Interference between nearly resonant three-photon excitation and third-harmonic generation probed by the cancellation of four-photon resonances

X. Xing,* D. Charalambidis,† E. Koutsourelaki, and C. Fotakis†
Foundation for Research and Technology-Hellas Institute of Electronics Structure & Laser, P.O. Box 1527, Heraklion 711 10, Crete, Greece

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The destructive interference between the three-photon and the third-harmonic one-photon absorption in the vicinity of the $6s[3/2]_1$ state of Xe has been studied in the resonantly enhanced multiphoton ionization via nf states, through the cancellation of the nf, $J=2$ resonances of the spectrum, accessible by four-photon absorption. The gas-pressure, i.e., phase-matching, dependence of the three-photon detuning range from the $6s[3/2]_1$ state for which cancellation occurs has been investigated in one- and two-color experiments, including positive detuning from this state in positively dispersive regions. For obtaining cancellation, the theoretically predicted requirement of optically thick medium at the third-harmonic frequency is corroborated by present observations for negative detunings. The different cancellation behavior of the $nf[3/2]_2$ and $nf[5/2]_2$ states observed indicates the dependence of the effect on the atomic parameters of the coupled four-photon resonance. The experimental results are found to be in partial agreement with existing theoretical models.

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INTRODUCTION

Upon appropriate phase-matching conditions, third-harmonic generation (THG) of a tunable-laser beam may occur in a gaseous medium. Once THG is produced, it can be reabsorbed by the medium together with photons of the fundamental frequency. Reabsorption may lead to an enhancement [1] or suppression of ionization, as, for example, has been experimentally observed some years ago in the cancellation of the $6s[3/2]_1$ resonance in the three-photon resonant five-photon ionization of Xe [2,3]. This cancellation effect was soon interpreted [4] as a destructive interference between the one- and three-photon absorption channels, where the linear process is due to the third-harmonic field. Several theoretical approaches to the problem [5] appeared subsequently.

In a recent communication [6] we reported another aspect of this effect. This was the destructive interference between THG and three-photon excitation negatively far detuned from the resonance state, experimentally probed through the cancellation of four-photon resonances. In that work it was also shown that the gas pressure must be higher than that resulting in optimum phase matching, that is, when only a very weak third harmonic is generated, for cancellation to occur. On the contrary, for pressures leading to good phase matching, an enhancement of ionization due to reabsorption of the THG was reported.

The theory of the observed effect, formulated in the density-matrix formalism, has appeared in the same issue as [6] postulating the following three conditions for cancellation to occur [7]: the pressure condition, that is, the pressure at which the absorption length for the THG becomes smaller than the interaction length (the medium is optically thick), and the single-state conditions, that is, the dominance of the resonant versus the nonresonant part of the first- and third-order susceptibilities and the dominance of the resonant versus the nonresonant ionization.

In the previous publication [6] we noted that the observed cancellation effect was related to the results reported some years ago by Blazewicz and Miller [8] in a two-color three-photon near-resonance, four-photon resonance excitation experiment. The ionization dips observed in the broad ionization features of their experiment were discussed [6] in terms of the same destructive interference occurring near the three-photon resonance. Very recently, Payne *et al.* [9] published an elaborate theoretical study on the influence of third-harmonic generation on such ionization schemes. Their calculations show ionization dips due to cancellation effects in four-photon resonances.

The present work describes in more detail the experimental results concerning the cancellation of four-photon resonances in one- (ω) and two- (ω_1,ω_2) color experiments, when three-photon (3ω or $3\omega_1$) excitation is near resonance and it interferes with the one THG photon (ω_{th}) excitation channel. In particular, the two-color experiments, due to their higher degree of state selectivity, allow control of the cancellation conditions by variation of the excitation wavelength at constant pressure, so that they may bring more insight as to the possible influence of the atomic parameters on the effect under investigation. Furthermore, by tuning the one laser so that three-photon excitation is in positively dispersing regions below the $6s[3/2]_1$ resonance, cancellation for positive detunings can be studied, as this is predicted by the theory, due to the Δ^2 dependence of the pressure condition for cancellation [7], Δ being the detuning from the three-photon resonance.

EXPERIMENTAL METHOD

The method used for the investigation of the near-resonance destructive interference between three-photon and third-harmonic one-photon absorption is the observation of the cancellation of four-photon resonances in the four-photon resonant, five-photon ionization spectrum of Xe due to the interruption of their excitation channel. Peak intensity considerations could be made through comparisons with resonances which for total angular momentum conservation reasons cannot be excited through the channel involving the third-harmonic photon (i.e., $J=4$ states) and hence cannot be cancelled. The manifestation of such cancellation effects in the ionization spectra is the restoration of the ionization peaks by means of counterpropagating beams originating from a split part of the initial laser beam [4,10]. This restoration is due to the additional channels to the two destructively interfering ones, which are introduced by the second beam. The situation is illustrated in Fig. 1. The photons of the second beam are indicated by ω'. The reason for including only channels involving one ω' photon is that in the present work the second beam was one order of magnitude less intense than the first one. As can be seen in Fig. 2, the $4f[3/2]_2$ resonance is cancelled (dashed line) in the one-beam spectrum and restored (solid line) when the counterpropagating beam is introduced. The relation between the pressure range and the detuning for which cancellation occurs has been investigated by choosing different four-photon resonances, for which different detuning from the near three-photon resonance (the $6s[3/2]_1$ in the present study) could be achieved.

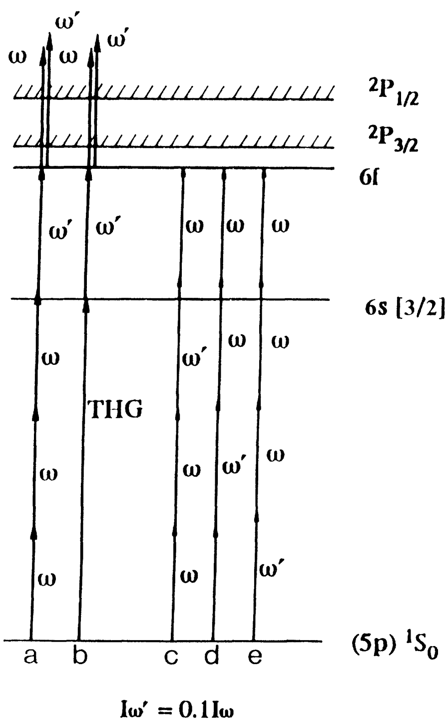


FIG. 1. Dominant ionization channels in the one beam [(a) and (b)] and in the two counterpropagating beam [(a), (b), (c), (d), and (e)] experiments.

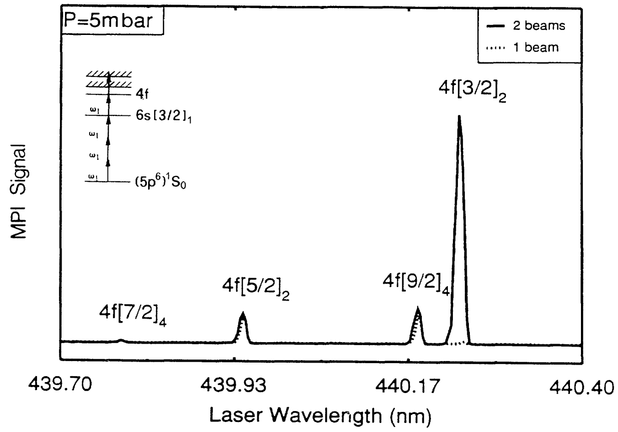


FIG. 2. Resonantly enhanced multiphoton ionization (REMPI) spectrum via the $4f$ state manifold of Xe taken with one (. . . .) and two counterpropagating (——) beams at 5-mbar Xe pressure. The cancelled $4f[3/2]_2$ resonance in the one-beam spectrum is restored upon introduction of the counterpropagating beam.

Further, for a more detailed insight about several aspects of the effects under investigation, which will be discussed later, two-color experiments have been performed. In these one color (ω_1) has been responsible for the three-photon excitation and THG (ω_{th}), while the second color (ω_2) has been chosen so as to match the excitation of the four-photon resonances ($3\omega_1+\omega_2$).

Furthermore, for a more complete picture of the effects under consideration, the THG profiles for several pressures have been recorded simultaneously with the ionization. These measurements give significant information about the interplay between THG and the three-photon excitation.

EXPERIMENTAL SETUP

The experimental setup of the type of experiments performed in this study is shown in Figs. 3(a), 3(b), and 3(c). In all three setups the tunable-laser beams were delivered by excimer (Lambda LPX 315) pumped dye lasers (Lambda FL 3002) of 15-ns pulse duration and 0.2-cm^{-1} bandwidth. In the one-color experiments [Fig. 3(a)] the laser beam was split via a quartz plate into two parts having an intensity ratio $I_{\omega'}/I_{\omega}\sim\frac{1}{10}$. The two laser beams after counterpropagating were focused via two $f=15$ cm lenses at the interaction region of the static cell filled with Xe. One beam and two counterpropagating beams are used in order to test for the presence of cancellation. In the one-color one-beam runs, care was taken that no backreflection from the exit window of the cell will interfere with the laser beam in the ionization area. This turns out to be important for the observation of the highest degree of cancellation. Ions were collected via a biased electrode. The ionization signal was amplified through a charge-sensitive preamplifier (Canberra 2004)-amplifier (Tennelec 243) stage, and integrated through a boxcar integrator. The boxcar output was digitized via an analog-to-digital converter and stored in the personal computer (PC). The whole experimental run was PC controlled. For the simultaneous observation of the THG profiles and ionization spectrum, a home made prism vacuum-ultraviolet (vuv) monochromator was employed [Fig. 3(b)]. It consisted of a second cell pumped with a turbomolecular pump (Alcatel 5010) to a vacuum of $\sim 10^{-6}$ mbar and vacuum interfaced to the ionization cell with a LiF window. THG was separated from the laser fundamental frequency with a LiF prism as the dispersive element of the monochromator and detected with a solar blind vuv photomultiplier tube [CR 1259 Hamamatsu photomultiplier tube (PMT)]. A broadband vuv transmission filter was placed in front of the PMT in order to reduce the scattered light of the fundamental frequency at the PMT. The geometry of the monochromator was such that without moving any element a range of about 3 nm of vuv light could be recorded. For detection of further spectral regions the PMT which was mounted on the cell with a flexible tube could be moved to a new position. The PMT signal was, similarly, amplified, integrated, digitized, and stored in the PC.

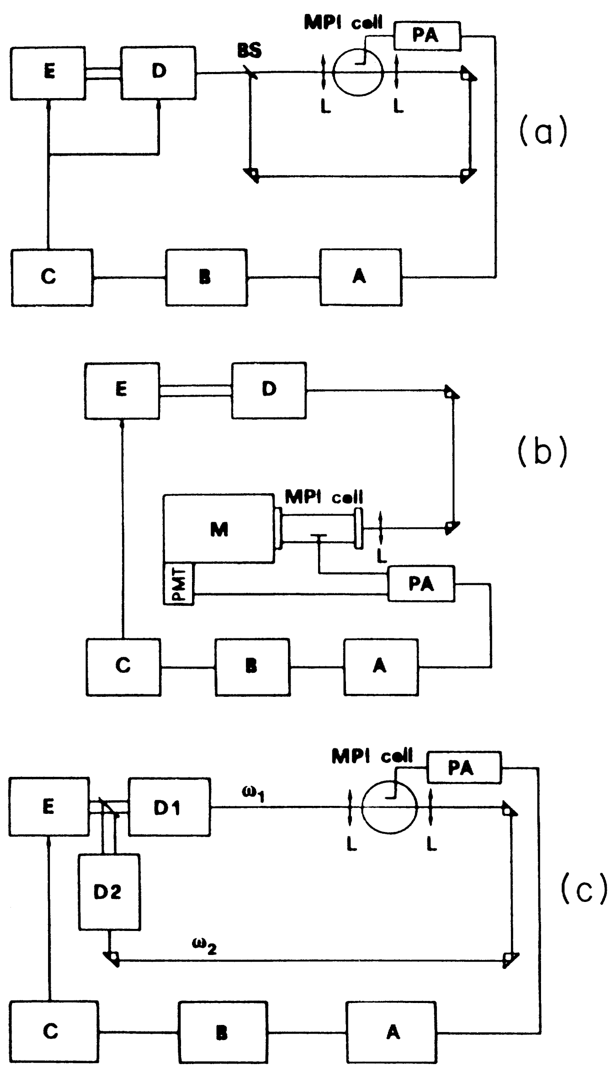


FIG. 3. Experimental setup of the (a) one-color two-beam ionization measurements, (b) one-color simultaneous ionization and THG measurements, and (c) two-color ionization measurements. E, excimer laser; D, dye laser; PA, preamplifier; A, amplifier; PMT, photomultiplier tube; M, vuv monochromator; L, lens; BS, beam splitter; B, boxcar; and C, computer.

Figure 3(c) shows the two-color-experiment arrangement. The two beams were counterpropagating and focused at the same point. The intensity of laser 2 was about one order of magnitude lower than that of laser 1. Good temporal overlap of the two beams at the focus was considered. In the last two types of experiments [Figs. 3(b) and 3(c)], laser 1 was not split into two parts, so that restoration of cancelled resonances could not be used as proof of cancellation. In all types of experiments the power density of the stronger laser beam was of the order of 1 GW/cm^2.

RESULTS AND DISCUSSION

One-color experiments

In the one-color experiments the manifolds of the $4f$ and $5f$ states have been excited via four-photon excitation, while a fifth photon ionizes the system. The corresponding detuning of the three-photon excitation from the $6s[3/2]_1$ state is ~ 100 and ~ 2000 cm^{-1}. The results of the one-color ionization experiments are demonstrated in Figs. 2 and 4. As can be clearly seen in the pressure dependence of the multiphoton ionization (MPI)

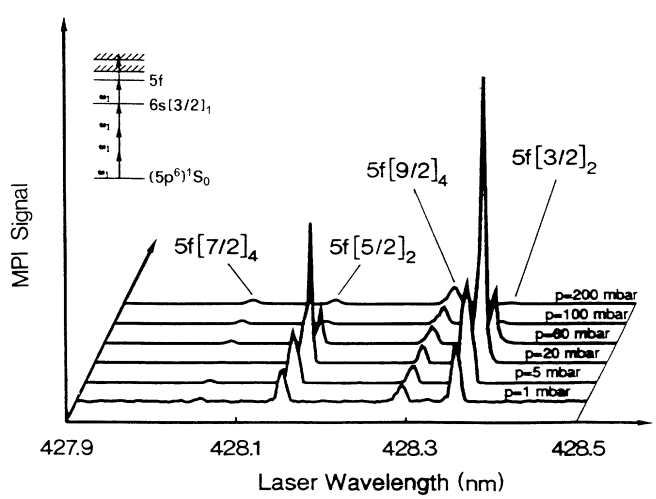


FIG. 4. Ionization spectra in the wavelength region 427.9–428.5 nm at different Xe pressures ranging from 1 to 200 mbar, showing the enhancement and cancellation of the two $5f$, $J=2$ states. Spectra are normalized, so that the $5f[9/2]_4$ peaks have the same height in all spectra for relative intensity comparison reasons.

spectra through the $5f$ manifold, the $J=2$ resonances start undergoing partial cancellation at pressures ≥ 60 mbar, while in the pressure range between 5 and 60 mbar, an enhancement of the $J=2$ peaks takes place, this being depicted in the spectrum taken at 20 mbar of Fig. 4. It must be noted that the scales of the different curves in Fig. 4 are not the same, so that the peaks which do not exhibit interference ($J=4$ peaks) appear having the same height. This is in order to facilitate peak-intensity comparisons, as discussed above, since no absolute ionization intensities have been measured. The cancellation of the $4f[3/2]_2$ resonance at 5 mbar is demonstrated in Fig. 2. The same spectrum taken with two counterpropagating beams shows the restoration of the cancelled peak. Restoration of all cancelled $5f$ resonances by means of counterpropagating beams as proof for cancellation has also been checked. Spectra depicting cancellation and restoration of $5f$, $J=2$ peaks can be found in the earlier publication [6]. In that publication the pressure dependence of the cancellation of the $4f[3/2]_2$ state is also illustrated. Cancellation of the $4f$ resonances starts at about 2 mbar. The observed effect has already been discussed [6] in terms of the destructive interference of the three-photon excitation channel and the one THG photon absorption, thus cancelling the paths to the continuum. A quantitative verification of this can be found in the theoretical paper of Elk, Lambropoulos, and Tang [7].

The relation of the cancellation of ionization to the THG spectral distribution can be seen in Figs. 5 and 6. Figure 5 shows the evolution of the ionization and THG spectrum as pressure increases from $P<0.3$ to 5 mbar when four-photon absorption is in the vicinity of the $4f$ manifold. At very low pressures no THG has been observed and the $6s[3/2]_1$ state ($3+2$ ionization) dominates the multiphoton ionization spectrum. At ~ 0.3 mbar in Fig. 5(b) cancellation of the $6s[3/2]_1$ state begins as THG starts occurring to the blue of the three-photon resonance. The $4f[3/2]_2$ ionization peak dominates among the four $4f$ resonances. Further increase of the pressure in Fig. 5(c) shifts the THG profile to shorter wavelengths so that now the $4f$, $J=2$ states can be excited via one THG and one laser photon absorption. The reabsorption of the THG can be clearly seen in the dip of the distribution when the wavelength matches the excitation of the $4f[3/2]_2$ state, the ionization peak of which shows strong enhancement and thus still dominates the MPI spectrum. Finally, by pressure-shifting the THG profile maximum to shorter wavelengths [Fig. 5(d)] than those matching excitation of the $4f$ states, the $4f[3/2]_2$ state shows cancellation. The same qualitative behavior is observed in the $5f$ manifold. Thus, the two $5f$, $J=2$ resonances undergo cancellation at such high pressures that phase matching at their excitation wavelengths is fairly poor due to the blue shift of the THG distribution; in contrast, when the maximum of the vuv distribution matches their excitation, the two $J=2$ states show enhancement and two absorption dips can be observed in the THG profile [Figs. 6(a) and (b)].

Both the dependence of pressure required for cancella-

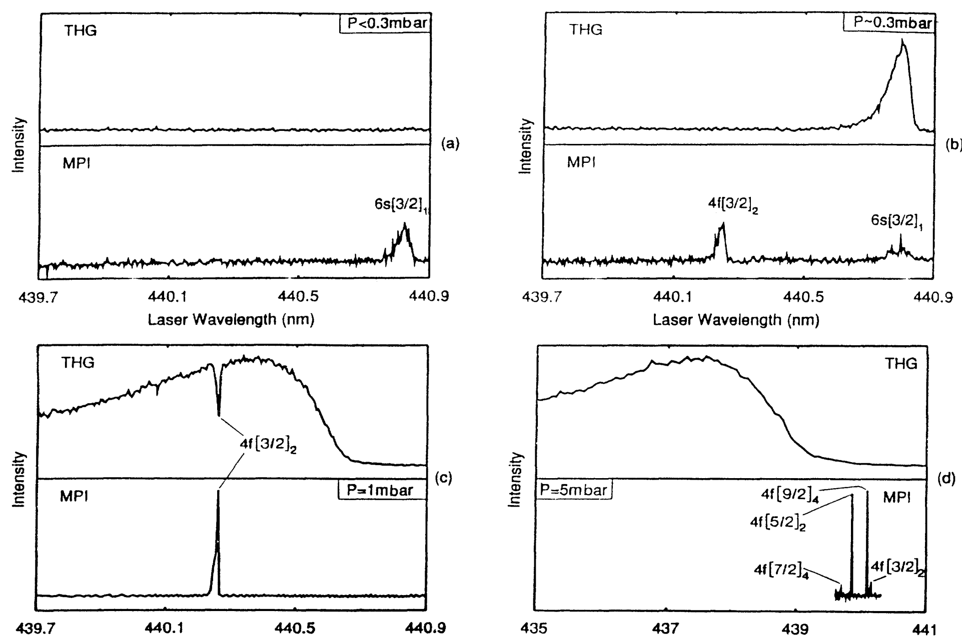


FIG. 5. Simultaneously recorded MPI and THG spectra in the wavelength region leading to four- or two- (one THG and one laser) photon excitation of the $4f$ states at $P<$ (a) 0.3, (b) 0.3, (c) 1.0, and (d) 5.0 mbar. Reabsorption of the THG resulting in ionization enhancement can be seen when the THG wavelength matches excitation of the $4f[3/2]_2$ state. When the THG profile is shifted to the blue (d) so that excitation of the $4f[3/2]_2$ resonance corresponds to wavelengths for which there is bad phase matching, cancellation occurs.

tion to occur on the three-photon detuning from the $6s[3/2]_1$ state and the fact that cancellation occurs at smaller detunings Δ_{can} than that of maximum harmonic generation $\Delta_{\max\ \text{THG}}$ are in good agreement with the pressure condition theoretically predicted by Elk, Lambropoulos, and Tang, as well as with similar conditions resulting from other models which include detuning from the three-photon resonances. This well-established condition requires the interaction volume to be much longer than the linear absorption length at ω_{th} (optical thickness condition) and reads [7]

$$|\text{Im}(\Delta k)|b \gg 1 \ , \quad \text{with } \Delta k \sim \frac{|\mu_{12}|^2}{\Delta + i\Gamma/2} N$$

and

$$b \gg \frac{n_3 c}{2\pi\omega_{\text{th}}} \frac{\Delta^2+\Gamma^2/4}{|\mu_{12}|^2 N\Gamma/2} \ ,$$

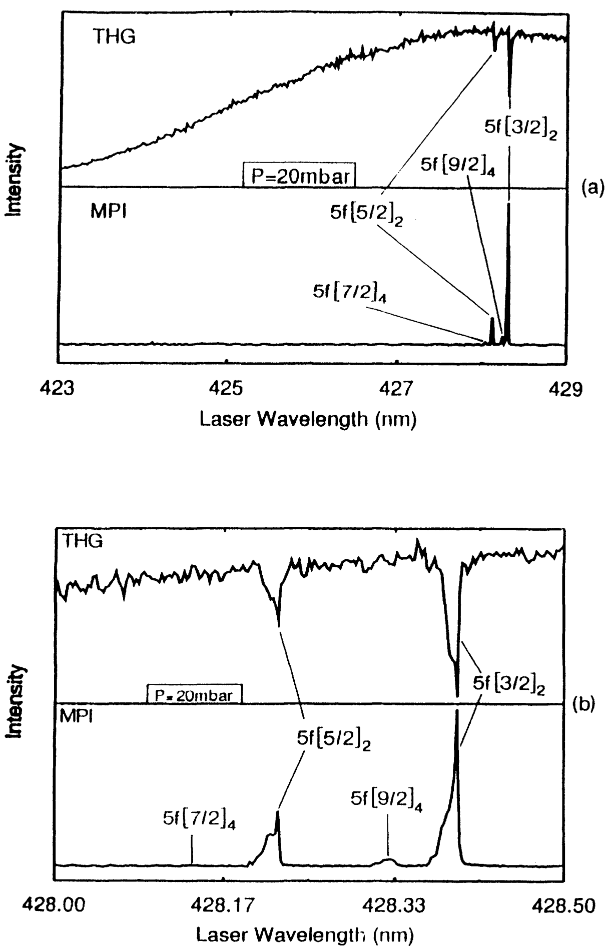


FIG. 6. Simultaneously recorded MPI and THG spectra in the wavelength region leading to four- or two- (one THG and one laser) photon excitation of the $5f$ states at (a) 20 mbar. (b) High-resolution spectra showing the reabsorption dips in the THG profile when the laser wavelength matches excitation of the $5f[3/2]_2$ and $5f[5/2]_2$ states.

b being the confocal parameter; Δk the phase mismatch at ω_{th}; Δ the detuning from the $6s[3/2]_1$ state; N the number density of the atoms, i.e., the pressure; Γ the sum of all decay widths of the coherence of the system (radiative, collisional, laser broadening, etc.) dominated by the laser bandwidth ($\simeq 0.2$ cm^{-1}); μ_{12} the one-photon transition matrix element between the ground and the $6s[3/2]_1$ state; and n_3 the refractive index at ω_{th}. By making the assumption, justified by the experimental parameters, $\Gamma \ll \Delta$, one can clearly see the need for higher pressures for cancellation at larger detunings. In other words, the observed cancellation at different detunings occurs at such pressures at which the medium becomes optically thick at ω_{th}, corresponding to the detuning Δ. Although the ratio of the pressures at which cancellation begins for the $4f[3/2]_2$ and the $5f[3/2]_2$ resonances is not exactly the one given by the formula above, taking into account the two corresponding detunings of ~ 100 and 2000 cm^{-1}, the quantitative agreement with this formula can be considered satisfactory, due to the large instrumental uncertainties in the pressure measurements (lack of calibrated manometer) especially in the 1-mbar regime.

Since maximum harmonic generation of tightly focused beams occurs for $\text{Re}(b\Delta k) = -2$, under the assumption $\Delta \gg \Gamma$, it can be easily seen from the relations shown above that $\Delta_{\text{can}} < \Delta_{\max\ \text{THG}}$. It must be pointed out that the $nf[5/2]_2$ resonances behave differently than the $nf[3/2]_2$ ones. Thus, while the $4f[3/2]_2$ state undergoes cancellation and causes reabsorption in the THG distribution, the $4f[5/2]_2$ state does not, as can be seen in Figs. 2 and 5(c). The absence of cancellation for the $4f[5/2]_2$ resonance has been attributed by Elk, Lambropoulos, and Tang [7] to be due to the violation of the single-state condition, that is, due to the dominance of the nonresonant excitation of this state. We will come back later to this point in the two-color-experiment section in which some experimental results concerning the

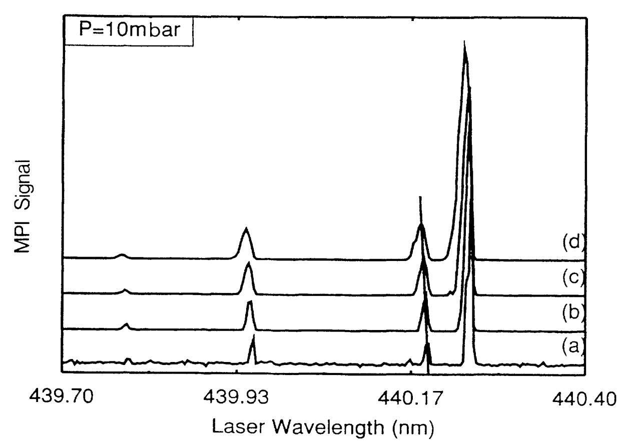


FIG. 7. REMPI spectrum via the $4f$ manifold at different power densities $I \simeq$(a)1.5 GW/cm^2, (b) 3.0 GW/cm^2, (c) 4.5 GW/cm^2, and (d) 6.0 GW/cm^2 depicting the ac Stark shift of the resonances.

$nf[5/2]_2$ states will be presented, which might bring more insight into this problem.

No intensity dependence of the cancellation effect has been observed, at least for the power-density range 1–10 GW/cm^2 employed in the experiment and for the few pressures investigated. ac Stark shifting of the nf resonances though has been observed. A shift of $\sim 0.45I$ (cm/GW), where I is the power density in GW/cm^2, is shown for the $4f$ manifold in Fig. 7.

Furthermore, no change of the cancellation condition could be observed by increasing the bandwidth of the laser by a factor of 3. This might be due to other dominating dephasing mechanisms.

Two-color experiments

For a more detailed study of the observed cancellation effect, two-color experiments have been performed. Three different aspects have been investigated. For the verification of the condition concerning the position of the THG profile relative to the three-photon excitation for which cancellation or ionization enhancement occurs, the following procedure has been undertaken: At a constant pressure, i.e., for a given THG profile, the wavelength of the first laser was tuned so that three-photon absorption is (a) to the red and (b) on the observable THG profile, while the second laser was scanned in the region for which absorption of a fourth photon from the second laser led to excitation of the $6f$ manifold. That is, instead of shifting the THG profile around the three-photon excitation, the three-photon excitation was shifted with respect to a constant THG profile, thus allowing a consideration of how atomic parameters may influence the process. Second, the possibility of cancellation for positive three-photon detunings relative to the $6s[3/2]_1$ state because of the Δ^2 dependence of the cancellation condition has been investigated by tuning the first laser so that three-photon excitation is below the $6s$ resonance. Lastly, in relation to the observed cancellation of the $5f[5/2]_2$ state and the absence of the effect for the $4f[5/2]_2$ state in the one-color experiments, the behavior of the $6f[5/2]_2$ resonance for different negative detunings of the first laser from the $6s$ state has been studied.

In the first case investigated, the Xe pressure was set at 5 mbar, for which the THG profile can be seen in Fig. 5(d). For three fixed wavelengths $\lambda_1 = 440.9$, 440.6, and 435.1 nm, i.e., on resonance, ~ 43 and ~ 900 cm^{-1} above the $6s[3/2]_1$ state, λ_2 was scanned so that $3\omega_1 + \omega_2$ matches excitation of the $6f$ manifold. In the first two cases there was bad phase matching at $3\omega_1$, that is, $3\omega_1$ was to the red of the observed THG profile, while in the last case, $3\omega_1$ has produced significant THG [see Fig. 5(d)]. The corresponding spectra are shown in Figs. 8(a), 8(b), and 8(c). As expected, only in the first two cases [Figs. 8(a) and 8(b)] has partial cancellation been observed. Since counterpropagation of the laser-1 beam for restoration of the resonances was not possible in the experimental setup employed, the intensities of the $6f$, $J=2$ resonances relative to those of the $J=4$ resonances were used as criteria for the presence or absence of cancellation. The relative peak intensities are not expected

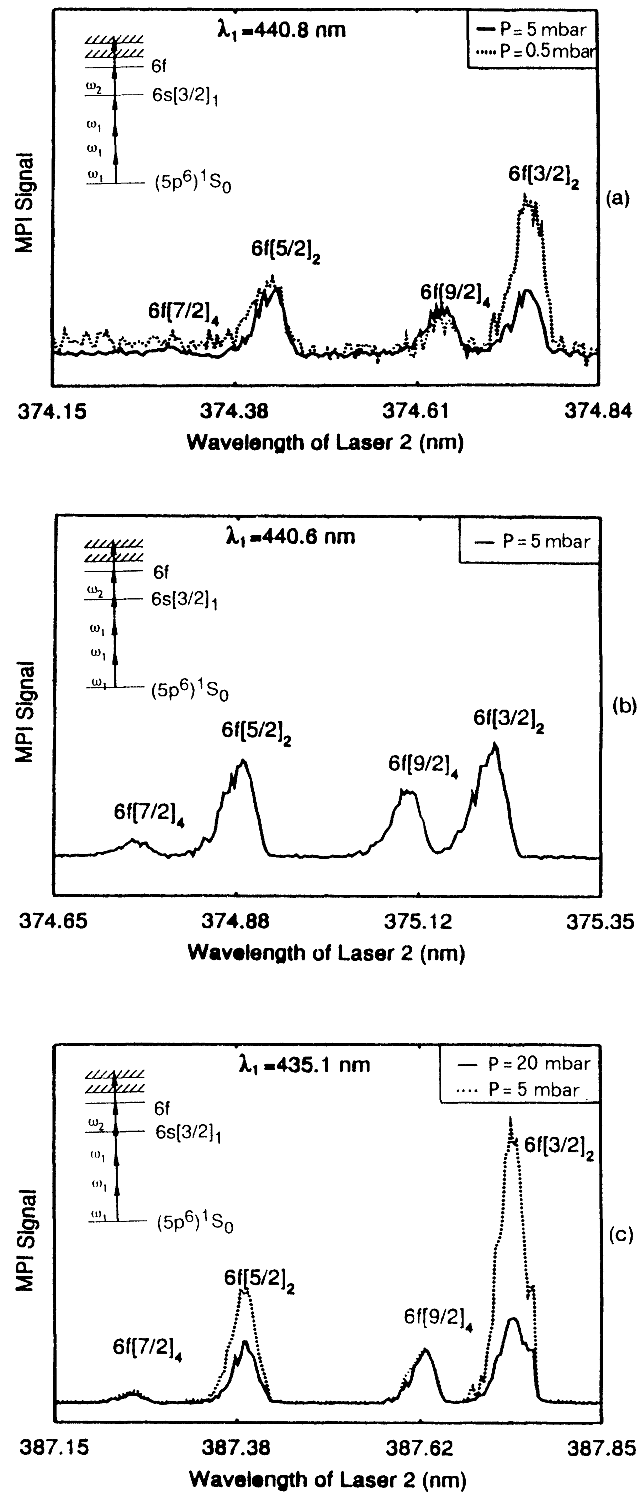


FIG. 8. Two-color REMPI spectra via the $6f$ manifold ($3\omega_1 + \omega_2$ excitation). $3\omega_1$ is (a) on resonance with, (b) ~ 43 cm^{-1} above, and (c) ~ 900 cm^{-1} above the $6s[3/2]_1$ state, that is, $3\omega_1$ is to the red of the THG profile [(a) and (b)] and on the THG profile (c). Spectra are normalized so that the $6f[9/2]_4$ peaks have the same height in all spectra for relative intensity comparison reasons.

in general to be the same for different pairs of ω_1 and ω_2, due to the different excitation probability of the nf manifold. Thus measurement of the relative peak intensities at different pressures was necessary in order to confirm cancellation, unless there was a striking decrease of $J=2$ peak intensities.

Typical relative intensities of the four peaks of the $6f$ manifold under conditions for which cancellation does not take place are given in the spectra of Fig. 9. Figure 9(a) depicts the one-color spectrum recorded with two counterpropagating beams at 5 mbar. In this spectrum there is no cancellation for two reasons: (i) counterpropagating beams were used and (ii) three-photon excitation was far to the blue of the THG profile. Figure 9(b) shows a two-color spectrum taken with $3\omega_1$ being on resonance with the $6s[3/2]_1$ state at low pressure, where no THG occurs. In this spectrum the $J=2$ states are even more dominant than in Fig. 9(a). These are the two extreme cases of detuning of $3\omega_1$ from the $6s$ state, for which different relative ionization intensities of the four peaks could result. Between them lie all investigated cases at

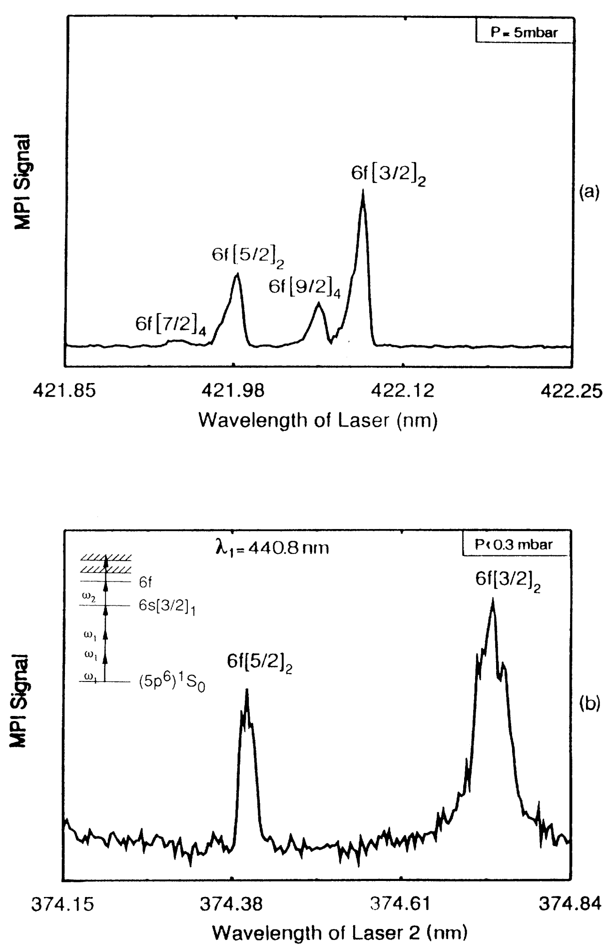


FIG. 9. REMPI spectra via the $6f$ manifold: (a) One-color two counter-propagating beam spectra at 5 mbar; (b) Two-color spectra at pressure lower than 0.3 mbar $3\omega_1$ is on resonance with the $6s[3/2]_1$ state.

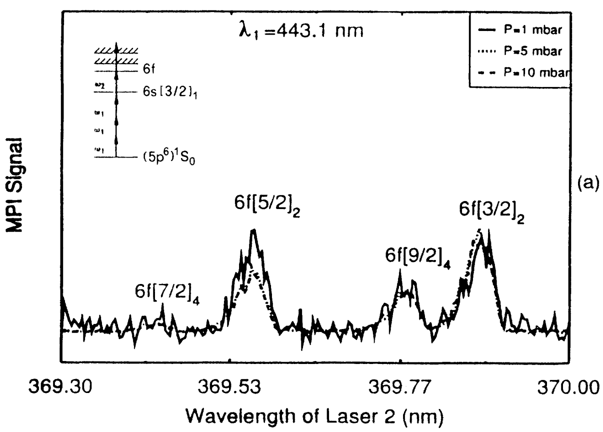
negative detuning in the two-color experiments. Moreover, no other dipole-allowed resonance transition is near the three-photon excitation for the wavelength range defined by the two limiting cases, which may cause large changes in the relative peak intensities. Comparison with the spectra in Fig. 8 proves again that partial cancellation of the $6f[3/2]_2$ resonance occurred when the $3\omega_1$ excitation was to the red of the THG profile, and hence phase matching was rather bad [Figs. 8(a) and 8(b)], while when $3\omega_1$ was in the region of optimum THG, the intensity of the $6f[3/2]_2$ resonance increased back [Fig. 8(c)] as compared with the intensities of the $J=4$ peaks in the spectrum. The reduction of the $J=2$ peak intensities in Fig. 8(c) when the pressure was increased to 20 mbar and the shift of the THG profile to wavelengths shorter than $\lambda_1/3$ provide further verification for the presence of cancellation. In the two-color experiments cancellation was incomplete probably due to the additional four-photon-excitation channels of the $6f$ resonances, in addition to the two destructively interfering ones.

These results are closely related to earlier observations of Compton and Miller [11]. In their experiment the second laser beam coupled the $6s[3/2]_1$ state with np and np' states, while the first one was scanned so that three-photon absorption was in the vicinity of the $6s[3/2]_1$ state. Experimental differences notwithstanding, the disappearance of the ionization peak corresponding to the coupling of the $6s[3/2]_1$ with the $7p[3/2]_2$ state at higher pressures might be interpreted as having the same origin as the results of the present work—namely, cancellation occurs when three-photon excitation is on the long-wavelength side of the blue-shifted THG profile.

An important aspect resulting from the two-color experiment is the observed behavior of the $6f[5/2]_2$ resonance. In Fig. 8(c) a comparison of the two spectra taken at the two different pressures shows that when the pressure condition was fulfilled, the $6f[5/2]_2$ resonance underwent partial cancellation, similar to the $6f[3/2]_2$ resonance. However, when $3\omega_1$ was on resonance with the $6s[3/2]_1$ state, the $6f[3/2]_2$ resonance showed the expected cancellation, while the $6f[5/2]_2$ state did not, as can be seen in Fig. 8(a) in which the intensity of the $6f[5/2]_2$ peak remained the same when the pressure increased to 5 mbar. Furthermore, the ratio of the ionization intensities of the $6f[5/2]_2$ and $6f[9/2]_4$ resonances in Fig. 8(b) is about the same as that of Fig. 8(a) at 5 mbar, but differs from that in Fig. 8(c) at 5 mbar. These observations indicate that cancellation of the $6f[5/2]_2$ resonance, besides the pressure condition, requires large detunings of $3\omega_1$ from the $6s[3/2]_1$ state, at least for the pressure region that has been investigated. This result is consistent with the observation for the $4f[5/2]_2$ and $5f[5/2]_2$ resonances in the one-color experiment, that is, for the $4f[5/2]_2$ state, 3ω is only 100 cm^{-1} above the $6s[3/2]_1$ state, while for the $5f[5/2]_2$ state, it is about 2000 cm^{-1}. The combined results of the one- and two-color experiments suggest that the $nf[5/2]_2$ resonances do not exhibit cancellation if the three-photon excitation is close to the three-photon resonance but only at large detunings, provided that the pressure condition is fulfilled. More generally, if one also takes into account the absence of any absorption feature in the THG profile when 4ω matches excitation of the $4f[5/2]_2$ state (Fig. 5) and contrasts it to the appearance of the dip corresponding to the $5f[5/2]_2$ resonances in Fig. 6, one may conclude that the channel including one THG photon is less relevant for the excitation of the $nf[5/2]_2$ state when the detuning from the three-photon resonance is not sufficiently large.

The condition that nonresonant ionization has to be negligible as predicted by Elk, Lambropoulos, and Tang [7] cannot straightforwardly explain the cancellation behavior of the $nf[5/2]_2$ resonances, since it is not obvious why resonant contributions to the four-photon transition probability to the $nf[5/2]_2$ states increase in comparison to the nonresonant contributions when the detuning from the $6s[3/2]_1$ state becomes larger.

It is likely that the strength of the coupling of the $6s[3/2]_1$ with the $nf[5/2]_2$ states controls both cancellation and absorption of THG, leading to excitation of the



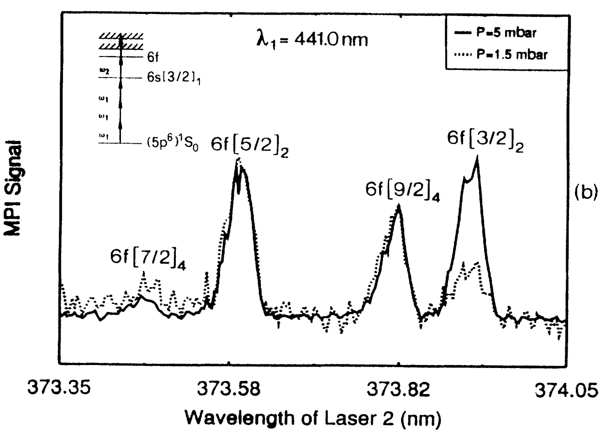


FIG. 10. Two-color REMPI spectra via the $6f$ manifold at 5 mbar for positive $3\omega_1$ detunings of (a) 340 cm^{-1} and (b) 40 cm^{-1} from the $6s[3/2]_1$ state. Spectra are normalized so that the $6f[9/2]_4$ peaks have the same height in all spectra for relative intensity comparison reasons.

latter states. However, since this effect has not been observed for the $nf[3/2]_2$ states, it can be considered as being state specific, depending on the detailed atomic parameters. Thus only a quantitative calculation including accurate atomic parameters could bring more insight into the problem.

Investigation of ionization spectra for positive $3\omega_1$ detuning from the $6s[3/2]_1$ state showed in general no evidence for the presence of cancellation. Although in the spectra shown in Fig. 10(a) the intensity of the $6f[3/2]_2$ peak is relatively low in comparison to all other investigated cases as yet, no significantly large change in the relative intensities of the $J=2$ to $J=4$ states has been observed in the spectra such as to justify consideration of cancellation as the pressure was increased from 1 to 10 mbar. Following the theoretically predicted Δ^2 dependence of the pressure condition for cancellation and for the present $\sim$340-cm^{-1} detuning from the $6s[3/2]_1$ state ($\lambda_1=443.1$ nm), it was expected that the transition from a "no cancellation" to a "cancellation" condition would occur within this pressure range. The only evidence for cancellation when $3\omega_1$ is tuned below resonance is when the detuning from the $6s[3/2]_1$ state was very small. That can be seen in the spectra of Fig. 10(b) taken at 1.5 mbar and for $\Delta \simeq 40$ cm^{-1} where the $6f[3/2]_2$ peak intensity has strongly decreased. However, the observed cancelled peaks reappeared as the pressure was increased to 5 mbar, which is in complete disagreement with the cancellation behavior for negative $3\omega_1$ detuning and with the theoretical predictions. The change of the dispersive and absorptive properties of the medium in the presence of a second laser beam that couples the three-photon with the four-photon resonance as proposed by Tewari and Agarwal [12] could account for the observed cancellation, this no longer being below resonance due to the splitting of the $6s$ state. Pressure shifting to shorter wavelengths may compensate for the effect, thus eliminating cancellation at increased pressures.

CONCLUSIONS

In summary, destructive interference between three-photon excitation and one THG excitation channels has been demonstrated for large three-photon detunings from the $6s[3/2]_1$ resonant state of Xe, probed through the cancellation of four-photon resonantly enhanced five-photon ionization via $nf[3/2]$ ($n=4$, 5, and 6), $J=2$ states. Cancellation turns out to occur not at pressures leading to maximum phase matching but at higher ones at which phase matching is poor and the THG profile is shifted towards shorter wavelengths. At these pressures the absorption length of the gas at ω_{th} becomes much smaller than the interaction length, which is an important condition determining the appearance of the destructive interference. At pressures leading to good phase-matching conditions, absorption dips in the THG profile have been observed at wavelengths corresponding to two-photon excitation (one THG and one photon of the driving electromagnetic wave) of the nf, $J=2$ states. Ionization through the $J=2$ states exhibits enhancement in this case.

Two-color experiments have verified the conclusion of the one-color experiments, as cancellation can be observed now at constant pressure only when the first laser is tuned so that $3\omega_1$ is to the red of the THG profile. An important result of the two-color experiments is the demonstration of cancellation of $nf[5/2]_2$ resonances only when the three-photon excitation is far detuned from the three-photon $6s[3/2]_1$ resonance. This is in very good agreement with the absence of cancellation of the $4f[5/2]_2$ in contrast to the $5f[5/2]_2$ resonance in the spectra of the one-color experiments, but cannot be explained in terms of the single-state condition of Elk, Lambropoulos, and Tang [7].

Furthermore, cancellation for detunings below the three-photon resonance resulting from the theoretically predicted Δ^2 dependence of the pressure condition disagrees with our experimental observation at positive three-photon detuning from the $6s[3/2]_1$ state. Cancellation can be observed only for small positive detuning, the magnitude and the existence of which is questionable due to possible shift and splitting of the $6s$ state caused by strong coupling with the $6f$ states via the second laser beam. The experimental observations of this work leave several questions open concerning the cancellation of four-photon resonances resulting from the destructive interference of the three-photon and one THG photon excitation channels. Further quantitative theoretical work is needed for a more complete understanding of the general scheme of cancellation.

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*Permanent address: Institute of Physics, Chinese Academy of Sciences, Beijing, China.
†Also at Department of Physics, University of Crete.

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