



The Abdus Salam  
International Centre for Theoretical Physics



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**WINTER COLLEGE ON OPTICS ON OPTICS AND PHOTONICS  
IN NANOSCIENCE AND NANOTECHNOLOGY**

(7 - 18 February 2005)

***"Optical Properties  
of Plasmonic Nanosystems"- I***

presented by:

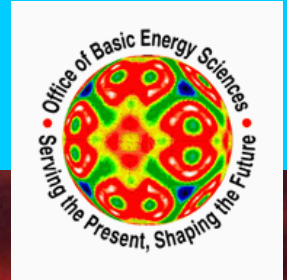
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**These are preliminary lecture notes, intended only for distribution to participants.**



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Support:



# Theory of Nanoplasmonics 1: Optical Properties of Plasmonic Nanosystems

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02/10/2005 1



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## LECTURE 1

### 1. Introduction

Problem of nanolocalization of energy

Surface plasmons and enhanced optical fields

Surface plasmon polaritons

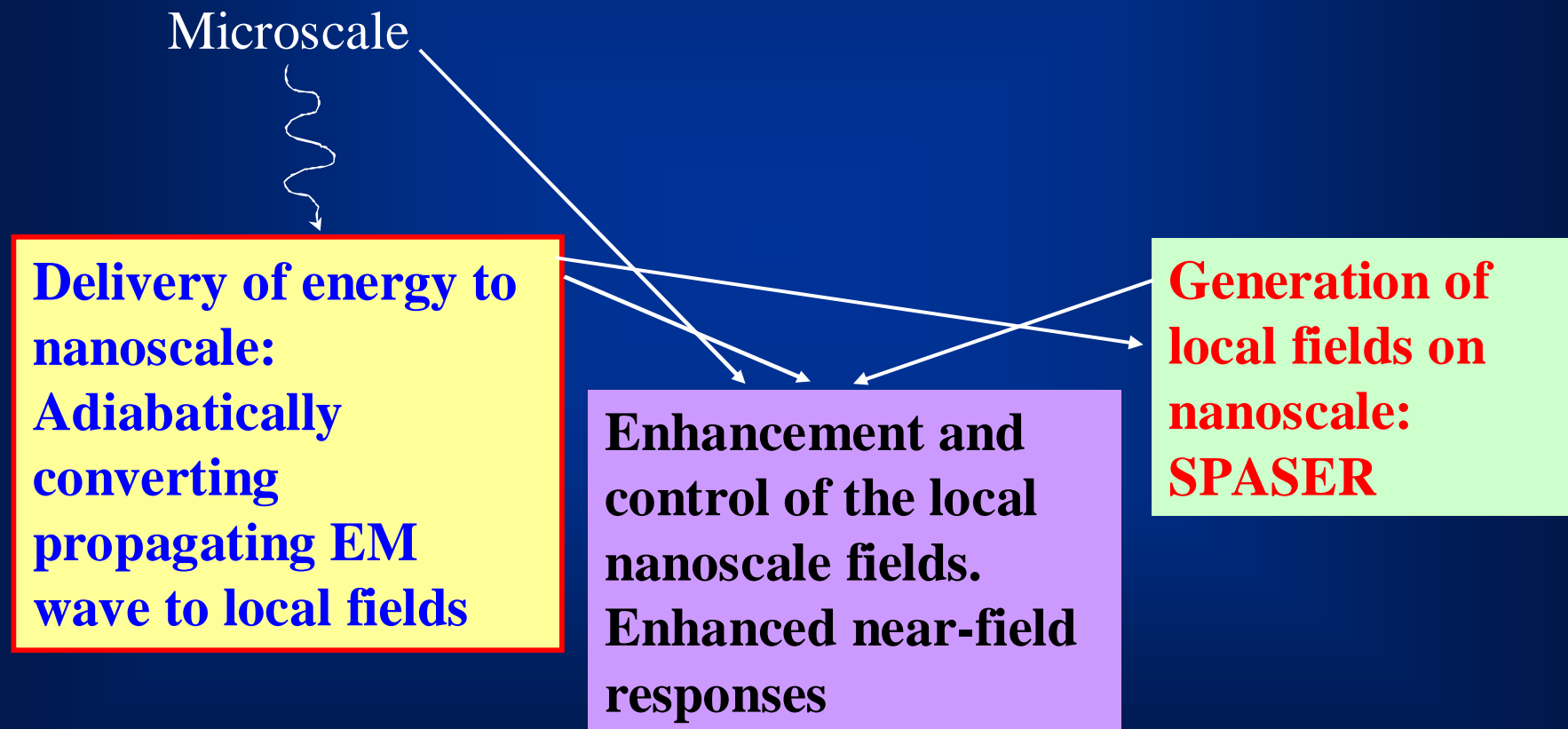
### 2. Surface plasmon polaritons as interface waves

Maxwell equation solution for metal-dielectric interface

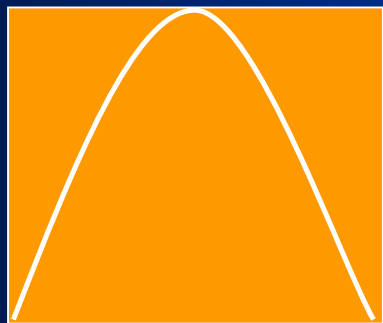
Surface plasmon polaritons in layered media

### 3. Adiabatic energy concentration in tapered plasmonic waveguides

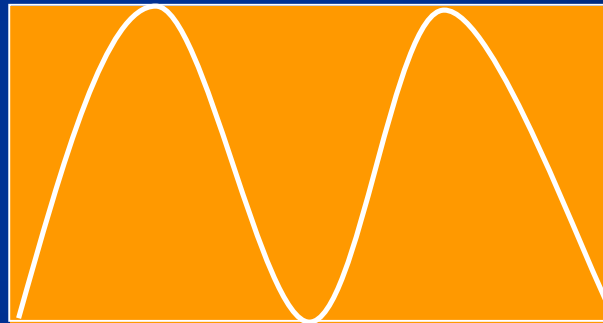
## PROBLEMS IN NANOOPTICS



## Concentration of optical (electromagnetic wave) energy



$$\frac{\lambda}{2}$$

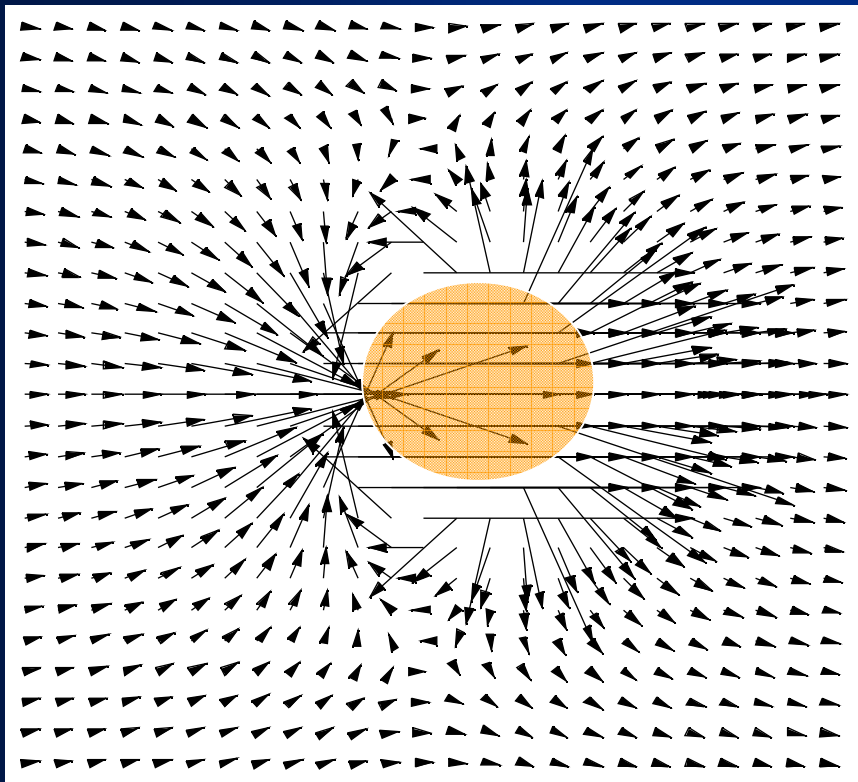


$$\lambda$$

...

# Enhanced Local Fields in Proximity of Metal Nanoparticle are Nanoscale-Localized

Local (near-zone) fields and surface plasmons

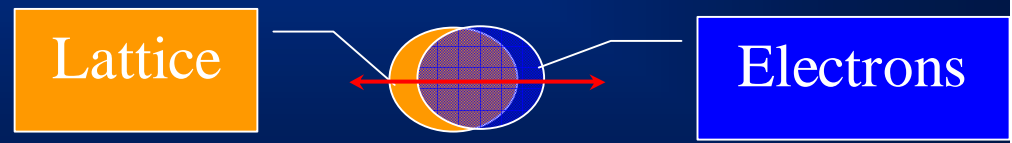


Nanoscale

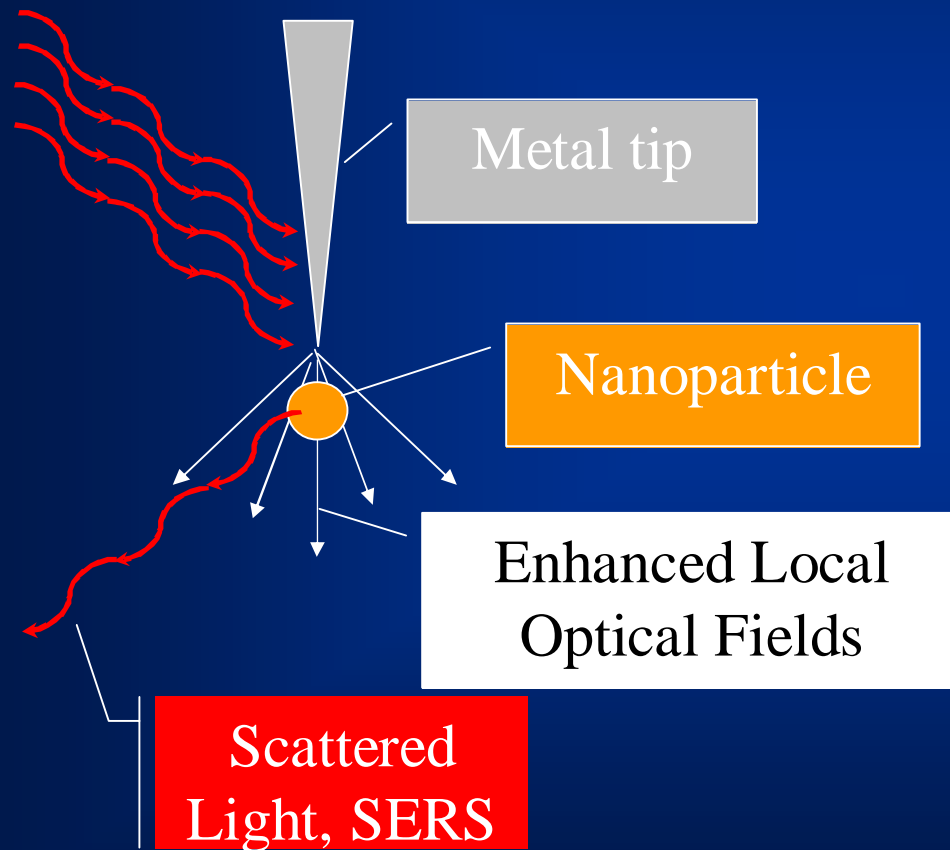
Enhancement (Quality) Factor:

$$Q = \frac{-\text{Re } \epsilon}{\text{Im } \epsilon} \sim 10$$

Surface Plasmon



## Use of Enhanced Local Fields for Nano-Microscopy



•R. Hillenbrand and F. Keilmann, *Optical Oscillation Modes of Plasmon Particles Observed in Direct Space by Phase-Contrast Near-Field Microscopy*, Appl. Phys. B **73**, 239-243 (2001).

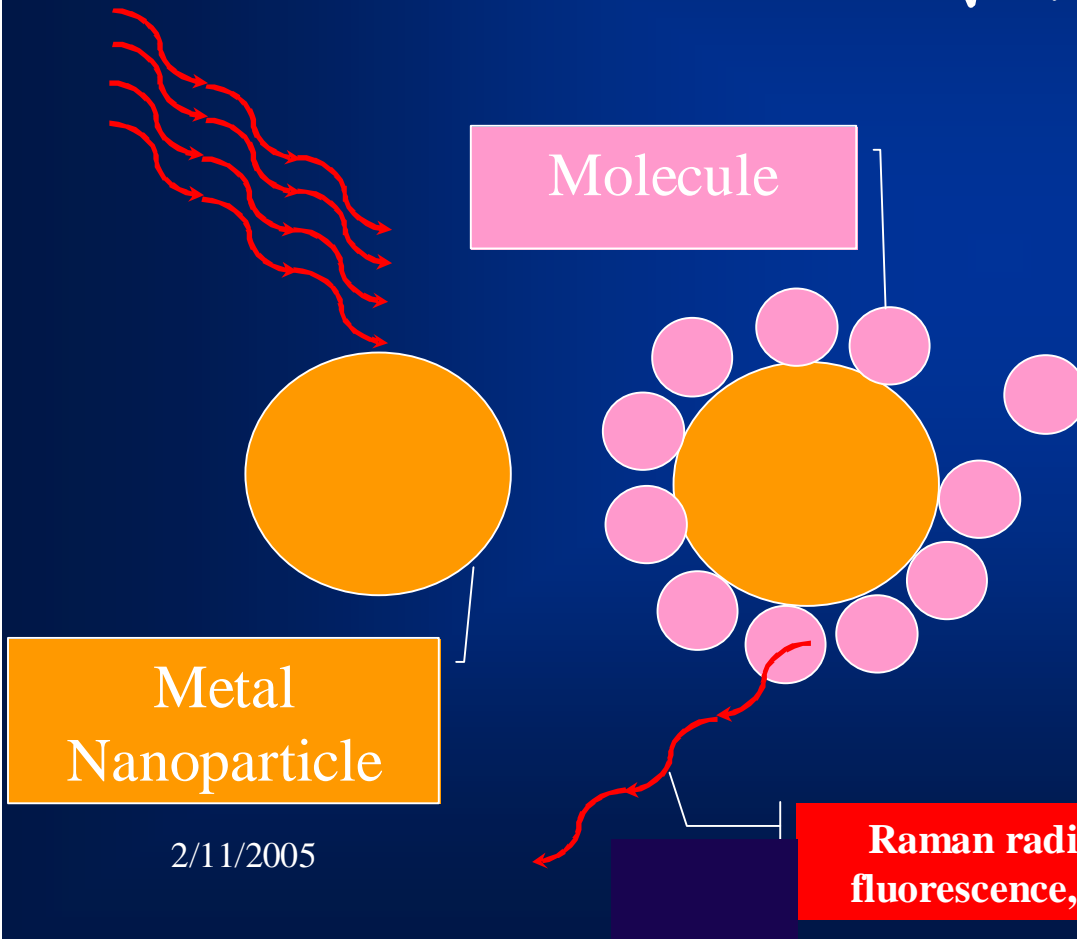
•A. Hartschuh, E. J. Sanchez, X. S. Xie, and L. Novotny, *High-Resolution Near-Field Raman Microscopy of Single-Walled Carbon Nanotubes*, Phys. Rev. Lett. **90**, 095503 -1-4 (2003).

Nanoscale



Surface plasmon frequency shifts to red upon molecules adhesion

$$\omega_p = \frac{\omega_{0p}}{\sqrt{\epsilon_h}}$$



- J. C. Riboh, A. J. Haes, A. D. McFarland, C. R. Yonzon, and R. P. Van Duyne, *A Nanoscale Optical Biosensor: Real-Time Immunoassay in Physiological Buffer Enabled by Improved Nanoparticle Adhesion*, *J. Phys. Chem. B* **107**, 1772-1780 (2003).
- C. R. Yonzon, C. L. Haynes, X. Y. Zhang, J. T. Walsh, and R. P. Van Duyne, *A Glucose Biosensor Based on Surface-Enhanced Raman Scattering: Improved Partition Layer, Temporal Stability, Reversibility, and Resistance to Serum Protein Interference*, *Anal. Chem.* **76**, 78-85 (2004).
- E. Dulkeith, A. C. Morteani, T. Niedereichholz, T. A. Klar, J. Feldmann, S. A. Levi, F. C. J. M. v. Veggel, D. N. Reinhoudt, M. Moller, and D. I. Gittins, *Fluorescence Quenching of Dye Molecules near Gold Nanoparticles: Radiative and Nonradiative Effects*, *Phys. Rev. Lett.* **89**, 203002 (2002).

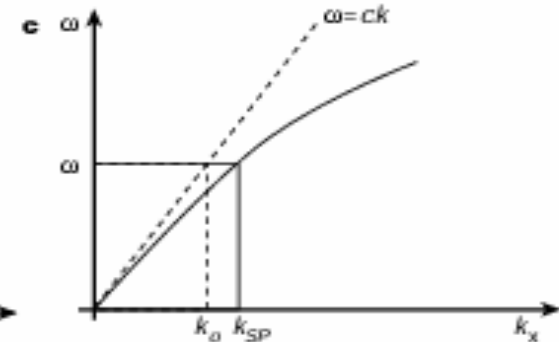
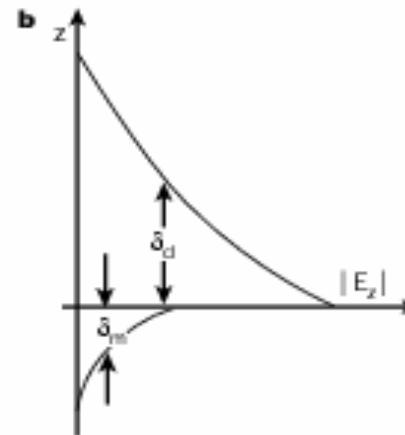
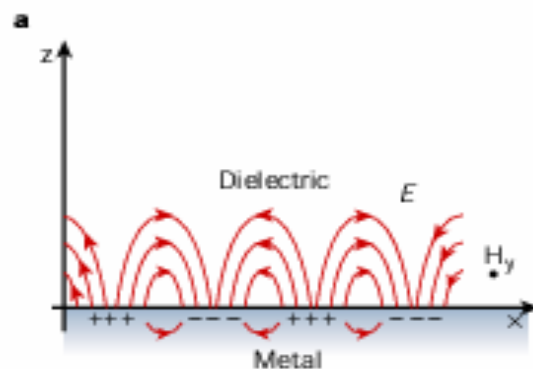
**insight review articles**

**William L. Barnes, Alain Dereux & Thomas W. Ebbesen, Nature 424, 824 (2003)**

Box 1

**Surface plasmon basics**

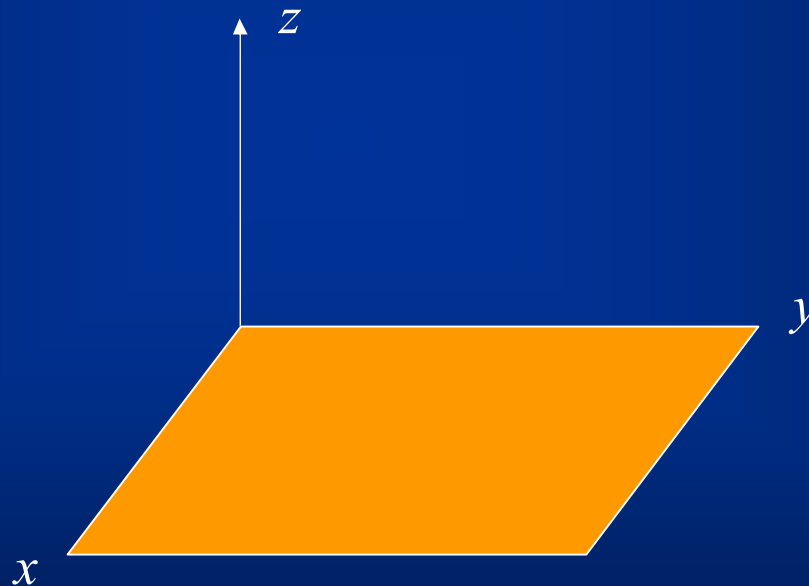
SPs at the interface between a metal and a dielectric material have a combined electromagnetic wave and surface charge character as shown in **a**. They are transverse magnetic in character ( $\mathbf{H}$  is in the  $y$  direction), and the generation of surface charge requires an electric



field normal to the surface. This combined character also leads to the field component perpendicular to the surface being enhanced near the surface and decaying exponentially with distance away from it (**b**). The field in this perpendicular direction is said to be evanescent, reflecting the bound, non-radiative nature of SPs, and prevents power from propagating away from the surface. In the dielectric medium above the metal, typically air or glass, the decay length of the field,  $\delta_d$ , is of the order of half the wavelength of light involved, whereas the decay length into the metal,  $\delta_m$ , is determined by the skin depth. **c**, The dispersion curve for a SP mode shows the momentum mismatch problem that must be overcome in order to couple light and SP modes together, with the SP mode always lying beyond the light line, that is, it has greater momentum ( $\hbar k_{SP}$ ) than a free space photon ( $\hbar k_D$ ) of the same frequency  $\omega$ .

## Guiding Equations

Assume that we have a plane interface and consider propagation in the  $xy$  plane.



Resolving the Maxwell equation into **parallel and normal (to the plane)** components, we obtain so called Guiding Equations

$$(k_0^2 \epsilon \mu - k_{\parallel}^2) \mathbf{H}_{\perp} = ik_{\parallel} \nabla_{\perp} \mathbf{H}_{\parallel} - ik_0 \epsilon \nabla_{\perp} \times \mathbf{E}_{\parallel}$$

$$(k_0^2 \epsilon \mu - k_{\parallel}^2) \mathbf{E}_{\perp} = ik_{\parallel} \nabla_{\perp} \mathbf{E}_{\parallel} + ik_0 \mu \nabla_{\perp} \times \mathbf{B}_{\parallel}$$

$$(\nabla_{\perp}^2 + k_0^2 \epsilon \mu - k_{\parallel}^2) \mathbf{E}_{\parallel} = 0$$

$$(\nabla_{\perp}^2 + k_0^2 \epsilon \mu - k_{\parallel}^2) \mathbf{H}_{\parallel} = 0$$

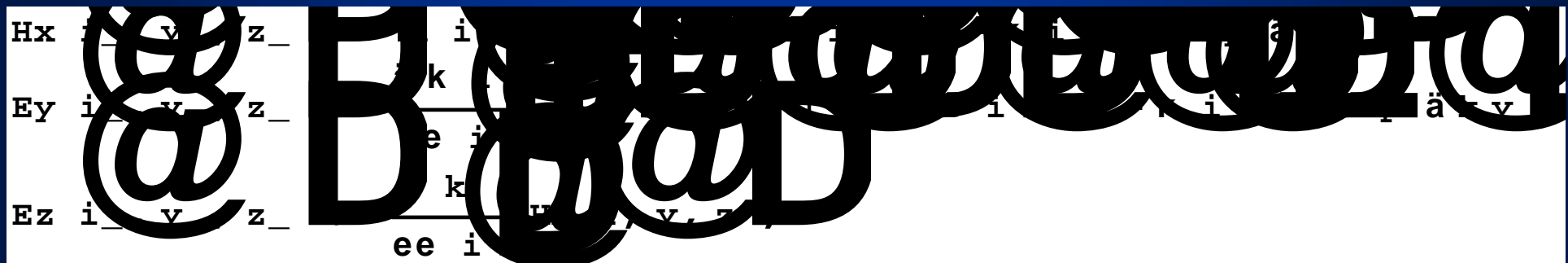
$$\text{where } k_0 \equiv \frac{\omega}{c}$$

See, e.g., J. A. Kong, *Electromagnetic wave theory*, Second ed. New York: Wiley, 1990.

Boundary conditions are continuity across the plane of

$$\mathbf{H}_{\parallel} \text{ and } \mathbf{E}_{\parallel} = \frac{i}{k_0 \epsilon} \nabla_{\perp} \times \mathbf{H}_{\perp}$$

Surface plasmon polariton (SPP) in a planar layered medium is a TM wave where in an  $i$ -th medium layer at a point  $(y, z)$  for a wave propagating in the  $y$  direction



## Metal-Dielectric Interface

For a two-medium system, the SPP wave vector is found as (dispersion relation)

$$k = \frac{\omega}{c} \sqrt{\frac{\epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2}}$$

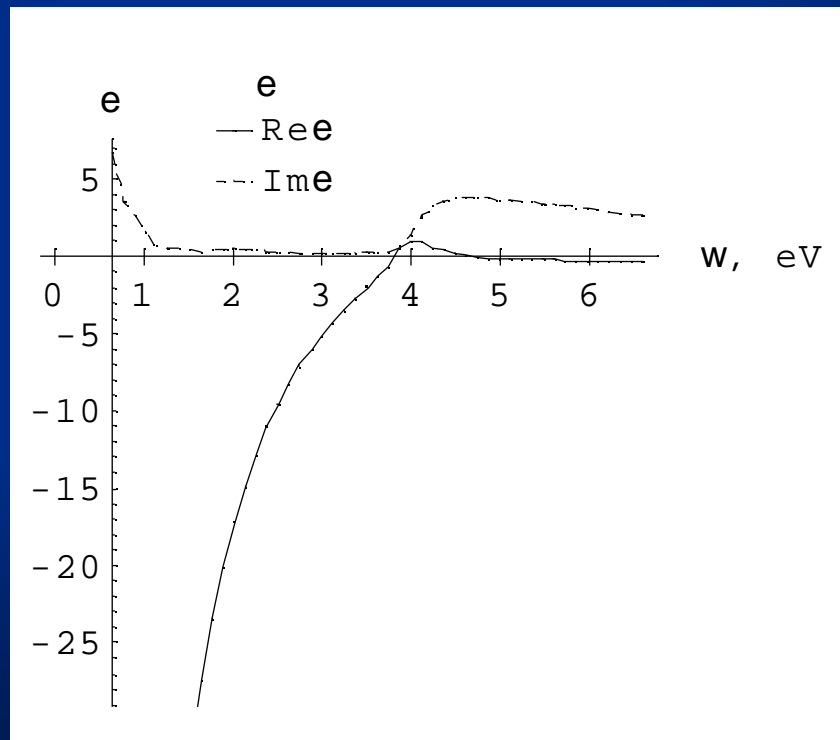
Evanescent decay decrements in these two media are found as

$$\kappa_1 = \frac{\omega}{c} \sqrt{-\frac{\epsilon_1^2}{\epsilon_1 + \epsilon_2}} \quad \kappa_2 = \frac{\omega}{c} \sqrt{-\frac{\epsilon_2^2}{\epsilon_1 + \epsilon_2}}$$

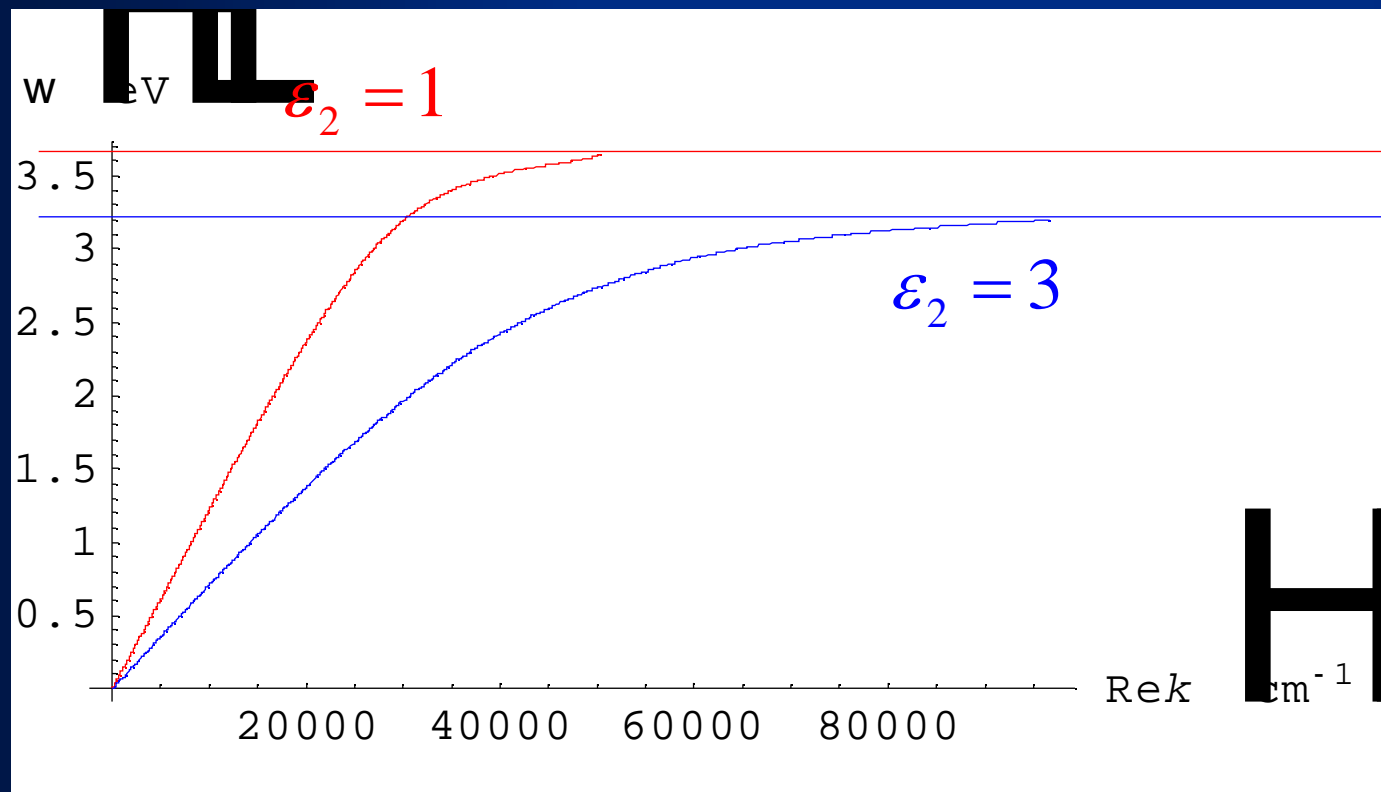
From these, it follows that for the existence of SPPs, it is necessary and sufficient that  $\epsilon_1 + \epsilon_2 < 0$  and  $\epsilon_1 \epsilon_2 < 0$

## Dielectric permittivity for silver

P. B. Johnson and R. W. Christy, "Optical-Constants of Noble-Metals," *Physical Review B* **6**, 4370-4379 (1972).



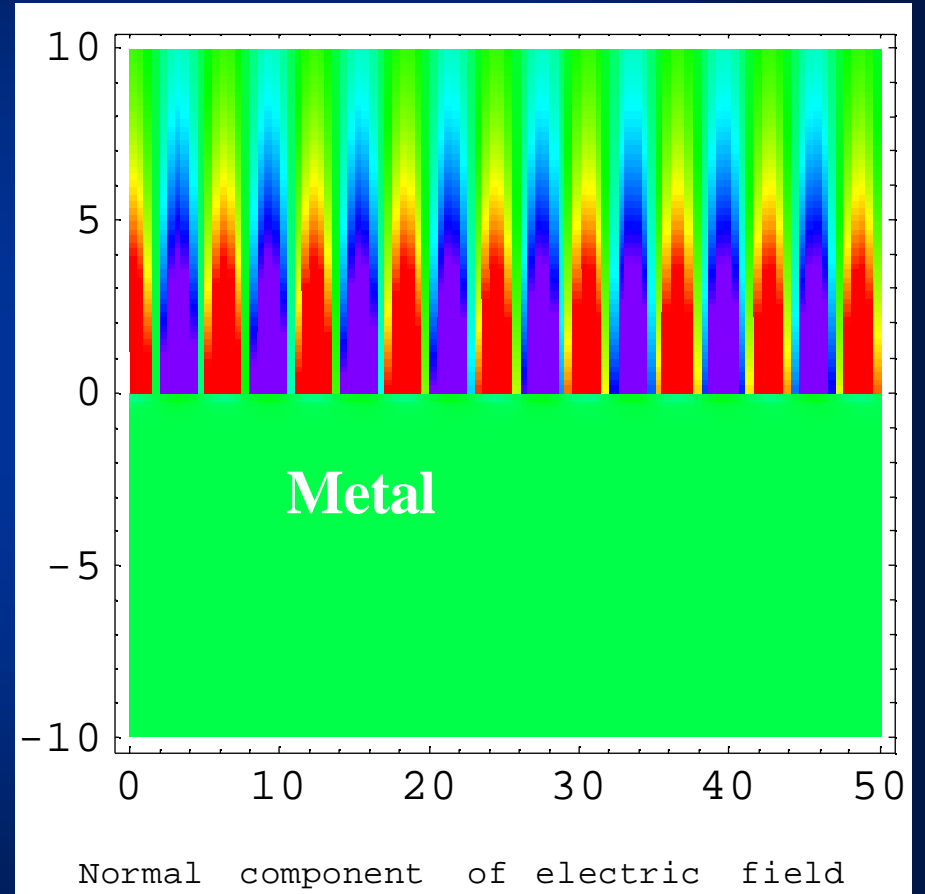
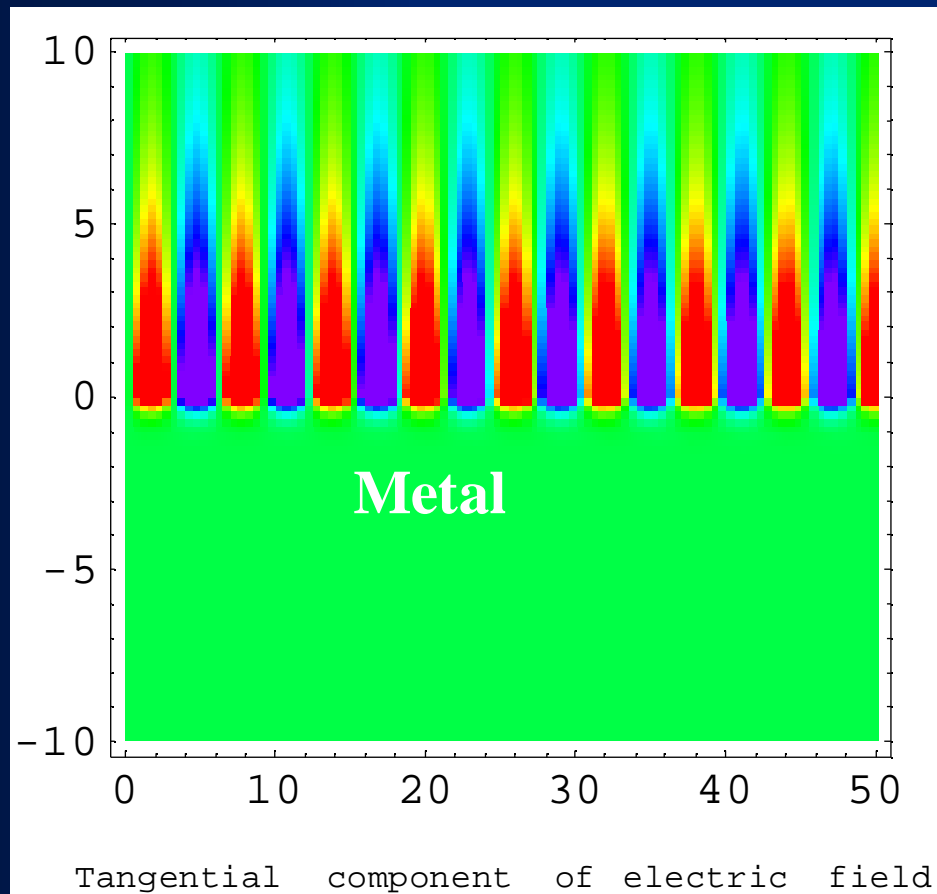
## Surface plasmon dispersion and resonance



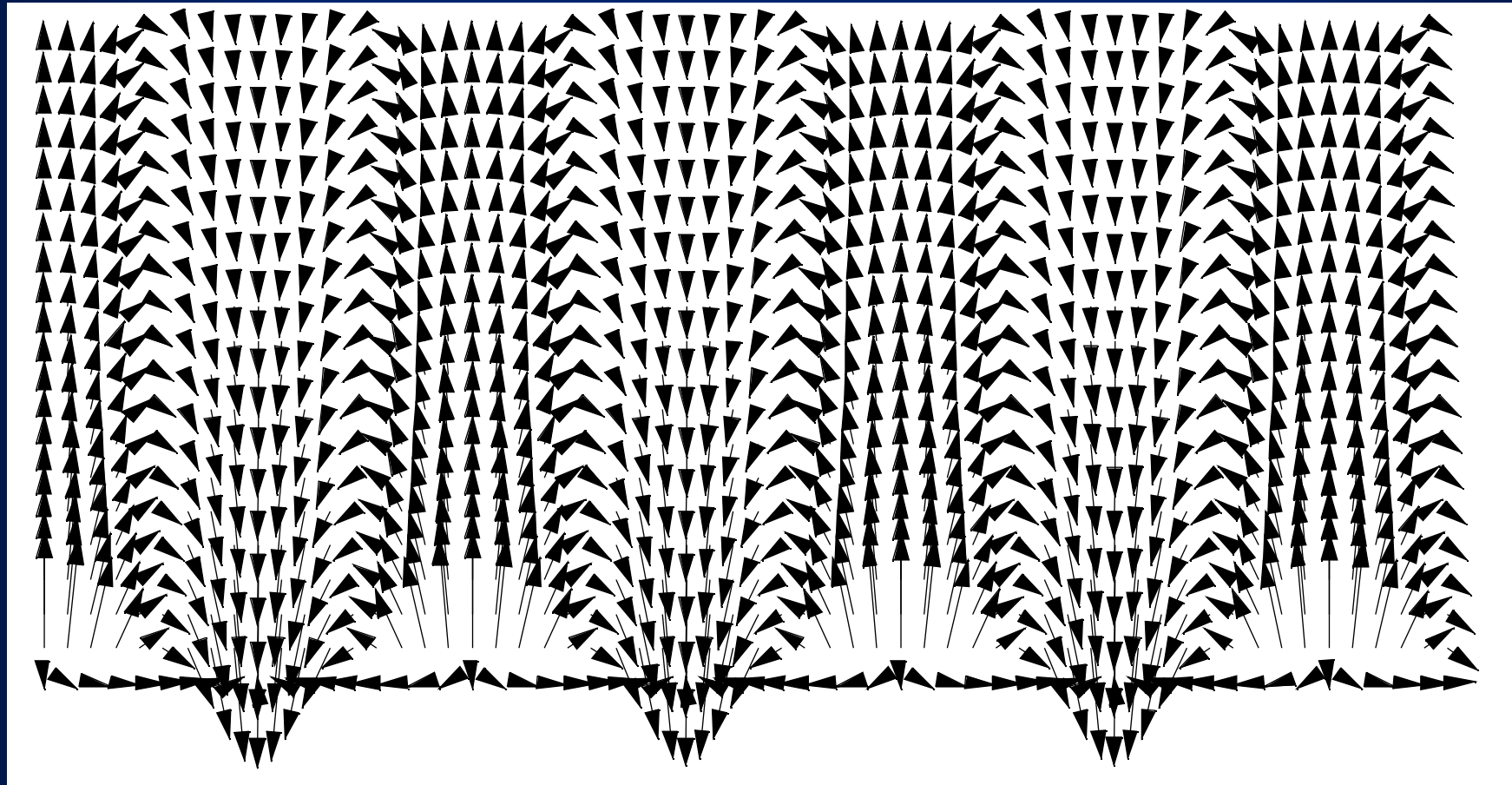
$$Re[\epsilon_1 + \epsilon_2] = 0$$



## Surface plasmon polaritons fields



## Topography of Surface Plasmon Polariton Electric Fields



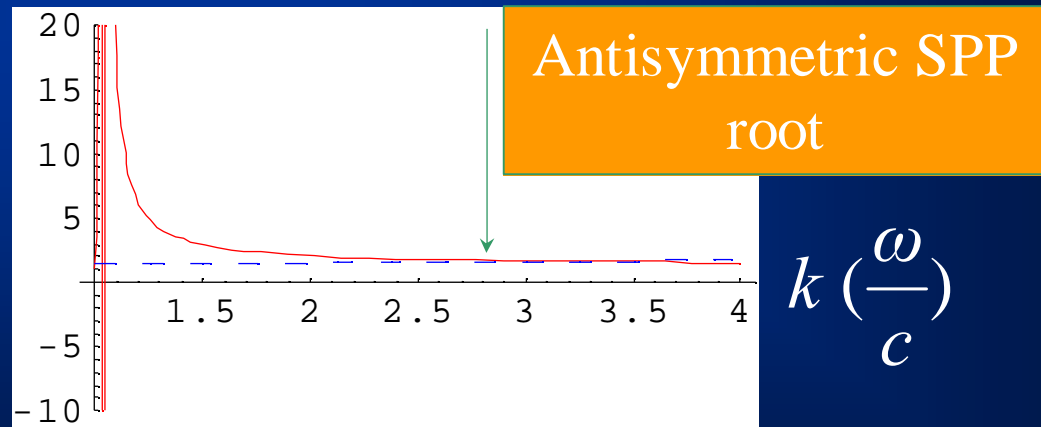
## Thin Metal Layer Between Two Semi-infinite Dielectrics

Dispersion relation (exact analytical expression), where  $Z[2]$  is the layer thickness: defines the SPP wave vector  $k$  in units of  $c/\lambda$



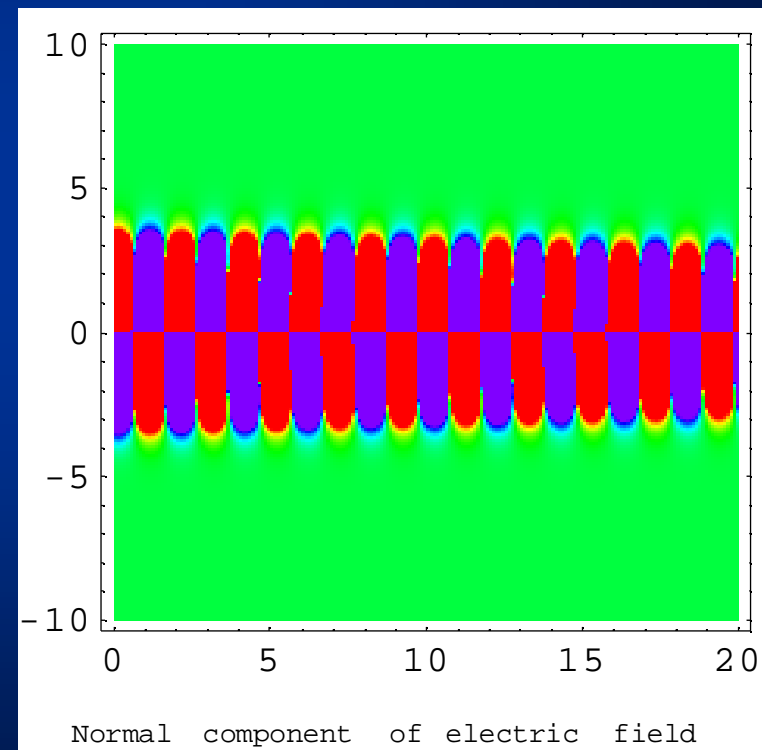
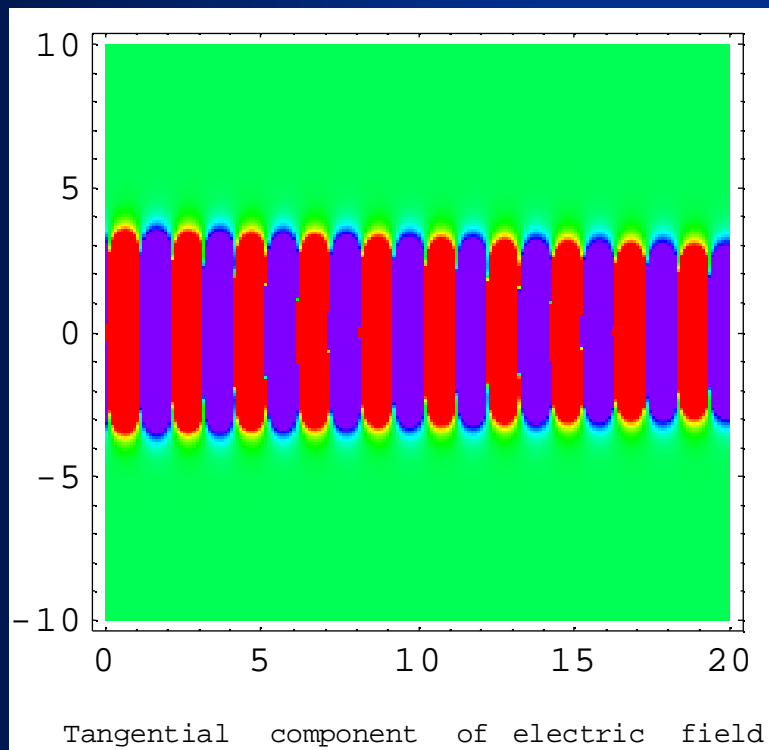
Here  $\epsilon[i]$  is dielectric permittivity of  $i$ -th layer (layer 2 is metal)

Two roots: Symmetric and Antisymmetric SPP



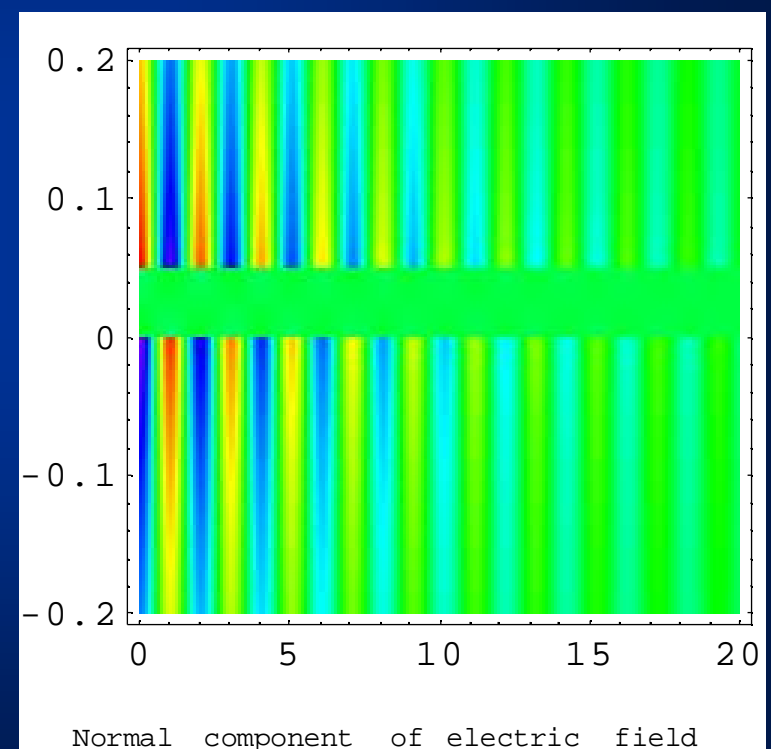
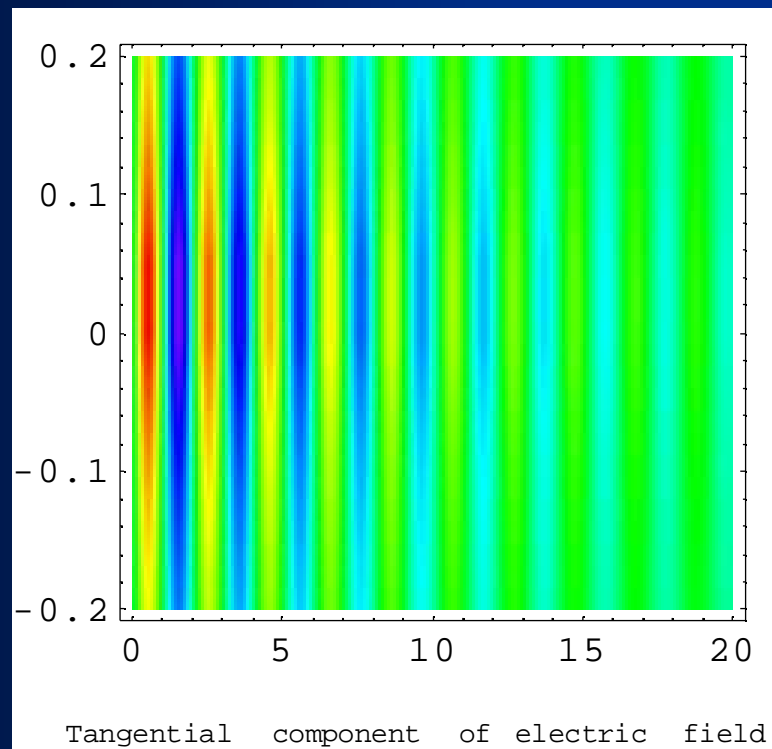
Electric field of antisymmetric (slow) SPP in plane normal to the metal layer (thickness 5 nm) at frequency 2.2 eV (wavelength 536 nm).

Spatial scales are in units of 100 nm



Electric field of antisymmetric (slow) SPP in the vicinity of the metal layer (thickness 5 nm) at frequency 2.2 eV (wavelength 536 nm).

Spatial scales are in units of 100 nm

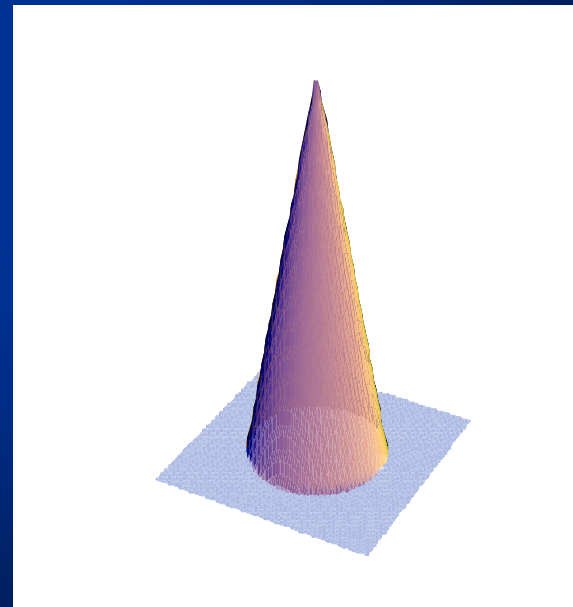


## Adiabatic Nanofocusing of Surface Plasmon Polaritons

M. I. Stockman, *Nanofocusing of Optical Energy in Tapered Plasmonic Waveguides*, *Phys. Rev. Lett.* **93**, 137404-1-4 (2004).

Waveguide geometry

↑  
Propagation direction



Electric field of SPP wave for TM<sub>00</sub> mode (magnetic field is tangential to the surface, normal to the axis; axially-symmetric solution)

$$r < R: E_z = I_0(\kappa_m r) \exp(ikz)$$

$$r > R: E_z = \frac{I_0(\kappa_m R)}{K_0(\kappa_d R)} K_0(\kappa_d r) \exp(ikz)$$

$$r < R: E_z = \frac{ik}{\kappa_m} I_1(\kappa_m r) \exp(ikz)$$

$$r > R: E_z = \frac{ik}{\kappa_d} \frac{I_0(\kappa_m R)}{K_0(\kappa_d R)} K_1(\kappa_d r) \exp(ikz)$$

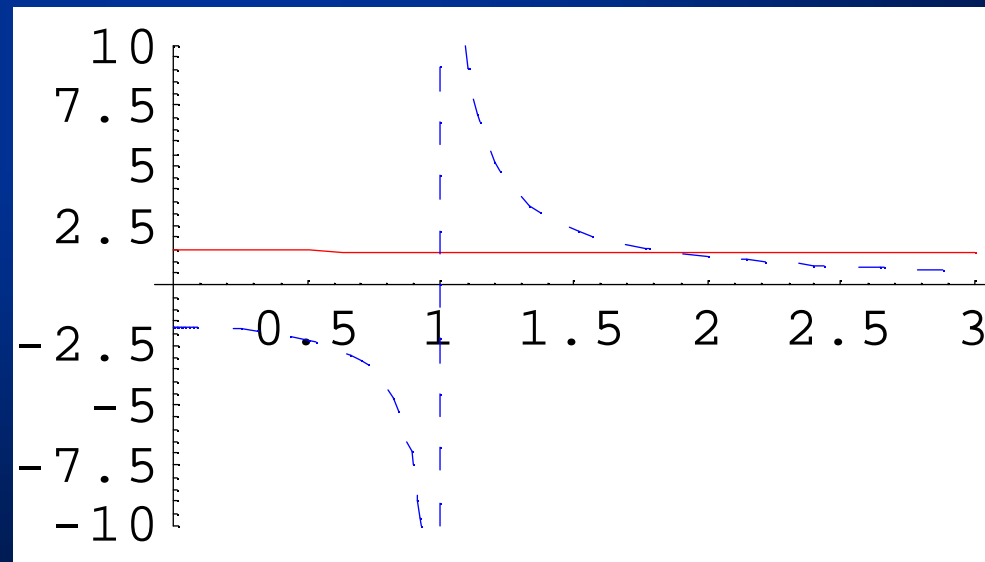
For TM<sub>00</sub> mode (magnetic field is tangential to the surface, normal to the axis; axially-symmetric solution), dispersion relation is

$$\frac{\varepsilon_m I_1(k_0 R \sqrt{k^2 - \varepsilon_m})}{\sqrt{k^2 - \varepsilon_m} I_0(k_0 R \sqrt{k^2 - \varepsilon_m})} + \frac{\varepsilon_d K_1(k_0 R \sqrt{k^2 - \varepsilon_d})}{\sqrt{k^2 - \varepsilon_d} K_0(k_0 R \sqrt{k^2 - \varepsilon_d})}$$

$$k_0 = \frac{\omega}{c}$$

There is single root:

Slow SPP





Introduce effective index:  $k = n\omega/c$

Close to the tip ( $R \rightarrow 0$ ), this effective index diverges as  $1/R$ :

$$n(R) \approx \frac{1}{k_0 R} \sqrt{-\frac{2\varepsilon_d}{\varepsilon_m} \left[ \ln \sqrt{-\frac{4\varepsilon_m}{\varepsilon_d}} - \gamma \right]^{-1}}$$

This describes slowing down and asymptotic stopping of SPP. Important, **the time to travel to the tip (singularity) of the conic waveguide logarithmically diverges,**

$$t = \frac{1}{c} \int_{R_{\max}}^R n(r) dr \sim -\ln(k_0 R) \rightarrow \infty$$

Adiabatic parameter:  $\delta = R' \frac{d\tilde{\lambda}(R)}{dR}$

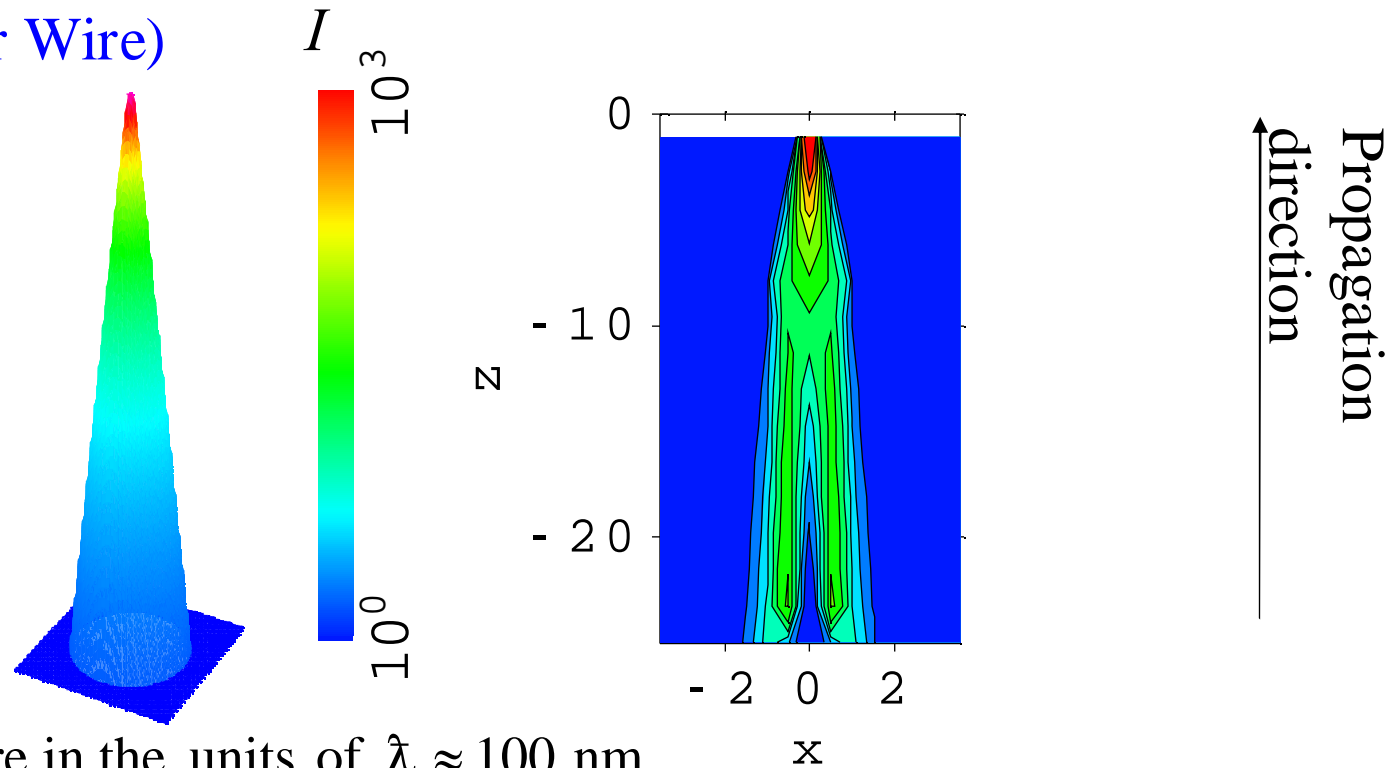
where  $R' = \frac{dR(z)}{dz}$  is the waveguide grading

For a plasmonic (TM<sub>00</sub>) mode, close to the tip

$$\delta \approx \left| R' \sqrt{-\frac{\epsilon_m}{2\epsilon_d}} \left[ \ln \sqrt{-\frac{4\epsilon_m}{\epsilon_d}} - \gamma \right] \right|$$

Thus, adiabatic parameter stays finite everywhere, including the tip.  
Correspondingly, the adiabatic (eikonal or WKB) approximation is applicable uniformly over the entire tip.

## Intensity of Local Fields at the Surface of Tapered Plasmonic Waveguide (Conic Silver Wire)



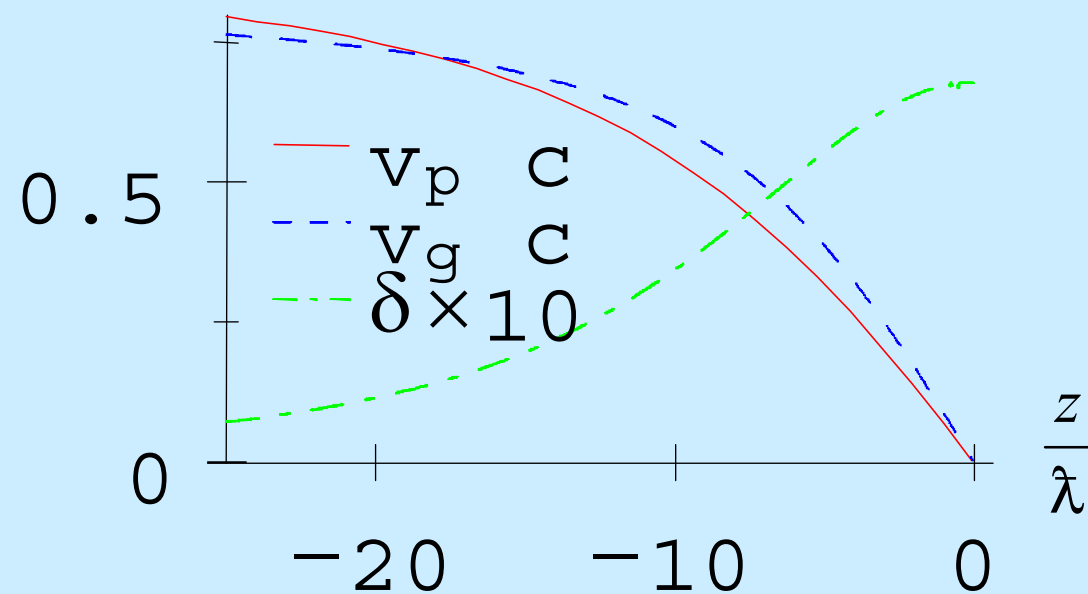
Coordinate  $s$  are in the units of  $\lambda \approx 100$  nm

M. I. Stockman, *Nanofocusing of Optical Energy in Tapered Plasmonic Waveguides*, Phys. Rev. Lett. **93**, 137404-1-4 (2004).

Phase velocity of surface plasmon polaritons

Group velocity of surface plasmon polaritons

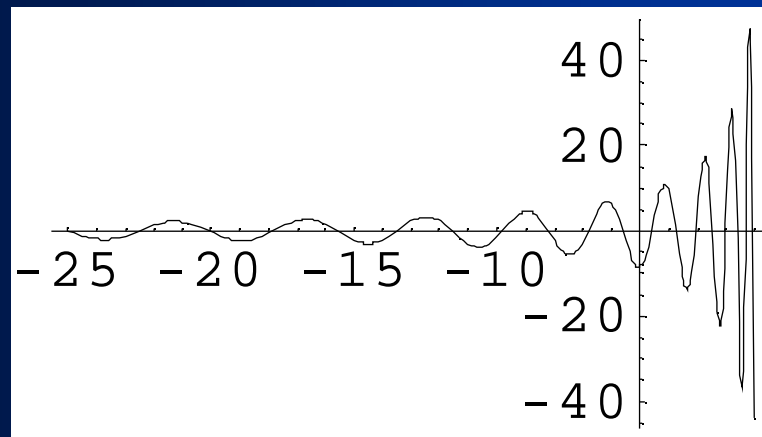
Adiabatic parameter (scaled by 10)



$\hat{\lambda} \approx 100 \text{ nm}$

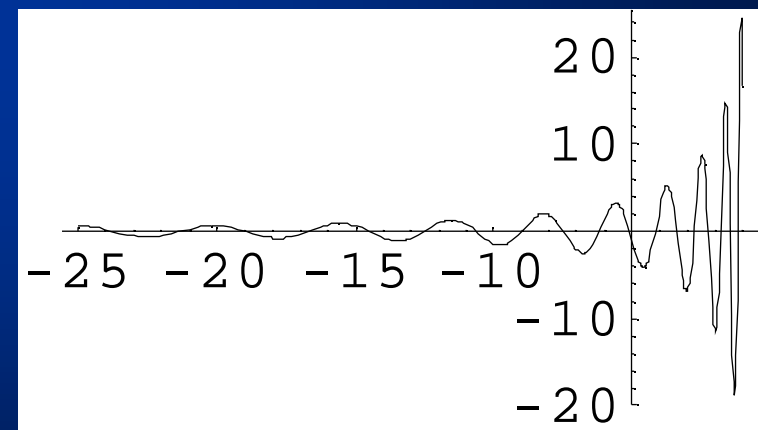
## Local Electric Fields at Surface of Plasmonic Tapered Waveguide

Transverse field



$\frac{z}{\lambda}$

Longitudinal field



$\frac{z}{\lambda}$

$\lambda \approx 100 \text{ nm}$

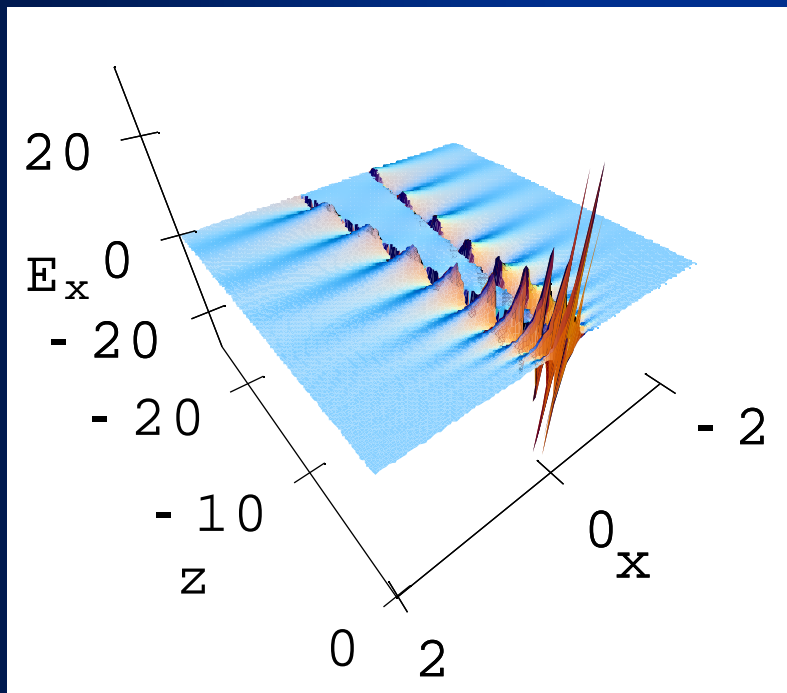
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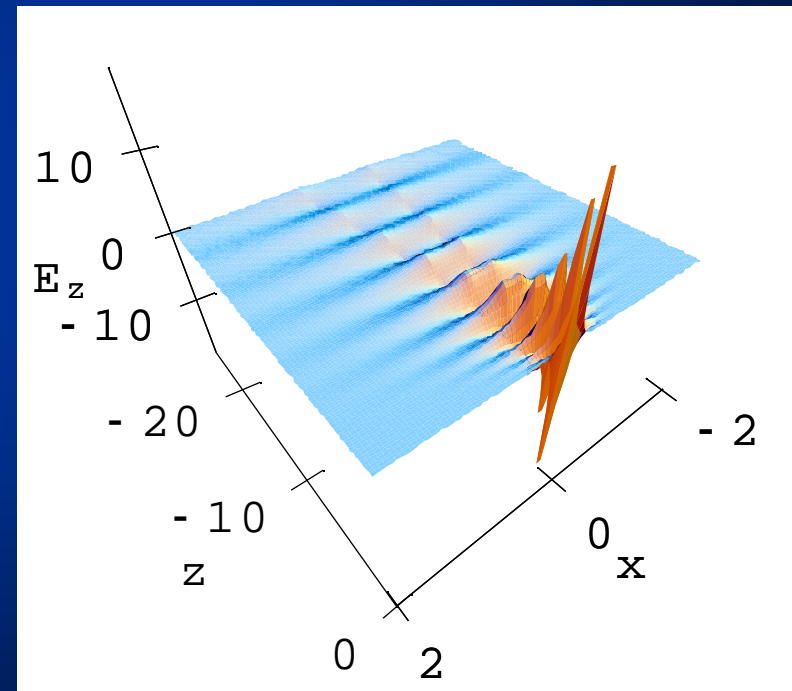
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## Local Electric Fields in Cross Section of System

Transverse electric field

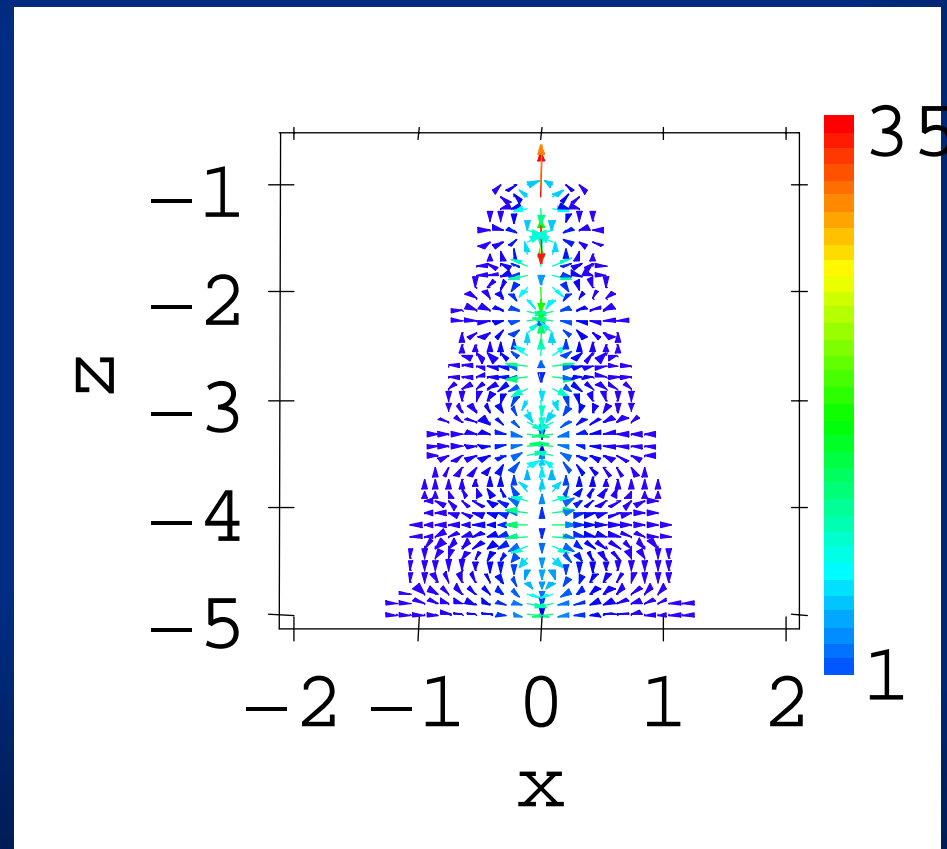


Longitudinal electric field



Coordinate  $s$  are in the units of  $\lambda \approx 100$  nm

## Vector of optical electric field for TM<sub>00</sub> plasmonic mode of conic waveguide made of silver



Spatial scales are in  
units of 100 nm

## Adiabatic Nanofocusing Conclusions

Due to adiabaticity, the back reflection of SPP is minimal.

The high wave vector of the TM<sub>00</sub> SPP makes them dark (no coupling to the far field radiation).

The velocity of SPP tends to zero  $\sim R$  as they approach the tip: *adiabatic slowing down and asymptotic stopping*.

This leads to the accumulation of the SPP near the tip and their adiabatic nanofocusing.

Under realistic conditions it is possible to transfer to the tip vicinity  $\sim 50\%$  of the initial energy flux, that along with adiabatic stopping leads to the local field-intensity enhancement by three orders of magnitude