# Coherent Transient Enhancement of Optically Induced Resonant Transitions

Nirit Dudovich, Dan Oron, and Yaron Silberberg

*Department of Physics of Complex Systems, Weizmann Institute of Science, Rehovot 76100, Israel*

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By applying pulse shaping techniques to a broadband 100 fs pulse in resonance with a two-level atomic transition, we are able to enhance the peak transient excited level population relative to that achievable with transform limited pulses. We also demonstrate how the dispersion induced by the absorption line itself leads to similar rapidly oscillating transients in the excited population. These transient population effects are applicable in any multiphoton resonant transition.

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In coherent quantum control, the primary goal is to steer a quantum system towards a desired final state through its interaction with light, while canceling out other paths leading to undesirable outcomes. Such quantum interference between different paths appears inherently in all nonlinear light matter interactions such as multiphoton processes. The ability to control such interference has been proposed [1–4], and demonstrated in a variety of atomic [5], molecular [6–11], and solid-state [12] systems, in particular the very simple process of two-photon absorption (TPA). Broers *et al.* [13] observed enhancement of a resonant TPA excited by chirped laser pulses at the high intensity regime, and oscillations of the resonant TPA at intermediate intensities. Meshulach and Silberberg [14] demonstrated that by applying pulse shaping techniques, one could reduce and even annihilate the TPA rate, while Dudovich *et al.* [15] demonstrated that by using a similar procedure a resonant TPA can be enhanced significantly beyond the level achieved by a transform limited pulse.

In this Letter we consider the simplest resonant interaction of a broadband pulse with a two-level system. In the weak field regime, the excited state population, for times which are considerably longer than the pulse duration, depends only on the energy content of the resonant frequency component of the excitation pulse. Neither the phase nor the amplitude of all other spectral components of the pulse affect the transition probability. These components, however, do affect the transient excited state population during the action of the pulse. Zamith *et al.* [16] demonstrated oscillation of the excited state population using a linearly chirped pulse. In this Letter we demonstrate controlled transient enhancement of a resonant transition by pulse shaping. Transient enhancement of up to a factor of 2.5 was observed. Furthermore, we show that reshaping of the pulse by propagation in the resonant medium results in strong transient oscillations in the excited state population. We show that after propagating a transform-limited pulse in an absorbing medium, the peak transient population is nearly constant, even when the on-resonant intensity is reduced by orders of magnitude.

Consider the resonant interaction between an atomic system and a weak femtosecond laser pulse with electric field $\epsilon_1(t)$. First order time dependent perturbation theory predicts the temporal amplitude of the first excited state to be

$$a_1^{(1)}(t) = \frac{\mu_{1g}}{i\hbar}\int_{-\infty}^{t} dt_1\, \epsilon_1(t_1)\exp(i\omega_{1g}t_1) = \frac{-\mu_{1g}}{\hbar}\int_{-\infty}^{\infty} d\omega\, \frac{E_1(\omega)}{\omega_{1g}-\omega}\exp[i(\omega_{1g}-\omega)t], \tag{1}$$

where $\mu_{1g}$ is the dipole moment matrix element, $|g\rangle$ and $|1\rangle$, the ground and the first excited state, respectively, $E_1(\omega)$ the Fourier transform of $\epsilon_1(t)$, and $\omega_{1g}$ the transition frequency. The pulse duration is assumed to be considerably shorter than all lifetimes involved. At $t = 0$ the transition amplitude is given by

$$a_1^{(1)}(t=0) = \frac{-\mu_{1g}}{\hbar}\left[ i\pi E_1(\omega_{1g}) + \wp\int_{-\infty}^{\infty} d\omega\, \frac{E_1(\omega)}{\omega_{1g}-\omega}\right], \tag{2}$$

where $\wp$ is the principal value of Cauchy. The first term in Eq. (2) depends only on the spectral component of the pulse at the resonance frequency, whereas the second term integrates over the contributions of all the other spectral components of the pulse. As is evident from Eq. (2), the integrand of the "off-resonant" term undergoes a sign inversion about the resonance. When the atom is subjected to a transform limited pulse, a destructive interference is induced between the blue and the red detuned contributions. For any $E_1(\omega)$, which is symmetric around $\omega_{1g}$, total destructive interference is induced, and the transition amplitude depends only on the "on-resonant" component. Since the contribution of the off-resonant term is highly sensitive to the spectral phase function and its symmetry about the resonance, pulse shaping techniques can be applied to control the temporal transition amplitude [17]. This term can be highly enhanced by applying an antisymmetric phase function that inverts the sign of the spectral components about the resonance, inducing constructive interference and therefore maximizing the transient transition amplitude. The time interval at which this enhancement can be observed is of the order of the

  

unshaped (transform-limited) pulse duration. In order to measure the population transient, it is necessary to use a second femtosecond probe which excites the electron to a higher level $|2\rangle$ within that time interval, preserving the transient enhancement. The transition probability for the final excited state $|2\rangle$ at $t \to \infty$ as predicted by the second order time dependent perturbation theory is

$$P_2^{(2)}(\tau) = \left| \frac{1}{\hbar^2}\mu_{21}\mu_{1g}\int_{-\infty}^{\infty} dt_2\, \epsilon_2(t_2 - \tau)\exp(i\omega_{21}t_2) \times \int_{-\infty}^{t_2} dt_1\, \epsilon_1(t_1)\exp(i\omega_{1g}t_1) \right|^2 = \left| \frac{1}{\hbar}\mu_{21}\int_{-\infty}^{\infty} dt_2\, A_2(t_2 - \tau)a_1^{(1)}(t_2) \right|^2, \quad (3)$$

where $\mu_{21}$ is the dipole moment matrix element, $\epsilon_2(t) = A_2(t)\exp(-i\omega_{21}t)$, and $\tau$ is the time delay between the pump and the probe pulses. For a transform limited femtosecond probe pulse, Eq. (3) becomes a convolution, representing the transient first level population within the duration of the probe pulse. It is assumed there is no spectral overlap between the two pulses.

To demonstrate a transient population enhancement experimentally, we considered the $5S_{1/2}$-$5P_{1/2}$ transition in rubidium (Rb) gas. The $5P_{1/2}$ population is probed by a second excitation to the $4D_{3/2}$ level (Fig. 1a). The first transition is induced by a femtosecond pulse centered about 795 nm, with an average power of 40 mW, produced by a Ti:sapphire mode locked laser (Spectra Physics Tsunami, 80 MHz repetition rate). The probe beam is produced by an optical parametric oscillator (Spectra Physics Opal, pumped by the Ti:sapphire laser) centered about 1475 nm, with an average power of 50 mW. Both beams are nearly transform limited 100 fs pulses. The excited $4D_{3/2}$ atoms decay spontaneously to the ground level via both the $5P_{1/2}$ and the $5P_{3/2}$ levels. We evaluate the $4D_{3/2}$ population, representing the transient population of the $5P_{1/2}$ level, by measuring the fluorescence of the $5P_{3/2}$-$5S_{1/2}$ transition at 780 nm. The spectral phase of the pump pulse is controlled by Fourier-transform pulse shaping [18] using a programmable liquid crystal spatial light modulator (SLM). The shaper enables both correction of dispersion, as well as the application of any desired spectral phase function. The spectral resolution, determined by the spot size of a given frequency in the Fourier plane of the SLM is approximately 0.2 nm. The probe beam, which remains transform limited, is spatially overlapped with the pump beam using a dichroic beam splitter. The temporal delay is adjusted by a variable delay line. The overlapped beams are focused into a Rb cell with a 5 cm achromatic lens. The fluorescence signal is filtered by a narrow line filter at 780 nm, and measured with a photomultiplier tube (PMT) and a lock-in amplifier. An outline of the experimental setup is presented in Fig. 1b.

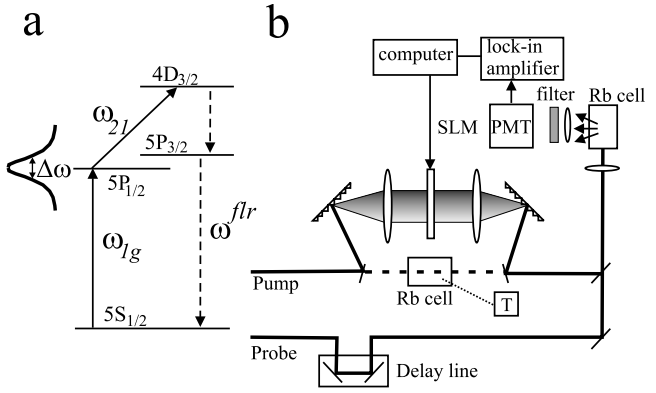


FIG. 1. (a) Energy level diagram of the pump-probe transitions in atomic Rb. (b) Outline of the experimental setup.

In Fig. 2 we plot the measured signal as a function of the probe delay for several pulse shapes. We first measure the signal induced by a transform limited pump pulse (solid line). Around zero delay the population increases within 300 fs, then slowly decays. Note that a constant nonzero signal was measured even at negative delays. This is related to the residual population at the $5P_{1/2}$ level, and results from the long lifetime of the electron at this level (25 ns) relative to the time separation of the pulse train (12 ns). We repeated the measurement with a shaped pump pulse, where a $\pi$-step phase function centered at the resonant frequency was applied (dashed line). This pulse shape maximizes the off-resonant term in Eq. (2). Since the phase inversion is located symmetrically about the resonance, the on-resonant component is almost completely diffracted at the Fourier plane of the pulse shaper. As the beam is collimated at the exit of the pulse shaper, the diffracted on-resonant component is blocked, and its contribution to the total absorption is negligible. Indeed, the observed signal at large probe delays, which depends only on the on-resonant term, is nearly zero. The observed signal is nearly symmetric about the zero delay point, as expected from Eq. (1), with a maximal enhancement of

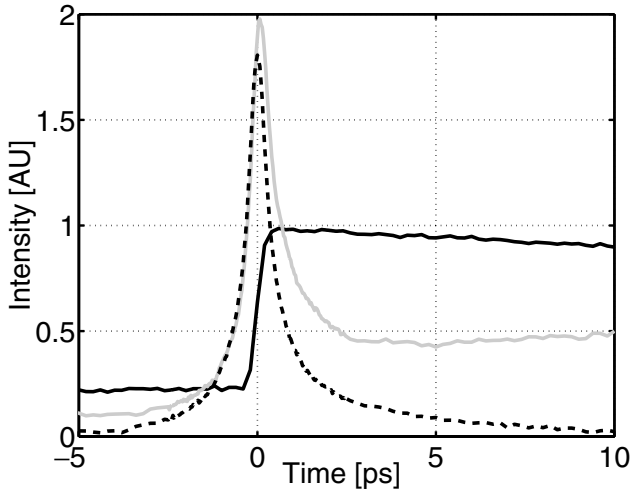


FIG. 2. Transient population measurement as a function of the probe delay for the pulse shapes: transform limited pump pulse (solid black line), on resonant $\pi$-step shaped pulse (dashed line), shifted $0.7\pi$ step (solid gray line).

almost 2.5 relative to the population jump in the transform limited case. To maximize the total transient signal, a $0.7\pi$ step, slightly shifted from the resonance, is applied (solid gray line). Thus, the on-resonant term is only partially diffracted and a 5% increase in the transient enhancement is achieved.

The spectral phase function generating the maximal transient enhancement is $\phi(\omega) = \arctan[(\omega - \omega_{1g})T_2]$, where $T_2$ is the inhomogeneous level lifetime. With this pulse shape, the theoretical limit to the enhancement at zero delay can be approximated from Eq. (2) to yield

$$\frac{\max(a_{SH}^{(1)})}{\max(a_{TL}^{(1)})} \approx \frac{1}{\pi}\ln(\Delta\omega T_2), \quad (4)$$

where $a_{SH}^{(1)}$, $a_{TL}^{(1)}$ are the transition amplitude of the shaped and transform limited pulses respectively, $\Delta\omega$ is the spectral bandwidth of the pulse. Ideally, this predicts a tenfold increase in the transient population for 100 fs pulses. In practice, however, the maximal enhancement is limited by the spectral resolution achievable by the pulse shaper [18], which is orders of magnitude worse than the atomic linewidth. Therefore $T_2$ in Eq. (4) is replaced by the inverse of the shaper spectral resolution.

Spectral manipulation with a resolution of the atomic linewidth is naturally induced by propagation in the resonant medium. The interaction of small-area pulses, such as those used in our experiments, with a resonant medium has been mathematically formulated by Crisp [19]. The reshaping of so called "$0\pi$" pulses has been demonstrated experimentally [20,21]. Here we concentrate on the coherent transients induced by such pulses. After propagating a distance $l$ through the medium, the electric field is

$$E_1(\omega, l) = E_1(\omega, 0) \cdot \exp\left[\frac{-\alpha_0 l}{1 - i(\omega - \omega_{1g})T_2}\right], \quad (5)$$

where $\alpha_0$ is the small signal amplitude absorption coefficient proportional to the atomic density. The dispersion of the absorption line thus induces a phase function $\phi(\omega) = -\alpha_0 l(\omega - \omega_{1g})T_2/[1 + (\omega - \omega_{1g})^2 T_2^2]$ on the pump beam. This, too, describes an antisymmetric phase function. In contrast with the optimal phase function, it decays as $1/(\omega - \omega_{1g})$. The maximal phase jump across the resonance is $\alpha_0 l$. As long as $\alpha_0 l < \pi$ the phase induced by the dispersion moves towards the optimal phase function for all the off-resonant components. For larger values, alternate spectral regions of destructive and constructive interference with the on-resonant term appear. The number of these regions increases linearly with $\alpha_0 l$. At zero delay between the pump and probe pulses, all these regions contribute to the transient population. As the delay is increased, the bandwidth contributing to the transient population diminishes, containing fewer regions. Since different spectral regions contribute to the population with alternating signs, oscillations in the transient population are found for large values of $\alpha_0 l$. Such oscillations have been recently observed in a TPA experiment [22].

A careful calculation of the population at zero delay shows that independent of the propagation length, the absorption of the on-resonant component is exactly canceled by the enhancement due to the off-resonant terms. Surprisingly, even when the resonant spectral component of the pulse is totally absorbed, the population at zero delay remains nearly constant. The peak transient population decreases only when the spectral width required to compensate for absorption is larger than the pulse spectral bandwidth.

To demonstrate propagation effects on the transient population, we replaced the pulse shaper by a second Rb cell. The absorption coefficient is varied by changing the cell temperature. The fluorescence signal as a function of the probe delay is illustrated in Fig. 3 for several values of $\alpha_0 l$, along with theoretical curves calculated by Eqs. (1) and (5). The population at late times, best observed at short "negative" delays, decreases rapidly as the temperature is increased due to the larger absorption of the on-resonant term. The population jump at zero delay is nearly constant, although $\alpha_0 l$ is increased by almost 3 orders of magnitude (Figs. 3a–3c), and then

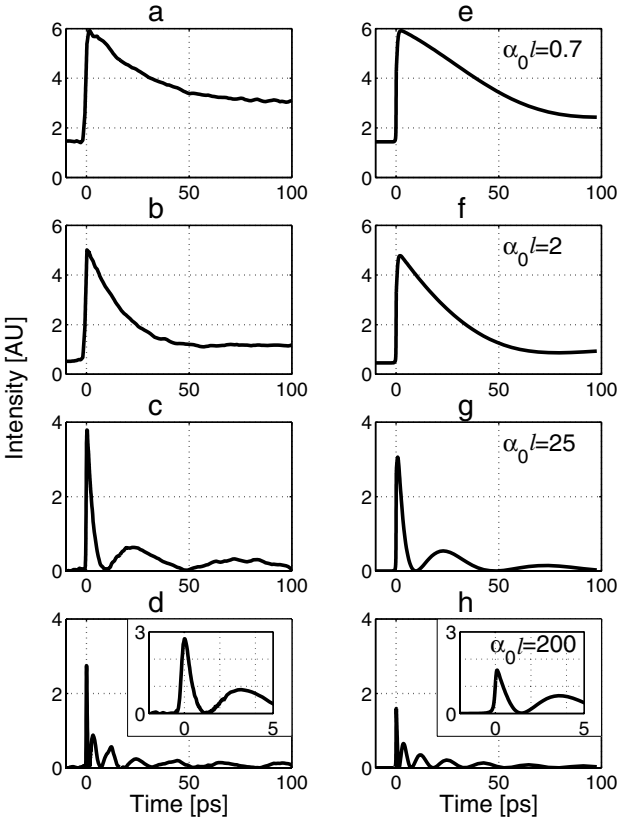


FIG. 3. Transient population measurements (a)–(d) and calculated results (e)–(h) as a function of the probe delay for unshaped pulses, showing the effect of linear dispersion in an absorbing medium. The insets in (d) and (h) zoom in on the first rapid oscillation. Four values of the absorption coefficient $\alpha_0 l$, were used in the respective calculations, as indicated.

decreases slightly for $\alpha_0 l \approx 200$ (Fig. 3d). At $\alpha_0 l \approx 25$ (Fig. 3c) oscillations in the transient population appear. Each oscillation is related to a $\pi$ cross of the spectral phase function, as determined by the dispersion. As $\alpha_0 l$ is increased (Fig. 3d), more rapid oscillations are observed. The first oscillation lasts about 500 fs (see inset of Fig. 3d), indicating that a significant part of the pulse spectrum contributes to the transient population. The calculated values (plotted in Figs. 3e–3h) are in excellent agreement with the experimental results.

In conclusion, we have shown that the transient population of a resonant transition can be enhanced. A transform limited pulse induces a destructive interference of the contributions of off-resonant spectral components. Using pulse shaping techniques, we were able to enhance the total transient population, by inducing constructive, rather than destructive, interference. For 100 fs pulses, an enhancement by a factor of about 2.5 was demonstrated. This enhancement is a general phenomenon typical of a passage through a resonant transition and has been recently demonstrated in a coherent anti-Stokes Raman process [23]. We have also shown that the natural reshaping of the pulse by linear dispersion of an absorption line also leads to rapid transient population oscillations. We have shown that the population transient peak remains nearly constant even when the absorption coefficient is increased by over 3 orders of magnitude. We conclude that narrow transitions induced by broadband pulses offer a rich spectrum of transient phenomena which has not been realized previously.

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