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Cancellation effects in four-photon-resonant five-photon ionization through the nf $J=2$ states of Xe

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Partial cancellation of the ionization signal in the four-photon-resonant five-photon ionization via $J=2$ $4f$ and $5f$ resonances in Xe has been observed at increased gas pressures. Experiments with one and two counterpropagating laser beams provide strong evidence that cancellation is due to destructive interference at the three-photon step between the coherent three- and one-photon excitation channels, the latter involving one vacuum ultraviolet photon of the third-harmonic generation that occurs above the $6s[3/2]_1^o$ state. The well-known destructive interference for the "on-resonance" case is now shown to occur also for large detuning from the resonance (up to 2000 cm^{-1}) probed by four-photon-resonant five-photon ionization. This type of cancellation may provide new insight in interference effects involved in multiphoton-excitation processes.

Third-harmonic generation (THG) in gases, its interplay with multiphoton ionization (MPI), and its influence on the observed MPI spectra has been studied extensively over past years [1]. Once the third harmonic is generated, it can be reabsorbed from the gas atoms together with additional photons of the fundamental frequency. This is an additional excitation channel to the one which involves the absorption of photons of the fundamental frequency. Interference effects between these two channels have been observed experimentally and studied theoretically for three-photon [2–4] and two-photon [5] resonance conditions. A well-known example of destructive interference between these channels is the complete cancellation of the resonance in the MPI spectra observed in the three-photon-resonant five-photon ionization—(3+2) resonantly enhanced multiphoton ionization (REMPI)—via the $6s[3/2]_1$ state of Xe [2,3]. In sharp contrast, a considerable enhancement of the MPI signal has been observed in Kr, when the THG photons are nonresonantly absorbed together with an additional photon of the fundamental frequency exciting the atoms in the autoionizing region between the two fine-structure ionization limits [6]. In the present work we report the partial cancellation of resonances in the 4+1 REMPI of Xe via the $J=2$ $4f$ and $5f$ states. Interesting questions about the origin of the present observations have already motivated a quantitative theoretical investigation [7]. The observed type of cancellation is consistent with the picture of a destructive interference between the THG produced in the vicinity of the $6s[3/2]_1$ state and the three-photon excitation path of the fundamental frequency, which interrupts excitation of the nf manifolds. It is pointed out that in contrast to previous experimental investigations, destructive interference is occurring not on resonance but in regions far detuned from it.

For the purposes of this work, a static cell filled with Xe at pressures ranging from 1 to 10^3 mbar was used. Ions were collected by an electrode assembly biased at 200 V. The ion signals were amplified, digitized, and stored in a microcomputer. Excitation was achieved by means of an excimer-pumped dye laser, emitting pulses of 12 ns duration. The laser output was focused between the electrodes and the resulting power densities were estimated to be of the order of 1 GW/cm^2 The laser beam was linearly polarized by using a Glan air prism. In some of the experiments, the beam was split by a quartz plate into two parts having an intensity ratio of $\sim\frac{1}{10}$. The weak beam was counterpropagated with respect to the strong beam and focused in the same region. For the experimental runs in which simultaneous observation of the MPI and the THG signal occurred, a vacuum chamber was used separated with a LiF window from the static cell and equipped with a LiF prism for the separation of the vacuum ultraviolet (VUV) light from the fundamental. The VUV photons were detected with a solar-blind photomultiplier tube. A more detailed description of the apparatus will be given elsewhere [8].

Four-photon excitation of Xe at the wavelength regions of 427.99–428.69 and 439.71–440.41 nm leads to the 4+1 REMPI via the $5f$ and $4f$ manifolds, respectively. Figures 1 and 2 show the REMPI spectra of Xe resulting from the $4f$ and $5f$ states resonances at different gas pressures. The solid curves correspond to one laser-beam ionization, while the dashed curves to two counterpropagating beams with parallel polarization. The four peaks of the spectra from left to the right are due to the $nf[7/2]_4^e$, $nf[5/2]_2^e$, $nf[9/2]_4^e$, and $nf[3/2]_2^e$ ($n=4,5$) intermediate resonances. In the spectra taken with only one laser beam the intensity of the $4f[3/2]_2^e$, $5f[3/2]_2^e$, and $5f[5/2]_2^e$ resonances show a strong dependence on the gas pressure. Since the collection efficiency of the detector may depend on the gas pressure, the $4f[9/2]_4^e$ and $5f[9/2]_4^e$ resonances have been used as reference peaks for the peak-intensity comparisons. For angular momentum conservation reasons, these peaks cannot be affected by the process discussed below which causes the pressure dependence of the peak intensity of the nf $J=2$ resonances. As shown in Figs. 1 and 2, the relative peak intensities of the three $J=2$ states decrease dramatically with increasing gas pressure. For the $4f[3/2]_2^e$ state this happens for pres-

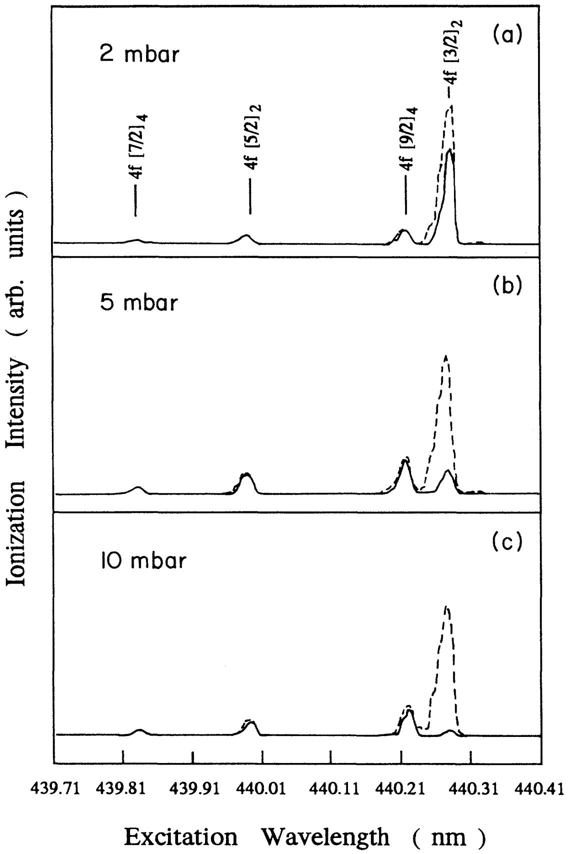

FIG. 1. Excitation spectrum of Xe in the wavelength region 439.71–440.41 nm at (a) 2-, (b) 5-, and (c) 10-mbar gas pressure by using one laser beam (——) and two counterpropagating beams with parallel polarizations (---). The peaks from left to right correspond to the 4+1 REMPI via the $4f[7/2]_4^o$, $4f[5/2]_2^o$, $4f[9/2]_4^o$, and $4f[3/2]_2^o$ intermediate resonances. The $4f[3/2]_2^o$ resonance is partly canceled at higher pressures in the one-beam experiment.

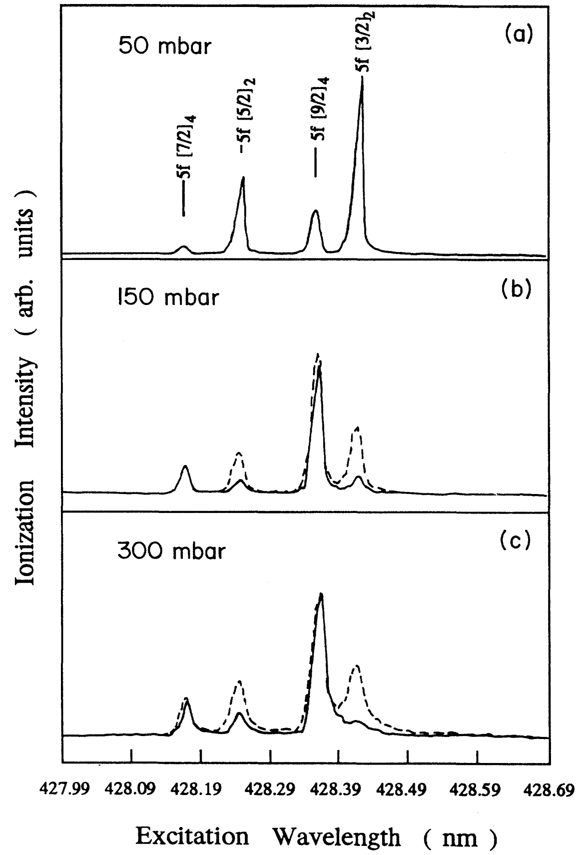

FIG. 2. Excitation spectrum of Xe in the wavelength region 427.99–428.69 nm at (a) 50-, (b) 150-, and (c) 300-mbar gas pressure by using one laser beam (——) and two counterpropagating beams with parallel polarizations (---). The peaks from left to right correspond to the 4+1 REMPI via the $5f[7/2]_4^o$, $5f[5/2]_2^o$, $5f[9/2]_4^o$, and $5f[3/2]_2^o$ intermediate resonances. The $J=2$ resonances are partially canceled at higher pressures in the one-beam experiment.

sures higher than 2 mbar; while for the $5f[3/2]_2^o$ and $5f[5/2]_2^o$ states, at pressures higher than 100 mbar. Furthermore, when the second counterpropagating beam is introduced, these peaks recover their intensity. The intensity of the peaks corresponding to the $J=4$ states is independent of the presence of the second beam, thus verifying that the effect under investigation does not affect the $J=4$ states.

The observed strong decrease of the intensity of the peaks may be explained, as discussed below, in terms of the presence of the THG which occurs above the $6s[3/2]_1^o$ state. As has been established by Miller *et al.*, this takes place at higher pressures and can be reabsorbed from the Xe atoms [9]. The THG profiles broaden and shift to the blue as the pressure increases, so that the THG photon energy together with the photon energy of the fundamental frequency eventually match up the excitation energy of the $4f$ and $5f$ $J=2$ states. This provides additional excitation channels for these states besides the four-photon excitation path. It is obvious that the excitation of the $4f$ states via this new channel needs a less-broadened THG profile, and therefore takes place at lower pressures than that of the $5f$ states. The importance of the presence of the two-photon excitation channel of the $4f$ states has also been noted by Jackson, Wynne, and Kes [10].

The partial cancellation is attributed to a destructive interference between excitation channels of the following two types: (i) the three-photon near-resonance absorption of the fundamental laser frequency, and (ii) the one-photon near-resonance absorption of the THG. Excitation in both channels is detuned far from the $6s[3/2]_1$ state in the vicinity of which THG takes place. It should be pointed out that the detunings from the $6s$ state for 4+1 REMPI via the $4f$ states are of the order of 100 cm^{-1} and via the $5f$ states about 2000 cm^{-1}. Destructive interference between two channels of this type has been experimentally observed for the case of resonant three-photon excitation [2,3]. In this case, the cancellation of the three-photon resonance is complete. The present experiment demonstrates that cancellation occurs also for a wide region above the three-photon resonance. This region is pressure dependent. The fact that only partial can-

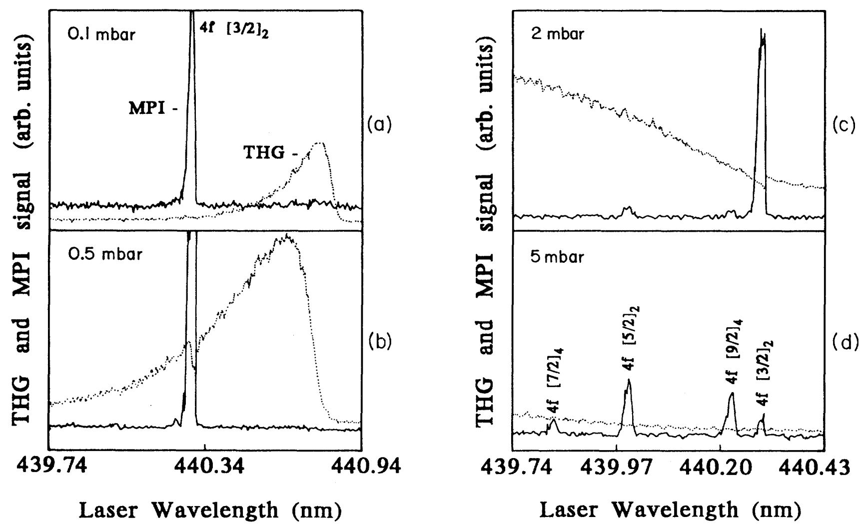

FIG. 3. Simultaneously observed MPI (——) and THG (····) spectra in the wavelength region where four-photon excitation of the $4f$ manifold occurs at (a) 0.1-, (b) 0.5-, (c) 2-, and (d) 5-mbar Xe pressure. Cancellation of the $4f[3/2]_2$ state occurs when the maximum of the THG profile is shifted to wavelengths shorter than those causing excitation of the $4f$ manifold (d).

cellation occurs can be attributed to nonresonant contributions which become more and more relevant as the detuning from the exact resonance is increased. It is well known that three-photon resonances which cancel completely through such interference effects can be restored by introducing a counterpropagating beam due to the new excitation channels which are opened by the second beam [3,11]. For the same reason, the peak intensity in the MPI spectra was restored when the second beam was used in this work. From the simultaneous observation of the MPI spectra and the THG profile (Fig. 3), we see that cancellation occurs not when the maximum of the VUV intensity distribution matches the excitation of the f states, but when this maximum is shifted to shorter wavelengths by increasing the pressure. In contrast, when the maximum of the THG intensity matches the excitation of both $4f$ and $5f$ manifolds, dips in the THG profile have been observed at wavelengths corresponding to the excitation of the nf $J=2$ states, in agreement with the observations of Miller *et al.* [9]. In this case, the nf $J=2$ resonances in the MPI spectra exhibit an enhancement. A more-detailed discussion of the simultaneously observed THG and MPI spectra will be presented elsewhere [8]. In brief, there is a pressure-dependent range of detuning above the three-photon resonance for which cancellation keeps occurring. This cancellation range is not extended to the region where phase-matching maximizes THG. Cancellation occurs, rather, in ranges of poor phase matching. However, there is an upper limit of maximum detuning for which cancellation occurs, irrespective of any further increase of the pressure as stated by the absence of any cancellation effects for the $6f$ and $7f$ $J=2$ states even at high pressures (10^3 mbar).

The absence of the strong-intensity decrease of the $4f$ resonances in the 4+1 REMPI in previous studies [10,11] is due to the different experimental conditions employed in these studies. However, the results obtained in a recent work by Blazewicz and Miller, who have studied optical processes in Xe and Kr in a two-color MPI experiment [1], seem to be relevant to the present observations. In their spectra, the ionization peaks corresponding to two-color 4+1 REMPI via the $5f$ $J=1$ states of Xe appear under certain circumstances as ionization dips in the nonresonant ionization background. These observations may be interpreted as having the same origin with the results of the present work.

Theoretical studies related to the "cancellation effect" in the three-photon resonance have been carried out by several authors [3,4,7,10,12–14]. It is noteworthy that different theoretical models including detuning from the three-photon resonance [7,12,13] lead to very similar conditions for cancellation to occur in regard to the relation between detuning and gas pressure. The present experimental results are in good agreement with their prediction that increased detuning can be compensated by increased gas pressure in order for the cancellation condition to be valid as long as collisional relaxations do not dominate. A verification of the quadratic pressure dependence on the detuning of the cancellation condition, as predicted theoretically, can only be done if the atomic parameters involved in the REMPI scheme are known. Using the condition $\Delta kb=-2$ for maximum THG in tightly focused beams [15], where Δk is the phase mismatch and b the confocal parameter, and the results of the above-mentioned theoretical treatments, it can be seen that the detuning from the resonance for which THG maximizes is larger than the maximum detuning for which cancellation occurs as has been observed in the present investigation. It is, however, quite surprising that there is no evidence for any cancellation effect for the $4f[5/2]_2^e$ resonance in the spectra obtained even at high gas pressures (up to 10^3 mbar). There is no obvious reason for which the excitation of this state should show a different behavior from that observed for the other $J=2$ states of the $4f$ and $5f$

manifolds. A quantitative calculation including all atomic parameters provides more insight into this behavior, such as the calculations carried out by Elk, Lambropoulos, and Tang [7].

In conclusion, the present investigation has demonstrated new aspects concerning the interplay between THG and MPI in gases. Cancellation of the excitation has been observed for a wide range of detuning from a three-photon resonance probed by 4+1 REMPI in Xe. The influence of parameters such as the magnitude of detuning, the pressure, and the presence of relaxation processes on the experimental results has been qualitatively discussed in terms of relevant theoretical work. Detailed theoretical calculations quantifying this interpretation are expected to elucidate these aspects further. The probing method used in this work may also be extended into two-color experiments providing a sensitive technique for the investigation of other parameters important for this type of interference, such as the laser bandwidth, the roles of other near resonances, and of different relaxation channels. These types of experiments are currently in progress.

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