Two Photon Absorption and Coherent Control with Broadband Down-Converted Light

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We experimentally demonstrate two-photon absorption with broadband down-converted light (squeezed vacuum). Although incoherent and exhibiting the statistics of a thermal noise, broadband down-converted light can induce two-photon absorption with the same sharp temporal behavior as femtosecond pulses, while exhibiting the high spectral resolution of the narrow band pump laser. Using pulse-shaping methods, we coherently control two-photon absorption in rubidium, demonstrating spectral and temporal resolutions that are 3–5 orders of magnitude below the actual bandwidth and temporal duration of the light itself. Such properties can be exploited in various applications such as spread-spectrum optical communications, tomography, and nonlinear microscopy.

In two-photon absorption (TPA), two photons whose sum energy equals that of an atomic transition must arrive at the atom together. The two, seemingly contradicting, demands of a narrow temporal and a narrow spectral behavior of the inducing light are typically maximized by transform-limited pulses, which exhibit the highest peak intensity possible for a given spectral bandwidth. Nonetheless, it was shown [1,2] that pulses can be shaped in a way that will stretch them temporally yet will not affect the transition probability, and even increase it in certain cases [3]. Other experiments have exploited coherent control [4–7] to increase the spectral selectivity of nonlinear interactions induced by ultrashort pulses [8–10]; however, the spectral resolution demonstrated by these methods remains considerably inferior to that obtained by narrow band, continuous lasers. A few experiments [11,12] have performed TPA with coherent, narrow band down-converted light, demonstrating nonclassical features which appear at very low powers [13–15] and result from the time and energy correlations (entanglement) between the down-converted photon pairs [16–18]. At high power levels (as those discussed in this work) these correlations vanish, yet similarly nonclassical phase and amplitude correlations [19,20] appear between the signal and idler beams. Unlike the time and energy correlations at low powers, these phase and amplitude correlations cannot be described in the usual form of second-order coherence. At sufficiently high powers, which greatly exceed the single-photons regime, broadband down-converted light that is pumped by a narrow band laser is inherently incoherent and exhibits the properties of a broadband thermal noise [16,21]; consequently, it is not expected to be effective at inducing TPA. Nevertheless, it was shown that at the appropriate conditions, the quantum correlations within the down-converted spectrum can give rise to efficient sum-frequency generation [22,23].

In this work we show that down-converted light beams with a spectral bandwidth that significantly exceeds that of the pump can induce TPA just like transform-limited pulses of the same bandwidth. Consequently, the interaction exhibits a sharp, ultrashort pulse-like temporal behavior and can be coherently controlled by pulse-shaping techniques, even though the down-converted light beams are neither coherent nor pulsed [24]. This effect occurs as long as the transition energy lies within the spectrum of the pump laser that generated the light; thus, the spectral selectivity of the interaction is dictated by the narrow band pump laser and not by the orders of magnitude wider bandwidth of the down-converted light itself. We demonstrate these principles experimentally by inducing and coherently controlling TPA in atomic rubidium with down-converted light and by obtaining results that are practically identical to those obtained with coherent ultrashort pulses.

The underlying principle that enables coherent TPA with broadband down-converted light is based on the fact that the quantum interference that governs TPA involves pairs of electromagnetic modes. Since the excitation of an atomic level with frequency Ω may be induced by any two photons with frequencies ω and $\Omega - \omega$, regardless of the exact value of ω, the final population p_f is proportional to [1]

$$p_f \propto \left| \int E(\omega)E(\Omega - \omega)d\omega \right|^2, \qquad (1)$$

where $E(\omega)$ is the spectral amplitude of the light. As is obvious from Eq. (1), the individual phases of the spectral modes do not matter; only the phase of the product $E(\omega)E(\Omega - \omega)$ is important. Despite the incoherence of each of the down-converted signal and idler beams, they exhibit exactly this mutually coherent phase behavior at frequency pairs, due to the inherent phase and amplitude quantum correlations within the down-converted

spectrum [19]:

$$\frac{1}{\sqrt{\omega}} E_s(\omega) \approx \frac{1}{\sqrt{\omega_p - \omega}} E_i^*(\omega_p - \omega), \qquad (2)$$

where ω_p is the pump frequency and $E_s(\omega)$, $E_i(\omega)$ denote the spectral amplitudes of the signal and the idler beams, respectively. In the case of TPA induced by the combined signal and idler beams from the same source, these correlations have a drastic effect when the pump frequency is equal to the total transition frequency. Combining Eqs. (1) and (2), letting $\omega_p = \Omega$, reveals that the random phase of $E_s(\omega)$ is always compensated by the opposite phase of $E_i(\omega_p - \omega)$. Thus, the integrand in Eq. (1) has a constant phase, leading to a full constructive interference of all the spectral combinations, exactly as if the interaction was induced by a pair of transform-limited pulses with the same spectra as the signal and idler beams. Moreover, this TPA process will be sensitive to minute delays between the signal and idler beams, to dispersion, and even to pulse shaping, exactly as if it was induced by a pair of ultrashort pulses (this principle holds for sum-frequency generation as well, when the final wavelength is equal to that of the pump [23]). Since this constructive interference occurs only when the final state energy falls within the spectrum of the pump laser, the spectral resolution of the TPA process will be equal to the spectral bandwidth of the pump laser, regardless of the actual bandwidth of the down-converted light itself. Note that in the case of a continuous pump laser, this implies a possible spectral resolution of a few MHz or even less, which is phenomenally high for an interaction that is induced by light with a spectral bandwidth that may be 5–7 orders of magnitude broader.

A complete quantum-mechanical analytic calculation [25], which takes into account the spectral band-widths of the pump laser and of the atomic level, shows that the TPA signal is composed of two parts, which may be referred to as the "coherent TPA" and the "incoherent TPA." The coherent TPA results from the summation of conjugated spectral components and indeed can be coherently controlled. The incoherent TPA, however, results from the summation of all other random spectral combinations and is a direct result of the incoherence of the down-converted light; therefore, it is unaffected by spectral-phase manipulations and may be regarded as a background noise, which limits the equivalence of the down-converted light to a coherent pulse. The ratio between the coherent term I^c and the incoherent term I^{ic} can be approximated by

$$\frac{I^c}{I^{ic}} \approx \frac{B}{(\gamma_p + \gamma_f)} \frac{n^2 + n}{n^2}, \qquad (3)$$

where n is the spectral average of the mean photon flux, and B, γ_p, γ_f are the bandwidths of the down-converted light, the pump laser, and the final state, respectively. This expression reveals the importance of using spectrally broad down-converted light. The coherent term becomes dominant only when the down-converted bandwidth exceeds both the pump bandwidth and the final level width: $B > (\gamma_p + \gamma_f)(\frac{n^2}{n^2+n})$. Equation (3) shows that the coherent term exhibits a linear intensity dependence at low powers, as was previously predicted [13,14] and experimentally observed [11].

Our experimental layout is described in Fig. 1. The pump laser (Spectra-Physics MOPO-SL laser) was tuned to emit 3 ns pulses with a bandwidth of 0.04 nm around 516.65 nm, which corresponds to the $5S - 4D$ transition in Rb [Fig. 1(a)]. These pulses pumped a BBO crystal that was located inside a low-finesse resonator, with the phase-matching conditions chosen to obtain broadband
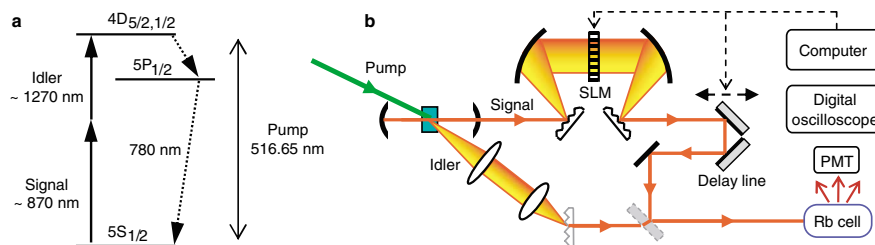

FIG. 1 (color online). (a) Atomic energy levels of Rb, showing the induced two-photon transition and the resulting cascaded decay. (b) The experimental layout; 3 ns pulses at 516.65 nm were used to pump a 14 mm long beta barium borate (BBO) crystal in a low-finesse resonator. The relative angles between the crystal axis, the pump laser, and the resonator were chosen to obtain broadband phase matching for noncollinear type-I down-conversion. Unlike the signal beam, whose direction of propagation was set by the resonator, the idler beam had a wavelength-dependent angular spread, due to the phase matching conditions in the crystal. Therefore, it was directed through an imaging system that included a transmission grating which compensated for this spread. The signal beam was directed through a pulse shaper and a computer-controlled delay line with 0.1 μ resolution and then combined with the idler beam by a dichroic mirror. The pulse shaper separates the spectral components of the beam and utilizes a computer-controlled spatial light modulator (SLM) to introduce the desired spectral phase filter to the light. A photomultiplier tube (PMT) and a digital oscilloscope were used to perform a triggered measurement of the TPA-induced fluorescence.

(~100 nm each), noncollinear signal and idler beams. The signal beam was directed through a computer-controlled pulse shaper, which is normally used to temporally shape femtosecond pulses and performs as a spectral-phase filter [26]. From the pulse shaper the signal beam continued to a computer-controlled delay line and then was combined with the idler beam by a dichroic mirror. The combined beams entered the Rb cell, and the induced TPA was measured through the resulting fluorescence at 780 nm. Our analysis predicts that the down-converted beams, each 3 ns long with a peak power of about 1 MW and a spectral bandwidth of about 100 nm, should induce TPA with the same efficiency and temporal resolution as 23 fs pulses with a peak power of about 150 GW, while exhibiting a spectral resolution of 0.04 nm.

To verify these predictions experimentally, we scanned both the pump wavelength and the relative delay between the signal and idler beams. Figure 2 shows the TPA signal versus the signal-idler relative delay and the pump wavelength. As is evident, for off-resonance pump or for large signal-idler delays, only low efficiency TPA signal is observed, which is insensitive to either pump wavelength or signal-idler delay. This signal corresponds to the classical, incoherent TPA background signal. However, when
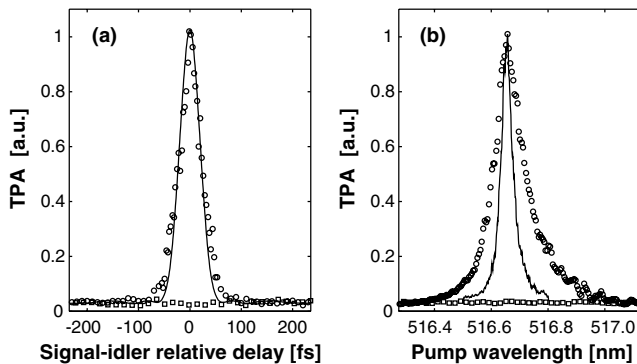

FIG. 2. Experimental TPA with broadband down-converted light, as a function of the signal-idler delay and the pump wavelength. The graphs clearly show a coherent TPA signal that is narrow both temporally and spectrally, superimposed on an incoherent background signal, which is insensitive to both pump wavelength and signal-idler relative delay. In this experiment the pulse shaper was used only to compensate for the dispersion in our system. (a) Calculated (line) and experimental TPA with off-resonance (squares) and on-resonance (circles) pump, as a function of the signal-idler delay. (b) Experimental TPA at zero signal-idler delay (circles), and at 100 fs signal-idler delay (squares), as a function of the pump center wavelength, together with a typical spectrum of the pump (line). The coherent TPA signal appears only when the pump is on-resonance with the $5S - 4D$ transition, and exhibits a sharp dependence on the signal-idler delay, exactly as if the interaction was induced by a pair of coherent, 23 fs pulses with the same spectra as the signal and idler beams.

the pump wavelength was set to the $5S - 4D$ transition resonance, an additional, high efficiency, TPA signal appeared. The sharp response of this coherent TPA signal to a few femtoseconds delay between the beams is practically identical to the case of TPA induced by two coherent, 23 fs pulses. This temporal resolution is 5 orders of magnitude better than the actual 3 ns temporal duration of the down-converted light.

Since the peak power of the equivalent transform-limited pulses is extremely high (150 GW), the signal and idler beams had to be expanded and attenuated in order to avoid complete saturation of the transition. Unfortunately, we could not attenuate the beams enough to avoid saturation completely, due to high levels of noise in our system. As a result, our measurements had to be performed in a partially saturated regime, where the measured intensity dependence of the TPA process was less than quadratic, and the $4D$ level was power broadened. Thus, the observed 0.12 nm spectral width of the coherent TPA is dictated by the bandwidth of the pump laser (0.04 nm) and the width of the (power-broadened) $4D$ level (~0.08 nm). This spectral resolution is 2000 times narrower than the total bandwidth of the down-converted light.

Finally, we demonstrated quantum coherent control over the coherent TPA process, in a similar way to coherent control of TPA with ultrashort pulses [1]. For that we used the pulse shaper to apply a square-wave phase filter on the signal spectrum [Fig. 3(a)]. With ultrashort pulses this has the effect of splitting the pulse temporally to a train of several smaller pulses. Figure 3(b) shows precisely this behavior of the coherent TPA signal as a function of the signal-idler delay. Figure 3(c) depicts the experimental and theoretical TPA signal at zero delay as a function of the magnitude of the square wave phase filter. The results, which are identical to those obtained in coherent control experiments with femtosecond pulses, show the cyclic transition between complete constructive and destructive quantum interference of the different spectral sections and demonstrate the ability to fully control the coherent TPA process.

Broadband down-conversion of a narrow band pump can be considered as an optical spread-spectrum source, which generates both a broadband white noise key and its conjugate simultaneously [27]. A spread-spectrum communication channel can therefore be established by modulating the phase of one of the keys at the transmitter and using TPA or sum-frequency generation to reveal the resulting modulations of the coherent signal at the receiver. Note that any phase modulations performed on the incoherent signal beam (or even the mere presence of the signal beam, in the existence of background noise) cannot be detected without the idler beam. Furthermore, many such communication channels can share the same signal and idler beams by assigning a unique signal-idler delay for each channel, thus creating an optical code division
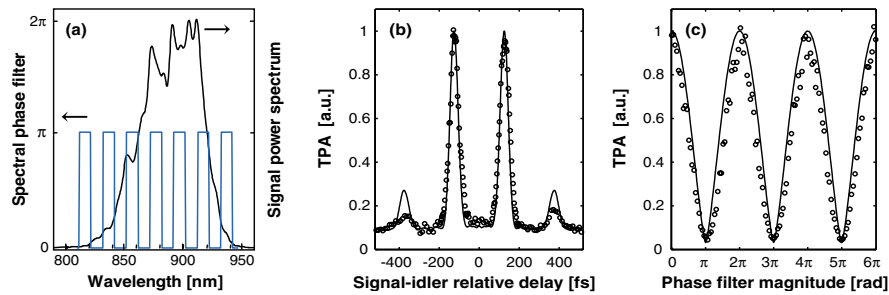
FIG. 3 (color online).   Quantum coherent control of TPA with broadband down-converted light. (a) The square-wave spectral phase filter applied on the signal beam, together with a typical power spectrum of the signal. (b) Experimental (circles) and calculated (line) TPA signal as a function of the signal-idler delay, using the spectral phase filter described in (a). The splitting of the central peak confirms that the coherent TPA behaves exactly as if it was induced by a pair of transform-limited pulses, with one of them temporally shaped by the applied phase filter. (c) Experimental (circles) and calculated (line) TPA signal as a function of the magnitude of the square wave spectral phase filter. The graph shows the periodic annihilation of the coherent TPA at magnitudes of $\pi, 3\pi, 5\pi$, where a complete destructive quantum interference occurs ("dark pulse"), and the complete reconstruction of the TPA signal at amplitudes of $2\pi, 4\pi, 6\pi$, etc.

multiple access network [27]. Other applications may include nonlinear microscopy and tomography, where the high efficiency and spatial resolution of ultrashort pulses can be obtained with continuous, nondamaging intensities.

[1] D. Meshulach and Y. Silberberg, Nature (London) **396**, 239 (1998); Phys. Rev. A **60**, 1287 (1999).

[2] Z. Zheng and A. M. Weiner, Chem. Phys. **267**, 161 (2001).

[3] N. Dudovich, B. Dayan, S. M. G. Faeder, and Y. Silberberg, Phys. Rev. Lett. **86**, 47 (2001).

[4] D. J. Tannor and S. A. Rice, J. Chem. Phys. **83**, 5013 (1985).

[5] M. Shapiro and P. Brumer, J. Chem. Phys. **84**, 4103 (1986).

[6] W. S. Warren, H. Rabitz, and M. Dahleh, Science **259**, 1581 (1993).

[7] S. A. Rice and M. Zhao, *Optical Control of Molecular Dynamics* (Wiley, New York, 2000).

[8] T. Lang, M. Mutzkus, H. M. Frey, and P. Beaud, J. Chem. Phys. **115**, 5418 (2001).

[9] N. Dudovich, D. Oron, and Y. Silberberg, Nature (London) **418**, 512 (2002); Phys. Rev. Lett. **92**, 103003 (2004).

[10] E. Gershgoren *et al.*, Opt. Lett. **28**, 361 (2003).

[11] N. Ph. Georgiades *et al.*, Phys. Rev. Lett. **75**, 3426 (1995).

[12] N. Ph. Georgiades, E. S. Polzik, and H. J. Kimble, Phys. Rev. A **55**, R1605 (1997).

[13] I. Gea-Banacloche, Phys. Rev. Lett. **62**, 1603 (1989).

[14] J. Javanainen and P. L. Gould, Phys. Rev. A **41**, 5088 (1990).

[15] H. B. Fei, B. M. Jost, S. Popescu, B. E. A. Saleh, and M. C. Teich, Phys. Rev. Lett. **78**, 1679 (1997).

[16] L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics* (Cambridge University Press, Cambridge, 1995).

[17] D. C. Burnham and D. L. Weinberg, Phys. Rev. Lett. **25**, 84 (1970).

[18] C. K. Hong, Z. Y. Ou, and L. Mandel, Phys. Rev. Lett. **59**, 2044 (1987).

[19] B. R. Mollow and R. J. Glauber, Phys. Rev. **160**, 1097 (1967).

[20] K. J. McNeil and C. W. Gardiner, Phys. Rev. A **28**, 1560 (1983).

[21] M. O. Scully and S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, 1997).

[22] I. Abram, R. K. Raj, J. L. Oudar, and G. Dolique, Phys. Rev. Lett. **57**, 2516 (1986).

[23] B. Dayan, A. Pe'er, A. A. Friesem, and Y. Silberberg, quant-ph/0302038.

[24] The case considered here is of broadband down-conversion generated by a narrow band pump; thus, the down-converted light may practically be considered as continuous compared to an ultrashort pulse of a similar bandwidth, even in cases where the pump laser (and hence also the down-conversion) is pulsed.

[25] B. Dayan and Y. Silberberg (to be published).

[26] A. M. Weiner, Rev. Sci. Instrum. **71**, 1929 (2000).

[27] A. Pe'er, B. Dayan, Y. Silberberg, and A. A. Friesem, J. Lightwave Technol. **22**, 1463 (2004).