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RELAXATION TIME CONSTANT OF FERROMAGNETIC SINGLE-DOMAIN PARTICLES

by

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ABSTRACT

A Brownian motion treatment was applied to the Néel model for superparamagnetism and the relaxation time was calculated. A comparison with the published results was made. It was shown that the fluctuations due to magnetostriction are too small to account for the dissipation mechanism.

CONTENTS

1. INTRODUCTION

It is well known that a particle of ferromagnetic material, if its size is sufficiently small, has a uniform magnetization. The upper critical size was estimated by C. Kittel (1) and is of order of 100 Å in radius for an iron particle. Let the total magnetic moment of the particle be $\underset{\sim}{M}$ and let it make an angle Θ with an applied field $\underset{\sim}{H}$. In thermal equilibrium Θ will be determined by the Boltzmann distribution and for a system of such independent spherically symmetric particles the total magnetization $\underset{\sim}{M}_t$ is given by the familiar Langevin equation $\underset{\sim}{M}_t/\underset{\sim}{M} = \coth x - 1/x$ with $x = MH/kT$. The situation is quite similar to the case of atomic paramagnetism. The difference lies in the enormous magnitude of $\underset{\sim}{M}$ which is of the order of 10^4-10^5 Bohr magnetons.

For a single-domain particle there exists a crystal anisotropy energy (let it be uniaxial) $KV\sin^2\Theta$ (K:constant, V: volume of the particle, Θ: angle between $\underset{\sim}{M}$ and easy direction) and the total energy is given by $E = M.H. \cos\Theta + KV \sin^2\Theta$ ($\underset{\sim}{H}$ // easy direction). This time Θ follows a different distribution and $\underset{\sim}{M}_t$ can no longer be expressed by the Langevin function but takes a more complicated form.

E has a maximum near $\Theta = \pi/2$ and two minima near $\Theta = 0, \pi$ for not too large a field compared with K/M. On displacement of equilibrium by application or removal of a field, $\underset{\sim}{M}$ goes from one energy valley to another between which there is a potential barrier of the order of KV. This excess energy must be supplied by thermal activation. In this case $\underset{\sim}{M}_t$ changes according to (2)

$$\underset{\sim}{M}_t = \underset{\sim}{M}_2 + (\underset{\sim}{M}_1 - \underset{\sim}{M}_2) \exp(-t/\tau)$$

where

$\underset{\sim}{M}_1$: initial equilibrium magnetization before displacement

$\underset{\sim}{M}_2$ " final equilibrium magnetization after displacement

t : time measured from displacement

This relaxatiom time τ is given by $1/\tau = f_o \exp(-KV/kT)$. Bean and Livingston take $f_o \sim 10^9$ sec the reason for which is not entirely obvious (2). According as the experimental times are smaller or larger than the relaxation time, the system of particles exhibit remanence or reaches thermal equilibrium. This transition is quite sensitive to the particle size to temperature of measurement ratio and from the standpoint of experiment the critical size can be well defined for each temperature of measurement. In other words, there exists for a certain size, a particular temperature above which the system behaves paramagnetically. A rough estimation gives for this temperature T_b (blocking temperature) $KV/kT_b \sim 25$.

A more accurate calculation of the pre-exponential factor f_o was first done by L. M. Néel (3) in his attempt to explain some peculiar behavior of rock-magnetism. This derivation was criticized by W. F. Brown (4), who gave a different result later. The former is based on a particular model while the latter is completely free from any particular mechanism. Although it was shown (4) that these two yield practically the same numerical order of magnitude for the relaxation time, the interrelation is not apparent because of the difference in the derivation.

Our interest is, therefore, to see what result the Néel model will give when the Brown analysis is applied to it. It turns out that the

problem is essentially to specify a dissipation coefficient in the Brown formula.

2. Statement of the Problem.

Take a spherical ferromagnetic particle immersed in a non-magnetic matrix. Its free energy per unit volume is given by (5)

$$F(\underset{\sim}{M}) = -\underset{\sim}{H}\cdot\underset{\sim}{M} + K(\underset{\sim}{M}) - \frac{3G\lambda}{M_s^2}\underset{\sim}{M}\,A\,\underset{\sim}{M} \qquad (2.1)$$

where $\underset{\sim}{H}$: external magnetic field

$\underset{\sim}{M}$: magnetization vector , M_s: its magnitude

$K(\underset{\sim}{M})$: crystal anisotropy energy

A: strain tensor

G: elastic shear modulus

λ: magnetostrictive constant

We define the effective field acting on the system by

$$\underset{\sim}{H}_{eff} = \left\{ H - \nabla_M K(\underset{\sim}{M}) \right\} + \frac{6G\lambda}{M_s^2 V}\underset{\sim}{M}\int A\,d\tau$$

$$= \underset{\sim}{H}_{pot} + \underset{\sim}{H}_{random} \qquad (2.2)$$

This is determined as follows (6). In equilibrium $F(\underset{\sim}{M})$ takes a minimum. Therefore, for $\underset{\sim}{M} \to \underset{\sim}{M} + \delta\underset{\sim}{M}$

$$\delta F(\underset{\sim}{M}) = \left\{ -\underset{\sim}{H} + \nabla_M K(\underset{\sim}{M}) - \frac{6G\lambda}{M_s^2 V}\underset{\sim}{M}\int A\,d\tau \right\}\cdot\delta\underset{\sim}{M}$$

$$\equiv -\underset{\sim}{H}_{eff}\cdot\delta\underset{\sim}{M} = 0$$

Since $\underset{\sim}{M}^2$ = constant, $\underset{\sim}{M}\cdot\delta\underset{\sim}{M} = 0$. In other words, $\underset{\sim}{H}_{eff} // \underset{\sim}{M}$. This means that the torque acting on the system is zero and surely represents an equilibrium condition. The sign of $\underset{\sim}{H}_{eff}$ is chosen so that it is the same as that of external field. This definition leaves some arbitrariness in $\underset{\sim}{H}_{eff}$ by a quantity proportional to $\underset{\sim}{M}$. But this does not influence the result because in the equation of motion for $\underset{\sim}{M}$ (4.3) the vector

product operation of the right hand side automatically removes this ambiguity. The importance of the effective field was first pointed out by C. Kittel (7),(8) in his interpretation of ferromagnetic resonance. On defining this effective field it is implicitly assumed that the local bending of $\underset{\sim}{M}$ can be neglected. In order to discuss the dynamic behavior of $\underset{\sim}{M}$, we have room to add to (2.2) a dissipation term of the form $-\eta^* \, d\underset{\sim}{M}/dt$ (t: time) without which the energy transfer between the system and the surroundings is impossible. Thus,

$$\underset{\sim}{H}_{eff} = \underset{\sim}{H}_{pot} + \underset{\sim}{H}_{random} - \eta^* \, d\underset{\sim}{M}/dt \qquad (2.3)$$

The assumption of the Néel model is that the strains are caused by Debye waves so that the second term of the right hand side is a random variable. In order to specify the magnitude of η^*, we make use of the fluctuation-dissipation theorem which relates the correlation function of the random field with a dissipation force (9),(10),(11). So long as the response time of the system is small compared with the correlation time of the random field, this theorem is reduced to the conventional Brownian motion treatment. We assume this to be our case. Then the theorem states,

$$\langle \underset{\sim}{H}_{random}(t) \cdot \underset{\sim}{H}_{random}(t+\tau) \rangle_{av} = \frac{2kT}{V} \eta^* \delta(\tau) \qquad (2.4)$$

where k: Boltzmann constant

T: absolute temperature

v: volume of the particle

$\delta(\tau)$: Dirac delta function

The choice of the form of the right hand side is by analogy to the Brownian motion case. Thus we have only to calculate the left hand side to determine η^*.

3. Specification of the Apparent Dissipation Constant.

We start with the Fourier analysis of the strain field. Imagine a plane wave of strain with a wave vector $\underset{\sim}{k}$, advancing in the direction (l,m,n). The strains caused thereby are (12)

$$e_{11} = -l\dot{u}/c \quad , \quad e_{22} = -m\dot{v}/c \quad , \quad e_{33} = -n\dot{w}/c$$

$$e_{23} = -\tfrac{1}{2}(m\dot{w}/c + n\dot{v}/c), \quad e_{31} = -\tfrac{1}{2}(n\dot{u}/c + l\dot{w}/c), \quad e_{12} = -\tfrac{1}{2}(l\dot{v}/c + m\dot{u}/c)$$

(3.1)

where (u,v,w) is a displacement vector and the dot denotes differentiation with respect to time. c is the velocity of sound waves. The e_{ij} are functions of the time and position of the system we are interested in. Further,

$$\tfrac{1}{2}E(\underset{\sim}{k}) = K.E. = \tfrac{1}{2}\cdot\rho\left\{\left(\frac{\partial u}{\partial t}\right)^2 + \left(\frac{\partial v}{\partial t}\right)^2 + \left(\frac{\partial w}{\partial t}\right)^2\right\}$$

(3.2)

where ρ : density of the particle in the unstrained state

$E(\underset{\sim}{k})$: total energy per unit volume due to this wave

We eliminate (u,v,w) from (3.1) and (3.2) by taking the time averages of the quantities $e_{ij}.e_{kl}$. They are (cf. appendix),

$$\langle \rho_{11} \cdot \rho_{11} \rangle_{av} = \frac{1}{\rho c^2} (1 - \cos^2\varphi \sin^2\vartheta) \cos^2\varphi \sin^2\vartheta \, E(\underset{\sim}{k})$$

$$\langle \rho_{22} \cdot \rho_{22} \rangle_{av} = \frac{1}{\rho c^2} (1 - \sin^2\varphi \sin^2\vartheta) \sin^2\varphi \sin^2\vartheta \, E(\underset{\sim}{k})$$

$$\langle \rho_{33} \cdot \rho_{33} \rangle_{av} = \frac{1}{\rho c^2} \sin^2\vartheta \cos^2\vartheta \, E(\underset{\sim}{k})$$

$$\langle \rho_{23} \cdot \rho_{23} \rangle_{av} = \frac{1}{4\rho c^2} \cdot (1 - \cos^2\varphi \sin^2\vartheta - 4\sin^2\varphi \sin^2\vartheta \cos^2\vartheta) E(\underset{\sim}{k})$$

$$\langle \rho_{31} \cdot \rho_{31} \rangle_{av} = \frac{1}{4\rho c^2} (1 - \sin^2\varphi \sin^2\vartheta - 4\cos^2\varphi \sin^2\vartheta \cos^2\vartheta) E(\underset{\sim}{k})$$

$$\langle \rho_{12} \cdot \rho_{12} \rangle_{av} = \frac{1}{4\rho c^2} (1 - 4\sin^2\varphi \cos^2\varphi \sin^2\vartheta) \sin^2\vartheta \, E(\underset{\sim}{k})$$

$$\langle \rho_{11} \cdot \rho_{22} \rangle_{av} = -\frac{1}{\rho c^2} \sin^2\varphi \cos^2\varphi \sin^4\vartheta \, E(\underset{\sim}{k})$$

$$\langle \rho_{22} \cdot \rho_{33} \rangle_{av} = -\frac{1}{\rho c^2} \sin^2\varphi \sin^2\vartheta \cos^2\vartheta \, E(\underset{\sim}{k})$$

$$\langle \rho_{33} \cdot \rho_{11} \rangle_{av} = -\frac{1}{\rho c^2} \cos^2\varphi \sin^2\vartheta \cos^2\vartheta \, E(\underset{\sim}{k})$$

(3.3)

where φ, ϑ are polar coordinates of the vector $\underset{\sim}{k}$ defined by

$$(l,m,n) = (\cos\varphi \sin\vartheta, \sin\varphi \sin\vartheta, \cos\vartheta)$$

We have neglected contributions from longitudinal waves which are small compared with those from transverse waves (6:100). Twenty-one ways of

combination of $\langle e_{ij} \cdot e_{kl} \rangle_{av}$ are possible nine of which average to zero, so only the non-zero averages are given in (3.3).

The number of modes of lattice waves with wave vectors lying between $\underset{\sim}{k}$ and $\underset{\sim}{k} + d\underset{\sim}{k}$ is (13)

$$1/(2\pi)^3 \cdot dk_1\, dk_2\, dk_3 \qquad \underset{\sim}{k} = (k_1, k_2, k_3) \qquad (3.4)$$

and each mode has an average energy

$$\frac{\hbar\omega}{2} + \frac{\hbar\omega}{\exp(\hbar\omega/KT) - 1} \qquad (3.5)$$

where $\hbar$: Dirac constant

ω : Frequency defined by $k = \omega/c$

Contributions from various waves are obtained by replacing $E(\underset{\sim}{k})$ in (3.3) by (3.5) times (3.4) and integrating over all possible $\underset{\sim}{k}$.

The correlation functions of $e_{ij}^{(1)}$ at point r_1, times 1, and $e_{kl}^{(2)}$ at point r_2, time 2 are given by Wiener-Kinchine's theorem (6),(7) for the 3-dimensional case

$$\langle e_{ij}^{(1)} \cdot e_{kl}^{(2)} \rangle_{av} = \frac{1}{(2\pi)^3} \iiint \langle e_{ij} \cdot e_{kl} \rangle_{av} \cos\omega\tau \times \cos(X_1 k_1) \cos(X_2 k_2) \cos(X_3 k_3)\, dk_1\, dk_2\, dk_3 \qquad (3.6)$$

where $\underset{\sim}{r} = (X_1, X_2, X_3) = \underset{\sim}{r}_1 - \underset{\sim}{r}_2$, $\tau = t_1 - t_2$

Simplifications of (3.6) are as follows. We note from (3.3) that $\langle e_{ij} \cdot e_{kl} \rangle_{av}$ are made of terms $\sin^{2m}\varphi \cos^{2n}\vartheta$ $(m,n = 0,1,2)$. Let's restrict ourselves to the case $m = 1$, $n = 2$. Let

$$k_1 x_1 = kr \cos\varphi \cos\zeta \sin\vartheta \sin\xi$$
$$k_2 x_2 = kr \sin\varphi \sin\zeta \sin\vartheta \sin\xi$$
$$k_3 x_3 = kr \cos\vartheta \cos\xi$$

Integration of (3.6) with respect to φ is

$$I = \int\int \sin^2\varphi \cos^4\vartheta \cos k_1 x_1 \cos k_2 x_2 \cos k_3 x_3 \sin\vartheta \, d\vartheta \, d\varphi$$

$$= \tfrac{1}{4} \int d\vartheta \int d\varphi \, (1 - \cos 2\varphi) \left[\cos\{kr\cos(\varphi - \zeta)\} + \cos\{kr\cos(\varphi + \zeta)\} \right] \cos k_3 x_3$$

$$= \pi \int_0^{\pi} \{ J_0(kr \sin\vartheta \sin\xi) \cos(kr\cos\vartheta\cos\xi) + \cos 2\zeta \, J_2(kr\sin\vartheta\sin\xi) \cos(kr\cos\vartheta\cos\xi) \} \cos^4\vartheta \sin\vartheta \, d\vartheta$$

We may drop the second term which integrates to zero with respect to φ. Using Gegenbauer's integral (14),

$$= \pi/35 \{ 8 \left(2\pi/kr\right)^{1/2} P_4(\cos\xi) J_{9/2}(kr) - 20 \left(2\pi/kr\right)^{1/2} P_2(\cos\xi) J_{5/2}(kr) + 7 \left(2\pi/kr\right)^{1/2} P_0(\cos\xi) J_{1/2}(kr) \}$$

(3.7)

We may leave only the lowest order Bessel function for the same reason as we did J_2 above. Similarly,

m = 0 ,	n = 0 ,	I = 120
0	1	40
0	2	20
1	0	60
1	1	24
1	2	12
2	0	45
2	1	15
2	2	9

(in relative units) (3.8)

Using (3.3) and (3.8), we get for (3.6)

$$\begin{aligned} \langle e^{(1)}_{11}\cdot e^{(2)}_{11}\rangle_{av} &= \langle e^{(1)}_{22}\cdot e^{(2)}_{22}\rangle_{av} = \langle e^{(1)}_{33}\cdot e^{(2)}_{33}\rangle_{av} = 16 \\ \langle e^{(1)}_{12}\cdot e^{(2)}_{12}\rangle_{av} &= \langle e^{(1)}_{23}\cdot e^{(2)}_{23}\rangle_{av} = \langle e^{(1)}_{31}\cdot e^{(2)}_{31}\rangle_{av} = 12 \\ \langle e^{(1)}_{11}\cdot e^{(2)}_{22}\rangle_{av} &= \langle e^{(1)}_{22}\cdot e^{(2)}_{33}\rangle_{av} = \langle e^{(1)}_{33}\cdot e^{(2)}_{11}\rangle_{av} = -8 \end{aligned} \qquad (3.9)$$

Next we go to the polar coordinate $(\phi, \theta, 1)$, and resolve H_{random} into ϕ - and θ - components. A geometrical consideration gives the following relations for the direction cosines.

$$\begin{array}{c} \\ \theta \\ \phi \\ r \end{array} \begin{array}{c} \begin{array}{ccc} x & y & z \end{array} \\ \begin{bmatrix} \cos\phi\cos\theta & \sin\phi\cos\theta & -\sin\theta \\ -\sin\theta & \cos\phi & 0 \\ \cos\phi\sin\theta & \sin\phi\sin\theta & \cos\theta \end{bmatrix} \end{array} \quad \ldots\ldots T$$

The inverse T^{-1} of the above matrix T is

$$\begin{bmatrix} \cos\phi\cos\theta & -\sin\phi & \cos\phi\sin\theta \\ \sin\phi\cos\theta & \cos\phi & \sin\phi\sin\theta \\ -\sin\theta & 0 & \cos\theta \end{bmatrix} \quad \ldots\ldots T^{-1}$$

Noting that the transformed $\underset{\sim}{M}$ takes the form $(0, 0, 1)$, the required quantities are determined from

$$T\underset{\sim}{A}\underset{\sim}{M} = TAT^{-1}.\quad T\underset{\sim}{M} = TAT^{-1}.\quad (0, 0, 1)^* \qquad A = (e_{ij}),\ *: \text{transposed}$$

where we have skipped the averaging operation over the volume and the proportional constant $\frac{6G\lambda^2}{M\omega^2 V}$. They are

$H_1(t)$ ... θ -component

$$= e_{11}\sin\theta\cos\theta\cos^2\phi + e_{13}\cos\phi(\cos^2\theta - \sin^2\theta)$$
$$+ e_{22}\sin^2\phi\cos\theta\sin\theta + e_{23}\cos\phi(\cos^2\theta - \sin^2\theta)$$
$$- e_{33}\sin\theta\cos\theta + 2e_{12}\sin\phi\cos\phi\sin\theta\cos\theta$$

$H_2(t)$ ... ϕ -component

$$= -e_{11}\sin\phi\cos\phi\sin\theta + e_{12}(\cos^2\phi - \sin^2\phi)\sin\theta$$
$$+ e_{22}\sin\phi\cos\phi\sin\theta - e_{13}\sin\phi\cos\theta$$
$$+ e_{23}\cos\phi\cos\theta$$

(3.10)

Strictly speaking, ϕ. θ are functions of strains. But if the magnitude of the random field is sufficiently small, $\underset{\sim}{M}$ will undergo precession the angular velocity of which is determined by H_{pot}, to a first approximation. Further, if the response time of the system is large, $\underset{\sim}{M}$ will stay at a fixed position during a time interval of the order of correlation time of the strain field. We assume so. Under this approximation, the correlation functions of the random field are by making use of (3.8) and (3.10)

$$\langle H_1(t).H_1(t+\tau)\rangle_{av} = \langle e_{12}^{(1)}.e_{12}^{(2)}\rangle_{av}$$
$$\langle H_2(t).H_2(t+\tau)\rangle_{av} = \langle e_{12}^{(1)}.e_{12}^{(2)}\rangle_{av}$$
$$\langle H_1(t).H_2(t+\tau)\rangle_{av} = 0$$
$$\langle H_2(t).H_1(t+\tau)\rangle_{av} = 0 \qquad (3.11)$$

These are independent of the direction of $\underset{\sim}{M}$ as they should be.

The final correlation function of the random field is obtained by averaging (3.11) with respect to the volume of the particle, according to (2.2). Changing the variables r_1 to $r_1 - r_2 = r$, collecting all the factors hitherto neglected,

$$\langle H_{random}(t) H_{random}(t+\tau)\rangle_{av} \qquad \omega = \omega_D$$
$$= \frac{1}{(2\pi)^3} \cdot \frac{2\pi \times 12}{120} \cdot \frac{36 G^2 N^2}{\rho^2 M_S^2 V} \cdot 4\pi \int_0 \left[\frac{\hbar\omega}{2} + \frac{\hbar\omega}{\exp(\hbar\omega/KT) - 1}\right]$$
$$\times k^2 \omega_D \omega \tau \, dk \int_0^r \left(\frac{2\pi}{kr}\right)^{1/2} J_{1/2}(kr)\, r^2 dr \; I$$

I: standard dyad (3.12)

This integration is elementary(15) so long as we replace $\hbar\omega_D/KT$ by infinity (zero point energy neglected). This does not necessarily mean that we limit ourselves to low temperature. First, the Debye temperatures (H) of iron and nickel are comparatively large: 467, 456°K, respectively. The Curie temperature of nickel is of this order, though for iron it is larger. As long as we are interested in the temperature range in which the magnetization remains constant, temperature cannot be too large. Thus $0.5 \lesssim (H)/T < \infty$. Second, as k gets larger the contribution to the integral becomes small because the trigonometric factor in $J_{1/2}$ oscillates and the exponential factor in the denominator becomes large. Therefore, the error introduced will not be serious. Then (3.12) is

$$= \text{const} \; kT/2 \int_0^\infty d\xi \frac{1}{e^\xi - 1} \cdot \left[(\sin p\xi + \sin q\xi) - \beta\xi (\cos p\xi + \cos q\xi) \right]$$

with $\alpha = kT\tau/\hbar$, $\beta = kT/\hbar \cdot r/c$, $p = \alpha - \beta$, $q = \alpha + \beta$

Using $$\int_0^\infty \frac{\xi \cos p\xi}{e^\xi - 1} d\xi = \partial/\partial p \left[\pi/2 \left(\frac{e^{p\pi} + e^{-p\pi}}{e^{p\pi} - e^{-p\pi}} \right) - 1/2p \right]$$

$$\int_0^\infty \frac{\xi \sin p\xi}{e^\xi - 1} d\xi = \quad [\quad " \quad]$$

The calculation is straightforward.

It turns out that according as $\tau \gtrless r/c$, this is ~ 0 or $1/2 \cdot \pi kT$. That is, when $\langle H(t).H(t+\tau) \rangle_{av}$ is regarded as a function of τ, the main contribution comes from a narrow range of the order of r/c. We introduce Dirac function $\delta(\tau)$ so that the area bounded by $\langle H(t).H(t+\tau) \rangle_{av}$ and τ-axis be written as $2kT/v \cdot \eta^* \delta(\tau)$. After all,

$$\langle H_{random}(t) . H_{random}(t+\tau) \rangle_{av} = \frac{2kT}{V} \eta^* \delta(\tau)$$

with $$\eta^* = \frac{18}{5} \cdot \frac{GN^2}{Ms^2} \cdot r/c$$

(3.12)

4. Calculation of Relaxation Time Constant.

The following approach is due to W. F. Brown (4). Assume we have an ensemble composed of independent systems similar to the one we have had in mind. The state of the ensemble is specified by the distribution of points over the surface of a unit sphere in the phase space, each point corresponding to a system. The conservation of the number of particles requires

$$\partial \rho / \partial t + \operatorname{div} \underset{\sim}{J} = 0 \tag{4.1}$$

where ρ: density of points

$\underset{\sim}{J}$: flux of points , in phase space

Further,

$$\underset{\sim}{J} = \rho \underset{\sim}{v} , \qquad \underset{\sim}{v} = \dot{\underset{\sim}{M}} / M_s \tag{4.2}$$

$\underset{\sim}{v}$: velocity of points

$\underset{\sim}{M}$ is supposed to be governed by Gilbert's equation (17)

γ : gyroscopic const.

$$1/\gamma \cdot d\underset{\sim}{M}/dt = \underset{\sim}{M} \times (H_{pot} - \nu \, d\underset{\sim}{M}/dt) \tag{4.3}$$

ν: dissipation const.

Instead of taking into account explicitly the random field in (4.3) add a diffusional term something like $-k' \operatorname{grad} \rho$ to $\underset{\sim}{J}$ (19). Thus we get from (4.1), (4.2), (4.3)

$$\begin{aligned} \sin\theta \, \partial \rho / \partial t = {} & \partial/\partial\theta \{ \sin\theta [(h' \partial V/\partial\theta - g' \tfrac{1}{\sin\theta} \partial V/\partial\theta) \rho \\ & + k' \partial \rho/\partial\theta] \gamma + \partial/\partial\phi [(g' \partial V/\partial\theta + h' \tfrac{1}{\sin\theta} \partial V/\partial\phi) \rho \\ & + k' \tfrac{1}{\sin\theta} \partial \rho/\partial\phi] \end{aligned} \tag{4.4}$$

where $h' = \dfrac{\eta}{1/\gamma^2 + \eta^2 M_s^2}$, $g' = 1/M_s\gamma \cdot \dfrac{1}{1/\gamma^2 + \eta^2 M_s^2}$

k' is determined so that ρ approaches the Boltzmann distribution in the limit $t \to \infty$. For the potential

$$V(\theta) \equiv K(M) = 1/2 . M_s H_c \sin^2\theta \quad , \quad \underset{\sim}{H} = \text{external field} \equiv 0 \qquad (4.5)$$

(4.4) becomes

$$\sin\theta \, \partial\rho/\partial t = \partial/\partial\theta \{ \sin\theta [(h' \partial V/\partial\theta - g'/\sin\theta \cdot \partial V/\partial\theta)\rho + k' \partial\rho/\partial\theta] \}$$

(4.6)

assuming that ρ is independent of ϕ .

(4.6) does not allow of an analytical solution, but variational methods yield for the minimum p such that $\rho(t) = \rho(t=\infty) + A F(\theta) e^{-pt}$

$$P_1 = \frac{\eta M_s}{1/\gamma^2 + \eta^2 M_s^2} \cdot H_c \left(\frac{2 H_c M_s V}{\pi k T} \right)^{1/2} e^{-H_c M_s V / 2kT}$$

(4.7)

For our model η must be replaced by η^* , and the corresponding p is, noting $1/\gamma \gg \eta^* M_s$

$$P_2 = \frac{8}{5} \cdot \gamma^2 G N^2 \frac{V}{L} \cdot H_c \left(\frac{2 H_c N}{\pi M_s k T} \right)^{1/2} e^{-H_c M_s N / 2kT}$$

(4.8)

For comparison's sake we quote Néel's result

$$P_1 = \tfrac{1}{2}\gamma\left(\frac{2N}{\pi KTG}\right)^{1/2} |3GN + DM_s^2|\, H_c\, e^{-H_c M_s N/2KT}$$

(4.9)

D : demagnetization factor

5. Discussion.

Let's check the validity of the assumption we made in deriving (3.11). We neglected the correlation of ϕ, θ with Debye waves. This is justified if,

$$r/c \ll (\text{gyroscopic angular velocity})^{-1} \qquad (5.1)$$

For an iron particle $r = 100\text{Å}$, $c_{44} = 1.16\times10^{12}$ dynes/cm^2

$$\rho = 7.87 \text{g/cm}^3$$

$$r/c = 10^{-6} \times \sqrt{7.87/1.16.10^{12}} = 2.6\times10^{-12}\text{sec}$$

while the right hand side of (5.1) is (neglecting the random field)

$$1/\omega = 1/(\gamma H_c \cos\theta) \geq 1/(1.76\times10^{7}\times10^{3}) = 5.7\times10^{-10} \text{ sec.}$$

Thus we see that (5.1) holds approximately in our case, however, is liable to break for larger r, and H_c. Then we have to abandom the white spectrum assumption on ν^*. Under the same approximation H_{random} follows a Gaussian distribution (18), (19).

One of the conspicuous differences between (4.8) and (4.7) or (4.9) is that p_1 or p_3 is a function of v/T while p_2 is not. The existence of the factor r/c is similar to the usual Brownian motion case, though the physical meaning is entirely different. This factor reduces the magnitude of our dissipation constant enormously. That is, for G = 0.77 x 10^{12} dynes/cm^2, M_s = 1700 0_e, $\nu = 2\times10^{-5}$

$$\nu^* M_s = \frac{18}{5} \frac{G\nu^2}{M_s} \cdot r/c \sim 10^{-12} \qquad (5.2)$$

On the other hand from ferromagnetic resonance experiments νM_s is estimated to be of the order of $1/\gamma$. Thus there is a discrepancy of a factor 10^{-4}.

However, with all the above differences, the dependence of τ on v/T is not too severely influenced. A numerical comparison between the above three formulas yields the following result.

For an iron particle

τ (sec)	Néel	Brown $\gamma = \gamma'$	Brown $\gamma' = \gamma/10$	Present r = 30Å	Present r = 100Å
10^{-1}	3.2	3.6	3.2	1.9	2.0
10	3.9	4.4	4.0	2.6	2.7
10^{3}	4.7	5.1	4.8	3.4	3.5
10^{5}	5.4	5.9	5.5	4.2	4.3

Figures are v/T ratios in 10^{-21} cm^3/deg(Kelvin) unit.

Since the real magnitude of the dissipation coefficient is not known, two cases are given the Brown formula. Further, in our formula we have arbitrarily set the particle radius to some value. This effect is not serious as can be seen above. The exponential factor is so effective that the final result does not reflect the difference conspicuously with all the smallness of our dissipation constant.

The difference in form between (4.8) and (4.9) arises from the assumption on the magnitude of the response time. Néel assumed it to be small, which we have seen to fail.

Up to now we have avoided the discussion on what the microscopic meaning of our model is. This is not necessary because where there are fluctuations there is a dissipation. It is certain that magnetostriction will contribute to fluctuations. But as we have seen its effect is very small.

Needless to mention, magnetostriction is not the only mechanism

conceivable. In a paramagnetic matrix, dipole field fluctuations are effective at a long distance and the problem will be a statistical investigation of Lorentz field. In a diamagnetic material the interaction between the conduction electrons and spin comes into effect.

In both cases the problem does not allow of a simple classical description. Probably field-theoretic techniques combined with quantum statistical mechanics, especially Green function method, will afford a satisfactory approach. This is because the problem necessitates a collective motion treatment.

It may be possible that the reversal of the magnetization vector might take place in a non-coherent way in which case the activation energy can be small (22), contrary to the assumption we have made.

In view of the present lack of knowledge on the magnetic transport phenomena it is of vital importance to investigate this problem further. It is hoped that the proposed method, when applied to a reasonable model, will help clarify the problem (20),(21).

6. Conclusion.

It was shown that magnetostriction makes only a small contribution to the spontaneous fluctuations of magnetization vector, and that the Néel model when the Brown analysis is applied to it yields a different result from the one derived in a intuitive manner and the cause was pointed out. Also another approach was suggested.

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APPENDIX

Let

$$\underset{\sim}{k}/|\underset{\sim}{k}| = (l, m, n) = (\cos\varphi \sin\vartheta, \sin\varphi \sin\vartheta, \cos\vartheta)$$

In an isotropic solid there is one longitudinal wave (its polarization vector p_1) and two transverse waves ($\underset{\sim}{p_2}$, $\underset{\sim}{p_3}$) for a fixed $\underset{\sim}{k}$. In the former

$$\underset{\sim}{p_1} = \underset{\sim}{k}/|\underset{\sim}{k}|$$

As to the latter, div (u,v,w) = 0 is not sufficient to determine $\underset{\sim}{p_2}$ and $\underset{\sim}{p_3}$. However, since what matters in the final analysis is the averaged absolute value of amplitude of each wave, we take for convenience' sake

$$\underset{\sim}{p_2} = (p_1^{(2)}, p_2^{(2)}, p_3^{(2)}) = (-\sin\varphi, \cos\varphi, 0) \quad \cdots\cdots + \underset{\sim}{\varphi}$$

$$\underset{\sim}{p_3} = (p_1^{(3)}, p_2^{(3)}, p_3^{(3)}) = (-\cos\varphi \cos\vartheta, -\sin\varphi \cos\vartheta, \sin\vartheta) \cdots - \underset{\sim}{\vartheta}$$

Further put

$$(u^{(i)}, v^{(i)}, w^{(i)}) = \underset{\sim}{p_i}/|\underset{\sim}{V_i}| \qquad \underset{\sim}{V_i}: \text{ displacement vector}$$

Let's take $\langle e_{11} \cdot e_{22} \rangle_{av}$ as an example. From the definition

$$e_{11} \cdot e_{22} = lm \left\{ p_1^{(1)} p_2^{(1)} |\dot{V}_1|^2/C_l^2 + p_1^{(2)} p_2^{(2)} |\dot{V}_2|^2/C_t^2 + p_1^{(3)} p_2^{(3)} |\dot{V}_3|^2/C_t^2 \right\}$$

$c_l = C$: velocity of longitudinal waves

c_t : velocity of transverse waves

Using

$$\langle |\dot{V}_1|^2 \rangle_{av} = \langle |\dot{V}_2|^2 \rangle_{av} = \langle |\dot{V}_3|^2 \rangle_{av} = E(k)/\rho$$

and neglecting the first term (it can be shown that the contribution

from this is $(Ct/\ell)^5$ ~ 1/20 part of the succeeding terms),

$$\langle e_{11}, e_{22} \rangle_{av} = \sin\varphi \cos\varphi \sin^2\vartheta \left(-\sin\varphi \cos\varphi + \sin\varphi \cos\varphi \cos^2\vartheta\right) \frac{E(k)}{p^2}$$

$$= -\frac{1}{C^2\rho} \sin^2\varphi \cos^2\varphi \sin^4\vartheta \, E(k)$$

The remaining combinations of e_{ij} can be dealt with in the similar manner.