Nanoantennas to Control the Dynamics of Quantum Systems

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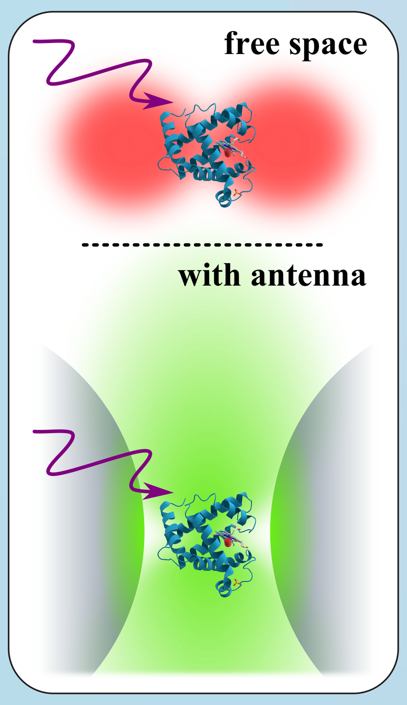
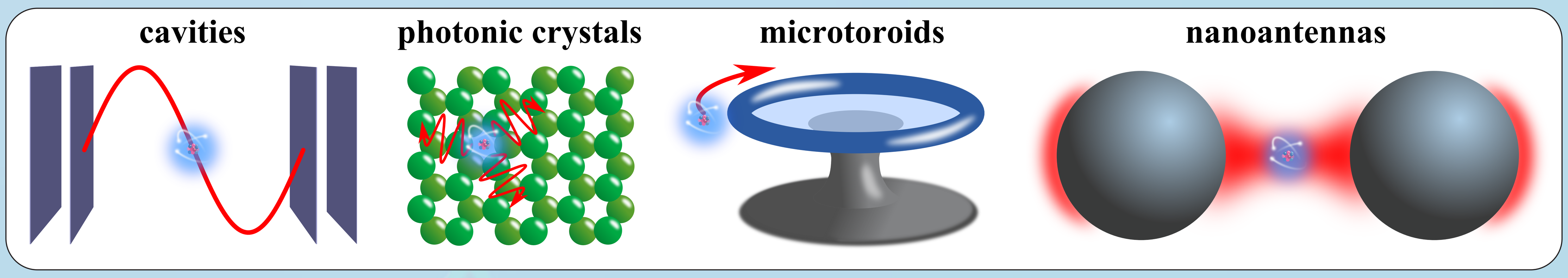
Why Nanoantennas?

- **nanoantennas:** change interaction of light with quantum systems two-fold[1]:
  absorption **and** emission
- **control entire dynamics**, e.g.
  - excitation of **dipole-forbidden** transitions and subsequent **luminescence** processes[2]
  - **selective** and **tunable** enhancement of THz emissions[3]

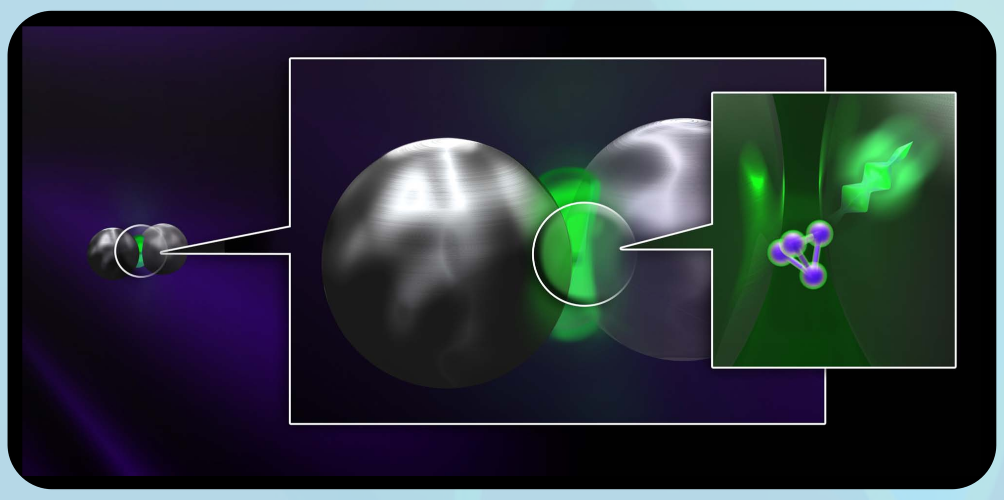


- **absorption**: strong **field enhancement**, nanoscale **localization**, **parameter sensitivity**
- **emission**: adaptable **directivity** and altered **radiation rates**[1-4]
  - spontaneous emission rate: **Purcell factor** $F = \gamma^{a}_{rad}/\gamma^{fs}_{rad}$ - from cavities to nanoantennas
  - quality factor Q, mode volume V, wavelength λ_m: $F = \frac{3}{4\pi^2}\left(\frac{\lambda_m^3}{V}\right)\cdot Q$



Forbidden Transitions

- a nanoantenna hugely enhances **dipole-forbidden excitation rate**
- subsequent **luminescence** depends strongly on **internal dynamics**



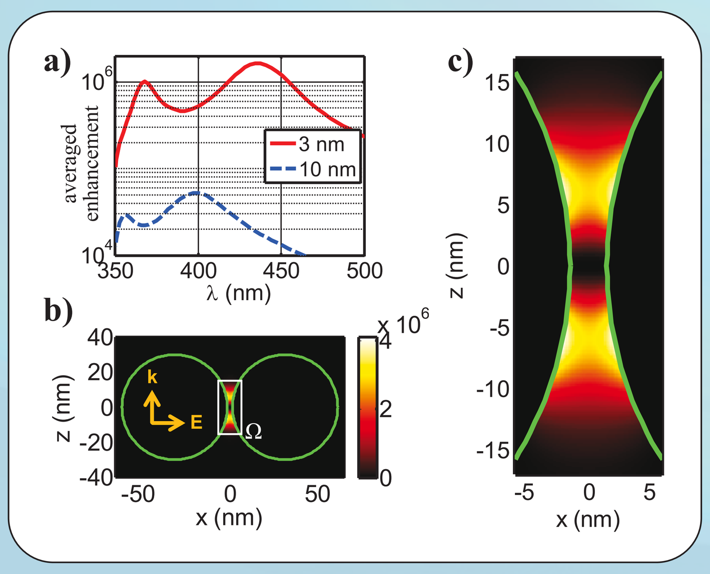
Left: a plane wave scatters a silver dimer. Middle: molecules in-between undergo a quadrupolar excitation resulting in a luminescence at a lower wavelength. Right: The whole process for a single molecule.

Quadrupolar Excitation Enhancement

- higher order field components vs. free space: **huge enhancement**
- field decomposition, Golden Rule

$$\mathbf{E}(\mathbf{x},\omega) = \sum_{m,n}\left[p_{mn}(\omega;\mathbf{r}_0)\,\mathbf{N}_{mn}(\mathbf{x}-\mathbf{r}_0,\omega) + q_{mn}(\omega;\mathbf{r}_0)\,\mathbf{M}_{mn}(\mathbf{x}-\mathbf{r}_0,\omega)\right]$$

$$\Gamma_{ij}(\mathbf{r}_0) = \frac{2\pi e^2}{\hbar^2}\left|\sum_{n,m} p_{mn}(\mathbf{r}_0)\,\langle i|\,\mathbf{N}_{mn}\cdot\mathbf{x}\,|j\rangle\right|^2 \delta(\omega_{ij}\pm\omega)$$



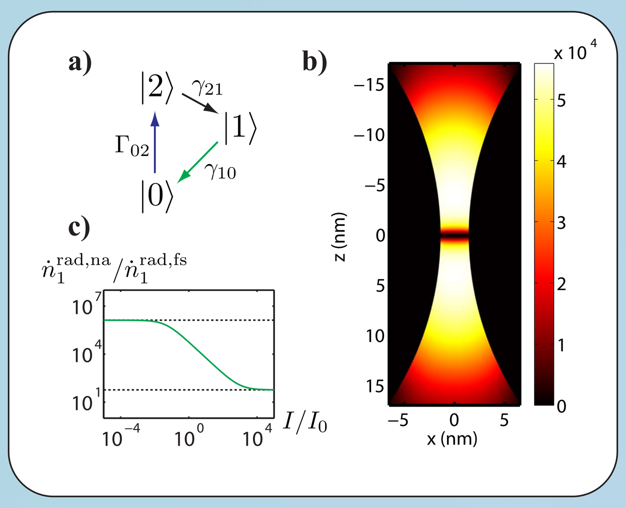
Can we observe this excitation enhancement also in luminescence?

Luminescence Enhancement

- **three-level-system**, stationary solution:

$$\frac{\dot{n}_1^{rad,na}}{\dot{n}_1^{rad,fs}} = \underbrace{\frac{\gamma_{10}^{rad,na}}{\gamma_{10}^{rad,fs}}\cdot\frac{\Gamma_{02}^{na}}{\Gamma_{02}^{fs}}}_{\text{Purcell effect and quadrupole enhancement}} \cdot \underbrace{\frac{\gamma_{10}^{fs}\gamma_{21}+\gamma_{21}\Gamma_{02}^{fs}+\gamma_{10}^{fs}\Gamma_{02}^{fs}}{\gamma_{10}^{na}\gamma_{21}+\gamma_{21}\Gamma_{02}^{na}+\gamma_{10}^{na}\Gamma_{02}^{na}}}_{\text{dynamics of quantum system}}$$

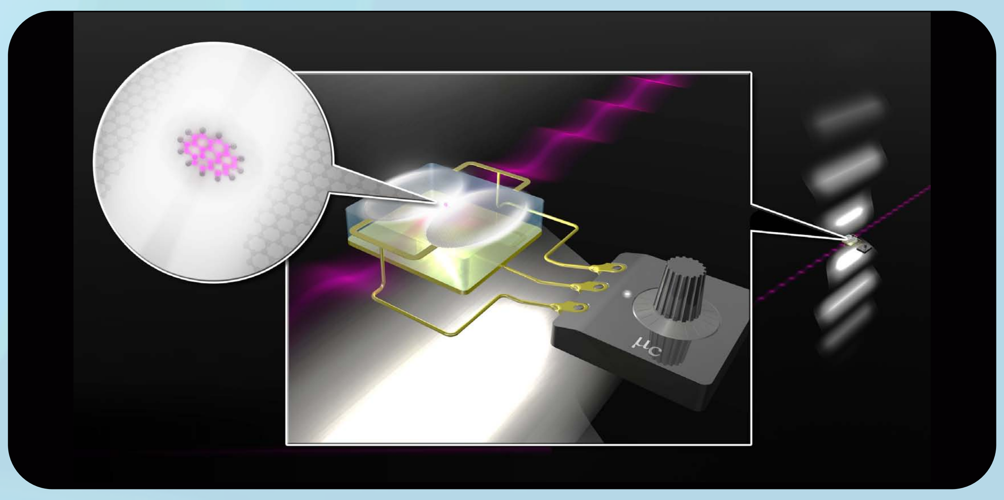
- limiting cases for excitation **intensities**
  - full enhancement only for **weak excitation**
  - strong excitation: internal dynamics form **bottleneck** - enhancement saturates



Internal dynamics are necessary to understand measurables!

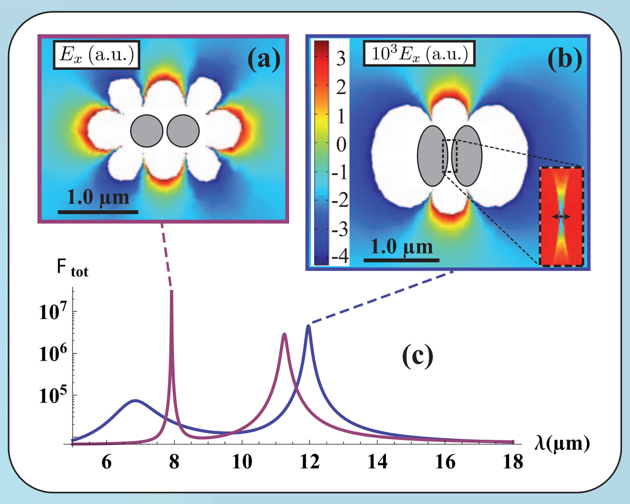
Selective Enhancement of THz Emissions

- **graphene antenna** greatly enhances the emission at specific frequencies
- **tunability** enables **selective enhancement** of weak transitions



Efficient Graphene Antenna

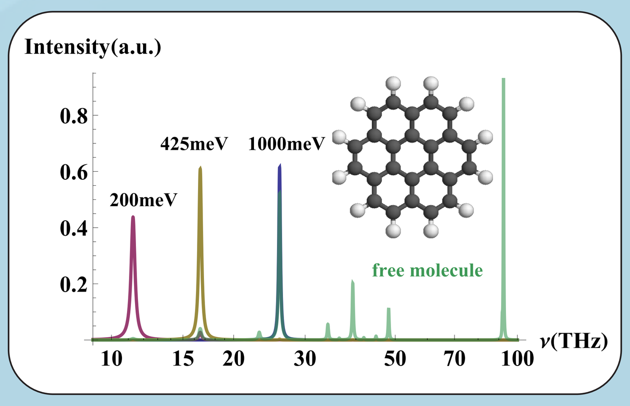
- radiating molecule between elements
- want: high single-frequency **Purcell effect**
- problem: coupling to **higher order modes**
- solution: two **elliptical elements**
  - distinctly enhanced dipole mode, **efficient**



The Changed Spectrum of Coronene

- ultrafast **internal redistribution**
- hugely enhance specific emission - any weak transition can become dominant

$$\mathcal{F}_{rad}(\omega_i) = \eta(\omega_i)\cdot F_{tot}(\omega_i)\cdot\frac{\sum_k \hbar\omega_k\gamma_{tot}^{fs}(\omega_k)\,B(\omega_k;T_m)}{\sum_k \hbar\omega_k F_{tot}(\omega_k)\,\gamma_{tot}^{fs}(\omega_k)\,B(\omega_k;T_m)}.$$



Conclusions

- nanoantennas can control **absorption and emission** of quantum systems
- **understanding altered dynamics** necessary for any measurement

References

1 P. Bharadwaj et al., Adv. Opt. Phot. **1** (2009), A. G. Curto et al., Science **20** (2010), P. Anger, et al., Phys. Rev. Lett. **96** (2006)
2 R. Zurita-Sánchez et al, J. Opt. Soc. Am. B **19** (2002), R. Filter et al., Phys. Rev. B **86** (2012)
3 R. Filter et al., under review
4 T. J. Kippenberg et al., Phys. Rev. Lett. **103** (2009), Jorgensen et al., Phys. Rev. Lett. **107** (2011)