DECOHERENCE IN THE QUANTUM DYNAMICS OF A "CENTRAL SPIN" COUPLED TO A SPIN ENVIRONMENT

N. V. Prokof’ev\(^1,2\) and P. C. E. Stamp\(^2\)

\(^1\) Russian Science Center "Kurchatov Institute", Moscow 123182, Russia
\(^2\) Physics Department, University of British Columbia, 6224 Agricultural Rd., Vancouver B.C., Canada V6T 1Z1

We consider here the problem of a "central spin", with spin quantum number \(S \gg 1\), interacting with a set of microscopic spins. Interactions between the microscopic spins are ignored. This model describes magnetic grains or magnetic macromolecules (ferromagnetically or antiferromagnetically ordered) interacting with nuclear spins and with any surrounding paramagnetic electronic spins. It has also been used to describe Si: P near the metal-insulator transition, and quantum spin glasses.

We investigate this model in zero external field. We first set up the general formalism required to analyse problems in which a macroscopic degree of freedom (here, a central spin) interacts with a set of microscopic spins. This is done by reducing the model to an effective low-energy Hamiltonian, and then calculating the correlation function \(\langle \vec{S}(t)\vec{S}(0) \rangle\) using instanton methods. Three physical effects come into play; we call these "topological decoherence" (coming from the phase randomization by the spin environment), "orthogonality blocking" (coming from mismatch between initial and final environmental states) and "degeneracy blocking" (whereby the spin environment destroys degeneracy between the initial and final central spin states). In this paper we consider the possible coherent motion of the central spin. We find that all 3 of the above mechanisms suppress coherence. Orthogonality blocking and topological decoherence destroy phase coherence, and degeneracy blocking prevents most central spins from tunneling at all. The general solution for \(\langle \vec{S}(t)\vec{S}(0) \rangle\) is given for the unbiased case, for all relevant values of the couplings. Under certain conditions, nuclear spin diffusion also plays a role.

The results are then used to calculate the spectral absorption \(\chi''(\omega)\) for 2 experimental systems, viz., TbFe\(_3\) grains, and ferritin molecules. For TbFe\(_3\) (as for most systems), coherence is completely destroyed. However ferritin is very unusual, in that only the degeneracy blocking mechanism is effective - consequently a resonance may still exist, but with a peculiar lineshape.

Our results have general implications for the observation of mesoscopic and macroscopic quantum coherence, and for the foundations of quantum mechanics. They show that a spin environment usually has a very destructive effect on coherence, which cannot be understood using the conventional "oscillator bath" models of quantum environments.

I. INTRODUCTION

In this paper we solve a model problem which, we believe, should be of interest to both condensed matter physicists working on quantum spin problems, and to physicists outside this field who are interested in the foundations of quantum mechanics (particularly the "measurement problem", and the physics of measurements and "decoherence"). It should also be of interest to physicists and chemists working on quantum dissipation and models of the environment, for one of the central points in what follows is the elucidation of the properties of a new kind of quantum environment, quite different from the usual "oscillator bath" models.

The model we discuss was originally introduced to deal with the effects of environmental spins on the coherent tunneling of magnetic grain magnetisation \(^1\)\(^3\). It consists of a central "giant spin", having spin quantum number \(S \gg 1\), coupled to a set \(\{\vec{\sigma}_k\}\) of "microscopic spins"; the microscopic spins are not coupled to each other (Fig.1). The model is then completely described by a Hamiltonian \(H_o(\vec{S})\) for the central spin, a Hamiltonian \(H_{env}(\{\vec{\sigma}_k\})\) for the
microscopic spins, and a coupling $H_{\text{int}}(\vec{S}, \{\sigma_k\})$ between the two. One can also introduce an external field $\vec{H}_o$ which couples to these spins - the effect of external fields will be considered in detail in a 2nd paper.

In most practical applications of this "central spin model", the environmental spins will be either nuclear spins (which may be either inside the object carrying the giant spin, or outside it, in some substrate, or solvent, or surrounding matrix) or else paramagnetic electric "spin impurities", which may also be inside or outside the giant spin. The central spin may be a magnetic grain $\text{MnSi}$ or a magnetic macromolecule such as ferritin $\text{Fe}_{12}O_{12}$; or it may be one of the large superparamagnetic "spin clusters" which are believed to exist in many disordered magnets at low temperature, such as $\text{Si}_i: P$ near the metal-insulator transition $\text{Fe}_3$, or "giant magnetic polarons" $\text{Mn}_3$. Similar spin clusters exist in "quantum spin glasses" $\text{Fe}_3$.

The crucial point which has come out of our study of this model, and which we believe is of great importance for both quantum magnets and for discussions of decoherence and the theory of measurement, is as follows. The effect of microscopic spins on the quantum coherence of the central spin is akin to the effect of an invasion of microscopic viruses on a host bacterium, i.e., in most cases complete destruction! Environmental spins act as a far more potent suppressor of quantum coherence than any other kind of quantum environment hitherto discussed - in particular, the conventional bosonic oscillator baths $\text{Fe}$ seem relatively benign.

Now although the spin environment we consider constitutes an example of an "unconventional environment" ref. [13], a moment’s thought shows that such spin environments will be the rule rather than the exception, when considering macroscopic or mesoscopic quantum coherence. This is because most large quantum objects will have at least a coupling to their own nuclei. Thus in the oft-cited example of the superconducting SQUID ring $\text{Ni}$, there is an electromagnetic coupling of the tunneling flux coordinate (and the associated ring supercurrent) to all of the nuclear spins within a penetration depth of the surface (either in the junction or in the ring), as well as to any paramagnetic impurity spins that happen to be present $\text{Ni}$. While the effect of these interactions on tunneling may not be serious, except under special circumstances $\text{Ni}$, their effect on coherence (MQC) will be quite drastic.

This means that our results are not only of practical importance for any experiments which attempt to test quantum coherence on the mesoscopic or macroscopic scale. They also show why coherence is so easily destroyed at the mesoscopic or macroscopic level (in particular, in measuring devices). This point is not obvious in the previous discussions of, e.g., MQC in the conventional models, in which bosonic oscillators models of the environment are used - in fact, it is predicted, using such models, that MQC ought to be visible in SQUID rings $\text{Ni}$ and in magnetic grains $\text{Fe}_3$, using presently achievable values of the classical dissipation in these systems. Whilst no observation of MQC has yet been claimed in SQUIDs, it has been claimed that MQC has been detected in samples of ferritin magnetic macromolecules $\text{Fe}_3$. Our central spin model can be directly applied to this case - as we shall see here, the case of ferritin is very unusual, in that most of the decoherence mechanisms we shall discuss are inoperative. In fact only one mechanism, which we call "degeneracy blocking", operates on ferritin, leading to an unusual lineshape which is not obviously inconsistent with the experiments (the ferritin experiments have also been criticized on other grounds, mostly related to the power absorption and the behaviour as a function of field $\text{Fe}_3$). In section VI we will give a quantitative discussion of spin environment effects on ferritin, and also $\text{TbFe}_3$ grains.

Our general thesis is that the almost universal presence of spin environments is going to make it far harder to see quantum coherence on the macroscopic scale than has previously been realized. Even on the mesoscopic scale it will be difficult. There are general strategies that one may adopt in looking for suitable candidates for MQC (or mesoscopic coherence). For example, it helps to choose a quantum soliton as one's macroscopic quantum object $\text{Ni}$, since quantum solitons, by definition, do not couple linearly to other excitations of their own field. An example of current interest concerns the quantum tunneling of macroscopic magnetic domain walls - recent experiments have apparently discovered this in $\text{Ni}$ wires $\text{Ni}$, and it will be interesting to see how experiment compares with theory in this case. It is also useful to make the quantum object neutral - this removes all electromagnetic couplings to the environment (although these are often not important, since they are infra-red weak $\text{Ni}$). However the crucial step is clearly one of getting rid of any environmental spins - this suggests at the very least a system for which the nuclear spin is zero. There is in fact an almost ideal candidate $\text{He}$, viz., superfluid $\text{He}$. Here the macroscopic quantum object (whose size is defined by the geometry of the container) is a vortex (a special kind of topological quantum soliton). The system is neutral, and the nuclear spin is zero; moreover, the system can be purified to a quite extraordinary degree $\text{Ni}$, and the experimental cell walls coated with solid $\text{He}$; to remove any influence from impurities, bumps, etc. In fact the only couplings left are to phonons; in common with other quantum vortex problems, such as magnetic vortices $\text{Ni}$, this coupling has both a local and a non-local component. However, just as with photons, that part of this coupling which is responsible for decoherence is infra-red weak (both of these couplings can be described by a Caldeira-Leggett spectral function $J(\omega) \sim \omega^3$).

In this paper we will not be able to discuss all of these issues, concerning the physics of spin environments. In particular, we intend to discuss elsewhere some of the consequences for both mesoscopic quantum devices and quantum
coherence experiments. These applications of the theory are at the forefront of present attempts by physicists to push the "F.A.P.P. barrier", between the quantum world and the classical world [23], ever further into the mesoscopic and even macroscopic domain. The implications for the measurement problem will also be dealt with elsewhere.

In what follows we begin (in section II) by describing the central spin model, and giving some discussion of the relevant physical coupling energies in it, with the aid of physical examples (ranging from magnetic grains to "spin clusters" in Si:P). In section III we show how an initial microscopic Hamiltonian, describing the giant central spin and its microscopic environmental satellites, can be reduced to a low-energy effective Hamiltonian description [2]. The particular way this reduction is done, and the terms that are left at the end, is partly a matter of theoretical choice. We have done it in a way which displays most conveniently the important physical processes that operate at low energies. To help the reader who finds this reduction a little abstract, we sketch how this reduction is done in practice for a real system.

In section IV we proceed to the discussion of 3 solvable limits of our model Hamiltonian [23]. Each of these limits brings out a different physical effect. The simplest of these to understand is "topological decoherence", in which the topological phase of the spin environment becomes entangled with the coherent phase of the giant spin, and destroys the phase coherence of the latter. This decoherence mechanism is completely novel, and has many amusing consequences - it leads to decoherence without dissipation or energy exchange of any kind, and the decay of the correlation functions of the central spin, caused by it, is most unusual [24]. The second effect is of "orthogonality blocking"; it arises when there is a mis-match between the final-state spin wave-functions in the environment, and their initial states. Although this effect is reminiscent of the "orthogonality catastrophe" first discussed by Anderson [24], which is at the heart of decoherence in the conventional oscillator bath models [12], it differs in important details (for example, instead of exponential suppression of tunneling rates, one gets power low suppression; and the effect on the correlation functions of the central spin $\vec{S}$ is most unusual). Finally we study the limit appropriate to pure "degeneracy blocking" [25], which arises because the coupling between the central spin and the environmental spins lifts that degeneracy, between initial and final states of the central spin, which is essential for coherence.

In this paper we will largely concentrate on the coherence properties of the central spin, rather then its relaxation from any particular state. As we shall see, this usually requires consideration only of processes in which the total polarization of the environmental spin bath is unchanged when $\vec{S}$ flips. Other processes necessarily destroy coherence.

Having thoroughly understood the 3 limits, we proceed in section V to the discussion of the "generic case", i.e., having arbitrary values of the parameters in the effective Hamiltonian, so that all 3 mechanisms are operating. At this point we also include the weak nuclear spin diffusion effects, which can also play a role in decoherence.

In section VI, we return to two of the physical examples described in section III.C (i.e., $TbFe_3$ and ferritin), and show how their behaviour is determined by the solutions derived in sections V and VI. In the case of $TbFe_3$, which is fairly typical, we find a catastrophic destruction of coherence - no trace of any resonance is left. On the other hand ferritin has very few nuclear spins, and one finds that only degeneracy blocking play any role in the final behaviour. As a result there is a significant reduction in the spectral weight of the resonance, but the lineshape still shows a strong but highly asymmetric peak. Finally in section VII we summarize our results.

An important omission from this paper is the effect of an external field. This field will strongly affect the giant spin dynamics (with usually a much lesser affect on the microscopic spins). Moreover, dissipation can play an essential role in the problem, whereas in the unbiased case we consider, decoherence arises without dissipation. A full discussion of the relaxation of $\langle \vec{S}(t)\vec{S}(0) \rangle$ (as opposed to coherence) also demands that we consider incoherent transitions involving a change in the spin bath polarization - moreover, since the relaxation of both the central spin and the spin bath is determined by coupling to phonons and possibly electrons, these must also be included. In view of the experimental importance of the field effects, we have reserved discussion of them to a second paper.

For practical applications of our results, it is important to note that there will also be an influence on coherence coming from phonons or electrons. These can interfere in interesting ways with the effect of the environmental spins [25]. We hope to address this aspect in more detail at a later time.

Many of the results appearing in sections II,III and IV of the present paper have appeared previously in short communications [1–3,26]. Here we give detailed derivations for the first time. The results in section V (the generic case) are new here, as is the detailed analysis for the various applications. Some of the mathematical derivations are quite lengthy, and are relegated to Appendices.

II. THE MODEL, AND SOME SIMPLE RESULTS

In this section we give a physical discussion of the central spin problem, with reference to a number of different examples, including magnetic grains, "spin clusters", etc. The various physical interactions in these problems are
described and quantified.

A. The central spin

A typical central spin is made up of a very large number of microscopic spins, whose motion is locked together by nearest-neighbour exchange couplings (or, in the case of superparamagnetic spin complexes, by indirect spin-spin couplings such as the RKKY interaction). The exchange coupling can be very large, as much as $1 - 2eV$; this is much larger than other individual spin energy scales such as those coming from anisotropy, so that in principle one can have large monodomain magnetic particles, with many spins (up to perhaps $10^8$, depending on the particular systems) lined up to form a "giant spin". Another more subtle example is the antiferromagnetic grain, in which the sign of the nearest-neighbour exchange is positive, so that nearest neighbours are anti-parallel, and one obtains a "giant Néel vector" [23]. The superparamagnetic spin complexes are more delicate still, and typically arise at low temperatures when paramagnetic electronic spins in a disordered magnet "lock together" via indirect exchange, RKKY interactions, etc.

Now it is clear that a realistic Hamiltonian for such a giant spin will be very complex. Let us consider first the simplest case of a ferromagnetic grain. An enormous literature exists on the properties of such grains, ranging from simple elemental grains, such as Ni, Fe, Tb, or Ho, to more complex magnetic compounds such as FeCo$_5$, Fe$_2$O$_3$ ($\gamma$-haematite), Mn$_{12}$O$_{12}$ and its derivatives, right up to organic magnetic molecules. With present techniques it is possible to prepare many grains of very uniform size [28] (or else rely on nature for the preparation of magnetic macromolecules).

An obvious first step in analysing such a system is to treat the entire spin complex as a rigid quantum rotator $\vec{S}$, with dynamics governed by the single-spin anisotropy field in the particle/grain; i.e., we write a central spin "bare Hamiltonian" $H_o(\vec{S})$, with $|\vec{S}| = S$ a constant. Early studies of the quantum dynamics of this model were stymied by the constraint of constant $|\vec{S}|$, which implies a kinetic term in the Lagrangian which is linear in time derivatives - this leads to well-known difficulties when one tries to apply path-integral methods to the problem [24]. A number of crucial advances allowed the solution of the problem, most particularly the proper treatment of the kinetic term, which leads to the "Haldane phase" [30], and also a correct formulation of WKB theory for the tunneling dynamics of $\vec{S}$, which yields the correct semi-classical solution for tunneling of an angular momentum through the barrier [31]. This semi-classical solution was also later derived using the instanton calculus [4,32] (although there still seem to be differences between the 2 methods, concerning the tunneling prefactor [31]).

As a simple example of the bare Hamiltonian $H_o(\vec{S})$ we may take (we write $\vec{S} = S\vec{s}$, where $S = |\vec{S}|$)

$$H_o(\vec{S}) = -K_\parallel s_z^2 + K_\perp s_y^2,$$

(2.1)

which has two degenerate classical minima at $s_z = \pm S$; the semi-classical paths between these 2 minima move on or near the easy XZ-plane (note that $K_\parallel$ and $K_\perp$ are proportional to $S$). Notice that if $K_\perp = 0$, there is no tunneling because $s_z$ is conserved; this point is obviously connected with the fact that we are dealing with angular momenta and not particles, and caused some confusion in the literature before 1986. Application of a magnetic field adds a term $-\vec{M} \cdot \vec{H}_o = -\gamma\vec{S} \cdot \vec{H}_o$ to (2.1); if $\vec{H}_o$ is directed along $\hat{z}$, this biases the symmetric 2-well problem in (2.1), but if it is applied along $\hat{x}$, it lowers the barrier height and displaces the degenerate minima towards each other in the XZ-plane. Application of $\vec{H}_o$ along $\hat{y}$ distorts the semi-classical path and lowers the barrier. A further effect of an applied $\vec{H}_o$ is mostly nicely seen in the instanton language; since the 2 possible paths between the degenerate minima involve opposite Haldane topological phase [33], this phase can be changed by an external field [34], causing the tunneling splitting to change and even oscillate.

In fact the original papers of van Hemmen and Suto analyzed a very general class of central spin bare Hamiltonian, given by

$$H_o(\vec{S}) = -K_\parallel |s_z|^4 - 1/2 \sum_{r=1}^n K_r (s_+^r + s_-^r),$$

(2.2)

and a considerable number of more recent papers have concerned themselves with re-analyzing special cases of (2.2).

Consider now what has been left out of (2.1) and (2.2). One obvious omission is the infra-red weak coupling of $\vec{S}$ to photons (via the magnetic dipolar interaction [19,20]) and to phonons. Another omission is coupling to higher excited states of the particle, involving spin flips of spins inside the grain (internal magnons); the neglect of these is
usually justified by appeal to the large value of spin exchange \( |J| \) of spin exchange \([1,20]\), in models like equation (2.1). Incorporation of such processes implies relaxation of the constraint \( |\vec{S}|=\text{constant} \). Another omission is that of surface excitations of the grain - in any real grain, there can be low-energy magnetic excitations involving "loose spins", i.e., spins which, because of imperfect preparation or the inevitable defects, dislocations, etc., have a coupling energy \( \ll J \) to their neighbours. The effect of these is hard to quantify, but will be included below in our description of environmental spin effects.

Some of the above additional physics has been discussed already in the literature. The effect of phonons has been analyzed in the usual Caldeira-Leggett instanton scheme by Garg & Kim [36] and Chudnovsky [33]. These calculations indicate very small corrections to tunneling rates coming from phonons, which is not surprising given the infra-red weakness of phonon environments. Notice however that these calculations ignore barrier fluctuation effects [37–39].

We shall also ignore coupling of \( \vec{S} \) to electrons. Actually, in contradistinction to domain walls [41,42], electrons have a very powerful effect on the dynamics of \( \vec{S} \). Here we simply assume the grain is insulating.

We now turn to the really important omission from (2.1) and (2.2), viz, the coupling of \( \vec{S} \) to other spin degrees of freedom - as already noted, these will include nuclear spins and paramagnetic electronic impurities, both inside and outside the grain. It will not include the low-energy excitations of coupled spins inside or outside the grain - these are generally describable in terms of a bath of oscillators (e.g. magnons) with a very weak super-Ohmic coupling to \( \vec{S} \). Thus it is crucial that the environmental spins be weakly coupled to each other - then the variety of effects we study in this paper become possible.

The most ubiquitous of such environments is the nuclear spin environment - this is well understood. A nucleus with a finite spin \( I \), and spin moment \( -\gamma_N \hbar I \), will interact both locally with the currents of electron clouds at the same ionic site, via the contact hyperfine interaction, and also non-locally with other ions via the dipolar field. There may also be other residual interaction such as "transferred" hyperfine interactions (as in, e.g., \( Fe^{19} \) in \( MnF_2 \) or \( CoF_2 \)), or quadrupolar couplings if \( I > 1/2 \), or nuclear spin-phonon interaction. Finally, one may have interactions between the different nuclear spins, via, e.g., spin waves (the Suhl-Nakamura interaction), or dipolar couplings - these are very weak (Suhl-Nakamura interactions are \( \lesssim 10^{-5} K \), and nuclear dipole-dipole interactions \( \sim 10^{-7} K \) but, it turns out, can play a role in the dynamics of \( \vec{S} \); this is discussed in Section III.E. The most important interactions are the contact hyperfine interaction and the dipolar interaction between the nuclei and electrons. Thus the nuclear moments in nonmagnetic hosts (e.g., \( H^1 \) in \( H_2O \), or in \( CuCl_2 \cdot H_2O \)) will be described by a Hamiltonian

\[
H_I = -\gamma_N \hbar \vec{r} \cdot \left\{ \vec{H}_o - \gamma_e \hbar \sum_j \frac{1}{r_j^3} \left[ \langle \vec{S}_j(T) \rangle - 3 \frac{\vec{r}_j \cdot \langle \vec{S}_j(T) \rangle}{r_j^2} \right] \right\},
\]

where \( \vec{H}_o \) is an applied field, \( \hbar \gamma_e = \mu_B g_e c \), where \( g_e \) is the electronic g-factor, \( \sum_j \) sums over all ionic sites and \( \langle \vec{S}_j(T) \rangle \) is the temperature-dependent expectation value of spin polarization at the sites (for weak fields, \( \langle \vec{S}_j(T) \rangle = \chi_j \vec{H}_o \), where \( \chi_j \) is the susceptibility tensor). On the other hand a nucleus in a magnetic ion will have a Hamiltonian

\[
H_I = -\gamma_N \hbar \vec{r} \cdot \left\{ A \cdot \langle \vec{S}_o \rangle + \vec{H}_o + H_{dip} \right\},
\]

\[
H_{dip} = -\gamma_e \hbar \sum_{j \neq o} \frac{1}{r_j^3} \left[ \langle \vec{S}_j \rangle - 3 \frac{\vec{r}_j \cdot \langle \vec{S}_j \rangle}{r_j^2} \right].
\]

The strength of these interactions is measured directly in NMR experiments; the hyperfine coupling range from \( 1 - 3 MHz \) (i.e., \( 5 - 15 \times 10^{-5} K \)) for protons in \( H^1 \), up to values greater than 5000 MHz \((0.25 K) \) for some rare-earth magnetic nuclei (e.g., \( Tb^{159} \), \( Dy^{163} \)); these latter correspond to local fields acting on the nuclei as high as 500 Tesla, and come overwhelmingly from the contact interaction. Thus when a central spin rotates, with all internal electronic spins rotating in unison, the contact interaction tries to force the nuclei on the magnetic sites to follow. If the hyperfine interaction strength is \( \omega_o \), then it is clear that the ratio \( \omega_o/\Omega_o \), with \( \Omega_o^{-1} \) the timescale of central spin rotation, is going to be crucial to the nuclear spin dynamics. If the electronic spin frequencies \( \Omega_o \gg \omega_o \), then the nuclear spins will experience a "sudden" perturbation, and few of them will follow the central spin; conversely, if \( \omega_o \gg \Omega_o \), the nuclear spins will follow adiabatically. In fact, as we shall see in more detail later in the paper, the ratio \( \omega_o/\Omega_o \) is usually
somewhere between 0.01 – 0.05 (although for rare earth nuclei like $Tb^{159}$ or $Dy^{163}$ it can be ~ $O(1)$, depending on the host). On the other hand for nuclei in non-magnetic ions, $\omega_o/\Omega_o$ may be much smaller, and will of course depend strongly on the host (in a magnetic host, dipolar interactions from permanent moments, and also possible transfer hyperfine interactions $-\gamma_N h \vec{r} \cdot \vec{A}_{ij} \cdot \vec{S}_j$ from nearby magnetic moments $\vec{S}_j$, can greatly increase $\omega_o$; for example, $\omega_o$ for $F^{19}$ in $MnF_2$ is 160 MHz).

A large central spin will also interact significantly with nuclear spins in a surrounding medium, such as a substrate or solvent, via the dipolar interaction generated by the central spin dipolar field, i.e.,

$$H_{int} = -\hbar^2 \gamma_e \gamma_N \sum_i \vec{I}_i \cdot \left( \sum_j \frac{1}{r_{ij}^3} \left[ \vec{S}_j - 3 \frac{\vec{r}_{ij} \cdot \vec{S}_j}{r_{ij}^2} \right] \right) ,$$

where $r_{ij}$ is the distance between some nuclear moments $\vec{I}_i$ in the surrounding matrix and a local moment $\vec{S}_j$ which is incorporated in the central spin. This interaction is not negligible; for a central spin with $S = 10^7$, the dipolar coupling to a nucleus at a distance of 10$^3$Å is already ~ 1 MHz, rising to 30 MHz at a distance of 300Å; and there is clearly a very large number of nuclear spins within 1000Å of the central spin!

This brings us to another potentially very serious source of decoherence for the central spin, viz., paramagnetic impurity spins in the surrounding matrix. The Hamiltonian has the same form as above except that $\gamma_e \vec{S}_i$ is replaced by $\gamma_e \vec{S}_i$, describing an impurity electronic spin $\vec{S}_i$ at some distance $r_{id}$ from the central spin. In a very pure surrounding matrix this is a problem because $\gamma_e \sim 2 \times 10^6 \gamma_N$; for the central spin with $S = 10^7$, this dipolar coupling is still > 2 MHz for a paramagnetic impurity 10$^4$Å distant! A sphere of this radius will contain over 10$^{11}$ ions, so unless we have a matrix of, e.g., solid ultrapure 4He (or perhaps very high grade Si), there will be many such impurities with appreciable coupling to $\vec{S}$.

Finally, as we approach the surface of the giant spin, we may expect a number of electronic spins to have "weak exchange" coupling to the giant spin. This may arise from imperfections or defects or even dirt at the surface (or near it, as in the coating around ferritin molecules). Typically we expect some kind of "superexchange" coupling to apply, with energy anywhere between 10 – 1000 K (i.e., roughly in the range 10$^{10}$ – 10$^{13}$ Hz). We shall refer to such spins in the spin environment as "loose spins". How many of them there will depend very much on the materials and material preparation used.

A somewhat similar picture may well apply to Si : P near the metal-insulator transition, where it is believed that the ESR lineshape [43] may well be explained in terms of a Hamiltonian

$$H_{eff} = A \vec{S} \cdot \sum_{k=1}^N c_k \vec{I}_k + \vec{H}_o \cdot \vec{S} ,$$

in which a central spin "superparamagnetic cluster" interacts with nuclear spins. This may well be so, provided there are insignificant decohering interactions between the spin complexes. We hope at some time to give a detailed analysis of this case, and also of similar quantum spin glass systems.

We may summarize this survey of the various components of the spin environment surrounding a ferromagnetic grain by the diagram in Fig.2. It is important to notice [13] how completely the range of frequencies from 10$^9$ Hz downwards is covered by the various coupling strengths; and even above 10$^9$ Hz, for very large grains and/or including "loose spins".

B. Some examples

The reader of this paper who is not a specialist in magnetism may also find it helpful to get a feel for specific numbers, for different materials in which different interactions are important. Thus in this sub-section we will give some "vital statistics" for a number of real magnetic systems.

Before doing so, it will be useful to glance at Table I, which lists some nuclear hyperfine coupling energies, in various host systems. For those who like to think of energy in temperature units, note that 1 K ~ 21.6 GHz. It will be noticed that the couplings range over a factor ~ 10$^3$, and that even the nuclei from non-magnetic ions can have reasonably large couplings, via transferred hyperfine interaction ($F^{19}$ in the Antiferromagnet (AFM) $MnF_2$ being a good example). For the transition metal Ferromagnets (FM), $\omega_k$ ranges from 50 – 500 MHz, but the rare-earth FM’s can have $\omega_k$ an order of magnitude greater. Where more then one value of $\omega_k$ is given, there is either a quadrupolar splitting of the lines (e.g. $Tb$, $Dy$) or nuclei are at inequivalent lattice sites (as in YIG).
The internuclear dipole interactions are way down from this (typically \( \sim O(1 kHz) \)); and even the indirect Shul-Nakamura interactions have energy scale \( \sim 100−200kHz \) (see Table I). Of course these interactions are always there, and they lead to an inevitable minimum spreading \( \delta \omega_k \) in the couplings; but \( \delta \omega_k/\omega_k \) is clearly very small \( (10^{-5}−10^{-3}) \) and so our assumption of independent nuclei is very accurate (note that the temperature \( T \) will always be far larger than this linewidth \( \delta \omega_k \)). However we notice that \( \delta \omega_k \) is not necessarily small compared to \( \Delta_o \); this will be important for the magnetic relaxation at long times, but is irrelevant for coherence, as we shall see later.

We have already seen (cf. Fig.2) how the coupling to external nuclei and external electronic spins is also important. As a rough guide, this coupling can be estimated as \( \omega_k \sim 10^8 S/r^3 Hz \), where \( S \) is the spin quantum number of the central spin, and \( r \) is the distance of a nuclear spin from \( S \) in \( A \). For an electronic spin, \( \omega_k \sim 2\times10^{11} S/r^3 Hz \) (obviously the prefactor is modified depending on the spin quantum numbers and \( g \)-factors of the environmental spin).

Consider now a few experimental systems, by way of example:

(a) \( MnF_2 \) grain: This is an AFM; however it will have an uncompensated excess spin moment which may couple to external nuclei or electronic spins, depending on the host. The size of \( \Omega_o \) will depend on many factors for a grain, mostly uncontrollable strain fields - in principle one expects \( \Omega_o \) to range from \( \sim 0.1 K \) to \( 1K \) for such a system. There are two hyperfine couplings; \( \omega_k = 160 MHz \) (\( F^{19} \)) and \( 680 MHz \) (\( Mn^{55} \)). If \( \Omega_o = 0.1K \) this gives values of \( \alpha_k = (\pi/2)\omega_k/\Omega_o \) equal to 0.125 and \( \sim 0.5 \), respectively; if \( \Omega_o = 1K \), they are ten times smaller.

(b) \( YIG \) grain: One can make very pure grains of Yttrium Iron Garnet. Since it is a soft magnet, a well-prepared system would have \( \Omega_o < 0.1 K \), even perhaps as low as \( 0.01K \). In the former case, the \( Fe^{57} \) hyperfine coupling has \( \alpha_k \sim 0.05 \); in the latter case \( \alpha_k \sim 0.5 \). Note that only 2.19% of \( Fe \) nuclei have a moment.

(c) Ferritin: Ferritin is a magnetic macromolecule made in all eukaryotic cells; a typical molecule contains 4500 \( Fe \) ions in a ferritin structure, which are antiferromagnetically ordered, although there is an excess uncompensated moment of size somewhere between 200 and \( 600\mu_B \). The 2.19% of \( Fe^{57} \) ions (roughly 100 of them) have \( \omega_k = 64 MHz \). In the experiments of Awschalom et al. [17], an anisotropy field \( \sim 1.72 T esla \) was found, indicating \( \Omega_o \sim 2K \), which indicates a value for \( \alpha_k \sim 2.2 \times 10^{-3} \).

(d) \( TbFe_3 \) grain: Here \( \Omega_o \) is typically \( 3K \); this is very high indeed. However so are the \( \omega_k \) since \( Tb \) is a rare-earth magnet. With the quadrupolar split hyperfine lines, one finds \( \alpha_k \sim 0.08, 0.07, 0.05 \) for the \( Tb \) nuclei. The 2% of \( Fe^{57} \) nuclei will have \( \alpha_k \) some 50 times smaller than this; the result of this for coherence of \( S \) will be, in effect, to simulate a spread \( \delta \omega_k \) in the \( \omega_k \), rather as though we had a spread \( \delta \omega_k \sim 50 MHz \) in the system.

From these examples we see that typical values of \( \alpha_k \) are between \( 10^{-3} \) and \( O(1) \), usually something like 0.05. Moreover the presence of other nuclear species such as \( H^1 \) (in \( H_2O \), as in ferritin) will cause a small spread in the \( \alpha_k \). Finally, of course, there will be a much larger spread in the \( \omega_k \) coming from spins outside the grain or molecule.

In section III we shall return to examine two of these examples, namely \( TbFe_3 \) and ferritin, in greater detail.

### III. THE EFFECTIVE HAMILTONIAN

In this section we make the crucial step of going from "bare Hamiltonian" like those in Section I to a low energy "effective Hamiltonian". This effective Hamiltonian is our starting point for calculating the dynamics of \( S \) at low \( T \). It contains a variety of effective couplings, which already contain all the high-energy physics. We emphasize that in realistic cases one may actually calculate these renormalised couplings, starting with the original bare Hamiltonian - they do not have to be treated as phenomenological couplings to be determined from experiment. To illustrate this we actually do the truncation explicitly for a typical hyperfine interaction.

#### A. Reduction to \( H_{eff} \)

Let us go back to our "bare" Hamiltonian \( H_o(S) \) for \( S \), before either truncation to a low energy effective Hamiltonian, or coupling to any environment. There are two important energy scales in \( H_o(S) \), viz, \( \Omega_o \) and \( \Delta_o \); the former describes the "fast" motion of the central spin in the effective potential in \( H_o(S) \) (small oscillations near the potential minima, or the "bounce frequency" involved in tunneling through the barrier), whereas the latter characterizes the much slower coherence between tunneling events, since \( \pi/2\Delta_o \) is the average time between successive tunneling events. These 2 time scales are seen very clearly if we show a "typical trajectory" for \( S(\tau) \), i.e., a typical path that would contribute to a path integral evaluation of the dynamics of \( S \). This is shown in Fig.3, for a case in which \( \dot{S}_1 \) and \( \dot{S}_2 \) are not antiparallel. There will be other even more rapid time scales which are buried in this path, corresponding to transitions to very high excited states of \( H_o(S) \); but their amplitude is very small.
The truncation of \( H_o(\vec{S}) \) is standard [3][4][5]. Consider, e.g., the bare Hamiltonian (2.2); truncation gives
\[
H_o(\vec{S}) \rightarrow 2\Delta_o \hat{\tau}_x \cos \pi S = H_o(\vec{r}) ,
\]
where the Pauli matrix \( \hat{\tau}_x \) flips \( \vec{S} \) between the degenerate configurations \( \vec{S}_1 = \hat{z}S \) and \( \vec{S}_2 = -\hat{z}S \), the factor \( 2\cos \pi S \) is an expression of Kramers theorem. The bare splitting \( \Delta_o \) is given in terms of \( \Omega_o \) by
\[
\Delta_o \sim \Omega_o e^{-A_o} ,
\]
\[
A_o \sim 2(K \parallel / K \perp)^{1/2} S ,
\]
showing the exponential ratio between \( \Delta_o \) and \( \Omega_o \) (more accurate expression for \( \Delta_o \) appear in Refs. [6][7][8]); and for this model
\[
\Omega_o \sim \frac{2}{S}(K \parallel / K \perp)^{1/2} .
\]
Notice that although \( \Omega_o \) is absent from (3.1), it is nevertheless implicitly contained via the "WKB renormalization" of (3.2).

Let us now consider how one should make the same truncation in the presence of a large number of environmental spins. In general the same argumentation will apply - we look first at how the environmental spins will affect the "fast" physics of the central spin (on a time scale \( \Omega_o^{-1} \)), and absorb this into a new effective Hamiltonian describing a "renormalized instanton" for the central spin. We then deal with the "slow" physics (on time scales \( \gg \Omega_o^{-1} \)) using this renormalized instanton, and derive a new effective Hamiltonian for this slow, low-energy physics.

However the same is certainly not true if we have \( N \) environmental spins. How can we handle this case? The method we have adopted (3.3) starts by asking how we should write the effective action to be associated with the instanton. In the absence of the spin environment, the transition amplitude matrix from \(| \vec{S}_1 \rangle \) and \(| \vec{S}_2 \rangle \) is given by
\[
K_o^\pm = \tau_x \Omega_o \exp( -A_o \mp i\pi S ) = \Delta_o \hat{\tau}_x e^{\pm i\pi S} ,
\]
where \( \Delta_o = \Omega_o e^{-A_o} \), and \( A_o \sim (K \parallel / K \perp)^{1/2} S \), in accordance with equations (3.2) and (3.4); the \( \pm \) refers to the 2 different (clockwise or anticlockwise) paths between \(| \vec{S}_1 \rangle \) and \(| \vec{S}_2 \rangle \). Now when we add the spins, the instanton will act as an operator in the subspace of each environmental spin (able to cause transitions in each subspace, in line with our discussion in section II B). The general form now must be
\[
\hat{K}_o^\mp (k) = \Delta_o \hat{\tau}_y e^{\delta_k} \exp \left\{ \left[ \delta_k \vec{u}_k \pm \xi_k \vec{v}_k \right] \cdot \hat{\sigma}_k \pm i\eta[\pi S + \phi_k + \alpha_k \vec{n}_k \cdot \hat{\sigma}_k] \right\} , \quad (\eta = \pm) ,
\]
in the subspace of the \( k \)-th spin - here \( \hat{\tau}_y \equiv \hat{\tau}_x \pm i\hat{\tau}_y \), \( \vec{u}_k \), \( \vec{v}_k \), and \( \delta_k \) are unit vectors in some direction, and \( \delta_k \) is a number. The instanton from \(| \vec{S}_2 \rangle \) to \(| \vec{S}_1 \rangle \) is Hermitian conjugated to that in (3.6) The role of the new terms in (3.6) is as follows:

(i) The adiabatic correction \( \delta_k \) is due to the high-frequency (\( \gg \Omega_o \)) fluctuations of \( \hat{\sigma}_k \), which renormalize the bare instanton action \( A_o \). This correction is usually very small, and is of course unobservable because it does nothing but renormalize \( A_o \). It increases the "moment of inertia" of \( \hat{S} \).

(ii) The "barrier fluctuation" terms \( \left[ \vec{u}_k \delta_k \pm \xi_k \vec{v}_k \right] \cdot \hat{\sigma}_k \) come from fluctuations of \( \hat{\sigma}_k \) which are of frequency \( \sim \Omega_o \) or less. As discussed in Refs. [9] and [10] (see pp. 53-56 of Ref. [9]), the effect of these fluctuations is, amongst other things, to cause the potential barrier to fluctuate in height. In Section II B we discuss in more detail how this leads to a term of this form for our problem. Here we simply note that (a) the effect of \( \vec{u}_k \) is to increase the tunneling rate, because barrier fluctuations occasionally lower the barrier, thereby creating a "window of opportunity" for \( \vec{S} \) to tunnel; and (b) this term can be entirely absorbed into a renormalization of the "orthogonality blocking" term that we will have in our final effective Hamiltonian. In the most general case one has to allow different couplings for the clockwise and counter-clockwise instantons, e.g., when the environmental spin is oriented along the \( \hat{x} \) direction it influences the two instanton trajectories in the \( XZ \) plane differently, thus giving a non-zero value of \( \xi_k \).

(iii) The topological term \( \alpha_k \vec{n}_k \cdot \hat{\sigma}_k + \phi_k \) describes the dynamical effect on \( \hat{\sigma}_k \) when \( \vec{S} \) flips. Its origin is two fold: one contribution comes from a transfer matrix (note that \( \alpha_k \neq \tau_3 k \))
\[
\hat{T}_k^\pm = e^{i \int_\tau d\tau H_{int}(\tau)} = e^{\pm i(\vec{u}_k \delta_k \hat{\sigma}_k + \phi_k)} ,
\]
in the spin subspace of the $k$-th spin, where in the strong coupling case the phase $\phi_k$ approaches the value $\phi_k = \sigma_k^B - \pi/2$; $\sigma_k^B$ is the adiabatic Berry phase accumulated by the nuclear spin. The other contribution $(\alpha_k - \pi_k)$ is closely related to the barrier preparation effect, but has no analog in the standard description of the particle tunneling through the barrier; it arises from the influence of the environmental spins on the topological phase (see below).

This summarizes the mutual effects of $\vec{S}$ and $\{\vec{\sigma}_k\}$ during the tunneling of $\vec{S}$. However after this tunneling is over, there will be a residual interaction between $\vec{S}$ and the $\{\vec{\sigma}_k\}$. This can be described as follows. Suppose the 2 relevant quasiclassical states of $\vec{S}$ are again oriented along $\vec{S}_1$ and $\vec{S}_2$, and that the total effective fields acting on $\vec{\sigma}_k$ are correspondingly $\gamma_k^{(1)}$ and $\gamma_k^{(2)}$, defined in units such that their interaction energy with $\vec{\sigma}_k$ is $\gamma_k \cdot \vec{\sigma}_k$. Now define the sum and the difference terms

$$
\omega_{\parallel} l_k = \gamma_k^{(1)} - \gamma_k^{(2)}
$$

$$
\omega_{\perp} m_k = \gamma_k^{(1)} + \gamma_k^{(2)}.
$$

(3.8)

where the $l_k$ and $m_k$ are unit vectors. In the truncated Hilbert space of $\vec{\tau}$ and $\{\vec{\sigma}_k\}$, the residual interaction term takes the form

$$
H_{static} = \frac{1}{2} \left\{ \tau_z \sum_{k=1}^{N} \omega_{\parallel} l_k \cdot \vec{\sigma}_k + \sum_{k=1}^{N} \omega_{\perp} m_k \cdot \vec{\sigma}_k \right\},
$$

(3.9)

i.e., a term which changes when $\vec{S}_1 \rightarrow \vec{S}_2$, and an extra term which does not. This latter term can arise in various ways. Most commonly it will arise because the spin-1/2 variable $\vec{\sigma}_k$ is produced by truncating some environmental spins $I_k$, in a bare Hamiltonian, with $I_k > 1/2$. It will also arise if, e.g., $\vec{S}_1$ and $\vec{S}_2$ are not exactly antiparallel (e.g., in an imperfect grain), or simply because there are small stray fields acting on the $\vec{\sigma}_k$. Usually $\omega_{\perp} \ll \omega_{\parallel}$ (at least for nuclei in a magnetic system).

We are now ready to write the full low-energy effective Hamiltonian for our central spin model, in the absence of an external field, but incorporating all the mutual effects between $\vec{S}$ and its spin environment.

$$
H_{eff} = 2\Delta_o \left\{ \tilde{\tau} \cos \left[ \Phi + \sum_{k=1}^{N} (\alpha_k \tilde{\sigma}_k - i \xi_k \tilde{u}_k) \cdot \vec{\sigma}_k \right] + H.c. \right\} + \tilde{\tau} \sum_{k=1}^{N} \frac{\omega_{\parallel} l_k \cdot \vec{\sigma}_k}{2} + \sum_{k=1}^{N} \frac{\omega_{\perp} m_k \cdot \vec{\sigma}_k}{2}.
$$

(3.10)

The form of this effective Hamiltonian is central to all further discussions. We have dropped the barrier fluctuation term $\beta_k \tilde{u}_k \cdot \vec{\sigma}_k$, since as already noted its effects will simply consist in a renormalization of $\Delta_o$ and the effective ratio $\omega_{\parallel}^{\tau}/\omega_{\parallel}^{\tau}$, whose effects appear in orthogonality blocking (see the next section). The phase $\Phi = (\pi S + \sum_k \phi_k)$, i.e., we absorb the Berry phase into our total value of the topological phase. Finally, $\Delta_o = \Delta_o \exp(-\sum_k \delta_k)$, absorbing the adiabatic corrections.

Before going on, it is worth asking (a) how general is $H_{eff}$, and (b) what, if anything, has been left out of it?

Consider first the general structure of $H_{eff}$. We see that in our $2^{N+1}$-dimensional Hilbert space, it contains not only all possible pairwise interactions between $\vec{\tau}$ and each $\vec{\sigma}_k$, but also a set of higher couplings, involving as many as $N$ environmental spins, produced by expanding the cosine; i.e., we have couplings of form

$$
H_{dyn} \sim \tilde{\tau}_i A_{\alpha \beta \gamma \delta \cdots}^{\alpha \beta \gamma \delta} \hat{\sigma}_{k_1}^{\alpha} \hat{\sigma}_{k_2}^{\beta} \hat{\sigma}_{k_3}^{\gamma} \hat{\sigma}_{k_4}^{\delta} \cdots.
$$

(3.11)

Some readers might like to think of (3.11), as being an infinite set of terms in a perturbation expansion of the bare coupling to all orders, but we feel that this is mistaken - the coefficients in (3.11) are fixed precisely by the cosine, and in any way there is nothing perturbative about the form of $H_{eff}$ - it is valid even when the coupling between the $\vec{\sigma}_k$ and $\vec{\tau}$ is very strong.

In fact the only terms left out of $H_{eff}$ are direct interactions between the $\{\vec{\sigma}_k\}$ (i.e., ones not generated, like (3.11), via coupling to $\vec{S}$). For nuclei these are the dipolar and Nakamura-Suhl interactions, and for paramagnetic impurities they could also be RKKY interactions between widely-separated spins. Thus the only terms left out of $H_{eff}$ are terms of the form

$$
\hat{V}(\{\vec{\sigma}_k\}) = \sum_{k \neq k'} V_{kk'}^{l} \hat{\sigma}_k^{\alpha} \hat{\sigma}_{k'}^{\beta}.
$$

(3.12)

As we shall see later in this paper, these terms can also be included in our analysis. Their effect on coherence is usually small (although under some circumstances they can affect coherence in large grains). However in a second paper, dealing with relaxation of $\vec{S}$ in a bias, we will see that $\hat{V}$ does play a very important role.
Readers not used to the philosophy underlying the derivation of low-energy effective Hamiltonians may also suspect that somehow the form of $H_{\text{eff}}$ depends in some way on the semiclassical analysis we use of the dynamics of $\tilde{S}$. We emphasize this is not so - the form of $H_{\text{eff}}$ depends in no way on any semiclassical or WKB approximation. What does depend on the semiclassical analysis is the exact value of each of the renormalised parameters in $H_{\text{eff}}$. If $\tilde{S}$ were a microscopic spin, it would not be appropriate to use WKB theory to evaluate, e.g., $\Delta_\alpha$ in $H_\alpha(\tilde{r})$, or the $\{\alpha_k\}$ in $H_{\text{eff}}$. However when $S \gg 1$, the use of WKB/instanton methods becomes extremely accurate, both in determining $H_\alpha(\tilde{r})$ (cf. Refs. [28, 31–33]), and (as we shall see in the next sub-section) in determining the values of the parameters in $H_{\text{eff}}$.

Finally, note how differently $H_{\text{eff}}$ describes the "spin bath" environment from the usual oscillator bath models. Not surprisingly, the physical effects of the spin bath will also turn out to be very different - from this point of view it is useful to think of the spin bath as an example of an "unconventional environment", having unusual effects (whether it interacts with a central spin, or SQUID, or any other collective coordinate). This theme has been elaborated elsewhere [13].

### B. Instanton Derivation of $H_{\text{eff}}$

As a first step we will present general arguments and the physical ideas involved in such a derivation, and proceed then with the more specific calculation for the Hamiltonian [24].

Suppose we are interested in the transition amplitude from $|\tilde{S}_1\rangle$ to $|\tilde{S}_2\rangle$ in the WKB approximation for the central spin only, ignoring for the moment the initial and final states of the environment. Without any interaction between the $\tilde{S}$ and $\{\tilde{\sigma}_k\}$ we have, of course,

$$\hat{o}K_n^\pm = \tilde{\tau}_a \Omega_a \exp\{-A_o \pm i\pi S\}, \quad (a = \pm) \quad (3.13)$$

Now if we calculate the transition amplitude with all the interactions included, according to

$$\hat{K}^\pm \{|\chi_k^{(in)}\rangle\} = \langle \tilde{S}_1 | e^{-i \int d\tau [H_o(\tau') + H_{\text{int}}(\tau')]} | \tilde{S}_2 \rangle \{|\chi_k^{(in)}\rangle\} \quad (3.14)$$

it will obviously depend on the initial state of the $\{\tilde{\sigma}_k\}$, and, when projected on the particular final state of the environmental spins, it will depend on the spin flips in the environment. Thus [23] in the most general way we have to consider the instanton from $|\tilde{S}_1\rangle$ to $|\tilde{S}_2\rangle$ as an operator in the subspace of $\{\tilde{\sigma}_k\}$. Eq. (3.6) is the most general form for this operator one can write down no matter what are the interactions in the system (the only omission is the simplified form for the adiabatic contribution $\delta_k$ which in a more general form should read $\delta_k \pm \tilde{\delta}_k$, applying to the case when the central spin is in an external magnetic field so that the adiabatic contribution is asymmetric for the clockwise and anticlockwise instantons [27]). Thus the real question to ask is not why we write the instanton in this form, but rather - what is the physical meaning of these parameters, and what is their relation to the original parameters characterizing $H_o$ and $H_{\text{int}}$?

We start by describing the adiabatic contribution to the action due to $\delta_k$. This comes from the environmental spins strongly coupled to the central spin, i.e., with couplings $\omega_k \gg \Omega_o$. The slow (on a time scale defined by $\Omega_o^{-1}$) rotation of the central spin can not cause any transitions from the ground state for these spins (in quantum tunneling problems we deal with temperatures $T \ll \Omega_o$ - otherwise the "classical" over-barrier transitions will prevail), and they have to follow the direction of the local field acting on them. It is appropriate to include these spins in the "combined central spin". In general, we increase the moment of inertia which will result in a correction to the instanton action. Let us calculate this correction using the particular form of $H_o(S)$ given by (2.1). The corresponding Lagrangian written in terms of angles $(\theta, \varphi)$ has the form

$$\mathcal{L}(\theta, \varphi) = iS'\varphi \sin \theta \dot{\theta} + U(\theta, \varphi) \quad (3.15)$$

$$U(\theta, \varphi) = K_\| \left[ \sin^2 \theta (1 + \lambda_a \sin^2 \varphi) - 2h \sin \theta \cos \varphi \right], \quad (3.16)$$

where $\lambda_a = K_\perp/K_\|$ is the anisotropy parameter, and $S' = S = 1/2 \cdot N_{ad}$ gives the number of environmental spins locked in a joint rotation with the central spin. For further discussion we also include a weak magnetic field acting in the $\tilde{x}$-direction $h = \gamma_e H_x S/(2K_\|) \ll 1$. As usual [27], we perform integration over the small deviations of the angle $\varphi$ around zero, and obtain the effective Lagrangian for $\theta$ as
The classical equation of motion starting at $\theta = \pi/2$ at $t = 0$, and approaching the angle $\arcsin(h)$ as $t \to \infty$ has the form

$$
\dot{\theta} = \Omega_o \frac{S}{S'} (h - \sin \theta) \left( 1 - \frac{h}{\lambda_{\text{a}} \sin \theta} \right)^{1/2},
$$

(3.18)

where $\Omega_o$ is the bare bounce frequency (3.4). For small $h$ and a large value of the anisotropy parameter (the action is proportional to $2S/\lambda_{\text{a}}^{1/2}$, so that very large values of $\lambda_{\text{a}}$ are necessary to make any discussion of mesoscopic spin tunneling physically meaningful) we may drop the last factor in the r.h.s. of equation of motion. The solution is now

$$
\sin \theta(\tau) = \frac{1 + h \cosh \Omega \tau}{\cosh \Omega \tau + h},
$$

(3.19)

$$
\Omega = \Omega_o \frac{S}{S'} \sqrt{1 - h^2}.
$$

(3.20)

Substituting this expression back into the effective action we have

$$
A = 2 \sqrt{K_{\parallel} / K_{\perp}} S' \left( \sqrt{1 - h^2} - h \arccos(h) \right).
$$

(3.21)

From this we derive the correction to the instanton action due to the increase of the moment of inertia

$$
\delta A_{\text{in}} = \sqrt{K_{\parallel} / K_{\perp}} N_{\text{ad}},
$$

(3.22)

which is proportional to the number of spin-1/2 "satellites" added to the "bare" central spin. The result is quite general, although the coefficient in front of $N_{\text{ad}}$ will depend on the particular form of $H_{\text{a}}(\vec{S})$.

For the hyperfine interaction we do not expect any variation in the potential energy. In a more general case, i.e., for the dipolar coupling between the $\vec{S}$ and $\{\vec{\sigma}_k\}$, or when there are interactions acting on $\{\vec{\sigma}_k\}$ other than their interaction with $\vec{S}$, the energy $U(\theta, \varphi)$ will be modified. This problem is hard to deal with exactly because now the potential energy will depend on the orientations $\vec{r}_{ij}/r_{ij}$ between the environmental spins and those forming the central spin. However to estimate the effect we do not need the exact solution. It is sufficient to look at the correction to the action due to weak applied field, which also changes the potential energy, by amount $\delta U \sim \gamma_c H S$. It follows from (3.21) that for small $h$ the correction is

$$
\delta A_{\text{pot}} \sim \pi \frac{\delta U}{\Omega_o},
$$

(3.23)

and may be quite strong if $\delta U$ is large. Still we assume that only a small number of "loose spins" on the surface of the grain $N_{\text{ad}} \ll S$ have dipolar couplings strong enough to move adiabatically with the grain magnetisation; thus the total change $\delta A_{\text{pot}}$ is much less than $A_o$.

To summarize, the adiabatic parameter $\delta_k$ can vary from a very small value $\sim \lambda_{\text{a}}^{-1/2}$ up to $\pi \delta U/\Omega_o$ depending on the model (for nuclear spins inside the grain the former case is the most relevant one since the dipolar coupling is much less than the contact hyperfine interaction).

Now we proceed to the discussion of the terms given by $\beta_k$ and $\xi_k$ in (3.6). Suppose we have some environmental spins which are coupled to the central spin weakly, $\omega_o/\Omega_o \ll 1$. These spins can not follow the central spin adiabatically - they rather see the rotating $\vec{S}$ as a sudden perturbation, so that most of them simply conserve their spin orientations during the time $\Omega_o^{-1}$. On the other hand their coupling to the central spin slightly changes the actual instanton path.

We may start by considering this influence as a weak static magnetic field due to the $\{\vec{\sigma}_k\}$ acting on $\vec{S}$, i.e., we write the interaction Hamiltonian as $H_{\text{int}} = \vec{S} \cdot \delta \vec{H}$, where $\delta \vec{H}$ is an operator in the environmental spin subspace, and can be easily found for the hyperfine interaction (2.4) and dipolar interaction (2.3). Using the fact that $\vec{S}$ is rotating so fast that $\delta \vec{H}$ hardly changes during the transition time, we may neglect its time dependence (which will be accounted for later in our discussion of the parameters $\alpha_k$ and $\phi_k$). The rest is simple now, for we have to find a linear in $\delta \vec{H}$ correction to the action, i.e., the variation of $A$ due to the extra potential energy $\vec{S}_{\alpha} \cdot \delta \vec{H}_{\alpha}$. In fact this was already
done for the $H_o(S)$ described by (2.1) and $\delta \hat{H}$ along the $\hat{x}$-direction. The linear in $h$ correction is easy to obtain from (3.21)

$$\delta A_k = -\frac{\gamma_e \delta \hat{H}_x S}{\Omega_o} = -\frac{\pi \omega_o}{2 \Omega_o} \sigma_x ,$$

(3.24)

where $\omega_o$ is the contact hyperfine interaction. In this particular case we find

$$\xi_k = -\frac{\pi \omega_k}{2 \Omega_o} ; \quad \vec{v}_k = \hat{x} .$$

(3.25)

One might try to argue that the calculation of the first order correction to $A$ can be done for arbitrary $H_o(S)$ and $H_{int}(\{\sigma_k\}, \hat{S})$ as follows. In the expression for the instanton action

$$A = -\int d\tau (L_o(\tau) + H_{int}(\tau)) ,$$

(3.26)

the first order correction is given simply by

$$\delta A = -\int d\tau H_{int}(\vec{S}_o(\tau)) ,$$

(3.27)

where $\vec{S}_o(\tau)$ is evolving according to the unperturbed semi-classical solution. For the case of hyperfine interactions considered above this expression takes the form

$$\delta A = -\frac{\omega_k}{2 \Omega_o} \hat{\sigma}_k \int d\tau \vec{S}_o(\tau) ,$$

(3.28)

Substituting here (3.19) (for $h = 0$) we reproduce the result (3.25). The symmetry of the problem in zero external field implies that the corrections due to $\delta \hat{H}_{x,y}$ are zero, which means that here $\vec{v}_k \approx 0$.

This would be a correct way of calculating things if we were dealing with ordinary particles but it is not correct for the angular momenta. The formula (3.27) takes into account the change in the potential energy only, while for the constrained rotation of $\hat{S}$ any time-reversal symmetry breaking field can also influence the topological phase. To see how it works we apply the magnetic field along $\hat{y}$-direction, which results in a term

$$-2K_i h \sin \theta \sin \varphi \approx -2K_i h \sin \theta \varphi .$$

(3.29)

Integrating over small $\varphi$ as before we obtain the effective Lagrangian for the angle $\theta$ as (up to a constant term)

$$\mathcal{L}(\theta) = \mathcal{L}_o(\theta) - i hS \frac{\lambda}{\lambda} ,$$

(3.30)

which results in a topological phase correction

$$i \pi S \rightarrow i \pi S + i \pi \frac{\gamma_e \hat{H}_y S}{\Omega_o} \equiv i \pi S + i \frac{\pi \omega_o}{2 \Omega_o} \sigma_y .$$

(3.31)

Thus we find one of the contributions to the coupling $\alpha_k$ to be

$$\alpha_k^{(top)} = \frac{\pi \omega_o}{2 \Omega_o} ; \quad \vec{n}_k^{(top)} = \hat{y} .$$

(3.32)

Now we turn to the last contribution to our instanton operator (3.6), which is due to the small, but unavoidable dynamics of $\{\sigma_k\}$ during the transition. To define this we note that from the environmental spin point of view the rotating central spin is seen as a time dependent external magnetic field. Thus we calculate the evolution of the microscopic spin wave-function according to

$$\hat{T}_k | \chi_{env} \rangle = e^{i \int d\tau H_{int}(\tau) | \chi_{env} \rangle} ,$$

(3.33)

For the weak coupling case $\omega_k / \Omega_o \ll 1$ we are considering here (sudden perturbation), this can be calculated as
\[
\hat{T}_k \approx 1 + i\delta \hat{H} \int d\tau \hat{S}_o(\tau) \approx \exp\{i\delta \hat{H} \int d\tau \hat{S}_o(\tau)\},
\]

Thus we find exactly the same expression as in (3.27) which is hardly surprising since we are dealing with the same Schrödinger equation but now in the real time domain. From this we derive the second contribution to the parameter \(\alpha_k\). Again, for contact interactions between the spins we have

\[
\alpha_k = \frac{\pi\omega_o}{2\Omega_o}; \quad \eta_k = \bar{\eta},
\]

These results for \(\alpha_k\), \(\xi_k\), \(\phi_k\), and \(\delta_k\) are all for the realistic case where \(\omega_k/\Omega_o \ll 1\). However in the rare cases where \(\omega_k/\Omega_o \geq 1\), the generalization of the above method is easy. For example, in Fig.4 we show the variation of \(\alpha_k\) as a function of \(\omega_k/\Omega_o\), for the \(H_o(\hat{S})\) of (2.1). The actual form of \(\alpha(\omega_o/\Omega_o)\) for a contact hyperfine interaction is quite lengthy, but almost indistinguishable from the simple form

\[
\alpha(x) \sim (\pi/2) \tanh x.
\]

Remarkably, this simple form is almost universal - it hardly depends on the detailed form of \(H_o(\hat{S})\). This point is dramatically illustrated if we take the really pathological instanton

\[
\theta(\tau) = \frac{\Omega_o}{2}[\eta(\tau + \pi/\Omega_o) - \eta(\tau - \pi/\Omega_o)] + \frac{\pi}{2}[\eta(\tau + \pi/\Omega_o) + \eta(\tau - \pi/\Omega_o)] .
\]

where \(\eta(\tau)\) is the unit step function. Equation (3.37) describes a "jerk start" and "jerk stop" instanton, with the polar angle \(\theta(\tau)\) varying linearly in between; thus

\[
\cos \alpha = \frac{\Omega_o}{(\omega_o^2 + \Omega_o^2)^{1/2}} \sin[\pi/2(1 + \omega_o^2/\Omega_o^2)^{1/2}] ,
\]

which is almost indistinguishable from Fig.4 except for some small "wiggles" around \(\alpha \approx \pi/2\), when \(\omega_o > \Omega_o\) (these arise from the infinite acceleration and deceleration at \(\tau = \pm \pi/\Omega_o\) respectively).

The saturation of \(\alpha(x)\) at \(\pi/2\) when \(x > 1\) is easy to understand - it corresponds to the adiabatic (Berry phase) limit, when \(\delta_k\) rotates adiabatically with \(\hat{S}\). Similar calculations may be made of \(\xi_k\), \(\phi_k\), \(\delta_k\), etc., for the full range \(0 < x < \infty\) of couplings.

This concludes our discussion of the instanton operator. Each term in the exponent (3.1) has its own physical meaning and may be calculated from first principles once the form of \(H_o(\hat{S})\) and the most relevant interactions are identified. Here we analyzed the most important case of hyperfine interactions between the central spin and nuclear spins when \(H_o(\hat{S})\) is given by (2.1). We see that the expression (3.6) simplifies a lot for nonadiabatic coupling \((\omega_k/\Omega_o \ll 1)\):

\[
\delta_k = \bar{\eta}_k = \phi_k = 0; \quad \alpha_k \bar{n}_k = \frac{\pi\omega_k}{2\Omega_o} (\hat{x}, \hat{y}) ; \quad \xi_k \bar{v}_k = -\frac{\pi\omega_k}{2\Omega_o} \hat{z} .
\]

In the adiabatic limit \((\omega_k/\Omega_o \gg 1)\) we have

\[
\bar{\eta}_k = \phi_k = \xi_k = 0; \quad \delta_k = \lambda_A^{-1/2}; \quad \alpha_k \to \pi/2 .
\]

IV. THREE IMPORTANT LIMITING CASES

The effective Hamiltonian \(H_{eff}\) in (2.10) has a very rich behaviour in general. To appreciate this behaviour it is best to lead up to it by first studying some limiting cases, each one of which brings out an important aspect of the physics. This is done as follows. First we assume that \(\omega_k^\parallel\) and \(\omega_k^\perp\) are zero - although this is a rather artificial way of doing things, it enables us to see "pure topological decoherence", without interference from anything else [1]. Next we suppress topological decoherence, by making \(\alpha_k\) very small, and we also make all the \(\omega_k\) equal; however the ratio \(\omega_k^\perp/\omega_k^\parallel\) now becomes the crucial parameter, governing the behaviour of "pure orthogonality blocking". Finally we study "pure degeneracy blocking"; this is done by making \(\alpha_k \to 0\) (suppressing topological decoherence), and letting \(\omega_k^\perp = 0\) (which suppresses orthogonality blocking) but now allowing a distribution of different values of \(\omega_k = \omega_k^\parallel\). We shall see the precise meaning of these appellations as we study each limiting case.
Naturally, the generic behaviour of $H_{\text{eff}}$, with completely arbitrary values of $\alpha_k$, $\omega_k^\parallel$, $\omega_k^\perp$ and $\xi_k$ involves all three mechanisms; it can also involve nuclear spin diffusion. We shall see how this works in section [V].

It is crucial in what follows to remember that a necessary (but not sufficient) condition for coherence is that the total effective bias acting on $\vec{S}$ be very small - the initial and final states of $\vec{S}$ must be within $\sim \Delta_o$ of each other, otherwise tunneling cannot occur at all. Even if it can, it may be incoherent.

A. Topological decoherence

Formally the case of pure topological decoherence applies to the following special case of $H_{\text{eff}}$:

$$H_{\text{eff}}^{\text{top}} = 2\Delta_o \hat{\tau}_z \cos \left[ \Phi + \sum_{k=1}^{N} \alpha_k \vec{n}_k \cdot \hat{\sigma}_k \right], \quad (4.1)$$

Notice here that (a) since $\omega_k^\parallel$ and $\omega_k^\perp$ are zero, all environmental states are degenerate, and so (b) the initial and final states of the environmental spins, and $\vec{S}$, are degenerate - there is no exchange of any energy between $\vec{S}$ and the $\{ \hat{\sigma}_k \}$. The only thing that is exchanged is phase, i.e., the phase of $\vec{S}$ becomes entangled with that of the $\{ \hat{\sigma}_k \}$, during the tunneling of $\vec{S}$. Thus the initial and final states of the spin environment are not the same. The term proportional to $\xi_k$ which changes the real part of the instanton action and tends to choose either clockwise or counter-clockwise trajectories will be included in section [V].

The properties of $H_{\text{eff}}^{\text{top}}$ were entirely elucidated in refs. [13] (see section 2.2 (ii) of ref. [3]): here we explain the derivation and results in more detail. What we are interested in are the correlation functions of $\vec{S}(t)$, most particularly in $P_{\text{eff}}(t)$ and its Fourier transform in the frequency domain. This function simply tells us the probability to find the system in the same state at time $t$ as it was at time $t = 0$; its Fourier transform tells us about the response of the system to a continuous perturbation at some frequency $\omega$. In the case where all interactions are zero we simply revert to the free central spin, for which

$$P_{\text{eff}}^{(0)}(t) = \frac{1}{2} [1 + \cos(4\Delta_o t \cos \Phi)] \quad (4.2)$$

$$\chi''(\omega) = \pi \delta(\omega - 4\Delta_o \cos \Phi) \quad (for \ \omega > 0). \quad (4.3)$$

Here $\chi''$ is the imaginary part of $\chi(\omega)$, the Fourier transform of $P_{\text{eff}}(t)$. In this non-interacting case $\Phi = \pi S$, and $\chi''$ is just a sharp line at the tunneling splitting energy $| 4\Delta_o \cos \pi S |$. The spin oscillates coherently between $| \vec{S}_1 \rangle$ and $| \vec{S}_2 \rangle$. The series (4.2) is generated from (4.1) (or alternatively, from the bare transition amplitude (3.5)), in the absence of the $\{ \hat{\sigma}_k \}$, by the usual summation of all possible instanton flips in a time $t$ (see refs. [11,12,33,34,43]).

Returning now to $H_{\text{eff}}^{\text{top}}$, we see that we can formally write the solution as

$$P_{\text{eff}}(t) = \frac{1}{2} \left[ 1 + \langle \cos [4\Delta_o t \cos \Phi + \sum_{k=1}^{N} \alpha_k \vec{n}_k \cdot \hat{\sigma}_k] \rangle \right], \quad (4.4)$$

simply treating the cosine function in $H_{\text{eff}}^{\text{top}}$ as a c-number, because there are no other terms in the Hamiltonian, and it obviously commutes with itself. Or, if one prefers an expansion over all instanton trajectories, we can write

$$P_{\text{eff}} = \frac{1}{2} \left[ 1 + \sum_{s=0}^{\infty} (-1)^s \frac{(2\Delta_o t)^{2s}}{(2s)!} \sum_{n=0}^{2s} \frac{(2s)!}{(2s-n)!n!} e^{i\Phi(2s-2n)} \prod_{k=1}^{N} e^{i\alpha_k \vec{n}_k \cdot \hat{\sigma}_k(2s-2n)} \right]. \quad (4.5)$$

$\langle \ldots \rangle$ is a thermal average over environmental states. Here all these states are degenerate, the average is independent of $T$ and one finds

$$F(n - m) = \langle \prod_{k=1}^{N} e^{i\alpha_k \vec{n}_k \cdot \hat{\sigma}_k(2s-2n)} \rangle = \prod_{k=1}^{N} \cos[2\alpha_k(n - m)]. \quad (4.6)$$
In the non-interacting case, \( F(\nu) = 1 \), and we get \( (4.2) \). For small \( \alpha_k \) we may approximate the product in \( (4.6) \) as
\[
F_\lambda(\nu) = e^{-4\lambda \nu^2} ; \quad \lambda = \sum_{k=1}^{N} \frac{\alpha_k^2}{2}.
\]

Notice that \( \lambda \) is just the mean number of environmental spins that are flipped. We now employ a very useful identity
\[
\int_0^{2\pi} \frac{d\varphi}{2\pi} e^{i(\Phi-\varphi)(2s-2n)} \cos^2 s \varphi \equiv \frac{(2s)!}{2^{2s}(2s-n)!} e^{i\Phi(2s-2n)}
\]
(4.8) to rewrite the expansion \( (4.5) \) as a weighted integration over topological phase:
\[
P_{\uparrow\uparrow}(t) = \sum_{m=-\infty}^{\infty} F_\lambda(m) \int_0^{2\pi} \frac{d\varphi}{2\pi} e^{i2m(\Phi-\varphi)} \left\{ \frac{1}{2} + \frac{1}{2} \cos(2\Delta_o(\varphi)t) \right\}
\]
(4.9)
\[
= \frac{1}{2} \left\{ 1 + \sum_{m=-\infty}^{\infty} F_\lambda(m)e^{i2m\Phi} J_{2m}(4\Delta_o t) \right\},
\]
(4.10)
where \( \Delta_o(\varphi) = 2\tilde{\Delta}_o \cos \varphi \), and \( J_{2m}(z) \) is the Bessel function of \( m \)-th order. For \( F(m) = 1 \) this is just another representation of the coherent dynamics \( (4.2) \). On the other hand in the adiabatic strong-coupling limit, where all of the \( \alpha_k \rightarrow \pi/2 \), we simply have \( F(m) = (-1)^m \), so that
\[
P_{\uparrow\uparrow}(t) \rightarrow \frac{1}{2} \left[ 1 + \cos(4\tilde{\Delta}_o t \cos \Phi) \right] ; \quad (\alpha_k \rightarrow \pi/2),
\]
(4.11)
where the renormalized phase is just \( \Phi + N\pi/2 \), that is
\[
\Phi = \pi S + N \sum_{k=1}^{N} \phi_k^B
\]
(4.12)
where \( \phi_k^B \) is the Berry phase, defined as before via
\[
\phi_k^B = \phi_k - \pi/2.
\]
(4.13)

Equation \( (4.11) \) simply tells us that in the strong coupling limit the total topological phase now comes from the bare phase \( \pi S \), plus the adiabatic Berry phase accumulated by all the environmental spins rotating adiabatically with it. If the coupling Hamiltonian is such that, e.g., all the \( \{ \sigma_k \} \) are forced to be parallel to \( \hat{S} \) in this limit, then this extra phase will just be \( N\pi/2 \) (i.e., we have effectively increased our spin quantum number from \( S \) to \( S + N/2 \)). However this is by no means necessary - if there is some other field acting on \( \sigma_k \) in addition to that due to \( \hat{S} \) then the Berry phase will deviate from \( \pi/2 \).

By far the most important regime is the ”intermediate coupling” regime, in which \( \alpha_k > N^{-1/2} \) (note that if \( N \) is large, then \( \alpha_k \) may be very small here). This is typically going to be the case, since usually \( \alpha_k \) will be independent of \( N \). In this case we have a large parameter \( \lambda \) in \( (4.7) \) with
\[
F_\lambda(\nu) = \delta_{\nu,0} + \text{small corrections} \quad \text{(intermediate)}.
\]
(4.14)
so that, very surprisingly, we get a universal form in the intermediate coupling limit for \( P_{\uparrow\uparrow}(t) \) (again, \( \eta(x) \) is the step function)
\[
P_{\uparrow\uparrow}(t) \rightarrow \frac{1}{2} \left[ 1 + J_0(4\tilde{\Delta}_o t) \right] \equiv \int \frac{d\varphi}{2\pi} P_{\uparrow\uparrow}^{(0)}(t, \Phi = \varphi)
\]
(4.15)
(compare the angular average of the coherent series in \( (4.10) \)), and \( \chi''(\omega) \)
\[
\chi''(\omega) \rightarrow \frac{2}{(16\tilde{\Delta}_o^2 - \omega^2)^{1/2}} \eta(4\tilde{\Delta}_o - \omega) \quad \text{(intermediate)}.
\]
(4.16)
We plot these universal forms in Fig.5. The physics of this universal form is simply one of phase cancellation in the expression \(\langle 1.0 \rangle\) for \(F(m)\). As explained in ref. [1], this phase cancellation arises because successive flips of \(\vec{S}\) cause, in general, a different topological phase to be accumulated by the spin environment, so that when we sum over successive instantons for \(\vec{S}\), we get phase randomisation and hence loss of coherence. This forces \(2s - n = n\) in the sum in \(\langle 1.3 \rangle\), i.e., the only paths that can contribute to \(P_{\vec{n} \uparrow \downarrow}(t)\) are those for which the number of clockwise and anticlockwise flips of \(\vec{S}\) are equal - in this case the topological phase "eaten up" by the environmental spins is zero. By looking at expression \(\langle 1.15 \rangle\) one sees another explanation - the universal behaviour comes from complete phase phase randomisation [1], so that all possible phases contribute equally to the answer! The final form shows decaying oscillations, with an envelope \(\sim t^{-1/2}\) at long times. This decay can also be understood [2] by noting that the "zero phase" trajectories that contribute to \(P_{\vec{n} \uparrow \downarrow}\) now constitute a fraction \((2s)!/(2^s s!)^2 \approx s^{-1/2}\) of the total number of possible trajectories, where \(s \sim \tilde{\Delta}_o t\). Because of this decay, the peak in the spectral function at \(\omega = 4\tilde{\Delta}_o \cos \Phi\) is now transformed to the spectral function of a 1-dimensional tight-binding model, as shown in Fig.5b.

From the point of view of fundamental principles this result is really rather interesting, for here we have a case of "decoherence without dissipation"; the coherent oscillations of \(\vec{S}(t)\) are destroyed without any exchange of energy with the environment. This mechanism would persist even if the bath were at zero temperature. Whilst there is no reason why such "pure decoherence" cannot exist elsewhere in nature, we are not aware of other examples [44].

It is actually very interesting to examine the intermediate coupling regime a little bit more closely, especially for the 1/2-integer spin, when there is no tunneling for \(\alpha_k = 0\). In Fig.6 we see that weak coupling (\(\alpha_k\) taking random values between 0 and 0.05) to 40 spins completely changes this behaviour - we now have an effective splitting \(\Delta_{eff} \sim \tilde{\Delta}_o/3\), but with strong damping. Fig.7 shows rather stronger coupling (\(\alpha_k\) having random values between 0 and 1). The result shown is for \(S = \)integer, but in fact the result for \(S = n + 1/2\) is almost indistinguishable from this. We see that the result is, within numerical accuracy, completely indistinguishable from the universal behaviour in \(\langle 1.13 \rangle\), for only 20 environmental spins.

B. Orthogonality blocking

Orthogonality blocking arises when there is a mismatch between the initial and final state wave-functions of the environment \(\langle 1.4 \rangle\). Expressed in this way it is an old idea, as old as the Debye-Waller or Franck-Condon, or polaronic factors for superohmic environments, or the "orthogonality catastrophe" of the X-ray edge and Kondo problems [2], which reappears in the discussion of Ohmic environments in the oscillator bath model of the environment \(\langle 1.2 \rangle\).

In the present case of the spin bath, the mismatch between \(|\sigma_k^{\uparrow\downarrow}\rangle\) and \(|\sigma_k^{\downarrow\uparrow}\rangle\) arises because the initial and final fields, \(\tilde{\gamma}_k^{(1)}\) and \(\tilde{\gamma}_k^{(2)}\), acting on \(\vec{\sigma}_k\), are not exactly parallel or antiparallel. Thus if \(\vec{\sigma}_k\) is initially aligned in \(\tilde{\gamma}_k^{(1)}\), it suddenly finds itself, after \(\vec{S}\) flips, to be misaligned with \(\tilde{\gamma}_k^{(2)}\). There is then an overlap with a flipped state in the new basis, equivalent to an amplitude \(\beta_k\) for \(\vec{\sigma}_k\) to have actually flipped (semiclassically, \(|\sigma_k^{\uparrow\downarrow}\rangle\) starts precessing in \(\tilde{\gamma}_k^{(2)}\)). Depending on the readers' taste, one may think of this as a "quasi-flip" generated by the basis change, or a real flip; however quantum-mechanically, for all purposes the spin has actually flipped.

Thus superficially we are dealing with a standard orthogonality problem. However as we now see the details are rather different.

We start by applying the following restrictions to \(H_{eff}\) in \(\langle 3.10 \rangle\):

i) We assume \(\alpha_k^2 N \ll 1\), i.e., extremely small \(\alpha_k\); this removes topological decoherence.

ii) We assume that the effective fields \(\tilde{\gamma}_k^{(1)}\) and \(\tilde{\gamma}_k^{(2)}\) acting on \(\vec{\sigma}_k\) at the beginning and end of the instanton are not either parallel or antiparallel; usually we will assume however that they are nearly parallel or antiparallel, i.e., that either \(\tilde{\gamma}_k^{(1)} \approx \tilde{\gamma}_k^{(2)}\) (i.e. that \(\omega_k \gg \omega_{k'}\), or else \(\tilde{\gamma}_k^{(1)} \approx -\tilde{\gamma}_k^{(2)}\) (so that \(\omega_k \ll \omega_{k'}\)). Both of these cases are very common, indeed almost the rule - quite apart from any stray fields that might disrupt perfect parallelism, there is also the contribution coming from barrier fluctuations (cf. Appendix A) which will contribute to orthogonality blocking even if parallelism is otherwise is perfect. Stray fields will come from a variety of sources (weak external fields, or strain fields in the sample, or demagnetisation fields from the dipole forces, etc., etc.).

iii) We assume that the couplings \(\omega_k\) satisfy the inequalities \(\omega_k \gg \Delta_{eff}\), where \(\Delta_{eff}\) will be defined below. The parameter \(\tilde{\Delta}_{eff} < \tilde{\Delta}_o\) in general; often \(\Delta_{eff} < \tilde{\Delta}_o\). This condition is virtually always obeyed for hyperfine interactions, since we expect \(\Delta_{eff} < 1\,MHz\).

iv) We assume that all the \(\omega_k\) are equal. This is of course a very unrealistic assumption (even the very small spread in hyperfine couplings will turn out to be important), but it is made to avoid the introduction of degeneracy blocking, to which we will come presently.
To be specific let us consider the case where \( \gamma_k^{(1)} \approx -\gamma_k^{(2)} \), which will arise in many situations; the analysis for the other most common problem, where \( \gamma_k^{(1)} \approx \gamma_k^{(2)} \), goes through in exactly the same way. Thus "pure orthogonality blocking" applies to the effective Hamiltonian

\[
H_{\text{ef}} = 2\Delta \hat{\tau}_x + \hat{\tau}_z \sum_{k=1}^N \frac{\omega_k^\parallel}{2} \vec{\hat{l}}_k \cdot \vec{\hat{s}}_k + \frac{\omega_k^\perp}{2} \sum_{k=1}^N \vec{\hat{m}}_k \cdot \vec{\hat{s}}_k .
\]  

(4.17)

\[
\Delta \Phi = \hat{\Delta}_\Phi \cos \Phi ,
\]  

(4.18)

where \( \omega_0^\parallel \gg \omega_0^\perp \); in (4.17) we use a "Kramers renormalised" splitting \( \hat{\Delta}_\Phi \).

We now make a number of observations, which are crucial to understanding of orthogonality blocking:

(i) The spin bath spectrum is split by \( \omega_0^\parallel \) into "polarisation groups" of degenerate lines, having polarisation \( \Delta N = N^+ - N^- \), and energy \( E_\gamma (\Delta N) = \tau_\gamma \omega_0^\parallel \Delta N/2 \); that is, we have \( N + 1 \) polarisation groups each separated by energy \( \omega_0^\parallel \), whose energy changes (by \( \omega_0^\parallel \Delta N \)) if \( S \) flips: \( E_\gamma (\Delta N) = -E_\gamma (\Delta N) = \omega_0^\parallel \Delta N/2 \). There are \( C_N^{(N+\Delta N)/2} \) degenerate states in polarisation group \( \Delta N \).

(ii) For \( S \) to flip at all, we require near resonance between initial and final states, i.e., they must be within \( \sim \hat{\Delta}_\Phi \) in energy. Since \( \omega_0^\parallel \gg \hat{\Delta}_\Phi \), this means that if no nuclear spins are flipped during the transition of \( S \), only states having \( \Delta N = 0 \) (the "zero polarisation" states") can flip at all (since \( E_\gamma (\Delta N = 0) = E_\gamma (\Delta N = 0) = 0 \)). At high \( T \), only a fraction \( f = \sqrt{2/\pi N} \) of grains in an ensemble will have \( \Delta N = 0 \).

(iii) However the presence of \( \omega_0^\perp \) means that some spins are flipped (in general a different number of them in each transition of \( S \)). Now consider a grain in polarisation state \( \Delta N \). If when \( S \) tunnels, at the same time nuclear spins also flip so that \( \Delta N \rightarrow -\Delta N \), then since \( E_\gamma (M) = E_\gamma (-M) \), resonance is still preserved and the transition is possible. Notice however that (a) for this change in polarisation of \( 2M \), at least \( M \) spins must flip, and (b) if we wish to preserve resonance (i.e., \( S \) makes back-and-forth transitions) then the polarisation state must change by \( \pm 2M \) each time. In fact \( S \) cannot flip at all unless there is a change in polarisation state of magnitude \( 2M \).

These 3 observations will lead to a fourth, which is most easily given once we have defined a function \( P_M(t) \), which is simply the correlation function \( P_{\gamma\Phi}(t) \), now restricted only to those systems in an ensemble for which \( \Delta N = M \). Since, as remarked above, we can only have transitions between \( \pm \Delta N \) states, we can write the complete correlation function for an ensemble of grains described by (4.17) as

\[
P_{\gamma\Phi}(t; M) = \sum_{M=-N}^N w(T, M) P_M(t) ,
\]  

(4.19)

\[
w(T, M) = Z^{-1} C_N^{(N+M)/2} e^{-M\omega_0^\parallel/k_B T} ,
\]  

(4.20)

where \( Z \) is the partition function.

We now make the observation

(iv) If we are interested in the incoherent relaxation of \( S \), particularly at long times, then obviously all the \( P_M(t) \) in (4.19) must be considered. However for coherence it is obvious that the \( P_0(t) \) must completely dominate, since it involves transitions of \( S \) with possibly no flipping of the \( \{ \vec{\hat{s}}_k \} \), which are necessarily coherent. On the other hand \( P_{M\neq 0}(t) \) not only involves many spin flips each time (necessarily causing loss of coherence), but it also involves only processes in which some number \( \geq M \) of spin flips leads to exactly a change \( 2M \) in polarisation. From this argument we see that \( P_{M\neq 0}(t) \) will describe slow (i.e., for large \( M \) on time scales \( \gg \hat{\Delta}_\Phi^{-1} \) and incoherent motions of \( S \).

These observations are borne out by the calculation of \( P_M(t) \) in Appendix A.2. Assuming that \( \omega_0^\parallel \gg \omega_0^\perp \) as above, one easily sees that the flip amplitude \( \beta_k \) is just the small angle defined as shown in Fig.8, via

\[
\cos 2\beta_k = -\gamma_k^{(1)} \cdot \gamma_k^{(2)} ,
\]  

(4.21)

where \( \gamma_k^{(i)} \) are unit vectors; it is then found that

\[
P_M(t) = \int_0^\infty dx \, e^{-x^2} \left( 1 + \cos[4\hat{\Delta}_\Phi J_M(2\sqrt{k_B x} t)] \right)
\]  

(4.22)

\[
= 2 \int_0^\infty dx \, e^{-x^2} P^{(0)}_{\gamma\Phi}(t, \Delta M(x)) ,
\]  

(4.23)
where $P^{(0)}_M(t, \Delta_M)$ is just the free function (Eq. (4.3)), but now a function of an $x$-dependent splitting $\Delta_M(x) = 2\Delta_\Phi J_M(2\sqrt{\kappa x})$; and $\kappa$ is just the orthogonality exponent, i.e.,

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

(4.24)

so that $\kappa \approx 1/2 \sum_k \beta_k^2$. Thus the frequency scale of $P_M(t)$ is set by $\Delta_M(x)$. The terms $P_M(t)$ are easily verified to be incoherent, and so

$$P^{(0)}_M(t) \sim f P_0(t) + \text{incoherent},$$

(4.25)

where $f = \sqrt{2/\pi N}$ as before; the exact answer is given still by (1.19) above.

The frequency scale of oscillations in $P_0(t)$ is easily found; we write it in terms of a renormalised splitting $\Delta_{eff}$:

$$P_0(t) = 1/2[1 + \cos(4\Delta_\Phi e^{-\kappa} t)] \quad \Delta_{eff} = 2\Delta_\Phi e^{-\kappa}$$

(4.26)

for small $\kappa$, and

$$P_0(t) = 1 - 4(\hat{\Delta}_\Phi^2 t^2/(\pi \kappa)^{1/2}) + O(\hat{\Delta}_\Phi^4 t^4)$$

(4.27)

for large $\kappa$. Thus the naive ”orthogonality catastrophe polaronic band narrowing” argument to derive $\hat{\Delta}_{eff}$ only works for $\kappa \ll 1$ (where it is superfluous!). In the interesting regime $\kappa \gg 1$, we get a quite different answer (with a much smaller suppression). This is understood as follows. In the usual oscillator bath models, band narrowing comes essentially without any bath transitions (most of the polaron ”cloud” is in virtual high-frequency modes) - it is adiabatic. Here, however, roughly $\kappa$ spins flip each time $\vec{S}$ flips (the probability of $r$ flips is $\kappa^r e^{-\kappa} / r!$, which peaks at $r \sim \kappa$), even though we only consider $P_0(t)$, i.e., even though $\Delta N = 0$ and does not change (the flips compensate - there are just as many one way as the other).

This inevitable flipping of the $\{\vec{S}_k\}$ means that even $P_0(t)$ is incoherent if $\kappa \gg 1$. This is most easily seen by looking at $\chi''_M(\omega)$ which has extraordinary behaviour. One finds

$$\chi''_{M=0}(\omega) = \sqrt{\frac{2}{\pi N}} \frac{\pi}{4\Delta_\Phi \sqrt{\kappa}} \sum_j x_j e^{-x_j^2} |J_1(2\sqrt{\kappa x_j})| \quad J_{0(2\sqrt{\kappa x})} = \pm \omega / 4\Delta_\Phi.$$  

(4.28)

The first factor is just the statistical fraction of zero polarization states in the spin environment. It is the complicated structure of the set of roots $x_j$ to the equation $J_0(2\sqrt{\kappa x_j}) = \pm \omega / 4\Delta_\Phi$ that yields the interesting behaviour. For small $\kappa$ only the lowest square root Bessel function singularity appears, coming from $x_1 \approx (|1 - \omega / 4\Delta_\Phi|/\kappa)^{1/2}$. This leads to

$$\chi''_{M=0}(\omega) = \frac{\pi}{4\Delta_\Phi \kappa} \exp \left( \frac{\omega - 4\Delta_\Phi}{4\Delta_\Phi \kappa} \right) \eta(4\Delta_\Phi - \omega), \quad (\kappa \ll 1),$$

(4.29)

and almost coherent dynamics for $\vec{S}$, as we see in Fig.9. However as $\kappa$ increases, more and more square root singularities contribute to $\chi''$ (which also gets pushed to lower $\omega$, according to (4.27)), and it develops the bizarre structure seen in Figs. 9 & 10. This structure has quite counter-intuitive consequences for $P_0(t)$; interference between the various peak structures can conspire to give the correlation function for $\vec{S}$ a multi-periodic behaviour. An example is shown in Fig.11.

Readers who wish to more fully understand the mechanism of orthogonality blocking are urged to study Appendix A. Recall also that for a real system, the values of the $\beta_k$ will be renormalized by the $\vec{\beta}_k$, (Eq. (3.11) et seq.), which describe the effect of barrier fluctuations. Thus the real value of $\kappa$, and the real values of $\omega^\parallel_k$ and $\omega^\perp_k$, will already include these barrier fluctuation effects.
C. Degeneracy blocking

The previous 2 limiting cases studied have imposed some rather artificial restrictions in order to bring out 2 physical mechanisms involved in decoherence from spin environments. We now study a third case which introduces a note of realism. To do this we suppress topological decoherence (by letting $\alpha^2_k N \ll 1$) and we suppress orthogonality blocking (by letting $\omega_\alpha = 0$). On the other hand we now allow a spread of coupling energies $\omega_k^\parallel$ around a mean value $\omega_o$. Thus we consider the effective Hamiltonian

$$\hat{H}_{\text{eff}} = 2\Delta_\Phi \hat{r}_x + \tau_z \sum_{k=1}^N \omega_k^\parallel \hat{\sigma}_k^z ;$$

(4.30)

with a spread of values of $\omega_k^\parallel$ of

$$\sqrt{\sum_k (\omega_k^\parallel - \omega_o)^2} \equiv N^{1/2} \delta\omega_o.$$  

(4.31)

i.e., a distribution of width $\delta\omega_o$. Clearly, this Hamiltonian is identical to the standard biased two-level system with the bias energy $\epsilon$ depending on the particular environmental state; thus $\epsilon = \sum_{k=1}^N \omega_k^\parallel \sigma_k^z$. The introduction of this spread is to destroy the exact degeneracy between states in the same polarization group. For coherence to take place, we require the initial and final states to be within roughly $\Delta_{\text{eff}}$ of each other, otherwise Landau-Zener suppression of the transition takes place (cf. Appendix A.1). Since we assume a random distribution of couplings around $\omega_o$, it is clear that there will be a crossover between unblocked behaviour for $\delta\omega_o \ll \Delta_o / N^{1/2}$, and Landau-Zener blocked behaviour for $\delta\omega_o \gg \Delta_o / N^{1/2}$. In practice once $\delta\omega_o \sim \Delta_o / N^{1/2}$, the crossover is essentially complete, and tunneling is blocked for almost all grains in an ensemble. Thus for large $N$, degeneracy blocking is a very powerful mechanism for suppressing coherent oscillations of $S$, since only a tiny spread $\delta\omega_o$ will be required to do the job. Now we look at this quantitatively.

The trivial case is of course where $\delta\omega_o = 0$, and the spectrum of $\{\sigma_k\}$ is organized in highly degenerate lines corresponding to different polarizations $\epsilon = \omega_o \Delta N / 2$. Then, as we discussed in the last subsection any zero polarization state of $\{\sigma_k\}$ gives an exact resonance for $\tilde{S}$, and the corresponding correlation function is an ideal one, i.e., we get $P_{\parallel\parallel}^{(0)}(t)$ as in (4.2); however only a small fraction of possible environmental states have $\Delta N = 0$, and for large $N$ this fraction is defined by the central limit theorem as $f = \sqrt{2/\pi N}$. Thus the corresponding spectral function is like (4.3) but with a total weight $f$, i.e.,

$$\chi''_o(\omega) \to \sqrt{2\pi / N} \delta(\omega - 4\tilde{\Delta}_\Phi) ; \quad (\delta\omega_o = 0).$$

(4.32)

We may simply disregard the contribution to $\chi''$ coming from states with nonzero $\Delta N$, because for $\omega_o \gg \tilde{\Delta}_{\text{eff}}$ these states are too far off the resonance. Thus, as we see from (4.32), in this special case the only effect of the coupling $\omega_o$ to the $N$ spins is a reduction of the spectral weight - otherwise the central spin behaviour is completely coherent. This is hardly surprising, since we have removed all decoherence mechanisms - we have no topological decoherence, no degeneracy blocking, and no orthogonality blocking! This case was also studied very recently by Garg [44] in an attempt to discuss experiments in magnetic grains.

Going now to the case where we do have some degeneracy blocking, it is useful to first study the regime where $\mu = \delta\omega_o N^{1/2} / \omega_o > 1$ (in real situations, this regime will almost always prevail in any case). Under this regime the different groups of environmental states will be spread out and overlap, and we will then get a continuous Gaussian distribution for the bias energy $\epsilon$:

$$W(\epsilon) = \frac{1}{\omega_o (2\pi N)^{1/2}} \exp\{-\epsilon^2 / (2\omega_o^2 N)\} ;$$

(4.33)

$$\epsilon = \sum_{k=1}^N \omega_k^\parallel \sigma_k^z .$$

(4.34)

This situation is depicted schematically in Figs.12a and 12b; Fig.12b also depicts a case where $\mu < 1$, i.e., with very small degeneracy blocking. The correlation function $P_{\parallel\parallel}^{(0)}(t)$ for an ensemble of central spins is now obtained by averaging the known analytic behaviour for the simple biased 2-level system. At high temperatures
\[ P_{\Theta \Phi}(t) = \int d\varepsilon W(e^{-\varepsilon / Z(\beta)} \left[ 1 - \frac{2\tilde{A}_\Phi^2}{\varepsilon^2 + 4\tilde{A}_\Phi^2} \left(1 - \cos(2t\sqrt{\varepsilon^2 + 4\tilde{A}_\Phi^2})\right)\right]) \]

\[ \rightarrow 1 - 2A \sum_{k=0}^{\infty} J_{2k+1}(4\tilde{A}_\Phi t). \]

\[ A = 2\pi \tilde{A}_\Phi / (2\omega_o \sqrt{2\pi N}) \equiv 2\pi f \cdot \tilde{A}_\Phi / 4\omega_o. \]

which is shown in Fig.13. (Integrating over \( \varepsilon \) we used the inequality \( W(0)\tilde{A}_\Phi \ll 1 \). We see that not only is there line-broadening, but also the amplitude of the oscillations, for the ensemble of central spins, is further reduced by a factor \( \tilde{A}_\Phi / 2\omega_o \), as compared with the trivial case in (4.32), and by a total factor \( A \) as compared with the ideal free central spin in (4.42). We should note at this point that in almost any realistic situation, a random distribution of values for \( \omega_k^2 \) for a single central spin will lead to results identical to the above, i.e., the ensemble average here is merely a device to define the distribution \( W(\varepsilon) \). The only difference is that for a single central spin one has to consider (4.36) as a statistical average over many experimental runs during a time which is longer then the NMR relaxation times in the nuclear subsystem.

Turning now to the general case, where the parameter \( \mu = \delta\omega_o N^{1/2}/\omega_o \) can take arbitrary values, we notice that the integral over \( \varepsilon \) in (4.36) is convergent on a scale defined by \( \tilde{A}_\Phi \), and this result will not change at all for any distribution which is flat around zero. Thus for \( \omega_o \gg \delta\omega_o N^{1/2} \gg \tilde{A}_\Phi \), i.e., \( \mu \ll 1 \) we have the same answer as before, but the amplitude \( A \) is now replaced by \( A = f \cdot \tilde{A}_\Phi / (2\sqrt{2\pi N\delta\omega_o}) = \tilde{A}_\Phi / (2\pi N\delta\omega_o) \). Thus we find that even a very small spread in the coupling constants will be sufficient to broaden the spectral line. Note that neither the energy dissipation nor the phase randomisation are responsible for this behaviour, which is totally due to the statistical average over the initial states of \( \{\tilde{\sigma}_k\} \).

\[ \chi''(\omega) = A \frac{8\tilde{A}_\Phi}{\omega \sqrt{\omega^2 - 16\tilde{A}_\Phi^2}} \eta(\omega - 4\tilde{A}_\Phi), \]

\section*{V. THE GENERIC CASE}

As we shall see, if we combine all 3 mechanisms just studied, and calculate \( P_M(t) \) for the full \( H_{eff} \), we get a very complex (but still a closed form) answer. Thus in order to make the derivations more transparent, we first combine the mechanisms in different combinations. This tactic is not just pedagogically useful - it also gives 3 more limiting cases which correspond to experimentally realistic situations. All this is done in Sections III A - III D. Finally, in III E, we deal with the residual inter-nuclear couplings mentioned in Section III D, Eq. (3.13). Within a particular polarisation group \( \Delta N \), the inter-nuclear dipolar couplings cause "diffusion in bias", simply by flipping random pairs of nuclei. We study the role this has on the general solution for the coherent part of \( P_{\Theta \Phi}(t) \) given in III D.

\subsection*{A. Projected Topological decoherence}

We start from the case of "projected topological decoherence", defined by the following special case of (3.11):

\[ H_{eff} = 2\Delta_o \tilde{\tau}_z \cos \left[ \Phi + \sum_{k=1}^{N} \alpha_k \tilde{n}_k \cdot \tilde{\sigma}_k \right] + \tilde{\tau}_z \sum_{k=1}^{N} \omega_k \tilde{\sigma}_k^z, \]

with the restrictions:

(i) The effective fields \( \omega_k^{(1)} = -\omega_k^{(2)} \), so \( \omega_k^+ = 0 \).

(ii) The couplings \( \omega_k \gg \tilde{\Delta}_o \), and all couplings are equal (thereby eliminating degeneracy blocking).

The second condition forces the polarization of the environment \textit{with respect to the central spin orientation} to remain constant. Now at first glance we have no orthogonality blocking at all in (5.1), since \( \omega_k^+ = 0 \) (compare section IV B), and so it looks as though \textit{only} zero polarisation group states will have any dynamics. However this is incorrect;
because now the dynamic couplings $\alpha_k$ can flip spins when $S$ flips, with amplitude $\alpha_k$; they play exactly the same role as the $\beta_k$ in pure orthogonality blocking (Section 4A.B), and so we can say that in (5.1), orthogonality blocking is generated by the $\alpha_k$. Thus, in the same way as in Eqs.(4.19) and (4.20) we write $P_{\parallel \perp}(t)$ as a weighted sum over $P_M(t)$, where now (cf. Appendix B.1):

$$P_M(t) = \int dx e^{-x^2} \sum_{m=-\infty}^{\infty} F_{\lambda'}(m) \int \frac{d\varphi}{2\pi} e^{i2m(\varphi-\varphi)} \left\{ 1 + \cos[4\tilde{\Delta}_o t J_M(2x\sqrt{\lambda - \lambda'}) \cos \varphi] \right\}$$

$$= \int dx e^{-x^2} \left\{ 1 + \sum_{m=-\infty}^{\infty} F_{\lambda'}(m) e^{i2m\Phi} J_{2m}[4\tilde{\Delta}_o t J_M(2x\sqrt{\lambda - \lambda'})] \right\} ,$$

where $F_{\lambda'}(m)$ is defined analogously to $F_{\lambda}(m)$ in (4.7), viz.,

$$F_{\lambda'}(m) = e^{-4\lambda'm^2} ;$$

$$\lambda' = \frac{1}{2} \sum_{k=1}^{N} \alpha_k^2(n_k)^2 ,$$

and we notice that $\lambda \geq \lambda'$. Eq.(5.2) can be interpreted in 2 ways, both of which are important. First, we can interpret it as an orthogonality-blocked expression, now with frequency scale

$$\Delta_M(\varphi, x) = 2\tilde{\Delta}_o \cos(\varphi) J_M(2x\sqrt{\lambda - \lambda'}) ,$$

which is then averaged over $\varphi$, to give phase randomisation (compare Eq.(4.9)). Alternatively we can regard it as an integration $\int dx$ over an already topologically decohered function $P_M^{(\text{top})}(t, \Delta_M(x))$, with $\Delta_M(x)$ now given by

$$\Delta_M(x) = \tilde{\Delta}_o J_M(2x\sqrt{\lambda - \lambda'}) ;$$

This is the result for $P_M(t)$; but now we can go through exactly the same arguments as for pure degeneracy blocking, to show that only $P_B(t)$ may behave coherently, with a weight $\sim \sqrt{2/\pi N}$. Again the exact expression is given by the weighted sum (4.19).

There are various interesting cases of (5.3) for $M = 0$. If $\lambda = \lambda'$ (i.e., $n_k$ is parallel to $\hat{z}$), then we go back to pure topological decoherence - the projection operator then commutes with the cosine operator. On the other hand if $\lambda' = 0$, we have pure orthogonality blocking as stated earlier, and in fact when $\lambda' = 0$, the parameter $\lambda$ plays the role of $\kappa$ in (4.22). Notice that whereas the case $\lambda = \lambda'$ can only occur accidentally, $\lambda' = 0$ is quite common - indeed it pertains to the model in Eqs. (6.1) and (6.2) which we will examine below in connection with real experiments.

We really begin to see the analogy with degeneracy blocking when $\lambda, \lambda' \gg 1$; just as with pure topological decoherence, $F_{\lambda'}(m)$ collapses to a Kronecker delta, and we get the universal projected topological decoherence form:

$$P_{\parallel \perp}(t) \rightarrow \int dx e^{-x^2} \left[ 1 + J_2[4\tilde{\Delta}_o t J_M(2x\sqrt{\lambda - \lambda'})] \right]$$

$$\chi''(\omega) \rightarrow \sqrt{\frac{2}{\pi N}} \int dx e^{-x^2} \left[ \frac{4}{16\tilde{\Delta}_o^2 J_6^2(2x\sqrt{\lambda - \lambda'}) - \omega^2} \right]^{1/2} \eta(4\tilde{\Delta}_o | J_0(2x\sqrt{\lambda - \lambda'}) | - \omega) ,$$

which generalizes the result of (4.16) for pure topological decoherence. Eq.(5.7) should be compared to (4.22).

We show in Figs.14 and 15 some results for $\chi''(\omega)$ for selected values of $\lambda - \lambda'$. The results are startling; even a very small value of $(\lambda - \lambda')$ significantly washes out pure topological decoherence; but for any large value of $\lambda'$, we never get back the pure orthogonality blocking spectrum.

**B. Orthogonality blocking plus degeneracy blocking**

We now look at what might be called "biased orthogonality blocking"; as explained in section 4.A.C, if we introduce a spread $\delta \omega$ about some $\omega_o$, then in effect we introduce a bias $\epsilon$ which destroys the degeneracy of the environmental states. Thus we now study the Hamiltonian
\[ H_{\text{eff}} = 2\hat{\Delta}_p \hat{\sigma}_x + \hat{t}_z \sum_{k=1}^{N} \omega_k^p \hat{\sigma}_k + \sum_{k=1}^{N} \omega_k^p \hat{m}_k \cdot \hat{\sigma}_k. \]  

(5.9)

with the spread in frequencies \( \omega_k = [(\omega_k^p)^2 + (\omega_k^z)^2]^{1/2} \) defined by Eq. (4.31). The effective bias \( \epsilon = \sum_{k=1}^{N} \omega_k^p \hat{\sigma}_k \) again, and now we would like to take account of the effect of this bias on the orthogonality blocking. Again, one starts by calculating a function \( P_M \) referring to polarisation subgroup \( M \), but this time in a bias \( \epsilon \). One finds (Appendix B.2):

\[
P_M(t, \epsilon) = 2 \int dx e^{-x^2} \left\{ 1 - \frac{\Delta^2_M(x)}{2E^2(x)} \left( 1 - \cos [2tE(x)] \right) \right\} = 2 \int dx e^{-x^2} P^{(0)}_{\eta\eta}(t, \Delta_M(x), \epsilon).
\]  

(5.10)

\[ E^2(x) = \epsilon^2 + \Delta^2_M(x). \]  

(5.11)

This expression could have been guessed without any calculation - it is just the usual orthogonality blocking average \( \int dx \), but now for a biased system (instead of, as in pure orthogonality blocking, an unbiased expression - cf. [4.22]).

To get \( P_{\eta\eta}(t) \), we must now both integrate over bias and sum over \( M \). Thus the generalisation of our previous weighted average (4.19) is just

\[
P(t; T) = \int d\epsilon W(\epsilon) \frac{e^{-\beta \epsilon}}{Z(\beta)} \sum_{M=1}^{N} P_M(t, \epsilon - M\omega_\circ). \]  

(5.12)

However, yet again, we use the usual argument that only \( P_0 \) may produce coherent response. Assuming again that the spread in couplings satisfies \( \mu = N^{1/2} \delta \omega_\circ/\omega_\circ > 1 \) (a trivial modification is necessary for \( \mu \omega_\circ/\Delta_\circ \gg 1 \) but \( \mu < 1 \), as discussed in section [Y, C]), we then get for \( M = 0 \)

\[
P_0(t) = 1 - 4A \int dx e^{-x^2} \left| J_0(2\sqrt{\kappa}x) \right| \sum_{k=0}^{\infty} J_{2k+1} \left[ 4\Delta_\Phi \left| J_0(2\sqrt{\kappa}x) \right| t \right] \]  

(5.13)

\[
= 1 - 2 \int dx e^{-x^2} 2A(x) \sum_{k=0}^{\infty} J_{2k+1} \left[ 2\Delta_0(x)t \right],
\]  

(5.14)

where the \( x \)-dependent spectral weight is

\[ A(x) = AJ_0(2\sqrt{\kappa}x). \]  

(5.15)

Yet again, by comparing with the previous results for pure orthogonality blocking and pure degeneracy blocking, we could have guessed this (compare Eq. [4.36]); we have just done an "orthogonality average" over a "biased averaged" expression for the free system with \( x \)-dependent tunneling frequency \( \Delta_0(x) \).

The expression for \( \chi''_{M=0}(\omega) \) follows immediately; we have

\[ \chi''_{0}(\omega) = \frac{1}{\omega} \int dx e^{-x^2} 4A(x) \frac{\Delta_0^2}{[\omega^2 - 4\Delta_0^2(x)]^{1/2}} \eta(\omega - 2) \left| \Delta_0(x) \right|, \]  

(5.16)

Figs.16 & 17 show some representative plots for this coherent part of \( \chi''(\omega) \); it is in fact almost completely incoherent, with total spectral weight

\[ \int_{-\infty}^{\infty} d\omega (\omega/2\pi) \chi''(\omega) = 2A \int dx e^{-x^2} \left| J_0(2\sqrt{\kappa}x) \right| \]  

\[ = \frac{2\Gamma(3/4) A}{\pi^{3/2} \kappa^{1/4}}; \]  

(5.17)

a result which is very accurate even for \( \kappa \sim 0.02 \). Of course, once we include all other polarization sectors \( M \neq 0 \), the total power increases - in fact it now becomes \( \sim A\kappa^{3/4} \) for large \( \kappa \), since \( \sim \kappa^{1/2} \) different polarization sectors contribute. However none of this is coherent either.
C. Pure topological decoherence plus degeneracy blocking

Here we discuss the case when the central spin dynamics is governed by pure topological decoherence is a biased system. With what we have calculated already the answer is readily obtained as follows. We note that the correlation function in Eq. (A10) is clearly given as a coherent correlation function \( P_{\uparrow\uparrow}(t) \) (see (A3)) integrated over \( \varphi \) and summed over \( m \). Now, if we add the bias, all we need to do is to substitute for the unbiased correlation function with the non-interacting biased one from Eq. (A10). Thus we have for the biased topological decoherence

\[
P_{\uparrow\uparrow}(t) = \sum_{m=-\infty}^{\infty} F_{\lambda}(m) \int \frac{d\varphi}{2\pi} e^{i2m(\Phi - \varphi)} \left( 1 - \frac{\Delta_0^2(\varphi)}{\epsilon^2 + \Delta_0(\varphi)} \right) \left( 1 - \cos \left[ 2t\sqrt{\epsilon^2 + \Delta_0^2(\varphi)} \right] \right) + \text{inc.} , \quad (5.18)
\]

Integrating over the bias with the usual assumption about the distribution function we immediately find

\[
P_{\uparrow\uparrow}(t) = 1 - \sum_{m=-\infty}^{\infty} F_{\lambda}(m) \int \frac{d\varphi}{2\pi} e^{i2m(\Phi - \varphi)} 2A(\varphi) \sum_{k=0}^{\infty} J_{2k+1}[2\Delta_0(\varphi)t] + \text{inc.} , \quad (5.19)
\]

for the correlation function, with \( \Delta_0(\varphi) = 2\Delta_0 \cos \varphi \) as before, and \( A(\varphi) = A \cos \varphi \); and

\[
\chi''(\omega) = \frac{2A}{\omega} \sum_{m=-\infty}^{\infty} F_{\lambda}(m) \int \frac{d\varphi}{2\pi} e^{i2m(\Phi - \varphi)} \frac{\cos^2 \varphi}{[(\omega/4\Delta_0)^2 - \cos^2 \varphi]^{1/2}} \eta(\omega/4\Delta_0 - |\cos \varphi|) , \quad (5.20)
\]

for the spectral function.

One might be confused whether this calculation applies to any real system, because on one hand the pure topological decoherence implies no (or at least very weak \( \omega_k < \Delta_0 \)) interaction between the central spin and its environment; on the other hand including degeneracy blocking we assume that the coupling is strong enough to move the system out of the resonance. How one can reconcile this? Below we demonstrate that this situation is described as a special limiting case of our effective Hamiltonian.

Suppose we have a very weak interaction between the central spin and its environment so that \( \omega_k \ll \Delta_0 \). Since \( \Delta_0 \) is exponentially small as compared to \( \Omega_0 \), we are dealing with the case of extremely small \( \alpha_k \sim \omega_k/\Omega_0 \ll \Delta_0/\Omega_0 \). Imagine now that we have a really macroscopic number of environmental spins contributing to the decoherence so that the small value of \( \alpha_k \) is compensated or even superseded by \( N \) so that \( \lambda \sim NA_k^2 \) is of order unity or even large. One immediately understands that in this case it is impossible for the central spin to tunnel and not to flip some environmental spins, but since \( \omega_k \) is much smaller than \( \Delta_0 \), we are still in resonance (if \( \omega_k \lambda^{1/2} < \Delta_0 \)) and may neglect energy dissipation.

It is quite obvious, however, that such a large number of microscopic spins both the parameter \( \mu = \delta N^{1/2}/\omega_0 \gg 1 \), and the level distribution width \( N^{1/2}/\omega_0 \gg \Delta_0 \) are very large, and we do have to consider tunneling in a biased system. The reason why the two mechanisms are combined so easily is that the bias is produced by the total effect of all spins, while only an extremely small fraction of them flips when the central spin tunnels. As described elsewhere the numbers are such that this special case has direct application to the environmental spin decoherence in SQUIDs. For large parameter \( \lambda \) Eq. (5.20) simplifies to

\[
\chi''(\omega) = \frac{2A}{\omega} \int \frac{d\varphi}{2\pi} \frac{\cos^2 \varphi}{[(\omega/4\Delta_0)^2 - \cos^2 \varphi]^{1/2}} \eta(\omega/4\Delta_0 - |\cos \varphi|) , \quad (5.21)
\]

which can be expressed in terms of Elliptic functions.

D. Combining the 3 mechanisms: the generic case

With the preceding studies we hope that the following basic message has come through, viz., that combining combinations of the 3 mechanisms simply boils down to combining 3 weighted averages. These averages are

\[
(a) \text{ A "topological phase average" given by } \sum_{m=-\infty}^{\infty} F'_{\lambda}(\nu) \int \frac{d\varphi}{2\pi} e^{i2m(\Phi - \varphi)} ; \quad (5.22)
\]
(b) An "orthogonality average" given by
\[ 2 \int_0^\infty dx x e^{-x^2} ; \] (5.23)

(c) A "bias average" \[ \int d\nu e^{-\beta \nu} Z(\beta) . \] (5.24)

These averages apply to any polarisation group $M$; the final expression for $P_{\nu\nu}(t)$ is then given by averaging the quantity
\[
P_{\nu\nu}(t; \Delta_M(\varphi, x); \epsilon) = 1 - \frac{\Delta_M^2(\varphi, x)}{E_M^2(\varphi, x)} \sin^2(E_M(\varphi, x)t) ,
\] (5.25)
i.e., the free biased correlator for a splitting $\Delta_M$:
\[
\Delta_M(\varphi, x) = 2 \tilde{\Delta}_o \cos \varphi J_M(2x\sqrt{\lambda - \lambda'}) ,
\] (5.26)
\[
E_M^2(\varphi, x) = \epsilon^2 + \Delta_M^2(\varphi, x) .
\] (5.27)

Thus, using this prescription, we can immediately write down almost the final generic case; we have, assuming that in the effective Hamiltonian both $\omega_k^\perp = 0$ and $\xi_k = 0$
\[
H_{\text{eff}} = 2 \tilde{\Delta}_o \hat{\tau}_x \cos \Phi + \sum_{k=1}^N \alpha_k \hat{n}_k \cdot \hat{\sigma}_k + \hat{\tau}_z \sum_{k=1}^N \frac{\omega_k}{2} \hat{\sigma}_k^z .
\] (5.28)
the full correlator is given by
\[
P_{\nu\nu}(t; T) = \int d\nu e^{-\beta \nu} Z(\beta) \sum_{M=-N}^N P_M(t, \epsilon - M\omega_0) ;
\] (5.29)
\[
P_M(t; \epsilon) = 2 \int_0^\infty dx e^{-x^2} \sum_{\nu=\infty}^{\infty} F_M(\nu) \int \frac{d\varphi}{2\pi} e^{i2\nu(\varphi - \varphi')} \left[ 1 - \frac{\Delta_M^2(\varphi, x)}{E_M^2(\varphi, x)} \sin^2(E_M(\varphi, x)t) \right],
\] (5.30)
at any temperature. It should be emphasized that this expression is exact provided the $\alpha_k$ are small and $\omega_k \gg \Delta_\Phi$ (which is virtually always the case in nature).

If we are only interested in possible coherence, then it suffices only to look at $M = 0$ term in this expression, for reasons we have previously discussed. Carrying out the bias average then just gives the obvious generalization of Eq. (5.14), i.e.,
\[
P_{\nu\nu}(t) = 1 - 2 \int dx e^{-x^2} \sum_{\nu=\infty}^{\infty} F_M(\nu) \int \frac{d\varphi}{2\pi} e^{i2\nu(\varphi - \varphi')} A(\varphi, x) \sum_{k=0}^{2k+1} [2\Delta_0(\varphi, x)t] + \text{inc} ;
\] (5.31)
\[
A(\varphi, x) = A \cos \varphi J_0(2x\sqrt{\lambda - \lambda'}) ;
\] (5.32)
and an equally obvious generalization for $\chi''(\omega)$
\[
\chi''(\omega) = \frac{2}{\omega} \int dx e^{-x^2} \sum_{\nu=\infty}^{\infty} F_M(\nu) \int \frac{d\varphi}{2\pi} e^{i2\nu(\varphi - \varphi')} \times \frac{A(\varphi, x)\Delta_0(\varphi, x)}{[\omega^2 - 4\Delta_0^2(\varphi, x)]^{1/2}} .
\] (5.33)

These results for the effective Hamiltonian in (5.10) are all we will derive here. We now argue that these are all we will ever really need for qualitative understanding. We hope it is clear that if we put the $\omega_k^\perp$ term back into $H_{\text{eff}}$, it would be equally straightforward to calculate $P_{\nu\nu}(t)$, since the role of the $\beta_k$ in the calculations is analogous to that of the $\alpha_k$, but even simpler to deal with. The only term we have still not dealt with is the term $\xi_k \hat{v}_k \cdot \hat{\sigma}_k$ which also appears in the cosine term in $H_{\text{eff}}$. In Appendix C.3 we show how this can be included - the final expression is a monster, but can easily be put onto a computer for comparison with real systems (as in the next Section). We also show plots here of $\chi''(\omega)$ in Fig.18 and Fig.19 for a few representative values of $\lambda - \lambda'$; one should again notice how rapidly decoherence sets in, and how large is the power reduction factor.
E. Nuclear Spin Diffusion

Finally we comment on the role of the role of the very weak dipolar/Nakamura-Suhl couplings between the nuclei (Eq. (3.12)). There is potentially very important decoherence effect that can come from these interactions, because the dipolar coupling can cause pairwise flipping (\(|\uparrow\downarrow\rangle \rightarrow |\downarrow\uparrow\rangle\)). Even though this interaction is weak (it is of strength \(T_2^{-1}\), and will be in the range \(10^3 - 10^6\) Hz - see Table I) it is important because it allows both real space spin diffusion and diffusion of the system in a bias energy \(\epsilon\), within a given polarisation group. For a set of \(N\) nuclei the pairwise flips occur at a rate \(\sim NT_2^{-1}\); each flip will change the bias energy by \(\sim \delta\omega_o\) in energy, and the bias will random walk throughout the energy range \(N^{1/2}/\delta\omega_o = \mu\omega_o\) of the polarisation group. Now suppose \(\Delta N = M\), so that transitions of \(\vec{S}\) can only proceed if the time it takes \(\epsilon(t)\) to diffuse out of the energy window around resonance, of width \(\Delta M\), exceeds the "waiting time" \(\Delta t_M^{-1}\) necessary for the transition to proceed. Since in a time \(\Delta t_M^{-1}\), roughly \(N/T_2\Delta M\) pairwise flips occur, \(\epsilon(t)\) will change by roughly \(\delta\omega_o(N/T_2\Delta M)^{1/2}\) in this time, and so coherence requires

\[
\Delta t_M^3 \gg \frac{N}{T_2}(\delta\omega_o)^2 .
\]  

(5.34)

If the spread \(\delta\omega_k\) arises itself from Nakamura-Suhl interactions between the nuclei of the same kind, \(T_2^{-1} \sim \delta\omega_k\), and this criterion becomes \(\Delta M \gg T_2^{-1}N^{1/3}\). These criteria are very hard to fulfill unless \(M\) is small, i.e., for the zero polarisation group. One obvious way to avoid this nuclear spin diffusion decoherence mechanism is to choose a system like Fe or Ni in which nuclear spin isotopes are rare (cf. Table I). Then \(T_2^{-1}\) will be very small, and coherence may be visible for quite large \(N\) (and even larger \(S\), of course).

In the opposite case, when \(\epsilon(t)\) fluctuates too rapidly for tunneling, we will simply get incoherent relaxation of \(P_M(t)\), with a correlation time \(\tau_M = (2\Delta t_M^2/\mu\omega_o)^{-1}\); or, defining a correlation time \(\tau_M(x)\) via

\[
\tau_M^{-1}(x) = \frac{2\Delta t_M^2(x)}{\mu\omega_o} ,
\]  

(5.35)

we would find a completely incoherent form for \(P_{\parallel\parallel}(t)\):

\[
P_{\parallel\parallel}(t) = 2\sum_M w(M;T) \int dx x e^{-x^2} \exp\{-t/\tau_M(x)\} ,
\]  

(5.36)

(and obvious generalisation, if necessary, to include integration over \(\varphi\)).

This concludes our analysis of the central spin model. We should emphasize that whereas in this paper we concentrated on the coherence problem, the general formalism we have constructed, and the results we established, can also be used in analysing any of the low-\(T\) relaxation properties of \(\vec{S}\). The generalisation to a finite applied field, for example, can be done straightforwardly, starting from our generic expression.

VI. A REALISTIC EXAMPLE

Readers (particularly experimentalists) may feel that in spite of these analytic results, there is still a very long path to follow from the bare, untruncated Hamiltonian \(H_o(\vec{S})\), with some set of couplings to nuclear spins, paramagnetic spins, etc., to our final answers. We therefore, in this section take a specific bare Hamiltonian, viz.,

\[
H(\vec{S}, \{\vec{I}_k\}) = H_0(\vec{S}) + \frac{1}{2\vec{S}} \sum_k \omega_k \vec{S} \cdot \vec{I}_k ,
\]  

(6.1)

\[
H_0(\vec{S}) = -K_\parallel s_z^2 + K_\perp s_y^2 ,
\]  

(6.2)

with a range of couplings \(\omega_k\) around a central hyperfine coupling \(\omega_o\) between \(\vec{S}\) and the nuclear spins \(\vec{I}_k\). We then solve this problem almost completely, by giving the analytic forms for \(P_{\parallel\parallel}(t)\) and \(\chi''(\omega)\). We hope that this will make it clear how one may deal with whatever realistic central spin Hamiltonian that Nature may care to throw at us.

We observe that this model Hamiltonian actually provides a very good description of magnetic grains (compare the examples in section II.B.) provided we ignore:

(a) The coupling of the grain order parameter to both nuclei and electron spins outside the grain;
Thus to find \( \delta \omega \), the effective Hamiltonian is estimated from simple magnetostatics to be present in a bulk sample, and so NMR linewidth measurements in the bulk will miss this effect. Its size can be

\[
\sim 10^{-6} \text{kHz}
\]

in which the parameters are given by

\[
\frac{\Delta}{\Omega} = \frac{\alpha}{S}
\]

\[
\Phi = \frac{\pi\omega}{2\Omega} (\hat{x}, \hat{y})
\]

\[
\xi_k \mathbb{v}_k = -\frac{\pi\omega}{2\Omega} (\hat{x}, \hat{y})
\]

and the unit vector \( \hat{v}_k \) is \( \hat{z} \). As just discussed, the parameter \( \omega_k \) (and its spread) is accessible experimentally using NMR, as is \( \Delta \) (which is unrenormalised here, for weak interactions). If we can determine experimentally the values of \( K_\parallel \) and \( K_\perp \), we can determine \( \Omega \) via

\[
\Omega = \frac{2}{S} (K_\parallel K_\perp)^{1/2}
\]

or, alternatively, \( \Omega \) can be determined directly from microwave absorption experiments (which "rock" the central spin back and forth in one of the degenerate wells).

Equation (6.3) corresponds to the case of projected topological decoherence with no orthogonality blocking, but degeneracy blocking coming from the spread \( \delta \omega_k \). This case was completely solved in section V; these equations are valid for all the examples we will discuss, since they assume \( \mu \omega_k / \Delta = N^{-1} / \delta \omega_k / \Delta > 1 \). To see what we get, let us look at two examples:

**Example A: TbFe\(_3\) grains**: We take this example as a representative case in which topological decoherence is very important; as noted in section II B, one has for this system \( \Omega \sim 50 \text{GHz} \), and the quadrupolar split lines have \( \alpha_k \sim 0.08, 0.07, \) and 0.05 respectively. Now since no experiments to look for coherence have been done on this system, we will simply imagine one in which we choose \( S = 10^4 \), and \( \Delta = 1 \text{MHz} \) (this value of \( \Delta \) may be optimistic, but is not in conflict with equation (6.2), given the uncertainty in the ratio of \( K_\parallel / K_\perp \) for TbFe\(_3\)). We also note that the
spread in $\alpha_k$ caused by the Fe$^{57}$ nuclei will give $\delta \omega_k \sim 50 \text{ MHz}$, so that with $N_{Tb} = 2.5 \times 10^3$ we have $\mu \sim 1$. Thus we have the following parameters

\[
\alpha = \sqrt{2\xi} = 0.05, \ 0.07, \ 0.08 \\
\mu \sim 1 \\
(\omega_o/\Delta_o)\mu \sim 3 \times 10^3 \\
(TbFe_3),
\]

leading to the values

\[
\lambda \sim 10; \quad (\lambda' = 0).
\]

The form of $\chi''(\omega)$ for this case is shown in Fig.20 (again only the $M = 0$ contribution is shown). We see that there is complete loss of coherence, and extremely strong suppression of the absorption power ($A \sim 10^{-5} - 10^{-6}$). As noted previously in Sec. III.B adding in the $M \neq 0$ contributions will increase the total spectral weight, but this contribution will be even more incoherent and centered around $\omega = 0$. Nuclear spin diffusion effects will of course change this somewhat.

**Example B: Ferritin grains**

This is a more interesting example, since topological decoherence is unimportant here. We shall first analyze the experiments of Awschalom et al. [17] whilst ignoring the interaction between the ferritin molecules. As we saw in section III.B, ferritin is an antiferromagnetic macromolecule with 4500 Fe$^{57}$ ions. It also has an excess ferromagnetic moment with a somewhat uncertain value (estimates range between 217 and 640$\mu_B$). In reality, because of this moment, ferritin molecules will interact. There will also be a coupling to nuclear and electronic moments outside the ferritin, which we ignore here.

We are then left with an Fe hyperfine coupling $\omega_k \sim 64 \text{ MHz}$; there will also be other hyperfine couplings to the $H^1$ protons in the $H_2O$ in ferritin; we also ignore this for the moment (but see our discussion below). We assume a $\delta \omega_k \sim 200 \text{ kHz}$ arising from a combination of Suhl-Nakamura and non-uniform dipolar interactions - this is probably an underestimate, but the quantity has not yet been measured, for ferritin molecules, as far as we know.

According to Awschalom et al., the anisotropy constants $K_\parallel$ and $K_\perp$ obey $(K_\parallel K_\perp)^{1/2} \sim 1.72 \text{ Tesla}$, so we shall assume a value for $\Omega_0$ of 40 $\text{ GHz}$, as a rough estimate [45]. Finally, the experiments of Awschalom et al. claim that a resonance they see of frequency just under 1 $\text{ MHz}$ corresponds to coherent tunneling; therefore we assume $\Delta_o \sim 1 \text{ MHz}$. We thereby arrive at the following numbers, using (6.4), (6.5), and the definition of $\mu$:

\[
\alpha = \sqrt{2\xi} \sim 2 \times 10^{-3} \\
\mu = N^{1/2}\delta \omega_k/\omega_0 \sim 2 \times 10^{-2} \\
(\omega_o/\Delta_o)\mu \sim 2,
\]

where we note that there will be $\sim 100$ Fe$^{57}$ nuclei in the molecule, so $N^{1/2} = 10$. Now from (6.8) we find

\[
\lambda \sim 4 \times 10^{-5}; \quad (\lambda' = 0); \quad (\text{Ferritin}),
\]

which rules out topological decoherence; since we assume $\beta_k = 0$ (perfect sample) this leaves only degeneracy blocking, and since $(\omega_o/\Delta_o)\mu > 1$ we can immediately apply the result (4.38) in the text, which is depicted in Fig.13. If one compares the curve in Fig.13 with the data of Awschalom et al. [17], one is immediately surprised that the lineshape in the experiments is not at all that different from the calculation. Now it is important to understand that the reason for the sharp theoretical lineshape in Fig.13 is precisely because topological decoherence and orthogonality blocking have been dropped. It will be immediately obvious, on referring to Table I, that the value of the dimensionless topological decoherence parameter $\lambda$ is extremely low for ferritin; the value of 10 for TbFe$3$ is more normal. If we can ignore any orthogonality blocking, then it rather seems as though the experimentalists have hit upon an almost ideal system for the investigation of MQC in magnets (it could be made even better if one could isotopically purify Fe$^{57}$ in ferritin, to get rid of all Fe$^{57}$ nuclei). Do our calculations then support the thesis of Awschalom et al., to have seen MQC in ferritin? At this point we feel that one must be cautious. First, we notice there is a strong reduction, by a factor $A = \Delta_o/(2\omega_o \sqrt{2\pi N}) \sim 3.5 \times 10^{-4}$, in the total spectral weight of this peak, as compared to the situation where we ignore the nuclear spins (notice that the question of power absorption in these experiments has already caused some controversy [13][14]). This absorption is a consequence of the restriction to the $M = 0$ polarization sector, and the requirement of near resonance.

Perhaps more seriously, it is not obvious that there really is no orthogonality blocking in the ferritin. The *intrinsic* blocking will be very small - the only obvious other sources are either the dipolar interactions between the ferritin...
molecules, "loose spins" on the surface of the ferritin, or strain fields in the sample. The effect of dipolar fields is probably small, but the other effects are difficult to quantify - we simply call attention to the severe effect on the lineshape that will be wrought by even a fairly small amount of orthogonality blocking (see Fig.16, for $\kappa = 2$). Orthogonality blocking will also further reduce the spectral weight (Eq.(5.17)). On the other hand nuclear spin diffusion between the protons may be important.

Thus there is apparently a disagreement between the theory and experiment for ferritin. Nevertheless it may not be insurmountable, provided the discrepancy concerning power absorption can be dealt with [18,46], and provided one can rule out any significant orthogonality blocking, and we certainly do not think that one can yet rule out MQC as a possible explanation of the experimental results. Another question that needs further investigation is the role of the dipole interaction between the ferritin molecules - this may well lead to some kind of cooperative behaviour between the ferritin molecules. Elsewhere we have also suggested [3] a search for a "spin echo" which would help to reveal the source of the experimental resonance at $\sim 1 MHz$.

It should also be noted that very recently Garg has given an analysis [48] of the effects of the nuclear spins on coherence in ferritin molecules. Garg concludes (in agreement with our earlier papers [1,3]), that the experiments do not agree with the theory. However his reasoning and detailed results are different, and are based upon the reduction in the total spectral weight of the signal (the factor $f$ in Eq.(5.37), where $f = (2/\pi N)^{1/2}$; recall this factor comes solely from the assumption of zero nuclear spin polarization). In effect the analysis of Garg ignores topological decoherence ($\alpha_k = 0$), ignores orthogonality blocking ($\kappa = 0$) and it ignores degeneracy blocking ($\delta \omega_k = 0$). He therefore arrives at a set of sharp lines for the spectrum of $\chi''(\omega)$.

However, as we have just seen this is not realistic. Dropping $\alpha_k$ is justified for ferritin (but not for most other systems - compare the values for the examples in section II.B), but we see that the spectrum in Fig.13 is not a sharp Lorentzian line. Thus the analysis of Garg misses the crucial physical mechanism (of decoherence); his lineshape is one of completely coherent motion for the ferritin Néel vector (albeit with a reduced total signal amplitude). We note moreover that the amplitude reduction factor found by Garg is just the "bare" value $f$, which is incorrect; the correct reduction factor is that given in Eq.(4.37), for pure degeneracy blocking (or (5.17), if orthogonality blocking is also present).

A further point, upon which we will elaborate in detail in another paper dealing with finite field effects, is that the field dependence of $\chi''(\omega)$ will be quite different from that of a 2-level system, even one coupled to a bath of oscillators. This is because degeneracy blocking causes $\chi''(\omega)$ to vary on an energy scale determined by $W(\epsilon = \gamma e S H_o)$, which has nothing to do with $\tilde{\Delta}_o$ and is usually much greater.

We hope that from our analysis of these 2 examples the reader will now have a fairly concrete idea of how our effective Hamiltonian is used in the analysis of a real central spin; in fact with the analytic results we have given, it should be possible for experimentalists to produce plots of $\chi''(\omega)$ for any given system, provided the relevant parameters are known.

VII. CONCLUSIONS

In this paper we have given an analysis of the behaviour of a central spin $\vec{S}$, coupled in an essentially arbitrary way to a spin environment. That one can do this, and moreover recover closed analytic forms for the correlation functions $P_{\vec{S}}(t)$ and $\chi''(\omega)$, is perhaps surprising. It was certainly not obvious to us when this work was initiated (cf. ref. [1]); although the 3 mechanisms involved in the decoherence of $P_{\vec{S}}(t)$ are already contained in the analysis of ref. [3], it has taken some time to find closed analytic forms for all cases. It is perhaps even more surprising when one examines the equation produced by the only other method we could think of using for this problem, viz., the Bethe ansatz (we are dealing with a set of $N$ coupled 1-dimensional problems here [47], with in general random couplings). However examination of the relevant Bethe ansatz equations reveals them to be apparently intractable.

Our solution relies on our novel technique of introducing "operator instantons" in the Hilbert space of the environmental spins, and then separating out the relevant energy scales. This then allowed identification of 3 important limiting cases of the general problem, each of which involved a single specific physical mechanism. These mechanisms were "topological decoherence" (already completely solved in ref. [3]), and "degeneracy blocking" and "orthogonality blocking", whose essential physics was also described in ref. [3]. The solution is then achieved by combining these 3 mechanisms; this allows full coverage of the parameter space when there is decoherence without energy dissipation. In this paper we have used these methods to examine specifically the coherence properties of $\vec{S}$ in zero external field. This allows us to deal only with the zero polarization sector of the environment. In fact a fairly trivial extension of these methods can be used to deal with the finite field case, and to include all polarisation sectors. This then yields
a theory of the relaxation of $\vec{S}(t)$ (including tunneling of $\vec{S}(t)$) in an applied field, or in its absence. This will be discussed in detail in another paper.

As a result of these investigations we find that the spin environment acts very differently from the usual "oscillator bath" environments, and has a far more powerful suppressive effect on quantum coherence at low temperature. One can also analyze the effect on coherence, of spin environments, for other systems. For example, an analysis of possible macroscopic quantum coherence for SQUIDs yields quite enormous values of our parameters $\lambda$, $\lambda'$, etc. (typically $\lambda > 10^5$). This implies that the previous analyses of SQUIDs, with a view to observing such "MQC", are far too optimistic, and that in fact it is unlikely that it will be seen in any superconductor (although the prognosis is more hopeful if the superconductor lacks nuclear spins). This work will be reported elsewhere.

These results lead us to suspect that in nature the overwhelmingly dominant mechanism of decoherence on the mesoscopic or macroscopic scales is likely to come from environmental spins, particularly nuclear spins. Whilst this may not change the fundamental attitude of most physicists towards the foundations of quantum mechanics, or the "measurement problem", it is certainly an interesting and non-trivial discovery about the physics of decoherence in the real world. It is also of fundamental importance for efforts to either observe MQC, or to at least build devices which operate coherently on more than just the 1- or 2-particle level. As noted in the introduction, it very severely restricts the possible physical systems in which such coherence might be seen - the only really clear example that avoids the problem discussed here is superfluid $^4$He - again, the details of this example will be discussed elsewhere.

Finally, let us note again that once an applied field starts to noticeably bias the potential $H_o(\vec{S})$, things change a great deal. Our problem then becomes effectively one of the effect of nuclear and other spins on the crossover to dissipative tunneling of $\vec{S}(t)$. This is a quite different problem from the one we have just analyzed, and we will return to it in another paper.

VIII. ACKNOWLEDGEMENT

This work was supported by NSERC in Canada, by the International Science Foundation (MAA300), and by the Russian Foundation for Basic Research (95-02-06191a). Some of it was carried out at the Aspen Center for Physics. We would like to thank I. Affleck, B.G. Turrell, and W.G. Unruh for some very helpful discussions.

APPENDIX A: DERIVATIONS FOR THREE LIMITING CASES

We describe here the derivations of the key formulae in section [14]. We begin with a few remarks on the free central spin, and the derivation of $P_{\uparrow\uparrow}(t)$ for it; it will then be seen how the expressions (4.4), (4.5) involve obvious generalizations of the free spin algebra. Then we give an account of the lengthy derivation required to obtain the expression (4.22) for pure orthogonality blocking.

1. Free central spin, plus topological and bias effects

Once $H_o(\vec{S})$ has been truncated, an expression for $P_{\uparrow\uparrow}(t)$ can be derived using standard instanton techniques [12-14], employing the free amplitude $K_o^\pm = \Delta_o \hat{\tau}_x \exp\{\pm i \pi S\}$. In the absence of the phase $\pi S$, one has a "2-level system" correlation function $P_{TLS}(t)$

$$P_{TLS}(t) = \frac{1}{2} \left[ 1 + \cos(2 \Delta_o t) \right] ,$$

which is derived in the instanton framework by summing over all paths involving an even number of flips, with an amplitude $i \Delta_o dt$ to flip in a time $dt$, giving

$$P_{TLS}(t) = \frac{1}{2} \left\{ 1 + \sum_{s=0}^{\infty} \frac{(2i \Delta_o t)^{2s}}{(2s)!} \right\} .$$

Adding the phase $\pi S$ clearly changes the flip amplitude in time $dt$ to $2i \Delta_o \exp\{\pm i \pi S\}$, and adding the $\pm$ processes then gives
which can, if one wishes, be derived by summing over all paths with an even number of flips, and summing over all combinations of clockwise and counterclockwise flips:

\[
P^{(0)}_{\uparrow\downarrow}(t) = \frac{1}{2} \left[ 1 + \cos(4\Delta_o \cos \pi S) t \right],
\]

(A3)

where \( \Phi_o = \pi S \) for the free spin. This is equation (4.2) of the text.

The generalization to topological decoherence is now easy. We either observe that the interference factor \( e^{i\pi S} + e^{-i\pi S} = 2 \cos \pi S \) in (4.3) will now be generalized via

\[
\pi S \to (\Phi + \sum_{k=1}^{N} \alpha_k \vec{n}_k \cdot \vec{\sigma}_k); \quad \text{(pure topological)},
\]

(A5)

which gives Eq. 4.4 (including the trace over spin states); or we simply include the extra phase (4.3) into (4.4), to give Eq. 4.5 of the text.

Now consider the modification introduced by a bias \( \epsilon \). For the 2-level system this is discussed very thoroughly in one way by Leggett et al. (ref. 12, Appendix A). We use a different representation here. Consider first the amplitude \( A^{TLS}_{\uparrow\downarrow}(t) \) for a return to an initial state in the biased case; this is clearly

\[
A^{TLS}_{\uparrow\downarrow}(t, \epsilon) = \sum_{n=0}^{\infty} (-i\Delta_o)^{2n} \int_0^t dt_{2n} \ldots \int_0^{t_2} dt_1 e^{i\epsilon(t-t_{2n})-\epsilon(t_{2n}-t_{2n-1})+\ldots+\epsilon t_1},
\]

(A6)

since the action coming from a pause of length \( dt \) is \( e^{\pm i\epsilon dt} \). We now write this in Laplace transform form as

\[
A^{TLS}_{\uparrow\downarrow}(t, \epsilon) = \int_{-i\infty}^{i\infty} e^{pt} A^{TLS}_{\uparrow\downarrow} (p, \epsilon).
\]

(A7)

\[
A^{TLS}_{\uparrow\downarrow}(p, \epsilon) = \frac{1}{p-i\epsilon} \sum_{n=0}^{\infty} \left( \frac{(i\Delta_o)^2}{p^2+\epsilon^2} \right)^n = \frac{1}{p-i\epsilon} \frac{p^2+\epsilon^2}{p^2+\epsilon^2}.
\]

(A8)

\[
E = \epsilon^2 + \Delta_o^2 1/2,
\]

(A9)

which gives the standard answer for \( P^{(0)}_{\uparrow\downarrow}(t, \epsilon) \):

\[
P^{TLS}_{\uparrow\downarrow}(t, \epsilon) = \int_{-i\infty}^{i\infty} dp_1 dp_2 \frac{e^{(p_1+p_2)t}}{(p_1-i\epsilon)(p_2-i\epsilon)} \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} \left( \frac{(i\Delta_o)^2}{p_1^2+\epsilon^2} \right)^n \left( \frac{(i\Delta_o)^2}{p_2^2+\epsilon^2} \right)^m
\]

\[= 1 - \frac{\Delta_o^2}{2E^2} \left(1 - \cos 2Et\right) = 1 - \frac{\Delta_o^2}{2E^2} \sin^2 Et
\]

(A10)

It is then obvious that in the case of the free spin, for a bias that does not distinguish between the clockwise and counterclockwise flips, \( P_{\uparrow\downarrow}(t, \epsilon) \) will look exactly like (4.10), except that \( \Delta_o \to 2\Delta_o \cos \pi S \). This representation avoids the difficulty in the usual representation which comes essentially from the square root \( E = \epsilon^2 + \Delta_o^2 1/2 \) in the cosine.

2. Orthogonality blocking

We consider the situation described in section 5B. For the spin \( \vec{\sigma}_k \), the "initial" and "final" local fields are \( \gamma_k^{(1)} \) and \( \gamma_k^{(2)} \). We will begin, to make the derivations easier, by assuming that the angle relating \( \gamma_k^{(1)} \) and \( \gamma_k^{(2)} \) is close to \( \pi \) for all the \( \vec{\sigma}_k \). This angle \( \beta_k \) is defined as

\[
\cos 2\beta_k = -\gamma_k^{(1)} \cdot \gamma_k^{(2)}.
\]

(A11)
We now choose axes such that the final state wave-function of $\hat{\sigma}_k$ is given in terms of the initial state wave-function by
\[
| \hat{\sigma}_k^f \rangle = \hat{U}_k | \hat{\sigma}_k^i \rangle = e^{-i\beta_k \hat{\sigma}_k^i} | \hat{\sigma}_k^i \rangle. \tag{A12}
\]
\[
| \{ \hat{\sigma}_k^f \} \rangle = \prod_{k=1}^N \hat{U}_k | \{ \hat{\sigma}_k^i \} \rangle = \hat{U} | \{ \hat{\sigma}_k^i \} \rangle. \tag{A13}
\]

In general the initial state of the spin bath will belong to the polarisation group $\Delta N$ (not necessarily $\Delta N = 0$), where the polarisation is defined along the $\hat{S}_1$-direction. As explained in the text, when $\hat{S}$ rotates from $\hat{S}_1$ to $\hat{S}_2 \approx -\hat{S}_1$, the energy conservation requires to change the polarisation from $\Delta N$ to $-\Delta N$. In future discussion of degeneracy blocking effects we will have to consider more general tunneling processes, when the nuclear polarisation is changed from $\Delta N$ to $\Delta N - 2M$ and back. In what follows we calculate the correlation function $P_{\Delta N, M}(t)$, which gives the dynamics of $\hat{S}$ when the spin bath transitions are restricted to be between the $\langle \hat{P} \rangle = \Delta N$ and $\langle \hat{P} \rangle = \Delta N - 2M$ subspaces, which are supposed to be in resonance (for pure orthogonality blocking $M = \Delta N$). The statistical weight of states with $\Delta N \gg N^{1/2}$ is negligible, so we will assume in our calculation that $\Delta N, M \ll N$.

The above restriction is enforced by the projection operator
\[
\hat{P}_{\Delta N} = \delta \left( \sum_{k=1}^N \hat{\sigma}_k^i - \Delta N \right) = \int_0^{2\pi} \frac{d\xi}{2\pi} e^{\xi \left( \sum_{k=1}^N \hat{\sigma}_k^i - \Delta N \right)}.
\]

We can now, including this restriction, write down an expression for the amplitude for $\hat{S}$ to go from $| \hat{S}_1(t = 0) \rangle$ to $| \hat{S}_1(t) \rangle$ starting from some $\langle \hat{P} \rangle = \Delta N$ environment state;
\[
A_{\Delta N, M}(t) = \left\{ \sum_{n=0}^\infty \frac{(i\Delta_t)^{2n}}{(2n)!} \prod_{i=1}^{2n} \left[ \int_0^{2\pi} \frac{d\xi_i}{2\pi} e^{-i\Delta N(\xi_{2n} + \xi_{2n-1} + \ldots + \xi_1)} e^{2iM(\xi_{2n-1} + \xi_{2n-3} + \ldots + \xi_1)} \right] | \{ \hat{\sigma}_k^i \} \right\},
\]
where $\hat{T}_{2n}$ is
\[
\hat{T}_{2n} = \left[ e^{i\xi_{2n} \sum_{k=1}^N \hat{\sigma}_k^i \hat{U}^\dagger e^{i\xi_{2n-1} \sum_{k=1}^N \hat{\sigma}_k^i \hat{U}^\dagger \ldots \hat{U}^\dagger e^{i\xi_1 \sum_{k=1}^N \hat{\sigma}_k^i \hat{U}}} \right].
\]

From (A15) we can now write the correlation function $P_{\Delta N, M}(t)$ as
\[
P_{\Delta N, M}(t) = \langle A_{\Delta N, M}(t)^\dagger A_{\Delta N, M}(t) \rangle
\]
\[
= \sum_{n=0}^\infty \sum_{m=0}^\infty \frac{(i\Delta_t)^{(2n+m)}}{(2n)!(2m)!} \prod_{i=1}^{2n} \prod_{i=1}^{2m} \left[ \int_0^{2\pi} \frac{d\xi_i}{2\pi} \int_0^{2\pi} \frac{d\xi_j}{2\pi} e^{-i\Delta N(\sum_{i=1}^{2n} \xi_i - \sum_{j=1}^{2m} \xi_j)} e^{2iM(\sum_{i=\text{odd}}^{2n-1} \xi_i - \sum_{j=\text{odd}}^{2m-1} \xi_j)} \langle \hat{T}_{2m}^\dagger \hat{T}_{2n} \rangle \right].
\]

The restrictions required to arrive at this formula are discussed in the text.

We now return to the assumption that the $\beta_k$ are small; specifically we will assume that the orthogonality exponent $\kappa$, defined by
\[
e^{-\kappa} = \prod_{k=1}^N \cos \beta_k,
\]
can be approximated by the perturbative expansion
\[
\kappa \approx \frac{1}{2} \sum_{k=1}^N \beta_k^2.
\]

This assumption makes it much easier to calculate the average in (A17). Consider first the problem with only one environmental spin $\hat{\sigma}_k$, and calculate the average $\langle \hat{T}_{2m}^\dagger \hat{T}_{2n} \rangle_k$ in this case; since $\hat{T}_{2m}^\dagger \hat{T}_{2n}$ is a product of operators acting separately on each $\hat{\sigma}_k$, the average over all spins is also the product of single spin results.
Because of (A19) we need only to consider processes with 0, 1, or 2 flips of the environmental spin, i.e., we will make an expansion in powers of $\beta_k$ for $\vec{\sigma}_k$, and stop at $\beta_k^2$. Then it is clear that, if the initial state of $\vec{\sigma}_k$ is $|\uparrow_k\rangle$

$$\hat{T}^{(k)}_{2n} |\uparrow_k\rangle = e^{i\xi_{2n} \vec{\sigma}_k^+ e^{-i\beta_k \vec{\sigma}_k^+} \cdots e^{-i\beta_k \vec{\sigma}_k^+} e^{i\xi_1 \vec{\sigma}_k^+} e^{-i\beta_1 \vec{\sigma}_k^+} |\uparrow_k\rangle}$$

$$= e^{i \sum_{i=1}^{2n} \xi_i} \left[ (1 - n\beta_k^2) |\uparrow_k\rangle + i\beta_k |\downarrow_k\rangle \right] \sum_{l=1}^{2n} (-1)^l e^{-2i \sum_{i=1}^{2n} \xi_i}$$

$$- \beta_k^2 |\uparrow_k\rangle \sum_{l=1}^{2n-1} \sum_{i=1}^{2n} (-1)^l e^{-2i \sum_{i=1}^{2n-1} \xi_i} + O(\beta_k^2) \right],$$

(A20)

where the first term arises from the sequence $[\uparrow\uparrow\ldots\uparrow]$, the second from the sequence $[\uparrow\uparrow\ldots\downarrow\downarrow\ldots\downarrow]$, with a flip when $j = l$; and so on. In the same way we find

$$\langle \downarrow_k | \hat{T}^{(k)}_{2m} \hat{T}^{(k)}_{2n} |\uparrow_k\rangle = e^{i \sum_{i=1}^{2m} \xi_i - \sum_{j=1}^{2m} \xi'_j} \left[ 1 - \beta_k^2 ((n + m) + \sum_{l=1}^{2m-1} \sum_{i=1}^{2m} (-1)^l e^{-2i \sum_{i=1}^{2m-1} \xi_i}$$

$$+ \sum_{p=1}^{2m} \sum_{p=1}^{2m-1} (-1)^{l+p} e^{-2i \sum_{j=1}^{p-1} \xi'_j}$$

$$- \sum_{p=1}^{2m} \sum_{p=1}^{2m} (-1)^{l+p} e^{-2i \sum_{i=1}^{2m} \xi_i - \sum_{j=1}^{p-1} \xi'_j} \right],$$

(A21)

to order $\beta_k^2$. The sequence $\langle \downarrow_k | \hat{T}^{(k)}_{2m} \hat{T}^{(k)}_{2n} |\downarrow_k\rangle$ will have a similar expression, but with reversed signs coming from the $e^{i\xi_i \vec{\sigma}_k^+}$ factors.

We now observe that the state with polarisation $\Delta N$ consists of $(N + \Delta N)/2$ spins up and $(N - \Delta N)/2$ spins down. Consequently, for each $\vec{\sigma}_k$, we add $\uparrow$ or $\downarrow$ averages like (A21), and then the product

$$\langle \hat{T}^{(k)}_{2m} \hat{T}^{(k)}_{2n} \rangle = \prod_{k=1}^{N_1} \langle \hat{T}^{(k)}_{2m} \hat{T}^{(k)}_{2n} \rangle \prod_{k'=1}^{N_1} \langle \hat{T}^{(k)}_{2m} \hat{T}^{(k)}_{2n} \rangle$$

(A22)

Substituting (A21) into this expression we get

$$\langle \hat{T}^{(k)}_{2m} \hat{T}^{(k)}_{2n} \rangle = e^{i \Delta N \sum_{i=1}^{2m} \xi_i - \sum_{j=1}^{2m} \xi'_j} \exp \left\{ - K_{nm}^{\text{eff}} (\xi, \xi', \Delta N) \right\},$$

(A23)

where the "effective action" $K_{nm}^{\text{eff}} (\xi, \xi', \Delta N)$ has two contributions $K^{\text{eff}} = K_1 + K_2$:

$$K_1 = 2\kappa (1 - N/\Delta N) \left\{ (n + m) + \sum_{l=1}^{2m-1} (-1)^l \cos[2 \sum_{i=1}^{l-1} \xi_i] + \sum_{p=1}^{2m} \sum_{p=1}^{2m} (-1)^{l+p} \cos[2 \sum_{j=1}^{p-1} \xi'_j]$$

$$- \sum_{p=1}^{2m} \sum_{p=1}^{2m} (-1)^{l+p} \cos[2 \sum_{i=1}^{\lambda l} \xi_i - \sum_{j=1}^{p-1} \xi'_j] \right\},$$

(A24)

$$K_2 = 2\kappa \frac{\Delta N}{N} \left\{ \sum_{l=1}^{2m} \sum_{l=1}^{2m} (-1)^l \sin[2 \sum_{i=1}^{l-1} \xi_i] + \sum_{p=1}^{2m} \sum_{p=1}^{2m} (-1)^{l+p} \sin[2 \sum_{j=1}^{p-1} \xi'_j]$$

$$- \sum_{p=1}^{2m} \sum_{p=1}^{2m} (-1)^{l+p} \sin[2 \sum_{i=1}^{\lambda l} \xi_i + \sum_{j=1}^{p-1} \xi'_j] \right\},$$

(A25)

We recall now that $\Delta N \leq N^{1/2} \ll N$, which allows to neglect the contribution due to $K_2$ and drop the correction $\Delta N/N$ to the coefficient $\kappa$ in $K_1$. Notice also that the phase factor in front of $\exp[-K^{\text{eff}}]$ in (A22) cancels exactly the phase proportional to $\Delta N$ in the formular (A17) for $P_{\Delta N,M}(t)$. Thus, quite surprisingly, we find the correlation function to be independent of $\Delta N$ in this limit:
\[
P_M(t) = \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} \frac{(i\Delta_n t)^{2(n+m)}}{(2n)!(2m)!} \prod_{i=1}^{2n} \prod_{j=1}^{2m} \int \frac{d\xi_i}{2\pi} \int \frac{d\xi'_j}{2\pi} \exp \left\{ 2iM(\xi_{2n-1} + \xi_{2n-3} + \ldots + \xi_1) - K_{\text{eff}}^{\xi_1}(\xi_1, \xi'_1) \right\}, \quad (A26)
\]

We can render this expression more useful by changing variables; first we consider the whole sequence \(\xi_\alpha = (\xi_1, \ldots, \xi_{2n}, -\xi_1, \ldots, -\xi_{2m})\) together, and then define new angular variables

\[
\chi_\alpha = 2(n+m) \sum_{\alpha' = \alpha}^2 \xi_\alpha + \pi \alpha,
\]

so that now

\[
P_M(t) = \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} \frac{(i\Delta_n t)^{2(n+m)}}{(2n)!(2m)!} \left( \prod_{\alpha=1}^{2(n+m)} \int \frac{d\chi_\alpha}{2\pi} \right) \right. \times \exp \left\{ iM \sum_{\alpha} (-1)^{\alpha+1} \chi_\alpha - 2\kappa [(n+m) + \sum_{\alpha' > \alpha} \cos(\chi_{\alpha'} - \chi_\alpha)] \right\}.
\]

Thus we have mapped our problem onto the partition function of a rather peculiar system of spins, interacting via infinite range forces, with interaction strength \(2\kappa\).

To deal with this partition function, we define “pseudo-spins” \(\vec{s}_\alpha\) and \(\vec{S}\), via

\[
\vec{s}_\alpha = (\cos \chi_\alpha, \sin \chi_\alpha)
\]

\[
\vec{S} = \sum_{\alpha=1}^{2(n+m)} \vec{s}_\alpha,
\]

so that

\[
\vec{s}_\alpha \cdot \vec{s}_{\alpha'} = \cos(\chi_\alpha - \chi_{\alpha'})
\]

\[
\sum_{\alpha',\alpha} \cos(\chi_\alpha - \chi_{\alpha'}) = S^2,
\]

We can think of \(\vec{s}_\alpha\) as rotating in our fictitious angular space defined by the projection operator \(\Pi_{\text{A14}}\). Now consider the term \(G(\vec{S})\) in \(\text{[A28]}\), defined by

\[
G(\vec{S}) = \left( \prod_{\alpha=1}^{2(n+m)} \int \frac{d\chi_\alpha}{2\pi} e^{iM(-1)^{\alpha+1} \chi_\alpha} \right) \exp \left\{ -\kappa \sum_{\alpha' = \alpha}^\chi \cos(\chi_\alpha - \chi_{\alpha'}) \right\}
\]

\[
= \left( \prod_{\alpha=1}^{2(n+m)} \int \frac{d\chi_\alpha}{2\pi} e^{iM(-1)^{\alpha+1} \chi_\alpha} \right) e^{-\kappa S^2}.
\]

This is easily calculated, viz.,

\[
G(\vec{S}) = \int d\vec{S} e^{-\kappa S^2} \prod_{\alpha=1}^{2(n+m)} \int \frac{d\chi_\alpha}{2\pi} e^{iM(-1)^{\alpha+1} \chi_\alpha} \delta(\vec{S} - \sum_\alpha \vec{s}_\alpha)
\]

\[
= \frac{1}{2\kappa} \int dz z e^{-z^2/4\kappa} f^{2(n+m)}_M(z),
\]

where \(J_m(\lambda)\) is the zeroth-order Bessel function. Using

\[
\sum_{l=0}^{2(n+m)} \frac{(2(n+m))!}{(2m)!(2n)!} = \delta_{n+m,0} + 2^{2(n+m)}/2,(A33)
\]
to reorganize the sum over $n$ and $m$ in (A28) and changing the integration variable $z \to 2x\sqrt{\kappa}$, we then find

$$P_{\|\|}(t) = 2 \int_0^\infty dx \, e^{-x^2} \frac{1}{2} \left( 1 + \sum_{s=0}^\infty \frac{[2it\bar{\Delta}_e J_M(2\sqrt{\kappa}x)]^{2s}}{(2s)!} \right)$$

$$\equiv 2 \int_0^\infty dx \, e^{-x^2} P_{\|\|}^{(0)}(t, \Delta_M(x)) .$$

(A34)

$$\Delta_M(x) = \bar{\Delta}_e J_M(2\sqrt{\kappa}x) .$$

(A35)

Here we come to the crucial point in our derivation. Eq.(b.21b) gives us the final answer as a superposition of non-interacting correlation functions for effective tunneling rates $\Delta_M(x)$ with the proper weighting

$$P_{\|\|}(t) = \int_0^\infty dx \, e^{-x^2} (1 + \cos[2\bar{\Delta}_e J_M(2\sqrt{\kappa}x)t]) ,$$

(A36)

For $M = 0$ this is the form quoted in Eq.(I.124) of the text.

It is worth noting that non-zero $M$ enters this calculation as the overall phase factor starting from (A17) and crown it in (A32) while finally integrating over $\{\chi_{\alpha}\}$ to produce the Bessel function of order $M$. This observation allows to generalise any calculation done for $M = 0$ to finite $M$ by simply replacing $J_0 \to J_M$ in the final answer - the prescription which we make use in other Appendices.

**APPENDIX B: DERIVATIONS FOR THE GENERIC CASE**

We outline here the derivations for section [V] incorporating all three mechanisms.

1. Orthogonality blocked topological decoherence

As discussed in the text, a formal expression for this case can be easily written down, by including the induced topological phase $\pm[\Phi + \sum_{k=1}^N (\alpha_k \bar{n}_k - i\xi_k \bar{v}_k) \cdot \hat{\sigma}_k]$ which accumulates during each instanton, and then averaging over the trajectories of the $\{\hat{\sigma}_k\}$. Here we evaluate the $M = 0$ contribution; The result for $P_M(t)$ then follows from obvious generalisation. We have

$$P_0(t) = \sum_{n=0}^\infty \sum_{m=0}^\infty \frac{(i\Delta_{\|})^{2(n+m)}}{(2n)!(2m)!} \sum_{\{g_i=\pm\}} e^{i\Phi \sum_{i=1}^n g_i} \prod_{i=1}^n \prod_{j=1}^m \int \frac{dk_i}{2\pi} \int \frac{d\xi_i}{2\pi} \langle \hat{T}_{2n}(g_i) \hat{T}_{2n}(\xi_i) \rangle ,$$

(B1)

where the transition matrices now include these phases, i.e.,

$$\hat{T}_{2n}(g_i) = e^{i\xi_2 P} \hat{U}_{\text{top}} e^{i\xi_{2n-1} P} \ldots e^{i\xi_1 P} \hat{U}_{\text{top}} .$$

(B2)

and $\hat{U}_{\text{top}}$ now generalizes the $\hat{U}$ involved in orthogonality blocking, viz.,

$$\hat{U}_{\text{top}} = \prod_{k=1}^N \exp \left\{ ig_i [(\alpha_k \bar{n}_k - i\xi_k \bar{v}_k) \cdot \hat{\sigma}_k - i\beta_k \hat{\sigma}_k^z] \right\} .$$

(B3)

To evaluate (B1), we rewrite it as

$$P_0(t) = \sum_{n=0}^\infty \sum_{m=0}^\infty \frac{(i\Delta_{\|})^{2(n+m)}}{(2n)!(2m)!} \sum_{\{g_i=\pm\}} e^{i\Phi \sum_{i=1}^n g_i} \prod_{\rho=1}^{2(n+m)} \int \frac{dk_\rho}{2\pi} \exp \left\{ -K_{\text{eff}}^{(n,m)}(\{g_i\}, \{\xi_\rho\}) \right\} ,$$

(B4)

and calculate the $K_{\text{eff}}^{(n,m)}(\{g_i\}, \{\xi_\rho\})$ up to order in $\alpha_k^2, \beta_k^2, \xi_k^2$ (see Appendix A.2).

Consider first the case where $\beta_k$ and $\xi_k$ are zero. Then, with the usual assumption that the individual $\alpha_k$ are small, but not necessarily $\lambda = 1/2 \sum_k \alpha_k^2$, the “effective action” $K_{\text{eff}}^{(n,m)}$ simplifies to (compare Eq.(A28))
\[
K_{mm}^{eff}(\{\xi_\rho\}) = \lambda' \sum_{\rho' \neq \rho} g_\rho g_{\rho'} + (\lambda - \lambda') \sum_{\rho' \neq \rho} \cos(\chi_\rho - \chi_{\rho'}) g_\rho g_{\rho'},
\]

which generalizes from orthogonality blocking; the \(\chi_\rho\) are defined as in (A27), and

\[
\lambda = \frac{1}{2} \sum_{k=1}^{N} \alpha_k^2
\]

\[
\lambda' = \frac{1}{2} \sum_{k=1}^{N} \alpha_k^2 (n_k)^2.
\]

The term in \(\lambda'\) comes from the expansion to \(O(\alpha_k^2)\) (analogous to the expansion in \(\beta_k^2\) in Appendix B), which produces the average

\[
\langle \sigma_k | \hat{\sigma}_k \cdot \hat{n}_k e^{-i \sum_{j=x,y} \xi_j (\hat{\sigma}_j \cdot \sigma_j)} | \sigma_k \rangle = [(n_k^z)^2 + (1 - (n_k^z)^2)e^{-2i\sigma_k \sum_{j=x,y} \xi_j}],
\]

(compare (A29); again, we assume equal statistical weightings for \(\uparrow\) and \(\downarrow\) spins).

We now use the same trick of (A29) and (A30), to write

\[
\hat{S}^2 = \left( \sum_{\rho = 1}^{2(n+m)} g_\rho \hat{s}_\rho \right)^2 = \sum_{\rho' \neq \rho} g_\rho g_{\rho'} \cos(\chi_\rho - \chi_{\rho'}),
\]

with the \(\hat{s}_\rho = (\cos(\chi_\rho, \sin(\chi_\rho))\) as before; making the change of variables \(\chi_\rho = \chi_\rho + \pi g_\rho/2\), we get the generalization of (A34), viz.,

\[
P_0(t) = 2 \int dx \int dx e^{-x^2} \left[ \frac{1}{2} \left[ 1 + \sum_{s=0}^{\infty} \frac{(2i\Delta_o t)^{2s}}{(2s)!} J_0^2(2x\sqrt{\lambda - \lambda'}) \right] \sum_{\{g_i = \pm\}} e^{i\Phi} \sum_{i=1}^{2(n+m)} g_i - \lambda' \left( \sum_{i=1}^{2(n+m)} g_i \right)^2 \right] 
\]

\[
= 2 \int dx \int dx e^{-x^2} \left[ \frac{1}{2} \left[ 1 + \sum_{s=0}^{\infty} \frac{(2i\Delta_o t)^{2s}}{(2s)!} J_0^2(2x\sqrt{\lambda - \lambda'}) t^{2s} \right] \right] 
\]

Writing the sum over \(\{g_i\}\) in the form

\[
\sum_{\{g_i = \pm\}} e^{i\Phi} \sum_{i=1}^{2(n+m)} g_i - \lambda' \left( \sum_{i=1}^{2(n+m)} g_i \right)^2 = \frac{2s}{(2s - m)!m!} e^{i\Phi(2s - 2m) - \lambda' (2s - 2m)^2}
\]

we now use the identity in (E8) to eliminate this term in favour of a phase integration over \(\phi\); defining \(F_{\lambda'}(\nu) = e^{-4\lambda' \nu^2}\) as in the text, we get \(P_{\uparrow\uparrow}(t)\) in the form

\[
P_0(t) = 2 \int dx \int dx e^{-x^2} \int \frac{d\phi}{2\pi} \sum_{m=-\infty}^{\infty} F_{\lambda'}(m) e^{i2m(\Phi - \varphi)} P_{\uparrow\uparrow}^{(0)}(t, \tilde{\Delta}_o(\varphi, x))
\]

where

\[
\tilde{\Delta}_o(\varphi, x) = 2\tilde{\Delta}_o \cos \varphi J_0(2x\sqrt{\lambda - \lambda'})
\]

If we do the integration over \(\varphi\), this gives

\[
P_0(t) = \int dx \int dx e^{-x^2} \left[ 1 + \sum_{m=-\infty}^{\infty} F_{\lambda'}(m) e^{i2m\Phi} J_{2m}[4\tilde{\Delta}_o t J_0(2x\sqrt{\lambda - \lambda'})] \right]
\]
which is Eq.(5.3) in the text.

The above calculation is generalized for non-zero $M$ by noting that the only modification will be in a phase factor $iM\sum_\rho(-1)^{\rho+1}\chi_\rho$ throughout the whole derivation, which will finally result in the replacement $J_0(2x\sqrt{\lambda - \lambda'}) \rightarrow J_0(2x\sqrt{\lambda - \lambda'})$ in Eq.(B12) for the effective tunneling rate.

2. Orthogonality blocking plus degeneracy blocking

We wish to evaluate the "biased orthogonality blocking" problem described in section IV.B. We generalize the expression (A8) for a biased 2-level system (topological phase is not yet included here) to include orthogonality blocking via our projection operators (cf. Eq.(A15)); this gives

$$A_{M=0}(p, \epsilon) = \frac{1}{p - i\epsilon} \sum_{n=0}^{\infty} \left( \frac{-i\Delta_{\Phi}}{p^2 + \epsilon^2} \right)^n \int_1^{2n} \frac{d\xi_i}{2\pi} \hat{T}_{2n} \right| \{\sigma_i^{in}\} , \tag{B14}$$

with $A_{M=0}(t, \epsilon)$ given by the Laplace transform (A7) of (B14). It then follows that

$$P_{T\bar{T}}(t, \epsilon) = \int_{-\infty}^{\infty} dp_1 dp_2 e^{(p_1 + p_2)t} \frac{1}{p_1 - i\epsilon} \frac{1}{p_2 - i\epsilon} \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} B_{nm}(p_1, p_2, \epsilon) \tag{B15}$$

$$B_{nm}(p_1, p_2, \epsilon) = \left( \frac{\Delta_{\Phi}}{p_1^2 + \epsilon^2} \right)^n \left( \frac{\Delta_{\Phi}}{p_2^2 + \epsilon^2} \right)^m \int_{i=1}^{n} \int_{j=1}^{m} \int \frac{d\xi_i}{2\pi} \int \frac{d\xi_j}{2\pi} \langle \hat{T}_{2n} \hat{T}_{2n} \rangle , \tag{B16}$$

Surprisingly, we find that the problem of calculating the correlation function in a bias (after writing it as a series expansion in the tunneling rate) can be again solved in terms of non-interacting $P_{T\bar{T}}^{(0)}(t, \epsilon)$, because the averages in (B10) are identical to those evaluated in Appendix A, Eq.(A32), to give

$$\prod_{i=1}^{2n} \int \frac{d\xi_i}{2\pi} \prod_{j=1}^{2m} \int \frac{d\xi_j}{2\pi} \langle \hat{T}_{2n} \hat{T}_{2n} \rangle = 2 \int dx x e^{-x^2} J_0^{2(n+m)}(2x\sqrt{\kappa}) . \tag{B17}$$

This can be absorbed into defining an effective $\Delta_{\phi}(x) = 2\Delta_{\Phi}J_0(2x\sqrt{\kappa})$, just as for pure orthogonality blocking, and the series become identical to Eq.(A10) giving the result (5.10) in the text. The generalisation to $M \neq 0$ just consists in the replacement $\Delta_{\phi}(x) \rightarrow \Delta_{M}(x)$.

3. The generic case

Here we calculate $P_{T\bar{T}}(t)$ with all terms $(\alpha_k, \omega^\parallel_k, \Phi, \text{ and } \xi_k)$ except $\omega^\perp_k$ included from the effective Hamiltonian. Thus we return to the expression (B3) and (B4) and calculate the "effective action" $K_{eff}^{nm}$ with nonzero $\xi_k$. Now, evaluating the contribution from individual spins from the expansion to $O(\alpha^2_k, \xi^2_k)$, we find for the average (cf. (B7))

$$\langle \sigma_k | (i\alpha_k \bar{n}_k + (-1)^{\rho} \xi_k \bar{v}_k) \cdot \hat{\sigma}_k \rangle = e^{-i \sum_{j=\rho}^{\rho'} \xi_j (\sigma_j - \sigma_k)} \times (i\alpha_k \bar{n}_k + (-1)^{\rho'} \xi_k \bar{v}_k) \cdot \hat{\sigma}_k | \sigma_k \rangle , \tag{B18}$$

the following expression

$$\langle i\alpha_k n_k + (-1)^{\rho} \xi_k v_k \rangle (i\alpha_k n_k + (-1)^{\rho'} \xi_k v_k)$$

$$+ e^{-2i \sum_{j=\rho}^{\rho'} \xi_j} \times [i\alpha_k (n^y_k + n^y_k) + (-1)^{\rho} \xi_k (v^y_k + iv^x_k)] [i\alpha_k (n^y_k - n^y_k) + (-1)^{\rho'} \xi_k (v^y_k - iv^x_k)] \tag{B19}$$

Thus, in full analogy to the derivation done in Appendix B (Eq.(A21)), we find the contribution of an individual spin to the action to be.

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where the constants are defined by:

\[
\frac{1}{2} \sum_{\rho, \rho'} g_\rho g_{\rho'} \left\{ \left[ \alpha_k^2 (n_k^2)^2 - (-1)^{\rho + \rho'} \xi_k^2 (v_k^2) - i \alpha_k \xi_k n_k v_k ((-1)^\rho + (-1)^{\rho'}) \right] \\
+ \cos(\chi_\rho - \chi_{\rho'}) (\alpha_k^2 (1 - (n_k^2)^2) - (-1)^{\rho + \rho'} \xi_k^2 (1 - (v_k^2)^2) \\
- i \alpha_k \xi_k (\bar{u}_k \cdot \bar{v}_k - n_k v_k ((-1)^\rho + (-1)^{\rho'})) \right\},
\]

Summing up the contributions from \( N \) spins we generate the "effective action", which now takes the form

\[
K_{n m}^{\text{eff}} = \sum_{\rho, \rho'} g_\rho g_{\rho'} \left\{ \left[ \lambda - \eta' (-1)^{\rho + \rho'} - i \gamma' ((-1)^\rho + (-1)^{\rho'}) \right] \\
+ \cos(\chi_\rho - \chi_{\rho'}) [\lambda - \lambda'] \right. \\
- \left. (-1)^{\rho + \rho'} (\eta - \eta') - i (\gamma - \gamma') ((-1)^\rho + (-1)^{\rho'}) \right\},
\]

where the constants are defined by:

\[
\lambda = \frac{1}{2} \sum_{k=1}^{N} \alpha_k^2 ; \quad \lambda' = \frac{1}{2} \sum_{k=1}^{N} \alpha_k^2 (n_k^2)^2 ;
\]

\[
\eta = \frac{1}{2} \sum_{k=1}^{N} \xi_k^2 ; \quad \eta' = \frac{1}{2} \sum_{k=1}^{N} \xi_k^2 (v_k^2)^2 ;
\]

\[
\gamma = \frac{1}{2} \sum_{k=1}^{N} \alpha_k \xi_k \bar{u}_k \cdot \bar{v}_k ; \quad \gamma' = \frac{1}{2} \sum_{k=1}^{N} \alpha_k \xi_k n_k v_k ;
\]

As before we change variables according to \( \chi_\rho = \chi_{\rho'} + \pi \) when \( g_\rho = -1 \), to introduce the odd and even spin fields

\[
\tilde{S}_o = \sum_{\rho = \text{odd}}^{2(n+m)-1} \tilde{s}(\chi_\rho) ; \quad \tilde{S}_e = \sum_{\rho = \text{even}}^{2(n+m)} \tilde{s}(\chi_\rho).
\]

Now we notice that the effective action is a quadratic form in \((\tilde{S}_o, \tilde{S}_e)\):

\[
K_{n m}^{\text{eff}} = \lambda' \left( \sum_{\rho} g_\rho \right)^2 - \eta' \left( \sum_{\rho} (-1)^\rho g_\rho \right)^2 - 2i \gamma' \left( \sum_{\rho} g_\rho \left( \sum_{\rho} (-1)^\rho g_\rho \right) \right)
+ (\lambda - \lambda') (\tilde{S}_e + \tilde{S}_o) - (\eta - \eta') (\tilde{S}_e - \tilde{S}_o)^2 - 2i (\gamma - \gamma') (S_e^2 - S_o^2),
\]

The rest is simple now. First we employ spectral representations for the \( \delta \)-functions \( \delta(\tilde{S}_o - \sum_{\rho = \text{odd}}^{2(n+m)-1} \tilde{s}_\rho) \) and \( \delta(\tilde{S}_e - \sum_{\rho = \text{even}}^{2(n+m)} \tilde{s}_\rho) \) to integrate over the angle variables \( \chi_\rho \) (compare Eq. \((\text{A32})\)) to give \( (s = n + m)\)

\[
\prod_{\rho = 1}^{2s} \frac{d \xi_\rho}{\sqrt{2\pi}} e^{-\lambda' \sum_{\rho} g_\rho \left( \sum_{\rho} (-1)^\rho g_\rho \right)} \\
\times \exp \left\{ -a (\tilde{S}_e + \tilde{S}_o)^2 + b (\tilde{S}_e - \tilde{S}_o)^2 + 2ic (S_e^2 - S_o^2) + i\vec{x}_1 \cdot \tilde{S}_e + i\vec{x}_2 \cdot \tilde{S}_o \right\}.
\]

with obvious definition

\[
a = \lambda - \lambda' ; \quad b = \eta - \eta' ; \quad c = \gamma - \gamma'.
\]

The Gaussian integration over \( d\tilde{S}_o, d\tilde{S}_e \), and then the integration over the angle between the \( \vec{x}_1 \) and \( \vec{x}_2 \) gives for the expression \((\text{B27})\) the formula

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\[ e^{-\lambda'(\sum_{\rho} g_{\rho})^2 + \eta' (\sum_{\rho} (-1)^{\rho} g_{\rho})^2 - 2 \gamma' (\sum_{\rho} g_{\rho}) (\sum_{\rho} (-1)^{\rho} g_{\rho})} \int dx_1 \int dx_2 x_1 x_2 J_0^s(x_1) J_0^s(x_2) \times \frac{1}{8(ab - c^2)} \int_0^{(a + b)x_1 x_2} \frac{(a - b + 2ic)x_1^2 + (a - b - 2ic)x_2^2}{16(ab - c^2)} \ exp \left\{ \frac{(a - b + 2ic)x_1^2 + (a - b - 2ic)x_2^2}{16(ab - c^2)} \right\} . \] (B29)

Finally we note that the sum over \( \{ g_{\rho} \} \) can be written as

\[
\sum_{\{ g_{\rho} = \pm \}} = \sum_{m_1, m_2 = -\infty}^{\infty} \sum_{\rho} \delta(2m_1 - \sum_{\rho} g_{\rho}) \delta(2m_2 - \sum_{\rho} (-1)^{\rho} g_{\rho})
\]

\[
= \int \int \frac{d\varphi_1 d\varphi_2}{(2\pi)^2} \sum_{m_1, m_2 = -\infty}^{\infty} e^{i2m_1 \varphi_1 + i2m_2 \varphi_2} \sum_{\{ g_{\rho} = \pm \}} e^{-i \varphi_1} \sum_{\rho} g_{\rho} - i \varphi_2 \sum_{\rho} (-1)^{\rho} g_{\rho}
\]

\[
= \int \int \frac{d\varphi_1 d\varphi_2}{(2\pi)^2} \sum_{m_1, m_2 = -\infty}^{\infty} e^{i2m_1 \varphi_1 + i2m_2 \varphi_2} \left[ 2 \cos(\varphi_1 + \varphi_2) \cos(\varphi_1 - \varphi_2) \right] . \] (B30)

Combining now (B29), (B30), and (B4), together we observe that the sum over \( n \) and \( m \) is nothing but the series for the coherent dynamics expansion in powers of the renormalized tunneling splitting

\[
\tilde{\Delta}_n^2(x_1, x_2, \varphi_1, \varphi_2) = 4 \Delta_n^2 \cos(\varphi_1 + \varphi_2) \cos(\varphi_1 - \varphi_2) J_0(x_1) J_0(x_2) , \] (B31)

which is then integrated over the variables \( x_1, x_2, \varphi_1, \varphi_2 \) and summed over \( m_1, m_2 \) with the weight given by

\[
Z = e^{2|m_1(\Phi - \varphi_1) - m_2 \varphi_2| + 4m_1 m_2 \gamma} e^{4(\gamma' m_2^2 - \lambda' m_1^2)} \times \frac{x_1 x_2}{8(ab - c^2)} \int_0^{(a + b)x_1 x_2} \frac{(a - b + 2ic)x_1^2 + (a - b - 2ic)x_2^2}{16(ab - c^2)} \ exp \left\{ \frac{(a - b + 2ic)x_1^2 + (a - b - 2ic)x_2^2}{16(ab - c^2)} \right\} . \] (B32)

One then finds

\[
P_{\varphi_\varphi}(t) = \int \frac{d\varphi_1}{2\pi} \int \frac{d\varphi_2}{2\pi} \sum_{m_1 = -\infty}^{\infty} \sum_{m_2 = -\infty}^{\infty} \int dx_1 \int dx_2 \times Z(\varphi_1, \varphi_2, x_1, x_2, m_1, m_2) P_{\varphi\varphi}^{(0)}(t, \tilde{\Delta}_0(x_1, x_2, \varphi_1, \varphi_2)) , \] (B33)

and (B33) has an obvious generalisation to include the bias integration \( \int de \).

**FIGURE CAPTIONS**

**Figure 1** Schematic depiction of the Central Spin Model

**Figure 2** The range of coupling energies between a typical central spin and its spin environment. The hyperfine contact interaction is shown for a variety of nuclear spins in various hosts.

**Figure 3** A typical trajectory for \( \tilde{S}(\tau) \), contributing to the path integral.

**Figure 4** The function \( \alpha_{\varphi}(\omega) \) for the instanton of the toy model Hamiltonian. This form is typical of virtually any instanton describing the tunneling of our central spin.

**Figure 5** Behaviour of the central spin in the topological decoherence limit. In (a) is shown \( P_{\varphi \varphi}(t) \) for the intermediate coupling case; and (b) shows the spectral function \( \chi''(\omega) \) derived from this. These forms are the universal forms arising for topological decoherence (see text).

**Figure 6** \( P_{\varphi \varphi}(t) \) for an \( S = 1/2 \)-integer spin coupled to 40 environmental spins with random \( \alpha_k \) between 0 and 0.05.

**Figure 7** \( P_{\varphi \varphi}(t) \) for an integer \( S \), and coupling to 20 environmental spins with random \( \alpha_k \) between 0 and 1.
Figure 8 Definition of the orthogonality angle $\beta_k$, in terms of the unit vectors $\mathbf{\gamma}_k^{(1)}$ and $\mathbf{\gamma}_k^{(2)}$. This also defines the unit vectors $\mathbf{l}_k$ and $\mathbf{m}_k$ appearing in the effective Hamiltonian (3.10).

Figure 9 The effect of pure orthogonality blocking when $\kappa$ is small ($\kappa \leq O(1)$).

Figure 10 The effect of pure orthogonality blocking when $\kappa \gg 1$. As well as being pushed to lower frequencies in accordance with Eq.(4.27), $\chi''(\omega)$ develops a complicated peak structure.

Figure 11 $P_{\theta\theta}(t)$, in the orthogonality blocking limit, for $\kappa = 1000$. Notice the small resurgence of oscillation for $\Delta_\theta t \sim 300$, coming from constructive interference between the environment-induced peaks in $\chi''(\omega)$.

Figure 12 The effect of a finite but small spread in the couplings $\{\omega_k\}$ on the energy levels of the system. In (a) the set of levels for the total "central spin + environment" is shown as we add environmental spins one by one. In (b) the resulting distribution $W(\epsilon)$ is shown. The parameter $\mu = N^{1/2}\delta\omega_k/\omega_o$. The separation between peaks for $\mu \ll 1$ is $\omega_o$. The tunneling splitting is far smaller, indeed invisible on this scale for typical values of $\omega_o$. Usually $\mu > 1$, giving the smooth Gaussian; if $\mu \ll 1$ a peak structure appears in $W(\epsilon)$.

Figure 13 The spectral weight $\chi''(\omega)$ for the case of pure degeneracy blocking.

Figure 14 A plot of the result for projected topological decoherence, in Eq.(5.8), for small values of $(\lambda - \lambda')$; notice that even if $(\lambda - \lambda')$ is only 0.1, the result is very noticeably different from pure topological decoherence (for which $\lambda - \lambda' = 0$), shown in Fig.5b.

Figure 15 Graphs of $\chi''(\omega)$ for projected topological decoherence, but now for $\lambda - \lambda' \gg 1$.

Figure 16 The spectral function $\chi''(\omega)$ for degeneracy blocked orthogonality blocking (eq.(5.16)). Here we see the results for small $\kappa$.

Figure 17 $\chi''(\omega)$ for degeneracy blocked orthogonality blocking, for $\kappa \gg 1$.

Figure 18 $\chi''(\omega)$ for the case where all 3 decoherence mechanisms enter, and we assume that $\lambda, \lambda' > 1$ (so there is topological decoherence). Here we show $\chi''(\omega)$ for small values of $\lambda - \lambda'$.

Figure 19 $\chi''(\omega)$ when all 3 mechanisms operate, and $\lambda, \lambda' > 1$; here we show results for large $(\lambda - \lambda')$.

Figure 20 $\chi''(\omega)$ for $TbFe\,3$ grains, calculated using the numbers in the text.
### Table I. Selected Hyperfine Couplings

| Nucleus | Host                | State | $\omega_k$ (MHz) | Abundance (%) |
|---------|---------------------|-------|------------------|---------------|
| H$^+$   | CuCl$_2 \cdot$ H$_2$O | (AFM) | 3.5              | 99.99         |
| F$^{19}$ | MnF$_2$             | (AFM) | 160              | 100           |
| Mn$^{55}$ | MnF$_2$             | (AFM) | 680              | 100           |
|        | MnB                 | (FM)  | 217.7            |               |
| Fe$^{57}$ | Fe                 | (FM)  | 46.65            | 2.19          |
|        | $\alpha - Fe_2O_3$  | (FM)  | 71.5             |               |
|        | Fe$_3$O$_4$         | (Ferrite) | 63.55         |               |
|        | $Y_3Fe_5O_{12}$     | (YIG-FM) | 64.9/76.05      |               |
| Co$^{59}$ | FeCo$_5$            | (FM)  | 289.2            | 100           |
|        | Co                  | (FM)  | 228              |               |
| Ni$^{61}$ | Ni                 | (FM)  | 28.35            | 1.19          |
| Tb$^{159}$ | Tb                | (FM)  | 3776, 3108, 2439 | 100           |
| Dy$^{161}$ | Dy                | (FM)  | 1603, 1219, 830, 445 | 18.88    |
| Dy$^{163}$ | "                 | "     | 1984, 1574, 1163 | 24.97        |

### Table II. Some Typical NMR Linewidths in Antiferromagnets Coming from the Suhl - Nakamura Interaction.

For Mn$^{55}$, a 1 Oe linewidth roughly corresponds to a $\delta \omega_k \sim 1 \text{ kHz}$.

| Nucleus | Host | Linewidth (Oe) |
|---------|------|----------------|
| F$^{19}$ | MnF$_2$ | 14             |
| Mn$^{55}$ | "     | 1000           |
| Co$^{59}$ | CoF$_2$ | 268 (1/2 $\leftrightarrow$ -1/2) |
|        | "     | 258 ($\pm$3/2 $\leftrightarrow$ $\pm$1/2) |
|        | "     | 222 (5/2 $\leftrightarrow$ 3/2) |
|        | "     | 165 (7/2 $\leftrightarrow$ 5/2) |
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Fig. 4
Fig. 6

$S = 1/2$ integer
Fig. 7
Fig. 10

$\kappa = 10^5$

$\kappa = 50$
\( \kappa = 1000 \)
Fig. 15

$\chi''$

$\lambda - \lambda' = 10^5$

$\lambda - \lambda' = 50$

$\omega / \Delta_0$
Fig. 16
Fig. 17

$\chi''$

$\kappa = 50$

$\kappa = 10^5$

$\frac{\omega}{\Delta_0}$
Fig. 18

$\chi''/\chi'$ for $\lambda - \lambda' = 0.1$ and $\lambda - \lambda' = 1$.
\[ \chi''/ \]

\[ \lambda - \lambda' = 50 \]

\[ \lambda - \lambda' = 10^5 \]

Fig. 19
Fig. 20

$\chi''$

$\lambda - \lambda' = 10$

$\omega / \Delta_o$