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Parabolic Field Dependence of Kinks Occurring in the Logarithmic Time Plots of Viscous Magnetization*

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Abstract. Viscous magnetization of a series of quartz dolerite samples was measured in different constant magnetic fields. Starting from the demagnetized state, each curve of magnetization versus logarithm of time shows a distinct kink in the expected straight line. The kink times depend parabolically on the magnitude of the constant magnetic field in which the viscous magnetization was acquired. An attempt to interpret this result theoretically considers the viscous magnetization in these rocks to be carried essentially by multidomain grains.

Key words: Viscous magnetization – Relaxation mechanism – Magnetization process – Palaeomagnetism – Magnetostriction.

1. Introduction

During an earlier study (Heller and Markert, 1973) experiments on the time dependence of viscous magnetization (VM) were carried out in order to determine the age of the viscous component of natural remanent magnetization. Certain quartz dolerite samples taken from the Roman Wall in northern England showed a strong viscous component aligned more or less parallel to the present earth's field.

The logarithmic time plots of viscous magnetization of these samples measured in different constant magnetic fields, always displayed kinks which occurred at different times depending on the amplitude of the applied field, and which marked a sudden increase of the otherwise strictly linear viscosity rate.

For the dating problem it is essential to know which part of the viscosity slope has to be applied to give the proper viscosity constant for the age calculations. On the other hand the mechanisms causing the VM-kinks are also of general interest as they probably lead to a better understanding of the nature of viscous magnetization in rocks and the processes involved in its generation.

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2. Experimental Procedure, Samples and Results

The experimental device is simple and comprises a field coil, an extremely constant dc-power supply (variation $<10^{-5}$), a fluxgate gradiometer system (to measure the sample's stray field) and a recorder which is switched on automatically at the moment when the applied dc-field reaches its constant value. The measurements usually were done at room temperature.One series of measurements was performed at 6° C by using a water cooled field coil. In this case care was taken to achieve thermal equilibrium between sample and field coil.

The samples were discs of quartz dolerite, 2.5 cm in diameter and varying in thickness from 1 cm to 3 cm. They contain about 5% by volume of titanomagnetite ore grains, the visible size of which ranges from less than 5 μm up to a few millimeters. Electron microprobe analysis showed the titanium content to amount to $x=0.5-0.6$ in the system $(1-x)\ Fe_3O_4 \cdot x FeTiO_4$, whereas Curie temperatures measured in air are around 550° C (for more details see Heller and Markert, 1973).

All VM-measurements started from the demagnetized state which was achieved by ac-demagnetization in maximum fields between 1 kOe and 3 kOe amplitude. Some representative curves of a great number of experimental results are collected in Fig. 1. It shows two series of logarithmic time plots of viscous magnetization of samples VRW 123 BD and VRW 106 AA at various dc-field amplitudes H_a which were raised step by step. Sample VRW 123 BD was measured at room temperature and sample VRW 106 AA at 6° C using the water cooled coil. All these VM-curves are strictly linear throughout the experimental measuring time (up to 1,200 sec) except at one point where a sudden increase of the viscosity slope is observed causing a distinct kink in the viscosity plot. These kink times depend on the field amplitude and pass through a minimum with increasing dc-field for each sample.

In Fig. 2 the kink times (plotted on a logarithmic scale) of several samples are determined as a function of the respective dc-field. Additionally the bulk coercive force of each sample is marked by an arrow. For each sample the logarithm of the kink times clearly shows an approximately parabolic field dependence. The fields at which the minimum kink times are obtained, appear to agree closely with the respective bulk coercive forces when observing VM at room temperature. At somewhat lower measuring temperatures (sample VRW 106 AA) the kink times seem to increase remarkably and the correlation between bulk coercivity (measured at room temperature) and the field which produces the minimum kink time appears to be less distinct.

3. Fundamental Theory

In 1949 Néel published a logarithmic time law for the representation of viscous magnetization (VM)

$$\Delta J=\chi_{\mathrm{irr}} \cdot S_v \cdot (Q+\ln(t)), \tag{1}$$

where χ_{irr} denotes the irreversible susceptibility, S_v a viscosity factor, $Q=-\ln(t_0)$ a logarithmic time constant, t the time interval considered and ΔJ the viscous

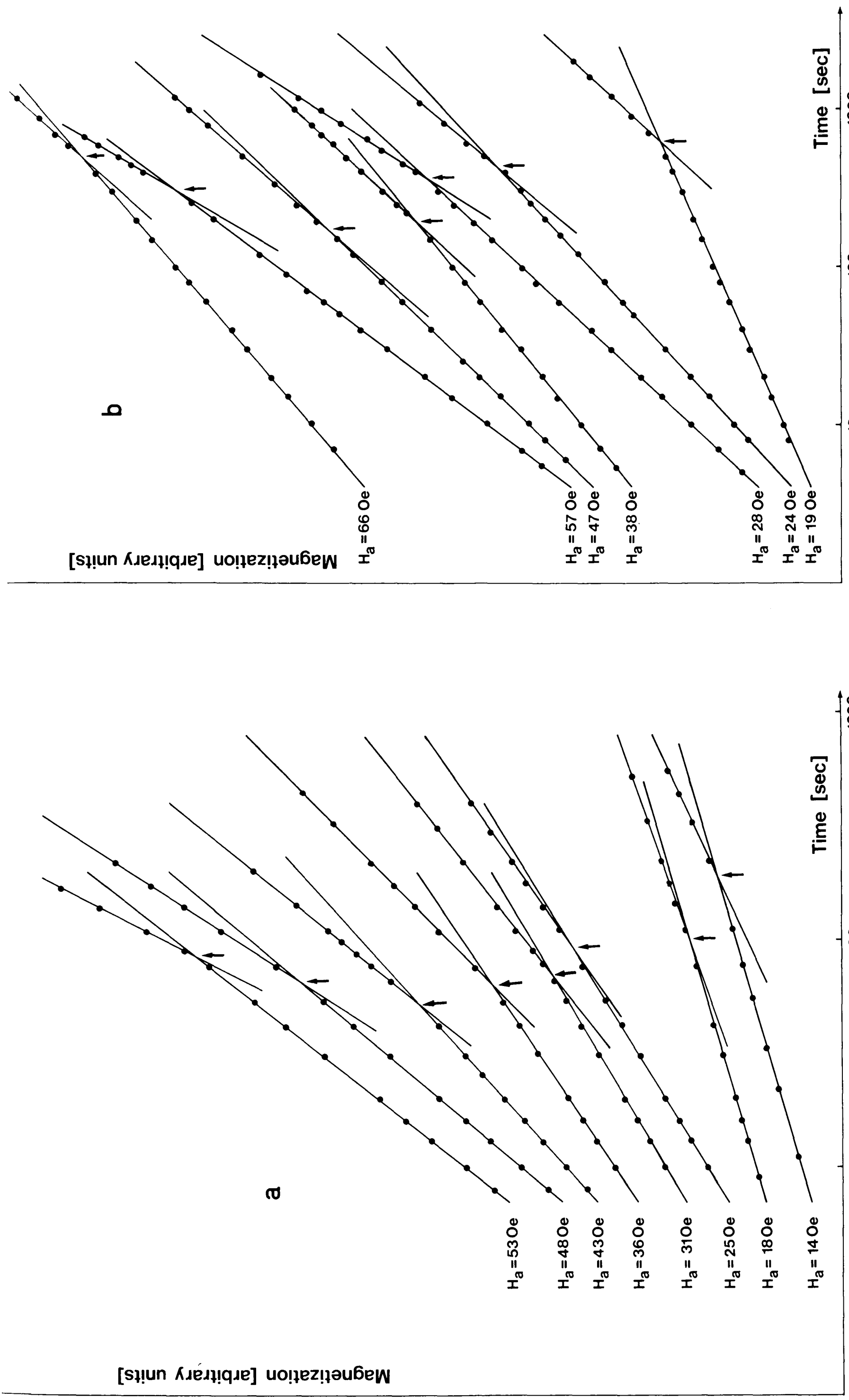

Fig. 1. Two series of logarithmic time plots of viscous magnetization of samples VRW 123BD (a) and VRW 106AA (b). The applied dc-field H_a plays the role of the series parameter. Sample VRW 123BD was measured at room temperature and sample VRW 106AA at 6° C. For better representation of their kinks all successive curves have been shifted along the magnetization axis: therefore the magnetization values of the kinks are arbitrary, whereas the systematic variation of the slopes as well as that of the kink angles is real (although given in relative units)

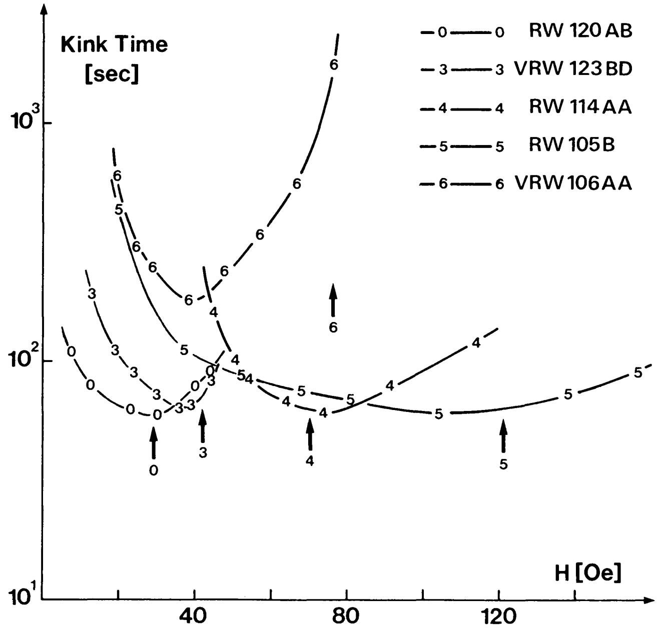

Fig. 2. Parabolic field dependence of the kink times of several samples. All samples plotted have been measured at room temperature except sample VRW 106 AA which has been measured at 6° C. The arrows mark the bulk coercive force of the samples

magnetization caused by thermally activated internal field fluctuations of amplitude

$$H_f = S_v \cdot (Q + \ln(t)). \tag{2}$$

This simple relationship has turned out to be very successful as a quantitative first order approximation of the description of magnetic viscosity phenomena.

Nearly simultaneously a more general formula was proposed by Street and Woolley (1949a):

$$dJ/dt = (\bar{i} \cdot p \cdot k \cdot T/t) \cdot (\exp(-\lambda_0 \cdot t) - \exp(-C \cdot t)). \tag{3}$$

In this expression the symbol $\bar{i}$ represents the average amount contributed to the magnetization in the direction of the applied field by activation of one magnetic domain. The factor p gives the value of the distribution function f of the magnetic domains involved in the magnetization process. f is assumed to be independent of the activation energy E; k denotes the Boltzmann's constant, T the absolute temperature, C a frequency constant, and λ_0 a frequency defined by

$$\lambda_0 = C \cdot \exp(-E_0/k \cdot T), \tag{4}$$

with E_0 being the upper limit of activation energies.

The general solution of Eq. (3) is of the form illustrated in Fig. 3 by an example of Street and Woolley. Two important approximate solutions within certain time intervals are:

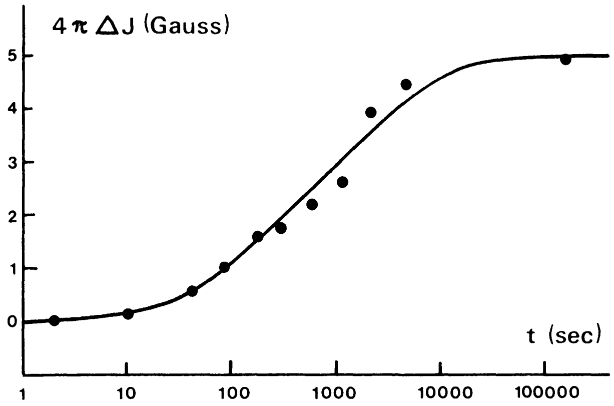

Fig. 3. Magnetic viscosity in Mn – Zn ferrite according to Street and Woolley (1949b). The full line is calculated from Eq. (3), the experimental points are obtained by Snoek (1947)

1. a logarithmic relationship

$$\Delta J = \bar{i}\cdot p\cdot k\cdot T\cdot \ln(t) + \text{const.} \tag{5}$$

which follows from Eq. (3) after integration with respect to *t*:

$$dJ = (\bar{i}\cdot p\cdot k\cdot T)\cdot \int t^{-1}\cdot \{\exp(-\lambda_0\cdot t) - \exp(-C\cdot t)\}\, dt, \tag{5a}$$

if two additional conditions are fulfilled:

$$\int_{t_c}^{\infty} t^{-1}\cdot exp(-C\cdot t_0)\, dt \ll \int_{t_c}^{\infty} t^{-1}\cdot \exp(-\lambda_0\cdot t_0)\, dt \tag{6a}$$

and

$$\lambda_0\cdot t_0 \ll 1 \tag{6b}$$

with

$$t_c < t < t_0 \tag{6c}$$

marking the interval of validity of Eq. (5).

2. a linear expression of type

$$\Delta J = \bar{i}\cdot p\cdot k\cdot T\cdot (C\cdot t) + \text{const.}, \tag{7}$$

which yields a good approximation, if

$$\lambda_0\cdot t \ll C\cdot t \ll 1, \tag{8}$$

and if therefore $\{\exp(-\lambda_0\cdot t) - \exp(-C\cdot t)\}$ can be estimated to result in

$$\{\exp(-\lambda_0\cdot t) - \exp(-C\cdot t)\} \approx \{1 - \exp(-C\cdot t)\} \approx C\cdot t \ll 1. \tag{8a}$$

In other words: in its very beginning the theoretical curve of Fig. 3 can be idealized linearly using Eq. (7), whereas the following steeper part may be described logarithmically in terms of Eq. (5). As actual experiments usually take place within the above limited time interval $t_c < t < t_0$, it does not matter in practice whether to apply Néel's or Street and Woolley's law.

4. Discussion

At first one might try to interpret the above results in terms of Eq. (3) assuming the low-time flexure of its graphic representation (cf. Fig. 1) to be rather sharp and to give the kinks observed in the logarithmic time plots of our measurements. If this were true, according to Eqs. (5) to (8) the logarithmic time plots of VM should obey a linear time law throughout the time range $0 \leqq t \ll t_c$. But in our experiments we find a strictly logarithmic time dependence of VM below as well as above the kink times observed. Therefore Eq. (3) does not fit our experimental evidence.

We think that the phenomena observed in our studies can only be described by superposition of more than one independent logarithmic relaxation mechanism. Obviously a second relaxation mechanism is starting with its "logarithmically linear" phase just after a certain expectation time t_c (see Eq. 6c) which may be equivalent to the kink time occurring in the above shown time plots.

a) *Single or Multidomain VM.* Before asking for the nature of these mechanisms and their field dependence, we should find an answer to the question whether single- or multi-domain grains cause the observed after-effects in our samples. We therefore should find experimental criteria which indicate either multidomain or single domain behaviour of their viscous magnetization.

The application of a test which was described by Johnson *et al.* (1975), unfortunately yields ambiguous results, as shown in Fig. 4. This test is used to identify single – or multidomain grains as carriers of remanent magnetization from the stability of IRM and ARM, against alternating demagnetizing fields. The stability trend of ARM produced by different constant fields contradicts the stability comparison between ARM and IRM. For single-domain grains IRM should be less stable than ARM generated in different low constant fields, whereas the ARM stability itself should decrease with increasing dc-field. For multidomain grains the opposite behaviour should hold true. The contradiction shown in Fig. 4 possibly indicates a mixture of single-domain (pseudosingle domain?) and multidomain grains and makes a strong contribution to anhysteretic and isothermal magnetization processes by multidomain particles probable.

Another test which, especially from viscous magnetization processes, may distinguish between single- and multidomain grains, was suggested by Pfrenger (1966). This test might be more helpful to our problem than the previous method, because it is related to the viscosity effects themselves.

Pfrenger has shown theoretically that the viscosity factor of single-domain particles *decreases* with increasing applied constant field. Denoting ϕ as the average angle between the direction of spontaneous magnetization (J_s) of non-interacting grains and the applied field, then $\overline{\cos\phi}$ increases with increasing field and S_v

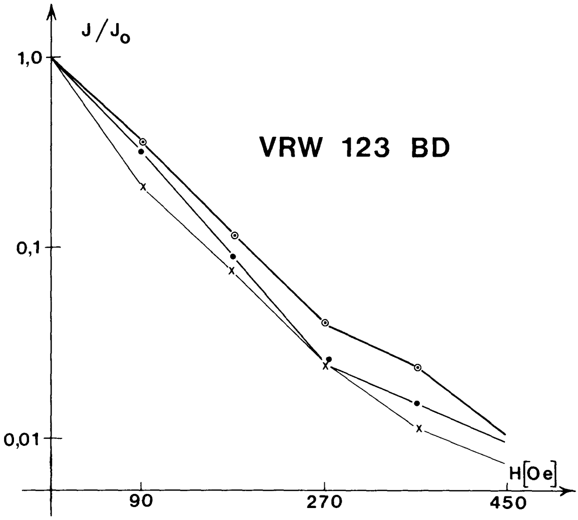

Fig. 4. Test for single domain or multidomain behaviour of sample VRW 123BD using the method described by Johnson *et al.* (1975). ARM given by 0.3 Oe (indicated by dots) respectively 3 Oe (indicated by encircled dots) constant field, shows contradictory stability trend compared to the ARM/IRM stability ratio. IRM is indicated by crosses

decreases according to

$$S_v = \frac{k \cdot T}{J_s \cdot \bar{v} \cdot \overline{\cos \phi}}, \tag{9}$$

where k denotes Boltzmann's constant, T the absolute temperature, and $\bar{v}$ the average particle volume of the particles participating.

When dealing with multidomain grains, however, S_v will *increase* with increasing applied constant field. At very low fields only displacements of domain walls with favorable orientation (ψ) to the field ($\sin \psi \cong 1$) are expected to cause magnetic aftereffects, whereas with increasing field domain walls whose normals lie close to the field direction ($\sin \psi \cong 0$), are displaced, too. Therefore we get according to Pfrenger

$$S_v = \frac{k \cdot T}{J_s \cdot \sin \psi} \cdot \frac{1}{w}, \tag{10}$$

where w is a factor which mainly depends on the total volume of the wall.

Fig. 5 shows the field dependence of S_v for one of our samples. Both viscosity factors, S_{v_1} denoting the factor before the respective kink is occurring and S_{v_2} the factor after it, increase almost linearly within the error limits with increasing field. Therefore it is suggested according to the theory mentioned above, that the viscous part of magnetization is carried by multidomain grains.

There are other theoretical reasons, too, which oppose an interpretation of our experimental data by assuming single-domain grains as the main carriers of VM. According to Kneller (1966), the relaxation time τ_{H_a} of field dependent

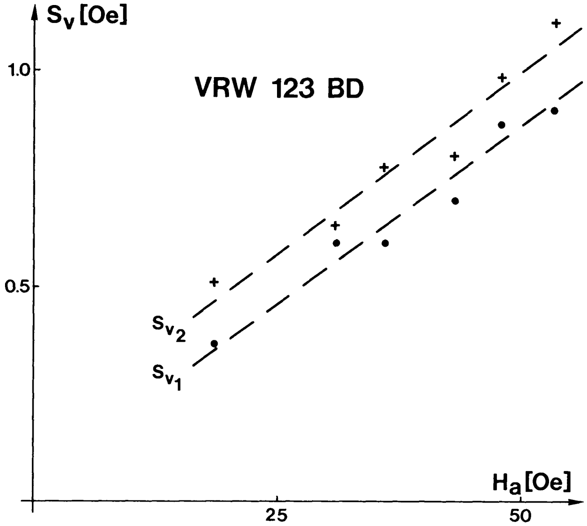

Fig. 5. Field dependence of the viscosity factors S_{v_1} (before kink) and S_{v_2} (after kink). Both factors increase with increasing constant field, thus according to Pfrenger (1966) indicating multidomain particles as carriers of VM

magnetization processes of non-interacting single domain particles can be described by

$$\tau_{Ha}^{-1} = 2 \cdot f_{H_a} \cdot \exp\left\{ -\frac{V \cdot J_s}{2 \cdot kT} \cdot H_k \cdot (1 + H_a^2/H_k^2) \right\} \cdot \cosh\left(\frac{V \cdot J_s}{k \cdot T} \cdot H_a \right), \qquad (11)$$

where V is the particle volume, H_k the anisotropy field which equals the coercivity H_c, and H_a the applied constant field. f_{H_a} is a frequency factor depending on both, H_k and H_a. It approaches zero for $H_a = H_k$. In other words: τ_{H_a} is found to have a singularity for $H_a = H_c$. Therefore in rocks which contain mainly single domain grains covering a certain coercivity spectrum range, it is expected that the kink times of logarithmic VM-plots reach a maximum value at a field which is in the range of the bulk coercivity.

Our observations display just the opposite features and are not compatible with single domain carriers. We suppose from this evidence and from the above shown field dependence of S_v that the viscous magnetization of the quartz dolerite samples investigated is carried mainly by multidomain grains.

b) *Multidomain Interpretation.* For the interpretation of the curves given in Figs. 1 and 2, we first would like to confine the discussion to field strengths $H_a < H_c$, with H_c denoting the bulk coercivity. As we have to deal mainly with multidomain grains, it should be possible to understand the occurrence of the kink times and their field dependence in terms of a magnetization theory which has been proposed by Markert (1970). It is shown by this theory that two irreversible relaxation mechanisms may contribute to viscous magnetization and hysteresis losses of single-slip deformed nickel single crystals, and also of magnetite and titanomagnetite multidomain ore grains.

The first mechanism is based on repulsive interaction forces acting between a 180°-domain wall and suitably oriented dislocations. Such a 180°-domain wall, moving towards a hindering dislocation barrier under the influence of an external field, can push and shift the barrier in front of it across a distance d after a certain expectation time t_{e_1} (which corresponds to a first t_c in the sense of Eq. (6c)), until the dislocations will link again with other lattice defects. The coupled motion of such an interacting system represents an irreversible relaxation process which is generated with the assistance of thermal fluctuations.

The second relaxation mechanism is the well known Barkhausen effect which prominently sets in after an expectation time t_{e_2} (which is equivalent to a t_{c_2}) as soon as the sum of the external field H_a and an internal fluctuation field H_i reaches a critical value just high enough to enable even the representative ones among all of the 180°-domain walls to jump over their hindering dislocation barriers, i.e. if (H_a+H_i) just equals the coercivity H_c.

It was intimated by Markert (1970) that t_{e_1}, the expectation time of the first mechanism, is far too small to be actually observed in our experiments. This is in agreement with our results, because otherwise we should find another low-time kink on the logarithmic time plots.

On the other hand t_{e_2} decreases rapidly with increasing H_a. Therefore throughout the range $H_a \leqq H_c$ we can explain the decrease of kink time with increasing applied field, by the second mechanism, but this model cannot explain how magnetic dc-fields greater than H_a can cause increasing expectation times of a relaxation process to set in. Thus a third relaxation mechanism has to be introduced, the activation energy of which increases with increasing applied field. We would like to suggest the following mechanism as a third possible process.

In ferrite single crystals, particularly in natural titanomagnetites a great density of point defects such as lattice defects, interstitials and impurity atoms is found. Because of their magnetostrictive lattice deformation the domain walls are among the energetically most favourable places for point defects which therefore show a tendency to diffuse into the domain walls and to damp their mobility. An analogous mechanism is applicable to dislocations. According to our above considerations the domain walls can shift suitable primary dislocations in front of them (first relaxation mechanism). But there are two damping mechanisms for the domain walls: first a direct damping by means of diffusing point defects (after-effect of Richter type) and second an indirect damping caused by point defects which move towards the dislocations lowering their mobility. Among these point defects anisotropic configurations like "dipoles" or pairs of interstitials are of particular interest because of the fact that their interaction with dislocations and domain walls depends on the mutual orientation between dipole axes, dislocations and domain walls.

The dipoles for instance may vary their orientation due to exchanges of places. But the activation energy of these exchange processes increases exponentially with increasing volume magnetostriction, i.e. with increasing magnetic dc-field, if the volume magnetostriction is positive, and with the square of increasing magnetization. Thus exchanges of places causing an additional mobility of dislocations and domain walls can set in after an expectation time t_{e_3} which increases with increasing applied field.

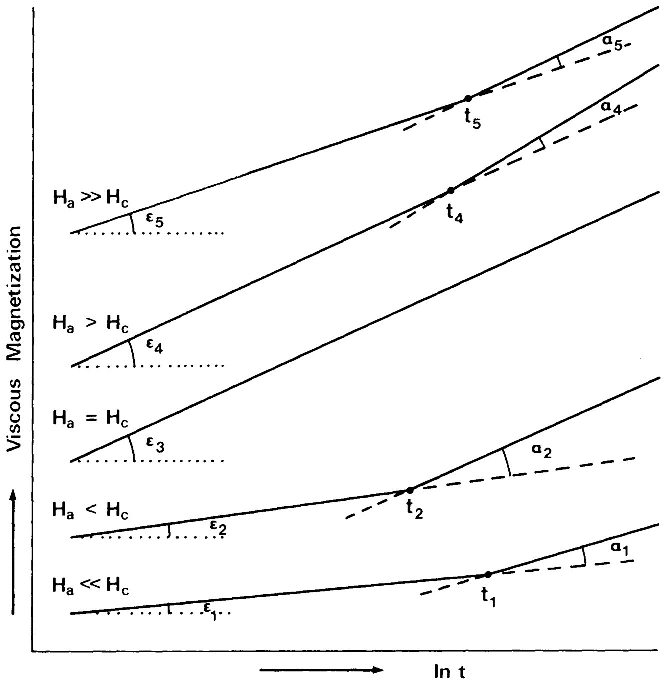

Fig. 6. Schematic diagram of field dependent kink times, kink angles and slopes of the logarithmic time plots of viscous magnetization to be expected due to the model of relaxation mechanisms suggested in points 1 to 3 of the discussion. Angles α_i and ε_i are assumed to obey the following rules: $\alpha_1 < \alpha_2$ and $\varepsilon_1 < \varepsilon_2$ according to point 1, $\varepsilon_4 = \varepsilon_3 = \alpha_2 + \varepsilon_2$ according to points 1 and 2, $\alpha_4 \cong \alpha_5$ and $\varepsilon_5 < \varepsilon_4$ according to points 1 and 3

If the above interpretation is true, then in an idealized case we should find the following three types of logarithmic time plots of VM which schematically are illustrated in Fig. 6:

1. If the dc-field ranges in the interval $0 < H_a < H_c$, the kinks mark the change from the logarithmic phase of relaxation mechanism one to the superposition of the logarithmic phases of relaxation mechanisms one and two (Barkhausen effect). The slope below the kink in a given time plot of viscous magnetization therefore characterizes the first relaxation process and its viscosity constant. Above the kink the new slope is a measure of the total viscosity constant of both relaxation processes. As the Barkhausen mechanism prominently sets in at about $0.6 \cdot H_c$ and increases to a maximum at $H_a \cong H_c$, the total viscosity after the kink should increase with increasing H_a, whereas at the same time the kink time decreases towards zero.

2. At $H_a = H_c$ the Barkhausen effect and also the total viscosity are passing through a maximum. Both kink times t_{e_1} and t_{e_2} approach zero.

3. For $H_a > H_c$ the Barkhausen effect starts to decrease and the viscosity constant S_v below the kink point also may show a tendency to decrease with

increasing H_a. As only the expectation time but not the intensity of the logarithmic phase of the third relaxation mechanism was shown to depend on the volume magnetostriction, the additional contribution to the viscosity due to the third relaxation process after the kink point should turn out to be more or less constant, i.e. with increasing H_a the kink angle α (Fig. 6) should not change.

c) *Alternative Interpretation.* The interpretation given above fits our data, but we are aware that there are possibly alternative or modifying models which also might be able to explain the parabolic field dependence of the kink times. For instance, we might think of a mixture of single and multidomain grains – perhaps caused by oxidation. Kinks have been observed in those rocks by Creer, Petherbridge and Petersen (1970). These authors, however, did not find straight forward relations between the oxidation state and the development of kinks, and we cannot see how such a mixture would display the kink time characteristics we have observed in our rocks.

d) *Application.* Regarding the age dating problem mentioned in paragraph 1 and relying on the evidence stated by Figs. 1 and 2, it seems to be clear that a dating method like that one used by Heller and Markert (1973) has to use the gradient of the viscosity slope which is observed before the kink point as the appropriate viscosity constant S_v because of the low magnitude of the earth's magnetic field. From Figs. 1 and 2 it is evident that in very low fields the kinks are expected to happen only after very long expectation times. This is in coincidence with measurements done by Kent and Lowrie (1974) on fine-grained deep sea sediments in a 1 Oe field over as much as 1,000 h. These authors did not find any kinks during their observations concerned with the increase of remanent magnetism with time.

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Postulated Rotation of Corsica not Confirmed by New Palaeomagnetic Data

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Abstract. Magneto-mineralogical studies of Corsican rhyolitic/andesitic rocks of late Carboniferous-Permian age have revealed an extensive low temperature (weathering) alteration of the primary oxides. In line with this observation analysis of the palaeomagnetic record suggests that at least a substantial part of the "stable" remanence is of chemical origin, probably dating back to the early Tertiary when the West Mediterranean region was subjected to marked uplift and erosion. The relatively stable bulk remanence is composite, constituting both normal and reverse components of magnetization. However, careful demagnetization does not support previous suggestions (based on the same rock formations) of a stable component of magnetization with south-southeasterly declination. Therefore, the idea of a certain anticlockwise rotation of Corsica, detectable by palaeomagnetic means, does not seem to have a firm basis.

Key words: Palaeomagnetism – Rock magnetism – Possible rotation of Corsica.

1. Introduction

The origin of small ocean basins like those of the West Mediterranean have been a matter of speculation among earth scientists for a long time. Such basins which to a large extent seem to be located between the compressional boundaries of larger crustal blocks may have developed through two entirely different processes: a) by subsidence of previous continental areas (Klemme, 1958; Van Bemmelen, 1969) which subsequently have been turned into a "transitional" type of crust (Menard, 1967) through vertical assimilation by the upper mantle, or b) by crustal separation through the mechanism of sea floor spreading (Vogt *et al.*, 1971; Le Pichon *et al.*, 1971). Theoretically, these mechanisms seem equally plausible and they may both have taken part in the development of at least some of the smaller ocean basins.