Coherent control of quantum interference in two-photon absorption

H. G. Barros, J. Ferraz, W. Lozano B., L. H. Acioli, and S. S. Vianna*
Departamento de Física, Universidade Federal de Pernambuco, 50670-901 Recife, PE, Brazil
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We investigate how tailoring the phase of the optical field allows us to select the different routes induced by time-delayed femtosecond pulse pairs exciting a two-photon transition in rubidium atoms. It is shown that by introducing an external phase in one of the pulses, the pathways leading to optical interferences can be eliminated, leaving only the quantum interferences.

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In recent years, femtosecond lasers have become a standard tool for the development of several branches of science. In particular the use of these lasers has been responsible for major breakthroughs in the field of coherent control, with prospects for applications in physics [1], biology [2], and chemistry [3,4]. In these studies the main goal is to control the outcome of a certain process by exploiting interferences between different quantum paths [5], which may be performed either in a weak-field limit, where perturbation theory is adequate, or in the strong-field limit [4].

This microscopic control of the system's evolution has been explored using different techniques. In temporal coherent control, for example, pairs of pulses of equal shape are employed to both excite and probe the sample, and the interference phase is related to the different times at which the two pulses arrive at the sample. Such a scheme, when applied to two-photon transitions, results in interference terms that occur at twice the central laser frequency (purely quantum interference) and which are closely related to Ramsey fringes, and also at the laser frequency (optical interference) when the two pulses overlap temporally [6,7]. Control of quantum interactions has also been achieved by manipulating the coherent properties of the optical field: by applying pulse shaping techniques, Meshulach and Silberberg [8] have demonstrated, for example, a reduction and even an annihilation of the two-photon absorption rate. Recently, Präkelt *et al.* [9] combined both techniques—temporal coherent control and pulse shaping—and showed that the effect of a single phase-modulated pulse can be efficiently controlled by a suitably timed prepulse. These combined techniques were also used to analyze the two-photon transition rates in the presence of an intermediate resonance [10] in the case where significant phase alterations are produced by propagation effects.

In this work we use the combination of a phase-tailored pulse with a second time-delayed pulse directly from the laser to select different routes involved in a two-photon transition. The experiment is performed in Rb vapor, and the externally introduced phase has the shape of a step function in the spectral domain. We show that for a given phase step it is possible to find a time delay τ for which determined routes are annihilated. In this situation, the optical interferences are destroyed and only the quantum interferences are observed.

*Electronic address: vianna@ufpe.br

The energy-level diagram of Rb with the relevant states is presented in Fig. 1(a). Two-photon absorption (TPA) takes the population from the $5S$ ground state, $|g\rangle$, to the excited $7S$ state, $|f\rangle$, with energies E_g and E_f, respectively. The absence of an intermediate resonance reduces the manifestation of accumulation effects [11,12]. The central frequency of the femtosecond laser, ω_L, is tuned to half the atomic frequency $\omega_0=(E_f-E_g)/\hbar$. The fluorescence emitted by spontaneous decay from $6P$ to $5S$ is a direct measure of the population excited to the $7S$ state.

In the weak-field regime, the two-photon transition from the ground state to the excited state can be described by second-order time-dependent perturbation theory. If the intermediate states are sufficiently far from resonance, the amplitude of the excited state $|f\rangle$ at $t\rightarrow\infty$ is given by

$$a_f = \frac{i}{\hbar} Q_{g,f} \int_{-\infty}^{\infty} \tilde{\mathcal{E}}_T(\omega)\tilde{\mathcal{E}}_T(\omega_0-\omega)d\omega, \tag{1}$$

where

$$Q_{g,f} = \sum_n \frac{\mu_{fn}\mu_{ng}}{\hbar(\omega_{ng}-\omega_L)}$$

is the two-photon operator, μ_{ij} are the dipole moment matrix elements, $\omega_{ij}\equiv(E_i-E_j)/\hbar$, with E_i being the energy of state i, and the summation is performed over all possible intermediate states $|n\rangle$ of the unperturbed atom. $\tilde{\mathcal{E}}_T(\omega)$ is the Fourier transform of the total electromagnetic field, $\mathcal{E}_T(t)$, which consists of a superposition of two laser pulses separated by a time delay τ and with an external phase $\phi(\omega)$ introduced in only one of the pulses. In this case, the total field in the spectral domain depends on τ and $\phi(\omega)$ as follows:

$$\tilde{\mathcal{E}}_T(\omega,\tau,\phi) = \tilde{\mathcal{E}}_1(\omega,\tau) + \tilde{\mathcal{E}}_2(\omega,\phi), \tag{2}$$

where $\tilde{\mathcal{E}}_1(\omega,\tau)=A(\omega)\exp(i\omega\tau)$, $\tilde{\mathcal{E}}_2(\omega,\phi)=\beta A(\omega)\exp[i\phi(\omega)]$, $A(\omega)$ is the spectral amplitude, and β is the ratio of the amplitudes of the two pulses.

After the two-pulse sequence, the population at level $|f\rangle$ can be written as

$$\rho_f(\tau) = |a_f|^2 = |F_{11}\exp(i\omega_0\tau) + \beta F_{12}(\tau)\exp(i\omega_{ng}\tau) + \beta F_{21}(\tau)\exp(i\omega_{fn}\tau) + \beta^2 F_{22}|^2, \tag{3}$$

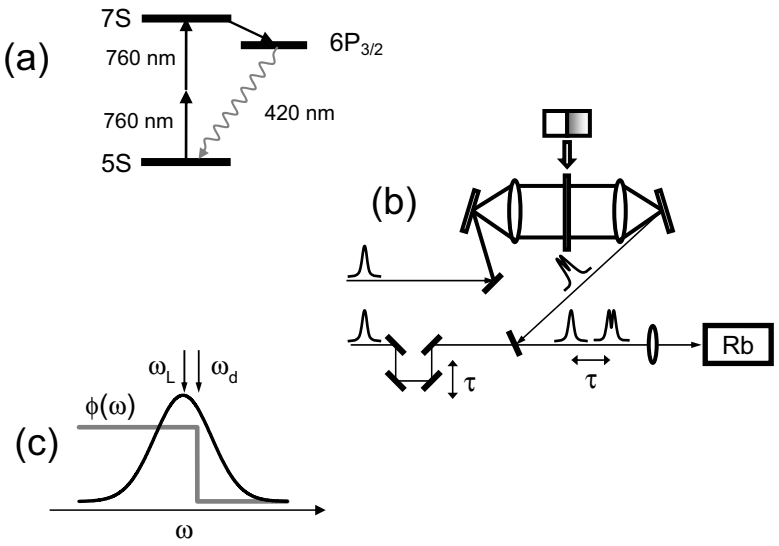

FIG. 1. (a) Diagram of relevant energy levels of Rb. (b) Schematic of the experimental setup. Fluorescence at 420 nm is collected at 90° from the center of the cell. (c) Spectrum of the femtosecond laser and the phase function $\phi(\omega)$.

with

$$F_{11} = \frac{i}{\hbar} Q_{g,f} \int_{-\infty}^{\infty} A(\omega) A(\omega_0 - \omega) d\omega, \tag{4a}$$

$$F_{22} = \frac{i}{\hbar} Q_{g,f} \int_{-\infty}^{\infty} A(\omega) A(\omega_0 - \omega) \exp[i\phi(\omega) + i\phi(\omega_0 - \omega)] d\omega, \tag{4b}$$

$$F_{12} = \frac{i}{\hbar} Q_{g,f} \int_{-\infty}^{\infty} A(\omega) A(\omega_0 - \omega) \times \exp[i(\omega - \omega_{ng})\tau] \exp[i\phi(\omega_0 - \omega)] d\omega, \tag{4c}$$

$$F_{21} = \frac{i}{\hbar} Q_{g,f} \int_{-\infty}^{\infty} A(\omega) A(\omega_0 - \omega) \times \exp[-i(\omega - \omega_{ng})\tau] \exp[i\phi(\omega)] d\omega. \tag{4d}$$

The four terms in Eq. (3) represent the different routes that an atom initially in the ground state can take to reach the final state. They correspond to the absorption of two photons from the field $\tilde{\mathcal{E}}_1$ (F_{11}), two photons from the field $\tilde{\mathcal{E}}_2$ (F_{22}), and one photon from each field (F_{12} and F_{21}). Each term reflects the fact that two-photon transitions occur for all pairs of photons with frequencies $\omega_i + \omega_j = \omega_0$. When the square modulus is taken in Eq. (3), the resulting 16 terms give the probability for each one of these processes plus the quantum interferences between them. If the external phase $\phi(\omega)$ is absent, the function $\rho_f(\tau)$ constitutes one of the basic results for temporal coherent control of two-photon transitions and a full description of all its terms can be found, for example, in Ref. [7].

In the experiment the Rb vapor is in a 5-cm-long sealed cell that is heated up to ≈130 °C (estimated density of 3.4×10^{13} atoms/cm^3). The atoms are excited from the $5S$ state to the $7S$ state by pairs of femtosecond pulses generated by a Ti:sapphire laser with a repetition rate of 82 MHz and tuned to $\lambda_L = 760$ nm ($\Delta\lambda \approx 6$ nm). Figure 1(b) shows a schematic view of the experimental setup. Before reaching the cell, one of the pulses, $\tilde{\mathcal{E}}_1$, is sent through a delay line while the other, $\tilde{\mathcal{E}}_2$, goes through a pulse shaper where it receives the phase $\phi(\omega)$. The pulse shaper consist of a (1×) telescope ($f = 10$ cm lenses) and two diffraction gratings (1200 lines/nm). At the focal plane between the lenses is placed a glass plate partially covered with a thin transparent film. The position of the film edge determines the reference frequency (ω_d) for which all frequencies of the pulse less than ω_d receive the extra phase. In Fig. 1(c) is shown the spectrum of the laser and the external phase $\phi(\omega)$. After recombination, the pulse pairs are focused on the middle of the cell and the fluorescence at 420 nm is collected perpendicularly to the direction of the laser beam and monitored as a function of the temporal delay, τ.

First we investigated the contribution due to the absorption of one photon from each field. For this the two beams $\tilde{\mathcal{E}}_1$ and $\tilde{\mathcal{E}}_2$ are chopped at different frequencies and the signal is detected at the sum frequency by a lock-in amplifier. This corresponds to observing only the contribution of the terms F_{12} and F_{21} in Eq. (3). The results for the partial fluorescence as a function of τ when the phase step is approximately equal to 2.9π are shown in Fig. 2(a) for different normalized positions of the phase step [$\delta_\phi = (\omega_d - \omega_L)/\Delta\omega$, where $\Delta\omega$ is the full width at half maximum (FWHM) spectral bandwidth of the femtosecond pulse]. The results of the phase modifications are monitored through cross-correlation measurements and are displayed in Fig. 2(b). The solid lines are the theoretical results obtained when the oscillations at the optical frequencies were eliminated because they are averaged out for fast scans of the delay τ. In Fig. 2(a), it is shown that, for this phase step, it is possible to null the two quantum paths corresponding to the terms F_{12} and F_{21}, when the position of the phase step is equal to half the atomic frequency ($\delta_\phi = 0$). This effect is related to the generation of *dark* pulses—i.e., pulses that induce no net multiphoton transitions [13]—and may be understood by examining Fig. 3 where two pulses are

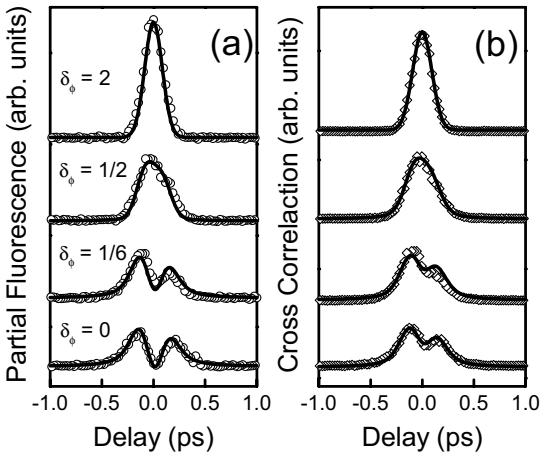

FIG. 2. Partial fluorescence (a) and cross-correlation measurements (b) versus τ for different normalized positions of the π-phase step.

shown, one with and the other without the phase step, as well as their product. As it is the integral of the product of these two pulses in the spectral domain that determines the F_{12} and F_{21} contributions, it is clear that for a π-phase step these terms vanish at $\tau=0$. For the experimental conditions under which Fig. 2 is obtained the appropriate coherent superposition of the optical frequencies that cancel each of the two paths occurs close to $\tau=0$.

Figure 4 shows the theoretical results obtained for different values of the phase step when its position is fixed at $\delta_\phi=0$. For values of the phase step different from π an additional phase contribution due to the delay between the pulses, $\phi_{propagation}(\omega,\tau)=\omega\tau$, is needed to generate a *dark* pulse. In fact, Fig. 4 shows that the cancellation of the two routes depends on both the time delay and the phase step.

To demonstrate that this coherent superposition of the optical frequencies can efficiently annihilate determined routes induced by pairs of time-delayed pulses, we have measured the total fluorescence from the atoms which is proportional to the final $7S$ population. In this case, the two beams $\tilde{\mathcal{E}}_1$ and $\tilde{\mathcal{E}}_2$ are chopped at same frequency and the variation of the total fluorescence with τ, for a 3.15π-phase step and $\delta_\phi=0$,

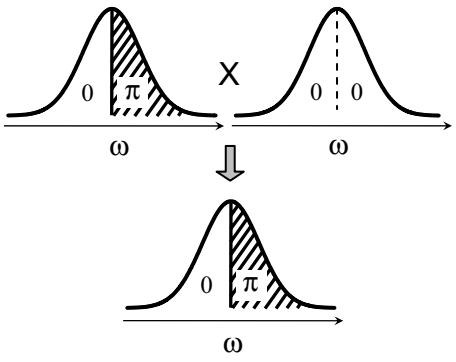

FIG. 3. Schematic phase distribution representation, at zero time delay, for one pulse with a π-phase step and other with zero external phase (top) and the spectral product of these two pulses (bottom) in the case that the partial two-photon absorption is null.

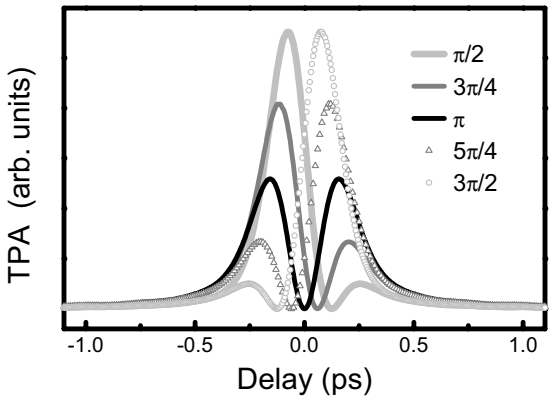

FIG. 4. Calculated TPA due to the contribution of one photon from each field for different values of the phase step and $\delta_\phi=0$.

is presented in Fig. 5. In Fig. 5(a) is shown a low-resolution scan of τ. For this phase step, the asymmetry of the phase in one of the pulses is compensated by the temporal delay between the two pulses when $\tau\approx-40$ fs. At this delay the total fluorescence intensity presents a minimum, with values comparable to the fluorescence obtained for $\tau\geqslant 500$ fs, when both pulses are completely separated in time. On the other hand, for $\tau\approx 140$ fs and $\tau\approx-190$ fs, when the overlap of the pulses is smaller, the fluorescence intensity presents local maxima.

In Figs. 5(b) and 5(c), we see the results for high-resolution scans around two different delays. For delays of $\tau\approx-190$ fs, Fig. 5(b), optical interferences with a period of 2.6 fs corresponding to the central laser frequency ω_L are observed. This result indicates that the four routes described in Eq. (3) are present. This means that the two pulses still overlap in time and the optical contribution dominates the process. Near $\tau=-40$ fs, Fig. 5(c), we see a pattern with an oscillating frequency that correspond to the atomic transition frequency, ω_0. The observation of the quantum interferences

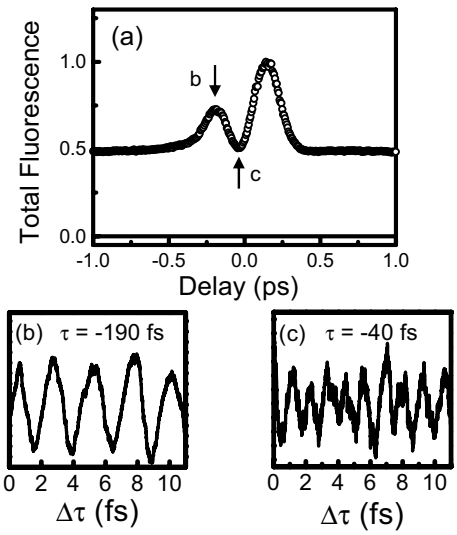

FIG. 5. Variation of the total fluorescence with the relative temporal delay between the pulse pair. (a) Low-resolution scan. High-resolution scans for delays (b) $\tau\approx-190$ fs and (c) $\tau\approx-40$ fs.

when the two pulses overlap is a clear demonstration that each route involving the absorption of one photon of each field has been annihilated.

In conclusion, we have showed that by using a single phase-tailored pulse it is possible to achieve selective control over the routes induced by pairs of ultrashort pulses in a two-photon transition. In particular, for a given constant difference of phase applied to one-half of the Fourier components, we always find a temporal delay where the optical interferences are destroyed, leaving only the quantum interference.

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