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## Fragmentation of twisted light in photon–phonon nonlinear propagation

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Twisted light, or orbital angular momentum (OAM) carrying light, has been gradually becoming an important subfield of nonlinear optics. Compared with ordinary light, its chiral phase front provides an additional interface for shaping the phase-matching condition of nonlinear interactions and in consequence reveals a feasible way to tailor light's transverse structure. Here, we explore the nonlinear propagation of twisted light during focused stimulated Brillouin scattering (SBS). Unlike ordinary light that will experience a time-reversal nonlinear reflection, OAM carrying light will break up into corresponding petal-like degenerate OAM modes that carry no net OAM, whereas the superposed OAM modes that carry no net OAM, as the input field, are still time-reversed in focused-SBS. This unexpected phenomenon, resulting from a unique OAM selection rule of noise-initiated SBS, gives more insight into the underlying principle of OAM conservation in electromagnetic interactions and provides an approach to shaping light via nonlinear propagation. *Published by AIP Publishing*. https://doi.org/10.1063/1.5020082

Twisted light, as a very frequent hot topic in optics over the past decade and as an emerging important subfield of modern optics,<sup>1-3</sup> refers to the generation and application of light beams or photons with a helical phase front, and it has led to many scientific advances throughout the photonics domain.<sup>4–13</sup> In addition to containing more information per photon, a quintessential attribute of orbital angular momentum (OAM) carrying light, its helical wave front can provide a chiral interface, i.e., a pair of complementary modes, for light-matter and light-light interactions. The chiral interface and the requirement for system rotational symmetry codetermine the OAM selection rule of light (matter) fields during nonlinear interactions.<sup>14-16,30</sup> More remarkably, in addition to governing the OAM selection rule, this chiral interface can also tailor the phase-matching condition profoundly and in consequence provides a new mechanism for shaping the transverse profiles of light fields via nonlinear propagation.<sup>17–22</sup> For example, even in saturated self-focusing-type nonlinear media, donut-twisted light carrying  $\ell$  will break up into superposed modes with opposite topological charges ( $2\ell$ filaments) that carry no net OAM,<sup>21</sup> and spin-orbital coupled light carrying net OAM will also break up into corresponding light that carries no net OAM.<sup>22</sup> These regular fragmentations can be concluded to be dynamic behaviors arising from a fact that two complementary OAM modes have the same nonlinear gain or loss rate in a certain interaction.

In addition to all-optical interactions, the fragmentations originating from OAM conservation and spin-orbital coupling have also been explored in all-matter interactions, such as cold-atom and Bose-Einstein systems with weak interactions.<sup>23,24</sup> Then, it is straightforward to infer that the fragmentation could exist in light-matter interactions as well. Recently, due to their value in scientific applications, nonlinear interactions between OAM photons and electrostatic quasi-particles (phonons or plasmons) via stimulated Raman scattering (SRS) and stimulated Brillouin scattering (SBS) have received intense attention.<sup>25-30</sup> The theories of SBS and SRS were first extended to involve OAM lasers by Mendonça et al. with plasma as nonlinear media.<sup>25</sup> Later, we experimentally observed the exchange of OAM between photons and phonons via all-parametric SBS.<sup>30</sup> In this letter, we report the fragmentation of OAM carrying light in photon-phonon nonlinear propagation, in which the breakup mechanism of Stokes light originating from a twisted pump in the SBS timereversal system-spontaneous OAM transfer from photons to phonons via noise-initiated SBS-is revealed.

Time-reversal (or phase-conjugate) operations have been broadly studied in generalized waves, such as acoustic, electromagnetic, and water waves.<sup>31–33</sup> For light waves, especially for laser beams, nonlinear wave couplings are common solutions.<sup>34–36</sup> Among these, the phase conjugate mirror (PCM) based on focused-SBS, discovered by Boris Zel'dovich in 1972,<sup>37</sup> is the most accessible method and has been widely adopted in commercial high-energy and single-mode laser systems for pulse compression and correcting amplifier aberrations. The term SBS generally refers to strong photon–phonon couplings in the case where the phonons are coherently generated via optical forces (beating fields). It can be described as a forward pump light  $E_p = A_p \exp [ik(\omega_p)z]$  beating with a backward Stokes light  $E_s = A_s \exp [-ik(\omega_s)z]$  and exciting a forward acoustic wave  $\rho(z, t) \exp [iq(\Omega)z]$ , where  $k(\omega)$  and  $q(\Omega)$ 

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are the optical and acoustic dispersion relations, respectively, and the corresponding wave equation may be expressed as  $^{36}$ 

$$\rho(z,t) = \kappa A_p A_s^* \exp\left[i\Delta kz\right],\tag{1}$$

where  $\kappa$  is the coupling coefficient and  $\Delta k = q(\Omega)$  $-k(\omega_p) - k(w_s)$  is the degree of phase-mismatching. Note that there are two types of SBS: one is an all-parametric process, or rather stimulated Brillouin amplification (SBA), where the coherent phonons are excited by pre-existing pump light  $E_p$  and Stokes light  $E_s$ ; the other is a noise-initiated parametric process,<sup>38</sup> e.g., focused-SBS, in which only one pump light is applied externally and the Stokes light is selected and fed from spontaneous Brillouin scattering noise by a trailing pump. In the case of focused-SBS, the Stokes photons originate from spatially distributed noise emitted in diffuse directions. Considering the narrow-linewidth feature of SBS and the fact that Brillouin gain depends only on the laser intensity (not its phase), the greatest Brillouin gain occurs only in the backward direction, in which the focusedpump and Stokes light overlap exactly (perfect phase matching). As a result, a backward copy (time-reversal) of the incident pump with a Stokes-frequency shift is created, i.e., the created Stokes light  $E_s \approx E_p^*$ .

The SBS process involving OAM carrying light can be described as double-twisted lights,  $E_p = A_p \exp [ik(\omega_p)z + i\ell_p\theta]$  and  $E_s = A_s \exp [-ik(\omega_s)z - i\ell_s\theta]$ , beating with each other and exciting a twisted acoustic wave  $\rho(z, t) \exp [iq(\Omega)z + i\ell_p\theta]$ , where  $\ell_p$ ,  $\ell_s$ , and  $\ell_\rho$  are the topological charges of the pump, Stokes, and coherent phonons, respectively. Notice that the plus-minus sign of the topological charges is defined by phase chirality along the propagation direction. Thus, Eq. (1) in this case,  $E_p$  and  $E_s$  counter-propagating with each other, can be rewritten as

$$\rho(z,t) \exp\left[i\ell_{\rho}\theta\right] = \kappa A_{\rho}A_{s}^{*} \exp\left[i\Delta kz\right] \exp\left[i(\ell_{\rho} + \ell_{s})\theta\right], \quad (2)$$

and the complete momentum conservation relationship of SBS including the OAM selection rule can be expressed as<sup>30</sup>

$$|\mathbf{q}(\Omega);\ell_{\rho}\rangle = |\mathbf{k}(\omega_{p});\ell_{p}\rangle - |\mathbf{k}(\omega_{s});\ell_{s}\rangle \approx |2\mathbf{k}(\omega_{p});\ell_{p}+\ell_{s}\rangle,$$
(3)

where the approximation  $k(\omega_p) \simeq k(\omega_s)$  arises because of the Stokes frequency far less than light. Note that the Stokes OAM mode  $\ell_s$  is pre-existing in *all-parametric* SBS, i.e.,  $\exp[i(\ell_p + \ell_s)\theta]$  corresponding to the beating of two input vortex lasers, while in the noise-initiated SBS, only one vortex light  $\ell_p$  is pre-existing, and the Stokes light  $\ell_s$  is selectively amplified from the noise by the input focused pump. Now, we further consider the *noise-initiated* SBS driven by the OAM pump, i.e., the pump  $E_p = A_p \exp [ik(\omega_p)z + i\ell_p\theta]$ is input alone. Due to the intensity-dependent gain of SBS, donut Stokes light that exactly overlaps the incident pump light will obtain the greatest Brillouin gain and be selected from the noise. However, this donut transverse profile corresponds to a pair of complementary OAM modes  $\pm \ell$ , and this phenomenon can be understood from the fact that the transverse profiles of twisted light, such as Laguerre-Gaussian (LG) modes or hypergeometric Gaussian modes<sup>40,41</sup> [for details, see Eq. (S2) in the supplementary material], are actually the functions of  $|\ell|$ , i.e.,  $E_{\ell} = A(z, r, \theta, |\ell|) \exp[ik(\omega)z + i\ell\theta]$ . That is to say, two complementary modes,  $+\ell_p$  and  $-\ell_p$ , counter-propagating with the incident pump  $\ell_p$  will be selectively amplified from the noise, finally creating a fragmented Stokes light with  $2\ell_p$  petals. On this basis, the conservation of OAM within the whole system will further lead to an OAM transfer from light to sound during the creation of petal-like Stokes light. Specifically, according to Eqs. (2) and (3), the incident  $\ell_p$  mode pump light selectively amplifies the  $\pm \ell_p$ mode and Stokes light will excite coherent phonons of  $0 + 2\ell_p$ mode simultaneously. Figure 1(a) shows the simulated transverse profiles of the pump/Stokes light and acoustic wave during focused-SBS propagation based on Eqs. (2) and (S2). Moreover, a pump light carrying an equal and opposite OAM (such as  $\ell_p = \pm 1, \pm 2, ...$ ), i.e., one which already has a complementary superposed OAM mode, will still be time-reversed in the focused-SBS. In this case, although no net OAM will be transferred from the pump to the phonons in the noise-initiated SBS, according to Eqs. (2) and (3), the excited phonons will carry a transverse structure of the  $1/\sqrt{2}|0\rangle + 1/\sqrt{4}|+2\ell_p\rangle$  $+1/\sqrt{4}|-2\ell_p\rangle$  mode, as shown in Fig. 1(b).

The experimental setup is shown in Fig. 2; a 532 nm single-longitudinal-mode pulse light (0.5 mJ @ 10 ns) is used as the pump source and a combination of wave-plates and a q-plate (for details, see Ref. 39) converts it into the desired twisted light. After propagation in the  $z_1$  direction after generation, it passes through a "time-reversal mirror" that consists of a polarization splitting prism (PBS), a lens (f = 150 mm), a quarter-wave plate (QWP), and an SBS-cell containing CS<sub>2</sub> (SBS active medium). Then, the output Stokes waves are reflected from the PBS for further analysis. In addition, a weakly focused Gaussian pulse with V-polarization brought from the same light source is directed into the focal region of the input pump from the other side of the coupling cell. The probe light interacts near-collinearly with the coherent phonons excited in the previous noise-initiated SBS and generates a diagnostic light with anti-Stokes-frequency shift for analyzing the excited phonons.



FIG. 1. Simulated results of twisted light in the *noise-initiated* SBS. Transverse profiles of input pump light (a1) and (b1), reflected Stokes light (a2) and (b2), and excited phonon waves (a3) and (b3).



FIG. 2. Schematic illustration of the experimental setup. (a) Experimental setup. Key components include the Q-plate, quarter-wave plate (QWP), polarized beam splitter (PBS), coupling cell, lens, and mirror. (b) Energy level and waveform diagram of focused-SBS.

In the experiment, first, a set of twisted pumps carrying a single topological charge  $(\ell_p = 4, 6, 8)$  with Hpolarization, converted from the LG<sub>00</sub> mode by q-plates, are injected into the SBS coupling cell, respectively, as shown in Figs. 3(a1)-3(a3). The residual pump light shown in Figs. 3(b1)-3(b3) maintains the transverse structure compared with input states, indicating that the pump itself does not undergo breakup during the SBS propagation. The petal-like transverse profiles shown in Figs. 3(c1)-3(c3) indicate that the Stokes light breaks up into  $\ell = \pm 4$ ,  $\ell = \pm 6$ , and  $\ell = \pm 8$ modes, respectively, as theortically predicted above. Figures 3(d1)-3(d3) show broken-up diagnostic light that corresponds to  $\ell = 0 + 8$ ,  $\ell = 0 + 12$ , and  $\ell = 0 + 16$  modes, respectively. That is to say,  $4\hbar$ ,  $6\hbar$ , and  $8\hbar$  net OAM per photon are transferred from the light to sound, respectively, via the noise-initiated SBS. In particular, the nonsymmetrical intensity profiles shown in Figs. 2(d1)-2(d3) are due to the following reasons: on the one hand, the spatial modes of coherent phonons excited in this case are focally distributed along the z-axis and the probe can only explore a partial region; on the other hand, the noncollinear parametric process in the probe-phonon interaction leads to the diagnostic

light carrying off-axis vortex states.<sup>30</sup> Second, a set of superposed twisted lights carrying equal and opposite topological charges ( $\ell_p = \pm 2, \pm 3$ ) with H-polarization are used as the pump. The results shown in Figs. 4(a) and 2(b) verified the above prediction again: specifically, the time-reversal is conserved, and the broken-up diagnostic light carrying  $1/\sqrt{2}|0\rangle + 1/\sqrt{4}|+4\rangle + 1/\sqrt{4}|-4\rangle$  and  $1/\sqrt{2}|0\rangle + 1/\sqrt{4}|+8\rangle + 1/\sqrt{4}|-8\rangle$  modes.

The OAM transfers from electromagnetic waves to electrostatic waves via SBS or SRS reported in previous work are all driven by pre-existing light beating fields,<sup>25–30</sup> i.e., two lasers with a relative Stokes-frequency difference and at last one carrying net OAM. Whereas in this work, the OAM transfer from light to sound is driven by a *noise-initiated* parametric process; to be specific, two complementary OAM modes contained in the noise have the same transverse profile  $A_z(x, y)$  and longitudinal wave vector  $k_z(\omega)$  at a given propagation point; therefore, they obtain the same SBS gain and rapidly grow up from the spontaneous Brillouin scattering noise. This unique attribute, i.e., the two complementary OAM modes having the same nonlinear gain, can also give rise to other analog nonlinear fragmentation. The difference



FIG. 3. Experimental results. (a1)–(a3) Observed transverse profiles of input pump light  $\ell_p = 4, 6, 8$  and (b1)–(b3) corresponding residual pump lights. (c1)–(c3) Observed fragmented Stokes lights from the OAM carrying pump and (d1)–(d3) corresponding diagnostic lights.



FIG. 4. Experimental results. (a) and (b) The time-reversal reflection of the  $\ell = \pm 2$  and  $\ell = \pm 3$  modes, respectively, in which the first line shows the input pump light and the second line shows the time-reversed Stokes light, and the corresponding residual pump and diagnostic lights are shown on the right-hand side.

is, in this experiment, the breakup Stokes light close to standard  $\pm \ell$  modes because they only experience once nonlinear selective gain; in contrast, the beam collapse occurring in self-focusing media tends to break up into thin filaments due to multiple nonlinear selection.<sup>19,20</sup> Notice that although the noise-initiated Brillouin PCM fails to time-reverse OAM carrying light, other all-parametric nonlinear PCM, such as four-wave mixing,<sup>34-36</sup> can still work for twisted light because they do not experience the mode-selection processes from the noise. In addition, it is important to note that the OAM selection rules (or rather OAM conservation) of electromagnetic interaction are actually governed by the system rotational symmetry; therefore, in addition to nonlinear interactions, this unique attribute can also play a role in other optical processes. For instance, a normal laser generator usually can only produce structured light that carry no net OAM, such as Hermite-Gaussian  $(\pm \ell)$  and vector vortex modes, because the two complementary OAM modes have the same gain rate in the laser resonator. In other words, the role of the focused donut pump in focused-SBS is equivalent to the resonant cavity of a laser generator, which high efficiently and selectively amplify a certain complementary OAM pair  $\pm \ell$  from the noncoherent noise.<sup>40,41</sup>

In summary, we have experimentally identified and explored the fragmentation of twisted light during nonlinear time-reversal propagation based on focused-SBS. Because two complementary OAM modes, carrying equal and opposite topological charges,  $+\ell$  and  $-\ell$ , have the same transverse profile at a given propagation point, they thus have the same Brillouin gain offered by a certain pump. This underlying mechanism makes donut-twisted light carrying  $\ell$  break up into  $2\ell$  petals during time-reversal propagation based on focused-SBS: that is, it is converted into a corresponding degenerate OAM mode that carries no net OAM. This phenomenon can also be realized in other nonlinear downconversion processes: for example, when using an OAM carrying pump field to amplify a spontaneous emission noise (transverse uncorrelated) via optical parametric amplification, such as SBA, stimulated Raman amplification (SRA), and difference frequency generation (DFG), the corresponding degenerate OAM mode contained in the noise field will be selected and amplified.

See supplementary material for wavefunctions of twisted light generated via phase-only OAM manipulation.

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