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# The fluctuation field of ferromagnetic materials

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**Abstract.** Starting from the basic constitutive equation that describes the magnetic viscosity of a ferromagnetic material, under the single assumption of a constant external field  $H$ , a connection is shown between the different expressions used to determine experimentally the fluctuation field  $H_f$ . The simplest method uses the relation  $H_f = -\partial H / \partial \ln t|_{M_{irr}}$ . If  $H_f$  is invariant during the viscous decay of the magnetization, the relation  $H_f = -\partial H / \partial \ln(M_{irr})|_{M_{irr}}$  may also be employed. The relaxation curves obtained at different fields, in this case, superimpose onto a single curve  $M_{irr}(t) = F[t/t_{1/2}]$  on renormalizing the time. An alternative treatment that considers explicitly the demagnetizing field is also presented. The theory is then applied to magneto-optic thin films, where two activation mechanisms are involved, assuming the absence of dispersion in the energy barriers, and also to the common case of relaxation by a single activation mechanism in the presence of a dispersion of the energy barriers. In both situations, it is shown that the fluctuation field may vary in strength during magnetic reversal. A method of classification of the hard ferromagnetic materials, through experimental means, is suggested.

## 1. Introduction

Magnetic viscosity, i.e., the time dependence of the magnetization under a constant external field  $H$ , arises from the thermal activation of the magnetization over energy barriers that are present when the field  $H$  is smaller than the coercive field. The time-dependent magnetization is given in general as  $M(t) = M_{irr}(t) + M_{rev}(t)$ , where  $M_{irr}(t)$  is defined as the remanent component obtained by the instantaneous removal of the field  $H$  at time  $t$ . Variations of  $M_{rev}$  with time are relatively small and arise for example from a magnetization-dependent demagnetizing field. Here, we consider only the component  $M_{irr}(t)$ , since magnetic changes by thermal activation are irreversible.

If the activation is over a single energy barrier  $E_B$ , the time dependence should be exponential:

$$M_{irr}(t) = A + B e^{-t/\tau_0} \quad (1)$$

where  $A, B$  are constants and  $\tau_0$  is the relaxation time given by the Arrhenius–Néel law [1]

$$\frac{1}{\tau_0} = f_0 e^{-E_B/kT} \quad (2)$$

where  $f_0$  is a frequency factor of the order of  $10^{-9}$ – $10^{-12}$  s<sup>-1</sup> [1, 2].

For most ferromagnetic materials, however, the observed time dependence is logarithmic over few decades in time:

$$M_{irr}(t) = M_0 + S \ln(t/t_0) \quad (3)$$

where  $M_0 = M_{irr}(t = t_0)$  and  $S$  is the coefficient of magnetic viscosity. It was recognized by Street and Woolley [3] that the logarithmic dependence arises from a dispersion in the energy barriers  $f(E_B) = c$ , where  $c$  is a constant, since the dependence of each barrier  $E_B$  on the respective relaxation time is logarithmic (equation (2)). The slow variation of the coefficient  $S$  with magnetization, observed experimentally, can then be accounted for by considering more realistic, non-uniform, forms for the dispersion  $f(E_B)$ .

An irreversible change of the magnetization may also be induced by a change in the applied field, i.e.  $dM_{irr} = \chi_{irr} dH$ , where  $\chi_{irr}$  is the irreversible susceptibility. Néel [4, 5] therefore suggested that the effect of the thermal fluctuations may entirely be represented as a fictitious field, which he called the fluctuation field. The definition of the fluctuation field is [6]

$$H_f = \frac{S}{\chi_{irr}}. \quad (4)$$

The fluctuation field can be evaluated from macroscopic measurements ( $S, \chi_{irr}$ ), does not depend on the number of magnetic moments that change direction and provides information on the activation mechanism. Wohlfarth [6] therefore argued that it is of fundamental importance. The subject has since attracted much attention and a review is given in reference [7]. Measurements of  $H_f$  can be used to evaluate an activation volume [6]

$$V_A = \frac{kT}{M_S H_f}. \quad (5)$$

Alternative expressions for the measurement of the fluctuation field have recently been suggested, for instance  $H_f = -\partial H / \partial \ln(t)|_{M_{irr}}$  [8, 11] and  $H_f = -\partial H_i / \partial \ln(\dot{M}_{irr})|_{M_{irr}}$  [12, 16], where  $\dot{M}_{irr} = dM_{irr}/dt$  and  $H_i$  is the internal field. The relationship of these expressions with equation (4), however, has not been established. The former has the advantage that it involves simply the measurement of the time required for the magnetization to reach a certain value. The latter was derived by postulating the existence of a magnetic constitutive relation, on the assumption that there is a unique relationship between  $H, M_{irr}$ , and  $dM_{irr}/dt$  [12]. The assumption was justified by considering an Arrhenius rate relation  $dM_{irr}/dt \propto \exp(-E_B(H_i)/kT)$  that involves in effect energy barriers that are all equal in size.

The conditions of validity of the assumption of an absence of a distribution in energy barriers, however, need to be clarified. For instance, the magnetization reversal in amorphous magneto-optic thin films may involve two distinct coercivity mechanisms, domain nucleation and domain growth, with different coercive forces [17]. A distribution in energy barriers is also expected from the spatial dispersion in magnetic properties.

The main objective of this paper is to derive the relationship between the different expressions used to determine the fluctuation field experimentally. Two alternative but equivalent treatments are considered, under the single assumption of a constant external field. The first, in section 2.1, does not consider explicitly the demagnetizing field that may be present in a ferromagnetic material. The second, in section 2.2, considers the demagnetizing field to derive the intrinsic value of the fluctuation field that provides direct information on the activation mechanism. The limitations in using the constitutive equation of Estrin, McCormick and Street [12] and the relation  $H_f = \partial H_i / \partial \ln \dot{M}_{irr}|_{M_{irr}}$  in treatments of magnetic viscosity [13, 16] are identified. The theory applies to any form of time dependence; it does not assume a logarithmic decay. The precise form of the time dependence depends on the choice of the dispersion in energy barriers  $E_B$  for each activation mechanism and is not considered at this stage.

Two examples of the difference between  $\Lambda = \partial H_i / \partial \ln \dot{M}_{irr} |_{M_{irr}}$  and  $H_f$  are then given. The first is the development of a model by Fatuzzo [18] that applies to the magnetic viscosity of some magneto-optic media. It considers two activation mechanisms via nucleation and domain growth without a dispersion in the energy barriers. The second is a treatment of the case that is most common, a single activation mechanism with a dispersion in the energy barriers. It is shown that in both cases  $\Lambda = H_f$  only if the activation volumes of the moments are identical, and the physical implications are discussed.

## 2. Theory

### 2.1. Analytic expressions for the fluctuation field

We consider a ferromagnetic system with a well defined initial state, for instance the saturation remanence. If a constant external field  $H$  is applied there is a spontaneous irreversible response from the moments whose energy barrier has vanished, followed by a slow viscous decay by thermal activation. The field  $H$  is then instantaneously removed after a time interval  $t$ . The remanent magnetization  $M_{irr}$  (that is a measure of any irreversible changes) is clearly uniquely determined by the choice of  $H, t$ , although other factors, such as a time-varying internal field, may also be of influence. An equation of state of the form  $f(M_{irr}, H, t) = 0$  can be constructed that is constitutive since the detailed form is dependent on the ferromagnetic material. A Taylor expansion of the magnetization is then carried out to a power series of small deviations  $\delta H, \delta \ln(t)$ . Although some different variable may have been chosen,  $\ln(t)$  is a natural choice since it is the inverse function associated with the exponential law for the relaxation time (equation (2)). If  $\delta H, \delta \ln(t)$  are sufficiently small, we retain only the linear terms and define a constitutive equation in differential form given by

$$dM_{irr} = S d \ln(t) + \chi_{irr} dH \quad (6)$$

where

$$S(H, t) = \left. \frac{\partial M_{irr}}{\partial \ln(t)} \right|_H \quad (7)$$

is the observed coefficient of magnetic viscosity and

$$\chi_{irr}(H, t) = \left. \frac{\partial M_{irr}}{\partial H} \right|_t \quad (8)$$

is the observed irreversible susceptibility.

Equation (8) is the definition for the irreversible susceptibility appropriate for the study of magnetic viscosity. Experimentally,  $\chi_{irr}$  is obtained using  $\chi_{irr} = (M_{irr}(H + \delta H, t) - M_{irr}(H, t)) / \delta H$ , where  $M_{irr}$  is measured as described above. The common method of measuring  $\chi_{irr}(H, t = 0)$  from the response to a field pulse of very short but undefined duration, for example,  $\chi_{irr} = dM_d(H) / dH$ , where  $M_d(H)$  is the DC demagnetization remanence curve, is restrictive and also difficult to apply when the time dependence of the magnetization is very strong, since the results may not be repeatable.

The general form of the definition of the fluctuation field (equation (4)) is

$$H_f(H, t) = \frac{S(H, t)}{\chi_{irr}(H, t)} \quad (9)$$

showing that  $H_f$  may be time dependent. A time dependence may arise from the gradual variation in the magnetic properties of the moments during the viscous decay.  $H_f$  will be

positive provided that we adopt the convention that  $H > 0$  in the direction of switching of the magnetization. Note also that  $\chi_{irr}$ ,  $S$ ,  $H_f$  may alternatively be regarded as functions of  $(H, M_{irr})$  or  $(M_{irr}, t)$ .

An alternative but equivalent definition of the fluctuation field is obtained directly from the constitutive relation (6):

$$H_f = - \left. \frac{\partial H}{\partial \ln t} \right|_{M_{irr}}. \quad (10)$$

Equation (10) has previously been derived for a system in which the metastable states all have the same value of activation energy [10] and also for magneto-optic thin films in which magnetic reversal is dominated by the expansion of the periphery of a region of reversed magnetization [11]. In the present treatment, however, there are no assumptions, so the fluctuation field can rigorously be determined experimentally by simply measuring the time required for the magnetization to reach a certain value. For instance, it is related to the time dependence of the coercivity

$$H_f(M_{irr} = 0) = - \frac{dH_C}{d \ln(t)} \quad (11)$$

that is of importance in magnetic recording [19, 20]. The time  $t_{1/2}$  for magnetization decay to zero in commercial magnetic tapes is found to fit an empirical relation of the form  $t_{1/2} = T_0 \exp(-\alpha \delta H_C)$ , where  $T_0$  is a constant [19]. The fluctuation field in this case is given by  $H_f = 1/\alpha$ . The relation  $H_f = 1/\alpha$  has previously been demonstrated for a system of perfectly aligned Stoner–Wohlfarth particles with energy barriers all of equal size [21], whereas it is clear from the present discussion that it is of more general validity.

Equation (6) can be expressed in differential form as

$$d \ln(t) = \frac{1}{S} dM_{irr} - \frac{1}{H_f} dH. \quad (12)$$

The right-hand side of equation (12) is an exact differential. The equality of the second derivatives can be expressed as

$$\left. \frac{\partial}{\partial H} \left( \frac{1}{S} \right) \right|_{M_{irr}} = - \left. \frac{\partial H_f^{-1}}{\partial M_{irr}} \right|_H \quad (13)$$

leading to

$$\left. \frac{\partial S}{\partial H} \right|_{M_{irr}} = -\chi_{irr}^2 \left. \frac{\partial H_f}{\partial M_{irr}} \right|_H. \quad (14)$$

Similarly, from the differential equation  $dH = (1/\chi_{irr}) dM_{irr} - H_f d \ln(t)$  we obtain

$$\left. \frac{\partial \chi_{irr}}{\partial H} \right|_{M_{irr}} = - \frac{\chi_{irr}}{H_f} \left. \frac{\partial H_f}{\partial H} \right|_t \quad (15)$$

Using equations (7), (9), (10), the fluctuation field can be expressed as

$$\begin{aligned} H_f = \frac{S}{\chi_{irr}} &= S / \left( - \left. \frac{\partial S}{\partial H} \right|_{M_{irr}} + \left. \frac{\partial S}{\partial H} \right|_{M_{irr}} - \left. \frac{\partial M_{irr}}{\partial \ln(t)} \right|_H \left. \frac{\partial \ln(t)}{\partial H} \right|_{M_{irr}} \right) \\ &= 1 / \left( - \frac{1}{S} \left. \frac{\partial S}{\partial H} \right|_{M_{irr}} + \left. \frac{\partial}{\partial H} (\ln S - \ln t) \right|_{M_{irr}} \right) \\ &= 1 / \left( - \frac{1}{S} \left. \frac{\partial S}{\partial H} \right|_{M_{irr}} + \left. \frac{\partial \ln(\dot{M}_{irr})}{\partial H} \right|_{M_{irr}} \right). \end{aligned} \quad (16)$$

Hence

$$\frac{1}{H_f} = -\frac{1}{S} \frac{\partial S}{\partial H} \Big|_{M_{irr}} + \frac{\partial \ln(\dot{M}_{irr})}{\partial H} \Big|_{M_{irr}}. \quad (17)$$

Equation (17) is valid for any form of time dependence. Next, we consider some cases of special interest.

2.1.1. *The case where  $\partial S/\partial H|_{M_{irr}} = 0$ .* If the coefficient of magnetic viscosity is a single-valued function of the magnetization ( $S(M_{irr})$ ), then

$$\frac{\partial S}{\partial H} \Big|_{M_{irr}} = 0. \quad (18)$$

It is possible to test experimentally whether equation (18) applies. The relaxation curves  $M_{irr}(t)$ , obtained for different values of the applied field  $H$ , should superimpose on a single curve  $M_{irr}(t) = F(t/t_0)$  on renormalizing the time.  $t_0$  is the time required for  $M_{irr}$  to reach a fixed value  $M_0$ . A convenient choice is the time  $t_{1/2}$  for the magnetization to vanish ( $M_0 = 0$ ) and we write

$$M_{irr}(t) = F(t/t_{1/2}). \quad (19)$$

The superposition occurs if the fluctuation field is independent of the magnetization or the time and is a single-valued function of the field (equation (14)):

$$S(M_{irr}) \iff \frac{\partial H_f}{\partial M_{irr}} \Big|_H = \frac{\partial H_f}{\partial t} \Big|_H = 0 \iff H_f(H). \quad (20)$$

Clearly, for a given value of  $H$ , the curves  $S(M_{irr})$ ,  $\chi_{irr}(M_{irr})$  exhibit a maximum for the same value of  $M_{irr}$ . The fluctuation field is also given by (equation (17))

$$H_f = \frac{\partial H}{\partial \ln(\dot{M}_{irr})} \Big|_{M_{irr}}. \quad (21)$$

It is clear that, from equations (10), (21),  $\delta \ln \dot{M}_{irr}|_{M_{irr}} = -\delta \ln(t)|_{M_{irr}}$ . Equation (6) can therefore be expressed as

$$dH = \frac{1}{\chi_{irr}} dM_{irr} - H_f d \ln(t) = \frac{1}{\chi_{irr}} dM_{irr} + H_f d(\ln \dot{M}_{irr}) \quad (22)$$

where

$$\chi_{irr}(H, t) = \frac{\partial M_{irr}}{\partial H} \Big|_{\dot{M}_{irr}} \quad (23)$$

and  $H_f$  is given by equation (21). The constitutive equation (22) is appropriate when the time dependence of the magnetization satisfies the condition (18).

2.1.2. *The case where  $\partial \chi_{irr}/\partial H|_{M_{irr}} = 0$ .* The irreversible susceptibility is a single-valued function of the magnetization if the fluctuation field is independent of  $H$  and  $M_{irr}$ , and can be regarded as a single-valued function of time (equation (15)):

$$\chi_{irr}(M_{irr}) \iff \frac{\partial \chi_{irr}}{\partial H} \Big|_{M_{irr}} = \frac{\partial H_f}{\partial H} \Big|_t = \frac{\partial H_f}{\partial M_{irr}} \Big|_t = 0 \iff H_f(t). \quad (24)$$

For a given value of  $t$ , the curves  $S(H)$ ,  $\chi_{irr}(H)$  then exhibit a maximum value at the same value of the field, in the region of the remanent coercivity.

2.1.3. *The case where  $\partial S/\partial H|_{M_{irr}} = \partial \chi_{irr}/\partial H|_{M_{irr}} = 0$*  If  $S$  and  $\chi_{irr}$  are both single-valued functions of  $M_{irr}$ , the fluctuation field can be regarded as a constant (equations (20), (24)):

$$S(M_{irr}) = H_f \chi_{irr}(M_{irr}). \quad (25)$$

The coercivity  $H_C$  is then logarithmically dependent on time (equation (11)).

There is experimental evidence that for some Tb–Fe–Co thin films [11, 22] the considerations in sections 2.1.1–2.1.3 apply. A more detailed presentation of the evidence is given in section 5.

2.1.4. *The case of a monodispersed system.* Next, we consider thermal relaxation over energy barriers  $E_B$  that are all of equal size, dependent on the field  $H$  but invariant over time. Two implicit assumptions are involved, the absence of a time-dependent demagnetizing field and a negligible probability of activation back to the original state. In section 3, it is shown that  $E_B$  may only be an effective barrier arising from two different activation mechanisms.

The time dependence of the magnetization is given by

$$M_{irr}(t) = F[R(H)t] \quad (26)$$

where  $R \propto e^{-E_B/kT}$  is a rate of activation that remains constant with time. The rate of change of the magnetization is derived as

$$\dot{M}_{irr} = \dot{M}_{irr}(E_B = 0)e^{-E_B(H)/kT}. \quad (27)$$

Equation (26) is a sufficient condition for the superposition of the relaxation curves (equations (18), (19)) and in fact equation (27) implies a unique relationship between  $M_{irr}$ ,  $\dot{M}_{irr}$ ,  $H$  that demonstrates the validity of the constitutive relation (22) [12]. In section 4, however, it will be shown that equation (18) is compatible with a dispersion in barriers, so equation (26) is not necessary for the superposition to be observed.

The fluctuation field obtained from equations (21), (27) is  $H_f = -kT/(\partial E_B/\partial H)|_{M_{irr}}$  and, using equation (20), it may be expressed as

$$H_f = -kT \left/ \frac{\partial E_B}{\partial H} \right|_t = -kT \left/ \frac{\partial E_B}{\partial H} \right|_{M_{irr}}. \quad (28)$$

## 2.2. Consideration of the demagnetization factor

Here we present an alternative treatment that considers explicitly the internal field  $H_i$  in a mean-field approximation

$$H_i = H - DM \quad (29)$$

where  $D$  is a scalar demagnetization factor and  $M = M_{irr} + M_{rev}$ .

If a sample with a well defined initial state is subject to a constant field  $H$  for a time interval  $t$ , the remanent ( $M_{irr}$ ) and the reversible  $M_{rev}$ -component of the magnetization can be evaluated. Using equation (29), it can be shown that the internal field  $H_i$  is uniquely determined by  $M_{irr}$  and  $t$ , and we define therefore a constitutive equation  $g(M_{irr}, H_i, t) = 0$  that in differential form is given by

$$dM_{irr} = \left. \frac{\partial M_{irr}}{\partial \ln(t)} \right|_{H_i} d \ln(t) + \left. \frac{\partial M_{irr}}{\partial H_i} \right|_t dH_i = S_0 d \ln(t) + \chi_{irr}^i dH_i \quad (30)$$

where  $S_0$ ,  $\chi_{irr}^i$  are the intrinsic values of  $S$ ,  $\chi_{irr}$  respectively, i.e., the values that would be observed if there was no demagnetizing field ( $D = 0$ ).

The intrinsic value of the fluctuation field is given by

$$H_f^i(H_i, t) = \frac{S_0(H_i, t)}{\chi_{irr}^i(H_i, t)} = - \left. \frac{\partial H_i}{\partial \ln t} \right|_{M_{irr}}. \quad (31)$$

As in section 2,  $H_f$  will be positive if we adopt the convention that  $H_i > 0$  in the direction of switching of the magnetization, i.e.  $D > 0$  for magnetizing and  $D < 0$  for demagnetizing processes.

We consider next the relationship between the intrinsic and observed values of  $\chi_{irr}$ ,  $S$ ,  $H_f$ . Equation (30) implies that

$$\left. \frac{\partial M_{irr}}{\partial H} \right|_t = \left. \frac{\partial M_{irr}}{\partial H_i} \right|_t \left. \frac{\partial H_i}{\partial H} \right|_{M_{irr}} \quad (32)$$

leading to

$$\chi_{irr}^i = \frac{\chi_{irr}}{1 - D\chi} \quad (33)$$

where  $\chi = \partial M / \partial H|_t$ . Using equations (30), (31) we obtain

$$\left. \frac{\partial \ln t}{\partial H} \right|_{M_{irr}} = \left. \frac{\partial \ln t}{\partial H_i} \right|_{M_{irr}} \left. \frac{\partial H_i}{\partial H} \right|_{M_{irr}} \quad (34)$$

leading to

$$H_f^i = H_f \left( 1 - D \left. \frac{\partial M_{rev}}{\partial H} \right|_{M_{irr}} \right). \quad (35)$$

Using equations (9), (31), (33), (35), we obtain

$$S_0 = S \left[ \frac{1 - D \left. \frac{\partial M_{rev}}{\partial H} \right|_{M_{irr}}}{1 - D\chi} \right]. \quad (36)$$

Since  $D\chi > 0$ , it can be verified that  $\chi_{irr}^i \geq \chi_{irr}$ ,  $H_f^i \leq H_f$ .

The reversible component of the magnetization is dependent on the magnetic configuration ( $M_{irr}$ ), since the contribution of a magnetic moment to  $M_{rev}$  may change following the activation process. Using equation (6) we obtain an expression for the reversible susceptibility that considers explicitly the dependence on the irreversible magnetization:

$$\chi_{rev} = \left. \frac{\partial M_{rev}}{\partial H} \right|_t = \chi_{irr} \left. \frac{\partial M_{rev}}{\partial M_{irr}} \right|_t. \quad (37)$$

The definition for  $\chi_{rev}$  is consistent with equation (8). It is appropriate for the study of magnetic viscosity, but different to  $\partial M_{rev} / \partial H|_{M_{irr}}$  which is normally used in the study of static properties.

The relations (33), (35), (36) are different in their detailed form to the expressions given by Street, Day and Dunlop [23]. In their model, the relationship between the intrinsic and observed susceptibility is of the form  $\chi_{irr}^i = \chi_{irr} / (1 - D\chi_{irr})$ , and similarly for  $\chi_{rev}^i$ ,  $\chi^i$ . This definition, however, results in the condition  $\chi^i = \chi_{rev}^i + \chi_{irr}^i$  not being satisfied.

The intrinsic fluctuation field can be expressed as in equation (17):

$$\frac{1}{H_f^i} = - \left. \frac{1}{S_0} \frac{\partial S_0}{\partial H_i} \right|_{M_{irr}} + \left. \frac{\partial \ln(M_{irr})}{\partial H_i} \right|_{M_{irr}}. \quad (38)$$



2.2.1. *The case where  $\partial S_0/\partial H_i|_{M_{irr}} = 0$ .* Here it is shown that this is the case that was considered in references [12, 13, 15, 16] and a criterion is derived to determine experimentally when it is applicable.

The treatment in section 2.1.1 is here valid on replacing  $H$ ,  $\chi_{irr}$ ,  $S$ ,  $H_f$  by the respective intrinsic values  $H_i$ ,  $\chi_{irr}^i$ ,  $S_0$ ,  $H_f^i$ . In particular, the constitutive relation (22) can be expressed as

$$dH_i = \frac{1}{\chi_{irr}^i} dM_{irr} + H_f^i d(\ln \dot{M}_{irr}) \quad (39)$$

where

$$\chi_{irr}(H_i, t) = \left. \frac{\partial M_{irr}}{\partial H_i} \right|_{\dot{M}_{irr}} \quad (40)$$

$$H_f^i = \left. \frac{\partial H_i}{\partial \ln(\dot{M}_{irr})} \right|_{M_{irr}}. \quad (41)$$

If the reversible magnetization can be ignored or alternatively if  $D = 0$ , then equation (35) reduces to  $H_f = H_f^i$  and using equations (17), (38) we obtain

$$\left. \frac{1}{S_0} \frac{\partial S_0}{\partial H_i} \right|_{M_{irr}} = \left. \frac{1}{S} \frac{\partial S}{\partial H} \right|_{M_{irr}}. \quad (42)$$

The validity of equation (39) used in references [12, 13, 15, 16] can be established experimentally—for instance, by testing whether the following sufficient condition is satisfied:  $\chi_{rev} = 0$  (or  $D = 0$ ) coupled with an invariance of the shape of the relaxation curves with applied field (equation (18)).

The validity of equation (39) does not arise, however, as is implicitly assumed in reference [12], from the absence of a dispersion in the energy barriers. If the energy barriers of all moments are equal in size  $E_B(H_i)$ , but vary in time, the general form of the relaxation curve is

$$M_{irr}(t) = F(R(H_i)t) = F(R(H, t), t). \quad (43)$$

Using  $\tau = R(H_i)t$ , it can be shown that

$$\left. \frac{\partial S}{\partial H} \right|_{M_{irr}} = \frac{dM_{irr}}{d\tau} \tau^2 \left. \frac{\partial}{\partial H} \left[ \frac{1}{R^2} \frac{\partial R}{\partial t} \right] \right|_{M_{irr}}. \quad (44)$$

Since the rate of relaxation is time dependent,  $R(t)$ , it is clear that equation (43) is not a sufficient condition for the validity of equation (22).

### 3. A model of the time dependence of magneto-optic media

In magneto-optic thin films, the reversible component of the magnetization can be neglected to a good approximation ( $M_{irr} = M$ ). For example, the measured value of the saturation remanence in ultrathin Co films is  $M_{RS} > 0.99M_S$  [24]. The intrinsic and observed values of the fluctuation field are then equivalent ( $H_f = H_f^i$ ) and are independent of the sample shape (equation (35)). The main objective, here, to evaluate  $H_f$ , can therefore be achieved using a model by Fatuzzo [18] that does not consider the demagnetizing field ( $D = 0$ ). The values of  $\chi_{irr}$ ,  $S$  obtained from the Fatuzzo model, however, should be corrected for the demagnetizing field (equations (33), (36)), an important experimental consideration. Irreversible magnetic changes against the direction of the applied field are negligible in practice and are ignored by the model.

The Fatuzzo model considers two concurrent activation mechanisms: domain nucleation and domain growth. The model assumes the nucleation of circular domains at a constant rate  $R$  with an initial radius  $r_c$  and subsequent growth at a constant velocity  $v$ . The time dependence of the magnetization is then given by

$$M(t) = M_S(2e^{g(\tau,\kappa)} - 1) \quad (45)$$

where  $\tau = Rt$ ,  $\kappa = v/Rr_c$  and

$$g(\tau, \kappa) = -2\kappa^2 \left[ 1 - (\tau + \kappa^{-1}) + \frac{(\tau + \kappa^{-1})^2}{2} - e^{-\tau}(1 - \kappa^{-1}) - \frac{1 - \tau}{2\kappa^2} \right]. \quad (46)$$

The shape of the curves  $M(t)$  depends only on a single parameter  $\kappa$ . On fitting the experimental curves  $M(t)$  to the analytic function (46) [25], an estimate of  $\kappa$  is obtained that provides a measure of the relative balance between the rates of nucleation and domain growth. The rate of nucleation  $R$  and the rate of expansion  $v$  of the domain radius are related to energy barriers  $E_N, E_W$  according to Arrhenius relations (equation (2)) of the form

$$\begin{aligned} R &= f_0 e^{-E_N/kT} \\ v &= f_0 d e^{-E_W/kT} \end{aligned} \quad (47)$$

where  $d$  is the average separation between pinning sites.  $\kappa$  is then given by

$$\kappa = \frac{d}{r_c} e^{-(E_W - E_N)/kT}. \quad (48)$$

The coefficient of magnetic viscosity and the irreversible susceptibility are obtained using equations (7), (8):

$$S = 2M_S e^g \left. \frac{\partial g}{\partial \ln(t)} \right|_H \quad (49)$$

$$\chi_{irr} = 2M_S e^g \left. \frac{\partial g}{\partial H} \right|_t. \quad (50)$$

The fluctuation field is then obtained from equations (49), (50):

$$H_f = \tau \frac{\partial g}{\partial \tau} / \left( \left. \frac{\partial g}{\partial \tau} \frac{d\tau}{dH} \right|_t + \left. \frac{\partial g}{\partial \kappa} \frac{d\kappa}{dH} \right|_t \right) \quad (51)$$

and using equations (47), (48)

$$H_f = -kT / \left( (1 - \lambda) \left. \frac{\partial E_N}{\partial H} \right|_t + \lambda \left. \frac{\partial E_W}{\partial H} \right|_t \right) = \frac{kT}{M_S} \frac{1}{(1 - \lambda)V_N + \lambda V_W} \quad (52)$$

where

$$\lambda = \kappa \frac{\partial g}{\partial \kappa} / \left( \tau \frac{\partial g}{\partial \tau} \right) \quad (53)$$

and  $V_N, V_W$  are the activation volumes of thermo-activated domain nucleation and domain growth respectively defined from

$$\begin{aligned} V_N(H) &= -\frac{1}{M_S} \frac{dE_N}{dH} \\ V_W(H) &= -\frac{1}{M_S} \frac{dE_W}{dH}. \end{aligned} \quad (54)$$

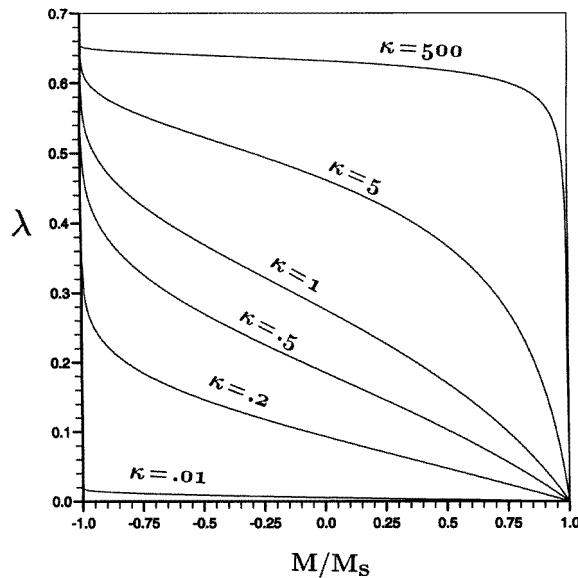
Equation (54) is the definition of the activation volume given by Gaunt [26] that is consistent with the definition of Wohlfarth (equation (5)) in the case of strong domain

wall pinning and no nucleation. The difference between the two definitions is clear from equations (5), (52):

$$V_A = (1 - \lambda)V_N + \lambda V_W. \quad (55)$$

$V_N, V_W$  are related to individual activation events, so  $V_A$  may be regarded as an average value of the activation volume.

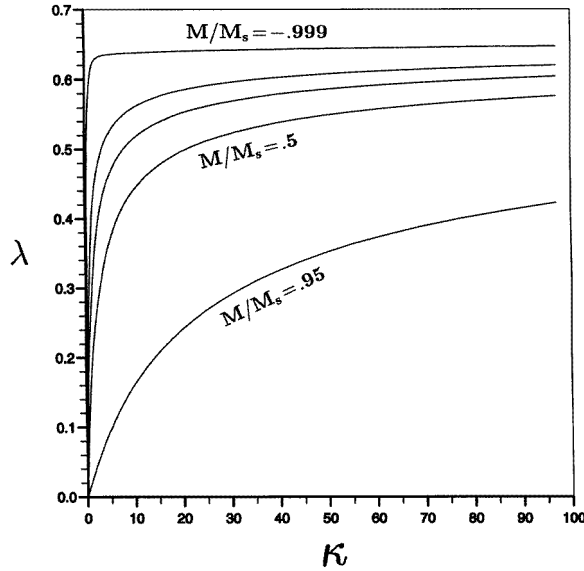
The energy barriers  $E_N, E_W$  are invariant during magnetic reversal in the model of Fatuzzo (since the rates of activation are constant), so the activation volumes defined by equation (54) are independent of the time or the magnetization.



**Figure 1.** The dependence of the factor  $\lambda$  defined by equation (53) on the magnetization  $M$ . Results are presented for different values of the parameter  $\kappa$ .

The time dependence of the fluctuation field is determined by a single parameter  $\lambda(\tau, \kappa)$ . The variation of  $\lambda$  during magnetic reversal is shown in figure 1, and the variation with  $\kappa$  in figure 2. Two basic trends can be identified. First, an increase in the value of  $\lambda$  is observed during magnetic reversal, from a minimum value  $\lambda = 0$  at  $t = 0$  ( $M/M_S = 1$ ) to a maximum value when the reversal is complete. This arises from the relative increase of the contribution of the domain growth to the magnetic reversal, in particular during the later stages of the reversal process (figure 1). Hence, no significant increase of  $\lambda$  is observable, when the reversal is primarily by domain nucleation ( $\kappa \ll 1$ ) (figure 1). Secondly, for the same reason, for any fixed value of  $M$ , an increase of the value of  $\lambda$  with  $\kappa$  is observed in figure 2 to a maximum value  $\lambda = 2/3$  (as will be shown shortly).

The dependence of the  $\chi_{irr}, S, H_f$  on magnetization during the reversal process is shown in figure 3 for  $\kappa = 0.2, V_W/V_N = 1.4$ . The fluctuation field has an initial value at saturation  $H_f(\tau = 0) = kT/(M_S V_N)$  and decreases during the entire reversal process, as a result of the associated increase of  $\lambda$  (figure 1) and the choice made for the ratio  $V_W/V_N > 1$ . The limit  $H_f = kT/M_S V_W$  ( $\lambda = 1$ ) is never reached, probably as the result of the existence of a finite probability of nucleation throughout the reversal process. A reduction by 10% was detected in a Gd-Tb-Fe film [10, 25], and although the activation volumes  $V_N, V_W$  are not



**Figure 2.** The dependence of the factor  $\lambda$  on the parameter  $\kappa$ . Results are presented for different values of the resolved magnetization:  $M/M_S = 0.95, 0.5, 0, -0.5, -0.999$ .

reported, the data for similar Gd–Tb–Fe films [25] indicate that  $V_W > V_N$  as expected from figure 3. The large reduction by 10% occurs since the parameter  $\kappa$  measured for this film has an optimal value  $\kappa = 1$  [25]. This is shown more clearly in figure 1. For  $\kappa \ll 1$  or  $\kappa \gg 1$  the value of  $\lambda$  remains approximately constant for the range of magnetization values that is accessible experimentally and the observed variation of  $H_f$  would be imperceptible in this case within experimental error. In fact, a reduction of  $H_f$  by a mere 3% was reported for a thin Tb–Fe–Co film, where magnetic reversal is dominated by domain wall motion ( $\kappa \gg 1$ ) [11].

Next, we consider three cases of special interest in more detail.

(i)  $\kappa \ll 1$ . If the magnetic reversal occurs primarily by domain nucleation,  $g \approx -\tau$  and  $\lambda \rightarrow 0$  and the fluctuation field is given by

$$H_f = -kT \frac{dE_N}{dH} = \frac{kT}{M_S V_N}. \quad (56)$$

(ii)  $\kappa \gg 1$ . If the magnetic reversal is dominated by domain expansion, it was shown by Fatuzzo [18] that  $Rt \ll 1$ . Equation (46) then reduces to  $g = -\kappa^2 \tau^3 / 3$  and  $\lim_{\kappa \rightarrow \infty} \lambda = 2/3$ . The fluctuation field can be expressed as

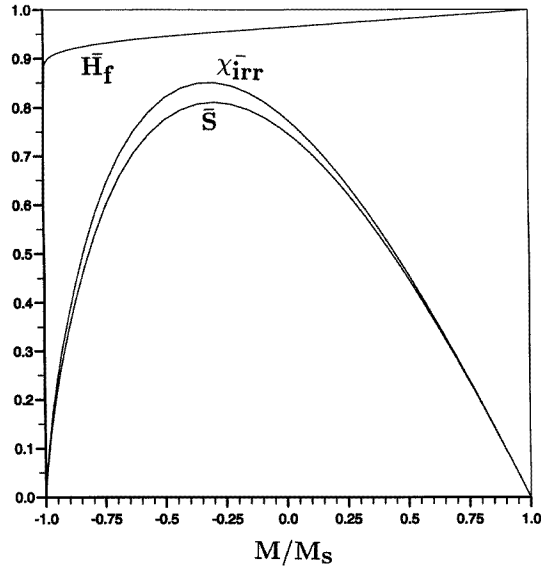
$$H_f = -kT \frac{d\bar{E}_B}{dH} = \frac{kT}{M_S [(V_N + 2V_W)/3]} \quad (57)$$

where

$$\bar{E}_B = \frac{E_N + 2E_W}{3}. \quad (58)$$

The relaxation can be described by a single barrier  $\bar{E}_B$ . If  $R'$  is an effective rate given by

$$R' = f_0 e^{-\bar{E}_B/kT} \quad (59)$$



**Figure 3.** The dependence of the normalized coefficient of magnetic viscosity  $\bar{S} = -S/M_S$ , irreversible susceptibility  $\bar{\chi}_{irr} = -\chi_{irr}/(M_S^2 V_N/kT)$  and fluctuation field  $\bar{H}_f = H_f/(kT/M_S V_N)$  on magnetization. The results were obtained using  $\kappa = 0.2$  and  $V_W/V_N = 1.4$ .

and  $\tau' = R't$ , the time decay can be expressed in a form that does not include the field-dependent parameter  $\kappa$ :

$$M(\tau') = M_S[2e^{-(d^2/3r_c^2)\tau'^3} - 1]. \quad (60)$$

(iii)  $d\kappa/dH = 0$ . This condition is satisfied when the activation volumes of nucleation and domain growth are equal in size, i.e.,  $V_N(H) = V_W(H)$ , for all fields  $H$ . The fluctuation field can then be written directly from equation (52) as

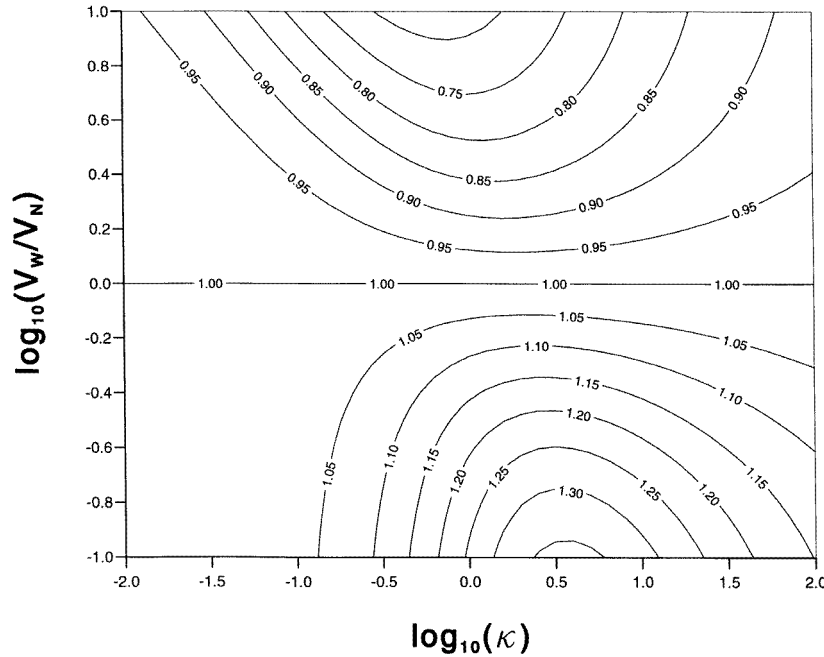
$$H_f = -kT \left/ \frac{dE_N}{dH} \right. = -kT \left/ \frac{dE_W}{dH} \right. = \frac{kT}{M_S V_N}. \quad (61)$$

For the three cases considered above, the time dependence of the irreversible magnetization can be expressed as

$$M(\tau'(H)) = M(R'(H)t) \quad (62)$$

where  $R' = R$  for the cases (i) and (iii), and is given by equation (59) for case (ii). The shape of the relaxation curves is then invariant with applied field (equation (26)). The average activation volume is  $V_A = V_N$ , if  $\kappa \ll 1$  or  $d\kappa/dH = 0$ , and  $V_A = (V_N + 2V_W)/3$ , if  $\kappa \gg 1$  (equation (5)). The fluctuation field is invariant with magnetization (equation (20)); however,  $\chi_{irr}$  may still depend on the applied field.

The constitutive equation (39) is often used [10, 12] to describe the time dependence of magneto-optic thin films. It is therefore interesting to consider to what extent the relation  $\Lambda = \partial H / \partial \ln \dot{M}|_M$  (equation (21)) approximates the exact value of the fluctuation field. A numerical evaluation of  $\Lambda$ ,  $H_f$  was carried out for the system whose time dependence is described by the Fatuzzo function (45). The results for  $H_f$  obtained using equations (9), (10) are indistinguishable. The ratio  $\Lambda/H_f$  is shown by the isoline curves of the contour maps in figures 4 and 5. The results are consistent with the previous discussion, i.e., if



**Figure 4.** A contour map of the ratio  $\Lambda/H_f$  as a function of  $\kappa$  and the ratio of the activation volumes  $V_W/V_N$  when  $M = 0$ .

$\kappa \ll 1$ ,  $\kappa \gg 1$  or  $V_W = V_N$ , the condition  $\partial S/\partial H|_M = 0$  is satisfied and  $\Lambda = H_f$ . In general, however, there may be a difference between  $\Lambda$  and  $H_f$ . For instance, if  $V_W > V_N$ , then  $d\kappa/dH > 0$  (equations (48), (54)), and the Fatuzzo model predicts that  $\partial S/\partial H|_M > 0$  (see figure 4 of reference [25]). According to equation (17),  $\Lambda/H_f < 1$ , as is observed in figure 4. Figure 5 indicates in addition that a significant difference between  $\Lambda$  and  $H_f$  may be observable for any value of the magnetization, within a certain range of values for  $\kappa$ .

Suppose that in addition to the condition (18), the assumption is also made that the average activation volume  $V_A$  is independent of the applied field. The fluctuation field  $H_f$  is then constant in magnitude (equations (5), (24), (25)) and  $\chi_{irr}$  is a single-valued function of the magnetization. These considerations appear to apply for some Tb–Fe–Co thin films [22], where both a weak dependence of  $H_f$  on  $H$ ,  $M_{irr}$  and the superposition of all of the  $\chi_{irr}(H, t)$ ,  $S(H, t)$  curves when plotted as a function of magnetization are observed. Equations (11), (25) also predict in this case a logarithmic time dependence of the coercivity that is in agreement with experimental observation [11, 27].

A more detailed account of the coercivity is obtained from equation (45):

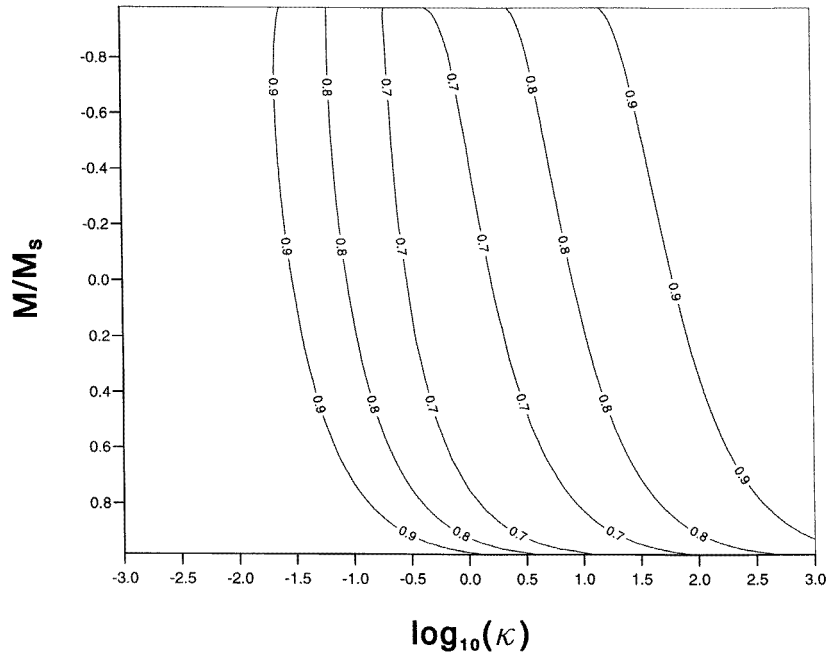
$$g(\tau, \kappa) + \ln 2 = 0. \tag{63}$$

If the activation volumes are independent of the applied field, the energy barriers  $E_N$ ,  $E_W$  are given by

$$\begin{aligned} E_N &= E_N^0 - M_S V_N H \\ E_W &= E_W^0 - M_S V_W H. \end{aligned} \tag{64}$$

The rate of nucleation  $R$  can be expressed as

$$R = R_0 e^{M_S V_N H/kT} \tag{65}$$



**Figure 5.** A contour map of the ratio  $\Delta/H_f$  as a function of  $\kappa$  and the resolved magnetization  $M/M_S$  using  $V_W/V_N = 10$ .

where

$$R_0 = f_0 e^{-E_N^0/kT}. \quad (66)$$

The solution  $\tau_0(\kappa)$  of equation (63) can be expressed as

$$\tau_0(\kappa) = R(H_C)t_{1/2} = R_0 e^{M_S V_N H_C/kT} t_{1/2}. \quad (67)$$

One may also define the temperature- and time-independent intrinsic coercive force of nucleation,  $H_N$ , as the field that makes the energy barrier of nucleation vanish:

$$H_N = \frac{E_N^0}{M_S V_N} = \frac{kT}{M_S V_N} \ln \left[ \frac{f_0}{R_0} \right]. \quad (68)$$

Using equations (67), (68), we obtain an expression for the time dependence of the coercivity:

$$H_C(t) = H_N - \frac{kT}{M_S V_N} \ln \left[ \frac{f_0 t_{1/2}}{\tau_0(\kappa)} \right]. \quad (69)$$

The dependence of  $\tau_0$  on  $\kappa$  is shown in figure 6. If condition (18) is not satisfied (i.e.,  $d\kappa/dH \neq 0$  and  $\kappa$  in neither too small nor too large) the dependence of  $\kappa$  on the field  $H$ , as shown in figure 6, becomes significant and equation (69) indicates that it results in a strictly non-linear logarithmic time dependence for the coercivity. Conversely, if (18) is satisfied, the gradient  $dH_C/d \ln(t)$  provides information through equations (5), (11) on the effective activation volume  $V_A$ . In figure 7, the logarithmic time dependence of the coercivity  $H_C$  is shown for different values chosen for  $\kappa$ , assuming that  $d\kappa/dH = 0$ . An increase in the relative rate of domain growth results in lower coercivity for a fixed time of measurement

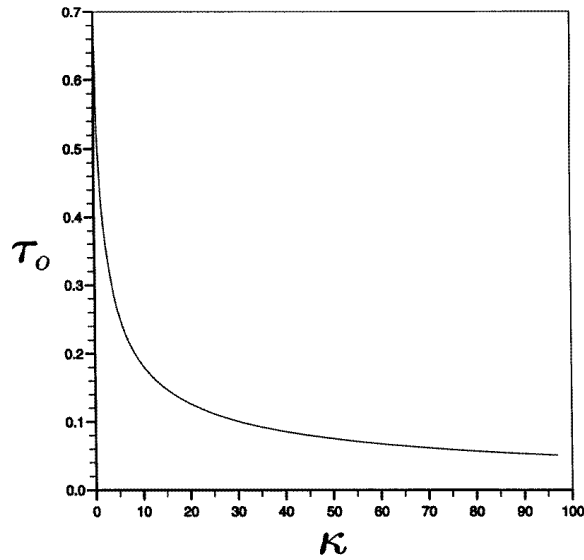


Figure 6. The resolved time  $\tau_0$  at the coercive point as a function of  $\kappa$ .

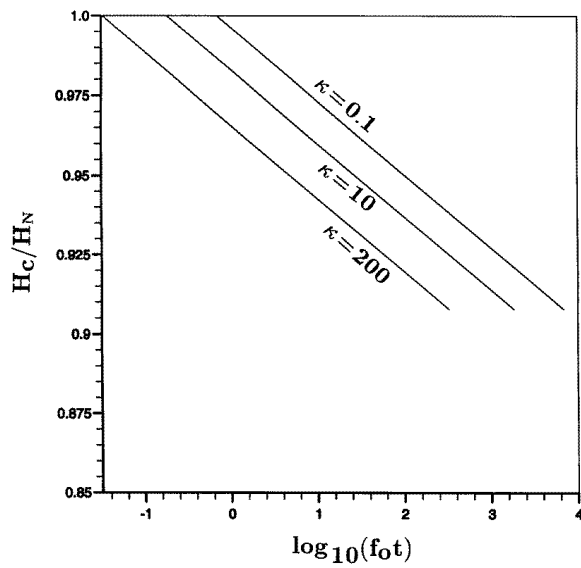
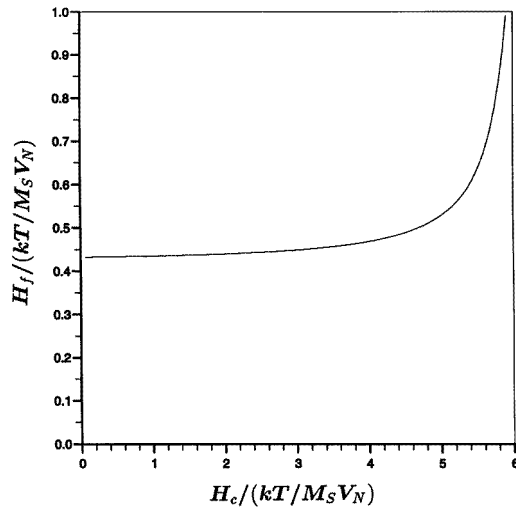


Figure 7. The logarithmic time dependence of the resolved coercivity  $H_C/H_N$  for different values of  $\kappa$ , using  $kT/E_N^0 = 0.01$ .

as is physically reasonable. In previous work [25], the logarithmic time dependence of the coercivity was demonstrated for  $\kappa \ll 1$  and  $\kappa \gg 1$  only.

An increase of the fluctuation field with coercivity is predicted under some special conditions, for instance, on allowing variations in the value of the factor  $(d/r_c) \exp(-E_W^0/kT)$  and considering  $V_W/V_N > 1$ . The variation of the coercivity  $H_C$  in this case arises from the dependence on  $\tau_0(\kappa)$ . A typical  $H_f$  versus  $H_C$  plot is shown in





**Figure 8.** The dependence of the normalized values of  $H_C$  versus  $H_f$ . The curve was obtained using  $f_0 t_{1/2} = 10^9$ ,  $E_N^0/kT = 27$ ,  $V_W/V_N = 3$  and by allowing variations in  $(d/r_c) \exp(-E_W^0/kT)$ .

figure 8. The observed increase has no connection with the Barbier relation  $\ln H_f \propto H_C$  that is observed for a wide range of materials of different magnetic hardness [6, 28], including magneto-optic media [10, 25]. The sharp increase in the coercivity of Gd–Tb–Fe films [25] is not accompanied with a substantial variation in  $\kappa$ . The Barbier relation appears to involve rather a relation between the intrinsic activation energies  $E_N^0$ ,  $E_W^0$  and the fluctuation field, and requires therefore a study of the detailed activation mechanisms.

The Fatuzzo model predicts an exponential decay of the magnetization when the nucleation process is dominant; however, the decay observed experimentally is logarithmic [24]. The distribution of energy barriers should then be considered and a detailed study follows in the next section.

#### 4. Magnetic viscosity arising from a single activation mechanism

The general treatment of a dispersion in the energy barriers is presented in section 4.1. The case of the barriers being coupled to a single physical parameter is considered in section 4.2. Finally in section 4.3., we consider the physical implications when the shape of a sample is allowed to vary.

##### 4.1. Relaxation over a dispersion of the energy barriers

As in the previous section, we ignore the demagnetizing field ( $D = 0$ ) and any magnetic changes that are either reversible or against the direction of the applied field, to simplify the presentation. Since the objective is simply to establish the relation between  $\Lambda = \partial H / \partial \ln \dot{M}|_M$  and  $H_f$ , the precise form of the time dependence is not important and these assumptions should not affect our conclusions.

The time dependence of the magnetization of a system of perfectly aligned moments is

$$M(H, t) = M_S \left( 2 \int_0^\infty e^{-t/\tau_0} f(\tau_0) d\tau_0 - 1 \right) \quad (70)$$

where  $f(\tau_0)$  is the dispersion in relaxation times. Since  $\tau_0$  is exponentially dependent on the energy barrier  $E_B$  (equation (2)), it is possible to define a time-dependent activation energy  $E'_B(t) = kT \ln(f_0 t)$  such that  $e^{-t/\tau_0} \approx 0$  if  $E_B < E'_B$  and  $e^{-t/\tau_0} \approx 1$  if  $E_B > E'_B$ . The time dependence of the magnetization can therefore be expressed as

$$M(H, t) = M_S \left[ 2 \int_{E'_B(t)}^\infty f(E_B, H) dE_B - 1 \right] \quad (71)$$

where  $f(E_B, H)$  is the normalized distribution of energy barriers. The coefficients of magnetic viscosity and irreversible susceptibility are obtained using equations (7) and (8) respectively:

$$S(H, t) = -2M_S f(E'_B, H) \frac{dE'_B}{d \ln(t)} = -2M_S kT f(E'_B(t), H) \quad (72)$$

$$\chi_{irr}(H, t) = 2M_S f(E'_B, H) \frac{dE_B}{dH} \Big|_{E_B=E'_B(t)}. \quad (73)$$

The fluctuation field is therefore given by

$$H_f(H, t) = -kT \Big/ \frac{dE_B}{dH} \Big|_{E_B=E'_B(t)}. \quad (74)$$

Equation (74) is similar to the expression derived by Gaunt [29]; however, it considers also the time dependence of  $H_f$ . We are interested in evaluating the term  $\partial S / \partial H|_M$  that is given by

$$\frac{\partial S}{\partial H} \Big|_M = -2M_S kT \frac{\partial f(E'_B, H)}{\partial H} \Big|_M. \quad (75)$$

Substitution of equations (72), (73) in equation (6) gives a relationship between  $\delta E'_B$  and  $\delta H$  when the magnetization is held constant:

$$kT \frac{\partial \ln(t)}{\partial H} \Big|_M = \frac{\partial E'_B}{\partial H} \Big|_M = \frac{dE_B}{dH} \Big|_{E_B=E'_B(t)}. \quad (76)$$

The field dependence of the energy barriers is determined by the individual activation volumes of the moments, that are given from equations (5), (74) as

$$V_A = - \frac{1}{M_S} \frac{dE_B}{dH} \Big|_{E_B=E'_B(t)}. \quad (77)$$

An implicit assumption in our treatment is the continuous variation of the energy barriers  $E_B$  with applied field. It is therefore possible to apply the continuity relation

$$\frac{\partial f}{\partial H} + \frac{\partial}{\partial E_B} \left( f \frac{dE_B}{dH} \right) = 0. \quad (78)$$

A Taylor series expansion of  $f$ , truncated to first order, gives

$$f(E_B + \delta E_B, H + \delta H) - f(E_B, H) \approx \frac{\partial f}{\partial H} \delta H + \frac{\partial f}{\partial E_B} \delta E_B \quad (79)$$

valid for arbitrary but small variations  $\delta E_B$ ,  $\delta H$ . Equation (78) implies correlated variations, such that  $\delta E_B/\delta H = dE_B/dH$ . Using equations (78), (79) we obtain the following expression valid to first order:

$$f(E_B + \delta E_B, H + \delta H) - f(E_B, H) \approx -f \frac{\partial}{\partial E_B} \left( \frac{dE_B}{dH} \right) \Big|_H \delta H. \quad (80)$$

At constant  $M$ , equation (76) implies a similar correlation in  $\delta E'_B$  and  $\delta H$ . Application of equation (80) in equation (75) yields

$$\frac{\partial S}{\partial H} \Big|_M = 2M_S kT f(E'_B(t), H) \frac{\partial}{\partial E_B} \left( \frac{dE_B}{dH} \Big|_{E_B=E'_B} \right) \Big|_H. \quad (81)$$

Using equations (72), (77) we obtain

$$\frac{1}{S} \frac{\partial S}{\partial H} \Big|_M = - \frac{\partial}{\partial E_B} \left( \frac{dE_B}{dH} \Big|_{E_B=E'_B} \right) \Big|_H = M_S \frac{\partial V_A}{\partial E_B} \Big|_H. \quad (82)$$

The relation between  $\Lambda = \partial H / \partial \ln \dot{M} \Big|_M$  and  $H_f$  is given by equations (17), (82). If the activation volume  $V_A$  of all moments is identical, i.e.,  $F(V_A) = \delta(V_A - V_A^0)$ , then the term  $(1/S) \partial S / \partial H \Big|_M = 0$  and  $\Lambda = H_f$ . The relaxation curves at different fields then fit on a single curve  $M(t) = F[\ln(t/t_{1/2})]$  (section 2.1.1). This relationship was observed in Co films of thickness of the order of few atomic monolayers [24] and results from the activation volume being decoupled from the physical mechanism that gives rise to the dispersion in energy barriers.

#### 4.2. Dependence of the energy barriers on a single parameter

Suppose that the energy barriers are coupled to a single physical property, for example, the volume  $V$  of each magnetic moment, and that the variation  $E_B(V)$  is monotonic on average. The energy  $E'_B$  would then be associated with some critical volume  $V_C(H, t)$ . The normalized distribution function  $g(V)$  is obtained from

$$g(V) = f(E_B, H) \frac{\partial E_B}{\partial V} \Big|_H. \quad (83)$$

The coefficient of magnetic viscosity may then be expressed as

$$S = -2M_S kT g(V_C) \left[ \frac{\partial E'_B}{\partial V_C} \Big|_H \right]^{-1}. \quad (84)$$

The term  $\partial S / \partial H \Big|_M$  can be expressed as

$$\frac{\partial S}{\partial H} \Big|_M = \frac{\partial S}{\partial H} \Big|_{V_C} = -2M_S kT g(V_C) \frac{\partial}{\partial H} \left[ \frac{\partial E'_B}{\partial V_C} \Big|_H \right]^{-1} \Big|_{V_C}. \quad (85)$$

Hence, we obtain

$$\frac{1}{S} \frac{\partial S}{\partial H} \Big|_M = \frac{\partial}{\partial H} \left[ \ln \left( \frac{\partial E'_B}{\partial V_C} \Big|_H \right)^{-1} \right] \Big|_{V_C} = M_S \frac{\partial V_A}{\partial E_B} \Big|_H \quad (86)$$

in agreement with equation (82). If the dependence  $E_B(V, H)$  is known explicitly, then the relation between  $\Lambda$ ,  $H_f$  can be evaluated from equation (86). For exchange-decoupled domains that reverse coherently [30],

$$E'_B = K_U V_C \left( 1 - \frac{H}{H_K} \right)^2 \quad (87)$$

where  $H_K = 2K_U/M_S$ , and substitution in equation (86) yields

$$\left. \frac{1}{S} \frac{\partial S}{\partial H} \right|_M = \frac{2}{H_K} \frac{1}{1 - H/H_K} \quad (88)$$

so  $\Lambda \neq H_f$ .

#### 4.3. Variation of the sample shape

It is interesting to consider briefly the effects of a variation of the shape of the sample, by allowing the demagnetization factor  $D$  to increase. To simplify the discussion, reversible magnetization changes are ignored.

The observed values of  $\chi_{irr}$ ,  $S$  are given by  $\chi_{irr} = \chi_{irr}^i / (1 + D\chi_{irr}^i)$  and  $S = S_0(1 - D\chi_{irr}^i)$  (section 2.2). Since  $D\chi_{irr}^i > 0$ , a reduction in the observed values  $\chi_{irr}$ ,  $S$  arises from an increase of the magnitude of the demagnetizing field  $DM$ , that is closely related to the observed shearing of the hysteresis loops. The shearing occurs since the irreversible response of the magnetization ( $S_0$ ,  $\chi_{irr}^i$ ) is dependent on the dispersion in the energy barriers  $f(E_B)$  and the size of each barrier is modified by the internal field  $H_i$ . A larger field variation  $\Delta H$  is required for a net change  $\Delta H_i$ , when the demagnetizing field is strong. The fluctuation field, however, is independent of the dispersion  $f(E_B)$  (equation (74)) and it is not surprising that no demagnetizing field correction is required ( $H_f = H_f^i$ ).

The precise form of the dependence of the energy barriers on the internal field  $E_B(H_i = H - DM)$  is determined by the activation mechanism, and information can be provided by measurement of  $H_f$ . A linear dependence of the energy barriers on the demagnetization factor arises, for example, when  $H_f$  is invariant with the applied field (section 2.1.2).

## 5. Discussion

The fluctuation field of a magnetic material, under constant external-field conditions, can be determined experimentally from relaxation curves  $M_{irr}(t)$  only, using  $H_f = S/\chi_{irr}$  or the waiting time method:  $H_f = -\partial H / \partial \ln(t)|_{M_{irr}}$ . If the shape of the curves is independent of the choice of applied field, i.e. they fit a single curve ( $M_{irr}(t) = F(t/t_{1/2})$ ) on renormalizing the time, then  $\partial S / \partial H|_{M_{irr}} = 0$  and the fluctuation field is given by an alternative expression:  $H_f = \partial H / \partial \ln M_{irr}|_{M_{irr}}$ . In practice, however, the usefulness of this expression is restricted in the region of remanent coercivity [14], since any variation  $\Delta \ln M_{irr}$  must be large to be measured without substantial experimental error. The waiting time method is therefore more rigorous. It is also simpler than the use of  $H_f = S/\chi_{irr}$ . For instance, the intrinsic value of the fluctuation field can be deduced directly for  $\chi_{rev} = 0$  using equation (35), and does not require a correction of  $S$ ,  $\chi_{irr}$  for the demagnetizing field.

The present theory is suggestive of a connection between two common experimental observations: the superposition of the relaxation curves ( $M_{irr}(t) = F(t/t_{1/2})$ ) and the invariance of  $H_f$  with magnetization. The former has been observed for magneto-optic media such as Gd-Fe, Gd-Tb-Fe and Tb(Co)-based alloys [25], Tb-Fe-Co [11] and ultrathin Co films [24]. A slow variation of  $H_f$  with magnetization has been reported for magneto-optic media such as Gd-Tb-Fe [10] and Tb-Fe-Co [11], CrO<sub>2</sub> recording particles [16], sintered Pr<sub>2</sub>Fe<sub>14</sub>B [16] and isotropic Nd-Fe-B permanent magnets [12]. At present strong evidence for the existence of a connection between the two experimental observations exists only in the case of Tb-Fe-Co [11].

It is interesting to consider the physical reason for the slow variation of the fluctuation field with magnetization. The fluctuation field is then dependent on some average  $V_A$  over the activation volumes of the moments switching during a certain time interval, and each of these is determined by the individual mechanism of reversal (equations (5), (55), (74)). Givord *et al* [31] have shown that  $V_A = \delta^3$  in NdFeB sintered magnets, where  $\delta$  is the average domain wall width. The activation volume of elongated particles such as CrO<sub>2</sub> is much smaller than the actual particle size [32] and may be dependent on the particle cross-sectional area and domain wall energy [33]. These two examples indicate the possibility that the activation volume is determined by the intrinsic material properties and may only be weakly correlated to the grain size.

The time dependence curves cannot be made to superimpose for all magnetic materials, for instance Tb/Fe [34] and Co/Pt [27] multilayers. In this case the constitutive relation (6) rather than (22) should be used.

By development of the theory of Fatuzzo [18], the fluctuation field for magneto-optic thin films was evaluated. The expression derived by Gaunt (equation (74)) is no longer applicable, since it involves the implicit assumption of a single activation mechanism. The fluctuation field was shown to exhibit a variation in strength during magnetic reversal if there is a difference in the activation volumes of nucleation and wall motion that arises from the gradual increase of the relative contribution of domain growth to the irreversible magnetic changes during the reversal process. A similar but rather small variation was detected in a Gd–Tb–Fe thin film [10]; however, to test the theory the measurement of the activation volumes of nucleation and wall motion is also required. If the activation volume of a magneto-optic thin film is determined experimentally using the definition adopted by Wohlfarth [6]:  $V_A = kT/(M_S H_f)$ , this value represents some average over the activation volumes of nucleation and wall motion. The relative balance is described by the factor  $\lambda$  in the model (equation (53)). The value of  $\lambda$  appropriate for magnetic reversal occurring primarily by domain growth ( $\lambda = 2/3$ ) was experimentally verified by Labrune *et al* [25]. The present treatment allows the evaluation of that factor in other cases as well.

The condition  $\partial S/\partial H|_{M_{irr}} = 0$  is satisfied in the Fatuzzo model if the magnetic reversal is primarily by domain wall motion or when the activation volumes of thermo-activated nucleation and wall motion are identical. If the magnetic reversal is by continuous nucleation, the Fatuzzo model is not appropriate and a different approach is required that considers the dispersion in the energy barriers. The condition  $\partial S/\partial H|_{M_{irr}} = 0$  then is satisfied only if there is no associated dispersion in the activation volumes. The relaxation curves then should fit a relation  $M(t) = F[\ln(t/t_{1/2})]$  in agreement with experimental data on ultrathin Co films [24]. This relation does not represent, however, a universal law, since it is not satisfied, for example, by fine-particle systems that exhibit coherent magnetic reversal.

In summary, it appears that the hard ferromagnetic materials can be classified into two categories.

(a) Magnetic materials for which  $\partial S/\partial H|_{M_{irr}} = 0$  that exhibit an absence of a dispersion in activation volumes (e.g., some Tb–Fe–Co thin films and possibly CrO<sub>2</sub> recording particles, Nd–Fe–B permanent magnets, and multi-domain particles).

(b) Magnetic materials such as those exhibiting coherent magnetic reversal (single-domain particles) and multilayers for magneto-optic recording for which the above considerations do not apply.

The available experimental evidence is rather limited at present and more data are needed to test the predictions of the theory.

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