Spectral broadening and shaping of nanosecond pulses: towards shaping of single photons from quantum emitters

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We experimentally demonstrate spectral broadening and shaping of exponentially-decaying nanosecond pulses via nonlinear mixing with a phase-modulated pump in a periodically-poled lithium niobate (PPLN) waveguide. 1550 nm pump light is imprinted with a temporal phase and used to upconvert a weak 980 nm pulse to 600 nm while simultaneously broadening the spectrum to that of a Lorentzian pulse up to 10 times shorter. While the current experimental demonstration is for spectral shaping, we also provide a numerical study showing the feasibility of subsequent spectral phase correction to achieve temporal compression and re-shaping of a 1 ns mono-exponentially decaying pulse to a 250 ps Lorentzian, which would constitute a complete spectro-temporal waveform shaping protocol. This method, which uses quantum frequency conversion in PPLN with > 100:1 signal-to-noise ratio, is compatible with single photon states of light.

1. Introduction

Hybrid quantum networks that combine different physical systems are one approach to achieving the varied functions needed in photonic quantum information processing systems [1]. Unfortunately, the disparate components of such a quantum network may not share the same spectro-temporal properties, leading to an intrinsic incompatibility that can only be overcome via an "adaptation interface." For example, single photon sources based on single quantum emitters like InAs quantum dots [2], nitrogen vacancy centers in diamond [3], and neutral alkali atoms [4] exhibit desirable features such as on-demand generation with the potential for high single photon purity. To interface the emission wavelengths below 1000 nm with the low-loss telecommunications band, quantum frequency conversion interfaces [5, 6] have been proposed and developed, both in bulk and on-chip geometries. However, wavelength incompatibility is not the only challenge that needs to be overcome. While the temporal waveform of such two-level quantum emitters is typically a few nanosecond mono-exponential decay, telecommunications networks are better suited to Gaussian and square pulses that are much shorter in duration. Moreover, the bandwidth of quantum memories that are an integral part of the quantum repeater protocol may be either broader or narrower than the bandwidth of the photons [7]. Spectro-temporal shaping of single photons, while preserving their quantum nature, is thus a vital tool for hybrid quantum networks [8, 9].

Here, we simultaneously frequency translate and convert the spectrum of a mono-exponentially decaying pulse to that of a shorter Lorentzian wavepacket. The core of this approach is frequency conversion using a spectrally chirped pump in a nonlinear medium [10]. We focus on classical input pulses with a mono-exponential decay in the nanosecond regime, mimicking the emission of the aforementioned single quantum emitters. We demonstrate spectral broadening of such waveforms using a variation of a setup previously used in nearly background-free quantum frequency conversion of single photon from a semiconductor quantum dot [11]. Our results complement recent work on spectral compression of single photons through nonlinear wave mixing [12]. Moreover, they constitute the first step towards complete spectro-temporal waveform shaping as proposed by Kielpinski and colleagues [10]. We highlight the experimental requirements for this, by calculating the spectral phase correction that would need to be applied to our output field to achieve temporal pulse compression.

2. Basic principles

There are several contexts in which spectro-temporal control of single photon wavepackets has been demonstrated. Three-level atoms enable waveform shaping during single photon generation [13], while photon echo quantum memories have been used for pulse compression and decompression [14]. Photons with sufficiently broad spectral bandwidths are amenable to pulse shaping techniques based on dispersive optics and spatial light modulators, as has been demonstrated for ultrafast biphoton wavepackets [15, 16]. Unfortunately, for single photon emitters such as quantum dots, trapped atoms, or nitrogen vacancy centers, the emission bandwidth is too narrow (< 1 GHz) for typical line-by-line
shaping. For such systems, spectral broadening has to precede temporal waveform shaping.

For arbitrary waveform shaping, controlling both the temporal phase (to change the spectral shape) and the spectral phase (to control the temporal waveform) is needed. An approach to doing so, outlined in Ref. 10, starts by imparting a temporal phase on an incoming single photon wavepacket to convert the input spectrum to the desired output spectrum. This is followed by spectral phase compensation to remove the unwanted accumulated spectral phase, and convert the temporal waveform to the desired output temporal shape.

The temporal phase manipulation step, shown schematically in Fig. 1(a), is the focus of this paper. Here, an incoming single photon pulse is mixed with a strong, phase-modulated classical pump pulse in a nonlinear crystal. The frequency of the pump is chosen to convert the photon to the desired output wavelength, while the temporal phase is imprinted across the frequency converted single photon. By correctly choosing the temporal phase, it is possible to convert the input spectrum into any desired output spectrum through the properties of Fourier transforms:

$$|\Omega_{out}(\omega)| = \left| \int dt \Omega_{in}(t) e^{i\phi(t)} e^{i\omega t} \right|$$

(1)

When the output spectrum is broader than the input spectrum (temporal compression), the method of stationary phase can produce an analytical solution for the temporal phase $\phi(t)$. For converting a mono-exponentially decaying input $\Omega_{in}(t) = H(t)e^{-|t|/\tau}$, where $H(t)$ is the Heaviside step function, to a Gaussian $\Omega_{out}(t) = e^{-t^2/2\sigma^2}$, we have $\phi(t) \approx \sqrt{2} \int_0^t \text{erf}^{-1}(-t'/\tau)dt'$, where $\text{erf}^{-1}(t)$ is the inverse error function [10]. While Ref. 10 considers large temporal compression ratios ($\approx 100$), we focus on more modest ratios we can achieve experimentally ($\lesssim 10$). Here, this $\phi(t)$ produces a spectrum matching a temporal Lorentzian, due to the smaller compression ratios.

The addition of a temporal phase cannot modify the temporal profile, so that even though the output waveform has the desired spectrum, its time-domain waveform is unchanged due to an extra spectral phase imprinted on it. This unwanted spectral phase can be extracted numerically by computing the phase of $\Omega_{out}(\omega)$, as a transform-limited pulse will have flat spectral phase. If the broadening in the first step increases the bandwidth sufficiently, spatial light modulator pulse shaping techniques [15, 16] may be used for spectral phase compensation, so that at the output, the pulse should have both the desired spectral and temporal profiles.

3. Experiment

In the experiment, we demonstrate spectral waveform shaping of classical light pulses compatible with the single photon regime. We target a mono-exponentially decaying pulse in time (a Lorentzian spectrum) of various durations (1 ns to 30 ns, spanning spontaneous emission lifetimes ranging from InAs/GaAs quantum dots to trapped atoms, respectively) and spectrally shape it to match the spectrum of a compressed Lorentzian wavepacket in time (mono-exponential decay in frequency), while simultaneously shifting the wavelength. The Lorentzian shape has been chosen because the required phase profile can be implemented with the available resources, where the main limitation is the 4 GHz sampling rate of our arbitrary-waveform generator and resulting number of phase points across the pulses.

To start, an exponentially decaying pulse that mimics the temporal profile of a single quantum emitter is generated by passing an attenuated 980 nm laser through an intensity electro-optic modulator (EOM) driven by an arbitrary waveform generator (AWG) (Fig. 1(b)). Simultaneously, the second channel of the AWG drives a phase EOM and imprints the desired phase on the continuous wave 1550 nm pump. An erbium-doped fiber amplifier produces $\approx 1000$ mW of 1550 nm pump that is combined with the 980 nm pulses in a PPLN waveguide (2 cm long, 5 % MgO, Y-cut). Sum frequency generation yields 600 nm light whose spectrum is broadened and shaped into the spectrum of a Lorentzian pulse in time. The wall-to-wall conversion efficiency (which includes input and output coupling losses) is estimated to be $40 \% \pm 1 \%$ [17] with a signal-to-noise ratio $> 100 : 1$, as described in Ref. 11, where quantum frequency conver-
sion with a single photon source was performed, establishing that the measurements below can be extended to the single photon regime if the single photon wavepackets are phase-coherent over the duration of the applied temporal phase. For single quantum dots, this suggests that resonantly pumped systems [18] will be required.

The initial 980 nm spectrum is measured by a Fabry-Perot (FP) analyzer with 10 GHz free spectral range and finesse > 100. The 980 nm pulse is then switched into a delay line, so that inside the PPLN waveguide, its starting edge is matched with the temporal phase carried by the pump. The PPLN output goes through a pair of prisms and a 750 nm short pass filter to remove residual and frequency doubled pump light, and the upconverted signal at 600 nm is analyzed via a second FP (10 GHz free spectral range and finesse > 100).

Figure 2 shows the data for spectral shaping of a 10 ns mono-exponentially decaying pulse, similar to that expected for the nitrogen vacancy center in diamond [3]) to the spectrum of a 1 ns Lorentzian (a mono-exponential decay in frequency). The input optical pulse is shown in Fig. 2(a), while the temporal phase function needed for shaping is shown in Fig. 2(b). As this phase function exceeds $2\pi$ half way through the pulse, and the AWG/EOM can supply a maximum of $2\pi$ phase, the phase is wrapped around near the $2\pi$ point. Figure 2(c) shows the measured spectrum for the initial 980 nm pulse, a Lorentzian in which the two superimposed peaks result from the pulse repetition rate (40 MHz), and subsidiary shoulders are due to imperfections in the FP response. Figure 2(d) shows the measured spectrum of the frequency converted signal. The black line is the theoretically calculated spectrum for the 10 ns to 1 ns spectral broadening and shaping, given the input 980 nm pulse and applied temporal phase. The red points are the experimental data measured via the second FP analyzer. The peaks in the frequency converted 600 nm spectrum are due to the 40 MHz pulse repetition rate. The envelope of these peaks matches the theoretical curve (which does not include the repetition rate) well.

This agreement is quantified by an overlap integral between the data ($|\Omega_{1b}(\omega)|$) and theory ($|\Omega_{th}(\omega)|$):

$$I = \frac{\int |\Omega_{exp}(\omega)||\Omega_{1b}(\omega)|d\omega}{\int |\Omega_{exp}(\omega)|^2d\omega \int |\Omega_{1b}(\omega)|^2d\omega} \quad (2)$$

Including the pulse repetition rate and FP response, we find $I = 0.88 \pm 0.02$, indicating some success in this spectral shaping approach [19]. Deviation from $I=1$ is likely due to a number of factors, including the limited sampling rate of our AWG and the corresponding distortions it causes in the applied phase, particularly in the 'wrapping around' points (when the phase reaches $2\pi$).

This spectral shaping approach can be configured to convert a variety of different input spectra to desired output spectra. In Fig. 3, we present experimental data on the spectral broadening and shaping of input pulse durations that are consistent with those measured for other single quantum emitters. In Fig. 3(a), we show the conversion of a 6 ns decaying pulse, similar to that of organic dye molecules used as single photon sources [20], to the spectrum of a 0.5 ns Lorentzian pulse. In Fig. 3(b), we show the conversion of the spectrum of a 1 ns pulse, similar to that from quantum dot single photon sources [2], to the spectrum of a 250 ps pulse, which is at the lower limit of output pulse duration that can be achieved by our AWG. In Fig. 3, the input and output spectra have been normalized for comparison of spectral shape and bandwidth. In practice, the output spectral amplitude is diminished due to the non-unity conversion efficiency (the area under the 600 nm pulse spectrum is 40 % of the area under the 980 nm pulse spectrum).

4. Towards temporal waveform shaping

To move from spectral shaping to temporal shaping, residual spectral phase must be removed from the generated pulse, and can be done by a number of techniques [21]. Here, we provide numerical calculations of the full spectro-temporal waveform shaping protocol.

We target compression of a 1 ns mono-exponential decay (Fig. 4(a), blue line) to a 250 ps (full-width at half-maximum) temporal Lorentzian (Fig. 4(d), black dashed line). First, we calculate the appropriate temporal phase (Fig. 4(a), red line) to create the spectrum of the 250 ps Lorentzian (Fig. 4(b)). Achieving flat phase across this spectrum requires a spectral phase correction function that is highly oscillatory. A smoothed function that should be easier to implement in practice is shown...
Fig. 3. (a) Broadening the spectrum of a 6 ns exponentially decaying pulse to the spectrum of a 0.5 ns Lorentzian. (b) Same as (a), but from 1 ns to 250 ps.

in Fig. 4(c). Figure 4(d) shows the resulting temporally compressed waveform (red line), along with the waveform that would be generated if perfect spectral phase correction is achieved (blue dotted line). Both of these curves are distorted from the target 250 ps Lorentzian (black dashed line), indicating that the spectral phase compensation and temporal phase application steps are not ideal. This is due to various approximations used, such as the method of stationary phase and our choice of smoothly varying functions. The resulting compressed waveform has a full-width at half-maximum of ≈310 ps, indicating that despite the distortion, a significant temporal re-shaping and compression is feasible.

In summary, we have implemented the first stage of a proposal for quantum waveform shaping [10], where nonlinear mixing of an input mono-exponentially decaying pulse with a phase-modulated pump simultaneously translates the wavelength of the pulse and spectrally broadens and shapes it to match a desired output spectrum. This approach is compatible with single photons and adds to the toolkit of resources being developed for applications in photonic quantum information science.

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References

[17] Conversion efficiency is the ratio of frequency converted photon flux to input photon flux, accounting for the single photon counter quantum efficiency at the two wavelengths. The uncertainty (one standard deviation value) is due to fluctuations in the detected count rate.
[19] The uncertainty is due to possible temporal misalignment between the 1550 nm and 980 nm signals, estimated to be ±250 ps (one standard deviation).