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Optical homogeneous and inhomogeneous linewidths in $^{171}Yb_3 : Y_2SiO_5$

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Rare earth ions are actively investigated as optically addressable spin systems for quantum technologies thanks to their long optical and spin coherence lifetimes. $^{171}Yb^{3+}$, which has 1/2 electron and nuclear spins, recently raised interest for its simple hyperfine structure that moreover can result in long coherence lifetimes at zero magnetic field, an unusual property for paramagnetic rare earth ions. Here, we report on the optical inhomogeneous and homogeneous linewidths in $^{171}Yb^{3+} : Y_2SiO_5$ (site 2) for different doping concentrations. While inhomogeneous linewidth is not correlated to $^{171}Yb^{3+}$ concentration, the homogeneous one strongly decreases between 10 and 2 ppm doping level, reaching 255 Hz at 3 K. This is attributed to a slowing down of 171Yb3+ ground state spin flip-flops.

Keywords: rare earth, quantum technologies, crystals, high-resolution spectroscopy.

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Introduction

Rare earth ions (RE) doped into crystals are promising centers for optical quantum technologies. They stand out in comparison to other solid-state systems because of the long coherence lifetimes (T_2) of both their optical and spin transitions at low temperatures. This opens the way to coherent light-atom-spin interfaces that can be used to build quantum memories, processors and optical to microwave transducers [1,2]. RE crystals are also versatile materials as they can be obtained as bulk single crystals, nanoparticles or thin films with good quantum state properties, allowing using them in a variety of devices designed for e.g increasing light-matter interactions [3].

One isotope of ytterbium, $^{171}Yb^{3+}$, has recently emerged as a particularly interesting system because it is the only paramagnetic RE ion with a 1/2 nuclear spin [4–8]. This gives a simple hyperfine structure that allows both spectral tailoring by ground state population storage and use of ground state coherence. This is especially important for e.g. long storage time quantum memories. In addition, $^{171}Yb^{3+}$ in Y_2SiO_5 (YSO) single crystals showed exceptionally long optical and spin coherence lifetimes, up to several 100 s of microseconds and few ms respectively, at zero applied magnetic field [4]. This very unusual observation is explained by a vanishing sensitivity to magnetic field fluctuations at first order (Zero First Order Zeeman or ZEFOZ effect), due in turn to a symmetric mixing of electron and nuclear spin wavefunctions. As a result, $^{171}Yb^{3+}$: YSO shows T_2 comparable to crystals doped with non-paramagnetic RE like Eu^{3+} or Pr^{3+}, while presenting strong electron-nuclear spin transitions that can be efficiently driven by short microwave pulses. This has led to the demonstration of a coherent optical memory with long storage time and large bandwidth [5]. $^{171}Yb^{3+}$ spin-spin interactions were also harnessed to strongly polarize ground state spins under optical pumping. This process, called Diffusion Enhanced Optical Pumping (DEOP), can extend optical coherence lifetimes up to about 800 μs [9].

In this paper, we report results on the dependence of inhomogeneous (Γ_{inh}) and homogeneous (Γ_{h}) optical linewidths as a function of $^{171}Yb^{3+}$ concentration, [Yb] in YSO. We show that at the concentration used, below 10 ppm, doping has a limited effect on Γ_{inh}, which nevertheless show significant variations across the series. On the opposite, a clear decrease in Γ_{h} is observed with decreasing [Yb]. We measure optical coherence lifetimes up to 1.25 ± 0.08 ms at zero magnetic field and $[\mathrm{Yb}] = 2$ ppm, equivalent to an effective homogeneous linewidth of 255 ± 16 Hz.

It is a great pleasure and honor for P.G. and all the authors to dedicate this paper to Prof. Marina Popova at the occasion of her 80th birthday. Prof. Popova has pioneered the field of high-resolution optical spectroscopy of rare earth ions and has in particular showed that hyperfine structures could be resolved in absorption spectra of high quality fluoride crystals and explained in the framework of crystal field and magnetic interactions theories [10–13]. It has been the starting point for many subsequent studies, and the research reported here is also part of this long string of discoveries and advances. Besides Prof. Popova's very high scientific skills, her human qualities and leadership

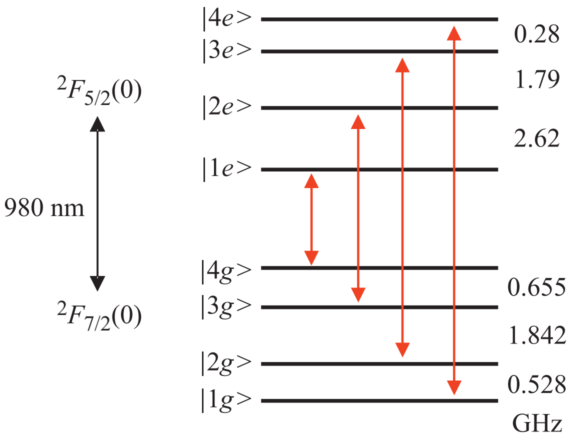

Figure 1. Hyperfine structure of the ground and excited states of the $^{171}Yb^{3+}$ ion in the pattern position 2 of Y_2SiO_5 crystal.

must also be mentioned and are recognized by a broad community beyond her team and colleagues at the Institute of Spectroscopy, Russian Academy of Sciences, in Troitsk.

Experimental

Y_2SiO_5 is a crystal with a monoclinic structure in which Y^{3+} ions occupy two crystallographic sites with a C_1 point symmetry. The principal dielectric axes are oriented along the crystal b axis and two perpendicular directions labelled $D1$ and $D2$. Samples were grown by the Czochralski technique starting from high purity (4N) raw oxide materials. Ytterbium doping was performed using isotopically enriched (95% ^{171}Yb) Yb_2O_3 powders (Isoflex USA) and the concentrations given in the text are nominal ones. Samples were cut along the b, $D1$ and $D2$ principal dielectric axes with typical dimensions of $10 \times 5 \times 5\,\mathrm{mm}^3$ and cooled down in closed-cycle refrigerators (Bluefors SD and CTI model 22).

Optical spectroscopy was performed using a narrow linewidth tunable diode laser (Toptica DL Pro) as the excitation source and amplified photodiodes for detection. For photon echoes experiments, the laser was focused by a 300 mm lens on the sample with a power of about 1 mW. Excitation and rephasing pulses were created using an acousto-optic modulator and had durations of about 1 μs.

Results and discussion

Fig. 1 shows the energy level structure of the lowest energy crystal field levels of the $^2F_{7/2}$ ground and $^2F_{5/2}$ excited multiplets [14] for $^{171}Yb^{3+}$ ions in site 2. Even at zero magnetic field, all hyperfine levels are non-degenerate, a consequence of the anisotropic hyperfine tensor. The splittings between levels are on the order of 0.5 to several GHz, in contrast to the zero-field hyperfine splittings found for non-paramagnetic ions like Eu^{3+} and Pr^{3+} in YSO that are in the range of 10 s to 100 s of MHz [1].

Absorption of $^{171}Yb^{3+}$ ions in site 2 of YSO in displayed in Fig. 2 for light polarized along the $D2$ axis and propagating along the b axis. A number of peaks are clearly observed for the 10 ppm sample and correspond to partially resolved transitions between ground and excited hyperfine levels. Indeed, the deconvoluted linewidth is 540 MHz, well below the largest hyperfine splittings. Four prominent peaks are labelled according to the main contributing transitions, which are starting from each of the ground state hyperfine levels. The narrow linewidths translate into relatively high peak absorption coefficients, given the low doping level, which is an important parameter for the efficiency of quantum memories. It can be increased by a factor of up to 4 by polarizing ground state spins into a single level using DEOP [9].

Absorption spectra of crystals grown at lower concentrations are also shown in Fig. 2. Peaks are much less resolved, translating into broader inhomogeneous linewidths of 1.3 and 0.93 GHz for the 5 and 2 ppm samples. This broadening, combined with the lower concentrations, explains the large difference between absorption coefficients of the 2 and 5 ppm samples compared to the 10 ppm one. The integrated spectra are actually in reasonable agreement with the relative nominal doping. The absence of correlation between linewidths and doping level suggests that the strain caused by the substitution of Y^{3+} by Yb^{3+} is not the dominant broadening mechanism. This is supported by the close ionic radii of Yb^{3+} and Y^{3+} (85.8 and 89.2 pm [15]) and the overall level of impurities in the sample on the order of several 10 s of ppm. We attribute the extra broadening in the 2 and 5 ppm samples to variations in growth conditions that affect defect level and therefore strain acting on $^{171}Yb^{3+}$ transitions.

Linewidths were recorded as a function of temperature (Fig. 3). Below 25 K, no broadening is observed but

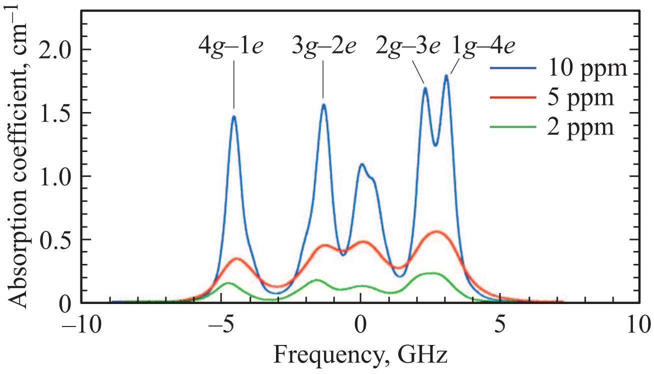

Figure 2. Absorption spectra of $^{171}Yb^{3+}:Y_2SiO_5$ crystals (ytterbium in position 2) for light polarized along D2 axis for various [Yb] concentrations. $T = 10$ K. Center frequency 306269 THz (978.854 nm in vacuum). The symbols at the top designate the transitions that make the largest contribution to the absorption peaks, the sequence order of hyperfine levels is shown in Fig. 1.

it then quickly grows to reach 2.5 GHz at 50 K. The curves can be well fitted with an expression including a temperature independent inhomogeneous broadening and a homogeneous one resulting from a 2-phonon Raman process [16]:

$$\Gamma = \Gamma_{\text{inh}} + \bar{\alpha}\left(\frac{T}{\theta_D}\right)^7 \int\limits_0^{\frac{\theta_D}{T}} \frac{x^6 e^x}{(e^x - 1)^2}\,dx, \tag{1}$$

where T is the temperature, $\bar{\alpha}$ a coupling coefficient, and θ_D YSO Debye temperature. The fits give $\bar{\alpha} = (9.3 \pm 0.8) \cdot 10^3$ and $(10.3 \pm 1.4) \cdot 10^3$ GHz for the 2 and 10 ppm samples, taking a Debye temperature of 420 K [17]. At low temperatures where the Raman broadening is proportional to T^7, this would correspond to an average coupling coefficient of $3 \cdot 10^{-3}$ Hz/K^7, similar to values found in $Eu^{3+}:Y_2SiO_5$ [17], but larger than the one estimated for $Yb^{3+}:Y_3Al_5O_{12}$ [8]. As in this latter work, we found that a resonant phonon process, i. e. phonon absorption between the two lowest crystal field levels of either the ground or excited sates, could also describe the experimental data.

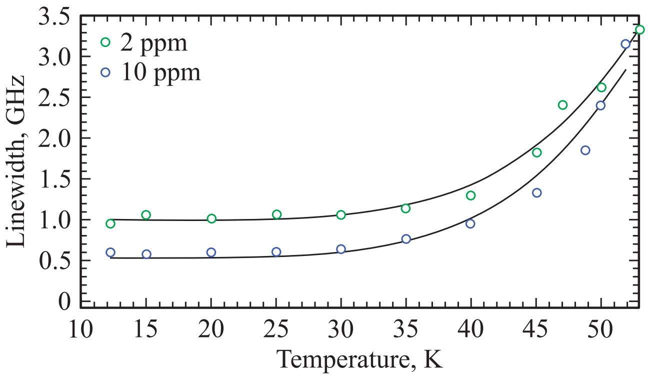

Figure 3. Width of the spectral absorption line of the $^{171}Yb^{3+}$ ion in YSO (position 2) vs. temperature. Solid lines are approximation using formula (1).

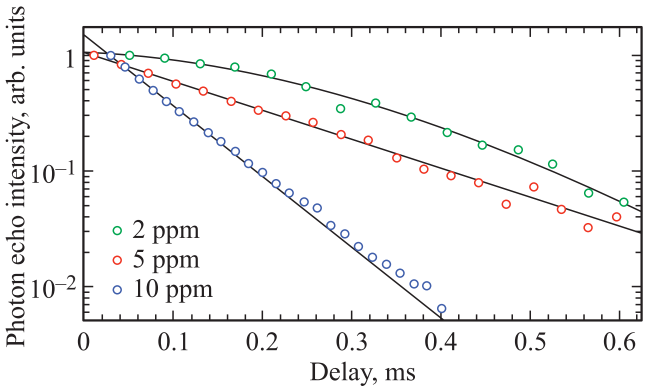

Figure 4. Signal amplitude attenuation of the photon echo vs. delay between the excitation and rephasing pulses for different concentrations of $^{171}Yb^{3+}$ in YSO (position 2). Circles correspond to experimental data, solid black lines are result of approximation, see text. $T = 3$ K.

However, in opposition to the Raman process, the fitted coupling coefficients were very poorly constrained. Further studies in a wider temperature range could help clarifying the contributions of these two processes. We also note that these results confirm that the observed variations in inhomogeneous broadening between samples cannot be due to differences in sample temperature that could occur because of varying thermal contact with the cold finger.

Coherence lifetimes were finally measured by the technique of photon echoes at a temperature of 3 K and with the laser set at the $3g-2e$ transition frequency (see Fig. 2). The echo intensity decays as the function of the delay . in the sequence excitation-τ-rephasing-τ-echo are shown in Fig. 4. Optical pumping by the pulses was low enough so that DEOP, which could modify T_2 values, did not occur. As can be seen from the units on the time axis, all samples show very long coherence lifetimes because of the ZEFOZ effect. In the 10 ppm sample, the echo decay is well described by an exponential, giving a T_2 of $285 \pm 6\,\mu$s ($\Gamma_h = 1.12 \pm 0.02$ kHz), in agreement with values measured on the $4g-1e$ transition at 2 K [9]. The 5 ppm exhibits a much longer decay which can also be well fitted by an exponential, giving $T_2 = 685 \pm 20\,\mu$s ($\Gamma_h = 465 \pm 13$ Hz), close to the value reported in [5]. Finally, the 2 ppm exhibits an even longer decay, with a non-exponential shape especially visible at short delays. It was fitted with a stretched exponential of the form:

$$I = I_0 \exp\left[-2\left(\frac{2\tau}{T_M}\right)^x\right], \tag{2}$$

where T_M is the phase memory time, leading to $T_M = 1.25 \pm 0.08$ ms and $x = 1.68$. The phase memory time corresponds to a $1/e^2$ decrease in echo intensity and is equivalent to T_2 when $x = 1$. The stretched exponential shape is characteristic of spectral diffusion, i. e. frequency shifts of optical transitions that occur on a time scale comparable to the echo decay [18]. The corresponding homogeneous linewidth can be estimated as $\Gamma_h = 1/\pi T_M = 255 \pm 16$ Hz. Taking into account the excited state lifetime of $1/2\pi T_1 = 122$ Hz contribution, we conclude that other dephasing processes represent a linewidth of 133 Hz. They are likely to be related to magnetic noise originating from slowly flipping spins. This produces magnetic noise that couples to ytterbium through the second order sensitivity of the transition to magnetic field. The flipping spins include those of $^{171}Yb^{3+}$ ions themselves in sites 1 and 2, as well as the nuclear spins of $^{89}Y^{3+}$ ions (100% abundance, $I = 1/2$).

The sharp decrease in coherence lifetime in higher doped samples is attributed to an increase in flip-flop rates between $^{171}Yb^{3+}$ ground state spins, which can be considered to vary as the square of the spin concentration. Flip-flops cause dephasing through magnetic noise as explained above but could also, especially in the 10 ppm sample directly affect optical coherence by reducing spin ground population lifetimes [9].

Conclusion

Inhomogeneous and homogenous optical linewidths were measured in $^{171}\mathrm{Yb}^{3+}:\mathrm{Y_2SiO_5}$ crystals with different doping concentrations. No correlation was found between inhomogeneous linewidths Γ_{inh} and Yb concentration in the range investigated, 2−10 ppm. This suggests that .inh, which varied from 0.54 up to 1.3 GHz, is determined by defects related to growth conditions. Transmission measurements as a function of temperature showed that the homogeneous contribution to the linewidth exceeds the inhomogeneous one for temperatures larger than 35 K and follows a two-phonon Raman process. At 3 K, homogeneous linewidths were measured by photon echo decays. A strong dependence on Yb concentration was observed, as T_2 increased from 0.285 to 1.25 ms when doping decreased from 10 to 2 ppm. In the latter case, the echo intensity decay showed a non-exponential shape that could be due to slow spectral diffusion driven by flip-flops among $^{171}\mathrm{Yb}^{3+}$ ground state spins in the two $\mathrm{Y_2SiO_5}$ sites and among $^{89}\mathrm{Y}$ nuclear spins. Further decrease in this magnetic noise could be obtained at lower $^{171}\mathrm{Yb}^{3+}$ concentrations or by polarizing ground state spins by optical pumping. This would leave flipping Y^{3+} nuclear spins as the remaining dephasing source and may allow T_2 to approach the radiative limit of 2.6 ms ($\Gamma_{\mathrm{h}} = 122\,\mathrm{Hz}$) as $^{89}\mathrm{Y}$ magnetic moment is weak and will couple to $^{171}\mathrm{Yb}^{3+}$ only to second order [18].

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Conflict of interests

The authors declare that they do not have any conflict of interests.

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