

Quantum frequency conversion from ultraviolet to visible band through waveguides in a period-poled MgO:LiTaO₃ crystal

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Abstract: In research on hybrid quantum networks, visible or near-infrared frequency conversion has been realized. However, technical limitations mean that there have been few studies involving the ultraviolet band, and unfortunately the wavelengths of the rare-earth or alkaline-earth metal atoms or ions that are used widely in research on quantum information are often in the UV band. Therefore, frequency conversion of the ultraviolet band is very important. In this paper, we demonstrate a quantum frequency conversion between ultraviolet and visible wavelengths by fabricating waveguides in a period-poled MgO:LiTaO₃ crystal with a laser writing system, which will be used to connect the wavelength of the dipole transition of ¹⁷¹Yb⁺ at 369.5 nm and the absorption wavelength of Eu³⁺ at 580 nm in a solid-state quantum memory system. An external conversion efficiency of 0.85% and a signal-to-noise ratio of greater than 500 are realized with a pumping power of 3.28 W at 1018 nm. Furthermore, we complete frequency conversion of the classical polarization state by means of a symmetric optical setup based on the fabricated waveguide, and the process fidelity of the conversion is (96.13 ± 0.021)%. This converter paves the way for constructing a hybrid quantum network and realizing a quantum router in the ultraviolet band in the future.

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1. Introduction

Interfacing fundamentally different quantum systems is key to building future hybrid quantum networks [1–4] that combine the advantages of different quantum systems to complete the operation, storage, and reading of quantum states [5,6]. However, because of their different wavelengths and bandwidths, it is difficult to connect these quantum systems directly. Quantum frequency conversion (QFC) offers an effective way to bridge the gaps between these systems, and some impressive results have been achieved in recent years [7–10]. It has been shown that QFC can preserve a broad range of photon properties, such as first- and second-order coherence, polarization, orbital angular momentum, and entanglement [11–16]. However, most experiments to date with QFC have been in the visible and near-infrared spectral ranges [17,18], which are not compatible with the working band of the common rare-earth or alkaline-earth metal atoms or ions at ultraviolet (UV) wavelengths. For example, for ¹⁷¹Yb⁺, for which research on quantum computing has progressed most rapidly to date, its fluorescence wavelength is 369.5 nm. QFC at UV wavelengths involves two aspects, one being nonlinear crystals: UV photons suffer higher losses in crystals than those at longer wavelengths. The other aspect is crystal poling technology.

The polarization period of UV wavelengths is often very short, thereby making it difficult to achieve uniform poling [19]. Nevertheless, in [9], the authors converted photons at 1311 nm to 369 nm by means of a periodically poled potassium-titanyl-phosphate (PPKTP) waveguide, achieving a conversion efficiency of 5% with a pumping power of 200 mW.

For converting UV wavelengths, a periodically poled MgO-doped stoichiometric LiTaO₃ (PPSLT) crystal is supposedly suitable because of its high optical damage threshold and shorter cut-off wavelength down to 0.28 μm [20–22]. Compared with other nonlinear crystals such as PPKTP, its transmittance at 369 nm is up to ~76%, and its absorption is hardly affected by temperature change ($< 1\%$ at temperatures of 26 – 62 $^{\circ}\text{C}$). To realize high conversion efficiency, it is essential to form waveguides in the crystal to improve the light intensity and interaction length [23]. In our experiment, the difference-frequency generation (DFG; $\nu_{369} - \nu_{1018} \rightarrow \nu_{580}$) process requires a wide wavelength range covering the UV, visible, and near-infrared bands. Meanwhile, to ensure the coupling efficiency of the optical fibers, the mode transmitted in the waveguide is preferably a single mode. Therefore, the geometry of depressed-cladding waveguides is favorable because it comprises an undamaged circular core surrounded by many tracks with low index change. In recent studies, PPSLT waveguides with three/five cladding layers were fabricated by femtosecond writing [24]. In [14], the coupling efficiency to the waveguide was limited to only 30%, which can be improved by controlling the cross-sectional dimension [25–28].

In this letter, we demonstrate the conversion of UV photons to visible photons by designing and fabricating a depressed-cladding waveguide in a PPSLT crystal with laser direct writing. Specifically, we converted from 369.5 nm to 580.0 nm, which correspond respectively to the fluorescence wavelength of $^{171}\text{Yb}^{+}$ in an ion trap and the absorption wavelength of Eu^{3+} in a solid-state quantum memory system. We adopted a noise-free scheme by means of long-wavelength pumping at 1018 nm [8,29]. Furthermore, based on the fabricated waveguide, we completed frequency conversion of the classical polarization state using a phase-stable optical setup, and the process fidelity of the conversion was $(96.13 \pm 0.021)\%$.

2. Experimental setup

The PPSLT crystal was supplied by the OXIDE Corporation, Japan. It was 10-mm long and 400- μm thick. To facilitate the poling electrode, we chose third-order quasi-phase-matching (QPM) with a corresponding poling period Λ_0 of 7.36 μm . The laser writing system (FemtoLAB; Altechna R&D Lithuania) is shown in Fig. 1(a). The main laser source was a YAG femtosecond laser, whose emission passed through a variable attenuator and was then focused into the PPSLT by an objective lens ($\text{NA} = 0.65$). To reduce aberration, the focal spot was close to the crystal surface. The system parameter values were as following: the laser pulse width was 210 fs, the repetition rate was 20 kHz, and the writing speed was 10 mm/s. We designed a structure with seven cladding layers, as shown in Fig. 1(b). The distance Λ between neighboring tracks was 10 μm , and the diameter of a track was 6 μm .

As the aberration of the crystal alters the focused laser spot, its intensity changes gradually along with its depth, so the required optimal fabrication pulse energy differs with increasing focal depth. To determine the fabrication pulse energy at different depths, we constructed a matrix to test different pulse energies in the crystal, as shown in Fig. 1(c). Three rows of tracks were fabricated with depths of 15, 30 and 50 μm , respectively. In each row, 10 tracks were written with pulse energies ranging from 630 nJ to 30 nJ in intervals of 60 nJ as shown from left to right. From Fig. 1(c), when the pulse energy is low, the tracks are not written successfully, and the track shape can help us to choose a proper pulse energy. After further optimization, we chose the corresponding track in each row with a symmetric shape, and their pulse energies were 150, 180, 230 and 250 nJ for depths of 15, 30, 50, and 70 μm , respectively.

The profile of the fabricated waveguide is shown in Fig. 1(d), the depth of which was 70 μm . After fabrication, we subjected the waveguide to annealing to reduce the scattering loss; the

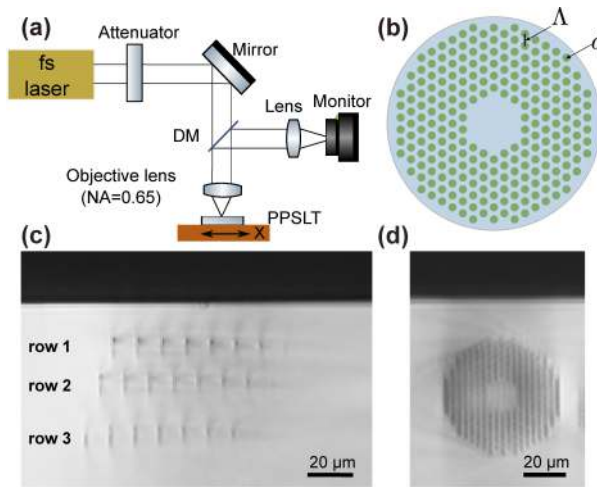


Fig. 1. Schematic of fabrication setup and structure of waveguide in PPSLT. (a) Laser writing system for waveguide fabrication; multi-tracks were written in the crystal by moving the translation stage back and forth in the X direction. (b) Cross section of designed structure of waveguide; the green dots represent the tracks, which were distributed with a periodic structure. The inter-track distance Λ was $10\ \mu\text{m}$, and the track diameter was $6\ \mu\text{m}$. (c) Single-track writing test with different pulse energy and depth; the pulse energies for 10 tracks of each row from left to right were 630, 570, $\dots$, 30 nJ, and the depth of rows 1–3 were 15, 30 and $50\ \mu\text{m}$, respectively. Tracks with low energy were not written successfully. (d) Image of end-facet of crystal showing fabricated waveguide structure.

crystal was surrounded by aluminum foil and placed in an oven, where the annealing temperature was $350\ ^\circ\text{C}$ and the annealing time was 1 h [24].

After annealing, the UV transmittance increased from 50% to 60%, which accounts for the waveguide surface scattering and mode coupling. This higher transmittance also shows that the waveguide has a good binding effect for the UV band. The transmittance of the 1018-nm laser was 25%, which may have been caused by the weak binding effect of the waveguide for the large mode field in the near-infrared band. Nevertheless, the result is acceptable, because the 1018-nm wavelength is pump light and the PPSLT crystal has a high optical damage threshold. After transmission from the waveguide, a $10\times$ objective lens was used to image the transmitted spot on a charge-coupled device (CCD) to measure its diameter, as shown in Fig. 2. Figure 2(a) – (c) show the simulated modes at 369, 580 and 1018 nm, respectively, and Fig. 2(d) – (f) show the corresponding measured mode profiles. The measured spot diameters are 8.7, 11, and $12.2\ \mu\text{m}$, which are consistent with the simulation results.

The DFG efficiency of the waveguide at UV wavelengths was characterized by measuring the power ratio before and after the conversion. The DFG experimental setup is shown in Fig. 3. The pumping light was produced by a tunable external-cavity diode laser (Toptica-pro) with a narrow linewidth, and the UV light was produced by a frequency-tripling laser. Both input beams overlapped on a dichroic mirror (DM) and were then coupled to the waveguide through an objective lens ($\text{NA} = 0.2$; transmittance to UV wavelengths = 95%). The temperature of the crystal was stabilized to within 4 mK, and three filters were used to filter out the strong pumping light and UV light (with a bandwidth of 2 nm) at the output of the waveguide.

To obtain the optimal QPM temperature, we scanned the temperature of the waveguide and measured the DFG power at a pumping power of 1 W. The results are shown in Fig. 4, where there is good agreement between the measured data and the characteristic sinc^2 phase matching

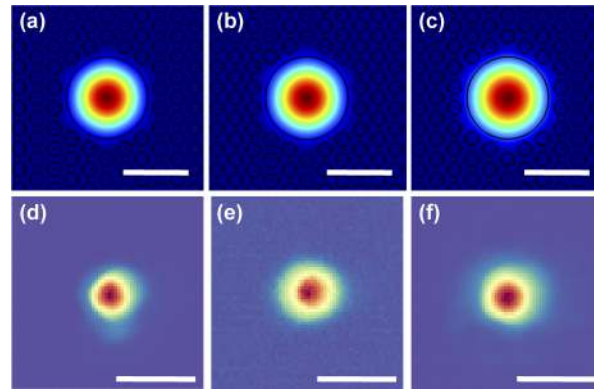


Fig. 2. Mode profiles of different light modes transmitted from waveguide: (a) – (c) simulated modes at 369, 580, and 1018 nm, respectively; (d) – (f) corresponding measured mode profiles with spot diameters of 8.7, 11, and 12.2 μm , respectively. The scale bar represents 10 μm .

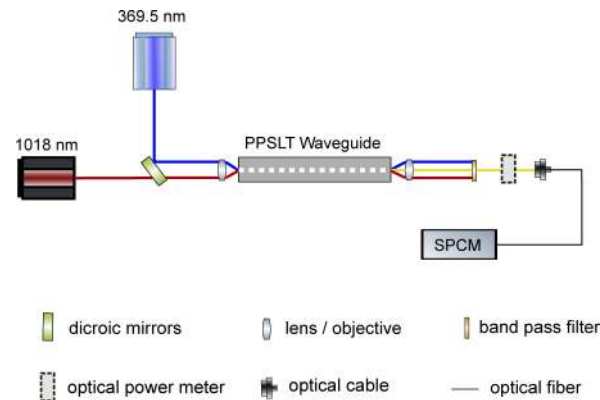


Fig. 3. Schematic of difference-frequency generation (DFG) experimental setup. The 369.5-nm and 1018-nm lasers were single-mode lasers with a narrow linewidth of less than 100 kHz, and their wavelengths were stabilized to within 1 MHz by locking the lasers to stable reference cavities. SPCM: single-photon counting module. The generated photons were filtered by band filters with 2-nm bandwidth and then detected by an optical power meter or an SPCM.

profile. The optimal QPM temperature was 113.78 $^{\circ}\text{C}$, and the corresponding DFG process conversion efficiency was $0.26\% \text{ W}^{-1} \text{ cm}^{-1}$ (the coupling efficiency of the waveguide was $\sim 40\%$, and the detection efficiency of the detector was 59%). This result is basically the same as that in [10]. The full width at half maximum (FWHM) of the QPM temperature was 0.6 $^{\circ}\text{C}$, and the corresponding frequency bandwidth was $\sim 130 \text{ GHz}$.

To compare the DFG efficiencies of the crystal for strong light and single-photon level, we measured them separately. First, we measured the DFG conversion efficiency with a UV-light input power of 860 μW , and the conversion efficiency was 0.85% at a pumping power of 3.28 W near the optimal temperature of 113.78 $^{\circ}\text{C}$. As shown in Fig. 5(a), the DFG conversion efficiency is proportional to the pumping power, which means that the pump power has not reached saturation; a higher conversion efficiency will be obtained under higher pumping power. To further filter out the 369.5-nm and 1018-nm photons, three bandpass filter plates (one piece of FESH0650 and two pieces of FESH0600) were used, achieving a total attenuation of the pumping light of up to

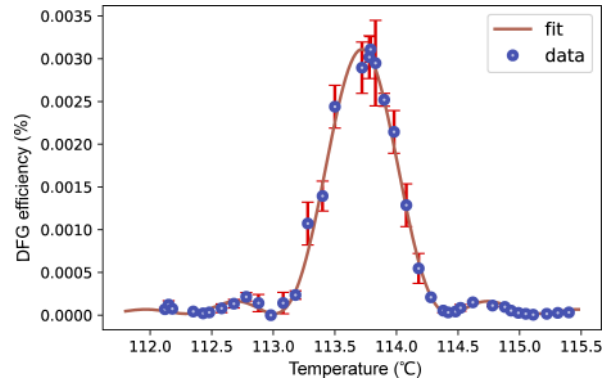


Fig. 4. Measured quasi-phase-matching (QPM) temperature curve for DFG process. The optimal DFG conversion efficiency was $0.26\% \text{ W}^{-1} \text{ cm}^{-1}$ at the optimal QPM temperature of 113.78°C .

20 orders of magnitude. Meanwhile, to further filter out the second-harmonic-generation light, two narrowband filters (FWHM = $\sim 1 \text{ nm}$; central wavelength = 580 nm) were used. We then measured the conversion efficiency with a single-photon-level UV input with a photon counting rate of 8.2 MHz ($\sim 1 \text{ pW}$). The results are shown in Fig. 5(b). The single-photon-level DFG conversion efficiency was also 0.85% at a pumping power of 3.28 W , consistent with the result for the strong light field.

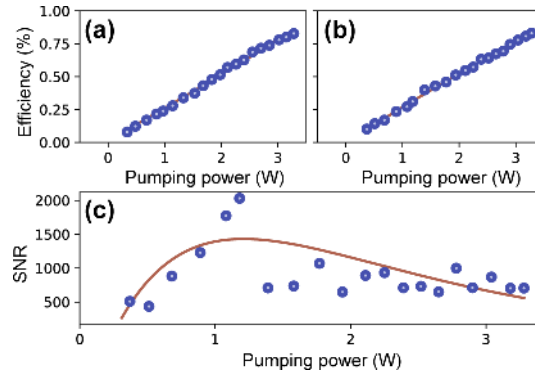


Fig. 5. DFG conversion efficiency versus pumping power. (a) Classical light-field conversion efficiency; the input UV light intensity was $860 \mu\text{W}$. (b) Single-photon level conversion efficiency with UV-light input count rate of 8.2 MHz ($\sim 1 \text{ pW}$). (c) Measured signal-to-noise ratio (SNR) of conversion process.

Because higher pumping power increases the noise count, we measured the signal-to-noise ratio (SNR) of the conversion process at different pumping powers with single-photon level, as shown in Fig. 5(c). In the whole conversion process, the SNR was always greater than 500, and the optimal pump power was 1.2 W , corresponding to an SNR of greater than 1400. This shows that even with high pumping power, a high SNR can still be achieved. If a narrower filter is used, a higher SNR is achieved, which is very beneficial for obtaining high-fidelity quantum states [30,31].

In our experiment, because of the long-wavelength pumping scheme that was adopted, there was no noise of spontaneous parametric down conversion. The conversion remained free of pump-induced noise even at the required high pump power [8,32], and the frequency difference

between these photons reached 300 THz, so the effect of Raman noise was insignificant [33]. The only noise in this experiment came from spontaneous upconversion. Under strong pumping power, this process cannot be avoided, but the noise level can be reduced significantly by using a narrower filter.

To realize frequency conversion for arbitrary polarization states of UV photons, we designed a symmetrical optical setup, as shown in Fig. 6(a). Because a single PPSLT waveguide can only convert vertically polarized photons, we fabricated a waveguide on another PPSLT crystal with the same parameter values. After testing, the two waveguides were shown to perform consistently. To separate the two polarization modes by a certain distance horizontally, we designed a trapezoidal polarization beam splitter (TPBS) comprising a wedge crystal and a triangular crystal. A half-wave plate (HWP) was attached to the transmission end of the TPBS to convert horizontal polarization into vertical polarization; similarly, after the conversion, another TPBS was used to convert vertical polarization into horizontal polarization. The whole optical structure was symmetrical, so the relative phase difference between the two polarization modes was stable. However, there was still a relative phase difference between the two waveguides due to the difference in the actual optical paths, so three wave plates were used to compensate for this relative phase difference between the two polarizations.

The input state was produced by an HWP and a quarter-wave plate (QWP), and then checked and recorded by a polarization analyzer (PA; SK010PA; Schäfer+Kirchhoff GmbH, Germany). When the recording process was completed, the mirror was flipped down so that the UV light could participate in the DFG process. The converted state was recorded by another PA. We measured the corresponding output states of the $|H\rangle$, $|V\rangle$, $|D\rangle$, $|A\rangle$, $|R\rangle$, and $|L\rangle$ input states, and the measurement time for each input state was 600 s.

To assess the quantum nature of the DFG process, we characterized it by using tomographic techniques [13]. The transformation matrix of the converter is shown in Fig. 6(b), where the solid

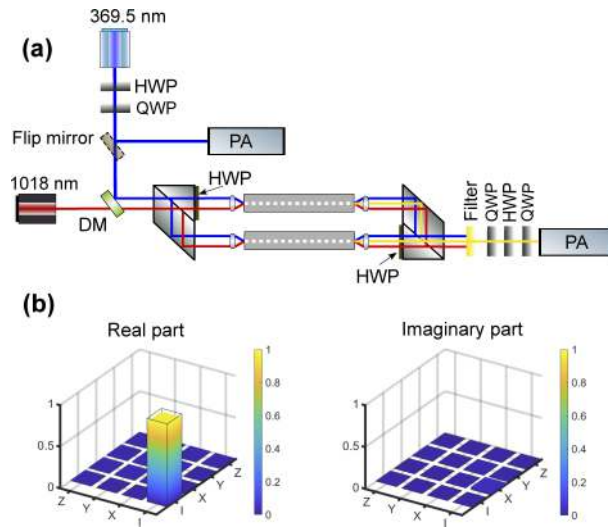


Fig. 6. Optical path of polarization state conversion and conversion matrix between ideal channel and real channel. (a) The whole optical structure was symmetrical to stabilize the relative phase difference between the two polarizations after conversion. The input state was produced by a half-wave plate (HWP) and a quarter-wave plate (QWP) and recorded by a polarization analyzer (PA). The three wave plates on the right were used to compensate for the relative phase difference between the two polarizations. (b) Conversion matrix of channel. The solid line represents the conversion matrix of the ideal channel.

line represents the ideal transformation matrix. We then calculated the fidelity of the channel with the ideal channel, and the process fidelity of the converter was $(96.13 \pm 0.021)\%$.

3. Results and discussion

The PPSLT crystal used in this work was third-order QPM, which led to low conversion efficiency. If first-order QPM could be used, the conversion efficiency would be improved ($\sim 10\%$), but the poling period required for first-order QPM is $\sim 2.6 \mu\text{m}$, and it would be quite a challenge to make such a short and uniform poling period on bulk PPSLT crystal. However, a poling depth of $100 \mu\text{m}$ would be enough to support creating waveguides, instead of poling the entire bulk crystal, so we intend to use this method to improve the conversion efficiency in our next experiment. Meanwhile, polishing and coating the two end faces of the crystal after the waveguide processing would improve the conversion efficiency.

The photon noise generated by the DFG process is quite low. After the 2-nm (1.8-THz) bandpass filter, the noise counting rate was measured as being ~ 40 Hz even at a high pumping power of 3.28 W. Because the bandwidth of the Eu^{3+} solid-state memory is below 20 MHz, the photon-noise counting rate falling into this band will be below 10^{-3} Hz. This low-noise feature of the conversion process is conducive to making a quantum interface in the future.

In the experiment of measuring fidelity, there was no active feedback device, only a hood to isolate air flow. Within 600 s, the fidelity was unchanged, as shown in Fig. 7. This indicates that the fabricated waveguide could be used in a quantum state conversion experiment. The fidelity of $(96.13 \pm 0.021)\%$ was limited by the accuracy of the PA, which was 5%. In an actual conversion experiment, the fidelity would be higher if a photoelectric multiplier were to be.

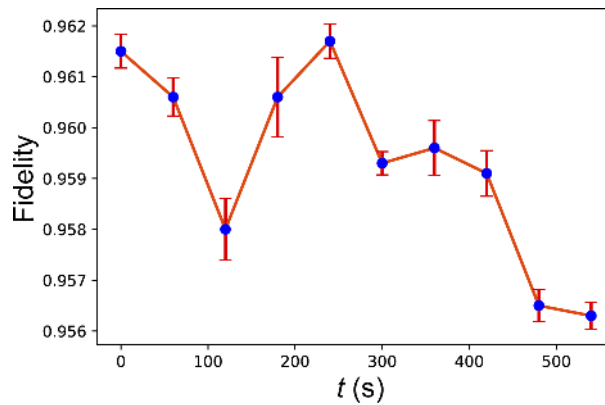


Fig. 7. Change of fidelity with time and the measurement time is 600 s.

4. Conclusion

In summary, we have demonstrated a QFC process of the UV band to the visible band through long-wavelength pumping by fabricating a waveguide in a PPSLT crystal. Moreover, based on the waveguide, we completed conversion of the classical polarization state, and the process fidelity of this converter was $(96.13 \pm 0.021)\%$. However, due to the third-order QPM, the conversion efficiency was only $\sim 0.26\% \text{W}^{-1} \text{cm}^{-1}$. If lower-order QPM were to be used in a future experiment, a higher conversion efficiency would be obtained. Our experiment paves the way for the construction of quantum networks and the realization of a quantum router in the UV band, and it will expand greatly the range of existing quantum networks that have been demonstrated.

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Data availability. The data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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