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Conference on Nanotechnology for Biological and Biomedical Applications (Nano-Bio-Med)

10 - 14 October 2011

Nonlinear Light Scattering and Luminescence from Nanoparticles and Biological Cells

Hai-Lung DAI

Dean, College of Science & Technology Temple University
1803 N.Broad St.(04103)
Philadelphia
U.S.A.

Nonlinear Light Scattering and Luminescence from Nanoparticles and Biological Cells

Minchul Yang
Hongfei Wang
Heather Eckenrode
Jia Zeng
Gan Wei
Rosa Chou
Angong Ye (DuPont)
Susan Dounce
Thomas Troxler
Shih-Hui Jen
Grazia Gonella
Min Zhang
Bolei Xu

Hai-Lung Dai
Temple University

College of Science and Technology
TEMPLE UNIVERSITY

The use of nanoparticles in Imaging and Sensing

- Optically bright for detection
- Surface functionalization for biocompatibility
- External field control for maneuver

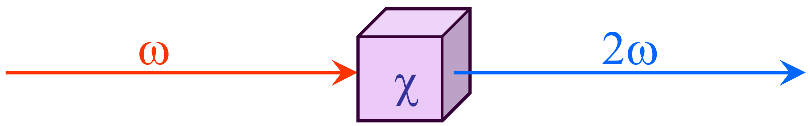
- **Fluorescence microscopy** –

  relies on fluorescence quantum efficiency
- **Nonlinear optical microscopy** –

  need to understand nonlinear light scattering

Nonlinear light scattering from nanoparticles

- 2^{nd} order nonlinear light scattering
  second harmonic generation (SHG)



$$\vec{P} = \varepsilon_0(\chi^1 \vec{E} + \vec{\vec{\chi}}^2 \vec{E}\vec{E} + \ldots)$$

$\chi^2 = 2^{nd}$ order susceptibility

(sum frequency generation (SFG))

Symmetry dependent – **for matters with inversion symmetry, no SHG** - and therefore surface sensitive

FIRST THEORETICAL TREATMENT OF Nonlinear Light Scattering from AN INTERFACE, 1962

PHYSICAL REVIEW VOLUME 128, NUMBER 2 OCTOBER 15, 1962

Light Waves at the Boundary of Nonlinear Media

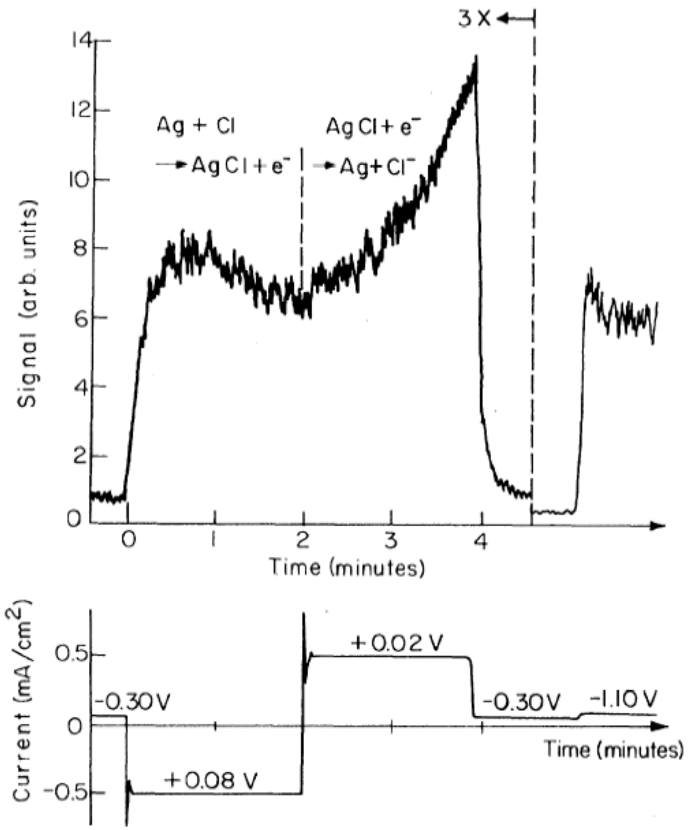
N. Bloembergen and P. S. Pershan
Division of Engineering and Applied Physics, Harvard University, Cambridge, Massachusetts

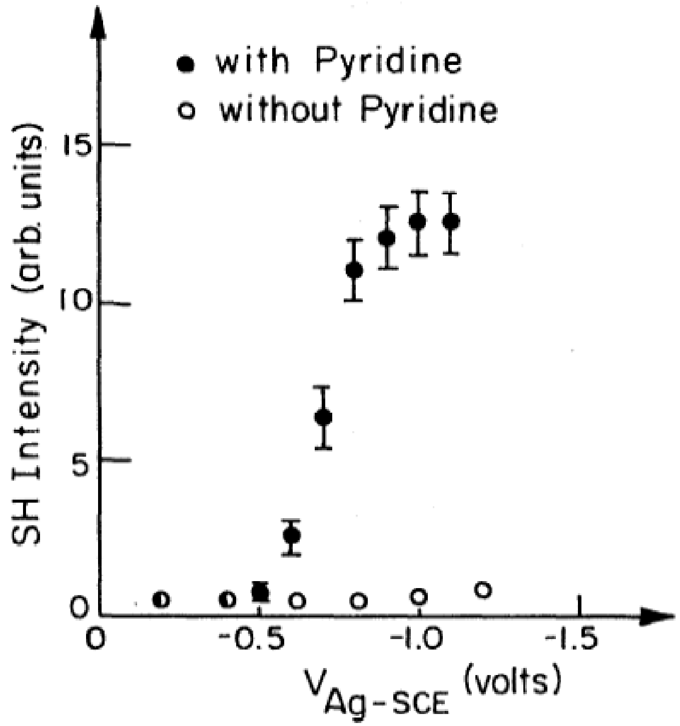
(Received June 11, 1962)

Solutions to Maxwell's equations in nonlinear dielectrics are presented which satisfy the boundary conditions at a plane interface between a linear and nonlinear medium. Harmonic waves emanate from the boundary. Generalizations of the well-known laws of reflection and refraction give the direction of the boundary harmonic waves. Their intensity and polarization conditions are described by generalizations of the Fresnel formulas. The equivalent Brewster angle for harmonic waves is derived. The various conditions for total reflection and transmission of boundary harmonics are discussed. The solution of the nonlinear plane parallel slab is presented which describes the harmonic generation in experimental situations. An integral equation formulation for wave propagation in nonlinear media is sketched. Implications of the nonlinear boundary theory for experimental systems and devices are pointed out.

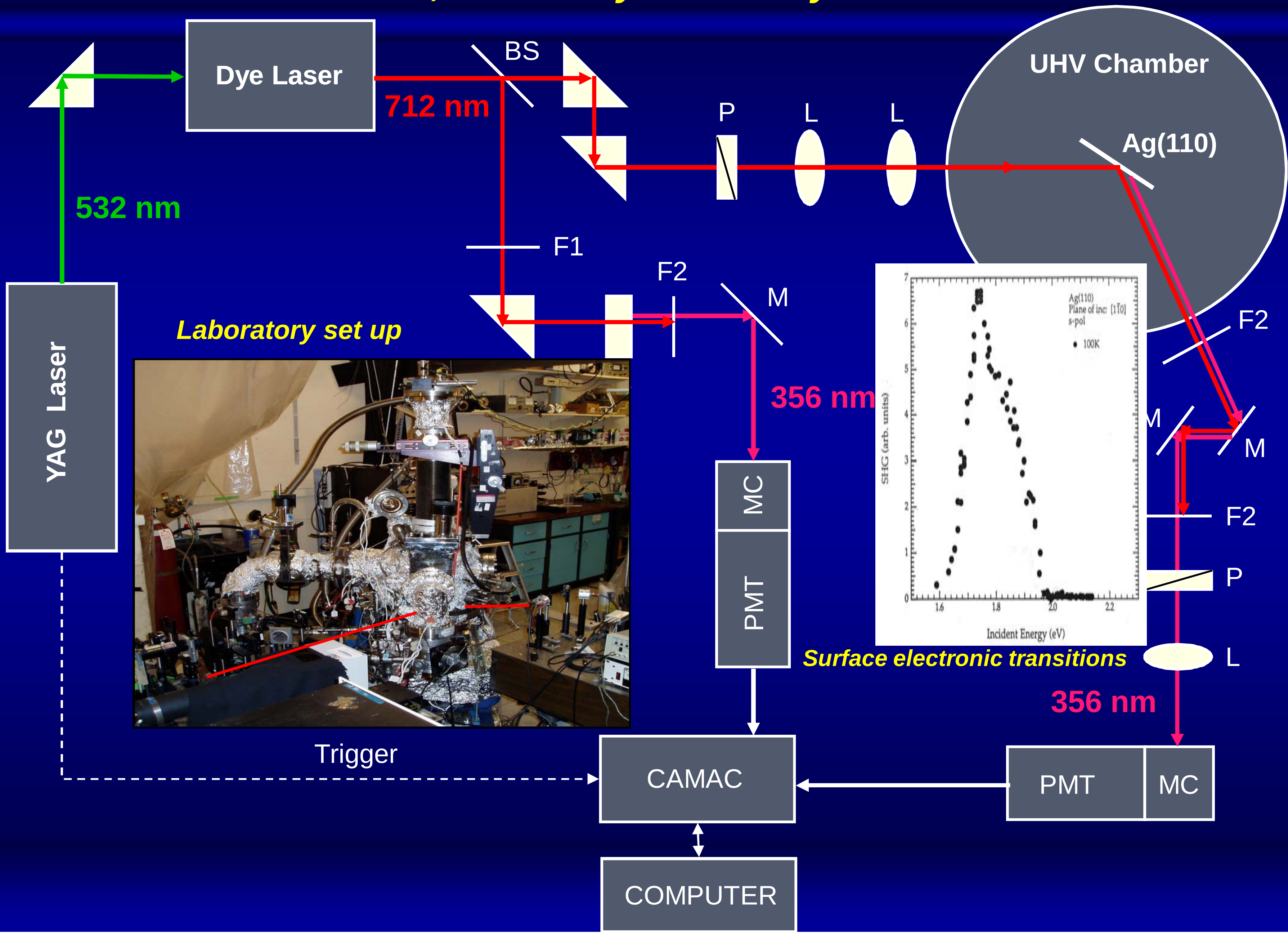
Detection of Molecular Monolayers by Optical Second Harmonic Generation

Yuen Ron Shen and coworkers,
PRL 46, 1010 (1981)

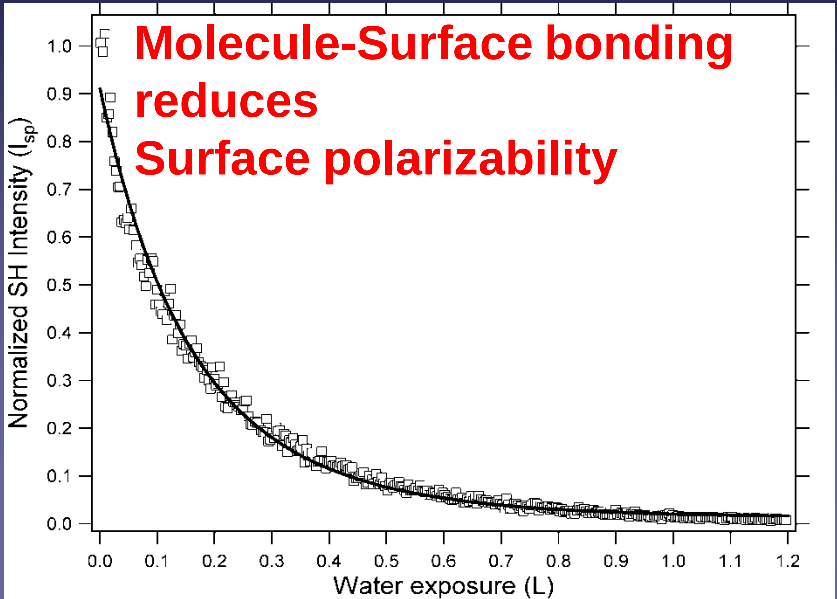


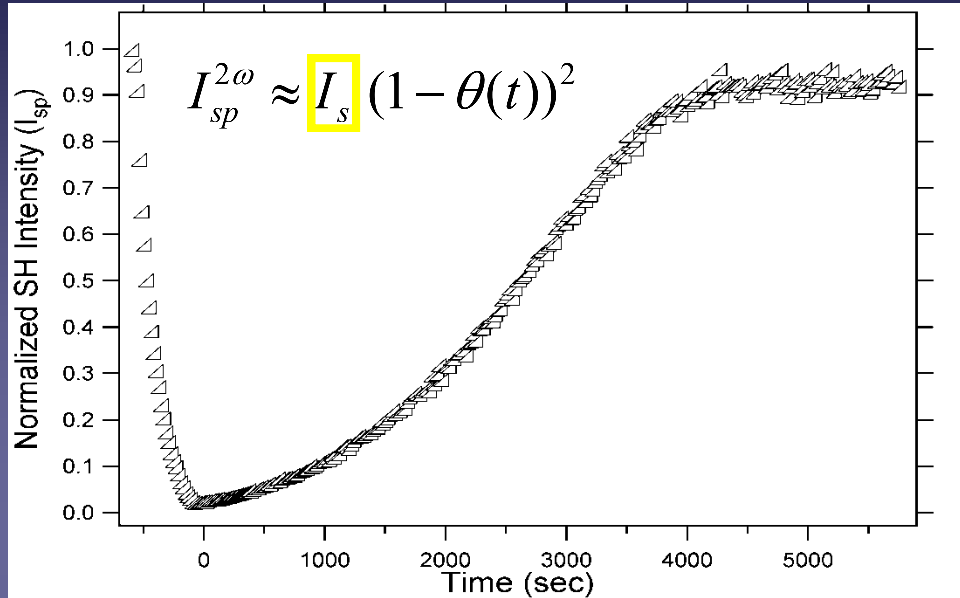


Nonlinear Optical Probe of Molecules on Surfaces and Interfaces
H. L. Dai, University of Pennsylvania ~1988



Monitoring isothermal water adsorption / desorption on Ag(111) through surface generated SHG





Phenomenological model of second-order susceptibility:

$$I_{sp}^{2\omega} \approx I_s (1-\theta(t))^2$$

Kinetics of desorption

$$k_d = k_o \exp\left(-E_d / RT \right)$$

Desorption energy increases with coverage:

$$E_d = E_o + \alpha \theta(t)$$

First order ODE:

$$\frac{-d\theta(t)}{dt} = C \theta(t) \exp\left(-\alpha\theta(t) / RT \right)$$

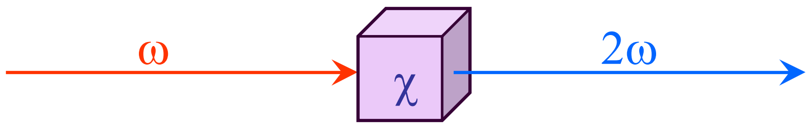
where: $C = k_o \exp\left(-E_o / RT \right)$

Susan Dounce, *JCP* 2005

SHG from nanoparticles

1. SHG from molecules adsorbed on surface of micron size polystyrene particles
2. Effect of particle size on SHG from colloidal particles
3. SHG from molecules adsorbed on surface of nanometer size polystyrene particles
4. SHG from metallic nanoparticles

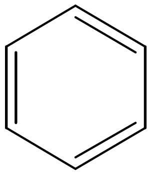
Second Harmonic Generation:
Symmetry Sensitive Structural Probe of molecular ensemble



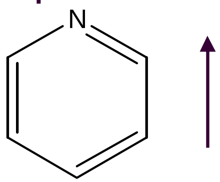
$$\vec{P} = \varepsilon_0(\chi^1 \vec{E} + \vec{\vec{\chi}}^2 \vec{E}\vec{E} + \ldots)$$

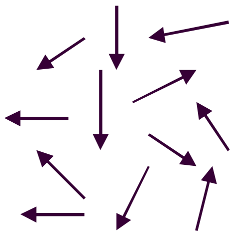
$$\chi^2 = 2^{nd} \text{ order susceptibility}$$

Benzene NO SHG



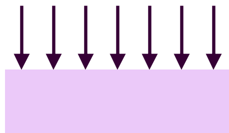
Pyridine SHG!
Hyperpolarizability is a vector



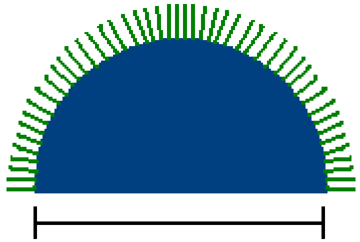


NO SHG
From ensemble of randomly oriented molecules

At a surface, molecules orient in same direction SHG!

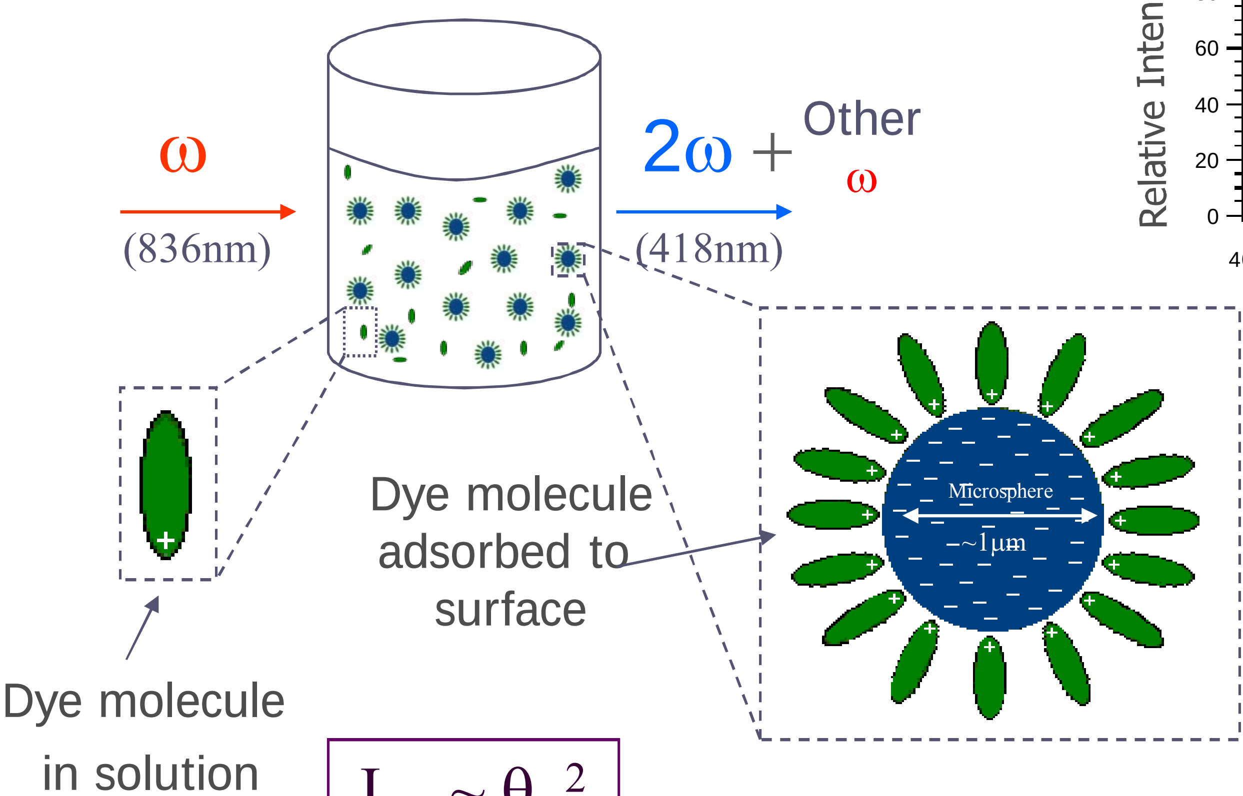


Colloidal particle surface, SHG!

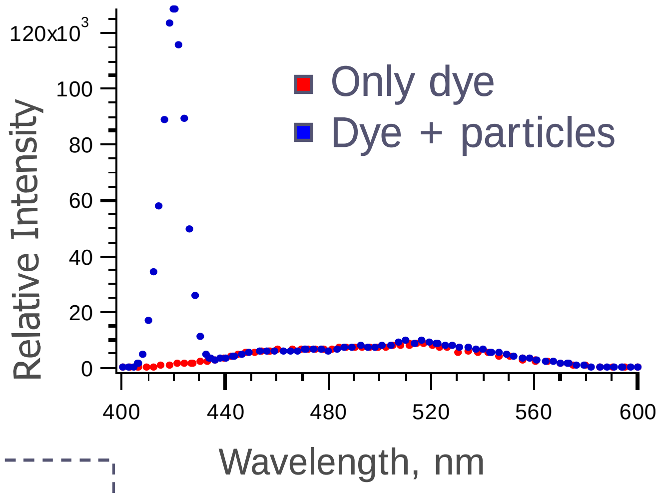


Second Harmonic Generation from molecules adsorbed on polystyrene particles

HF Wang, *Langmuir* 2000



$$I_{SH} \sim \theta_D^2$$

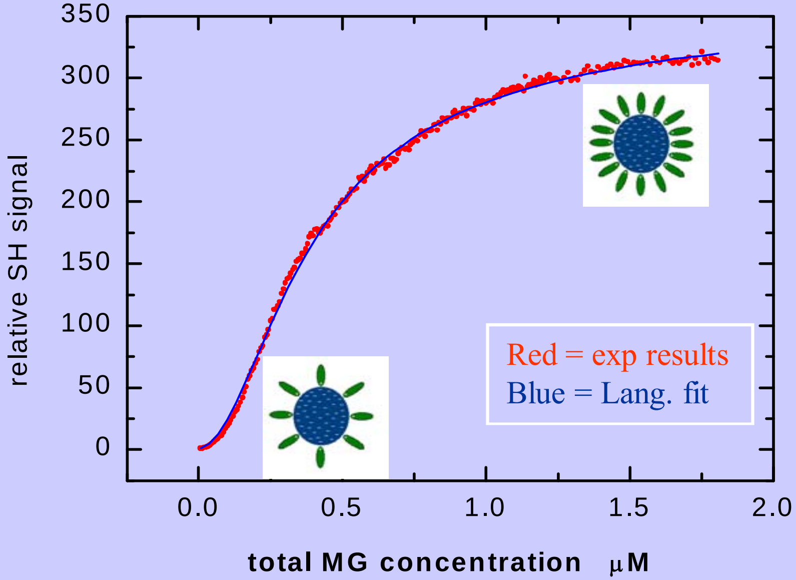


Particle size d ≥ λ

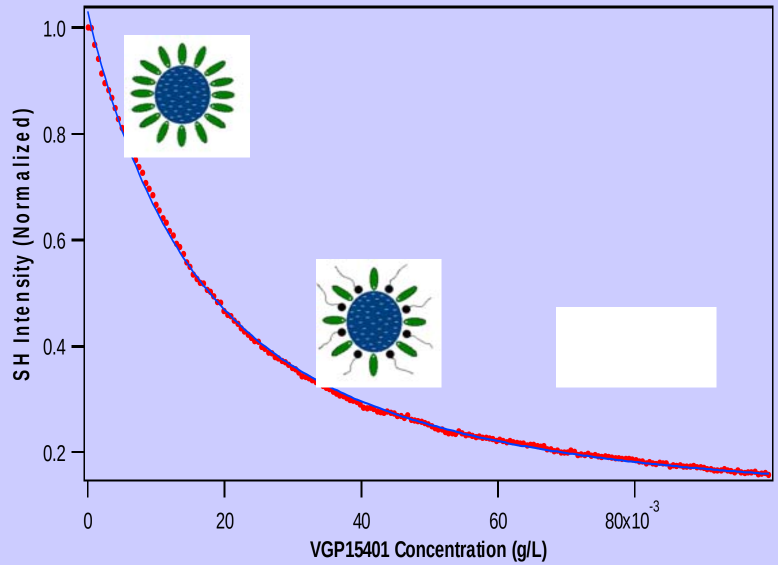
Eisenthal and coworkers, 1996: SHG from MG on polystyrene particles

Adsorption of surfactant on PS particle

PSS/MG



PSS/MG/VGP



ΔG=-12.4 ±0.2kcal/mol

N_{max}=0.9±0.1*10^6dye/PSS

ΔG=-12.3±0.1kcal/mol

N_{max}= 0.73±0.29*10^6VGP/PSS

VGP MW 8500, 480 A2 = 10 out 100 methacrylate units adsorb on surface

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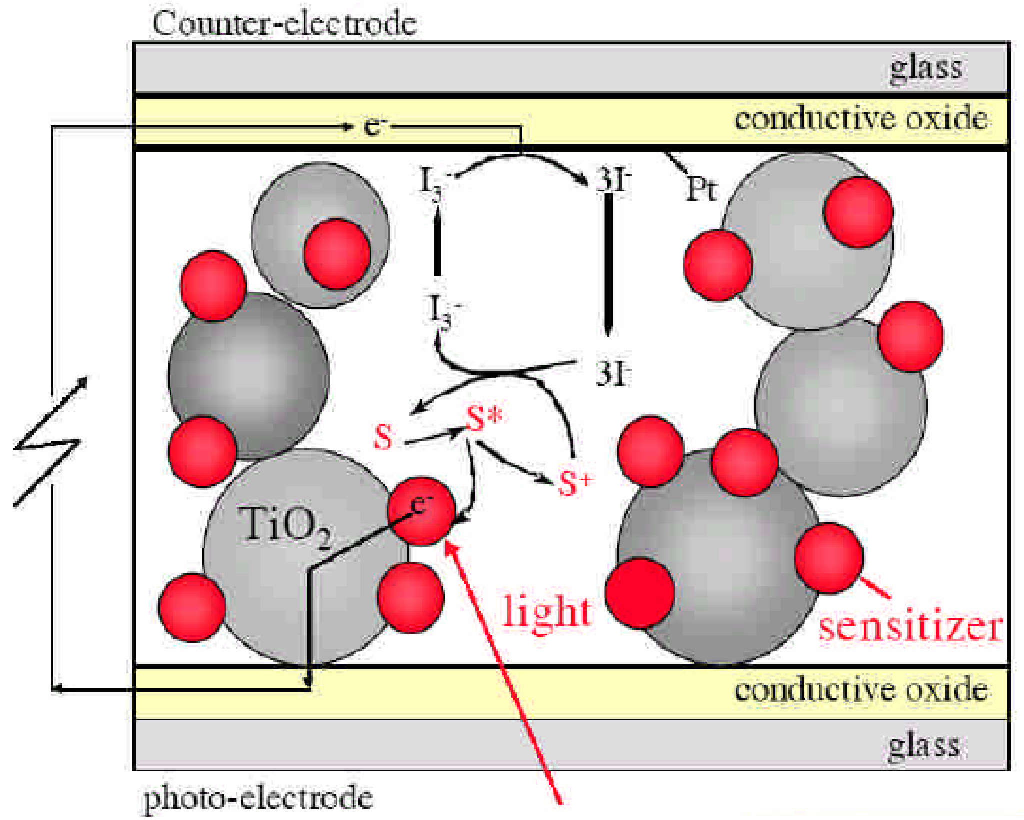
Prototypical Dye Sensitized Solar Cell (DSSC)

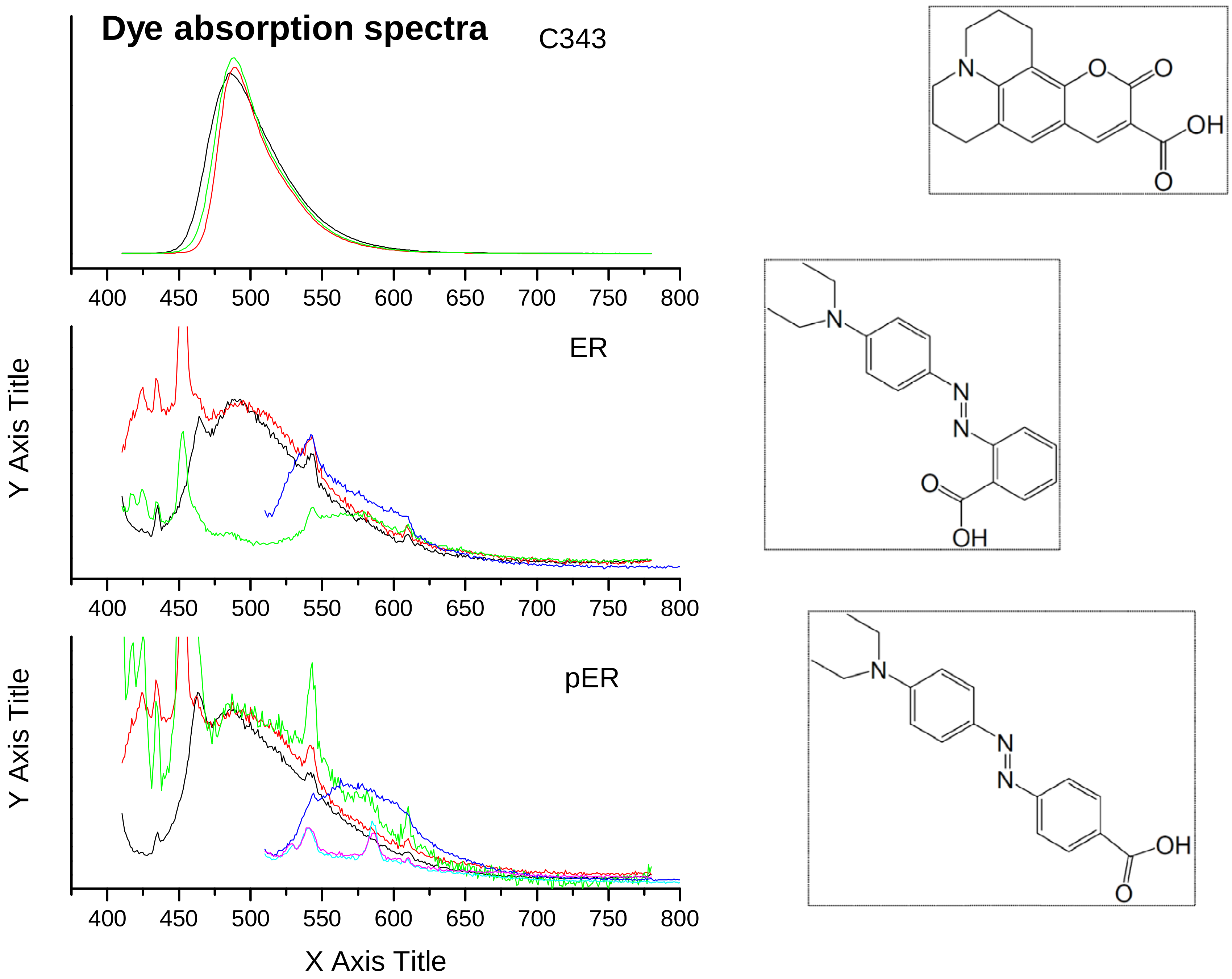


$$\text{dye} + h\nu \rightarrow \text{dye}^*$$

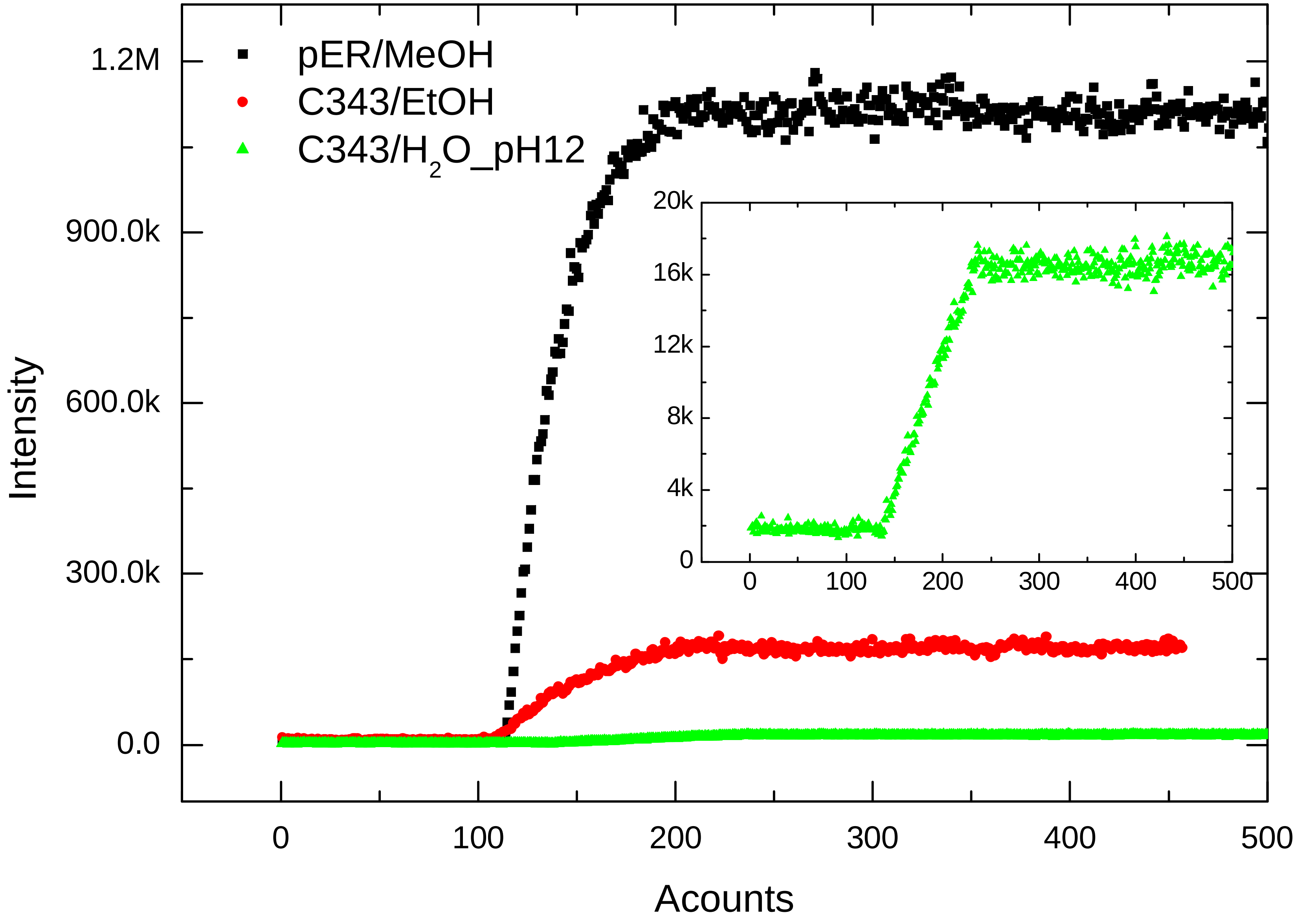
$$\text{dye}^* \rightarrow \text{dye}^+ + e^-$$

$$2e^- + 3I_3^- \rightarrow 3I^-$$

$$3I^- \rightarrow I_3^- + 2e^-$$

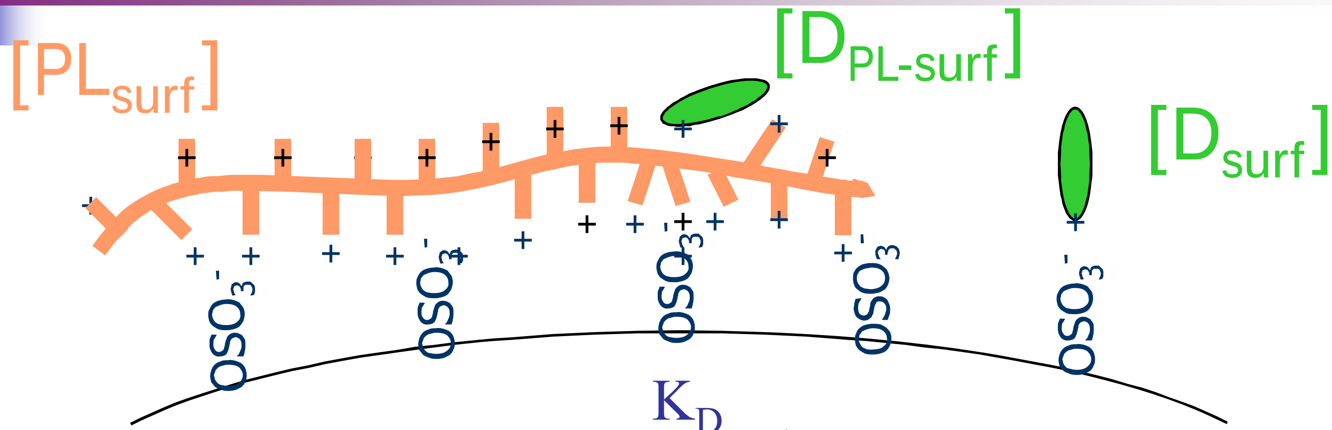

Dye absorption spectra
C343
ER
pER
Y Axis Title
Y Axis Title
X Axis Title
400
450
500
550
600
650
700
750
800
N
O
O
OH
O
N
N
N
O
OH
N
N
N
OH
O

SHG probe of dye adsorption on TiO2 particles in different solvents



Model for MG/PL Adsorption on PS
(peptides on biochips)

H. Eckenrode, *Langmuir* 2004



$$D + SS \underset{}{\overset{K_D}{\rightleftharpoons}} D\text{-}SS$$

$$PL + SS \overset{K_{PLL}}{\rightleftharpoons} PL\text{-}SS$$

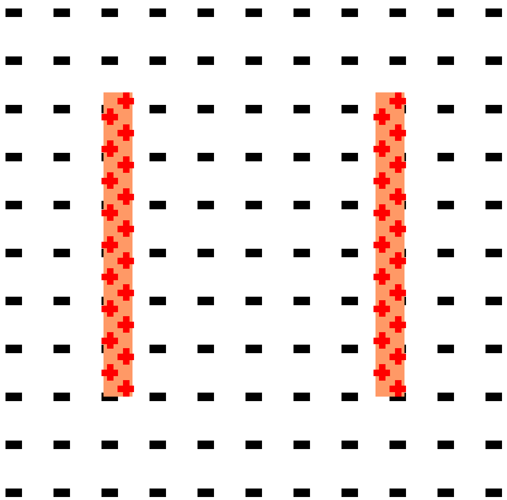
$$D + PL\text{-}PS \rightleftharpoons D\text{-}PL\text{-}PS$$

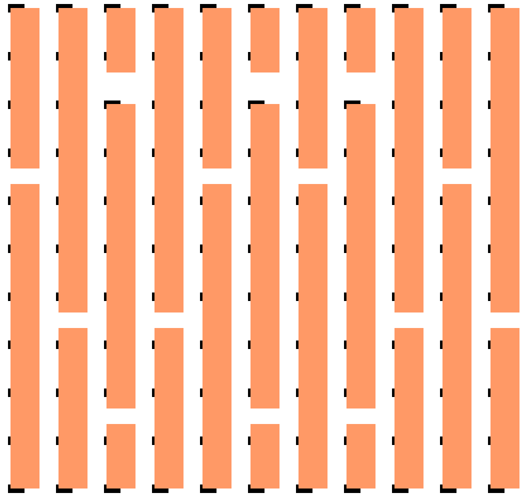
$$I = B + \left(\alpha_1 \theta_{D-Surf} + \alpha_2 \theta_{D-PL-Surf} e^{i\delta}\right)^2$$

$$\theta_{D-Surf} = \frac{\left[\begin{array}{l} -\left(K_D N_{\max}^{D} - K_{PLL} N_{\max}^{PLL} + K_D[D] + K_{PLL}[PLL]\right) + \\ \left(\left(K_D N_{\max}^{D} - K_{PLL} N_{\max}^{PLL} + K_D[D] + K_{PLL}[PLL]\right)^2 + 4K_D[D]\left(K_{PLL} N_{\max}^{PLL} - K_D N_{\max}^{D}\right)\right)^{1/2} \end{array}\right]}{2\left(K_{PLL} N_{\max}^{PLL} - K_D N_{\max}^{D}\right)}$$

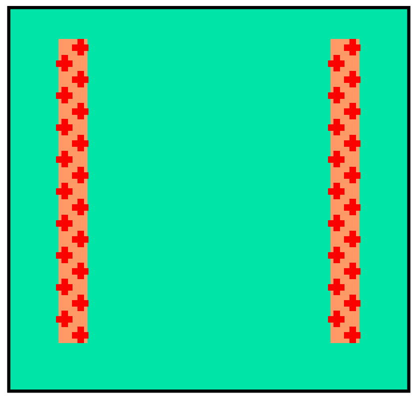
$$\theta_{D-PL-Surf} \propto \theta_{PL-Surf} = 1 - \theta_{D-Surf}$$

Peptide adsorption on biochips: Effect of Charge Repulsion





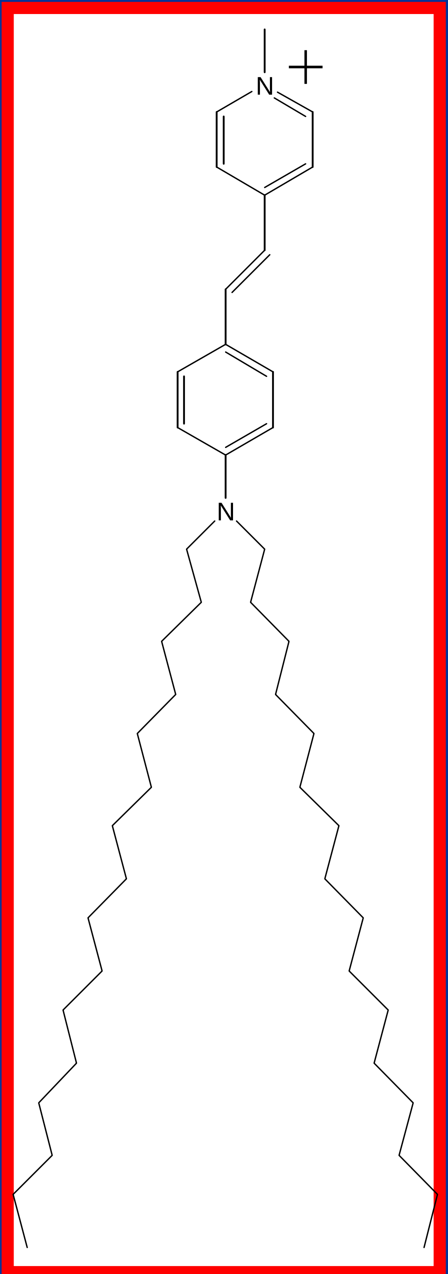
$$E = V_{++} + V_{+-} - \Delta V_{solv}$$

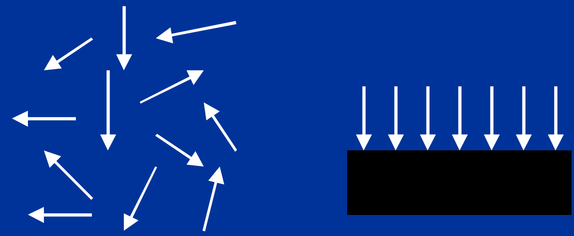


PL75 in H_2O

V_{++}=150 kcal/mol

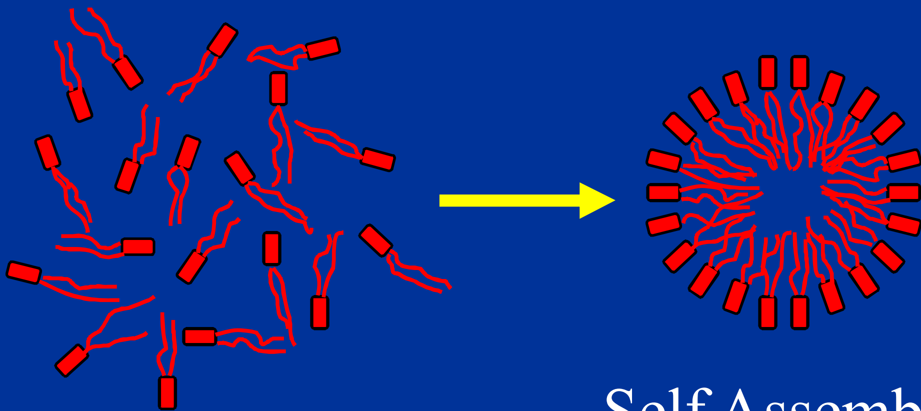
Probing formation of vesicle: DiA





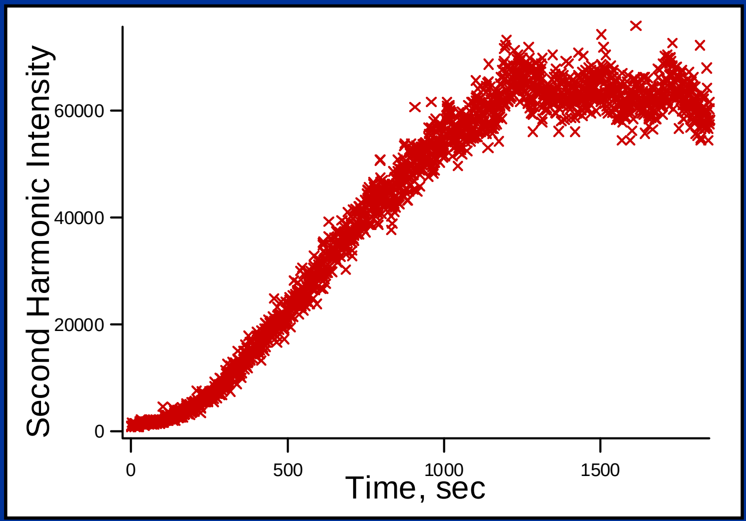
NO SH

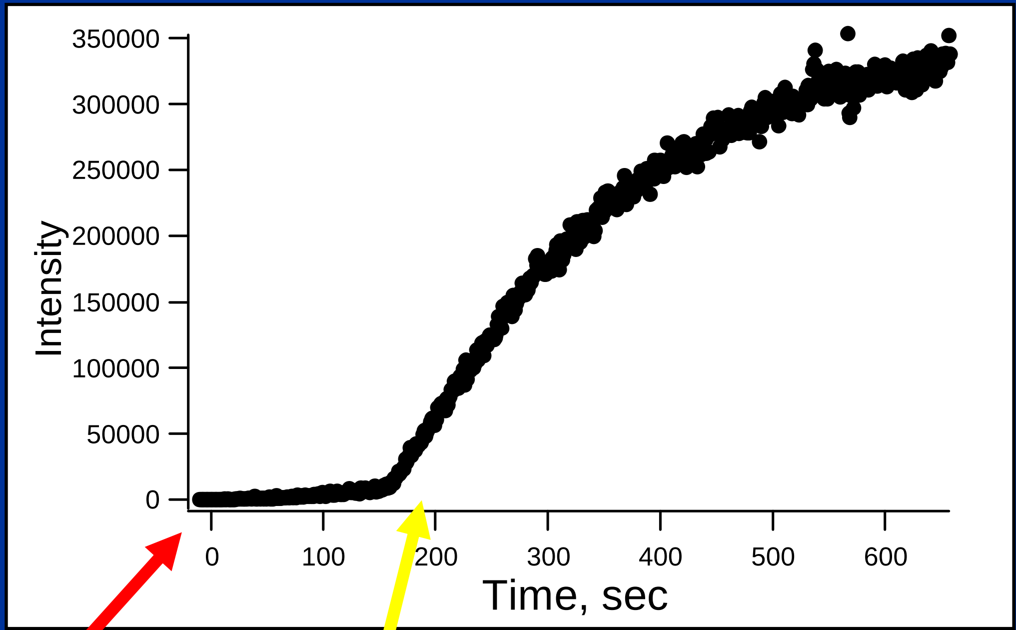
SH!



Self Assembly

Salt Effect on emulsion formation





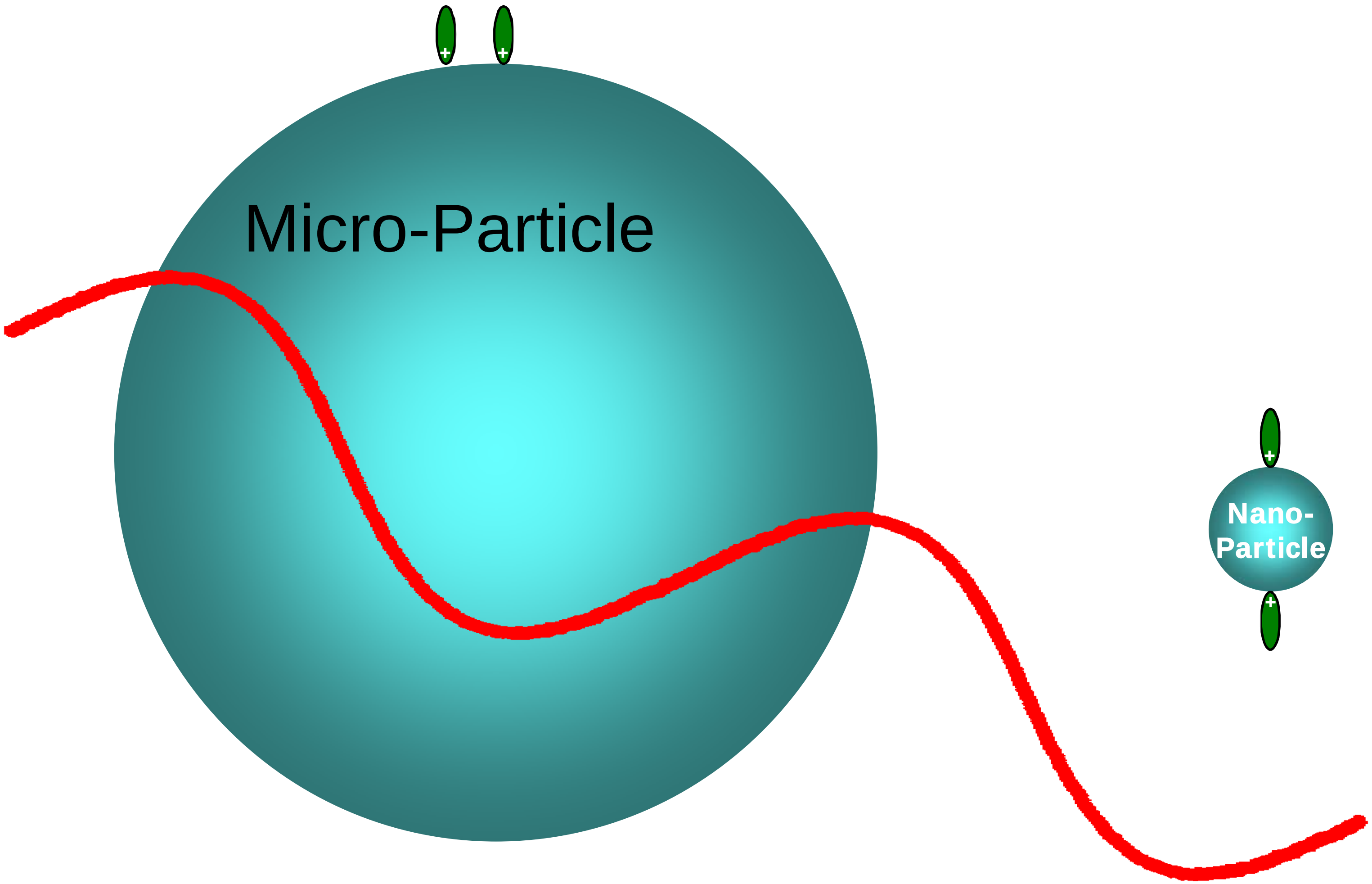
DiA/Ethanol Addition

Salt Addition 0.1M NaCl

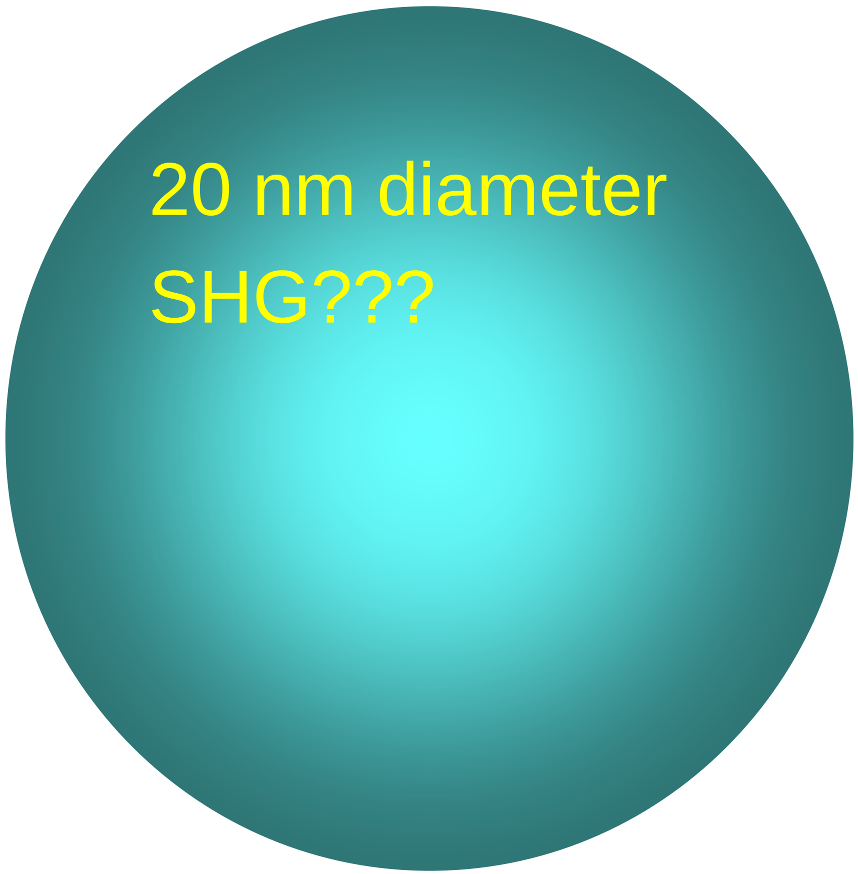
- 1700 time increase in slope!
- Particle size measured after salt addition: 0.37μm

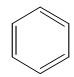
Q1. SHG from nanoparticle surface?

Particle size d << wavelength 800 nm!



Q1: How small is small?
Q2: SHG from surface of spherical, (metallic) nanoparticle?



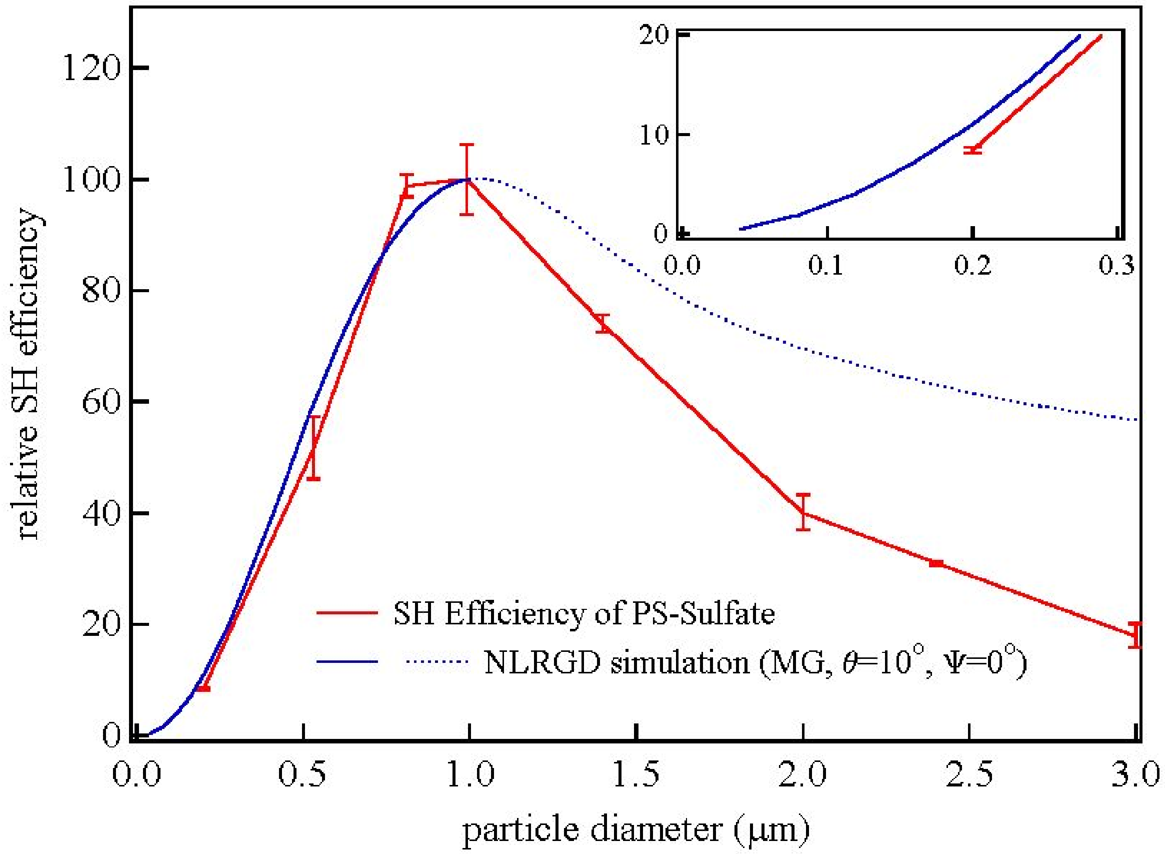


Benzene
No SHG

Particle size effect in SHG from particle surface

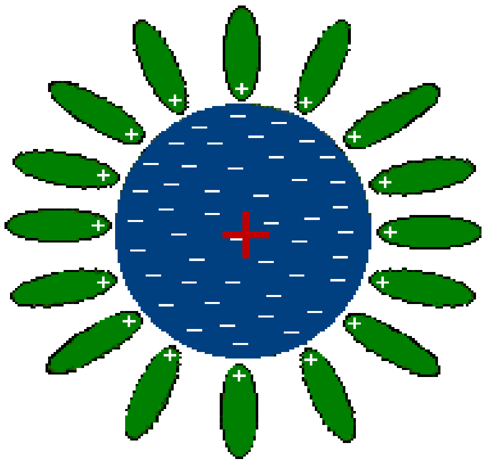
Polystyrene- Sulfate (SO_3^-)Particle size (diameter):

0.2, 0.53, 0.81, 0.99, 1.4, 2.0, 2.4, 3.0 μm; SHG detected at **forward scattering** direction

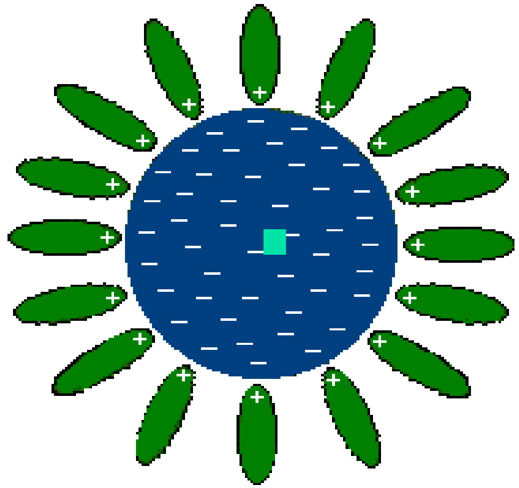


SH Jen, *JPCA* 2009; *JPCC* 2010

The molecules at the edge of the cross section perpendicular to the beam propagation are responsible for the forward direction detection



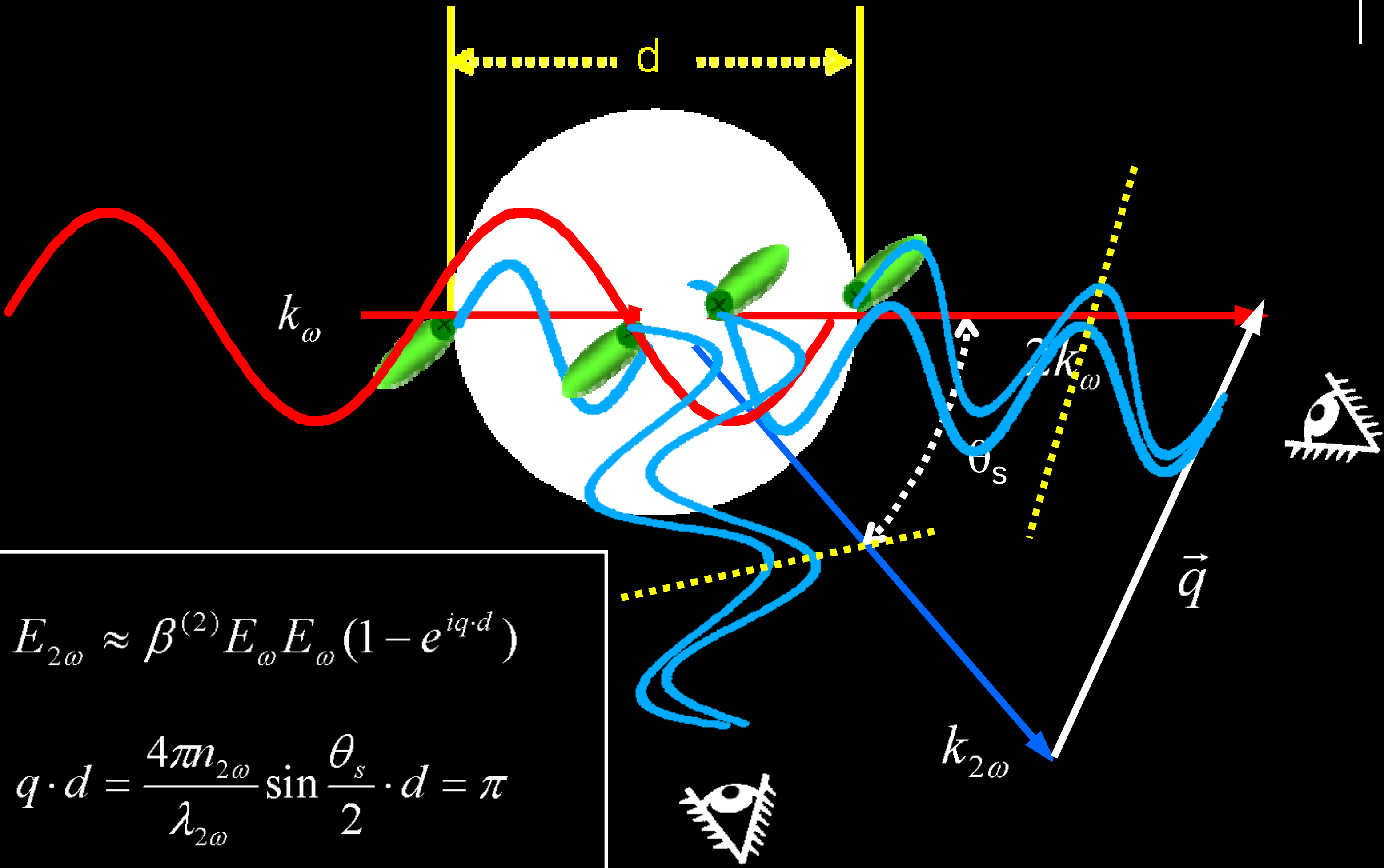
fundamental



2nd harmonic

Size Effect of SHG from Colloidal Particles

Phase matching at larger scattering angle with decreasing particle size



$$E_{2\omega} \approx \beta^{(2)} E_\omega E_\omega (1 - e^{iq\cdot d})$$

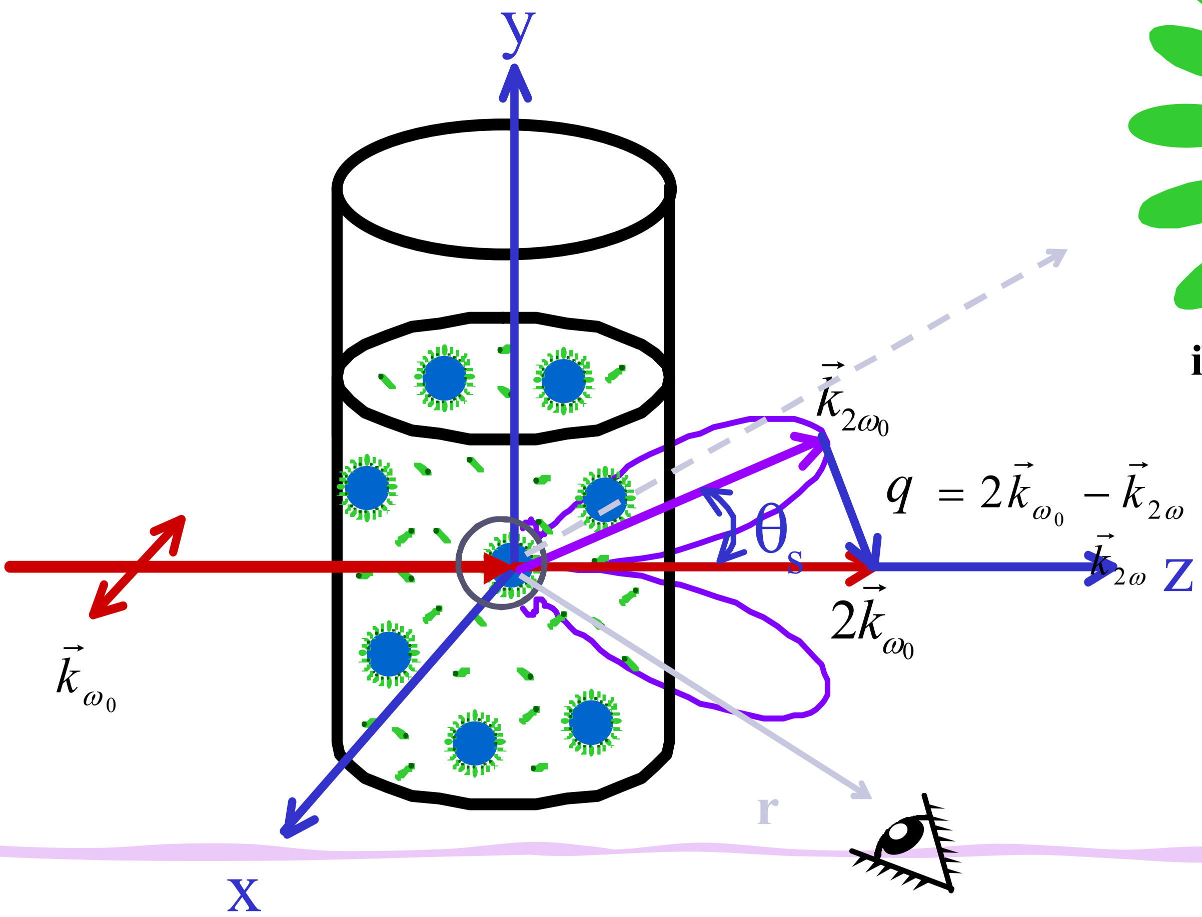
$$q \cdot d = \frac{4\pi n_{2\omega}}{\lambda_{2\omega}} \sin\frac{\theta_s}{2} \cdot d = \pi$$

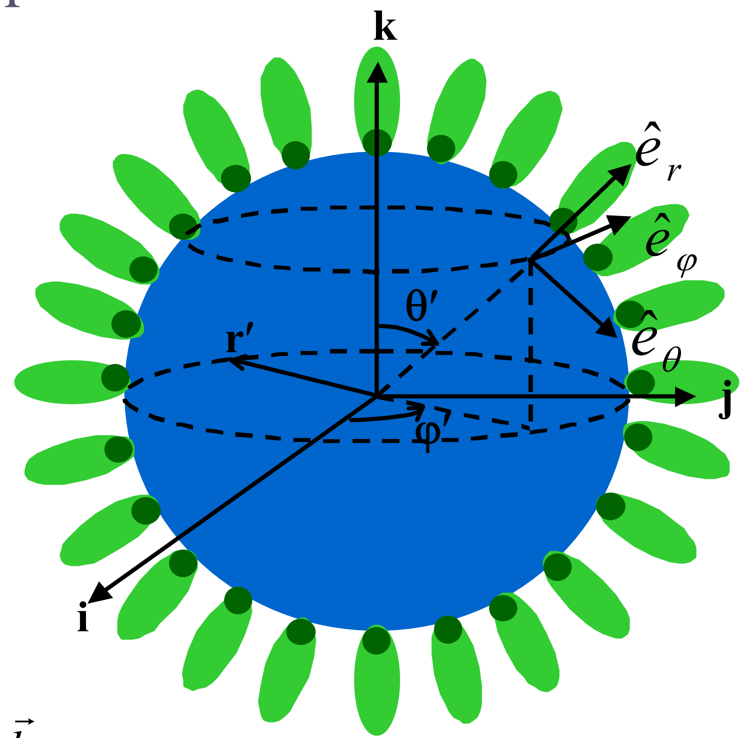
SHG angular distribution in the laboratory frame

$\vec{k}_{\omega_0}, \vec{k}_{2\omega}$: fundamental and SH wave vector

q : scattering vector

θ_s : scattering angle





Nonlinear Rayleigh-Gans-Debye (NLRGD) Theory

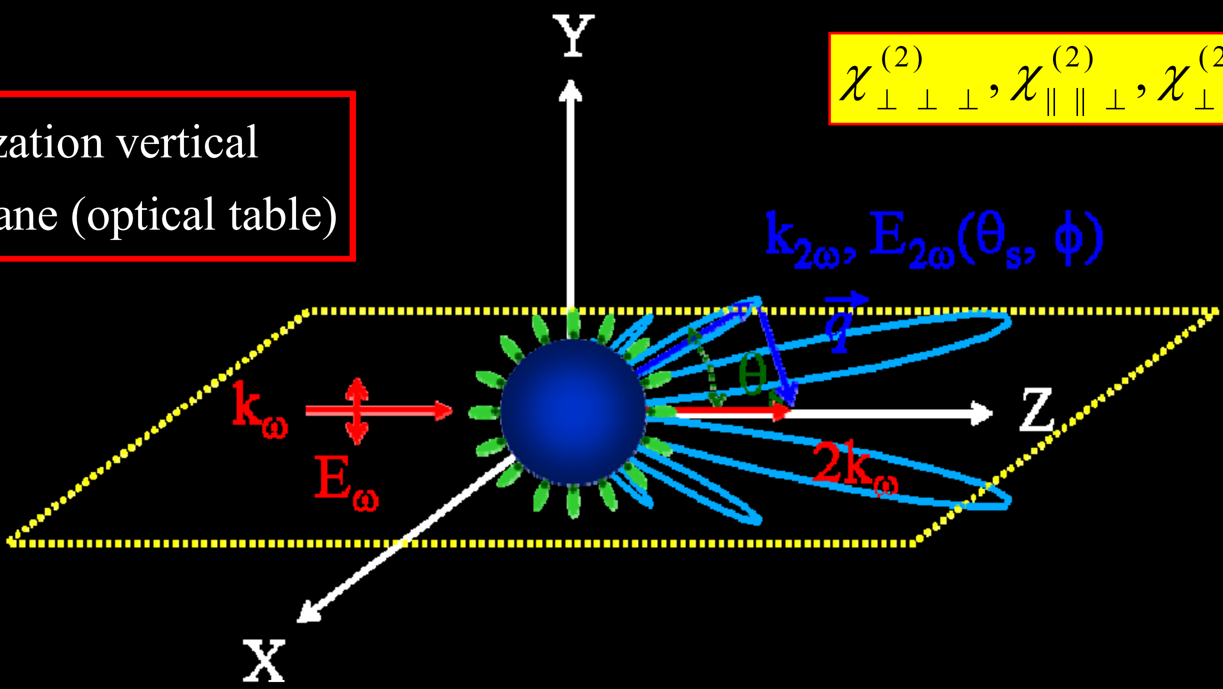
SH Jen and Grazia Gonella

$$E^{sp}_{2\omega}(r) \propto \cos\frac{\theta_s}{2}\left[-F_1(\theta_s)\chi^{(2)}_{\perp\perp\perp} + F_1(\theta_s)\chi^{(2)}_{\parallel\parallel\perp} + \left(2F_2(\theta_s) - F_1(\theta_s)\right)\chi^{(2)}_{\perp\parallel\parallel}\right]\hat{o}_\perp$$

$$E^{pp}_{2\omega}(r) \propto \cos\frac{\theta_s}{2}\left[\left(\sin^2\frac{\theta_s}{2}F_1(\theta_s) + 2\cos^2\frac{\theta_s}{2}F_2(\theta_s)\right)\chi^{(2)}_{\perp\perp\perp} - \left(\left(3\cos^2\frac{\theta_s}{2} - 1\right)F_1(\theta_s) + 2\sin^2\frac{\theta_s}{2}F_2(\theta_s)\right)\chi^{(2)}_{\parallel\parallel\perp} + \left(-\left(\sin^2\frac{\theta_s}{2} + 2\right)F_1(\theta_s) + 2\sin^2\frac{\theta_s}{2}F_2(\theta_s)\right)\chi^{(2)}_{\perp\parallel\parallel}\right]\hat{o}_\perp$$

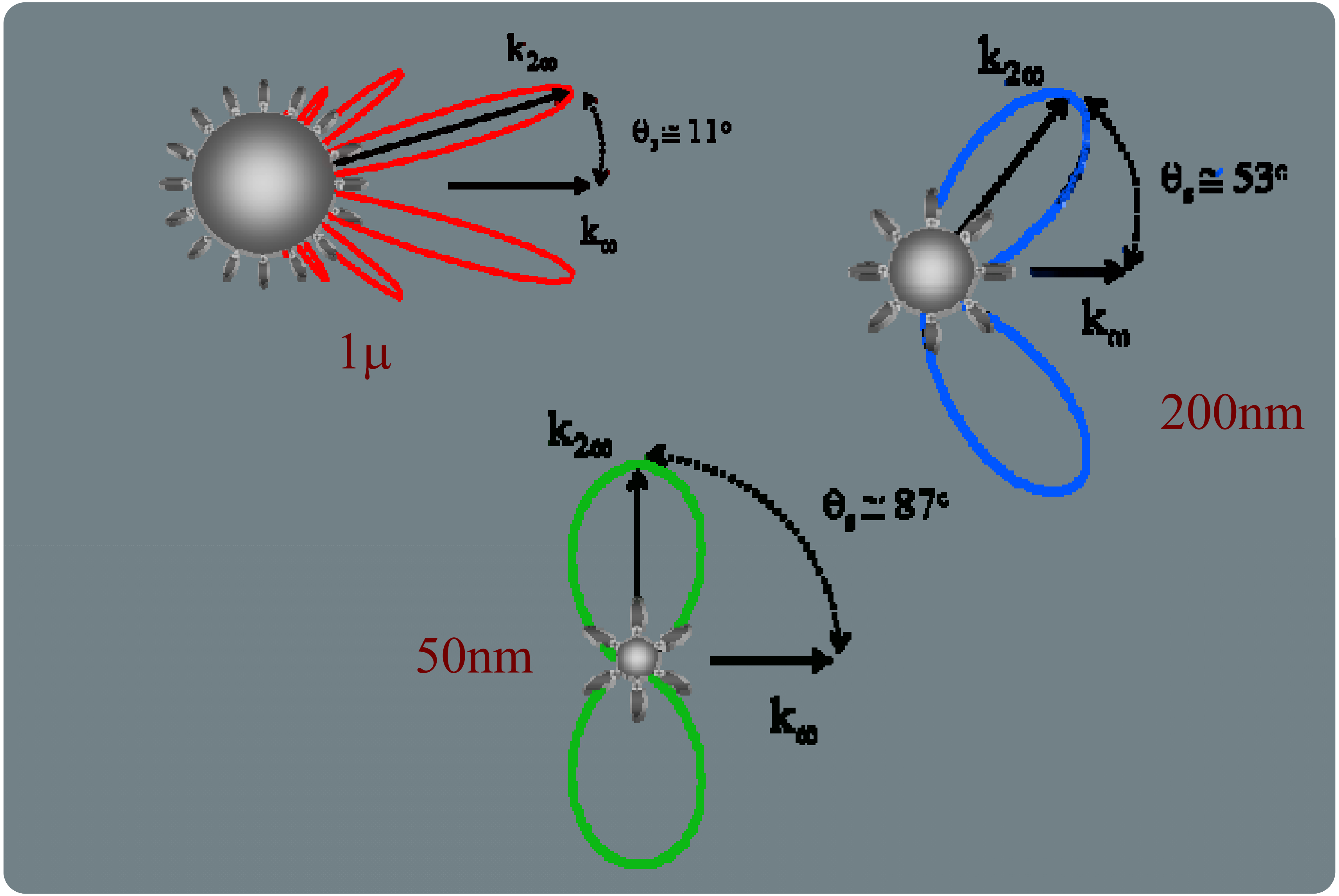
s (p)-polarization: polarization vertical (parallel) to scattering plane (optical table)

$$\chi^{(2)}_{\perp\perp\perp}, \chi^{(2)}_{\parallel\parallel\perp}, \chi^{(2)}_{\perp\parallel\parallel}$$



Malachite Green/Polystyrene in Water

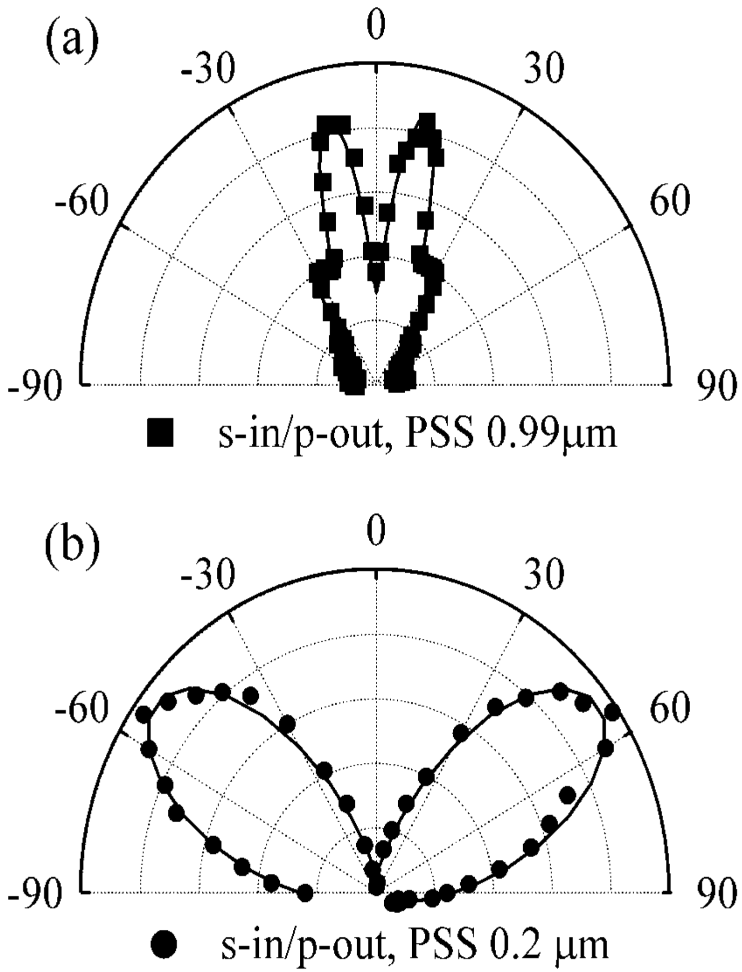
Nonlinear RGD model calculation of angular dependence of SHG scattering



Measurements on 55 and 85 nm also reported in Shan et al. Phys. Rev. A 73, 023819 (2006)

Angle dependence of SHG from colloidal particles

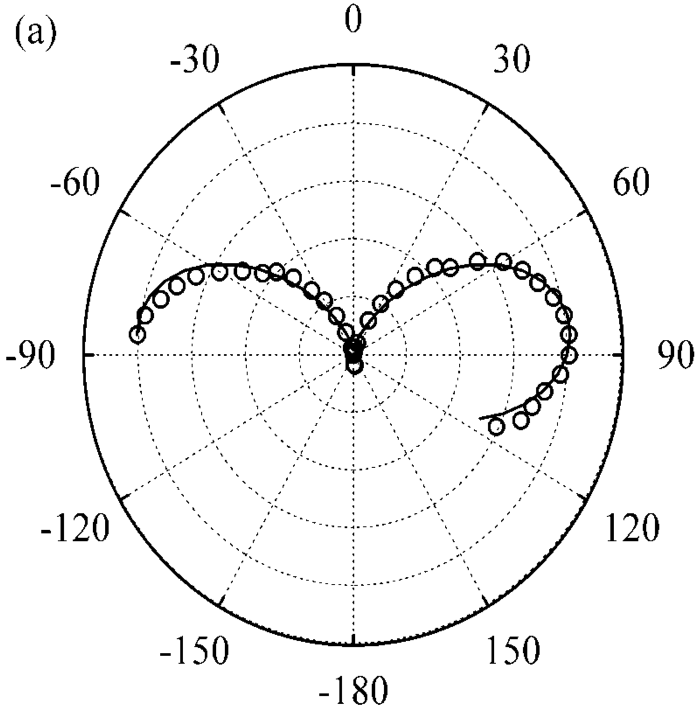
Probe particles of different sizes at selected scattering angles

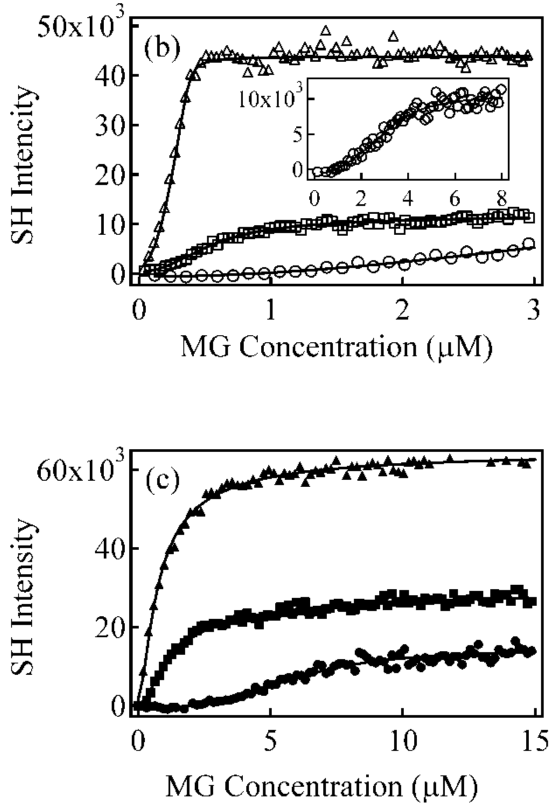


Polystyrene particles (diameter from 50 nm To 1 micron)
The smaller the particle, the larger the SHG scattering angle.

Fundamental light
Propagation direction

Probe adsorption on nanoparticles through Second Harmonic Scattering





50 nm particles SHG at 90°

Adsorption isotherm of nanoparticles
Detected at large scattering angles

SH Jen, *JPCB Lett* 2006

Can we detect SHG from the surface of metallic nanoparticles?

(SHG from flat metal surface can be detected.)

Enormous Hyper-Rayleigh Scattering from Nanocrystalline Gold Particle Suspensions

Fredrick W. Vance, Buford I. Lemon, and Joseph T. Hupp*
Department of Chemistry and Materials Research Center, Northwestern University, Evanston, Illinois 60208

***J. Phys. Chem. B* 1998, *102*, 10091-10093**

The recent emergence of advanced technological applications for colloidal gold suspensions and related particle assemblies and interfaces has created a demand for new chemical and physical techniques with which to characterize them. **For macroscopic samples/interfaces, coherent second harmonic generation (SHG) has proven itself a useful characterization tool** due, at least in part, to metal-based plasmon enhancement. **In an effort to defeat or bypass the size restrictions inherent to SHG, we have utilized a related *incoherent methodology,* hyper-Rayleigh scattering (HRS), to interrogate aqueous colloidal suspensions** of 13 nm diameter gold particles. The nanoscale particles have proven to be remarkably efficient scatterers; when evaluated in terms of the first hyperpolarizability (*â), HRS signals from the gold particles substantially surpass those observable from* the best available molecular chromophores. Moreover, the present experiments indicate that *â is highly sensitive* to colloid aggregation and imply that HRS is an effective tool for the characterization of symmetry-reducing perturbations of nanoscale interfaces.

Theoretical modeling for Metal/semiconductor particles???

Nonlinear Rayleigh-Gans-Debye (RGD) Theory

Conditions of applicability:

- particle refractive index ~ liquid refractive index
- smaller particles (in comparison with wavelength)

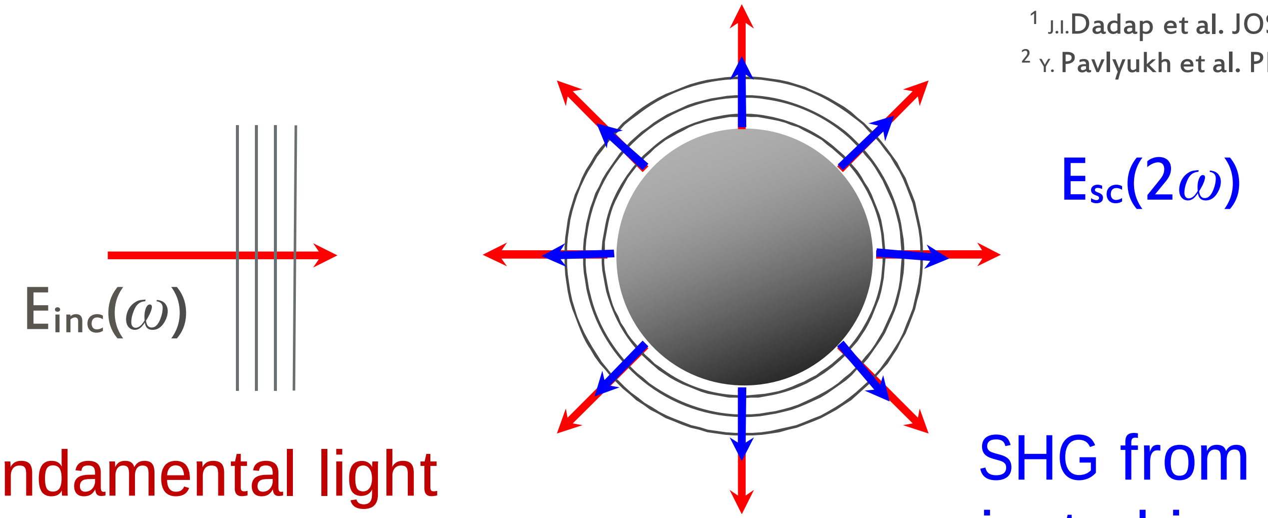
N. Yang, W.E. Angerer and A.G.Yodh PRL 87, 103902 (2001)
S. Roke, W. G. Roeterdink, J. E. G. J. Wijnhoven, A. V. Petukhov, A. W. Kleyn, and M. Bonn, PRL 91, 258302 (2003)
S.-H. Jen, Ph. D. Thesis, University of Pennsylvania 2006

PROBLEMs:
Not good for
Larger (micron) particles , and
Metal /Semi-Conductor particles

Solution: the more general Mie Scattering Theory

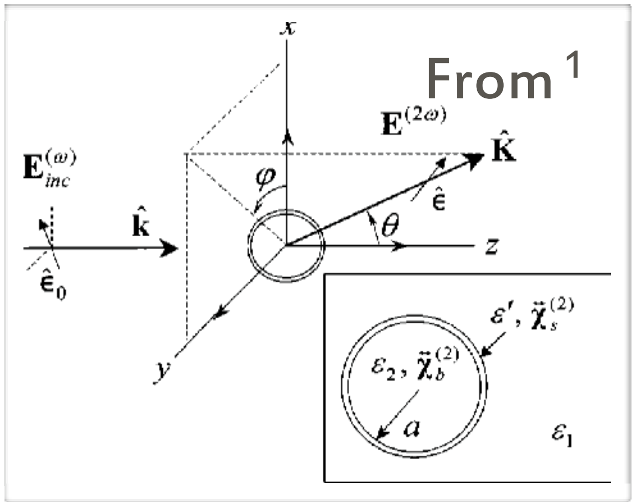
Nonlinear MIE Theory[1,2]

[1] J.I.Dadap et al. JOSA B 21(2004), 1328
[2] Y. Pavlyukh et al. PRB 70(2004), 245434



Fundamental light
In plane waves

SHG from surface
projected in plane waves



Nonlinear Surface Source:

isotropic symmetry with a mirror plane perpendicular to the surface

$$|\Sigma \longrightarrow \quad |_{\infty\infty\infty\infty}, \quad |_{\infty||||}, \quad |_{||\infty||}$$

Expression for the SH Scattered field for ANY particle size[1]

Grazia Gonella, *PRB* 2011

$$\vec{E}^{sc}(2\omega) = E_0 \sum_{l=1}^{\infty} \sum_{m_{even}=-2}^{2} \left(A_M^{sc}(l,m) h_l(K_1 r) \mathbf{X}_{lm} + \frac{A_E^{sc}(l,m)}{K_1} \left(i \frac{\sqrt{l(l+1)}}{r} h_l(K_1 r) Y_{lm} \hat{\mathbf{r}} + \frac{1}{r}\frac{d}{dr}(r h_l(K_1 r)) \hat{\mathbf{r}} \times \mathbf{X}_{lm} \right) \right)$$

l → multipole contribution

0

Far Zone:

$$h(K_1 r) = (-i)^{l+1} \frac{e^{iK_1 r}}{K_1 r}$$

$$\frac{1}{r}\frac{\partial}{\partial r}(r h_l(K_1 r)) = -(-i)^{l} \frac{e^{iK_1 r}}{r}$$

[1] G. Gonella and H.-L. Dai (in preparation)

$$A_M^{2\omega,sc}(l,m) = \frac{4\pi \mu_1(2\omega)\mu_2(2\omega) K^2 r\, \chi_{\perp\perp\parallel} b^{lm}_{\parallel\perp\parallel,M}\, j(K_2 r)}{\mu_1(2\omega) h(K_1 r)\frac{d}{dr}(r j(K_2 r)) - \mu_2(2\omega) j(K_2 r) \frac{d}{dr}(r h(K_1 r))}\Bigg|_{r=a}$$

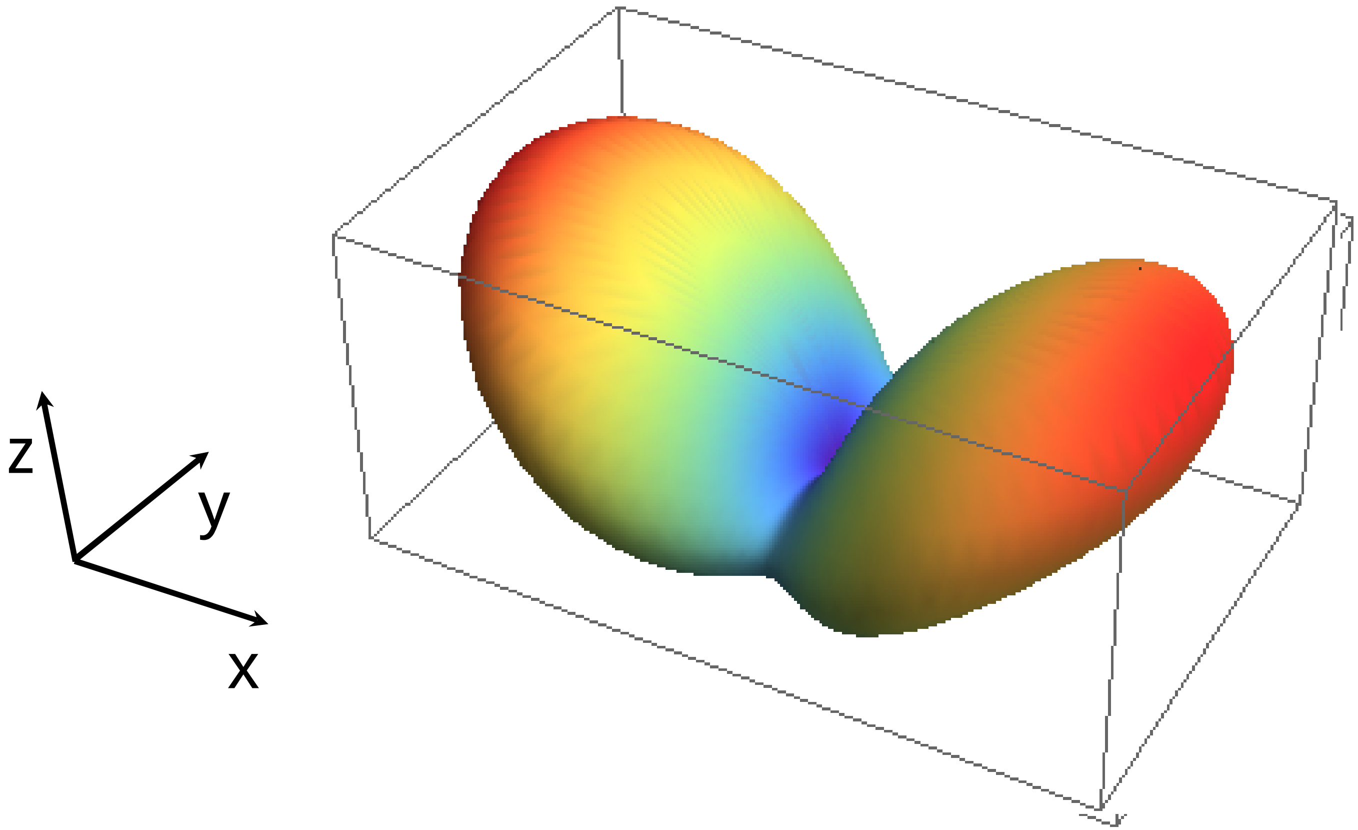
$$A_E^{2\omega,sc}(l,m) = 4\pi K_1 \frac{i\frac{\kappa_2(2\omega)}{\kappa'(2\omega)}\sqrt{l(l+1)}\, j(K_2 r)(\chi_{\perp\perp\perp} b^{lm}_{\perp\perp\perp} + \chi_{\perp\parallel\parallel} b^{lm}_{\perp\parallel\parallel}) - \frac{d}{dr}(r j(K_2 r))\chi_{\parallel\perp\parallel} b^{lm}_{\parallel\perp\parallel,E}}{\varepsilon_2(2\omega) j(K_2 r)\frac{d}{dr}(r h(K_1 r)) - \varepsilon_1(2\omega) h(K_1 r)\frac{d}{dr}(r j(K_2 r))}\Bigg|_{r=a}$$

SH Mie Scattering from A SINGLE PARTICLE

- ✓ In the case of DILUTE REGIME: mean distance between particles is greater than the coherence length so the scattering events simply add up.
- ✓ In the HIGH-DENSITY REGIME: coherent interference among the particles becomes important.

3D view of SHG calculated by MIE theory for Gold Nanoparticles (50 nm diameter)

SHG from metallic nanoparticles should be detected at large scattering angles

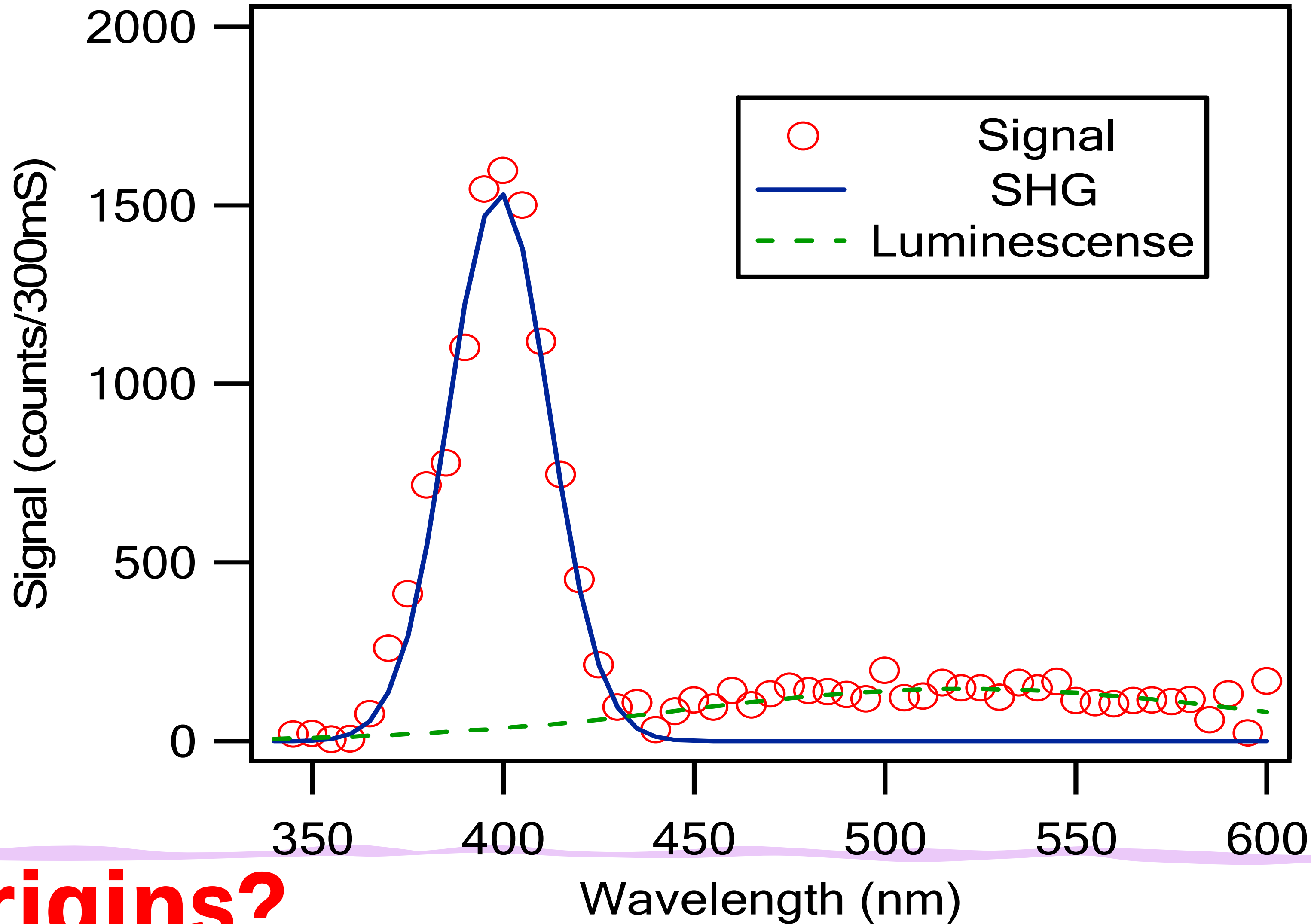


z: propagation direction

xz: (horizontal) scattering plane

SHG from 80 nm diameter Ag particle (citrate stabilized in water)

Wei Gan, Min Zhang

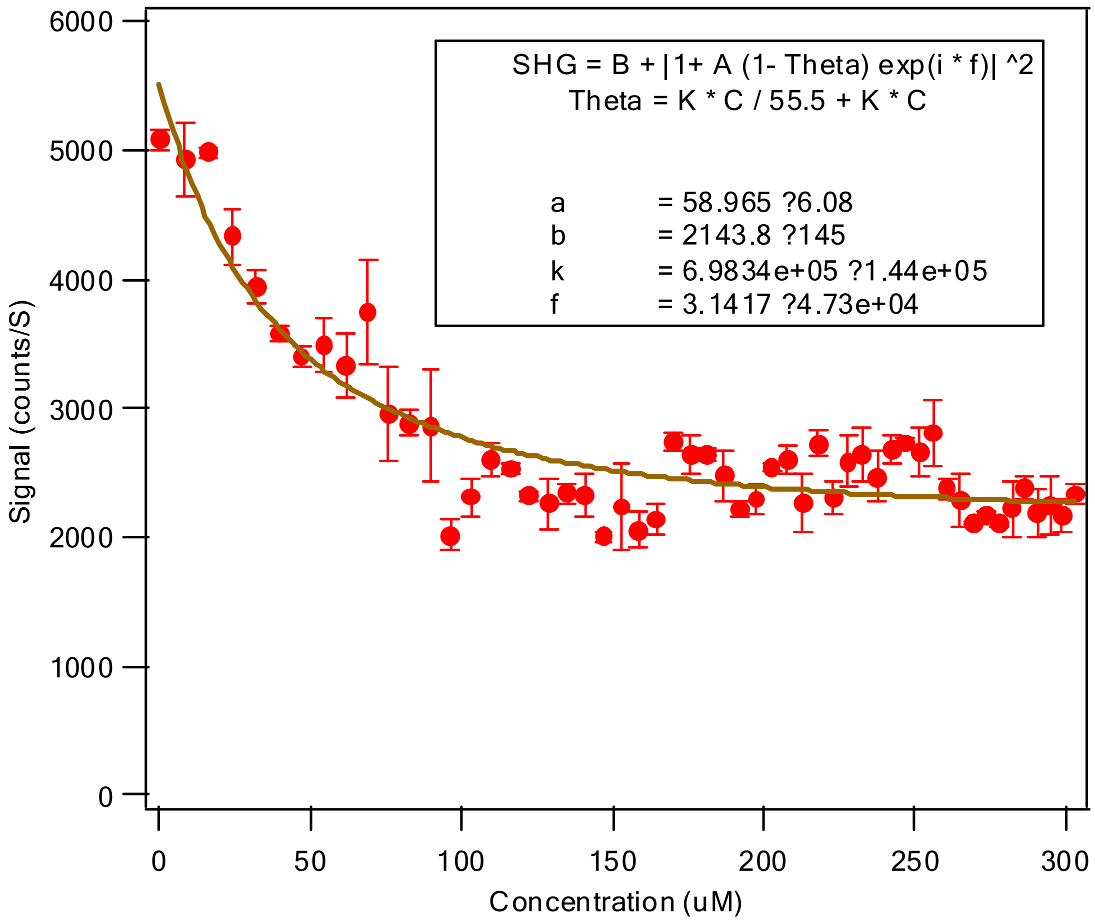


Origins?

SHG or Hyper-Rayleigh Scattering? (a moot question)

SHG – from surface and bulk?

Surface SHG decreases as thiol bonds to Ag surface



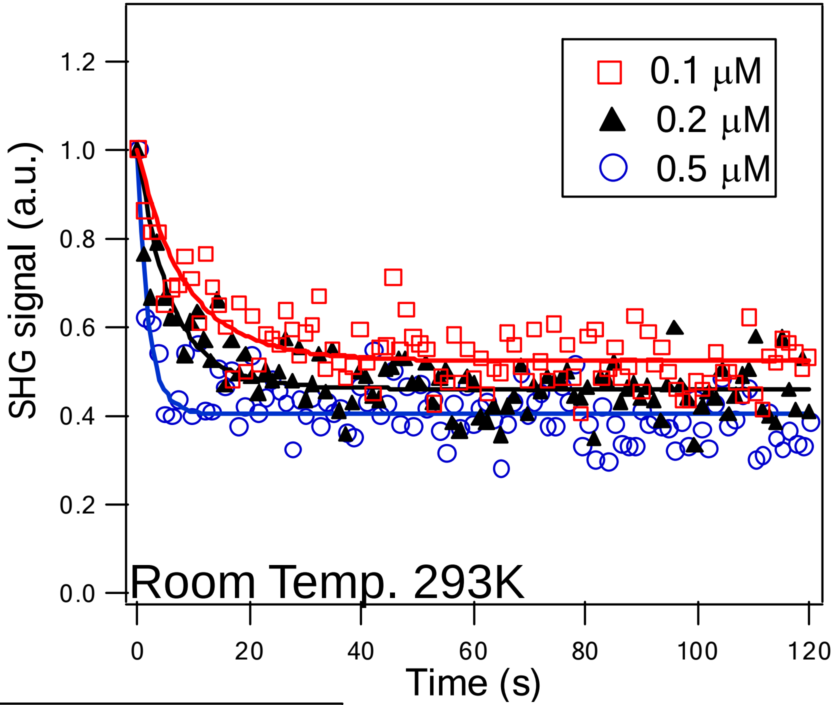
Surface SHG

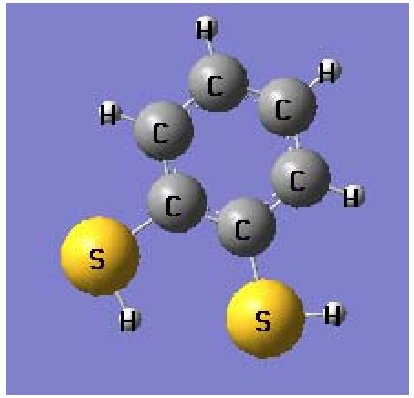
Langmuir Model

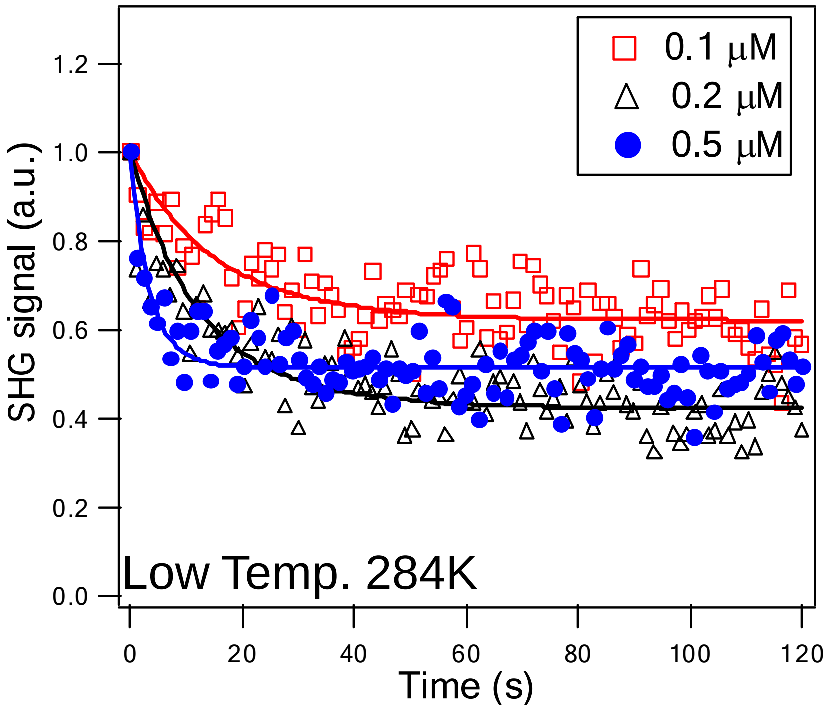
Bulk SHG

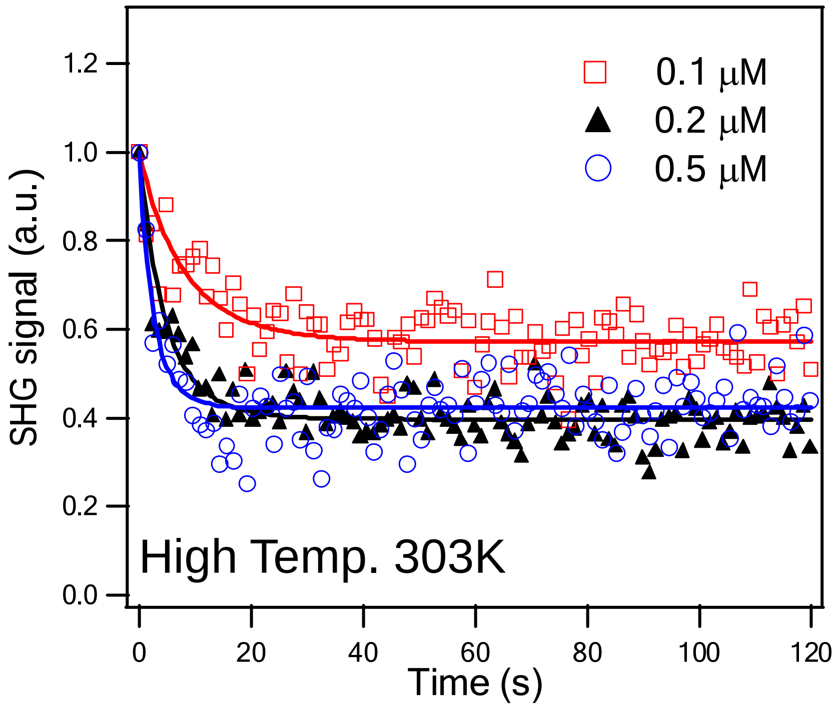
Wei Gan, *JCP* 2011

SHG Quenching At three temperatures

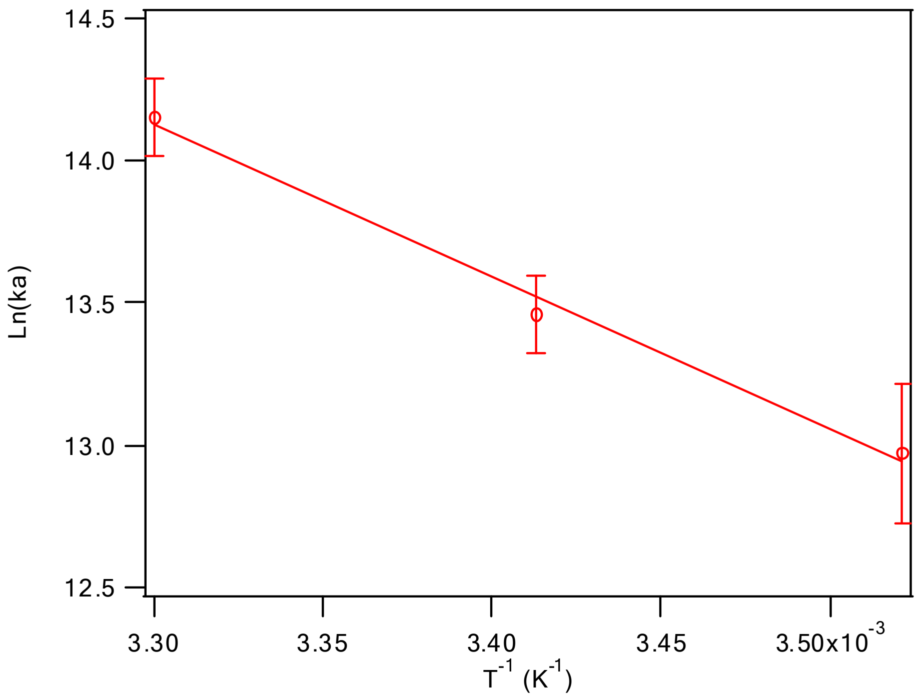








Temperature dependence of Thiol reactions at Ag particle surface



$$k_a = A\exp(-Ea/RT)$$

$$\ln(k_a) = \ln(A) - Ea/RT$$

$$y = \ln(A) - \frac{Ea}{R}x$$

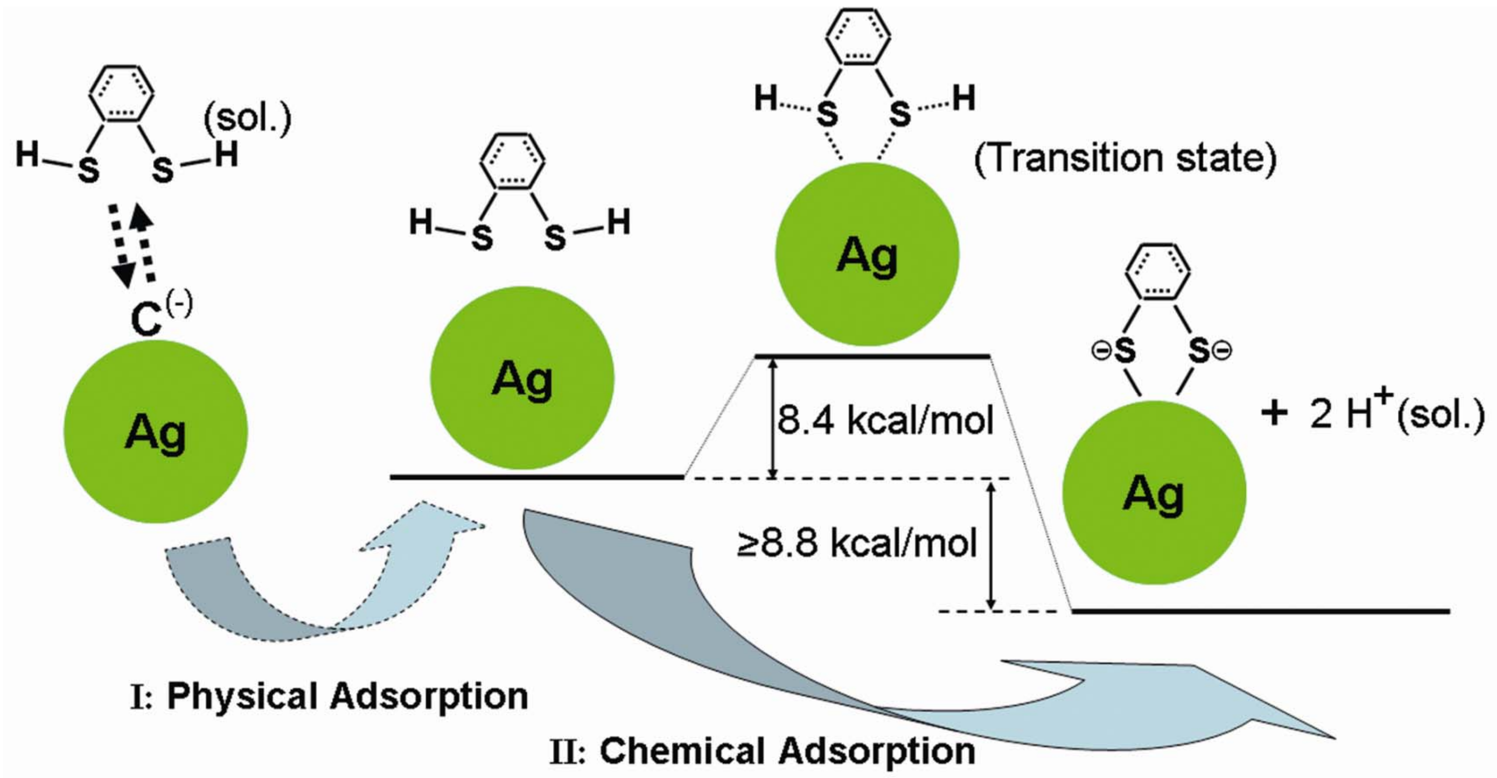
Ea= 8.4 Kcal/mole

Thiol reactions at Ag particle surface are activated processes!!!

Why???

Wei Gan, *Ang Chem Int Ed* 2011

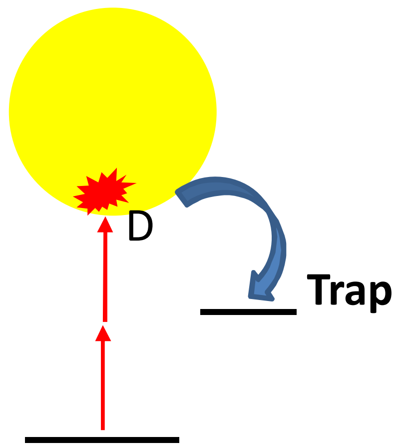
Origin of the activation energy – transition state?



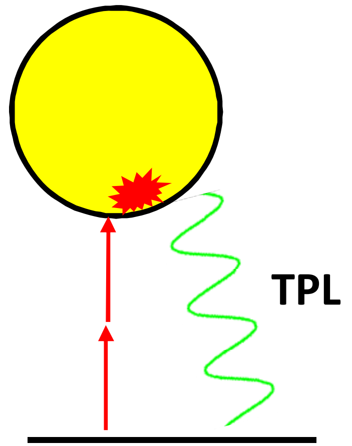
Dramatic increase of Luminescence efficiency of metallic nanoparticles through surface modification

- While thiol adsorption onto particle surface reduces the surface polarizability and thus reduces SHG, it dramatically increases the luminescence [see publications soon]

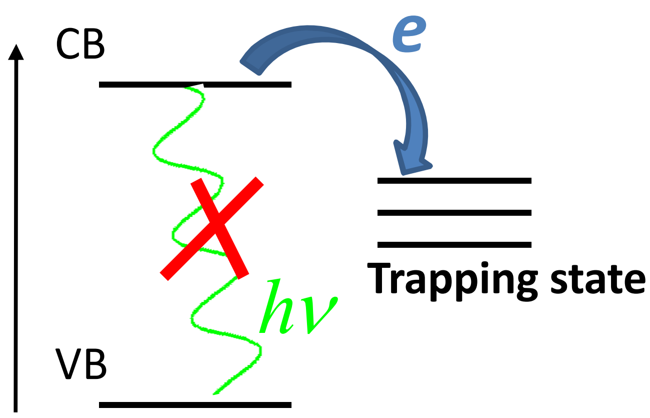
Premises: Surface defects quenches luminescence, and S-Ag bonding anneals surface defects

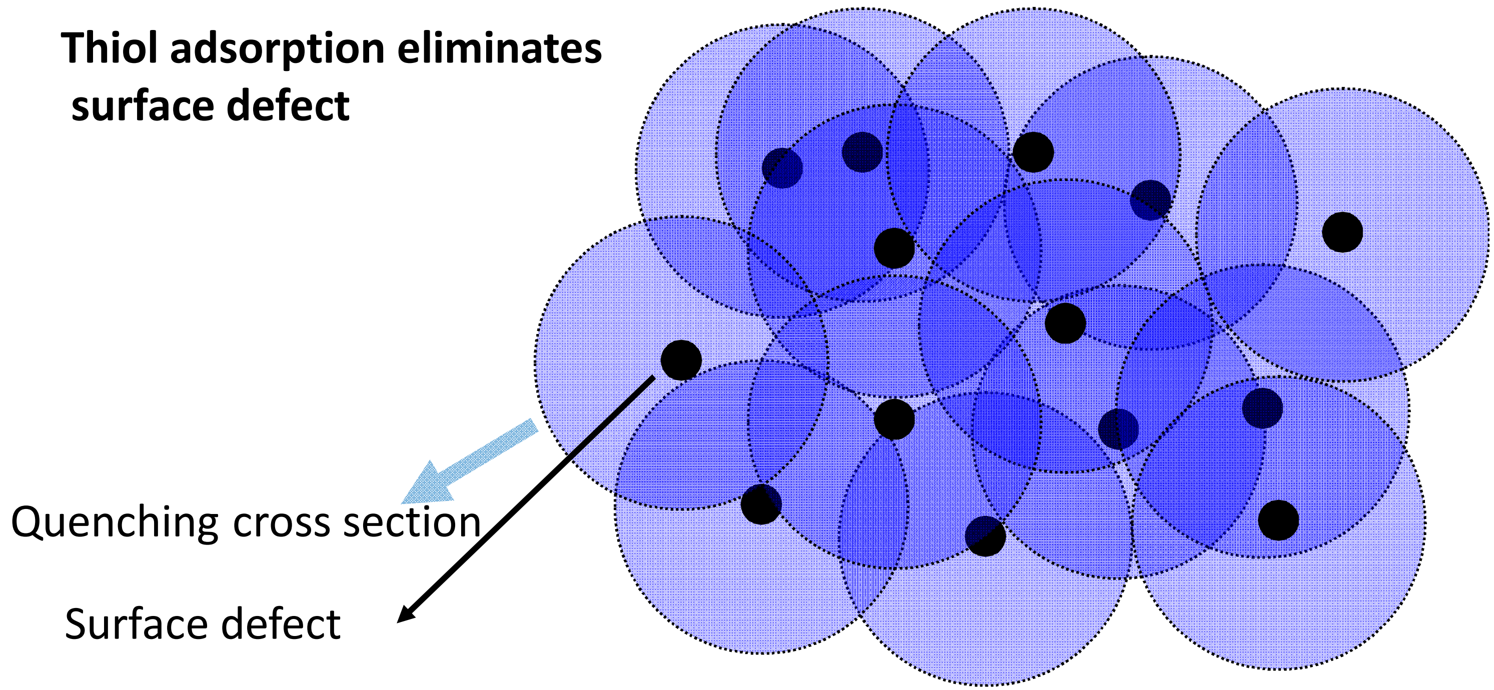


Surface defect traps local excitation



Thiol adsorption eliminates surface defect





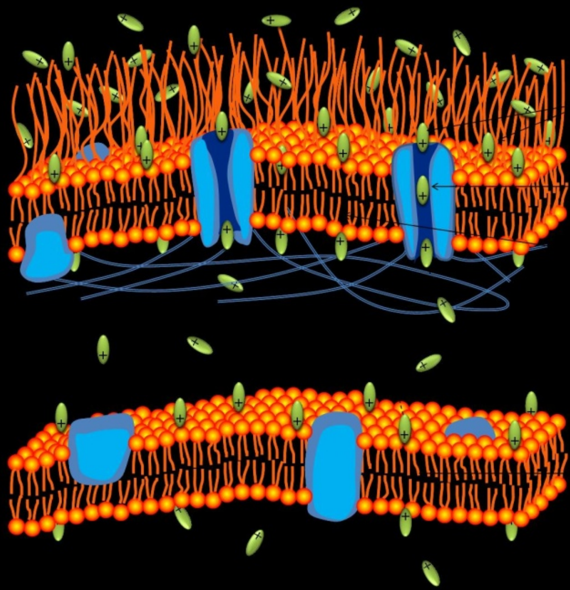
REF: 1:Jiang Z J, Leppert V, Kelley D F, *J. phys. Chem. C* 2009, 113, 19161
2: Fu H X, Zunger A, *Phys. Rev. B* 1997, 56, 1496

Nonlinear light scattering from Biological cells

- SHG can be generated from molecules adsorbed at the membrane of biological cells
- and be used to probe
- membrane-molecule interactions

Molecular Adsorption and Transport at Cell Membrane

by Second Harmonic Generation (SHG)

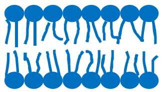


Jia Zeng
Heather Eckenrode

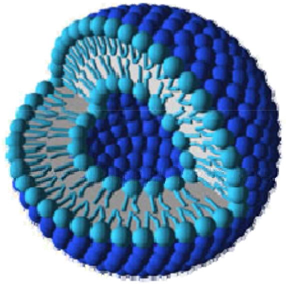
Motivation:
-- Membrane Transport Studies of Hydrophobic Molecules

- Time-resolved Techniques

Simple Model Membrane Systems:



Electrical relaxation studies



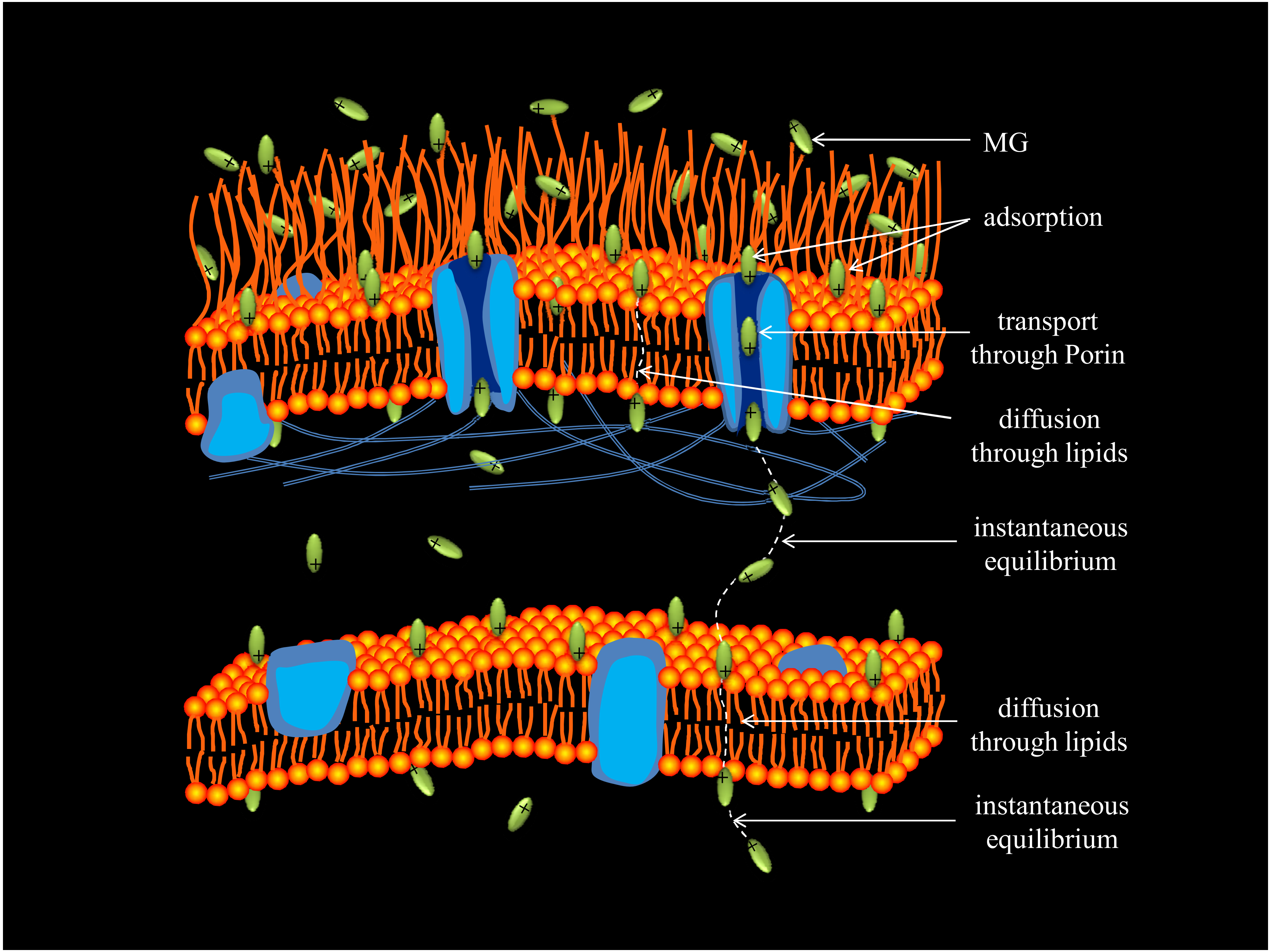
Fluorescence stop-flow technique

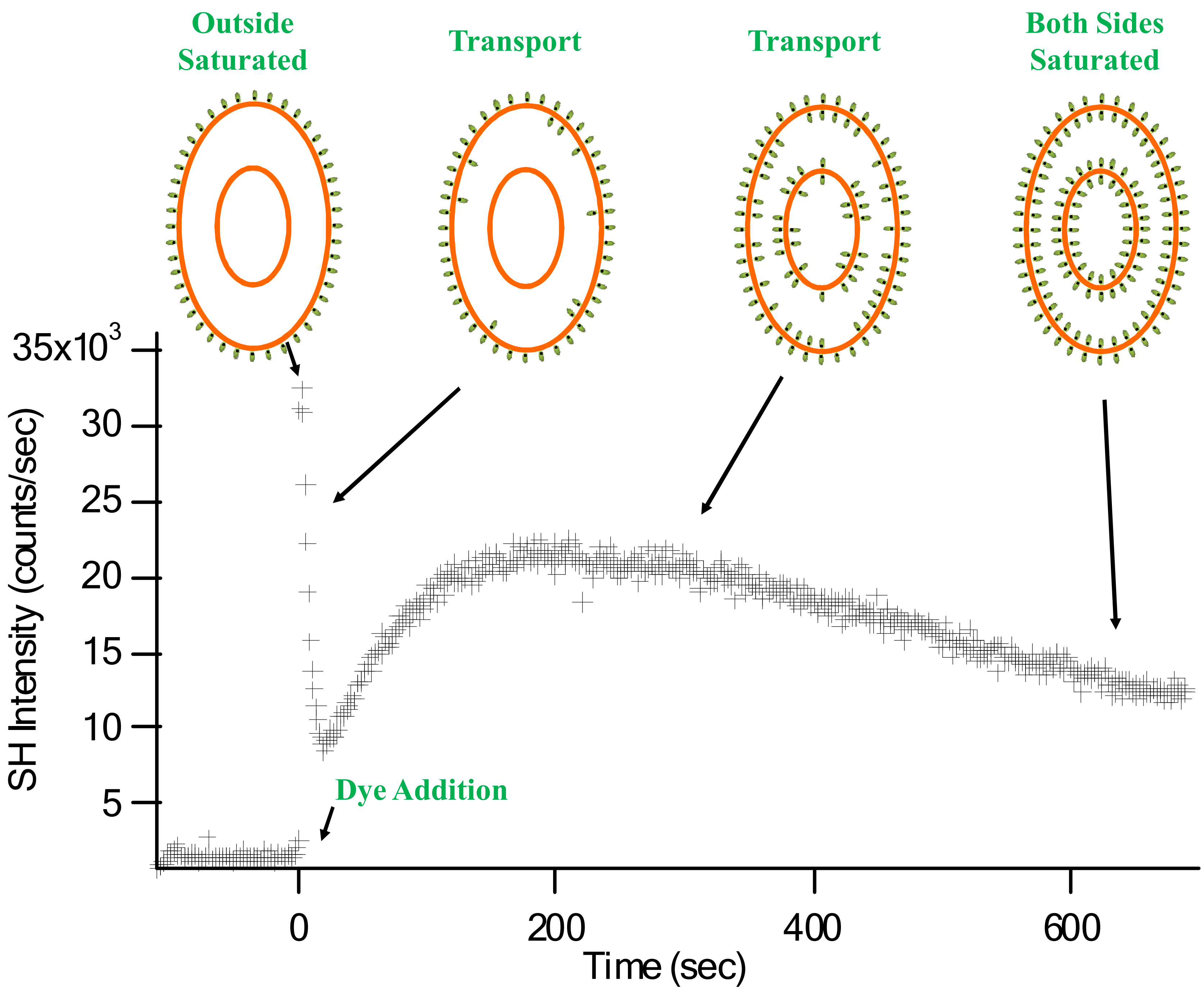
Real Time Real Cell --SHG

- Intact Cells → Steady State Level

Beta-Lactam antibiotics transport through the bacterial outer membrane by Enzyme Assay Method

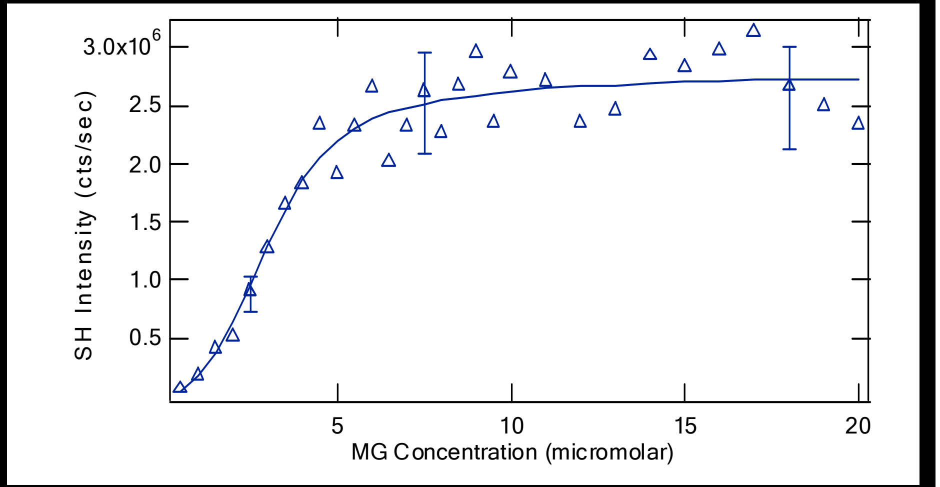
W.Zimmermann, A. Rosselet *Antimicrobial Agents and Chemotherapy*, 368-372 (1977)


MG
adsorption
transport through Porin
diffusion through lipids
instantaneous equilibrium
diffusion through lipids
instantaneous equilibrium

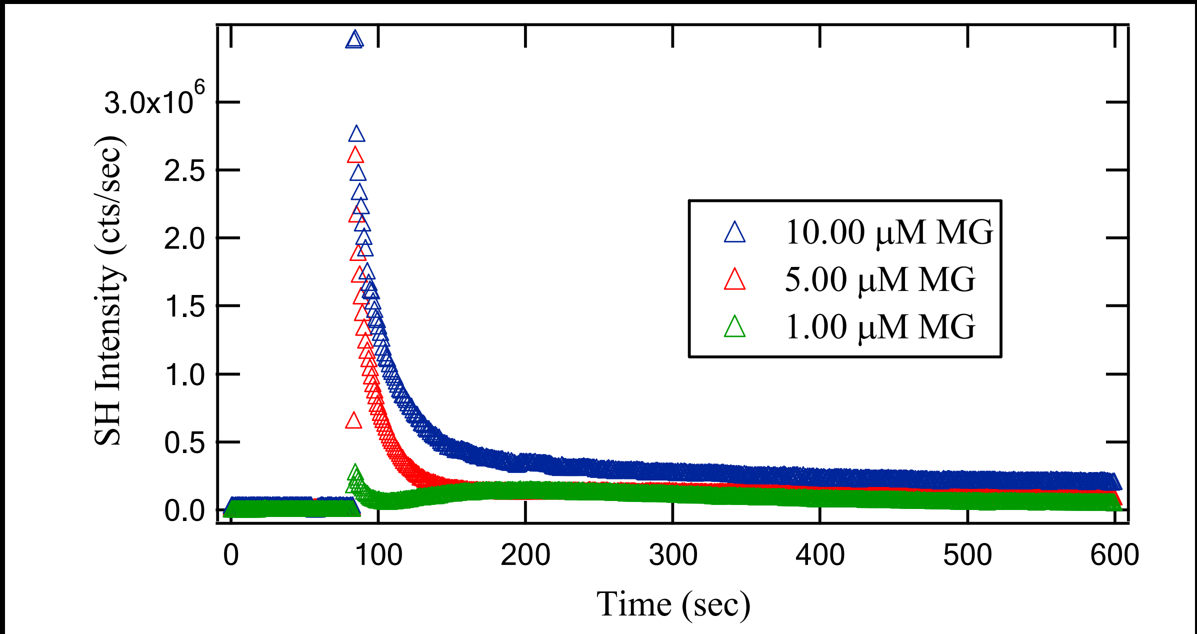


Outside Saturated
Transport
Transport
Both Sides Saturated
35x10^3
30
25
20
15
10
5
SH Intensity (counts/sec)
Dye Addition
0
200
400
600
Time (sec)

Adsorption Isotherm



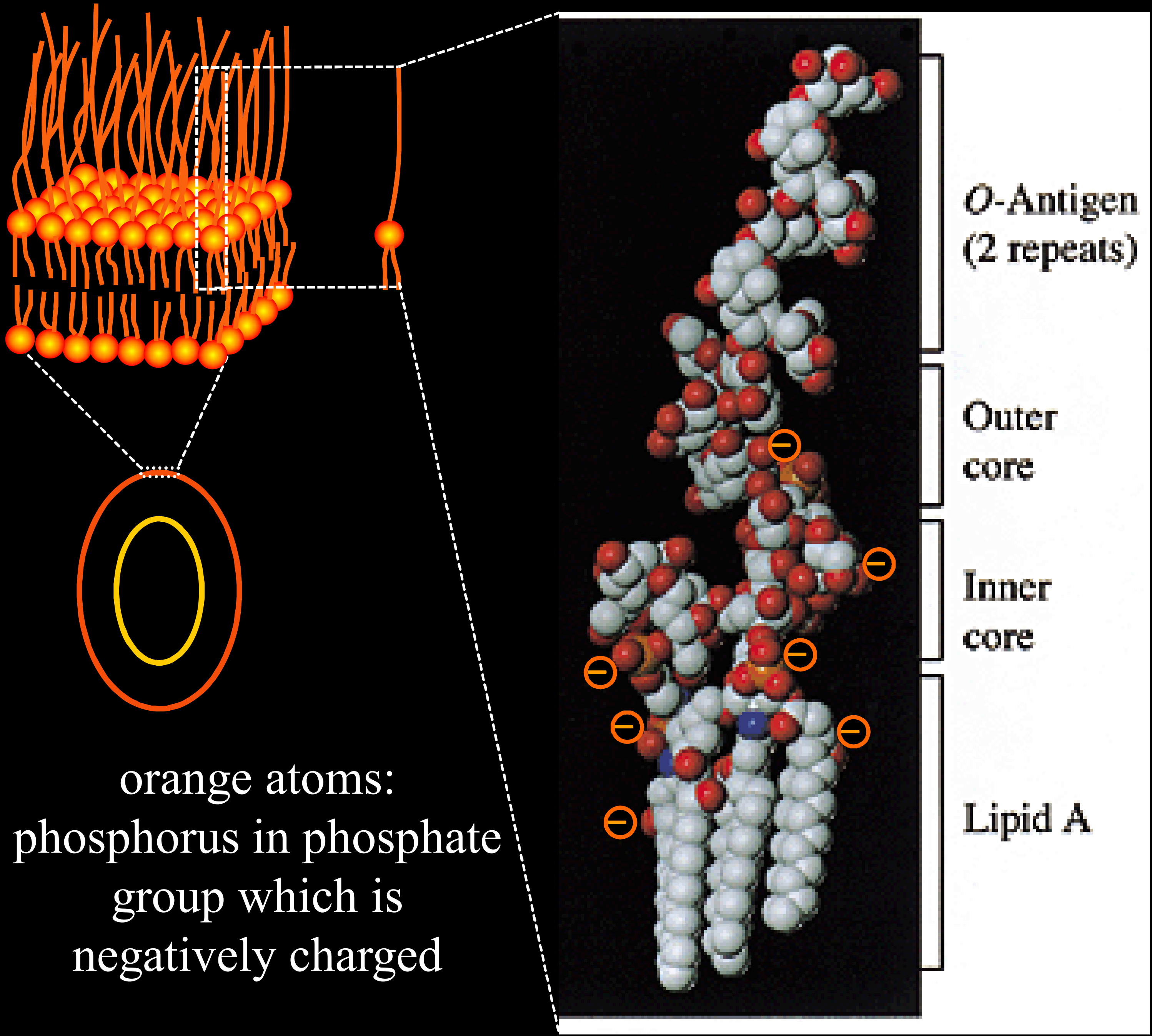
Plot peak values



Transport time profiles at different MG concentrations

$$N_{max} = (3.7 \pm 0.6)\mu M = (7.4 \pm 1.2) \times 10^7 cell^{-1} = (1.2 \pm 0.2) \times 10^7 \mu m^{-2} = 1/8.3 Å^2$$

$$\Delta G = -RT \ln K = -(13.6 \pm 0.4) kcal \cdot mol^{-1}$$

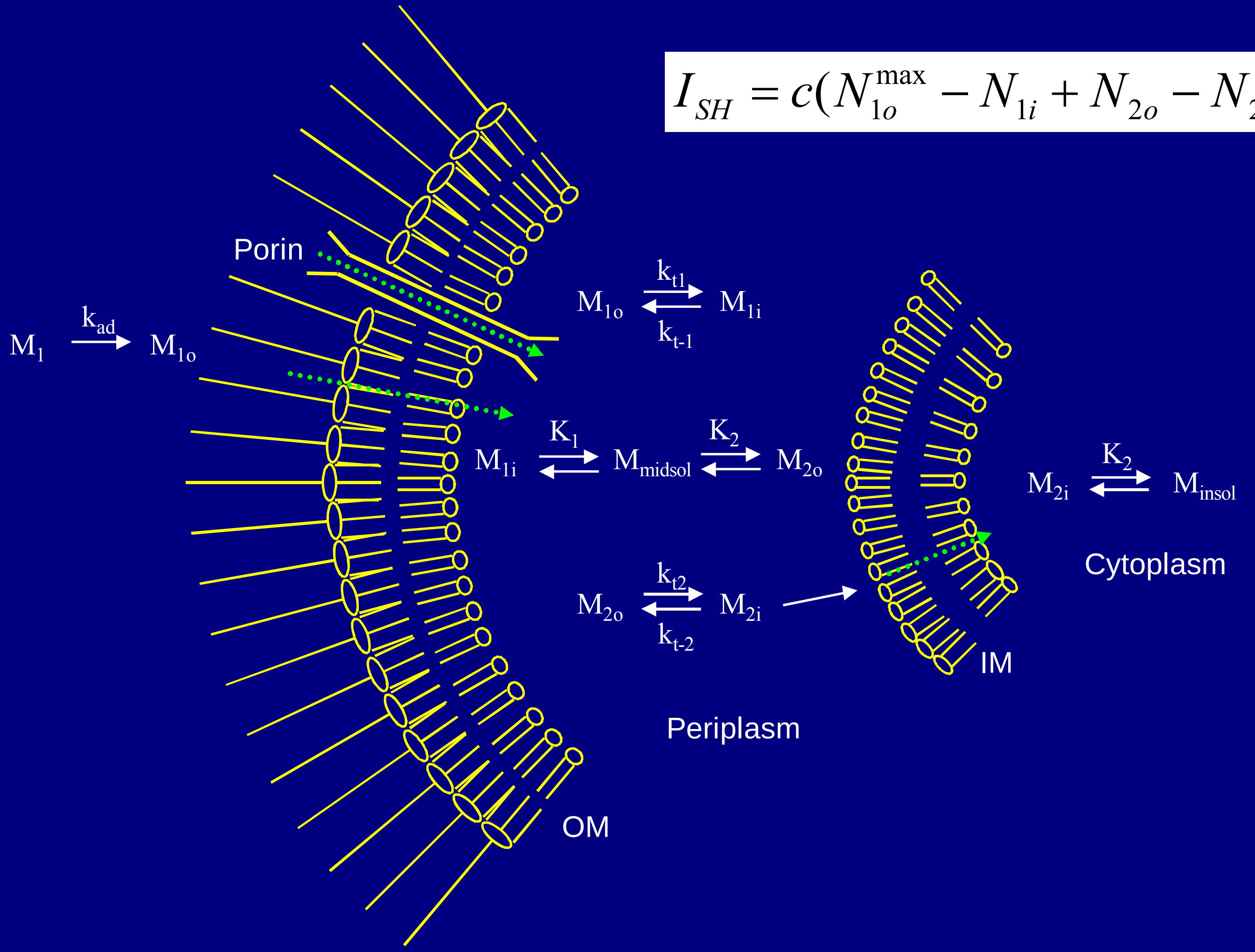


orange atoms:
phosphorus in phosphate
group which is
negatively charged

N.A. Amro, et al., *Langmuir* 16: 2789-2796 (2000)

Kinetic Model Based on the Double Membrane Structure of *E.coli*:

$$I_{SH} = c(N_{1o}^{max} - N_{1i} + N_{2o} - N_{2i})^2$$



$$d[Transp.1] = k_{t1} \cdot (N_{1o} - N_{1i}) \cdot dt$$

$$K_1 = \frac{55.5 V_{mid} N_A (N_{1i} + dN_{1i})}{(N_{mid,sol} + dN_{mid,sol})(N_{1i}^{const.} - N_{1i} - dN_{1i})}$$

$$K_2 = \frac{55.5 V_{mid} N_A (N_{2o} + dN_{2o})}{(N_{mid,sol} + dN_{mid,sol})(N_{2o}^{const.} - N_{2o} - dN_{2o})}$$

$$I_{SH} = c(N_{1o}^{const.} - N_{1i} + N_{2o} - N_{2i})^2$$

k_{t1} : transport rate constant of OM

K_1 K_2 : adsorption equilibrium constants

55.5 : water molarity

V_{mid} : volume in between the two membranes

N_{1i} : adsorbed to the inner surface of the OM

N_{2o} : adsorbed to the outer surface of the IM

$N_{mid,sol}$: dissolved in the periplasmic space

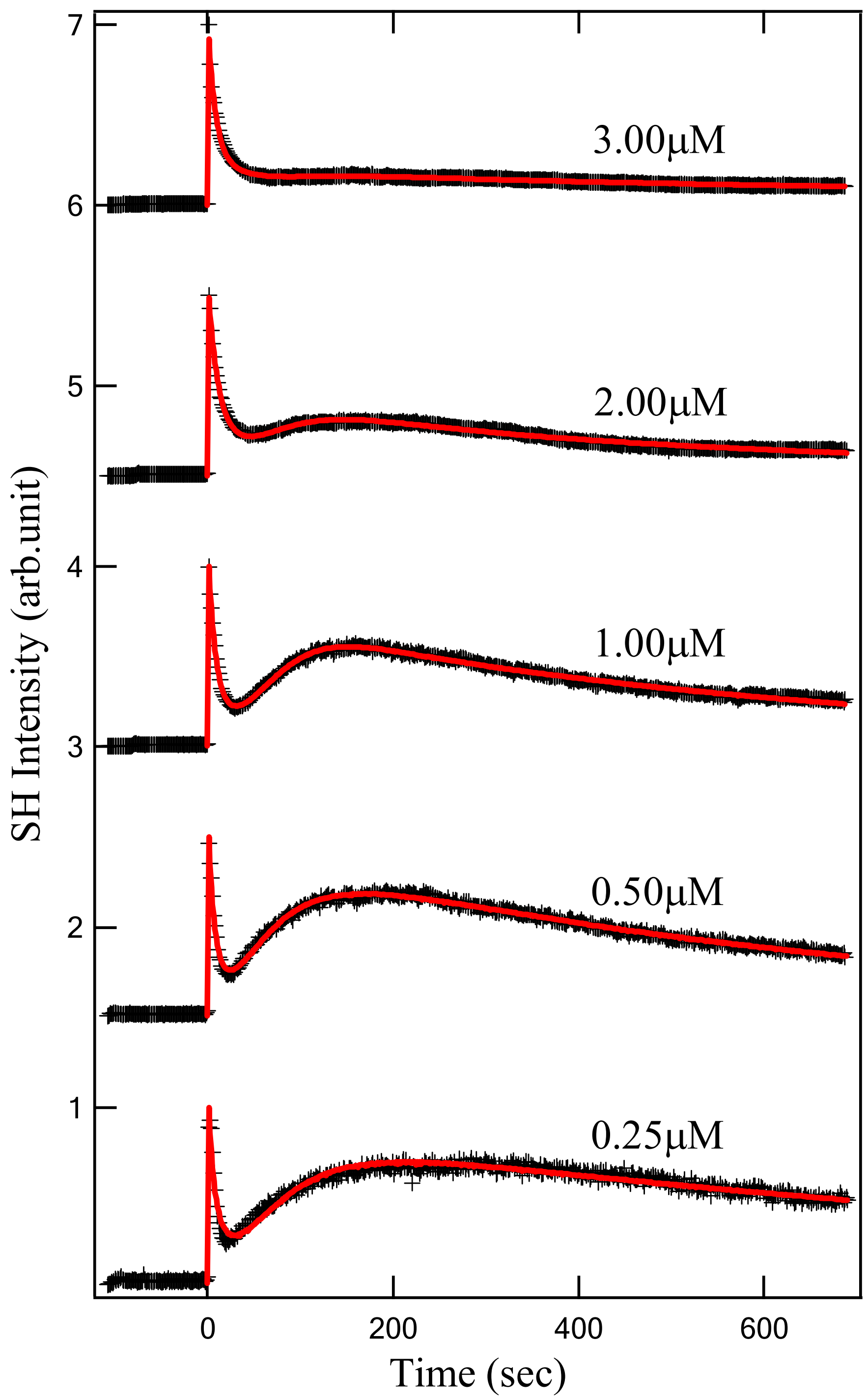
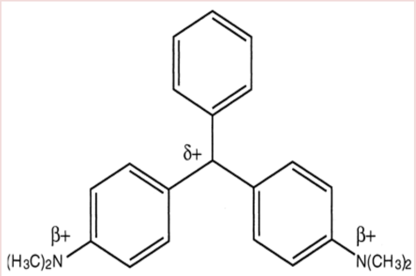
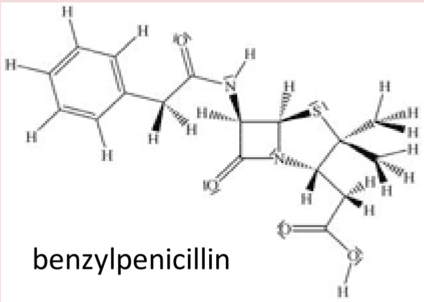


Table. Outer Membrane Characterizations

Parameters \ MG conc. μM	0.25	0.50	1.00	2.00	3.00
$k_{t1}(s^{-1})$	$(6.0\pm0.1)\times10^{-2}$	$(7.0\pm0.6)\times10^{-2}$	**$(6.0\pm0.2)\times10^{-2}$**	$(4.0\pm0.4)\times10^{-2}$	$(4.0\pm0.3)\times10^{-2}$
$K_1(M^{-1})$	$(8.0\pm0.8)\times10^{4}$	$(8.0\pm0.5)\times10^{4}$	$(8.0\pm0.6)\times10^{4}$	$(7.8\pm0.2)\times10^{4}$	$(8.0\pm1.0)\times10^{4}$
$N_{1out}^{\max}(cell^{-1})$	$(4.1\pm0.3)\times10^{6}$	$(7.1\pm0.2)\times10^{6}$	$(1.2\pm0.1)\times10^{7}$	$(1.6\pm0.2)\times10^{7}$	$(2.2\pm0.1)\times10^{7}$
$N_{1in}^{\max}(cell^{-1})$	$(3.2\pm0.1)\times10^{6}$	$(6.1\pm0.2)\times10^{6}$	$(1.1\pm0.1)\times10^{7}$	$(1.3\pm0.1)\times10^{7}$	$(1.7\pm0.1)\times10^{7}$



$(5.4\pm1.6)\times10^{-2}\ s^{-1}$



$1.9\times10^{-4}\ s^{-1}$

W.Zimmermann, A. Rosselet . *Antimicrobial Agents and Chemotherapy*, 1977

Hydrophobic ion vs hydrophobic molecule

Relative Characterizations of OM and IM:

	N_{max} ($cell^{-1}$)	N_{max} (μm^{-2})	$-\Delta G$ (kcal/mol)	k_t (s^{-1})
***E.coli* OM**	$(7.4\pm1.2)\times10^{7}$	$(1.2\pm0.2)\times10^{7}$	13.6 ± 0.4	$(5.4\pm1.6)\times10^{-2}$
***E.coli* IM**	$\sim10^{6}$ to $\sim10^{7}$	$\sim10^{5}$ to $\sim10^{6}$	5.1 ± 0.7	$(5.7\pm0.8)\times10^{-3}$
Liposome	$(2.8\pm0.2)\times10^{5}$	$(1.9\pm0.1)\times10^{6}$	8.6 ± 0.2	9.5×10^{-3}

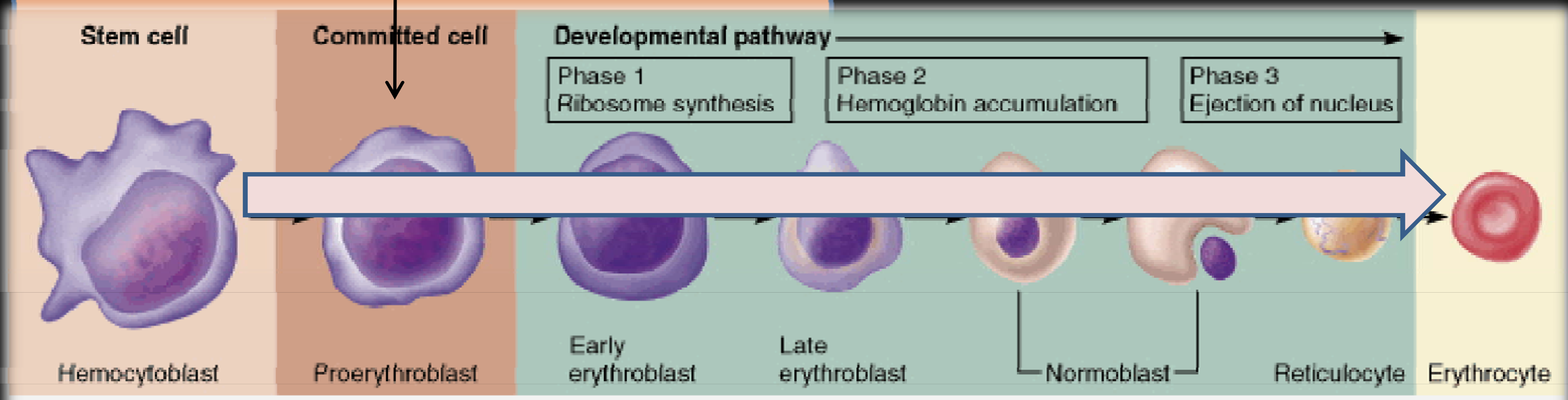
$k_{t1} > k_{t2}$ simple diffusion + ion channel

$-\Delta G_{IM} < -\Delta G_{LIP} < -\Delta G_{OM}$ ionic strength effect

MEL Cell: precursor stage

- 7~12 μm in diameter (100 ~500 μm^3)
- Spherical (NOT disc)
- NOT red yet

- ❑ a gradual reduction in cell size (about ten times)
- ❑ the progressive degeneration of the cell's nucleus which is eventually extruded from the cell
- ❑ the gradual loss of cytoplasmic organelles
- ❑ the gradual appearance of haemoglobin and disappearance of ribonucleic acid (RNA) in the cell



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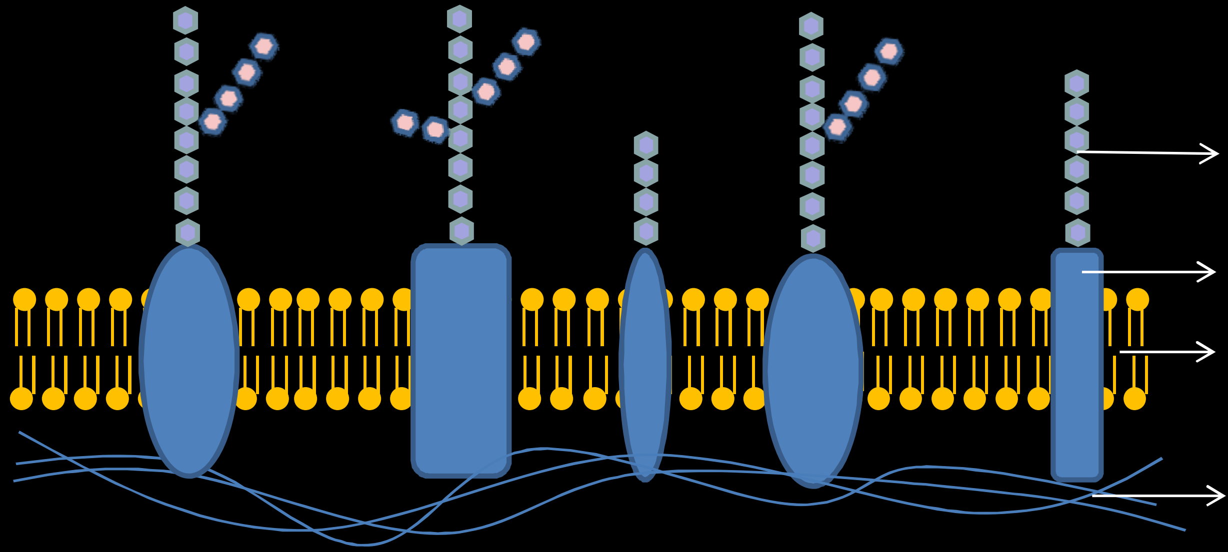
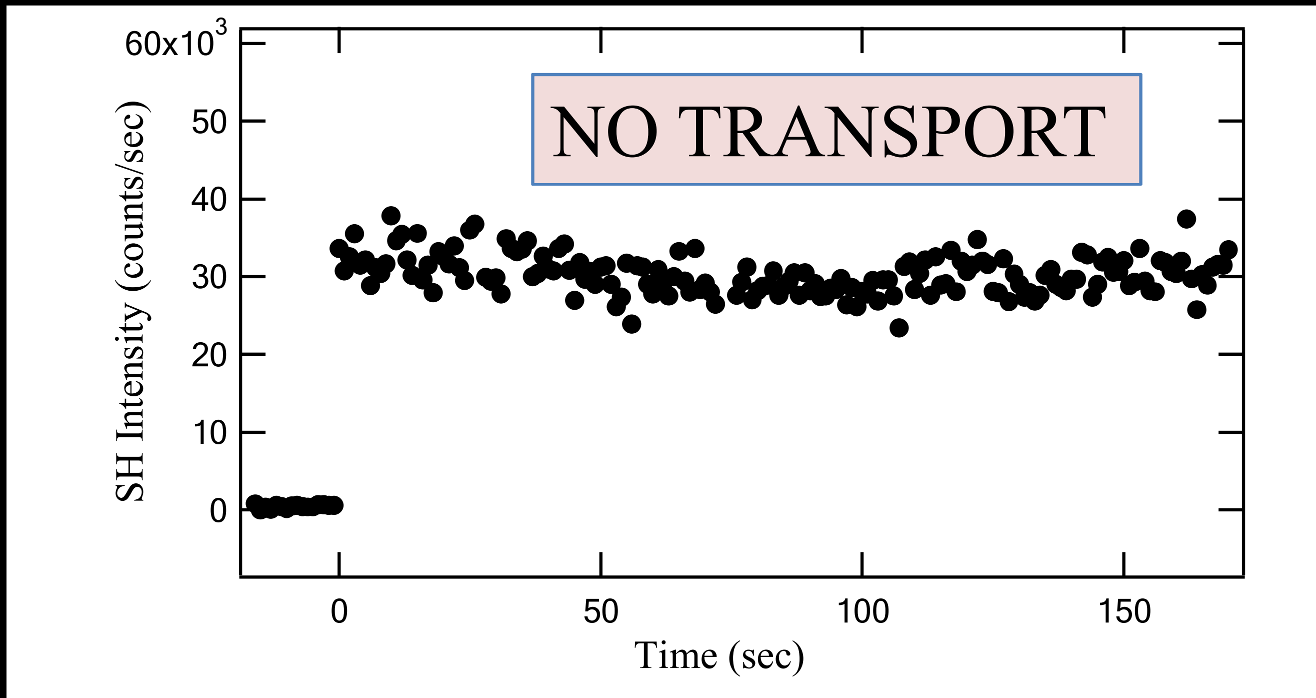
MEL Cell Membrane Structure: 3 Layers



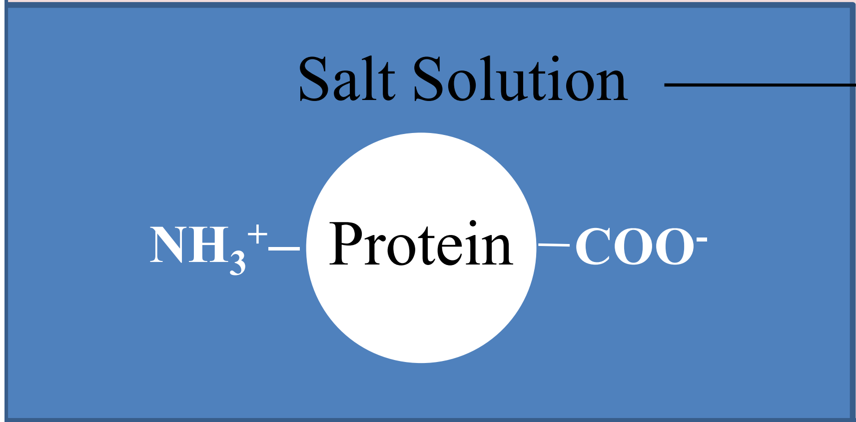
ION CHANNELS in red blood cell (RBC) membrane

1. the well-known Ca^{2+}-activated K^{+} channel (Gardos channel)
   high selectivity of K^{+} over Na^{+} if Ca^{2+} at the intracellular face of the channel
2. a non-selective cation channel (NSC)
   permeable to the divalent cations Ca^{2+} and Ba^{2+}, and even Mg^{2+}
3. anion channel in the RBC membrane

MG @ MEL Cell Membrane



Understanding Hofmeister Effect



- Protein activity
- Protein-Molecule interaction
- Protein stability
- …

Hydrophobic Interaction + Electrostatic Interaction

Phenomenological description-- **Hofmeister Series**

A particular ordering of ions in the ability to precipitate certain proteins from an aqueous solution

Partial Listing:

$$F^- \approx SO_4^{2-} > HPO_4^{2-} > acetate > Cl^- > NO_3^- > Br^- > ClO_3^- > I^- > ClO_4^- > SCN^-$$

$$NH_4^+ > K^+ > Na^+ > Li^+ > Mg^{2+} > Ca^{2+}$$

Salt Out (aggregate) ⟷ *Salt In (solubilize)*

Salt Solutions

Surface-modified Polystyrene (PS) as model system for protein

Nonlinear Light Scattering and Luminescence from Nanoparticles and Biological Cells

Nonlinear light scattering (SHG) from the surface of nanoparticles is detectable, and can be used to probe the particle surface.

Luminescence particles from nanoparticles can be much enhanced through surface modification.

SHG can be used to probe molecular interaction and transport at cell membranes.

College of Science and Technology
TEMPLE UNIVERSITY