Quantum light-matter interactions in low-dimensional materials

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THESIS DEFENSE

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Outline

- **Introduction**
- **Spontaneous Emission (SE)**
  - One-photon SE and the Purcell effect;
  - Two-quanta SE (TQSE);
  - TQSE in one-dimensional carbon nanostructures;
  - TQSE in atomically thin plasmonic nanostructures.
- **Casimir effect**
  - Casimir effect and the Lifshitz formula;
  - Casimir forces in the flatland;
  - Photo-induced phase transitions and quantum Hall physics in the Casimir force.
- **Final remarks**

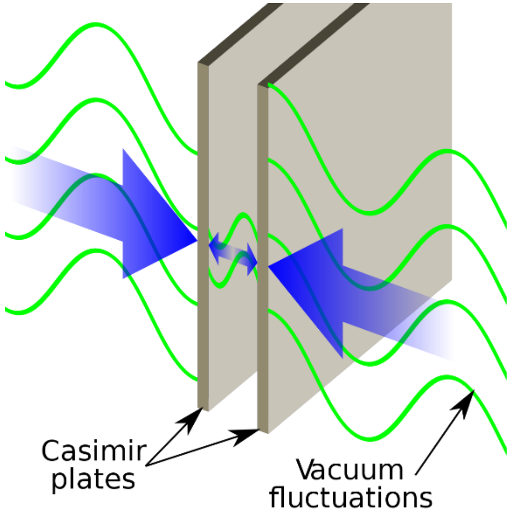
Introduction

Spontaneous emission and dispersive interactions

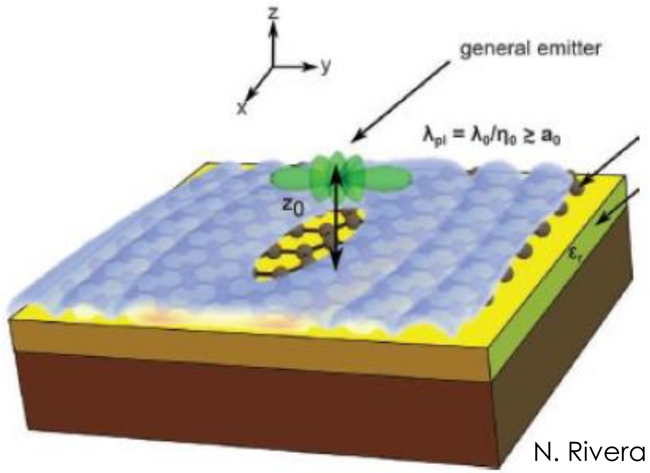
Spontaneous emission

London Van der Waals forces

Casimir forces

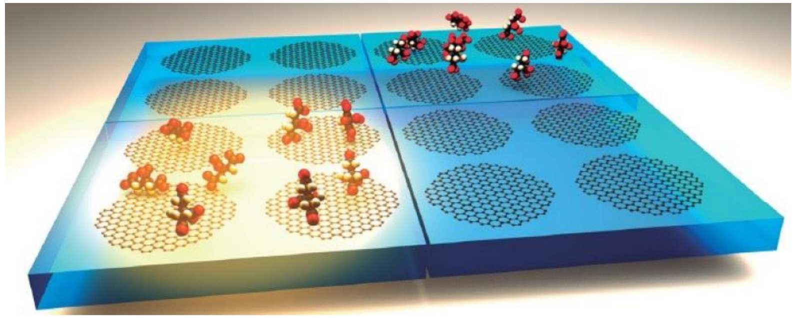


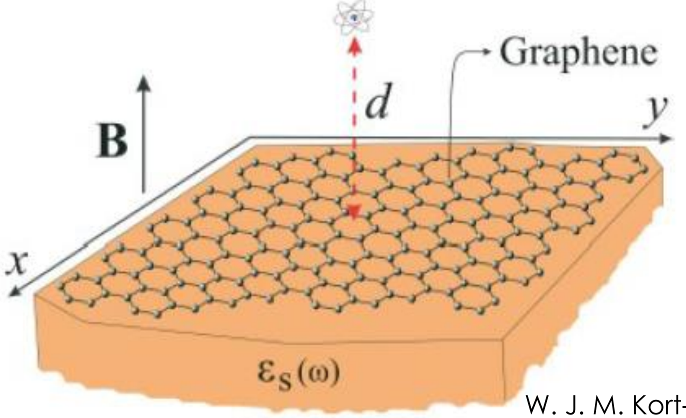
Low-dimensional materials



N. Rivera *et. al*, Science 353 6296 (2016)

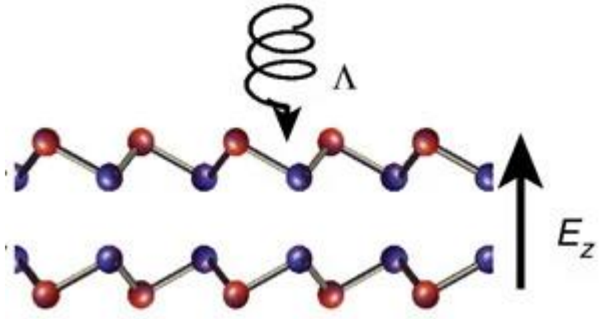
- **Unusual electromagnetic properties.**
  - Highly confined plasmon-polaritons.
  - Non-trivial topological features.


L. Zundel and A. Manjavacas, *ACS Photonics* **4 (7)** 1831 (2017)



W. J. M. Kort-Kamp *et. al.*, *PRB* **92** 205415 (2015)

- **Tunable parameters** for efficient control of quantum light-matter interactions.
  - Intrinsic: geometry, # of charge carriers.
  - External: electric and magnetic fields.

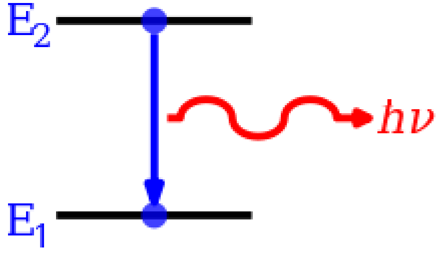


P. Rodriguez-Lopez *et. al.*, *Nat. Comm.* **8**, 14699 (2017)

Spontaneous Emission

One-photon SE

- An **excited atom**, even when isolated, **decays** to its fundamental state.



- Phenomenon induced by **quantum vacuum fluctuations**.
- Quantum electrodynamics (QED): excited atom + zero photons is not a stationary state of the atom-field system.

Purcell effect

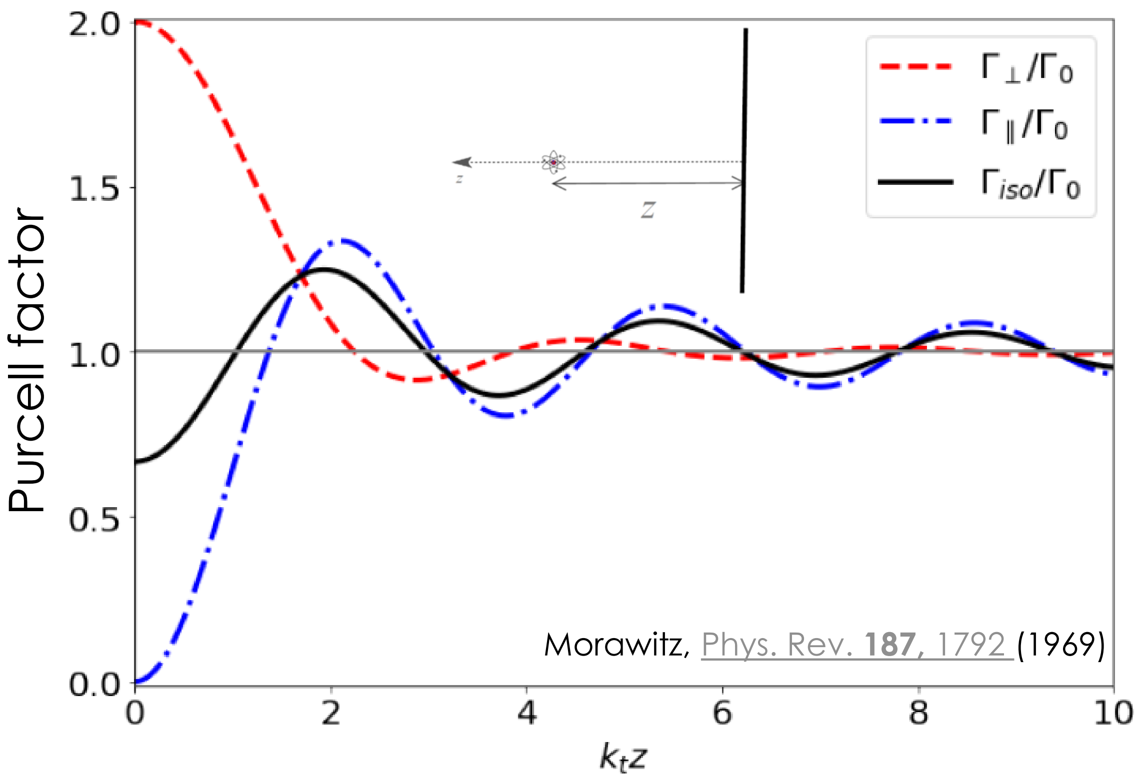
- **E.M. Purcell** (**1946**): Bodies in the vicinities of an emitter change its SE rate.

- Reason: The presence of bodies affects the **boundary conditions** (BC) on the electromagnetic field.

$$\Gamma(\mathbf{r}) = \frac{\pi}{\epsilon_o \hbar} \sum_\alpha \omega_\alpha |\mathbf{d}_t \cdot \mathbf{A}_\alpha(\mathbf{r})|^2 \delta(\omega_\alpha - \omega_t).$$

- It can be shown that the SE rate is proportional to the local density of states (**LDOS**) of the electromagnetic field.

Purcell effect



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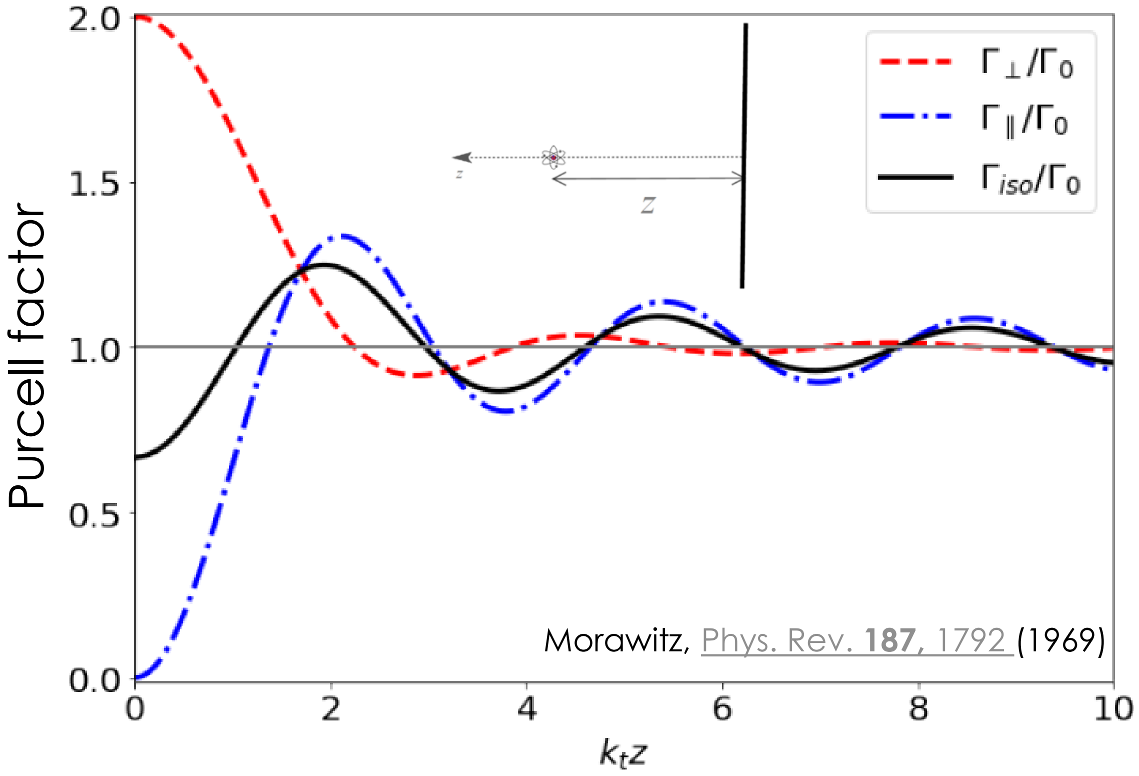
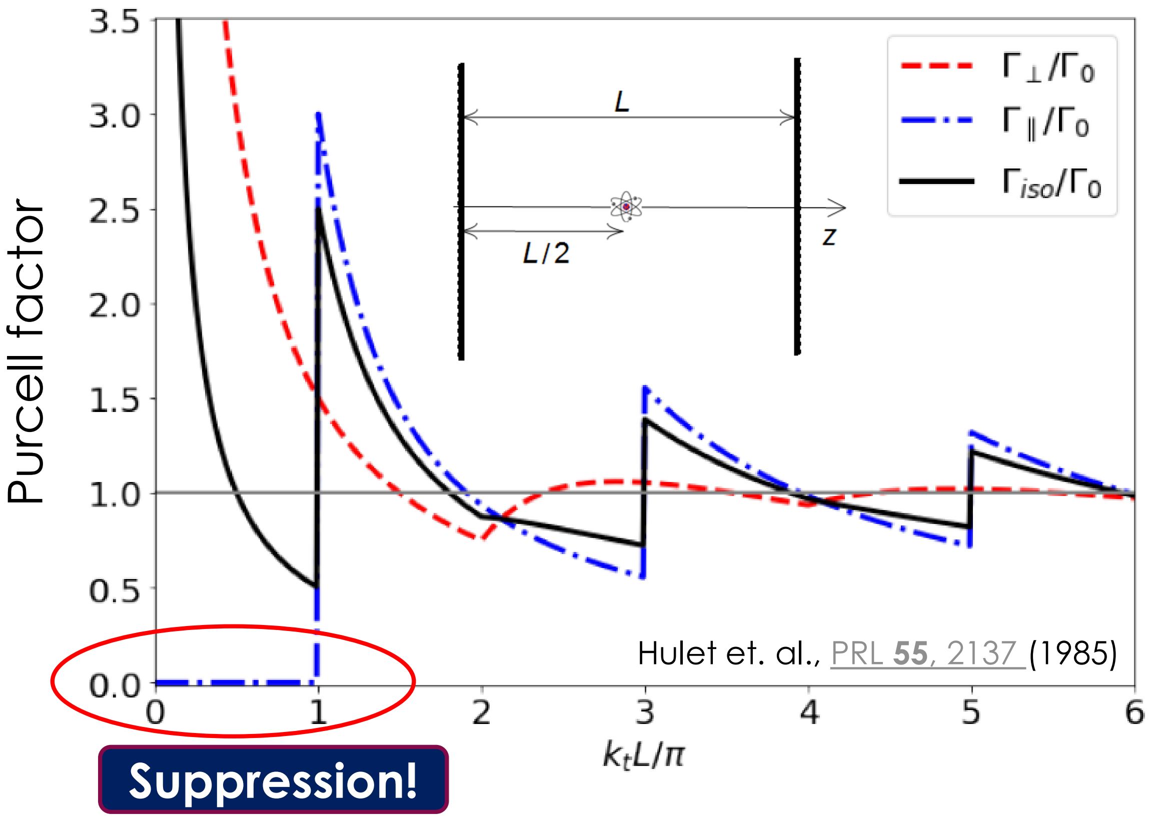
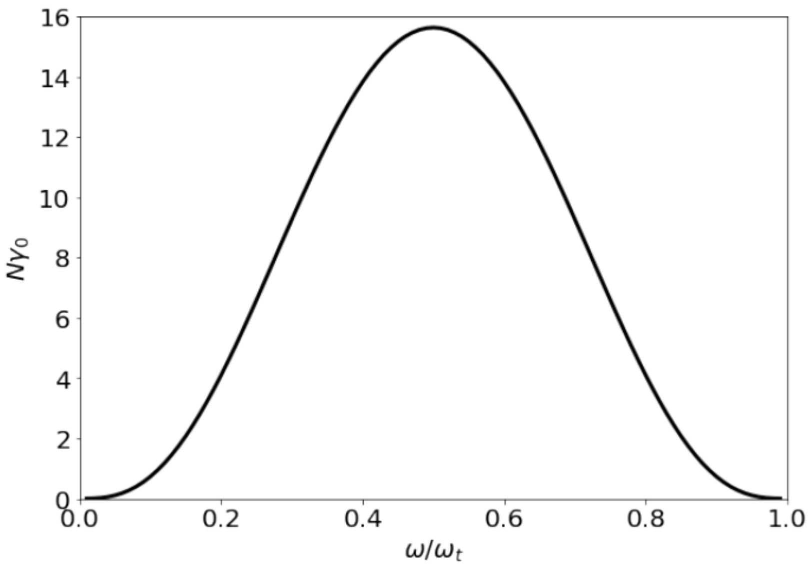
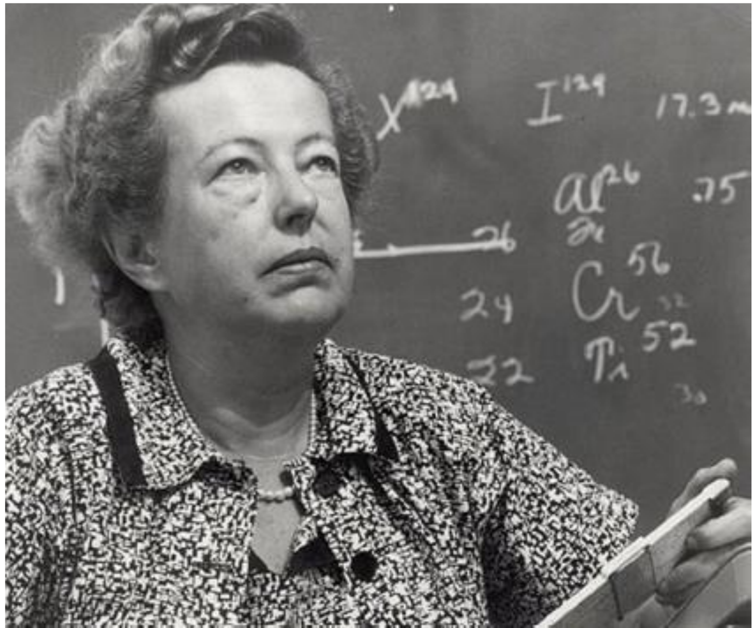




Two-photon spontaneous emission

- **Second-order** process in perturbation theory (**Maria Göppert-Mayer**, 1931).
  - Atoms usually decay by one-photon spontaneous emission.
- **Broadband spectrum** of emission.





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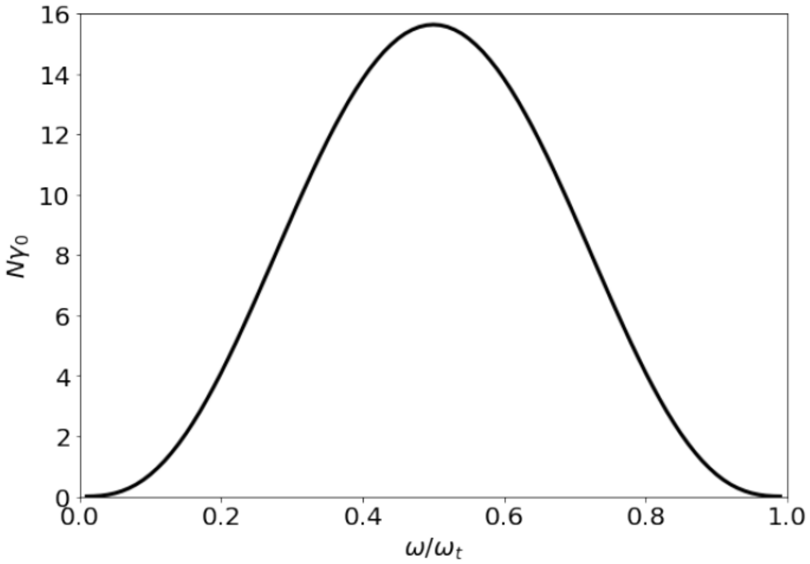
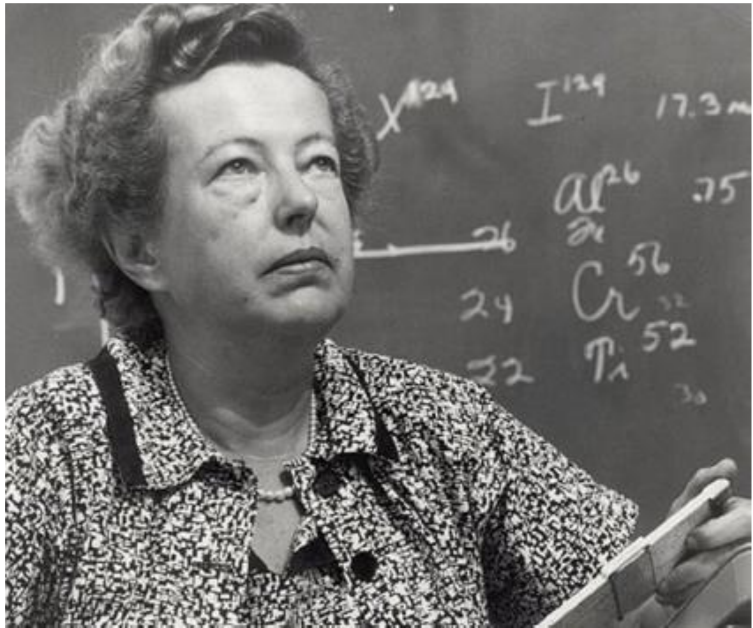
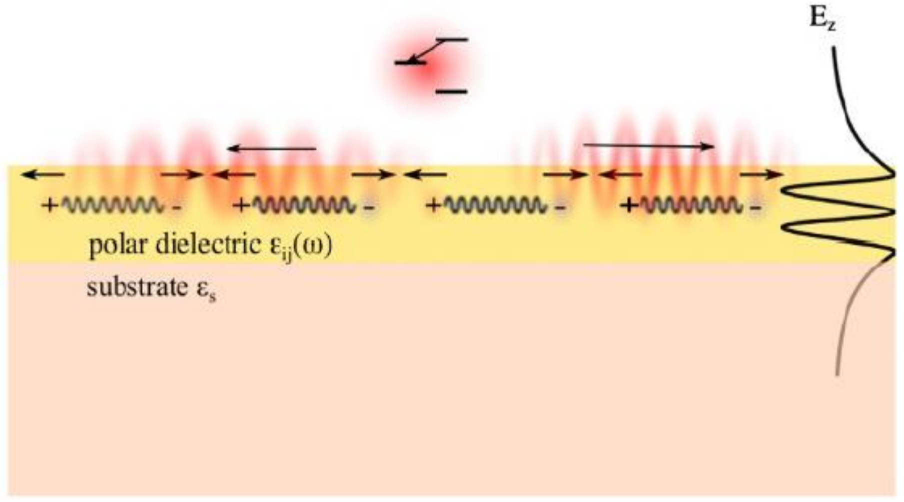
- **Broadband spectrum** of emission.
- Explains the emission spectrum of planetary nebulae.
  - 2s → 1s transition in He+.

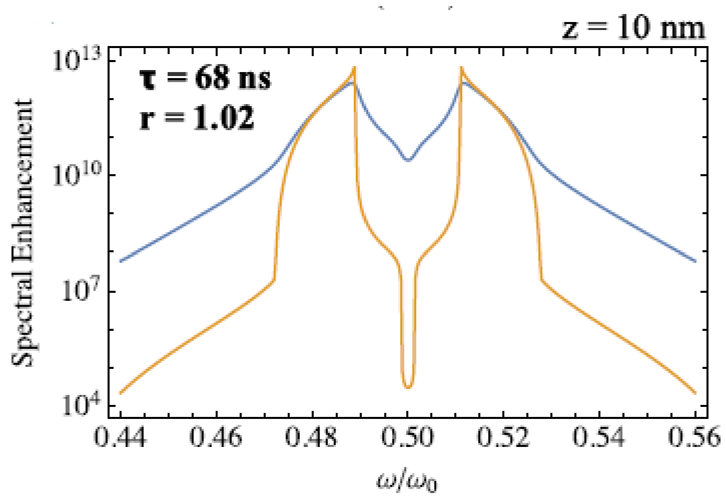




Why going beyond one-photon SE?

- TQSE is a possible source of **entangled photons**. → $$|\psi\rangle = \int d\omega c(\omega)|\omega\rangle|\omega_t - \omega\rangle$$
- TQSE **can be dominant** in some particular scenarios.
  - Polar dielectrics: coupling with optical phonon-polaritons.





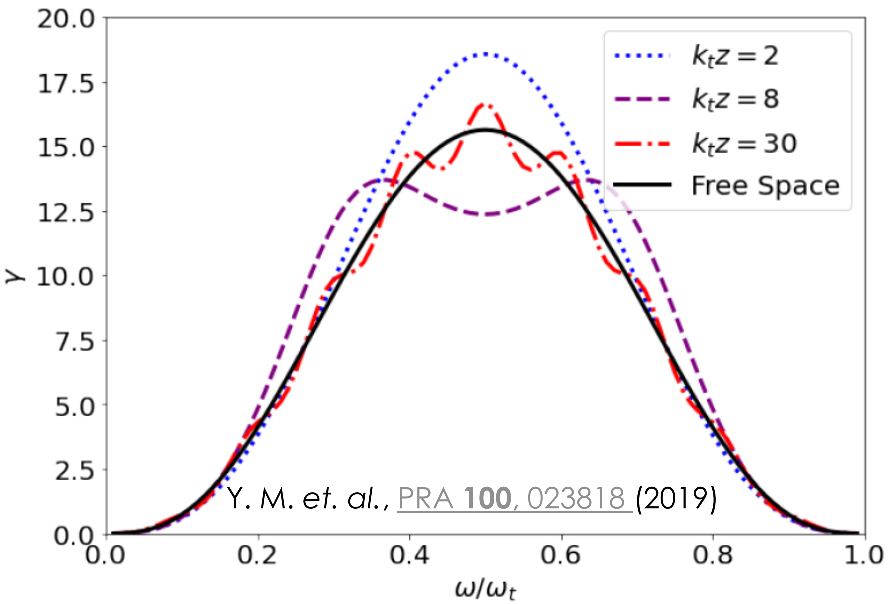
N. Rivera *et. al.*, PNAS **114**, 13607 (2017)

TQSE Purcell effect

- TQSE can be calculated with second order Fermi's Golden rule.
  - Using a **field modes** approach. → $\Gamma(\mathbf{r}) = \frac{\pi}{4\epsilon_0^2\hbar^2}\sum_{\alpha,\alpha'}\omega_\alpha\omega_{\alpha'}|\mathbf{A}_\alpha(\mathbf{r})\cdot\mathbb{D}(\omega_\alpha,\omega_{\alpha'})\cdot\mathbf{A}_{\alpha'}(\mathbf{r})|^2\delta(\omega_\alpha+\omega_{\alpha'}-\omega_t).$
  - Using a **Green's function** approach.

$$\mathbb{D}(\omega_\alpha,\omega_{\alpha'}) := \sum_m\left[\frac{\mathbf{d}_{im}\mathbf{d}_{mf}}{\omega_{im}-\omega_\alpha}+\frac{\mathbf{d}_{mf}\mathbf{d}_{im}}{\omega_{im}-\omega_{\alpha'}}\right]$$

An atom close to a perfect mirror



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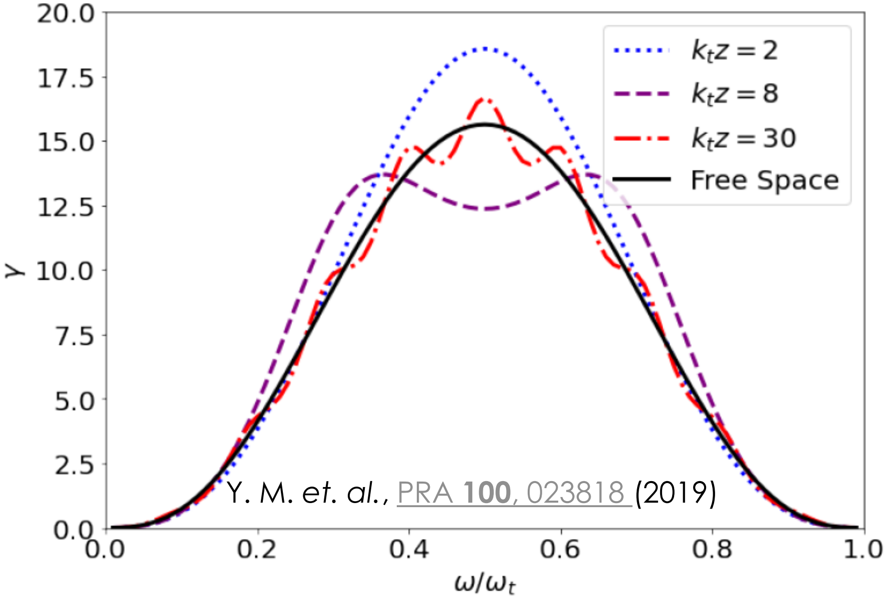
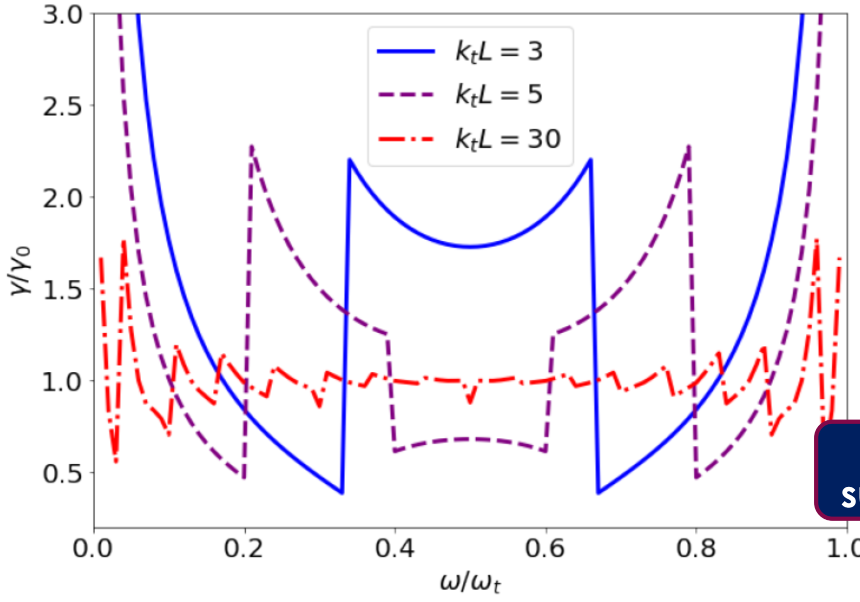


Y. M. *et. al.*, PRA **100**, 023818 (2019)

An atom between two perfect mirrors



No suppression

TQSE Purcell effect

- **Key result:** One equation to rule them all!!

TQSE rate

$$\Gamma = \int_0^{\omega_t} d\omega \gamma_0(\omega) \sum_{a,b} t_{ab}(\omega) P_a(\mathbf{r}, \omega) P_b(\mathbf{r}, \omega_t - \omega)$$

TQSE Purcell effect

- **Key result:** One equation to rule them all!!

$$t_{ab}(\omega) = |\mathbb{D}_{ab}(\omega, \omega_t - \omega)|^2 / |\mathbb{D}(\omega, \omega_t - \omega)|^2$$

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TQSE rate

Intrinsic properties of the emitter

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Free-space spectrum

TQSE Purcell effect

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Free-space spectrum

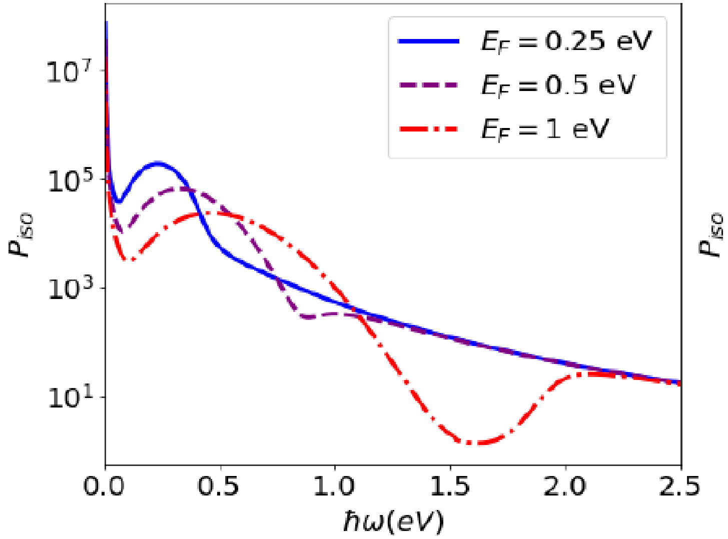
Purcell factors / normalized one-photon SE rates

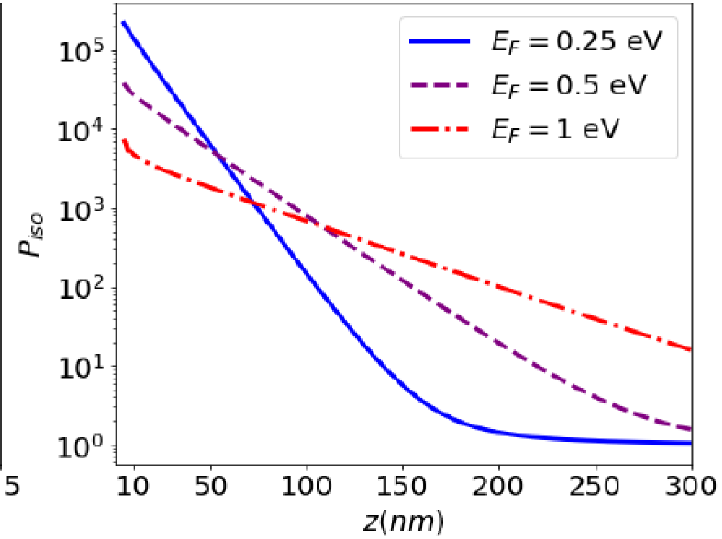
Y. M. *et. al.*, PRA **100**, 023818 (2019)

Spontaneous emission near a graphene sheet

- **Plasmon-emitter coupling** significantly enhances the SE.
  - SE rate can be tuned by changing the Fermi energy.
- Exponential decay with the distance.
  - Plasmons are evanescent modes.

One-plasmon isotropic Purcell factor





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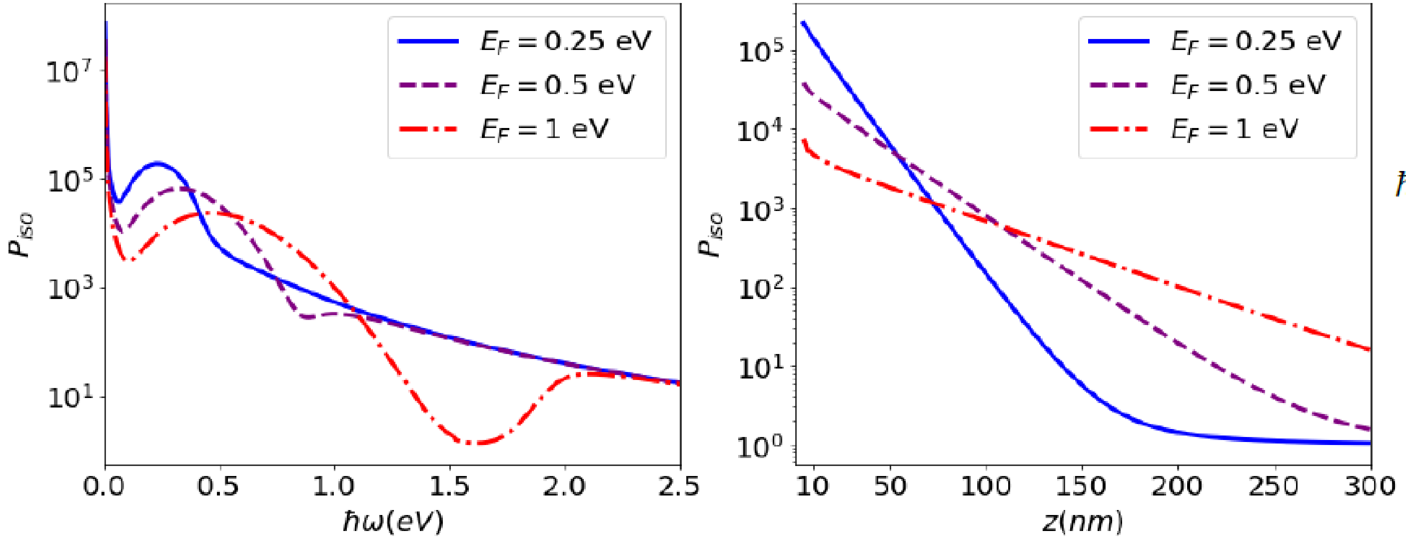
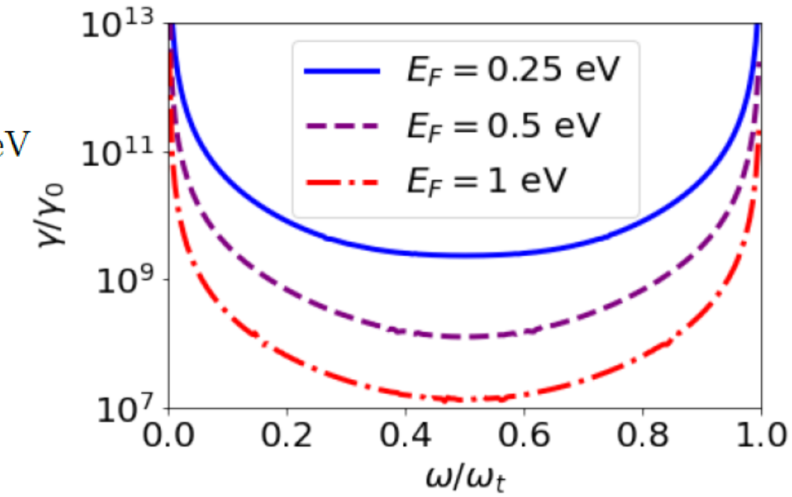


$\hbar\omega_t \approx 166$ meV

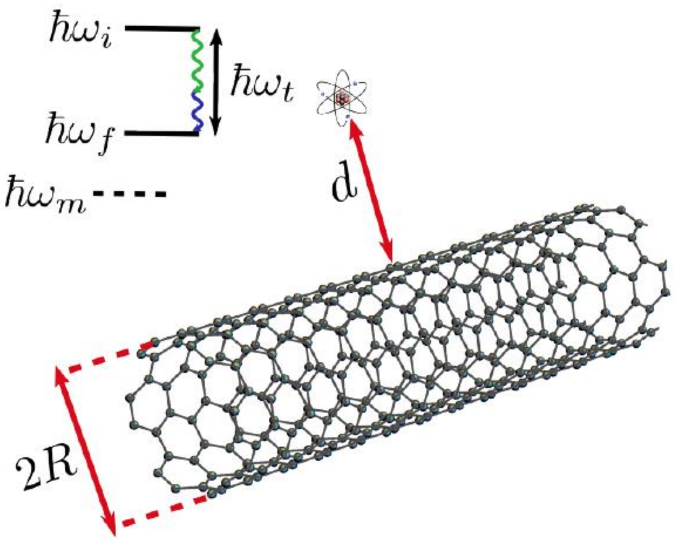
z = 10nm
QE ~ 2%

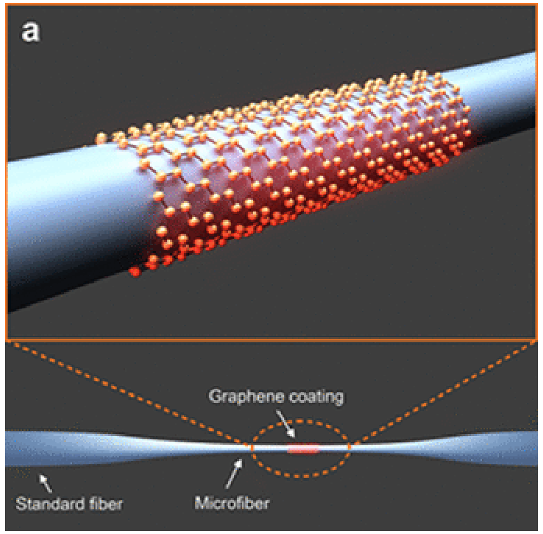
Two-plasmon SE spectrum

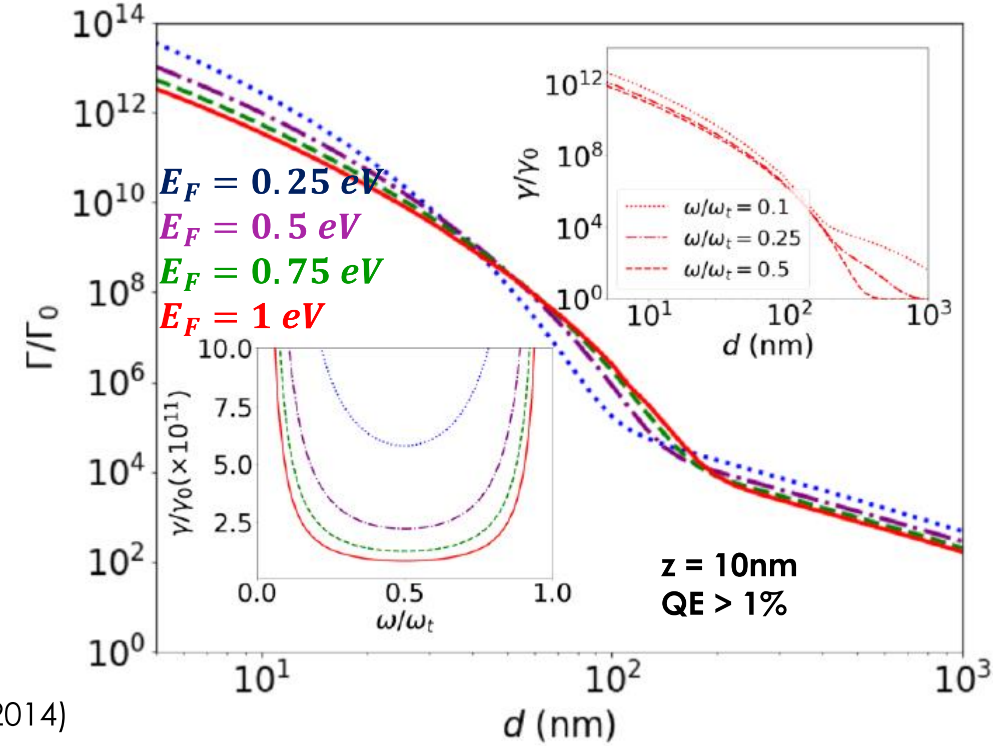


TQSE in one-dimensional carbon nanostructures

- **Carbon nanotubes** – extreme enhancement of two-plasmon SE.
  - **Plasmons** propagate in **one-dimension.**





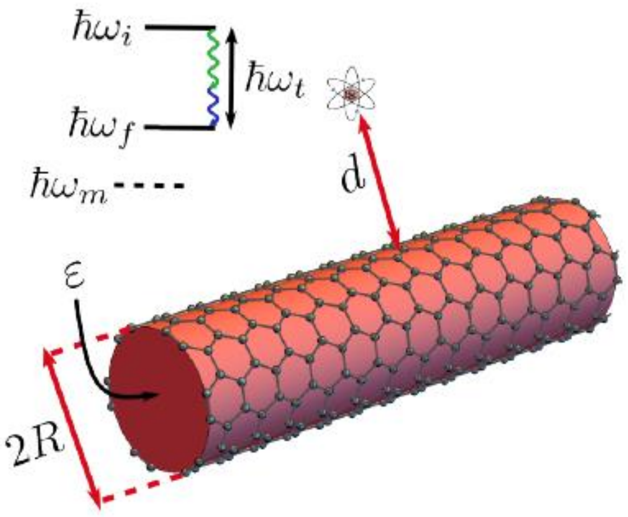


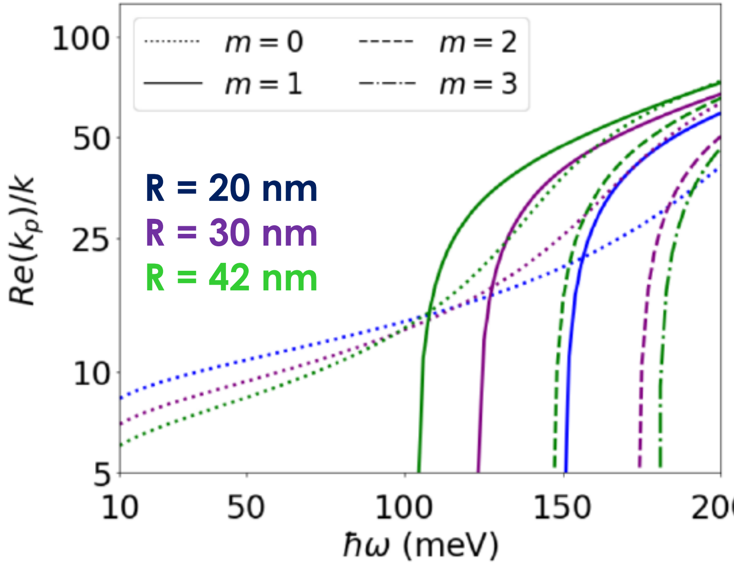
Y. M. *et. al.*, PRB **105**, 165412 (2022)

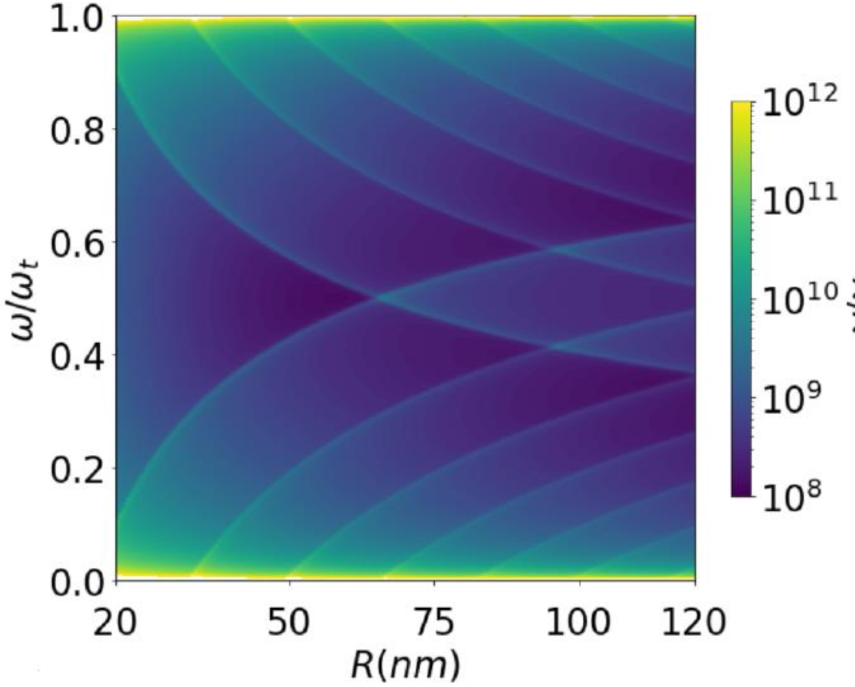
Wei Li *et. al.*, Nano Lett. **14** (2), 955 – 959 (2014)

TQSE in one-dimensional carbon nanostructures

- **Graphene coated wires**: tunable spectrum of emission.
  - **Different modes** contribute depending on the **nanowire radius**.



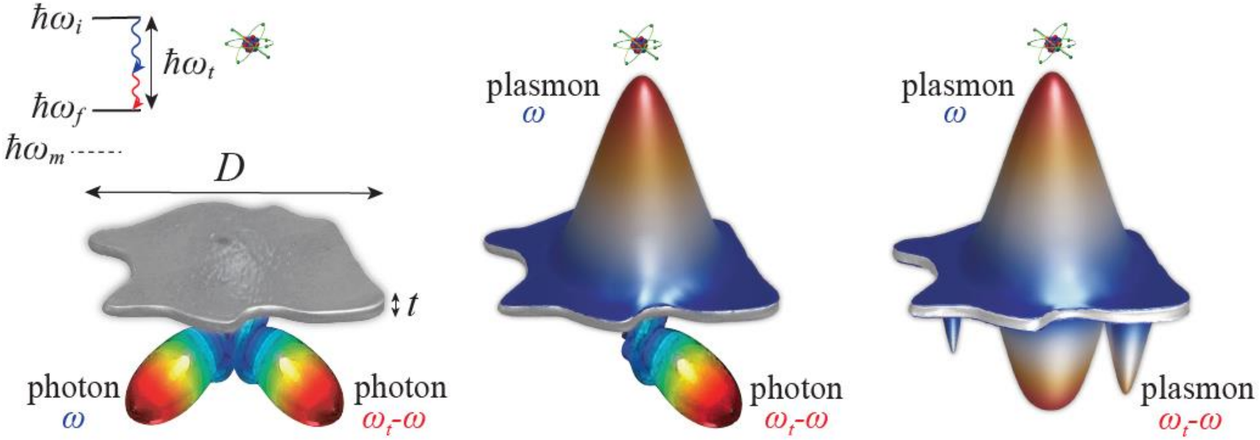




Y. M. *et. al.*, PRB **105**, 165412 (2022)

TQSE in atomically thin plasmonic nanostructures

- **Finite size** → far-field radiation.
- Three main decay channels.
- Entangled photons generation.



Y. M. et. al., PRL **125**, 033601 (2020)

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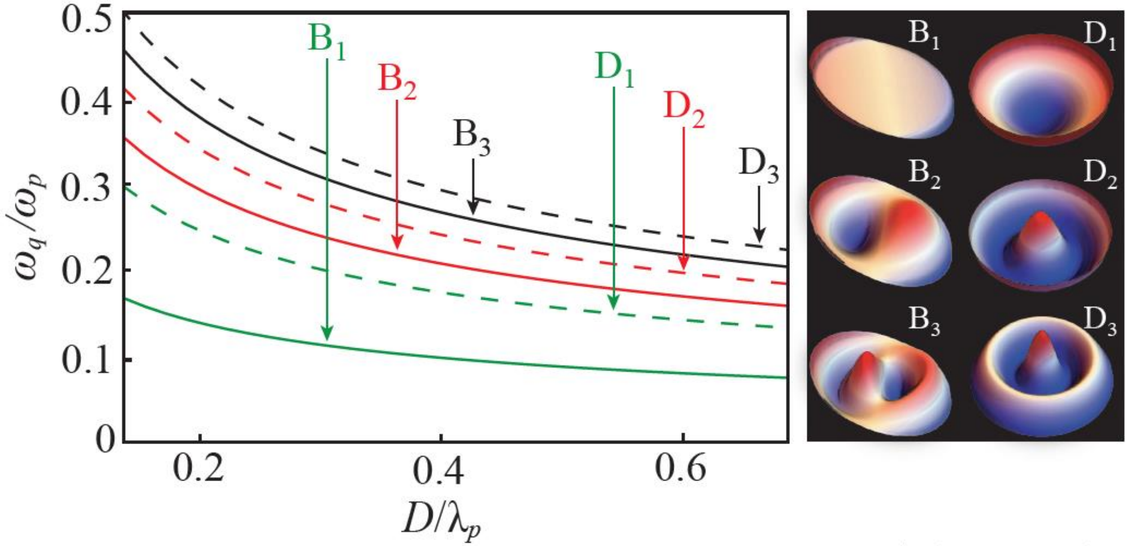
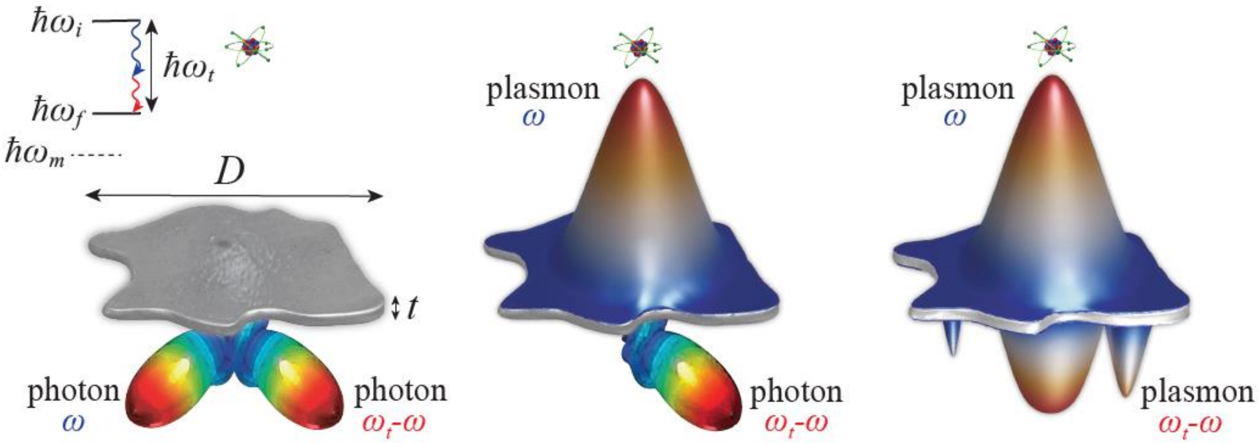


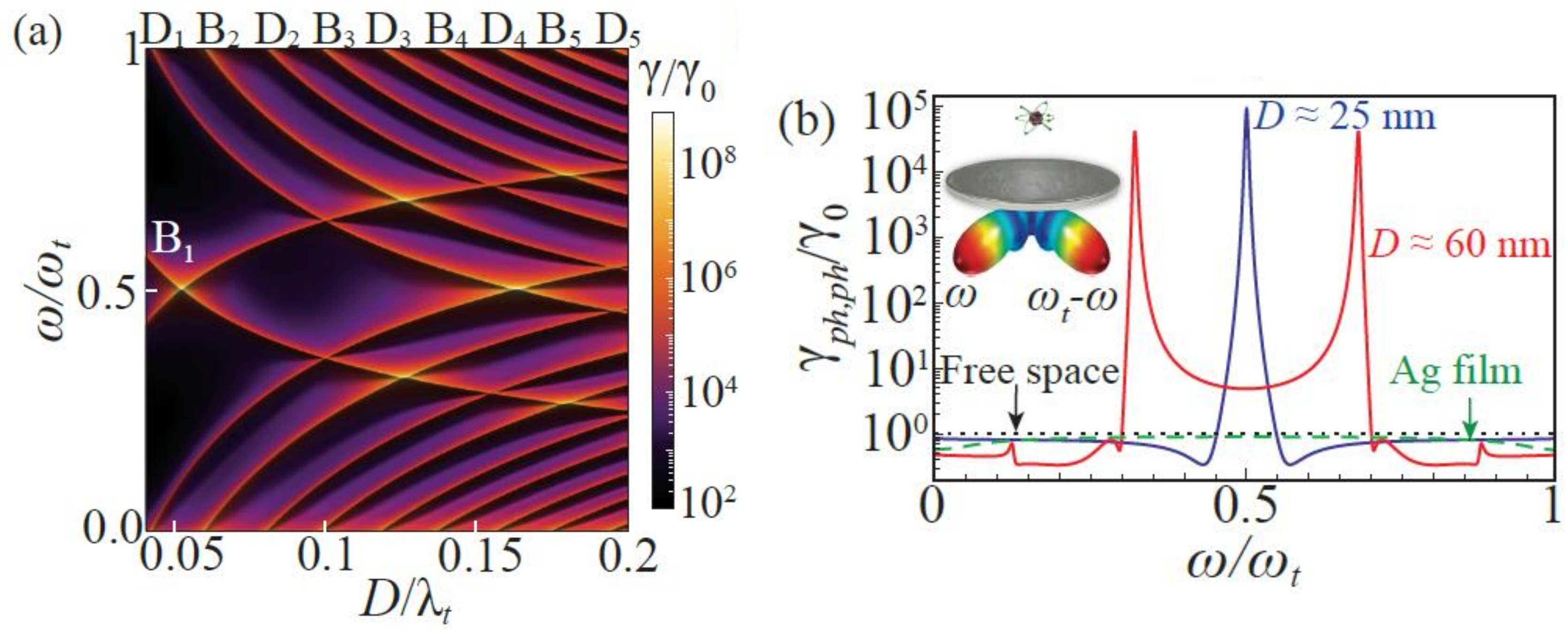


- Nanodisk → bright and dark plasmonic modes.
- Only bright modes radiate into the far-field.
- Well-defined tunable plasmonic frequencies.

Y. M. *et. al.*, PRL **125**, 033601 (2020)

TQSE in atomically thin plasmonic nanostructures

- Extreme enhancements at the plasmonic resonance frequencies of the nanodisk.
  - **Crossings** between bright and dark modes.
- **Photon-photon** decay channel **amplified** by the nanodisk bright modes.



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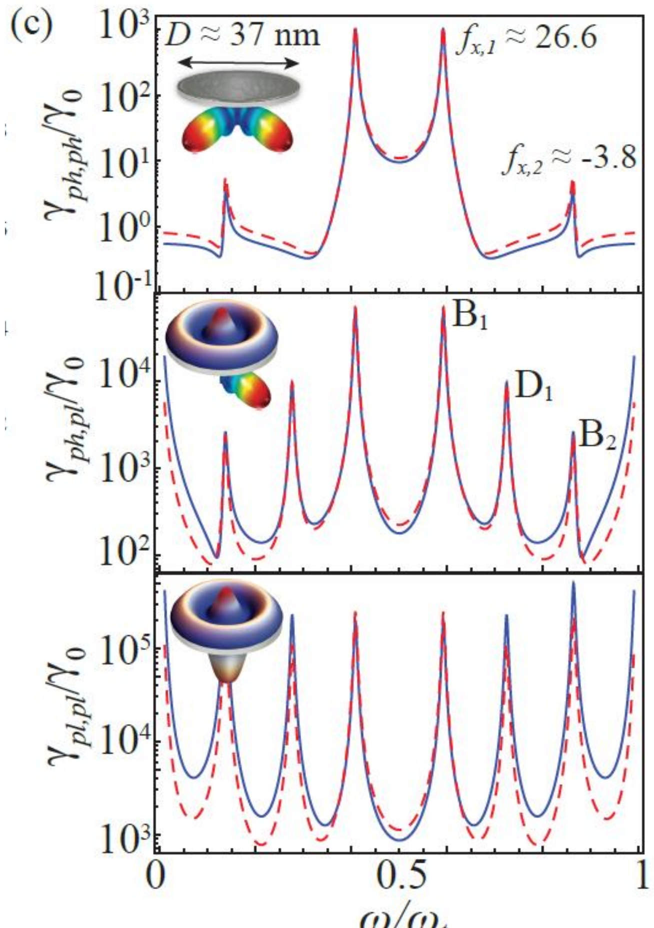

- All pathways contribute to the spectrum.
  - **Entangled ph-ph and ph-pl states.**

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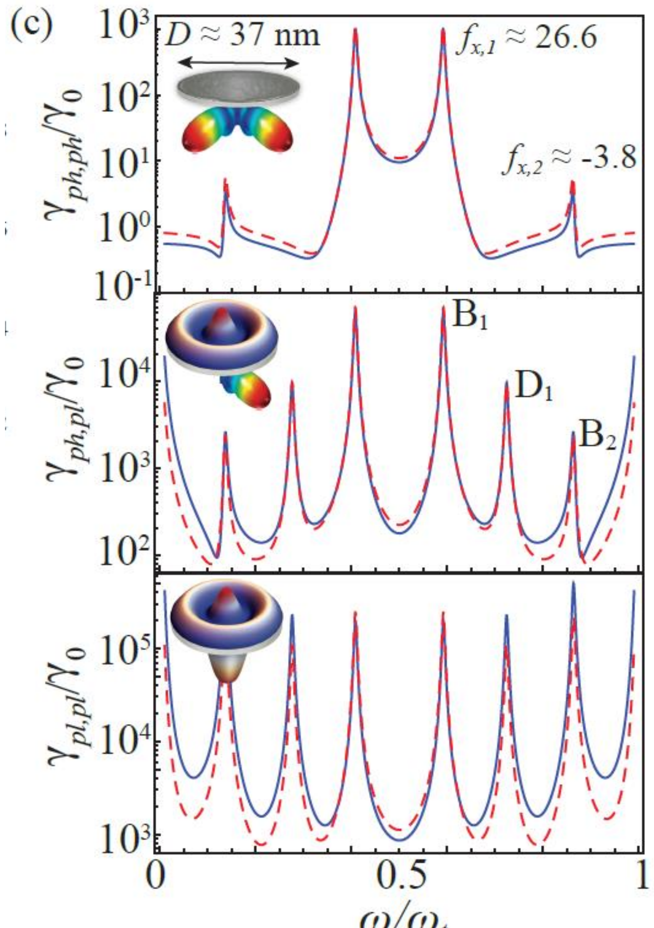
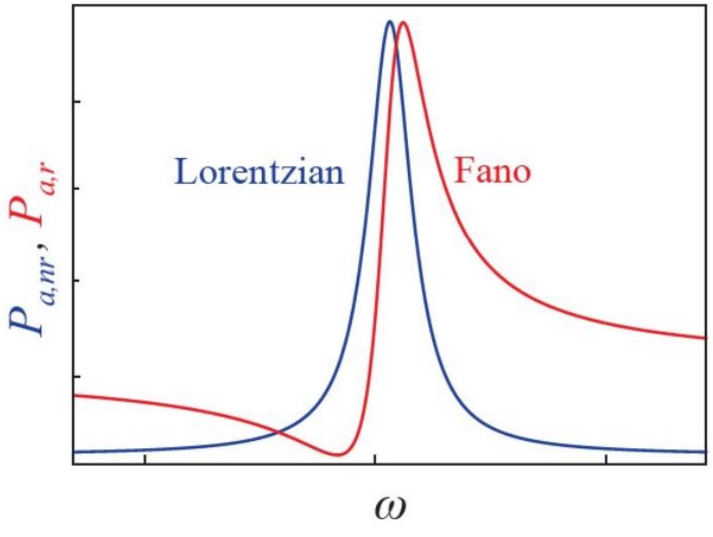


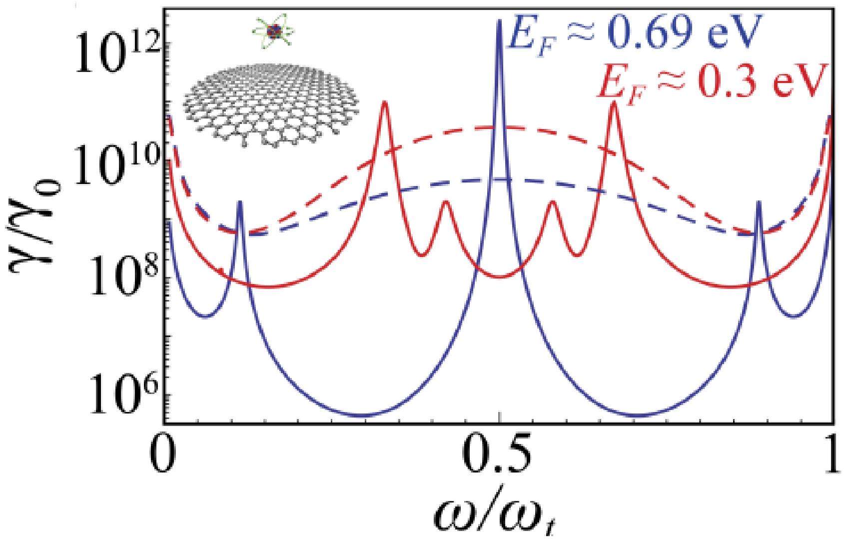
- All pathways contribute to the spectrum.
  - **Entangled ph-ph and ph-pl states.**
- Lorentzian resonances in the pl-pl channel.
- **Fano resonances** in the ph-ph channel.
  - Interference between direct emission to the far-field and radiation by the nanostructure.

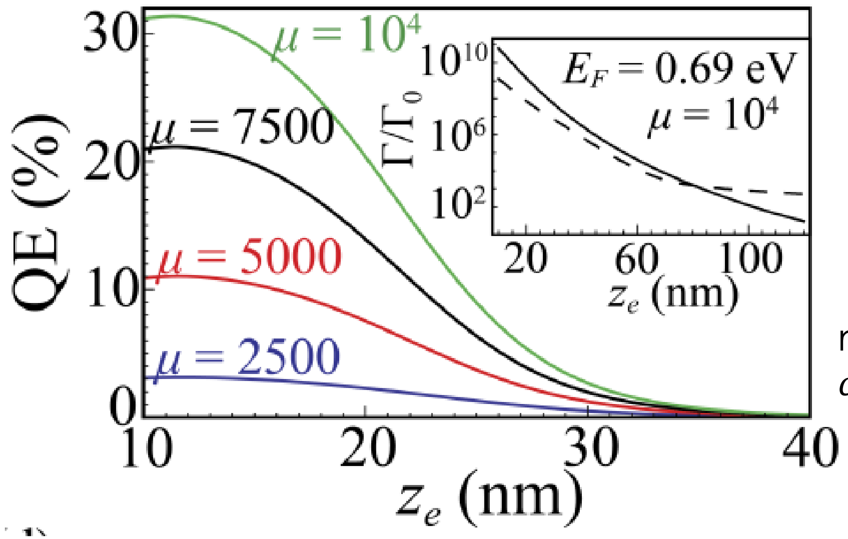


Y. M. *et. al.*, PRL **125**, 033601 (2020)

TQSE in a graphene nanodisk

- Localized spectrum enhancements higher than in a graphene sheet.
- **TQSE** can be almost **as likely** to occur as **one-quantum SE**.
  - Robustness to distance variations.





μ: electron mobility in $cm^2V^{-1}s^{-1}$

Y. M. *et. al.*, *PRL* ***125****, 033601 (2020)*

Casimir Effect

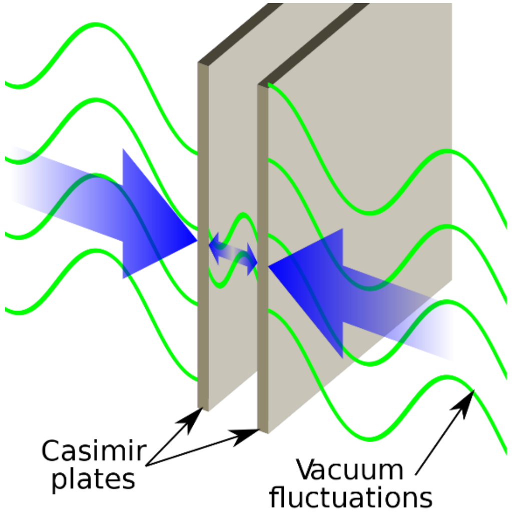
The Casimir method

- Casimir energy per unit area between two **perfect mirrors**:

$$E = \frac{1}{A}\left[\left(\sum_{\mathbf{k}\lambda}\frac{\hbar\omega_k}{2}\right)_I - \left(\sum_{\mathbf{k}\lambda}\frac{\hbar\omega_k}{2}\right)_{II}\right]$$

- A regularization is needed in order to subtract the infinities.

$$E(a) = \lim_{\epsilon\to 0^+} E_r(a,\epsilon) = -\frac{\pi^2}{720}\frac{\hbar c}{a^3}.$$



Lifshitz formula

- Casimir energy between two **real materials** in a plane geometry:

Energy per unit area

$$E = k_B T \sum_n{}' \int \frac{d^2\mathbf{k}_\parallel}{(2\pi)^2} \log\det\left(1 - \mathbb{R}_1 \cdot \mathbb{R}_2 e^{-2k_{z,n}d}\right)$$

Sum over Matsubara frequencies → $\xi_n = 2\pi k_B T n/\hbar$

Lifshitz formula

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$$k_{z,n} = \sqrt{\mathbf{k}_{\parallel}^2 + \xi_n^2/c^2}$$

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Reflection matrices

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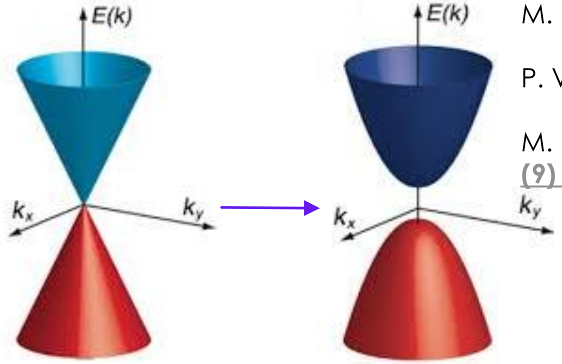
Zero-temperature limit

$$k_B T \sum_n \to \frac{\hbar}{2\pi} \int_0^\infty d\xi$$

Casimir forces in the flatland

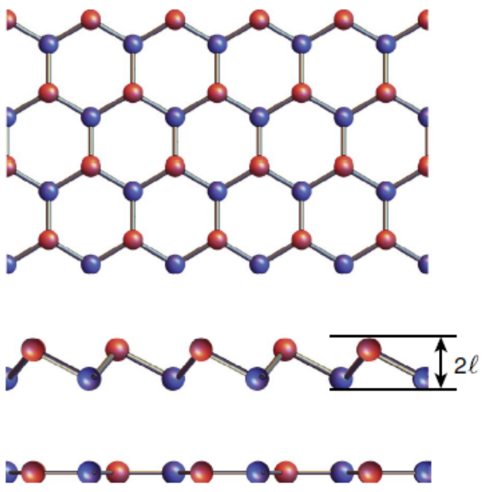
- Graphene family materials.
  - Silicene, germanene, stanene, plumbene.
  - Honeycomb structure, but with a finite buckling.
  - **Topological insulators**.

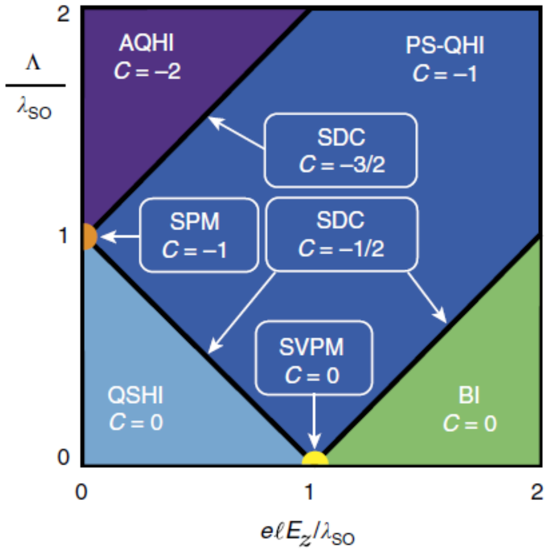


M. Ezawa, J. Phys. Soc. Jpn. **84**, 121003 (2015)

P. Vogt *et. al.*, PRL **108**, 155501 (2012)

M. E. Dávila *et. al.*, New Journal of Physics **16 (9)** 095002 (2014)





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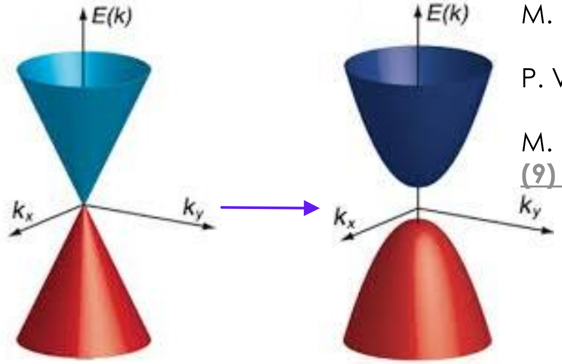

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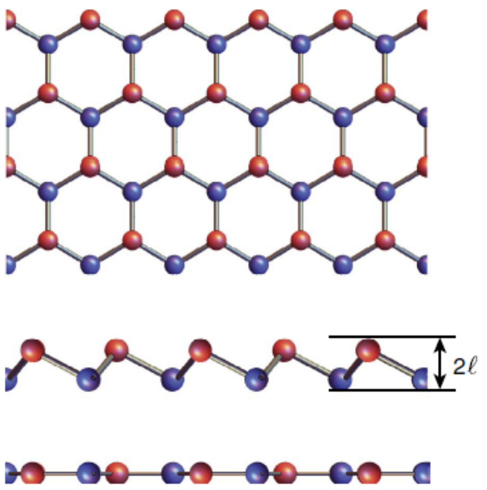
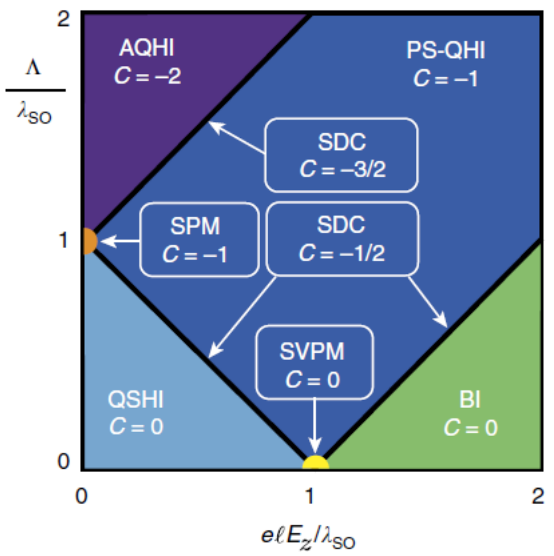
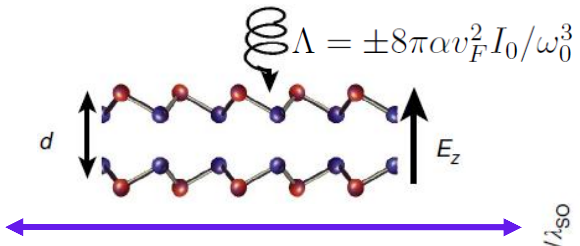
M. E. Dávila *et. al.*, *New Journal of Physics* **16 (9)** 095002 (2014)

Casimir energy phase diagram

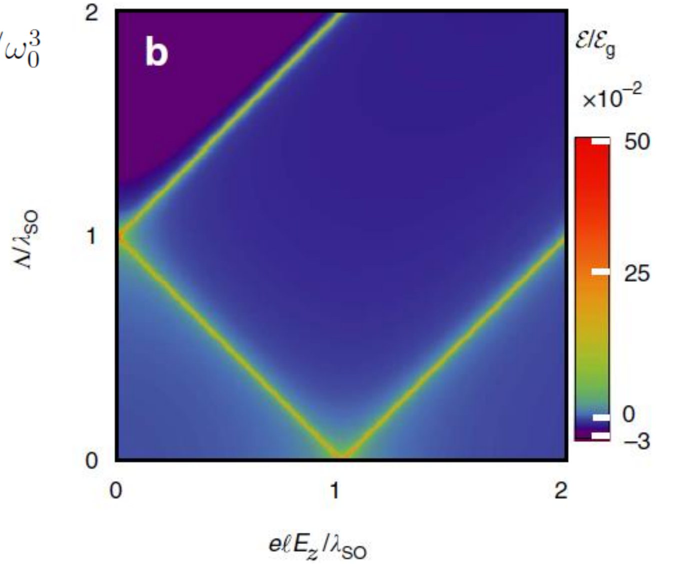






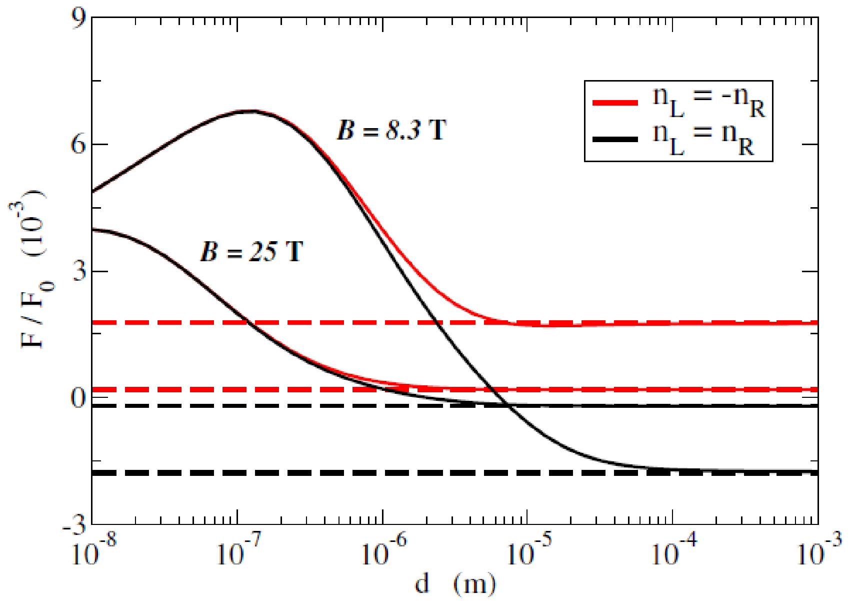
$\frac{4\alpha}{\pi} C_1 C_2$

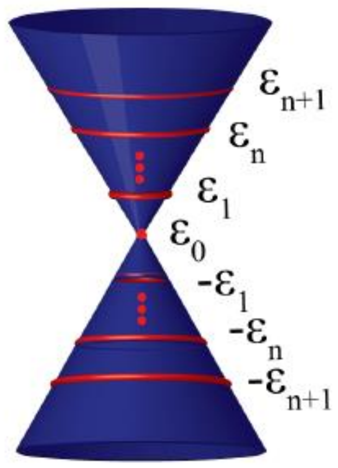
P. Rodriguez-Lopez *et. al.*, *Nat. Comm.* **8**, 14699 (2017)



Casimir forces in the flatland

- Meanwhile, for the graphene-graphene Casimir force...
  - We have **quantum Hall effect**





W.-K. Tse, A. H. MacDonald,
*PRL **109**, 236806 (2012)*

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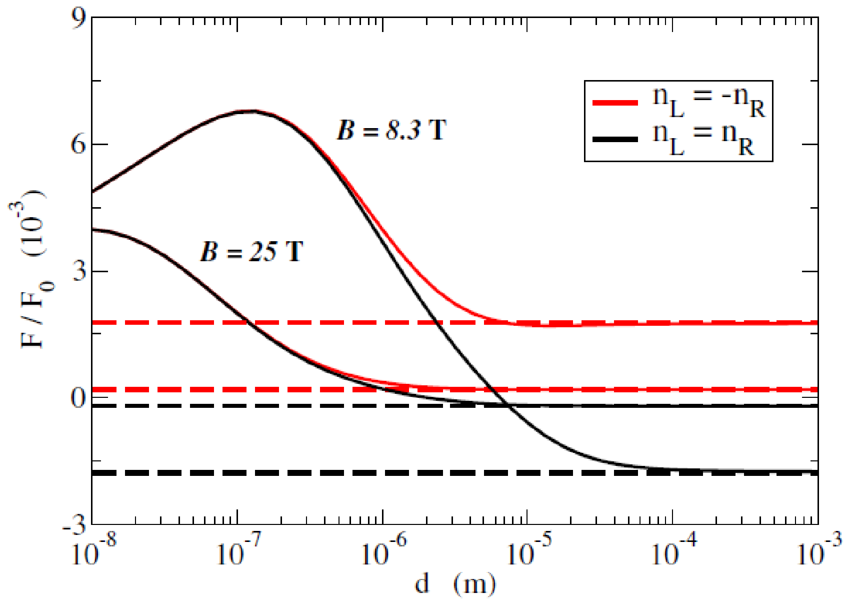

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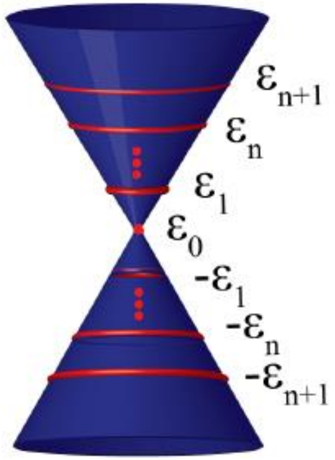


What if?

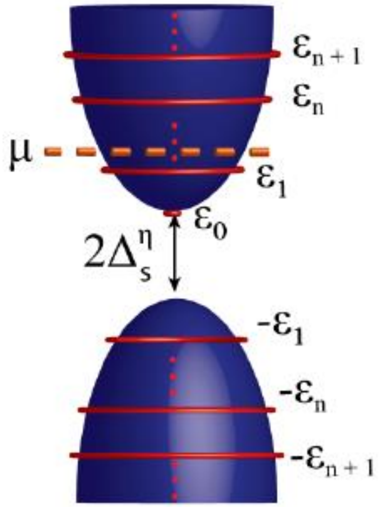
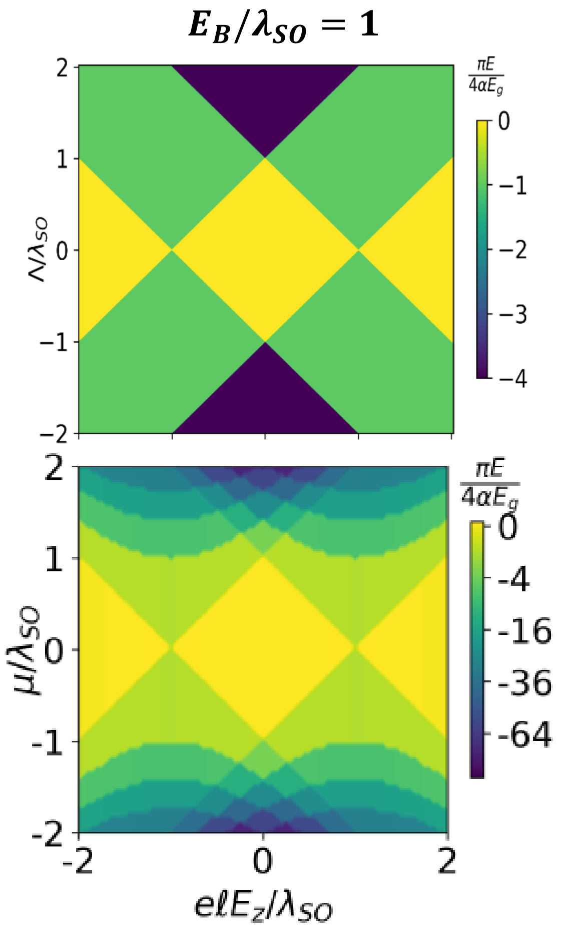


Photo-induced phase transitions and quantum Hall physics in the Casimir force



- Two contributions to the Chern number results in

$$\frac{E^{(0)}}{E_g} = -\frac{4\alpha}{\pi}(C_{1,\mathrm{ph}} + C_{1,\mathrm{QH}})(C_{2,\mathrm{ph}} + C_{2,\mathrm{QH}}).$$

- **Chemical potential** as a **substitute** of the circularly polarized **laser**.
- New boundaries in the phase diagram.
- No more attraction near the boundaries (gap always present due to the Landau levels).

Y. M. *et. al.*, *Phys. Rev. Research* ***3**, 023061* (2021)

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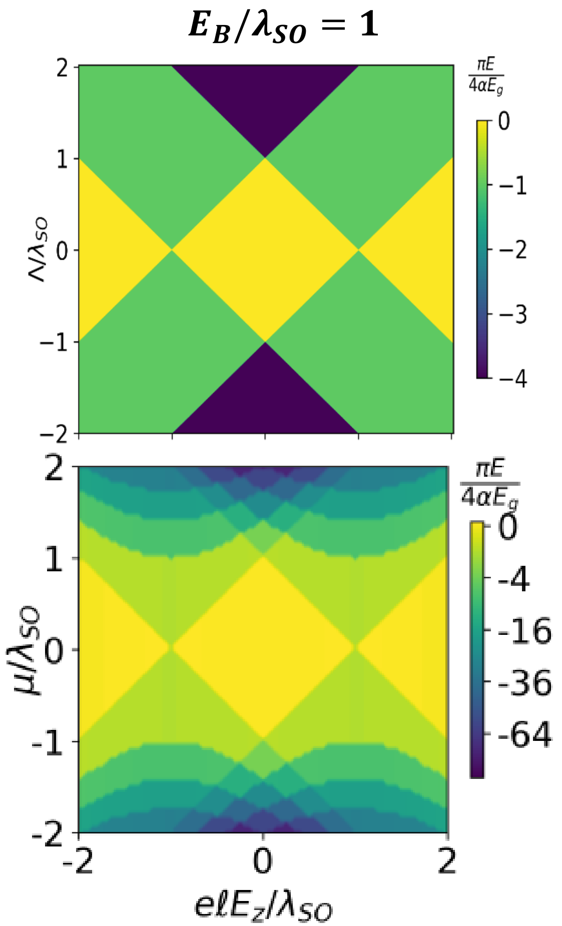

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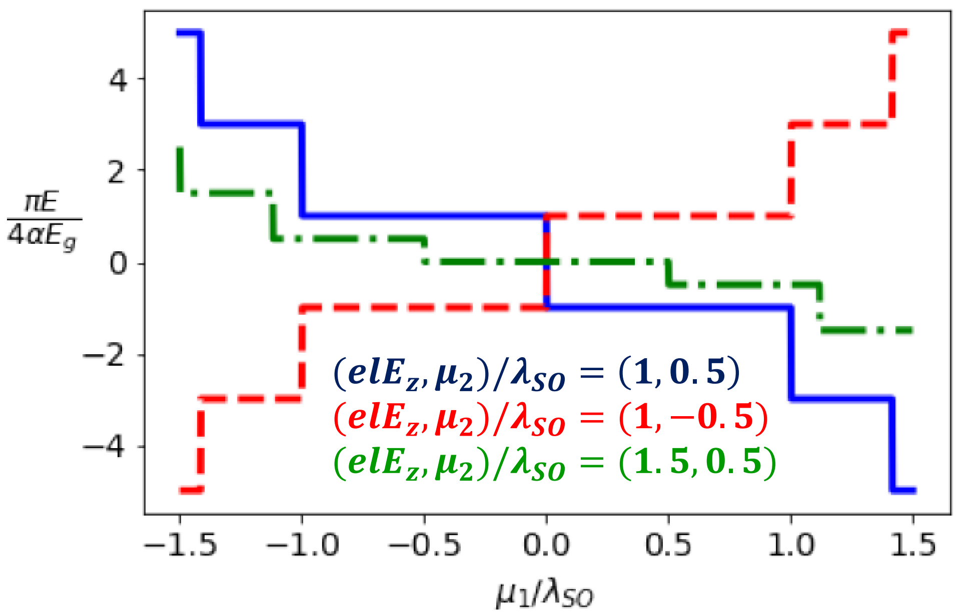
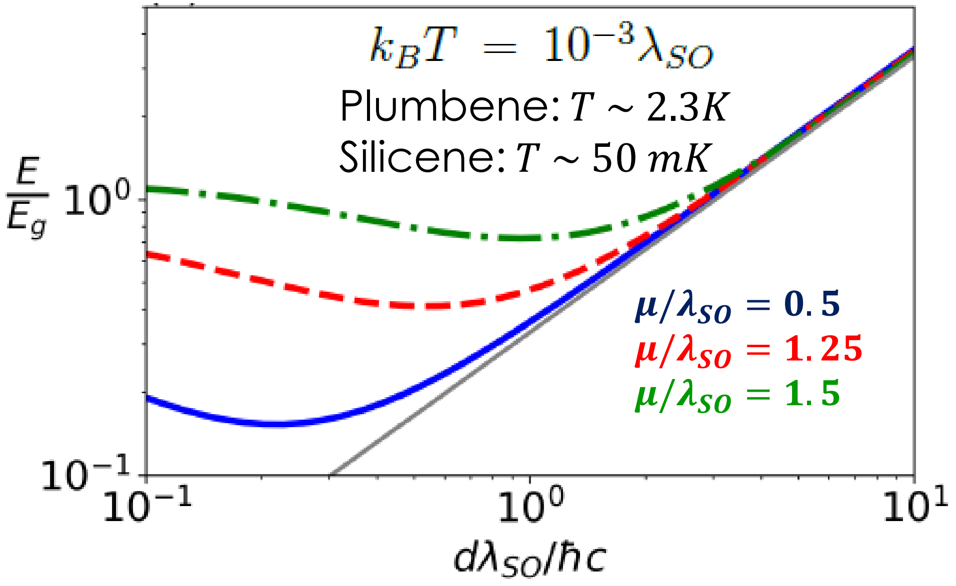
Control over the sign of the force



Y. M. et. al., Phys. Rev. Research **3**, 023061 (2021)

Thermal effects

With dissipation



Temperature erases repulsion

Y. M. et. al., Phys. Rev. Research **3**, 023061 (2021)

Thermal effects

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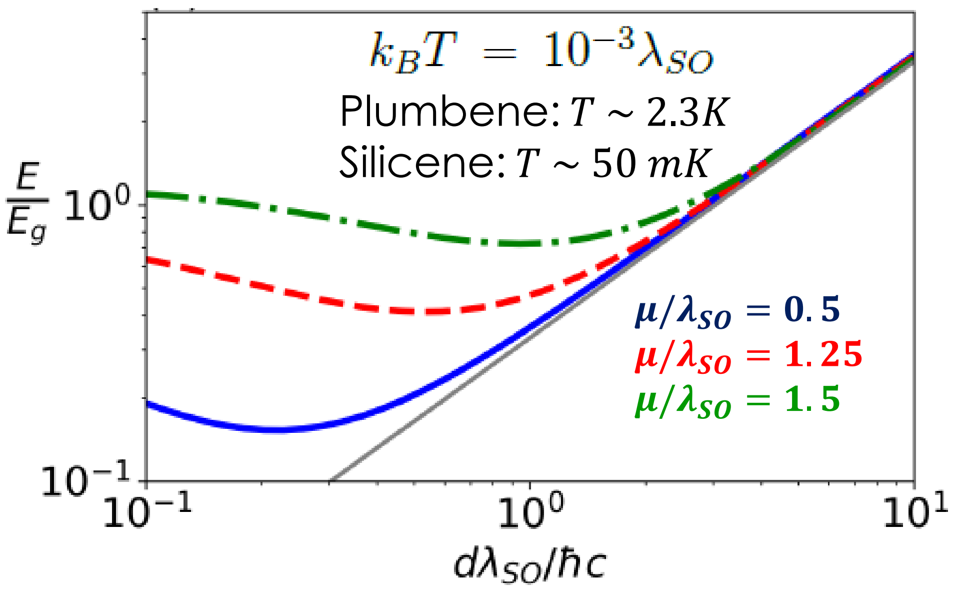
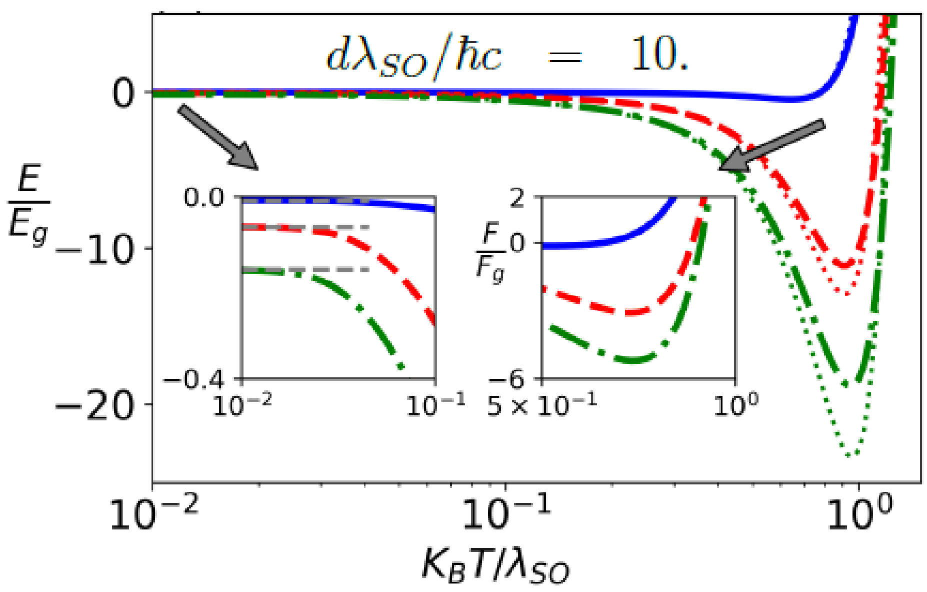


Without dissipation



Temperature erases repulsion

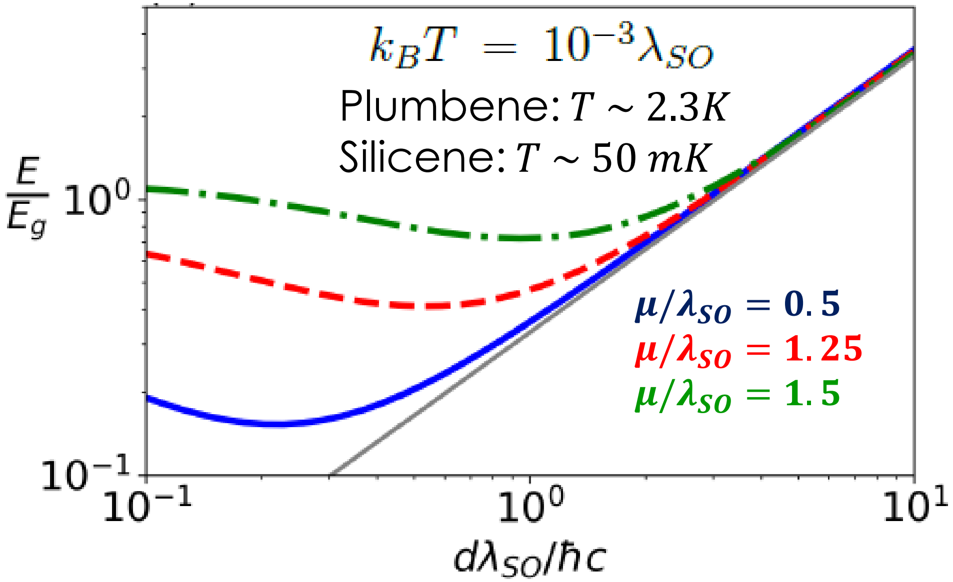
Temperature enhances repulsion

Room temperature repulsion in stanene

Y. M. et. al., Phys. Rev. Research **3**, 023061 (2021)

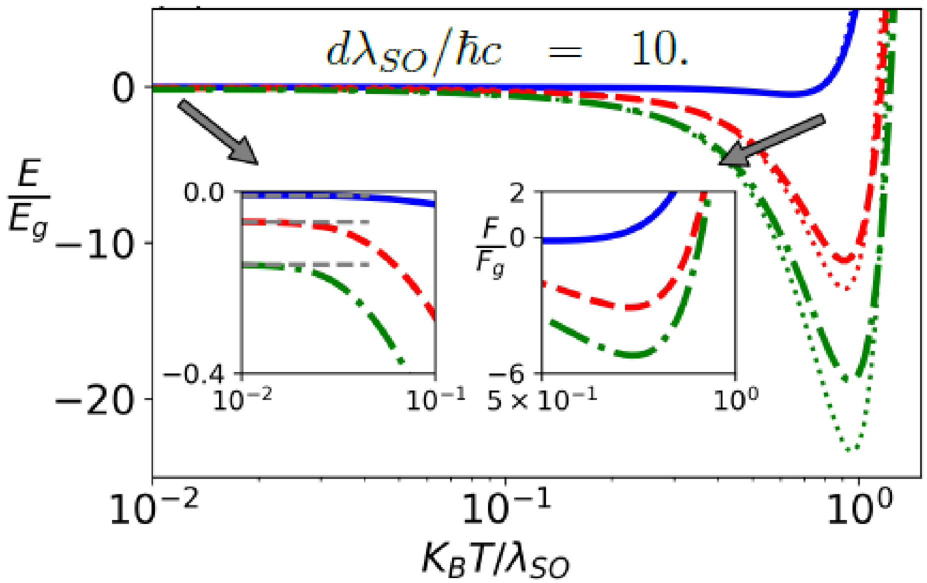
Thermal effects

Drude?



Temperature erases repulsion

Plasma?



Temperature enhances repulsion

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Y. M. et. al., Phys. Rev. Research **3**, 023061 (2021)

Final remarks

Conclusions

- We made a **comprehensive study of TQSE** that enabled us to propose novel material platforms to **enhance** and **tailor** the generation of **two-plasmon** and **two-photon entangled states**.
- We added important contributions to the **Casimir effect** between **graphene family** materials by bringing a new player to the field, namely, an externally applied **magnetic field.**

- Due to time constraints in this presentation we did not show some other interesting results.
  - In TQSE near atomically thin nanostructures, it was shown that the **generation of single photon states through the ph-pl channel** can be **more efficient than via first-order one-photon SE.**

List of publications during PhD

- Y. Muniz, D. Szilard, W. J. M. Kort-Kamp, F. S. S. Rosa, and C. Farina, *Quantum two-photon emission in a photonic cavity*, Phys. Rev. A **100**, 023818 (2019).
- Y. Muniz, A. Manjavacas, C. Farina, D. A. R. Dalvit, and W. J. M. Kort-Kamp, *Two-photon spontaneous emission in atomically thin plasmonic nanostructures*, Phys. Rev. Lett. **125**, 033601 (2020).
- Y. Muniz, C. Farina, and W. J. M. Kort-Kamp, *Casimir forces in the flatland: Interplay between photoinduced phase transitions and quantum hall physics*, Phys. Rev. Research, **3**, 023061 (2021).
- Y. Muniz, P. Abrantes, L. Martín-Moreno, F. Pinheiro, C. Farina, and W. J. M. Kort-Kamp, *Entangled two-plasmon generation in carbon nanotubes and graphene coated wires*, Phys. Rev. B, **105**, 165412 (2022).
- L. Weitzel, Y. Muniz, C. Farina, Carlos A. D. Zarro, *Two-photon spontaneous emission of an atom in a cosmic string background*, Phys. Rev. D. **106**, 045020 (2022).

Perspectives

- **TQSE** near **magneto-optically controlled** atomically thin plasmonic **nanostructures**.
- More on **high-order quantum transitions** (e.g., quadrupolar SE).
- **SE** near **graphene family** materials.
- Further investigations of **thermal effects** in the **graphene family Casimir force**.

Thanks!