Single molecule fluorescence quenching by metallic nanoparticles: crossover between macroscopic and microscopic interactions

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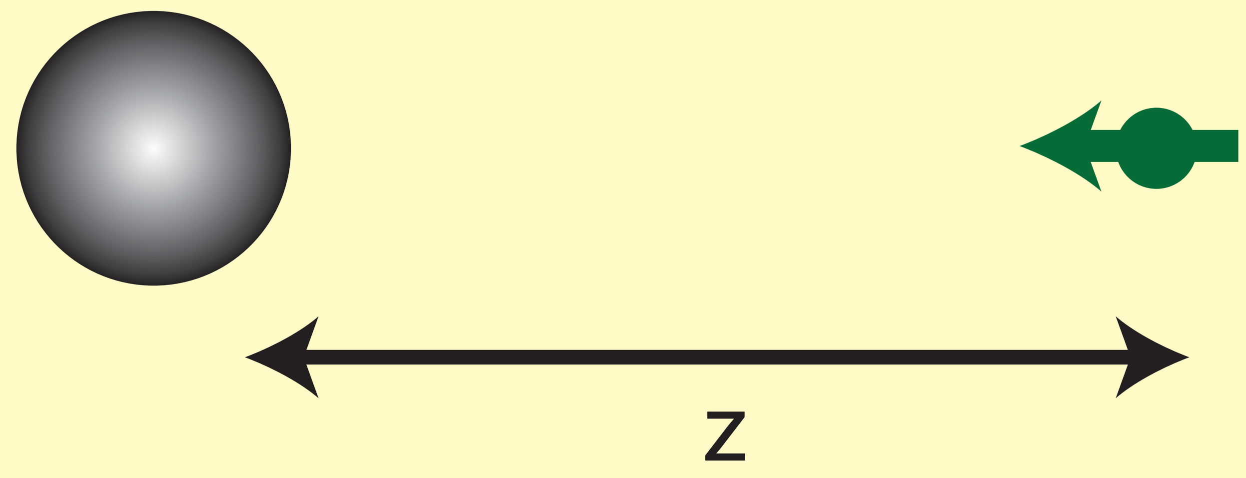
The fluorescence decay rate of a single emitter is modified close to a metallic nano-sphere.

Where does the macroscopic description of the emitter-sphere interaction fail ?

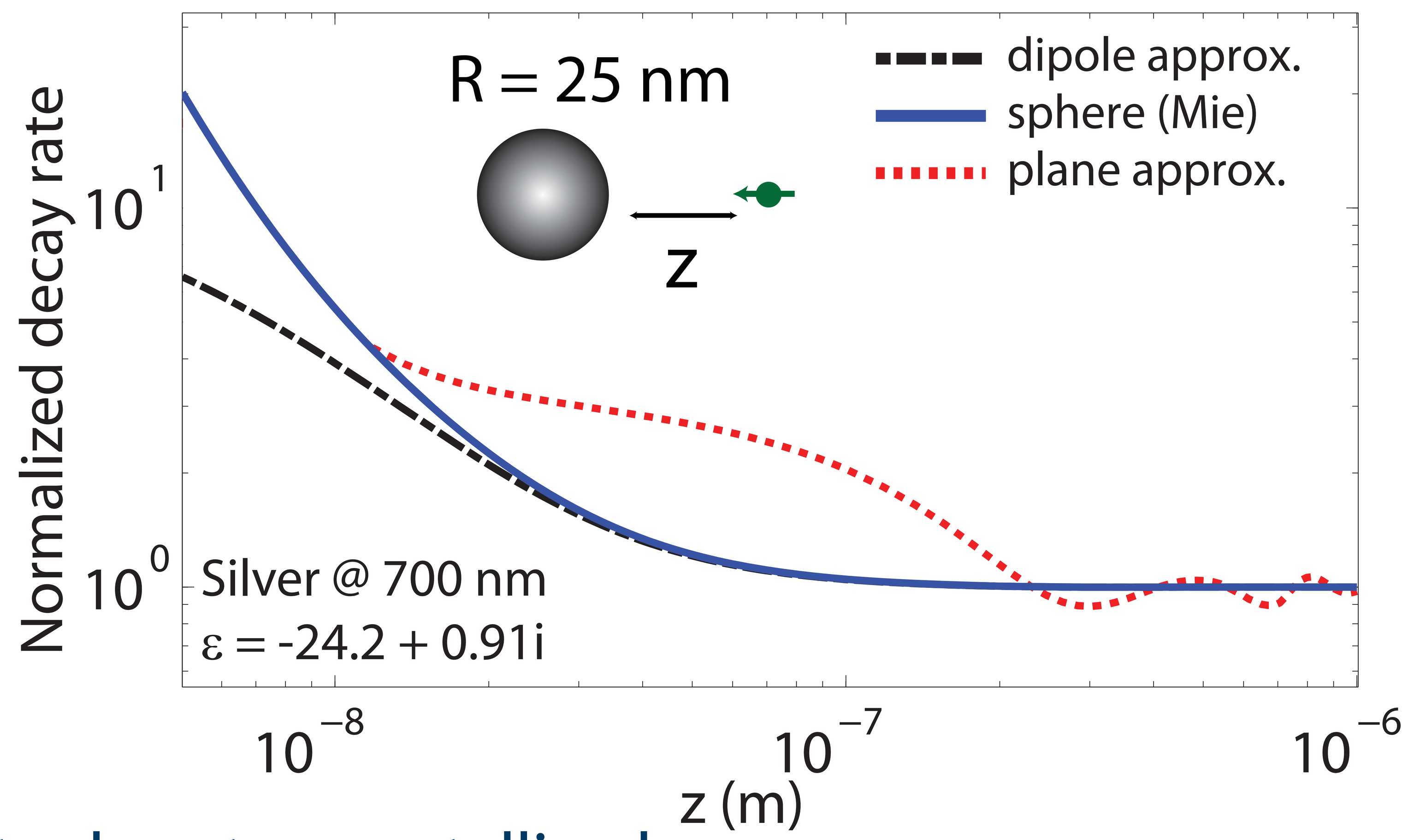


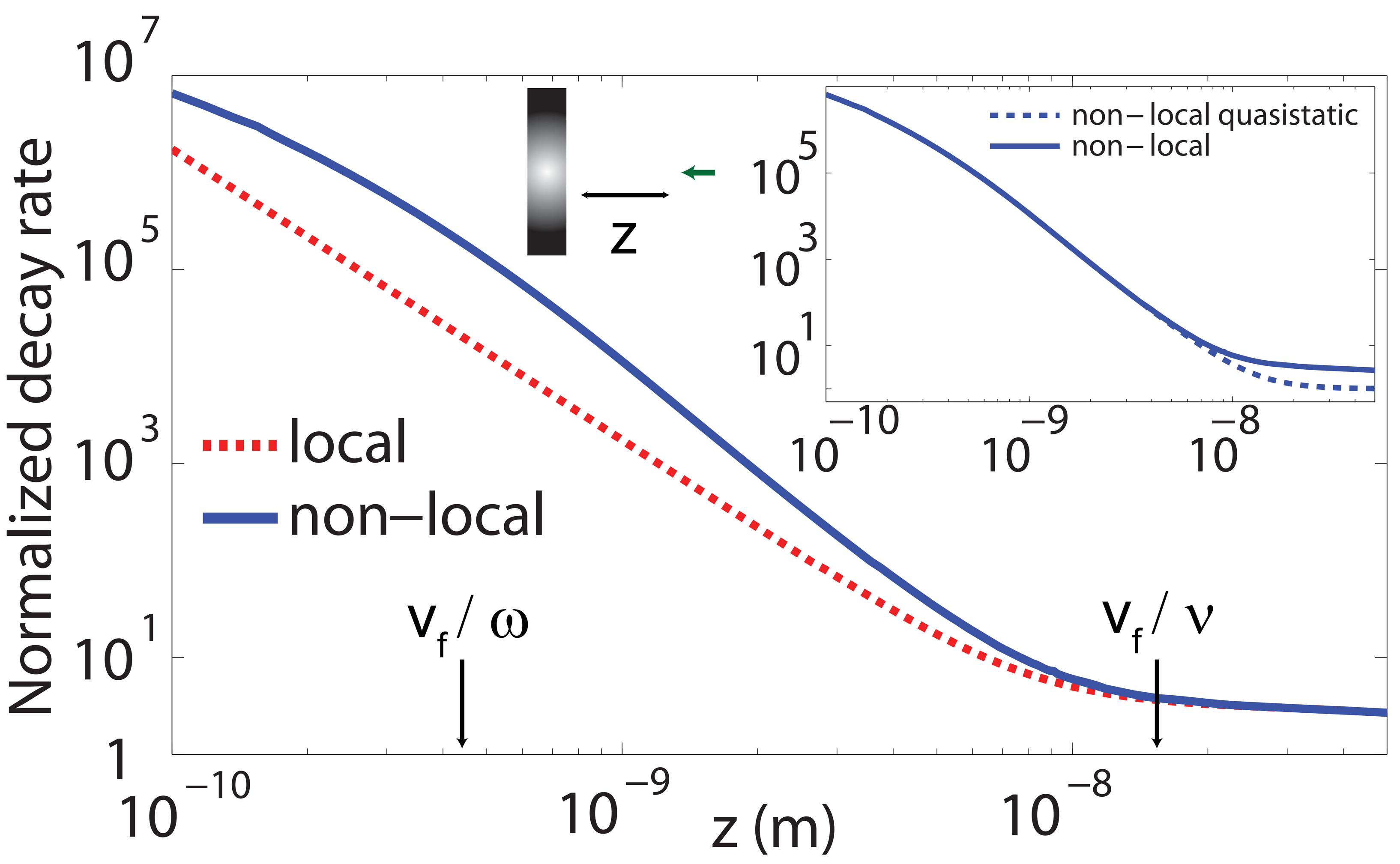
Full transition from far-field to near-field (macroscopic approach)

We study the decay rate (inverse of the fluorescence lifetime) modification of a single emitter in the vicinity of a metallic nano-sphere (gold or silver, radius R) in the regime where $2R > l_e$ (l_e being the electron mean free path)

- At a large distance z, the nano-sphere is seen like a dipole by the emitter[1]
- At short distance the sphere can be approximated by a plane
- Between z = R/2 and z = 2R, only the Mie theory for spheres gives the correct normalized decay rate

- Expected failure of the macroscopic model for z below a few nm[2]





Non-local effects close to a metallic plane

- When z becomes of the order of the microscopic length scales of the free electron gas of the nano-sphere, non-local effects have to be considered

- Use of the Lindhard-Mermin model for the dielectric constant along with a surface impedance[3] to calculate the non-local normalized decay rate of a single emitter close to a metallic plane

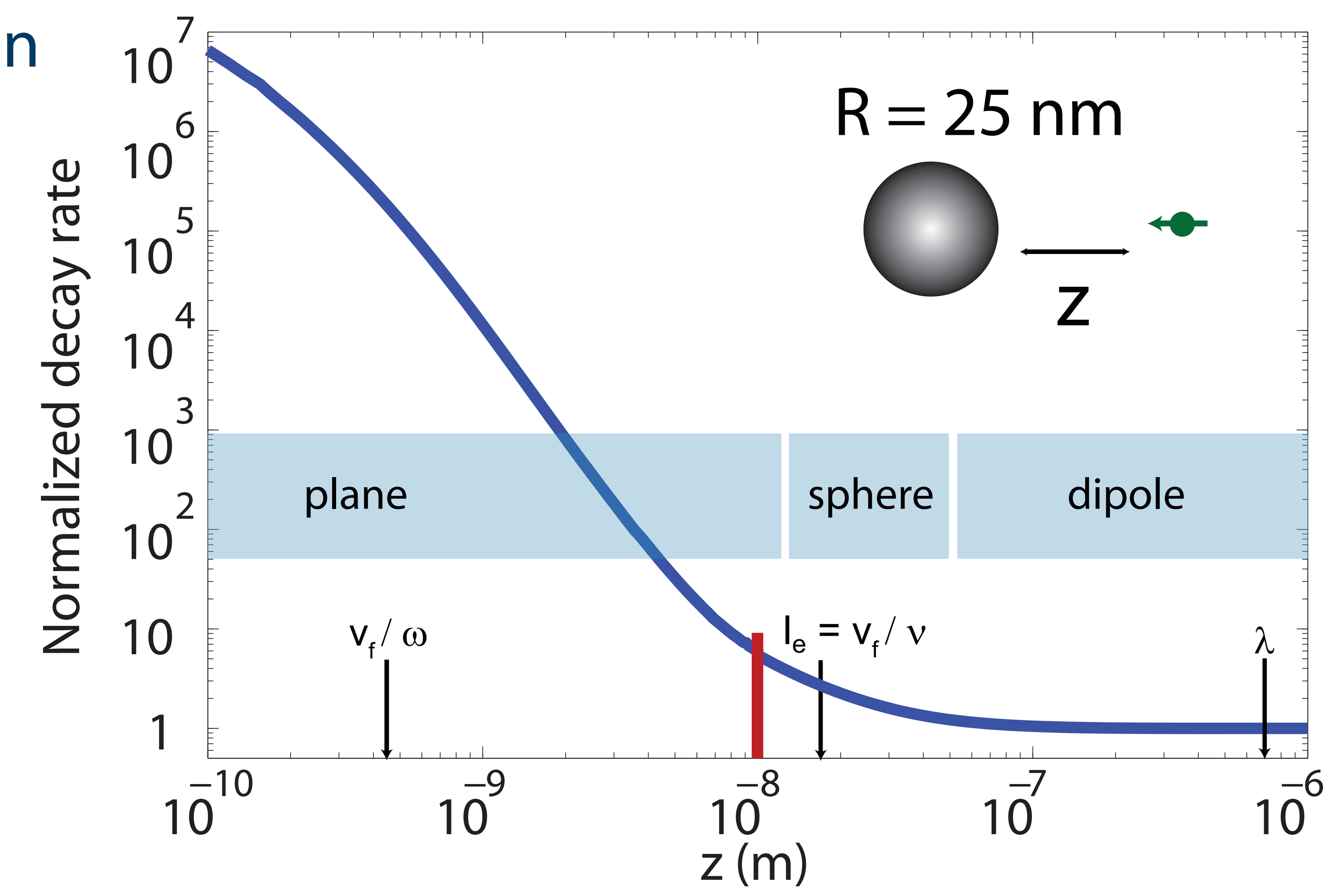
- We find a faster increase of the decay rate when z decreases[4]. The deviation from the local model occurs[5] when $z < l_e$.

Length	Expression	Silver
Fermi electron mean free path	$l_e = \frac{v_F}{\nu}$	16 nm
Distance travelled by an electron during one period of the electromagnetic field	$\frac{v_F}{\omega}$	0.45 nm
Fermi wavelength	$\frac{1}{k_F}$	0.083 nm

From macroscopic dipolar interaction to microscopic plane interaction

- Normalized decay rate of a single emitter close to a silver nano-sphere: the right part of the curve (z > 10 nm) is calculated with the exact Mie theory, the left part (z < 10 nm) with the non-local plane model

- This plot gives the full normalized decay rate from far-field (1 µm) to extreme near-field (1 Å)



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2 P. Anger, P. Bharadwaj and L. Novotny, Phys. Rev. Lett. 96, 113002 (2006)
3 G.W. Ford and W.H. Weber, Phys. Rep. 113,195 (1984)
4 I.A. Larkin, M.I. Stockman, M. Achermann and V.I. Klimov, Phys. Rev. B 69, 121403(R) (2004)
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- Calculation of the normalized decay rate full transition from far-field to extreme near field
- Breakdown of the macroscopic description below the Fermi electron mean free path