

Coherent control with broadband squeezed vacuum

Barak Dayan,^{*} Avi Pe'er,[†] Asher A. Friesem, and Yaron Silberberg[‡]
*Department of Physics of Complex Systems,
 Weizmann Institute of Science,
 Rehovot 76100, Israel*

We report the experimental demonstration of coherent control with high power, broadband squeezed vacuum. Although incoherent and exhibiting the statistics of a thermal noise, broadband squeezed vacuum is shown to induce certain two-photon interactions as a coherent ultrashort pulse with the same spectral bandwidth. Utilizing pulse-shaping techniques we coherently control the sum-frequency generation of broadband squeezed vacuum over a range of two orders of magnitude. Coherent control of two-photon interactions with broadband squeezed vacuum can potentially obtain spectral resolutions and extinction ratios that are practically unattainable with coherent pulses.

PACS numbers: 32.80.Qk, 42.50.Dv, 42.65.Ky, 42.65.Lm, 42.65.Re

The quantum mechanical expressions for broadband two-photon interactions clearly reflect the underlying spectral quantum interference. For example, the final population of an atomic level with energy Ω , due to a two-photon absorption (TPA) induced by light with spectral amplitude $E(\omega)$ is [1]:

$$p_f \propto \left| \int E(\Omega/2 + \xi)E(\Omega/2 - \xi) d\xi \right|^2 \quad (1)$$

This expression exhibits quite directly the fact that TPA may occur by photon pairs with energies $\Omega/2 + \xi$ and $\Omega/2 - \xi$, for any ξ within the bandwidth of the light. For coherent light, $E(\omega)$ has a defined spectral phase: $E(\omega) = A(\omega) \exp[i\Theta(\omega)]$. Consequently, the final population can be controlled by applying a spectral phase filter $\Phi(\omega)$ to the light, making the quantum interference constructive or destructive, as desired:

$$p_f \propto \left| \int A(\Omega/2 + \xi)A(\Omega/2 - \xi) \right. \\ \times \exp[i\Theta(\Omega/2 + \xi) + i\Theta(\Omega/2 - \xi)] \\ \times \left. \exp[i\Phi(\Omega/2 + \xi) + i\Phi(\Omega/2 - \xi)] d\xi \right|^2 \quad (2)$$

Typically, pulses with a constant spectral phase (transform-limited pulses) maximize nonresonant interactions, as becomes evident if we let $\Theta(\omega) = \Phi(\omega) = 0$ in Eq. (2). The technique of coherent control [2, 3, 4, 5] usually exploits pulse-shaping techniques [6] to apply a spectral phase filter to mode-locked ultrashort pulses, in order to manipulate the quantum interference and steer multiphoton interactions towards desired states. In particular, it was demonstrated [7] that applying a spectral phase filter that is anti-symmetric about $\Omega/2$ does not affect the TPA probability, although it may significantly stretch the pulse and lower its peak power. The simple reason for this effect is that when $\Phi(\Omega/2 + \xi) = -\Phi(\Omega/2 - \xi)$, opposite phases are applied to the complementary modes in all the mode-pairs that

contribute to the TPA process. Thus, as is evident from Eq. (2), the overall phase contribution of the phase filter is cancelled out, leaving the efficiency equal to that of a transform-limited pulse. Nonetheless, a symmetric phase filter does effect the TPA probability, and can even eliminate it. Similar coherent control was demonstrated over sum-frequency generation (SFG) with ultrashort pulses, establishing that SFG in thick nonlinear crystals may be considered equivalent (in the perturbative limit) to TPA [8, 9].

Here we establish that coherent control can be performed with incoherent, broadband squeezed vacuum, generated by a parametric down-conversion [10, 11] of a narrowband pump laser in a nonlinear crystal. We show that squeezed vacuum with a spectral bandwidth that exceeds a certain limit induces TPA and SFG just like a coherent ultrashort pulse with the same spectral bandwidth. This effect occurs as long as the final state energy lies within the spectrum of the pump laser that generated the squeezed vacuum. Consequently, such interactions can be coherently controlled by pulse-shaping techniques, even though the squeezed vacuum is neither coherent nor pulsed, but rather is incoherent and exhibits the properties of a broadband thermal noise [10, 12]. We prove this principle experimentally by coherently controlling the SFG signal induced by high-power broadband squeezed vacuum, and obtaining similar results to those obtained with coherent ultrashort pulses.

The underlying principle of our coherent control scheme is that while coherence of all the spectral paths is indeed required for coherent control, it does not necessarily imply coherence of the inducing light. Since the quantum interference that governs two-photon interactions always involves pairs of photons, it requires the photon-pairs to be coherent. In other words, as is obvious from Eq. (1), it does not matter whether $E(\omega)$ has a defined phase for every ω , but rather whether the product $E(\Omega/2 + \xi)E(\Omega/2 - \xi)$ has a defined phase for every ξ . Although broadband squeezed vacuum has a random spec-

tral phase, it does exhibit exactly this phase behavior at frequency-pairs due to the inherent quantum correlations within its spectrum. The two-mode squeezing which occurs during the down-conversion process leads to complete amplitude correlations and phase anti-correlations between the spectral-components of $E(\omega)$ at complementary frequency-pairs that sum to the pump frequency [13, 14]:

$$\begin{aligned} \frac{1}{\sqrt{\omega_p/2+\xi}} A(\omega_p/2+\xi) &\sim \frac{1}{\sqrt{\omega_p/2-\xi}} A(\omega_p/2-\xi) \\ \Theta(\omega_p/2+\xi) &\sim \pi/2 - \Theta(\omega_p/2-\xi) \end{aligned} \quad (3)$$

These correlations drastically affect the quantum interference that governs two-photon interactions. Generally, the expected efficiency of a broadband thermal noise at inducing nonlinear interactions is extremely low, and usually ultrashort pulses with high peak-powers must be used. However, for TPA (or SFG) with down-converted light, the situation is strikingly different when the final state energy is equal to the pump frequency. As becomes clear if one combines equations 2 and 3 taking $\Omega = \omega_p$, the random phase of $E(\Omega/2 + \xi)$ is always compensated by the opposite phase of $E(\Omega/2 - \xi)$. Thus, the overall phase contribution of every mode pair is cancelled out, and the integrand of Eq. (2) is affected only by the phase-filter $\Phi(\omega)$, exactly as if the interaction was induced by a transform-limited pulse with the same spectrum:

$$\begin{aligned} p_f \propto &\left| \int A(\Omega/2 + \xi) A(\Omega/2 - \xi) \right. \\ &\times \left. \exp[\Phi(\Omega/2 + \xi) + \Phi(\Omega/2 - \xi)] d\xi \right|^2. \end{aligned} \quad (4)$$

Naturally, the cancelling out of the phase of the squeezed light resembles the cancelling out of an anti-symmetric phase manipulation on a coherent ultrashort pulse. This similarity may be further clarified by considering cw-pumped broadband squeezed vacuum as similar to a classical ultrashort pulse, which has undergone a spectral phase manipulation that is random at every mode, but antisymmetric about $\frac{\omega_p}{2}$. Such a hi-resolution phase manipulation, which is practically unattainable by pulse-shaping techniques, will stretch the pulse to a continuous wave. Consequently, the pulse's ability to induce TPA or SFG will be reduced drastically, except for final states with energy that is exactly ω_p , where the full efficiency of the process will remain unaffected. This constructive interference occurs only when the final state energy falls within the spectrum of the pump laser. As was indeed observed by Abram *et al.* [15], this implies that the spectrum of the SFG signal actually reproduces the spectrum of the pump laser. Note that in the case of a single-frequency pump laser, this implies a possible spectral resolution of a few KHz. Such a spectral

resolution is phenomenally high for an interaction that is induced by light with spectral bandwidth that is typically many orders of magnitude wider.

A full analytic quantum mechanical calculation [16] reveals a somewhat more complicated behavior. The SFG or TPA signal at the pump frequency is composed of two parts, which may be referred to as the 'quantum term' and the 'classical term': $I_\Omega = I_\Omega^c + I_\Omega^q$. While the quantum term results from the coherent summation of conjugated spectral components, the classical term results from the incoherent summation of all random spectral combinations. The quantum term is therefore the one that is equivalent to a coherent pulse and can be coherently controlled. The classical term, however, is a direct result of the incoherence of the down-converted light and therefore it is unaffected by spectral-phase manipulations. Thus, it may be regarded as an incoherent background noise, which limits the equivalence of the down converted light to a coherent pulse.

The ratio between the quantum term and the classical term can be approximated by:

$$\frac{I_\Omega^q}{I_\Omega^c} \approx \frac{B}{2(\gamma_p + \gamma_f)} \frac{n^2 + n}{n^2}, \quad (5)$$

where n is the spectral average of the mean photon flux, and B, γ_p, γ_f are the bandwidths of the squeezed vacuum, the pump laser and the final state respectively (in the case of SFG γ_f represents the spectral resolution of the measurement). The factor of 2 results from the assumed collinear configuration. This expression reveals the importance of using spectrally broad down-converted light. The quantum term becomes dominant only when the down-converted bandwidth exceeds both the pump bandwidth and the spectral resolution of the measurement: $B > 2(\gamma_p + \gamma_f)(\frac{n^2}{n^2+n})$. Moreover, Eq (5) shows that the quantum term exhibits a linear intensity dependence at low powers, as was indeed observed by Georgiades *et al.* [17].

To experimentally demonstrate these principles we used a programmable pulse-shaper to apply a spectral phase filter to broadband (60 nm centered at 1064 nm) down-converted light, emitted from a nonlinear PPKTP crystal pumped by spectrally narrow (≈ 0.01 nm) 8-ns pulses at 532nm (Fig.). The light was directed from the pulse-shaper to a second PPKTP crystal, and the resulting SFG signal was measured by a spectrometer with a spectral resolution of 0.03 nm. Our calculations show that the behavior of the SFG signal in this scheme is equivalent to TPA with final level broadening equal to the spectral resolution of the spectrometer.

Figures 2 and 3 show the experimental and the calculated results of our coherent control experiments

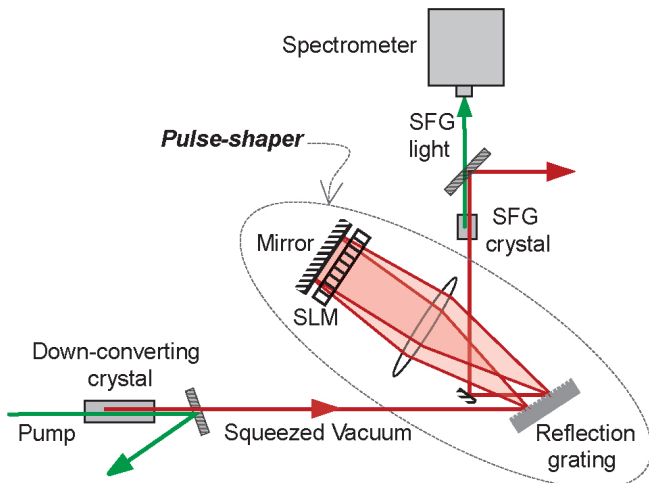


FIG. 1: Experimental system for coherent control of SFG with broadband squeezed vacuum. Down-converted light with spectral bandwidth of 60 nm around 1064 nm is generated in a 9 mm long periodically-poled KTP nonlinear crystal pumped by 1 mJ, 8-ns pulses at 532 nm from a doubled Q-switched Nd:YAG laser. The remainder of the pump beam is removed after the crystal by a filter. The squeezed vacuum then undergoes spectral phase manipulations in a folded pulse-shaper that is composed of a reflection grating, a lens, a 128-element liquid-crystal spatial light-modulator (SLM) and a mirror. In this configuration, a single set of reflection grating and lens is used both to image the different spectral components of the incoming beam on the phase elements of the SLM, and to reassemble those components when they are reflected back by the mirror behind the SLM. The output beam is directed to a second, 2 mm long, periodically-poled KTP crystal, where the SFG process occurs. The SFG signal is then measured by a spectrometer with 0.03 nm spectral resolution.

with high-power broadband squeezed vacuum. Figure 2(a) depicts the measured (circles) and calculated (line) spectrum of the SFG signal on a logarithmic scale, obtained without any spectral phase manipulation, save for dispersion compensation. In this measurement the pulse shaper was set only to compensate for the overall dispersion of the system. The graph clearly shows the remarkable difference between the quantum peak at 532nm, which is as narrow as our spectral resolution, and the broad, two orders of magnitude lower, classical background.

To demonstrate coherent control of the SFG process we applied a linear phase function to the lower half of the spectrum, and a linear phase function with opposite slope to the higher half of the spectrum: $\Phi(\omega) = \tau |\omega - \omega_p/2|$. Such a phase function is equivalent to delaying one half of the spectrum by 2τ relative to the other. Figure 2(b) shows the measured SFG signal at 532 nm as a function of the overall delay in fs. The rapid

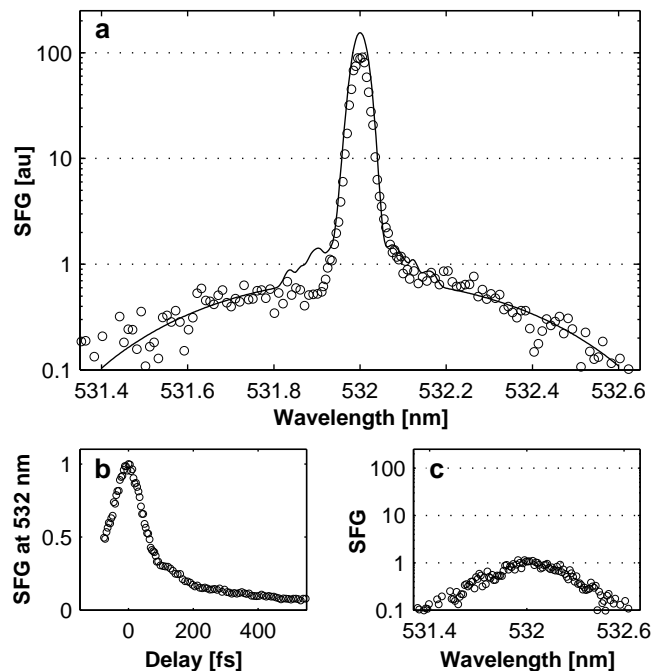


FIG. 2: Coherent control of SFG with broadband squeezed vacuum by a linear spectral phase filter that is equivalent to a delay between the higher and lower halves of the spectrum. (a) Experimental (circles) and calculated (line) SFG spectrum without any delay. In this measurement the pulse shaper was set only to compensate for the dispersion of the optical system. (b) The measured SFG signal at 532 nm as a function of the equivalent delay between the two spectral halves of the down-converted light. (c) The experimental SFG spectrum at an equivalent delay of 1.5 ps, where the quantum term is completely suppressed.

decay in the signal verifies the predicted equivalence of the down-converted light to an ultrashort pulse of ~ 65 fs, despite the fact that the actual duration of the down-converted pulse is 5 orders of magnitude longer. Figure 2(c) shows the SFG spectrum at an equivalent delay of 1.5 ps, where the quantum term is completely suppressed, leaving only the unaffected classical background.

Next, we applied a sinusoidal phase filter $\Phi(\omega) = \alpha \sin(\beta(\omega - \frac{1}{2}\omega_p) + \theta)$ to the down-converted spectrum. When $\theta = 0, \pm\pi, \pm2\pi, \dots$ this function is anti-symmetric about $\frac{1}{2}\omega_p$ and therefore, as noted earlier, it does not affect the SFG signal at ω_p . On the other hand, for $\theta = \pm\frac{1}{2}\pi, \pm\frac{3}{2}\pi, \dots$ the filter is a symmetric function about $\frac{1}{2}\omega_p$, and with the appropriate values of α and β it is expected to completely suppress the quantum part of the SFG signal. Figure 3 depicts the measured (circles) and the calculated (line) SFG signal at 532 nm as a function of the phase θ , showing the expected periodic reconstruction of the full signal at $\theta = 0, \pm\pi, \pm2\pi$, and a suppression of the signal at

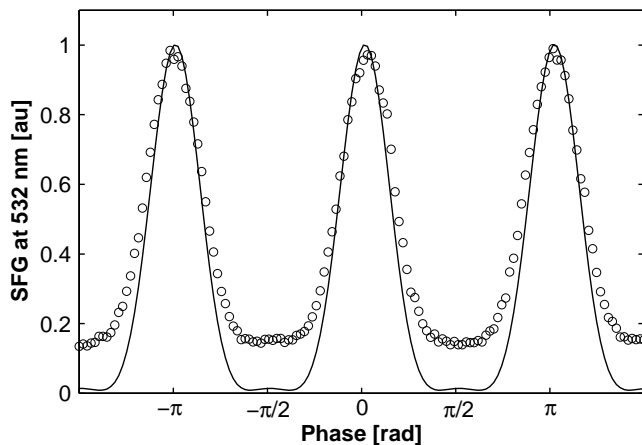


FIG. 3: Coherent control of SFG with broadband squeezed vacuum by a periodic spectral phase function $\Phi(\omega) = \alpha \sin(\beta(\omega - \frac{1}{2}\omega_p) + \theta)$. Experimental (circles) and calculated (line) SFG signal at 532 nm as a function of the phase θ . The signal remains unaffected when $\Phi(\omega)$ is anti-symmetric about $\omega_p/2$, and suppressed elsewhere. The values of α and β were chosen to maximize the suppression.

$\theta = \pm\frac{1}{2}\pi, \pm\frac{3}{2}\pi$. The SFG signal is reduced then to about 13.5% of the maximal value. This should be compared with the calculated value of $\sim 1\%$, being the classical background level. We believe the residual signal at the minima points is due to large shot-to-shot fluctuations in the down-converted spectral envelope, which affected the averaged measurement of the SFG signal.

These results verify that SFG at the frequencies of the pump laser is induced coherently by broadband squeezed vacuum, and therefore can be coherently controlled by pulse-shaping techniques, despite the fact that the squeezed vacuum may be neither coherent, nor pulsed. Accordingly, the SFG signal at these frequencies is as coherent as the pump laser that generated the squeezed vacuum. We note that a similar principle is known to hold at low-power squeezed vacuum, where the nonlocal second-order coherence effects of entangled photon-pairs are determined by the first-order coherence of the pump laser [18]. However, the equivalence of high-power squeezed vacuum to a coherent ultrashort pulse is not directly connected to second-order coherence, which exhibits a similar temporal behavior only at the single-photon regime [19]. While non-classical features are expected in two-photon interactions with

squeezed vacuum, they were not demonstrated in these experiments.

Two-photon interactions induced by broadband squeezed vacuum exhibit the low intensity and the narrow spectral resolution of the pump laser, while exhibiting the efficiency and temporal resolution of an ultrashort pulse with the same broad bandwidth as the squeezed vacuum. The possibility to induce nonlinear interactions like an ultrashort pulse, yet with spectral resolution and peak intensities of a continuous, single-frequency laser may offer new opportunities for various applications such as multi-users optical communication [20] and multiphoton microscopy.

* Barak.Dayan@Weizmann.ac.il

† Avi.Peer@Weizmann.ac.il

‡ www.weizmann.ac.il/home/feyaron/

- [1] D. Meshulach, Y. Silberberg, Phys. Rev. A **60**, 1287 (1999).
- [2] D. J. Tannor, S. A. Rice, J. Chem. Phys. **83**, 5013(1985).
- [3] M. Shapiro, P. Brumer, J. Chem. Phys. **84**, 4103 (1986).
- [4] W. S. Warren, H. Rabitz, M. Dahleh, Science **259**, 1581 (1993).
- [5] S. A. Rice, M. Zhao, *Optical Control of Molecular Dynamics*, (Wiley, New York, 2000).
- [6] A. M. Weiner, Rev. Sci. Instr. **71**, 1929 (2000).
- [7] D. Meshulach, Y. Silberberg, Nature **396**, 239 (1998).
- [8] Z. Zheng, A. M. Weiner, Opt. Lett. **25**, 984 (2000).
- [9] Z. Zheng, A. M. Weiner, Chem. Phys. **267**, 161 (2001).
- [10] L. Mandel, E. Wolf, *Optical Coherence and Quantum Optics*, (Cambridge Univ. Press, Cambridge, 1995).
- [11] C. K. Hong, L. Mandel, Phys. Rev. A **31**, 2409 (1985).
- [12] M. O. Scully, S. Zubairy, *Quantum Optics*, (Cambridge Univ. Press, Cambridge, 1997).
- [13] B. R. Mollow, R. J. Glauber, Phys. Rev. **160**, 1097 (1967).
- [14] K. J. McNeil, C. W. Gardiner, Phys. Rev. A **28**, 1560 (1983).
- [15] I. Abram, R. K. Raj, J. L. Oudar, G. Dolique, Phys. Rev. Lett. **57**, 2516 (1986).
- [16] B. Dayan, Y. Silberberg (to be published).
- [17] N. Ph. Georgiades, E. S. Polzik, K. Edamatsu, H. J. Kimble and A. S. Parkins, Phys. Rev. Lett. **75**, 3426 (1995).
- [18] Z. Y. Ou, X. Y. Zou, L. J. Wang, L. Mandel, Phys. Rev. Lett. **65**, 321 (1990).
- [19] C. K. Hong, Z. Y. Ou, L. Mandel, Phys. Rev. Lett. **59**, 2044 (1987).
- [20] A. Pe'er, B. Dayan, Y. Silberberg, A. A. Friesem (to be published).