Optical clock transition in a rare-earth-ion-doped crystal: coherence lifetime extension for quantum storage applications

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Abstract. Atomic clock transitions are desirable for quantum information storage and processing thanks to the protection from decoherence they provide. In the context of rare-earth-ion-doped crystals for quantum information storage, clock Zeeman or hyperfine transitions have been identified and exploited for long-lived storage in spin degrees of freedom. We present a theoretical and experimental analysis on the existence of an optical clock transition in Tm³⁺:YAG, in view of storage in optical coherences. The combination of a Zeeman-like term and a quadratic electronic Zeeman term in the Hamiltonian, lead to the existence of a magnetic field amplitude (12 mT) for which the derivative of the optical transition energy with respect to the field amplitude vanishes, regardless of the magnetic field orientation. We have verified this prediction through hole-burning spectroscopy experiments. In addition to that, a study of the behavior of the Hamiltonian as a function of the magnetic field orientation yields the direction for which both derivatives with respect to the magnetic field angular coordinates also vanish. The condition for an optical clock transition with three vanishing partial derivatives is met.

1. Introduction
In atom-based quantum memory systems, quantum information is stored in atomic superposition states, that is, in atomic coherences. Thus, the storage time is limited by the atomic coherence lifetime $T_2$. Long $T_2$ systems are desirable for such applications. Rare-earth-ion-doped crystals (REIDC) display satisfying $T_2$ values at low temperature ($< 3$ K). Typical $T_2$’s are of the order of some tens of microseconds for optical coherences [1], that is, a superposition state between two levels separated by energies in the optical electromagnetic spectrum. Therefore, REIDC have been intensively studied as candidates for quantum memories. Aside their long optical $T_2$, they constitute compact systems in which the storage centers are immobile within a solid matrix, simplifying their experimental manipulation. If longer $T_2$’s are desirable, REIDC offer the possibility of transferring the quantum information from optical coherences to Zeeman or hyperfine ones, of longer lifetime of the order of some hundreds of microseconds. This can be performed by applying a control pulse tuned to one arm of a Λ-system (two sub-levels of the ground level optically linked to an excited one) [2]. Of course, back-conversion is compulsory before launching the retrieval of the stored information.
Additional strategies to further extend $T_2$ have been demonstrated in REIDC. One of them, known as dynamical decoupling (DD), is based on the application of successive RF $\pi$ pulses all through the time the information is stored in the Zeeman (or hyperfine) coherences [3, 4]. If the pulses are applied at a rate faster than the correlation time of the magnetic environment fluctuations (magnetic moments of the lattice ions) responsible for the decoherence, the effect of the environment on the rare-earth ion coherence is canceled out. This way, Zeeman coherence extension of more than two orders of magnitude has been demonstrated in Pr$^{3+}$:Y$_2$SiO$_5$ [4], Tm$^{3+}$:YAG [5] and other REIDCs. Another strategy for $T_2$ extension is to work on an atomic clock transition [6, 7], that is, a transition whose frequency is insensitive (to first order) to fluctuations in the magnetic environment because of vanishing partial derivatives with respect to one or more magnetic field coordinates. In other words, if we note $\Delta_g$ as the Zeeman splitting of the ground level, and $B, \theta_B, \varphi_B$ the spherical coordinates of the magnetic field $B$, an atomic clock transition is met when there exists some $B_0$ such that $\partial \Delta_g/\partial B |_{B_0} = 0$ and/or $\partial \Delta_g/\partial \theta_B |_{B_0} = 0$ and/or $\partial \Delta_g/\partial \varphi_B |_{B_0} = 0$. That way, $\Delta_g$ is insensitive, to first order, to fluctuations of some or all of the magnetic field coordinates introduced by the environing magnetic moments. Therefore, decoherence is minimized. A clock transition with three vanishing partial derivatives will be henceforth called a full clock transition. Such a clock transition is also known, in the context of REIDC, as the ZEFOZ (zero first order Zeeman effect) technique [4, 8]. The combination of DD and ZEFOZ has yielded impressive results in Pr$^{3+}$:Y$_2$SiO$_5$, reaching the value of 40 s for the Zeeman $T_2$ [9].

The results of both techniques for $T_2$ extension just described have been obtained on Zeeman or hyperfine coherences. However, no demonstration has been presented in the case of optical coherences. DD is not well-suited for the optical case since a photon echo would be emitted after the first $\pi$ pulse, leading to immediate retrieval of the stored information (this is irrelevant in the Zeeman or hyperfine case since the radiated-energy-to-$h\omega$ ratio scales as $\omega^3$). On the contrary, it would be worth it to implement an optical clock transition, proposed in the context of REIDC initially in Ref. [10]. If long enough optical $T_2$’s were obtained that way, there would be no need to resort to storage in the Zeeman coherences, and the loss in efficiency that comes along would be avoided. Then, quantum storage protocols such as the atomic frequency comb (AFC) [11, 12] or the revival of silenced echo (ROSE) [13] could be implemented with higher efficiencies.

In the present communication, we investigate the viability of implementing a full optical clock transition in Tm$^{3+}$:YAG. Aside from its advantages in terms of diode-laser-compatible wavelength or large inhomogeneous broadening for broadband storage discussed elsewhere [14], such REIDC presents a rather long optical level lifetime $T_1$, typically 800 $\mu$s [15]. As $2T_1$ is the theoretical limit for $T_2$, Tm$^{3+}$:YAG is an interesting candidate for implementing an optical clock transition. In the following section we will develop a theoretical analysis for identifying an optical clock transition with three vanishing partial derivatives. In section 3, we will present results from hole-burning spectroscopy that validate our theoretical findings.

2. Theoretical analysis for identifying a full optical clock transition

Trivalent thulium ions substitute for trivalent yttrium ions in sites of $D_2$ symmetry of the YAG lattice. The facts that Tm$^{3+}$ is a non-Kramers ion (even number of electrons), has nuclear spin 1/2 and lies in a low symmetry site lead to an effective Hamiltonian of the form [16]

$$H_{\text{eff}} = -\hbar \vec{B} \cdot \vec{\gamma}(J) \cdot \vec{I} - \vec{B} \cdot \vec{\Lambda}(J) \cdot \vec{B},$$

where $\vec{B}$ is the magnetic field, $\vec{I}$ is the nuclear spin operator, and $\vec{\gamma}$ and $\vec{\Lambda}(J)$ are diagonal tensors whose elements are given by
\[
\Lambda_{\alpha\beta}^{(j)} = \mu_B g_J^2 \delta_{\alpha\beta} \sum_{n=1}^{2J+1} \frac{|\langle 0|J_n|n\rangle|^2}{E_n - E_0},
\]
(2)

\[
\gamma_{\alpha\beta}^{(J)} = \left( \gamma_n + \frac{2A_J \Lambda_{\alpha\beta}^{(j)}}{\hbar B g_J} \right) \delta_{\alpha\beta}.
\]
(3)

In the above expressions, \( \gamma_n \) is the nuclear gyromagnetic ratio, \( \mu_B \) is the Bohr magneton, \( g_J \) is the Landé factor, \( A_J \) is the hyperfine interaction parameter, \( J \) is the total angular momentum operator, \( \Lambda \) is the Landé factor, \( \gamma \) are the eigenstates in the \( J \) multiplet, \( E_n \) are their corresponding energies, and \( \delta_{\alpha\beta} \) is the Kronecker delta. The tensors in (2) and (3) are expressed in the coordinate system defined by the three orthogonal \( C_2 \) axes of the site symmetry group, \( D_2 \). The parameters \( \gamma_{xx} \), \( \gamma_{yy} \) and \( \gamma_{zz} \) in the effective Hamiltonian have already been measured for the ground \([^{3}\text{H}_0(0)]\) and excited \([^{3}\text{H}_4(0)]\) states. In what follows, we will use the values reported in Ref. [17]. From them, the values for \( \Lambda_{xx}, \Lambda_{yy}, \Lambda_{zz} \) can be deduced.

The eigenstates of \( H_{\text{eff}} \) are two sublevels for the ground state, that we will note \([1]\) and \([2]\), and two sub-levels for the excited state, \([3]\) and \([4]\). The first term in \( H_{\text{eff}} \) is a Zeeman-like one, that would lead to a linear increase of the ground and excited level splittings with growing magnetic field amplitude. We choose \([1]\) and \([3]\) \(([2]\) and \([4]\)) to be the sub-levels whose energies decrease (increase) with increasing magnetic field. The second term does not depend on \( B \), therefore, it rigidly shifts both sub-levels of the ground or excited states. The amount of that shift depends quadratically on the magnetic field amplitude, and its curvature differs for the ground and excited levels. The latter fact ensures that, regardless of the orientation of the magnetic field, there always exists a magnetic field amplitude \( B_0 \) for which \( \partial \Delta_{13}/\partial B \big|_{B_0} = 0 \), where \( \Delta_{13} \) is the energy difference between levels \([1]\) and \([3]\). Thus, one of the conditions for a full clock transition is met. This can be seen in Fig. 1(a), where we plot the calculated \( \Delta_{13} \) and \( \Delta_{24} \) off-set to their values at zero magnetic field as a function of the field amplitude (the field orientation relative to the crystal axes \( \{[100],[010],[001]\} \) has been chosen as \( \theta_B = 90^\circ \), \( \varphi_B = 45^\circ \) for reasons described later on). A vanishing derivative is found for \( \Delta_{13} \) at \( B_0 = 13 \) mT. As expected, no vanishing derivative is observed for \( \Delta_{24} \).

At this point of the discussion we must clarify that Y\(^{3+}\) ions, and thus Tm\(^{3+}\) ions, in a YAG lattice occupy six sites that are equivalent in terms of their local environment but distinct in terms of the orientation of this environment, i.e. the axes of the \( D_2 \) point group are differently oriented (see Ref. [18] and references therein). When an external field is applied, the equivalence between sites is broken. Therefore, curves such as the one plotted in Fig. 1(a) differ from site to site. In Fig. 1(a) we have computed \( \Delta_{13} \) and \( \Delta_{24} \) for the site usually labeled “1” in the literature [18], that is, the site whose \( C_2 \) axes are \( \{[01\bar{1}],[01\bar{1}],[100]\} \). Similar plots can be obtained for the remaining sites.

Still, the magnetic field orientation for a full clock transition has to be identified. In other words, we need to find values for the magnetic field angular coordinates for which the remaining two partial derivatives of \( \Delta_{13} \) vanish. For that, in Fig. 2 we plot \( \Delta_{13}(B = 13 \) mT) as a function of \( \theta_B \) and \( \varphi_B \), which are spherical coordinates with respect to the crystal axes \( \{[100],[010],[001]\} \).

Here again, we consider site 1. We observe that both derivatives vanish for \( \theta_B = 90^\circ \), \( \varphi_B = 45^\circ \) (this orientation is indicated in Fig. 2 as the intersection between the two lines). Hence, a full clock transition exists for site 1 and is given in spherical coordinates by \( B_0 = (13 \) mT, \( 90^\circ, 45^\circ \)).

For that particular magnetic field orientation, it happens that sites 3, 4, 5 and 6 remain equivalent (it can be easily seen that the projection of \( B \) onto each of the three orthogonal \( C_2 \) axes of one site is the same for all sites 3 to 6.) This means that the Zeeman-like sub-level splittings are the same for sites 3-6, both in the ground and excited levels, which results in equal \( \Delta_{13} \) for all sites 3-6, as well as equal \( \Delta_{24} \). Of course, these values differ from those of site 1.
Figure 1. (a) Calculated energy difference between ground and excited sub-levels $|1⟩$ and $|3⟩$ (full line), and $|2⟩$ and $|4⟩$ (dashed line) of site 1. An offset corresponding to the values at zero magnetic field has been subtracted. (b) Hole-burning spectral position of the hole burnt in site 1’s level $|1⟩$ as a function of the magnetic field amplitude. The line is a quadratic fit.

Figure 2. Calculated energy difference between ground and excited sub-levels $|1⟩$ and $|3⟩$ as a function of the magnetic field spherical angular coordinates $\theta_B$ and $\varphi_B$ defined with respect to the Cartesian frame $\{[100],[010],[001]\}$. The magnetic field amplitude has been fixed to 13 mT. An offset corresponding to the transition frequency at zero magnetic field has been subtracted.

regards site 2, it does not participate at all in the experiments because the electric field vector ([110]) is perpendicular to the $^{3}H_{6}(0) \rightarrow ^{3}H_{4}(0)$ transition dipole moment ([1 ¯10]).

3. Hole-burning spectroscopy results

We use the spectral hole burning (SHB) technique to measure the optical transition frequency variation with the external magnetic field amplitude $B$. After burning a hole at $B = B_i = 19$ mT, we probe the hole at $B = B_f$. We have enough time to vary the field amplitude from $B_i$ to $B_f$ as the holes are burnt into long lifetime (~ 1 min) ground state sub-levels. Repeating the operation with different $B_f$ values, and the same initial $B_i$, we are able to follow the dependence
of the hole spectral position with $B$. The $B_f$ values range from 7 to 27 mT, by steps of 0.95 mT. As the hole is probed through the optical transitions $|1\rangle \rightarrow |3\rangle$ and $|2\rangle \rightarrow |4\rangle$, this gives us access to the dependence of $\Delta_{13}$ and $\Delta_{24}$ with $B$. The experiment is performed with $B$ orientation fixed at $\theta_B = (90 \pm 5)^\circ$, $\varphi_B = (45 \pm 5)^\circ$.

Figure 3. Hole burning spectra. In each curve the hole is burnt at $B = B_i = 19$ mT and the spectrum is read at $B = B_f$. $B_f$ is set to values ranging from 7 mT to 29 mT by steps of 0.95 mT (the last step is twice as large). The horizontal axis indicates the spectral detuning from the position the hole takes when $B_f = B_i = 19$ mT. Four different holes are identified with guides to the eye. They correspond to: site 1, hole burnt in $|1\rangle$, read through transition $|1\rangle \rightarrow |3\rangle$ (--); site 1, hole burnt in $|2\rangle$, read through transition $|2\rangle \rightarrow |4\rangle$ (---); sites 3-6, hole burnt in $|1\rangle$, read through transition $|1\rangle \rightarrow |3\rangle$ (· · · · · ·); sites 3-6, hole burnt in $|2\rangle$, read through transition $|2\rangle \rightarrow |4\rangle$ (— · —).

The results are shown in Fig. 3. When $B_f = B_i = 19$ mT, a single hole is observed (a single peak). As $B_f$ is shifted away from $B_i$ we see that the initial hole splits in four holes that display different dependences with $B_f$. These dependences are indicated with the four guides to the eye. After some analysis of the Hamiltonian in (1), we are able to assign the four holes to:

- Site 1, hole burnt in $|1\rangle$, read through transition $|1\rangle \rightarrow |3\rangle$ (—).
- Site 1, hole burnt in $|2\rangle$, read through transition $|2\rangle \rightarrow |4\rangle$ (— —).
- Sites 3-6, hole burnt in $|1\rangle$, read through transition $|1\rangle \rightarrow |3\rangle$ (· · · · · ·).
- Sites 3-6, hole burnt in $|2\rangle$, read through transition $|2\rangle \rightarrow |4\rangle$ (— · —).

(Crossed transitions $|1\rangle \rightarrow |4\rangle$ and $|2\rangle \rightarrow |3\rangle$ are negligibly weak). As the optical transition inhomogeneous broadening ($\sim 20$ GHz) is much larger than the sub-level splitting ($\sim 1$ MHz), for some Tm$^{3+}$ ions, the optical pump energy is resonant to $\Delta_{13}$, while for others, it is resonant to $\Delta_{24}$. In the former case, the hole is burnt in sub-level $|1\rangle$; in the latter, the hole is burnt in $|2\rangle$. As this happens for site 1 and for sites 3-6, four different holes are burnt. The full line in Fig. 3 shows the dependence of site 1’s $\Delta_{13}$, the transition expected to be a full clock transition. In fact, a vanishing derivative is observed around $B_f = 12$ mT (seventh curve from the bottom).
The spectral positions of the holes burnt in |1⟩ for site 1 are plotted in Fig. 1(b) as a function of $B_f$. We observe that the behavior is perfectly quadratic, as expected, and displays a vanishing derivative at $B_f = 12$ mT. The linear (quadratic) term coefficient of the fit in Fig. 1(b) differs from the predicted one in 6% (15%). Our experimental setup does not allow fast variation of the magnetic field orientation. Therefore, an analog study as a function of the magnetic field angular coordinates was not possible. However, the good agreement of the experimental data to the predicted behavior suggests that a full optical clock transition should be possible in site 1 of Tm$^{3+}$:YAG for extending the optical $T_2$.

Our search for a clock transition has been limited to site 1. Although not shown here, it can be demonstrated that sites 3-6 display very small angular derivatives at the field orientation found for site 1, and that the derivative with respect to the amplitude vanishes for a somewhat higher field ($\sim 20$ mT). The latter statement can be verified by observing the dotted line in Fig. 3. Therefore, even if not on a proper clock transition, $T_2$ will also be extended for sites 3-6.

For no site, clock transition conditions are met in the case of transition $|2⟩ \rightarrow |4⟩$. In a quantum information storage scheme, one would wish to avoid storage in that transition, since decoherence is not prevented. A simple preparation stage can be implemented prior to the storage, where sub-level $|2⟩$ would be emptied through optical pumping.

4. Summary
We have performed a theoretical and experimental study of the viability of gaining profit from a full clock transition to extend the optical $T_2$ in a REIDC system compatible with quantum information storage. From the analysis of the Hamiltonian we have identified the magnetic field coordinates for a full optical clock transition. Experimentally, we have verified that the theoretical prediction for the amplitude coordinate is in good agreement with hole-burning spectroscopy results.

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