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Influence of stimulated Raman scattering on the conversion efficiency in four wave mixing

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Abstract: Secondary nonlinear optical effects following parametric four wave mixing in sodium vapor are investigated. The generated ultraviolet radiation induces stimulated Raman scattering and other four wave mixing process. Population transfer due to Raman transitions strongly influences the phase matching conditions for the primary mixing process. Pulse shortening and a reduction in conversion efficiency are observed.

Introduction

Tunable light sources in the ultraviolet (UV) and vacuum ultraviolet (VUV) region of the optical spectrum are important tools for the resonant excitation and ionisation of atoms and moleculs whose lowest excited states have energies above $6eV$. Below $200nm$ tunable VUV light is most often produced by four wave mixing (FWM) in phase matched gas mixtures. The tuning range has been extended below $100nm$ (Hilber et. al 1987) using Kr as nonlinear medium. The efficiency of UV generation is increased by choosing an active medium with resonance enhancement of the third order nonlinear susceptibilty ,$\chi^{(3)}(\omega_{UV})$, at the particular wavelength.

Here we are concerned with loss processes of the generated UV due to further nonlinear processes like stimulated electronic Raman scattering and four wave mixing processes initiated by the UV which do not directly involve the generation of the UV itself.

Processes which directly limit UV generation depend on the resonance structure of $\chi^{(3)}(\omega_{UV})$. In case of a two photon resonance pump depletion due to two photon absorption and ionisation, bleaching of $\chi^{(3)}(\omega_{UV})$ due to population transfer (Heinrich et al 1983) and population transfer induced index changes (Puell et al 1980) limit the UV generation. In addition the generated UV together with one pump photon is always two photon resonant with the ground state and the resonance state in $\chi^{(3)}(\omega_{UV})$ .Resulting absorption can be strong when the UV is generated close to an allowed dipole transition to the ground state (Hilber et al 1987, Payne et al 1988). This two photon absorption process also creates a second pathway for the excitation of the two photon resonance in $\chi^{(3)}(\omega_{UV})$. In case of phase matching this pathways interfere destructively. The generated wave increases in intensity until the Rabi rates for the two pathways become equal and of opposite sign and cancel the two photon resonance (Manykin and Afanas'ev 1965, Malcuit et al 1986,Boyd et al 1987). This also leads to a cancellation of the nonlinear polarisation at the UV and IR frequency. In the latter case the pump waves and the generated wave propagate through the remaining part of the medium without further exchanging energy.

Parametric four wave mixing (PFWM) in Na vapor was used to investigate the secondary loss processes for the generated UV. A laser was tuned near or at the $4D_{3/2,5/2}$ two photon resonance. Phase matched emission at 2.33μ (IR) and $330.3nm$ (UV) connected with the $4D_{3/2,5/2} - 4P_{1/2,3/2}$ and $4P_{1/2,3/2} - 3S_{1/2}$ transitions was observed. This emission is parametric in origin i.e. does not result from a mixing process involving stimulated hyper-Raman and two laser photons. Power conversion efficiencies of 1.% and 3.% for UV and IR generation respectively have been obtained.

DISCLAIMER

Fig.1 shows the pump intensity dependance of the UV and IR generated in the PFWM process. While the IR emission increases linearly with pump intensity for $I_P > 5MW/cm^2$ the UV emission shows onset of saturation. In seperate studies (Garrett et al 1988, Payne et al 1988) we have demonstrated that the two photon interference effect causes I_{IR} and I_{UV} in the PFWM process to change from an I_P^2 to a linear power dependance when the two Rabi rates become equal in magnitude. Also in this case no additional gain is produced by added path length or higher vapor density. Note that in Fig. 1 at right the UV production decreases to even lower dependance on I_P due to the effects described below.

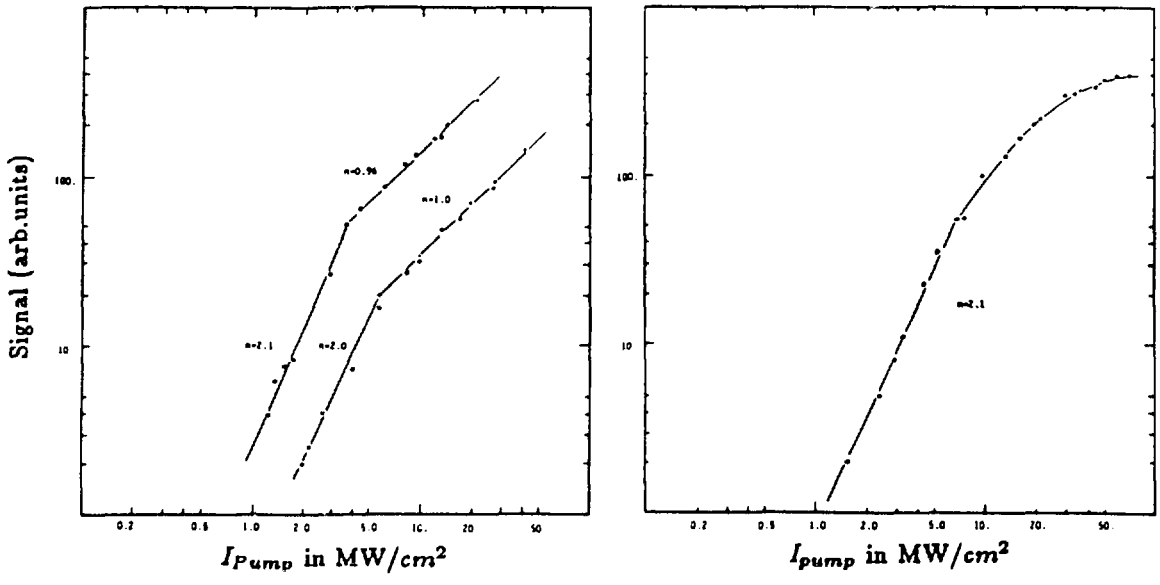

Fig.1 :Pump power dependance of PFWM signals. Left: IR emission, 2.33μ, at 2.0 and 0.4 Torr. Right: UV emission at 2.0 Torr sodium.

The absolute intensities of the IR and UV signal as shown in Fig. 1 can not be compared.From the parametric origin of the mixing process the same power dependance for the IR and UV is expected.

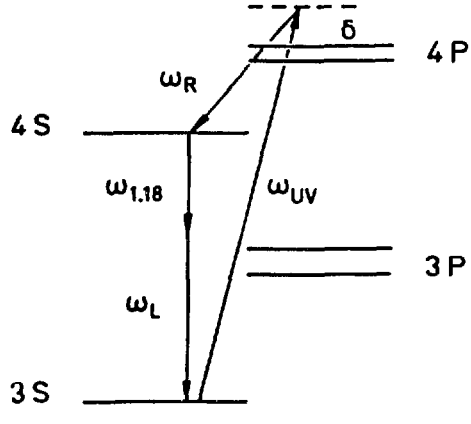

Fig.2 Level scheme for stimulated Raman generation .

Compared to a linear extrapolation of I_{UV} for $I_P > 5MW/cm^2$ the observed output is decreased by ≈ 40%. This decrease is in part due to stimulated Raman scattering of the generated UV connecting the $3S$ ground state with the $4S$ state. The relevant level scheme is shown in Fig.2. Stimulated Raman emission ,ω_R, at 2.20μ is observed in the forward and backward direction. At a pump intensity of $I_P = 30MW/cm^2$ the 2.20μ Raman intensity is $2.10^5 Watt/cm^2$. From a calculation of the Raman intensity build up we find a 25% conversion efficiency in photon numbers from UV to 2.20μ. In the calculation of the Raman gain,G_R, the AC Stark shift, Δ_S , of the $4S-4P$ transition ,induced by the Raman signal,has been included. The shift is given by:

$$\Delta_S = \left(\frac{1}{\delta}\right)|\Omega_{12}|^2$$

and $G_R \sim (1/\Delta_S)^2$. Ω_{12} is the Rabi frequency of the $4S-4P$ transition. the detuning δ is determined by the phase matching conditions for the initial PFWM process. The shift can be quite large because of

the typically large $nl-nl'$ matrix elements. With $I_R = 2.10^6 Watt/cm^2$ we obtain $\Delta_S = 1.1cm^{-1}$ which is much larger than the inhomogeneous width of the transition. Thus the Raman gain is not only limited by pump depletion and gain saturation but also by the induced shift. In the latter case the Raman signal increases only with the square root of the UV intensity.The saturation of the Raman process is demonstrated in the saturation of the of the (backward) 1.14μ amplified spontaneous emission (ASE) as shown in Fig 3. The ASE originates from population transfer to the $4S$ state (see below) which then decays to the $3P$ states .

In addition another four wave mixing process of the type $\omega = \omega_{UV} - \omega_{Raman} - \omega_{Laser}$ is observed with emission at 1.18μ. This process can only phase match by angular adjustment of the emitted radiation which in turn is only possible if the 1.18μ radiation combines with some off axis part of the Raman emission at 2.20μ. It is thus interesting that the 1.18μ FWM signal has a higher intensity than the 1.14μ forward ASE.

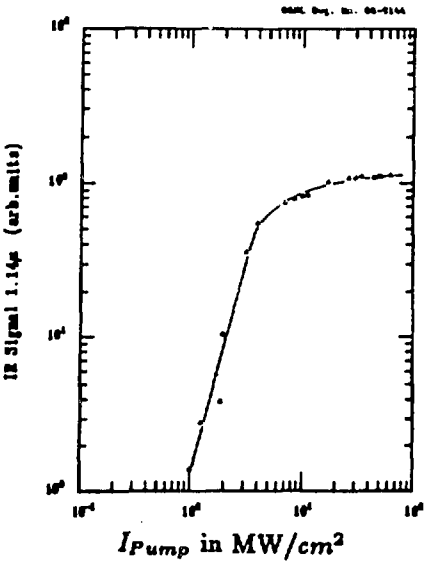

Fig.3 Pump intensity dependance of backward 1.14μ ASE at 2 Torr Na.

Population transfer induced phase mismatch

The Raman process leads to population transfer from the ground state to the $4S$ state. The two photon transition rate R_2 is:

$$R_2 = \left(\frac{2}{\Gamma}\right) |\Omega_{02}^{(2)}|^2 = 1.410^{-2}\ I_{UV}\, I_{Raman}\quad sec^{-1}$$

$\Omega_{02}^{(2)}$ is the two photon Rabi frequency

$$\Omega_{02}^{(2)} = \frac{\Omega_{01}\ \Omega_{12}}{\delta}$$

connecting the $3S-4S$ states , Γ is the transition width and Ω_{01} is the $3S-4P$ Rabi frequency. The intensities I are in $Watt/cm^2$. The number density of atoms at time t in the $4S$ state can be estimated from $N_{4S} = N_{3S} R_2\, t$ if $R_2\, t << 1$; N_{3S} is the number density of ground state atoms . The phase matching condition for the axial component of the initial PFWM process is:

$$\omega_{UV}\, n(\omega_{UV}) = 2\omega_{Laser}\, n(\omega_{Laser}) - \omega_{2.33} n(\omega_{2.33})$$

We set the index at $2.33\mu = 1$. To calculate the population transfer induced index change the linear susceptibility $\chi^{(1)}(\omega_i)$ is written as a sum over all populated excited states $\chi^{(1)} = \Sigma\chi_g^{(1)}$. The excited state population decays very fast by ASE to the lowest excited state from which decay is slower than the inverse laser pulse duration so that the time scale of population transfer is given by the two photon pumping rate R_2. Only the most resonant wavelengths are considered and we assume initial

phase matching. The resulting phase mismatch in our case is:

$$\Delta k_P = \left(\frac{2\pi}{c\hbar}\right) N_{4S}\left\{4\omega_{Laser} D^2_{3S,3P}\left[\delta_L(\tfrac{1}{2}) + \delta_L(\tfrac{3}{2})\right] - \omega_{UV} D^2_{3S,4P}\left[\delta_{UV}(\tfrac{1}{2}) + \delta_{UV}(\tfrac{3}{2})\right]\right\}$$

N_{4S} is the excited state population, $D_{i,j}$ are the matrix elements and δ_L, δ_{UV} are the inverse detunings of the laser and UV respectively from the most resonant states, including the $3P$ and $4P$ fine structure. Large Δk_P can be expected when the UV is generated close to an allowed dipole transition to the ground state. The above formulae is applicable for many two colour FWM experiments where one wavelength ,here the IR, is far off resonance with any populated state and the UV is generated close to a resonance. Inserting the numbers for our experiment we obtain:

$$\Delta k_P = 1.10^{-13} N_{4S}\ \ cm^{-1}$$

The FWM intensity is proportional to $(1/\Delta k)^2$ .A reduction in output by a factor 10^4 can be expected for $N_{3S} = 2.10^{16} cm^3$, $R_2 = 1.10^7 sec^{-1}$ and $t > 4.10^{-9} sec$ compared to an initially phase matched situation. This typically requires UV intensities of some $10^5 Watt/cm^2$. In a measurement of the pulse duration of the driving laser field and the generated UV emission with a fast photodiode we observe a pulse shortening from $7ns$ for the pump pulse to $4.5ns$ (FWHM) for the UV pulse. Also the UV pulse reaches its maximum $3ns$ before the maximum pump intensity which indicates that when the pump pulse is at maximum intensity the phase matching conditions for the PFWM process are already shut off. This pulse shortening has to be considered in the measurement of power conversion efficiencies. It should be mentioned that there is also a contribution from backward stimulated hyper-Raman emission to population transfer.

In summary we have discribed some of the secondary nonlinear effects wich can occur in a four wave mixing experiment especially when the gain for nonlinear generation is high. It appears that the main effect of these processes is two photon induced population transfer via stimulated Raman scattering of the generated UV. This in turn destroys the phase matching conditions for the primary mixing process. The effect can be large and results in pulse shortening of the generated radiation.

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