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10 HERTZ OPTICAL RESONANCE LINEWIDTHS IN Cr:YAlO₃

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We have observed the nearly degenerate four-wave mixing (NDFWM) spectrum of Cr:YAlO₃ using a new technique to eliminate the effects of laser jitter. The optical emission resonance at room temperature has a Lorentzian linewidth of 9.8 Hz, in excellent agreement with the measured longitudinal relaxation rate of 9.6 Hz on the ²E - ⁴A transition of Cr, determined by fluorescence decay.

I - INTRODUCTION

Nonlinear laser spectroscopy based on four-wave mixing permits state-specific spectroscopy and the measurement of important relaxation rates as has been demonstrated in atomic sodium¹. In gaseous systems it is possible with this approach to vary the pressure of buffer gas and follow the evolution of a closed quantum mechanical system into an open system². In the previous experimental work, measurements were instrument limited by laser frequency jitter. Here we present a new nearly degenerate four-wave mixing (NDFWM) technique to eliminate the adverse effects of laser frequency jitter and measure the frequency-domain spectrum of the backward wave resonance in a crystal of Cr:YAlO₃. An emission resonance linewidth of 9.8 Hertz (FWHM) is measured at room temperature. Quantitative agreement is obtained with a density matrix calculation and independent fluorescent decay measurements which show that the Lorentzian linewidth is determined by ground state relaxation of Cr³⁺ ions involving a single process: fluorescent decay of the ²E metastable level.

II - EXPERIMENT

NDFWM emission resonances arise from the third order nonlinear optical interaction³ of two counter propagating pump beams at frequency ω and an independently tunable probe beam at frequency $\omega + \delta$. Energy conservation gives the signal frequency at $\omega - \delta$. Phase matching insures that if the two pumps are counter propagating, then the signal wave is counter propagating with respect to the probe beam. For our measurements the resonance of interest occurs at $\delta = 0$ and is observed by

sweeping δ and observing the backward wave signal. The nonlinearity responsible for the mixing is due to spatial hole burning in the Cr^{3+} doped materials. In this paper, we demonstrate that if the two frequencies ω and $\omega + \delta$ are derived from the same source, then the source jitter does not contribute to the observed linewidth.

Our measurements were performed using a 3.9 mm thick sample of room temperature Cr: YAlO_3 doped with approximately 0.05% Cr and exhibiting an optical density of 0.29 at 570 nm. The crystal was positioned with the c-axis parallel to the two counter propagating pump beams. The experimental configuration is shown in Figure 1. The probe beam was nearly colinear with the forward pump beam and intersected the pump beams in the sample at an angle of 0.3° .

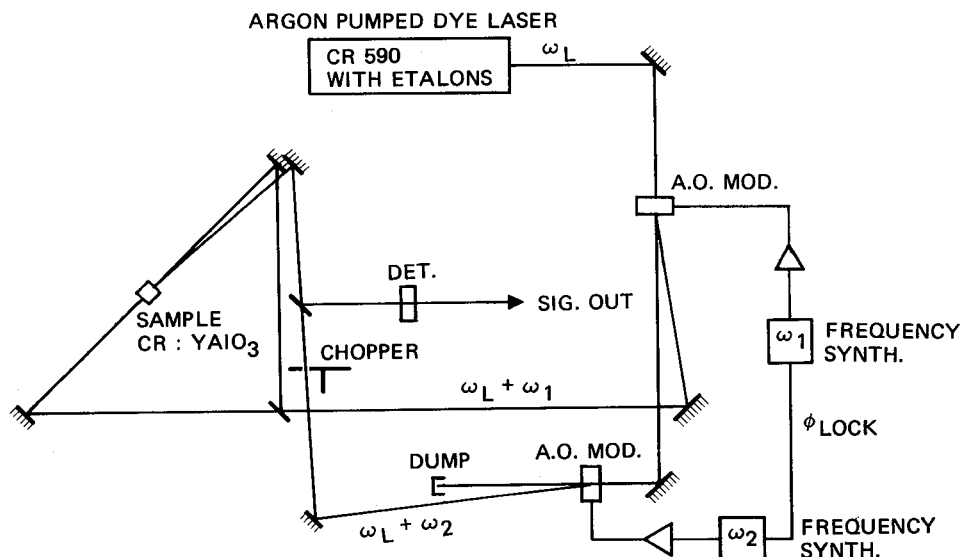


Figure 1. Experimental configuration for NDFWM using synthesized Frequencies for the three input beams.

As shown in the figure, two acousto-optic modulators were used to synthesize the appropriate wavelengths for the pump and probe beams. The intensities were in the range of 1-20 W/cm^2 . The measurements were confirmed to be independent of pump intensity. The acousto-optic modulators were driven by two frequency synthesizers at ω_1 and ω_2 respectively. The two synthesizers were phase locked together and stable to better than one Hertz throughout the duration of each experiment. Typically, ω_1 was held fixed at 40 MHz, and ω_2 was scanned over a 200 Hertz range centered at 40 MHz. The fundamental laser frequency was tuned near 570 nm for peak signal from the sample. As we show below, the 40 MHz time averaged bandwidth of this laser does not contribute to the observed linewidth as long as the jitter is small compared to the absorption linewidth of the sample. This is certainly the case in Cr: YAlO_3 where the $4A-4T$ absorption profile is homogeneously broadened with a width of approximately 65 nm.

Figure 2 shows a 200 Hertz scan over the resonance. The smooth curve is the result of a least squares fit of a Lorentzian. The observed width of 9.8 Hertz (FWHM) is not instrument limited since measurements on BaTiO_3 have shown intensity dependent widths as small as 2.5 Hertz.

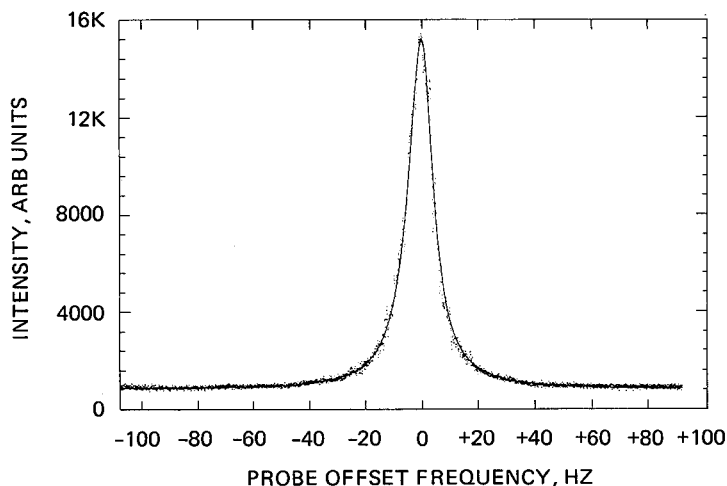


Figure 2. NDFWM signal as a function of pump-probe detuning, δ .

III - DISCUSSION

To interpret the spectrum, we consider a three level model for Cr:YAlO₃ and the four-wave mixing geometry shown in Figure 3. The frequencies of the input optical beams (two pump beams, E_f and E_b , and the probe beam, E_p) are assumed to be close to the resonant frequency ω_0 . A chromium atom excited from the ground state, $|1\rangle$, to the excited state, $|2\rangle$, can decay by spontaneous emission back to the ground state. State $|2\rangle$ can also decay by a nonradiative transition to the metastable state $|3\rangle$. This state decays by fluorescence back to the ground state, emitting a photon in the region of 694 nm.

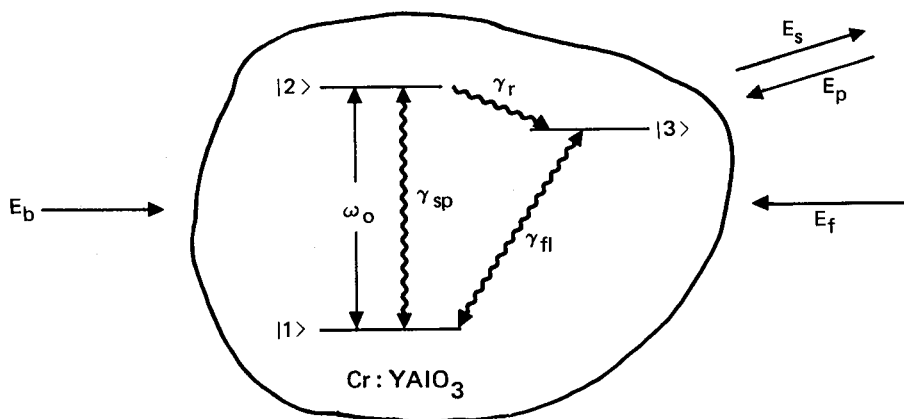


Figure 3. Three level model for Cr:YAlO₃ and geometry for calculating NDFWM response.

The optical polarization is calculated from $P = \text{Tr } \rho$ where ρ is third order in the electromagnetic fields. We make the rotating wave approximation and keep only those terms which are phase matched in the direction of interest: i.e., k_s , the signal wave vector, is given such that $k_f + k_p - k_s \sim 0$. The polarization of interest is given by

$$P = 2\mu_{12} \rho_{11}^{(0)} \Omega_f \Omega_b \Omega_p^* e^{i(\omega - \delta)t + ik_p x} \\ \times \left(\frac{1}{\delta + i\gamma_T} + \frac{1}{\gamma_T - \gamma_{f1}} \left(\frac{\gamma_r}{\delta + i\gamma_{f1}} + \frac{\gamma_{sp} - \gamma_{f1}}{\delta + i\gamma_T} \right) \right) \\ \times \left\{ \frac{1}{-\Delta + \delta + i\gamma_{ph}} \left(\frac{1}{-\Delta - \delta - i\gamma_{ph}} - \frac{1}{-\Delta + i\gamma_{ph}} \right) \right\}$$

where $\Omega_i = \mu_{12} E_i / 2\hbar$, $\Delta = \omega_1 - \omega_0$, $\delta = \omega_2 - \omega_1$, $\gamma_T = \gamma_{sp} + \gamma_r$, and γ_{ph} represents dephasing due to the crystal and is related to the absorption linewidth (for a Lorentzian profile) by $1/\pi\gamma_{ph}$ (FWHM in Hertz).

In spectroscopy by NDFWM, the signal wave, E_s , arises from a coupled mode analysis between the probe wave and the signal wave. In the limit of no pump depletion (weak signal limit) the signal intensity ($\approx E_s^2$) is proportional to the magnitude squared of the polarization. This leads to a resonant behavior of the signal intensity described by

$$\frac{1}{\delta^2 + \gamma_{f1}^2} \frac{\gamma_r^2}{(\gamma_T - \gamma_{f1})^2} + \frac{1}{\delta^2 + \gamma_T^2} \left(1 + \frac{\gamma_{sp} - \gamma_{f1}}{\gamma_T - \gamma_{f1}} \right)^2 \\ + \frac{2\gamma_r}{\gamma_T - \gamma_{f1}} \left(1 + \frac{\gamma_{sp} - \gamma_{f1}}{\gamma_T - \gamma_{f1}} \right) \frac{1}{\delta^2 + \gamma_{f1}\gamma_T + \frac{\delta^2}{\delta^2 + \gamma_{f1}\gamma_T} (\gamma_T - \gamma_{f1})^2}$$

In this expression, we have assumed that $\gamma_{ph} \gg \gamma_{f1}$, γ_T and ignore resonances due to $\Delta \pm \delta = 0$. The only term that contributes when $\gamma_T = \gamma_{sp} + \gamma_r \gg \gamma_{f1}$ is the first Lorentzian, proportional to $(\delta^2 + \gamma_{f1}^2)^{-1}$.

This resonant term originates explicitly in the equation of motion for the second order contribution to the ground state term, $\rho_{11}^{(2)}$. It contains a contribution from the fluorescent decay of the metastable state, ρ_{33} , generated by nonradiative transitions from the upper laser excited population, ρ_{22} . This term describes the relaxation of the ground state and in general is in contrast to a time domain measurement of fluorescent decay which measures the relaxation time of ρ_{33} . However, in the three level system considered here, the frequency domain measurement of the third order polarization does measure the same parameter as the time domain measurement of fluorescence. It is clear from the expression given above for the polarization that, these two measurements coincide in this case only because of the relative magnitude of the different decay rates. To confirm this interpretation, a separate measurement of the fluorescent decay rate was made and shown to be $\gamma_{f1}^{-1} = 33 \pm 1 \text{ msec}$ corresponding to a linewidth of 9.65 Hertz, in excellent agreement with the observed linewidth of 9.8 Hertz. In ruby also, the fluorescent decay rate

$\gamma_{f1}^{-1} = 2.6\text{ms}$ agreed well with the observed NDFWM spectral width of 138 Hertz.

From the above data, it is clear that this new spectroscopy technique is not sensitive to laser jitter. This was demonstrated earlier in a Ramsey fringe experiment with an atomic beam⁴, but the ability to observe such narrow resonances was limited by transit time effects. It is our expectation that laser jitter can be eliminated in any NDFWM spectroscopy where the resonance occurs in the second order contribution to the third order nonlinear optical polarization. This will prove very useful in ultrahigh resolution frequency domain spectroscopy studies of inverted V type transitions where the linewidth is determined by the relaxation between states of identical parity. It may also be possible to adapt the results described here for integrated atomic clock applications.

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