RELAXATION OF SINGLE DOMAIN MAGNETIC PARTICLES

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In this article, we intend to describe a particularly illustrative application of the general theory of stochastic processes which has been discussed in great detail by various authors in this volume. The example is taken from the field of magnetism and is based on the work of Brown [1] . Brown considered the relaxation of single domain particles. This mechanism of relaxation has been invoked by a number of workers to understand relaxation and response behaviour of spin-glasses in recent years.

1. Description of the system

Let us begin the discussion by explaining what is meant by a 'single domain particle'. A body made up of magnetic material, say iron, has no magnetic moment under usual conditions even if it is well below its Curie temperature. This is so because the body gets divided into various domains in which the spontaneous magnetisation points in different directions. However, if the size of the body is reduced, there comes a point beyond which the body has just one domain. For example, iron particles having radius below 150 Å stay in a single domain. At this point, the magnetostatic energy, and the energy of forming domain wall compare in such a way that single domain state becomes preferable. Here we shall consider an assembly of such single domain particle which are assumed to be non-interacting [2] . The magnetic state of such a particle is characterised by its magnetisation vector $\vec{M}$

$$\vec{M} = vM_s\hat{n} \tag{1.1}$$

where v is the volume of the particle, M_s the saturation magnetisation, and $\hat{n}$ a unit vector. There are generally two contributions to the magnetic energy. The first one is due to the external field $\vec{H}$, given by $-\vec{M}.\vec{H}$, the second due to the magnetic anisotropy energy which arises from the crystalline structure of the material. The typical forms for anisotropy energy are

(i) Cubic Anisotropy (Example : iron)

$$E = -vA\,[n_x^4 + n_y^4 + n_z^4] \tag{1.2}$$

(ii) Uniaxial Anisotropy (Example : Cobalt)

$$E = +\,v\,K\,[\,1 - n_z^2\,] \tag{1.3}$$

where A and K are constants, and n_j (j = x,y,z) are the components of $\hat{n}$. Since the purpose of the article is illustrative, we shall consider henceforth particles of uniaxial anisotropy. Choosing the z-axis along the anisotropy axis, and denoting by θ the angle between the z-axis and $\hat{n}$, the anisotropy energy may be written as

$$E(\theta) = vK \sin^2\theta \tag{1.4}$$

2. Relaxation : a simple description

We now define a distribution function $f(\Omega)$, such that $f(\Omega)\, d\Omega$ denote the fraction of particles whose magnetisation vector $\hat{n}$ lies between solid angles Ω and $\Omega + d\Omega$. If the assembly is in thermal equilibrium

$$f_{eq}(\Omega)\, d\Omega \propto e^{-(vK/kT)\sin^2\theta} \sin\theta\, d\theta\, d\phi \tag{2.1}$$

If at time t=o, one starts with an arbitrary distribution $f(\Omega)$, the system eventually relaxes to the equilibrium distribution given in (2.1). However if $vK/kT \ll 1$, the equilibrium distribution is more or less uniform, and the relaxation occurs on a microscopic time scale. On the other hand, if $vK/kT \gg 1$, most of the particles in equilibrium are either near $\theta \simeq 0$ or $\theta \simeq \pi$. The relaxation now occurs in two stages as illustrated in Fig.1. In the first stage the particles in the region $0 \leqslant \theta < \frac{\pi}{2}$ quickly slide to the potential valley $\theta \simeq 0$, while those in the region $\frac{\pi}{2} < \theta \leqslant \pi$ slide into the potential valley $\theta \simeq \pi$. This process occurs on the microscopic time scale, but leads to

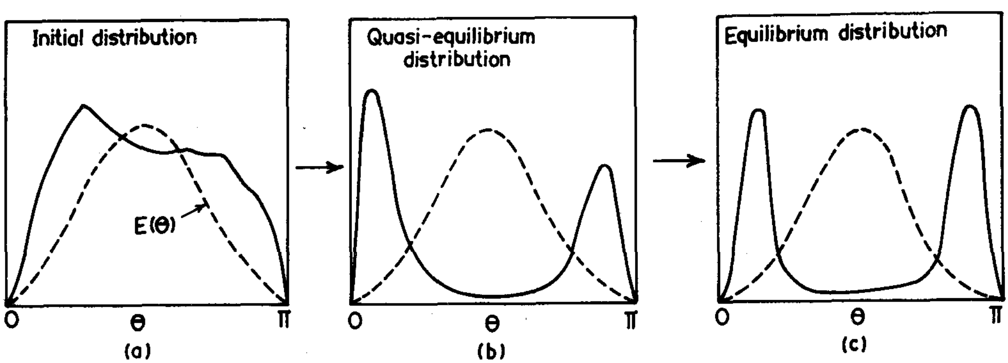

Fig.1 This series of figures depicts the evolution of the distribution function towards thermal equilibrium. Fig. a shows the initial distribution. The first stage of evolution leading to quasi-equilibrium distribution (Fig.b) occurs on microscopic time scale $\tau_\perp$. The second stage of evolution from quasi-equilibrium state occurs on much larger time scale of $\tau_\perp e^{\beta K v}$, as it involves thermally activated tunnelling across the barrier. Fig.c shows the equilibrium distribution.

only a quasi-equilibrium (or metastable) state, because though the equilibrium is achieved within each valley of the potential, the overall equilibrium between the two valleys is not reached (see Fig.1). In order that the global equilibrium may be established, the magnetic vector of some particles must go from one valley to the other. This requires a thermally activated tunnelling across a barrier, which is a far slower process. This sort of situation was considered by Neél long ago who described the evolution from quasi-equilibrium to global equilibrium in terms of simple rate equations [3] . Let us define the fractional populations in the two valleys n_1 and n_2 as follows

$$n_1(t) = \int_0^{\theta_1} f(\theta)\ \mathrm{Sin}\theta\ d\theta; \quad n_2(t) = \int_{\theta_2}^{\pi} f(\theta)\ \mathrm{Sin}\theta\ d\theta \tag{2.2}$$

where θ_1 and θ_2 are two arbitrary angles such that $\theta_1 \sim 0$ and $\theta \sim \pi$ (See Fig.1). Since the number of particles in other angular regions is negligible ($vK/kT \gg 1$), the thermal evolution can be well described in terms of merely $n_1(t)$ and $n_2(t)$. One can easily write down the rate equations for these two as

$$\frac{dn_1(t)}{dt} = -\frac{dn_2(t)}{dt} = -\nu_{12}\, n_1 + \nu_{21}\, n_2 \tag{2.3}$$

where ν_{12} and ν_{21} are the rates at which the particles reorient from one region to the other, and Neél assumed the usual Arrhenius form for these

$$\nu_{12} = \nu_{21} = \nu_o\ e^{-vK/kT} \tag{2.4}$$

where ν_o is some kind of an attempt frequency.

3. A First Principles Description of Relaxation : The Fokker Planck Equation

While the above approach is intuitively appealing, it needs justification from a more basic point of view. The basic theory should be able to describe the thermal evolution generally, i.e. in situations in which the restriction of large barrier (i.e. $vK/kT \gg 1$) can be removed. Moreover, in a basic theory, one should be able to derive the rate equations and find explicit expressions for the rates ν_{12} and ν_{21}, which incorporate precessional motion of spins, details of the potential barrier, etc. Brown essentially provided such a theory.

Brown began his considerations by first constructing an appropriate Langevin equation for the precession of the magnetisation vector $\vec{M}$ [4,5] . To construct this equation let us first write the deterministic equation of motion for $\vec{M}$, which reads

$$\frac{d\vec{M}}{dt} = \gamma \vec{M} \times (\vec{H}_e - \frac{\partial E(\vec{M})}{\partial \vec{M}}) = \gamma\, \vec{M} \times \vec{H} \tag{3.1}$$

where γ is the gyromagnetic ratio and $\vec{H}_e$ is the external field. This equation describes the precession of $\vec{M}$ around the total effective field $\vec{H}$, as shown in Fig.2. If we now let the particle interact with a heat bath, two new terms should be added to Eq. (3.1). First, there is the frictional term, which causes the precession-moment to spiral towards the axis of the effective field, so that it reaches the lowest energy state. Second, the bath exerts a random force which essentially provides thermal energy to the moment. These physical requirements, in addition to the requirement that the magnitude $\vec{M}$ is a fixed quantity, permit us to introduce the additional terms in the following way

$$\frac{d\vec{M}}{dt} = \gamma \vec{M} \times [\ \vec{H} - \eta(\vec{M}\times\vec{H}) + \vec{h}'(t)] \tag{3.2}$$

where η is the co-efficient of viscosity and $\vec{h}'(t)$ is a random field, which is taken

taken to be a stationary Gaussian random process. Its correlations are given by

$$\langle h'_\alpha (t_1)\, h'_\beta (t_2) \rangle = D_{\alpha\beta}\, \delta(t_1-t_2) \tag{3.3}$$

where we take

$$D_{xx} = D_{yy} = \frac{1}{\tau_\perp} \; ; \; D_{zz} = \frac{1}{\tau_\parallel} \; ; \; D_{xy} = D_{yz} = o \text{ etc.} \tag{3.4}$$

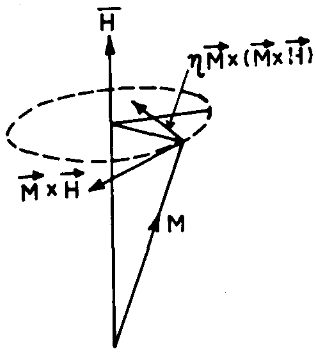

Fig. 2 Torques on the magnetisation vector according to Langevin equation.

Using the Langevin equation (3.2) and properties given in Eqs.(3.3) and (3.4), the Fokker-Planck equation can be derived following the methods described in this volume. In the present case the Fokker-Planck equation turns out to be

$$\frac{\partial}{\partial t} (\vec{M},t) = +\gamma\left[\vec{L}.\vec{H}\, f - \eta\vec{M} \times \vec{L}.\vec{H}\, f + 1/2\, L_\alpha D_{\alpha\beta} L_\beta\, f\right] \tag{3.5}$$

where we have defined the operator

$$\vec{L} = \vec{M} \times \frac{\partial}{\partial \vec{M}} \quad . \tag{3.6}$$

We shall now effect a simplification by considering an axially symmetric situation. We take the anisotropy energy E and the initial distribution $f(\theta,\phi,0)$ to be independent of ϕ, where θ,ϕ denote the direction of magnetisation in polar coordinates. Under these conditions $f(\theta,\phi,t)$ remains independent of ϕ for all t and Eq.(3.5) may be written as

$$\frac{\partial f(\theta,t)}{\partial t} = \frac{1}{2\tau_\perp} \frac{1}{\text{Sin}\theta} \frac{\partial}{\partial\theta} \left[\text{Sin}\theta \left\{ \frac{1}{kT} \frac{\partial E(\theta)}{\partial\theta} f + \frac{\partial f}{\partial\theta} \right\} \right] \tag{3.7}$$

where we have used the Einstein relation

$$2\tau_\perp \eta \gamma = \frac{1}{kT} \tag{3.8}$$

to ensure that $f(\theta,t)$ approaches the equilibrium distribution in the limit $t \to \infty$. As a first consequence of Eq. (3.5) let us consider the time evolution of the average component of magnetisation. In the case of zero anisotropy and an external field H_o in z-direction [7] one finds

$$\frac{d\langle M_z\rangle}{dt} = - \frac{\langle M_z\rangle}{T_1} + \frac{H_o}{kT} \frac{\langle M_x^2 + M_y^2\rangle}{2T_1} \tag{3.9}$$

$$\frac{d\langle M_x\rangle}{dt} = -\gamma H_o \langle M_y\rangle - \frac{\langle M_x\rangle}{T_2} - \frac{\eta H_o}{2kT} \frac{\langle M_z M_x\rangle}{T_1} \tag{3.10}$$

$$\frac{d\langle M_y\rangle}{dt} = \gamma H_o \langle M_x\rangle - \frac{\langle M_y\rangle}{T_2} - \frac{\eta H_o}{2kT} \frac{\langle M_z M_y\rangle}{T_1} \tag{3.11}$$

where $T_1 = \tau_{\parallel}$ and $T_2^{-1} = 1/2(\tau_{\parallel}^{-1} + \tau_{\perp}^{-1})$. These equations reduce to the well known Bloch equations in the limit $M_s H_o/kT \ll 1$. The Fokker-Planck equation discussed above can, in principle, describe relaxation in arbitrary situations. However, analytic results can be obtained only in certain simple cases. In the next section, we shall study the solution of the axially symmetric equation (3.7) in the large barrier limit.

4. Variational Solution in Large Barrier Limit

We can write Eq. (3.7) schematically as

$$\frac{\partial f}{\partial t'} = Df \tag{4.1}$$

where $t' = t/2\tau_{\perp}$ and

$$D = \frac{1}{\sin\theta} \frac{\partial}{\partial\theta} \left[\sin\theta \left\{ \frac{1}{kT} \frac{\partial E(\theta)}{\partial\theta} + \frac{\partial}{\partial\theta} \right\} \right]. \tag{4.2}$$

Noting that $e^{\beta E(\theta)} D$ is a Hermitian operator ($\beta = 1/kT$) and $D\, e^{-\beta E(\theta)} = 0$, we can write down the time dependent solution of Eq. (4.1) as

$$f(\theta,t) = A_o e^{-\beta E(\theta)} + \sum_n A_n e^{-p_n t'} F_n(\theta) \tag{4.3}$$

where $F_n(\theta)$'s are eigenfunctions of D, satisfying

$$D F_n(\theta) = - p_n F_n(\theta) . \tag{4.4}$$

F_n's form a complete orthonormal set, with the following definition of the scalar product

$$(F_n, F_m) = \int_0^{\pi} F_n(\theta) F_m(\theta) e^{\beta E(\theta)} \sin\theta \, d\theta = \delta_{n,m} . \tag{4.5}$$

Following physical ideas of Kramers [5], Brown [1] argued that only the smallest eigenvalue p_1 (other than zero, of course) describes the long time evolution from the quasi-equilibrium state. The higher eigenvalues are important only in stablizing quasi-equilibrium in the early stages of relaxation. More precisely, we show in the appendix that $p_1 \sim 0(e^{-\beta K})$ while $p_i \sim 0(1)$ for $i \geqslant 2$.

To evaluate p_1 and F_1, Brown gave a variational treatment. Writing $F_1(\theta) = e^{-\beta E(\theta)} \Phi(\theta)$, one can easily show that the above eigenvalue problem is equivalent to the minimisation of the functional

$$D[\Phi] = \int_0^{\pi} e^{-\beta E} \left(\frac{d\Phi}{d\theta}\right)^2 \sin\theta \, d\theta \tag{4.6}$$

under the constraints

$$H[\Phi] = \int_0^{\pi} e^{-\beta E} \Phi^2 \sin\theta \, d\theta = 1 \tag{4.7}$$

and

$$(F_1, e^{-\beta E}) = \int_0^{\pi} e^{-\beta E} \, \Phi \, \text{Sin}\theta \, d\theta = 0 \tag{4.8}$$

The constraint (4.7) corresponds to the normalisation of eigenfunction, while (4.8) to its orthogonality to the lowest eigenfunction.

The choice of Φ is now dictated by the following requirements. (i) $\Phi(\theta)$ must change sign within the interval $0 \leqslant \theta \leqslant \pi$ to satisfy Eq.(4.8). (ii) To keep $D[\Phi]$ small, one should concentrate large values of $|d\Phi/d\theta|$ near the minima of $e^{-\beta K \text{Sin}^2\theta}$, i.e., $\theta \approx \pi/2$. Thus Brown allowed Φ to take constant values ϕ_1 in $(0,\theta_1)$ and ϕ_2 in (θ_2,π) and determined the derivative $d\Phi/d\theta$ in the region (θ_1,θ_2) from the minimisation of $D[\Phi]$. The form of the solution is shown schematically in Fig.3. Under the condition $\beta K \gg 1$, the constraints (4.7) and (4.8) can be evaluated by neglecting the constribution to the integrals from the region (θ_1,θ_2) where $e^{-\beta E}$ is very small. Thus the two constraints require

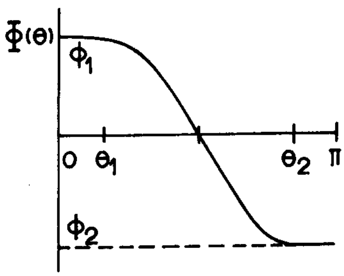

Fig. 3 Variational choice of $\Phi(\theta)$.

$$I_1\phi_1^2 + I_2\phi_2^2 = 1 \quad ; \quad I_1 \phi_1 + I_2 \phi_2 = 0 \tag{4.9}$$

where

$$I_{1,2} = \int_{R_1, R_2} e^{-\beta E(\theta)} \, \text{Sin}\theta \, d\theta \tag{4.10}$$

where R_1, R_2 denote respectively the integration regions $(0,\theta_1)$ and (θ_2,π). Now we minimise $D[\Phi]$ in (θ_1,θ_2) with the condition that $\Phi(\theta_1) = \phi_1$ and $\Phi(\theta_2) = \phi_2$. This yields

$$\frac{d}{d\theta} \left[e^{-\beta E} \frac{d\Phi}{d\theta} \text{Sin}\theta \right] = 0 \tag{4.11}$$

or

$$\frac{d\Phi}{d\theta} = \frac{A \; e^{\beta E}}{\text{Sin}\,\theta} \tag{4.12}$$

where A is an undetermined constant. Integration of Eq.(4.12) yields

$$\phi_1 - \phi_2 = A \int_{\theta_1}^{\theta_2} \frac{e^{\beta E}}{\text{Sin}\theta} d\theta = A \, I_m \tag{4.13}$$

where

$$I_m \simeq \sqrt{\frac{2\pi}{\beta E''(\pi/2)}} \; e^{\beta E(\pi/2)} \tag{4.14}$$

Equations (4.9), (4.10), (4.13) and (4.14) determine the solution. Using Eqs.(4.9), we get

$$\phi_1 = \frac{C}{I_1} \; ; \; \phi_2 = -\frac{C}{I_2} \quad ; \quad C = \sqrt{\frac{I_1 \, I_2}{I_1 + I_2}} \tag{4.15}$$

Substituting Eq.(4.15) in (4.13), one can find A as well. Finally, the eigenvalue p_1

is given as

$$p_1 = D[\Phi]_{min} = \frac{1}{I_m} \left(\frac{1}{I_1} + \frac{1}{I_2} \right) \qquad (4.16)$$

$$= \frac{4(\beta K v)^{3/2}}{\pi^{1/2}} e^{-\beta K v} \qquad (4.17)$$

where the second line follows by substituting the approximate values for the integrals when $V K \gg kT$. The variational solution for long times is

$$f(\theta,t) = e^{-\beta E(\theta)} \left[A_o + A_1 \Phi(\theta) e^{-p_1 t'} \right] \qquad (4.18)$$

From this solution, it is easy to derive the Neél picture. Let us calculate n_1 and n_2 as defined in (2.2). Using (4.18), one finds

$$n_{1,2} = I_{1,2} (A_o + A_1 \phi_{1,2} e^{-p_1 t'}) \qquad (4.19)$$

The constants A_o and A_1 are determined from the normalisation of the distribution function and the initial condition. However, here we shall eliminate them in favour of n_1, n_2 and their time derivatives. Using Eq.(4.10), one gets

$$n_1 + n_2 = A_o (I_1 + I_2) \qquad (4.20)$$

which shows that the total number of particles in the two potential valleys is a constant within this approximation. Further

$$\frac{dn_1}{dt} = - I_1 \phi_1 \frac{p_1}{2\tau_\perp} A_1 e^{-p_1 t'} = - \frac{dn_2}{dt} \qquad (4.21)$$

We can now use Eqn. (4.19) to eliminate $A_1 e^{-p_1 t'}$ in favour of n_1 and n_2. This yields

$$\frac{dn_1}{dt} = - \frac{dn_2}{dt} = - \nu_{12} n_1 + \nu_{21} n_2 \qquad (4.22)$$

with

$$\nu_{12} = \frac{1}{2\tau_\perp} \frac{p_1 \phi_1}{\phi_1 - \phi_2} = \frac{1}{2\tau_\perp} \frac{1}{I_1} \frac{1}{I_m} \simeq \frac{(\beta K v)^{3/2}}{\tau_\perp \pi^{1/2}} e^{-\beta K v} \qquad (4.23)$$

$$= \nu_{21} \qquad (4.24)$$

In the next section, we discuss the application of (4.22) for calculating the magnetic response of an assembly of single domain particles.

5. Non-equilibrium Response

In the large barrier limit, the magnetisation of the assembly is

$$M = v M_s [n_1(t) - n_2(t)] \qquad (5.1)$$

Using rate equations, it is a simple matter to see that

$$\frac{dM}{dt} = - \nu M \qquad (5.2)$$

where $\nu = (\nu_{12} + \nu_{21})$. Now suppose at time t = 0 we apply a field H along the anisotropy axis of the particles, assuming that anisotropy axes of all the particles are aligned. The rates in the presence of the field can again be calculated using the formalism of Sec.3. In the rate-equation description, however, the effect of the field can be easily determined using Arrhenius-like arguments. The energy of the n_1 particles is lowered by $+vM_sH$, so that the rate ν_{12} becomes ($M_sH \ll K$)

$$\nu_{12} = \frac{(v\beta K)^{3/2}}{\tau_\perp \pi^{1/2}} e^{-\beta v(K+M_sH)} \tag{5.3}$$

Similarly the energy of the n_2 particles is raised by vM_sH, and corresponding rate ν_{21} becomes

$$\nu_{21} = \frac{(v\beta K)^{3/2}}{\tau_\perp \pi^{1/2}} e^{-\beta v(K-M_sH)} \tag{5.4}$$

Substituting these rates in Eq.(4.22), it is an easy matter to show that

$$M(t) = M(o)\, e^{-\nu t} + vM_s(1-e^{-\nu t}) \tanh \frac{vM_sH}{kT} \tag{5.5}$$

where

$$\nu = \nu_{12} + \nu_{21} = 2 \frac{(v\beta K)^{3/2}}{\tau_\perp \pi^{1/2}} e^{-\beta K v} \operatorname{Cosh} \beta vM_sH \tag{5.6}$$

The susceptibility $\chi_\parallel$ along the easy axis is then

$$\chi_\parallel = \frac{(vM_s)^2}{kT} (1-e^{-\nu t}) \tag{5.7}$$

The situation considered above is somewhat artificial, because in most physical situations, the anisotropy axes of different particles are in different directions. So one really needs the response of the particles to a field in an arbitrary direction [9]. This response can be expressed in terms of the parallel response determined above and the response to a field perpendicular to the easy axis. If α is the angle between the easy axis and the field,

$$M = M_\parallel \operatorname{Cos}\alpha + M_\perp \operatorname{Sin}\alpha = (\chi_\parallel \operatorname{Cos}^2\alpha + \chi_\perp \operatorname{Sin}^2\alpha)\, H \tag{5.8}$$

Averaging over α then yields for susceptibility χ

$$\chi = 1/3\, [2\chi_\parallel + \chi_\perp] \tag{5.9}$$

The time dependent perpendicular susceptibility χ can be obtained by noting that the perpendicular field changes the energy of the particles according to the equation

$$E(\theta) = v\,[K\operatorname{Sin}^2\theta - M_sH \operatorname{Sin}\theta] \tag{5.10}$$

which shifts the energy minima to θ_1 and θ_2 given by

$$\theta_1 = \operatorname{Sin}^{-1} \frac{M_sH}{2K} \approx \frac{M_sH}{2K} = \pi - \theta_2 \; . \tag{5.11}$$

When the field is switched on at t=o, the response occurs in the following way. On time scales of order $\tau_\perp$, the quasi-equilibrium state is established, as the particles in the two valleys simply adjust to the new potential. This occurs quickly since no tunnelling across the barrier is involved. The contribution to $M_\perp$ is then

$$M_\perp = (n_1 \text{Sin}\theta_1 + n_2 \text{Sin}\theta_2)\, vM_s = \frac{vM_s^2}{2K}\, nH \tag{5.12}$$

Since $n = n_1 + n_2$ is a constant of motion in this approximation, the perpendicular response has only quick component. If we had considered an asymmetric barrier, in which case $\text{Sin}\theta_1 \neq \text{Sin}\theta_2$, the perpendicular response would also have a slow component. On time scales of interest here, we can put together all these results to write for susceptibility

$$\chi = \frac{N}{3}\left[2\,\frac{(vM_s)^2}{kT}\,(1-e^{-\nu t}) + \frac{vM_s^2}{2K}\,\theta(t)\right] \tag{5.13}$$

where $\theta(t)$ is the step function. From this, relaxational response it is easy to derive the equilibrium frequency dependent response via the use of the fluctuation-dissipation theorem [9]

$$\chi(\omega) = \frac{N}{3}\left[\frac{2(vM_s)^2}{kT}\;\frac{\nu}{\nu+i\omega} + \frac{vM_s^2}{2K}\right. \tag{5.14}$$

Appendix

In this appendix we show that in the large barrier limit only one eigenvalue of the Fokker-Planck equation (3.10) is of order $e^{-\beta K}$, while all others are of O(1). Thus only one eigenvalue is needed to describe the slow relaxation from the quasi-equilibrium state. A transparent proof of the statement can be made by first transforming Eq. (3.10) into a Schrödinger like equation (10). The transformation is

$$f(\theta,t) = e^{-\beta E(\theta\,/2}\, g(\theta,t) \tag{A.1}$$

Substitution in Eq. (3.10) leads to the following equation for $g(\theta,t)$

$$-\frac{\partial g}{\partial t'} = \left[-\frac{1}{\text{Sin}\theta}\,\frac{\partial}{\partial\theta}\left(\text{Sin}\theta\,\frac{\partial g}{\partial\theta}\right) + U(\theta)\, g\right] \tag{A.2}$$

where

$$U(\theta) = -\frac{1}{2}\,\frac{1}{\text{Sin}\theta}\,\frac{\partial}{\partial\theta}\left(\text{Sin}\theta\,\frac{\partial E}{\partial\theta}\right) + \frac{\beta^2}{4}\left(\frac{\partial E}{\partial\theta}\right)^2 \tag{A.3}$$

Substituting for E from Eq. (1.4), gives

$$U(\theta) = (\beta vK)^2\, \text{Sin}^2\theta\, \text{Cos}^2\theta - \beta vK(2\text{Cos}^2\theta - \text{Sin}^2\theta) \tag{A.4}$$

Writing $g(\theta,t) = e^{-pt}G(\theta)$, (A.2) can be reduced to the standard quantum mechanical eigenvalue pronlem

$$pG(\theta) = -\frac{1}{\text{Sin}\theta}\,\frac{\partial}{\partial\theta}\left(\text{Sin}\theta\frac{\partial G}{\partial\theta}\right) + U(\theta)G. \tag{A.5}$$

The potential $U(\theta)$ is plotted in Fig.4. As expected, the minimum potential regions are $\theta \approx 0$ and $\theta \approx \pi$. In the large barrier limit, as a first approximation the tunnelling between the two potential wells can be neglected, and one finds that the small eigenvalues p_i^o's are doubly degenerate, with their wavefunctions confined to either

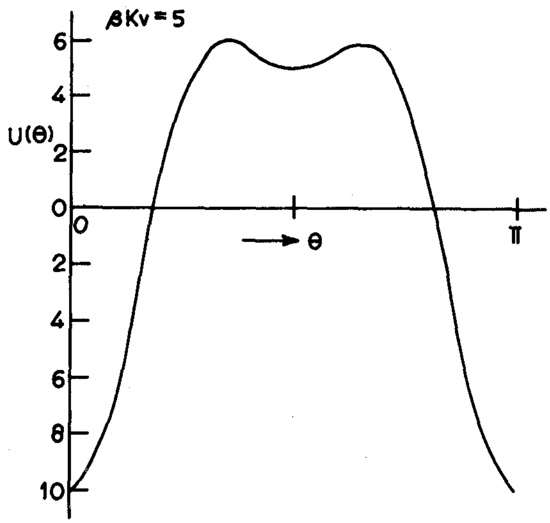

Fig.4 Plot of the effective potential $U(\theta)$

of the two potential wells. When the tunnelling is included the degeneracy of the levels is lifted and

$$p_i = p_i^o \pm t_i$$

where t_i's are the tunnelling amplitudes, which can be easily estimated in the WKB approximation to be

$$t_1 \approx e^{-\beta K v}$$

Thus the first two levels are split by an amount $e^{-\beta K v}$. But we know the lowest one to be zero, and thus $p_1 \approx e^{-\beta K v}$. The higher eigenvalues are of order unity.

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