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## 蔡定平 台灣大學物理系 中央研究院應用科學中心 國家實驗研究院儀器科技研究中心







## **A Short Story on Christmas Eve**

Richard P. Van Duyne Northwestern University, vanduyne@chem.northwestern.edu Martin Moskovits California Nanosystems Institute, University of California, Santa Barbara mmoskovits@ltsc.ucsb.edu

## **Contents**

What is "Plasmonic Nanophotonics"?

Why "Plasmonic Nanophotonics" is important?

What is current status and progress of "Plasmonic Nanophotonics"?

What is the future prospect of "Plasmonic Nanophotonics"?



## **Photonics**

- Physics and technology of generating and harnessing light and other forms of radiant energy whose quantum unit is the photon.
- The physics includes light emission, transmission, deflection, amplification and detection by optical components and instruments, lasers and other light sources, fiber optics, electro-optical instrumentation, related hardware and electronics, and sophisticated systems.
- The range of applications of photonics extends from energy generation to detection, communications and information processing.

from: www.photonics.com/dictionary/



# **Nanophotonics**



# **Plasmonic Nanophotonics**

### Photonics

- Photonics is the physics and technology of generating, manipulating, detecting, transferring and storing information using photons (light), which are particularly in the visible and near infra-red light spectrum.
- It is the study or application of electromagnetic energy incorporating optics, laser technology, electrical engineering, materials science, and information storage and processing.
- Photonics is also known as optoelectronics.

### Nanophotonics

The generation, manipulation, detection, transmission and storage of light (both far-field and near-field) using nano-scale materials.

### Plasmonic Nanophotonics

Using plasmonic nanostructures to generate, manipulate, detect, transfer and store light information in nanometer-scale region via surface plasmons.



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# History of plasmon

- Evidence of SPP's first observed (unknowingly) more than 100 years ago in experiments with metallic diffraction gratings. [Wood, Phil. Mag. <u>4</u>, 396 (1902)]
- Not understood until the pioneering work of Ritchie [Phys. Rev. <u>106</u>, 874 (1957)] and Powell & Swan [Phys. Rev. <u>118</u>, 640 (1960)] on electron energy loss spectra in metals.
- Plasmonic field enhancement effects began to attract attention in the mid-1970's: Surface-Enhanced Raman Spectroscopy [Fleischmann, Hendra & McQuillan, Dept. of Chemistry, Southampton, Chem. Phys. Lett. 26, 163, (1974)].
- □ Resurgence of interest in the last few years due to:
  - Advances in fabrication technologies (e-beam lithography, focused ionbeam milling, self-assembly), and their availability.
  - Advances in characterisation and analysis tools (near-field microscopy, quantitative EM simulation), and their availability.
  - Potential applications in fields from data transport and processing to solar cells, high-res. microscopy, biosensing, metamaterials,.....

# Plasmon

Plasmon = quantum of the collective excitation of free electrons in solids. A collective oscillation of a metal's conduction electrons.

- Surface plasmon = electron plasma oscillations near a metal surface.
- Polariton = 'quasiparticle' resulting from the coupling of EM waves with an electric or magnetic dipole-carrying excitation.
- Surface plasmon polariton = A combined excitation consisting of a surface plasmon and a photon – essentially, light waves trapped on the surface of a conductor.



## **Electron collective motion in metal spheres**

### Polarization of metal sphere under external electric field:

$$\alpha = 4\pi\varepsilon_0 R^3 \frac{\varepsilon_d - \varepsilon_m}{\varepsilon_d + 2\varepsilon_m}$$

( Classical electrodynamic calculation, Mie theory )



**Coherent oscillatory motion** 

### **Resonant excitation**



# From SPs to localized SPs

- In addition to surface plasmons on a plane surface, in other geometries, such as metallic particles or voids of different topologies, localized surface plasma excitations can be considered.
  - -- Such surface plasma excitations in bounded geometries are called localized surface plasmons (LSPs).

### Infinitely large metal

-- fields of surface plasmon resonances can propagate along the metal surfaces.

### Metal particles with nanometer size

-- fields of surface plasmon resonances are confined at the surface of the plasmonic nanostructures.



# The field of plasmon

## Rapid growth in research activity in recent years:



# New Springer Journal of "Plasmonics"



*Plasmonics* is an international forum for the publication of peerreviewed leading-edge original articles that both advance and report our knowledge base and practice of the interactions of freemetal electrons, *Plasmons*.

Topics covered include notable advances in the theory, Physics, and applications of surface plasmons in metals, to the rapidly emerging areas of nanotechnology, bio-photonics, sensing, biochemistry and medicine. Topics, including the theory, synthesis and optical properties of noble metal nanostructures, patterned surfaces or materials, continuous or grated surfaces, devices, or wires for their multifarious applications are particularly welcome. Typical applications might include but are not limited to, surface enhanced spectroscopic properties, such as Raman scattering or fluorescence, as well developments in techniques such as surface plasmon resonance and near-field scanning optical microscopy.

### Editor-in-chiefs: Chris D. Geddes, Joseph R. Lakowicz Regional Editor of Asia: Motoichi Ohtsu

Plasmonics publishes papers that describe new plasmonic based devices, new synthetic procedures for the preparation of nanostructures and there optical properties, as well their applications in analytical sensing. Papers describing new synthetic preparations and theory are particularly welcome.

## Why "Plasmonic Nanophotonics" is important?

- Conductive nanostructured materials open new avenues for manipulating light on the nanometer-scale.
- Specially designed plasmonic structures that act as "smart" optical nano-antennas for focusing light on nanometer scale with high spatial and spectral control of the energy concentration.
- Nano-antennas for strong enhancing a number of optical phenomena, such as the extraordinary optical transmittance, Raman scattering, nonlinear photoluminescence, Kerr optical nonlinearity, and many other important optical effects.
- Plasmonic nanoantennas open up the feasibility to detect molecules with unsurpassed sensitivity and perform lithography with nanometer spatial resolution.
- Plasmonic nanostructures can be employed for developing photonic nano-circuits where photons are controlled in a similar manner as electrons in conventional electronic circuits.
- Can be applied for developing novel left-handed materials with negative refraction index in the optical spectral range that can revolutionize the current optics.

## **Properties of Plasmonic structures**

## Properties of electromagnetic interactions

Short wavelengths



Wavelength of SP is less than the incident light => high spatial resolution => better control of energy flow in nano-scale Plasmonic resonances of fields are localized at the interfaces and hot spots of plasmonic nanostructures

J. P. Kottmann et. al., *Phys. Rev. B* **64**, 235402 (2001). K. L. Kelly, et. al., *J. Phys. Chem. B* **107** 668 (2003).

Localization of field





Field and intensity are enhanced at hot spots, and accumulation of enhance electromagnetic energy happened at certain spots of plasmonic nanostructures



## Why nanoscale photonics need plasmonics ?

- Fundamental blocks to process light in nanoscale devices with subwavelength dimensions:
  - Efficiency of light-matter interactions of wave-optics decreased for the structure size down to nanometer scale.
  - The diffraction limit resulted from wave behavior of light allows little possibilities to control and manipulate light in nanometer structures.

### □ Role of SPs resonances in nanophotonics:

- Metallic nanostructures which support surface plasmon modes => localize light fields to a sub-wavelength volume while enhancing local field strength by several orders of magnitude.
- The field-localization and short-wavelength properties of SPs waves => accumulation of local EM energy at hot spots with very small volume.
  - The optical field of plasmonic resonance are localized and enhanced, and the efficiencies of optical interactions are increased.
    The plasmonic nanostructures can be used as optical nano-antennas to manipulate light in nanometer scale.



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## **Current progress of plasmonic nanophotonics**

- □ A. Sensing, detection, imaging and recording
  - A-1. Surface plasmon resonance (SPR) sensing
  - A-2. Surface enhanced Raman scattering (SERS)
  - □ A-3. Single molecule detection
  - A-4. Surface plasmon cover glass slip
  - A-5. Plasmonic nano optical recording
- B. Nano-photonics integration
  - □ B-1. Plasmonic waveguide
  - B-2. Nano focusing
  - B-3. Metamaterial and nano fabrication
- C. Photo-chemistry process
  - C-1. Solar cell
  - □ C-2. Photocatalytic Micro-reactor
- D. Photo-physics interaction
  - □ D-1. Energy conversion efficiency of luminescence and fluorescence

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D-2. OLED efficiency

## A-1a. Surface plasmon resonance sensing

### □ Schematic diagram of SPR sensor



$$k_{SP} = \frac{\omega}{c} \left(\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d}\right)^{\frac{1}{2}}$$

 $\epsilon_m$ : dielectric constant of the metal (Au) film  $\epsilon_d$ : dielectric constant of the material in the flow channel

### □ Advantages:

- Evanescent field interacts with adsorbed molecules only
- Coupling angle strongly depends on  $\epsilon_{\rm d}$
- Use of well-established surface chemistry for Au (thiol chemistry)



## A-1b. Surface plasmon resonance sensing

### Plasmonic sensing of biological analytes through nanoholes



• Schematic of the 2-D nanohole-array SPP transmission setup. (Inset: SEM image of a section of the gold nanohole array.)







- Transmission spectra for the ethylene glycol solution series (%): 0, 1.96, 3.85, 5.60, 7.40, 9.10.
- Monitoring the resonant wavelength of a SPP mode at the fluid-metal overlayer in a transmission setup.
- The system presented here has the potential to detect Con A down to 1.43 g/ml (or 53 nM) and to detect anti-BSA down to 38 ng/ml (or 271 pM).

## A-2a. Surface enhanced Raman scattering



Advanced Fuel Research, Inc., East Hartford, CT Shuming Nie and Steven R. Emory, *Science* **275**, 1102 (1997).

R. M. Stockle, Y. D. Suh, V. Deckert, R. Zenobi, Chem. Phys. Lett. 318, 131 (2000).

# A-2b. Raman-enhancing substrates based on silver nanoparticle arrays



F. Hsu, J. K. Wang, and Y. L. Wang, Adv. Mater. 18, 491 (2006).

# A-2c. Polarization dependence of SERS in gold nanoparticle -nanowire systems



For the separation between particle and wire of 5 nm, the averaged SERS enhancement is in the order of 10<sup>6</sup> for the different shapes of the particles, and the maximum local SERS enhancement for perpendicular polarization is of the order of 10<sup>10</sup>.

Hong Wei, Feng Hao, Yingzhou Huang, Wenzhong Wang, Peter Nordlander, and Hongxing Xu, *Nano Lett.* **8**, 2497 (2008).

## A-3a. SP enhanced single molecule detection

### Single molecule imaging -- using fluorescence

K: kinesin 為一種微管附屬性蛋白質 (趨向正端) M: microtubule



囊泡就是由dynein 與kinesin 沿著微管作運送(耗ATP); kinesin 可 與β-tubulin結合, kinesin 與ATP 結合水解後可與β-tubulin 分開

H. Yokota, Ki. Saito, and T. Yanagida, *Phys. Rev. Lett.* **80**, 4606 (1998).









SP enhanced fluorescence images of a single kinesin molecule. P/NSTL@NTU

## A-3b. SP enhanced single molecule detection

### A molecular ruler based on plasmon coupling of metallic nanoparticles

 Color effect on directed assembly of DNA-functionalized Au and Ag nanoparticles.



> Single Ag particles → blue color.
Ag particle-pairs → blue-green color.
Single Au particles → green color.
Au particle-pairs → orange color.
(particle sizes ~ 40 nm)

• Spectra shift upon DNA hybridization



 Spectra peak position as function of time after addition of complementary DNA



Discrete states are observed in the time dependent spectra peak position (indicated by horizontal dashed lines).

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C. Sonnichsen, B. M. Reinhard, J. Liphardt, and A. P. Alivisatos, *Nature Biotechnology.* **23**, 741 (2005).

# **B-1. Guiding of electromagnetic energy**

SP propagation along the 18.6 μm
SP propagation along 4.7 μm long
Au and Ag nanorod



incident wavelength: 785 nm excited SP wavelength: 414 nm

H. Ditlbacher, A. Hohenau, D. Wagner, U. Kreibig, M. Rogers, F. Hofer, F. R. Aussenegg, and J. R. Krenn, *Phys. Rev. Lett.* **95**, 257403 (2005). R. M. Dickson and L. A. Lyon, *J. Phys. Chem. B* **104**, 6095 (2000).

Au 532 nm 820 nm Ag 532 nm 820 nm **Excitation** Nanowire Glass - Immersion oil Objective lens P/NSTL@NTU Collection

# **B-2. Manipulation of electromagnetic wave**

### Subwavelength focusing



50-nm Ag film containing 19 200-nm holes ranged on a quarter circle with a 5- $\mu$ m radius.

### □SPP subwavelength focusing and guiding





L. Yin, V. K. Vlasko-Vlasov, J. Pearson, J. M. Hiller, J. Hua, U. Welp, D. E. Brown, and C. W. Kimball, *Nano Lett.* **5**, 1399

### Plasmonic visible nanosource



#### SEM image of the nanosource structure.

The dimensions of the Bragg gratings are *a*=165 nm, *h*=80 nm, and *D*=295 nm.





Field amplitude taken at 100 nm from the hole exit as a function of the incident wavelength.

Marianne Consonni, Jérôme Hazart, and Gilles Lérondel, *Appl. Phys. Lett.* **94**, 051105 (2009).

# **B-3a. Developments of metamaterials**

### □ What is metamaterial (異向性材料?/異向性物質?/超穎材料?/超穎物質?)?

- Metamaterials are artificial nanostructures made of metals or composites that exhibit behavior which (i) either their component materials do not exhibit (ii) or is enhanced relative to exhibition in the component materials.
- The permittivity ( $\epsilon$ ) and permeability ( $\mu$ ) of a metamaterial are decided primarily by the constitutive nano structure of the component materials rather than what it is made of.

### Some examples of metamaterials

 metallic split-ringresonator (SRR) and wire arrays





Cu SRR structure T. J. Yen, et al., Science 308, 1494 (2004).

 Array of metallic nanowire or nano pillar pairs



#### Au nanowire pairs

V. M. Shalaev, et al., physics/0504091 (2005). A. N. Grigorenko, et al., Nature 438, 335 (2005).

### Array of chiral structures



#### Resin with chiral structure

Jun-ichi Kato, et al., Appl. Phys. Lett. 86, 044102 (2005).

# **B-3b. Nano assembly and patterning**



Optical spectra



TEM image of



Theoretical modelling of the optical spectra of two different-sized nanoprisms.

-- The calculated plasmon bands reproduce the experimentally observed spectrum.







Spectrum measurement of far-field transmission of hole arrays with different periods.



AFM image of a pattern obtained by SP lithography. (using  $\lambda$  = 365 nm and a mask of hole array with period of 170 nm)

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R. Jin, Y. C. Cao, E. Hao, G. S. Métraux, G. C. Schatz, and C. A. Mirkinthe, *Nature* **425**, 487 (2003). W. Srituravanich, N. Fang, C. Sun, Q. Luo, and X. Zhang, *Nano Lett.* **4**, 1085 (2004).

# **B-3c. Plasmonic Nanolithography**

# Schematic cross-section of a plasmon interference printing system



- The illumination generates counter-propagating surface plasmons.
- The SPs interfere on the central metallic area.
- Then, the corresponding field is printed in the DR1MA/MMA layer.



M. Derouard, and J. Hazart et al., Opt. Express 15, 4238 (2007).

Flying plasmonic lens for high-speed nanolithography





 AFM image of arbitrary writing pattern of 'SINAM' with 145 nm linewidth on the TeO<sub>x</sub>-based thermal photoresist.

W. Srituravanich, L. Pan, Y. Wang, C. Sun, D. B. Bogy, and X. Zhang., *Nat. Nanotechol.* **3**, 733 (2008).



## C-1a. SP enhanced photovoltaic conversion efficiency of a solar cell



O. Stenzel, A. Stendal, K. Voigtsberger, C. von Borczyskowski, *Sol. Energy Mater. Sol. Cells* **37**, 337 (1995).

# C-1b. Plasmonic enhanced efficiency of photovoltaic organic solar cell

### □ Plasmonic nanocavity arrays for enhanced efficiency in OPV cell

Device structure: Glass substrate/patterned Ag (30nm) /copper phthalocyanine (CuPc) (20nm)/C<sub>60</sub> (40nm)/bathocuproine (BCP) 10nm/Al (50nm)

Simulated field intensity map ( $\lambda$ =838 nm)



SEM image of the patterned Ag anode

Ratio of patterned to unpatterned device external quantum efficiency



> The nanocavity array is formed between a patterned Ag anode and an unpatterned AI cathode.

> The power conversion efficiency ( $\eta_p$ ) can be increased by a factor of 3.2 relative to an unpatterned device.

### Plasmon enhanced performance of organic solar cell



The incident photon to charge carrier efficiency (ICPE) is increased ~20% compared to the reference cell.

N. C. Lindquist, W. A. Luhman, S. H. Oh, and R. J. Holmes, *Appl. Phys. Lett.* **93**, 123308 (2008). S. S. Kim, S. I. Na, J. Jo, D. Y. Kim, and Y. C. Nah, *Appl. Phys. Lett.* **93**, 073307 (2008).

## **C-2.** Photocatalytic Micro-reactor

### Optical fiber photo-chemical reactor



## D-1a. SP enhanced energy conversion efficiency of fluorescence and luminescence

L intensity (a.u.)

### □ SP enhanced fluorescence



Fluorescence spectra of Rose Bengal with and without 50nm Ag nanoparticles.



T. Nakamura and S. Hayashi, Jap. J. Appl. Phys. 44, 6833 (2005).


# D-1b. SP enhanced energy conversion efficiency of fluorescence and luminescence

### Nanoaperture-enhanced fluorescence

 Schematics of the experimental setup. (Inset: Micrograph of a single nanoaperture milled in Au film)



 Fluorescence enhancement factor as a function of the aperture diameter for Alexa-Fluor 647 dyes at excitation wavelength λ<sub>e</sub>= 633 nm.



> Enhancement factor  $\eta_{max}$  ~12 for Au and  $\eta_{max}$ ~7.7 for Al.

### PL enhancement by SPs of a biharmonic metallic grating substrate



- (Left) AFM image of biharmonic metallic surface includes  $\Lambda_1$ =500 nm and  $\Lambda_2$ =250 nm gratings.
- (Right) PL spectrum of biharmonic metallic grating coated with silicon rich silicon nitride.
- There is 30 times enhancement in PL signal at wavelengths coinciding with the plasmonic resonance wavelengths.

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D. Gérard, et al., Phys. Rev. B 77, 045413 (2008). A. Kocabas et al., Opt. Express 16, 12469 (2008).

# **D-2a. SP enhanced OLED and LED efficiency**





Configuration of the corrugated and flat OLEDs.



AFM image of the top metallic cathode surface of an OLED fabricated with 2-D corrugation.



Electroluminesence (EL) spectra observed at normal directions of 1-D and 2-D corrugated and flat devices.

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J. Feng, T. Okamoto, S. Kawata, Opt. Lett. 30, 2302 (2005).

# **D-2b. Plasmonic enhanced LED efficiency**

- Localized surface plasmon-induced LED emission enhancement
  - SEM images of as-deposited and thermally annealed nanostructure Ag thin film of 12 nm in thickness.



- EL spectra of a InGaN/GaN QW LED sample
  - A: LED sample with out Ag
  - B: LED sample with 12 nm flat Ag film
  - C: LED sample with 12 nm thermally annealed Ag film



- In sample C, the LSP coupling leads to 150% enhancement in peak intensity and 120% enhancement in integrated intensity at the injection current of 20 mA.
- The output EL peak intensities of the three LED samples as functions of injection current show that with LSP coupling, sample C can achieve 90-220% output intensity enhancement over the conventional LED (A).

### Metal grating induced LED emission enhancement



- EL spectra of a InGaN/GaN QW LED sample
  - A: LED sample with out Ag
  - B: LED sample with flat Ag film
  - C: LED sample with 1D grating Ag film (period and groove depth are 500 and 30 nm)

The total output intensity of an LED of SP-QW coupling can be enhanced by ~200%

D. M. Yeh, et al., *Nanotechnology* **19**, 345201 (2008). K. C. Shen, et al., *Appl. Phys. Lett.* **93**, 231111 (2008).



# **Achievement of current progress**

### **Sensitivity**

- Resolution of dielectric difference to a refractive index unit of 5×10<sup>-7</sup> to 5×10<sup>-8</sup> (corresponding to 1-pg/mm<sup>2</sup> to 100-fg/mm<sup>2</sup>)
- □ Single molecular (diameter of 10 to 20 nm) fluorescence detection

### **Enhancement and Efficiency**

- □ Near-field enhancement corresponds to SERS locally in excess of 10<sup>15</sup>
- Plasmonic nanostructures can be used to achieve 30 times enhancement in PL signal or 10 times enhancement in molecule fluorescence.
- Surface enhancement Raman and combination with scanning probe microscope to detect DNA, single nanowire
- □ The light emission efficiency of an LED can be enhanced to 4 to 15 times by using metal particles or metal grating
- □ Spontaneous emission rate of semiconductor QW (QD) can be 55~100 times faster than normal one

### □ High spatial resolution for imaging, storage and nanolithography

- **The nano recording mark smaller then 100 nm can be readout**
- □ A pattern with 90 nm features on a 170 nm period has been obtained by surface plasmon lithography using an exposure radiation of 365 nm wavelength

### Nano-photonics integrated circuit

- SPs propagate along metal guide with diameter smaller than 50 nm
- SPs photonic IC, such as emitter, collector, amplifier, transistor, switch and gate P/NSTL@NTU

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## Nanophotonics from plasmonic nano structures



# **Developments of plasmonic metamaterials**

Basic scheme of optical setup. MLA: microlens array, L1: lens, DM: dichroic mirror, OL: objective lens. A is the plane where multiple focuses are generated



*Appl. Phys. A* **80**, 683 (2005).



The over view and it optical microscopic image



The SEM image of photopolymetized resin voxels generated on a glass substrate





Jun-ichi Kato, N. Takeyasu, Y. Adachi, H. B. Sun, and S. Kawata, *Appl. Phys. Lett.* **86**, 044102 (2005).

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# **Metamaterials**

- Metamaterials are macroscopic composites having a manmade, periodic cellular architecture designed to produce an optimized combination, not available in nature, of two or more responses to specific excitation.
- A metamaterial is a material which gains its properties from its structure rather than directly from its composition. To distinguish metamaterials from other composite materials, the metamaterial label is usually used for a material which has unusual properties.
- The permittivity (ε) and permeability (μ) of a metamaterial are decided primarily by the constitutive structure of the component materials rather than what it is made of.

R.M. Walser, in: W.S. Weiglhofer and A. Lakhtakia (Eds.), *"Introduction to Complex Mediums for Electromagnetics and Optics"*, SPIE Press, Bellingham, WA, USA, 2003. http://en.wikipedia.org/wiki/Metamaterials



# εand μ diagram



# From electronics to plasmonic nanophotonics

### □ Nanophotonics with Plasmonic/metal optics: A logical next step



The ever-increasing need for faster information processing and transport is undeniable.

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Electronic components are running out of steam due to issues with RC-delay times.

# **Integrated Plasmonic nanophotonic device**



# Niche of plasmonic nanophotonics

- Well developed semiconductor industry
  - Excellent nano fabrication skill
  - Interests on new developments
  - Good support from industry and academy
- Blossom and flourish of photonic industry
  - Two digits growth booming
  - Strong industrial interests and supports
  - Demand of upgrade and advancement
- Abundance of human resource
  - Well educated people from many high quality universities
  - Experienced and well trained personnel from industry

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Rich support from society

Many links to different industrial applications

- One for all possibilities and great future
- Little investment for tremendous return

# Thanks for your attention....

# **Plasmonic Nanophotonics**

Din Ping Tsai Department of Physics, National Taiwan University Research Center for Applied Sciences, Academia Sinica Instrumentation Technology Research Center, National Applied Research Laboratories

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