Effect of Pressure Dependent Quantum Interference on the ac Stark Shifting of a Four-Photon Resonance

Lu Deng, J. Y. Zhang, and M. G. Payne
Department of Physics, Georgia Southern University, Statesboro, Georgia 30460-8031

W. R. Garrett
Chemical Physics Section, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831
(Received 22 February 1994)

We report an experiment in xenon which confirms theoretical predictions [Payne *et al.*, Phys. Rev. A **48**, 2334 (1993)] on the pressure dependent suppression of the ac Stark shifting of even-photon resonances. In the copropagating configuration the ac Stark shift introduced by a second laser is totally suppressed at elevated concentrations due to a very complete destructive interference between two pumping pathways. The ac Stark shift persists in the counterpropagating configuration.

PACS numbers: 32.80.Rm

The quantum interference effect (QIE) between different optical harmonics was first reported [1–3] by Compton *et al.* [1] and Miller *et al.* [2], who observed a complete suppression of the three-photon resonantly enhanced multiphoton ionization (MPI) when transitions were studied in argon, krypton, and xenon at concentrations greater than 10^{15} cm^{-3}. Payne *et al.* [4–6] and others [7–10] have developed theories which explain the above effects, and a number of others [11–13], in terms of a destructive interference which occurs at elevated concentrations between one-photon coupling driven by the multiwave mixing field generated in the medium and multiphoton coupling due to the laser fields. This destructive interference occurs only in cases where the lasers are tuned near an odd-photon resonance which has a dipole-allowed transition back to the ground state. When the photons involved in pumping the odd-photon resonance all propagate in the same direction, the destructive interference between different excitation pathways results in the complete suppression of the amplitude for the excited state. However, when one of the laser fields necessary for the resonant coupling is counterpropagated, the amplitude for the excited state remains normal, with the odd-photon resonance being slightly shifted [11,12]. The experimental manifestation of the latter effect on odd-photon resonantly enhanced MPI studies is the absence of the MPI enhancement with copropagating beams. For counterpropagating beams the size of the MPI signal is normal, but the frequency of the signal undergoes a small pressure dependent shift [11].

Until recently, it has been assumed that the presence of an intermediate even-photon resonance would spoil the occurrence of the destructive interference. The upper level in the intermediate even-photon resonance would develop a population which would be involved in developing an amplitude for the upper level in the odd-photon resonance, while the coherent part of the odd-photon coupling plays the dominant role in the generation of the multiwave mixing field. This would supposedly spoil the very special amplitude and phase relation between the two pathways. Recently, Payne, Zhang, and Garrett [14] have shown that under certain conditions, when a second copropagating laser is used to resonantly couple the excited state in the even-photon resonance to the excited state involved in the odd-photon resonance, the destructive interference persists.

A second, but related, prediction of Ref. [14] is the suppression of ac Stark shifts due to the laser coupling of the upper state in the even-photon resonance to the upper state involved in the odd-photon resonance. We provide here an experimental demonstration of this curious interference effect. This study shows that the effect not only occurs when predicted by theory, but also in some situations where a theory has not been provided.

In this brief treatment we restrict the discussion to four-photon resonances to coincide with the experiment conducted in xenon. Thus, consider a laser at angular frequency ω_{L1} tuned near a four-photon resonance between the ground state $|0\rangle$ and an excited state $|1\rangle$. A second laser is tuned near a resonance transition between state $|1\rangle$ and state $|2\rangle$, which has an electric dipole-allowed transition to the ground state $|0\rangle$. In the described situation one must allow for the six-wave mixing field at the frequency $\omega_m = 4\omega_{L1} \pm \omega_{L2}$. The latter field provides a second pathway for coupling between states $|0\rangle$ and $|2\rangle$ which is linear in the six-wave mixing field. At high concentrations, it has been shown that in many circumstances a destructive interference occurs between these two pathways, resulting in the amplitude for state $|2\rangle$ being zero [1–13].

We use the time-dependent Schrödinger equation with a wave function of the form

$$|\Psi(x,t)\rangle = a_0(x,t)e^{-i\omega_0 t}|0\rangle + a_1(x,t)e^{-i\omega_1 t}|1\rangle + a_2(x,t)e^{-i\omega_2 t}|2\rangle + \sum_\mu \int dE\, C_\mu(E,x,t)e^{-iEt/\hbar}|E\rangle . \quad (1)$$

From the orthogonality conditions, the adiabatic approximation for dealing with the discrete-continuum coupling, and the time-dependent Schrödinger equation,

$$\begin{aligned}
\left(\frac{\partial a_0}{\partial t}\right)_x &= i\Omega_{01}^{(4)}A_1 + i\Omega_{02}^{(1)}A_2, \\
\left(\frac{\partial A_1}{\partial t}\right)_x &= i\delta_1 A_1 + i[\Omega_{01}^{(4)}]^* a_0 + i\Omega_{12}^{(1)}e^{i\Delta k_r x}A_2 - \frac{\Gamma_{I1}}{2}A_1, \\
\left(\frac{\partial A_2}{\partial t}\right)_x &= i\delta_2 A_2 + i\Omega_{12}^{(1)}e^{-i\Delta k_r x}A_1 + i[\Omega_{02}^{(1)}]^* a_0 - \frac{\Gamma_{I2}}{2}A_2,
\end{aligned} \tag{2}$$

where we have defined the new amplitudes $A_2 = a_2 e^{i\delta_2 t}e^{-ik_r(\omega_m)x}$ and $A_1 = a_1 e^{i\delta_1 t}e^{-4ik(\omega_{L1})x}$. We have let $\Omega_{01}^{(4)}$ be one-half of the four-photon Rabi frequency between the levels $|0\rangle$ and $|1\rangle$, $\Omega_{12}^{(1)}$ is one-half of the one-photon Rabi frequency between states $|1\rangle$ and $|2\rangle$, and $\Omega_{02}^{(1)}$ is one-half of the one-photon Rabi frequency (due to the six-wave mixing field generated in the medium) between $|0\rangle$ and $|2\rangle$. $\delta_1 = 4\omega_{L1} - (E_1 - E_0)/\hbar$ and $\delta_2 = 4\omega_{L1} \pm \omega_{L2} - (E_2 - E_1)/\hbar$ are detunings from the first and second resonances, and Γ_{I1} and Γ_{I2} are ionization rates out of states $|1\rangle$ and $|2\rangle$.

Neglecting depletion of the ground state and assuming that δ_2 is much larger than δ_1, we use $|\Psi(x,t)\rangle$ to calculate a polarization of the medium at angular frequency ω_m in terms of A_1. Laplace transform techniques are then used to solve simultaneously Maxwell's equations and Eqs. (2). In Maxwell's equations we have defined the phase mismatch as $\Delta k = k(\omega_m) - [4k(\omega_{L1}) \pm k(\omega_{L2})] \equiv \Delta k_r + i\beta/2$, where Δk_r is the real part of the phase mismatch and β is the absorption coefficient at the mixing frequency ω_m. We find

$$A_1(x,t_r) = i\int_{-\infty}^{t_r} dt'\, e^{i\delta_1(t_r - t')}[\Omega_{01}^{(4)}(t')]^* e^{-i\nu(t_r,t')} \times S(\Delta k x, \nu(t_r,t')), \tag{3}$$

where $\nu(t_r,t') = \int_{t'}^{t_r} dt''|\Omega_{12}^{(1)}(t'')|^2/\delta_2$ is the integral of the ac Stark shift, Δkx is the product of the phase mismatch and the distance from the entrance window to the gas cell, and

$$S(\nu,\mu) = \frac{i}{2\pi}\int_{-\infty+i\epsilon'}^{\infty+i\epsilon'} \frac{dq}{q}\, e^{-iq}\exp\left(\frac{i\nu\mu}{\nu + q}\right). \tag{4}$$

After a bit of analysis one can show that the function S can be expressed as $S(\nu,\mu) = 1 + e^{i\nu}\sum_{n=1}^{\infty}[(-i\nu)^n/n!]B_n(\mu)$, where $B_n(\mu) = \sum_{k=1}^{n}(i\mu)^k/k!$. The series solution for S can, in principle, be used whenever μ is real and opposite in sign to the real part of ν.

When $|\Re(\nu)| > 5\sqrt{|\mu|}$, the S takes on the following asymptotic form $S(\nu,\mu) = e^{i\mu}$. Note that when $-\Re(\nu) > 5\sqrt{\mu}$, $|S(\nu,\mu)|$ is close to unity, and in A_1 the phase factor from S cancels with that from the ac Stark shift. In this limit we obtain for A_1 the solution corresponding to $A_2 = 0$. From Eqs. (2) we see that when $A_2 = 0$ the population of the upper state pumped by the laser is unaffected by the presence of laser 2. Thus, the effect of including the six-wave mixing field is to suppress totally the ac Stark shift when $|\Delta kx| > 5\sqrt{\Delta_s^{\max}\tau}$ and the laser beams are copropagating. The parameter τ is the width of the laser pulse in time.

Our experiment is aimed at providing the first critical test of the theory and the underlying mechanism. Because of the ease of making ionization measurements in inert gases, particularly xenon, we decided to carry out the experiment in xenon. We make use of a four-photon resonance between the ground state $5P^6(J = 0)$ and the excited state $5p^5 6p[1/2](J = 0)$, with the first dye laser being tuned near 499.1 nm. The second dye laser couples the $5p^5 6p[1/2](J = 0)$ state with the lower lying second excited state $5p^5 6s[3/2](J = 1)$, as shown in Fig. 1. The latter state has a strong dipole-allowed transition back to the ground state, as required for the predicted suppression of ac Stark shifts due to the second laser.

A Spectra Physics DCR-1A Q-switched Nd:YAG laser was used to pump two Quanta Ray PDL-1 pulsed dye lasers (bandwidth 0.25 cm^{-1}). The third harmonic of the Nd-YAG was used to pump C-500 dye, while the second harmonic of the Nd-YAG was used to pump LDS-821 dye. The outputs of the two dye lasers, after suitable optical delay, were focused into a cell equipped with a proportional counter and filled with xenon gas at a selected pressure. The laser beams were overlapped in the cell in either copropagating or counterpropagating configurations. The output energies of the two dye lasers were $E_1 = 0.51$ mJ at 499.13 nm and $E_2 = 23$ mJ at 828.24 nm, respectively. Both beams were focused to a 250 μm beam waist. We investigated the MPI ion yield in the pressure range of 1 to 250 Torr by scanning the first laser from about 15 cm^{-1} on one side of the four-photon resonance to 15 cm^{-1} on the other side, while the second laser was fixed in frequency at a detuning that

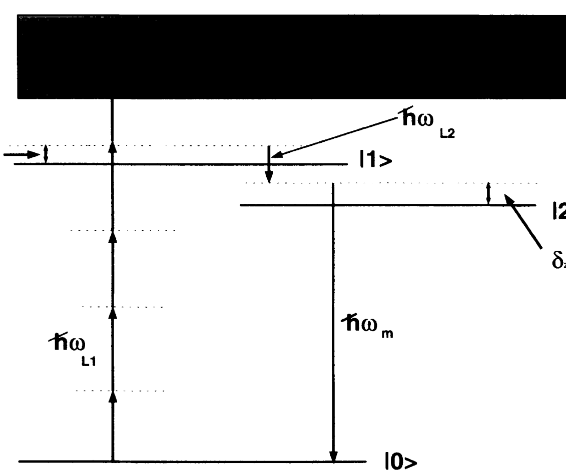

FIG. 1. Energy-level diagram defining the detunings involved in the MPI excitation of the two excited states.

was typically in the range $\pm 10\ \mathrm{cm}^{-1}$ from the one-photon resonance.

Copropagating beams.—In this setup, using the parameters mentioned above, we estimate that at 1 Torr $|\Delta k_r| b/\sqrt{|\Delta_s^{\max}\tau|} \gg 10$ for a value of $|\delta_2| \leq 10\ \mathrm{cm}^{-1}$. For a multimode laser, τ is of the order of the coherence time and b is the confocal parameter. This estimate indicates that we are working in the region where $|S(\Delta kx, \nu(t_r, t'))| \to 1$ and $\Psi(\alpha, \beta) \simeq \beta - \pi/2$. Consequently, we expect to see the total suppression of the ac Stark shift due to the coherent cancellation between the five-photon absorption process involving two lasers and one-photon absorption involving the six-wave mixing field. This is indeed what we observed. In Fig. 2 we plot the MPI ion yield signal versus the wavelength of the first laser at $P = 5$ Torr. The second laser was tuned to $\pm 10\ \mathrm{cm}^{-1}$ from one-photon resonance between the two excited states. In both cases the first dye laser was tuned from 498.8 to 499.4 nm with the second dye laser being turned both on and off. The data exhibited a somewhat asymmetric line shape for the MPI signal in all three situations. The asymmetry is because of ac Stark shifts from the first laser, which must have a high power density in order to observe the MPI signal. We observed no appreciable changes in MPI line shapes and signal strengths when the second laser was blocked, or when it was incident with power densities $I > 10^{10}\ \mathrm{W/cm}^2$ with any detuning δ_2. Similar results were obtained at elevated concentrations, for instance at $P = 75$, 125, and 250 Torr.

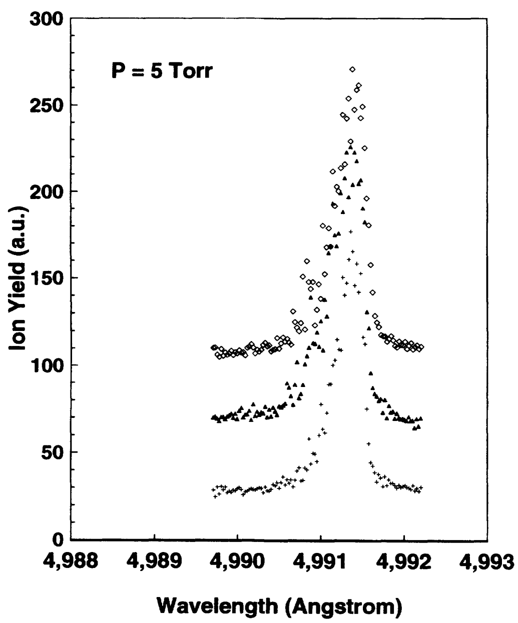

FIG. 2. Plot of the MPI signal against the wavelength of the first laser at $P = 5$ Torr in the copropagating configuration. (+++) The first laser only (i.e., the IR beam is blocked). (◇◇◇) The second laser tuned $10\ \mathrm{cm}^{-1}$ from the one-photon resonance between the two excited states. (▲▲▲) The second laser tuned $-10\ \mathrm{cm}^{-1}$ away from the one-photon resonance. The slightly different linewidths are attributed to laser power drifts. The three line shapes are offset so they can be distinguished.

Counterpropagating beams.—In the counterpropagating configuration, as argued in Ref. [14] and shown elsewhere [4–6], the ac Stark shift should persist as if the six-wave mixing field were not present. In Fig. 3 we plot the MPI signal against the wavelength of the first laser. In collecting these data we detuned the second laser to both 10 and $-10\ \mathrm{cm}^{-1}$ from one-photon resonance; the pressure was set at $P = 5$ Torr. In the center of the figure we have plotted the resonantly enhanced four-photon MPI signal recorded with the second laser blocked. This line shape exhibits the same asymmetric line shape observed with copropagating laser beams.

When the second laser is turned on, the MPI line shape is significantly altered by the large ac Stark shifts due to this laser field. A multimode dye laser exhibits amplitude fluctuations due to the beating of different frequency modes. Correspondingly, the ac Stark shift is a very complicated function of time. At the power densities used in the present experiment, the average magnitude of the ac Stark shift due to the second laser for $\delta_2 < 10\ \mathrm{cm}^{-1}$ is very large compared with the laser bandwidth, and is also larger than the ac Stark shift due to the first laser. Correspondingly, when the first laser is detuned in the direction of the ac Stark shift, the laser field is shifted out of resonance at most times during the excitation pulse. However, if the detuning is not too large, the ac Stark shift will come within a laser bandwidth of being equal to δ_1 for brief periods of time during which the second laser power density is within the required range. During

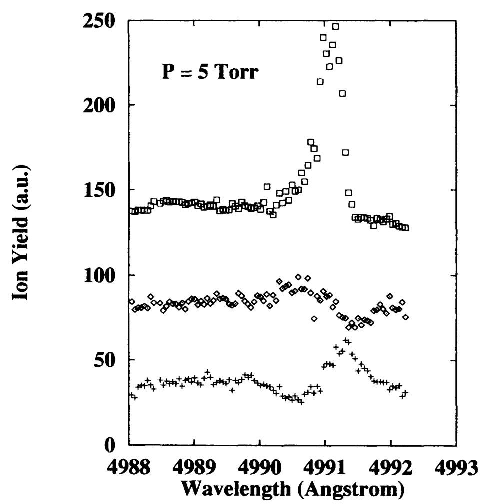

FIG. 3. Plot of the MPI signal against the wavelength of the first laser at $P = 5$ Torr in the counterpropagating configuration. (□□□) The first laser only (i.e., the IR beam is blocked). (+++) The IR laser tuned $+10\ \mathrm{cm}^{-1}$ away from the one-photon resonance. (◇◇◇) The IR laser tuned $-10\ \mathrm{cm}^{-1}$ away from the one-photon resonance. The three line shapes are successively offset by 50 units so each can be seen clearly.

these brief curve crossings resonance absorption occurs. At moderate power densities for the first laser, this process tends to make the dominant contribution to the MPI resonance enhancement. We expect that absorption will occur over the full range of the ac Stark shift due to the second laser, but the amplitude of the signal should be correspondingly lower. When the second laser is not blocked, the MPI signal should be shifted and greatly broadened, the amplitude of the signal being lower in inverse proportion to the added width. The direction of the shift depends on the sign of δ_2. When the ac Stark shift due to the first laser and that of the second laser are in opposite directions they tend to cancel. In this case the reduction in signal is smaller than when the shifts are added together. The expected effects are seen in Fig. 3. Similar results were obtained at elevated pressures, for instance at $P = 25$, 125, 150, and 250 Torr.

The theory used in Ref. [14] assumes that the Rabi frequency between $|1\rangle$ and $|2\rangle$ is very small compared with $|\delta_2|$, and that $|\delta_1| \ll |\delta_2|$, so that the amplitude for $|2\rangle$ can be adiabatically eliminated. We find, however, that the suppression occurs for copropagating beams even when the second laser is tuned very close to the unshifted one-photon resonance so that the Rabi frequency is very large compared with the detuning from one-photon resonance. At 5 Torr the MPI line shape for this case looks exactly like Fig. 2. Very different qualitative results are observed in this case with counterpropagating laser beams. When the second laser is detuned by only $\pm 2\ \mathrm{cm}^{-1}$, the upper state is split into a doublet by the second laser. The MPI signal is then smallest near the unperturbed four-photon resonance, with a very broad peak occurring on each side. The peaks are somewhat asymmetrical due to the ac Stark shift of the first laser and the detuning from exact one-photon resonance. We see these effects in Fig. 4, where the MPI signal at $P = 125$ Torr is plotted for the counterpropagating configuration.

In conclusion, we have investigated the suppression of the ac Stark shifting of a four-photon resonance due to the destructive interference between different excitation pathways. The experimental observations verify the theoretical predictions in the pressure range studied. Although the theory is based on the assumptions of transform limited pulses and an adiabatic approximation, we found that the theory works well even when the second laser is very close to the one-photon resonance and both lasers are broad bandwidth devices.

This research was sponsored by National Science Foundation Grant No. PHY-9206769. We would like to acknowledge the assistance of Robert Hacker and Robert Deal, both of Georgia Southern University.

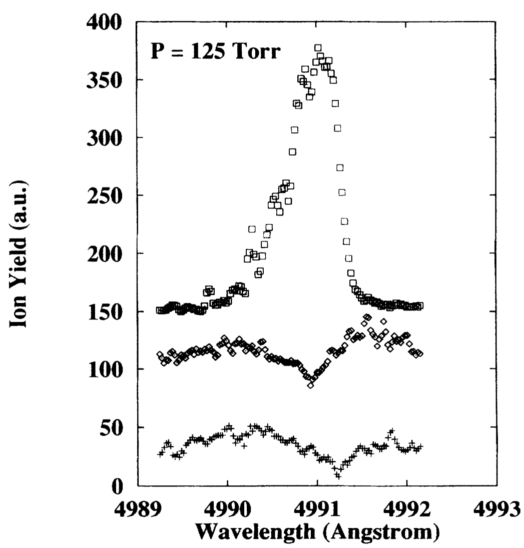

FIG. 4. Plot of the MPI signal against the wavelength of the first laser at $P = 125$ Torr in the counterpropagating configuration. (□□□) The first laser only (i.e., the IR beam is blocked). (+++) The IR laser tuned only $+2\ \mathrm{cm}^{-1}$ away from the one-photon resonance. (◇◇◇) The IR laser tuned only $-2\ \mathrm{cm}^{-1}$ away from the one-photon resonance. The three line shapes have been successively offset by 75 units so each can be seen clearly.

[1] R.N. Compton, J.C. Miller, A.E. Carter, and P. Kruit, Chem. Phys. Lett. **71**, 87 (1980).
[2] J.C. Miller, R.N. Compton, M.G. Payne, and W.R. Garrett, Phys. Rev. Lett. **45**, 114 (1980).
[3] D.J. Jackson and J.J. Wynne, Phys. Rev. Lett. **49**, 543 (1982).
[4] M.G. Payne, W.R. Garrett, and H.C. Baker, Chem. Phys. Lett. **75**, 468 (1980).
[5] M.G. Payne and W.R. Garrett, Phys. Rev. A **28**, 3049 (1983).
[6] M.G. Payne, W.R. Ferrell, and W.R. Garrett, Phys. Rev. A **27**, 3053 (1982).
[7] D.J. Jackson, J.J. Wynne, and P.H. Kes, Phys. Rev. A **28**, 781 (1983).
[8] G.S. Agarwal and S.P. Tewari, Phys. Rev. A **29**, 1922 (1984).
[9] M. Pourier, Phys. Rev. A **27**, 934 (1982).
[10] J.J. Wynne, Phys. Rev. Lett. **52**, 751 (1984).
[11] M.G. Payne and W.R. Garrett, Phys. Rev. A **42**, 1434 (1990). See also, W.R. Garrett, Roger C. Hart, J.C. Wray, Irene Datskou, and M.G. Payne, Phys. Rev. Lett. **64**, 1717 (1990).
[12] M.G. Payne, W.R. Garrett, Roger C. Hart, and Irene Datskou, Phys. Rev. A **42**, 2756 (1990).
[13] M.G. Payne, J.C. Miller, R.C. Hart, and W.R. Garrett, Phys. Rev. A **44**, 7684 (1991). See also, R.C. Hart, W.R. Garrett, and M.G. Payne, Phys. Rev. A **46**, 4213 (1992).
[14] M.G. Payne, J.Y. Zhang, and W.R. Garrett, Phys. Rev. A **48**, 2334 (1993).

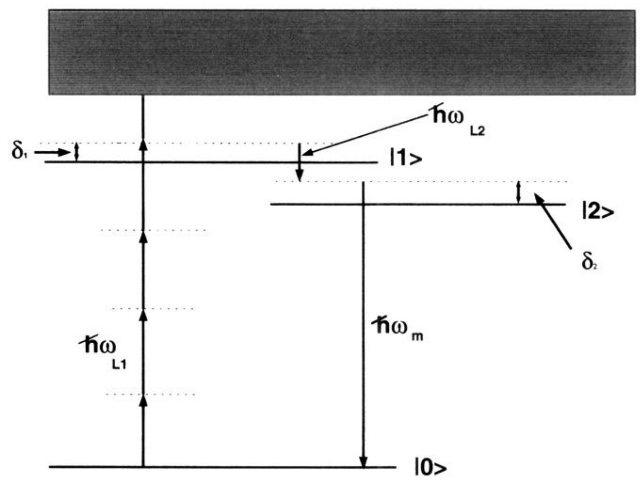

FIG. 1. Energy-level diagram defining the detunings involved in the MPI excitation of the two excited states.