Effect of quantum interference on the suppression of the ac Stark shifting of a multiphoton resonance

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We report detailed experimental results in xenon that confirm theoretical predictions [Payne *et al.*, Phys. Rev. A **48**, 2334 (1993)] on the interference-based suppression of ac Stark shifting of even-photon resonances under circumstances where the shift is produced through strong coupling to a state that is also coupled back to the ground state. In the copropagating-beam configuration and in the pressure range of 0.5–250 Torr, the multiphoton ionization spectral line shape is unchanged regardless of the presence of the second laser field, i.e., the ac Stark shift introduced by the second laser is totally suppressed due to a complete destructive interference between two excitation pathways. In the counterpropagating-beam configuration, however, the ac Stark shift persists in the range of the pressure studied. By choosing a different set of energy levels where no four-wave-mixing field is permitted, we show that the very same ac Stark shift introduced by the second laser field persists even with copropagating-beam configuration, as predicted by theory.

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I. INTRODUCTION

The suppression of three-photon resonantly enhanced multiphoton ionization (MPI) due to the quantum interference effect (QIE) was first reported in the early 1980s [1–3]. In a series of studies involving argon, krypton, and xenon at elevated concentrations Compton *et al.* [1] and Miller *et al.* [2] observed a total suppression of resonantly enhanced MPI processes due to different optical harmonics. Payne and co-workers [4–6] and others [7–10] have developed theories that explain the above and a number of related effects [11–13] in terms of a destructive quantum interference that occurs between different excitation pathways. Since then many QIE-related phenomena have been discovered, including the suppression of multiphoton excitation of one-photon allowed transitions by an interference from the sum or difference-mixing field [3–19], where such transitions are pumped by an odd number of photons; the suppression of resonant three-photon excitation due to broadening and optical shifting produced by amplified spontaneous emission (ASE) and/or stimulated hyper-Raman (SHR) [20,21]; the suppression of the resonant two-photon excitation of an even-parity transition involving laser driven two-photon resonant four-wave-mixing or internally generated parametric four-wave-mixing field (PFWMF) [22,23] and the suppression of the forward gain of optically pumped stimulated emission [24]. Many of these suppressions of the MPI processes due to the quantum interference effect are well understood. For instance, it has been well established both theoretically and experimentally that the suppression of odd-photon excitation of an optically allowed transition results from the influence of an internally generated mutliwave-mixing field at and near to the resonant frequency. This multiwave-mixing field evolves in such a way that the pumping of the resonant transition by the generated field is equal in magnitude but 180° out of phase with that due to the direct odd-photon pumping. Recently, Payne, Zhang, and Garrett [25] have shown that the very same destructive quantum interference could influence a supposedly interference-free system. Their study showed that at elevated concentrations when the ground state of a three-level system is coupled with the first excited state via an even-photon process while this excited state is coupled with a second excited state via a one-photon process the very same destructive interference between different pumping pathways could occur for suitable beam geometry only if the second excited state has a dipole-allowed transition back to the ground state. As a result of this destructive interference the ac Stark shift introduced by the second laser can become totally suppressed. The system studied by Payne *et al.* does not appear, at first glance, to support the possibility of the destructive quantum interference process since it has been assumed and accepted that the presence of *an intermediate even-photon resonance* would spoil the occurrence of the destructive interference. One would think that an amplitude developed by even-photon excitation would establish a population in the intermediate even-photon resonance which would be involved in developing an amplitude for the excited state in the odd-photon resonance. On the other hand, the coherent art of the odd-photon coupling plays the dominant role in the generation of the multiwave-mixing field. This would supposedly destroy the very special amplitude for the excited state in the odd-photon resonance. On the other hand, the coherent part of the odd-photon coupling plays the dominant role in the generation of the multiwave-mixing field. This would supposedly destroy the very special amplitude and phase relation between two excitation pathways. Following Payne's work Deng *et al.* [26] have observed in a subsequent experiment the persistence of the odd-photon interference effect and suppression of ac Stark shift predicted in xenon.

We have conducted a series of experiments and corresponding calculations for the system proposed by Payne *et al.* in which we studied the suppression of ac Stark shifts due to a destructive quantum interference effect at various xenon concentrations using both four-photon and two-photon pumping schemes. In this paper we report our results. We first present a brief review of the theoretical model of QIE-based suppression of an ac Stark shift in Sec. II. Readers interested in this subject should consult Ref. [25] for delineated numerical methods and results. The experimental considerations and setup are discussed in Sec. III. Experimental results obtained with different pumping schemes and discussions on these results are the subject of Sec. IV, and finally in Sec. V we present a summary.

II. A BRIEF REVIEW OF THE THEORETICAL MODEL OF QIE-BASED SUPPRESSION OF AC STARK SHIFTS

Payne, Zhang, and Garrett [25] have proposed a theoretical model and shown how the QIE-based suppression of ac Stark shifts may be calculated and understood. Here, we follow their method and notations.

We consider a situation in which a three-level plus continuum atomic system interacts with two laser fields of different frequencies. The first laser couples the ground state $|0\rangle$ and the first excited state $|1\rangle$ through a two-photon process and the second laser couples the fist excited state $|1\rangle$, through a one-photon process, with a second excited state $|2\rangle$ which has a dipole-allowed transition back to the ground state. An energy-level diagram for our study is shown in Fig. 1.

Let the electric fields at various frequencies be given as follows:

$$\hat{\mathbf{E}}_1=\hat{\mathbf{e}}E_1^{(0)}\cos[k(\omega_{L1})x-\omega_{L1}t+\alpha_1] ,$$
$$\hat{\mathbf{E}}_2=\hat{\mathbf{e}}E_2^{(0)}\cos[k(\omega_{L2})x-\omega_{L2}t+\alpha_2] ,$$
$$\hat{\mathbf{E}}_m=\hat{\mathbf{e}}E_m^{(0)}\cos[k(\omega_m)x-\omega_m t] ,$$

where

$$\omega_m=2\omega_{L1}-\omega_{L2} ,$$

and $\hat{\mathbf{e}}$ is the unit vector parallel to the z axis. We define various Rabi frequencies as follows:

$$\Omega_{12}^{(1)}=D_{12}E_2^{(0)}/(2\hbar) ,$$
$$\Omega_{02}^{(1)}=D_{02}E_m^{(0)}/(2\hbar) .$$

Following Payne *et al.* we use the time-dependent Schrödinger equation with a wave function of the form

$$|\Psi(x,t)\rangle=a_0e^{-i\omega_0 t}|0\rangle+a_1e^{-i\omega_1 t}|1\rangle+a_2e^{-i\omega_2 t}|2\rangle + \sum_\mu\int dE\, C_\mu(E,x,t)e^{-iEt/\hbar}|E\rangle . \quad (2.1)$$

Using the usual adiabatic elimination method we obtain the following set of differential equations on the amplitudes of various states:

$$\left[\frac{\partial a_0}{\partial t}\right]_x=i\Omega_{01}^{(2)}A_1+i\Omega_{02}^{(1)}A_2 , \quad (2.2a)$$

$$\left[\frac{\partial A_1}{\partial t}\right]_x=i\delta_1A_1+i[\Omega_{01}^{(2)}]^*a_0 +i\Omega_{12}^{(1)}e^{i\Delta k_r x}A_2-\frac{\Gamma_{I1}}{2}A_1 , \quad (2.2b)$$

$$\left[\frac{\partial A_2}{\partial t}\right]_x=i\delta_2A_2+i\Omega_{12}^{(1)}e^{-i\Delta k_r x}A_1 +i[\Omega_{02}^{(1)}]^*a_0-\frac{\Gamma_{I2}}{2}A_2 . \quad (2.2c)$$

In writing Eq. (2.2) we have used the quantities

$$A_1=a_1e^{i\delta_1 t}e^{-2ik(\omega_{L1})x} ,$$
$$A_2=a_2e^{i\delta_2 t}e^{-ik_r(\omega_m)x} , \quad (2.3a)$$

with

$$\omega_m=2\omega_{L1}-\omega_{L2} ,$$
$$\delta_1=2\omega_{L1}-(\omega_1-\omega_0) , \quad (2.3b)$$
$$\delta_2=\omega_m-(\omega_2-\omega_0) .$$

In Eqs. (2.2) Γ_{I1} and Γ_{I2} are photoionization rates of states $|1\rangle$ and $|2\rangle$. The phase mismatch is given by

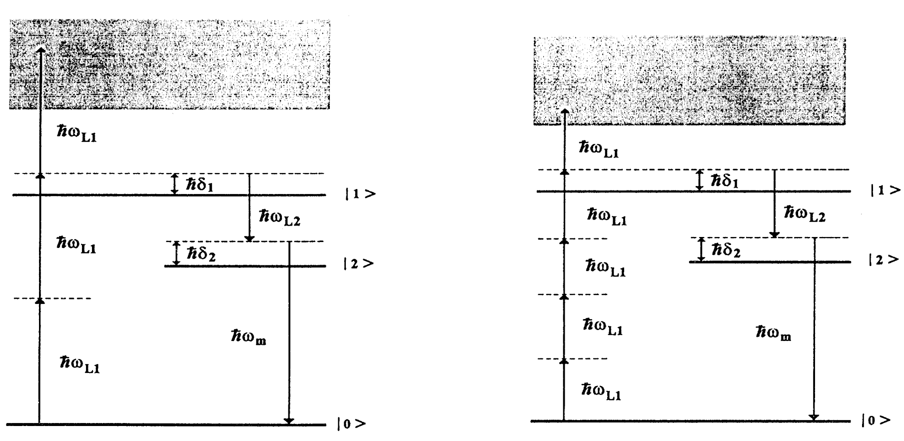

FIG. 1. Energy-level diagram defining detunings for various transitions.

$\Delta k = k(\omega_m) - [2k(\omega_{L1}) + k(\omega_{L2})] \equiv \Delta k_r + i\beta/2$ for the copropagating-beam configuration. In this expression of complex phase mismatch, Δk_r is the real part of the phase mismatch and β is the absorption coefficient at ω_m.

Our objective is to evaluate the new amplitudes A_1 and A_2. We start from Eqs. (2.2). We neglect the depletion of the ground state by using $a_0 \approx 1$ and adopt $[\partial A_2/\partial t]_x \approx 0$ by assuming that δ_2 is much larger than δ_1 so that amplitude A_2 adiabatic follows. This gives

$$A_2(x,t_r) \approx -\frac{[\Omega_{12}^{(1)}]^* e^{-i\Delta k_r x} A_1 + [\Omega_{02}^{(1)}]^*}{\delta_2 + i(\Gamma_{I2}/2)} . \quad (2.3c)$$

If we substitute Eq. (2.3c) into Eq. (2.2b) and recognize that $\Delta_s \equiv |\Omega_{12}^{(1)}|^2/\delta_2$ as an ac Stark shift introduced by the second laser field we get

$$\left[\frac{\partial A_1}{\partial t_r}\right]_x = i(\delta_1 - \Delta_s + i\Gamma_{I1}/2)A_1 + i[\Omega_{01}^{(2)}]^* - i\frac{\Omega_{12}^{(1)}(\Omega_{02}^{(1)})^*}{\delta_2 + i(\Gamma_c + \Gamma_{I2}/2)} e^{i\Delta k_r x} . \quad (2.4)$$

In order to solve Eq. (2.4) and to discuss the interference effects on amplitudes A_1 and A_2, one must know the Rabi frequency of the four-wave-mixing field, i.e., $\Omega_{02}^{(1)}(x,t_r)$. This is achieved by solving the Maxwell equations satisfied by the four-wave-mixing field.

Letting P_m be the polarization of the medium at the angular frequency ω_m, we have

$$\begin{aligned} P_m &= N\langle \Psi(x,t)|\hat{D}|\Psi(x,t)\rangle \\ &= P_m^+ + \text{c.c.} \\ &= ND_{02} A_2 \exp[ik(\omega_m)x - i\omega_m t] + \text{c.c.} , \end{aligned} \quad (2.5)$$

where N is the concentration of the atomic gas under study, $\hat{\mathbf{D}}$ is the electric dipole operator, and the time-dependent state vector is given by Eq. (2.1). The four-wave-mixing field generated in the medium must satisfy the wave equation

$$\left[\frac{\partial^2 E_m}{\partial x^2}\right] - \frac{(1+4\pi\chi_0)}{c^2}\frac{\partial^2 E_m}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 P_m^{NL}}{\partial t^2} , \quad (2.6)$$

where the linear terms in E_m and P_m have been moved to the left to be part of the linear susceptibility χ_0 at the mixing frequency ω_m.

We now express the four-wave-mixing field as

$$E_m = E_m^+ + E_m^- ,$$

and use the slowly varying amplitude and phase approximation in Eq. (2.6). In doing so we use the nonlinear polarization on the right-hand side in Eq. (2.5) with the amplitude A_2 given by Eq. (2.3c). This gives

$$\left[\frac{\partial E_0}{\partial x}\right]_{t_r} + \frac{1}{2}\beta E_0 = -4\pi i\frac{\omega_m}{c} ND_{02}(\Omega_{12}^{(1)})^* \times \frac{A_1(x,t_r+x/v)}{\delta_2 + i(\Gamma_c + \Gamma_{I2}/2)} e^{-i\Delta k_r x} . \quad (2.7)$$

In writing Eq. (2.7) we have used the notations

$$\begin{aligned} E_m^+ &= \frac{E_m^{(0)}}{2}\exp[ik_r(\omega_m)x - i\omega_m t] , \\ E_m^{(0)}(x,t) &\equiv E_0(x,t_r) \equiv E_m^{(0)}(x,t_r+x/v) , \end{aligned} \quad (2.8)$$

where $v = c/\sqrt{1+4\pi\,\text{Re}(\chi_0)}$, and $t_r = t - x/v$.

We now multiply Eq. (2.7) by $(D_{02})^*/2\hbar$ and obtain

$$\left[\frac{\partial(\Omega_{02}^{(1)})^*}{\partial x}\right]_{t_r} + \frac{\beta}{2}(\Omega_{02}^{(1)})^* - \int_{-\infty}^{t_r} dt' F(t_r,t')(\Omega_{02}^{(1)})^* = -(\Delta k - \Delta k_0)\exp(-i\Delta k_r x)(\Omega_{12}^{(1)})^* \int_{-\infty}^{t_r} dt' [\Omega_{01}^{(2)}(t')]^* \exp\left[i\int_{t'}^{t_r} dt''[\delta_1 - \Delta_s(t'')]\right] , \quad (2.9)$$

where

$$\Delta k = \Delta k_0 - \frac{\kappa_{02}}{\delta_2 + i(\Gamma_c + \Gamma_{I2}/2)} = \Delta k_r + i\frac{\beta}{2} , \quad (2.10a)$$

with

$$\kappa_{02} = \frac{2\pi\omega_m N|D_{02}|^2}{\hbar c} \quad (2.10b)$$

and

$$F(t_r,t') = (\Delta k - \Delta k_0)\frac{[\Omega_{12}^{(1)}(t_r)]^*}{\delta_2 + i(\Gamma_c + \Gamma_{I2}/2)}\Omega_{12}^{(1)}(t')\exp\left[i\int_{t'}^{t_r} dt''[\delta_1 - \Delta_s(t'')]\right] . \quad (2.10c)$$

Equation (2.9) yields a solution of the form

$$(\Omega_{02}^{(1)})^* = Y(x,t_r)e^{-i\Delta k_r x} , \quad (2.11a)$$

where

$$Y(x,t_r)=[\Omega_{12}^{(1)}(t_r)]^*\int_{-\infty}^{t_r}dt'e^{i\delta_1(t_r-t')}e^{-i\nu(t_r,t')}[\Omega_{01}^{(2)}]^*Z(\Delta kx,\Delta k_0x,\nu(t_r,t')) \tag{2.11b}$$

and

$$Z(\Delta kx,\Delta k_0x,\nu(t_r,t))=\frac{(\Delta kx-\Delta k_0x)}{2\pi}\int_{-\infty+i\varepsilon}^{+\infty+i\varepsilon'}dq\frac{e^{-iq}}{q(\Delta kx+q)}\exp\left[i\nu(t_r,t)\frac{(\Delta kx-\Delta k_0x)}{(\Delta kx+q)}\right] . \tag{2.11c}$$

In these equations $\nu(t_r,t)-\int_t^{t_r}dt'\Delta_s(t')$ is the integrated ac Stark shift.

We now write the formal solution of Eq. (2.4) as

$$A_1=i\int_{-\infty}^{t_r}dt'\left[[\Omega_{01}^{(2)}(t')]^*-\frac{\Omega_{12}^{(1)}(t')[\Omega_{02}^{(1)}(t')]^*}{\delta_2+i(\Gamma_c+\Gamma_{I2}/2)}e^{i\Delta k_rx}\right]$$

$$\times\exp\left[i\int_{t'}^{t_r}dt''[\delta_1-\Delta_s(t'')]\right] , \tag{2.12}$$

with $[\Omega_{02}^{(1)}(t')]^*$ given by Eq. (2.11). By interchanging the order of the two time integrations and after a bit of algebra we obtain

$$A_1(x,t_r)=-\int_{-\infty}^{t_r}dt'e^{i\delta_1(t_r-t)}[\Omega_{01}^{(2)}(t')]^*$$

$$\times e^{-i\nu(t_r-t)}S(\Delta kx,\nu(t_r,t')) , \tag{2.13}$$

where

$$S(\alpha,\mu)=\frac{1}{2\pi}\int_{-\infty+\varepsilon'}^{+\infty+\varepsilon'}\frac{dq}{q}e^{-iq}\exp\left[\frac{i\alpha\mu}{\alpha+q}\right]$$

$$=|S(\alpha,\mu)|e^{i\Psi(\alpha,\mu)} . \tag{2.14}$$

We see in Eq. (2.13) that the amplitude A_1 is expressed in terms of the Rabi frequency for two-photon excitation multiplied by a complex function S which depends both on the phase mismatch and on the ac Stark shift introduced by the second laser field. As Payne *et al.* have shown, when $-\text{Re}(\alpha=\Delta kx)\gg1$ the phase of the function $S(\alpha,\mu)$ approaches the asymptotic limit $\Psi\approx\mu-\pi/2$. Under this limit the phase factor of the function $S(\alpha,\mu)$ will cancel that from the ac Stark shift. That is, $S(\alpha,\mu)\equiv S(\Delta kx,\nu)\rightarrow|S|e^{i(\nu-\pi/2)}$ where $\nu=\nu(t_r,t)$ is the integrated Stark shift. Moreover, when $-\text{Re}(\alpha)\gg1$ is satisfied it has been shown in Ref. [25] that $|S(\alpha,\mu)|\rightarrow1$ is also satisfied. Consequently, the amplitude A_1 is reduced to the form

$$A_1(x,t_r)=i\int_{-\infty}^{t_r}dt'[\Omega_{01}^{(2)}(t')]^*e^{i\delta_1(t_r-t')} . \tag{2.15}$$

This is just the solution to Eq. (2.2b) in the absence of the ac Stark shift and ionization term. Thus we conclude that when the magnitude of the real part of the phase mismatch for the multiwave-mixing field is large one has $\Psi\approx\mu-\pi/2$ and $|S(\alpha,\mu)|\rightarrow1$; consequently the ac Stark shift introduced by the second laser field will be initially suppressed.

The evaluation of the amplitude A_2 under these same conditions may be easily done by taking $[\partial A_2/\partial t]_x\approx0$ in Eq. (2.2c), and we immediately see that

$$A_2(x,t_r)\approx-\frac{[\Omega_{12}^{(1)}]^*e^{-i\Delta k_rx}A_1+[\Omega_{02}^{(1)}]^*}{\delta_2+i(\Gamma_{I2}/2)} . \tag{2.16}$$

It is easily seen from this expression that a destructive interference may occur between the three-photon pumping of $|0\rangle\rightarrow|1\rangle\rightarrow|2\rangle$ transitions involving both laser fields, represented by $[\Omega_{12}^{(1)}]^*e^{-i\Delta k_rx}A_1\propto[\Omega_{12}^{(1)}]^*[\Omega_{01}^{(2)}]^*$, and the one-photon pumping $|0\rangle\rightarrow|2\rangle$ involving the four-wave-mixing field, represented by $[\Omega_{02}^{(1)}]^*$, provided these two contributions are equal in magnitude and 180° out of phase. Indeed, in the limit of $|\Delta k|\gg|\Delta k_0|$ and $\Psi\approx\mu-\pi/2,|S(\alpha,\mu)|\rightarrow1$, using A_1 given in Eq. (2.15) and Eqs. (2.11a)–(2.11c) one immediately finds that the numerator of Eq. (2.16) is equal to zero:

$$[\Omega_{12}^{(1)}]^*e^{-i\Delta k_rx}A_1+[\Omega_{02}^{(1)}]^*=ie^{-i\Delta k_rx}[\Omega_{12}^{(1)}]^*\int_{-\infty}^{t_r}dt'[\Omega_{01}^{(2)}(t')]^*e^{i\delta_1(t_r-t')}[1-ie^{-i\nu(t_r,t')}S(\Delta kx,\nu(t_r,t'))]\rightarrow0 .$$

Numerical evaluations of A_2 also show that at elevated concentrations this amplitude is strongly suppressed compared with the value obtained when the four-wave-mixing field is neglected.

When the phase mismatch is small the suppression of the ac Stark shift due to three-photon pumping involving both laser fields and one-photon excitation due to the four-wave-mixing field is not complete. However, when $-\text{Re}(\alpha)\ll1$ and $|S(\alpha,\mu)|\ll1$ a different destructive interference may occur, leading to the suppression of two-photon excitation. This can be easily seen from Eqs. (2.12) and (2.13). For very small phase mismatch $|\Delta k_r|x$, we have $|S(\Delta kx,\nu(t_r,t))|\ll1$, resulting in the cancellation between two-photon pumping of the $|0\rangle\leftrightarrow|1\rangle$ transition and two one-photon excitations by the second laser and the four-wave-mixing field.

III. APPARATUS AND EXPERIMENTAL CONSIDERATIONS

The ideal choice of energy levels to test the theory of the QIE-based suppression of ac Stark shifts would be one where level $|2\rangle$ gives rise to the ac Stark shift but where the second laser does not lead to an enhancement to the ionization signal by a one-photon process involving the second laser field. Such a situation exits in xenon. In Fig. 2 we show relevant energy levels of xenon selected for the present experiment. We coupled the ground state

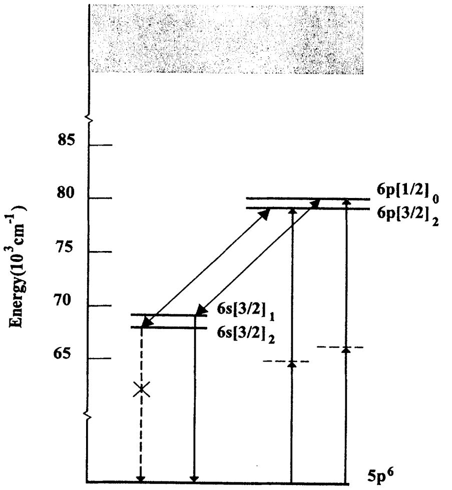

FIG. 2. Relevant xenon energy levels used in experiments.

$5p^6(J=0)$ with the excited state $5p^56p[\frac{1}{2}](J=0)$ by a two- or four-photon resonant process. The second dye laser was used to couple the $5p^56p[\frac{1}{2}](J=0)$ state with a lower lying second excited state $5p^56s[\frac{3}{2}](J=1)$. The latter state has a strong dipole-allowed transition back to the ground state, as required for the predicted suppression of the ac Stark shifts due to the second laser.

In the first set of experiments a Quanta Ray 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 laser was used to pump a dye laser containing C-500 dye, while the second harmonic of the Nd:YAG laser was used to pump a dye laser using LDS-821 dye. The outputs of the two dye lasers, after suitable optical treatment, were focused into a gas 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 the *copropagating* or the *counterpropagating* configurations. The output energy of the first dye lasers was $E_1=0.5$ to 1 mJ at 499.13 nm and was used to couple the ground state $|0\rangle$ and the excited state $|1\rangle$ through a four-photon resonant process. The output of the second dye laser was $E_2=2$ to 3 mJ at about 828.24 nm. This laser was used to couple the excited state $|1\rangle$ and a lower lying excited state $|2\rangle$. Both beams were focused to a beam waist of 250 μm and overlapped both spatially and temporally. We investigated the MPI ion yield in the pressure range of 0.5–250 Torr by scanning the first laser from about 15 cm^{-1} on the high-energy side of four-photon resonance to 15 cm^{-1} on the low-energy side of the same transition, while the second laser was fixed in frequency at a detuning that was typically of the order of ± 10 cm^{-1} from the one-photon resonance. The experimental setup is depicted in Fig. 3.

In the second set of experiments we made use of a two-photon resonant excitation scheme using dye lasers with narrower linewidths. A Quanta Ray DCR-1 A Q-switched Nd:YAG laser was again used to pump two dye lasers. The third harmonic of the Nd:YAG laser, pulse length about 4 ns, was used to pump a LUMIONICS

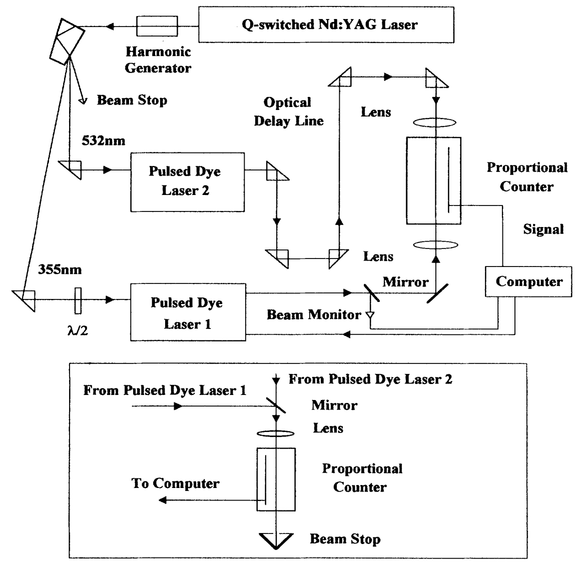

FIG. 3. Experimental setup for studying the ac Stark shifting of two- and four-photon resonances. Inset: setup for copropagating-beam configuration.

HyperDye-300 pulsed dye laser (bandwidth 0.05 cm^{-1}) using C-500 dye, while the second harmonic of the Nd:YAG laser was directed into a Spectra-Physics PDL-2 pulsed dye laser (bandwidth 0.2 cm^{-1}) using LDS-821 dye. The output of the first dye laser was then frequency doubled by a crystal of β-barium borate (BBO) cut at 65° (CSK Ltd., conversion efficiency is about 15%) to reach a wavelength of 250 nm. The crystal may be angle tuned, either by hand or with a feedback-controlled system, to produce the excitation spectra. In the case of hand tuning, data may be smoothed with either a Fourier transform method with a filter function or three-point data smoothing routine. The laser beams at both 250 and 828 nm were focused into the same cell used previously. The output energies were $E_1=0.1$ mJ at 249.63 nm and $E_2=3.5$ to 4.5 mJ at 828.24 nm, respectively. The pressure of xenon gas was selected to be within the range of 0.5–200 Torr. We scanned the first laser from about 10 cm^{-1} on the high-energy side of the two-photon resonance to abut 10 cm^{-1} on the low-energy side of the same resonance, while the second laser was fixed in frequency at a detuning that was typically in the range of ± 10 cm^{-1} from the one-photon resonance.

An important and interesting test on the theory of QIE proposed by Payne *et al.* is to contrast the above situations with one in which states are chosen such that no four-wave-mixing field is allowed. This is the subject of the third set of experiments since such a system is also readily available in xenon (see Fig. 2). Here we make use of a two-photon resonance between the ground state $5p^6(J=0)$ and the excited state $5p^56p[\frac{3}{2}](J=2)$. The second laser was tuned to couple the excited state $5p^56p[\frac{3}{2}](J=2)$ and the lower lying excited state $5p^56s[\frac{3}{2}](J=2)$, which permits no four-wave-mixing field due to the selection rules. The lack of four-wave-mixing field will certainly destroy the quantum interference effect responsible for the suppression of the ac Stark shifts caused by the second laser.

Finally, it would be interesting to study the suppression of the ac Stark shift when the second laser is tuned very close to one-photon resonance. This situation is an interesting test of the QIE-based theory that is based on an adiabatic approximation which assumes that $|\delta_2| \gg |\Omega_{12}^{(1)}|^2$. In the fourth set of experiments we used a two-photon excitation scheme and typically detuned the second laser by only ± 1 to ± 2 cm^{-1} from the exact one-photon resonance.

IV. RESULTS AND DISCUSSIONS

A. Four-photon resonant excitation: multiwave-mixing field is allowed

Copropagating-beam configuration. In this setup, using parameters described in Sec. III we estimate that at 1 Torr $|\Delta k_r| b / \sqrt{|\Delta_s^{max} \tau|} \gg 10$ for a value of $|\delta_2| \leq 10$ cm^{-1}, where τ is of the order of the pulse length, b is the confocal parameter, and Δ_s^{max} is the maximum ac Stark shift. This estimate indicates that we are working in the region where the conditions $|S(\Delta kx, \nu(t_r,t'))| \to 1$ and $\Psi(\alpha,\beta) \approx \beta - \pi/2$ are well met. Consequently we expect to see the total suppression of the ac Stark shift in this configuration 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. 4 we plot the MPI signal versus the wavelength of the first laser at concentration of $P = 5$ 125 Torr. We observed no appreciable changes in MPI line shapes no matter whether the second laser was blocked or unblocked with any detunings δ_2 although a slight increase of the MPI signal strength was observed when the second laser was most intense.

Counterpropagation-beam configuration. It has been argued and shown in Refs. [4–6,25] that due to the different phase matching conditions with the counterpropagating-beam configuration the ac Stark shift should persist as if the six-wave-mixing field were absent. Figure 5 shows the MPI signal as a function of the wavelength of the first laser for concentrations of $P = 5$ and 250 Torr. In this situation, we detuned the second laser about ± 10 cm^{-1} away from the one photon resonance. In the center of each figure we have plotted the resonantly enhanced four-photon MPI signal recorded with the second laser blocked, which exhibits the similar asymmetric MPI line shape observed with the copropagating-beam configuration.

When the second laser is turned on, the MPI signal line shape is significantly altered by the large ac Stark shifts caused by this laser field. Due to the multimode nature of the dye laser used, the ac Stark shift is a complicated function of time and wavelength. At the power densities used in these experiments, the average magnitude of the ac Stark shift due of the second laser for $\delta_2 < 10$ cm^{-1} is very large compared with the laser bandwidth (about 0.05–0.3 cm^{-1}), and is also much larger than the ac Stark shift due to the first laser. Correspondingly, when

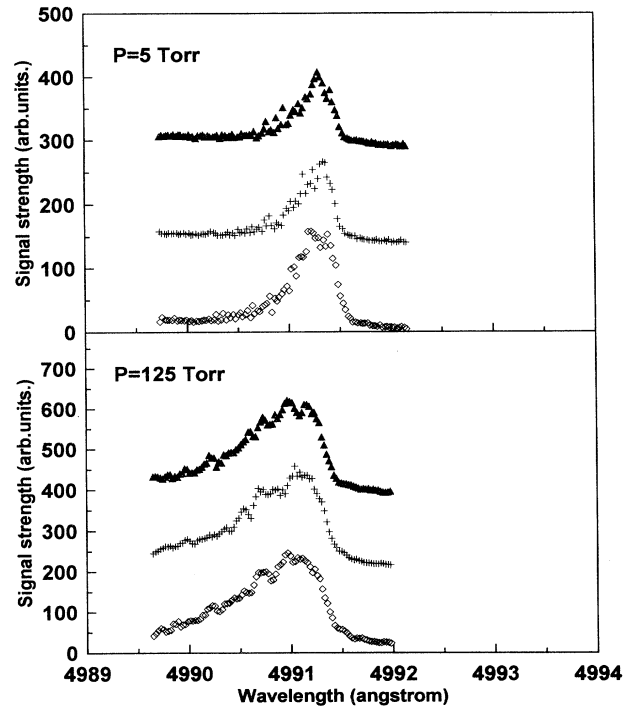

FIG. 4. Plot of the MPI signal against the wavelength of the first laser at $P = 5$ and 125 Torr for four-photon resonance in the copropagating-beam configuration. (◇◇◇) The first laser only. (+++) The second laser tuned to $+10$ cm^{-1} away from one-photon resonance between two excited states. (△△△) The second laser tuned to -10 cm^{-1} away from one-photon resonance. The three line shapes are offset so they can be distinguished.

the first laser is detuned in the direction of the ac Stark shift, the laser field is shifted out of the 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 for a brief period of time during which the second laser power density is within the required range. During these brief curve crossings resonance absorption occurs. At low or moderate power densities for the first laser, this process tends to make the dominant contribution to the MPI resonance enhancement (see discussion below). When the second laser is unblocked, the MPI signal should be shifted and greatly broadened, the amplitude of the signal should be lower and inversely proportional to the added width, and the direction of the shift depends on the sign of δ_2. These effects are seen in Fig. 5. Similar results were obtained at different pressures, for instance at $P = 0.5$, 50, and 125 Torr.

B. Two-photon resonant excitation: multiwave-mixing field is allowed

Copropagating- and counterpropagating-beam configurations. With dye lasers of narrower bandwidth (about 0.05 cm^{-1}) and with a β-barium borate frequency doubling crystal we repeated the above experiments with two-photon excitation for the coupling of the ground

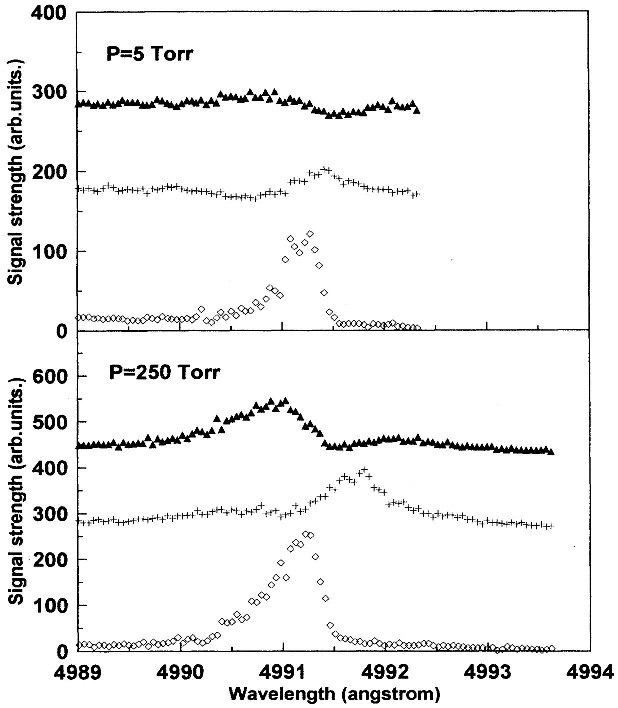

FIG. 5. Plot of the MPI signal against the wavelength of the first laser at $P=5$ and 250 Torr for four-photon resonance in the counterpropagating-beam configuration. (◇◇◇) The first laser only. (+ + +) The second laser tuned to $+10\ \mathrm{cm}^{-1}$ away from one-photon resonance between two excited states. (△△△) The second laser tuned to $-10\ \mathrm{cm}^{-1}$ away from one-photon resonance. Three line shapes are offset so they can be distinguished.

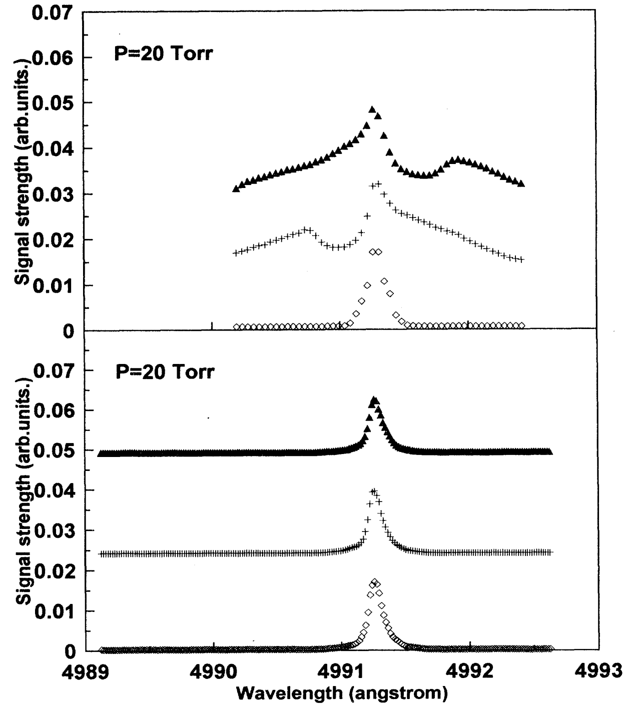

FIG. 6. Plot of the MPI signal against the wavelength of the first laser at $P=20$ Torr for two-photon resonance. The upper plot: counterpropagating-beam configuration. The lower plot: copropagating-beam configuration. (◇◇◇) The first laser only. (+ + +) The second laser tuned to $+10\ \mathrm{cm}^{-1}$ away from one-photon resonance between two exited states. (△△△) The second laser tuned to $-10\ \mathrm{cm}^{-1}$ away from one-photon resonance. Three line shapes are offset so they can be distinguished.

state $5p^6(J=0)$ and the excited state $5p^5 6p[\frac{1}{2}](J=0)$. An estimate similar to that given above shows that conditions for total suppression of the ac Stark shift were well met. Consequently, the effect of pressure-dependent quantum interference on the total suppression of the ac Stark shift should be very pronounced. In the copropagating-beam configuration the suppression of the ac Stark shift is due to the coherent cancellation between the three-photon absorption process involving both lasers and one-photon absorption involving the four-wave-mixing field. In Fig. 6 we plot the MPI signal versus the wavelength of the first laser at $P=20$ Torr for both copropagating- and counterpropagating-beam configurations. The second laser was detuned about $\pm 10\ \mathrm{cm}^{-1}$ from the one-photon resonance. Again, no appreciable change of spectra was observed for the copropagating-beam configuration, as described before. Since the two-photon pumping is much more efficient we were able to reduce the power density for the first laser, and these traces exhibit narrower linewidth with much smaller fluctuations and no apparent ac Stark shift due to the first laser. In the counterpropagating-beam configuration the presence of the second laser altered the lineshape of the MPI signals significantly. When the second laser was detuned about $10\ \mathrm{cm}^{-1}$ to the low-energy side of one-photon resonance a reduction of the MPI signal at longer wavelength was observed, as before. We have also observed a similar reduction of the MPI signal on the short wavelength side when the second laser was detuned about $10\ \mathrm{cm}^{-1}$ to the high-energy side of the one-photon resonance. The effects that are responsible for these reductions in the MPI signal with significantly altered line shapes have been discussed before [26]. Similar results were obtained at different pressures, for instance at $P=0.5$, 50, 125, and 200 Torr.

C. Comparison between a case in which multiwave-mixing field is forbidden and a case in which multiwave-mixing field is allowed

An interesting and still important piece of evidence that confirms the QIE-based suppression of ac Stark shift is the observation of the persistence of the ac Stark shift in the copropagating-beam configuration when a different set of energy levels of xenon is involved. We coupled the ground state $5p^6(J=0)$ and the excited state $5p^5 6p[\frac{3}{2}](J=2)$ with a two-photon process and tuned the second laser to couple the excited state $5p^5 6p[\frac{3}{2}](J=2)$ and the lower lying excited state $5p^5 6s[\frac{3}{2}](J=2)$. This set of levels permits no four-wave-mixing (FWM) field because of the selection rules. The one-photon transition coupled by the second laser has comparable oscillator strength to the case where the FWM field is allowed, and hence the second laser will generate a large ac Stark shift. We repeated the investigation of the effect of the second laser for the *copropagating-beam configuration* at a typical pressure of $P=20$ Torr. The effect of the second laser on the resonant multiphoton ionization signal was readily observable even with reduced power densities. On the upper plot in Fig. 7 we have shown the MPI signals versus the wavelength of the first laser. When the second laser was blocked the MPI signal due to the first laser was weak with a symmetric linewidth. Considerable change was observed for both signal strength and line shape when the second laser was unblocked. The second laser was set at typically 10 cm^{-1} on the lower-energy side of one-photon resonance. When the second laser was turned on we observed more than a factor of 8 increase in MPI signals. It is important to note that this signal exhibits a line shape similar to various traces shown before with *counterpropagating-beam configuration*. The asymmetric ac Stark shifted line shape is evident in this trace, indicating the persistence of a large ac Stark shift introduced by high power density of the second laser. Similar results were obtained at different pressures.

As a direct comparison we reexamined the situation of two-photon excitation with four-wave-mixing field being allowed (see the description on the second set of experiments). We used the same power densities and beam configuration as in the above experiment. We observed a considerable change of the MPI signal strength when the second laser was turned on and off. However, we observed no appreciable change of the lineshape of the MPI signals. This set of data has been displayed in the lower plot in Fig. 7. When the second laser was blocked the MPI signals produced by the first laser had a symmetric line shape indicating no appreciable ac Stark shift caused by the first laser. The linewidth of this trace is $\lambda_{FWHM} \approx 0.075$ Å or about 0.3 cm^{-1}. When the second laser was turned on we observed a factor of 4 increase of the signal strength with slightly reduced linewidth $\lambda_{FWHM} \approx 0.06$ Å or about 0.2 cm^{-1}. To explain this we first note that the 250-nm radiation can ionize both states $|1\rangle$ and $|2\rangle$. In the situation described here, the second laser cannot photoionize either state with one photon. This leads to a situation where the suppression of the ac Stark shift may actually increase the ionization signal both because of the decrease linewidth and due to decreased suppression due to the two-photon cancellation

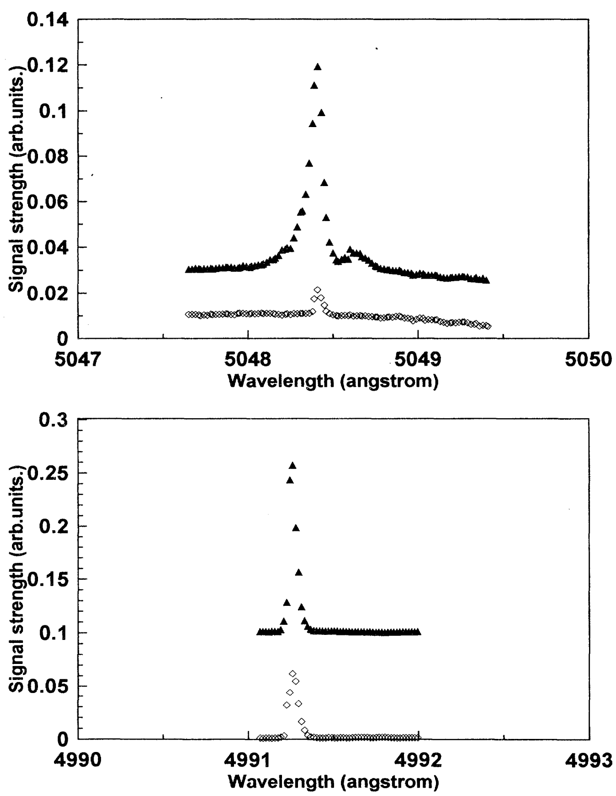

FIG. 7. Plot of the MPI signal against the wavelength of the first laser at $P=20$ Torr for two-photon resonance and in the copropagating-beam configuration. The upper plot: multiwave-mixing field is *not* allowed. The lower plot: multiwave-mixing field is allowed. (◇◇◇) The first laser only. (△△△) The second laser tuned to -10 cm^{-1} away from one-photon resonance. Line shapes are offset so they can be distinguished.

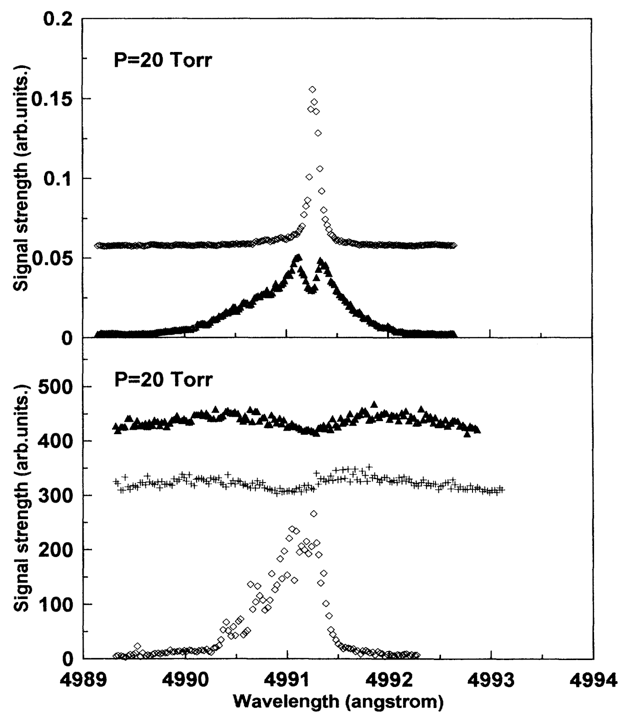

FIG. 8. Plot of the MPI signal against the wavelength of the first laser at $P=20$ Torr for both two- and four-photon resonances in the counterpropagating-beam configuration. The second laser tuned very close to one-photon resonance. The upper plot: two-photon resonance. (◇◇◇) The first laser only. (△△△) The second laser tuned to -2 cm^{-1} away from one-photon resonance. The lower plot: four-photon resonance. (◇◇◇) The first laser only. (+++) The second laser tuned to $+2$ cm^{-1} away from one-photon resonance between two excited states. (△△△) The second laser tuned to -2 cm^{-1} away from one-photon resonance. Line shapes are offset so they can be distinguished.

effect. Similar results were obtained at other elevated concentrations and different power densities.

D. Cases in which the second laser was detuned very close to one-photon resonance

The theory proposed in Ref. [25] assumes that the Rabi frequency between $|1\rangle$ and $|2\rangle$ is very small compared with $|\delta_2|$, and $|\delta_1| \ll |\delta_2|$, so the amplitude for $|2\rangle$ can be adiabatically eliminated. We found, 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 20 Torr the MPI signal line shapes for this case look exactly like Fig. 4 for the four-photon excitation and Fig. 6 for the two-photon excitation. Very different qualitative results are observed in this case with counterpropagating laser beams. When the second laser is detuned by only ± 1 to ± 2 cm^{-1}, the upper state is split into a doublet by the second laser. The MPI signal is then smallest near the unperturbed two- or four-photon resonance, with a broader peak occurring on each side. The asymmetries of these peaks, if any, are due to the ac Stark shift of the laser and the detuning from exact one-photon resonance. We see these effects in Fig. 8 where the MPI signal at $P = 20$ Torr has been plotted.

V. CONCLUSION

We have investigated the quantum interference effect on the suppression of the ac Stark shifting of both two-photon and four-photon resonances. In the pressure range studied, our experimental observations verify the theoretical predictions on the suppression of the ac Stark shift due to destructive interference between different excitation pathways. An important confirmation of the QIE-based theory is provided with the comparison between two copropagating-beam experiments in which one permits the multiwave-mixing fields but the other does *not.* In the latter case the ac Stark shift persists as predicted by the theory. Although the theory is based on the transform-limited pulse shape and adiabatic approximation, we found that the theory works well even when the second laser is very close to one-photon resonance and both lasers are broadband devices.

ACKNOWLEDGMENTS

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