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Relaxation behaviour of single-domain magnetic particles

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Abstract. The relaxation behaviour of single-domain magnetic particles is investigated. The first non-vanishing eigenvalue of an appropriate Fokker–Planck equation, characterising the dynamical response as well as the decay of the metastable states of the system, is calculated using a variational approach. The lower bound to the eigenvalue is also computed. The present treatment yields estimates for the eigenvalues that are better than those obtained by the Kramers method.

1. Introduction

The relaxation behaviour of single-domain magnetic particles has been receiving renewed attention in recent years in view of its close similarity with the non-equilibrium properties of spin glass alloys (see, for instance, Kumar and Dattagupta (1983) and the references cited therein). When the time for relaxation of the magnetisation of the particle from one easy orientation to another is comparable with the time of observation, one observes magnetic after-effects or magnetic viscosity, which are also characteristics of spin glasses. Essentially because of the simplicity of the theory, the data on spin glass freezing in a number of alloys have been analysed on the basis of thermal blocking of non-interacting magnetic particles. It is, therefore, of considerable interest to obtain a reliable estimate of the time τ_r for (or rate λ_r of) relaxation of a magnetic particle from one equilibrium orientation to another.

A related question is concerned with the general and important topic of the decay of metastable states. The orientation of a magnetic particle can be prepared in an initial state, say by the prior application of a magnetic field. The state becomes metastable if the field is removed, and its decay into a more stable state (magnetic after-effects) is obviously determined by λ_r introduced above. The computation of the rate of decay of metastable states is a fundamental problem in many other areas, and has been a subject of intense activity over the last four decades or so (see Kramers (1940), Landauer and Swanson (1961), Langer (1969), Haake (1979), Schuss (1980), Arecchi and Politi (1980); for a recent review, see Dattagupta and Shenoy (1983)). In this paper we present a variational estimate of λ_r in the context of the relaxation or approach to equilibrium of

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a single-domain magnetic particle. The method used, however, is of general utility in the theory of the decay of metastable states.

Our analysis here is based on a Fokker–Planck equation for the orientational distribution function of a magnetic particle in a potential that represents the combined effect of an anisotropy energy and an applied magnetic field. The treatment most often employed to calculate, for instance, the rate of decay of a metastable state, using an underlying Fokker–Planck equation, is due to Kramers (1940) and others (Landauer and Swanson 1961, Langer 1969). The Kramers method is based on a physical *ansatz* which, in the context of magnetic particles, should be valid when the 'barrier height' (proportional to the anisotropy energy) is much larger than the thermal energy (proportional to $\beta^{-1} = k_B T$; see, for example, Dattagupta and Shenoy (1983)). When this condition becomes invalid, the Kramers estimate yields poor results and one needs alternative approaches. One such treatment is due to Brown (1963) who, in his pioneering work on the relaxation behaviour of single-domain particles, introduced a variational analysis of the Fokker–Planck equation. However, the special choice that Brown made of the variational trial function yields results that are essentially equivalent to those obtained from the Kramers method. More recently, Schenzle and Brand (1979) had given several examples of trial functions for Fokker–Planck equations used in the context of optical bistability (Bonifacio and Lugiato 1978, Farina *et al* 1981). Brand *et al* (1982) also gave variational estimates for the lower bounds to the eigenvalues of the Fokker–Planck equation. Thus variational methods have the added advantage that they yield both upper and lower bounds to the eigenvalues which determine the time-dependent properties of the system concerned.

In the present paper we extend Brown's original work and show how a judiciously chosen trial function leads to a better estimate than Kramers' of the rate of relaxation λ_r. Using this new trial function we also obtain the lower bound to the first non-vanishing eigenvalue of the appropriate Fokker–Planck equation. This is an improvement over Brown's earlier work as his trial function is not twice differentiable and hence is not suitable for the computation of lower bounds (see § 2). The results presented here are of relevance in analysing the dynamic response, i.e. the AC susceptibility (Kumar and Dattagupta 1983) as well as the hysteresis behaviour of single-domain magnetic particles (for a general discussion, see Gilmore (1979) and Agarwal and Shenoy (1981)).

2. The general variational treatment

Consider a system with effectively a single degree of freedom. We assume that the time-dependent behaviour of the system is characterised by a Fokker–Planck equation for the probability distribution $P(\psi, t)$:

$$\partial P/\partial t = \partial(A(\psi)P)/\partial\psi + \partial^2(D(\psi)P)/\partial\psi^2. \tag{2.1}$$

Here ψ is the dynamical variable and $A(\psi)$ the drift coefficient which represents the effect of internal and external forces on the system. The diffusion coefficient $D(\psi)$ arising from, say, thermal fluctuations, can depend on ψ, in general. The steady state ($\dot{P} = 0$) solution P_s of equation (2.1) corresponding to natural boundary conditions is

$$P_s(\psi) = \eta \exp(-\Phi) \qquad \Phi(\psi) = \int^{\psi} d\psi' \frac{A(\psi') + D'(\psi')}{D(\psi')} \tag{2.2}$$

where η is a normalisation constant. Note that the 'potential' function $\Phi(\psi)$ may have several minima, and in particular, for a certain range of system parameters, may be bistable.

The time-dependent solution of equation (2.1) can be cast in the form

$$P(\psi, t) = \sum_n a_n \exp(-\lambda_n t) P_s(\psi) f_n(\psi) \tag{2.3}$$

where the a_n are the coefficients to be determined from the initial conditions, and the $f_n(\psi)$ and λ_n are to be evaluated from the eigenvalue equation

$$\frac{\partial}{\partial \psi}\left(D(\psi) P_s(\psi) \frac{\partial f_n(\psi)}{\partial \psi}\right) + \lambda_n P_s(\psi) f_n(\psi) = 0. \tag{2.4}$$

In order that the system may asymptotically ($t \to \infty$) reach a stationary state characterised by the probability distribution $P_s(\psi)$, the lowest eigenvalue λ_0 must vanish. In addition, of course, $f_0 = a_0 = 1$.

The eigenvalue problem (2.4) is self-adjoint and hence the eigenfunctions form an orthonormal set with weight factor P_s:

$$\int \mathrm{d}\psi\, P_s(\psi) f_n(\psi) f^*_m(\psi) = \delta_{nm}. \tag{2.5}$$

For dynamical calculations, we need to know the first few non-vanishing eigenvalues. In particular, if the eigenvalue spectrum is well separated the most important eigenvalue for dynamical considerations is the lowest non-vanishing eigenvalue λ_1 (cf Brand *et al* 1982). Hence, in what follows, we focus our attention on computing λ_1. Let $\chi(\psi)$ be the trial function; then the variational problem posed by equation (2.4) is formulated as

$$\lambda_1 \leqslant \left[\int \mathrm{d}\psi\, P_s(\psi) D(\psi) \left(\frac{\partial \chi}{\partial \psi}\right)^2\right]\left(\int \mathrm{d}\psi\, P_s(\psi) \chi^2(\psi)\right)^{-1} \tag{2.6}$$

$$N = \int \mathrm{d}\psi\, P_s(\psi) \chi^2(\psi) \tag{2.7}$$

$$0 = \int \mathrm{d}\psi\, P_s(\psi) \chi(\psi) \tag{2.8}$$

where the conditions in equations (2.7) and (2.8) imply orthonormality of the trial function (N is a constant).

The equation (2.6) yields an upper bound to the first non-vanishing eigenvalue. On the other hand, Brand *et al* (1982) have discussed how a lower bound to the eigenvalue of the Fokker–Planck operator can also be obtained by an extension of the results of Weinstein (1934) and Kamke (1939). For the self-adjoint problem $Lf = \lambda f$ and for functions f that are differentiable at least twice continuously, the lower bound to the eigenvalue λ_1 is given by

$$\lambda_1 \geqslant \alpha - (\beta^2 - \alpha^2)^{1/2} \equiv \alpha_1 \tag{2.9}$$

where

$$\alpha = \left(\int \mathrm{d}\psi f^*(\psi) L f(\psi)\right)\left(\int \mathrm{d}\psi |f|^2\right)^{-1} \tag{2.10}$$

$$\beta^2 = \left(\int \mathrm{d}\psi (L f^*(\psi))(L f(\psi))\right)\left(\int \mathrm{d}\psi |f|^2\right)^{-1}. \tag{2.11}$$

For the eigenvalue problem (2.4), one finds

$$\alpha = \frac{1}{N}\int \mathrm{d}\psi\, P_s(\psi) D(\psi) \left(\frac{\partial \chi}{\partial \psi}\right)^2 \tag{2.12}$$

$$\beta^2 = \frac{1}{N} \int \mathrm{d}\psi P_s^{-1}(\psi) \left| \frac{\partial}{\partial \psi} \left(D(\psi) P_s(\psi) \frac{\partial \chi}{\partial \psi} \right) \right|^2 . \tag{2.13}$$

In the next section the above formulae will be applied to the study of the reorientational motion (or the rotational diffusion) of single-domain magnetic particles.

3. The lowest non-vanishing eigenvalue for rotational diffusion

Here we shall obtain a variational estimate for λ_1 using a suitable trial function for the Fokker–Planck equation that describes the rotational diffusion of magnetic particles. Restricting the discussion to the experimentally interesting case of uniaxial anisotropy, the magnetic energy of a single-domain particle can be written as

$$V(\Theta) = K \sin^2 \Theta - HM_s \cos \Theta \tag{3.1}$$

where K is the anisotropy parameter, H the applied field and M_s is the saturation magnetisation of the particle. Both K and M_s are linearly proportional to the volume v of the particle. The orientational distribution function $W(\Theta, t)$ then obeys the Fokker–Planck equation in terms of the dimensionless time τ (Brown 1963):

$$\frac{\partial W}{\partial \tau} = \frac{1}{\sin \Theta} \frac{\partial}{\partial \Theta} \left[\sin \Theta \left(\beta \frac{\partial V}{\partial \Theta} W + \frac{\partial W}{\partial \Theta} \right) \right] \qquad 0 \leqslant \Theta \leqslant \pi \tag{3.2}$$

where $\beta = (k_B T)^{-1}$ and

$$\int_0^\pi W(\Theta, t) \sin \Theta \, \mathrm{d}\Theta = 1.$$

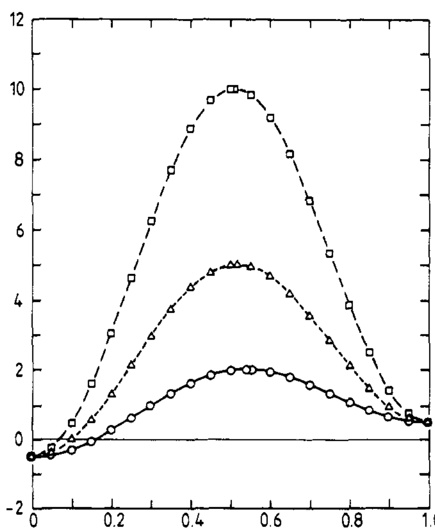

Figure 1. The potential function $\Phi(\Theta) = \gamma \sin^2 \Theta - y \cos \Theta$ as a function of Θ/π for three values of γ and for $y = 0.5$.

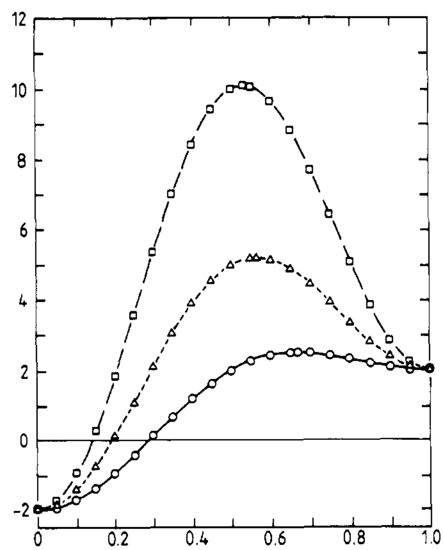

Figure 2. The same as figure 1 except that $y = 2.0$.

In terms of the variable $\psi = \cos\Theta$, equation (3.2) becomes ($\sin\Theta\, W(\Theta)\,\mathrm{d}\Theta = P(\psi)\,\mathrm{d}\psi$):

$$\frac{\partial P}{\partial \tau} = \frac{\partial}{\partial \psi}\left[(1-\psi^2)\left(\beta\frac{\partial V}{\partial \psi}P + \frac{\partial P}{\partial \psi}\right)\right]. \tag{3.3}$$

The equation (3.3) is of the general structure given in equation (2.1) with the 'diffusion coefficient' $D(\psi) = (1-\psi^2)$. The steady-state solution is expressed in equation (2.2) where the potential has the form

$$\Phi(\psi) \equiv \beta V(\psi) = \gamma(1-\psi^2) - y\psi$$
$$\gamma \equiv \beta K \qquad y \equiv \beta H M_s. \tag{3.4}$$

The potential Φ is bistable and is asymmetric (for $y \neq 0$), as illustrated in figures 1 and 2 for a set of values of γ and y. As the external field strength y increases, the potential becomes more and more asymmetric.

We now make a suitable choice of the trial function χ in order to estimate λ_1. As argued by Brown, the function χ should be such that (i) it changes sign in the interval $-1 \leqslant \psi \leqslant 1$ in order to satisfy equation (2.8) and (ii) its derivative must be small in the neighbourhood of the maxima of P_s and peaked around the minimum of P_s, in order to keep the upper bound small (cf equation (2.6)). In addition, the trial function must be twice differentiable in the interval in question. As dictated by these considerations, we propose the following form:

$$\chi(\psi) = \chi_1\{1 + \exp[-a(\psi_3 - \psi)]\}^{-1} + \chi_2\{1 + \exp[-a(\psi - \psi_3]\}^{-1} \tag{3.5}$$

where the variational parameter a is 'suitably' large (see below), and ψ_3 is the value of ψ at which P_s is a minimum (or Φ is a maximum). Evidently,

$$\psi_3 = -y/2\gamma. \tag{3.6}$$

The constants χ_1 and χ_2 may be determined from equations (2.7) and (2.8). It may be noted that $\chi \rightarrow \chi_1\,(\chi_2)$ as $\psi - \psi_3 \ll 0\ (\gg 0)$. The trial function in (3.5) is similar in form to the one used by Schenzle and Brand (1979) in the context of optical bistability.

The first derivative of χ, required for evaluating the various bounds given in § 2, is given by

$$(\mathrm{d}\chi/\mathrm{d}\psi) = (a/2)(\chi_2 - \chi_1)(1 + \cosh a(\psi_3 - \psi))^{-1}. \tag{3.7}$$

It is evident that as the quantity a increases, $\mathrm{d}\chi/\mathrm{d}\psi$ becomes more and more peaked around ψ_3; at the same time, χ retains its constant value $\chi_1\,(\chi_2)$ for a wider range of $\psi < \psi_3\,(>\psi_3)$ (see (3.5)). Thus as a becomes very large, our choice of the trial function in (3.5) becomes equivalent to that of Brown (1963). Since the Brown *ansatz* is deemed to be accurate when the barrier height is much greater than the thermal energy (i.e. when γ is very large), it is expected that a should be somehow related to γ. This, plus dimensional reasoning (cf the exponents in (3.5)) leads us to express a in the form

$$a^2 = \xi\,|-\Phi''(\Theta_3)| \tag{3.8}$$

where $\Phi''(\Theta_3)$ is the curvature of the potential at its maximum ($\Theta_3 = \cos^{-1}\psi_3$), and ξ is now the new variational parameter. From equations (3.4) and (3.6), we have

$$a^2 = 2\xi\gamma(1 - y^2/4\gamma^2). \tag{3.9}$$

The variational problem formulated in equations (2.6)–(2.8) is now solved numerically,

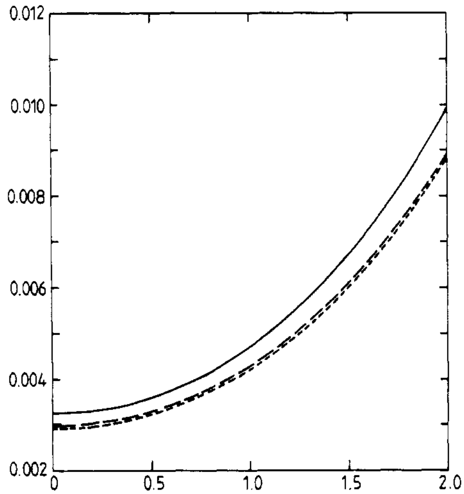

Figure 3. The upper bound to the eigenvalue λ_1 as a function of the field strength y and for the anisotropy parameter $\gamma = 10.0$. Kramers' estimate (3.10) (——); modified Kramers' estimate (3.11) (— — —); and variational estimate deduced from the minimisation of (2.6) (- - - -).

and the results are shown in figures 3–5. For each set of barrier parameters (i.e. γ and y), a distinct value of ξ is computed which minimises the right-hand side of (2.6). The corresponding optimised trial function χ is then employed in calculating β^2 from (2.13) and hence the lower bound to the eigenvalue α_1 from (2.9). Note that the lower bound makes sense only if $\alpha_1 > 0$ since the eigenvalue λ_1 has to be positive.

We also compare the present results with those obtained from the Kramers treatment. The latter yields (Brown 1963)

$$\lambda = (\gamma/\pi)^{1/2}(1 - y^2/4\gamma^2) \exp[-(\gamma + y^2/4\gamma)] \times [(2\gamma + y) \exp(-y) + (2\gamma - y) \exp(-y)]. \tag{3.10}$$

It is clear from figures 3–5 that the trial function in equation (3.5) leads to a significant improvement over the Kramers estimate. Also, as expected, our results match asymptotically with those of Kramers, as γ becomes large. Now, in deriving (3.10), certain integrals are evaluated approximately by using steepest-descent arguments, which are valid when γ is large (see Brown 1963). On the other hand, we find that a somewhat better result follows if the integrals are left as such and evaluated numerically. Thus the original Kramers method would yield for the lowest non-vanishing eigenvalue (cf Dattagupta and Shenoy 1983)

$$\nu = (1/I_3)(1/I_1 + 1/I_2) \tag{3.11}$$

where

$$I_1 = \int_0^{\Theta_3} \sin\Theta \, d\Theta \exp(-\Phi(\Theta))$$

$$I_2 = \int_{\Theta_3}^{\pi} \sin\Theta \, d\Theta \exp(-\Phi(\Theta))$$

$$I_3 \simeq 2\gamma(4\gamma^2 - y^2)^{1/2} \int_0^{\pi} d\Theta \exp(\Phi(\Theta)). \tag{3.12}$$

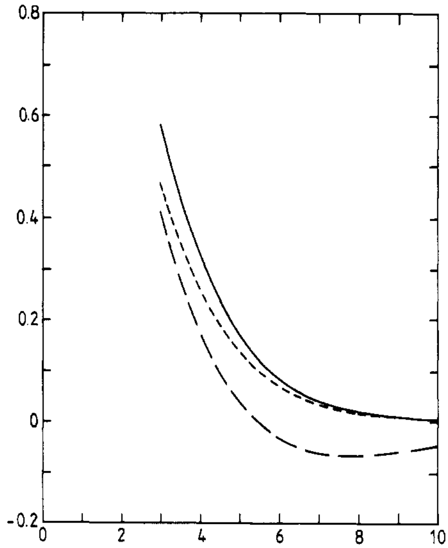

Figure 4. The variational lower bound (— —), upper bound (- - - -) and the Kramers (——) estimates for λ_1 as a function of the anisotropy parameter γ and for $y = 0$.

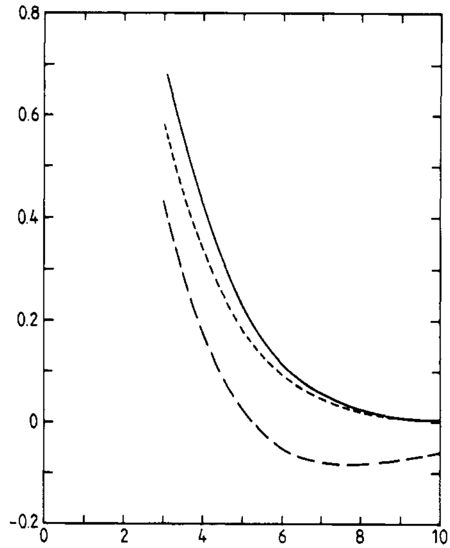

Figure 5. The same as figure 4 except that $y = 1.0$.

Needless to say, the limiting form of ν for γ large, obtained by calculating approximately the integrals in (3.12), would lead to the expression for λ given in (3.10). Interestingly, the estimate for ν shown in figure 3 is found to be rather close to our variational result.

4. Conclusions

In summary we have shown how a suitably chosen variational trial function can be employed to evaluate the upper and lower bounds to the first non-vanishing eigenvalue of the Fokker–Planck equation which describes the relaxation behaviour of magnetic particles. These estimates for the eigenvalue are found to be much more reliable than those obtained from the widely used Kramers method. Among other applications of the present treatment we might mention the AC susceptibility response of a single-domain magnetic particle to an oscillatory magnetic field. The susceptibility as a function of the frequency of the applied field is usually characterised by a Debye peak whose width is proportional to the first non-vanishing eigenvalue of the Fokker–Planck equation computed here. Since the present estimate of the eigenvalue is lower than that based on the Kramers method, the width is expected to be narrower than that calculated earlier (Kumar and Dattagupta 1983). The trial function of this paper is also expected to yield reasonable results in the context of diffusion in other bistable potentials (Agarwal *et al* 1984).

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