High Resolution Optical Spectroscopy and Magnetic Properties of Yb$^{3+}$ in Y$_2$SiO$_5$

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Rare earth doped crystals are promising systems for quantum information processing. In particular paramagnetic rare earths could be used to build coherent interfaces with optical and microwave photons. In addition, isotopes with non zero nuclear spins could provide long lived states for quantum state storage and processing. Yb$^{3+}$ is particularly interesting in this respect since it is the only paramagnetic rare earth with a spin 1/2 isotope, which corresponds to the simplest possible level structure. In this paper, we report on the optical and magnetic properties of Yb$^{3+}$ in the two sites of Y$_2$SiO$_5$, a commonly used crystal for quantum applications. We measured optical inhomogeneous linewidths, peak absorption coefficients, oscillator strengths, excited state lifetimes and fluorescence branching ratios. The Zeeman tensors were also determined in the ground and excited states, as well as the ground state hyperfine tensor for the $^{171}$Yb$^{3+}$ (I = 1/2) isotope. These results suggest that Yb$^{3+}$:Y$_2$SiO$_5$ is a promising material for applications like solid state optical and microwave quantum memories.

I. INTRODUCTION

Rare-earth (RE) doped crystals are promising solid state candidates for quantum information processing. In particular, they can show extremely narrow optical homogeneous linewidths at low temperature, in the range of a few kHz to less than 100 Hz. Combined with inhomogeneous linewidths in the GHz range, this allows optical addressing of ions within ensemble for quantum processors, or quantum memories with large time-bandwidth products. Moreover, several RE ions have non-zero nuclear spins, which can act as long-lived, optically addressable qubits. As an example, hyperfine transitions of Eu$^{3+}$:Y$_2$SiO$_5$ can show coherence lifetimes ($T_2$) up to 6 hours at 2 K using various dephasing control techniques, and still reach a few ms at 20 K. Recent results include entanglement storage and light-matter teleportation at the telecom wavelength, single photon level memories with storage in nuclear spin states as well as memories with high efficiency and storage time exceeding one minute. Large and switchable interactions between RE ions have also been observed as well as single RE detection, which opens the way to quantum processing in these systems.

Paramagnetic RE ions, such as Nd$^{3+}$ or Er$^{3+}$, have one more degree of freedom due to their electron spins. This can be used to interface microwave photons to a RE doped crystal through a superconducting resonator and obtain quantum memories for superconducting qubits. In this case too, nuclear hyperfine transitions can provide long storage time. We recently showed that $^{145}$Nd$^{3+}$:Y$_2$SiO$_5$ hyperfine transitions have ground state coherence lifetimes up to 9 ms at 5 K, whereas the electron spin $T_2$ is about 100 µs. High fidelity coherent transfer of microwave excitations to nuclear spins was also demonstrated by quantum state tomography. Ultimately, it should be possible to build coherent interfaces between optical and/or microwave photons qubits, and long lived nuclear spin quantum states.

Yb$^{3+}$ ions have attractive properties in this respect. Their $4f^{13}$ configuration comprises only two multiplets: $^2F_{5/2}$ (ground) and $^2F_{7/2}$ (excited) separated by $\approx 10000$ cm$^{-1}$. This energy corresponds to the near infrared, where laser diodes are easily found. The excited state decays usually radiatively, with a lifetime of about 1 ms, which sets a limit for the optical coherence lifetime of 2 ms. There are also two naturally abundant isotopes with non-zero nuclear spin: $^{171}$Yb$^{3+}$ ($I = 1/2$) and $^{173}$Yb$^{3+}$ ($I = 5/2$), which hyperfine transitions could be used as long lived qubits. The lower spin angular momenta of Yb$^{3+}$ compared to Er$^{3+}$ and Nd$^{3+}$ ($I = 7/2$ for all non-zero spin isotopes) result in simpler energy level structures. This is important since optical initialization and coherent manipulation of spins require selecting specific transitions within the optical inhomogeneous linewidth by potentially complex optical pumping sequences. The lower number of spin states in $^{171}$Yb$^{3+}$ and $^{173}$Yb$^{3+}$ could simplify them considerably.

Here we report on the low temperature and high resolution optical spectroscopy of Yb$^{3+}$:Y$_2$SiO$_5$. This crystalline host has been chosen since it shows outstanding properties in terms of narrow optical linewidths and long coherence spin lifetimes when doped with Pr$^{3+}$, Nd$^{3+}$,Er$^{3+}$ and Er$^{3+}$ It is currently the most used host in quantum storage experiments. We measured inhomogeneous linewidths, absorption spectra, excited state lifetimes and branching ratios for the two Yb$^{3+}$ sites and found values among the best for RE doped crystals. Magnetic fields are often used to slow down electron spin relaxation, which influences optical and spin coherence lifetimes and spectral hole burning efficiency. As a first step towards these dynamical experiments, we determined the ground and excited state g tensors from electron paramagnetic resonance and optical spec-
II. EXPERIMENTAL

We used Y$_2$SiO$_5$ (YSO) samples doped at 0.005 at.% (50 ppm) with Yb$^{3+}$ and cut from a boule grown by the Czochralski method. YSO has a monoclinic structure belonging to the $C_{2h}$ ($C_{2}/c$) space group. Yb$^{3+}$ can substitute Y$^{3+}$ in two different crystallographic sites, both with a C$_1$ point symmetry. In addition, non equivalent subsites appear under magnetic fields neither parallel nor perpendicular to b. They are related by the crystal C$_2$ (b) symmetry axis. 

Samples were cut along the three principal dielectric axes: b (the C$_2$ crystallographic axis), D$_1$ and D$_2$. Ytterbium has five stable even isotopes $^{168}$Yb, $^{170}$Yb, $^{172}$Yb, $^{174}$Yb and $^{176}$Yb with nuclear spin I=0 and a total abundance of 69.59%. There are also two odd isotopes, $^{171}$Yb, with I=1/2 and an abundance of 14.28% and $^{173}$Yb with I=5/2 and a abundance of 16.13%. Absorption spectra with 0.1 nm resolution were obtained with a Varian Cary 6000i spectrophotometer. Fluorescence measurements were performed using a Coherent 829 Titanum Sapphire laser pumped by a Coherent Verdi G10 laser, a SpectraPro 750 monochromator (1 nm resolution) and an InGaAs photodiode. Fluorescence lifetime measurements were possible using a tunable optical parametric oscillator pumped by a Nd:YAG laser (Ekspla NT342BSH, 6 ns pulse length) as the excitation source, a Jobin-Yvon HR250 monochromator and an InGaAs photodiode. High resolution transmission spectra were recorded by scanning a single mode Toptica DL 100 diode laser (1 MHz linewidth) around 980 nm. Continuous frequency scans of about 15 GHz could be performed. The laser beam was collimated with a power of about 1mW in front of the cryostat. The transmitted signal was detected by a Thorlabs PDA36A photodiode and a reference beam by a Thorlabs PM10A photodiode. A small part of signal was also sent to a Toptica Fabry-Perot Interferometer (1 GHz Free Spectral Range at 980 nm). This allowed us to precisely calibrate the spectra frequency scale. The sample was maintained at 10 K in a CTI-Cryogenics closed-cycle cryostat.

Electron paramagnetic resonance spectra were recorded at 9 K with a Bruker ELEXSYS E500 and an ELEXYS Super High Sensitivity Probe Head in X-band. For the optical determination of Yb$^{3+}$ excited state g-tensors, the crystal was placed between two permanent NdFeB magnets. The field average value was 217 mT with an inhomogeneity of about 10 % along the laser propagation axis. To record angular variations, the crystal sat on a pedestal attached to an Attocube ANRv51 stage and was rotated by steps of 10°. The whole assembly was put in a Janis LHe cryostat at 10 K. The transmission spectra were recorded with the set up described above.

III. RESULTS AND DISCUSSION

A. Optical Spectroscopy

1. Absorption and Emission Spectra

We first recorded absorption and emission spectra to determine Yb$^{3+}$ crystal field (CF) level energies. The absorption spectrum (Fig. 1) shows well resolved lines corresponding to transitions from the lowest CF level $^{2}F_{7/2}(0)$ of the ground multiplet to the three CF levels $^{2}F_{5/2}(0,1,2)$ of the excited one. These lines are homogeneously broadened except for the two lowest energy ones which are likely to be inhomogeneously broadened. A narrow and isolated peak, with a full width at half maximum (FWHM) of 2.8 cm$^{-1}$, is recorded at 10565 cm$^{-1}$ and corresponds to the $^{2}F_{7/2}(0)$--$^{2}F_{5/2}(1)$ transition for site 2. Other transitions above 10300 cm$^{-1}$ are much broader with FWHM between 15 and 25 cm$^{-1}$. The structures in the range 10450 -10650 cm$^{-1}$ observed for Yb$^{3+}$ concentrations of a few % [30] are not seen in this 50 ppm doped sample and are therefore attributed to distorted Yb$^{3+}$ sites.

Low doping concentration also prevents energy transfer between sites and allows separate recording of each site’s emission spectrum (Fig. 2). These lines correspond to the $^{2}F_{5/2}(0)$--$^{2}F_{7/2}(0,1,2,3)$ transitions and, except for the higher energy lines, whose widths are instrument limited, are much broader than those observed in absorption. In addition, many partially resolved lines, that we attribute to vibronic transitions, appear in the range 9760-10060 cm$^{-1}$ for site 2. This induces an uncertainty in the $^{2}F_{7/2}(2)$ level position, which we determined using the strongest peak at 9982 cm$^{-1}$.

The crystal field levels for both sites are summarized in Table I and are in good agreement with previous studies. CF splittings for the ground and excited multiplets are significantly smaller for site 1, which suggests that it corresponds to the crystallographic site with a coordination number (CN) of 7, in which Y-O distances are larger. Indeed, scalar CF strengths have been found smaller for this site in Ce$^{3+}$:Y$_2$SiO$_5$ in a theoretical study. In the same way, Pr$^{3+}$ ions with the smaller CF splittings have also a higher absorption coefficient, suggesting a higher relative concentration. Since Pr$^{3+}$ ions have a larger ionic radius than Y$^{3+}$ ones, they should occupy preferentially the CN = 7 site, which has a larger volume.

Because of phonon relaxation between CF levels within the multiplets, only the 0-0 ($^{2}F_{7/2}(0)$--$^{2}F_{5/2}(0)$) transition
was parallel to and the wave vector $k$ was parallel to $D_2$ (Fig. 3). The lines peak at 10188.87 cm$^{-1}$ (981.463 nm in vac.) and 10216.06 cm$^{-1}$ (978.854 nm in vac.) for sites 1 and 2.

Both lines show a narrow central part, as expected for the $I = 0$ Yb$^{3+}$ isotopes, on top of a broader and weaker structure, in which peaks can be clearly seen in some cases (e.g., site 2, $E \parallel D_2$). We attribute this additional feature to the zero field hyperfine structures of the $^{171}$Yb$^{3+}$ (abundance 14.3%) and $^{173}$Yb$^{3+}$ (abundance 16.1%) isotopes, which span about 3-4 GHz in the ground state (see Section III B). All lines could be well fitted by a combination of Lorentzian lines (see inset in Fig. 3). This has been observed in several RE doped crystals and, according to Stoneham, corresponds to perturbations by a low concentration of point defects. The FWHM of the central parts of the lines are 2.2 and 1.7 GHz for sites 1 and 2. These values are comparable to those found for Y$_2$SiO$_5$ doped at low levels of RE ions and are related to the difference in ionic radius between Y$^{3+}$ (0.892 Å) and Yb$^{3+}$ (0.858 Å). It is important to note that growth conditions can also have a significant influence on $\Gamma_{inh}$.

Large and anisotropic peak absorption coefficients were measured, reaching maximum values of 6.5 cm$^{-1}$ ($E \parallel b$) and 10.3 cm$^{-1}$ ($E \parallel D_2$) for site 1 and 2 respectively. In both cases, the lower absorption occurs for a light electric field polarized along $D_1$. The average oscillator strengths $P$ were calculated without local field corrections and assuming equal Yb$^{3+}$ occupancy for the two sites, because of the close ionic radii of Y$^{3+}$ and Yb$^{3+}$. Site 2 value ($P = 6.4 \times 10^{-7}$) is about 30% higher than the one for site 1 ($P = 5.0 \times 10^{-7}$).

Excited state population lifetimes are around 1 ms for both sites (Table I) and can be considered to be purely radiative. On one hand the low doping concentration prevents energy transfers between Yb$^{3+}$ ions and to quenching centers. On the other hand, the energy gap between the $^2$F$_{7/2}$ and $^2$F$_{5/2}$ multiplets ($\approx 10000$ cm$^{-1}$) is much larger than the Y$_2$SiO$_5$ phonon cut-off frequency ($\approx 960$ cm$^{-1}$) so that multiphonon relaxation is negligible.

### Table I. Optical transitions in Yb$^{3+}$:Y$_2$SiO$_5$. CF level energies; 0-0 transition wavelength (vac.), inhomogeneous linewidths, peak absorption coefficient for different light polarizations, oscillator strength; experimental fluorescence decay time ($T_1$), spontaneous emission decay time for the 0-0 transition ($T_{1s}$) and $T_{1s}/T_1$ ratio.

| Energy (cm$^{-1}$) | $\lambda_{vac}$ (nm) | $\Gamma_{inh}$ (GHz) | $\alpha_0$ (cm$^{-1}$) | $P \times 10^7$ | $T_1$ (ms) | $T_{1s}$ (ms) | $T_{1s}/T_1$ |
|-------------------|-----------------------|-----------------------|------------------------|----------------|-----------|-------------|-------------|
| $^2$F$_{7/2}$     | $^2$F$_{5/2}$         | Site 1                | Site 2                 |
| 0                 | 10189                 | 111                   | 10391                  | 499            | 709       | 0           | 1           |
| $^0$              | 981.463               | 981.463               | 981.463                | 981.463        |           |             |             |
| 2                 | 2.2                   | 5.7                   | 6.5                    |                |           |             |             |
| 3                 | 3.3 ($E \parallel D_1$) | 5.0 ($E \parallel D_2$) | 6.5 ($E \parallel b$) |                |           |             |             |

FIG. 1. Unpolarized absorption spectrum of Yb$^{3+}$:Y$_2$SiO$_5$ at 12 K with light propagating along the $b$ axis. Transitions between CF levels are indicated for sites 1 and 2.

FIG. 2. Emission spectra of sites 1 (purple line) and 2 (orange line) of Yb$^{3+}$:Y$_2$SiO$_5$ at 10 K excited respectively at 919 nm and 902 nm. Transitions between CF levels are indicated.
of the ground multiplet of Yb$^{3+}$. Combined with a relatively short and purely radiative $T_1$, the reduced number of possible transitions favors a strong 0-0 transition. Still, some RE-host combinations allow larger oscillator strengths, like Nd$^{3+}$:YVO$_4$ ($8 \times 10^{-6}$) or Pr$^{3+}$:Y$_3$Al$_5$O$_{12}$ ($1.5 \times 10^{-6}$).

$T_{1s}$ is also short, similar to what is found in Pr$^{3+}$:YSO (5.7 ms) and much shorter than for Er$^{3+}$ (54.6 ms) or Eu$^{3+}$ (120 ns) doped YSO. Compared to systems with very high oscillator strengths, such as Nd$^{3+}$:YVO$_4$, $T_{1s}$ is however about 13 times longer. Finally, $T_{1s}/T_1$ is very close to the lowest value observed for YSO, 4.8 in Er$^{3+}$:YSO and about one order of magnitude better than for Pr$^{3+}$ (34.5) and Eu$^{3+}$ (63.2). Only Tm$^{3+}$:LiNbO$_3$ shows a significantly lower ratio (2.25). In summary, the 0-0 transition of Yb$^{3+}$ ions in site 2 of YSO shows among the best properties for RE doped crystals in terms of oscillator strength, spontaneous decay $T_{1s}$, and $T_{1s}/T_1$ ratio.

B. Magnetic Properties

We first determined Yb$^{3+}$ ground state Zeeman and hyperfine tensors by EPR, complementing previous studies where only particular orientations of the magnetic field were investigated. Due to the two crystallographic sites, which generally divide in two sub-sites under a magnetic field, and the two Yb$^{3+}$ isotopes with $I \neq 0$, many lines were observed for magnetic fields $B$ in the range 50-1000 mT. Fig. 4 shows site 1 lines for $B$ at 160$^\circ$ from $D_1$ in $D_1D_2$ plane, a configuration where the two subsites, related by a $C_2$ symmetry along $b$, are equivalent. Apart from the Zeeman line at 103.5 mT, the two transitions corresponding to $^{171}$Yb$^{3+}$ ($I = 1/2$) are observed at 76.1 and 131.2 mT. The other lines located between 64.5 and 144.6 mT are attributed to the hyperfine structure of $^{173}$Yb$^{3+}$ ($I = 5/2$). In this case, due to quadrupole interactions, some transitions with $\Delta M_I \neq 0$ are clearly seen. For comparison, the positions of lines corresponding to $\Delta M_I = 0$ and deduced from $^{171}$Yb$^{3+}$ hyperfine tensor (see below) are indicated. The lines were very narrow at low magnetic field and FWHMs as low as 12 MHz could be recorded, which is favorable to coupling with high quality factor microwave resonators.

The $g$ tensors of the $I = 0$ isotopes were determined for both sites from angular variations of the Zeeman lines in the three perpendicular planes $bD_1$, $bD_2$ and $D_1D_2$ (Figs. 4 and S1). All line positions were simultaneously fitted to the Hamiltonian $H = \mu_B \mathbf{B} \cdot \mathbf{g} \cdot \mathbf{S}$, where $\mu_B$ is the Bohr magneton and $\mathbf{S}$ is an effective 1/2 spin operator. The $C_2$ symmetry linking the sub-sites was also taken into account. The effective spin approach is possible because of the large ground state CF splittings ($100 - 200$ cm$^{-1}$) compared to the Zeeman one ($0.3$ cm$^{-1}$). Moreover, at low temperature, only the lowest energy CF doublet is populated. For each plane, misalignment between the crystal and the lab frame was also allowed.
for by introducing three variable angles in the fit. Excellent agreement with experimental data was obtained, as shown in Figs. 4 and S1. The principal values of $g$ and the Euler angles for the principal axes are gathered in Table III.

Fig. 4 shows the calculated effective $g$ factors for magnetic fields oriented in the $bD_1$, $bD_2$ and $D_1D_2$ planes. For site 1, the largest principal value is 6.53, with the principal axis oriented close to the $D_1$ axis. For site 2, the largest principal value is similar, 6.06, but corresponds to an orientation close to the $b$ axis. This explains the main features of the angular variations of Fig. 5. For both sites, the two other principal values are much smaller. This results in low $g$ factors in the $bD_2$ and $D_1D_2$ planes for sites 1 and 2 respectively.

As mentioned above, EPR lines were fitted allowing for crystal misalignment in each plane. For site 1, misalignment angles were below 2°. Surprisingly, these angles were much larger in the case of site 2, reaching for example 7° for the $D_1D_2$ variation. Moreover, fitting site 1 transitions with site 2 angles resulted in poor agreement with experimental data. This rules out a higher sensitivity of site 2 transitions to crystal misalignment. This discrepancy is particularly clear in the $D_1D_2$ angular variation (Fig. S1). For magnetic fields around 412 mT (but for different crystal orientations), subsite lines are separated by only 1.2 mT for site 1, whereas site 2 lines are separated by 21.3 mT. This result suggests that the distortion introduced by Yb$^{3+}$ substituting Y$^{3+}$ in site 2 results in an effective $C_2$ axis direction significantly different from the host crystal. This observation should be further investigated, as it is of importance for predicting particular energy level structures, such as Zero First Order Zeeman shift (ZEOZ) lines.

The $g$ tensor of the $^2F_{5/2}(0)$ excited state for the $I = 0$ isotopes was determined using optical measurements, by recording transmission spectra under a magnetic field of about 200 mT. The low 0-0 inhomogeneous linewidth allowed observing four partially resolved transitions in most orientations for both sites. Fig. 6(a) shows a transmission spectrum for site 1 with $B \parallel D_2$. In this case, the ground state Zeeman splitting is larger than the excited one and the energy difference corresponding to the effective $g$ factors of the ground and excited states are indicated in Figure 6(a). Effective $g$ factors were determined in the three $D_1D_2$, $bD_1$ and $bD_2$ planes by rotating the crystal with respect to the magnetic field (Fig. S2 and S3). The values for $B \parallel b$ are in agreement with those reported in [31]. In a first step, the ground state $g$ tensor measured by EPR was compared to the experimental data to check for sample misalignment. The corresponding angles were then used to fit the excited state Zeeman splittings to the spin Hamiltonian in the same way as for the ground state. The calculated effective excited state $g$ factors are shown in Fig. 6(b) and (c).

Table III gives the principal values and axes directions of the excited state $g$ tensors for both sites. The largest values are close, 3.4 and 3.3 for sites 1 and 2, and lower than those of the ground state by a factor of about 2.
This can be qualitatively understood by considering the expression of the $g_{zz}$ component for a pure $M_J$ crystal field level: $g_{zz} = 2g_JM_J$, where $g_J$ is the Landé factor. With $M_J = 7/2$ and $5/2$, we find $g_{zz} = 8$ and $4.3$, which reproduces approximately the experimental ratio of 2. The principal axes corresponding to the largest principal $g$ values are oriented close to the $D_1$ and $b$ axes for sites 1 and 2. These orientations are close to those obtained for the ground state, which explains that the angular variations and effective $g$ factors in the $D_1D_2$, $bD_1$ and $bD_2$ planes look similar (Figs. 6, 7(b,c), S2 and S3).

It was shown in Er$^{3+}$:YSO that a long optical coherence lifetime could be obtained with strong fields applied in direction where large effective $g$ factors for the ground and excited states are observed in both sites. At low temperatures, the upper Zeeman levels are strongly depopulated in this configuration, and Er$^{3+}$ spin flips are suppressed. Dephasing of the optical transitions between the lowest Zeeman levels are thus strongly decreased too. In Er$^{3+}$:YSO, this could be obtained with a field in the $D_1D_2$ plane, for which subsites are equivalent. From Figs. 6 and 7 it is clear that a similar strategy in Yb$^{3+}$:YSO requires a magnetic field in the $bD_1$ plane, which results in magnetically non equivalent subsites and, for example, a reduced optical density. In the $D_1D_2$ plane or along the $b$ axis, where subsites are magnetically equivalent, one site always has a low effective $g$ factor. Additional experiments are however needed to fully explore this question, as spin dephasing mechanisms may have complex dependences with respect to RE concentration, temperature and magnetic field magnitude and orientation.

We finally determined the ground state hyperfine interaction tensors $A$ for $^{171}$Yb$^{3+}$ ($I = 1/2$) from EPR experiments. The angular variations of the corresponding EPR lines in the $D_1D_2$, $bD_1$ and $bD_2$ planes were fitted to the Hamiltonian:

$$H = \mu_B \mathbf{B} \cdot \mathbf{g} \cdot \mathbf{S} + \mathbf{I} \cdot \mathbf{A} \cdot \mathbf{S},$$

(2)

using the $\mathbf{g}$ tensors and misalignment angles previously determined from the transitions of the $I = 0$ isotopes. Fig. 8(a) shows the angular variation of the two transitions between hyperfine levels in the $bD_1$ plane for the two subsites of site 2. Experimental data are very well reproduced by the fit, as in the case of the other angular variations (Figs. S4). The principal values and principal axes of $\mathbf{A}$ are given in Table III for sites 1 and 2.

For both sites, the ratio between the tensor elements $A_{ij}/g_{ij}$ is nearly constant for the large $A_{ij}$ and $g_{ij}$ values ($A_{ij}/g_{ij} \approx -0.0264$ for site 1, $A_{ij}/g_{ij} \approx -0.0259$ for site 2), as expected for a pure $J$ multiplet. Indeed, $A_{ij}/g_{ij} = A_J/g_J$ where $A_J$ does not depend on $M_J$. As a result, the $\mathbf{g}$ and $\mathbf{A}$ tensors of both sites have nearly the same orientation as $J$ mixing by the crystal field should be very small given the large separation between the $^2F_{7/2}$ and $^2F_{5/2}$ multiplets ($10000 \text{ cm}^{-1}$) compared to the multiplet splittings ($< 1000 \text{ cm}^{-1}$). The larger deviation between the $g$ and $A$ principal axes orientations for site 2 could be due to an enhanced $J$ mixing effect, consistent with the stronger crystal field of the CN = 6 environment (see Section III A).

The large hyperfine interaction results in zero field splittings of 3.7 and 3.1 GHz for sites 1 and 2, which can be tuned by several GHz using small magnetic fields oriented along the large effective $g$ factors (Fig. 9 for site 1 and S7 for site 2). Moreover, ZEFOZ transitions could appear when the Zeeman and hyperfine interactions start to be comparable. These transitions, also known as clock transitions, are insensitive to magnetic field fluctuations, which can increase their coherence lifetime. A partial ZEFOZ transition is observed around 48 mT in site 1 for a magnetic field oriented along $D_2$ in Fig. 9. This could be useful for coupling Yb$^{3+}$ ions to superconducting resonators and obtaining quantum memories for microwave photons with long storage time.

We also deduced the hyperfine tensor for $^{173}$Yb$^{3+}$ by scaling $^{171}$Yb$^{3+}$ $\mathbf{A}$ tensor by the ratio (-0.27) of the nuclear gyromagnetic factors. This accounted for the observed angular variations of $^{173}$Yb$^{3+}$ strongest EPR transitions (Fig. 8(b), S5 and S6), although the quadrupole
TABLE II. Magnetic properties of Yb$^{3+}$:Y$_2$SiO$_5$. Principal values of the $g$ and $A$ ($^{171}\text{Yb}^{3+}$, in MHz) tensors and Euler angles (in degree) defining the principal axis orientations (zxz convention).

| Site 1                  | $g_x$ | $g_y$ | $g_z$ | $\alpha$ | $\beta$ | $\gamma$ | $A_x$ | $A_y$ | $A_z$ | $\alpha_A$ | $\beta_A$ | $\gamma_A$ |
|-------------------------|-------|-------|-------|-----------|---------|----------|-------|-------|-------|------------|------------|------------|
| Ground state            | -0.31 | -1.60 | 6.53  | 252.8     | 88.7    | 113.8    | 0     | -2140 | -5302 | 247        | 67         | 122        |
| Excited state           | -0.8  | 1.0   | 3.4   | 77        | 84      | 173      |       |        |        |            |            |            |
| Site 2                  | -0.13 | -1.50 | 6.06  | 59.1      | 11.8    | 347.4    | 2     | 1490  | -4760 | 51         | 11         | 12         |
| Ground state            | -1.0  | 1.4   | -3.3  | 234       | 157     | 190      |       |        |        |            |            |            |
| Excited state           |       |       |       |           |         |          |       |        |        |            |            |            |

FIG. 9. Calculated energies $E$ of the ground state hyperfine levels of $^{171}\text{Yb}^{3+}$ ($I = 1/2$) in site 1 (red lines) as a function of the magnetic field strength. The field is oriented along $D_2$. The vertical dashed line denotes a partial ZEFOZ transition ($dE/dB = 0$). The energies of the Zeeman levels of the $I = 0$ isotopes are given for comparison (blue lines).

IV. CONCLUSION

Optical properties of a 50 ppm doped Yb$^{3+}$:Y$_2$SiO$_5$ crystal have been studied in the context of applications in quantum information processing. In particular, a detailed study of the transition between the lowest crystal field levels of the $^2F_{7/2}$ and $^2F_{5/2}$ multiplets has been carried out at low temperature, allowing the measurements of inhomogeneous broadenings $\Gamma_h$, peak absorption coefficients of polarized light, oscillator strengths $P$, and excited state lifetimes $T_1$.

For Yb$^{3+}$ ions in site 2, we found of $P = 6.4 \times 10^{-7}$, one of the largest value observed for a rare earth ion in Y$_2$SiO$_5$, while $\Gamma_{inh} = 1.7$ GHz. A relatively strong branching ratio was also found for the 0-0 transition, leading to a relaxation rate of $T_{1s} = 6.5$ ms and a ratio $T_{1s}/T_1 = 5.0$. These values also compare favorably with those obtained for other RE doped materials.

We also determined the ground and excited state Zeeman tensors of the $I = 0$ isotopes from angular variations obtained respectively with EPR and optical transmission under a magnetic field of about 200 mT. The largest principal values for both sites are close to $g = 6$ for the ground state and $g = 3$ for the excited state. The corresponding principal axes are close for the ground and excited states and oriented respectively along $D_1$ and $b$ for sites 1 and 2.

Yb$^{3+}$ is the only paramagnetic rare earth ion with a $I = 1/2$ isotope, which could be advantageous for optical addressing of spin transitions. We therefore determined the ground state $A$ tensor for the $^{171}\text{Yb}^{3+}$ ($I = 1/2$) isotope from EPR measurements. Principal values as large as 5 GHz are observed, leading to calculated total zero field splittings of 3.7 and 3.1 GHz for sites 1 and 2. With magnetic fields oriented along directions of large effective $g$ factors, ground state transitions can be tuned by several GHz with fields of tens of mT. In regions were the Zeeman and hyperfine interactions are
comparable in strengths transitions insensitive to magnetic field fluctuations are predicted, which could lead to increased coherence lifetimes. Extrapolated $A$ tensor for $^{173}\text{Yb}^{3+}$ ($I = 5/2$) isotope is in reasonable agreement with EPR angular variations (Figs. S5 and S6). Calculated zero field splittings are in the same range as for $^{171}\text{Yb}^{3+}$, with a much complex behavior under magnetic field.

In conclusion, these measurements suggest that $\text{Yb}^{3+}:\text{Y}_2\text{SiO}_5$ could be used for quantum information processing with optical, electron and nuclear spin degrees of freedom.

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