

# Non Locality & the Limits to Sub Wavelength Focussing

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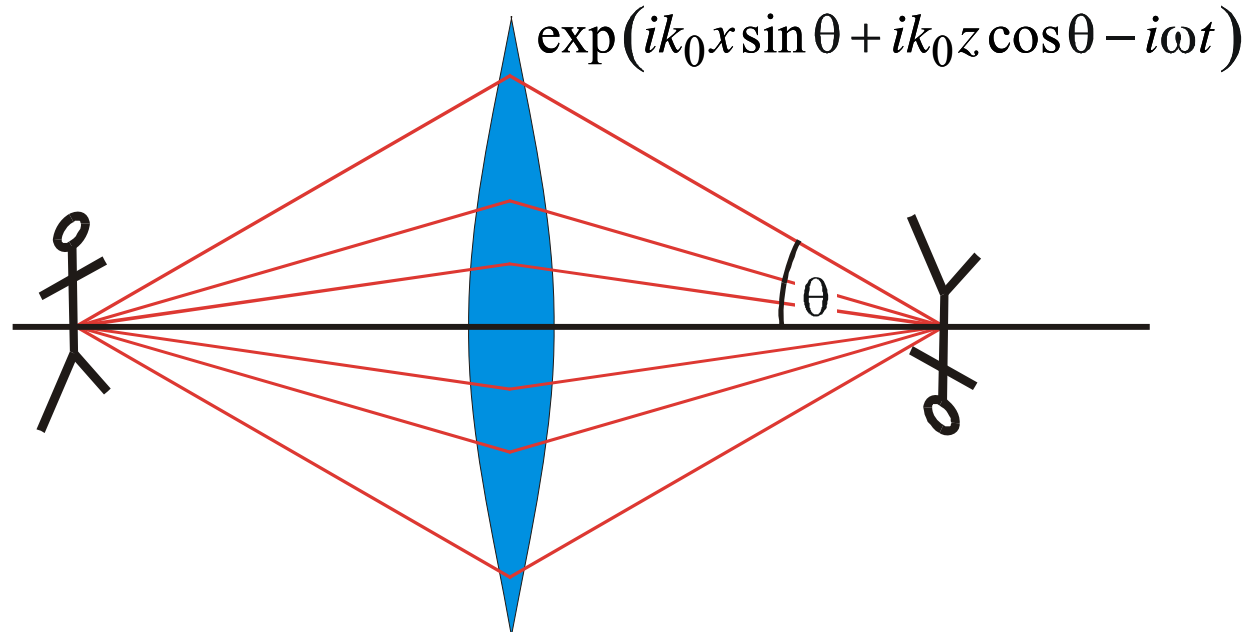
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for new experimental data see: *Science* **337**, 1071-4 (2012).

for a review see: *Science* **337**, 549-52 (2012).

# Focussing light: wavelength limits the resolution

Contributions of the far field to the image ....



..... are limited by the free space wavelength:

$\theta = 90^\circ$  gives maximum value of  $k_x = k_0 = \omega/c_0 = 2\pi/\lambda_0$  – the shortest wavelength component of the 2D image. Hence resolution is no better than,

$$\Delta \approx \frac{2\pi}{k_0} = \frac{2\pi c}{\omega} = \lambda_0$$

# Harvesting light – enhancing non linearity

*What is ‘harvesting’?*

A lens gathers beams of light from points in an object and brings them to a focus at the corresponding points in the image.

In contrast a light harvester **concentrates all the beams at a single point.**

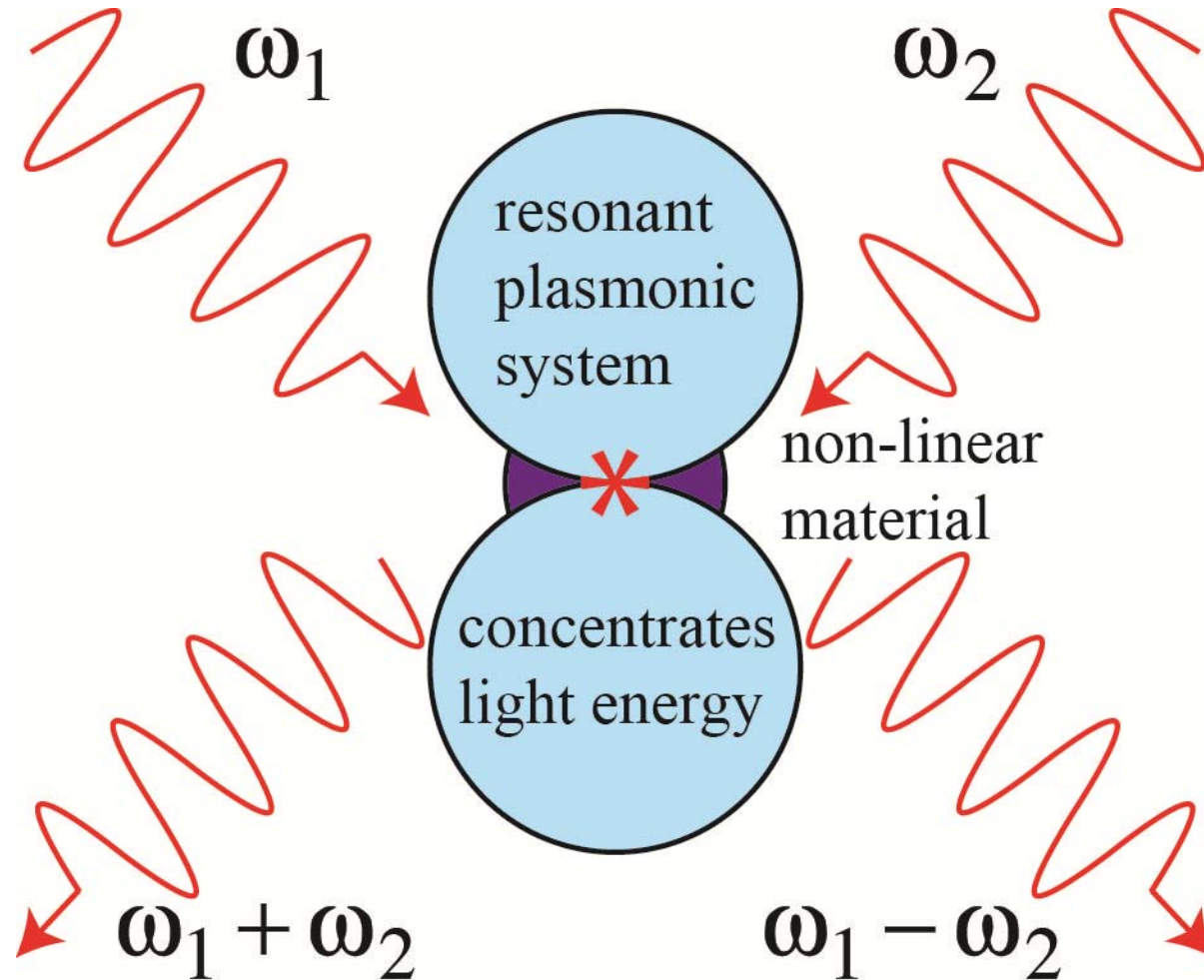
*Harvesters can be used for:*

- Making very intense concentrations of light, forcing photons to interact with one another
- Detecting molecules with high sensitivity – ideally sensing a single molecule
- Enhancing the efficiency of solar cells
- Improving the efficiency of lasers

Light harvesters suffer from the same wavelength restrictions as lenses, but we can use the same tricks of negative refraction to improve their performance

# Harvesting light – non linear mixing

Incident radiation at  $\omega_1, \omega_2$  interacts non linearly to produce  $\omega_1 + \omega_2$  and  $\omega_1 - \omega_2$ . The process is enhanced by harvesting energy to the site of the non-linear material.



# Strategy for broadband Harvesting of Light

- Start with a ‘mother system’ that collects light, is broad band, **and is sufficiently simple to be solved analytically**, but may not have the desired geometry.
- Make a coordinate transformation to a desirable geometry.
- Use ‘*transformation optics*’ to calculate the properties of the transformed system. The daughter structure inherits the mother’s analytical solution.
- A whole family of structures is now possible containing the same analytic genes but of sometimes wildly different shapes and sizes.

**Formal theory:** A. J. Ward and J.B. Pendry, *J Mod Op*, **43** 773 (1996)

also D.M. Shyroki (2003)

<http://arxiv.org/abs/physics/0307029v1>

New coordinates in terms of the old:  $x'^{j'}(x^j)$

In the new coordinate system we must use renormalized values of the permittivity and permeability,  $\epsilon, \mu$ :

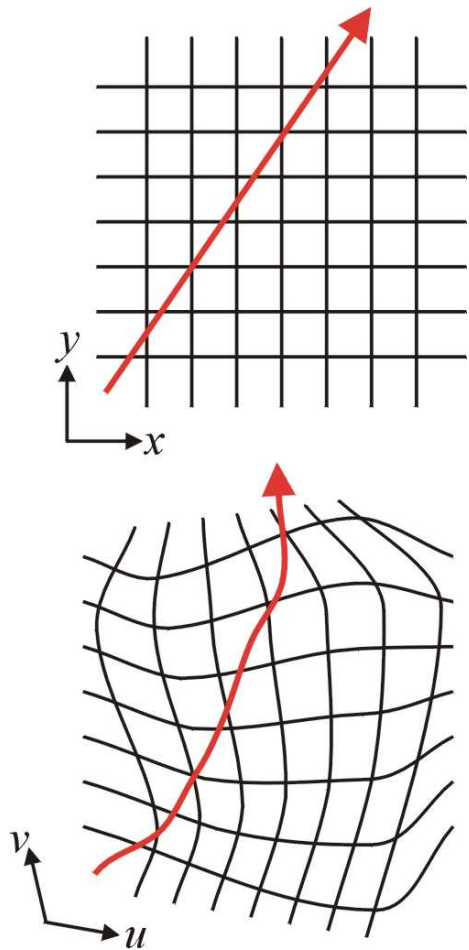
$$\epsilon'^{i'j'} = [\det(\Lambda)]^{-1} \Lambda_i^{i'} \Lambda_j^{j'} \epsilon^{ij}$$

$$\mu'^{i'j'} = [\det(\Lambda)]^{-1} \Lambda_i^{i'} \Lambda_j^{j'} \mu^{ij}$$

where,

$$\Lambda_j^{j'} = \frac{\partial x'^{j'}}{\partial x^j}$$

**For the special case of conformal transformations in 2D systems,  $\epsilon, \mu$  are unchanged.**



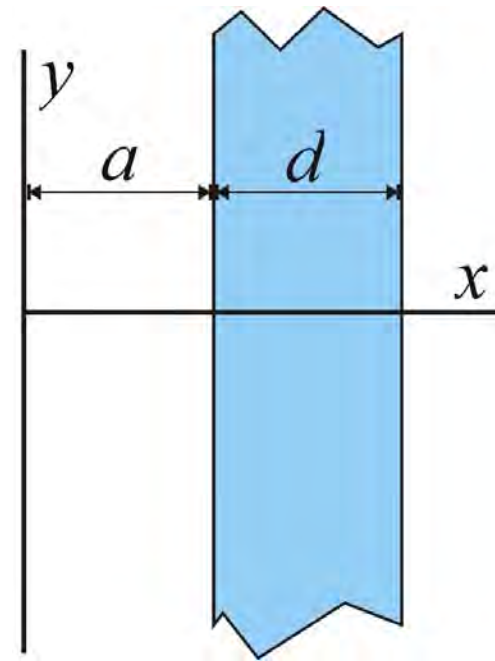
Top: a ray in free space with the background Cartesian coordinate grid shown.  
 Bottom: the distorted ray trajectory with distorted coordinates.

# Constructing a broadband absorber

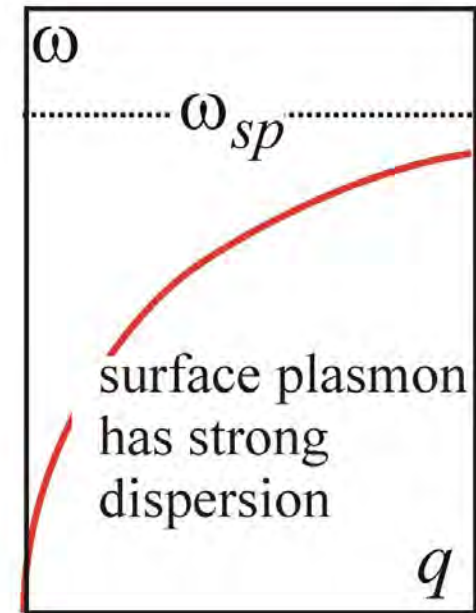
Resonant systems, such as silver spheres, enhance the absorption of radiation hence greatly improving the sensitivity to adsorbed molecules; but absorption by a single resonance is narrow band and therefore of limited use.

- start with a dipole exciting an infinite system – most infinite systems have a *broadband* continuum
- invert about the origin to convert to a finite system excited by a plane wave. The *spectrum is unchanged and remains broadband*.

a metallic slab of finite thickness has a broadband spectrum



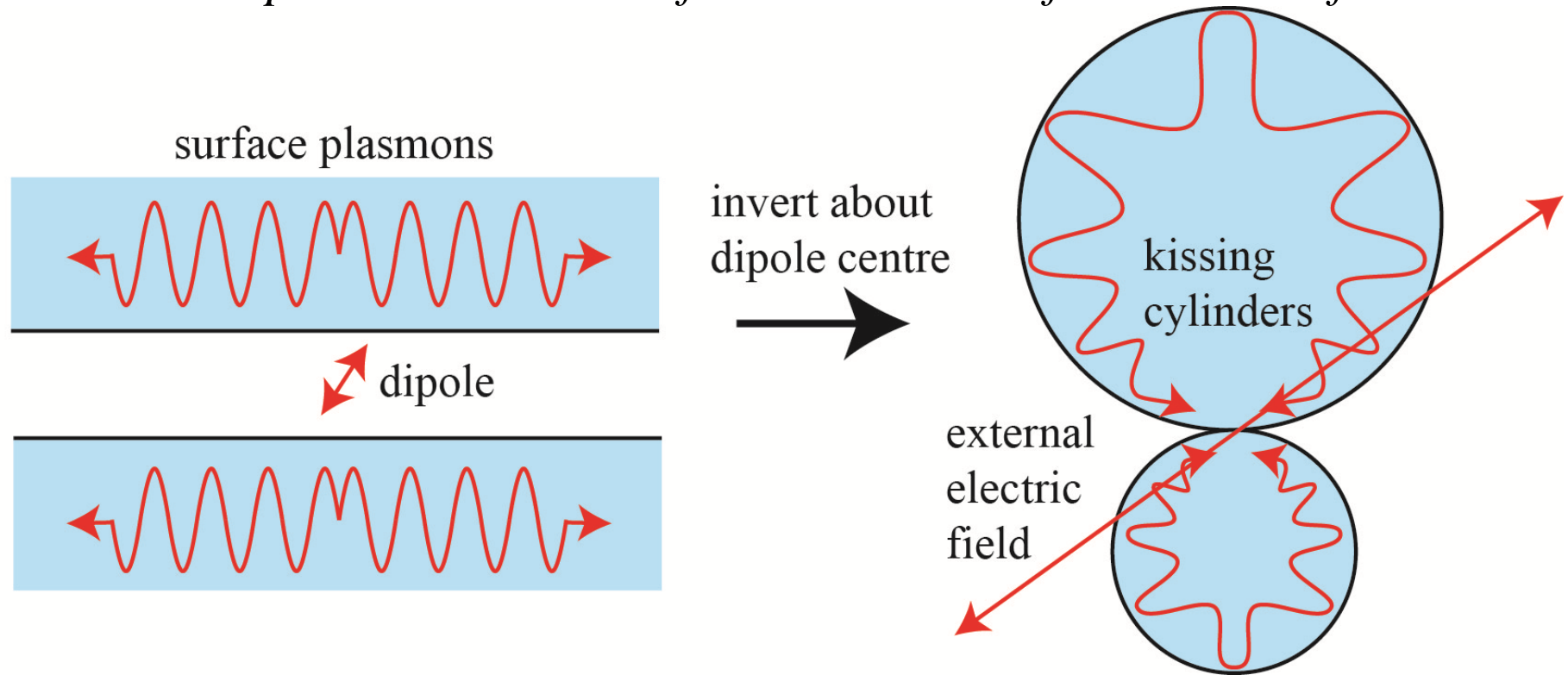
finite slab of silver





# Inversion about the origin, $z' = 1/z$ , converts a cavity to a pair of kissing cylinders

*The dipole source is transformed into a uniform electric field*



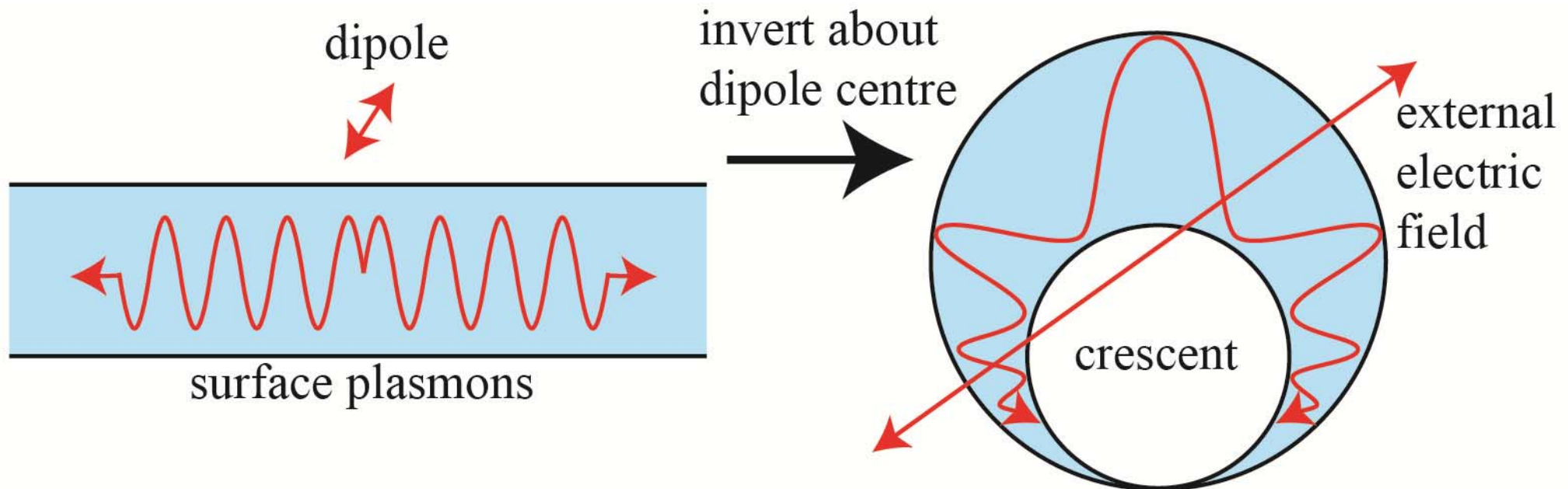
**Left:** a cavity supports surface plasmons that couple to a dipole source, transporting its energy to infinity. The spectrum is continuous.

**Right:** the transformed material now comprises two kissing cylinders. The dipole source is transformed into a uniform electric field.



# Inversion about the origin, $z' = 1/z$ , converts a slab to a cylindrical crescent

*The dipole source is transformed into a uniform electric field*

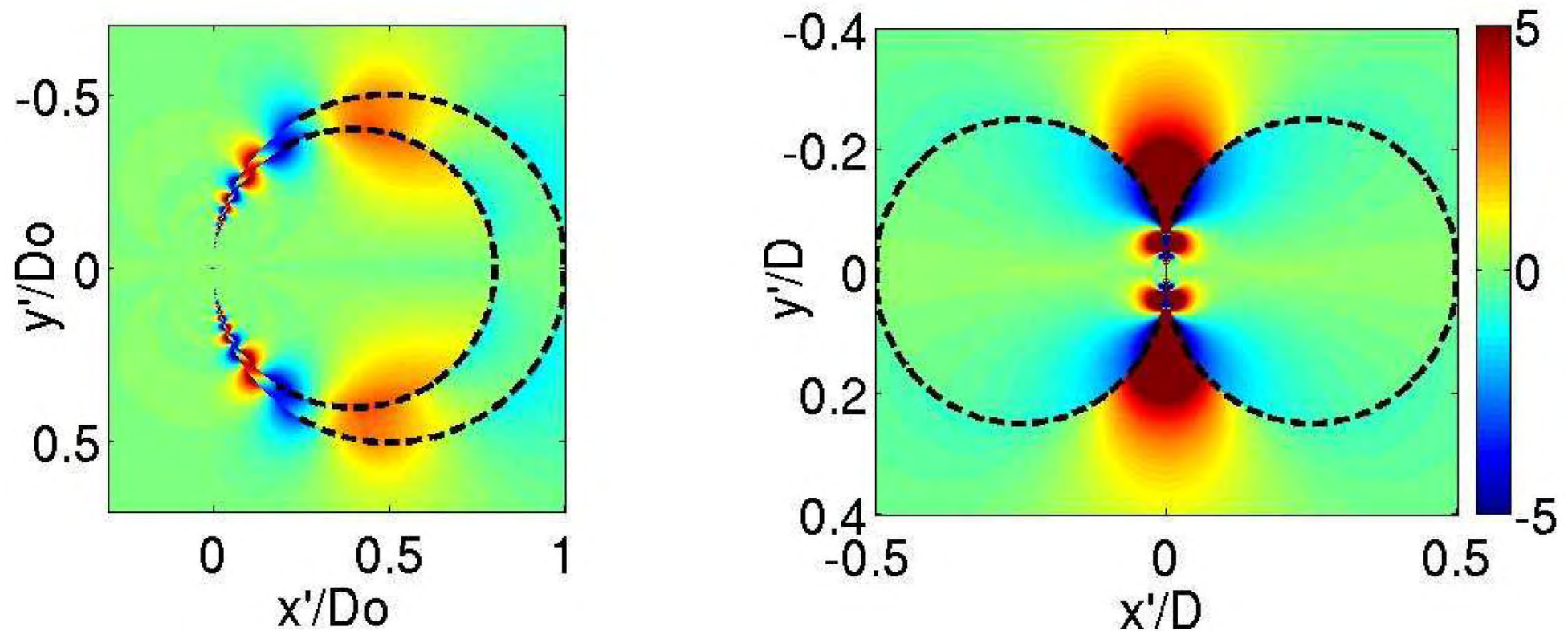


**Left:** a thin slab of metal supports surface plasmons that couple to a dipole source, transporting its energy to infinity. The spectrum is continuous and broadband therefore the process is effective over a wide range of frequencies.

**Right:** the transformed material now comprises a cylinder with cross section in the form of a crescent. The dipole source is transformed into a uniform electric field.

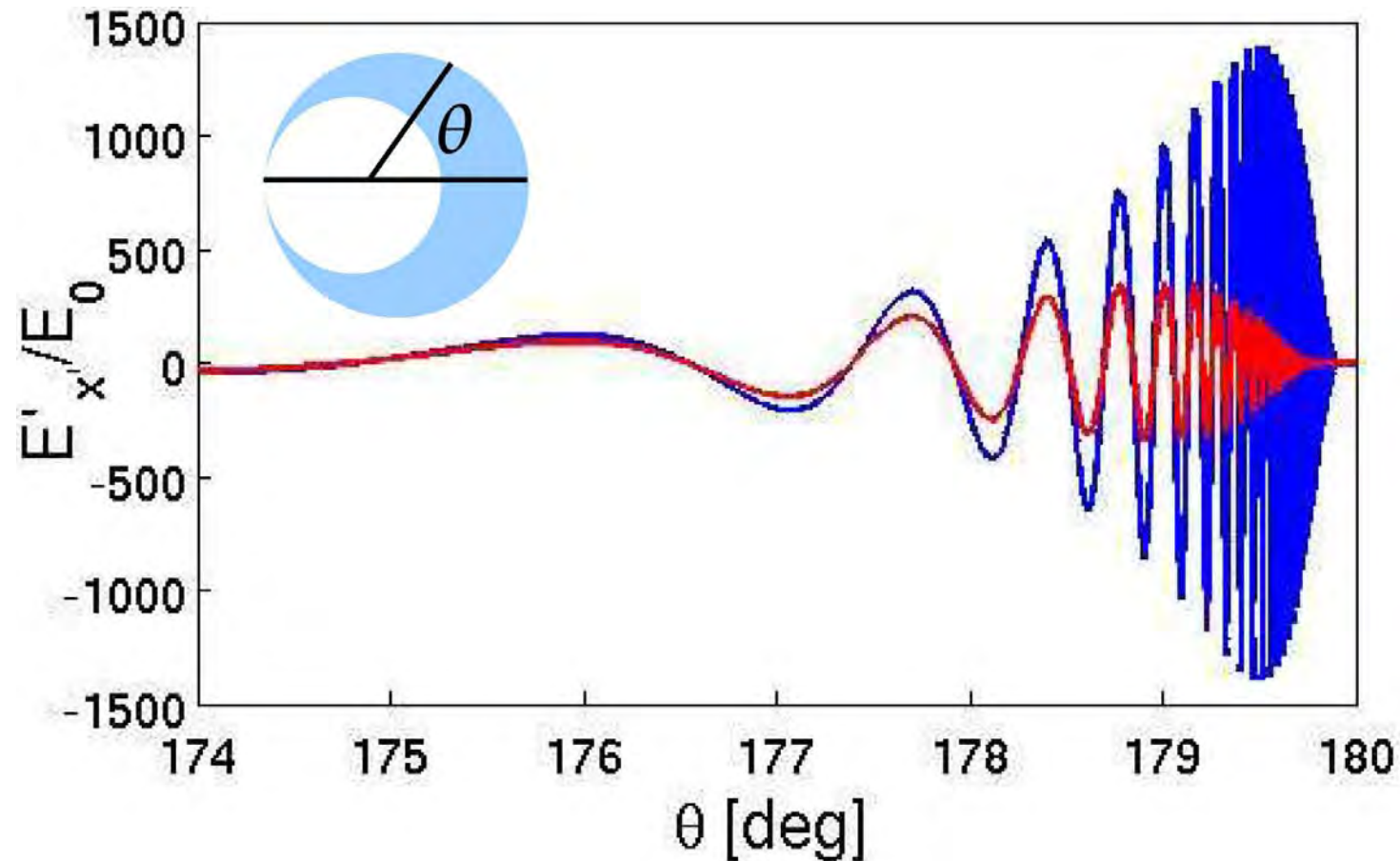
# Broadband field enhancement in singular structures

(Alexandre Aubry, Dang Yuan Lei, Antonio I. Fernandez-Dominguez, et al.)



Calculated  $E_x$  normalized to the incoming field ( $E$ -field along  $x$ ). The left and right panels display the field in the crescent and in the two kissing cylinders respectively. The metal is silver and  $\omega = 0.9\omega_{sp}$ . The scale is restricted to  $-10^5$  to  $+10^5$  but note that the field magnitude is far larger around the structural singularities.

# Field enhancement versus angle – crescent

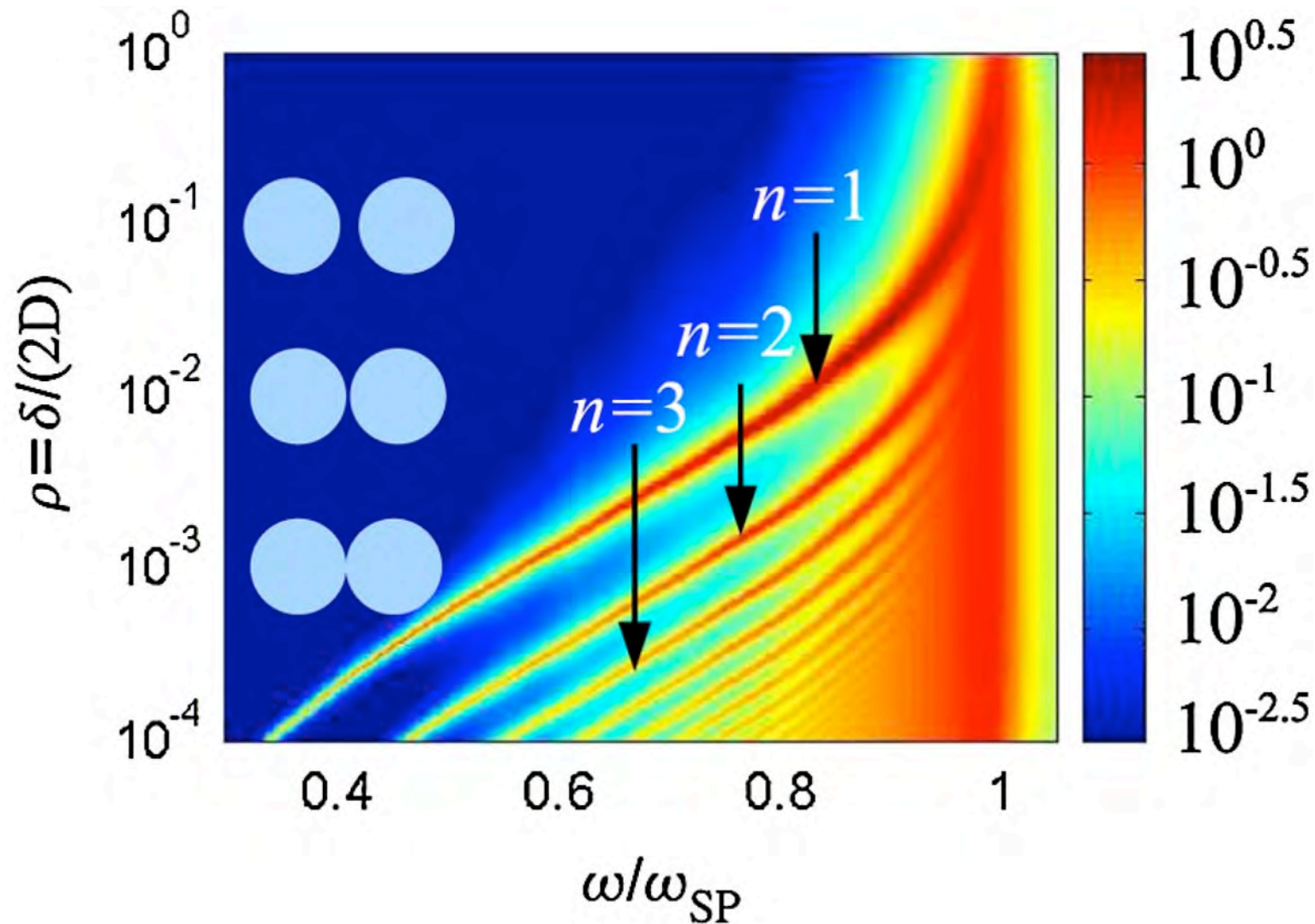


**Blue curve:**  $E_x$  at the surface of the crescent, plotted as a function of  $\theta$ , for  $\omega = 0.75\omega_{sp}$  and  $\varepsilon = -7.058 + 0.213i$  taken from Johnson and Christy.

**Red curve:**  $\varepsilon = -7.058 + 2 \times 0.213i$  i.e. more loss. Both curves are normalised to the incoming field amplitude  $E_0$ . The crescent is defined by the ratio of diameters  $r = 0.5$

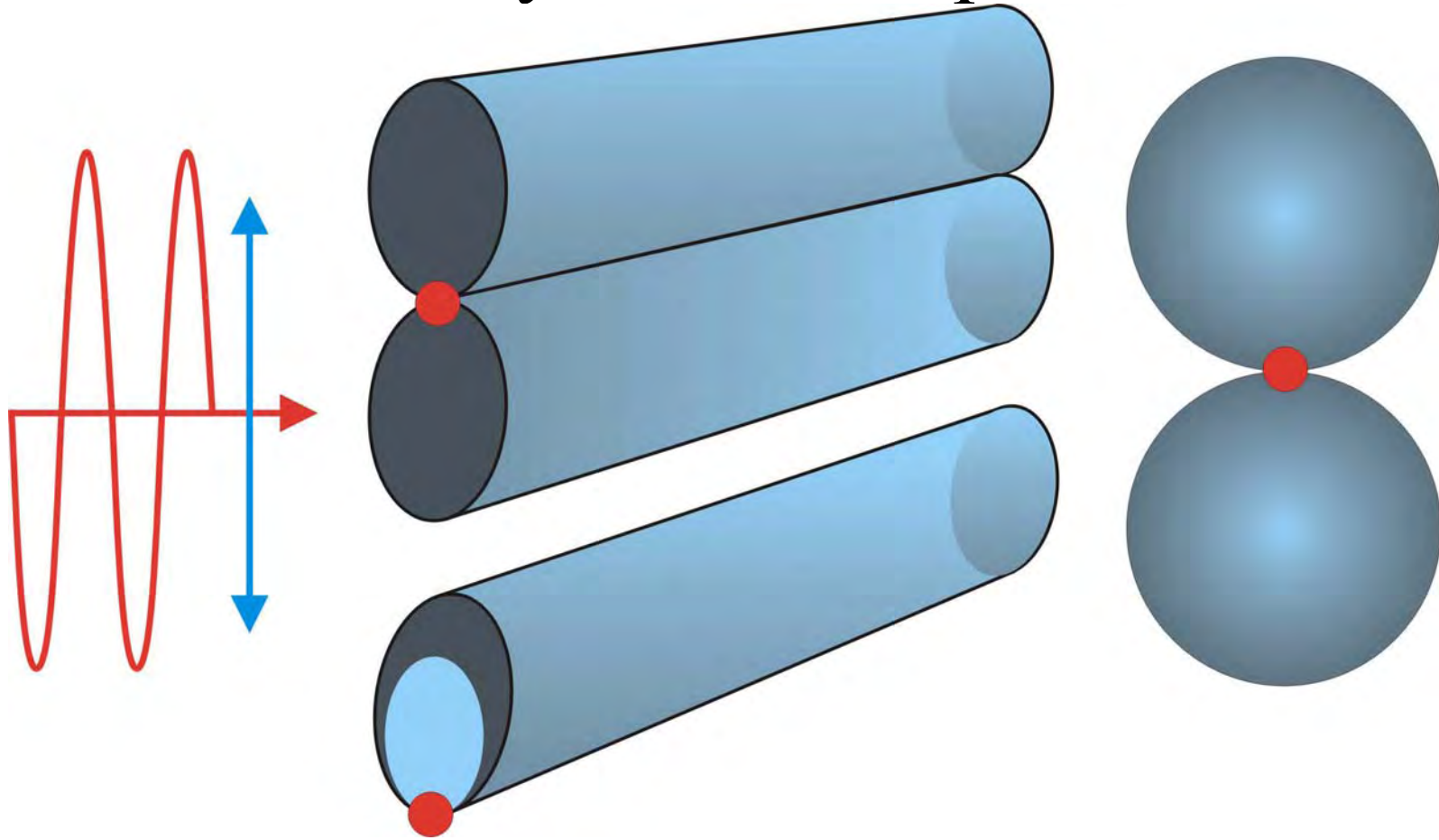


# Absorption cross section: non-touching cylinders



Absorption cross section normalised to the physical cross section,  $D_0$ , as a function of separation between the cylinders,  $\delta$ .

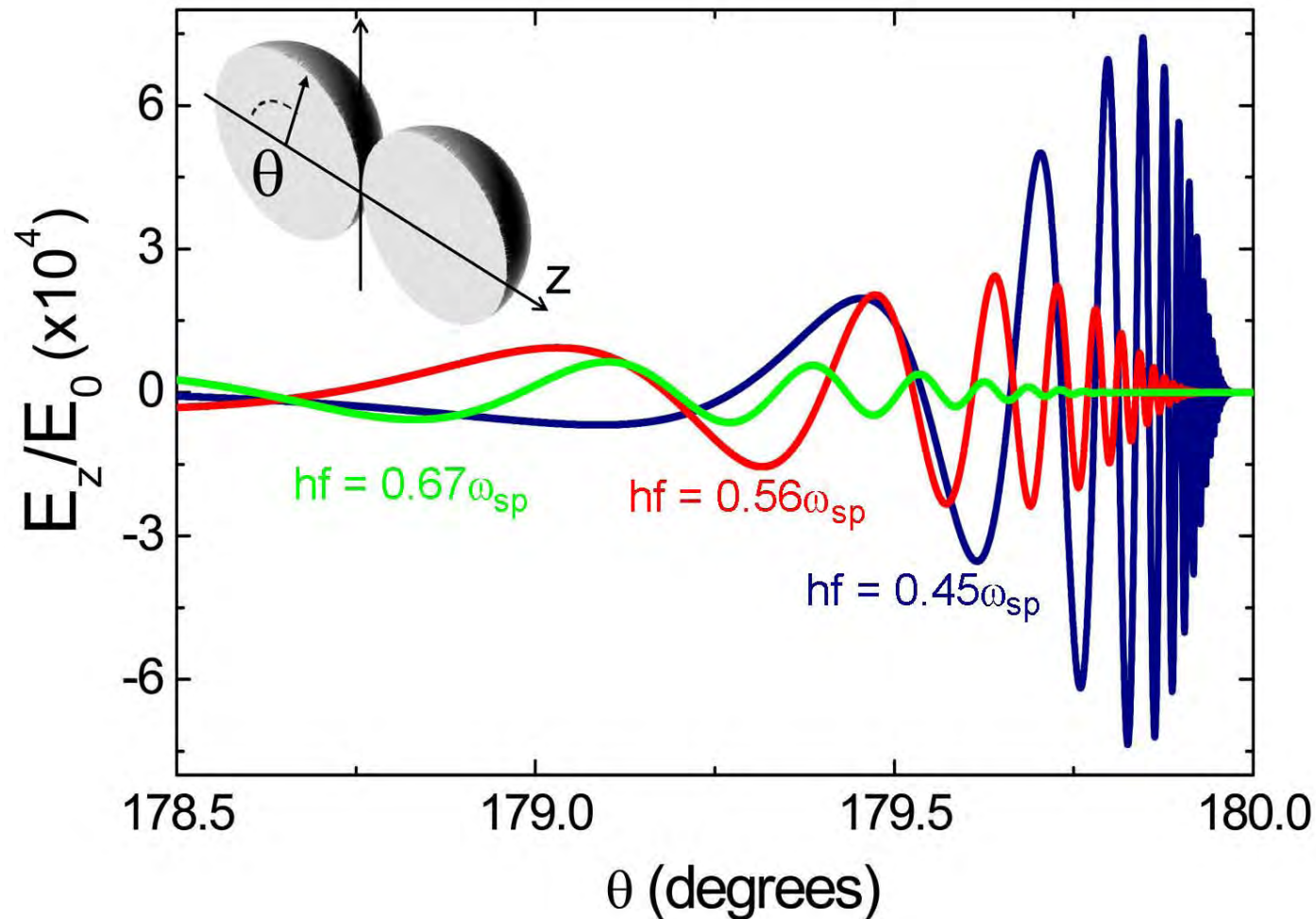
# Extension of Harvesting Theory from Cylinders to Spheres



Analytic theory in the electrostatic limit is exact for touching cylinders. For spheres theory approximates to the cylindrical solution near the touching point.

# Spheres as Light Harvesting Devices

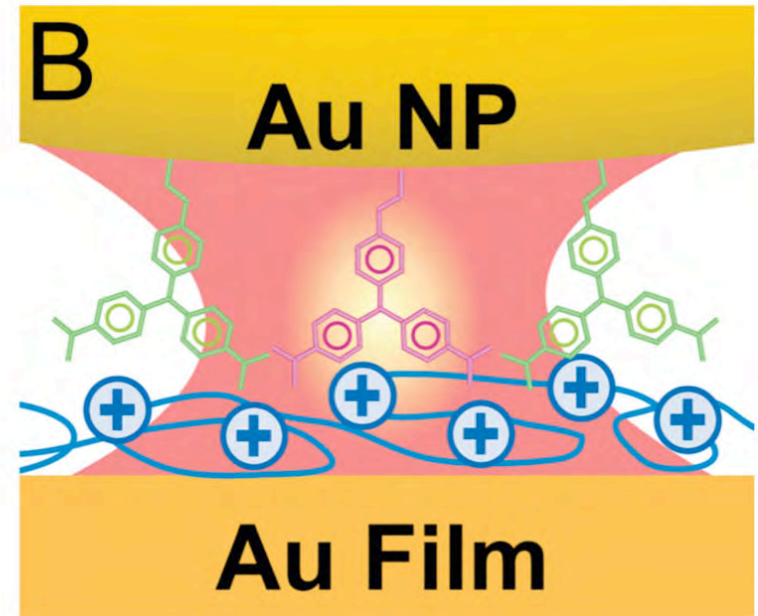
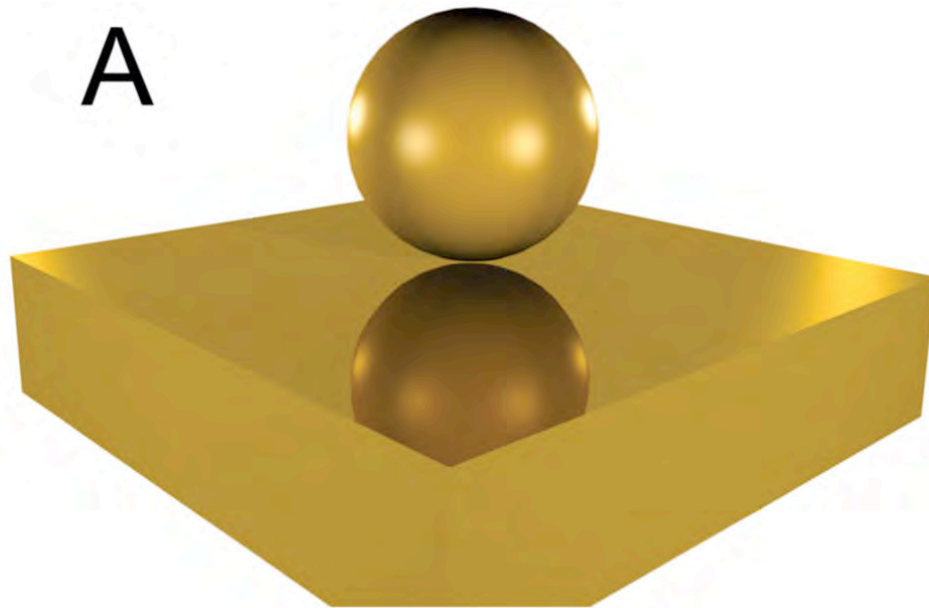
*Theory by Antonio Fernandez-Dominguez*



Electric field enhancement at various photon frequencies for touching spheres. Enhancement is an order of magnitude greater than for touching cylinders because energy is compressed in 2 directions.

# Leveraging Nanoscale Plasmonic Modes to Achieve Reproducible Enhancement of Light

Ryan T. Hill, Jack J. Mock, Yaroslav Urzhumov, David S. Sebba, Steven Oldenburg, Shiuan-Yeh Chen, Anne A. Lazarides, Ashutosh Chilkoti, and David R. Smith. *NanoLetters* **10** 4150 (2010).

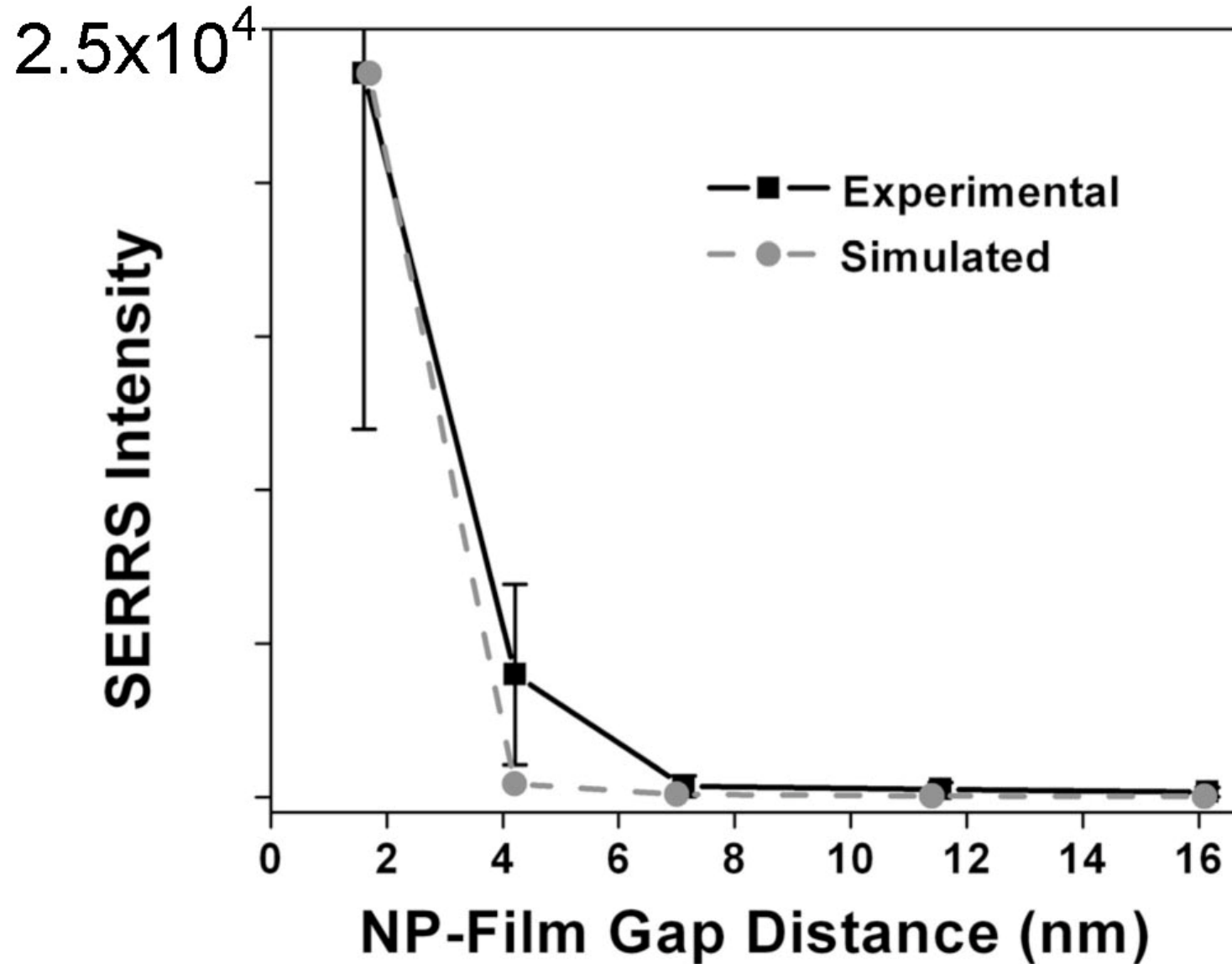


A: Gold nanoparticle near a gold. B: Localized NP-film field enhancement using SERRS from MGITC molecules adsorbed on the surface of the NP and within the NP-film gaps, which were created by a single molecular layer of PAH (0.6nm layer thickness).



# Measured distance dependence of SERRS

from MGITC molecules in the NP-film gaps with varying spacer distances.



# Benefits of Broadband Harvesting

- Multi - frequency systems benefit from enhancement of all frequencies.
- e.g. one system can enhance detection of a wide range of molecules
- e.g. if we are amplifying a weak signal at one frequency using a pump of another frequency, both are enhanced.

# What can go wrong?

The theory predicts spectacular enhancements in the harvested fields, even when realistic values of the silver permittivity are included. **Harvesting is much less sensitive to resistive losses than is perfect imaging.**

Enhancements in field strength of  $10^4$  are predicted, implying an enhancement of the SERS signal of  $10^{16}$ . Several factors will prevent this ideal from being attained:

- radiative losses
- problems in nm scale precision manufacture
- non locality of  $\epsilon$

Nevertheless substantial effects can be expected

# Nonlocality – what is it?

*Formal definition:* the longitudinal permittivity,  $\varepsilon_L$ , depends on wave vector as well as frequency.

The *physical interpretation* for metals at optical frequencies is that the bulk plasmon frequency also depends on wave vector and is defined by,

$$\varepsilon_L(k, \omega_p) = \varepsilon_\infty \left[ 1 - \frac{\omega_0^2}{\omega_p(\omega_p + i\gamma) - \beta^2 k^2} \right] = 0$$

Since the bulk plasmon controls the screening charge in a metal, the surface charges induced by external fields no longer appear as delta functions at the surface but decay smoothly into the bulk with a decay defined by,

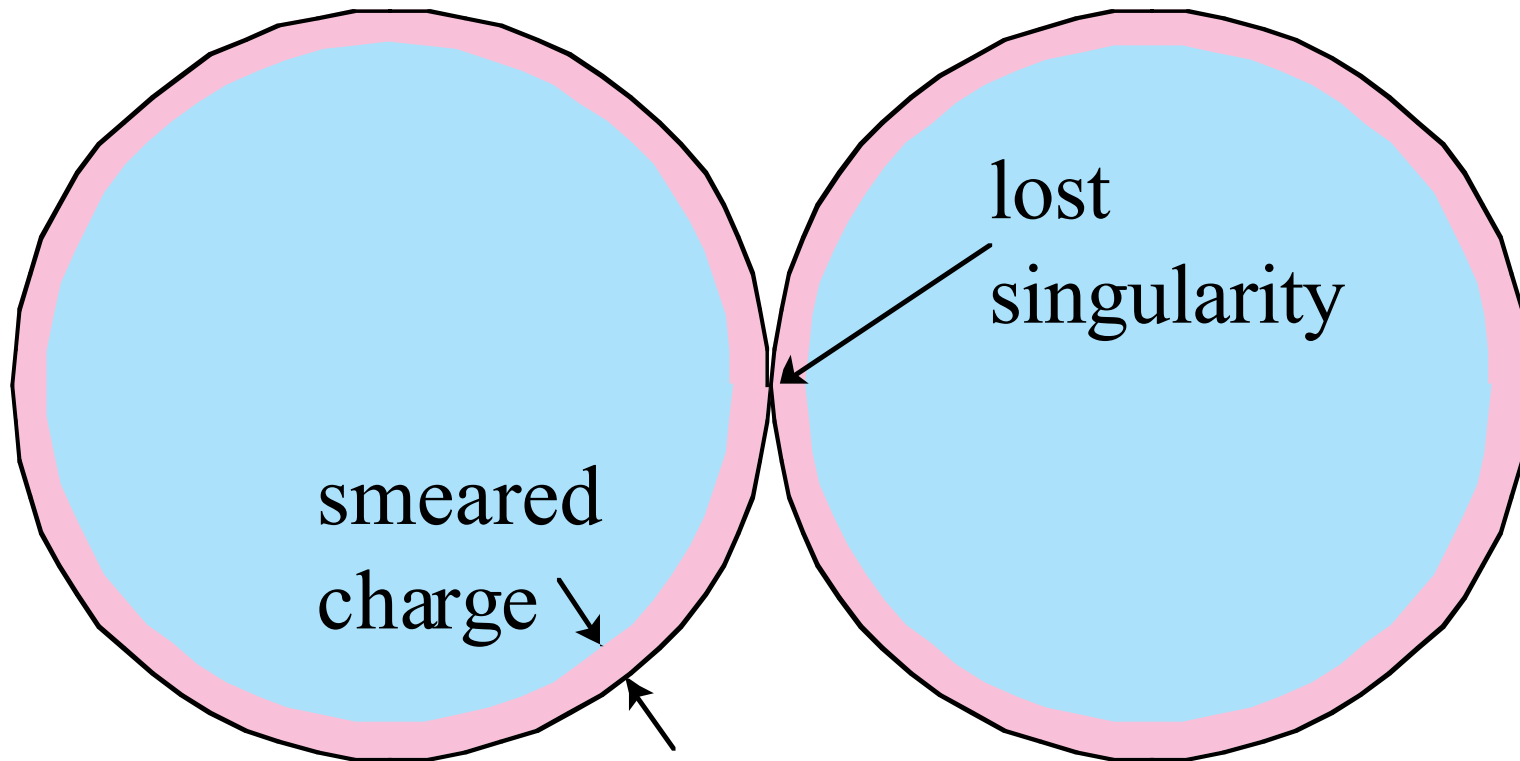
$$\exp(-\delta^{-1}z), \quad \delta^{-1} = \beta^{-1} \sqrt{\omega_0^2 - \omega_p(\omega_p + i\gamma)}$$

Typically  $\delta \approx 0.2\text{nm}$ , a very small length, but one that is sometimes important.

# Nonlocality and light harvesting

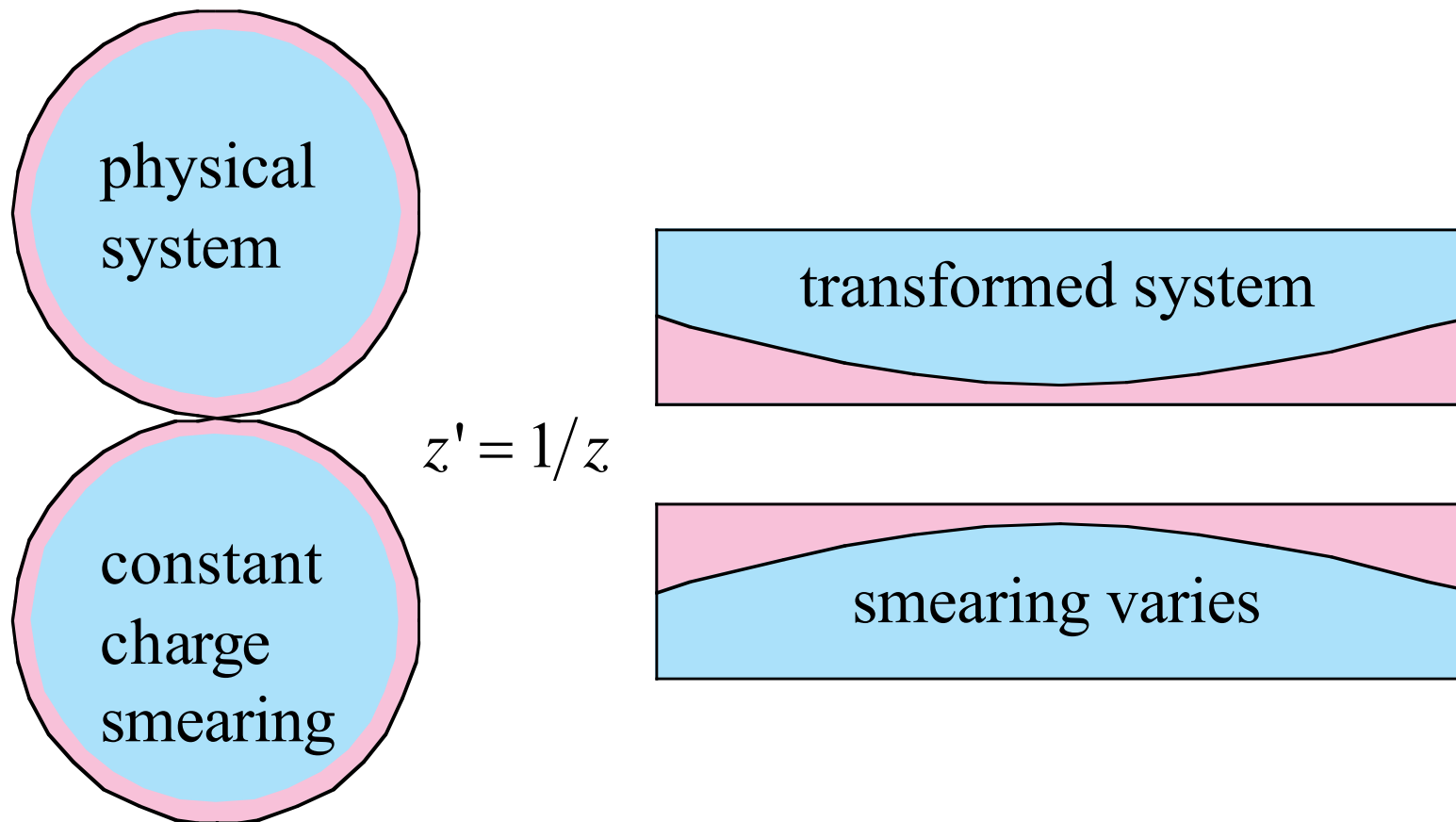
*most of this work was performed by Antonio Domingues*

Non locality smears out polarisation charge responsible for driving field enhancements at the touching point. Although the cylinders appear to touch, the charge distributions do not. Hence the field enhancement is degraded.



# Nonlocality and transformation optics

We have succeeded applying transformation optics to nonlocal systems but we must transform  $k$  as well as  $\epsilon$  and  $\mu$ . For example for touching spheres, the conformal transformation  $z' = 1/z$  which transforms to a slab waveguide, also alters the scale of charge smearing.



# Nonlocality and field enhancement

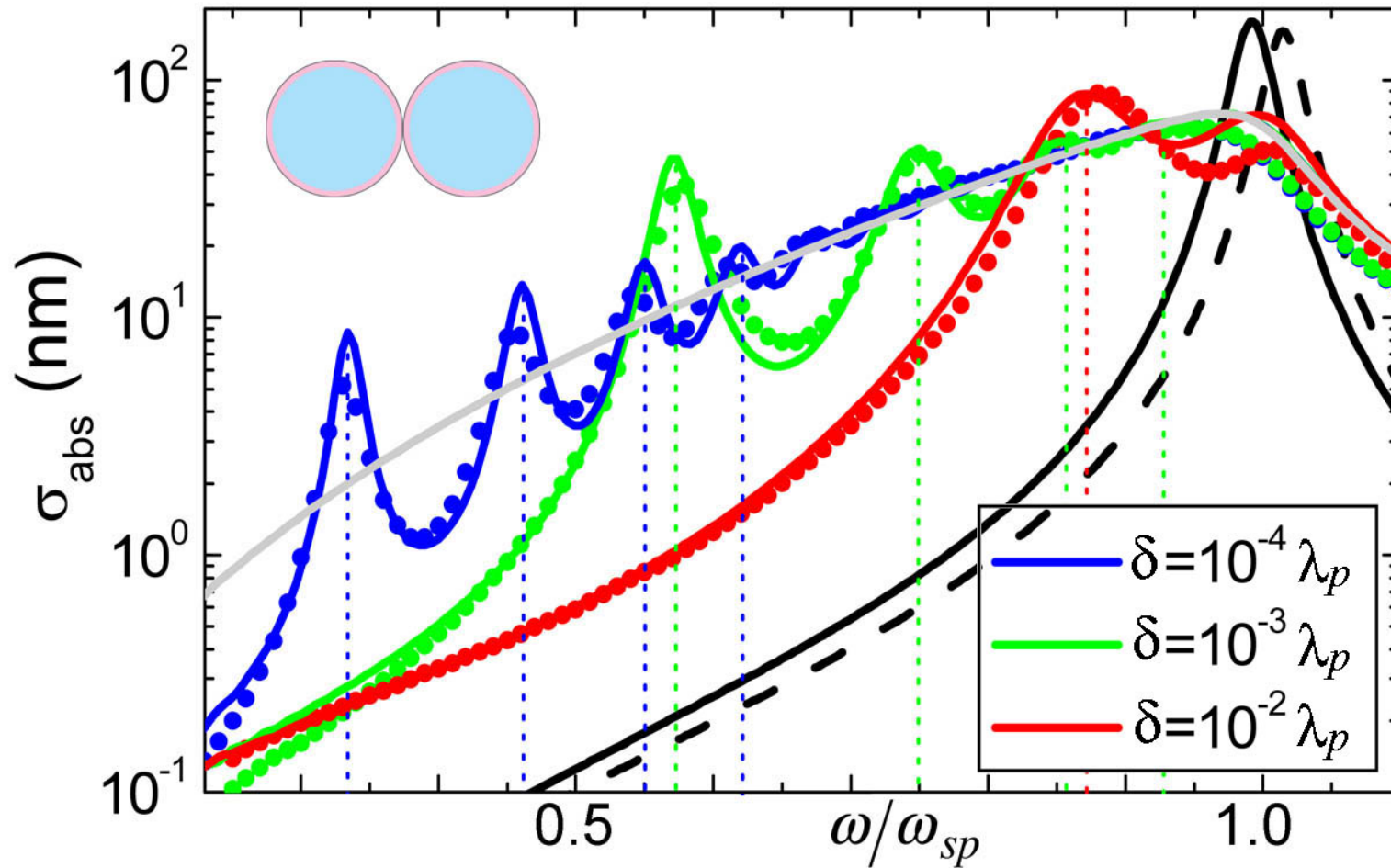
Because the polarisation charge distributions are smeared, and no longer touch at the singularity, the characteristics of two touching but nonlocal cylinders resemble two almost touching cylinders:

- waves travelling towards the touching points no longer slow to zero velocity, but head on past the touching point and as a result:
- the spectra are no longer continuous
- enhancement is reduced in the vicinity of the touching point

*The effects grow larger as the overall size of the system shrinks*

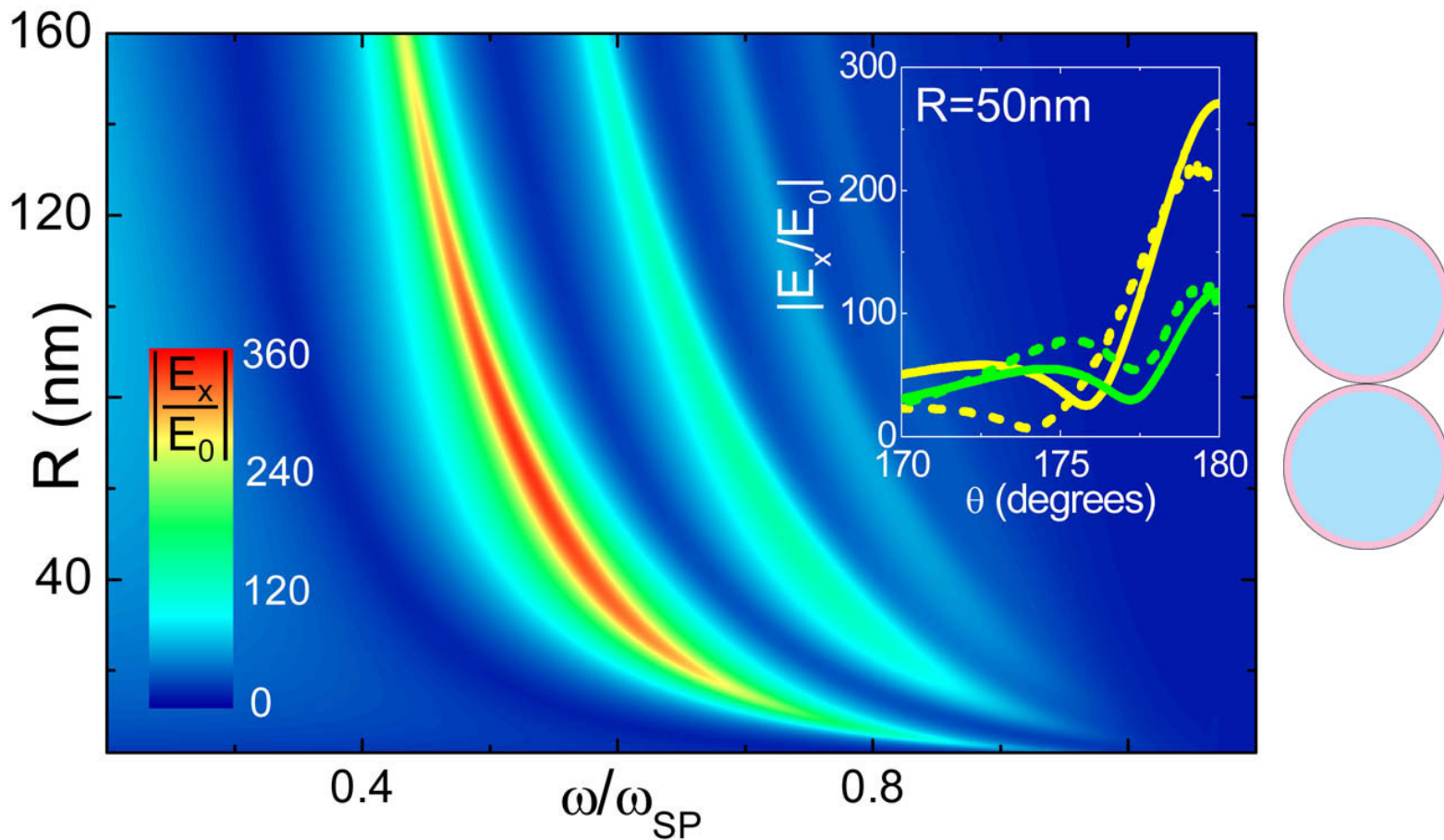


# Nonlocality quantises spectra of touching cylinders



Absorption spectra for 10nm radii non-local cylinders for several degrees of nonlocality. The green line shows a typical result. The local approximation is shown in grey. Black line: results for a single cylinder.

# Nonlocality versus radiation loss



Electric field enhancement for two touching cylinders at the touching point as a function of cylinder radius,  $R$ , and frequency  $\omega$ . **Radiation loss** kills enhancement for large cylinders, **non local effects** for small. Optimum enhancement is found for  $35\text{nm} < R < 80\text{nm}$ .

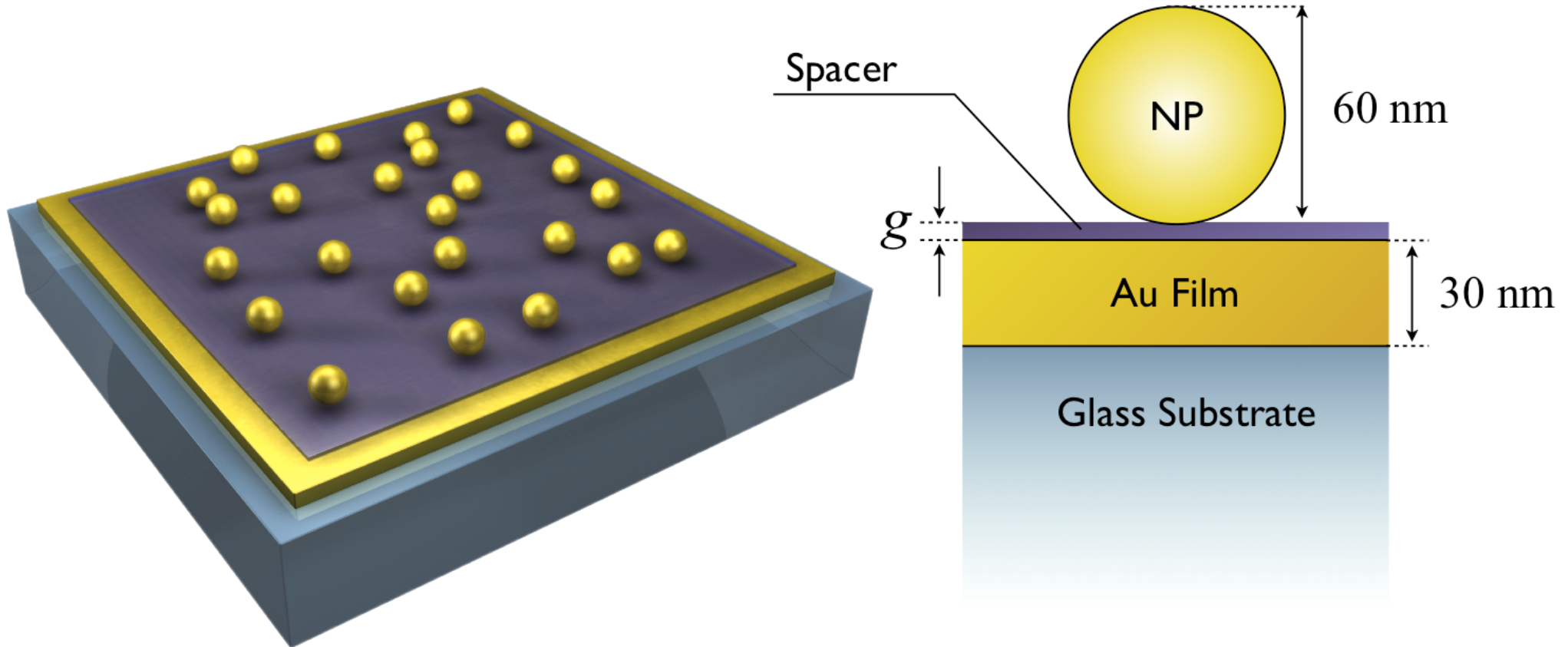
# Measuring non-locality

reported last month in:

*Probing the Ultimate Limits of Plasmonic Enhancement*

C. Ciraci, R. T. Hill, J. J. Mock, Y. Urzhumov, A. I. Fernández-Domínguez,  
S. A. Maier, J. B. Pendry, A. Chilkoti and D. R. Smith,  
*Science*, **337**, 1071-4, 2012

# Measuring non-locality – experimental setup



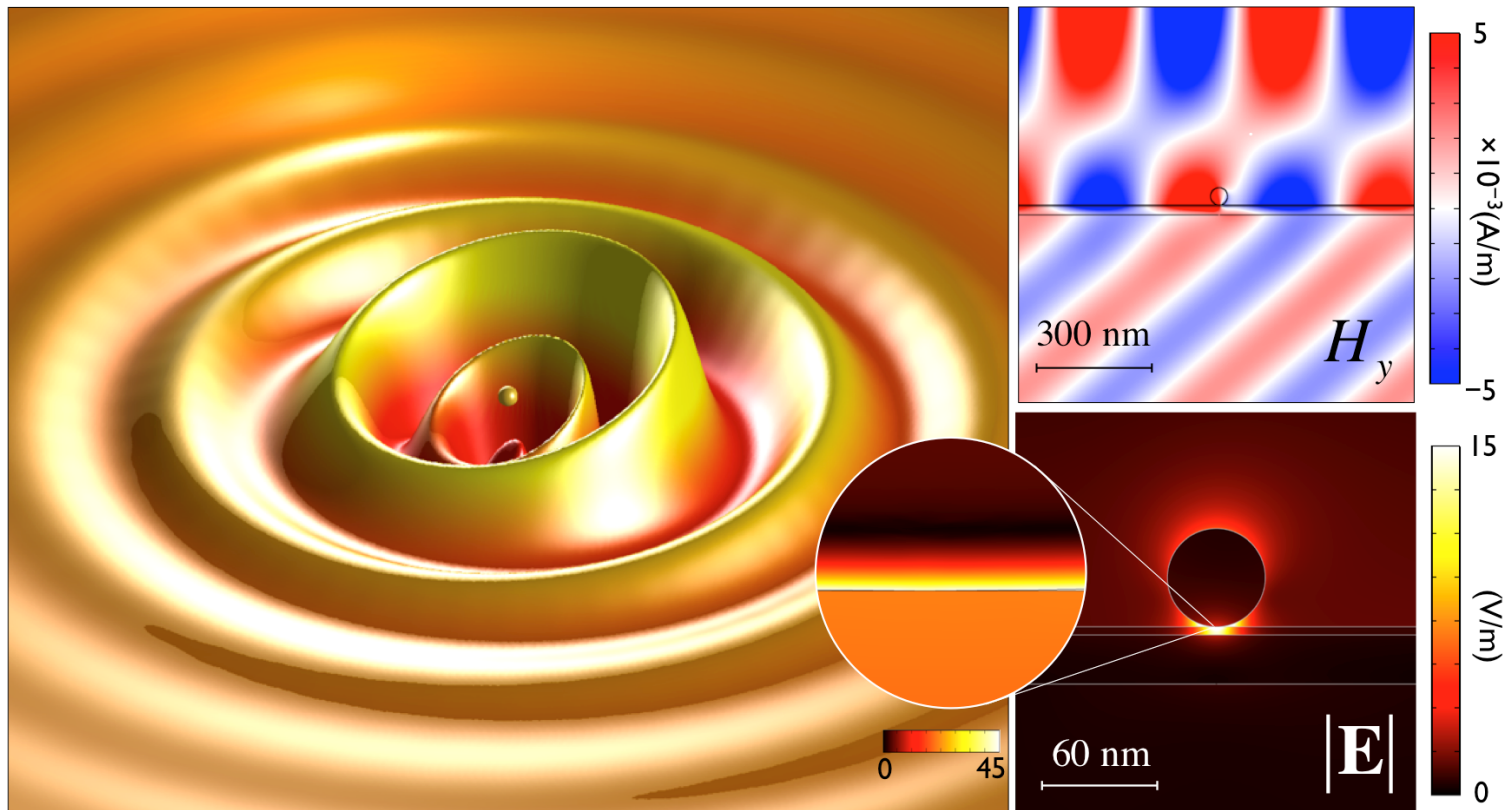
Geometry of the film-coupled nanoparticle.

**Left:** Schematic of the sample.

**Right:** Cross-section of a single film-coupled nanosphere.

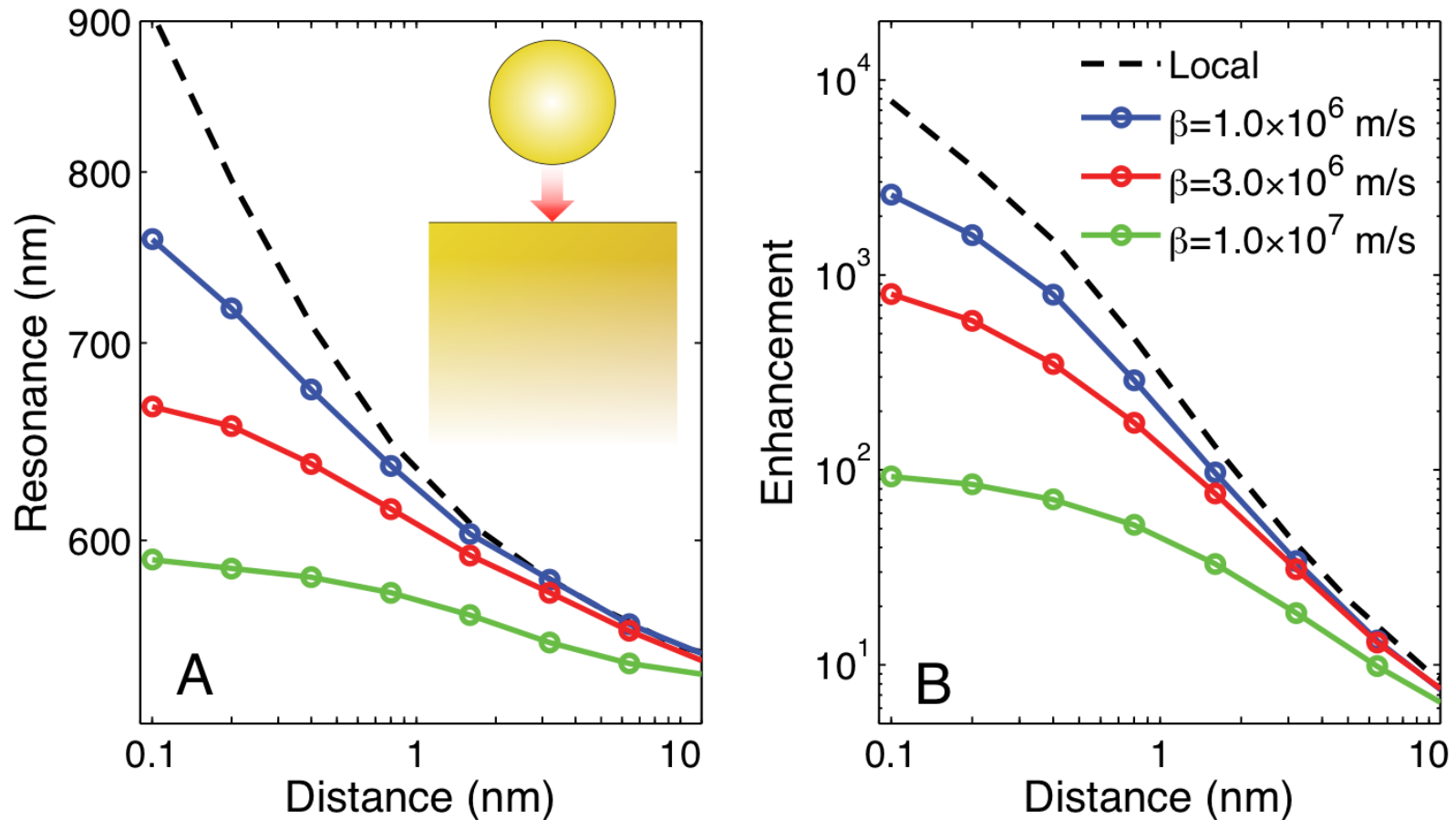


# Measuring non-locality – simulations I



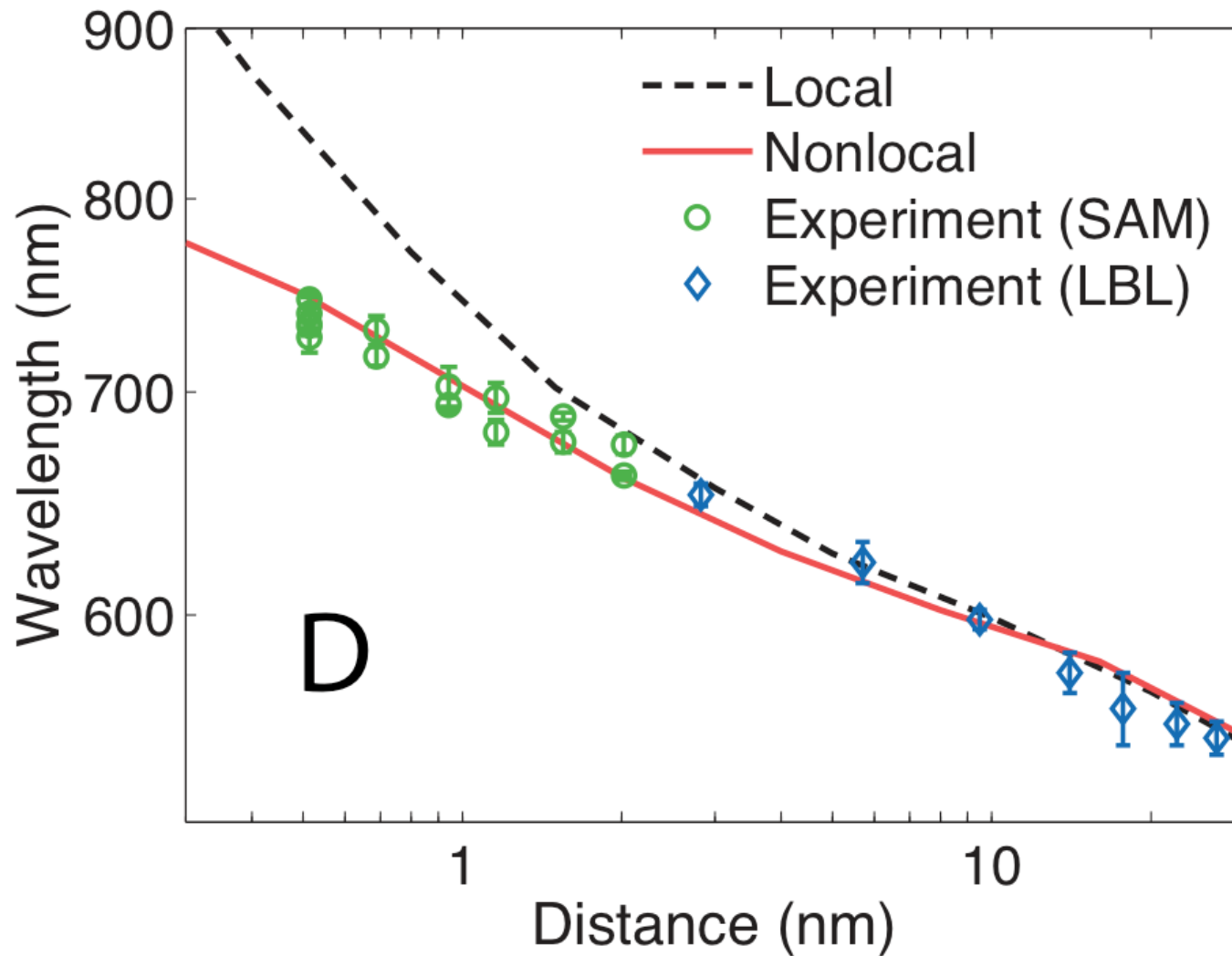
Simulation of a single film-coupled nanoparticle. **Left:** Relative electron surface density showing the excited surface plasmon polariton propagating over the metal film. **Right (top):** A plane wave is incident at  $75^\circ$  from normal. **Right (bottom):** The near-fields surrounding the nanosphere. Looking closer yet, it can be seen that the fields penetrate into the nanosphere a distance on the order of the Thomas-Fermi screening length.

# Measuring non-locality – simulations II



Resonant frequency (left), and local enhancement (right) at various sphere/surface separations of a gold nanosphere of radius  $r$  on a 300 nm thick film for various degrees of non locality.

# Measuring non-locality: comparison with expt.



Comparison of experimental measurements from both SAM and LBL type spacers with numerical results with a best fit  $\beta = 1.27 \times 10^6$  m/s. i.e. the polarisation charge penetrates  $\delta \approx 0.6$  nm into the metal.



# Non locality versus tunnelling

At the sub-nanometre level non *local effects* can be expected to be important but there is another phenomenon which will play a role at the shortest separations: *electron tunnelling*.

Currently there is a strong debate amongst theorists as to which dominates that can only be resolved by experiment.

Establishing an accurate description of electromagnetism on the sub nanometre scale will in the future be vital to a quantitative understanding the next generation of experiments.

# Conclusions

- Broadband light harvesting systems can be designed and studied analytically using transformation optics.
- Resistive losses are not the major limiting factor in harvesting potential.
- Radiative losses are minimised for small systems  $> 80\text{nm}$ .
- Nonlocal effects are the main limitation and are important for systems  $< 35\text{nm}$ .
- Given precise manufacturing field enhancements of the order of 300 are possible, which implies Raman enhancements approaching  $10^{10}$ .

# Non Locality & the Limits to Sub Wavelength Focussing

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# Radiative Losses

In the electrostatic approximation there are no radiative losses. This is a valid assumption provided that the cylinders are small compared to the wavelength.

In practice this means:

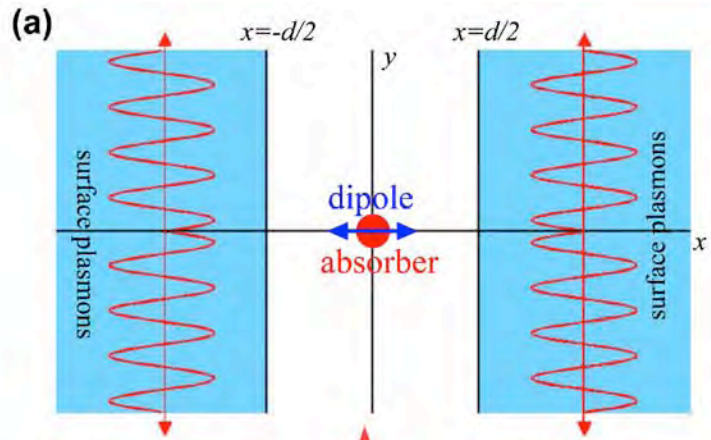
$$D(\text{diameter}) < 200\text{nm}$$

Their effect is to damp the resonant harvesting states and steal energy from the light harvesting mechanism.

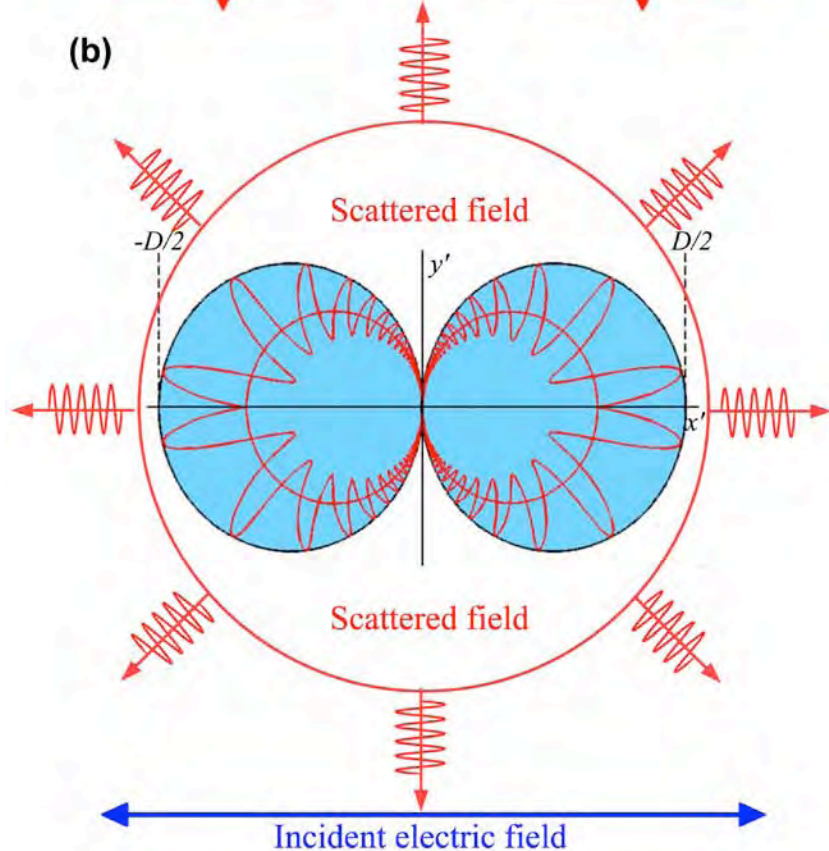
In the electrostatic limit the harvesting cross section  $\sigma_a \propto D^2$  reflecting the scale invariance of an electrostatic system.

However computer simulations using COMSOL show this scaling breaking down as radiative corrections kick in and  $\sigma_a$  falling dramatically.

# Calculating radiative corrections analytically



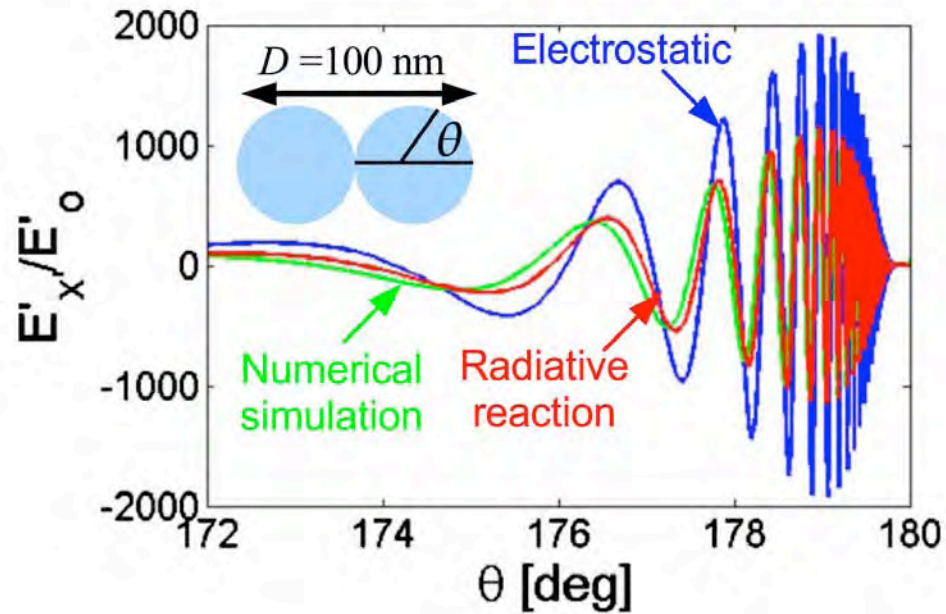
(a) Two semi-infinite metal slabs support surface plasmons that couple to a dipole source, transporting its energy to infinity.



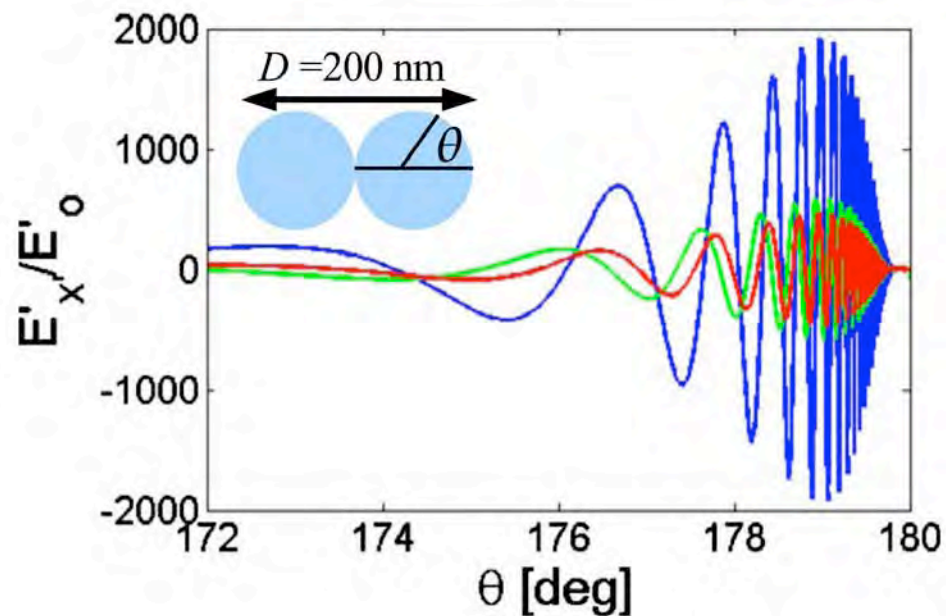
A fictional absorbing particle superimposed on the emitting dipole is chosen to account for the radiative damping in the transformed geometry.

(b) The transformed material consists of two kissing cylinders. The dipole source is transformed into a uniform electric field, and the radiative losses are approximated by lossy material outside the large sphere.

# The effect of radiative loss on enhancement



Electric field along the  $x$ - direction normalized by the incident electric field  $E_0$ , for  $D = 100$  nm and  $D = 200$  nm for  $\omega = 0.9 \omega_{sp}$ .



The analytical prediction for the effect of radiative loss (red) is compared to the numerical result (green) and to the electrostatic case where there is no radiative loss (blue).