# Magnetic interaction analysis of basaltic samples and pre-selection for absolute palaeointensity measurements

Florian Wehland,<sup>1</sup> Roman Leonhardt,<sup>2</sup> Fabienne Vadeboin<sup>3</sup> and Erwin Appel<sup>1</sup>

<sup>1</sup>Institut für Geowissenschaften, Abteilung Geophysik, Sigwartstr. 10, 72074 Tübingen, Germany. E-mail: florian.wehland@uni-tuebingen.de <sup>2</sup>Department for Earth and Environmental Sciences, Geophysics Section, Theresienstr. 41/IV, 80333 München, Germany. E-mail: leon@geophysik.uni-muenchen.de

<sup>3</sup>CEREGE, (UMR CNRS- Université, d'Aix-Marseille III), Europôle Méditerranéen de l'Arbois, BP80 13545, Aix en Provence, Cedex 4, France. E-mail: vadeboin@cerege.fr

Accepted 2004 July 22. Received 2004 July 5; in original form 2003 September 17

# SUMMARY

For a successful palaeointensity experiment a sample should only consists of non-interacting single domain (SD) particles. As this cannot be completely guarantied for natural samples we applied two different interaction methods ( $\Delta$ M plots and FORC-diagrams) on basaltic samples from Fernando de Noronha, Brazil, in order to determine the degree of interaction and estimate their use as a preselection tool for palaeointensity experiments. An independent classification of the samples in terms of SD/MD contribution and the occurrence of partial self-reversals is obtained by rockmagnetic means, Thellier-like experiments and continuous thermal demagnetization. The samples show a variable amount of mean interaction fields, which tends to be higher for samples with partial self-reversal. Nevertheless, high mean interaction fields can also be present in samples of successful palaeointensity experiments with high mean coercivities. This suggests, that the recording potential of a sample depends on the interplay of magnetic interaction fields. As this is difficult to assess by normal rockmagnetic means, interaction measurements, especially the  $\Delta$ M plot derived parameters  $\Delta$ M 1 and 2, provide a fast tool for preselection.

Key words: basalts, FORC, Henkel plot, magnetic interaction, palaeointensities.

# **1 INTRODUCTION**

The determination of palaeointensities of the Earth magnetic field is based on the Thellier–Thellier method (Thellier & Thellier 1959; modified by Coe 1967). Crucial for the success of these methods is the independence of individual pTRMs (partial thermoremanent magnetizations), which allows the accurate replay of the magnetic information by laboratory means. This requires the existence of a non-interacting single-domain (SD) particle assemblage. In natural samples, this requirement is often violated by a contribution of MD particles and magnetic interaction of various amounts.

The best procedure to estimate the quality of a sample for palaeointensity determination is a Thellier–Thellier like experiment itself. The curvature in the resulting Arai-Nagata-plot and the implementation of MD-checks during the experiment gives a broad impression of the occurrence of magnetic interaction and the MD contribution in the sample [a major MD contribution can be detected by rockmagnetic means in advance using the classification of the sample in the Day-plot (Day *et al.* 1977)]. As such experiments are very time consuming, a fast preselection method is desirable.

The scope of this paper is to apply two different magnetic interaction methods on basaltic samples and assesses the outcome preparatory to the use as a preselection tool for Thellier–Thellier experiments. This will be achieved by:

(1) Qualitative and semi-quantitative analysis of the interaction patterns obtained by  $\Delta M$  plots and FORC analysis.

(2) A comparison of the results with the classification of the samples obtained by independent measurements on twin samples using continuous thermal demagnetization and Thellier-experiments.

It is not intended to clarify the exact nature of the magnetic interaction in the samples. The following paragraphs epitomize the two interaction methods applied:

The first method is based on the consideration of Wohlfahrt (1958), which are valid for a non-interacting SD particle assemblage. He concluded, that the relationship of the different normalized remanence acquisitions can be described by

$$\operatorname{IRM}_{d}(H) = 1 - 2^{*} \operatorname{IRM}_{r}(H), \tag{1}$$

where  $IRM_r$  is the normalized remanence acquired from the initially demagnetized state and  $IRM_d$  the normalized remanence during the equivalent dc backfield demagnetization of the SIRM (saturation magnetization), respectively. Eq. (1) also holds for MD particles provided that the density of pinning sites experienced by the domain-wall is the same for the ascending and descending branch of the remanence curve.

The Henkel plot is obtained by plotting  $IRM_r$  versus  $IRM_d$  (Henkel 1964), where non-interacting SD particles will lead to a straight line with a slope of -2. For an SD particle assemblage any deviation from this line can be expressed by the differential remanence parameter (Speliotis & Lynch 1991)

$$\Delta M(H) = \operatorname{IRM}_{d}(H) - [1 - 2\operatorname{IRM}_{r}(H)].$$
<sup>(2)</sup>

This deviation is ascribed to many-body effects and generally indicates the presence of magnetic interparticle interaction (Fearon *et al.* 1990; Mayo *et al.* 1990a,b, 1991a,b). Positive values of  $\Delta M$ are interpreted as a result of positive interaction, which tends to stabilize the magnetization, whereas a negative value would come from negative interaction yielding a net demagnetizing effect (see Petrovský *et al.* 1993).

The simplicity of this method is questioned by the calculations made by Garcia-Otero *et al.* (2000). They concluded that the direct relation between the linearity and noninteraction only exists for particles of uniaxial anisotropy at low temperature. For particles of cubic anisotropy the deviation is positive over the entire temperature range, but changes gradually to negative with increasing dipolar interaction. As we are only interested in the relative diffeences of  $\Delta M$  parameter, this consideration is of minor importance for this study.

A more detailed attempt to detect magnetic interaction is found in the recently developed FORC (First Order Reversal Curves) diagrams (Pike *et al.* 1999; Roberts *et al.* 2000). Such a diagram is calculated from a set of partial hysteresis curves (see Mayergoyz 1986). The measurement of a FORC starts with the saturation of the sample in a high positive field. The field is afterwards decreased to the reversed field  $H_a$ , and the FORC is the magnetization curve measured from  $H_a$  back to saturation. A set of FORCs is obtained by repeating this measurement for different values of  $H_a$ . At the applied field  $H_b$  on a FORC starting from  $H_a$  the magnetization is denoted as  $M(H_a, H_b)$ , where  $H_b > H_a$ . The FORC distribution  $\rho$  $(H_a, H_b)$  is then defined as the mixed second derivative:

$$\rho(H_{\rm a}, H_{\rm b}) \equiv -\frac{\partial^2 M(H_{\rm a}, H_{\rm b}),}{\partial H_{\rm a} \partial H_{\rm b}},\tag{3}$$

where  $\rho$  ( $H_a$ ,  $H_b$ ) is defined for  $H_b > H_a$ . It became convenient for plotting the FORC distribution to use the coordinate system { $H_c = (H_b - H_a)/2$ ,  $H_u = (H_a + H_b)/2$ } instead of the original { $H_a$ ,  $H_b$ }. As  $H_b > H_a$ ,  $H_c > 0$ , and the FORC diagram is plotted as a contour plot in the right-hand half plane with  $H_u$  and  $H_c$  as the vertical and horizontal axis, respectively. The  $H_c$  coordinate is referred to microcoercivity. Further details about the derivation of eq. (3), the change of the coordinates and and basic interpretation of the resulting FORC-diagram can be found in Pike *et al.* (1999).



**Figure 1.** Continuous demagnetization diagrams of samples FN-R1-2 (a) and FN-Q20-3 (c), as well as strong field thermomagnetic curves from sister samples (b, d). The insets on the upper right of (a) and (c) show pseudo orthogonal projections of the measured *z*- and *x*-component. The reversible decrease, respectively increase of magnetization at heating steps above 200°C, particularly between 400°C and 600°C, in (a) indicates the presence of interacting particles leading to partial self-reversal. Such feature is not found in (c).



Figure 2. FORC diagrams of samples from group A (FN-Q20;FN-Q19;FN-Q24), B (FN-Q3) and C (FN-Q18; FN-R1) together with their hysteresis loop.  $H_c$  stands for the coercivity distribution in the sample, whereas  $H_u$  indicates the strength of the interaction fields.

## 2 SAMPLES AND METHODOLOGY

Samples were taken from ankaratritic lava flows and dykes from the island of Fernando de Noronha, Brazil (Leonhardt et al. 2003). Continuous demagnetizations were performed with a high temperature spinner magnetometer (HOTSPIN; described by Matzka et al. 2003), which measures simultaneously two orthogonal components of the NRM at temperatures of up to 600°C. These measurements indicate the presence of partial self-reversal in samples of 5 volcanic units. The samples were repeatedly heated and cooled in HOTSPIN instrument with incrementally increasing maximum temperatures of 100°C, 200°C, 400°C and 600°C. This technique facilitates the recognition of reversibility of heating and cooling cycles and the observation of possible blocking of remanences. Samples for the magnetic interaction analysis are grouped in: (A) four samples given suitable palaeointenisty results, (B) two samples indicating an MD contribution during Thellier experiments and (C) six samples containing self-reversed magnetization as proven by the HOTSPIN measurements. Twin specimens were taken from all of these samples and subjected to FORC analyses and IRM acquisition with subsequent backfield measurements. Prior to any treatment the specimens were subjected to AF demagnetization of 120 mT to initialise the magnetic configuration using a 2G600 AF-demagnetizer. Ensuing IRM<sub>r</sub> and IRM<sub>d</sub> were performed up to 700 mT with a field step of 5-10 mT followed by hysteresis measurements. Lastly FORC measurements (100 FORCs) were made using a saturation field of 500 mT and a smoothing factor (SF) of 2. All experiments were carried out by a Micromag 3900 Vibrating Sample Magnetometer.

## **3 RESULTS AND DISCUSSION**

#### 3.1 Continuous thermal demagnetizations

The results of continuous thermal demagnetization and corresponding strong field thermomagnetic curves  $[M_s(T)]$  are shown in Fig. 1. Sample FN-R1-2 (Figs 1a, and b) shows evidence for partial selfreversal. Stepwise continuous thermal demagnetization and the corresponding orthogonal projection of x and z-component are shown in Fig. 1(a).  $M_{S}(T)$ -curves of a sister sample from the same drill core are shown in Fig. 1(b). This sample is characterised by two ferromagnetic components with Curie temperatures (T<sub>c</sub>) of approximately 200°C and 520°C (Fig. 1b). T<sub>c</sub> is estimated by the approach of Moskowitz (1981). The low  $T_{\rm c}$  is likely related to a pristine Tirich titanomagnetite and the high  $T_{\rm c}$  phase can be attributed to a Tipoor titanomagnetite or titanomagnemite. This interpretation is also supported by ore microscopy (fig. 3 in Leonhardt et al. 2003). The orthogonal projection of continuous thermal demagnetization indicates that during the first heating cycles to 100°C and 200°C a soft overprint, very likely of viscous origin, is removed. After heating to 200°C the cooling curve in the decay diagram shows first an increase and then, at about 150°C, a characteristic decrease of magnetization towards room temperature (Fig. 1a).  $M_s(T)$ -curves indicate that this feature is not controlled by the temperature dependence of the saturation magnetization. A decrease of partial thermoremanence on cooling was also observed for multidomain grains, related to transdomain processes (Markov et al. 1983; McClelland & Sugiura 1987; Muxworthy 2000). This MD effect, however, is not reversible in the subsequent heating run. In our experiment, particularly, the decrease during cooling from 400°C is almost fully reversible during heating to 600°C. Such feature cannot be explained exclusively by the MD effect. Hence, beside a minor MD effect and possibly also some alteration during subsequent heating/cooling cycles, the occurrence of magnetic interaction between the two magnetic phases is the most plausible cause for our observations. Between the heating runs to 200°C and 400°C and, to a far smaller extend, between 400°C and 600°C, the maximum of magnetization is gradually shifted from lower temperatures to higher temperatures. The cooling cycle of the  $M_s$  (T)-curve to 700°C (Fig. 1b) indicates slightly higher  $T_c$  for the low temperature phase as well. Therefore, this observation is interpreted as a results of a partly oxidation of the low temperature phase during the subsequent heating/cooling cycles. Samples from sites FN-Q4 and FN-Q7 show similar characteristics as described for Fig. 1(a).

Fig. 1(c) shows the continuous thermal demagnetization results of sample FN-Q20-3 containing a high  $T_c$  ferromagnetic phase (Fig. 1d). Rock magnetic investigations and ore microscopy revealed high temperature oxidised titanomagnetite as the dominating remanence carrying magnetic phase accompanied by a very limited amount of hematite (Leonhardt *et al.* 2003). Samples from site FN-Q20 proved to be suitable for palaeointensity determination. The orthogonal projection indicates a single component remanence. The decay diagram of the NRM intensity also points to a simple unblocking of a high-temperature component exhibiting no indications of magnetic interaction.

## 3.2 Interaction analysis

In Fig. 2, examples of FORC diagrams of group A samples (FN-Q20; FN-Q22; FN-Q24) are compared with samples from group B (FN-Q3) and C (FN-Q13; FN-R1). The B and C samples show a higher percentage of open contour lines indicating an MD like behaviour and a strong asymmetry in the plot according the horizontal line (Pike *et al.* 1999). Thus, and the shape of the lower half



**Figure 3.** Two examples of  $\Delta M$  plots. FN-Q20 belongs to group A and FN-Q4 to group C. The position of the parameters  $\Delta M1$  and  $\Delta M2$  are marked in the upper graph.

**Table 1.** Summary of the rockmagnetic parameters and the parameters derived from the interaction methods. For sample FN-Q22 (\*) the  $\Delta$ M-plot was not calculated due to a high initial remanence.

Sample	ΔM1 (T)	$\Delta M2$	$H_{\rm cr}/H_{\rm c}$	$M_{\rm rs}/M_{\rm s}$	$H_{\rm c}$ Peak FORC (mT)	Group
FN-Q19	0,0358	-0,17	2,23	0,27	8	А
FN-Q24	0,0351	-0,417	2,28	0,20	26	А
FN-Q20	0,035	-0,127	1,93	0,31	23	А
FN-Q22	*		3,03	0,23	16	А
FN-Q3	0,0101	-0,452	2,61	0,10	7	В
FN-Q1	0,0049	-0,221	5,11	0,14	3,5	В
FN-Q18	0,00515	-0,49	2,27	0,12	5	С
FN-Q23	0,00995	-0,414	3,03	0,11	4,5	С
FN-Q4	0,0099	-0,484	2,77	0,08	5,5	С
FN-R1	0,0101	-0,395	1,97	0,20	9	С
FN-R4	0,0099	-0,485	1,71	0,18	8	С
601???	0,0101	-0,152	2,03	0,12	5	С

of the FORC diagram can be attributed towards a negative mean interaction field (Stancu *et al.* 2003). Such negative mean interaction fields are generally interpreted as having a demagnetizing effect on the sample (Petrovský *et al.* 1993) and can be expected in samples containing partial self-revesals. For the group A samples, such a strong asymmetry is only observed in sample FN-Q24 going along with a relative high mean coercivity (peak of the distribution). In the other samples the asymmetry in the FORC diagrams is negligible, although a certain degree of magnetic interaction is present (elongation of the contour lines along Hu).

In the corresponding  $\Delta M$  plots (Fig. 3) we compared the results by using the position (called  $\Delta M1$  parameter) and the intensity (called  $\Delta M2$  parameter) of the minima (see Table 1). Samples of group B and C have low  $\Delta M1$  values and tend to have high negative  $\Delta M2$  values indicating the dominance of negative mean interaction. Group A samples have all higher  $\Delta M1$  values. The  $\Delta M2$  shows an analogy with the asymmetry in the FORC diagrams based on the fact that both indicate the presence of negative mean interaction fields.

In Fig. 4 the rockmagnetic values of the samples in a Day plot are compared with the results from the  $\Delta M$  method. Whereas the three groups of samples are difficult to discriminate in the Day plot, the differences between group A samples and the samples of group B and C using the  $\Delta M1$  parameter is obvious. As A samples with high mean coercivities and higher interaction fields resist self demagnetization, it can be assumed that a stability field in the  $\Delta M1$ versus  $\Delta M2$  plot exists (marked by the dashed line with the question mark). As samples of low coercivity, but no interaction fields should also behave like ideal SD grains, the stability field should start from the origin. The clear shape of that field has to be determined by more experiments.

## **4** CONCLUSION

In terms of magnetic interaction, the analysis of the FORC and  $\Delta M1$ results indicate that the suitablility of a sample for palaeointensity experiments depends on the relationship between the degree in magnetic interaction and the mean coercivity. For high mean interaction fields-as assessed by a strong asymmetry in the FORC diagram and a high intensity of the  $\Delta M$  minima—, a particle assemblage of higher mean coercivity will give a successful palaeointensity results, whereas a sample of low coercivity is prone to effects like partial self-reversal. For the  $\Delta M$  method, this can be quantitatively assessed using a combination of the  $\Delta M1$  and  $\Delta M2$  parameters, although more data will be needed to mark distinctive values for the suitability of samples for palaeontensity measurements. Such parameters are harder to be found by the FORC methode, where the shape of the distribution expresses the mean interaction field. This cannot be easily quantivied. The peak of the FORC distribution on its own as the mean coercivity of the sample will not be sufficient to assess the potential of a sample for palaeointesity determinations. Taking into account that  $\Delta M$  plots is also faster to obtain than FORC diagram, we suggest this method as a possible new preselection tool for palaeointensity determinations.



**Figure 4.** Comparison of the samples of all groups in the  $\Delta M1$  versus  $\Delta M2$  plot and the conventional Day plot. The first one gives a better discrimination between group A and the other groups, whereas in the Day plot the transition between group A and B is gradual.

# ACKNOWLEDGMENTS

We would like to thank A. Muxworthy and E. Petrovsky for their insightful and helpful comments during the review of the paper. The work was supported by a DFG research studentship and a DAAD scholarship.

## REFERENCES

- Coe, R.S., 1967. Paleointensities of the Earth's magnetic field determined from Tertiary and Quaternary rocks, J. geophys. Res., 72, 3247–3262.
- Day, R., Fuller, M. & Schmidt, V.A., 1977. Hysteresis properties of titanomagnetites: grain-size and compositional dependence, *Phys. Earth planet. Inter.*, 13, 260–267.
- Fearon, M., Chantrell, R.W. & Wohlfahrt, E.P., 1990. A theoretical study of interaction effects on the remanence curves of particle dispersions, *J. Magn. Magn. Mater.*, 86, 197–206.
- Garcia-Otero, J., Porto, M. & Rivas, J., 2000. Henkel-plots of single domain ferromagnetic particles, J. appl. Phys., 87(10), 7376–7381.
- Henkel, O., 1964. Remanenzverhalten und Wechselwirkungen in hartmagnetischen Teilchenkollektiven, *Phys. Status Solidi*, (b), 161, K41–K44.
- Leonhardt, R., Matzka, J. & Menor, E.A., 2003. Absolute paleointensities and paleodirections of miocene and pliocene lavas from Fernando de Noronha, Brazil, *Phys. Earth planet. Int.*, **139**, 285–303.
- Markov, G.P., Shcherbakov, V.P., Bol'shakov, A.S. & Vinogradov, Y.K., 1983. On the temperature dependence of the partial thermoremanent magnetization of multidomain grains, Izv., Akad. of Sci., USSR, *Phys. Solid Earth*, 19, 625–630.
- Matzka, J., Krasa, D., Kunzmann, Th., Schult, A. & Petersen, N., 2003. Magnetic state of 10 to 40 Ma old ocean basalts and its implications for natural remanent magnetization, *Earth planet. Sci. Lett.*, 206(3–4), 541– 553.
- Mayergoyz, I.D., 1986. Mathematical model of hysteresis, *IEEE Trans. Mag.*, MAG-22, 603–608.
- Mayo, P.I., Bradbury, A., Chantrell, R.W., Kelly, P.E., Jones, H.E. & Bissel, P.R., 1990a. Interaction effects in the remanence curves of Co-Ti doped BaFe system. *IEEE Trans. Magn.*, **119**, 228–230.

- Mayo, P.I., Erkkila, R.M., Bradbury, A. & Chantrell, R.W., 1990b. Interaction effects in longitudinally oriented and non-oriented barium hexaferrite tapes. *IEEE Trans. Magn.*, MAG-26, 1894–1896.
- Mayo, P.I., O'Grady, K., Chantrell, R.W., Cambridge, J.A., Sanders, I.L., Yogi, T. & Howard, J.K., 1991a. Magnetic measurement of interaction effects in CoNiCr and CoPtCr thin film media, *J. Mag. Mag. Mater.*, 95, 109–117.
- Mayo, P.I., O'Grady, K., Kelly, P.E., Cambridge, J., Sanders, I.L., Yogi, T. & Chantrell, R.W., 1991b. A magnetic evaluation of interaction and noise characteristics of CoNiCr thin films, *J. appl. Phys.*, 69(8), 4733– 4735.
- McClelland, E. & Sugiura, N., 1987. A kinematic model of TRM acquisition in multidomain magnetite, *Phys. Earth planet. Int.*, **46**, 9–23.
- Moskowitz, B.M., 1981. Methods for estimating Curie temperatures of titanomaghemites from experimental J<sub>s</sub>-T data, *Earth planet. Sci. Lett.*, 53, 84–88.
- Muxworthy, A.R., 2000. Cooling behaviour of partial thermoremanences induced in multidomain magnetite, *Earth planet. Sci. Lett.*, **184**, 169–179.
- Petrovský, E., Hejda, P., Zelinka, T., Kropáček, V. & Šubrt, J., 1993. Experimental determination of magnetic interaction within s system of synthetic haematite particles, *Phys. Earth planet. Inter.*, 76, 123–130.
- Pike, C.R., Roberts, A.P. & Verosub, K.L., 1999. Characterizing interactions in fine magnetic particle systems using first order reversal curves, *J. appl. Phys.*, **85**, 6660.
- Roberts, A.P., Pike, C.R. & Verosub, K.L., 2000. First-order reversal curve diagrams: A new tool for characterizing the magnetic properties of natural samples, *J. geophys. Res.*, **105**(B12), 28 461–28 475.
- Speliotis, D.E. & Lynch, W., 1991. Magnetic interaction in particulate and thin-film recording media, J. appl. Phys., 69, 4496–4498.
- Stancu, A., Pike, C., Stoleriu, L., Postolache, P. & Cimpoesu, D., 2003. Micromagnetic and Preisach analysis of the First Order Reversal Curves (FORC) diagram, *J. appl. Phys.*, **93**(10), 6620–6622.
- Thellier, E. & Thellier, O., 1959. Sur l'intensité du champ magnétique terrestre dans le passé historique et géologique, Ann. Géophys., 15, 285– 376.
- Wohlfahrt, E.P., 1958. Relations between different modes of the acquisition of the remanent magnetization of ferromagnetic particles, *J. appl. Phys.*, 29, 595–596.