



# Nano-Optics

*Bert Hecht*

*Nano-Optics and Bio-Photonics Group  
Lehrstuhl Experimentelle Physik V*

*Universität Würzburg  
Am Hubland  
97074 Würzburg*




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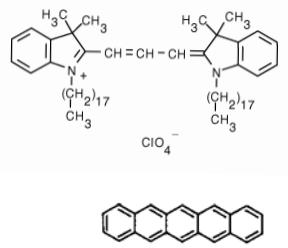
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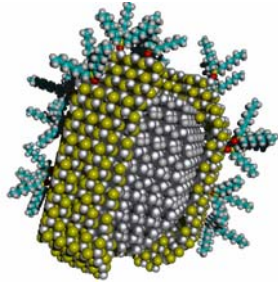


## Single emitters

Organic dye molecules




Semiconductor nanocrystal quantum dots



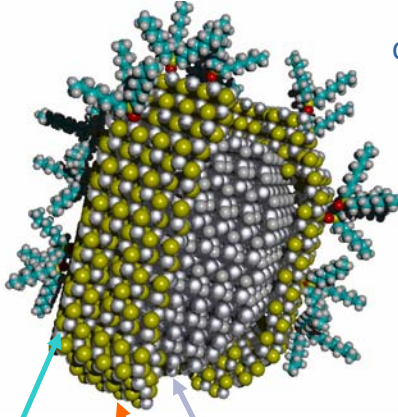
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## Colloidal Semiconductor Nanocrystals



complex architecture

Core: opto-electronically active part

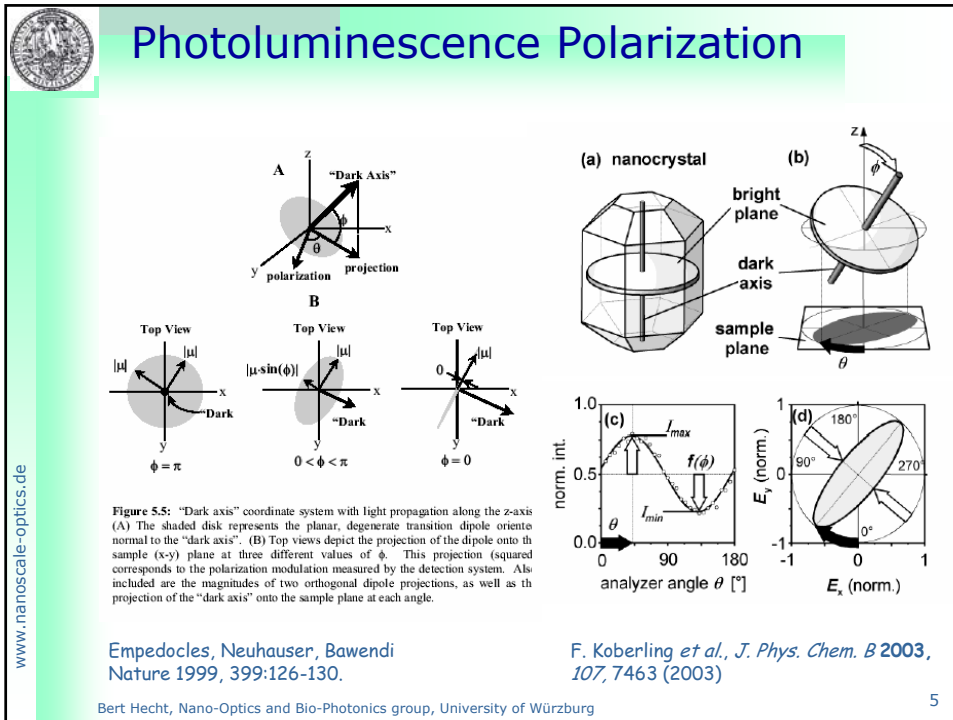
Shell: protective, complementary part

Organics: protective, functional part → biological applications

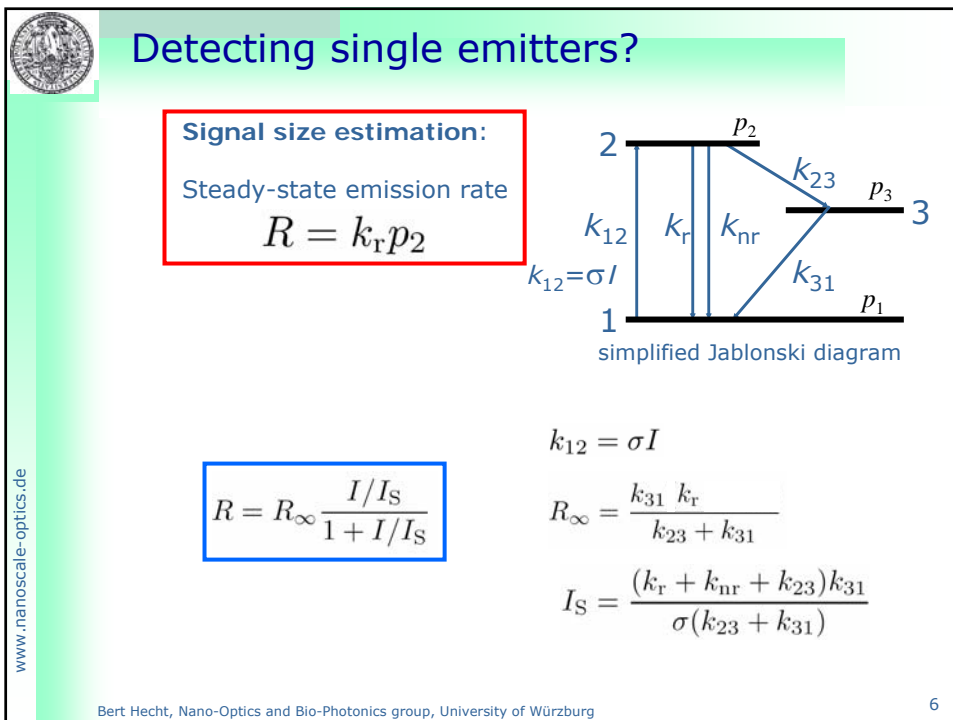
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Slides courtesy H.-J. Eisler 4

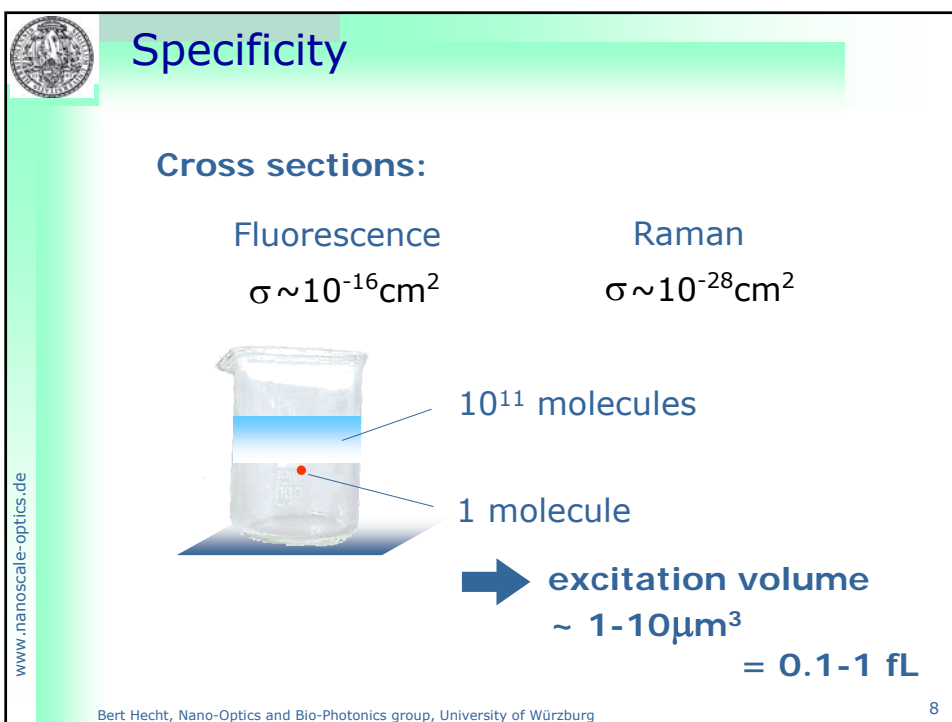
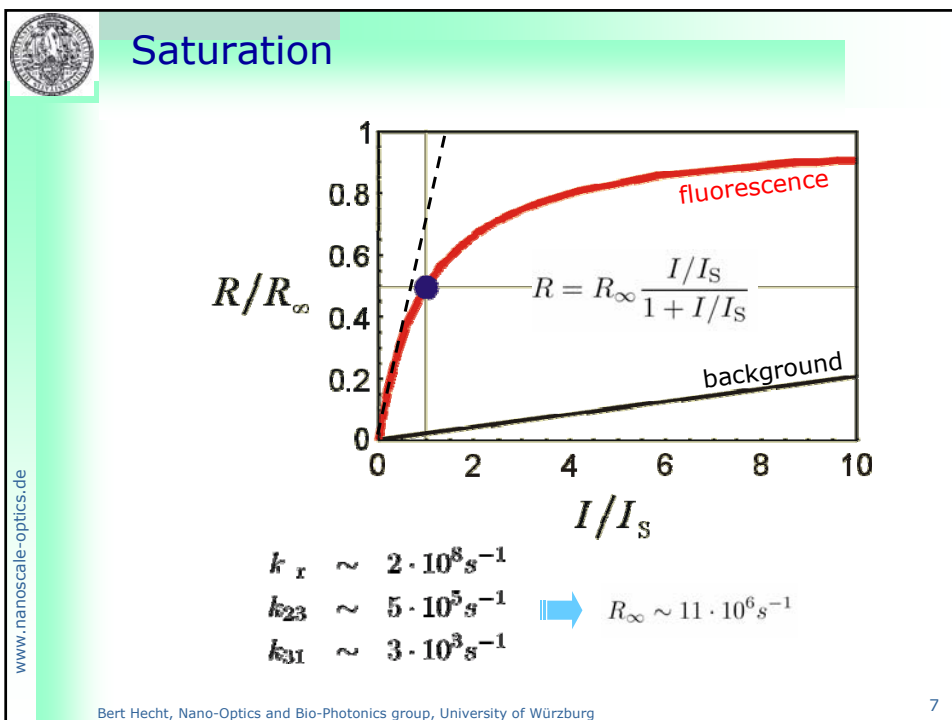
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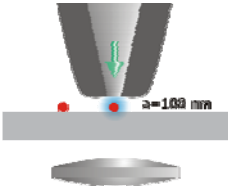


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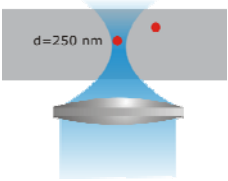


 **Microscopy techniques**

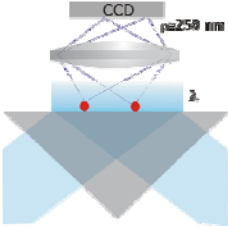
- control of background signals
- isolate single molecules by spatial (and spectral) selection & dilution



$V = \pi a^2 / 4 \cdot a$   
 $= 8 \cdot 10^4 \mu\text{m}^3$




$V = \pi d^2 / 4 \cdot 2.5d$   
 $= 3 \cdot 10^2 \mu\text{m}^3$



$V = \pi \rho^2 / 4 \cdot \lambda$   
 $= 2.5 \cdot 10^2 \mu\text{m}^3$

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 **Quantum properties of single emitters**

Single emitters emit single photons at a time

➔ **Photon Statistics**

analysis of the time structure (fluctuations) in the stream of emitted photons

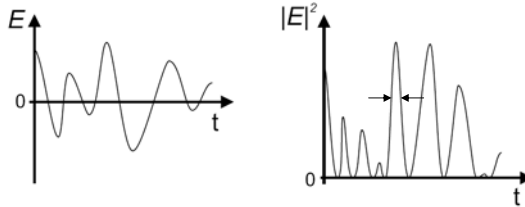
➔ a way to identify possible "nonclassical" or quantum behavior of light

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## Tool: Second order autocorr. function



$$g^{(2)}(\tau) = \frac{\langle I(t)I(t + \tau) \rangle}{\langle I(t) \rangle^2}$$

$$\langle \dots \rangle = \frac{1}{T} \int \dots dt \quad \text{for large but finite } T$$

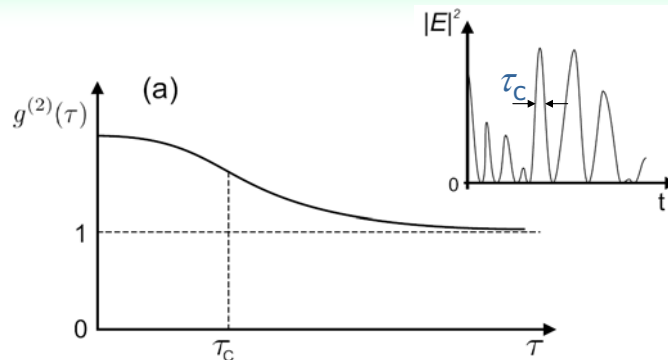
Literature: R. Loudon, The quantum theory of light, Oxford University Press

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## Properties of $g^{(2)}(\tau)$



If the intensity is a classical continuous function of time the following relations must be fulfilled:

$$g^{(2)}(0) \geq 1$$

$$g^{(2)}(\tau) \leq g^{(2)}(0)$$

Literature: R. Loudon, The quantum theory of light, Oxford University Press

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## Single quantum system

consider

$$g^{(2)}(\tau) = \frac{\langle I(t)I(t+\tau) \rangle}{\langle I(t) \rangle^2} \quad \text{for } t=0$$

For  $t=0$  we prepare the emitter in the ground state:  $p_2(0) = 0$

The probability for any photon to be **detected** at time  $\tau$  provided a photon was emitted at  $\tau=0$  is then given by  $\eta k_r p_2(\tau)$

$$g^{(2)}(\tau) = \frac{\eta k_r p_2(\tau)}{\eta k_r p_2(\infty)} = \frac{p_2(\tau)}{p_2(\infty)}$$

$\eta k_r p_2(\infty)$  is the steady-state count-rate that is measured after a sufficiently long time



we need the time-dependent solution of

$$\dot{p}_1 = -k_{12}p_1 + (k_r + k_{nr})p_2 + k_{31}p_3$$

$$\dot{p}_2 = k_{12}p_1 - (k_r + k_{nr} + k_{23})p_2$$

$$\dot{p}_3 = k_{23}p_2 - k_{31}p_3$$

$$\dot{p}_1 = -(k_{12} + k_{31})p_1 + (k_r + k_{nr} - k_{31})p_2 + k_{31}$$

$$\dot{p}_2 = k_{12}p_1 - (k_r + k_{nr} + k_{23})p_2 .$$



can be written in matrix form

$$\dot{\mathbf{p}}(\tau) = \begin{bmatrix} a & b \\ c & d \end{bmatrix} \mathbf{p}(\tau) + \begin{bmatrix} f \\ 0 \end{bmatrix}$$

Laplace transformation:

$$s\mathbf{p}(s) - \mathbf{p}(0) = \begin{bmatrix} a & b \\ c & d \end{bmatrix} \mathbf{p}(s) + \frac{1}{s} \begin{bmatrix} f \\ 0 \end{bmatrix}$$



## Laplace transformation

$$F(s) = \mathcal{L}\{f(t)\} = \int_0^{\infty} e^{-st} f(t) dt. \quad \text{with } s = \sigma + i\omega; \quad \sigma > 0; \quad t \geq 0;$$

$F(s)$  is called the Laplace-transform of  $f(t)$

Inverse transformation:

$$f(t) = \mathcal{L}^{-1}\{F(s)\} = \frac{1}{2\pi i} \int_{\gamma-i\infty}^{\gamma+i\infty} e^{st} F(s) ds \quad \gamma > \max_k \operatorname{Re}(z_k),$$

where the  $z_k$  are the singularities of  $F$

Calculation of the integrals via complex analysis (Residuensatz, usw.)

Most of the times transformation possible using tables of transformations



## Transformation table

$f$	$\mathcal{L}_t [f(t)](s)$				
	$\frac{1}{s}$	$s > 0$			
$t$	$\frac{1}{s^2}$	$s > 0$	$e^{at} \cos(bt)$	$\frac{s-a}{(s-a)^2+b^2}$	$s > \alpha$
$t^n$	$\frac{n!}{s^{n+1}}$	$n \in \mathbb{Z} > 0$			
$t^a$	$\frac{\Gamma(a+1)}{s^{a+1}}$	$a > 0$	$\delta(t-c)$	$e^{-cs}$	$c > 0$
$e^{at}$	$\frac{1}{s-a}$	$s > \alpha$	$H_c(t)$	$\frac{e^{-cs}}{s}$	$s > 0$
$\cos(\alpha t)$	$\frac{s-a}{s^2+a^2}$	$s > 0$	$J_0(t)$	$\frac{1}{\sqrt{s^2+1}}$	
$\sin(\alpha t)$	$\frac{a}{s^2+a^2}$	$s > 0$			
$\cosh(\alpha t)$	$\frac{s}{s^2-a^2}$	$s >  \alpha $	$J_n(\alpha t)$	$\frac{(\sqrt{s^2+a^2}-s)^n}{a^n \sqrt{s^2+a^2}}$	
$\sinh(\alpha t)$	$\frac{a}{s^2-a^2}$	$s >  \alpha $			$s > 0, n > -1$
$e^{at} \sin(bt)$	$\frac{b}{(s-a)^2+b^2}$	$s > \alpha$			

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## Properties of the Laplace transform

<http://de.wikipedia.org/wiki/Laplace-Transformation>

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## Example

Solve the differential equation,

$$5 \frac{dy}{dt} + 4y = 2, \quad y(0) = 1$$

using Laplace transform.

Solution) Take Laplace transform both sides

$$5\{sY(s) - y(0)\} + 4Y(s) = \frac{2}{s}$$

$$Y(s) = \frac{5s + 2}{s(5s + 4)}$$

$$y(t) = L^{-1}\left\{\frac{5s + 2}{s(5s + 4)}\right\} = 0.5\{1 + \exp(-0.8t)\}$$

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$$\mathbf{p}(s) = \left[ s \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} - \begin{pmatrix} a & b \\ c & d \end{pmatrix} \right]^{-1} \begin{pmatrix} f/s + 1 \\ 0 + 0 \end{pmatrix}$$

using  $p_1(0)=1$

back transformation using the Heaviside expansion theorem:

$$p_2(\tau) = A_1 e^{s_1 \tau} + A_2 e^{s_2 \tau} + A_3$$



$$s_1 = \frac{1}{2} \left( a + d - \sqrt{(a-d)^2 + 4bc} \right)$$


$$s_2 = \frac{1}{2} \left( a + d + \sqrt{(a-d)^2 + 4bc} \right)$$

$$A_1 = +c \frac{1 + f/s_1}{s_1 - s_2}, \quad A_2 = -c \frac{1 + f/s_2}{s_1 - s_2}, \quad A_3 = \frac{cf}{s_1 s_2}$$

using  $p_2(\infty) = A_3$

and  $-A_1/A_3 = (1 + A_2/A_3)$

$$\Rightarrow g^2(\tau) = -\left(1 + \frac{A_2}{A_3}\right) e^{s_1 \tau} + \frac{A_2}{A_3} e^{s_2 \tau} + 1$$



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$$k_{21} \geq k_{12} \gg k_{23} \geq k_{31}$$

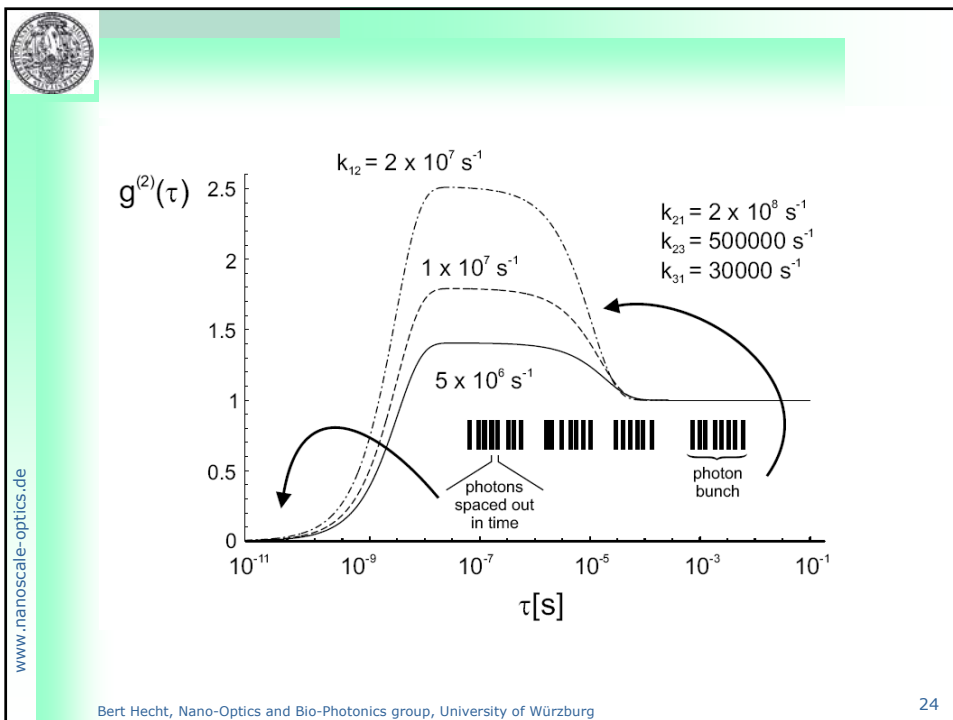
$$s_1 \simeq -(k_{12} + k_{21})$$

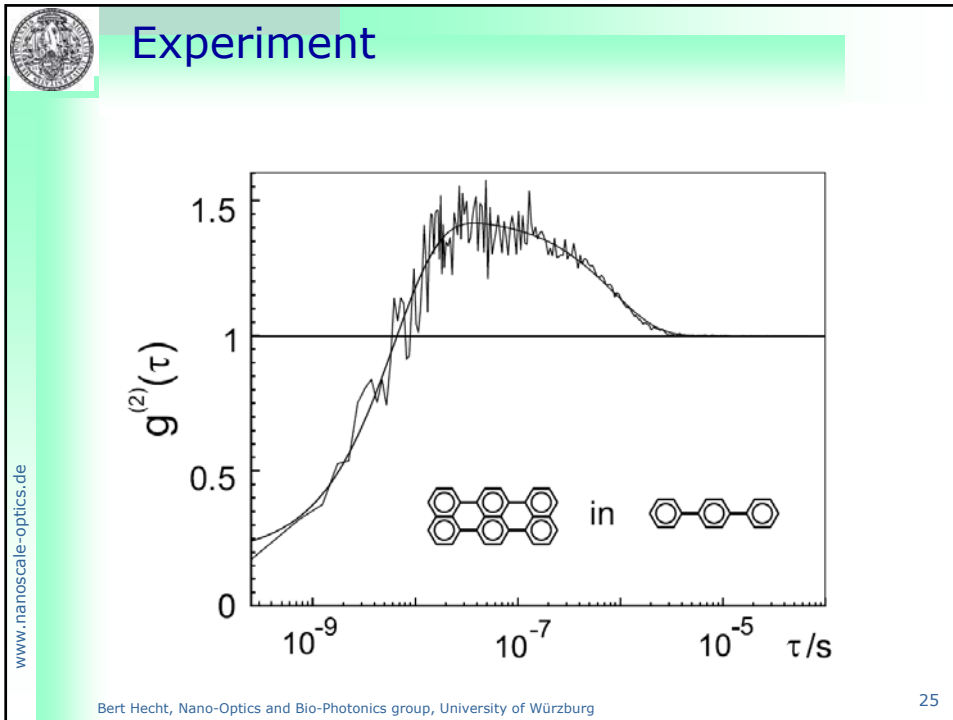
$$s_2 \simeq -\left(k_{31} + \frac{k_{12}k_{23}}{k_{12} + k_{21}}\right)$$

$$\frac{A_2}{A_3} \simeq \frac{k_{12}k_{23}}{k_{31}(k_{12} + k_{21})}$$

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**Electrodynamics of single emitters**

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- although single emitters emit a quantum field there interaction with matter can in most cases be described by classical electrodynamics!

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## Radiating electric dipole

$$\mathbf{j}(\mathbf{r}, t) = \frac{d}{dt} \boldsymbol{\mu}(t) \delta[\mathbf{r} - \mathbf{r}_o]$$

with

$$\boldsymbol{\mu}(t) = \sum_n q_n [\mathbf{r}_n(t) - \mathbf{r}_o]$$

harmonic time-dependence:

$$\mathbf{j}(\mathbf{r}, t) = \text{Re}\{\mathbf{j}(\mathbf{r}) \exp(-i\omega t)\}$$

$$\boldsymbol{\mu}(t) = \text{Re}\{\boldsymbol{\mu} \exp(-i\omega t)\}$$

$$\Rightarrow \mathbf{j}(\mathbf{r}) = -i\omega \boldsymbol{\mu} \delta[\mathbf{r} - \mathbf{r}_o]$$

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## Electric dipole in homogeneous space

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_o + i\omega \mu \mu_o \int_V \vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}') \mathbf{j}(\mathbf{r}') dV'$$

als formale Lösung der Helmholtzgleichung

$$\text{using } \mathbf{j}(\mathbf{r}) = -i\omega \boldsymbol{\mu} \delta[\mathbf{r} - \mathbf{r}_o]$$

$$\Rightarrow \mathbf{E}(\mathbf{r}) = \omega^2 \mu \mu_o \vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}_o) \boldsymbol{\mu}$$

$$\text{with } \vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}_o) = \left[ \vec{\mathbf{I}} + \frac{1}{k^2} \nabla \nabla \right] G(\mathbf{r}, \mathbf{r}_o)$$

$$\text{where } G(\mathbf{r}, \mathbf{r}_o) = \frac{\exp(ik|\mathbf{r} - \mathbf{r}_o|)}{4\pi|\mathbf{r} - \mathbf{r}_o|}$$

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In a Cartesian coordinate system:

$$\vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}_o) = \frac{\exp(ikR)}{4\pi R} \left[ \left( 1 + \frac{ikR - 1}{k^2 R^2} \right) \vec{\mathbf{I}} + \frac{3 - 3ikR - k^2 R^2}{k^2 R^2} \frac{\mathbf{R}\mathbf{R}}{R^2} \right]$$

$$\mathbf{R} = \mathbf{r} - \mathbf{r}_o$$

$\mathbf{R}\mathbf{R}$  denotes the outer product of  $\mathbf{R}$  with itself.

$$\vec{\mathbf{G}} = \underbrace{\vec{\mathbf{G}}_{NF}}_{(kR)^{-3}} + \underbrace{\vec{\mathbf{G}}_{IF}}_{(kR)^{-2}} + \underbrace{\vec{\mathbf{G}}_{FF}}_{(kR)^{-1}}$$



$$\vec{\mathbf{G}}_{NF} = \frac{\exp(ikR)}{4\pi R} \frac{1}{k^2 R^2} \left[ -\vec{\mathbf{I}} + 3\mathbf{R}\mathbf{R}/R^2 \right]$$

$$\vec{\mathbf{G}}_{IF} = \frac{\exp(ikR)}{4\pi R} \frac{i}{kR} \left[ \vec{\mathbf{I}} - 3\mathbf{R}\mathbf{R}/R^2 \right]$$

$$\vec{\mathbf{G}}_{FF} = \frac{\exp(ikR)}{4\pi R} \left[ \vec{\mathbf{I}} - \mathbf{R}\mathbf{R}/R^2 \right]$$



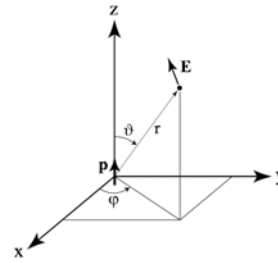
in homogeneous space it is sufficient to consider  $\boldsymbol{\mu} = |\boldsymbol{\mu}| \mathbf{n}_z$   
and use spherical coordinates:  $\mathbf{r} = (r, \vartheta, \varphi)$

$$\mathbf{E} = (E_r, E_\vartheta, E_\varphi)$$

$$E_r = \frac{|\boldsymbol{\mu}| \cos \vartheta}{4\pi\epsilon_o\epsilon} \frac{\exp(ikr)}{r} k^2 \left[ \frac{2}{k^2 r^2} - \frac{2i}{kr} \right],$$

$$E_\vartheta = \frac{|\boldsymbol{\mu}| \sin \vartheta}{4\pi\epsilon_o\epsilon} \frac{\exp(ikr)}{r} k^2 \left[ \frac{1}{k^2 r^2} - \frac{i}{kr} - 1 \right]$$

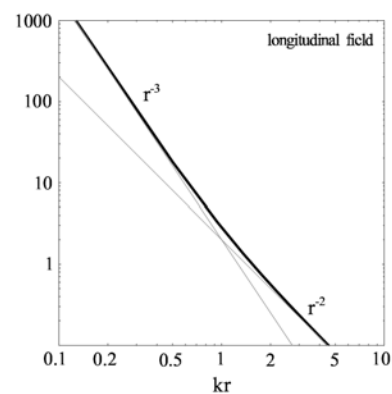
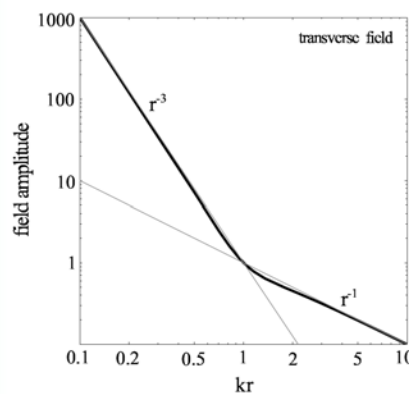
$$H_\varphi = \frac{|\boldsymbol{\mu}| \sin \vartheta}{4\pi\epsilon_o\epsilon} \frac{\exp(ikr)}{r} k^2 \left[ -\frac{i}{kr} - 1 \right] \sqrt{\frac{\epsilon_o\epsilon}{\mu_o\mu}}$$



$E_r$  has no farfield term  $\rightarrow$  field in the farfield is purely transverse  
magnetic field has no terms in  $(kr)^{-3} \rightarrow$  the nearfield is dominated  
by the electric field

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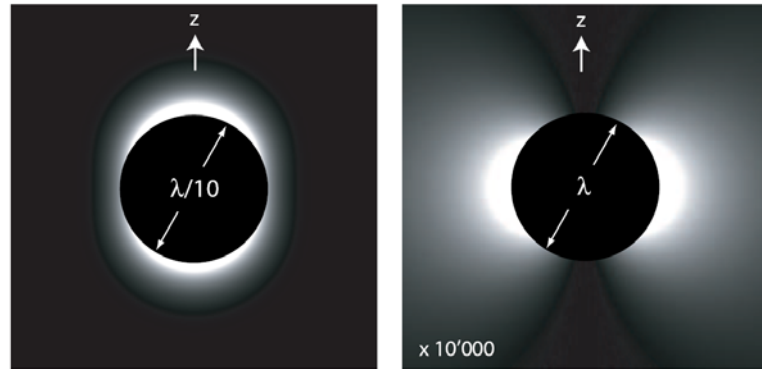
While both the transverse and the longitudinal field contribute  
to the near field, only the transverse field survives in the far field.

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## Near field vs. far field



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## Dipole radiation

Only the far field contributes to net energy transport (in free space)

$$\mathbf{S}(t) = \mathbf{E}(t) \times \mathbf{H}(t)$$

$$\boldsymbol{\mu}(t) = \text{Re}\{\boldsymbol{\mu} \exp(-i\omega t)\}$$

$$P(t) = \int_{\partial V} \mathbf{S} \cdot \mathbf{n} da = \frac{1}{4\pi\epsilon_0\epsilon} \frac{2n^3}{3c^3} \left[ \frac{d^2 |\boldsymbol{\mu}(t)|}{dt^2} \right]^2$$

$$\bar{P} = \frac{|\boldsymbol{\mu}|^2}{4\pi\epsilon_0\epsilon} \frac{n^3\omega^4}{3c^3}$$

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## Poyntings theorem

$$\nabla \times \mathbf{E}(\mathbf{r}, t) = -\frac{\partial \mathbf{B}(\mathbf{r}, t)}{\partial t}, \quad (1)$$

$$\nabla \times \mathbf{H}(\mathbf{r}, t) = \frac{\partial \mathbf{D}(\mathbf{r}, t)}{\partial t} + \mathbf{j}(\mathbf{r}, t), \quad (2)$$

$$\mathbf{H} \cdot (1) - \mathbf{E} \cdot (2)$$

$$\begin{aligned} \mathbf{H} \cdot (\nabla \times \mathbf{E}) - \mathbf{E} \cdot (\nabla \times \mathbf{H}) &= -\mathbf{H} \cdot \frac{\partial \mathbf{B}}{\partial t} - \mathbf{E} \cdot \frac{\partial \mathbf{D}}{\partial t} - \mathbf{j} \cdot \mathbf{E} \\ &= \nabla \cdot (\mathbf{E} \times \mathbf{H}) \end{aligned}$$

integrating both sides over space and applying Gauss' theorem

$$\begin{aligned} \int_{\partial V} (\mathbf{E} \times \mathbf{H}) \cdot \mathbf{n} \, da &= - \int_V \left[ \mathbf{H} \cdot \frac{\partial \mathbf{B}}{\partial t} + \mathbf{E} \cdot \frac{\partial \mathbf{D}}{\partial t} + \mathbf{j} \cdot \mathbf{E} \right] dV \\ \mathbf{D}(\mathbf{r}, t) &= \epsilon_0 \mathbf{E}(\mathbf{r}, t) + \mathbf{P}(\mathbf{r}, t), \\ \mathbf{H}(\mathbf{r}, t) &= \mu_0^{-1} \mathbf{B}(\mathbf{r}, t) - \mathbf{M}(\mathbf{r}, t) \end{aligned}$$

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## Energy conservation

Flow of e.m. energy in or out of V time rate of change of electromagnetic energy inside V

$$\begin{aligned} \int_{\partial V} (\mathbf{E} \times \mathbf{H}) \cdot \mathbf{n} \, da + \frac{1}{2} \frac{\partial}{\partial t} \int_V [\mathbf{D} \cdot \mathbf{E} + \mathbf{B} \cdot \mathbf{H}] dV &= \\ - \int_V \mathbf{j} \cdot \mathbf{E} dV &\quad \text{for linear media} \end{aligned}$$

Energy dissipation in V  
= Ohmic losses

$$\mathbf{S} = (\mathbf{E} \times \mathbf{H}) \quad \begin{array}{l} \text{energy flux density,} \\ \text{Poynting vector} \end{array} \quad \langle \mathbf{S} \rangle = \frac{1}{2} \text{Re} \{ \mathbf{E} \times \mathbf{H}^* \} \quad \begin{array}{l} \text{time- averaged} \\ \text{Poynting vector} \end{array}$$

$$W = \frac{1}{2} [\mathbf{D} \cdot \mathbf{E} + \mathbf{B} \cdot \mathbf{H}] \quad \text{density of electromagnetic energy}$$

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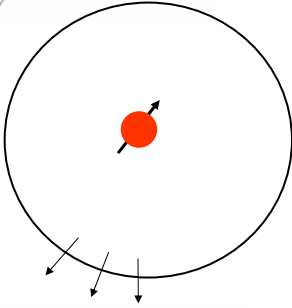
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the radiated power of any current distribution with a harmonic time dependence in a linear medium has to be identical to the rate of energy dissipation:

$$\frac{dW}{dt} = -\frac{1}{2} \int_V \text{Re}\{\mathbf{j}^* \cdot \mathbf{E}\} dV.$$

flow of energy out of V



$$\mathbf{S} = (\mathbf{E} \times \mathbf{H})$$

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using  $\mathbf{j}(\mathbf{r}) = -i\omega\boldsymbol{\mu} \delta[\mathbf{r} - \mathbf{r}_o]$

$$\frac{dW}{dt} = \frac{\omega}{2} \text{Im}\{\boldsymbol{\mu}^* \cdot \mathbf{E}(\mathbf{r}_o)\}$$

evaluated at the dipole's origin  $\mathbf{r}_o$

in terms of the Green's function  $\mathbf{E}(\mathbf{r}) = \omega^2\mu_o \vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}_o) \boldsymbol{\mu}$

$$\frac{dW}{dt} = \frac{\omega^3 |\boldsymbol{\mu}|^2}{2c^2 \epsilon_o \epsilon} \left[ \mathbf{n}_\mu \cdot \text{Im}\left\{ \vec{\mathbf{G}}(\mathbf{r}_o, \mathbf{r}_o; \omega) \right\} \cdot \mathbf{n}_\mu \right]$$

unit vector in direction of the dipole moment

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At first sight it seems not possible to evaluate

$$\frac{dW}{dt} = \frac{\omega^3 |\boldsymbol{\mu}|^2}{2c^2 \epsilon_0 \epsilon} \left[ \mathbf{n}_\mu \cdot \text{Im} \left\{ \bar{\mathbf{G}}(\mathbf{r}_o, \mathbf{r}_o; \omega) \right\} \cdot \mathbf{n}_\mu \right]$$

since  $\exp(ikR)/R$  appears to be infinite at  $\mathbf{r} = \mathbf{r}_o$ .

This is however not the case:

Due to the dot product between  $\boldsymbol{\mu}$  and  $\mathbf{E}$  we need only to evaluate the component of  $\mathbf{E}$  in direction of  $\boldsymbol{\mu}$

Choose  $\boldsymbol{\mu} = |\boldsymbol{\mu}| \mathbf{n}_z$

$$E_z = \frac{|\boldsymbol{\mu}|}{4\pi \epsilon_0 \epsilon} \frac{e^{ikR}}{R} \left[ k^2 \sin^2 \vartheta + \frac{1}{R^2} (3 \cos^2 \vartheta - 1) - \frac{ik}{R} (3 \cos^2 \vartheta - 1) \right]$$



Consider the limiting case  $R \rightarrow 0$


$$\exp(ikR) = 1 + ikR + (1/2)(ikR)^2 + (1/6)(ikR)^3 + ..$$

$$\frac{dW}{dt} = \lim_{R \rightarrow 0} \frac{\omega}{2} |\boldsymbol{\mu}| \text{Im}\{E_z\} =$$

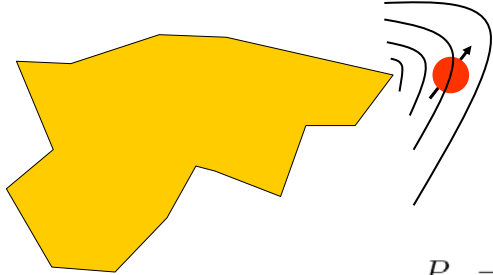
$$\frac{\omega |\boldsymbol{\mu}|^2}{8\pi \epsilon_0 \epsilon} \lim_{R \rightarrow 0} \left\{ \frac{2}{3} k^3 + R^2 (..) + .. \right\} = \frac{|\boldsymbol{\mu}|^2 \omega}{12\pi \epsilon_0 \epsilon} k^3$$

correct result despite of the apparent singularity at  $R=0$ !

compare to  $\bar{P} = \frac{|\boldsymbol{\mu}|^2}{4\pi \epsilon_0 \epsilon} \frac{n^3 \omega^4}{3c^3}$

 **Dipole in inhomogeneous space**

$\mathbf{E}(\mathbf{r}_o) = \mathbf{E}_o(\mathbf{r}_o) + \mathbf{E}_s(\mathbf{r}_o)$



$P_o = \frac{|\boldsymbol{\mu}|^2}{12\pi} \frac{\omega}{\epsilon_o \epsilon} k^3$


➔  $P = P_o + P_s$

➔  $P/P_o = 1 + P_s/P_o$

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 **Rate of energy dissipation**  
in inhomogeneous space

$$\frac{P}{P_o} = 1 + \frac{6\pi\epsilon_o\epsilon}{|\boldsymbol{\mu}|^2} \frac{1}{k^3} \text{Im}\{\boldsymbol{\mu}^* \cdot \mathbf{E}_s(\mathbf{r}_o)\}$$

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## Another point of view

undriven harmonically oscillating dipole:

$$\frac{d^2}{dt^2} \boldsymbol{\mu}(t) + \gamma_o \frac{d}{dt} \boldsymbol{\mu}(t) + \omega_o^2 \boldsymbol{\mu}(t) = 0$$

$$\longrightarrow \boldsymbol{\mu}(t) = \text{Re} \left\{ \boldsymbol{\mu}_o e^{-i\omega_o \sqrt{1 - (\gamma_o^2/4\omega_o^2)} t} e^{-\gamma_o t/2} \right\}$$

damped oscillation with slight frequency shift

require that  $\gamma_o \ll \omega_o$

then the average energy stored in the oscillator is:

$$\bar{W}(t) = \frac{m}{2q^2} [\omega_o^2 \mu^2(t) + \dot{\mu}^2(t)] = \frac{m\omega_o^2}{2q^2} |\boldsymbol{\mu}_o|^2 e^{-\gamma_o t}$$

$$\longrightarrow \text{lifetime of the oscillator: } \tau_o = 1/\gamma_o$$



## Classical decay rate

Energy conservation requires that the decrease in oscillator energy must equal the energy losses

$$\bar{W}(t=0) - \bar{W}(t) = q_i \overset{\text{quantum yield}}{\int_0^t} P_o(t') dt'$$

using  $\bar{W}(t)$  as just defined and  $P_o(t) = \frac{|\boldsymbol{\mu}(t)|^2}{4\pi\epsilon_o} \frac{\omega_o^4}{3c^3}$

we get

$$\gamma_o = q_i \frac{1}{4\pi\epsilon_o} \frac{2q^2\omega_o^2}{3mc^3}$$

**classical expression for the atomic decay rate**

At optical wavelengths a value for the decay rate of  $\sim 2 \cdot 10^{-8} \text{ s}^{-1}$  is obtained.