Modulating ionization through phase control

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We present a theory of modulating ionization by controlling the phases of incident laser fields. Specific calculations have been performed for the Na atom using the density-matrix equations. Significant modulation of the ion signal can be obtained by choosing the appropriate combinations of laser intensities and frequencies, in resonant as well as nonresonant processes.

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A recent paper Chen *et al.* [1] demonstrated the possibility of controlling the excitation (and hence subsequent ionization) of a bound state in Hg through the simultaneous coupling to the ground state by a single- and three-photon transition. Control is achieved by changing the relative phase of the two monochromatic beams thereby altering the degree of destructive interference between the two paths of excitation. Their experimental demonstration relied on the exploitation of a cancellation effect [2–10] in a resonant third-harmonic generation, which has been known for some time. A generalization of that effect to the case of a somewhat nonresonant three-photon excitation has been demonstrated most recently [11,12]. The possibility of controlling such transitions into the continuum [13,14] (ionization or dissociation) without changing the radiation intensity represents a prospect of both basic and applied interest. It is therefore desirable to establish possibilities of such control in as great a variety of schemes as possible. It would, in particular, be most useful if it were possible to induce such a cancellation directly into the continuum, so as to minimize restrictions on wavelength. The purpose of this paper is to demonstrate that this is indeed the case and to present specific proposals for its experimental implementation. The basic question then is whether, in simultaneous one- and three-photon ionization, we can modulate the amount of ionization by changing the relative phase of the two beams. If yes, how deep can the modulation be and how does its depth depend on the laser intensities and the atomic parameters. We consider an s initial state with a specific quantitative application to Na.

The first potential complication that comes to mind stems from the fact that a three-photon transition from an s state into the continuum leads to a final state that contains a p and an f partial wave, while the one-photon transition leads to only a p partial wave. Thus only part of the three-photon amplitude can interfere with the one-photon amplitude. If the f wave happens to dominate, any modulation would be undetectable. There are two ways out of this difficulty. One is to tune one of the wavelengths to a two-photon resonance with an excited s state from which ionization produces only a p wave. For maximum flexibility, one can introduce a third wavelength [as shown in Fig. 1(a)] and adjust it so as to achieve interference at the desired energy in the continuum. Proper choice of the relative intensities of the three beams will also be required as discussed below. It turns out that only two of the three phases enter in the interference. We examine also the more restrictive case of two wavelengths by taking ($\lambda_1=\lambda_2$).

The other way out of the difficulty is to abandon the two-photon resonance [Fig. 1(b)] but search for ranges in the continuum where the p wave dominates over the f wave. Such ranges do exist, owing to the oscillatory behavior of the bound-free matrix elements as functions of

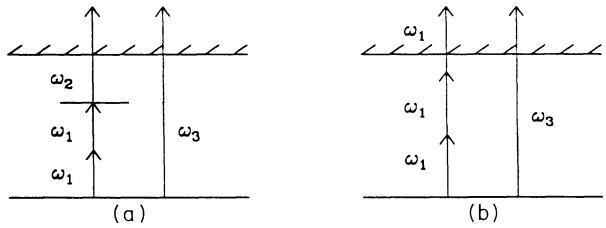

FIG. 1. (a) and (b) Schemes studied in this paper.

the photon energy. The case of completely nonresonant three-photon ionization can be studied in terms of a simple equation that includes the appropriate amplitudes added, squared, and integrated over the photoelectron angular distribution. The resonant case, however, requires a set of density-matrix equations which yield the simpler nonresonant equations as a special case. We present here the equations for the most general case of a two-photon resonance and three wavelengths.

Let $|0\rangle$ be the ground state $3s$ and $|1\rangle$ the excited state $5s$. We assume that the linearly polarized laser field has the following form:

$$E(t)=(\epsilon_1 e^{i\omega_1 t}+\epsilon_2 e^{i(\omega_2 t+\phi_2)}+\epsilon_3 e^{i(\omega_3 t+\phi_3)})+\text{c.c.}\ , \tag{1}$$

where the phases of the electric fields 2 and 3 are taken with respect to that of field 1 and assumed to be fixed and time independent. For the time being, we also assume that the electric fields ϵ_i ($i=1$, 2, and 3) vary in time according to a prescribed pulse shape (Gaussian, for example) without any stochastic fluctuations. We are, in other words, assuming here Fourier-limited pulses. A standard derivation leads to the following set of density-matrix equations:

$$\frac{\partial}{\partial t}\sigma_{00}=-\Gamma_0^{(3)}\sigma_{00}+\text{Im}\{[\Omega_{10}^{(-1,-1)}+e^{-i(\phi_2-\phi_3+\Delta\omega t)}(\Omega_{10}^{(2,-3)*}+\Omega_{10}^{(-3,2)})]\sigma_{10}\}\ , \tag{2}$$

$$\frac{\partial}{\partial t}\sigma_{11}=-(\Gamma_1^{(1)}+\Gamma_1^{(2)}+\Gamma_1^{(3)})\sigma_{11}-\text{Im}\{[\Omega_{10}^{(-1,-1)}+e^{-i(\phi_2-\phi_3+\Delta\omega t)}(\Omega_{10}^{(2,-3)}+\Omega_{10}^{(-3,2)})]\sigma_{10}\}\ , \tag{3}$$

$$\left[\frac{\partial}{\partial t}-i(D+S_{10})+\tfrac{1}{2}(\Gamma_0^{(3)}+\Gamma_1^{(1)}+\Gamma_1^{(2)}+\Gamma_1^{(3)})\right]\sigma_{10}=-\tfrac{1}{2}i[\Omega_{10}^{(-1,-1)}+e^{i(\phi_2-\phi_3+\Delta\omega t)}(\Omega_{10}^{(2,-3)*}+\Omega_{10}^{(-3,2)})]\sigma_{00}$$
$$+\tfrac{1}{2}i[\Omega_{10}^{(-1,-1)}+e^{i(\phi_2-\phi_3+\Delta\omega t)}(\Omega_{10}^{(2,-3)}+\Omega_{10}^{(-3,2)})]\sigma_{11}\ , \tag{4}$$

where $\Omega_{ij}^{(m,n)}$ is the two-photon Rabi frequency between $|i\rangle$ and $|j\rangle$ through the absorption of photon ω_m and ω_n ($-m$ means the emission of the photon ω_m), $\Gamma_i^{(j)}$ the ionization width from $|i\rangle$ by the absorption of photon ω_j, D the detuning defined by $D=2\omega_1-\tilde{\omega}_{10}$ ($\tilde{\omega}_{10}$ being the atomic energy difference between $|1\rangle$ and $|0\rangle$), $\Delta\omega=2\omega_1-(\omega_3-\omega_2)$, and S_{10} the relative ac Stark shift between $|1\rangle$ and $|0\rangle$ defined by $S_{10}=S_1-S_0$. However, it is most desirable to work with intensities at which the Stark shifts are negligible, which is indeed possible.

The amount of ionization as a function of time is given by

$$P(t)=1-\sigma_{00}(t)-\sigma_{11}(t)\ . \tag{5}$$

Certain conclusions can be drawn from these equations before performing any numerical calculations. First, it is only the phase difference $\phi_2-\phi_3$ that matters. There are two paths that couple $|1\rangle$ and $|0\rangle$. One is the coupling by the absorption of two ω_1. The other is the coupling that involves photon ω_2 and ω_3. The coupling via this process has the relative phase $\phi_2-\phi_3$, since $|0\rangle$ and $|1\rangle$ are coupled by either virtual absorption of photon ω_2 and virtual emission of photon ω_3 or vice versa. Second, if the intensity I_1 of the radiation at ω_1 is much stronger or weaker than $\sqrt{I_2 I_3}$, no modulation of the ion signal will be observed: If $I_1 \gg \sqrt{I_2 I_3}$, the coupling of the state $|1\rangle$ and $|0\rangle$ due to the photon ω_2 and ω_3 is negligible compared with that due to photon ω_1. Therefore the ion yield does not depend on $\phi_2-\phi_3$. If on the other hand, $I_1 \ll \sqrt{I_2 I_3}$, the coupling of the states $|1\rangle$ and $|0\rangle$ due to photon ω_1 is negligible. Then, by rewriting the above density-matrix equations with a redefined σ_{10}, one immediately sees again that the ion yield does not depend on the phase. This result is consistent with that of the perturbation limit: Without I_1, the phase should not matter, since the information of the phase will be lost when the absolute value of the transition amplitude is squared. On the basis of these considerations, it turns out that the coupling strength of the states $|1\rangle$ and $|0\rangle$ due to photon ω_1 needs to be comparable with that due to photons ω_2 and ω_3 to make the phase-dependent modulations observable.

We present now typical results illustrating the expected modulation under these conditions. The atomic parameters are calculated using single-channel quantum-defect theory (SQDT). We know from our experience that SQDT gives good atomic parameters for simple atoms such as sodium. Having obtained all of the necessary equations and parameters, we performed numerical calculations assuming Gaussian pulses of the same duration τ (full width at half maximum) for all three lasers.

For the three-color case, with the laser frequencies $\omega_1=16\,600\ \text{cm}^{-1}$, $\omega_2=10\,398\ \text{cm}^{-1}$, and $\omega_3=43\,598\ \text{cm}^{-1}$ corresponding to Fig. 2, the atomic parameters employed in the calculations reported here (in units of rad/sec for Ω and S and sec^{-1} for Γ with the laser intensities in W/cm^2) are $\Gamma_0^{(3)}=0.128I_3$, $\Gamma_1^{(1)}=0.156I_1$, $\Gamma_1^{(2)}=3.950I_2$, $\Gamma_1^{(3)}=0.015I_3$, $\Omega_{10}^{(-1,-1)}=472.0I_1$, $\text{Re}\Omega_{10}^{(2,-3)}=-2.845\sqrt{I_2 I_3}$, $\text{Im}\Omega_{10}^{(2,-3)}=0.711\sqrt{I_2 I_3}$, $\Omega_{10}^{(-3,2)}=7.675\sqrt{I_2 I_3}$, $S_0=-1110.9I_1-76.1I_2+8.9I_3$, and $S_1=4.8I_1+192.8I_2+7.4I_3$. For the two-color case, with the laser frequencies $\omega_1=16\,600\ \text{cm}^{-1}$ and $\omega_3=49\,800\ \text{cm}^{-1}$ corresponding to Fig. 3, the atomic parameters in the same units as above are $\Gamma_0^{(3)}=0.019I_3$, $\Gamma_1^{(1)}=0.156I_1$, $\Gamma_1^{(3)}=0.0137I_3$, $\Omega_{10}^{(-1,-1)}=472.0I_1$, $\text{Re}\Omega_{10}^{(1,-3)}=-4.791\sqrt{I_1 I_3}$, $\text{Im}\Omega_{10}^{(1,-3)}=0.0544\sqrt{I_1 I_3}$,

$\Omega_{10}^{(-3,1)}=6.302\sqrt{I_1 I_3}$, $S_0=-1110.9I_1+6.6I_3$, and $S_1=4.8I_1+5.7I_3$.

We now present and discuss selected numerical results. First, we show in Fig. 2(a) how the ion yield depends on the phase difference $\phi_2-\phi_3$. With that particular combination of I_1, I_2 and I_3, the modulation is significant (solid line). The maximum and the minimum of the ion yield Y differ by one order of magnitude. Figure 2(b) shows the variation of the depth of modulation $[d=(Y_{\max}-Y_{\min})/\frac{1}{2}(Y_{\max}+Y_{\min})]$ as a function of I_1. At the weak and the high intensity limits, the modulation becomes zero as discussed earlier. The depth changes quite dramatically near the peak. This three-color case is, of course, more demanding experimentally in that it requires three beams with fixed phase relations. In principle this should be possible through a combination of wave-mixing processes.

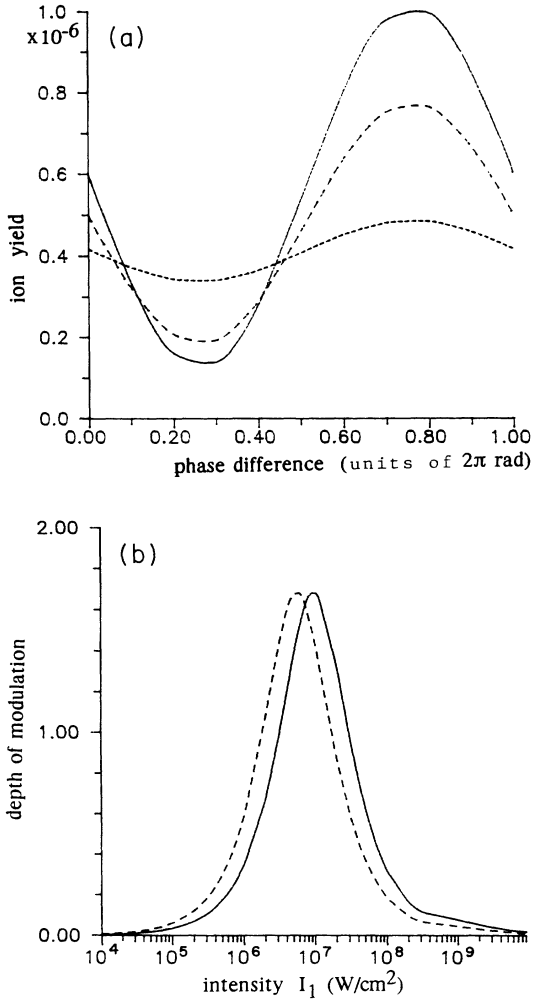

FIG. 2. (a) Variation of the ion yield as a function of the phase difference. $\tau=10$ ps, $I_2=5\times10^7$ W/cm^2, and $I_3=3\times10^5$ W/cm^2. $I_1=6\times10^6$ W/cm^2 (solid line) or 4×10^6 W/cm^2 (long-dashed) or 10^6 W/cm^2 (short-dashed). Atomic parameters are given in the text. (b) Change of the depth (defined in the text) as a function of the intensity I_1 for the fixed $I_2=5\times10^7$ W/cm^2. $I_3=3\times10^5$ W/cm^2 (solid). $I_3=10^5$ W/cm^2 (dashed). $\tau=10$ ps. Atomic parameters are the same used for (a).

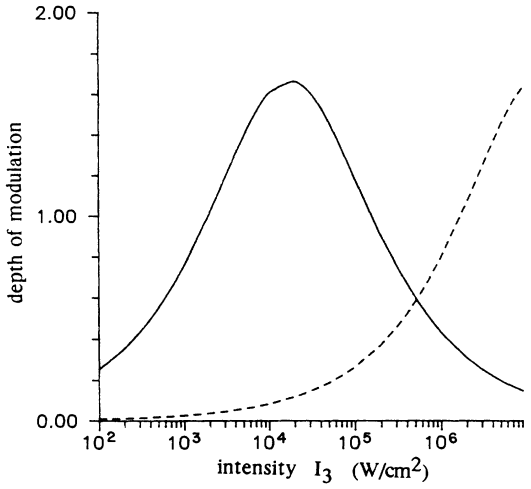

FIG. 3. Variation of the depth as the function of the intensity I_3 for the two-color case. $I_1=10^7$ W/cm^2 (solid line) and $I_1=10^8$ W/cm^2 (dashed line); $\tau=10$ ps. Atomic parameters are given in the text.

The results for the two-color case are shown in Fig. 3. This scheme may be more desirable from the experimental point of view since the ω_3 photon can be generated using the third harmonic of ω_1. We see that the modulation obtained here is as significant as in the three-color case. The reason for which the curves of Fig. 3 are broader than those of Fig. 2(b) is that the horizontal scale is chosen with respect to the intensity I_3. Finally, we show examples for nonresonant ionization [Fig. 4(a)]. The atomic parameters vary as a function of the laser frequencies, and the averaged ion yield $[\frac{1}{2}(Y_{\max}+Y_{\min})]$ is given in Fig. 4(b) as a function of laser frequencies. It turns out, as anticipated earlier, that significant modulation can be obtained here as well by choosing the appropriate laser frequencies.

In summary, we have shown that significant modulation of the ionization yield can be obtained by changing the relative phase difference of three as well as two laser fields. To maximize the depth of modulation, appropriate combinations of laser intensities need to be chosen. Resonant as well as nonresonant processes can exhibit this effect, although with different depths of modulation. The ideas and formalism presented here can be readily applied to more complex systems where more transition channels are involved. In fact it will be more interesting in that case. But even in simple systems such as Na, this effect can in the simplest case of a dominant p wave be employed for a measurement of a three-photon transition relative to a single-photon transition. In the simultaneous presence of an f wave, we can envision canceling the p wave, providing thus a measurement of the f wave contribution. Of course the latter can be achieved through a three-photon transition by one beam of circular polarization, but redundant measurements are always valuable in reducing errors. The case of resonance lends itself to interesting generalizations. It provides, for example, a con-

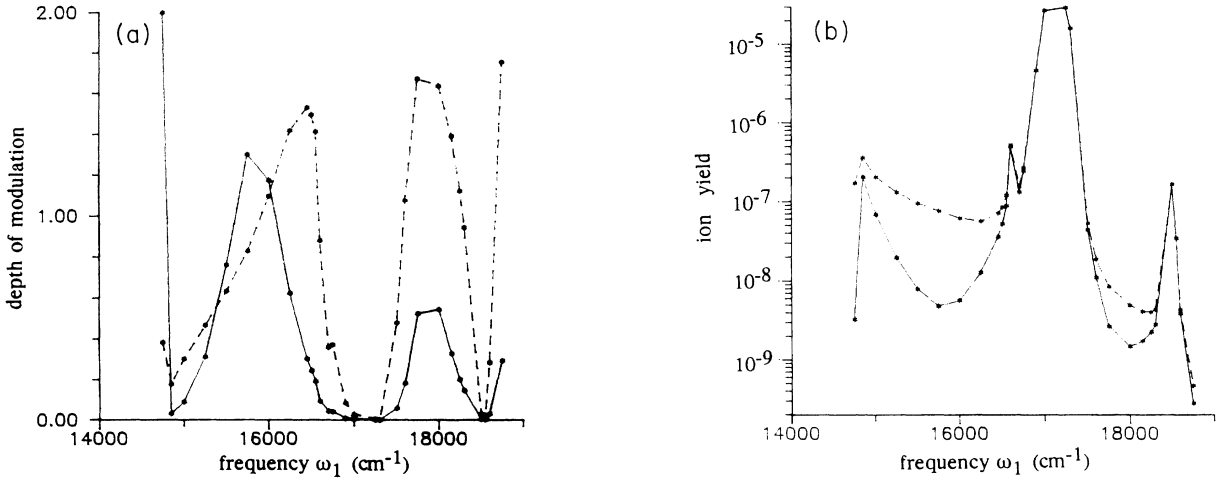

FIG. 4. (a) Change of the depth of modulation as a function of the laser frequency ω_1. ω_3 is always kept as $\omega_3=3\omega_1$. $I_1=10^8$ W/cm^2, $I_3=10$ W/cm^2 (solid line); $I_1=10^8$ W/cm^2, $I_3=10^3$ W/cm^2 (dashed line). $\tau=5$ ns. Calculated data points are connected as a guide to the eyes. (b) Variation of the ion yield corresponding to (a). All the atomic parameters are the same with those employed for (a). Clearly, the same depth of modulation can be obtained with higher ion yield by increasing the intensities of both lasers.

venient context for the influence of fluctuations of the laser fields on the whole process as manifested in the modification of the depth of modulation. We will report soon on this effect. Collisional dephasing effects can also be studied through observation of the depth of modulation.

After completion of this work, we became aware of recent papers by Potvliege and Smith [15] and by Schafer and Kulander [16]. Although the general topic is the same, the context is quite different. These authors have emphasized the high-intensity aspects of the problem while we are explicitly interested in low intensities so that we can carefully control the outcome and trace its physical origin.

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