Temporal Coherent Control in Two-Photon Transitions: From Optical Interferences to Quantum Interferences

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Temporal coherent control of an excited state wave packet is produced by a sequence of two identical ultrashort laser pulses. We show theoretically and experimentally in the case of the ($6s$-$7d$) two-photon transition in Cs that optical and quantum interferences take place and are clearly distinguished. [S0031-9007(97)02872-X]

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Recently a great deal of attention has been devoted to laser control of chemical and molecular processes [1]. The goal is to take advantage of laser radiation properties to favor a specific outcome when several output channels can occur simultaneously in the process.

Several kinds of control have been investigated. Optimal control consists of shaping the laser pulse in order to reach a well-defined target, but remains mostly inaccessible to the experimentalist, except in very limited cases [2]. In coherent control, quantum interferences between several quantum paths are used to modulate a specific channel [3–9]. Each quantum path results from the interaction between a laser field and the atomic or molecular system. Variation of the relative phases between these laser fields induces constructive or destructive interferences between quantum paths. Excitation probability of the process is thus coherently controlled.

One way to achieve coherent control consists of using the combination of a fundamental frequency and one of its harmonics [3–5]. In the case of an odd harmonic, both quantum paths reach the same excited state, and the total cross section can be controlled [3,4]. With an even harmonic, the quantum paths lead to degenerate states with opposite parity, and differential cross sections can be controlled [5].

In temporal coherent control, a sequence of two time-delayed pulses is used to follow the two quantum paths [6–9]. The interference phase is related to the time delay between the two pulses. Variation of the time delay results in interferograms which show a high frequency oscillation (corresponding to Ramsey fringes [10]) modulated by a slow envelope resulting from the wave packet motion in the excited state. Various detection schemes have been used to isolate this slow envelope from the rapid oscillations. Three examples are the phase lock technique [6], the sensitive phase technique [7], and the random phase technique [11]. These techniques allow extraction of the wave packet motion contribution from the signal. However, to achieve control of the process, the time delay must be fixed with an accuracy much better than the optical period in order to select between constructive or destructive interferences which result in enhancement or weakening of the total cross section of the process. Most of these experiments have been performed in the weak field regime [6,7,9]. These quantum interferences persist in the intermediate or high field regime, where higher transfer rates and large control contrasts are observed [8]. However, the analysis of the data is difficult because of ac Stark effects and multiple Raman processes populating numerous states. Thus a careful numerical integration of the Schrödinger equation is required [8].

We show in this paper that temporal coherent control can be applied to two-photon transitions in the weak field regime, where striking new features are observed and where a complete analysis of all the interference phenomena is given. Namely, we are able to distinguish for the first time between optical interferences (oscillations at the optical period), which take place when the two pulses overlap temporally, an purely quantum interferences (oscillations at half the optical period) when the two pulses are well separated. This new scheme also provides an efficient tool for probing the excited states wave packet dynamics which contains all the spectroscopic information on the system, extending the Fourier transform spectroscopy to a powerful coherent Fourier transform multiphoton ionization (CFT-MPI) spectroscopic tool.

The atomic system considered here consists of three sets of levels: the ground state $|g\rangle$, the intermediate states $|i_l\rangle$, and the excited states $|e_k\rangle$. Their corresponding energies are denoted 0, $\hbar\omega_{i_l}$, and $\hbar\omega_{e_k}$. The electromagnetic field $\mathcal{E}(t)$ consists of a superposition of two ultrashort, Fourier-transform-limited, linearly polarized (along the same direction) laser pulses separated by a delay time τ. $\mathcal{E}(t) = \mathcal{E}_p(t) + \mathcal{E}_c(t)$, where (in the rotating wave approximation) $\mathcal{E}_p(t) = \mathcal{E}_0(t)e^{-i\omega_L t}$ and $\mathcal{E}_c(t) = \beta\mathcal{E}_p(t - \tau) = \beta\mathcal{E}_0(t - \tau)e^{-i\omega_L(t-\tau)}$. β is the ratio between the amplitudes of the electric fields of the two pulses (assumed to be real). $\mathcal{E}_0(t)$ is the pulse envelope, of duration τ_L and spectral width τ_L^{-1}, and ω_L is the "central" frequency of the pulse. We have assumed here two perfectly collinear laser beams with identical longitudinal and transverse profiles, so that it is

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not necessary to take explicitly into account the spatial variation of the electric field.

At resonance, the central frequency of the laser ω_L is chosen to be close (within the spectral width) to half the energy difference between the excited states $|e_k\rangle$ and ground state $|g\rangle$, $|2\omega_L - \omega_{e_k}| \leq \tau_L^{-1}$. We consider only the nonresonant (with respect to the intermediate states) case (two-photon excitation instead of sequential excitation) so that no population remains in the intermediate states after the laser pulse sequence, $|\omega_L - \omega_{i_l}| \gg \tau_L^{-1}$. Also, we limit our treatment to the weak-field regime and use a second-order time-dependent perturbation theory.

After the laser pulses $[(t - \tau) \gg \tau_L]$, the total wave function of the system is expressed as

$$|\Psi(t)\rangle = |g\rangle + \sum_k a_{e_k}(t)\,|e_k\rangle \tag{1}$$

if no depletion of the ground state is assumed, where

$$a_{e_k}(t) = \frac{i}{\hbar}\,Q_{g,e_k} e^{-i\omega_{e_k} t}\tilde{\mathcal{E}}^2(\omega_{e_k})\,. \tag{2}$$

Here the two-photon transition operator [12] is given by

$$Q_{g,e_k} = \sum_l \frac{\langle e_k|\mu|i_l\rangle\langle i_l|\mu|g\rangle}{\hbar(\omega_{i_l} - \omega_L)} \tag{3}$$

(μ is the electric dipole moment of the atom), and

$$\tilde{\mathcal{E}}^2(\omega) = \int_{-\infty}^{+\infty} \mathcal{E}^2(t) e^{i\omega t}\,dt \tag{4}$$

is the Fourier transform of the square of the electric field. The τ dependence of the amplitude $a_{e_k}(t)$ is completely contained in the $\tilde{\mathcal{E}}^2(\omega_{e_k})$ factor of Eq. (2). The two-pulse sequence leads to the following expression:

$$a_{e_k}(t) = K_1^{e_k} + K_2^{e_k}(\tau) + 2K_{12}^{e_k}(\tau)\,, \tag{5}$$

with

$$K_1^{e_k} = \frac{i}{\hbar}\,Q_{g,e_k} e^{-i\omega_{e_k} t}\tilde{\mathcal{E}}_0^2(\omega_{e_k} - 2\omega_L)\,, \tag{6a}$$

$$K_2^{e_k}(\tau) = \beta^2 K_1^{e_k} e^{i\omega_{e_k}\tau}, \tag{6b}$$

$$K_{12}^{e_k}(\tau) = \frac{i}{\hbar}\,Q_{g,e_k}\beta e^{-i\omega_{e_k} t} e^{i\omega_L\tau} \times \int_{-\infty}^{+\infty} \mathcal{E}_0(t')\mathcal{E}_0(t' - \tau) \times e^{+i(2\omega_L - \omega_{e_k})t'}\,dt'. \tag{6c}$$

These contributions account for all possible quantum paths leading the system from the ground state to the excited states (see Fig. 1). They correspond to the absorption of two photons from the first pulse ($K_1^{e_k}$), two photons from the second pulse ($K_2^{e_k}$), and one photon from each laser pulse ($K_{12}^{e_k}$). As the intermediate states, $|i_l\rangle$, are nonresonant, this last term vanishes as the pulse overlap decreases. This means that, since these two photons are absorbed "simultaneously" (within a time equal to the inverse of the energy defect in the intermediate state), both combinations (photon of pulse 1 followed by photon of pulse 2 or vice versa) contribute equally. This explains the factor 2 in Eq. (5). The two terms $K_1^{e_k}$ and $K_2^{e_k}$ differ only by an amplitude factor β^2 and a phase factor $e^{i\omega_{e_k}\tau}$.

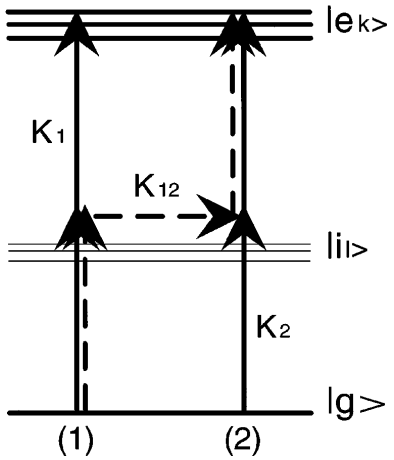

FIG. 1. Schematic diagram of the levels involved and of the various paths considered [see Eq. (6)]. (1) and (2) refer to the pulse numbers.

The population in each excited state after the end of the interaction is given by $|a_{e_k}(\infty)|^2$ and contains contributions from each path considered separately and interference terms between these various paths. The phase factors which govern these interferences depend on the product of the time delay τ and the frequencies ω_L or ω_{e_k}. An example of the evolution of the excited states population as a function of τ is given in Fig. 2 in the case of two excited states. Three insets show expanded views of the population for the characteristic values of the time delay: 2(a) perfect overlap of the two pulses

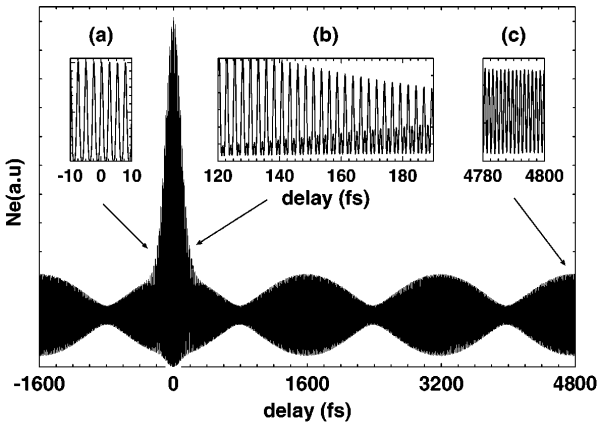

FIG. 2. Simulation of the population variation of the $7d$ cesium state as a function of the time delay τ between the two excitation laser pulses (at $\lambda = 768$ nm, $\beta = 0.7$ corresponding to experimental conditions). The three insets show the two regimes of interferences: (a) optical interferences, (c) quantum interferences, and (b) the transition between these two regimes.

($\tau \ll \tau_L$), 2(b) partial overlap ($\tau \simeq \tau_L$), and 2(c) no overlap ($\tau \gg \tau_L$).

When $\tau \ll \tau_L$, the temporal overlap of the two laser pulses results in optical interferences. A simple expression can be obtained by equating $\mathcal{E}_0(t - \tau)$ to $\mathcal{E}_0(t)$. The expression of the electric field then reduces to $\mathcal{E}(t) \simeq (1 + \beta e^{i\omega_L \tau})\mathcal{E}_0(t)$, where the factor $(1 + \beta e^{i\omega_L \tau})$ accounts for optical interferences. From Eqs. (2) and (6a), one obtains the total population in the excited states, for ($\tau \ll \tau_L$),

$$N_e(\tau) = \sum_k |a_{e_k}(\infty)|^2 = |1 + \beta e^{i\omega_L \tau}|^4 \sum_k |K_1^{e_k}|^2. \quad (7)$$

Optical interferences take place at the optical frequency ω_L and result in strong oscillations of the intensity of the laser beam and, in consequence, of the excited states' population. These oscillations have exactly the same frequency for all the excited states $|e_k\rangle$ as they result only from optical interferences. Because of the nonlinearity of the two-photon transition, the oscillations are not symmetric with respect to the average value. In other words, the population oscillates with the period $2\pi/\omega_L$, but these oscillations contain both frequencies ω_L and ω_{e_k} ($\simeq 2\omega_L$) as appears clearly in Eq. (6).

When the time delay is such that the pulses do not overlap ($\tau \gg \tau_L$), then the path $K_{12}^{e_k}$, corresponding to one photon absorbed from each pulse, vanishes. This again yields a simple expression for the excited state population, which results from quantum interferences between paths $K_1^{e_k}$ and $K_2^{e_k}$, for ($\tau \gg \tau_L$),

$$N_e(\tau) = \sum_k (1 + \beta^4 + 2\beta^2 \cos \omega_{e_k} \tau) |K_1^{e_k}|^2. \quad (8)$$

Equation (8) is the superposition of terms which oscillate at the frequency of each transition ω_{e_k} (close to $2\omega_L$) between the extreme values $(1 + \beta^2)^2 |K_1^{e_k}|^2$ (constructive interferences) and $(1 - \beta^2)^2 |K_1^{e_k}|^2$ (destructive interferences). The phase factor $\omega_{e_k} \tau$ results from the parallel evolution of the two waves in the two levels (initial and excited) separated by the energy $\hbar\omega_{e_k}$ during the time interval τ. Each excited state population oscillates at its own frequency ω_{e_k}. After delays greater than the reciprocal of the excited states energy splittings $\hbar\Delta\omega_{e_k}$, the interferograms associated with each individual excited state $|e_k\rangle$ dephase and beats result. These beats correspond to the time evolution of the wave packet created in the excited state by the first pulse and reflect the energy structure of the excited states manifold. When the interference contrast is maximal, the two wave packets created by the two laser pulses are identical (or have a maximal overlap), and interferences are constructive or destructive according to the relative phase between the wave packets. At the nodes, the two excited state wave packets are orthogonal and cannot interfere. This result can be generalized to any multiphoton transition: For a resonant transition of frequency ω_e excited through a n-photon transition, the interferogram exhibits rapid oscillations at the frequency ω_e ($\simeq n\omega_L$) resulting from quantum interferences.

For intermediate time delays ($\tau \simeq \tau_L$), optical and quantum interferences take place simultaneously. As the two laser pulses overlap partially in time, optical interferences are partially contrasted so that quantum interferences (at twice the optical frequency) can show up. This effect is shown in the inset (b) of Fig. 2 which gives an extended view of the [120–190 fs] time interval. The exact expression of the excited state population N_e contains independent contributions from the three possible quantum paths [see Eq. (6)] and interference terms between these paths. The direct contributions $|K_1^{e_k}|^2$ and $|K_2^{e_k}(\tau)|^2$ are independent of τ, and $|K_{12}^{e_k}(\tau)|^2$ drops monotonically with τ as the overlap between the two pulses decreases. The interference terms between $K_1^{e_k}$ or $K_2^{e_k}(\tau)$ and $K_{12}^{e_k}(\tau)$ produce oscillations at the optical frequency ω_L whose amplitude also decreases regularly with τ. They correspond to the optical interferences. Finally, the interference term between $K_1^{e_k}$ and $K_2^{e_k}(\tau)$ has a constant amplitude and is modulated at the excitation frequencies, ω_{e_k}, close to twice the optical frequency. For the shortest delays, the interference pattern presents single maxima separated by the optical period. Although the quantum interferences are present, they are hidden by the optical interferences and contribute only to modify the shape of the fundamental period which is not a simple cosine function. As the time delay gradually increases, secondary maxima appear in between the main maxima, as a result of the quantum interferences between the part of the laser pulses which do not overlap and cannot therefore interfere optically.

The phenomena discussed above have been successfully demonstrated in the two-photon excitation to the $7d\,^2D_{3/2,5/2}$ fine-structure doublet in Cs at 768 nm. The intermediate states, $6p\,^2P_{1/2,3/2}$ are off resonance by ca 1300 to 1850 cm^{-1}. The $7d$ fine structure splitting is 20.97 cm^{-1}, corresponding to a beat period of 1.591 ps. The Cs($7d$) atoms are then ionized by the absorption of an extra photon from either pulse (1) or (2). One can show [9] that the ion signal is qualitatively similar to the excited state population calculated here.

The experimental setup has been described in detail elsewhere [9]. Briefly, a titanium sapphire laser delivers 13 nJ pulses at 76 MHz repetition rate. The measured pulse duration is ca 150 fs, and the corresponding spectral width is 120 cm^{-1} (FWHM), so that the off-resonance condition for the intermediate states is fulfilled. The laser pulses are split into two parts and recombined in a Mach-Zender type interferometer, with a variable optical path difference resulting in a two-pulse sequence. Copropagating pulses are slightly focused on a cesium atomic beam. Ions resulting from the interaction with the laser are detected with a quadrupole mass spectrometer.

Experimental results are displayed in Fig. 3. A low-resolution scan ([−10 ps, +30 ps] interval) shows the broad features: strong peaks resulting from optical

interferences around $\tau = 0$ fs, high-frequency oscillations modulated by a slow envelope resulting from the beats between the two fine-structure excited states, at a period of 1.59 ps. The insets show high-resolution scans which allow us to distinguish between optical interferences [3(a): 2.56 fs period], quantum interferences [3(c): 1.28 fs period], and the transition regime 3(b), where quantum interferences gradually show up as the temporal overlap between the laser pulses drops. The qualitative agreement between the observed ionic signal and the calculated excited state population (Fig. 2) is excellent.

We have shown in this paper that the temporal coherent control technique relies on two types of interferences (quantum and optical) which are clearly identified in the case of a two-photon transition. This technique opens a broad field of applications. As a spectroscopic tool, this "Fourier transform multiphoton ionization" (FT-MPI) technique provides information on the energetic structure and on the dynamics of the excited state. It also provides coherent control capabilities which are manifold. If several channels can be simultaneously controlled with different phases, the branching ratios between these channels can thus be controlled. This opens wide applications, particularly in polyatomic systems. Ultrafast switching devices can be developed with semiconductors [13]. Finally, the efficiency of the control process can be greatly enhanced by using sequences of coherent laser pulses such as in NMR.

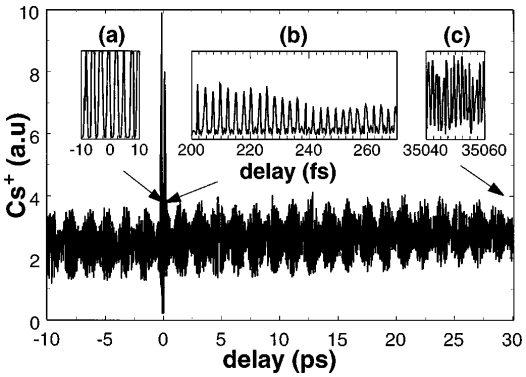

FIG. 3. Variation of the (2 + 1) resonance-enhanced multiphoton ionization (REMPI) probability in Cs as a function of the time delay between the two laser pulses (at $\lambda = 768$ nm, with $\beta = 0.7$). The $7d\,^2D_{3/2,5/2}$ fine-structure doublets are the resonant states for the two-photon transition. The three insets show the two regimes present in this experiment: (a) optical interferences with a 2.56 fs period, (c) quantum interferences with a 1.28 fs period, and (b) the transition between these two regimes.

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