Photo Credit: I. Tsukerman, Seefeld, Austria, January, 2009

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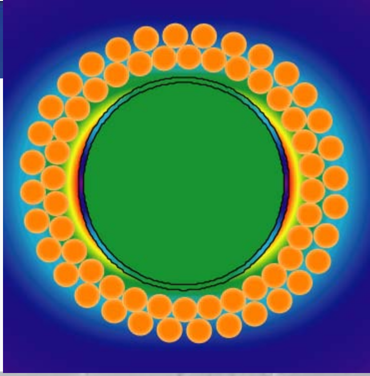

Complexity in Nanoplasmonics: Linear, Nonlinear, and Quantum

Mark I. Stockman

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

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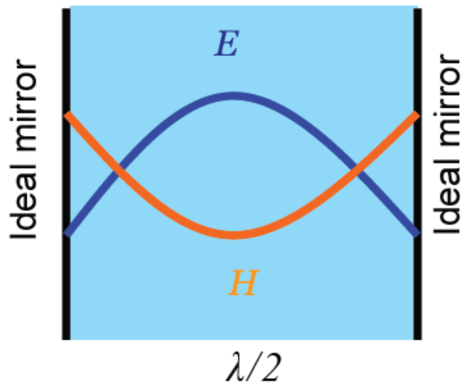
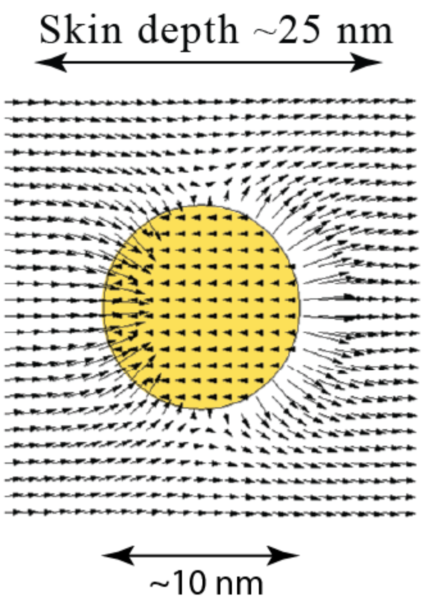
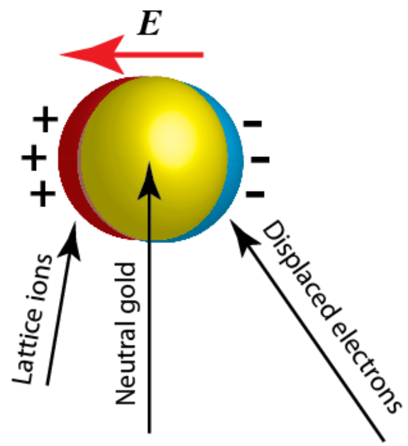
Nanoplasmonics is about nanolocalization of optical energy

Concentration of optical energy on the nanoscale



$\sim \frac{1}{2}\ \mu\mathrm{m} = 500\ \mathrm{nm}$





Photon: Quantum of **electromagnetic** field

Surface Plasmon: Quantum of **electromechanical** oscillator

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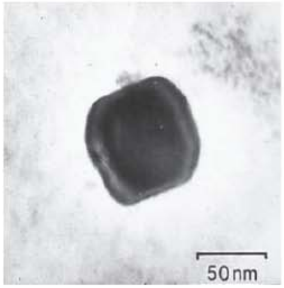
Lycurgus Cup (4th Century AD): Roman Nanotechnology

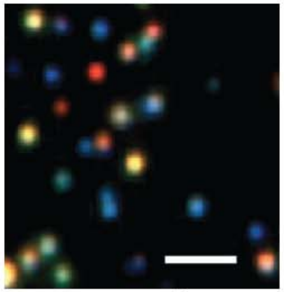


I. Freestone, N. Meeks, M. Sax, and C. Higgitt, *The Lycurgus Cup - a Roman Nanotechnology*, Gold Bull. **40**, 270-277 (2007

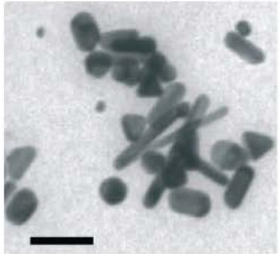
Nanoplasmonic colors are very bright. Scattering and absorption of light by them are very strong. This is due to the fact that all of the millions of electrons move in unison in plasmonic oscillations

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

Colors of Silver Nanocrystals and Gold Nanoshapes

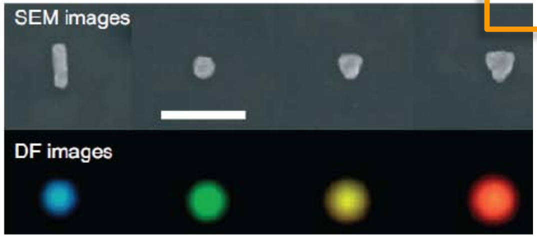


2 µm



100 nm

C. Orendorff, T. Sau, and C. Murphy, *Shape-Dependent* …, Small **2**, 636-639 (2006)



W. A. Murray and W. L. Barnes, *Plasmonic Materials*, Adv. Mater. **19**, 3771-3782 (2007) [Scale bar: 300 nm]

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Applications of Nanoplasmonics:

1. Ultrasensitive and express sensing and detection using both SPPs and SPs (LSPRs): see, e.g., J. N. Anker, W. P. Hall, O. Lyandres, N. C. Shah, J. Zhao, and R. P. Van Duyne, *Biosensing with Plasmonic Nanosensors*, Nature Materials 7, 442-453 (2008);
2. Near-filed scanning microscopy (or, nanoscopy): NSOM (SNOM)
3. Nanoantennas: Coupling of light to nanosystems. Extraction of light from LEDs and lasers [N. F. Yu, J. Fan, Q. J. Wang, C. Pflugl, L. Diehl, T. Edamura, M. Yamanishi, H. Kan, and F. Capasso, *Small-Divergence Semiconductor Lasers by Plasmonic Collimation*, Nat. Phot. 2, 564-570 (2008)]; nanostructured antennas for photodetectors and solar cells; heat-assisted magnetic memory [W. A. Challener *et al.*, Nat. Photon. 3, 220 (2009)]
4. Photo- and chemically stable labels and probes for biomedical research and medicine
5. Nanoplasmonic-based immunoassays and tests. Home pregnancy test (dominating the market), PSA test (clinic), troponin heart-attack test, and HIV tests (in trials)
6. Near perspective: Generation of EUV and XUV pulses [I.-Y. Park, S. Kim, J. Choi, D.-H. Lee, Y.-J. Kim, M. F. Kling, M. I. Stockman, and S.-W. Kim, *Plasmonic Generation of Ultrashort Extreme-Ultraviolet Light Pulses*, Nat. Phot. advance online publication (2011). doi: 10.1038/nphoton.2011.258.
7. Thermal cancer therapy: L. R. Hirsch, R. J. Stafford, J. A. Bankson, S. R. Sershen, B. Rivera, R. E. Price, J. D. Hazle, N. J. Halas, and J. L. West, *Nanoshell-Mediated Near-Infrared Thermal Therapy of Tumors under Magnetic Resonance Guidance*, Proc. Natl. Acad. Sci. USA 100, 13549-13554 (2003). C. Loo, A. Lowery, N. Halas, J. West, and R. Drezek, *Immunotargeted Nanoshells for Integrated Cancer Imaging and Therapy*, Nano Lett. 5, 709-711 (2005)

- M. I. Stockman, *Nanoplasmonics: The Physics Behind the Applications*, Phys. Today **64**, 39-44 (2011). DOI: 10.1063/1.3554315.
- M. I. Stockman, *Nanoplasmonics: Past, Present, and Glimpse into Future*, Opt. Express **19**, 22029-22106 (2011). 10.1364/OE.19.022029.

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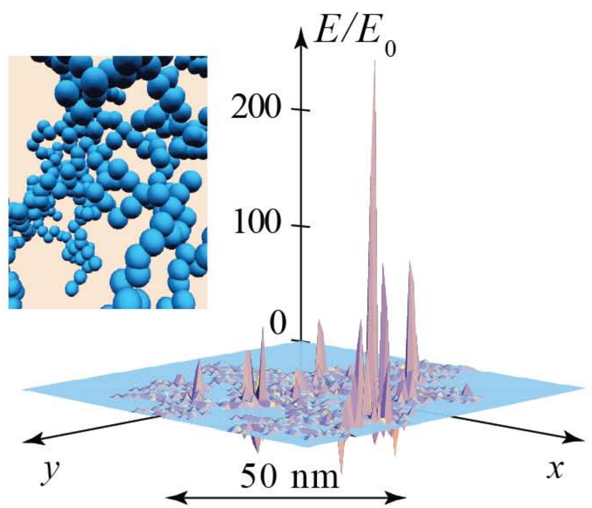
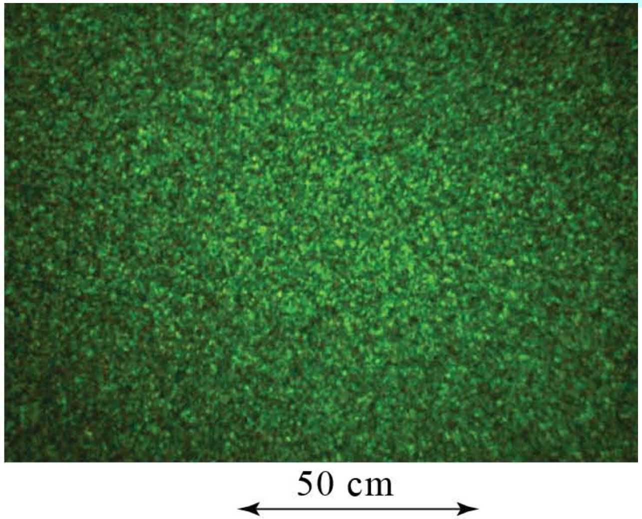
Plasmonic Near-Field **Hot Spots:** Happy 17th Anniversary!

- D. P. Tsai et al., Phys. Rev. Lett. **72**, 4149 (1994).
- M. I. Stockman et al., *Phys. Rev. Lett.* **75**, 2450 (1995)
- M. I. Stockman, L. N. Pandey, and T. F. George, Phys. Rev. B **53**, 2183 (1996)



M. I. Stockman, L. N. Pandey, and T. F. George, Phys. Rev. B 53, 2183 (1996).



Random scattering speckles

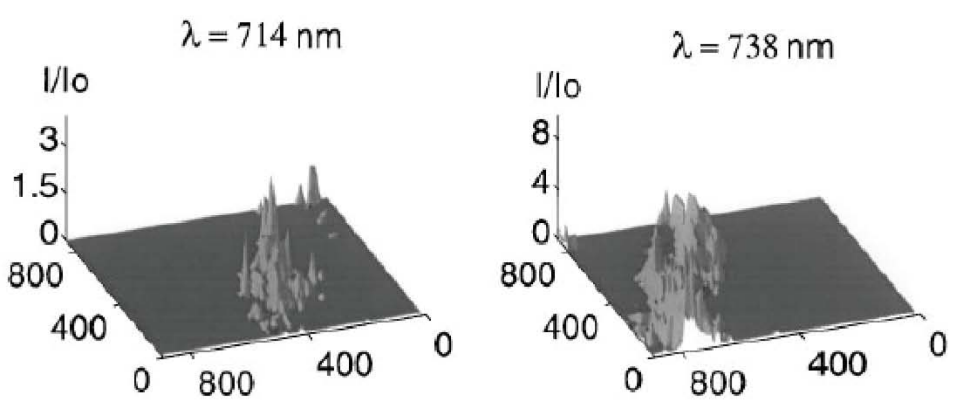
$$R_{Speckle} \sim \frac{\lambda\!\!\!^{-}}{A} L$$

$R_{Speckle}$ is speckle size

$\lambda\!\!\!^{-} \sim 100$ nm is reduced wave length

A is laser spot size,

L is distance to the screen



S. Gresillon et al., Phys. Rev. Lett. 82, 4520 (1999)

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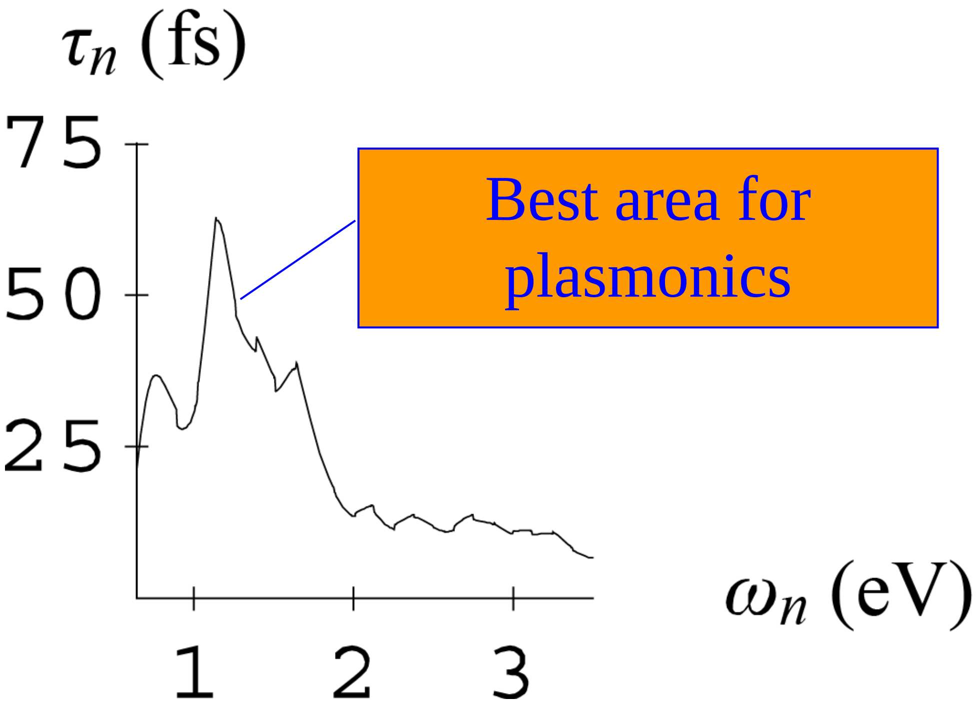
Nanoplasmonics is intrinsically ultrafast:



Spectrally, surface plasmon resonances in complex systems occupy a very wide frequency band; for gold and silver:

$$\Delta\omega \approx \omega_p / \sqrt{2} \approx 4\,\text{eV}$$

Including aluminum with plasmon responses in the ultraviolet, this spectral width increases to ~10 eV.

Corresponding rise time of plasmonic responses ~ 100 as

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

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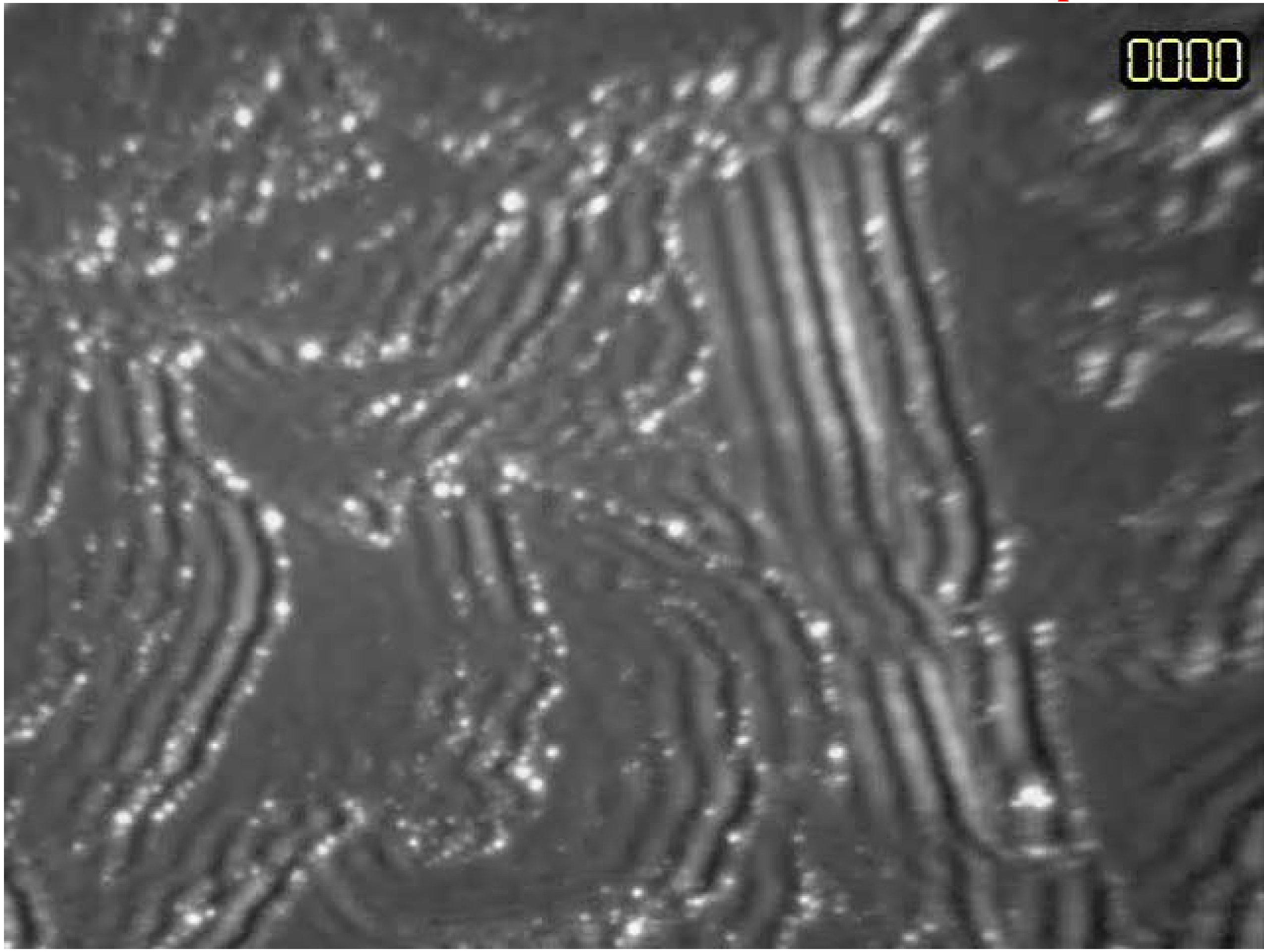
Localized SP hot spots and SPPs coexist in space and time on nanostructured surfaces

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).

250 as per frame



200 nm

30 femtoseconds from life of a nanoplasmonic system

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

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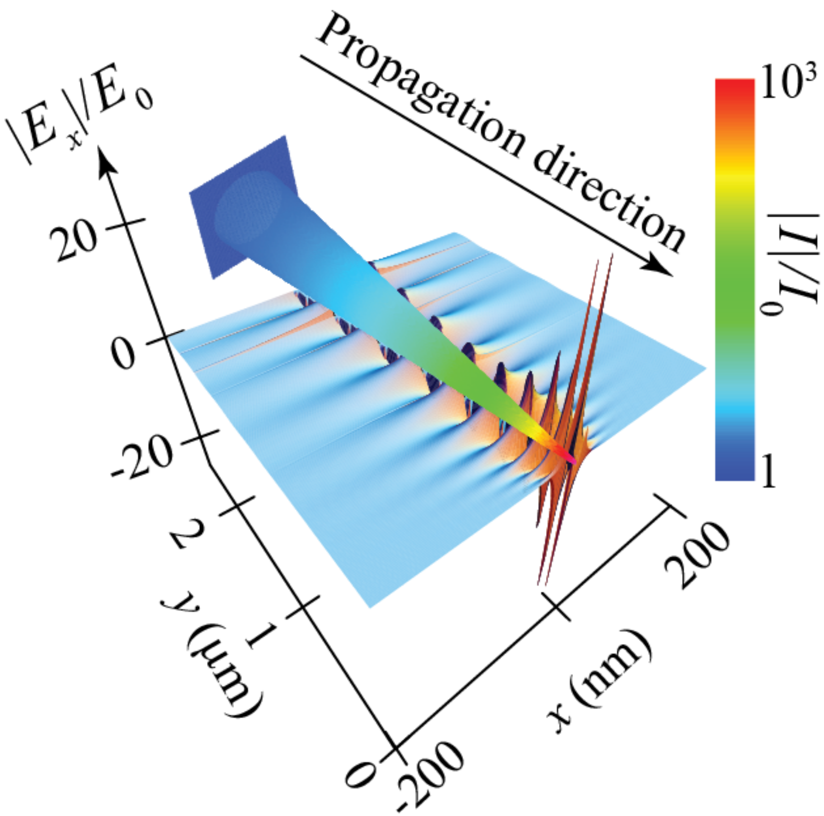
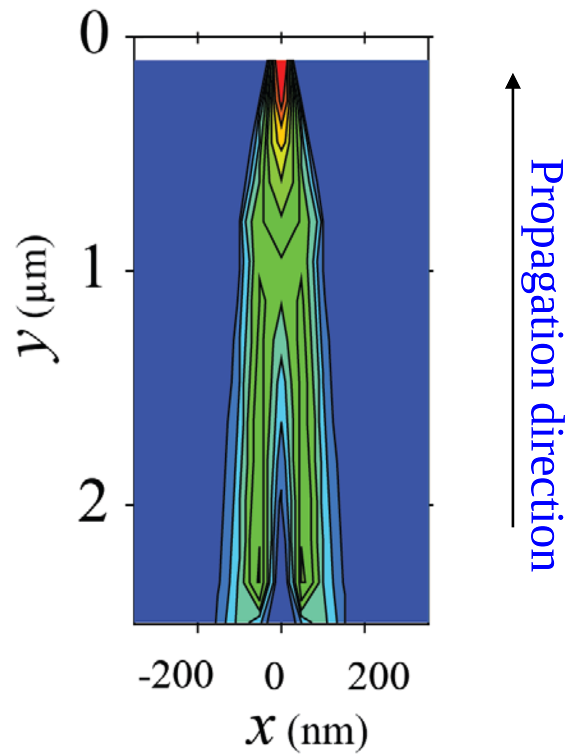
Field enhancement :

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

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

Adiabatic Compression





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

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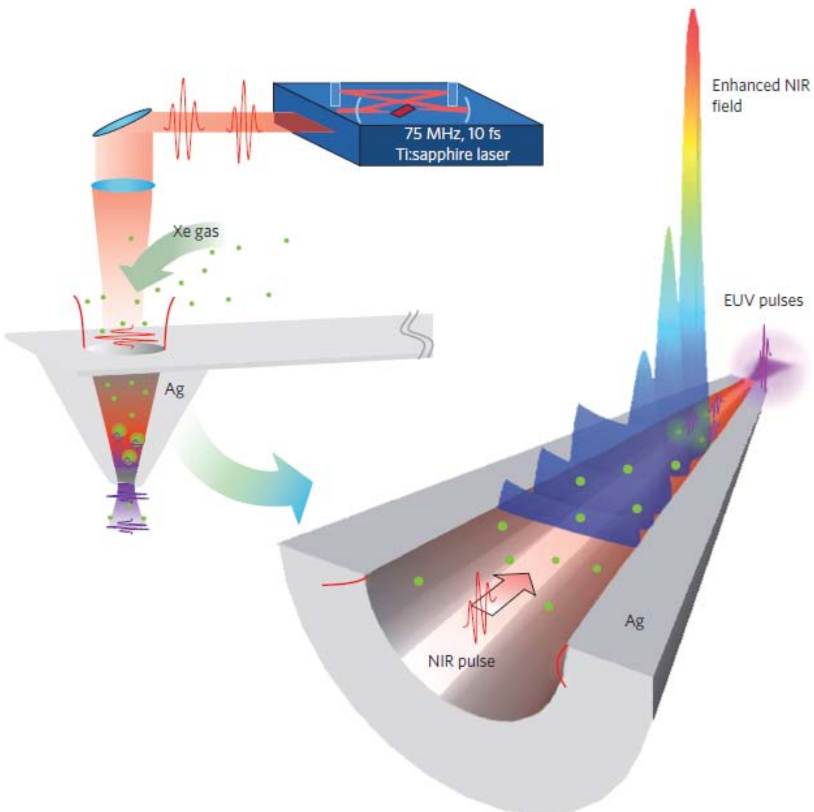
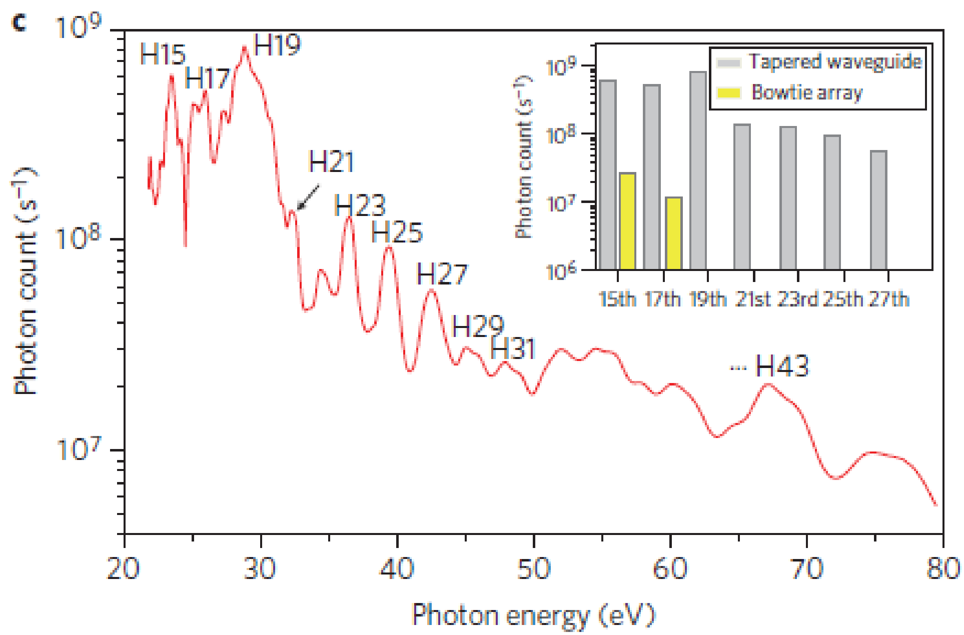
nature photonics
LETTERS
PUBLISHED ONLINE: 16 OCTOBER 2011 | DOI: 10.1038/NPHOTON.2011.258

Plasmonic generation of ultrashort extreme-ultraviolet light pulses

In-Yong Park[1†], Seungchul Kim[1†], Joonhee Choi[1†], Dong-Hyub Lee[1], Young-Jin Kim[1], Matthias F. Kling[2], Mark I. Stockman[3] and Seung-Woo Kim[1]*





[1]Ultrafast Optics for Ultraprecision Group, Korea Advanced Institute of Science and Technology (KAIST), Daejeon, 305-701, South Korea, [2]Max Planck Institute of Quantum Optics, Hans-Kopfermann-Str. 1, D-85748 Garching, Germany, [3]Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303, USA; †These authors contributed equally to this work as main authors. *e-mail: swk@kaist.ac.kr

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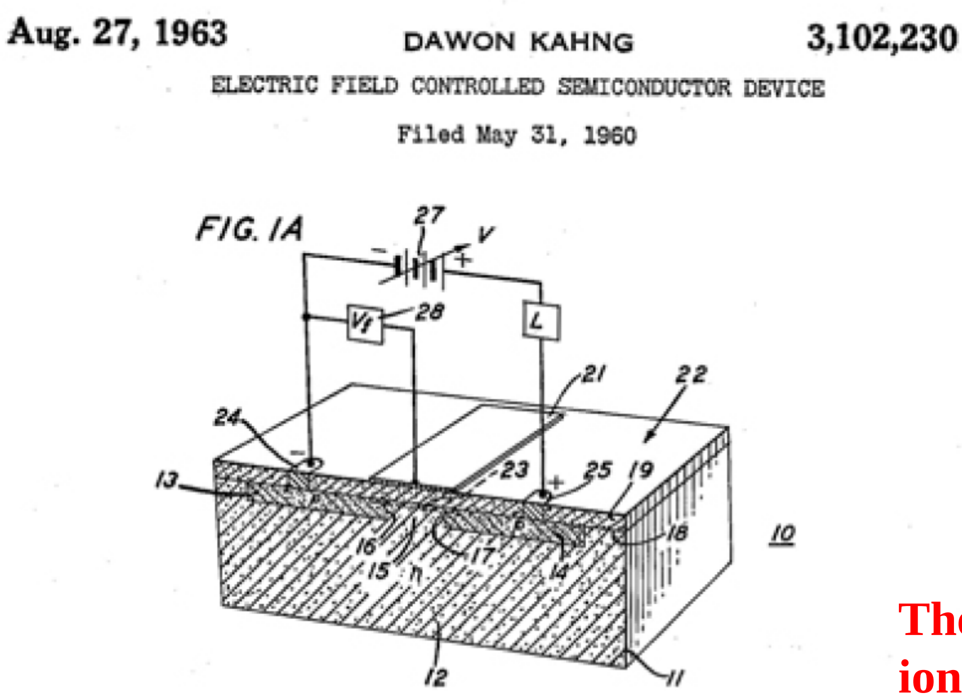
Goal of plasmonics is to keep, amplify, and manipulate optical energy on nanoscale, just like the transistor does with electric energy

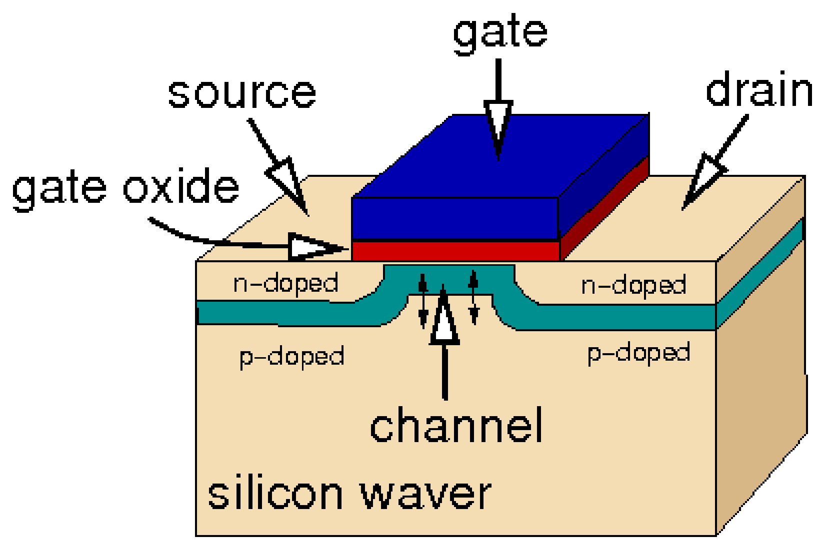
This invention changed civilization as we know it

This invention is used many more times than all others combined

This is the most valuable element of nanotechnology: nanoamplifier, whose pairs in c-MOS technology form digital bistable amplifiers and logical gates for information processing

MOSFET US Patent





Metal–Oxide Field–Effect Transistor (MOSFET)

Bandwidth ~ 10-100 GHz

Low resistance to ionizing radiation

The FET transistor is extremely vulnerable to ionizing radiation damage of the gate oxide, catastrophic degradation

Speed of a processor ~ 3 GHz is determined by electric interconnects

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

1. 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).
2. M. I. Stockman, *Spasers Explained*, Nat. Phot. **2**, 327-329 (2008) .
3. M. I. Stockman and D. J. Bergman, *Surface Plasmon Amplification by Stimulated Emission of Radiation (SPASER)*, USA Patent No. 7,569,188 (August 4, 2009)
4. M. I. Stockman, *Spaser as Nanoscale Quantum Generator and Ultrafast Amplifier*, Journal of Optics (JOPT) **12**, 024004-1-13 (2010).
5. M. A. Noginov, G. Zhu, A. M. Belgrave, R. Bakker, V. M. Shalaev, E. E. Narimanov, S. Stout, E. Herz, T. Suteewong, and U. Wiesner, *Demonstration of a Spaser-Based Nanolaser*, Nature **460**, 1110-1112 (2009).
6. M. T. Hill, M. Marell, E. S. P. Leong, B. Smalbrugge, Y. Zhu, M. Sun, P. J. van Veldhoven, E. J. Geluk, F. Karouta, Y.-S. Oei, R. Nötzel, C.-Z. Ning, and M. K. Smit, *Lasing in Metal-Insulator-Metal Sub-Wavelength Plasmonic Waveguides*, Opt. Express **17**, 11107-11112 (2009).
7. R. F. Oulton, V. J. Sorger, T. Zentgraf, R.-M. Ma, C. Gladden, L. Dai, G. Bartal, and X. Zhang, *Plasmon Lasers at Deep Subwavelength Scale*, Nature **461**, 629-632 (2009).
8. R.-M. Ma, R. F. Oulton, V. J. Sorger, G. Bartal, and X. Zhang, *Room-Temperature Sub-Diffraction-Limited Plasmon Laser by Total Internal Reflection*, Nat. Mater. **10**, 110-113 (2010). DOI 10.1038/nmat2919
9. R. A. Flynn, C. S. Kim, I. Vurgaftman, M. Kim, J. R. Meyer, A. J. Mäkinen, K. Bussmann, L. Cheng, F. S. Choa, and J. P. Long, *A Room-Temperature Semiconductor Spaser Operating near 1.5 Micron*, Opt. Express **19**, 8954-8961 (2011).
10. I. D. Leon and P. Berini, *Amplification of Long-Range Surface Plasmons by a Dipolar Gain Medium*, Nat. Phot. **4**, 382-387 (2010). doi:10.1038/nphoton.2010.37.

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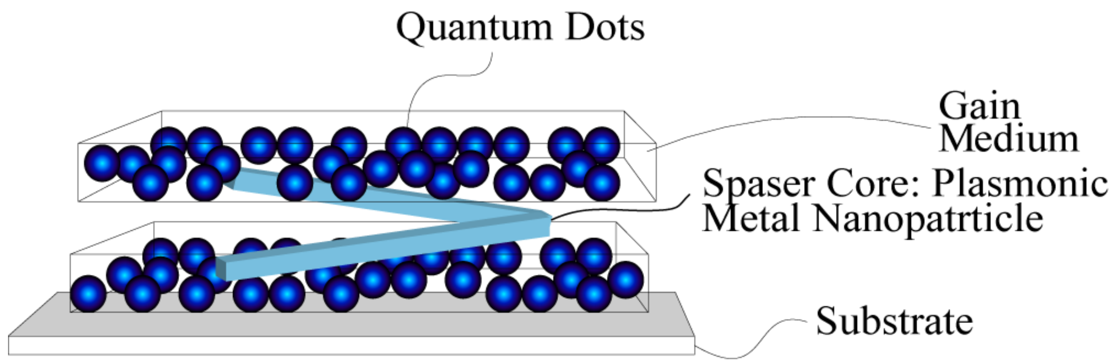
The original spaser geometry

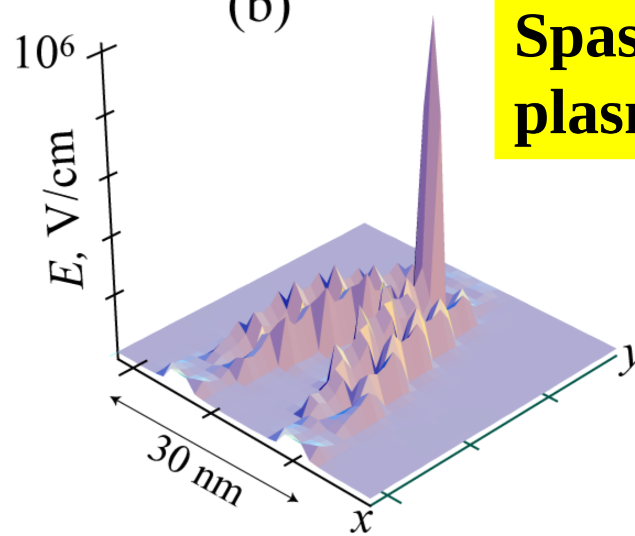
(a)



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).

Spaser field per one plasmon in the core

(b)



Spaser is the ultimately smallest quantum nano-generator

Spasers explained
nature photonics | VOL 2 | JUNE 2008 |

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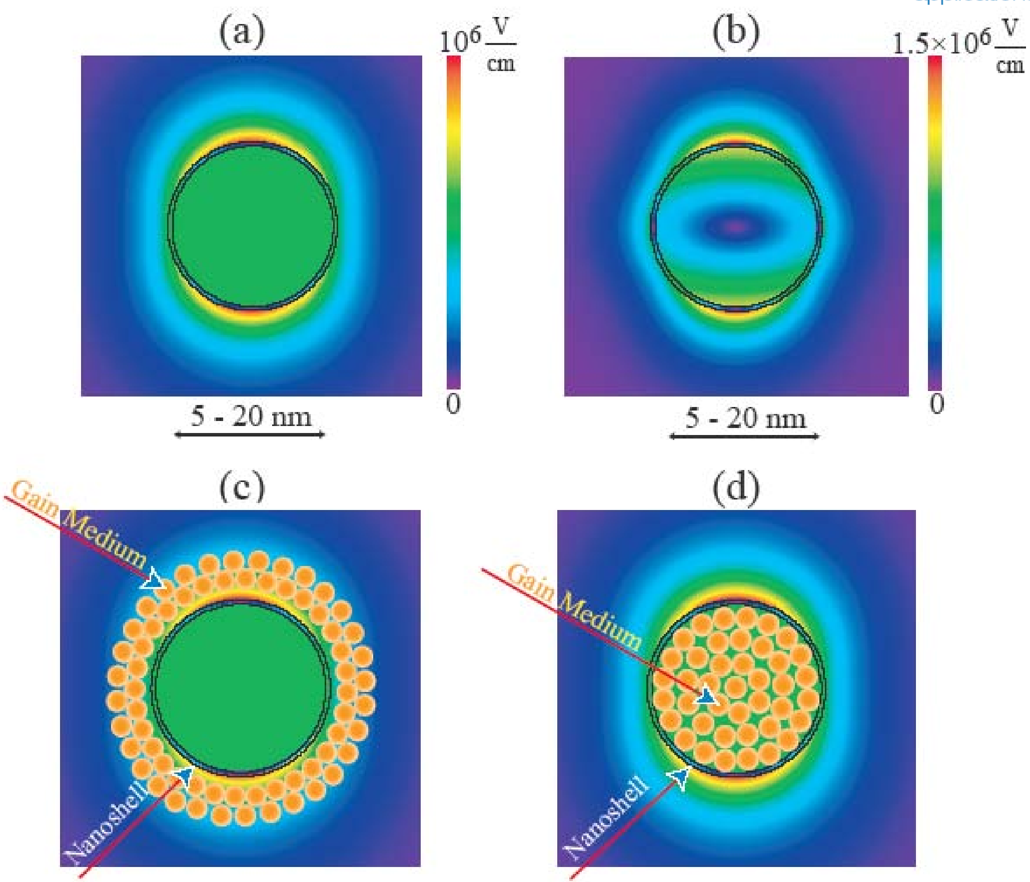
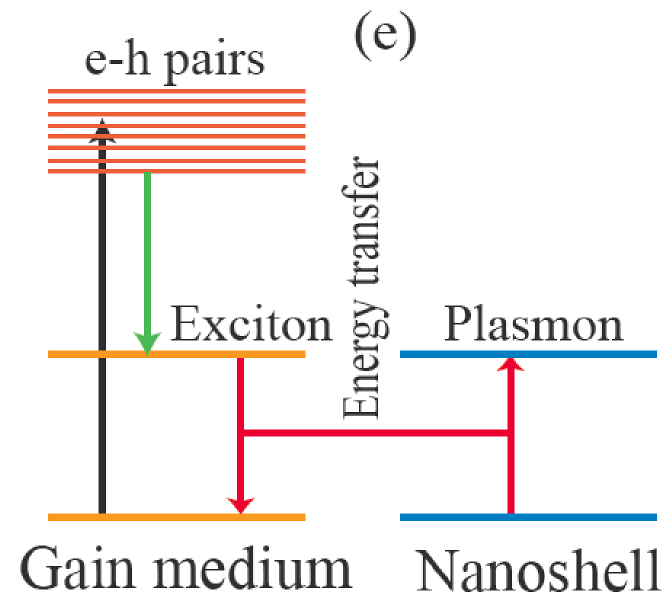
The spaser is a propos
number of leading lab ... e of
applications, including

For small nanoparticles, radiative loss is negligible.
Spaser is fully scalable





These equations of spaser theory are nonlinear describing a non-equilibrium second-order phase transition to spasing

ɔnomy

arXiv:0908.3559
Journal of Optics, **12**, 024004-1-13 (2010)

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],

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

The electric field operator[40] of the quantized SPs is[4]

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

$s(\omega) = \varepsilon_d/[\varepsilon_d - \varepsilon_m(\omega)]$ 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_g 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)

Nondiagonal element of density matrix (polarization):

$$\hbar\dot{\rho}_{12}^{(p)} = -\left[i(\hbar\omega - \varepsilon_{12}) + \hbar\Gamma_{12}\right]\rho_{12}^{(p)} + i\hbar n_{12}^{(p)} a_n \tilde{\mathbf{\Omega}}_{12}^{(p)}\ ,$$

$$\tilde{\mathbf{\Omega}}_{12}^{(p)} \equiv -A_n \mathbf{d}_{12}^{(p)} \nabla\varphi_n(\mathbf{r}_p)/\hbar$$

Diagonal elements of density matrix (inversion):

$$\dot{n}_{12}^{(p)} = -4\,\mathrm{Im}\left[a_n \tilde{\Omega}_{12}^{(p)} \rho_{12}^{(p)}\right] - \gamma_2(1 + n_{12}^{(p)}) + g(1 - n_{12}^{(p)})$$

SP field amplitude (semiclassical approximation):

$$\dot{a}_n = \left[i(\omega - \omega_n) - \gamma_n\right]a_n + i a_n \sum_p \tilde{\mathbf{\Omega}}_{12}^{(p)} \rho_{12}^{(p)*}$$

Spectral width of spaser emission (Schawlow-type formula)

$$\gamma_s = \frac{\mathbf{\Gamma}_0}{2N_p + 1}$$

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arXiv:0908.3559
Journal of Optics, **12**, 024004-1-13 (2010).

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

$$(\omega_s - \omega_n + i\gamma_n)^{-1} \times (\omega_s - \omega_{21} + i\Gamma_{12})^{-1} \sum_p \left|\tilde{\Omega}_{12}^{(p)}\right|^2 n_{21}^{(p)} = -1 \,, \tag{9}$$

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

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

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

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

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

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

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

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

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SPASER Threshold Condition [Consistent with original 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,

$$\frac{(\gamma_n + \Gamma_{12})^2}{\gamma_n \Gamma_{12}\left[(\omega_{21} - \omega_n)^2 + (\Gamma_{12} + \gamma_n)^2\right]} \sum_p \left|\tilde{\Omega}_{12}^{(p)}\right|^2 \geq 1 \, . \qquad (14)$$

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:

$$\frac{d_{12}^2 Q N_c}{\hbar \Gamma_{12} V_n} \gtrsim 1, \qquad (15)$$

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

arXiv:0908.3559
Journal of Optics, **12**, 024004-1-13 (2010).

The spasing is essentially a quantum effect.
It is non-relativistic: does not depend on *c*
The spasing condition does not directly contain gain per cm and the Purcell factor [E. M. Purcell, Phys Rev **69**, 681 (1946)] but is related to them

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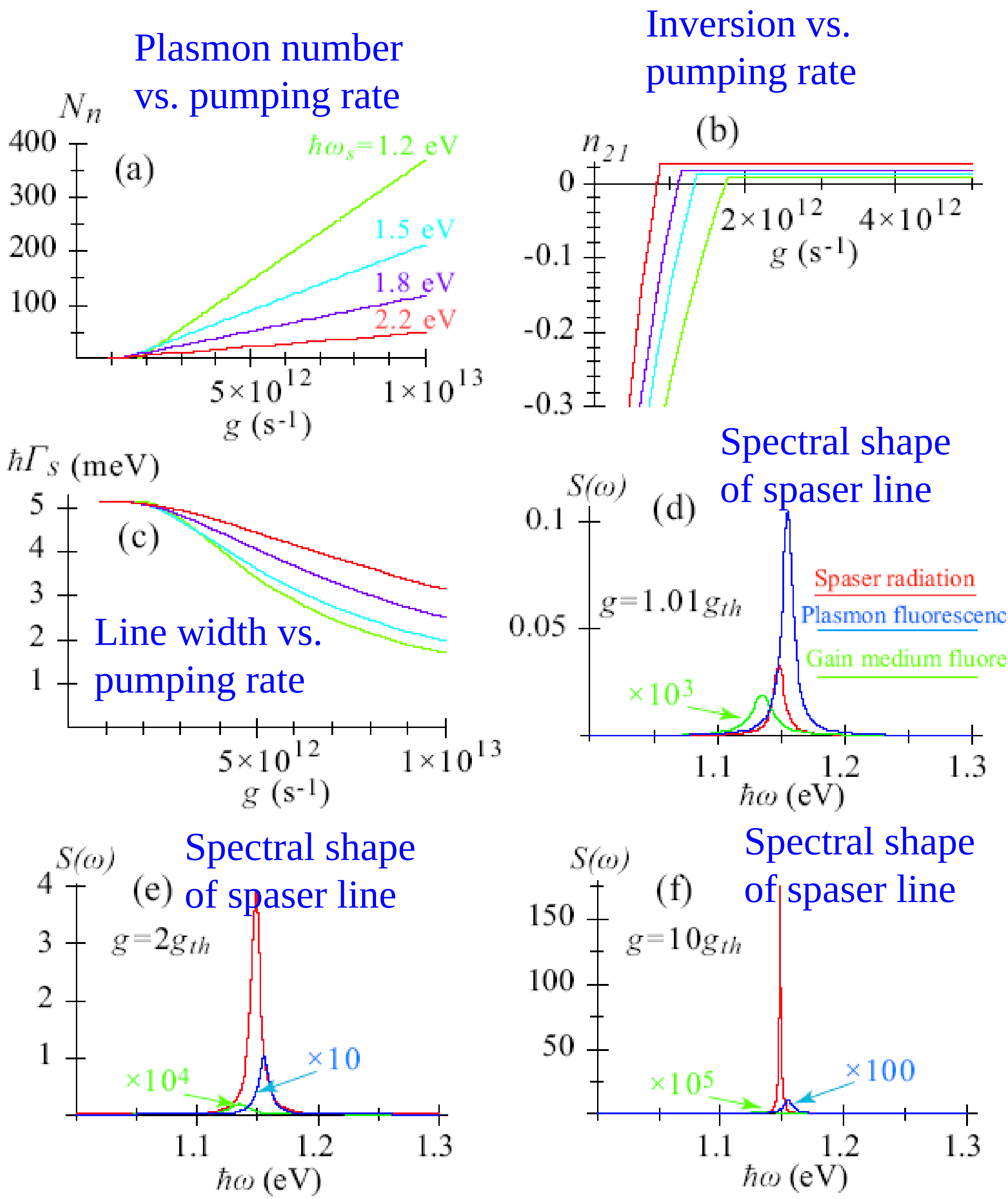
Stationary (CW) spaser regime

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

Mark I. Stockman, Journal of Optics, **12**, 024004-1-13 (2010).



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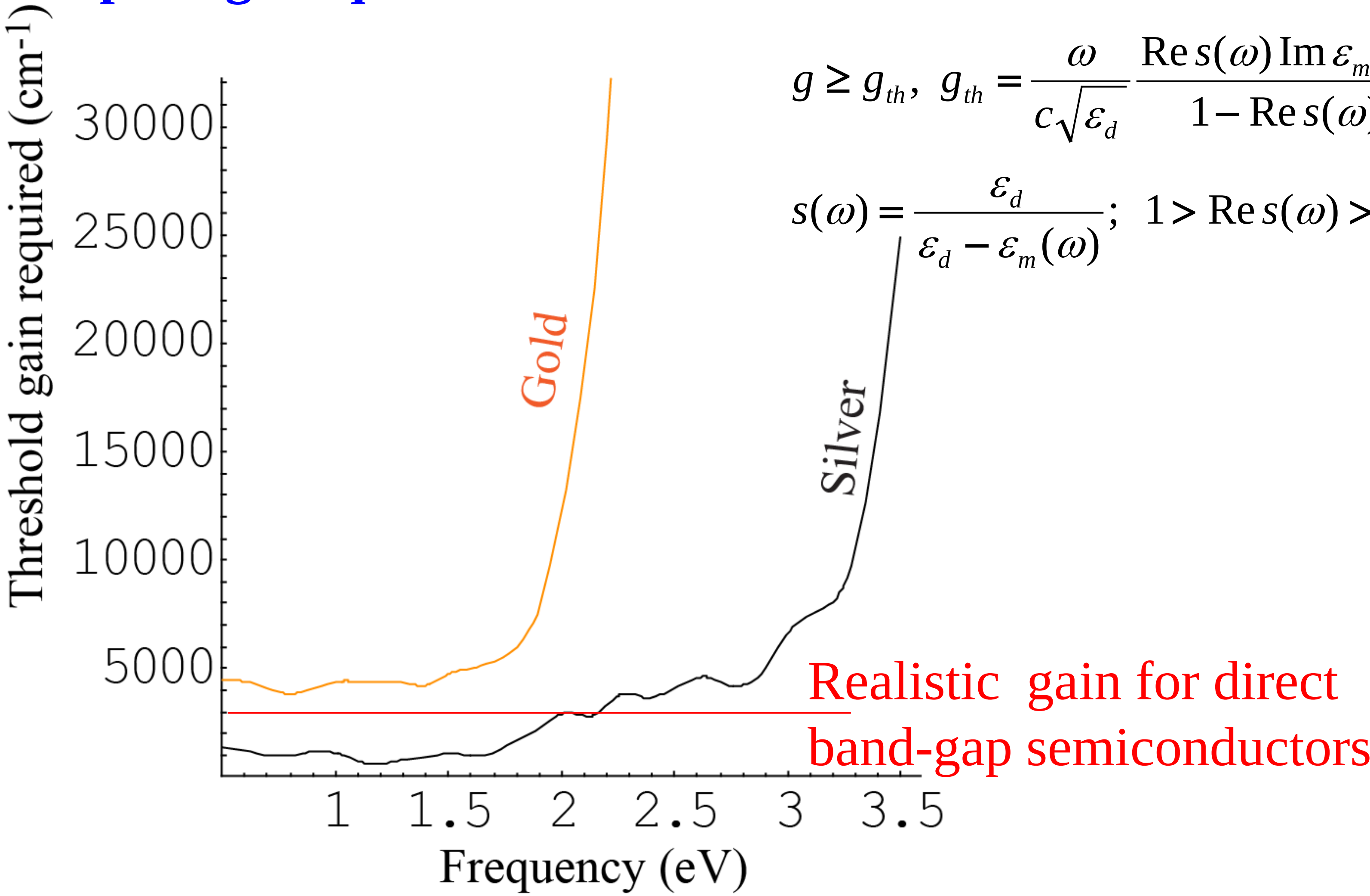
Spasing-Required Gain of Bulk Gain Medium

$$g \geq g_{th}, \quad g_{th} = \frac{\omega}{c\sqrt{\varepsilon_d}} \frac{\mathrm{Re}\, s(\omega)\, \mathrm{Im}\, \varepsilon_m(\omega)}{1 - \mathrm{Re}\, s(\omega)}$$

$$s(\omega) = \frac{\varepsilon_d}{\varepsilon_d - \varepsilon_m(\omega)}; \quad 1 > \mathrm{Re}\, s(\omega) > 0$$



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Scaling of Spaser

Field in spaser: $E \sim \frac{\hbar\omega}{R^{3/2}}\sqrt{N_p} \sim 1\frac{\text{MV}}{\text{cm}}\left(\frac{R}{10\text{ nm}}\right)^{-3/2}\sqrt{N_p}$

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: $\tau \sim \frac{1}{\gamma_p N_p} \sim \frac{100\text{ fs}}{N_p}$

Quantum limit: $\omega\tau \sim \frac{\omega}{\gamma_p}\frac{1}{N_p} \sim \frac{Q}{N_p} \sim \frac{100}{N_p} \ge 1$

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

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The spaser as a Nanoamplifier

Strategic goal is all-optical ultrafast (multi-THz) processors of signals and information, which are also radiation hardened

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 three ways:

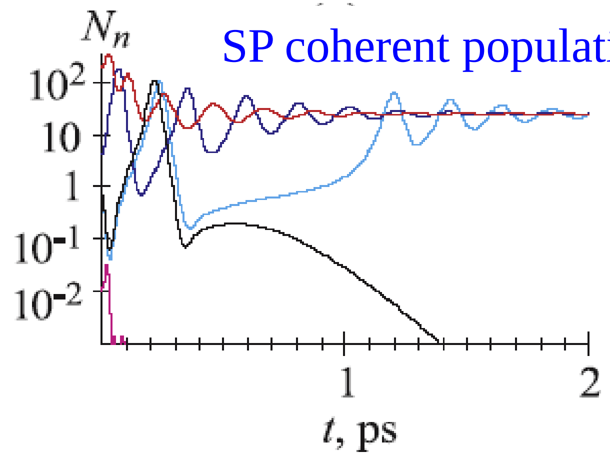
1. In transient mode (before reaching the CW regime), the spaser still possesses non-zero amplification
2. With a saturable absorber, the spaser can be bistable. There are two stable states: with the zero coherent SP population (“logical zero”) and with a high SP population that saturates the absorber (“logical one” state). Such a spaser will function as a threshold (digital) amplifier
3. Removing or reducing feedback, polaritonic spaser can function just like an optical amplifier but with a nanoscale size

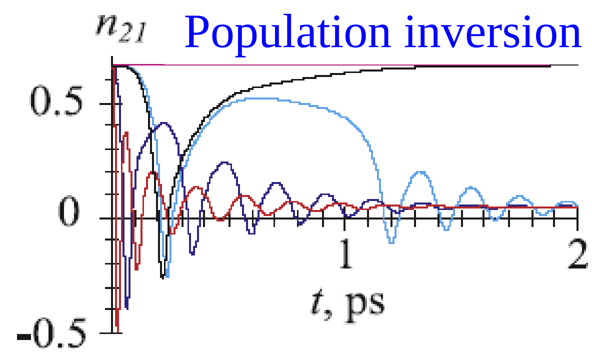
Bandwidth ~ 10-100 THz

Very high resistance to ionizing radiation, graceful degradation

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

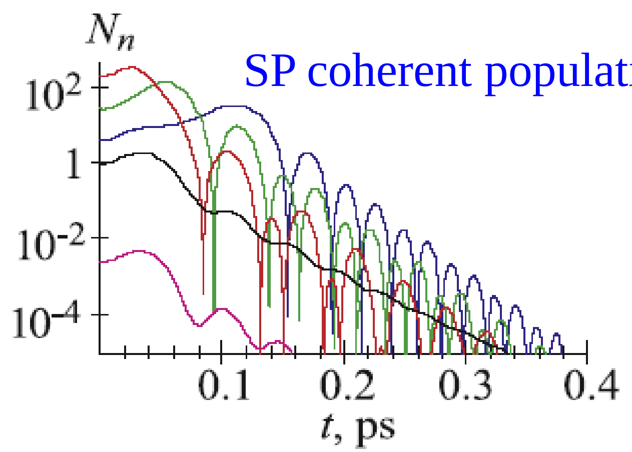
Stationary pumping

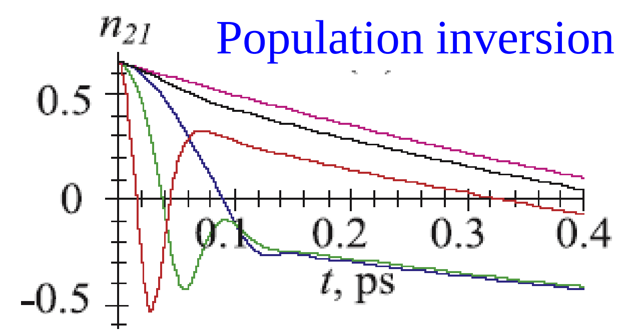




Mark I. Stockman, Journal of Optics, **12**, 024004-1-13 (2010).

Pulse pumping





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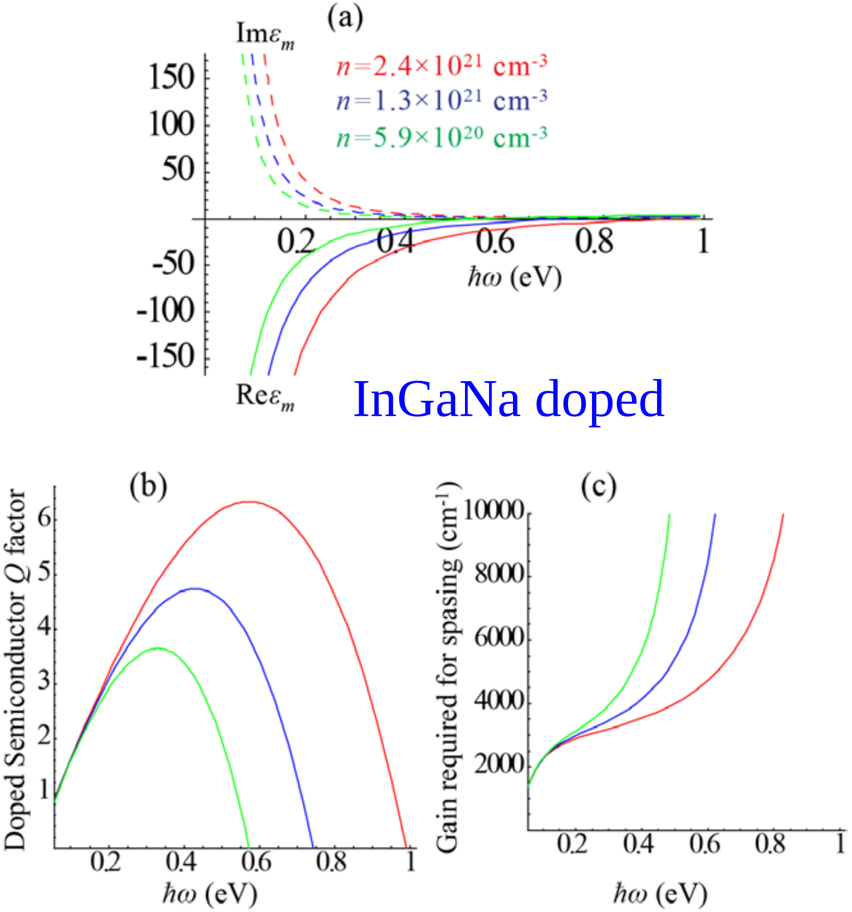
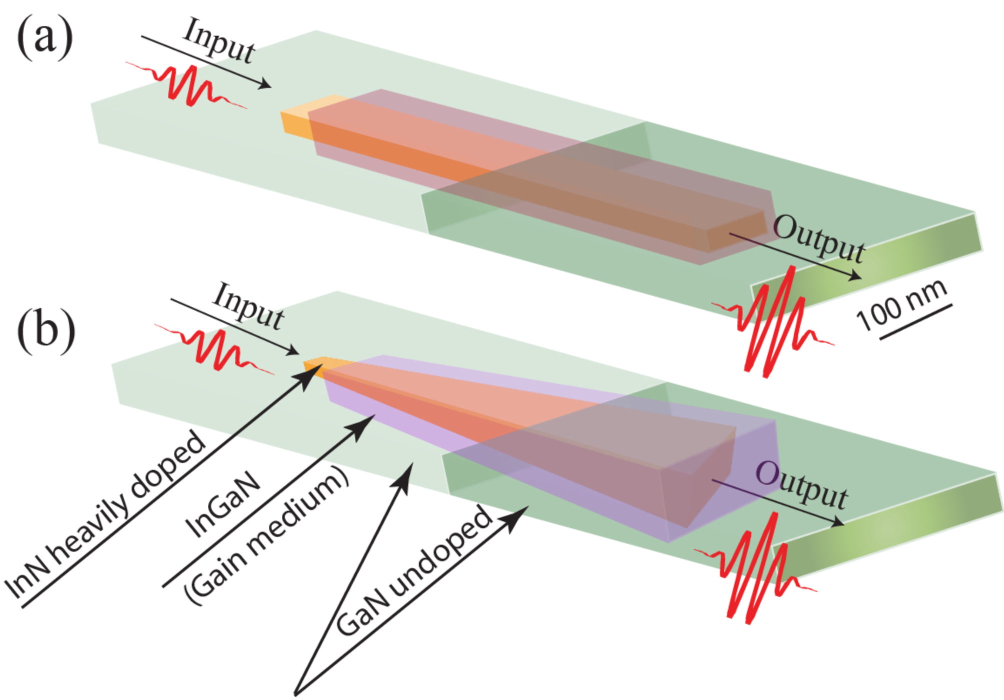
Spaser Nanoamplifier in Direct Bandgap Semiconductors





N. Dietz and M. Stockman, In preparation.

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LETTERS

Demonstration of a spaser-based nanolaser

M. A. Noginov[1], G. Zhu[1], A. M. Belgrave[1], R. Bakker[2], V. M. Shalaev[2], E. E. Narimanov[2], S. Stout[1,3], E. Herz[3], T. Suteewong[3] & U. Wiesner[3]

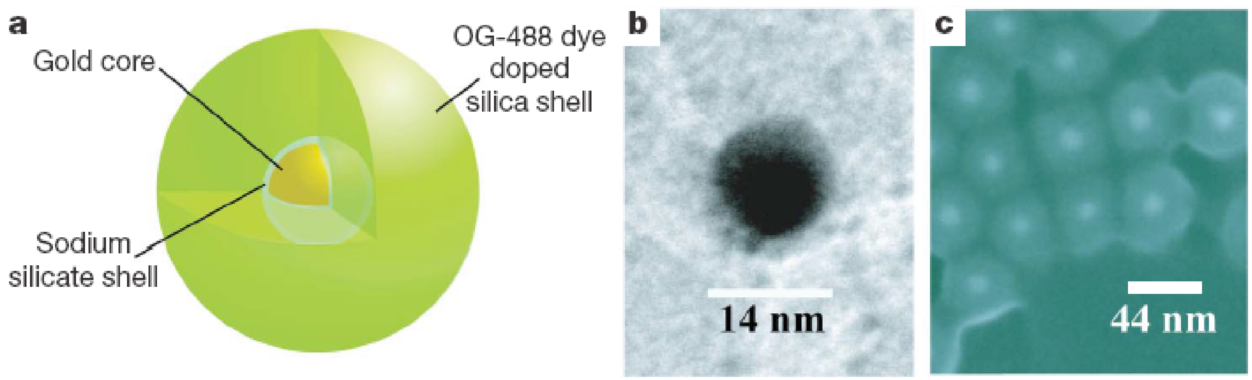

Figure 1 | Spaser design. a, Diagram of the hybrid nanoparticle architecture (not to scale), indicating dye molecules throughout the silica shell. **b**, Transmission electron microscope image of Au core. **c**, Scanning electron microscope image of Au/silica/dye core–shell nanoparticles. **d**, Spaser mode (in false colour), with $\lambda = 5$… circles represent the 14-nm … strength colour scheme is sh…

[1]Center for Materials Research, Norfolk State University, Norfolk, Virginia 23504, USA. [2]School of Electrical & Computer Eng… University, West Lafayette, Indiana 47907, USA. [3]Materials Science and Engineering Department, Cornell University, Ithaca

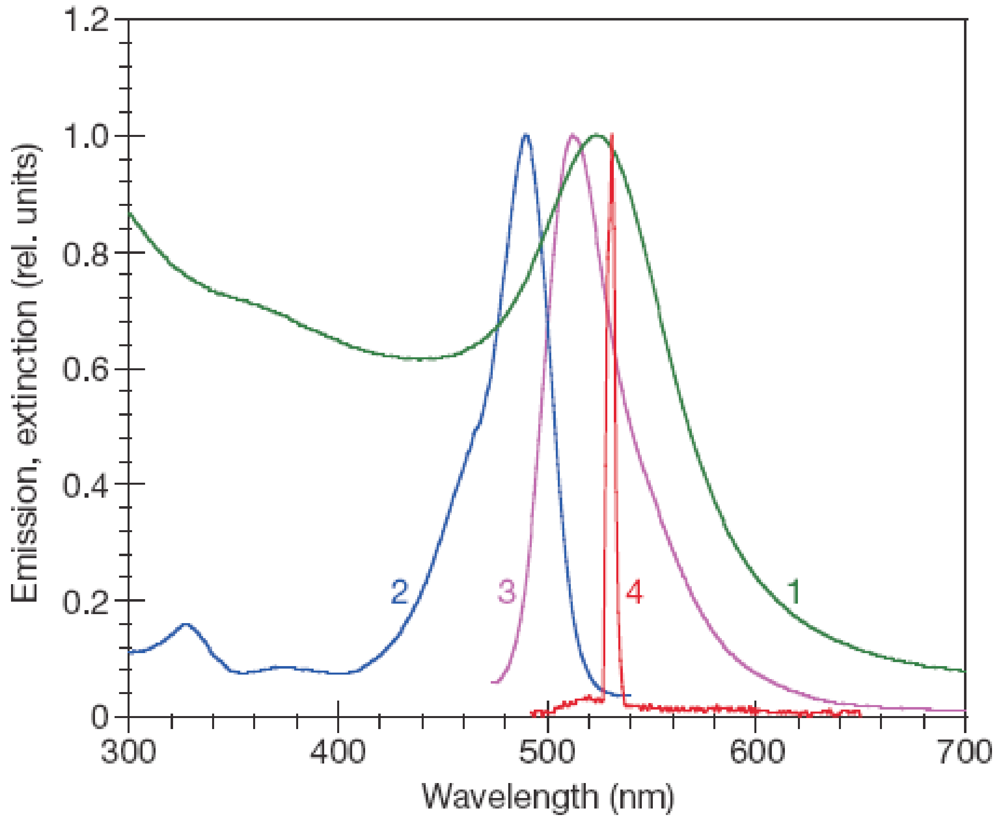

Figure 2 | Spectroscopic results. Normalized extinction (1), excitation (2), spontaneous emission (3), and stimulated emission (4) spectra of Au/silica/dye nanoparticles. The peak extinction cross-section of the nanoparticles is 1.1×10^{-12} cm^2. The emission and excitation spectra were measured in a spectrofluorometer at low fluence.

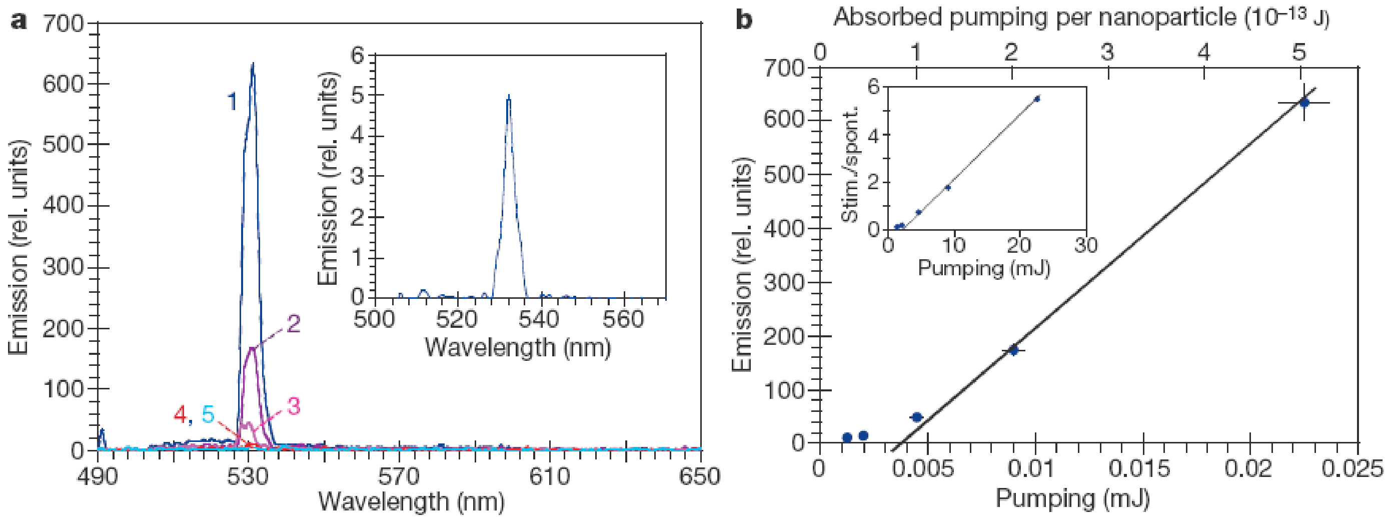

Figure 4 | Stimulated emission. **a**, Main panel, stimulated emission spectra of the nanoparticle sample pumped with 22.5 mJ (1), 9 mJ (2), 4.5 mJ (3), 2 mJ (4) and 1.25 mJ (5) 5-ns optical parametric oscillator pulses at $\lambda = 488$ nm. **b**, Main panel, corresponding input–output curve (lower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

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

Martin T. Hill[1*], Milan Marell[1], Eunice S. P. Leong[2], Barry Smalbrugge[1], Youcai Zhu[1], Minghua Sun[2], Peter J. van Veldhoven[1], Erik Jan Geluk[1], Fouad Karouta[1], Yok-Siang Oei[1], Richard Nötzel[1], Cun-Zheng Ning[2], and Meint K. Smit[1]

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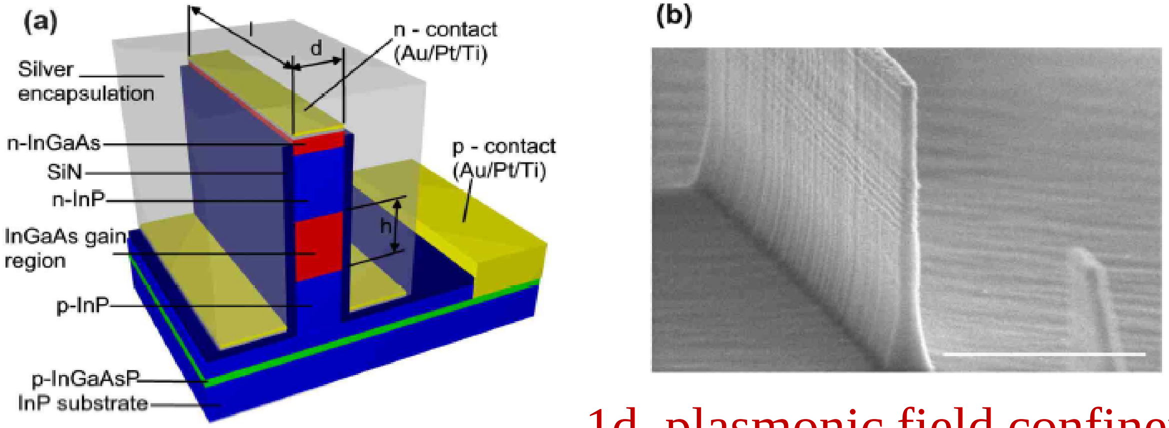

1d plasmonic field confinement

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.

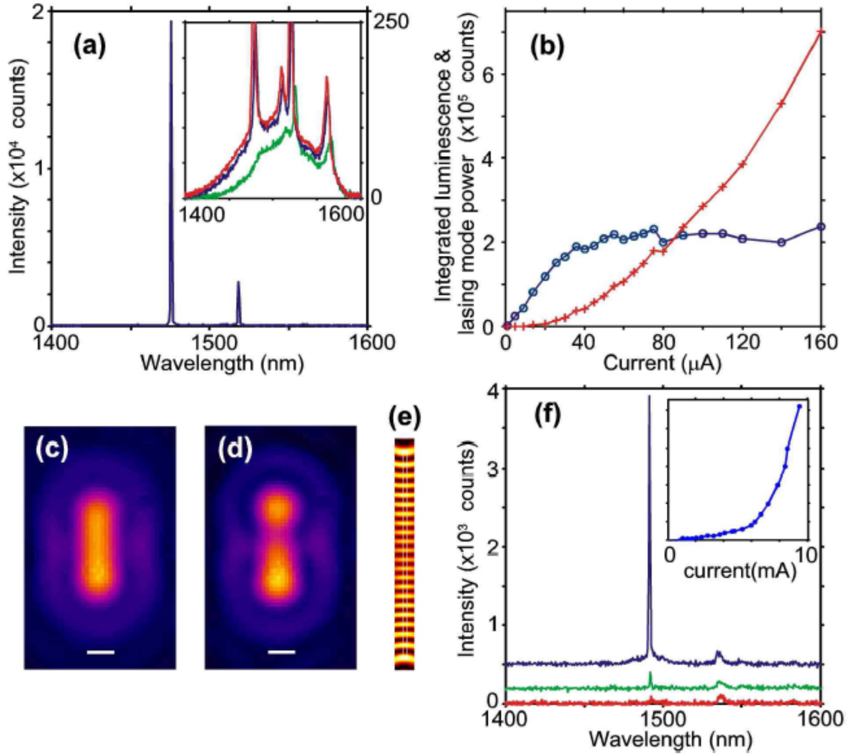

Fig. 2. Spectra and near field patterns showing lasing in devices. (a) Above threshold emission spectrum for 3 micron long device with semiconductor core width d~130nm (± 20nm), with pump current 180 µA at 78K. Inset: emission spectra for 20 (green), 40 (blue) and 60 (red) µA, all at 78K. (b) Lasing 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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Plasmon lasers at deep subwavelength scale

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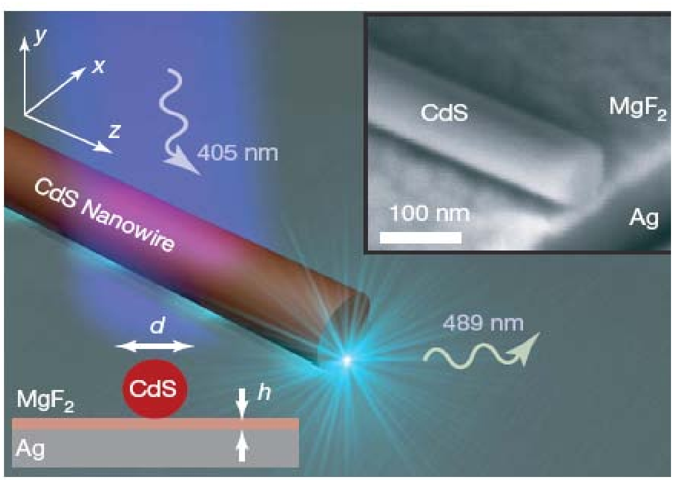
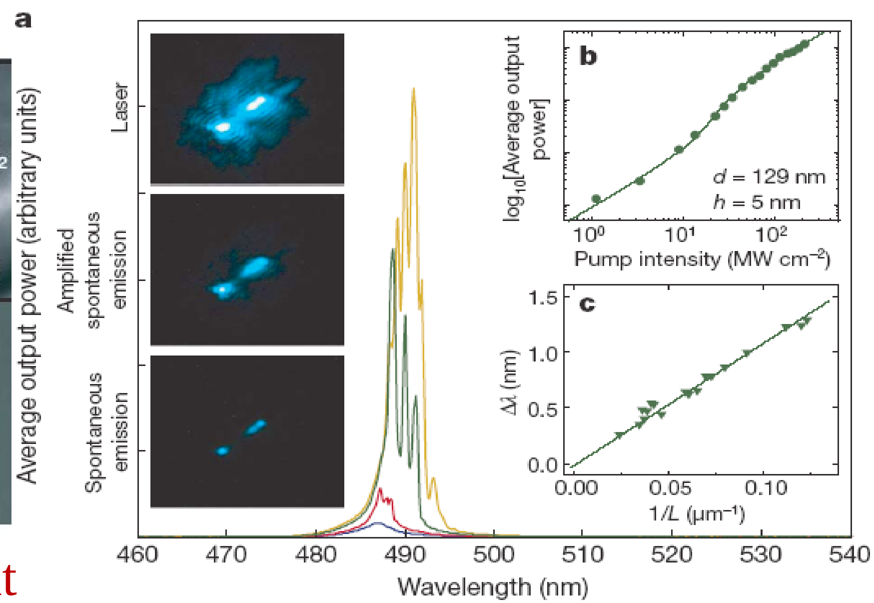
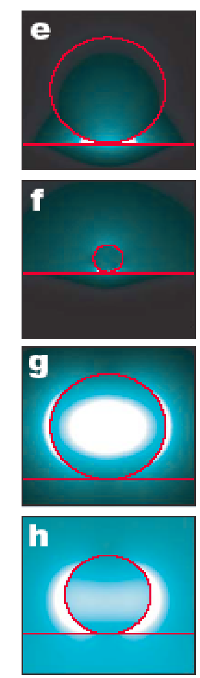






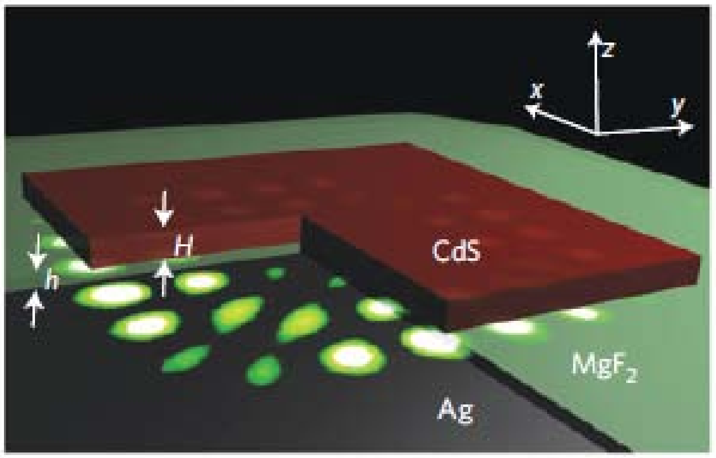
2d plasmonic field confinement

nature materials
LETTERS
PUBLISHED ONLINE: 19 DECEMBER 2010 | DOI: 10.1038/NMAT2919

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

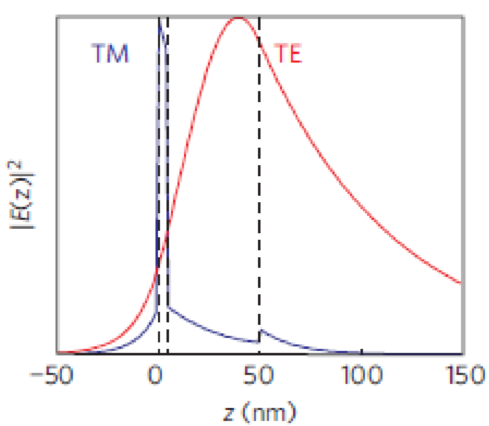
Ren-Min Ma[1†], Rupert F. Oulton[1†], Volker J. Sorger[1], Guy Bartal[1] and Xiang Zhang[1,2★]

a



1d +2d plasmonic field confinement

c



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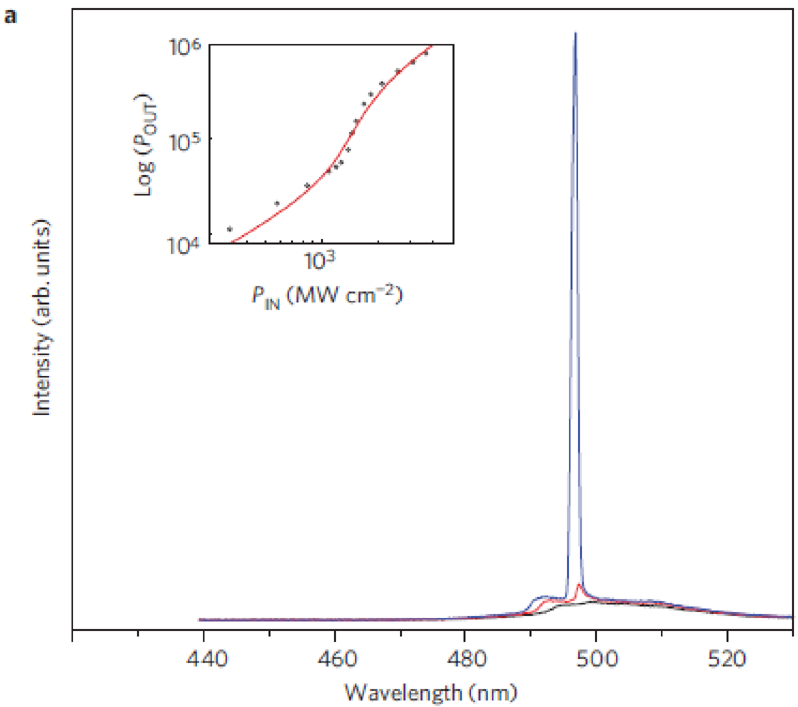

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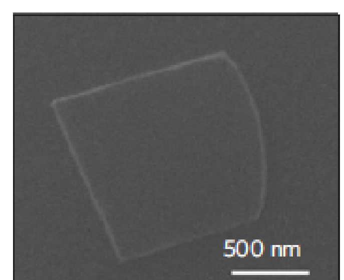

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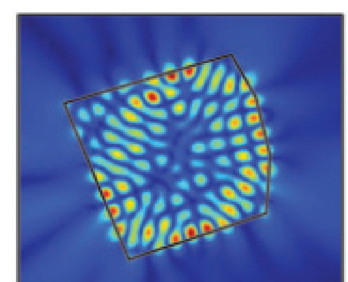

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BRIEF CONCLUSIONS

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. Spaser has been experimentally observed recently. This experiment is in an 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. Numerous plasmon-polariton spasers (plasmonic nanolasers) have been designed. In contrast to spaser, their length is on the order of micron (transverse mode size is nanometric).
5. The most promising applications of the spaser are an ultrafast nanoamplifier, local optical energy source, active nano-label, and an element of metamaterials with compensated loss.

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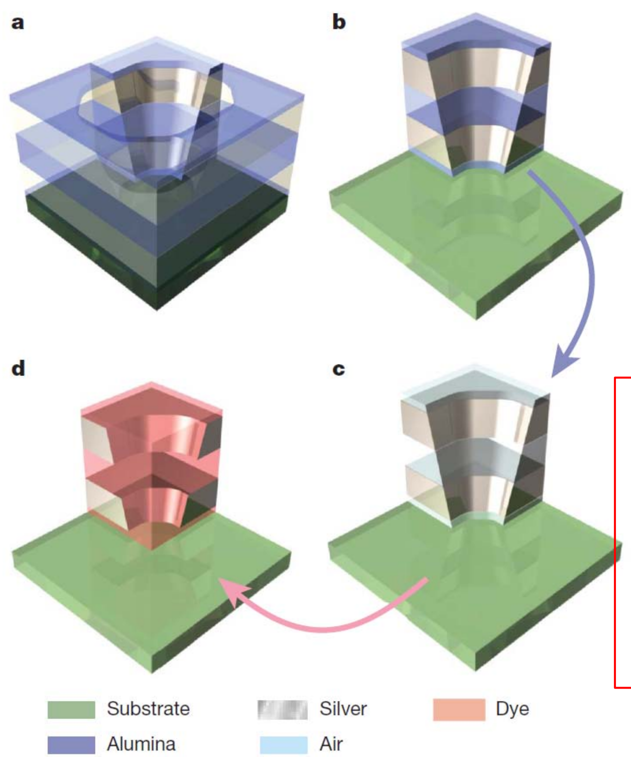
Appendix

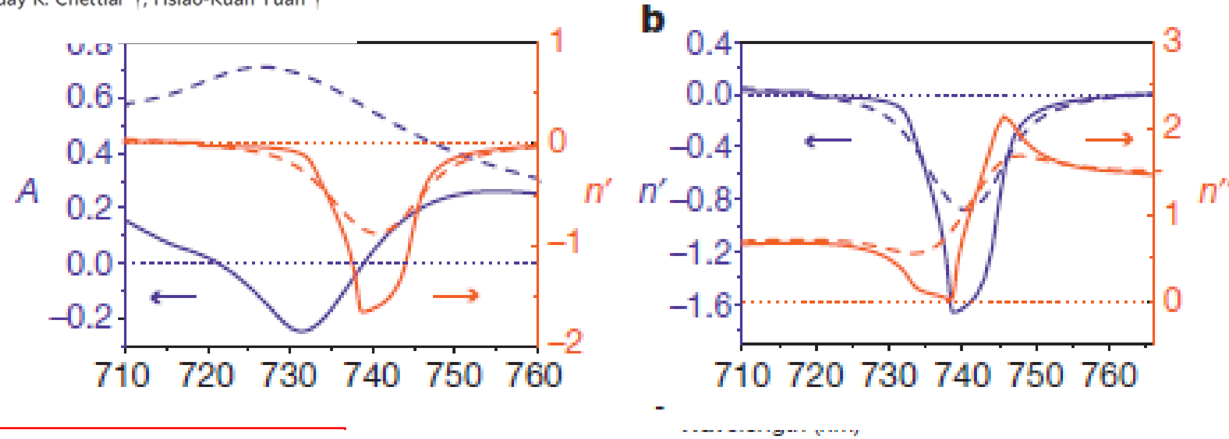
LETTERS

Loss-free and active optical negative-index metamaterials

Shumin Xiao[1], Vladimir P. Drachev[1], Alexander V. Kildishev[1], Xingjie Ni[1], Uday K. Chettiar[1]†, Hsiao-Kuan Yuan[1]† & Vladimir M. Shalaev[1]

active optical NIM. The original loss-limited negative refractive index and the figure of merit (FOM) of the device have been drastically improved with loss compensation in the visible wavelength range between 722 and 738 nm. In this range, the NIM becomes active such that the sum of the light intensities in transmission and reflection exceeds the intensity of the incident beam. At a wavelength of 737 nm, the negative refractive index improves from −0.66 to −1.017 and the FOM increases from 1 to 26. At 738 nm, the FOM is expected to become macroscopically large, of the order of 10^6. This study demonstrates the possibility of fabricating an optical negative-index metamaterial that is not limited by the inherent loss in its metal constituent.





This device is more like a spaser than an effective low-loss medium

Figure 4 | Simulation and determined parameters. **a**, The simulated refractive index, n' (real part), and absorptance, A (in the forward direction), as functions of wavelength with (solid) and without (dashed) gain. **b**, The effective refractive index, $n = n' + in''$, determined with (solid) and without (dashed) gain. **c**, The effective FOM determined with (solid) and without

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PRL **106,** 156802 (2011) PHYSICAL REVIEW LETTERS week ending 15 APRIL 2011

Spaser Action, Loss Compensation, and Stability in Plasmonic Systems with Gain

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(Received 16 November 2010; revised manuscript received 28 February 2011; published 11 April 2011)

We demonstrate that the conditions of spaser generation and the full loss compensation in a dense resonant plasmonic-gain medium (metamaterial) are identical. Consequently, attempting the full compensation or overcompensation of losses by gain will lead to instability and a transition to a spaser state. This will limit (clamp) the inversion and lead to the limitation on the maximum loss compensation achievable. The criterion of the loss overcompensation, leading to the instability and spasing, is given in an analytical and universal (independent from system's geometry) form.

DOI: 10.1103/PhysRevLett.106.156802 PACS numbers: 73.20.Mf, 42.50.Nn, 78.67.Pt, 81.05.Xj

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Consider an isotropic metamaterial that can be described by complex permittivity and permeability. A known homogenization procedure leads to an exact result for the (effective) permittivity of the composite

$$\bar{\varepsilon}(\omega) = \frac{1}{V\left|E(\omega)\right|^2}\int_V \varepsilon(\mathbf{r},\omega)\left|\mathbf{e}(\mathbf{r},\omega)\right|^2 dV$$

Hear *E* is the macroscopic field and **e**(**r**) is the (mesoscopic) local field inside the metamaterial. This local field is expressed as an eigenmode expansion

$$\mathbf{e}(\mathbf{r},\omega) = \mathbf{E}(\omega) - \sum_n \frac{a_n}{s(\omega) - s_n}\mathbf{E}_n(\mathbf{r})\,, \quad s(\omega) \equiv \frac{\varepsilon_h(\omega)}{\varepsilon_h(\omega) - \varepsilon_m(\omega)}$$

where $\mathbf{E}_n(\mathbf{r})$ is the eigenmode field. Assume that: there is a resonance with an *n*-th eigenmode, the metal has a high quality factor, *Q*>>1, and the metal's fill factor *f* is not too small, so *Qf*>>1. Then the local field is

$$\mathbf{e}(\mathbf{r},\omega) = i\frac{a_n}{\operatorname{Im} s(\omega_n)}\mathbf{E}_n(\mathbf{r})$$

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Then the effective permittivity becomes (where b_n >0 is a coefficient):

$$\bar{\varepsilon}(\omega) = b_n \left[s_n \varepsilon_m(\omega) + (1 - s_n)\varepsilon_h(\omega) \right] \qquad b_n = \frac{1}{V}\left(\frac{Q\int_V \theta(\mathbf{r})\mathbf{E}_n(\mathbf{r})dV}{s_n(1-s_n)}\right)^2$$

In the case of the full inversion (maximum gain) and in exact resonance, the imaginary part of the host-medium permittivity describes stimulated emission as given by the standard expression

$$\varepsilon_h(\omega) = \varepsilon_d - i\frac{4\pi}{3}\frac{|\mathbf{d}_{12}|^2 n_c}{\hbar\Gamma_{12}}$$

where $\varepsilon_d = \mathrm{Re}\,\varepsilon_h$, $\mathbf{d}_{12}$ is the dipole matrix element of the gain transition in a chromophore center of the gain medium, Γ_{12} is a spectral width of this transition, and n_c is the concentration of these centers.

The condition for the full electric loss (over)compensation at the resonant frequency $\omega = \omega_n$ is $\mathrm{Im}\,\bar{\varepsilon}(\omega) \leq 0$, which reduces to

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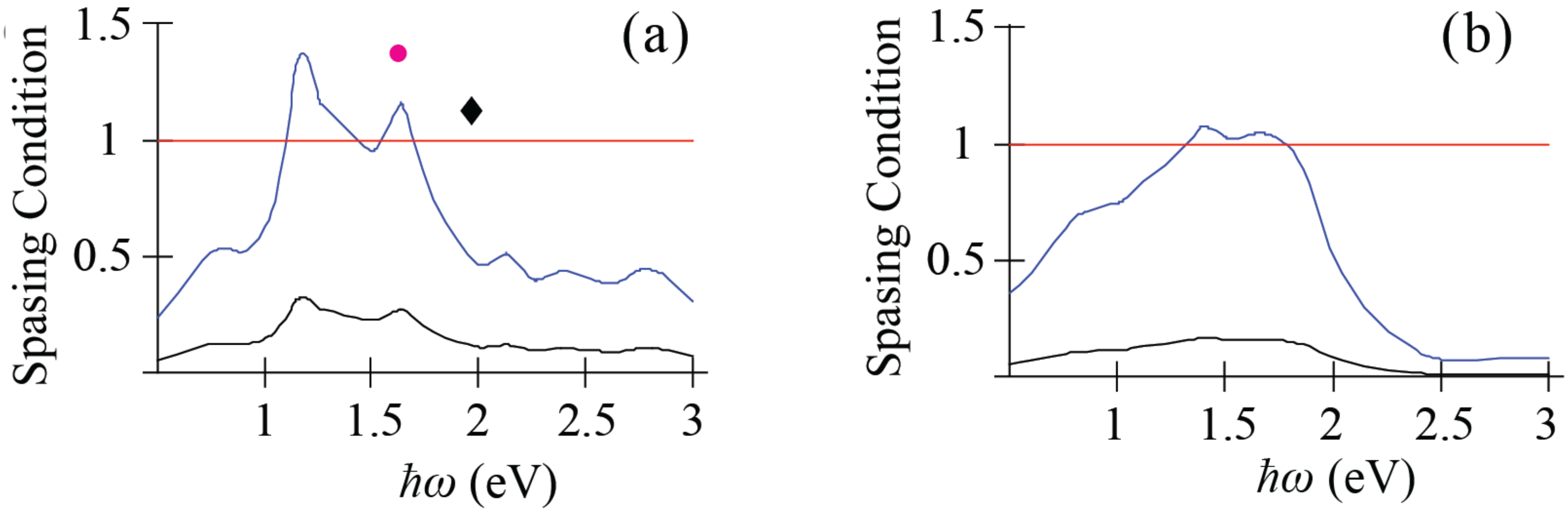
$$\frac{4\pi}{3}\frac{|\mathbf{d}_{12}|^2 n_c[1-\operatorname{Re}s(\omega)]}{\hbar\Gamma_{12}\operatorname{Re}s(\omega)\operatorname{Im}\varepsilon_m(\omega)}\geq 1 \text{ or } g \geq g_{th},\; g_{th}=\frac{\omega}{c\sqrt{\varepsilon_d}}\frac{\operatorname{Re}s(\omega)\operatorname{Im}\varepsilon_m(\omega)}{1-\operatorname{Re}s(\omega)},$$

where g is the required gain

- This is a criterion for both the loss compensation and spasing, the latter obtained previously in: M. I. Stockman, *The Spaser as a Nanoscale Quantum Generator and Ultrafast Amplifier*, Journal of Optics **12,** 024004-1-13 (2010)
- This criterion is analytical and exact, provided that the metamaterials is resonant and dense, and that its eigenmodes are non-uniform in space -- hot spots or reflection from facets -- create a feedback
- Thus, an attempt at a full compensation of losses will cause spasing instead, which will saturate the gain transition, eliminate the net gain, clamp the inversion, and make the complete loss compensation impossible
- This criterion does not depend on the geometry of the system or any specific hot spots of local fields, predicated on the gain medium filling all the space left by the metal



Spasing criterion as a function of optical frequency . The straight line (red on line) represents the threshold for the spasing and full loss compensation, which take place for the curve segments above it. (a) Computations for silver. The chromophore concentration is n_c = 6 x10^{18} cm^{-3} for the lower curve (black) and n_c = 3x10^{19} cm^{-3} for the upper curve (blue). The magenta solid circle and black diamond show the values of the spasing criterion for the conditions of Refs. 2 and 3, respectively. (b) Computations for gold. The chromophore concentration is n_c = 3x10^{19} cm^{-3} for the lower curve (black) and n_c =2x10^{20} cm^{-3} for the upper curve (blue).

1. S. Xiao, V. P. Drachev, A. V. Kildishev, X. Ni, U. K. Chettiar, H.-K. Yuan, and V. M. Shalaev, *Loss-Free and Active Optical Negative-Index Metamaterials*, Nature **466**, 735-738 (2010).

2. M. A. Noginov, G. Zhu, M. Mayy, B. A. Ritzo, N. Noginova, and V. A. Podolskiy, *Stimulated Emission of Surface Plasmon Polaritons*, Phys. Rev. Lett. **101**, 226806-1-4 (2008).

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BRIEF CONCLUSIONS

- The same criterion is obtained for both the loss compensation and spasing
- This criterion is analytical and exact, provided that the metamaterials is resonant and dense, and that its eigenmodes are non-uniform in space (contain "hot spots"), which creates an inherent feedback
- Thus, an attempt at a full compensation of losses will cause spasing instead, which will saturate the gain transition, eliminate the net gain, clamp the inversion, and make the complete loss compensation impossible
- This criterion does not depend on the geometry of the system or any specific hot spots of local fields, predicated on the gain medium filling all the space left by the metal


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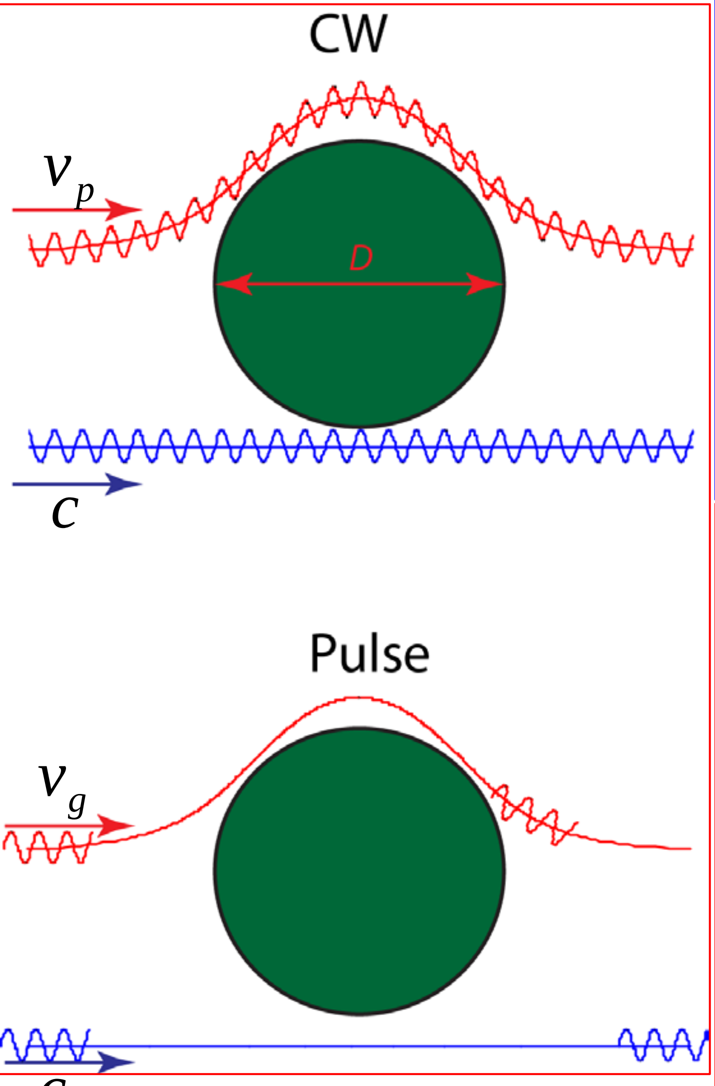
Breaking the Cloak: Relativistic Causality



For CW radiation, the ray that bends around the cloak carries radiation with higher than *c* phase velocity, which is possible

$$v_p = (\pi - 1)c > 2c$$

For pulse radiation, the ray that bends around the cloak carries radiation with *group* velocity than *must* be less than *c* *(relativistic causality)*. Thus, it arrives with a delay,

$$\Delta t = \left(\frac{\pi}{v_g} - \frac{1}{c}\right)D > \frac{(\pi - 1)D}{c} = (\pi - 1)\frac{D}{\lambda}T >> T$$

which for a *macroscopic* cloak is much larger than the period *T* (typically, $> 10^6 T$)