Nanoplasmonics:
Optical Properties of Plasmonic Nanosystems

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Lecture 3:
Ultrafast, Nonlinear, and Quantum Nanoplasmonics
LECTURE 3

Ultrafast, Nonlinear, and Quantum Nanoplasmonics

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

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PROBLEMS IN NANOOPTICS

Microscale

Delivery of energy to nanoscale: Conversion of propagating EM wave to local fields

Enhancement and control of local nanoscale fields. Enhanced near-field responses

Ultrafast, nonlinear, and quantum nanoplasmonics (SPASER)
Nanoplasmonics is intrinsically ultrafast:

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

\[ \Delta \omega \sim \omega_p \sim 10 \text{ eV} \]

Corresponding shortest time of plasmonic responses

\[ \sim 100 \text{ as} \]

Surface plasmon relaxation times are in \(~10-100 \text{ fs range}\).
COHERENT CONTROL ON NANOSCALE

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

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

However, it does possess spectral (phase, or temporal) degrees of freedom and polarization. These can be used to *dynamically* control the optical energy localization at the nanoscale
Different spectral components of the excitation pulse excite resonant surface plasmon modes.

These excitations dynamically interfere creating time-dependent hot spots of local fields during their coherence time.

This interference can be directed by choosing phases and amplitudes of the different frequency components of the excitation pulse (pulse shaping).
Principles of coherent (quantum) control


REFERENCES ON COHERENT CONTROL OF OPTICAL ENERGY NANOLOCALIZATION


• Femtosecond local fields on nanoscale:

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

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

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

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

No Chirp  |  Negative Chirp  |  Positive Chirp  |  Transform-Limited

$E_z(t)$ Exciting Pulse

$E_z(t)$ Local Field at Opening

$E_x(t)$ Local Field at Tip

Same spectrum

Same envelope

Same average period
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.
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.
Time-Averaged Responses

\[ \frac{I}{I_0} \times 10^{-3} \quad \alpha = 0 \]

Linear

\[ (\frac{I}{I_0})^2 \times 10^{-7} \quad \alpha = 0 \]

Two-Photon

\[ (\frac{I}{I_0})^2 \times 10^{-7} \quad \alpha = -0.3 \]

\[ (\frac{I}{I_0})^2 \times 10^{-7} \quad \alpha = 0.3 \]

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.
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
Femtosecond Imaging of Surface Plasmon Dynamics in a Nanostructured Silver Film

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ABSTRACT

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

Delay

Phase delay (× 2π)
(-1/4) 15 (+1/4) (+1/2) (+3/4) 16 (+1/4) (+1/2)

(τ_d=19.62 fs) (21.95 fs)

230 nm

Experimental Observation

Two-Photon Interferometric Coherent Control

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

\[ Dt = 009.75 \times 2\pi \]

**200 nm**

30 femtoseconds from life of a nanoplasmonic systems

Localized SP hot spots are deeply subwavelength as seen in PEEM (photoemission electron microscope)
Due to the low photoelectron energy and its large spread, there are large chromatic aberrations in the electron optics of the PEEM.
Theory: Spatial distributions of two-photon excitation as a function of delay between the two excitation pulses (Movie)

Geometry of the system
Coherent Control of Two-Photon Electron Emission in V-Shape Nanoantennas

Time-integrated electron current as a function of interpulse delay
Adaptive subwavelength control of nano-optical fields

Martin Aeschlimann¹, Michael Bauer², Daniela Bayer¹, Tobias Brixner³, F. Javier García de Abajo⁴, Walter Pfeiffer⁵, Martin Rohmer¹, Christian Spindler³ & Felix Steeb¹
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 $10^7$. 
“Synthetic Aperture Radar” in Nanoplasmonics

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

Planar array of a phased-array antenna

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

\[
\phi = \text{phase shift between two successive elements}
\]
\[
d = \text{distance between the radiating elements}
\]
\[
\Theta = \text{beam steering}
\]

\[
\chi = d \cdot \sin \Theta
\]

(Sinussatz)

\[
\frac{360^\circ}{\phi} = \frac{\lambda}{\chi}
\]

(Dreisatz)

\[
\phi = \frac{360^\circ}{\lambda} \cdot d \cdot \sin \Theta
\]

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APAR: Active Phased Array Radar

AESA: Active Electronically Scanned Array

(a) Straight beam
(b) Steering
(c) Focusing
(d) Steering and focusing
An interference fringe of two coherent EM sources is a line of

\[ R_1 - R_2 = m\lambda, \quad m = 0, \pm 1, \ldots \]

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


Wave front

Polariton rays

(a) $I(\text{arb. u.})$

$\hbar \omega = 2.5 \text{ eV}$

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

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

Solution: Time reversal approach

Adaptive subwavelength control of nano-optical fields

Martin Aeschlimann¹, Michael Bauer², Daniela Bayer¹, Tobias Brixner³, F. Javier García de Abajo⁴, Walter Pfeiffer⁵, Martin Rohmer¹, Christian Spindler³ & Felix Steeb¹
Focusing Beyond the Diffraction Limit with Far-Field Time Reversal

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

1120 23 FEBRUARY 2007 VOL 315 SCIENCE

A

B

C

8-antenna TRM

~10λ

λ/30

1-8 channel micro-structured array

1-m² reverberation chamber

copper wires

insulating layer

metallic ground

coaxial line

3 4

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Fig. 2. Focusing beyond the diffraction limit. (A and B) show the signal received at one antenna of the TRM when a 10-ns pulse is sent from antennas 3 and 4, respectively, of the microstructured array. The signals in (A) and (B) look considerably different, although antennas 3 and 4 are only $\lambda/30$ apart. (C and D) show the time compression obtained at antennas 3 and 4, respectively, when the eight signals coming from antennas 3 and 4 are time-reversed and sent back from the TRM. (E) In full line are shown the focusing spots obtained around antennas 3 and 4. Their typical width is $\lambda/30$. Thus, antennas 3 and 4 can be addressed independently. The focal spots obtained when there are no copper wires are shown for comparison (dashed-dotted line). All maxima have been normalized by scaling factors in the ratios: 1 (red and blue dashed-dotted lines), 2.2 (red full line), 2.5 (blue full line).
Nanoplasmonic Energy Localization, Time Reversal, and Coherent Control


Idea of time reversal for subwavelength EM-wave localization:


Time-averaged two-photon excitation

\[ <I^2> \]

Time-dependent local field intensity at the target points

\[ I \]

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Resonant Metalenses for Breaking the Diffraction Barrier

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We introduce the resonant metalens, a cluster of coupled subwavelength resonators. Dispersion allows the conversion of subwavelength wave fields into temporal signatures while the Purcell effect permits an efficient radiation of this information in the far field. The study of an array of resonant wires using microwaves provides a physical understanding of the underlying mechanism. We experimentally demonstrate imaging and focusing from the far field with resolutions far below the diffraction limit. This concept is realizable at any frequency where subwavelength resonators can be designed.

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

(d) Focal spot obtained after far field time reversal
Schematic of proposed local excitation with adiabatic cones


Active spatial control of plasmonic fields

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Femtosecond Nanofocusing with Full Optical Waveform Control

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CONCLUSIONS

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

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

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

• The optical field nanolocalization is coherently controllable by pulse shaping.

• Time reversal gives a convenient and powerful tool to determine the pulse shaping.
Attosecond nanoplasmonic-field microscope

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PEEM Electron Optics Imaging with Energy Resolution

Schematic of Attosecond Nanoplasmonic Field Microscope

XUV photoelectrons accelerated by enhanced IR plasmonic local fields

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

Escape (dwelling) time from the local field region: \( \tau_d \)

Oscillation period: \( T \)

Local field-pulse duration: \( \tau_p \)

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

Instantaneous regime: \( \tau_d \ll T \) \( \tau_d = 180 \text{ as} \), \( \hbar \omega_{\text{XUV}} = 95 \text{ eV} \)

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

In this case the XUV-electron energy is defined solely by the local, instantaneous electrostatic potential.
Novel Regime of Electron Emission from Nanoplasmonic Systems

The time of flight through the region of local fields for ~100 eV XUV electrons is ~300 as and much less than plasmonic near-IR period. Consequently, electrons are electrostatically accelerated by the instantaneous local field electric potential.

Escape (dwelling) time of electron from the local field region: \( \tau_d \)

Optical oscillation period: \( T \)

Local field-pulse duration: \( \tau_p \)    XUV photon energy: \( \hbar \omega_X \)

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

Instantaneous regime: \( \tau_d << T \)\[ \tau_d = 180 \text{ as}, \quad \hbar \omega_X = 95 \text{ eV} \]

The XUV photoelectron energy \( E_e \):

\[ E_e = \hbar \omega_X - W_f + e\phi(r, t_X) \]

In this case the XUV-electron energy is defined solely by the local, instantaneous electrostatic potential.
Uncertainty Principle and Real-Time Measurement of Local Fields

For any practical purposes, from $E_e = \hbar \omega_X - W_f + e\phi(r, t_X)$
it follows that uncertainty of the local potential $\Delta \phi = -\frac{\hbar}{e} \Delta \omega_X$
where $\Delta \omega_X$ is the spectral bandwidth of the XUV pulse.

The Heisenberg uncertainty relation is $\hbar \Delta \omega_X \Delta t_X \geq \frac{\hbar}{2}$
The precision of the instance of measurement is limited by $\Delta t = \Delta t_X$

Thus, the fundamental limitation on the measurement of $\phi(r, t)$ is

$$\Delta \phi \cdot \Delta t \geq \frac{\hbar}{2e} \quad \text{and} \quad \Delta \phi = -\frac{\hbar}{e} \Delta \omega_X$$

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

$t = 64.13 \, \text{fs}$

$X = 14 \, \text{fs}$

Local optical electric field at the “hottest spot”

$t_X = 14 \, \text{fs}$

Attosecond pulse applied within a period of IR oscillations
Energy shift (eV) of electrons emitted by a 95 eV XUV attosecond pulse as a function of the as pulse delay with respect to the infrared-visible excitation pulse (frames are in 200 as) as observed in Photoemission Electron Microscope (PEEM).

**Experiment directly measures instantaneous electric potential of nanoplasmonic oscillations with nm spatial and ~200 as temporal resolution**

Nanosystem is **60x60 nm** random silver film (50% filling factor)
High-harmonic generation by resonant plasmon field enhancement

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

Plasmonic generation of ultrashort extreme-ultraviolet light pulses

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

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


Spaser is the ultimately smallest quantum nano-generator

For small nanoparticles, radiative loss is negligible.

Spaser is fully scalable

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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 $\varphi_n$ are eigenfunctions:

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

where $\theta(r \in \text{metal}) = 1$ and $\theta(r \in \text{elsewhere}) = 0$;

$\varphi_n(x, y, 0) = \varphi_n(x, y, L_z) = 0$, and

$$\frac{\partial}{\partial x} \varphi_n(x, y, z) \bigg|_{x=0, L_x} = \frac{\partial}{\partial y} \varphi_n(x, y, z) \bigg|_{y=0, L_y} = 0.$$
Spectral parameter: \[ s(\omega) = \left[ 1 - \varepsilon(\omega) / \varepsilon_h \right]^{-1} \]

Frequency \( \omega_n \) and decay rate \( \gamma_n \) of surface plasmons:

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

Quasielectrostatic Hamiltonian of an inhomogeneous dispersive nanosystem:

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

where is \( \mathbf{E}(\mathbf{r}, \omega) = -\nabla \phi(\mathbf{r}, \omega) \) the electric field operator.
Quantized potential operator as an expansion over surface plasmons:

\[ \hat{\phi}(r, t) = \sum_n \sqrt{\frac{4\pi\hbar s_n}{\varepsilon_n}} \varphi_n(r) \left[ a_n e^{-i\omega_n t} + a_n^+ e^{i\omega_n t} \right] \]

where \( a_n^+ \) and \( a_n \) are the surface plasmon creation and annihilation operators. With this, the Hamiltonian becomes

\[ H = \sum_n \hbar \omega_n \left( a_n^+ a_n + \frac{1}{2} \right) \]

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

\[ H' = \sum_a d^{(a)} \nabla \hat{\phi}(r_a, t) \]

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

\[ \frac{dN_n}{dt} = \left( B_n - \gamma_n \right) N_n + A_n \]
The Einstein stimulated emission coefficient is

\[ B_n = \frac{4\pi}{3\hbar} \frac{s_n |d_{10}|^2 p_n q_n}{\varepsilon_h s'_n \gamma_n} > \gamma_n \Rightarrow \frac{|d_{10}|^2 N_{QD} Q}{\hbar R^3 \Gamma} \geq 1 \]

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

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

Spaser gain

\[ \alpha_n = \frac{B - \gamma_n}{\gamma_n} \]

shows how many times faster the surface plasmons are born by the stimulated emission than they decay.
The local RMS field produced by spaser: 
\[ E(r) = \left( \left[ \nabla \phi(r) \right]^2 \right)^{1/2} \]
is calculated as:
\[ E(r) = E_n(r) \left( N_n + \frac{1}{2} \right)^{1/2} \]
where
\[ E_n(r) = \left\{ \frac{4\pi\hbar s_n}{\epsilon_h s'_n} \left[ \nabla \phi_n(r) \right]^2 \right\}^{1/2} \]
RESULTS

The resonant nanoparticle is an “engineered” V-shape. The material is silver; the spatial scale is 2-5 nm/grid unit.
Calculated gain for thin (three monolayers of quantum dots) active medium.
Eigenmodes with highest yields for the spectral maximum at 1.2 eV

\( \hbar \omega = 1.15 \text{ eV} \)
\( \alpha = 12, \ f = 0.005 \)

\( \hbar \omega = 1.18 \text{ eV} \)
\( \alpha = 11, \ f = 10^{-12} \)

Bright mode

Dark mode
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],

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

The electric field operator\(^4\) of the quantized SPs is\(^4\)

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

$s(\omega) = \varepsilon_d/\sqrt{\varepsilon_d - \varepsilon_m(\omega)}$ is Bergman’s spectral parameter, $\varepsilon_d$ is the permittivity of the ambient dielectric, and $\varepsilon_m(\omega)$ is the metal permittivity.

The spaser Hamiltonian has the form

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

where $H_g$ is the Hamiltonian of the gain medium.

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

$$\dot{\rho}_{12}^{(p)} = -[i (\omega - \omega_{12}) + \Gamma_{12}] \rho_{12}^{(p)} + i n_{21}^{(p)} \Omega_{12}^{(a)*} , \quad (4)$$

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

$$\dot{n}_{21}^{(p)} = -4 \text{Im} \left[ \rho_{12}^{(p)} \Omega_{21}^{(p)} \right] - \gamma_2 \left( 1 + n_{21}^{(p)} \right) + \gamma \left( 1 - n_{21}^{(p)} \right)$$

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

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

$$\gamma_s = \frac{\sum_p \left( 1 + n_{21}^{(p)} \right) \gamma^{(p)}_2}{2(2N_p + 1)} . \quad (8)$$

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

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

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

where \( \omega_s \) is the spasing frequency, \( \tilde{\Omega}_{12}^{(p)} = -A_n d_{12}^{(p)} \nabla \varphi_n(r_p)/\hbar \) is the single-plasmon Rabi frequency, \( d_{12}^{(p)} \) is the transition dipole moment of a \( p \)-th chromophore, \( \varphi_n(r_p) \) is the electric potential of the spasing mode at the position this chromophore, \( \gamma_n \)

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

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

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

\[ \omega_s = (\gamma_n \omega_{21} + \Gamma_{12} \omega_n) / (\gamma_n + \Gamma_{12}), \]

which generally does not coincide with either the gain transition frequency \( \omega_{21} \) or the SP frequency \( \omega_n \), 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

\[ \frac{(\gamma_n + \Gamma_{12})^2}{\gamma_n \Gamma_{12} \left[ (\omega_{21} - \omega_n)^2 + (\Gamma_{19} + \gamma_n)^2 \right]} \times \]

\[ \sum_p \left| \tilde{\Omega}_{12}^{(p)} \right|^2 n_{21}^{(p)} = 1, \]

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

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

This system defines the stationary (CW) number of SPs per spasing mode \( N_n \).
SPASER Threshold Condition [Consistent with PRL 90, 027402-1-4 (2003)]:

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

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

(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:

\[
\frac{d_{12}^2 Q N_c}{\hbar \Gamma_{12} V_n} \gtrsim 1,
\]

(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 \( \omega_{21} - \omega_n = 0 \). The spasing is essentially a quantum effect.

It is non-relativistic: does not depend on \( c \)

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

\[
s(\omega) = \frac{\varepsilon_d}{\varepsilon_d - \varepsilon_m(\omega)}; \quad 1 > \text{Re} s(\omega) > 0
\]

Realistic gain for direct band-gap semiconductors.
This quasilinear dependence $N_n(g)$ is a result of the very strong feedback in spaser due to the small modal volume.

Spectral line width $\propto 1 / N_{SP}$
This invention changed civilization as we know it

This invention is used in more copies than all others combined

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

Bandwidth ~ 10-100 GHz
Low resistance to ionizing radiation
The spaser as a Nanoamplifier

Major problem: any quantum amplifier (laser and spaser) in a CW regime possesses exactly zero amplification (it is actually a condition for the CW operation).

We have proposed to set the spaser as a nanoamplifier in two ways:

1. In transient mode (before reaching the CW regime), the spaser still possesses non-zero amplification.

2. With a saturable absorber, the spaser can be bistable. There are two stable states: with the zero coherent SP population ("logical zero") and with a high SP population that saturates the absorber ("logical one" state). Such a spaser will function as a threshold (digital) amplifier.
Scaling of Spaser

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

Heat per flop: \[ H = \hbar \omega N_p \]

Threshold: \[ g \geq g_{th}, \quad g_{th} = \frac{\omega}{c} \frac{\text{Re} s(\omega) \text{Im} \varepsilon_m(\omega)}{1 - \text{Re} s(\omega)}, \quad s(\omega) = \frac{\varepsilon_d}{\varepsilon_d - \varepsilon_m(\omega)} \]

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

Conclusion: Spaser is orders of magnitude more efficient (less heat per flop) and much faster than transistor. It can operate close to the quantum limit.
Amplification in Spaser without a Saturable Absorber

Stationary pumping

Pulse pumping

SP coherent population

Population inversion

SP coherent population

Population inversion
Bandwidth $\sim 10$-100 THz

Very high resistance to ionizing radiation

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

Stationary pumping

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

E. Plum, V. A. Fedotov, P. Kuo, D. P. Tsai, and N. I. Zheludev

Fig. 1. Photonic metamaterial hybridized with semiconductor quantum dots. The insets show the metamaterial unit cell and an SEM image of the actual metamaterial structure.
Demonstration of a spaser-based nanolaser

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**Figure 1** | **Spaser design.** a, Diagram of the hybrid nanoparticle architecture (not to scale), indicating dye molecules throughout the silica shell. b, Transmission electron microscope image of Au core. c, Scanning electron microscope image of Au/silica/dye core–shell nanoparticles. d, Spaser mode (in false colour), with $\lambda = 525$ nm and $Q = 14.8$; the inner and the outer circles represent the 14-nm core and the 44-nm shell, respectively. The field strength colour scheme is shown on the right.

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Figure 2 | Spectroscopic results. Normalized extinction (1), excitation (2), spontaneous emission (3), and stimulated emission (4) spectra of Au/silica/dye nanoparticles. The peak extinction cross-section of the nanoparticles is $1.1 \times 10^{-12} \text{ cm}^2$. 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 \( \lambda = 488 \text{ nm} \). b, Main panel, corresponding input–output curve (lower axis, total launched pumping energy; upper axis, absorbed pumping energy per nanoparticle); for most experimental points, \( \sim 5\% \) 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

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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 798K, 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.
Plasmon lasers at deep subwavelength scale

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

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A room-temperature semiconductor spaser operating near 1.5 μm


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

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


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Operation of electrically-pumped room-temperature telecom-frequency spaser

Fig. 8. (a) Spectra of a DFB cavity taken at a temperature of 296K at various injection currents, blue 2500µA, green 3600µA, red 7600µA. (b) Linewidth of the main peak as a function of injection current (circles). The red line is a fit to a $1/P_{mode}$ dependence.
COMPARISON OF THE SPASER AND NANOLASERS
(POLARITONIC SPASERS)

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

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

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

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

Nanolaser lases at ~10^4 – 10^6 quanta per mode, proportional to its modal volume.

Conventional laser lases at ~10^18 – 10^20 quanta per mode

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

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

Radiation of a spaser can be fed to any waveguide

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

Other possible applications of spaser: local optical excitation of nanosystems, nanomodification (ultrastrong local fields), and active labels for bio-objects and ultramicroscopy.

A nanolaser is a source of optical fields with cross extension on the nanoscale. However the total size of a nanolaser is much greater than a biological molecule such as DNA or protein and their complexes

Radiation of a nanolaser can be fed into most waveguides

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

As a source of optical energy on the nanoscale, a nanolaser has an uphill competition with adiabatic plasmonic concentrators (tapers)
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 excellent qualitative agreement with theory. The observed spaser is single mode. Its pumping curve is linear with a threshold. Its linewidth is inversely proportional to pumping rate.

4. Two plasmon-polariton spasers (nanolasers) have been designed. In contrast to spaser, their length is on micron order (transverse mode size is nanometric). Their emission is multimode.

5. The most promising applications of the SPASER are an ultrafast nanoamplifier, local optical energy source, and active nano-label.
END LECTURE 3