CF-UM-UP

Universidade do Minho
Escola de Ciências

Quantum electrodynamics with polaritons in 2D materials

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in collaboration with

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Quantum polaritons

- Description of light-matter interactions at the nanoscale requires quantization of Eletromagnetic (EM) field
- **Polaritons**: quasi-particles resulting from hydridization between photons and matter excitations
- EM Dyadic Green's function:
  - Includes quantum effects ✔
  - Muddles physical interpretation ✘

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  - Muddles physical interpretation ✘
- What we want:

Quantum description of polaritonic modes

The problem: quantization of EM field in dispersive media

- Canonical quantization of EM field in static dielectric media, poses no problem.
- Hamiltonian in the Weyl gauge $(\phi = 0)$:

$$H_{\mathrm{EM}} = \int d^3\mathbf{r} \left(\frac{1}{2\epsilon_0} \mathbf{\Pi} \cdot \boldsymbol{\epsilon}^{-1} \cdot \mathbf{\Pi} + \frac{1}{2\mu_0} (\nabla \times \mathbf{A})^2 \right)$$

$$\mathbf{\Pi} = \epsilon_0 \boldsymbol{\epsilon} \partial_t \mathbf{A}$$

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$$\mathbf{P}(\omega) = \epsilon_0 \boldsymbol{\chi}(\omega) \mathbf{E}(\omega) \quad \Longrightarrow \quad \mathbf{P}(t) = \epsilon_0 \int dt' \boldsymbol{\chi}(t, t') \mathbf{E}(t')$$

Dispersion = Retarded response

Commutation relations ?

The solution: auxiliar degrees of freedom for matter

- We can make the theory local in time, if we explicitly include degrees of freedom that describe matter
- These can be ficticious: we only require that they reproduce $\epsilon(\omega)$

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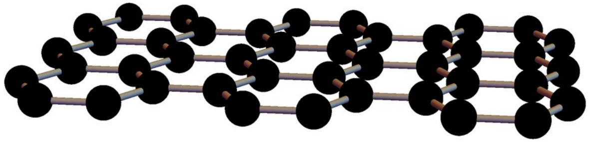
Example:

Hydrodynamic **conductivity of graphene**:



$$\boldsymbol{\sigma}_g(\omega, \mathbf{q}) = D\frac{i\omega}{\omega^2 - \beta^2 q^2}\frac{\mathbf{q}\otimes\mathbf{q}}{q^2} + D\frac{i}{\omega}\left(\mathbf{1} - \frac{\mathbf{q}\otimes\mathbf{q}}{q^2}\right)$$

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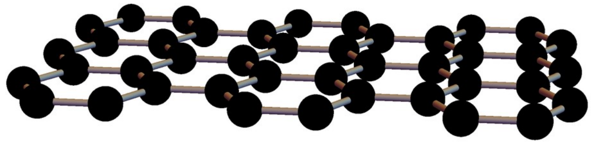

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Same conductivity as for a **charged fluid** described by the Lagrangean:

$$\mathcal{L}_{\text{hyd}} = \frac{1}{2}n_0 m\left(\partial_t \boldsymbol{v}\right)^2 - \frac{1}{2}n_0 m\beta^2\left(\nabla\cdot\boldsymbol{v}\right)^2 - e n_0 \partial_t \boldsymbol{v}\cdot\mathbf{A}$$

Current density: $\mathbf{J} = -e n_0 \partial_t \boldsymbol{v}$

Drude weight: $D = e^2 n_0/m$

Quantization of EM field + matter

- The Hamiltonian for **EM field + matter** is **local in time**!
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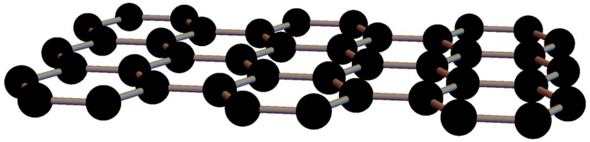

- For graphene example:

$$\begin{bmatrix} \hat{\mathbf{A}}(t,\mathbf{r}) \\ \hat{\boldsymbol{v}}(t,\mathbf{x}) \end{bmatrix} = \sum_{\mathbf{q},\lambda} \sqrt{\frac{\hbar}{2\epsilon_0 \omega_{\mathbf{q},\lambda} A L_{\mathbf{q},\lambda}}} \begin{bmatrix} \mathbf{A}_{\mathbf{q},\lambda}(z) \\ \boldsymbol{v}_{\mathbf{q},\lambda} \end{bmatrix} e^{i\mathbf{q}\cdot\mathbf{x}} \hat{a}_{\mathbf{q},\lambda} + \text{h.c.}$$

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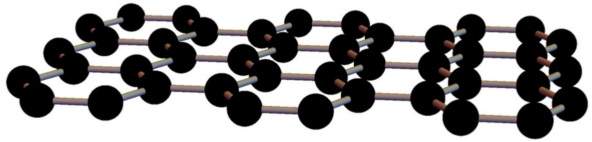

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Classical solutions

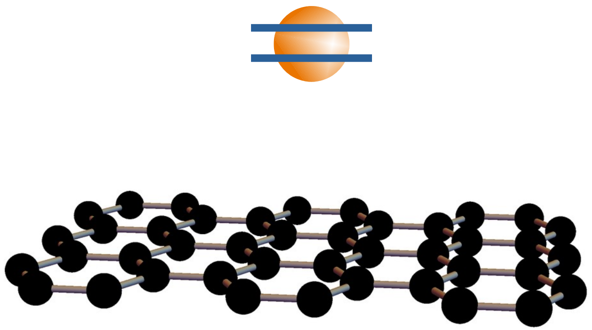
- Non-standard normalization factor! **(induced by dispersive matter)**

$$L_{\mathbf{q},\lambda} = \int d^3\mathbf{r} \mathbf{A}^*_{\mathbf{q},\lambda}(\mathbf{r}) \cdot \left[\boldsymbol{\epsilon}(\omega_{\mathbf{q},\lambda},\mathbf{r}) + \omega^2_{\mathbf{q},\lambda} \frac{\partial \boldsymbol{\epsilon}(\omega_{\mathbf{q},\lambda},\mathbf{r})}{\partial(\omega^2)} \right] \cdot \mathbf{A}_{\mathbf{q},\lambda}(\mathbf{r})$$

General result! (ignoring losses)

Application 1: Purcell effect due to emission of graphene plamons

- **Purcell effect**: decay rate of quantum emitter depends on **local density** of EM modes



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Phys. Rev. A 101, 033817 (2020)

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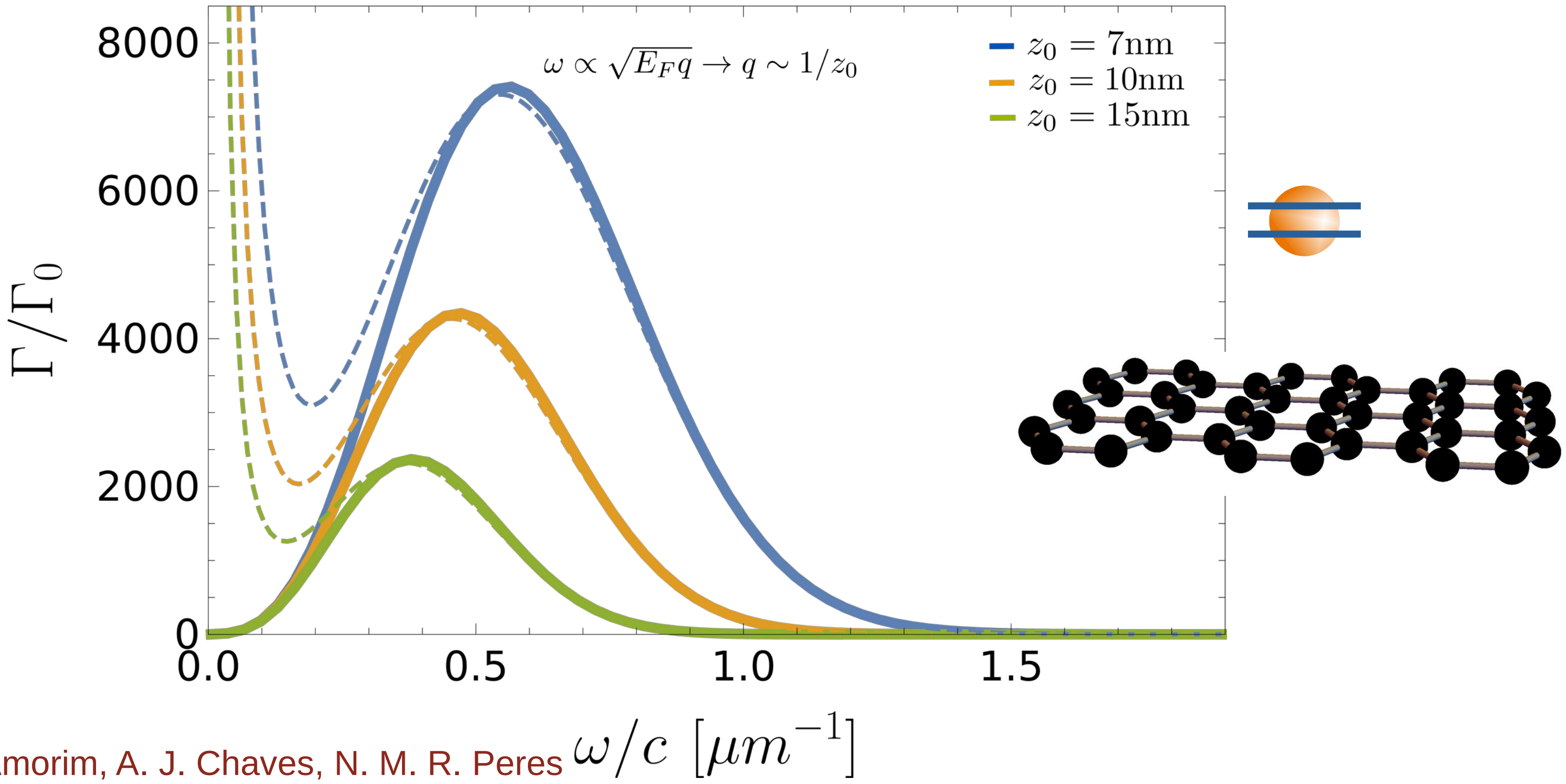

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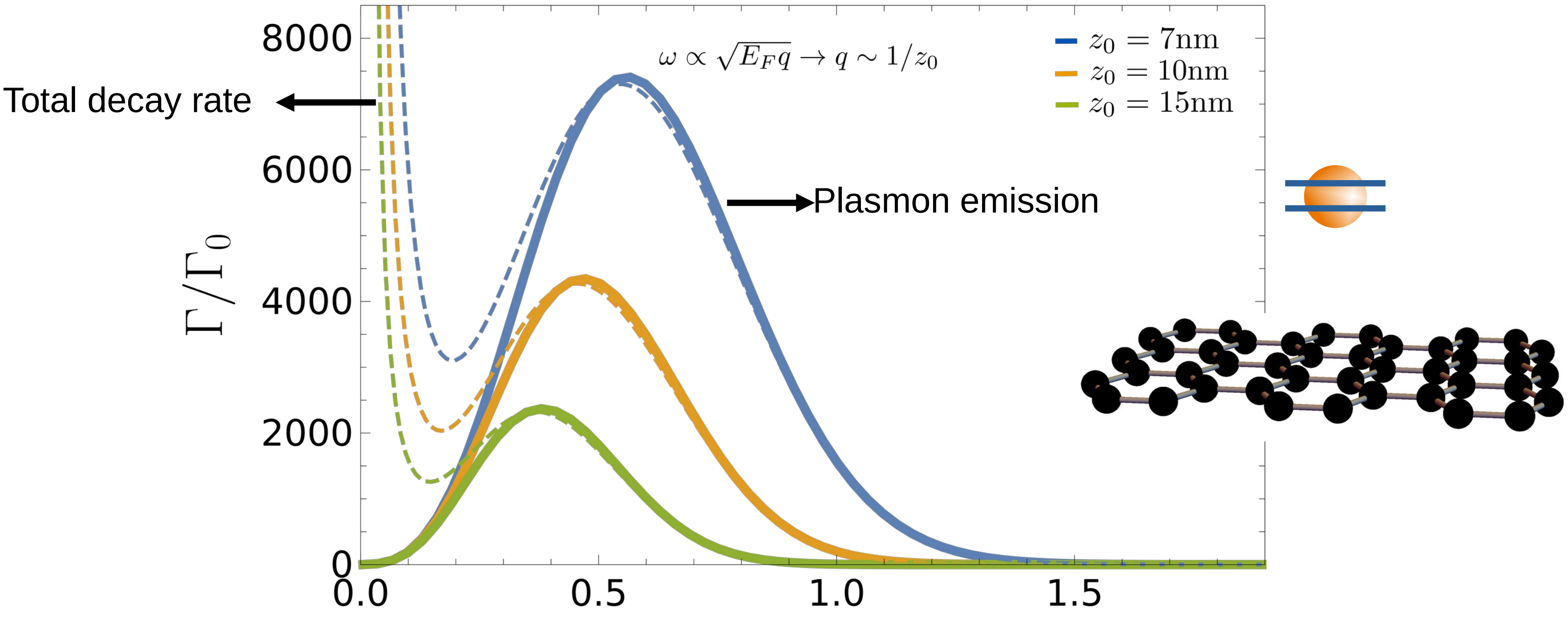

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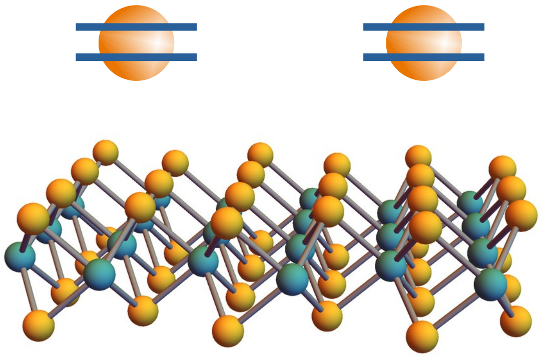
Application 2: superradiance mediated by exciton-polaritons in TMDs

- Nitrogen-vacancy color centers placed on top of TMD monolayer:



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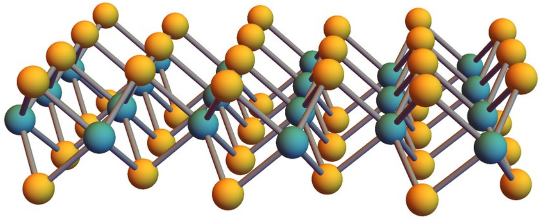

TMD with exciton ~ 1.95 eV

$$\sigma(\omega) = -id\epsilon_0\omega\left[\chi_{\mathrm{bg}} - f_{\mathrm{ex}}\frac{\omega_{\mathrm{ex}}^2}{\omega^2 + i\omega\gamma - \omega_{\mathrm{ex}}^2}\right]$$

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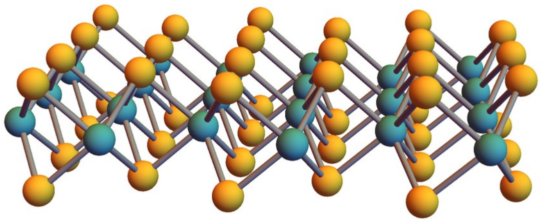

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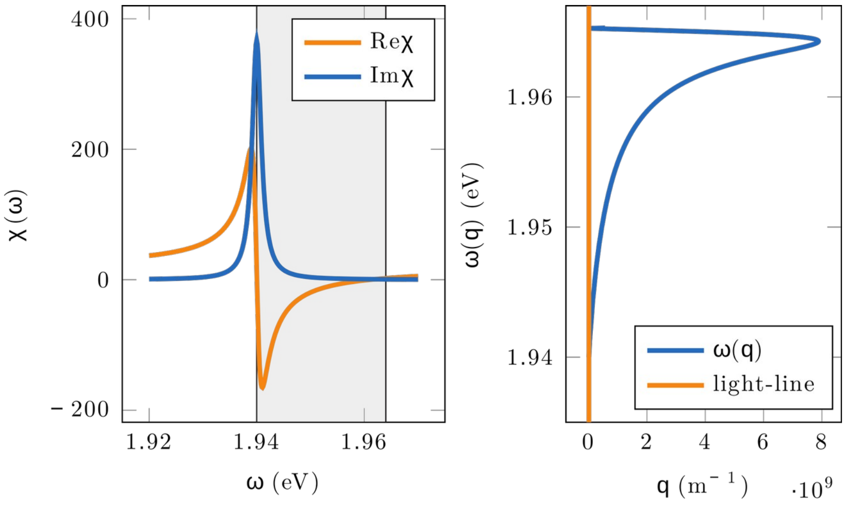

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- Dynamics for NV centers:

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}\left[\frac{1}{2}\sum_{i=1}^{2}\left(\hbar\omega_0+\Delta_i\right)\sigma_i^z + \sum_{i\neq j} g_{ij}\sigma_i^+\sigma_j^-, \rho\right] + \frac{1}{\hbar}\sum_{ij}\gamma_{ij}\left(\sigma_j^-\rho\sigma_i^+ - \frac{1}{2}\left\{\sigma_i^+\sigma_j^-, \rho\right\}\right)$$

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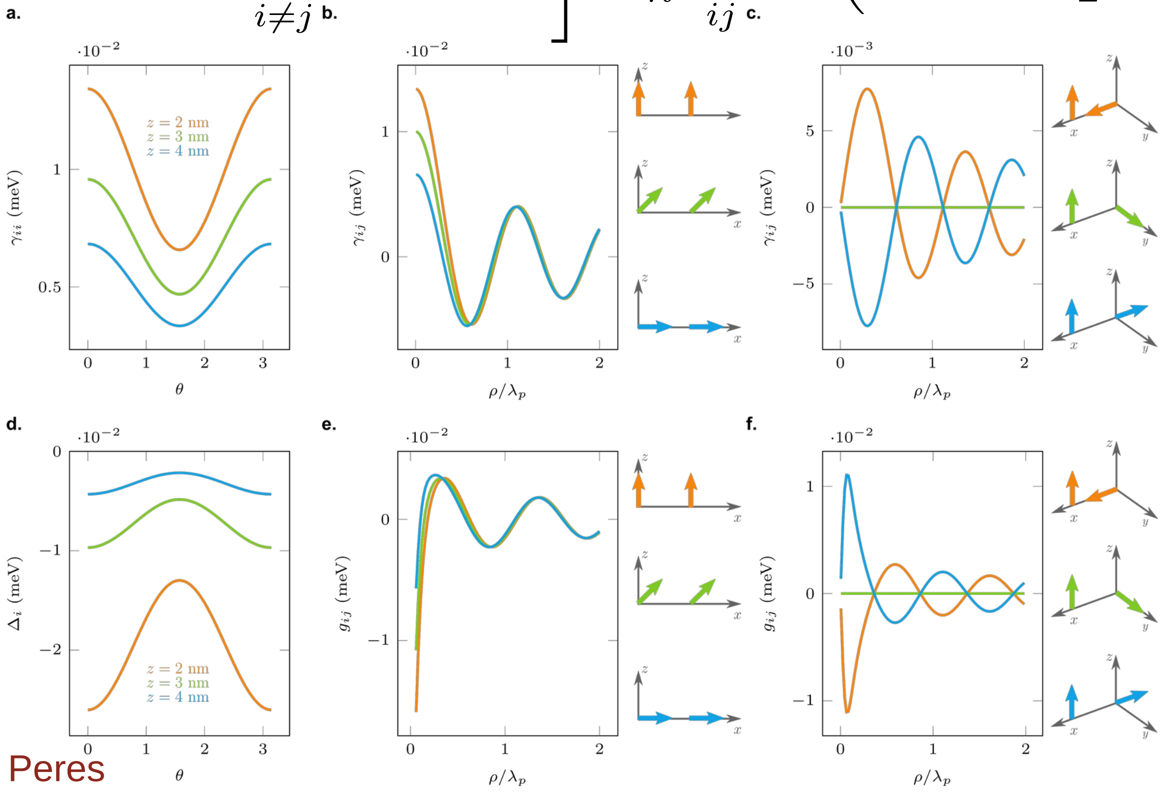

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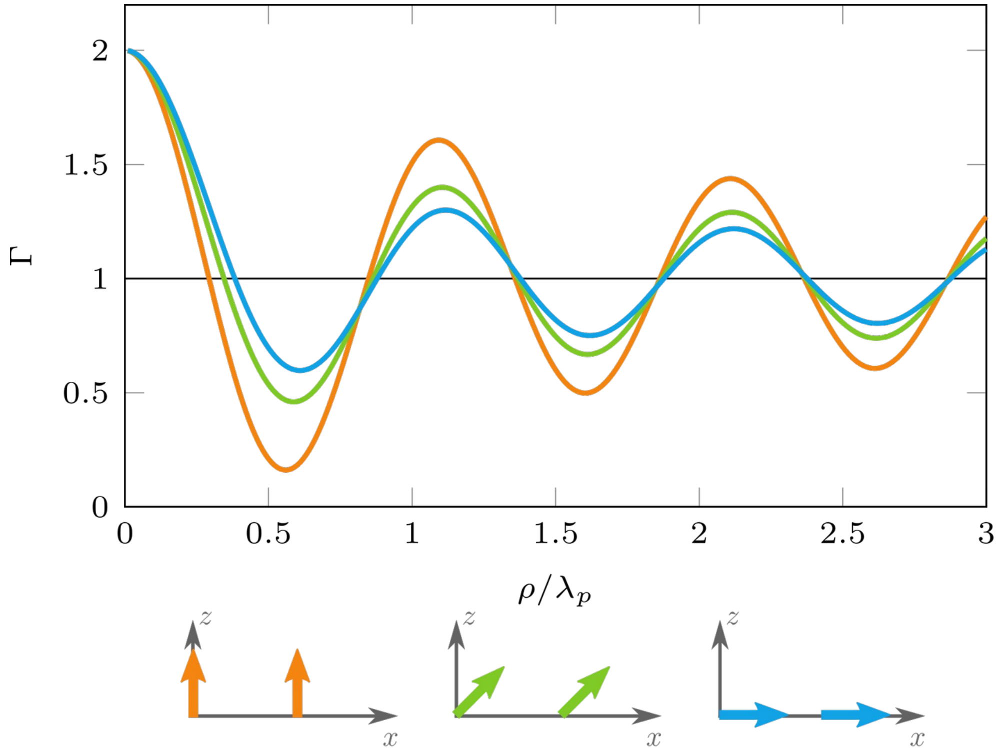
$\lambda_p = 37$ nm

Application 2: superradiance mediated by exciton-polaritons in TMDs

- Superradiance if:

$$\Gamma = \frac{\gamma_{11} + \gamma_{22} + \gamma_{12} + \gamma_{21}}{\gamma_{11} + \gamma_{22}} > 1$$



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Enhanced emission radiation for seperations $\simeq n\lambda_p$

Conclusions

- Canonical quantization of polaritons allows to extract polaritonic contribution from different quantum electrodynamic processes
- Method can be applied for different kinds of polaritons: plasmon-polaritons, exciton-polariton, phonon-polaritons
- Approach excludes material losses, but can be included in a perturbative way

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Thank you for your attention

Extra 1: Mode normalization and response to external current

- EM field generated by source current:

$$\nabla \times \nabla \times \mathbf{A}(\omega,\mathbf{r}) + \frac{\omega^2}{c^2}\boldsymbol{\epsilon}(\omega)\mathbf{A}(\omega,\mathbf{r}) = \mu_0 \mathbf{J}(\omega,\mathbf{r})$$

- $\mathbf{J}(\omega,\mathbf{r})$ peaked at $\omega \simeq \omega_\lambda$

$$\nabla \times \nabla \times \mathbf{A}_\lambda(\mathbf{r}) + \frac{\omega_\lambda^2}{c^2}\boldsymbol{\epsilon}(\omega_\lambda)\mathbf{A}_\lambda(\mathbf{r}) = 0$$

- If we look for solution of the form $\mathbf{A}(\omega,\mathbf{r}) \propto \mathbf{A}_\lambda(\mathbf{r})$

$$\mathbf{A}(\omega,\mathbf{r}) \simeq \int d^3\mathbf{r}' \frac{1}{\epsilon_0 L_\lambda} \frac{\mathbf{A}_\lambda(\mathbf{r})\mathbf{A}_\lambda^*(\mathbf{r}')}{\omega^2 - \omega_\lambda^2} \mathbf{J}(\omega,\mathbf{r}')$$

$$L_\lambda = \int d^3\mathbf{r}' \mathbf{A}_\lambda^*(\mathbf{r}') \left[\boldsymbol{\epsilon}(\omega_\lambda) + \omega_\lambda^2 \frac{\partial \boldsymbol{\epsilon}(\omega_\lambda)}{\partial\left(\omega^2\right)}\right] \mathbf{A}_\lambda(\mathbf{r}')$$

Extra 2: Lindblad equation

$$\frac{\partial\rho(t)}{\partial t} = -\frac{i}{\hbar}\left[\sum_i \frac{1}{2}\left(\epsilon_0+\Delta_i\right)\sigma_i^z + \sum_{i\neq j} g_{ij}\sigma_i^+\sigma_j^-, \rho(t)\right]$$

$$+\frac{1}{\hbar}\sum_{i,j}\gamma_{ij}\left(\sigma_j^-\rho(t)\sigma_i^+ - \frac{1}{2}\left\{\sigma_i^+\sigma_j^-, \rho(t)\right\}\right)$$

$$+\frac{1}{\hbar}\sum_{i,j}\tilde{\gamma}_{ij}\left(\sigma_j^+\rho(t)\sigma_i^- - \frac{1}{2}\left\{\sigma_i^-\sigma_j^+, \rho(t)\right\}\right)$$

$$\Delta_i = P\int\frac{d\nu}{2\pi}\left[1+2b(\nu)\right]\frac{\mu_{i,\alpha}^* A_{\alpha\beta}\left(\nu;\mathbf{r}_i,\mathbf{r}_i\right)\mu_{i,\beta}}{\omega_0-\nu}$$

$$g_{ij} = \mu_{i,\alpha}^* D_{\alpha\beta}(\omega_0;\mathbf{r}_i,\mathbf{r}_j)\mu_{j,\beta},$$

$$\gamma_{ij} = \left[1+b(\omega_0)\right]\mu_{i,\alpha}^* A_{\alpha\beta}\left(\omega_0;\mathbf{r}_i,\mathbf{r}_j\right)\mu_{j,\beta},$$

$$\tilde{\gamma}_{ij} = b(\omega_0)\mu_{j,\alpha}^* A_{\alpha\beta}\left(\omega_0;\mathbf{r}_j,\mathbf{r}_i\right)\mu_{i,\beta},$$

$$D_{\alpha\beta}^R(\omega;\mathbf{r}_i,\mathbf{r}_j) = D_{\alpha\beta}(\omega;\mathbf{r}_i,\mathbf{r}_j) - \frac{i}{2}A_{\alpha\beta}(\omega;\mathbf{r}_i,\mathbf{r}_j)$$