Trends in Nanoplasmonics: Citius, Minimius, Fortius!

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• Spaser as an Ultrafast Quantum Generator and Nanoamplifier
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• Bonus: Ultrafast Nanoscale Coherent Control
• Bonus: Attosecond Plasmonic Field Nanoscope
Nanoplasmonics in a nano-nutshell

Concentration of optical energy on the nanoscale

Photon

Surface Plasmon

Skin depth \(\sim 25 \text{ nm}\)

Size \(\sim 10 \text{ nm}\)

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Nanoplasmonic colors are very bright. Scattering and absorption of light by them are very strong. This is due to the fact that all of the millions of electrons move in unison in plasmonic oscillations.

Nanoplasmonic colors are also eternal: metal nanoparticles are stable in glass: they do not bleach and do not blink. Gold is stable under biological conditions and is not toxic in vivo.

Colors of Silver Nanocrystals and Gold Nanoshapes

Scanning electron microscopy

Dark field optical microscopy


**SEM-ESB: Energy Selective Backscattered**

D. A. Pawlak et al., Institute of Electronic Materials Technology, Warsaw, Poland

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The magnificent nanoplasmonic colors: The windows of La Sainte-Chapelle, Paris

Applications of Nanoplasmonics:


2. Near-filed scanning microscopy (or, nanoscopy): NSOM (SNOM)


4. Photo- and chemically stable labels and probes for biomedical research and medicine

5. Nanoplasmonic-based immunoassays and tests. Home pregnancy test (dominating the market), PSA test (clinic), troponin heart-attack test, and HIV tests (in trials)

6. Near perspective: Generation of EUV and XUV pulses

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Plasmonic Hot Spots 15th Anniversary

- M. Hentschel et al., Nano Lett. 10, 2721 (2010)

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Enhancement factors for small nanoparticles (size $R < l_s \sim 25$ nm)

Plasmonic quality factor: $Q = \frac{-\text{Re } \varepsilon_m}{\text{Im } \varepsilon_m} \sim 10 - 100$

Radiative rate enhancement for dipole mode frequency: $\sim Q^2$

Excitation rate enhancement: $\sim Q^2$

SERS enhancement: $\sim Q^4$

The above-listed enhancement factors do not depend on size $R$

Emission rate of SPs into a mode: $\propto \frac{Q}{R^3}$

This with respect to free photons: $\sim \frac{\lambda^3 Q}{R^3}$ (Purcell factor)

This enhancement factor is *inversely* proportional to $R^3$

This is of fundamental importance for spasers (plasmonic nanolasers)
Nanoplasmonics is intrinsically ultrafast:

- Surface plasmon relaxation times are in ~10-100 fs range.
- Spectrally, surface plasmon resonances in complex systems occupy a very wide frequency band; for gold and silver:
  \[ \Delta \omega \approx \frac{\omega_p}{\sqrt{2}} \approx 4 \text{ eV} \]
- Including aluminum with plasmon responses in the ultraviolet, this spectral width increases to ~10 eV.
- Best area for plasmonics

Corresponding rise time of plasmonic responses ~ 100 as
Localized SP hot spots and SPPs coexist in space and time on nanostructured surfaces


**PEEM Image as a Function of Delay (250 as per frame)**

30 femtoseconds from life of a nanoplasmonic system

Localized SP hot spots are deeply subwavelength as seen in PEEM (photoemission electron microscope)

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Constructive control by the second pulse
Destructive control by the second pulse
No control pulse (free induction decay)

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Adiabatic Compression

Field enhancement:

\[ \sim \frac{L_s}{R} \quad \text{(for 2d compression), } L_s \approx 25 \text{ nm} \]

\[ \sim \left( \frac{L_s}{R} \right)^{3/2} \quad \text{(for 3d compression)} \]

Nanowire Plasmon Excitation by Adiabatic Mode Transformation

Ewold Verhagen,* Marko Spasenović, Albert Polman, and L. (Kobus) Kuipers

FIG. 4 (color). (a) Secondary electron micrograph of a 2 μm long nanowire connected by tapered waveguide sections for input and output coupling. (b) Near-field amplitude of forward-propagating waves in the structure at \( \lambda = 1550 \) nm. The intensity transmission of the complete structure is 20 ± 6%.

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Near-Field Localization in Plasmonic Superfocusing: A Nanoemitter on a Tip

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FIGURE 1. Grating coupling of surface plasmons on a tip. Overlay of SEM and optical far-field image of a Au tip with grating written by FIB for surface plasmon coupling of incident near-IR light from a Ti:Sapphire laser (spectrum shown). The grating with period $a_0 \sim 770$ nm is illuminated with polarization parallel with respect to the tip axis and an incident focus size of $\sim 8 \mu m$. The nonradiative SPP propagation leads to energy transfer and focusing and finally reemission near the tip apex with radius $\leq 15$ nm.

FIGURE 2. Determination of tip emitter size. (a) Schematic of scanning the nanofocusing tip across a silicon step edge with radius $3 \pm 1$ nm. (b) Top view SEM image of step edge. The wall and lower terrace are on the right-hand side. The edge serves as a local scatterer of the optical near-field of the apex. (c) The optical signal of a lateral scan across the step edge provides a measure of the spatial field confinement and thus the emitter size at the apex. Solid black line: AFM topography of the step. Red circles: plasmonic edge-scattered light intensity of the apex. The optical intensity peaks at the step edge and displays a width of $22 \pm 5$ nm, demonstrating the near-field localization at the apex. Solid red: Signal obtained under direct illumination of the apex under otherwise identical conditions.
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Heat-assisted magnetic recording by a near-field transducer with efficient optical energy transfer

Here decay of SPP is a useful effect!
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Gold Nanolenses Generated by Laser Ablation-Efficient Enhancing Structure for Surface Enhanced Raman Scattering Analytics and Sensing

Janina Kneipp,*†,†† Xiangting Li,§ Margaret Sherwood,† Ulrich Panne,† Harald Kneipp,† Mark I. Stockman,§ and Katrin Kneipp††

Different types of aggregates of gold nanospheres

Scale bar: 100 nm

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Surface Plasmon Amplification by Stimulated Emission of Radiation (SPASER)


Amplification and Stimulated Emission in Plasmonic systems:

Spaser is the ultimately smallest quantum nano-generator and nano-amplifier.

For small nanoparticles, radiative loss is negligible.

Spaser is quasistatic and fully scalable.

\[ |d_{10}|^2 \frac{N_{QD} \Omega}{\hbar R^3 \Gamma} \geq 1 \]

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Stationary (CW) spaser regime

This quasilinear dependence $N_n(g)$ is a result of the very strong feedback in spaser due to the small modal volume

arXiv:0908.3559
Bandwidth $\sim 10$-100 THz

Very high resistance to ionizing radiation

**Amplification in Spaser with a Saturable Absorber (1/3 of the gain chromophores)**

Stationary pumping

This very high speed of the spaser is due to the small modal volume

Pulse pumping

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Demonstration of a spaser-based nanolaser

M. A. Noginov¹, G. Zhu¹, A. M. Belgrave¹, R. Bakker², V. M. Shalaev², E. E. Narimanov², S. Stout¹,³, E. Herz³, T. Suteewong³ & U. Wiesner³

Figure 1 | Spaser design. a, Diagram of the hybrid nanoparticle architecture (not to scale), indicating dye molecules throughout the silica shell. b, Transmission electron microscope image of Au core. c, Scanning electron microscope image of Au/silica/dye core–shell nanoparticles. d, Spaser mode (in false colour), with \( \lambda \) = circles represent the 14-nm strength colour scheme is
Figure 2 | Spectroscopic results. Normalized extinction (1), excitation (2), spontaneous emission (3), and stimulated emission (4) spectra of Au/silica/dye nanoparticles. The peak extinction cross-section of the nanoparticles is $1.1 \times 10^{-12}$ cm$^2$. The emission and excitation spectra were measured in a spectrofluorometer at low fluence.

Figure 4 | Stimulated emission. a, Main panel, stimulated emission spectra of the nanoparticle sample pumped with 22.5 mJ (1), 9 mJ (2), 4.5 mJ (3), 2 mJ (4) and 1.25 mJ (5) 5-ns optical parametric oscillator pulses at $\lambda = 488$ nm. b, Main panel, corresponding input–output curve (lower axis, total launched pumping energy; upper axis, absorbed pumping energy per nanoparticle); for most experimental points, ~5% error bars (determined by the noise of the photodetector and the instability of the pumping laser) do not exceed the size of the symbol. Inset of a, stimulated emission spectrum at more than 100-fold dilution of the sample. Inset of b, the ratio of the stimulated emission intensity (integrated between 526 nm and 537 nm) to the spontaneous emission background (integrated at <526 nm and >537 nm).

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Plasmon lasers at deep subwavelength scale

Rupert F. Oulton, Volker J. Sorger, Thomas Zentgraf, Ren-Min Ma, Christopher Gladden, Lun Dai, Guy Bartal & Xiang Zhang

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Room-temperature sub-diffraction-limited plasmon laser by total internal reflection

Ren-Min Ma\textsuperscript{1}\textsuperscript{†}, Rupert E. Oulton\textsuperscript{1}\textsuperscript{†}, Volker J. Sorger\textsuperscript{1}, Guy Bartal\textsuperscript{1} and Xian Zhang\textsuperscript{1,2}\textsuperscript{*}

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• Bonus: Attosecond Plasmonic Field Nanoscope
BRIEF CONCLUSIONS

1. Nanoplasmonics is based on nanolocalization of optical fields due to SPs
2. Enhancement in nanoplasmonics is due to quality factor of SP modes and geometric concentration
3. Plasmonic hot spots is universal phenomena due to the scale-invariance of the nanoplasmonic phenomena
4. Adiabatic concentration is a non-resonant, wide-band, and non-radiative root to nanofocusing with extremely high throughput. There are demonstrated applications to nanoscopy and chemical nano-imaging.
5. Nanolenses are highly efficient enhancers of local field and SERS
6. SPASER is an efficient nanoscale generator and ultrafast quantum amplifier with a switch time \(~100\) fs for silver and \(~10\) fs for gold. It has the same size as MOSFET and can perform the same functions but is \(~1000\) times faster.
7. SPASERs have been observed in a number of experiments
8. The most promising applications of the SPASER are an ultrafast nanoamplifier, local optical energy source, active nano-label, and an element of metamaterials with compensated loss.
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Different spectral components of the excitation pulse excite resonant surface plasmon modes. These excitations dynamically interfere creating time-dependent hot spots of local fields during their coherence time.

This interference can be directed by choosing phases and amplitudes of the different frequency components of the excitation pulse (pulse shaping).

Schematic of Coherent Control by Phase Modulation


Adaptive subwavelength control of nano-optical fields

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Trends in Nanoplasmonics: Faster, Smaller, Stronger!
Nanoplasmonic Energy Localization, Time Reversal, and Coherent Control


Idea of time reversal for subwavelength EM-wave localization:


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Resonant Metalenses for Breaking the Diffraction Barrier

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We introduce the resonant metalens, a cluster of coupled subwavelength resonators. Dispersion allows the conversion of subwavelength wave fields into temporal signatures while the Purcell effect permits an efficient radiation of this information in the far field. The study of an array of resonant wires using microwaves provides a physical understanding of the underlying mechanism. We experimentally demonstrate imaging and focusing from the far field with resolutions far below the diffraction limit. This concept is realizable at any frequency where subwavelength resonators can be designed.

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PACS numbers: 41.20.—q, 78.67.Pt, 81.05.Xj

amplitude of $E_x$ TEM Bloch modes (1,1), (2,3), (5,6), and (19,19).

(d) Focal spot obtained after far field time reversal

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Controlling the Optical Near Field of Nanoantennas with Spatial Phase-Shaped Beams

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Figure 4. Experimental TPL maps recorded for (a) a Gaussian beam and (b, c) a HG_{10} beam whose phase shift (located by the vertical dashed line) coincides with (b) the right gap and (c) the left gap.

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Attosecond nanoplasmonic-field microscope

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Schematic of Attosecond Nanoplasmonic Field Microscope

XUV photoelectrons accelerated by enhanced IR plasmonic local fields

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Excitation field

Local optical electric field at the “hottest spot”

Attosecond pulse applied within a period of IR oscillations

$t_x = 14$ fs
Energy shift (eV) of electrons emitted by a 95 eV XUV attosecond pulse as a function of the as pulse excitation instant with respect to the infrared excitation field (frames are in 200 as) as observed in Photoemission Electron Microscope (PEEM).

Experiment directly measures instantaneous electric potential of nanoplasmic oscillations with nm spatial and ~200 as temporal resolution.

Nanosystem is 60x60 nm random silver film (50% filling factor)

Energy change (eV) of 90 eV XUV photoelectrons from silver nanosystem for 10 GW/cm² 800 nm IR power; × 10^{15} slowed down.