Nonlinear optics in structured graphene and other 2D materials

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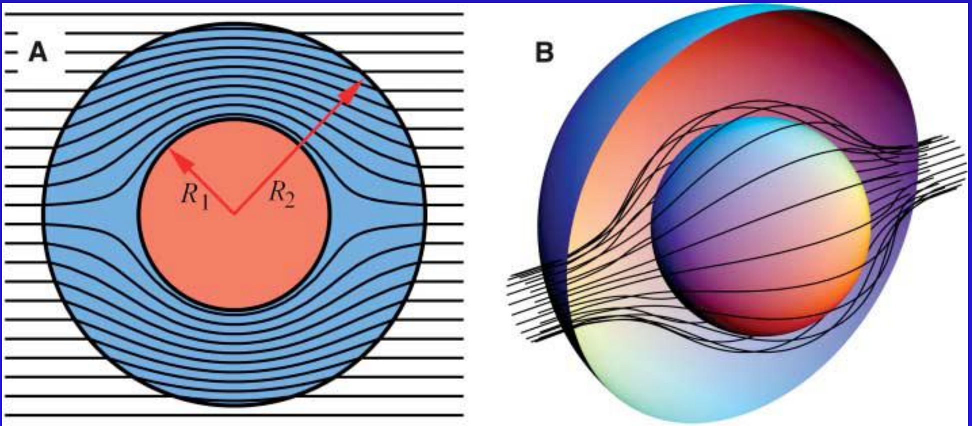
Outline

- Motivation
  - ✓ Two-dimensional materials – efficient light guiding at nanoscale;
  - ✓ Applications: sub-wavelength light concentrators, active photonic nanodevices.
- Computational tools for metamaterials
- Nonlinear optics in 2D materials
  - ✓ SHG in TMDC monolayers;
  - ✓ THG in graphene nanostructures;
  - ✓ Nanodevices based on 2D materials.
- Conclusions and future work

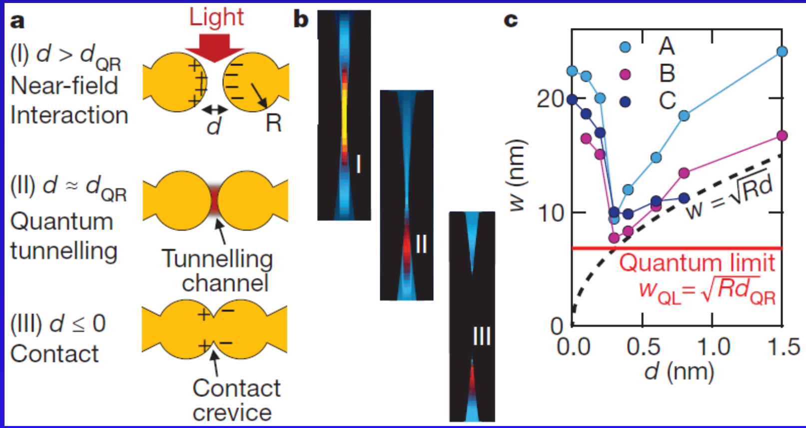
Light-matter Interaction at the Nanoscale

Transformation optics – Cloaking



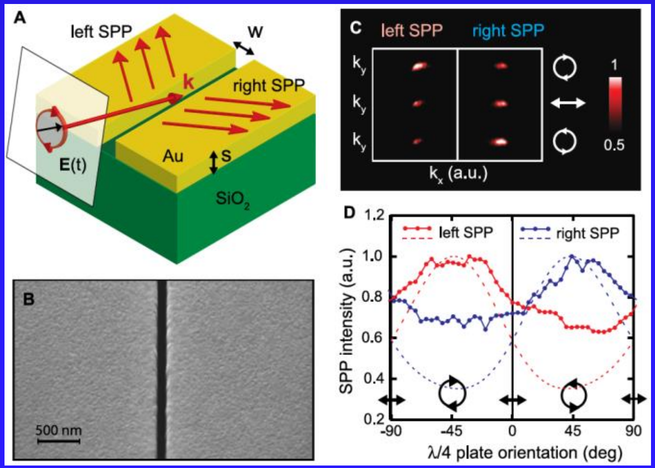
J.B. Pendy, D. Schurig, D.R. Smith, Science **312**, 1780 (2006).

Quantum plasmon tunnelling



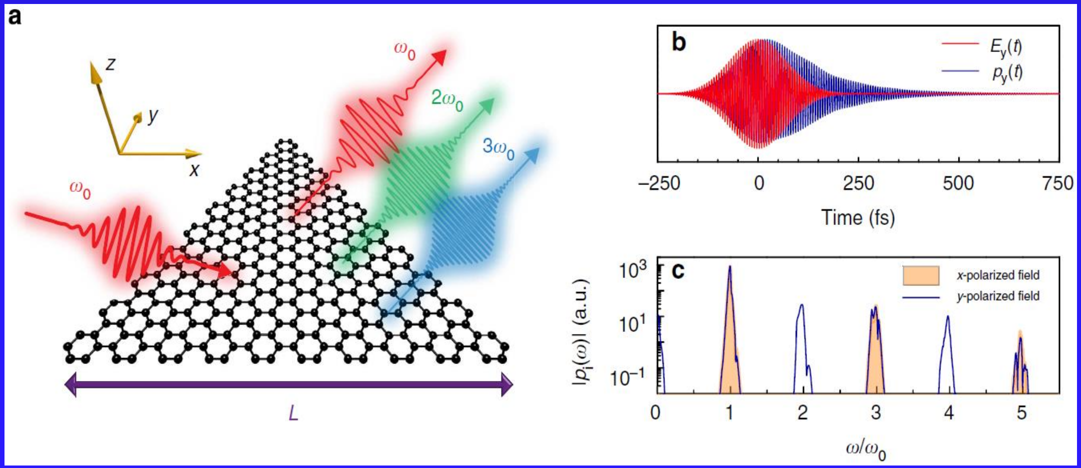
K.J. Savage et al., Nature **491**, 574 (2012).

Near-field manipulation



F.J. Rodriguez-Fortuno et al., Science **340**, 329 (2013).

Nonlinear optics in 2D materials



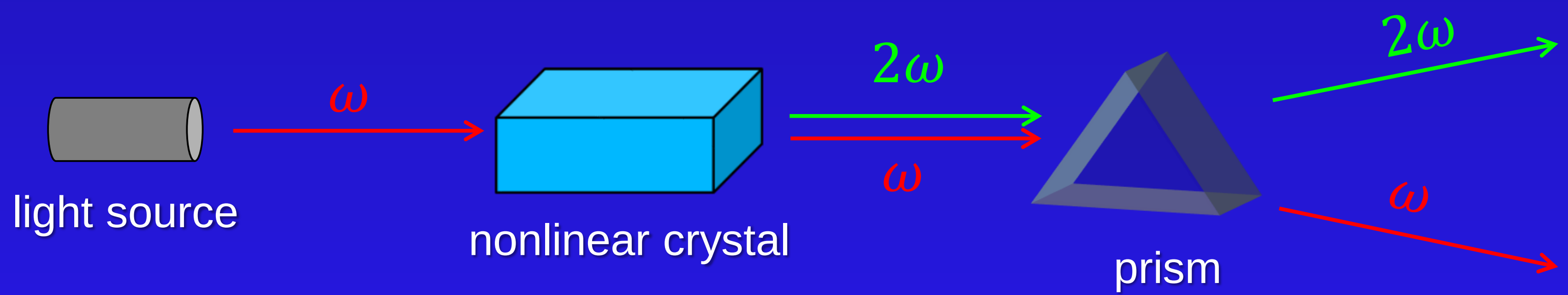
J. D. Cox & F. J. G. de Abajo, Nature Commun. **5**, 5725 (2014).

PCS, April 2018

Modelling Nonlinear Optical Phenomena

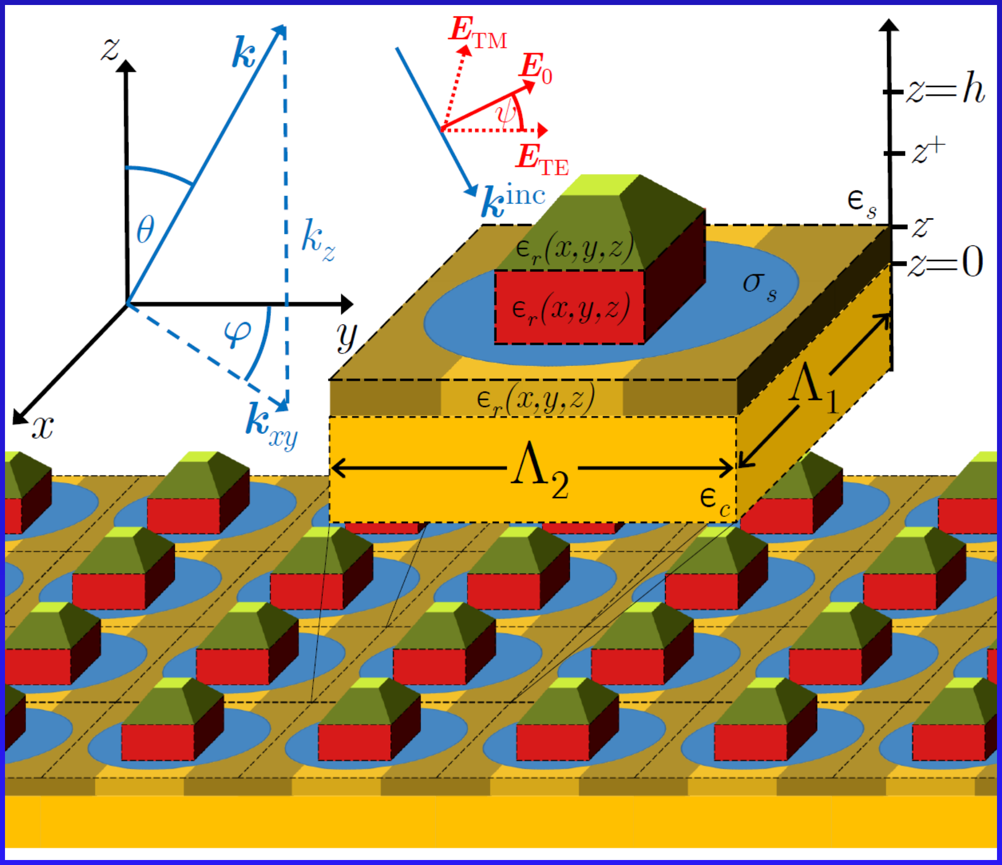
- Nonlinear optics key for many applications

  wavelength conversion, signal processing, optical microscopy



- Challenges:
  - ✓ Practical: nonlinear optical response is generally weak
    - ➢ need for local field enhancement
    - ➢ gratings are devices for engineering of optical near and far-field
  - ✓ Theoretical: complex dependency between excitation and optical response
    - ➢ efficient numerical tools for nonlinear gratings essential

Diffraction Gratings with 2D Materials



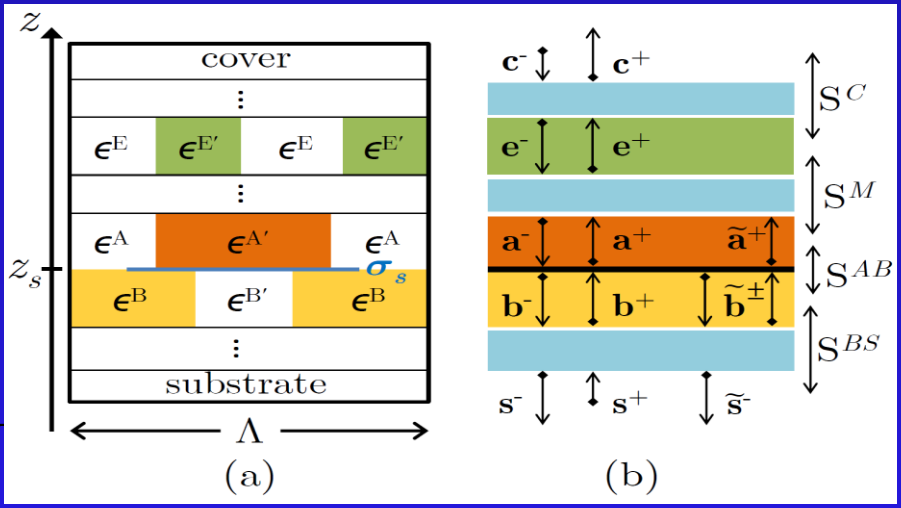
- ✓ Multilayered periodic structure $\epsilon(x,y,z)$
- ✓ Periodically patterned 2D material layers $\sigma_s(x,y)$
  No effective height!
- ✓ Nonlinear surface current
  $\boldsymbol{j}^{\mathrm{NL}} = \sigma^{(3)}(\boldsymbol{E}\cdot\boldsymbol{E})\boldsymbol{E}$ or
  $\boldsymbol{j}^{\mathrm{NL}} = \sigma^{(2)}:\boldsymbol{E}\boldsymbol{E}$

Incident plane wave at FF → Linear diffraction → Total field $\boldsymbol{E}$ at FF → Nonlinear polarization $\boldsymbol{P}^{NL}$ → Nonlinear field at TH or SH

*M. Weismann & N. C. Panoiu, Phys. Rev. B **94**, 035435 (2016)*

RCWA with Inhomogeneous *S*-Matrix

In each layer:
Mode expansion of $\boldsymbol{E}$ and $\boldsymbol{H}$ with coefficients $\mathbf{a}^{\pm}$ and $\mathbf{b}^{\pm}$



Boundary conditions

$$\boldsymbol{n} \times \left(\boldsymbol{E}^{\mathrm{A}} - \boldsymbol{E}^{\mathrm{B}}\right) = 0$$

$$\boldsymbol{n} \times \left(\boldsymbol{H}^{\mathrm{A}} - \boldsymbol{H}^{\mathrm{B}}\right) = \boldsymbol{n} \times \left[\boldsymbol{j}(\boldsymbol{E}) + \boldsymbol{j}^{\mathrm{NL}}\right]$$

$\boldsymbol{j}(\boldsymbol{E}, \boldsymbol{x}) = \sigma(\boldsymbol{x})\boldsymbol{E}(\boldsymbol{x})$ with $\sigma(\boldsymbol{x}_0) = 0$
$\Rightarrow$ Fast factorisation difficult

$\boldsymbol{j}(\boldsymbol{E}) = (\sigma(\boldsymbol{x}) + \eta\sigma_s)\boldsymbol{E}(\boldsymbol{x}) \quad (\eta \ll 1)$
$\Rightarrow$ Fast Fourier factorisation possible

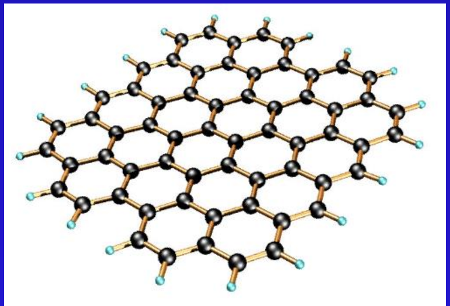
Khavasi, OL ***38****, 3009 (2013)*

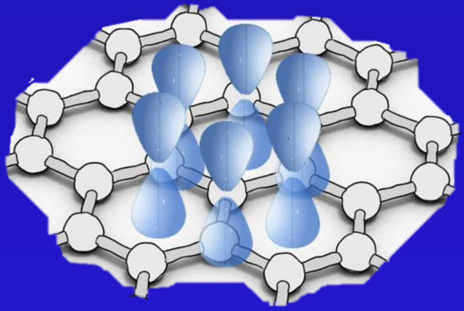
***S*-Matrix for modified interface**

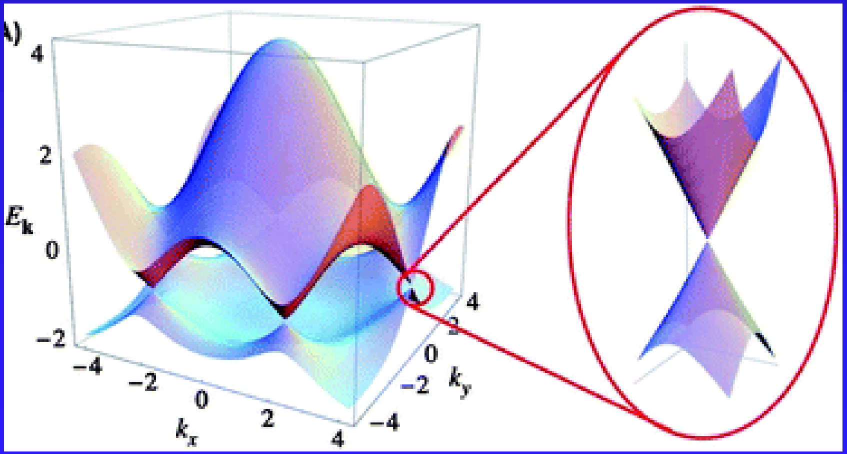
$$\begin{bmatrix}\mathbf{a}^{+}\\ \mathbf{b}^{-}\end{bmatrix} = \mathrm{S}\begin{bmatrix}\mathbf{a}^{-}\\ \mathbf{b}^{+}\end{bmatrix} + \mathrm{T}\begin{bmatrix}j_x^{\mathrm{NL}}\\ j_y^{\mathrm{NL}}\end{bmatrix} = \mathrm{S}\begin{bmatrix}\mathbf{a}^{-}\\ \mathbf{b}^{+}\end{bmatrix} + \begin{bmatrix}\tilde{\mathbf{a}}^{-}\\ \tilde{\mathbf{b}}^{+}\end{bmatrix}$$

Graphene: Structure and Properties

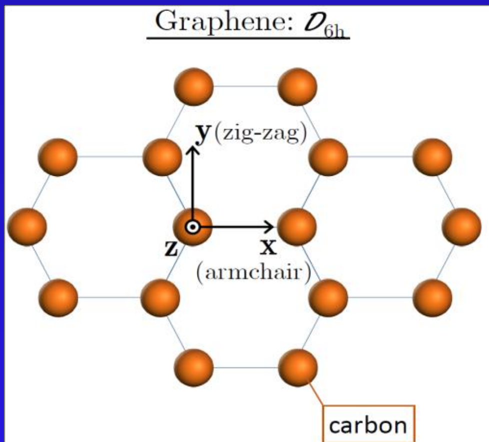
- ✓ One atomic layer thick
- ✓ 2D honeycomb crystal lattice
- ✓ Zero-gap semiconductor
- ✓ Almost transparent with absorption tunable via doping
- ✓ High electron mobility
- ✓ Low electron scattering
- ✓ Remarkable mechanical properties







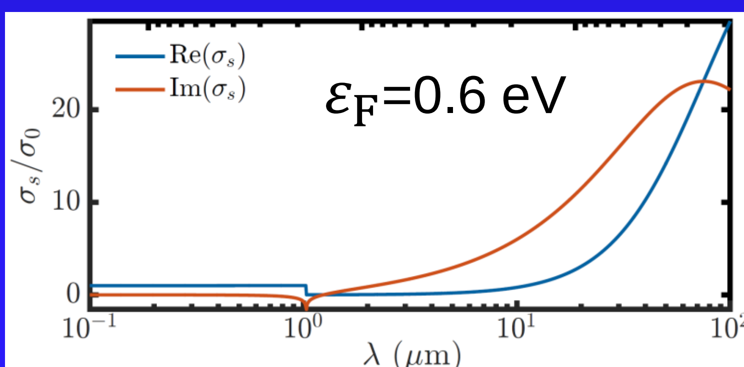
Optical Properties of 2D Materials
Graphene



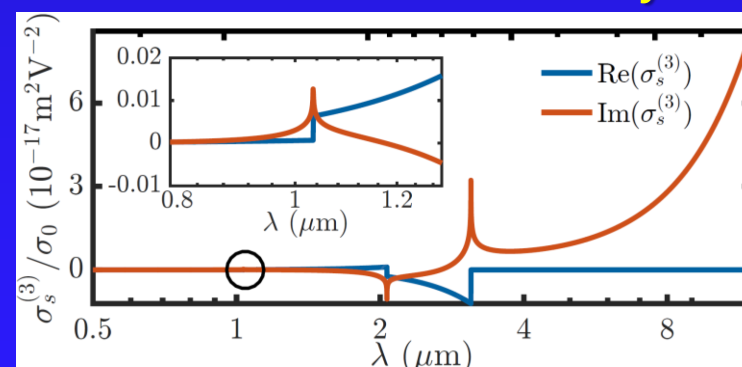
- ✓ high electron mobility, low optical losses
- ✓ electrically tuneable conductance
- ✓ centrosymmetric ⇒ no SHG, strong THG

$$\frac{\sigma_s(\omega)}{\sigma_0} = \frac{4\varepsilon_F}{\pi\hbar}\frac{i}{\omega + i\tau^{-1}} + \theta(\hbar\omega - 2\varepsilon_F) + \frac{i}{\pi}\ln\left|\frac{\hbar\omega - 2\varepsilon_F}{\hbar\omega + 2\varepsilon_F}\right|$$

Sheet conductance



Third-order conductivity

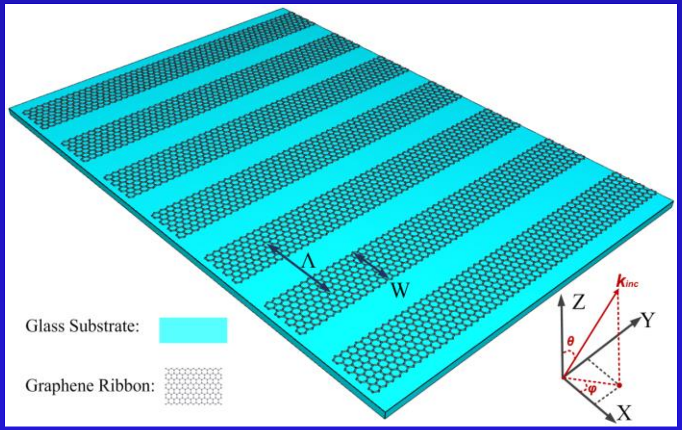


Cheng, New J. Phys. 053014 (2014)

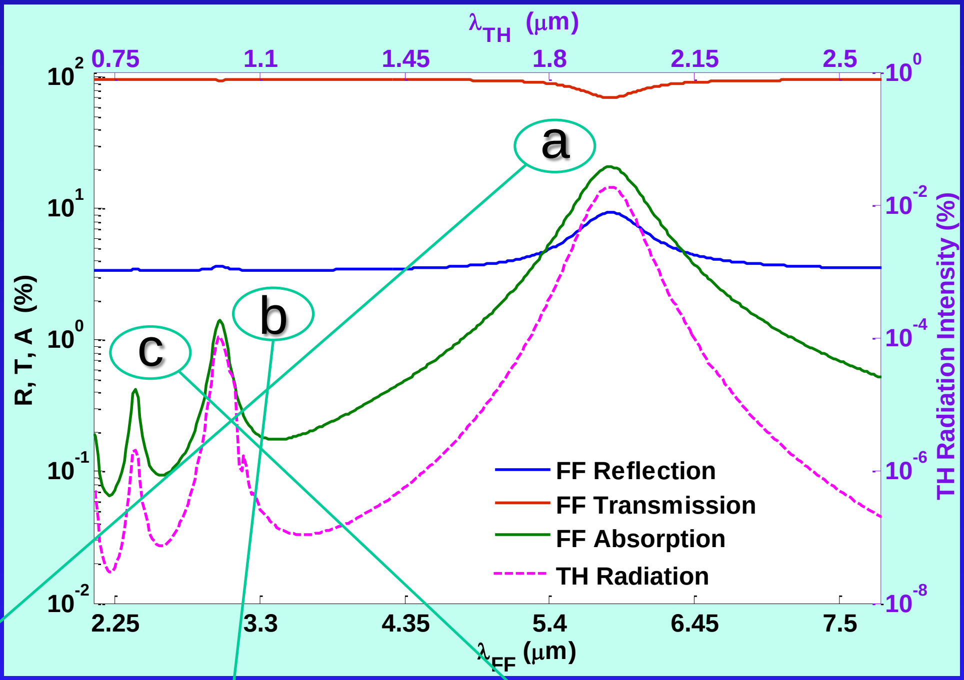
- ✓ Applications to modulators, efficient nonlinear optical devices at the nanoscale

PCS, April 2018

1D Diffraction Gratings – Graphene Ribbons



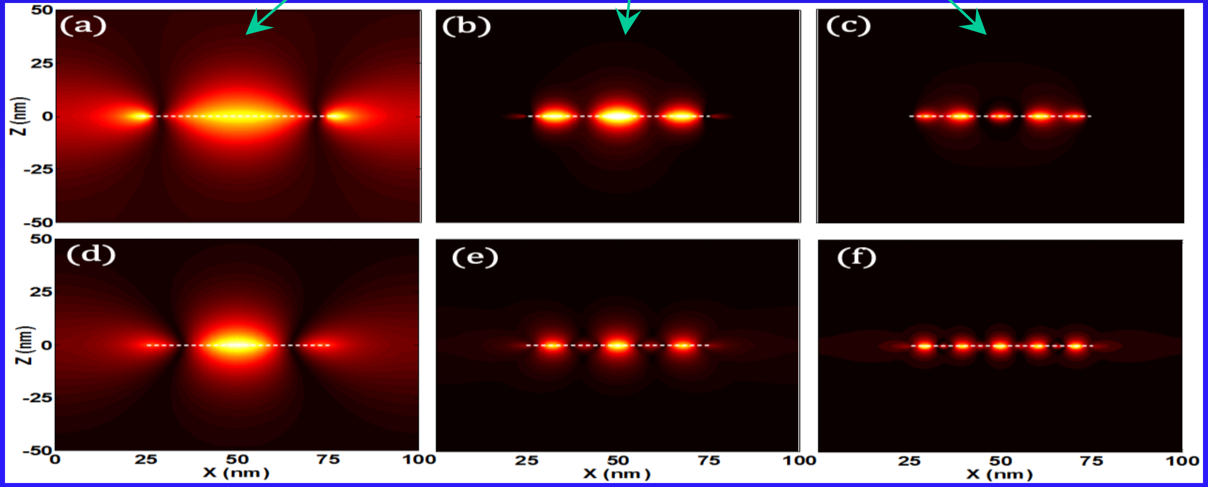
$\Lambda = 100$ nm; $w = 50$ nm



- Optical near-field

Fundamental frequency

Third harmonic



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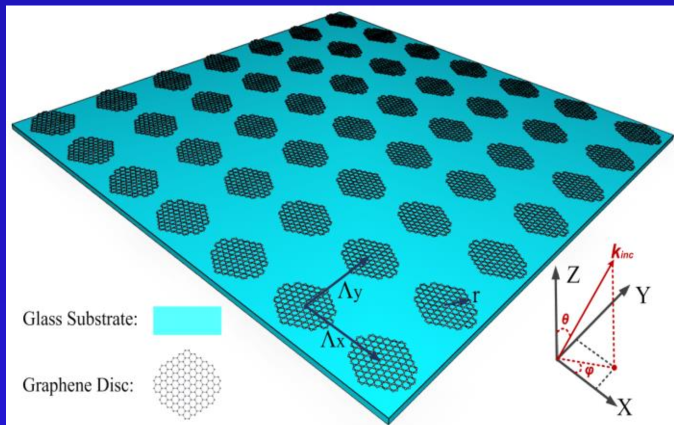
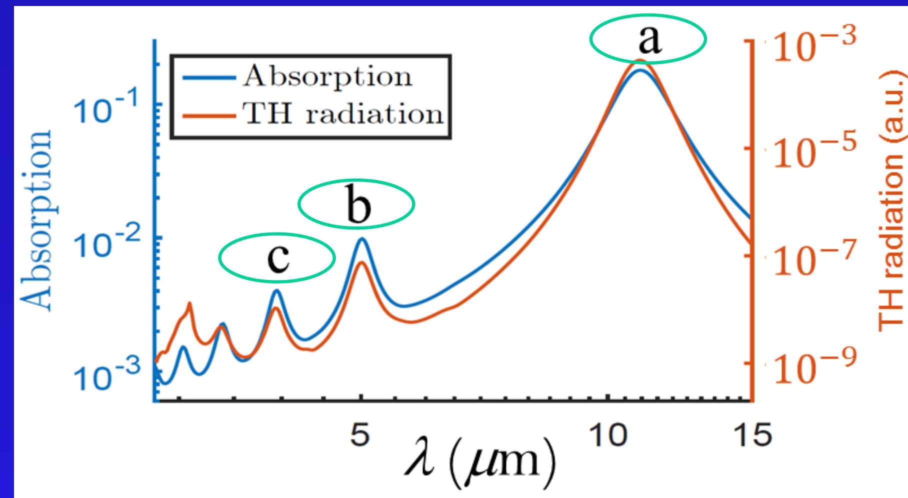
2D Diffraction Gratings – Graphene Disks



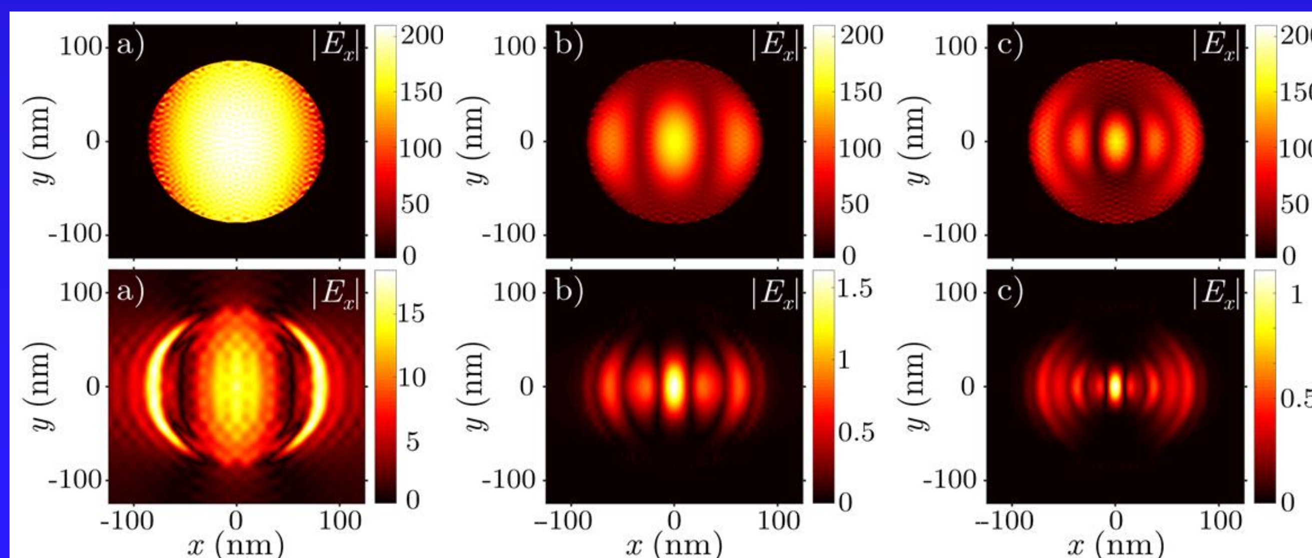
$\Lambda = 250\ \mathrm{nm}; D = 175\ \mathrm{nm}$



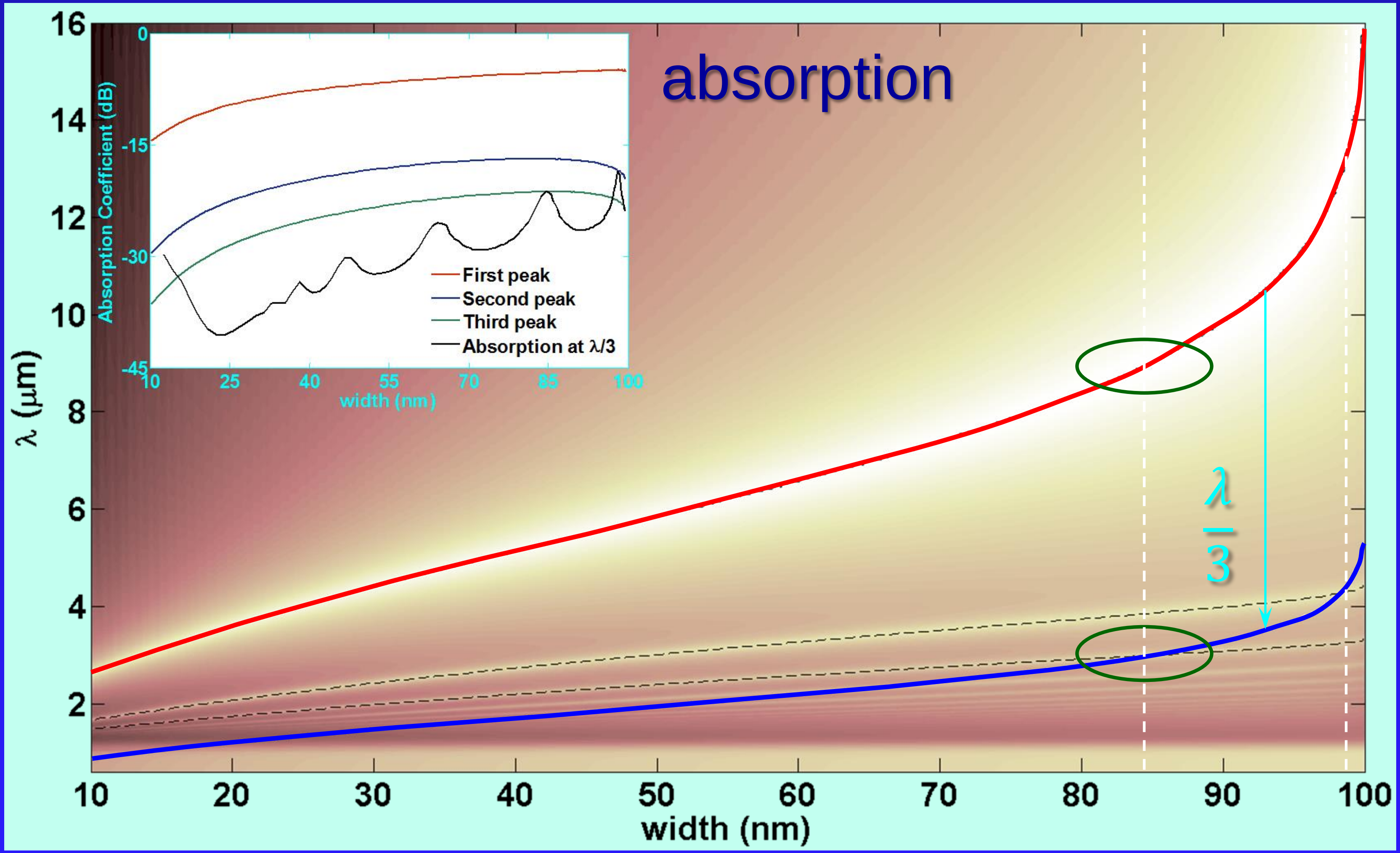
❑ Optical near-field

Fundamental frequency

Third harmonic



Mechanisms for Nonlinearity Enhancement

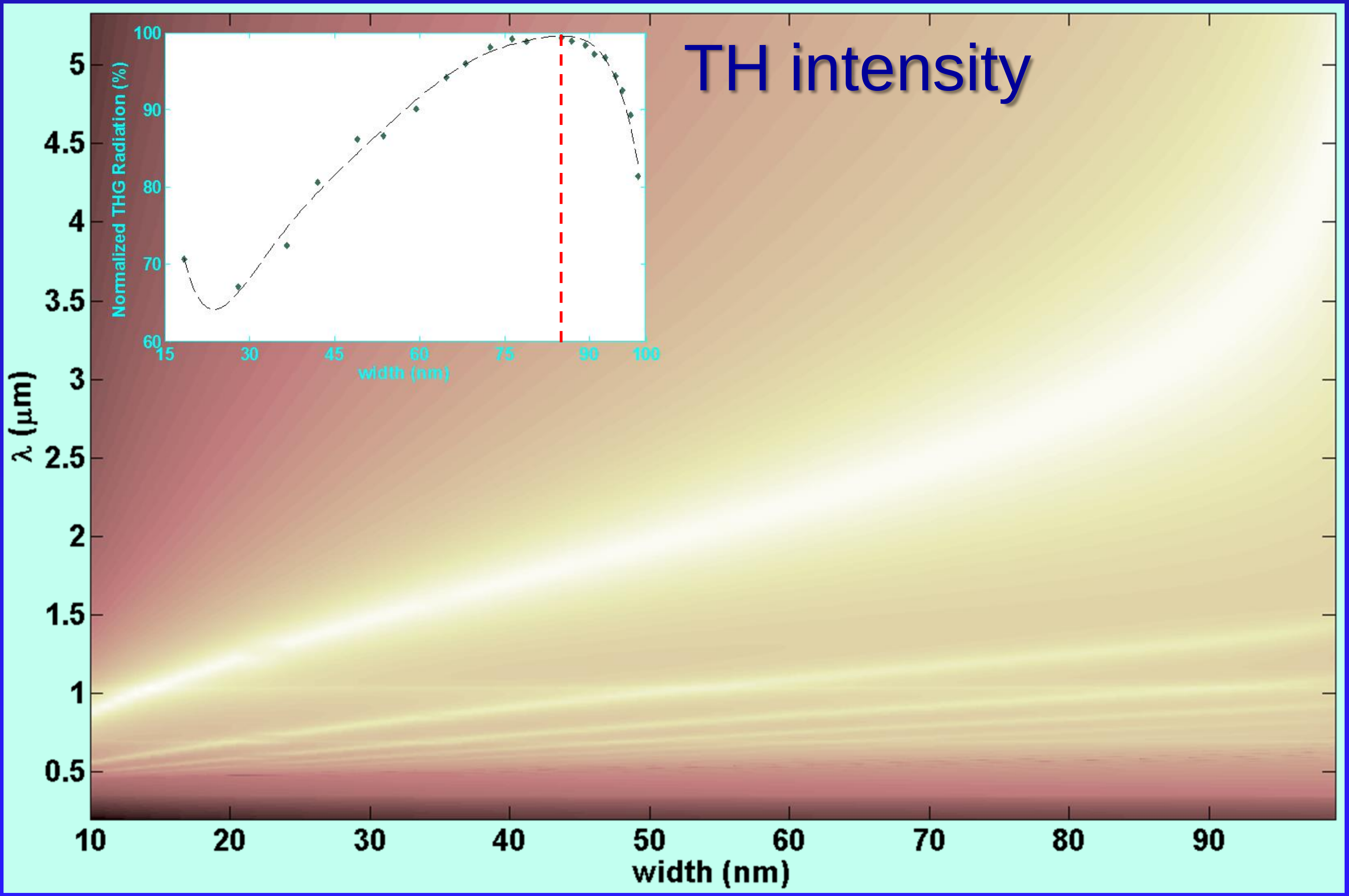


✓ Dual resonance condition: optical modes exist at λ and λ/3;

✓ Enhanced nonlinear optical response.

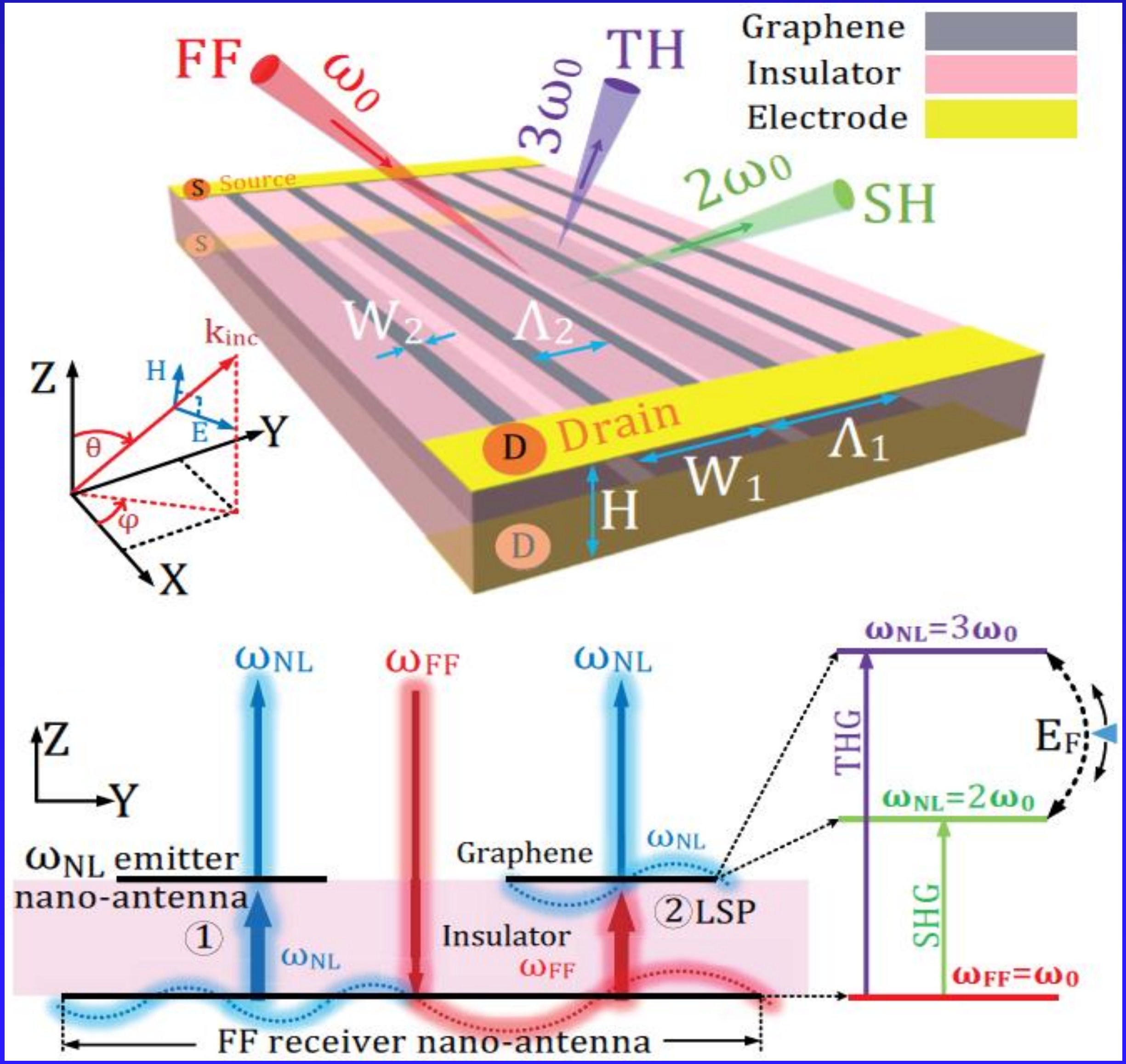
J. W. You et al., Phil. Trans. R. Soc. A **375**, 20160313 (2017)

Mechanisms for Nonlinearity Enhancement



Strong nonlinear optical response when the dual resonance condition is satisfied

Mechanisms for THG Enhancement



Fundamental plasmon at FF: efficient light in-coupling

Fundamental plasmon at TH: efficient emitter

Tune $E_F \rightarrow$ switch between TH and SH

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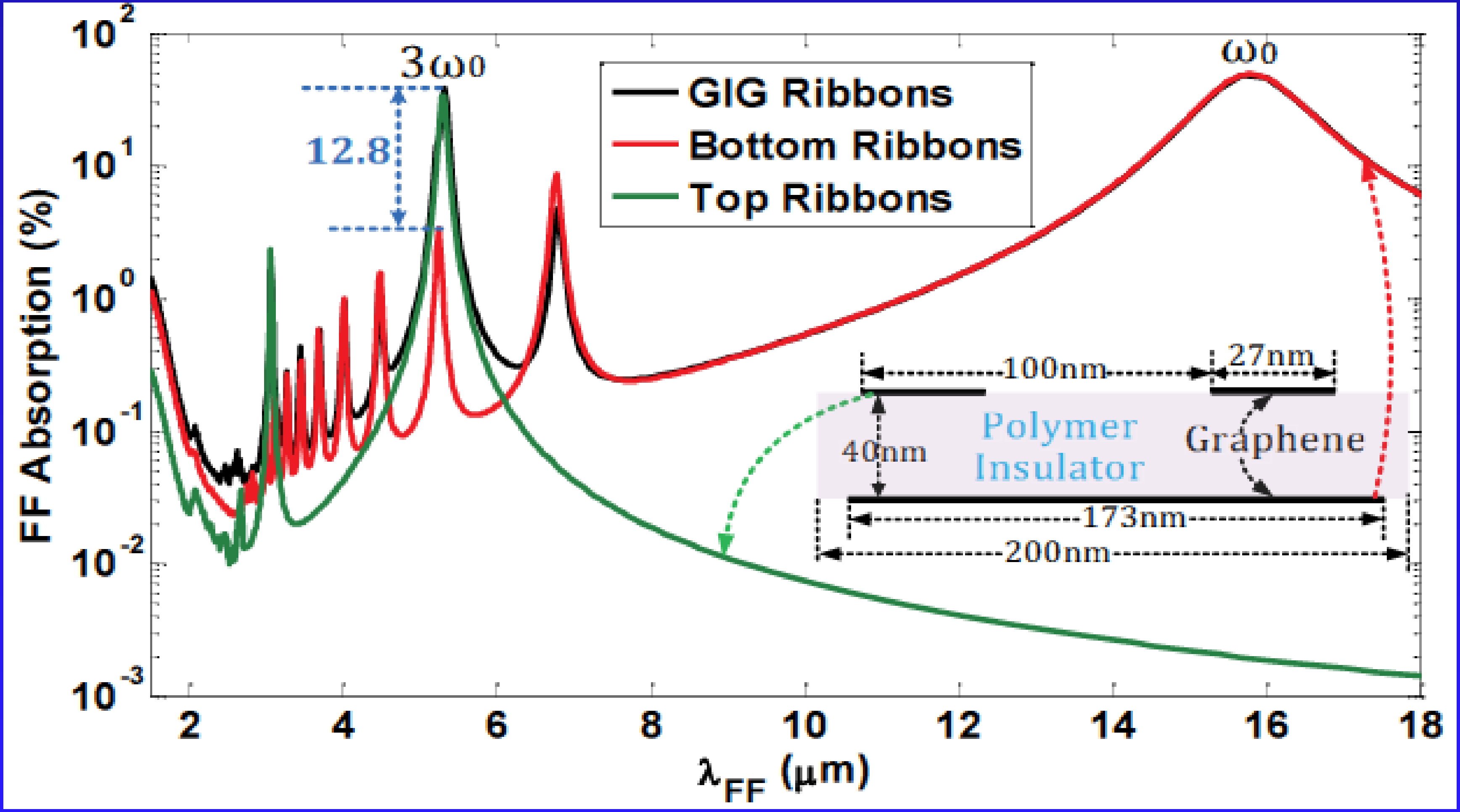
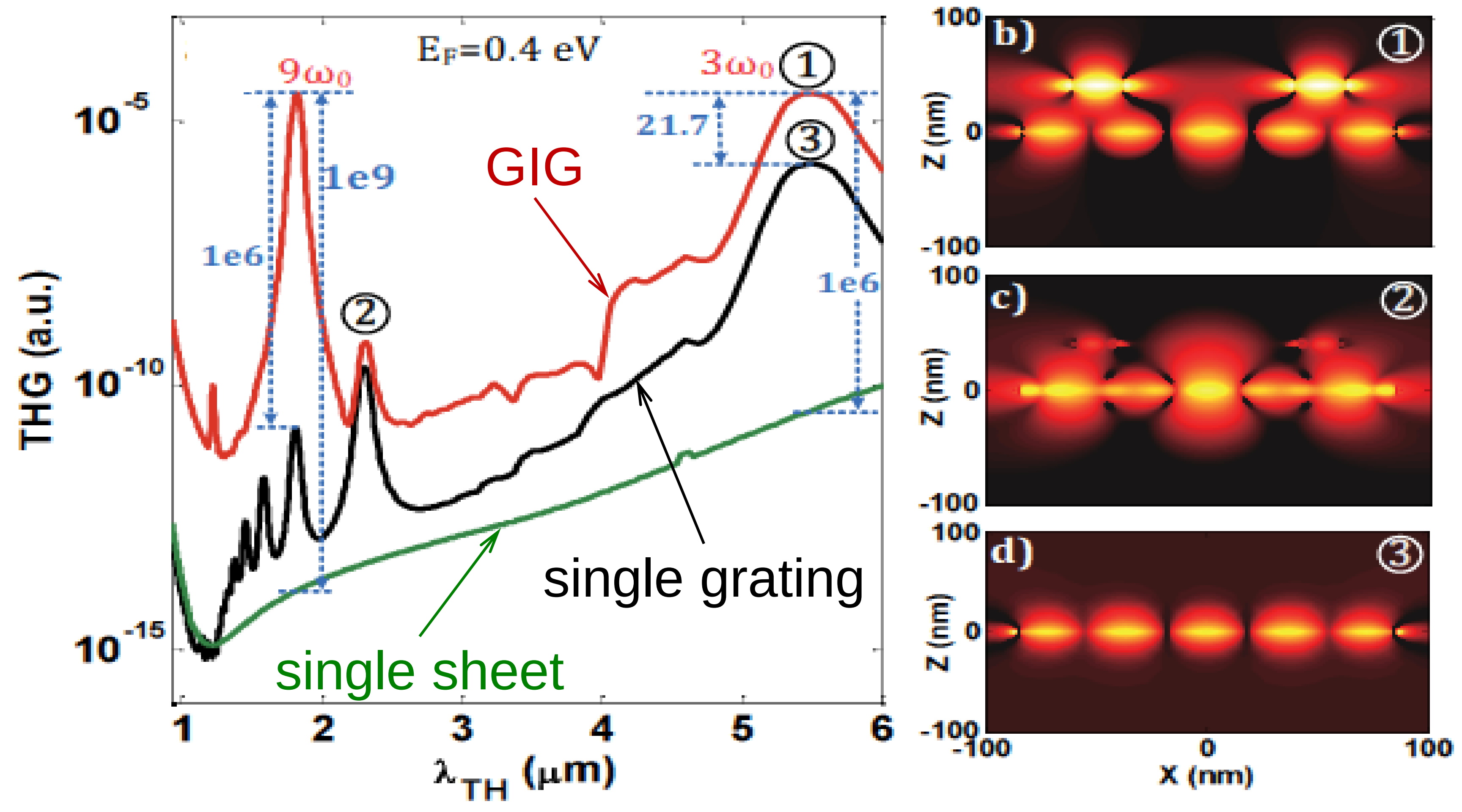
Bi-layer Grating – Linear Response



- ✓ Strongly enhanced absorption
- ✓ Fundamental plasmons exist at both FF and TH

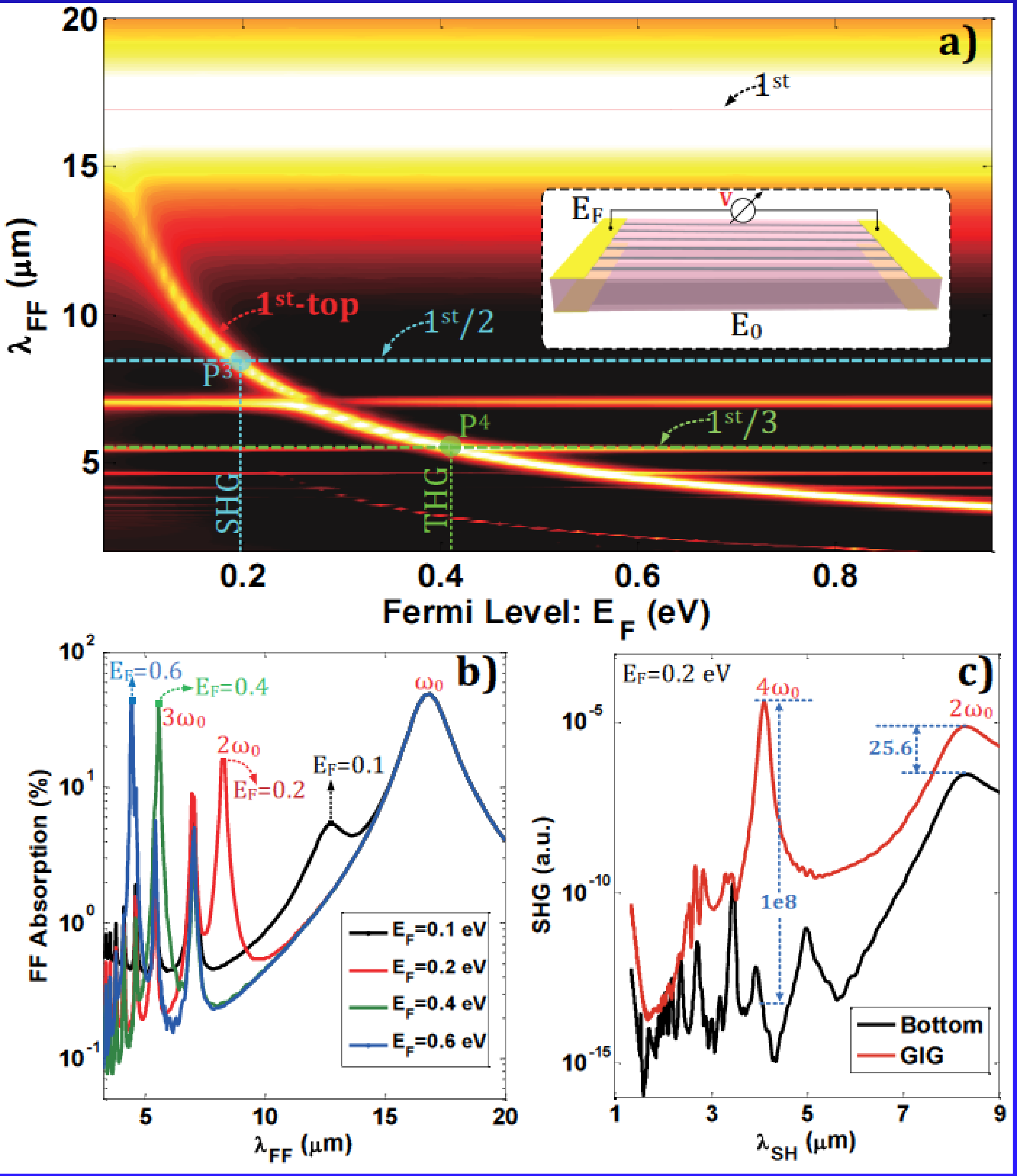
Bi-layer Grating – THG



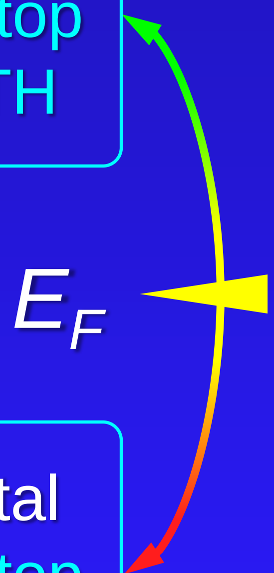
✓ 20× THG enhancement wrt single-layer grating

✓ ~10^6 THG enhancement wrt graphene sheet

Switching between THG and SHG



Fundamental plasmon of top grating at TH



Fundamental plasmon of top grating at SH

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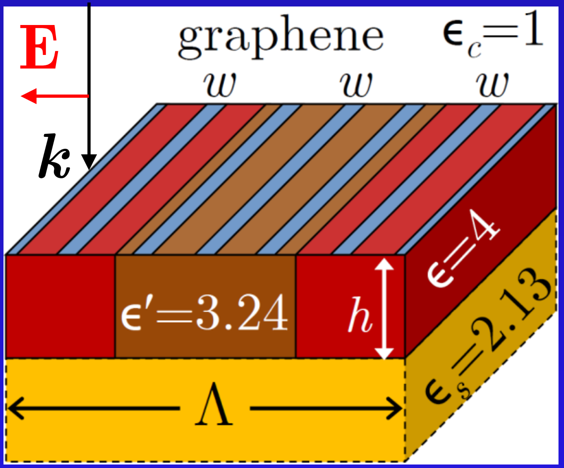
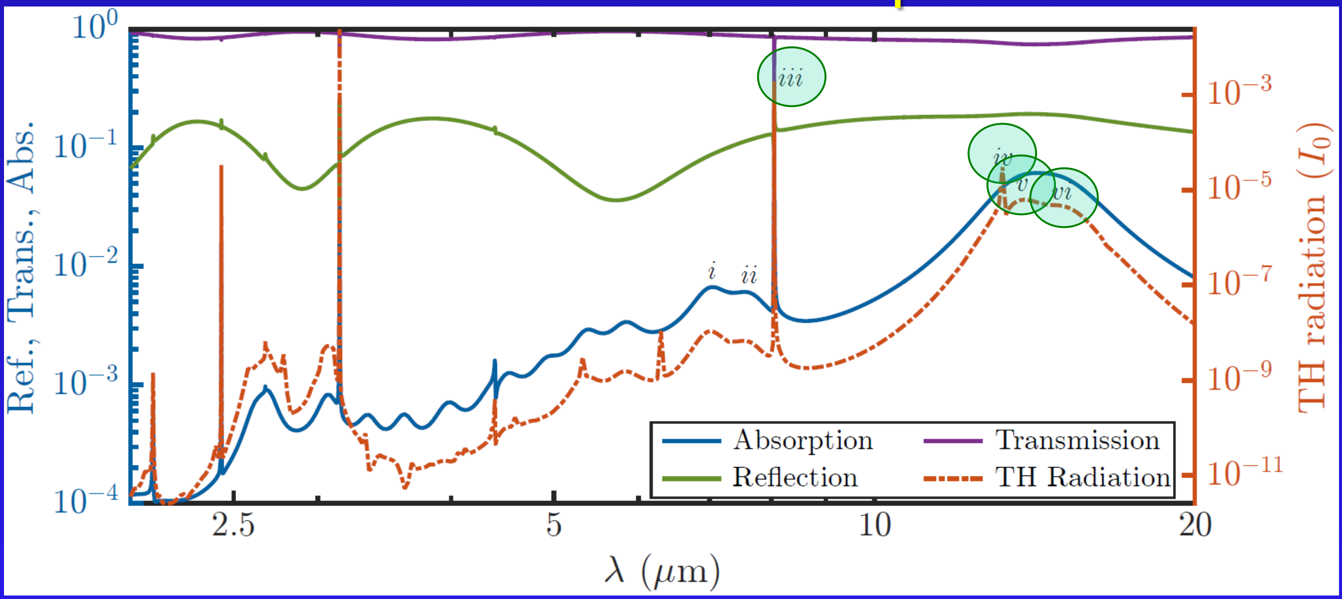
Mechanisms for Nonlinearity Enhancement



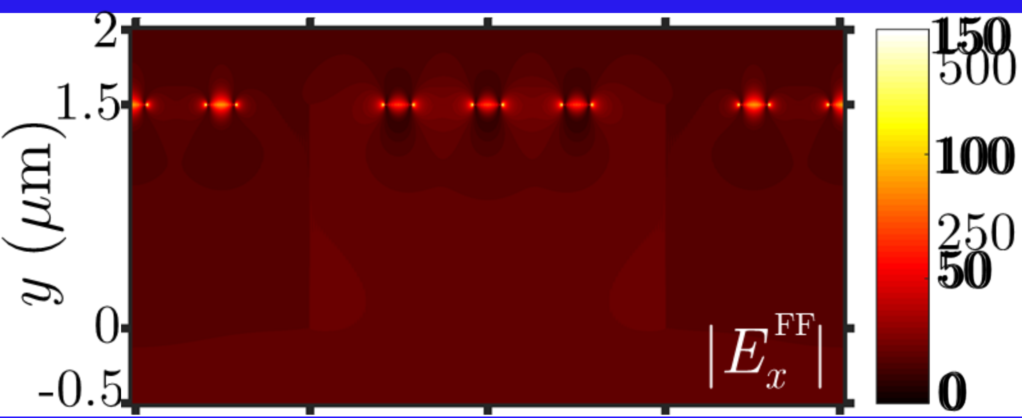
Λ=5.5 μm, h=1.5 μm
w=0.04Λ

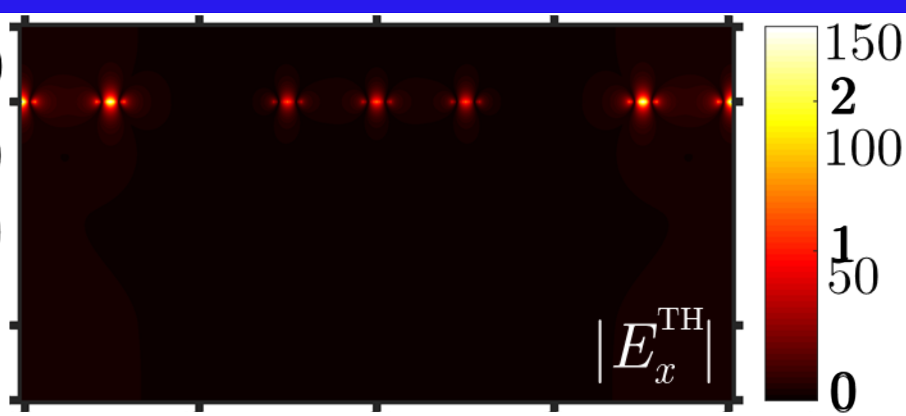
Linear and nonlinear spectrum



FF field

TH field





M. Weismann & N. C. Panoiu, Phys. Rev. B **94**, 035435 (2016)

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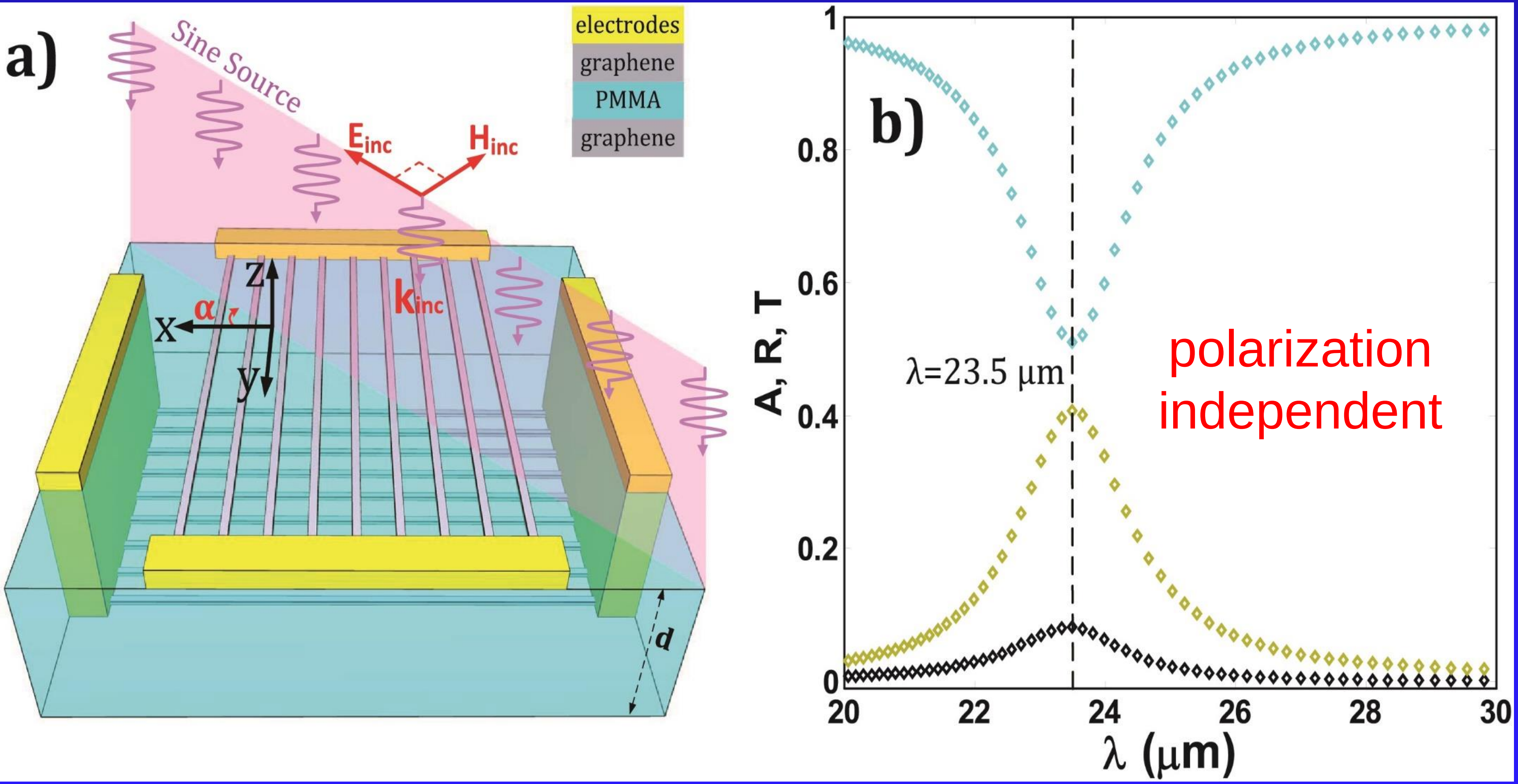
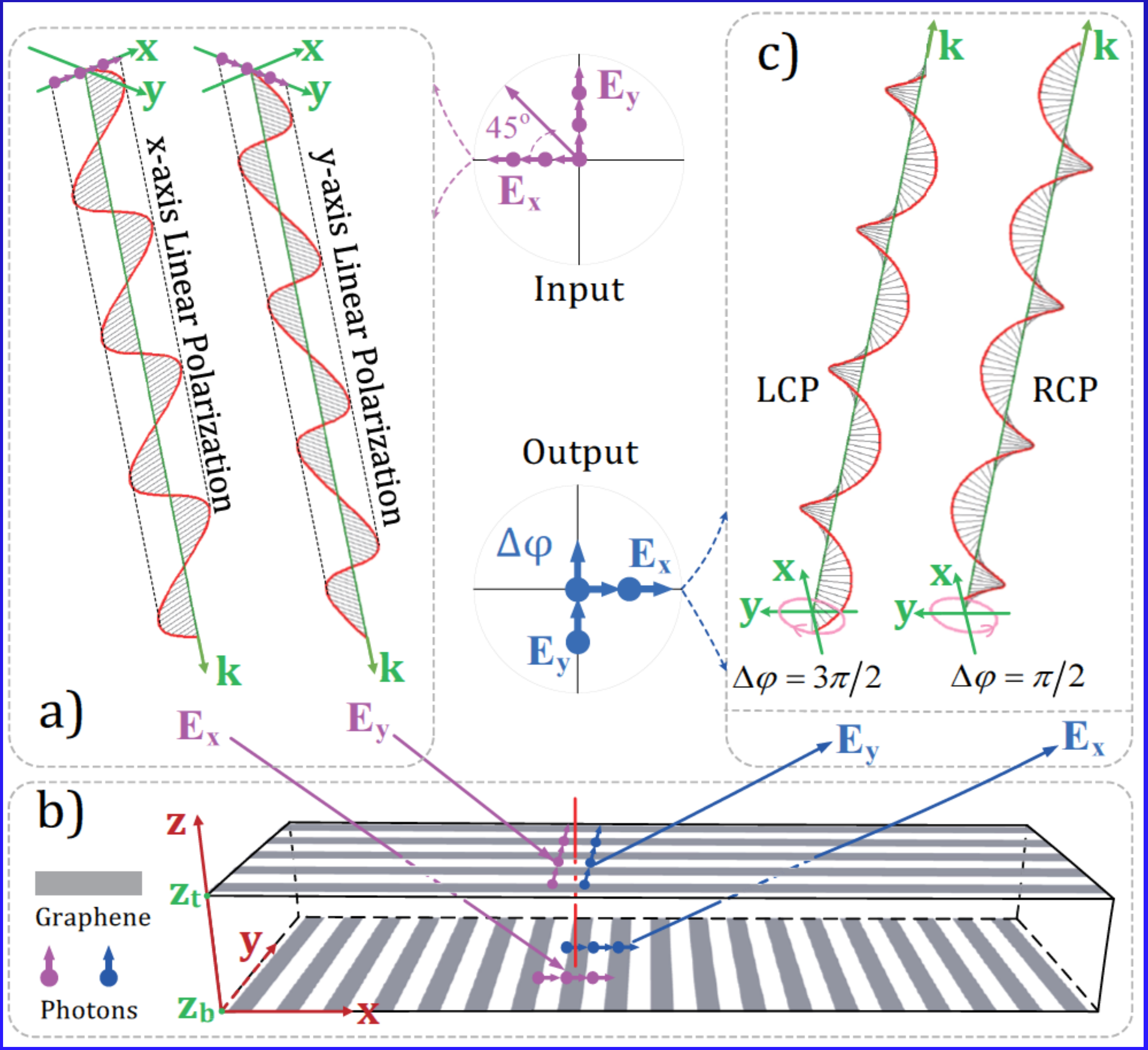
Nonlinear graphene polarization converters



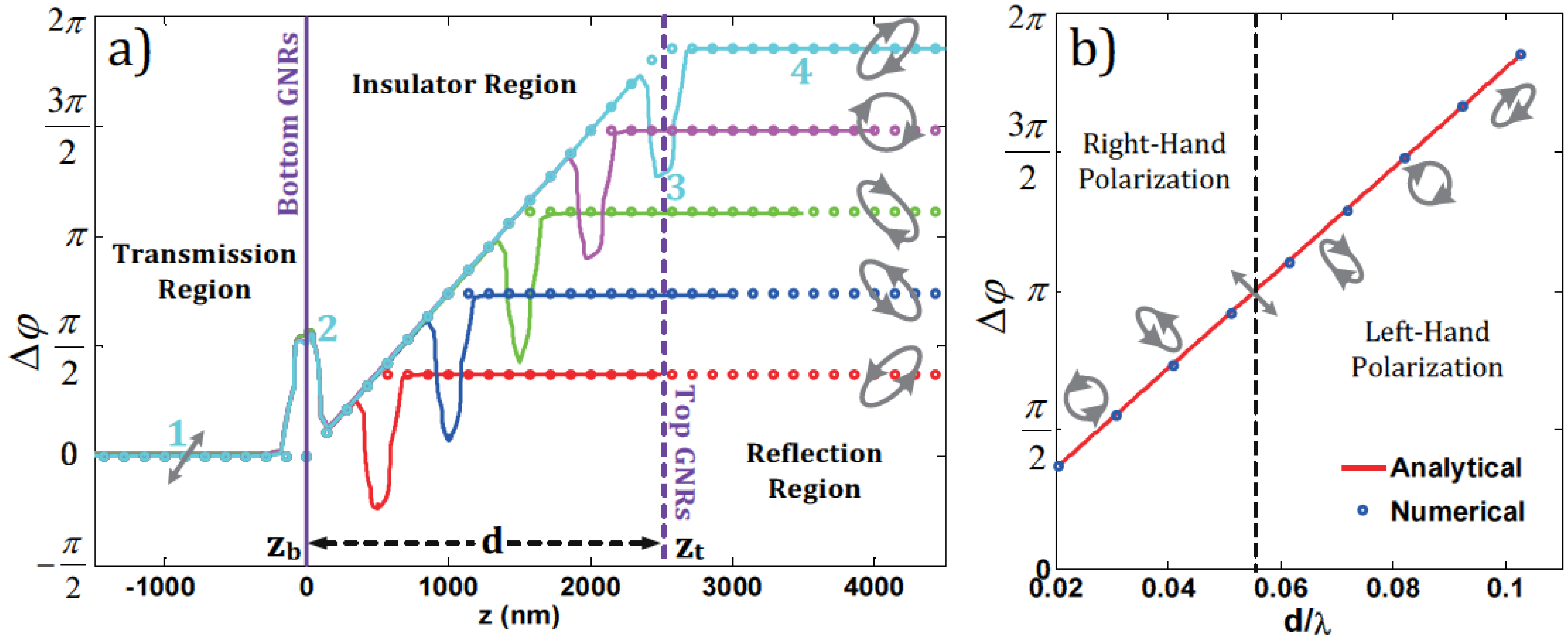
- ✓ Crossed-graphene gratings separated by a dielectric spacer;
- ✓ Polarization-independent spectra.

*J. W. You & N. C. Panoiu, Opt. Express **26**, 1882 (2018)*

How does it work?



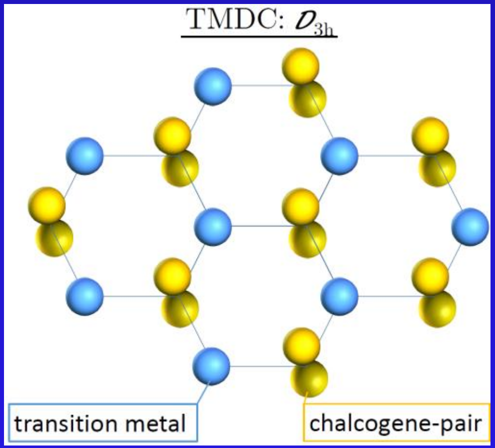
Nonlinear graphene polarization converters



$$\Delta\varphi = \frac{4\pi n d}{\Lambda} \qquad \Lambda = \frac{\lambda}{m} = \frac{\lambda}{3}$$ TH wavelength

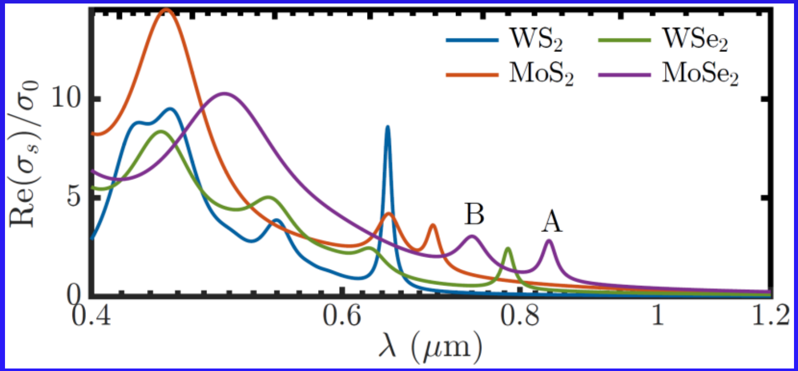
✓ Linearly polarized light can be converted to RCP or LCP light;
✓ Tunable operation wavelength.

Optical Properties of 2D Materials
TMDC Monolayers

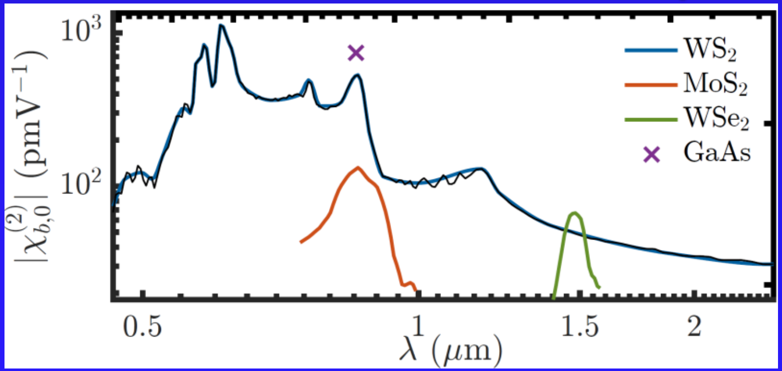


- ✓ transition metal (W, Mo) + two chalcogens (S, Se)
- ✓ direct band-gap semiconductors
- ✓ high absorption (> graphene)
- ✓ non-centrosymmetric: SHG allowed

Sheet conductance



Second-order conductivity

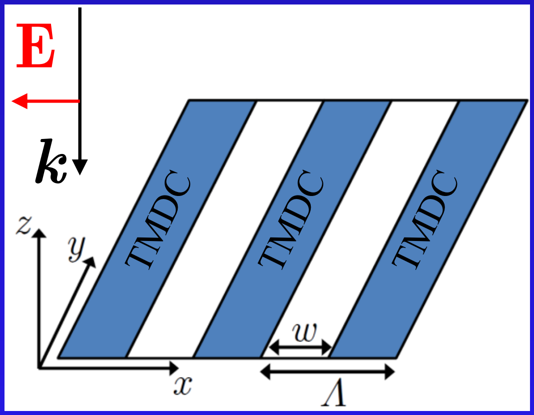


- ❑ Potential applications:

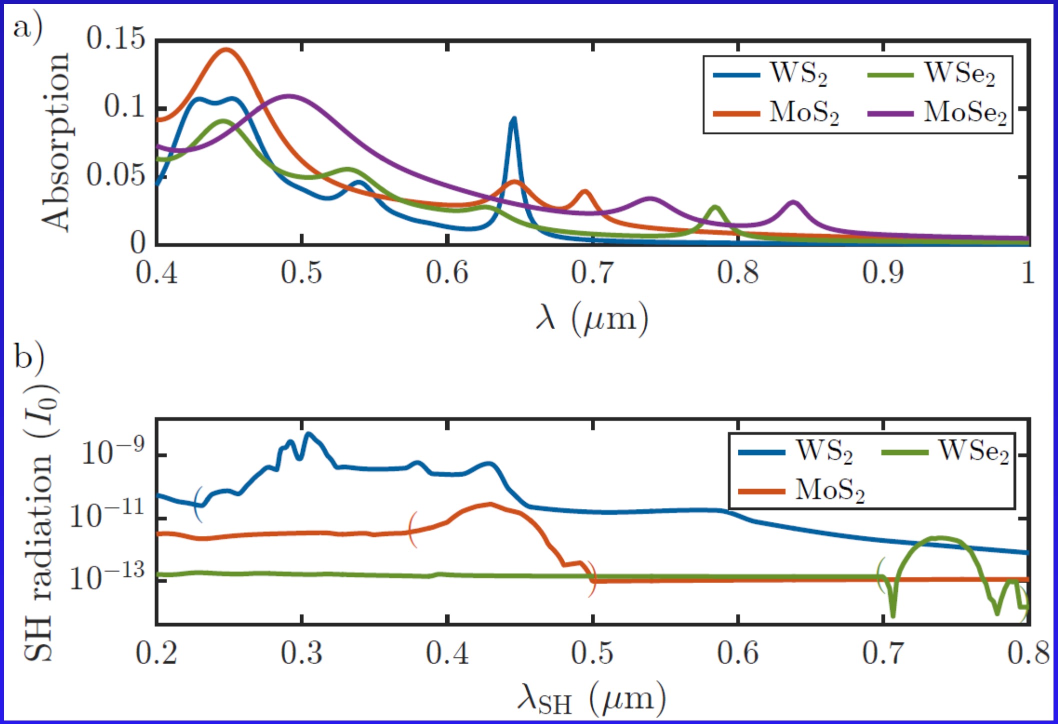
transistors, detectors, flexible electronics

Li, PRB 205422 (2014)
Janisch, Sci. Rep. 5530 (2014)
Li, Nano Lett. 3329 (2013)
Seyler, Nat. Nanotech. 404 (2015)

Diffraction in TMDC Gratings



$\Lambda = 100$ nm; $w = 90$ nm

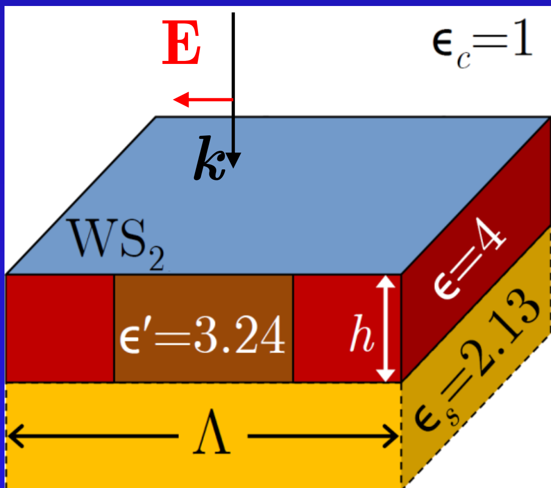


TMDCs: semiconductors ⇒ no plasmon resonances

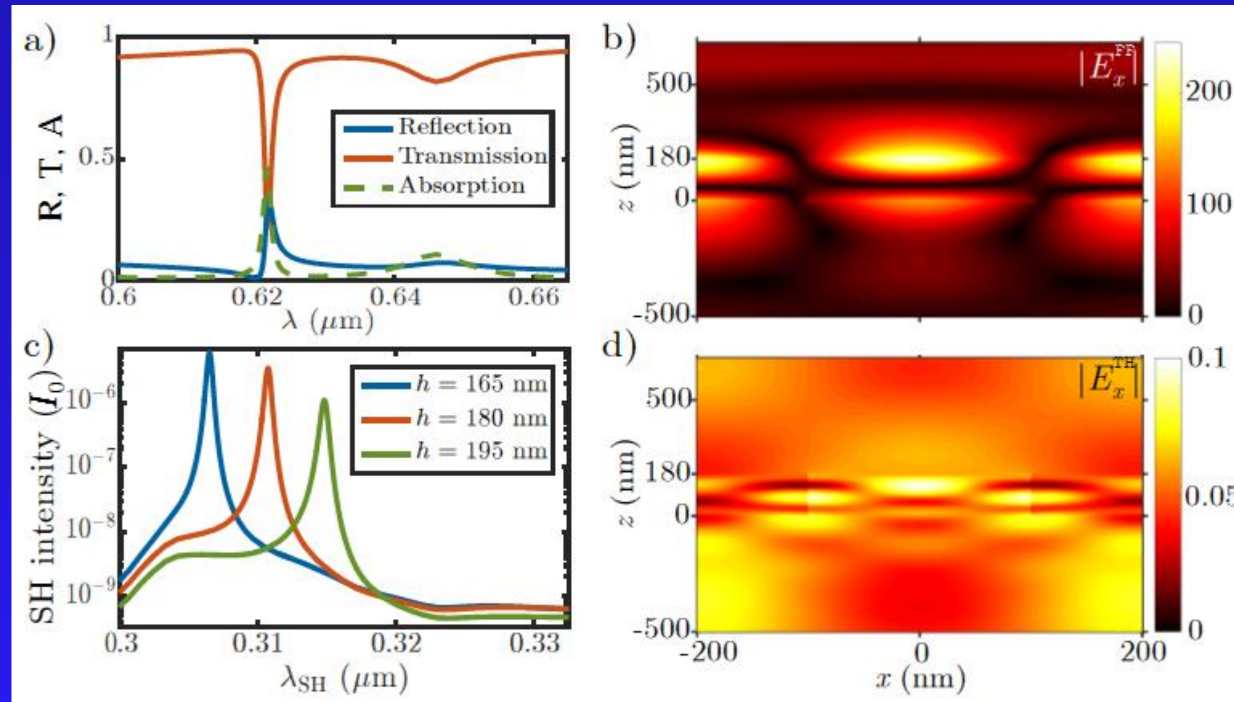
2D materials ⇒ no waveguide resonances

- ✓ Absorption is determined by exciton absorption peaks
- ✓ Radiated SH is determined by the magnitude of $\chi^{(2)}$

TMDC-Slab Waveguide Systems



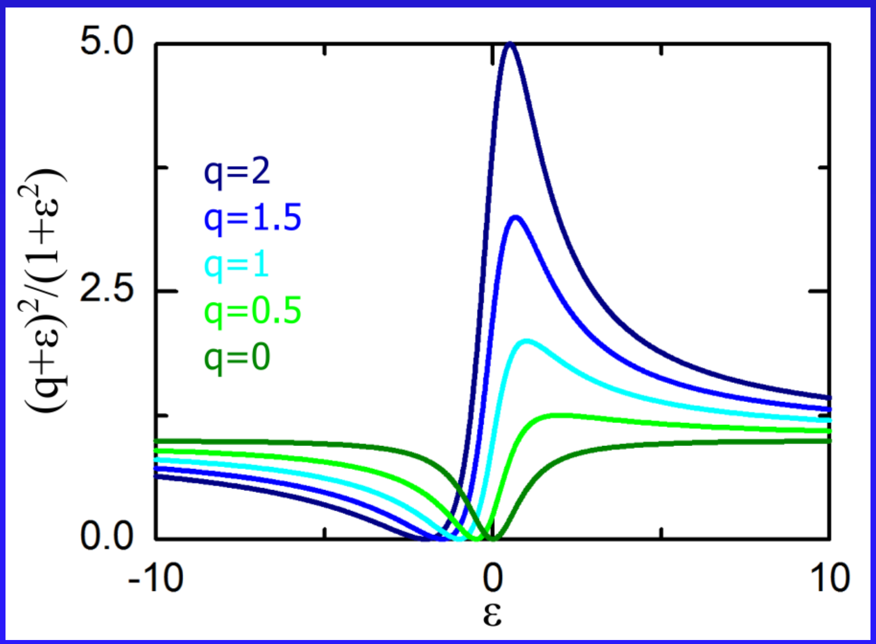
Textured slab waveguide + TMDC monolayer



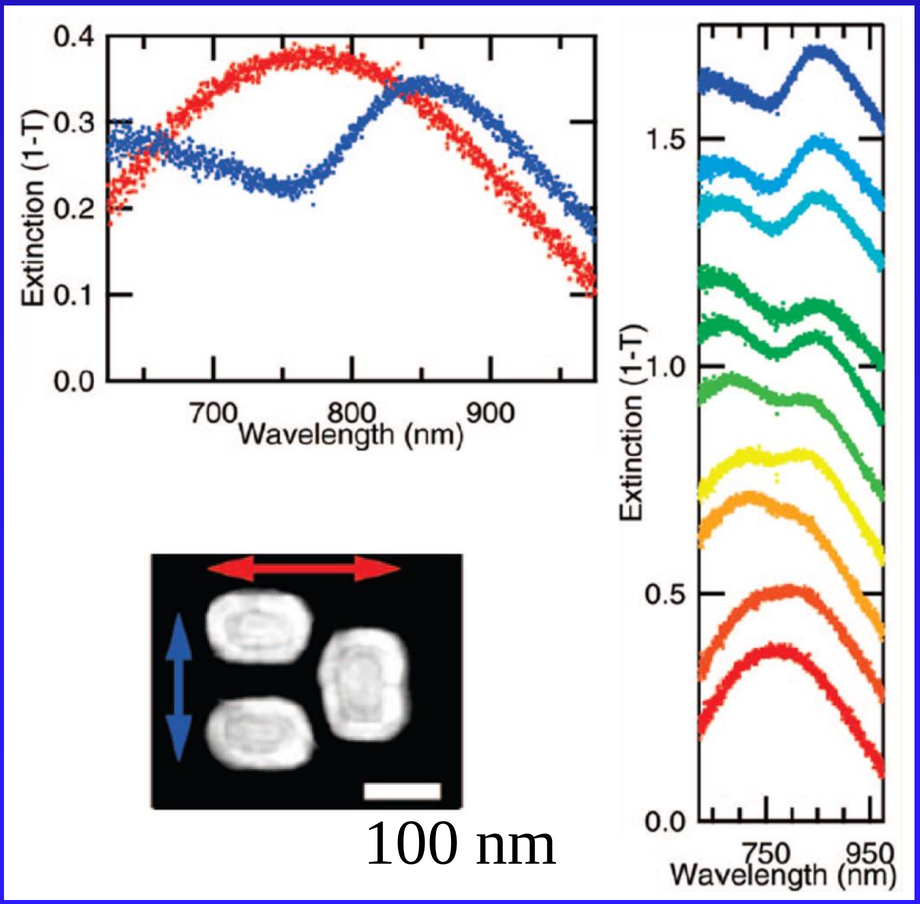
- Linear & nonlinear optical response
  - ✓ Sharp resonances in the reflectivity spectra
  - ✓ Excitation of waveguide modes
  - ✓ Translate to resonances in SH spectra
  - ✓ Strongly dependent on the system parameters

Fano Resonances in Plasmonic Systems

$$F(q,\varepsilon)=\frac{(q+\varepsilon)^2}{1+\varepsilon^2}$$



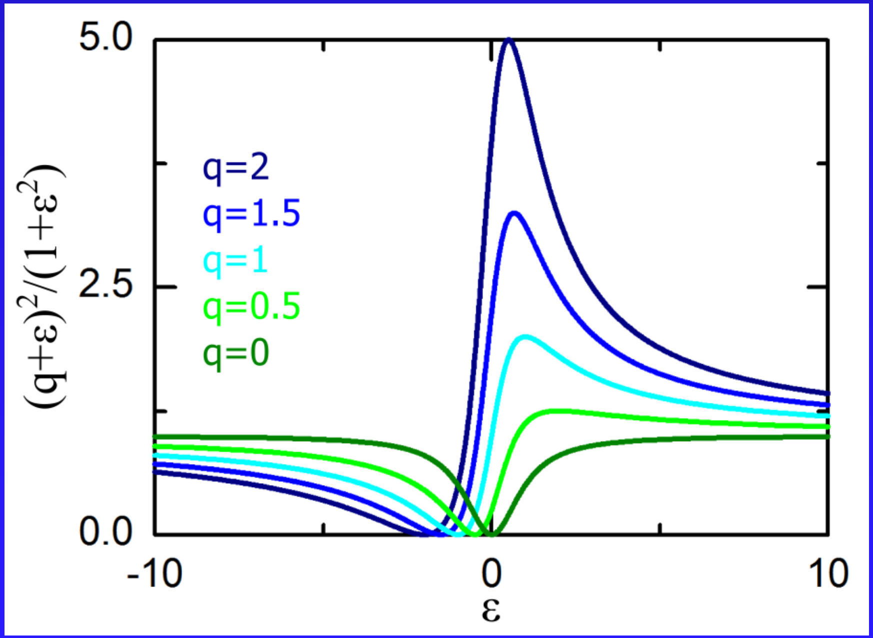
U. Fano *Phys. Rev.* **124,** 1866 (1961).

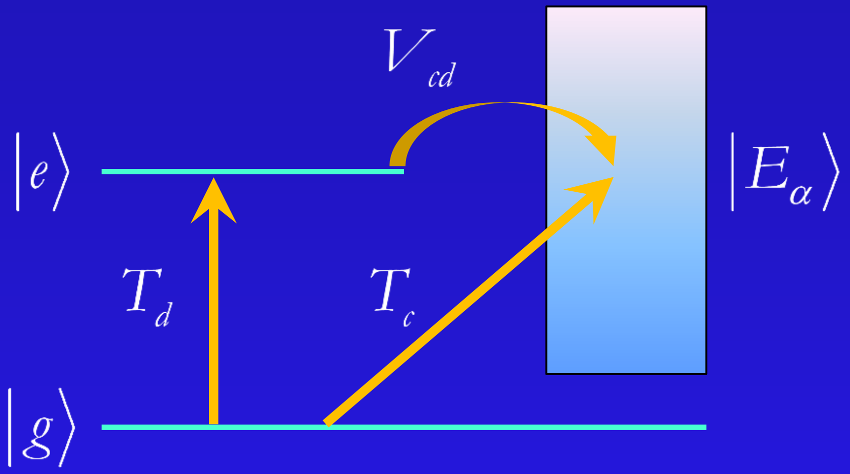


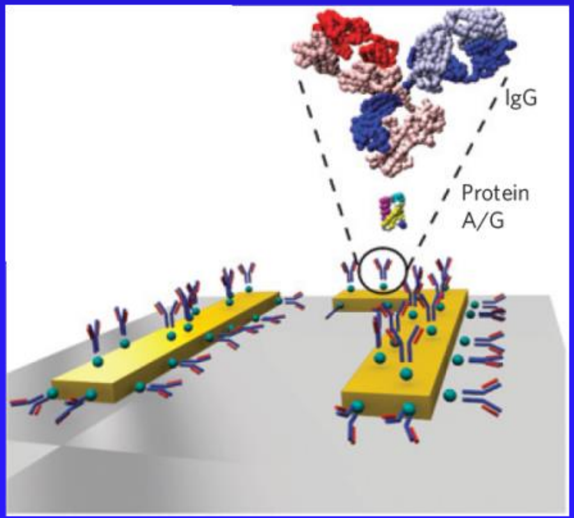
N. Verellen et al. *Nano Lett.* **9**, 1663 (2009).

Fano Resonance – Energy Levels

$$F(q,\varepsilon)=\frac{(q+\varepsilon)^2}{1+\varepsilon^2}$$



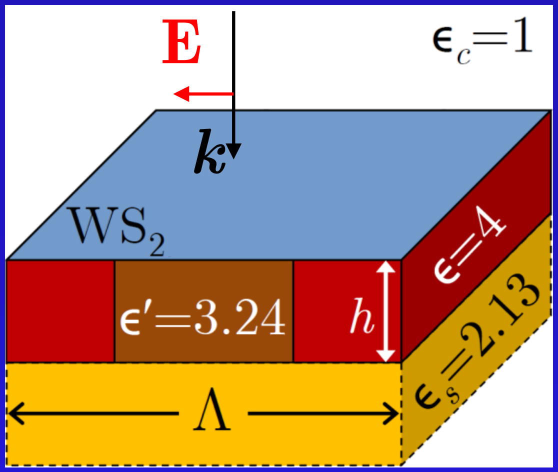




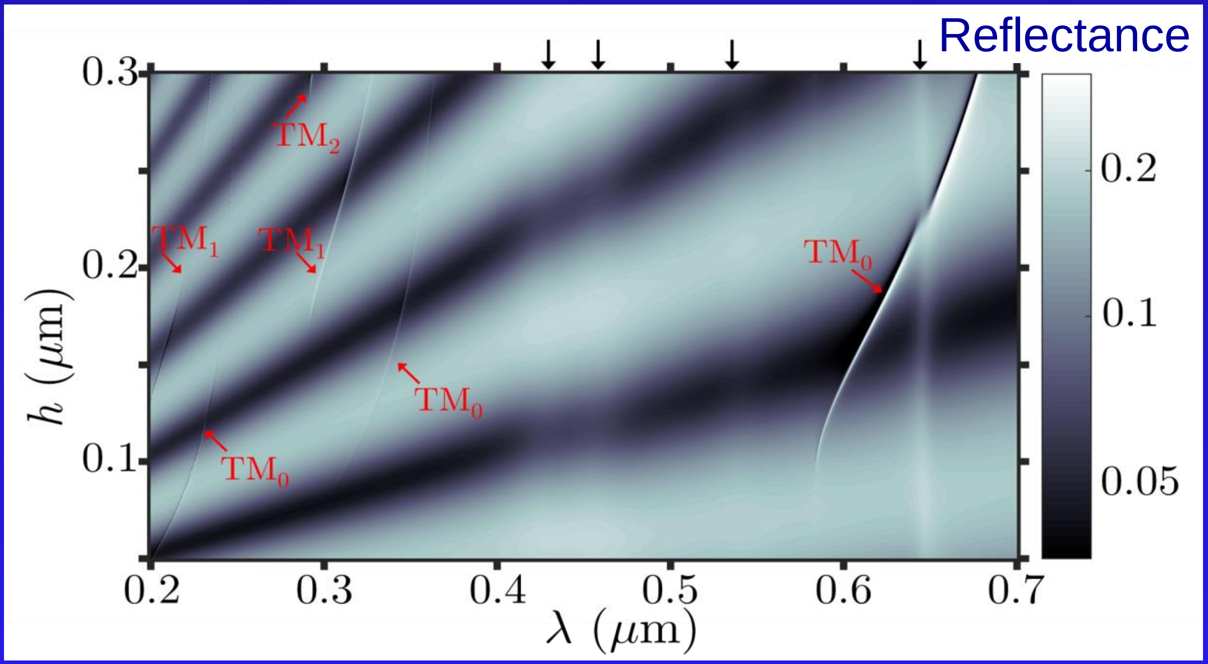
U. Fano Phys. Rev. ***124,*** *1866 (1961).*

Wu et al. Nature Materials ***11****, 69 (2012).*

TMDC-Slab Waveguide Systems (FF)



Textured slab waveguide + TMDC monolayer



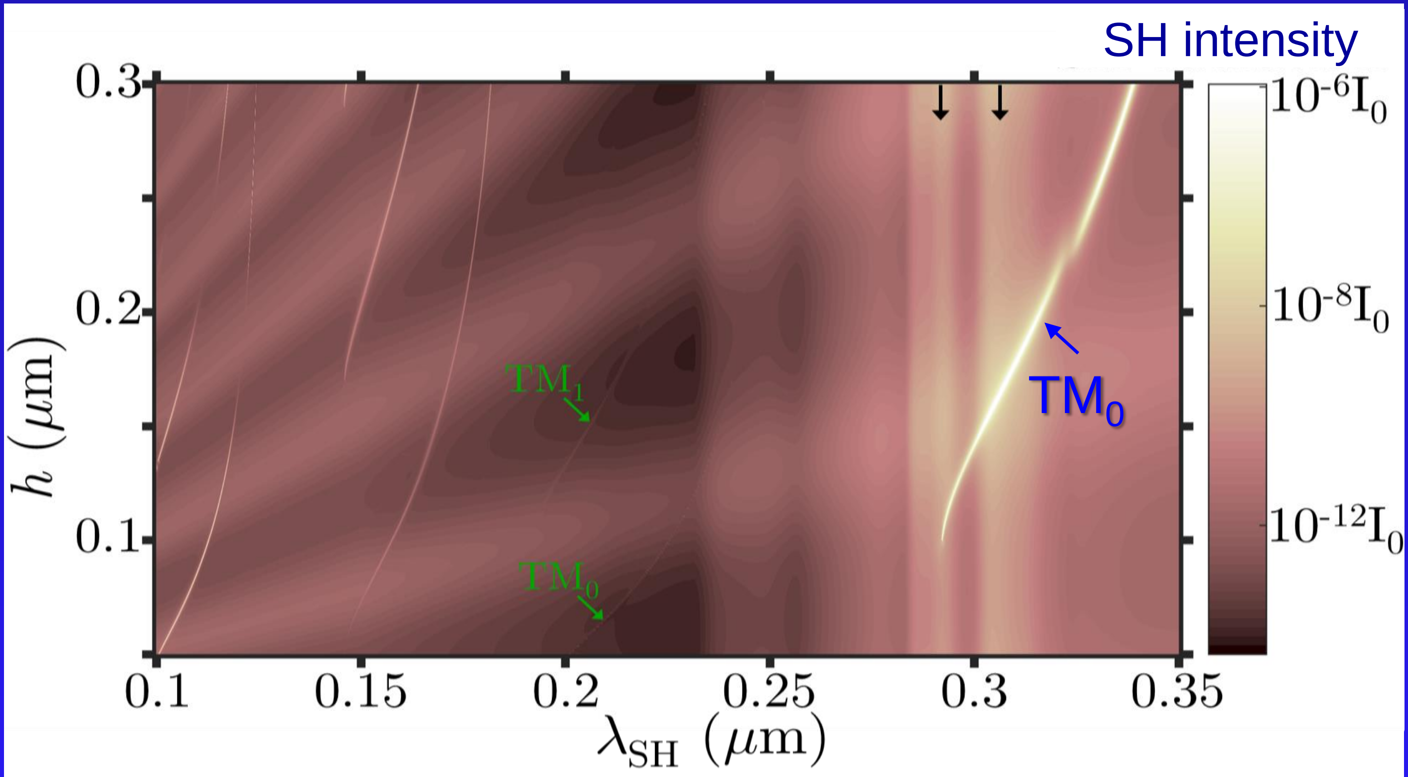
Spectrum determined by three mechanisms

- ✓ Fabry-Perot resonances (largest spectral variations)
- ✓ Absorption due to intrinsic material absorption (*h*-independent)
- ✓ Waveguide resonances (spectrally sharp)

Intricate resonance interplay

- ✓ Waveguide resonance (discrete) and FP (broad): Fano resonance
- ✓ Antisymmetric mode crossing of WGR with absorption band
- ✓ Additional Fano-resonance: exciton-absorption (discrete) and FP (broad)

TMDC-Slab Waveguide Systems (SH)



<u>SH radiation-spectrum determined by</u>

- ✓ Fabry-Perot resonances
- ✓ Dispersion of $\chi^{(2)}_{WS_2}$ (*h*-independent)
- ✓ Waveguide resonances: intrinsic + inherited

SH intensity enhanced by >6 orders of magnitude

*M. Weismann & N. C. Panoiu, Phys. Rev. B **94**, 035435 (2016)*

Conclusions

- Computational EM – key to modelling nonlinear optical effects at nanoscale.
- New approaches to engineering optical properties of nanostructured materials.
- Nonlinear optics in 2D materials.
- Active devices based on 2D materials.

Dr. M. Weismann

Dr. Jian Wei You

Thank you!