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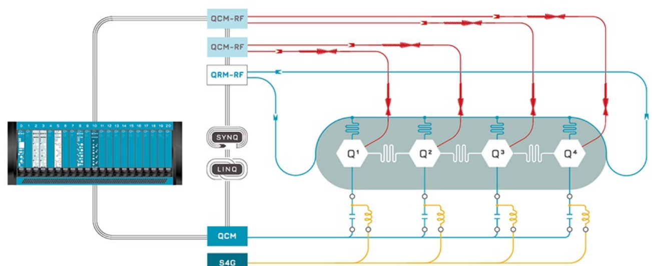

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ABSTRACT

Orbital angular momentum (OAM) of light has been widely investigated in optical manipulation, optical communications, optical storage, and precision measurement. In recent years, the studies of OAM are expanded to nonlinear and quantum optics, paving a way to high-quality nonlinear imaging, high-capacity quantum communication, and many other promising applications. In this Perspective, we first summarize the fundamental research on OAM in nonlinear optics. Then, we introduce its recent applications in nonlinear imaging (including nonlinear spiral imaging and OAM-multiplexing nonlinear holography) and high-dimensional quantum entanglement. In particular, we highlight the manipulations of OAM through various functional nonlinear photonic crystals. Finally, we discuss the further developments of OAM-based nonlinear and quantum techniques in the near future.

I. INTRODUCTION

In 1992, Allen *et al.*[1] proposed that the orbital angular momentum (OAM) of light originates from the phase factor $e^{il\varphi}$, where l is the topological charge (TC) and φ is the azimuth angle. The value of TC can be an arbitrary integer and decides the OAM magnitude and handedness. Compared with the spin angular momentum that is related to the light polarization, an OAM beam features a helical wavefront. Popular OAM-carrying optical beams include Laguerre–Gaussian beams,[1] Bessel beams,[2] Ince–Gaussian beams,[3] spatiotemporal vortex beams,[4] and so on. In particular, the introduction of OAM brings an additional degree of freedom of light for practical applications. For example, OAM modes compose a discrete Hilbert space with infinite dimensions for classic and quantum communications, because these modes are quantized in units of ħ and possess inherent orthogonality.[5] In addition, OAM modes are also very useful in optical tweezers,[6] holography,[7] optical microscopy,[8] and precision measurements of spinning objects.[9,10]

The introduction of nonlinear optics further inspires the fundamental study of OAM. Unlike the above-mentioned linear scenarios, the interacting OAM fields are physically dominated by a nonlinear wave-mixing equation. In 1996, Dholakia *et al.*[11] reported that the second-harmonic (SH) OAM conversion satisfied

$$l_1 + l_1 = l_2 \tag{1}$$

under a birefringent phase matching (BPM) configuration in a KTP crystal. Here, l_1 and l_2 are the TCs of the fundamental and SH waves, respectively. Notably, the nonlinear conversion of OAM can be associated with various degrees of freedom of light such as path, polarizations,[12,13] and radial components[14,15] through polarization control, radial-angular coupling, and other mechanisms.[16] Now, nonlinear manipulations of OAM states attract increasing research interest in the scientific communities ranging from fundamental physics to practical applications. Various nonlinear optical processes, such as second/third-harmonic generation (SHG/THG),[17–19] high-harmonic generation,[20,21] spontaneous parametric downconversion (SPDC),[22–29] terahertz wave generation,[30,31] optical parametric oscillation,[32,33] and cascaded/coupled[34,35] nonlinear process, are utilized to control the OAM conversion. The OAM transfer laws under different mechanisms, such as BPM,[11,36] quasi-phase matching (QPM),[37–39] and spin–orbit coupling,[40–42] are revealed. Aside from the traditional nonlinear materials such as KTP, BBO, and atom gases, it is also an interesting topic to manipulate OAM modes in nonlinear micro/nano-structures such as nonlinear photonic crystals (NPCs) and metasurfaces.[40,41,43–46] Now, the investigations of OAM in nonlinear and quantum optics have inspired the development in many promising fields such as nonlinear holography,[47,48] nonlinear spiral imaging,[49–52] and high-dimensional quantum entanglement.[53–61]

The rapid development in NPCs makes them possible to control the nonlinear interactions in 3D space and sub-micron resolution, which provides a flexible and compact platform to manipulate OAM modes. In 1998, Berger[62] proposed the concept of NPC (featuring a space-dependent second-order nonlinear coefficient, χ^2), which opens the door for nonlinear manipulation of light in high-dimensional space. Based on the QPM theory, NPCs have been widely utilized in laser frequency conversion,[63] nonlinear beam shaping,[64] nonlinear optical imaging,[48,65] and quantum entanglement.[66] The popular way to fabricate 1D and 2D NPCs is to use the electrically poling method,[67] in which the ferroelectric domains in nonlinear crystals such as $LiNbO_3$ and $LiTaO_3$ are selectively inverted by applying an external electric field. However, the experimental realization of 3D NPCs remains a great challenge until 20 years after its first proposal. In 2018, two types of femtosecond laser writing techniques[68,69] were proposed to erase and pole ferroelectric domains, which have been utilized to successfully fabricate 3D χ^2 structures in $LiNbO_3$ and BCT crystals, respectively. In 2022, we developed a nonreciprocal laser writing technique[70] that is capable of fabricating the 3D NPC with its feature size beyond the diffraction limit of light. The NPC provides a powerful platform to control nonlinear interactions. In this Perspective, we briefly present the functions of NPCs in manipulating OAM modes for nonlinear and quantum optic applications.

II. OAM CONVERSION IN NONLINEAR OPTICS

In the work of Dholakia *et al.*,[11] they input an OAM-carrying fundamental beam into a BPM crystal and observed the doubled OAM number at the generated SH wave. In comparison, the walk-off effect can be bypassed in the QPM configuration, which helps improve the performance of nonlinear OAM conversion. After the theoretical deduction,[37] the experimental demonstration was first realized in a 1D periodically poled nonlinear crystal (i.e., 1D NPCs).[38] Since then, the OAM conservation has been verified in various 1D NPC structures. For example, by arranging two kinds of domain blocks in a Fibonacci sequence to compose a quasi-periodically poled nonlinear crystal,[34] the OAM THG process was realized by the coupled SHG and sum-frequency generation (SFG) processes, in which the TCs of the generated SH and third-harmonic (TH) waves were found to be doubled and tripled, respectively, in comparison to that of the input fundamental wave. By designing a scheme of cascading two different 1D domain structures, the OAM of the fundamental wave was transferred into the TH wave in a controllable way[19] [Fig. 1(a)]. Recently, the OAM transfer law has been investigated by inputting various types of OAM-carrying beams. For instance, the SH perfect vortices with an OAM order up to 40 were generated by inputting two nondegenerate fundamental perfect vortices into a 1D NPC.[39] By inputting a fractional OAM-carrying fundamental beam, the OAM components at SH waves were decided by the interactions between the integer components of the fundamental beam.[17] Generally, these early works have demonstrated that OAM transfer in a nonlinear optical process obeys the conservation relationship, i.e., the harmonic wave has a TC value that is the sum of the TCs carried by the interacting fundamental waves.

When high-dimensional NPCs are introduced, the situation becomes quite different because topological domain structures can introduce TCs as well. For example, when performing SHG of an input OAM mode (l_1) in a fork-shaped 2D NPC, the output SH beam has an OAM of

$$l_2 = 2l_1 + ml_c, \tag{2}$$

because the NPC structure contributes a designed TC of l_c into the nonlinear interaction. Here, m is the diffraction order. In this way, high-order SH OAM beams were generated in phase-type[71] and amplitude-type[72] nonlinear fork gratings. One can further introduce a supercell structure to compose a powerful Dammann fork-grating NPC, which is capable of controlling the energy distribution and TCs at each SH diffraction order[73] [Fig. 1(b)]. Another type of topological NPC features a spirally poled domain structure, which has been used to introduce or detect the OAM number at SH waves.[74] In addition, taking the advantage of abundant reciprocal vectors, the function of 2D NPCs is extended to achieve multiple copies of SH OAM modes.[75] In recent years, the emergence of 3D NPC provides a promising platform to manipulate nonlinear optical processes in 3D space. For example, one can use a three-layer 3D NPC to compose a multi-functional mode converter,[76] which transforms a fundamental Gaussian beam into an SH vortex beam, a Gaussian beam, and a conical beam selectively. If the layer number is increased to 30, one can demonstrate efficient nonlinear beam shaping. Because the requirements of nonlinear wave-front shaping and quasi-phase-matching can be simultaneously fulfilled, the conversion efficiency of SH OAM beams is enhanced by two orders of magnitude in comparison to the 2D case[64] [Fig. 1(c)]. When larger-scale 3D NPCs are fabricated, one can expect 3D integrated photonic devices with more powerful functions.

III. OAM-ENABLED NONLINEAR IMAGING TECHNIQUES

The unique spiral phase in an OAM beam plays an important role in nonlinear optical imaging. One example is nonlinear spiral phase contrast (SPC) imaging for edge enhancement at up-converted frequencies. The nonlinear SPC scheme is equivalent to place a nonlinear spatial filter at the Fourier plane under a 4f optical configuration. In experiment, a type-II KTP crystal was first exploited to demonstrate nonlinear SPC imaging.[49] Now, its function has been significantly expanded according to the advanced requirements of nonlinear imaging. For example, an wide-field-of-view nonlinear SPC imaging was achieved by modulating the temperature of the nonlinear crystal.[50] Up-conversion SPC imaging at a single-photon level was realized in the mid-infrared region[51] [Fig. 2(a)]. By utilizing a vecotrial nonlinear vortex filter, one can obtain selective edge enhancements depending on the polarization of the invisible illumination.[52]

Another important application is OAM-multiplexing nonlinear holography in NPCs. The capacity of linear optical holography can be effectively enhanced by utilizing the wavelength, polarization, and incident angle of light.[7,43,77] However, it is difficult to effectively enhance the capacity of nonlinear holography because of the non-negligible crosstalk from nonlinear wave coupling.[78,79] The OAM of light introduces an extra phase-matching condition, which provides an efficient way to suppress the crosstalk. In experiment, one can encode the OAM-multiplexing nonlinear hologram into the interacting fundamental waves or the NPC structures[47,48] [Fig. 2(b)]. Then, the output images at harmonic waves can be selectively reconstructed through

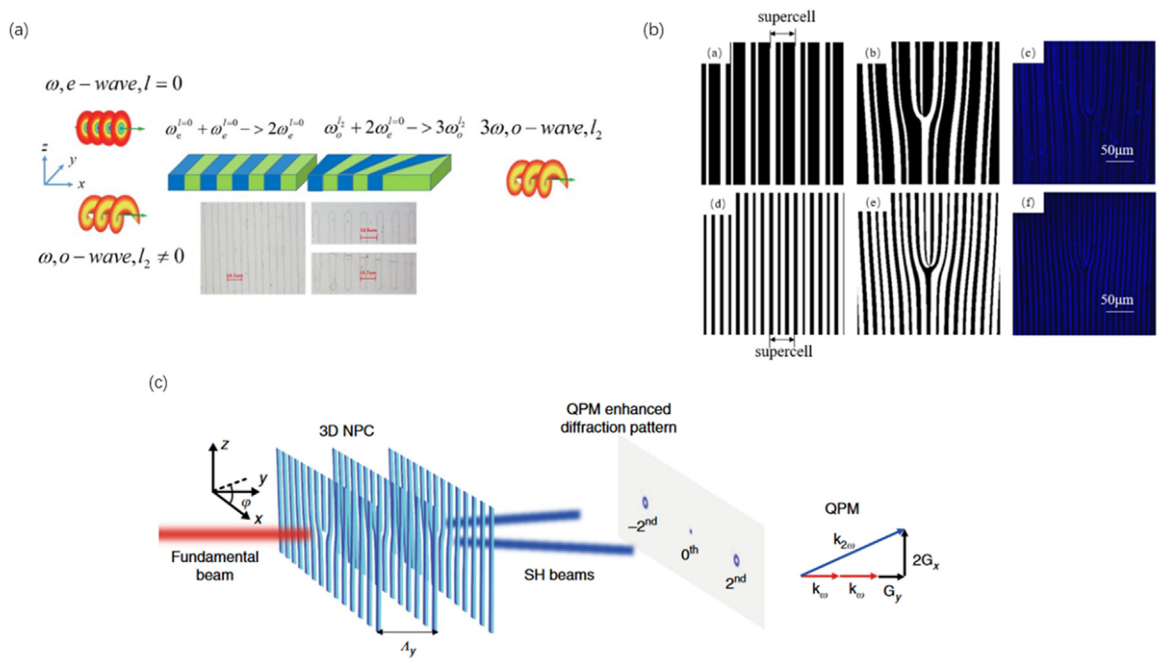

FIG. 1. OAM conversion under various NPC configurations. (a) Illustration of two cascaded 1D NPCs for the generation of TH OAM beams. Reproduced with permission from Ref. 23. (b) Typical 2D structures of supercell vortex gratings. Reproduced with permission from Ref. 21. (c) Efficient generation of SH OAM beams in a 3D nonlinear fork-grating array. Reproduced with permission from Ref. 60.

inputting the corresponding fundamental OAM beam. In this way, one can significantly enhance the capacity of nonlinear holography.

IV. MANIPULATING THE OAM SPECTRUM IN QUANTUM OPTICS

In 2001, Mair *et al.*[22] experimentally realized the OAM entanglement for two-photon states generated through a SPDC process. Since then, the OAM correlations in quantum optics have been extensively investigated such as OAM anti-correlation between signal and idle photons,[23] coincidence detection of OAM entanglement by Hong-Ou-Mandel interference,[24] OAM conservation in the entire downconversion cone,[25] and so on. The OAM degree of freedom gives new insight into quantum optics, particularly facilitating the generation of quantum entanglement in high dimensions. In principle, the signal-idler spiral bandwidth is bound to transverse correlations[26] related to the radial and azimuthal index of pump light, and the width ratios

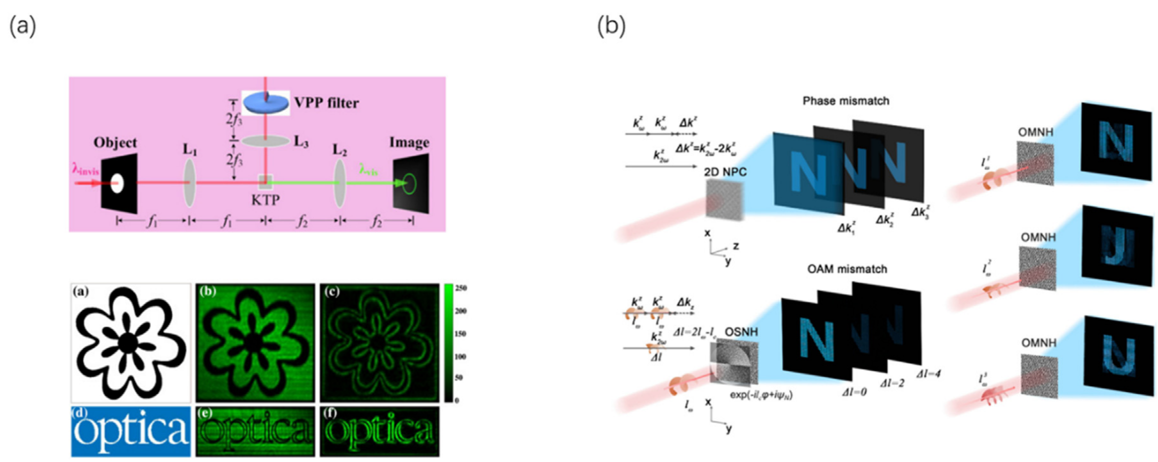

FIG. 2. (a) Schematic illustration and experimental results of nonlinear SPC imaging. Reproduced with permission from Ref. 49. (b) Schematic diagram of OAM multiplexing nonlinear holography. Reproduced with permission from Ref. 47.

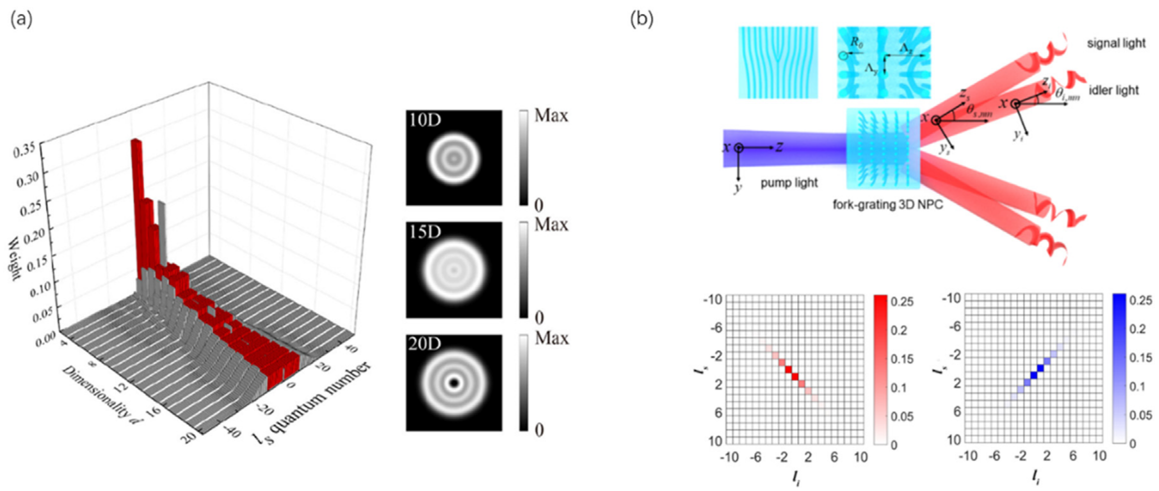

FIG. 3. Manipulating the OAM spectrum. (a) In the pump beam shaping method, one can optimize the radial components of the input modes to realize high-dimensional OAM MES. Reproduced with permission from Ref. 53. (b) By using the 3D fork grating array, one can achieve positive or negative OAM correlation. Reproduced with permission from Ref. 56.

between the pump and signal/idler beams. In experiment, the entanglement of two photons[27] was generated with an OAM number up to 600. In addition, the studies on OAM correlation were expanded to four-photon and higher-dimensional multipartite states.[28,29]

One important application of OAM in quantum optics is to compose a high-dimensional maximally entangled state (MES). It is well known that high-dimensional MES is of great help in enhancing the capacity[80,81] and robustness[82,83] of quantum communications. In 1996, the Procrustean method[84] was theoretically proposed to extract high-dimensional MES out of non-maximally entangled states through postselection. However, it is a great challenge to efficiently produce high-dimensional MES.[22,53] OAM entanglement provides a potential solution to realize high-dimensional MES through tailoring the amplitudes of various OAM components.

One way to manipulate the OAM spectrum is pump beam shaping. In principle, one can generate arbitrary maximally OAM-entangled states by manipulating the pump topology.[53] In experiment, one useful way is to utilize the superposition of multiple LG modes as a pump light. After optimizing the LG components, 3D and 4D OAM-based MESs have been successfully prepared.[54,55] In addition to shaping the azimuthal components of LG pump light, one can also manipulate the pump radial components to achieve high-dimensional OAM-based MESs with high fidelity[56] [Fig. 3(a)]. The typical algorithms to optimize LG pump modes include simultaneous perturbation stochastic approximation algorithms and gradient descent and simulated annealing algorithms. Recently, a basis-independent approach[57] has been developed to shape the pump profile in any basis as well as to improve the signal-to-noise ratio. The other way is to introduce a proper NPC structure to manipulate the OAM spectrum, which is capable of improving the conversion efficiency. Several theoretical schemes have been reported by using different NPC structures. For example, nonlinear wave-front engineering in 2D NPCs was proposed to generate a two-photon OAM-based N00N state.[58] The OAM spectrum features different distributions in 2D fork-grating NPCs with various TCs.[59] Recently, the manipulation of the OAM spectrum was extended to 3D NPC structures (including 3D fork gratings[60] and 3D spiral structures[61]) in theory. As shown in Fig. 3(b), the OAM correlation is positive (or negative) for counter-propagation (or co-propagation) in a 3D fork-grating NPC.

The pump shaping technique can effectively broaden the spiral bandwidth for generation of high-dimensional MESs. However, it usually introduces unwanted modes and requires extra diagonalization of the OAM detection bases. In comparison, one can well suppress these unwanted modes by properly optimizing the NPC structure. It is worth expecting the experimental realization of OAM spectrum manipulation in 3D NPCs.

A. Prospects

OAM of light has been extensively investigated in the past few decades. Novel concepts such as spatiotemporal OAM states[4] are emerging. Potential applications have been extended to optical holography, precision measurement, and optical storage. In nonlinear optics, there still exist many remaining challenges. For example, previous works are mainly focused on nonlinear manipulations of low-order OAM states. The generation, evolution, and conversion of high-order OAM states during a nonlinear optical process may present very different phenomena beyond the current physical mechanisms. Particularly, nonlinear manipulation of the OAM spectrum in NPCs[85,86] is a promising topic in recent years. In 2020, it was experimentally demonstrated that the OAM spectrum generated from 1D NPCs strongly depends on the phase-matching conditions.[85] However, it is still difficult to efficiently control the OAM spectrum in NPCs for quantum applications. The major obstacle is that 1D and 2D NPCs cannot simultaneously realize high-efficiency SPDC and effective wave front engineering of signal/idler beams. The recent developments of femtosecond laser writing techniques[70] make it possible to fabricate 3D NPC, which may provide enough reciprocal vectors to facilitate the experimental demonstrations.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Sixin Chen and Taxue Ma contributed equally to this work.

Sixin Chen: Writing – original draft (equal); Writing – review & editing (equal). **Min Xiao:** Funding acquisition (supporting); Writing – original draft (supporting). **Yong Zhang:** Funding acquisition (lead); Supervision (lead); Writing – original draft (equal); Writing – review & editing (equal). **Taxue Ma:** Writing – original draft (equal). **Qian Yu:** Writing – original draft (supporting). **Pengcheng Chen:** Writing – original draft (supporting). **Xinzhe Yang:** Writing – original draft (supporting). **Xuewei Wu:** Supervision (supporting); Writing – review & editing (equal). **Hai Sang:** Supervision (supporting). **Xiaopeng Hu:** Writing – original draft (supporting). **Shining Zhu:** Supervision (supporting).

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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