Modeling Spontaneous Emission Rate near a Metallic Split-Ring Resonator in a Graphene-Incorporated Multilayer Substrate

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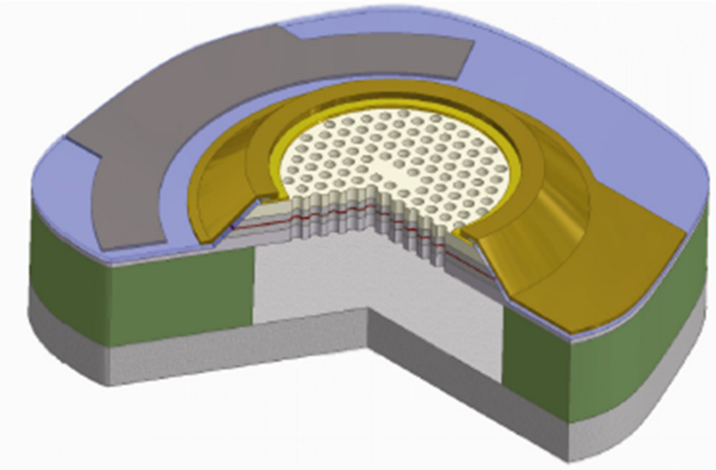
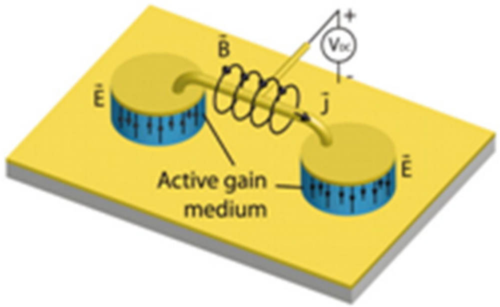
OUTLINE

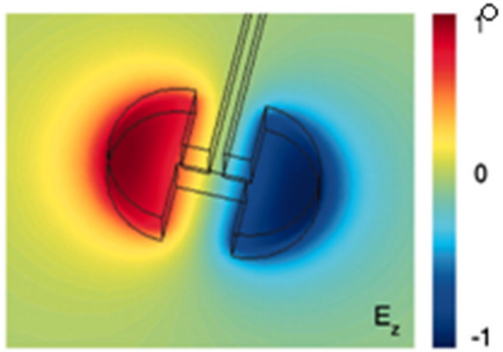
- 1. Significance and History
- 2. Quantum Electrodynamics
- 3. Spontaneous Emission, Local Density of States, and Green's Tensor
- 4. Graphene Plasmonics to Control the Spontaneous Emission Rate
- 5. Conclusion

WHY SPONTANEOUS EMISSION (DECAY) IS IMPORTANT?

Control of spontaneously emitted light lies at the heart of quantum optics. It is essential for diverse applications ranging from lasers, light-emitting diodes, solar cells, and quantum information.







Purcell factor

LED (photonic crystal cavity) ⟷ **Laser (metallic microcavity)**

M. Francardi et al. Appl. Phys. Lett. 93, 143102 (2008)

C Walther et al. Science 327, 1495-1497 (2010)

Spontaneous emission: classical view

Transition rates $R_{\text{spon}} = AN_2$ $R_{\text{stim}} = BN_2\rho_{\text{em}}$ $R_{\text{abs}} = B'N_1\rho_{\text{em}}$

Boltzmann statistics $N_2/N_1 = \exp(-E_g/k_BT) \equiv \exp(-h\nu/k_BT)$

Thermal equilibrium $AN_2 + BN_2\rho_{\text{em}} = B'N_1\rho_{\text{em}}$

Spectral density of EM energy

$$\rho_{\text{em}} = \frac{A/B}{(B'/B)\exp(h\nu/k_BT) - 1}$$

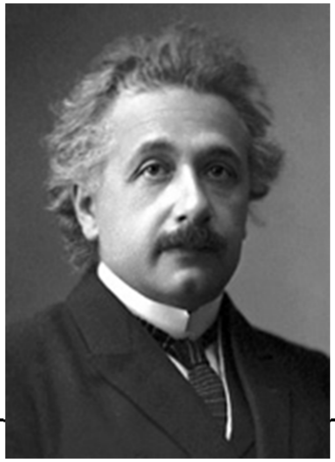
Blackbody radiation ***Planck's formula***

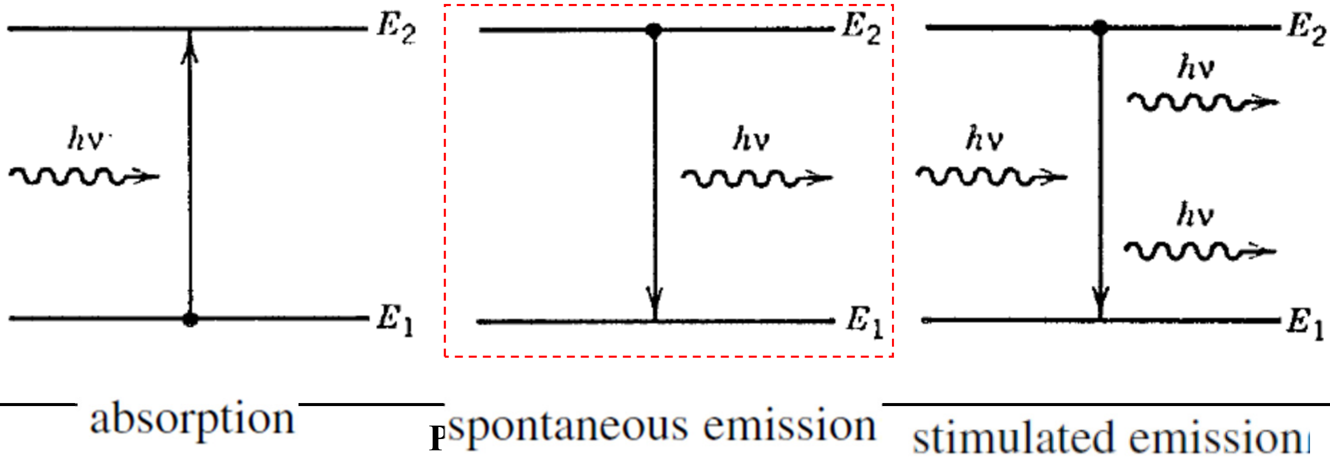
$$\rho_{\text{em}} = \frac{8\pi h\nu^3/c^3}{\exp(h\nu/k_BT) - 1}$$

Einstein's coefficients $A = (8\pi h\nu^3/c^3)B;$ $B' = B$

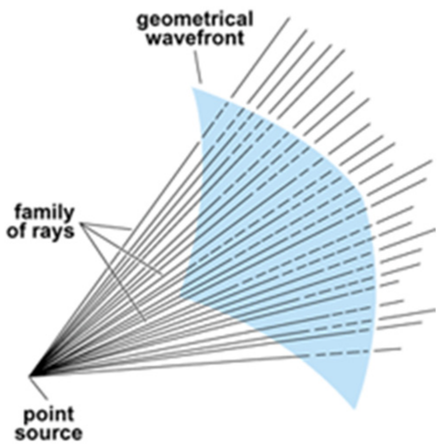
1917

$$E_2 - E_1 = \Delta E = h\nu$$

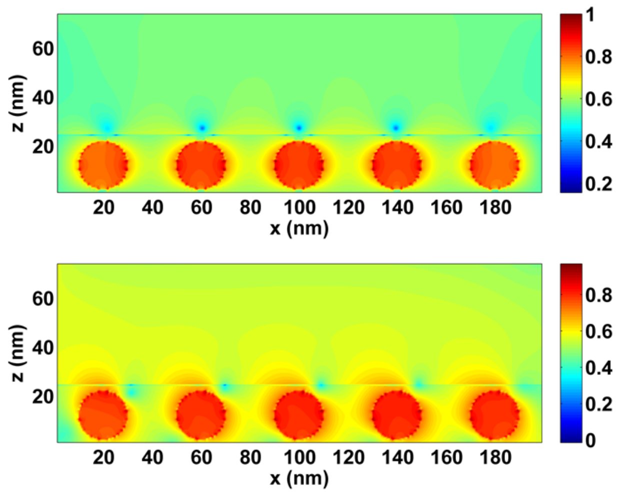




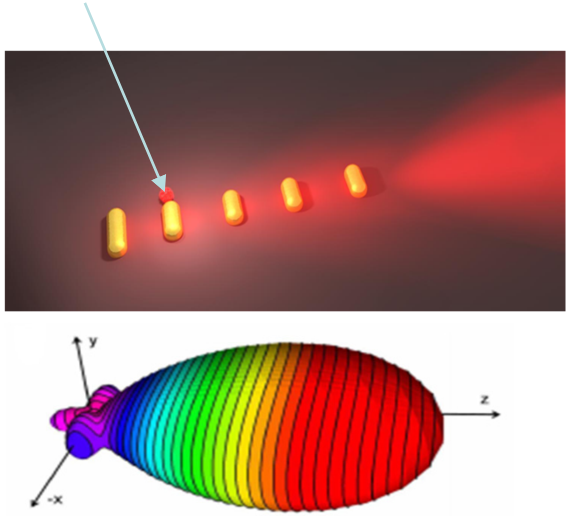
THREE REGIMES IN OPTICS



classical optics
ray physics
D>>λ



nano-optics
wave physics
D~λ



quantum optics
quantum physics
D<<λ

At quantum regimes, the object size (<10 nm) is quite small compared to wavelength. In this situation, semi-classical Maxwell-Schrödinger system is required to describe the EM—particle interaction. Moreover, if the number of photons is also quite small, Maxwell's equations should be quantized.

A MODERN INTERPRETATION: QUANTUM ELECTRODYNAMICS (1)

Quantized form of Maxwell Equations

$$\boxed{\begin{aligned}\nabla\times\hat{\mathbf{E}} &= -\frac{\partial\hat{\mathbf{B}}}{\partial t}\\ \nabla\times\hat{\mathbf{H}} &= \hat{\mathbf{J}}+\frac{\partial\hat{\mathbf{D}}}{\partial t}\\ \nabla\cdot\hat{\mathbf{B}} &= 0\\ \nabla\cdot\hat{\mathbf{D}} &= \rho\end{aligned}}$$

$$\hat{\mathbf{B}} = \nabla\times\hat{\mathbf{A}} \qquad \hat{\mathbf{E}} = -\frac{\partial\hat{\mathbf{A}}}{\partial t}-\nabla\phi$$

$$\nabla^2\phi = -\frac{\rho}{\varepsilon_0} \qquad \nabla\bullet\hat{\mathbf{A}} = 0$$

Coulomb gauge

Quantized Hamiltonian

$$\hat{H}\left(\hat{\mathbf{A}},\hat{\mathbf{Y}},\hat{\mathbf{p}}\right) = \hat{H}^{em}\left(\hat{\mathbf{A}},\hat{\mathbf{Y}}\right)+H^{q}\left(\hat{\mathbf{p}},\hat{\mathbf{A}}\right)$$

$$\hat{H}^{em}\left(\hat{\mathbf{A}},\hat{\mathbf{Y}}\right) = \int_\Omega\left(\frac{1}{2\epsilon_0}\left|\hat{\mathbf{Y}}\right|^2+\frac{1}{2\mu_0}\left|\nabla\times\hat{\mathbf{A}}\right|^2\right)d\mathbf{r}$$

eigenmodes

$$\hat{H}^{q}\left(\hat{\mathbf{p}},\hat{\mathbf{A}}\right) = \frac{\left(\hat{\mathbf{p}}-q\hat{\mathbf{A}}\right)^2}{2m}+V$$

$$\hat{\mathbf{A}} = \sum_k\frac{\alpha_k}{\omega_k}\left(\mathbf{U}_k(\mathbf{r})\hat{a}_k+\mathbf{U}_k^*(\mathbf{r})\hat{a}_k^+\right),\ \alpha_k = \sqrt{\frac{\hbar\omega_k}{2\epsilon_0}}$$

wave-particle duality

$$H_0^{q}\left(\hat{\mathbf{p}},\hat{\mathbf{A}}\right) = \frac{\hat{\mathbf{p}}^2}{2m}+V$$

$$\hat{H}\left(\hat{\mathbf{A}},\hat{\mathbf{Y}},\hat{\mathbf{p}}\right) = \underbrace{\hat{H}^{em}\left(\hat{\mathbf{A}},\hat{\mathbf{Y}}\right)+H_0^{q}\left(\hat{\mathbf{p}},\hat{\mathbf{A}}\right)}_{\text{non-interaction part}}+\underbrace{\boxed{H_I^{q}\left(\hat{\mathbf{p}},\hat{\mathbf{A}}\right)}}_{\text{interaction part}}$$

$$\boxed{\hat{H}\Psi(t) = i\hbar\frac{\partial\Psi(t)}{\partial t}}$$

$$H_I^{q}\left(\hat{\mathbf{p}},\hat{\mathbf{A}}\right) \approx \frac{q\hat{\mathbf{p}}\cdot\hat{\mathbf{A}}}{m}$$

Can be solved by perturbation method!

$$\Psi(t) = a(t)|e,0> + \sum_k b_k(t)|g,1_k>$$ wave function

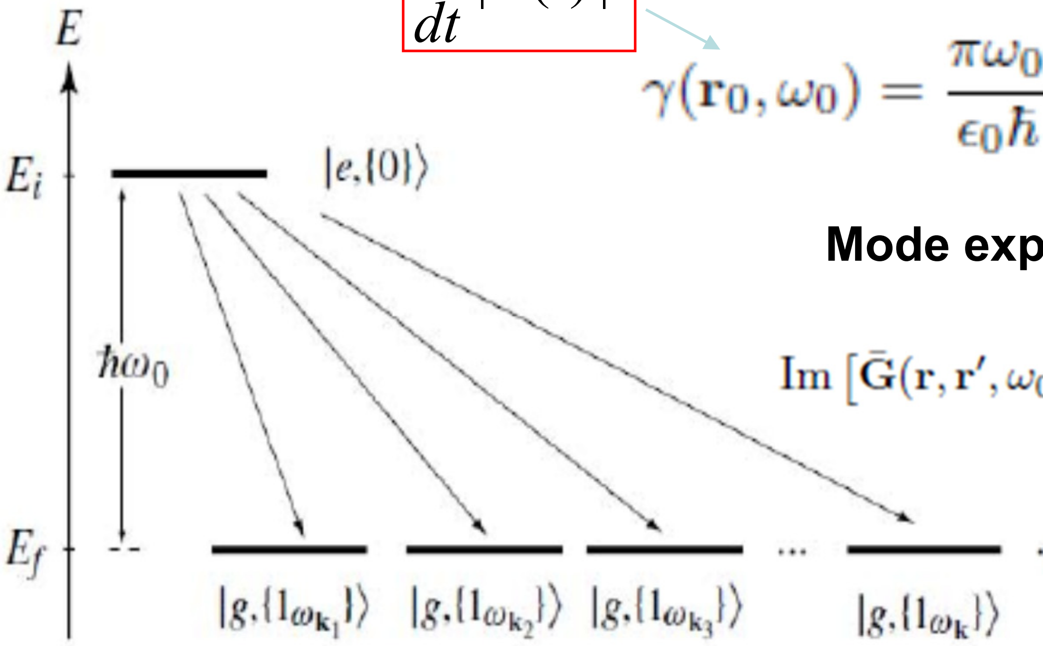
e: excited state of atom; g: ground state of atom; 0: no photon; 1: one photon

A MODERN INTERPRETATION: QUANTUM ELECTRODYNAMICS (2)

$$\boxed{\frac{d}{dt}|a(t)|^2}$$

Spontaneous emission rate by Fermi golden rule

$$\gamma(\mathbf{r}_0,\omega_0) = \frac{\pi\omega_0}{\epsilon_0\hbar}\sum_k \left[\mathbf{p}\cdot(\mathbf{u_k}\mathbf{u_k^*})\cdot\mathbf{p}\right]\delta(\omega_k-\omega_0)$$



Mode expansion of dyadic green's function

$$\mathrm{Im}\left[\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}',\omega_0)\right] = \frac{\pi c^2}{2\omega_0}\sum_k \mathbf{u_k}(\mathbf{r},\omega_k)\mathbf{u_k^*}(\mathbf{r}',\omega_k)\delta(\omega_k-\omega_0)$$

Representation by Green's tensor

$$\gamma(\mathbf{r}_0,\omega_0) = \frac{2\omega_0^2}{\epsilon_0\hbar c^2}\left\langle\mathbf{p}\cdot\mathrm{Im}\left[\bar{\mathbf{G}}(\mathbf{r}_0,\mathbf{r}_0,\omega_0)\right]\cdot\mathbf{p}\right\rangle$$

Electromagnetic Local density of state (EMLDOS)

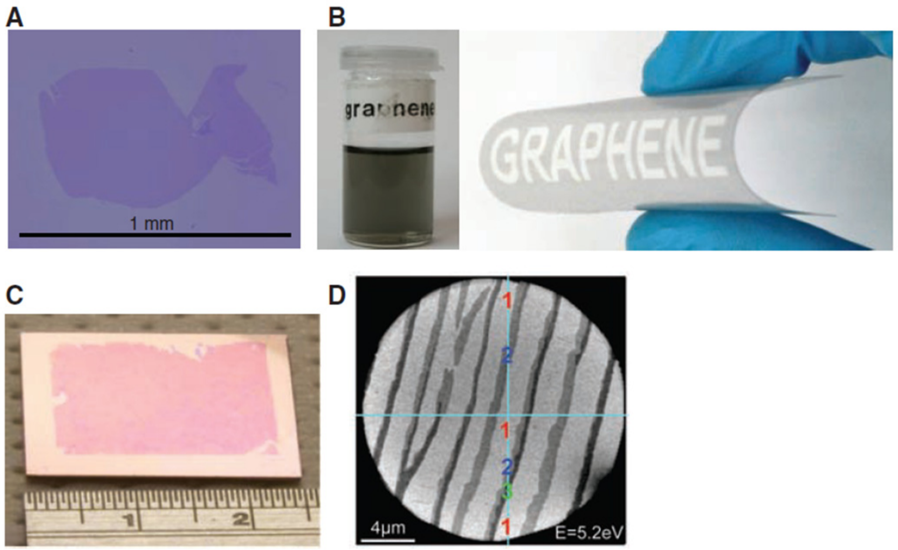
$$\rho(\mathbf{r}_0,\omega_0) = \sum_{\mathbf{k}}|\mathbf{u_k}|^2\delta(\omega_{\mathbf{k}}-\omega_0) = \frac{2\omega_0}{\pi c^2}\mathrm{Im}\left\{\mathrm{Tr}\left[\bar{\mathbf{G}}(\mathbf{r}_0,\mathbf{r}_0,\omega_0)\right]\right\}$$

Purcell factor

$$\frac{\gamma}{\gamma_0} = \frac{\rho(\mathbf{r}_0,\omega_0)}{\rho_0(\mathbf{r}_0,\omega_0)} = \frac{\mathrm{Im}\left\{\mathrm{Tr}\left[\bar{\mathbf{G}}(\mathbf{r}_0,\mathbf{r}_0,\omega_0)\right]\right\}}{\mathrm{Im}\left\{\mathrm{Tr}\left[\bar{\mathbf{G}}_0(\mathbf{r}_0,\mathbf{r}_0,\omega_0)\right]\right\}}$$

GRAPHENE

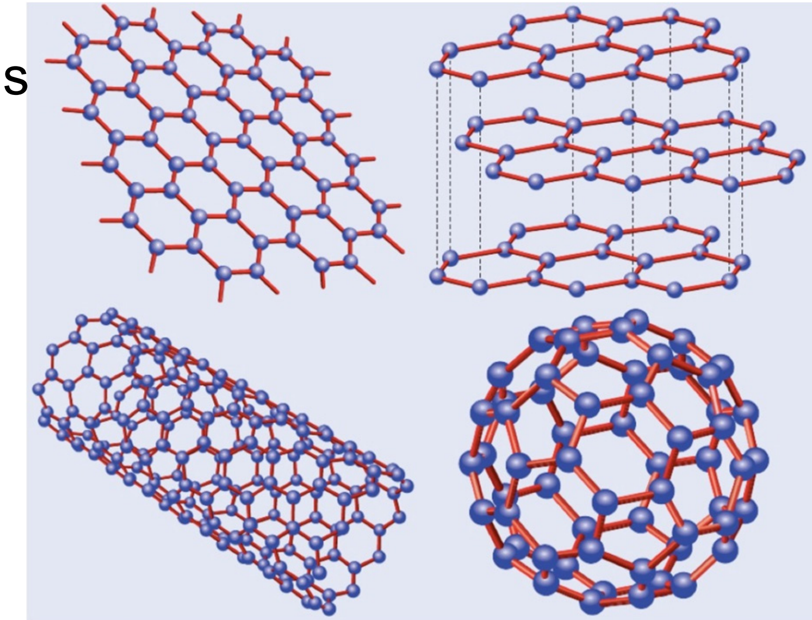
- Atomic thickness;
- High optical transmittance and conductivity;
- Dynamically modify chemical potentials through tuning the gate voltage



Fabrication

SCIENCE VOL 324 19 JUNE 2009

Graphene Graphite

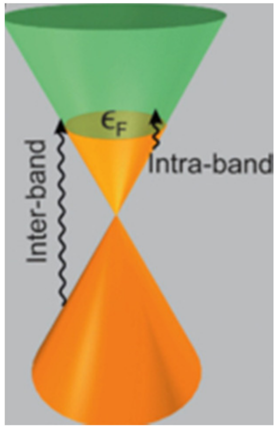


Carbon Nanotubes Fullerences (C_{60})

Physics World 19, 33 (2006)

FORMULATIONS FOR GRAPHENE PERMITTIVITY

➢ **Surface conductivity of Graphene given by Kubo formula**



Intraband Relaxation (plasmonic effect)

$$\sigma(\omega,\mu_c,\Gamma,T)=\frac{je^2(\omega-j2\Gamma)}{\pi\hbar^2}\left[\frac{1}{(\omega-j2\Gamma)^2}\times\int_0^\infty\epsilon\left(\frac{\partial f_d(\epsilon)}{\partial\epsilon}-\frac{\partial f_d(-\epsilon)}{\partial\epsilon}\right)d\epsilon-\int_0^\infty\left(\frac{f_d(-\epsilon)-f_d(\epsilon)}{(\omega-j2\Gamma)^2-4(\epsilon/\hbar)^2}\right)d\epsilon\right]$$

Interband Transition
(Be neglectable at THz frequencies)

- ✓ ω-Frequency, μ_c-Chemical potential, Γ-Carrier scattering rate and T-Temperature;
- ✓ Where Fermi-Dirac distribution f_d is $f_d(\epsilon)=1/\left(e^{(\epsilon-\mu_c)/k_BT}+1\right)$
- ✓ Converts the surface conductivity to volume conductivity in modeling;

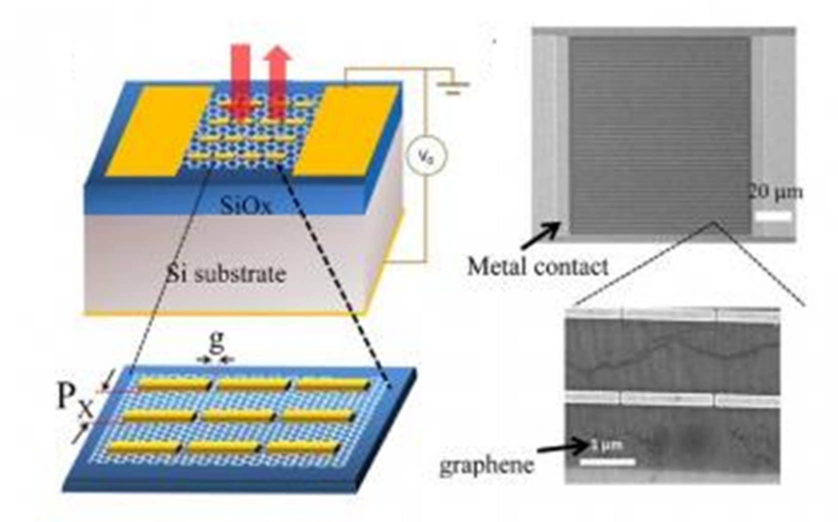
$$\tilde{\sigma}=\sigma/d_0 \quad \varepsilon_r(\omega)=1-j\tilde{\sigma}/\omega\varepsilon_0$$

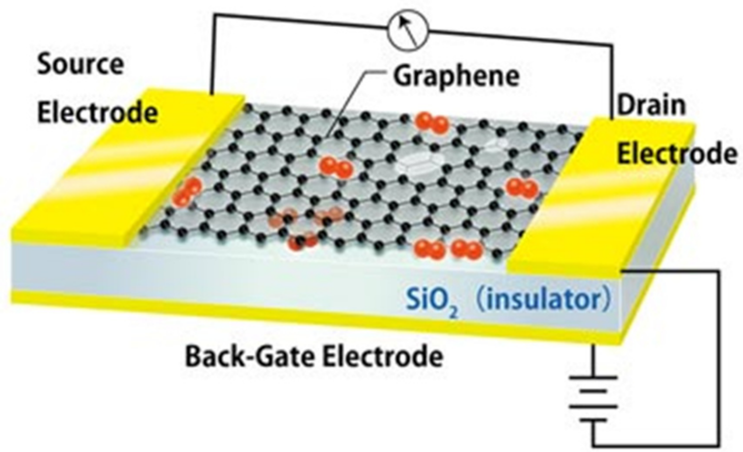
WHEN GRAPHENE IS APPLIED TO CONTROL SPONTANEOUS EMISSION

Quantum Electrodynamics ⟷ Computational Electromagnetics

Spontaneous Emission ⟷ Numerical Green's Function

Low-Dimensional Materials
Electrically Tunable Active Materials

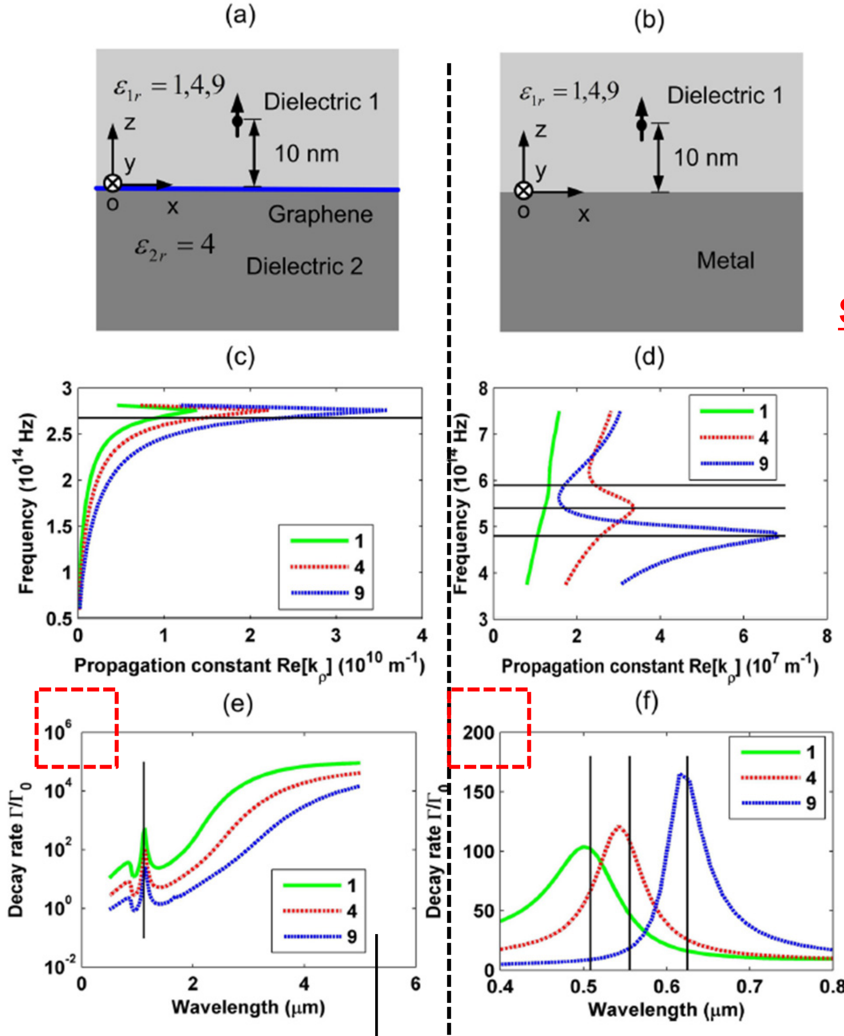




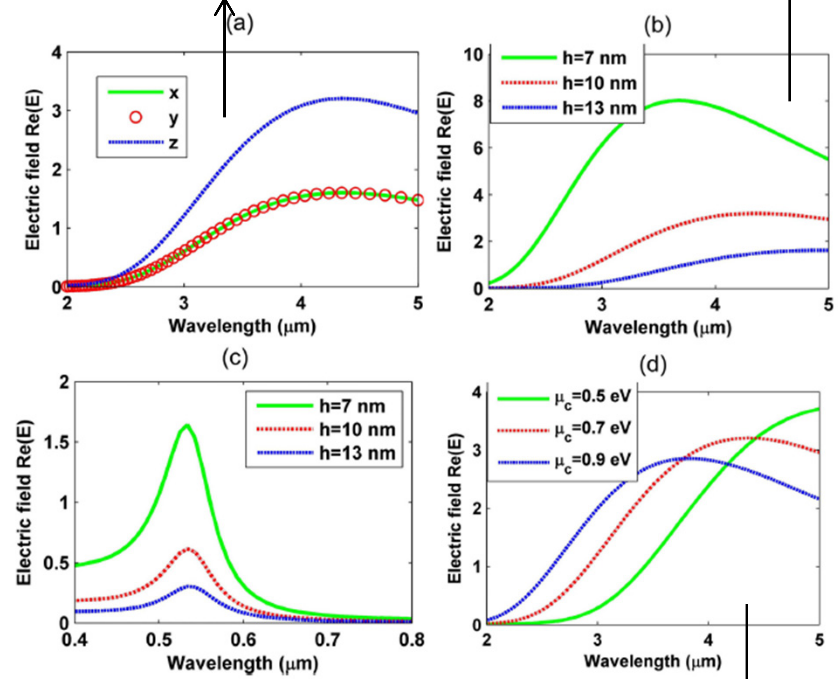
GRAPHENE VS METAL FOR CONTROLLING SPONTANEOUS EMISSION

$$\frac{\omega\varepsilon_1 k_{2z}}{(\omega\varepsilon_2-\sigma k_{2z})k_{1z}}+1=0. \quad k_\rho=\sqrt{k^2-\left(\frac{2\omega\varepsilon}{\sigma}\right)^2}.$$

$$k_\rho=\frac{\omega}{c}\sqrt{\frac{\varepsilon_{1r}\varepsilon_{2r}}{\varepsilon_{1r}+\varepsilon_{2r}}}.$$



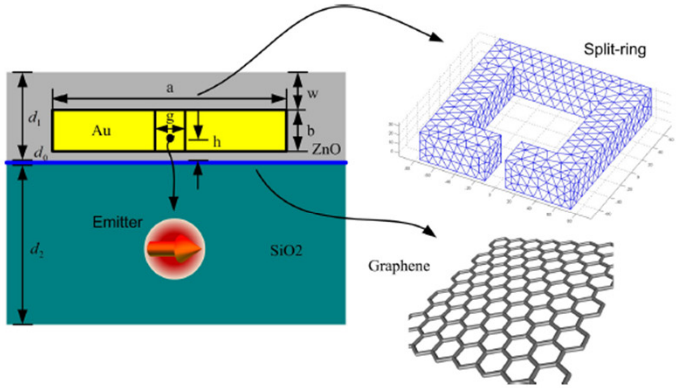
sensitive to polarization sensitive to position



much larger enhancement than metals

tunable by chemical potential

COMPUTATIONAL ELECTROMAGNETICS METHODOLOGY



Multilayered medium Green's function

$$\bar{\mathbf{G}}_e(\mathbf{r},\mathbf{r}') = \bar{\mathbf{G}}_e^{\mathrm{TE}}(\mathbf{r},\mathbf{r}') + \frac{1}{k_{nm}^2}\bar{\mathbf{G}}_e^{\mathrm{TM}}(\mathbf{r},\mathbf{r}')$$

$$\bar{\mathbf{G}}_e^{\mathrm{TE}}(\mathbf{r},\mathbf{r}') = (\nabla\times\hat{z})(\nabla'\times\hat{z})\frac{i}{4\pi}\int_0^\infty dk_\rho J_0(k_\rho\rho)\frac{F^{\mathrm{TE}}(k_\rho,z,z')}{k_{mz}k_\rho}$$

$$\bar{\mathbf{G}}_e^{\mathrm{TM}}(\mathbf{r},\mathbf{r}') = (\nabla\times\nabla\times\hat{z})\,(\nabla'\times\nabla'\times\hat{z})\,\frac{i}{4\pi}\int_0^\infty dk_\rho J_0(k_\rho\rho)\frac{F^{\mathrm{TM}}(k_\rho,z,z')}{k_{mz}k_\rho}$$

PMCHWT formulation

$$\left[\begin{array}{c} -\mathbf{E}_{\mathrm{inc}}^o \\ -\mathbf{H}_{\mathrm{inc}}^o \end{array}\right]\Bigg|_{\mathrm{tan}} = \left[\begin{array}{cc} (\mathscr{L}_E^o+\mathscr{L}_E^i) & (\mathscr{K}_E^o+\mathscr{K}_E^i) \\ (\mathscr{K}_H^o+\mathscr{K}_H^i) & (\mathscr{L}_H^o+\mathscr{L}_H^i) \end{array}\right]\cdot\left[\begin{array}{c} \mathbf{J} \\ \mathbf{M} \end{array}\right]\Bigg|_{\mathrm{tan}}$$

$$\mathbf{E}(\mathbf{r}) = \mathscr{L}_{\mathrm{E}}(\mathbf{r},\mathbf{r}')\cdot\mathbf{J}(\mathbf{r}') + \mathscr{K}_{\mathrm{E}}(\mathbf{r},\mathbf{r}')\cdot\mathbf{M}(\mathbf{r}')$$

$$\mathbf{H}(\mathbf{r}) = \mathscr{L}_{\mathrm{H}}(\mathbf{r},\mathbf{r}')\cdot\mathbf{M}(\mathbf{r}') + \mathscr{K}_{\mathrm{H}}(\mathbf{r},\mathbf{r}')\cdot\mathbf{J}(\mathbf{r}')$$

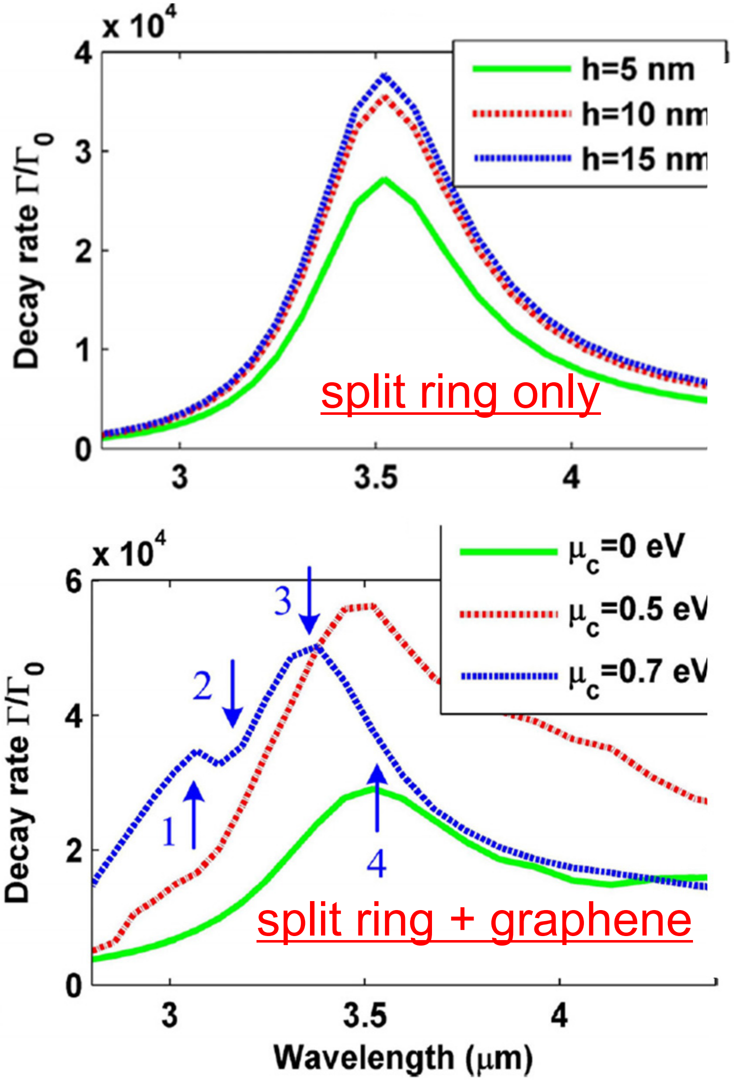
$$\mathscr{L}_{\mathrm{E}}(\mathbf{r},\mathbf{r}')\cdot = i\omega\int d\mathbf{r}'\bar{\mathbf{G}}_e(\mathbf{r},\mathbf{r}')\mu(\mathbf{r}')$$

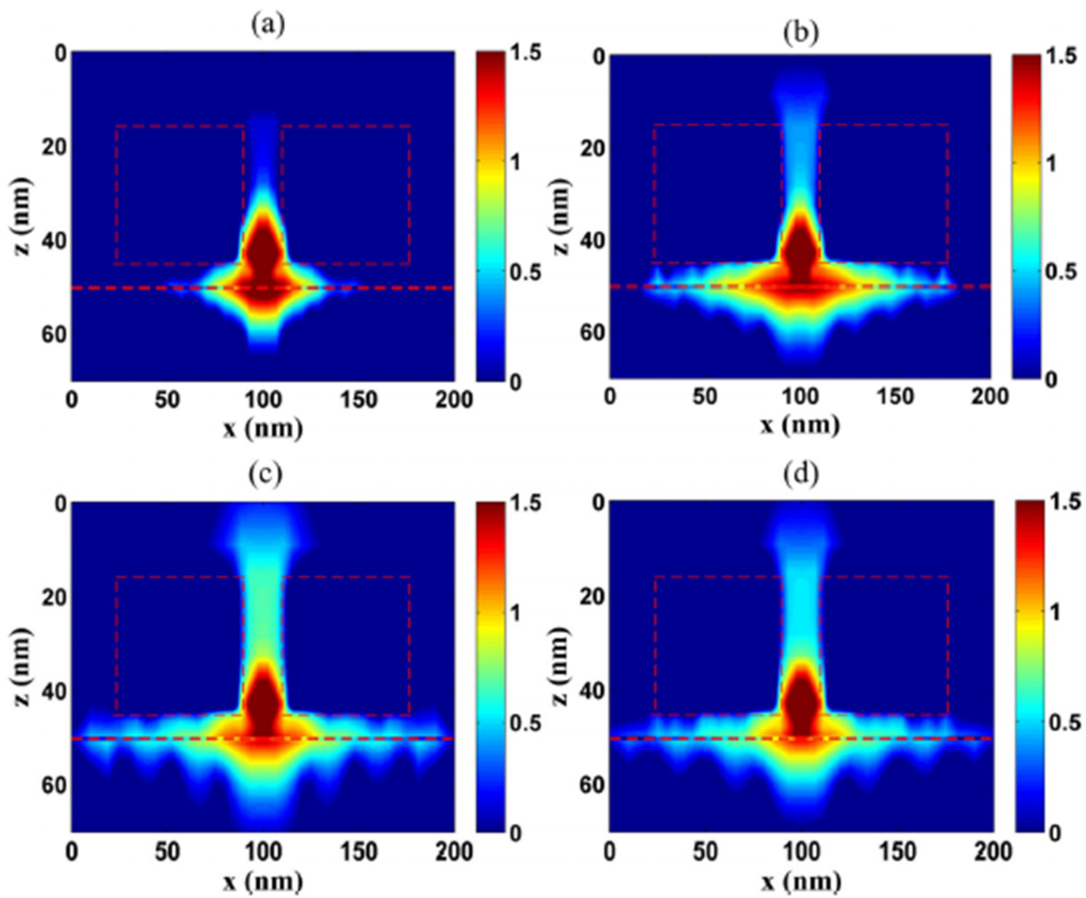
$$\mathscr{K}_{\mathrm{H}}(\mathbf{r},\mathbf{r}')\cdot = \mu^{-1}(\mathbf{r})\int d\mathbf{r}'\nabla\times\bar{\mathbf{G}}_e(\mathbf{r},\mathbf{r}')\mu(\mathbf{r}')$$

$$\mathscr{L}_{\mathrm{H}}(\mathbf{r},\mathbf{r}')\cdot = i\omega\int d\mathbf{r}'\bar{\mathbf{G}}_m(\mathbf{r},\mathbf{r}')\varepsilon(\mathbf{r}')$$

$$\mathscr{K}_{\mathrm{E}}(\mathbf{r},\mathbf{r}')\cdot = -\varepsilon^{-1}(\mathbf{r})\int d\mathbf{r}'\nabla\times\bar{\mathbf{G}}_m(\mathbf{r},\mathbf{r}')\varepsilon(\mathbf{r}')$$

SPONTANEOUS EMISSION IN COMPLEX MULTILAYER NANOSTRUCTURE





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CONCLUSION

- 1. Graphene offers several flexible tuning routes for manipulating EMLDOS, including tunable chemical potential and the emitter's position and polarization. It shows broadband enhancements of EMLOS compared to metal materials.

- 2. We study spontaneous emission rate of a quantum emitter near a metallic split-ring resonator, which is embedded in a multilayered substrate incorporating a graphene layer. This design enables a mutual interaction between graphene plasmonics and metallic plasmonics. The boundary element method with a multilayered medium Green's function is adopted in the numerical simulation.

- 3. Strong plasmonic coupling with a switch on-off feature was observed, which is helpful to dynamically manipulate spontaneous emission rate in complex optical devices.

THANKS FOR YOUR ATTENTION!