SAND2018-9605C

Ultrathin nonlinear metasurfaces for 'flat nonlinear optics'

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“Flat” nonlinear optics

Phase-matching

Bulk: $\vec{k}_1 + \vec{k}_2 = \vec{k}_3$

– requires special efforts

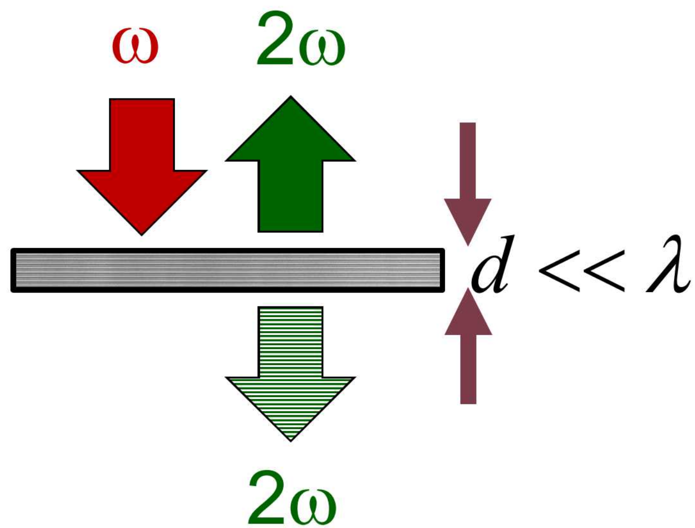
Surface: $k_{1//} + k_{2//} = k_{3//}$

– satisfied automatically

Generation efficiency:

$$I_{\omega 3} = \frac{\omega_3^2}{2\varepsilon_0 n_{\omega 1} n_{\omega 2} n_{\omega 3} c^3} \left|\chi_{eff}^{(2)}\right|^2 I_{\omega 1} I_{\omega 2} d^2$$

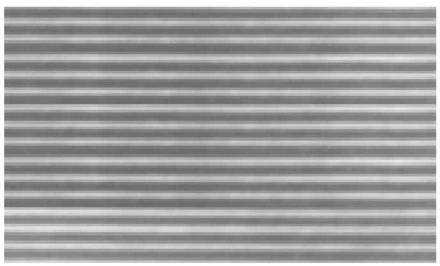
$$\boxed{\eta_{SHG} \equiv \frac{I_{2\omega}}{I_\omega} \approx \left(\frac{d}{\lambda_{2\omega}}\right)^2 \left|\chi_{eff}^{(2)} E_\omega\right|^2}$$

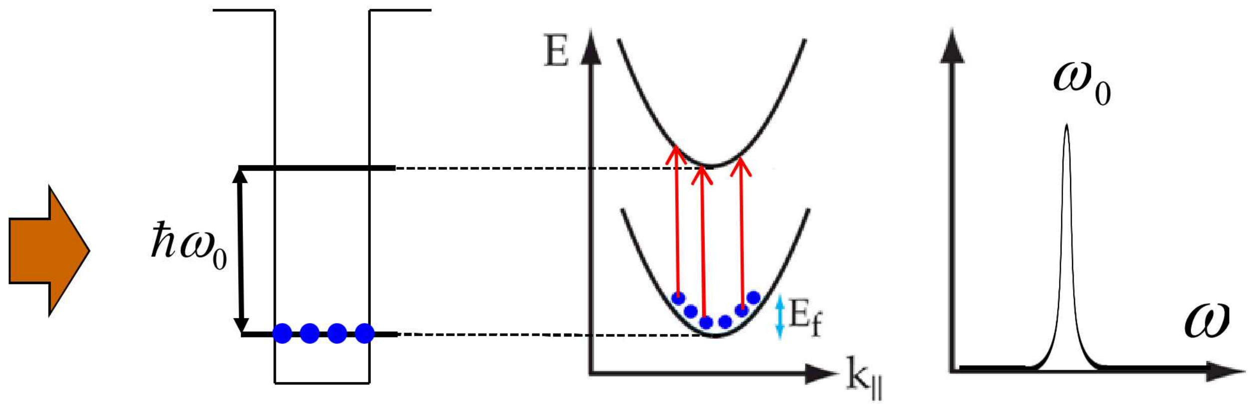


Need $\left|\chi_{eff}^{(2)} E_\omega\right| \sim 1$

Intersubband transitions

n-doped quantum wells





Key features:

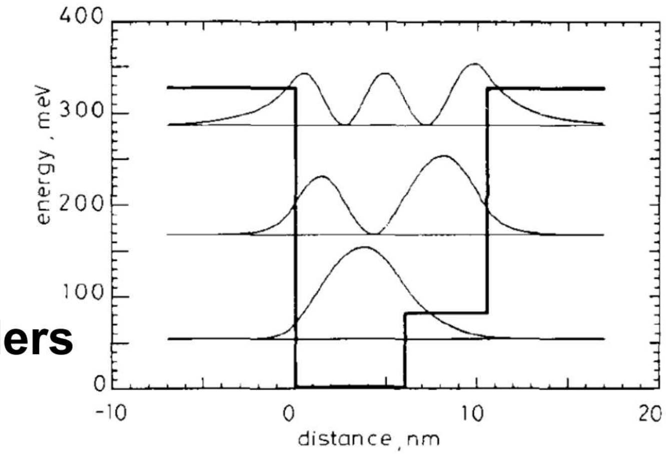
- *Control of energy levels positions, transition dipole moments, lifetime*
- *Tailored linear and nonlinear optical properties for TM-polarized light*
- *Ultra-fast electron dynamics (0.3-3 ps)*
- *Various materials AlInAs/InGaAs/InP, GaAs/AlGaAs, GaN/AlInGaN…*
- *Foundation of quantum cascade lasers*

Intersubband nonlinearities

Giant nonlinearities for wave mixing

Mid-IR SHG $\chi^{(2)}$ 10^4-10^5 pm/V

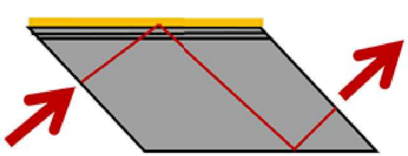
(E. Rosencher et al. 1989; M. Fejer et al. 1989)

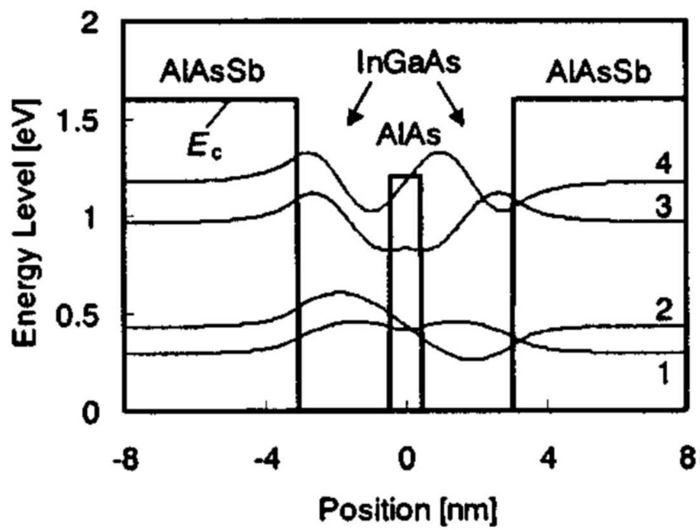


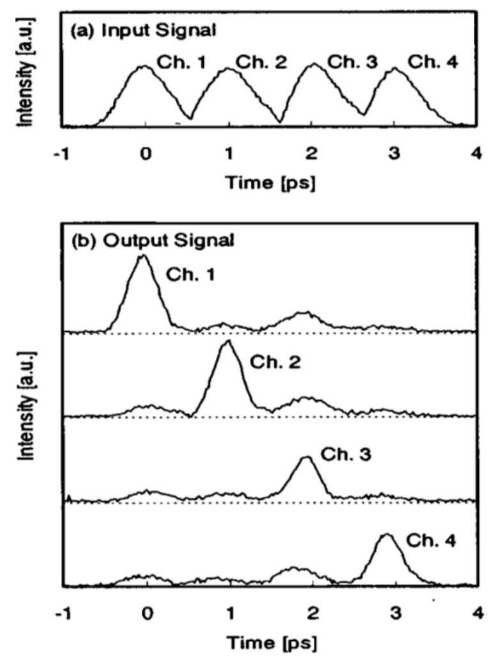
+ Other nonlinear processes of 2nd and higher orders

Ultrafast optical switching

Akiyama et al, *IEEE Photon. Technol. Lett.* **14,** 495 (2002)







Nonlinear polaritonic metasurfaces

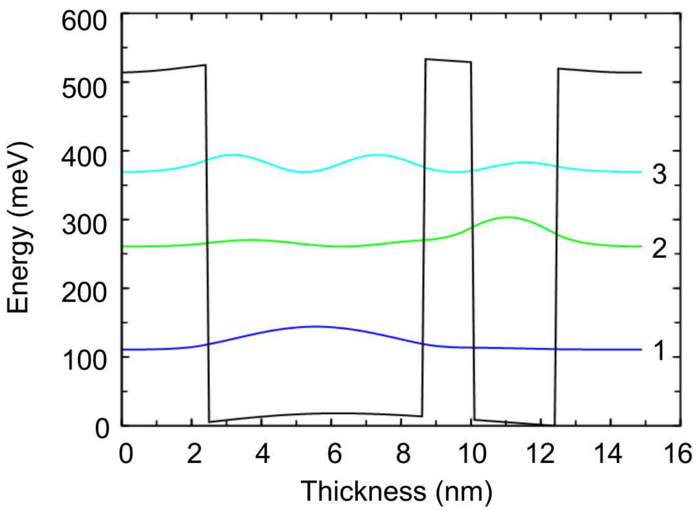
Quantum-engineered intersubband transitions

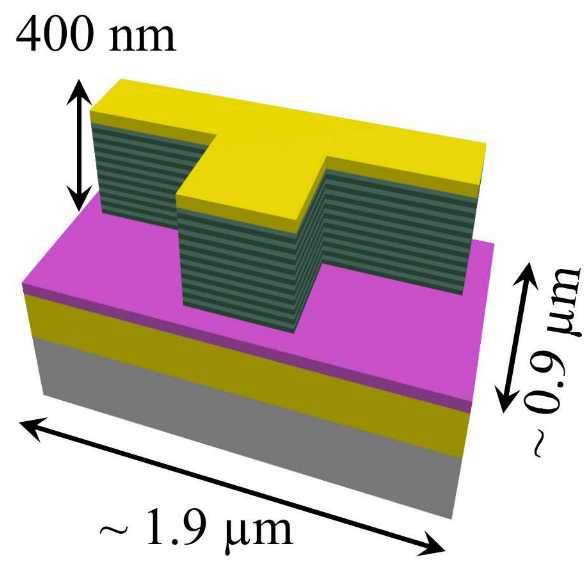
+

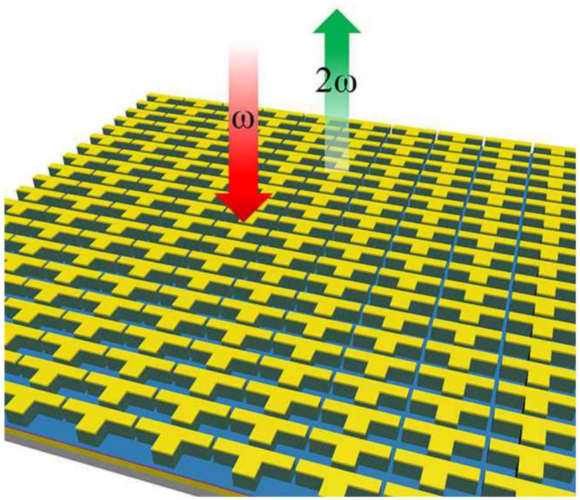
Electromagnetically-engineered modes in metallic nanostructures

➡ Large-area ultrathin metasurfaces with tailored nonlinear response

Lee *et al*., *Nature*, **511**, 65-69 (2014)

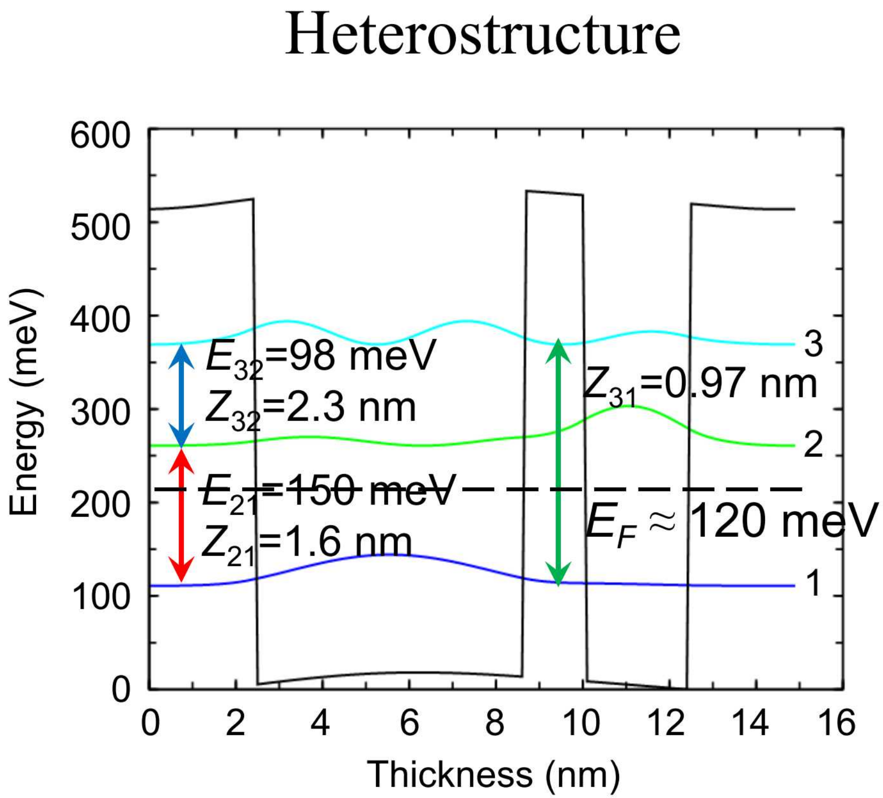


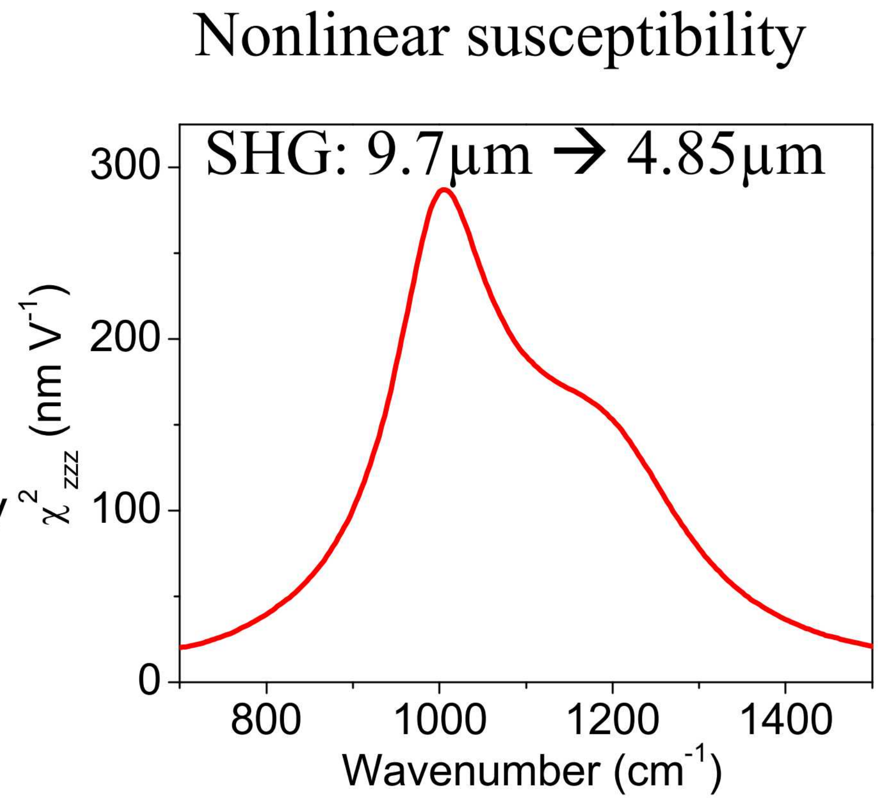




SHG mirror: 8-10μm → 4-5μm (LWIR→MWIR)

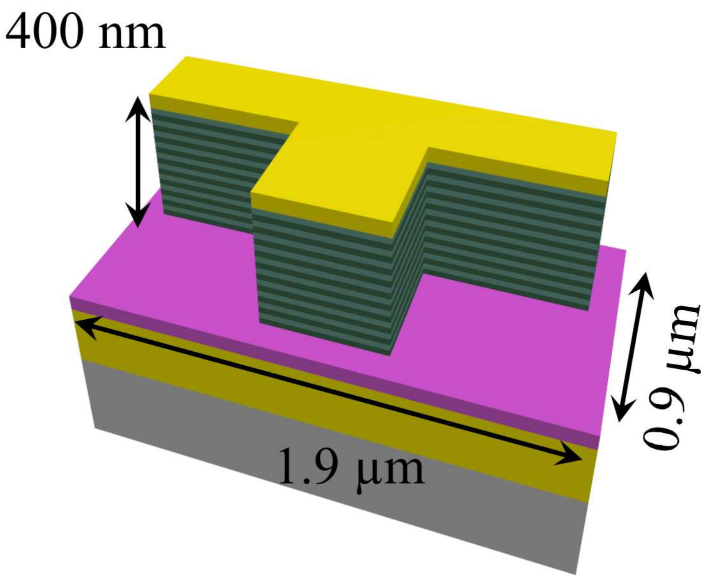
Semiconductor heterostructure design

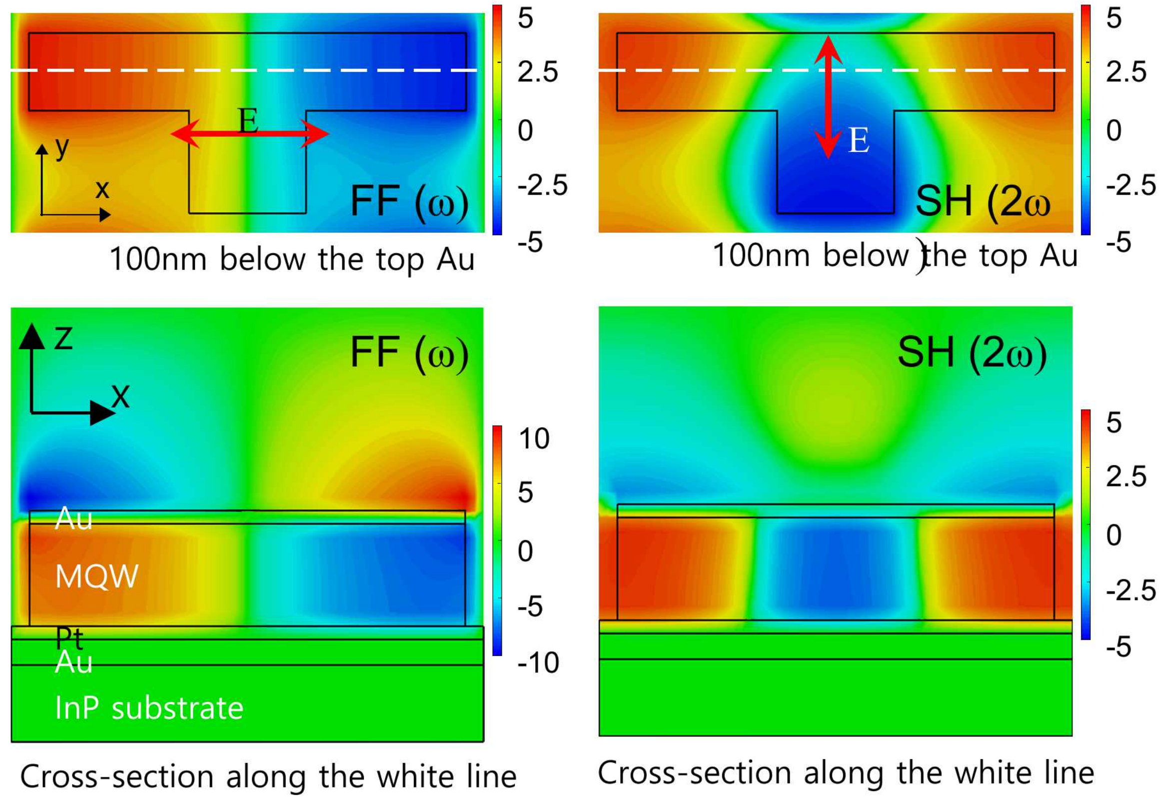




$$\chi_{zzz}^{(2)} \approx \left(N_1 - 2N_2 + N_3\right)\frac{e^3}{\hbar^2\varepsilon_0}\frac{z_{12}z_{13}z_{23}}{\left(\omega-\omega_{12}-i\gamma_{12}\right)\left(2\omega-\omega_{13}-i\gamma_{13}\right)}$$

Nanoresonator design





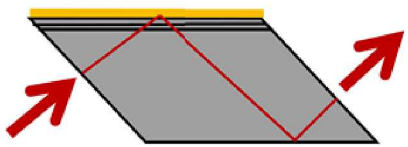
$$I_{2\omega} = \frac{2\omega^2}{\varepsilon_0 n_\omega^2 n_{2\omega} c^3} \left|\chi_{eff}^{(2)}\right|^2 I_\omega I_\omega d^2$$

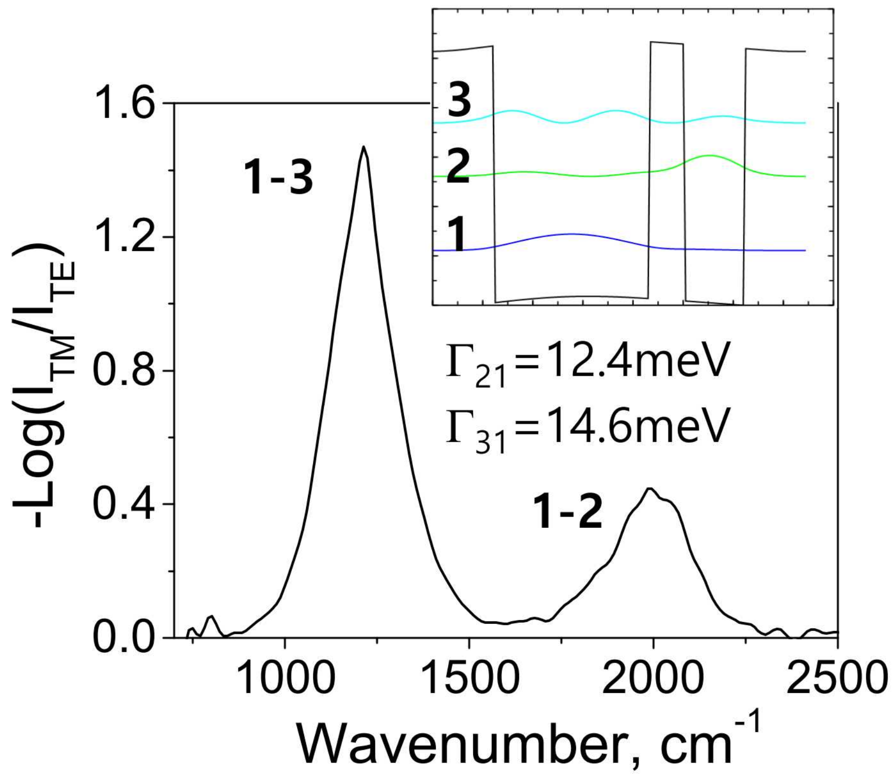
$$\chi_{ijk}^{(2)eff} = \chi_{zzz}^{(2)} \cdot \boxed{\frac{\displaystyle\int_{unit\_cell} \frac{E_{z(i)}^{2\omega}}{E_{i(inc)}^{2\omega}} \cdot \frac{E_{z(j)}^{\omega}}{E_{j(inc)}^{\omega}} \cdot \frac{E_{z(k)}^{\omega}}{E_{k(inc)}^{\omega}}\, dV}{V_{unit\_cell}}}, \quad (i, j, k = x, y, z)$$

Overlap integral, ~4

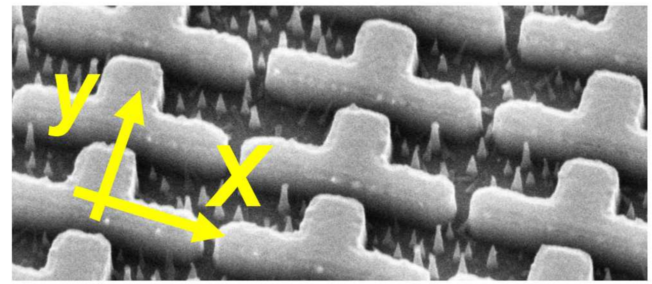
Characterization

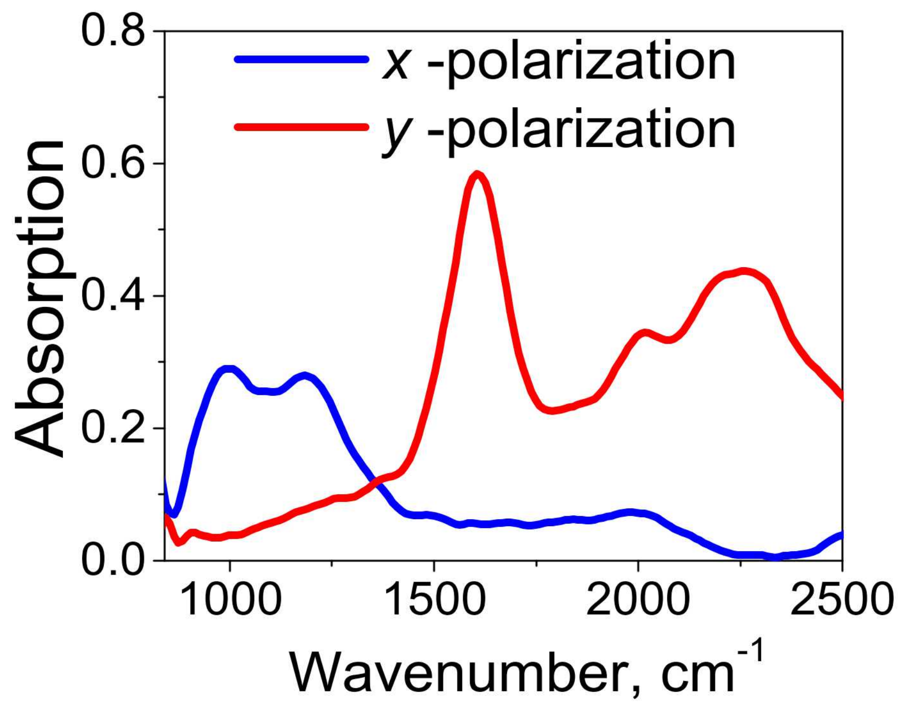
Heterostructure



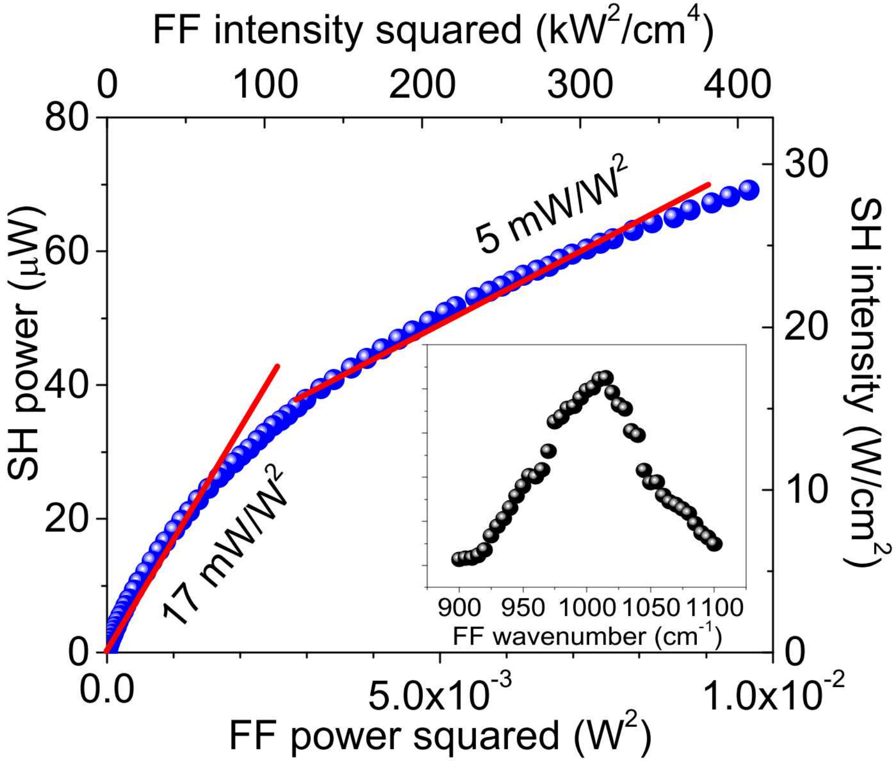


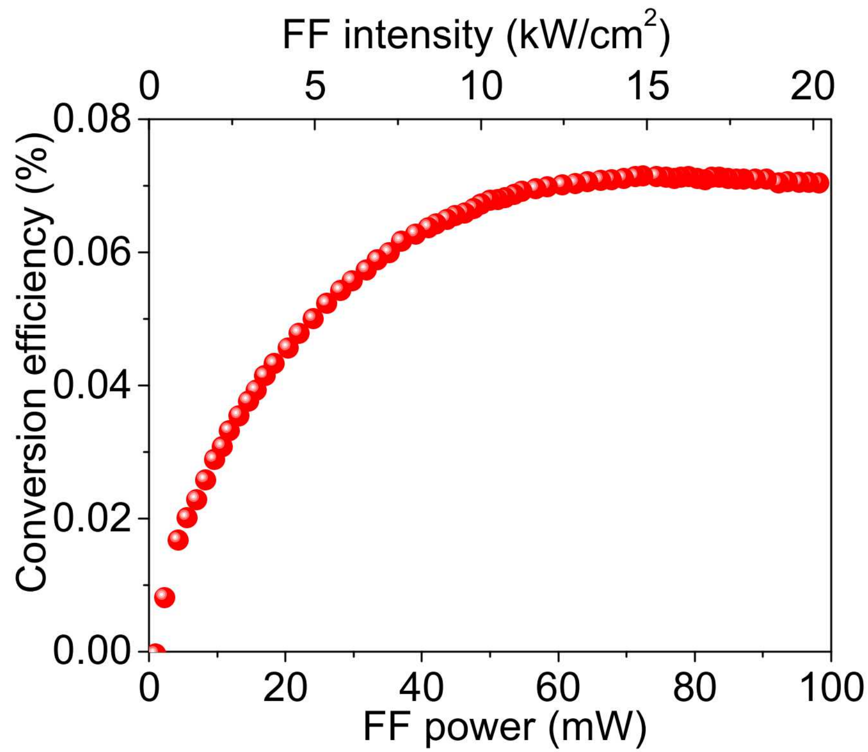
Metasurface





Experiment



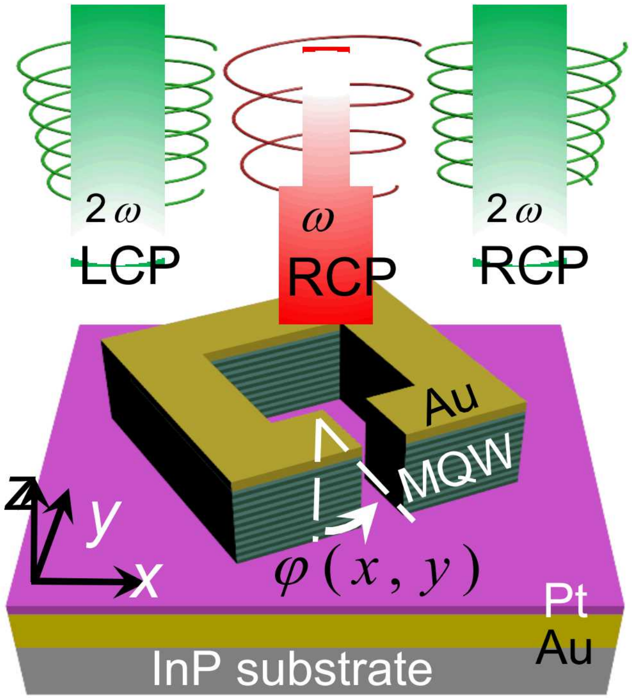


$$\chi^{(2)eff}_{yxx} \approx 1.2\times10^{6}\ pm/V$$

$(|\chi^{(2)}E_{\omega}| \sim 1 \text{ @ } I_{\omega} \sim 100\ kW/cm^{2})$

Lee *et al.*, *Adv. Opt. Mat.* **4,** 664 (2016)

Gradient nonlinear metasurfaces

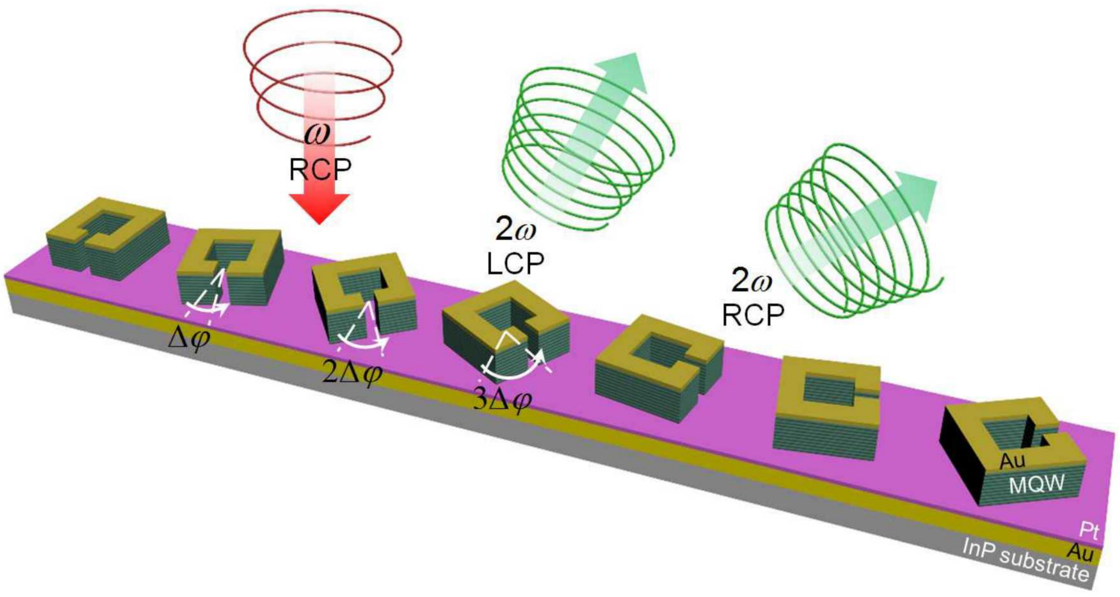


$$\chi^{(2)}_{L'L'L'} = \chi^{(2)}_{LLL} \exp(-i3\varphi)$$

$$\chi^{(2)}_{R'L'L'} = \chi^{(2)}_{RLL} \exp(-i\varphi)$$

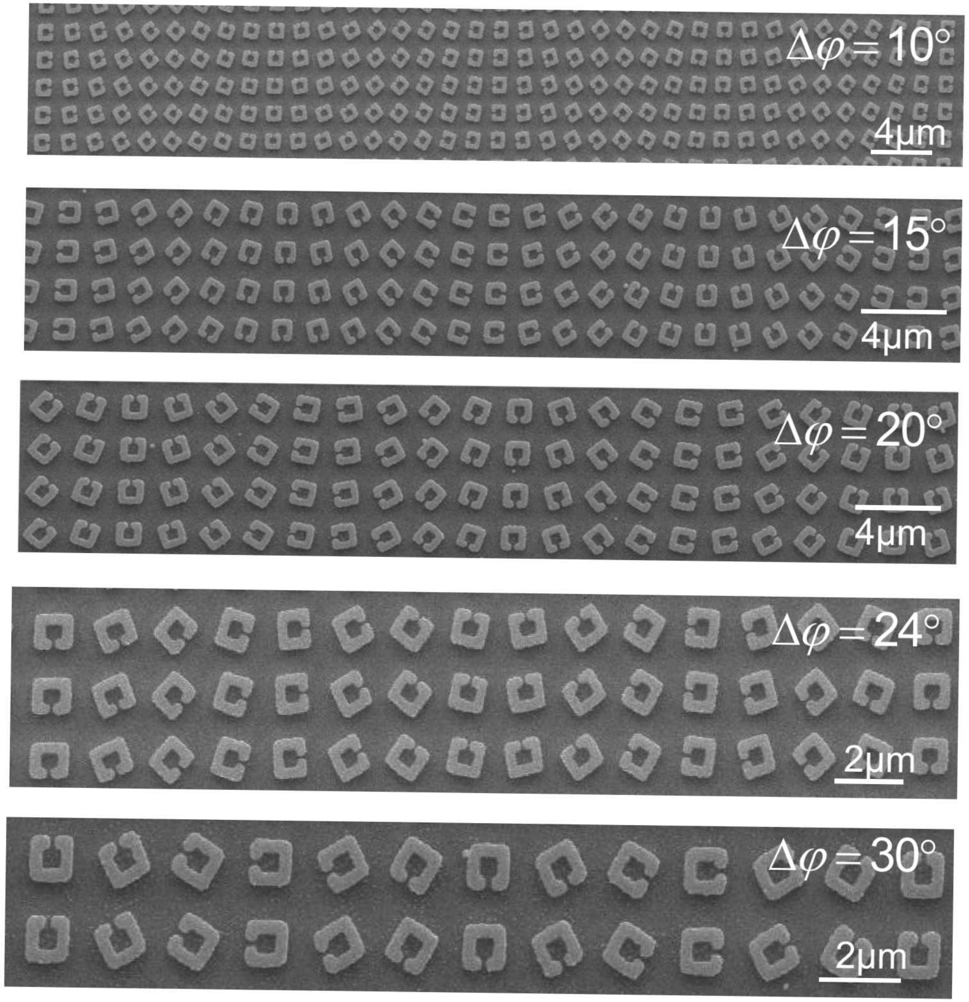
$$\chi^{(2)}_{R'R'R'} = \chi^{(2)}_{RRR} \exp(i3\varphi)$$

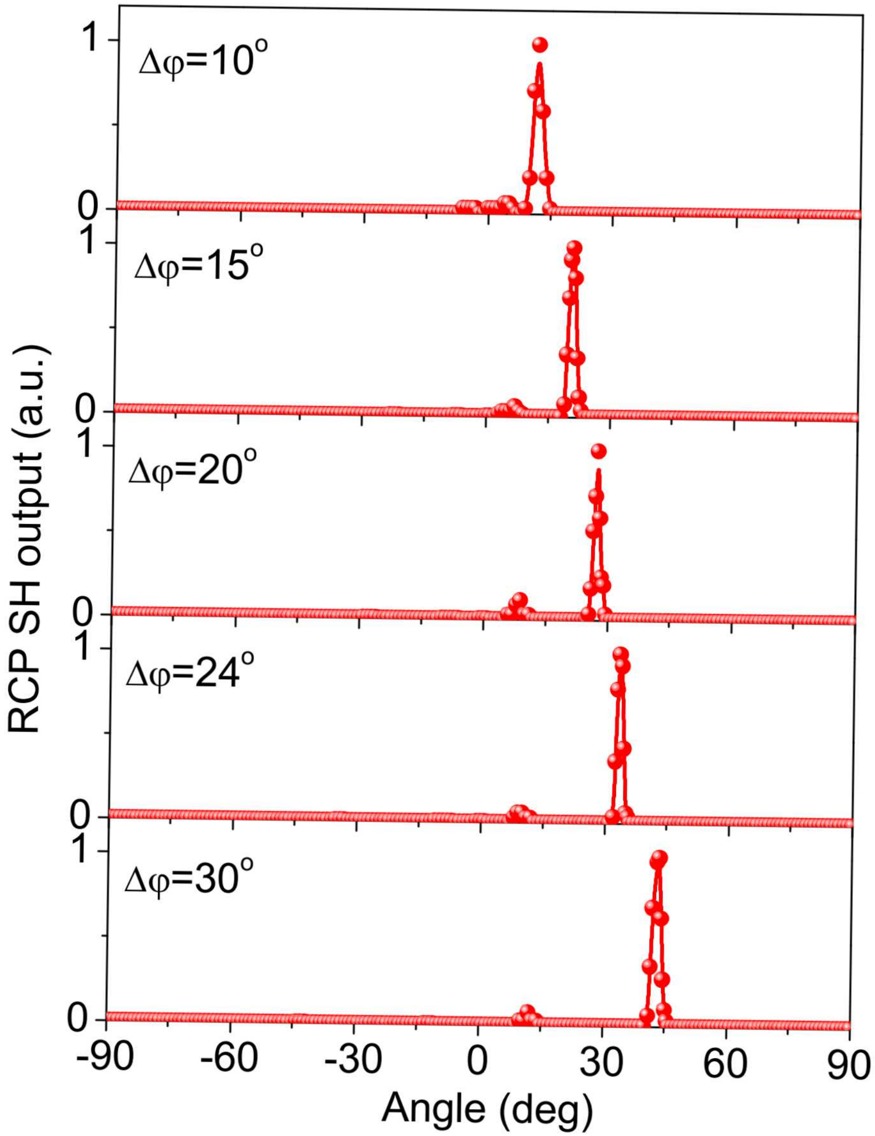
$$\chi^{(2)}_{L'R'R'} = \chi^{(2)}_{LRR} \exp(i\varphi)$$



Tymchenko *et al.*, *Phys. Rev. Lett.* **115,** 207403 (2015)

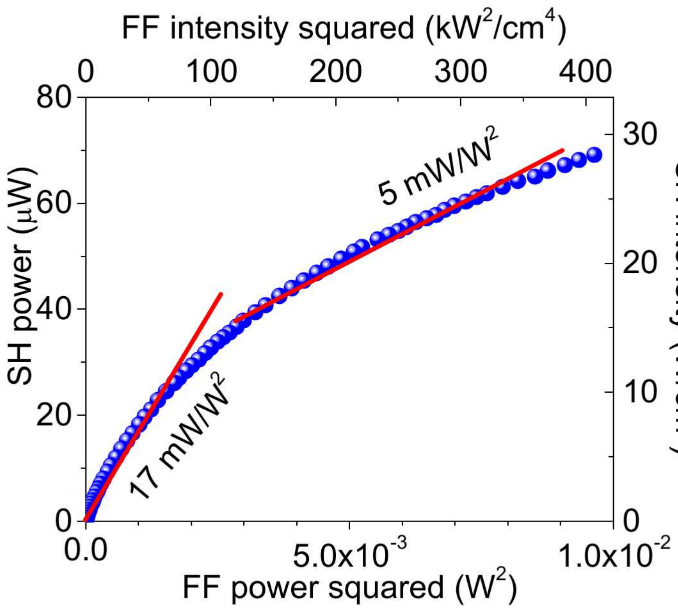
Experment

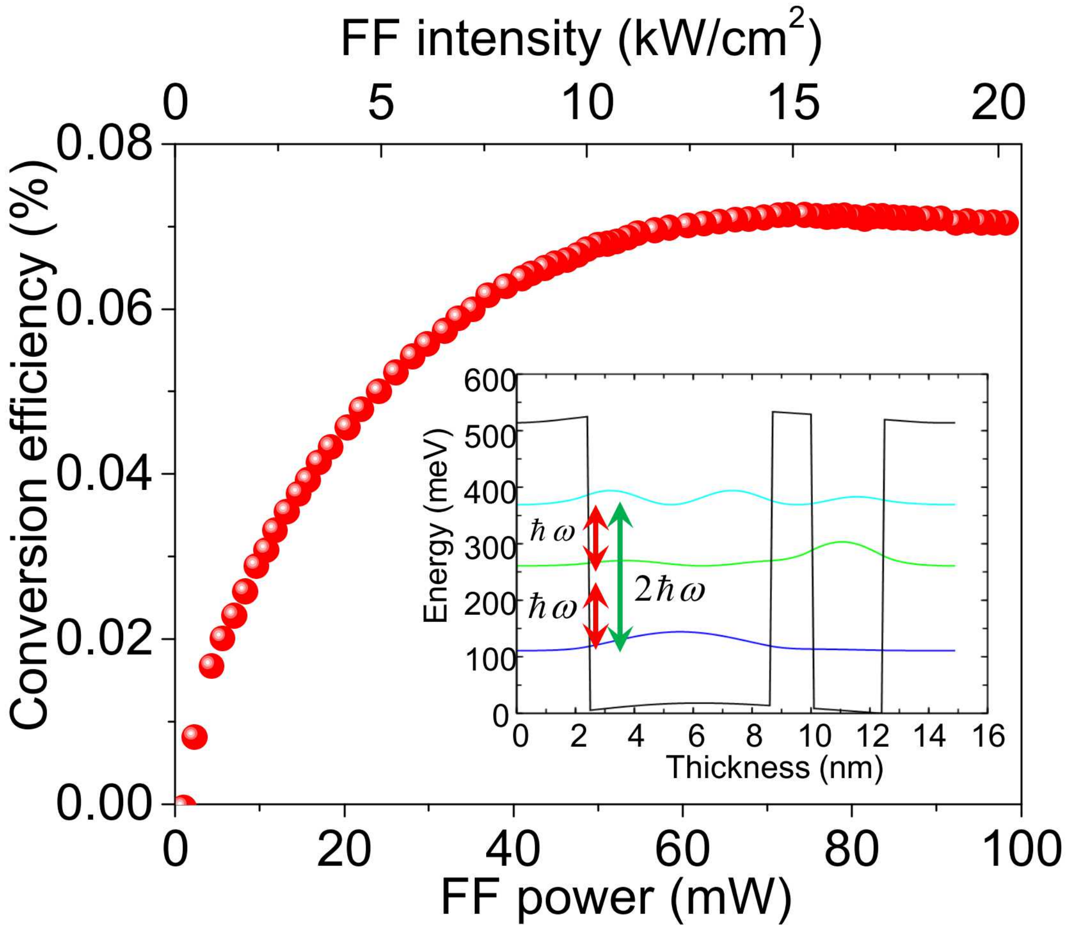


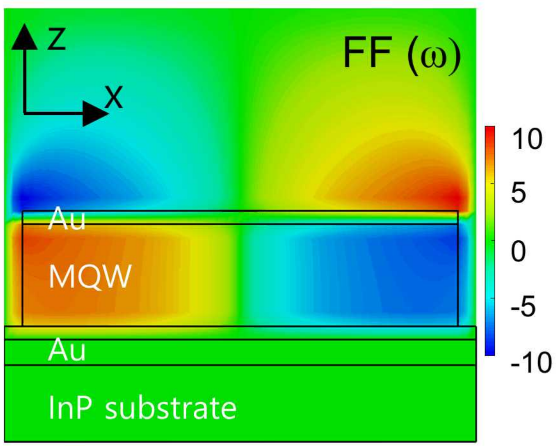


Nookala *et al.*, *Optica* **3,** 283 (2016)

Intensity Saturation





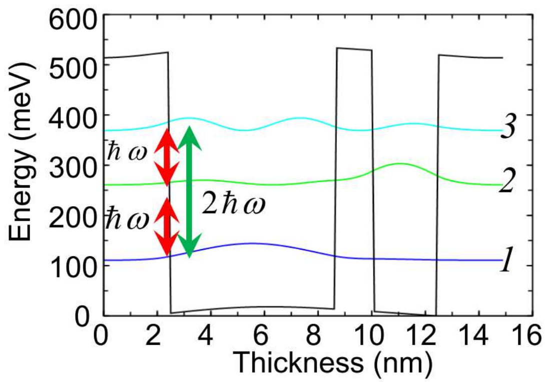


Cross-section along the white line

$$\chi_{zzz}^{(2)} \approx \left(N_1 - 2N_2 + N_3\right) \times$$

$$\times \frac{e^3}{\hbar^2 \varepsilon_0} \frac{z_{12} z_{13} z_{23}}{\left(\omega - \omega_{12} - i\gamma_{12}\right)\left(2\omega - \omega_{13} - i\gamma_{13}\right)}$$

Potential improvements in performance

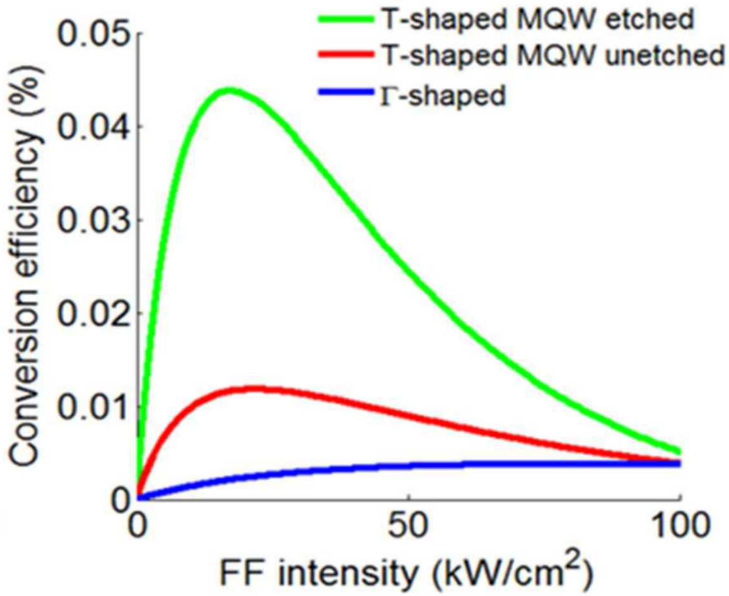


$$I_{sat}(1-2) = \frac{cn\varepsilon_0\hbar}{2e^2 z_{12}^2 \tau_2} \frac{(\hbar\omega - E_{21})^2 + \gamma_{21}^2}{\gamma_{21}} \approx 1.5\ MW/cm^2$$

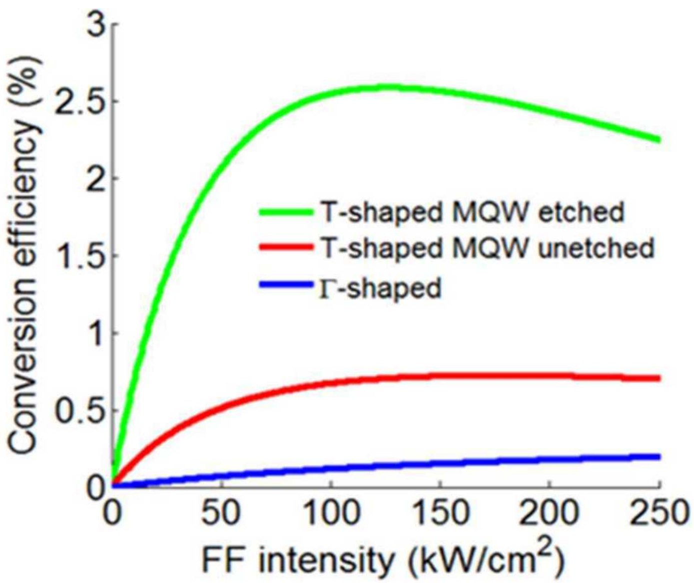
$$2\gamma_{21} \approx 25\ meV$$

$$\tau_2 \approx 1.5\ ps;\ \hbar/\tau_2 \approx 1\ meV$$

Current MQW structure (2γ≈30 meV)

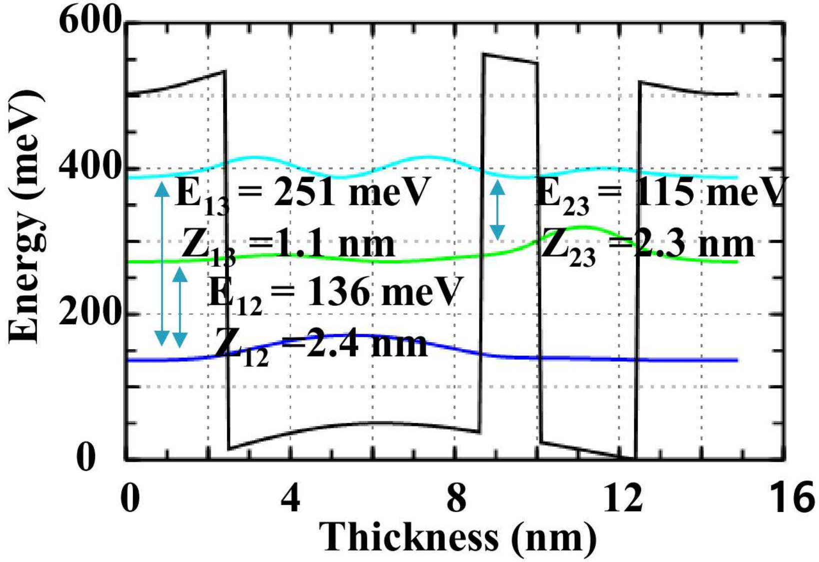


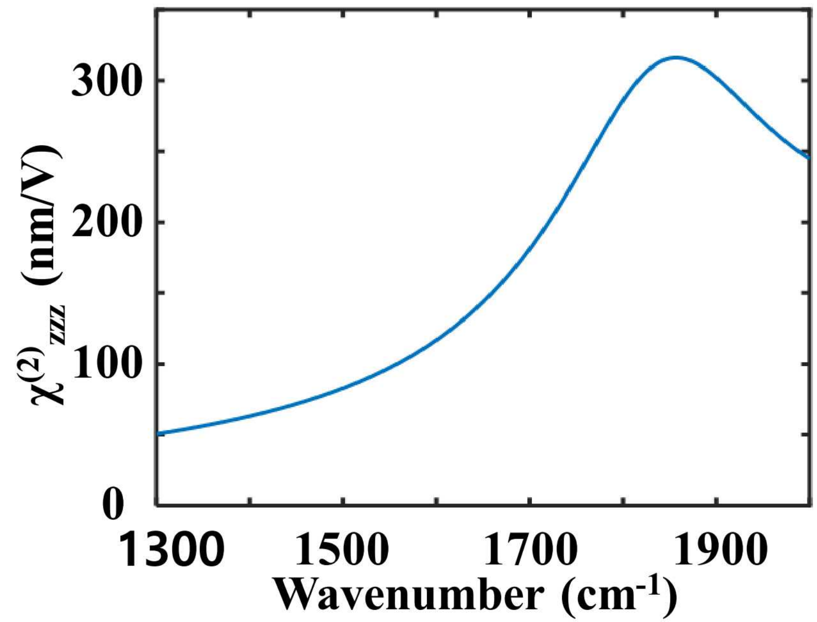
Same MQW structure with (2γ≈10 meV)

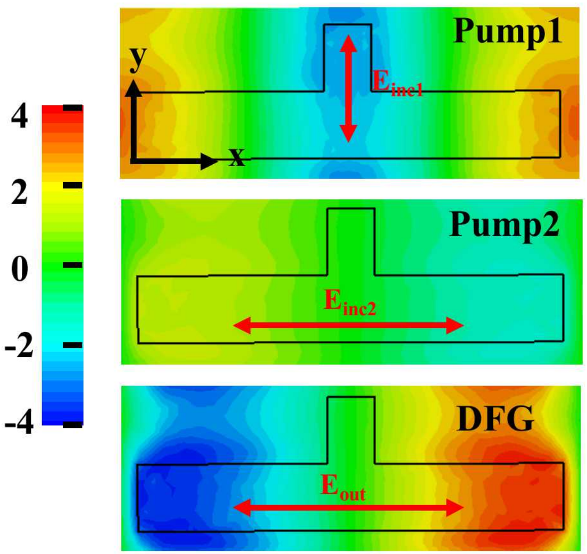


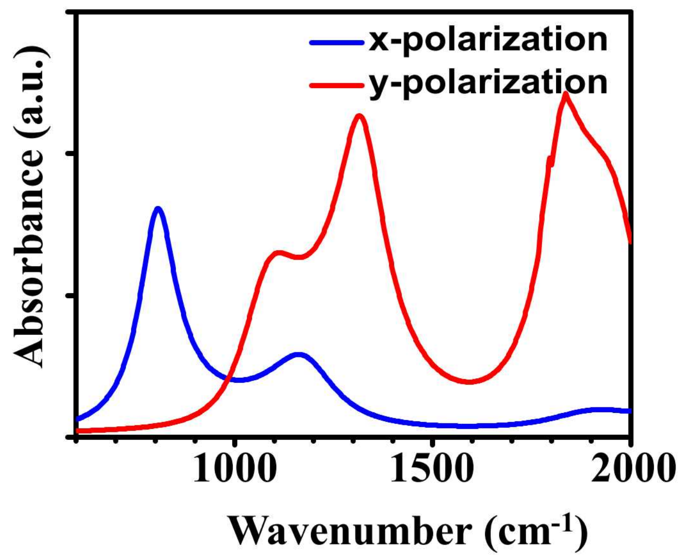
Gomez-Diaz *et al.*, *Phys. Rev. B* **92,** 125429 (2015)

Up- and down-conversion







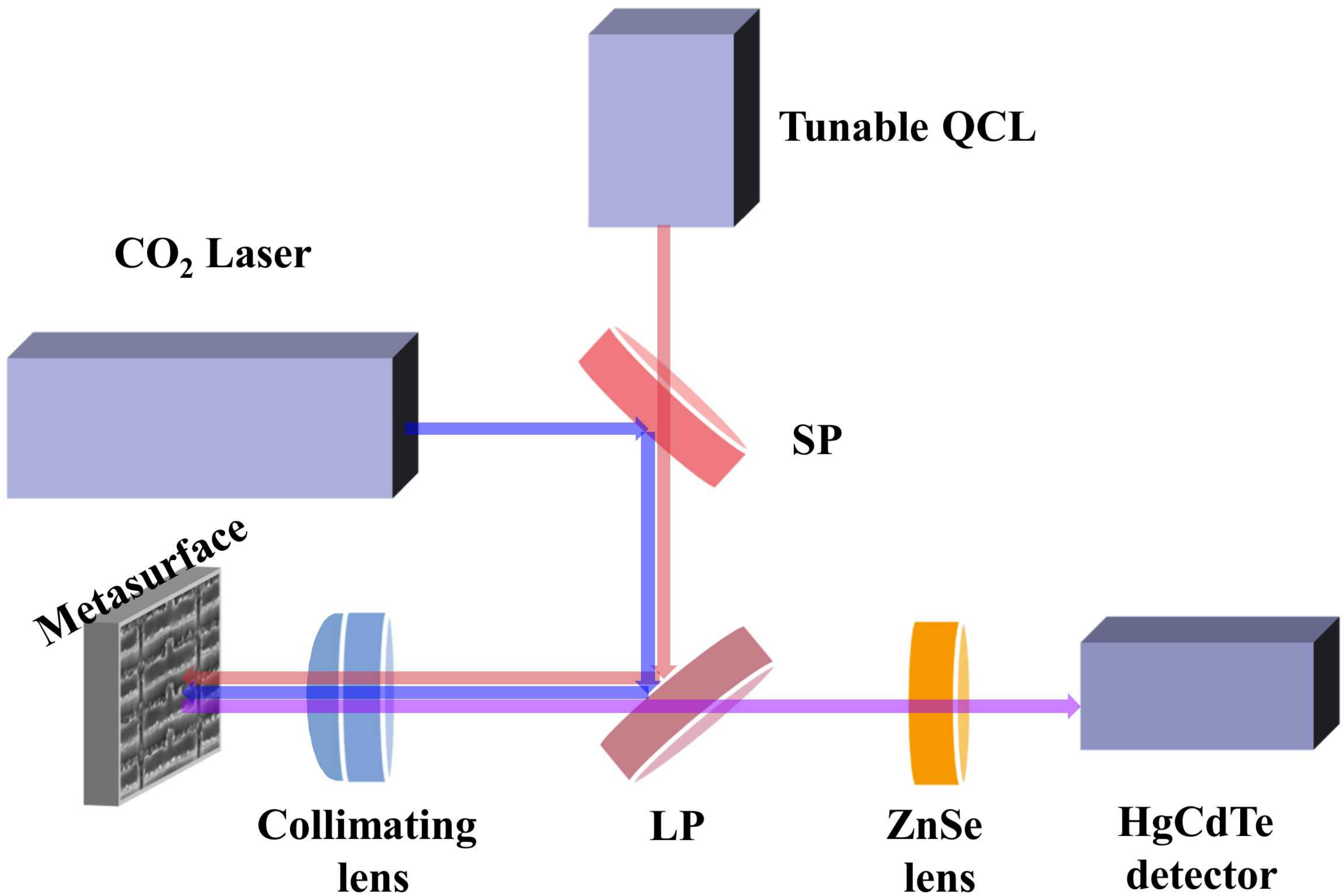


Down-conversion setup

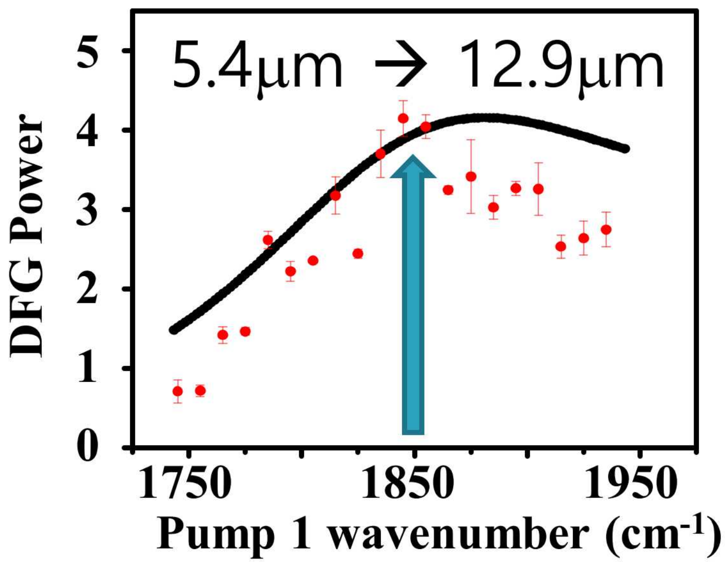
$\lambda_1 \approx 5.1\text{-}5.7\ \mu m$

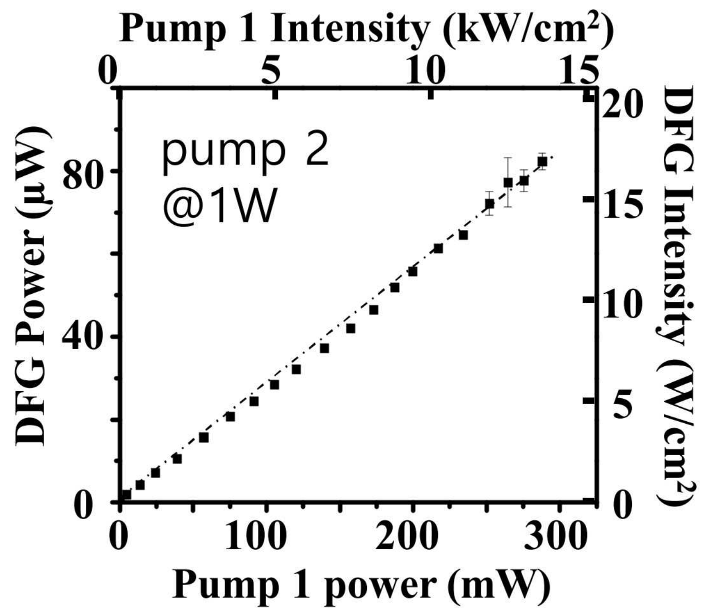
$\lambda_2 \approx 9.3\ \mu m$

$\lambda_{DFG} \approx 11.3\text{-}14.7 \mu m$



Down-conversion results

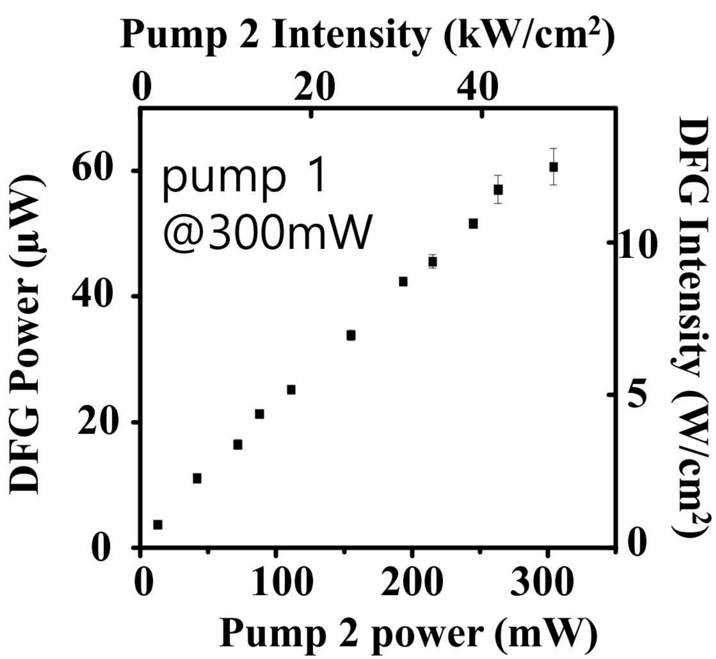




$$I_3 = \frac{\omega_3^2}{2\varepsilon_0 c^3}\left|\chi_{eff}^{(2)}\right|^2 d^2 I_1 I_2$$

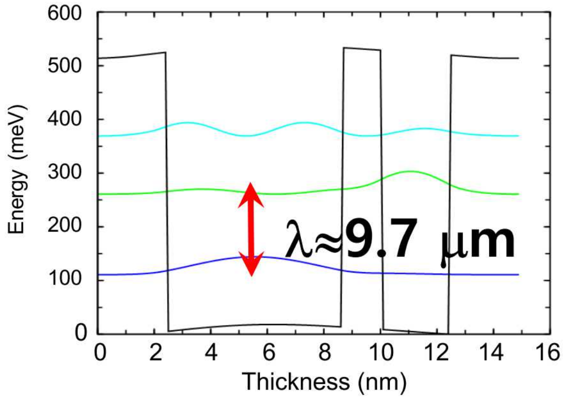
$$\frac{I_3}{I_1} = 0.13\%$$ (at I_2=150 kW/cm^2)

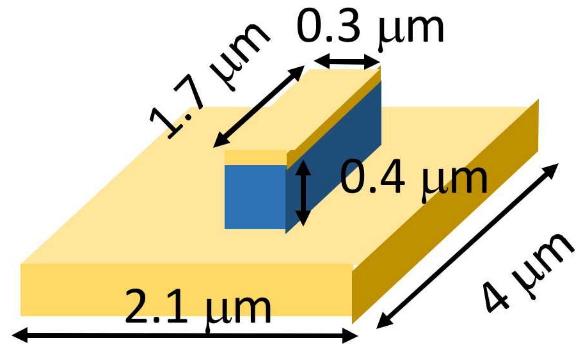
(photon conversion efficiency 0.3%)

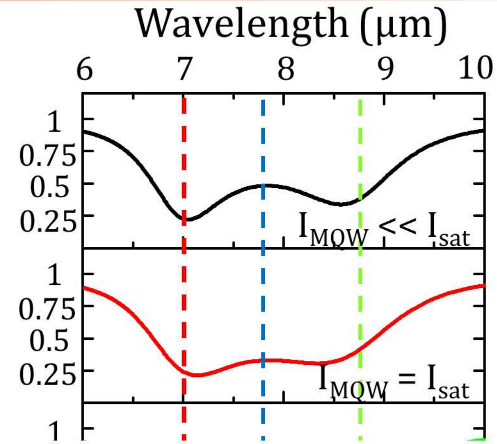


Up-conversion (12.9 μm → 5.4 μm) power efficiency of ~0.75% is expected

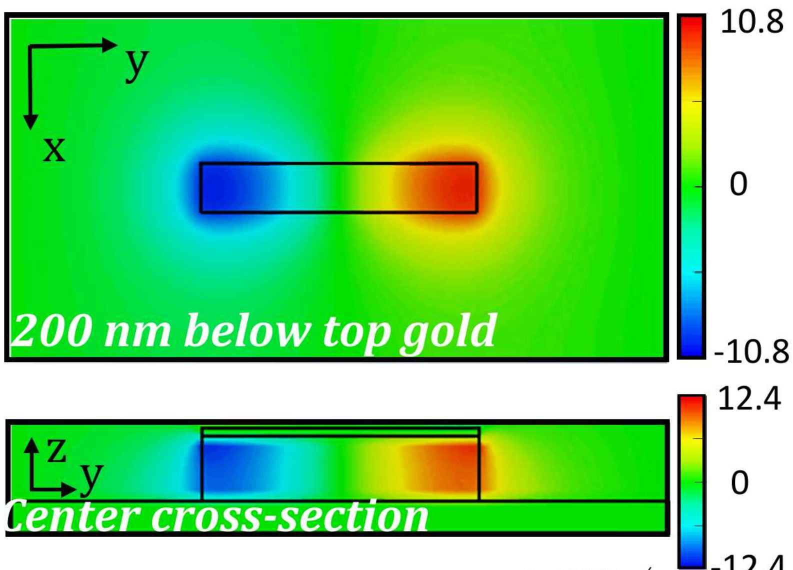
Optical switching and power limiting

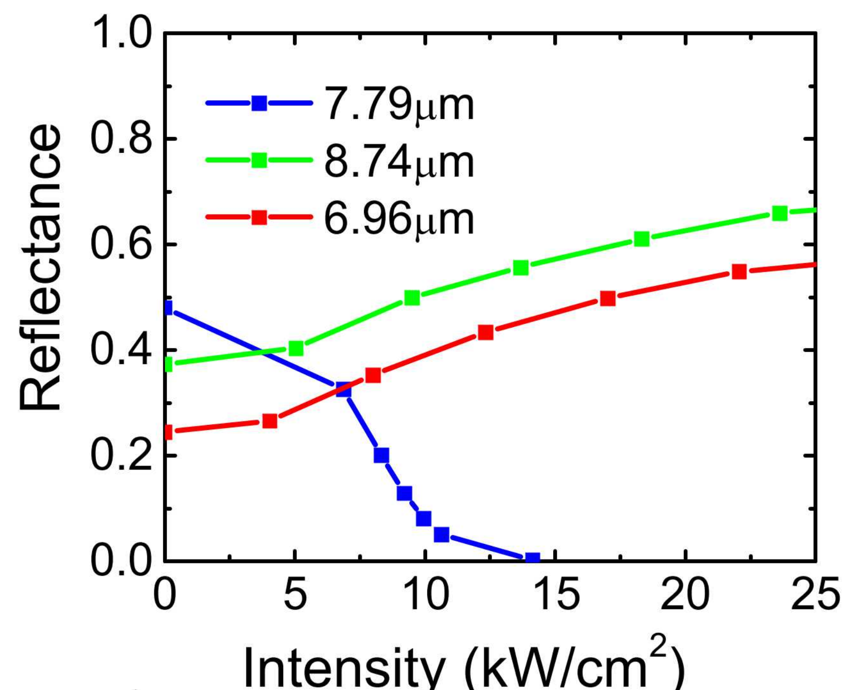


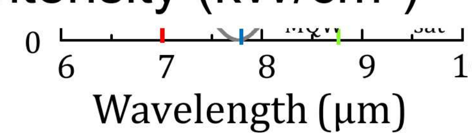




Local field enhancement, $I_{MQW} = 10 I_{sat}$ E_z/E_{inc}

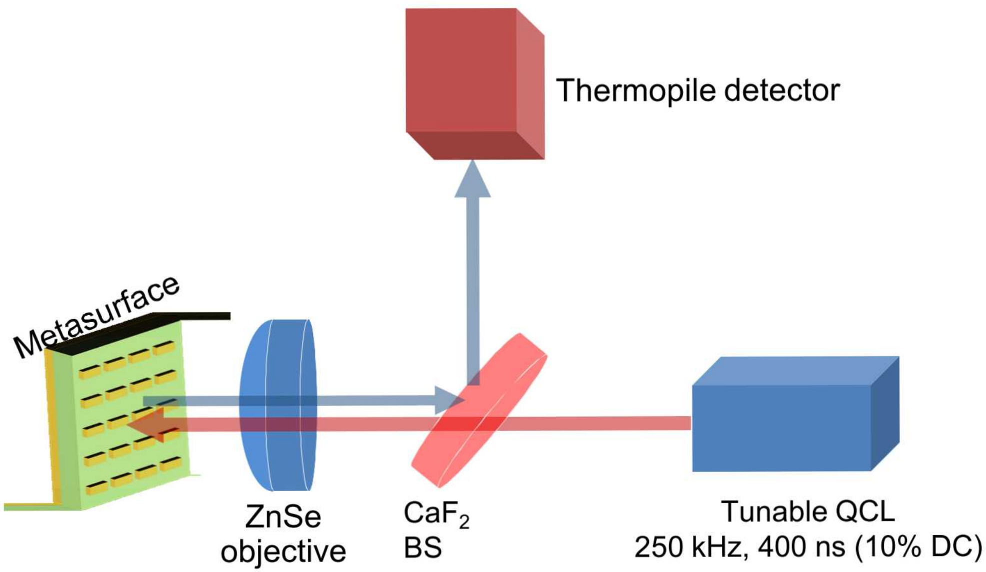


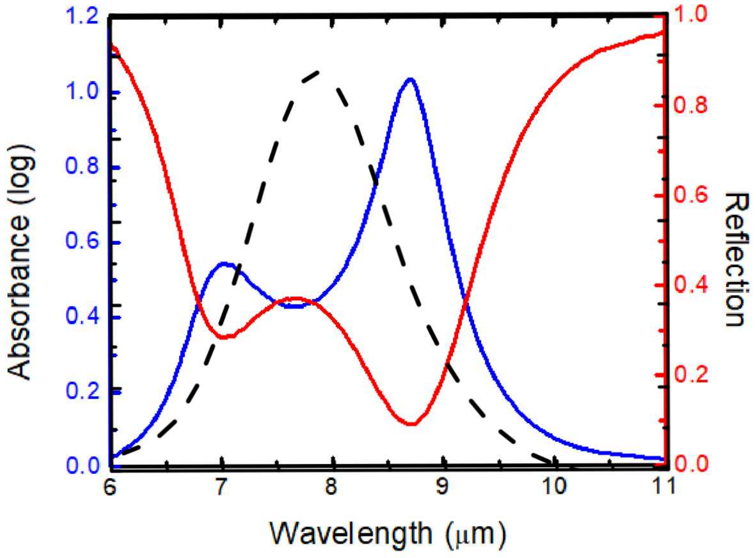


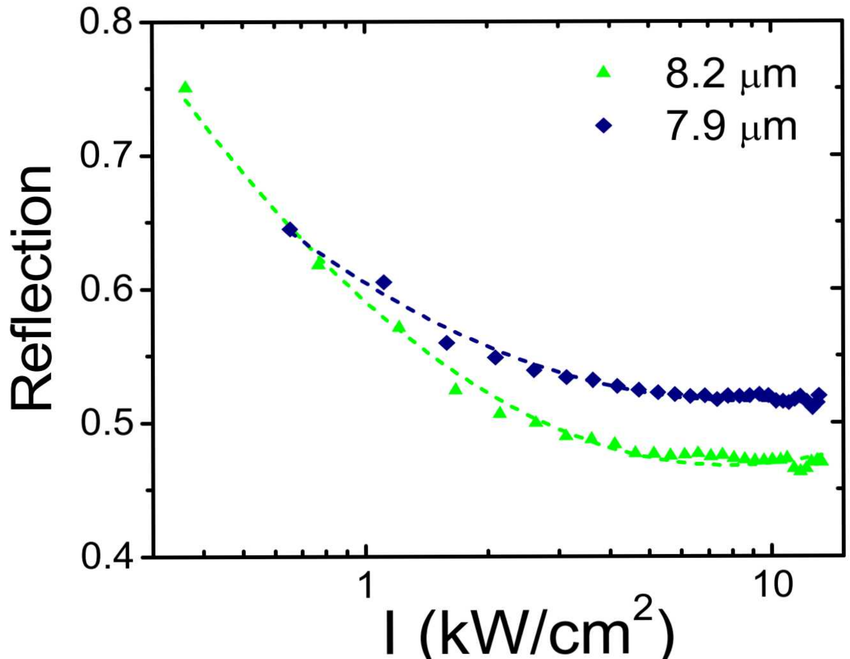


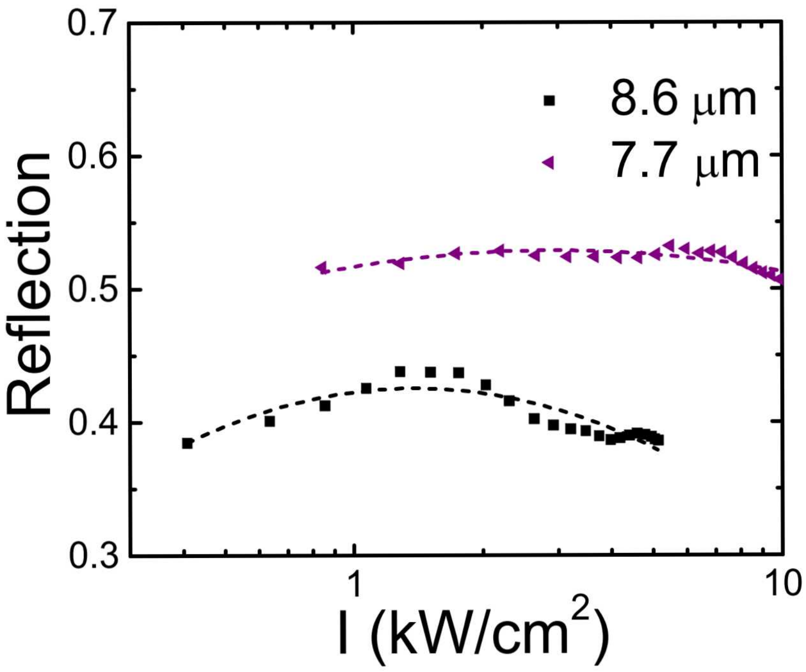
$$I_{inc} = \frac{I_{MQW}}{n_{MQW}\xi^2} = \frac{I_{sat}\times 10}{n_{MQW}\xi^2} = \frac{6.8\,{}^{MW}\!/_{cm^2}}{3.3\times 12.4^2} \approx 13.4\,{}^{kW}\!/_{cm^2}$$

Preliminary experimental results









Summary

- *Highly-nonlinear metasurfaces by combing QM engineering of intersubband transitions and EM engineering of plasmonic reso nators*
- *Giant nonlinear response of 1.2×10^{6} pm/V for SHG*
- *Continuous local phase control of the nonlinear response*
- *Up- and down-conversion: 0.13 % down-conversion efficiency demonstrated; 0.75% up-conversion efficiency is expected*
- *Metasurfaces for optical power limiting and all-optical control*

Funding: ***Sandia Laboratories***