



Measurement of magnetic viscosity in a Stoner–Wohlfarth material

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Abstract

A Stoner–Wohlfarth model has been used to simulate conventional magnetic viscosity experiments. The magnetic viscosity parameter Λ was found to vary depending on the method used for measurement of the irreversible magnetisation. The difference in Λ is found to be related to a parameter η which describes the dependence of reversible magnetisation on irreversible magnetisation. The correct value of Λ at any field has been related to the population of particles which reverse at that field and it is revealed that distributions of volume and orientation can obscure the interpretation of the measured Λ . Thus the measurement of Λ as an indicator of reversal mechanism is questioned.

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1. Introduction

The first observation of the time dependence of magnetisation, or magnetic viscosity, was reported by Ewing [1]. Later the work of Street and Woolley [2,3] and Street, Woolley and Smith [4] related this phenomenon to the intrinsic properties of the magnetic material under study. This was done by considering the thermal activation of metastable magnetisation states over energy barriers. For a ‘flat’ distribution of energy barriers at a fixed external field the magnetisation decays with time as:

$$M(t) = A + S \ln t. \quad (1)$$

Street, Woolley and Smith [4] showed that the

magnetic viscosity coefficient S was related to the irreversible susceptibility by:

$$S = \Lambda \chi_{\text{irr}}, \quad (2)$$

where both S and χ_{irr} have been corrected for demagnetisation effects. Λ is a parameter which is dependent on both field and magnetisation. Eq. (2) summarised the key magnetic viscosity parameters and provided the basis for all work in magnetic viscosity for the next 40 years.

Assuming that each successful activation event results in the same change in magnetisation then Eq. (2) can be used to relate Λ to the energy barriers of reversal within the material by [4,5]:

$$\Lambda = \frac{-kT}{\partial E / \partial H|_{M_{\text{irr}}}} \quad (3)$$

where T is the absolute temperature, k is Boltz-

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mann's constant and $(\partial E/\partial H)$ is the rate of change of the energy barrier with field. Eq. (3) is the basis for the interest in magnetic viscosity measurements because it enables such measurements to be used as a probe of the rate of change of energy barrier with field, which is directly determined by the mechanism of reversal.

The parameters in Eq. (2) are calculated from conventional magnetic viscosity experiment data. The experiment consists of starting with the material in a known state of magnetisation, usually positive saturation, then applying a fixed reverse external field and measuring the decay of magnetisation with time, from which S at that field can be calculated. This procedure is then repeated for progressively larger reverse fields along the demagnetisation curve.

Two experimental methods are used for the measurement of χ_{irr} for use in Eq. (2). (a) χ_{irr} is evaluated as the derivative of the dc demagnetisation curve, obtained by measuring the remanence after applying progressively larger reverse fields following positive saturation. (b) χ_{irr} is determined as the difference $\chi_{\text{tot}} - \chi_{\text{rev}}$ where χ_{tot} is derived from the hysteresis loop and χ_{rev} is measured in the manner of Tebble and Corner [6] by defining it as the mean slope of a small minor loop performed at the end of the fixed external field portion of the experiment. Both definitions of χ_{irr} assume that irreversible processes are not available to activate during removal of the field. Method (b) is more local to the point of measurement.

Measured values of Λ using these two methods of measuring χ_{irr} have been related to the energy barriers of reversal (Eq. 3). This information has been used to compare magnetisation mechanisms of experimental materials with theoretical predictions [7,8], to study the angular dependence of material properties [9] and to study the energy barriers of reversal in a variety of materials [4,10–12].

Since magnetisation is time dependent, measured values of χ_{irr} are also functions of time. For example values of χ_{irr} determined by both methods (a) and (b) depend on the time the reverse field is applied before the recoil or minor loop is traversed. Neglect of the time dependence of χ_{irr} may lead to very significant errors in the determination of the functional dependencies of Λ on applied field and magnetisation.

The problem of the time dependence of χ_{irr} was solved by Estrin, McCormick and Street [13]. By analogy with plastic deformation they developed a phenomenological description of magnetic viscosity which defined how the irreversible susceptibility should be measured. In addition this approach did not require the assumptions made by Street, Woolley and Smith [4] and yielded Eqs. (2) and (3) as special cases. The constitutive equation, under constant temperature conditions, relates the internal magnetic field H_i to the irreversible magnetisation M_{irr} and the time rate of change of irreversible magnetisation \dot{M}_{irr} . In differential form this can be represented as:

$$dH_i = \frac{1}{\chi_{\text{irr}}^i} dM_{\text{irr}} + \Lambda d \ln \dot{M}_{\text{irr}}, \quad (4)$$

where

$$\chi_{\text{irr}}^i = \left. \frac{dM_{\text{irr}}}{dH_i} \right|_{\dot{M}_{\text{irr}}} \quad (5)$$

is the intrinsic irreversible susceptibility and

$$\Lambda = \left. \frac{dH_i}{d \ln \dot{M}_{\text{irr}}} \right|_{M_{\text{irr}}} \quad (6)$$

It can be shown for a conventional magnetic viscosity experiment in which the applied field is changed discontinuously to a fixed value that the constitutive equation (Eq. (4)) leads to an expression for the time dependence of magnetisation of the form [13]:

$$M_{\text{tot}}(t) = A + S \ln(t + t_0), \quad (7)$$

where t_0 is a constant equal to S/\dot{M}_0 and \dot{M}_0 is the initial rate of change of magnetisation. The assumptions made are:

- 1) both χ_{irr}^i and Λ are independent of magnetisation during a viscosity experiment,
- 2) the intrinsic reversible susceptibility (χ_{rev}^i) is independent of time, and
- 3) $\dot{M}_{\text{rev}} = \chi_{\text{rev}}^i \dot{H}_i$, i.e. that the reversible magnetisation (M_{rev}) is a function of the field only.

The coefficient S in Eq. (7) is:

$$S = \Lambda \chi_{\text{irr}}^i, \quad (8)$$

where both S and χ_{irr}^i are corrected for demagnetisation effects and χ_{irr}^i is measured, as required by Eq. (5), at constant \dot{M}_{irr} . Eq. (8) is equivalent to Eq. (2) and identifies the quantity Λ in terms of Eq. (6).

In general the assumptions 1–3 above are not appropriate and experimental results cannot be analysed in terms of Eq. (8). Eq. (6) provides a method of calculating Λ without making the assumptions above, provided that the irreversible magnetisation can be measured. The only assumption made when using Eq. (6) is that the experiment is performed isothermally, which is true in general but may not be valid when χ_{irr} is large and the magneto-caloric effect significant, e.g. in high coercivity square loop materials [14].

The relation between Λ and the energy barrier, Eq. (3), can also be derived from the phenomenological description of Estrin, McCormick and Street [13] without relying on Eq. (8) and hence the assumptions used in deriving it. The assumption that each activation event results in the same change in magnetisation is, however, still required.

Although the work of Estrin, McCormick and Street [13] defines χ_{irr} precisely through Eq. (5), it does not define how M_{irr} should be measured. How to measure Λ is then similarly uncertain. Two methods can be used to measure M_{irr} , which are based on the two methods (a) and (b) used to measure χ_{irr} . It is the purpose of this paper to examine the effect each of these methods has on the measured value of Λ .

To this end a Stoner–Wohlfarth model [15] has been used to simulate conventional magnetic viscosity experiments. The use of a model, besides removing unknown experimental variables, has allowed calculation of Λ from M_{irr} derived directly from the model in an experimentally inaccessible way, as well as M_{irr} extracted from M_{tot} data which is experimentally accessible. It will be shown that to resolve the differences between the experimentally accessible and the ‘true’ intrinsic Λ account must be taken of the dependence of reversible magnetisation on irreversible magnetisation.

The use of a model has also allowed the value of Λ at any field to be related to the population of particles which reverse at that field. This has implications on the interpretation of Λ when the mechanism of reversal and population of particles are unknown. This is because when a distribution of energy barriers is present in a material, relating Λ to the energy barrier through Eq. (3) is questionable because at each field different populations of parti-

cles, with different $E(H)$ dependencies, may be actively involved in reversal.

2. Extraction of M_{irr} from M_{tot} experimental data

Λ has been calculated in this work using curves of H_i versus M_{irr} according to the method of Folks and Street [16]. In this method Λ is calculated according to the phenomenological representation of Estrin, McCormick and Street [13] from the difference in field ΔH between two points with the same value of M_{irr} but with differing values of \dot{M}_{irr} . Two methods have been used to obtain values of M_{irr} . They are referred to here as the χ_{rev} and the DCD methods. The two methods produce different $H(M_{\text{irr}})$ curves, hence different values for Λ . It is important to note that the calculation of Λ and χ_{irr} requires only knowledge of changes in M_{irr} rather than its absolute magnitude.

2.1. χ_{rev} method

This method consists of measuring a small minor loop (usually of the order of 100 Oe in hard permanent magnet materials) at the end of the constant external field portion of a conventional magnetic viscosity experiment. The mean slope of this minor loop is identified with χ_{rev} and the reversible magnetisation is assumed to be the integral of χ_{rev} around the hysteresis loop. Thus:

$$M_{\text{irr}}(H, t) = M_{\text{tot}}(H, t) - \int_{H_{\text{initial}}}^H \chi_{\text{rev}}(H) dH, \quad (9)$$

where the quantities on the right hand side of the equation are all experimentally measurable and H_{initial} is the lowest field value at which χ_{rev} data has been measured. It is nominally assumed at H_{initial} that $M_{\text{rev}} = 0$. This assumption is of little consequence in light of the importance of changes in M_{irr} rather than its absolute magnitude.

The first assumption made when using this method is that χ_{rev} is not time dependent during a viscosity measurement. Thus the magnetisation changes during viscosity all result from changes in irreversible magnetisation, i.e. that $\dot{M}_{\text{tot}} = \dot{M}_{\text{irr}}$ during the fixed external field portion of the experiment. The second

assumption is that M_{rev} is a function of field only. This second assumption means that M_{rev} is a monotonically increasing function of field because χ_{rev} can never be negative. The χ_{rev} method has the advantage that it is a local measurement of the reversible susceptibility and hence a local measurement of M_{irr} and so is useful when measuring materials which may have irreversible processes occurring along recoil loops, such as exchange spring materials [17].

In this description of M_{irr} , χ_{irr} is related to $\chi_{\text{tot}} - \chi_{\text{rev}}$ except that χ_{tot} must be measured at constant \dot{M}_{irr} . Excepting this last qualification, the χ_{rev} method is analogous to that method used by Givord et al. [8,10,11] and Street and Woolley [2].

2.2. DCD method

In this method M_{irr} at the end of a viscosity experiment is defined to be equal to the value of the remanent magnetisation when the field is removed. This assumes that M_{irr} is constant during the recoil loop to zero internal field. The reversible magnetisation is assumed to be constant during a viscosity measurement and equal to the difference in M_{tot} between the end of viscosity and when the internal field is removed. Thus during a conventional magnetic viscosity experiment the irreversible magnetisation as a function of time is:

$$\begin{aligned} M_{\text{irr}}(H, t) &= M_{\text{tot}}(H, t) - M_{\text{rev}}(H) \\ &= M_{\text{tot}}(H, t) - [M_{\text{tot}}(H, t_{\text{max}}) - M_{\text{tot}}(0, t_{\text{max}})], \end{aligned} \quad (10)$$

where t_{max} is the time at the end of the viscosity. Eq. (10) requires that during the constant external field portion of the experiment $\dot{M}_{\text{tot}} = \dot{M}_{\text{irr}}$, the same assumption as is made in the χ_{rev} method.

The DCD method requires ramping to zero internal field at the end of each viscosity step. It has the disadvantage that it is more time consuming compared to the χ_{rev} method and requires that the irreversible magnetisation and temperature are constant during recoil. It has the advantage that no implicit assumption is made about the form of M_{rev} . Experimental data for some materials using this method

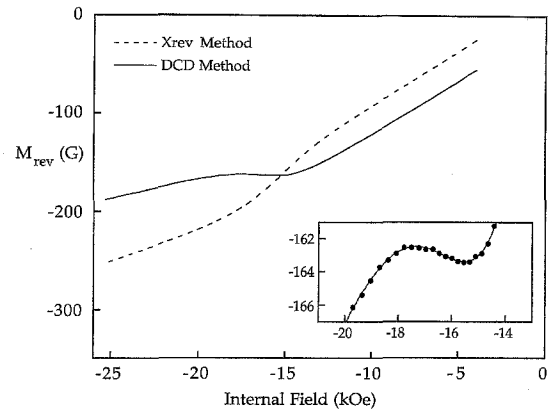


Fig. 1. Reversible magnetisation (M_{rev}) on the demagnetisation curve measured for a sphere of MQ1 by the χ_{rev} and DCD methods (see text). Inset: Enlargement of the DCD method measurement around coercivity (15 kOe) showing the non-monotonic behaviour.

indicate that M_{rev} is not a monotonic function of H , as assumed in the χ_{rev} method. Fig. 1 shows M_{rev} measured using the DCD method and the χ_{rev} method as a function of reversing field for a sphere of MQ1 and illustrates that M_{rev} need not be a monotonically increasing function of field as shown in the inset to Fig. 1.

Excepting the requirement to measure χ_{irr} at constant \dot{M}_{irr} this method has been used by Ferguson et al. [12] and de Witte et al. [18].

3. The definition of M_{irr}

It is important to stress the difference between the implied definitions of M_{irr} when using the χ_{rev} and DCD methods for extracting M_{irr} from measured M_{tot} data.

The χ_{rev} method uses only measurements of the reversible susceptibility to estimate changes in reversible magnetisation which occur around the hysteresis loop. It is thus assumed that χ_{rev} does not depend on the value of M_{irr} but only on the field applied. Consideration of the Stoner–Wohlfarth model shows this to be false. At any field below the critical field a particle may either be on the upper branch or the lower branch of the hysteresis curve. The reversible susceptibility at these two positions is not the same, excepting the special cases for a

particle with easy axis oriented at 0° or 90° to the field. Thus if the particle was to be thermally activated between the upper and the lower branch in a constant external field, χ_{rev} would change even though the field had been held constant. The reversible magnetisation must depend on the state of the particle, represented by M_{irr} , as well as the field. For an ensemble, as the particles thermally activate in a fixed field, the reversible susceptibility, which will be the average χ_{rev} of the particles in the ensemble, will change as the irreversible magnetisation changes.

During viscosity in high energy permanent magnets the change in χ_{rev} with M_{irr} is smaller than the error of measurement of χ_{rev} , but during field changes M_{irr} can change significantly and hence χ_{rev} is also affected not only by the field change but by the changes in M_{irr} . Thus, there is some doubt as to the validity of Eq. (9).

Relating M_{irr} to the internal state of the system under study also takes some care when it is measured using the χ_{rev} method. In a two well system, specifying the state is equivalent to specifying the number of particles in each well. The problem with relating M_{irr} measured by the χ_{rev} method to the state of the system is that M_{rev} will depend not only on the number of particles in each well, but also on how those particles got to that well from some initial state.

This may be illustrated by considering an isotropic Stoner–Wohlfarth ensemble. If the field is ramped from positive to negative saturation the difference in the values of M_{irr} at these two points, using the χ_{rev} method, will be different depending on whether all of the particles reversed at remanence (for example if the field was ramped infinitely slowly) or at each of their critical fields, i.e. following the athermal demagnetisation curve. In essence the change in M_{irr} between any two points will depend on the temperature and the rate of change of field.

The DCD method refers all measurements of M_{irr} back to the remanence of the system after the internal field is removed sufficiently quickly so that no irreversible processes occur during recoil. This is analogous to the method of measuring the components of elastic and plastic strain. It is the description of plastic strain from which the constitutive equation, Eq. (4), was derived by analogy. It is possible to

relate the measurement of ‘irreversible’ magnetisation to the number of particles in each well (of a two well system) because all measurements are made at the same field. This means that any state of the system has a unique value of M_{irr} independent of the path which was taken to arrive at that state. Considering the example of an isotropic Stoner–Wohlfarth ensemble above, the difference in M_{irr} between positive and negative saturation, measured using the DCD method, is independent of the path taken to get between the two points.

‘Irreversible’ magnetisation in this sense is a misnomer because it has no relation to the entropy of the system or any inherent irreversibility of the path. The DCD definition of M_{irr} is much easier than the χ_{rev} definition to relate to the intrinsic energy barriers within the material because it measures the proportion of magnetisation in the two wells at the same field. This proportion can, with suitable assumptions (including the absence of interactions), be related to the number of particles in each well. A complication with the DCD method is that it implies that during a discontinuous change in magnetisation both the ‘reversible’ and ‘irreversible’ components of magnetisation change (see below).

In using the non-interacting model described below it is possible to know the number of particles present in each of the two energy minima. Thus it is possible to calculate the ‘irreversible’ magnetisation defined by the DCD method for any state, independent of the path taken to get there. The χ_{rev} method results in a path dependent estimate of M_{irr} and does not describe the *state* of the system but the *path* taken to get there from some reference state.

Measurement of A is most suited to aiding analysis of energy barrier information, using Eq. (3). Energy barrier information is directly related to the state of the system. The model thus uses the DCD definition as the ‘true’ definition of M_{irr} because it is directly related to the state of the system. The term ‘irreversible’ magnetisation will thus be used subsequently as a convenient label and should not be taken as a measure of thermodynamic irreversibility.

4. Dependence of M_{rev} on M_{irr}

The DCD method of measuring M_{irr} and M_{rev} suggests that M_{rev} is not simply a function of field

only but is also a function of M_{irr} . This is illustrated in the inset to Fig. 1 where, because M_{rev} is not a monotonic function of field it must depend on parameters other than χ_{rev} . In differential form this observation can be expressed phenomenologically as [19]:

$$dM_{\text{rev}} = \chi_{\text{rev}} dH_i + \eta dM_{\text{irr}}, \quad (11)$$

where

$$\chi_{\text{rev}} = \left. \frac{dM_{\text{rev}}}{dH} \right|_{M_{\text{irr}}} \quad (12)$$

and

$$\eta = \left. \frac{dM_{\text{rev}}}{dM_{\text{irr}}} \right|_H. \quad (13)$$

Eq. (11) implies that if M_{irr} displays hysteresis then M_{rev} will also display hysteresis. By defining M_{rev} as in Eq. (11) the assumptions used in deriving $\Lambda = S/\chi_{\text{irr}}$ (Eq. (8)) are not valid. Namely χ_{rev} is not time independent (assumption 2) and M_{rev} is a not function of field only (assumption 3). Using Eq. (11) and the constitutive equation it is possible to derive a correction to Λ measured by Eq. (8) [19]:

$$\Lambda_\eta = \Lambda/(1 + \eta), \quad (14)$$

where $\Lambda = S/\chi_{\text{irr}}$ as in Eq. (8). This implies that the correction to Λ increases with η . The assumption used in this derivation is that Λ_η and χ_{irr} are constant during viscosity (equivalent to assumption 1 used in deriving Eq. (8)).

As an estimate of the magnitude of the correction to Λ in Eq. (14) consider an ensemble of identical single domain, uniaxially anisotropic non-interacting particles [15], with easy axes at an angle of 45° to the field direction. In the absence of thermal activation the coercivity of the ensemble is $H_a/2$, where $H_a = 2K/M_s$ is the anisotropy field. Using the DCD definition of M_{irr} the derivative in Eq. (13) can be found at coercivity, where most thermal activation will occur. At this field the magnetisation vector of the ensemble rotates irreversibly from an angle of 90° to the positive field direction to an angle of 210° . When the moment makes an angle of 90° to the field direction M_{irr} , equal to the remanent magnetisation, is $M_s \cos 45^\circ$. This means $M_{\text{rev}} (= M_{\text{tot}} - M_{\text{irr}})$ is $-M_s \cos 45^\circ$. Similarly when the moment makes an

angle of 210° to the field direction M_{irr} is $M_s \cos 225^\circ$ and M_{rev} is $(M_s \cos 210^\circ - M_s \cos 225^\circ)$. Thus η at this field is:

$$\begin{aligned} \eta &= \left. \frac{\Delta M_{\text{rev}}}{\Delta M_{\text{irr}}} \right|_H \\ &= \frac{(\cos 210^\circ - \cos 225^\circ) - (-\cos 45^\circ)}{(\cos 225^\circ - \cos 45^\circ)} \\ &= -0.388. \end{aligned} \quad (15)$$

The derivative in Eq. (15) is not strictly valid in the athermal case because of the discontinuity at coercivity, however with thermal activation the value of η will tend toward this limit as the field approaches $H_a/2$.

The example above illustrates that the correction to Λ measured near coercivity would be of the order of 40% for an idealised aligned Stoner–Wohlfarth material with easy axes at 45° to the field. Further analysis of Stoner–Wohlfarth particles shows that η , near the switching field of the particle, is zero at an orientation of 0° and becomes increasingly negative as the orientation approaches 90° , as illustrated in Fig. 2.

The importance of η in experimental systems is that when reversible magnetisation is a large proportion of the total magnetisation (e.g. in isotropic systems) neglecting this term can lead to erroneous results for Λ . For example, in a study of isotropic

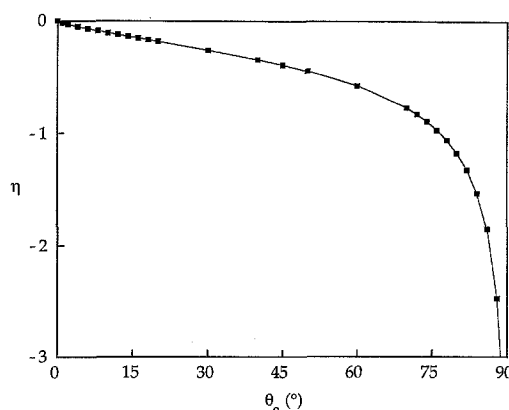


Fig. 2. η calculated at the critical field as a function of easy axis orientation θ_0 of a Stoner–Wohlfarth ensemble. η has an asymptote at 90° .

sintered NdFeB η was found to vary between 1 and -0.2 along the demagnetisation curve [20].

5. The model

The model is based on an ensemble of Stoner–Wohlfarth particles extended to include thermal activation of magnetisation reversal. The time constant τ for decay over an energy barrier ΔE follows an Arrhenius–Néel law:

$$\frac{1}{\tau} = f_0 \exp\left(-\frac{\Delta E}{kT}\right), \quad (16)$$

where f_0 is the attempt frequency, assumed to be independent of field and taken to be 10^9 s^{-1} . The value of f_0 is not well known and depends on many parameters including anisotropy and crystal structure [21–24]. The assumption that f_0 is independent of field is not correct with f_0 being a monotonically decreasing function of field, approaching zero as the field approaches the critical field where the energy barrier to reversal is zero. However for uniform rotation, for the temperatures and times available experimentally, all particles reverse well before this critical field in a region where f_0 is large and only weakly field dependent [21]. Additionally the large value of f_0 means that τ is controlled by the energy barrier in the exponential rather than by the pre-exponential factor. Thus f_0 can be assumed to be constant to a good approximation and 10^9 s^{-1} is a good average value which does not affect the results.

The energy of a uniaxial Stoner–Wohlfarth particle with moment at an angle θ to the field H and easy axis at an angle θ_0 to the field is given by:

$$E/V = K_u \sin^2(\theta - \theta_0) - M_s H \cos \theta, \quad (17)$$

where M_s is the saturation magnetisation per unit volume and K_u is the uniaxial anisotropy energy coefficient. Below a critical field H_{crit} , the energy $E(\theta)$ has two minima, θ_1 in the positive field direction and θ_3 in the negative field direction with an intervening maximum θ_2 . This is shown diagrammatically in Fig. 3. From Eq. (17) the energy barrier can be found numerically as a function of field.

H_{crit} depends on the orientation of the easy axis of the particle to the field as:

$$H_{\text{crit}} = H_a (\cos^{2/3} \theta_0 + \sin^{2/3} \theta_0)^{-3/2}, \quad (18)$$

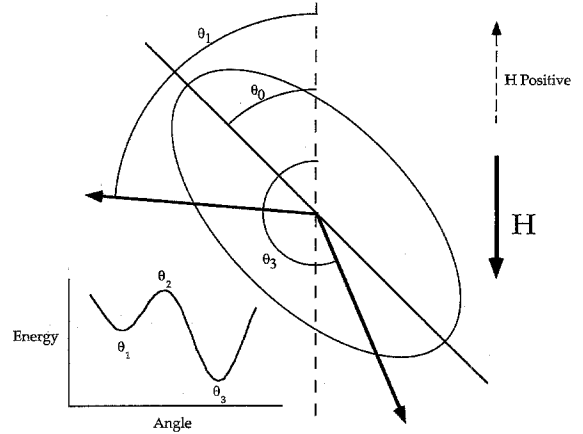


Fig. 3. The definitions of θ_0 , θ_1 , θ_2 and θ_3 used in the model described in the text.

where $H_a = 2K_u/M_s$ is the anisotropy field. H_{crit} is a minimum at $0.5H_a$ for $\theta_0 = 45^\circ$ and rises steeply to H_a near $\theta_0 = 0$ and 90° . For $|H| > H_{\text{crit}}$ only one minimum exists and no thermal activation can occur.

If τ_1 is the time constant for activation out of the energy well at θ_1 and τ_2 is the time constant for activation out of the energy well at θ_3 then the proportion of particles P in the energy minimum at θ_1 is governed by the differential equation:

$$\frac{dP}{dt} = -\frac{1}{\tau_1}P + \frac{1}{\tau_2}(1 - P). \quad (19)$$

In the general case where H is a function of time, Eq. (19) must be solved by numerical methods. In the special case where H is not a function of time, τ is constant and the solution of Eq. (19) is:

$$P(t) = \frac{R}{Q} + K \exp(-Qt), \quad (20)$$

where $Q = 1/\tau_1 + 1/\tau_2$, $R = 1/\tau_2$ and $K = P(0) - R/Q$. R/Q is the thermal equilibrium population distribution between the two energy wells and $P(0)$ is the value of P at $t = 0$.

For a particle of volume V and an easy axis orientation of θ_0 to the field direction, the irreversible magnetisation is defined as the remanent magnetisation of the particle. Thus $M_{\text{irr}} = M_s V \cos \theta_0$ for particles with magnetisation vectors in the minimum at θ_1 and $M_{\text{irr}} = -M_s V \cos \theta_0$ for particles with magnetisation vectors in the minimum at θ_3 .

The irreversible magnetisation of the whole system is then given by:

$$M_{\text{irr}}(H, t) = M_s \int_0^{\pi/2} \int_0^{\infty} F(\theta_0) V f(V) \times \{2P(\theta_0, V, H, t) - 1\} \cos \theta_0 dV d\theta_0, \quad (21)$$

where $F(\theta_0)d\theta_0$ is the number of particles with orientations between θ_0 and $\theta_0 + d\theta_0$ and $f(V)dV$ is the number of particles with volumes between V and $V + dV$.

The reversible magnetisation is defined as:

$$M_{\text{rev}}(H, t) = M_s \int_0^{\pi/2} \int_0^{\infty} F(\theta_0) V f(V) \times \{m_{\text{tot}}(\theta_0, V, H, t) - m_{\text{irr}}(\theta_0, V, H, t)\} dV d\theta_0, \quad (22)$$

where

$$m_{\text{tot}}(\theta_0, V, H, t) = P(\theta_0, V, H, t) \cos \theta_1 + (1 - P(\theta_0, V, H, t)) \cos \theta_3,$$

is the total magnetisation per unit volume and

$$m_{\text{irr}}(\theta_0, V, H, t) = (2P(\theta_0, V, H, t) - 1) \cos \theta_0$$

is the irreversible magnetisation per unit volume of all particles with volume V and orientation θ_0 .

The expressions (21) and (22) allow the calculation of $M_{\text{tot}}(H, t)$ and $M_{\text{irr}}(H, t)$ for any arbitrary $H(t)$ and any initial conditions.

$f(V)dV$ is taken to be log normally distributed in this model:

$$f(V)dV = C_1 \frac{1}{(V/V_m)\sigma} \exp\left(-\frac{1}{2}\left(\frac{\ln(V/V_m)}{\sigma}\right)^2\right) dV, \quad (23)$$

where V_m is the median volume of the particles in the ensemble and σ the dimensionless standard deviation. A log-normal volume distribution was chosen because it well describes the particle size distribution which arises from multiplicative processes such as grinding [25]. The actual form of the distribution is not important for the results presented here however the log-normal distribution does allow

a wide range of volumes to be present in the model compared with other distributions which could be used. The constant C_1 is chosen such that:

$$1 = \int_0^{\infty} V f(V) dV, \quad (24)$$

i.e. that the total volume of the system is 1 unit. $F(\theta_0)d\theta_0$ is taken to be:

$$F(\theta_0)d\theta_0 = C_2 \sin \theta_0 d\theta_0, \quad (25)$$

which represents an isotropic system. The constant C_2 is chosen such that:

$$1 = \int_0^{\pi/2} F(\theta_0) d\theta_0. \quad (26)$$

6. Results

The model above has been used to simulate conventional magnetic viscosity experiments. The material parameters used were those of barium ferrite at 300 K ($K_u = 3.3 \times 10^6$ erg/cm³, $M_s = 380$ G) with an assumed volume distribution parameterised by $V_m = 2.7 \times 10^{-17}$ cm³ (corresponding to a median diameter of 30 nm) and $\sigma = 1$. This set of parameters were used because it is not possible to simultaneously normalise field, magnetisation, volume, time and temperature. It is not suggested that this model reflects the properties of actual barium ferrite. The demagnetisation factor was taken to be zero. The hysteresis loop, showing reversible and irreversible magnetisation, for these material parameters at a field rate of 450 Oe/s is shown in Fig. 4. This field rate simulates that available in commercial superconducting solenoids. The decrease in the magnitude of M_{rev} near coercivity is evidence that η is negative in this system.

For the simulated viscosity experiments the field was started above H_a (17.4 kOe) and ramped at 450 Oe/s to a fixed reverse field between 6 and 14 kOe. The field was held constant at this value for 100 s. After this time either a minor loop to a field 100 Oe below the measurement field (to simulate a χ_{rev} experiment) or a recoil loop to zero field (to simulate a DCD experiment) was performed. The ramp rate simulated was a constant 450 Oe/s.

For each of the measurement fields M_{irr} and M_{tot}

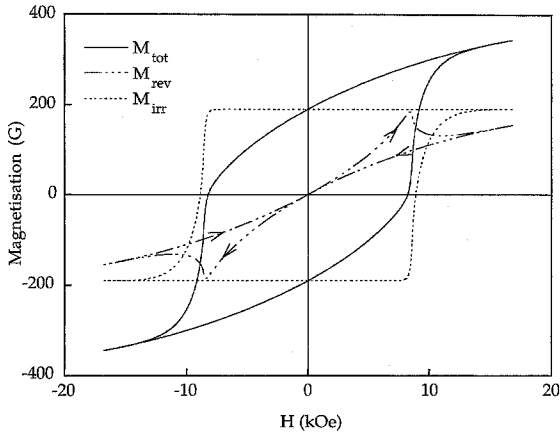


Fig. 4. The simulated hysteresis loop for barium ferrite at 300 K using the model in the text. Both the reversible and irreversible components are shown illustrating the effect of η on the reversible component.

were calculated from Eqs. (21) and (22). Λ was calculated according to Eq. (6) in three ways:

1. From M_{irr} directly, known exactly from Eq. (21) (Λ^{DEF} , i.e. from the definition).
2. From M_{irr} extracted from M_{tot} by the χ_{rev} method (Λ^X).
3. From M_{irr} extracted from M_{tot} by the DCD method (Λ^{DCD}).

Λ^X and Λ^{DCD} simulate results which would be obtained from common experimental practice, without calculation of η , while Λ^{DEF} is the 'true' value of Λ for this material. The results are shown in Figs.

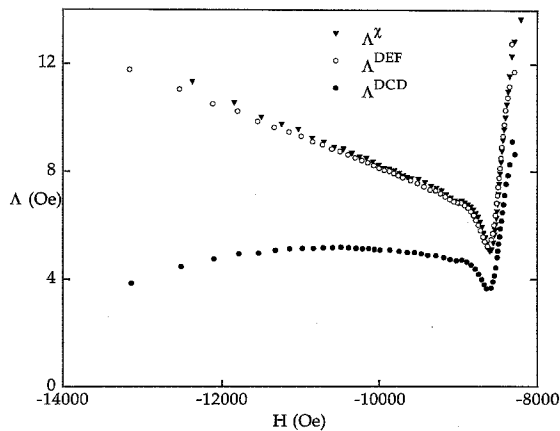


Fig. 5. Λ derived by the χ_{rev} (Λ^X) and DCD (Λ^{DCD}) methods compared with the true Λ (Λ^{DEF}) derived from M_{irr} directly, plotted as a function of field.

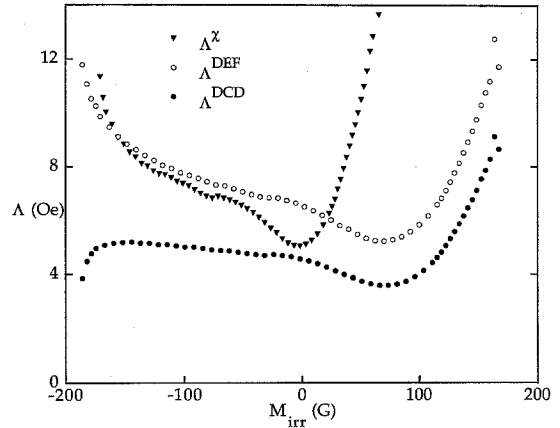


Fig. 6. The results of Fig. 5 plotted as a function of M_{irr} . Note the difference between Λ^X and Λ^{DEF} not evident in Fig. 5.

5 and 6 as functions of both field and M_{irr} . Both Λ^X and Λ^{DCD} are only approximations to Λ^{DEF} . When plotted as a function of field in Fig. 5 Λ^X and Λ^{DEF} are identical, within the errors of the numerical analysis, while Λ^{DCD} lies consistently below the other two curves. When plotted as a function of M_{irr} in Fig. 6 Λ^X has the right magnitude but the wrong M_{irr} values compared with Λ^{DEF} . Λ^{DCD} is consistently lower than Λ^{DEF} but covers the correct M_{irr} range.

To calculate η along the demagnetisation curve the following procedure was used. For each field at which a conventional magnetic viscosity test was simulated, M_{rev} and M_{irr} were calculated, as shown

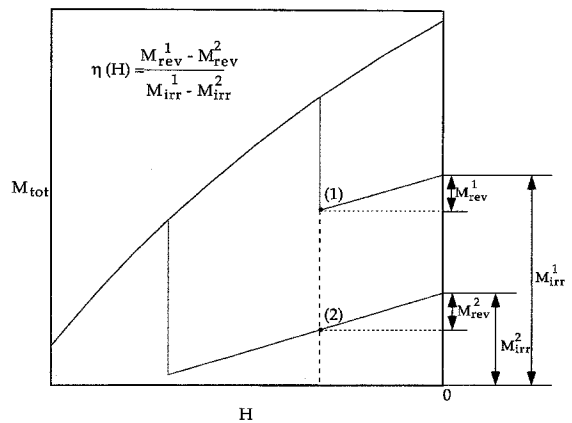


Fig. 7. How η was calculated from the DCD recoil loops (see text) showing the measurement of the reversible and irreversible components.

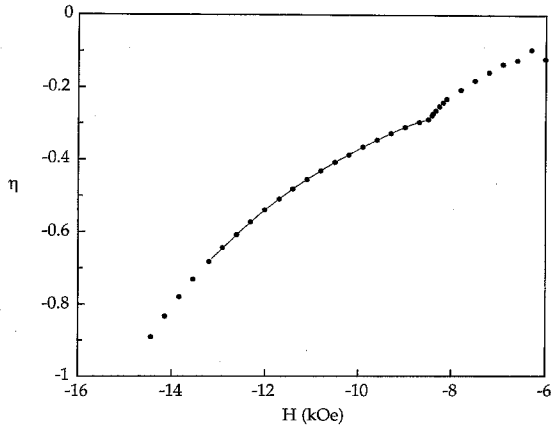


Fig. 8. η calculated for the model as in Fig. 7, plotted as a function of demagnetising field. The solid line indicates the curve fit used to correct Λ^{DCD} in Fig. 9.

in Fig. 7, for the last point (point (1)) in the fixed external field portion of the experiment. To calculate the derivative in Eq. (13), however, another point at the same field as point (1), for which M_{rev} and M_{irr} are known accurately, must be used. This point (point (2)) was chosen as the point on another recoil curve for which $M_{\text{tot}}^1 - M_{\text{tot}}^2$ was a minimum. M_{rev} and M_{irr} were calculated at point (2) in the same manner as for point (1), as shown in Fig. 7. Eq. (13) can then be used to estimate η at this field.

η calculated in this way is shown as a function of

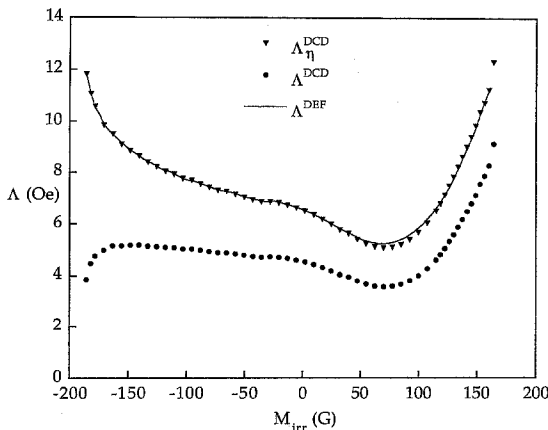


Fig. 9. Λ^{DCD} corrected by η ($\Lambda_{\eta}^{\text{DCD}}$) compared with Λ^{DEF} as a function of irreversible magnetisation. A similar result is obtained when plotted against field. Λ^{DCD} and Λ^{DEF} are the same as those shown in Fig. 6.

field in Fig. 8 and is negative as expected for a Stoner–Wohlfarth reversal mechanism. $\Lambda_{\eta}^{\text{DCD}}$, calculated from Λ^{DCD} corrected by η using Eq. (14) is shown, together with Λ^{DEF} and Λ^{DCD} , in Fig. 9 as a function of M_{irr} . An excellent correspondence between $\Lambda_{\eta}^{\text{DCD}}$ and Λ^{DEF} is obtained. It is important to note that $\Lambda_{\eta}^{\text{DCD}}$ can be obtained for a real experimental material while Λ^{DEF} can only be obtained from information not available experimentally, hence the importance of this result.

7. Discussion

In order to analyse the differences between the three methods of calculating Λ it is necessary to make some simplifying assumptions. These assumptions are the same as those leading to Eq. (8) allowing the use of this equation to explain the data. In using Eq. (8) it is important to differentiate between values of χ_{irr} measured from the definition ($\chi_{\text{irr}}^{\text{DEF}}$) and those measured using the χ_{rev} (χ_{irr}^{χ}) and DCD ($\chi_{\text{irr}}^{\text{DCD}}$) methods.

From Eqs. (8) and (14), Λ^{DEF} may be written as:

$$\Lambda^{\text{DEF}} = \Lambda_{\eta} = \frac{S}{\chi_{\text{irr}}^{\text{DEF}}(1 + \eta)}, \quad (27a)$$

where S is defined according to Eq. (7). For the χ_{rev} method, from Eq. (8):

$$\Lambda^{\chi} = \frac{S}{\chi_{\text{irr}}^{\chi}} = \frac{S}{\chi_{\text{tot}} - \chi_{\text{rev}}}, \quad (27b)$$

where $\chi_{\text{irr}}^{\chi} = \chi_{\text{tot}} - \chi_{\text{rev}}$ from Eq. (9). Similarly Λ^{DCD} is:

$$\Lambda^{\text{DCD}} = \frac{S}{\chi_{\text{irr}}^{\text{DCD}}} = \frac{S}{\chi_{\text{irr}}^{\text{DEF}}}, \quad (27c)$$

where $\chi_{\text{irr}}^{\text{DCD}} = \chi_{\text{irr}}^{\text{DEF}}$ because of the definition of irreversible magnetisation.

Comparison of Eq. (27a) and (27c) explains the relation between Λ^{DEF} and Λ^{DCD} evident in Figs. 5, 6 and 9. The slight mismatch in Fig. 9 between $\Lambda_{\eta}^{\text{DCD}}$ and Λ^{DEF} between 50 and 100 G is because the assumptions used in deriving Eq. (8), hence Eq. (14), are not obeyed in this region of the hysteresis curve.

Why Λ^{χ} and Λ^{DEF} are the same within error when plotted against field but so different when

plotted against M_{irr} becomes evident when the relationship between χ_{irr}^{χ} and $\chi_{\text{irr}}^{\text{DEF}}$ is examined.

Following from Eq. (11) for dM_{rev} :

$$\left. \frac{dM_{\text{irr}}}{dH} \right|_{M_{\text{irr}}} = \left. \frac{dM_{\text{tot}}}{dH} \right|_{M_{\text{irr}}} - \left. \frac{dM_{\text{rev}}}{dH} \right|_{M_{\text{irr}}}$$

$$\chi_{\text{irr}}^{\text{DEF}} = \chi_{\text{tot}} - (\chi_{\text{rev}} + \eta \chi_{\text{irr}}^{\text{DEF}}) \quad (\text{from Eq. (11)})$$

$$\Rightarrow \chi_{\text{irr}}^{\text{DEF}} (1 + \eta) = \chi_{\text{tot}} - \chi_{\text{rev}} = \chi_{\text{irr}}^{\chi}, \quad (28)$$

assuming that η is constant during the period of measurement. Substituting Eq. (28) into Eq. (27b) it can be seen that $\Lambda^{\text{DEF}} = \Lambda^{\chi}$. Λ^{χ} does give the correct magnitude of Λ but does not assign it to the correct M_{irr} value because $\chi_{\text{irr}}^{\text{DEF}} \neq \chi_{\text{irr}}^{\chi}$, thus the alignment of the curves in Fig. 5 but the mismatch in Fig. 6. It should be noted that this is only true when the demagnetising factor is zero. For a non-zero demagnetising factor the situation is more complex and Λ^{χ} is no longer equal to Λ^{DEF} .

Using the DCD method it is possible to extract the correct value of M_{irr} at the end of viscosity because no irreversible processes occur in the model during recoil. Assuming $\eta = 0$, however, results in Λ^{DCD} being consistently lower in value than Λ^{DEF} as expected from Eq. (14).

The preceding results suggest that in a material with a small demagnetisation factor and a large reversible component of magnetisation, the interpretation of Λ data must account for the effect of η . When the demagnetisation factor is included in the analysis Eq. (14) is modified to [19]:

$$\frac{\Lambda}{\Lambda_{\eta}} = \frac{(1 + \eta)(1 + D\chi_{\text{tot}}^i)}{1 + D(\chi_{\text{rev}}^i + \chi_{\text{irr}}^i) + D\eta\chi_{\text{irr}}^i}, \quad (29)$$

which shows that the correction due to η becomes less significant as the demagnetisation factor increases. Eq. (29) also shows that the relation between Λ^{χ} and Λ^{DEF} is no longer simple, i.e. substituting Eq. (28) into Eq. (29), and using $\Lambda = S/\chi_{\text{irr}}$ and $\Lambda_{\eta} = \Lambda^{\text{DEF}}$, no longer yields $\Lambda^{\text{DEF}} = \Lambda^{\chi}$.

The choice of the method for extracting M_{irr} from M_{tot} data will depend on the information being sought and the material under study. The DCD method will give a better estimate of the irreversible magnetisation, but only if the field can be returned to zero without further field or temperature induced irreversible processes occurring. Λ^{DCD} will be more affected if the η correction is not used than Λ^{χ} .

However the η correction cannot be applied to Λ^{χ} because the χ_{rev} method uses an incompatible definition of M_{irr} .

Calculation of Λ for an ensemble of Stoner–Wohlfarth particles has been done before by de Witte et al. [18] and El Hilo et al. [26]. De Witte et al. [18] simulated an isotropic distribution of orientations and a log-normal distribution of volumes exactly as has been done here. The calculation was done differently however. They used an approach where a critical volume, which was a function of orientation, field and time, was assumed to divide the population of particles into two – those which had flipped and those which had not. Thus there was no provision to have, at any particular volume, some particles which had flipped and some which had not. In the terminology used here P was fixed at 0 or 1 and could take no value in between. De Witte et al. [18] used Eq. (8) to calculate Λ instead of the method of Folks and Street [16] used in this work. Also their expression for χ_{irr} (their Eq. (21)) is incorrect and is in fact an expression for χ_{tot} , as an examination of their Eq. (12) shows. In addition the volume distribution used in their analysis is unknown. Despite these differences the form obtained by De Witte et al. [18] for Λ is similar to Λ^{DCD} , with a minimum near $0.5H_a$ and extrapolating to zero near H_a (suggesting their Eq. (21) is a typographical error).

El Hilo et al. [26] took an aligned system with a variety of anisotropy field and volume distributions, and used a critical field, critical volume approach with Eq. (8) to calculate Λ as a function of field. Their results showed that a decreasing Λ with field was a function of the volume distribution while an increasing Λ with field resulted from the anisotropy field distribution. For limited ranges of dispersion for anisotropy field and volume, a minimum in Λ is seen at approximately the coercivity of the model. The form of Λ^{DEF} for the model used here is similar to that calculated by El Hilo et al. [26] in this limited range of dispersion. The initial decrease in Λ^{DEF} with decreasing moment and increasing reverse field is due to the distribution of volume within the material. Small volumes, with large Λ , reverse at low fields and as the field is increased larger and larger volumes reverse, which have smaller values of Λ . This ‘volume controlled’ regime extends in field to

8700 Oe, nearly $H_a/2$, close to coercivity, where the particles oriented at 45° to the field reverse. At this point the behaviour of A^{DEF} changes to an increase with decreasing moment and increasing reverse field. This is because the reversal is now controlled by the orientation of the particles to the field rather than the volume. The distribution of orientations of particles imparts a distribution of critical fields to the model, which although being different in detail to that as-

sumed by El Hilo et al. [26] imparts a similar behaviour to A .

This change in behaviour, from volume to orientation controlled reversal is illustrated in Fig. 10 which shows P calculated by Eq. (18) at different reverse fields between 6000 Oe and 11000 Oe. Each 'frame' shows the state of P as a function of volume and orientation at one particular field. No appreciable difference in the 'frame' can be seen by eye for

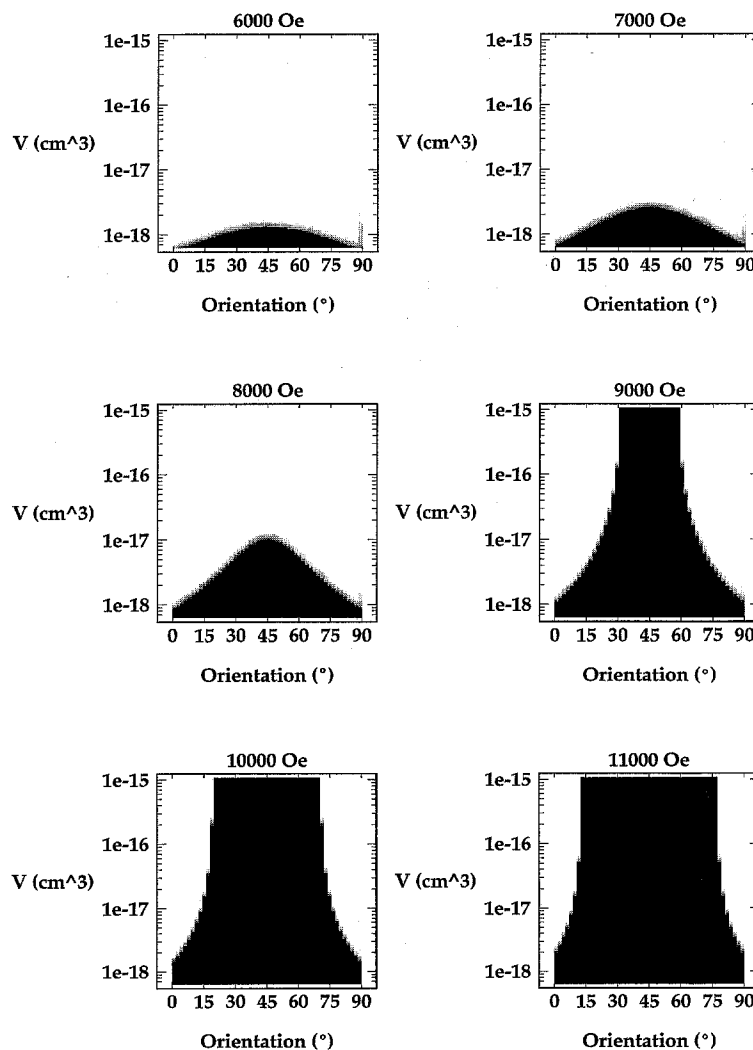


Fig. 10. The state of reversal of simulated Stoner-Wohlfarth barium ferrite particles as a function of volume and orientation for various reversing fields. Black areas represent completely reversed particles, white areas completely unreversed while grey areas represent particles which have only a part of their total number reversed. The time dependence of the reversal, for time scales of the order of 100 s, is negligible on the scale shown.

simulated times from 1 to 100 s at this scale. Black areas represent $P = 0$ or reversed particles, white areas represent $P = 1$ or unreversed particles and grey areas represent some value in between. Below 9000 Oe the increase in the number of reversed particles with field is due mainly to increasing volumes reversing while after 9000 Oe the increase in the number of reversed particles is due to H_k being reached for the two orientations symmetrically disposed about 45° to the field direction.

To calculate Λ at any field directly from a 'frame' in Fig. 10 requires averaging Λ over all of the reversing particles at that field, weighted by both the number of such particles according to volume and orientation and the magnetisation change associated with that particle reversing. This weighting is done automatically when Λ is calculated from the definition, Eq. (6) and therefore has effectively been done when Λ^{DEF} was calculated in Figs. 5 and 6. To illustrate the wide range of particles over which Λ is averaged, the value of Λ when the particle reverses has been calculated as a function of orientation and volume using Eq. (3). This equation is valid when considering individual particle orientations, but is invalid when the whole ensemble is considered because the population of reversing particles changes with field. Each particle was assumed to reverse when the energy barrier was equal to $25kT$, equivalent to a waiting time τ of 100 s in Eq. (16). The

energy barrier was calculated by a numerical approximation due to Pfeiffer [27]:

$$\Delta E = K_u V \left(1 - \frac{H}{H_k} \right)^{0.86 + 1.14 H_k / H_a} \quad (30)$$

This approximation is exact for particles at 0° and 90° to the field and is reasonable for particles between 25° and 65° where, in an isotropic sample, the bulk of the irreversible magnetisation arises. Eq. (30) allows the calculation of $\partial E / \partial H$ at $E = 25kT$ and hence Λ from Eq. (3). Using the value of K_u and H_a for barium ferrite at 300 K, the value of Λ at reversal is shown in Fig. 11 as a function of orientation and volume. It can be seen that at any orientation the value of Λ at reversal decreases with increasing volume and at a fixed volume the value of Λ at reversal increases as the orientation approaches 0° and 90° .

When the reversing particles from a 'frame' in Fig. 10 are examined in Fig. 11 it can be seen that they possess a wide range of values of Λ . The interpretation of Λ^{DEF} in Figs. 5 and 6 in these circumstances is unclear. The value measured at any field is not associated with a single population of reversing particles but is a complex ensemble average over the orientation and volume distributions within the material. It is therefore not possible to work backwards from a measured value of Λ to obtain either the distribution of energy barriers or the

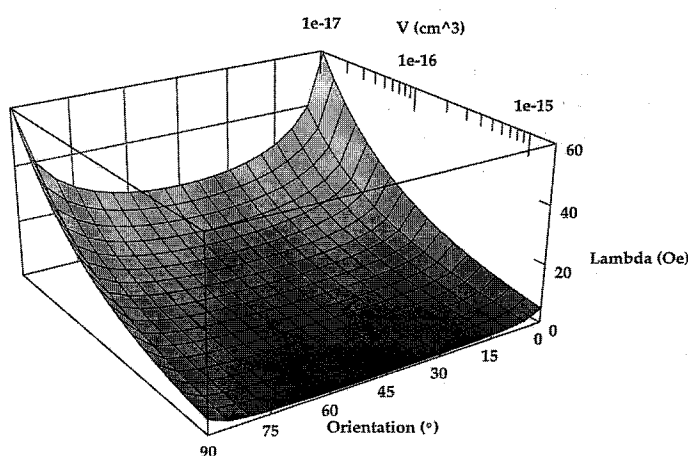


Fig. 11. Λ of a particle when it reverses, calculated as described in the text, as a function of orientation and volume for Stoner–Wohlfarth barium ferrite particles.

mechanism of reversal because Eq. (3) is not valid and the form of Λ obtained may not be unique to any single mechanism of reversal.

In systems with complicated distributions of parameters (e.g. random grain orientation or particle size distribution) the value of measurements of Λ as an investigative tool is unclear. In simpler systems, however, the use of measurements of Λ is justified. In the thin film of TbFeCo studied by Brown et al. [28] for example the experimental evidence points to only a single energy barrier for reversal. In such a system the measured Λ is directly related to the Λ of each particle and Eq. (3) can be used to probe the behaviour of the energy barrier with field.

8. Conclusions

The analysis of magnetic viscosity measurements simulated here on a system of isotropic Stoner–Wohlfarth particles has indicated that the measured form of the viscosity parameter Λ depends on how the reversible component of magnetisation is eliminated from the experimental M_{tot} values. Two common methods for removing the reversible component of magnetisation (here denoted the χ_{rev} and DCD methods) have been examined and each is found to produce results which vary from the true Λ . Correcting results of the DCD method by measuring a parameter η which describes how the reversible magnetisation varies with the state of the system (i.e. the irreversible magnetisation) allows the true Λ for the material to be obtained via an experimentally accessible route. It is only then that interpretation of Λ results can be made reliably. In systems where there is a distribution of orientation and volume, for example, the interpretation of Λ in terms of the mechanism of reversal is obscure.

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