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Sequential harmonic spin–orbit angular momentum generation in nonlinear optical crystals

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Section 1: Sequential optical spin–orbit interactions of second harmonic waves in two BBO crystals

As shown in Fig. S1(a), the circularly polarized fundamental waves (FWs) with angular momentum state of $(\sigma, \ell)_{FW}$ is focused into BBO1, which, through the spin–orbit interaction (SOI) process, is converted to a new angular momentum state with conversion efficiency close to 50%. Thus, two angular momentum states of FWs with nearly equal energy are prepared after BBO1. Meanwhile, four angular momentum states of second harmonic waves (SHWs) are generated from the FWs through the second harmonic generation (SHG) process and SOI process. However, the two angular momentum states of SHWs from the SOI-induced FWs are very weak. Then the SHWs generated in BBO1 are all filtered out. As shown in Fig. S1(b), by using the assembly of polarization optics, the spin angular momentum of the angular momentum states of FWs are flipped. As a result, totally four kinds of angular momentum states of FWs with the same energy are prepared and focused into BBO2. Through the SHG process and the SOI process, totally eight different angular momentum states of SHWs are generated, which are with equal energies.

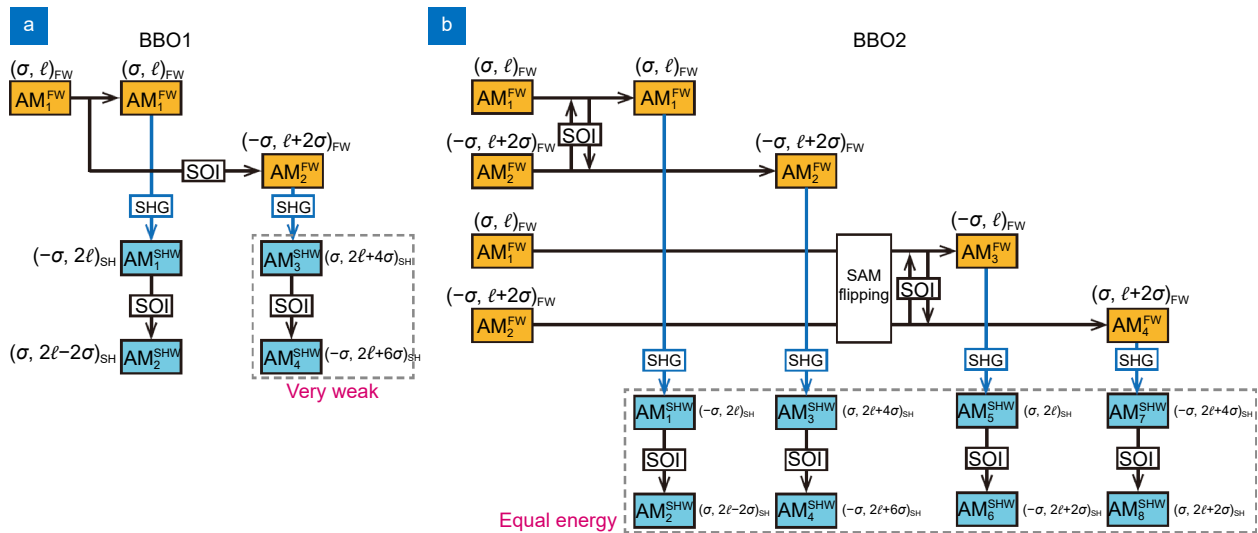


Fig. S1 | Schematic diagram of the evolution process of the fundamental waves (FWs) and second harmonic waves (SHWs) with multiple angular momentum states through sequential optical spin–orbit interactions (SOIs) in two BBO crystals with threefold rotational symmetry. (a) Evolution process in BBO1. For a circularly polarized FW with angular momentum state of $(\sigma, \ell)_{FW}$, two states of the FWs are generated in BBO1 through the optical SOI process. Four states of the SHWs are generated in BBO1 through the second harmonic generation process and SOI process, however, two states of the SHWs are very weak in experiment. The SHWs generated in BBO1 are filtered out by using a long-pass filter. (b) Evolution process in BBO2. By flipping the spin angular momentum of the FWs before the second BBO crystal, two new angular momentum states of the FWs are obtained. The four kinds of FWs are then refocused into the BBO2, where the SOI process of the FWs occurs without new states being generated. In BBO2, SHWs with four angular momentum states will be generated in the direct channels. Through a sequential SOI process of the SHWs in the second BBO crystal, four new angular momentum states are obtained. It is expected that these eight kinds of angular momentum states of the SHWs have nearly equal energies.

Section 2: Experimental setup of the double BBO crystal system

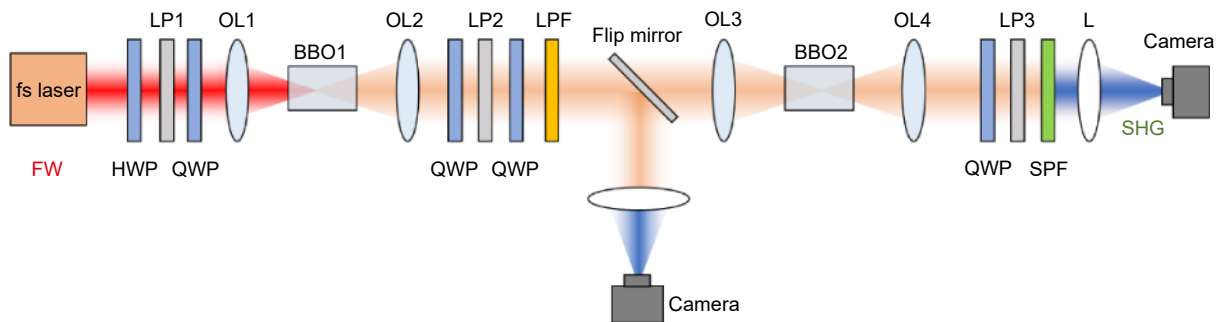


Fig. S2 | Schematic illustration of the double BBO crystal system composed of two confocal optical sections, by which the intensity profiles of the FWs and SHWs are captured. HWP, half-wave plate; LP, linear polarizer; QWP, quarter-wave plate; OL, objective lens; LPF, long-pass color filter; SPF, short-pass color filter; L, tube lens (spherical lens or cylindrical lens).

Section 3: Nonlinear optical spin–orbit interactions in a single BBO crystal

The captured intensity profiles of the FWs and SHWs behind the first BBO crystal are shown in Fig. S3, in which the mechanism of the nonlinear optical spin–orbit interactions and the evolution of the angular momentum of photons are also demonstrated. The combinations of circular polarization components in Fig. S3 and Fig. S4 are denoted by ‘LL’, ‘LR’, ‘RR’ and ‘RL’, where the first denotes the circular polarization states of the FW before the BBO crystal, and the second character represents the state of the FW or SHW after the BBO crystal.

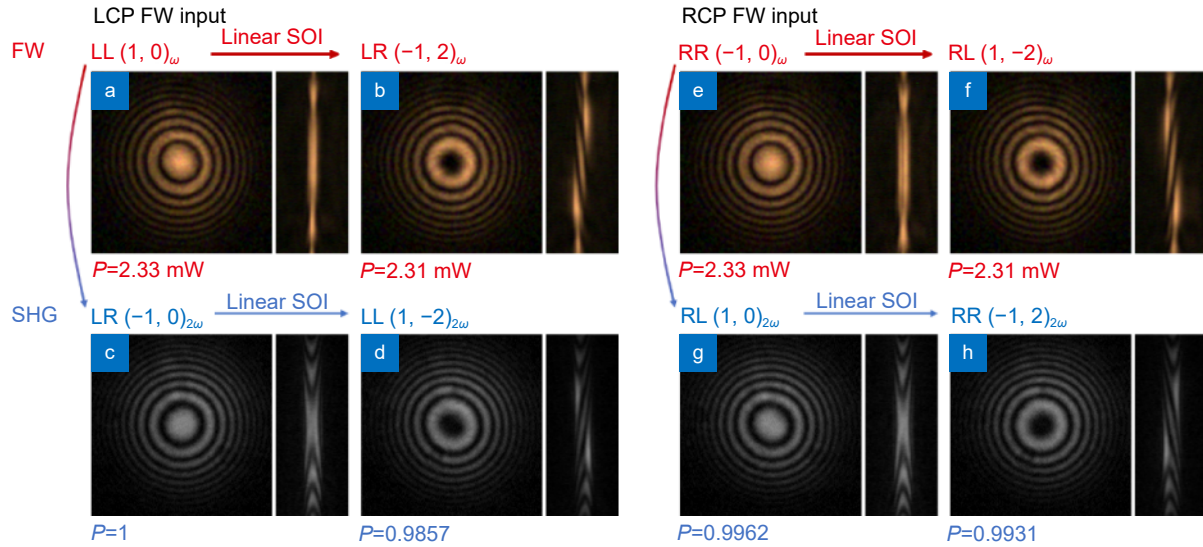


Fig. S3 | Evolution of the spin and orbital angular momentum states of the fundamental waves (FWs) and second harmonic waves (SHWs) after the first BBO crystal. The intensity profiles of the FWs and SHWs under different combinations of circularly polarized components are measured (square images). Intensity profiles imaged by a cylindrical lens are used to identify the orbital angular momentum values (rectangle images) of light. The power of the FWs is measured by using a power meter. The relative power value of the SHWs is obtained by integrating the SHG spectra (shown in Fig. S4), which is recorded by using an Andor spectrometer (SR193i).

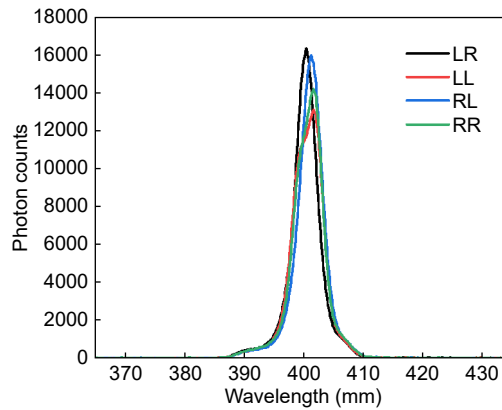


Fig. S4 | Spectra of the second harmonic waves after the first BBO crystal under different combinations of the circularly polarized components. In the LL’, ‘LR’, ‘RR’ and ‘RL’ measurements, the first and the second characters denote the circular polarization states of the fundamental wave and second harmonic wave, respectively.

Section 4: Angular momentum states of the fundamental waves in the double BBO crystal system

The captured intensity profiles of the FWs behind the second BBO crystal are shown in Fig. S5. The combinations of circular polarization components in Fig. S5 are denoted by ‘LLLL’, ‘LLLR’, ‘LLRR’, etc. The first and third characters denote the circular polarization states of the FW before BBO1 and BBO2, and the second and fourth ones denote the circular polarization states of the FW after them. The sixteen angular momentum states of the FWs are listed in Fig. S5, however, it should be noted there are several degenerate ones, such as the states in Fig. S5(a, f). As indicated by the red arrow, the states in the second (Fig. S5(b, f, j, n)) and fourth (Fig. S5(d, h, i, p)) columns are generated by those in the first (Fig. S5(a, e, i, m)) and third (Fig. S5(c, g, k, o)) columns, respectively, through the linear optical SOI process.

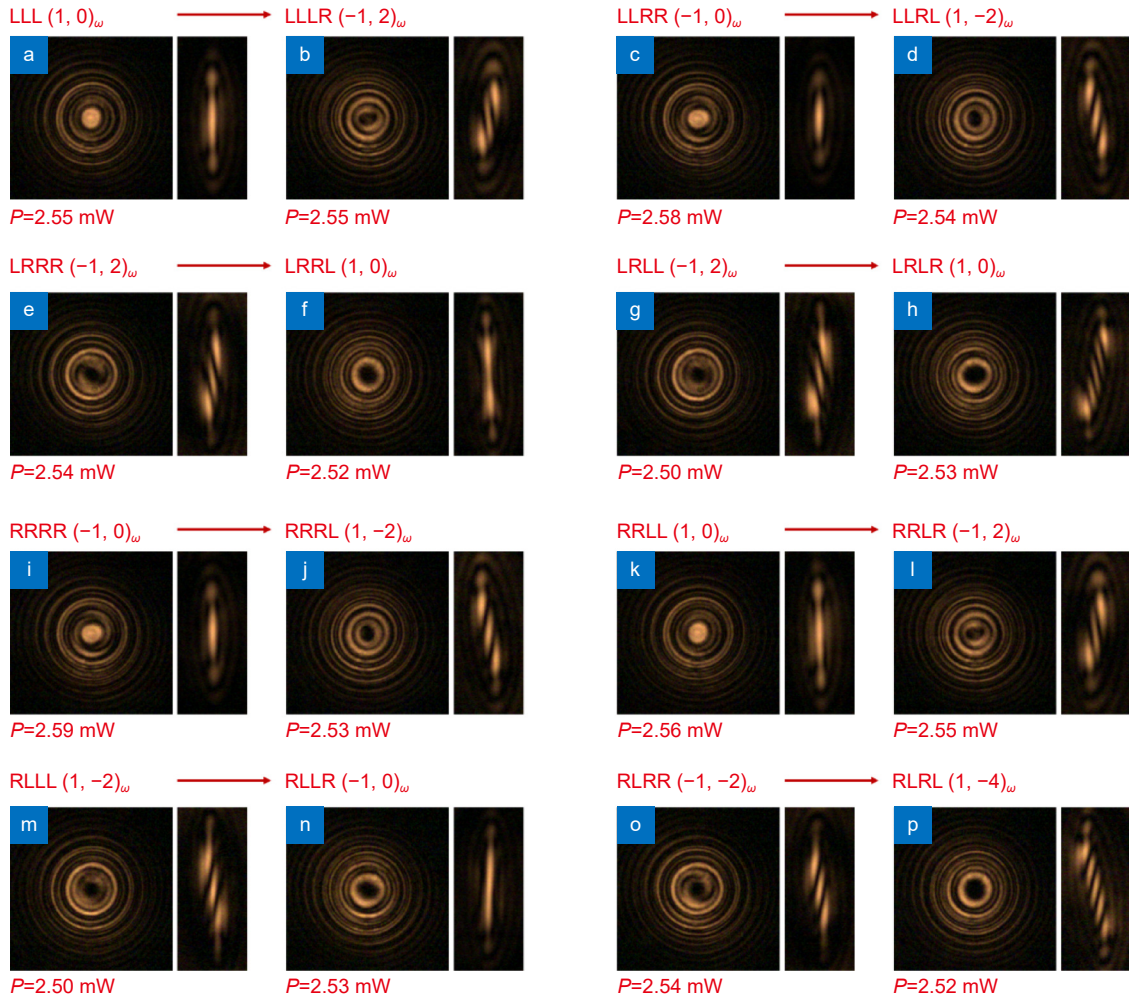


Fig. S5 | Evolution of the spin and orbital angular momentum states of the fundamental waves (FWs) after the second BBO crystal. The intensity profiles of the FWs under different combinations of the circularly polarized components are measured (square images). Intensity profiles imaged by cylindrical lens are used to identify the orbital angular momentum values (rectangle images). The power of the FWs is measured by using a power meter.

Section 5: Angular momentum states of the second harmonic waves in the double BBO crystal system

The captured intensity profiles of the SHWs behind the second BBO crystal are shown in Fig. S6. The first three characters and the fourth character in the polarization resolved measurements denote the spin states of the FWs and SHWs, respectively. The angular momentum states in the first (Fig. S6(a, e, i, m)) and third (Fig. S6(c, g, k, o)) columns are generated by the FWs through the SHG process in the second BBO crystal. In comparison, the states in the second (Fig. S6(b, f, j, n)) and fourth (Fig. S6(d, h, l, p)) columns are generated by those in the first and third columns through the linear optical SOI process at the second harmonic frequency.

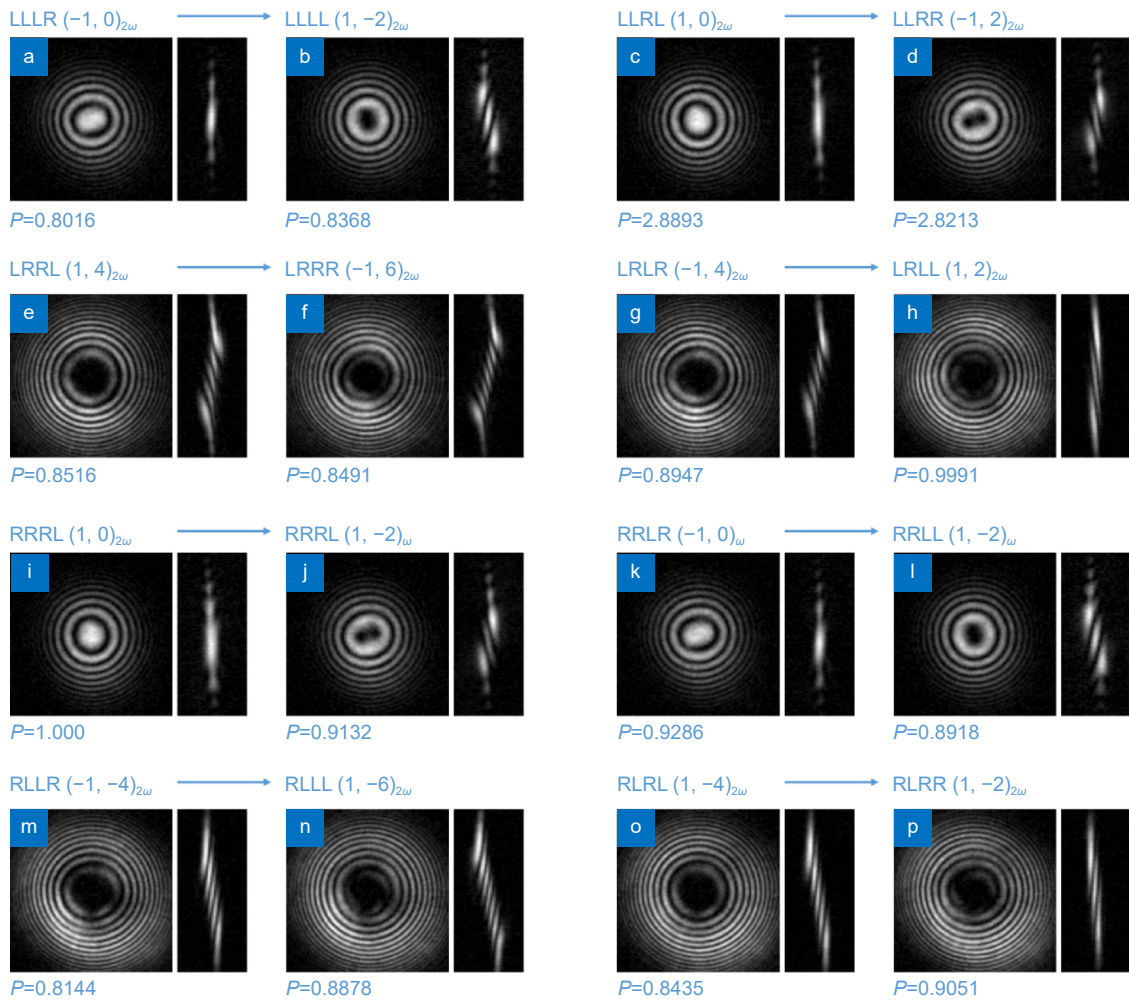


Fig. S6 | Spin and orbital angular momentum states of the second harmonic waves (SHWs) after the second BBO crystal. The intensity profiles of the SHWs under different combinations of the circularly polarized components are measured (square images). Intensity profiles imaged by a cylindrical lens are used to identify the orbital angular momentum values (rectangle images) of the SHWs. The relative power of the SHWs is obtained by integrating the spectra in Fig. S7, which are measured by using an Andor spectrometer (SR193i). The first three characters and the fourth character in the polarization resolved measurements denote the spin states of the fundamental waves and SHWs, respectively.

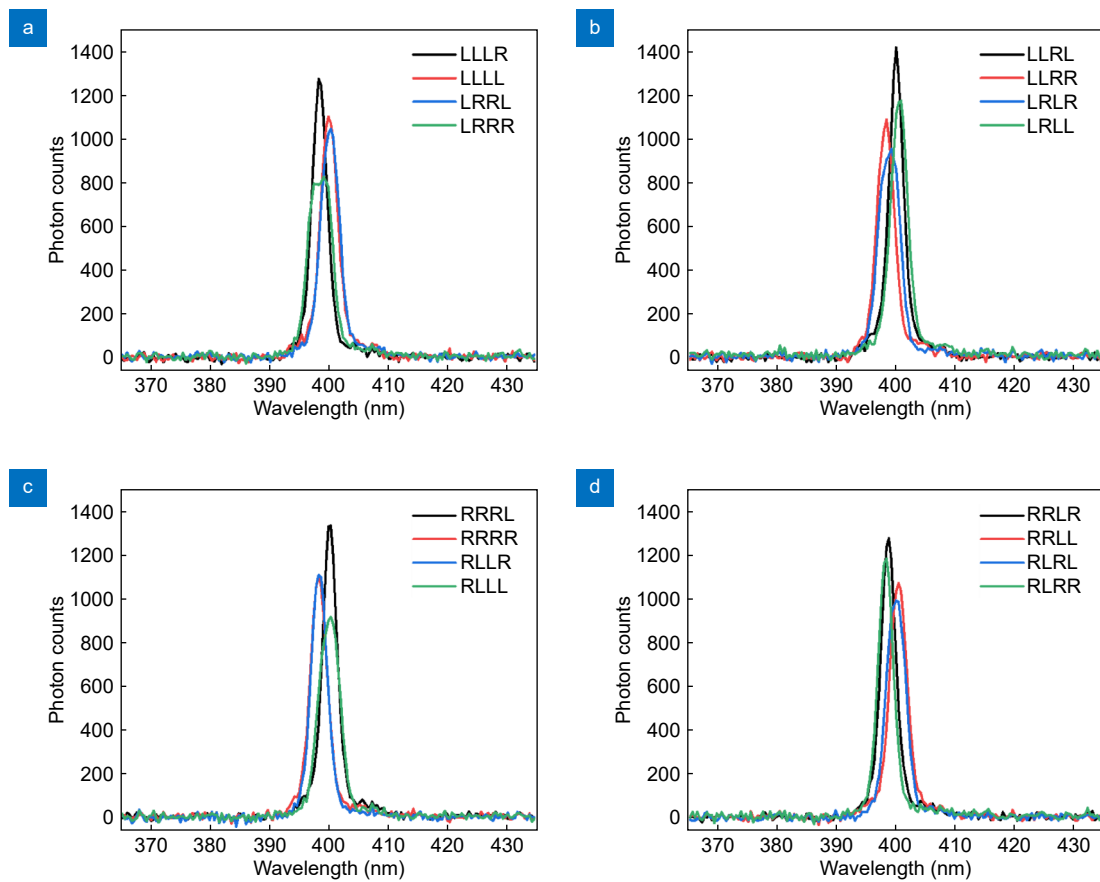


Fig. S7 | Spectra of the second harmonic waves (SHWs) after the second BBO crystal under different combinations of the circularly polarized components. The first three characters and the fourth character in the polarization resolved measurements denote the spin states of the FWs and SHWs, respectively. For all the measurements, the power of the fundamental wave is kept as 48 mW.