Ultrafast All-Optical Wavefront Shaping Enabled by Second-Harmonic Polarization Switching in Transition Metal Dichalcogenide Monolayers

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Second-harmonic generation (SHG) plays a key role in various applications, ranging from frequency conversion to pulse characterization. However, further-reaching applications rely on modulation of the nonlinear signal which is usually achieved by external electrical or optical triggers, both having characteristic drawbacks in terms of switching speed or modulation depth. In this work, we initially introduce a novel all-optical method, leveraging the crystal symmetry of transition metal dichalcogenide (TMD) monolayers, realizing polarization and amplitude modulation of the generated SH signal with close to 100% depth and pulse duration-limited speed [1]. Furthermore, we demonstrate a cascaded meta-optical system based on the aforementioned process with a dielectric metasurface, enabling all-optical wavefront control with high spatial complexity (see Fig.1). We show that this combination enables beam deflection and orbital angular momentum (OAM) structuring of the SH signal on a femtosecond timescale [2], overcoming downsides from conventional wavefront shaping methods, such as digital micromirror devices which are limited to modulation rates in the kHz range. Therefore, our results pave the way for all-optical transformative applications in high-speed and robust OAM-encoded data transmission for secure, free-space communication. They also highlight the capabilities of pristine two-dimensional materials, as well as their combination with carefully designed dielectric metasurfaces for ultrafast light modulation and structuring.

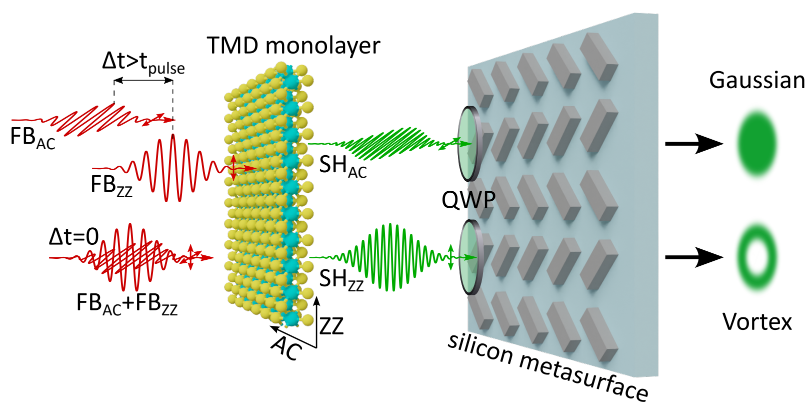

Fig. 1. Initially, two orthogonally linear polarized fundamental beam (FB) replicas, with pulse durations t_{pulse}, are aligned along the main crystal axes (armchair (AC) and zig-zag (ZZ) directions, respectively) of the TMD monolayer. The emitted SH signal, governed by the TMD's nonlinear susceptibility tensor, varies with the time-delay (Δt) between the pulse replicas, emitting along the AC ($\Delta t > t_{pulse}$) or ZZ ($\Delta t = 0$) direction. Subsequently, a quarter-wave plate (QWP) leads to a left (right) handed circular polarization of the SH for emission along the AC (ZZ) direction. Finally, a silicon metasurface modulates the SH wavefront based on the helicity of the incident radiation, resulting in either a Gaussian or a vortex beam shape.

References

[1] S. Klimmer *et al.*, Nat. Photonics **15**, 837-842 (2021).

[2] A. Sinelnik *et al.*, Nat. Commun. **15**, 2507 (2024).