscopic level needs investigation, particularly how this can be compromised by shallowing of the $\mathbf{M_r}$ vector during sedimentation or by secondary alteration and diagenesis [*van Hoof and Langereis*, 1992].

Linear Magnetic Anomalies: The Seafloor Record of Polarity Reversals

Lithospheric plate motions result in a smooth spreading of newly erupted seafloor basalts away from mid-ocean ridges. The basalts contain fine grains of titanomagnetite ($Fe_{2.4}Ti_{0.6}O_4$) which acquire an intense thermoremanent magnetization (TRM) on cooling below their Curie temperatures. The time series of geomagnetic polarity epochs thus becomes fossilized as a spatial sequence of normally and reversely magnetized bands of seafloor, symmetrical about mid-ocean ridges and increasing in age with distance from the ridge. A replica of the magnetization pattern appears at the sea surface in the form of linear magnetic anomalies.

Magnetic lineations are tangible evidence of seafloor spreading. Historically, they were decisive in converting geologists to a mobilist view of Earth. They are the main means of determining plate velocities, and they provide the most complete record of geomagnetic reversal history during the last 200 m.y. In view of the exhaustive research that has been carried out since *Vine and Matthews* [1963] first proposed their model, it might be thought that very little remains to be learned. Surprisingly, this is not so. Using a tape recorder as an analogy, the tape drive (the spreading seafloor and the tectonic forces that drive it) and the time series of geomagnetic reversals that are recorded have been thoroughly looked into, but the recording medium itself, the magnetic seafloor, is still imperfectly understood.

The first surprise is that chemical alteration has destroyed all primary TRM in much of the seafloor. Before new seafloor has spread more than 100 km from its parent ridge, seawater has penetrated deeply along fissures and oxidized the primary titanomagnetites, changing both their chemistry and their magnetization. The secondary chemical remanent magnetization (CRM) may have a different direction (e.g., opposite polarity) from the primary TRM it replaces. A major puzzle is how the oceanic crust can maintain a clear pattern of magnetic lineations in the face of this pervasive remagnetization.

Paleomagnetic Field Intensity

The intensity of the geomagnetic field is deduced from the intensity of primary remanence $\mathbf{M_r}$, usually TRM, recorded in rocks. Paleofield intensity has been studied for both young and old rocks. Earth's field existed at least since 3500 Ma [*Hale and Dunlop*, 1984], but we have only a fragmentary knowledge of how its strength has varied with time.

Rocks which behave ideally during paleointensity experiments are unfortunately few and far between. Successful determinations usually require single-domain grains which do not alter in repeated heatings. Recently, there have been attempts to extend the range of suitable rocks; submarine volcanic glasses are particularly promising [*Pick and Tauxe*, 1993].

Paleomagnetism and Plate Motions

Plate motions during the past 200 m.y. are tracked by linear magnetic anomalies over the oceanic portions of the plates.

Tracing earlier motions requires spot readings, usually obtained by isolating well-dated primary or secondary remanences in continental rocks and determining paleolatitude and paleoazimuth from the vector direction of $\mathbf{M_r}$. Paleomagnetic evidence that continents have moved about Earth was available in the 1950s but was ignored for a long time by the geological community. Why did this happen, and why was magnetic evidence from the spreading seafloor felt to be so much more compelling a decade later?

At least part of the explanation is rock magnetic. Young seafloor lavas seemed to be simple magnetic recorders (more recent evidence to the contrary has been largely ignored) and their TRM record is binary: normal or reverse polarity. Continental rocks comprise a wide range of lithologies and ages. Once laboratory demagnetization techniques had developed, it became obvious that only part of the NRM was stable and by implication reliable. Small errors in directions of the vector components could cause large differences in the inferred paleopositions of the continents.

Paleomagnetically based plate reconstructions are now generally accepted without comment if they fit geological and paleoclimatological constraints, but the problem of assessing the reliability of individual results remains [*Van der Voo*, 1993]. Paleomagnetists increasingly have come to recognize that the older the rock, the more intricate (and interesting) the mixture of different NRMs it is likely to contain. For example, NRMs of very different ages are carried by separate minerals (magnetite and hematite, αFe_2O_3) in some Appalachian rocks [*Kent and Opdyke*, 1985]. The secondary NRM is a CRM of late Carboniferous age perhaps related to tectonically driven fluids associated with the Alleghenian/Hercynian orogeny. The age and significance of such CRMs were only belatedly recognized [*McCabe and Elmore*, 1989].

Biomagnetism

Many organisms, man included, contain intracellular magnetite. Some species of neutral-density water-dwelling bacteria use their magnetite to sense geomagnetic field lines and thereby swim down to feed or to optimize their microaerobic environment. Honeybees, homing pigeons, migratory birds, and dolphins all contain magnetite sensors which in some cases are utilized as compasses. An account of this fascinating subject is given by *Kirschvink et al.* [1985].

Magnetotactic bacteria have evolved efficient compass "needles" consisting of chains of single-domain magnetite crystals polarized along the chain axis. Rock magnetic tests, for single-domain behavior and for interactions between crystals in a chain, are used by biomagnetists to categorize biogenic magnetites [*Moskowitz et al.*, 1988].

Beginnings of Rock Magnetism

Magnetic Domains

Weiss [1907] was intrigued by the fact that iron and other "soft" magnetic materials with small permanent magnetizations become strongly magnetized when exposed to quite weak magnetic fields. Weiss proposed that external fields play a minor role compared to an internal "molecular field" (now known to be due to quantum mechanical exchange coupling), which aligns the magnetic moments of individual atoms, producing a spontaneous magnetization, $\mathbf{M_s}$. In the absence of an external field, the magnetic moments of domains with different directions of $\mathbf{M_s}$ cancel almost perfectly, but even a small applied field will rotate domains or enlarge some at the expense of others.

Without the benefit of any substantial data base, *Landau and Lifschitz* [1935] correctly predicted the form and dimensions of ferrimagnetic domains and domain walls. Reasoning that domain boundaries would be positioned so as to maximize internal flux closure, they postulated precisely the structure illustrated in Figure 1. The platelike main or body domains (seen here edge on) are terminated by wedge-shaped surface closure domains which provide a continuous flux path between neighbouring body domains.

The Advent of Rock Magnetism

The subjects of ferrimagnetism and geomagnetism developed in mutual isolation until *Koenigsberger* [1938], *Thellier* [1938], and *Nagata* [1943] brought Earth magnetism into the laboratory. Reproducing the process by which igneous rocks are magnetized in nature, they gave TRMs to their samples (lavas and archeological materials like bricks and baked clays) by heating them to high temperatures and cooling them in a weak field. The TRM vector, $\mathbf{M_{tr}}$, was always parallel to the field $\mathbf{H}$ in which it was acquired, and its intensity was proportional to the strength of the field. TRM was therefore an accurate recorder of magnetic field directions, as geomagnetists had assumed, and was also potentially a means of determining paleofield intensities.

TRM in the laboratory had a series of remarkable properties which were most clearly described by Thellier. When a sample was given a partial TRM, by exposing it to a field only in a narrow cooling interval (between T_1 and T_2 say), and was subsequently reheated to high temperatures in zero field, the TRM remained unchanged below T_2 but completely disappeared between T_2 and T_1.

Néel [1949] explained this observation as a consequence of blocking of TRM during cooling at a single blocking temperature T_B determined by the size and shape of a particular single-domain grain. The TRM is unblocked when reheated through T_B. The wide spectrum of grain sizes and shapes in a rock leads to a continuous distribution of partial TRM blocking temperatures, but each single-domain grain has one and only one value of T_B.

This individuality of blocking temperatures illuminates other experimental TRM "laws." Partial TRMs acquired in different temperature intervals are mutually independent. For example, if two partial TRMs are added vectorially during cooling, e.g., by rotating the field by a large angle after the first partial TRM is produced, the two partial TRMs demagnetize independently of each other during zero-field reheating. Each partial TRM disappears over its own blocking temperature interval, and the total magnetization vector retraces the exact pattern it followed during cooling. This property allows us to separate primary and secondary remanences acquired by rocks in nature at widely different times.

Koenigsberger, Thellier, and Nagata were fortunate in their choice of fine-grained rocks containing an abundance of single-domain magnetite and hematite. Only single-domain grains exhibit a unique TRM blocking temperature and obey the Thellier laws. The TRM of larger multidomain (MD) grains is complicated by the mobility of domain walls and is less obviously suitable for recording the paleomagnetic field.

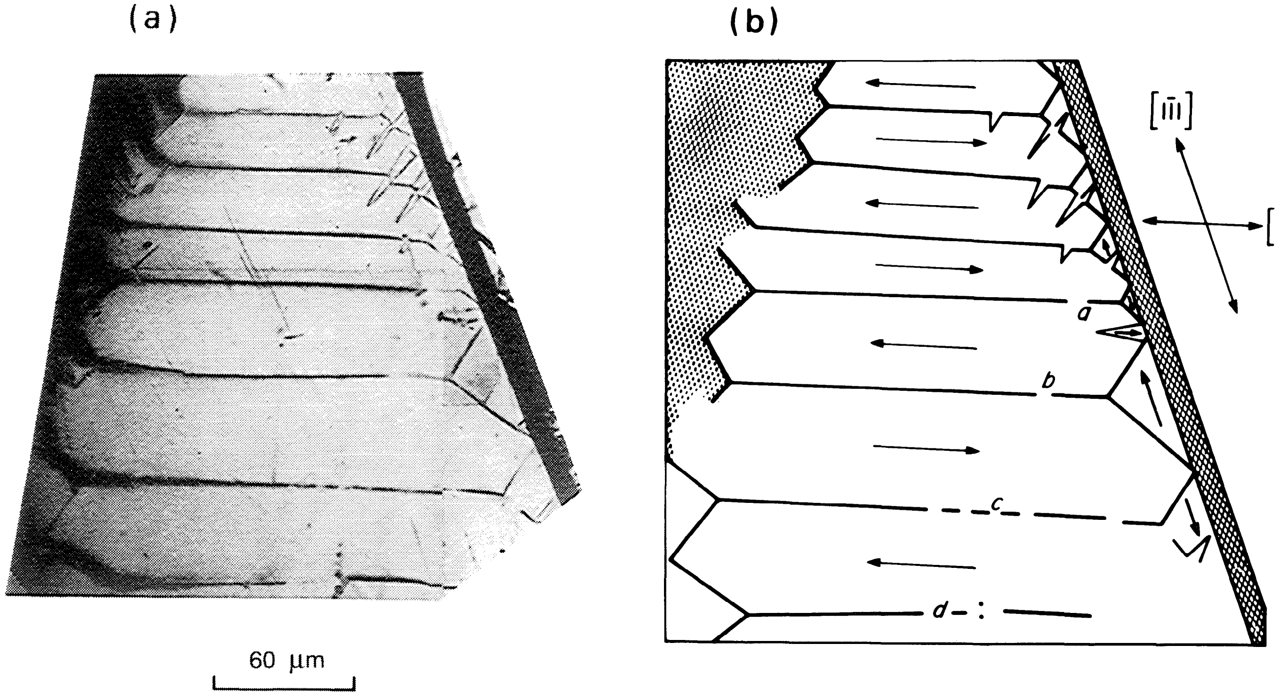

Figure 1. Body and surface (closure) domains observed on a polished {110} surface of a large magnetite crystal and inferred directions of $\mathbf{M_s}$ within each domain. The domain walls are made visible by a colloidal suspension of ultrafine magnetite particles. After *Özdemir and Dunlop* [1993].

Verhoogen [1959] and *Stacey* [1962, 1963] independently proposed a new view: multidomain grains that have aspects of single-domain behavior. *Stacey and Banerjee* [1974] in their classic textbook explored the ramifications in detail. Pseudo-single-domain (PSD) models have inspired and shaped rock magnetic research ever since.

Pseudo-Single-Domain Magnetic Properties

Following application of a strong laboratory field **H**, single-domain (SD) grains have a saturation remanence $\mathbf{M_{rs}}$ which is a large fraction of $\mathbf{M_s}$. For randomly oriented grains, M_{rs} = 0.50 M_s or 0.87 M_s, depending on whether the grains have a single axis or multiple axes of easy magnetization. Multidomain (MD) grains, on the other hand, should have $M_{rs} \approx kH_c \approx H_c/N$ (see introduction), where the coercive force or coercivity H_c is the field necessary to move pinned domain walls. Taking as an example approximately equidimensional MD grains of magnetite, for which $M_s = 4.8 \times 10^5$ A/m, $N \approx 1/3$, and $H_c \approx 1.6 \times 10^3$ A/m, we find $M_{rs} \approx 4.8 \times 10^3$ A/m = 0.01 M_s. Thus above the critical SD grain size d_o (about 0.1 μm in magnetite), when domain walls begin to appear, there should be a sharp drop in saturation remanence (Figure 2).

Experimentally, nothing of the sort is seen (Figure 2). Above d_o, there is a smooth, gradual decrease in M_{rs} values over many decades of grain size d. TRM intensities have a similar size variation. This range of grain sizes is loosely referred to as the PSD range, although it has only recently been demonstrated that the remanence has a truly single-domain part [*Dunlop and Argyle*, 1991]. Magnetites as large as 10-100 μm in size can have intermediate or PSD properties. Most magnetite grains that carry paleomagnetic remanences fall in the PSD range.

There are several possible explanations for PSD behavior.

1. Grains which should nucleate walls following saturation fail to do so and remain in metastable SD states. There is considerable experimental evidence for this [*Halgedahl and Fuller*, 1980, 1983; *Boyd et al.*, 1984; *Halgedahl*, 1991].

2. Grains do nucleate walls, but the walls have SD-like moments ("psarks" [*Dunlop*, 1977]) and grains with odd numbers of domains have net unbalanced moments [*Dunlop*, 1983a].

3. Conventional domains and walls cannot develop properly in submicron grains; instead, there is a continuous spatial variation of magnetization within each grain [*Williams and Dunlop*, 1989].

4. H_c in small grains is much larger than the typical MD value assumed above. (Experimentally, this is true and ensures that PSD remanence is stable as well as strong, but the argument is circular. It is not clear whether the higher H_c values are actually due to stronger pinning of walls or are associated with the SD-like component of remanence.)

Dunlop [1986] examined all available M_{rs} and other hysteresis data for magnetite and concluded that models 1, 2, and 4 explained the data about equally well for submicron grains. More recently, model 3 has also been shown to correctly predict measured hysteresis in submicron magnetites (W. Williams and D.J. Dunlop, Simulation of magnetic hysteresis in pseudo-single-domain grains of magnetite, submitted to *Journal of Geophysical Research*, 1994), although there should still be a sharp drop in remanence-carrying ability near 0.1 μm (Figure 2) [*Newell et al.*, 1993].

Domain Observations and Micromagnetic Calculations

A new era in rock magnetism began when it was observed that some titanomagnetite ($Fe_{2.4}Ti_{0.6}O_4$) grains much larger than SD critical size fail to nucleate domain walls following

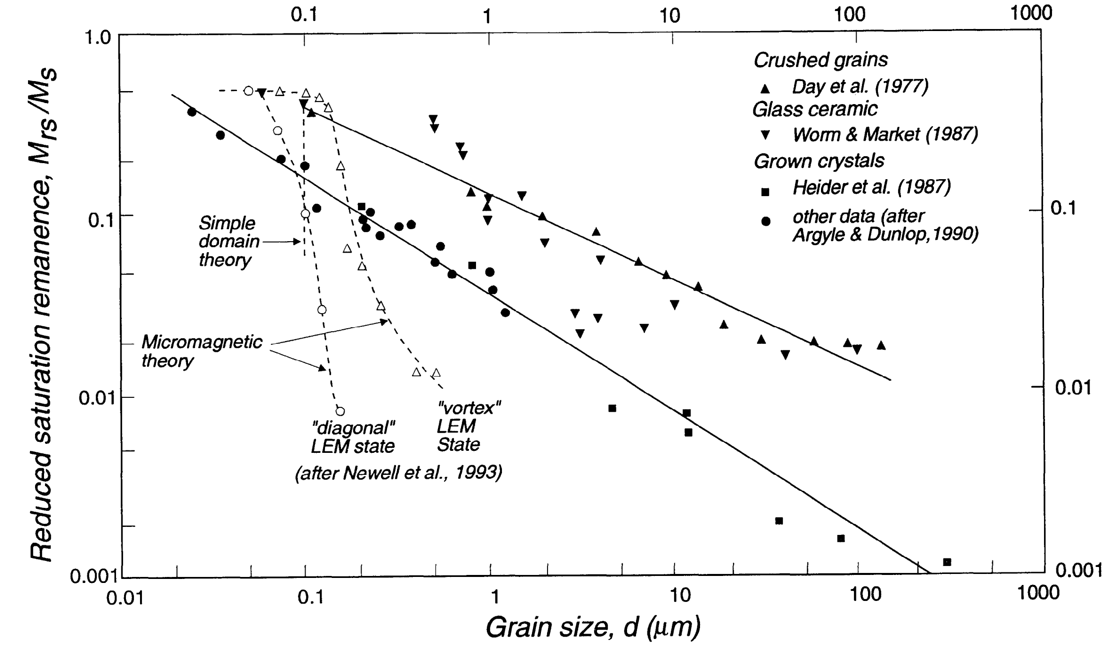

Figure 2. Theoretical (dashed line and curves) and measured dependences of M_{rs}/M_s on grain size for crushed (high stress) and grown (lower stress) magnetite grains. The solid lines are best fits to the Day et al. and to the Heider et al. and other data, respectively. They can be used as master curves for determining grain size from measured saturation remanence, if the internal stress state is known.

either exposure to saturating fields [*Halgedahl and Fuller*, 1980, 1983] or heating to the Curie temperature [*Metcalf and Fuller*, 1987a; *Halgedahl*, 1991]. The magnetic moments of even a few such metastable SD grains might well outweigh the remanence of both MD and ordinary SD grains in a rock. The situation in magnetite is less clearcut because grains in the size range $d_0 - 10d_0$ are optically invisible, but observations of >1 μm grains suggest that the number of body domains in a particular size grain is substantially less than the number predicted (Figure 3) [*Worm et al.*, 1991].

Another vital observation made by Halgedahl, Fuller, and their colleagues was that a particular grain cycled repeatedly through the same experimental conditions has a choice of several different remanent states. Until this point, rock magnetic theory had implicitly assumed that grains always had equilibrium domain structures, corresponding to minimum total energy. *Moon and Merrill* [1984, 1985] introduced the idea of local energy minimum (LEM) domain states, analogous to the excited states of an atomic system. Transdomain processes or transitions between LEM states (amounting to the nucleation or

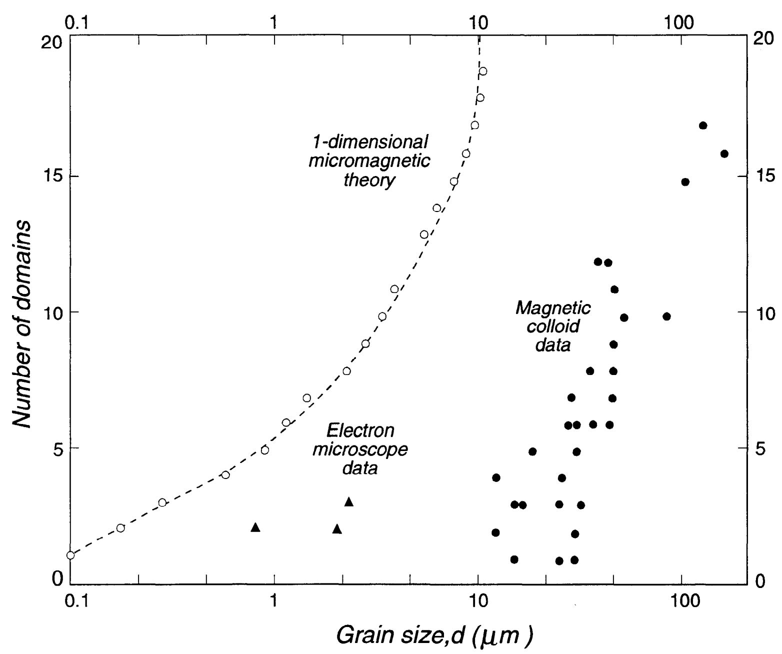

Figure 3. Expected number of domains in magnetite grains of different sizes according to one-dimensional micromagnetic calculations compared to the number actually observed using magnetic colloids and Lorentz electron microscopy. (Redrawn after *Worm et al.* [1991].)

denucleation of one or more domains), rather than domain rotation or domain wall motion, might control thermoviscous processes like TRM and time-dependent or viscous remanent magnetization (VRM) [*Moon and Merrill*, 1986; *Enkin and Dunlop*, 1987; *Moskowitz and Halgedahl*, 1987].

Moon and Merrill [1984, 1985] and *Enkin and Dunlop* [1987] took a new approach to calculations. Instead of imposing domains and walls of predetermined form, as in classical domain theory, they solved a variational problem in which the magnetization direction is permitted to vary continuously throughout the grain. These early micromagnetic calculations allowed only a one-dimensional variation of $\mathbf{M_s}$ and as a result reproduced classical domain structures, albeit with interesting and unanticipated details of wall structure. But subsequent calculations in which $\mathbf{M_s}$ was unconstrained in either two dimensions [*Newell et al.*, 1993] or three dimensions [*Williams and Dunlop*, 1989, 1990, also submitted manuscript, 1994; *Shcherbakov et al.*, 1990] made it clear that grains in the crucial range $d_o - 10d_o$ (0.1-1 μm for magnetite) should have a continuous variation in magnetization direction, in the form of vortex or other patterns. Conventional domain structures only develop in grains larger than $\approx 10d_o$ [*Xu et al.*, 1994].

Two recent developments may narrow the gap (Figure 3) between theory and experiment. First is the observation (Figure 1) [*Özdemir and Dunlop*, 1993], and micromagnetic prediction [*Xu et al.*, 1994] of abundant surface closure domains in magnetite. Their existence provides internal flux closure and reduces the required number of body domains by about a factor 3. Second is the recognition that micromagnetic structures near the grain surface may not image well in domain observations [*Ye and Merrill*, 1991; *Williams et al.*, 1992a; *Newell et al.*, 1993]. For example, a vortex structure, which has only a small remanence, might falsely image as a high-remanence SD grain. (Note, however, that this cannot be a general explanation for metastable SD grains. These grains are often observed to have alternative states containing domain walls [*Halgedahl and Fuller*, 1983; *Halgedahl*, 1991].)

Rock Magnetic Tests for Domain Structure, Stability, and Type of NRM

Despite our present uncertainty about what microscopic variation of **M** in magnetic mineral grains actually generates macroscopic magnetic properties of rocks, many tests have been devised with a view to diagnosing domain structure and, by implication, grain size and paleomagnetic stability. Virtually all apply to magnetite; databases for other minerals are much less detailed. Figure 2 shows that M_{rs}/M_s varies smoothly with grain size d for magnetites of a common provenance: M_{rs}/M_s data "measure" grain size even though the underlying domain structures are uncertain. (Other magnetic properties, among them H_c, susceptibility k, and anhysteretic susceptibility, in which the steady field is augmented by a field of alternating polarity and strength, are used in a similar manner [e.g., *King et al.*, 1983].) Unfortunately, internal stress (which is only known in a general way for most rocks) affects the master curves in Figure 2 and compromises the test. A popular variant [*Day et al.*, 1977] which compares saturation remanence and coercivity data has the same drawback and is intended only to classify rocks broadly into SD, PSD, and MD categories.

Lowrie and Fuller [1971] proposed a test that initially seemed to offer what hysteresis tests do not: a binary classification into SD or MD behavior. This was exciting because the test uses alternating field (AF) demagnetization data measured routinely in paleomagnetism, can be applied to NRM directly, and would permit a rapid selection of rocks with SD carriers of NRM. It soon became clear that PSD as well as SD grains have "SD" Lowrie-Fuller tests [*Dunlop et al.*, 1973; *Johnson et al.*, 1975]. In recent work on grown magnetites, only grains >100 μm in size gave "MD" test results [*Heider et al.*, 1992]. The test therefore has limited discriminating power.

The most powerful and practical tests are still those developed in the early days of paleomagnetism: the stepwise AF or thermal demagnetization of the NRM vector $\mathbf{M_r}$. The underlying assumption is that high blocking temperature T_B or coercivity H_c imply a long magnetic memory. "Stable" is used in the paleomagnetic literature to describe all three. It was *Néel* [1949, 1955] who provided the link among the three types of "stability". Néel showed that the relaxation time τ of the remanence of an SD grain is

$$\tau = \tau_o \exp[VM_s(T)H_c(T)/2kT], \qquad (1)$$

V being grain volume, k Boltzmann's constant, and $\tau_o \approx 10^{-10}$ s an atomic reorganization time. Despite the small value of τ_o, the exponential dependence of τ on H_c and T ensures that all except the very smallest SD grains have relaxation times of millions to billions of years.

This brilliant piece of theoretical work provided the justification for thermal and AF "cleaning" techniques developed by experimenters like *Thellier* [1938], *Creer* [1959], *Wilson* [1961], and *Zijderveld* [1967]. However, theoretical understanding of how the remanence of pinned walls in MD grains responds to alternating fields and heating has only recently been achieved [*Xu and Dunlop*, 1993, 1994; *Dunlop and Xu*, 1994]. Key experimental observations were made by *Bolshakov and Shcherbakova* [1979] and *Worm et al.* [1988], who found that partial TRMs blocked between T_1 and T_2 during cooling do not unblock when reheated from T_2 to T_1 in the manner of SD grains but continue to demagnetize gradually above T_1 right up to the Curie temperature (Figure 4). Thus it may be difficult to confidently isolate different generations of NRM by means of thermal or AF demagnetization if one or more of the NRMs is carried by MD grains.

This conclusion makes it all the more important to determine whether PSD remanence is basically SD-like (in metastable SD grains or psarks) or is a manifestation of multidomain or other non-uniform magnetization in small grains. Experimentally, the response of partial TRM and VRM in PSD-size magnetite grains to continuous thermal demagnetization (in which measurements are made continuously at high temperature rather than at room temperature in a series of discrete steps) is intriguing and nonintuitive [*McClelland and Sugiura*, 1987; *Halgedahl*, 1993] and is interpreted as being a transdomain process resulting from nucleation or denucleation of domains during heating [*Shcherbakov et al.*, 1993].

In addition to separating different vectorial components, e.g., $\mathbf{M_{r1}}$ and $\mathbf{M_{r2}}$, in a composite NRM by laboratory demagnetization, one would also like to have a test or tests for the nature of the separated vectors: primary TRM or secondary pTRM or CRM, for example. Frequently, the demagnetization ranges of the vectors overlap to a greater or lesser extent, as indicated by curved rather than straight segments in the demagnetization trajectory of $\mathbf{M_r} = \mathbf{M_{r1}} + \mathbf{M_{r2}}$ in vector space. Of course, overlap may result from non-SD carriers of one or other NRM vector, as just discussed, but it can also indicate that one component, say $\mathbf{M_{r2}}$, is a CRM [e.g., *McClelland-Brown*,

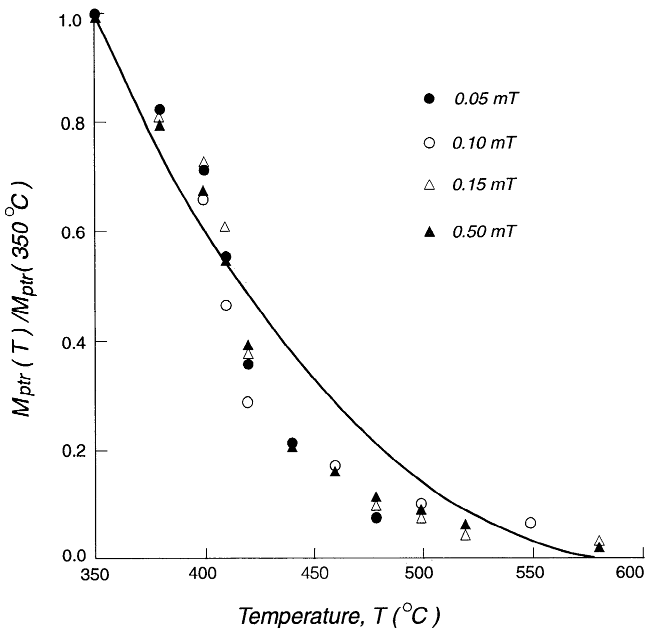

Figure 4. Continuous thermal demagnetization data for pTRMs acquired between 400 and 350°C in various fields by 3 μm magnetite grains [after *Worm et al.* 1988]. About two thirds of the remanence is demagnetized at or just above 400°C, as predicted by SD theory. The remainder has unblocking temperatures distributed up to the Curie temperature (580°C), as predicted by MD theory (solid curve is after *Xu and Dunlop* [1994]).

1982]. In the simplest type of CRM, grain-growth CRM, the remanence is blocked because grains grow from initially small volumes V (and consequently small values of τ according to equation (1)) to larger V and very large values of τ. Since grains typically grow considerably larger than the minimum V required to ensure stability, their ultimate blocking temperatures in thermal demagnetization may be much greater than the temperature at which the CRM was blocked. Partial TRM, on the other hand, if carried by SD grains, will replace (or overprint) all primary remanence with $T_B < T_r$ (the reheating temperature in nature) and will leave unaffected all remanence with $T_B > T_r$. A refined version of this test, the baked contact test, takes advantage of the variable reheating temperature T_r with distance from a dike or other intrusive rock body [*Schwarz and Buchan*, 1989].

There are few other laboratory procedures that can pinpoint primary NRM (TRM in igneous rocks, depositional remanent magnetization or DRM in sedimentary rocks) or discriminate between CRM and other types of secondary NRM with any degree of confidence. A successful Thellier paleointensity experiment is a strong indication that the NRM is a primary TRM, but successes are so rare (because of alteration of samples during repeated heatings [*Coe*, 1967]) that this "test" is seldom attempted in ordinary paleomagnetic work. Field-based tests date a particular NRM component relative to a geological event such as physical weathering (the conglomerate test), deformation (fold and tilt tests), or igneous activity (contact test).

Microanalysis of NRM

An obvious, but technically challenging, means of investigating the origin of NRM components is to carry out vector measurements on oriented microsamples containing single mineral grains or aggregates of a particular mineral [*Geissman et al.*, 1988], with or without accompanying microstructural study using the electron microscope. Both silicate minerals with ferrimagnetic inclusions and ferrimagnetic minerals with silicate inclusions may contain sufficient potassium to be datable by the $^{40}Ar/^{39}Ar$ method [e.g., *Özdemir and York*, 1992]. Eventually, it may be possible to determine both a silicate mineral cooling age and the nature of NRM for individual mineral phases in rocks.

The first fully vectorial microanalysis was carried out by *Wu et al.* [1974] on oriented crystals of plagioclase, biotite, and hornblende from a Miocene age granodiorite. All three minerals contained magnetite inclusions in sufficient quantity to give measurable NRMs, but the plagioclase and hornblende remanences were much more stable than the biotite remanence. *Buchan* [1979] was the first to separate ancient NRMs of very different ages residing in different minerals. His data for a Precambrian diorite (Figure 5) show that a ≈820 Ma NRM in ferromagnesian silicates (presumably a CRM) and a ≈980 Ma NRM in hematite and possibly also plagioclase combine to give the whole rock NRM.

To avoid the labor of cutting microsamples from a rock and the inevitable uncertainty in their orientations, several recent studies [e.g., *Renne and Onstott*, 1988] have employed selective demagnetization techniques, in which individual mineral grains are physically removed from a rock, by laser fusion or other methods, and the incremental remanence is measured. Although there is no doubt that the incremental remanences would be easily measurable in isolation by modern superconducting magnetometers, it is not clear how accurately their directions are determined against the large residual background remanence of the rock sample. This uncertainty tends to offset the advantages of not having to orient grains individually.

The next step will be to compare the vector NRM properties of individual oriented crystals with their observed domain structures [*Halgedahl*, 1992]. A step in this direction was taken by *Metcalf and Fuller* [1987b], who observed domain patterns on polished titanomagnetite crystals in a seafloor basalt during field cycling and then removed individual crystals for measurements of their hysteresis and AF demagnetization behavior.

Thermoviscous Remagnetization and Hopkinson Peaks

Rocks which have been deeply buried, tectonically or in a thick sedimentary or volcanic pile, undergo thermoviscous remagnetization because of the combined effect of elevated temperatures and long times. Thermoviscous NRM overprints resemble pTRMs and VRMs and can be recognized by their linear thermal demagnetization trajectories. However, the maximum unblocking temperature T_B in short-term laboratory heatings is somewhat greater than the maximum reheating temperature T_r in nature because of the long reheating time t_r. Appealing to Néel's relaxation equation (1), one finds [*Pullaiah et al.*, 1975]

$$T_B \ln(t_L/\tau_o)/M_s(T_B)H_c(T_B) = T_r \ln(t_r/\tau_o)/M_s(T_r)H_c(T_r), \quad (2)$$

in which t_L is the time of laboratory heating.

There has been considerable controversy because in some rocks thermoviscous overprints, whose values of T_r and t_r could be deduced from independent information, exhibited

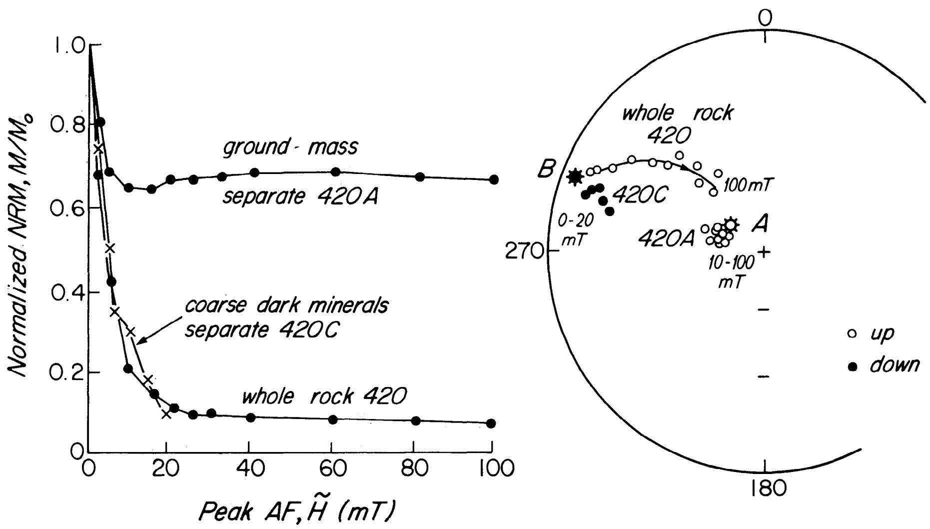

Figure 5. Microanalysis of NRM in a Precambrian diorite (redrawn after *Buchan* [1979]). Plotted are the intensity and direction (in stereographic projection) of the composite NRM, $\mathbf{M_r} = \mathbf{M_{rA}} + \mathbf{M_{rB}}$ in the course of stepwise AF demagnetization. A and B are the mean directions of $\mathbf{M_{rA}}$ and $\mathbf{M_{rB}}$. These NRM components have respective ages of 980 and 820 Ma, and reside in the ground mass (containing hematite and plagioclase) and in ferromagnesian minerals, respectively.

maximum laboratory unblocking temperatures higher than predicted by (2). In some cases [e.g., *Kent*, 1985], for magnetite, T_B was in accord with predictions by *Middleton and Schmidt* [1982] based on a theory by *Walton* [1980]. Kent's data are shown in Figure 6.

This issue is not conclusively settled, but truly SD grains of hematite [*Enkin and Dunlop*, 1988] and magnetite [*Dunlop and Özdemir*, 1993] definitely do obey (2). "Anomalously" high unblocking temperatures probably reflect non-SD carriers of thermoviscous magnetization [*Dunlop et al.*, 1994]. Either domain wall pinning/unpinning (Figure 4) or transdomain processes could produce a tail of high unblocking temperatures. Another possibility is that some overprints, e.g., the higher-T_B overprint in Kent's Appalachian limestones, are not thermoviscous overprints but CRMs (see next section).

Long times and high temperatures not only reactivate remanent magnetization; they enhance induced magnetization as well. If T is increased in (1), the relaxation time τ decreases,

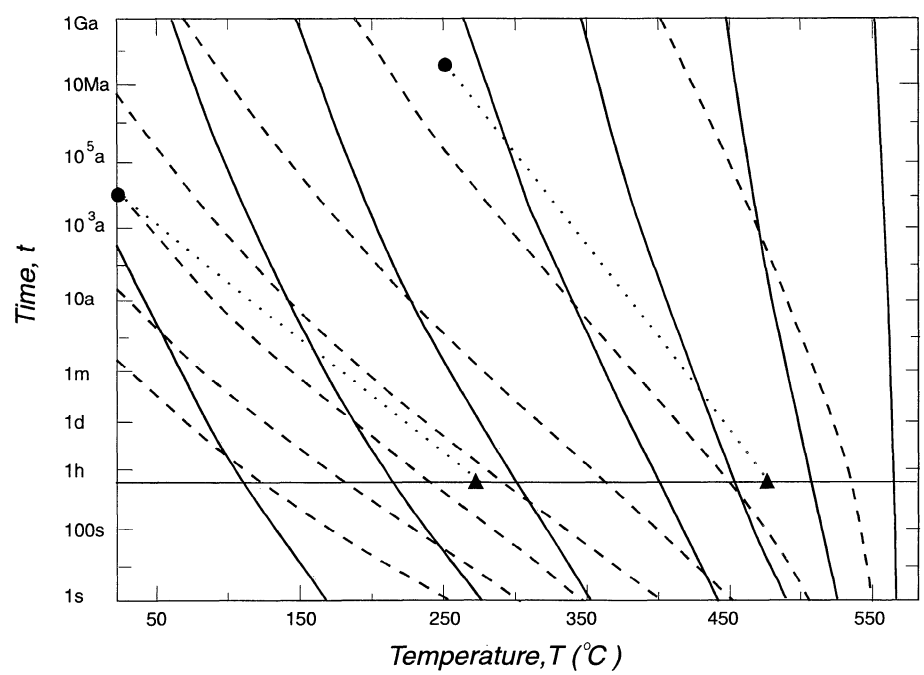

Figure 6. Comparison of t,T data for thermoviscous NRM overprints as blocked in nature (circles: t_r,T_r) and unblocked in laboratory heating (triangles, t_L,T_B) for limestones [*Kent*, 1985] with theoretical relaxation time-temperature relations proposed by *Pullaiah et al.* [1975] and *Middleton and Schmidt* [1982] (solid and dashed curves, respectively).

particularly just below the Curie temperature T_C, where $M_s(T)$ drops rapidly towards zero. Under these conditions, magnetization vectors $\mathbf{M_s}$ of individual SD grains are unblocked and can reorient themselves rapidly in response to a field **H**. This condition is described as superparamagnetism (SP). The susceptibility rises to high values, called a Hopkinson peak. When a constant field is applied for long times, as in nature during a geomagnetic polarity chron, there is also an enhancement of viscous induced magnetization, which occurs at somewhat lower temperature [*Dunlop*, 1983b].

Multidomain grains exhibit much smaller Hopkinson peaks [e.g., *Clark and Schmidt*, 1982]. Domain wall mobility and the ease of domain rotation both increase with heating, but because of the strong self-demagnetizing effect in MD grains (see introduction), there is only a small increase in susceptibility k towards its limiting value 1/N.

Some of the strong magnetic anomaly signature of the continental lower crust and oceanic upper mantle may be due to thermal enhancement of susceptibility and viscous magnetization just below the Curie point of magnetite, rather than to unusually high magnetic mineral content. But a mechanism related to Hopkinson peaks is viable only if SD grains are the dominant carriers of induced magnetization in these rocks, and this seems unlikely [*Shive and Frost*, 1992].

Chemical Remanent Magnetization (CRM)

CRM is a ubiquitous secondary NRM, particularly in sedimentary and metamorphic environments. In simple growth CRM, described earlier, a ferrimagnetic crystal nucleates and grows until its relaxation time is long enough to stabilize the direction of **M**; in other words, CRM is blocked when the crystal passes from an SP to a stable SD state [*Kobayashi*, 1961]. CRM also forms when one magnetic phase changes into another [*Haigh*, 1958]. If the two phases have similar crystal lattices, e.g. in the oxidation of magnetite to maghemite (γFe_2O_3), the CRM in the daughter phase may "remember" the NRM direction of the parent phase, through magnetic exchange coupling across the moving phase boundary. This phenomenon is sometimes called phase-coupled CRM or parent-daughter CRM. CRM is usually just as stable to AF demagnetization as any primary NRM, e.g., TRM, that it replaces [*Kobayashi*, 1959], and as discussed earlier, growth CRM is more stable to thermal demagnetization than a pTRM overprint produced at the same temperature.

Two instances of CRM in natural settings have attracted widespread attention because of their broad geological and geophysical significance. The first is CRM accompanying oxidation of titanomagnetites in seafloor basalts. Is this phase-coupled CRM, which would inherit the parent TRM direction and preserve magnetic lineations, or growth CRM, which would reset the NRM to the direction of the geomagnetic field at the time of oxidation? The second example is the CRM of authigenic magnetites, discovered by microanalytical techniques in Appalachian limestones with Alleghenian NRM overprints.

Maghemitization of titanomagnetite ($Fe_{2.4}Ti_{0.6}O_4$) on the seafloor occurs throughout Layer 2A [*Smith and Banerjee*, 1986], generally within a few million years of extrusion. As a result, the amplitude of linear magnetic anomalies drops about an order of magnitude from the central anomaly over an active spreading centre to older lineations over oxidized seafloor. Clearly, some memory of the primary TRM is preserved, because otherwise magnetic lineations would be completely obliterated, but most is lost.

Simulation of the CRM process during maghemitization is not easy because of the difficulty of achieving reasonable reaction rates in the laboratory below the ≈150°C Curie temperature of titanomagnetite. The present picture is based on two key studies. SD-size (<0.1 μm) titanomagnetites with initial TRMs, when oxidized in air to varying degrees, preserved the TRM direction up to ≈65% oxidation, beyond which the daughter product was no longer a single phase (Figure 7) [*Özdemir and Dunlop*, 1985]. On the other hand, PSD-size (<d> = 20 μm) titanomagnetites with initial TRMs, when oxidized in hydrothermal solutions simulating the seafloor environment, acquired CRM along the applied field **H** at all stages of oxidation [*Kelso et al.*, 1991]. In the simplest terms, fine SD grains can preserve "TRM" (i.e., their CRM is phase-coupled to the primary TRM during oxidation) and are responsible for the survival of linear magnetic anomalies, while larger PSD grains acquire growth CRM and lose all memory of the prior TRM. *Raymond and LaBrecque* [1987] reached similar conclusions based on the skewness of oceanic magnetic anomalies.

Permo-Carboniferous remagnetization is widespread on both sides of the Atlantic, not only in the Hercynian/Alleghenian orogen but also in stable platform areas beyond any obvious orogenic influence. The remagnetization may have been caused by fluids driven "onshore" during plate convergence [*McCabe and Elmore*, 1989]. NRM overprints in undeformed, mildly reheated platform carbonate rocks were at first believed to be thermoviscous overprints with anomalously high laboratory unblocking temperatures, but were subsequently found to be low-temperature CRMs carried by authigenic or diagenetically altered magnetites [*McCabe et al.*, 1989; *Jackson*, 1990]. Some of the magnetites have exotic and beautiful forms, for example botryoidal or spheroidal polycrystalline aggregates 10-100 μm in size composed of 0.5-2 μm crystallites [*McCabe et al.*, 1983]. In some cases, the spherules formed by magnetite replacement of pyrite framboids [*Suk et al.*, 1990].

Summary

Rock magnetism has its roots in ferrimagnetism, which was first studied in the early days of this century, and in geomagnetism and paleomagnetism, which are even older subjects. The experimental "laws" governing TRM and partial TRM were discovered around 1940 and their theoretical explanation (which remains the basis of most theorizing today) was given by Néel about a decade later.

Since that time, the applications of rock magnetism have blossomed. Continental and marine magnetic anomalies, the seafloor spreading record, records of geomagnetic secular variation and polarity reversals with their implications for fluid motions in the core, the paleomagnetic record of plate motions and orogenic events, and magnetic sensors in biological organisms all ultimately depend on how magnetic particles respond to present and ancient magnetic fields.

The distinction between single-domain grains, with strong and stable magnetic moments, and multidomain grains, with mobile domain walls, dominates thinking in rock magnetism. Yet the magnetic carriers of remanence in most rocks have a blend of SD and MD character. The pseudo-single-domain problem has been with us since the 1960s and remains unresolved, but it has driven much useful research. For example,

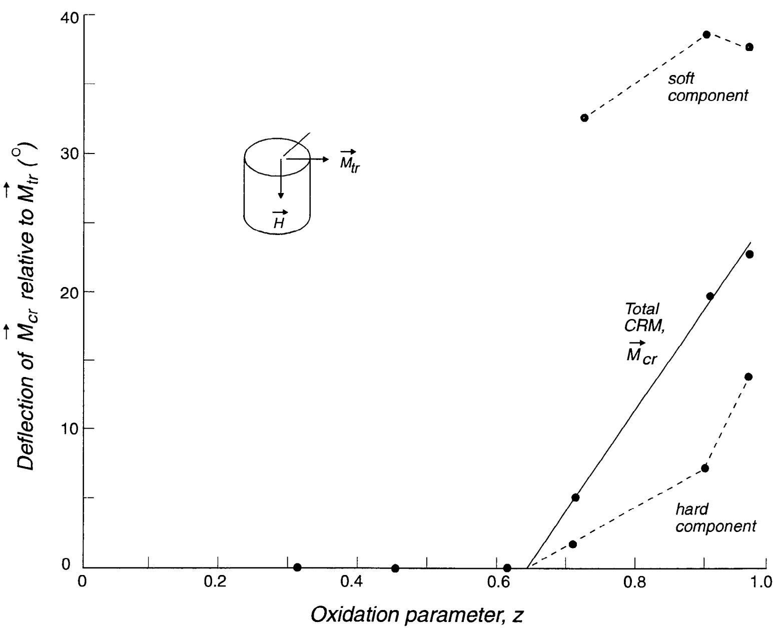

Figure 7. The direction of the CRM vector $\mathbf{M_{cr}}$ at different stages of in-air oxidation of SD titanomagnetite relative to the direction of the original TRM $\mathbf{M_{tr}}$. For $z < 0.65$, the oxidized phase is exchange coupled to the parent mineral and $\mathbf{M_{cr}}$ inherits the direction of $\mathbf{M_{tr}}$. For $z > 0.65$, there are multiphase oxidation products, exchange coupling is broken, and $\mathbf{M_{cr}}$ is increasingly deflected towards the field $\mathbf{H}$, which was applied perpendicular to $\mathbf{M_{tr}}$. After *Özdemir and Dunlop* [1985].

because of our need to understand complex magnetization structures in submicroscopic grains, micromagnetic calculations in geophysics have pulled ahead of those in magnetic recording. Our observations of larger grains have revealed surprises, such as metastable SD grains, which the physics community failed to uncover.

Most of the applications discussed in this paper relate to the paleomagnetic or magnetic anomaly record of igneous rocks. The standard paleomagnetic cleaning techniques and analytical procedures used in unravelling multivectorial NRMs are firmly based on our understanding of remanence blocking in SD grains. However, it is becoming increasingly clear that in MD grains, the blocking and unblocking of TRM (and undoubtedly of other types of NRM) are not straightforward reciprocal processes in the SD style. Reported failures of Néel theory to correctly describe thermoviscous overprinting seem to be due to non-SD carriers of NRM, not to a fundamental failing of the theory. CRMs too have very different properties depending on whether they are produced in SD or larger than SD grains.

A frontier that has been intermittently active over the years is taking paleomagnetism to the microscopic scale, making vector measurements of NRM on oriented individual mineral crystals. Although technically demanding, here we have the potential for demonstrating beyond doubt what mineral fractions carry different fractions of NRM and the probable origin of each. Electron microscopy and laser $^{40}Ar/^{39}Ar$ dating can be carried out on the same crystals, giving microstructural and age information to complement our knowledge of the nature and direction of each NRM component. Dated paleomagnetic poles for individual crystals are not too far away.

Finally, with new developments in techniques for measuring very small magnetic moments and for observing magnetic domains (or their counterparts in inhomogeneously magnetized grains) on a submicroscopic scale, e.g., the magnetic force microscope [*Williams et al.*, 1992b], we are entering an era where both the NRM of a specified grain and the magnetic configuration responsible for it will be observable. For rock magnetists, this is the most exciting prospect of all, the culmination of a long effort to understand the fundamentals of magnetic recording in nature.

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