Supporting Information

Cross-polarized and Stable Second Harmonic Generation from Monocrystalline Copper

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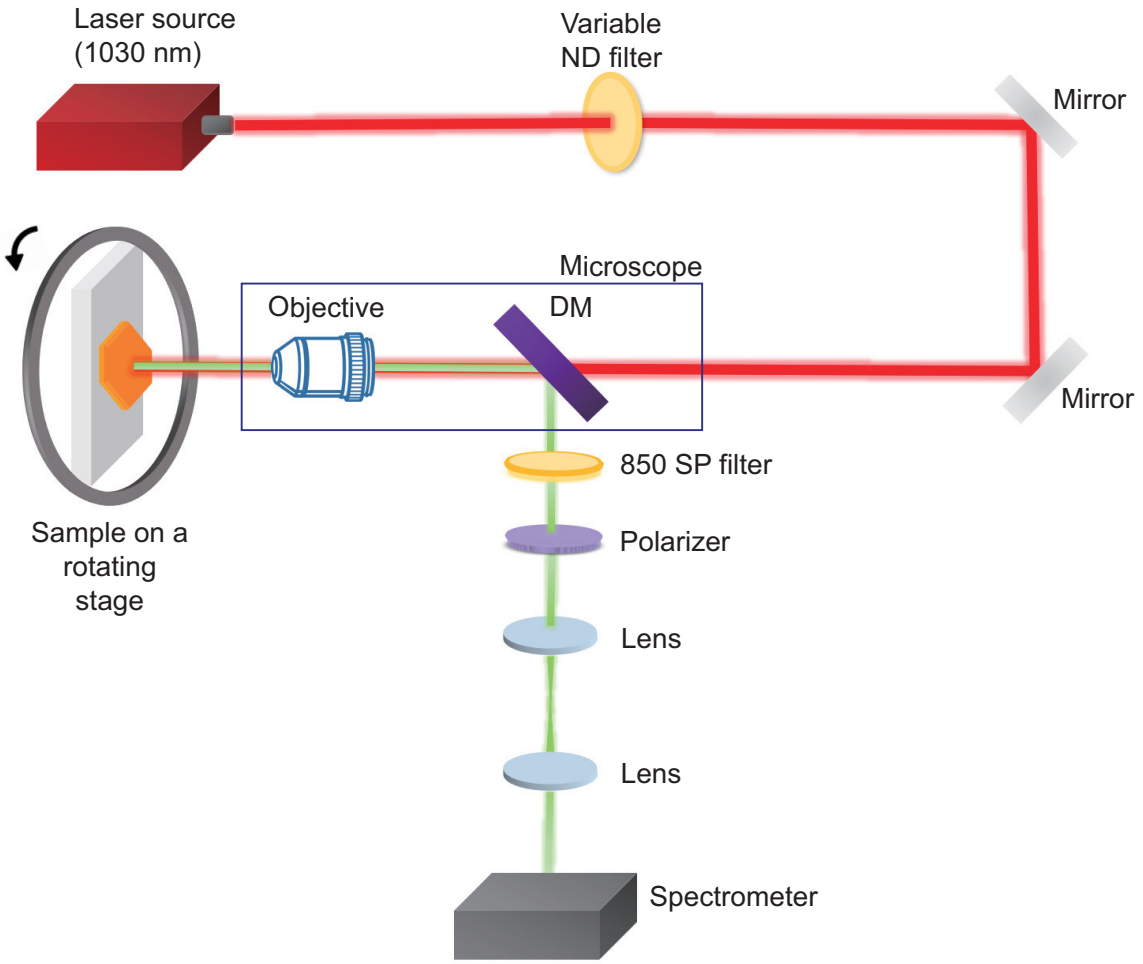

Figure S1: Schematic of the setup used for SHG measurements.The measurements were recorded using a microscope (Nikon Eclipse T2) coupled to a spectrometer (Princeton Instruments Spectra Pro HRS-500). A 1030nm femtosecond laser (NKT Origami) was used as excitation source and focused onto the sample by a 60x objective (Nikon S Plan Fluor ELWD, NA=0.7). An 750nm dichroic mirror (DM) was used to separate the fundamental and emitted signals. SP: Shortpass, ND: Neutral density. For the polarization-resolved measurements, a polarizer was inserted to the collection path.

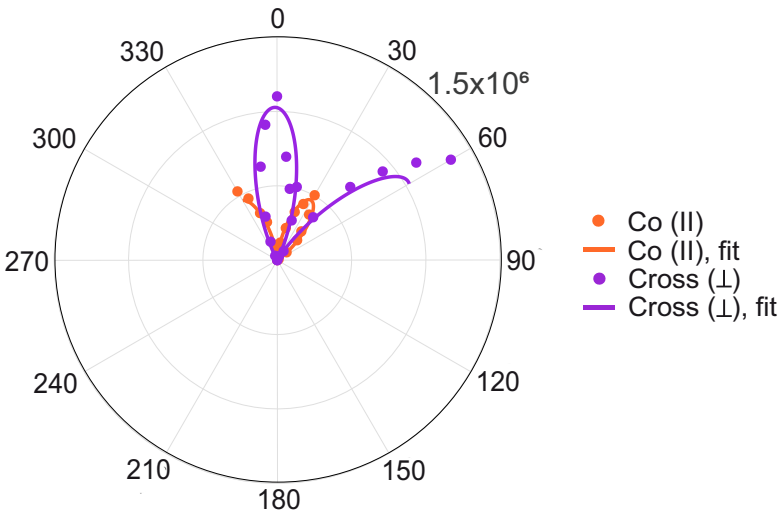

Figure S2: Polarization-resolved SHG measurements from a monocrystalline copper microflake in 0-90° range used for Fourier imaging measurements. Polar plot of SHG intensity as a function of detection polarization angle for co-polarized (orange) and cross-polarized (purple) emission, acquired at fixed incident polarization under 2 mW/μm^2 beam intensity.

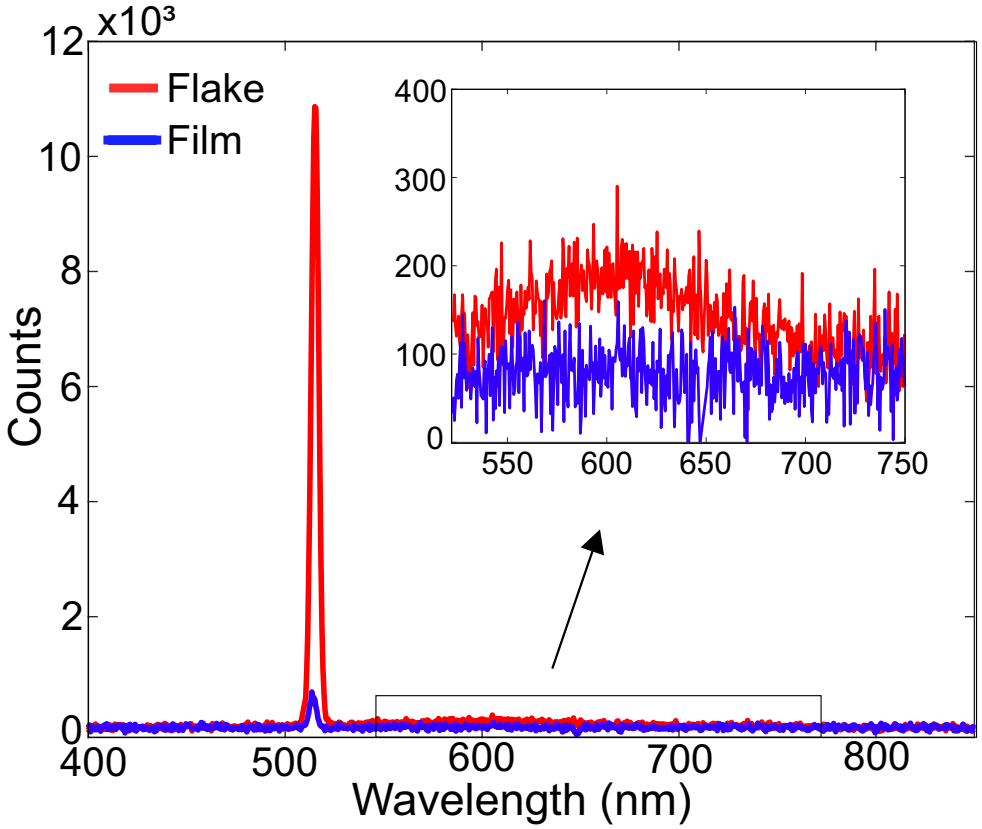

Figure S3: Relative SHG intensities for monocrystalline microflakes (red) and polycrystalline films (blue) when exciting at 1030 nm with 2.15 mW/μm^2 beam intensity. Inset shows expanded view of the two-photon photoluminescence response of the microflake in the 550-750 nm range. Small spot-to-spot and thickness dependent variation observed in the TPL strength wasn't thoroughly explored as it's not the main focus of this study.

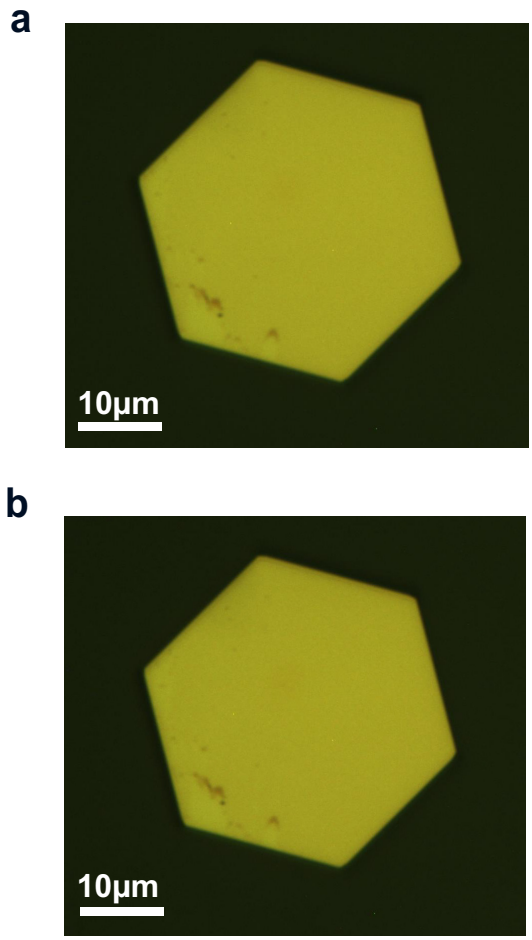

Figure S4: Bright-light images of the Cu microflake (a) before and (b) after 3 hours of continuous illumination under 2 mW/μm^2, showing no visible change to the flake surface under prolonged exposure.