Coherent quantum control of multiphoton transitions by shaped ultrashort optical pulses

Doron Meshulach and Yaron Silberberg*
Department of Physics of Complex Systems, Weizmann Institute of Science, Rehovot 76100, Israel
(Received 1 October 1998)

Multiphoton transitions can be reached by many routes through a continuum of virtual levels. We show that the transition probability of two-photon and multiphoton processes can be controlled by tailoring the shape of an ultrashort excitation pulse, so that the many paths leading to the final state interfere to give a desired probability amplitude. We analyze the effect of pulse shapes on *N*-photon absorption as well as on Raman transitions. We show theoretically that certain tailored *dark pulses* do not excite the system at all, while other shaped pulses induce transitions as effectively as transform limited pulses, even when their peak amplitudes are greatly reduced. These results are confirmed experimentally for two-photon transitions in atomic cesium. [S1050-2947(99)00808-2]

PACS number(s): 32.80.Qk, 32.80.Wr, 42.65.Re

I. INTRODUCTION

In coherent quantum control, the goal is to steer a quantum system towards a desired final state through its interaction with light, while canceling out other possible paths leading to undesirable outcomes [1,2]. This can be achieved through quantum interference between the various trajectories leading to the same final state. The underlying principle in most schemes is to control quantum interactions by manipulating the coherence properties of the optical field.

While schemes of coherent control may involve excitations by continuous waves, many studies involved ultrashort optical pulses. For example, coherent control of molecular systems with a sequence of short pulses could allow selective manipulation of molecular structures [3], including breaking specific bonds and changing a reaction path. Most of these schemes involve exciting the system with short pulse pairs [4–9], and control of the interactions is achieved by varying the time delay between the two pulses. Such control by pulse-pairs excitations was also demonstrated for various one-photon [10,11] and two-photon transitions in two-level systems [12,13].

With the recent progress in ultrafast optics, it is now possible to shape ultrashort signals with almost arbitrary temporal shapes [14,15]. These shaped signals are generated from laser pulses through manipulation of the spectral phases and amplitudes of the frequency components of these pulses. With the ability to shape such pulses with high fidelity, excitations should not be limited to pulse pairs or simple pulse sequences, and it is natural to ask, then, what degree of control can be achieved by exciting atoms and molecules by such complex-shaped pulses. As explained below, one-photon transitions with ultrashort pulses offer only very limited control. However, we have recently shown [16] that it is possible to control a nonresonant two-photon transition (i.e., a transition with no intermediate state) in a two-level system, even to cancel it completely, by tailoring the spectral phase, and hence the shape of an ultrashort pulse. We predicted and demonstrated experimentally the existence of so-called *dark pulses* that induce no net transitions. Furthermore, we demonstrated that tailored long pulses can induce two-photon transitions as effectively as transform-limited pulses with the same power spectrum and energy.

In this work, we show that coherent control through spectral phase manipulation of ultrashort pulses can be applied to *any* narrow transition line nonresonant multiphoton processes, including multiphoton absorption and Raman-type processes, some of which are shown in Fig. 1. We approximate, using a simple theoretical model, the dependance of these coherent processes on the shape and the spectral phase of the excitation pulse, and present experimental results demonstrating the effectiveness of this coherent control scheme. To emphasize the role of coherence, we limit the discussion here to ultrashort pulses that are shaped by phase-only spectral filters. This ensures that the power spectrum and the energy of the shaped excitation pulse are left intact, unlike in coherent control schemes involving pulse-pair excitations, where varying the delay between the pulses modifies their power spectrum. Finally, we then compare our results with the other limit of a very broad inhomogeneous absorption line, where two-photon and multiphoton transitions depend on the intensity of the excitation pulse, and not on its phase.

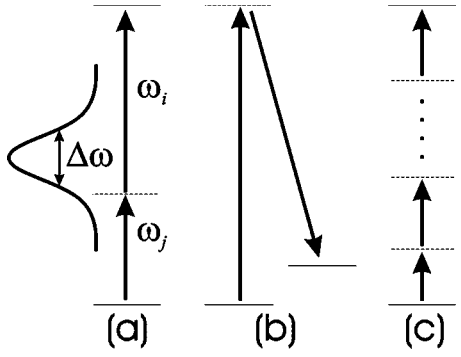

FIG. 1. Schematic energy-level diagrams of nonresonant multiphoton processes. (a) Two-photon absorption, with a schematic representation of the excitation spectrum. (b) Stimulated Raman transition. (c) Multiphoton absorption.

*Electronic address: feyaron@wis.weizmann.ac.il

II. THEORY

Consider the resonant interaction of a weak ultrashort pulse with a two-level atom. Let $|g\rangle$ and $|f\rangle$ with energies E_g and E_f be the ground and excited states, respectively. The electric field of the excitation pulse is $\epsilon(t)$, which is assumed to be much shorter than the lifetime of the excited state. Assuming the atom is initially in the ground state, first-order time-dependent perturbation theory predicts for the amplitude of the excited state [17]

$$a_f(t)=\frac{\mu_{fg}}{i\hbar}\int_{-\infty}^{t}\epsilon(t_1)\exp(i\omega_0 t_1)dt_1, \tag{1}$$

with μ_{fg} the dipole moment matrix element and $\omega_0=(E_f-E_g)/\hbar$. For times after the end of the excitation pulse, this integral is just the Fourier component of the optical field at resonance. The one-photon transition probability to the excited state depends, therefore, only on the energy content of the frequency component of the excitation pulse which is resonant with the transition. Neither the phases nor the amplitudes of all other spectral components of the pulse affect the transition probability.

If the pulse is not resonant with any one-photon transition, it may still induce two-photon transitions. Second-order time-dependent perturbation theory leads to

$$a_f(t)=-\frac{1}{\hbar^2}\sum_n \mu_{fn}\mu_{ng}\int_{-\infty}^{t}\int_{-\infty}^{t_1}\epsilon(t_1)\epsilon(t_2) \times\exp(i\omega_{fn}t_1)\exp(i\omega_{ng}t_2)dt_2dt_1, \tag{2}$$

where $\omega_{ij}=(E_i-E_j)/\hbar$, and the summation is performed over all possible intermediate states of the unperturbed atom. Most standard treatments of two-photon and multiphoton transitions (see, for example, Ref. [18]) assume continuous excitations, and the integration is performed explicitly. In the case of a short pulse excitation, it is instructive to examine first the summation over the intermediate levels. Since we assume they are all far from resonance, the contribution of all intermediate levels will add coherently only for a very short duration, hence we approximate

$$\sum \mu_{fn}\mu_{ng}\exp[iE_n(t_2-t_1)/\hbar] \simeq\begin{cases}\langle f|\mu^2|g\rangle, & |t_1-t_2|<\bar{\omega}^{-1}\\ 0, & |t_{1-}-t_2|\geqslant\bar{\omega}^{-1},\end{cases} \tag{3}$$

where $\hbar\bar{\omega}$ is an appropriately weighted average energy, similar to the derivation of Bebb and Gold [19]. This form has the advantage of limiting the role of the intermediate levels, which now merely define the lifetime of the virtual levels. Using Eqs. (2) and (3) we obtain for $|a_f|^2$ the two-photon transition probability to the excited level following the pulse excitation

$$P_{g\to f}^{(2-PH)}=\frac{1}{\hbar^4}\left|\frac{\langle f|\mu^2|g\rangle}{\bar{\omega}}\right|^2\left|\int_{-\infty}^{\infty}\epsilon^2(t)\exp(i\omega_0 t)dt\right|^2. \tag{4}$$

In a similar fashion, if all lower-order processes are nonresonant as in Fig. 1(c), the probability for an N-photon transition to the excited level following the pulse excitation is proportional to the resonant Fourier component of $\epsilon^N(t)$:

$$P_{g\to f}^{(N-PH)}\sim\left|\int_{-\infty}^{\infty}\epsilon^N(t)\exp(i\omega_0 t)dt\right|^2. \tag{5}$$

Obviously, the power spectrum of $\epsilon^N(t)$ can be controlled by modifying the power spectrum of $\epsilon(t)$. It is less obvious, however, that the power spectrum of $\epsilon^N(t)$ can also be controlled by manipulations of the spectral phase of the exciting pulse $\epsilon(t)$, affecting neither the power spectrum nor the energy of the pulse. As we show below, it is possible to modify the spectral phase of the exciting pulse, thereby to control the N-photon transition probability, even to cancel it completely. It is important to note that the above derivation is rather crude, and several strong-field effects, such as ac Stark shifts, are not included. Yet, the general form of these results, and in particular the $\epsilon^N(t)$ dependence, is expected to hold even in more complete analysis.

We shall first concentrate on two-photon transitions. Assuming that the excitation pulse $\epsilon(t)$ is produced by a short pulse containing a band of frequencies around a central frequency ω_l, then $\epsilon^2(t)$ can be decomposed into three bands of frequencies, two around $\pm 2\omega_l$ and one near 0. Accordingly, a two-photon transition as predicted by Eq. (4) can describe two-photon absorption (TPA, $2\omega_l\simeq\omega_0$ [Fig. 1(a)], as well as Raman transitions $\omega_l\gg\omega_0$ [Fig. 1(b)]). Note that now, in contrast to a single-photon transition, all frequency components of the pulse may contribute to the transition. To make this point more explicit, we rewrite the integral in Eq. (4) in the frequency domain for TPA:

$$\begin{aligned}S_2&=\left|\int_{-\infty}^{\infty}\epsilon^2(t)\exp(i\omega_0 t)dt\right|^2\\ &=\left|\int_{-\infty}^{\infty}\tilde{\epsilon}(\omega_0/2+\Omega)\tilde{\epsilon}(\omega_0/2-\Omega)d\Omega\right|^2\\ &=\left|\int_{-\infty}^{\infty}A(\omega_0/2+\Omega)A(\omega_0/2-\Omega)\right.\\ &\quad\left.\times\exp[i\{\Phi(\omega_0/2+\Omega)+\Phi(\omega_0/2-\Omega)\}]d\Omega\right|^2,\end{aligned} \tag{6}$$

where $\tilde{\epsilon}(\omega)=A(\omega)\exp[i\Phi(\omega)]$ is the Fourier transform of $\epsilon(t)$, and $A(\omega)$ and $\Phi(\omega)$ are the spectral amplitude and the spectral phase, respectively. Note that since $\epsilon(t)$ is real, $\tilde{\epsilon}(\omega)=\tilde{\epsilon}^*(-\omega)$. Equation (6) reflects the fact that two-photon transitions occur for all pairs of photons with frequencies ω_i and ω_j, with $\omega_i+\omega_j=\omega_0$, and ω_i and ω_j lie within the spectrum of the exciting pulse, as shown schematically in Fig. 1(a). The two-photon transition probability can therefore be controlled by tailoring the spectral phase of a single pulse.

It is easy to verify that S_2, and hence the two-photon transition probability is maximized, for a given energy and power spectrum $A^2(\omega)$, by the transform limited pulse, i.e., the pulse having the minimum time duration [$\Phi(\Omega)=0$].

Consider a pulse with the same energy and power spectrum, but having any *antisymmetric* spectral phase distribution around the two-photon transition frequency $\omega_0/2$, i.e., $\Phi(\omega_0/2+\Omega)=-\Phi(\omega_0/2-\Omega)$. In this case, the phase terms in Eq. (6) cancel each other, so that the two-photon transition probability is independent of the spectral phase, and is identical with that of the transform limited pulse. However, this antisymmetric spectral phase significantly affects the pulse shape. Essentially, the pulse can be spread in time into a very small amplitude signal without affecting the two-photon transition probability (as long as it does not exceed the lifetime of the excited state). This result is somewhat counterintuitive, as it might be expected that for a given energy and power spectrum, shorter pulses with higher peak intensities improve two-photon excitations.

While antisymmetric phase distributions do not affect the two-photon transition probability, other phase distributions can be tailored to eliminate multiphoton transitions. The existence of such pulses can be given a simple explanation [20] by considering the power spectrum of $\epsilon^N(t)$. This power spectrum can be tailored to vanish at certain frequencies, leading to the elimination of transitions at those frequencies. Such dark pulses that induce no net multiphoton transitions are the appropriate coherent superposition of optical frequencies that cancels TPA [16] and multiphoton absorption. They are analogous to dark states, which are coherent superpositions of quantum states that do not absorb resonant light. The role of symmetry of the spectral phase was discussed in Ref. [16], where the effect of periodic spectral phases on TPA was demonstrated. Here we investigate the effect of a particularly simple spectral phase, namely a π phase step, on TPA and other multiphoton processes. We emphasize the role of coherence by considering also the limit of wide TPA resonances.

Consider, first, an initial sech-like pulse with a central frequency ω_l that is tuned for an N-photon absorption, i.e., $\omega_l=\omega_0/N$. The pulse is now modified by a π spectral phase step, shifted by δ from the central frequency ω_l, which is introduced into the spectrum of the pulse. The excitation spectrum is then

$$\tilde{\epsilon}(\omega_l+\Omega)=\mathrm{sech}\left(\frac{1.76\Omega}{\Delta\omega}\right)\exp\left[i\frac{\pi}{2}\mathrm{sgn}(\Omega-\delta)\right], \qquad (7)$$

where $\Delta\omega$ is the full width at half maximum (FWHM) bandwidth of the power spectrum, and $\mathrm{sgn}(x)=\pm 1$, according to the sign of x. The spectral phase step modifies the temporal shape of the pulse, producing double-humped pulses, the precise shapes of which depend on δ. In Fig. 2 we present the calculated values of $S_N=\left|\int_{-\infty}^{\infty}\epsilon^N(t)\exp(i\omega_0 t)dt\right|^2$ for $N=2,3,4$ as a function of the normalized phase-step position $\delta/\Delta\omega$. It is clear that when $|\delta|\gg\Delta\omega$, the phase step should have no effect on the interaction, and indeed all curves approach the value obtained for the transform-limited pulses for large values of $\pm\delta/\Delta\omega$. Higher-order processes, however, are sensitive to phase steps that are further out in the wings of the spectrum, and the sensitivity to the phase-step locations scales with N. When $\delta=0$, the phase structure is antisymmetric, hence it should not affect TPA, but it does affect higher-order processes. Interestingly, each of the curves vanishes exactly for N values of $\delta/\Delta\omega$; these are dark

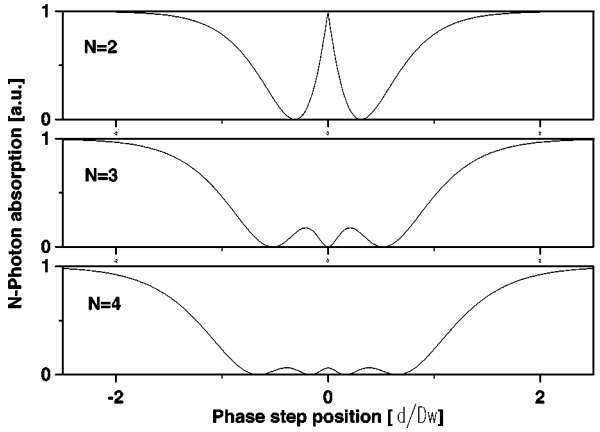

FIG. 2. Calculated results for coherent quantum control of N-photon absorption in a two-level system. Transition probability for excitation by a pulse with a π spectral phase step is shown as a function of the normalized step position $\delta/\Delta\omega$. The transition probabilities are normalized to the probability of excitation by a transform limited pulse.

pulses that induce no net transitions. It is also obvious from the trend in Fig. 2 that a π phase step reduces high-order multiphoton transitions over a very wide band. Since dark pulses for an N-photon transition are characterized by a null at the transition frequency in the power spectrum of $\epsilon^N(t)$, it is obvious that other dark pulses exist for other phase modulations.

Let us consider now the case of Raman transitions as in Fig. 1(b). For Raman transitions to be excited by a single pulse, the Raman frequency ω_0 must be within the bandwidth of the pulse. This form of excitation is known as impulsive Raman scattering. In this case the transition amplitude is

$$\int_{-\infty}^{\infty}\epsilon^2(t)\exp(i\omega_0 t)dt=\int_{-\infty}^{\infty}\tilde{\epsilon}(\Omega)\tilde{\epsilon}^*(\Omega-\omega_0)d\Omega, \qquad (8)$$

hence the Raman transition is excited by all pairs of frequencies which are separated by the Raman frequency. It can be shown that their integrated contribution is proportional to $\int_{-\infty}^{\infty}I(t)\exp(i\omega_0 t)dt$, which is simply the resonant frequency component of the intensity $I(t)$. Again, a transform-limited pulse gives the most effective excitation. Most spectral phase perturbations will attenuate the Raman yield, however periodic spectral phases with periodicity ω_0 will not. Such periodic phase structures split an initial transform-limited pulse into a pulse sequence with a repetition rate of $\omega_0/2\pi$. Such a pulse sequence can be used to narrow down the response of the impulsive Raman excitation, i.e., to cancel all but a desired transition. The effectiveness of pulse sequences exciting vibrational levels was demonstrated by Weiner *et al.* [21]. Our analysis above shows that the same principle should hold to any Raman-like transition.

Finally, it is instructive to consider the other limit of a very broad inhomogeneous absorption line. The TPF signal is then proportional to a sum of many individual transitions

$$S_2=\int_{-\infty}^{\infty}g(\omega_0)\left|\int_{-\infty}^{\infty}\epsilon^2(t)\exp(i\omega_0 t)dt\right|^2 d\omega_0, \qquad (9)$$

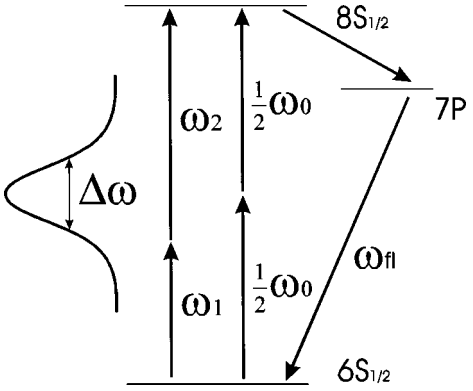

FIG. 3. Schematic diagram of the energy levels of the $6S_{1/2}$-$8S_{1/2}$ two-photon transition in atomic Cs. The two-photon transition energy $\hbar\omega_0$ corresponds to 411 nm. The excited atoms spontaneously decay to the ground level through the $7P$ level, so that the two-photon transitions can be directly observed through the measurement of the fluorescence signal ω_{fl} at $\sim$460 nm. The $6S_{1/2}$ and the $8S_{1/2}$ levels are split into two hyperfine states, but only two transitions are allowed, one from each of the sublevels of the ground state to the corresponding excited state. In the experiment all signals were shorter than a few picoseconds, too short to observe any dynamics between the two transitions corresponding to the two sublevels. We therefore considered the ground and excited states as single states.

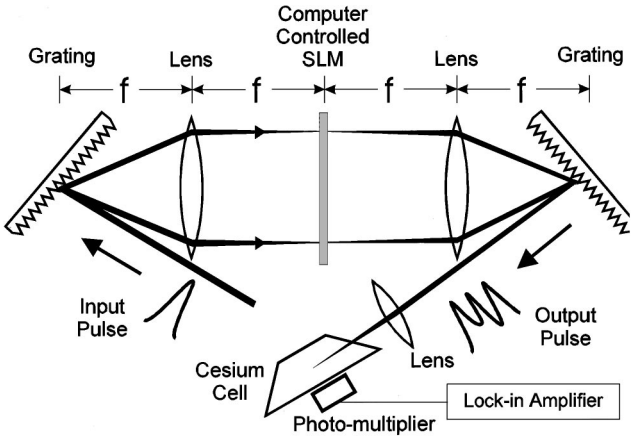

FIG. 4. Experimental arrangement for two-photon transitions in Cs gas. The programmable 4-f pulse shaper was composed of a pair of diffraction gratings with 1200 lines/mm, and a pair of achromat lenses with a 100-mm focal length. A programmable one-dimensional SLM with 128 computer controlled discrete elements was placed at the Fourier plane of the shaper, and was used as a dynamic filter for spectral phase manipulation of the pulses. The shaped output pulses were focused using a lens with a 50 mm focal length into the Cs gas cell. The fluorescence signal at $\sim$460 nm was detected by the photomultiplier and the lock-in amplifier.

where $g(\omega)$ is the line-shape function. Assuming a wide line limit, we have

$$S_2=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\epsilon^2(t_1)\epsilon^{*2}(t_2)\exp[i\omega_0(t_1-t_2)]dt_1dt_2d\omega, \tag{10}$$

which can be shown to yield

$$S_2=\int_{-\infty}^{\infty}I^2(t)dt. \tag{11}$$

Similarly, in this case for N-photon transitions, $S_N=\int_{-\infty}^{\infty}I^N(t)dt$. These well-known results simply reflect the fact that here TPA and multiphoton transitions depend on the intensity of the exciting pulses and not on their phase as in Eq. (6).

III. EXPERIMENTAL RESULTS

To demonstrate coherent control with this approach, we performed experiments with the $6S_{1/2}$-$8S_{1/2}$ two-photon transitions of atomic cesium, at a wavelength of 411 nm, induced by femtosecond pulses centered at 822 nm, as shown schematically in Fig. 3. Each of the excited atoms decays spontaneously to the ground level through the $7P$ level, so that the two-photon transition can be directly monitored through the measurement of the two-photon fluorescence (TPF) signal at 460 nm.

The experimental system, shown in Fig. 4, was similar to that described in Ref. [16]. The setup was composed of a programmable 4-f pulse shaper [14], a Cs gas cell, a photomultiplier, and a lock-in amplifier. A mode-locked Ti:sapphire laser produced 31 fs FWHM transform-limited sech^2 intensity pulses, centered at 822 nm, measured at the output of the pulse shaper. The programmable 4-f pulse shaper was composed of a pair of diffraction gratings with 1200 lines/mm and a pair of achromat lenses with a 100 mm focal length. The first lens and grating spatially map the complex spectrum of the input pulse at the Fourier plane, where a spatial filter was inserted. The second lens and grating reassemble the spectral components to form a modified time-shaped pulse. A programmable one-dimensional liquid-crystal spatial light modulator (SLM) array (SLM-256, CRI), composed of computer-controlled 128 discrete elements, was placed at the Fourier plane of the shaper, and was used as a dynamic filter for spectral phase manipulation of the pulses. The width of each pixel is 97 μm and the interpixel gap is 3 μm. The shaped output pulses were focused using a lens with a 50 mm focal length into the Cs gas cell, and the fluorescence signal at $\sim$460 nm was detected by the photomultiplier (1P28, Hamamatsu) and a lock-in amplifier.

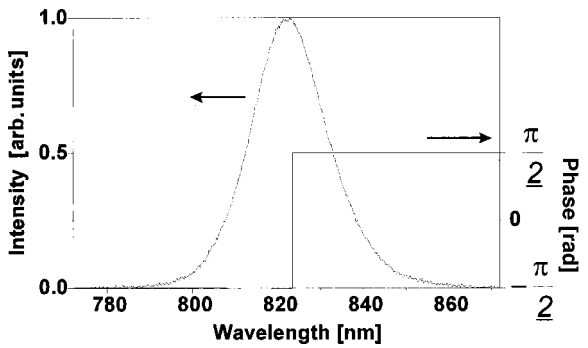

FIG. 5. A π spectral phase step with $\delta=0$ imposed upon the spectral phase input pulses, and the power spectrum of the input pulse.

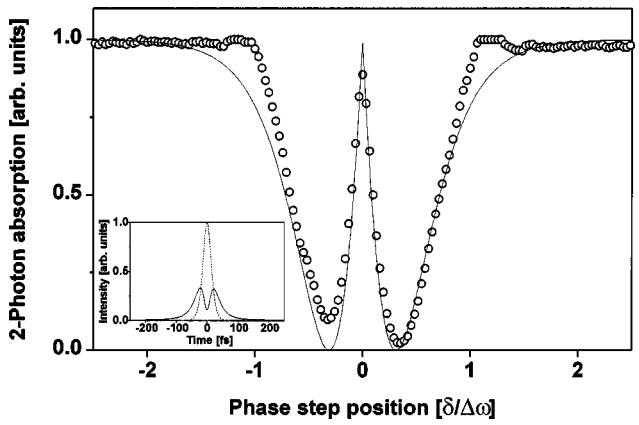

FIG. 6. Experimental (circles) and calculated results (solid) for two-photon transitions in Cs gas excited by a pulse with a π spectral phase step, as a function of the normalized step position $\delta/\Delta\omega$, with $\Delta\omega$ corresponding to $\Delta\lambda=23$ nm. Calculated results were obtained assuming transform-limited 31 fs sech^2 intensity input pulses. Inset: calculated temporal intensity distribution of the dark pulse corresponding to $\delta/\Delta\omega=0.31$ (solid) and the transform-limited input pulse having the same power spectrum (dashed).

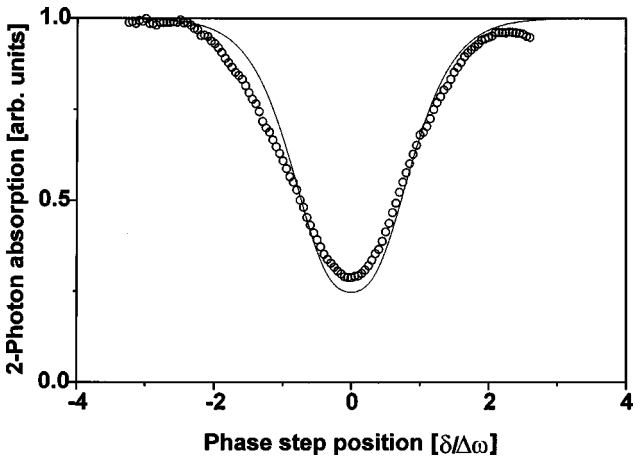

FIG. 7. Experimental (circles) and calculated results (solid) for two-photon transitions in a laser dye (Coumarin 6H) excited by a pulse with a π spectral phase step, as a function of the normalized step position $\delta/\Delta\omega$, with $\Delta\omega$ corresponding to $\Delta\lambda=14$ nm. Calculated results were obtained for transform-limited 50 fs sech^2 intensity input pulses.

We applied the appropriate voltages to the SLM to induce a π spectral phase step in the spectrum, as shown in Fig. 5. We then measured the TPF signal as a function of the step position δ. The experimental results are presented in Fig. 6, together with the theoretical curve calculated from Eq. (6). As expected, we note a constant TPF signal for large values of $\pm\delta/\Delta\omega$. The TPF almost vanishes at $\delta/\Delta\omega=\pm0.31$, corresponding to dark pulses. The calculated temporal distribution of these dark pulses is shown in Fig. 6 (inset), together with the transform-limited input pulse having the same energy and power spectrum. In contrast with our earlier results with periodic spectral phases, where the dark pulses were composed of sequences of short pulses [16], this dark pulse is a single burst of optical field. Note that in Fig. 6 the TPF induced by the pulse with an antisymmetric spectral phase distribution with $\delta=0$ almost reproduced the TPF for a transform limited pulse, as predicted by Eq. (6).

The results presented above were derived in the narrow line limit, where the absorption line is much narrower than the excitation spectrum. To emphasize the role of the spectral phase, we considered also the other limit of a very broad inhomogeneous line, where we recall that the two-photon transition probability is proportional to $S_2=\int_{-\infty}^{\infty}I^2(t)dt$. To demonstrate this experimentally, we replaced the Cs gas cell by a dye cell (Coumarin 6H in ethanol). The maximum of the absorption band is at 396 nm, and its FWHM is $\sim$60 nm. We tuned our laser to produce 50 fs FWHM transform-limited sech^2 intensity pulses, centered at 814 nm with a FWHM of 14 nm, and measured the TPF signal as a function of δ. The experimental results are presented in Fig. 7, together with the theoretical curve. Note that the effect of the broad line is to cancel out the coherent feature at $\delta=0$, which characterizes the narrow line limit (Fig. 6). These results can be explained in the time domain, noting that a π spectral phase step gives rise to a double-humped pulse, with a symmetrical distribution for $\delta=0$.

IV. CONCLUSIONS

In conclusion, we have shown that coherent quantum control of two-photon and multiphoton transitions can be effectively accomplished by tailoring the shape of the exciting ultrashort pulse. Since the pulse shape is controlled through phase manipulation of its spectrum, we investigated theoretically the effect of a particularly simple spectral phase modulation, namely a phase step, on two-photon and multiphoton absorption. We predicted the existence of dark pulses which induce no transitions; on the other hand, we show that certain spectral phase modulation leads to long pulses that induce two-photon transitions as effectively as transform-limited pulses with the same energy and power spectrum. These principles hold for multiphoton absorption including other multiphoton processes such as Raman transitions. We demonstrated experimentally these finding with two-photon absorption in cesium gas. Finally, to emphasize the role of coherence, we reported experiments with organic dye with broad energy levels, in which these coherent features are absent. The basic principles presented here open a wide new area for theoretical and experimental work, as well as possible applications in nonlinear spectroscopy and in atomic and molecular physics.

ACKNOWLEDGMENTS

We thank Professor C. Cohen-Tannoudji, Dr. N. Davidson, Professor T. W. Hänsch, Professor D. Tannor, and Professor M. Shapiro for helpful discussions, and D. Yelin for his help in developing pulse-shaping techniques.

[1] S. A. Rice, Science **258**, 412 (1992); P. Brumer and M. Shapiro, Sci. Am. **272**, 34 (1995).
[2] W. S. Warren, H. Rabitz, and M. Dahleh, Science **259**, 1581 (1993).
[3] D. J. Tannor, in *Molecules in Laser Fields*, edited by A. Bandrauk (Dekker, New York, 1994), pp. 403–446.
[4] D. J. Tannor and S. A. Rice, J. Chem. Phys. **83**, 5013 (1985).
[5] D. J. Tannor, R. Kosloff, and S. A. Rice, J. Chem. Phys. **85**, 5805 (1986).
[6] M. Shapiro and P. Brumer, J. Chem. Phys. **84**, 4103 (1986).
[7] D. J. Tannor and S. A. Rice, Adv. Chem. Phys. **70**, 441 (1988).
[8] P. Brumer and M. Shapiro, Annu. Rev. Phys. Chem. **43**, 257 (1992).
[9] E. Potter, J. L. Herek, S. Pedersen, Q. Liu, and A. H. Zewail, Nature (London) **355**, 66 (1992).
[10] J. T. Fourkas, W. L. Wilson, G. Wäckerle, A. E. Frost, and M. D. Fayer, J. Opt. Soc. Am. B **6**, 1905 (1989).
[11] A. P. Heberle, J. J. Baumberg, and K. Köhler, Phys. Rev. Lett. **75**, 2598 (1995).
[12] V. Blanchet, C. Nicole, M. Bouchene, and B. Girard, Phys. Rev. Lett. **78**, 2716 (1997).
[13] M. Bellini, A. Bartoli, and T. W. Hänsch, Opt. Lett. **22**, 540 (1997).
[14] A. M. Weiner, Prog. Quantum Electron. **19**, 161 (1995).
[15] C. W. Hillegas, J. X. Tull, D. Goswami, D. Strickland, and W. S. Warren, Opt. Lett. **19**, 737 (1994).
[16] D. Meshulach and Y. Silberberg, Nature (London) **396**, 239 (1998).
[17] M. Shapiro, J. Phys. Chem. **97**, 7396 (1993).
[18] F. H. M. Faisal, *Theory of Multiphoton Processes* (Plenum, New York, 1987), Chap. 2.
[19] H. H. Bebb and A. Gold, Phys. Rev. **143**, 1 (1966).
[20] P. H. Bucksbaum, Nature (London) **396**, 217 (1998).
[21] A. M. Weiner, D. E. Leaird, G. P. Wiederrecht, and K. A. Nelson, Science **247**, 1317 (1990).