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Effects of disorder on optical and electron spin linewidths in Er^{3+},Sc^{3+}:Y_2SiO_5

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ABSTRACT

The material Er^{3+}:Y_2SiO_5 co-doped with Sc^{3+} is investigated for applications in optical quantum storage and signal processing. Replacing 1% of the Y^{3+} in the crystal with Sc^{3+} introduces static strain into the lattice that increases the inhomogeneous linewidth of the Er^{3+} optical transition at 1.536 μm to 25 GHz, a 50-fold increase compared to Er^{3+}:Y_2SiO_5 samples without Sc^{3+} co-doping. Electron paramagnetic resonance spectroscopy shows that electron spin linewidths are also strongly increased, confirming the previously proposed mechanism for decoherence suppression by using disorder to inhibit resonant spin-spin interactions. Analysis of the spin line broadening as a function of magnetic field orientation indicates the presence of contributions that cannot be modeled by a simple change in the electronic **g** tensor. Optical homogeneous linewidths of less than 2 kHz are observed for a weak magnetic field of 0.1 T and also for fields greater than 2 T with the field oriented near the $\boldsymbol{D_2}$ crystal axis and at a temperature of 1.7 K. These results suggest that this material can be useful for high-bandwidth classical and quantum information processing in the telecom C-band.

1. Introduction

Rare-earth ion (RE) doped crystals can have very narrow optical transitions at liquid helium temperature, making them attractive for applications in quantum information processing and advanced RF signal processing [1–6]. One key property of these materials is the potential for a high ratio between the optical inhomogeneous (Γ_{inh}) and homogeneous (Γ_h) linewidths. This ratio determines the maximum time-bandwidth product for coherent signal processing, reaching values as high as 10^8 with Γ_{inh} = 1–100 GHz and Γ_h = 0.1–1 kHz [3]. This allows signals with high bandwidth to be stored in quantum memories for a long time, or alternatively, the high resolution spectral analysis of RF signals. Er^{3+} is particularly interesting because it has a transition at 1.5 μm that is directly compatible with telecommunication components in existing optical fiber networks. This has recently motivated several promising demonstrations in quantum storage and signal processing [7–10]. Er^{3+} doped into Y_2SiO_5 (Er:YSO) is currently the solid-state system with the narrowest known optical homogeneous linewidth Γ_h (73 Hz) [11,12]. The inhomogeneous absorption linewidth Γ_{inh} is typically 0.5 GHz or less in this crystal [13], which limits the signal processing bandwidth and spectral multiplexing capacity for some potential device applications. To overcome this problem, it has been shown that co-doping Er:YSO with Eu^{3+} can induce static strain in the lattice and broaden the optical line to several 10's of GHz [14]. Moreover, under specific experimental conditions, a decrease of the homogeneous linewidth was also observed [15]. At temperatures below 4 K, Γ_h in this system is dominated by magnetic fluctuations due to Er^{3+} ground-state electron spin flips. The decrease of Γ_h was then attributed to a reduction of the Er^{3+}-Er^{3+} spin flip-flop rate since the disorder is also expected to increase the inhomogeneous broadening of the electron spin linewidth, reducing the overlap of neighboring spin resonances in the lattice and therefore inhibiting the spin flip-flop dynamics (spin diffusion) that require spins to have identical Zeeman splittings for energy conservation.

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Here, we investigate Sc^{3+} as an alternative co-dopant in Er:YSO. Sc^{3+} is of interest since it is expected to have a larger effect on the lattice than Eu^{3+}. It also has no optical transitions that could interact with Er^{3+} or any other RE ions, and therefore could be used to broaden Eu^{3+} optical transitions that are also used in quantum information processing [16–21]. In addition, Sc^{3+} doping has previously been investigated for inducing static disorder in Eu^{3+},Sc^{3+}:Y_2O_3 where no detrimental effect on the optical homogeneous linewidth of Eu^{3+} was observed due to the Sc^{3+} [22], suggesting its promise as a dopant for Y_2SiO_5. However, the ^{45}Sc isotope has a non-zero nuclear spin (I = 7/2, μ = +4.76 μ_N) with 100% natural abundance [23], potentially impacting the Er^{3+} coherence lifetime due to interactions between the Er^{3+} electronic spin and the ^{45}Sc nuclear spin at higher Sc^{3+} doping levels.

We report the optical inhomogeneous and homogeneous linewidths measurements at low temperatures and in varying magnetic fields. We also perform electron paramagnetic resonance spectroscopy and analyze the Er^{3+} electron spin transition linewidths. Analysis of these results demonstrates that Sc^{3+} is a promising dopant for increasing the optical bandwidth of Er^{3+} materials for photonic signal processing and quantum information applications.

It is a great honor and pleasure to dedicate this paper to Professor Georges Boulon. Among his outstanding works and achievements, we would like to point out his renewed interest in new topics and support of young scientists. This includes collaborative work on Tm^{3+}:Y_2O_3 for quantum storage [24], and the highlight on quantum information processing in RE doped crystals during the 2008 International Conference on Luminescence that he chaired. The work reported here on employing substitutional dopants to induce disorder and manipulate the properties of materials for quantum and classical signal processing follows in the spirit of Professor Boulon's significant investigations of the effects of dopants and the resulting disorder on the spectroscopic properties of laser materials, including inhomogeneous broadening, appearance of multisites, and changes to relaxation and energy transfer dynamics as studied extensively in Refs. [25–27], for example.

2. Materials

Samples of nominally 0.003 at.% Er^{3+}, 1 at.% Sc^{3+}:Y_2SiO_5 (Er,Sc:YSO) were obtained from a boule grown by the Czochralski method. YSO is a monoclinic crystal with a structure corresponding to the *C*2/*c* space group. Er^{3+} and Sc^{3+} can substitute for Y^{3+} in two different crystallographically inequivalent sites, both with C_1 point symmetry. For each of the two crystallographic sites, in magnetic fields that are not parallel or perpendicular to the ***b*** crystallographic axis, there are two magnetically inequivalent sub-classes of sites with different local orientations relative to the external field. All samples were cut with faces nominally perpendicular to the crystal's ***b***, ***D*****1**, and ***D*****2** principal dielectric axes, but with an uncertainty of up to several degrees that resulted in slight differences in magnetic field orientations between samples. In this work, we primarily focus on Er^{3+} ions at site 1 since this is the site that is most often employed in applications. Scandium has a 100% abundant isotope ^{45}Sc that has a nuclear spin of I = 7/2 and a magnetic moment μ = +4.76 μ_N, where μ_N is the nuclear magneton [23].

3. Results and discussion

3.1. Optical inhomogeneous linewidths and lineshifts

The inhomogeneous absorption of Er^{3+} at site 1 for the Sc^{3+}-doped sample was measured by recording the transmitted spectrum of a broadband source (amplified spontaneous emission of an erbium-doped-fiber amplifier, EDFA) using an ANDO AQ6317B Optical Spectrum Analyzer with a resolution of 2 GHz. For this measurement, the sample was mounted in a Janis Research cryostat with a variable temperature insert and held at 1.8 K. The absorption spectrum of the $^4I_{15/2}(1) \rightarrow {}^4I_{13/2}(1)$ transition for light polarized along ***D*****2** and propagating along ***b*** is shown in Fig. 1. The label 1 denotes the lowest crystal field level of each multiplet. A broad inhomogeneous line is observed, with a full width at half maximum (FWHM) of 25 GHz. This is much larger than the 0.5 GHz FWHM measured for non-codoped Er:YSO samples with similar Er^{3+} concentration. A quantum memory or signal processor based on Er:YSO with 1% Sc^{3+} co-doping could therefore offer a bandwidth about 50 times larger than the material without co-doping. The broadening of the line is attributed to static lattice strain induced by the random distribution of Sc^{3+} ions replacing Y^{3+} ions. This is in agreement with the Lorentzian shape of the absorption line that indicates a broadening induced by point defects at low concentration [28]. We find that Sc^{3+} co-doping has a stronger effect on optical line broadening than Eu^{3+} since an 11 GHz FWHM was obtained for a 1% Eu^{3+} co-doping of Er^{3+}:YSO [14]. We expect Sc^{3+} to introduce a greater amount of strain in the lattice since it has an ionic radius (0.870 Å) that is 15% smaller than Y^{3+} (1.019 Å), whereas Eu^{3+} is only 4.6% bigger than Y^{3+} (1.066 Å) [29]. In contrast, Er^{3+} itself has an ionic radius (1.004 Å) that is a much closer match to Y^{3+} so that we expect the Er^{3+} doping to have a negligible effect on the linewidths at the low concentrations studied here.

The peak absorption coefficient observed in this sample was 0.14 cm^{-1}. The integrated absorption measured in 0.003%Er,1% Sc:YSO is consistent with those found in 0.02%Er^{3+},1%Eu:YSO and 0.02%Er:YSO [14]. This suggests that co-doping does not significantly change the Er^{3+} transition oscillator strength. Although this conclusion is reasonable, it should be noted that transitions of Eu^{3+} in YSO are known to be sensitive to growth conditions [30].

To further probe the effects of Sc^{3+} doping on the linewidths and the absolute frequency of the optical transition, additional high-resolution laser absorption measurements were performed. For these measurements, the 0.003%Er,1%Sc:YSO sample and a 0.005% Er:YSO sample were both mounted in a Montana Instruments Cryostation C2 system and held at 3 K for direct comparison of the

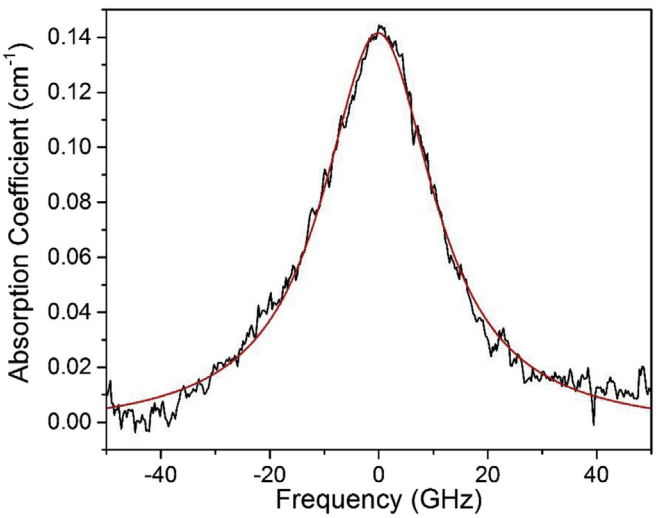

Fig. 1. Absorption spectrum of the $^4I_{15/2}(1) \rightarrow {}^4I_{13/2}(1)$ transition for Er^{3+} at site 1 in 0.003%Er,1%Sc:YSO for **E**//***D*****2** and **k**//***b*** at T = 1.8 K, with the fit to a Lorentzian lineshape shown. Zero frequency corresponds to 195125.8 GHz.

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