

Modelling magnetization data using SIMPLEX

R. Thompson

Department of Geophysics, University of Edinburgh, James Clerk Maxwell Building, Mayfield Road, Edinburgh EH9 3JZ (Gt. Britain)

Thompson, R., 1986. Modelling magnetization data using SIMPLEX. *Phys. Earth Planet. Inter.*, 42: 113–127.

Two new approaches to modelling magnetization data are described. The first approach models the magnetic properties of natural materials in terms of mixtures of possible source materials. The second approach models the magnetic properties of natural materials in terms of mixtures of magnetite and haematite crystals of varying concentrations and grain sizes. In the second approach the magnetic characteristics of magnetite and haematite and their variation with grain size are described by bicubic spline functions. The methods are applied to remanence, and susceptibility measurements on a suite of lake sediment particle size fractions and on a set of samples from a soil profile.

1. Introduction

The most common minerals of the Earth's crust, quartz and feldspar, are diamagnetic. They form a practically non-magnetic matrix within which the less common magnetic minerals are scattered. In rocks, sediments and soils the most abundant of the magnetic minerals are the oxides and hydroxides of iron. Whilst the most familiar magnetic material, metallic iron, is rare in rocks it is not an uncommon industrial pollutant. Other naturally occurring magnetic minerals are the iron sulphides and the manganese oxides. In practical terms the magnetic properties of natural materials are largely connected with the element iron, its valency, concentration and partitioning.

Iron's abundance, geochemical properties, variable stability, sensitivity to redox changes and importance to Man make it a most interesting and important element in environmental studies. Magnetic studies offer a set of rapid, safe, non-destructive, and often extremely sensitive, techniques with which the mineralogy of iron bearing materials can be investigated.

The magnetic properties of rocks began to be studied in some detail when instrumentation was developed which was sensitive enough to be able to detect their weak natural permanent magnetiza-

tions. With the realization that rocks were capable of storing information about the ancient geomagnetic field, studies of natural magnetism blossomed into the wide ranging subject of palaeomagnetism, which has grown to be concerned with documenting the history of the Earth's magnetic field, as recorded by the remanent magnetization of rocks, throughout geological time. Arising from palaeomagnetic studies, work of major importance to the earth sciences has been done on the establishment of the polarity reversal time scale, on continental drift and on sea floor spreading.

Alongside palaeomagnetic work and investigation of the ancient geomagnetic field, magnetic investigations concerned with the crystallographic, mineral magnetic and solid state properties of natural magnetic minerals have also been carried out. Such rock magnetic and mineral magnetic investigations are often directed towards studying problems of particular importance to palaeomagnetism, for example, the origin and mechanism of thermoremanent magnetism, although rock magnetism is also pursued as a subject in its own right.

This paper describes some progress towards an approach to quantifying certain mineral magnetic properties of natural materials. The method allows the concentration and grain size of magnetic crystals in some natural samples to be estimated

and permits a more quantitative approach to magnetic differentiation, correlation, tracing and monitoring studies to be followed. At this stage of the work the range of variables in the models has been limited by working with materials dominated by the magnetic properties of multi domain and stable single domain magnetite and haematite. Furthermore it has been assumed that the magnetic minerals are stoichiometric.

2. Experimental aspects

The emphasis of experimental work in environmental magnetic studies (Thompson and Oldfield, 1986) has been on easy-to-measure magnetic properties, such as initial susceptibility and isothermal remanence.

Portable, battery-operated alternating current bridges for measuring initial susceptibility are commercially available at a modest cost, are extremely simple and quick to use, and have sensitivities (noise levels) of below 1×10^{-6} SI units. They operate continuously at frequencies of between 1 and 10 kHz with peak fields of about 0.1 mT. Great flexibility in the use of susceptibility equipment is provided by a range of alternative sensing heads which allow susceptibility measurements to be made both in the laboratory and the field. In the laboratory susceptibility measurements can be made on samples of a range of sizes, with masses varying from 0.1 to 100 g, whilst in the field outcrops, pits and boreholes can be studied.

Robust, portable, battery-operated fluxgate magnetometers with sensitivities (noise levels) below 10^{-4} Am^{-1} are also available commercially. These magnetometers allow isothermal remanence to be measured in a matter of seconds.

Equipment for magnetizing samples is also available commercially and is very straightforward and easy to use. Magnetic fields of up to 1 T can be conveniently generated by electromagnets or pulse discharge units. Minimal training or instrumental knowledge is needed to use any of the above portable equipment. Fields of between 1 and 13 T used in one experiment described below were generated in a superconducting solenoid. Fields in excess of 20 T require special power

sources (Rubin and Wolff, 1984), and while 280 T fields have been attained (Chikazumi et al., 1978), they are no longer non-destructive.

3. Magnetic mixtures

Sediments are made up of mixtures of rock fragments and authigenic minerals. Consequently they contain complicated assemblages of magnetic grains and a complete description of the magnetic content of a sediment is impractical. Nevertheless, magnetic parameters can be measured which can be used to characterise the average magnetic properties of a sediment and to indicate the relative concentrations of the main types of magnetic crystals. A wide variety of magnetic parameters are available for such characterisation studies.

3.1. Magnetite concentration

Perhaps one of the most useful magnetic parameters in environmental magnetic studies, and certainly one of the simplest and quickest to measure, is initial susceptibility, χ . It is found that for many magnetite-bearing sediment samples, susceptibility is a reasonable measure of magnetite concentration (e.g., Puranen, 1977; Currie and Bornhold, 1983): the volume fraction of magnetite in a sample being given by dividing the volume susceptibility (measured in SI units) by three. A typical volume susceptibility for recent sediments is 30×10^{-6} which corresponds to ten particles in every million consisting of magnetite. Examples of magnetite-bearing materials for which this relationship breaks down are those containing unusually large concentrations of haematite or goethite; those containing coarse multidomain grains; those containing superparamagnetic grains; and finally those materials in which magnetite is present in such low concentrations that the paramagnetism of weakly magnetic minerals makes a significant contribution to the initial susceptibility.

Susceptibility may vary with frequency (e.g., Snoek, 1948; Galt, 1952). At the frequencies used in a.c. bridges the 'in phase' susceptibility of magnetite grains lying on the superparamagnetic/stable single domain boundary decreases with increasing frequency (Smit and Wijn, 1954), whereas

the susceptibility of multidomain and single domain grains shows little change (Bhathal and Stacey, 1969). Consequently, frequency dependent susceptibility, or the associated parameter quadrature susceptibility, can be used to assess the concentration of magnetite grains lying on the superparamagnetic/stable single domain boundary (Thompson and Oldfield, 1986).

3.2. Magnetite grain size

The intrinsic magnetic properties of Curie temperature, saturation magnetization and magnetocrystalline anisotropy are independent of grain size whereas magnetic parameters associated with the magnetization process e.g., susceptibility, coercive force, coercivity of remanence, isothermal, and anhysteretic remanence and rotational hysteresis loss depend on grain size on account of extrinsic influences (O'Reilly, 1984). All of these latter parameters are of potential use in grain size studies.

One particularly easily measured magnetic parameter which varies with grain size is saturation isothermal remanence, *SIRM*. Often in practice an isothermal remanence grown in the highest field easily available, e.g., 1 T (IRM_{1T}) is measured rather than the strict saturation remanence. Saturation remanence is highest for single domain grains. It decreases steadily with increasing grain size (e.g., O'Reilly, 1984) and is zero for superparamagnetic grains. Normalizing saturation remanence by initial susceptibility allows the variation of magnetic concentrations to be largely taken into account, so providing a magnetic granulometric parameter (Thompson et al., 1980). This parameter IRM_{1T}/χ has been found to be a very helpful magnetic parameter in investigating recent sediments and soils (Thompson and Oldfield, 1986).

The IRM_{1T}/χ ratio decreases with magnetic grain size from 100 kAm^{-1} for large multi-domain grains. Figure 1 plots this variation in IRM_{1T}/χ ratio which is based on the measurements of Parry (1965), Dunlop (1973), Dankers (1978) and the author (unpublished data) on sized synthetic and natural magnetite crystals. The mean IRM_{1T}/χ ratio calculated from measurements on 1000 natu-

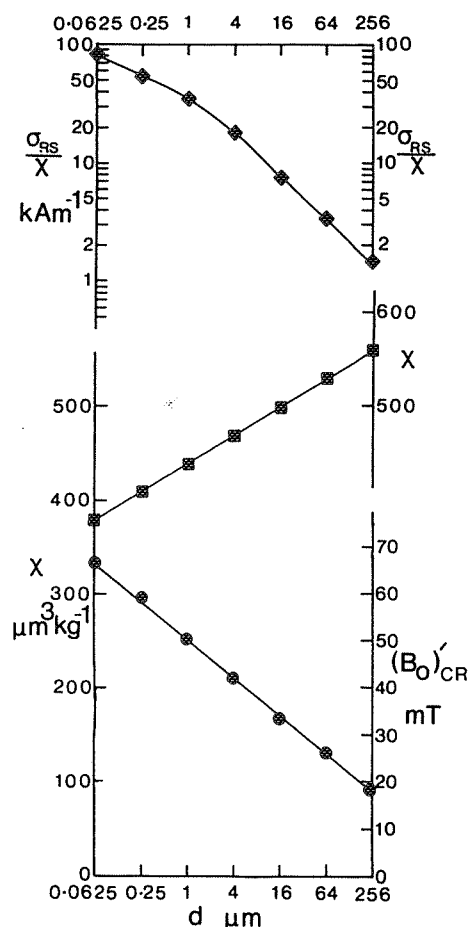


Fig. 1. Idealized diagram of the variation of acquisition coercivity of remanence, $(B_0)_{CR}$ susceptibility, χ , and saturation remanence to susceptibility ratio, σ_{RS}/χ , with grain size for magnetite crystals.

ral materials was found to be 13 kAm^{-1} (Thompson and Oldfield, 1986). The IRM_{1T}/χ ratio has an interquartile range from about 7 to 20 kAm^{-1} with a skewed log normal distribution.

Dunlop (1983) reviewed, in detail, the potential of a wide variety of parameters in magnetic granulometric work by applying them to a collection of carefully chosen igneous rocks. In addition to the traditional magnetic properties used in domain structure determination of coercive force $(B_0)_C$, median destructive field, *MDF*, and the ratio of saturation remanence to saturation magnetization, IRM_{1T}/M_S , a number of other rather

more complicated and less used parameters were also tested in Dunlop's study. Dunlop assessed the value of the various granulometric parameters by plotting correlation diagrams of their variation with coercive force for three series of basaltic rocks. Parameters such as (1) median destructive field, (2) the ratio of saturation remanence to saturation magnetization, and (3) the ratio of the susceptibility of anhysteretic remanence to saturation remanence, which all showed a strong correlation with coercive force were concluded by Dunlop to be of value in magnetic granulometric studies, whereas parameters which showed weak or non-existent correlation, such as (4) the ratio of coercivity of remanence to coercive force and (5) the Lowrie-Fuller parameter, were concluded to be of little value.

Dunlop did not consider the ratio of saturation remanence to susceptibility. His comprehensive study, however, allows IRM_{1T}/χ ratios to be calculated. Figure 2 is an IRM_{1T}/χ versus $(B_0)_C$ correlation diagram constructed using the data from Dunlop's three sets of basaltic rocks. It was assumed in producing Fig. 2 that Dunlop's (1983) susceptibility measurements had been normalized by saturation magnetization in order that they could be expressed in terms of unit volume of magnetite. A strong correlation is revealed in Fig. 2 between IRM_{1T}/χ ratio and $(B_0)_C$ confirming the suggestions of Thompson et al. (1980) that IRM_{1T}/χ ratio is potentially useful in magnetic granulometric studies.

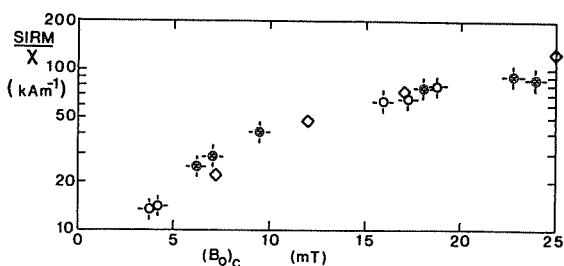


Fig. 2. Correlation diagram for saturation remanence to susceptibility ratio, $IRM_{2,3T}/\chi$ with coercive force, $(B_0)_C$ for three series of basaltic rocks described in Dunlop (1983). Open circles, Icelandic basalts; closed circles, Steen's Mountain basalts; open diamonds, Coronation sills series.

3.3. Other magnetic minerals

Haematite, goethite and pyrrhotite display different magnetic properties from those of magnetite. Collinson (1967, 1968a,b) discussed the magnetic properties of haematite in red bed sediments and described one approach to estimating haematite content by magnetic analysis. His technique involves calculating haematite concentration from measurements of high field magnetization, a paramagnetic correction to the high field magnetization measurements having been made based on spectrophotometric determinations of ferrous and ferric iron content.

Isothermal remanence acquired in fields greater than 0.1 T provides another magnetic technique for detecting the presence of haematite crystals (e.g., Dunlop, 1972). High field isothermal remanence acquisition, $HIRM$ ($=IRM_{1T} - IRM_{0.1T}$), has been used by Bradshaw and Thompson (1985) and Hiron and Thompson (1985) as a very rough indicator of haematite concentration. Ratios of moderate field IRM to saturation IRM , for example the S -ratio of Stober and Thompson (1979), have also proved useful in classifying samples with mixed magnetic mineralogies (e.g., Oldfield et al., 1983) and in detecting samples with high goethite or haematite concentrations.

The magnetically stable minerals haematite and goethite both display very high IRM_{1T}/χ ratios. The mineral pyrrhotite also yields high IRM_{1T}/χ ratios (Clark, 1984). So in addition to its use as a granulometric tool for magnetite bearing samples, IRM_{1T}/χ ratio may also be of some value in determining mineral types (Thompson et al., 1980) in magnetic mixtures. Bradshaw and Thompson (1985) have described how IRM_{1T}/χ ratio measurements can be graphically combined with coercivity of remanence $(B_0)_{CR}$ measurements to investigate the relative importance of various types of magnetic minerals in lake and soil samples.

As an example of the IRM/χ versus $(B_0)_{CR}$ approach to assessing magnetic mixtures the method has been applied to Dunlop's (1983) data set for igneous rocks. Plotting Dunlop's results on the IRM/χ versus $(B_0)_{CR}$ diagram of Bradshaw and Thompson (1985) yields Fig. 3. The anticipated sweep of points for magnetites of decreas-

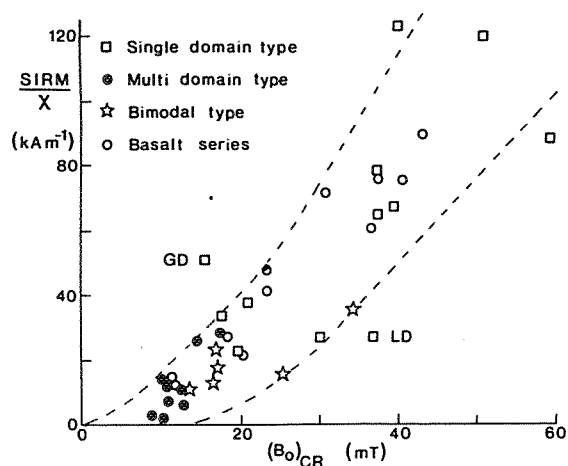


Fig. 3. Saturation remanence to susceptibility ratio, $IRM_{2.3T}/\chi$, versus coercivity of remanence $(B_0)_{CR}$ for igneous rocks described in Dunlop (1983). Multi-domain magnetite bearing rocks plot in lower left hand corner. Two samples, GD and LD fall outside the main trend. This difference indicates that these two samples probably contain more complicated magnetic mixtures compared with the rest of the igneous rocks in the study. See text for further details.

ing grain size is seen running from the multi-domain rocks which plot in the lower left hand corner of the diagram through to the stable single domain rocks which plot towards the top right of the diagram. The bimodal rocks of Dunlop's (1983) classification span the multidomain/single domain divide as do the basalt series.

Two samples, a Glamorgan diabase, GD, and a Logan diabase, LD, stand out from the main sweep of points in Fig. 3. The unusually high $IRM_{2.3T}/\chi$ ratio of over 50 kAm^{-1} for the 16 mT coercivity of remanence of the Glamorgan diabase sample could be caused by the presence of pyrrhotite grains in addition to magnetite grains. Pyrrhotite has been reported in samples from this suite of rocks (Dunlop, 1983) but has not been noted in this particular sample. The Logan diabase sample is distinct as it combines a low $IRM_{2.3T}/\chi$ ratio of 27 kAm^{-1} with its 37 mT coercivity of remanence. This pair of magnetic properties could be caused by an important contribution to the susceptibility of the diabase by paramagnetic or superparamagnetic grains.

Summarizing this section on magnetic mixtures

—in many environmental studies we have found: (1) susceptibility to be a useful, rapid first estimate of magnetite concentration, (2) IRM_{1T}/χ ratio to be a quick magnetic granulometric measure, and (3) IRM_{1T}/χ ratio versus $(B_0)_{CR}$ plots to be a simple graphical method of recognising magnetic mixtures. Our approach appears to tie in well with Dunlop's (1983) studies of igneous rocks. In particular: (1) Dunlop found little variation in the susceptibility of magnetite with grain size except for an increase at very large grain sizes (see fig. 7b in Dunlop, 1983), so confirming the value of susceptibility measurements for estimating magnetite concentration, (2) the simple, quantitative parameter IRM_{1T}/χ ratio has been shown using Dunlop's igneous rock data, to have a clear, continuous variation with coercive force $(B_0)_C$ (compare Fig. 2 with fig. 7a in Dunlop, 1983) so confirming the value of IRM_{1T}/χ ratio as a granulometric indicator, and (3) graphical comparison of IRM_{1T}/χ ratio with $(B_0)_{CR}$ appears to be able to contribute information about magnetite grain size and about magnetic mixtures in igneous rocks as well as in recent sediments and soils.

Although the above mineral magnetic techniques based around susceptibility and isothermal remanence measurements have been used in many environmentally motivated studies (Thompson and Oldfield, 1986), a more quantitative, mathematical approach appears to be desirable. The following section discusses two such mathematical approaches to extending, formalizing and quantifying investigations of mineral magnetic properties of natural materials.

4. Modelling hysteresis properties

The magnetic hysteresis properties of crystals when combined can produce strange looking hysteresis loops such as the constricted (wasp-waisted) loops of mixtures of magnetically hard and soft materials (Wasilewski, 1973). Fortunately, however, the combination of discrete (non-interacting) mixtures is mathematically straightforward. Kneller and Luborsky (1963), for example, calculated mathematically the coercive force of mixtures of hard and soft grains, and were able to

demonstrate how a small addition of soft materials can bring about a pronounced reduction in coercive force in a mixture and how hysteresis loop constriction can arise from mixing.

Two approaches to mathematically modelling remanence hysteresis data are described and applied to measurements on lake sediment and soil samples with mixed magnetic mineralogies.

4.1. Mass mixing model

In this first approach the magnetic hysteresis characteristics of a sediment are modelled in terms of mixtures of 'source' materials. In a second approach, described below, mixtures of crystals are analysed. The magnetic characteristics of possible source materials are established by experiment and then combined mathematically to yield the magnetic properties of different mixtures. The problem at hand is one of finding the best combination of source materials to account for the magnetic characteristics of the sediment under investigation.

As a first step towards solving the mixing problem a set of m simultaneous linear equations can be formed and written as $Ax = b$, where the number of columns, n , of the m by n , known matrix A is equal to the number of possible source materials, and the number of rows of the matrix, m , is equal to the number of measured magnetic parameters. The elements of the right hand vector b (with m rows and one column) are the magnetic measurements on the sediment and the elements of the matrix A are the magnetic measurements on the various source materials. The elements of the required solution vector x (with n rows and one column) are the unknown relative concentrations of the source materials. Now if m is greater than n , then a least squares solution to the equations can be found; i.e., the best attainable values of the unknown quantities are taken to be those that distribute the residuals to each of the observation equations according to Gauss's law of error and which minimize the length of the residual vector $r = b - Ax$. For a matrix of full rank the minimal least squares solution of the problem $Ax = b$ can be found by any standard linear algebraic method. In our mass mixing model we require the elements

of x to be non-negative and we also might like the elements to sum to 100%. This means that a constrained optimization approach needs to be applied.

4.2. Simplex

A suitable direct search approach to the minimization problem posed above proves to be the simplex search method of Nedler and Mead (1965). An excellent implementation of their simplex method for minimizing a function is to be found in the NAG library as routine EO4CFF. Although computationally slower than other non-linear least squares search algorithms such as steepest descent or Newton-Raphson methods, divergence is impossible in the simplex method. Also in the simplex method no knowledge is required of the first derivatives of the response surface. Boundary (or contour) conditions can easily be added as required, simply by giving the objective function a very large value whenever the constraints are violated. Similarly equations of conditions can also be added without difficulty. Furthermore the criterion used in judging the quality of fit of the model and data can easily be adjusted.

A simplex is a geometric shape that has one more corner (or vertex) than the space in which it is defined has dimensions (n). For example, in two dimensions the vertex is a triangle. To find the minimum residual sum of squares the algorithm rolls the simplex 'downhill' from its starting position. The rate and direction of movement of the simplex are controlled by the algorithm through expansion, contraction and reflection of the vertexes (Caceci and Cacheris, 1984). The only information required to be supplied by the user, in addition to the data used in forming the simultaneous linear equations is (1) an initial guess of each model parameter (n values), (2) the maximum error allowed (typically 10^{-5}) and (3) the maximum number of iterations (very roughly around $20 \times n^2$).

In calculating mass mixing models by simplex minimization the model parameters can be forced to be non-negative by including a statement in the goodness of fit algorithm which assigns high residual sum of square values to out-of-boundary model

parameters. Furthermore the model parameters can be constrained to total 100% by elimination of one variable.

A potential source of difficulty with all constrained optimization algorithms is the problem of local minima (e.g., Bunday, 1984). One approach used here in an effort to avoid local minima, in addition to the methods used by the NAG implementation, is to compare the outcomes of the simplex minimization for a number of starting positions. One such starting position is the unconstrained least squares solution found directly by linear algebra. In practice, however, no problems related to local minima were encountered during development or testing of the mass mixing algorithm.

4.3. Crystal mixing model

In this second approach the magnetic characteristics of a sample are modelled in terms of mixtures of crystals. The magnetic properties of many natural materials are dominated by their constituent magnetite and haematite crystals. The crystal mixing approach has been developed for these materials.

The magnetic properties of magnetite and haematite crystals of various grain sizes are tabulated in Table I. The bottom row of Table I lists the variation of susceptibility with grain size. The second to bottom row lists the variation of saturation remanent magnetization. The remaining rows tabulate normalized acquisition of isothermal remanence for a series of increasing field strengths. The normalized isothermal remanences increase from zero in the demagnetized state to one in a field of 1 T. Table I is an idealized compilation of magnetic properties based on magnetic measurements of natural crystals (e.g., Dankers, 1978), synthetic crystals (e.g., review by O'Reilly, 1984), natural samples (author's unpublished data) and theoretical considerations. This idealized, empirically derived, acquisition of isothermal remanence data of Table I is plotted as sets of curves in Figs. 4 and 5.

Least squares bicubic spline functions (Hayes and Halliday, 1974) are used to interpolate between the Table I data points. Interpolation allows

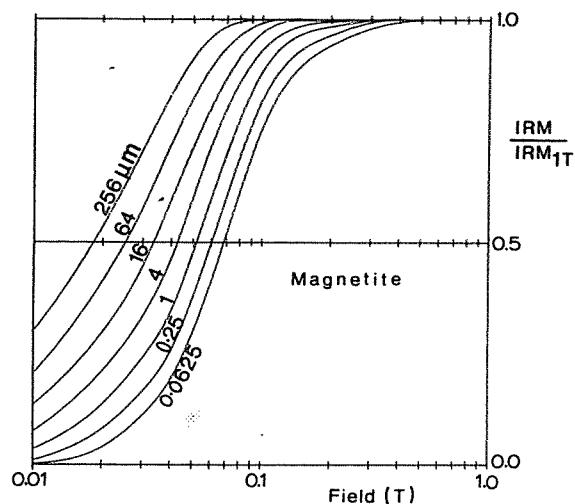


Fig. 4. Empirically derived, idealized normalized acquisition of isothermal remanent magnetization curves for magnetite of grain diameters 0.0625, 0.25, 1, 4, 16, 64 and 256 μm . The curves are spline fits to the data of Table I. Peak field of 1 T used for normalization.

the isothermal remanence acquired at any field strength by magnetic crystals or haematite crystals of any grain size (within the limits of the outer columns of Table I), to be calculated. The NAG routine E02DAF is used in fitting the bicubic

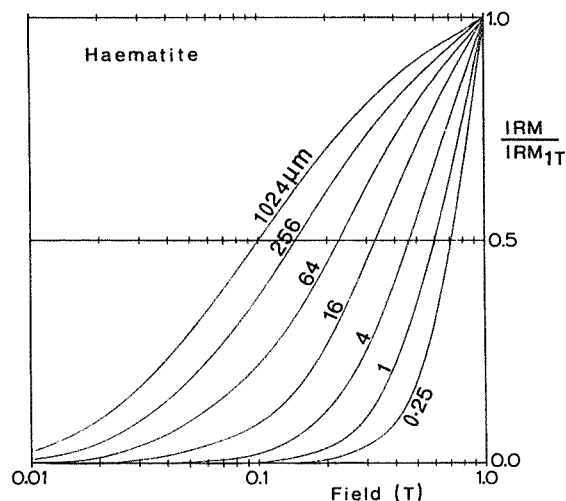


Fig. 5. Empirically derived, idealized normalized acquisition of isothermal remanent magnetization curves for haematite of grain diameters 0.25, 1, 4, 16, 64, 256 and 1024 μm . The curves are spline fits to the data of Table I. Peak field of 1 T used for normalization.

TABLE I
Isothermal remanence and initial susceptibility properties of magnetite and haematite

Field (mT)	Haematite					Magnetite								
	Grain size					Grain size								
	0.250	1.000	4.000	16.00	(μm)	256.0	1024.	0.0625	0.250	1.000	4.000	16.00	64.00	256.0
0	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
1	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.002	0.005
5	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.001	0.003	0.020	0.050	0.100	0.150
10	0.000	0.000	0.000	0.000	0.000	0.001	0.008	0.002	0.010	0.035	0.075	0.135	0.205	0.300
20	0.000	0.000	0.000	0.000	0.000	0.010	0.047	0.039	0.080	0.135	0.210	0.297	0.412	0.535
30	0.000	0.000	0.000	0.008	0.008	0.038	0.095	0.110	0.163	0.239	0.332	0.445	0.577	0.707
40	0.000	0.000	0.000	0.018	0.018	0.065	0.145	0.191	0.260	0.345	0.462	0.596	0.722	0.829
50	0.000	0.000	0.001	0.030	0.030	0.093	0.185	0.300	0.368	0.475	0.600	0.716	0.822	0.917
60	0.000	0.000	0.003	0.046	0.046	0.120	0.233	0.407	0.500	0.600	0.723	0.800	0.895	0.958
80	0.000	0.001	0.007	0.065	0.065	0.170	0.310	0.620	0.698	0.780	0.854	0.920	0.963	0.995
100	0.000	0.002	0.024	0.090	0.090	0.222	0.375	0.752	0.814	0.880	0.935	0.970	0.994	0.999
150	0.000	0.018	0.057	0.172	0.172	0.345	0.500	0.900	0.935	0.970	0.985	0.995	0.998	1.000
200	0.000	0.042	0.115	0.265	0.265	0.460	0.605	0.942	0.965	0.983	0.996	0.999	1.000	1.000
300	0.007	0.110	0.250	0.452	0.452	0.630	0.725	0.980	0.995	0.999	1.000	1.000	1.000	1.000
400	0.037	0.225	0.405	0.603	0.603	0.737	0.807	0.999	1.000	1.000	1.000	1.000	1.000	1.000
500	0.092	0.355	0.555	0.710	0.710	0.807	0.865	1.000	1.000	1.000	1.000	1.000	1.000	1.000
600	0.185	0.500	0.677	0.790	0.790	0.865	0.900	1.000	1.000	1.000	1.000	1.000	1.000	1.000
800	0.320	0.755	0.860	0.910	0.910	0.945	0.955	1.000	1.000	1.000	1.000	1.000	1.000	1.000
900	0.650	0.900	0.935	0.957	0.957	0.972	0.981	1.000	1.000	1.000	1.000	1.000	1.000	1.000
IRM _{IT}	0.295	0.268	0.243	0.224	0.224	0.216	0.211	30.50	22.50	15.30	08.48	03.60	1.765	0.891
χ μm ³ kg ⁻¹	0.600	0.600	0.600	0.600	0.600	0.600	0.600	380.0	410.0	440.0	470.0	500.0	530.0	560.0

Isothermal remanence acquired in fields between 1 and 900 mT is listed as a proportion of the isothermal remanence grown on a 1T field for magnetite grain sizes between 0.0625 and 256 μm and haematite grain sizes between 0.25 and 1024 μm . The initial reversible susceptibility per unit mass (χ) and isothermal remanence per unit mass (IRM_{IT}) are also listed for each grain size of magnetite and haematite.

spline surfaces, in terms of B-splines (de Boor, 1978), and the sub-routine E02DBF is used in calculating the value of the splines at any point of interest. A regularly spaced ten by five array of internal knots is used in fitting the magnetite data and a regularly spaced 15 by five array of internal knots is used in fitting the haematite data. The knots can be thought of as dividing the data regions into panels. Most of the computation time spent in calculating crystal mixtures models occurs in the evaluation routine E02DBF. The time involved is approximately proportional to the number of panels. So the number of knots as well as controlling the goodness of fit and the smoothness of the approximating surface also controls the computation time. The chosen ten by five and 15 by five arrays of knots represent a balance between these three competing factors.

The crystal mixture model which is best able to account for the magnetic characteristics of a sample can be found through the simplex, linear programming algorithm described above. The unknown model parameters are (1) magnetite concentration, (2) haematite concentration, (3) magnetite grain size, and (4) haematite grain size. Starting from an initial guess such as the unconstrained linear algebra solution calculated by solving the normal equations, the simplex algorithm iterates to the best solution by trying different crystal mixtures. The out-of-boundary conditions are that the magnetite and haematite concentrations should be non negative—and that the grain sizes should lie between the extreme grain diameters of Table I. In practice it helps to set the out-of-boundary grain size diameters to lie within the range of the empirical data rather than at the extreme edges of the data (e.g., by including additional grain size data) as this arrangement avoids possible local minima problems connected with end points of the spline fitting. An alternative approach would be to use the complex method of Box (1965) which allows the search to move along the constraints.

4.4. Error assessment

It is desirable in addition to calculating a best fitting model to be able to provide an estimate of

the errors associated with the fitting calculation. One approach to error assessment is to investigate the range of variation of the model parameters about the best fit solution that still permit a good fit to the experimental data. Goodness of fit can be quantified and appraised by a chi-squared test and the range of permitted variation can be mapped out by a grid search, contour tracing algorithm, or random Monte Carlo search strategy. A chi-squared test combined with a Monte Carlo search routine has been successfully used with both the mass mixing and crystal mixing models. Although random search techniques considerably increase programme running time they provide a further method of detecting and escaping from a local minimum in addition to their role in error assessment.

A second approach to error assessment is sensitivity analysis. Simulated data sets can be created by adding pseudo-random errors to the original experimental data: the pseudo-random errors being drawn from a normal distribution with zero mean and with variance the same as that of the residuals about the best fitting model. When formed the simulated data sets are processed in the same way as the original experimental data. The variation in model parameters as calculated from these simulated data sets provides an estimate of the errors associated with the fitting of the original experimental data.

4.5. Measurements and weighting

Any characteristics that can be measured quantitatively act as conservative tracers and combined linearly can be used in mass mixing modelling. For example, remanent magnetization measurements can be used, as can initial and frequency dependent susceptibility measurements, magnetizations such as measurements taken from major hysteresis loops, anhysteretic remanence measurements and partially demagnetized remanences. Chemical and mineralogical measurements can equally be used to contribute to the matrix of observations.

Several different weighting schemes can be used. The weights associated with each row of the observation matrix A and the row vector b can be

varied according to the personal assessments of the observer, kept natural, or adjusted to pertain to fractional errors. In the following two examples from Loch Catherine and Quatford fractional error weighting has been used for all of the experimental observations. Also in the Loch Catherine example remanent magnetization differences between successive magnetization or demagnetization steps were used rather than individual remanence measurements in forming the observation matrix.

4.5.1. Loch Catherine

A horizon of sandy silt from the surface core LCM5 from Loch Catherine (Thompson and Edwards, 1982) was chosen to investigate the mass mixing approach to accounting for magnetic characteristics. Figure 6b plots bulk sample isothermal remanence data from the 8–22 cm deep sandy silt layer which, in this example, is simply to be modelled as a combination of the isothermal remanences of five particle size splits. The splits were obtained by wet sieving (Stewart, 1979). This particular sediment layer was possibly deposited as a result of disturbances caused by road building close to the side of the lake.

IRM acquisition was measured for 13 field applications of increasingly strong fields. Graphs of the acquisition of *IRM* of the five particle size splits are plotted in Fig. 6a. Note the overall increase in magnetic stability with particle size from $0.5 \text{ mAm}^2\text{kg}^{-1}$ for sediment of less than $32 \mu\text{m}$ diameter to $13.4 \text{ mAm}^2\text{kg}^{-1}$ for sediment over 0.25 mm in diameter. The diagonal crosses in Fig. 6b mark the acquisition of *IRM* to a saturation intensity of $1.7 \text{ mAm}^2\text{kg}^{-1}$ for the bulk sample, D.

Sixteen measurements on each of the five particle size splits were used in forming the observation matrix *A* for the mass mixing modelling. Firstly, 13 remanence acquisition measurements were used to calculate 13 remanence increments for each of the five particle size fractions and for sample D. The resulting increments were used to provide the elements of the first 13 rows of matrix *A* and vector *b*. Secondly, initial susceptibility measurements, anhysteretic remanences (imparted statically in direct field of 0.1 mT using a 0.1T peak alternating field) demagnetised by 30 mT

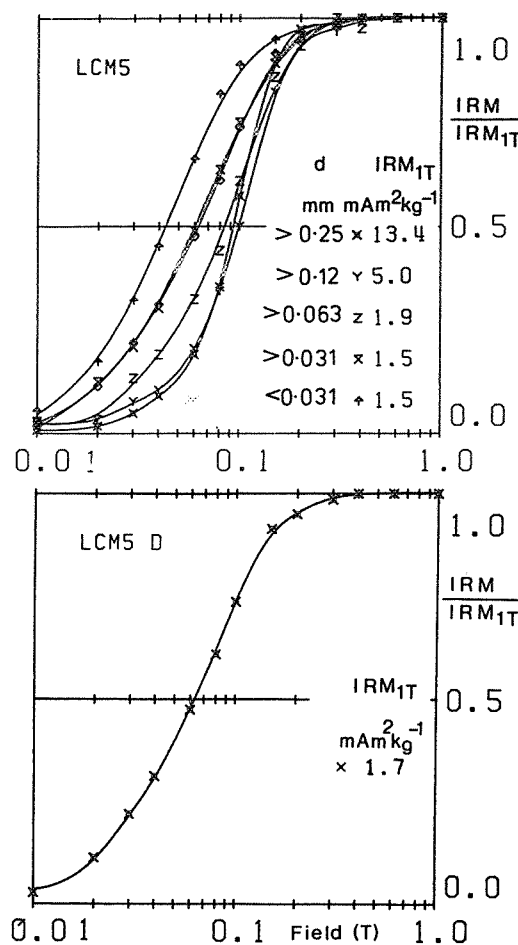


Fig. 6. Acquisition of isothermal remanent magnetization in increasing magnetizing fields for material from core M5 from Lough Catherine. (a) Five particle size fractions from the layer at 8–22 cm depth. (b) Bulk material at 15 cm depth. The curve in (b) is a modelled acquisition of remanence curve constructed from the particle size data.

alternating fields and anhysteretic remanences unaltered by the 30 mT fields were also included. These measurements were used to fill the last three rows of the 16 by five observation matrix and right hand vector *b*.

A best fit mass mixing model with contributions of 1%, 0%, 26%, 36% and 37% for the five particle sizes 2, 3, 4, 5, and $> 5\phi$ was found using the simplex minimization approach outlined above. The curve in Fig. 6b plots acquisition of isothermal remanence as calculated from this best fitting

mass mixing model. This best fit model can be very roughly checked by comparing it with the sediment particle size distribution of the layer. Weighing the sediment held on each sieve in the wet sieving work revealed that the layer was indeed dominantly composed of sediment in the 4, 5 and $> 5\phi$ particle size fractions. This mass mixing approach to modelling magnetization data is now being applied to more difficult problems involving sediment and soil provenance.

4.5.2. Quatford

Three soil samples from woodland at Bowman's Coppice near Quatford in Shropshire have been analysed using the crystal mixing model. The woodland, situated on a Triassic red sandstone escarpment, was previously part of the ancient forest of Morfe. Table II summarizes various mineral magnetic properties of the Quatford soil samples and in the right hand columns lists the results of the crystal mixing modelling. Normalized remanence acquisition data, a best fitting curve and eight curves derived by the sensitivity error analysis described above are plotted in Fig. 7 for the three soil samples.

A clear contrast can be seen between the top-soil and the two lower soil samples. The top-soil reveals the usual enhanced susceptibility of many temperate and subtropical top-soils, brought about by an increased ferrimagnetic component. A superparamagnetic component is revealed in all three samples by the frequency dependence of the susceptibility. The Quatford top soil has a frequency dependence of 3% per decade. Slight viscous magnetic changes are also detectable in the Quatford topsoil. For example, the $IRM_{0.1T}$ was found to decrease by 2% during 24 h zero field storage.

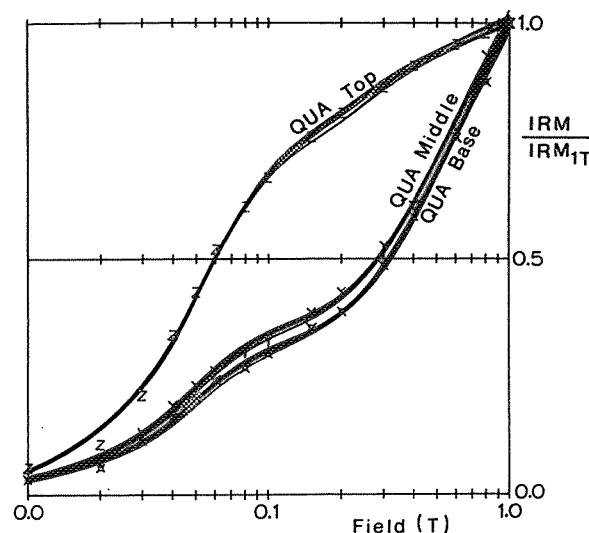


Fig. 7. Acquisition of isothermal remanent magnetization curves for three soil samples from Quatford. The curves are modelled acquisition of remanence curves based on the data of Table I. The curve envelopes are based on the result of simulation experiments which were designed to provide some information about the fitting errors involved in the modelling. See text for further details.

Similar small viscous changes were found in the lower two samples. These viscous changes and frequency dependence of susceptibility together indicate that even the basal soil samples are probably magnetically enhanced compared with unweathered, haematite-rich, Triassic bedrock, as pronounced frequency dependence of susceptibility is rare in unweathered rocks.

A magnetization hysteresis loop for the basal Quatford soil sample is shown in Fig. 8. The loop was traced out on cycling the field between plus and minus 1.07 T by a X - Y plotter attached to a

TABLE II

Quatford soil profile selected measured and modelled properties

Sample	χ ($\mu\text{m}^3\text{kg}^{-1}$)	IRM_{1T} ($\text{mA}\text{m}^2\text{kg}^{-1}$)	IRM_{20T} ($\text{mA}\text{m}^2\text{kg}^{-1}$)	$(B_0)_{CR}$ (mT)	'Magnetite'		'Haematite'	
					Concentration (mg/g)	Grain size (μm)	Concentration (mg/g)	Grain size (μm)
QUA1 top	0.18	3.34	3.61	43	$0.26^{+0.02}_{-0.04}$	$4.9^{+2.1}_{-0.9}$	$6.5^{+1.7}_{-1.1}$	124^{+357}_{-95}
Middle	0.023	0.78	1.46	190	$0.050^{+0.007}_{-0.016}$	$9.3^{+2.2}_{-3.9}$	$2.2^{+0.1}_{-0.1}$	$4.7^{+2.1}_{-0.8}$
Bottom	0.020	1.02	1.75	180	$0.052^{+0.004}_{-0.011}$	$7.7^{+1.0}_{-2.7}$	$2.9^{+0.1}_{-0.1}$	$4.3^{+1.3}_{-1.7}$

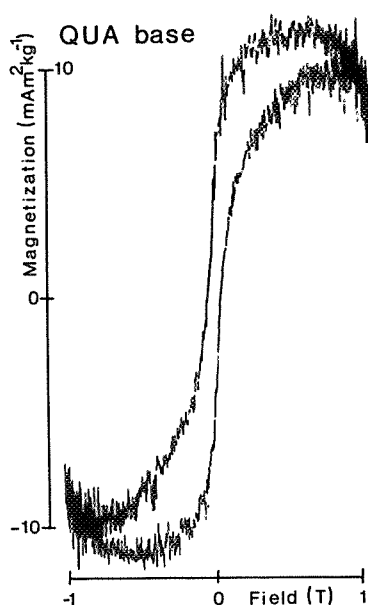


Fig. 8. Hysteresis loop measured on a vibrating sample magnetometer for the basal Quatford soil sample. The constricted form of the loop results from the presence of both magnetite and haematite in the soil sample. The tails of the loop reveal a diamagnetic component, which is probably an instrumental effect (see text) enhanced by the relatively low magnetization of the soil sample. The high frequency noise is present because the instrument had to be run at a very sensitive level (90 dB with 300 mS averaging time) to detect the weak magnetization of the sample.

vibrating sample magnetometer. The hysteresis loop is very noisy on account of the low concentrations of magnetic minerals in the subsoil. The constriction in the hysteresis loop is caused by the presence of both ferrimagnetic and imperfect antiferromagnetic minerals with their contrasting stabilities as revealed by the crystal mixing modelling of the isothermal remanence data. The decrease in magnetization in fields above 0.5 T is caused by the diamagnetism of the sample holder and sample rod.

The crystal modelling assesses the 'magnetite' enhancement in the Quatford soil profile between the top and bottom samples to be a factor of five in concentration with no significant change in average magnetite grain size. The magnetite grain size estimates are likely to be overestimates as the crystal mixing model, in its present form, does not take into account low coercivity grains just above

the superparamagnetic boundary. Stephenson (1971) described one approach to investigating such grain size distributions through Néel's (1955) theory of the variation of blocking volume with temperature. The 'haematite' modelling of the Quatford samples is also unsatisfactory. Very large fitting errors are associated with the 'grain size' estimate of the top-soil haematite. Furthermore, the haematite concentration errors are much larger for the top-soil sample. These large formal fitting errors are possibly connected with the difficulty that interacting single domain magnetite crystals do not saturate easily. Strongly interacting magnetite particles are not fully described by the data of Table I with the consequence that the crystal modelling procedure in its present form is unable to distinguish satisfactorily between coarse grained haematite and interacting fine grained magnetite. Measurements of remanence acquisition in fields of excess 1T might allow better modelling of haematite constituents to be performed. Although the modelled curves of Fig. 7 fit the *IRM* data quite well, and the formal concentration fitting errors of Table II for the Quatford samples are small, the grain size estimates are not reliable and point to several improvements needed in the crystal mixture modelling approach.

4.6. Interaction fields

Interaction fields have been found to be of importance in some natural materials (Dunlop and West, 1969; Cisowski, 1981) and in synthetic iron oxides aggregates (Eldridge, 1964; Jaep, 1969). Indeed according to certain theories (Eldridge, 1964) interaction fields may dominate in determining some bulk magnetic properties such as anhysteretic magnetization. In soils and lake sediments low interaction fields associated with grains slightly larger than superparamagnetic in size could lead to unusually high anhysteretic remanences.

The crystal mixing and mass mixing modelling procedures assume that sediment magnetic hysteresis parameters can be accounted for by linear combinations of the magnetic properties of their constituents. This linearity assumption does not hold in the case of varying interaction fields. Physical clustering of magnetic grains can lead to

increased interaction fields which can have the effect of making demagnetization easier but of making magnetization harder. This means that the magnetic hysteresis properties of clustered grains are not only dependent on the magnetic properties of the grains but also on their degree of dispersion. Magnetic modelling procedures can be expected to perform better when interaction fields are small, or when interaction fields are similar in all the samples under investigation.

One approach to investigating interaction fields is to construct Preisach diagrams (Preisach, 1935; Bate, 1962; Mullins, 1974). A second approach is to compare experimentally derived magnetization acquisition and demagnetization characteristics with comparable theoretical characteristics such as those derived by Wohlfarth (1958). A limited number of experiments have been performed to investigate the importance of interaction fields using this second approach. After Cisowski (1981), isothermal remanence acquisition has been analysed along with saturation remanence alternating field demagnetization. The ratio R , of the saturation remanence demagnetized in alternating fields as high as the remanent coercive force $(B_0)_{RC}$ to the undemagnetized saturation remanence is used as a measure of the interaction fields (see Fig. 9). An average R ratio of 0.32 and a range of R ratios from 0.26 to 0.39 was found for the ferrimagnetic fraction of 39 lake sediments, deep-sea sediments, shallow marine sediments, soils and peats. Wohlfarth (1958) pointed out that non-interacting, uniaxial, single-domain grains should yield R ratios of 0.5. This theoretical R ratio is significantly higher than the above R ratios of recent sediments and soils. Such deviations from the ideal 0.5 R ratio could be caused by non-uniaxial anisotropy and superparamagnetic or multidomain effects as well as by grain interactions.

Natural multidomain magnetites can certainly display low R ratios. For example, measurements by the author on a Hebridean metamorphic rock (a magnetite-bearing greenschist with a remanent coercive force of 10 mT), and on an igneous rock from Glen Doll (a diorite with a remanent coercive force of 6 mT), both of which contained large, clearly visible magnetite crystals yielded R ratios of 0.24 and 0.23, respectively.

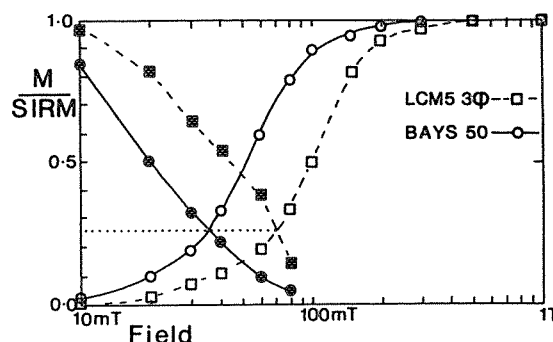


Fig. 9. Acquisition of isothermal remanence (open symbols) plotted along with alternating field demagnetization (solid symbols) of the saturation remanence for the 3ϕ particle size fraction of a layer 8–22 cm deep in core M5 from Lough Catherine and for a Lough Neagh sediment from core BAYS 50. The intersection of the Catherine magnetization and demagnetization curves yields an R ratio of 0.26 and a remanent coercive force, $(B_0)_{RC}$ of 70 mT. The intersection of the two Neagh curves yields an R ratio of 0.26 and $(B_0)_{RC}$ value of 35 mT. The low R ratios in both samples are interpreted as resulting from grain interactions.

In Fig. 9 we see that both the 3ϕ Loch Catherine particle size split and a bulk sample of Lough Neagh sediment from core BAYS 50 have R ratios of 0.26. The moderate stability but low R ratio of the Lough Neagh sediment is most probably caused by interacting single domain grains. Similarly the Loch Catherine sediment is also likely to contain interacting grains. However, only minor differences in R ratio are to be found within the whole suite of Catherine samples studied indicating that interaction fields are unlikely to have significantly perturbed the Catherine modelling calculations.

5. Discussion

Mass mixing and crystal mixing calculations enable mineral magnetic measurements of natural materials to be expressed in a form suitable for quantitative magnetic differentiation, tracing and monitoring studies. Magnetic remanence measurements in particular have proved to be very powerful for investigating very low concentrations of iron oxides that are difficult to analyse quantita-

tively by other magnetic methods or by X-ray or Mossbauer studies.

Difficulties in fitting model remanent magnetization curves, particularly with grain size estimation, indicate the need for additional measurements. In particular remanence measurements of magnetizations grown in fields in excess of 1T are desirable as are measurements for magnetite grains close to the superparamagnetic/stable single domain boundary are needed. Figure 10 illustrates some remanent magnetizations for a haematite and a goethite sample. Pronounced changes in remanent magnetization above 1T are revealed particularly in the goethite. Further experiments are planned to help characterise the behaviour of goethite and haematite crystals over a range of grain sizes at these higher field strengths.

Recent developments in microprocessor controlled vibrating sample magnetometers and in coil detection systems used with pulse magnetizers will probably lead to improved noise levels and allow magnetization as well as remanence measurements to be used in routine data collection. An improvement over bicubic spline functions for approximat-

ing hysteresis magnetization loops might be the first order linear differential equations discussed by Jiles and Atherton (1983). A great advantage of modelling magnetization hysteresis loops in addition to remanence hysteresis loops would be that an intrinsic parameter—saturation magnetization—would be under consideration in addition to the various magnetic parameters controlled by extrinsic influences.

An investigation of the range of magnetite grain sizes within a sample was performed in one version of the crystal mixing programme, but it was not particularly successful. Extension of the algorithm to include frequency dependent and quadrature susceptibility measurements and viscous and anhysteretic remanences might be one approach to estimating grain size spread and tackling samples with grains spanning the superparamagnetic/stable single domain boundary.

Acknowledgements

B. Jackson of the Royal Scottish Museum and P. Aspen of the Grant Institute provided magnetic materials. W.D. Corner, H.M. Whitworth and H.W. Myron provided access to the high magnetic field facilities at Durham, Oxford and Nijmegen Universities. W. O'Reilly arranged use of X-ray and magnetic equipment at Newcastle University. The author most gratefully acknowledges all their valuable help, assistance and advice.

References

- Bate, G., 1962. Statistical stability of the Preisach diagram for particles of λ -Fe₂O₃. *J. Appl. Phys.*, 3: 2263–2269.
- Bhathal, R.S. and Stacey, F.D., 1969. Frequency independence of low-field susceptibility of rocks. *J. Geophys. Res.*, 74: 2025–2027.
- Box, M.J., 1965. A new method of constrained optimization and a comparison with other methods. *Comput. J.*, 8: 42–52.
- Bradshaw, R.H.W. and Thompson, R., 1985. The use of magnetic measurements to investigate the mineralogy of some Icelandic lake sediments and to study catchment processes. *Boreas*, 14: 203–215.
- Bunday, B.D., 1984. *Basic Optimization Methods*. Edward Arnold, London.

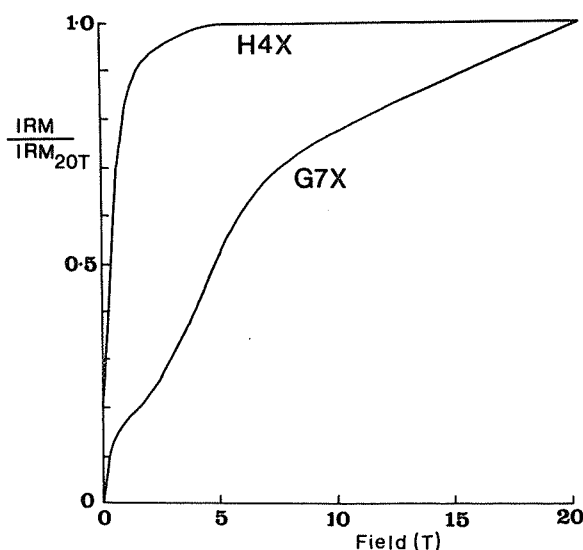


Fig. 10. Acquisition of remanent magnetization curves. G7X is a goethite sample, supplied by the Royal Scottish Museum. It acquired an IRM of $18 \text{ mAm}^2 \text{ kg}^{-1}$ in the peak field used of 20 T. H4X is a haematite pencil ore from Framlington with a saturation IRM of $0.21 \text{ Am}^2 \text{ kg}^{-1}$.

- Caceci, M.S. and Cacheris, W.P., 1984. Fitting curves to data, the Simplex algorithm is the answer. *Byte*, 9, pp. 340–362.
- Chikazumi, S., Miura, N., Kido, G. and Akihiro, M., 1978. Production of magnetic fields in Megagauss region and related measuring techniques. *IEEE Magnetics*, 14; 577–585.
- Cisowski, S., 1981. Interacting versus non-interacting single domain behaviour in natural and synthetic samples. *Phys. Earth Planet. Inter.*, 26: 56–62.
- Clark, D.A., 1984. Hysteresis properties of sized dispersed monoclinic pyrrhotite grains. *Geophys. Res. Lett.*, 11: 173–176.
- Collinson, D.W., 1967. The variation of magnetic properties among red sandstone. *Geophys. J.R. Astron. Soc.*, 12: 197–207.
- Collinson, D.W., 1968a. An estimate of the haematite content of sediments by magnetic analysis. *Earth Planet. Sci. Lett.*, 4: 417–421.
- Collinson, D.W., 1968b. Ferrous and ferric iron in red sediments and their magnetic properties. *Geophys. J.R. Astron. Soc.*, 16: 531–542.
- Currie, R.G. and Bornhold, B.D., 1983. The magnetic susceptibility of continental shelf sediments, west coast Vancouver Island, Canada. *Mar. Geol.*, 51: 115–127.
- Dankers, P.H.M., 1978. Magnetic Properties of Dispersed Natural Iron-oxides of Known Grain Size. Ph.D., University of Utrecht, pp. 33–55.
- De Boor, C., 1978. A Practical Guide to Splines. Springer-Verlag, New York, pp. 108–128.
- Dunlop, D.J., 1972. Magnetic mineralogy of unheated and heated red sediments of coercivity spectrum analysis. *Geophys. J.R. Astron. Soc.*, 27: 37–55.
- Dunlop, D.J., 1973. Superparamagnetic and single-domain threshold sizes in magnetite. *J. Geophys. Res.*, 78: 1780–1793.
- Dunlop, D.J., 1983. Determination of domain structure in igneous rocks by alternating field and other methods. *Earth Planet. Sci. Lett.*, 63: 353–367.
- Dunlop, D.J. and West, G.F., 1969. An experimental evaluation of single domain theories. *Rev. Geophys.*, 7: 709–757.
- Eldridge, D.F., 1964. Quantitative determination of the interaction fields in aggregates of single-domain particles. *J. Appl. Phys.*, Supplement, 32: 247–249.
- Galt, J.K., 1952. Motion of ferromagnetic domain wall in Fe_3O_4 . *Phys. Rev.*, 85: 664–669.
- Hayes, J.G. and Halliday, J., 1974. The least squares fitting of cubic spline surfaces to general data sets. *J. Inst. Maths. Appl.*, 14: 89–103.
- Hirons, K. and Thompson, R., 1985. The palaeoenvironmental application of magnetic measurements from inter-duration hollow lake sediments near Dungannon, Co. Tyrone, N. Ireland. *Boreas*, in press.
- Jaep, W.F., 1969. Anhysteretic magnetization of an assembly of single-domain particles. *J. Appl. Phys.*, 40: 1297–1298.
- Jiles, D.C. and Atherton, D.L., 1983. Ferromagnetic hysteresis. *IEEE Trans. Magnet.*, 19: 2183–2185.
- Kneller, E.F. and Luborsky, F.E., 1963. Particle size dependence on coercivity and remanence of single-domain particles. *J. Appl. Phys.*, 134: 656–658.
- Mullins, C.E., 1974. The magnetic properties of the soil and their application to archaeological prospecting. In: *Technische und Naturwissenschaftliche Beitrag zur Feldarchaologie*. Rheinland-Verlag, pp. 143–344.
- Nedler, J.A. and Mead, R., 1965. A simplex method for function minimization. *Comput. J.*, 7: 308–313.
- Néel, L., 1955. Some theoretical aspects of rock magnetism. *Adv. Phys.*, 4: 191–243.
- Oldfield, F., Barnosky, C., Leopold, E.B. and Smith, J.P., 1983. Mineral magnetic studies of lake sediments. *Hydrobiologia*, 103: 37–44.
- O'Reilly, W., 1988. *Rock and Mineral Magnetism*. Blackie, Glasgow, pp. 132–171.
- Parry, L.G., 1965. Magnetic properties of dispersed magnetite powders. *Philos. Mag.*, 11: 303–312.
- Preisach, F., 1935. Über die magnetische Nachwirkung. *Z. Phys.*, 94: 277–302.
- Puranen, R., 1977. Magnetic susceptibility and its anisotropy in the study of glacial transport in northern Finland. In: L.K. Lawrence (Editor), *Prospecting in Areas of Glaciated Terrain*. Institution of Mining and Metallurgy, London, pp. 111–119.
- Rubin, L.G. and Wolff, P.A., 1984. High magnetic fields for physics. *Phys. Today*, August, 24–33.
- Smit, J. and Wijn, H.P.J., 1954. Physical properties of ferrites. *Adv. Electron. Electron. Phys.*, 6: 69–136.
- Snoek, J.L., 1948. Dispersion and absorption in magnetic ferrites at frequencies above one megacycle. *Physica*, 14: 207–217.
- Stephenson, A., 1971. Single domain grain distributions. *Phys. Earth Planet. Inter.*, 4: 353–369.
- Stewart, R.C., 1979. An Investigation of the Magnetic Remanence in Recent Lacustrine Sediments. Unpublished B.Sc. dissertation, University of Edinburgh, pp. 1–15.
- Stober, J. C. and Thompson, R., 1979. An investigation into the source of magnetic minerals in some Finnish lake sediments. *Earth Planet. Sci. Lett.*, 45: 464–474.
- Thompson, R. and Edwards, K., 1982. A Holocene palaeomagnetic record and a geomagnetic master curve from Ireland. *Boreas*, 11: 335–349.
- Thompson, R. and Oldfield, F., 1986. *Environmental Magnetism*. Allen and Unwin, London.
- Thompson, R., Bloemendal, J., Dearing, J.A., Oldfield, F., Rummary, T.A., Stober, J.C. and Turner, G.M., 1980. Environmental applications of magnetic measurements. *Science*, 207: 481–486.
- Wasilewski, P.J., 1973. Magnetic hysteresis in natural materials. *Earth Planet. Sci. Lett.*, 20: 67–72.
- Wohlfarth, E.P., 1958. Relations between different modes of acquisition of the remanent magnetization of ferromagnetic particles. *J. Appl. Phys.*, 29: 595–596.