

A multimode quasi-normal mode framework for nonlinear harmonic generation with 2D materials

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Motivation and Objectives

Objectives

- ✓ Analysis and design of systems using the concept of QNMs
- ✓ Presentation of a multimode framework that incorporates **2D materials**
- ✓ Examination of **linear** and **nonlinear** responses (third-harmonic generation)
- ✓ Examination of periodic systems that support **higher diffraction orders**
- ✓ Validation of the framework with full-wave simulations

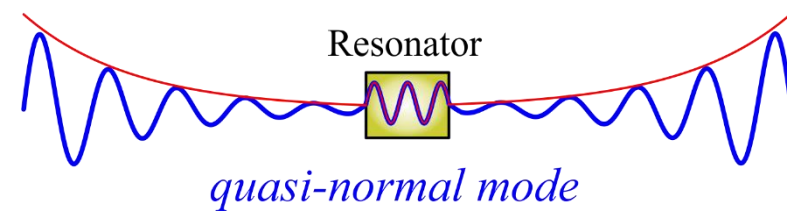
Motivation

- ✓ Physical **elegance** of modal analysis
- ✓ Rigorous capturing and better **understanding** of (modal) interference effects
- ✓ Better **intuition** on the analysis and design processes

QNMs framework with 2D materials

The concept of QNMs

- ✓ **Quasi-normal modes** (QNMs) are the natural (leaky) modes of **non-Hermitian** systems
- ✓ QNMs spatially diverge → their normalization is **non-trivial**
 - [Sauvan, *et al.*, Phys. Rev. Lett. 110, 237401, 2013]
 - [Doost, *et al.*, Phys. Rev. A 90, 013834, 2014]
 - [Lalanne, *et al.*, Laser Photon. Rev. 12, 1700113, 2018]
- ✓ QNMs can be used as an **orthonormal basis** (strictly complete inside the cavity) to find the spectral/temporal response of a system



$$\mathbf{E}(\omega) = \sum_m a_m(\omega) \tilde{\mathbf{E}}_m$$

- ✓ $a_m(\omega)$ are **appropriate coefficients** for the expansion, depending on the **excitation** and its interaction with each QNM

The concept of QNMs

- ✓ QNMs are typically retrieved with **modal** computational **techniques**
- ✓ We use an FEM version, modified to accommodate bulk **and** sheet materials with dispersion [Raman and Fan, *Phys. Rev. B* 83, 205131, 2011]

- ✓ An additional **auxiliary field** is used to include the **Drude dispersion** of 2D materials

$$\tilde{\mathbf{J}}_m = -\frac{\bar{\bar{\sigma}}_0}{(\tilde{\omega}_m - j\gamma)} \tilde{\mathbf{E}}_m$$

- ✓ Final system of equations to be solved through a **linearized eigenproblem** utilizing $\tilde{\mathbf{J}}_m$

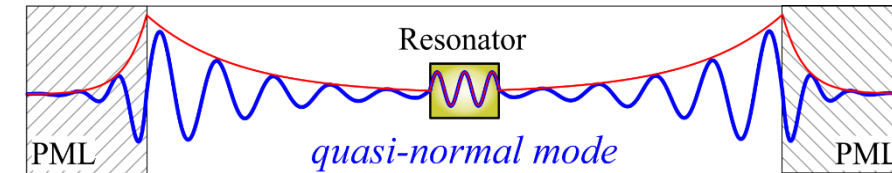
$$\begin{bmatrix} 0 & j\mu^{-1}\nabla\times & 0 \\ -j\varepsilon^{-1}\nabla\times & 0 & -\varepsilon^{-1} \\ 0 & -\bar{\bar{\sigma}}_0\delta_s & j\gamma \end{bmatrix} \begin{bmatrix} \tilde{\mathbf{H}}_m \\ \tilde{\mathbf{E}}_m \\ \tilde{\mathbf{J}}_m \end{bmatrix} = \tilde{\omega}_m \begin{bmatrix} \tilde{\mathbf{H}}_m \\ \tilde{\mathbf{E}}_m \\ \tilde{\mathbf{J}}_m \end{bmatrix}$$

Eigenvector
(on-resonance spatial field distribution)

Eigenvalue
(QNM complex resonance frequency)

The concept of QNMs

- ✓ QNMs should be **normalized** for the expansion
- ✓ This is a non-trivial task due to **spatial field divergence**
- ✓ The normalization is known for bulk materials. Integration should be performed **inside PML** as well, to accommodate spatial divergence
[Sauvan, et al., Phys. Rev. Lett. 110, 237401, 2013]
- ✓ We provide here the appropriate normalization for 2D materials



Traditional Version

$$\int_{V \cup V_{\text{PML}}} \left(\tilde{\mathbf{E}}_m \cdot \frac{\partial \{\omega \varepsilon(\omega)\}}{\partial \omega} \bigg|_{\omega=\tilde{\omega}_m} \tilde{\mathbf{E}}_m - \tilde{\mathbf{H}}_m \cdot \frac{\partial \{\omega \mu(\omega)\}}{\partial \omega} \bigg|_{\omega=\tilde{\omega}_m} \tilde{\mathbf{H}}_m \right) dV = 1$$

dispersive terms of bulk materials

Version for 2D Materials

$$\int_{V \cup V_{\text{PML}}} \left(\tilde{\mathbf{E}}_m \cdot \varepsilon \tilde{\mathbf{E}}_m - \tilde{\mathbf{H}}_m \cdot \mu \tilde{\mathbf{H}}_m - \tilde{\mathbf{E}}_m \cdot j \frac{\partial \bar{\sigma}_s(\omega)}{\partial \omega} \bigg|_{\omega=\tilde{\omega}_m} \tilde{\mathbf{E}}_m \delta_s \right) dV = 1$$

dispersive (surface) conductivity

[Christopoulos, Kriezis, and Tsilipakos, Phys. Rev. B 107, 035413, 2023]

The concept of QNMs

- ✓ In the FEM and in the QNMs expansion framework, **respecting** the true nature of **2D materials** as **infinitesimally thin layers** is crucial
- ✓ 2D materials are typically described electromagnetically by their (complex and dispersive) **surface conductivity** tensor. A surface current is induced: $\mathbf{J}_s = \bar{\bar{\sigma}}(\omega)\mathbf{E}$
- ✓ We take care to incorporate this term in the QNMs formulation

Traditional Version

$$a_m(\omega) = -\frac{\tilde{\omega}_m}{\tilde{\omega}_m - \omega} \int_V [\varepsilon(\tilde{\omega}_m) - \varepsilon_b] \tilde{\mathbf{E}}_m \cdot \mathbf{E}_{\text{inc}} dV \\ + \int_V [\varepsilon_b - \varepsilon_\infty] \tilde{\mathbf{E}}_m \cdot \mathbf{E}_{\text{inc}} dV$$

Version for 2D Materials

$$a_m(\omega) = -\frac{1}{\tilde{\omega}_m - \omega} \int_V \frac{\bar{\bar{\sigma}}_0}{(\tilde{\omega}_m - j\gamma)} \tilde{\mathbf{E}}_m \cdot \mathbf{E}_{\text{inc}} dV$$

QNMs and spurious modes

Incident field

Step 4: QNM nonlinear expansion w/ 2D materials (1/2)

The concept of QNMs

- ✓ QNMs expansion can be applied to describe **nonlinear interactions** of **2D materials**
- ✓ We focus on **third-harmonic generation**, induced by $\bar{\sigma}_3$ of graphene
- ✓ A field @ ω induces a surface current @ 3ω (new source) through a third-order process;
In turn, the current produces a field @ 3ω which is coupled to a radiative channel (leaky mode)

$$a_m(3\omega) = -\frac{j}{\tilde{\omega}_m - 3\omega} \int_V \tilde{\mathbf{E}}_m \cdot \mathbf{J}_{s,\text{NL}}^{(3\omega)} \delta_s dV$$

↑ Dominant contribution @ 3ω
↑ New source term @ 3ω

$$\mathbf{J}_{s,\text{NL}}^{(3\omega)} = \frac{\sigma_3}{4} (\mathbf{E}_{t,\parallel}^{(\omega)} \cdot \mathbf{E}_{t,\parallel}^{(\omega)}) \mathbf{E}_{t,\parallel}^{(\omega)}$$

← Total field @ ω

[Christopoulos, Kriezis, and Tsilipakos, *Phys. Rev. B* 107, 035413, 2023]

The concept of QNMs

✓ Calculation steps

1. Find all QNMs (around ω and 3ω)

2. Calculate the expansion coefficients @ ω : $a_m(\omega) = -\frac{1}{\tilde{\omega}_m - \omega} \int_V \frac{\bar{\sigma}_0}{(\tilde{\omega}_m - j\gamma)} \tilde{\mathbf{E}}_m \cdot \mathbf{E}_{\text{inc}} dV$

3. Calculate scattered field @ ω : $\mathbf{E}_{\text{sct}}(\omega) = \sum_m a_m(\omega) \tilde{\mathbf{E}}_m$ QNMs are the same but the expansion coefficients are not

4. Calculate total field @ ω : $\mathbf{E}_t(\omega) = \mathbf{E}_{\text{inc}} + \mathbf{E}_r + \mathbf{E}_{\text{sct}}$ Total field consists of the incident, reflected from the background, and scattered waves!

5. Calculate the induced surface current @ 3ω : $\mathbf{J}_{s,\text{NL}}^{(3\omega)} = \frac{\sigma_3}{4} (\mathbf{E}_{t,\parallel}^{(\omega)} \cdot \mathbf{E}_{t,\parallel}^{(\omega)}) \mathbf{E}_{t,\parallel}^{(\omega)}$

6. Calculate the expansion coefficients @ 3ω : $a_m(3\omega) = -\frac{j}{\tilde{\omega}_m - 3\omega} \int_V \tilde{\mathbf{E}}_m \cdot \mathbf{J}_{s,\text{NL}}^{(3\omega)} \delta_s dV$

7. Calculate scattered field @ 3ω : $\mathbf{E}_{\text{sct}}(3\omega) = \sum_m a_m(3\omega) \tilde{\mathbf{E}}_m$

A simple system

Single graphene strip scatterer

Resonant System

- ✓ Single graphene strip on a glass substrate
- ✓ Normal illumination with a TM plane wave to excite graphene surface plasmons → Fabry-Perot resonances along x axis

✓ Bright modes:

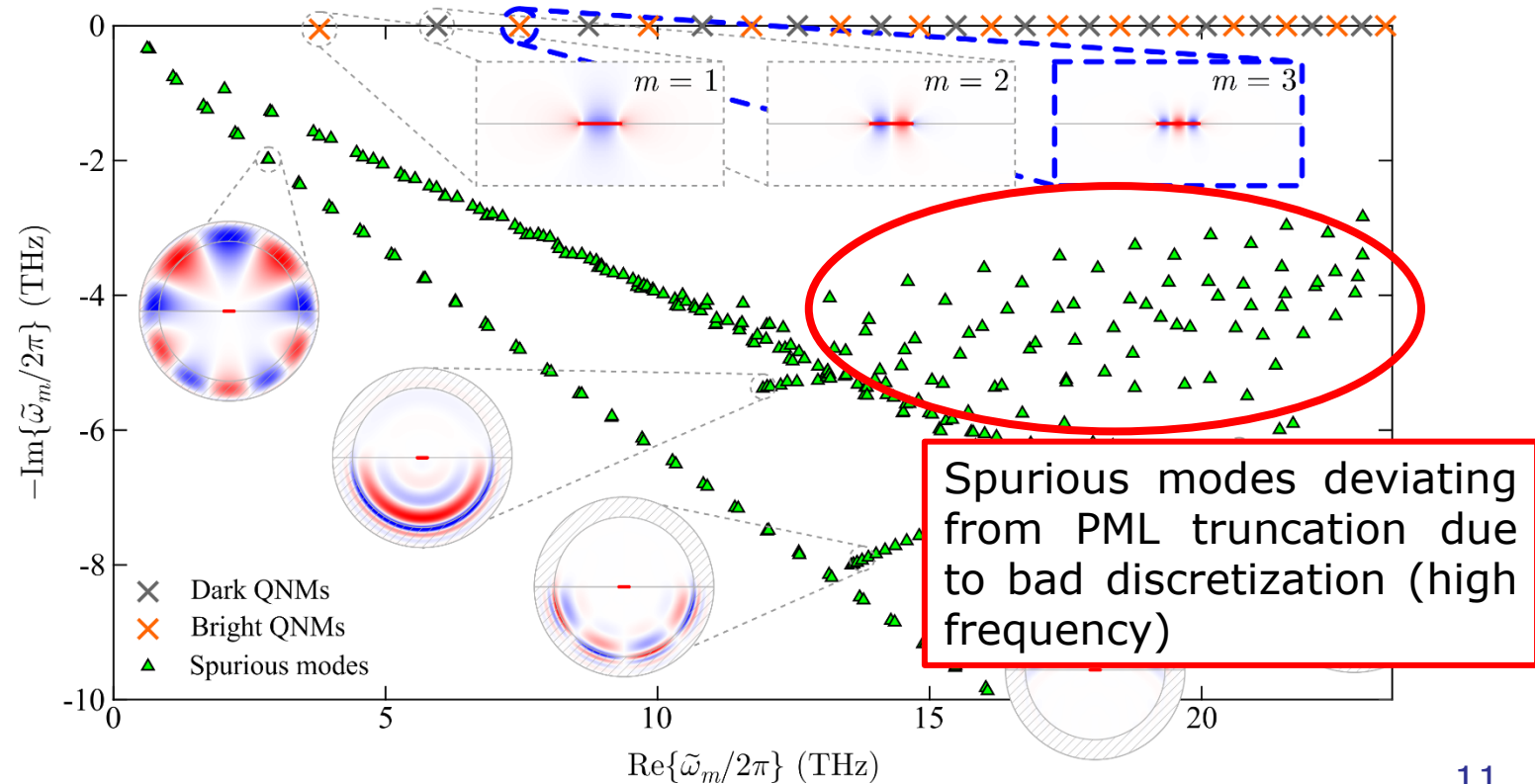
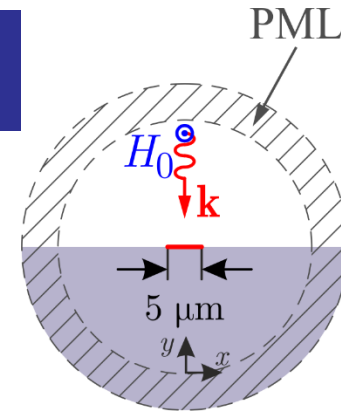
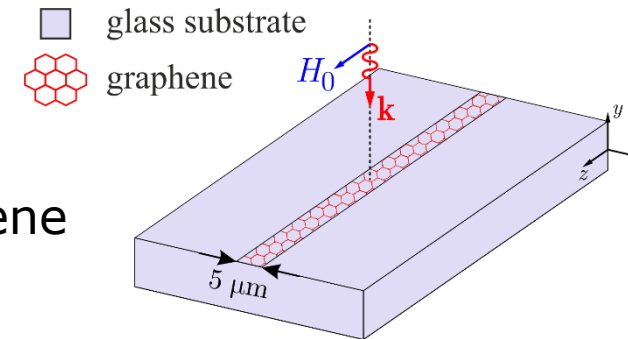
- ✓ m is odd (symmetric distribution)
- ✓ Q_{rad} finite
- ✓ Can be excited by a normally incident plane wave

❖ Dark modes:

- ❖ m is even (antisymmetric distribution)
- ❖ $Q_{\text{rad}} \rightarrow \infty$
- ❖ Cannot be excited by a normally incident plane wave

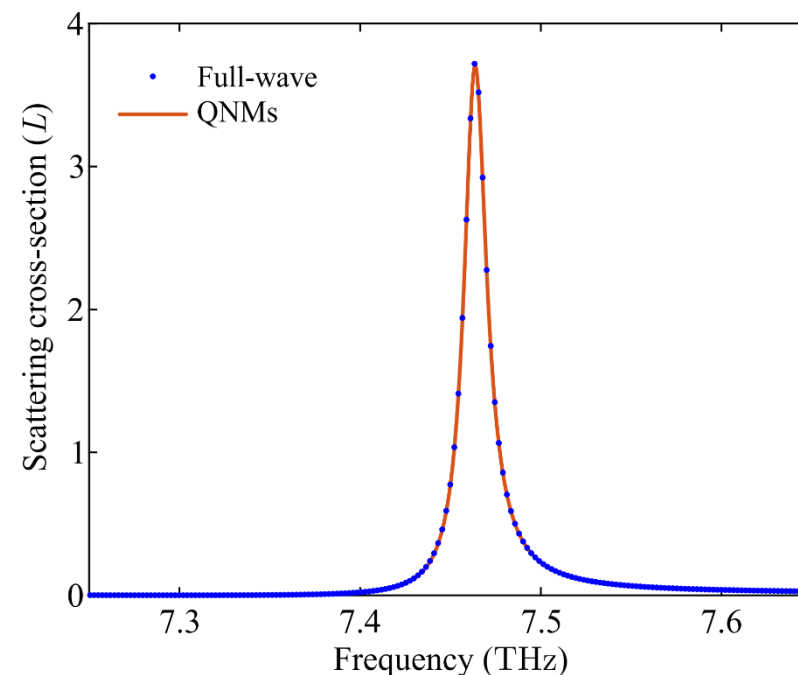
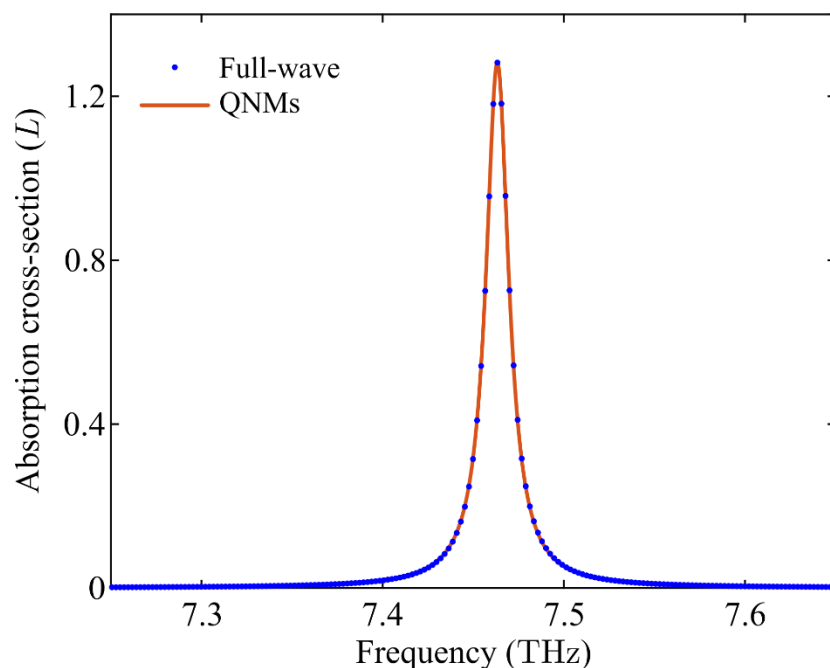
✓ Spurious (PML) modes:

- ✓ Necessary for the expansion



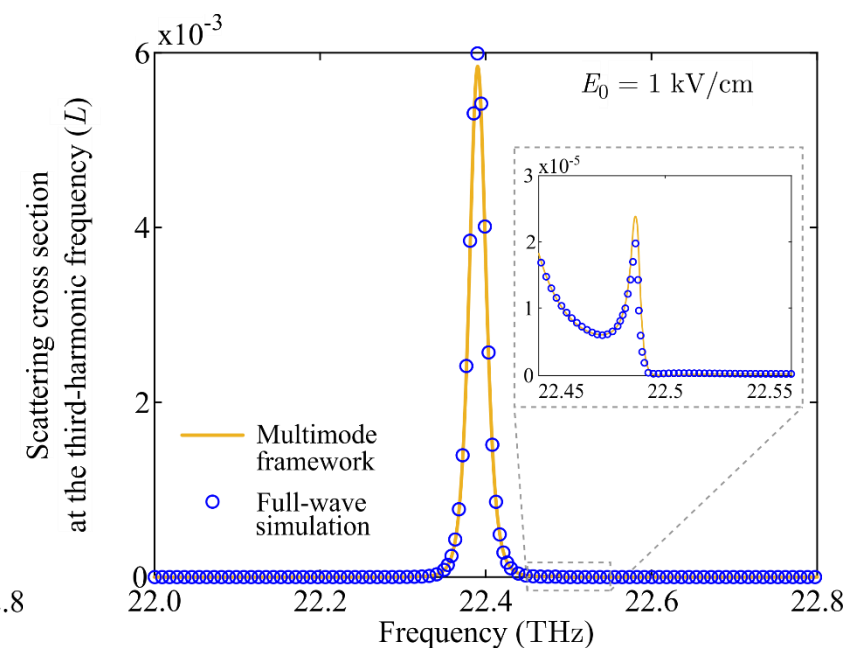
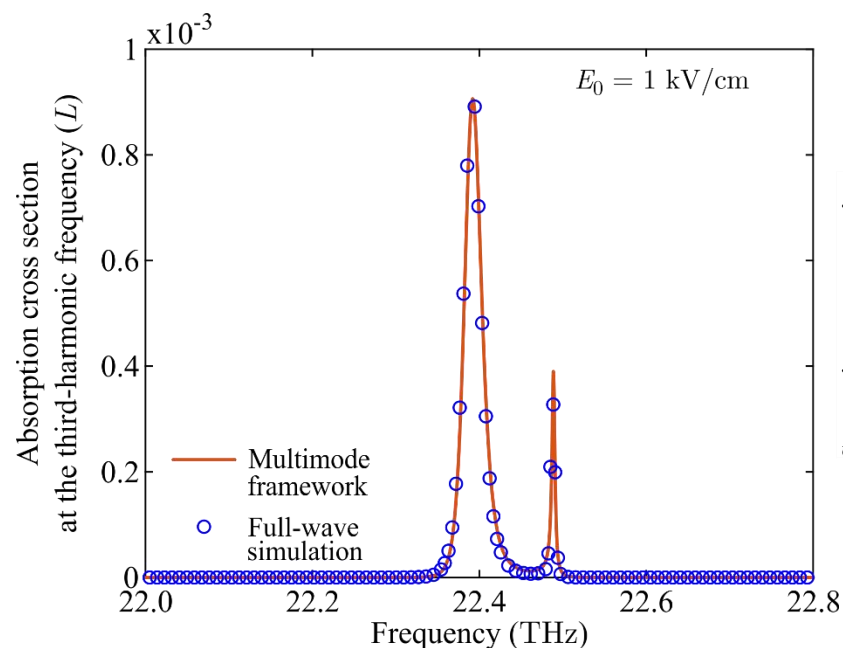
Linear Absorption and Scattering Cross-Sections

- ✓ Comparison with full-wave linear simulations (around ω) gives **excellent agreement**
- ✓ Even **asymmetries** in scattering cross-section are accurately captured
- ✓ **Correct QNMs normalization** considering **graphene's surface contribution** is crucial to get accurate results



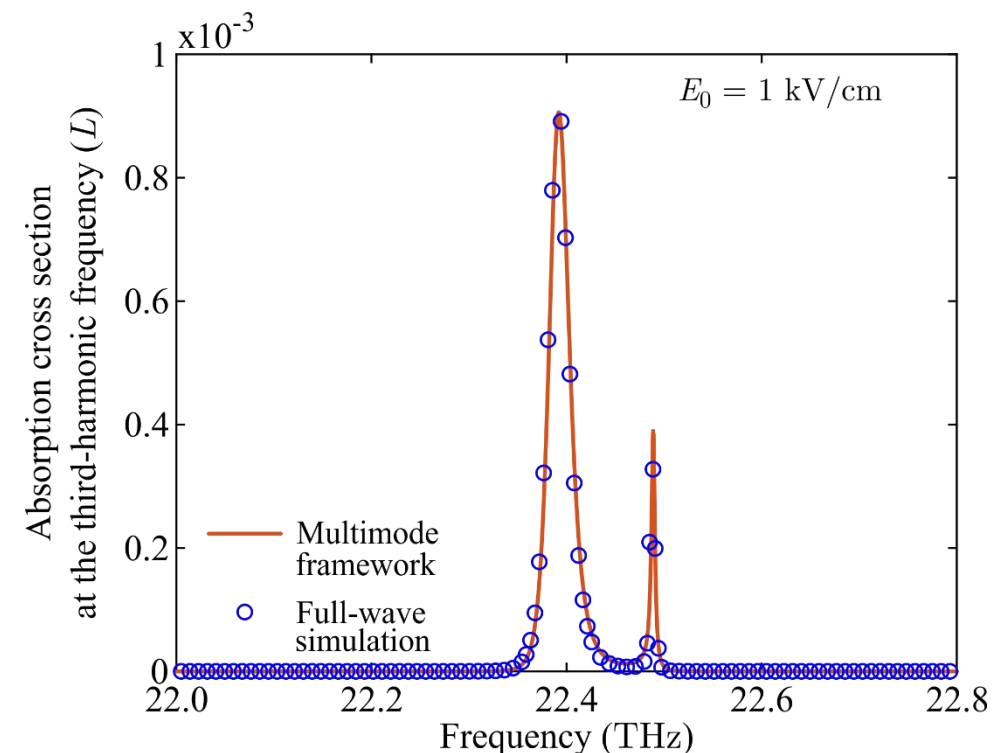
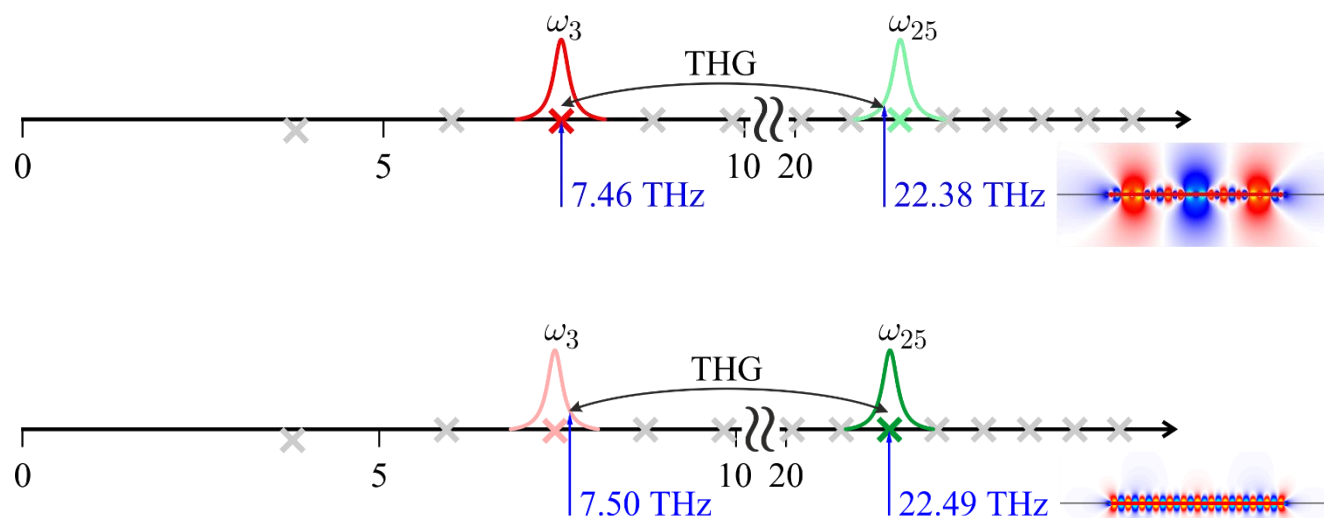
Nonlinear Absorption and Scattering Cross-Sections @ 3ω

- ✓ Using QNMs and the **nonlinear expansion, third-harmonic generation** can be efficiently and accurately described
- ✓ Comparison with full-wave nonlinear simulations (around 3ω) gives **excellent agreement**
- ✓ The multimode framework can reveal **interesting physics** of the THG process (two peaks, next slide)
- ✓ Very good agreement even in **asymmetric non-Lorentzian lineshapes** or very weak peaks/interactions



Nonlinear Absorption and Scattering Cross-Sections @ 3ω

- ✓ The **multimode** framework can reveal **interesting physics** of the THG process (non-trivial mode interaction)
- ✓ The first peak emerges at the third harmonic of the fundamental frequency ($3\omega_3 = 22.38$ THz)
- ✓ The second peak emerges exactly at the resonance frequency of the QNM lying closest to 3ω ($\omega_{25} = 22.49$ THz)

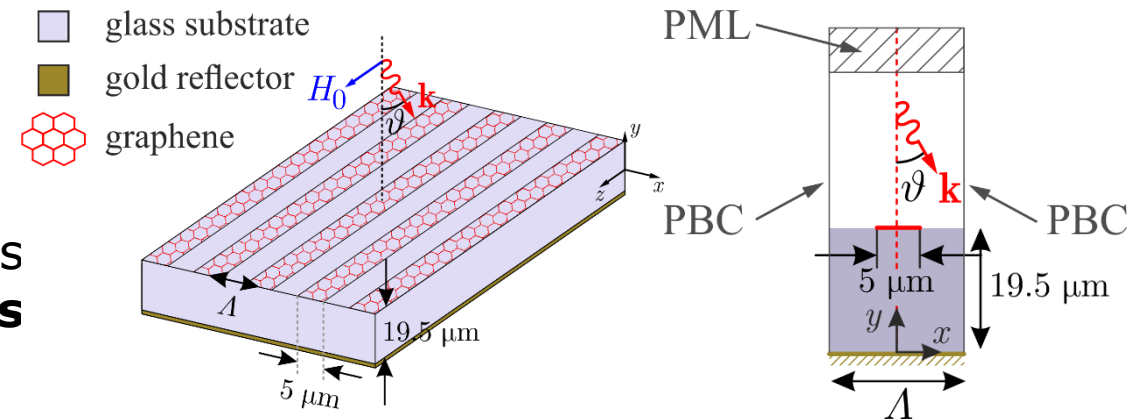


A more practical system

Graphene strip metasurface

Resonant System

- ✓ Expansion of the previous graphene strip in a **reflective metasurface**
- ✓ Graphene allows for **deeply subwavelength** cavities due to tightly confined **graphene surface plasmons** (GPSs), i.e., large parallel wavevector
- ✓ Deeply subwavelength cavities \rightarrow lattice remains **subwavelength** even in the **third-harmonic frequency**
- ✓ QNMs **can** also describe metasurfaces with larger periodicity (gratings) where **higher diffraction orders** are excited

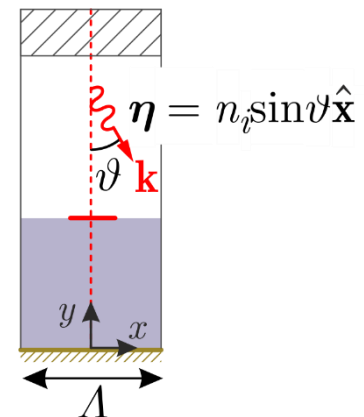


QNM framework modifications for periodic systems

- ✓ **Floquet** periodic boundary condition **cannot** be used in **modal calculations** because the knowledge of the wavevector in the periodicity dimension is required and $\tilde{\mathbf{k}}_m \propto \tilde{\omega}_m$
- ✓ The wave equation that is to be solved with the FEM is **modified** to account for this fact
- ✓ Fields are expressed with respect to a spatial periodic envelope $\mathbf{E} = \mathbf{e} \exp\{-j\mathbf{k}_F \cdot \mathbf{r}\} = \mathbf{e} \exp\{-jk_0 \mathbf{\eta} \cdot \mathbf{r}\}$
- ✓ The incorporation of $\mathbf{\eta}$ vector that depends only on the incident angle allows for a **new** and **correct** wave equation to calculate the QNMs

$$\nabla \times \mu^{-1} \nabla \times \tilde{\mathbf{e}}_m - j \frac{\tilde{\omega}_m}{c_0} \nabla \times \mu^{-1} (\mathbf{\eta} \times \tilde{\mathbf{e}}_m) - j \frac{\tilde{\omega}_m}{c_0} \mathbf{\eta} \times \mu^{-1} \nabla \times \tilde{\mathbf{e}}_m - \frac{\tilde{\omega}_m^2}{c_0^2} \mathbf{\eta} \times \mu^{-1} \mathbf{\eta} \times \tilde{\mathbf{e}}_m - \tilde{\omega}_m^2 \epsilon \tilde{\mathbf{e}}_m - \tilde{\omega}_m \tilde{\mathbf{j}}_m = 0$$

new terms disentangling the eigenvalue and the angle of incidence contributions on the wave-vector



QNM framework modifications for periodic systems

- ✓ The envelope modification introduced above modifies the QNMs eigenvalue equation

$$\begin{bmatrix} 0 & j\mu^{-1}\nabla\times & 0 \\ -j\varepsilon^{-1}\nabla\times & 0 & -\varepsilon^{-1} \\ 0 & -\bar{\sigma}_0\delta_s & j\gamma \end{bmatrix} \begin{bmatrix} \tilde{\mathbf{h}}_m \\ \tilde{\mathbf{e}}_m \\ \tilde{\mathbf{j}}_m \end{bmatrix} = \tilde{\omega}_m \overset{\hat{\mathcal{M}}}{\begin{bmatrix} 1 & -(c_0\mu)^{-1}\boldsymbol{\eta}\times & 0 \\ (c_0\varepsilon)^{-1}\boldsymbol{\eta}\times & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}} \begin{bmatrix} \tilde{\mathbf{h}}_m \\ \tilde{\mathbf{e}}_m \\ \tilde{\mathbf{j}}_m \end{bmatrix}$$

- ✓ It also modifies the expansion coefficients and the QNM normalization term – only **envelope quantities** are involved

$$a_m(\omega) = -\frac{1}{\tilde{\omega}_m - \omega} \int_V \frac{\bar{\sigma}_0}{(\tilde{\omega}_m - j\gamma)} \tilde{\mathbf{e}}_{-m} \cdot \mathbf{e}_{\text{inc}} dV$$

$$\int_V \left[\tilde{\mathbf{e}}_{-m} \cdot \varepsilon \tilde{\mathbf{e}}_m - \tilde{\mathbf{h}}_{-m} \cdot \mu \tilde{\mathbf{h}}_m - \tilde{\mathbf{e}}_{-m} \cdot j \frac{\partial \bar{\sigma}_s(\omega)}{\partial \omega} \tilde{\mathbf{e}}_m \delta_s - \boldsymbol{\eta} \cdot \frac{1}{c_0} \left(\tilde{\mathbf{h}}_{-m} \times \tilde{\mathbf{e}}_m + \tilde{\mathbf{e}}_{-m} \times \tilde{\mathbf{h}}_m \right) \right] dV = 1$$

extra normalization term

QNM framework modifications for periodic systems

- ✓ Expansion coefficients are also modified in the nonlinear system

$$a_m(3\omega) = -\frac{j}{\tilde{\omega}_m - 3\omega} \int_V \left[\tilde{\mathbf{e}}_{-m} \cdot \mathbf{j}_{s,\text{NL}}^{(3\omega)} \delta_s - \boldsymbol{\eta} \cdot (\tilde{\mathbf{h}}_{-m} \times \mathbf{j}_{s,\text{NL}}^{(3\omega)} \delta_s) \right] dV$$

Expected envelope term

New additional term due to periodicity

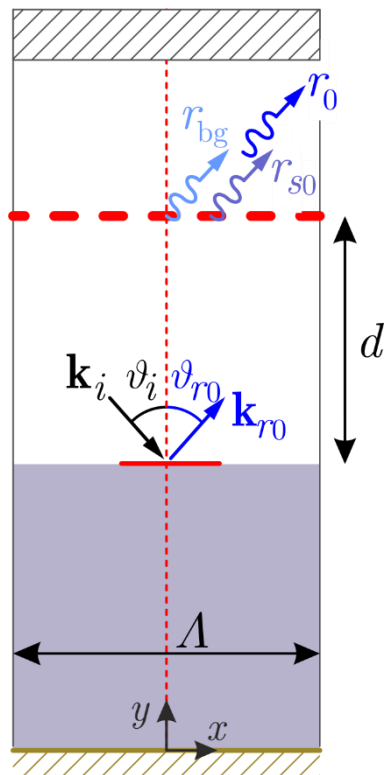
Non-zero when there is a surface current component perpendicular to the periodicity vector $\boldsymbol{\eta}$

Nonlinear current envelope

$$\mathbf{j}_{s,\text{NL}}^{(3\omega)} = \frac{\sigma_3}{4} (\mathbf{e}_{t,\parallel}^{(\omega)} \cdot \mathbf{e}_{t,\parallel}^{(\omega)}) \mathbf{e}_{t,\parallel}^{(\omega)}$$

$$\mathbf{e}_t^{(\omega)} = \mathbf{E}_t^{(\omega)} e^{jk_0 \boldsymbol{\eta} \cdot \mathbf{r}}$$

Linear reflection (specular)



$$\Lambda = 10 \mu\text{m}$$

$$\lambda_0 \sim 40 \mu\text{m}$$

- ✓ Specular reflection (Snell's law), $\vartheta_i = \vartheta_{r0}$ and $k_{i,y} = -k_{r0,y}$

- ✓ Reflection coefficient:
$$r_0 = r_{\text{bg}} + \frac{1}{\Lambda H_0} e^{jk_{r0,y}d} \int_{-\Lambda/2}^{\Lambda/2} \mathbf{H}_{\text{sct}}(x, d) e^{jk_{r0,x}x} dx$$

background reflection without graphene
 (analytical calculation)

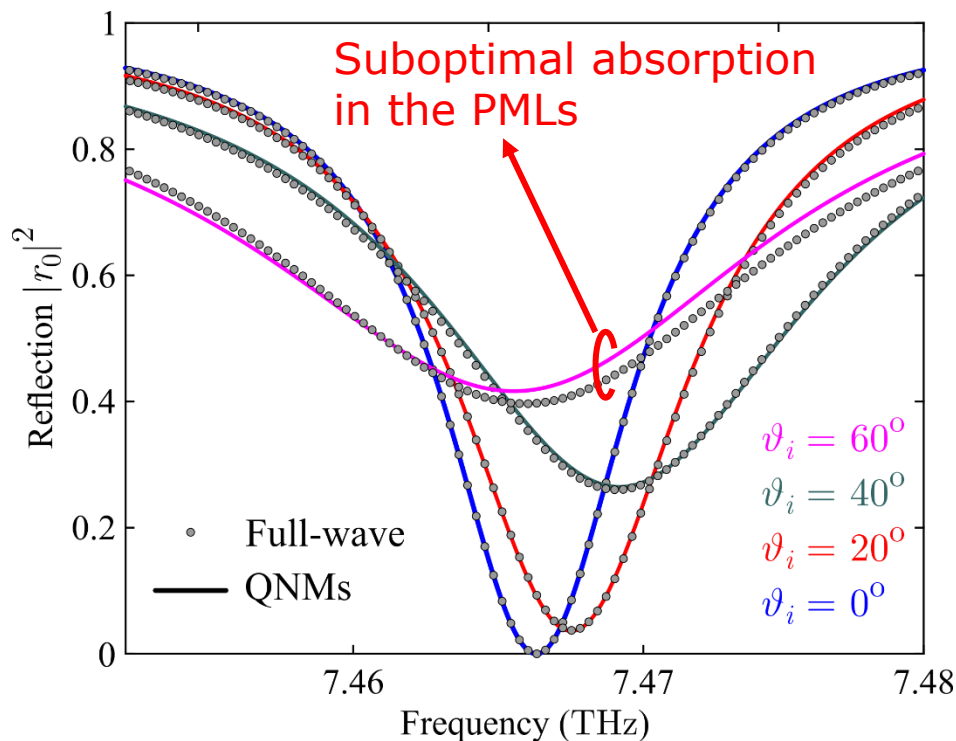
Scattered field

$$\mathbf{H}_{\text{sct}}(x, d) = \sum_m a_m(\omega) \tilde{\mathbf{h}}_m(x, d) e^{-j\tilde{k}_m \sin \vartheta_i x}$$

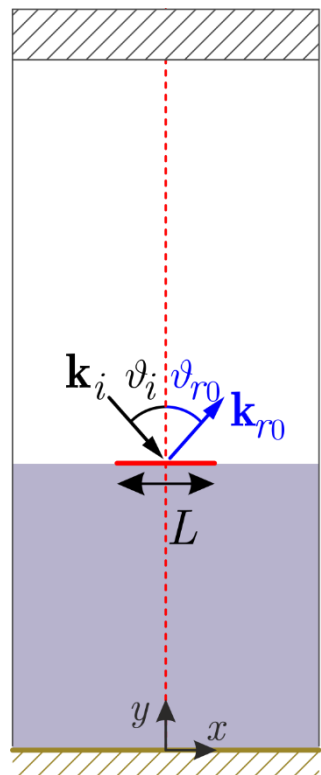
QNM expansion
 coefficients

QNMs (envelopes)

Periodic phase
 term (different
 for each QNM)



Linear absorption

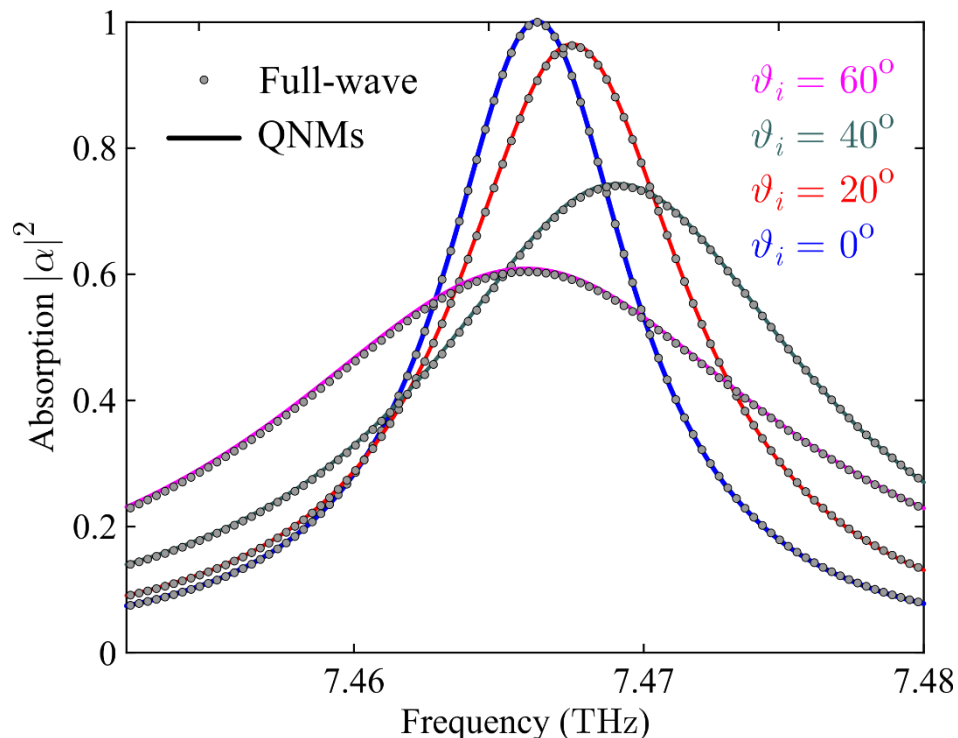


$$L = 5 \mu\text{m}$$

✓ Absorption coefficient:

$$|\alpha|^2 = \frac{1}{P_{\text{inc}}} \int_{-L/2}^{L/2} \text{Re}\{\sigma_s(\omega)\} |\mathbf{e}_{\text{sct},\parallel}|^2 dx$$

✓ **Perfect agreement for any angle of incidence** since the expansion is complete on the resonator (graphene sheet)



No need for the phase term here!

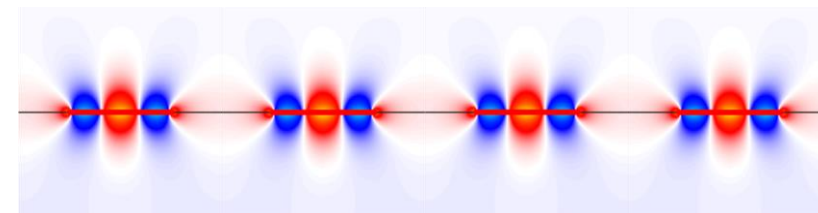
Scattered field's envelope

$$\mathbf{e}_{\text{sct}} = \sum_m a_m(\omega) \tilde{\mathbf{e}}_m$$

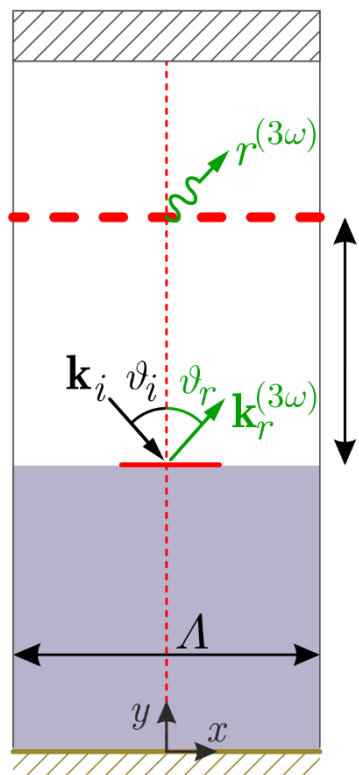
QNM expansion coefficients

QNMs (envelopes)

No need to consider background field (incident and reflected) due to the **high confinement** and enhancement of the GSPs



Nonlinear absorption and specular reflection



$$\Lambda = 10 \mu\text{m}$$

$$\lambda_0 \sim 40 \mu\text{m}$$

✓ Nonlinear absorption coefficient:

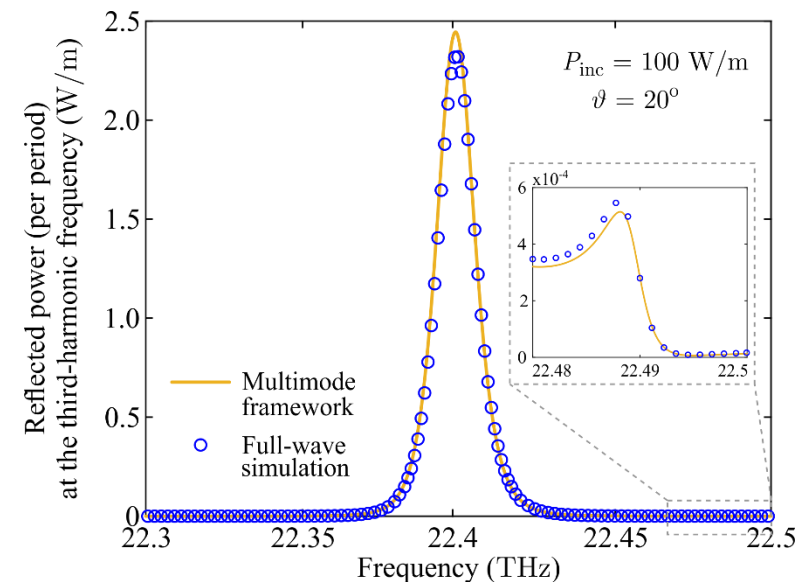
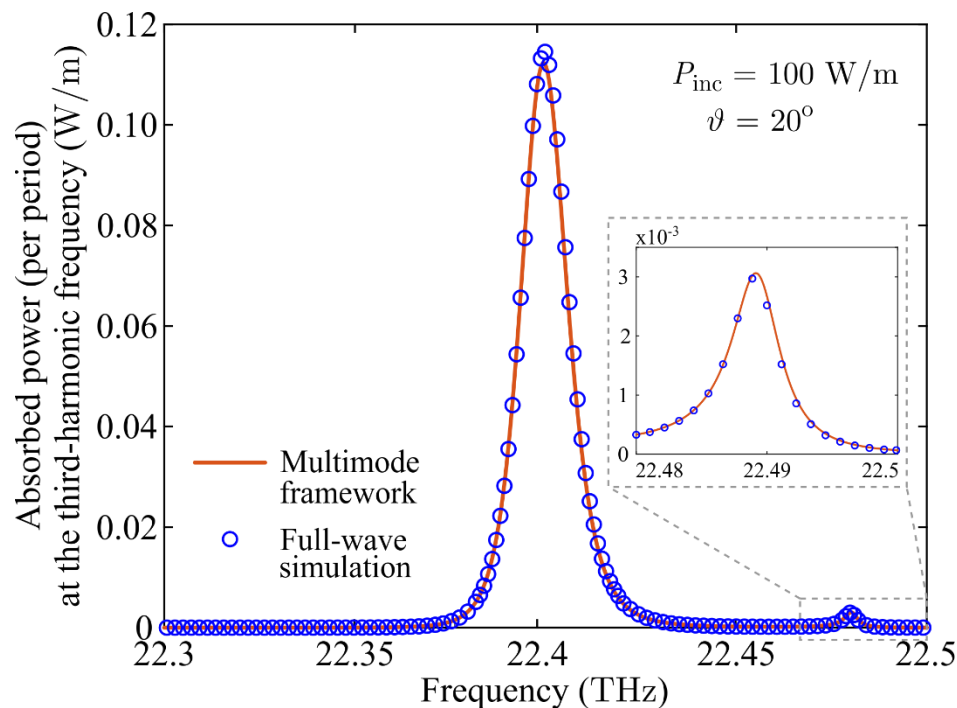
✓ Nonlinear reflection coefficient:

$$|\alpha|^2 = \frac{1}{P_{\text{inc}}} \int_{-L/2}^{L/2} \text{Re}\{\sigma_s(\omega)\} |e_{\text{sct},||}^{(3\omega)}|^2 dx$$

$$r_0 = \frac{1}{\Lambda H_0} e^{jk_r^{(3\omega)} d} \int_{-\Lambda/2}^{\Lambda/2} \mathbf{H}_{\text{sct}}^{(3\omega)}(x, d) e^{jk_r^{(3\omega)} x} dx$$

Scattered field @ 3ω

$$\mathbf{H}_{\text{sct}}^{(3\omega)}(x, d) = \sum_m a_m(3\omega) \tilde{\mathbf{h}}_m(x, d) e^{-j\tilde{k}_m \sin \vartheta_i x}$$



A more practical system

Higher diffraction orders in graphene strip metasurface

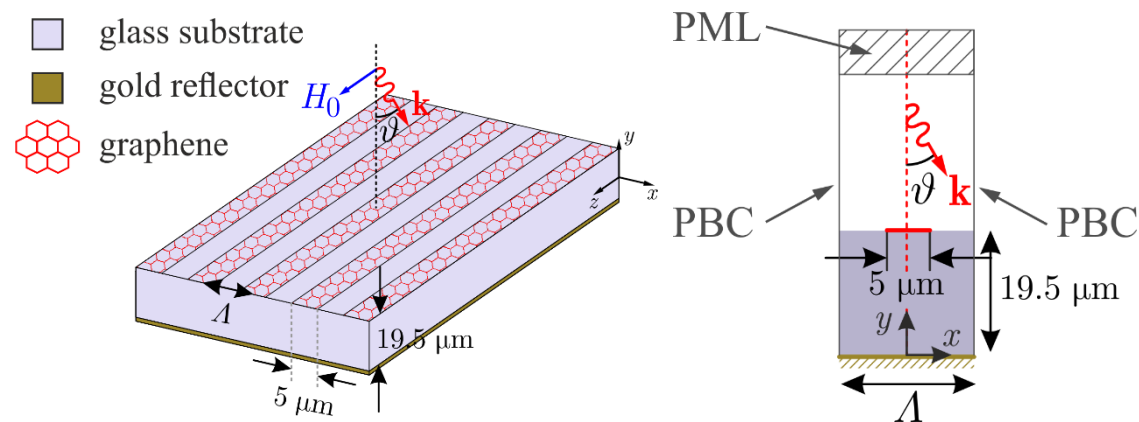
Resonant System

- ✓ Expansion of the previous graphene strip configuration in a **reflective metasurface/grating**

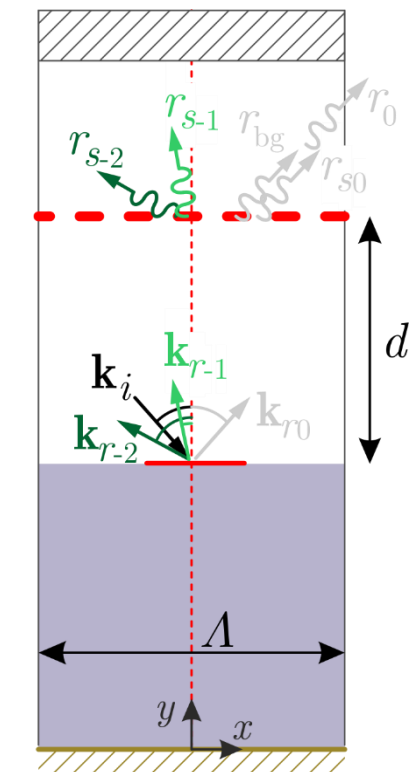
- ✓ Periodicity Λ **controls reflection** and allows:
 - only for specular reflection (subwavelength metasurface)
 - excitation of higher diffraction orders (reflective grating)

- ✓ A higher diffraction order ($\ell \neq 0$) is excited when $\frac{\Lambda}{\lambda_0} > -\frac{\ell}{n_i(\sin \vartheta \pm 1)}$

- ✓ For example, in normal incidence a higher diffraction order is excited when $\frac{\Lambda}{\lambda_0} > \pm|\ell|$



Higher diffraction orders linear calculation

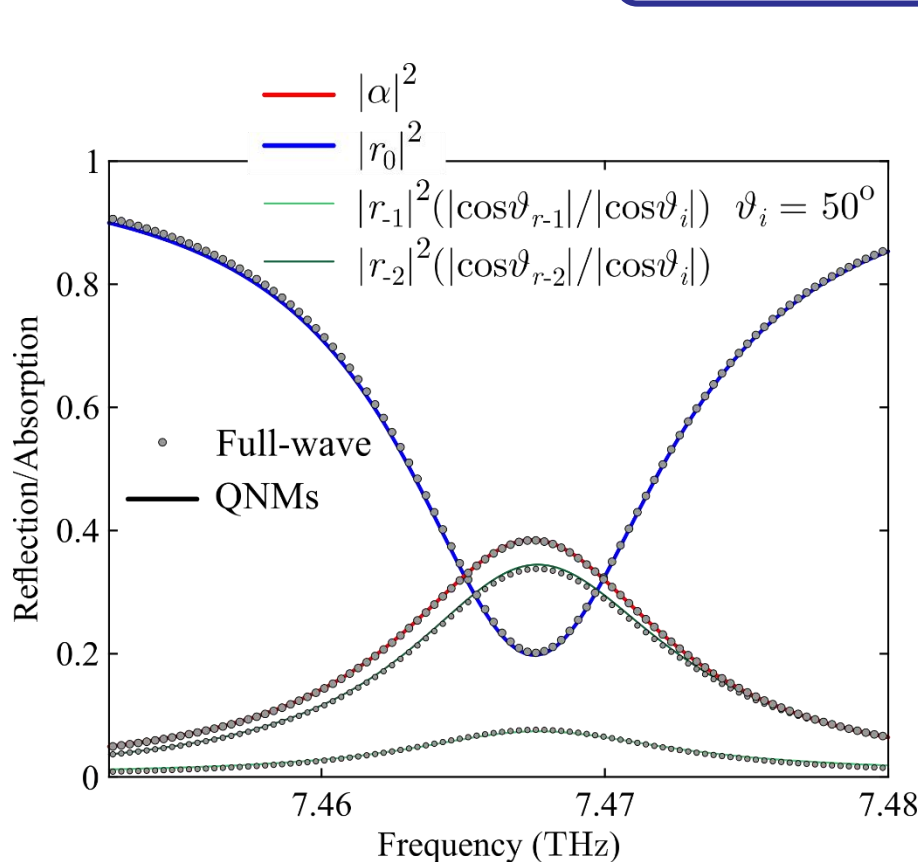


$$\Lambda = 50 \mu\text{m}$$

$$\lambda_0 \sim 40 \mu\text{m}$$

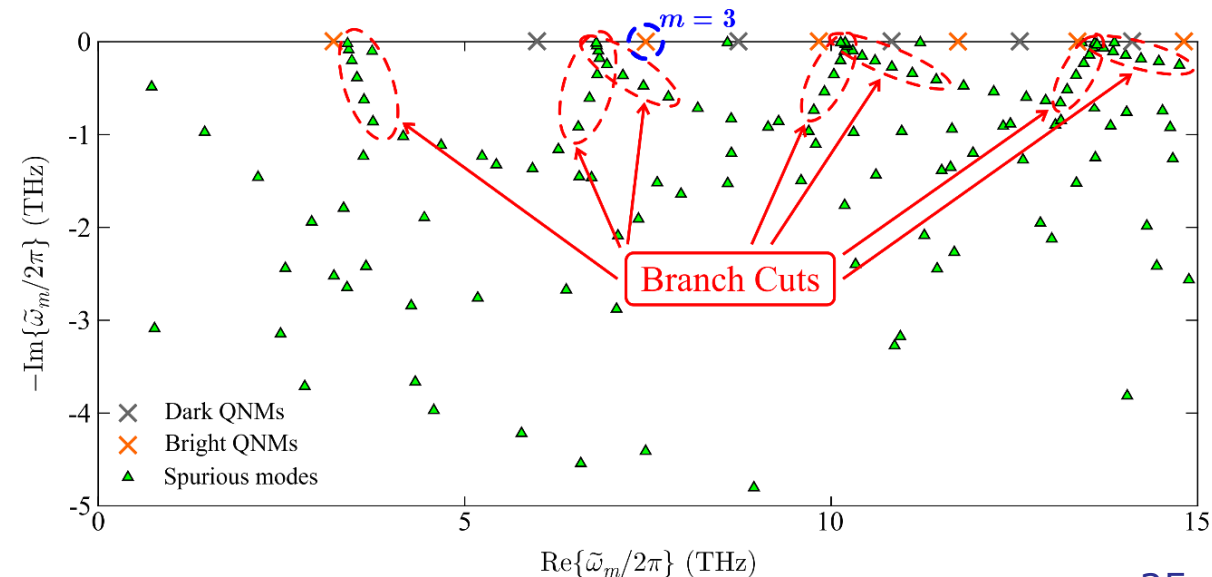
- ✓ Higher diffraction orders: $k_{r\ell,x} = k_0 n_i \sin \vartheta_i + \ell \frac{2\pi}{\Lambda}$ and $k_{r\ell,y} = \sqrt{k_0^2 - k_{r\ell,x}^2}$

- ✓ Reflection coefficient:
$$r_\ell = \frac{1}{\Lambda H_0} e^{jk_{r\ell,y}d} \int_{-\Lambda/2}^{\Lambda/2} \mathbf{H}_{\text{sct}}(x, d) e^{jk_{r\ell,x}x} dx$$



Scattered field

$$\mathbf{H}_{\text{sct}}(x, d) = \sum_m a_m(\omega) \tilde{\mathbf{h}}_m(x, d) e^{-j\tilde{k}_m \sin \vartheta_i x}$$



Summary and Conclusion

Summary

- ✓ Presentation of a **multimode** QNMs framework, rigorously incorporating **2D materials**
- ✓ Accurate description of linear and **nonlinear effects** (third-harmonic generation) in single scatterers and periodic structures (metasurfaces)
- ✓ Excellent accuracy of the framework (validated through full-wave simulations)
- ✓ Examination of gratings with 2D materials that support **higher diffraction orders**

Outlook




- ✓ Expansion of the framework in 3D metasurfaces for additional degrees of freedom and richer mode interaction effects
- ✓ Examination of other nonlinear effects (self-phase modulation)

Publications



PHYSICAL REVIEW B **107**, 035413 (2023)

Multimode non-Hermitian framework for third harmonic generation in nonlinear photonic systems comprising two-dimensional materials

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(Received 7 November 2022; revised 23 December 2022; accepted 23 December 2022; published 13 January 2023)

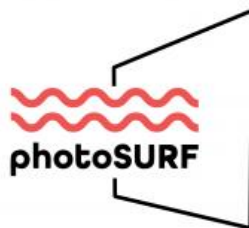
✓ **In preparation:** Higher-diffraction orders handling with QNMs

Thank you!

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This work was supported by the Hellenic Foundation for Research and Innovation (H.F.R.I.) under the
“2nd Call for H.F.R.I. Research Projects to support Post-doctoral Researchers”
(Project No. 916, PHOTOSURF)



ΕΘΝΙΚΟ ΙΔΡΥΜΑ ΕΡΕΥΝΩΝ
National Hellenic Research Foundation