Phase-sensitive ionization through multiphoton-excitation schemes involving even numbers of photons

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(Received 4 November 1998)

Strongly field-phase-dependent ionization is demonstrated by using a convenient experimental setup appropriate for excitation schemes involving an even number of photons. In this paper, ionization in Xe occurs via two phase-correlated electromagnetic fields, namely, the fundamental and the second harmonic of the laser. Since the fields are in the visible and ultraviolet regime, they have readily controllable amplitudes. Thus excitation interference conditions and, consequently, phase-sensitive ionization can be easily and efficiently established. [S1050-2947(99)03706-3]

PACS number(s): 32.80.Qk, 32.80.Fb, 32.80.Rm

Several experimental and theoretical studies have been recently conducted in what has now been established as phase-sensitive coherent control. Phase control refers to the change of the excitation probability of bound or continuum states, through the external adjustment of the relative phase of different phase-correlated excitation pathways involving a different number of photons of different energies. The excitation process can be controlled by the interference of the different pathways and, consequently, the excitation probability is determined by their relative phase. Excitation and ionization rates, ionization and dissociation branching ratios, autoionization line shapes, angular distributions of photoabsorption products as well as frequency up conversion may be manipulated exploiting this effect [1–12].

In atoms, phase control has to involve absorption channels with either only odd or only even number of photons in the case of excitation of bound states or continuum states, which are not observed angularly resolved (i.e., when total ionization is measured). This is because all the interfering channels must have the same initial and final state and thus parity as well as angular momentum has to be conserved due to the central potential of the atom. In the vast majority of the phase-sensitive control experiments in laser atom/molecule interactions, absorption of three photons of the fundamental frequency and one of its third harmonic has been employed, as this interference scheme involves the lowest possible (odd) number of photons in centrosymmetric systems. In this type of experiment complications arise inherent to the generation and utilization of the third harmonic wave, which in most cases is in the vacuum ultraviolet spectral region. Some of those are the lack of amplitude stability, the low conversion efficiency and hence the low third harmonic power, the strong absorption and scattering, the limited number of transmitting materials and optical elements in this region, the nonvisible radiation as well as the high-vacuum requirements.

For excitations involving absorption of an even number of photons in all the interfering channels, the use of third harmonic generation is no longer necessary. This type of excitation scheme allows high degree of excitation through visible or UV frequencies. Recently phase control of ionization has been demonstrated in four-photon resonant five-photon ionization [4+1 resonantly enhanced multiphoton ionization, (REMPI)] through the $5p[5/2]_2$ and $4f[3/2]_2$ states of Kr and Xe, respectively [13], where the second excitation channel of the resonant state included one third harmonic and one laser photon. Phase-insensitive coherent control has also been demonstrated in two-color, two-photon vs two-photon excitation schemes [14–16]. One notices, however, that in the work of Ref. [13] the two-photon excitation channel of the bound state does not need to involve one third harmonic photon and one fundamental photon but can also be reached by absorbing two second harmonic photons. The second harmonic, which lies in the ultraviolet spectral region, can be generated using nonlinear crystals and, consequently, is much more intense and convenient to handle.

The excitation probability of the bound state now reads

$$W \propto [\mu^{(4)}E_{10}^4]^2 + [\mu^{(2)}E_{20}^2]^2 + 2\mu^{(4)}\mu^{(2)}E_{10}^4E_{20}^2 \times \cos[2(\vartheta_2 - 2\vartheta_1)], \quad (1)$$

where the indices $i=1,2$ refer to the fundamental and the second harmonic field. $\mu^{(n)}$ is the effective n-photon (four visible and two UV) electric dipole moment of the transition

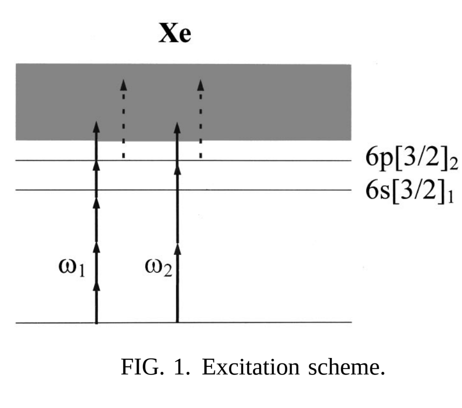

FIG. 1. Excitation scheme.

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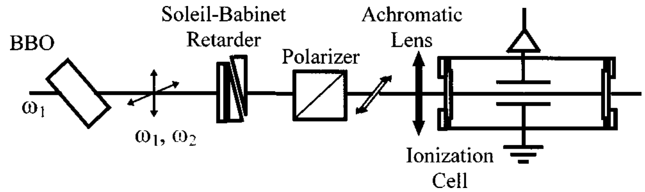

FIG. 2. Experimental setup.

between the ground state and the resonant state, while E_{i0} is the electric-field amplitude of the laser beams. Hence, the probability can be controlled through the phase difference $\Delta\vartheta = 2(\vartheta_2 - 2\vartheta_1)$.

Employing the second instead of the third harmonic has strong advantages with respect to the production process, the intensity, stability and visibility of the beam, as well as concerning the possibility of its propagation in several media and, consequently, the retardation mechanism of its phase relative to that of the fundamental. Experimental setups for this type of phase-control experiments set much less severe vacuum requirements and become simpler. The experimental procedure is expected to provide increased stability and controlled reproducibility.

In this paper we demonstrate phase-sensitive ionization in a scheme of $4+1$ (and $2+1$) REMPI in Xe, which now involves pathways including absorption of photons of the fundamental and the second harmonic. The resonant state is the $6p[3/2]_2$ as shown in the excitation scheme depicted in Fig. 1. It is excited via (i) four-photon absorption at the fundamental frequency and (ii) two-photon absorption of the second harmonic.

The experimental setup used is shown in Fig. 2. The laser beam is delivered by an excimer-pumped dye laser. The wavelength of the fundamental is tuned to 504.97 nm. The pulse duration is of the order of 15 ns. The second harmonic is produced in a type I BBO crystal. Its polarization is thus perpendicular to that of the fundamental. Both beams are propagated through a Soleil-Babinet compensator, the orientation of which is chosen so that its two optic axes are parallel to the two polarization planes of the two fields. In this arrangement the Soleil-Babinet compensator is essentially used as a variable-thickness optical plate and not in order to vary the retardance of a wave as is normally the case. Utilizing the normal dispersion characteristics of the quartz material from which the compensator was made, the relative phase between the two wavelengths could be adjusted. A linear polarizer, placed behind the Soleil-Babinet retarder, is used in order to select the appropriate field amplitudes in a common polarization plane, the polarization plane of the polarizer. The two excitation amplitudes can, in this way, be easily adjusted by rotating the polarizer so as to transmit the required field-amplitude component of the two beams. Both beams are focused by means of an achromatic lens of 12-cm focal length into the ionization cell, which is filled with a few mbar of Xe gas and equipped with a charge collector. The energies of the fundamental and second harmonic at the entrance of the ionization cell are 2.8 and 0.3 mJ, respectively. The beam waist of the focused beam has not been measured, as the absolute intensities of the two laser beams in the interaction region are not relevant in the present paper.

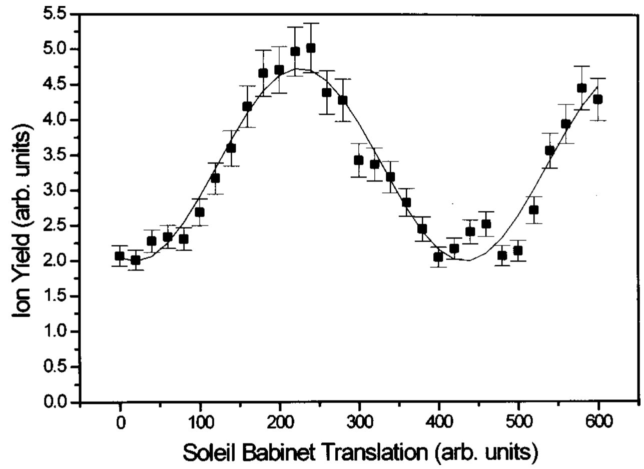

FIG. 3. Phase control in a $4+1$ and $2+1$ ionization scheme in Xe ionization measured as a function of the relative phase of the fields introduced by a Soleil-Babinet retarder.

Similar results with those described below have also been obtained in a Xe atomic beam.

By tuning the relative phase of the two fields through the Soleil-Babinet adjustment, the phase difference $\Delta\vartheta$ could be varied. A strong modulation of the ionization signal could be observed as a function of the Soleil-Babinet prism position as shown Fig. 3. The solid line is a least-square fit of the function:

$$y = (1 + Ax) \times \left[B + C \sin\left(\frac{x}{D} + E \right) \right]. \tag{2}$$

The modulation depth, defined as $I_{max} - I_{min}/1/2(I_{max} + I_{min})$, where I_{max} is the maximum and I_{min} the minimum ion signal of the modulation, has a maximum value better than 0.8 in the present experiment and is close to the highest values obtained in phase-control experiments. The modulation depth can be adjusted through (i) the spatial overlapping of the two beams, which can be optimized by carefully adjusting the phase matching of the BBO crystal to minimize beam walk off, and (ii) the angle of the polarizer, an adjustment that controls the relative amplitudes of the interfering excitation channels.

The present paper demonstrates a simple experimental procedure for phase-sensitive coherent control experiments. By applying this procedure we were able to obtain well reproducible phase-sensitive control of the ionization rates in xenon with high efficiency and under straightforward experimental conditions. The procedure provides a tool for other applications of phase-sensitive control of laser-excitation processes. A further advantage of the scheme employed in this paper is that, since the wavelength difference between the two beams is smaller than that when third harmonic is used, dispersion effects such as the ''phase slip'' in the ionization region, which diminish the depth of the effect, can be reduced.

This work has been carried out at the Ultraviolet Laser Facility of FORTH (TMR Program, Contract No. ERBFMGECT950017). We wish to thank N. E. Karapanagioti for numerous fruitful discussions.

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