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Rigorous retrieval of linear and nonlinear parameters in graphene waveguides

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Presentation Outline

- **Framework**
  - Nonlinear propagation in graphene waveguides
  - Motivation & objectives
- **Electromagnetic modelling**
  - Graphene as a conductive sheet
  - Equivalent bulk medium representation
  - FEM formulation considering graphene as a sheet
  - Nonlinear waveguide parameters and the NLSE
- **Review of graphene physical properties**
  - Linear conductivity
  - Nonlinear conductivity
- **Nonlinear parameter calculation**
  - Optical frequencies
  - Terahertz
- **To probe further**

Framework

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Framework / Nonlinear propagation in graphene waveguides

❖ Strong recent interest in nonlinear propagation effects in graphene but with some skepticism, as well.

Hendry *et al*., Coherent Nonlinear Optical Response of Graphene, PRL, 2010

Gu *et al.*, Regenerative oscillation and four-wave mixing in graphene optoelectronics, Nat Photonics, 2012

Gorbach, Nonlinear graphene plasmonics: Amplitude equation for surface plasmons, PRA, 2013

Ooi *at al*., Waveguide engineering of graphene's nonlinearity, APL, 2014

Khurgin, Graphene—A rather ordinary nonlinear optical material, APL, 2014

❖ Various publications report high (giant?) nonlinearity levels in graphene.

❖ Theoretical frameworks still not well developed and lacking the understanding of nonlinear effects in photonics.

❖ Rather poor correlation between theoretical and experimental results and very few device-oriented experiments.

❖ So, is there any exploitable potential in nonlinear graphene waveguides?

Framework / Motivation & objectives

❖ Current literature in nonlinear graphene waveguides include various simplifications and misconceptions related to:

- inappropriate effective medium representations
- inconsistent introduction of graphene tensorial properties
- superficial or excessive models for graphene’s nonlinearity
- poor waveguide engineering

❖ Our objectives include:

- treatment of graphene as sheet (2D material)
- full/complete tensorial representation of nonlinear surface conductivity
- rigorous calculation of nonlinear parameter γ ($W^{-1}m^{-1}$) for arbitrary waveguide cross-sections (Kerr response)
- quantify individual sheet and bulk nonlinear contributions in γ for a range of waveguide archetypes
- engineer the waveguide cross-section to enhance γ

Electromagnetic Modelling

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Electromagnetic Modelling / Graphene as a conducting sheet

Maxwell's curl equations, including both linear and nonlinear contributions:

$$\nabla \times \tilde{\mathbf{E}} = +i\omega\mu_0\bar{\mu}_r\tilde{\mathbf{H}}$$

$$\nabla \times \tilde{\mathbf{H}} = -i\omega(\varepsilon_0\tilde{\mathbf{E}} + \tilde{\mathbf{P}}) + \tilde{\mathbf{J}}$$

$$\tilde{\mathbf{J}} = \tilde{\mathbf{J}}_{\mathrm{lin}} + \tilde{\mathbf{J}}_{\mathrm{NL}}, \quad \tilde{\mathbf{J}}_{\mathrm{NL}} << \tilde{\mathbf{J}}_{\mathrm{lin}} = \sigma_s^{(1)}\tilde{\mathbf{E}}$$

$$\tilde{\mathbf{P}} = \tilde{\mathbf{P}}_{\mathrm{lin}} + \tilde{\mathbf{P}}_{\mathrm{NL}}, \quad \underbrace{\tilde{\mathbf{P}}_{\mathrm{NL}}}_{\text{perturbation}} << \tilde{\mathbf{P}}_{\mathrm{lin}} = \varepsilon_0\bar{\chi}^{(1)}\tilde{\mathbf{E}}$$

Bulk current density, expanded similarly to polarization $\mathbf{P}$:

$$\mathbf{J}_b = \bar{\sigma}^{(1)} \mid \mathbf{E} + \bar{\sigma}^{(2)} \mid \mathbf{EE} + \bar{\sigma}^{(3)} \mid \mathbf{EEE} + \ldots$$

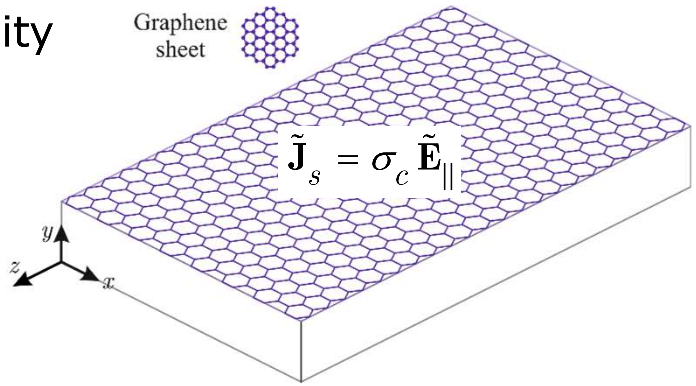
Surface current density on graphene:

$$\mathbf{J}_s = \underbrace{\bar{\sigma}_s^{(1)} \mid \mathbf{E}}_{\text{important}} + \bar{\sigma}_s^{(2)} \mid \mathbf{EE} + \underbrace{\bar{\sigma}_s^{(3)} \mid \mathbf{EEE}}_{\text{important}} + \ldots$$

$\boxed{\bar{\sigma}_s^{(n)}}$ Surface conductivity tensor, rank (n+1), in $[\mathrm{S(m/V)}^{n-1}]$

Overall current density

$$\mathbf{J} = \mathbf{J}_b + \mathbf{J}_s\delta_s(\mathbf{r})$$



$$\bar{\sigma}_s^{(1)} = \begin{bmatrix} \sigma_{s,xx} & 0 & \sigma_{s,xz} \\ 0 & 0 & 0 \\ \sigma_{s,zx} & 0 & \sigma_{s,zz} \end{bmatrix}$$

$$\sigma_{s,xx} = \sigma_{s,zz} \equiv \sigma_c, \sigma_{s,xz} = \sigma_{s,zx} \simeq 0$$

Electromagnetic Modelling / Graphene as a conducting sheet

We focus on **self-acting third-order** nonlinear effects, i.e. Kerr effect and TPA:

$$\boxed{J_{s,j,\mathrm{NL}} = \mathbf{J}_{s,\mathrm{NL}} \cdot \mathbf{j} = \frac{3}{4}\sum_{klm} \sigma^{(3)}_{s,jklm} \; E_k E_l^* E_m}$$

j-th Cartesian of the nonlinear surface current

$\bar{\sigma}_s^{(3)}$ Surface conductivity tensor, 4th-rank (81 elements), units [S(m/V)²]

- Hexagonal symmetry group
- Normal to sheet component of $J_{s,NL}$ vanishes

Up to **14 non-zero** elements & up to **6** are **independent**!

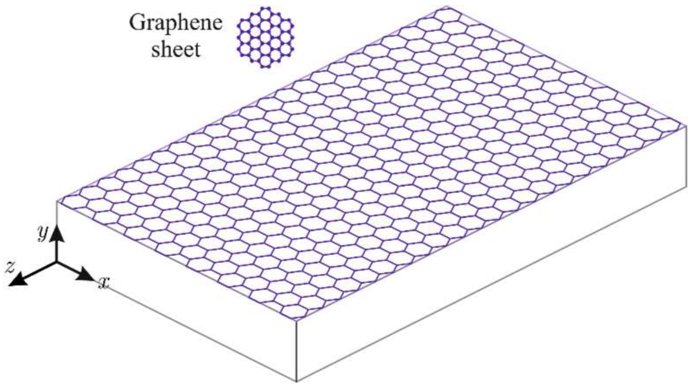
Simplest form (8 non-zero, 1 independent), i.e. 2D-equivalent of an isotropic bulk (3D) medium:

$$\boxed{\sigma^{(3)}_{s,jklm} = \sigma_3 \frac{1}{3}\left(\delta_{jk}\delta_{lm} + \delta_{jm}\delta_{kl} + \delta_{jl}\delta_{mk}\right)}$$

For example, a graphene sheet normal to the y-axis

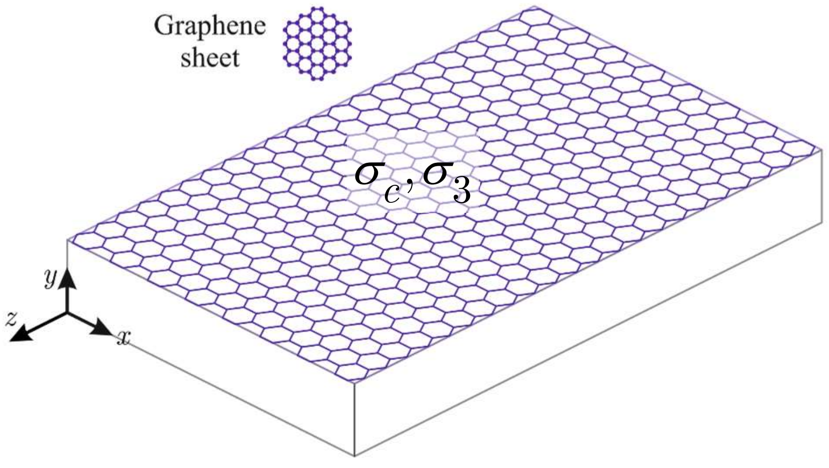
$$\sigma^{(3)}_{s,xxxx} = \sigma^{(3)}_{s,zzzz} = \sigma_3$$

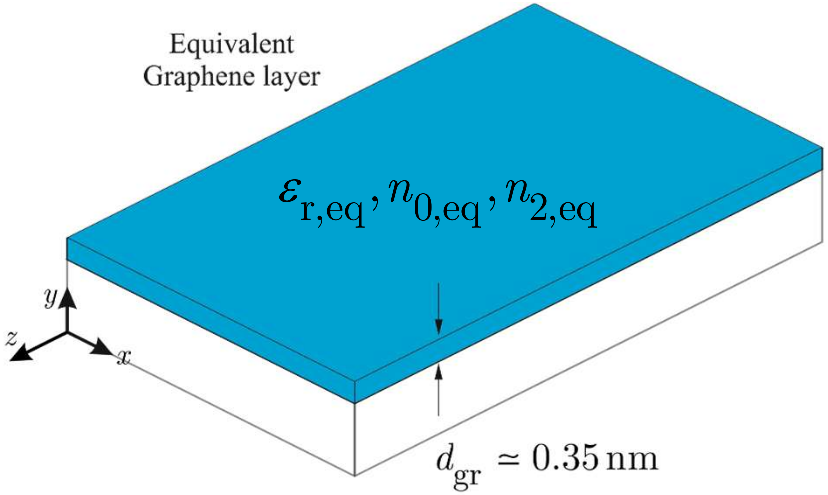
$$\sigma^{(3)}_{s,mmnn} = \sigma^{(3)}_{s,mnmn} = \sigma^{(3)}_{s,mnnm} = \frac{1}{3}\sigma_3 \qquad \{m,n\} = \{x,z\}$$



Electromagnetic Modelling / Equivalent bulk medium representation

Widespread choice, but commonly leads to poor or misleading results!





Equivalent bulk medium pros & cons:

- very simple approach
- allows usage of existing SW tools
- introduces an artificial thickness
- excessive computational burden
- tensorial information easily lost

$$\text{map } \varepsilon_{r,\mathrm{eq}} \to \overline{\varepsilon}_{r,\mathrm{eq}}$$

$$\overline{\chi}_{\mathrm{eq}}^{(m)} = i\overline{\sigma}_s^{(m)} / (\omega\varepsilon_0 d_{\mathrm{gr}})$$

$$\varepsilon_{\mathrm{r,eq}} = n_{0,\mathrm{eq}}^2 = 1 + \chi_{\mathrm{eq}}^{(1)} = 1 + \frac{i\sigma_c}{\omega\varepsilon_0 d_{\mathrm{gr}}}$$

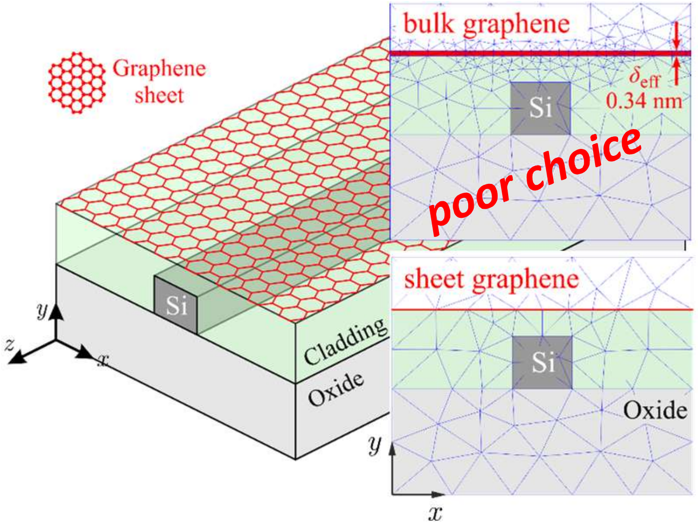
$$n_{2,\mathrm{eq}} = \frac{3}{4}\frac{\chi_{\mathrm{eq}}^{(3)}}{\varepsilon_{\mathrm{r,eq}}} Z_0 = \frac{3}{4} \times \frac{i\sigma_3 / (\omega\varepsilon_0 d_{\mathrm{gr}})}{1 + i\sigma_c / (\omega\varepsilon_0 d_{\mathrm{gr}})} Z_0$$

Electromagnetic Modelling / FEM formulation considering graphene as a sheet

Finite Element Method is used to analyze the **linear** problem. Graphene is rigorously introduced as **sheet** (zero thickness) characterized by **surface** conductivity tensors.

$$\begin{aligned} \mathbf{n}\times(\tilde{\mathbf{E}}_2-\tilde{\mathbf{E}}_1) &= \mathbf{0} \\ \mathbf{n}\times(\tilde{\mathbf{H}}_2-\tilde{\mathbf{H}}_1) &= \tilde{\mathbf{J}}_s \end{aligned}$$

Boundary Conditions



Usual Galerkin procedure:

$$\underbrace{\iiint_V \left\{(\nabla\times\tilde{\mathbf{E}}_a)\cdot[\mu_r^{-1}\ (\nabla\times\tilde{\mathbf{E}})] - k_0^2\tilde{\mathbf{E}}_a\cdot[\bar{\varepsilon}_{\mathrm{r}}\ \tilde{\mathbf{E}}]\right\}\mathrm{d}V}_{\text{LHS} \rightarrow \text{Standard FEM bulk term}} = \underbrace{i\omega\mu_0\iint_S \tilde{\mathbf{E}}_a\cdot[\ \bar{\sigma}_s^{(1)}\ \tilde{\mathbf{E}}]\mathrm{d}S}_{\text{RHS} \rightarrow \text{Graphene surface term}}$$

In the simplest case: $\bar{\sigma}_s^{(1)}\,\tilde{\mathbf{E}} = \sigma_c\,\tilde{\mathbf{E}}_{||} = \sigma_c\left[\mathbf{n}\times(\tilde{\mathbf{E}}\times\mathbf{n})\right]$

RHS $$\iint_S \tilde{\mathbf{E}}_a\cdot[\ \bar{\sigma}_s^{(1)}\ \tilde{\mathbf{E}}]\mathrm{d}S == \sigma_c\iint_S(\mathbf{n}\times\tilde{\mathbf{E}}_a)\cdot(\mathbf{n}\times\tilde{\mathbf{E}})\mathrm{d}S$$

Electromagnetic Modelling / Nonlinear waveguide parameters and the NLSE

Nonlinear Schrodinger Equation (NLSE) framework for Kerr & TPA: extract the nonlinear parameter γ_{NL} $(mW)^{-1}$, that quantifies nonlinear phase shift or loss.

Basic Figure of Merit $\mathcal{F} = \gamma_{\mathrm{NL}} L_{\mathrm{prop}}$ (1/W)

$$\nabla \times \tilde{\mathbf{H}} = -i\omega\varepsilon_0 \left(\bar{\varepsilon}_{\mathrm{r}} - \frac{\bar{\sigma}^{(1)}}{i\omega\varepsilon_0} \right) \tilde{\mathbf{E}} - i\omega \underbrace{\left(\tilde{\mathbf{P}}_{\mathrm{NL}} + \frac{i}{\omega} \tilde{\mathbf{J}}_{\mathrm{NL}} \right)}$$

Overall perturbation term $\tilde{\mathbf{P}}'_{\mathrm{NL}} = \tilde{\mathbf{P}}_{\mathrm{NL}} + i\omega^{-1}\tilde{\mathbf{J}}_{\mathrm{NL}}$

Vector NLSE (Afshar & Monro, OPEX, 2009), (Daniel & Agrawal, JOSA B, 2010):

$$\tilde{\mathbf{E}}(\mathbf{r},\omega) = \tilde{A}(z,\omega-\omega_0)\mathbf{e}(x,y,\omega_0)\exp(+i\beta_0 z) \,/\, \sqrt{N}$$

Time-domain nonlinear propagation equation for SVE

$$\boxed{\frac{\partial A}{\partial z} = \frac{i\omega_0 e^{i(\omega_0 t - \beta_0 z)}}{2\sqrt{4N}} \iint \mathbf{e}^* \cdot \left(\mathbf{P}_{\mathrm{NL}} + \frac{i}{\omega_0} \mathbf{J}_{\mathrm{NL}} \right) \mathrm{d}S + \mathcal{L}A}$$

Linear operator $\mathcal{L}$: dispersion + losses

Electromagnetic Modelling / Nonlinear waveguide parameters and the NLSE

$$P_{j,\mathrm{NL}} = \mathbf{P}_{\mathrm{NL}} \cdot \mathbf{j} = \frac{3\varepsilon_0}{4} \sum_{klm} \chi^{(3)}_{jklm} \; E_k E_l^* E_m$$

$$J_{s,j,\mathrm{NL}} = \mathbf{J}_{s,\mathrm{NL}} \cdot \mathbf{j} = \frac{3}{4} \sum_{klm} \sigma^{(3)}_{s,jklm} \; E_k E_l^* E_m$$

We substitute in the time-domain nonlinear equation for the SVE

Nonlinear Schrodinger Equation
(here simply written for CW)

$$\boxed{\frac{\partial A}{\partial z} = -\frac{1}{2L_{\mathrm{prop}}} A + i(\gamma_b + \gamma_s) \,|\, A \,|^2 \; A}$$

Bulk nonlinearity

Sheet (graphene) nonlinearity

$$\gamma_b = \frac{3\omega_0 \epsilon_0}{4(2N)^2} \sum_{jklm}^{xyz} \iint \chi^{(3)}_{jklm} e_j^* e_k e_l^* e_m \, \mathrm{d}S \qquad \gamma_s = i\frac{3}{4(2N)^2} \sum_{jklm}^{xyz} \int \sigma^{(3)}_{s,jklm} e_j^* e_k e_l^* e_m \, \mathrm{d}\ell$$

Simplest possible, yet rigorous, expressions

$$\gamma_b = \frac{\omega_0 \epsilon_0}{(2N)^2} \iint \chi_3 \left(\frac{1}{2} \,|\, \mathbf{e} \,|^4 + \frac{1}{4} \,|\, \mathbf{e} \cdot \mathbf{e} \,|^2 \right) \mathrm{d}S \qquad \gamma_s = i\frac{1}{(2N)^2} \int \sigma_3 \left(\frac{1}{2} \,|\, \mathbf{e}_{\|} \,|^4 + \frac{1}{4} \,|\, \mathbf{e}_{\|} \cdot \mathbf{e}_{\|} \,|^2 \right) \mathrm{d}\ell$$

Review of graphene physical properties

Review of graphene physical properties / Linear conductivity

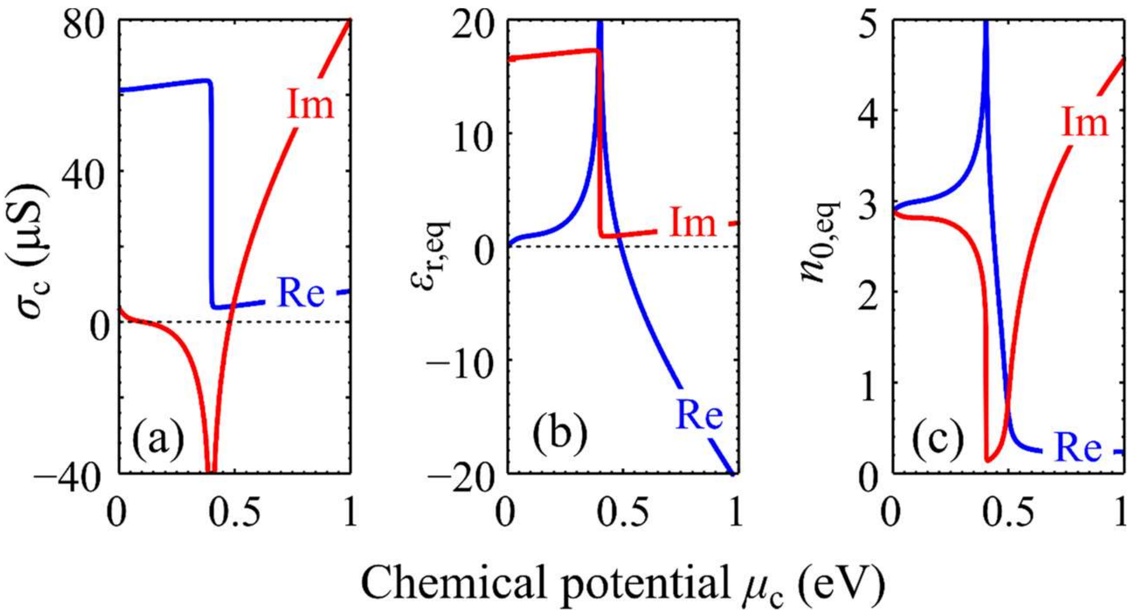
Overall linear surface conductivity $\boxed{\sigma_c = \sigma_{c,\text{intra}} + \sigma_{c,\text{inter}}}$ Simplest scalar form of $\bar{\sigma}_s^{(1)}$

$$\sigma_{c,\text{intra}} = i\frac{e^2\mu_c}{\pi\hbar^2(\omega + i/\tau_1)} \times \mathcal{T}\left(\frac{\mu_c}{2k_BT}\right)$$

Intraband contribution
(Drude)

$$\sigma_{c,\text{inter}} = i\frac{e^2}{4\pi\hbar}\ln\left[\frac{2|\mu_c| - \hbar(\omega + i/\tau_2)}{2|\mu_c| + \hbar(\omega + i/\tau_2)}\right]$$

Interband contribution
(unimportant at THz)
Disappears when $\mu_c > \hbar\omega/2$



Re & Im part of graphene surface conductivity, equivalent ε_r and equivalent (linear) refractive index n_0 at 1550 nm.

Review of graphene physical properties / Nonlinear conductivity

Nonlinear graphene surface conductivity: a topic of much debate!

Simplest possible form of 4th-rank tensor $\bar{\sigma}_s^{(3)}$:

$$\sigma_{s,jklm}^{(3)} = \boxed{\sigma_3} \frac{1}{3}\left(\delta_{jk}\delta_{lm} + \delta_{jm}\delta_{kl} + \delta_{jl}\delta_{mk}\right)$$

$$\sigma_{3,\mathrm{opt}} \approx -i10^{-23}\,[\mathrm{S(m\,/\,V)^2}]\ @\ 1550\,\mathrm{nm}$$

Hendry *et al.*, PRL, 2010

$$\sigma_{3,\mathrm{opt}} = -i\frac{9e^4 v_F^2}{32\omega^4\hbar^3}$$

$$v_F \approx c_0\,/\,300$$

$$\sigma_{3,\mathrm{THz}} \approx -i10^{-19}\,[\mathrm{S(m\,/\,V)^2}]\ @\ 10\,\mathrm{THz}$$

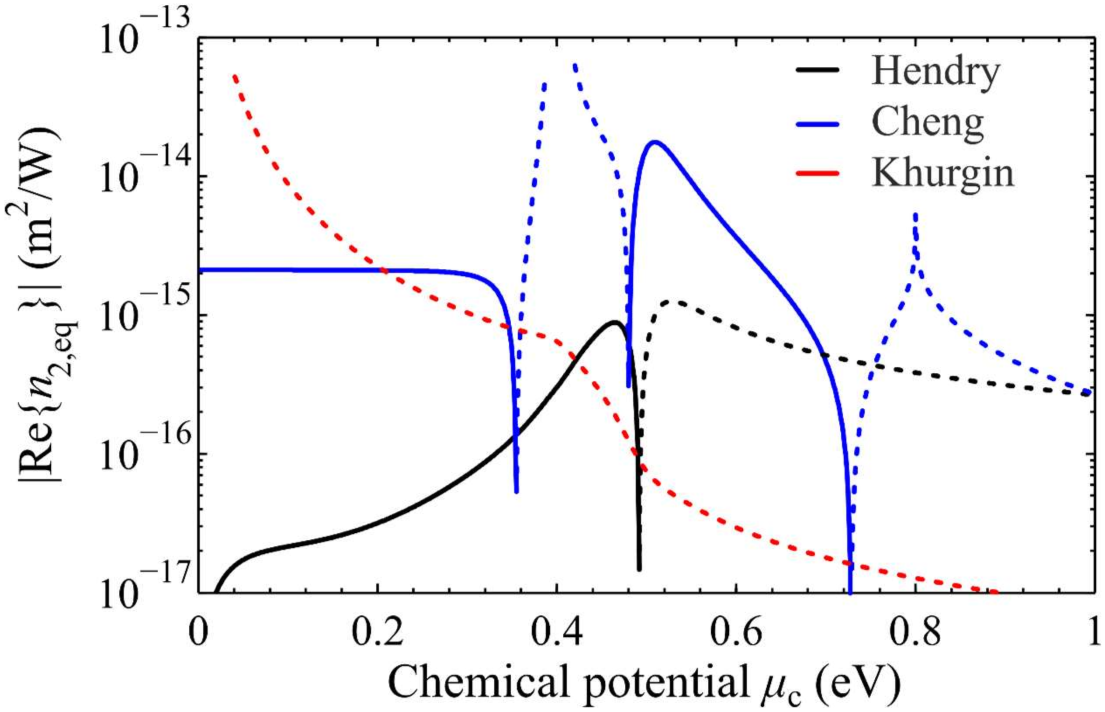
Mikhailov & Ziegler, J. Phys.: Condens. Matter, 2008

$$\sigma_{3,\mathrm{THz}} = -i\frac{3e^4 v_F^2}{32\omega^3\hbar^2\mu_c}$$

Number of elements in graphene’s nonlinear surface conductivity tensor $\bar{\sigma}_s^{(3)}$

Source	Non-zero	Independent
Hexagonal 2D crystal	8+6	3+3
Gorbach et al., OL, 2013	8+6	1+1
Cheng et al., New J. Phys, 2014	8	2
Simplest model	8	1

Review of graphene physical properties / Nonlinear conductivity



Absolute value of Re{$n_{2,eq}$}.
Solid (positive),
Dashed (negative).

Equivalent nonlinear index n_2 undergoes sign transitions at $\left|\mathrm{Re}\{n_{0,\mathrm{eq}}\}\right| = \left|\mathrm{Im}\{n_{0,\mathrm{eq}}\}\right|$

Index n_2 can be in the range of **10^{-15} m^2/W**. For comparison, Si ~2.5×10^{-18} m^2/W, Chalcogenides ~10^{-17} m^2/W, polymers (DDMEBT) ~1.7×10^{-17} m^2/W.

However, without the right waveguide engineering, this high value does **not** translate into a high γ_{NL} parameter.

Nonlinear parameter calculation

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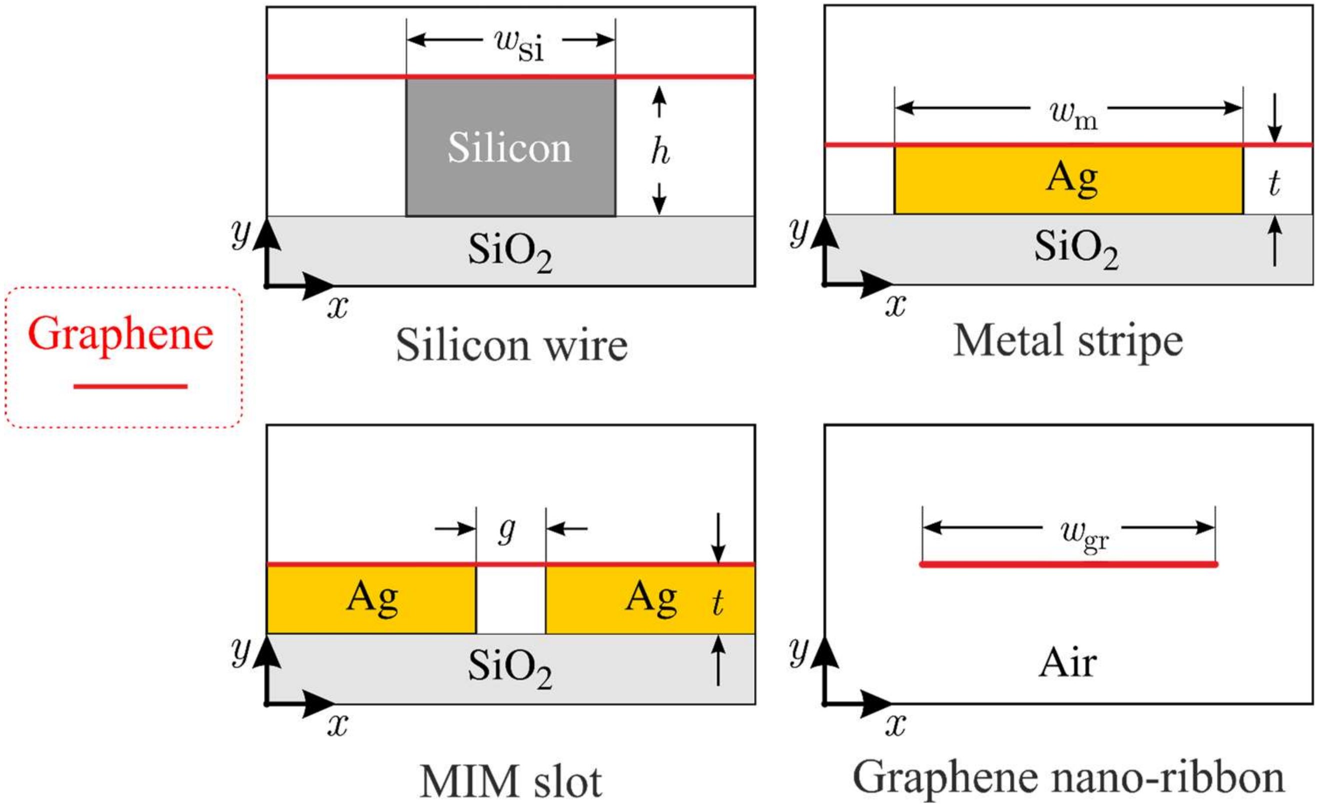
Nonlinear parameter calculation / Optical frequencies



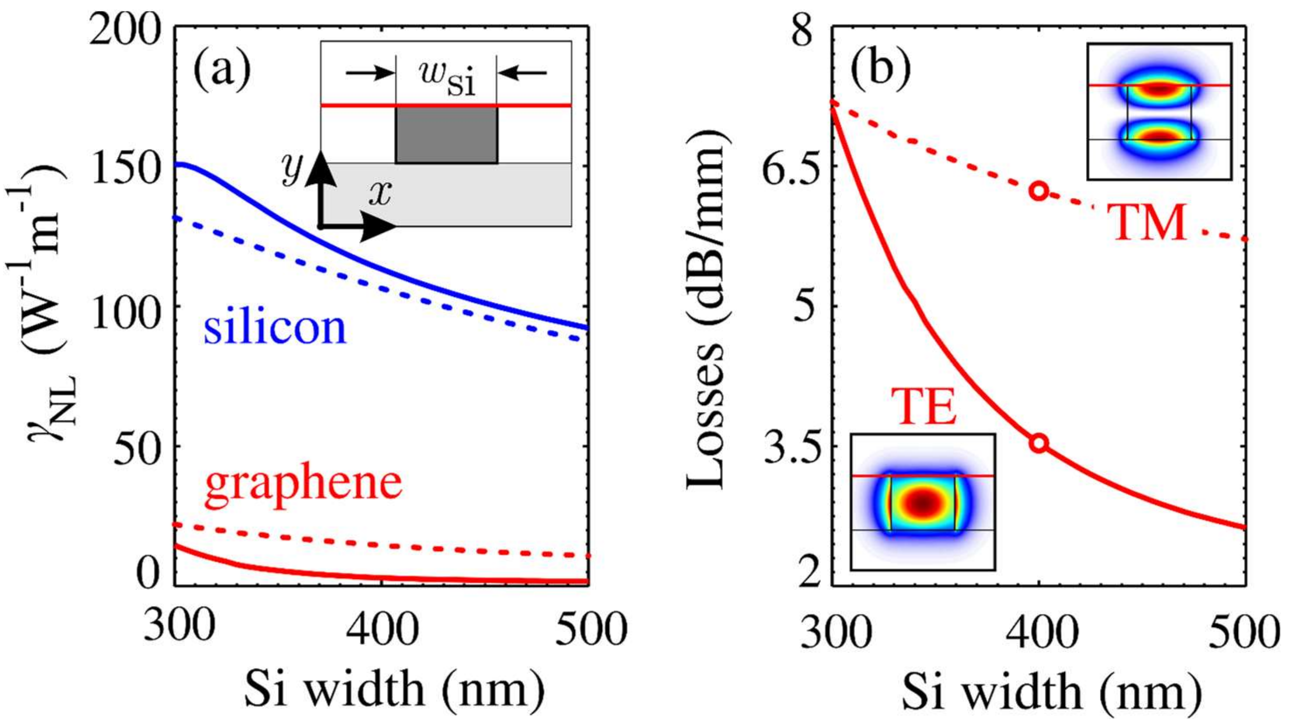
Here shown simple/trivial layouts of graphene, but our formulation allows for:

- Inhomogeneous σ_1 & σ_3
- arbitrary orientations (curved, etc.)
- multiple sheets

➢ We will assess how the presence of a graphene layer affects the nonlinearity of “standard” photonic and plasmonic waveguides.

Nonlinear parameter calculation / Optical frequencies

Silicon wire overlaid with graphene



Insets:

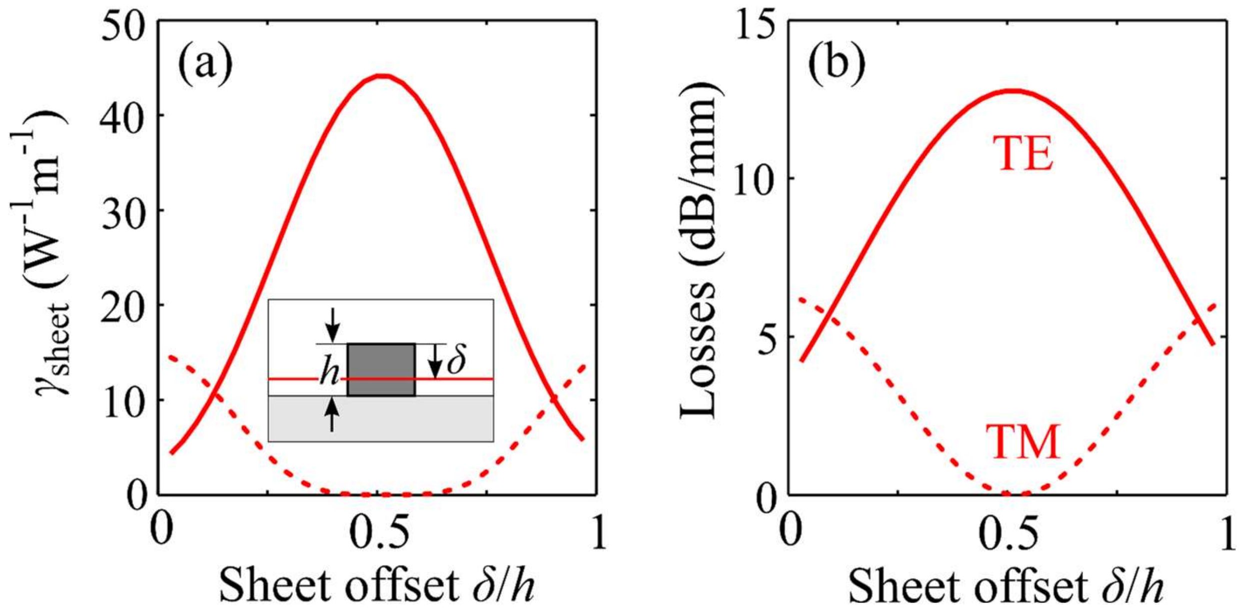
$$|e_x|^2 + |e_z|^2$$

$$\mu_c = 0.5\,\mathrm{eV}$$

- Graphene nonlinearity is much lower compared to that of bulk Si.
- Marginal interaction of w/g mode with graphene.
- Counterintuitively, γ_{gr} is larger for TM modes, partially due to E_z!
- Losses are too high (they reduce from ~1 dB/cm to ~5 dB/mm).

Nonlinear parameter calculation / Optical frequencies

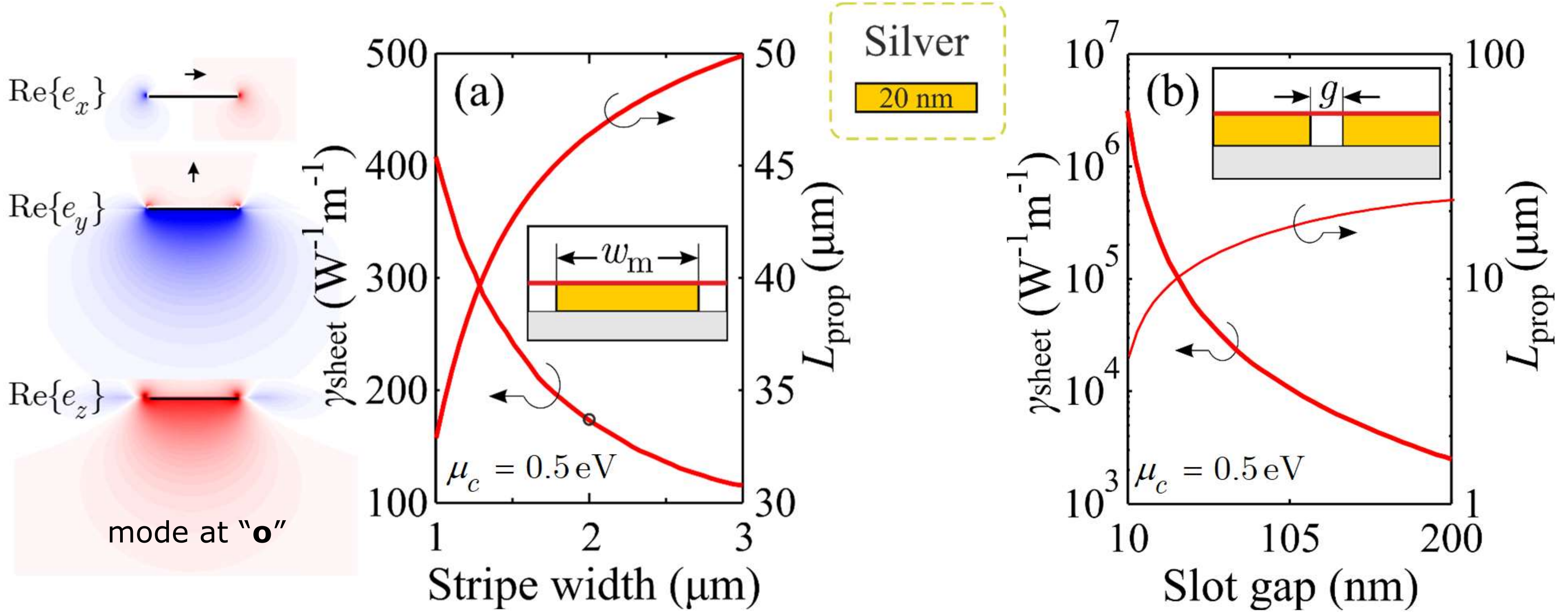
Can we enhance graphene's contribution in the Si-wire waveguide?



- Graphene sheet artificially offset through the Si-core.
- Sheet nonlinearity is now comparable to that of bulk Si (~110 $W^{-1}m^{-1}$).
- May not be easy to fabricate.
- Still higher losses.

Nonlinear parameter calculation / Optical frequencies

Plasmonic waveguides: metal stripe & MIM slot

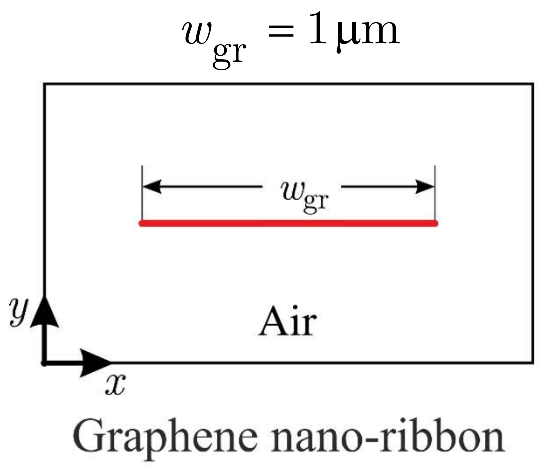


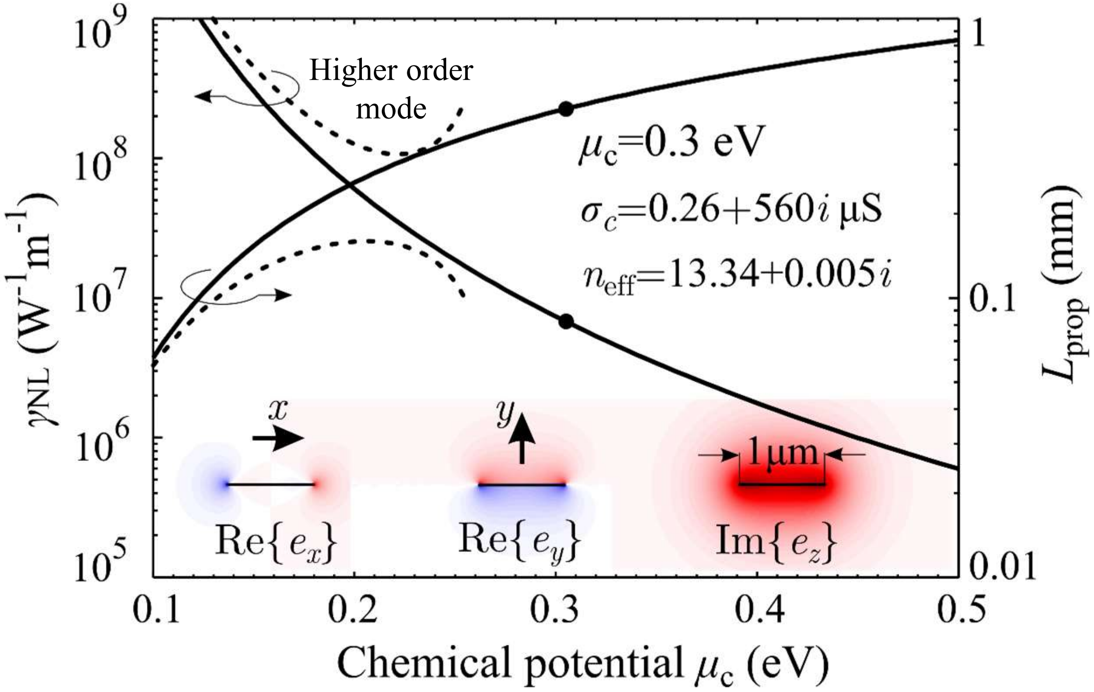
- We exploit the y-antisymmetric mode and not the symmetric "long-range".
- γ_{sheet} exceeds Si contribution.
- γ_{sheet} improves by reducing thickness.
- Metal loss dominates.

- TE polarization ensures strong interaction. High field confinement.
- γ_{sheet} orders of magnitude above Si.
- γ_{sheet} improves by reducing thickness.
- For small gaps metal & graphene equally contribute to losses.

Nonlinear parameter calculation / Terahertz

THz: Graphene Nano-Ribbon (GNR)





- Nonlinear parameter γ_{NL} is excessive.
- The "edge" plasmonic mode provides highly confined field components.
- Propagation length ~10λ at 10 THz.
- Ample tuning via chemical potential (gating), as σ_3 depends on μ_c.

To probe further

To probe further

❖ Current work:

- **rigorous derivation** of the nonlinear parameter γ
- **full tensorial properties** retained
- **MIM** slots overlaid with graphene lead to ultra-high γ at tolerable losses
- **higher nonlinearity** is always associated with increasing **losses**
- more promising results provided by **graphene nano-ribbons at THz**

❖ Future work:

- better understanding of the most **appropriate** surface conductivity model
- include **more nonlinear effects**:
    - Two Photon Absorption (trivial)
    - Carrier Effects, FCD & FCA
    - Four Wave Mixing
- more **complex graphene layouts** (bilayers, etc.) or biasing conditions
- more efficient **waveguide engineering**

Acknowledgments

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