Method of extending hyperfine coherence times in $Pr^{3+} : Y_2SiO_5$

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(Dated: February 26, 2018)

In this letter we present a method for increasing the coherence time of praseodymium hyperfine ground state transitions in $Pr^{3+}:Y_2SiO_5$ by the application of a specific external magnetic field. The magnitude and angle of the external field is applied such that the Zeeman splitting of a hyperfine transition is at a critical point in three dimensions, making the first order Zeeman shift vanishingly small for the transition. This reduces the influence of the magnetic interactions between the praseodymium ions and the spins in the host lattice on the transition frequency. Using this method a phase memory time of 82ms was observed, a value two orders of magnitude greater than previously reported. It is shown that the residual dephasing is amenable quantum error correction.

PACS numbers:

There is growing interest in the use of nuclear spin states associated with dopant ions in a solid state host to store and manipulate quantum information[1, 2, 3]. These applications require the relevant spin transitions to have long coherence times. This can be challenging to achieve in a solid state system due to magnetic interactions between the dopant ion and spins within the host. A spin free host can be utilized to minimize these interactions[2], however not all dopant species of interest are chemically compatible with such hosts. An example of a class of dopants where no satisfactory spin free host has been identified, are the rare earth ions. Because of the potential to manipulate their spin states optically, the use of rare earth ions has been proposed in a number of quantum information applications[1, 3]. In this letter we investigate the decoherence in the ground state hyperfine transitions in $Pr^{3+}:Y_2SiO_5$ and demonstrate a new method for increasing their coherence times. This technique is expected to be applicable to a wide range of spin systems.

$Pr^{3+}:Y_2SiO_5$ was chosen to investigate since along with its use in a quantum computing architecture having been proposed by Ichimura et al.[1], it has already been used in slow and stopped light demonstrations[4, 5]. The longest coherence time ($T_2$) stated in the literature for a ground state hyperfine transition in $Pr^{3+}:Y_2SiO_5$ is 500$\mu$s[6]. This is likely to be insufficient for practical quantum computing applications given that the hyperfine transition frequency is $\sim 10 MHz$, limiting the Rabi frequencies of any driving field to less than a few MHz so as to be transition specific. This only allows of the order of 1000 operations to be completed within $T_2$.

Yttrium orthosilicate has symmetry given by the $C_{2h}$ space group[6] with two formula units $Y_2SiO_5$ per translational unit. This gives four different sites at which the Pr can substitute for Y. The four sites can be divided into two pairs. The two pairs labelled 'site 1' and 'site 2' have different crystal field splittings and hence different optical and hyperfine frequencies. The members of each pair, labelled 'a' and 'b', are related to each other by the crystal’s $C_2$ axis and their optical and hyperfine frequencies are degenerate in zero field. The Pr sites in $Y_2SiO_5$ have electronic singlet ground states with hyperfine ground state interactions described by the following Hamiltonian:

$$H = B \cdot M \cdot I + I \cdot Q \cdot I$$

where $B$ is the magnetic field vector, $I$ is the vector of nuclear spin operators, $M$ is the effective Zeeman tensor combining nuclear and electronic Zeeman interactions and $Q$ is the effective quadrupole tensor combining the quadrupole and second order magnetic hyperfine interaction, known as the pseudo-quadrupole[7]. The one naturally occurring isotope of Pr has a nuclear spin of 5/2. $M$ and $Q$ for site 1 were recently determined for $Pr^{3+}:Y_2SiO_5$ to be:

$$Q = R(\alpha, \beta, \gamma) \begin{bmatrix} -E & 0 & 0 \\ 0 & E & 0 \\ 0 & 0 & D \end{bmatrix} R^T(\alpha, \beta, \gamma)$$

$$M = R(\alpha, \beta, \gamma) \begin{bmatrix} g_x & 0 & 0 \\ 0 & g_y & 0 \\ 0 & 0 & g_z \end{bmatrix} R^T(\alpha, \beta, \gamma)$$

where $E = 0.5624 MHz$ and $D = 4.4450 MHz$, $(g_x, g_y, g_z) = (2.86, 3.05, 11.56) kHz / G$, and the Euler angles are $(\alpha, \beta, \gamma) = (-99.7, 55.7, -40) \deg$. These values are for the crystal aligned with the $C_2$ axis in the $y$ direction, and the $z$ axis is the direction of linear polarization of the praseodymium optical transitions. These tensors are highly anisotropic due to the low symmetry of the site.

The dominant dephasing mechanism for the Pr hyperfine ground states in $Y_2SiO_5$ is due to magnetic interactions with the Y nuclear spins in the host. Yttrium has a nuclear spin of 1/2 and a gyromagnetic ratio of...
$g_n = 200Hz/G$. Nearest neighbor Y ions induce a magnetic field at the Pr site of the order of 0.1G. The direct magnetic dipole-dipole interaction between the Pr and a nearest neighbor Y ion is of the order of hundreds of Hertz. This interaction can induce decoherence via two mechanisms. The first mechanism is that the Y ions can exchange spin with each other, resulting in a fluctuating magnetic field at the Pr site. For resonant Y nuclei the rate of exchange between two nearest neighbor ions is of the order of tens of Hertz. The second mechanism involves the excitation of nearly degenerate transitions where one or more Y spin flips are induced along with the desired change in the Pr hyperfine state.

To suppress these dephasing mechanisms for a given Pr ground state hyperfine transition we propose applying a specific magnetic field such that transition’s linear Zeeman shift about this field value is zero. The main requirements for this technique are the existence of a zero field splitting in the spin states and the ability to apply a sufficiently strong magnetic field such that the Zeeman splitting is comparable to the zero field splittings. This method should be applicable to many $I > 1$ spin systems.

Using equation (2) and the parameters for the site 1 given above, all the magnetic field values resulting in a zero first Zeeman shift were identified. The case chosen to experimentally investigate was at a magnetic field of $B_{CP} = \{732, 173, -219\}G$ on the $m_I = +1/2 \leftrightarrow +3/2$ transition at 8.63 MHz as it was found to have the smallest second order Zeeman shift. Around this magnetic field value the transition energy as a function of magnetic field has a turning point in the $y$ and $z$ axis while the $x$ axis has a slow inflection point. As the required magnetic field was not along the crystal’s $C_2$ axis the degeneracy between site 1a and site 1b was lifted with only the site 1a ions being at the critical point.

Raman heterodyne two and three pulse spin echo sequences were used to investigate the decoherence and spectral diffusion in the ground state hyperfine transitions (figure 1). The experiment was performed using a Coherent 699 frequency stabilized (1MHz FWHM) tunable dye laser tuned to the $3^2H_4 \rightarrow 1^2D_2$ transition at 605.977nm. The laser was gated using a 100MHz AOM such that there was no laser radiation applied to the sample during the RF pulse sequence (figure 1). The laser power incident on the crystal was 40mW, focused to ~100μm and a RF Rabi frequency of $\Omega_{RF} = 300kHz$. The $Pr^{3+:Y_2SiO_5}$ (0.05% concentration) crystal was held at ~1.2K for the duration of the experiment. The laser prepared a population difference in the excited ions for 5s before the pulse sequence and was scanned over 1.2GHz of the optical transition to avoid hole burning effects. The laser is off during the RF pulse sequence to minimize coherence loss from optical pumping.

The magnetic fields were supplied by two superconducting magnets one along the $z$ axis and the other in the horizontal plane. The field was then rotated by $13 \pm 0^\circ$ to provide the correct ratio of fields along the $x$ and $y$ axes for the critical point in magnetic field space. Corse adjustment of the field was done by comparing the Raman heterodyne spectrum to theoretical simulations as shown in figure 2. Fine adjustment of the magnetic field utilized perturbing coils along each axis and associated lock-in amplifiers to perform field sensitivity measurements on the frequency of the $m_I = +1/2 \leftrightarrow +3/2$ transition. The rotation of the sample and the current driving the magnets were iteratively adjusted to minimize the sensitivity of the desired transition. Final adjustments of the field values were made by minimizing the echo decay rate. The magnetic field values were accurate to within $\pm 0.1G$ of the critical point.

The two pulse echo data (figure 2) shows three echo sequences at the critical point magnetic field configuration with the zero field echo sequence for reference. Since the field configuration is transition specific the $m_I = +1/2 \leftrightarrow +3/2$ transition at site 1a is at a critical point while the two other transitions are not.

With the application of the field for all the transitions there was observed a significant increase in the coherence time. For the $m_I = +3/2 \leftrightarrow -3/2$ transition $T_2$ was 5.86ms while for the $m_I = +1/2 \leftrightarrow +3/2$ transition at site 1b it was 9.98ms (figure 3). Applying a field lifts the degeneracy of the $Y$ spin states and inhibits Pr transition involving single $Y$ spin flips [11]. This can be understood by considering the $Y$ quantization axis. At zero field the $Y$ quantization axis is locally defined by the

![FIG. 1: Two and three pulse echo sequences. $\pi$ pulse duration was 40μs and the laser was turned off 150μs before the pulse sequence and turned on 100μs before the echo.](image1)

![FIG. 2: Spectrum of hyperfine transitions showing experimentally measured points as the field is increased toward the critical point.](image2)
reducing to a slow decay for have a clearly biexponential decay: a fast initial decay, 2.5 ms no spectral diffusion was observed on the critical point (figure 4a). The other two transitions (figure 4a). The critical point transition. The biexponential decay is attributed to the existence of a “frozen core”. A frozen core is formed by the Pr ion detuning nearby Y ions from the bulk Y, thereby inhibiting mutual Y spin flips within the frozen core. A strong frozen core is expected due to the Pr magnetic moment ratio being an order of magnitude larger than that of Y. Although a frozen core has not been reported in Pr$^{3+}$:Y$_2$SiO$_5$, it has been observed in directly analogous systems [13]. On short time scales the rapid spin flips of the bulk Y ions dominate the dephasing. Since the bulk Y are well removed from the Pr ion they only weakly perturb the transition frequency. On longer time scales the frozen core Y spin flips dominate the dephasing as they induce larger but less frequent frequency shifts. Given that the slow spectral diffusion has not plateaued for the longest $\tau_2$ used, the correlation time of the frozen core is comparable or longer than 0.5s.

Further investigation of the three pulse echos on the critical point transition (figure 4) shows that for all values of $\tau_1$ only a single, slow exponential decay was observed. Therefore dephasing of the critical point transition dominated, at all time scales by the frozen core Y spin flips. The fact that the bulk Y dephasing contribution is ab-

\[ I(2t) = I_0 \exp \left[ -\left( \frac{2t}{T_M} \right)^2 \right] \]  

\[ (3) \]

To investigate the diffusion process over longer times scales a series of three pulse echo measurements were made. Within each of the series shown in figure 4, the delay between the first two pulses ($\tau_1$) is held constant while the delay between the second and third pulses ($\tau_2$) is varied.

A comparison is made with the same transitions as used in the two pulse echo study. Comparing echo sequences with the same $\tau_1$ on the different transitions shows a clear difference between the critical point and the other two transitions (figure 4b). The $m_I = +1/2 \leftrightarrow +3/2$ and the site 1b $m_I = +1/2 \leftrightarrow +3/2$ transitions have a clearly biexponential decay: a fast initial decay, reducing to a slow decay for $\tau_2 > 200 ms$. With a $\tau_1$ of 2.5ms no spectral diffusion was observed on the critical point transition.
sent for the site $1a$ $m_f = +1/2 \leftrightarrow +3/2$ transition and present for the other two transitions is consistent with the dephasing at the critical point being due to second order Zeeman interactions. This suggests that the applied field is accurate to the order of a Pr-Y interaction strength from the ideal critical point. Further optimisation of the applied field is therefore unlikely to produce a significant gain in coherence time.

Using the method described above a regime has reached where the correlation time of the dephasing interaction is extremely long compared to the duration of readily achievable RF pulses. This is exactly the regime required for effective implementation of quantum error correction schemes\[15\,16\,17\]. The simplest implementation shown to be sufficient for error correction in solid state qubits\[15\] consists of a series of hard rephasing pulses ($\pi$) separated by the cycling time. In the present case the difference between a transition specific hard RF $\pi$ pulse ($\geq 1 \mu s$) and the $T_M$ would allow an effective error correction scheme to be applied with a duty cycle $<0.1\%$.

We have shown that a phase memory time of 82ms can be achieved for $Pr^{3+}:Y_2SiO_5$ by using an external magnetic field to minimise the transition sensitivity to magnetic field fluctuations. Directions to further reduce the residual dephasing mechanism have been identified. We have shown that even in hosts containing nuclear spins it is possible to obtain spin based qubits that have coherence properties suitable for sophisticated quantum computing demonstrations.

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