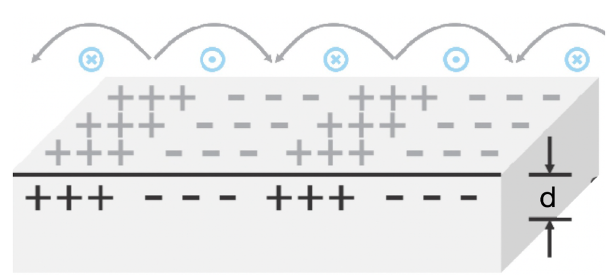
Plasmonics at the Macroscale and Microscale

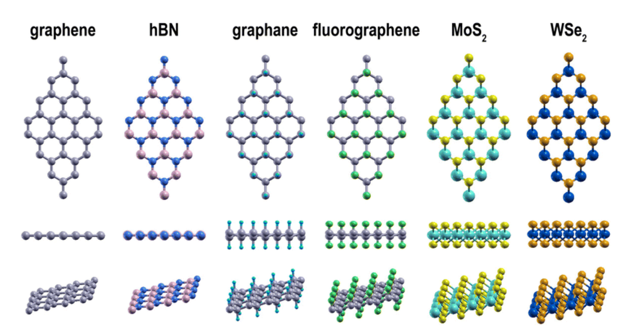
Darío Cruzado-Padró[1] Madison Sousa[2]
Mentor: Dionisios Margetis[3]
University of Puerto Rico, Mayagüez[1], University of Denver[2], University of Maryland, College Park[3]

Plasmonics in 2D Materials

- Plasmons are excitations due to the coupling of electrons with electromagnetism in conducting materials.
- Surface plasmons - electromagnetic waves confined between a dielectric and an atomically-thin material.

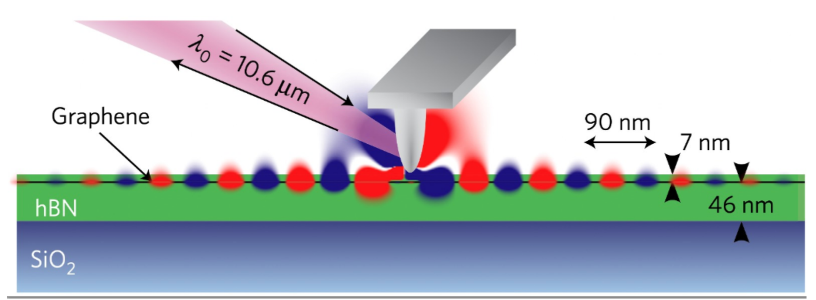


[Yoon,Yeung, Kim, Ham, 2014]



[Pollard,Clifford, Kim, Ham, 2017]

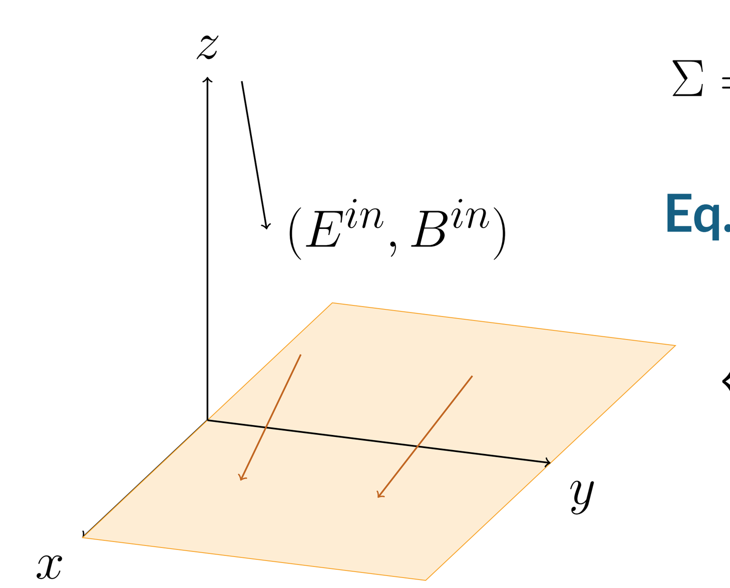
- Key property of surface plasmons: *Contraction of wavelength*



[Martín-Cano, et al.,2017]

- Surface plasmons have a wide range of applications: nanotechnology, spectroscopy, remote sensing, and optical imaging.

Geometry of the Problem



$$\Sigma = \{\mathbf{x} = (x, y, z) \in \mathbb{R}^3 \mid z = 0\}$$

Eq. for Physics of the Material:

$$\begin{cases} \partial_t \rho^\Sigma + \nabla \cdot \mathbf{J}^\Sigma = 0 \\ \mathbf{J}^\Sigma = -e\eta_0 \mathbf{v} \\ \partial_t \mathbf{v} = -\frac{e}{m}\mathbf{E}_\parallel \text{ (Classical)} \end{cases}$$

Dispersion Relation

- Relation between frequency and wavenumber.
- Can be affected by:
  - Geometry
  - Boundary Conditions (Physics of the material)

Dispersion Relation from a Classical View

- The classical model considers electrons to be point charges.

$$\begin{cases} \mathbf{E}(\mathbf{x}, t) \approx -\nabla\phi(\mathbf{x}, t) \\ -\Delta\phi = \frac{\rho_{\text{ex}}(x,y,z,t) + \rho^\Sigma(x,y,t)\delta(z)}{\epsilon_0} \end{cases}$$

Boundary Conditions:

$$\begin{cases} \phi : \text{ continuous on } \Sigma \\ \left.\frac{\partial\phi}{\partial z}\right|_{z=0^-}^{z=0^+} = \frac{-\rho^\Sigma}{\epsilon_0} \text{ on } \Sigma \end{cases}$$

Classical Approach

- The derivation of the Dispersion Relation is based on using Green's function and the Convolution Theorem for the Fourier Transform in x, y, t of the potential $\phi(\mathbf{x}, t)$.

$$\phi(\mathbf{x}, t) = G(\mathbf{x'}) \star \rho^\Sigma(\mathbf{x'}, t) = \frac{1}{4\pi\epsilon_0}\int_{\mathbb{R}^2} \frac{\rho^\Sigma(x', y', t)}{\sqrt{(x-x')^2 + (y-y')^2 + (z)^2}} dx'dy'$$

- Apply the Convolution Theorem for the Fourier transform of $\phi(\mathbf{x}, t)$

$$\widehat{\phi}(k_x, k_y, z, \omega) = \widehat{G}(k_x, k_y, z)\hat{\rho}^\Sigma(k_x, k_y, \omega)$$

- Calculate $G(\mathbf{x})$ such that $\Delta G(\mathbf{x}) = -\delta^3(\mathbf{x})$, and its Fourier transform

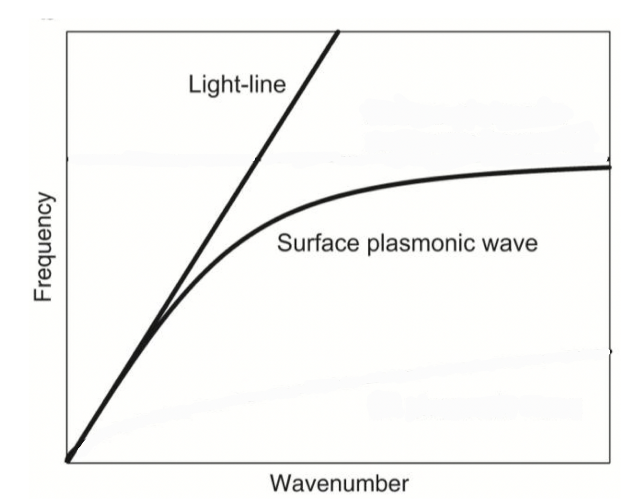
$$\widehat{G}(k_x, k_y, z) = \frac{1}{2\sqrt{k_x^2 + k_y^2}} e^{-\sqrt{k_x^2+k_y^2}|z|}$$

- Use the physics of the material to express the Fourier Transform of the surface charge density in terms of $\widehat{\phi}(k_x, k_y, z, \omega)$

$$\hat{\rho}^\Sigma(k_x, k_y, \omega) = \frac{e^2\eta_0}{\omega^2 m}(k_x^2 + k_y^2)\widehat{\phi}(k_x, k_y, \omega)$$

Classical Result:

$$\omega^2 = \frac{e^2\eta_0}{2m}|k|^{\frac{1}{2}}, \quad |k| = \sqrt{k_x^2 + k_y^2}$$



Problem in the Quantum Regime

How is the plasmon dispersion relation derived by coupling the Schrödinger equation (for the electron) with the electrostatic field?

Model with Schrödinger Dynamics

- The quantum model considers the wave fluctuations of the electron.

$$\begin{cases} \{\mathcal{H}_0 + \mathcal{H}_1\}\psi(\mathbf{x}, t) = i\partial_t\psi(\mathbf{x}, t) \text{ , } \mathbf{x} \in \mathbb{R}^3 \\ -\Delta\phi = \frac{\rho_e}{\epsilon} \end{cases}$$

$$\mathcal{H}_0 = -\frac{\hbar}{2m}\Delta - V_0\ell\delta(z), \ell : \text{ small length scale of confinement}$$

$$\mathcal{H}_1 = -e\phi(t, \mathbf{x})$$

$$\rho_e(\mathbf{x}, t) = -e\{|\psi(\mathbf{x}, t)|^2 - |\psi_0(\mathbf{x})|^2\}, \text{ where } \mathcal{H}_0\psi_0(\mathbf{x}) = \mu_0\psi_0(\mathbf{x})$$

Approach: Perturbation Theory

- Main Idea: Introduce a scattered wave function and linearize $\rho_e(\mathbf{x}, t)$.

$$(i\hbar\partial_t - \mathcal{H}_0)\psi(\mathbf{x}, t) = \mathcal{J}(\mathbf{x}, t) = \mathcal{H}_1\psi(\mathbf{x}, t)$$

  - *Perturbation*: $\psi(\mathbf{x}, t) = \psi_0(\mathbf{x})e^{-\frac{iE_0t}{\hbar}} + \psi_s(\mathbf{x}, t), \quad |\psi_s(\mathbf{x}, t)| << |\psi_0(\mathbf{x}, t)|$
  - *Linearization*: $\rho_e(\mathbf{x}, t) \approx \psi_0(\mathbf{x}, t)^*\psi_s(\mathbf{x}, t) + \psi_0(\mathbf{x}, t)\psi_s(\mathbf{x}, t)^*$

- Using the fact that $\psi_s(\mathbf{x}, t) = G(t, \mathbf{x}, t', \mathbf{x'}) \star \mathcal{J}(\mathbf{x}, t)$,

$$\psi_s(\mathbf{x}, t) = -\int_{\mathbb{R}^3}\int_{-\infty}^{t} G(t-t', x-x', y-y', z; z')\mathcal{J}(t, \mathbf{x}) \, dt'd\mathbf{x}'$$

- $G(\cdot)$ is the propagator for the Schrödinger equation.

$$(i\hbar\partial_t - \mathcal{H}_0)G(t, \mathbf{x}, t', \mathbf{x'}) = -\delta(\mathbf{x}\text{-}\mathbf{x'})\delta(t-t')$$

$$\widehat{G}(\cdot) = \begin{cases} \frac{m}{\hbar^2}\frac{1}{\beta}[e^{-\beta|z-z'|} + \frac{m}{\hbar^2}V_0\ell(\beta - \frac{m}{\hbar^2}V_0\ell)^{-1}e^{\beta(|z|+|z'|)}] & zz' > 0 \\ \frac{m}{\hbar^2}(-\beta - \frac{m}{\hbar^2}V_0\ell)^{-1}e^{\beta(|z|+|z'|)} & zz' < 0 \end{cases}$$

where $\beta = \sqrt{\frac{2m}{\hbar^2}}(\omega_* - \omega)^{\frac{1}{2}}, \omega_* = \frac{\hbar}{2m}(k_x^2 + k_y^2)$

- Ansatz: $\psi_s(\mathbf{x}, t) \approx f^+(z)e^{i(q_x x + q_y y)}e^{i(\alpha^2 - \Omega)} + f^-(z)e^{-i(q_x x + iq_y y)}e^{i(\alpha^2 + \Omega)}$

$$f^\pm(z) = \frac{-(Ce)^2}{2}\int_{\mathbb{R}} \widehat{G}(E_3 \pm \Omega, q_x, q_y, z; z')e^{-\alpha z'}\mathcal{F}_{op}\left[e^{-\alpha z'}(f^\pm + f^{\mp *})\right](z') \, dz'$$

Dispersion Relation: $\Omega^2 \approx \frac{\hbar}{2m}|q|^2 + \frac{e^2\eta_0}{2m}|q|^{\frac{1}{2}}$

Conclusion

- We obtained linear corrections due to the kinetic energy of the plane waves in x and y.
- These corrections are caused by the wave-particle duality of the electron.

dario.cruzado@upr.edu

Acknowledgements: This project was supported by the National Science Foundation as part of the 2022 Mathematics REU at the University of Maryland.

madi.sousa@du.edu