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SMR: 1643/4

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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Support:

Theory of Nanoplasmonics 1: Optical Properties of Plasmonic Nanosystems

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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

Microscale

Delivery of energy to nanoscale: Adiabatically converting propagating EM wave to local fields

Enhancement and control of the local nanoscale fields. Enhanced near-field responses

Generation of local fields on nanoscale: SPASER

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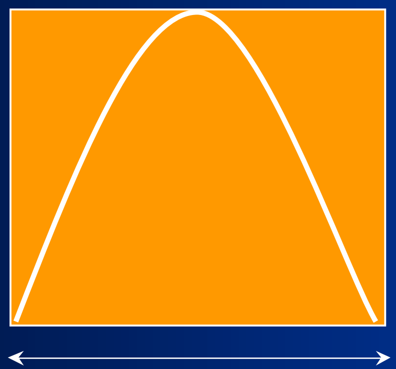
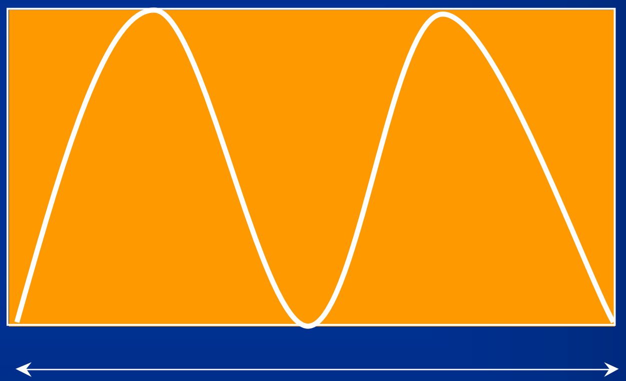
Concentration of optical **(electromagnetic wave)** energy



$\frac{\lambda}{2}$



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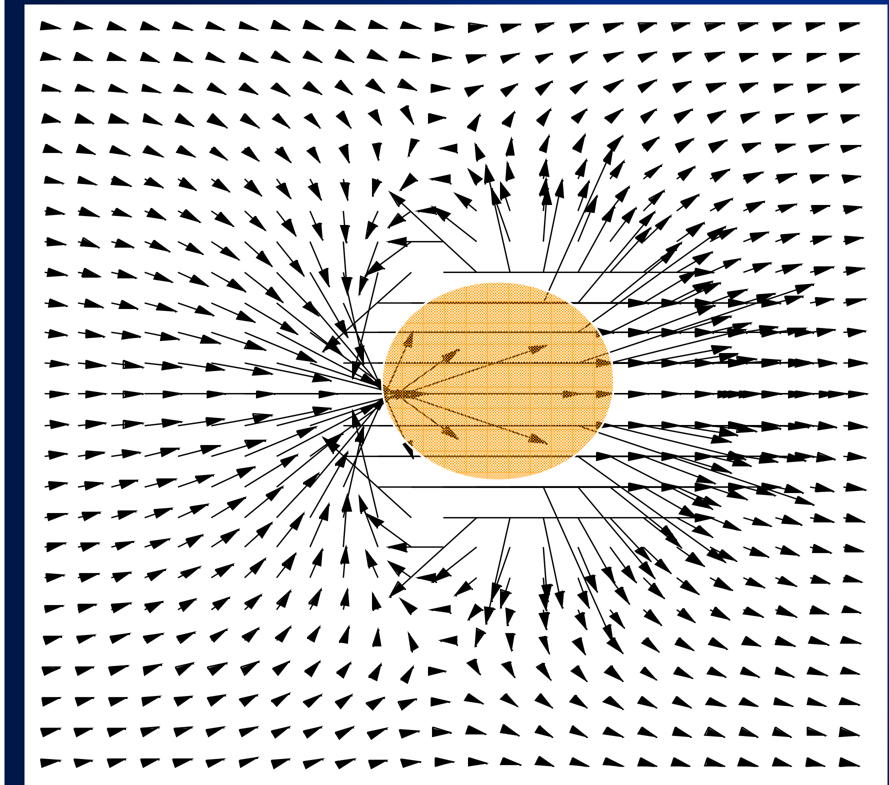
Local (near-zone) fields and surface plasmons

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

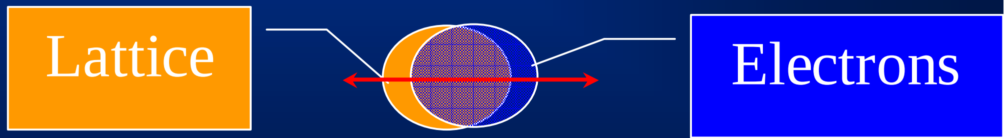


Nanoscale

Enhancement (Quality) Factor:

$$Q = \frac{-\mathrm{Re}\,\varepsilon}{\mathrm{Im}\,\varepsilon} \sim 10$$

Surface Plasmon



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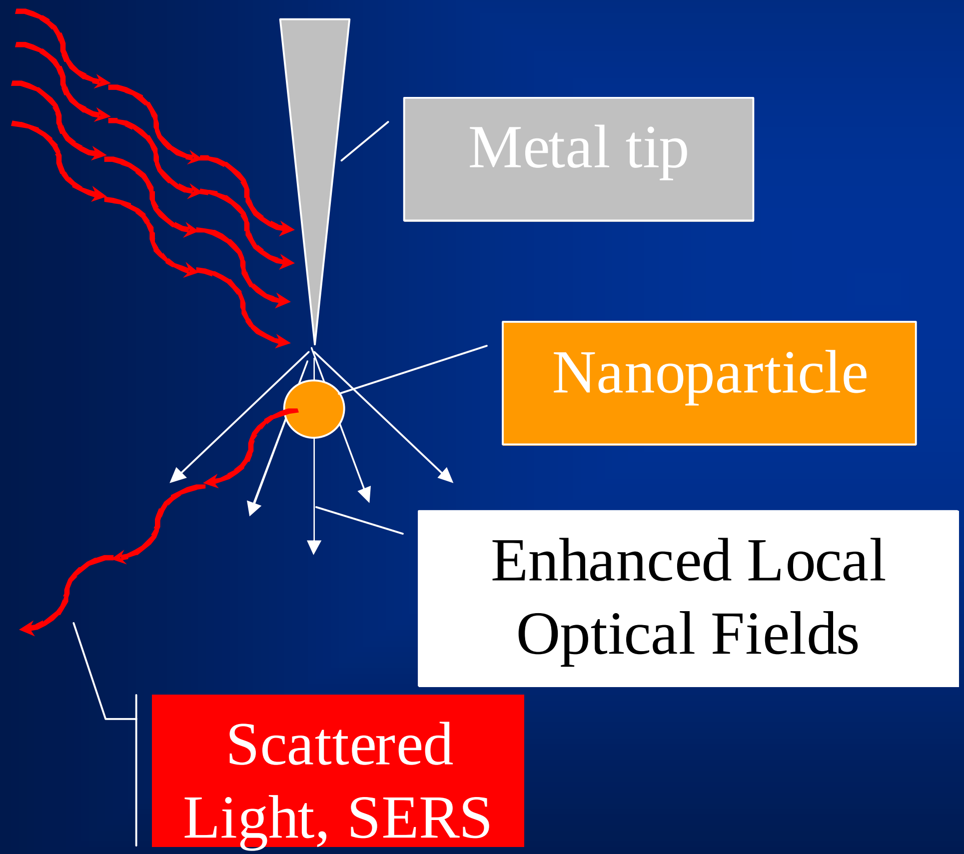
Use of Enhanced Local Fields for Nano-Microscopy



•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

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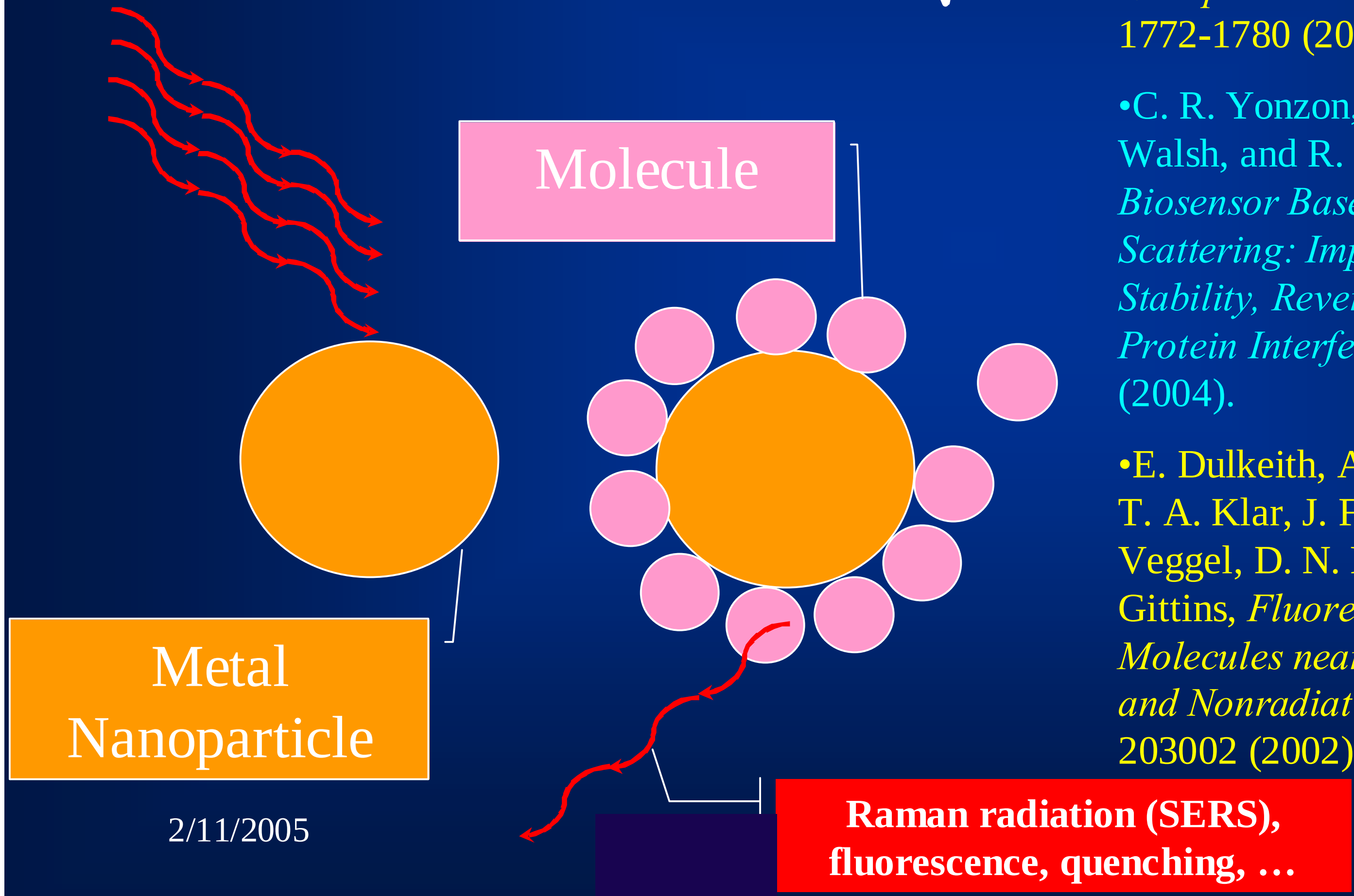
Nanosensors based on enhanced local fields

Surface plasmon frequency shifts to red upon molecules adhesion $\omega_p = \dfrac{\omega_{0p}}{\sqrt{\varepsilon_h}}$



•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).

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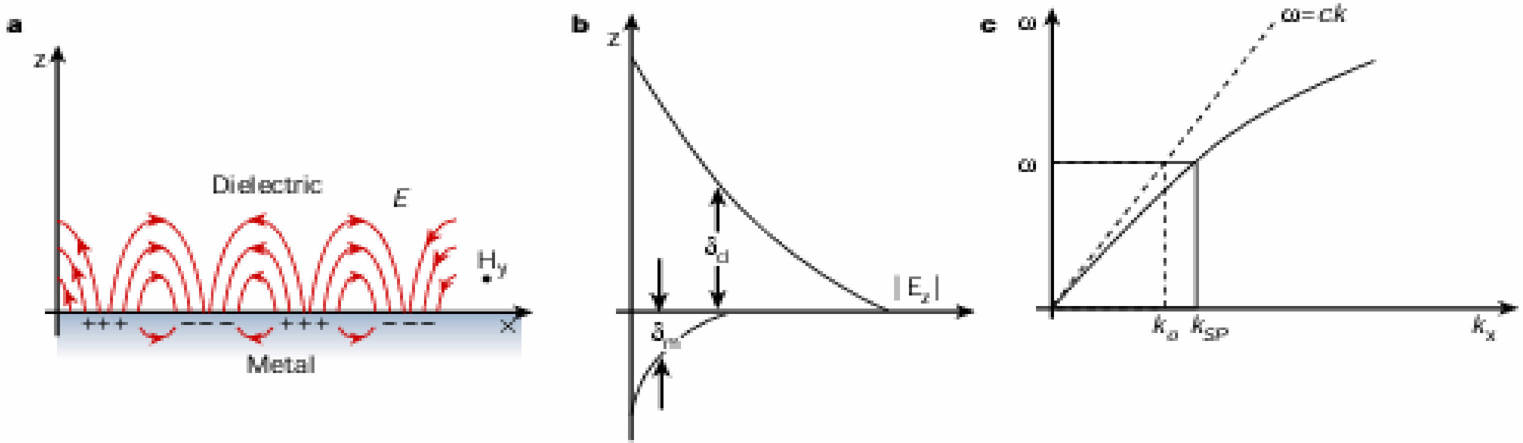
insight review articles

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

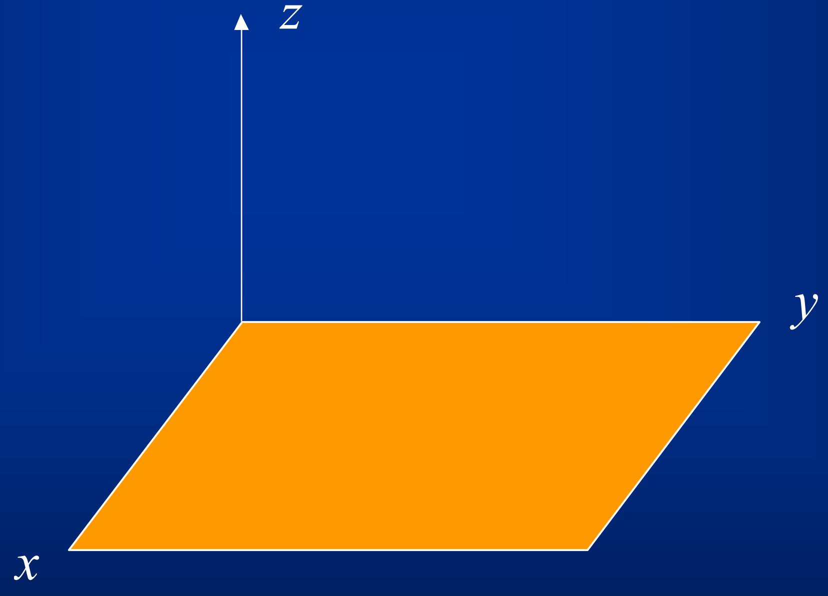
Box 1
Surface plasmon basics



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 (**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, δ_d, is of the order of half the wavelength of light involved, whereas the decay length into the metal, δ_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 ($\hbar k_0$) of the same frequency ω.

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

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

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

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

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

where $k_0 \equiv \dfrac{\omega}{c}$

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

Boundary conditions are continuity across the plane of

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

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)

$$k = \frac{\omega}{c}\sqrt{\frac{\varepsilon_1\varepsilon_2}{\varepsilon_1+\varepsilon_2}}$$

Evanescent decay decrements in these two media are found as

$$\kappa_1 = \frac{\omega}{c}\sqrt{-\frac{\varepsilon_1^2}{\varepsilon_1+\varepsilon_2}} \qquad \kappa_2 = \frac{\omega}{c}\sqrt{-\frac{\varepsilon_2^2}{\varepsilon_1+\varepsilon_2}}$$

From these, it follows that for the existence of SPPs, it is necessary and sufficient that $\varepsilon_1+\varepsilon_2<0$ and $\varepsilon_1\varepsilon_2<0$

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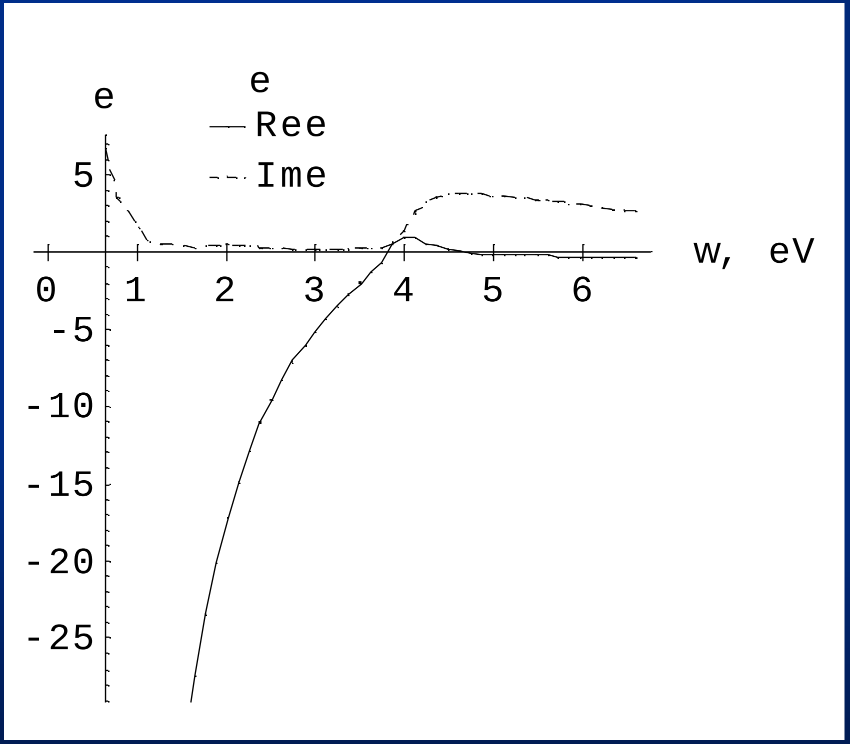
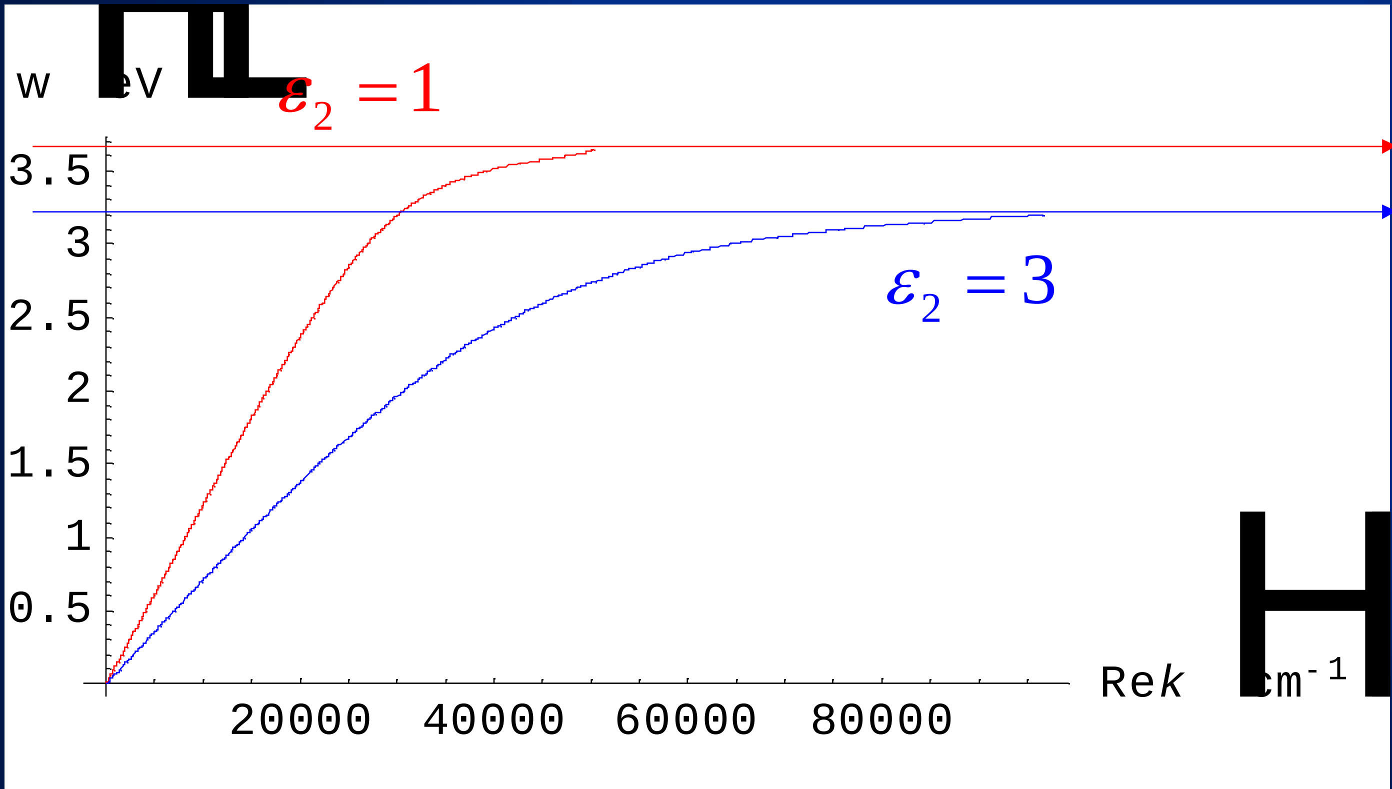
Dielectric permittivity for silver

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



Surface plasmon dispersion and resonance



$$\mathrm{Re}[\varepsilon_1 + \varepsilon_2] = 0$$

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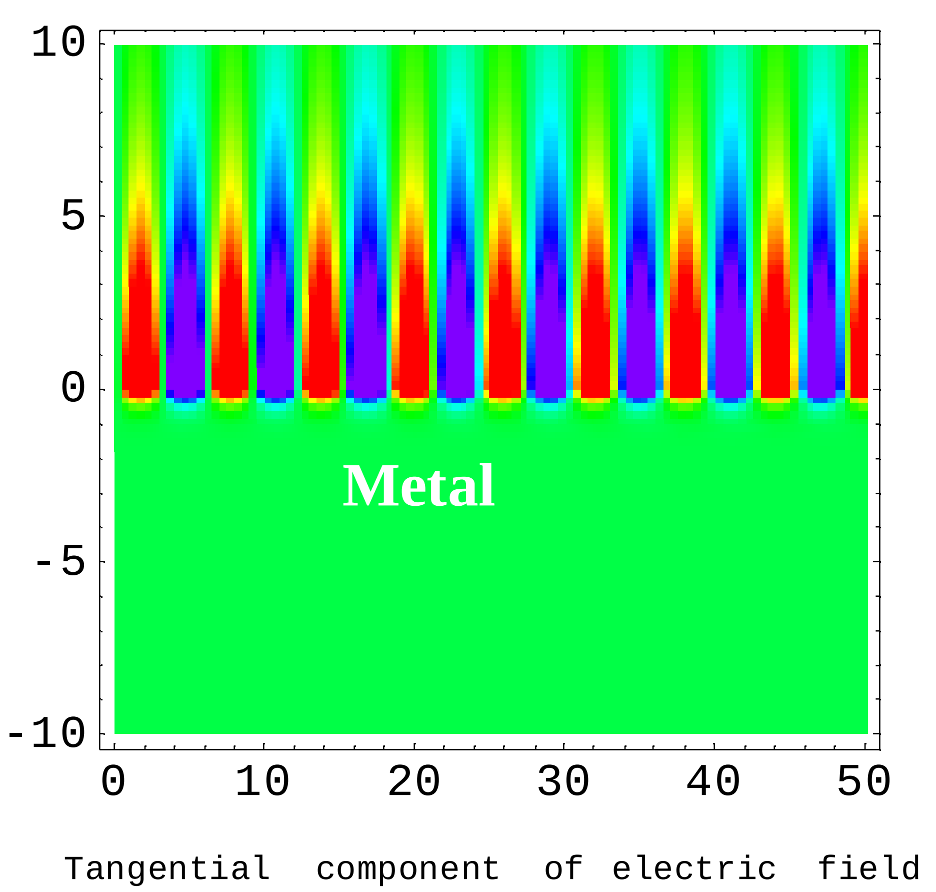
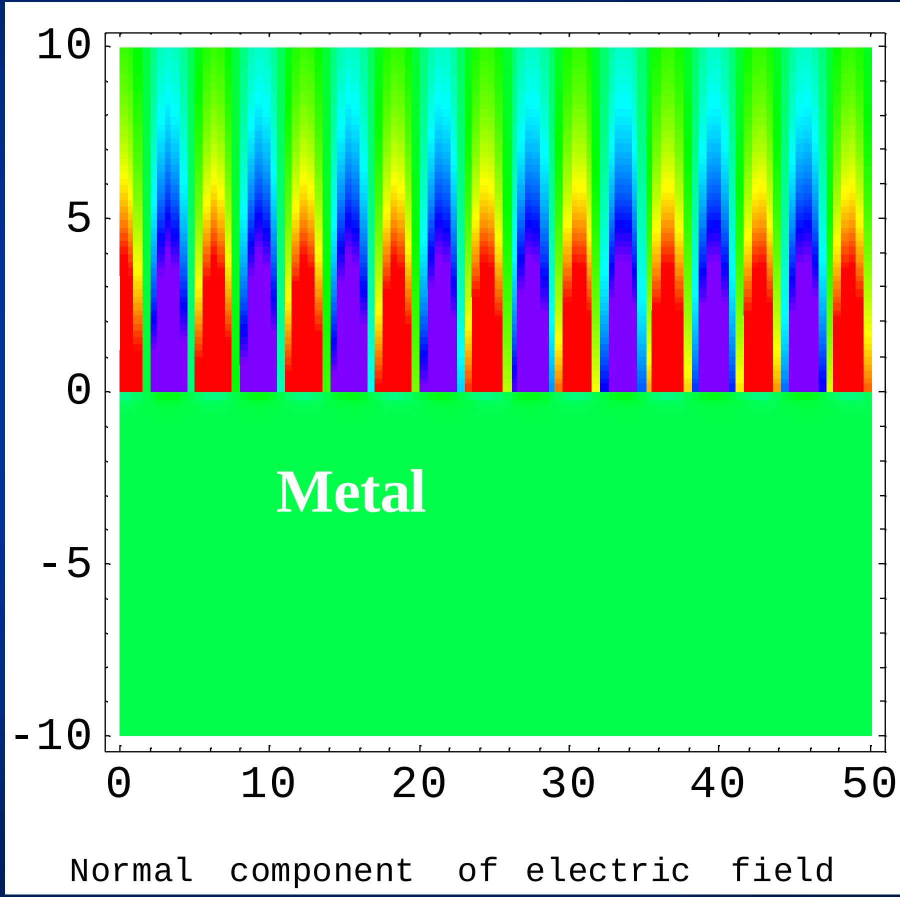
Surface plasmon polaritons fields





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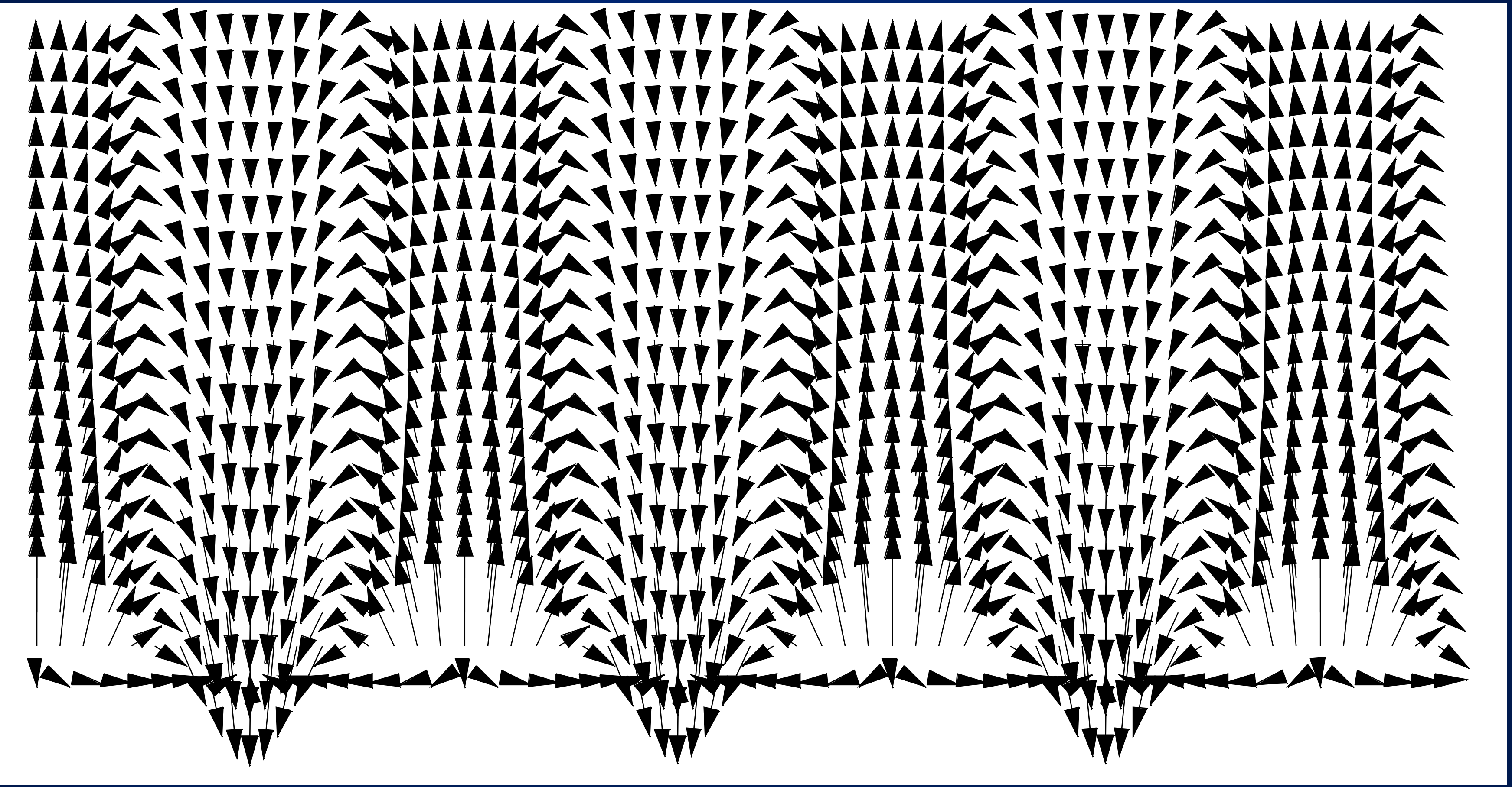
Topography of Surface Plasmon Polariton Electric Fields

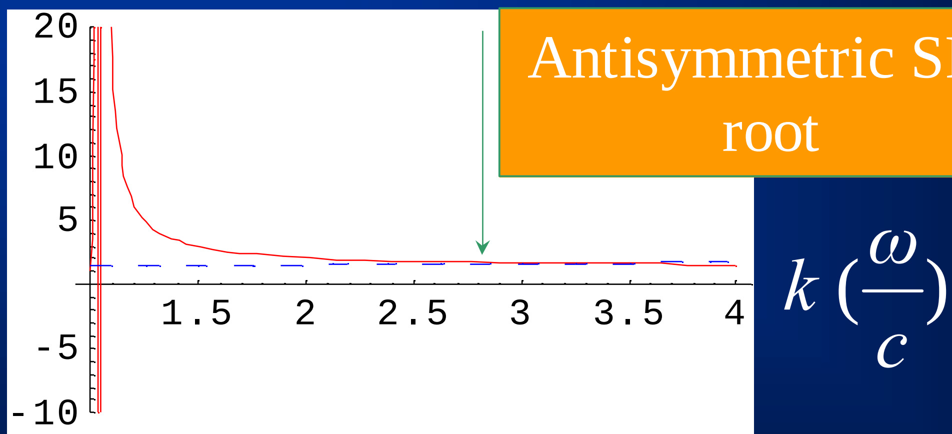


Thin Metal Layer Between Two Semi-infinite Dielectrics

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

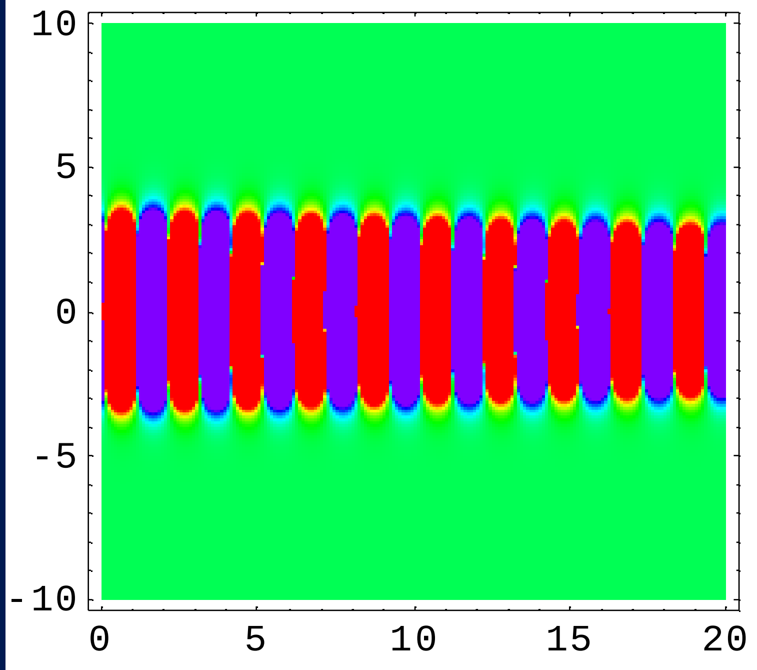
Here $\varepsilon\varepsilon[i]$ is dielectric premittivity of i - th layer (layer 2 is metal)

Two roots: Symmetric and Antisymmetric SPP

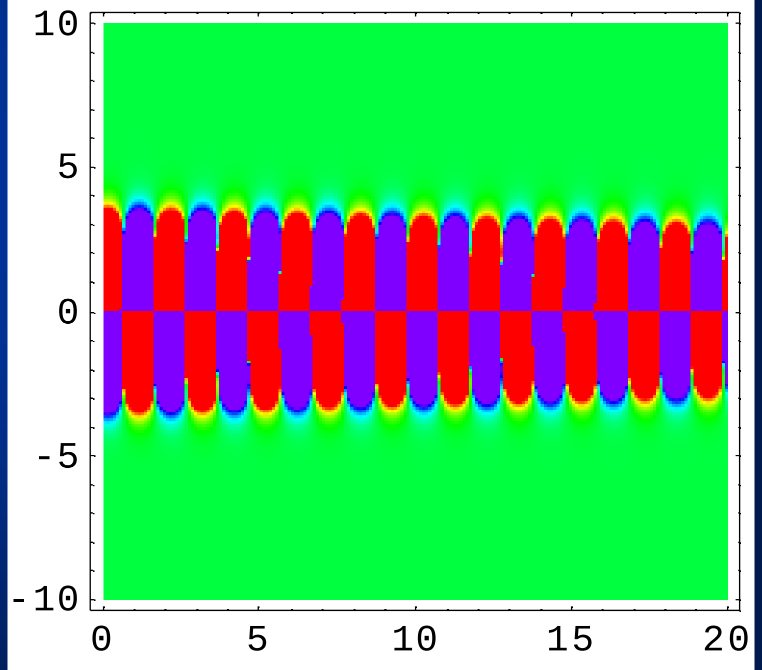


Electric field of antisymmetric (slow) SPP in plane normal to the metal layer (thickness 5 nm) at frequency 2.2 eV (wavelength 536 nm).

Spatial scales are in units of 100 nm



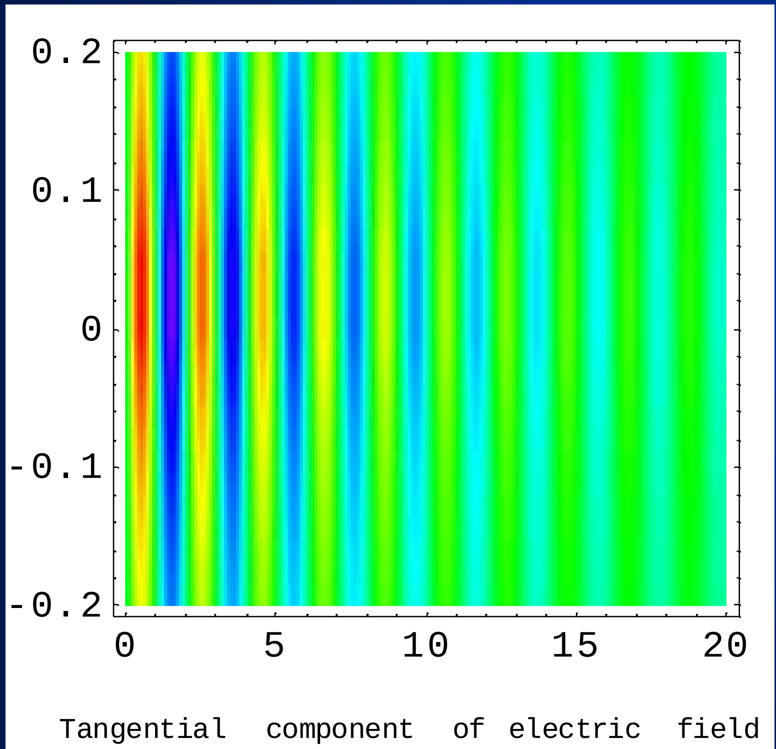
Tangential component of electric field

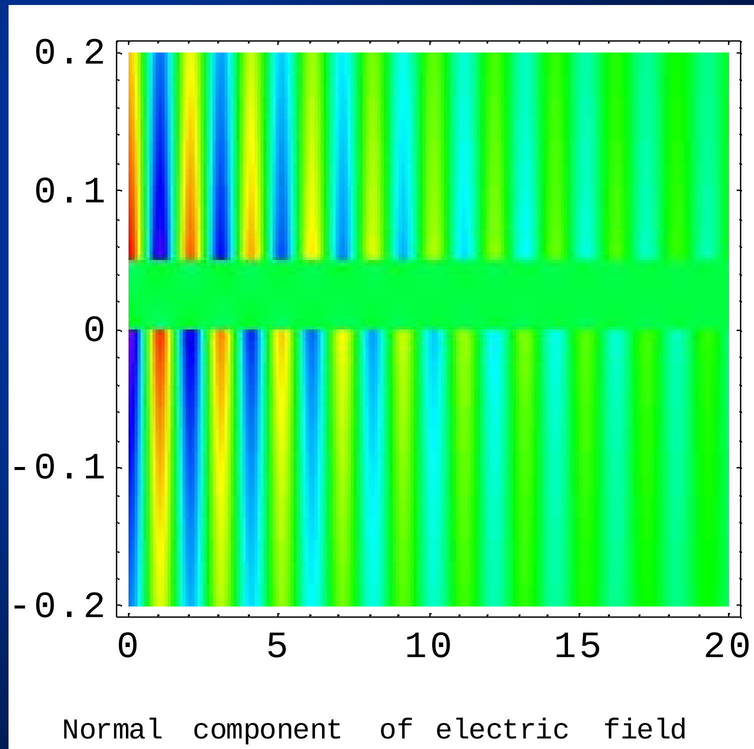


Normal component of electric field

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



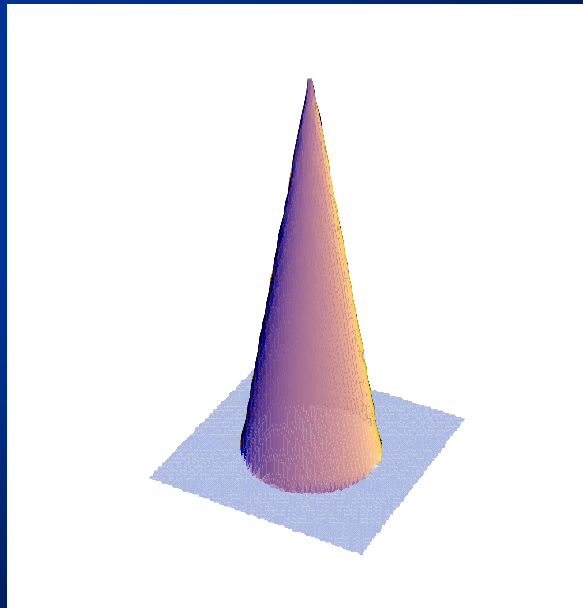


Adiabatic Nanofocusing of Surface Plasmon Polaritons

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

Waveguide geometry

Propagation direction



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For Cylindrical Plasmonic Waveguide

Electric field of SPP wave for TM00 mode (magnetic field is tangential to the surface, normal to the axis; axially-symmetric solution)

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

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

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

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

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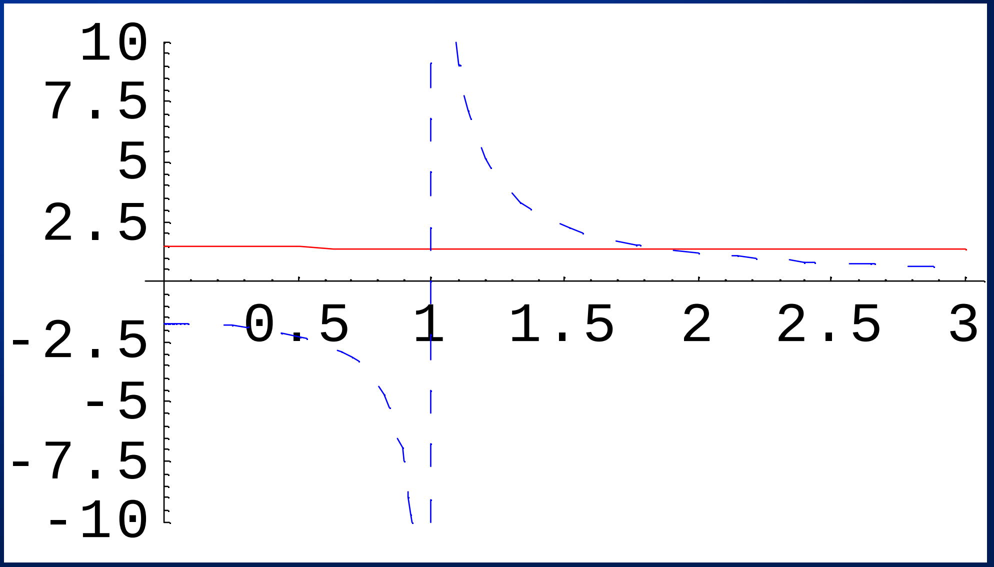
For TM00 mode (magnetic field is tangential to the surface, normal to the axis; axially-symmetric solution), dispersion relation is

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

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

There is single root:

Slow SPP



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

Close to the tip ($R \to 0$), 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,

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

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

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

For a plasmonic (TM00) mode, close to the tip

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

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

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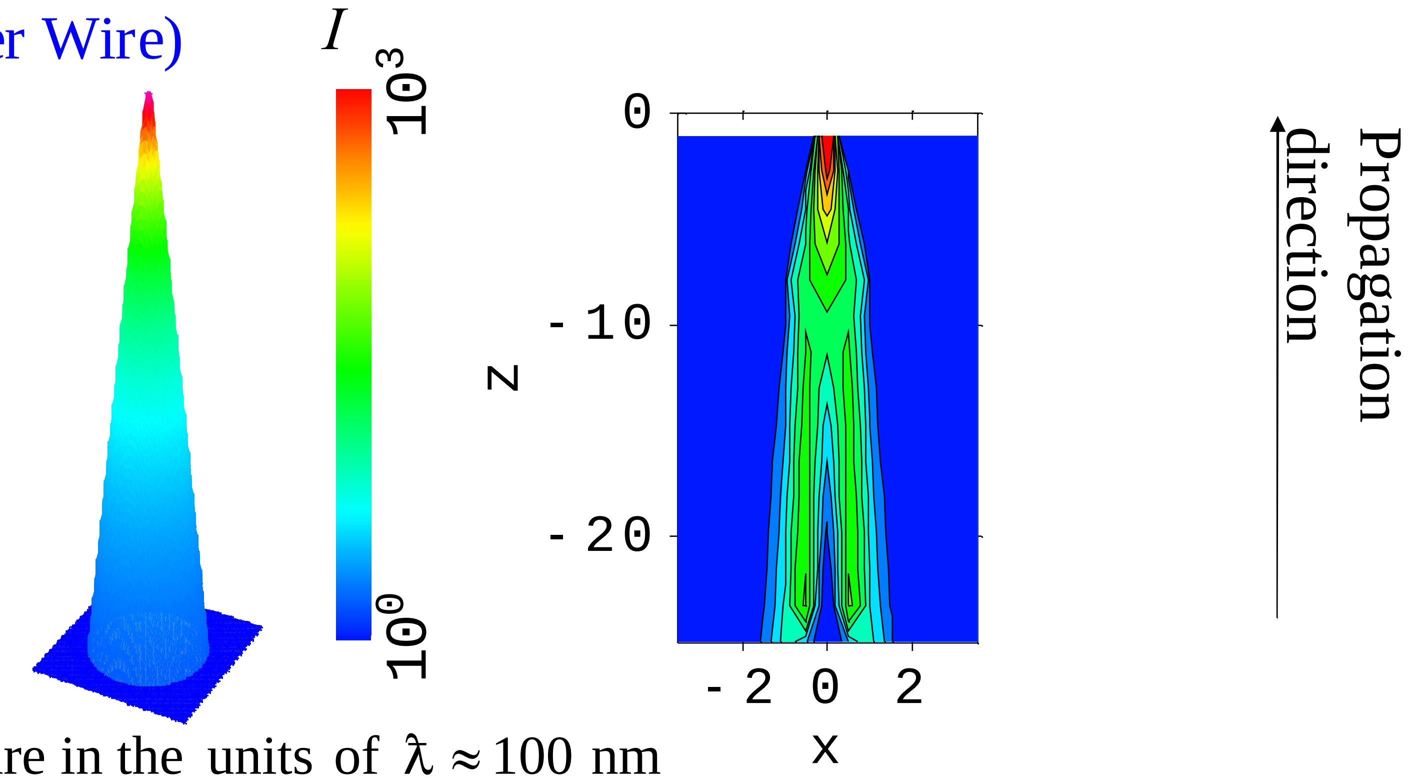
Adiabatic Nanofocusing in Tapered Nanoplasmonic Waveguides

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



Coordinates are in the units of $\bar{\lambda} \approx 100$ nm

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

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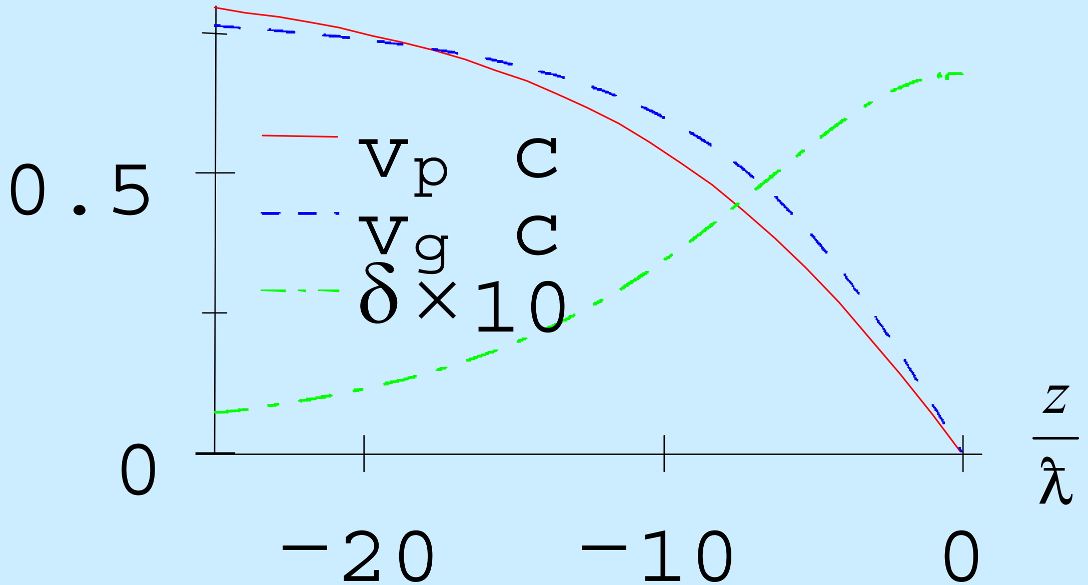
Phase velocity of surface plasmon polaritons

Group velocity of surface plasmon polaritons

Adiabatic parameter (scaled by 10)



$\lambda \approx 100$ nm

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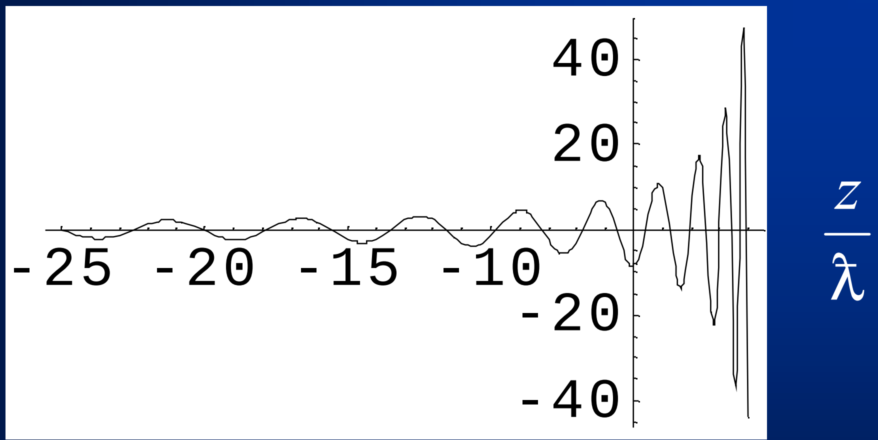
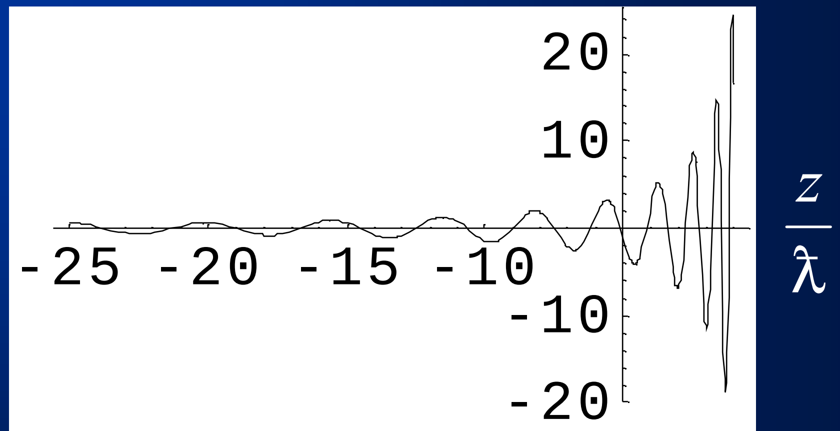
Local Electric Fields at Surface of Plasmonic Tapered Waveguide

Transverse field



Longitudinal field



$\lambda \approx 100$ nm

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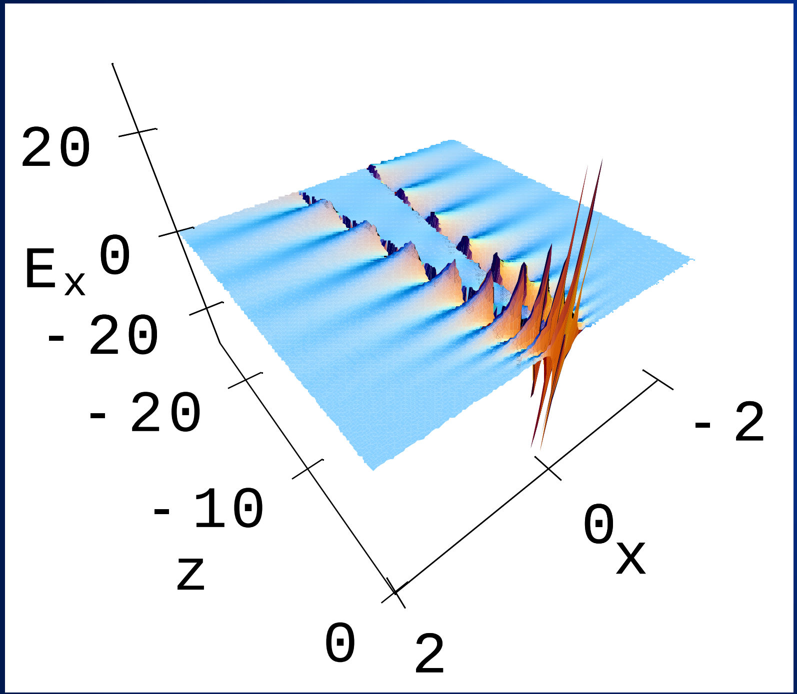
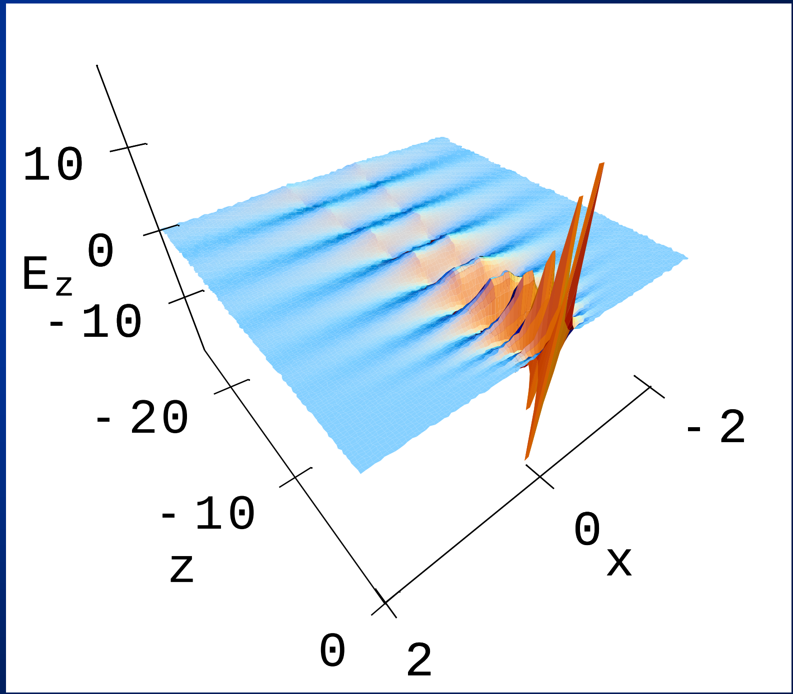
Local Electric Fields in Cross Section of System

Transverse electric field



Longitudinal electric field



Coordinates are in the units of $\lambda \approx 100$ nm

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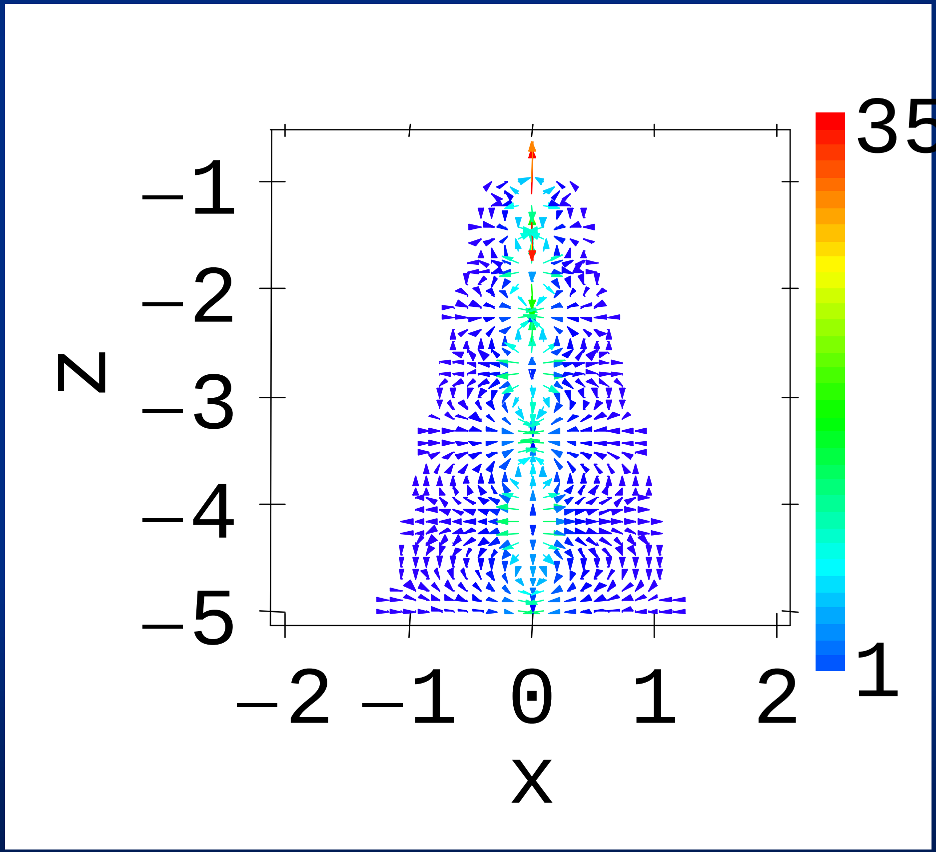
Vector of optical electric field for TM00 plasmonic mode of conic waveguide made of silver

Spatial scales are in units of 100 nm



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Adiabatic Nanofocusing Conclusions

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

The high wave vector of the TM00 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 ~50% of the initial energy flux, that along with adiabatic stopping leads to the local field-intensity enhancement by three orders of magnitude