

# Nano-patterning of surfaces by ion sputtering: Numerical study of the Kuramoto-Sivashinsky Equation

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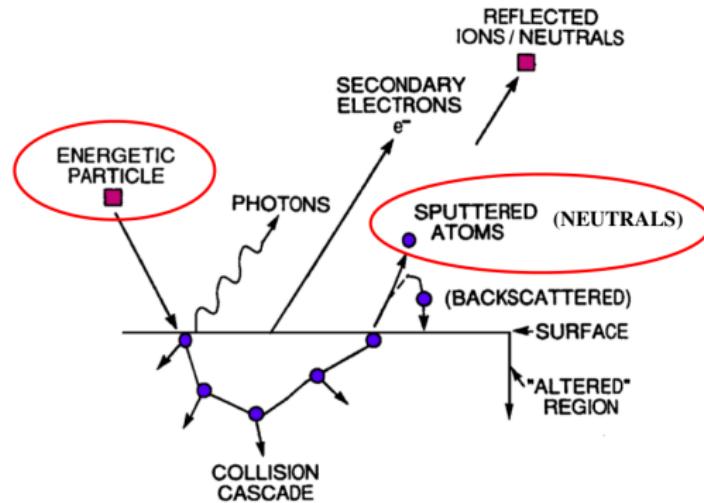
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# Overview

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# Sputtering

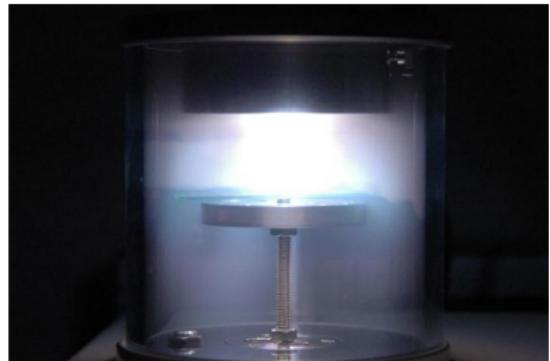
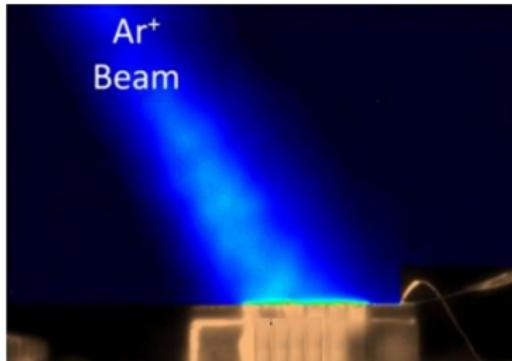
- Physics of Sputtering



- Applications: thin film deposition, micromachining, pattern etching for the fabrication of integrated circuits and chemical analysis

# Sputtering

- Ion Beam x Glow Discharge Sputtering



- Sputtering yields:

- $E < 1\text{KeV} : S = \frac{3\alpha}{\pi^2} \frac{M_1 M_2}{(M_1 + M_2)^2} \frac{E_i}{E_L}$

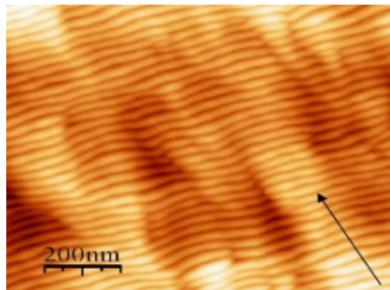
- $E > 1\text{KeV} : S \sim (Z_1 Z_2) \frac{M_1}{M_1 + M_2} S_n(E)$

# Experimental results

- Two main classes of surface morphology by ion sputtering according to experimental results:
  - ▶ Periodic ripples on the surface
  - ▶ Rough eroded surfaces
- Experimental data and parameters:
  - i) Ripple formation
    - ▶ Angle of incidence
    - ▶ Temperature dependence
    - ▶ Ion energy
    - ▶ System chemistry
  - ii) Kinetic roughening
    - ▶ Temperature dependence
    - ▶ Ion energy

# Experimental results

Rippled patterns formed by ion sputtering have close relatives in nature:



(a)  $\text{Ar}^+$  sputtering on Si



(b) Namib desert



(c) Cloudy sky



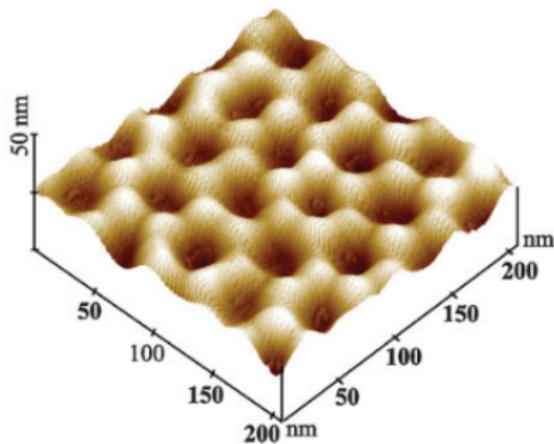
(d) Ripples on the water

# Experimental results

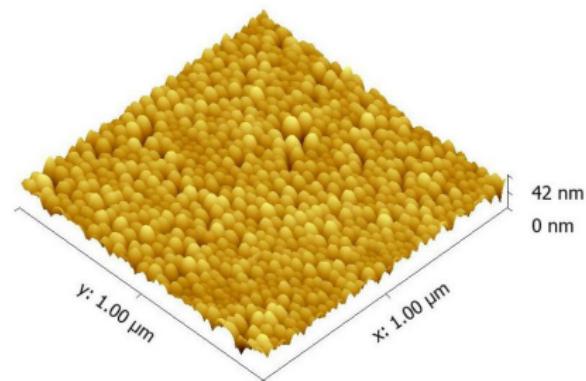
- Bradley and Harper theory (HP): theoretical approach describing the process of ripple formation on amorphous substrates
  - ▶ Ripples are a result of a surface instability caused by the curvature dependence of the sputter yield.
  - ▶ Wavelength and orientation prediction in agreement with numerous experimental results
  - ▶ Cannot account for surface roughening
- A nonlinear theory is required to model the time evolution of ion-sputtered surfaces and predict their morphology:
  - ▶ Short time scales: describe the development of a periodic ripple structure
  - ▶ Large time scales: the predicted surface morphology should be rough or dominated by new ripples

# Experimental results

Experimental data and imaging (AFM):



3D image of hexagonally ordered nanoholes on a Ge surface ( $\text{Ga}^+$  FIB bombardment) obtained from AFM data



AFM image of a nanodot pattern created by  $\text{Ar}^+$  sputtering on  $\text{GaSb}(001)$

# Theoretical approaches

## Kardar-Parisi-Zhang equation (KPZ):

$$\partial_t h = \nu \nabla^2 h + \frac{\lambda}{2} (\nabla h)^2 + \eta$$

- Nonlinear stochastic partial differential equation
- Describe the time evolution of a nonequilibrium interface
- $\eta(\vec{x}, t)$ : noise, reflects the random fluctuations in the process
- EW equation,  $\lambda = 0$ : equilibrium fluctuations of an interface which tries to minimize its area under the influence of external noise
- Scaling properties may change drastically due to anisotropy (AKPZ)

# Theoretical approaches

## Kuramoto-Sivashinsky equation (KS):

$$\partial_t h = -|\nu| \nabla^2 h - K \nabla^4 h + \frac{\lambda}{2} (\nabla h)^2$$

- Deterministic equation, originally proposed to describe chemical waves and flame fronts
- Chaotic solution rises from its unstable and highly nonlinear character
- 1D: for long time and length scales, the surface described by the KS equation is similar to the one described by the KPZ equation; for short time scale solution, the morphology is reminiscent of ripples
- 2D: contradictory computer simulations

# Numerical scheme

## Governing equation:

- An anisotropic damped Kuramoto-Sivashinsky equation was adopted for the present study
- One simplified dimensionless form of such equation can be described as follows:

$$\begin{aligned}\frac{\partial \bar{h}}{\partial \tau} = & -\bar{\alpha} \bar{h} + \bar{\mu} \frac{\partial^2 \bar{h}}{\partial X^2} - c^2 \frac{\partial^2 \bar{h}}{\partial Y^2} + \bar{\nu}_x \left( \frac{\partial \bar{h}}{\partial X} \right)^2 - c^3 \left( \frac{\partial \bar{h}}{\partial Y} \right)^2 - D_{XX} \frac{\partial^4 \bar{h}}{\partial X^4} \\ & + D_{XY} \frac{\partial^4 \bar{h}}{\partial X^2 \partial Y^2} + c^2 \frac{\partial^4 \bar{h}}{\partial Y^4} - \bar{K} \left( \frac{\partial^4 \bar{h}}{\partial X^4} + 2 \frac{\partial^4 \bar{h}}{\partial X^2 \partial Y^2} + \frac{\partial^4 \bar{h}}{\partial Y^4} \right)\end{aligned}$$

# Numerical scheme

## Semi-implicit scheme:

- We propose the following second order in time Cranck-Nicolson semi-implicit scheme for solving the target equation with  $a_\mu = 4$ , high temperatures and  $\theta < 65.3^\circ$ :

$$\frac{\bar{h}^{n+1} - \bar{h}^n}{\Delta\tau} = \Lambda_X \frac{\bar{h}^{n+1} + \bar{h}^n}{2} + \Lambda_Y \frac{\bar{h}^{n+1} + \bar{h}^n}{2} + f^{n+1/2}$$

- Including the factor 1/2 into the operators  $\Lambda_X$  and  $\Lambda_Y$ , they may be defined as follows:

$$\Lambda_X = \frac{1}{2} \left[ -\frac{\bar{\alpha}}{2} - (D_{XX} + K) \frac{\partial^4}{\partial X^4} \right]$$

$$\Lambda_Y = \frac{1}{2} \left[ -\frac{\bar{\alpha}}{2} - K \frac{\partial^4}{\partial Y^4} \right]$$

# Numerical scheme

## Internal iterations:

- Since the operators  $\Lambda_X^{n+1/2}$ ,  $\Lambda_Y^{n+1/2}$  and the function  $f^{n+1/2}$  contain terms in the new stage we do internal iterations at each time step according to:

$$\frac{\bar{h}^{n,m+1} - \bar{h}^n}{\Delta\tau} = \Lambda_X (\bar{h}^{n,m+1} + \bar{h}^n) + \Lambda_Y (\bar{h}^{n,m+1} + \bar{h}^n) + f^{n+1/2}$$

where the superscript  $(n, m + 1)$  identifies the “new” iteration. The iterations proceed until the following criteria is satisfied:

$$\frac{\max |\bar{h}^{n,m+1} - \bar{h}^{n,m}|}{\max |\bar{h}^{n,m+1}|} < \delta$$

# Numerical scheme

## The splitting scheme:

- The splitting of the target equation is made according to the Douglas second scheme (also known as “scheme of stabilizing correction”). It is designed as follows:

$$\frac{\tilde{h} - \bar{h}^n}{\Delta\tau} = \Lambda_X \tilde{\bar{h}} + \Lambda_Y \bar{h}^n + f^{n+1/2} + (\Lambda_X + \Lambda_Y) \bar{h}^n$$

$$\frac{\bar{h}^{n,m+1} - \tilde{h}}{\Delta\tau} = \Lambda_Y (\bar{h}^{n,m+1} - \bar{h}^n)$$

## Linear stability analysis

- In Fourier transform, for small perturbations, the governing equation may be written as:

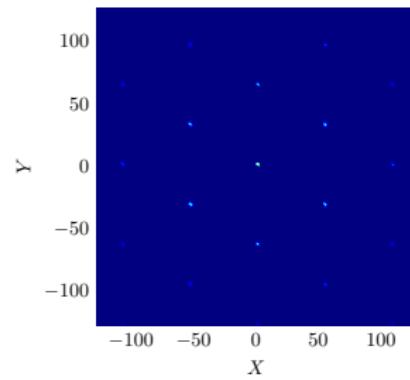
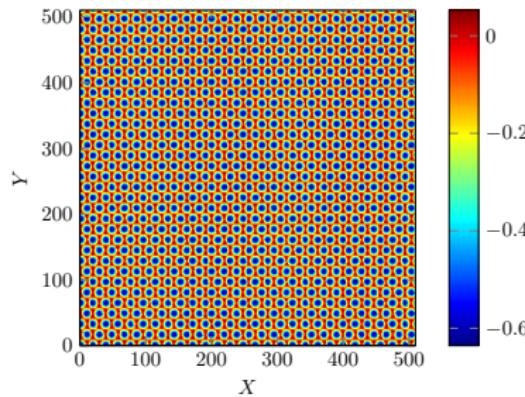
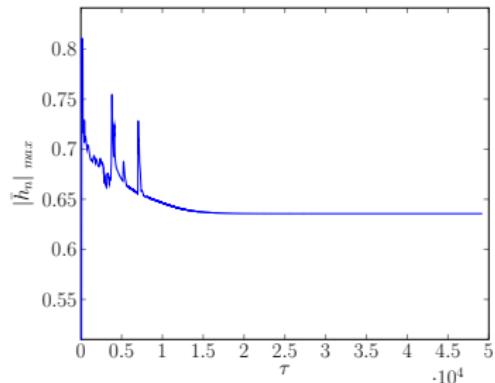
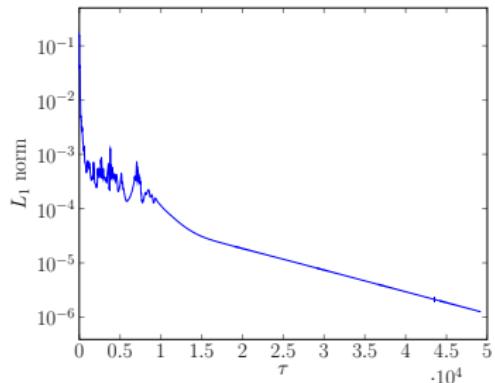
$$\bar{h}_p(\vec{q}) = \bar{h}_o e^{i(q_x X + q_y Y)} e^{\sigma_\tau}$$

$$\begin{aligned}\sigma_\tau \bar{h}_p(\vec{q}) &= [ -\bar{\alpha} + (-\bar{\mu}q_x^2) + (-\bar{\nu}q_y^2) - D_{XX}q_x^4 + D_{XY}q_x^2q_y^2 \\ &\quad + D_{YY}q_y^4 - \bar{K}(q_x^2 + q_y^2)^2 ] \bar{h}_p(\vec{q})\end{aligned}$$

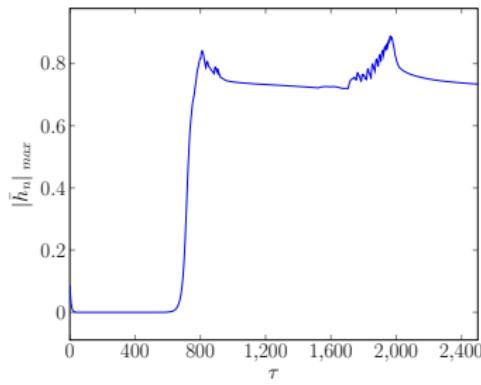
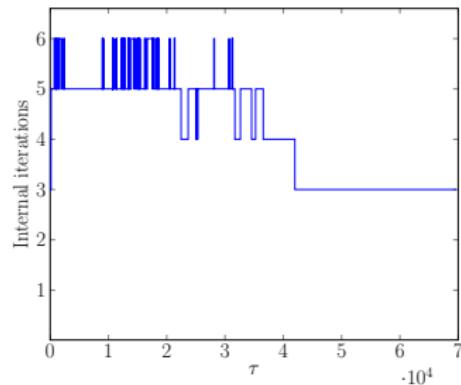
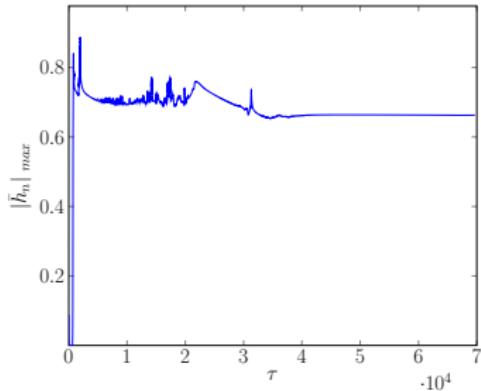
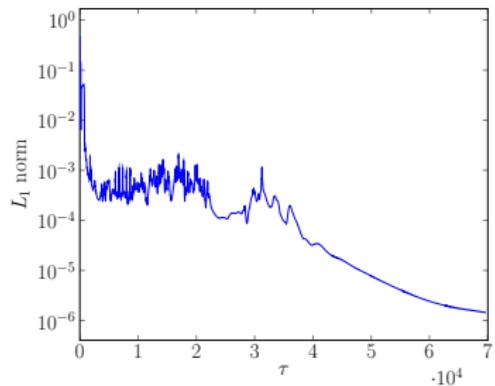
- The anisotropic coefficients  $D_{XX}$ ,  $D_{XY}$  and  $D_{YY}$  can be hidden for an easier manipulation of the equation. Besides, we may consider  $q^2 = q_x^2 + q_y^2$ .

$$\sigma_\tau = \epsilon - \bar{K}(q^2 - q_c^2)^2 - (|\bar{\mu}| - |\bar{\nu}|)q_y^2$$

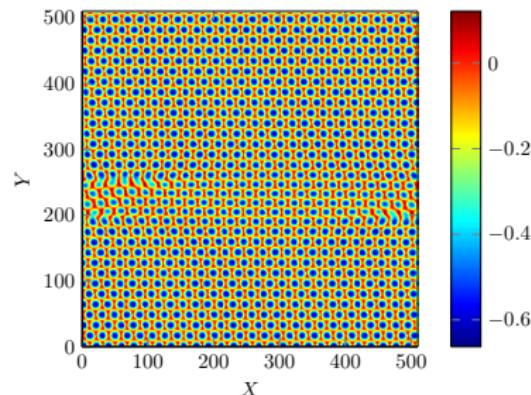
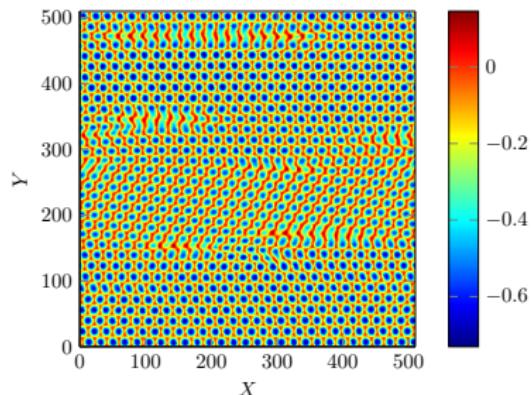
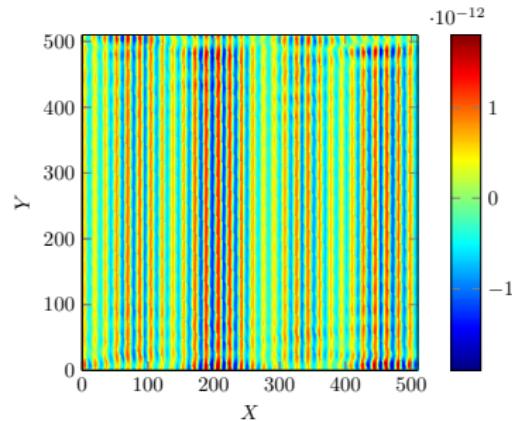
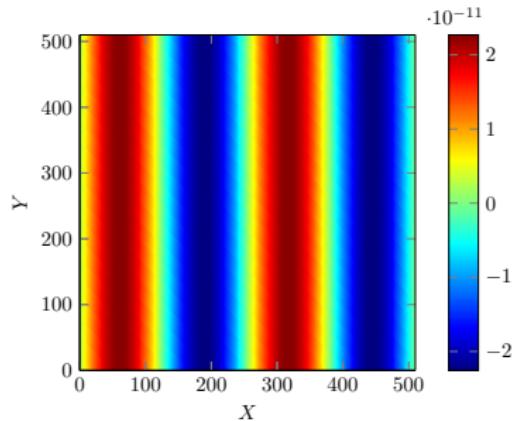
# Random initial case



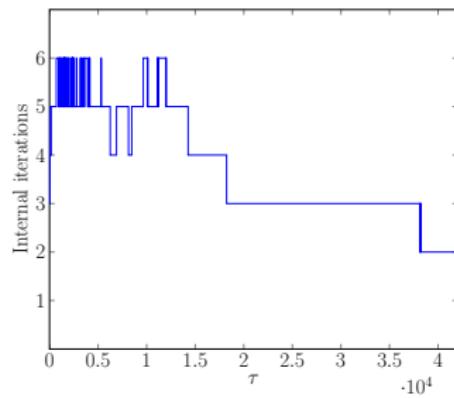
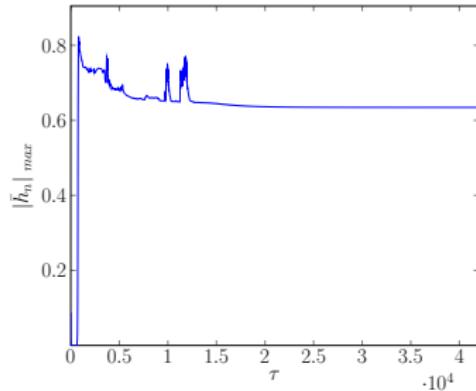
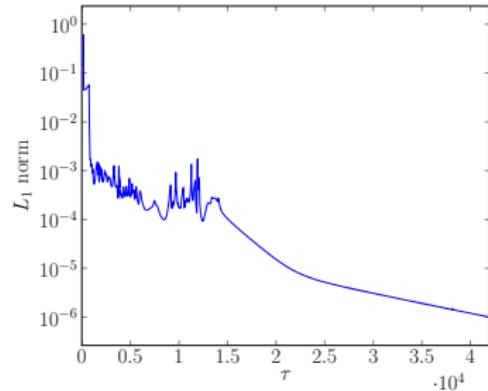
# Initial pattern: monomode $q_o \vec{1}_x$



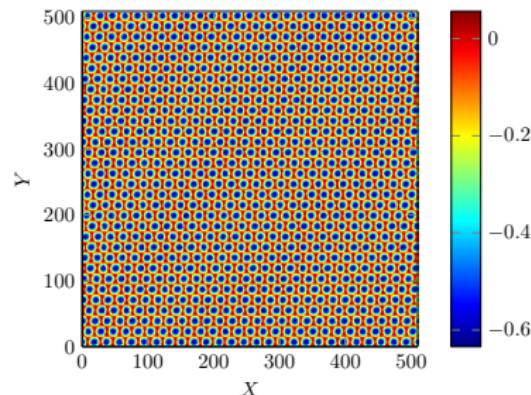
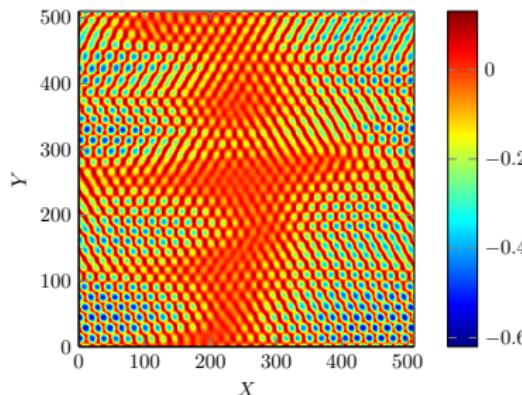
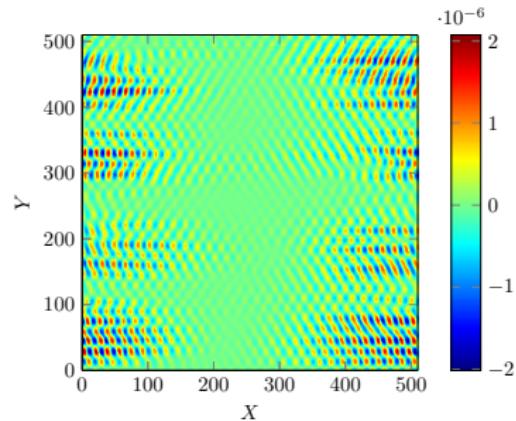
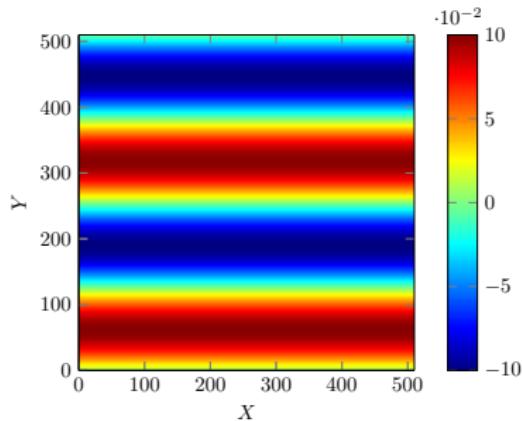
# Initial pattern: monomode $q_o \vec{1}_x$



# Initial pattern: monomode $q_o \vec{1}_y$



# Initial pattern: monomode $q_o \vec{1}_y$



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