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OPTICAL DEPHASING OF Pr$^{3+}$ IONS BY NONEQUILIBRIUM PHONONS IN LaF$_3$ AND YAlO$_3$

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Abstract - We describe a new effect, phonon-induced coherence loss (PICOLO), whereby a monoenergetic nonequilibrium phonon distribution is responsible for the loss of optical coherence. The interaction of these phonons with the ions is monitored with the free induction decay or photon echo. We show how PICOLO can be used to study the electron-phonon interaction and the dynamics of phonons.

I. Introduction - We report the first observation of optical dephasing produced by a monoenergetic nonequilibrium distribution of phonons (PICOLO). The system studied is LaF$_3$ containing 0.05 at. % Pr$^{3+}$. Optical dephasing due to a thermal distribution of phonons has been studied extensively in this system using homogeneous linewidths /1,2/ and coherent transient spectroscopy/3,4/. In this paper, optical free induction decay (FID) is observed on the $^3$H$_4(I)\rightarrow^1$D$_2(I)$ transition of the Pr$^{3+}$ ion at 5925 Å /5/ (see Fig. 1). The 23 cm$^{-1}$ nonequilibrium phonons are resonant with the $^1$D$_2(I)\rightarrow^1$D$_2$(II) transition and are absorbed by the excited Pr$^{3+}$ ions causing dephasing. We show how

![Energy level diagram of Pr$^{3+}$ in LaF$_3$ showing the low-lying crystal field levels in $^3$H$_4$ and $^1$D$_2$.](a)

![Experimental geometry for PICOLO experiments.](b)
PICOL0 can be used (1) to isolate and study specific energy relaxation pathways and (2) as a frequency selective detector to study the dynamics of high frequency phonons.

II. Experiments - Monoenergetic phonons were generated by selectively pumping the $^3H_4(I)$-$^1D_2(II)$ transition at 5917A with a Nd:YAG pumped tunable dye laser whose bandwidth was 2 GHz. Rapid relaxation ($T_1(II)=225\text{ps}$) to $^1D_2(II)$ generates a monoenergetic distribution of phonons which are resonant with the $^1D_2(I)$-$^1D_2(II)$ transition (see Fig. 1). The FID was observed on the $^3H_4(I)$-$^1D_2(II)$ transition, excited by an actively stabilized cw tunable dye laser whose frequency was switched by an intracavity electro-optic phase modulator. The 20 mW, 1MHz bandwidth laser output was gated on to the sample for 20 µs with an acousto-optic modulator. After a 10 µs preparation time, the laser frequency was shifted by 20 MHz. After 5 µs the laser was returned to the original frequency. The frequency shifted laser acts as a local oscillator for the heterodyne detection of the FID. The heterodyne signal was detected with an avalanche photodiode after filtering with a monochromator to reject the scattered laser light at the 5917A phonon generator wavelength.

The experimental geometry is shown in Fig. 1b. The cw dye laser was focused to a 50 µm spot in the sample. The Nd:YAG pumped dye laser propagated at a right angle to the cw beam and was focused with a cylindrical lens to a line 100 µm thick and 10 mm long which overlapped the cw beam. Because of the strong absorption of the pulsed dye laser on the $^1D_2(II)$ level ($α=10\text{cm}^{-1}$), the cw laser beam is brought close to the surface through which the pulsed laser enters the sample. The timing sequence and delay between the frequency switch of the cw dye laser and the output of the pulsed dye laser was continuously variable electronically.

III. Experimental results - The FID signal in the absence of the 23 cm$^{-1}$ phonons is shown in Fig. 2, top trace. It exhibits a decay time of 1.5 µs, corresponding to a dephasing time of 3 µs. This is somewhat shorter than the true low temperature dephasing time of 5 µs obtained from photon echo experiments due to laser frequency jitter during the preparation time. When phonons are generated during the FID decay, an enhancement of the dephasing rate occurs (Fig. 2). With an energy input of 300 µJ (not shown), the coherence decays in less than one cycle of the 20 MHz FID signal. There is a small remaining FID signal which arises from the 0.5 mm end sections of the sample which are masked so as to prevent surface heating of the endfaces by the pulsed laser. The heating produces bubbles in the superfluid helium introducing strong oscillatory signals which interfere with the measurement of the FID. As the phonon generating
laser energy is reduced, the FID signal decays over many cycles but at a rate which is still faster than that observed under the 1.5K equilibrium conditions.

Phonons were also generated at times \( \Delta t \) prior to the FID signal. For strong pulsed excitation and times \( \Delta t < 1 \mu s \) only the FID signal from the masked ends is observed. However, even for much weaker pulsed laser excitation conditions and for \( \Delta t \gg T_{ph} \), the phonon lifetime, significant dephasing effects are evident.

IV. Analysis of PICOLO - The dephasing of the FID signal results from the absorption of the resonant 23 cm\(^{-1} \) phonons. Therefore the nonequilibrium phonon contribution to the dephasing rate, \( T_2^{-1} \), can be related directly to the population relaxation time of \( 1D_2(II) \) and the occupation number of resonant 23 cm\(^{-1} \) phonons, \( \bar{n}_{23}(t) \), with the expression

\[
T_2^{-1}(t) = T_1^{-1}(II) \bar{n}_{23}(t).
\]

(1)

We first consider the measurement of phonon lifetimes obtained by generating the phonons during the FID signal. For the case of large resonant phonon occupation number \( \bar{n}_{23}(0) > 10^{-2} \), the dephasing occurs in a short time compared to \( T_{ph} \) and \( \bar{n}_{23}(t) \) can be considered constant during the dephasing. However, for smaller \( \bar{n}_{23}(0) \), the resonant phonons decay during the dephasing and, as a result, the FID decay is nonexponential.

We define a fractional FID amplitude, \( f(t) \), which is the amplitude at times \( t \) divided by the amplitude before phonon generation and a no-phonon coherence time, \( T_2 = 3 \mu s \). We find that \( f(t) \) decays according to

\[
\ln[f(t)] = -2\bar{n}_{23}(0) T_1^{-1}(II) T_{ph}[1 - e^{-t/T_{ph}}] - 2T_2'^{-1}t.
\]

(2)

The experimentally observed \( f(t) \) is plotted in Fig. 3 along with the values calculated from Eq. (2) for several values of \( T_{ph} \). The value of \( \bar{n}_{23} \) at \( t = 0 \) is obtained from the initial slope of the decay. We see that the non-exponential behavior of \( f(t) \) is well described with a phonon lifetime \( T_{ph} = 500 \) ns, in agreement with the hot luminescence data/8/.

We next consider the persistence of the phonon-induced optical dephasing, shown in Fig. 4, even when the delay between phonon generation and the FID signal, \( \Delta t \), is much greater than the phonon lifetime. When \( \Delta t \gg T_{ph} \), we expect the 23 cm\(^{-1} \) phonon occupation number to vary slowly during the observed FID decay so that \( T_2^{-1} \) will be proportional to a nearly constant \( \bar{n}_{23} \). We obtained \( \bar{n}_{23}(t) \) independently from the hot luminescence from \( 1D_2(II) \) which is proportional to \( \bar{n}_{23}(t) \). We found that \( \bar{n}_{23}(t) \) contained a long tail out to \( \Delta t > 100 \mu s \). This probably

![Graph](image-url)

**Fig. 3.** Time dependence of the 23 cm\(^{-1} \) contribution to the FID amplitude, corrected for end effects, for several pulsed dye laser energies. The solid curves are fits with Eq. (2) to the 4\( \mu \)J data for several values of \( T_{ph} \).
results from trapping of the anharmonic decay products of the 23 cm\(^{-1}\) phonons which remain in the excited volume due to elastic impurity scattering and which can recombine to form 23 cm\(^{-1}\) phonons at time \(\Delta t \gg T_{\text{ph}}\). At long times (\(\Delta t > 5 \mu s\)) the observed \(T_2^*\) values (circles in Fig. 4) are consistent with those predicted (squares) from Eq. (1) using the measured value of \(\bar{n}_{23}(\Delta t)\). For \(\Delta t < 1 \mu s\), \(T_2^*\) is only 1/6 of the value predicted on the basis of \(\bar{n}_{23}(\Delta t)\). A plausible explanation is that our laser (bandwidth=2GHz) excites a select group of ions in the \(^2\text{D}_2(\text{II})\) inhomogeneously broadened line (optical linewidth = 6GHz) with the result that the phonons generated by the \(^2\text{D}_2(\text{I})\) relaxation have a narrower frequency distribution than the full excited state resonance. As a result, they may be slightly out of resonance with the \(^2\text{D}_2(\text{II})\) transition of the ions undergoing FID which were prepared with a different laser. The electron-phonon coupling is reduced because of the resonance mismatch. For \(\Delta t > 5 \mu s\), available 23 cm\(^{-1}\) phonons have been produced by recombination and contain a broad frequency distribution like that present in a thermally excited phonon population.

V. Photon Echoes in YAlO\(_3\):Pr\(^{3+}\) - We also studied PICOLO with photon echoes produced by a gated cw dye laser tuned to the 6107A \(^3\text{H}_4(\text{I})\rightarrow^2\text{D}_2(\text{I})\) transition of Pr\(^{3+}\) in YAlO\(_3\)/7/. The first excited crystal field levels which can produce coherence loss by phonon absorption occur at 51 and 274 cm\(^{-1}\) in the ground and excited levels, respectively/9/. Since our sample was brown in color due to color centers which did not fluoresce, we assume that absorbed photon energy from a pulsed dye laser tuned to \(\sim 6500\)A, off resonance with any Pr\(^{3+}\) transitions, was efficiently converted to heat via non-radiative relaxation. The pulsed laser beam was propagated colinearly with the cw laser and prepared a heat pulse which contained some phonons in resonance with the \(^3\text{H}_4(\text{I})\rightarrow^2\text{H}_4(\text{II})\) transition.

The effect of these optically produced heat pulses is shown in Fig. 5. The upper trace shows the \(\pi/2\) and \(\pi\) preparation pulses produced with an acousto-optic modulator and an echo viewed through an optical shutter consisting of a Pockels cell between a pair of crossed polarizers. The shutter was opened \(\sim 1 \mu s\) after the end of the preparation pulses. The echo, on an expanded scale, is shown with and without the heat pulse. With 100 \(\mu\)J of pulsed laser energy focused to a 1mm diameter spot, the echo terminates within 50ns. In this case, we made no attempt to analyze the PICOLO results since \(T_1\) for the levels involved in the coherence loss and the resonant phonon occupation number were both unknown. However, the coherence loss appears to be highly non-exponential, suggesting that the high frequency phonons in resonance with the crystal field transition within the ground and excited states thermalize rapidly.
VI. Conclusions - With PICOLO it is possible to observe the presence of nonequilibrium phonons of a particular frequency with great sensitivity. In the present case, occupation numbers of $10^{-4}$ were detected, but in some systems, detection levels as low as $10^{-7}$ are to be expected. Parameters which lead to these sensitivities are short population relaxation times of upper electronic states, e.g., $T_1(II)$, and long coherence times, $T_2$. In addition, PICOLO has good spectral, spatial and temporal resolution. Spectral resolution is limited to excited state inhomogeneous resonance widths, typically 1-10GHz. Temporal resolution using FID techniques is determined by the heterodyne frequency. Although this was only 20MHz in our experiments, much larger frequencies are possible, limited ultimately by detector frequency response and the extent of the laser frequency shift.

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