



2328-5

Preparatory School to the Winter College on Optics and the Winter College on Optics: Advances in Nano-Optics and Plasmonics

30 January - 17 February, 2012

Nanoplasmonics: Optical Properties of Plasmonic Nanosytems III

M.I. Stockmani Georgia State University Atlanta U.S.A.



Support:



US Israel Binational Science Foundation

Optical Properties of Plasmonic Nanosystems <u>Mark I. Stockman</u>

Nanoplasmonics:

Department of Physics and Astronomy, Georgia State University, Atlanta, GA 30303, USA

Lecture 3: Ultrafast, Nonlinear, and Quantum Nanoplasmonics

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LECTURE 3

Ultrafast, Nonlinear, and Quantum Nanoplasmonics

- 1. Introduction: Problem of nanoscale control of local optical fields
- 2. Coherent control using pulse shaping
- 3. Two-pulse (interferometric) coherent control and visualization
- 4. Time-reversal and determination of controlling pulses
- 5. Attosecond nanoplasmonics: attosecond plasmonic field microscope
- 6. Generation of high harmonics (EUV and XUV radiation)
- 7. Surface plasmon amplification by stimulated emission of radiation (SPASER) and nanolasers

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PROBLEMS IN NANOOPTICS



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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 $\Delta \omega \sim \omega_p \sim 10 \, \mathrm{eV}$ Corresponding shortest time of plasmonic responses ~ 100 as

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COHERENT CONTROL ON NANOSCALE

Problem of dynamic spatial control at the nanoscale: The wavelength of the excitation radiation is orders of magnitude too large to control spatial distribution of local fields on nanoscale by focusing

Thus, optical radiation does not have spatial degrees of freedom on the nanoscale

However, it does possess spectral (phase, or temporal) degrees of freedom and polarization. These can be used to *dynamically* control the optical energy localization at the nanoscale

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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)

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Principles of coherent (quantum) control

D. J. Tannor and S. A. Rice, *Control of Selectivity of Chemical Reaction Via Control of Wave Packet Evolution*, The Journal of Chemical Physics **83**, 5013-5018 (1985); P. Brumer and M. Shapiro, *Principles of the Quantum Control of Molecular Processes* (Wiley, New York, 2003); R. S. Judson and H. Rabitz, *Teaching Lasers to Control Molecules*, Phys. Rev. Lett. **68**, 1500 (1992); A. P. Heberle, J. J. Baumberg, and K. Kohler, *Ultrafast Coherent Control and Destruction of Excitons in Quantum-Wells*, Phys. Rev. Lett. **75**, 2598-2601 (1995).

REFERENCES ON COHERENT CONTROL OF OPTICAL ENERGY NANOLOCALIZATION

•M. I. Stockman, S. V. Faleev, and D. J. Bergman, *Coherent Control of Femtosecond Energy Localization in Nanosystems*, Phys. Rev. Lett. **88**, 67402-1-4 (2002).

•M. I. Stockman, D. J. Bergman, and T. Kobayashi, *Coherent Control of Nanoscale Localization of Ultrafast Optical Excitation in Nanosystems*, Phys. Rev. B **69**, 054202-10 (2004)

•A. Kubo, K. Onda, H. Petek, Z. Sun, Y. S. Jung, and H. K. Kim, *Femtosecond Imaging of Surface Plasmon Dynamics in a Nanostructured Silver Film*, Nano Lett. **5**, 1123-1127 (2005)

•M. I. Stockman and P. Hewageegana, *Nanolocalized Nonlinear Electron Photoemission under Coherent Control*, Nano Lett. **5**, 2325-2329 (2005)

•M. Aeschlimann, M. Bauer, D. Bayer, T. Brixner, F. J. G. d. Abajo, W. Pfeiffer, M. Rohmer, C. Spindler, and F. Steeb, *Adaptive Subwavelength Control of Nano-Optical Fields*, Nature **446**, 301-304 (2007).

•X. Li and M. I. Stockman, *Highly Efficient Spatiotemporal Coherent Control in Nanoplasmonics on a Nanometer-Femtosecond Scale by Time Reversal*, Phys. Rev. B **77**, 195109-1-10 (2008).

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•Femtosecond local fields on nanoscale:

$$\varphi(\mathbf{r},t) = \varphi_0(\mathbf{r},t) - \int \varphi_0(\mathbf{r}',t') \nabla_{\mathbf{r}'}^2 G(\mathbf{r},\mathbf{r}';t-t') d^3r' dt'$$

Example to be considered: The exciting pulses are *z*-polarized, have Gaussian envelopes, and carry linear chirp,

$$E_{z}^{(0)}(t) = \exp\left[i\omega_{0}\left(1 + \alpha \frac{t - T/2}{T}\right)\left(t - T/2\right) - \frac{3}{2}\left(\frac{t - T/2}{T}\right)^{2}\right] + \text{c.c.}$$

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Possibility Coherent Control on Nanoscale Demonstrated

The nanosystems studied are an "engineered" V-shape and a random planar composite (RPC), positioned in the plane. The material is silver; the spatial scale is 1-3 nm/grid unit.



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Spatial Distribution: Local Fields in V-shape, Negative Chirp



Conclusion: There is a strong localization of the excitation energy at the tip of the nanostructure during a time interval on order of the pulse length

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Georgia State University Atlanta, GA 30303-3083 Spatial Distribution: Local Fields in V-shape, Positive Chirp



Conclusion: Excitation energy is transferred between the tip and the opening of the nanostructure. No spatial concentration of energy takes place.

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Local Optical Fields in Random Planar Composite at the Instants of their Maxima



Conclusion: The phase is a controlling factor in random systems as well

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Femtosecond Imaging of Surface Plasmon Dynamics in a Nanostructured Silver Film

Atsushi Kubo,^{†,§} Ken Onda,^{†,§} Hrvoje Petek,^{*,†,§} Zhijun Sun,^{‡,§} Yun S. Jung,^{‡,§} and Hong Koo Kim^{‡,§}

Department of Physics and Astronomy, Department of Electrical Engineering, and Institute of NanoScience and Engineering, University of Pittsburgh, Pittsburgh, Pennsylvania 15260

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ABSTRACT

Light interacting with nanostructured metals excites the collective charge density fluctuations known as surface plasmons (SP). Through excitation of the localized SP eigenmodes incident light is trapped on the nanometer spatial and femtosecond temporal scales and its field is enhanced. Here we demonstrate the imaging and quantum control of SP dynamics in a nanostructured silver film. By inducing and imaging the nonlinear two-photon photoemission from the sample with a pair of identical 10-fs laser pulses while scanning the pulse delay, we record a movie of SP fields at a rate of 330-attoseconds/frame.

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Two-Photon Interferometric Coherent Control



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Two-Photon Interferometric Coherent Control (Movie as a function of the delay time between the pulses)



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A. Kubo, K. Onda, H. Petek, Z. Sun, Y. S. Jung, and H. K. Kim, *Femtosecond Imaging of Surface Plasmon Dynamics in a Nanostructured Silver Film*, Nano Lett. 5, 1123 (2005).
PEEM Image as a Function of Delay (250 as per frame)

200 nm

- 30 femtoseconds from life of a nanoplasmonic systems Localized SP hot spots are deeply subwavelength as seen in PEEM (photoemission electron
- microscope)



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Due to the low photoelectron energy and its large spread, there are large chromatic aberrations in the electron optics of the PEEM

Two-Photon Electron Emission



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Theory: Spatial distributions of two-photon excitation as a function of delay between the two excitation pulses (Movie)





Geometry of the system

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0.



Coherent Control of Two-Photon Electron max Emission in V-Shape Nanoantennas



Time-integrated electron current as a function of interpulse delay M.I. Stockman, Nano Lett. **5**, 2325-2329 (2005).

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LETTERS

Adaptive subwavelength control of nano-optical fields

Martin Aeschlimann¹, Michael Bauer², Daniela Bayer¹, Tobias Brixner³, F. Javier García de Abajo⁴, Walter Pfeiffer⁵, Martin Rohmer¹, Christian Spindler³ & Felix Steeb¹





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CONCLUSIONS

• Phase modulation of the excitation femtosecond pulse provides a functional degree of freedom necessary to control the spatial distribution of the local optical fields in nanosystems on the femtosecond temporal and nanometer spatial scale.

• Both the spectral composition and the phase modulation determine femtosecond-nanometer dynamics of local fields.

 For nonlinear photoprocesses, time-integral spatial distribution is controlled by both the pulse spectrum and its phase modulation.
Two-photon processes are locally enhanced at the optimum by a factor of up to 10⁷.

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"Synthetic Aperture Radar" in Nanoplasmonics

M. Durach, A. Rusina, K. Nelson, and M. I. Stockman, *Toward Full Spatio-Temporal Control on the Nanoscale*, arXiv:0705.0725 (2007); Nano Lett. **7**, 3145-3149 (2007)

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 $\frac{360^{\circ}}{\Phi} = \frac{\lambda}{X}$



Planar array of a phased-array antenna

Radar Basics: Phased Array Antenna (Synthetic Aperture Array, or Beam





 Θ s= beam steering



Space Surveillance Radar

U.S. Space Command's largest surveillance radar. The world's first large <u>phased array radar</u>, the AN/FPS-85 was constructed in the 1960s at Eglin Air Force Base, Florida.

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(Dreisatz)

 $\varphi = \frac{360^{\circ}}{2} * d* \sin \Theta_{s}$

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 φ = phase shift between two successive elements

d = distance between the radiating elements



APAR: Active Phased Array Radar

AESA: Active Electronically Scanned Array



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An interference fringe of two coherent EM sources is a line of

$$R_1 - R_2 = m\lambda$$
, $m = 0, \pm 1, \cdots$

It is a hyperbola. This is the first example of coherent control used by the British in operation *Oboe* during WWII to guide bombers over Germany in complete radio silence

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Problem: The pulses to control even simplest nanoplasmonic systems found by adaptive algorithms may be very complex.

How to determine a pulse that can localize optical energy at a given site of a nanosystem deterministically and robustly?

Solution: Time reversal approach

X. Li and M. I. Stockman, *Highly efficient spatiotemporal coherent control in nanoplasmonics on a nanometerfemtosecond scale by time reversal*, Phys. Rev. B **77**, 195109 (2008); arXiv:0705.0553

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LETTERS

Adaptive subwavelength control of nano-optical fields

Martin Aeschlimann¹, Michael Bauer², Daniela Bayer¹, Tobias Brixner³, F. Javier García de Abajo⁴, Walter Pfeiffer⁵, Martin Rohmer¹, Christian Spindler³ & Felix Steeb¹







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Focusing Beyond the Diffraction Limit with Far-Field Time Reversal

Geoffroy Lerosey, Julien de Rosny, Arnaud Tourin, Mathias Fink*



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reversed and sent back from the TRM. (**E**) In full line are shown the focusing spots obtained around antennas 3 and 4. Their typical width is $\lambda/30$. Thus, antennas 3 and 4 can be addressed independently. The focal spots obtained when there are no copper wires are shown for comparison (dashed-dotted line). All maxima have been normalized by scaling factors in the ratios: 1 (red and blue dashed-dotted lines), 2.2 (red full line), 2.5 (blue full line).

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Georgia State University Atlanta, GA 30303-3083 Nanoplasmonic Energy Localization, Time Reversal, and Coherent Control

X. Li and M. I. Stockman, *Highly efficient spatiotemporal coherent control in nanoplasmonics* on a nanometer-femtosecond scale by time reversal, Phys. Rev. B **77**, 195109 (2008); arXiv:0705.0553

Idea of time reversal for subwavelength EM-wave localization:

G. Lerosey, J. de Rosny, A. Tourin, and M. Fink, *Focusing Beyond the Diffraction Limit with Far-Field Time Reversal*, Science **315**, 1120-1122 (2007).

A. Derode, A. Tourin, J. de Rosny, M. Tanter, S. Yon, and M. Fink, *Taking Advantage of Multiple Scattering to Communicate with Time-Reversal Antennas*, Phys. Rev. Lett. 90, 014301 (2003).



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PRL 104, 203901 (2010) PHYSICAL REVIEW LETTERS

week ending 21 MAY 2010

Resonant Metalenses for Breaking the Diffraction Barrier

Fabrice Lemoult, Geoffroy Lerosey,* Julien de Rosny, and Mathias Fink

Institut Langevin, ESPCI ParisTech & CNRS, Laboratoire Ondes et Acoustique, 10 rue Vauquelin, 75231 Paris Cedex 05, France (Received 8 January 2010; revised manuscript received 14 April 2010; published 18 May 2010)

> 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.

DOI: 10.1103/PhysRevLett.104.203901

(19, 19).

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



(d) Focal spot obtained after far field time reversal

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(1,1),

(2,3),

(5,6),

and

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Schematic of proposed local excitation with adiabatic cones

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

F. De Angelis, G. Das, P. Candeloro, M. Patrini, M. Galli, A. Bek, M. Lazzarino, I. Maksymov, C. Liberale, L. C. Andreani, and E. Di Fabrizio, *Nanoscale Chemical Mapping Using Three-Dimensional Adiabatic Compression of Surface Plasmon Polaritons*, Nature Nanotechnology **5**, 67-72 (2009).





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nature photonics

Active spatial control of plasmonic fields

Bergin Gjonaj^{1*}, Jochen Aulbach¹, Patrick M. Johnson¹, Allard P. Mosk², L. Kuipers¹ and Ad Lagendijk¹

¹FOM-Institute for Atomic and Molecular Physics AMOLF, Science Park 104, 1098 XG Amsterdam, The Netherlands, ²Complex Photonic Systems, Faculty of Science and Technology, and MESA+ Institute for Nanotechnology, University of Twente, PO Box 217, 7500 AE Enschede, The Netherlands. *e-mail: b.gjonaj@amolf.nl



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dx.doi.org/10.1021/nl2023299 pubsacs.org/NanoLett

LETTER

Femtosecond Nanofocusing with Full Optical Waveform Control

Samuel Berweger,[†] Joanna M. Atkin,[†] Xiaoji G. Xu, Robert L. Olmon, and Markus B. Raschke^{*}





CONCLUSIONS

•In nanoplasmonic systems, optical energy concentrates on the nanoscale in "hot spots" whose characteristic size is limited by the minimum size of the metal nanofeatures

•The local optical field enhancement at the hot spots may be very large, up to five orders of magnitude in intensity

•The local optical fields evolve in time on the femtosecond scale, potentially on the attosecond scale

•The optical field nanolocalization is coherently controllable by pulse shaping

•Time reversal gives a convenient and powerful tool to determine the pulse shaping

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Attosecond nanoplasmonic-field microscope

MARK I. STOCKMAN^{1,2*}, MATTHIAS F. KLING², ULF KLEINEBERG³ AND FERENC KRAUSZ^{2,3*}

¹Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303, USA ²Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Straße 1, D-85748 Garching, Germany ³Ludwig-Maximilians-Universität München, Department für Physik, Am Coulombwall 1, D-85748 Garching, Germany *e-mail: mstockman@gsu.edu; ferenc.krausz@mpq.mpg.de

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Energy of the Fermi-edge photoelectron is $(\sim 100\pm 10)$ eV. The local potential of the instantaneous plasmonic fields at the instant of the attosecond pulse arrival adds to the kinetic energy of electrons, acting as a local electrostatic (van de Graaf) accelerator



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Escape (dwelling) time from the local field region: τ_d Oscillation period: *T*

Local field-pulse duration: τ_p

$$v_d = \sqrt{2(\hbar\omega_X - W_f)/m} = 5.4 \times 10^8 \text{ cm/s}$$

Instantaneous regime: $\tau_d \ll T$ $\tau_d = 180 \frac{\text{as}}{\text{nm}}$, $\hbar \omega_{\text{XUV}} = 95 \text{ eV}$

$$E_{XUV} = \hbar\omega_X - W_f + e\phi(\mathbf{r}, t_X)$$

In this case the XUV-electron energy is defined solely by the local, instantaneous electrostatic potential

Short Course Nanoplasmonics SPIE Photonics West 2012 http://www.phy-astr.gsu.edu/stockman E-mail: mstockman@gsu.edu Lecture 3 p.45 1/24/2012 7:32 PM Novel Regime of Electron Emission from Nanoplasmonic Systems

- The time of flight through the region of local fields for ~100 eV XUV electrons is ~300 as and much less then plasmonic near-IR period. Consequently, electrons are electrostatically accelerated by the instantaneous local field electric potential
- Escape (dwelling) time of electron from the local field region: τ_d Optical oscillation period: *T*

Local field-pulse duration: τ_p XUV photon energy: $\hbar \omega_X$

Escape velocity: $v_d = \sqrt{2(\hbar\omega_X - W_f)/m} = 5.4 \times 10^8 \text{ cm/s}$

Instantaneous regime: $\tau_d \ll T$ $\tau_d = 180 \frac{\text{as}}{\text{nm}}, \quad \hbar \omega_x = 95 \text{ eV}$ The XUV photoelectron energy E_e :

$$E_e = \hbar \omega_X - W_f + e \phi(\mathbf{r}, t_X)$$

In this case the XUV-electron energy is defined solely by the local, instantaneous electrostatic potential

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Uncertainty Principle and Real-Time Measurement of Local Fields

For any practical purposes, from $E_e = \hbar \omega_x - W_f + e\phi(\mathbf{r}, t_x)$ it follows that uncertainty of the local potential $\Delta \phi = \frac{\hbar}{e} \Delta \omega_x$ where $\Delta \omega_x$ is the spectral bandwidth of the XUV pulse. The Heisenberg uncertainty relation is $\hbar \Delta \omega_x \Delta t_x \ge \frac{\hbar}{2}$ The precision of the instance of measurement is limited by $\Delta t = \Delta t_x$ Thus, the fundamental limitation on the measurement of $\phi(\mathbf{r}, t)$ is

$$\Delta \phi \cdot \Delta t \ge \frac{\hbar}{2e}$$
 and $\Delta \phi = \frac{\hbar}{e} \Delta \omega_X$

Thus, the instance and the potential cannot be (precisely) measured simultaneously, and the XUV pulse should not be too broadband and too short (though it should have as high energy as possible).

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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 nanoplasmonic oscillations with nm spatial and ~200 as temporal resolution



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

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nature

High-harmonic generation by resonant plasmon field enhancement

Seungchul Kim¹*, Jonghan Jin¹*, Young-Jin Kim¹, In-Yong Park¹, Yunseok Kim¹ & Seung-Woo Kim¹



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Figure 3 | Scanning electron microscope image of the nanostructure used for high-harmonic generation. Bow-tie elements were arranged in a twodimensional, 36×15 array with an area of $10 \ \mu m \times 10 \ \mu m$. The inset shows the magnified image of a single bow-tie element with the important dimensions marked. Owing to the high magnification, edge lines are seen blurred by multiple scattering of electrons in imaging.





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NEWS & VIEWS

ATTOSECOND PHYSICS

An easier route to high harmony

Mark I. Stockman

The generation of ultrashort light pulses by atomic ionization and recombination doesn't come cheap. But by niftily exploiting the play of light on a nanostructured surface, it can be done on a table-top.

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Figure 1 | **Stripping on the table-top.** The bow-tie-shaped gold nanoantennas used by Kim *et al.*¹ develop electric-field strengths in the gap

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Quantum Nanoplasmonics: Surface Plasmon Amplification by Stimulated Emission of Radiation (SPASER)

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THEORETICAL APPROACH

Because the characteristic size of a spaser is much smaller than the wavelength, the quasistatic approximation in field equations is valid. Surface plasmon field equations and boundary conditions in a material-independent form, where S_n are eigenvalues and φ_n are eigenfunctions:

$$\frac{\partial}{\partial \mathbf{r}} \theta(\mathbf{r}) \frac{\partial}{\partial \mathbf{r}} \varphi_n(\mathbf{r}) = s_n \frac{\partial^2}{\partial \mathbf{r}^2} \varphi_n(\mathbf{r}),$$

where $\theta(\mathbf{r} \in metal) = 1$ and $\theta(\mathbf{r} \in elsewhere) = 0$
 $\varphi_n(x, y, 0) = \varphi_n(x, y, L_z) = 0,$ and
 $\frac{\partial}{\partial x} \varphi_n(x, y, z) \Big|_{x=0, L_x} = \frac{\partial}{\partial y} \varphi_n(x, y, z) \Big|_{y=0, L_y} = 0.$

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Spectral parameter: $s(\omega) = \left[1 - \varepsilon(\omega) / \varepsilon_h\right]^{-1}$

Frequency ω_n and decay rate γ_n of surface plasmons:

$$\operatorname{Re}[s(\omega_n)] = s_n, \ \gamma_n = \frac{\operatorname{Im}[s(\omega_n)]}{s'_n}, \ \text{where} \ s'_n = \frac{d\operatorname{Re}[s(\omega_n)]}{d\omega_n}$$

Quasielectrostatic Hamiltonian of an inhomogeneous dispersive nanosystem:

$$H = \frac{1}{4\pi} \int_{-\infty}^{\infty} \frac{d \left[\omega \,\varepsilon(\mathbf{r}, \omega) \right]}{d\omega} \mathbf{E}(\mathbf{r}, \omega) \mathbf{E}^{+}(\mathbf{r}, \omega) \frac{d\omega}{2\pi} d^{3}r$$

where is $\mathbf{E}(\mathbf{r},\omega) = -\nabla \phi(\mathbf{r},\omega)$ the electric field operator.

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Quantized potential operator as an expansion over surface plasmons:

$$\hat{\phi}(\mathbf{r},t) = \sum_{n} \sqrt{\frac{4\pi\hbar s_n}{\varepsilon_h}} \,\varphi_n(\mathbf{r}) \Big[a_n e^{-i\omega_n t} + a_n^+ e^{i\omega_n t} \Big]$$

where a_n^+ and a_n^- are the surface plasmon creation and annihilation operators. With this, the Hamiltonian becomes $H = \sum_n \hbar \omega_n \left(a_n^+ a_n + \frac{1}{2} \right)$

The interaction Hamiltonian of the surface plasmons and two-level systems (quantum dots) of the active medium:

$$H' = \sum_{a} \mathbf{d}^{(a)} \nabla \hat{\phi}(\mathbf{r}_{a}, t)$$

Using the perturbation theory, kinetic equation for the population number of surface plasmons in an *n*-th mode is:

$$\frac{dN_n}{dt} = \left(B_n - \gamma_n\right)N_n + A_n$$

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The Einstein stimulated emission coefficient is

$$B_{n} = \frac{4\pi}{3\hbar} \frac{s_{n} \left| \mathbf{d}_{10} \right|^{2} p_{n} q_{n}}{\varepsilon_{h} s_{n}' \gamma_{n}} > \gamma_{n} \implies \frac{\left| \mathbf{d}_{10} \right|^{2} N_{QD} Q}{\hbar R^{3} \Gamma} \ge 1$$

Here p_n is the spatial overlap factor, q_n is the spectral overlap factor between the eigenmode intensity and the population inversion, Γ is the spectral width of the gain medium emission, $Q = \omega_n / \gamma$ is the plasmon quality factor.

$$p_n = \int \left[\nabla \varphi_n(\mathbf{r}) \right]^2 \left[\rho_1(\mathbf{r}) - \rho_0(\mathbf{r}) \right] d^3 r, \ q_n = \int F(\omega) \left[1 + \left(\omega - \omega_n \right)^2 \gamma_n^2 d\omega \right] d^3 r$$

Spaser gain

$$\alpha_n = \frac{B - \gamma_n}{\gamma_n}$$

shows how many times faster the surface plasmons are born by the stimulated emission than they decay.

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The local RMS field produced by spaser: $E(\mathbf{r}) = \left\langle \left[\nabla \phi(\mathbf{r}) \right]^2 \right\rangle^{1/2}$ is calculated as:

$$E(\mathbf{r}) = E_n(\mathbf{r}) \left(N_n + \frac{1}{2} \right)$$
, where

$$E_n(\mathbf{r}) = \left\{ \frac{4\pi\hbar s_n}{\varepsilon_h s'_n} \left\langle \left[\nabla \varphi_n(\mathbf{r}) \right]^2 \right\rangle \right\}^{1/2}$$

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RESULTS

The resonant nanoparticle is an "engineered" V-shape. The material is silver; the spatial scale is 2-5 nm/grid unit.



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Calculated gain for thin (three monolayers of quantum dots) active medium

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Eigenmodes with highest yields for the spectral maximum at 1.2 eV



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Quantum Theory of SPASER

Quantization of the SP system, valid in the quasistatic regime for times shorter than the SP lifetime $\tau_n \equiv 1/\gamma_n$, is carried out by using the following approximate expression for the energy H of an electric field $\mathbf{E}(\mathbf{r}, t)$, which is obtained for a dispersive system by following Ref. [13],

$$H = \frac{1}{4\pi T} \int_{-\infty}^{\infty} \frac{d[\omega \varepsilon(\mathbf{r}, \omega)]}{d\omega} \mathbf{E}(\mathbf{r}, \omega) \mathbf{E}(\mathbf{r}, -\omega) \frac{d\omega}{2\pi} d^3 r.$$
(2)

The electric field operator⁴⁰ of the quantized SPs is⁴

$$\mathbf{E}(\mathbf{r}) = -\sum_{n} A_n \nabla \varphi_n(\mathbf{r}) (\hat{a}_n + \hat{a}_n^{\dagger}) , \quad A_n = \left(\frac{4\pi\hbar s_n}{\varepsilon_d s'_n}\right)^{1/2}$$

 $s(\omega) = \varepsilon_d / [\varepsilon_d - \varepsilon_m(\omega)]$ is Bergman's spectral parameter, ε_d is the permittivity of the ambient dielectric, and $\varepsilon_m(\omega)$ is the metal permittivity.

The spaser Hamiltonian has the form

$$H = H_g + \hbar \sum_n \omega_n \hat{a}_n^{\dagger} \hat{a}_n - \sum_p \mathbf{E}(\mathbf{r}_p) \mathbf{d}^{(p)} ,$$

where H_q is the Hamiltonian of the gain medium.

D. J. Bergman and M. I. Stockman, Surface Plasmon Amplification by Stimulated Emission of Radiation: Quantum Generation of Coherent Surface Plasmons in Nanosystems, Phys. Rev. Lett. 90, 027402-1-4 (2003) **E-man:** mstockman@gsu.edu SPIE PHOLOMICS West 2012

024004 - 1 - 13(2010)

Introducing a rate constant Γ_{12} to describe the polarization relaxation and a difference $n_{21}^{(p)} = \rho_{22}^{(p)} - \rho_{11}^{(p)}$ as the population inversion on this spasing transition, we derive an equation of motion for the non-diagonal element of the density matrix as

$$\dot{\bar{\rho}}_{12}^{(p)} = -\left[i\left(\omega - \omega_{12}\right) + \Gamma_{12}\right]\bar{\rho}_{12}^{(p)} + in_{21}^{(p)}\Omega_{12}^{(a)*} , \qquad (4)$$

where $\Omega_{12}^{(p)} = -A_n \mathbf{d}_{12}^{(p)} \nabla \varphi_n(\mathbf{r}_p) a_{0n}/\hbar$ is the Rabi frequency for the spasing transition in a pth chromophore, and $\mathbf{d}_{12}^{(p)}$ is the corresponding transitional dipole element.

$$\dot{\bar{n}}_{21}^{(p)} = -4 \operatorname{Im} \left[\bar{\rho}_{12}^{(p)} \Omega_{21}^{(p)} \right] - \gamma_2 \left(1 + n_{21}^{(p)} \right) + g \left(1 - n_{21}^{(p)} \right)$$

$$\dot{a}_{0n} = [i(\omega - \omega_n) - \gamma_n] a_{0n} + i \sum_p \rho_{12}^{(p)*} \Omega^{(p)_{12}}$$

As in Schawlow-Towns theory of laser-line width, this spontaneous emission of SPs leads to the diffusion of the phase of the spasing state. This defines width γ_s of the spasing line as

$$\gamma_s = \frac{\sum_p \left(1 + n_{21}^{(p)}\right) \gamma_2^{(p)}}{2(2N_p + 1)} \ . \tag{8}$$

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Theory of Spaser in Stationary Regime

Physically, the spaser action is a result of spontaneous symmetry breaking when the phase of the coherent SP field is established from the spontaneous noise. Mathematically, the spaser is described by homogeneous differential Eqs. (4)-(6) derived and solved in Sec. II B. These equations become homogeneous algebraic equations for the stationary (CW) case. These equations always have a trivial, zero solution. However, when their determinant vanishes, they also possess a nontrivial solution describing spasing, whose condition is

$$(\omega_s - \omega_n + i\gamma_n)^{-1} \times$$

$$(\omega_s - \omega_{21} + i\Gamma_{12})^{-1} \sum_p \left| \tilde{\Omega}_{12}^{(p)} \right|^2 n_{21}^{(p)} = -1 ,$$

$$(9)$$

where ω_s is the spasing frequency, $\tilde{\Omega}_{12}^{(p)} = -A_n \mathbf{d}_{12}^{(p)} \nabla \varphi_n(\mathbf{r}_p) / \hbar$ is the single-plasmon Rabi fre-

quency, $\mathbf{d}_{12}^{(p)}$ is the transition dipole moment of a *p*th chromophore, $\varphi_n(\mathbf{r}_p)$ is the electric potential of the spasing mode at the position this chromophore, γ_n

$$n_{21}^{(p)} = (g - \gamma_2) \times$$

$$\left\{ g + \gamma_2 + 4 \left| \Omega_{12}^{(p)} \right|^2 / \left[(\omega_s - \omega_{21})^2 + \Gamma_{12}^2 \right] \right\}^{-1} ,$$
(10)

Short Course Nanoplasmonics SPIE Photonics West 2012 <u>arXiv:0908.3559</u> Journal of Optics, 024004-1-13 (2010).

From the imaginary part of Eq. (10) we immediately find the spasing frequency

$$\omega_s = \left(\gamma_n \omega_{21} + \Gamma_{12} \omega_n\right) / \left(\gamma_n + \Gamma_{12}\right) \quad , \tag{11}$$

which generally does not coincide with either the gain transition frequency ω_{21} or the SP frequency ω_n , but is between them (this is a frequency walk-off phenomenon similar to that of laser physics). Substituting Eq. (11) back to Eqs. (10)-(11), we obtain a system of equations

$$\frac{(\gamma_n + \Gamma_{12})^2}{\gamma_n \Gamma_{12} \left[(\omega_{21} - \omega_n)^2 + (\Gamma_{12} + \gamma_n)^2 \right]} \times \sum_p \left| \tilde{\Omega}_{12}^{(p)} \right|^2 n_{21}^{(p)} = 1 , \qquad (12)$$

$$n_{21}^{(p)} = (g - \gamma_2) \times \left[g + \gamma_2 + \frac{4N_n \left| \tilde{\Omega}_{12}^{(p)} \right|^2 (\Gamma_{12} + \gamma_n)}{(\omega_{12} - \omega_n)^2 + (\Gamma_{12} + \gamma_n)^2} \right]^{-1} . (13)$$

This system defines the stationary (CW) number of SPs per spasing mode N_n .

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SPASER Threshold Condition [Consistent with PRL 90, 027402-1-4 (2003)]:

Since $n_{21}^{(p)} \leq 1$, from Eqs. (12), (13) we immediately obtain a necessary condition of the existence of spasing,

$$\frac{(\gamma_n + \Gamma_{12})^2}{\gamma_n \Gamma_{12} \left[(\omega_{21} - \omega_n)^2 + (\Gamma_{12} + \gamma_n)^2 \right]} \sum_p \left| \tilde{\Omega}_{12}^{(p)} \right|^2 \ge 1 . \qquad \frac{\text{arXiv:0908.3559}}{\text{Journal of Optics,}}$$
(14) 024004-1-13 (2010).

This expression is fully consistent with [4]. The following order of magnitude estimate of this spasing condition has a transparent physical meaning and is of heuristic value:

$$\frac{d_{12}^2 Q N_{\rm c}}{\hbar \Gamma_{12} V_n} \gtrsim 1,$$

The spasing is essentially a quantum effect. It is nonrelativistic: does not depend on c

(15)

of Optics,

where $Q = \omega/\gamma_n$ is the quality factor of SPs, V_n is the volume of the spasing SP mode, and N_c is the number of gain medium chromophores within this volume. Deriving this estimate, we have neglected the detuning, i.e., set $\omega_{21} - \omega_n = 0$.

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Required Gain for Spasing



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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

<u>arXiv:0908.3559</u> Journal of Optics, 024004-1-13 (2010).

Spectral line width $\propto 1/N_{SP}$

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Georgia State University Atlanta, GA 30303-3083 This invention changed civilization as we know it

This invention is used in more copies than all others combined

This is the most valuable element of nanotechnology: nanoamplifier, which in c-MOS technology pairs forms a digital nanoamplifier and bistable element for information processing





- Major problem: any quantum amplifier (laser and spaser) in a CW regime possesses exactly **zero amplification** (it is actually a condition for the CW operation).
- We have proposed to set the spaser as a nanoamplifier in two ways:
- 1. In transient mode (before reaching the CW regime), the spaser still possesses non-zero amplification
- 2. With a saturable absorber, the spaser can be bistable. There are two stable states: with the zero coherent SP population ("logical zero") and with a high SP population that saturates the absorber ("logical one" state). Such a spaser will function as a threshold (digital) amplifier

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Amplification in Spaser without a Saturable Absorber



Stationary pumping

Pulse pumping

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11 May 2009 / Vol. 17, No. 10 / OPTICS EXPRESS 8548

Towards the lasing spaser: controlling metamaterial optical response with semiconductor quantum dots

E. Plum,^{1,*} V. A. Fedotov,¹ P. Kuo,² D. P. Tsai,³ and N. I. Zheludev^{1,**}



Fig. 1. Photonic metamaterial hybridized with semiconductor quantum dots. The insets show the metamaterial unit cell and an SEM image of the actual metamaterial structure.

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Wavelength (nm)



doi:10.1038/nature08318

nature

LETTERS

Demonstration of a spaser-based nanolaser

M. A. Noginov¹, G. Zhu¹, A. M. Belgrave¹, R. Bakker², V. M. Shalaev², E. E. Narimanov², S. Stout^{1,3}, 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 $\lambda = 525$ nm and Q = 14.8; the inner and the outer circles represent the 14-nm core and the 44-nm shell, respectively. The field strength colour scheme is shown on the right.

¹Center for Materials Research, Norfolk State University, Norfolk, Virginia 23504, USA. ²School of Electrical & Computer Engineering and Birck Nanotechnology Center, Purdue University, West Lafayette, Indiana 47907, USA. ³Materials Science and Engineering Department, Cornell University, Ithaca, New York 14850, USA.

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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 1.1×10^{-12} cm². The emission and excitation spectra were measured in a spectrofluorometer at low fluence.

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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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Lasing in metal-insulator-metal sub-wavelength Jacob Plasmonic waveguides

Martin T. Hill^{1*}, Milan Marell¹, Eunice S. P. Leong², Barry Smalbrugge¹, Youcai Zhu¹, Minghua Sun², Peter J. van Veldhoven¹, Erik Jan Geluk¹, Fouad Karouta¹, Yok-Siang Oei¹, Richard Nötzel¹, Cun-Zheng Ning², and Meint K. Smit¹

¹COBRA Research Institute, Technische Universiteit Eindhoven, Postbus 513, 5600 MB Eindhoven, The Netherlands ² Department of Electrical Engineering, Arizona State University, Tempe AZ 85287, USA

*m.t.hill@ieee.org



Fig. 1. Structure of cavity formed by a rectangular semiconductor pillar encapsulated in Silver. (a) Schematic showing the device layer structure. (b) Scanning electron microscope image showing the semiconductor core of one of the devices. The scale bar is 1 micron.

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Fig. 2. Spectra and near field patterns showing lasing in devices. (a) Above threshold emission spectrum for 3 micron long device with semiconductor core width d~130nm (\pm 20nm), with pump current 180 µA at 78K. Inset: emission spectra for 20 (green), 40 (blue) and 60 (red) µA, all at 78K. (b) Lasing mode light output (red crosses), integrated luminescence (blue circles), versus pump current for 78K. (c) Actual near field pattern (in x-y plane) for 6 micron (d = 130nm) device captured with 100x, 0.7 NA long working distance microscope objective and infrared camera, the scale bar is 2 micron, for below threshold 30 µA, and (d) above threshold 320 µA. (e) Simulated vertical (z) component of the Poynting vector taken at 0.7 microns below the pillar base, shows most emitted light at ends of device. (f) Spectra for a 6 micron long device with d~310nm at 298K, pulsed operation (28 ns wide pulses, 1MHz repetition). Spectra for peak currents of 5.2mA (red), 5.9mA (green) and 7.4mA (blue), (currents were estimated from the applied voltage pulse amplitude). The spectra for 5.9 and 7.4 mA are offset from 0 for clarity. Inset shows the total light collected by the spectrometer from the device for currents ranging from 0 to 10mA.

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LETTERS

Plasmon lasers at deep subwavelength scale

Rupert F. Oulton¹*, Volker J. Sorger¹*, Thomas Zentgraf¹*, Ren-Min Ma³, Christopher Gladden¹, Lun Dai³, Guy Bartal¹ & Xiang Zhang^{1,2}



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Atlan[®] **Room-temperature sub-diffraction-limited** plasmon laser by total internal reflection

Ren-Min Ma^{1†}, Rupert F. Oulton^{1†}, Volker J. Sorger¹, Guy Bartal¹ and Xiang Zhang^{1,2}*



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A room-temperature semiconductor spaser operating near 1.5 µm

R. A. Flynn,¹ C. S. Kim,¹ I. Vurgaftman,¹ M. Kim,¹ J. R. Meyer,¹ A. J. Mäkinen,¹ K. Bussmann,² L. Cheng,³ F.-S. Choa,³ and J. P. Long^{4,*}

¹Optical Sciences Division, Naval Research Laboratory, Washington, DC 20375, USA ²Materials Science and Technology Division, Naval Research Laboratory, Washington, DC 20375, USA ³Department of Electrical Engineering, University of Maryland Baltimore County, Baltimore, Maryland 21250, USA ⁴Chemistry Division, Naval Research Laboratory, Washington, DC 20375, USA



Fig. 2. Log-log plot comparing the strength of diagnostic radiation from the spaser (circles) and a control device with air above the Au-film waveguide (squares) vs the instantaneous pumpintensity. The control exhibited only spontaneous emission. Solid line shows expectations from our wave-equation model. Inset: L-L plot with linear axes showing threshold at ~60 kW/cm² pump.

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dx.doi.org/10.1021/nl2022477 Nano Lett. 2011, 11, 4256-4260

Plasmonic Green Nanolaser Based on a Metal–Oxide–Semiconductor Structure

Chen-Ying Wu,[†] Cheng-Tai Kuo,[†] Chun-Yuan Wang,[†] Chieh-Lun He,[‡] Meng-Hsien Lin,[‡] Hyeyoung Ahn,^{*,§} and Shangjr Gwo*,+,+

[†]Department of Physics and [†]Institute of Nanoengineering and Microsystems, National Tsing-Hua University, Hsinchu, Taiwan 30013, Republic of China

[§]Department of Photonics and Institute of Electro-optical engineering, National Chiao-Tung University, Hsinchu, Taiwan 30010, Republic of China



E-mail: mstockman@gsu.edu



Plasmonic distributed feedback lasers at telecommunications wavelengths

Milan J.H. Marell,¹ Barry Smalbrugge,¹ Erik Jan Geluk,¹ Peter J. van Veldhoven,² Beatrix Barcones,² Bert Koopmans,² Richard Nötzel,² Meint K. Smit,¹ and Martin T. Hill^{1,*}



COMPARISON OF THE SPASER AND NANOLASERS (POLARITONIC SPASERS)

Spaser's size is below the skin depth (25 nm for gold and silver and 16 nm for aluminum) in all directions, limited from the bottom by the nonlocality length of ~2 nm. The modal volume in the spaser is ~1000 times smaller than in a typical nanolaser. Spaser generates very high local fields on the nanoscale, emits very little light. It can generate dark modes.

Spaser spases at ~10 -100 SPs per mode. Feedback in the spaser is much stronger than in nanolaser, inversely proportional to modal volume. Energy dissipation is proportionally smaller

The optical fields in the spaser are inversely proportional to its linear size and are realistically $\sim 10^{6}$ - 10^{8} V/cm. This leads to much stronger feedback and much faster switching than for nanolasers, giving 10-100 THz bandwidth

E-mail: mst

Nanolasers are of two types. One has a ~10 nm confinement in the transverse direction but is ~microns in the longitudinal direction. The second type is a patch-type nanolaser which is fractions of micron in all directions. Nanolasers do generate light.

Nanolaser lases at $\sim 10^4 - 10^6$ quanta per mode, proportional to its modal volume.

Conventional laser lases at $\sim 10^{18} - 10^{20}$ quanta per mode

The feedback of the nanolasers is much weaker than in the spaser inversely proportionally to its modal volume. Correspondingly the maximum modulation rate and bandwidth of the spaser are proportionally slower.



Potential Applications of the Spaser and Nanolasers

The spaser is a source of optical fields and optical energy on the nanoscale with the same size on the order of magnitude as nanoparticles and biological molecules (proteins, nuclear acids, viruses ...)

Radiation of a spaser can be fed to any waveguide

The spaser can function as an ultrafast amplifier and memory cell with the same form factor as MOSFET, but ~1000 times faster

Other possible applications of spaser: local optical excitation of nanosystems, nanomodification (ultrastrong local fields), and active labels for bio-objects and ultramicroscopy. A nanolaser is a source of optical fields with cross extension on the nanoscale. However the total size of a nanolaser is much greater than a biological molecule such as DNA or protein and their complexes

Radiation of a nanolaser can be fed into most waveguides

A nanolaser has a much weaker feedback and would be much slower and larger amplifier

As a source of optical energy on the nanoscale, a nanolaser has an uphill competition with adiabatic plasmonic concentrators (tapers)

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- 1. SPASER is a nanoscopic quantum generator of coherent and intense local optical fields
- 2. SPASER can also serve as a nanoscale 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.
- 3. SPASER has been experimentally observed recently. This experiment in in excellent qualitative agreement with theory. The observed spaser is single mode. Its pumping curve is linear with a threshold. Its linewidth is inversely proportional to pumping rate.
- 4. Two plasmon-polariton spasers (nanolasers) have been designed. In contrast to spaser, their length is on micron order (transverse mode size is nanometric). Their emission is multimode.
- 5. The most promising applications of the SPASER are a ultrafast nanoamplifier, local optical energy source, and active nano-label.

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