Quantum Relaxation of Magnetisation in Magnetic Particles

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At temperatures below the magnetic anisotropy energy, monodomain magnetic systems (small particles, nanomagnetic devices, etc.) must relax quantum mechanically - thermal activation is ineffective. The discrete nature of the spectrum is important. This quantum relaxation must be mediated by the coupling to both nuclear spins and phonons (and electrons if either particle or substrate is conducting).

We analyze the effect of each of these couplings, and then combine them. Conducting systems can be modelled by a "giant Kondo" Hamiltonian, with nuclear spins added in as well. At low temperatures, even microscopic particles on a conducting substrate (containing only 10\(^{-50}\) spins) will have their magnetisation frozen over millenia by a combination of electronic dissipation and the "degeneracy blocking" caused by nuclear spins. Raising the temperature leads to a sudden unblocking of the spin dynamics at a well defined temperature.

Insulating systems are quite different. The relaxation is strongly enhanced by the coupling to nuclear spins. At short times the magnetisation of an ensemble of particles relaxes logarithmically in time, after an initial very fast decay - this relaxation proceeds entirely via the nuclear spins. At longer times phonons take over, but the decay rate is still governed by the temperature-dependent nuclear bias field acting on the particles - decay may be exponential or power-law depending on the temperature.

The most surprising feature of the results is the pivotal role played by the nuclear spins. The results are relevant to any experiments on magnetic particles in which interparticle dipolar interactions are unimportant. They are also relevant to future magnetic device technology.

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I. INTRODUCTION

A. Quantum Relaxation

One of the most thoroughly explored subjects in all of science is that of thermal relaxation of magnetisation, in magnetic systems of all shapes and sizes. Investigations in this area (which go back many centuries \cite{1}) have revealed many subtleties, and even today there are many unsolved puzzles (e.g., the physics of "magnetic avalanches").

Very recently a whole new set of questions in this area has arisen, with the advent of well-characterised "nanoscopic" magnetic structures \cite{2}. Such structures include "made-to-order" magnetic grains, magnetic wires and superlattice arrays, as well as thin films and spin chains. There are very obvious applications of such nanomagnets in, e.g., the recording industry, as well as in information transmission and computing. Theoretical activity has been particularly intense on 1-dimensional spin systems \cite{3}, and on the tunneling and coherence phenomenon which should exist in both grains (involving "giant spin" \cite{4,5} dynamics) and in the dynamics of domain walls \cite{6}. Both the theory and the experimental activity in the latter field have been reviewed recently \cite{7,8,9}.

In this paper we focus on something rather different - the "quantum relaxation" of magnetisation that must occur in any small magnetic particle, once thermally-activated relaxation has ceased. Although there does not seem to have been a serious theoretical analysis of such relaxation before, the conventional view has been that it must proceed via a simple tunneling process (dissipative or otherwise). The picture we shall derive here is more subtle, and contains some rather fascinating new physics. Tunneling is certainly involved; but we find that what really controls relaxation is the nuclear spin system inside the magnetic particle, often with the help of phonons or electrons. One can put the result in the following way: the nuclear spins provide the essential "switch" that turns on (or turns off) the quantum transitions. As far as we are aware, there has been no previous recognition that nuclei had any role whatsoever to play in magnetic relaxation, despite the enormous number of experiments in this area. Nor do we know of any theoretical analysis of the influence of electrons on magnetic relaxation. There has certainly been no attempt at a theory dealing...
with the combined effects of nuclear spins, phonons, and electrons, which is what we do here. As we shall see, it is impossible to give a realistic analysis without putting them all together.

We shall not try here to address experiments in any detail, for two reasons. First, each different experimental system has its own idiosyncrasies, and we shall see that our theory has many different parameters - it is simply not possible to give detailed results for all possible cases, and we feel that it would be preferable to examine particular systems for which parameters are well known, as test cases. This we intend to do elsewhere. Second, in most experiments on magnetic grains, the grains are rather close together. As we shall briefly explain in this paper, this makes it probable that even the low-T relaxation of an experimental ensemble of grains proceeds via dipolar interactions between the grains.

The theory in this paper is closely related to previous work on quantum coherence in magnetic grains \cite{10}. In that work a very detailed analysis was given of the way nuclear spins control the dynamics of "giant spins", i.e., spins with quantum number $S \gg 1$. We also analysed the effect of phonons and electrons in "decohering" the quantum motion of $\vec{S}$. The problem of quantum relaxation is different, in that even if coherence is destroyed, relaxation may still proceed incoherently. What we shall find is that in the absence of nuclear spins, phonons cannot relax $\vec{S}$ in a small biasing field, because they couple too weakly to it; on the other hand electrons couple so strongly to $\vec{S}$ that they freeze it completely. The nuclear spins can then liberate $\vec{S}$ - amusingly, the mechanisms by which they do this are the same as those by which they destroy the quantum coherent motion of $\vec{S}$. The bias field $\epsilon$ acting on $\vec{S}$, generated by the combined hyperfine fields of all the nuclei can easily be greater than $10K$ (e.g., for TbFe$_3$ grains), and depending on the size of the grain may be equivalent to an external field from $100G$ up to well over $1T$. This bias changes with time in a diffusive way, at a rate governed by inter-nuclear dipolar interactions (in the absence of phonons and electrons), and also nuclear spin-phonon or spin-electron interactions, when the latter are present. Every so often, as $\epsilon$ shunts around, it brings quantum 2 states corresponding to different orientations of $\vec{S}$ into near degeneracy. At this point the system can tunnel, provided (a) the nuclear bias field $\epsilon(t)$ gives it time to do so coherently, (b) the usual dissipative effects of the electrons and phonons are not too destructive.

On the other hand, we shall also see that the nuclear spins can also further inhibit the relaxation of $\vec{S}$, at low $T$. If the nuclear spins are allowed to come to equilibrium, then at temperature below the hyperfine coupling energy, they exert a large negative bias field on $\vec{S}$, which essentially traps $\vec{S}$ for astronomical times. Thus, depending on $T$ and on the nuclear polarisation, the nuclei can enhance or suppress relaxation.

Although our purpose in this paper is not to deal with the implications for magnetic technology involving quantum relaxation, we feel we should at least make some remarks here about what they might be. Fig.1 shows the way in which magnetic computer memory elements have decreased exponentially in size in the last 40 years - such plots have figured large in recent discussions of the need for new kinds of computer design, incorporating elements which operate quantum-mechanically. At present computers, as well as magnetic tapes, use elements which behave classically, and are stable over long periods of time. This stability over decades exists because they are big - the energy barrier between 2 states of the element, proportional to its size, usually exceeds $100k_BT$ even at room temperature. The hypothesized "quantum threshold", below which tunneling is important even at $T = 0$, is usually supposed to be for grains containing roughly $10^2 - 10^4$ spins, depending on the material involved. However we shall find that the threshold between stable behaviour and quantum relaxation can be moved around a lot, depending on how the magnetic system is coupled to the nuclear spins, and whether or not it is coupled to electrons. We shall see that in some cases, one can freeze the dynamics of even microscopic magnetic systems (containing only 10 spins), for times of thousands of years or more. Conversely, we shall see how it is possible for much larger systems (containing $10^5$ electronic spins or more) to relax very quickly (in $\mu$s). These theoretical insights may have some relevance to the design of future nanoscopic magnetic devices.

In what follows we shall analyse the role of electrons (Section II), phonons (Section III), and nuclei (Section IV), acting in isolation on $\vec{S}$. Then in Section V we shall put them together, to give the final rather complex picture. As noted above, we save detailed discussion of experiments for other papers; but in Section VI we will indicate the general nature of our predictions.

**B. The Model, and Energy Scales**

We briefly describe the giant spin model here, and the various energy scales relevant to the physics. The basic idea behind this model has been explained in detail elsewhere \cite{10}. The exchange couplings $J_{ij}$ between electronic spins in the grain are enormous compared to the anisotropy couplings $K_\alpha$ (typically $J_{ij} \sim eV$, whereas $K_\alpha \sim 10^{-2} - 1K$). The mesoscopic grains can be treated as giant rotators, keeping only states $|\vec{S},m\rangle$, with $S \geq m \geq -S$. The spectrum
of this giant spin is shown in Fig.1 in a small bias field $\xi_H$. Sometimes, as a model example, we shall use the easy-axis/easy-plane system, having Hamiltonian

$$H_o(\vec{S}) = \frac{1}{\mathcal{S}} \left[ -K_\parallel S^2_\parallel + K_\perp S^2_\perp - \gamma \vec{S} \cdot \vec{H}_o \right]. \quad (1.1)$$

We will usually assume $\vec{H}_o = \gamma H_o$, so $\xi_H = \gamma S H_o$.

The low energy physics of (1.1) can be understood in terms of the truncated Hamiltonian

$$H_o(\vec{\tau}) = (2\Delta_o \hat{\tau}_x \cos \pi S - \xi_H \hat{\tau}_z) \equiv \Delta_o \hat{\tau}_x - \xi_H \hat{\tau}_z, \quad (1.2)$$

where the tunneling splitting $\Delta_o$ is

$$\Delta_o \sim \Omega_o e^{-A_o}, \quad (1.3)$$

and the bare parameters are

$$\Omega_o \sim 2(K_\parallel K_\perp)^{1/2}, \quad (1.4)$$

$$A_o \sim 2S(K_\parallel/K_\perp)^{1/2}, \quad (1.5)$$

The "bounce frequency" $\Omega_o$ is also roughly the distance to the next pair of excited levels; the total barrier height in (1.1), between the 2 semiclassical minima at $\vec{S} = \pm \xi S$, is $SK_\parallel$. Thus if, say, $S = 10^4$, we might easily have $\Omega_o \sim 0.1 - 1 K$, a barrier height $\sim 0.01 - 0.1 eV$, but a splitting $\Delta_o \sim 10^{-6} K$, or less.

This is certainly not the whole story. Magnetic grains having more complicated symmetries are not so easily described using WKB/instanton methods. The "internal" magnon modes of the giant spin are also neglected, except for those uniform precession modes corresponding to $| S, m \rangle$. There will also be phonons (spectrally very weak at these low energies) and possibly electrons (which will mix strongly with all states of the grain).

Finally and most importantly, the nuclei in the grain (and also possibly outside) will couple via hyperfine interactions to each level shown in Fig.2. The net result of this is shown in Fig.3 for the 2 lowest levels in the presence of a bias field. Typically $\Omega_o \gg \omega_k \gg \Delta_o$, where $\omega_k$ is an individual hyperfine coupling; however $N$ nuclei will spread out a single grain level into a mass of $2^N$ levels, of Gaussian half-width $\sim N^{1/2} \omega_o$, where $\omega_o$ is the principal hyperfine splitting. For rare earths, where $\omega_o \sim 1 GHz$ or greater, this half-width can easily exceed $\Omega_o$ for mesoscopic grains, and the whole "giant spin" model begins to get rather complex.

In this paper we will ignore all physics at energies $\sim \Omega_o$ or greater, simply including it into renormalisation parameters of a low-$T$ effective Hamiltonian. For such a treatment to be valid, we then require

$$k_B T, \xi_H, N^{1/2} \omega_o \ll \Omega_o. \quad (1.6)$$

We will also assume that any "loose spins" on the surface of the grain still have couplings $\gg \Omega_o$ to $\vec{S}$. If not, they will be treated as part of the "spin bath" environment, along with nuclei and paramagnetic impurity spins.

Before beginning, it is useful to bear in mind 2 simple points. Note first that in the presence of any bias $\xi_H$ which is greater than $\Delta_\perp$ in (1.2), the system is essentially "frozen" in one or other of the states $|\uparrow\rangle$ or $|\downarrow\rangle$. Suppose, e.g., the system starts in the higher-energy state $|\downarrow\rangle$ at $t = 0$. Then the probability $P^{(0)}(t, \xi_H)$ of finding it $|\downarrow\rangle$ at time $t$, in the absence of any other couplings, is given by

$$P^{(0)}(t, \xi_H) = 1 - \frac{\Delta^2}{E^2} \sin^2 E t, \quad (1.7)$$

where $E^2 = \xi_H^2 + \Delta_\perp^2$. Thus the coupling to phonons, electrons, and nuclei is necessary if we are to have any relaxation at all. Usually one thinks of this as a kind of spontaneous or stimulated emission, perhaps in the presence of dissipation, in which the requisite energy is taken up by a bath of oscillators.

In fact this conventional picture of tunneling of a biased 2-level system, coupled dissipatively to a bath of oscillators [12], works reasonably well for a giant spin coupled to either electrons or phonons. This we shall see in detail in the next two sections. However as soon as we introduce the spin bath of nuclei, etc., it breaks down completely; this is basically why our final results are so surprising.

The second point arises from the finite-dimensional nature of the giant spin Hilbert space. As emphasized by van Hemmen and Suto [2-3] this means that we must use WKB calculations of tunneling rates with great care when

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discussing the motion of $\vec{S}$. This point is especially clear in recent work of Politi et al. [14] (see also Villain et al. [14]), analysing the Mn$_{12}$O$_{12}$ acetate systems; here tunneling appears to proceed via a 4th-order term $\sim (\vec{S}_+^4 + \vec{S}_-^4)$ in $H_o(\vec{S})$. Consequently transitions may only proceed between states differing by $\Delta S_z = \pm 4$. To correctly handle such selection rules in the instanton calculus is not a trivial problem. Even more important, there is no continuum of final states in this tunneling problem, and tunneling may only proceed if there is near resonance between initial and final states. This means that in the presence of a bath of, e.g., phonons, one may not in general simply apply the Caldeira-Leggett tunneling formalism [15] to calculate tunneling, since this formalism assumes a continuum of final states. For this reason calculations such as those of Garg and Kim [16], of the tunneling rate of a strongly biased grain using the Caldeira-Leggett tunneling formalism [15] to calculate tunneling, since this formalism assumes a continuum of final states. This is unnecessary once the level spacing is greater than the (very small) linewidth caused by phonon damping (note further that calculations of grain tunneling in the presence of a nuclear spin bath should never use the Caldeira-Leggett method, since the spin bath cannot be mapped onto an oscillator bath of the Feynman-Vernon $\Gamma$ type - thus calculations of this kind [15] are not valid for any $S$).

We now turn to the detailed treatment of the different relaxation mechanisms. Readers wishing to see only a summary of the results should proceed directly to section IV.

II. GIANT SPINS AND THE ELECTRON BATH

In this section we develop a model for the coupling between the giant spin and the electron bath only, and use it to calculate relaxation rates for the grain magnetisation. Since, as far as we are aware, no attempt has ever been made to deal with the coupled giant spin/electron system before, it is clear that we must begin from first principles. The full development of our model, including detailed discussion of the multiple-scattering of the electrons by the individual spins inside the grain, the effects due to surface spins, to the discreteness of the electronic spectrum inside the grain if it is on an insulating substrate, the interaction between electrons and the internal degrees of freedom (magnons) in the grain, etc., will not appear in this paper. We shall also set aside questions about how conduction electron dynamics outside the grain, in a conducting substrate, will be influenced by the giant spin dynamics. This is a very important question for any experiment (which can use the substrate magnetoresistance as a probe of the grain dynamics), but is too complicated to deal with here.

There are 3 physical situations which we will address here. The first involves conducting electrons both inside and outside the grain; we shall see that this problem can be analysed fairly clearly. Likewise the problem of an insulating grain interacting with conducting substrate can be modelled in a reasonably treatable way. The problem of a conducting grain and an insulating substrate is more messy, however we can fairly easily extract the essential results. Our essential result is that unless the substrate is insulating, electrons simply freeze the grain if it is on an insulating substrate, the interaction between electrons and the internal degrees of freedom which permeates freely through the boundary between grain and substrate; i.e., a Hamiltonian

$$H = H_o(\vec{S}) + \sum_{\vec{k},\sigma} e_{\vec{k}\sigma}^c c_{\vec{k}\sigma}^c + \frac{1}{2} \sum_{i \in V_o} J_i \vec{s}_i \cdot \vec{\sigma} + \sum_{\vec{k}} \sum_{\vec{q}} e^{i\vec{q} \cdot \vec{r}_i} c_{\vec{k}+\vec{q}\sigma}^c c_{\vec{k}\sigma}^c ,$$

(2.1)

describing short-range exchange interaction between the localised individual spins $\vec{s}_i$, located at positions $r_i$ inside the grain, and conduction electrons in momentum $| \vec{k} \rangle$. A more refined model would couple grain electrons in eigenstates $| \mu \rangle$ to substrate electrons $| \vec{k} \rangle$, via a transfer matrix $\hat{T}_{\mu \vec{k}}$ across the boundary. However it is intuitively obvious that this is unnecessary once $| \hat{T}_{\mu \vec{k}} | > \Delta \epsilon_\mu$, where $\Delta \epsilon_\mu$ is the typical level spacing between internal electron states of the grain. If the electronic bandwidth is $D$, then $\Delta \epsilon_\mu \sim D/S$, and for most metals $D \sim 1 - 5 \text{ eV}$; thus if $S \sim 10^5$, $\Delta \epsilon_\mu \sim 10 - 100 K$, so that an insulating layer at least $10 \text{ Å}$ thick would be required to make such a model necessary.

We assume in using (2.1) that the internal grain spins are locked together to form a giant spin, and that the $J_i$ already take account of the virtual mixing between internal excited states of the magnetic ions and of the giant spin (magnons, surface modes) and the electron bath. Thus, as usual, we shall be working at low $T$ and small $\tilde{H}_o$. However we shall assume that the $J_i$ can vary; near the surface of the grain boundary they may be a little weaker. The present
model does not include "loose spins", i.e., surface spins which are more weakly coupled to the motion of \( \vec{S} \) (the reader should not confuse the Kondo coupling \( J_i \) here with the inter-spin exchange \( J_{ij} \) between spins inside the grain). These will be discussed in section \( \text{III} \) onwards. Typically the Kondo couplings \( J_i \sim 0.1 - 1 \, \text{eV} \).

We now rewrite (2.2), incorporating the giant spin hypothesis, to get a volume averaged interaction

\[
H_{\text{int}}^b \to \frac{1}{2} \vec{J} \vec{S} \cdot \sigma \sum_{\vec{q}} \sum_{\alpha} F_{\vec{q}} \hat{c}^\dagger_{\vec{q}+\vec{q}_0} c_{\vec{q}_0,\alpha} ;
\]

(2.2)

where the form factor \( F_{\vec{q}} \) integrates the number density \( \rho(\vec{r}) \) of microscopic spins over the grain volume, and \( \vec{J} \) is the mean value of the \( J_i \). \( H_{\text{int}}^b \) is a sort of "giant spin Kondo Hamiltonian", and as such contains a lot of interesting physics, much of which we will ignore in this paper. Recall that the Kondo coupling for microscopic spins is just \( J_{\alpha} \) with \( \rho(\vec{r}) \) in (2.3) restricted to a single lattice cell; for this problem one conventionally defines a dimensionless coupling \( g = \vec{J} N(0) \sim 0.1 \) for most metals, where \( N(0) \sim \rho/D \) is the Fermi surface density of states. The Kondo problem can be mapped onto an Ohmic spin-boson problem of the kind discussed by Leggett et al. \[12\]; even for this spin-1/2 system, there is a large dimensionless Ohmic coupling to the electron bath; \( \alpha_K \sim O(1) \).

It is possible to start with (2.4) and build up an analogous description by considering the individual Kondo scattering from each spin, in many different orbital channels. Here we shall finesse entirely this move by instead integrating out the electrons directly, starting from (2.2). We first truncate directly to a biased spin-boson Hamiltonian of form (for \( H_o(\vec{S}) = H_o^b(\vec{S}) \) in (1.1)):

\[
H_{\text{eff}}^b = \Delta_o \hat{x} - \epsilon \hat{z} + \frac{1}{2} \sum_{k=1}^N m_k (\hat{x}_k^2 + \omega_k^2 \hat{z}_k^2) + \hat{z} \sum_{k=1}^N c_k x_k ,
\]

(2.4)

where the bias field is parallel to the easy axis (we shall ignore transverse fields here). The Caldeira-Leggett spectral function for this problem is

\[
J_b(\omega) = \frac{\pi}{2} \sum_{k=1}^N \frac{|c_k|^2}{m_k \omega_k} \equiv \pi \alpha_b \omega ,
\]

(2.5)

and the Ohmic coupling constant \( \alpha_b \) is determined by a standard Fermi surface average \[19,20\] over electrons, i.e.,

\[
\alpha_b = 2g^2 S^2 \langle | F_{\vec{q}-\vec{r}} |^2 \rangle_{F.S.} .
\]

(2.6)

The naive guess that \( \alpha_b \sim S^2 \), apparently confirmed by (2.6), is however wrong; carrying out the average, one gets

\[
\alpha_b = 2g^2 S^2 \int G_{\vec{r},\vec{r}'} \left( \frac{\sin k_F | \vec{r} - \vec{r}' |}{k_F | \vec{r} - \vec{r}' |} \right)^2 \sim \left( \frac{g S}{k_F R_o} \right)^2 \sim g^2 S^{4/3},
\]

(2.7)

This at first surprising result is a consequence of interference between Kondo scattering at different sites - notice that \( k_F^2 \ll R_o \), so that the problem is a multiple scattering one. A related point is that the estimate of \( \alpha_b \) in (2.4) looks perturbative - it is not clear it should work for large \( \alpha_b \). However for a grain of radius \( R_o \) some \( \alpha_{\text{max}} \sim 1/(k_F R_o)^2 \sim S^{2/3} \) orbital channels are playing role in electron scattering. If \( S \leq 10^3 \), the phase shifts in each channel are small (one has \( \delta_k \sim g k_F R_o \sim g S^{1/3} \)), making it possible to apply a perturbation theory, yet the sum over all the channels can be rather large. The case of large phase shifts is more complicated, and may result in a giant spin dynamics different from that described by the standard Ohmic model. We address this possibility in Appendix A.

For mesoscopic spins (\( S \geq 10^3 \)) the coupling in (2.7) is very large; we may take over standard results from the spin-boson problem to discuss its effect on the dynamics of \( \vec{S} \). Since \( \alpha_b > 1 \), coherence at zero bias is destroyed at any temperature; at \( T = 0 \) the grain dynamics are rigidly locked, and the spin \( \vec{S} \) cannot move. At finite \( T \) and \( \epsilon = 0 \), the system fluctuates incoherently between the states \( | \uparrow \rangle + | \downarrow \rangle \) at a rate \( (T/\Omega_o)^{2\alpha_b-1} \); on the other hand if \( \epsilon \gg T \), one has incoherent relaxation at a rate \( (\epsilon/\Omega_o)^{2\alpha_b-1} \). The general result for the relaxation rate is \[12,20\]
\[
\tau_c^{-1} = 2\Delta_o \left( \frac{\Delta_o}{\Omega_o} \right)^2 \left( \frac{2\pi T}{\Omega_o} \right)^{2\alpha_b-1} \cosh \left( \frac{\epsilon}{2T} \right) \left| \frac{\Gamma[\alpha_b + i\epsilon/2\pi T]}{\Gamma[2\alpha_b]} \right|^2,
\]

where \(\Gamma[z]\) is the Gamma function; for \(\epsilon = 0\), this gives

\[
\tau_c^{-1}(T) = \Delta_o \left( \frac{\Delta_o}{\Omega_o} \right)^2 \left( \frac{2\pi T}{\Omega_o} \right)^{2\alpha_b-1} \times \frac{\Gamma[2\alpha_b]}{\Gamma[\alpha_b + i\epsilon/2\pi T]}. \tag{2.9}
\]

On the other hand for very low \(T\), such that \(\epsilon/k_BT \gg 1\), it gives

\[
\tau_c^{-1}(\epsilon) = 2\pi\Delta_o \left( \frac{\Delta_o}{\Omega_o} \right)^2 \frac{1}{\Gamma[2\alpha_b]} \left( \frac{\epsilon}{\Omega_o} \right)^{2\alpha_b-1}. \tag{2.10}
\]

The crucial thing to notice here is that for microscopic spins \((S \ll 100)\), there will be easily observable relaxation of \(\vec{S}\). On the other hand if \(S\) is much greater than 100, the giant spin will be frozen completely, even over astronomical times, unless either bias or temperature is large \((\epsilon, k_BT \sim \Omega_o)\). Moreover, in this model we would expect that applying either a large bias or temperature would lead to a sudden "switching on" of the relaxation at some critical bias or temperature (however recall that our model is not strictly valid for either a large bias or temperature would lead to a sudden "switching on" of the relaxation at some critical bias or temperature). For smaller grains, i.e., \(S \leq 100\), one can have \(\alpha_b < 1\), and then results different from the "frozen spin" behaviour of (2.8) and (2.11) are obtained. We may define a renormalised splitting \(\Delta_s\), and a damping rate \(\Gamma_s\), by

\[
\text{B. Insulating Grain and Conducting Substrate.}
\]

The insulating grain can be analysed in a similar way, provided we gloss over some of the complications arising from the variation of both the electron density and the spin properties in the vicinity of the surface. Thus we naively extend the previous model by writing an effective interaction Hamiltonian

\[
H_{int}^s \sim \frac{1}{2} \vec{J} \vec{S} \cdot \vec{\sigma} \sum_k \sum_{\vec{q}} F_\vec{q}^s \sum_{\alpha, \beta} c_{\vec{k}+\vec{q}, \alpha}^\dagger c_{\vec{k}, \beta}; \tag{2.11}
\]

\[
F_\vec{q}^s = \int_G \frac{d^3\vec{r}}{V_o} |\Psi_e(\vec{r})|^2 \rho(\vec{r}) e^{i\vec{q} \cdot \vec{r}}, \tag{2.12}
\]

where now the form factor takes account of the decay of the electron density \(|\Psi_e(\vec{r})|^2\) as one penetrates into the sample. By the same manoeuvres as before this leads to an Ohmic coupling

\[
\alpha_s \sim g_s^2 \Omega_s^{2/3}, \tag{2.13}
\]

where \(g_s \sim g\), but is multiplied by some "scale factor" which describes the depth to which the electrons penetrate into the grain; clearly it can vary widely.

The reduction to only near surface spins makes \(\alpha_s \ll \alpha_b\) for large \(S\), but from (2.13) we see that \(\alpha_s \geq O(1)\) if \(S \geq 10^3\). For smaller grains, i.e., \(S \leq 100\), one can have \(\alpha_s < 1\), and then results different from the "frozen spin" behaviour of (2.8) and (2.11) are obtained. We may define a renormalised splitting \(\Delta_s\), and a damping rate \(\Gamma_s\), by
\[ \Delta_s = \Delta_o (\Delta_o / \Omega_o)^{\alpha_s/(1-\alpha_s)} ; \]  
\[ \Gamma_s = 2\pi \alpha_s T . \]  

At \( T = 0 \) one can get coherent motion if \( \alpha_s < 1/2 \); however any such coherence is destroyed once \( \Gamma_s \geq \Delta_s \), which even for microscopic values of \( S \) (i.e., \( S \sim 10 \)) will occur at extremely low temperatures, usually much less than 1 mK. For higher \( T \) we find, when \( \epsilon = 0 \) and \( \alpha_s < 1 \), that \( \tau^{-1} \) is still given by (2.9), which in terms of \( \Delta_s \) is

\[ \tau^{-1}(T) = 2\Delta_s \frac{\Gamma^2(\alpha_s)}{\Gamma(2\alpha_s)} \left( \frac{2\pi T}{\Delta_s} \right)^{2\alpha_s-1} . \]  

For finite \( \epsilon \) and \( T \), the form (2.8) is still valid, as is (2.10) when \( \epsilon \gg T \).

From all of these results we conclude that for low \( T \) and \( \epsilon \), the relaxation dynamics of mesoscopic grains will be extremely slow in the presence of a conducting substrate. Thus, although our models have certainly not included all possible nuances of the surface physics, one conclusion is very clear and unlikely to be affected by further refinements, i.e., that for \( T, \epsilon \ll \Omega_o \), mesoscopic magnetic grains on conducting substrates will have their spin dynamics completely frozen. However once either \( T \) or \( \epsilon \) is of order \( \Omega_o \) or greater, these dynamics will rapidly be liberated. Again, these statements will be modified if nuclear spin effects are important (Section V).

C. Conducting Grain and Insulating Substrate.

In the opposite extreme when the substrate is insulating, the character of the electron states is quite different - they form a set of discrete states \( |\mu, \sigma\rangle \) inside the grain, with mean spacing \( \Delta s \approx D / s \) for bandwidth \( D \). We describe these states via creation and annihilation operators \( A^\dagger_{\mu,\sigma}, A_{\mu,\sigma} \); these states will be rather complicated, even in the absence of spin-orbit coupling. It is very useful to separate the exchange scattering interaction into bulk and surface parts, where now the bulk part sums over the whole sample, assuming that all the \( J_i \) are the same, whilst the surface part describes the difference between \( H_{int}^{bulk} \) and the true interaction Hamiltonian \( H_{int} \): thus if

\[ H_{int} = \frac{1}{2} \sum_{i \in V_o} J_i (\hat{s}_i \cdot \hat{\sigma}^\alpha) \hat{c}_{i,\alpha} \hat{c}_{i,\beta} , \]  

with the \( \hat{c}_{i,\alpha} \) creating electron on site \( i \), we write

\[ H_{int}^{bulk} = \frac{1}{2} \sum_{i \in V_o} (\hat{s}_i \cdot \hat{\sigma}^\alpha) \hat{c}_{i,\alpha} \hat{c}_{i,\beta} = \frac{\mathcal{J}}{2S} (\hat{S} \cdot \hat{\sigma}^\alpha) \sum_{i \in V_o} \hat{c}_{i,\alpha} \hat{c}_{i,\beta} = \frac{\mathcal{J}}{2S} (\hat{S} \cdot \hat{\sigma}^\alpha) \sum_{\mu} A^\dagger_{\mu,\alpha} A_{\mu,\beta} , \]  

and the surface term

\[ H_{int}^S = \frac{1}{2} \sum_{i \in V_o} (J_i - \mathcal{J})(\hat{s}_i \cdot \hat{\sigma}^\alpha) \hat{c}_{i,\alpha} \hat{c}_{i,\beta} . \]  

We notice that the bulk term is diagonal in both the site representation and the exact eigenstate representation, because of completeness of states. This is important, because the condition \( \mathcal{J} \gg \Omega_o \) means that the electron spins inside the system will rotate rigidly with \( \hat{S} \), and from the last form in (2.18) we see that there is no way this adiabatic rotation can excite the low energy orbital states \( |\mu, \sigma\rangle \) of the system. Thus the effect of \( H_{int}^{bulk} \) is simply to renormalise the moment of inertia of the giant spin.

On the other hand the surface term does have off-diagonal matrix elements \( \langle \mu | H_{int}^S | \mu' \rangle \) and this may result in infrared electron-hole pair production when \( \hat{S} \) flips. The calculation of the corresponding coupling parameter \( \alpha_s \) proceeds in exactly the same way as in the previous subsection (the corrections to the plane-wave calculation are small in a large grain with \( k_F R_o \gg 1 \)). Thus at high temperatures the internal conduction electrons constitute an Ohmic bath with

\[ \alpha_s \sim g^2 S^{2/3} \]  

(here $g_s$ is given by $(J - \bar{J})N(0)$ averaged over the surface). Now we notice that this result holds only at $T > \Delta \epsilon_\mu$.

At lower temperature the effective coupling constant goes to zero exponentially with $T$:

$$\alpha_{s}^{eff} \sim \alpha_s e^{-\Delta \epsilon_\mu/T};$$

and the infrared renormalisation of the tunneling rate stops at

$$\Delta_s = \Delta_o \left( \frac{\Delta \epsilon_\mu}{\Omega_o} \right)^{\alpha_s}; \quad (\text{for } \Delta \epsilon_\mu \ll \Omega_o).$$

If $\Delta \epsilon_\mu \gg \Omega_o$ one has $\Delta_s \approx \Delta_o$, and we do not have to worry about conduction electrons in the grain at all because they will rotate adiabatically with $\vec{S}$.

The onset of coherent motion in zero bias can be found by comparing the damping rate $\Gamma_s = 2\pi \alpha_{s}^{eff} T$ with $\Delta_s$.

Expressing all the parameters in terms of $S$ we find the criterion of coherence in the form

$$\frac{T}{\Delta \epsilon_\mu} \sim \frac{ST}{D} \ll \frac{1}{\ln \frac{2\pi g^2 S^2 \gamma D}{S \Delta}},$$

For experiments at $mK$ temperatures and small bias $\epsilon < \Delta \epsilon_\mu$, this means that electronic dissipation effects can be ignored in such grains provided $S < 10^5$. On the other hand for $T$ and/or $\epsilon > \Delta \epsilon_\mu$, we return to the case of strong Ohmic dissipation which looks very much the same as that for an insulating grain on a metallic substrate.

### III. GIANT SPINS AND THE PHONON BATH

In the absence of electrons (and of nuclear spins) relaxation in a biased system proceeds via phonons. This is a very slow process - typical relaxation times are of order months. A number of previous studies have appeared; Garg & Kim [16] and Chudnovsky [21] have discussed grain tunneling within a straightforward Caldeira-Leggett approach, assuming a continuum of final states available for the tunneling - we have already explained problems with this approach in the introduction. Politi et al. [14] have given a very thorough discussion of the effects of phonons on tunneling in the Mn$_{12}$O$_{12}$ acetate system, including the effects of discrete initial and final states, and the non-diagonal couplings (to be defined below). They also take proper account of the symmetries and selection rules in the tunneling transitions. Whether their theory explains the relaxation measurements in Mn$_{12}$O$_{12}$ acetate [22] is another matter - in our opinion the explanation may have more to do with dipolar interactions between the molecules [23]. The treatment of phonon effects we shall give is more general although quite close to theirs. We first discuss the form the low-energy effective Hamiltonian should take, depending on the symmetries of $H_o(\vec{S})$ and the direction of the applied bias field, and determine the relaxation of the giant spin dynamics for the different possible couplings. The values of the relevant couplings are then estimated - no attempt is made at exact calculations, since these would depend so much on the detailed nature of the system in question. Our main result is that these couplings are so weak that phonons by themselves are incapable of causing anything but very slow relaxation of $\vec{S}$.

#### A. Effective Hamiltonians, and Relaxation Rates

We start by assuming, for ease of discussion, a simple easy-axis, easy-plane bare giant spin Hamiltonian in a bias (Eqs. (1.1), (1.2)). The phonons are described by

$$H_o = \sum_\nu \nu Q(\hat{b}^\dagger Q \hat{b} + 1/2),$$

with $\nu Q = cq$ (we take account of polarization later, and ignore optical modes), and the general form of the truncated interaction is then

$$H_{int} = \hat{\tau}_z \sum_\nu C^{\nu}_Q(h)x_\nu + [\hat{\tau}_x \frac{\Delta_o}{\Omega_o} \sum_\nu C^{\nu}_Qx_\nu + H.c.];$$

$$x_\nu = \frac{1}{(2m_Q \nu)^{1/2}}(\hat{b}^\dagger \nu + \hat{b} - \nu),$$

where $h = \gamma_c SH_z$ is the transverse bias.

It is unusual to consider non-diagonal couplings (i.e., couplings in $\hat{\tau}_x$), since in general their effects are reduced, compared to those of diagonal properties, by a factor $(\Delta_o/\Omega_o)^2$. However we shall see that in this system the diagonal
couplings are zero unless we apply a field perpendicular to the easy axis. The reason for this is very simple - time reversal symmetry implies that the magnetoacoustic interaction cannot distinguish between grain states \( \vec{S}_1 \) and \( \vec{S}_2 \) when \( \vec{S}_1 = -\vec{S}_2 \) (a point which also arises in the interaction of phonons with domain walls \([1]\)), and diagonal couplings describe interactions which do distinguish such states. Applying a transverse field projects a component of \( \vec{S} \) on \( \vec{S}_2 \), and enables such a coupling; from this argument we see that \( C_{\vec{q}}(h) \) is proportional to the transverse field \( H_x \). We shall later see that

\[ C_{\vec{q}}^z \sim (\gamma_c H_x/K^z) C_{\vec{q}}^z , \tag{3.3} \]

for a field applied in the easy plane.

Before deriving \([3.1]\) and evaluating the couplings, we first give the results for relaxation rates that derive from it. Just as for the electron bath, these rates are very low, but for the opposite reason - instead of being frozen by a very strong coupling, \( \tilde{S} \) is frozen because the effective coupling is so weak!

The relaxation rates are derived by first defining Caldeira-Leggett spectral functions

\[ J_\alpha(\omega) = \frac{\pi}{2} \sum_{\vec{q}} \frac{|C_{\vec{q}}^\alpha|^2}{m_{\vec{q}}^\omega_\vec{q}} \delta(\omega - \nu_\vec{q}) , \tag{3.4} \]

where \( \alpha = z, \perp \); these describe the phonon effect on grain dynamics via 2nd order perturbation theory. It has of course been known for decades that these couplings are very weak for \( \omega \ll \Theta_D \), the Debye frequency; in fact

\[ J_\alpha(\omega) = B_\alpha(\omega/\Theta_D)^m , \tag{3.5} \]

with \( m \geq 3 \) in 3 dimensions; this is the fundamental reason for the coherent motion of polaron and defects in insulators at low temperatures \([20]\). We will see below that \( m = 3 \) in our case.

We start with the case where there is no transverse field \( H_x \), so that \( C_{\vec{q}}^z = 0 \) in \( (3.1) \). The relaxation dynamics for this case are described as damped motion between \( \vec{S} = \pm \tilde{S} \) with a relaxation rate (see, e.g., \([20]\))

\[ \tau^{-1}(\epsilon) \sim 2 \frac{\Delta_o^2}{\omega_o^2} E^2 J_\perp(E) \coth(\beta E/2) \sim 4 B_z \frac{\Delta_o^2}{\omega_o^2} (\epsilon/\Theta_D)^2 \frac{T}{\Theta_D} , \tag{3.6} \]

\[ E^2 = \Delta_o^2 + \epsilon^2 , \tag{3.7} \]

where the longitudinal bias \( \epsilon = \gamma_c S H_z \). This rate is of course very small \( (\tau^{-1} \ll \Delta_o, \epsilon) \). Notice that \( \tau^{-1}(\epsilon) \) is quite different from that rate which would be calculated via a naive application of the spin-boson results \([1]\); in fact it is smaller by a factor \( (\epsilon/\omega_o)^2 \).

On the other hand application of a transverse field \( \vec{H} = \tilde{z} H_x \) gives a diagonal coupling, according to \( (3.3) \), which can be incorporated into a spin-boson model; in the absence of any bias \( \epsilon \) one finds a relaxation rate

\[ \tau^{-1} \sim 2 J_z(\Delta_o) \coth(\beta \Delta_o/2) \sim 4 B_z \frac{\Delta_o}{\Theta_D} \frac{T}{\Theta_D} , \tag{3.8} \]

or, in a non-zero bias,

\[ \tau^{-1}(h, \epsilon) \sim 2 \frac{\Delta_o^2}{E^2} J_z(E) \coth(\beta E/2) \sim 4 B_z(h) \frac{\Delta_o}{\Theta_D} \frac{T}{\Theta_D} , \tag{3.9} \]

which is independent of \( \epsilon \) as long as \( \epsilon \ll T \). Using \( (3.3) \) we may express \( B_z \) in terms of \( B_\perp \), and write

\[ \tau^{-1}(h) \sim 4 B_\perp \left( \frac{\gamma_c H_x}{K^z} \right)^2 \frac{\Delta_o}{\Theta_D} \frac{T}{\Theta_D} . \tag{3.10} \]

These rates are very long, because there are so few phonons available to absorb the tunneling energy at these frequencies - we shall give typical numbers once the coupling constant \( B_\perp \) has been determined.
B. Magnetoacoustic Couplings

The magnetoacoustic coupling is of course very well understood [1]; its effect on the WKB tunneling action of a grain has been analysed by Garg & Kim [6] and by Chudnovsky [21]. There has been some confusion about the relation between Chudnovsky’s ”angular momentum coupling” and the usual magnetoacoustic coupling, which we try to resolve here.

The essential points we wish to make are that the couplings $C_{\hat{q}}$ in (3.3), and $B_\perp$ in (3.5), are given to within dimensionless constants $\sim O(1)$ by

$$|C_{\hat{q}}| \sim 2S\Omega_o \, |\hat{q}|; \quad (3.11)$$

$$B_\perp \sim S^2\Omega_o^2 / \Theta_D, \quad (3.12)$$

so that the relaxation rate in a longitudinal field $H_z$ is finally

$$\tau^{-1}(\epsilon) \sim S^2\Delta_o \frac{\Delta_o}{\Theta_D} \left(\frac{\epsilon e H_z}{\Theta_D} \right)^3 \coth \left(\frac{\epsilon e H_z}{2k_B T}\right), \quad (3.13)$$

whereas the relaxation rate in a non-zero transverse field $H_z$ has an additional term

$$\tau_z^{-1}(\hbar) \sim S^2\Delta_o \frac{\Delta_o}{\Theta_D} \left(\frac{\epsilon e H_z \Omega_z}{\Theta_D K_{\|}} \right)^2 \frac{T}{\Theta_D}, \quad (3.14)$$

By comparing (3.13) and (3.14) we find their ratio to be $\sim (H_z SK_{\|}/H_z \Omega_o)^2$. This ratio is large unless the external field is almost parallel to $\hat{x}$.

To get some ideas of the relaxation rates implied by these formulae, let us consider a situation where $S = 10^3$, $\Delta_o \sim 1 MHz$, $T \sim 50 mK$, $\Theta_D \sim 100 K$, and $\epsilon \sim \epsilon e H_z \sim 50 mK$. Then we get $\tau(\epsilon) \sim 10^4 sec$ (i.e., 3 hours). Thus, if we ignore nuclear spin effects, the relaxation times at $50 mK$ are already very long; and $\Delta_o \sim 1 MHz$ is actually a large value for $\Delta_o$. If the bias energy is further reduced to the $\Delta_o$ scale we get astronomically large $\tau$.

To get (3.11) and (3.12) we start by noting that Chudnovsky’s angular momentum coupling

$$H_{AM} = \int_G d^3\vec{\hat{r}} \frac{\vec{\hat{S}}}{S} \cdot \left(\nabla \times \vec{u}(\vec{r})\right), \quad (3.15)$$

integrated over the grain volume, is nothing but that set of transverse terms in the magnetoacoustic coupling which is responsible for transporting angular momentum to the transverse phonons. This magnetoacoustic coupling has the general form

$$H_{ME} = \langle \gamma e/S \rangle^2 \int_G d^3\vec{\hat{r}} a_{ijkl} \vec{S}_i \vec{S}_j u_{kl}(\vec{r}), \quad (3.16)$$

where the coefficients $\gamma e a_{ijkl}$ are energy densities, and in fact are simply combinations of the magnetic anisotropy constants (i.e., of $K_{\|}$ and $K_\perp$ in the case of $H_z (\vec{S})$), since this is the only energy scale that can be involved. From this we see that for the grain, $H_{ME} \sim SK |\hat{q}|$. However we also have

$$\vec{\hat{S}} = i [\vec{\hat{S}}, H_o (\vec{\hat{S}})] \sim \Omega_o S, \quad (3.17)$$

and so $H_{AM} \sim S\Omega_o |\hat{q}|$ also; the direct relation to the $a_{ijkl}$ can be easily determined for any particular $H_o (\vec{S})$ and lattice type.

From this it follows that $C_{\hat{q}} \sim 2S\Omega_o |\hat{q}|$ in (3.11). It is instructive to rederive this by estimating the correction $\delta A$ to the bare tunneling action in a given strain field $u_{kl}$, i.e., finding

$$\Delta_o \{u_{kl}\} = \Omega_o e^{-A(u_{kl})} \sim \Delta_o e^{-\delta A(u_{kl})}, \quad (3.18)$$

This is determined trivially from the interaction $H_{AM}$ by integrating over time (assuming a negligible change in the instanton path); writing
\[ \delta A(\{u_{kl}\}) = \sum_{\vec{q}} \frac{C^\perp_{\vec{q}}}{\Omega_o} \vec{q}, \quad (3.19) \]

we get

\[ C^\perp_{\vec{q}} = 2S\Omega_o \int_G \frac{d\vec{r}}{V_o} e^{i\vec{q} \cdot \vec{r}} (\hat{\vec{e}}_{\vec{q}} \times \hat{\vec{e}}_{\vec{q}}) \cdot \hat{\vec{z}} \]

\[ = 2S\Omega_o F_{\vec{q}} (\vec{q} \times \hat{\vec{e}}_{\vec{q}}) \cdot \hat{\vec{z}}, \quad (3.20) \]

where \( F_{\vec{q}} \) is the form factor, and \( \hat{\vec{e}}_{\vec{q}} \) is the polarization of the relevant phonon; for long wavelength phonons (\( qR_o \ll 1 \)) this gives us equation (3.11); and \( B^\perp_\omega \) in (3.12) immediately follows using (3.4). Notice that in the case of the easy-axis/easy-plane Hamiltonian \( H^\perp_o (\vec{S}) \), the same argument gives

\[ C^\perp_z \sim 2S\Omega_o \gamma e H_x K_\parallel |\vec{q}|, \quad (3.21) \]

It is sometimes useful to write all these couplings in terms of the relevant sound velocity, using \( \Theta_D^4 \sim \rho c_5^3 \); usually this will mean the transverse sound velocity since it is usually lower than the longitudinal one. Then without specifying exactly which of the many combinations of the \( a_{ijkl} \) enter into our expression for some particular symmetry, but just calling this combination \( K_a \), we write

\[ J^\perp(\omega) \sim \frac{S^2 K^2_a}{\rho c^3} \omega^3; \quad (3.22) \]

\[ J^z(\omega) \sim \frac{K^2_a}{\rho c^3} \left( \frac{MH_x}{K_\parallel} \right)^2 \omega^3. \quad (3.23) \]

Finally, we remind the reader that none of the analysis in this section takes account of nuclear spins.

IV. GIANT SPINS AND THE SPIN BATH

We now turn to a very different kind of bath from the phonon and electron bath, viz., the “spin bath” made up from nuclear spins both inside and outside the grain, paramagnetic electronic impurity spins, and any loose spins in or near the grain surface. As noted in the introduction, this spin bath cannot be mapped onto an oscillator bath, and so a completely different kind of theoretical framework is necessary to describe its effects on mesoscopic or macroscopic quantum objects.

The main point of this section is to show how the spin bath, interacting in isolation with a giant spin \( \vec{S} \), will basically block any relaxation at all, unless either the grain happens to be in a total field (produced by the sum of the external field and the “internal bias field” generated by the nuclei) which is very small, less than roughly \( \Delta_o \); or the nuclear spin diffusion mechanism of energy bias diffusion allows the system to find the resonance window. In what follows we first briefly recall the 4 mechanisms by which the spin bath controls the giant spin dynamics, and set up the formalism required to analyse magnetic relaxation in the presence of nuclear spins. We then derive the relaxation dynamics of \( \vec{S} \), first ignoring the internuclear spin diffusion (to see how \( \vec{S} \) is frozen), then including it (to show how \( \vec{S} \) can then relax).

A. Effective Hamiltonian, and the 4 Coherence-Blocking Mechanisms.

In an earlier series of papers [10,23–26] we have constructed a theoretical description of a giant spin interacting with a more or less arbitrary spin bath, and solved this model for the quantum dynamics of \( \vec{S} \) in the absence of any external bias. The same sort of model has also been set up to describe an arbitrary unbiased macroscopic coordinate interacting with a spin bath. Elsewhere we have used this model to deal with coherence experiments in SQUID’s [27], and also shown how it leads to some remarkable conclusions about the quantum theory of measurement (particularly concerning the role that nuclear spins play in making the mesoscopic world behave classically [28]).
Here we generalise this work to deal with a biased giant spin. As before, we shall assume that the giant spin can be truncated to 2 levels, and that $T_\epsilon \ll \Omega_o$; all effects of higher levels will be absorbed into the parameters of an effective Hamiltonian. This Hamiltonian was derived in Refs. [10, 24, 26], and is written for the biased case as

$$H_{eff} = 2\Delta_o \{ \tilde{\tau}_+ \cos \Phi + \sum_{k=1}^N (\alpha_k \tilde{n}_k - i \xi_k \tilde{v}_k) \cdot \tilde{\sigma}_k \} + H.c. \} - \gamma_c \tilde{S} \cdot \tilde{H}_o + \frac{\gamma_c}{2} \sum_{k=1}^N \omega_k^\parallel \tilde{l}_k \cdot \tilde{\sigma}_k + \sum_{k=1}^N \omega_k^\perp \tilde{m}_k \cdot \tilde{\sigma}_k. \quad (4.1)$$

There is also an inter-nuclear interaction of dipolar form; thus in general we have to add to (4.1) a term

$$\hat{V}(\{\tilde{\sigma}_k\}) = \sum_{k \neq k'} V_{kk'}^{\alpha\beta} \hat{\sigma}_k^\alpha \hat{\sigma}_k'^\beta. \quad (4.2)$$

We shall not need to write down the matrix elements $V_{kk'}^{\alpha\beta}$ in detail; their strength is $\sim T_2^{-1}$, the transverse nuclear relaxation rate. Typically we shall assume that $\tilde{H}_o$ is parallel to the easy axis of our easy-axis/easy-plane system, so that the total effect of $\tilde{H}_o$ will be written as

$$\gamma_c \tilde{S} \cdot \tilde{H}_o \rightarrow \xi_H \tilde{\tau}_z, \quad (4.3)$$

where $\xi_H = \gamma_c S H_o$ (we change notation - the symbol $"\epsilon\"$ will be used to denote the "internal" bias coming from the nuclei). The spin bath is described in (4.1) by spin-1/2 variables $\{\tilde{\sigma}_k\}$. The vectors $\tilde{n}_k$, $\tilde{v}_k$, $\tilde{l}_k$, and $\tilde{m}_k$ are unit vectors; the "static" couplings between $\tilde{S}$ and the $\{\tilde{\sigma}_k\}$ are described by the last two terms, and the dynamic effects are given by the terms in the curly brackets. The longitudinal static coupling $\omega_k^\parallel$ tells us the change in energy of $\tilde{\sigma}_k$ when $\tilde{S}$ flips between $\pm \tilde{S}$; thus the vector $\omega_k^\parallel \tilde{l}_k$ represents the difference between the effective field acting on $\tilde{\sigma}_k$ before $\tilde{S}$ flips and that field after $\tilde{S}$ flips. If the $\{\tilde{\sigma}_k\}$ describe nuclei, this field will be almost entirely hyperfine in origin, unless the $\{\tilde{\sigma}_k\}$ are produced by truncating the levels of a higher-spin nucleus (i.e., a nuclear moment $\tilde{l}_k$ with $|\tilde{l}_k| > 1/2$). In this latter case the total field acting on the $\{\tilde{\sigma}_k\}$ will also include, e.g., quadrupolar contributions; it then follows that the field $\omega_k^\perp \tilde{m}_k$ will be non-zero. This transverse field is the sum of the fields acting on $\tilde{\sigma}_k$ before and after $\tilde{S}$ flips.

The dynamic terms in (4.1) originate as follows. The phase $\Phi = \pi S + \sum_k \phi_k$ is the sum of the "Kramers phase" $\pi S$ (see Introduction - notice that in (4.1) we have not yet included $\cos \pi S$ into the definition of $\Delta_o$) and a renormalisation $\sum_k \phi_k$ of this phase, caused by the $\{\tilde{\sigma}_k\}$. The term $\alpha_k$ describes the amplitude for $\tilde{\sigma}_k$ to flip, under the influence of $\tilde{S}$, when $\tilde{S}$ flips; both $\phi_k$ and $\alpha_k$ partly derive from the unitary "transfer matrix" $\tilde{T}_k$ describing the effect of a tunneling $\tilde{S}$ on $\tilde{\sigma}_k$:

$$|\chi_k^{fin}\rangle = e^{-i \int d\tau H_{int}(\tau)} |\chi_k^{in}\rangle \equiv \tilde{T}_k^\pm |\chi_k^{in}\rangle; \quad (4.4)$$

$$\tilde{T}_k^\pm = e^{\pm i (\alpha_k \tilde{n}_k \cdot \tilde{\sigma}_k + \phi_k)}. \quad (4.5)$$

Here $H_{int}(\tau)$ describes the microscopic interaction between $\tilde{S}$ and $\tilde{\sigma}_k$; $|\chi_k^{in}\rangle$ is the environment state before $\tilde{S}$ flips, and $|\chi_k^{fin}\rangle$ after $\tilde{S}$ flips. The signs $\pm$ refer to the path traced by $\tilde{S}$ whilst flipping. The term in $\xi_k$ and further contributions to $\phi_k$ and $\alpha_k$ are necessary because environmental spins also have an effect on the tunneling action for $\tilde{S}$ (in exact analogy with the nondiagonal terms discussed in Section II).

The derivation of (4.1) has been described in detail elsewhere [10]. Here we shall simply quote the values of the parameters in (4.1) that arise if we start with the microscopic hyperfine Hamiltonian

$$H_{hyp} = H_o^\perp (\tilde{S}) + \frac{1}{2} \sum_{k=1}^N \omega_k \tilde{S} \cdot \tilde{l}_k \cdot \tilde{\sigma}_k$$

$$= \frac{1}{S} \left[ (-K_\parallel S_z^2 + K_\perp S_y^2) + \sum_{k=1}^N \omega_k \tilde{S} \cdot \tilde{l}_k \right], \quad (4.6)$$

i.e., our usual easy-axis/easy-plane model coupled to nuclei via hyperfine couplings $\omega_k$; we get
\[
\Phi = \pi S \\
\xi_k = \alpha_k/\sqrt{2} = \pi \omega_k/2\Omega_\nu \quad (\phi_k = 0) \\
\omega_k^0 = \omega_k \\
\omega_k^\perp = 0 \\
\bar{n}_k = (\hat{x}, \hat{y})/\sqrt{2} \\
\bar{v}_k = -\hat{x} \\
\bar{l}_k = \hat{z}
\]  

(4.7)

The assumption in (4.7), that the hyperfine coupling \( \omega_k \ll \Omega_\nu \), is almost always true. In any case \( \alpha_k, \xi_k, \) and \( \phi_k \) can be calculated as general functions of \( \omega_k/\Omega_\nu \), for any initial microscopic Hamiltonian. It is useful to bear in mind the physical meaning of the parameter \( \alpha_k \) in this regime; \( \alpha_k \) is the amplitude for \( \bar{s}_k \) to coflip with \( \bar{S} \). Note also that more general interactions than (4.6) will produce further renormalisations of \( \Delta_\nu \).

Working with (4.1) (which is of course nothing but a low-energy effective Hamiltonian, in the usual spirit which also led to (2.4) and (3.1)) allows us to bring out the 4 essential physical mechanisms operating in the dynamics of \( \bar{S} \). These are:

(i) Even when \( \bar{H}_\nu = 0 \), an internal field bias \( \epsilon = \sum_{k=1}^N \omega_k^0 \bar{\sigma}_z^k \) acts on \( \bar{S} \). Typically for nuclei we expect the hyperfine couplings to be tightly clustered around a principal value \( \omega_0 \), with \( \omega_0 \gg \Delta_\nu \). There will however be a spread \( \delta\omega_k \), due not only to inter-nuclear couplings (principally via dipolar or Nakamura-Suhl interactions, with \( \delta\omega_k \sim 10^{-4} - 10^{-6} \text{Hz} \)) but also other couplings such as transfer hyperfine couplings. One may then define a "density of states" \( W(\epsilon) \) for bias \( \epsilon \) (Fig.3); if \( \mu = N^{1/2} \delta\omega_k/\omega_0 \ll 1 \), this consists of Gaussian peaks

\[
G_\mu(\epsilon - \omega_0 \Delta N/2) = \frac{1}{\Gamma_\mu \pi^{1/2}} \exp\left\{-(\epsilon - \omega_0 \Delta N/2)/\Gamma_\mu^2\right\},
\]

(4.9)
of width

\[
\Gamma_\mu = \mu \omega_0,
\]

(4.10)

around \( \epsilon = \omega_0 \Delta N/2 \), where \( \Delta N = N_\uparrow - N_\downarrow \) is the total nuclear polarisation, inside a Gaussian envelope of width \( N^{1/2} \omega_0 \); this envelope extends out to a maximum bias \( \pm N \omega_0 \):

\[
W(\epsilon) = 2^{-N} \sum_{\Delta N} C_{N}^{(N+\Delta N)/2} G_\mu(\epsilon - \omega_0 \Delta N/2).
\]

(4.11)

(Here we assume that \( N = \text{even integer; otherwise } \Delta N = 0 \) is impossible! The modifications required for \( N = \text{odd} \) are trivial and irrelevant to the ensuing discussion). In most cases, however, \( \mu > 1 \), and the different "polarisation groups" (i.e., the different peaks) completely overlap, and we end up with a Gaussian envelope for \( W(\epsilon) \), i.e.,

\[
W(\epsilon) \rightarrow \frac{f}{\omega_0} \exp\{-2\epsilon^2/(\omega_0^2 N)\}; \quad (\mu > 1).
\]

(4.12)

where \( f = \sqrt{2/\pi N} \).

Now for tunneling to occur at all, the total bias \( (\epsilon + \xi_H) \) must be \( \leq \Delta_\nu \), otherwise the grain is simply trapped. The fraction of grains in an ensemble having bias \( \leq \Delta_\nu \) is roughly \( A \sim \Delta_\nu/N \delta\omega_k \) if \( \mu \ll 1 \), and \( A \sim \Delta_\nu/N^{1/2} \omega_0 \) if \( \mu > 1 \); the result for \( \mu \ll 1 \) arises because only a fraction \( \sim \Delta_\nu/N^{1/2} \delta\omega_k \) of that portion \( f = \sqrt{2/\pi N} \) of grains having \( \Delta N = 0 \) can flip when the different polarisation groups do not overlap.

Thus only a small fraction \( A \) of spins are not "degeneracy blocked" from tunneling (in the absence of nuclear dynamics, the effects of which are discussed below and in a Section [3]). This degeneracy blocking mechanism operates just as easily whether there is an external bias or not - in both cases only grains in the small window of bias can make transitions.

(ii) Even in the absence of an external thermostat, the nuclear system can change the internal bias \( \epsilon \). This occurs via dipolar interactions between the nuclei, at a rate \( T_2^{-1} \). This pairwise flipping of nuclei conserves \( \Delta N \) but allows the nuclear bath to "wander in bias space" over the full range of bias energy associated with a particular polarization group, i.e., over an energy range \( \sim \mu \omega_0 = N^{1/2} \delta\omega_k \). If the spread \( \delta\omega_k \) is due entirely to dipolar interactions, so \( \delta\omega_k \sim T_2^{-1} \), then the fluctuating bias covers the whole energy range in a time \( T_2 \). This fluctuating bias \( \epsilon(t) \) can destroy coherence, but it can help magnetic relaxation, by helping the system find the small bias window.
(iii) The third mechanism, called "orthogonality blocking", arises because the transverse fields \( \omega_k^\perp \) acting on the environmental spins (which are typically \( \ll \omega_k^\parallel \)) cause a mismatch between the initial and final nuclear wave-functions, in a way reminiscent of Anderson’s catastrophe \[^{28}\]. Defining \( \sin 2\beta_k = \omega_k^\perp / \omega_k^\parallel \) (assuming \( \beta_k \ll 1 \)), this blocking effect can be parametrised by \( \kappa \), where

\[
e^{-\kappa} = \prod_{k=1}^N \cos \beta_k ,
\]

so that \( \kappa \approx 1/2 \sum_k \beta_k^2 \). Orthogonality blocking inhibits both coherence and relaxation of \( \vec{S} \); it spreads out the high-frequency (\( \sim\Delta_o \)) response of the grain to lower frequencies.

(iv) A final interaction mechanism arises simply because the giant spin can flip the nuclear spins. This process is parametrised by the \( \alpha_k \) in (4.5); for the whole spin bath one finds that on average a number \( \lambda \) of spins will be flipped each time \( \vec{S} \) flips, where \( \lambda = 1/2 \sum_k \alpha_k^2 \) for \( \alpha_k \ll 1 \). This causes phase decoherence; we have called this mechanism "topological decoherence" because it adds a random winding number to the effective action for \( \vec{S} \). It has a decoherence effect on tunneling, and thus inhibits magnetic relaxation.

The formal discussion of all these mechanisms has appeared in our previous work. In the next 2 sub-sections we will deal first with the dynamics of \( \vec{S} \) in the absence of spin diffusion between the nuclei, to show how the nuclei keep all but a tiny fraction of grains frozen; then we show how spin diffusion changes this picture.

### B. Tunneling Rate in a Bias

We assume that neither the grain nor the spin bath is connected to an external thermostat except at time \( t \leq 0 \) (in reality both are coupled to phonons and possibly electrons - see next section). We also neglect nuclear spin diffusion here, and estimate its role later. Thus relaxation of \( \vec{S} \), in a bias, can only occur if the bias field due to the \( \{\vec{\sigma}_k\} \) allows states \( |\uparrow;\{\vec{\sigma}_n\} \rangle \), with the \( \{\vec{\sigma}_k\} \) in some polarisation state \( \Delta N^{in} \), to overlap in energy (within \( \sim \Delta_o \)) with some states \( |\downarrow;\{\vec{\sigma}_m\} \rangle \), where the polarisation state \( \Delta N^{fin} \neq \Delta N^{in} \) in general. Thus we expect some nuclear spins to flip, if \( \vec{S} \) is to relax, although if \( \mu \) is large, and the applied bias is small one can even have energy overlap between initial and final states having the same \( \Delta N \neq 0 \) (this does not, of course, mean that no spins are flipped during the transition, but only that none have to be flipped).

The formal treatment of this problem is a generalisation of our treatment of the unbiased case. We write, for the time correlation function \( P(t) = \langle \hat{\tau}_z(t)\hat{\tau}_z(0) \rangle \), the form \[^{[0]}\]

\[
P(t; T; \xi_H) = 1 + \int dc W(c)e^{-\beta c} \sum_{M=-N/2}^{N/2} \left[ P_M(t, \epsilon + \xi_H - M\omega_o/2) - 1 \right] .
\]

This formula is crucial, and so we now spend a little time explaining it. The average \( \int dc \) is over initial bias, with both thermal and density of states weighting. \( P_M(t, \epsilon) \) is a function defined in Ref. \[^{[1]}\]; it describes all those grains in an ensemble having bias \( \epsilon \) and for which, every time \( \vec{S} \) flips, the polarisation state of the \( \{\sigma_k\} \) changes by \( 2M \):

\[
P_M(t) = \int_0^\infty dy e^{-y} \sum_{\nu=-\infty}^\infty \int_0^{2\pi} d\varphi \frac{F_N(\nu)e^{2i\nu(\Phi - \varphi)}}{2\pi} P_M^{(0)}(t, \epsilon, \varphi, y) ,
\]

in which \( P_M^{(0)} \) describes a simple biased 2-level system (cf. Eq.\ZN)

\[
P_M^{(0)} = 1 - \frac{\Delta_M^2(\varphi, y)}{E_M^2(\varphi, y)} \sin^2(E_M(\varphi, y)t) ,
\]

\[
E_M^2 = \epsilon^2 + \Delta_M^2 ,
\]

\[
\Delta_M(\varphi, y) = 2\Delta_o \left| \cos \varphi J_M(2\sqrt{(\lambda - \lambda')}y) \right| ,
\]

and...
with $\lambda = 1/2 \sum_k \alpha_k^2$, $\lambda' = 1/2 \sum_k \alpha_k^2 (\eta_k^2)$ (so that $\lambda \geq \lambda'$). Eq.(4.15) for $P_M(t)$ can be understood as combining a "phase average"

$$
\sum_\nu \int \frac{d\varphi}{2\pi} F'_{\lambda'}(\nu)e^{2i\varphi(\mathbf{q} - \mathbf{p})}; \quad F_{\lambda'}(\nu) = e^{-4\lambda' \nu^2},
$$

over the phase $\varphi$ and winding number $\nu$, with an "orthogonality average"

$$
\int_0^\infty d\nu e^{-\nu y}
$$

These averages are performed over the biased 2-level correlation function $P^{(0)}_{M}(t, \epsilon, \varphi, y)$, in which the tunneling amplitude $\Delta_{M}(\varphi, y)$ depends on $M$, $\varphi$, and $y$ via (4.14).

Eq.(4.14) for $P(t)$ may seem a little odd, since it sums over apparently independent processes in which the polarisation state of the nuclei changes by $2M$ each time $\delta S$ flips. Why should the polarisation state change by $2M$ each time? After all, when $\delta S$ flips, the number of nuclei which flip is random (with an average number $\lambda$)! The answer to this question comes from energy conservation considerations.

Consider a single grain that at $t = 0$ has its nuclear environment in the state $|\chi_1, \epsilon_0, \Delta N\rangle$ corresponding to the polarisation state $\Delta N$ and the bias energy $\epsilon_0$ (see Fig.4). Now $\delta S$ flips - what are the possible final states for the combined system, which have energies in resonance with $\epsilon_0$? There are three possibilities:

1. None of the environmental spins flip; then the final state is $|\chi_1, \epsilon_f, -\Delta N\rangle$ with $\epsilon_f = -\epsilon_0$, since $\tau_z \sigma_k^z$ changes sign.

2. Some small number of environmental spins, say $r \ll N$, flip together with $\delta S$, so that the net change in the nuclear spin polarisation is $2M$ (i.e., $(r + M)/2$ up and $(r - M)/2$ down spins are flipped). As a first approximation the final state energy will be $\epsilon_f \approx -\epsilon_0 + M \omega_0$; assuming that the spread in nuclear frequencies is small, we may neglect for the moment the correction $\delta \epsilon_r \sim \delta \omega_k \sqrt{F} \ll (\mu \omega_0, \omega_0)$. We denote these states as $|2\rangle$, and $|3\rangle$ in Fig.4.

3. An enormous number of nuclear spins flip together with $\delta S$, with $r$ as large as $N$; in this case we can neglect the correction $\delta \epsilon_r \sim \mu \omega_0$ any more, because it is comparable or even larger (for $\mu > 1$) than the typical difference $M \omega_0((2r - M)/2 \sim \omega_0$. Then we can we use $\delta \epsilon_r$ to "fine tune" a resonance (which is impossible, in general, for the case (2)). Thus, we find in this case $\epsilon_f = -\epsilon_0 + M \omega_0 + \delta \epsilon_r$. The correction strongly depends, of course, on the particular set of nuclei flipped; a "fine tuned" situation is shown as state $|4\rangle$ in Fig.4.

It seems at first, that the latter possibility is the best we can do for tunneling in resonance. Recall, however, that the probability that any given environmental spin flips during the transition is very small ($\alpha_k^2 \ll 1$). Now, if we are going to tunnel between $|1\rangle$ and $|4\rangle$ then the amplitude of such a transition will be $\sim \Delta_o(\alpha_k^2)^N$, and for large $N$ will be even less then $2^{-N} \delta \omega_k$! Thus such a resonance is simply impossible to realise.

In fact, only a small number of environmental spins may be flipped with a reasonable probability. Since the probability to flip none is just $e^{-\lambda}$, we find the probability to flip exactly $r$ spins to be $p_r \approx e^{-\lambda} (\alpha_k^2)^r C_N^r \approx e^{-\lambda \lambda^r/r!}$, which peaks at $r \sim \lambda$. The parameter $\lambda$ may be large in some systems, but still $\lambda \ll N$. One immediately recognizes that the case (1) plays a role at small $\lambda$, while the case (2) will dominate when $\lambda \gg 1$ (formally, we could include (1) in (2) as a particular transition with $r = 0$).

Now we make use of the inequality $\omega_0 \gg \Delta_o$ to notice that among all possible transitions with different $M$, the energy mismatch between the initial and final states $\epsilon_1 - \epsilon_f = 2\epsilon_1 - M \omega_0$ is either much larger then $\Delta_o$ for all $M$, or is close to the resonance $\epsilon_1 - \epsilon_f \sim \Delta_o$ for only one specific value $M_{\epsilon_1}$, with all the other transitions being $\geq \omega_0$ away in energy. Once we have a resonance the system will make transitions from $\uparrow \downarrow \uparrow \downarrow \uparrow \downarrow$ by changing the nuclear spin polarisation by exactly $\pm 2M_{\epsilon_1}$ each time to maintain this resonance. As for all the other transitions with $M \neq M_{\epsilon_1}$, they give only very small corrections, of order $(\Delta_o/\omega_0)^2$ or less (cf. Eqs.(4.17) and (4.16)) to the main contribution to the dynamics of $\delta S$, coming from the $M = M_{\epsilon_1}$ term. Of course Eqs.(4.14) and (4.15) simply sum over all processes, and the resonance value of $M$ is included automatically.

It is worth noting that nothing depend explicitly on the initial polarisation state. This point is further illustrated in Fig.4. For given $\epsilon_1$ the initial polarisation state can be $M$, and if $\epsilon_1$ is close to the center of the Gaussian $G_{\mu}(\epsilon - M \omega_0/2)$, then the resonance value is $M_{\epsilon_1} = M$. If however $\epsilon_1$ is, say, the down-tail state of another polarisation group $M + 2$, then transition to the up-tail state of $-M + 2$ has exactly the same energy mismatch and polarisation change $2M$. That is why the only relevant statistical average is over the bias energy. Of course, if $\mu \ll 1$, and different polarisation groups do not overlap, the bias energy is related to the specific group and Eq.(4.14) may be written as ($\epsilon_H = 0$)

$$
P(t) = \frac{1}{2N Z(\beta)} \sum_M C_{N}^{M} \int d\epsilon G_{\mu}(\epsilon - M \omega_0/2)e^{-\beta \epsilon} P_M(t, \epsilon + \xi_H - M \omega_0/2) .
$$

(4.21)
It is easy to understand now that external bias, which is indistinguishable from the internal one as far as the giant spin is concerned, will simply shift the resonance condition for given $\epsilon$ to some other value $M_{\epsilon+\xi_H}$, as is clearly seen from the diagram in Fig.5, which is similar to Fig.4, but now with $\xi_H \neq 0$.

We may further simplify Eq.(4.14) by changing variables from $\epsilon + \xi_H - M\omega_o/2 \to \epsilon$, and write it as

$$P(t; T; \xi_H) = 1 + \sum_M \int d\epsilon \frac{W(\epsilon - \xi_H + M\omega_o/2)e^{-\beta(\epsilon - \xi_H + M\omega_o/2)}}{Z(\beta)} [P_M(t, \epsilon) - 1] .$$

Then because the function $[P_M(t, \epsilon) - 1] \sim (\Delta_o/\epsilon)^2$ for large bias, and in most cases $W(\epsilon)$ is a smooth function on a $\Delta_o$ scale (for $\mu\omega_o \gg \Delta_o$), we have

$$P(t; T; \xi_H) = \sum_M \frac{W(M\omega_o/2 - \xi_H)e^{-\beta(M\omega_o/2 - \xi_H)}}{Z(\beta)} \int d\epsilon P_M(t, \epsilon) , \quad (\mu\omega_o \gg \Delta_o) .$$

(we assumed here that $\beta\Delta_o \ll 1$). In the opposite limiting case $\mu = 0$, when the Gaussian in (4.21) is a $\delta$-function, we have

$$P(t) = \frac{1}{2^N Z(\beta)} \sum_M C_N^{(N+M)/2} e^{-\beta M\omega_o/2} P_M(t, 0) .$$

To summarize, we may understand (4.14) in a fairly simple way as including all effects of the spin bath on the dynamics of $\vec S$. These come from the averages over phase (Eq.4.19), over the orthogonality mismatch between initial and final spin bath states (Eq.4.20), and over the internal bias $\epsilon$, acting on $\vec S$, caused by the spin bath. It also comes from summing over all possible changes in the polarisation state of the bath when $M$ flips. It is perhaps worth noting, for those who may be used to the theory of ”oscillator bath” environments [17], that the averages appearing here for the spin bath environment are very different in form, for the simple reason that in the case of the spin bath, most of the dynamics of the spins comes from their coupling to the macroscopic system itself (in our case, to $\vec S$). By contrast in the oscillator bath models, the coupling to the macroscopic system is weak ($\sim O(N^{-1/2})$, for each oscillator), and the dynamics of the individual oscillators is only weakly perturbed by the system. This is why, in the theory of the oscillator bath, one can first calculate the weakly perturbed oscillator motion as a function of the system coordinates, and then integrate out the oscillators by functional averaging. No such manoeuvre is possible for the spin bath (or indeed any other environment where the couplings are not weak), and it is not possible to map the spin bath onto an oscillator bath (see also Ref. [28]). Any attempt to do so (by, e.g., writing a spectral function $J(\omega) \sim N\delta(\omega - \omega_o)$, for a set of ”nuclear spin oscillators”, as in ref. [29], cannot be expected to give meaningful results.

From these remarks we can also see what has been left out of Eq.(4.15) for $P_M(t)$; we have left out everything coming from the independent dynamics of the spin bath (independent, that is, from $\vec S$). Formally this can be put in by including these dynamics in $P_M^{(0)}$ in (4.16). As we argue below this corresponds to allowing a time-dependent bias $\epsilon(t)$ in (4.14) and (4.15).

We neglect the effect of coupling of the nuclei to phonons which is actually an incredibly small effect in most cases. In reality the nuclear spin dynamics comes almost entirely from nuclear spin diffusion, parametrised by the transverse relaxation time $T_2$, and caused by the dipolar interaction in (4.3). As we shall see presently, if the spin bath is mainly composed of nuclei, this spin diffusion is crucial, since without it $\vec S$ cannot relax at all. Spin diffusion processes, in which $\Delta N$ remains unchanged but pairs of nuclei flip, cause $\epsilon(t)$ to fluctuate in time because of the variation $\delta\omega_k$ in the coupling of each pair to $\vec S$ (thus, for a process $|\downarrow\downarrow\rangle \to |\uparrow\uparrow\rangle$ involving 2 nuclei, the total change in internal bias $\sim \delta\omega_k$; if $N$ nuclei flip, the change in $\epsilon$ is $\sim \delta\omega_k N^{1/2}$). Now this fluctuation is fast, in the sense that for large values of $M$ the time it takes $\epsilon(t)$ to change by an energy $\Delta_M$ is usually much less than the time $\Delta_M^{-1}$ required for $\vec S$ to flip if it is in the coherence window (of energy width $\Delta_M$); this will be demonstrated below. Consequently it is not necessary (or even useful) to go through the elaborate detour of recalculating $P_M^{(0)}$ in Eq.(4.13), including a coupling to some effective ”oscillator bath”, intended to model the effects of spin diffusion. We simply write instead that for this fast diffusion problem, where $\epsilon(t)$ varies over an energy range $\mu\omega_o$, one has:

$$P(t; T; \xi_H) - \frac{1}{2} = \sum_M \frac{C_N^{(N+M-M_H)/2} e^{-\beta(M-M_H)\omega_o/2}}{2^N Z(\beta)} \left[ (P_M(t, \epsilon(\tau) + \delta\xi_H))_{\epsilon(\tau)} - \frac{1}{2} \right] ,$$

where we introduced the notation $\xi_H = M_H\omega_o/2 + \delta\xi_H$ with $M_H = $integer and $|\delta\xi_H| \leq \omega_o/2$ to define the shift in the polarisation change enforced by applying the external bias. Here (as before)
the expression for the effective relaxation rate simplifies to

\[ \langle P_M(t, \epsilon(\tau) + \delta \xi_H) \rangle_{\epsilon(\tau)} = \int_0^\infty dy e^{-y} \sum_{\nu = -\infty}^\infty \int \frac{d\nu}{2\pi} F_X(\nu) e^{2i\nu(\Phi - \varphi)} \langle P_M^{(0)}(t, \epsilon(\tau) + \delta \xi_H, \varphi, y) \rangle_{\epsilon(\tau)} , \]  
(4.26)

\[ \langle P_M^{(0)}(t, \epsilon(\tau) + \delta \xi_H, \varphi, y) \rangle_{\epsilon(\tau)} - \frac{1}{2} = \frac{1}{2} e^{-t/\tau_M(\delta \xi_H)} , \]  
(4.27)

\[ \tau_M^{-1}(\delta \xi_H) = 2\Delta_M^2 G_\mu(\delta \xi_H) , \]  
(4.28)

and \( \Delta_M(\varphi, y) \) is given by (4.18). In the usual case where \( \mu > 1 \), or if the external bias is close to a multiple of \( \omega_\mu \), the expression for the effective relaxation rate simplifies to

\[ \tau_M^{-1} = \frac{2\Delta_M^2}{G_\mu^2} , \]  
(4.29)

This concludes our formal discussion of the problem of quantum relaxation of \( \vec{S} \) when it is coupled to a spin bath. We now use the results to find \( P(t, \xi_H) \) for a few interesting cases.

We start by ignoring spin diffusion, in order to demonstrate the way in which relaxation is blocked in its absence. Let us first consider a really pathological case, in which there is no degeneracy blocking at all, i.e., \( \mu = 0 \). Then the function \( W(\epsilon) \) simply becomes a set of sharp lines, and all states in a grain ensemble may resonantly tunnel, if \( \delta \xi_H = 0 \). If \( \delta \xi_H \neq 0 \), then no resonance is possible. Initially there will be fast relaxation, involving processes where \( M \) is not large. At longer times the higher-\( M \) processes take over - recall that for large \( M \), the transition amplitude \( \Delta_M \) is very small, since \( \Delta_M \sim \Delta_\mu M^{\lambda M/2}/M! \), which collapses when \( M \gg \sqrt{N} \). To get some idea of the resulting relaxation, consider what happens if \( \xi_H = 0 \), and let us ignore topological decoherence for simplicity (which makes no difference for the long-\( t \) asymptotics of the result). Then we have

\[ P_M(t) = \int_0^\infty dy e^{-y} P_M^{(0)}(t, y) = \frac{1}{2} \int_0^\infty dy \left[ 1 + \cos(2\Delta_M(y)t) \right] , \]  
(4.30)

where \( \Delta_M(y) = \Delta_\mu J_M(2\sqrt{\lambda}y) \), assuming \( \lambda' = 0 \) (note if \( \lambda' = 0 \), then the topological phase average \( \langle \Phi \rangle \) collapses to a \( \delta \)-function \( \delta(\varphi - \Phi) \) and we get (4.31) anyway). The resulting curves are shown for \( \lambda = 5 \) in Fig.6, for various \( P_M(t) \). It is easy to then find the behaviour of \( P(t) \) by substituting (4.30) into (4.24). The steepest-descent integral over \( M \) gives an accurate answer for long times. However we more quickly derive this long time behaviour from dimensional arguments. The sum over \( M \) contains some \( \sqrt{2N} \) polarisation groups inside the broad envelope function \( C_M^{(N+M)} \), thus the crossover from \( P(t) \approx 1 \) to the equilibrium \( P(t) \approx 1/2 \) occurs around a time \( t_c \) such that \( \Delta_N^{1/2} t_c \approx 1 \), i.e.,

\[ \ln(\Delta_N t_c) \sim \sqrt{\frac{N}{2}} \ln \left( \frac{2N}{\lambda c^2} \right) . \]  
(4.31)

For shorter times the relaxation is roughly logarithmic, viz.

\[ 1 - P(t) \sim \sqrt{\frac{2}{N}} \ln \left( \frac{\ln(\Delta_N t_c)}{\sqrt{\lambda}} \right) ; \quad (t \ll t_c) , \]  
(4.32)

whereas for longer times one has

\[ P(t) - 1/2 \sim \exp \left( -3 \frac{\ln^{2/3}(\Delta_N t_c)}{N^{1/3}} \right) ; \quad (t \gg t_c) , \]  
(4.33)

with logarithmic accuracy. The numerical evaluation of \( P(t) \) is shown in Fig.7.

However even the tiniest degeneracy blocking will upset these results - in fact if \( \delta \omega_k \sqrt{N} > \Delta_M \), the result (4.30) fails completely. Since the proverbial "thunderstorm on Jupiter" is enough to give a \( \delta \omega_k \) exceeding \( \Delta_M/\sqrt{N} \) for the large values of \( M \) governing long time relaxation, we see that for relaxation (just as for coherence (10)), it is the limit of strong degeneracy blocking that is experimentally meaningful. Let us again consider the zero bias case in this limit,
where $\mu > \Delta_o/\omega_o$. We go back to (4.23), again assume $\lambda' = 0$ for simplicity, and, noting that the oscillatory $\sin^2 E_M t$ term in $P^{(0)}_M$ (Eq.(4.16)) gives an integrated contribution
\[ \int \frac{dx}{\varepsilon^2 + \Delta^2_M} \sin^2(t\sqrt{\varepsilon^2 + \Delta^2_M}) = \frac{\pi \Delta_M(y)}{2} \int_0^{2\Delta_M(y)t} dz J_0(z) ; \quad \text{(4.34)} \]
we find
\[ 1 - P(t) = \frac{\pi \Delta_o}{2} \sum_M W(M\omega_o/2)e^{-\beta M\omega_o/2} \int_0^\infty dy \frac{\pi \Delta_M(y)}{2} \int_0^{2\Delta_M(y)t} dz J_0(z) ; \quad \text{(4.35)} \]
This result describes an ensemble of grains in which a fraction $A$ of grains relaxes, leaving a fraction $1 - A$ completely unrelaxed in the infinite-time limit; $A$ is given by
\[ A = 1 - P(t \to \infty) = \frac{\pi \Delta_o}{2} \sum_M W(M\omega_o/2)e^{-\beta M\omega_o/2} \int_0^\infty dy \frac{\pi \Delta_M(y)}{2} \int_0^{2\Delta_M(y)t} dz J_0(z) ; \quad \text{(4.36)} \]
Since terms for $M > \sqrt{\lambda}$ make almost no contribution to $A$ due to the collapse of $\Delta_M$, it can be approximated for large $\lambda$ by
\[ A = \lambda^{1/4} \frac{\Delta_o}{\omega_o} e^{-N\omega_o^2/8(k_B T)^2} , \quad \text{(4.37)} \]
where $f = \sqrt{2/\pi N}$ is the number of states in the $\Delta N = 0$ polarisation group (in Ref. we showed that individual terms in the sum over $M$ decreased as $\lambda^{-1/4}$; but the sum over all $M$ gives the factor $\lambda^{1/2}$).

Thus we see that apart from the small number of grains in near resonance, most grains will be frozen for eternity by the nuclear bias field, even in the absence of an external bias (again, ignoring nuclear spin diffusion). Adding an external bias makes essentially no difference to this (since this bias is physically indistinguishable from the internal bias). In fact the bias will have no effect at all on (4.33) until $\xi_H \sim$ the energy scale of variation of $W(\varepsilon)$. For $\mu < 1$, this means that we will see oscillations in the decay rate as a function of $\xi_H$, with period $\omega_o$. For $\mu > 1$, the decay rate and decay function $A$ will change very little until $\xi_H \sim \omega_o\sqrt{N}$; for $\xi_H \gg \omega_o\sqrt{N}$ the relaxed fraction $A \to 0$ since not even the nuclei can bring the system into resonance. We derive also a useful formula for the bias and temperature effects by noting that only small $M \ll \sqrt{N}$ contribute to the sum in Eq.(4.23) and thus $W(M\omega_o/2 - \xi_H) \exp(-\beta(M\omega_o/2 - \xi_H)) \approx W(-\xi_H) \exp[\beta\xi_H]$ giving
\[ P(t; T; \xi_H) = P(t; \infty; 0) \frac{W(\xi_H) e^{\beta \xi_H}}{W(0) Z(\beta)} . \quad \text{(4.38)} \]

None of the results (4.30)-(4.38) is physical at very long time, because we have ignored the time variation of $\varepsilon$ caused by nuclear spin diffusion. Let us now demonstrate that nuclear spin diffusion is fast, so that equations (4.26)-(4.28) are justified. Notice first that since dipolar pairwise flips occur at a rate $T_2^{-1}$ for a given pair, and assuming short-ranged pair flips, we have roughly $NT_2^{-1}$ nuclear flips per second in the sample. Then in a time $\Delta M^{-1}$ we have $N(T_2 \Delta M)^{-1}$ such flips, and in this time the bias will change by $\delta \varepsilon \sim \delta \omega_o \sqrt{NT_2/\Delta M}$. The condition for fast diffusion is $\delta \varepsilon \gg \Delta_M$ (so the system has no time to tunnel), i.e., fast diffusion requires
\[ \Delta^3_M \ll \frac{N}{T_2 (\delta \omega_o)^2} . \quad \text{(4.39)} \]

If the spread $\delta \omega_o$ arises from the dipolar interactions between the same kind of nuclei only, then $\delta \omega_o \sim T_2^{-1}$ (but usually $\delta \omega_o \gg T_2^{-1}$, because of other nuclei and fields), then this criterion becomes $\Delta_M \ll T_2^{-1} N^{1/3}$. Now in fact $T_2^{-1}$ will be typical $10^3 - 10^6 Hz$ (depending on the isotopic concentration of nuclear spins, etc.). For any mesoscopic spin $\Delta_o < 10^6 Hz$ (at least for most bare Hamiltonians), so that even if (4.39) is violated for $M = 0$ (i.e., the zero polarisation change process), it is obeyed for large values of $M$. Since the bulk of the relaxation involves values of $M \sim N^{1/2}$, we see that (4.39) will always be the relevant condition. In this case Eqs.(4.23)-(4.29) follow immediately.

Let us now therefore evaluate $P(t)$ for the coupled grain/spin bath system, taking full account of the dipolar interactions between the spins, and assuming (4.30) to be satisfied; using (4.27)-(4.27), with $\lambda'$, this means we must evaluate, for $\mu > \Delta_o/\omega_o$
\[
P(t) - \frac{1}{2} = \frac{1}{2} \sum_M C_N^{(N+M-M_H)/2} e^{-\beta \omega_\alpha (M-M_H)/2} \frac{\lambda e^2}{2N Z(\beta)} \int_0^\infty dy e^{-y} \exp \left\{ -2 \frac{\tilde{\Delta}_o^2}{\pi^{1/2} \Gamma_\mu} t J_M^2 (2 \sqrt{\tilde{g} y}) \right\}.
\]

(4.40)

The integral is very easily evaluated; calling it \(I\), we have

\[
I \sim \int_0^\infty dy e^{-y} \exp \left\{ -2 \frac{\tilde{\Delta}_o^2 t}{\pi^{1/2} \Gamma_\mu} \left( \frac{\lambda e^2}{M^2} \right)^M \right\}
\]

\[
\sim \int_0^\infty dy e^{-y} \theta(y_M(t) - y),
\]

(4.41)

where the step-function \(\theta(y_M(t) - y)\), with

\[
y_M(t) = \frac{M^2}{\lambda e^2} \left( \frac{\lambda e^2}{M^2} \right)^{1/M},
\]

(4.42)

arises because values of \(M\) range up to roughly \(\sqrt{2N}\), and are thus typically large; thus

\[
P(t) - \frac{1}{2} \approx \frac{1}{2} \sum_M C_N^{(N+M-M_H)/2} e^{-\beta \omega_\alpha (M-M_H)/2} \frac{\lambda e^2}{2N Z(\beta)} \left\{ 1 - \exp \left[ -\frac{M^2}{\lambda e^2} \left( \frac{\lambda e^2}{M^2} \right)^{1/M} \right] \right\}.
\]

(4.43)

Noting again that if \(\mu > 1\), the dependence of \(P(t)\) on \(\xi_H\) in (4.43) will be rather small until \(\xi_H \sim \omega_o \sqrt{N}\), we begin by analysing this result for \(\xi_H = 0\). This zero bias case has of course been previously analyzed in our coherence papers \[\text{[4,5]}\], but there we were only interested in the very short time behaviour of \(P(t)\), i.e., that part involving frequencies \(\sim \Delta_o\), connected with possible coherent oscillations. Coherent behaviour can only arise in the \(M = 0\) polarisation sector of (4.30), which ignores spin diffusion.

However in analysing the quantum relaxation properties of \(\vec{S}\), we need to sum over all \(\sqrt{2N}\) important terms in (4.43); the effect of the spin diffusion will be to unblock the long-time relaxation. In evaluating (4.40) or (4.43), a steepest-descent integration over \(M\) is possible, but just as with the case of no spin diffusion, dimensional analysis is sufficient: the crossover time from unrelaxed to almost relaxed behaviour occurs at a time \(t_c\) given by

\[
\ln \left( \frac{2 \tilde{\Delta}_o^2}{\pi^{1/2} \Gamma_\mu} t_c \right) \sim \sqrt{2N} \ln \left( \frac{2N}{\lambda e^2} \right),
\]

(4.44)

(compare \[\text{[4.31]}\]), and the short-time relaxation is roughly logarithmic, as in \[\text{[4.32]}\], viz.,

\[
1 - P(t, \xi_H = 0) \sim \sqrt{\frac{1}{2\pi N}} \ln \frac{\ln \left( \frac{2 \tilde{\Delta}_o^2 t}{\pi^{1/2} \Gamma_\mu} \right)}{\ln \left( \frac{2 \tilde{\Delta}_o^2 t}{\pi^{1/2} \Gamma_\mu} \right)} ; \quad (t \ll t_c),
\]

(4.45)

The long-time behaviour is, analogously to \[\text{[4.33]}\], given by

\[
P(t, \xi_H = 0) - 1/2 \sim \exp \left\{ -\frac{3}{(2N)^{1/3}} \ln^{2/3} \left( \frac{2 \tilde{\Delta}_o^2 t}{\pi^{1/2} \Gamma_\mu} \right) \right\} ; \quad (t \gg t_c),
\]

(4.46)

with \(\ln \ln\) corrections in the exponent. Thus, amusingly, the results with spin diffusion included look just like those without spin diffusion, provided \(\mu = 0\) in the latter. The physical reason for this is simple - rapid spin diffusion basically eliminates the effects of degeneracy blocking, by allowing the bias \(\epsilon(t)\) to cover the whole energy range of each polarisation group. This is then the fundamental reason why spin diffusion "unlocks" \(\vec{S}\) and allows it to relax.

In Fig.7 we showed a plot of \(P(t)\) against \(\ln(2\Delta_o t)\), which clearly brings out the 3 relaxation regimes. For very short times \(\sim \Delta_o^{-1}\), there is a sudden relaxation involving only \(M \sim \lambda^{1/2}\) processes - the theory of this was given entirely in our coherence papers. For times \(t\) such that \(\Delta_o^{-1} \ll t \ll t_c\), we have roughly logarithmic relaxation. Then for \(t \gg t_c\), we have the behaviour in \[\text{[4.33]}\] or \[\text{[4.44]}\].

At this point it is important to realise that for all but microscopic spins, \(t_c\) will be astronomically long. From (4.44) we have

\[
t_c \sim \frac{\pi^{1/2} \Gamma_\mu}{2 \tilde{\Delta}_o} \left( \frac{2N}{\lambda e^2} \right)^{\sqrt{2N}}.
\]

(4.47)
Consider now, as in Fig. 7, the case of a mesoscopic spin where, e.g., \( \tilde{\Delta}_o \sim 1 \text{ MHz} \), \( \Gamma_p = 100 \text{ MHz} \), \( N = 1000 \), and \( \lambda = 10 \). We then find that \( t_c \sim 10^{39} \) s, i.e., more than \( 10^{39} \) times the age of the universe! Thus for mesoscopic grains we always get logarithmic relaxation at long times, from nuclear spins, in the quantum regime. Readers familiar with the experimental measurements of magnetic relaxation in grains may well be quite surprised at this result, since it is almost a central dogma of magnetism that logarithmic relaxation must arise from a distribution of energy barriers. Nevertheless we see that not only is this not so, but that the logarithmic relaxation has been derived here in a very general way, with essentially no approximations (but see below).

On the other hand for microscopic spins \( (S \sim 10) \), one finds that \( t_c \) can be short. For example, consider a situation where \( S = 20 \), \( \Omega_o = 10^{10} \text{ GHz} \), \( \tilde{\Delta}_o \sim 100 \text{ MHz} \), \( \Gamma_p 100 \text{ MHz} \), \( N = 10 \), and \( \lambda = 0.1 \) (this would roughly describe a particle containing 10 Tb atoms). Then one finds \( t_c \sim 1 \) s, and any direct relaxation experiment (typically conducted over time periods \( 1 \text{ ms} < t < 10^4 \text{ s} \)) would be mostly in the long-time relaxation regime.

These results are both surprising and interesting, in view of the very long time (often logarithmic) relaxation observed in experiments. However we immediately point out that the results (4.43)-(4.47) are by no means complete - although they are correct to within \( \ln \ln \) corrections for a coupled grain/spin bath system, a realistic calculation must incorporate all three of the relaxation mechanisms we have discussed, in Sections II, III, and this one. Since this changes the results yet again, we will not waste time here analysing the case of large bias (\( \xi_H > \omega_o \sqrt{N} \)) for the grain/spin bath system, but proceed directly to the general case.

V. COMBINING THE MECHANISMS - PHYSICAL RESULTS

We now come to the crux of the paper, which involves putting together the various mechanisms discussed in Sections II-V to give a physically realistic picture of quantum relaxation in magnetic particles. As mentioned in the introduction, we do not attempt a comprehensive discussion of all possible cases. Such a discussion would involve consideration of a large class of "giant spin" bare Hamiltonians, with widely varying behaviour, and with each being considered throughout the whole range of field strength, field orientation, and temperature. This would only be the beginning - we would then have to go on to examine the large variety of couplings to nuclear spins (with hyperfine couplings ranging over 3 orders of magnitude, plus quadrupolar couplings), the variety of different magnetoacoustic couplings, etc., etc.

We adopt the tactic here of showing how the mechanisms combine, and giving some details for one model. In other papers, in preparation, we analyse some specific experimental systems in much greater detail. We emphasize once again here that, in our view, magnetic relaxation in most experiments involves the dipolar interactions between grains in an essential way.

We begin by showing how phonons and nuclear spins combine to give a rather surprising form for the relaxation at low \( T \). This analysis is appropriate to the case where both grain and substrate are insulating. We then go on to include electrons - in this case phonons become irrelevant, and only the combination of electrons and nuclei are important.

Before discussing the details, let us first state the method we shall use. We have already seen how the dynamics of \( \vec{S} \) is given in the presence of a spin bath (Eqs. (4.25) or (4.40)). When we couple in a bath of oscillators which allow transitions between states \(| \vec{S}_1; \chi_1(\vec{\sigma}_k) \rangle \) and \(| \vec{S}_2; \chi_2(\vec{\sigma}_k) \rangle \) of the combined grain/spin bath system, in which there is no restriction on the difference \( 2M \) between the polarisations of the spin bath states \( \chi_1 \) or \( \chi_2 \), or on their biases \( \epsilon_1 \) and \( \epsilon_2 \) (apart from those imposed by energy conservation), the form of \( P(t) \) must change. In this paper we will not attempt to give a general expression covering all mechanisms simultaneously; this is more than we need. Instead we will use 2 expressions, which apply in the 2 limiting cases of interest. These are

(a) For short times, when the spin-mediate relaxation dominates; then we ignore the oscillator bath, and use Eq. (4.14).

(b) For longer times the oscillator bath-mediated relaxation will take over. In this regime, each time \( \vec{S} \) flips, some of the bias energy is taken up by an oscillator mode, and the rest by some number \( r \) of flipped spins in the spin bath. There is no restriction on either \( r \) or the polarisation change \( 2M \), for this process to work. The complete calculation of \( P(t) \) here is very complicated, but we notice that as soon as we reach times where the typical oscillator-mediated transition rate, at a typical bias, is faster than the spin bath-mediated transition rate (for those grains in an ensemble which have not yet relaxed), then a reasonable approximation for \( P(t) \) will be

\[
P(t; \xi_H) = \int d\epsilon \frac{W(\epsilon - \xi_H)}{Z(\beta)} e^{-\beta(\epsilon - \xi_H)} \left\{ P^{(eq)}(T, \epsilon) + \left[ 1 - P^{(eq)}(T, \epsilon) \right] e^{-t/\tau(\epsilon, T)} \right\} ,
\]

(5.1)
where $\tau^{-1}(\epsilon, T)$ is the oscillator-mediated relaxation rate, and $P^{eq}(T, \epsilon) = e^{-\epsilon/T}/(2\cosh(\epsilon/T))$ is the equilibrium population of the state $\hat{S}_1$ in a given bias (in our previous discussion of the nuclear spin effects we assumed that this bias was actually much less than $T$). We note that this expression is incoherent, because it applies to the majority of grains in a large bias, which cannot relax via the spin bath as discussed in the previous Section. The relaxation rate in this incoherent case can be calculated as a second order perturbation theory expression in the tunneling amplitude $\Delta_o$. Thus the only effect of the spin bath which is left is the distribution over the bias.

The formal proof of the above statement is easy to follow for the case of pure orthogonality blocking. In a typical bias $\epsilon \sim \omega_o N^{1/2} \gg \Delta_o$ the second order perturbation theory expression for the tunneling rate is given by

\[
\tau^{-1}(\epsilon) = 2\pi \sum_{ij} \rho_{i}^{eq} |U_{ij}|^2 \sum_{\alpha\gamma} \rho_{\alpha}^{eq} |V_{\alpha\gamma}^{(ph)}|^2 \delta(\epsilon + E_\alpha - E_\gamma + \omega_o(M_i - M_f)/2),
\]

where the sums are over the initial and final states of the spin bath ($i, f$) and oscillator bath ($\alpha, \gamma$) with the equilibrium density matrices $\rho^{eq}$. Here $V^{(ph)}$ describes the interaction with the phonons (we do not even need its explicit form to prove the point), and the orthogonality rotation operator $U$ is given by

\[
U = \prod_{k=1}^N e^{-i\beta_k \hat{\sigma}_k^y}.
\]

The crucial point here is that in a large bias one may drop the energy $\omega_o(M_i - M_f)/2$ transferred to the spin bath from the $\delta$-function argument in (5.2), because only transitions for which $M_i - M_f \ll N^{1/2}$ contribute to the answer. After that we have the sum over the complete set of states $f$, so that $U_{ij}U_{ij}^\dagger \equiv 1$ because the of the unitarity of $U$, and the final answer is that for the oscillator bath alone. Including topological decoherence changes the result in a minor way. Indeed, with nonzero $\alpha_k$, we have

\[
U = \prod_{k=1}^N e^{-i\beta_k \hat{\sigma}_k^y} \left( e^{i\Phi+i\alpha_k \hat{n}_k \hat{\sigma}_k} + e^{-i\Phi-i\alpha_k \hat{n}_k \hat{\sigma}_k} \right),
\]

and the result for the relaxation rate in the oscillator bath is renormalized by the factor

\[
D = \langle UU^\dagger \rangle = 2 + 2\cos(2\Phi)F_\lambda(1),
\]

Unless $\lambda = 0$ and the topological phase $\Phi$ is a multiple of $\pi$, this factor is of order $\sim 1$, and clearly has no essential effect on the results. Thus, apart from the renormalisation factor $D$, the relaxation of a single grain in a nuclear bias field $\epsilon$ is that already given in sections II and III, and all that remains for a grain ensemble is to average over the bias field.

### A. Spin Bath Plus Phonons

The case of insulating grains and an insulating substrate is the one where there is most obvious competition and interplay, between the nuclear spin and phonon relaxation mechanisms. They are both slow. We have already seen (Fig.6, and Eq. (4.14)) what the short-time relaxation will look like. Now consider the implications of (5.1) for the long-time relaxation. We worked out $\tau^{-1}(\epsilon)$ for phonon relaxation in the absence of nuclear spins; for a longitudinal bias this was given by (3.13), and will be roughly the same in the presence of the spin bath (i.e., with the renormalisation factor (5.5) above). Thus we write $\tau^{-1}(\epsilon, T)$ in the form

\[
\tau^{-1}(\epsilon, T) = \tau^{-1}_o \left( \frac{\epsilon}{E_o} \right)^3 \coth(\epsilon/2k_BT),
\]

where $\tau^{-1}_o$ is a "typical" normalising relaxation rate, defined as

\[
\tau^{-1}_o = \tau^{-1}(\epsilon = E_o, k_B T = E_o) \approx S^2 \Delta_o \left( \frac{\Delta_o}{\Theta_D} \right) \left( \frac{E_o}{\Theta_D} \right)^3,
\]

and $E_o$ is the width of the Gaussian peak in $W(\epsilon)$, i.e.,
\[ E_\omega = \omega_0 N^{1/2}/2. \]  

(5.8)

Notice that for a fixed bias \( \epsilon = E_\omega \), the actual relaxation rate \( \tau^{-1}(E_\omega, T) \) still depends on temperature as \( \tau^{-1}(\epsilon, T) = \tau_0^{-1} \coth(E_\omega/2k_BT) \). At low \( T \) this scales down to the constant value \( \tau_0^{-1} \), whereas at high \( T \) it increases linearly, i.e.,
\[ \tau^{-1}(E_\omega, T) = 2\tau_0^{-1}k_BT/E_\omega. \]

It is useful to get some preliminary idea of typical time scales for \( \tau_\omega \). Let us consider 3 examples, viz., (i) a Tb Oxide grain, with \( S \sim 1000 \), containing \( N \sim 1000 \) nuclei; (ii) a particle of Er As (such particles are apparently insulating \cite{39}, containing \( N \sim 1000 \) nuclei, with \( S \sim 1000 \); and (iii) Ni O grains with \( S \sim 1000 \), but \( N \sim 10 \) only (coming from the 1\% of Ni\(^{59} \) nuclear spins). Let us also assume that \( \Theta_D \sim 100 \) K and \( \Delta_\omega \sim 1 \) MHz for each example. However \( E_\omega \) varies dramatically - for the Tb O grain, \( \omega_o \sim 5 \) GHz, and \( E_\omega \sim 4 \) K; for the Er As grain, \( \omega_o \sim 1 \) GHz, and \( E_\omega \sim 0.8 \) K; whilst for the Ni O grain, \( \omega_o \sim 28 \) MHz, and \( E_\omega \sim 2.3 \times 10^{-3} \) K. We thus get a wide range of time scales: for the Tb O grains one has \( \tau_\omega \sim 1.6 \times 10^{-2} \) s, for the Er As grains, one has \( \tau_\omega \sim 5 \) s, whilst for the Ni O grains, one has \( \tau_\omega \sim 4.5 \) years!

We now consider the general behaviour of \( P(t; T; \xi_H) \). We start by noticing that the weighting function for the initial states, of bias \( \epsilon \), contributing to (5.1), will be

\[ w(\epsilon, T; \xi_H) = W(\epsilon - \xi_H) \frac{e^{-\beta(\epsilon - \xi_H)}}{Z(\beta)} \sim \frac{1}{\sqrt{2\pi E_\omega}} \exp \left\{ -\frac{1}{2} \left[ \frac{\epsilon - \xi_H}{E_\omega} + \frac{E_\omega}{k_BT} \right] \right\} . \]

(5.9)

Ignoring \( \xi_H \) for the moment, we see that in zero external field this distribution peaks at negative bias \( \epsilon \sim -E^2_\omega/k_BT \). This is natural; as we lower \( T \), more and more of the nuclei align with \( \vec{S} \), thereby lowering the energy of the combined system in the initial state. This process continues until \( \sim N \) nuclei are aligned with \( \vec{S} \), i.e., when \( \epsilon \sim -N\omega_o/2 \), at a temperature \( k_BT \sim \omega_o/2 \). We have then reached the bottom edge of the distribution \( W(\epsilon) \), way out in the wings. If \( \xi_H \neq 0 \), the distribution peaks at \( \epsilon \sim (\xi_H - E^2_\omega/k_BT) \).

Thus we write

\[ P(t; T; \xi_H) = \int d\epsilon w(\epsilon, T; \xi_H) \left\{ P^{(eq)}(T, \epsilon) + \left[ 1 - P^{(eq)}(T, \epsilon) \right] e^{-t/\tau(\epsilon, T)} \right\} \]

\[ = \int dx \frac{1}{\sqrt{2\pi}} e^{\frac{-1}{2} \left( x - \xi_H + 1/T \right)^2} e^{x/T} e^{-t/(t/\tau_o)} \frac{x^3 \cosh(x/2T)}{2 \cosh(x/T)} , \]

(5.10)

where we have normalised all energies by \( E_\omega \):

\[ x = \epsilon/E_\omega \]
\[ \xi_H = \xi_H/E_\omega \]
\[ T = k_BT/E_\omega . \]

(5.11)

Let us first look at the high-\( T \) limit of (5.10), i.e., for \( k_BT \gg E_\omega \) when \( T \gg 1 \). Then (5.10) is easily evaluated; one finds

\[ P(t, T; \xi_H) - 1/2 \sim \frac{1}{2} \left( 1 + 4t/\tau_o \right)^{1/2} \exp \left\{ -\frac{\xi_H^2}{2T(t/\tau_o)} \right\} \]

(5.12)

\[ \rightarrow \frac{1}{2} \left( 1 + 4t/\tau_o \right)^{1/2} \exp \left\{ -\frac{2t}{\tau_o} \right\} . \]

(5.13)

(We recall that this result is correct only when \( T \ll \Omega_o \) - otherwise higher levels of the grain will come into play.)

This surprising power-law behaviour is actually easily understood. The initial fast relaxation (at \( t \ll \tau_o \)) comes from grains with large bias; the slower relaxation (\( t \gg \tau_o \)) comes from the grains with smaller bias. The typical relaxation occurs at a rate \( T\tau_o^{-1} \), i.e., it is faster for higher temperatures, in accordance with the remarks just after Eq. (5.8). We may think of the power law as a “grain-ensemble” sum of a lot of different exponential decays, or as a funny kind of "stretched exponential". Note however that the decay is not logarithmic in time.

Now consider the low-\( T \) limit, with \( T \ll 1 \); this is a little more complicated. From (5.10) we now have
\[ P(t, T; \xi_H) \approx 1 + \int \frac{dx}{\sqrt{2\pi}} e^{-\frac{x^2}{2}} \left( e^{x/T} \frac{e^{x/T}}{2 \cosh(x/T)} \left( e^{-x^3 \coth(x/2T)t/\tau_o} - 1 \right) \right) \]

\[ = 1 + e^{\xi_H/T - 1/2 T^2} \int \frac{dx}{\sqrt{2\pi}} \frac{e^{-x^3 \coth(x/2T)t/\tau_o}}{2 \cosh(x/T)} \left( e^{-x^3 \coth(x/2T)t/\tau_o} - 1 \right) , \] \hspace{1cm} (5.14)

Let us consider first the behaviour of (5.14) when \( \xi_H = 0 \). Using the fact that \( T \ll 1 \) one may further simplify this expression to

\[ P(t, T; \xi_H = 0) \approx 1 + \frac{T}{\sqrt{2\pi}} e^{-1/2 T^2} \left[ \int_0^\infty \frac{dx}{\cosh(x)} e^{-x^3 \coth(x/2T) t/\tau_o} - \frac{\pi}{2} \right] , \] \hspace{1cm} (5.15)

This clearly defines a temperature-dependent relaxation rate

\[ \tau^{-1}_{\text{eff}}(T) = T^3 \tau_o^{-1} \sim S^2 \Delta_o \left( \frac{\Delta_o}{\Theta_D} \right) \left( \frac{k_B T}{\Theta_D} \right)^3 , \] \hspace{1cm} (5.16)

which goes to zero at low \( T \). We note however that the validity of (5.15) requires that the calculation is not affected by the edges of \( W(\epsilon) \), which means that at a temperature \( k_B T \sim \omega_o/2 \), Eq.(5.16) crosses over to a constant value, i.e.,

\[ \tau^{-1}_{\text{eff}}(T \to 0) \to N^{3/2} \tau_o^{-1} \sim S^2 \Delta_o \left( \frac{\Delta_o}{\Theta_D} \right) \left( \frac{\omega_o}{2 \Theta_D} \right)^3 . \] \hspace{1cm} (5.17)

but the magnitude of the relaxing component at this temperature is already extremely small; in fact \( 1 - P(t \to \infty) \sim N^{-1/2} e^{-N} \). It is again useful to consider what is the the maximum relaxation time at low \( T \). For the examples previously mentioned, we have (i) for the Tb O grain, \( \tau_{\text{eff}}(T \to 0) \sim 10 \text{ minutes} \); (ii) for the Er As grain, \( \tau_{\text{eff}}(T \to 0) \sim 2 \text{ days} \); and (iii) for the Ni O grain, \( \tau_{\text{eff}}(T \to 0) \sim 4 \times 10^7 \text{ years} \).

Defining the exponentially small total amplitude of the relaxing component as

\[ A \approx \frac{T \sqrt{\pi}}{2 \sqrt{2}} e^{-1/2 T^2} , \] \hspace{1cm} (5.18)

we find the long time asymptotics of \( P(t) \) as

\[ P(t, T; \xi_H = 0) \to 1 - A + A \left( \frac{2 \tau_{\text{eff}}(T)}{\pi t} \right)^{1/2} . \] \hspace{1cm} (5.19)

The reason for the \( T^3 \) decrease in the relaxation rate at low \( T \) is simply that as we lower the temperature, the typical bias energy decreases linearly with \( T \), because only a small fraction of grains in the Gaussian tail which have very small bias \( \epsilon \sim k_B T \) contribute to the relaxation. The majority of the grains are actually “trapped” by the hyperfine interaction in a negative bias energy \( \epsilon \sim E^2_0/k_B T \gg k_B T \). Thus at this low temperature even phonons will not help to liberate the giant spin.

Consider now Eq.(5.14) with nonzero external bias. For \( \xi_H \ll 1/T \) the previous answer (5.13) hardly changes except that the amplitude of the relaxing component is given now by

\[ A \approx \frac{T \sqrt{\pi}}{2 \sqrt{2}} e^{-1/2 (\xi_H^2 - 1/2 T^2)} , \hspace{1cm} (\xi_H \ll 1/T) . \] \hspace{1cm} (5.20)

We observe that already for rather small bias \( \xi_H \sim T \ll 1 \) we have an exponential dependence on \( \xi_H \). The negative bias will simply further suppress an already exponentially small fraction of relaxing grains and we shall not go into more details here. The case of positive \( \xi_H \) is much more intriguing and surprising. After an exponential increase in the amplitude up to \( A \sim 1 \) for \( \xi_H \sim 1/T \), the answer changes drastically. For \( \xi_H > 1/T \) one finds the magnetisation function to be

\[ P(t, T; \xi_H) \approx \int \frac{dx}{\sqrt{2\pi}} e^{-\frac{1}{2} (x-\xi_H+1/T)^2} e^{-|x|^3 t/\tau_o} , \hspace{1cm} (\xi_H > 1/T) , \] \hspace{1cm} (5.21)

which has different behaviour depending on whether we look at short times, where
\[
P(t; T; \xi_H) \approx e^{-t/\tau_{eff}(\xi_H; T)}, \quad (t/\tau_{eff} < (\xi_H - 1/T)^2), \tag{5.22}
\]
\[
\tau_{eff}^{-1}(\xi_H, T) = (\xi_H - 1/T)^3 \tau_0^{-1}; \tag{5.23}
\]

or long times, where
\[
P(t; T; \xi_H) \sim \frac{\sqrt{2} \Gamma(1/3)}{3\sqrt{\pi}} e^{-(\xi_H - 1/T)^2/2} \left(\frac{\tau_0}{t}\right)^{1/3}; \quad (t/\tau_{eff} > (\xi_H - 1/T)^2). \tag{5.24}
\]

Note that Eqs. (5.21)-(5.24) are derived in the approximation that \(\xi_H - 1/T \gg 1\). As before, the bias is restricted to be much less than \(\Omega_\alpha\). Finally, at \(t > \tau_0\), this behaviour changes yet again to \(P(t) \approx \exp(- (\xi_H - 1/T)^2/2) (\tau_0/4tT)^{1/2}\). We note the anomalous temperature and bias dependence of the effective relaxation rate (5.23) near the crossover.

From this analysis we conclude that unless the external bias is larger than \(E_0\) (or \(E_0^2/k_B T\)) at low temperature the ensemble averaged results for the magnetization relaxation are not described by the naive theory of a two level system coupled to a bath of oscillators, and spin bath interactions essentially modify the answer. It is worth noting three effects which have to be kept in mind when considering the evolution of the experimental data in external bias:

(i) Since the dependence on \(\xi_H\) starts when \(\xi_H > E_0\) or \(\xi_H > E_0^2/k_B T\), one cannot derive the value of the tunneling amplitude from this dependence, as it would be in the case of isolated two-level system;

(ii) At low temperature the giant spin is ”trapped” in its initial state by the large and negative bias produced by the spin bath.

(iii) Only large and positive bias can liberate the giant spin and to allow complete magnetisation relaxation. The temperature and bias dependence of the relaxation rate is anomalous near the crossover \(\xi_H \sim 1/T\). Of course, when \(\xi_H \gg 1/T\) we recover back the pure case of oscillator bath relaxation.

At this point we would like to comment on our starting assumption that the initial state of the spin bath is equilibrated with the giant spin direction \(\vec{S} = \vec{S}_1\). Experimentally this could be arranged by applying a very strong negative bias \(\xi_H\) during a time period much longer than the longitudinal NMR relaxation time \(T_1\), and switching it off at \(t = 0\). In some systems however this procedure may not work because of an astronomically long \(T_1\) (in fact, low temperature longitudinal NMR is still something of an unsolved mystery; spin-lattice relaxation times at \(mK\) temperatures ought to be many years in insulating crystals, unless some gapless magnetic excitations are involved, e.g., on the sample surface or crystal defects). It may be useful then to introduce two different temperatures - one for the oscillator bath \(T_{ph}\), and the other for the spin bath \(T_S\) (which may even be negative, i.e., \(T_S < 0\)). Our basic equation (5.1) is still valid in this more general case, but now the distribution over the initial bias is defined by the spin bath temperature, whereas the giant spin evolution toward equilibrium is governed by the crystal temperature, i.e.,

\[
P(t; \xi_H) = \int d\epsilon \frac{W(\epsilon - \xi_H)}{Z(T_S)} e^{-(\epsilon - \xi_H)/T_S} \left\{ P^{(eq)}(T_{ph}, \epsilon) + [1 - P^{(eq)}(T_{ph}, \epsilon)] e^{-t/\tau(\epsilon; T_{ph})} \right\}. \tag{5.25}
\]

One may proceed with the analysis of this expression as before. At this point we feel that considering more cases in this paper will not add much to the physical picture. Depending on the particular experimental system and sample preparation, the necessary formulae can be easily derived from (5.24).

Summarising what we have found for insulating systems, we see that at short times, the relaxation proceeds entirely via the nuclear spin bath, and is logarithmic in time (Eq. (4.43)). At longer times phonons take over, and we get power law decay in time; at low \(T\) this goes as \((\tau_{eff}(T)/t)^{1/2}\), with \(\tau_{eff}^{-1} \sim T^3\) (Eqs. (5.16) and (5.19)), but only a small fraction of grains relax unless the bias \(\xi_H \sim E_0/k_B T\) or larger. This bias is necessary to counteract the nuclear bias field. At higher temperatures we still get power-law relaxation (Eq. (1.13)). The crossover between nuclear spin-mediated and phonon-mediated relaxation can be understood by matching Eq. (4.43) with the relevant phonon expression.

### B. Spin Bath Plus Electrons

We now turn to the case where either the grain or the substrate is conducting. We use the giant Kondo model of section 4 to describe interaction of \(S\) with electrons, with a dimensionless coupling \(\alpha\). If both grain and substrate are conducting, then \(\alpha = \alpha_s \sim g^2 S_{Q}^{4/3}\) (Eq. (2.17)); if only the substrate conducts, then \(\alpha = \alpha_s \sim g^2 S_{Q}^{2/3}\) (Eq. (2.13)); and if only the grain conducts, \(\alpha \sim \alpha_s e^{-\Delta_p/T}\) (Eq. (2.21)).
The formal analysis is almost identical to that just used for phonons - we start again from (5.1), now using the general result \( \Delta_\mu \) for the electronic relaxation rate. The high temperature limit is most transparent because \( \tau_e(T) \) in Eq. (2.3) is independent of the bias. Thus we find a pure exponential relaxation

\[
P(t) = 1/2(1 + e^{-t/\tau_e(T)\epsilon}) ; \quad (\alpha T, T \gg E_o) ;
\]

(5.26)

at high temperature.

For very small grains or conducting grains on an insulating substrate at \( T \ll \Delta_\mu \), we have a peculiar relaxation regime when the internal bias is much less than temperature, but the electronic damping rate \( \Gamma_e = 2\pi \alpha T \) is already small, that is for \( \alpha \ll 1 \) there is the temperature range where \( \Gamma \ll E_o \ll T \). Now the electronic relaxation rate is inversely proportional to the bias energy

\[
\tau^{-1}_e(T, \epsilon) = 8\pi \alpha T \frac{\Delta^2}{\epsilon^2} , \quad (2\pi T \ll \epsilon \ll T)
\]

(5.27)

Substituting this expression to (5.1) we find the time correlation function as

\[
P(t, T; \xi_H) \approx 1/2 + 1/2 \int \frac{dx}{\sqrt{2\pi}} e^{-\frac{1}{2}(x-\xi_H)^2} e^{-t/(x^2 \tau_e(T;E_o))} ,
\]

(5.28)

which after easy evaluation yields

\[
P(t) \approx 1/2(1 + e^{-t/\tau_e(T;E_o)}) ; \quad (\xi_H = 0)
\]

(5.29)

\[
\approx 1/2(1 + e^{-t/\tau_e(T;\xi_H)}) ; \quad (\xi_H \gg E_o) .
\]

(5.30)

The temperature dependence of the effective relaxation rate is \( \sim T \alpha(T) \), and again in the small external bias we have an unusual decay law - it is neither simple exponential nor power-law.

Let us deal now with the low-temperature behaviour, \( k_B T/E_o \ll 1 \). Using (2.10) we then get

\[
P(t, T; \xi_H) \approx 1 + \int \frac{dx}{\sqrt{2\pi}} e^{-\frac{1}{2}(x-\xi_H+1/T)^2} \frac{e^{x/T}}{2\cosh(x/T)} \left( e^{-xt/x^2 \tau_{eff}} - 1 \right) ,
\]

(5.31)

\[
f(x) = \cosh(x) \vert \Gamma[\alpha + ix/\pi] \vert^2
\]

(compare [5.14]). If the bias \( \xi_H < 1/T \), this defines a temperature dependent relaxation rate

\[
\tau_{eff}(T) = \frac{2}{\Gamma[2\alpha]} \frac{\Delta_o}{\Omega_o} \left( \frac{2\pi T}{\Omega_o} \right)^{2\alpha - 1} .
\]

(5.32)

The answers crucially depend on the parameter \( \alpha \). We consider three limiting cases:

(i) For \( \alpha \ll 1 \) we approximate the function \( f(x) \) by \( f(x) \approx x \coth(x)(\alpha^2 + (x/\pi)^2)^{-1} \) and obtain

\[
P(t, T; \xi_H) \approx 1 - A \frac{\pi t}{\alpha \tau_{eff}} ; \quad (t \ll \alpha^2 \tau_{eff}) ,
\]

(5.33)

\[
P(t, T; \xi_H) \approx 1 - 2A \left( \frac{\pi t}{\tau_{eff}} \right)^{1/2} ; \quad (\alpha^2 \tau_{eff} < t < \tau_{eff}) ,
\]

(5.34)

\[
P(t, T; \xi_H) \approx 1 - A + A \left( \frac{t}{\tau_{eff}} \right)^{1/4} e^{-2\pi \sqrt{t/\tau_{eff}}} ; \quad (\tau_{eff} \gg t) .
\]

(5.35)

(ii) For \( \alpha = 1/2 \) we have \( f(x) = \pi \), and, as in the high temperature limit, the relaxation is given by the simple exponential law

\[
P(t) = 1 - A(1 - e^{-\pi t/\tau_{eff}}) ;
\]

(5.36)

(iii) Finally we consider the case of large \( \alpha \), which has a power-law asymptotic at long times

\[
P(t, T; \xi_H) \approx 1 - A + 2A \left( \frac{2\alpha}{2\alpha - 1} \right) \left( \frac{\tau_{eff}}{\pi t} \right)^{1/(2\alpha - 1)} ,
\]

(5.37)
In all these expressions \( A(T, \xi_H) \) is given by (5.20).

The interpretation of these results also depends on the value of \( \alpha \). If both grain and substrate are conducting, then the relaxation time in (5.32) will be greater than the age of the universe unless \( S \leq 100 \) (depending on the value of \( y^2 \)). This shows the astonishing power of the electron bath to "freeze" the dynamics of \( \vec{S} \), unless it is quite microscopic in size. We shall not analyze here the case of very small \( S \), since once \( S \sim O(10 - 100) \), the detailed structure of \( W(\epsilon) \) will no longer be Gaussian - there will be lots of fine structure, depending on the particular system involved. Such studies are best done on a case-by-case basis.

If only the substrate is conducting, then \( \alpha = \alpha_s \) will be small until \( S \sim 10^3 \). For larger \( S \) then the giant spin is again frozen. For \( S \leq 10^3 \) the results (5.33)-(5.36) can be applied if \( \xi_H \ll 1/T \). Again, as \( \xi_H \) approaches \( 1/T \), there will be an amusing crossover , with a relaxation rate given for finite \( \xi_H > 1/T \) by

\[
\tau_{eff}^{-1}(T, \xi_H) = \tau_e^{-1}(T, \epsilon = \xi_H - E_0^2/T) ,
\]

(5.38)

Finally, if only the grain is conducting, \( \alpha \) can be exponentially small, if the spacing \( \Delta \mu \) between the internal electron levels is large enough - this typically requires that \( \Delta \mu \gtrsim \Omega_o \), so that \( S \leq 10^5 \) (cf. Eq. (2.23)). In this case, unlike the cases where the substrate is conducting, the short-time relaxation will be again dominated by nuclear spin-mediated transitions, and the weak electron-mediated transitions only enter at later times in possible competition with the phonon-mediated transitions; the typical electron-mediated rate becomes

\[
\tau_{eff}^{-1}(T) = 4\pi \alpha_o \frac{\Delta^2}{E_0^2} e^{-\Delta \mu/T} ,
\]

(5.39)

Note that for low enough temperature, \( \tau_{eff} \) in (5.39) becomes so long that electrons in the grain become irrelevant, and we return to the calculations for the nuclear spin/phonon-mediated problem, i.e., the grain behaves as an insulator.

Thus to summarize, we see that depending on whether the grain and/or substrate are conducting, we get behaviour ranging from grains frozen for all eternity (if \( \alpha \gg 1 \)), to grains behaving essentially as insulators, with short-time relaxation controlled by the nuclei (when \( \alpha \rightarrow 0 \)). In the former case we see an extraordinary alliance between the electrons (with their strong dissipative suppression of tunneling) and nuclear spins (which trap \( \vec{S} \) in a negative bias \( \epsilon \sim -E_0^2/T \)) to block any motion of \( \vec{S} \) even at low \( T \), where traditionally one expects tunneling. In fact, as \( T \rightarrow 0 \), our calculations show that \( \vec{S} \) is blocked for eternity when \( S \) takes microscopic values (e.g., \( S \sim O(10) \)), because the negative bias becomes very large. This is a quite astonishing demonstration of the control that nuclear spins exert over the dynamics of \( \vec{S} \) - the spin is only unblocked when \( S \sim O(1) \), when \( \alpha \ll 1 \) again (and the whole WKB framework breaks down!).

VI. SUMMARY & CONCLUSIONS: PRACTICAL IMPLICATIONS

Let us first summarize our main results. At temperatures \( T \) such that \( k_B T \ll \Omega_o \), we may employ a 2-level model with bias to describe the grain. Coupling to electrons and/or phonons in this model can be treated by the usual oscillator bath methods. However the crucial coupling turns out to be to the nuclear spin bath, to which oscillator bath models do not apply. The main effect of this coupling is to spread each of the 2 levels for \( \vec{S} \) into a Gaussian "band" of half-width \( E_0 \sim N^{1/2} \omega_o \), and total width \( N \omega_o \), where \( \omega_o \) is the hyperfine coupling and \( N \) the number of nuclei in the grain. If \( N \) and \( \omega_o \) are small, \( E_o \) may only be a few \( mK \) (as in Ni or Fe grains). If \( N \) and \( \omega_o \) are large (as in mesoscopic rare earth grains), \( E_o \) may be hundreds of Kelvin or more (so that \( E_o \gg \Omega_o \)). At low temperatures the nuclei begin to line up with the grain vector \( \vec{S} \), and then \( \vec{S} \) finds itself in a negative internal nuclear bias field, with a mean bias \( \epsilon \) of roughly \( \epsilon \sim -E_0^2/T \). Then at short times the only way that the grain can relax is by taking energy from the nuclear system. The inter-nuclear dipolar interactions allow this to happen, by causing the bias \( \epsilon \) to become time-dependent - the result is a slow relaxation, which for an ensemble of grains gives a fraction of relaxed grains going roughly logarithmically in time (section VI):

\[
1 - P(t) \sim \sqrt{\frac{1}{2\pi N}} \frac{\ln \left( \frac{2\Delta^2 t/\pi^{1/2} \Gamma_o}{\epsilon} \right)}{\ln \left( \frac{1}{\epsilon} \ln \left( \frac{2\Delta^2 t/\pi^{1/2} \Gamma_o}{\epsilon} \right) \right)} .
\]

(6.1)

This logarithmic relaxation, shown in Fig.7, has nothing to do with a distribution of sizes of the grains (we assume all grains have the same parameters).
At longer times the grains can relax very slowly via either electron- or phonon-mediated transitions, again in the time-varying nuclear field. If the grain is insulating (no electrons), we get power-law relaxation; a fraction $A$ of grains can relax, where

$$A \approx \sqrt{2\pi/2E_o} \exp \left\{-1/2 \left( \frac{\xi_H}{E_o} - \frac{E_o}{T} \right)^2 \right\},$$

(6.2)
in a bias field $\xi_H$, and these grains relax as $A(\tau_{\text{eff}}/t)^{1/2}$ where

$$\tau_{\text{eff}}^{-1}(T) \sim S^2 \Delta_o \left( \frac{\Delta_o}{\Theta_D} \right)^3 \left( \frac{k_B T}{\Theta_D} \right)^3,$$

(6.3)
if $\xi_H \ll E_o^2/T$, and $P(t) \sim 1/2(1 + e^{-t/\tau_{\text{eff}}})$ with

$$\tau_{\text{eff}}^{-1}(\xi_H, T) = \left( \frac{\xi_H}{E_o} - \frac{E_o}{T} \right)^3 S^2 \Delta_o \left( \frac{\Delta_o}{\Theta_D} \right)^3 \left( \frac{E_o}{\Theta_D} \right)^3,$$

(6.4)
once $\xi_H > E_o^2/T$. Thus very few grains can relax at until the external bias $\xi_H$ compensates the internal bias $\epsilon$ and "untraps" $\vec{S}$.

If there are electrons around, the results depend on whether the substrate is conducting or not. If it is, then all but microscopic spins ($S \ll 10^5$) will be trapped in states of frozen magnetisation for astronomical times, once the temperature goes below $\Omega_o$. This remarkable result is a combination of "degeneracy blocking" caused by the nuclear bias field, and the very strong electronic dissipation. However if only the grain is conducting, and the substrate is insulating, then for $S \ll 10^5$, the electronic relaxation becomes negligible, and we go back to the fully insulating case; the full results for all values of electronic coupling appear in section VII.B.

It is clear that these results have implications both for future magnetic device design, and for the thousands of experiments that have been done on magnetic grain relaxation over the years. As far as devices are concerned, perhaps the most interesting result is the freezing of $S$ when both grain and substrate are conducting unless $S \ll 10^5$. This may have far-reaching implications for future computers and information storage, since it implies that if we are prepared to go to low temperatures, one should be able to use even magnetic molecules as permanent memory storage elements, perhaps of the "M.R.A.M." type (cf. ref. [31]). Even more tempting is the possibility that we may be able to manipulate $\vec{S}$ in such molecules indirectly, by controlling the nuclear spin polarisation, since it is the nuclear bias field that controls the dynamics of $\vec{S}$.

As far as experiments on grain relaxation are concerned, we believe that it is quite urgent that experimental tests of our results be done at low $T$. Unfortunately most of the many relaxation measurements that have already been done are at higher temperatures, where relaxation is dominated by thermal activation. What we have found here is that the low-$T$ behaviour does not look like the conventional tunneling picture at all - there is no crossover to a temperature-independent "tunneling relaxation" at temperatures $k_B T \ll \Omega_o$. The low-$T$ behaviour of magnetic particles is not governed by the external bias at all, unless it is very large; instead it is governed by the random distribution of internal nuclear bias fields. This renders previous theories of the tunneling of magnetic particles somewhat meaningless, regardless of the external field. Thus we believe that it would be useful to look again at those experiments that have been done at low $T$ on ensembles of relaxing grains [1][2][3][4][13][20], particularly in an attempt to understand the "plateau" that often appears at low $T$ in the relaxation rate (the "magnetic viscosity plateau").

However an important cautionary note is necessary. In virtually all of these experiments, inter-grain interactions (mediated by the grain dipolar fields) are very important. We believe this to be the reason for the common occurrence of "avalanche" magnetisation reversal in grain ensembles (see, e.g., ref. [22]); these avalanches are prima facie evidence for the importance of inter-grain interactions (such avalanches also occur in multi-domain magnets, because of dipolar interactions between the domains and walls [3][22]). Thus to interpret most experiments (including the remarkable recent results on Mn$_{12}$O$_{12}$ molecules [22]), we must incorporate these interactions - this we do elsewhere [23].

However some experiments may not suffer this problem. The recent results of Coppinger et al. [30], on ErAs grains, with $S \sim 10^3$, may be a case in point. It is not yet clear what is the effective coupling to the electron bath for this system, but we believe that the theory in the present paper should be applicable to this case.

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for $g$ along the weak-scattering direction. Since the $J$ dynamics is governed by the transverse orthogonality. The relevant renormalization group variable is fugacity $\Delta g$ scaling of $J$ the renormalization group equation for the channels are equally coupled to the giant spin, the Anderson orthogonality [6] term where $\delta$ correct expression for the giant spin relaxation time will be neglect conduction electron spin flips or not depends on the parameter (the solution of this equation is explicitly used in our equations (2.8)-(2.10)). Thus the question of whether we may

correct expression for the giant spin relaxation time will be

$$\tau^{-1} \approx 2\pi |J_{\parallel}|^2 N(0) \left(\frac{2\pi T_c}{\epsilon_{\text{max}}}\right)^{2\alpha_s - \theta}, \quad \frac{\delta_l + \delta_\nu}{\pi} = \theta$$

Note that for $\theta > 1$ we definitely have $\alpha_s \sim S^{2/3} \gg 1$, so the $\vec{S}$ dynamics are definitely incoherent, and the above refinement is hardly different from (2.8)-(2.10) (it is equivalent to going from $2\alpha_s - 1$ to $2\alpha_s - \theta$).

FIGURE CAPTIONS
Figure 1 The Atomic Weight (in Daltons) of a typical computer memory element, as a function of time. Note that the position of the ”Quantum Threshold” depends on temperature. As we see in this article, it also depends strongly on what the element is made of.

Figure 2 Schematic diagram of the energy scales in the giant spin, biased by a longitudinal field $H_z$. The bias $\epsilon = \gamma S H_z$, and $\Omega_o$ is the gap to the next set of excited states. In reality, for a giant spin, the top of the barrier will be much higher, at energy $\sim (S \Omega_o)$ above the lowest states.

Figure 3 The distribution function $W(\epsilon)$ for spin bath levels, around each giant spin level. Typically the parameter $\mu > 1$ except for very small numbers of nuclear spins (see text).

Figure 4 Some possible transitions when $\vec{S}$ flips, for a grain in zero external field. The initial state of the combined giant spin/nuclear spin system has energy $\epsilon_1$; in the diagram this state $|1\rangle$ has either initial nuclear polarisation $\Delta N = M$ or $\Delta N = M + 2$. Transitions to states $|2\rangle$ or $|3\rangle$ are accomplished by changing the nuclear polarisation by $2M$ (so the final polarisation is either $-M$ or $-M + 2$). The transition to state $|4\rangle$ is made by flipping a very large number $\sim N$ of nuclear spins, whilst still changing the polarisation by $2M$ only. This allows us to "fine tune" a resonance with state $|1\rangle$, as described in the text. Only a few polarisation groups are shown in the Figure; the insert shows how these fit into the distribution $W(\epsilon)$ (shown for both initial and final states).

Figure 5 The same set of transitions as described in Fig.4, but now for a grain in an external bias $\xi_H$, acting on $\vec{S}$. The change in polarisation is $M_1 + M_2$. The insert shows the initial and final distributions $W(\epsilon \pm \xi_H)$, displaced from each other by $2\xi_H$.

Figure 6 The time dependence of different contributions $P_M(t)$ to the time correlation function $P(t)$ for an ensemble of grains interacting only with nuclear spins, ignoring the effects of nuclear spin diffusion. We assume that the parameter $\lambda = 5$, i.e., roughly 5 nuclei out of $N = 1000$ are flipped each time $\vec{S}$ flips.

Figure 7 The time correlation function $P(t)$ for an ensemble of grains, now plotted against $\ln t$; all contributions $P_M(t)$ are included. This includes only nuclear spin bath-mediated transitions. The figure describes either (a) $P(t)$ neglecting nuclear spin diffusion, and having zero degeneracy blocking ($\mu = 0$), or (b), including both spin diffusion and finite degeneracy blocking.
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\[ \epsilon = \frac{M \omega_0}{2} \]

\[ M + 2 \]

\[ -M + 2 \]

\[ |1\rangle \]

\[ |2\rangle \]

\[ |3\rangle \]

\[ |4\rangle \]
\[ P_{11} \]

- \[ M = 10 \]
- \[ \kappa = 5 \]
- \[ M = 6 \]
- \[ M = 0 \]

\[ \Delta_0 t \]
$P_{11}$

$kappa = 10$

$\ln(2\Delta_0 t)$