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SATURATION BEHAVIOR OF PARAMETRIC FOUR-WAVE MIXING DUE TO TWO-PHOTON INTERFERENCE EFFECT*

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INTRODUCTION

Strong pumping of "extended" alkali metal vapors near two-photon resonances produces copious stimulated emissions by amplified spontaneous emission (ASE) or stimulated electronic hyper-Raman scattering (SEHRS), and by phase matched parametric four-wave mixing (PFWM). The new electromagnetic fields that are generated within the near-resonant nonlinear medium can strongly influence the total atomic response through processes including interferences, level shifting, and population transfer mechanisms. The internally generated fields can, e.g., suppress transition probabilities for processes involving excitations (SEHRS and two-photon excitations) and produce gain saturation for PFWM processes. Recent experiments have demonstrated that in two-photon resonant pumping of $S \rightarrow D$ and $S \rightarrow S$ transitions in Na, the excitation rate is strongly reduced through a two-photon interference effect involving PFWM.[1-5] In this process, two-photon pumping from the ground state, $|0>$, to a two-photon level, $|2>$, by laser photons ω_1 and ω_2 with two-photon Rabi frequency $\Omega_{02}^{(2)}$ produces two new frequencies ω_3 and ω_4 through PFWM. These generated fields create a second two-photon Rabi rate $\tilde{\Omega}_{02}^{(2)}$ between the same two states. Under phase matched PFWM generation, the second pathway destructively interferes with the first;[1,2,6,7] that is, in the equations of motion the two-photon pumping rate by the laser field, $\Omega_{02}^{(2)}$, is cancelled off by the two-photon rate $\tilde{\Omega}_{02}^{(2)}$ due to PFWM which becomes equal in magnitude but opposite in sign

$$\Omega_{02}^{(2)} = -\tilde{\Omega}_{02}^{(2)} \tag{1}$$

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Under these conditions the "effective" two-photon Rabi rate between $|0>$ and $|2>$ goes to zero. Since photons generated through PFWM are almost resonant with intermediate p-states, the two Rabi rates can become equal in magnitude even though the fields involved in $\tilde{\Omega}_{02}^{(2)}$ are fairly weak as compared to the laser field. Manykin and Afanas'ev[6] showed that solutions to the equations of motion always evolve to the destructive interfering phase relationship at zero detuning.

Experimental studies of the interference associated with resonant two-photon pumping in Na vapors have so far involved some way of monitoring the influence of PFWM on the resonant transfer of population to the two-photon resonant state.[1–4] However, the interference effect also has a stong and easily demonstrated influence on PFWM intensities which are produced when tuning near two-photon resonances. If we ignore the fine structure splittings in Na, we can describe the problem by a four-level system as depicted in the insert of Fig. 1. (In the present context we label ω_3 as ω_{IR} and ω_4 as ω_{UV}). We assume the time dependent state vector of an atom at z to be of the form

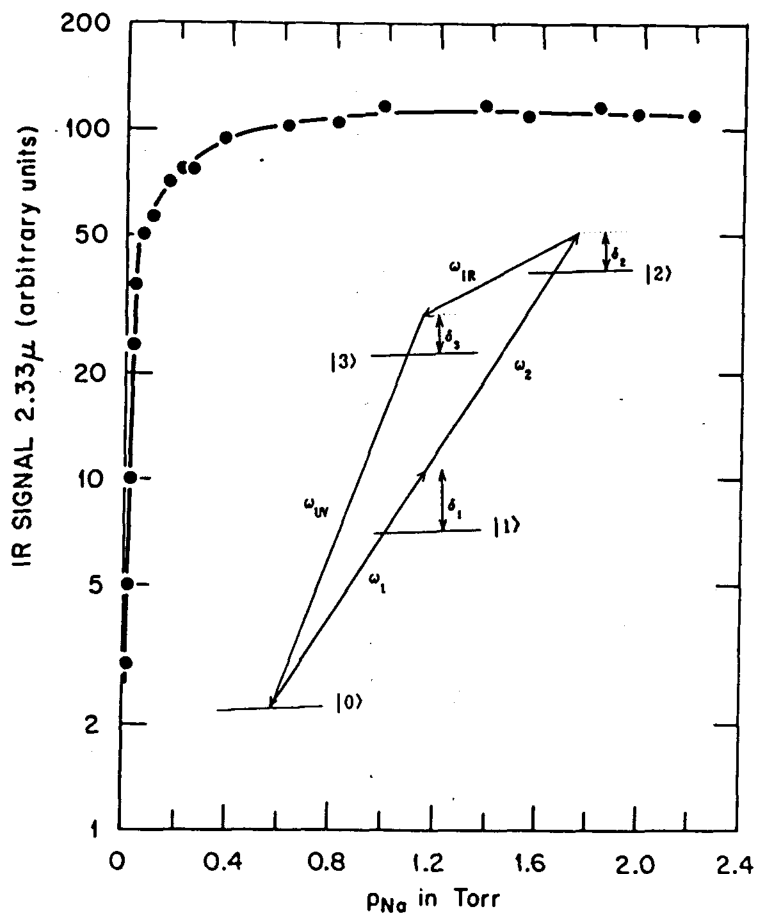

Fig. 1. Intensity of 2.33μ components of PFWM as a function of sodium vapor pressure. Laser tuned 0.3 Å on high energy side $4d_{3/2,5/2}$ resonance. Fine structure components of 4D were unresolved. Infrared signal included components of PFWM, i.e., that near resonance with $4p_{1/2}$ and $4p_{3/2}$ levels $I_L = 34$ MW/cm^2.

$$|\Psi(z,t)\rangle = \sum_{n=0}^{3} a_n(z,t)e^{-i\omega_n t}|n\rangle \tag{2}$$

A perturbation treatment in the rotating wave approximation and with $a_0 \sim 1$ yields

$$\dot{a}_2(z,t) = i\Omega_{20}^{(2)}\ e^{-i\delta_2 t}e^{i2k_L z}a_0(z,t) + i\Omega_{23}e^{-i(\omega_{IR}-\omega_{23})t}e^{ik_{IR}z}a_3(z,t)$$

$$\dot{a}_3(z,t) = i\Omega_{30}\ e^{-i\delta_3 t}e^{ik_{UV}z}a_0(z,t) + i\Omega_{32}e^{i(\omega_{IR}-\omega_{23})t}e^{-ik_{IR}z}a_2(z,t) \tag{3}$$

where $2\Omega_{23}$ is the Rabi frequency between $|2>$ and $|3>$, etc., and $2\Omega_{02}^{(2)}$ is the two-photon Rabi frequency between $|0>$ and $|2>$ wherein state $|1>$ is adiabatically eliminated. Equation (2) can be solved for the amplitudes a_n. From these one can also write the nonlinear polarization source terms for the PFWM waves at frequencies ω_3 and ω_4. Thus

$$\begin{aligned} P^{NL}(\omega_3) &= ND_{32}\langle\langle a_2 a_3^{\dagger}\rangle\rangle e^{-i(\omega_2-\omega_3)t} + c.c. \\ &= ND_{23}e^{-i(\omega_{IR}t-k_{IR}z)}\frac{1}{\Delta_2\delta_3}\left[\Omega_{20}^{(2)}\Omega_{03}e^{-i\Delta kz} - \frac{1}{\delta_3}\Omega_{23}|\Omega_{03}|^2\right] + c.c. \end{aligned} \tag{4}$$

where D_{32} is the dipole matrix element between $|2>$ and $|3>$ and $\Delta_2 = \delta_2 + \Gamma$ where Γ allows for collisional dephasing and laser bandwidth effects. An analagous expression holds for $P^{NL}(\omega_4)$. Now we note that $\left[\Omega_{20}^{(2)} - \Omega_{23}\Omega_{30}e^{i\Delta kz}/\delta_3\right]$ is the effective two-photon Rabi frequency for a_2 with $\tilde{\Omega}_{02}^{(2)} = \Omega_{23}\Omega_{30}/\delta_3$. The solutions evolve such that Eq. (1) is satisfied, thus with $\Delta k = 0$, the nonlinear polarization at ω_3 and ω_4 also go to zero at a critical depth into the medium.

We have conducted a series of experiments in a Na heat pipe (20 cm vapor column length), and have examined the pressure and power dependencies of PFWM processes when tuning near two-photon resonance with the 4d state. (Lumonics excimer pumped dye laser with 3 to 5 nJ/pulse in 5 nsec pulses of ~0.1 cm^{-1} bandwidth).

The interference effect has several consequences for PFWM production.

(1) The PFWM gain vanishes at a critical depth into the nonlinear medium, i.e., where the generated fields grow to the strength required to produce the interference condition of Eq. (1). Thus additional propagation through the medium produces no further growth in the generated fields.

(2) Similarly, under given conditions of laser field and path length, PFWM output will saturate with pressure at a density where Eq. (1) is just satisfied within the length of the medium. Further increase in vapor density will produce no additional increase in the generated light. This behavior is clearly exhibited in the data of Fig. 1. The results were produced by pumping 0.3 Å from the 4D

resonance in Na at 34 MW/cm^2 and observing forward PFWM emissions at λ_3 ~2.33μ and λ_4 ~330.3 nm. The data show that PFWM at 2.3μ saturates at about 0.4 Torr Na pressure. Further increases in number density produces no additional PFWM. (We note that the PFWM process associated with pumping near the 4d state is dominated by axial[8] as opposed to conical components). Additionally, calculations yield the expected result that $\Omega_{02}^{(2)} = \tilde{\Omega}_{02}^{(2)}$ at the infrared (IR) and ultraviolet (UV) intensities produced at ~0.4 Torr and 34 MW/cm^2 pumping.

(3) The generated fields, E_3 and E_4 at frequencies ω_3 and ω_4, obey a constant of motion such that $(E_3^2/\omega_3) - (E_4^2/\omega_4)$ = constant. That is, the change in photon number at ω_3 is equal to the change at ω_4. This, together with the condition Eq. (1) for two-photon cancellation, means that the intensity of the PFWM beams at ω_3 and ω_4 will grow linearly with laser pump intensity, once the interference condition is achieved. Thus, the fields at a given pressure, at ω_3 or ω_4 would grow nonlinearly with laser intensity at low pump intensities but linearly at laser intensities above a critical cancellation value I_c. This influence of the interference effect on PFWM output is revealed in the data of Fig. 2. Again the 2.33μ PFWM component is shown. In the lower curve at 0.35 Torr Na the yield suddenly changes to a linear dependence on laser intensity, I_L, at a critical intensity, I_c, of 0.85 MW/cm^2 (laser tuned onto 4d resonance). The critical laser intensity is that which produces PFWM of intensity sufficient to satisfy Eq. (1). At a higher P_{Na} of 2 Torr (upper curve) the onset of the interference occurs at a lower laser intensity.

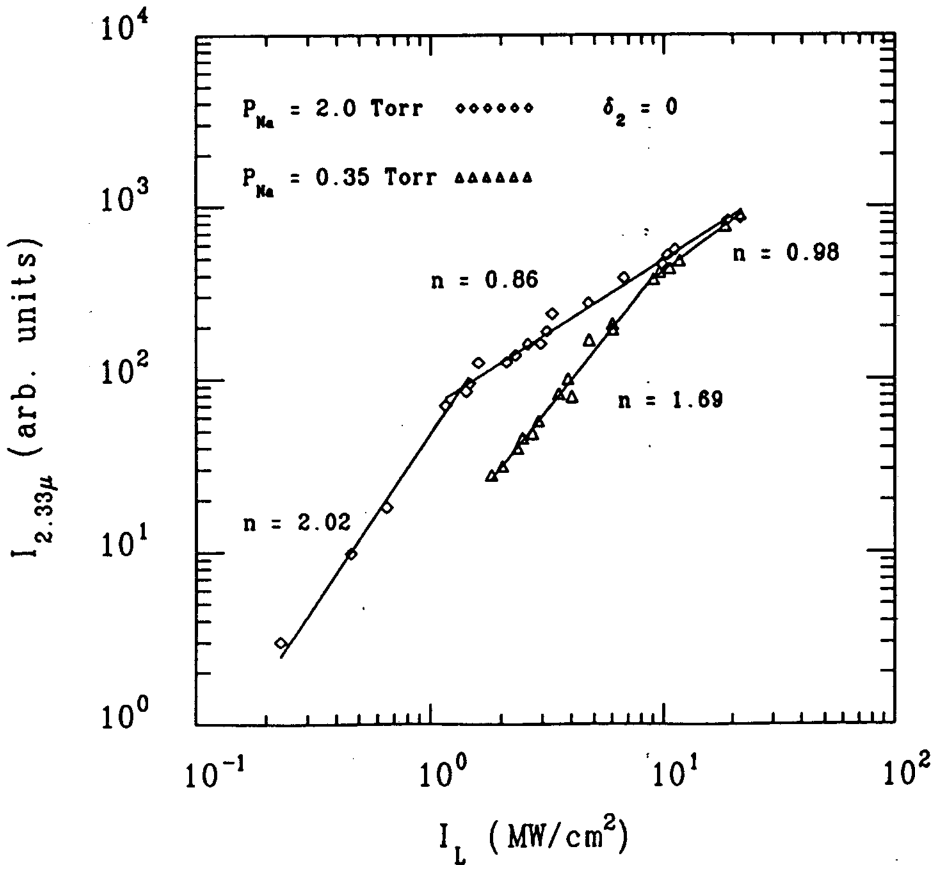

Fig. 2. Intensity of the 2.3μ PFWM emission as a function of pump laser intensity for two different Na vapor pressures. IR intensities go as I_L^n where the values of n were obtained from a computer generated fit to the data.

(4) One can show a fourth property due to the interference effect; that the product of critical intensity and sodium pressure should be a constant. This property holds for the data of Fig. 2, where for the two cases $I_c P_{Na} \cong 2.5$ Torr MW/cm^2.

(5) Yet another predicted feature of PFWM under the influence of the interference effect is shown in Fig. 2, namely, above the critical intensity where the outputs of the PFWM components go linear with laser intensity, the intensities should merge to a common yield curve for all Na densities (assuming sufficient vapor column length to produce critical PFWM fields).

(6) Finally, there is a sixth and rather novel result of the interference effect. Since PFWM output becomes limited when the two-photon Rabi rate from the parametric waves, $\Omega^{(2)}_{PFWM}$, becomes equal in magnitude to that from the laser pumping, $\Omega^{(2)}_L$, then for a given $\Omega^{(2)}_L$, a PFWM process involving a large nonlinear susceptibility will, under conditions of two-photon cancellation, produce less output than one with a smaller $\chi^{(3)}$.

It is interesting, but not surprising, that the most comprehensive evidence for the presence of an interference effect involving PFWM can be obtained from the overall saturation behavior of the PFWM process itself.

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REFERENCES

1. M. S. Malcuit, D. J. Gauthier, and R. W. Boyd, Phys. Rev. Lett. 55:1086 (1985).
2. R. W. Boyd, M. S. Malcuit, and D. J. Gauthier, Phys. Rev. A 35:1648 (1987).
3. V. V. Krasnikov, M. S. Pshenichnikov, and V. S. Solomatin, JETP Lett. 43:148 (1986); [P. Zh. Eksp. Theo. Fiz. 43:115 (1986)].
4. V. V. Krasnikov, M. S. Pshenichnikov, and V. S. Solomatin, Sov. J. Quant. Elect. 18:991 (1988).
5. R. K. Wunderlich, M. A. Moore, W. R. Garrett, and M. G. Payne (unpublished).
6. E. A. Manykin and A. M. Afanas'ev, Sov. Phys. JETP 21:619 (1965); 25:828 (1967).
7. G. S. Agarwal, Phys. Rev. Lett. 57:827 (1986).
8. M. A. Moore, W. R. Garrett, and M. G. Payne, Phys. Rev. A 39:3692 (1989).