PHOTON ECHO MODULATION EFFECTS IN $LaF_3:Pr^{3+}$

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Photon echo modulation effects have been observed in the $^3H_4$–$^3P_0$ transition of $LaF_3:Pr^{3+}$ in an applied magnetic field. Our results are interpreted as being due to the combined effect of the Pr nuclear Zeeman and hyperfine interaction.

We have observed strong photon echo modulation effects in the $^3H_4 \leftrightarrow {}^3P_0$ transition of LaF_3 doped with Pr^{3+}. We used excitation pulse separation times of 97 nsec and 260 nsec and we applied magnetic fields of up to 3.95 kG. Our results are shown in figs. 1 and 2. Previous work by Takeuchi and Szabo [1] which reported the observation of photon echoes in this transition did not find any evidence of modulation effects. Their experimental observation is consistent with ours however, since their excitation pulse separation was only 38 nsec and the maximum magnetic field they applied was only 500 gauss.

Our experimental apparatus is similar to that described by Takeuchi and Szabo. Our sample was a LaF_3 crystal doped with 0.1 atomic percent Pr^{3+}. The magnetic field was applied along the crystal *c*-axis ($\hat{z}$ axis) which is normal to the two fold symmetry axis ($\hat{x}$ axis) at the Pr^{3+} site. The 10 kW 4777 Å nitrogen-pumped dye-laser pulse was linearly polarized and directed normal to the crystal *c*-axis. We used a white cell as an optical delay line.

The terminal levels of the echo transition are mixed and split by the hyperfine interaction, the nuclear Zeeman interaction and the interaction of the praseodymium ion with the neighboring fluorine nuclear magnetic moments. Photon echoes arising from these coherently excited multilevel systems exhibit modulation effects the frequency of which are determined by the energy splittings within the multiplets. Strong modulation effects occur when the excitation pulses are able to coherently couple more than two terminal levels with comparable transition probabilities.

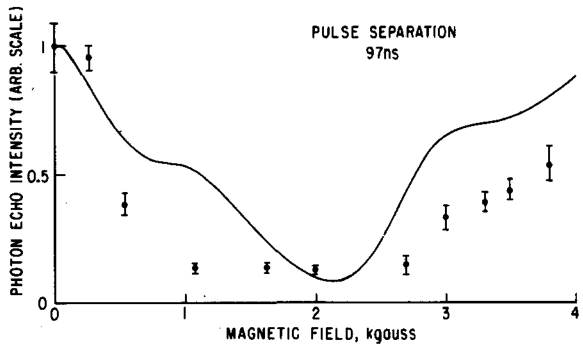

Fig. 1. The photon echo intensity is plotted versus the magnitude of the applied magnetic field when the excitation pulse separation is 97 nsec.

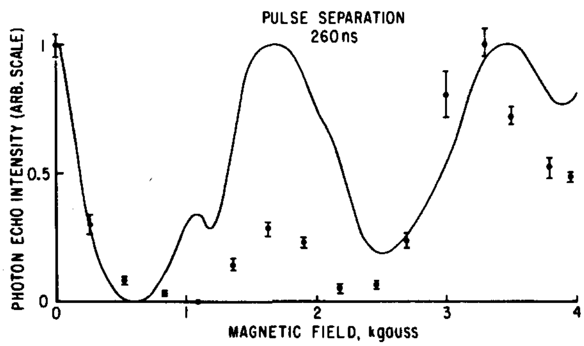

Fig. 2. The photon echo intensity is plotted versus the magnitude of the applied magnetic field when the excitation pulse separation is 260 nsec.

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The site symmetry of the praseodymium ions in LaF_3 is C_{2v} or C_2 [2]. This precludes the possibility of there being an electronic moment associated with the electronic levels. The lack of such an electronic moment would eliminate the $F-Pr^{3+}$ magnetic interaction, however in the presence of an external field an induced moment may appear because of electronic level mixing. From the measured magnetic susceptibility of PrF_3 [3] whose crystal field is close to that of $LaF_3:Pr^{3+}$ [4] the average induced dipole moment of the praseodymium ion is estimated to be about 3×10^{-3} Bohr magneton per kiloGauss. But unless the induced dipole moment is of the order of one Bohr magneton, the significant degradation of the echoes shown in fig. 1 and fig. 2 at 1 kG would be unexplained on the basis of $F-Pr^{3+}$ interaction.

We believe the echo modulation is due to the interference among electronic levels split and mixed by the interaction with the praseodymium nucleus. Although the first order effect of the nuclear hyperfine interaction of Pr^{3+} in LaF_3 is zero because of the zero electronic moment, the second order effect is appreciable. The praseodymium nuclear state degeneracies are lifted by the combination effect of the nuclear Zeeman interaction and the second order hyperfine interaction [5, 6]. For the $^3P_0$ level where the electronic state mixing is negligible, the nuclear levels are split by the external field into six equally spaced levels with an energy separation of 1.1 MHz per kilo gauss between them. They are characterized by the z-component of the nuclear spin quantum number m_I with m_I ranging from $-\frac{5}{2}$ to $+\frac{5}{2}$. For the $^3H_4$ levels, the splitting caused by the external field is enhanced because the induced magnetic moment couples with the nuclear spin through the hyperfine interaction. The magnitude of the enhancement varies with direction. The average value is about five [7]. When the external field is parallel to the crystal c axis, the effective field at the nucleus is also parallel to the applied field. In this case state mixing is large and is caused by the second order hyperfine interaction which has the form $\Sigma_{i=x,y,z} P_i I_i^2$ where P_i's are dependent on direction and the hyperfine interaction constant [6]. Using the formula given in eq. (9) of ref. [6], the average value of P_i's is estimated to be about 2 MHz based on susceptibility measurements [3]. Since the second order hyperfine interaction connects the nuclear state m_I with the state $m_I \pm 2$, the nuclear wave function associated with the $^3H_4$ level must be of the form $a|\pm\frac{5}{2}\rangle + b|\pm\frac{1}{2}\rangle + c|\pm\frac{3}{2}\rangle$. The selection rule for the electric dipole transition is $\Delta m_I = 0$. The excitation pulses are therefore able to connect every substate in the ground multiplet with three substates in the excited multiplet and vice versa. Since the second order hyperfine interaction is estimated to be comparable in strength with the nuclear Zeeman effect, a thorough mixing of states is expected and a significant echo modulation effect becomes possible. The appearance of echo intensity maxima at 1.7 kG and 3.3 kG in fig. 2 may come from the beating effect of the transition from one of the ground states to either $\{|\frac{5}{2}\rangle, |\frac{1}{2}\rangle, |-\frac{3}{2}\rangle\}$ or $\{|-\frac{5}{2}\rangle, |-\frac{1}{2}\rangle, |\frac{3}{2}\rangle\}$ in the $^3P_0$ multiplet. An accurate analysis is difficult without knowledge of the crystal field parameters. A rough calculation [8] yields the solid curves in fig. 1 and fig. 2 which mirror the general features of the data. Since the magnetic susceptibility for the crystal of LaF_3 was believed to be anisotropic, we used $\chi_{xx} = 0.034$ and $\chi_{yy} = \chi_{zz}$ with the restriction that the average value of χ equals the measured value 0.016. Using these parameters the corresponding calculation for the Takeuchi-Szabo experiment explains their null result since the computed echo degradation under their conditions is less than 8%.

A second effect of the second order hyperfine interaction estimated above would be to distribute, even in zero field, the ground state levels over a 12 MHz interval. This may explain the anomalous ~10 MHz width of the transition $^1D_2 \rightarrow {}^3H_4$ observed by Erickson in his fluorescence line narrowing experiment [9].

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