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Short term mobility program 2015 -- final report

Herewith I describe the activity that I carried out during the three-weeks period funded by the CNR 2015 Short Term Mobility program. The initial proposal was focused on a single sample, namely Pr^{3+}:LaF_3. As I will show in this report, due to the characteristics of the samples in use the program could be very much extended to a wider variety of materials. This was made possible by the positive collaboration of R.M. Macfarlane and P. Goldner who provided a large set of samples and high quality lab instrumentation. Given the amount of data and understanding obtained within the three weeks of the program, the overall experimental session can be considered successful and likely to give enough material for publication and further developments.

Experimental setup preparation

The experimental setup was implemented during the first week of work, and the activity consisted in :

- Tuning the stabilized laser source (tunable dye laser, Sirah 'Matisse') to get stable operation with a spectral bandwidth of about 100 kHz and possibility to run wide frequency scans (up to 100 GHz).
- Implementing radiofrequency circuits to excite nuclear quadrupole transitions in the 5-90 MHz range.
- Aligning the optical table in order to get a collimated beam controlled by acousto-optic modulators mounted in retro-reflection configuration.
- Preparing an appropriate insert for the optical cryostat to host a radiofrequency excitation coil and the sample mount.
- Preparing the samples (cutting, polishing, mounting).
- Implementing a fluorescence excitation setup for Optically Detected Nuclear Magnetic Resonance (ODNMR) experiments.

Pr^{3+}:LaF_3, single crystal

We measured the optical absorption from the $^3H_4 \rightarrow {}^1D_2$ transition in the sample (Pr^{3+}:LaF_3 single crystal, doping concentration 0.03%). The absorption line is shown in Fig.1. The central wavelength is located at 505865 GHz and is inhomogeneously broadened by 18 GHz.

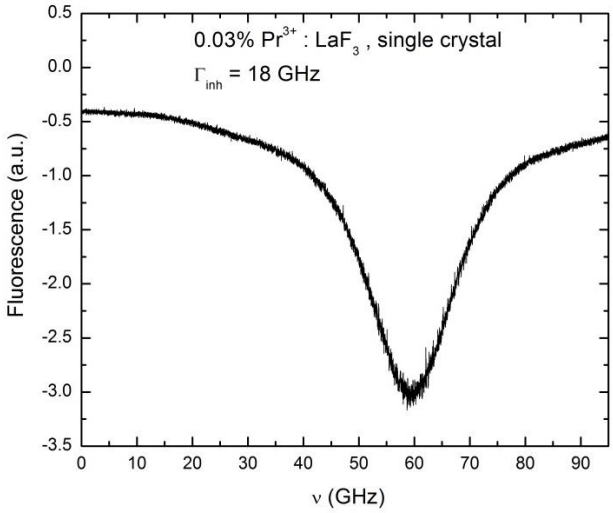

Fig. 1 Fluorescence excitation spectrum corresponding to the $^3H_4 \rightarrow {}^1D_2$ transition of Pr^{3+} in LaF_3.

This is a rather anomalous behaviour for this material, given that earlier characterizations reported absorption lines about three times narrower [1]. This, together with presumably short coherence times, was identified as the reason why a Raman Heterodyne signal could not be detected for this sample. We therefore moved to an alternative approach for measuring the hyperfine transitions of Pr^{3+} in the electronic ground state. We observed Optically Detected Duclear Magnetic Resonance (ODNMR) for the two nuclear quadrupole transitions in the ground state, as shown in Fig. 2, measuring a linewidth of 450 kHz for the transition centered at 16.7 MHz.

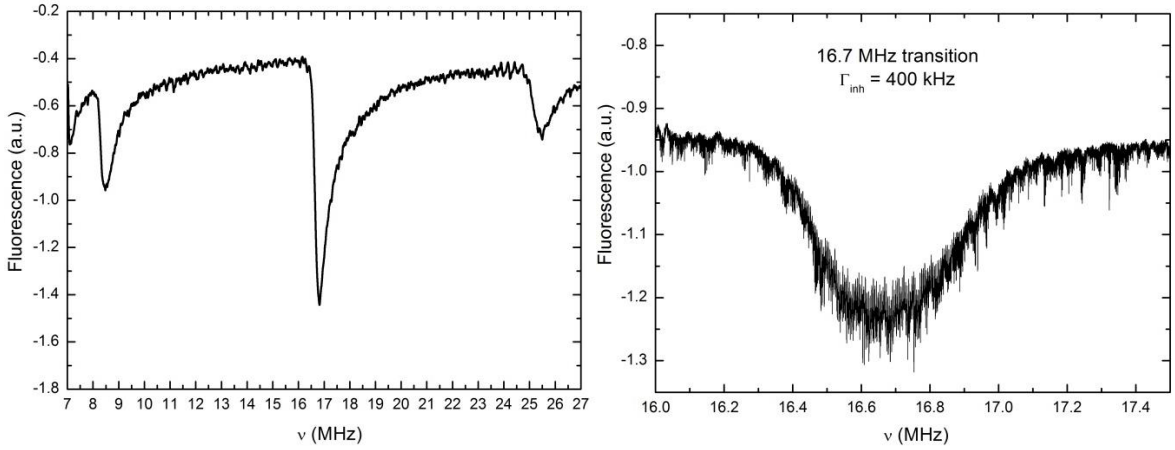

Fig. 2 ODNMR signal recorded by burning a hole in the $^3H_4 \rightarrow {}^1D_2$ transition, scanning 50 W of rf field across the nuclear quadrupole transitions and observing the increase of fluorescence due to the partial refilling of the hole by rf pumping of the spin state. Left: identification of the central frequencies. Right: Sample-and-hold measurement of the inhomogeneous linewidth of the transition centered at 16.7 MHz obtained by slowly scanning a pulsed rf source (1.5 MHz in 20 s)

This is intriguingly higher than the previously reported values of about 180 kHz[2]. Although we do not expect any specific correlation between the optical and the nuclear quadrupole inhomogeneous broadening of the transitions (they are related to different processes), our observation suggests possible strain-induced

broadening on both transitions. This is particularly interesting, as many studies are currently directed towards the identification of the origin and possible control of the inhomogeneous widths in rare-earth transitions in crystals for solid-state quantum information applications [3]. Accurate Stark measurements on the nuclear quadrupole transitions need higher sample quality, being strongly dependent on the electric field gradients on the nuclei site, so we decided not to insist further on this kind of measurements.

EuP_5O_{14}, single crystal

We investigated the hyperfine structure of a fully-concentrated, Eu-doped pentaphosphate EuP_5O_{14}. The motivation for this work is mainly the renewed interest in stoichiometric materials for quantum computing applications[4]. Moreover, previous results showing excitation energy migration effects in the same material[5] need to be completed, understood and clarified. From our measurements we gained a better understanding of the dephasing characteristics for the $^7F_0 \rightarrow {}^5D_0$ Eu^{3+} optical transition and we determined other important parameters for this system.

We examined four samples, grown at Lincoln Laboratories (MIT) and at University of Hamburg. We mounted the samples in a radiofrequency excitation coil inside a liquid helium cryostat and measured the optical and hyperfine transitions characteristics. We monitored the absorption parallel to the crystal *b* axis. The structure of the inhomogeneously broadened $^7F_0 \rightarrow {}^5D_0$ lines turned out to be rather unusual, showing multiple components likely to belong to different crystal phases (Fig. 3). The average integrated cross section for the four samples is 0.0059×10^{-19} cm^2 and the oscillator strength for E ∥ b is 0.061×10^{-9}.

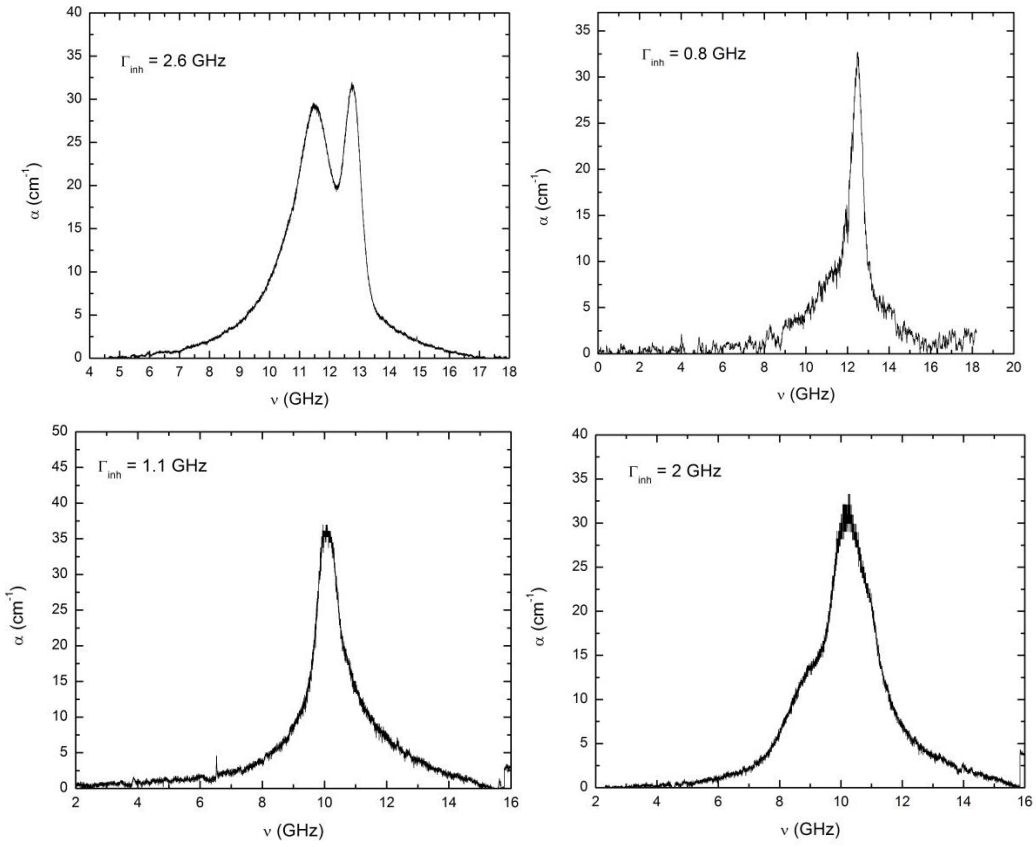

Fig. 3 Absorption measurements corresponding to Eu^{3+} $^7F_0 \rightarrow {}^5D_0$ transition on four different EuP_5O_{14} samples with the same nominal composition. The sample corresponding to the top-right spectrum was the one chosen for further analysis

We recorded photon echo decays using heterodyne detection and measured the power dependence of the coherence time T_2. Measurements were made at 518108.4 GHz (-4 GHz from line center) and at 518102.4 GHz (-10 GHz from line center). The power-dependent homogeneous linewidth extracted from the echo decay measurements is shown in Fig.4 and extrapolates to an homogeneous width of 9 kHz. The zero-excitation power extrapolation gives a good indication of the homogeneous width, being ideally free of instantaneous spectral diffusion.

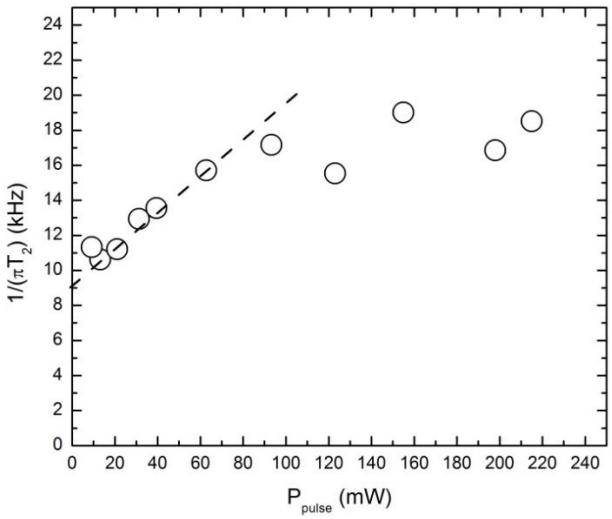

Fig. 4 Photon echo decay measurements on the Eu^{3+} $^7F_0 \rightarrow {}^5D_0$ transition recorded at -4 GHz from the center of the absorption line. The power of the excitation pulses was decreased in order to reduce the effect of instantaneous spectral diffusion. The zero-power extrapolation gives a homogeneous linewidth of 9 kHz.

We recorded ODNMR spectra for the four Eu^{3+} nuclear quadrupole transitions in the ground state, for the two different europium isotopes ^{151}Eu and ^{153}Eu. The spectra relative to ^{151}Eu are shown in Fig. 5. From these spectra important informations such as the inhomogeneous width of the nuclear quadrupole transitions and lineshape were deduced.

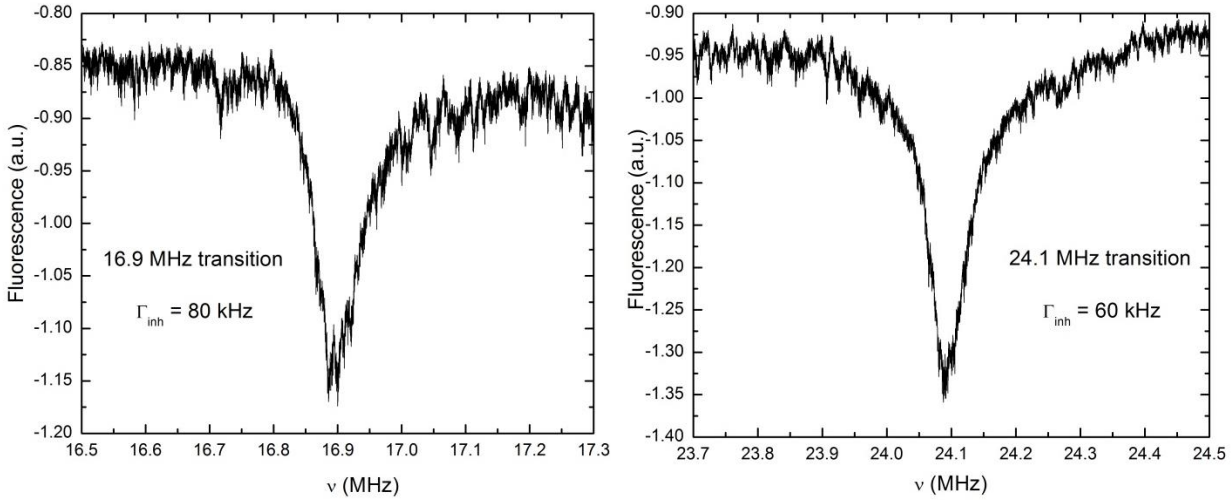

Fig. 5 ODNMR spectra recorded for the two nuclear quadrupole transitions of Eu^{3+} in EuP_5O_{14} single crystal. Inhomogeneous linewidths of 80 kHz and 60 kHz were measured for the 16.9 MHz and 24.1 MHz transitions, respectively.

The next step for this experiment will be to measure the homogeneous linewidth of the ground state quadrupole transitions. The first attempts we made to record spin echo signals were not successfull. This is most likely due to a non-optimal focusing of the laser on the thin (about 200 µm) crystal, as we deduced from the oscillator strength following the data processing. Ideally one would like to see to what extent the phosphorus ions contribute to the dephasing of the spin transitions. This could be the topic of next experimental session, now that all the parameters involved in the formation of the echo are more clearly understood.

Pr^{3+}:CaF_2, ceramics and single crystals

We investigated the inhomogeneous lineshapes of the $^3H_4 \rightarrow {}^1D_2$ Eu^{3+} transition in CaF_2 ceramics and single crystals. The aim of this is to determine the effect of the preparation process of the ceramic materials on the optical transitions linewidths. Ceramic samples were grown at Chimie Paristech.

Fluorescence excitation measurements on the reference single crystal (0.05% Pr^{3+} doping) nicely reveal the hyperfine structure of the ground state (Fig 6), with an overall width of about 18 GHz resulting from the six transitions originated from the hyperfine levels. The same measurement for a 0.02% Pr-doped ceramic shows two distinct, inhomogeneously broadened spectral features separated by circa 50 GHz. These can be attributed to a strain-induced broadening and splitting of the E ground state[6,7]. Another possible explanation to this is the presence of another occupation site with slightly different energy strucutre, although this latter explanation seems to be less likely.

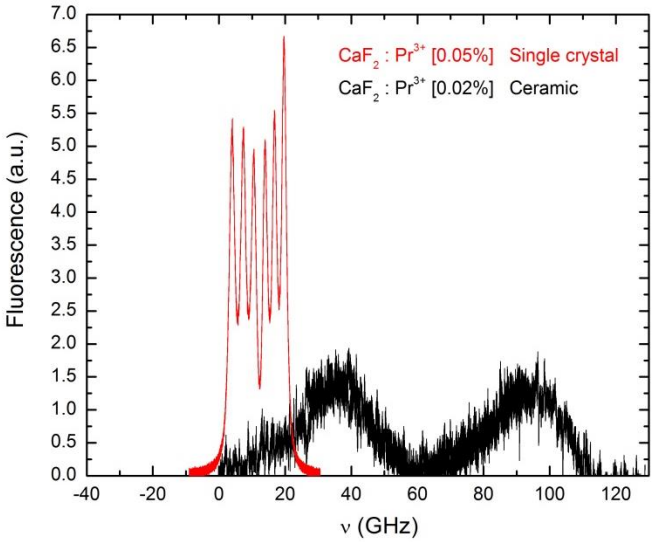

Fig. 6 Fluorescence excitiation spectra corresponding to the $^3H_4 \rightarrow {}^1D_2$ transition in CaF_2 ceramic (black) and single crystal (red). The ground state hyperfine strucure is masked by inhomogeneous broadening in the ceramic samples.

We plan to investigate further this material, given the novelty and interest of the effect that is shown by the ceramic sample. A further improvement of the experimental technique adopted for this kind of measurement will make us able to understand more precisely the process leading to the broadening and splitting of the absorption lines.

We also measured the phonon relaxation rate at 6 K for this material. To do this, we recorded fluorescence excitation spectra by scanning the excitation laser across the E → D2 transition. The D2 level lies 20 cm^{-1} above D1 and thus is the main responsible for Orbach dephasing of the zero phonon line transition. The collected spectra are shown in Fig 7. The red line is a fit to the data obtained by artificially broadening each hyperfine line by 10 GHz which represents the phonon relaxation rate for this material.

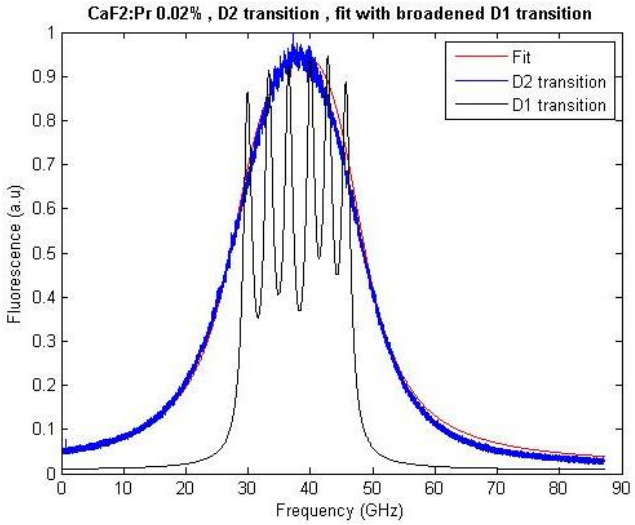

Fig. 7 Fluorescence excitation spectra corresponding to the E → D1 and E → D2 transitions in Pr^{3+}:CaF_2. The red line is a fit to the data obtained by imposing a 10 GHz broadening to each hyperfine line of the E → D1 transition.

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