

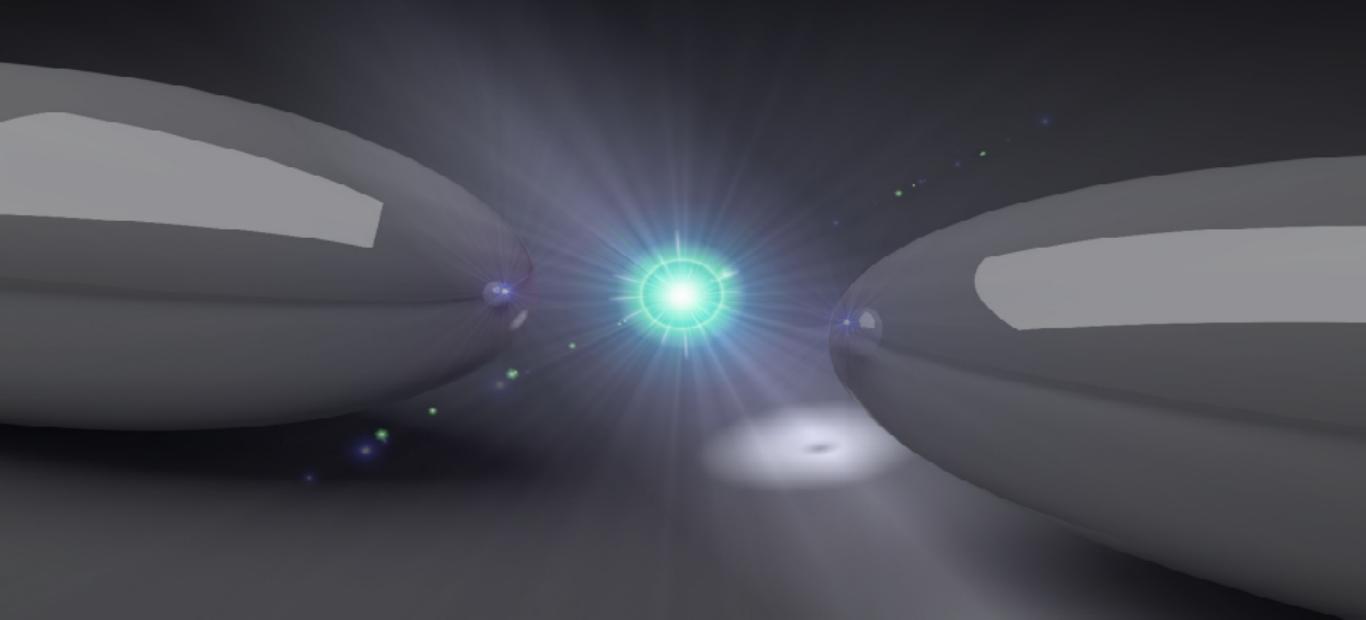
Nanoantennas for light-matter coupling in multiple parallel channels

K. Słowiak[†], E. Rusak^{*}, P. Gładysz[†], J. Straubel^{*}, M. Kühn^{*}, F. Weigend^{*}, and C. Rockstuhl^{*}

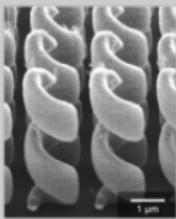
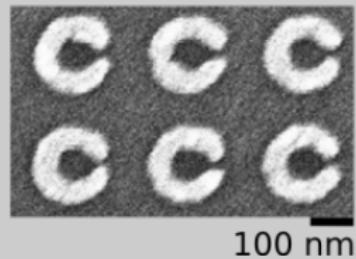
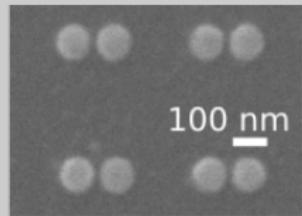
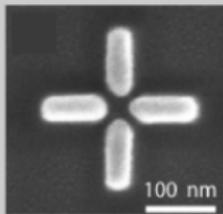
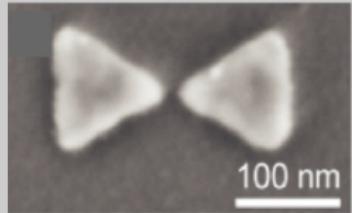
[†] Nicolaus Copernicus University, Toruń, Poland

^{*} Karlsruhe Institute of Technology, Karlsruhe, Germany

NANOLIGHT 2018, BENASQUE

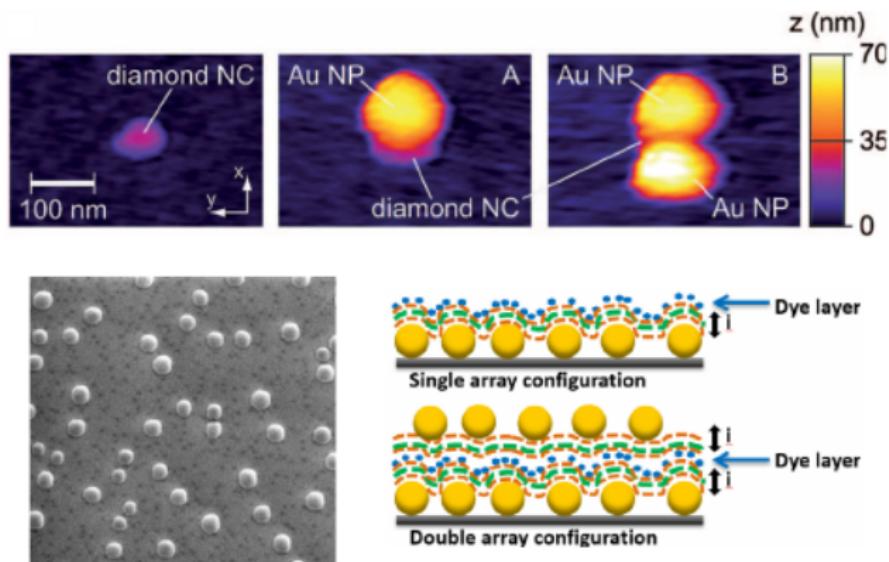


Nanoantennas are now feasible



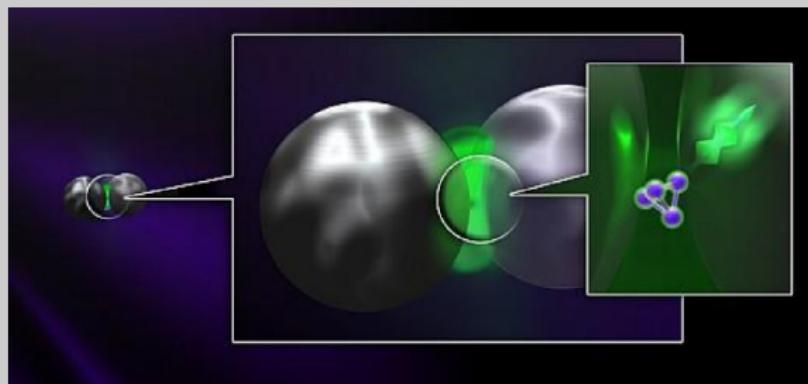
triangles: B. Roxworthy et al., Sc. Rep. (2012); cross: Huang et al., Nat. Commun. (2010);
dimers: S. Aćimović, ACS Nano (2009); split rings: A. Clark. et al., J. Am. Chem. Soc. (2009);
helices: J. Gansel et al., Science (2009); rods: courtesy of S. Maćkowski, NCU Toruń;

Quantum emitters can be positioned
at nanoantenna vicinity

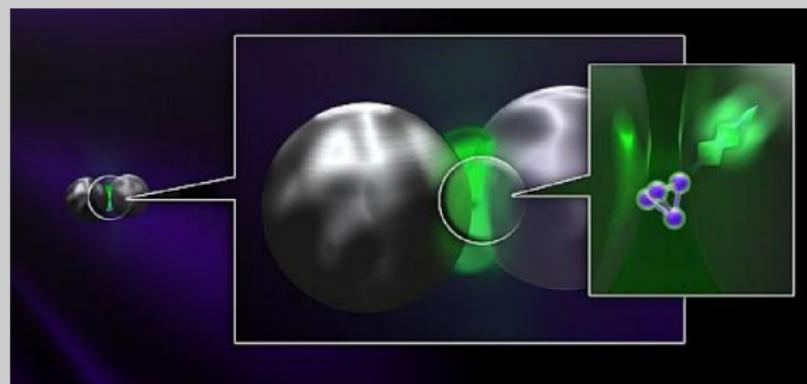


NVCs: Schietinger et al., Nano Letters (2009); QDs & In islands: courtesy of G. Khitrova, Univ. of Arizona;
gold spheres & dyes: Chekini et al., J. Appl. Phys. (2015)

Nanoantennas focus light



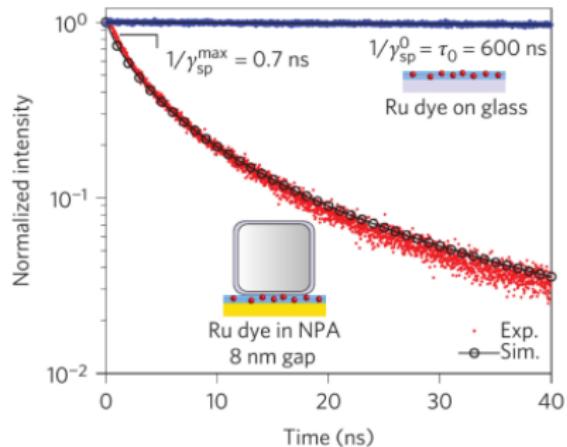
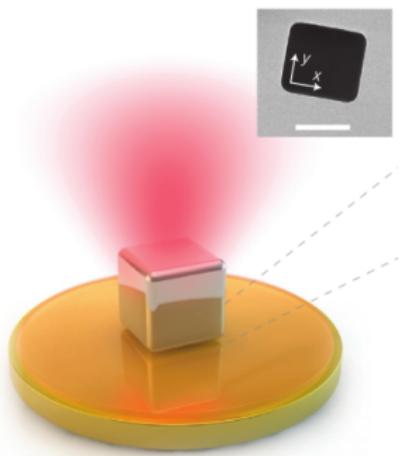
Nanoantennas speed up light-matter interactions



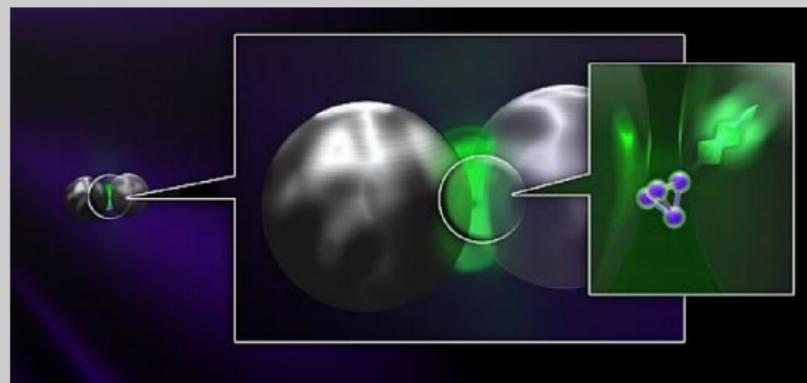
interaction
dipole approximation

$$\mathcal{V} = -\mathbf{d} \cdot \mathbf{E}$$

Nanoantennas modify molecular lifetime



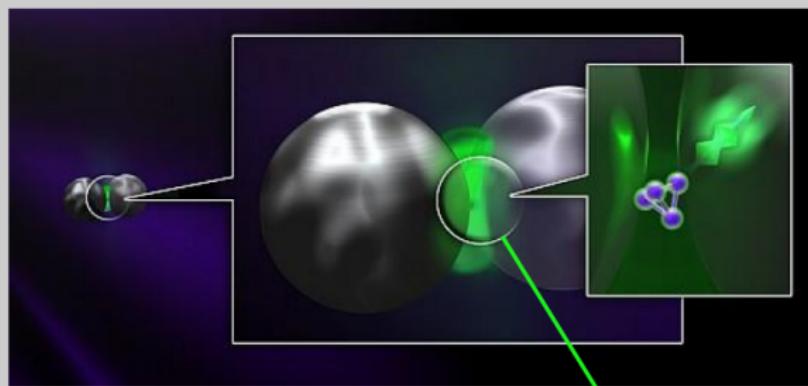
Nanoantennas modify molecular lifetime



interaction
dipole approximation

$$\mathcal{V} = -\mathbf{d} \cdot \mathbf{E}$$

Nanoantennas modify molecular lifetime



$\lambda/10$

**interaction
beyond** dipole appr.

$$\begin{aligned}\mathcal{V} = & -\mathbf{d} \cdot \mathbf{E} \\ & -\mathbf{m} \cdot \mathbf{B} - [\mathbf{Q} \nabla] \cdot \mathbf{E}\end{aligned}$$

multiple pathways
interference effects

State of the art

magnetic dipole

Feng et al., Opt. Lett. (2011): 2 gold patches
Schmidt et al., Opt. Expr. (2012): dielectric disks
Hein & Giessen, PRL (2013): **split ring** resonators
Mivelle et al., ACS Phot. (2015): **diabolo** nanoantenna

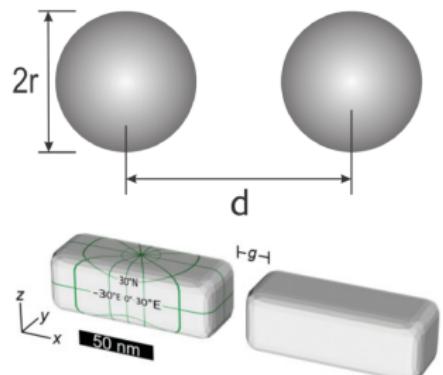
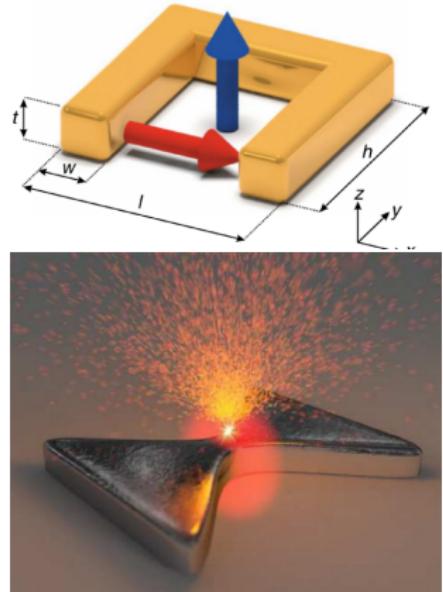
electric quadrupole

Filter et al., PRB (2012): **dimer** nanoantenna (2 spheres)
Kern & Martin, PRA (2012): **dimer** nanoantenna (2 nanorods), Cs atoms
Yannopapas & Paspalakis, J. Mod. Opt. (2015): core-shell arrays

combined (Green's function approach)

Tighineanu et al., PRL (2014): QD & nanowire
Cotrufo & Fiore, PRB (2015): QD & photonic crystal, nanorods
Yang & An, PRA (2016): QD & planar interface

M. Kosik, maser thesis, NCU (2017)



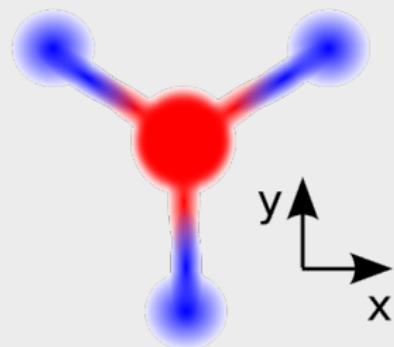
Transition rates beyond dipole approximation

$$\Gamma = \frac{2\pi}{\hbar^2} |\langle f | \mathcal{V} | i \rangle|^2 \rho (\omega_i - \omega_f)$$

$$\mathcal{V} = -\mathbf{d} \cdot \mathbf{E}(\mathbf{r}_m) - \mathbf{m} \cdot \mathbf{B}(\mathbf{r}_m) - [\mathbf{Q} \nabla] \cdot \mathbf{E}(\mathbf{r}_m)$$

molecular properties	field distribution	transition rate
$\mathbf{d}, \mathbf{m}, \mathbf{Q}$	$\mathbf{E}(\mathbf{r}_m), \mathbf{B}(\mathbf{r}_m)$	Γ
TDDFT (TURBOMOLE)	Maxwell's eqs. (MNPBEM, etc.)	Fermi's golden rule

Exemplary molecules



Dipole-forbidden transitions:

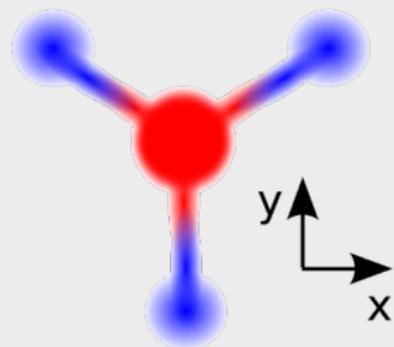
- ▶ OsO₃ @ 553 nm

$$m_y = 0.84 \mu_B, Q_{xz} = 0.66 \text{ B}$$

$$m_x = 0.84 \mu_B, Q_{yz} = 0.66 \text{ B}$$

Data source: DFT calculations, F. Weigend i M. Kühn, KIT

Exemplary molecules



Dipole-forbidden transitions:

- ▶ OsO₃ @ 553 nm

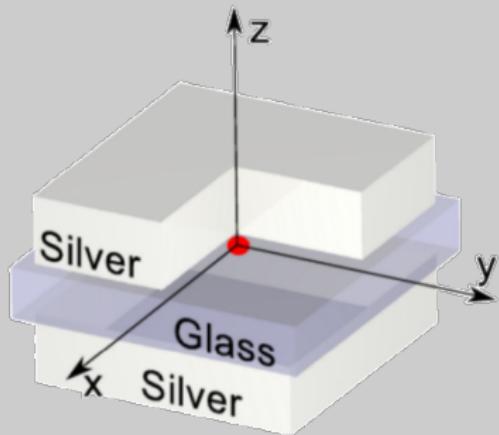
$$m_y = 0.84 \mu_B, Q_{xz} = 0.66 \text{ B}$$

$$m_x = 0.84 \mu_B, Q_{yz} = 0.66 \text{ B}$$

Data source: DFT calculations, F. Weigend i M. Kühn, KIT

Goal: a nanoantenna to control MD & EQ interactions

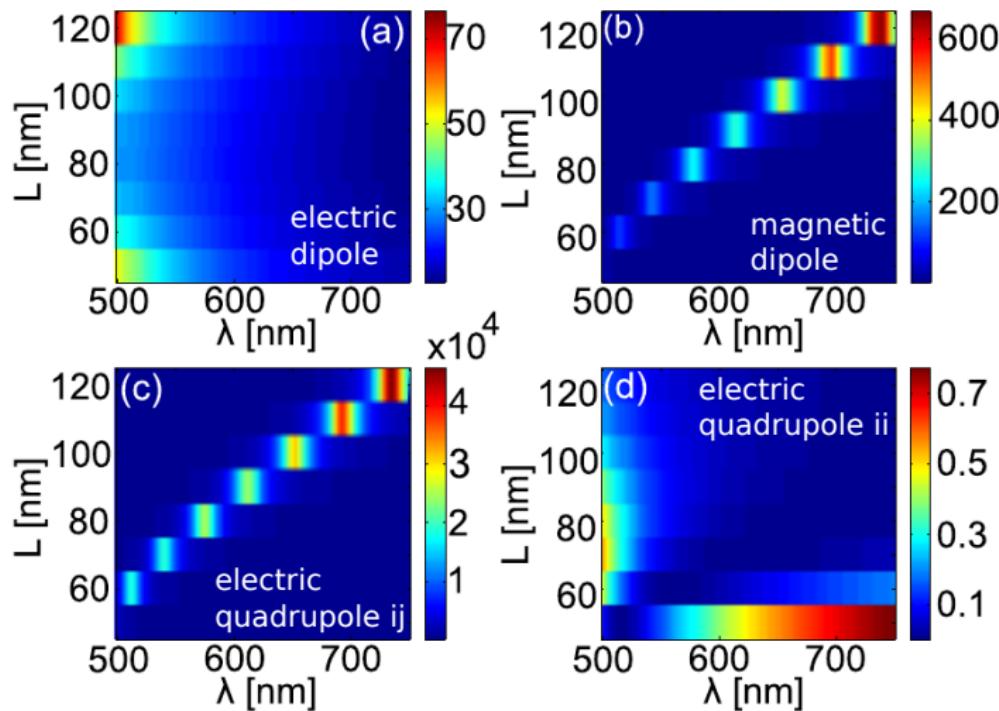
Silver nanoantenna to enhance MD & EQ interactions



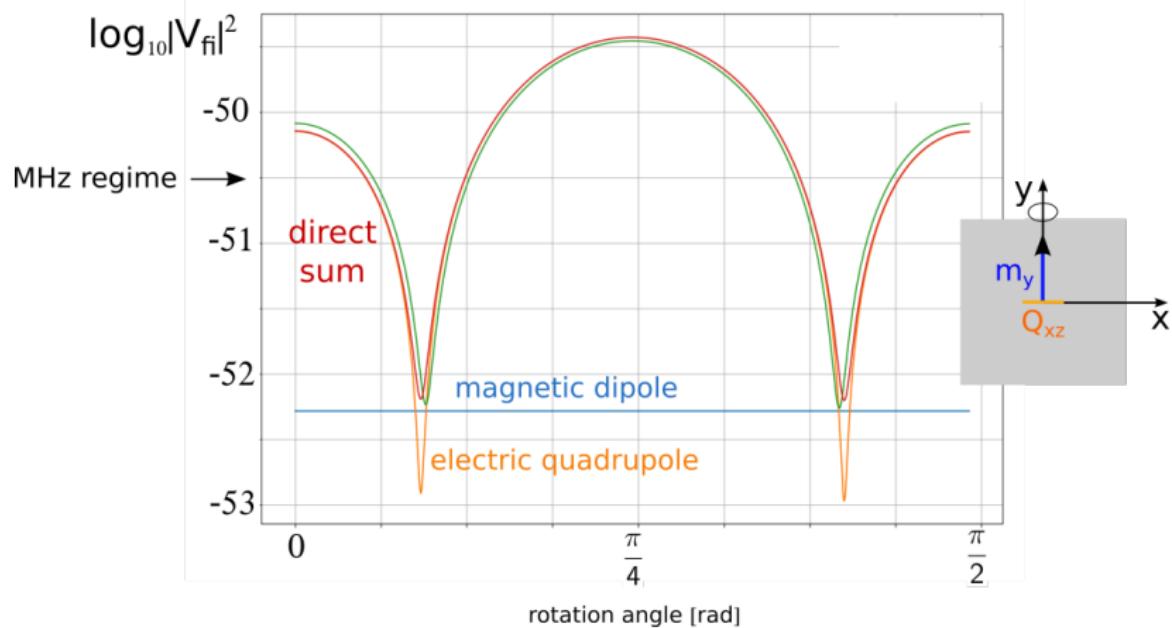
Double-patch nanoantenna:

- ▶ material: **silver** with **glass spacer**
- ▶ patch size: $L \times L \times 50 \text{ nm}$
- ▶ spacer gap: 30 nm
- ▶ illumination: ED, MD, EQ
with basic orientations
- ▶ goal: field/rate enhancement

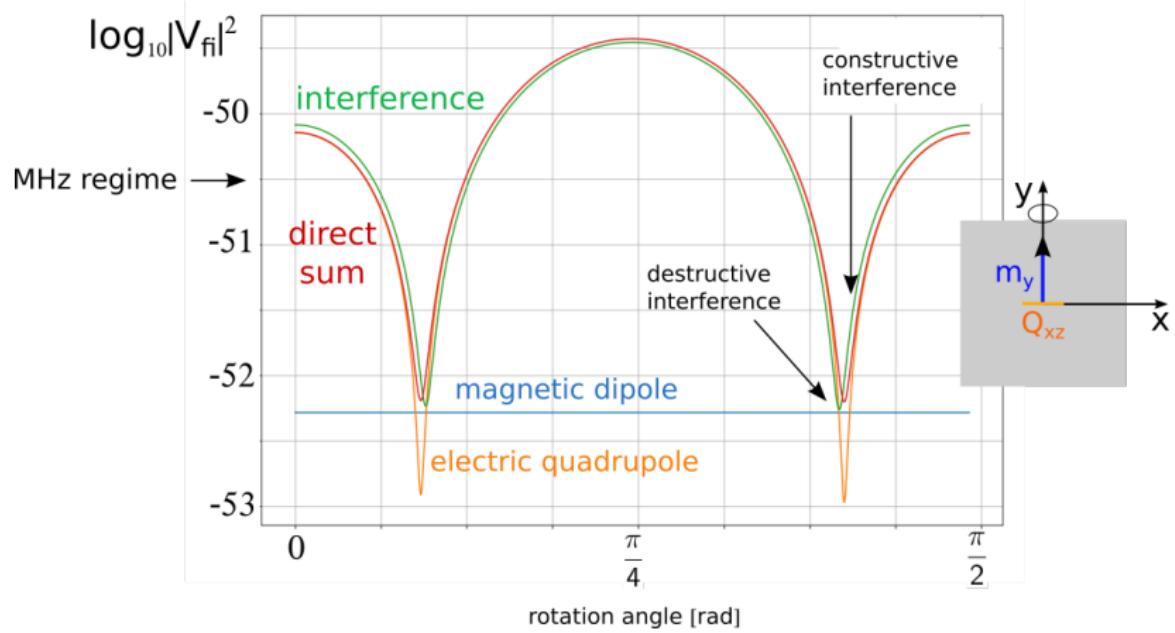
Transition rate enhancement: isolated MD or EQ sources



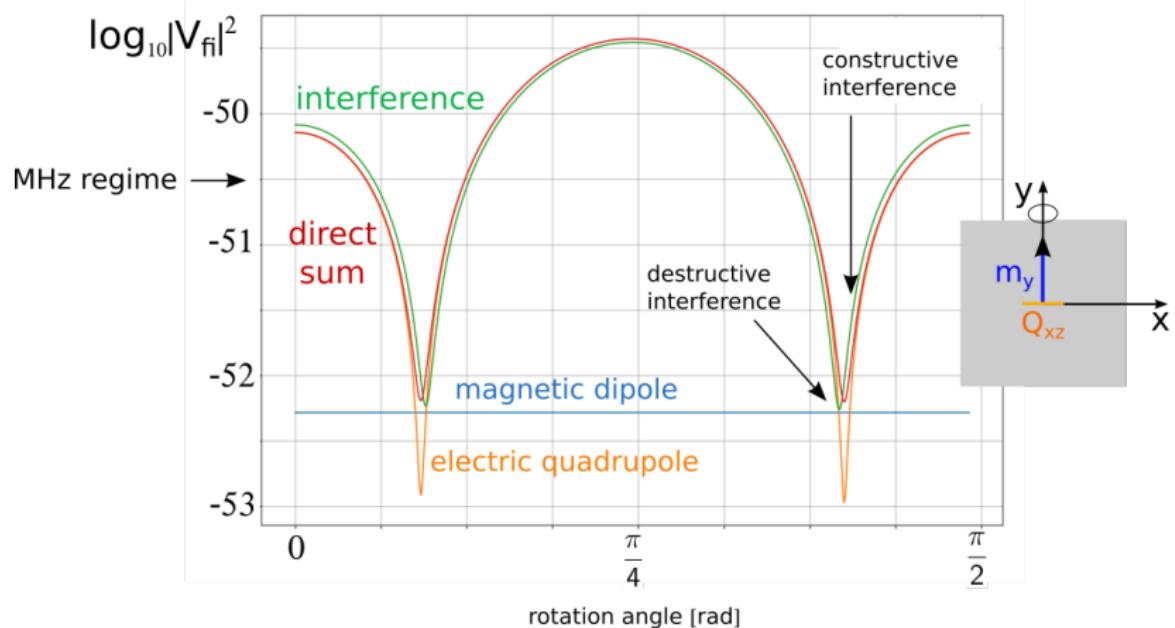
Transition rate: combined MD & EQ source



Transition rate: combined MD & EQ source

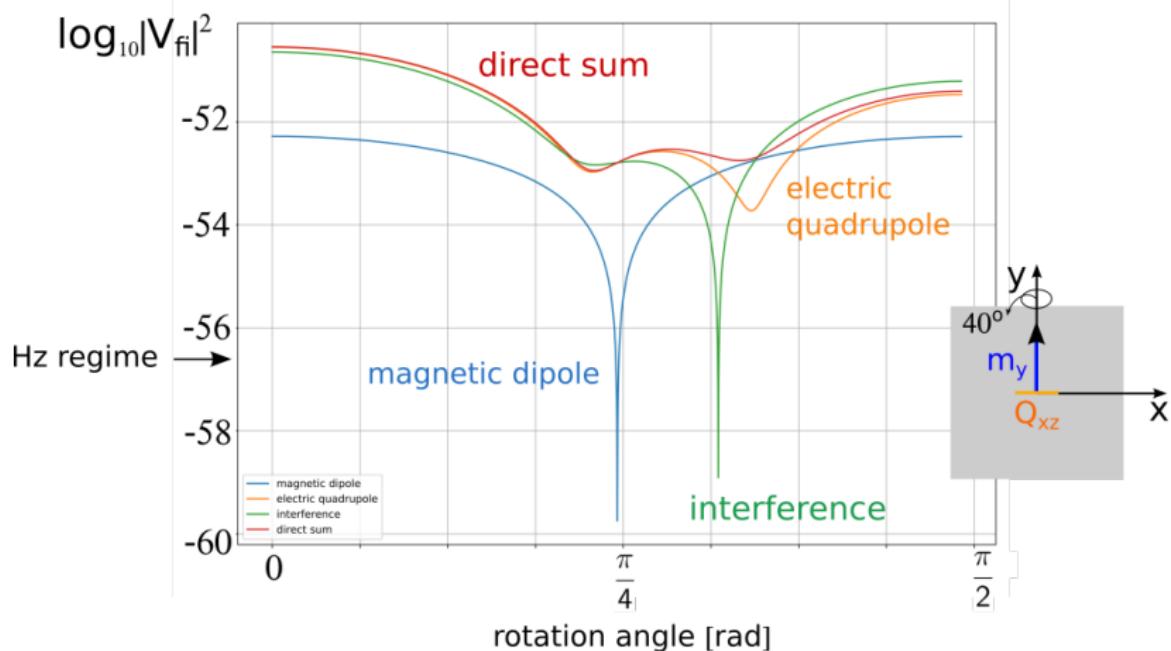


Transition rate: combined MD & EQ source



Response **stable with respect to molecular location**

Transition rate: combined MD & EQ source



7 orders of magnitude suppression due to interference
sensitive to small shifts

Conclusions & outlook

- ▶ Nanoantennas **enhance "forbidden" transitions** by even 4 orders of magnitude.
- ▶ **Interference** of parallel interaction channels:
suppression of spontaneous emission
by 7 orders of magnitude

Acknowledgements



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Smart Growth



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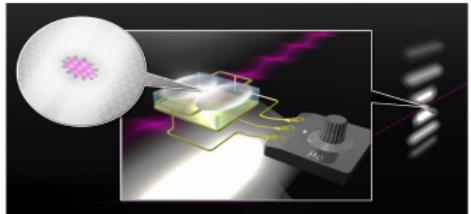
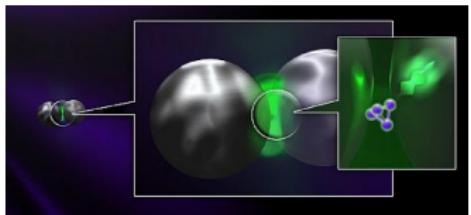
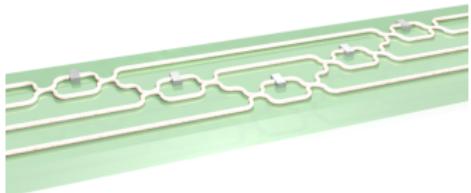
Ministry of Science
and Higher Education
Republic of Poland



PhD positions available:

- ▶ quantum optics: single photon sources, photonic circuits, coupling to quantum emitters
- ▶ nanophotonic devices: nanoantennas, metasurfaces, microdisks
- ▶ 2D plasmonic materials
- ▶ optically dressed media

Contact: karolina@fizyka.umk.pl





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