Supplementary Material for:
Nonlinear harmonic wave manipulation in nonlinear scattering medium via scattering-matrix method

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1 Principle of the measuring scattering matrix of nonlinear scattering medium

In the linear scattering media and monochromatic case, the output field of the scattering medium is related to the input field through an matrix (*K*), which can be written as:[21] $E_m^{\text{out}}(\omega)=\sum_n k_{mn}E_n^{\text{in}}(\omega)$, where $E_m^{\text{out}}(\omega)$ and $E_n^{\text{in}}(\omega)$ are the complex amplitudes of the *m*th output mode and *n*th input mode, respectively. k_{mn} is the matrix element and connects the input and output mode. Once the scattering process above involves a nonlinear process, the whole system will become more complicated. For a second-order nonlinear scattering medium, the nonlinear output field in the direction of backscattering and the input field can be connected through a scattering matrix (SM) K^{NL}, which is a 3-rank tensor and can be expressed as:

$$E_m^{\text{out}}(\omega_1+\omega_2)=\sum_n\sum_o k_{mno}^{\text{NL}}E_n^{\text{in}}(\omega_1)E_o^{\text{in}}(\omega_2), \tag{S1}$$

where $E_m^{\text{out}}(\omega_1+\omega_2)$, $E_n^{\text{in}}(\omega_1)$ and $E_o^{\text{in}}(\omega_2)$ are the complex amplitudes of the *m*th nonlinear output mode, *n*th and *o*th input modes with frequency $\omega_1+\omega_2$, ω_1 and ω_2, respectively. k_{mno}^{NL} is the SM

K^{NL} element and connects the linear input mode and nonlinear output mode. The SM K^{NL} contains the information of the generation and scattering process of nonlinear signals in the scattering medium. The above relationship is suitable for describing the second-order nonlinear processes that occur in the nonlinear scattering medium, such as second-harmonic generation (SHG), sum-frequency generation (SFG), and difference-frequency generation (DFG). In our experiment, we consider the SFG process and choose lithium niobate (LN) powder as the nonlinear scattering medium without loss of generality. The maximum size of the LN particle is much smaller than the coherent length l_c in a LN crystal, which means that every single LN particle can be seen as a nonlinear source[39] and each LN particle can be considered as a microcrystallite with random $\chi^{(2)}$ domain distribution. If we only modulate the input light field $E^{\mathrm{in}}(\omega_2)$ and keep the other input field $E^{\mathrm{in}}(\omega_1)$ fixed, the relationship between the output nonlinear signal and the input signals can be reduced to:

$$E_m^{\mathrm{out}}(\omega_1+\omega_2)=\sum_n k_{mn}^{\mathrm{NL}} E^{\mathrm{in}}(\omega_1) E_n^{\mathrm{in}}(\omega_2), \tag{S2}$$

We can further measure the SM of the nonlinear scattering medium using full-field interferometry method. For all input mode of $E^{\mathrm{in}}(\omega_2)$, if the relative phases are all shifted by a value α, the intensity of the mth output mode can be written as:

$$\begin{aligned} I_m^{\alpha}(\omega_1+\omega_2) &= \left|E_m^{\mathrm{out}}(\omega_1+\omega_2)\right|^2 \\ &= \left|s_m+\sum\nolimits_n e^{i\alpha} k_{mn}^{\mathrm{NL}} E^{\mathrm{in}}(\omega_1) E_n^{\mathrm{in}}(\omega_2)\right|^2 \\ &= \left|s_m\right|^2+\left|\sum\nolimits_n e^{i\alpha} k_{mn}^{\mathrm{NL}} E^{\mathrm{in}}(\omega_1) E_n^{\mathrm{in}}(\omega_2)\right|^2+2\,\mathrm{Re}\left[s_m^* \sum\nolimits_n e^{i\alpha} k_{mn}^{\mathrm{NL}} E^{\mathrm{in}}(\omega_1) E_n^{\mathrm{in}}(\omega_2)\right]. \end{aligned} \tag{S3}$$

Where s_m is the complex amplitude of the nonlinear output field used as reference in the mth output mode generated by input field $E^{\mathrm{in}}(\omega_1)$ and the unmodulated field of $E^{\mathrm{in}}(\omega_2)$ via SFG process. Thus, four phase steps for $\alpha = 0, \pi/2, \pi$ and $3\pi/2$ are used and we can get that:

$$\frac{I_m^0 - I_m^\pi}{4} + \frac{i\left(I_m^{3\pi/2} - I_m^{\pi/2}\right)}{4} = s_m^* \sum_n k_{mn}^{\mathrm{NL}} E^{\mathrm{in}}\left(\omega_1\right) E_n^{\mathrm{in}}\left(\omega_2\right) \tag{S4}$$

After completing all the 4N measurement, we can obtain $M \times N$ elements of the SM K^{NL} and nonlinear focal spot can be achieved via appropriate wavefront shaping of the input field $E^{\mathrm{in}}\left(\omega_2\right)$ according to optical phase conjugation.

2 Measurement of the transport mean free path of the LN powder

The fundamental parameter in the light diffusion model is the transport mean free path (TMFP), corresponding to the distance light travels in the medium before its direction is randomized. The TMFP of the scattering medium can be measured by coherent backscattering method.[40-42]

The measurement of TMFP l_t has been performed with the experimental setup shown in Fig. S1(a). A continuous wave (CW) laser with wavelength λ = 450 nm is used as the light source. A half-wave plate (HWP) after the laser is used to adjust the polarization of the laser beam. Then the laser beam is expanded by passing through a pair of expander lens L1 (f_1 = 50 mm) and L2 (f_2 = 200 mm). A modified Michelson interferometer was employed to detect the backscattered intensity. The CW laser is incident on the scattering medium, and the backscattered light is reflected by the BS and then passes through the lens L3 (f_3 = 50 mm), and is collected by the CCD. It should be noted that the scattering medium needs to be tilted slightly to reduce the interference of single scattered photons (ballistic photons) with the experimental results. Fig. S1(b) shows the intensity cross-sections of the back-scattering cone recorded by CCD, and Fig. S1(c) shows the Gaussian fit to the data of Fig. S1(b) with full width at half maximum of $\Delta\theta$ = 14.7 mrad. The TMFP can be calculated by:[42]

$$\Delta\theta = \frac{0.7\lambda}{2\pi l_t}, \tag{S5}$$

Where λ is the wavelength of the laser. The TMFP of the LN powder we used in the experiment is $l_t \approx 3.4\ \mu\text{m}$.

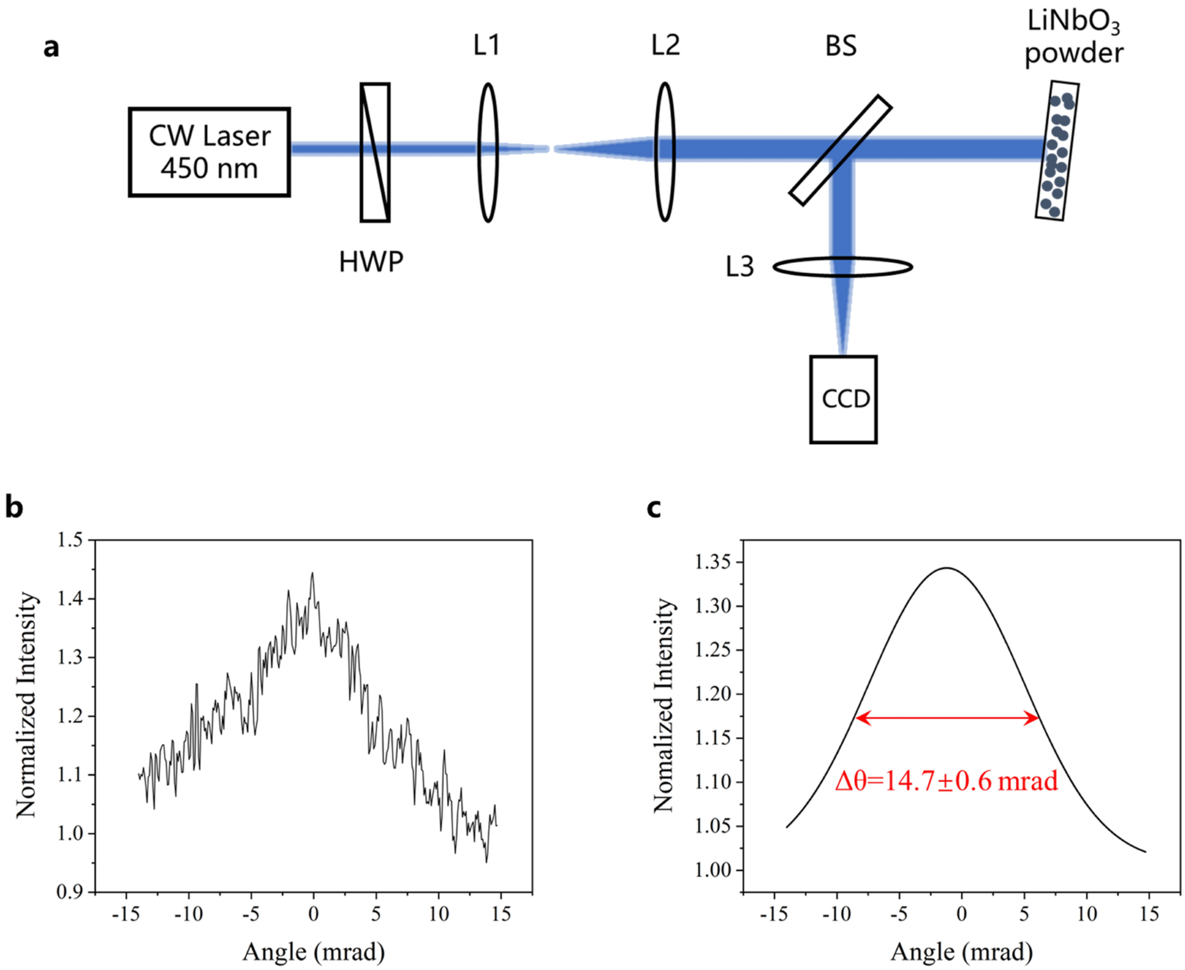

Fig. S1 (a) Experimental setup used for the measurement of TMFP. HWP, half wavelength plate; L1~3, lens, $f_{1\text{-}3}$ = 50, 200, and 50 mm; BS, beam splitter. (b) The intensity cross-sections of the back-scattering cone recorded by CCD. (c) The Gaussian fit to the data of (b).