Supplementary Materials for:

High spatial resolution Collinear Chiral Sum Frequency Generation Microscopy

Ziheng Ji[1,2], Wentao Yu[2,5], Dashan Dong[2,3], Hong Yang[2,3,4], Kaihui Liu[2], Yun-Feng Xiao[2,3,4], Qihuang Gong[2,3,4], Qinghai Song[1,3,*] and Kebin Shi[2,3,4,*]

[1]*Ministry of Industry and Information Technology Key Lab of Micro-Nano Optoelectronic Information System, School of Science, Harbin Institute of Technology (Shenzhen), Shenzhen 518055, China.*
[2]*State Key Laboratory for Mesoscopic Physics and Frontiers Science Center for Nano-optoelectronics, School of Physics, Peking University, Beijing 100871, China*
[3]*Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan, Shanxi 030006, China*
[4]*Peking University Yangtze Delta Institute of Optoelectronics, Nantong 226010, Jiangsu, China*
[5]*Institute of Interdisciplinary Physical Sciences, School of Science, Nanjing University of Science and Technology, Nanjing 210094, China*
**kebinshi@pku.edu.cn, qinghai.song@hit.edu.cn*

To explore the feasibility of utilizing collinear modality in vibrational resonant chiral SFG, we assume the use of a commercially available reflective objective (PIKE 40X 891-0007, NA=0.543~0.78) for both excitation and collection in simulation. This choice is based on its high efficiency and low chromatic aberration properties across UV to mid-IR wavelength range.

1. Foci and signal PSF

The advantages of using the collinear modality in vibrational resonant chiral SFG are particularly apparent. As shown in Fig. S1(b, c), the foci and signal PSF are well preserved. In contrast, the noncollinear modality, as depicted in Fig. S1(e, f), exhibits significant degradation. This degradation can be attributed to the weakened tailoring effect between the two foci when the IR focus becomes too large. Here, wavelength of 3274 nm is chosen to match the vibrational resonance of CH bond. The sample refractive index is set to 1.33.

Due to the presence of a secondary mirror structure within the reflective objective, the central portion of the incident beam becomes obstructed, as depicted in Figure S1(a, d). Consequently, the high spatial frequency component is given greater weighting, thereby offering higher resolving power in the transverse direction compared to the unobstructed case. Also, the hollow beam profile produced by the obstruction is advantageous for longitudinal polarization conversion efficiency. This is because such conversion can be viewed as a vectorial projection of the electric field, which is primarily contributed by the light in close proximity to the aperture's edge. Thus, even with a smaller numerical aperture and a lower refractive index medium, the longitudinal polarization conversion efficiency still reaches 25.5%, which is 53% of the value obtained using a 1.4 NA refractive objective lens.

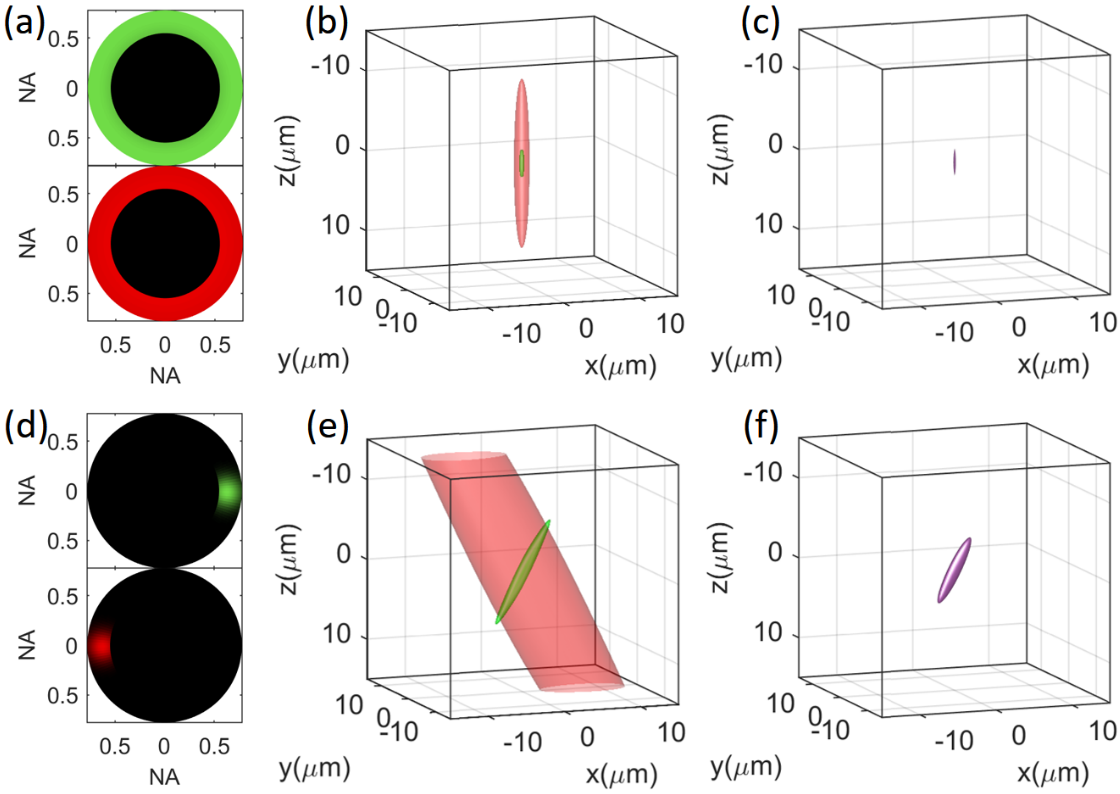

Figure S1. The numerical simulation of the foci and the chiral SFG point spread function. First row depicts collinear modality: (a) laser intensity at the objective lens pupil; (b) foci; (c) chiral SFG PSF. Second row depicts noncollinear modality: (d) laser intensity at the objective lens pupil; (e) foci; (f) chiral SFG PSF. The colors green, red, and purple are used to represent wavelengths of 520 nm, 3274 nm, and 448.7 nm, respectively.

2. Wavelength independent resolution

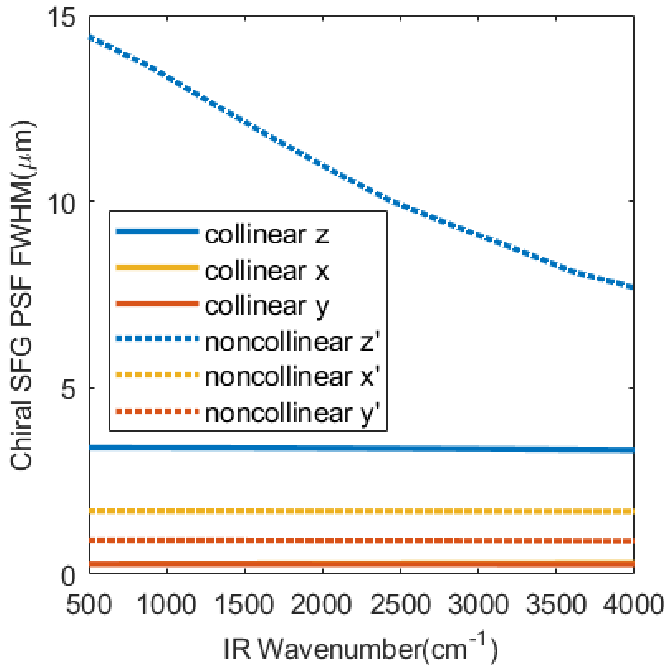

Figure S2. The dependence of the chiral SFG PSF FWHM on wavelength. The wavelength of mid-IR beam is tuned to scan across typical vibrational resonance range.

Figure S2 illustrates the size of chiral SFG PSF by tuning mid-IR laser beam across the typical spectrum range of 500 - 4000 cm^{-1} (2500 - 20000 nm), while keeping the visible laser beam fixed at 520 nm. The results show that the PSF of the collinear modality is insensitive to IR wavelength for all three dimensions. In contrast, for the noncollinear modality, the two transverse dimensions remain relatively constant at large values, but the longitudinal dimension increases substantially with increasing wavelength (or decreasing wavenumber).

3. Phase matching

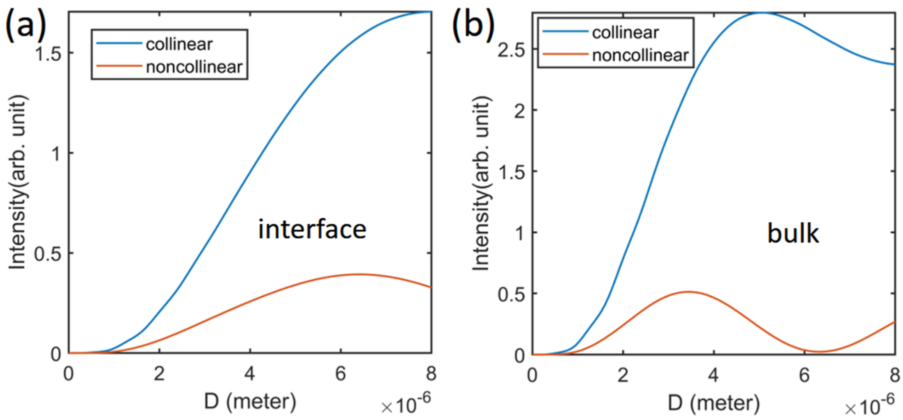

Figure S3. Dependence of far field chiral SFG signal on the sample size. (a) Focusing on the interface between a chiral liquid and an achiral medium. (b) Focusing inside the bulk of a uniform chiral liquid.