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SUPPRESSION OF TWO-PHOTON RESONANTLY ENHANCED NONLINEAR PROCESSES IN EXTENDED MEDIA*

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ABSTRACT

On the basis of combined experimental and theoretical studies of nonlinear processes associated with two-photon excitations near 3d and 4d states in Na, we show how resonantly enhanced stimulated hyper-Raman emission, parametric four-wave mixing processes and total resonant two-photon absorption can become severely suppressed through the actions of internally generated fields on the total atomic response in extended media.

INTRODUCTION

A number of nonlinear optical studies have established that certain strongly resonant multiphoton phenomena which are readily observed under very low density conditions may become almost undetectable at higher number densities, though from simple considerations they would be expected to scale with pressure.[1-3] In a series of studies in Na vapor, we have demonstrated some new suppression mechanisms which become operative in near resonant nonlinear processes and we have illustrated new features of previously known suppression effects.

Some facts associated with two-photon excitations of $s \rightarrow d$ transitions in "extended" alkali vapors are the following: 1) at two-photon resonance, laser absorption is much smaller than that predicted on the basis of atomic beam photoionization experiments; 2) stimulated hyper-Raman (SHR) emission is suppressed in the forward direction and backward SHR, parametric four-wave mixing (PFWM), and amplified spontaneous emission (ASE) show saturation behavior at high laser power and/or high number density; 3) the saturation effect mention in 2) has a very sharp onset with pressure at constant laser power, or with laser power at constant pressure; 4) the backward propagating SHR profiles show a power and pressure dependent dip in lineshape at the two-photon resonance, and 5) although in these experiments SHR emission should have higher gain than PFWM, nevertheless PFWM processes are comparable in intensity to SHR emissions. All of these effects can be operative in regimes where laser beam attenuation and population transfer are not significant.

We have delineated the respective roles of a.c. Stark effects and PFWM interferences in suppressing expected results for two-photon resonantly enhanced phenomena, and explained quantitatively the features listed in the above paragraph.

Experimental elements included a stainless steel heat pipe oven and an excimer pumped dye laser system of 3 mm beam diameter, maximum unfocused power densities of $5 \times 10^6 \mathrm{W/cm^2}$, and $\approx 0.08 \mathrm{cm^{-1}}$ FWHM bandwidth.

RESULTS

In an earlier study of nonlinear wave mixing phenomena in Na vapor, we have shown that the three-photon resonant interference effect, in which an internally generated four-wave mixing field interferes with direct three-photon excitation,[3,4] manifests itself in SHR emission.[5] Indeed we have shown that forward directed SHR emissions are suppressed for hyper-Raman processes involving excitation of a dipole allowed transition.[5] It is easy to show that the same behavior should result for the five-photon hyper-Raman process shown in Fig. 1. The predicted behavior has been established in two-photon pumping near 4d states in Na. In the lower trace in Fig. 1, backward directed five-photon hyper-Raman emissions near 8000 Å, associated with $3p_{3/2}$ and $3p_{1/2}$ excitations, as shown (corresponding to ω' and ω'' in the diagram). In the upper trace the forward axial emissions are recorded under identical circumstances. The SHR lines are absent, due to interference by the internally generated six-wave mixing fields at the $3p_{1/2} - 3s_{1/2}$ and $3p_{3/2} - 3s_{1/2}$ frequencies. (The single line is due to axially phase matched parametric six-wave mixing.)

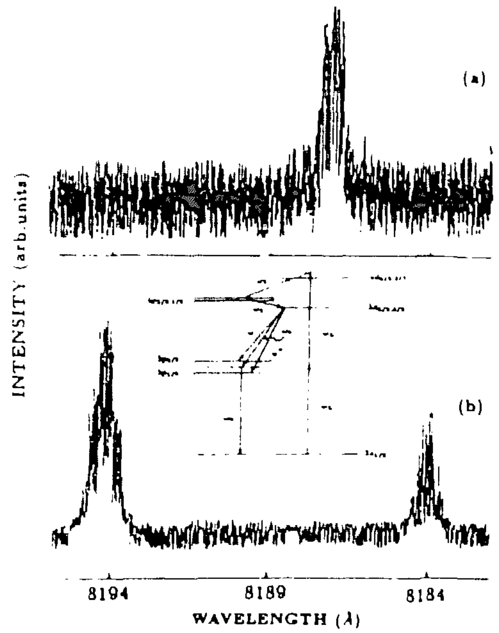

Figure 1. Insert: Five-photon SHR emissions, ω', ω'', in Na. Lower trace, backward SHR profiles. Top trace, forward emission. No SHR in forward direction.

The backward directed SHR (one of which is ω_3 in Fig. 1) causes large a.c. Stark shifting of the $4d$ and $4p$ states. This effect limits the gain of backward SHR, causing it to go from a quadratic laser intensity, I_L^2, over to linear dependence for SHR output. This effect is demonstrated in Fig. 2 for ≈819 nm emissions associated with $3p_{3/2}$ and $3p_{1/2}$ SHR excitations when tuning near two-photon resonance with the $3d$ states. The Stark shifting also causes the SHR excitation profiles to show a dip at two-photon resonance.

Additionally, when tuned to two-photon resonance, the generation of PFWM fields can cause a different class of interference to occur between direct two photon pumping by the laser field and two photon pumping by the PFWM fields.[6,7] The generated fields grow until the interference cancels the two-photon pumping mechanism and thereafter no additional PFWM generation occurs unless the driving field changes. Thus at fixed laser power the intensity of a PFWM signal should increase with pressure until the interference occurs, then saturate. This type of behavior is shown in Fig. 3 where the PFWM wave at 2.33μ generated when tuning near the $4d$ state, is shown as a function of Na number density (fixed laser intensity of 34 MW/cm^2).

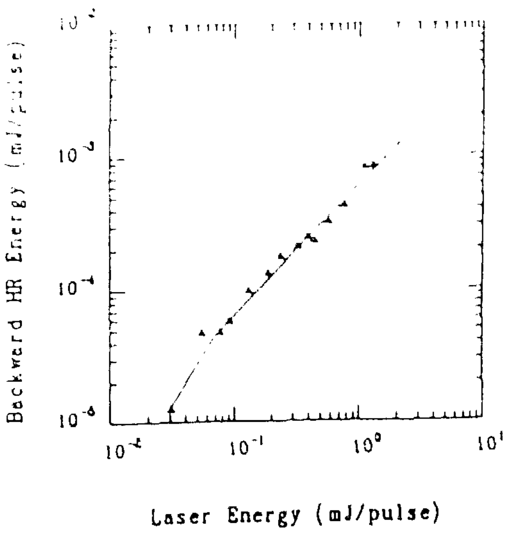

Figure 2. Intensity of backward SHR emissions near 8100 Å when tuning near Na 3d state, showing quantative and linear power dependent regions. $P_{Na} = 0.07$ Torr.

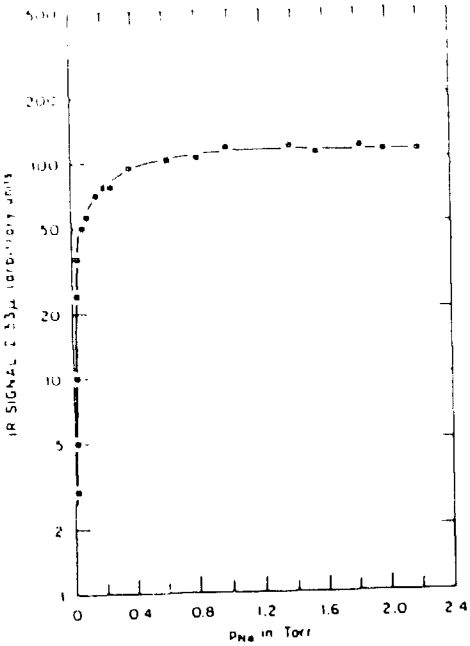

Figure 3. PFWM output at 2.33 mm as a function of P_{Na}. Laser intensity 34 MW/cm^2, detuned 0.3 Å from 4d resonance.

Finally, we show that with increasing laser intensity the two-photon interference will set in at a given intensity (for fixed number density and column length). When this occurs we predict that the power dependence of I_{PFWM} will go over from quadratic in I_L to linear in I_L. This feature has been shown to occur again for the 2.33μ component of PFWM generated at two-photon resonance with 4d.

We get quantitative agreement between experimental findings and theoretical predictions on these and other features of the nonlinear behavior associated with near resonant pumping of two-photon resonances in Na.

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