

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

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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:



Spectrally, surface plasmon resonances in complex systems occupy a very wide frequency band

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

Surface plasmon relaxation times are in ~10-100 fs range

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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 Department of Physics and Astronomy 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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In a <u>nonlinear</u> case, the phase is a controlling factor.

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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

LETTERS

NANO

2005 Vol. 5, No. 6 1123–1127

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

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Received April 10, 2005; Revised Manuscript Received May 6, 2005

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)



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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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nature

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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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Planar array of a phased-array antenna





 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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(Sinussatz)

(Dreisatz)

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

X = d∗sin Θ_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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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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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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nature

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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Fig. 2. Focusing beyond the diffraction limit. (**A** and **B**) show the signal received at one antenna of the TRM when a 10-ns pulse is sent from antennas 3 and 4, respectively, of the microstructured array. The signals in (A) and (B) look considerably different, although antennas 3 and 4 are only λ /30 apart. (**C** and **D**) show the time compression obtained at antennas 3 and 4, respectively, when the eight signals coming from antennas 3 and 4 are time-

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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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.

> > a)

DOI: 10.1103/PhysRevLett.104.203901

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

0.5

-0.5

0

1

b)

Normalized Amplitude

d)

0.75



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



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 $-\frac{\lambda}{15} - \frac{\lambda}{20} - \frac{\lambda}{30} - \frac{\lambda}{80} \circ \frac{\lambda}{30} - \frac{\lambda}{20} - \frac{\lambda}{30} \circ \frac{\lambda}{30} - \frac{\lambda}{20} - \frac{\lambda}{15}$ (d) Focal spot obtained after far field time reversal
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25

2

Time (µs)

3

4

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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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Active spatial control of plasmonic fields

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dx.doi.org/10.1021/nl2023299 pubs.acs.org/NanoLett

LETTER

Femtosecond Nanofocusing with Full Optical Waveform Control

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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*}

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Published online: 3 September 2007; doi:10.1038/nphoton.2007.169

nature photonics | VOL 1 | SEPTEMBER 2007 | www.nature.com/naturephotonics

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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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Regimes of Electron Emission

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

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http://www.phy-astr.gsu.edu/stockman E-mail: mstockman@gsu.edu Lecture 3 p.45 7/10/2015 10:08 AM 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: $\tau_d \ll T$ $\tau_d = 180 \frac{\text{as}}{\text{nm}}, \ \hbar \omega_x = 95 \text{ eV}$ 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 $\Delta \phi = -\frac{\hbar}{2} \Delta \omega_X$ where $\Delta \omega_{v}$ 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 $\Delta t = \Delta t_v$ 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 delay with respect to the infrared-visible excitation pulse (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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Plasmonic generation of ultrashort extreme-ultraviolet light pulses

In-Yong Park^{1†}, Seungchul Kim^{1†}, Joonhee Choi^{1†}, Dong-Hyub Lee¹, Young-Jin Kim¹, Matthias F. Kling², Mark I. Stockman³ and Seung-Woo Kim¹*



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

- 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).
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Ge

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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).

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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: $s(\omega) = \left[1 - \varepsilon_m(\omega) / \varepsilon_d\right]^{-1}$

Frequency ω_n and decay rate γ_n of surface plasmons:

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

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:

$$\hat{\varphi}(\mathbf{r},t) = \sum_{n} \sqrt{\frac{4\pi\hbar s_n}{\varepsilon_d s_n}} \phi_n(\mathbf{r}) \left[a_n e^{-i\omega_n t} + a_n^+ e^{i\omega_n t} \right]$$

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

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

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.

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

Spaser gain
$$\alpha_n = \frac{B - \gamma_n}{M}$$

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

 γ_n

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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: $E(\mathbf{r}) = E_n(\mathbf{r}) \left(N_n + \frac{1}{2} \right)^{1/2}$, 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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Department of Physics and Astronomy 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 **E**(**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) 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

arXiv:0908.3559

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

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 *p*th chromophore, and $\mathbf{d}_{12}^{(p)}$ is the corresponding transitional dipole element.

$$\dot{\bar{n}}_{21}^{(p)} = -4 \operatorname{Im} \left[\bar{\rho}_{12}^{(p)} \Omega_{21}^{(p)} \right] - \gamma_2 \left(1 + n_{21}^{(p)} \right) + g \left(1 - n_{21}^{(p)} \right)$$
$$\dot{a}_{0n} = \left[i \left(\omega - \omega_n \right) - \gamma_n \right] a_{0n} + i \sum \rho_{12}^{(p)*} \Omega^{(p)_{12}}$$

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)

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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.

(15)

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 $\omega_{21} - \omega_n = 0$.

It is nonrelativistic: does not depend on *c*

Short Course Nanoplasmonics 2015 http://www.phy-astr.gsu.edu/stockman E-mail: mstockman@gsu.edu Lecture 3 p.64 7/10/2015 10:08 AM Gain of bulk medium required for spasing and for loss compensation by gain: M. I. Stockman, *Spaser Action, Loss Compensation, and Stability in Plasmonic Systems with Gain,* Phys. Rev. Lett. **106**, 156802-1-4 (2011); Phil. Trans. R. Soc. A **369**, 3510 (2011).



Stationary (CW) hysics and Astronomy hiversity spaser regime Inversion vs. 3-3083 Plasmon number pumping rate This quasilinear dependence N_n vs. pumping rate (b) $N_n(g)$ is a result of the very 400 $\hbar \omega_s = 1.2 \text{ eV}$ n_{21} (a) 0 Ť strong feedback in spaser due to 300 2×10^{12} 4×10^{12} 1.5 eV the small modal volume $g(s^{-1})$ 200 -0.1 1.8 eV 100 2.2 eV -0.2 1×10^{13} Spectral shape 5×10^{12} -0.3 ± of spaser line g (s-1) M. I. Stockman, *The* $\hbar \Gamma_s$ (meV) $S(\omega)$ Spaser as a (d) 5 0.1 (c) 4 Nanoscale Quantum Spaser radiation 3 $g=1.01g_{th}$ Generator and Plasmon fluorescenc 0.05 Line width vs. 2 Ultrafast Amplifier, pumping rate 1 $\times 10^{3}$ Journal of Optics 12, 5×10^{12} 1×10^{13} 1.2 1.3 1.1024004-1-13 (2010) g (s-1) $\hbar\omega$ (eV) Spectral shape Spectral shape $S(\omega)$ $S(\omega)$ of spaser line (e) of spaser line (f) 150 Spectral line width $\propto 1/N_{SP}$ $g=2g_{th}$ $g=10g_{th}$ 3

100

50

 $\times 10^{5}$

1.1

×100

1.2

ħω (eV)

1.3

 $\times 10$

1.3

1.2

 $\hbar\omega$ (eV)

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 $\times 10^{4}$

1.1

GeorgaState 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





To be revisited at the end

P. Packan et al., in 2009 IEEE International Electron Devices Meeting (IEDM), *High Performance 32nm Logic Technology Featuring Second Generation High-K* + *Metal Gate Transistors (Baltimore, MD, 2009), Vol. IEDM09-662, p. 28.4.1-28.4.4*

Abstract:

A 32nm logic technology for high performance microprocessors is described. 2nd generation high-k + metal gate transistors provide record drive currents at the tightest gate pitch reported for any 32 nm or 28nm logic technology. NMOS drive currents are 1.62mA/um Idsat and 0.231mA/um Idlin at 1.0V and 100nA/um Ioff. PMOS drive currents are 1.37mA/um Idsat and 0.240mA/um Idlin at 1.0V and 100nA/um Ioff. The impact of SRAM cell and array size on Vccmin is reported.



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The spaser as a Nanoamplifier

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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Scaling of Spaser

Field in spaser:
$$E \sim \frac{\hbar\omega}{R^{3/2}} \sqrt{N_p} \sim \left(\frac{R}{10 \text{ nm}}\right)^{-3/2} \sqrt{N_p} \frac{\text{MV}}{\text{cm}}$$

Heat per flop: $H = \hbar \omega N_p$

Threshold:
$$g \ge g_{th}, g_{th} = \frac{\omega}{c\sqrt{\varepsilon_d}} \frac{\operatorname{Re} s(\omega) \operatorname{Im} \varepsilon_m(\omega)}{1 - \operatorname{Re} s(\omega)}, \quad s(\omega) = \frac{\varepsilon_d}{\varepsilon_d - \varepsilon_m(\omega)}$$

Switching time: $\tau \sim \left(\frac{10 \text{ nm}}{R}\right)^3 \frac{100}{N_p}$ fs

Conclusion: Spaser is orders of magnitude more efficient (less heat per flop) and much faster than transistor. It can operate close to the quantum limit.

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Amplification in Spaser without a Saturable Absorber



Stationary pumping

Pulse pumping

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Experimental Observations of Spaser

doi:10.1038/nature08318

nature

LETTERS

Demonstration of a spaser-based nanolaser

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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

(in false colour), with $\lambda = 5$: circles represent the 14-nm of strength colour scheme is sh

b, Transmission electron microscope image of Au core. **c**, Scanning electron microscope image of Au/silica/dye core–shell nanoparticles. **d**, Spaser mode

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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.



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

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



Lasing in metal-insulator-metal sub-wavelength plasmonic waveguides

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> Received 14 Apr 2009; revised 8 Jun 2009; accepted 9 Jun 2009; published 18 Jun 2009 22 June 2009 / Vol. 17, No. 13 / OPTICS EXPRESS 11107



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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doi:10.1038/nature08364

LETTERS

nature

Plasmon lasers at deep subwavelength scale

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A room-temperature semiconductor spaser operating near 1.5 μm

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25 April 2011 / Vol. 19, No. 9 / OPTICS EXPRESS 8954



Plasmonic Nanolaser Using Department of Georgia State Epitaxially Grown Silver Film Atlanta, GA 3 Vu-Jung Lu,¹* Jisun Kim,²* Hung-Ying Chen,¹ Chihhui Wu,² Nima Dabidian,² Charlotte E. Sanders,² Chun-Yuan Wang,¹ Ming-Yen Lu,³ Bo-Hong Li,⁴ Xianggang Qiu,⁴ Georgia<u>State</u> University

60 K

78 K 100 K

700



Wen-Hao Chang,⁵ Lih-Juann Chen,³ Gennady Shvets,² Chih-Kang Shih,²† Shangjr Gwo¹†

Having developed epitaxially grown, atomically smooth Ag films as a scalable plasmonic platform, we report a SPASER under CW operation with an ultralow lasing threshold at liquid nitrogen temperature and a mode volume well below the 3D diffraction limit. The device has





with contrast fringes indicative of spatial coherence resulting from lasing. a.u., arbitrary units. (B) Temperaturedependent lasing thresholds of the plasmonic cavity. The L-L plots at the main lasing peak (510 nm) are shown with the corresponding linewidth-narrowing behavior when the plasmonic laser is measured at 8 (red) and 78 K (blue), with lasing thresholds of 2.1 and 3.7 kW/cm², respectively. (O Temperature-dependent lasing behavior from 8 to 300 K. (D) Second-order photon correlation function measurements at 8 K.

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Y.-J. Lu *et al.*, Nano Lett. **14**, 4381 (2014)

All-Color Plasmonic Nanolasers with Ultralow Thresholds: Autotuning Mechanism for Single-Mode Lasing

Yu-Jung Lu,[†] Chun-Yuan Wang,[†] Jisun Kim,[‡] Hung-Ying Chen,[†] Ming-Yen Lu,[∥] Yen-Chun Chen,[⊥] Wen-Hao Chang,[⊥] Lih-Juann Chen,[∥] Mark I. Stockman,^{§,#,¶} Chih-Kang Shih,^{*,‡} and Shangjr Gwo^{*,†}





JOURNAL OF OPTICS

doi:10.1088/2040-8978/14/11/114015

Georganstate Stimulated emission of surface plasmon polaritons on smooth and corrugated silver surfaces

J K Kitur, G Zhu, Yu A Barnakov and M A Noginov



Random Spaser



Figure 5. Emission spectra in the RB:PMMA film deposited on a roughened silver with surface roughness equal to 234 nm, pumped with 7 mJ (1), 13 mJ (2) and 20 mJ (3) laser pulses. Inset: stimulated emission threshold as a function of the surface roughness.

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Surface plasmon lasing observed in metal hole arrays

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See also: W. Zhou, M. Dridi, J. Y. Suh, C. H. Kim, D. T. Co, M. R. Wasielewski, G. C. Schatz, and T. W. Odom, *Lasing Action in Strongly Coupled Plasmonic Nanocavity Arrays*, Nature Nanotechnology **8**, 506-511 (2013)



FIG. 2. (a) Luminescence spectra as a function of pump power, plotted on a semilog scale. For increasing pump power the bandwidth of the luminescence increases until the device starts lasing. Above threshold, the emission of the non-lasing resonances starts to saturate at a maximum intensity. 80 mW corresponds to $\sim 11 \text{ kW/cm}^2$ (b) The output in the lasing peak and in the luminescence in the range of 1485 - 1600 nm. The power in the lasing peak shows a clear threshold (red).

Short Course NanoplasThe black line is a guide to the eye. The luminescence out-2015in semiconductor devices (blue).



Phys. Rev. Lett. **110**, **206802-1-5** (2013)

LETTERS PUBLISHED ONLINE: 16 JUNE 2013 | DOI: 10.1038/NNANO.2013.99

Lasing action in strongly coupled plasmonic nanocavity arrays

Wei Zhou^{1†}, Montacer Dridi², Jae Yong Suh², Chul Hoon Kim^{2,3†}, Dick T. Co^{2,3}, Michael R. Wasielewski^{2,3}, George C. Schatz² and Teri W. Odom^{1,2,3}*

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nature nanotechnology

Explosives detection in a lasing plasmon nanocavity

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ARTICLE

Received 28 Dec 2013 | Accepted 11 Aug 2014 | Published 23 Sep 2014

DOI: 10.1038/ncomms5953

A room temperature low-threshold ultraviolet plasmonic nanolaser

Qing Zhang¹, Guangyuan Li¹, Xinfeng Liu¹, Fang Qian², Yat Li³, Tze Chien Sum^{1,4}, Charles M. Lieber⁵ & Oihua Xiong^{1,6}

d 355 nm at RT L=15 m Ellc 105 = 2 μm Log₁₀ output power (mm) Emission Intensity (a.u.) Emission Intensity (a.u.) 3 Elc 10 0.2 0.4 ზი Pumping fluence (MW cm⁻²) 1 L⁻¹ (m⁻¹) b 355 nm, 10 ns, 100 KHz at RT 360 370 380 390 400 410 420 360 370 380 390 400 410 350 Emission wavelength (nm) Emission wavelength (nm)

375-nm

asing

b

С

GaN

AI

GaN

Purcel

Distance (nm)

-75 75

Distance (nm)

9630 -75

355 nm

SiO2

GaN

Al film

Figure 2 | Room temperature ultraviolet plasmonic lasing characterization. (a) Scanning electron microscopy (SEM) image of a GaN nanowire sitting on SiO₂/Al film. Inset: magnified scanning electron microscopy image of one end of the GaN nanowire. The nanowire length and diameter is 15 µm and 100 nm, respectively. (b) Schematic of optical measurement and polarization detection setup. *c* is defined as the orientation of nanowire. The incidence excitation laser is circular polarized and the focused laser beam can cover the whole nanowire. The emission scattered out from two ends is collected and the polarization property along and perpendicular to nanowire axis *c* is analysed. (c) Spontaneous emission of as-fabricated plasmonic device below lasing threshold at room temperature under a power fluence of 0.5 MW cm⁻². Arrows highlight the Fabry-Pérot peaks. The nanowire length is 2 µm. Inset: cavity mode spacing $\delta\lambda$ variation with nanowire length *L* (green dots). $\delta\lambda$ versus 1/*L* can be well fitted with a linear function (red curve), suggesting a high group index n_g ($n_g = \lambda^2/2L$) of 7.61 due to the high gain requirement of the plasmonic laser device. (d) Power-dependent emission spectra of the plasmonic devices. One sharp peak with a maximum full width at half maximum (FWHM) ~ 0.8 nm appears above the spontaneous emission background. The nanowire length is 15 µm. Inset: integrated emission versus pumping intensity. The S-shaped plot suggests the evolution from a spontaneous emission, amplified spontaneous emission to lasing process.

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ARTICLES PUBLISHED ONLINE: 28 SEPTEMBER 2014 | DOI: 10.1038/NPHYS3103

Ultrafast plasmonic nanowire lasers near the surface plasmon frequency

Themistoklis P. H. Sidiropoulos^{1*}, Robert Röder², Sebastian Geburt², Ortwin Hess¹, Stefan A. Maier¹, Carsten Ronning² and Rupert F. Oulton^{1*} b

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nature

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Figure 5 | Measured spectra versus double-pump pulse delay for the plasmonic nanowire laser and its Fourier transform. a, Normalized difference spectrum, $\Delta l(\lambda, \tau)/l_0(\lambda) - l(\lambda, \tau)/l_0(\lambda) - 1$, of the plasmonic nanowire laser for $\tau \ge 0$, where $l(\lambda, \tau)$ is the spectrum under double-pump excitation and $l_0(\lambda)$ is the single strong pump pulse spectrum. The two upper panels show the $\Delta l/l_0$ spectra for the pulse delays, $\tau = 2.0$ ps and $\tau = 3.1$ ps, indicating the increasing spectral modulation frequency with pulse delay. **b**, Fourier transform of each spectrum shown in **a** versus pulse delay. The white trend line follows $t = \tau - \tau_{on} \approx \tau_{m}$, indicating a turn-on time of $\tau_{on} = 1.1$ ps. The inset shows the amplitude decay of the Fourier transform along the white trend line, with linear fits (red lines) to the modulation

peaks. The presented data in this figure correspond to measurements at the http://www.ph highest pump power (situation i) shown in Fig. 4a.

Graphene spaser

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Georgia:

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³Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, D-85748 Garching, Germany (Dated: May 10, 2013)

We propose a graphene spaser, which is a coherent quantum generator of surface plasmons in nanostructured graphene. The plasmonic core of this spaser is a graphene monolayer nanopatch and its active (gain) element is a multi-quantum well system with a design similar to the design of an active element of quantum cascade laser. For realistic parameters of the multi-quantum well system, the spasing in graphene monolayer can be achieved at a finite doping of graphene and at a plasmon frequency, ≈ 0.15 eV, close to the typical frequency of intersubband transitions in multi-quantum well systems. The proposed graphene spaser will be an efficient source of intense and coherent nanolocalized fields in the mid-infrared spectral region with wide perspective applications in mid-infrared nanoscopy, nano-spectroscopy, and nano-lithography.

V. Apalkov and M. I. Stockman, Proposed Graphene Nanospaser; NPG: Light Sci. Appl. 3, e191 (2014).

Applications in Biomedicine: Why spaser is efficient as fluorescent, photothermal and photoacoustic agent? It does not saturate!

Absorption cross section as a function of the pumping rate for different

Spaser as Versatile Biomedical Tool

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(arXiv:1501.00342; Submitted)

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Spaser for biological applications

a, Schematic of spaser as multifunctional intracellular nanoprobe. b, Stimulated emission in spaser suspension. c, Radiation spectrum of spaser in suspension at 528 nm at different pump intensities. d, Spatially homogenous spaser's emission at a relatively low pump intensity (30 MW/cm², 120- μ m thick spaser's suspension); Middle: emission during the bubble formation around overheated spasers (150 MW/cm²); **Bottom:** "directional" spaser emission in the presence of two large bubbles (200 MW/cm²). Scale bars, 10 μ m.

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Photoacoustic (PA) and photothermal (PT) spectral microscopy of spasers. a, Absorption spectra, and linear and nonlinear PA spectra of spaser in suspension at laser energy fluence of 20 mJ/cm^2 and 150 mJ/cm^2 , respectively. **b**, PA signal dependence on laser pump energy fluence for dye and spaser in suspension and into cells. c-e, Images of single cancer cell with spasers loaded through endocytosis. (c)-scattering (dark field), (d) -photothermal (PT); (e)- stimulated emission for local irradiated cell zone in background of conventional fluorescence image. **f**, Spectral peak from single cancer cell with spasers at relatively low energy fluence (80 mJ/cm²). Scale bars, 10 μ m. g, Spectral peak single cancer cell with spasers showing spectral hole burning in stimulated emission spectra at moderate energy fluence (135 mJ/cm^2).

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Cells а е Spaser solution Emission Excitation Imaging Injection site Blood Cancer cell b Mouse ear Intact tissue Tissue with PA signal, a.u. spaser С 8-530 570 490 Wavelength, nm g d Spaser

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Short Course Nanoplasmonics 2015 Imaging of spasers in viable cells in vitro and in biotissue in vivo. a, Schematic in vitro. b, Optical transmission image. c, d, Fluorescence imaging using conventional optical source (lamp) of blood with cancer cells at depth of $\sim 1 \text{ mm}$ (top) and spaser emission from single cancer cell at depth of 1 mm (**bottom**). e, Schematic of intradermal injection of spaser suspension into top layer of mouse ear tissue. f, PA identification of spasers in ear tissue using laser spectral scanning (top). Laser parameters: beam diameter 15 μ m, fluence 20 mJ/cm². g, Spaser emission through $\sim 250 \ \mu m$ ear tissue. Pump parameters: beam diameter: 10 µm; intensity, 30 MW/cm².

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Demonstration of spaser as theranostic

agent. a, Cell viability tests for different spaser concentration using two various kits without (blue, green) and after (red) laser irradiation (100 mJ/cm², 1 Hz, 3min). Inset: intact cell (left) and cells labeled with spasers at different concentration (middle and right) after laser irradiation. **b**, Viability test for concentration 15.6 x 10^{-5} M as a function of laser exposure time (3s [3 pulses], 1 min, and 3 min); c, Viability test for concentration 15.6 x 10⁻⁵ M as a function of laser pulse number (1, 3 and 5) showing that even single laser pulse at fluence of 500 mJ/cm² is sufficient for significant damage of cancer cells labeled by spasers. The average SD for each column is 15-20%

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The most important technological application: Information processing

P. Packan et al., in 2009 IEEE International Electron Devices Meeting (IEDM), *High Performance 32nm Logic Technology Featuring Second Generation High-K* + *Metal Gate Transistors (Baltimore, MD, 2009), Vol. IEDM09-662, p. 28.4.1-28.4.4*

Abstract:

A 32nm logic technology for high performance microprocessors is described. 2nd generation high-k + metal gate transistors provide record drive currents at the tightest gate pitch reported for any 32 nm or 28nm logic technology. NMOS drive currents are 1.62mA/um Idsat and 0.231mA/um Idlin at 1.0V and 100nA/um Ioff. PMOS drive currents are 1.37mA/um Idsat and 0.240mA/um Idlin at 1.0V and 100nA/um Ioff. The impact of SRAM cell and array size on Vccmin is reported.

Processor speed :

 $f_{\rm max} = I_{\rm drive} / (C_{\rm Intercon} \Delta U) \sim 3 \, \rm GHz$

Transistor speed is not a limiting factor! Charging the interconnects is.

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Nanospaser with electric excitation ("pumping") does not exist as of today *yet*, but fundamentally it is entirely possible

ch_{annel}

Al

 $\hbar\omega_n$ (eV)

Spaser frequency

 $J_{OW}(\mu {\rm A})$

^{ùrrent} per o_{ne} gu_{anf}

D. Li and M. I. Stockman,

Quantum Limit, Phys. Rev. Lett.

200

150

100

50

plasmon in the mode

Pump curent per one

Concept of ~300 GHz processor unit with ~1% energy cost per flop

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110, 106803-1-5 (2013) Lecture 3 p.95 7/10/2015 10:08 AM

Georgia State University University Department of Physics and Astronomy Georgia State University Atlanta, GA 30303-3083 BRIEF CONCLUSIONS

- 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. SPASERs have been experimentally observed. The experiments are in an excellent qualitative agreement with theory
- 4. Numerous 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 non-saturable nano-label.

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END LECTURE 3