Control of Laser Induced Continuum Structure in the Vicinity of Autoionizing States

O. Faucher,[(1)] D. Charalambidis,[(1)] C. Fotakis,[(1)] Jian Zhang,[(2),(3)] and P. Lambropoulos[(1),(2)]
[(1)]*Foundation for Research and Technology, Hellas, Institute of Electronic Structure and Laser, P.O. Box 1527, Heraklion 71110, Crete, Greece and Department of Physics, University of Crete, Crete, Greece*
[(2)]*Max-Planck-Institut für Quantenoptik, W-8046 Garching, Germany*
[(3)]*Laboratoire des Collisions Atomiques, Université de Bordeaux I, 351 Cours de la Libération, 33405 Talence Cedex, France*
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The first observation of laser induced continuum structure on autoionizing states in both third harmonic generation and ionization is reported for atomic Ca. The possibility of controlling the shape of the induced structure, exhibiting a transition from a dip to a peak, through the laser field intensities is demonstrated. Theoretical time dependent analysis including calculated atomic parameters verifies that the experimental observations originate from laser field dependent coherent interactions.

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The modification of transitions within the atomic continuum [1–6] through the combination of coherent interactions has been a challenging experimental undertaking even for the relatively straightforward case of the smooth continuum of a one electron atom. Thus, it was only recently that the modification of ionization due to laser induced continuum structure (LICS) was observed [7,8] in Na. The effect of LICS on harmonic generation rests so far on a single experimental report [2,3] which has proven difficult to replicate under somewhat different conditions [9,10] and to extend to other atoms.

The modification of transitions within the continuum of an atom that already possesses structure due to intra-atomic interactions (such as the manifold of autoionizing doubly excited states) poses an even more challenging problem, but with potentially far reaching consequences. Such doubly excited states represent high degrees of excitation which, if controllable by external means, can provide interesting schemes for the generation of coherent, short-wavelength radiation either through nonlinear optical processes [11,12] or more novel schemes such as lasing without inversion [13,14].

The experimental challenge in this problem is compounded by its theoretical counterpart, as it is far from evident what type of additional structure should be expected. Simple models [6] of the type that have been qualitatively useful for LICS in one electron atoms cannot be expected to be relevant to this much more complex problem.

It is the purpose of this paper to report the first observation of the effect of LICS on autoionizing states (AIS) in Ca, as manifested in both third harmonic generation (THG) and ionization; including a theoretical analysis providing the basis for the interpretation.

The basic coupling scheme is shown in Fig. 1. We have investigated several variations of this basic arrangement over a range of intensities for both lasers I and II, with respective frequencies ω_1 and ω_2. In all cases, laser I was on resonance with one of the three AIS of the configuration $3d4f$, but it also happens to be near two-photon resonance with the bound $4s4d\,^1D_2$ state. When in exact two-photon resonance with the above state, the third photon lies only 0.8 cm^{-1} above the $^3P_1$ AIS, whose width is $\sim$3 cm^{-1}, leading thus to double resonance. The second laser II has been tuned so as to couple either the bound states $4s5s\,^1S_0$ or $4s6s\,^1S_0$ with the continuum around the manifold of the three AIS. Obviously, when ω_2 is tuned exactly on resonance with one of the AIS, it couples the corresponding bound state not only to the continuum but also to the discrete (or more generally the closed channel) component of the wave function to which the ground state has been coupled by three ω_1 photons. This is not the place to list and discuss all types of results we have obtained under the various coupling arrangements. We shall instead focus our discussion on one of our more novel results, namely, the first observation of the reduction of an autoionizing resonance to a window resonance

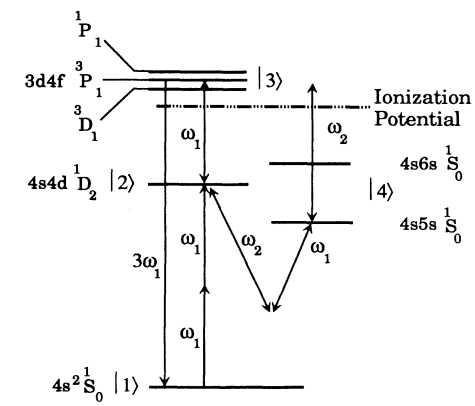

FIG. 1. The most relevant Ca energy levels and couplings. Laser I is frequency tripled above the threshold of ionization. Laser II couples either the low-lying $4s5s$ or $4s6s$ bound states to the continuum and to the $4s4d\,^1D_2$ state via the two depicted two-photon Raman process.

in both ionization and THG, through laser induced coupling to a bound state. At the same time, we demonstrate the control of the shape of the resonance through the proper combination of the intensities of the two lasers.

The experimental setup is based on a common single metal heat pipe oven [15]. The Ca atomic number density at a temperature of 900 °C is of the order of 7×10^{14} cm^{-3}. The He buffer gas pressure in the oven is 15 mbar. During each run, the two excimer pumped dye laser beams (0.2 cm^{-1} bandwidth, 12 ns pulse duration) are combined through a dichroic mirror and focused by an $f=30$ cm lens in the heat pipe. The spatial and temporal overlapping of the two laser beams has been optimized through maximization of the signal of both two-photon resonant (i) four-wave mixing and (ii) two-color multiphoton ionization. A vacuum ultraviolet (VUV) monochromator, connected to the heat pipe via a LiF window, is employed to separate the THG wave from the laser light. The VUV signal is detected by a solar blind photomultiplier equipped with an aluminium filter to reduce the scattered laser light background. A biased charge collection electrode placed in the heat pipe, near the laser focus, is used as an ion detector for measuring the multiphoton ionization (MPI) signal. Simultaneous MPI and THG data acquisition is performed on a shot-to-shot basis by two boxcar integrators interfaced with a microcomputer.

Turning now to the discussion of the results, we consider first (Fig. 2) the induced structure observed in the THG when $2\omega_1$ is on resonance with the $4s4d\,^1D_2$ state and ω_2 (coupling the $4s5s\,^1S_0$ to the continuum) is scanned around the $^3P_1$ autoionizing state. As expected, the harmonic signal is strongly enhanced as a result of the double resonance. But in contrast to the only existing observation of LICS in THG [2,3], in which an eighteen-fold enhancement of the signal has been reported, a strong decrease of the THG signal is observed in the present experiment when the energy balance condition $E_{4s^2}+3\hbar\omega_1=E_{4s5s}+\hbar\omega_2$ is satisfied. The inserted spectra in Fig. 2 provide the verification that this condition is met, as they show the expected shift of the observed dip for three different fixed values of ω_1. The origin of the dip can be traced to the minimization of the bound-bound coupling between the $4s4d\,^1D_2$ and $4s5s\,^1S_0$ state via the two-photon Raman processes shown in Fig. 1. Recalling the picture of the "conventional" (not induced) autoionization, weak bound-bound coupling leads to a q parameter close to zero and thus to a window resonance. In our case, the weak coupling of the two discrete states can be attributed to partial cancellation between the two coherent Raman processes, one through the continuum and the other through the bound spectrum. A significant role in this feature is played by the presence of the autoionizing state which lies close to the virtual state of the Raman process through the continuum. This increases the

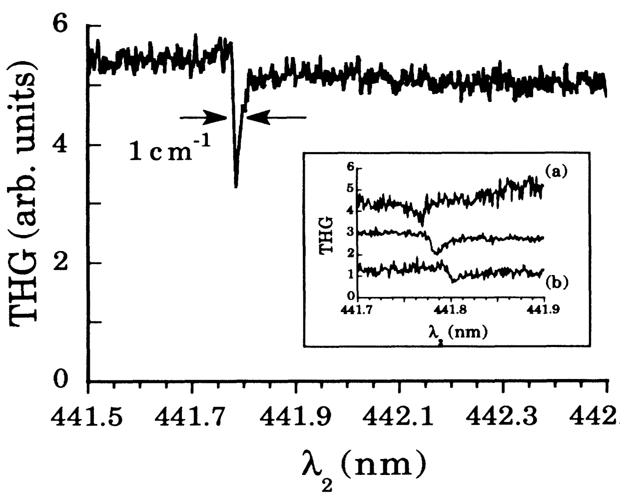

FIG. 2. Window resonance observed in the two-photon resonant third harmonic generation profile at $I_1=4\times10^9$ W/cm^2. The $4s5s\,^1S_0$ state is coupled to the continuum with $I_2=1.5\times10^9$ W/cm^2. The laser induced structure appears when ω_2 matches the energy where the $3\omega_1$ excitation embeds the ground state. The bottom spectra show the expected shift of the structure when the harmonic wave is generated in (a) the blue side or (b) the red side of the two-photon resonance.

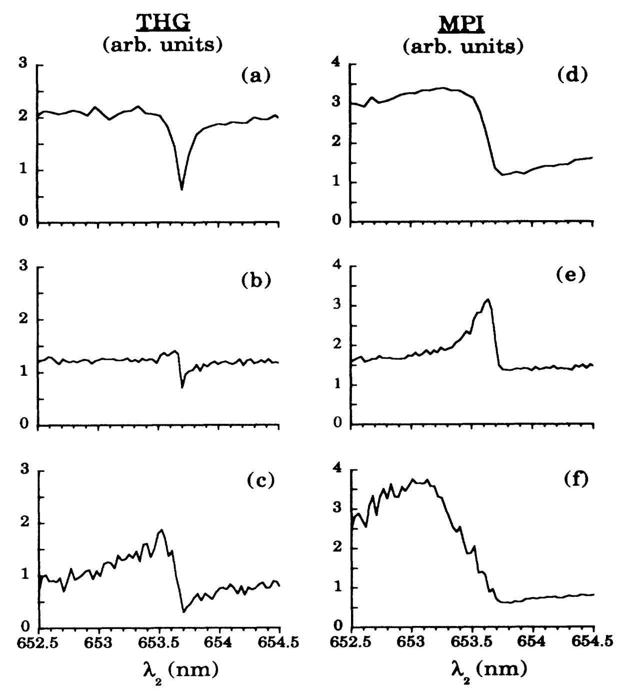

FIG. 3. Third harmonic and multiphoton ionization spectra recorded for different laser intensities. In this case, LICS is achieved when $3\omega_1$ is resonant with the $3d4f\,^1P_1$ autoionizing state, whereas ω_2 couples the $4s6s\,^1S_0$ state to the continuum. (a),(d) $I_1=5.2\times10^9$ W/cm^2, $I_2=2.2\times10^9$ W/cm^2. (b),(e) $I_1=5.2\times10^9$ W/cm^2, $I_2=5.5\times10^8$ W/cm^2. (c),(f) $I_1=1.1\times10^9$ W/cm^2, $I_2=2.2\times10^9$ W/cm^2.

strength of this Raman process (which often is weak) [16] thus making possible the partial cancellation.

The picture described above is not sensitive to the coupled states. As shown in Fig. 3(a), a similar dip is observed when $3\omega_1$ is on resonance with the ${}^1P_1$ state, with ω_2 coupling the $4s6s\ {}^1S_0$ state to the continuum. On the other hand, the laser induced structure is definitely affected by the power densities of the two lasers. As clearly demonstrated in Fig. 3 for three different sets of laser intensities, the shape of the observed structure in the THG spectra is changing from a dip [Fig. 3(a)] to an asymmetric profile [Fig. 3(c)], exhibiting a maximum and a minimum corresponding to 50% enhancement and 60% depletion of the harmonic signal, respectively. A strong dependence on the field intensities is also found for the LICS in MPI, as shown in Figs. 3(d)–3(f).

A dependence of an autoionizing line shape on intensity, exhibiting a transition from asymmetric peak, to step-like profile, to dip, had been predicted some time ago [17] in connection with the excitation of an AIS by a strong field. This is the first observation of an effect of this type, except that it is not the large intensity but the coherent superposition of interactions (combined with the appropriate intensities) that creates the effects reported in Fig. 3.

In order to provide theoretical insight into the above observations, we have developed a model that incorporates the necessary atomic structure as well as the formalism for the temporal dynamics of the system under the two pulsed lasers. Realistic atomic parameters are here indispensable, represent a project in itself, and have been obtained through an L^2 basis constructed with B-splines along lines that have been discussed elsewhere [11,12,18,19]. Calcium presents the further complexity of strong singlet-triplet mixing which cannot be addressed here, and we shall confine our theoretical analysis to the modification of the AIS $3d4f\ {}^1P_1$. The theoretical values of the main parameters are listed in Table I.

Because of the proximity of state $4s4d\ {}^1D_2$ to two-photon resonance, it must be included in the density matrix equations governing the dynamics of the system. Including also the AIS, we have four states and the respective continuum to be included in the equations, which leads to a set of ten differential equations; too many to be exhibited and discussed here. The structure and solutions of a similar set of equations, but without the AIS can be found in Ref. [16]. The presence of an AIS makes the respective Rabi frequency complex as, for example, in Ref. [20]. These equations must be solved under the proper temporal laser pulse shapes (which were here taken to be Gaussian) approximating the experimental conditions. In general, an integration over the spatial distribution of the laser intensity must also be performed, especially when saturation of the ionization occurs. This was not necessary in this paper, because in the experimental data under interpretation saturation was not a severe problem as it had been in the work of Ref. [16].

We have summarized a typical set of results in Fig. 4 demonstrating the type of effects observed in the experiment. With the laser I kept tuned to two-photon resonance with state $|2\rangle$ (Fig. 1), we have calculated THG and ionization as a function of detuning of the coupling laser II. The top three pairs of frames in Fig. 4 show THG and ionization for different combinations of laser intensities. Ionization is plotted in terms of the fraction of atoms ionized; thus 1.0 in that scale means complete ionization. As shown in the top three pairs of frames, THG and the corresponding ionization revert from dip to peak depending on the combination of intensities I_1 and I_2. It should be noted that, in the pair of frames (c) and (g), THG exhibits structure (dip) even though ionization is completely flat due to saturation which stems from the presence of the two-photon resonance involved in these six fames. This can be readily understood if one notes that harmonic generation occurs while the atom exists, while ionization represents the cumulative effect of the ions produced during the pulse. It follows then that THG may be expected to exhibit interference effects even after integration over the spatial distribution of radiation.

In the fourth pair of frames of Fig. 4, laser I has been tuned to an exact three-photon resonance with the AIS, and is therefore tuned above two-photon resonance with state $|2\rangle$ by 34.8 cm^{-1} which correspond to the conditions of the experimental data of Fig. 3. After a study with various combinations of intensities, we have obtained the result shown in frames (d) and (h). We thus have a case in which both THG and ionization exhibit dips as a function of the detuning of the coupling laser. This was obtained only when the AIS was involved in the coupling scheme and corresponds to the top pair of frames of the experimental data in Fig. 3. Results of this type exhibiting sensitivity to the combination of intensities suggest that the experimental outcome will be sensitive to the spatial overlap of the two beams. This has in fact been

TABLE I. Coupling parameters, i.e., one- and two-photon generalized Rabi frequencies Ω_{ij}, photoionization widths γ_i, and autoionizing width Γ_3 (subscripts corresponding to the states shown in Fig. 1). I_1 and I_2 are in units of $\mathrm{W/cm^2}$.

$\Omega_{12}^{(2)}$ (rad/sec)	$\Omega_{23}^{(1)}$ (rad/sec)	$\Omega_{24}^{(2)}$ (rad/sec)	$\Omega_{34}^{(1)}$ (rad/sec)
$-86.26I_1$	$(-6.83\times10^7+i1.34\times10^6)(I_1)^{1/2}$	$(-198.0+i26.02)(I_1I_2)^{1/2}$	$(2.81\times10^7+i1.75\times10^6)(I_2)^{1/2}$

γ_2 (rad/sec)	γ_4 (rad/sec)	Γ_3 (rad/sec)
20.42 (I_1+I_2)	33.93 (I_1+I_2)	9.05×10^{10}

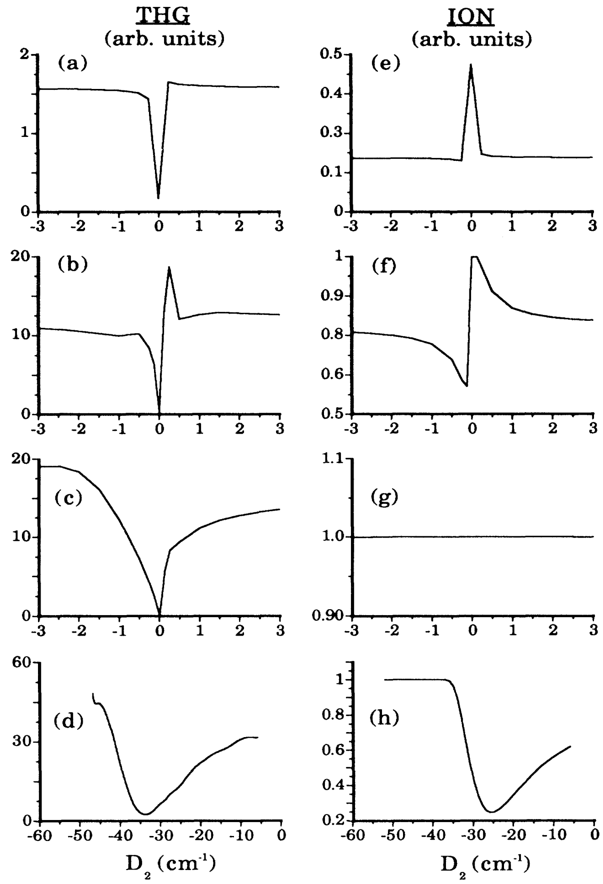

FIG. 4. Calculated third harmonic (left) and ionization (right) spectra for different laser intensities I_1 and I_2. (a),(e) $I_1=I_2=1.0\times10^7$ W/cm^2. (b),(f) $I_1=I_2=1.0\times10^8$ W/cm^2. (c),(g) $I_1=2.0\times10^9$ W/cm^2, $I_2=5.0\times10^9$ W/cm^2. (d),(h) $I_1=2.0\times10^9$ W/cm^2, $I_2=5.0\times10^9$ W/cm^2. The spectra are plotted vs the detuning of laser II, as defined by $D_2=(E_{|2\rangle}-E_{|4\rangle})-\hbar(\omega_2-\omega_1)$. Laser I is either two-photon resonant with the state $|2\rangle$ [frames (a)-(c) and (e)-(g)] or three-photon resonant with an AIS [frames (d) and (h)].

tested in our experiment and found to be indeed the case. We have been able to vary the line shapes and convert peaks (dips) to dips (peaks) by modifying the spatial overlap and the intensities of the two laser beams.

In conclusion, we have demonstrated the possibility of LICS on autoionizing states in both THG and ionization. Our theoretical analysis confirms that the observed induced structure can be produced by coherent interactions manipulated through the proper choice of the laser fields. The sensitivity to the intensities and frequencies suggests a variety of further possibilities not only in the studies of AIS but also in problems of nonlinear optics and lasing without inversion. Detailed accounts of both experiment and theory will appear in a forthcoming paper.

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[1] Yu. I. Heller, V. F. Lukinykh, A. K. Popov, and V. V. Slabko, Phys. Lett. **82A**, 4 (1981).

[2] L. I. Pavlov, S. S. Dimov, D. I. Metchkov, G. M. Mileva, and K. V. Stamenov, Phys. Lett. **89A**, 441 (1982).

[3] S. S. Dimov, L. I. Pavlov, and K. V. Stamenov, Appl. Phys. B **30**, 35 (1983).

[4] D. Feldmann, G. Otto, D. Petring, and K. H. Welge, J. Phys. B **19**, 269 (1986).

[5] M. H. R. Hutchinson and K. M. M. Ness, Phys. Rev. Lett. **60**, 105 (1988).

[6] P. L. Knight, M. A. Lauder, and B. J. Dalton, Phys. Rep. **190**, 1 (1990).

[7] Y. L. Shao, D. Charalambidis, C. Fotakis, Jian Zhang, and P. Lambropoulos, Phys. Rev. Lett. **67**, 3669 (1991).

[8] S. Cavalieri, F. S. Pavone, and M. Matera, Phys. Rev. Lett. **67**, 3673 (1991).

[9] K. G. H. Baldwin, P. B. Chapple, H. A. Bachor, J. Zhang, and P. Lambropoulos, in *Nonlinear Optics*, Kauai, HI, 16–20 July 1990 (IEEE, New York, 1990).

[10] S. Cavalieri (private communication).

[11] Jian Zhang, thesis, University of Southern California, 1991 (unpublished).

[12] P. Lambropoulos, Jian Zhang, and X. Tang, in *Coherence Phenomena in Atoms and Molecules in Laser Fields*, edited by A. D. Bandrauk and S. C. Wallace (Plenum, New York, 1992), p. 255.

[13] V. G. Arkhipkin and Yu. I. Heller, Phys. Lett. **98A**, 12 (1983).

[14] S. E. Harris, Phys. Rev. Lett. **62**, 1033 (1989).

[15] C. R. Vidal and J. Cooper, J. Appl. Phys. **40**, 3370 (1969).

[16] Jian Zhang and P. Lambropoulos, Phys. Rev. A **45**, 489 (1992).

[17] P. Lambropoulos and P. Zoller, Phys. Rev. A **24**, 379 (1981).

[18] X. Tang, T. N. Chang, P. Lambropoulos, S. Fournier, and L. F. DiMauro, Phys. Rev. A **41**, R5265 (1990).

[19] T. N. Chang, Phys. Rev. A **39**, 4946 (1989).

[20] A. Lyras, X. Tang, P. Lambropoulos, and Jian Zhang, Phys. Rev. A **40**, R4131 (1989); **44**, 796 (1991).