Dynamic decoherence control of a solid-state nuclear quadrupole qubit

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Recent experiments have demonstrated that optically active centers in solids and in particular rare-earth ions in crystals show promise as an alternative to atomic based ensembles for quantum information processing applications. Rare-earth doped ions provide multi-level systems possessing optical transitions with long coherence times and in crystalline hosts, high optical densities are achievable. These solid state centers have the significant advantage over atomic systems that they are stationary, thus allowing spatial and frequency variation in an ensemble induced by interactions with light fields to be maintained for appreciable times. This last attribute has already been exploited extensively in the field of time domain optical processing, where complex temporal and spatial relationships between different interacting light fields are employed to process classical information, in a manner not possible in atomic systems.

A common feature of many of the proposed ensemble based quantum information processing applications, including quantum memories, single photon sources and quantum repeaters, involves the storage of quantum information on long lived low lying states. In the case of rare-earth centers it is proposed to store the quantum information on the centers’ ground state hyperfine transitions. The duration and fidelity that such information can be stored depends on the coherence time of the hyperfine transitions and, therefore, it is important to maximize these coherence times. The coherence times of hyperfine transitions in rare-earth centers are typically of the order of ms but in recent work we have demonstrated ways where these can be extended and have achieved a coherence time of 82 ms in the case of $^{111}$In $\leftrightarrow +3/2$ transition in $P_{r}^{3+}:Y_{2}SiO_{5}$. In this letter we demonstrate a further dramatic increase of the coherence time to several tens of seconds through the use of a Bang Bang dynamic decoupling sequence.

Dynamic decoupling methods for open systems were initially developed for use in NMR spectroscopy to selectively remove contributions to the spin Hamiltonian. There is growing interest in applying these techniques and in particular “Bang Bang” decoupling sequences to quantum information systems to decrease their rate of decoherence. “Bang Bang” sequences operate by decoupling the quantum system from the bath through the application of a periodic control Hamiltonian. In the current work this is implemented by a series of pairs of hard pulses ($\pi$, $-\pi$) separated by the cycling time $\tau_{c}$. The Bang Bang pulse sequence can theoretically rephase all the coherence in the quantum system, thereby making $T_{2} = T_{1}$ if the following criterion is met.

$$\omega_{c} \tau_{c} \lesssim 1$$

where $\omega_{c}$ is the cutoff frequency of the dephasing bath and $\tau_{c}$ is the period between the $\pi$ pulses in the Bang Bang sequence. The pulses are assumed to be ‘hard’ such that during the pulse any evolution of the state other than the action of the driving field can be assumed to be negligible.

For Pr ions substituting for Y in $Y_{2}SiO_{5}$ the main source of decoherence of the its ground state hyperfine transitions is magnetic interactions with nuclei in the host. Y possess a nuclear spin and has a magnetic moment of $\gamma_{Y} = 209 \text{MHz} / G$. The other nuclear spin in the host is $^{29}Si$ found in natural abundance ($\sim 4\%$) with a moment of $845 \text{MHz} / G$. The magnetic field seen by any given Pr ion fluctuates over time due to resonant cross relaxations between the host spins. The sensitivity can be calculated using the reduced Hamiltonian of the electronic ground state hyperfine structure. Praseodymium has a nuclear spin of $5/2$ and as a result of a pseudoquadrupole interaction its electronic ground state splits into three doubly levels with splittings of the order of 10 MHz. The final degeneracy can be lifted by a magnetic field through an enhance nuclear Zeeman interaction. In the region where the applied field produces anticrossings a field direction and magnitude can be found such that there is no first order Zeeman shift for the $m_{1} = -1/2 \leftrightarrow +3/2$ hyperfine transition, demonstrated in recent work. It has been shown that the dephasing perturbations acting on the Pr ions are predominantly occurring on a timescale longer than $\sim 10\text{ms}$. Given that Rabi frequencies greater than $100\text{kHz}$ are readily achievable the period between the applied pulses in the...
The coherence generated by the initial pulse is rephased by a N times. The laser is common to all pulse sequences.

FIG. 1: Pulse sequences used in experiment: a) 2 pulse spin echo, b) Inversion Recovery $T_1$, c) Bang Bang pulse sequence. The laser is common to all pulse sequences.

**Bang Bang sequence** can be made such that the inequality (1) is satisfied for the frequencies of the dominant perturbations.

As in previous work a $Pr^{3+}:Y_2SiO_5$ 0.05% concentration crystal maintained at temperature of $\sim 1.5K$ in a liquid helium bath cryostat is. Yttrium orthosilicate ($Y_2SiO_5$) is a low symmetry host with two crystallographically inequivalent sites where Pr can substitute for Y, labelled ‘site 1’ and ‘site 2’. Site 1 ions are used in this work. The coordinate system chosen is the $C_2$ axis is $x$, $z$ is the direction of the predominate polarization of the optical $^1H_1 - ^1D_2$ transition and $x$ is perpendicular to both.

The magnetic fields to achieve the Critical Point field configuration ($B_{CP} = (732, 173, -219)$ G) were supplied by two orthogonal superconducting magnets supplying a $z$ field, and $x, y$ field. The sample was rotated about the $z$ axis to provide the correct ratio of fields along the $x$ and $y$ axes for the critical point in magnetic field space. The field in the $x, y$ plane could also be adjusted using a small correction coil mounted orthogonal to the main $x, y$. The inhomogeneity in magnetic field across the sample was measured using a hall probe to be <2G.

Raman heterodyne was employed to investigate the ground state hyperfine transitions using an experimental setup similar to that described in previous work. The experiment was performed using a Coherent 699 frequency stabilized tunable dye laser tuned to the $^3H_1 - ^1D_2$ transition at 605.977nm. The laser’s frequency was stabilized to a sub kilohertz linewidth. The laser power incident on the crystal was 40mW, focused to $\sim 100\mu m$ and could be gated using a 100MHz acousto-optic modulator. The hyperfine transition was excited using a six turn coil with a diameter of 5mm, driven by a 10W RF amplifier resulting in a Rabi frequency $\Omega, f = 91kHz$. The RF pulse and digital control sequences were generated using a direct digital synthesis system referenced to an oven controlled crystal oscillator. The pulse sequences used in the experiment are illustrated in Fig. 1. The Raman heterodyne signal, seen as a beat on the optical beam, was detected by a 125MHz photodiode. This signal was analyzed using a mixer and a phase controlled local oscillator referenced to the RF driving field.

At the Critical Point field the $m_I = -1/2 \leftrightarrow +3/2$ transition was observed at 8.646 MHz with a inhomogeneous linewidth of 4kHz. This linewidth was found to be insensitive to changes in the magnetic field of the order of $\sim 10G$.

Prior to applying each Raman heterodyne pulse sequence the sample was prepared with the optical/RF repump scheme as shown in Fig. 2. The repump frequencies were $\omega_r = 18.2MHz$, $\omega_1 = 12.2$, $\omega_2 = 15.35MHz$ and $\omega_3 = 16.3MHz$. The repump RF was pulsed with a duty cycle of 10% to reduced sample heating, while the repump laser frequency $\omega_r$ was scanned 200kHz to hole burn a trench in the inhomogeneous optical line where detection would take place. This repump scheme ensures that all Pr ions interacting with the laser radiation are forced into the $m_I = -1/2$ state, creating a pure state ensemble. It also ensures there is no initial population near the laser frequency used for Raman heterodyne detection. The use of a sub kilohertz linewidth laser and the repump scheme resulted in a significant improvement in the signal to noise compared to work performed earlier.

Figure 3 shows the decay of the amplitude of two pulse spin echo as a function of the delay between the pulses. Two data sets are shown, the first is for an applied magnetic field optimized for long decay times and the second for a field detuned by 5G in the $z$ direction from the optimal value. The significantly longer decay time for the optimised field in the present work, compared to previous work, is attributed to better identification of the critical point enabled by the addition of the correction coil enabling significantly more precise magnetic field adjustments. Besides being longer the decay can no longer be described by standard echo decay function with a single time constant. There are three distinct regions. For pulse separations less than 20ms the decay rate is less than 1/4s$^{-1}$. At 30ms there is a distinct shoulder with the decay rate increasing to 1/0.4s$^{-1}$ as the pulse separation reaches 60ms. From 150 ms onwards the de-
decay rate asymptotically approaches a value of $1/0.86s^{-1}$. This asymptotic decrease in the decay rate was only observed for magnetic fields within 0.5G of the optimal field. When the field was more than 0.5G away from the critical point a simple exponential decay was observed for delays longer than 50 ms.

The shoulder in the decay at 30 ms is interpreted as indicating that the majority of the dephasing is due to perturbations that occur on time scales between 10 and 100 ms. The asymptotic behavior of the decay is attributed to a variation in the $T_2$ within the ensemble resulting from inhomogeneity in the magnetic field across the sample. Ions experiencing a field closer to the Critical Point condition will have a longer $T_2$ and consequently their contribution to the echo intensity will dominate for long pulse separations. The asymptotic decay rate therefore gives an upper limit for the contribution to the decoherence rate due to second order magnetic interactions.

The Bang Bang pulse sequence was investigated using an initial delay of $\tau_1 = 1.2 ms$ and varying the cycling time $\tau_c$ from 20 ms to 0.5 ms, as shown in Fig. 4. Also shown in Fig. 4 is the result of an inversion recovery measurement used to determine the lifetime of the transition. The inversion recovery measurements were performed using the pulse sequence described in Fig. 1(b). As can be seen in Fig. 4 the Bang Bang sequence significantly increases $T_2$, though for the shortest value of $\tau_c$, $T_2/T_1 < 1/4$. In Figure 5 the coherence times $T_2$ for each of the data sets are plotted as a function of the cycling time $\tau_c$. This shows that for $\tau_c < 5 ms$, there are significant gains in $T_2$ made by reducing $\tau_c$, while reducing $\tau_c$ further only slightly increases in $T_2$. Also shown in Figure 5 is the same measurements made with the dc magnetic field detuned so as to increase the transitions magnetic field sensitivity. The field was detuned such that the two pulse echo $T_2$ was reduced to 100 ms. Although for long $\tau_c$ there is a significant reduction in the $T_2$ observed using the Bang Bang for the measurements made with the detuned field, $T_2$ in the limit of short $\tau_c$ appears to be the same for the two magnetic field conditions. This suggest that the residual decoherence in the limit of short $\tau_c$ is not predominately due to magnetic field fluctuations. The residual decoherence is possibly due to detuning and pulse area errors in the decoupling sequence.

To assess how well arbitrary quantum state are preserved by the Bang Bang pulse sequence we performed process tomography on the input state and for 1, 10, 100 and 1000 iterations of the Bang Bang cycle. The initial delay was $\tau_1 = 1.2 ms$, with a cycling time of $\tau_{uc} = 2 ms$. The total period over which the tomography was performed was $\sim 4 ms$, $\sim 40 ms$, $\sim 400 ms$ and $\sim 4 s$ respectively. The imaginary component of the process tomography is only shown for the input state since it never contributed more than 10% of the signal. Ideally the Bang Bang process operator is the identity matrix, leaving the state unchanged. It was observed that the

![Image](335x397 to 543x507)

**FIG. 3:** Spin echo decay at the Critical Point field ($\odot$), and detuned from the Critical Point field by $\sim 2 G$ ($\odot$) and 5G ($\odot$) in the Z direction.

![Image](335x576 to 543x740)

**FIG. 4:** Bang Bang decoupled echo decays with $\tau_c = 7.5 ms (\times), 10 ms (\odot), 15 ms (\nabla), 20 ms (\idot)$ corresponding to $T_2 = 27.9, 21.1, 15.2, 10.9 s$. Inversion recovery measurements ($\odot$) yield $T_1 = 145 s$.

![Image](62x556 to 291x740)

**FIG. 5:** Dependence of decoherence time on the Bang Bang cycling time $\tau_c$. both at the critical point ($\odot$) and with the magnetic field misaligned to give a coherence time of $T_2 = 100 ms (\times)$. Trend lines are included but do not represent a physical model.
FIG. 6: Process tomography of the Bang Bang pulse sequence for 1, 10, 100 and 1000 iterations for both experiment and theoretical modeling. The imaginary component of both the experiment and modeling is always zero and is omitted for clarity.

Component of the Bloch vector in the coherence plane for a given state was preserved well, while the population component of the Bloch vector rapidly decayed. The fidelity of the Bang Bang process for 1, 10, 100 and 1000 iterations was 99%, 65%, 54% and 43% respectively.

The evolution of the ensemble was modeled using the Bloch equations assuming an infinite $T_1$ and $T_2$, with an inhomogeneous linewidth of 4kHz (FWHM) and a Rabi frequency of 100kHz. The results from this modeling are shown in figure 6 along side the experimental data. Despite the model not including any homogeneous dephasing the main features of the simulated process tomography (figure 6) closely matches those of the experimental data. In particular the rapid decay of the population component of the Bloch vector compared to the coherence components. Further simulations indicated that the decay rate of the population terms can be reduced by increasing the ratio of the Rabi frequency to the linewidth. A suitable criteria for when the application of the Bang Bang pulse sequence is useful for preserving arbitrary quantum states is when the decay rate of the population terms in the presence of the pulse sequence is slower than that of the coherence terms in the absence of the sequence. For the present case where $T_2 = 0.86s$ the simulation indicates that to meet this criteria it will be necessary to achieve a ratio of Rabi frequency to linewidth of $\Omega_{RF}/\omega_{inh}\approx 100$. There is limited capacity to increase the Rabi frequency of the driving field without the possible excitation of off-resonant transitions. Therefore for the application of the Bang Bang pulse sequence to the $m_I = -1/2 \leftrightarrow +3/2$ transition to be useful it will be necessary to reduce the inhomogeneous broadening of the transition by a factor of $\sim 10$.

The large change in magnetic field sensitivity as the Critical Point field is approached with no corresponding change in the inhomogeneous linewidth of the transition suggests that the inhomogeneous broadening of the transition at the Critical Point field is not due to magnetic interactions. The inhomogeneous broadening at the Critical Point field is probably due to strain within the crystal which is not intrinsic to the site and can be reduced by refining standard crystal growing techniques. A reduction in strain broadening by over an order of magnitude has been achieved in analogous materials through reducing the dopant concentration. Irrespective of reaching the desired ratio of Rabi frequency to inhomogeneous linewidth, methods of designing pulse sequences more robust to Rabi and detuning errors have been proposed.

In conclusion the application of the Bang Bang pulse sequence demonstrates that dynamic decoupling techniques are applicable to correcting quantum errors on nuclear spin transitions of $^{3+}Pr^{3+}Y_2SiO_5$. This work realises very long hyperfine decoherence times, greater than 30s, in a solid state optical Λ system suitable for quantum information processing applications.

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