# PHOTON ECHOES IN $LaF_3:Nd^{3+}$ AND YAG:$Nd^{3+}$ ☆

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Magnetic field and temperature dependent photon echo behavior in $LaF_3:Nd^{3+}$ and YAG:$Nd^{3+}$ is reported and compared with corresponding behavior in $CaWO_4:Nd^{3+}$.

We report the observation of photon echoes for the $^4I_{9/2}-^4F_{3/2}$ transition of $Nd^{3+}$ in both lanthanum trifluoride and yttrium aluminum garnet. Previously, echoes in this transition have only been observed in calcium tungstate [1]. We compare the dependence of the echo signals in these crystals on sample temperature and applied magnetic field.

In all three crystals, the ground state $^4I_{9/2}$ is split into five Kramers doublets ($Z_1-Z_5$) and the $^4F_{3/2}$ state into two doublets ($R_1$ and $R_2$) as shown in fig. 1 [2–4]. The echoes were excited between the $Z_1$ and $R_1$ levels. Our samples were: $LaF_3:Nd^{3+}$ (0.1 atom %), $CaWO_4:Nd^{3+}$ (0.1 atom %, uncompensated), and YAG:$Nd^{3+}$ (0.7 atom %), with transition frequencies corresponding to 8626 Å [2], 8767 Å [3], and 8749 Å [4], respectively. The experimental arrangement was similar to ref. [1] except that the excitation laser pulses were collinear and consequently the echoes were also emitted collinearly. A three-stage Kerr cell optical shutter prevented the photomultiplier detector from saturating. The laser $E$-field was vertical. The $LaF_3$ and $CaWO_4$ samples were oriented with the $c$-axis perpendicular to both the $E$-vector and the direction of propagation to maximize the echo signal. The sample temperature was varied between 3°K and 20°K, and a magnetic field variable up to 3 kG was applied along the $c$-axis. The shot-to-shot signal fluctuation was typically a factor of two.

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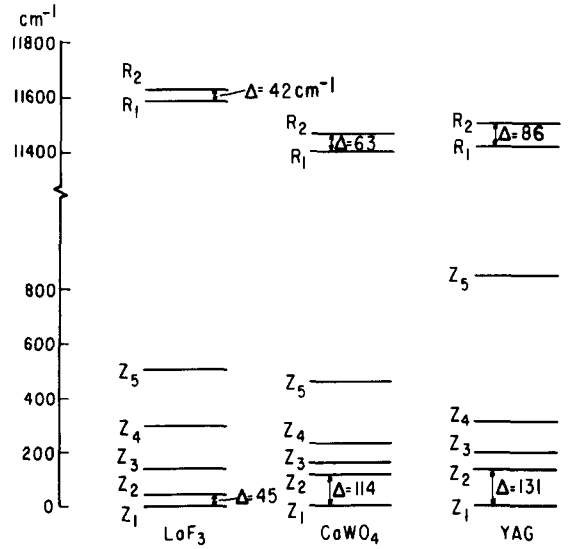


Fig. 1. The $^4I_{9/2}$ and $^4F_{3/2}$ energy levels of $Nd^{3+}$ in the three crystalline hosts.

The photon echo amplitude in each crystal depended upon both magnetic field and sample temperature. The echoes were largest in $CaWO_4$. They were typically two orders of magnitude smaller in $LaF_3$ and three orders smaller in YAG. Photon echoes in YAG, although considerably more difficult to observe than in the other two crystals, were still easily observable (see fig. 2). In manner similar to that reported for echoes in $CaWO_4:Nd^{3+}$ there is a threshold temperature above which the echo amplitude at fixed excitation-pulse separation decreases rapidly [1]. This behavior was insensitive to the magnitude of the applied magnetic field. The threshold temperature for an excitation-pulse separation of 64 nsec was

Fig. 2. Photon echo in YAG:$Nd^{3+}$. Time scale is 50 ns/div.

~ 5°K for $LaF_3$, ~ 8°K for $CaWO_4$, and ~ 13°K for YAG. We attribute this sharp decay to phonon induced relaxations of Orbach and Raman type [5]. For either mechanism the most important contribution comes from levels closest to the echo levels $Z_1$ and $R_1$, viz. $Z_2$ and/or $R_2$. The Orbach relaxation rate behaves as $\sim \Delta^3 \exp(-\Delta/kT)$, while the Raman relaxation rate follows $\sim \Delta^{-4}T^9$, where $\Delta$ is the energy separation between $R_1$ and $R_2$ or between $Z_1$ and $Z_2$ [5]. We note that the ordering of the observed threshold temperatures follows that of $\Delta$ (see fig. 1 for values of $\Delta$). Our experimental data do not allow us to determine which of the two relaxation mechanisms is responsible for the echo decay.

The magnetic field dependence of the photon echoes was studied for fields between 1 and 3 kG and for a fixed pulse separation of 97 nsec. In $CaWO_4$ the echo signal was independent of magnetic field. In $LaF_3$ we observed a monotonic increase by a factor of ten between 1 and 2 kG, after which the signal appears constant up to 3 kG. In YAG the echo signal was largest at 1 kG and decreased monotonically by a factor of ten to disappear in the noise as the field was increased to 2.5 kG. No echoes were detected between 2.5 and 3 kG. We attribute this behavior to the difference in the magnetic interaction of the $Nd^{3+}$ atom with neighboring paramagnetic nuclei present in the crystals [6]. In $CaWO_4$ only 14% of the tungsten nuclei are magnetic and only weakly so ($\gamma_n/2\pi = 0.175$ MHz/kG). We therefore do not expect any modulation of the echo, since $\gamma_n Ht \ll \pi$ (where $H = 3$ kG and $t =$ 97 nsec pulse separation), in agreement with our observation. For both $LaF_3$:$Nd^{3+}$ and YAG:$Nd^{3+}$ we expect and observe modulation effects. For fluorine $\gamma_n/2\pi = 4.0$ MHz/kG, and in $LaF_3$ there are eleven F nuclei within 3 Å of the $Nd^{3+}$ ion. In YAG the ten nearest Al nuclei ($\gamma_n/2\pi = 1.1$ MHz/kG) are within 4 Å of the $Nd^{3+}$ ion.

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## References

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