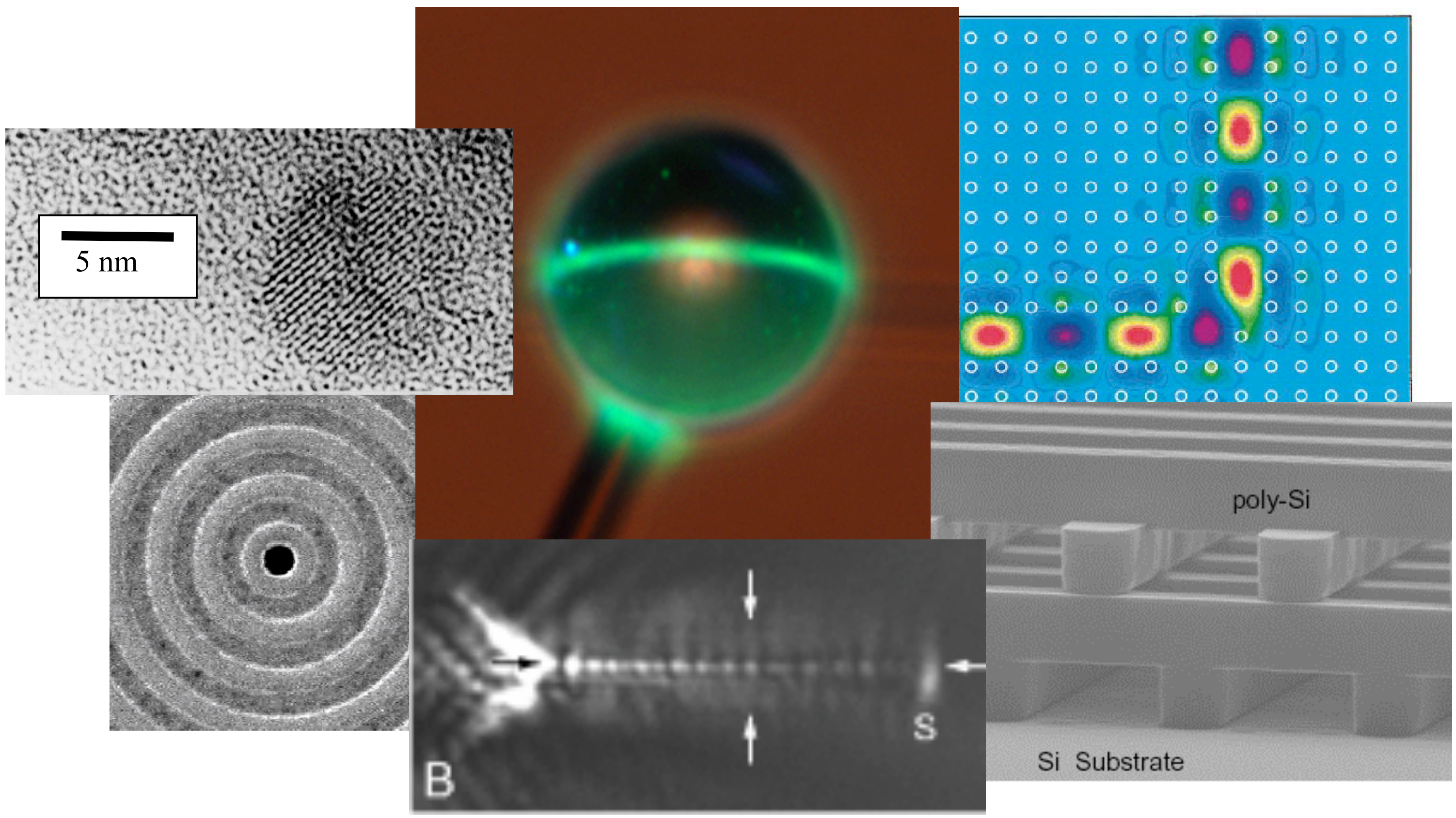
Lecture 4: Electromagnetic Properties of Molecules, Nano, and Microscopic Particles



Light Interaction with Small Structures

Molecules

- Light scattering due to harmonically driven dipole oscillator

Nanoparticles

- Insulators…Rayleigh Scattering (blue sky)
- Semiconductors….Resonance absorption at $\hbar\omega \geq E_{GAP}$, size dependent fluorescence…)
- Metals…Resonance absorption at surface plasmon frequency, no light emission)

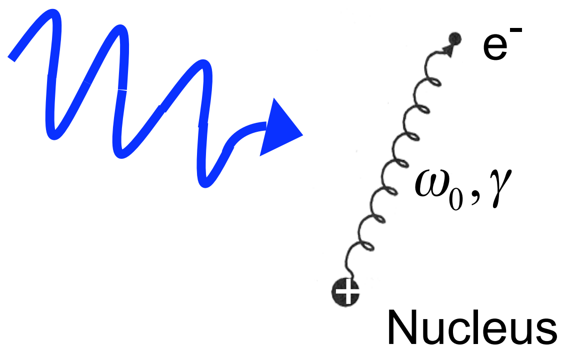
Microparticles

- Particles with dimensions on the order of λ or bigger
  - ⇨ Enhanced forward scattering
  - ⇨ Intuitive ray-picture useful
  - ⇨ Rainbows due to dispersion H_20
  - ⇨ Applications: resonators, lasers, etc…

Light Interaction with a Small Object

Electric field drives harmonic motion of electrons

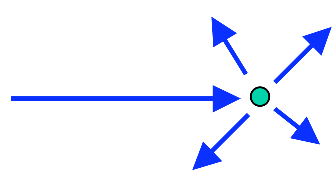
- Consider the Lorentz model



$$\mathbf{p} = \underbrace{\frac{e^2}{m}\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}}_{\sim \text{ Atomic polarizability}}\mathbf{E}_L$$

Oscillating charges radiates

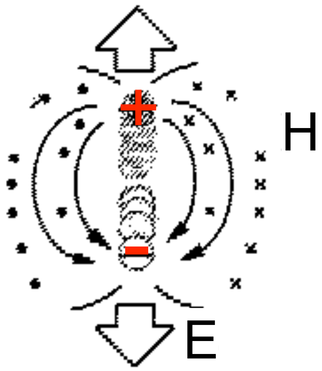
- This radiation is the scattered light intensity



- What does this process look like?

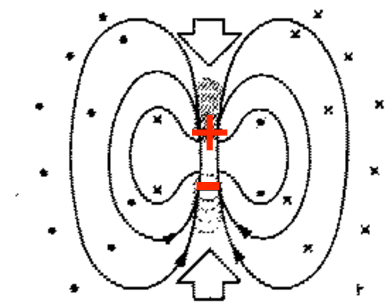
Oscillating charges Emits EM Waves

E and H fields from oscillating charges

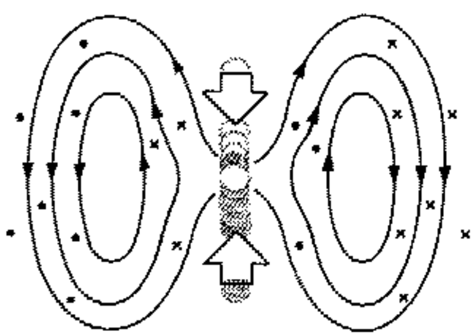


E-field lines start at positive charge

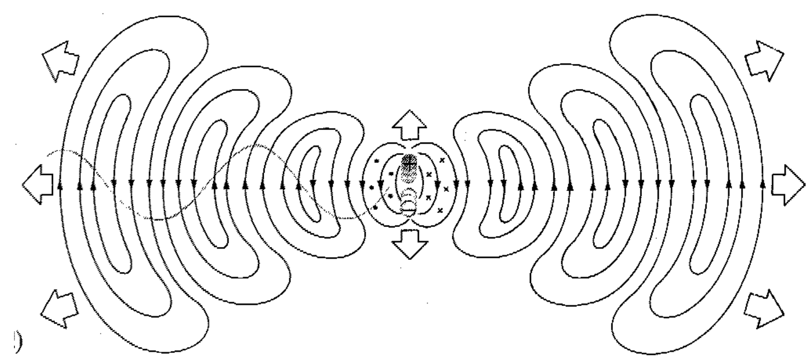
E-field lines end at negative charge



E-field lines close upon themselves
(field lines cannot cross)



The start of an EM wave



After several periods

Radiation mainly ⊥ to oscillation direction

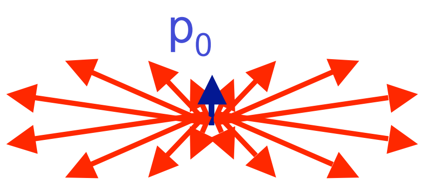
Oscillating charges Emit EM Waves

Radiation is angle dependent

- Radiated intensity: $I = \dfrac{p^2\omega^4}{32\pi^2\varepsilon_0 c^3 r^2}\sin^2\theta$

Derivation: Feynman lectures on physics (or Ramo, et al)

- Radiated pattern:



- Total scattered radiation: $P_S = \int_A I dA' = \dfrac{p_0^2\omega^4}{12\pi\varepsilon_0 c^3}$

Closed surface around the dipole

Radiation Emitted by a Lorentz Oscillator

Scattered intensity from a Lorentz Oscillator

- Scattered intensity by a dipole: $I = \dfrac{p^2\omega^4}{32\pi^2\varepsilon_0 c^3 r^2}\sin^2\theta$
- Lorentz model: $\mathbf{p} = \dfrac{e^2}{m}\dfrac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}\mathbf{E}_L$
- Lorentz model:

$$\boxed{I_S = \frac{e^4\omega^4}{32\pi^2 m^2\varepsilon_0 c^3 r^2}\left(\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}\right)^2 E_L^2 \sin^2\theta}$$

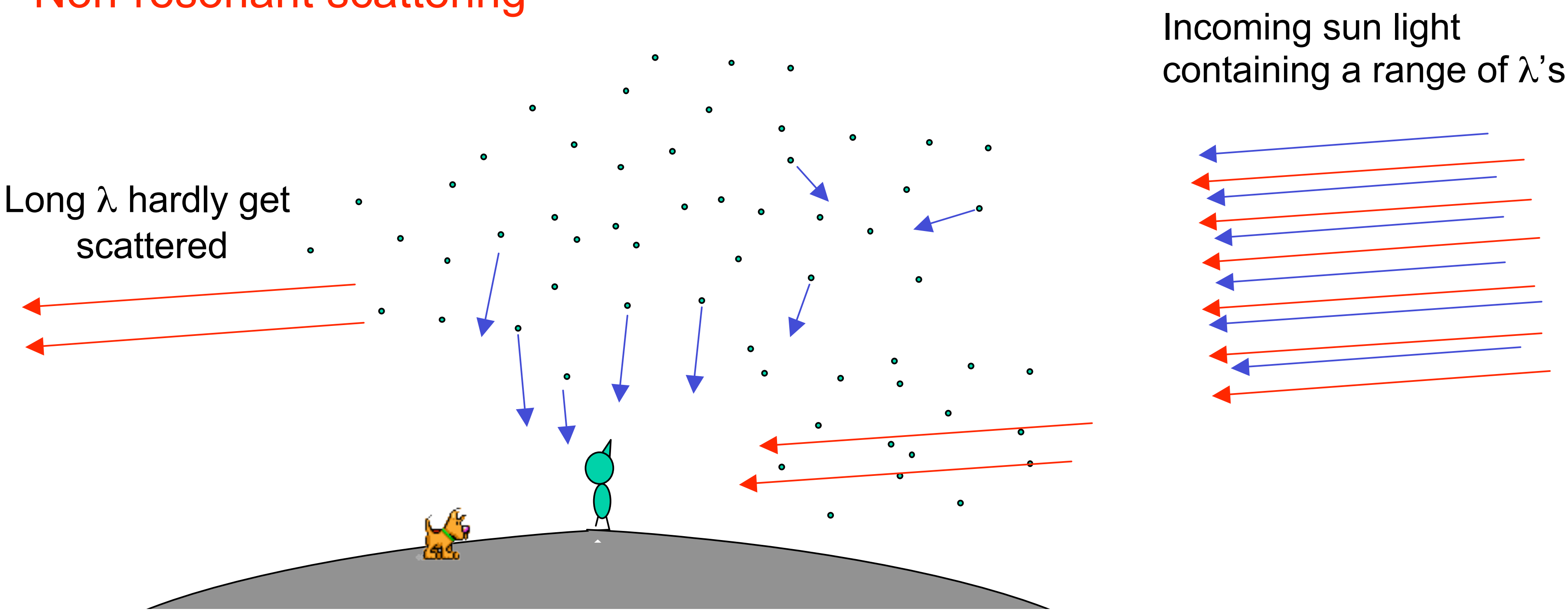
Incoming intensity

Conclusions

- Strongest scattering near a resonance
- Strongest scattering for higher ω or shorter λ
- Scattering occurs both in the forward and backward directions

The Blue Sky

Non-resonant scattering



- In the visible: O_2, and N_2 molecules have : $\omega_0 >> \omega$

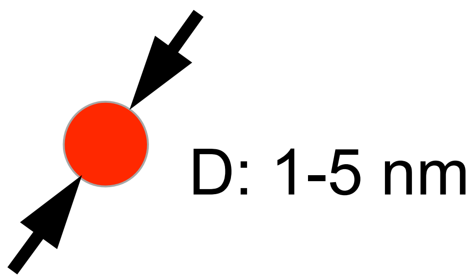
$$\Rightarrow \quad I_S = \frac{e^4\omega^4}{32\pi^2 m^2 \varepsilon_0 c^3 r^2} \frac{1}{\omega_0^2 \cancel{- \omega^2 - i\gamma\omega}} E^2 \sin^2\theta$$

- λ two times shorter $\Rightarrow$ Scattering $2^4 = 16$ times stronger

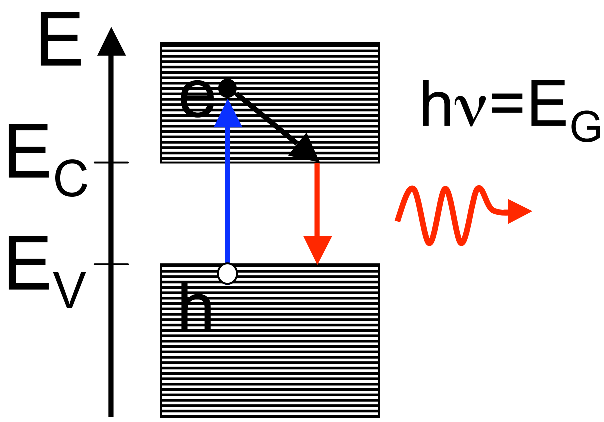
- Similar situation for insulating nanoparticles,

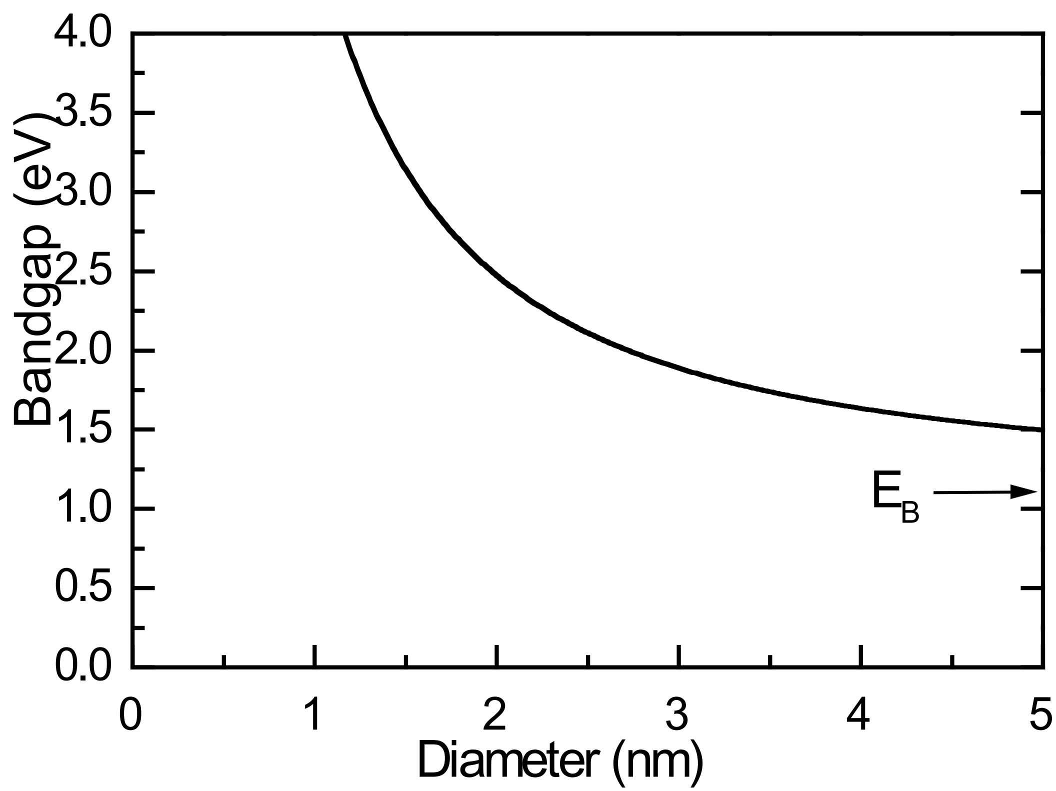
Semiconductor Nanoparticles

Example: Si nanocrystals



Photoluminescence



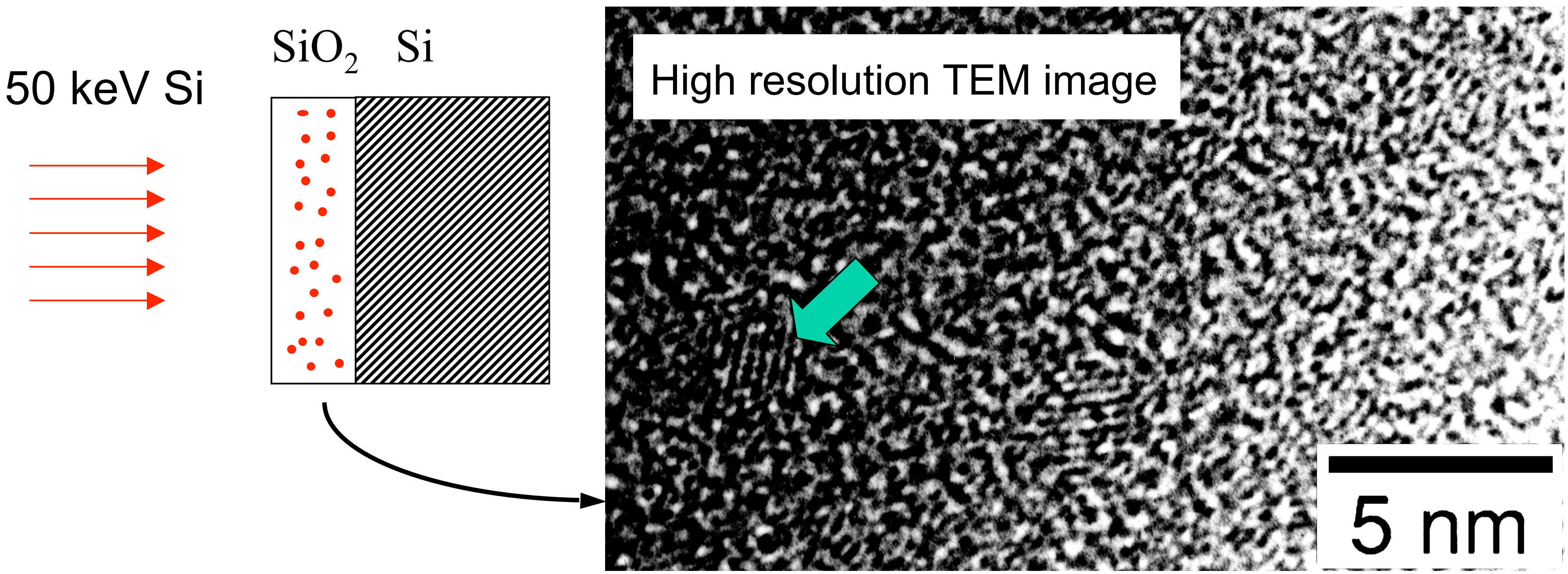


C.Delerue et al. Phys Rev. B 48, 11024 (1993)

Ion Beam Synthesis of Si nanocrystals

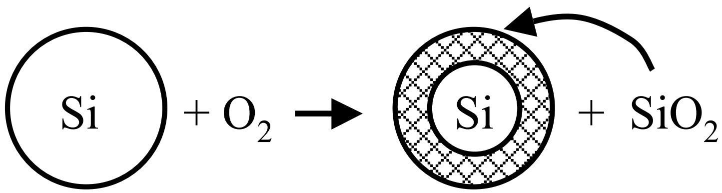
Synthesis of Si nanocrystals

- Implantation: 5×10^{16} Si @ 50 keV → 100 nm SiO_2
- Anneal: 1100 °C/10 min in vacuum
- Hydrogen passivation to 1) quench defect luminescence
  2) increase fraction of optically active nanocrystals

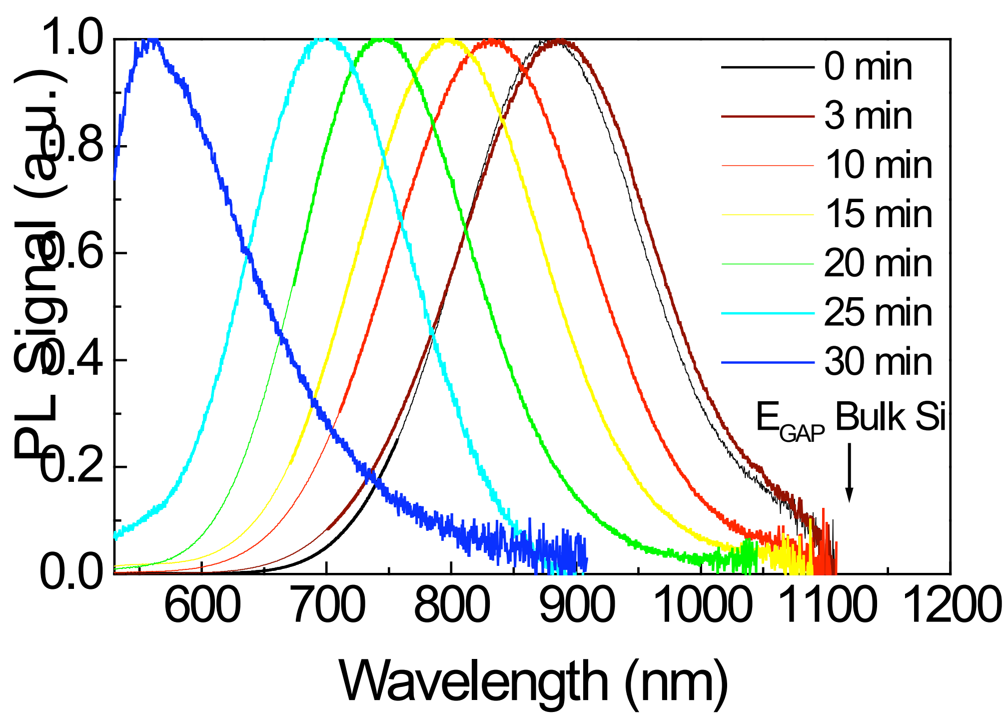


Tuning of the $\lambda_{Emission}$ of Si Nanocrystals by Oxidation

- Oxidation of Si nanocrystals at T = 1000 °C



- Peak wavelength tunable over more than 300 nm



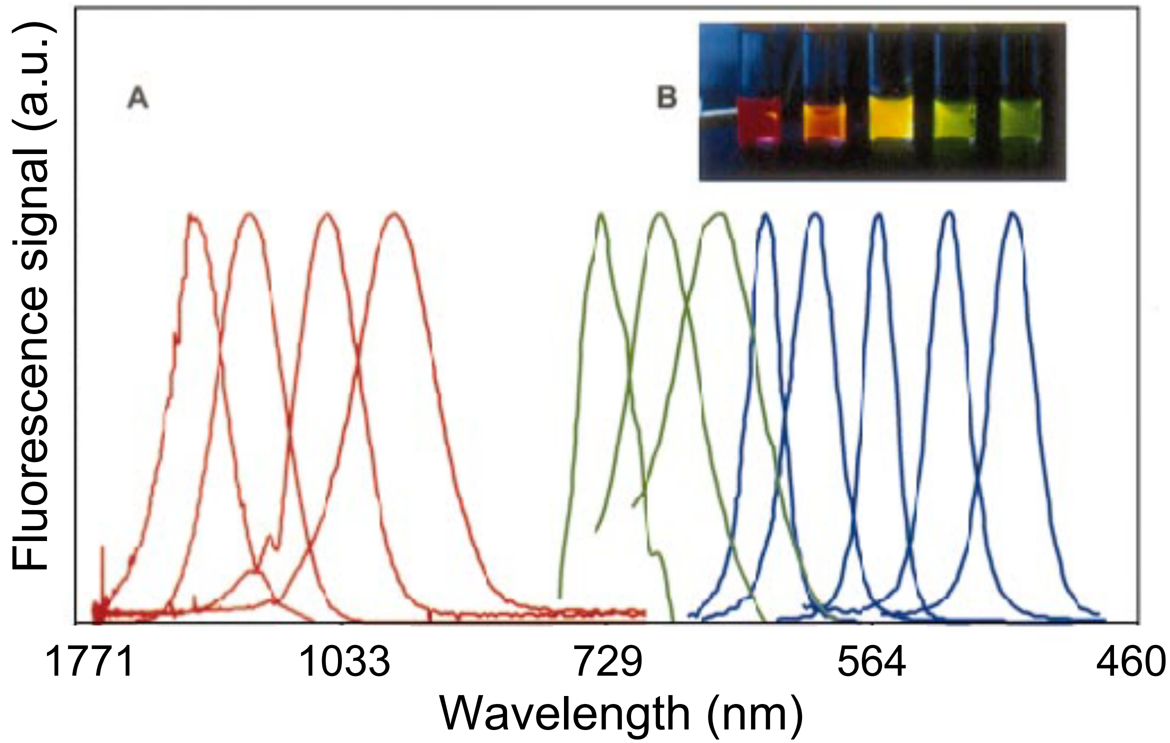
Experimental parameters

$P = 10\ mW/mm^2$

$\lambda_{EXC} = 458$ nm

T = 293 K

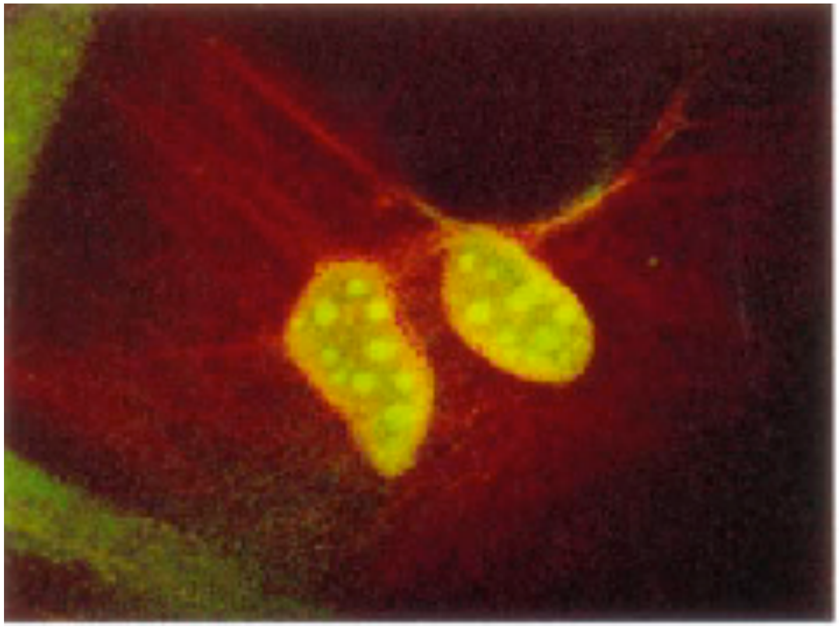
Size and Material Dependent Optical Properties



- Red series: InAs nanocrystals with diameters of 2.8, 3.6, 4.6, and 6.0 nm
- Green series: InP nanocrystals with diameters of 3.0, 3.5,and 4.6 nm.
- Blue series: CdSe nanocrystals with diameters of 2.1, 2.4,3.1, 3.6, and 4.6 nm

*M.Bruchez et al. (Alivisatos group), Science, 2013, **281 (**2014)*

Tagging Biomaterials with Semiconductor Nanocrystals



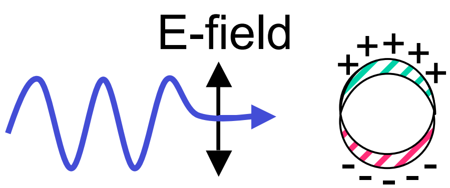
- Confocal microscopy image of Mouse fibroblasts
- Labeling with semiconductor nanoparticles
- 363-nm excitation, observation in the visible

Excitation of a Metal Nanoparticle

Particle

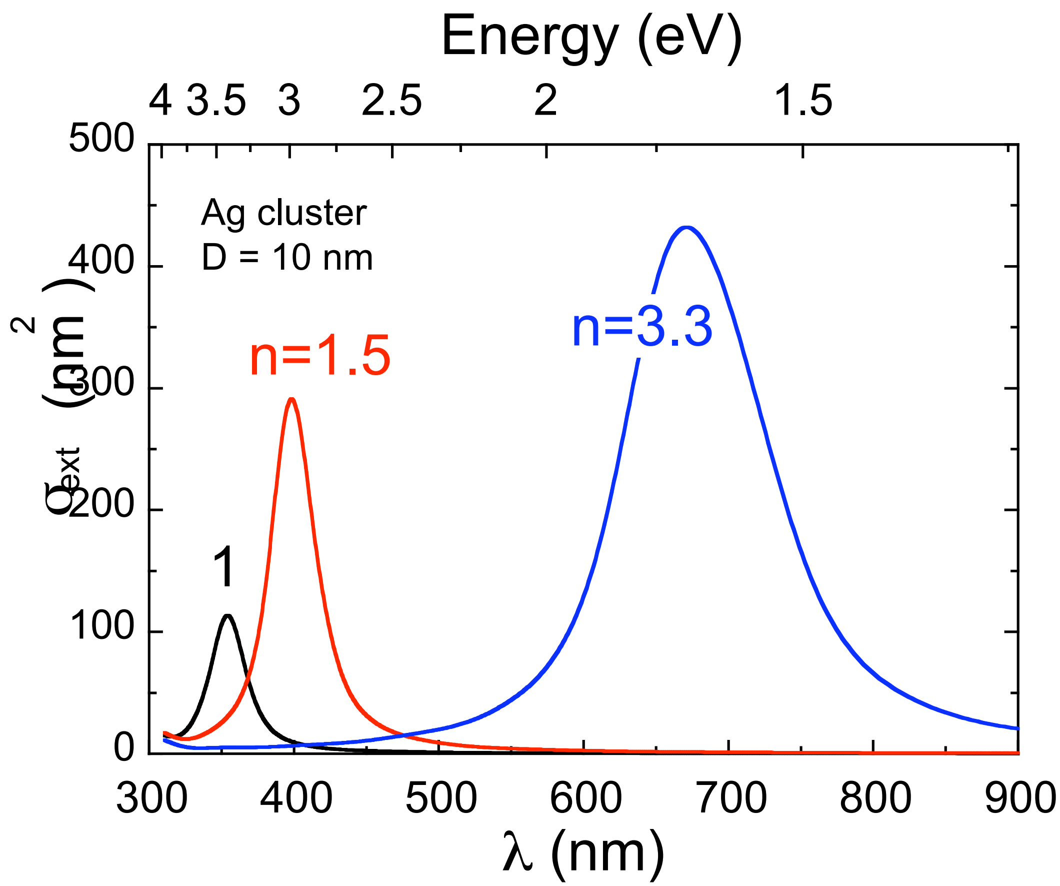
Volume = V_0

$\varepsilon_M = \varepsilon'_M + i\varepsilon''_M$



Host matrix

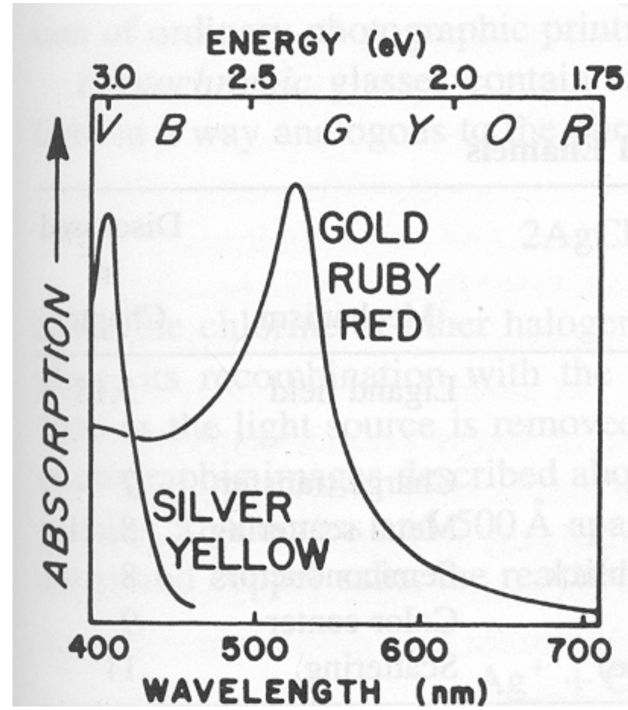
$\varepsilon_H = \varepsilon'_H =$ ■



$$\sigma_{ext}(\omega) = 9\frac{\omega}{c}\varepsilon_H'^{\,3/2} V_0 \frac{\varepsilon'_M(\omega)}{\left[\varepsilon'_M(\omega) + 2\varepsilon'_H\right]^2 + \varepsilon''_M(\omega)^2}$$

Homework problem ☺

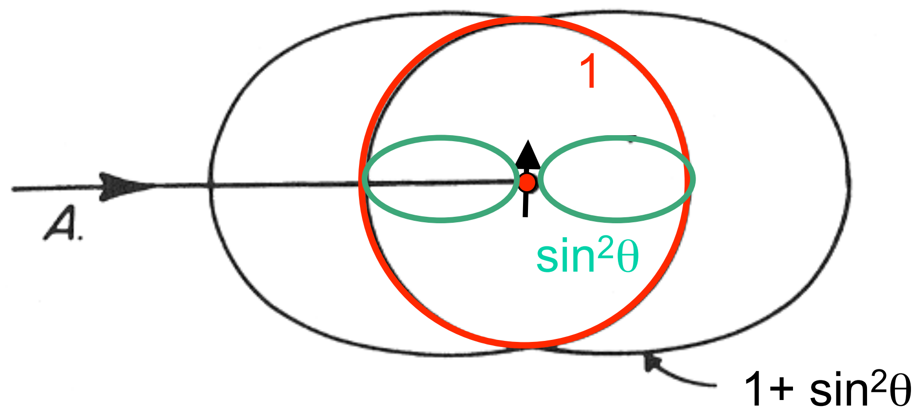
Applications Metallic Nanoparticles



- Engraved Czechoslovakian glass vase
- Ag nanoparticles cause yellow coloration
- Au nanoparticles cause red coloration
- Molten glass readily dissolves 0.1 % Au
- Slow cooling results in nucleation and growth of nanoparticles

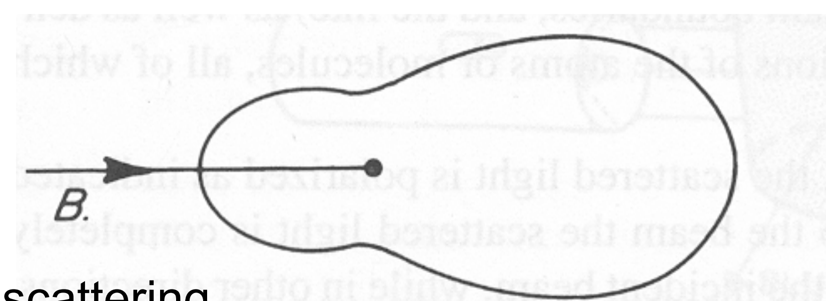
Light scattering by particles with d ≈ λ

Scattering from a driven dipole d << λ



- Red circle intensity from polarization out of the plane
- Black curve show total scattering pattern for a random incident polarization

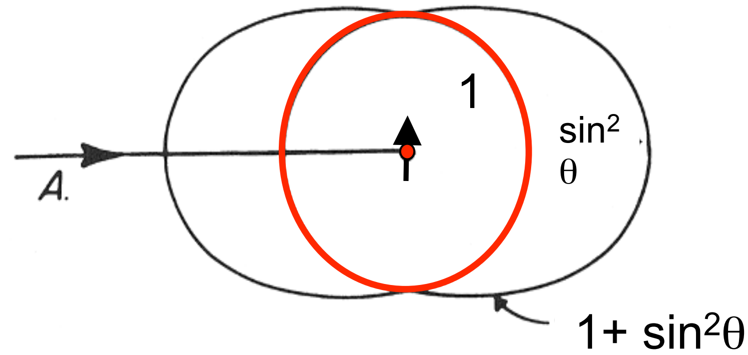
Scattering from a particle with size d ≈ λ



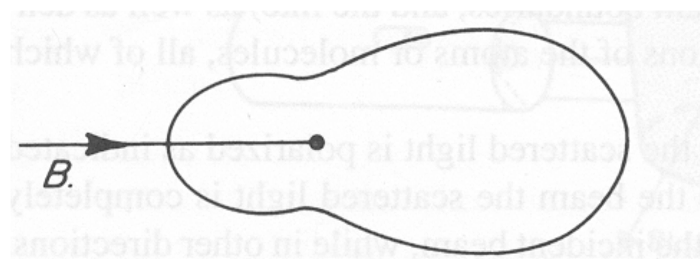
- More forward scattering

Light interaction with particles d ≈ λ

Particle d << λ

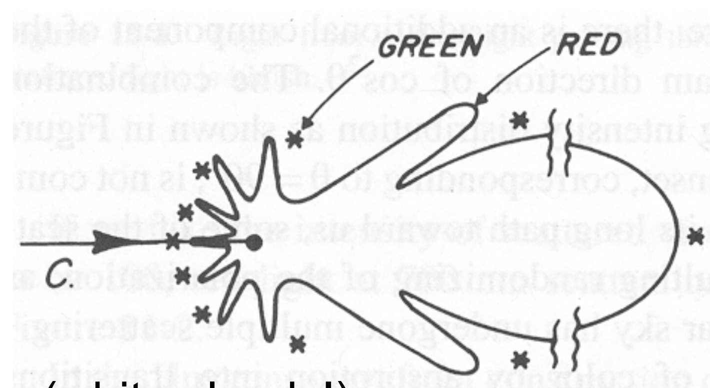


Particle d ≈ λ



- Scattering is more forward

Particle d ≈ 2λ

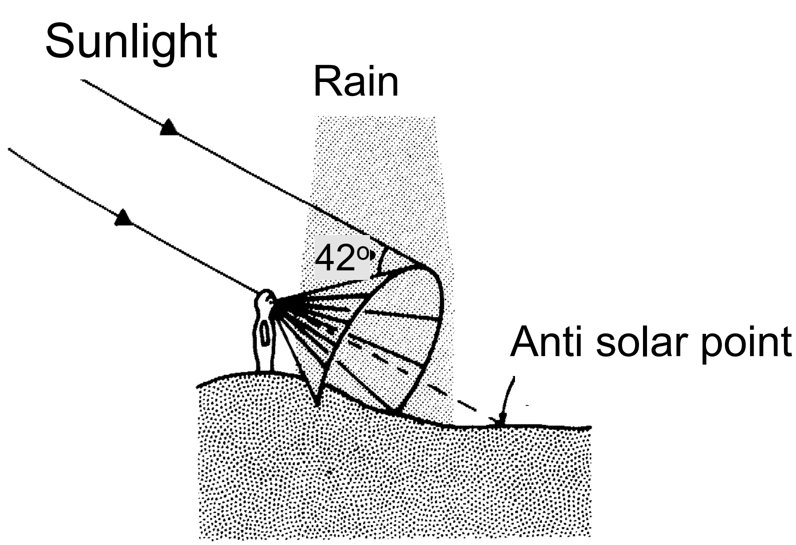


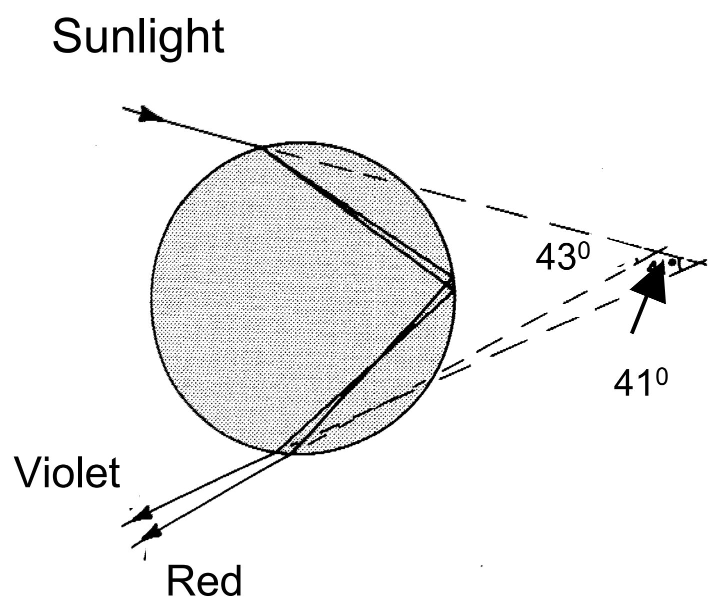
- Very strong forward scattering
- Scattering intensities similar for different colors (white clouds!)
- Scattering maxima occur in different directions for different colors

Light interaction with particles d >> λ

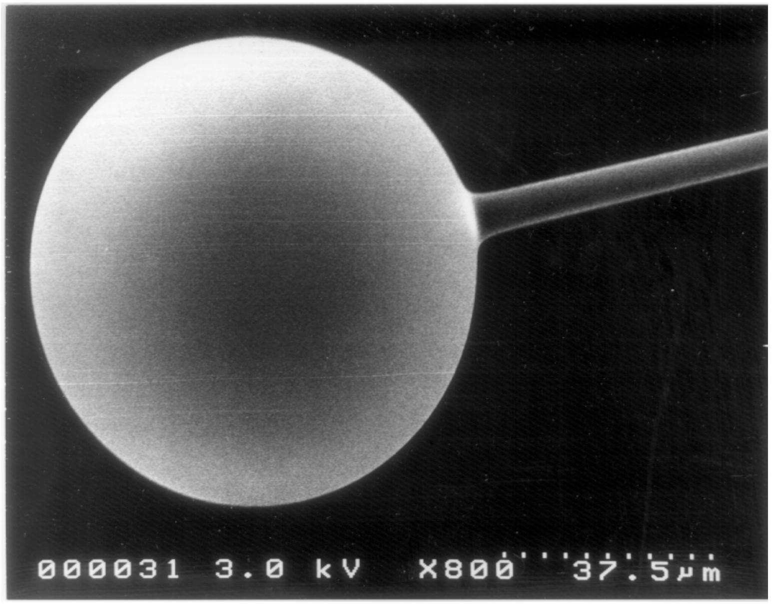
Ray pictures of light become appropriate

- Example: explanation of rainbows

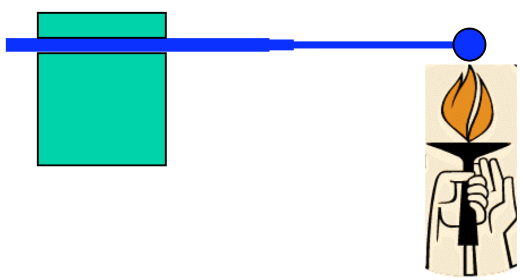




Microspheres with diameters d >> λ



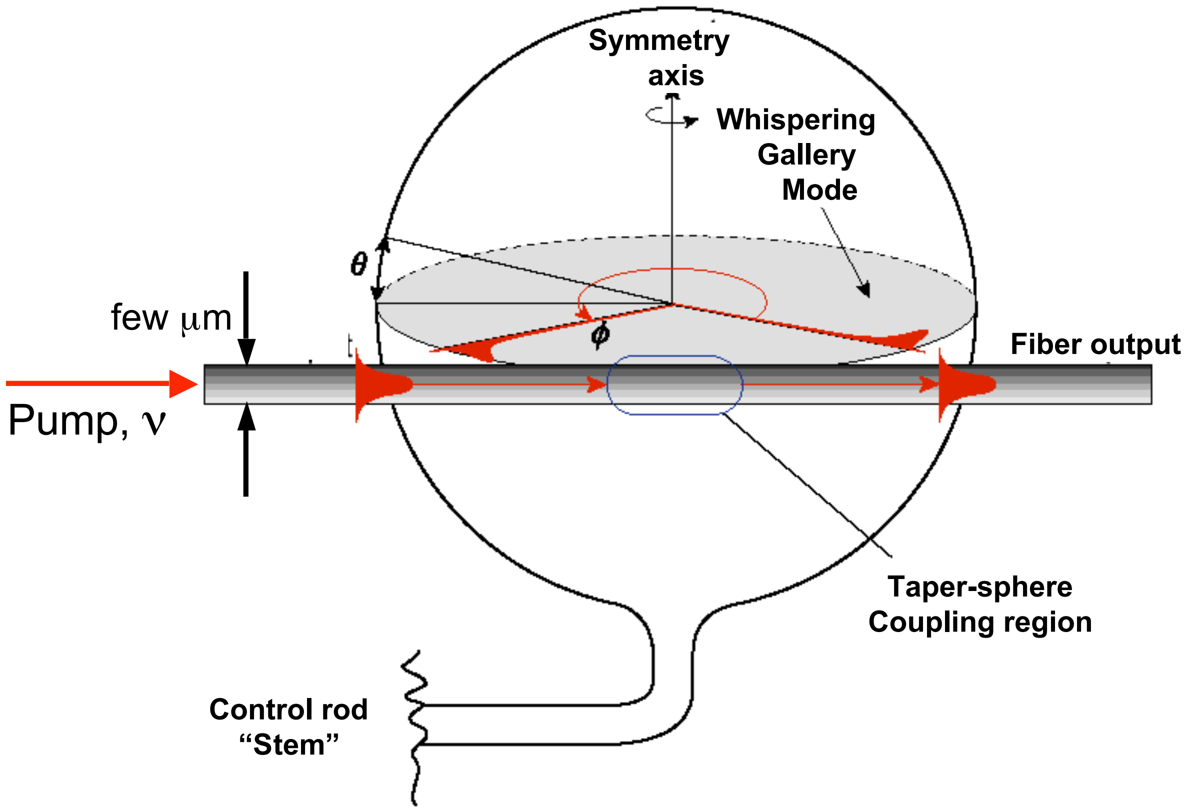
100 μm diameter SiO_2 Microsphere

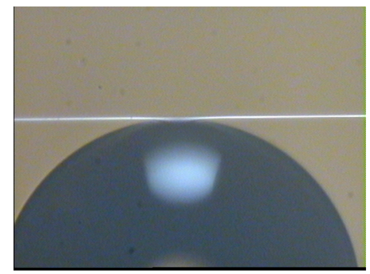


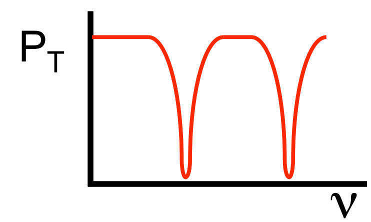
Synthesis of Microspheres

Coupling Light into a SiO_2 Microsphere

- Coupling light into a whispering gallery mode using a tapered fiber





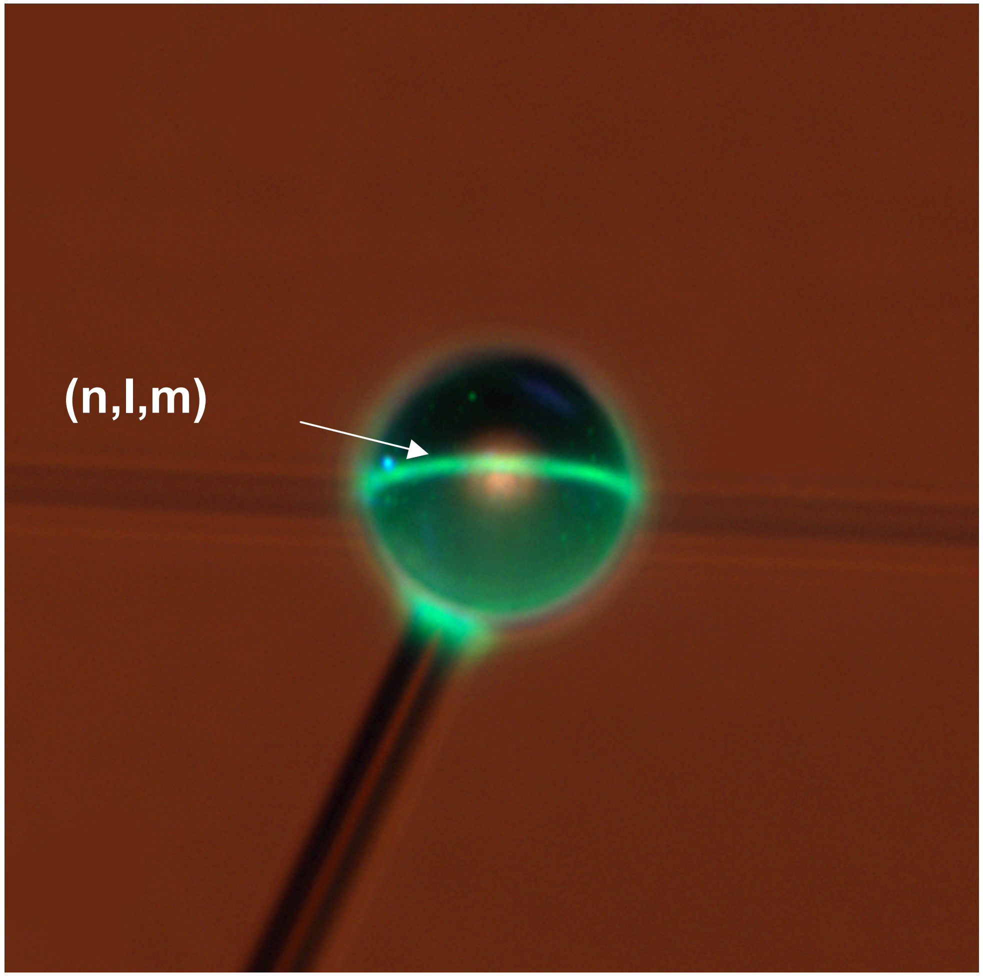


$Q = \Delta\nu/\nu$

$\eta = 99.8\%$

** Ming Cai, Oskar Painter, and Kerry Vahala, Phys Rev. Lett. 85, 74 (2000)*

Microsphere Doped with Er ions



- Similarity with electron orbits
- Er doped sphere lases ! (Consider roundtrip loss and gain)

Microsphere can act as a Microresonator

Performance

- Determined by quality factor Q = $\dfrac{\text{Photon lifetime}}{\text{Optical period}}$
- Q-values of 10^3 - 10^4 are considered excellent
- Measured Q-value of a SiO_2 microsphere resonator: $\sim 10^{10}$! *

  Photon storage time $\sim \mu s$

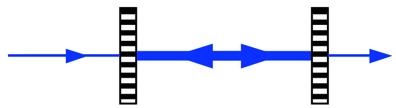
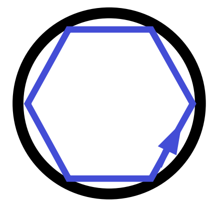
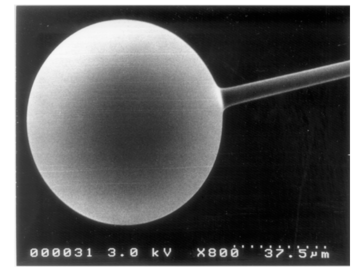
  Photon comes around 10^6 times for D = 100 μm

- Ultimate Q of 10^{10} is limited by intrinsic material properties

* *M.L. Gorodetsky, A.A. Savchenkov, and V.S. Ilchenko, Opt. Lett.* ***21****, 453 (1996)*

MicrOspheres: Devices and Applications

Comparison with more conventional resonators

- Examples:  or  or 

- Characteristic dimensions: λ - 100λ

Devices and applications

- Low threshold micro-lasers
- Narrow linewidth optical filters
- Sensors with submonolayer sensitivity
- Wavelength Division Multiplexing devices for telecommunications
- Non-linear optics
- Quantum electrodynamics experiments

Summary

Light interaction with small objects ($d < \lambda$)

- Light scattering due to harmonically driven dipole oscillator

Nanoparticles

- Insulators…Rayleigh Scattering (blue sky)
- Semiconductors....Resonance absorption at $\hbar\omega \geq E_{GAP}$, size dependent fluorescence…)
- Metals…Resonance absorption at surface plasmon frequency, no light emission)

Microparticles

- Particles with dimensions on the order of λ ⇨ Enhanced forward scattering
  ⇨ λ-independent (white clouds)

- Microspheres with diameters much larger than λ
  ⇨ Intuitive ray-picture useful
  ⇨ Rainbows due to dispersion $H_2 0$
  ⇨ Applications: resonators, lasers, etc…