[6] G. E. Brown, J. S. Dehesa, and J. Speth, Nucl. Phys. A330, 290 (1979).

[7] W. Knüpfer, R. Frey, A. Friebel, W. Mettner, D. Meuer, A. Richter, E. Spamer, and O. Titze, Phys. Lett. 77B, 367 (1978).

[8] C. P. Swann, Phys. Rev. Lett. 32, 1449 (1974).

[9] R. Del Vecchio, S. Freedman, G. T. Garvey, and M. Oothoudt, Phys. Rev. Lett. 34, 1296 (1975), and Phys. Rev. C 13, 2089 (1976).

[10] C. P. Swann, Phys. Rev. C 16, 2426 (1977).

[11] T. Chapuran, R. Vodhanel, and M. K. Brussel, Phys. Rev. C 22, 1420 (1980).

[12] W. Biesiot and Ph.B. Smith, Phys. Rev. C 24, 808 (1981).

[13] K. Ackermann, K. Bangert, U. E. P. Berg, G. Junghans, R. K. M. Schneider, R. Stock, and K. Wienhard, Nucl. Phys. A372, 1 (1981).

[14] R. M. Laszewski and P. Axel, Phys. Rev. C 19, 342 (1979).

[15] K. Wienhard, R. K. M. Schneider, K. Ackerman, K. Bangert, U. E. P. Berg, and R. Stock, Phys. Rev. C 24, 1363 (1981).

[16] A. M. Nathan, R. Starr, R. M. Laszewski, and P. Axel, Phys. Rev. Lett. 42, 221 (1979).

Experimental Evidence for the Competition between Resonantly Enhanced Multiphoton Ionization and Third-Harmonic Generation in Xenon

James H. Glownia and Robert K. Sander
University of California, Los Alamos National Laboratory, Los Alamos, New Mexico 87545

(Received 21 May 1982)

Competition between third-harmonic generation and multiphoton ionization in xenon is confirmed in a direct experiment using counterpropagating circularly polarized laser beams to suppress third-harmonic emission. Three-photon resonantly enhanced multiphoton ionization through the 6*s* and 6*s*′ levels in xenon, previously reported to disappear at pressures above a few Torr, gives rise to sharp intense ionization signals under these conditions.

PACS numbers: 32.80.Kf, 32.20.Jc, 32.80.Fb, 42.65.Cq

With the advent of high-power lasers, gas-phase multiphoton excitation has been extensively studied in recent years. In particular, xenon atoms have been investigated by three-photon excited fluorescence and resonantly enhanced multiphoton ionization (MPI). In these studies, the signal due to the three-photon allowed $^1S_0$-$5p^5(^2P_{3/2}{}^o)6s$ transition was observed to be strongly pressure dependent. The disappearance of fluorescence signal[1] from the 6*s* level above 8 mTorr was ascribed to radiation trapping, while Aron and Johnson[2] have reported the complete absence of any MPI signal due to this transition, and attributed the anomalous pressure effects to xenon dimers. In that MPI study,[2] 4*f* four-photon atomic resonances were observed in the wavelength region of the expected three-photon 6*s* resonance. Compton *et al.*[3] reported that under collision-free conditions, three-photon resonantly enhanced MPI signals through the 6*s* level dominate the ionization spectrum of xenon. More recently, Miller *et al.*[4,5] have shown that the strong 6*s* and 6*s*′ ionization signal shifts to the blue (in excess of the ac Stark effect) and disappears as pressure is increased above a few Torr. This is accompanied by the production of intense third-harmonic radiation, and it is suggested that third-harmonic generation (THG) and MPI are competitive processes. In a theoretical model, Payne, Garrett, and Baker[6] have attributed the loss of right-angle fluorescence and ionization signals to resonantly enhanced THG following a coherent, collective excitation of an ensemble of xenon atoms. A competition between the third-harmonic field and the laser field (or the one-photon Rabi frequency and the three-photon Rabi frequency) produces these effects.[7] Coherent loss mechanisms are generally excluded in rate equation models of MPI. In this work we report the first direct experimental evidence that xenon MPI through the 6*s* and 6*s*′ levels is quenched by THG. This is accomplished by observing xenon MPI under conditions for which THG is forbidden.

To achieve an unambiguous experiment in which three-photon resonantly enhanced MPI through the 6*s* and 6*s*′ intermediate states is allowed but THG is forbidden, it is necessary to consider the selection rules for MPI and THG with respect to excitation laser polarization. The polarization constraints on THG arise from angular momentum conservation and wave-vector phase-matching considerations.[8] They are the following: for linearly polarized light THG is allowed, for circularly polarized light THG is forbidden (requirement of angular momentum conservation), and for counterpropagating circularly polarized (CPCP) light THG is forbidden (wave-vector phase-matching requirement). Identical polarization constraints arise when treated in the formalism of the two-level collective-emission model.[7] Neglecting hyperfine interactions, the electric dipole selection rules for xenon atoms, which follow (j,j) coupling, are $\Delta J = 0$, ± 1; $0 \not\to 0$ for each single-photon transition.[9] If circularly polarized photons are used, the additional one-photon selection rule is $\Delta M = \pm 1$ where (+) refers to right-hand circular (RHC) and (−) refers to left-hand circular (LHC) polarized light. In a three-photon transition circularly polarized light gives $\Delta M = \pm 3$. Thus, three-photon resonant transitions to the $6s[\frac{3}{2}]^o$ $J=1$ or $6s'[\frac{1}{2}]^o$ $J=1$ states of xenon are allowed for linearly polarized light, but forbidden for either RHC or LHC polarized light. The use of CPCP light beams is an experimental condition in which three-photon excitation to the $6s$ or $6s'$ level is allowed. If the counterpropagating photons are both RHC or both LHC, they have opposite senses of rotation in the atom-fixed reference frame. Consequently, three-photon transitions to a $J=1$ state are allowed if the atom absorbs two photons from one laser beam and one photon from the other yielding a net $\Delta M = \pm 1$, and $\Delta J = +1$. Competition between MPI and THG may be observed by comparing the three-photon resonant MPI signal produced by linearly polarized and CPCP laser beams.

The experimental apparatus, shown in the upper panel of Fig. 1, consists of a parallel-plate ionization cell and polarizing optics. The lower panel of Fig. 1 displays the polarization of the light beam as it propagates through the apparatus and reflects back upon itself. The light source is a frequency-tripled Nd-doped yttrium aluminum garnet laser–pumped tunable dye laser (Quanta Ray). The Glan-air polarizing prisms ensure the polarization purity, and the BK-7 Fresnel rhombs (Karl Lambrecht) are mounted to act as achromatic quarter-wave plates. The 10-ns laser pulses are focused by 150-mm focal length Suprasil lenses. The MPI cell which consists of a four-way cross, fitted with BK-7 entrance and exit windows, is connected to a vacuum system. Photoelectrons resulting from the MPI process are monitored by a grounded flat-plate platinum collector. A negative bias of 10–50 V applied to the other platinum plate allows operation in a linear-counting regime.[2] The signal is amplified (PAR 115) and averaged in a boxcar integrator (PAR 164/160) before being recorded on an x-y plotter.

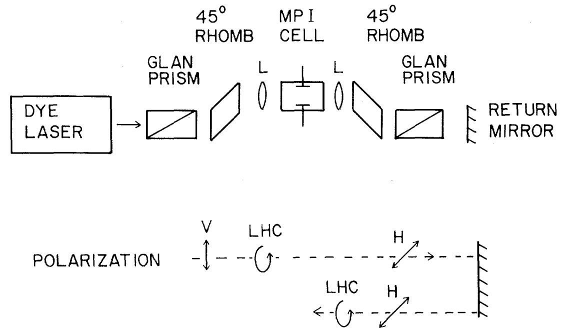

FIG. 1. Schematic diagram of the apparatus used to measure the MPI spectra (upper panel) and the polarization of the CPCP laser beams (lower panel).

Figure 2 shows the MPI spectrum in the region of the $6s[\frac{3}{2}]^o$ $J=1$ and the $4f$ intermediate resonances as a function of pressure and laser polarization. The former is a three-photon resonant, five-photon ionization process, and the latter are four-photon resonant, five-photon ionization processes. The $4f$ states have J's ranging in value from 0 to 5; signals due to various $4f$ states are observed for the three laser polarizations. As a result of the short duration of the laser pulses, temporal as well as spatial overlap of the two laser beams is less than 100% in the CPCP case, and, since the signal in some of the $4f$ levels may be produced independently by each beam, the observed $6s$ to $4f$ ratios in the CPCP experiments represent lower bounds. Comparing the $6s$ to $4f$ signal intensities for each of the three laser polarizations, it is clear that in the case of CPCP light, the $6s$ line dominates the MPI spectrum in this wavelength region for all xenon pressures. This observation, in light of the selection rules for resonantly enhanced MPI and THG, is strong evidence for the quenching of the MPI by the third-harmonic field. The low pressure ($< 10^{-5}$ Torr) atomic-beam study of Compton *et al.*[3] and similar studies in this laboratory show that with linearly polarized light the $6s$ five-photon ionization signal is large. In those experiments the number density is insufficient for THG. However, unlike the CPCP experiment, the low-pressure results do not rule out the possibility that other collisional effects are responsible for the disappearance of the $6s$ MPI.

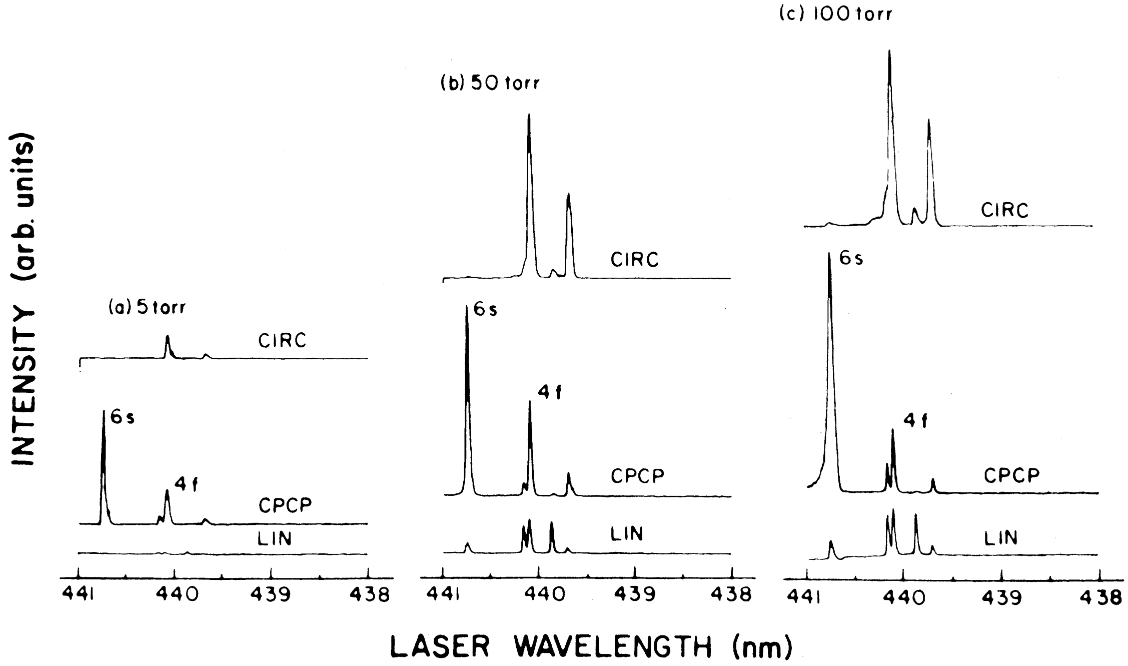

FIG. 2. A portion of the MPI spectrum showing the 6*s* and 4*f* resonances taken with linearly polarized light (LIN), circularly polarized light (CIRC), and CPCP light as a function of xenon pressure. The Coumarin 440 dye-laser energy is 0.7 mJ for the CPCP case, and 1.4 mJ for both linear and circular polarization.

In separate experiments using an apparatus described elsewhere,[10] we observe THG using linearly polarized light and the laser conditions and optics described here. As expected, no third-harmonic emission could be detected by using circularly polarized light. The broadening to the *red* of the 6*s* line for CPCP light at 100 Torr is thought to be due to xenon dimer absorption.[11]

Further evidence for the competition between MPI and THG is given in Fig. 3, which displays the MPI spectrum of xenon in the three-photon wavelength region of the $6s'[\frac{1}{2}]^o$ $J=1$ state taken with CPCP laser photons. Also shown is the data of Miller and Compton for a xenon pressure of 100 Torr and linearly polarized light. Ionization through the 6*s*′ level is a three-photon resonantly enhanced four-photon ionization process. Miller and Compton reported,[5] as in the 6*s* case, that for pressures above a few Torr no ionization could be detected at the resonance position. However, unlike the 6*s* case, the MPI signal does not disappear but continues to broaden and blue shift in a manner similar to the THG signal. With CPCP light, we detect only sharp intense MPI signal at the 6*s*′ resonance frequency at all xenon pressures investigated. In fact, the magnitude of the MPI signal obtained with CPCP light is of interest. In contrast to the relatively weak signal observed from linearly polarized light, at 5 Torr the CPCP laser beam experiment yields on the order of 10^{12}–10^{13} photoelectrons per pulse, which is near volume saturation within the laser-beam waist. This observation is consistent with the large single-photon oscillator strength[12] of 0.27 measured for the 6*s*′ transition.

The origin of the broad ionization reported in previous work was associated in part with molecular bands due to xenon dimers[11] and excitation-ionization processes involving both the third-harmonic and dye-laser photons. This explanation, suggested by Miller and Compton,[5] is consistent with the pressure dependencies first observed by Aron and Johnson.[2] Other detailed MPI-THG studies[7,13] arrive at a similar conclusion. In the present work where third-harmonic emission is suppressed, we do not observe the broad ionization. This result supports excitation-ionization mechanisms involving third-harmonic photons.

In summary, use of CPCP light is a method of suppressing third-harmonic emission while preserving three-photon resonantly enhanced MPI through the 6*s* and 6*s*′ intermediate states in xenon gas. The previously reported disappear-

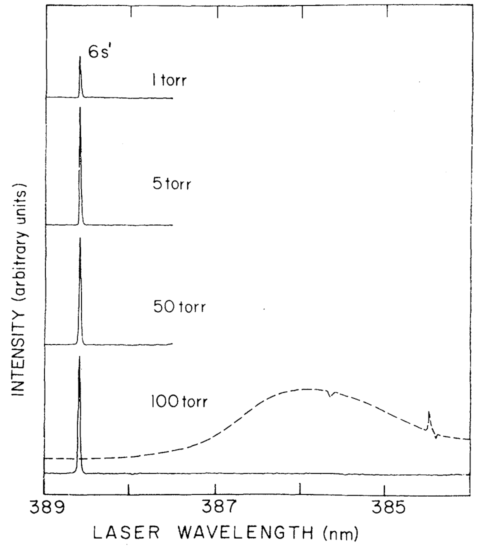

FIG. 3. A portion of the MPI spectrum showing the $6s'$ resonance taken with CPCP laser light as a function of xenon pressure. Also shown in dashed lines is the MPI data of Miller and Compton taken with linearly polarized light at a xenon pressure of 100 Torr. The dye-laser energy is 0.7 mJ for the CPCP experiment. (Adapted, with permission, from Ref. 5.)

ance of the $6s$ and $6s'$ resonant MPI signal at xenon pressures of a few Torr does not occur in the CPCP experiment. The large increase in the MPI signal at the resonance frequencies that occurs in the absence of THG at all xenon pressures investigated is direct experimental evidence that MPI and THG are competing processes. The present study confirms the suggestion of such a phenomenon by Miller *et al.*[4,5] and is consistent with the two-level collective-emission model.[6] These results demonstrate the important role that coherent loss mechanisms may play in high-laser-intensity spectroscopic studies, and suggest that such effects be included in MPI theoretical modeling.

The authors thank W. R. Garrett, M. G. Payne, D. J. Jackson, and J. J. Wynne for discussing their results prior to publication. This work was performed under the auspices of the U. S. Department of Energy.

---

[1] F. H. M. Faisal, R. Wallenstein, and H. Zacharias, Phys. Rev. Lett. 39, 1138 (1977).

[2] K. Aron and P. M. Johnson, J. Chem. Phys. 67, 5099 (1977).

[3] R. N. Compton, J. C. Miller, A. E. Carter, and P. Kruit, Chem. Phys. Lett. 71, 87 (1980).

[4] J. C. Miller, R. N. Compton, M. G. Payne, and W. R. Garrett, Phys. Rev. Lett. 45, 114 (1980).

[5] J. C. Miller and R. N. Compton, Phys. Rev. A 25, 2056 (1982).

[6] M. G. Payne, W. R. Garrett, and H. C. Baker, Chem. Phys. Lett. 75, 468 (1980).

[7] M. G. Payne and W. R. Garrett, Phys. Rev. A, to be published, and private communication.

[8] D. C. Hanna, M. A. Yuratich, and D. Cotter, in *Nonlinear Optics of Free Atoms and Molecules*, edited by D. L. MacAdam (Springer-Verlag, New York, 1979), Chap. 4.

[9] E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge Univ. Press, Cambridge, England, 1963).

[10] J. H. Glownia and R. K. Sander, Appl. Phys. Lett. 40, 648 (1982).

[11] M. C. Castex and N. Damany, Chem. Phys. Lett. 24, 437 (1974).

[12] P. G. Wilkinson, J. Quant. Spectrosc. Radiat. Transfer 6, 823 (1966).

[13] D. J. Jackson and J. J. Wynne, private communication.