For publication in the Proceedings of the Fourth International Laser Science Conference (ILS-IV) (Advances in Laser Sciences - IV) Mariott Marquis Hotel, Atlanta, Georgia, October 2-6, 1988, to be published by the American Institute of Physics

NEW COHERENT CANCELLATION EFFECT INVOLVING FOUR-PHOTON EXCITATION AND THE RELATED IONIZATION*

CONF-881002--35

DE89 004800

M. G. Payne, W. R. Garrett,
J. P. Judish, and M. P. McCann**

Chemical Physics Section, Oak Ridge National Laboratory,
Post Office Box 2008
Oak Ridge, Tennessee, USA, 37831-6378

November 1988

*Research sponsored by the Office of Health and Environmental Research, U.S. Department of Energy under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

**Postdoctoral fellow, University of Tennessee, Knoxville, Tennessee

"The submitted manuscript has been authored by a contractor of the U.S. Government under contract DE-AC05-84OR21400. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes."

MASTER

NEW COHERENT CANCELLATION EFFECT INVOLVING FOUR-PHOTON EXCITATION AND THE RELATED IONIZATION

M.G. PAYNE, W.R. GARRETT,
J.P. JUDISH, AND M.P. MCCANN
CHEMICAL PHYSICS SECTION, OAK RIDGE NATIONAL LABORATORY,
OAK RIDGE, TENNESSEE 37831-6378

ABSTRACT

We describe here an effect which occurs when a first laser is tuned near a dipole allowed three-photon resonance and a second laser is used to complete a dipole allowed four-photon resonance between the ground state $|0>$ and an excited state $|2>$. In this process three photons are absorbed from the first laser and one photon from the second; so that if the $|0>$ to $|2>$ transition is two-photon allowed the transition is also pumped resonantly by the third harmonic field due to the first laser and the second laser field. When the second laser is strong enough to cause strong absorption of the third harmonic light, and the phase mismatch, Δk is large and dominated by the nearby resonance, a destructive interference occurs between the pumping of the $|0>$ to $|2>$ transition by two- and four-photon processes.

I. INTRODUCTION

Blazewicz and Miller[1] studied five-photon ionization in the region near several three-photon resonances in Xe and Kr. These three-photon resonances were $J = 0$ to $J = 1$ transitions, which are also dipole allowed in one photon. Two colors of light were used, with the first laser being tuned through the phase matching region for third harmonic generation while the second was fixed at a frequency such that four-photon resonance was achieved for some preselected frequency for the first laser. Even without the second laser considerable ionization was seen in this situation, with the ionization lineshape being similar to the phase matching curve for the third harmonic generation. For several different transitions, and for a wide range of pressures, they observed that when the four-photon resonance occurred on the side of the peak of the phase matching curve closest to three-photon resonance a dip in ionization coincided with four-photon resonance. On the other hand, when resonance occurred on the side of the peak with $-\Delta kb < 4$ a peak was observed in the ionization. The observed dips were found to occur only when the power density of the second laser was sufficiently high so that observations of the third harmonic photons showed a deep dip at the resonance. We suggest here that the dips are due to an interference between four- and two-photon pumping of the $|0>\leftrightarrow|2>$ transition which with a broad bandwidth laser is pumped at a rate $R \simeq |\Omega_{0,1}^{eff}\Omega_{1,2}|^2/(\delta_1^2\sqrt{3\Gamma_{L1}^2+\Gamma_{L2}^2})$. Here,

$\Omega^{eff}_{0,1}$ is half the Rabi frequency for $|0>$ to $|1>$ (including both channels) and $\Omega_{1,2}$ is half of the Rabi frequency between $|1>$ and $|2>$ due to the second laser. δ_1 is the detuning of the second laser from the three-photon resonance, Γ_1 and Γ_2 are the laser bandwidths. When the cancellation occurs $\Omega^{eff}_{0,1} \simeq 0$, thereby leading to no population in $|2>$. The four-photon resonant cancellation to be described here is closely related to the odd-photon resonance cancellation effect first observed by Miller et al[2] and treated theoretically by Payne and Garrett[3-5] and others.[6,7]

II. SUMMARY OF RESULTS

We treat this problem as a three state problem, with states $|0>$ and $|1>$ being coupled coherently by both a three-photon Rabi frequency, $2\Omega^{(3)}_{0,1}$, due to the laser and by a one-photon Rabi frequency, $2\Omega^{(1)}_{0,1}$, due to third harmonic light generated in the medium. The states $|1>$ and $|2>$ are coupled by a one-photon Rabi frequency $2\Omega^{(1)}_{1,2}$ due to the second laser. In dealing with the generation of third harmonic light we assume that the nonlinear susceptibility is dominated by the near resonant terms due to the nearby three-photon resonance. The latter can be related to the three-photon Rabi frequency and δ_1. Since $|0>$ and $|1>$ are coupled by both three-photon and one-photon Rabi frequencies we write an effective coupling parameter as

$$\Omega^{eff}_{0,1} = e^{-3ik(\omega_{L1})z}\Omega^{(3)}_{0,1} + e^{-ik(3\omega_{L1})z}\Omega^{(1)}_{0,1}, \qquad (1)$$

where ω_{L1} is the frequency of the first laser and $k(\omega)$ is the length of the propagation vector for an electromagnetic wave with frequency ω. It is this coupling parameter which enters the equations of motion for the density matrix when both coupling channels are included.

We find for the polarization at $3\omega_{L1}$:

$$\mathcal{P}_{3\omega_{L1}} = -ND_{0,1}\frac{\Omega^{eff}_{1,0}}{\delta_1}e^{-3i\omega_{L1}t}\left(1 + i\frac{<<|\Omega_{1,2}|^2>>}{\delta_1\sqrt{3\Gamma^2_{L1}+\Gamma^2_{L2}}}\right) + cc., \qquad (2)$$

where the quantity $D_{0,1}$ is the dipole matrix element between states $|0>$ and $|1>$ and N is the concentration of the active gas.

This polarization can now be combined with Maxwell's equations in order to determine the third harmonic field. As a consequence of our few state approximation Eq. (2) contains only one term linear in the third harmonic field. To properly deal with the dispersive properties of the medium we add on linear polarization terms at $3\omega_{L1}$ due to all other dipole allowed transitions in both the active atoms and any buffer gas atoms that may be present.

We let $\kappa_{0,1} = 2\pi(3\omega_{L1})N|D_{0,1}|^2/(\hbar c)$, $\Delta_s = << |\Omega^{(1)}_{1,2}|^2 >> /\delta_1$, and

$$\alpha = \frac{\kappa_{0,1}}{\delta_1} \frac{\Delta_s}{\sqrt{3\Gamma^2_{L1} + \Gamma^2_{L2}}},$$

$$\Delta k \equiv \frac{3\omega_{L1}}{c}(n(3\omega_{L1}) - n(\omega_{L1})) = -\frac{\kappa_{0,1}}{\delta_1} + \Delta k_0. \qquad (3)$$

2α is the absorption coefficient for the third harmonic light due to two-photon absorption, Δ_s is the a.c. Stark shift in level $|2>$ due to the second laser, and Δk_0 is what needs to be added to the near resonant $|0> \rightarrow |1>$ contribution to the phase mismatch in order to get Δk. Using the slowly varying amplitude and phase approximation with Maxwell's equations and the nonlinear part of the polarization from Eq. (2) ($\Omega^{(1)}_{1,0} = D_{1,0}E^{(0)}_{3\omega_{L1}}/(2\hbar)$)

$$\Omega^{(1)}_{1,0} = -\Omega^{(3)}_{1,0}\left[1 - \frac{\Delta k_0}{\Delta k + i\alpha}\right]\left(e^{-i\Delta kz} - e^{-\alpha z}\right). \qquad (4)$$

If $\alpha z >> 1$ and $\sqrt{\Delta k^2 + \alpha^2} >> |\Delta k_0|$ Eq.(4) yields $\Omega^{(1)}_{1,0} \simeq -\Omega^{(3)}_{1,0}e^{-i\Delta kz}$. In this limit Eq. (1) becomes $\Omega^{eff}_{0,1} \equiv 0$. Thus, no population transfer to state $|2>$ occurs for z such that $\alpha z >> 1$.

Research sponsored by the Office of Health and Environmental Research, U.S. Department of Energy under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc. M. P. McCann, postdoctoral fellow, University of Tennessee, Knoxville, Tennessee.

REFERENCES

1. P.R. Blazewicz and J.C. Miller, Phys. Rev. A38, 2863 (1988). See also: P.R. Blazewicz, M.G. Payne, W.R. Garrett, and J.C. Miller, Phys. Rev. A34, 5171 (1986).
2. J.C. Miller, R.N. Compton, M.G. Payne, and W.R. Garrett, Phys. Rev. Lett. 45, 114 (1980).
3. M.G. Payne, W.R. Garrett, and H.C. Baker, Chem. Phys. Lett. 75, 468 (1980).
4. M.G. Payne and W.R. Garrett, Phys. Rev. A26, 356 (1982).
5. M.G. Payne and W.R. Garrett, Phys. Rev. A28, 3409 (1983). See also: M.G. Payne, W.R. Ferrell, and W.R. Garrett, Phys. Rev. A27, 3053 (1982).
6. D.J. Jackson and J.J. Wynne, Phys. Rev. Lett. 49, 543 (1982).
7. D.J. Jackson, J.J. Wynne, and P.H. Kess, Phys. Rev. A28, 781 (1983).