Simple Route to Strong-Field Coherent Control

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Coherent-control schemes to manipulate weak-field interactions are generally invalid at stronger fields, since strong-field interactions are accompanied by level power broadenings and level shifts that usually elude simple analytical treatments. Here we show that a broad subgroup of weak-field solutions (those with real fields, i.e., fields with only one quadrature in the complex plane) can be extended to the strong-field regime while retaining their properties. The salient feature of these fields is a symmetry that cancels out power broadening effects. Such fields can be generated from ultrashort coherent pulses or from incoherent broadband down-converted light. Weak-field coherent-control approaches based on these solutions can therefore be extended to the strong-field regime as we demonstrate in a two-photon absorption experiment in atomic cesium.

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Ultrashort laser pulses have become the preferred tool for control of atomic and molecular processes due to their high peak intensity and short time duration. A major difficulty that results from the broad spectral bandwidth of femtosecond pulses is the loss of spectral selectivity, since all the transitions within that bandwidth may be excited. This difficulty can be overcome by quantum coherent-control techniques, using the interference between different quantum paths that reach the same final state. The central idea is to exploit constructive quantum interference in order to direct the system into a desired final state, while destructive interference attenuates the quantum amplitude of undesired states [1–3].

Coherent-control schemes are easily implemented to control weak-field interactions, due to the simple relation between the optical field and the resulting excitations. In a series of investigations it was demonstrated how manipulation of the spectral phase function can control the excitation field to achieve a resolution higher by orders of magnitude than the spectral bandwidth of the pulse [4–9]. A nonresonant two-photon absorption (TPA) process can be reduced and even eliminated using shaped pulses [4,5]. Similarly, resonant TPA was enhanced significantly beyond the level achieved by a transform limited pulse [6,7]. These concepts were also applied to achieve high spectral resolution coherent anti-Stokes Raman spectroscopy with broadband pulses [8,9].

For many practical applications, and, in particular, for effective control of chemical processes, strong-field interactions have to be considered. In general, weak-field schemes will not remain valid for stronger fields, since strong-field interactions are accompanied by level broadenings and level shifts that usually elude simple analytical treatments. An extensive effort has been directed in recent years towards the achievement of selective population transfer. Various schemes have been proposed, based on an adiabatic approach [10]. A non adiabatic scheme, proposed by Wollenhaupt *et al.* [11], demonstrated control over the photoelectron spectrum excited by strong fields, and investigated its dependence on the laser intensity and temporal shape. In this paper we show that it is possible to extend coherent-control schemes that were developed for weak fields into the strong-field regime, and thus retain the selectivity and the intuition developed by perturbative analysis. This extension is possible if one is willing to use fields with only one quadrature, as explained below.

We begin by reviewing the essence of weak-field control. Simple perturbation analysis predicts that a femtosecond laser pulse with an electric field $\varepsilon(t)$ driving a two-level system from the ground level $|g\rangle$ to a final level $|f\rangle$ excites an amplitude $a_{g\to f} \propto E(\omega_0)$, where $E(\omega)$ is the Fourier transform of $\varepsilon(t)$ and $\omega_0 = (E_f - E_g)/\hbar$ is the transition resonant frequency. A transition is therefore excited by a single frequency component, hence excitation amplitudes to a number of final states could obviously be adjusted through control of the spectrum.

This result has been generalized to N-photon transitions, where the excitation amplitude following the pulse is proportional to the resonant Fourier component of $\varepsilon^N(t)$ [4]:

$$a^N_{g\to f}(t\to\infty) = \frac{\mu^N_{fg}}{(i\hbar)^N}\int_{-\infty}^{\infty}\varepsilon(t)^N \exp(i\omega_0 t)dt \equiv \frac{\mu^N_{fg}}{(i\hbar)^N}E_N(\omega_0) \quad (1)$$

where μ^N_{fg} is the effective dipole matrix of the N-photon transition and $E_N(\omega)$ is the Fourier transform of the N-photon field $\varepsilon(t)^N$. Here too, the transition probability of an N-photon transition is dictated only by a single resonance component of $E_N(\omega)$ [4,12]. $E_N(\omega_0)$ is a function of the spectrum $E(\omega)$, and is determined by the interference between all N-photon combinations that additively sum to give the transition energy. Spectral phase manipulations were applied in various weak-field coherent-control schemes to control the energy content of this frequency component in order to manipulate nonlinear

processes [4,8,9]. In all weak-field experiments, a resonant transition was dictated by a *single* resonant frequency component of $E_N(\omega)$.

Now, as the field intensity increases, this simple relationship between the excitation amplitude and the spectrum of the N-photon field does not remain valid due to power broadening. We first analyze a one-photon transition and show that a family of solutions exists which cancels out power broadening effects completely. We then generalize this approach for the case of an N-photon transition. Eventually, we demonstrate these ideas experimentally through the control of TPA in the strong-field regime.

The interaction between a two-level system and a laser field beyond the weak-field regime is described by the optical Bloch equations (OBE), usually analyzed in a reference frame rotating at the laser frequency ω_l. However, when the laser field is much broader than the transition, the laser frequency is not well defined and the atomic transition frequency ω_0 becomes the natural reference frequency. The OBE can be then rewritten in the rotating wave approximation as

$$\frac{d}{dt}\begin{pmatrix} u \\ v \\ w \end{pmatrix} = \frac{\mu}{\hbar}\begin{pmatrix} \mathrm{Re}\{\varepsilon(t)\} \\ \mathrm{Im}\{\varepsilon(t)\} \\ 0 \end{pmatrix} \times \begin{pmatrix} u \\ v \\ w \end{pmatrix}, \tag{2}$$

where $\varepsilon(t)$ is the laser electric field in the rotating frame, w is the population inversion, and u, v are the atomic dipole moments in phase and in quadrature with the field, respectively. In this picture, the field driving force can be represented by a torque exerted to the Bloch vector (u, v, w). The torque vector lies in the uv plane and has a magnitude of $|\varepsilon(t)|$ and a direction given by the pulse temporal phase. The Bloch dynamics is governed by successive rotations of the Bloch vector around a torque vector, which change its direction with time. In the weak-field regime this is a succession of small rotations that are commutative. It is simple to show that this is equivalent to the perturbative solution of Eq. (1). In the strong-field regime the analysis becomes more complex. Large rotations around different axes do not commute, therefore the final population is determined by the specific temporal shape of the pulse and not just its total area [11].

The Bloch vector dynamics is greatly simplified if we constrain the field to be real ($\mathrm{Im}\{\varepsilon(t)\} = 0$). In that case, all the rotations of the Bloch vector are around the u axis only, and therefore the different field contributions commute. The OBE then yields the well known solution for a system initially in the ground state [13]:

$$w(t) = -\cos(\theta(t)), \qquad v(t) = -\sin(\theta(t)), \qquad u(t) = 0. \tag{3}$$

where $\theta(t)$ is the transient pulse area defined by $\theta(t) = \frac{\mu}{\hbar}\int_{-\infty}^{t}\varepsilon(t')\exp(i\omega_0 t')dt'$. The important consequence is that population inversion following the pulse excitation is dictated again by a *single* frequency component, since $\theta(\infty) \propto E(\omega_0)$.

While any electromagnetic field is always real, the reality required here is beyond this trivial one. Here, the slowly varying envelope of the field, which is generally complex, should be real, i.e., the optical field should have only one quadrature. An alternative view is a carrier wave that is only amplitude modulated. This implies the following symmetry condition in the frequency domain:

$$E(\omega_0 + \delta) = E^*(\omega_0 - \delta). \tag{4}$$

This symmetry leads to destructive interference between the contributions of frequency components above and below the resonance, thus canceling power broadening effects. This result is rather surprising; either the red-detuned sideband or the blue-detuned sideband alone would have induced significant power broadening, yet adding the symmetric spectral band (therefore increasing the total pulse intensity) leads to complete cancellation of the effect.

It is interesting to note that although our discussion focuses on coherent optical pulses, coherence is not a necessary feature. It is well known that broadband incoherent down-converted light is symmetric around its central frequency ($\omega_p/2$, ω_p being the pump frequency), and therefore satisfies Eq. (4) when $\omega_0 = \omega_p/2$ [5]. Indeed, with incoherent light the transient area of the field is unknown, so the transient population transfer cannot be predicted. However, when the field is real, only one quadrature of the atomic coherence, $v(t)$, is coupled to it. The other quadrature $u(t)$ is a constant of the Bloch dynamics, regardless of the temporal shape of the field. This can be viewed as a classical analogue of the quantum derivation, previously given by Gardiner [14].

We now extend the above analysis to multiphoton transitions. Consider a nonresonant multiphoton transition in a two-level system. Equation (1) predicts that when induced by a weak femtosecond laser pulse, the amplitude of the excited state will be determined by $E_N(\omega_0)$. As in the one-photon case, at higher laser intensities this relation does not remain valid due to power broadening, and the transition is dictated by the interference of all spectral components of $E_N(\omega)$. A nonresonant multiphoton transition in a two-level system is analyzed by the OBE, taking $\varepsilon(t)^N$ as the interacting field [15]. Therefore, we can extend our conclusion from the one-photon case, and state that power broadening in the multiphoton case is cancelled when $\varepsilon(t)^N$ is real.

In the following we will focus on the case of $N = 2$, analyzing coherent-control of TPA transitions in the strong-field regime. A nonresonant TPA process is dictated by the complex spectrum of $\varepsilon^2(t)$ given by [4]

$$E_2(\omega) = \int_{-\infty}^{\infty} E(\omega/2 + \delta)E(\omega/2 - \delta)d\delta. \tag{5}$$

Spectral phase manipulations of the spectrum $E(\omega)$ can be applied to control both the phase and the amplitude of $E_2(\omega)$. Selectivity in the weak-field regime was achieved by maximizing or canceling $E_2(\omega_0)$, where ω_0 is the two-

photon transition energy [4]. Selectivity in the strong-field regime can now be achieved by applying the additional constraint that $\varepsilon^2(t)$ has to be real. As is evident from Eq. (5), any spectral phase function which is antisymmetric around $\omega_0/2$, for example $\Phi(\omega) = \alpha \sin(\beta(\omega - \omega_0/2))$, will maximize $E_2(\omega_0)$. If, in addition, $|E(\omega)|$ is symmetric about $\omega_0/2$, then $\varepsilon(t)$ and therefore $\varepsilon(t)^2$ are real. Other spectral phase functions can drive $E_2(\omega_0)$ to be zero. Such pulse shaping annihilate the transition rate in the weak-field regime generating a "dark pulse" [4]. Generally, a dark pulse at low intensities will not remain dark at higher intensities due to power broadening. However, it is possible to find dark pulses with real $\varepsilon^2(t)$. In particular, for a spectral phase function $\Phi(\omega) = \alpha \cos(\beta(\omega - \omega_0/2))$, a specific value ($\alpha_0$) of α exist for which $E_2(\omega_0) = 0$, while $\varepsilon^2(t)$ is real. The important result is that this dark pulse is *invariant of the laser intensity*. In summary, two spectral phase functions, both valid also at high laser intensities, can be applied to achieve selective excitations by either maximizing or minimizing $E_2(\omega_0)$:

$$E(\omega)_{\max} = A(\omega - \omega_0/2)\exp[i\alpha_0 \sin(\beta(\omega - \omega_0/2))],$$
$$E(\omega)_{\min} = A(\omega - \omega_0/2)\exp[i\alpha_0 \cos(\beta(\omega - \omega_0/2))]. \quad (6)$$

where $A(\omega - \omega_0)$ is the field amplitude, constrained only to be a symmetric function around $\omega_0/2$.

To demonstrate a high spectral resolution in the strong-field regime, we studied TPA in cesium gas between the $6S_{1/2}$ and the $8S_{1/2}$ states [Fig. 1(a)]. The TPA is induced by amplified pulses generated by a multipass amplifier at a repetition rate of 1 kHz. The amplified pulses consist of a 40 nm bandwidth centered at 800 nm. The TPA rate is evaluated by measuring the fluorescence at ~460 nm due to spontaneous decay to the ground level through the $7P$ level. Spectral phases are applied by a programmable pulse shaper with a spectral resolution of about 0.6 nm. We tailored the spectrum with the shaper and filters to achieve a symmetric spectrum around 822 nm. The fluorescence signal is filtered out and measured with a photomultiplier tube and a lock-in amplifier. The collected signal is a result of an integration over the entire beam profile. We estimate the intensity at the center of the beam, $I_{\max}$, to be equivalent to a two-photon pulse area of $\theta_{\max} \sim 3\pi$. An outline of the experimental setup is presented in Fig. 1(b).

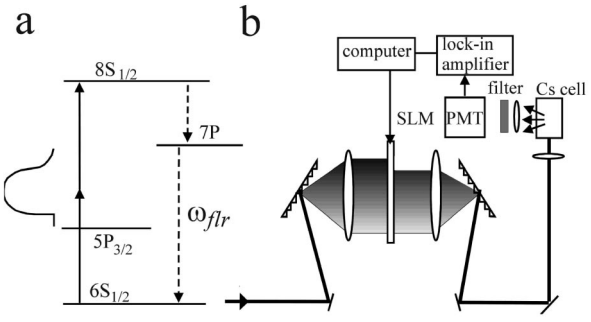

FIG. 1. (a) Energy level diagram of the two-photon transition $6S \rightarrow 8S$ in atomic Cs. Wavelengths longer than 840 nm are blocked as they spectrally overlap the $5P_{3/2}$ level. (b) Outline of the experimental setup.

We first applied an antisymmetric spectral phase function of $\Phi(\omega) = \alpha \sin(\beta(\omega - \omega_0/2))$. Figure 2 shows the measured signal as a function of the modulation depth α, for various values of pulse intensity ranging over an order of magnitude (black lines). The measured signal is normalized to the population transfer induced by an unshaped, transform limited pulse of the same laser intensity. This normalizing value should undergo Rabi oscillations at the beam center, but due to the averaging over the beam profile it just exhibits a saturating response. As predicted for the weak-field regime, the population transfer induced by an antisymmetric phase function is independent of the modulation depth (dotted black line). This response is maintained even when increasing the laser intensity to well within the saturation regime (solid black line). We next applied a symmetric spectral phase function of $\Phi(\omega) = \alpha \cos(\beta(\omega - \omega_0/2))$. The experimental results are presented by the gray lines in Fig. 2. In the weak-field regime we observed strong variations of the population transfer as α was increased, whereas no population transfer was induced for $\alpha = 1.4$ (dotted gray line). A similar response is observed for higher laser intensities. Cancellation of the population transfer at $\alpha = 1.4$ is demonstrated to be almost invariant to the field intensity. The minor deviation from the weak-field result is related to the existence of an intermediate $P_{3/2}$ level at 852 nm, close to the spectral range of the laser pulse [16]. It is to be noted that since the

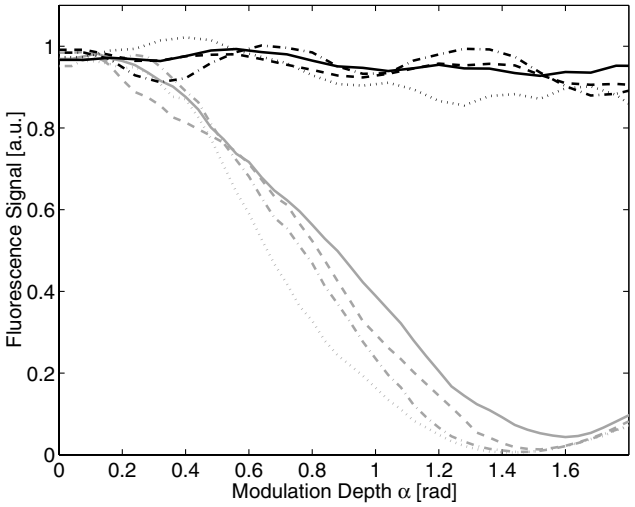

FIG. 2. TPA measurement for periodic spectral phase functions for which $\varepsilon^2(t)$ is real. Two spectral phase functions are applied: $\Phi(\omega) = \alpha \sin(\beta(\omega - \omega_0/2))$ (black lines) and $\Phi(\omega) = \alpha \cos(\beta(\omega - \omega_0/2))$ (gray lines). The TPA is measured as a function of the modulation depth α for pulse intensities (at the center of the beam) of $0.1I_{\max}$ (dotted line), $0.5I_{\max}$ (dashed-dotted line), $0.8I_{\max}$ (dashed line) and $I_{\max}$ (solid line).

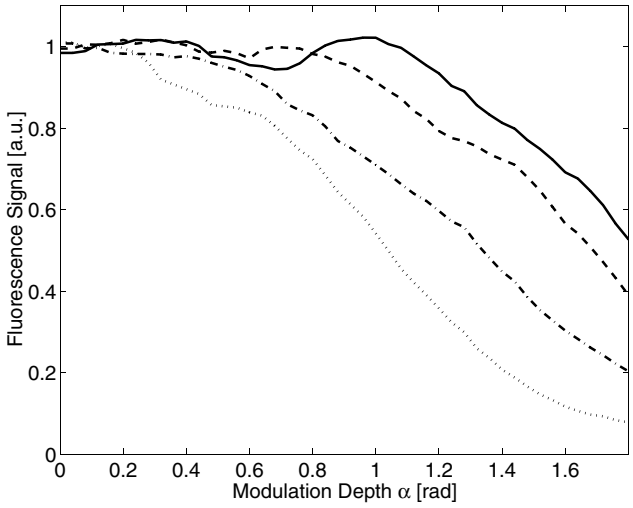

FIG. 3. TPA measurement for spectral phase function of $\Phi(\omega) = \alpha \sin(\beta(\omega - \omega_0/2) + \pi/4)$ for which $\varepsilon^2(t)$ has an imaginary part. The TPA is measured as a function of the modulation depth α for pulse intensities (at the center of the beam) of $0.1I_{\max}$ (dotted line), $0.5I_{\max}$ (dashed-dotted line), $0.8I_{\max}$ (dashed line) and $I_{\max}$ (solid line).

dark pulse is invariant to the field intensity, it is not obscured by the integration over the beam profile.

We should emphasize the point that the two fields discussed above, having a real $\varepsilon^2(t)$, are special. Generally, complex fields may induce significant power broadening. For example, by simply shifting the periodic spectral phase function by $\pi/4$ to apply a spectral phase function of $\Phi(\omega) = \alpha \sin(\beta(\omega - \omega_0/2) + \pi/4)$, we generate $\varepsilon^2(t)$ that is composed of three main pulses with relative phases of i, 0, $-i$. Figure 3 shows the measured signal as a function of modulation depth for various values of the pulse area. In the weak-field regime we observe a smooth reduction of the signal with increasing α (dashed line). However, increasing the laser intensity resulted in a significant deviation from the weak-field response. As the laser intensity increases, the fluorescence signal decays slower as a function of α (dashed-dotted, dashed, and solid lines), indicating that power broadening plays a significant role in this case. Although $E_2(\omega_0)$ decreases with α, population transfer is induced by other spectral components of $E_2(\omega)$, hence the slower decrease of the measured signal.

In summary, we have shown that power broadening can be cancelled when the transition is induced by real fields. Power broadening cancellation is a result of the destructive interference induced by symmetry properties of the pulse spectrum. The direct consequence is that the transition is dictated by a *single* frequency component, which allows the extension of weak-field control schemes into the strong-field regime. This is true for simple two-level transitions, as well as for nonresonant multiphoton transitions, as demonstrated by our TPA experiment. With this scheme, selective excitation in the strong-field regime can be readily achieved. Although our discussion deals primarily with shaped coherent pulses, its direct applicability to incoherent broadband down-converted light illustrates the generality of the presented principles. Similar ideas could apply to other control schemes, such as resonant multiphoton transition, transient effects [6,7], and Raman transition [8,9]. We believe that the scheme presented here can be utilized as a ''building block'' to manipulate more complex strong-field interactions such as control of wavepacket dynamics, photodissociation, or ionization.

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