Ultrafast Cellular Automata Dynamics of Phase-change Optical Response

Liwei Zhang[1,2], Kevin F. MacDonald[1], and Nikolay I. Zheludev[1,3]

1. Optoelectronics Research Centre & Centre for Photonic Metamaterials, University of Southampton, UK
2. School of Mathematics and Physics, Anqing Normal University, China
3. Centre for Disruptive Photonic Technologies, SPMS & TPI, Nanyang Technological University, Singapore

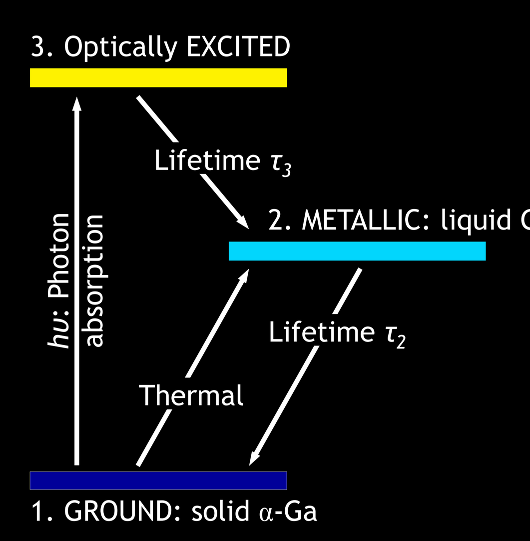
We introduce a cellular automata methodology for studying photonics of light-induced phase transitions. Multiphysical complexity over disparate length/timescales is reduced to a simple, heuristic rule/parameter set in a model successfully describing several independent experimental datasets.

Phase-change Photonics

- Light-induced structural transitions are of enormous technological importance and fundamental scientific interest …
  - optical data storage
  - laser-based manufacturing
  - controlling laser dynamics
  - optical and plasmonic modulation
  - insight to fundamental physics of transition mechanisms
- … BUT comprehensive modelling is extremely challenging, involving
  - atomic/molecular structural change
  - domain/crystallization dynamics
  - inhomogeneous change of optical properties
  - transport/dissipation of heat & light
  - time & length scales spanning many orders of magnitude
- A cellular automata (CA) model can capture this complexity in a sparse set of 'evolutionary' rules

CA Model for Gallium Phase-change Nonlinearity

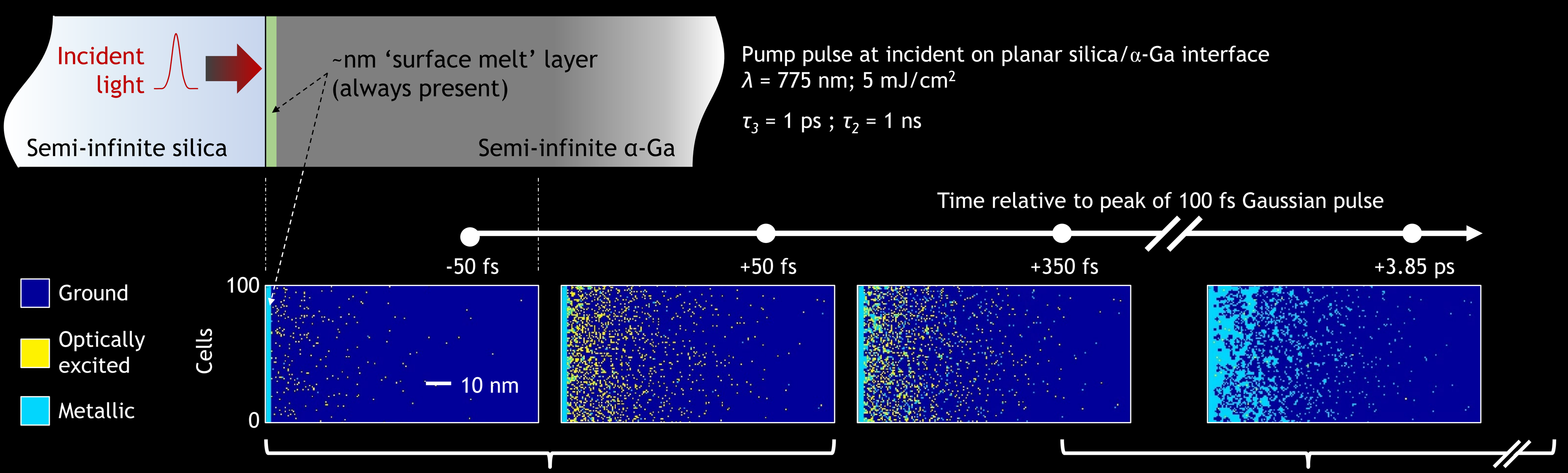
- Solid Ga near its bulk melting point (T_m = 29.8°C) manifests a gigantic, broadband phase-change nonlinearity underpinned by thermal + non-thermal light-induced surface metallization.



- CA model for metallization dynamics: 2D array of Ga (crystalline unit) cells; 3 states; 4 transition rules applicable in each time step.

GROUND → EXCITED	IF a photon is absorbed (non-thermal)
GROUND → METALLIC	Probabilistic, conditional on environment *[more likely if more neighbouring cells are EXCITED/METALLIC]*
EXCITED → METALLIC	Probabilistic *[Dependent on EXCITED state lifetime]*
METALLIC → GROUND	Probabilistic, conditional on environment *[Dependent on METALLIC state lifetime IF allowed by neighbourhood - blocked if most surrounding cells are EXCITED/METALLIC]*

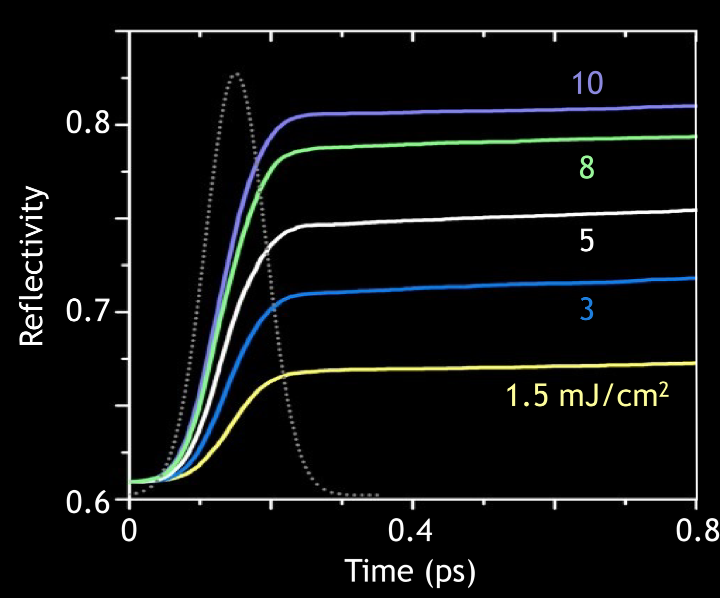
Femtosecond Optical Excitation



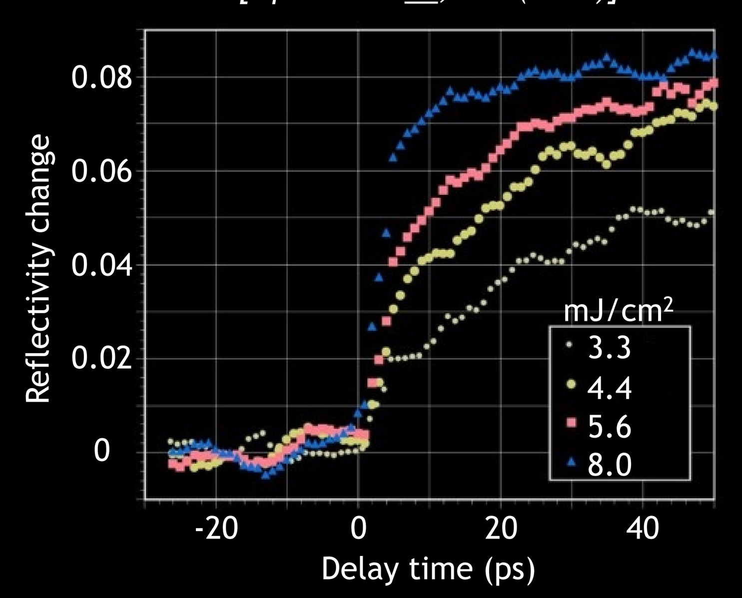
During pulse:
Rapid (non-thermal) excitation of diffuse EXCITED cell population; Neighbourhood irrelevant.
Corresponding sharp reflectivity increase.

After pulse:
EXCITED cells relax to METALLIC;
Thermalisation via neighbour interactions.
Slow reflectivity increase over 10s of ps.

CA model

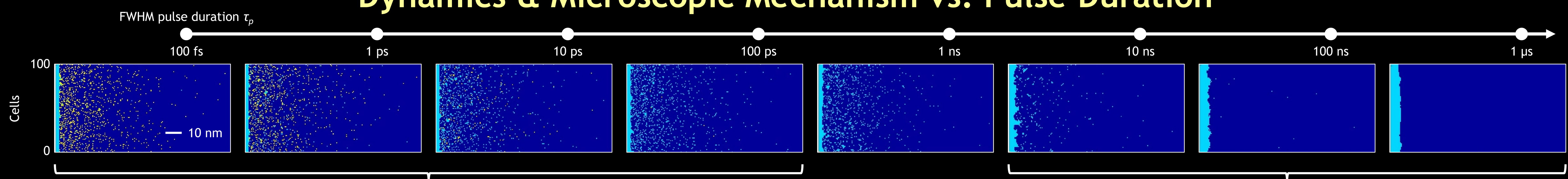


Experiment
[Opt. Lett. 26, 441 (2001)]



- CA replicates experimental pump-probe measurements of fs-ps Ga/silica interface reflectivity dynamics, supporting prior inference of diffuse 'fractional melting' mechanism.

Dynamics & Microscopic Mechanism vs. Pulse Duration



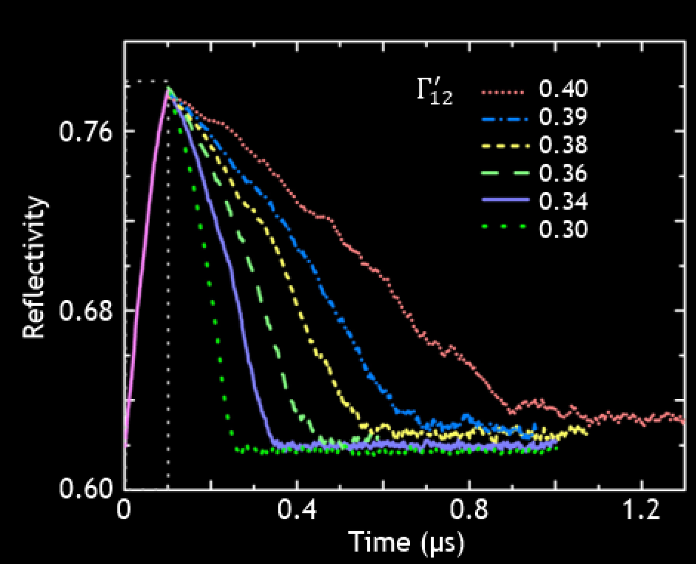
Short pulses ($\tau_p < \tau_2$):
Diffuse EXCITED/METALLIC cell population in GROUND state Ga bulk; No change in surface-melt layer thickness at interface. Two-step reflectivity increase - initially sharp then slower rise.

Long pulse regime ($\tau_p >> \tau_2$):
Growth of METALLIC surface-melt layer into Ga bulk.
Proportionate, steady increase in reflectivity during pulse.

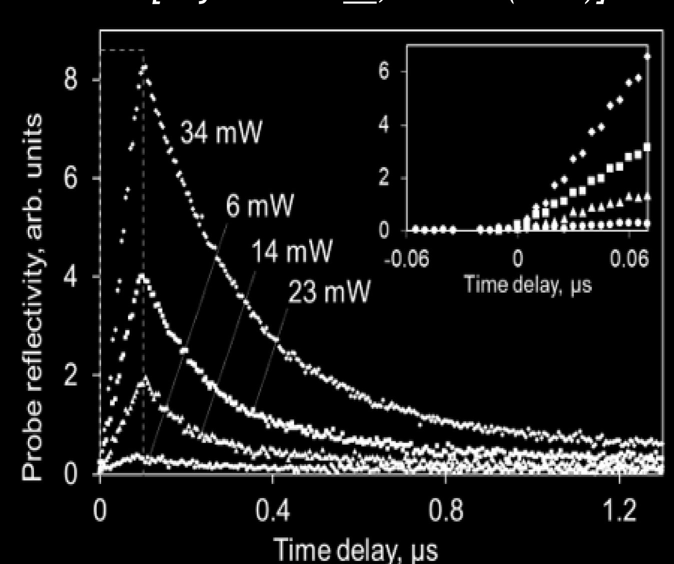
Recrystallization

- Temperature not an independent CA model parameter but GROUND → METALLIC (thermal) transition rate Γ_{12} represents proximity to T_m
- CA reproduces experimental fact that recrystallization after end of excitation pulse is faster at lower T

CA model



Experiment
[Phys. Rev. B 63, 165207 (2001)]



Summary

- Cellular automata successfully describe, non-stationary, spatially inhomogeneous dynamics and resulting nonlinear optical properties of a medium undergoing a light-induced structural transition.
- Minimal CA transition rule and physical parameter set reproduces experimentally observed behaviours over seven orders of excitation pulse duration (fs-μs), providing insight to microscopic mechanisms.
- CA methodology easily adaptable to different physical systems and nano- to macroscopic sample structures.

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