Spectrally pure states at telecommunications wavelengths from periodically poled MTiOXO₄ (M = K, Rb, Cs; X = P, As) crystals

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Significant successes have recently been reported in the study of the generation of spectrally pure state in group-velocity-matched (GVM) nonlinear crystals. However, the GVM condition can only be realized in limited kinds of crystals and at limited wavelengths. Here, we investigate pure state generation in the isomorphs of PPKTP crystal: i.e., periodically poled RTP, KTA, RTA and CTA crystals. By numerical simulation, we find that these crystals from the KTP family can generate pure photons with high spectral purity (over 0.8), wide tunability (more than 400 nm), reasonable nonlinearity at a variety of wavelengths (from 1300 nm to 2100 nm). It is also discovered that the PPCTA crystal may achieve purity of 0.97 at 1506 nm. This study may provide more and better choices for quantum state engineering at telecom wavelengths.

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I. INTRODUCTION

The generation of single photons is a fundamental resource required for optical quantum information processing (QIP) and spontaneous parametric down conversion (SPDC) is one of the most widely used methods to prepare single photons. In the general case of the SPDC process, a pump photon is split into two lower energy daughter photons, the signal and idler, which are spectrally correlated. For many QIP applications, however, it is necessary to utilize bi-photons with no spectral correlations, so as to achieve high-visibility interference between independent sources.

There are two methods to remove the spectral correlations between the bi-photons in SPDC. The first one is to filter the bi-photons using narrow bandpass filters, which can be performed easily, but inevitably and severely decreases the brightness of the photon source. The second method is to engineer the SPDC process, so as to prepare an intrinsically spectrally pure state. Such a quantum state engineering method can be realized by considering the group-velocity-matched (GVM) condition in specific crystals at certain wavelengths. Previous work in the field has shown that the GVM condition can be realized in several crystals, e.g., KDP crystal at 830 nm, periodically poled KTP crystal (PPKTP) at 1584 nm, and BBO crystal at 1514 nm."

The four crystals (RTP, KTA, RTA and CTA) considered in the work are the isomorphs of the KTP crystal, i.e., they belong to the “KTP” family. They have the general form of MTiOXO₄ with \{M = K, Rb, Cs\} and \{X = P, As (for M = Cs only)\}. Therefore, they retain all the general properties of their better-known “parent” KTP crystal. For example, they are all positive biaxial crystals (point group: mm2); they have a long transparency range (0.35 \mu m - 3 \mu m), reasonable nonlinearity, reduced walk-off in the X-Y plane and a high optical damage threshold. All these crystals possess ferroelectric properties, and are suitable for use in periodically poled structures.

The GVM condition is the prerequisite for engineering a spectrally pure state, therefore, the GVM wavelength is a key parameter for these crystals. In the case of Type II SPDC with a PPKTP crystal, the following GVM condition is met at a wavelength of 1584 nm:

\[2V_{g,p}^{-1}(\lambda/2) = V_{g,s}^{-1}(\lambda) + V_{g,i}^{-1}(\lambda),\] (1)
TABLE I. Comparison of the chemical composition, group-velocity-matched (GVM) wavelength $\lambda_{\text{GVM}}$, poling period $\Lambda$, purity and effective nonlinear coefficient $d_{\text{eff}}$ of PPKTP crystal and four of its isomorphs. The $d_{\text{eff}} = d_{32}$ values are taken from the SNLO v66 software package, developed by AS-Photonics, LLC [23].

| Name       | Composition | $\lambda_{\text{GVM}}$ (nm) | $\Lambda$ (\mu m) | Purity | $d_{\text{eff}}$ (pm/V) | References |
|------------|-------------|-----------------------------|-------------------|--------|------------------------|------------|
| PPKTP      | KTiOPo$_4$  | 1584                        | 46.1              | 0.82   | 2.4                    | [5, 17, 18] |
| PPRTP      | RbTiOPo$_4$ | 1643.2                      | 56.6              | 0.82   | 2.4                    | [19]       |
| PPRTA      | KTiOAsO$_4$ | 1634.7                      | 57.3              | 0.82   | 2.3                    | [20]       |
| PPCTA      | RbTiOAsO$_4$| 1784.5                      | 71.1              | 0.82   | 2.4                    | [21]       |
|            | CsTiOAsO$_4$| 1864.6                      | 381.9             | 0.82   | 2.1                    | [22]       |

where $V^{-1}_{\mu}(\mu = p, s, i)$ is the inverse of the group velocity $V_{\mu}$ for the pump $p$, the signal $s$, and the idler $i$. $\Lambda$ is the degenerate wavelength of the signal and idler.

For RTP, KTA, RTA and CTA crystals, the GVM wavelengths are 1643 nm, 1635 nm, 1785 nm and 1865 nm, respectively, as listed in Tab. I. These GVM wavelengths are calculated based on their Sellmeier equations. All these four crystals are suitable for periodically poling, and their poling periods at the GVM wavelength are shown in Tab. I. The effective nonlinearity $d_{\text{eff}}$ of the isomorphs (from 2.4 pm/V to 2.1 pm/V) is comparable to that of the PPKTP crystal (2.4 pm/V).

III. PURE STATE GENERATION IN THE ISOMORPHS

By satisfying the GVM condition, it is possible to prepare spectrally pure bi-photon state using the isomorphs of PPKTP crystal. The spectral purity can be quantitatively evaluated by considering the spectral distribution of the signal and idler photons. This distribution can be described by their joint spectral functions, and their poling periods at the GVM wavelength are shown in Tab. I. The effective nonlinearity $d_{\text{eff}}$ of the isomorphs (from 2.4 pm/V to 2.1 pm/V) is comparable to that of the PPKTP crystal (2.4 pm/V).

IV. WIDE TUNABILITY WITH HIGH PURITY IN THE FOUR CRYSTALS

At the GVM wavelength, the biphoton source has a high spectral purity and this high purity can be maintained at the nearby wavelengths. By simulation, we find all these four crystals can maintain wide wavelength tunabilities under high purity. The purity can remain higher than 0.80, with wavelength tunable from 1300 nm to 1800 nm for PPRTA crystal; from 1300 nm to 1700 nm for PPKTA crystal; from 1400 nm to 2000 nm for PPRTA crystal; from 1500 nm to 2100 nm for PPCTA crystal. This range of wavelengths covers the commonly used S-, C-, L-, and U-bands in optical fiber telecommunications. In Fig. 2(a-d) we provide an example of the spectral distribution of the PPRTA crystal at different wavelengths from 1400 nm to 1700 nm and note that similar property is also possessed by the other three isomorphs and their “parent” PPKTP crystal [28].

V. ANOTHER GVM CONDITION IN THE CTA CRYSTAL

While the GVM condition of Eq. (1) is satisfied in all the isomorphs of PPKTP crystal, a further GVM condition, given by Eq. (2), is satisfied by the PPCTA crystal
FIG. 1. (color online) JSA (first row) and JSI (second row) of the photons generated from the four isomorph crystals at their GVM wavelengths. (a) PPRTP, (b) PPKTA, (c) PRPA and (d) PPCTA. In this simulation, the crystal lengths are fixed at 30 mm long, while the pump laser bandwidths are 0.42 nm, 0.42 nm, 0.50 nm and 0.77 nm for (a), (b), (c) and (d), respectively.

FIG. 2. (color online) JSA (first row) and JSI (second row) of photons generated from the PPRTP crystal at different wavelengths. (a) 1400 nm, (b) 1500 nm, (c) 1600 nm, (d) 1700 nm. The corresponding spectral purity $p$ and poling period $\Lambda$ are shown in the figure. In this simulation, the crystal lengths are fixed at 30 mm long, while the pump laser bandwidths are fixed at 0.42 nm.

At 1506 nm,

$$V_{g,p}^{-1}(\lambda/2) = V_{g,i}^{-1}(\lambda),$$

Under this condition, the JSA can have a very narrow and sharp distribution, as shown in Fig. 3(a). In this simulation, we assumed a crystal length of of 30 mm for PPCTA and a pump bandwidth of 5 nm at 753 nm (with a Gaussian profile). With the JSA in Fig. 3(a), the corresponding intrinsical purity is calculated as 0.97, higher than the purities of 0.82 shown in Fig. 1 and Fig. 2. The corresponding JSI is shown in Fig. 3(b). Previously, it was discovered that this condition was satisfied by a KDP crystal at 830 nm [1, 2, 4, 7]. Here we find, for the first time, that this condition can be satisfied at telecom wavelengths in a PPCTA crystal.

This source offers great potential in quantum interference between independent sources [1, 2, 29]. In Fig. 3(c), we simulate the Hong-Ou-Mandel interference between two signal photons, one of each from two independent PPCTA crystals (with the JSA shown in Fig. 3(a)), with the two idler photons employed as the heralders [1, 2, 8]. Similarly, Fig. 3(d) shows the case of Hong-Ou-Mandel interference between two idlers heralded by the signal photons. The visibilities in Fig. 3(c) and (d) are
determined by the spectral purity shown in Fig. 3(a), which can reach as high as 0.97. The bandwidth of the interference patterns are 0.24 ps and 3.5 ps respectively.

While we only show results for PPCTA here, the conditions of $V_{g,p}^{-1}(\lambda/2) = V_{g,s}^{-1}(\lambda)$ and $V_{g,p}^{-1}(\lambda/2) = V_{p,s}^{-1}(\lambda)$ are also satisfied by the other four crystals. As a result, the similar figure as Fig. 3(a) can also be achieved by PPTRT crystal at 1282 nm and 2491 nm, by PPKTA crystal at 1278 nm and 2481 nm, by PPRTA crystal at 1372 nm and 2933 nm, and by PPKTP crystal at 1225 nm and 2337 nm, respectively.

It should be noticed that the JSA in Fig. 3(a) is asymmetric, therefore, the Hong-Ou-Mandel interference between the signal and the idler photons (from the same SPDC source) does not show a high visibility, which may limit their use in some quantum information processing protocols.

VI. FUTURE ISSUES

These results presented here for this theoretical study of these particular isomorph crystals provide a lot of possible starting points for future theoretical and experimental studies of similar isomorphs and their practical applications to quantum information. Firstly, in addition to the generation of spectrally correlated states shown in Fig. 3, it is also possible to prepare spectrally positively-correlated or negatively-correlated states by varying the crystal length or pump bandwidth, in a similar manner to which has been demonstrated in the case of a PPKTP crystal [13]. Secondly, it is useful to demonstrate customized poiling (by modulating the poling-order [31], the duty-cycle [32], or domain-sequence [33, 34]) in these four isomorphs, so as to improve the maximal intrinsically purity from 0.82 to near 1. Thirdly, it is also meaningful to make waveguide based on these four isomorphs, similar as in the case of PPKTP waveguide [35]. Fourthly, PPCTA crystal shows high spectral purity at around 2 µm wavelength. This wavelength is useful for biology and medical applications, and also useful for the detection of carbon dioxide [36]. This implies that the quantum state generated from a PPCTA crystal may be a good quantum light source for the quantum information processing in these applications.

In our theoretical model, we only consider the spectral correlation between the signal and idler photons. It is also interesting to consider the spatial correlation, since the spatial purity can be further improved by spatial filtering [16, 37, 38]. As reported in Ref. [16], the purity and coupling efficiency can be improved to around 0.9 by using a larger beam waist for the pump laser, and by coupling the photons into single-mode fibers. But the trade off is that the source brightness is lower than in the case of tight focusing.

It was reported recently that the KTP family has 118 known isomorphs [39–41], including not only 29 pure crystals but also 89 doped ones. Their general formula can be written as $M'M'O_4$, where $M = K$, Rb, Na, Cs, Ti, NH$_4$; $M' =$ Ti, Sn, Sb, Zr, Ge, Al, Cr, Fe, V, Nb, Ta, Ga; $X =$ P, As, Si, Ge. The Sellmeier equations for most of these crystals are not reported yet, therefore, exploring the nonlinear optical properties of these crystal is promising for further research. We propose that it is valuable to investigate the GVM wavelength of the pure crystals. For the doped crystals, it may possible to engineer the GVM wavelength to an arbitrary value by adjusting the chemical portions, so as to prepare highly pure photon source at arbitrary wavelength.

VII. CONCLUSION

In conclusion, we have theoretically and numerically demonstrated the generation of spectrally uncorrelated states from a small sub-set of the 118 known isomorphs of the PPKTP crystal. It was found that these particular isomorphs still retain the desirable properties of their parent PPKTP crystal, namely high spectral purity (over 0.8) with wide tunability (more than 400 nm) at a variety of wavelengths (from 1300 nm to 2100 nm). Further, we found that the PPCTA crystal can achieve an intrinsically high spectral purity of 0.97 at wavelength of 1506 nm. In the future, these crystals may have many promis-
ing applications for quantum information processing at telecom wavelengths.

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