Распад оптического импульса на два плазмона и формирование периодических поверхностных структур

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ПЛАН

01 Введение

Проблема LIPSS: возбуждение плазмонов

02 Возбуждение ППП

Конверсия в пов. плазмон на случайном возмущении ε

03 Обратная связь, распад

Рост возмущения ε, развитие неустойчивости

04 Моделирование

Оценки и предварительные результаты

Introduction 01

The problem of LIPSS: efficiency of SPP excitation

FIRST EXPERIMENTS:

Milton Birnbaum, Semiconductor Surface Damage Produced by Ruby Lasers, 1965

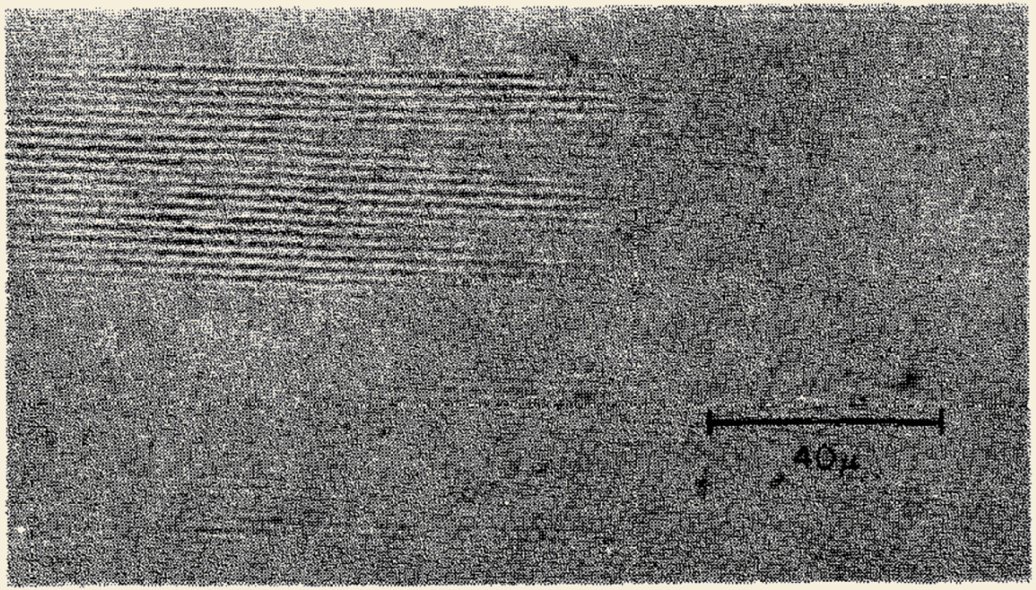
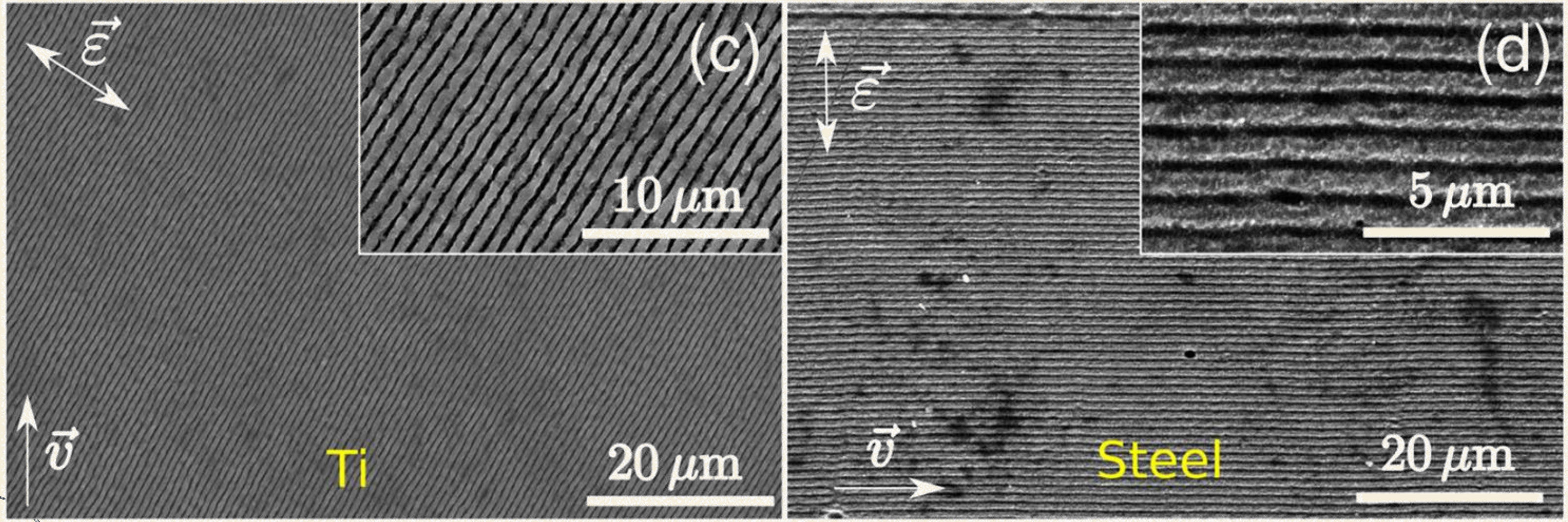


FIG. 1. Photomicrograph of surface damage of a (111) face of a germanium sample.

FEW-SHOT EXPERIMENTS:

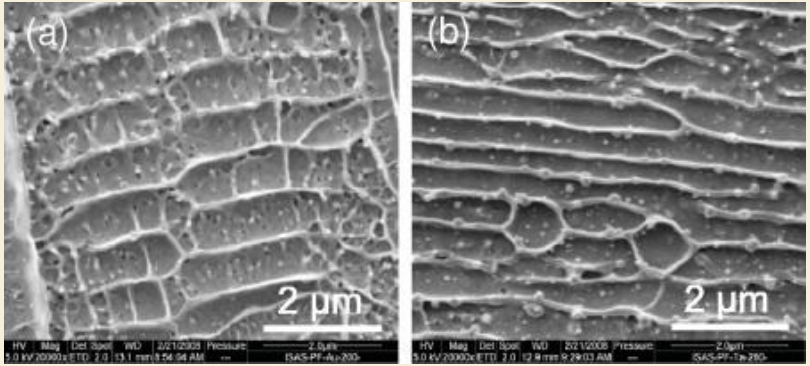
I. Gnilitskyi et al., Scientific reports 7 (1), 1-11 (2017)
scanning; overlap – 2 shots in one point



SINGLE-SHOT EXPERIMENTS:

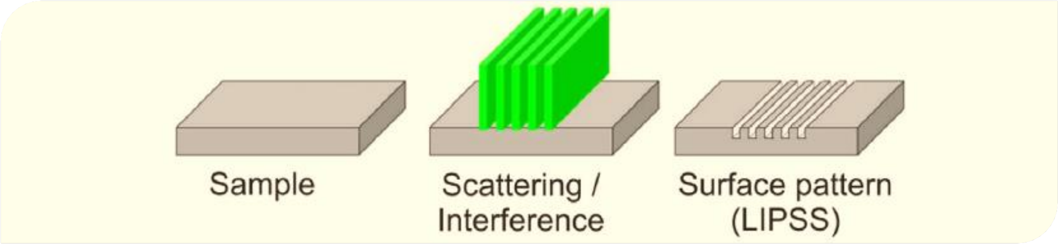
E. L. Gurevich, Self-organized nanopatterns in thin layers of superheated liquid metals, Phys. Rev. E (2011)

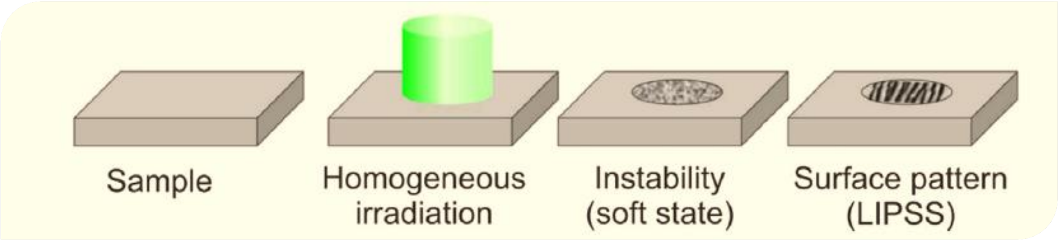
золото (3.3 Дж/см^2) и тантал (4.7 Дж/см^2)



ELECTRODYNAMICS VS. HYDRODYNAMICS

J. Bonse, S. Gräf, Laser Photonics Rev. 2020. Review: Maxwell meets Marangoni

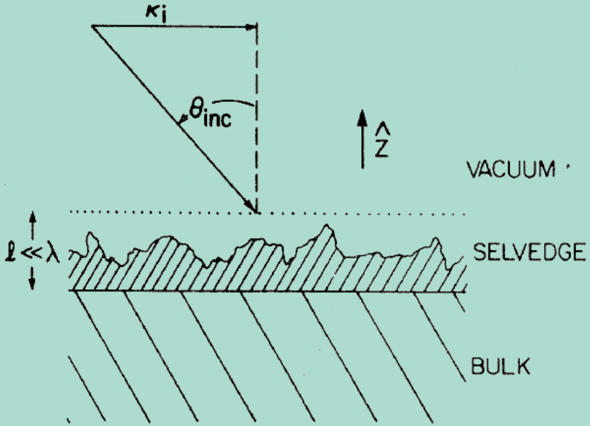




SURFACE PLASMON-POLARITONS

J. E. Sipe, Phys. Rev. B, 1983-1984

Role of surface plasmon-polaritons



SURFACE PLASMON-POLARITONS

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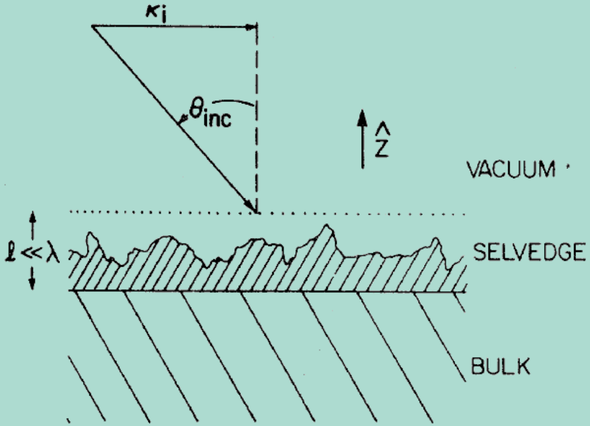
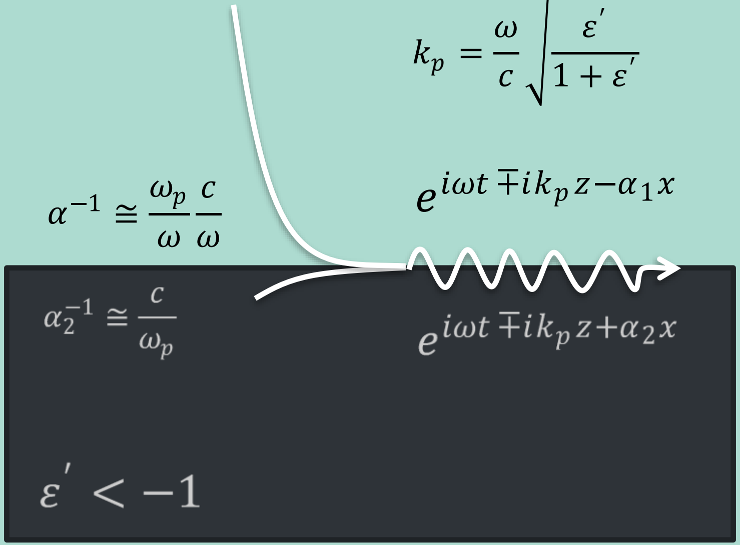


Reminder:

$$k_p = \frac{\omega}{c}\sqrt{\frac{\varepsilon'}{1+\varepsilon'}}$$



$$\alpha^{-1} \cong \frac{\omega_p}{\omega}\frac{c}{\omega}$$

$$e^{i\omega t \mp i k_p z - \alpha_1 x}$$

$$\alpha_2^{-1} \cong \frac{c}{\omega_p}$$

$$e^{i\omega t \mp i k_p z + \alpha_2 x}$$

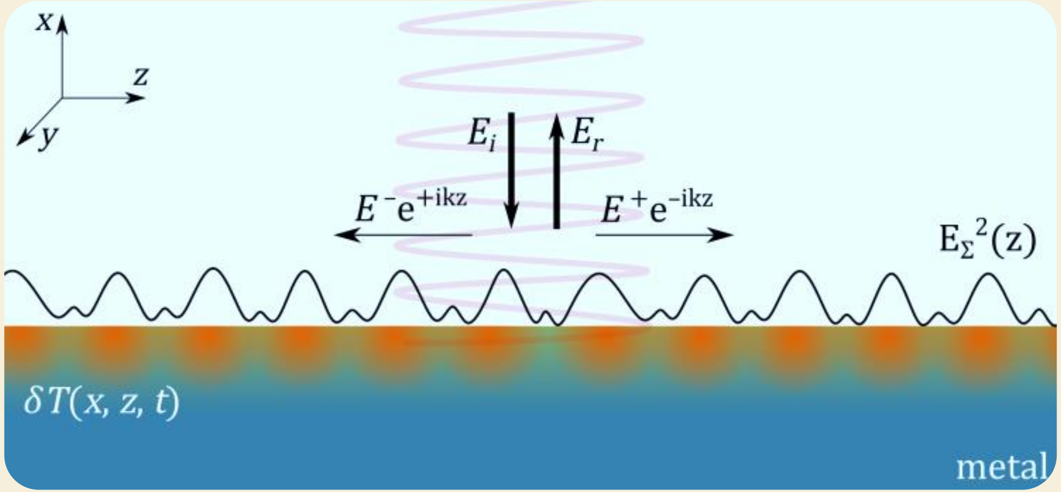
$$\varepsilon' < -1$$

PROBLEM

- Why do periodic structures appears even under single femtosecond pulse action?
- **Is the magnitude of surface plasmons enough** to explain periodic melting?

THE RESULTS BRIEFLY

- There is a **positive feedback** due to thermal nonlinearity. Periodic heating of electrons = periodic perturbation of permittivity
- The time of instable growth can be **as short as 15-20 fs** at damaging intensities
- So, it is short enough to **influence significantly** on SPP magnitudes

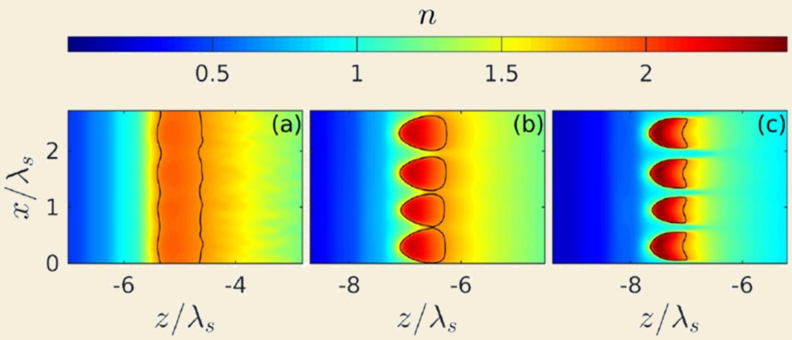


INCIDENT PULSE DECAY

I. Oladyshkin, Self-Induced Decay of Intense Laser Pulse into a Pair of Surface Plasmons, PRB **106**, L081408 (2022)

VERY CLOSE, BUT DIFFERENT:

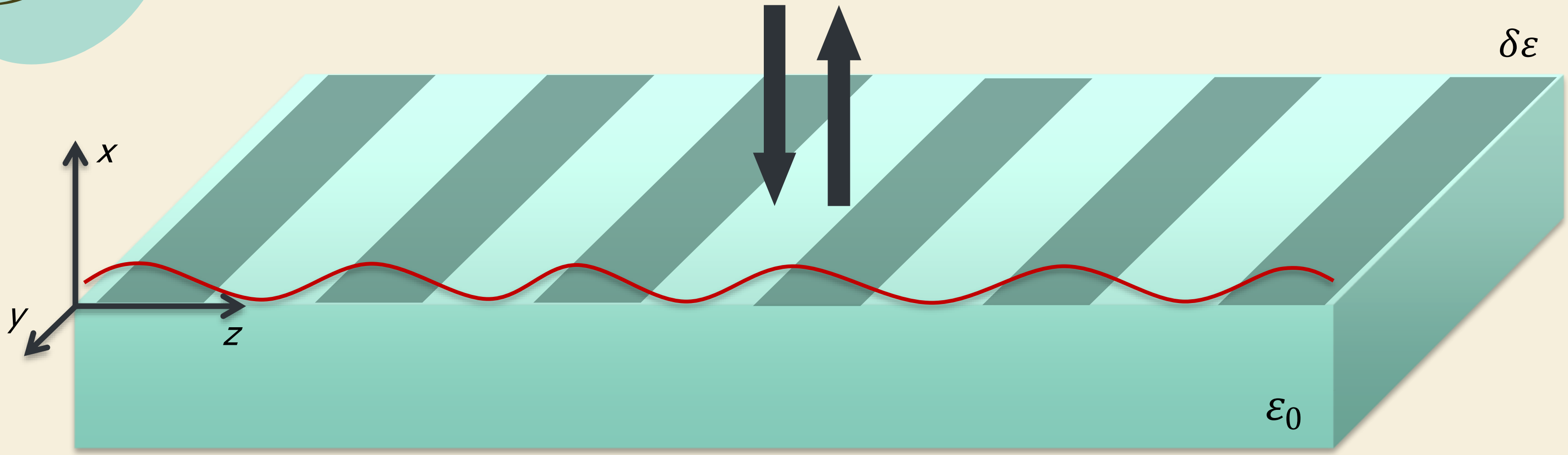
- Evgeny L. Gurevich et al., PRB 95, 054305 (2017) – heat conductivity decrease with temperature growth
- V. B. Gildenburg, I. A. Pavlichenko, Nanomaterials 10, 1461 (2020) – ionization of the unstable layer (in glass)



Excitation of SPP

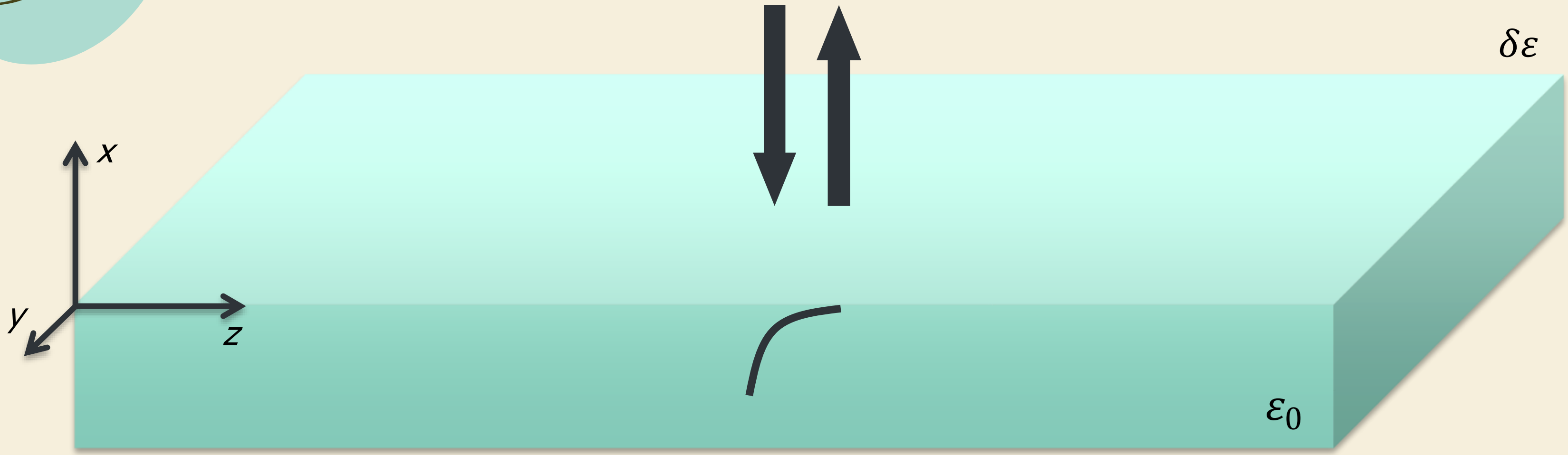
02

Conversion to SPP on a random perturbation of ε



STATEMENT OF THE PROBLEM

- Flat surface of a metal with Re(ε) < −1
- Normally incident laser pulse
- Conversion of the incident wave to SPPs on the ε perturbation



UNPERTURBED SOLUTION

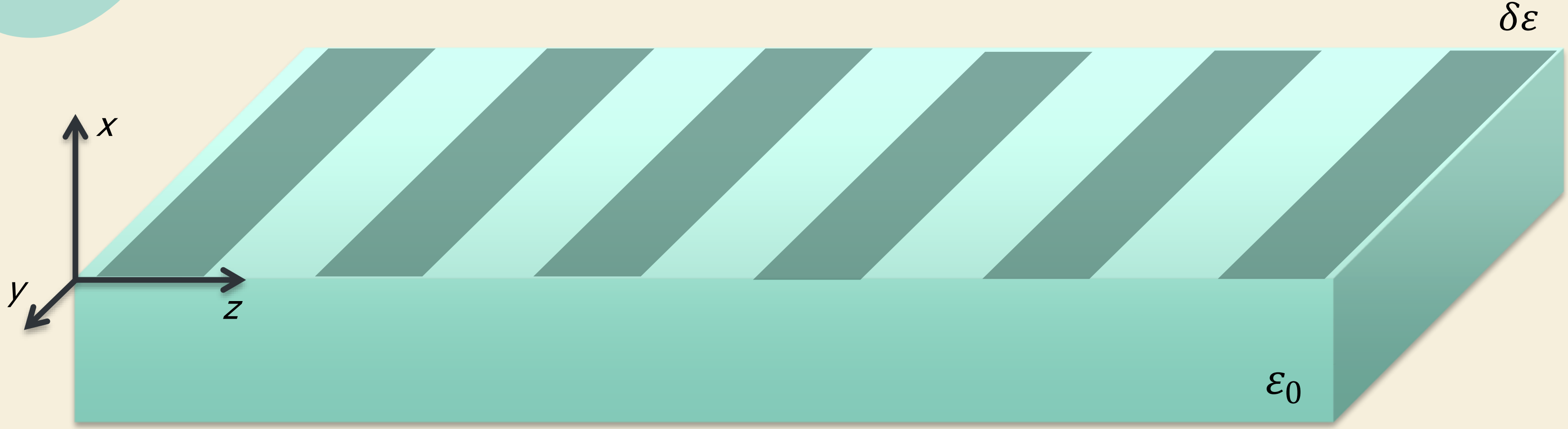
$$x > 0: \ \mathbf{E}_i(t, x, z) = \mathbf{z_0} E_i \exp(i\omega t + i k_0 x)$$

$$x > 0: \ \mathbf{E}_r(t, x, z) = \mathbf{z_0} E_r \exp(i\omega t - i k_0 x)$$

$$x < 0: \ \mathbf{E}_t(t, x, z) = \mathbf{z_0} E_t \exp(i\omega t + \alpha x)$$

$$E_t = \frac{2E_i}{1 - \sqrt{\varepsilon}}$$

$$E_r = E_i \frac{1 + \sqrt{\varepsilon}}{1 - \sqrt{\varepsilon}}$$



ARBITRARY PERTURBATION

$$\delta\varepsilon(x,z,t) = \delta\tilde{\varepsilon}(x,t)\cdot\cos k_\varepsilon z$$

Wave equation inside the medium:

$$\Delta\mathbf{H} - \frac{\varepsilon}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{H} = \frac{1}{c}\frac{\partial}{\partial t}[\boldsymbol{\nabla}\varepsilon, \mathbf{E}]$$

Diffraction. Solution for SPP magnitude

Wave equation inside the medium, in the first perturbation order:

$$\frac{\partial^2 H_1^{\pm}}{\partial x^2} - k_\varepsilon^2 H_1^{\pm} + \varepsilon_0 k_0^2 H_1^{\pm} = -\frac{ik_0 E_t e^{\alpha x}}{2}\frac{\partial \delta\tilde{\varepsilon}(x)}{\partial x} - \frac{k_0^2 \delta\tilde{\varepsilon}(x)}{2} H_t e^{\alpha x}$$

Solution:

$$x < 0:\ H_1^{\pm} = Ae^{\alpha_2 x} + \Psi(x)$$

$$x > 0:\ H_1^{\pm}(x) = Ce^{-\alpha_1 x}$$

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where $\Psi(x) = -\frac{ik_0 g - k_0^2\sqrt{\varepsilon_0}}{(g+\alpha)^2 - k_\varepsilon^2 + \varepsilon_0 k_0^2}\frac{E_i \delta\tilde{\varepsilon}}{\left(1-\sqrt{\varepsilon_0}\right)} e^{(g+\alpha)x}$

$$A = \frac{\varepsilon_0\alpha_1 + g + \alpha}{\alpha_2 + \varepsilon_0\alpha_1}\frac{ik_0 g - k_0^2\sqrt{\varepsilon_0}}{(g+\alpha)^2 - k_\varepsilon^2 + \varepsilon_0 k_0^2}\frac{E_i \delta\tilde{\varepsilon}}{\left(1-\sqrt{\varepsilon_0}\right)}$$

$$C = \frac{g + \alpha - \alpha_2}{\alpha_2 + \varepsilon_0\alpha_1}\frac{ik_0 g - k_0^2\sqrt{\varepsilon_0}}{(g+\alpha)^2 - k_\varepsilon^2 + \varepsilon_0 k_0^2}\frac{E_i \delta\tilde{\varepsilon}}{\left(1-\sqrt{\varepsilon_0}\right)}$$

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$$A = \frac{\varepsilon_0\alpha_1 + g + \alpha}{\alpha_2 + \varepsilon_0\alpha_1}\frac{ik_0 g - k_0^2\sqrt{\varepsilon_0}}{(g+\alpha)^2 - k_\varepsilon^2 + \varepsilon_0 k_0^2}\frac{E_i\delta\tilde{\varepsilon}}{(1-\sqrt{\varepsilon_0})}$$

$$C = \frac{g + \alpha - \alpha_2}{\alpha_2 + \varepsilon_0\alpha_1}\frac{ik_0 g - k_0^2\sqrt{\varepsilon_0}}{(g+\alpha)^2 - k_\varepsilon^2 + \varepsilon_0 k_0^2}\frac{E_i\delta\tilde{\varepsilon}}{(1-\sqrt{\varepsilon_0})}$$

Resonance with SPP mode!

$$k_\varepsilon = k_p = \frac{\omega}{c}\sqrt{\frac{\varepsilon'}{1+\varepsilon'}}$$

+ Drude model
+ Near the resonance:

$$\varepsilon = 1 - \frac{\omega_p^2}{\omega(\omega - i\nu)} \cong -\frac{\omega_p^2}{\omega^2} - i\frac{\nu\omega_p^2}{\omega^3}$$

$$\omega = \omega_0 + \delta\omega$$

Diffraction. Solution for SPP magnitude

Wave equation inside the medium, in the first perturbation order:

$$\frac{\partial^2 H_1^{\pm}}{\partial x^2} - k_\varepsilon^2 H_1^{\pm} + \varepsilon_0 k_0^2 H_1^{\pm} = -\frac{ik_0 E_t e^{\alpha x}}{2}\frac{\partial \delta\tilde{\varepsilon}(x)}{\partial x} - \frac{k_0^2 \delta\tilde{\varepsilon}(x)}{2} H_t e^{\alpha x}$$

Solution:

$$x < 0:\ H_1^{\pm} = A e^{\alpha_2 x} + \Psi(x)$$

$$x > 0:\ H_1^{\pm}(x) = C e^{-\alpha_1 x}$$

$$A(\omega) = C(\omega) = -\frac{\omega_0}{\delta\omega - i\nu\,{\omega_0}^2/2\omega_p^2}\frac{{\omega_0}^4}{4\omega_p^4} E_i(\omega)\delta\tilde{\varepsilon}$$

where

$$A = \frac{\varepsilon_0\alpha_1 + g + \alpha}{\alpha_2 + \varepsilon_0\alpha_1}\frac{ik_0 g - k_0^2\sqrt{\varepsilon_0}}{(g+\alpha)^2 - k_\varepsilon^2 + \varepsilon_0 k_0^2}\frac{E_i\delta\tilde{\varepsilon}}{(1-\sqrt{\varepsilon_0})}$$

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$$\omega = \omega_0 + \delta\omega$$

FINALLY,

conversion to SPPs at the resonant grating (equation for the envelopes):

SPP magnitude

incident field

$$\left(\frac{\partial}{\partial t} + \nu \frac{{\omega_0}^2}{2\omega_p^2}\right) \tilde{E}_z^{\pm}(t) = -\frac{{\omega_0}^6}{4\omega_p^5} \tilde{E}_i(t) \delta\tilde{\varepsilon}$$

damping

linear transformation with respect to E_i and $\delta\varepsilon$

Feedback 03

Growth of ε perturbation, instability development

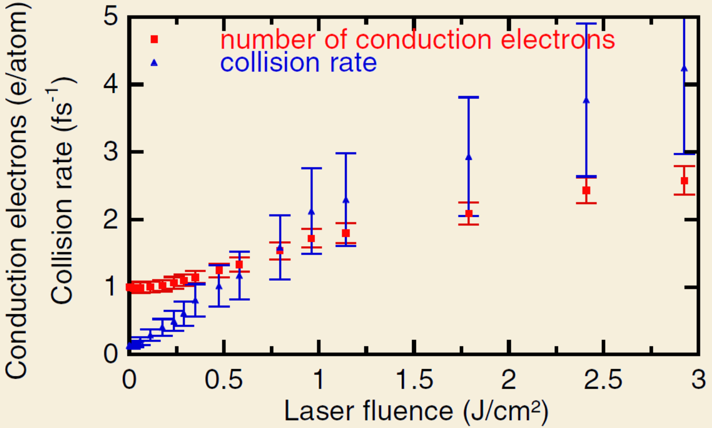
THERMAL NONLINEARITY

Ultrafast heating:

- increase of the electron density **n**
- increase of the electron scattering rate **ν**

$$\varepsilon = 1 - \frac{\omega_p^2}{\omega(\omega - i\nu)} \cong -\frac{\omega_p^2}{\omega^2} - i\frac{\nu\omega_p^2}{\omega^3}$$

$$\nu(W_e) = \nu_0 + \xi\frac{W_e}{\hbar}$$



C. Fourment, F. Deneuville et al., PRB 89, 161110(R) (2014)

INTERFERENCE AND HEATING

Interference of the incident wave and standing wave of counter-propagating SPP → periodic heating

e heating rate = incident field ² + incident field × SPP field

$$\begin{cases} \dfrac{\partial W_e}{\partial t} = \nu \dfrac{e^2 |\mathbf{E}_\Sigma|^2}{2m\omega_0} \cong \nu \dfrac{e^2}{2m\omega_0} \left| E_t^2 e^{2\alpha x} + 4E_t E_z^{\pm} e^{(\alpha+\alpha_2)x} \cos k_\varepsilon z \right| \\ \nu(W_e) = \nu_0 + \xi \dfrac{W_e}{\hbar} \end{cases}$$

FULL SYSTEM. INSTABILITY

SPP excitation

$$\left(\frac{\partial}{\partial t} + \nu \frac{{\omega_0}^2}{2\omega_p^2}\right)\tilde{E}_z^{\pm}(t) = -\frac{{\omega_0}^6}{4\omega_p^5}\tilde{E}_i(t)\delta\tilde{\varepsilon}$$

e heating

$$\frac{\partial W_e}{\partial t} = \nu \frac{e^2}{2m\omega_0}\left|E_t^2 e^{2\alpha x} + 4E_t E_z^{\pm} e^{(\alpha+\alpha_2)x}\cos k_\varepsilon z\right|$$

Drude model

$$\varepsilon \cong -\frac{\omega_p^2}{\omega^2} - i\frac{\nu\omega_p^2}{\omega^3}$$

scattering ↑

$$\nu(W_e) = \nu_0 + \xi\frac{W_e}{\hbar}$$

FULL SYSTEM. INSTABILITY

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$$\left(\frac{\partial}{\partial t}+\nu\frac{{\omega_0}^2}{2\omega_p^2}\right)\tilde{E}_z^{\pm}(t)=-\frac{{\omega_0}^6}{4\omega_p^5}\tilde{E}_i(t)\delta\tilde{\varepsilon}$$

e heating

$$\frac{\partial W_e}{\partial t}=\nu\frac{e^2}{2m\omega_0}\left|E_t^2e^{2\alpha x}+4E_tE_z^{\pm}e^{(\alpha+\alpha_2)x}\cos k_\varepsilon z\right|$$

Drude model

$$\varepsilon\cong-\frac{\omega_p^2}{\omega^2}-i\frac{\nu\omega_p^2}{\omega^3}$$

scattering ↑

$$\nu(W_e)=\nu_0+\xi\frac{W_e}{\hbar}$$

Initial stage of the instability development:

$$\Gamma_0=\frac{{\omega_0}^2}{\omega_p^2}\sqrt{\xi\nu_0\frac{E_i^2e^2}{\hbar m\omega_0^2}}$$

Estimations:

Laser pulse: 100 fs, 800 nm, 1 J/cm^2
Au: ξ = 0.5, $\varepsilon_o \cong -26-1.85i$

- Initially: **Γ_o^{-1} ≅ 50 fs**
- After heating: **Γ_o^{-1} ≅ 10-15 fs**

DECAY. SATURATION REGIME

Due to strong heating the SPP lifetime goes down to 20–40 fs

Balance between the pumping and absorption

$$\left(\frac{\partial}{\partial t} + \nu \frac{{\omega_0}^2}{2\omega_p^2}\right)\tilde{E}_z^{\pm}(t) = -\frac{{\omega_0}^6}{4\omega_p^5}\tilde{E}_i(t)\delta\tilde{\varepsilon}$$

$$\delta\varepsilon \cong -i\frac{\delta\nu\omega_p^2}{\omega^3}$$

$$\tilde{E}_{z,sat}^{\pm} = i\frac{\omega_0}{2\omega_p}\tilde{E}_i \cong E_t/4$$

- modulation of the total electric field from 0.5 E_t to 1.5 E_t
- contrast in the laser intensity: **9 times!**

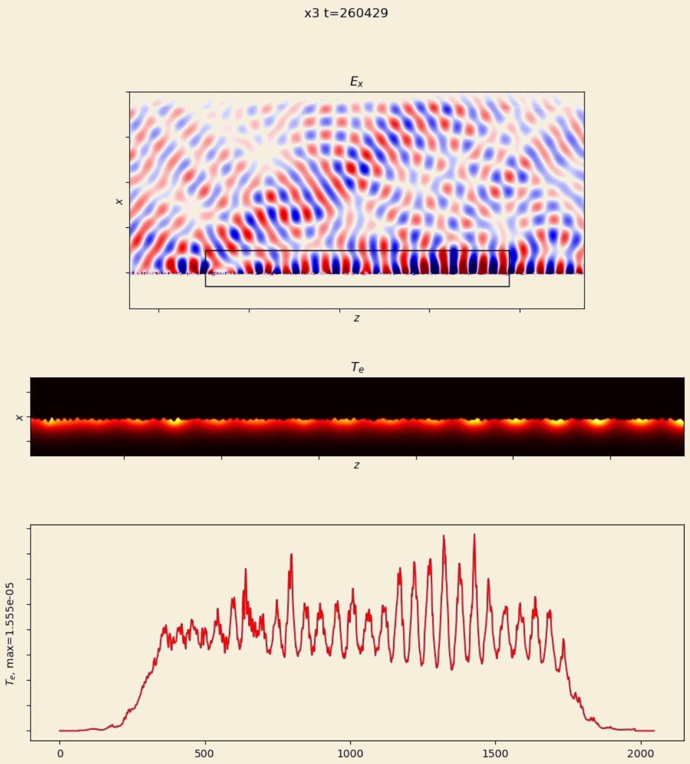
Numerical modeling

04

Estimations and preliminary results of calculations

LASER PULSE DECAY ON A RANDOM SURFACE

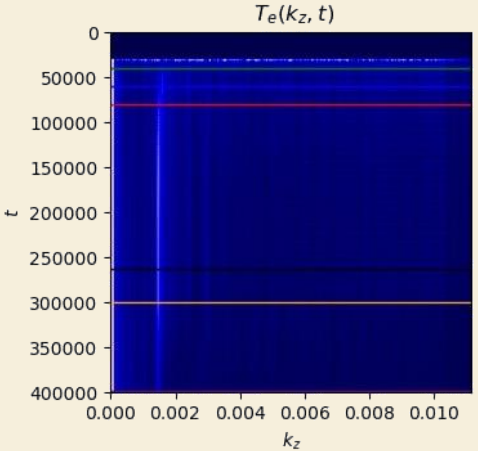
calculations by Daniil Fadeev, IAP RAS



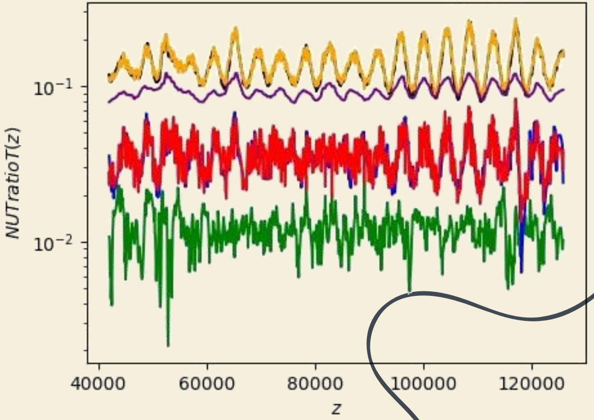
LASER PULSE DECAY ON A RANDOM SURFACE

calculations by Daniil Fadeev, IAP RAS

Temperature (k_z) →



Time ↓



CONCLUSION

- There is a **positive feedback** between the SPP excitation and perturbation of the medium permittivity
- According to the estimations, this influences strongly on the process of metal heating by the laser pulses of damaging intensities

I. Oladyshkin, Phys. Rev. B **106**, L081408 (2022)

THANKS!

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