

Report Title

Spin Decoherence Measurements for Solid State Qubits

ABSTRACT

This project set out to measure and to understand the decoherence times of nuclear spins in semiconductors, to assess their potential as qubits in solid state quantum computer architectures. Our initial goal was to characterize P-31 nuclei in Silicon doped with Phosphorous (Si:P). While working towards this goal, we unexpectedly discovered an important discrepancy with the conventional theory of NMR. Specifically, well-known multiple π pulse spin echo experiments had the ability either to freeze out or to accelerate the signal decay expected due to the spin-spin dipolar coupling, reminiscent of the quantum Zeno effect. This result has now been seen in many nuclei (e.g., Si-29, C-13, Y-89, H-1) in different samples (including Silicon and buckyballs), and it is a robust phenomenon. This was probably always present, just not recognized, in magnetic resonance experiments. It appears to be a many-body effect arising from the tiny spin-spin interactions acting during strong, but finite, control pulses. Understanding this puzzle is essential, because it is likely to be relevant to most physical qubits (not just spins) driven by "bang-bang" control sequences, or other control pulse sequences.

List of papers submitted or published that acknowledge ARO support during this reporting period. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

"Anomalies in the NMR of silicon: Unexpected spin echoes in a dilute dipolar solid",
A. E. Dementyev, D. Li, K. MacLean, and S. E. Barrett
Phys. Rev. B 68, 153302 (2003).

Number of Papers published in peer-reviewed journals: 1.00

(b) Papers published in non-peer-reviewed journals or in conference proceedings (N/A for none)

Number of Papers published in non peer-reviewed journals: 0.00

(c) Presentations

12 APS March Meeting Talks (1 in 2002, 4 in 2003, 4 in 2004, and 3 in 2005.)
Three meeting or seminar talks.

Number of Presentations: 15.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

"Generating Spin Echoes in Dipolar Solids with π -pulses: More is Different",
D. Li, A.E. Dementyev, Y. Dong, R.G. Ramos, and S.E. Barrett
submitted to PRL.

(two more manuscripts are in preparation)

Number of Manuscripts: 1.00

Number of Inventions:

Graduate Students

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	
Anatoly Dementyev	0.33	No
Yanqun Dong	0.33	No
Rona Ramos	0.33	No
Dale Li	0.05	No
Jonathan Jerke	0.05	No
FTE Equivalent:	1.09	
Total Number:	5	

Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	
Anatoly Dementyev	0.50	No
FTE Equivalent:	0.50	
Total Number:	1	

Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
Sean Barrett	0.50	No
FTE Equivalent:	0.50	
Total Number:	1	

Names of Under Graduate students supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	
Kenneth MacLean	0.50	No
Ariel Cohen	0.05	No
John Murray	0.05	No
Maggie Wittlin	0.05	No
Nathaniel Kan	0.05	No
FTE Equivalent:	0.70	
Total Number:	5	

Names of Personnel receiving masters degrees

<u>NAME</u>	
Anatoly Dementyev	No
Dale Li	No
Rona Ramos	No
Yanqun Dong	No
Total Number:	4

Names of personnel receiving PHDs

<u>NAME</u>	
Anatoly Dementyev	No
Total Number:	1

Names of other research staff

NAME

PERCENT_SUPPORTED

FTE Equivalent:

Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Final Report: Spin Decoherence Measurements for Solid State Qubits

Proposal #: 42388PHQC, Agreement #: DAAD190110507

Report Covers Period: May 1, 2001-Jul 31, 2005.

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Foreword: Solid-state NMR measurements in Silicon were carried out in support of efforts to use ‘Spins in Semiconductors’ for quantum information processing (QIP). Measurements of the intrinsic decoherence time (T_2) for spins in Silicon revealed a puzzle: two standard pulse sequences used to measure T_2 (i.e., Hahn echoes and CPMG) yielded contradictory results. By investigating this effect in a series of Silicon samples, and then in other dipolar solids, we discovered an unexpected effect of finite-pulses: the very weak spin-spin coupling can have a coherent effect over many single spin rotations, which lead to dramatic failures of normal NMR intuition. These many-body effects will make the dynamics of even small numbers (e.g., >3) of interacting qubits much more challenging to control, which is a ‘looming problem for quantum information processing (QIP) as systems grow larger and more complicated’. At the same time, these many-body modifications of pulse action may be used to implement novel qubit-qubit interactions, not normally accessible to the experimentalist. These results are relevant to almost all proposed implementations of quantum computation using many qubits, since avoiding them would require that *all terms* in the spin Hamiltonian besides the pulse are set *exactly* to zero, which is basically impossible to do in practice. Moreover, since the ideal system for QIP is separated from environmental influences, tiny effects can add up coherently to have large consequences. Ignoring this effect will lead to big problems for QIP. On the other hand, understanding it may lead to new resources for QIP.

Final Report: Spin Decoherence Measurements for Solid State Qubits

Proposal #: 42388PHQC, Agreement #: DAAD190110507, Report Period: (5/1/01-7/31/05)

PI: S.E. Barrett, Yale Physics Dept.

1. Introduction

Quantum information processing (QIP) has the potential to revolutionize computation. Nearly all of the advantages of QIP require the use of large numbers of qubits. With some exceptions, most subfields are still working on isolated, single qubits...that is already quite demanding. However, the *Quantum Information Science and Technology Roadmap, Version 2* lays out an aggressive high-level goal that all subfields are trying to reach: a system of ~ 10 qubits by 2007, jumping up to ~ 50 qubits by 2012.¹ Pushing to such large numbers of qubits is an essential step in achieving the full promise of QIP. At the same time, such systems are likely to exhibit new complexity, not easily anticipated at the level of ~ 1 -3 qubit demonstration experiments. Effects that we accidentally discovered, during our recently-funded NSA/ARDA/ARO projects: #DAAD19-01-1-0507 (this Final Report) and #DAAD19-02-1-0203 (QuaCGR support for Dale Li), are an example of this many-spin complexity. As we will describe below, we have discovered unexpectedly large effects in spin dynamics that may be traced back to the presence of very small spin-spin couplings during the implementation of ‘single spin’ rotations using very strong, but finite, pulses. This effect appears to be a ‘looming problem’ that will be faced by nearly all subfields of QIP research as they scale-up to larger numbers of qubits.

2. Background- a widespread failure of conventional NMR theory

Nuclear magnetic resonance experiments rest upon a solid theoretical foundation²⁻⁵. Clever NMR pulse sequences have been developed that evoke specific responses from samples with a wide range of spin Hamiltonians. NMR discoveries such as Hahn's Spin Echo⁶ have been successfully applied across magnetic resonance (e.g., in ESR and MRI), in atomic physics, and even in important QIP experiments.

Thus, we were surprised to discover⁷ that simple experiments on doped Silicon appeared to be inconsistent with conventional NMR theory. For example, coherent signals may be observed well beyond the “ $T_{2_{HE}}$ ” that is measured in two-pulse Spin Echo experiments, *provided that at least one more π -pulse is used*⁷⁻⁹. We accidentally discovered this effect, during our recently-funded NSA/ARDA/ARO projects: #DAAD19-01-1-0507 and #DAAD19-02-1-0203.

It turns out that this effect is much more general than we first thought, having very little to do with the particular characteristics of the doped-Silicon. More recently, we showed that the unique aspects of Silicon NMR (^{29}Si is a spin $I=1/2$, with 4.67% natural abundance, on a diamond lattice), are not key factors in the effect.

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Figure 1 shows the same surprising phenomena¹⁰ in Buckminsterfullerene¹¹ (C₆₀) and Yttria (Y₂O₃), two solids whose sole connections to Silicon are through the form of the homonuclear (“like-spin”) dipolar coupling^{2,4}. Hahn spin echo^{2,6} (HE) measurements (filled points) for both (Fig. 1(a)) ¹³C (spin I=1/2, with 1.11% n.a.) in C₆₀ powder and (Fig. 1(b)) ⁸⁹Y (spin I=1/2, with 100% n.a.) in Y₂O₃ powder are shown (T=300 K, B=12 Tesla). The HE sequence generates a single spin echo

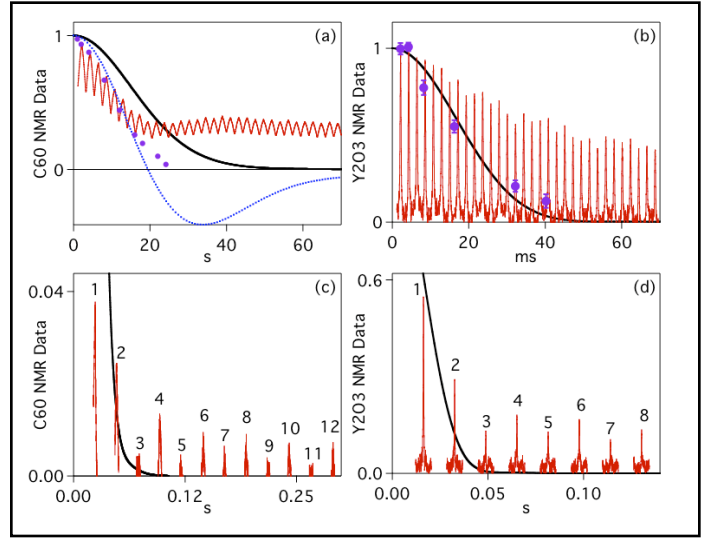


Figure 1.

at time TE using a single- π -pulse (HE: 90_x-TE/2-180_y-TE/2-ECHO); the decay of this signal as a function of increasing TE is a standard measure of “ $T_{2\text{HE}}$ ”. Note that the HE data are in reasonable agreement with the dipolar decay curves calculated assuming either “unlike” (solid black curve (a-d)) or “like” spins (dashed blue curve (a)) on the corresponding lattice². The multiple- π -pulse Carr-Purcell-Meiboom-Gill sequence¹² (CPMG: 90_x-(TE/2-180_y-TE/2-ECHO)^{repeat}) is another standard technique to measure “ T_2 ”. Figure 1 shows that CPMG produces a train of echoes in one experiment (red curves (a-d)), that are clearly detectable long after the Hahn echoes decay to zero...a completely different result for “ T_2 ”! Moreover, CPMG in these samples show both the ‘long tail’ at short TE (Fig. 1(a-b)) and the ‘even-odd effect’ at long TE (Fig. 1(c-d)), as previously reported in Silicon⁷. Similar results have been obtained in several other types of samples, not yet published. To understand why the ‘long-tail’ in the CPMG echo train is surprising, we need to quickly review the conventional NMR theory of these measurements.

3. Conventional NMR theory of π -pulse measurements in dipolar solids.

The NMR signal in both Hahn Echo and CPMG experiments is proportional to: $\langle I_{Y_T} \rangle = \sum_i^{N_{\text{spins}}} \text{Tr} \{ \rho(t) I_{Y_i} \}$

in the rotating frame²⁻⁵. So, to predict the outcome of a particular experiment, we need to calculate the time-dependent density matrix $\rho(t)$. We start from its conventional equilibrium value:

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$$\rho(0) = \sum_i^{N_{spins}} I_{Z_i} \equiv I_{Z_T} \quad (1)$$

which assumes both the strong field and the high temperature approximations². Treating a strong 90_X pulse as a

perfect $\pi/2$ -rotation about X, $\rho(0)$ becomes: $\rho(\delta^+) = e^{i\frac{\pi}{2}I_{X_T}} \rho(0) e^{-i\frac{\pi}{2}I_{X_T}} = I_{Y_T}$. For these dipolar solids, the secular

part of the dipolar coupling gives rise to the spin-spin Hamiltonian: $H_{ZZ} = \sum_{j>i}^{N_{spins}} B_{ij} (3I_{Z_i} I_{Z_j} - \vec{I}_i \cdot \vec{I}_j)$, where

$$B_{ij} = \frac{1}{2} \frac{\gamma^2 \hbar^2}{r_{ij}^3} [1 - 3\cos^2 \theta_{ij}], \gamma \text{ is the gyromagnetic ratio for the spins, and } \vec{r}_{ij}, \text{ the vector between spins } i \text{ and } j,$$

satisfies $\vec{r}_{ij} \cdot \hat{z} = r_{ij} \cos \theta_{ij}$ (the static lab field $\vec{B}_0 \parallel \hat{z}$).

In between pulses, the full spin Hamiltonian $H_0 = H_{\Omega_Z} + H_{ZZ}$ includes a Zeeman

term: $H_{\Omega_Z} = \sum_i^{N_{spins}} \hbar \Omega_{Z_i} I_{Z_i}$, where Ω_{Z_i} is the magnetic shift offset for spin i (relative to on-resonance spins). In the

clean limit of a dipolar solid (appropriate for these samples), Ω_{Z_i} is primarily due to bulk diamagnetism, and thus

will vary so little across so many spins that we drop the index i . In that case, H_{Ω_Z} commutes with H_{ZZ} , so we can

define the Unitary operators corresponding to free evolution for time $\tau = TE/2$:

$$U_0 \equiv e^{-\frac{i}{\hbar} H_0 \tau} = e^{-\frac{i}{\hbar} H_{\Omega_Z} \tau} e^{-\frac{i}{\hbar} H_{ZZ} \tau} \equiv U_{\Omega_Z} U_{ZZ} \quad (2).$$

Next, we invoke the conventional infinite pulse ($H_1 = \infty$) approximation, also known as the δ -function pulse

approximation. This conventional approximation enables us to treat any strong 180_Y pulse as a perfect π -rotation

about Y, or $P_Y^{H_1=\infty} \equiv e^{i\pi I_{Y_T}}$. Perfect π -pulses "refocus" the Zeeman term, since

$$(P_Y^{H_1=\infty}) U_{\Omega_Z} (P_Y^{H_1=\infty})^{-1} = (U_{\Omega_Z})^{-1}, \text{ but they "do nothing" to the dipolar term, since}$$

$$(P_Y^{H_1=\infty}) U_{ZZ} (P_Y^{H_1=\infty})^{-1} = U_{ZZ}. \text{ Using this, the density matrix at the time of the first Spin Echo (t=TE) is:}$$

$$\rho^{SE^{H_1=\infty}}(TE) = (U_{ZZ})^2 I_{Y_T} \{inv\}, \text{ where } \{inv\} \text{ is the inverse of all operators to the left of } I_{Y_T}. \text{ Similarly, the } n^{\text{th}}$$

Spin Echo (SEn) produced by the CPMG experiment at time $n \times TE$ is described by:

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$$\rho^{SEn H_1=\infty}(nTE) = (U_{ZZ})^{2n} I_{Y_T} \{inv\} \quad (3).$$

Since $\left(e^{-\frac{i}{\hbar} H_{ZZ} \tau}\right)^{2n} = \left(e^{-\frac{i}{\hbar} H_{ZZ} n\tau}\right)^2$, these conventional approximations predict that the signal at a particular total

time should be independent of whether we use one or more π -pulses to measure it.

This is so important, that we will say it again. If “perfect, δ -function” 180° -pulses are applied to a system described by $H_0 = H_{\Omega_z} + H_{ZZ}$, then one can show analytically that the results in Fig. 1 are impossible^{2,7,10}. Clearly, however, Fig. 1 shows that while the Hahn echo data (purple dots) are roughly consistent with the expected dipolar decay, the CPMG echoes (red lines) persist to times well beyond this ‘limit’.

It is natural to attribute these puzzling results to extrinsic pulse imperfections. For example, the above ‘*this is impossible*’ statement would have to be carefully checked, if:

- 1) we used the wrong pulse angle (e.g., “ δ -function” 170° -pulses),
- 2) we had big phase transients (e.g., instead of ‘ 180° ’, the real pulse looked like a composite $+4x, 178y, -3x$),
- 3) we used pulses that varied across the sample (e.g., so ‘ 180° ’ for one spin looked like 165° for another)^{2,10,11}.

Over the course of the project, we have spent a great deal of time and effort investigating these, and other such extrinsic effects. However, even after great efforts to improve the pulses, and to quantify effects of residual errors, the CPMG data remain surprising. This forced us to consider another, intrinsic effect: real pulses are never δ -functions (i.e., they all have non-zero duration, so H_0 has time to act). This enables new dynamics¹⁰ that are strictly forbidden in the conventional δ -function, π -pulse approximation; this effect is thus broadly relevant to QIP¹³. The conventional approximation that very strong π -pulses are in the “ $H_1=\infty$ ” limit, appears to be in trouble, at least for interacting spin systems.

4. Many-body Modifications to Pulse Action in the Finite Pulse Limit

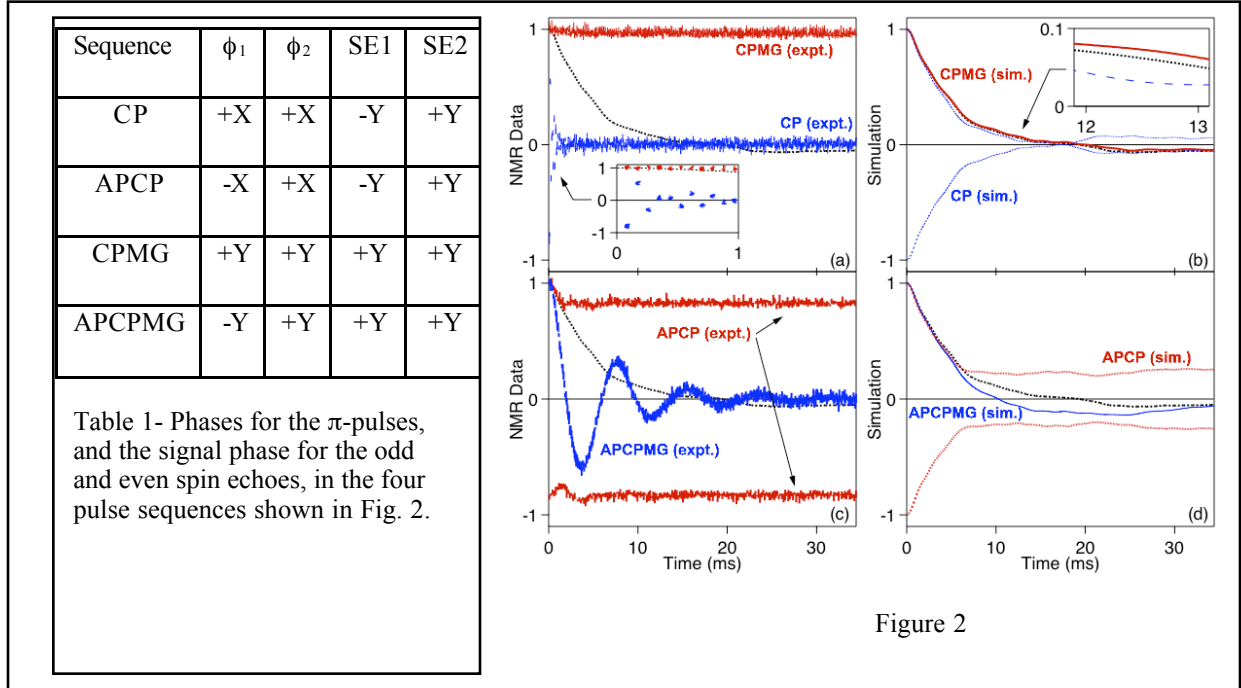
The measurements exhibit an interesting pulse sequence sensitivity (PSS), that is an important clue about the failure of conventional NMR theory. Defining a generic multiple π -pulse sequence as: $90_x(-TE/2-180_{\phi_1}-TE/2-SE1-TE/2-180_{\phi_2}-TE/2-SE2)^{\text{repeat}}$, it turns out that changing the phase (or direction in the rotating frame) of the π -pulses as defined in Table 1 (below), dramatically changes the measured echo trains.

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This PSS is present for all TE, but is particularly striking in the short TE limit. Figure 2(a,c) show ^{29}Si measurements in lightly doped Si:Sb at room temperature, in $B=11.75$ Tesla, with $TE=72$ ms, and a π -pulse duration $t_\pi=14$ ms. Only the very “tops” of the echoes can be seen for such short TE, but the message is clear: multiple π -pulse echo trains may either *freeze-out or accelerate the expected dipolar decay of the NMR signal, depending upon the phases used for the π -pulses*; on the surface, this appears similar to the quantum dynamical



decoupling limit of the Quantum Zeno and Inverse Quantum Zeno effects¹⁴. This pulse phase sensitivity has an intrinsic origin, arising from the surprisingly non-negligible effects of the dipolar coupling during strong, but finite, pulses. Clearly, this complicates the application of “quantum bang-bang” control sequences to systems, limiting one important method for extending coherence times¹⁵.

To see this, recall that during an ideal rf pulse along the ϕ_i direction, the total spin Hamiltonian becomes:

$$H_{P\phi_i} = -\hbar\omega_1 I_{\phi_i} + H_0 = -\hbar\omega_1 I_{\phi_i} + H_{\Omega_z} + H_{ZZ}, \text{ where } \omega_1 = \gamma H_1 \text{ for all spins; the corresponding Unitary}$$

operator for a π -pulse is $P_{\phi_i} = e^{-\frac{i}{\hbar} H_{P\phi_i} t_\pi}$. Formally, we may write the density matrix at the time of SE2 for all four pulse sequences as:

$$\rho^{SE2}(2TE) = U_0 P_{\phi_2} U_0 U_0 P_{\phi_1} U_0 I_{Y_T} \{inv\} \quad (4).$$

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When we take the “ $H_1=\infty$ ” limit, the H_0 term is killed (since $t_\pi=0$), and each pulse acts like a perfect rotation, collapsing all four variants of Eqn. 4 onto the earlier definition of $\rho^{SE2^{H_1=\infty}}(2TE)$ (Eqn. 2), *so there can be no PSS*. However, for any real experiment, $t_\pi>0$, so the $H_{\Omega_z} + H_{ZZ}$ terms in the pulse Hamiltonian might have some effect. V.V. Dobrovitski was the first to point this out to us, based upon his simulations of our experiments¹⁶. In fact, a small, but definite effect of these terms is seen in our own finite amplitude, non-zero duration pulse simulations (with $N_{\text{spins}}=7$) of the experiments (Fig. 2(b,d)). Both the red and blue simulations, keeping the full Hamiltonian during the pulse, move away from the black reference curve (which sets H_0 to zero during the finite pulses), and towards the corresponding experimental trace. At the same time, the simulations badly underestimate the experimental effects. Understanding what was going on required more work, and that took place after the end of both ARO/NSA/ARDA-funded projects. In fact, the delay in submission of this Final Report was caused by our confusion at the end of the project period. We felt fairly confident that something interesting was behind the results, but at that point, we didn’t understand “what was going on”...only “what wasn’t happening”. Now that we have a better understanding, we are finally able to describe what this project uncovered, and what it means for QIP.

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5. Recent Progress

We now know that the calculations in Fig. 2(b,d) underestimate the true effects of the dipolar coupling¹⁰. This is because H_{zz} is a sum over all spin pairs in the system, and limitations on classical computer speed force us to artificially truncate the system size (e.g., $N=9$ is the biggest we've ever been able to compute). Based on further studies (carried out after the end of this ARO/NSA/ARDA project), we can show that the small scale of the B_{ij} coupling in Silicon makes the restriction to small N even more of a problem...which is not what we were expecting, going into this project.

To make progress on this problem, we: **1)** implemented numerical simulations of spin dynamics using the exact, time-dependent Hamiltonian, **2)** derived analytic expressions for average Hamiltonian^{2,4} of these pulse sequences, and **3)** carried out numerical simulations using the leading order terms in the average Hamiltonian (i.e., $H^{(0)}$ and $H^{(1)}$). This three-pronged strategy was a fruitful way to combine the power of theory and computation.

One example of an unexpected result that came out of this effort is shown in Fig. 3. The main figure shows calculated CPMG decay curves in the presence of pure dipolar coupling H_{zz} . For the dashed curve (with $N=4$), we set H_{zz} to zero by hand during each π -pulse, which forces the curve to 'artificially' be in the " $H_I=\infty$ ", δ -function pulse limit. The solid curves are full, exact calculations, showing how the 'tail' grows with spin number N . Curves average over 400 disorder realizations (DRs) of spin locations on a lattice, except for the case $N=8$, which uses 80 DRs (since our fastest computer can simulate only 3 DRs every 24hrs for $N=8$). All curves are for

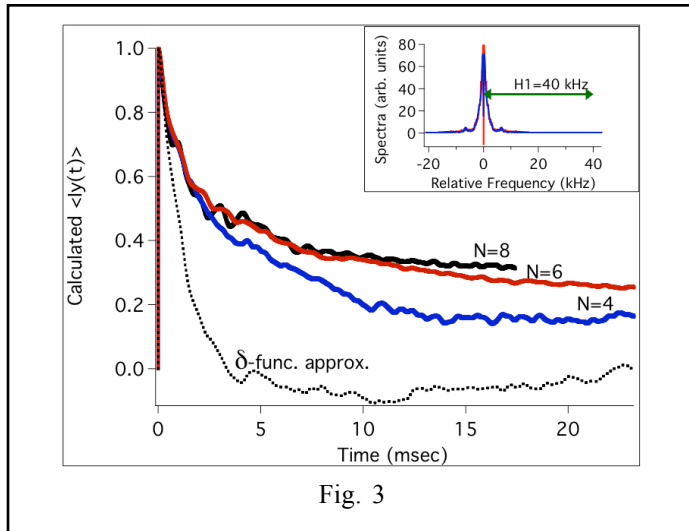


Fig. 3

$TE = 2$ ms. The exact calculations disagree markedly from the δ -function approximation, after starting out together. Moreover, there is an N -number dependence to the different 'tail' heights. These are surprising and non-trivial results, given the conditions used in the simulation. For example, Fig. 3(inset) shows that the applied π -pulse strength ($H_I=40$ kHz) is much larger than the linewidth (~ 2.2 kHz, due entirely to dipolar coupling), and furthermore that

the calculated spectra are essentially identical for both the $N=4$ (red) and $N=6$ (blue) cases.

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This result has definite implications for quantum information processing, as can be seen in Figure 4, which shows the calculated evolution of the $2^6 \times 2^6$ element density matrix for a six spin system during CPMG, with conditions as in Fig. 3 (only 1 DR is shown in Fig. 4).

The leftmost boxes correspond to the initial state $\rho(\delta^+) = I_{Y_T}$. The red-white-blue color scale shows the phase angle for ‘large’ elements; black cells have negligible magnitude. In the top row, we set H_{zz} to zero by hand during each π -pulse, (i.e., the δ -function pulse limit). The selection rules for this case allow coherence to spread only to elements corresponding to $\Delta M_z = \pm 1$, as shown in top row. The bottom row uses the full, exact, Hamiltonian. Clearly, the allowed coherence transfers are quite different for approximate (top row) and for exact (bottom row) pulses. In the exact calculation, the initial coherence spreads over a much larger number of cells of the density matrix, since more coherence transfer pathways are opened up. In our CPMG experiments, which measure just $\langle I_{Y_T}(t) \rangle$, the bottom case actually leads to more signal, which is an interesting result, given the complexity of the coherence pattern! For QIP applications, which generally care about the full state of the density matrix, there is clearly an important difference between the top (approximate-pulse) and bottom (exact-pulse) rows of Figure 4.

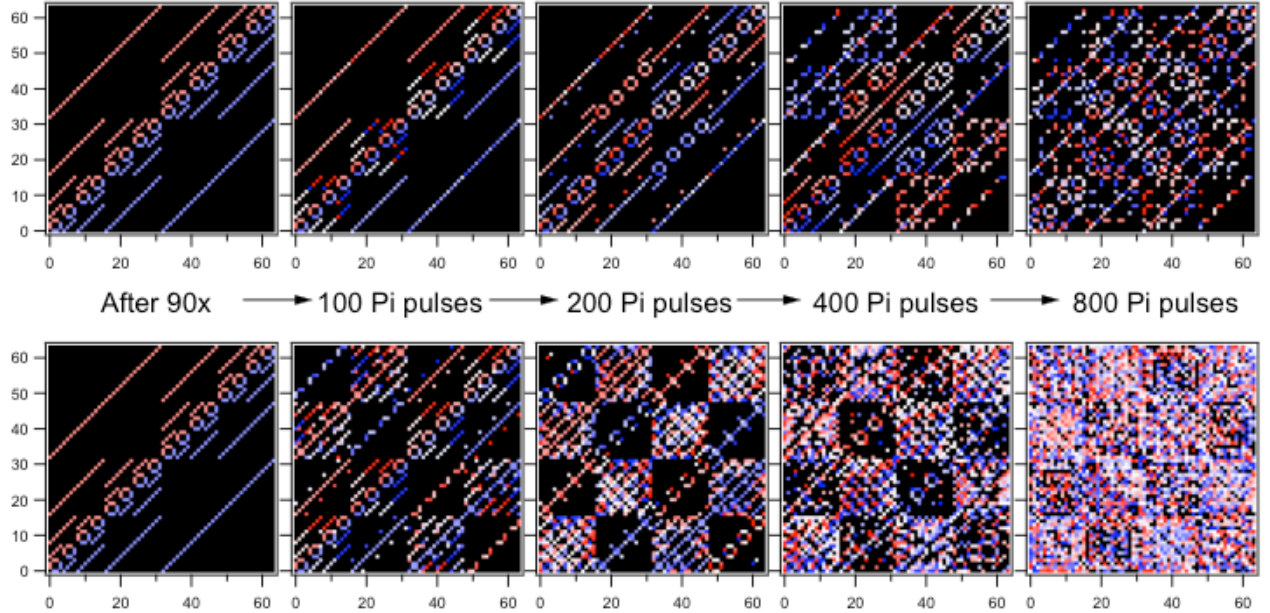


Figure 4

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The δ -function pulse approximation is widely applied in NMR. Given the striking difference between the top and bottom rows of Fig. 4, why doesn't this approximation break down regularly? Well, even if the approximation is bad, it might be hard to tell, since most NMR experiments measure quantities like $\langle I_{Y_T}(t) \rangle$, and therefore only part of the density matrix matters. The reason that we spent so long tracking this effect down is just because of the unusual behavior of the CPMG sequence shown in Fig. 3, where anyone would agree that there is a striking difference between the dashed (approximate-pulse) and solid (exact-pulse) curves. Without that discrepancy, we wouldn't have looked into this in detail. In fact, for other pulse sequences (e.g., CP), the calculated signal using either approximate- or exact-pulses can look much more similar...there isn't any obvious puzzle to solve. On the other hand, even for cases like CP, the density matrix evolution will be drastically different, depending upon whether we use either approximate- or exact-pulses, so this is a big problem for any application which cares about the full density matrix...such as Quantum Information Processing.

6. Conclusions

To summarize, each finite H_1 π -pulse, even a very strong one with no errors, leaves an imprint on the time evolution of the density matrix, because it is definitely not a pure π -rotation about an axis in the X-Y plane. This is an intrinsic effect, due to the presence of H_0 during the pulse, that cannot be avoided in any real experiment. Note that extrinsic effects such as: Ω_{Zi} or ω_1 that vary from spin to spin, or an incorrect pulse angle, or phase transients at the edges of the pulse, may also alter the NMR signal in non-trivial ways, but the corresponding equation will be qualitatively similar to the intrinsic form of Eq. (4). Ignoring these issues will make QIP much more difficult. On the other hand, developing an improved understanding of these many-body corrections to pulse action will enable the rational design of pulse sequences optimized to achieve desired coherence transfer pathways in real systems with large numbers of qubits. This will be relevant to most subfields of QIP within the next ~5-10 years, as people scale-up to larger system sizes.

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