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EFFECT OF THE COHERENT CANCELLATION OF THE TWO-PHOTON RESONANCE ON THE GENERATION OF VACUUM ULTRAVIOLET LIGHT BY TWO-PHOTON RESONANTLY ENHANCED FOUR-WAVE MIXING*

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ABSTRACT

Many of the most impressive demonstrations of the efficient generation of vacuum ultraviolet (VUV) light have made use of two-photon resonantly enhanced four-wave mixing to generate light at $\omega_{VUV} = 2\omega_{L1} \pm \omega_{L2}$. The two-photon resonance state is coupled to the ground state both by two photons from the first laser, or by a photon from the second laser and one from the generated VUV beam. We show here that these two coherent pathways destructively interfere once the second laser is made sufficiently intense, thereby leading to an important limiting effect on the achievable conversion efficiency.

INTRODUCTION

Various aspects of the two-photon cancellation effect were first discussed in a series of papers by Manykin and Afanas'ev[1] published more than 20 years ago. However, detailed experimental confirmation of the predicted effects have only recently been presented.[2-4] We discuss here the ramifications of this effect on the use of two-photon resonances to enhance nonlinear susceptibilities used in the generation of VUV light. Not only do these effects distort the radial intensity profile of the generated light, but they also limit the conversion efficiency to a few percent.

RESULTS

We describe as a three state system the atomic response when a first laser at frequency ω_{L1} is tuned on or near resonance between the ground state $|0>$ and an excited state $|2>$, and a second laser at ω_{L2} causes a near three-photon resonance between $|0>$ and $|3>$ at $2\omega_{L1} + \omega_{L2}$. Neglecting population transfer from the ground state, but including the possible absorption of the generated VUV due to resonant absorption in which a VUV photon is absorbed and an emission is stimulated at the frequency of the second laser, we find for the nonlinear polarization at angular frequency $2\omega_{L1} + \omega_{L2}$

$$\mathcal{P}_{VUV}^{NL} = e^{i\Phi} \frac{N D_{3,0} \Omega_{23}^{(1)}}{\delta_3(\delta_2 + i\Gamma)} \left[\Omega_{02}^{(2)} e^{-i\Delta k z} - \frac{\Omega_{32}^{(1)} \Omega_{03}^{(1)}}{\delta_3} \right] + c.c. \qquad (1)$$

In Eq. (1) $2\Omega_{02}^{(2)}$ is the two-photon Rabi frequency for the transition $|0> \leftrightarrow |2>$ due to the first laser, $2\Omega_{23}^{(1)}$ is the one-photon Rabi frequency for the transition $|2> \leftrightarrow |3>$ due to the second laser, and $2\Omega_{03}^{(1)}$ is the one photon Rabi frequency for the $|0> \leftrightarrow |3>$ transition due to the generated field at $2\omega_{L1} + \omega_{L2}$. N is the concentration of the active gas, $\Phi \equiv k_{VUV}z - \omega_{VUV}t$, δ_2 is the detuning from exact resonance between the first laser and the $|0>$ to $|2>$ resonance, δ_3 is the detuning of $2\omega_{L1} + \omega_{L2}$ from resonance between $|0>$ and $|3>$, Γ is the collisional width of the two-photon resonance, $\Delta k \equiv k_{VUV} - 2k_{L1} - k_{L2}$ is the phase mismatch, and $D_{0,3}$ is the matrix element of the electronic dipole operator between states $|0>$ and $|3>$. The term $\Omega_{03}^{(1)}\Omega_{32}^{(1)}/(\delta_3)$ is a two-photon Rabi frequency for driving transitions between $|0>$ and $|2>$ by the absorption of a VUV photon and a stimulated emission of a photon at ω_{L2}. The term in $\mathcal{P}_{VUV}^{NL}$ involving the latter combination includes in the polarization both the absorption of the VUV wave by this process and the power dependent phase mismatch due to the strong coupling between $|2>$ and $|3>$ when $\delta_2 \neq 0$. If we use Eq. (1) in Maxwell's equations and implement the slowly varying phase and amplitude approximation we find for $\Delta k = 0$

$$\Omega_{03}^{(1)} = \frac{\Omega_{02}^{(2)}\delta_3}{\Omega_{32}^{(1)}}(1 - e^{-\beta z}), \tag{2}$$

where $\kappa_{03} = 2\pi\omega_{VUV}N|D_{0,3}|^2/(\hbar c)$, and $\beta = i\kappa_{03}(|\Omega_{23}^{(1)}|^2)/[\delta_3^2(\delta_2 + i\Gamma)]$. If $|\beta L|$ is much larger than unity the exponential term is small and we see that by the time the exit end of the cell (z = L) is reached $\Omega_{03}^{(1)}\Omega_{32}^{(1)}/\delta_3 = \Omega_{02}^{(2)}$. This is just the condition for the total destructive interference of the two pathways for pumping the transition between state $|0>$ and state $|2>$. The approach to this limit involves a true cancellation of the two-photon resonance if $\delta_2 = 0$. However, when $|\delta_2| >> \Gamma$ the solution oscillates about this limit as a function of z due to a large power dependent phase mismatch induced by the near two-photon resonance. We see that the VUV signal becomes limited by the cancellation of the pumping of the two-photon resonance. On the other hand, if $|\beta L| << 1$ the result becomes exactly equivalent to the conventional result.

One thing which is easily seen from the cancellation condition is that making the power density at ω_{L2} too large can actually decrease the generation of light at ω_{VUV}. The optimum intensity at frequency ω_{L2} is such that the absoption of the VUV due to two-photon excitation satisfies $N\sigma_a L/2 \simeq 1.5$. Either less or more power density at this wavelength decreases the generated intensity of the VUV light. When the intensity is made far too large we must consider the power densities as a function of radial distance away from beam center carefully if we are to be able to interpret the manifestations of the effect in an actual experiment. We assume that the radial power densities are such that $\Omega_{23}^{(1)} = \Omega_{23}^{(1)}(0)e^{-\rho^2/R_2^2}$, and $\Omega_{02}^{(2)} = \Omega_{02}^{(2)}(0)e^{-2\rho^2/R_1^2}$.

We will now integrate over the radial power densities for the lasers and derive an expression for the number of VUV photons passing through a cross section of the beam at z during the pulse for the case $\delta_2 = 0$ and $\Delta k = 0$. Let $x = \beta(0)z$ and $y = (R_2/R_1)^2$, then after some substitutions

$$N_\gamma = \frac{1}{2} N_e W(\beta(0)z, (R_2/R_1)^2), \tag{3}$$

with $N_e = [2|\Omega_{02}^{(2)}(0)|^2 \tau/\Gamma](N\pi R_1^2 z)$ = number of excited atoms produced in state 2 from cell entrance to z, and τ is the pulse length of the lasers. Above, $\beta(0)$ is the value of beta at beam center, R_1 and R_2 are the radii of the laser beams and the function W is given by

$$W(x,y) = yx^{-2y} \int_0^x \frac{ds}{s^{2(1-y)}} \left(1 - e^{-s}\right)^2 = 2xy \sum_{n=0}^{\infty} (-1)^n \frac{[2^{n+1} - 1]}{(n+2)!} \frac{x^n}{n + 2y + 1}.$$

W is usually less than 0.2, where this value is reached for $\beta(0)L$ values between 1 and 5 if the beam radii are not widely different. This suggests that an overall efficiency in generating VUV photons is limited to about 10% of N_e. It is probable that an N_e large enough to attenuate the laser significantly would spoil the phase matching by changing the index of the medium. Another effect which becomes probable when the power density of the first laser is large enough to produce strong attenuation of the first laser beam is the initiation of parametric four-wave mixing which can grow from noise to the point where it leads to greatly reduced absorption of the laser beam. Exactly this effect is known to greatly reduce the absorption of laser beams tuned near the 3s to 4d two-photon resonance in sodium.[4]

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REFERENCES

1. E.A. Manykin and A.M. Afanas'ev, Soviet Physics JETP 21, 619 (1965). See also: A.M. Afanas'ev and E.A. Manykin, Soviet Physics JETP 21, 323 (1965).
2. M.S. Malcuit, D.J. Gauthier, and R.W. Boyd, Phys. Rev. Lett. 55, 1086 (1985).
3. M.A. Moore, W.R. Garrett, and M.G. Payne, Optics Commun. 68, 310 (1988).
4. W. R. Garrett, Mary Anne Moore, M. G. Payne, and Rainer K. Wunderlich (Proceeding of the Fourth International Laser Sience Conference (ILS-IV), Atlanta, Ga, October 1988 (to be published by American Institute of Physics).