Spectrally Pure States at Telecommunications Wavelengths from Periodically Poled $M\mathrm{TiO}X\mathrm{O}_4$ ($M = \mathrm{K, Rb, Cs}$; $X = \mathrm{P, As}$) Crystals

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Significant successes have recently been reported in the study of the generation of a spectrally pure state in group-velocity-matched (GVM) nonlinear crystals. However, the GVM condition can be realized only in limited kinds of crystals and at limited wavelengths. Here, we investigate pure-state generation in the isomorphs of the 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), and reasonable nonlinearity at a variety of wavelengths (from 1300 to 2100 nm). It is also discovered that the PPCTA crystal may achieve a 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 biphotons with no spectral correlations so as to achieve high-visibility interference between independent sources [1–3].

There are two methods to remove the spectral correlations between the biphotons in SPDC. The first one is to filter the biphotons 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 [4–6]. Previous work in the field has shown that the GVM condition can be realized in several crystals, e.g., the KDP crystal at 830 nm [1,2,7], the periodically poled KTP crystal (PPKTP) at 1584 nm [5,8], and the β-barium borate crystal at 1514 nm [4,9,10].

Recently, the study of spectrally pure-state generation in a PPKTP crystal has achieved significant progress [11–16]. Besides high spectral purity, the PPKTP crystal still has several other merits, such as high brightness (because the crystal can be very long, thanks to the periodically poling technique), a high damage threshold (higher than the periodically poled lithium niobate crystal), and wide tunability (more than 200 nm at telecom wavelengths [14]).

However, as mentioned above, the GVM condition can be realized only in these limited crystals and at limited wavelengths. Is it possible to find more crystals to satisfy the GVM condition, and can this be done over a wider range of wavelengths with higher nonlinear efficiency and higher purity? In this paper, we answer this question by investigating the spectral purity of the photons generated from the isomorphs of a PPKTP crystal: i.e., periodically poled RTP ($\mathrm{RbTiOPO_4}$), KTA ($\mathrm{KTiOAsO_4}$), RTA ($\mathrm{RbTiOAsO_4}$), and CTA ($\mathrm{CsTiOAsO_4}$). We expect these crystals to retain the same highly spectral purity with wide tunability as PPKTP and possibly provide higher nonlinearity and purity than the PPKTP crystal. This study may suggest a wider range of choices for the crystals employed when conducting quantum-state engineering at telecom wavelengths.

II. BASIC PARAMETERS OF THE FOUR CRYSTALS

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 $M\mathrm{TiO}X\mathrm{O}_4$ with $\{M = \mathrm{K, Rb, Cs}\}$ and $\{X = \mathrm{P, As\,(for}\ M = \mathrm{Cs\ only)}\}$ [17]. Therefore, they retain all the general properties of their better-known "parent" KTP crystal [18]. For example, they are all positive biaxial crystals (point group $mm2$); they have a long transparency

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TABLE I. Comparison of the chemical composition, GVM wavelength λ_{GVM}, poling period Λ, purity, and effective nonlinear coefficient d_{eff} of the PPKTP crystal and four of its isomorphs. The $d_{eff} = d_{32}$ values are taken from the SNLO v66 software package, developed by AS-Photonics, LLC [24]. The relevant sources in the literature for the appropriate Sellmeier equations are also listed in the table.

Name Composition	PPKTP KTiOPO4	PPRTP $RbTiOPO_4$	PPKTA $KTiOAsO_4$	PPRTA $RbTiOAsO_4$	PPCTA $CsTiOAsO_4$
λ_{GVM} (nm)	1584	1643.2	1634.7	1784.5	1864.6
Λ (μm)	46.1	56.6	57.3	71.1	381.9
Purity	0.82	0.82	0.82	0.82	0.82
d_{eff} (pm/V)	2.4	2.4	2.3	2.4	2.1
References	[5,22,23]	[25]	[26]	[17]	[27]

range (0.35–3 μm), 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 [19–21].

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 [5,8,22,23]:

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

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

For RTP, KTA, RTA, and CTA crystals, the GVM wavelengths are 1643, 1635, 1785, and 1865 nm, respectively, as listed in Table 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 Table I. The effective nonlinearity d_{eff} of the isomorphs (from 2.4 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 a spectrally pure biphoton state using the isomorphs of the 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 amplitude (JSA) $f(\omega_1, \omega_2)$, which is the product of the pump envelope function $\alpha(\omega_s + \omega_i)$ and the phase-matching function $\phi(\omega_s, \omega_i)$, i.e., $f(\omega_s, \omega_i) = \alpha(\omega_s + \omega_i)\phi(\omega_s, \omega_i)$. By applying Schmidt decomposition to the JSA, we can obtain

$$f(\omega_1, \omega_2) = \Sigma_j c_j \phi_j(\omega_1)\varphi_j(\omega_2), \qquad (2)$$

where $\phi_j(\omega_1)$ and $\varphi_j(\omega_2)$ are two orthogonal basis sets of spectral functions and c_j is the normalized coefficient. The spectral purity p, a parameter describing the degree of spectral uncorrelation between the signal and idler photons, can be calculated as

$$p = \sum_j c_j^4. \qquad (3)$$

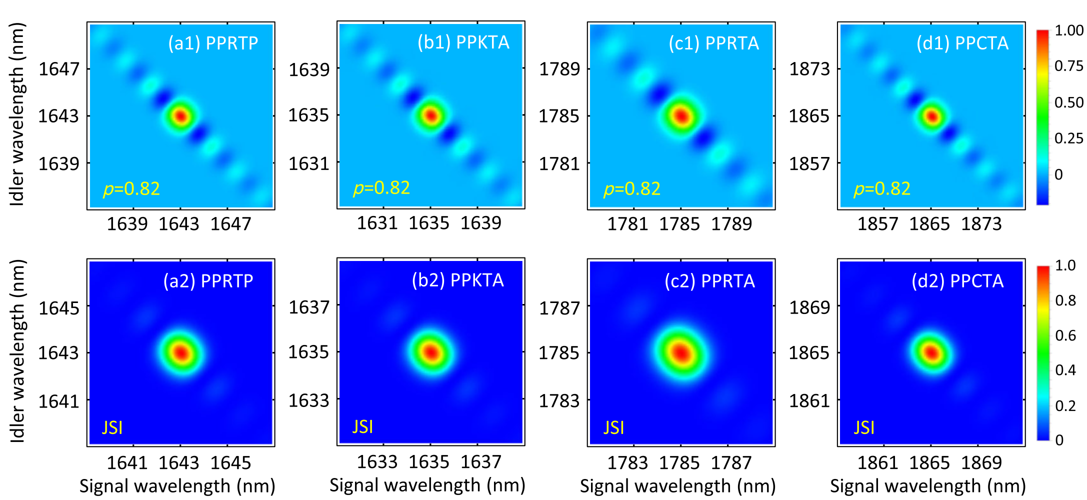

FIG. 1. JSA (first row) and JSI (second row) of the photons generated from the four isomorph crystals at their GVM wavelengths. (a) PPRTP, (b) PPKTA, (c) PRTA, and (d) PPCTA. In this simulation, the crystal lengths are fixed at 30 mm long, while the pump laser bandwidths are 0.42, 0.42, 0.50, and 0.77 nm for (a), (b), (c), and (d), respectively.

Under the GVM condition, the photons from a PPKTP crystal have a high spectral purity of 0.82 at telecom wavelengths. See Ref. [14] for more theoretical details of the spectral purity and the simulation of the JSA for the PPKTP crystal. The same property is maintained by the four isomorph crystals studied here. Figures 1(a1)–1(d1) show the JSA $f(\omega_s, \omega_i)$ of the four crystals at their GVM wavelengths. In this simulation, we fix the crystal length at 30 mm long for the four crystals and scan the pump bandwidth (with a Gaussian spectral distribution) to maximize the spectral purity. It is noteworthy that all four JSAs shown in Figs. 1(a1)–1(d1) exhibit a high spectral purity of 0.82 at their GVM wavelengths. Figures 1(a2)–1(d2) present the corresponding joint spectral intensities (JSIs) $|f(\omega_s, \omega_i)|^2$, which have subcircular shapes. In this simulation, we assume a type II ($o \to o + e$) wavelength-degenerated SPDC process in a collinear configuration along the crystal's x axis. In the process $o \to o + e$, o indicates polarization along the ordinary (the y) axis, while e denotes polarization along the extraordinary (the z) axis. The pump and signal photons have polarization along the y axis, and the idler photons are polarized in the z axis [28].

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 to 1800 nm for the PPRTP crystal; from 1300 to 1700 nm for the PPKTA crystal; from 1400 to 2000 nm for the PPRTA crystal; and from 1500 to 2100 nm for the PPCTA crystal. This range of wavelengths covers the commonly used S, C, L, and U bands in optical-fiber telecommunications. In Figs. 2(a)–2(d), we provide an example of the spectral distribution of the PPRTA crystal at different wavelengths from 1400 to 1700 nm and note that a similar property is also possessed by the other three isomorphs and their parent PPKTP crystal [14].

V. ANOTHER GVM CONDITION IN THE CTA CRYSTAL

While the GVM condition of Eq. (1) is satisfied in all the isomorphs of the PPKTP crystal, a further GVM condition, given by Eq. (4), is satisfied by the PPCTA crystal at 1506 nm:

$$V_{g,p}^{-1}(\lambda/2) = V_{g,i}^{-1}(\lambda). \tag{4}$$

Under this condition, the JSA can have a very narrow and sharp distribution, as shown in Fig. 3(a). In this simulation, we assume a crystal length 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 Figs. 1 and 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 that this condition can be satisfied at telecom wavelengths in a PPCTA crystal.

This source offers great potential in quantum interference between independent sources [1–3,29]. In Fig. 3(c), we simulate the Hong-Ou-Mandel (HOM) interference [30] 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 [1,3,8]. Similarly, Fig. 3(d) shows the case of Hong-Ou-Mandel interference between two idlers heralded by the signal photons. The visibilities in Figs. 3(c) and 3(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 is 0.24 and 3.5 ps, respectively.

While we show only results for PPCTA here, the conditions of $V_{g,p}^{-1}(\lambda/2) = V_{g,i}^{-1}(\lambda)$ and $V_{g,p}^{-1}(\lambda/2) = V_{g,s}^{-1}(\lambda)$ are also satisfied by the other four crystals. As a

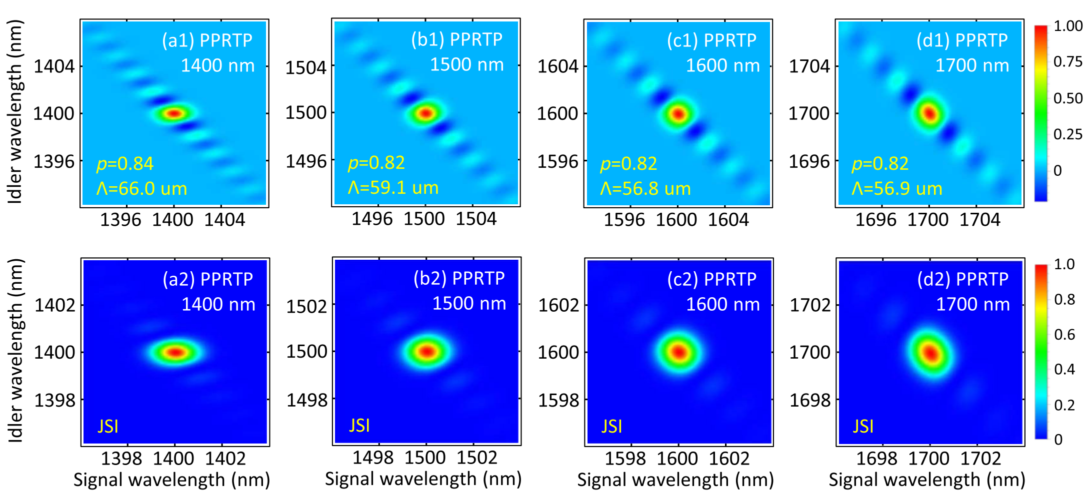

FIG. 2. JSA (first row) and JSI (second row) of photons generated from the PPRTP crystal at different crystal wavelengths. (a) 1400, (b) 1500, (c) 1600, and (d) 1700 nm. The corresponding spectral purity p and poling period Λ are shown in the figure. In this simulation, the crystal lengths are fixed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

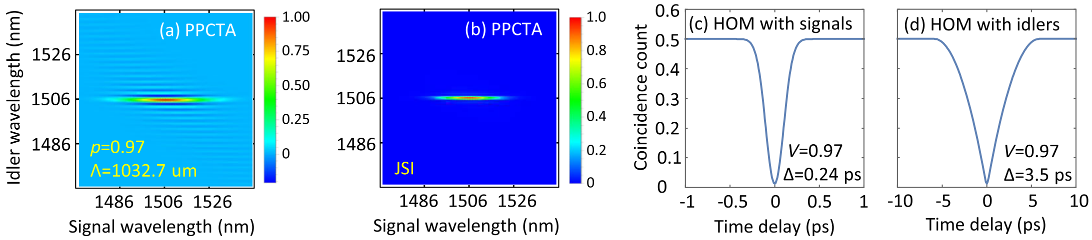

FIG. 3. (a) JSA of the photons generated from the PPCTA crystal at 1506 nm, with a purity of 0.97 and a poling period of 1032.7 μm. (b) JSI of (a). In this simulation, the crystal lengths are fixed at 30 mm long, while the pump laser bandwidth is 5.0 nm with a center wavelength of 753 nm. (c) HOM interference pattern with two heralded signals from (a), with a visibility of 0.97 and a width of 0.24 ps. (d) HOM interference pattern with two heralded idlers from (a), with a visibility of 0.97 and a width of 3.5 ps.

result, a similar figure as Fig. 3(a) can also be achieved by the PPRTP crystal at 1282 and 2491 nm, by the PPKTA crystal at 1278 and 2481 nm, by the PPRTA crystal at 1372 and 2933 nm, and by the PPKTP crystal at 1225 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

The 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. First, in addition to the generation of spectrally decorrelated states shown in Fig. 1, 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 what has been demonstrated in the case of a PPKTP crystal [13]. Second, it is useful to demonstrate customized poling (by modulating the poling order [31], the duty cycle [32], or the domain sequence [33,34]) in these four isomorphs so as to improve the maximal intrinsically purity from 0.82 to near 1. Third, it is also meaningful to make a waveguide based on these four isomorphs, similar as in the case of the PPKTP waveguide [35]. Fourth, the 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 consider only the spectral correlation between the signal and idler photons. It is also interesting to consider the spatial correlation, since the spectral 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 $MM'OXO_4$, where $M = $ K, Rb, Na, Cs, Tl, 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 a highly pure photon source at an arbitrary wavelength.

VII. CONCLUSION

In conclusion, we have theoretically and numerically demonstrated the generation of spectrally uncorrelated states from a small subset of the 118 known isomorphs of the PPKTP crystal. It is 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 to 2100 nm). Furthermore, we find that the PPCTA crystal can achieve an intrinsically high spectral purity of 0.97 at a wavelength of 1506 nm. In the future, these crystals may have many promising applications for quantum-information processing at telecom wavelengths.

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