

Cross-frequency nonlinear metasurfaces for simultaneous near- and far-field enhancement and highly sensitive molecular detection

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Backgrounds

- **Weak Nonlinear signals:** Inherently weak due to the **near-field confinement** and inefficient **far-field collection**.
- **Metasurfaces:** Offer unprecedented capabilities in tailoring both linear and nonlinear responses at the **subwavelength scale**.
- **The Research Gap:** Mostly focus on single-frequency optimization. Simultaneously engineering pump coupling and harmonic emission remains underexplored.

Design of nonlinear metasurface

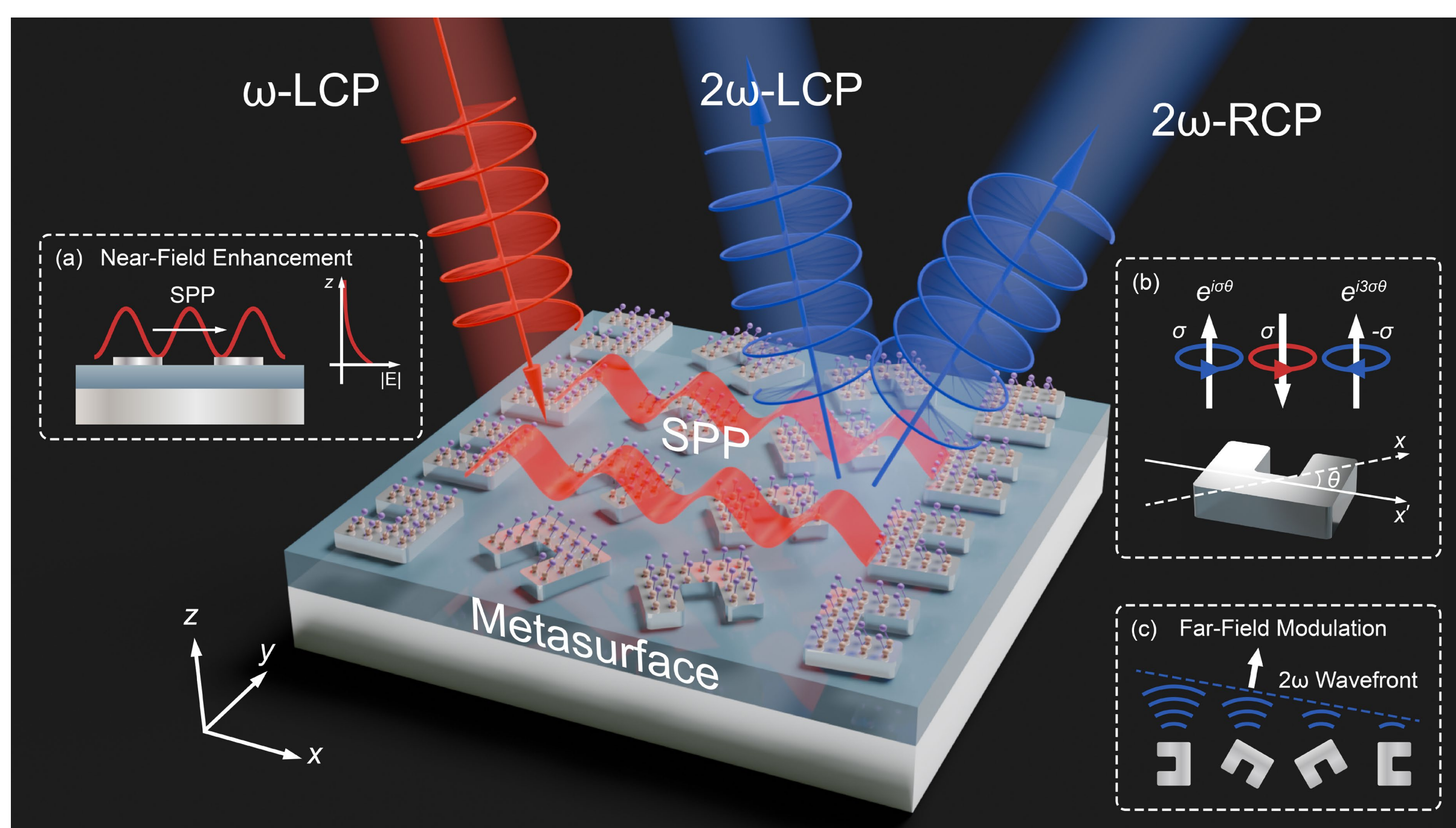


Figure 1 Concept of the nonlinear metasurface.

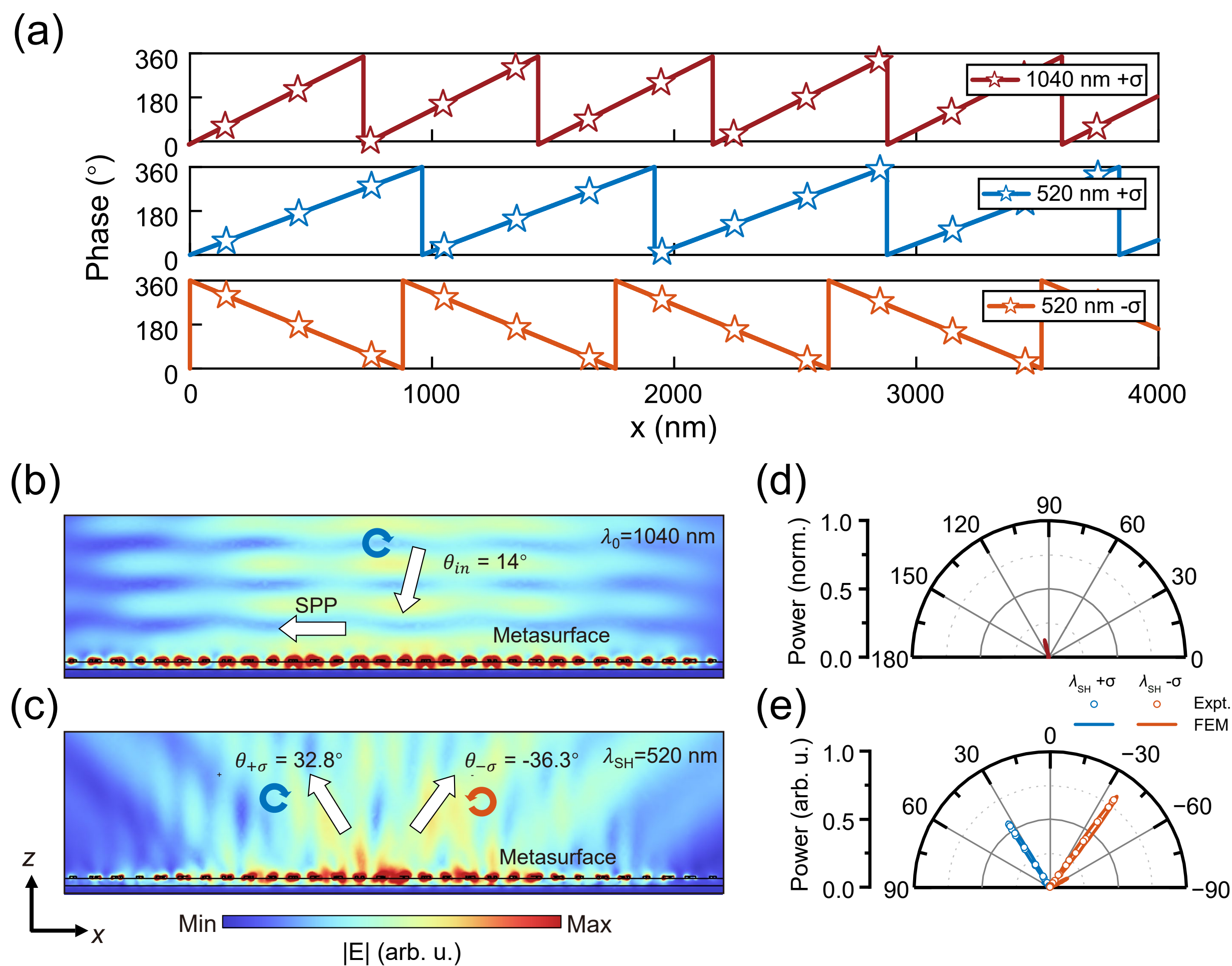


Figure 2 (a) Schematic of the phase distribution of the metasurface at 1040nm (FW) and 520nm (SH). (b)-(c) FEM-simulated $|E|$ field intensity of the metasurface at λ_0 (b) and λ_{SH} (c). (d)-(e) FEM-simulated angle spectrum of the far-field intensity at λ_0 (d) and λ_{SH} (e). The open circles in (e) denote experimental data.

- At λ_0 : Phase gradient \rightarrow Efficient SPP excitation^[1] \rightarrow **Near-field (NF) enhancement**
- At λ_{SH} : Nonlinear geometric phase^[2] \rightarrow Wavefront reshaping \rightarrow **Far-field (FF) enhancement**

Contributions of NF and FF enhancement

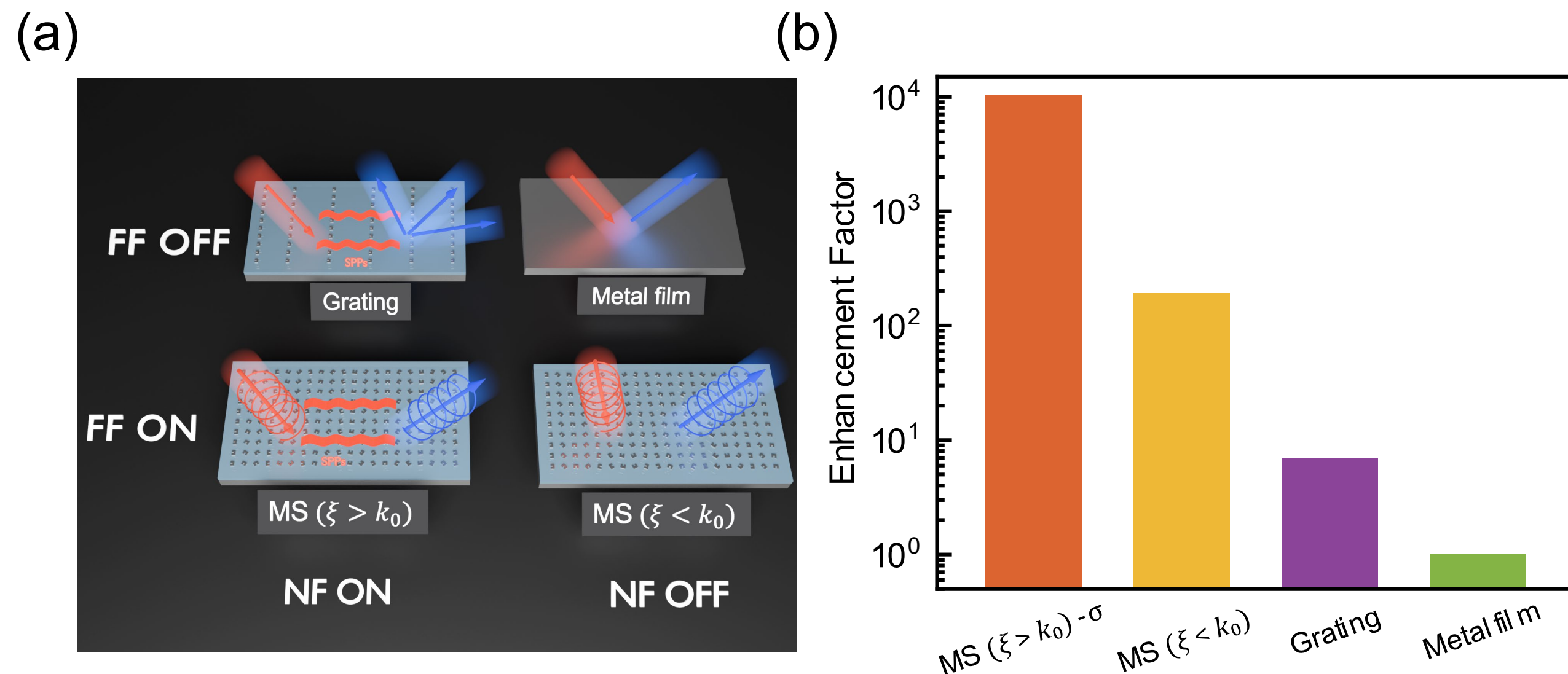


Figure 3 Disentangling the contributions of near-field enhancement at fundamental wave and far-field shaping at second harmonic wave. (a) Schematic of the investigated samples. (b) Second-harmonic enhancement factors for different control samples.

- A general strategy for boosting nonlinear conversion efficiency with a SHG enhancement factor $> 10^4$.

Highly sensitive molecular detection

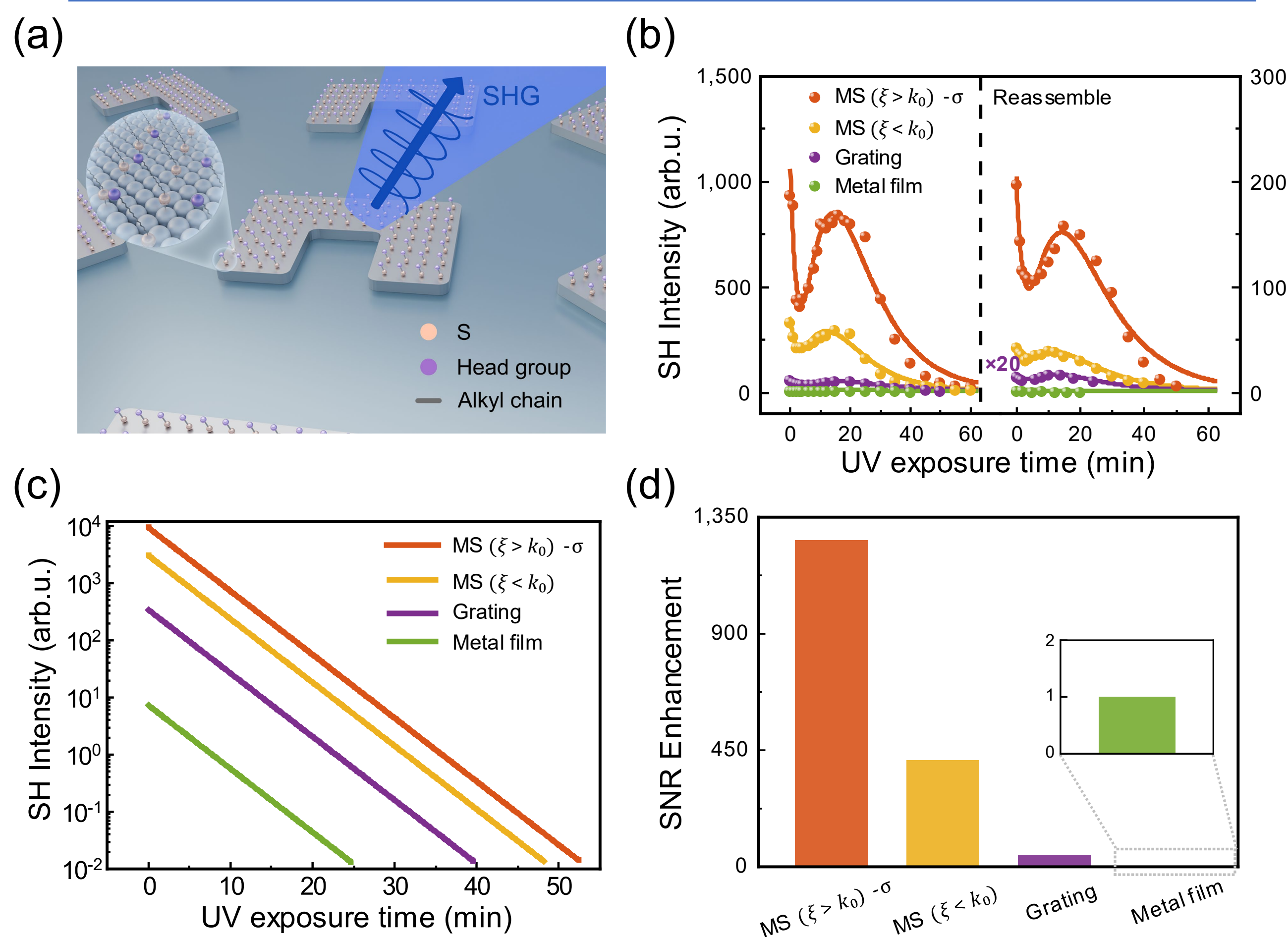


Figure 4 (a) Schematic illustration of *n*-hexadecanethiol self-assembled monolayer (SAMs) formation on nonlinear metasurface. (b) Total SHG signals for different control groups under varying UV exposure durations. (c) Extracted molecular contributions to the SHG signal across different control groups. (d) Normalized SNR enhancement for the detection of SAMs.

- The SAMs undergo oxidative desorption under UV illumination^[3]. The **molecular origin** of signal variations was confirmed via the dynamic and repeatable detections.
- **Isolated molecular contributions** by fitting

$$I(t) = \left| \chi_{thiol}^{(2)}(0)e^{-\frac{t}{\tau_1}} + \chi_{metal}^{(2)}(0)e^{-\frac{t}{\tau_2}}e^{i\varphi} \right|^2$$
- The normalized signal-to-noise ratio (SNR) enhancement $> 10^3$, corresponding to a detection sensitivity of ~ 0.01 monolayers/s.

Conclusions

- ✓ A **general strategy** for boosting nonlinear signals is established using cross-frequency nonlinear metasurfaces, achieving an SHG enhancement $> 10^4$ through both near- and far-field contributions.
- ✓ The platform enables **high-sensitivity molecular detection**, yielding an SNR enhancement $> 10^3$ and a detection sensitivity of ~ 0.01 monolayers/s.