Phase and intensity control of integral and differential above-threshold ionization rates

Ricardo A. Blank and Moshe Shapiro
Department of Chemical Physics, Weizmann Institute of Science, Rehovot 76100, Israel
(Received 14 June 1995)

A theory of coherent control of above-threshold ionization (ATI) of hydrogen using light fields composed of a fundamental frequency and its third harmonic is developed. *Phase* control and *intensity* control over *integral* and *differential* quantities is demonstrated. Modulation of the first ATI kinetic-energy peak height by more than 70%, variation in the ratio of the first two peaks of up to a factor of 4, and separation in the alignment of the first two ATI peaks by as much as $\sim 50°$, as a function of the relative phase between the two fields, are demonstrated. Intensity control, in which both partial and total integral ATI rates attain a local maximum at a specific combination of light intensities, is predicted to occur.

PACS number(s): 32.80.Rm; 32.80.Bx; 42.50.Hz

The possibility of coherent control (CC) [1] of above-threshold ionization (ATI) by interfering two frequencies has attracted both experimental [2–6] and theoretical [7–14] interest in the last few years. Early on, two-color schemes were used to modify the pondermotive shifts of ionization thresholds [2,3,7]. In such studies a combination of a strong (typically *ir*) laser, used to shift the ionization potential, and a *weaker uv* laser, used to ionize the atom, was often considered. Subsequent works dealt with the use of a fundamental frequency and its second harmonics [4–6,8,10,11,13] (the "1+2" scenario) [15,16], or the use of a fundamental frequency and its third harmonics [8–10,12,14] (the "1+3" scenario) [17–19]. Control was achieved with respect to the following: the modulations of both the total ionization rate and the relative magnitude of the different ATI kinetic energy peaks [4,9–12,18]; the backward/forward ratio [15,16] of photoelectrons [5,6,8,11]; the transition from the multiphoton regime to the tunneling-ionization regime [6,11,13]; and stabilization induced by very intense fields [14].

The above studies have mainly concentrated on carefully varying the relative phase of the two beams at a small number of fixed intensities. This type of control may be termed *phase control*. Following the general guidelines of CC theory [1,17], we know that the two-color interference effects ought also to be a sensitive function of the intensity ratio. This type of control may be termed *intensity control*. In this paper we explore the combination of phase *and* intensity control on ATI processes. We show that through this combination we achieve a dramatic improvement in the control and altogether new effects. In particular we demonstrate "1+3" control over *integral* rates (e.g., partial and total ionization rates), as well as *differential* quantities such as the angular alignment of each ATI kinetic energy peak. The demonstration is achieved by performing numerically exact computations of three-dimensional (3D) hydrogen in a strong electromagnetic field, using the artificial channel method [21–23].

The essence of CC [1] is the coherent interference between different pathways leading to the same final state. CC interference effects can generally be classified according to whether the same angular momentum (l) states or different l states contribute to a given asymptotic channel [20]. In general, paths involving the same l states allow for integral control while paths involving different l states result in differential control.

As shown in Fig. 1, in the present work both types of interferences are considered. Depicted are possible pathways, such as the two-photon pathways associated with the $S=0$ peak, which involve the same l states ($l=0$ and the $l=2$), leading to integral control, and possible pathways, such as those contributing to the $S=1$ peak, which involve different l states, leading mainly to differential control.

We first present results of integral (Fig. 2) and differential (Fig. 3) *phase control*, in which control is achieved by varying the relative phase of the two beams. Though in this part we keep the intensities constant, the magnitude of the effect depends strongly on the actual intensity values chosen. Therefore, two sets of intensities are considered:[1] One intensity set, "set A," defined as, $I(\omega)=6.216\times10^{-3}$ a.u., $I(3\omega)=3.108\times10^{-3}$ a.u., results in good integral control. In contrast, the other intensity set, "set B," for which $I(\omega)=6.216\times10^{-3}$ a.u., $I(3\omega)=1.554\times10^{-4}$ a.u., gives rise to good differential control.

Figure 2 shows the variation of the rates for the first three ATI kinetic energy peaks as a function of the phase between the two lasers. For set A intensities [Fig. 2(a)], a large modulation (of more than 70%) in the $S=0$ ionization rate is obtained. [The percentage-modulation is defined as $(I_{\max}-I_{\min}/I_{\max})$]. In addition, the ratio between the first two kinetic energy peaks can be made to vary by as much as 4. [See the right-hand scale of Fig. 2(a).] In contrast, for set B intensities shown in Fig. 2(b), the relative phase seems to have only a minor effect on the partial ionization rates.

Figure 3(b) demonstrates differential control in which *directional separation* of two ATI kinetic energy peaks is achieved. This is accomplished by diverting one of the ATI peaks (the $S=1$ peak) away from the polarization direction. We sense that contrary to the "1+2" scenario [4–6,8,10,11,13] which leads to a change in backward/forward ratio (i.e., *orientation*) [15,16], the "1+3" scenario used here leads to change in *alignment*.

[1]The conversion factor used in this paper is 1 a.u. of *intensity* $=6.43\times10^{15}$ W/cm^2. We have not used the number often appearing in the literature of 3.5×10^{16} W/cm^2 because it is a hybrid: it corresponds to 1 a.u. of *field amplitude*.

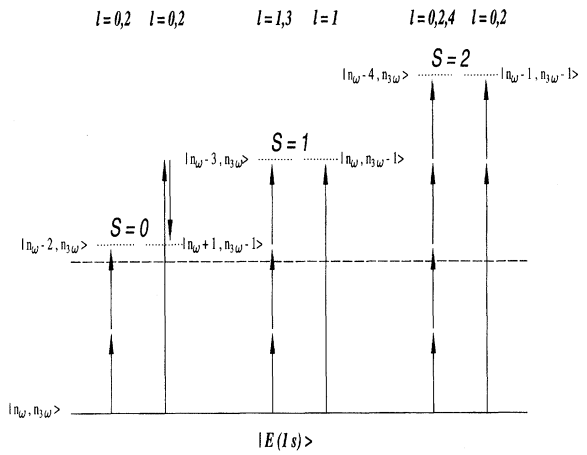

FIG. 1. Present "1+3" scheme for CC of hydrogenic ATI electrons for $\omega=0.2567$ a.u. Drawn are the ground state ($1s$) dressed with $n_{3\omega}$ big photons and n_{ω} small photons, the threshold limit (broken line), and the positions of the first three ATI peaks. Also shown are the number of ω and 3ω photons connecting the ground state with each of the ATI continuum states and the l values involved.

The degree of control over the alignment is calculated using the expression for the differential rate to a specific final kinetic energy. Assuming that we start from $l=0$ bound states, the differential rate, $dR/d\Omega$, is

$$\frac{dR}{d\Omega} \propto \left| \frac{1}{k} \sum_{l=0}^{\infty} i^l \exp[i\eta_l] Y_{l,0}(\theta) T_{l,0}(k) \right|^2 . \quad (1)$$

In the above, θ is the angle between the polarization direction and that of the ejected electron, k is the magnitude of the momentum of the ejected electron, η_l is the Coulombic phase shift [$\eta_l=\arg\Gamma(l+1+i/k)$], and $T_{l,0}(k)$ are the T matrix element between the bound and each of the scattering states correlating asymptotically with an l partial wave and a k momentum value.

Figure 3 shows the differential rates as a function of the spatial angle and the relative phase between the lasers, for the $S=0$ and $S=1$ peaks, at set A intensities (left-hand side), and set B intensities (right-hand side). For set A we see that although the $S=0$ peak does depend on the relative phase, both the $S=0$ and $S=1$ peaks remain centered about $\theta=0$ for all phases. In contrast, for set B the two peaks behave differently, the $S=1$ peak varies strongly with α, while the $S=0$ angular distribution is almost invariant to α. Thus, at $\alpha=2$ rad the $S=1$ peak is maximal at $\theta=50°$ and minimal in the polarization direction ($\theta=0°$). For $\alpha=5$ rad the $S=1$ angular dependence is reversed. Because the $S=0$ peak is essentially zero for $\theta\sim 50°$, tuning the α phase to 2 rad results in dramatic directional control.

The effect we have just described illustrates the power of the present scenario in controlling ATI processes. Past one- and two-color ATI schemes [5,6,11,22] result in the photoelectrons being aligned about the field polarization direction [24] (see, however, Ref. [27]). By using "1+3" interferences we are able to completely divert the alignment of a *specific* ATI kinetic energy peak ($S=1$ in this case) away from the polarization direction.

In Fig. 4 we demonstrate a different effect, that of *intensity* control over *integral* partial (and total) ionization rate. In Fig. 4(a) we plot the rates of the first three ATI kinetic energy peaks as a function of $I(\omega)$ in the presence and absence of the second laser (of frequency 3ω). In Fig. 4(b) we scan the 3ω laser intensity while keeping $I(\omega)$ constant. We note that when the intensity of one of the lasers is very high with respect to the other, the two-color rates converge, as they should, to the one-color rates, which rise monotonically with intensity. In contrast, the two-color rates attain a maximum at

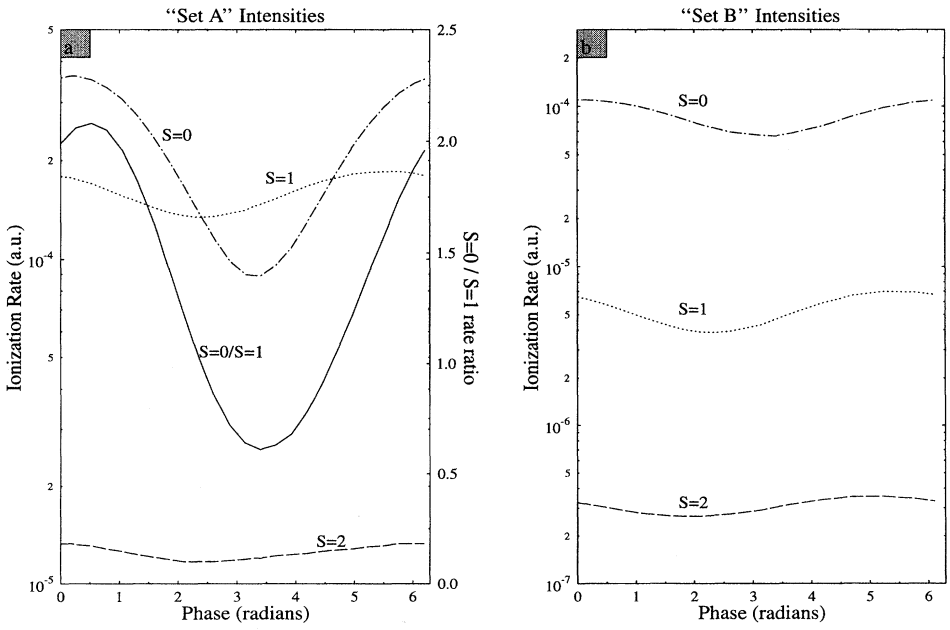

FIG. 2. Integral (phase) control of the ATI peaks as a function of the phase between the two lasers for intensity sets A and B.

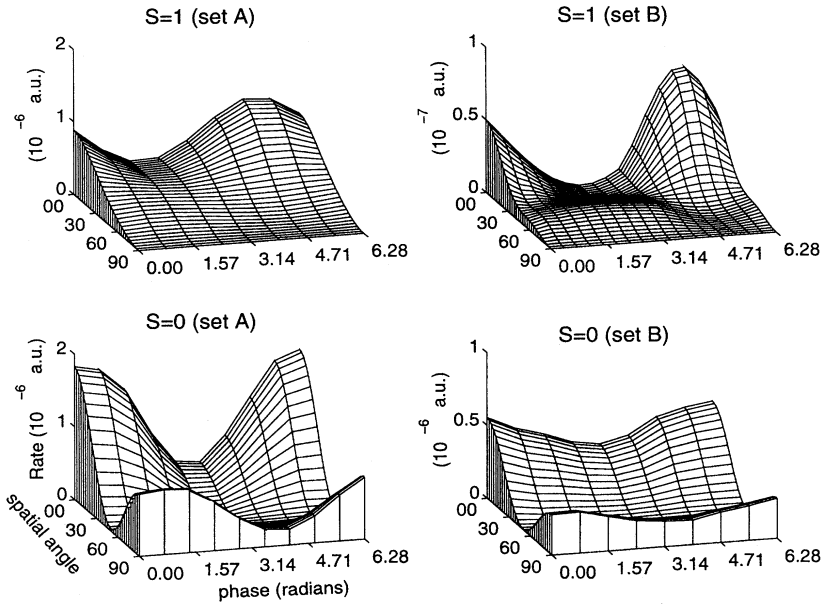

FIG. 3. Differential rates for the $S=0$ and $S=1$ peaks as a function of the relative phase between the lasers, for intensity sets A and B. The spatial angles are given in degrees.

a *particular* combination of intensities, irrespective of the value of the relative phase (which is kept constant at $\alpha=0$).

The resonant-type behavior of the ionization rate appears to be a result of the "crossing" between the $|S=1,l=1,n_\omega-3,n_{3\omega}\rangle$ and $|S=1,l=1,n_\omega,n_{3\omega}-1\rangle$ dressed continuum states, where n_ω and $n_{3\omega}$ are the initial number of the ω and 3ω photons. As illustrated in Fig. 1, the $S=1$ peak serves as "doorway" state to all other ATI peaks. Hence the resonance between the above two states affects all other ATI peaks. The effect is more general than the present $1w$-$3w$ scheme, since it appears also in the $1w$-$2w$ case and even when the frequency of one photon is not an exact multiple of the frequency of the other.

In conclusion, we have shown that *extensive* integral and differential CC of ATI processes is attainable, provided a judicious choice of relative intensities is made. Most striking is the demonstration of directional separation, by as much as $\sim 50°$, of one ATI kinetic energy peak from another, and the diversion of the alignment away from the light polarization direction. Of great importance too is the demonstration of intensity control in which both partial and total integral ATI rates attain a maximum at a specific combination of light intensities.

This work was supported by the U.S.-Israel Binational Foundation and by the U.S. Office of Naval Research under Contract No. N00014-90-J-1014.

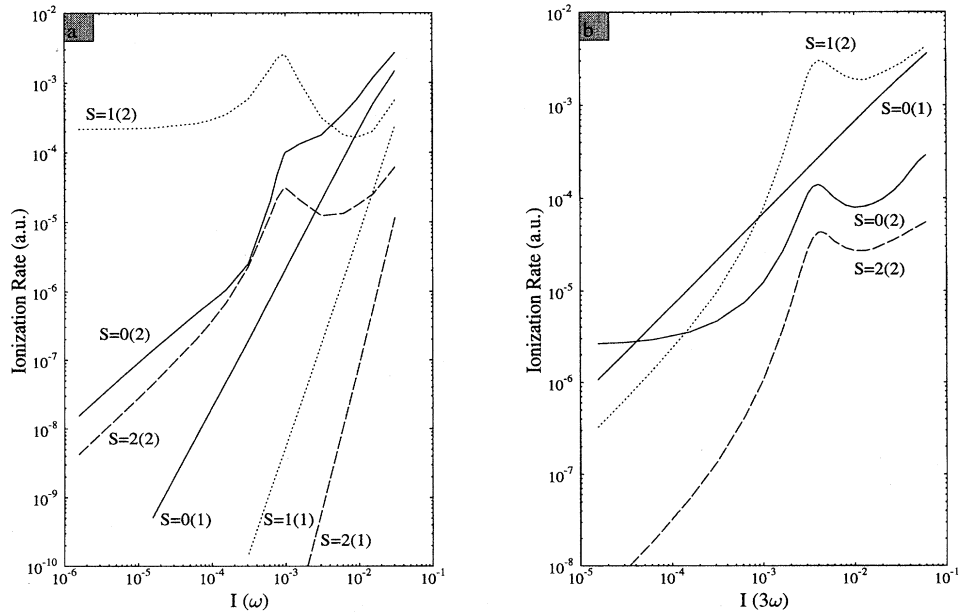

FIG. 4. Ionization rates of the first three ATI peaks for a fixed phase ($\alpha=0$) (intensity control) as a function of (a) $I(\omega)$, at a fixed value of $I(3\omega)$ ($=3.108\times10^{-3}$ a.u.) and (b) $I(3\omega)$, at a fixed value of $I(\omega)$ ($=1.10\times10^{-3}$ a.u.). Also plotted are the one-color results. (The number in parentheses distinguishes between the one- and two-color cases.)

[1] For a recent review, see, e.g., M. Shapiro and P. Brumer, in *International Reviews in Physical Chemistry* (Taylor & Francis, London, 1994), Vol. 13, p. 187.
[2] R. Trainham, G. H. Fletcher, N. B. Mansour, and D. J. Larson, Phys. Rev. Lett. **59**, 2291 (1987).
[3] D. Normand, L. A. Lompre, A. L'Huilier, J. Morellec, M. Ferray, J. Lanvancier, G. Mainfray, and C. Manus, J. Opt. Am. B **6**, 1513 (1989).
[4] H. G. Muller, P. H. Bucksbaum, D. W. Schumacher, and A. Zavriyev, J. Phys. B **23**, 2761 (1990).
[5] Y. Y. Yin, C. Chen, and D. S. Elliot, Phys. Rev. Lett. **69**, 2353 (1992).
[6] D. W. Schumacher, F. Weihe, H. G. Muller, and P. H. Bucksbaum, Phys. Rev. Lett. **73**, 1344 (1994).
[7] M. Dorr and R. Shakeshaft, Phys. Rev. A **40**, 459 (1989).
[8] A. Szoke, K. C. Kulander, and J. N. Bardsley, J. Phys. B **24**, 3165 (1991).
[9] R. M. Potvliege and P. H. G. Smith, J. Phys. B **24**, L641 (1991).
[10] R. M. Potvliege and P. H. G. Smith, J. Phys. B **25**, 2501 (1992).
[11] K. J. Schafer and K. C. Kulander, Phys. Rev. A **45**, 8026 (1992).
[12] T. Nakajima, P. Lambropoulus, S. Cavalieri, and M. Matera, Phys. Rev. A **46**, 7315 (1992).
[13] N. B. Baranova, H. R. Reiss, and B. Y. Zel'dovich, Phys. Rev. A **48**, 1497 (1993).
[14] M. Protopapas, P. L. Knight, and K. Burnett, Phys. Rev. A **49**, 1945 (1994).
[15] G. Kurizki, M. Shapiro, and P. Brumer, Phys. Rev. B **39**, 3435 (1989).
[16] B. A. Baranova, A. N. Chudinov, and B. Ya. Zel'dovitch, Opt. Commun. **79**, 116 (1990).
[17] M. Shapiro, J. W. Hepburn, and P. Brumer, Chem. Phys. Lett. **149**, 451 (1988).
[18] C. Chen, Y. Y. Yin, and S. S. Elliot, Phys. Rev. Lett. **64**, 507 (1990).
[19] S. M. Park, S-P. Lu, and R. J. Gordon, J. Chem. Phys. **94**, 8622 (1991); S-P. Lu, S. M. Park, Y. Xie, and R. J. Gordon, *ibid.* **96**, 6613 (1992).
[20] C. K. Chan, P. Brumer, and M. Shapiro, J. Chem. Phys. **94**, 2688 (1991).
[21] M. Shapiro, J. Chem. Phys. **56**, 2582 (1972).
[22] R. A. Blank and M. Shapiro, Phys. Rev. A **50**, 3234 (1994).
[23] R. A. Blank and M. Shapiro, Phys. Rev. A **51**, 4762 (1995).
[24] This is indeed the main justification for the use of 1D models in ATI, see Ref. [25], or strong field photodissociation see Ref. [26].
[25] Q. Su and J. H. Eberly, Laser Phys. **2**, 598 (1992).
[26] E. Charron, A. Giusti-Suzor, and F. H. Mies, Phys. Rev. Lett. **71**, 692 (1993); Phys. Rev. A **49**, R641 (1994).
[27] A recent one-color work [28] reports the observation of sidelobes in the angular distribution of some of the higher ATI peaks in xenon, though the majority of the electrons are still ejected in the polarization direction.
[28] B. Yang, K. J. Schafer, B. Walker, K. C. Kulander, P. Agostini, and L. F. DiMauro, Phys. Rev. Lett. **71**, 3770 (1993).