

Nanoantennas to Control the Dynamics of Quantum Systems

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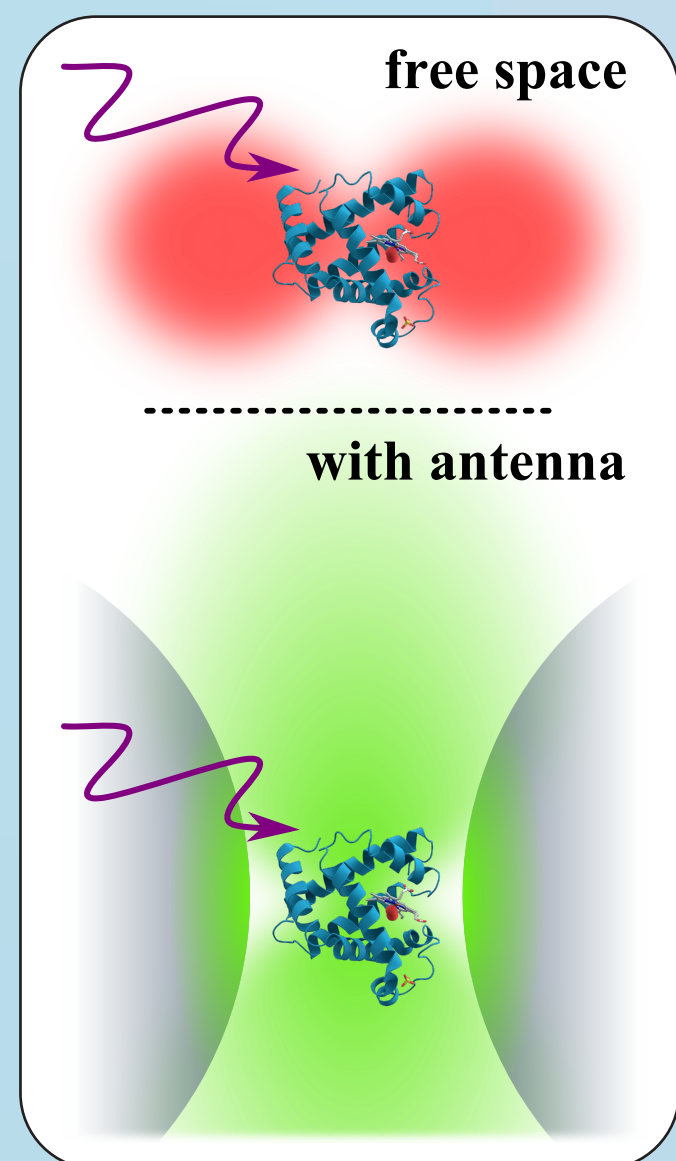
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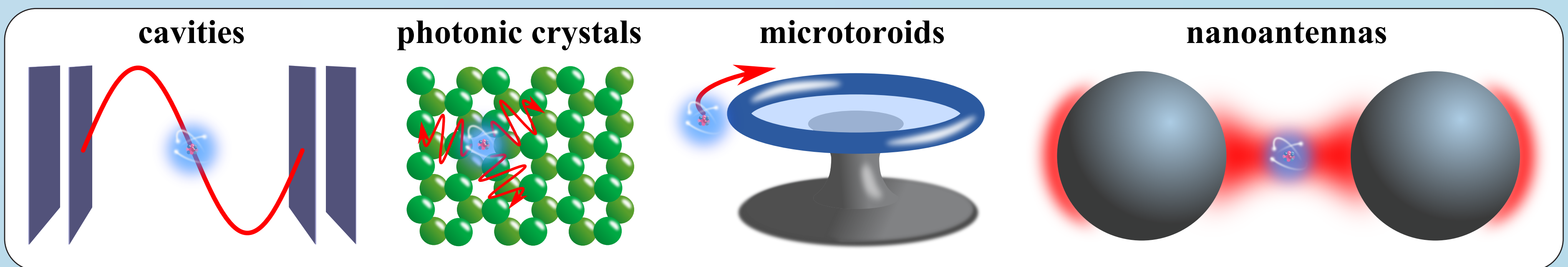
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Why Nanoantennas?

- **nanoantennas**: change interaction of light with quantum systems two-fold¹:
absorption **and** emission
- **control entire dynamics**, e.g.
 - excitation of **dipole-forbidden** transitions and subsequent **luminescence** processes²
 - **selective and tunable** enhancement of THz emissions³

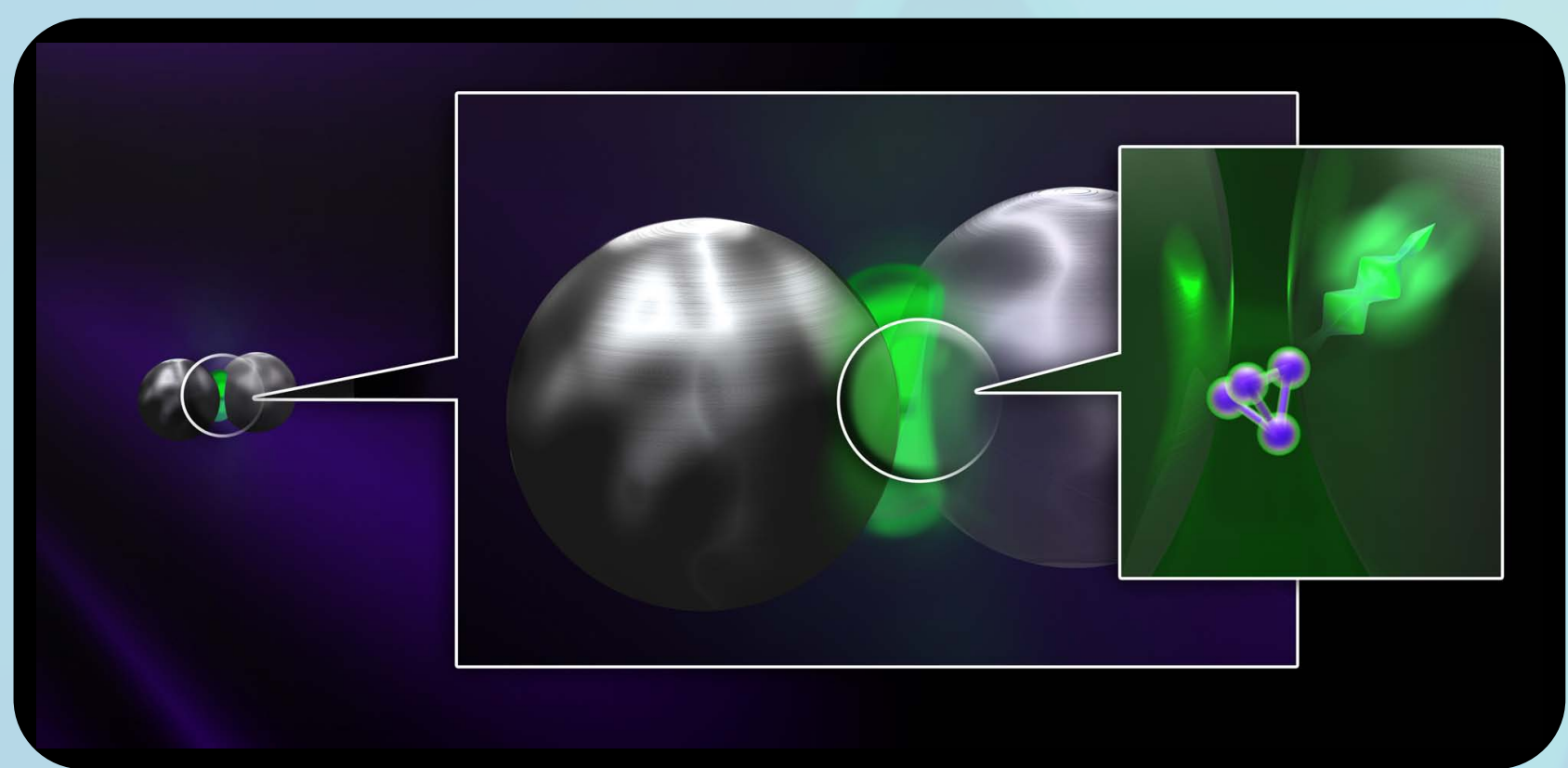


- **absorption**: strong **field enhancement**, nanoscale **localization**, **parameter sensitivity**
- **emission**: adaptable **directivity** and altered **radiation rates**¹⁻⁴
 - spontaneous emission rate: **Purcell factor** $F = \gamma_{\text{rad}}^{\text{a}} / \gamma_{\text{rad}}^{\text{fs}}$ - 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**



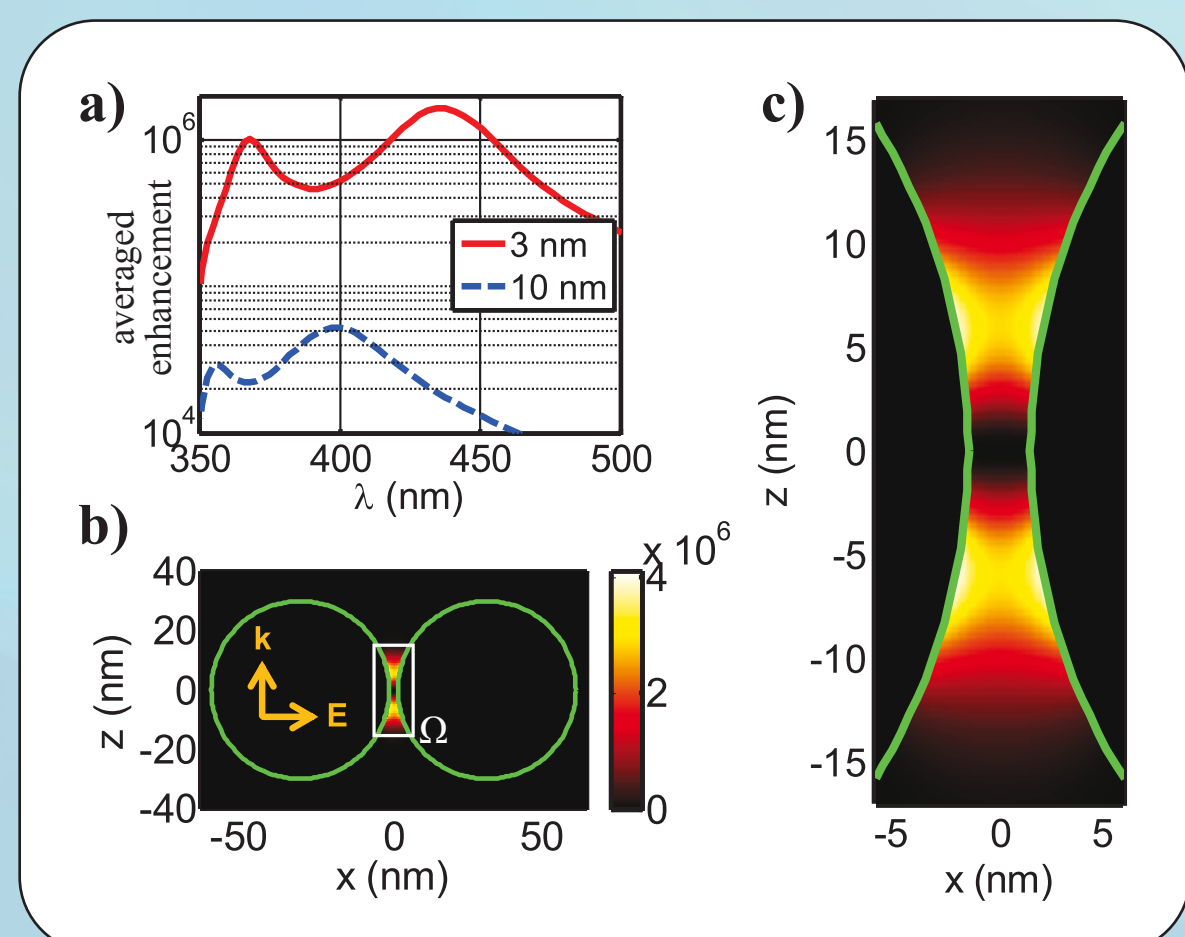
Left: a plane wave scatters a silver dimer. Middle: molecules in-between undergo a quadrupolar excitation resulting in a luminescence at a lower wavelength. Right: The whole process for a single molecule.

Quadrupolar Excitation Enhancement

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

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

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



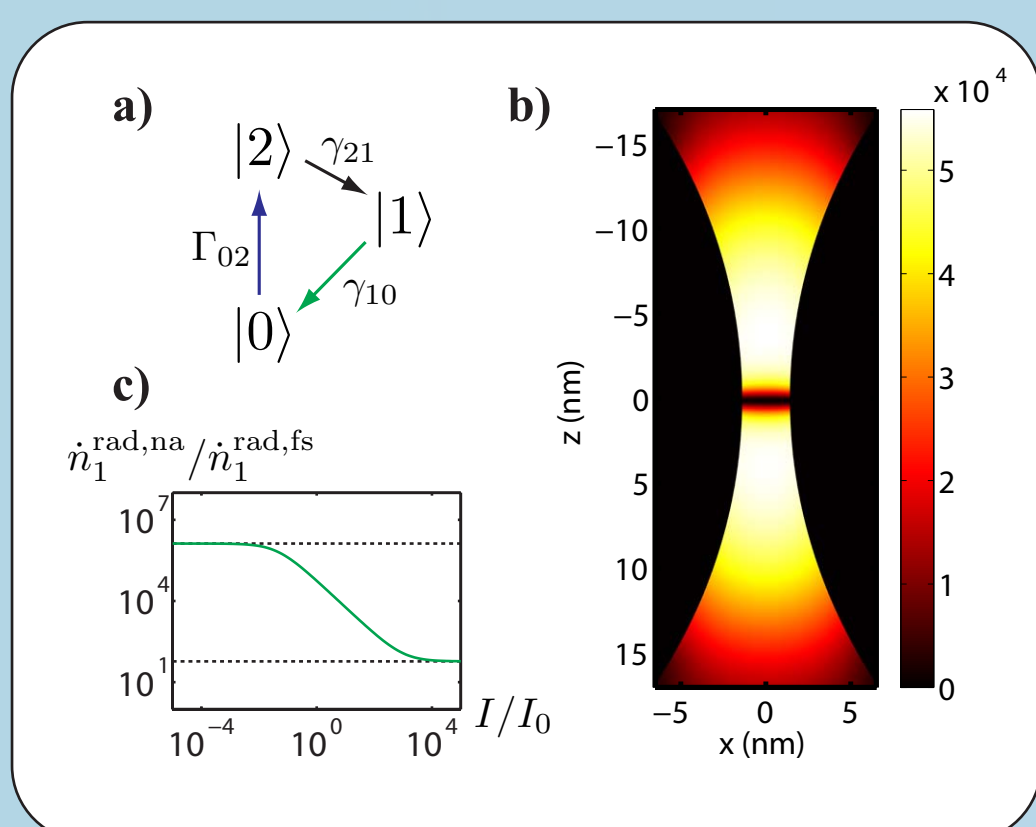
Can we observe this excitation enhancement also in luminescence?

Luminescence Enhancement

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

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

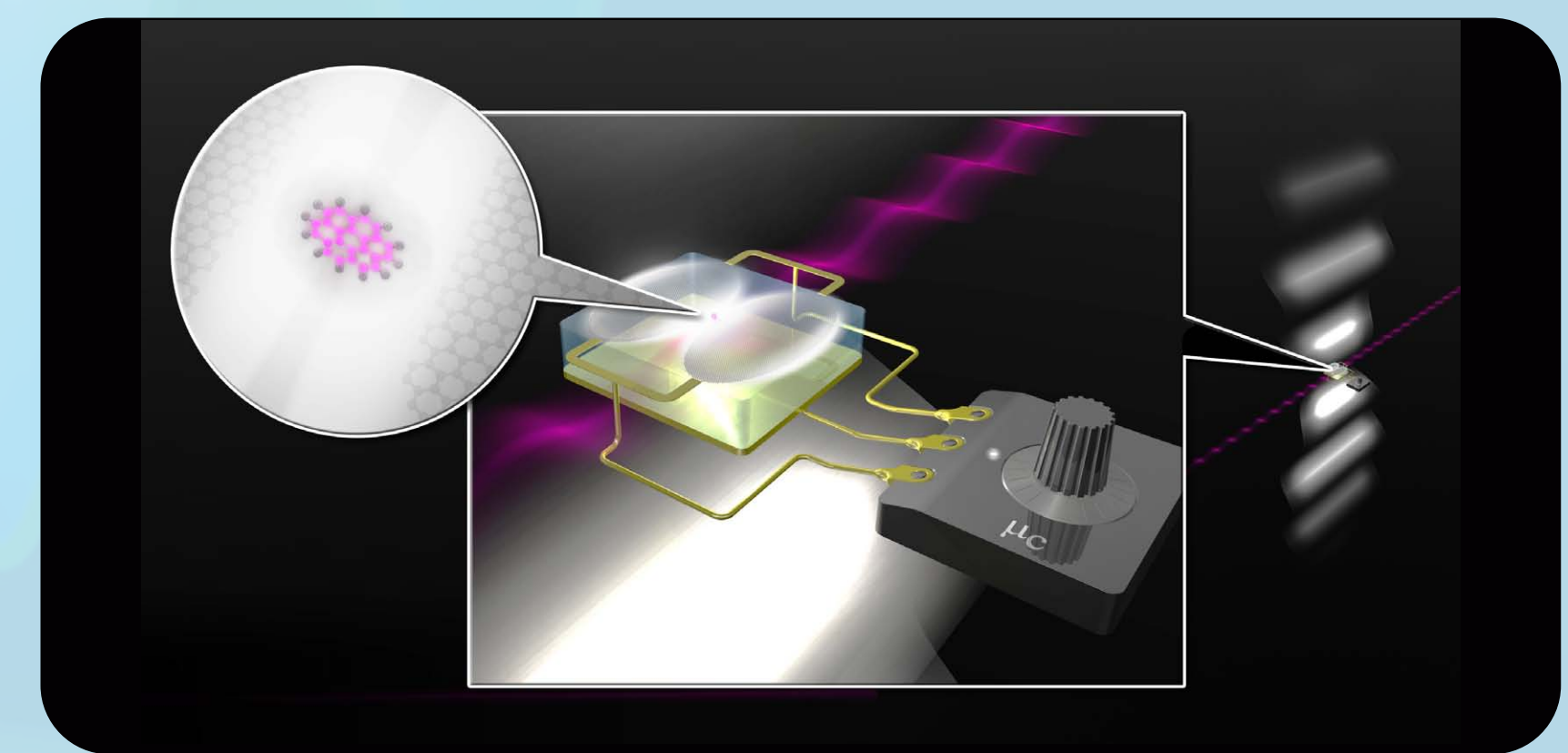
- limiting cases for excitation **intensities**
- full enhancement only for **weak excitation**
- strong excitation: internal dynamics form **bottleneck** - enhancement saturates



Internal dynamics are necessary to understand measurables!

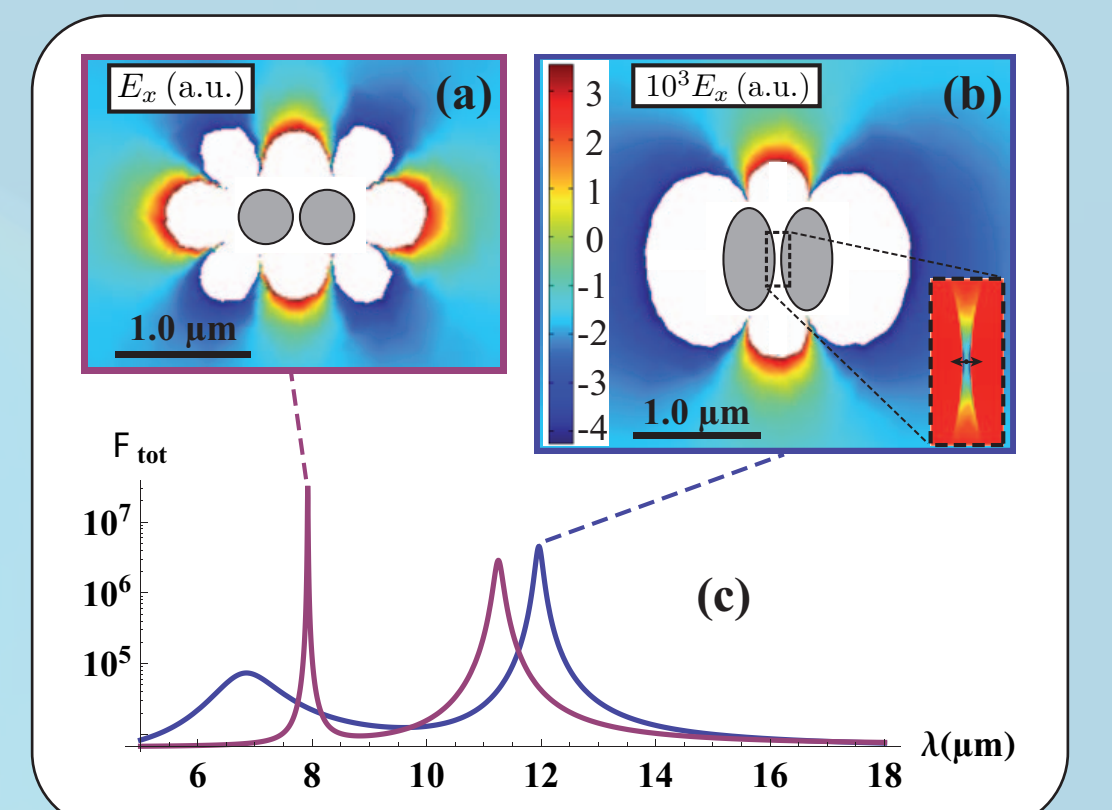
Selective Enhancement of THz Emissions

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



Efficient Graphene Antenna

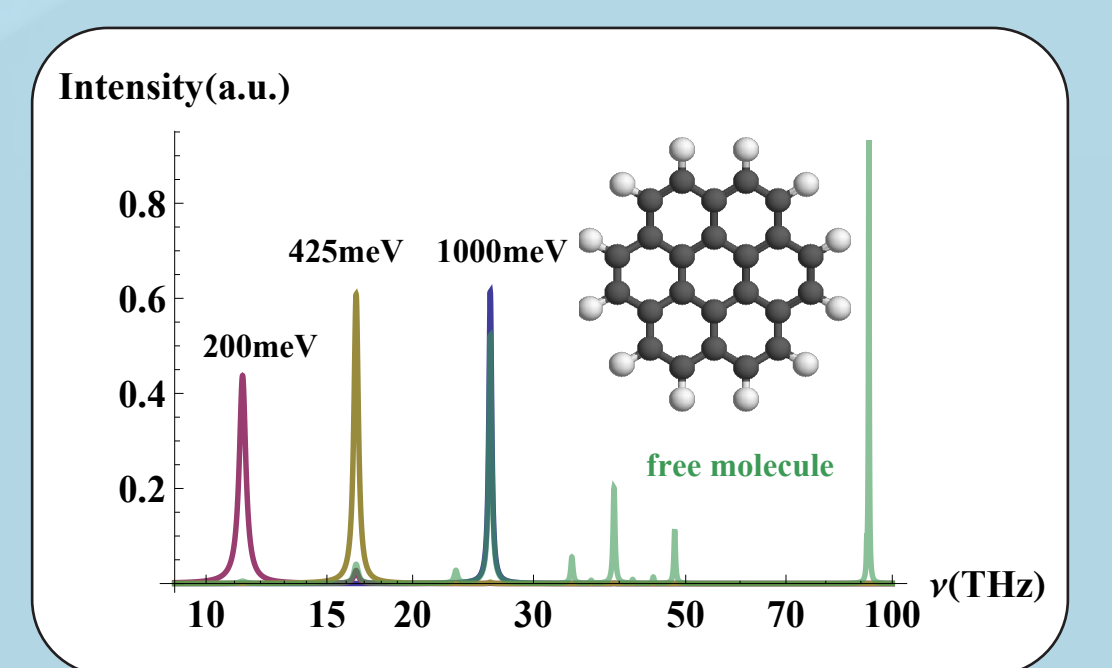
- radiating molecule between elements
- want: high single-frequency **Purcell effect**
- problem: coupling to **higher order modes**
- solution: two **elliptical elements**
- distinctly enhanced dipole mode, **efficient**



The Changed Spectrum of Coronene

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

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



Conclusions

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

References

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