

Doubly Resonant Four-Photon Interactions in Cesium Vapor

ARLEE V. SMITH AND J. F. WARD

Abstract—Four-photon interactions involving both a two-photon resonance with the $9d^2D_{3/2}$ level and a one-photon resonance with an np^2P level ($n = 7-10$) are investigated in detail. Gain, gain saturation, gain threshold, and forward-to-backward generation ratios are measured. Comparison with numerical estimates indicates that $9d^2D_{3/2}$ to np^2P lasing is the dominant gain mechanism but that a doubly resonant four-photon parametric oscillation makes a significant contribution. Gain saturation is dominated by population of np^2P levels. Ionization is important, especially at higher pump powers.

WE wish to report detailed observations of a process in which pairs of optical fields with frequencies ω_S and ω_I (referred to here as signal and idler, respectively) are generated in cesium vapor irradiated by a ruby laser pump (ω_P). The frequencies are related by

$$\omega_S + \omega_I = 2\omega_P \quad (1)$$

and are near resonance with transitions in atomic cesium:

$$2\hbar\omega_P \approx E(9d^2D_{3/2}) - E(6s^2S_{1/2}) \quad (2a)$$

$$\hbar\omega_{S,n} \approx E(np^2P) - E(6s^2S_{1/2}) \quad (2b)$$

$$\hbar\omega_{I,n} \approx E(9d^2D_{3/2}) - E(np^2P) \quad (2c)$$

where n ($10 \geq n \geq 6$) is the principle quantum number of a P level lower in energy than the $9D_{3/2}$ level.

A number of authors have reported that metal vapors irradiated by one or two powerful lasers with frequencies nearly resonant with one- or two-photon transitions generate intense signals colinear with the incident laser beams [1]. In several instances it is demonstrated that the process responsible for the generation of these signals is electronic stimulated Raman scattering or stimulated emission from an induced population inversion in the vapor, perhaps in conjunction with nonlinear mixing with the incident laser fields. In other cases the origin of the signals is uncertain. Related theoretical contributions may be found in [2] and [3]. Penzkofer *et al.* [4] observed a similar effect in water and present evidence that a single-photon resonant, four-wave parametric process is involved.

Population of the $9D_{3/2}$ level of cesium by two-photon absorption was first demonstrated by Abella [5]. We have previously studied several processes in cesium excited near this two-photon resonance (enhanced third-harmonic genera-

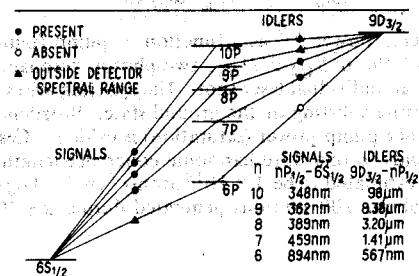


Fig. 1. Relevant cesium energy levels, signal and idler transitions.

tion [6], fluorescence [6], and saturation of these processes [7]). The process discussed here also relies on this resonance as indicated in (2a).

We observe the generation of as much as 3 W of signal power (P^S) with components at $\omega_{S,n}$ ($n = 10, 9, 8, 7$), and 100 mW of idler power (P^I) with components at $\omega_{I,n}$ ($n = 8, 7$). There is no radiation at $\omega_{I,6}$ and components $\omega_{S,6}$, $\omega_{I,10}$, and $\omega_{I,9}$ were outside the spectral range of the detectors used. Signal and idler transitions are shown together with relevant atomic cesium energy levels in Fig. 1. We have observed the variation of signal power over nine decades as a function of pump power (P^P) and cesium density, and have measured thresholds, small-signal gains, and forward-to-backward generation ratios. Saturation behavior and resonant dependence on pump frequency ω_P have also been studied. The small-signal 348 nm ($n = 10$) gain indicates that if saturation were absent, complete conversion to 348 nm would be achieved with 1 MW of pump power. This may be compared with the factor of 10^{10} attenuation calculated for a weak 348 nm beam passing through the cell in the absence of the pump beam.

The experimental arrangement is similar to that described previously [7]. A ruby laser produces a beam with 1 MW peak power and 0.03 cm^{-1} bandwidth tunable in the region of 693.5 nm. The collimated beam (area $A = 0.03 \text{ cm}^2$) propagates through approximately 5 cm of cesium vapor at densities up to 10^{16} cm^{-3} (at 510 K). The cesium cell also contains 1 torr of neon buffer gas. The signal and idler beams are generated colinear with the pump beam, all beams are of comparable diameter, and no evidence of self-focusing is observed. The depletion of laser beam power going through the cesium cell is less than 1 percent.

The resonant response of the 348 nm, $n = 10$ signal as a function of pump frequency is shown in Fig. 2 at two pump powers. The splitting of the resonance is caused by the hyperfine splitting of the atomic cesium ground state, and broadening of the resonance at the higher pump power is clearly seen. In data shown subsequently, the pump frequency has been set

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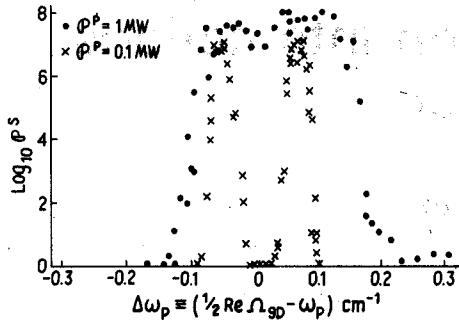


Fig. 2. 348 nm signal power \mathcal{P}^S as a function of pump frequency ω_P in the vicinity of the $6^2S_{1/2}-9^2D_{3/2}$, two-photon resonance. Each point represents an individual laser shot. The splitting arises from the 0.31 cm^{-1} hyperfine splitting of the ground state. Broadening of the resonance at higher pump power (saturation) is evident. Cesium density ρ_0 corresponds to one third-harmonic coherence length of vapor in the cell and is estimated to be $4 \times 10^{15} \text{ atoms} \cdot \text{cm}^{-3}$. $\text{Log}_{10} \mathcal{P}^S = 0$ corresponds to about 100 photons generated during the 20 ns laser pulse.

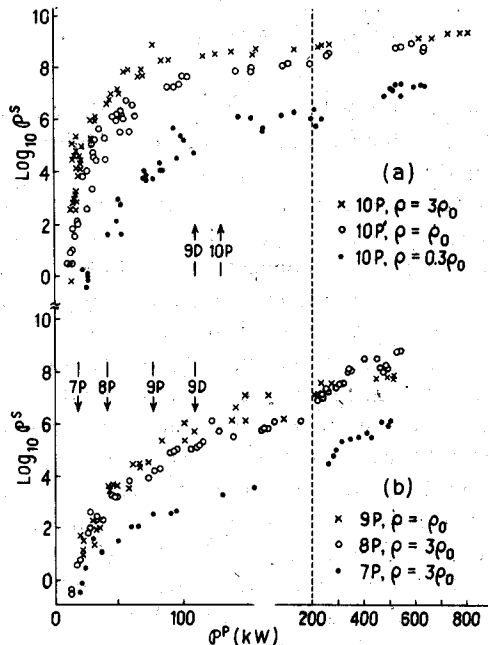


Fig. 3. Signal power, \mathcal{P}^S for various signal components at various cesium densities as a function of pump power, \mathcal{P}^P . Note the change in pump power scale at 200 kW. Arrows indicate the pump power at which there is a 50 percent probability of ionization from each relevant level during the 20 ns laser pulse. ρ_0 and $\text{log}_{10} \mathcal{P}^S = 0$ are defined in the caption to Fig. 2.

to resonance with the $F=4$ hyperfine component (data for $F=3$ are similar). Spectral resolution of the signal wave in these experiments is limited by the quartz prism monochromator used to about 100 \AA . This is sufficient to distinguish signals associated with P levels of different principal quantum numbers, but not for distinguishing between components related to the $P_{1/2}$ and $P_{3/2}$ levels, and details of line shape are not observed.

Fig. 3 shows the dependence of signal power on pump power for several signal components at various cesium vapor densities. Fig. 4 shows the dependence of signal power on cesium density for $n=10$ and 8 signal components at $\mathcal{P}^P = 1 \text{ MW}$. Signal powers are observed over a range of nine decades from a minimum detectable signal of approximately 100 photons up to several watts. Low-signal exponential gain is seen in

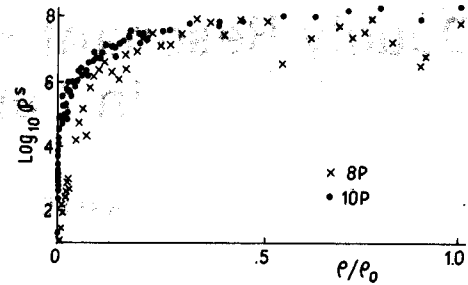


Fig. 4. Signal power \mathcal{P}^S as a function of cesium density ρ for $n=10$ and 8 components. ρ_0 and $\text{log}_{10} \mathcal{P}^S = 0$ are defined in the caption to Fig. 2.

each instance, with gain saturation at high-signal power. There is a pump-power threshold (Fig. 3) but no density threshold (Fig. 4) for signal generation.

As a basis for discussion of the data, we now present a theory similar to that of Yariv and Pearson [3] and Penzkofer *et al.* [4]. Consider two mixing processes in which polarizations P are produced in an atomic vapor at ω_S and ω_I , respectively:

$$\begin{aligned} P_y^{\omega S} &= \rho \frac{3}{4} \langle \chi_{yyyy}(-\omega_S; \omega_P, \omega_P, -\omega_I) \rangle E_y^{\omega P} E_y^{\omega P} E_y^{\omega I} \\ &= \rho \frac{3}{4} \langle \chi^S \rangle E_y^{\omega P} E_y^{\omega P} E_y^{\omega I} \end{aligned} \quad (3a)$$

and

$$\begin{aligned} P_y^{-\omega S} &= \rho \frac{3}{4} \langle \chi_{yyyy}^*(-\omega_I; \omega_P, \omega_P, -\omega_S) \rangle E_y^{\omega P} E_y^{\omega P} E_y^{\omega S} \\ &= \rho \frac{3}{4} \langle \chi^{I*} \rangle E_y^{\omega P} E_y^{\omega P} E_y^{\omega S} \end{aligned} \quad (3b)$$

where E is an electric field Fourier amplitude at a frequency indicated by the superscript, ρ is the atomic number density, $\langle \rangle$ indicates an average over the atomic velocity distribution [8], and the χ are atomic nonlinear polarizabilities [9] which exhibit double resonances:

$$\chi^S \propto (\Omega_{9D} - 2\omega_P)^{-1} (\Omega_{nP} - \omega_S)^{-1} \quad (4a)$$

$$\chi^{I*} \propto (\Omega_{9D}^* - 2\omega_P)^{-1} (\Omega_{nP} - \omega_S)^{-1} \quad (4b)$$

where $\hbar\Omega_i$ is the complex energy of the ground to i level transition. Complex wave vectors may be used to describe the propagation of the signal and idler fields:

$$k = k' + ik'' \quad (5)$$

and a mismatch wave vector is defined as

$$\Delta k = 2k'_P - k'_S - ik''_S - k'_I + ik''_I. \quad (6)$$

k''_P is taken to be zero and k''_S and k''_I describe loss or gain for the signal and idler fields. The signal field experiences linear loss due to one-photon absorption. The idler may exhibit lasing gain due to $9D_{3/2} - nP$ population inversion or gain due to the Raman process with coefficient $\chi(-\omega_I, \omega_I, \omega_P, \omega_P, -\omega_P, -\omega_P)$, either possibility being included by taking k''_I negative and dependent on pump power. Of these two mechanisms, we refer subsequently to lasing, but the Raman process is qualitatively similar. Coupled plane wave equations in which the polarizations of (3a) and (3b) serve as source terms can be solved [3], [4] with the assumption of slowly varying amplitudes and negligible pump depletion. Both signal and idler waves have contributions with exponential gain

$$\mathcal{P}^S = \text{const} \times \exp[\gamma z] \quad (7)$$

TABLE I

OBSERVED AND CALCULATED GAINS, THRESHOLDS, AND SIGNAL POWERS, \mathcal{P}^S , FOR PROCESSES INVOLVING VARIOUS nP LEVELS ($n = 6-10$). OBSERVED GAIN DERIVED FROM THE EXPONENTIAL GAIN REGION OF FIG. 3 IS $10\delta (\log_{10} \mathcal{P}^S) / \partial \mathcal{P}^P |_{\rho=\rho_0}$ dB/MW AT CESIUM DENSITY ρ_0 DEFINED IN THE CAPTION TO FIG. 2. GAINS HAVE BEEN CALCULATED AS DISCUSSED IN THE TEXT. CASE 1) IS AN UPPER LIMIT FOR LASING GAIN AND CASE 2) IS PARAMETRIC GAIN. OBSERVED THRESHOLDS ARE FROM FIG. 3 AND THRESHOLDS ARE CALCULATED BY EXTRAPOLATING THE EXPONENTIAL GAIN REGION FOR PARAMETRIC OSCILLATION TO ZERO GAIN. $\mathcal{P}^S_{\text{obs}}$ IS THE MAXIMUM OBSERVED SIGNAL POWER.

n	Gain dB			Threshold kW		$\mathcal{P}^S_{\text{obs. W}}$
	Obs.	Calc. (1)	Calc. (2)	Obs.	Calc. (2)	
10	2000	3×10^7	600	15	38	2.2
9	1000	2×10^6	570	15	102	0.6
8	700	1×10^5	740	15	373	0.14
7	400	2×10^4	1800	15	1700	2×10^{-4}
6	0	5×10^3	31 000	—	25 700	0

where γ is given by

$$\gamma = 2 \operatorname{Re} \{G - (\Delta k/2)^2\}^{1/2} - \{k_S'' + k_I''\} \quad (8)$$

and

$$G = \frac{2^4 \cdot 3^2 \cdot \pi^2}{c^6} \cdot \frac{\omega_S^2 \omega_I^2}{k_S k_I} (\rho \mathcal{P}^P / A^2) \langle \chi_S \rangle \langle \chi_I^* \rangle. \quad (9)$$

Two extreme cases may be distinguished.

1) *Lasing*: $-k_I'' \gg \sqrt{G}$: The gain is given by

$$\gamma z = -2k_I'' z + \operatorname{Re} [2G/(k_S'' - k_I'')] + \dots \quad (10)$$

and there is no threshold included in the model for this case. In this process the idler gain derives from lasing ($9D_{3/2} \rightarrow nP$) represented by the negative k_I'' and the signal is generated subsequently by mixing idler and pump waves. We will refer to this process as lasing or lasing plus mixing, reserving the term parametric for case 2).

2) *Parametric*: $\sqrt{G} \gg \Delta k, k_I''$: The gain is

$$\gamma z = \operatorname{Re} 2\sqrt{G} z - (k_S'' + k_I'') + \dots \quad (11)$$

and although the dependence of the gain on pump power at low power is complicated, one can extrapolate back to a "threshold" value of \mathcal{P}^P determined by

$$\operatorname{Re} (2\sqrt{G}) = k_S''. \quad (12)$$

This process may be called doubly resonant four-photon parametric oscillation.

With the help of numerical estimates [10] we now compare this model with the data.

Table I shows observed gain ($10\delta \log_{10} \mathcal{P}^S / \partial \mathcal{P}^P |_{\rho=\rho_0}$) and threshold derived from data such as that shown in Fig. 3. Gains and thresholds for the parametric process have been calculated from (7)–(9), (11), and (12) using cesium wavefunction data from the literature [11], and values for processes involving $P_{1/2}$ levels are shown in Table I (for $P_{3/2}$ levels, gains are lower and thresholds higher). Calculated values indicate that the parametric process does not have a sufficiently low threshold, nor in the case of $n = 10$ and 9 can it provide sufficient gain to account for the observations. Lasing gain can be calculated from (7), (8), and (10), knowing, from fluorescence measurements [7], that the $9D_{3/2}$ population is linear

in \mathcal{P}^P under these conditions (rather than quadratic as would be expected for low pump powers). An upper limit given in Table I is calculated on the basis that at $\mathcal{P}^P = 1$ MW, half the ground $F = 4$ hyperfine state population is excited to the $9D_{3/2}$ level and all P levels are empty. Observed gains are well within the calculated lasing gains and we believe that at threshold the lasing process is dominant. The observed threshold is not included in the model but could arise from a loss mechanism such as molecular absorption.

A comparison of observed and calculated gain and threshold data in Table I leads to the conclusion that the parametric process should make a relatively large contribution for $n = 8$ at high pump power. Here the predicted parametric threshold is exceeded and the predicted parametric gain is comparable with that observed. Measurement of the power in the forward propagating idler beam (i.e., propagating in the same direction as the pump beam) relative to that in the backward propagating idler beam provides a sensitive test of the relative contribution of the parametric and lasing-gain processes. Lasing is expected to produce idler beams propagating forward and backward with equal power, whereas the parametric process is limited by index matching to produce idler in the forward direction only. The measured forward-to-backward ratio (for $n = 8$, $\mathcal{P}^P = 1$ MW, $\rho = \rho_0$) is 7 ± 3 . Deriving the parametric contribution to the gain from this measurement is complicated by saturation and by the details of the frequency dependences of the gain processes. However, using (10), we deduce that this contribution lies in the range of 100–700 dB where the higher figure is comparable with the theoretical prediction shown in Table I. In any case it is clear that the contribution of the parametric process is significant here, but not dominant. The signal beam, whether produced by the parametric process or by mixing of the pump with laser-produced idler, is constrained by index matching to the forward direction, and in neither case have we detected a measurable backward propagating signal.

Signal power as a function of cesium density, as presented for $n = 8$ and 10 in Fig. 4, exhibits no discernable threshold. This is to be expected since all terms in (8) are density dependent. The density gain ($10\delta \log_{10} \mathcal{P}^S / \partial \rho |_{\mathcal{P}^P=1\text{MW}}$) derived from the exponential gain region of Fig. 4 is found to be a factor 3–4 times larger than the power gain ($10\delta \log_{10} \mathcal{P}^S / \partial \mathcal{P}^P |_{\rho=\rho_0}$) discussed above. If all gain terms in (8) (except k_S'') are linear in both \mathcal{P}^P and ρ as we have supposed, both these measures of the gain would be equal. Inclusion of a pump-power threshold requires that the density gain be *smaller* than the power gain. The contrary observation could arise from a less than linear dependence of gain on density. Level changing collisions [13] occur at rates slower than 10^7 s^{-1} and are unimportant during the 20 ns laser pulse. The effect of collision on T_2 for the $9D_{3/2}$ level could well change the lasing gain sufficiently to account for density gain exceeding the power gain.

Gain saturation is evident in Figs. 3 and 4. It is observed that the onset of saturation depends not on \mathcal{P}^P or ρ alone, but occurs at a certain fixed value of \mathcal{P}^S (or \mathcal{P}^I). Saturation by two-photon population of the $9D_{3/2}$ level would depend on \mathcal{P}^P but be independent of ρ . The important saturation mechanism here is, therefore, population of the P levels rather than of the $9D_{3/2}$ level. The P levels may be populated either by stimulated transitions from the $9D_{3/2}$ level or by absorption

of signal photons by ground state atoms. We believe the first of these mechanisms is the important one because observed signal powers (see Table I) are inadequate by more than a factor of 100 to saturate the ground to P level transition.

In the previous discussion we have ignored photoionization [12], which, for $\mathcal{P}^P = 1$ MW, occurs at rates in the range of 0.2–1.5 ns⁻¹ for single-photon ionization from $10P_{1/2}$ – $7P_{1/2}$, 0.25 ns⁻¹ for $9D_{3/2}$, and which does not occur for $6P$. Photoionization is expected to have several effects: populations of nP ($n \geq 7$) and $9D_{3/2}$ levels are reduced, thus reducing the tendency to saturate; reduced lifetimes introduce extra damping of the resonances and depletion of neutral cesium atoms reduces the effectiveness of all the resonant interactions considered. Arrows in Fig. 3 indicate, for the $9D_{3/2}$ and each P level, the pump power at which there is a 50 percent probability of photoionization during the laser pulse. For $n = 10$ in the exponential gain region and through the onset of saturation, photoionization is significant but not dominant; for $n = 7$, on the other hand, the effect is very important throughout the relevant range of pump power, and the slow onset of gain saturation for $n = 7$ (see Fig. 3) may be attributable to photoionization. At $\mathcal{P}^P = 1$ MW, photoionization dominates all processes. It is interesting to note that direct photoionization from the $9D_{3/2}$ level may proceed at a slower rate than the two-step process of stimulated transition from $9D_{3/2}$ to nP levels followed by photoionization.

In conclusion, we have presented detailed data on doubly resonant four-photon processes in cesium involving the $9D_{3/2}$ and nP levels ($n = 10, 9, 8, 7, 6$). We find that lasing followed by mixing of idler with pump to produce the signal is the dominant process and doubly resonant four-photon parametric oscillation contributes significantly for $n = 8$ at high pump power. Gains are large but saturation at approximately 1 W in the visible (0.1 W in the IR) is caused by population of P levels. Photoionization also plays an important role.

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J. F. Ward, photograph and biography not available at the time of publication.

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