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WINTER COLLEGE ON OPTICS ON OPTICS AND PHOTONICS IN NANOSCIENCE AND NANOTECHNOLOGY

(7 - 18 February 2005)

"Optical Properties of Plasmonic Nanosystems"- III

presented by:

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These are preliminary lecture notes, intended only for distribution to participants.

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Support:

Theory of Nanoplasmonics 3: Optical Properties of Plasmonic Nanosystems

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LECTURE 3

Ultrafast and Quantum Nanoplasmonics

1. Introduction: Problem of nanoscale control of local optical fields
2. Coherent control using phase (temporal) degrees of freedom of ultrashort pulse in linear and nonlinear processes.
3. Two-pulse (interferometric) coherent control: full suppression or enhancement of near field hot spots for two-photon effects
4. Surface plasmon amplification by stimulated emission of radiation (SPASER)

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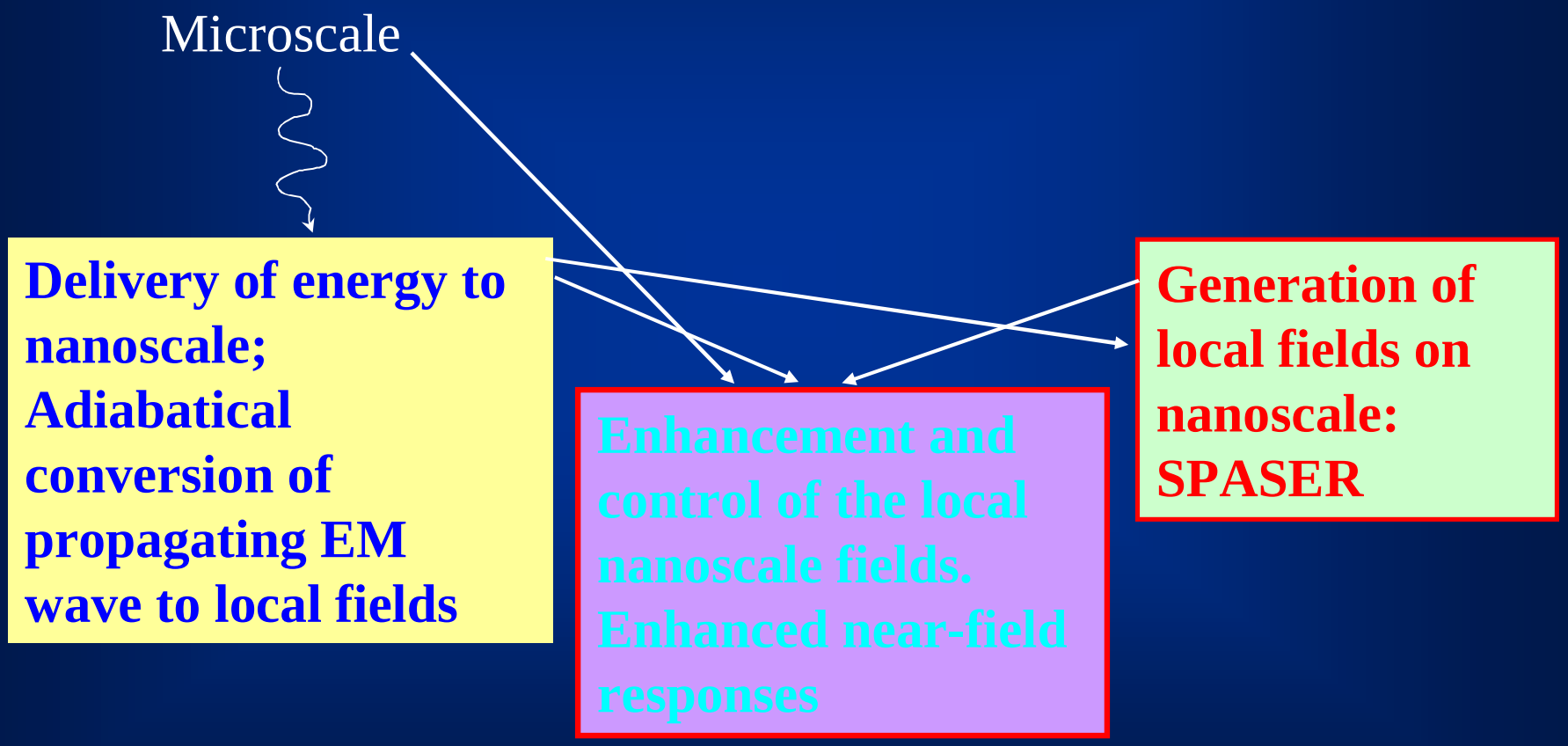
PROBLEMS IN NANOOPTICS



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COHERENT CONTROL OF THE NANOSCALE LOCALIZATION OF ULTRAFAST OPTICAL EXCITATION ENERGY

- 1. M. I. Stockman, S. V. Faleev, and D. J. Bergman, *Coherent Control of Femtosecond Energy Localization in Nanosystems*, Phys. Rev. Lett. **88,** 067402 (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**, 054202-1-10 (2004).

Problem: 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, this optical radiation does possess spectral (phase) degrees of freedom

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•Femtosecond local fields on nanoscale:

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

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

$$E_z^{(0)}(t) = \exp\left[i\omega_0 \left(1+\alpha\frac{t-T/2}{T}\right)(t-T/2) - \frac{3}{2}\left(\frac{t-T/2}{T}\right)^2\right] + \text{c.c.}$$

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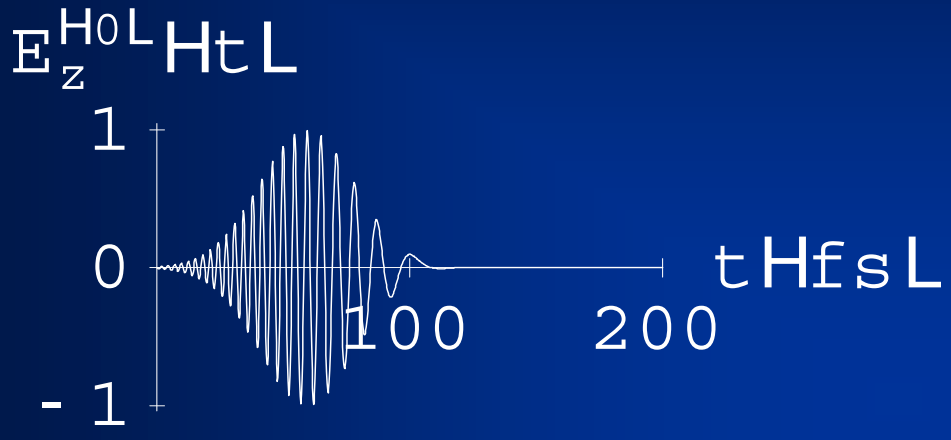

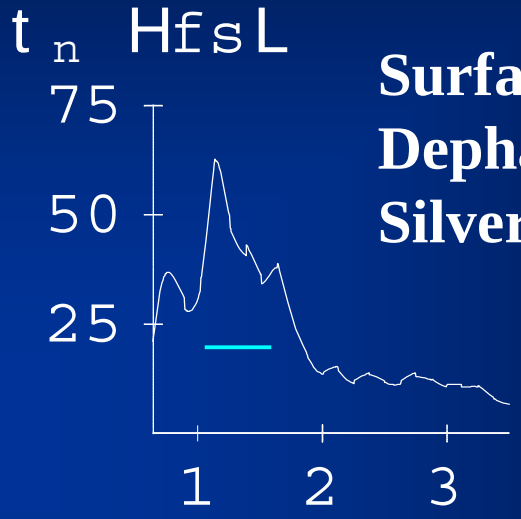

Surface Plasmon Dephasing Time for Silver

See also: J. Bosbach, C. Hendrich, F. Stietz, T. Vartanyan, and F. Trager, *Ultrafast Dephasing of Surface Plasmon Excitation in Silver Nanoparticles: Influence of Particle Size, Shape, and Chemical Surrounding*, Phys. Rev. Lett. **89**, 257404 (2002).

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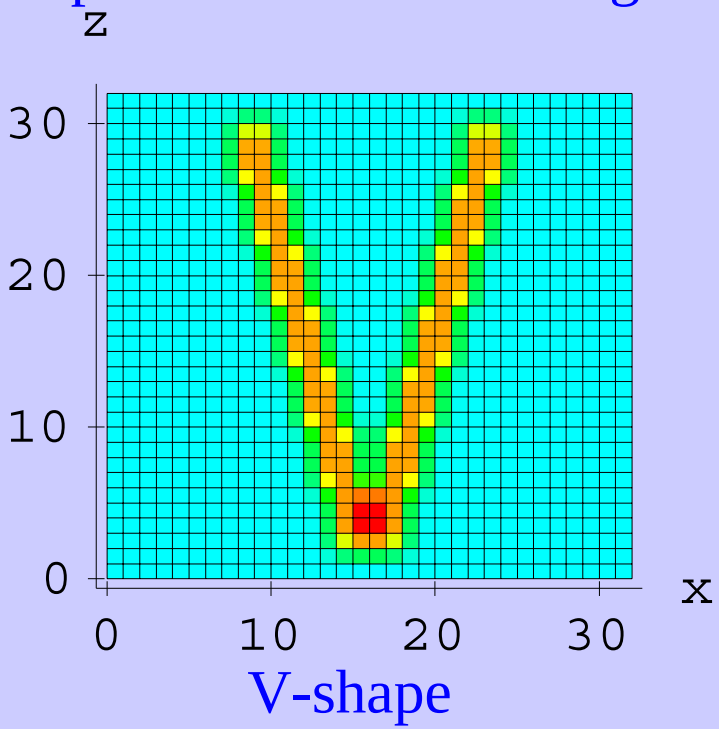
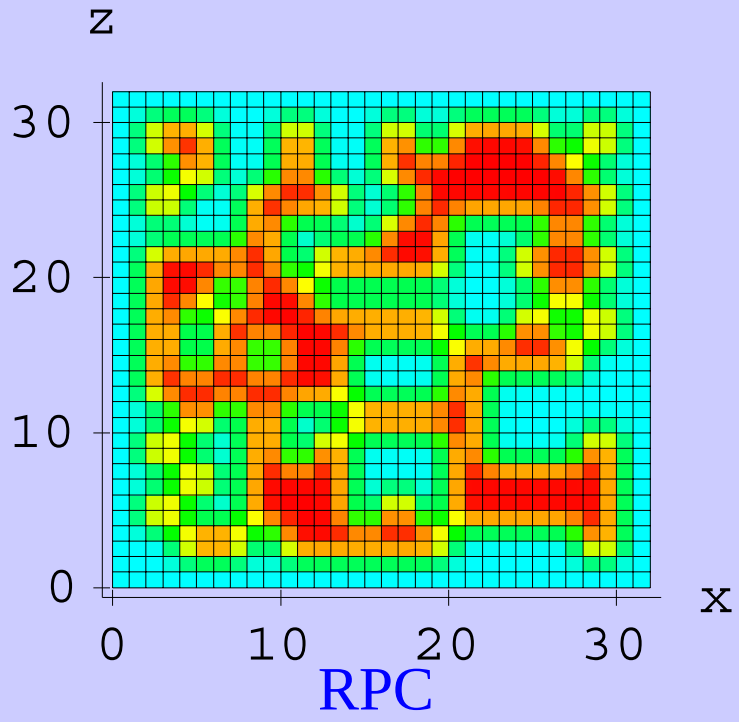
RESULTS

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.



V-shape



RPC

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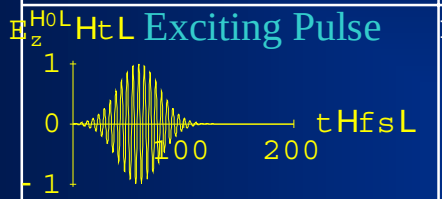
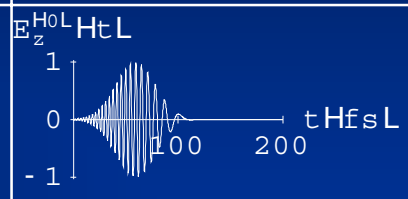
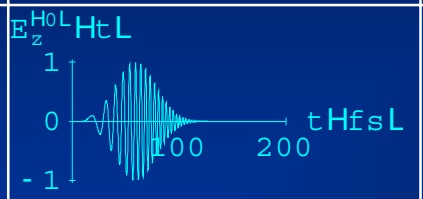
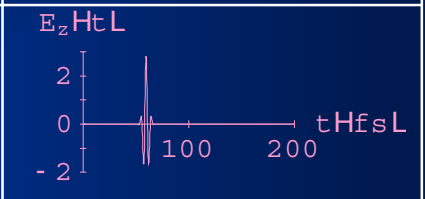
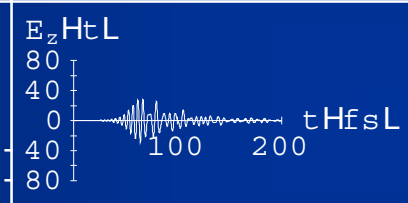
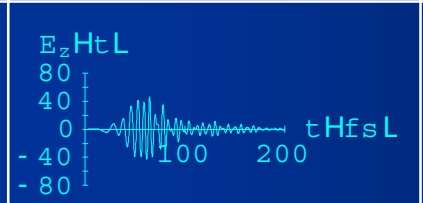
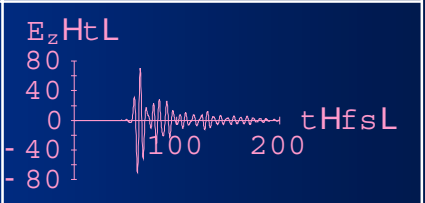
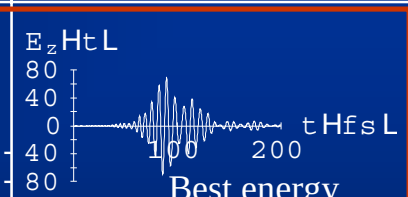
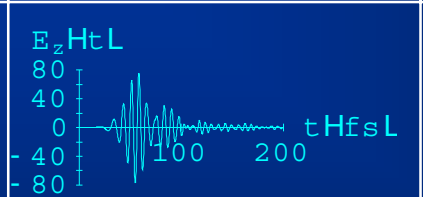
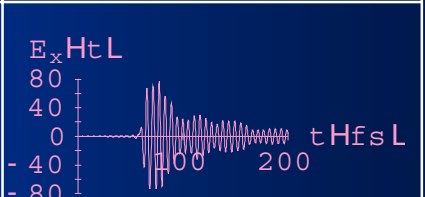
Linear responses

No Chirp	Negative Chirp	Positive Chirp	Transform-Limited
 Exciting Pulse			
 Local Field at Opening			
 Local Field at Tip	 Best energy concentration		

Same spectrum

Same envelope

Same average period

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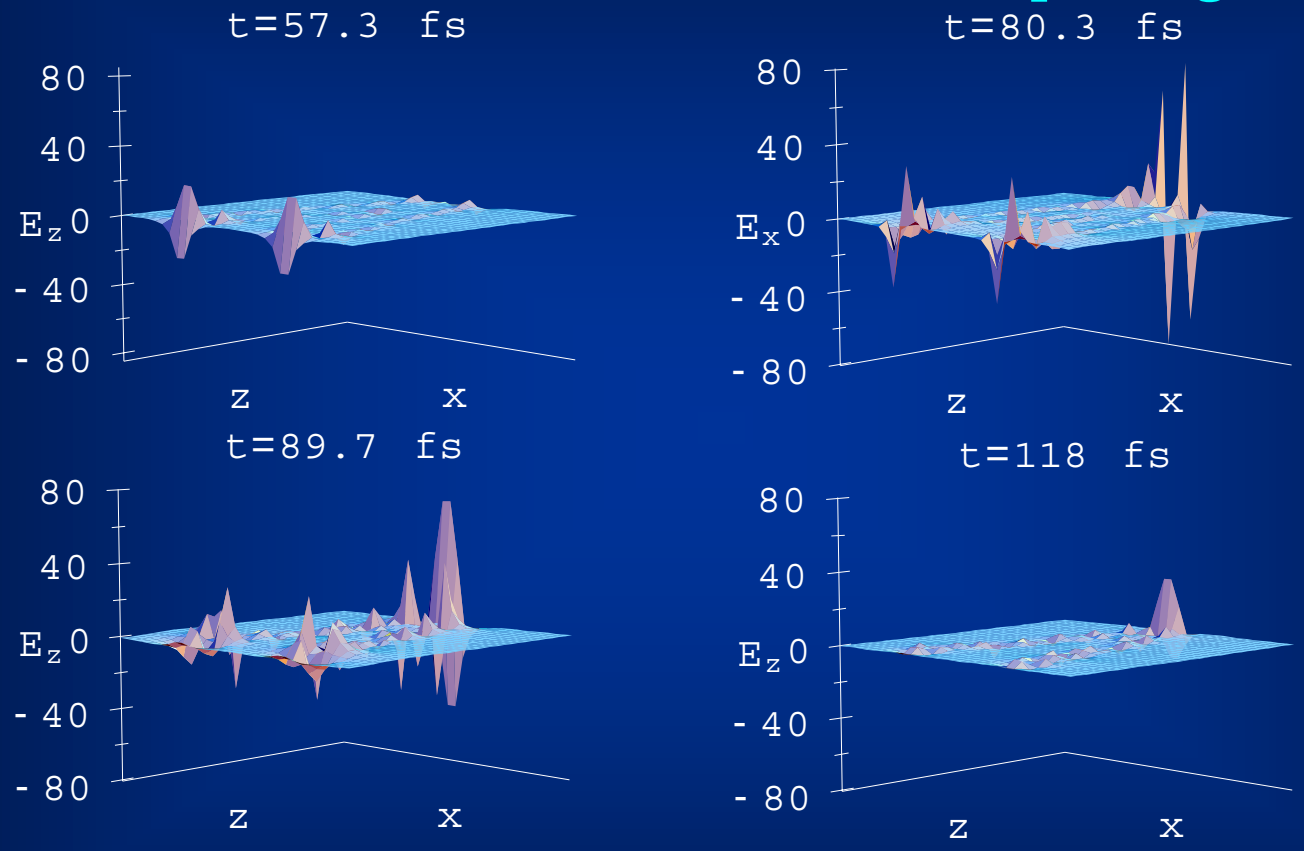
Spatial Distribution: Local Fields in V-shape, Negative Chirp



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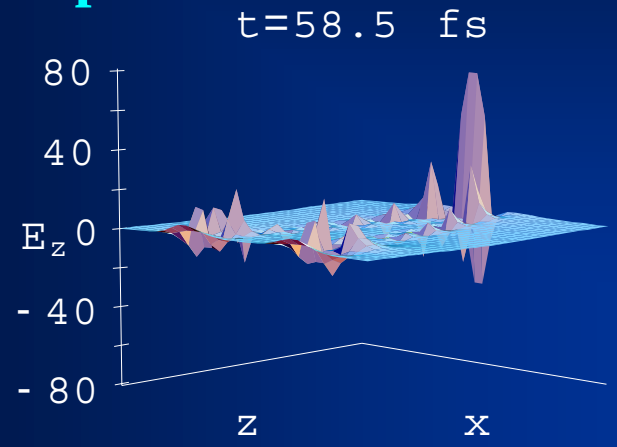
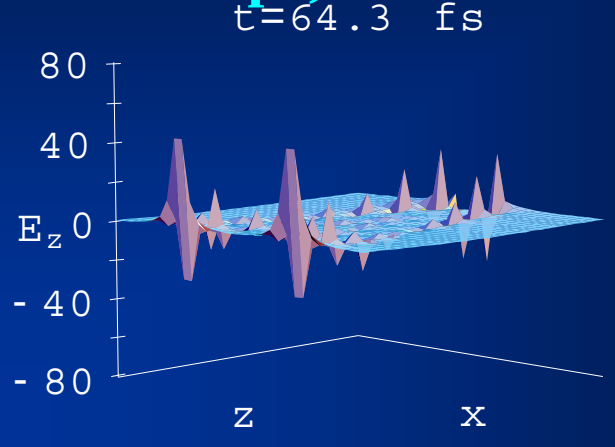
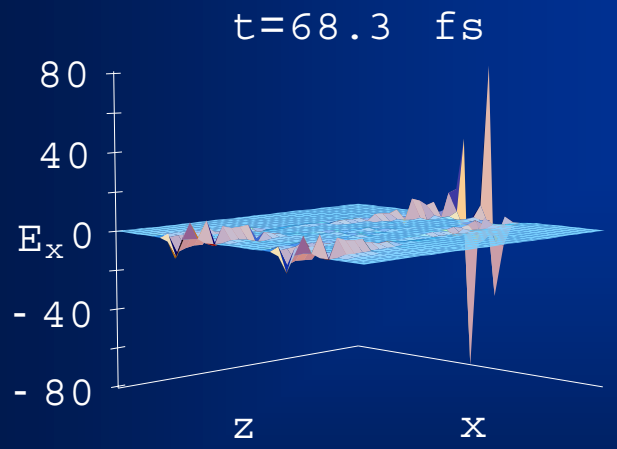
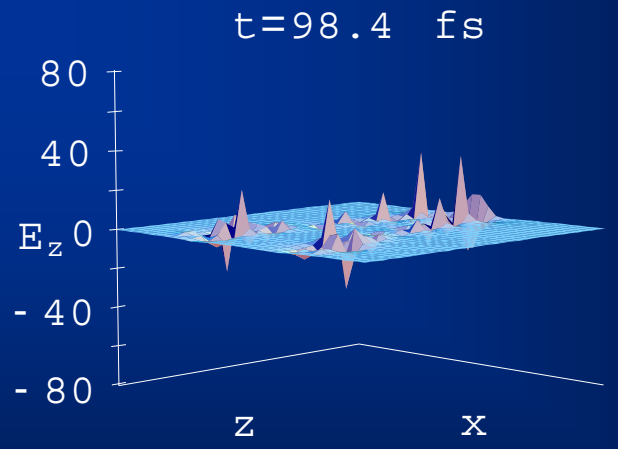
Spatial Distribution: Local Fields in V-shape, Positive Chirp









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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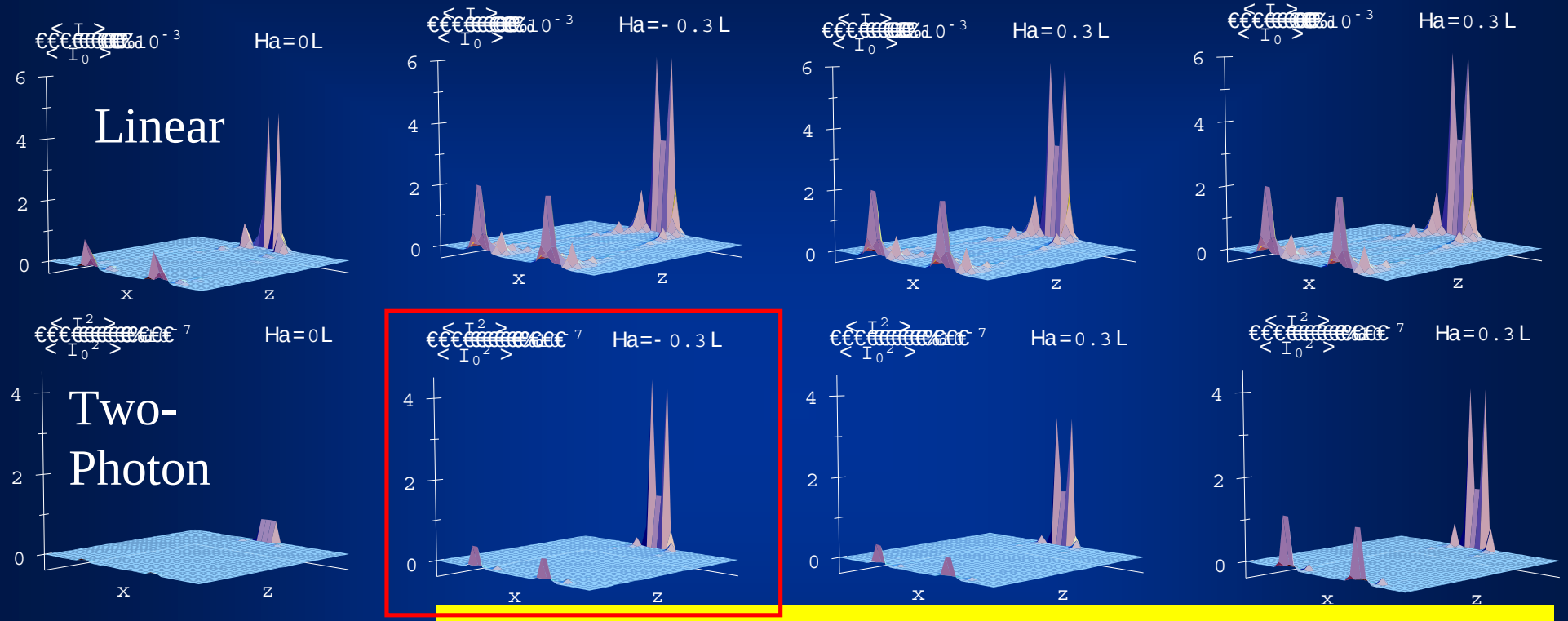
Time-Averaged Responses



Same spectrum

Same envelope

Same average period

Conclusion: For averaged linear responses, only spectrum is important.
In a nonlinear case, the phase is a controlling factor.

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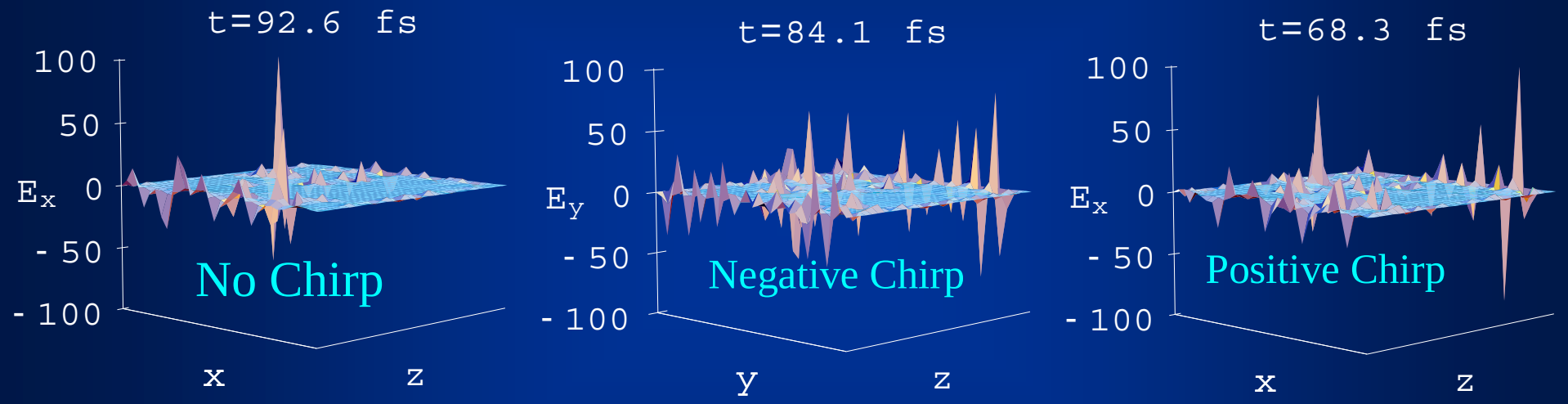
Local Optical Fields in Random Planar Composite at the Instants of their Maxima



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

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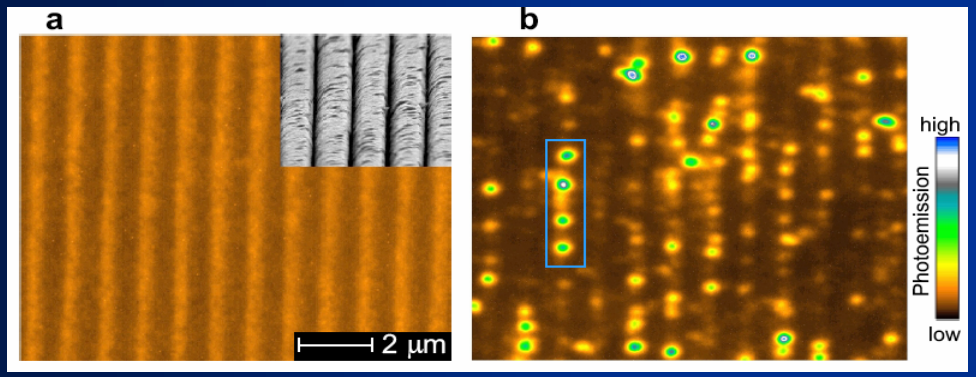
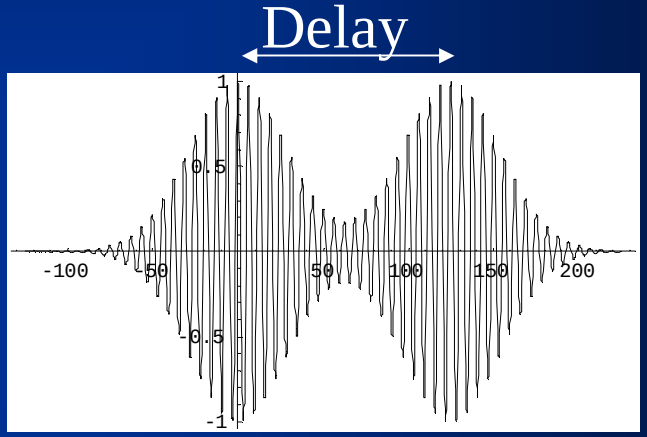
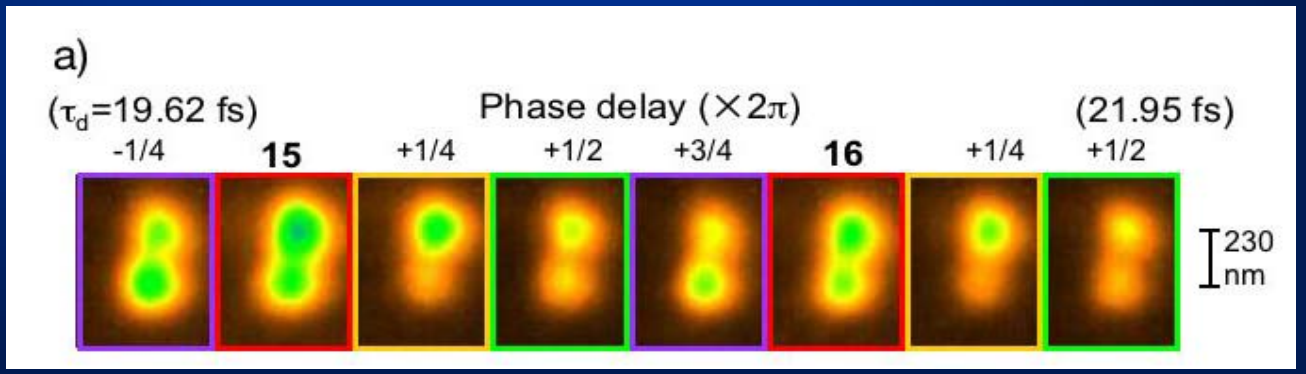
Two-Photon Interferometric Coherent Control

A. Kubo, K. Onda, H. Petek, Z. Sun, Y. S. Jung, and H. K. Kim, in *Attosecond Kinematic Micrography of Surface Plasmon Dynamics*, Nature (2004 (submitted)).







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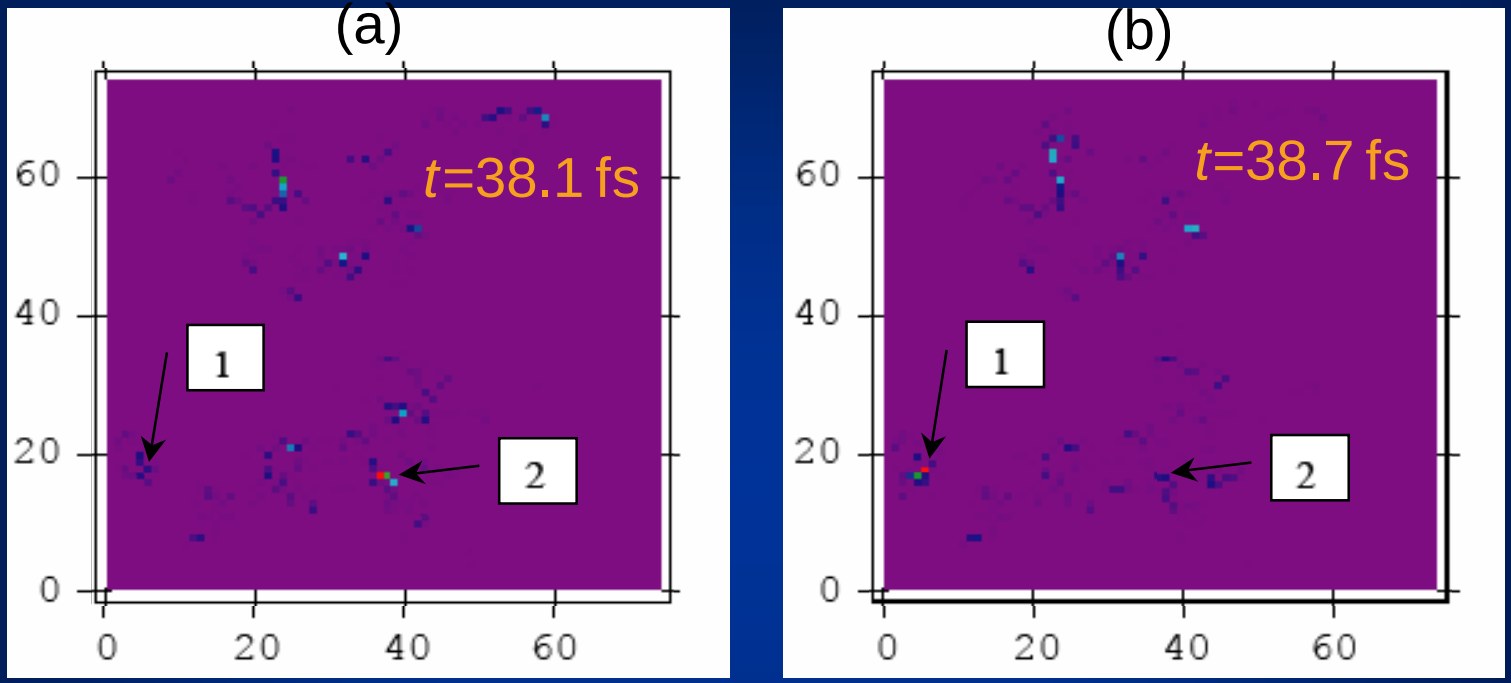

Local optical field intensity for a fractal cluster of gold for a 50 fs ultrashort exciting pulse (carrier wavelength of 1240 nm). Panels (a) and (b) are snapshots for observation times indicated in the figures, separated by 600 as. The data are adapted from M. I. Stockman, *Giant Attosecond Fluctuations of Local Optical Fields in Disordered Nanostructured Media*, Phys. Rev. B **62**, 10494-10497 (2000).

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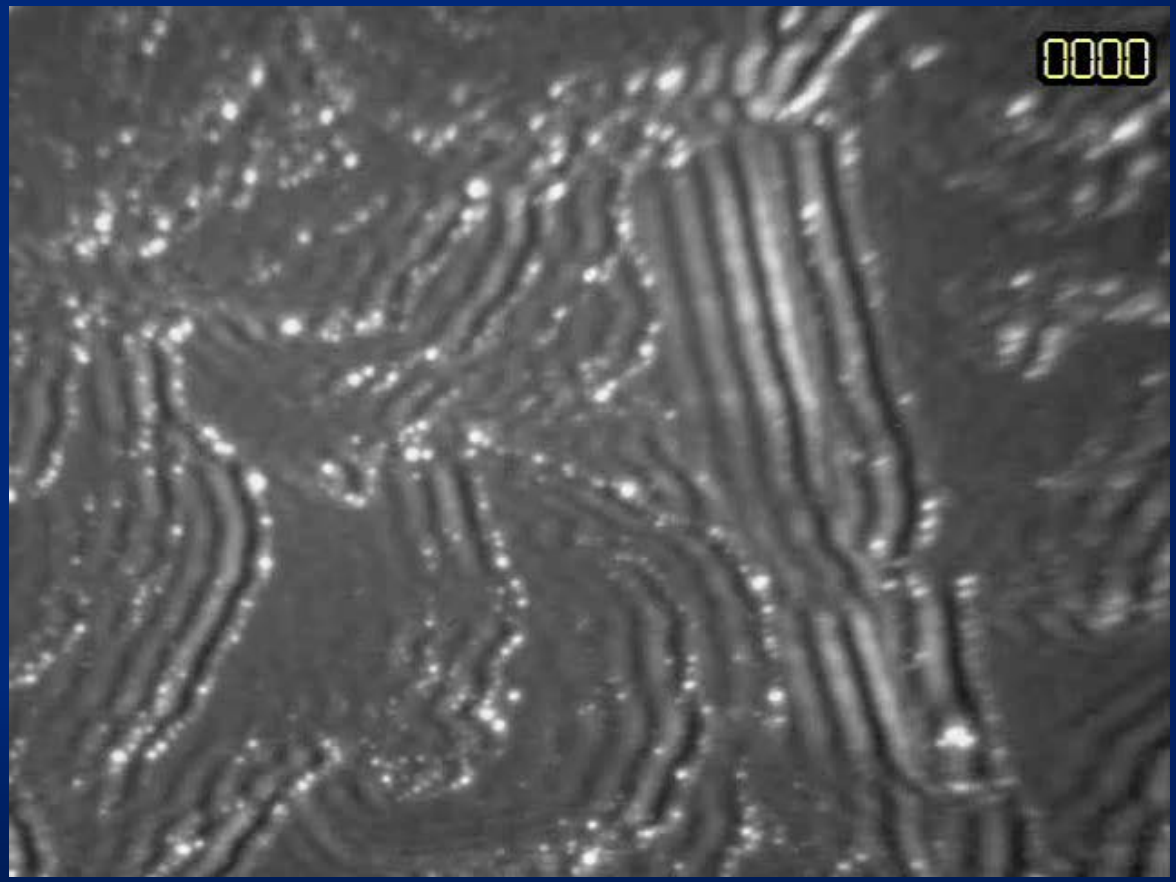
Experiment: Distribution of the two-photon electron emission from rough silver surface. Frame are taken with 200 as periodicity in delay



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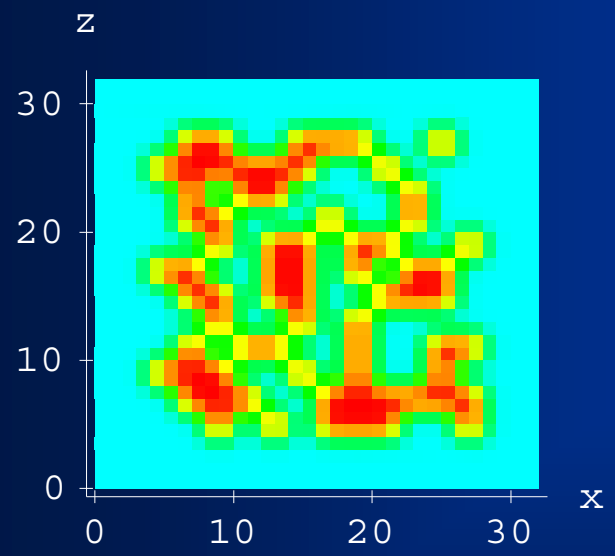
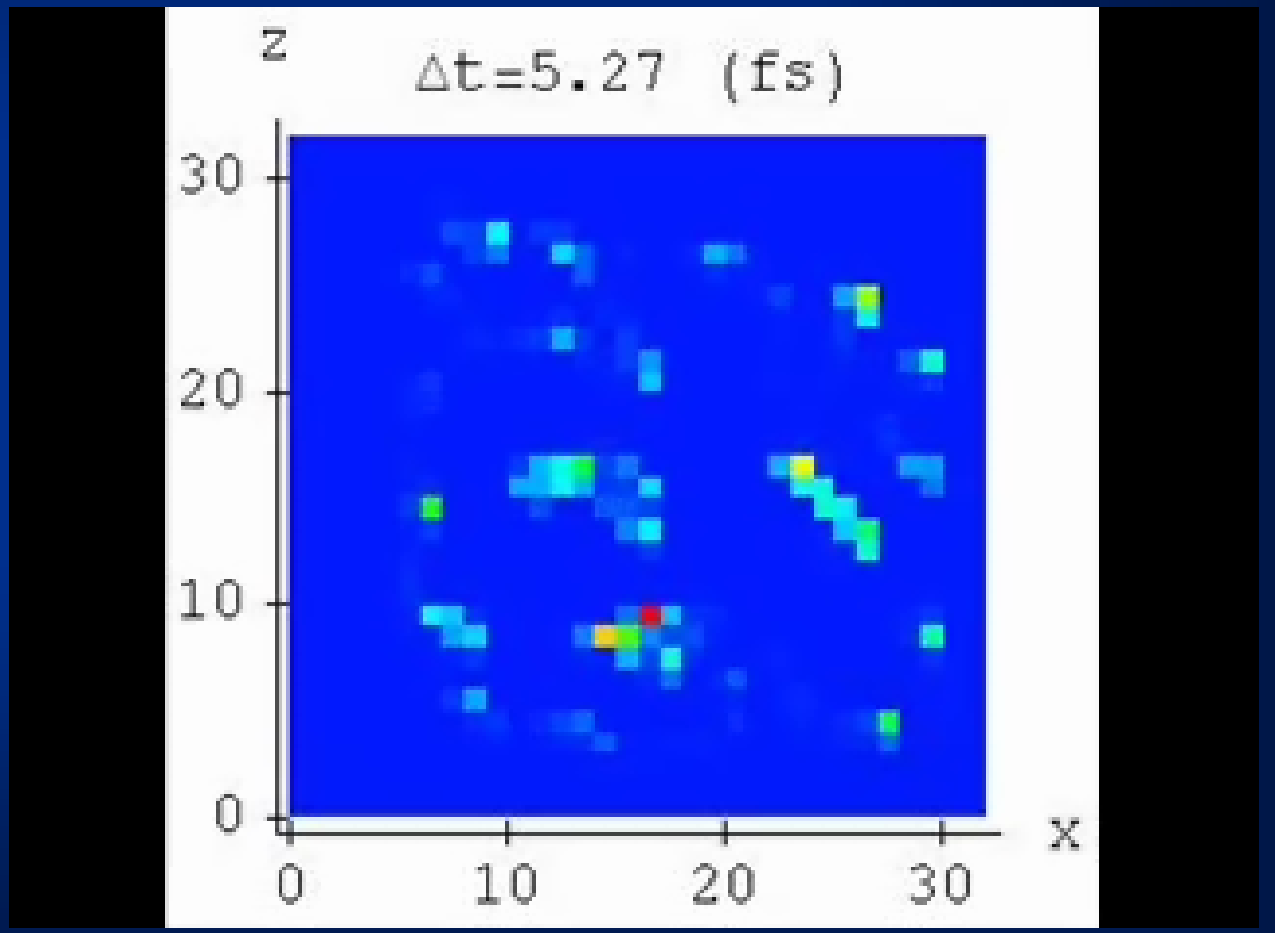
Theory: spatial distributions of two-photon excitation as a function of delay between the two excitation pulses



Geometry of the system



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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 10^7.

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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).

Acknowledgement: Victor I. Klimov (LANL) and John E. Sipe (U. Toronto)

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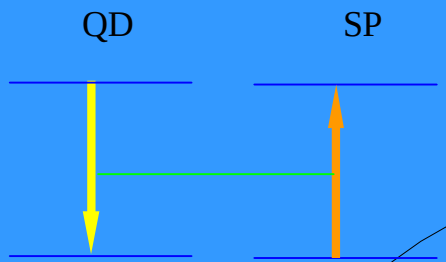
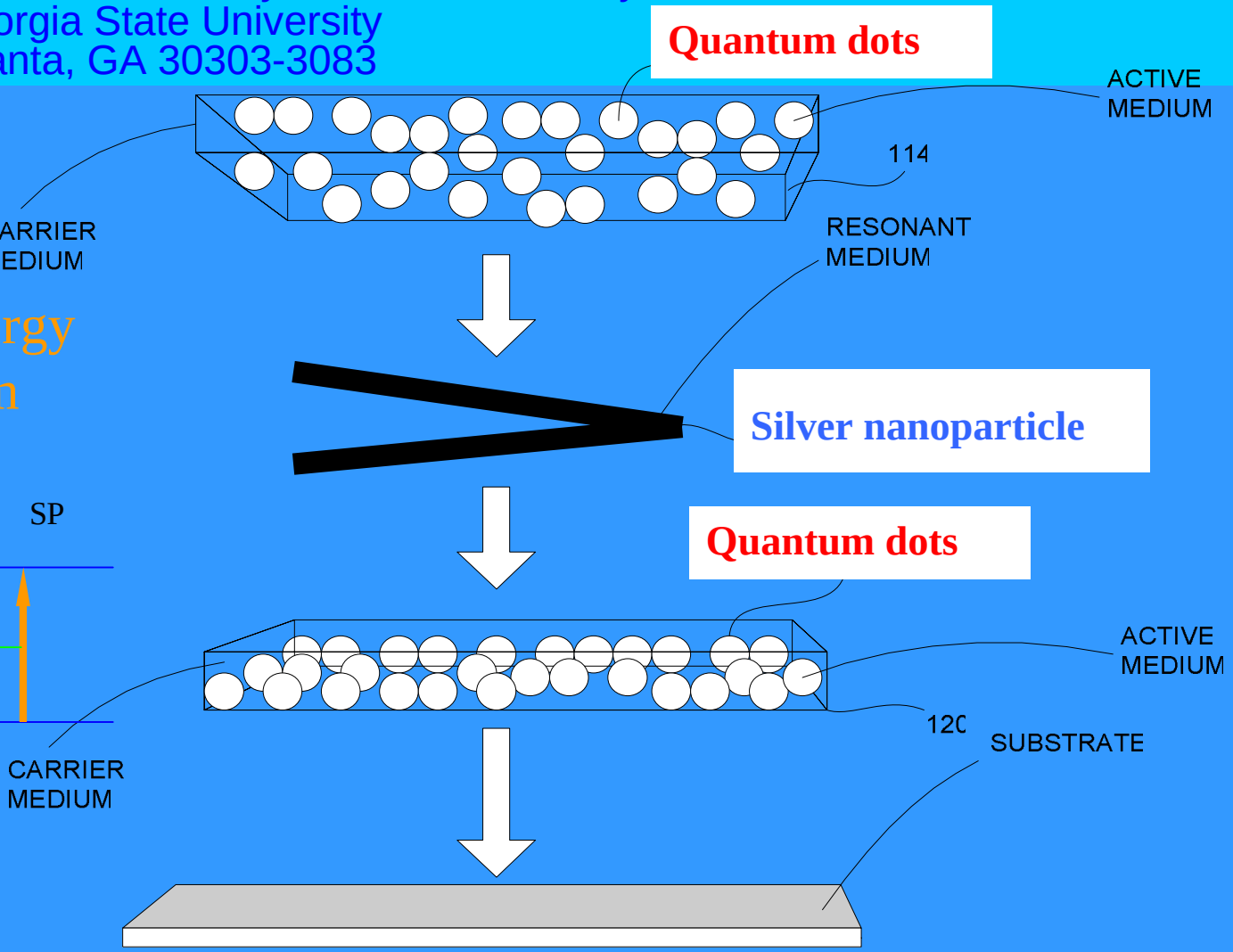
Foerster energy transfer from QD to SP's





SPASER Schematic

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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

$$\left.\frac{\partial}{\partial x}\varphi_n(x, y, z)\right|_{x=0,L_x} = \left.\frac{\partial}{\partial y}\varphi_n(x, y, z)\right|_{y=0,L_y} = 0.$$

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

Frequency ω_n and decay rate γ_n of surface plasmons:

$$\mathrm{Re}\left[s(\omega_n)\right] = s_n \ , \ \gamma_n = \frac{\mathrm{Im}[\, s(\omega_n)]}{s_n'}, \quad \text{where} \quad s_n' \equiv \frac{d\ \mathrm{Re}\left[s(\omega_n)\right]}{d\omega_n}$$

Quasielectrostatic Hamiltonian of an inhomogeneous dispersive nanosystem:

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

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

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Quantized potential operator as an expansion over surface plasmons:

$$\hat{\phi}(\mathbf{r},t)=\sum_n\sqrt{\frac{2\pi\hbar s_n}{\varepsilon_h}}\,\varphi_n(\mathbf{r})e^{-\gamma_n t}\left[a_n e^{-i\omega_n t}+a_n^{+}e^{i\omega_n t}\right]$$

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:

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

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The Einstein stimulated emission coefficient is

$$A_n = \frac{4\pi}{3\hbar}\frac{s_n\left|\mathbf{d}_{10}\right|^2 p_n q_n}{\varepsilon_h s_n' \gamma_n}$$

Here p_n is the spatial overlap factor and q_n is the spectral overlap factor between the eigenmode intensity and the population inversion,

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

Spaser gain $\alpha_n = \dfrac{A_n-\gamma_n}{\gamma_n}$

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

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

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

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

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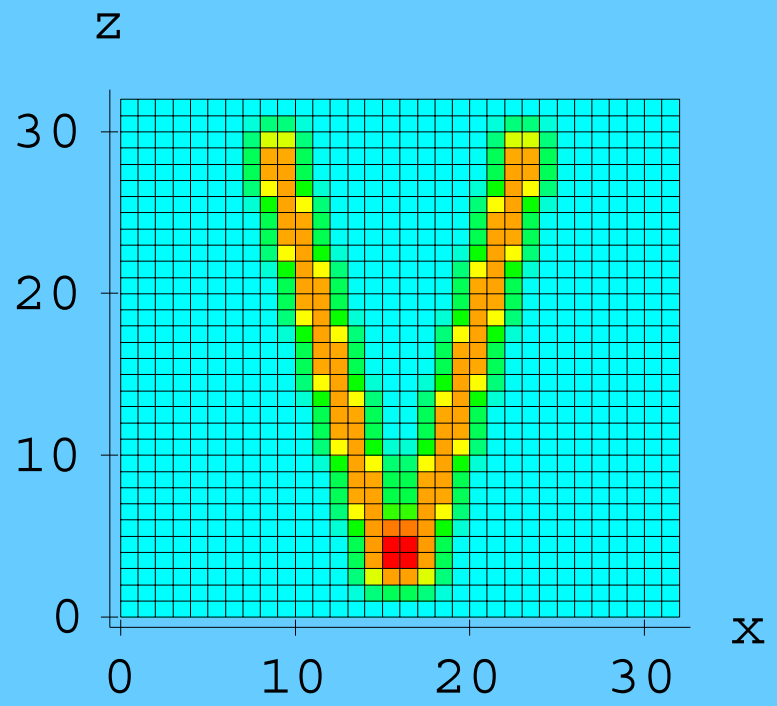
RESULTS

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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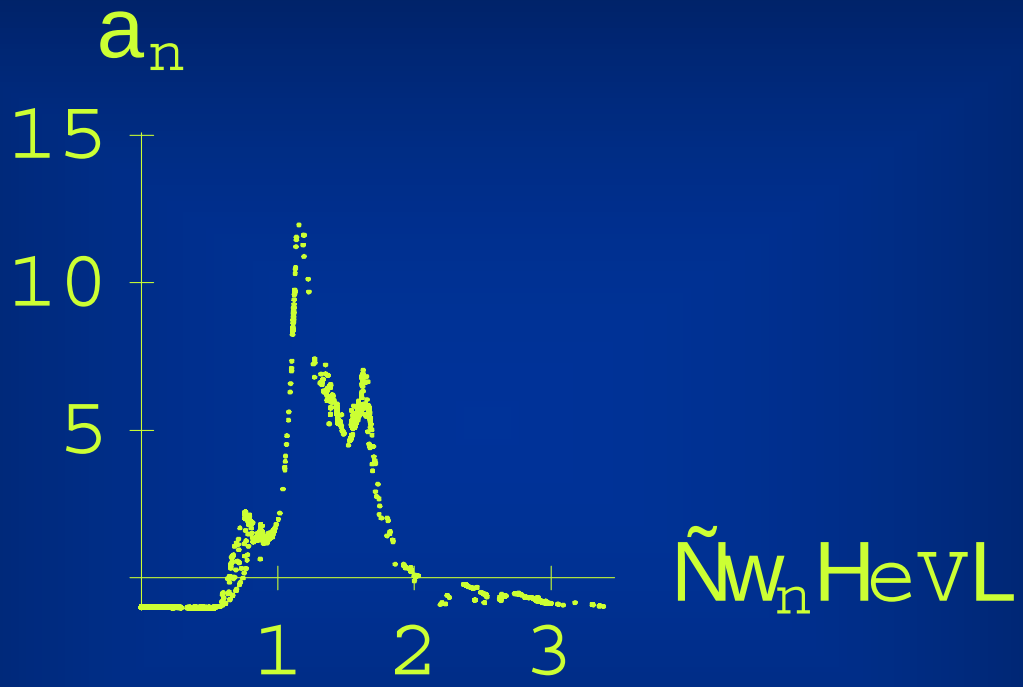

Calculated gain for thin (three monolayers of quantum dots) active medium

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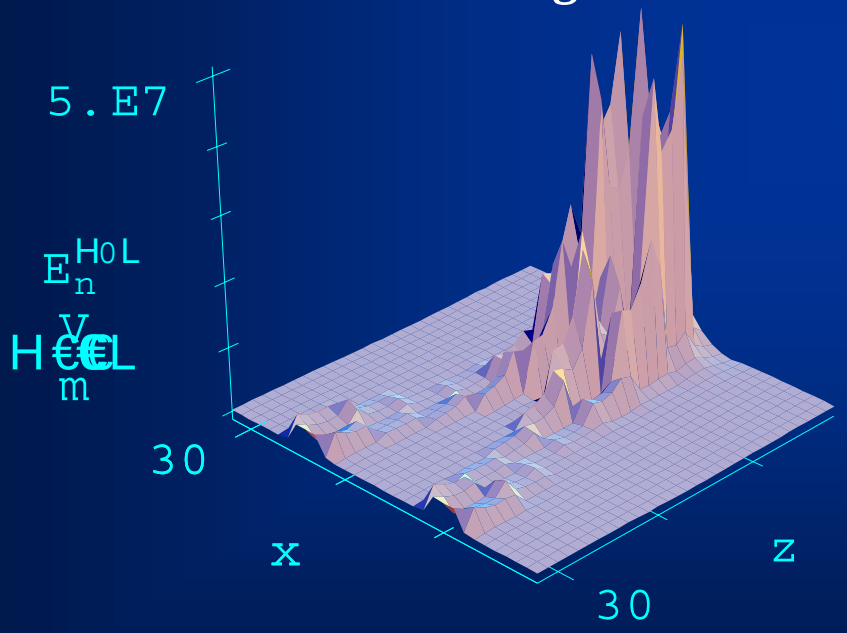
Eigenmodes with highest yields for the spectral maximum at 1.2 eV

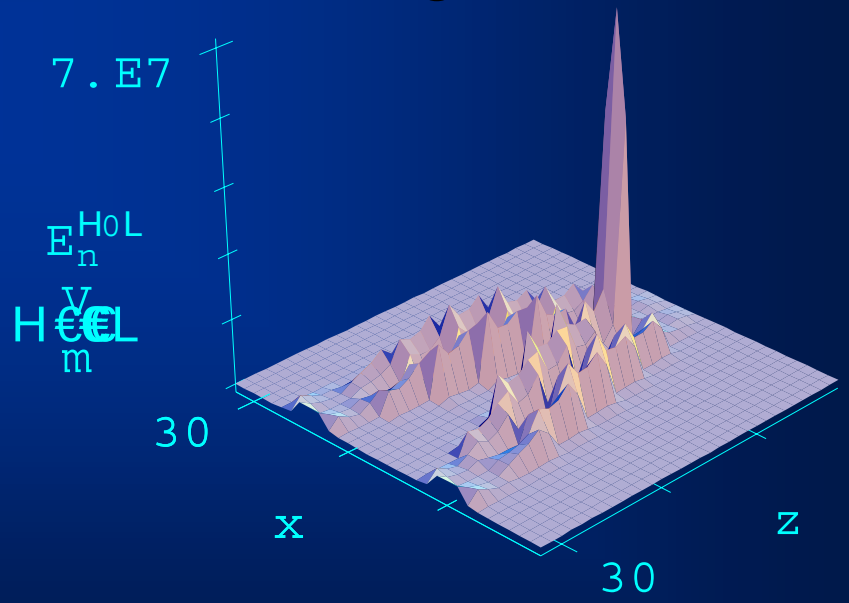
$\hbar\omega_n = 1.15$ eV
$a_n = 12.,\ f_n = 5.E-3$

Luminous eigenmode



$\hbar\omega_n = 1.18$ eV
$a_n = 11.,\ f_n = 3.E-12$

Dark eigenmode



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CONCLUSIONS

- We have proposed the SPASER: effect and prospective quantum-nanoplasmonic device.
- Spaser is not a laser: it does not emit light waves but builds up macroscopic numbers of coherent SPs in individual eigenmodes of a nanosystem.
- It is possible to generate dark SPs that do not couple to far-zone fields.
- Spaser generates intense, ultrafast, nanoscale-localized optical-frequency local fields.
- There exist many possibilities for prospective applications in nanoscience and nanotechnology