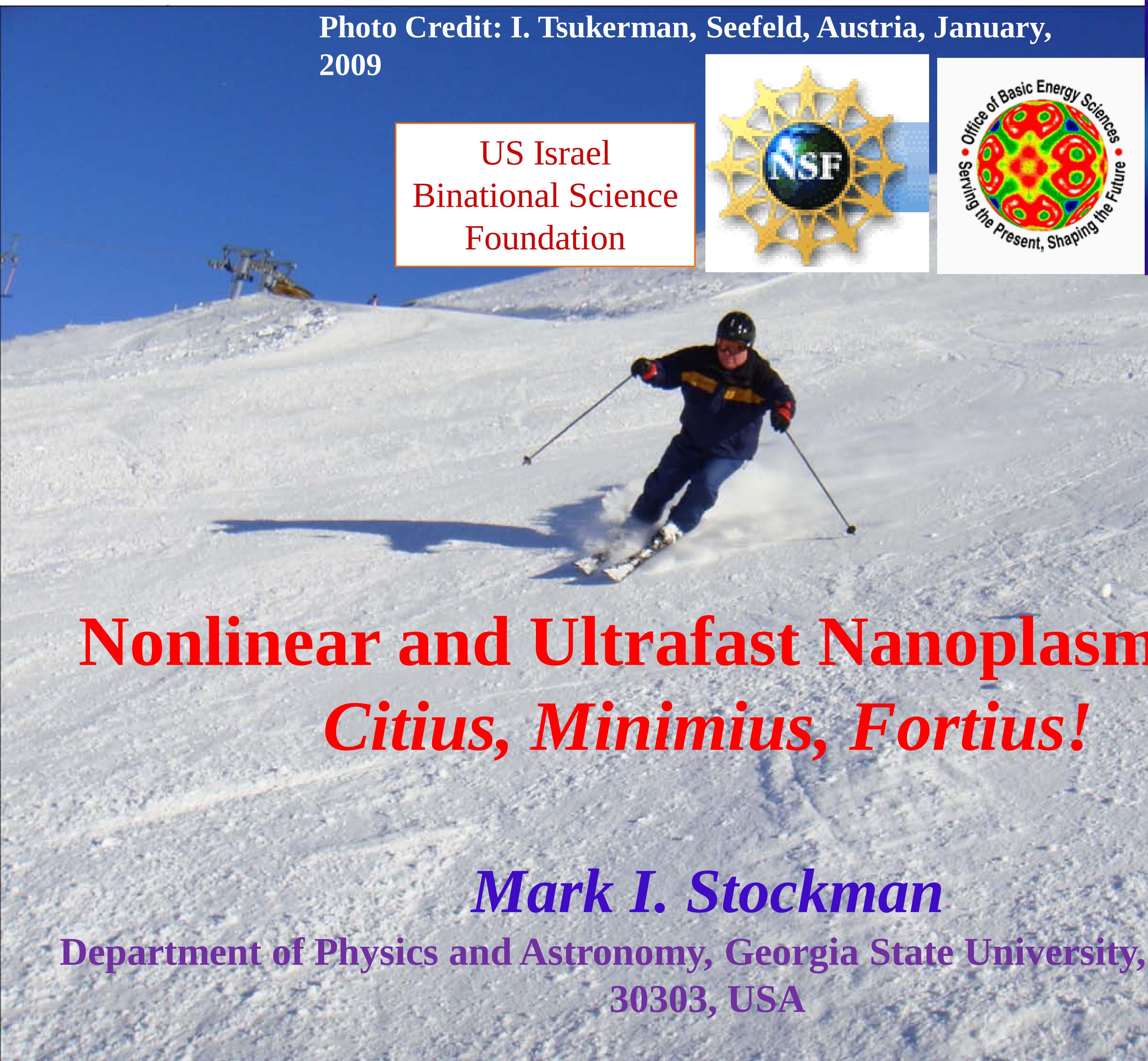
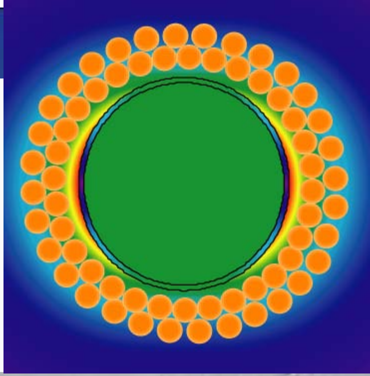


Photo Credit: I. Tsukerman, Seefeld, Austria, January, 2009

US Israel
Binational Science
Foundation

NSF

Office of Basic Energy Sciences • Serving the Present, Shaping the Future •



Nonlinear and Ultrafast Nanoplasmonics: *Citius, Minimius, Fortius!*

Mark I. Stockman

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

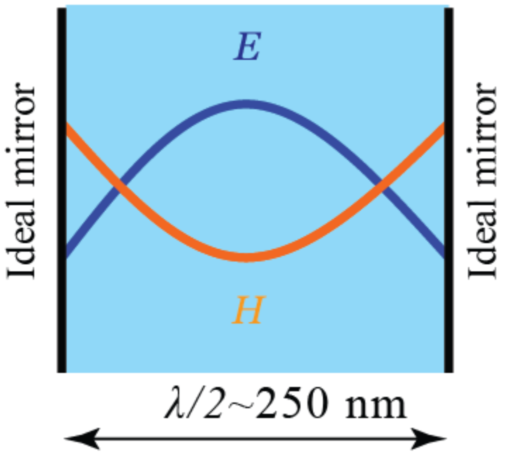
Collaborators:

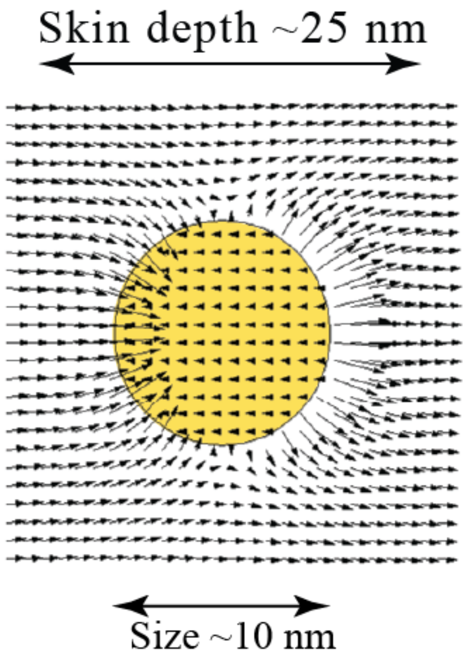
1. David J. Bergman, Department of Physics, Tel Aviv University, Israel
2. Sophie Brasselet, Institut Fresnel, Marseilles, France
3. Paul Corkum, Femtosecond Science Program, National Research Council of Canada
4. Maxim Durach, Georgia State University, Atlanta, GA 30340, USA
5. Sergey V. Faleev, Sandia National Laboratories, Livermore, CA, USA
6. Harald Giessen, University of Stuttgart, Germany
7. Dmitry Gramotnev, Queensland University of Technology, Brisbane, Qld 4001, Australia
8. Misha Ivanov, Imperial College, UK
9. Ulf Kleineberg, Ludwig Maximilian University, Munich, Germany
10. Victor Klimov, Los Alamos National Laboratory, Los Alamos, New Mexico, USA
11. Matthias Kling, Max Plank Institute for Quantum Optics, Garching, Germany
12. Katrin Kneipp, Technical University Copenhagen, Denmark
13. Takayoshi Kobayashi, University of Tokyo, Japan
14. Ferenc Krausz, Max Plank Institute for Quantum Optics, Garching, Germany
15. Ivan Larkin, Georgia State University, Atlanta, GA 30340, USA
16. Kuiru Li, Georgia State University, Atlanta, GA 30340, USA
17. Keith Nelson, MIT, Boston, USA
18. Peter Nordlander, Rice University, Houston, Texas, USA
19. Hrvoje Petek, University of Pittsburgh, USA
20. Anastasia Rusina, Georgia State University, Atlanta, GA 30340, USA
21. Igor Tsukerman, University of Akron, OH 44325, USA
22. Nikolay Zheludev, University of Southampton, UK
23. Joseph Zyss, Ecole Normale Supérieure de Cachan, 94235 Cachan, France

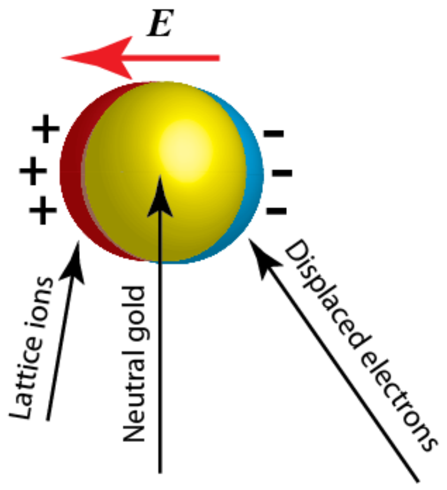
CONTENTS

Nanoplasmonics in a nano-nutshell

Concentration of optical energy on the nanoscale



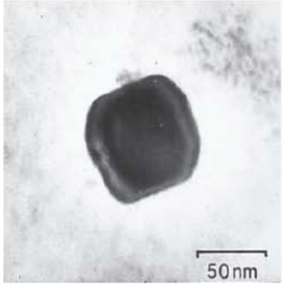




Extrinsic (outside the metal) enhancement of local fields is by a plasmonic quality factor $Q = \frac{-\mathrm{Re}\,\varepsilon_m}{\mathrm{Im}\,\varepsilon_m} \sim 10-100$

Lycurgus Cup (4th Century AD): Roman Nanotechnology

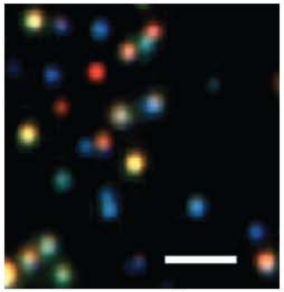
© Trustees of British Museum

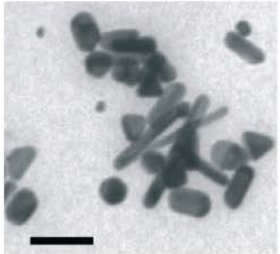


I. Freestone, N. Meeks, M. Sax, and C. Higgitt, *The Lycurgus Cup - a Roman Nanotechnology*, Gold Bull. **40**, 270-277 (2007

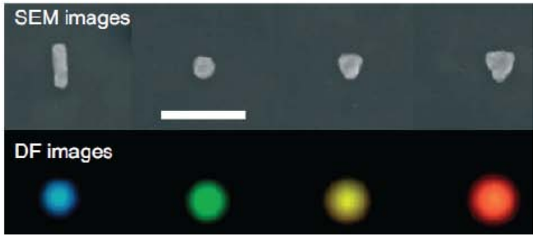
Nanoplasmonic colors are very bright. Scattering and absorption of light by them are very strong. This is due to the fact that all of the millions of electrons move in unison in plasmonic oscillations Nanoplasmonic colors are also eternal: metal nanoparticles are stable in glass: they do not bleach and do not blink. Gold is stable under biological conditions and is not toxic *in vivo*

Colors of Silver Nanocrystals and Gold Nanoshapes





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]

CONTENTS

- Introduction: Plasmonics and Nanoconcentration of Optical Energy
- Plasmonic Enhancement, Ultrafast Nature of Plasmonics, and Plasmonic Nonlinearities
- Perturbative Nonlinearities in Nanoplasmonic Systems
- Nonperturbative (Strong Field) Nonlinearities
- Spaser as an Ultrafast Quantum Generator and Nanoamplifier
- Spasing vs. Loss Compensation as Extrinsic Strong Nonlinearity in Plasmonics
- Conclusions

Classification of Nonlinearities in Nanoplasmonics

1. Extrinsic, Intrinsic, and Combined

In the surrounding dielectric, in the metal itself, and in both.

Intrinsic (Perturbative): Due to nonlinear polarizabilities of the metal itself.

A. Bouhelier et al., Phys. Rev. Lett. **90**, 13903 (2003).
J. Renger et al., Phys. Rev. Lett. **103**, 266802 (2009).

Extrinsic (Perturbative) : Due to nonlinear polarizabilities of the surrounding medium

D. Pacifici, H. J. Lezec, and H. A. Atwater, Nat. Phot. **1**, 402 (2007)

2. Perturbative (weak field) and Nonperturbative (strong field)

Perturbative (due to low-order nonlinear polarizabilities), both extrinsic and intrinsic: SHG, THG, parametric mixing, Kerr effect, nonlinear absorption, two-photon fluorescence

Nonperturbative (extrinsic): High-harmonic generation, spaser, loss compensation by gain

S. Kim et al., Nature **453**, 757 (2008)
M. I. Stockman, Journal of Optics **12**, 024004 (2010)
M. I. Stockman, Phys. Rev. Lett. **106**, 156802 (2011)
P. M. Bolger et al., Opt. Lett. **35**, 1197 (2010)

Nonperturbative (intrinsic): Plasmon-polariton solitons, metallization of dielectrics

E. Feigenbaum, and M. Orenstein, Opt. Lett. **32**, 674 (2007)
M. Durach et al., Phys. Rev. Lett. **105**, 086803 (2010)
M. Durach et al., arXiv:1104.1642 (2011)

Extrinsic Enhancement Factors for Small (in comparison to skin depth ~25 nm) Nanoparticles

These factors do not depend on the nanoparticle size *R* (scaling)

Plasmonic quality factor: $Q = \dfrac{-\mathrm{Re}\,\varepsilon_m}{\mathrm{Im}\,\varepsilon_m} \sim 10-100$

Radiative rate enhancement factor: $\sim Q^2 \sim 10^2 - 10^4$

Excitation rate enhancement factor: $\sim Q^2 \sim 10^2 - 10^4$

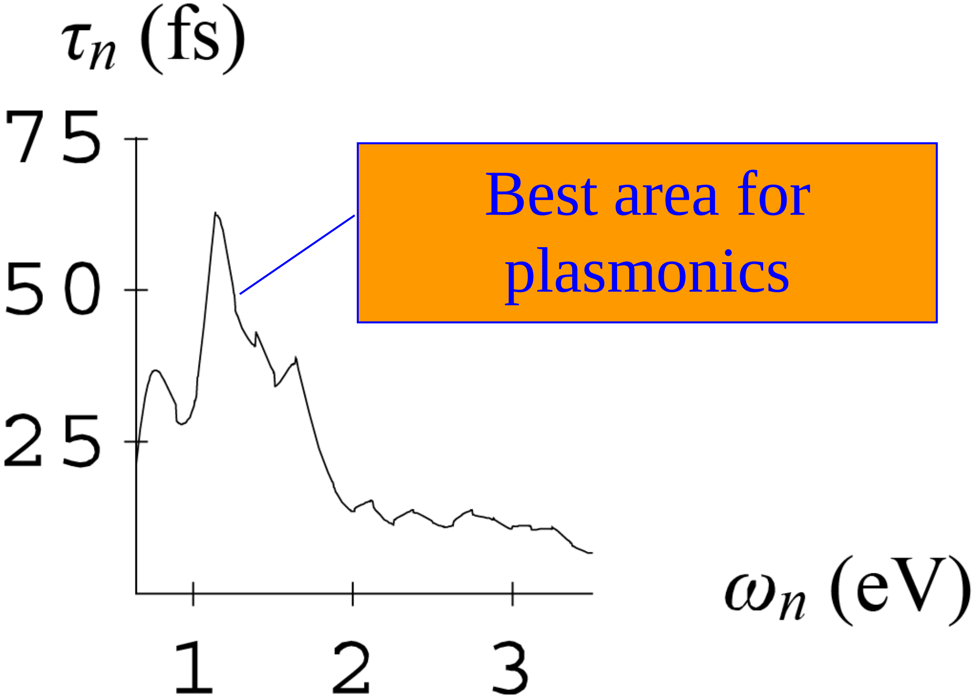
This Purcell factor is inversely proportional to the nanoparticle volume, which is fundamentally important for spaser action (feedback rate)

SERS resonant enhancement factor: $\sim Q^4 \sim 10^4 - 10^8$ (additional enhancement is due to the geometric enhancement (in tight gaps, at sharp tips, etc.)

Enhancement of the emission rate into an SP mode: $\propto \dfrac{Q}{R^3}$

Same with respect to free photons (Purcell factor): $\sim \dfrac{\bar{\lambda}^3 Q}{R^3} \sim 10^8$

Nanoplasmonics is intrinsically ultrafast:



Best area for plasmonics

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

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

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

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

Corresponding rise time of plasmonic responses ~ 100 as

M. I. Stockman, Phys. Rev. Lett. **84,** 1011 (2000).
M. I. Stockman, S. V. Faleev, and D. J. Bergman, Phys. Rev. Lett. **88**, 67402 (2002)
D. J. Bergman, and M. I. Stockman, Phys. Rev. Lett. **90**, 027402 (2003)

CONTENTS

- Introduction: Plasmonics and Nanoconcentration of Optical Energy
- Plasmonic Enhancement, Ultrafast Nature of Plasmonics, and Plasmonic Nonlinearities
- Perturbative Nonlinearities in Nanoplasmonic Systems
- Nonperturbative (Strong Field) Nonlinearities
- Spaser as an Ultrafast Quantum Generator and Nanoamplifier
- Spasing vs. Loss Compensation as Extrinsic Strong Nonlinearity in Plasmonics
- Conclusions

Perturbative (two-photon) ultrafast intrinsic nonlinearity

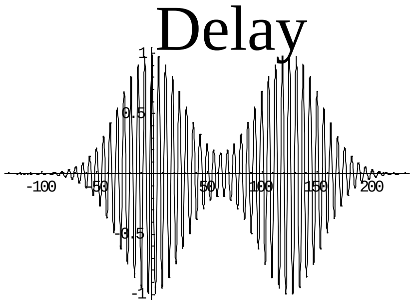
Localized SP hot spots and SPPs coexist in space and time on nanostructured surfaces

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).

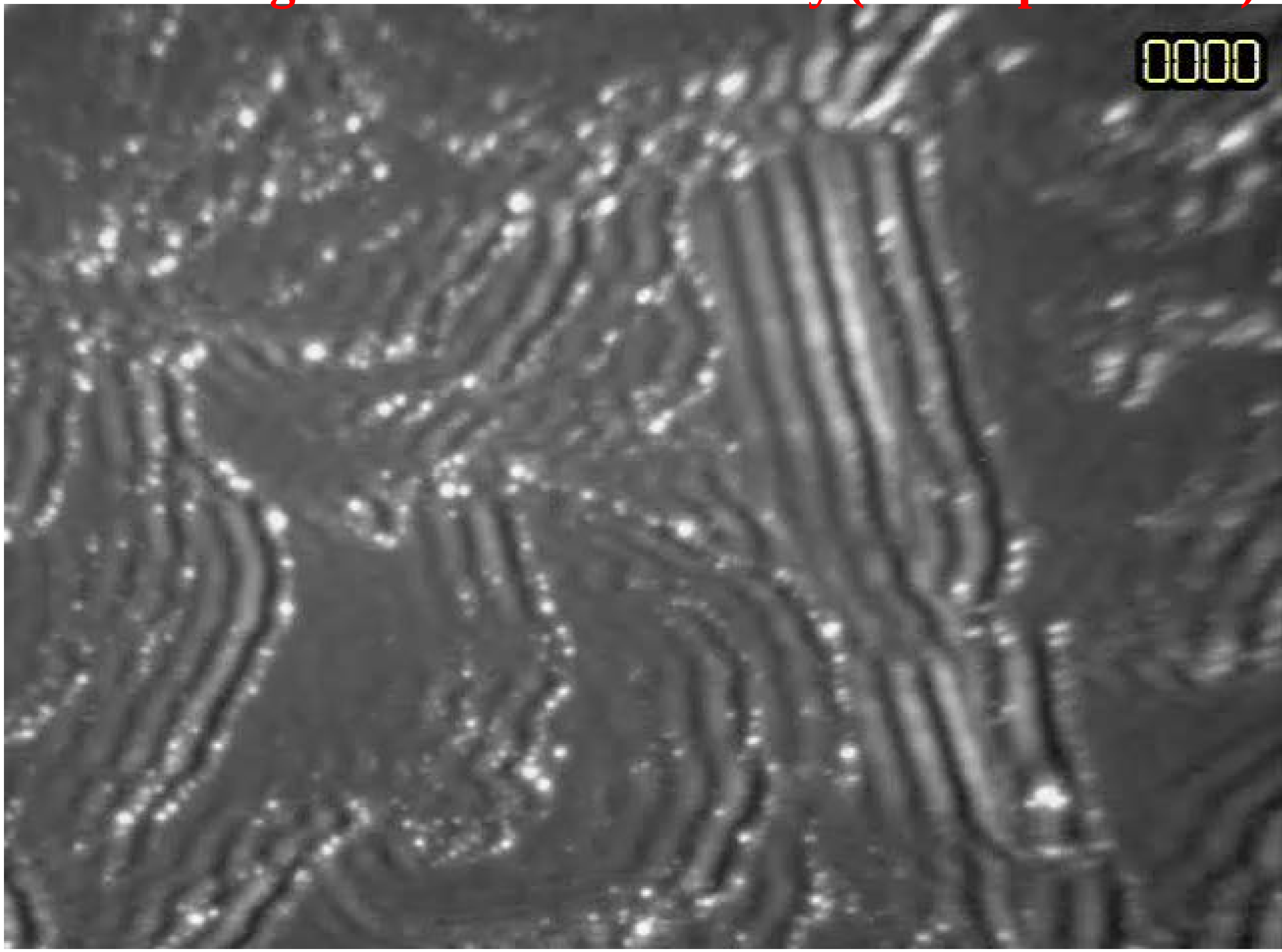
200 nm

30 femtoseconds from life of a nanoplasmonic system

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



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



nature photonics LETTERS
PUBLISHED ONLINE: 14 DECEMBER 2008 | DOI: 10.1038/NPHOTON.2008.249

Ultrafast active plasmonics

Perturbative (second-order in field) ultrafast intrinsic nonlinearity

Kevin F. MacDonald[1]*, Zsolt L. Sámson[1], Mark I. Stockman[2] and Nikolay I. Zheludev[1]

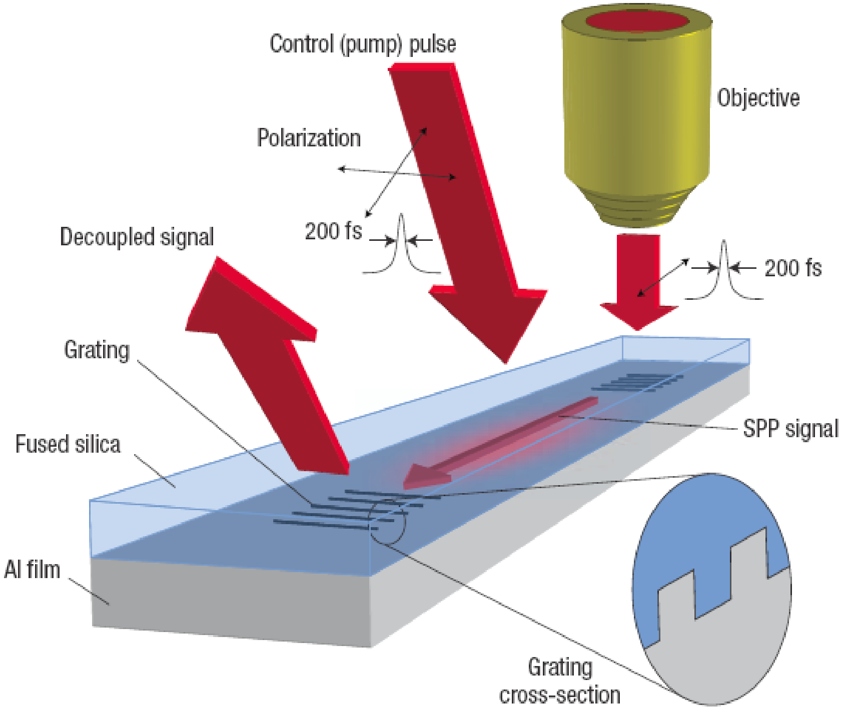
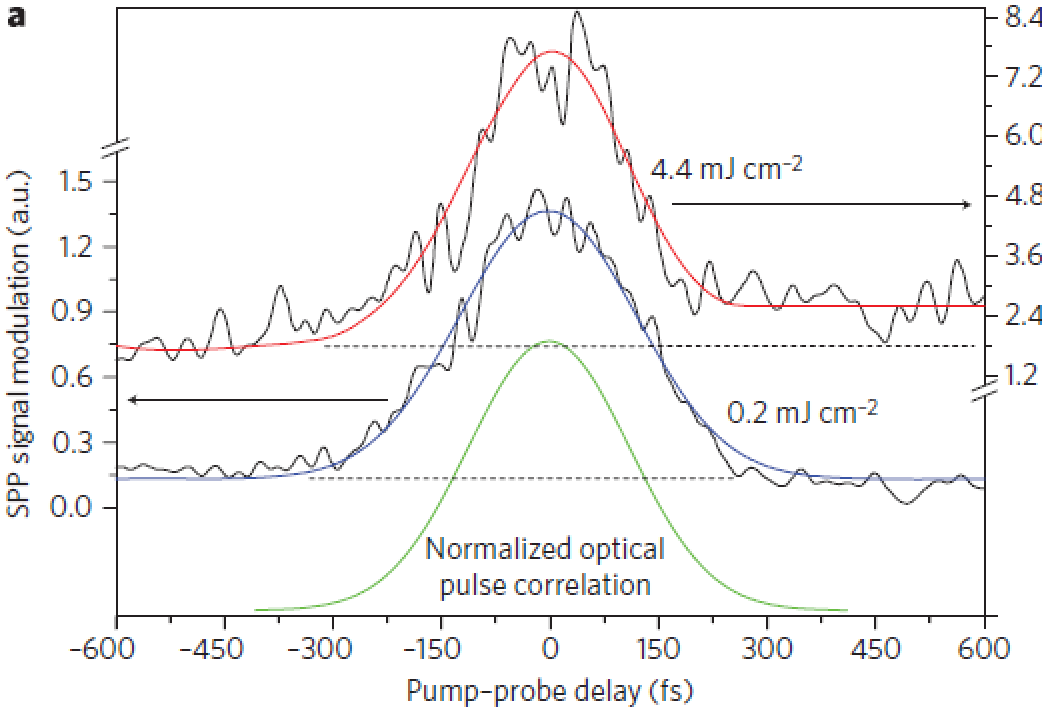
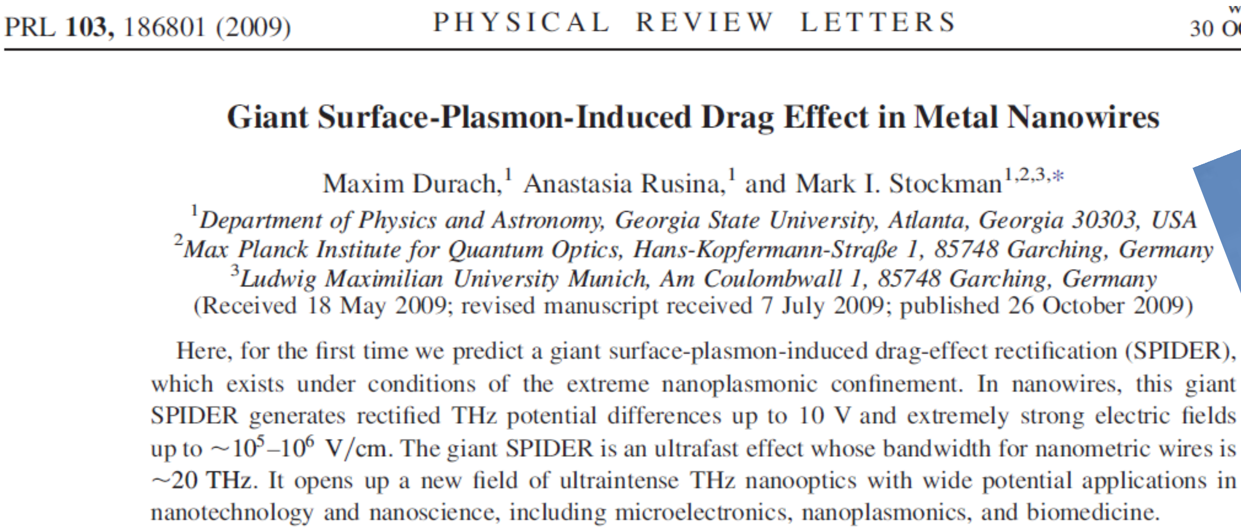


Figure 1 | Ultrafast optical modulation of SPP propagation. A plasmonic signal, coupled to and from the waveguide by gratings on an aluminium/silica interface, is modulated by optical pump pulses as it travels between the gratings.



Giant SPIDER

Perturbative (second-order in field) ultrafast intrinsic nonlinearity



PRL **103,** 186801 (2009) PHYSICAL REVIEW LETTERS week ending 30 OCTOBER 2009

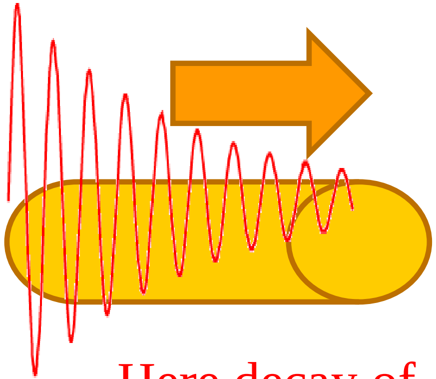
Giant Surface-Plasmon-Induced Drag Effect in Metal Nanowires

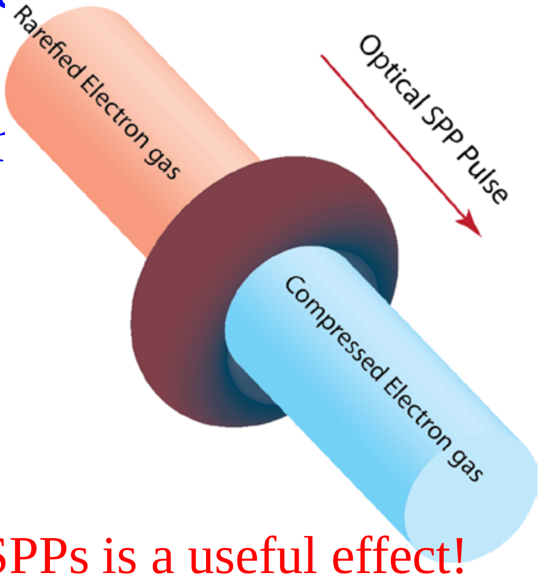
Maxim Durach,[1] Anastasia Rusina,[1] and Mark I. Stockman[1,2,3,*]
[1]*Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303, USA*
[2]*Max Planck Institute for Quantum Optics, Hans-Kopfermann-Straße 1, 85748 Garching, Germany*
[3]*Ludwig Maximilian University Munich, Am Coulombwall 1, 85748 Garching, Germany*

(Received 18 May 2009; revised manuscript received 7 July 2009; published 26 October 2009)

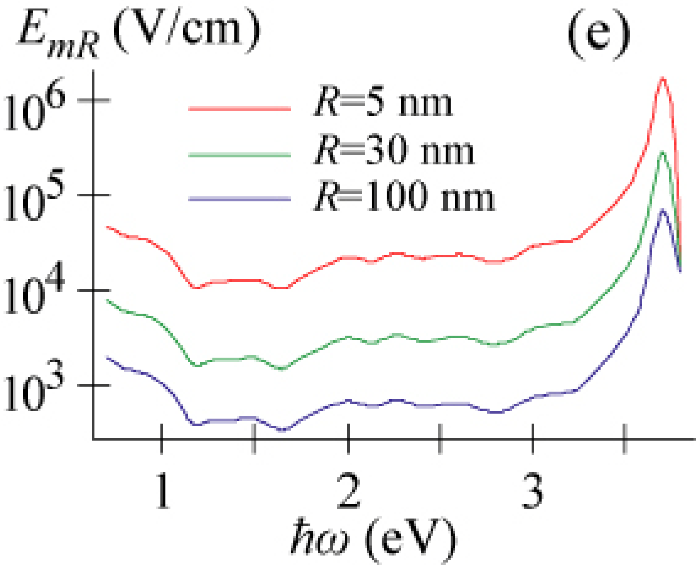
Here, for the first time we predict a giant surface-plasmon-induced drag-effect rectification (SPIDER), which exists under conditions of the extreme nanoplasmonic confinement. In nanowires, this giant SPIDER generates rectified THz potential differences up to 10 V and extremely strong electric fields up to ~10^5–10^6 V/cm. The giant SPIDER is an ultrafast effect whose bandwidth for nanometric wires is ~20 THz. It opens up a new field of ultraintense THz nanooptics with wide potential applications in nanotechnology and nanoscience, including microelectronics, nanoplasmonics, and biomedicine.

Gradient Force and Pressure Force exerted on electro



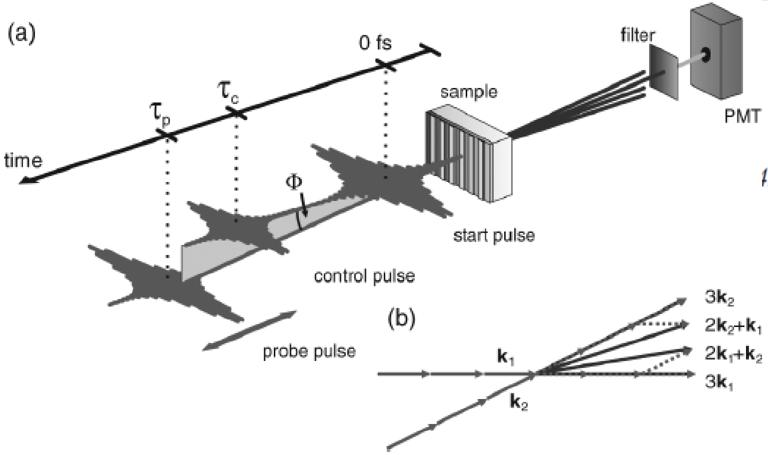


Maximum THz Field by SPIDER



Here decay of SPPs is a useful effect!

PRL **104,** 113903 (2010) PHYSICAL REVIEW LETTERS week ending 19 MARCH 2010



All-Optical Control of the Ultrafast Dynamics of a Hybrid Plasmonic System

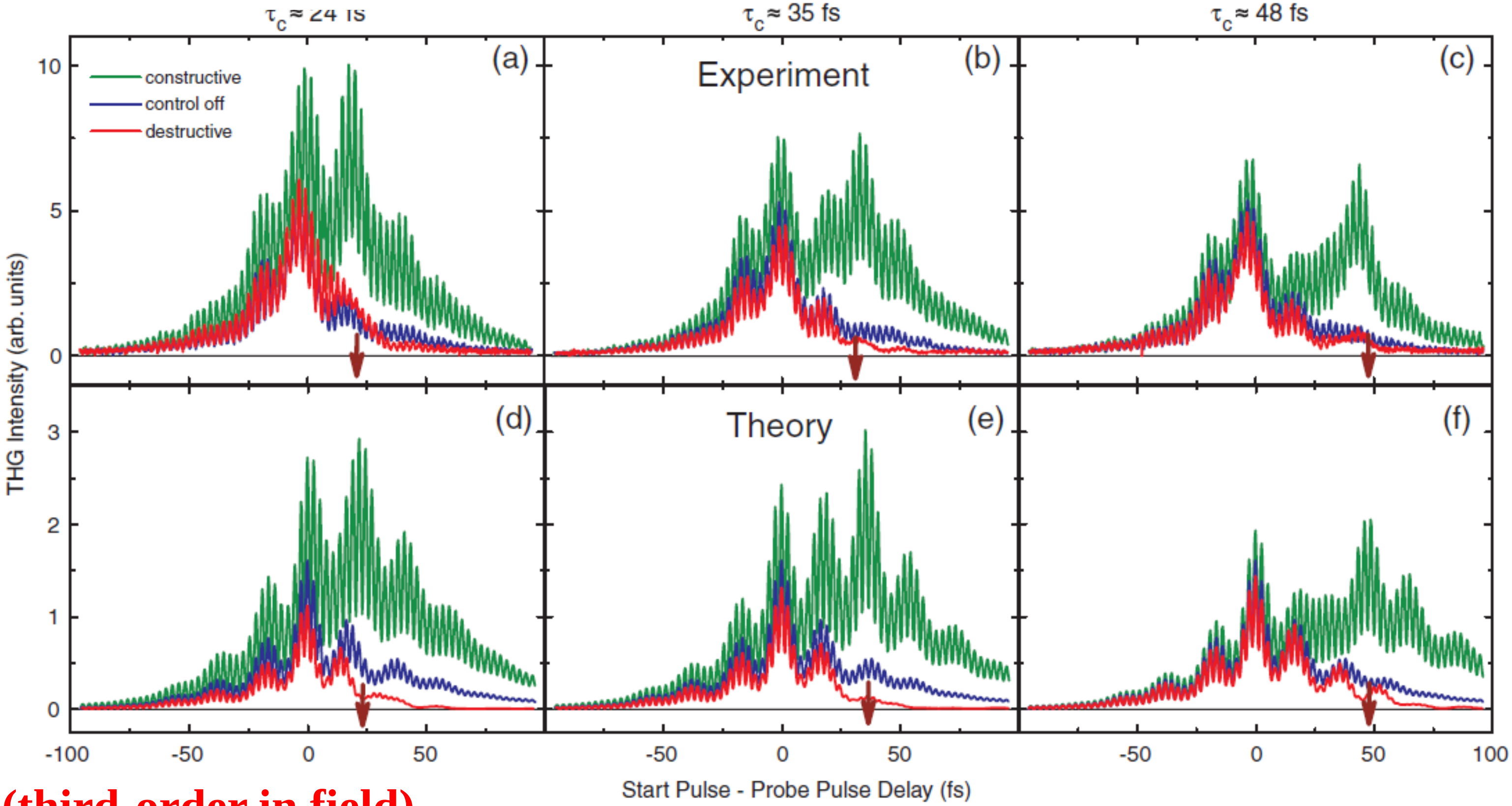
Tobias Utikal,[1,2] Mark I. Stockman,[1,2,3] Albert P. Heberle,[1] Markus Lippitz,[1,2] and Harald Giessen[1]

4th Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, 70550 Stuttgart, Germany
[2]*Max Planck Institute for Solid State Research, Heisenbergstraße 1, 70569 Stuttgart, Germany*
[3]*Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303, USA*

Constructive control by the second pulse

Destructive control by the second pulse

No control pulse (free induction decay)



Perturbative (third-order in field) ultrafast combined nonlinearity

CONTENTS

- Introduction: Plasmonics and Nanoconcentration of Optical Energy
- Plasmonic Enhancement, Ultrafast Nature of Plasmonics, and Plasmonic Nonlinearities
- Perturbative Nonlinearities in Nanoplasmonic Systems
- Nonperturbative (Strong Field) Nonlinearities
- Spaser as an Ultrafast Quantum Generator and Nanoamplifier
- Spasing vs. Loss Compensation as Extrinsic Strong Nonlinearity in Plasmonics
- Conclusions

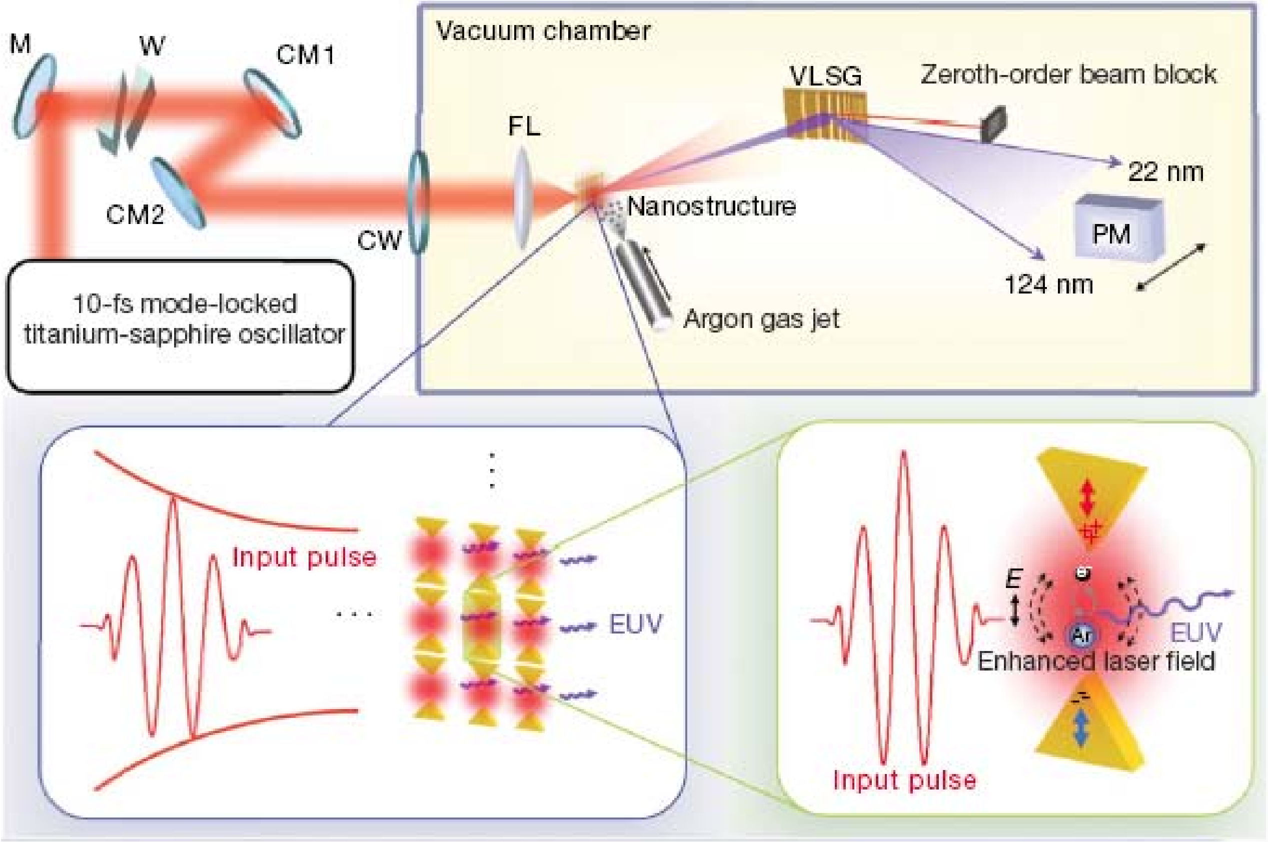
Nonperturbative (strong field) ultrafast extrinsic nonlinearity

Vol 453|5 June 2008|doi:10.1038/nature07012 nature

LETTERS

High-harmonic generation by resonant plasmon field enhancement

Seungchul Kim[1]*, Jonghan Jin[1]*, Young-Jin Kim[1], In-Yong Park[1], Yunseok Kim[1] & Seung-Woo Kim[1]



NATURE|Vol 453|5 June 2008 NEWS & VIEWS

ATTOSECOND PHYSICS

An easier route to high harmony

Mark I. Stockman

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

2. Corkum, P. B. *Phys. Rev. Lett.* **71,** 1994–1997 (1993).
3. Chang, Z., Rundquist, A., Wang, H., Murnane, M. M. & Kapteyn, H. C. *Phys. Rev. Lett.* **79,** 2967–2970 (1997).
4. Paul, P. M. *et al. Science* **292,** 1689–1692 (2001).

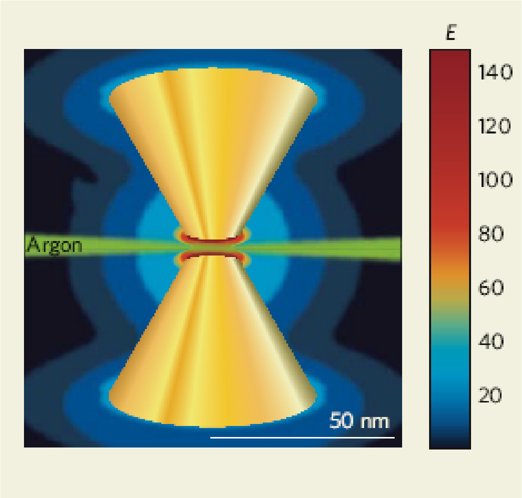
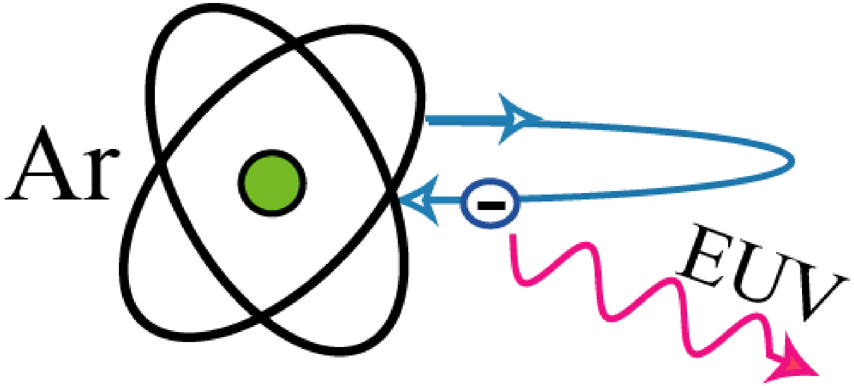


Figure 1 | Stripping on the table-top. The bow-tie-shaped gold nanoantennas used by Kim *et al.*[1] develop electric-field strengths in the gap

Above threshold ionization and electron recollision in high harmonic generation



Computations courtesy Javier Aizpurua

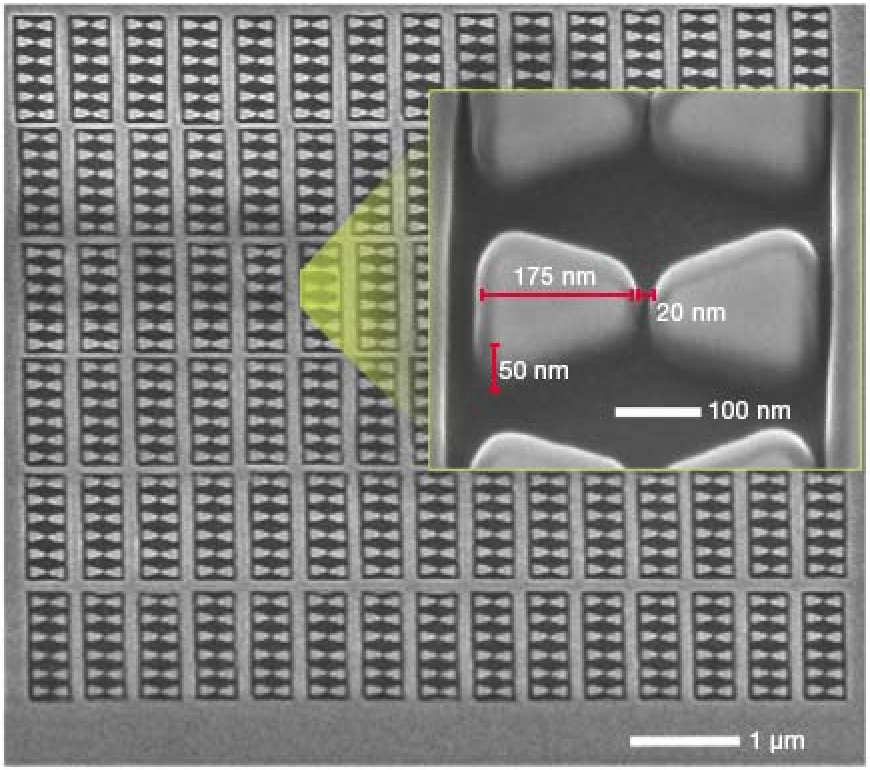


Figure 3 | Scanning electron microscope image of the nanostructure used for high-harmonic generation. Bow-tie elements were arranged in a two-dimensional, 36 × 15 array with an area of 10 µm × 10 µm. The inset shows the magnified image of a single bow-tie element with the important dimensions marked.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

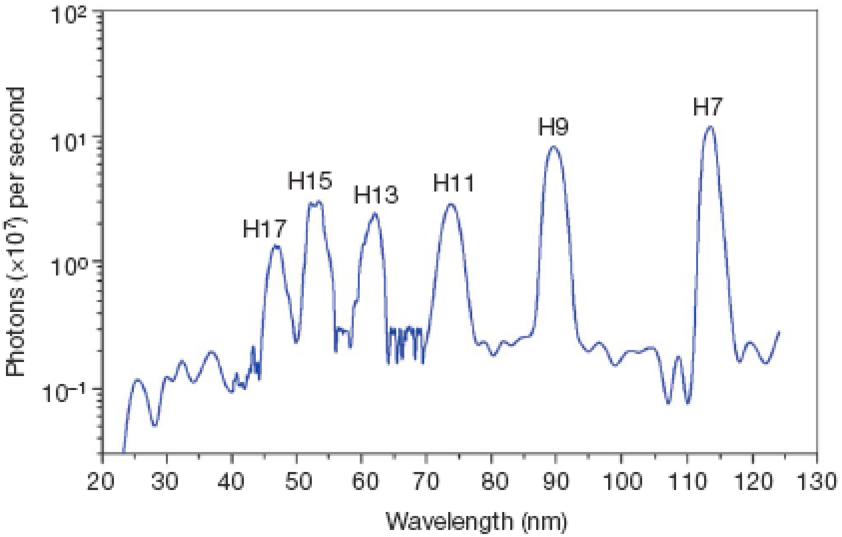


Figure 4 | Measured spectrum of generated high harmonics. A varied-line-

CONTENTS

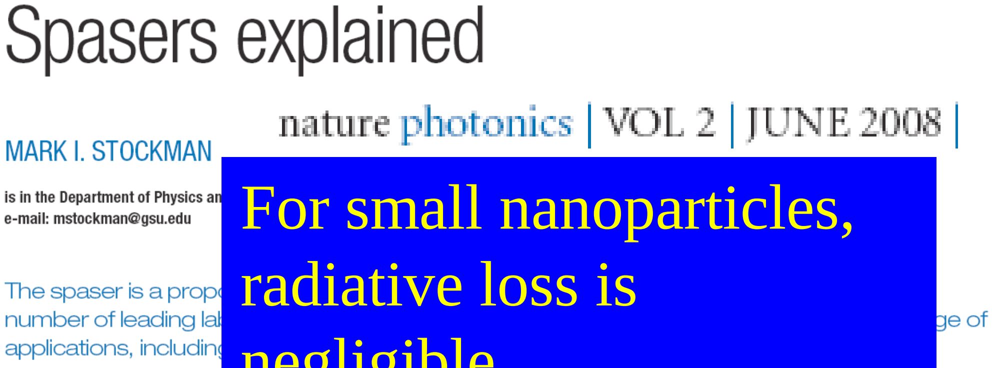
Surface Plasmon Amplification by Stimulated Emission of Radiation (SPASER)

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).

Amplification and Stimulated Emission in Plasmonic systems:

1. E. Plum, V. A. Fedotov, P. Kuo, D. P. Tsai, and N. I. Zheludev, *Towards the Lasing Spaser: Controlling Metamaterial Optical Response with Semiconductor Quantum Dots*, Opt. Expr. **17**, 8548-8551 (2009)
2. P. M. Bolger, W. Dickson, A. V. Krasavin, L. Liebscher, S. G. Hickey, D. V. Skryabin, and A. V. Zayats, *Amplified Spontaneous Emission of Surface Plasmon Polaritons*, Opt. Lett. **35**, 1197-1199 (2010).
3. 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)

Department of Physics and A
Georgia State University
Atlanta, GA 30303-3083



Spasers explained

nature photonics | VOL 2 | JUNE 2008 |

MARK I. STOCKMAN

is in the Department of Physics an
e-mail: mstockman@gsu.edu

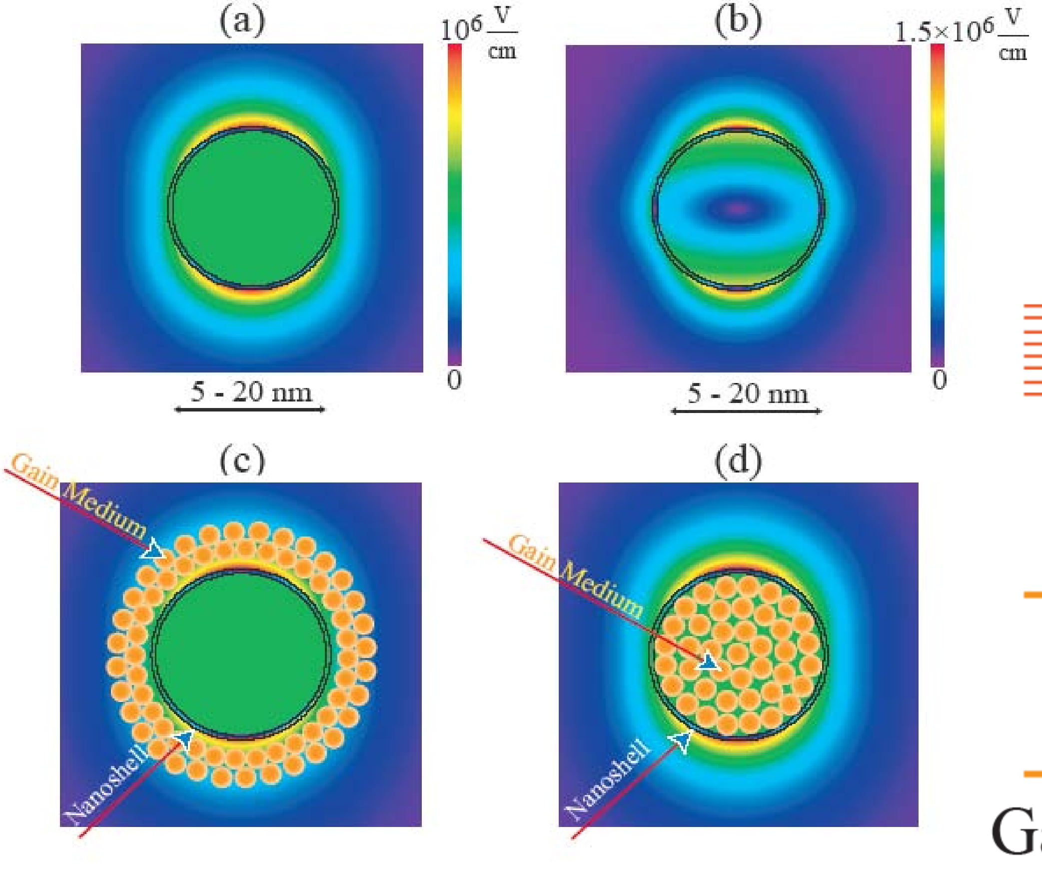
The spaser is a propo
number of leading lal
applications, including

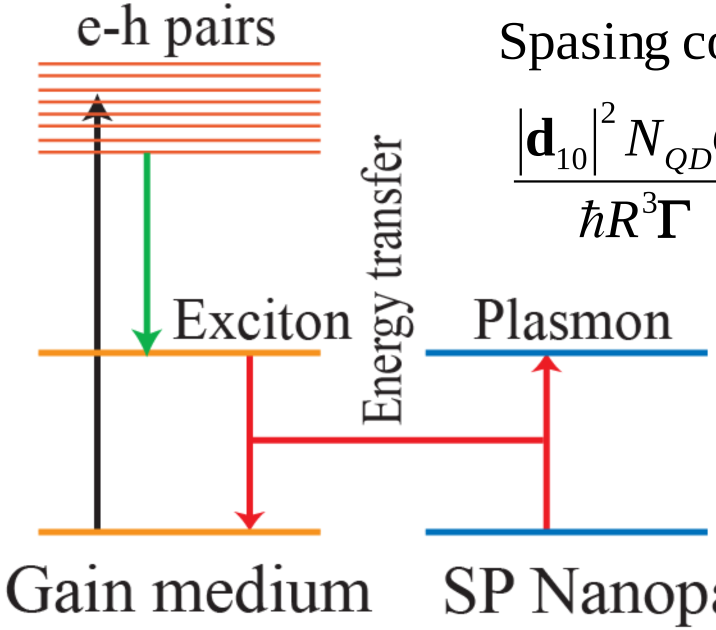
ge of

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

For small nanoparticles, radiative loss is negligible.

Spaser is quasistatic and fully scalable





Spasing condition :

$$\frac{\left|\mathbf{d}_{10}\right|^2 N_{QD} Q}{\hbar R^3 \Gamma} \geq 1$$

Nonperturbative (strong field) ultrafast extrinsic nonlinearity

d Ast

University

Atlanta, GA 30303-3083

Plasmon number

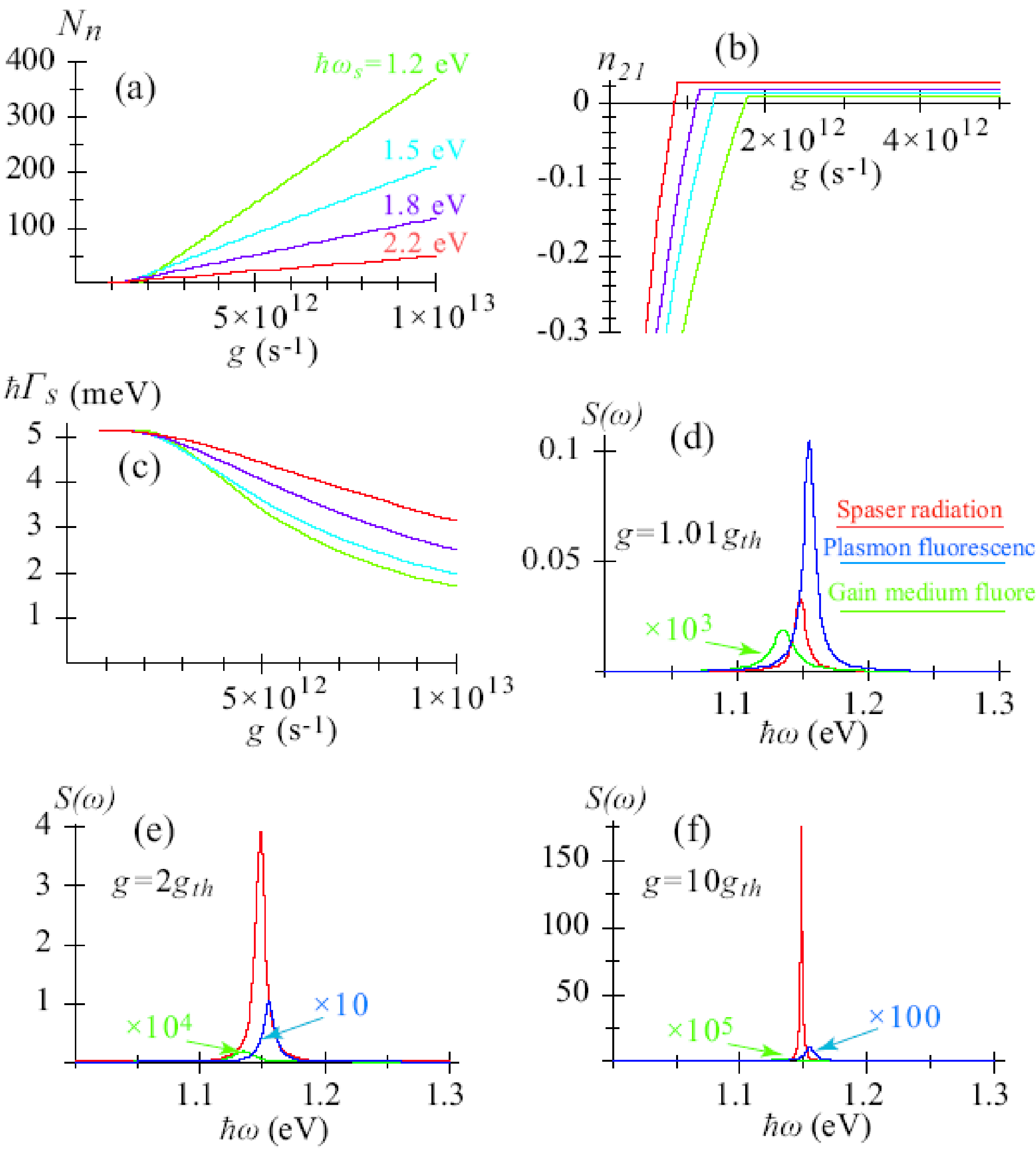
Inversion vs. pumping rate

Stationary (CW) spaser regime

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

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

$$\text{Spaser linewidth} \propto N_{SP}^{-1}$$

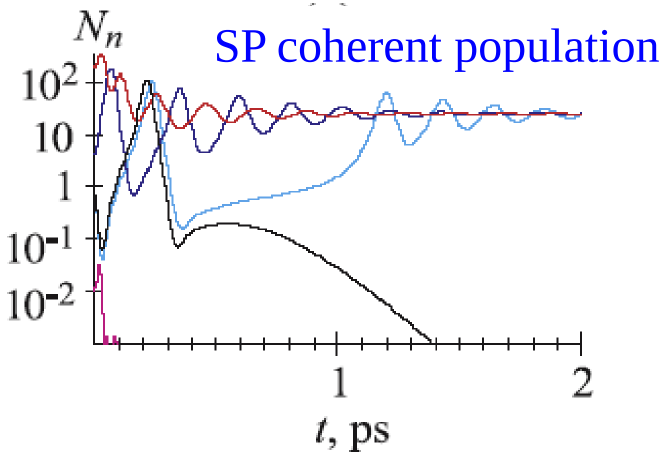


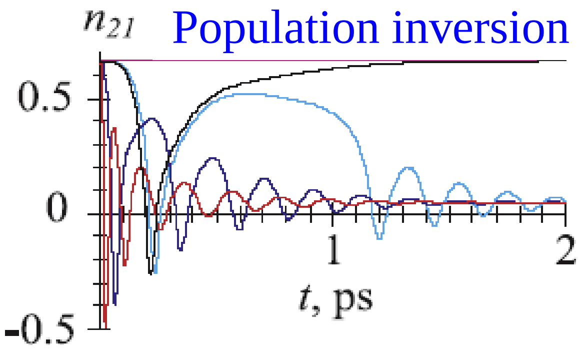
Bandwidth ~ 10-100 THz

Very high resistance to ionizing radiation

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

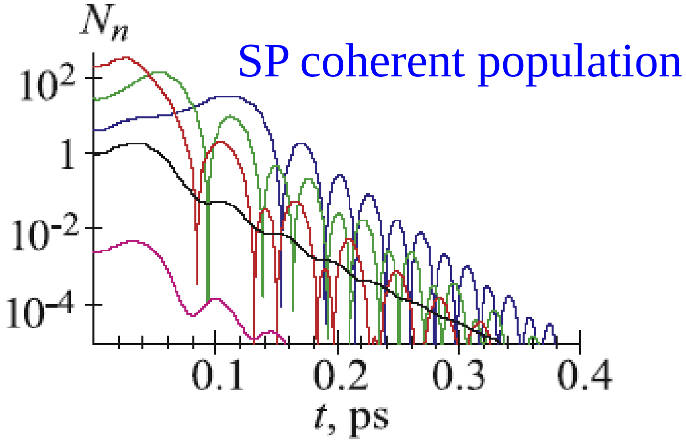
Stationary pumping

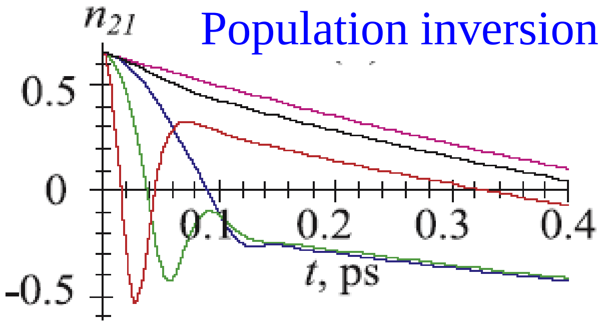

SP coherent population

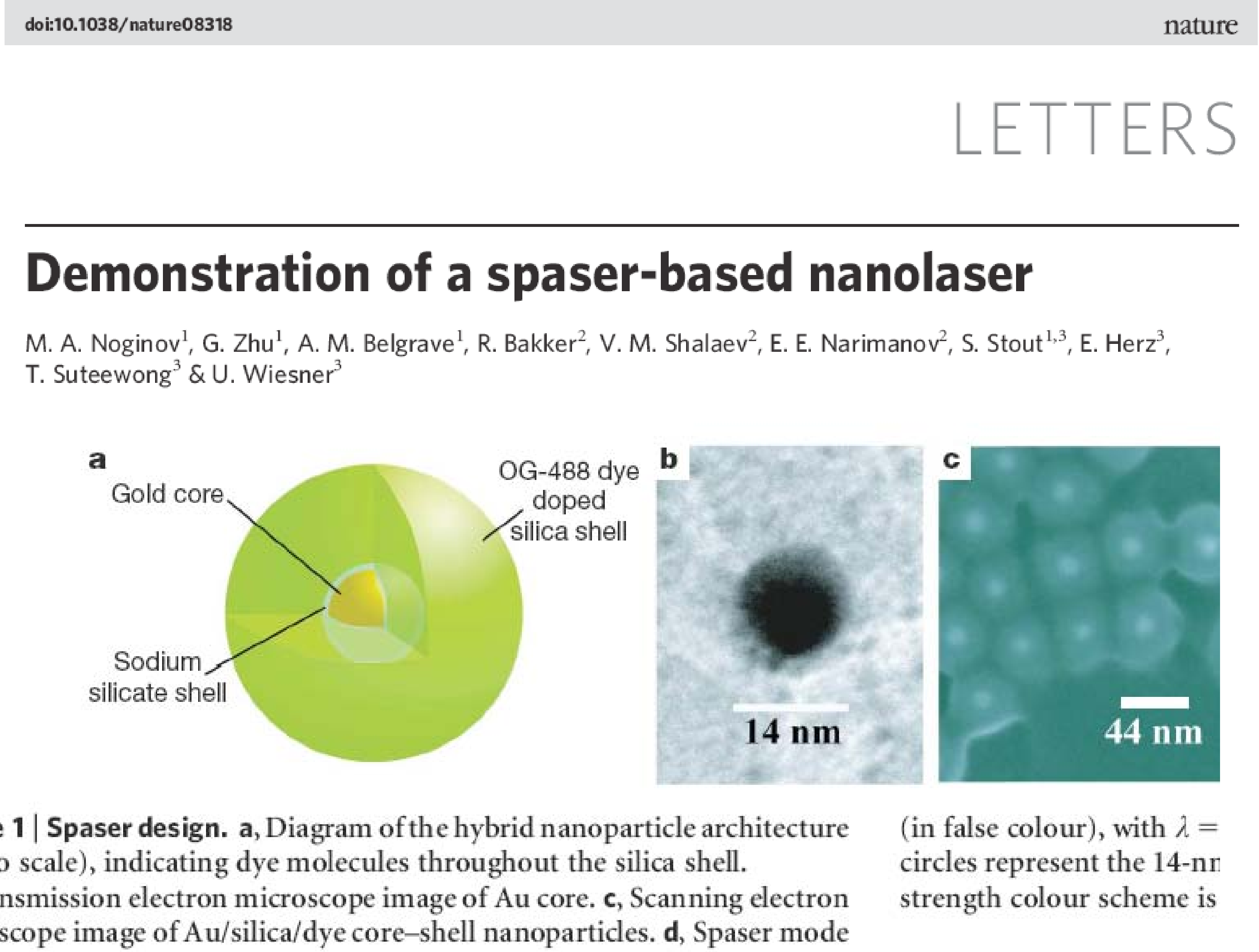

Population inversion

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

Pulse pumping


SP coherent population


Population inversion



doi:10.1038/nature08318 **nature**

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 $\lambda =$
circles represent the 14-n
strength colour scheme is

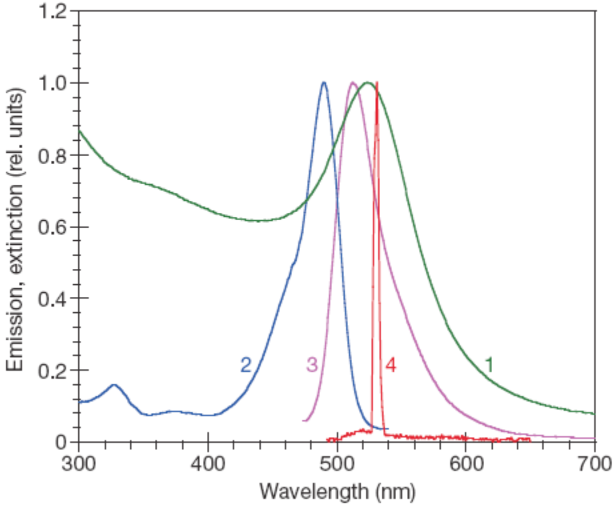


Figure 2 | Spectroscopic results. Normalized extinction (1), excitation (2), spontaneous emission (3), and stimulated emission (4) spectra of Au/silica/dye nanoparticles. The peak extinction cross-section of the nanoparticles is 1.1×10^{-12} cm^2. The emission and excitation spectra were measured in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

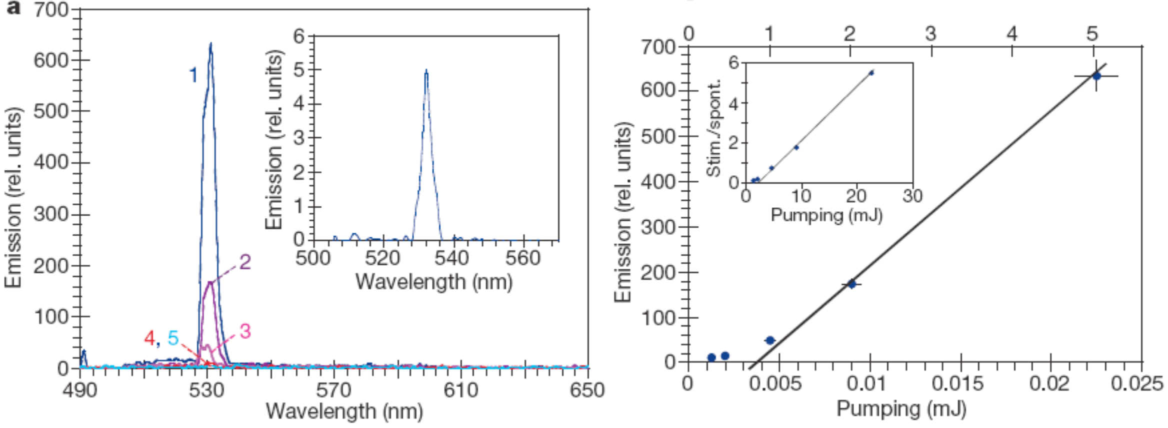


Figure 4 | Stimulated emission. **a**, Main panel, stimulated emission spectra of the nanoparticle sample pumped with 22.5 mJ (1), 9 mJ (2), 4.5 mJ (3), 2 mJ (4) and 1.25 mJ (5) 5-ns optical parametric oscillator pulses at $\lambda = 488$ nm. **b**, Main panel, corresponding input–output curve (lower 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]

[1]*COBRA Research Institute, Technische Universiteit Eindhoven, Postbus 513, 5600 MB Eindhoven, The Netherlands*
[2] *Department of Electrical Engineering, Arizona State University, Tempe AZ 85287, USA*
**m.t.hill@ieee.org*

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

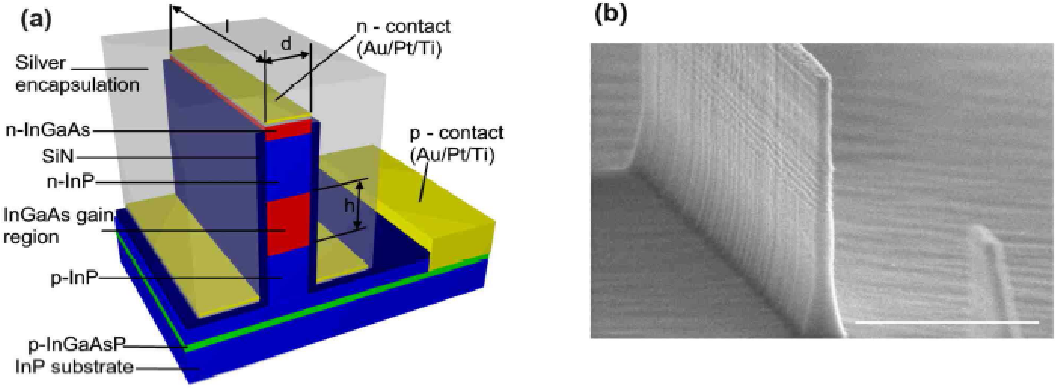
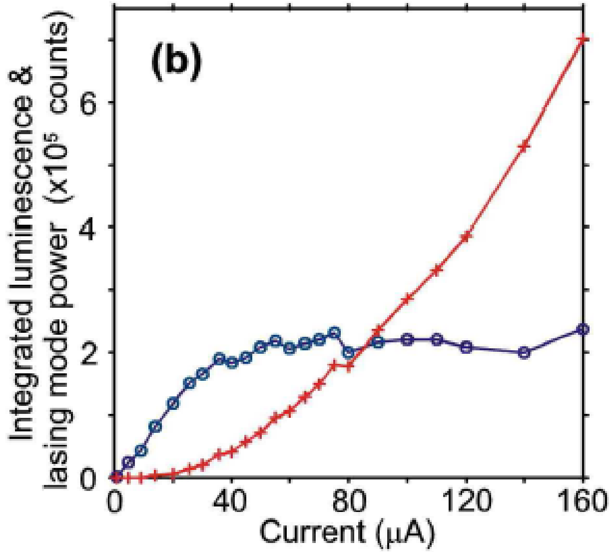
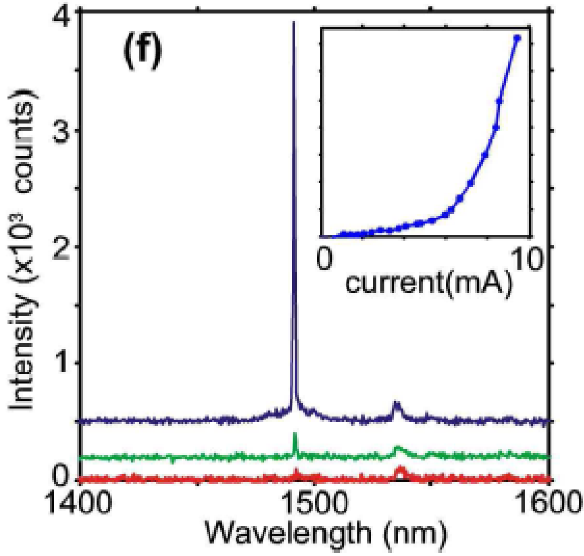


Fig. 1. Structure of cavity formed by a rectangular semiconductor pillar encapsulated in Silver. (a) Schematic showing the device layer structure. (b) Scanning electron microscope image showing the semiconductor core of one of the devices. The scale bar is 1 micron.



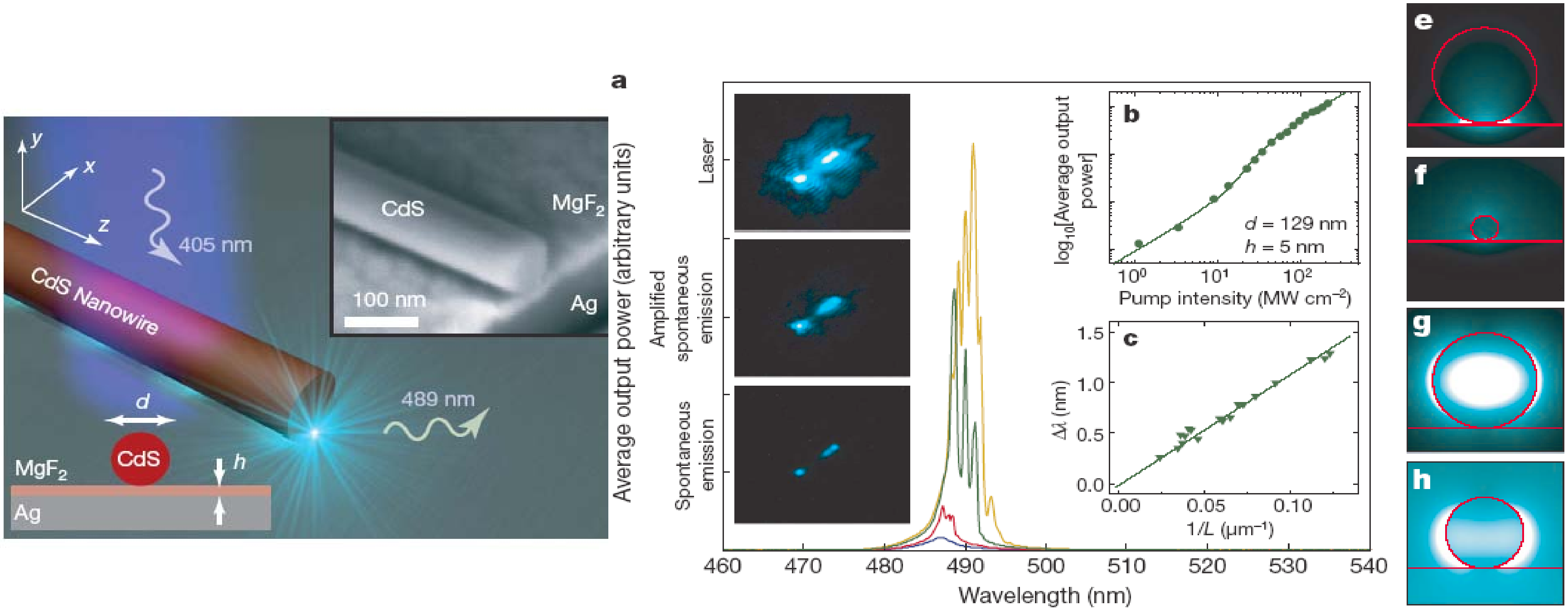


doi:10.1038/nature08364 **nature**

LETTERS

Plasmon lasers at deep subwavelength scale

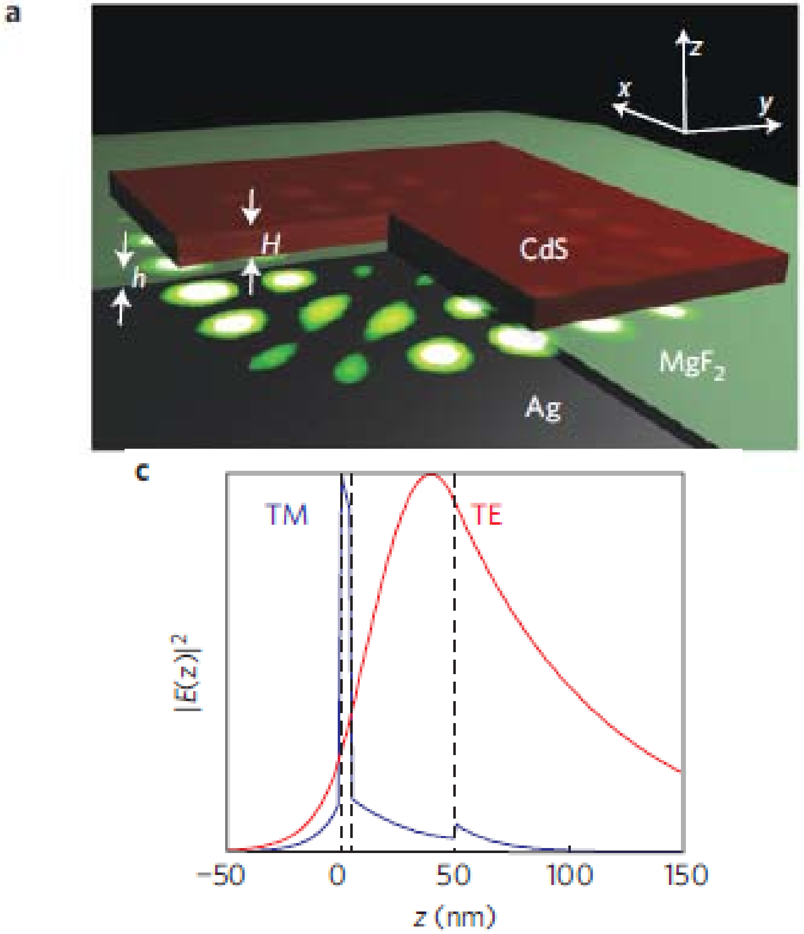
Rupert F. Oulton[1]*, Volker J. Sorger[1]*, Thomas Zentgraf[1]*, Ren-Min Ma[3], Christopher Gladden[1], Lun Dai[3], Guy Bartal[1] & Xiang Zhang[1,2]

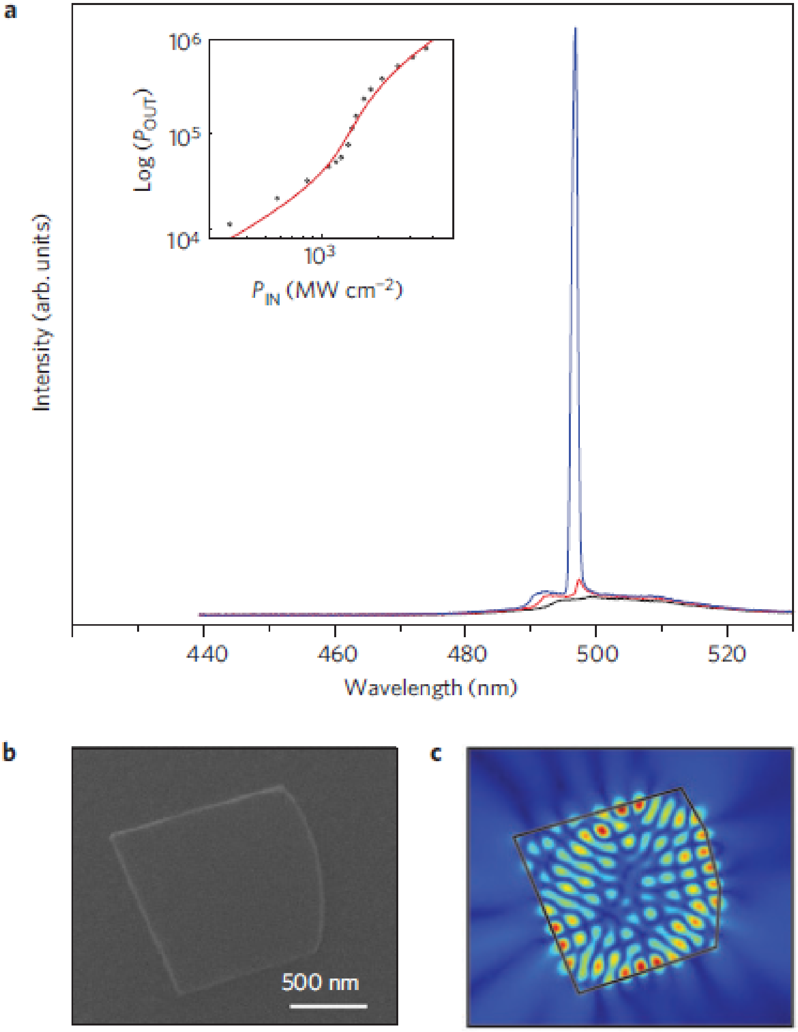


nature materials
LETTERS
PUBLISHED ONLINE: 19 DECEMBER 2010 | DOI: 10.1038/NMAT2919

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]

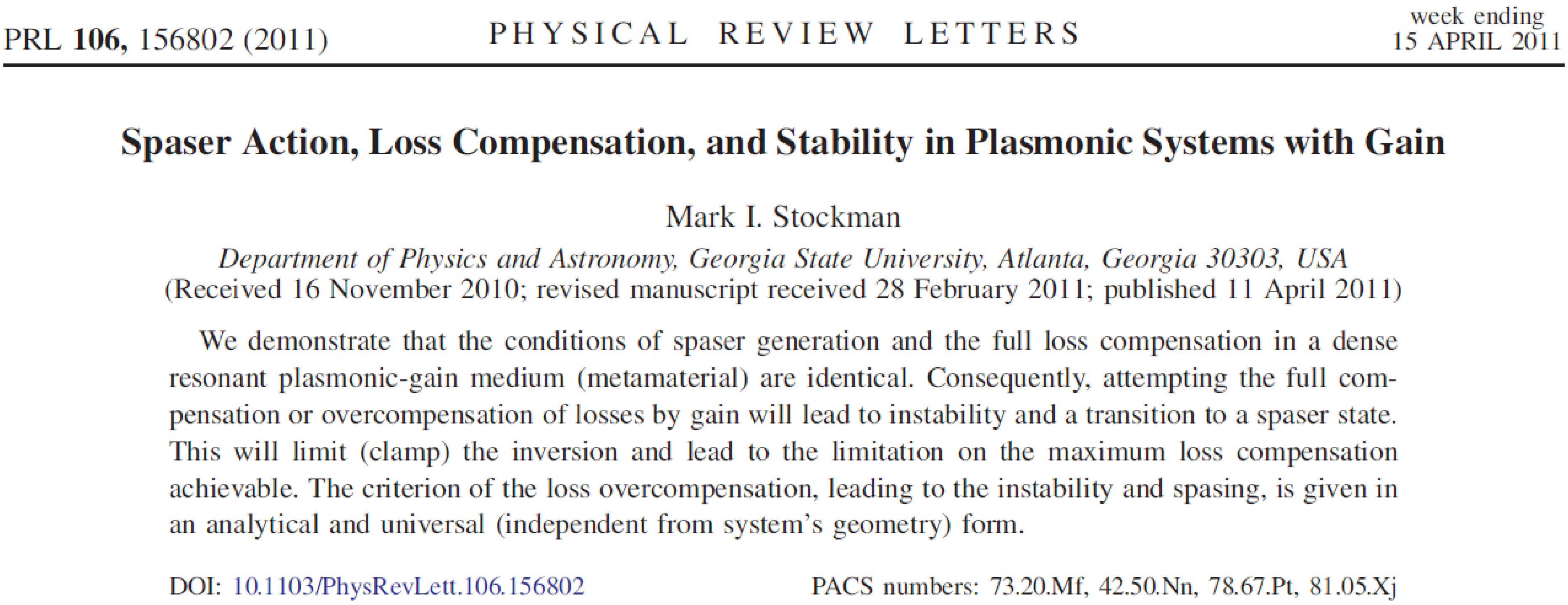




CONTENTS

- Introduction: Plasmonics and Nanoconcentration of Optical Energy
- Plasmonic Enhancement, Ultrafast Nature of Plasmonics, and Plasmonic Nonlinearities
- Perturbative Nonlinearities in Nanoplasmonic Systems
- Nonperturbative (Strong Field) Nonlinearities
- Spaser as an Ultrafast Quantum Generator and Nanoamplifier
- Spasing vs. Loss Compensation as Extrinsic Strong Nonlinearity in Plasmonics
- Conclusions

Nonperturbative (strong field) ultrafast extrinsic nonlinearity



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

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

Mark I. Stockman

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

(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

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

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

Hear E is the macroscopic field and $\mathbf{e}(\mathbf{r})$ is the (mesoscopic) local field inside the metamaterial. This local field is expressed as an eigenmode expansion

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

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 effective permittivity is (where a_n is a coefficient):

$$\bar{\varepsilon}(\omega) = |a_n|^2[s_n\varepsilon_m(\omega) + (1-s_n)\varepsilon_h(\omega)]$$

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

$$\varepsilon_h(\omega) = \varepsilon_d - i\frac{4\pi}{3}\frac{|\mathbf{d}_{12}|^2 n_c}{\hbar\Gamma_{12}}, \tag{6}$$

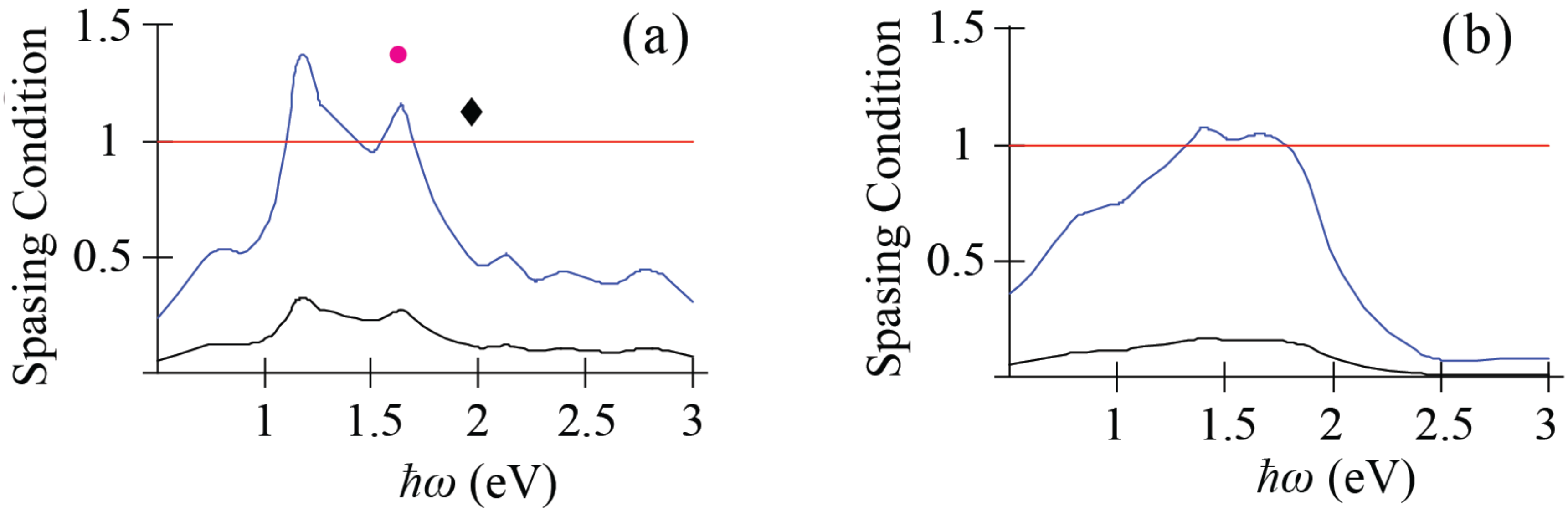
where $\varepsilon_d = \mathrm{Re}\,\varepsilon_h$, $\mathbf{d}_{12}$ is the dipole matrix element of the gain transition in a chromophore center of the gain medium, Γ_{12} is a spectral width of this transition, and n_c is the concentration of these centers.

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

$$\frac{4\pi}{3}\frac{|\mathbf{d}_{12}|^2 n_c[1 - \mathrm{Re}\,s(\omega)]}{\hbar\Gamma_{12}\mathrm{Re}\,s(\omega)\mathrm{Im}\,\varepsilon_m(\omega)} \ge 1,$$

$$\frac{4\pi}{3}\frac{\left|\mathbf{d}_{12}\right|^{2} n_c\left[1-\mathrm{Re}\,s(\omega)\right]}{\hbar\Gamma_{12}\mathrm{Re}\,s(\omega)\mathrm{Im}\,\varepsilon_m(\omega)}\geq 1$$

- This is a criterion 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



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 6 x10^{18} cm^{-3} for the lower curve (black) and nc = 3x10^{19} cm^{-3} for the upper curve (blue on line). 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 3x10^{19} cm^{-3} for the lower curve (black) and 2x10^{20} cm^{-3} for the upper curve (blue on line).

1. S. Wuestner, A. Pusch, K. L. Tsakmakidis, J. M. Hamm, and O. Hess, *Overcoming Losses with Gain in a Negative Refractive Index Metamaterial*, Phys. Rev. Lett. **105**, 127401-1-4 (2010).
2. 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).
3. 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).

Department of Physics and Astr
Georgia State University
Atlanta, GA 30303-3083

The recently emerged fields of metamaterials and transformation optics promise a family of exciting applications such as invisibility, optical imaging with deeply subwavelength resolution and nanophotonics with the potential for much faster information processing. The possibility of creating optical negative-index metamaterials (NIMs) using nanostructured metal–dielectric composites has triggered intense basic and applied research over the past several years[1–10]. However, the performance of all NIM applications is significantly limited by the inherent and strong energy dissipation in metals, especially in the near-infrared and visible wavelength ranges[11,12]. Generally the losses are orders of magnitude too large for the proposed applications, and the reduction of losses with optimized designs seems to be out of reach. One way of addressing this issue is to incorporate gain media into NIM designs[13–16]. However, whether NIMs with low loss can be achieved has been the subject of theoretical debate[17,18]. Here we experimentally demonstrate that the incorporation of gain material in the high-local-field areas of a metamaterial makes it possible to fabricate an extremely low-loss and 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.

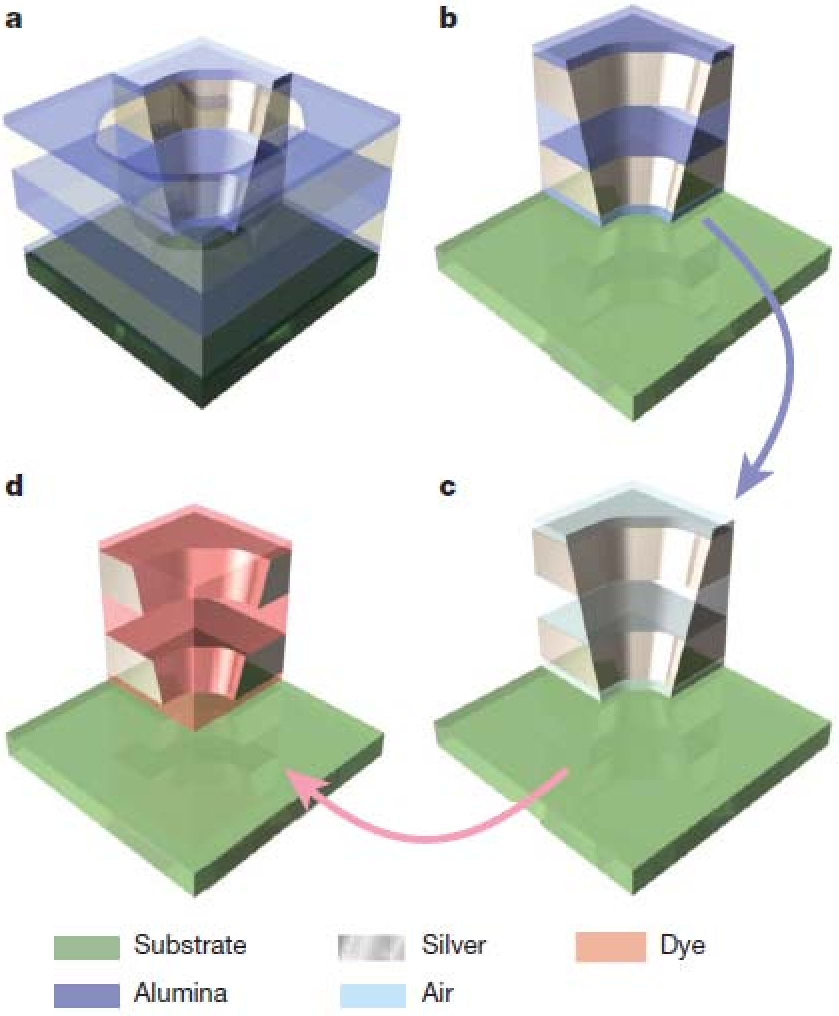
Vol 466 | 5 August 2010 | doi:10.1038/nature09278

nature

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]



PRL **105,** 127401 (2010) PHYSICAL REVIEW LETTERS week ending 17 SEPTEMBER 2010

Overcoming Losses with Gain in a Negative Refractive Index Metamaterial

Sebastian Wuestner, Andreas Pusch, Kosmas L. Tsakmakidis, Joachim M. Hamm, and Ortwin Hess*

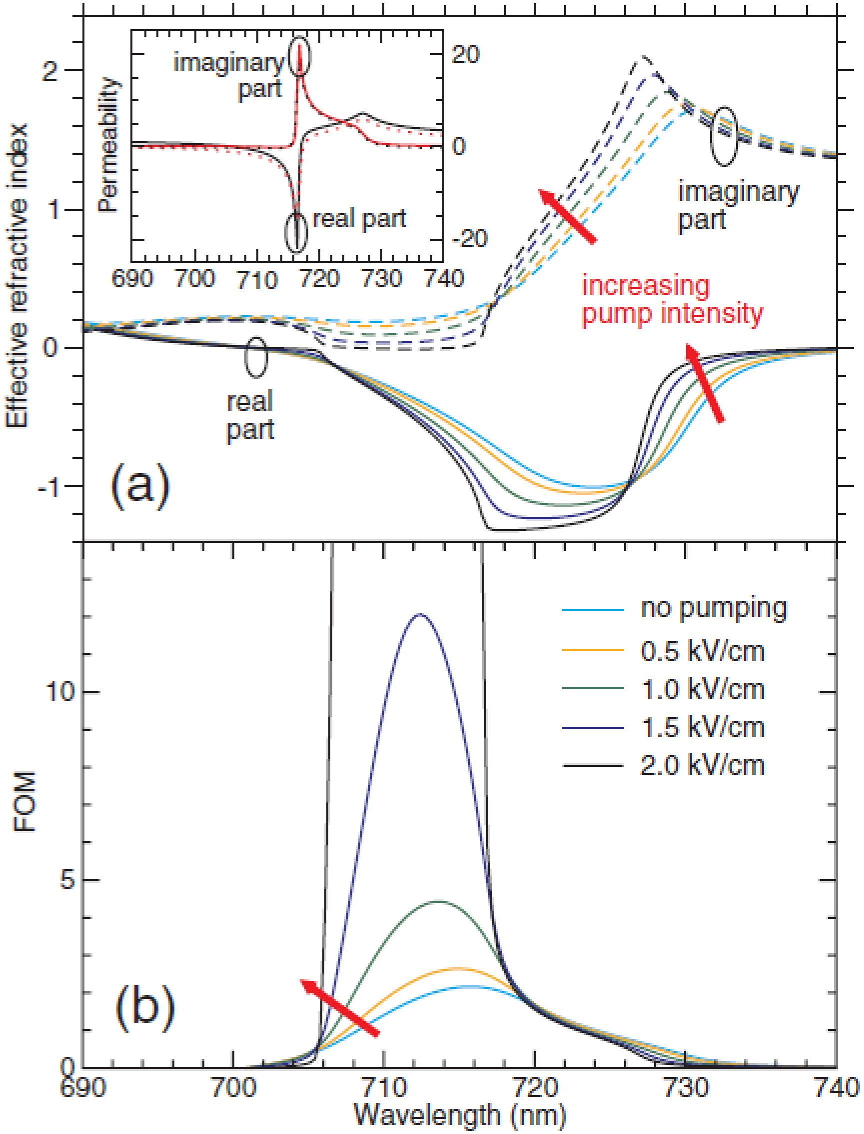


FIG. 3 (color online). (a) Real and imaginary part of the retrieved effective refractive indices of the double-fishnet structure for different pump amplitudes. The peak electric field amplitude of the pump increases in steps of 0.5 kV/cm from no pumping (cyan line, lightest) to a maximum of 2.0 kV/cm (black line, darkest). The inset shows the real and imaginary part of the effective permeability (black and red line, respectively) and the result of the Kramers-Kronig relation (black and red dotted lines) for the highest peak electric field amplitude of 2.0 kV/cm. (b) The figures-of-merit (FOM) for the same pumping amplitudes.

CONTENTS

- Introduction: Plasmonics and Nanoconcentration of Optical Energy
- Plasmonic Enhancement, Ultrafast Nature of Plasmonics, and Plasmonic Nonlinearities
- Perturbative Nonlinearities in Nanoplasmonic Systems
- Nonperturbative (Strong Field) Nonlinearities
- Spaser as an Ultrafast Quantum Generator and Nanoamplifier
- Spasing vs. Loss Compensation as Extrinsic Strong Nonlinearity in Plasmonics
- Conclusions

BRIEF CONCLUSIONS

1. Nanoplasmonics is based on nanolocalization of optical fields due to SPs
2. Enhancement in nanoplasmonics is due to quality factor of SP modes and geometric concentration. Nanoplasmonics is ultrafast with ~100 as – 10 fs characteristic times
3. Plasmonic nonlinearities are classified as intrinsic (in the metal), extrinsic (in the embedding dielectric, or combined. Independently, they are classified as perturbative or nonperturbative (strong field) nonlinearities
4. There are abundant examples of ultrafast nonlinear perturbative nonlinearities: multiphoton electron emission, Kerr effect, nonlinear absorption, three- and four-wave mixing, etc. There are also strong-field nonlinearities observed of an extrinsic type (high-harmonic generation, nonlinear photo-ionization, SPASER) and predicted of an intrinsic type (plasmon-solitons, metallization, etc.)
5. SPASER is an efficient nanoscale generator and ultrafast quantum amplifier with a switch time ~100 fs for silver and ~10 fs for gold. It has the same size as MOSFET and can perform the same functions but is ~1000 times faster. Spaser is classified as a nonperturbative extrinsic nonlinear ultrafast effect
6. SPASERs have been observed in a number of experiments
7. Attempting the full loss compensation in a dense isotropic resonant plasmonic metamaterial is shown to lead to spasing that clamps the gain and makes this full compensation impossible. This is, as the SPASER itself, an extrinsic nonperturbative nonlinear effect

END