Predicted Time Dependence of the Switching Field for Magnetic Materials

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A scaling relationship between switching fields, of which remanent coercivity is a prominent example, and measurement time is derived. The energy barrier to thermal fluctuations is found to exhibit a $\frac{3}{2}$-power dependence on the difference between the applied field and the nonthermally assisted switching field. This $\frac{3}{2}$-power dependence contrasts with the 2-power dependence which has been widely assumed in the literature. Implications for magnetic viscosity and the orientational dependence of the time-dependent switching field in certain small, isolated particles are also discussed.

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Hysteresis is one of the primary manifestations of ferromagnetism. The change in magnetization owing to an applied field, as plotted in a hysteresis loop, consists of reversible and irreversible components. The irreversible changes occur as the magnetization in some part of the sample crosses an energy barrier. This crossing can be greatly aided by thermal fluctuations. The number of such thermally aided opportunities depends on the attempt frequency and the allowed time. Thus, the field at which magnetization switches (switching field) depends on the measurement time. This leads to phenomena such as magnetic viscosity, where the magnetization continues to change even though the applied field is constant, and time-dependent coercivities. Elevated temperature magnifies these effects, as in thermoremanent magnetization, where sufficient time is allowed for thermal equilibrium and, thus, enhanced response to any applied magnetic field.

Examination of thermally induced phenomena, particularly thermoremanent magnetization in rocks, led Néel, in 1949, to study[1] a simple model consisting of a single particle of volume V. The particle possessed an uniaxial anisotropy constant K with the easy axis parallel to an applied field of magnitude H. The particle magnetization was assumed to be a coherently rotating vector of magnitude M at an angle θ relative to $-\mathbf{H}$. It is easy to show that, for fields below the switching field $H_0=2K/M$, there are two energy minima at magnetization angles $\theta=0,\pi$ with energies $E=\pm HMV$ and two maxima at $\theta=\cos^{-1}(HM/2K)$ with energy $E=KV[1+(HM/2K)^2]$. Thus, the energy barrier which prevents switching from the $E=HMV$ local minima has magnitude $KV(1-H/H_0)^2$; in other words, the energy barrier shows a $(\Delta H)^2$ dependence.

This simple model, although never justified in other than the very special case of a collinear applied field and anisotropy vector, has become the dominant model for thermal effects. Aside from its original and continuing[2] use in the study of rock magnetism, it has been applied to spin glasses to explain the magnetic field dependence of the susceptibility peak.[3] Predictions for anhysteretic remanent magnetization[4] and the temperature dependence of the maximum magnetic viscosity[5] have been made on the basis of this model. The properties of ferrofluids have also been discussed.[6] Direct practical application includes work[7] on phenomena in magnetic recording such as print-through, in which stray fields produce media noise, and record coercivities. Interestingly, the latter effect was also examined by using a $(\Delta H)^{3/2}$ dependence of the energy barrier, such having been empirically found to match a special case of cubic anisotropy very well. It is worthwhile to note that Néel, in a paper[8] discussing domain-wall motion under weak applied fields, also found a $(\Delta H)^{3/2}$ dependence for a simple one-variable model, but discarded it in an effort to include fluctuations in other system variables.

In this paper, it will be shown that the Néel[1] results, in the region where the effect of thermal fluctuations can be viewed perturbatively, present an atypical special case and that a much more general result can be derived which exhibits to leading order a $(\Delta H)^{3/2}$ dependence of the energy barrier. In the case of a Lorentzian switching-field distribution, specific predictions for the temperature dependence of the magnetic viscosity will be made. An assumption of uniaxial anisotropy, a coherently rotating magnetization, and neglect of interactions will allow the coefficient of the $(\Delta H)^{3/2}$ term to be explicitly stated with a resultant prediction of the orientational dependence. Implications and the expected range of the proposed scaling relationship will be discussed.

We are interested in the case where the height of the energy barrier is much greater than k_BT. This corresponds to the physically and technologically interesting case of the measurement frequency being much less than the attempt frequency, and is the situation considered by Néel.[1] It implies that, to a high degree of accuracy, a Maxwell-Boltzmann distribution occurs in the neighborhood of the switching point.[9] This means[1,10] that the probability of switching is given by

$$P=A\tau e^{-\Delta E/k_BT}. \tag{1}$$

Here τ is the measurement time, ΔE is the energy barrier, and A is the attempt frequency which is approximately 10^9 Hz. Actually, A depends on the applied field; however, in the conditions considered here ($\Delta E \gg k_B T$), this contribution to P is extremely weak in comparison to the ΔE contribution and will be neglected.

We begin the derivation by considering the sample, consisting of numerous small areas of magnetization all interacting with each other both magnetostatically and through quantum-mechanical exchange, to be subject to the applied field H. The high-frequency, i.e., without thermal fluctuations, switching field which is closest to the applied field is labeled H_0. The energy is Taylor expanded about the state at $H=H_0$ in the applied field $\Delta H = H_0 - H$ and the numerous variables $\Delta\theta_i = \theta - \theta_0$ representing the magnetization degrees of freedom. The expansion is carried to third order. The first-order terms, except for the ΔH dependence, are all equal to zero because the system is in a local minimum. The matrix of second derivatives is diagonalized to obtain the normal modes of the system. This allows all system variables to be treated on an equal basis. Henceforth, the problem shall be addressed in terms of these normal modes:

$$\Delta\beta_i = \sum_j A_{ij}\Delta\theta_j . \quad (2)$$

Note that these normal modes do not usually represent the motion of a single grain's magnetization; typically, though, they will be localized.

The second derivative of the energy with respect to the normal mode (or modes) corresponding to $H=H_0$ will be zero. This is a consequence of the definition of a switching point: the point where a neighboring minimum and maximum meet. The assumption will be made that the third derivative with respect to the normal mode does not vanish. There are certain highly symmetric situations where this is not true; for example, the axially symmetric situation examined in the Néel model exhibits a third derivative equal to zero. However, the usual physical situation is not symmetric and the third derivative is not zero. The same physical intuition also prompts us to ignore the possibility of accidental degeneracies and thus simplify the analysis by considering a single switching mode.

The energy expansion may now be written as follows:

$$E = E_0 - \frac{\partial E}{\partial H}\bigg|_0 \Delta H + \frac{1}{2}\sum_i \frac{\partial^2 E}{\partial \beta_i^2}\bigg|_0 (\Delta\beta_i)^2 - \sum_i \frac{\partial^2 E}{\partial\beta_i\,\partial H}\bigg|_0 (\Delta\beta_i)(\Delta H)$$
$$+ \frac{1}{6}\sum_{i,j,k} \frac{\partial^3 E}{\partial\beta_i\,\partial\beta_j\,\partial\beta_k}\bigg|_0 (\Delta\beta_i)(\Delta\beta_j)(\Delta\beta_k) - \frac{1}{2}\sum_{i,j}\frac{\partial^3 E}{\partial H\,\partial\beta_i\,\partial\beta_j}\bigg|_0 (\Delta H)(\Delta\beta_i)(\Delta\beta_j) + O(\Delta H)^2 . \quad (3)$$

The next step is to solve for the $\Delta\beta_i$ by requiring that the system be at an energy extremum. Therefore,

$$\frac{\partial E}{\partial\beta_i} = \frac{\partial^2 E}{\partial\beta_i^2}\bigg|_0 \Delta\beta_i - \frac{\partial^2 E}{\partial\beta_i\,\partial H}\bigg|_0 \Delta H + \frac{1}{2}\sum_{j,k}\frac{\partial^3 E}{\partial\beta_i\,\partial\beta_j\,\partial\beta_k}\bigg|_0 (\Delta\beta_j)(\Delta\beta_k) - \sum \frac{\partial^3 E}{\partial H\,\partial\beta_i\,\partial\beta_j}\bigg|_0 (\Delta H)(\Delta\beta_j) = 0 . \quad (4)$$

We know that, aside from the switching mode $\Delta\beta_s$, the values of $\Delta\beta_i$ represent single-valued extensions about $\Delta\beta_i = 0$, since we have required that H_0 be the nearest switching point. This means that the second-order terms in $\Delta\beta_i$ may be neglected without affecting the lowest-order result. Therefore,

$$\Delta\beta_i = \frac{-\frac{1}{2}\,\partial^3 E/\partial\beta_i\,\partial\beta_s^2|_0(\Delta\beta_s)^2 + \partial^2 E/\partial H\,\partial\beta_i|_0\Delta H + \sum_{j\neq 1}\partial^3 E/\partial H\,\partial\beta_i\,\partial\beta_j|_0(\Delta H)(\Delta\beta_j)}{\partial^2 E/\partial\beta_i^2|_0 - \partial^3 E/\partial H\,\partial\beta_i^2|_0\Delta H} . \quad (5)$$

Thus $\Delta\beta_i$ is of order ΔH or $(\Delta\beta_s)^2$ for $i \neq s$. Insertion into (3) yields, for the case of $\Delta\beta_i = O(\Delta H)$, a contribution to the energy of order at least $(\Delta H)^2$. Similarly, the case $\Delta\beta_i = O(\Delta\beta_s)^2$ produces terms of order $(\Delta\beta_s)^4$ and $(\Delta H)(\Delta\beta_s)^2$, which are of higher order than other nonzero terms already in (3). Thus, the nonswitching modes of the system either make no contribution to the lowest-order term or contribute at order $(\Delta H)^2$.

Equation (4) may now be solved for the switching mode to yield

$$\Delta\beta_s = \pm[2\Delta H(\partial^2 E/\partial\beta_s\partial H)|_0/(\partial^3 E/\partial\beta_s^3)|_0]^{1/2} + O(\Delta H) . \quad (6)$$

This corresponds to the values of $\Delta\beta_s$ at the local energy minimum and at the local saddle which the magnetization must cross to switch. The height of this barrier is given by inserting the values of $\Delta\beta_s$ into (3) to yield

$$\Delta E = (\Delta H)^{3/2}[4(2\,\partial^2 E/\partial\beta_s\,\partial H|_0)^3/(9\,\partial^3 E/\partial\beta_s^3|_0)]^{1/2} . \quad (7)$$

Thus, one concludes that

$$k_B T \ln(2A\tau) = C(\Delta H)^{3/2} , \quad (8)$$

where C is the scaling constant contained within the square root of (7).

The careful reader may be concerned that, while the above argument is technically correct, the predicted effects may not be observable. One concern might be that in a macroscopic system, there are a huge number of switching modes available, and thus the constraint that we are considering only the nearest switching field may mean that the predicted dependence is only applicable at impossibly low temperatures (where $k_B T$ includes only one mode). However, the dependence on the magnetostatic interaction on an inverse-distance-cubed law means that only nearby grains make independently meaningful contributions to a switching unit's applied field. This means that only a relatively small number of switching modes interact significantly. Thus, the $(\Delta H)^{3/2}$ dependence should be observable at reasonable temperatures.

Further refinement in our knowledge of the range of the validity for the $(\Delta H)^{3/2}$ law can be obtained by extending the energy expansion to fifth order. The analysis proceeds in a similar, although much more tedious, fashion to that presented above. The result is that the next-highest-order term is proportional to $(\Delta H)^{5/2}$. The absence of a $(\Delta H)^2$ term can be attributed to the loss of sign (between minima and maxima) in the $(\Delta\beta_s)^4$ dependence. The coefficient of the $(\Delta H)^{5/2}$ term is complex; it may be attributed to a contribution from higher-order terms within the same mode and a contribution from interacting modes. The interacting contribution depends greatly on the interaction strength and no generalities will be attempted here. The higher-order contributions within the same mode will be explored by limiting our preceding general discussion to a special case of sufficient simplicity that the coefficient C of the $(\Delta H)^{3/2}$ term can be analytically evaluated and the resulting energy barrier compared to exact results calculated numerically.

The special case to be considered is the very popular model originally proposed by Stoner and Wohlfarth.[11] This model assumes a coherently rotating magnetization vector characterized by an angle θ relative to a uniaxial anisotropy axis of magnitude K. A magnetic field is applied at an angle ψ relative to this same axis. The resulting energy expression is

$$E = KV\sin^2\theta - HMV\cos(\theta-\psi) . \qquad (9)$$

The model lacks interactions; this means that the normal modes correspond to individual excitation of the magnetic particles and may be replaced by the variable θ for each particle. The partial derivatives identified in (7) can be taken; use of the minima condition $\partial E/\partial\theta = 0$ yields the following expression:

$$\Delta E = 4KV(2\Delta H/3H_0)^{3/2}\sin\theta_0\cos\theta_0 . \qquad (10)$$

The potentially unknown switching field H_0 and switching angle θ_0 may be found by setting $\partial E/\partial\theta = \partial^2 E/\partial\theta^2 = 0$ and solving. Some algebra produces

$$\tan\theta_0 = -\tan^{1/3}\psi ,$$

and

$$H_0 = \frac{2K}{M}\frac{(1-\tan^{2/3}\psi+\tan^{4/3}\psi)^{1/2}}{1+\tan^{2/3}\psi} . \qquad (11)$$

We thus have an explicit expression for the scaling constant C of this common model.

Table I exhibits values of the energy barrier calculated numerically to all order of ΔH in comparison to the first-order expression derived in the previous paragraph. There is clearly very good agreement (within a few percent) to $\Delta H/H$ values as high as 0.5. The only exception occurs for applied field angle ψ within a few degrees of zero where a special axial symmetry develops and the $\partial^2 E/\partial\theta\partial H$ derivative vanishes. Higher orders of ΔH would be required here. This then suggests that our expansion actually extends into physically important regions such as small particles near room temperature.

Our results affect the predictions of a number of previous papers. Most obviously, the effect of time in relaxing a system changes from a $[\ln(2A\tau)]^{1/2}$ dependence to a $[\ln(2A\tau)]^{2/3}$ dependence. Less obviously, the energy barrier preventing fluctuation-induced switching will tend to be larger than the Néel result for a given ΔH. This will lead workers to claim lower effective switching volumes based on their measurements. A particularly interesting change will be found in the prediction of Charap[5] for the maximum rate of magnetization decay normalized to the saturation magnetization. Using Néel's model, he obtained approximately a $T^{1/2}$ variation

TABLE I. Energy barriers ΔE as a function of the uniaxial anisotropy constant K, saturation magnetization M, the angle ψ between applied field and anisotropy axis, and the nonthermally assisted switching field H_0. Barrier heights are given both exactly and using the first-order expression.

ψ		$\Delta E(0.9h_0)/2KV$		$\Delta E(0.5h_0)/2KV$	
(deg)	$h_0 = H_0M/2K$	All orders	First order	All orders	First order
180	1.000	0.005	0.000	0.125	0.000
175	0.766	0.013	0.013	0.161	0.143
165	0.615	0.016	0.016	0.176	0.175
150	0.524	0.017	0.017	0.183	0.189
135	0.500	0.017	0.017	0.184	0.192
110	0.574	0.016	0.016	0.178	0.182

by assuming a Lorentzian switching-field distribution; the new result would be $T^{2/3}$.

Experimental verification of these predictions would obviously be desirable. It is easy to show that the experimental measurements of Oseroff *et al.*[12] for coercivity as a function of time (their Fig. 4) are fitted by a $(\Delta H)^{3/2}$ dependence very well. (The comparison is made for the five swept-field points since this avoids their discrepancy as they move to a "jumped" field.) However, a $(\Delta H)^2$ dependence cannot be ruled out. An accurate determination of the exponent by this method may require several measurements at still higher frequency. Alternatively, it may be possible to exploit very-low-temperature measurements of the maximum magnetization decay rate to test the $T^{2/3}$ dependence discussed in the previous paragraph. Here, special care must be taken that the applied field and temperature are very constant. This measurement was attempted by Tobin *et al.*;[13] unfortunately, their data are too scattered at the lowest temperatures to be reliably fitted with an exponent, and a fit to higher temperatures shows a strong dependence of the exponent on the cutoff temperature. (A least-squares fit to their data yields an exponent of 0.66 for a cutoff temperature of 13 K versus an exponent of 0.52 for a cutoff temperature of 25 K.) Finally, a test of an orientational-dependent switching field (such as the remanent coercivity) may be possible if very small individual particles can be isolated.

In conclusion, a scaling relationship between measurement time and switching field has been derived. The predicted $(\Delta H)^{3/2}$ dependence differs from the commonly used Néel-model $(\Delta H)^2$ dependence: This is because the Néel model is derived for a special case of high symmetry which is usually not applicable. The new scaling relationship implies that the maximum magnetic viscosity at low temperature should exhibit a $T^{2/3}$ dependence and that phenomenologically obtained switching volumes are actually smaller than previously thought. An expression for the orientational dependence of the time-dependent switching field in a particle possessing uniaxial anisotropy and a coherently rotating magnetization is obtained. Several of these predictions offer opportunities for experimental verification; such testing would be very desirable.

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