RESEARCH ARTICLE

10.1002/2014GC005680

Special Section:

Magnetism From Atomic to Planetary Scales: Physical Principles and Interdisciplinary Applications in Geo- and Planetary Sciences

Key Points:

- A variable-resolution FORC measurement protocol has been developed
- The irregular FORC protocol efficiently captures fine-scale features
- Free measurement and processing software are provided

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Citation:

Zhao, X., D. Heslop, and A. P. Roberts (2015), A protocol for variable-resolution first-order reversal curve measurements, *Geochem. Geophys. Geosyst.*, 16, 1364–1377, doi:10.1002/2014GC005680.

Received 1 DEC 2014

Accepted 31 MAR 2015

Accepted article online 7 APR 2015

Published online 9 MAY 2015

A protocol for variable-resolution first-order reversal curve measurements

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Abstract High-resolution first-order reversal curve (FORC) diagrams are being increasingly used in rock and environmental magnetism, including for detection of biomagnetic signals in sediments. Resolution can be a major barrier to obtaining high-quality FORC diagrams, and time-consuming measurements that employ small field steps are necessary to resolve the finest features of a FORC distribution. We present a new experimental protocol with irregularly spaced field steps that allow different parts of a FORC diagram to be measured at different resolutions. Larger numbers of measurements can, therefore, be made in key regions of a FORC distribution to resolve diagnostic features at higher resolution. Specification of the field steps in the irregular measurement grid is based on major hysteresis properties; no *a priori* knowledge concerning the underlying FORC distribution is required. FORC diagrams obtained with conventional measurements and with our new measurement protocol give consistent results. Because of its variable resolution, the irregular protocol provides a clear representation of fine-scale features produced by quasireversible superparamagnetic and noninteracting single-domain particles. Although the proposed irregular measurement protocol is not as efficient at suppressing noise as recently developed postprocessing techniques (e.g., VARIFORC), it enables efficient high-resolution analysis for relatively strongly magnetized samples where measurement noise is not detrimental to FORC distribution estimation.

1. Introduction

First-order reversal curve (FORC) diagrams [Pike *et al.*, 1999; Roberts *et al.*, 2000] have become a standard method for identifying magnetic minerals, their grain-size (domain state), and magnetostatic interactions among magnetic particles. FORC diagrams contain much useful information that magnetic hysteresis loops and isothermal remanent magnetization acquisition curves cannot provide [e.g., Roberts *et al.*, 2000, 2014], particularly concerning magnetostatic interaction distributions and domain states of magnetic particles. High-resolution FORC diagrams [Roberts *et al.*, 2000; Egli *et al.*, 2010] are useful for identifying particular features of FORC distributions, such as the so-called “central ridge” that is often indicative of the presence of intact biogenic magnetofossil chains [Egli *et al.*, 2010; Roberts *et al.*, 2011, 2012, 2013; Yamazaki, 2012; Ludwig *et al.*, 2013; Heslop *et al.*, 2013, 2014]. Improving FORC measurements and processing is desirable to enable better mapping of the FORC distribution, which is important for a wide range of applications, including the use of Preisach [1935] theory to estimate absolute paleointensities [Muxworthy and Heslop, 2011; Muxworthy *et al.*, 2011]. Improving the efficiency of FORC measurement protocols will facilitate rapid characterization of magnetic samples. In turn, an ability to make efficient FORC measurements as a routine component of studies of large sample sets will support wide-ranging analyses of fine magnetic particle systems.

Estimation of a FORC distribution requires calculation of a mixed partial second derivative [Mayergoyz, 1986; Pike *et al.*, 1999; Roberts *et al.*, 2000]. Calculation of the derivative amplifies measurement noise and may obscure features of interest; therefore, smoothing is a fundamental aspect of FORC processing. Several studies have focused on techniques to improve the quality of FORC distributions. For example, Heslop and Muxworthy [2005] described how the signal-to-noise ratio (SNR) can be used to evaluate the quality of a FORC diagram, Harrison and Feinberg [2008] adopted locally weighted regression smoothing (LOESS), which enables removal of noisy FORCs and calculation of FORC distributions on nonregular grids, Heslop and Roberts [2012] demonstrated how to calculate the statistical significance of FORC distributions, and Egli [2013] introduced the VARIFORC algorithm, which allows variable smoothing of

different parts of a FORC distribution with contrasting SNRs. These studies have made important contributions to improving FORC data processing and interpretation, but few investigations have focused on improving data acquisition [Postolache *et al.*, 2003]. In this study, we introduce a new FORC measurement protocol and estimation approach. The new measurement protocol makes use of an irregular field grid in which more data are measured in regions of larger magnetization change. This method is analogous to the VARIFORC method of Egli [2013] except that it employs a variable measurement scheme rather than data processing based on variable smoothing. We outline the proposed method and illustrate its performance with a series of examples and discuss its potential to enhance rock magnetic, environmental magnetic, and paleomagnetic studies.

2. Methods

A FORC is a magnetization curve that is measured after saturating a sample in a large magnetic field. The field is then reduced from saturation to a reversal field B_r , and M is the magnetization measured at a series of fields, B , where $B_r \leq B$. The FORC distribution, ρ , is then given by:

$$\rho(B_r, B) = -\frac{1}{2} \frac{\partial^2 M(B_r, B)}{\partial B_r \partial B}. \quad (1)$$

In the conventional FORC measurement protocol, B_r and B define a regular measurement grid with a field spacing of δB that dictates the maximum resolution of a FORC diagram [Pike *et al.*, 1999; Roberts *et al.*, 2000; Egli *et al.*, 2010]. Piecewise regression of a FORC diagram over a series of local square [Pike *et al.*, 1999; Roberts *et al.*, 2000] or nonsquare grids [Harrison and Feinberg, 2008; Egli, 2013] is then employed to estimate the FORC distribution. The SNR of the resulting FORC distribution is controlled by the size of the local grid, which is defined by the smoothing factor, SF. Employing larger SF values provides a smoother estimate of the FORC distribution at the cost of a loss of resolution. Egli *et al.* [2010] demonstrated that the resolution of a FORC distribution is $\Delta B \approx (2SF + 1)\delta B$; thus, selection of SF is based on a balance between noise reduction and resolution. With conventional measurement protocols, noise reduction without resolution loss can only be achieved by increasing the number of measurement points, either by decreasing δB or by averaging multiple repeat measurements. For example, high-resolution FORC measurements are made to resolve detailed features such as the central ridge signature that is indicative of intact magnetofossil chains [Egli *et al.*, 2010; Roberts *et al.*, 2011, 2012, 2013]. High-resolution measurements provide a powerful tool to elucidate the fine-scale features of FORC distributions. However, because M does not vary in a constant manner with respect to B_r and B , regularly spaced FORC measurements will oversample regions with small magnetization gradients and undersample regions with large magnetization gradients. Oversampling contributes to long measurement times, which places a limit on the number of samples that can analyzed in a given study.

2.1. FORC Measurements on an Irregularly Spaced Field Grid

To overcome key limitations associated with making high-resolution FORC measurements on a regular grid, we introduce a measurement grid with irregular field spacing (Figure 1). We use this protocol to sample FORC distributions at high resolution over regions with larger magnetization gradients, as proposed by Postolache *et al.* [2003]. The form of a FORC distribution is not known *a priori*; we, therefore, gain important first-hand information from the slope-corrected major hysteresis loop to facilitate design of an irregularly spaced measurement grid (Figures 1a and 1b).

An irregular sequence of FORC measurements is constructed by first selecting a positive calibration field (B_{cal}) from the region where the major hysteresis loop is closed. Correspondingly, the magnetization at this field is defined as M_{cal} . The major hysteresis loop is then considered at fields in the interval $[-B_{cal}, B_{cal}]$, with magnetizations along the upper and lower branches given by $M^+(B)$ and $M^-(B)$, respectively. For an irregular FORC measurement grid consisting of N FORCs, a magnetization spacing, ΔM , is defined as:

$$\Delta M = 2 \times M_{cal} / N. \quad (2)$$

The first data point for each FORC is B_r , which is located on M^+ . Thus, based on a sequence of magnetizations spaced at ΔM , a series of B_r values can be defined from M^+ . For the j th FORC, B_r is given by:

Figure 1. Newly proposed FORC measurement protocol with irregularly spaced field steps in the measurement grid. (a) Uncorrected and (b) slope-corrected major hysteresis loop; and (c) definition of irregularly spaced field measurement steps from the slope-corrected hysteresis loop based on evenly spaced magnetization steps, and (d) the generated irregular grid. (e) The raw FORC surface, (f) the data point distribution in $\{B_r, B\}$ space with illustrated data selection areas for the irregular grid (SF = 2), and (g) the resulting FORC distribution plotted on the irregular measurement grid to illustrate the resolution of the distribution. (h) Illustration of the FORC distribution in $M-B$ space, and (i) rotated FORC diagram in $\{B_c, B_r\}$ space (SF = 2). Note: for clarity, different subfigures have different scales; the dashed square in Figure 1d indicates the data range for Figure 1e, the square in Figure 1e indicates the data range in Figure 1g, and the square in Figure 1g indicates the limits of the rotated FORC diagram in Figure 1i.

$$M^+(B_r) = M^+(B_{cal}) - j \times \Delta M, \quad (3)$$

where B_r can be determined by estimating $M^+(B)^{-1}$ via a least squares cubic spline. Once a given B_r is determined, M^- is used to define a sequence of fields at which the FORC will be measured between B_r and B_{cal} (Figure 1c). This sequence of fields is again based on ΔM , with the k th field of the j th FORC given by:

$$M^-(B) = M^-(-B_{cal}) + (N + k - j - 1) \times \Delta M, \quad (4)$$

where B can be determined by estimating $M^-(B)^{-1}$ via a least squares cubic spline.

FORC measurements, especially high-resolution measurements, may take many hours and instrument drift must be accounted for [Jackson and Solheid, 2010; Egli, 2013; Roberts et al., 2014]. Drift can be tracked by comparing measured magnetizations at a preset calibration field, which should have the same value throughout the measurement sequence if no drift has occurred. For the widely used MicroMagTM systems, calibration measurements are made after each FORC to enable drift correction (see Egli [2013] and Roberts et al. [2014] for details). In our new protocol, measurements continue until B_{cal} is reached (Figure 1c), and the last data point of each FORC is used for calibration.

As indicated in equations (2–4), our proposed approach is based on creating an irregularly spaced measurement grid, where each field step is expected to yield a constant magnetization change of ΔM (Figure 1c). Although the hysteresis loop-based measurement grid (Figure 1d) cannot be considered ideal, it provides a practical solution to defining an appropriate irregular measurement grid given that the underlying FORC distribution is unknown. We used the multisegment function in the Princeton Measurements Corporation (PMC) MicroMagTM vibrating sample magnetometer (VSM) software to perform a sequence of FORC measurements over an irregular grid. Software for creating irregular grid multisegment sequences and processing of the resulting FORC diagrams is available at <https://sites.google.com/site/irregular-forc/>.

2.2. Estimation of a FORC Distribution

A FORC distribution is a map of the mixed partial second derivative FORC function in $\{B_r, B\}$ coordinates, with magnitude $\rho(B_r, B)$ as given by equation (1). When plotting a FORC distribution, it is convenient to change coordinates from $\{B_r, B\}$ to: $\{B_c = (B - B_r)/2, B_f = (B_r + B)/2\}$, which involves a 45° counterclockwise rotation so that the x - y axes of the FORC distribution are the B_c and B_f axes, respectively. The most straightforward way to calculate a FORC distribution would be to use finite differences to calculate the derivative along either B_r or B , followed by calculation of the derivative along the alternate B or B_r direction [Egli et al., 2010]. The main disadvantage of this method is that it increases the influence of noise [Egli et al., 2010]. Pike et al. [1999] introduced a local second-order polynomial regression over a square array of $(2SF + 1) \times (2SF + 1)$ points. As discussed above, such regression increases smoothness at a cost of resolution (ΔB) [Egli et al., 2010]. Selection of a suitable SF is, therefore, important when calculating a FORC distribution. Heslop and Muxworthy [2005] and Harrison and Feinberg [2008] provided algorithms to determine the optimum SF value. However, use of regression over a local grid of fixed size can cause excessive smoothing in some regions of FORC space and too little smoothing in others. To address this problem, Egli [2013] developed the VARIFORC protocol to enable variable smoothing across FORC space. This protocol is a significant development, but it still relies on time-consuming high-resolution FORC measurements. In our new FORC measurement protocol, we use a constant SF throughout FORC space as proposed by Pike et al. [1999], but smoothing is carried out on a rectangular instead of a square grid (because of the irregular field spacing). In Figure 1f, we illustrate how data are selected over an irregular grid for SF = 2. Second-order polynomial regression is carried out for each data point (where the polynomial takes the form $a_1 + a_2B_r + a_3B_r^2 + a_4B + a_5B^2 + a_6B_rB$ [Pike et al., 1999; Roberts et al., 2000]); the FORC distribution is then estimated as $-0.5a_6$ [Roberts et al., 2014]. Given the form of the irregular grids, we have chosen not to employ a weighted polynomial regression, such as LOESS [Harrison and Feinberg, 2008]. The field spacing within a local grid selected for surface fitting increases with the magnitude of B_r and B . Therefore, a distance-based weighting scheme will consistently place more emphasis on the lower field portion of a local grid. To avoid potential biases introduced by up-weighting the lower field measurements, we favor nonweighted regression. Calculated FORC distributions based on irregular measurement grids can be plotted in three coordinate systems (Figures 1g–1i).

2.3. Measurement Errors in FORC Diagrams

Roberts *et al.* [2014] made 30 sets of high-resolution FORC measurements of the same sample using a PMC MicroMagTM VSM 2900, and calculated the standard deviation of field and magnetization values for each FORC data point in order to estimate nonsystematic measurement errors [Roberts *et al.*, 2014, Figure 2]. Standard deviations for the measured field values can be relatively high (up to 1.6 mT), with an average of around 0.01 mT (Figures 2b and 2c). Magnetization measurements have an almost identical standard deviation distribution to that of B (Figure 2c), which indicates that B and M measurement errors may be produced by the same source; or that magnetization errors are caused by instrument-related inaccuracies in reaching B values. According to Roberts *et al.* [2014], the standard deviation of both B and M do not vary strongly across the FORC plane and spikes are thought to have an external origin (e.g., radio frequency interference). Thus, we consider the measurement errors within local regression grids to be random and white (i.e., with constant power spectral density). Under this assumption, errors in both M and B can only be reduced by improving the instrument's SNR or by averaging repeated measurements.

2.4. FORC Distribution Signal-to-Noise Ratios

Because the FORC distribution for a given sample is unknown prior to measurement, the form of the irregular field grid is defined based on the sample's major hysteresis loop. It is then assumed that regions of the loop with larger magnetization gradients will correspond to larger values of ρ in the FORC distribution (Figure 3). Although there are clear violations of this assumption (e.g., superparamagnetic grains well below the stable single-domain threshold have $\rho = 0$ throughout FORC space) so that the sampling density does not exactly follow the FORC distribution (Figure 3), it does provide a basic relationship between a major hysteresis loop and ρ for typical geological materials. This assumed relationship is essential given that no *a priori* information is available concerning the FORC distribution for a sample.

Heslop and Roberts [2012] developed an approach to estimate the standard error of ρ when a FORC distribution is calculated using second-order polynomial regression. Assuming a square local measurement grid with side $2SF + 1$ and a field spacing of δB , the expected standard error on ρ is given by:

$$E(SE_{\rho}) = \frac{6\sigma_M}{(2SF+1)^3 \delta B^2}, \quad (5)$$

where σ_M is the standard deviation of the measurement errors, which are assumed to be independent and identically distributed. Thus, a simple expression for the expected SNR of a given value of ρ is:

$$E(SNR) = \frac{|\rho|}{E(SE_{\rho})} = \frac{(2SF+1)^3 \delta B^2 |\rho|}{6\sigma_M}. \quad (6)$$

For conventional FORC processing, where SF and δB are fixed, $E(SE_{\rho})$ will be constant across the plane (Figure 4a) and variations in SNR will be controlled solely by ρ . For the irregular grid measurement protocol, SF is fixed across the FORC plane, but δB is adjusted as a function of the gradient of the major hysteresis loop (Figure 4b). Specifically, in regions with low magnetization gradients where ρ is expected to be small, δB will be increased (equations (2)–(4)), which reduces $E(SE_{\rho})$ and improves the SNR.

Egli [2013] proposed an optimized protocol for calculating FORC distributions. The VARIFORC method performs regression on rectangular local grids, the size and aspect ratio of which can be varied as a function of B_r and B . VARIFORC is used for postprocessing, which means that smoothing can be gradually refined for a given sample. This approach, however, relies on using small δB during measurement to ensure that all features of interest can be resolved in the final FORC distribution.

To simplify comparison of our proposed irregular grid method to VARIFORC, we consider regression on square grids and, for irregular grid measurements, assume that δB is effectively constant within any local region. VARIFORC increases the SNR of ρ at a given point in the FORC plane by increasing the size of the local regression grid (and, therefore, the number of measurement points within the grid), while δB is constant. For a square local regression grid with a side of A , the number of points, n , along each side of the VARIFORC grid is given by:

$$n = \frac{A}{\delta B} + 1. \quad (7)$$

Substitution into equation (6) gives:

Figure 2. Statistical analyses of magnetization and field measurement errors. (a) Histogram of standard deviation of magnetization, σ_M ; (b) histogram of standard deviation of field, σ_B ; and (c) cross plot of the standard deviation of magnetization and field.

$$\text{SNR} = \frac{\left(\frac{A}{\delta B} + 1\right)^3 \delta B^2 |\rho|}{6\sigma_M}, \quad (8)$$

which has a partial derivative with respect to A of:

$$\frac{\partial \text{SNR}}{\partial A} = \frac{(\delta B + A)^2 |\rho|}{2\sigma_M \delta B}, \quad (9)$$

so that the VARIFORC SNR increases as the square of A . In contrast, the irregular measurement grid method increases the size of the local regression grid by increasing δB , while holding n constant. Thus, the field spacing within the local grid is given by:

Figure 3. Sampling density distribution for a FORC distribution for a basalt sample with magnetic properties dominated by single-domain magnetite. (a and f) Sampling density and FORC diagram for the irregularly spaced measurement protocol. Vertical and horizontal profiles for the (b and c) sampling density, and (d and e) interaction field and coercivity distributions, respectively.

Figure 4. Noise transferred into FORC distributions for (a) a regular grid and (b) an irregular grid. The black dots in Figures 4a and 4b are mirror images of the sample grid (i.e., reflected about $B_r = B$). (c) The expected standard error of ρ plotted as a function of box size, with solid red and dashed green lines representing the irregular measurement grid and VARIFORC methods, respectively. Calculations were performed with $\delta B = 1$ mT for the VARIFORC method and $SF = 3$ for the irregular method. Note $E(SE_{\rho})$ will scale linearly with σ_M .

$$\delta B = \frac{A}{n-1}, \quad (10)$$

and substitution into equation (6) yields SNR for the irregular grid approach of:

$$SNR = \frac{n^3 \left(\frac{A}{n-1}\right)^2 |\rho|}{6\sigma_M}. \quad (11)$$

In this case, the partial derivative with respect to A is:

$$\frac{\partial SNR}{\partial A} = \frac{n^3 A |\rho|}{3(n-1)^2 \sigma_M}, \quad (12)$$

so that the SNR increases in proportion to A . The irregular and VARIFORC approaches are compared in Figure 4c for a square regression box with a side of A . As box size grows, $E(SE_{\rho})$ for both techniques decreases rapidly (equations (9) and (12)), however, VARIFORC yields higher SNRs with increasing box size. This comparison demonstrates that compared to the irregular grid method, VARIFORC provides a more efficient means of increasing the SNR of a FORC distribution while maintaining resolution by keeping box sizes low. Therefore, VARIFORC should be preferred for weakly magnetized samples, where care needs to be taken to increase the SNR to reveal fine features of a FORC distribution. The main advantage of the irregular grid method is for analyzing more strongly magnetized samples, where the SNR is not of primary concern, but where the FORC distribution contains features that can only be resolved with high-resolution measurements. The flexibility of irregular measurement grids to deliver resolution where it is most needed provides an efficient means to obtain a FORC diagram compared to measurement on a regular FORC grid where a δB small enough to resolve the finest features must be maintained throughout the entire measurement sequence.

3. Results

3.1. FORC Results for an Irregularly Spaced Grid

To test the irregular grid measurement protocol, FORC distributions for conventional and irregularly spaced grids are compared in Figure 5 for three types of samples. Hysteresis loops before and after paramagnetic slope correction, and measurement grids generated from corrected loops, are shown in the first column of Figure 5. For a multidomain sample, FORC diagrams are shown in $\{B_r, B\}$ space for the irregular grid in Figure 5b, and are compared in $\{B_c, B\}$ space in Figure 5c for irregular (left) and conventional grids (right). Compared to regular grid FORC measurements, the varying resolution of the irregular protocol gives a clearer distribution and sharper B_r and B_c profiles. FORC diagrams obtained with irregular and regular measurement

Figure 5. Comparison of FORC distributions calculated for measurement protocols with irregular and regular measurement grids. (a, e, and i) An illustration of the irregular measurement grid and major hysteresis loops for different samples. (b) FORC distribution in $\{B_i, B\}$ space with its associated 0.05 significance level (solid white line) calculated according to Heslop and Roberts [2012]. The left-hand FORC distribution in Figure 5c is a conventional representation in $\{B_c, B_i\}$ space for the irregularly spaced grid and the right-hand FORC distribution is for a regularly spaced grid for a multidomain sample [see Pike et al., 2001b]. (f and g) FORC diagrams for irregularly and regularly spaced measurements, respectively, for a SD magnetite sample; and (j and k) for a SD hematite-bearing sample. (d, h, and l) Projections where the FORC distribution has been rotated in three dimensions to provide views along the B_i and B_c axes; black solid lines are for the irregular grids, while the solid blue areas correspond to regular grid results.

grids are shown in Figures 5f and 5g for a basalt sample that contains stable single-domain (SD) magnetite. These distributions are nearly identical (as are the B_i and B_c profiles), although the irregular grid contains only 80 FORCs compared to 250 FORCs for the regular measurements. There is some loss of high-coercivity detail in the irregular FORC, which is a result of the smaller number of FORCs measured and the inability to determine an optimal measurement grid based on major hysteresis properties alone. FORC diagrams for a stable SD hematite-bearing sample are shown in Figures 5j and 5k. The two FORC diagrams are similar, although the irregular grid protocol gives smoother results from fewer FORC measurements. In summary, the irregular grid protocol enables reliable FORC measurements with fewer FORCs than the conventional approach. Loss of high-coercivity detail in some irregular-grid FORC diagrams could potentially be resolved by measuring more FORCs. The irregular FORC measurement protocol has higher resolution than the conventional method in the main part of the FORC distribution because of the higher sampling density in that region; likewise, the irregular protocol has reduced measurement density in parts of the FORC distribution with lower ρ .

3.2. Preisach Model

An ideal FORC distribution should be noise free, with no distortion. In reality, there is always a trade-off in experimental FORC distributions between reducing noise and smoothing-induced distortion. Optimal smoothing levels are usually judged visually rather than statistically and associated distortion due to oversmoothing is difficult to assess. To address this question, we employ a Preisach model [Preisach, 1935] to demonstrate differences between irregular and regular FORC measurement protocols. A smooth FORC distribution was constructed numerically for an idealized sample that contains two magnetic components (Figure 6a); one component comprises highly interacting SD particles with a large vertical spread along the B_i axis, while the other is represented by a central ridge signature due to non-interacting uniaxial SD particles [Egli et al., 2010]. Vertical and horizontal profiles through the FORC distribution at $B_i = 0$ mT and $B_c = 20$ mT are shown in Figures 6b and 6c, which correspond to interaction field and coercivity distributions, respectively [Roberts et al., 2000]. Regular FORC diagrams are plotted in Figures 6d and 6g for 100 and 250 simulated FORCs, respectively. FORC distributions for irregular measurement grids for 100 and 150 simulated FORCs are shown in Figures 6j and 6m, respectively. Coercivity and interaction field profiles for the simulations are shown on the right of Figure 6 (to facilitate comparison of these simulations, we use a constant color scale throughout the figure with dashed blue lines that represent the true input Preisach distribution). These results indicate that the irregular grid method produces a good approximation to the coercivity and interaction profiles even with a small number of FORCs, while the regular grid method is overly smooth due to a lack of resolution, especially where the FORC distribution has sharp gradients. This is consistent with the results of Postolache et al. [2003]. While the irregular grid method performs well in representing the main body of the FORC distribution, it underestimates the highest coercivity portion (Figures 6l and 6o).

3.3. Mapping Central Ridge Signatures

A central ridge is a sharp feature in an FORC diagram that reflects the presence of noninteracting or weakly interacting SD particles that often result from the presence of intact magnetofossil chains [Egli et al., 2010; Roberts et al., 2011, 2012, 2013; Ludwig et al., 2013; Heslop et al., 2014; Egli, 2013]. Central ridges can only be adequately defined with high-resolution FORC measurements; however, with regular spacing, time-consuming measurements must also be made in other regions. The irregular grid FORC protocol has the inherent advantage of higher measurement density in areas related to central ridges, and reduced measurement density in regions with negligible magnetization gradients (Figures 3, 4, and 7). The irregular grid also requires less measurement time, although some information is lost in higher coercivity regions. Figures 7a and 7b are examples of a central ridge from regular and irregular FORC measurements for a pelagic marine carbonate [Roberts et al., 2012]. The central ridge is defined with fewer FORCs for irregular measurements, which also produces a smoother FORC diagram compared to the regular measurement grid. The irregular measurement protocol is, therefore, a valuable option for defining central ridge signatures efficiently.

3.4. Detection of Superparamagnetic Signals

In addition to SD, pseudosingle domain and multidomain contributions, superparamagnetic (SP) particles can be detected with FORC diagrams [e.g., Pike et al., 2001a]. SP particles can be difficult to detect using conventional FORC measurements because either δB is too large or because of their small contribution to the total magnetization. Nevertheless, the irregular grid is generated using a major hysteresis loop so that even small magnetization contributions help to define the grid, which enables better detection of SP signatures. FORC diagrams from regular and irregular measurements for a SP-rich Australian topsoil sample (frequency dependence of magnetic susceptibility of 7.7%) are shown in Figures 7c and 7d. SP particles give rise to an asymmetric FORC distribution along the B_i axis [Pike et al., 2001a; Roberts et al., 2000, 2014], which is evident in both the regular (Figure 7c) and irregular FORC measurements (Figure 7d). Because of the variable resolution of the irregular grid, a FORC diagram with $N = 80$ is sufficient to detect and characterize the SP component, with most of the FORC distribution falling within the 0.05 significance level. In contrast, a resolution high enough to detect SP particles must be maintained throughout the regular grid and a large number of FORCs ($N = 300$) is required, yet most of the SP part of the FORC distribution falls outside the 0.05 significance level. Clear detection of quasireversible SP signals is a significant advantage of the irregular grid protocol.

Figure 6. Preisach [1935] model and FORC simulations to illustrate the relative performance of regular and irregular measurement grids in representing a known FORC distribution. (a–c) An ideal Preisach [1935] distribution, (d–l) results of simulations using regular measurement grids (with 100 and 250 FORCs, respectively), and (j–o) simulations using irregular measurement grids (with 100 and 150 FORCs, respectively). (a, d, g, j, and m) The estimated FORC distribution, (b, e, h, k, and n) the estimated interaction field distribution, and (c, f, i, l, and o) the estimated coercivity distribution. The blue dashed lines represent the Preisach [1935] distribution profiles from Figures 6b and 6c. Artifacts in Figures 6d–6o indicate that estimates of ρ based on regression may not recover the true FORC distribution, even for noise-free data.

Figure 7. FORC diagrams for SD and SP samples calculated from measurements made with regular and irregular grids. (a) Regular and (b) irregular FORC diagrams for sample ODP690B-019H02W-140–142 [Roberts *et al.*, 2012] with a noninteracting SD central ridge signature (white contours represent the 0.05 significance level calculated according to Heslop and Roberts [2012]); and (c) regular and (d) irregular FORC diagrams for an Australian topsoil sample. Streaking of the FORC distribution in the low ρ regions for the irregular FORC protocol is an artifact that results from a lack of data in these regions. However, the measurement grid is based on a major hysteresis loop, therefore, it can be assumed *a priori* that these regions are not important for interpretation of the FORC distribution.

4. Discussion

An efficient FORC measurement protocol should be adaptable, with measurement density based on the magnetic properties of the sample. The conventional (regular grid) protocol is simple; the only parameter needed to define measurements within the B_r and B_c limits is the field step. The issue with employing constant δB is that different parts of the FORC distribution will be under or oversampled depending on local characteristics. The irregular grid protocol is straightforward to implement, and provides a workable strategy in which over and undersampling are balanced by adopting a measurement sequence that is dictated by the magnetic properties of the sample. Thus, use of an irregular grid is an attractive option for relatively strongly magnetized samples, where measurement noise will not have an overly detrimental effect on the estimated FORC distribution. For weakly magnetized samples, however, postprocessing using the VARIFORC protocol provides a more efficient means to increase the SNR of a FORC distribution with the advantage that smoothing regimes can be defined once the approximate form of the distribution is known.

FORCs are time consuming to measure, especially when high-resolution measurements are required. Measurement time depends on the specific diagram parameters [Pike *et al.*, 1999; Roberts *et al.*, 2000, 2014] and is 1–2 h for a conventional measurement with less than 140 FORCs. High-resolution measurements and measurement of high-coercivity samples (e.g., hematite-rich materials) can take several hours. It is a challenge to compare the efficiency of irregular and regular FORC measurements, but measurement efficiency

Figure 8. Data usage rate in FORC measurements for an Australian topsoil. A 68% usage rate is obtained in (a) for an irregular grid and (b) a 45% rate for a regular grid.

can be illustrated with the usage rate of measured data (the percentage of data points in the region of interest with respect to the number of measured data points). The usage rate for an Australian topsoil sample for both methods is compared in Figure 8; the irregular method has a higher usage rate than the regular method (68% and 45%, respectively). The usage rate for the irregular method could be further improved by not measuring across the entire FORC plane. The usage rate of irregular grids is not constant, but depends on sample-specific properties and the definition of the FORC region of interest. It is clear, however, that for more strongly magnetized samples, use of an irregular grid can substantially shorten measurement time, thereby increasing overall efficiency.

SF has an important role in the second-order regression used for FORC processing [Pike *et al.*, 1999; Roberts *et al.*, 2000; Heslop and Muxworthy, 2005; Harrison and Feinberg, 2008; Egli *et al.*, 2010; Egli, 2013; Roberts *et al.*, 2014]. Larger SF values generally cause more signal distortion regardless of measurement method or calculation algorithm (Figure 9). Signal distortion is illustrated in Figure 9 for $SF = 3$ to 6 for both regular and irregular methods. Distortion occurs for both methods, with greater distortion along the B_i axis for the regular method; in contrast, greater distortion occurs along the high-field tail of the B_c axis for the irregular method. Distortion is usually caused by undersampling, where data have insufficient resolution to fully map FORC surface variations. This is an inherent problem that can only be overcome by increasing the sampling density until an acceptable approximation of the true FORC distribution is obtained. Achieving adequate sampling density is why high-resolution FORC measurements are needed to identify central ridge signatures with a regular measurement grid [Egli *et al.*, 2010] and optimal SF values can be determined *post hoc* using an appropriate objective function [Heslop and Muxworthy, 2005; Harrison and Feinberg, 2008]. For irregular grids, large δB changes throughout the FORC plane mean that wherever possible small SF values (typically 2 or 3) should be preferred, because use of higher SF values in regions where δB is large will produce low-resolution zones within the FORC distribution. Given that an irregular measurement protocol is most appropriate for relatively strongly magnetized samples (section 3), larger SF values should not be necessary for calculating FORC distributions. Where a higher SF value is deemed necessary to reduce noise, it is more appropriate to measure a regular grid and calculate the FORC distribution using the VARIFORC protocol.

5. Conclusions

We propose a new FORC measurement protocol with an irregularly spaced measurement grid that provides an efficient, high-resolution alternative to conventional FORC measurements for relatively strongly magnetized samples. The variable resolution provided by the irregular grid approach allows detailed

Figure 9. Signal distortion in FORC calculations for conventional (regular) and irregular measurement protocols. Distortion of the FORC distribution estimate (a–d) for a regular grid measurement protocol and (e–h) for an irregular grid protocol. (a and e) Reference FORC distributions (SF = 3), (b and f) difference from the reference FORC distributions for SF = 4, (c and g) for SF = 5, and (d and h) for SF = 6.

measurements in potential regions of interest without making time-consuming high-resolution measurements over the entire span of the measured FORC space. Overall, FORC distributions calculated from measurements obtained using our new protocol compare well with those obtained using the conventional regular grid method.

The field steps used in the irregular measurement grid are determined through measurement of a major hysteresis loop, so that it is an objective approach. Our numerical simulations demonstrate that the irregular grid protocol provides good measurement efficiency and high-quality FORC diagrams, with improved resolution in regions that contain sharp features, such as single-domain central ridges and superparamagnetic peaks. Therefore, for relatively strongly magnetized samples, the proposed irregular grid approach provides a more efficient alternative to conventional FORC measurement protocols.

Acknowledgments

We thank Pengxiang Hu and Liao Chang for useful discussions and for use of data from their samples. We are grateful to Ramon Egli, Adrian Muxworthy, and Ioan Lascu for their careful and constructive reviews, which helped to improve the manuscript. We particularly thank Ramon Egli for assisting our comparison of results from the irregular grid FORC protocol with VARIFORC. We gratefully acknowledge funding from the Australian Research Council through grants DP11010541, DP120103952, and DP140104544. A free Labview program for developing measurement sequences and processing irregular FORC diagrams is available at <https://sites.google.com/site/irregularforc/>.

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