Phenomenology and underlying physics of time-varying media

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Time-varying media

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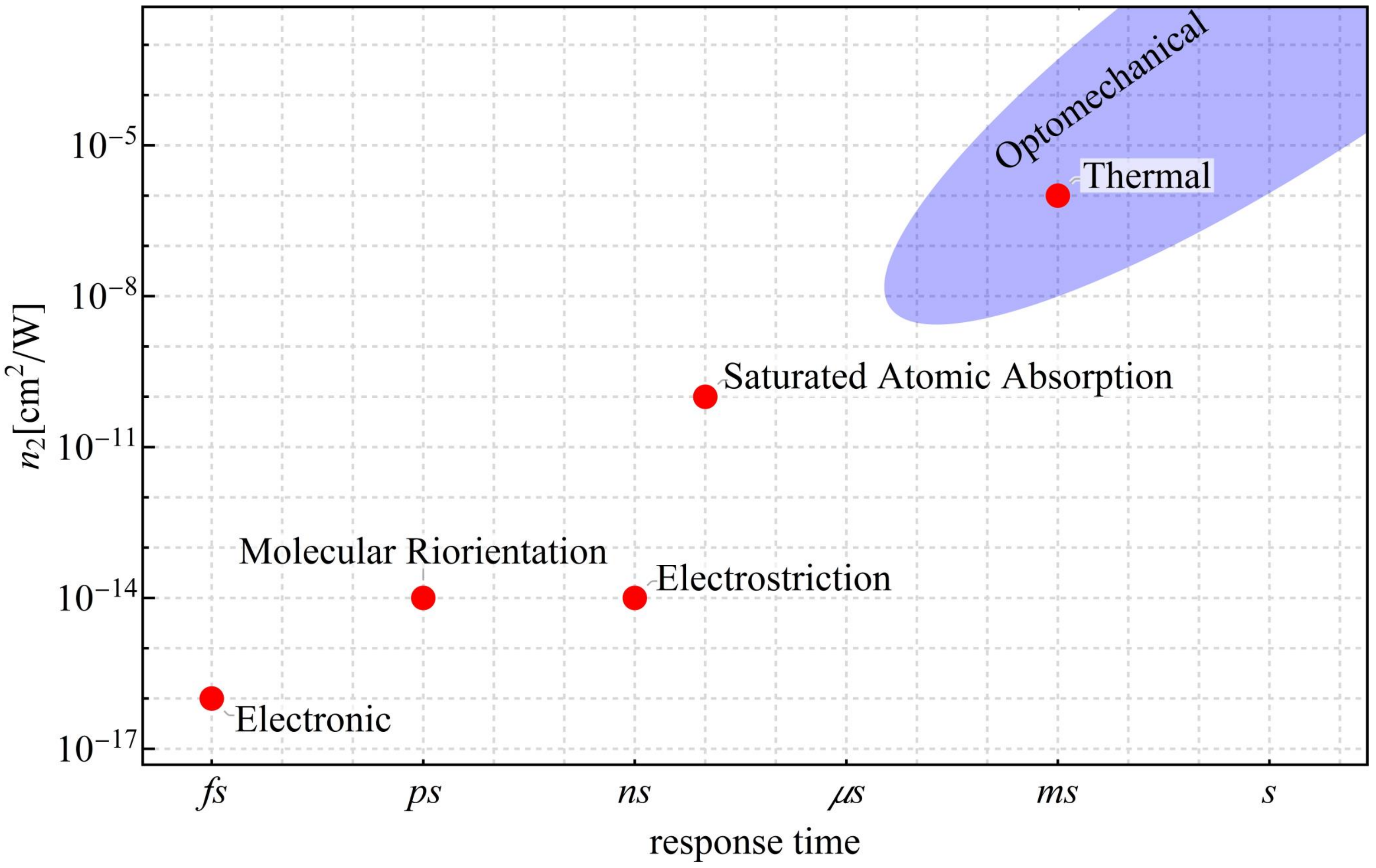
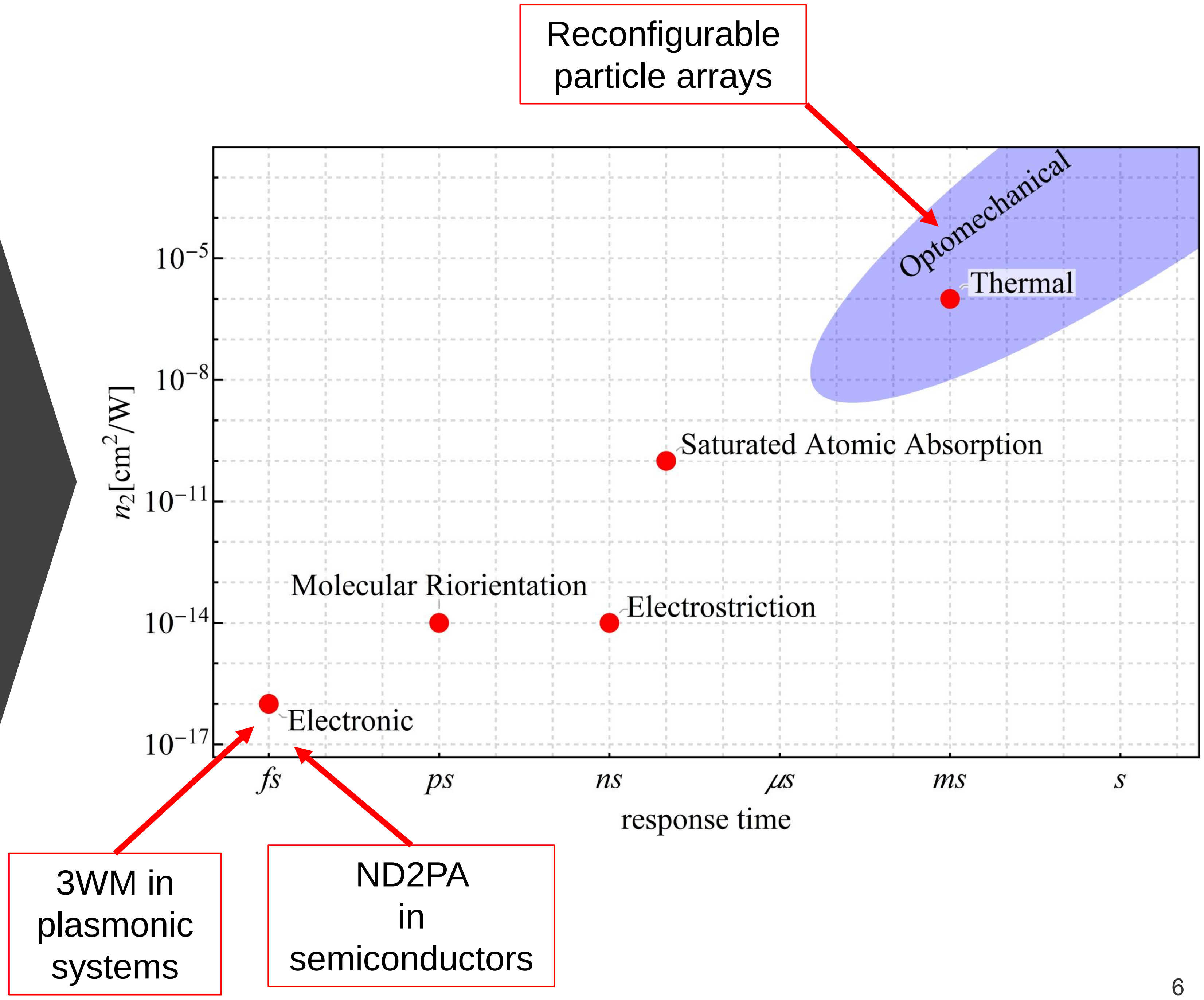
Pump-probe framework: NL optical interactions mediated by a pump field can be exploited to produce a time-varying susceptibility for a probe field.

Amplitude
VS
response- time
tradeoff



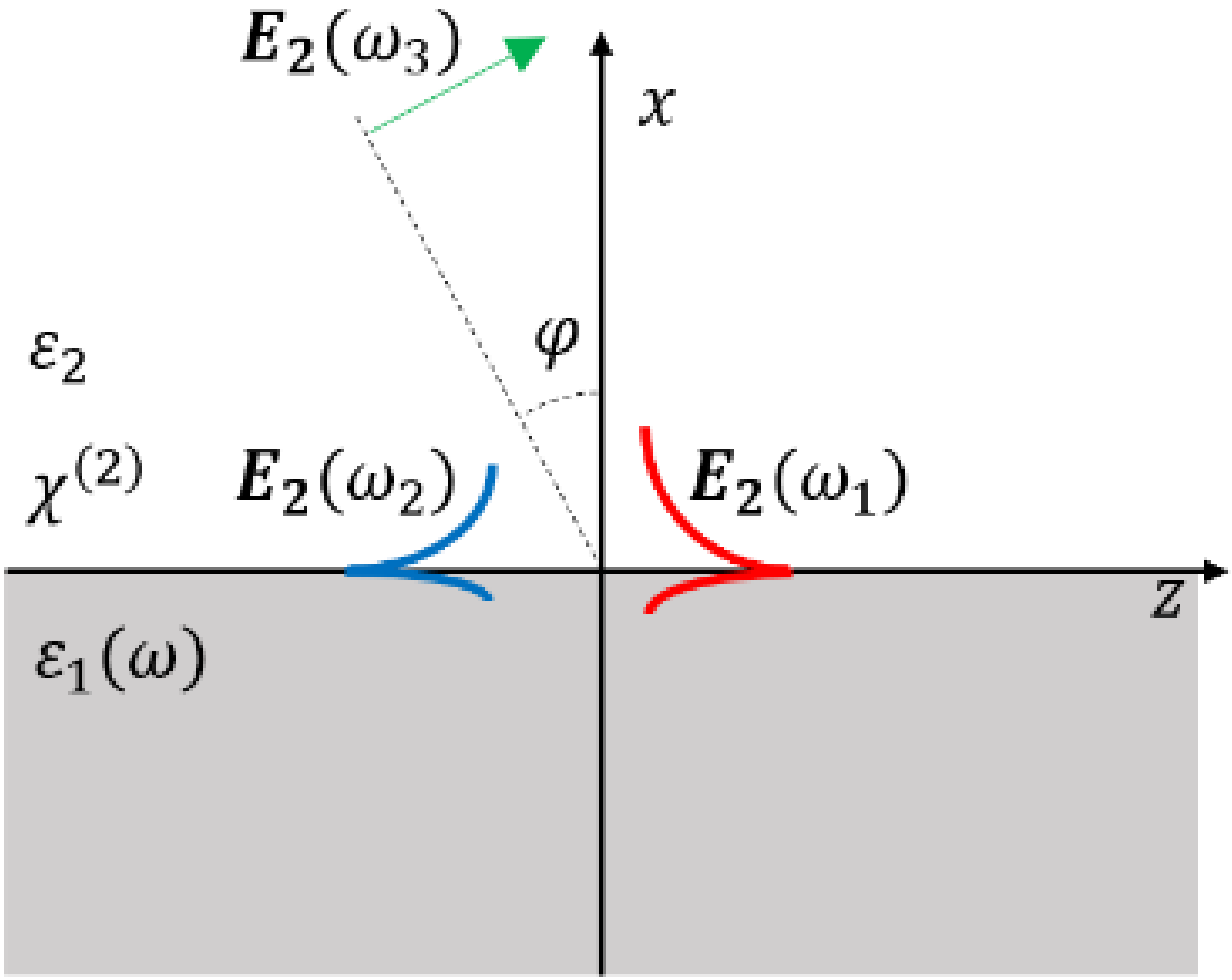
Amplitude VS response- time tradeoff



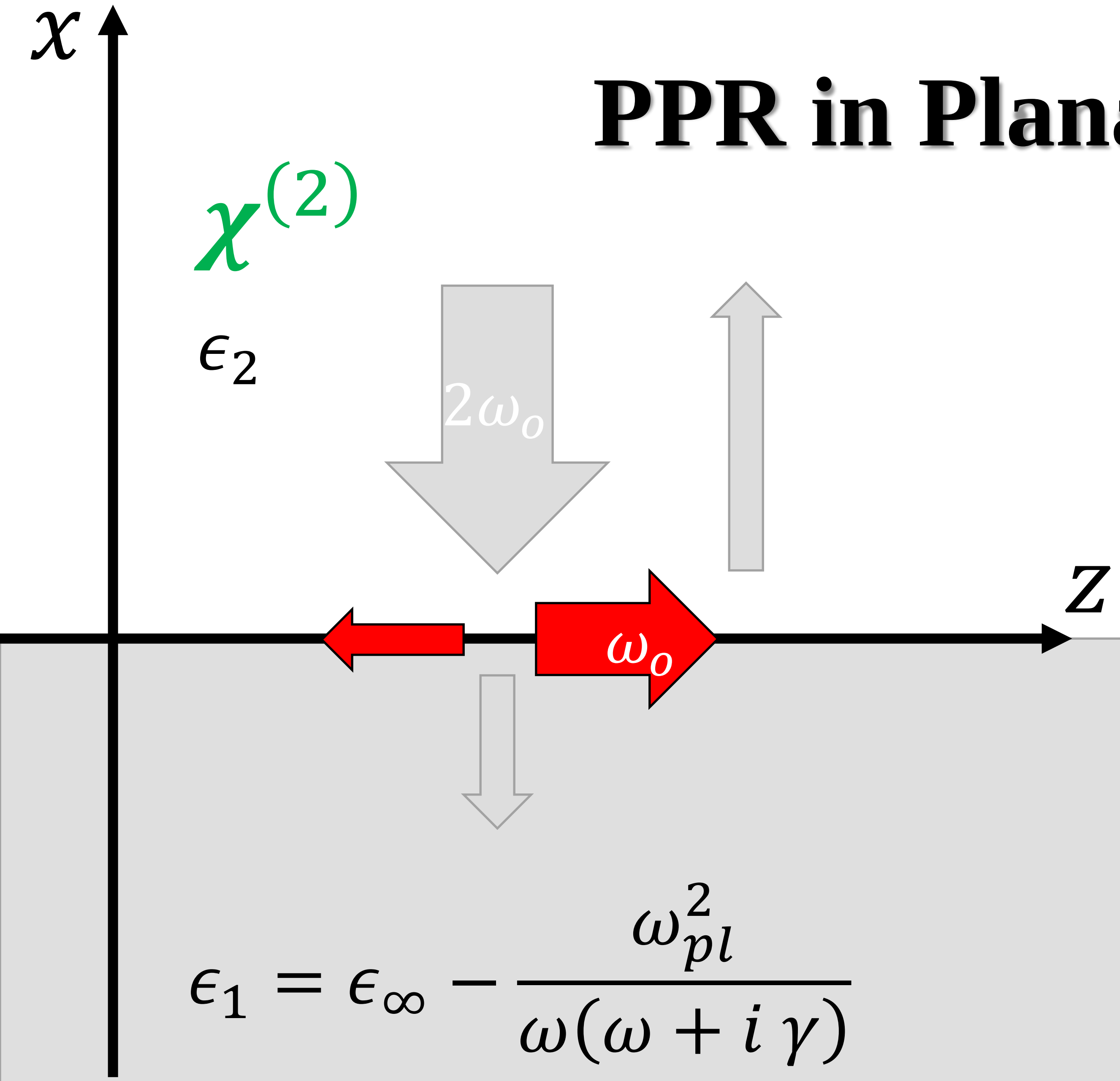
Effective linear regimes in plasmonic three-wave mixing*

*Luca Stefanini, Davide Ramaccia, Filiberto Bilotti, Shima Fardad, and Alessandro Salandrino,
"Effective linear regimes in plasmonic three-wave mixing," **J. Opt. Soc. Am. B** 41, 1968-1978 (2024)

System of interest



PPR in Planar Systems



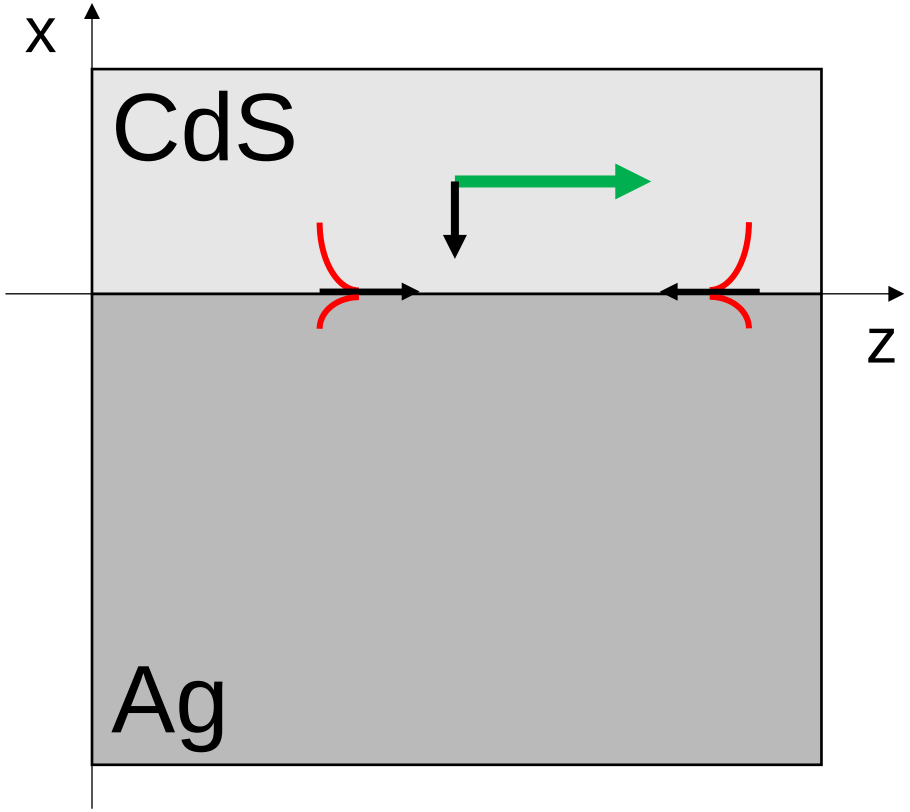
$$\nabla^2 \mathbf{E}_2 - \frac{\varepsilon_2}{c^2}\frac{\partial^2 \mathbf{E}_2}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2}$$

$$\nabla^2 \mathbf{E}_1 - \frac{\varepsilon_\infty}{c^2}\frac{\partial^2 \mathbf{E}_1}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}_1}{\partial t^2}$$

$$\frac{\partial^2 \mathbf{P}_1(\mathbf{r},t)}{\partial t^2} + \gamma \frac{\partial \mathbf{P}_1(\mathbf{r},t)}{\partial t} = \varepsilon_0 \omega_p^2 \mathbf{E}_1(\mathbf{r},t)$$

$$\epsilon_1 = \epsilon_\infty - \frac{\omega_{pl}^2}{\omega(\omega + i\,\gamma)}$$

Analysis of a realistic system

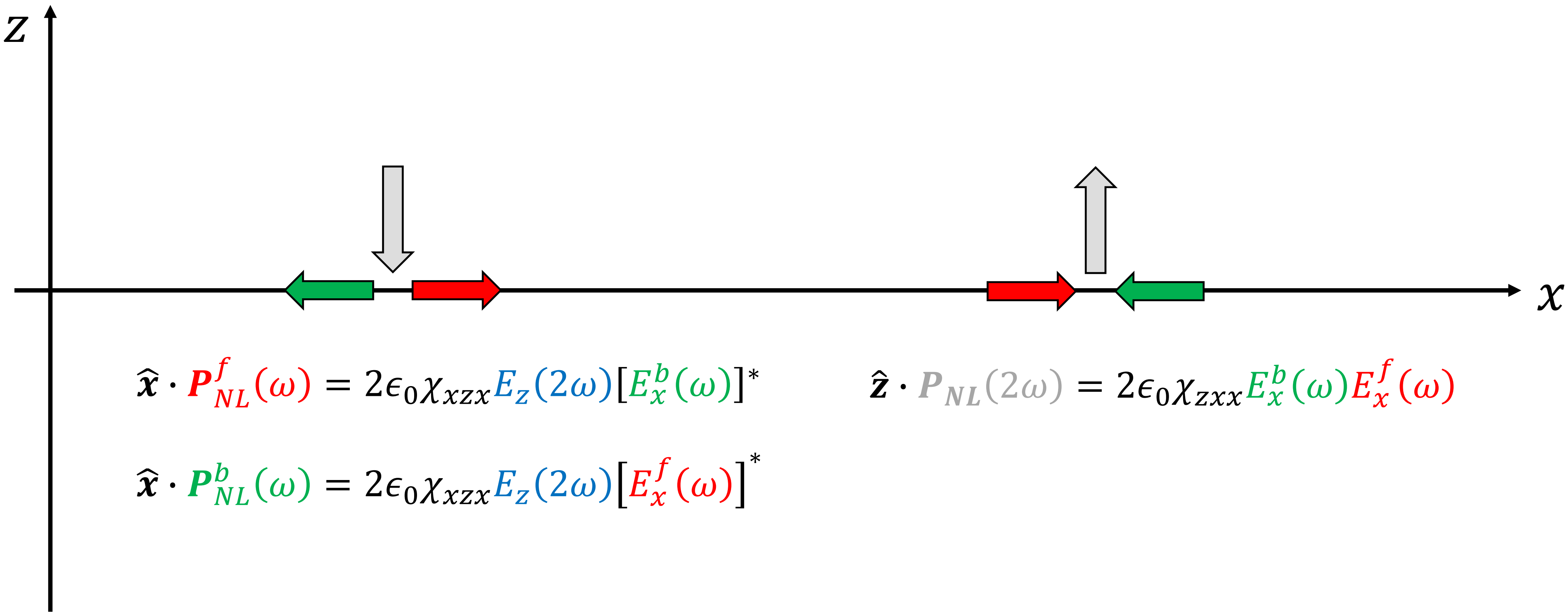


Crystals belonging to the C_{6v} point group symmetry are suitable for mixing two co-polarized and one cross-polarized field.
For **Cadmium Sulfide**:

$$\chi^{(2)}_{zxx} = \chi^{(2)}_{xzx} = \chi^{(2)}_{xxz} = -40\,\frac{pm}{V}$$

Bandgap 2.42 eV (512nm)
Permittivity ~5.4
Plasmonic Resonance 415 nm

Nonlinear Polarization



$$\hat{\boldsymbol{x}} \cdot \boldsymbol{P}_{NL}^{f}(\omega) = 2\epsilon_0 \chi_{xzx} E_z(2\omega)[E_x^b(\omega)]^*$$

$$\hat{\boldsymbol{x}} \cdot \boldsymbol{P}_{NL}^{b}(\omega) = 2\epsilon_0 \chi_{xzx} E_z(2\omega)\left[E_x^f(\omega)\right]^*$$

$$\hat{\boldsymbol{z}} \cdot \boldsymbol{P}_{NL}(2\omega) = 2\epsilon_0 \chi_{zxx} E_x^b(\omega) E_x^f(\omega)$$

DC, 3ω, and 4ω terms are negligible.

Dynamical equation

$$\frac{\partial^2 \mathbf{P}_1(\mathbf{r},t)}{\partial t^2}+\gamma\frac{\partial \mathbf{P}_1(\mathbf{r},t)}{\partial t}=\varepsilon_0\omega_p^2\mathbf{E}_1(\mathbf{r},t) \xrightarrow{\text{B.C.}} \frac{\partial^2 \mathbf{P}_1(\mathbf{r},t)}{\partial t^2}+\gamma\frac{\partial \mathbf{P}_1(\mathbf{r},t)}{\partial t}+\left[\omega(\varepsilon_2)\right]^2\mathbf{P}_1(\mathbf{r},t)=0$$

$$\xrightarrow{\text{modulation}} \frac{\partial^2 \mathbf{P}_1(\mathbf{r},t)}{\partial t^2}+\gamma\frac{\partial \mathbf{P}_1(\mathbf{r},t)}{\partial t}+\left[\omega(\varepsilon_2)\right]^2\mathbf{P}_1(\mathbf{r},t)+\frac{\partial\left[\omega(\varepsilon_2)\right]^2}{\partial\varepsilon_2}\delta\varepsilon_2\mathbf{P}_1(\mathbf{r},t)=0$$

$$\hat{\mathbf{x}}\cdot\mathbf{D}_2(\mathbf{r},t)=\varepsilon_0\varepsilon_2\hat{\mathbf{x}}\cdot\mathbf{E}_2(\mathbf{r},t)+\varepsilon_0\underbrace{2\chi_{xzx}^{(2)}\left[\hat{\mathbf{z}}\cdot\mathbf{E}_P(\mathbf{r},t)\right]}_{\delta\varepsilon_{xx}}\left[\hat{\mathbf{x}}\cdot\mathbf{E}_2(\mathbf{r},t)\right]$$

$$\boxed{\frac{\partial^2 \mathbf{P}_1(\mathbf{r},t)}{\partial t^2}+\gamma\frac{\partial \mathbf{P}_1(\mathbf{r},t)}{\partial t}+\omega^2\mathbf{P}_1(\mathbf{r},t)=\omega^2 f_1 A_P(t)\sin\left(2\omega t\right)\mathbf{P}_1(\mathbf{r},t)}$$

Threshold Condition

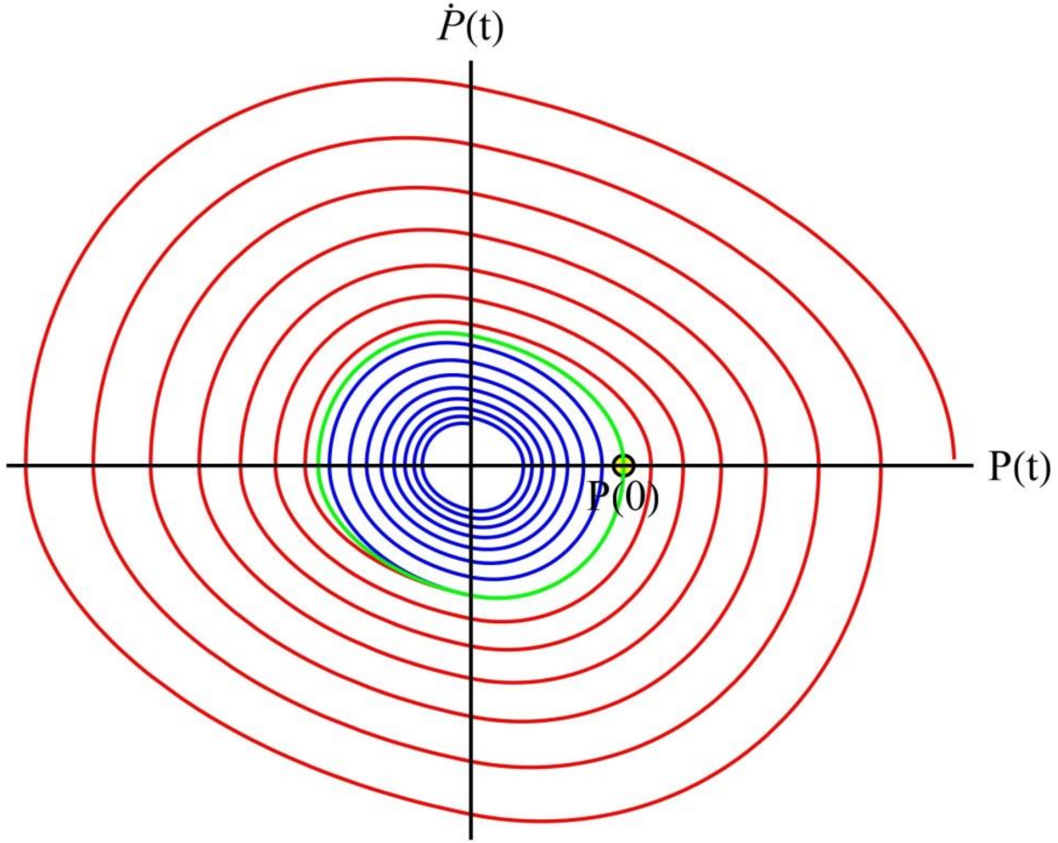
$$\frac{\partial^2 \mathbf{P}_1(\mathbf{r},t)}{\partial t^2} + \gamma \frac{\partial \mathbf{P}_1(\mathbf{r},t)}{\partial t} + \omega^2 \mathbf{P}_1(\mathbf{r},t) = \omega^2 f_1 A_P(t) \sin\left(2\omega t\right) \mathbf{P}_1(\mathbf{r},t)$$

$$P\left(t\right) = p(t)\cos\left[\omega t - \theta(t)\right] \mathrm{e}^{-\frac{\gamma}{2}t}$$

$$p(t) = p_0 \sqrt{\cosh\left(\frac{A_p}{A_{PPR}}\gamma t\right)}$$

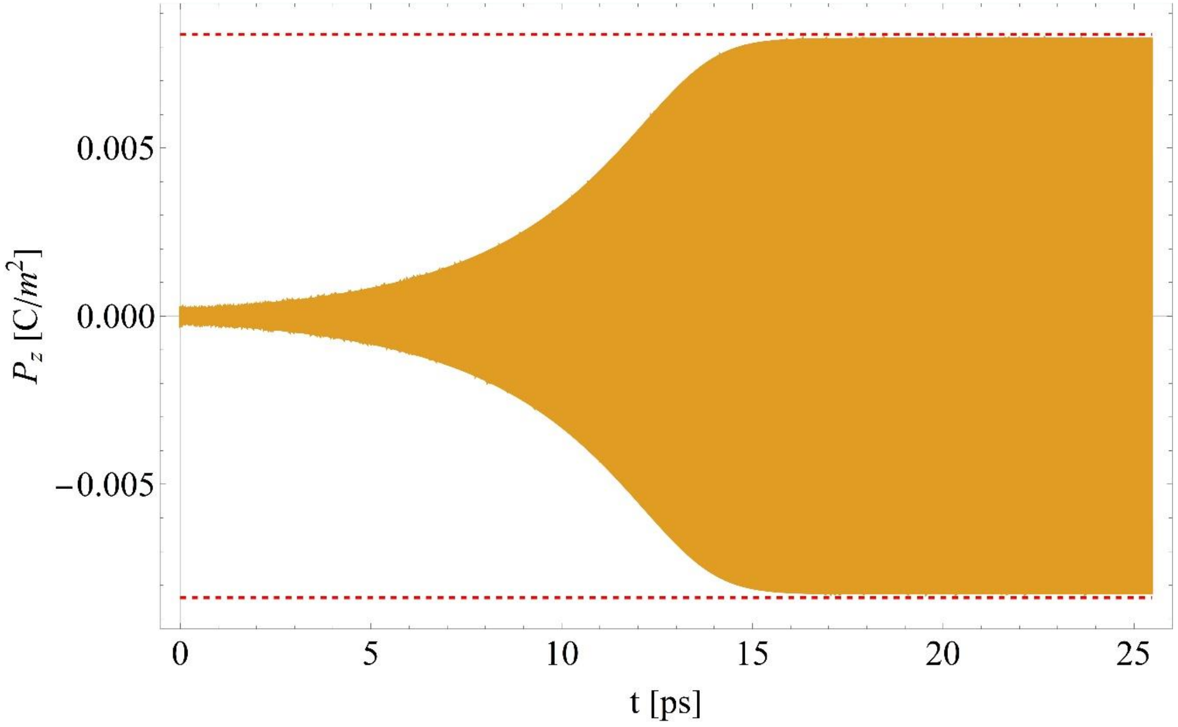
$$\theta(t) = \operatorname{arccot}\left[\exp\left(-\frac{A_p}{A_{PPR}}\gamma t\right)\right]$$

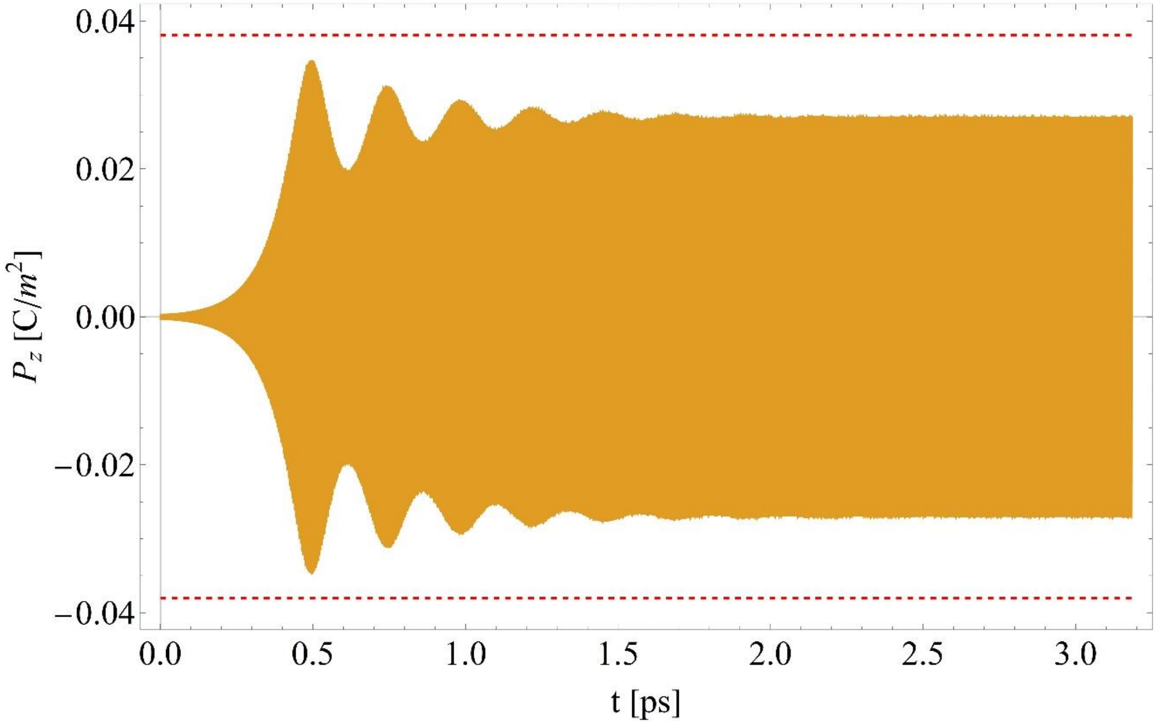
$$A_{PPR} = \frac{2\gamma}{\omega f_1}$$



Saturation

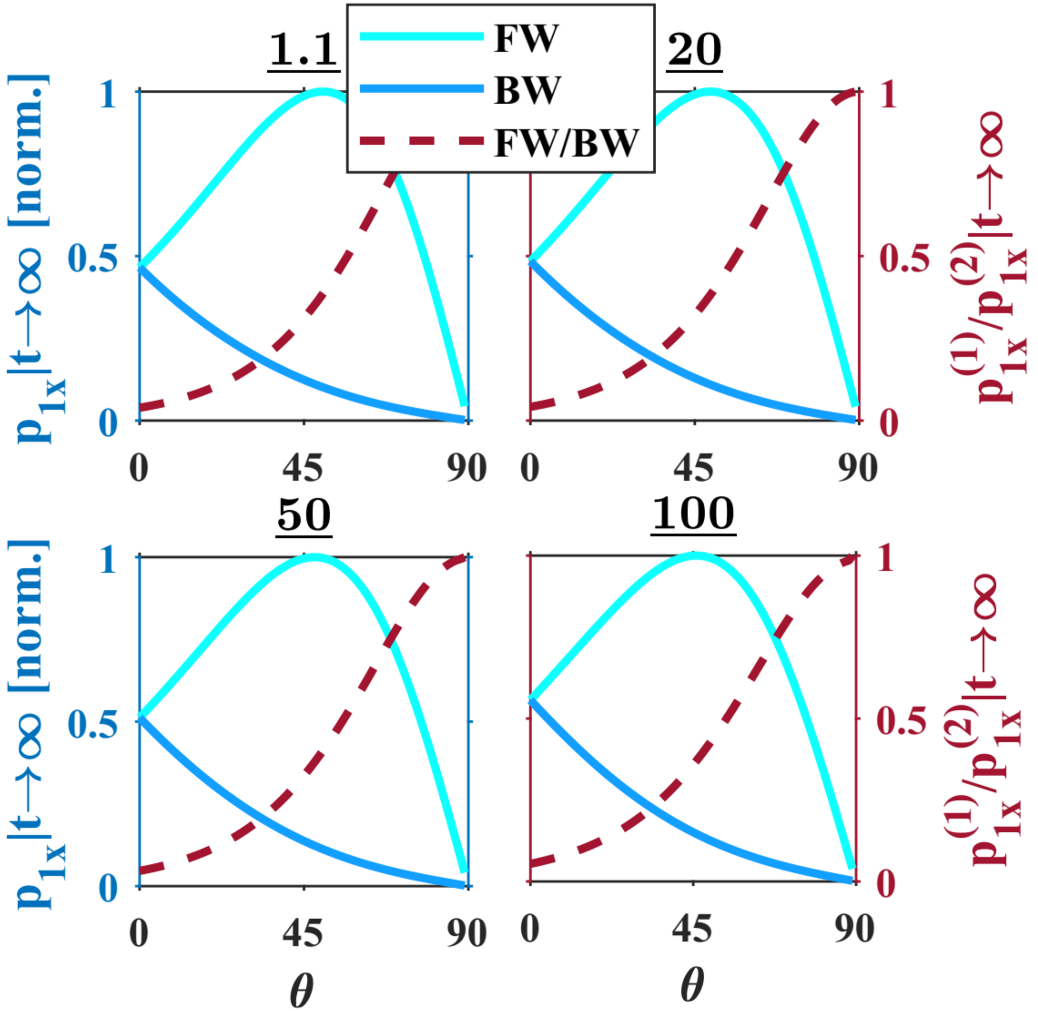
$$\frac{\partial^2 \mathbf{P}_1(\mathbf{r},t)}{\partial t^2} + \gamma \frac{\partial \mathbf{P}_1(\mathbf{r},t)}{\partial t} + \omega^2 \mathbf{P}_1(\mathbf{r},t) = \omega^2 \left[f_1 A_P(t) \sin(2\omega t) \mathbf{P}_1(\mathbf{r},t) + f_2 \mathbf{P}_1(\mathbf{r},t)^2 + f_3 \mathbf{P}_1(\mathbf{r},t)^3 \right]$$



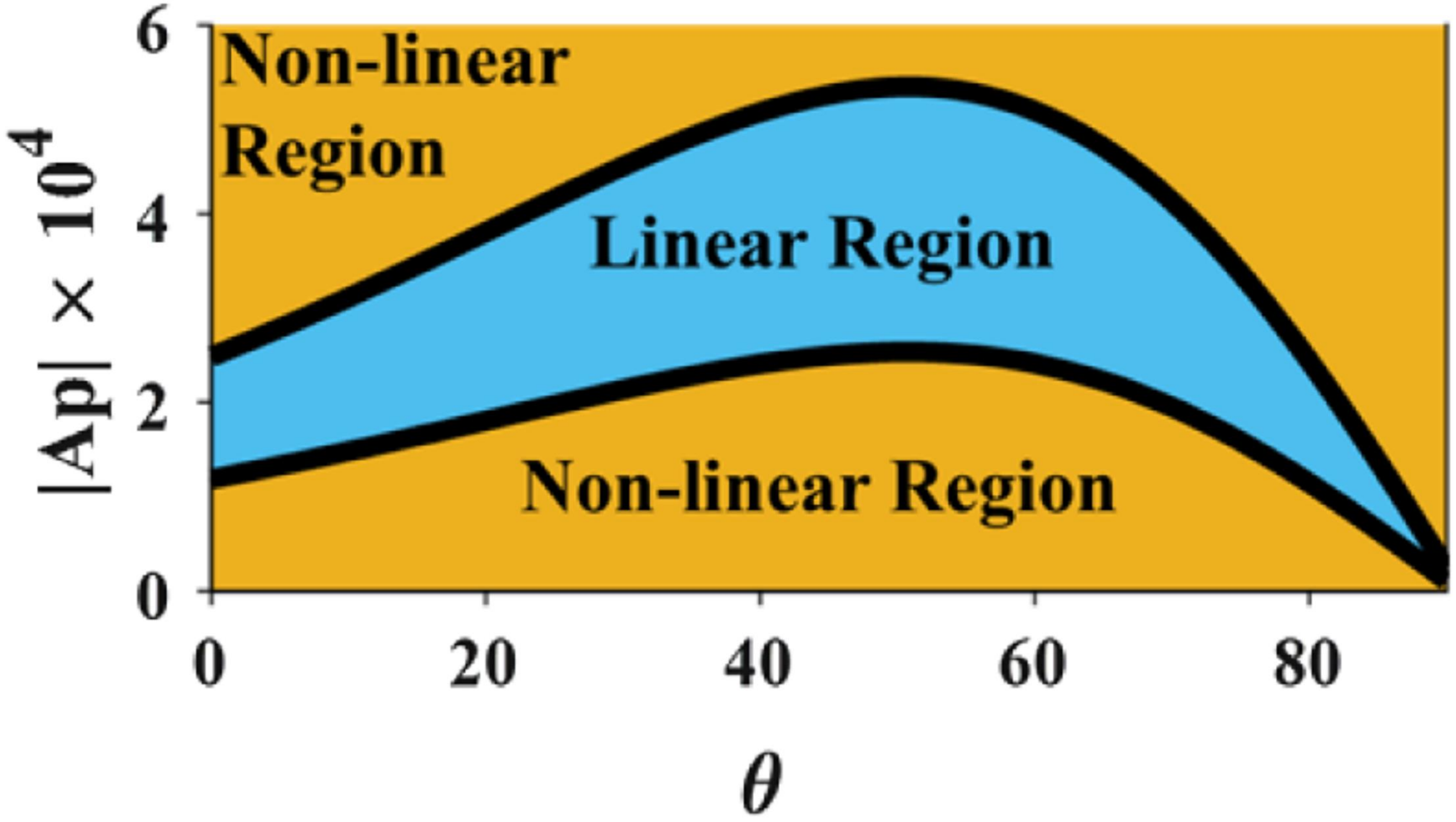


$$\begin{cases} \dfrac{\partial|\bar{p}_{1x}(\Omega_2)|^2}{\partial|\bar{A}_p|^2} = \left|\dfrac{\bar{\chi}^{(\mathrm{III})}_{\mathrm{eff2}}(\Omega_1)}{\bar{\chi}^{(\mathrm{III})}_{\mathrm{eff1}}(\Omega_1)}\right| + \dfrac{1}{2|\bar{A}_p|}\left|\dfrac{\bar{\chi}^{(\mathrm{II})}_{\mathrm{eff}}(\Omega_1)}{\bar{\chi}^{(\mathrm{III})}_{\mathrm{eff1}}(\Omega_1)}\right|\left|\dfrac{\bar{p}^*_{1x}(\Omega_2)}{\bar{p}_{1x}(\Omega_1)}\right| + \dfrac{1}{2}\dfrac{\partial}{\partial|\bar{A}_p|}\left(\left|\dfrac{\bar{\chi}^{(\mathrm{II})}_{\mathrm{eff}}(\Omega_1)}{\bar{\chi}^{(\mathrm{III})}_{\mathrm{eff1}}(\Omega_1)}\right|\left|\dfrac{\bar{p}^*_{1x}(\Omega_2)}{\bar{p}_{1x}(\Omega_1)}\right|\right), \\[2ex] \dfrac{\partial|\bar{p}_{1x}(\Omega_1)|^2}{\partial|\bar{A}_p|^2} = \left|\dfrac{\bar{\chi}^{(\mathrm{III})}_{\mathrm{eff2}}(\Omega_2)}{\bar{\chi}^{(\mathrm{III})}_{\mathrm{eff1}}(\Omega_2)}\right| + \dfrac{1}{2|\bar{A}_p|}\left|\dfrac{\bar{\chi}^{(\mathrm{II})}_{\mathrm{eff}}(\Omega_2)}{\bar{\chi}^{(\mathrm{III})}_{\mathrm{eff1}}(\Omega_2)}\right|\left|\dfrac{\bar{p}^*_{1x}(\Omega_1)}{\bar{p}_{1x}(\Omega_2)}\right| + \dfrac{1}{2}\dfrac{\partial}{\partial|\bar{A}_p|}\left(\left|\dfrac{\bar{\chi}^{(\mathrm{II})}_{\mathrm{eff}}(\Omega_2)}{\bar{\chi}^{(\mathrm{III})}_{\mathrm{eff1}}(\Omega_2)}\right|\left|\dfrac{\bar{p}^*_{1x}(\Omega_1)}{\bar{p}_{1x}(\Omega_2)}\right|\right). \end{cases}$$

Under what conditions is the steady state susceptibility linear in the pump amplitude?



Pump linearity region



Summary

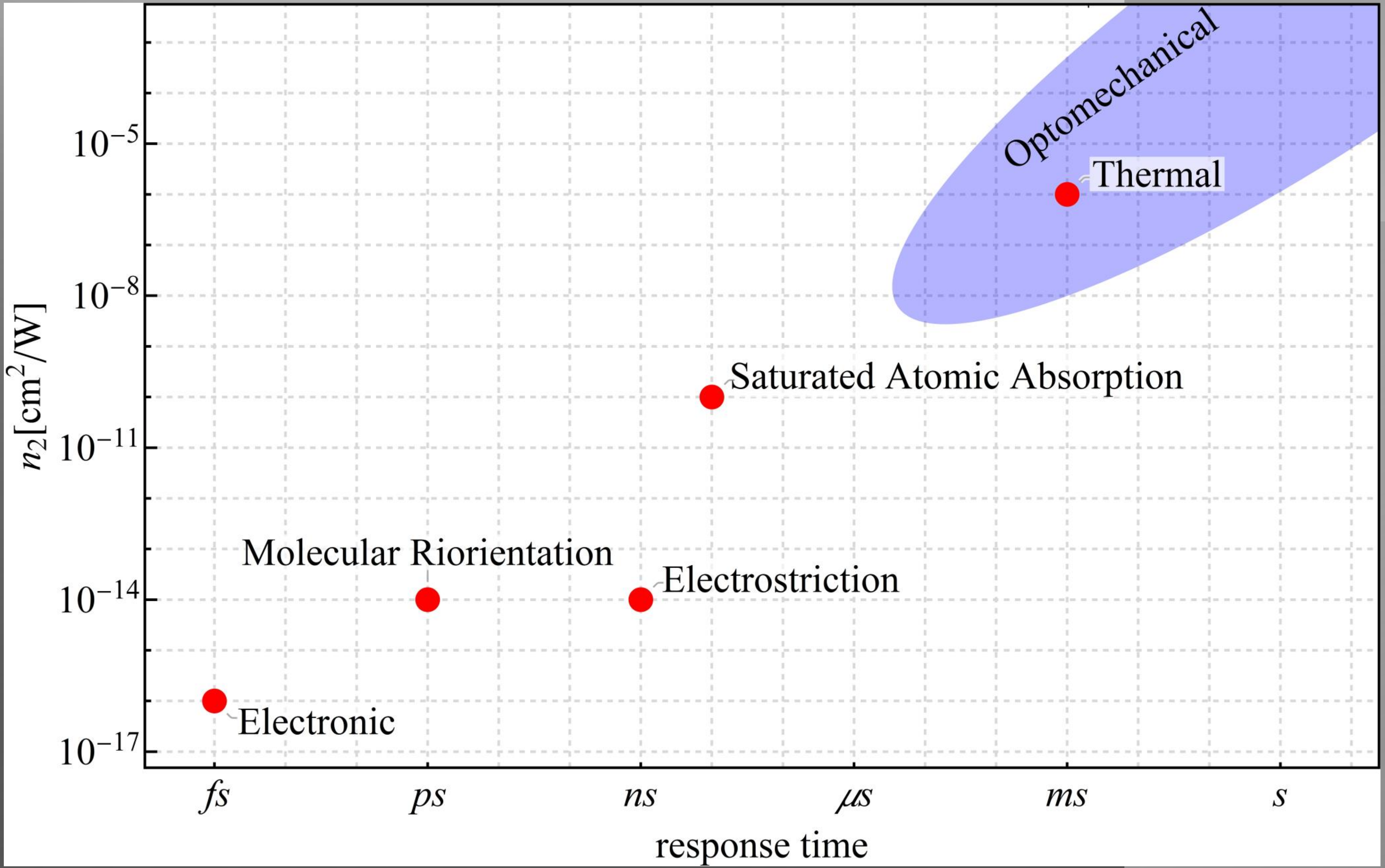
- We developed the theory of three-wave mixing in planar plasmonic structures.

- We identified the parametric resonance conditions.

- We identified the regime of linear response with respect to the pump amplitude for the steady state susceptibility

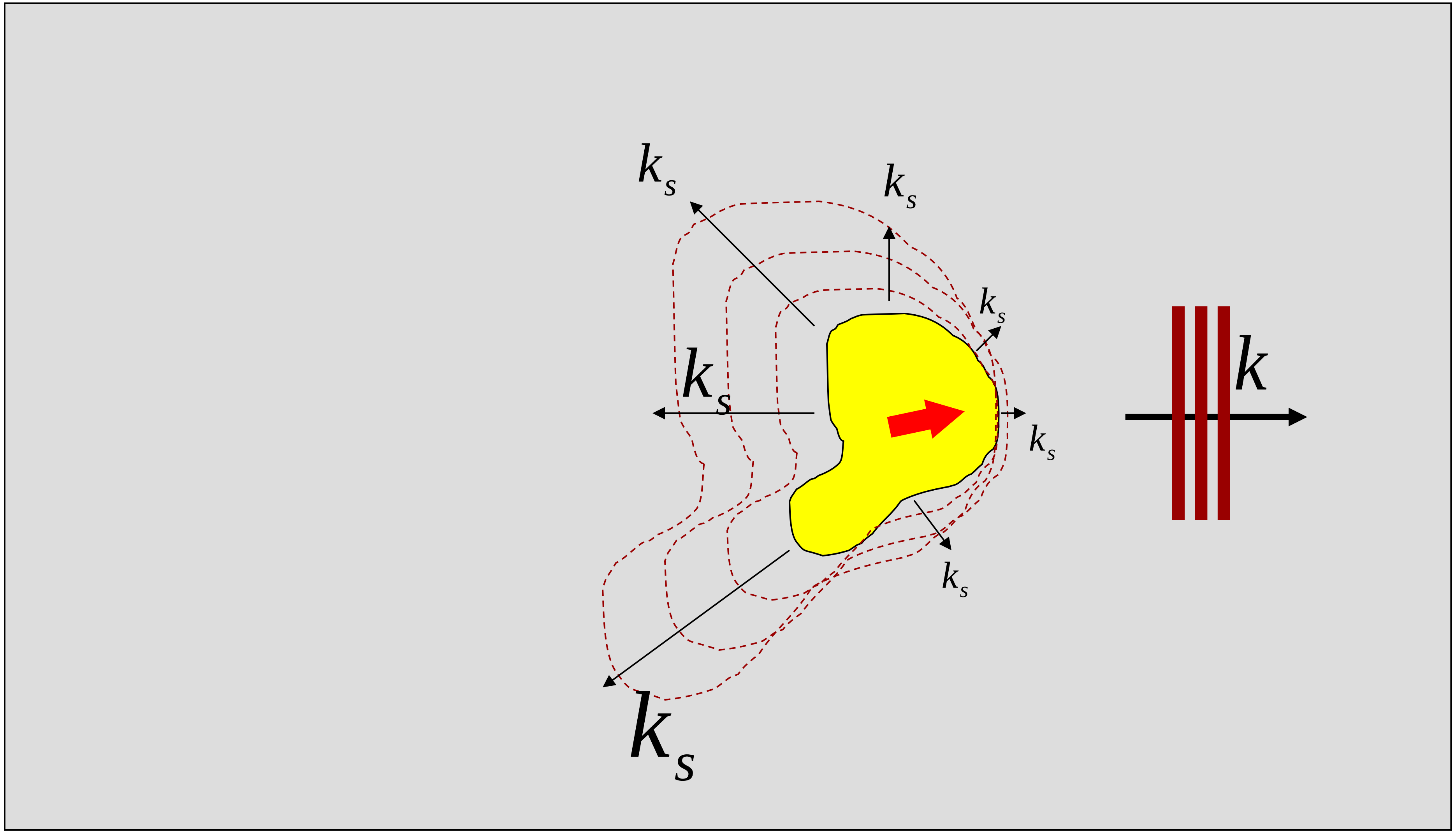
Dynamic dielectric landscape engineering via modified Gerchberg-Saxton Algorithm*

*Bretton Scarbrough, Chase Ward, David Levy, Alessandro Salandrino, Shima Fardad; "Efficient Calculation of Computer-Generated Holograms via Phase Induced Compressive-Sensing Gerchberg-Saxton Algorithm" ***to be submitted.***



Optomechanical
Thermal
Saturated Atomic Absorption
Molecular Riorientation
Electrostriction
Electronic
n_2[cm^2/W]
10^{-5}
10^{-8}
10^{-11}
10^{-14}
10^{-17}
fs
ps
ns
μs
ms
s
response time

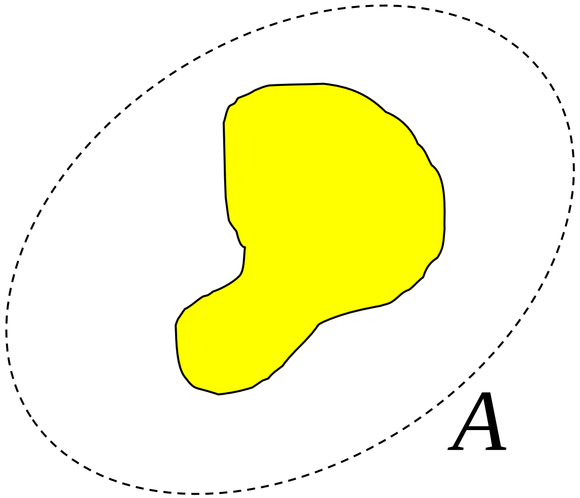
Optomechanical interactions



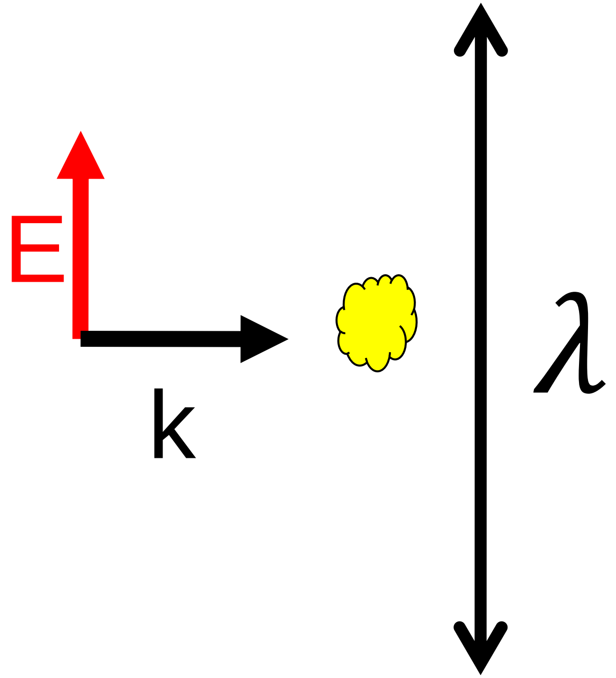
General approach

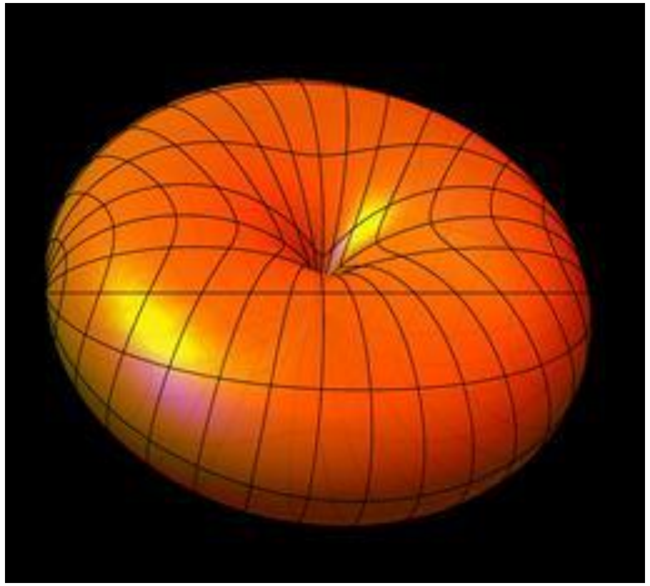
- The amount of momentum that an EM field can deliver per unit of time to a volume is the flux of the Maxwell Stress Tensor through a surface enclosing the volume:

$$\left\langle \vec{F} \right\rangle = \oint \overline{\mathbf{T}} \cdot \hat{\mathbf{n}}\, dA$$



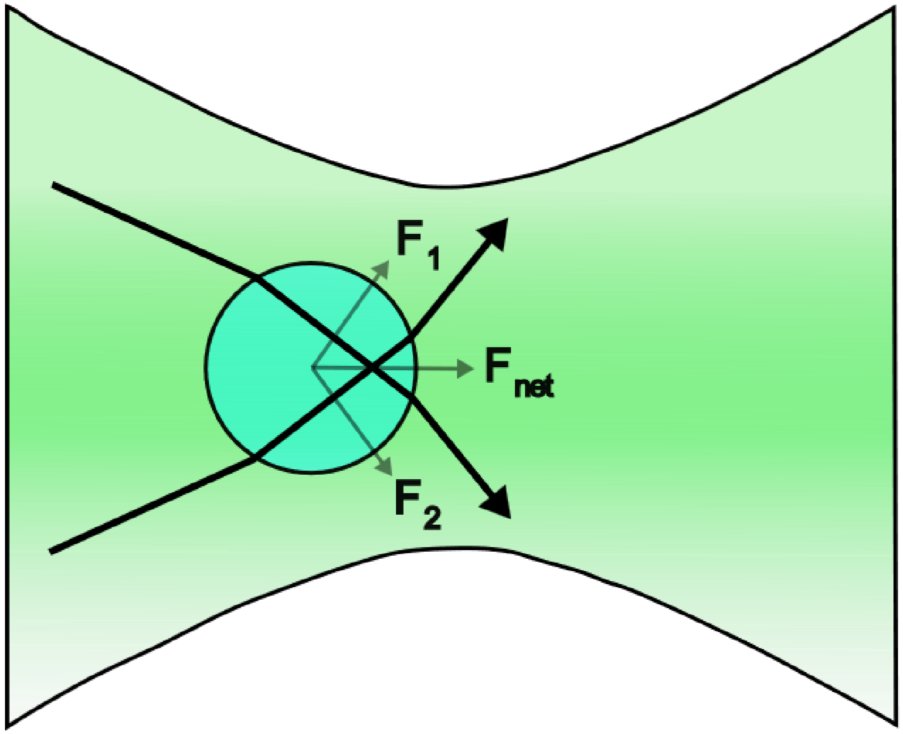
Rayleigh regime

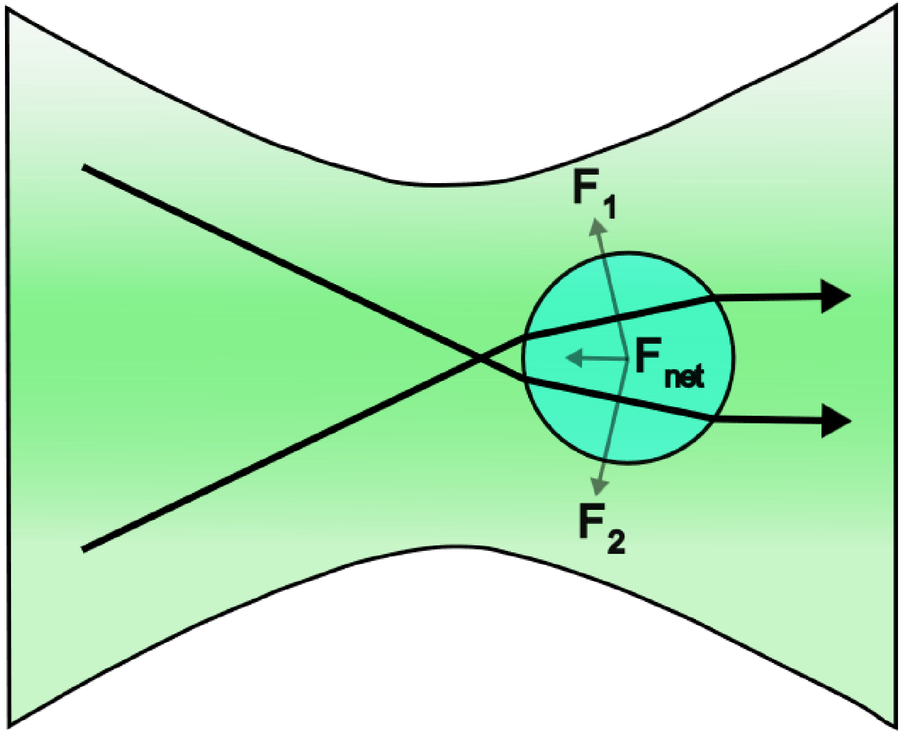




$$\langle \bar{\mathbf{F}} \rangle = \frac{1}{4}\alpha_r \nabla |\mathbf{E}|^2 + \frac{k_0 \alpha_i}{\varepsilon_0}\left[\frac{\langle \mathbf{S} \rangle}{c} + c\nabla \times \langle \mathbf{L}_S \rangle\right]$$

Geometrical optics regime

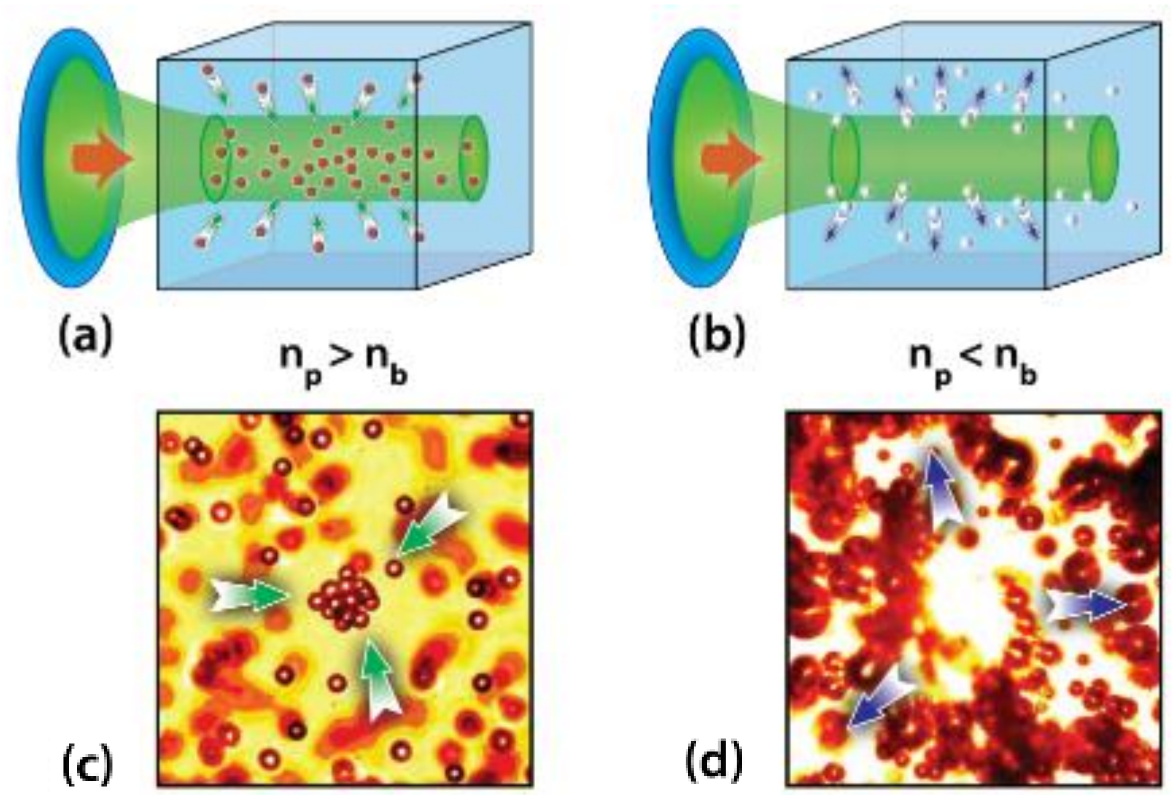




$$\langle \bar{\mathbf{F}} \rangle_{grad} \propto \nabla |\mathbf{E}|^2$$

$$\langle \bar{\mathbf{F}} \rangle_{\substack{rad \\ pressure}} \propto \frac{\langle \mathbf{S} \rangle}{c}$$

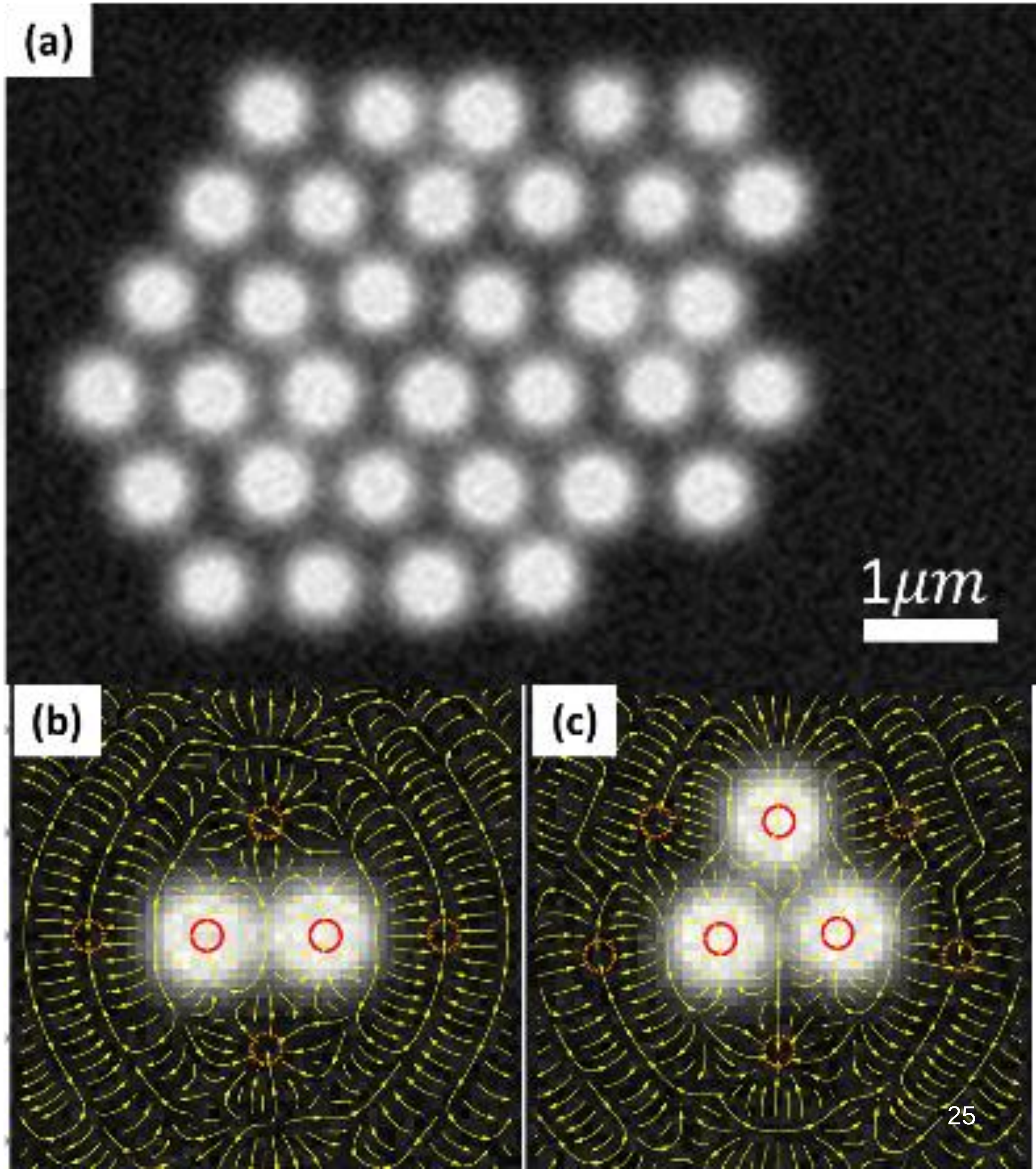
Reconfigurable dielectric landscape



$$\frac{\partial \rho}{\partial t} = -\nabla \cdot \left[\rho\, \mu\, \mathbf{F}_E + \rho\, \mu_T\, \mathbf{F}_T - D \nabla \rho \right]$$

$$\varepsilon_{eff} = \frac{\rho(\mathbf{E}) V_p \varepsilon_p \phi + [1 - \rho(\mathbf{E}) V_p] \varepsilon_b}{\rho(\mathbf{E}) V_p \phi + [1 - \rho(\mathbf{E}) V_p]}$$

Optically-bound particle arrays



- Placement and manipulation of polarizable units with wavelength-scale resolution.

Superposition Algorithm

- A single focus can be calculated by a Fresnel lens.
- The system we are utilizing is linear. Therefore, we can use *superposition* to generate multiple foci.
- This algorithm is fast. However, it is **more susceptible to creating ghost traps** when compared to alternatives.

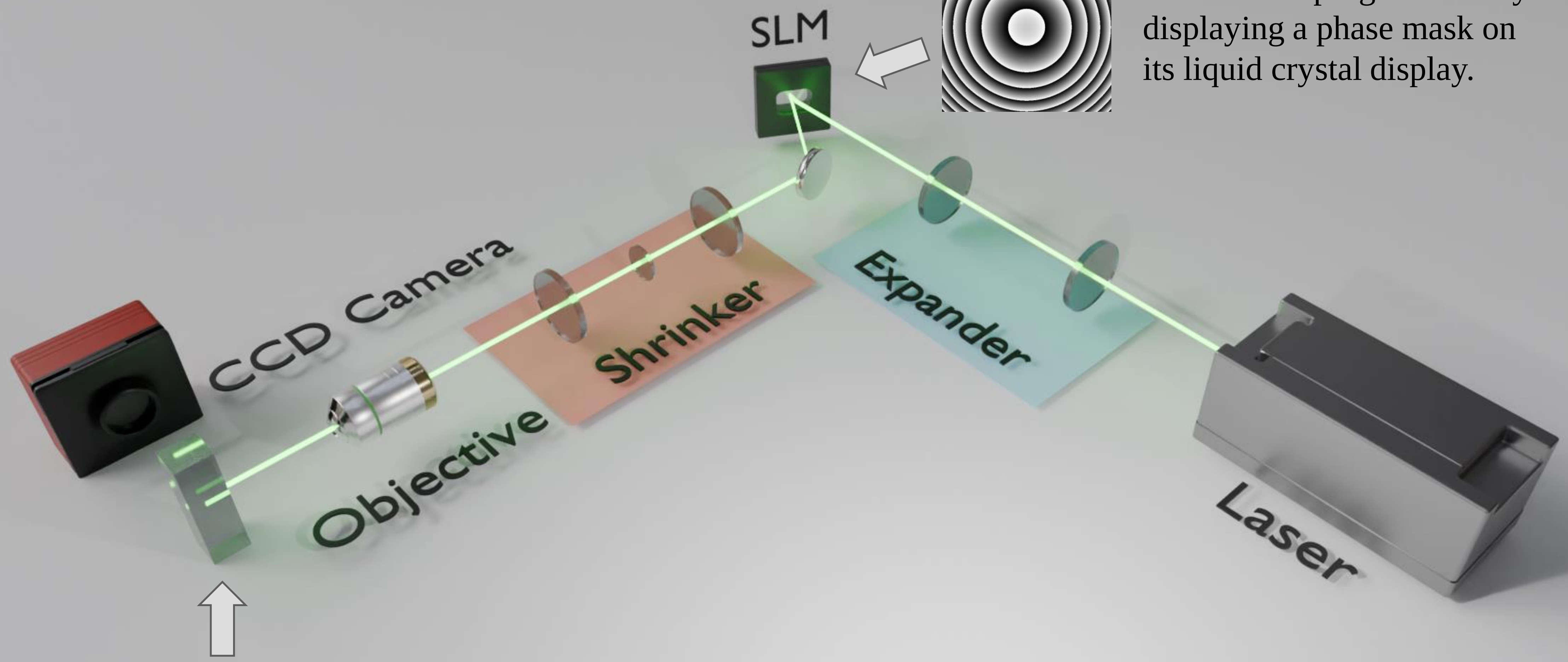
Fresnel Lens Equation

$$f(x,y) = \exp\left(\frac{-j2\pi[(x-x_0)^2+(y-y_0)^2]}{\lambda f}\right)$$

$$\Downarrow$$

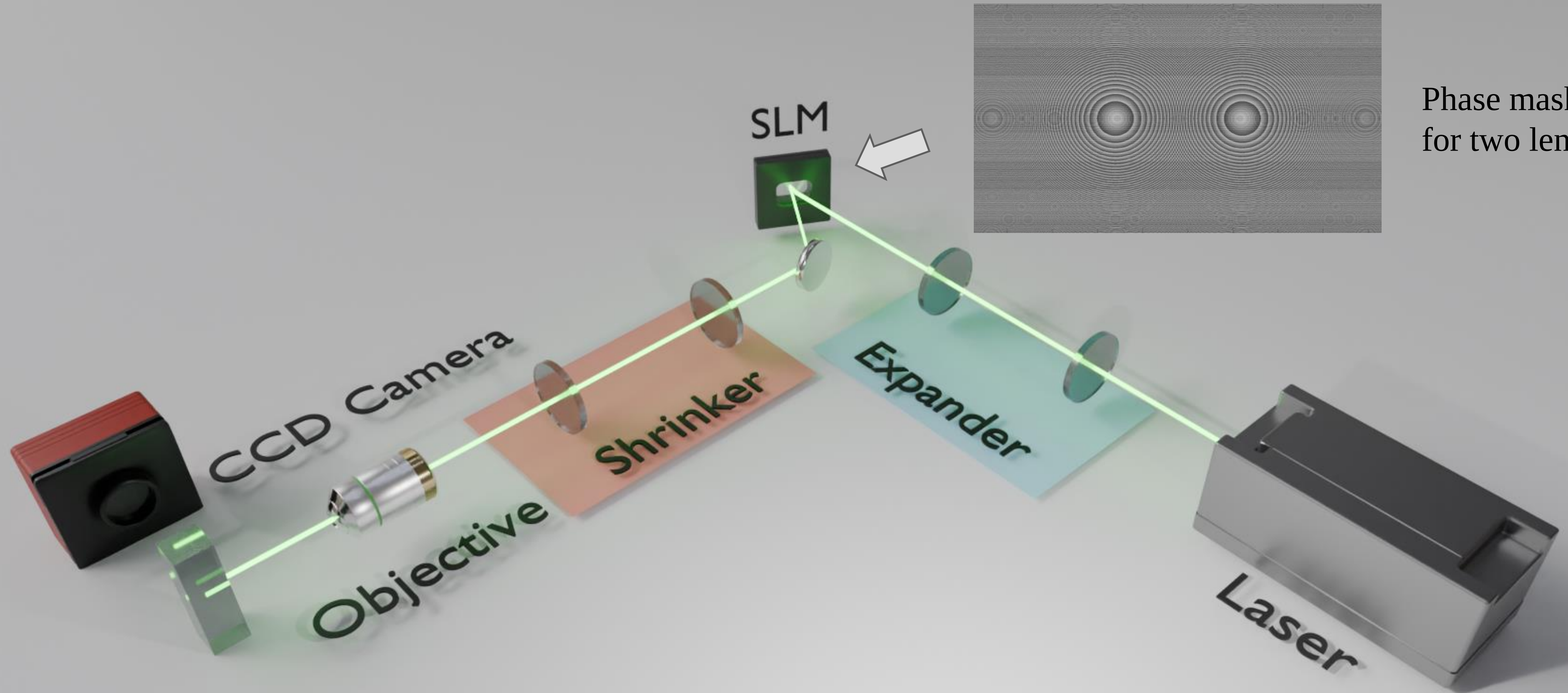
$$f(x,y) = \sum_{n=1}^{N} \exp\left(\frac{-j2\pi[(x-x_n)^2+(y-y_n)^2]}{\lambda f}\right)$$

The SLM is programmed by displaying a phase mask on its liquid crystal display.



Fluorescent nanoparticles are placed in solution to visualize the beam shape.

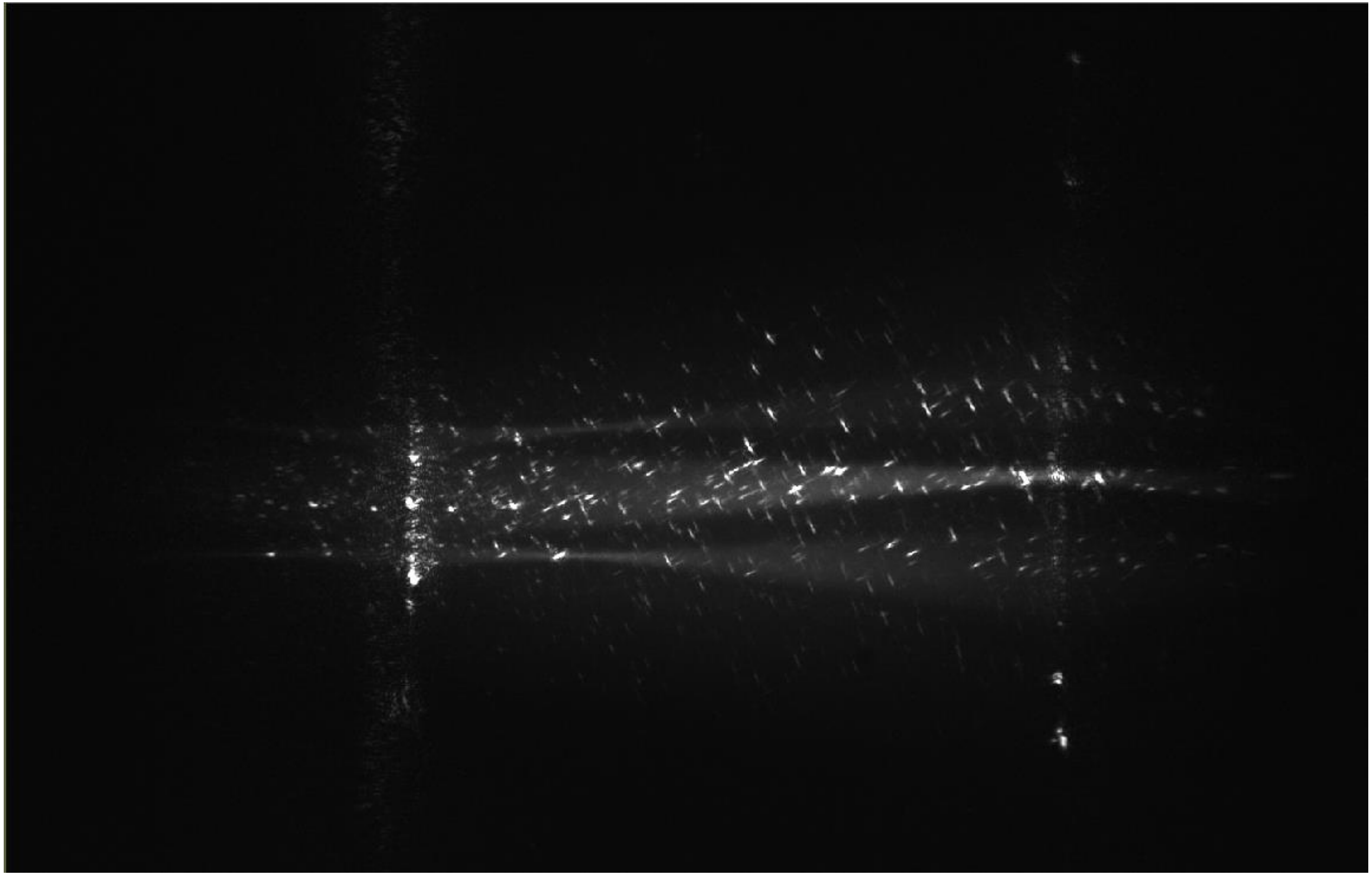
Basic HOT Setup



Phase mask
for two lenses

Basic HOT Setup

Two Trap Phase Mask Results:



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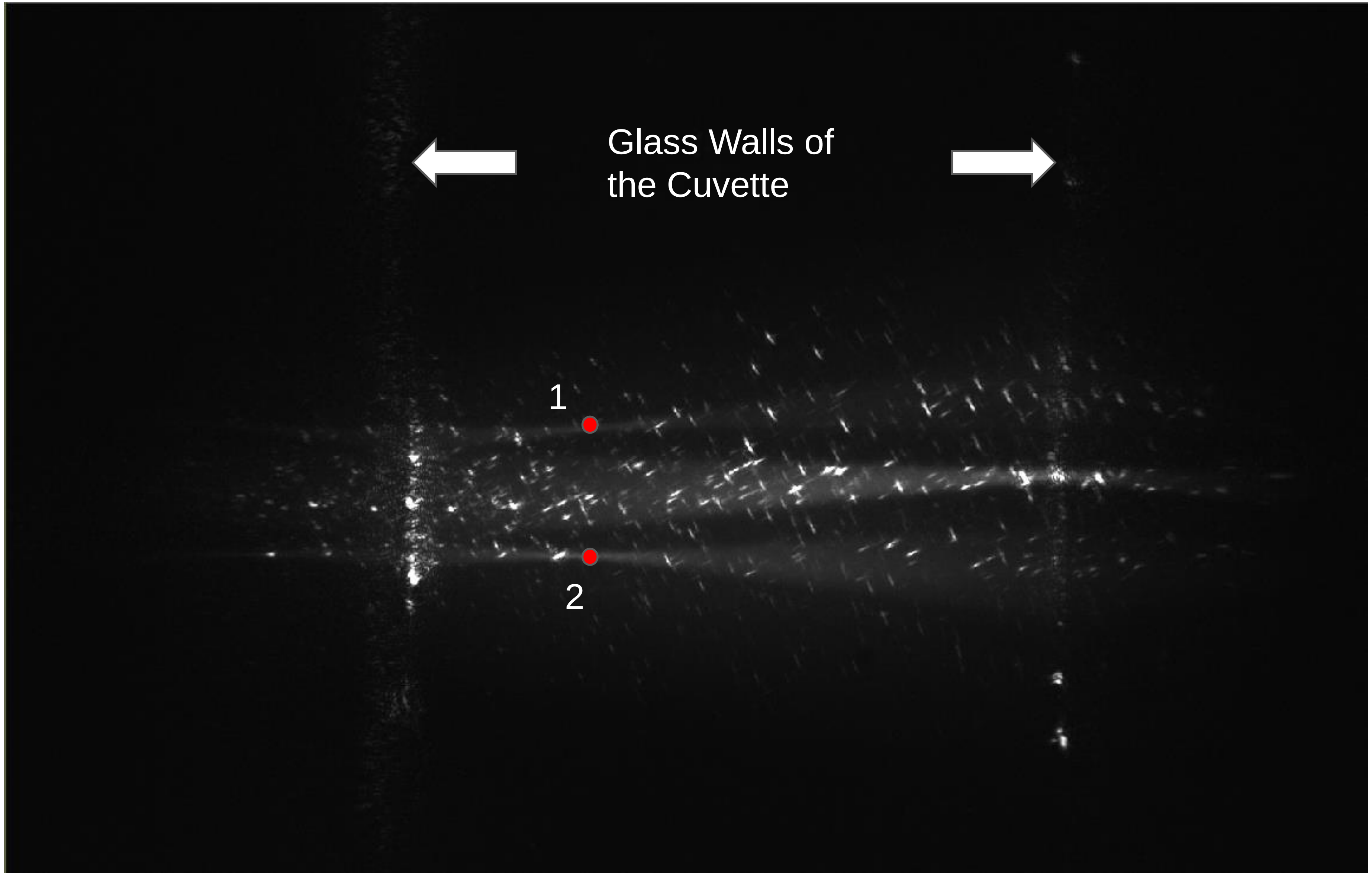

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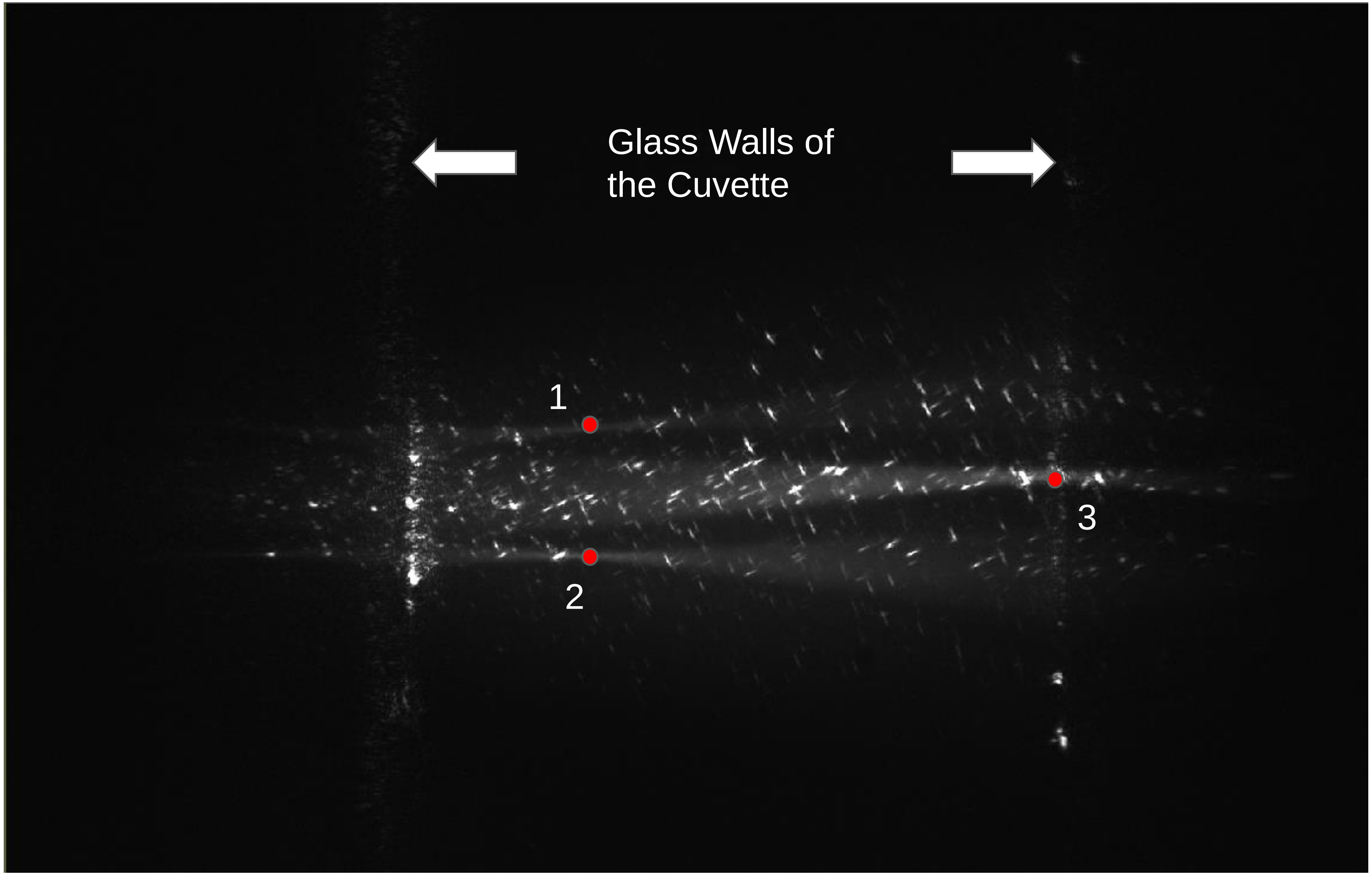
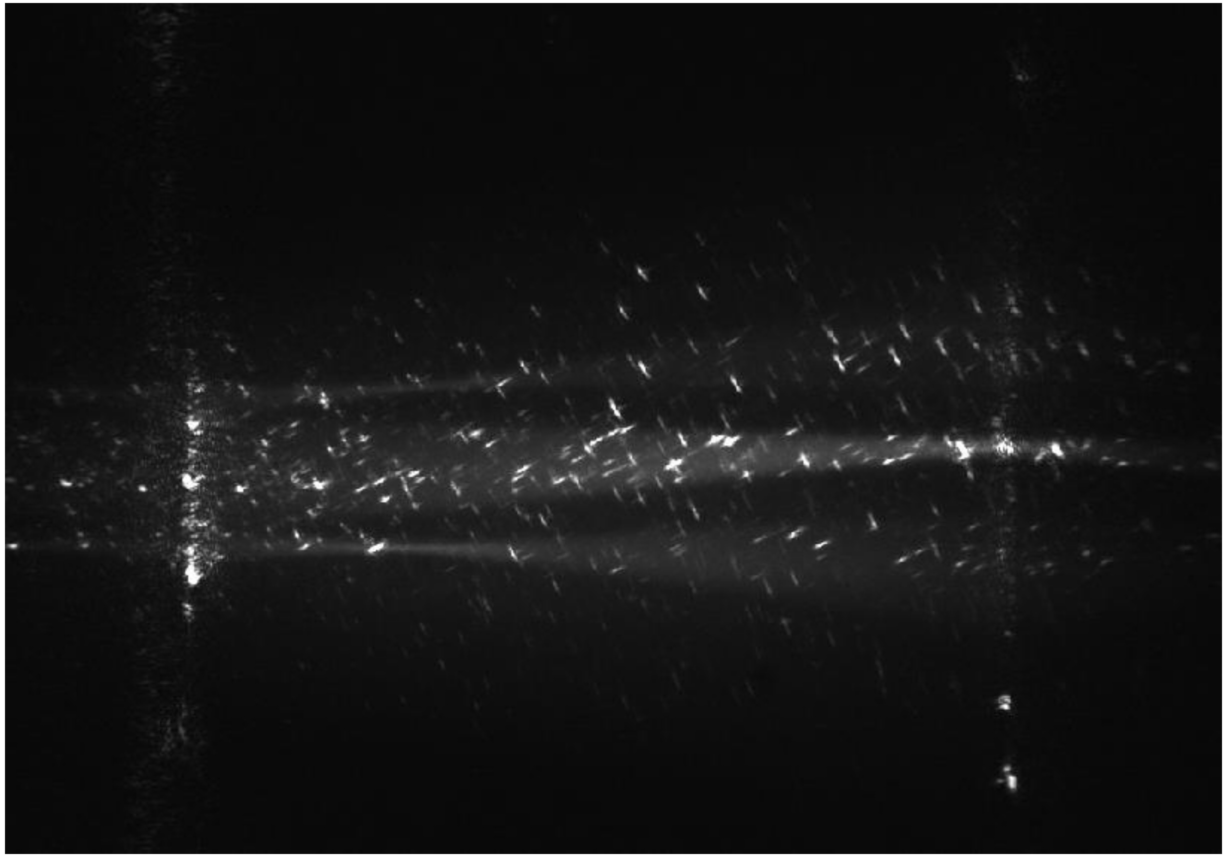


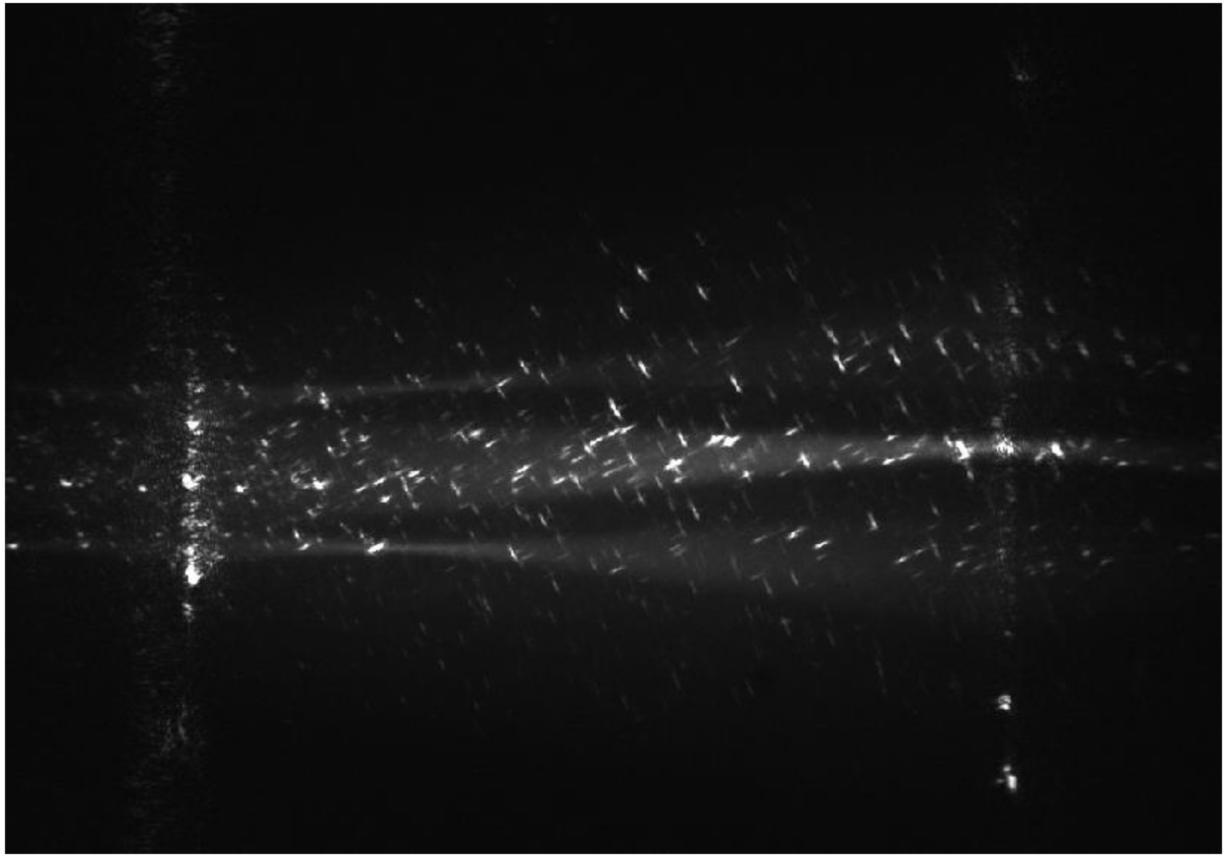
What are Ghost Holograms and Ghost Traps:



Two intentional traps with a ghost trap in between them.

- A **ghost hologram** is an unintentional structure or aberration in a generated hologram.
- A **ghost trap** is an unintentionally generated focus that is powerful enough to trap a particle just like a normal trap.

Why do Ghost Holograms Occur:

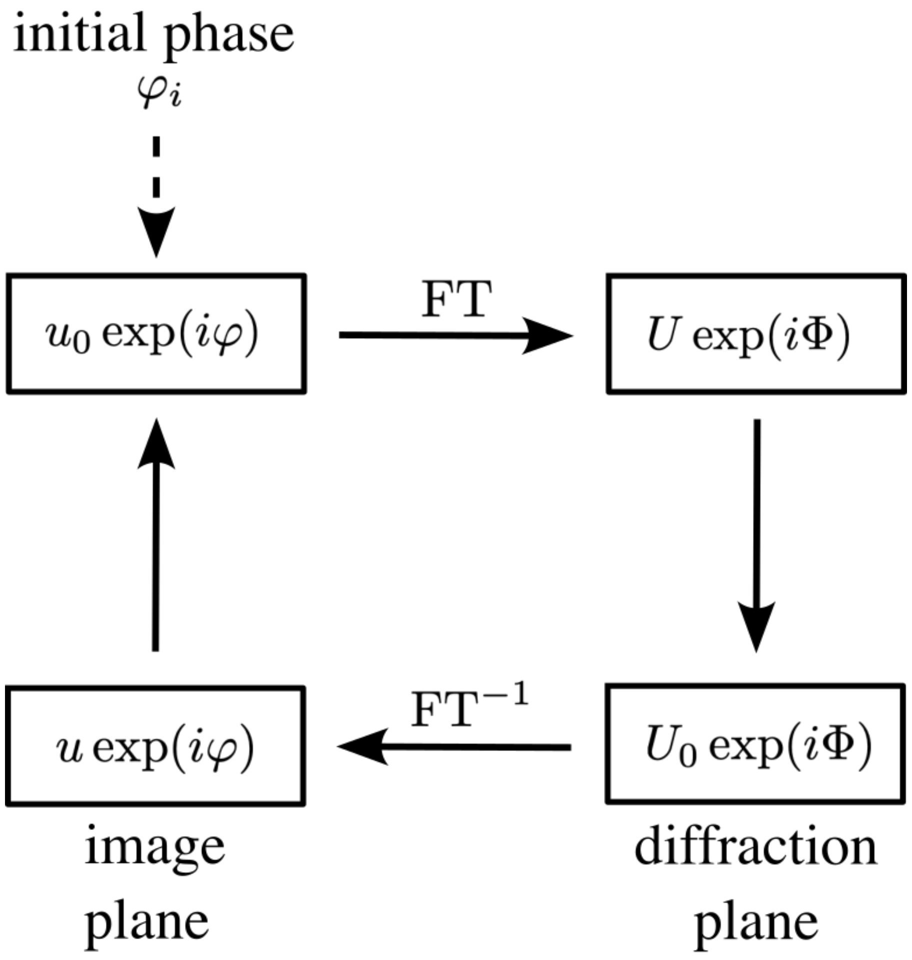


Two intentional traps with a ghost trap in between them.

- Ghost holograms can be caused by **physical limitations** of the devices utilized:
  - SLM pixel size
  - SLM effective area
  - Quantization error in phase
- Highly **dependent on the structure**. Ghost holograms are more likely to appear in highly symmetric structures (see [3] for discussion).
- Ghost holograms can also be caused by *how a phase mask is calculated*. The way the phase mask is calculated always introduces implicit or explicit assumptions.

Gerchberg-Saxton Algorithm: How it Works

1. Record the desired amplitude in the image plane and diffraction plane, u_0 and U_0 respectively (this can be done from utilizing $u_0 = \sqrt{I_0}$). u_0 and U_0 are *related by the Fourier Transform*.
2. On the first iteration, add a random phase to the image plane.
3. Take the Fourier Transform of this new complex field.
4. The transformed field now has a new magnitude (U) and phase (Φ). However, we know that we want the magnitude to be U_0. Therefore, we replace the magnitude and keep the phase
5. This process can be repeated with the inverse Fourier transform, using u_0 instead of U_0. Every repetition of this process improves the accuracy of the phase, φ.
6. Whenever "enough" iterations have been completed, take φ and convert it to a grayscale image to display.



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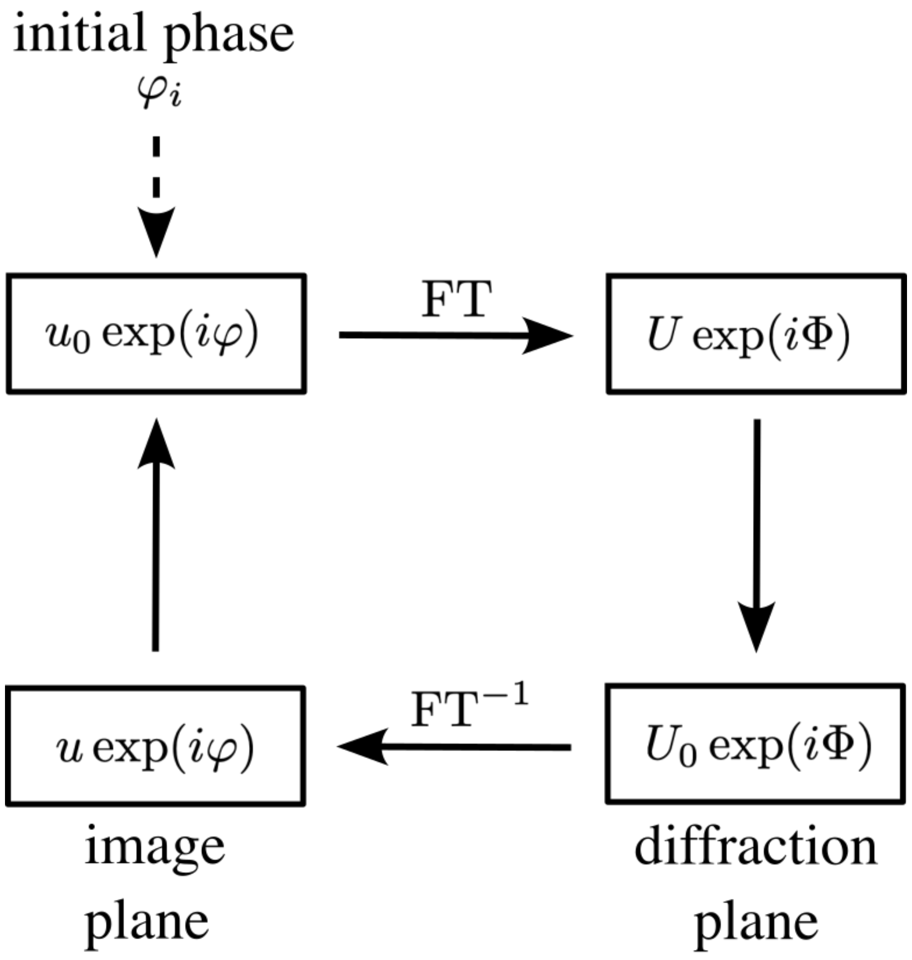

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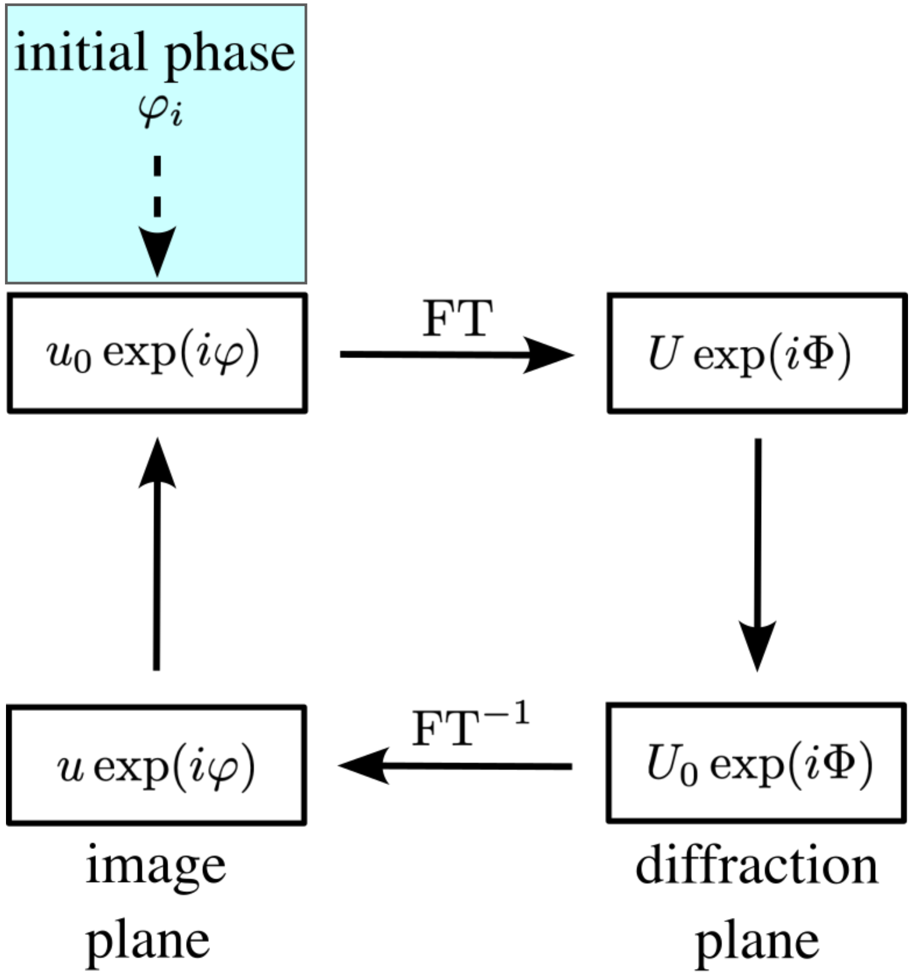

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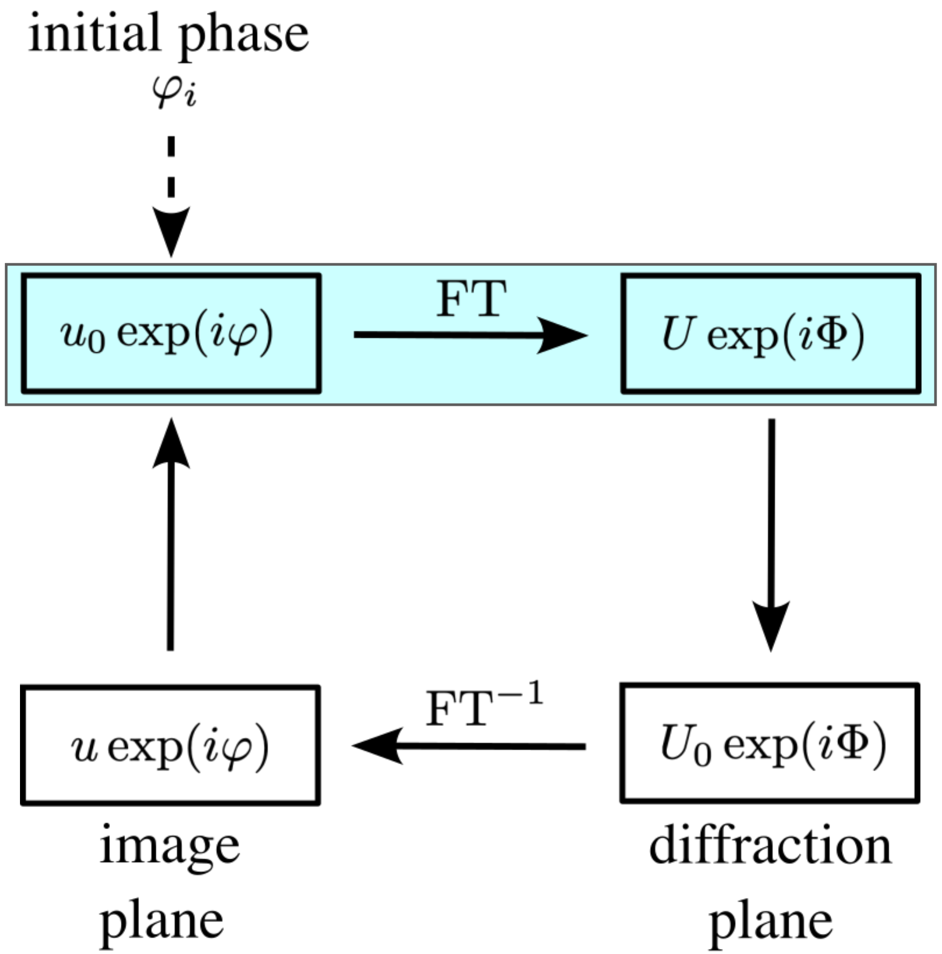

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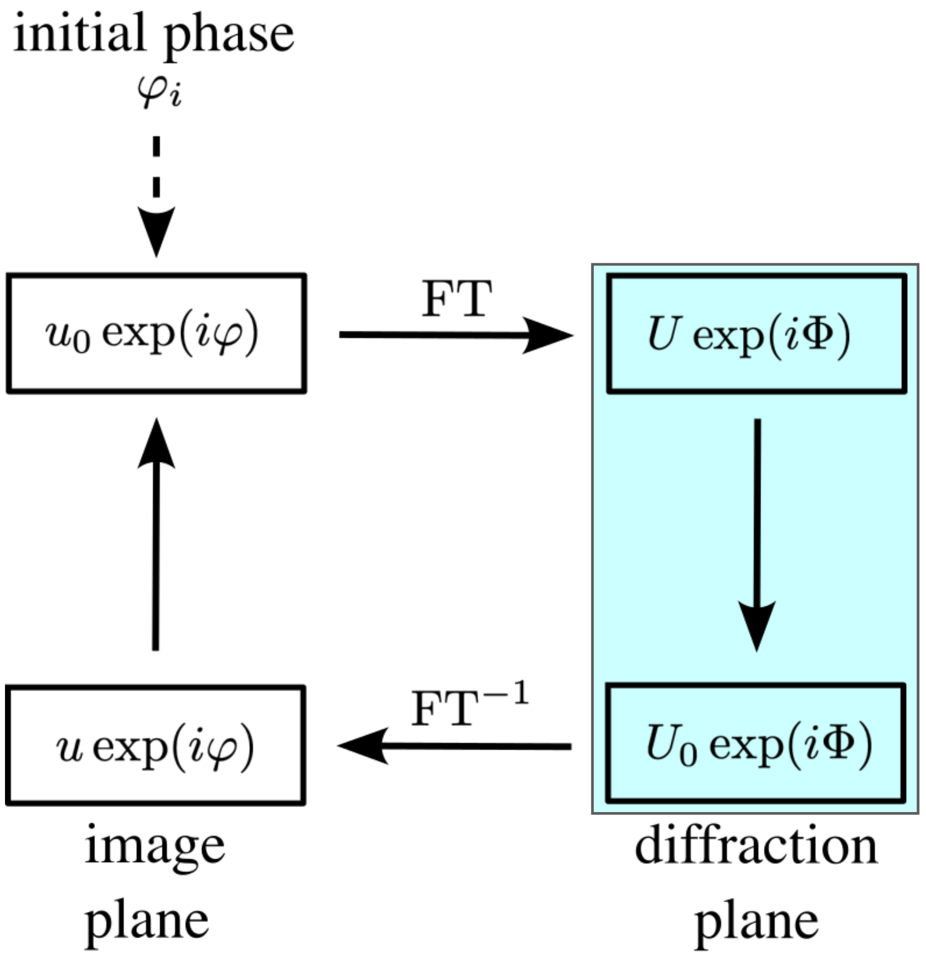

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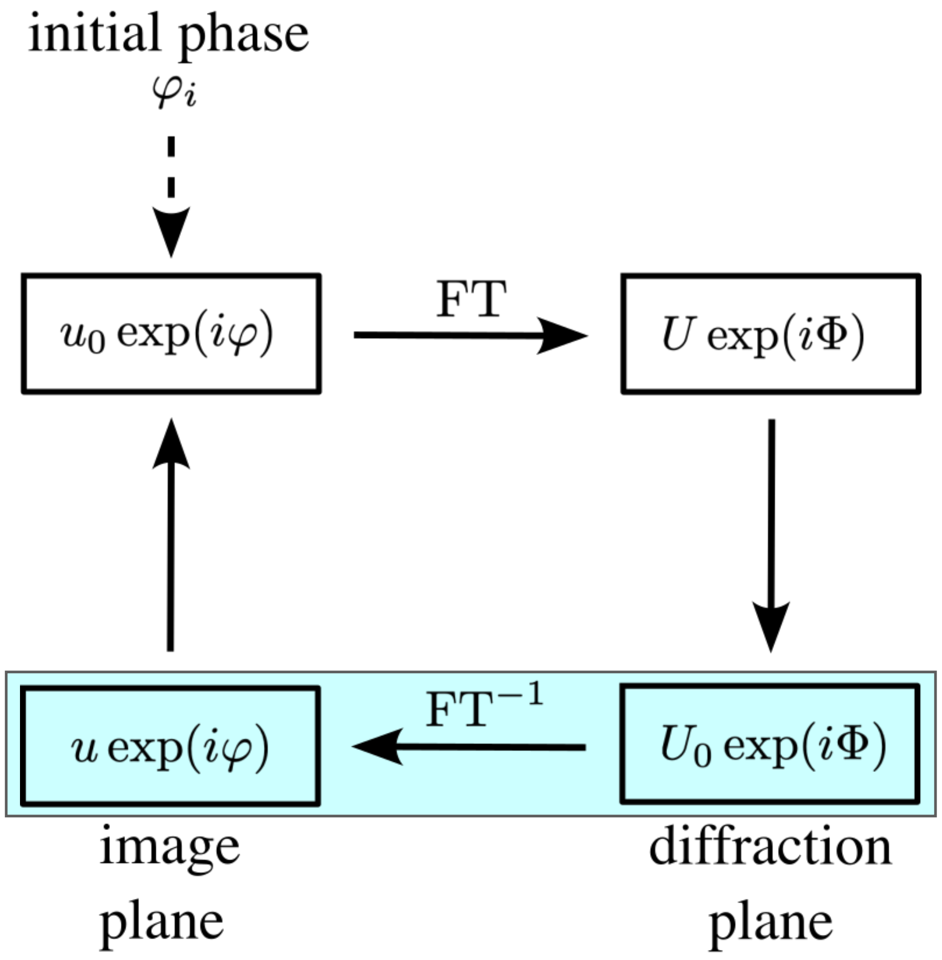

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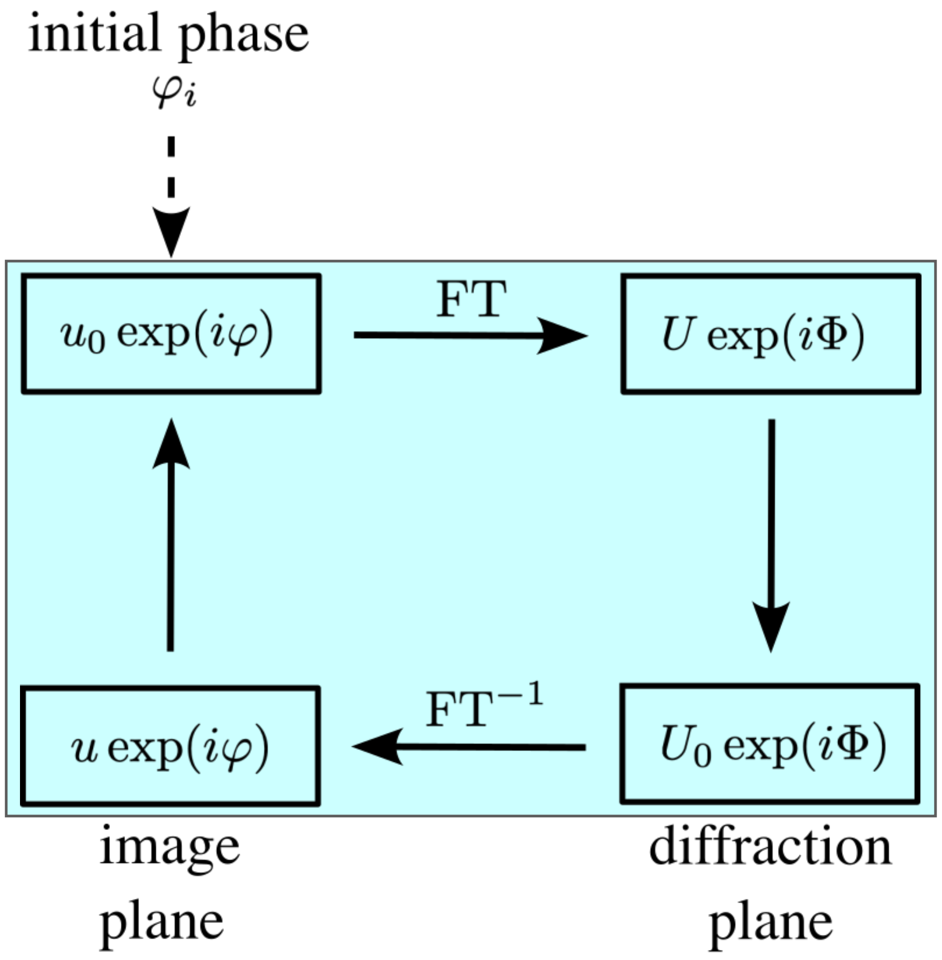
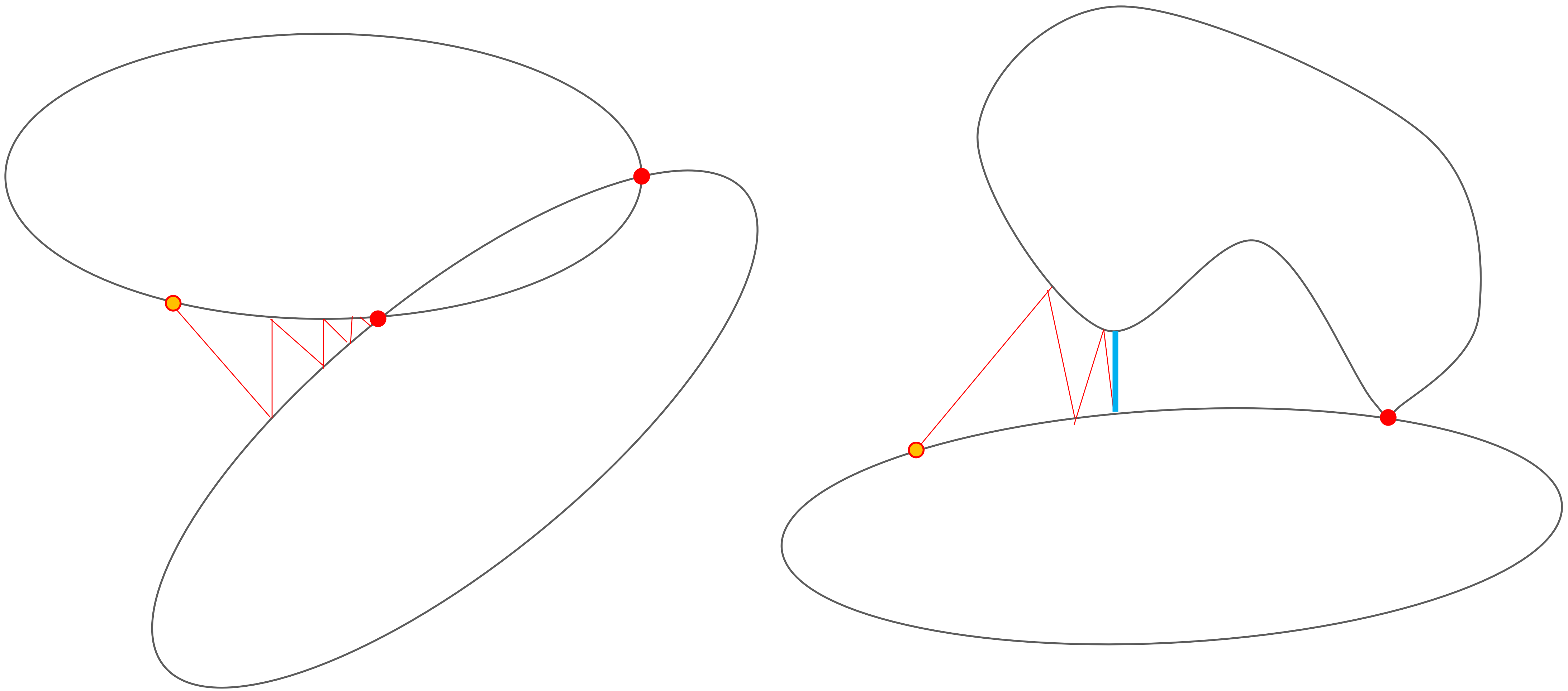
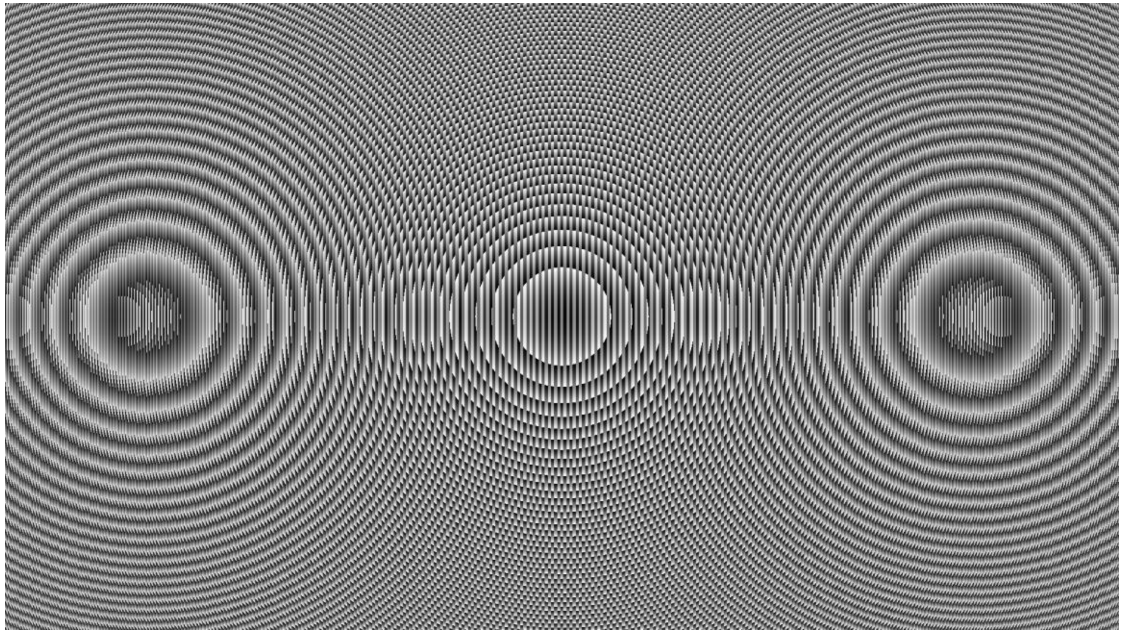
5. This process can be repeated with the inverse Fourier transform, using u_0 instead of U_0. Every repetition of this process improves the accuracy of the phase, φ.
6. Whenever "enough" iterations have been completed, take φ and convert it to a grayscale image to display.

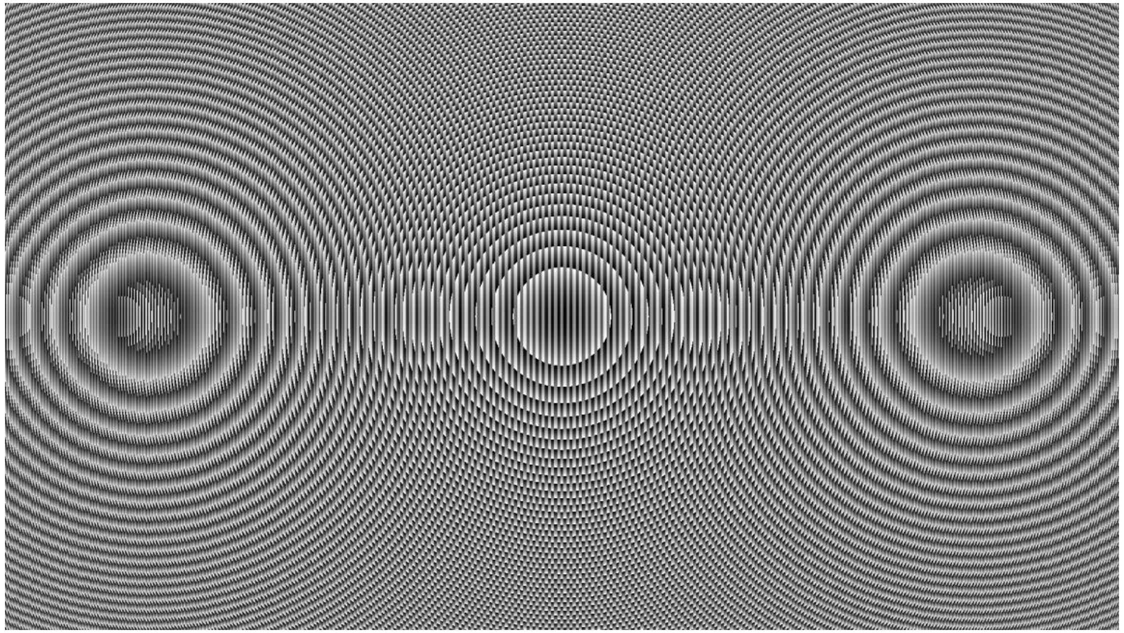


Alternating Projections and Convexity



Phase Mask Comparison:



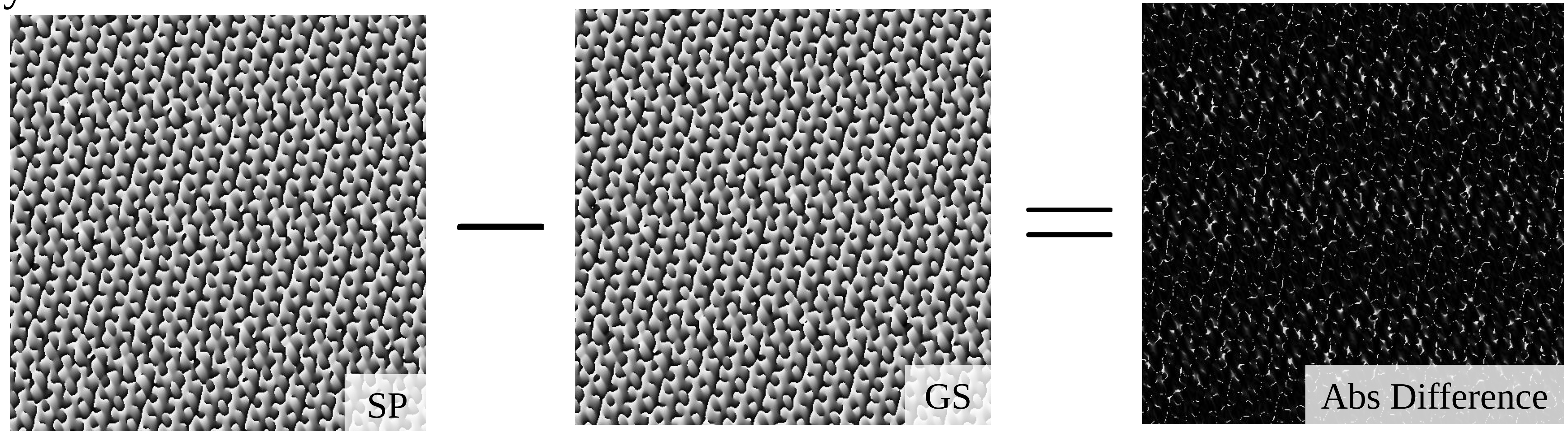


Superposition Algorithm Result with Ghost Trap

Gerchberg Saxton Algorithm Result without Ghost Trap

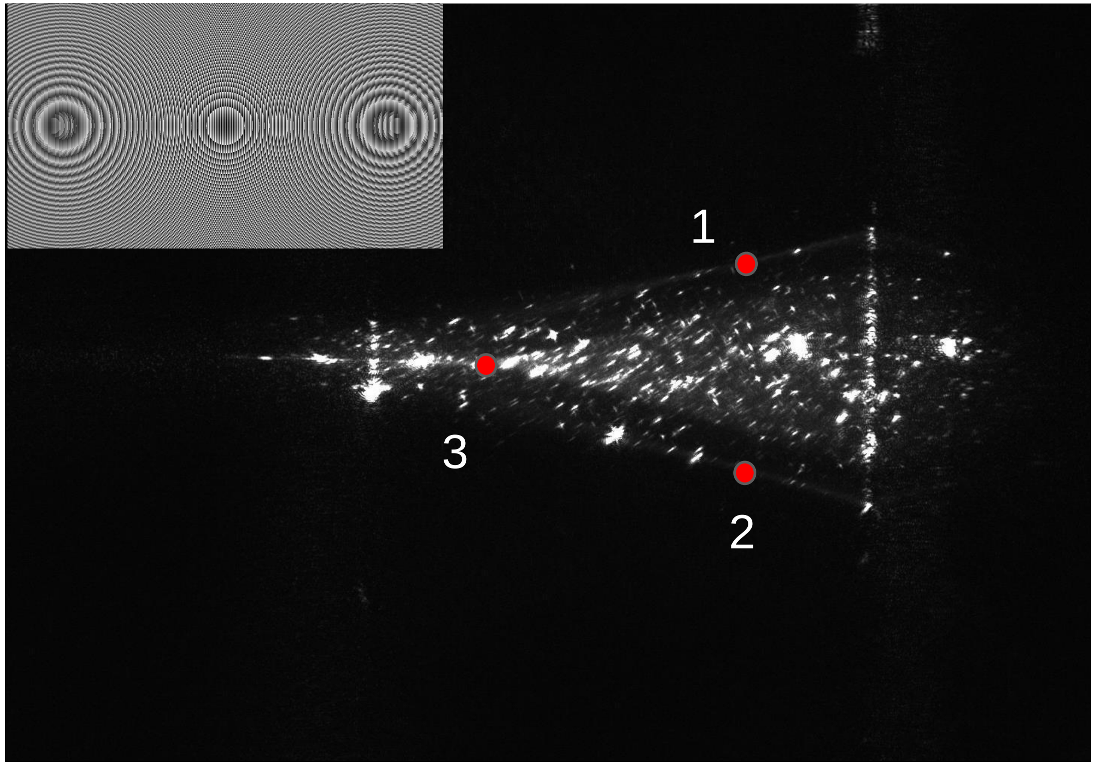
Why does the Gerchberg-Saxton Aglorithm Create Less Ghost Holograms?

- The Superposition algorithm looks at the system as individual traps, not as a complete entity.
- The GS algorithm considers all traps simultaneously. This results in “smoother” phase masks.
- The GS algorithm is iterative. Therefore, if the results are unsatisfactory, more iterations can always be done.

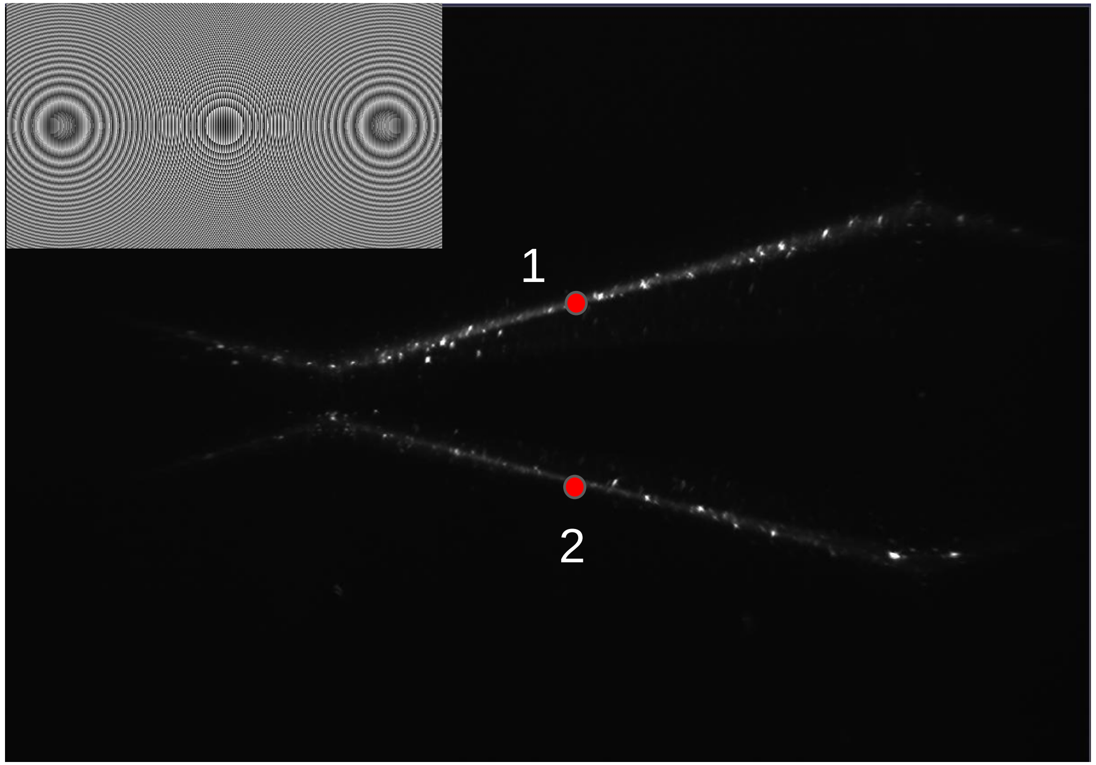


The same phase mask calculated via superposition and the GS algorithm subtracted from each other to highlight their differences. Five traps were placed randomly in a $100\ \mu m \times 100\ \mu m \times 200\ \mu m$ volume simulating a cuvette structure.

Algorithm Comparison for 2 Symmetric Foci



Superposition Algorithm Result with Ghost Trap



Gerchberg-Saxton Algorithm Result without Ghost Trap

Quantifying the Quality of Phase Masks

- A phase masks quality can be quantified into three numbers:
  - *Uniformity* – a measure of how closely distributed the intensity of each trap is.
  - *Efficiency* – fraction of the total intensity directed at the generated traps.
  - *Standard deviation* – how far on average each trap’s intensity deviates from the mean intensity.

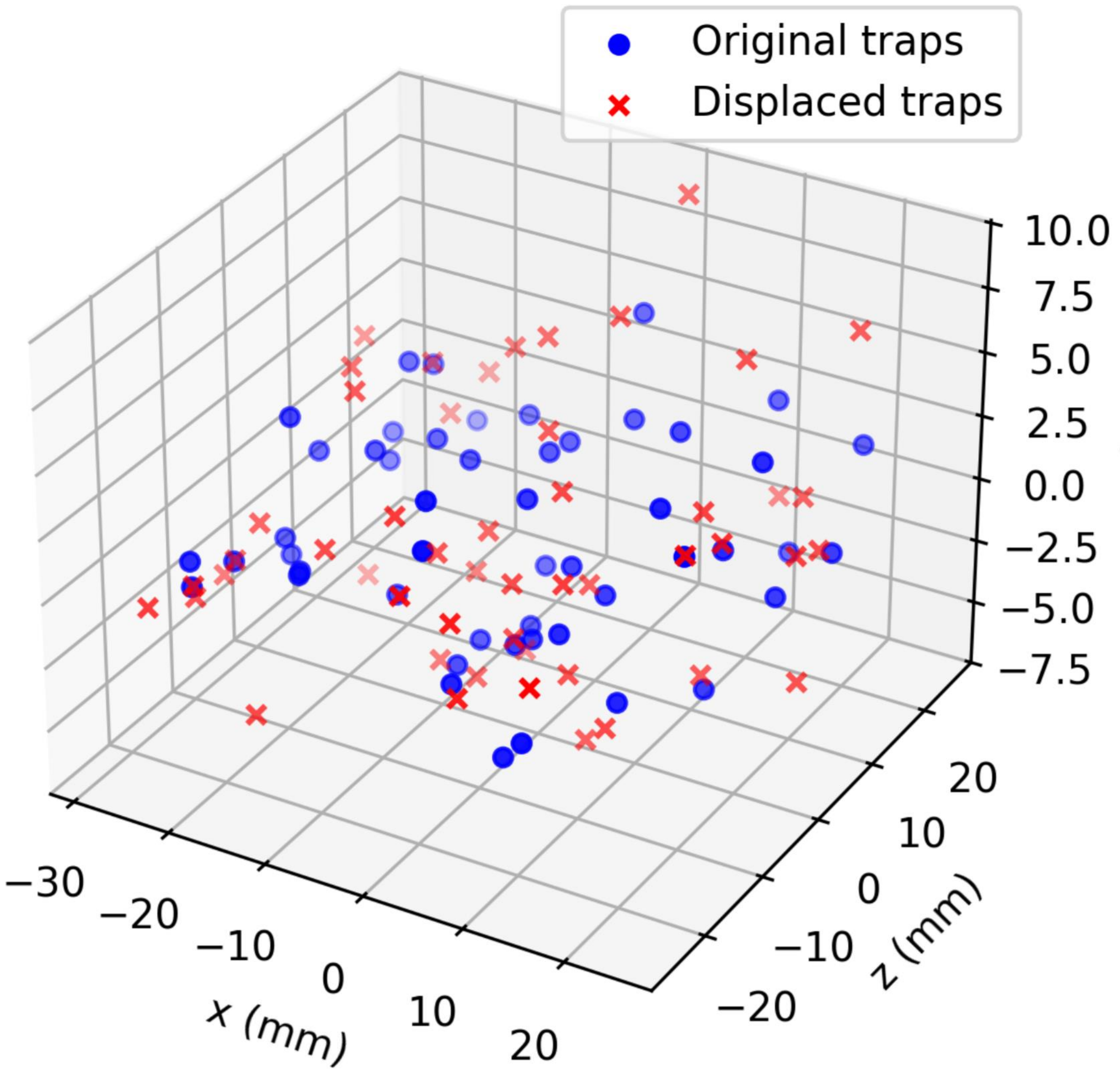
$$u = 1 - \frac{I_{\mathrm{max}} - I_{\mathrm{min}}}{I_{\mathrm{max}} + I_{\mathrm{min}}}$$

$$e = \frac{I_{\mathrm{traps}}}{I_{\mathrm{total}}}$$

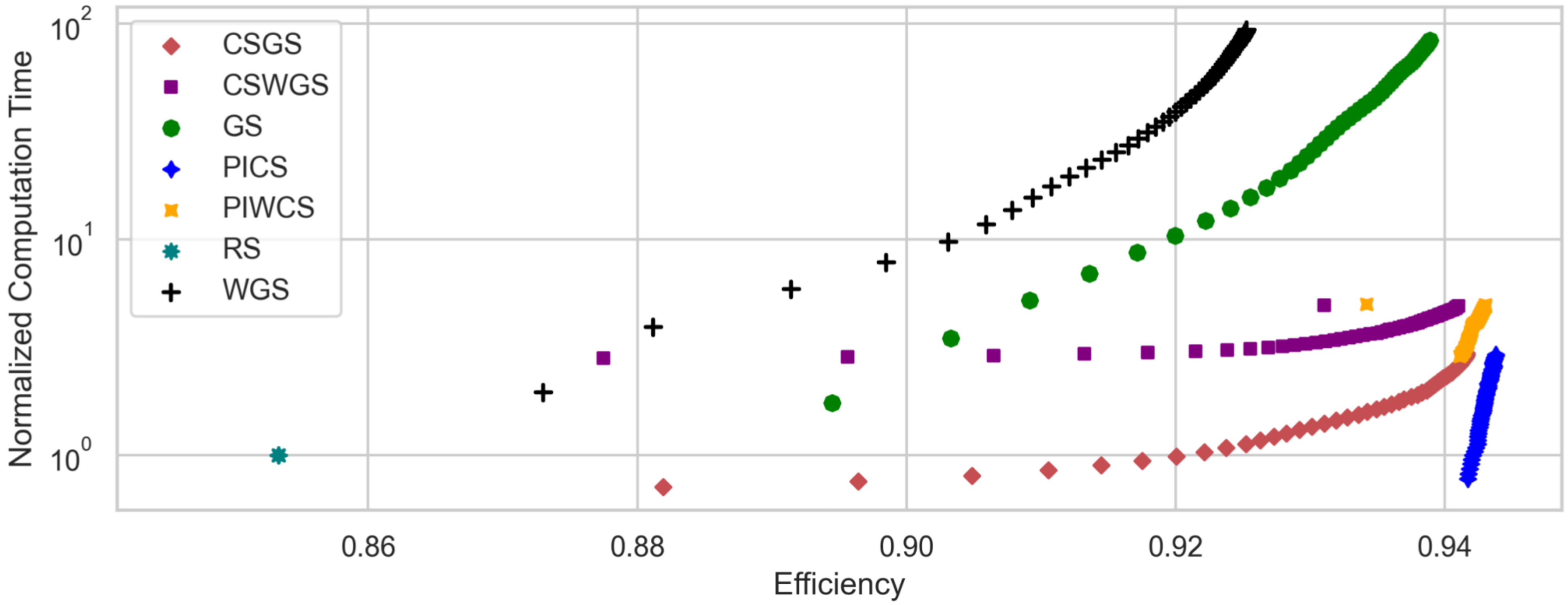
$$\sigma = \frac{\sqrt{I_{\mathrm{variance}}}}{I_{\mathrm{avg}}}$$

Dynamical phase masks

- To create a time-dependent structure, the particles array must be dynamically rearranged.
- Being holographic in nature, updating the positions of the trap sites requires the computation from scratch of a new phase mask.
- At each step the GS algorithm must iterate till convergence is achieved.
- We have developed a new algorithm that exploits the current state of the array to compute the new configurations.



Phase-Induced Compressive Sensing (PICS) GS



Summary

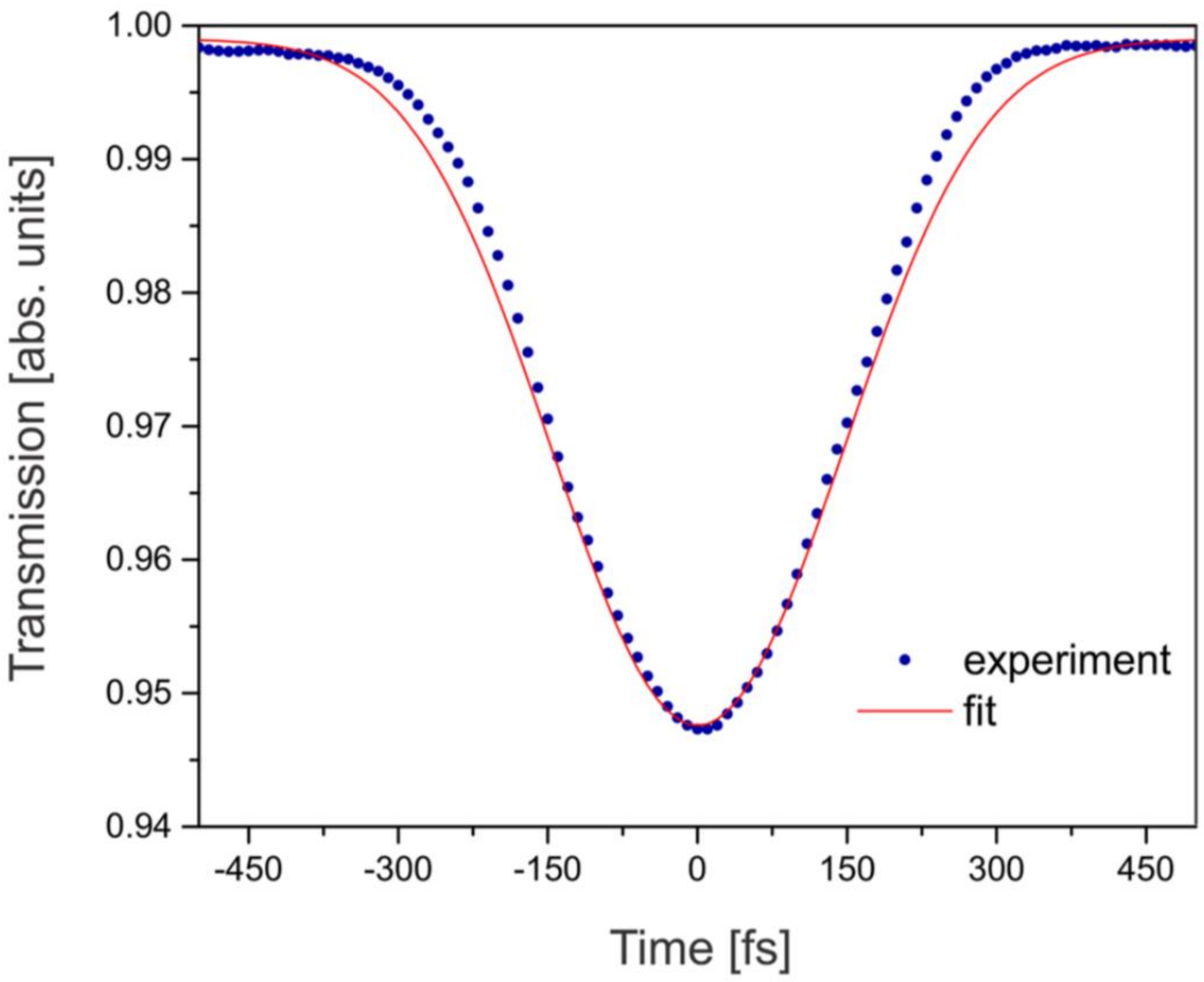
- We developed a novel algorithm to compute phase masks to assemble and reconfigure particle arrays by means of optical force.

- The proposed method outperforms competing strategies in terms of efficiency and computational cost.

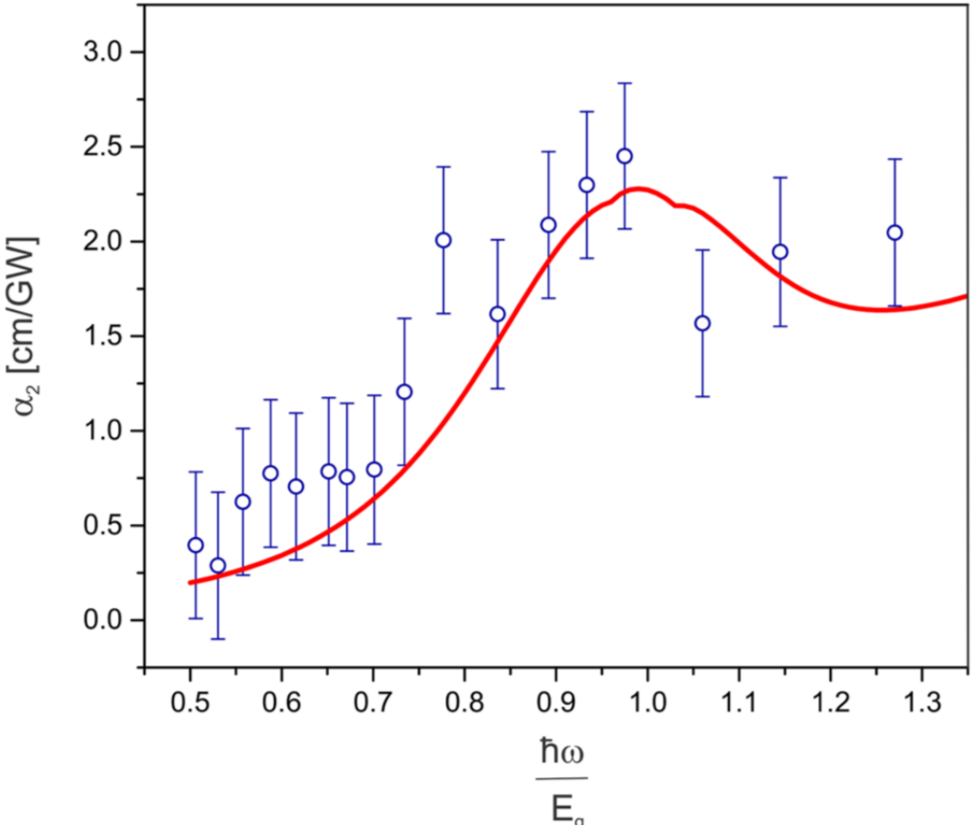
Theory of nondegenerate 2-photon absorption in indirect bandgap semiconductors*

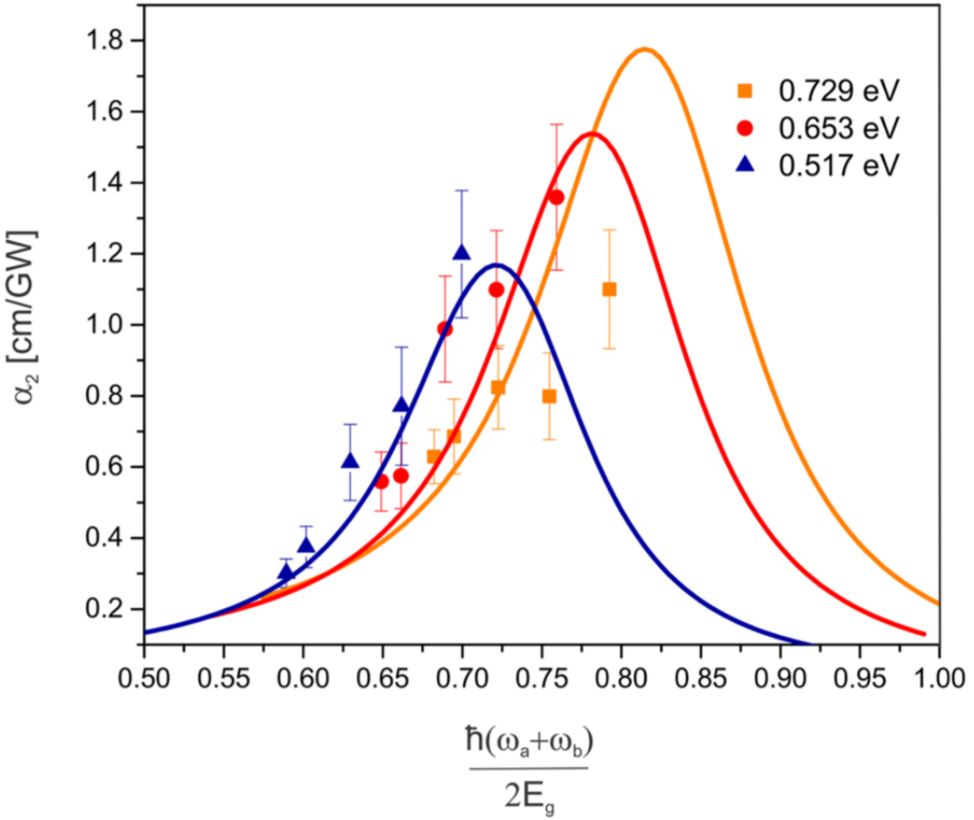
*David Ziemkiewicz, David Knez, Evan P. Garcia, Sylwia Zielińska-Raczyńska, Gerard Czajkowski, Alessandro Salandrino, Sergey S. Kharintsev, Aleksei I. Noskov, Eric O. Potma, Dmitry A. Fishman; Two-photon absorption in silicon using the real density matrix approach. ***J. Chem. Phys***. 7 October 2024; 161 (14): 144117.

Modeling pump-probe experiments in Si



Nonlinear Absorption – Theory vs Experiment





Summary

- We developed a theoretical model of nondegenerate 2-photon absorption in indirect bandgap semiconductor.
- The theory is based on the real density matrix approach and relies on a number of known measured hyperparameters.
- The model is in good agreement with the pump-probe experiment conducted at UC Irvine on Si samples.