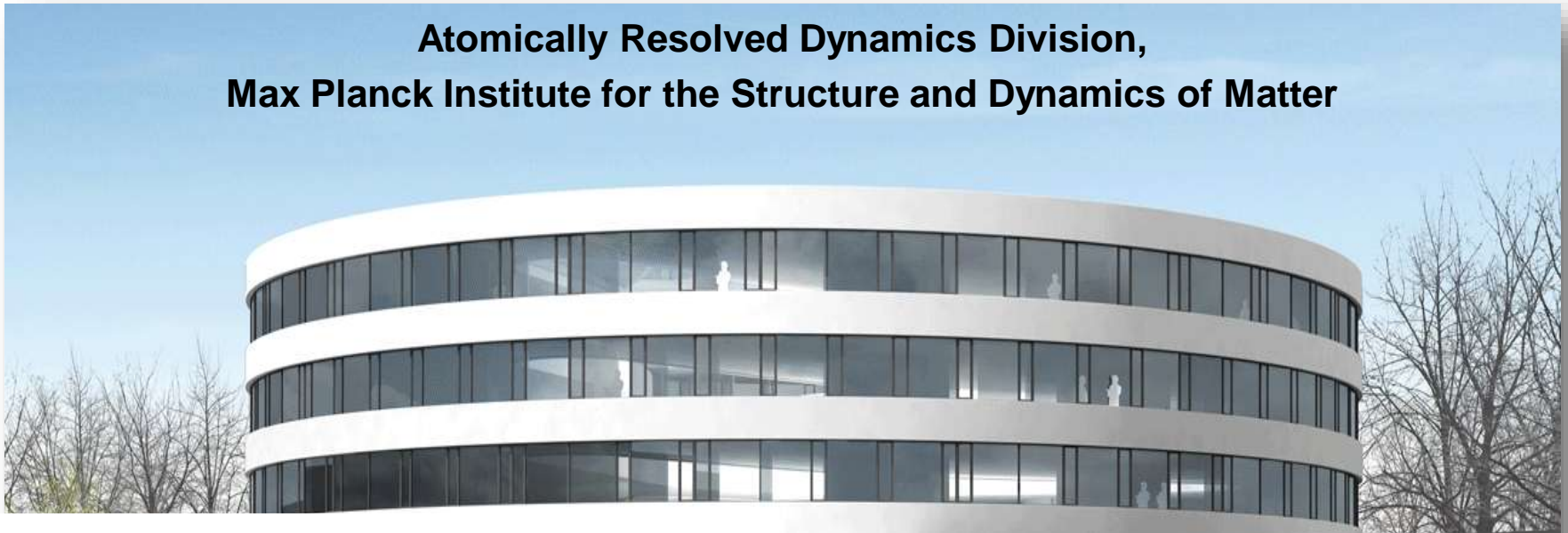


# Cold ablation driven by localized forces: a femtosecond electron diffraction study

Masaki Hada\*, R. J. Dwayne Miller

Atomically Resolved Dynamics Division,  
Max Planck Institute for the Structure and Dynamics of Matter



\*Present address: Tokyo Institute of Technology, JST-PRESTO

# Acknowledgements



## **Hamburg (MPSD)**

**R. J. Dwayne Miller**  
**German Sciaini**  
**Kostyantyn Pichugin**  
**Stuart Hayes**  
**Alexander Marx**  
**Stephanie Manz**  
**Regis Gengler**  
**Shin-ichiro Ideta**  
**Gaston Cothey**  
**Günther Kassier**

**Julian Hirscht**  
**Dongfang Zhang**  
**Sercan Keskin**  
**Albert Casandruc**



## **Univesity of Toronto**

**Gustavo Moriena**  
**Hubert Jean-Ruel**  
**Ray Gao**  
**Lai Chung Liu**

## **Tokyo institute of Technology**

**Tadahiko Ishikawa**  
**Shin-ya Koshihara**

## **RIKEN**

**Mitsushiro Nomura**  
**Reizo Kato**

## **Kyoto University**

**Jiro Matsuo**  
**Toshio Seki**

## **Edinburg University**

**Michał Kochman**  
**Derek Wann**  
**Carole Morrison**

# Outline

## ➤ **Electron diffraction setup**

- **Motivation**
- **DC gun, RF gun, Relativistic gun**
- **Pulse characterization**

## ➤ **Science with electron diffraction**

- **Single-shot experiment**
  - KI Cold ablation driven by localised forces
- **Multiple-shot experiment**
  - $\text{Pt(dmit)}_2$  Coherent phonon in large lattice material

# Femtosecond electron diffraction

Optical pump

Electron probe method

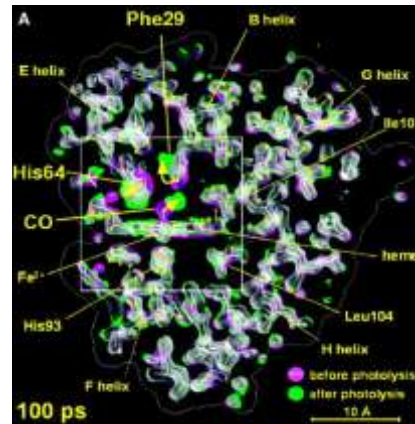
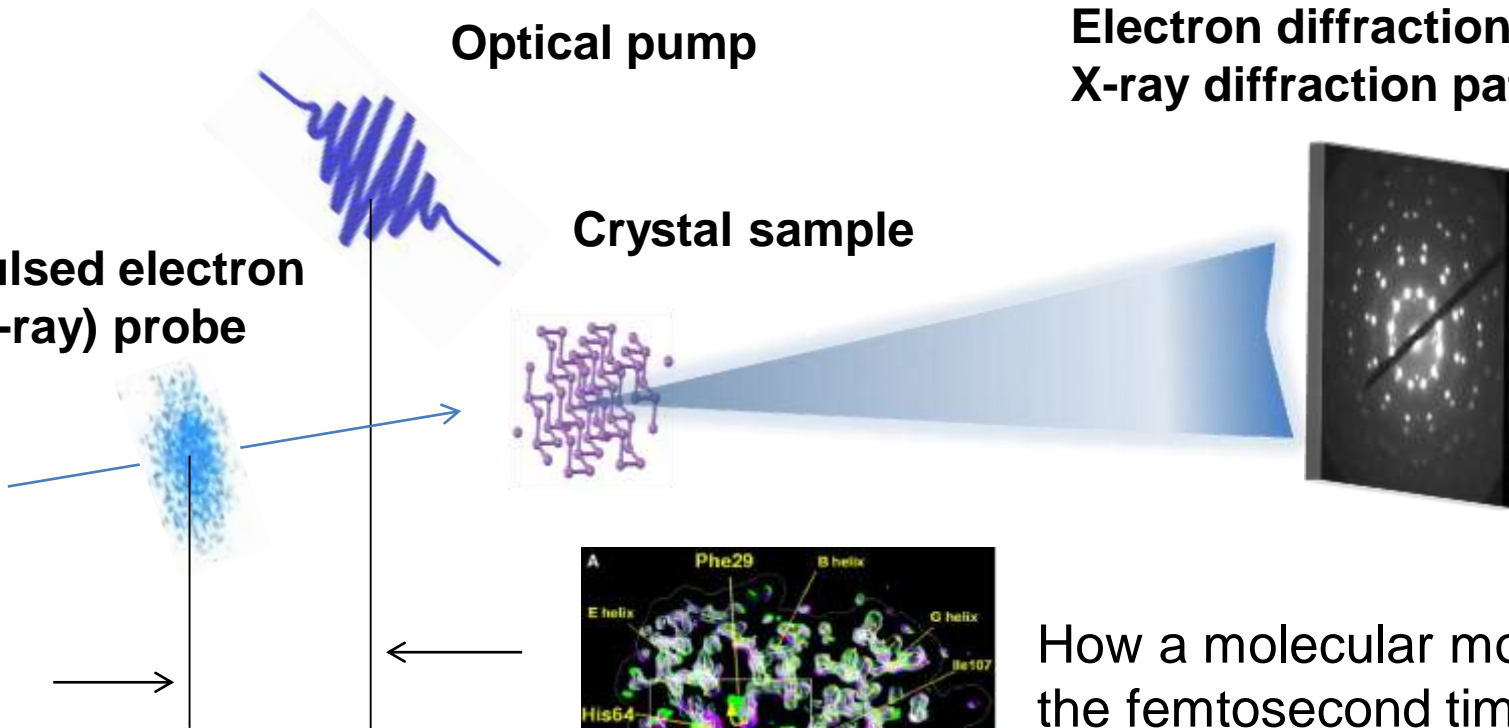
Optical pump

Crystal sample

Electron diffraction pattern  
X-ray diffraction pattern (line)

Pulsed electron  
(X-ray) probe

Optical delay  $\Delta\tau$   
30  $\mu\text{m}$  = 100 fs

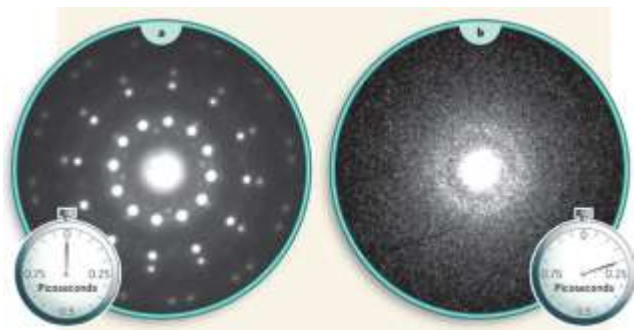


How a molecular moves at  
the femtosecond time scale?

F. Schotte, et. al., *Science* 300, (2003)  
1944–1947.

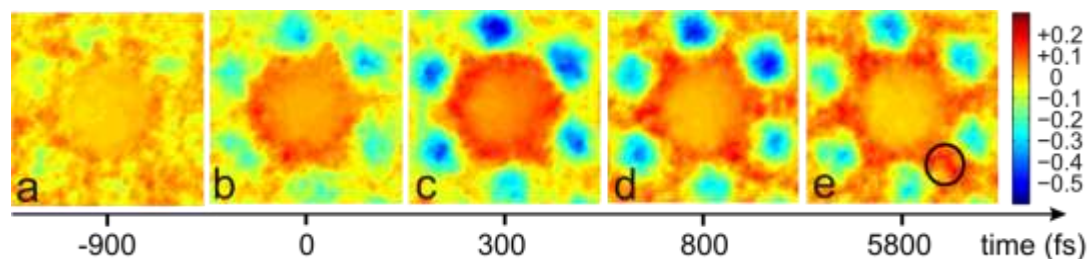
# Electron diffraction studies from Toronto

## Al, Au, Bi, Si melting



G. Sciaini et al, Nature **458**, 56 (2009).

## TaS<sub>2</sub> CDW



M. Eichberger et al, Nature **468**, 799 (2010).

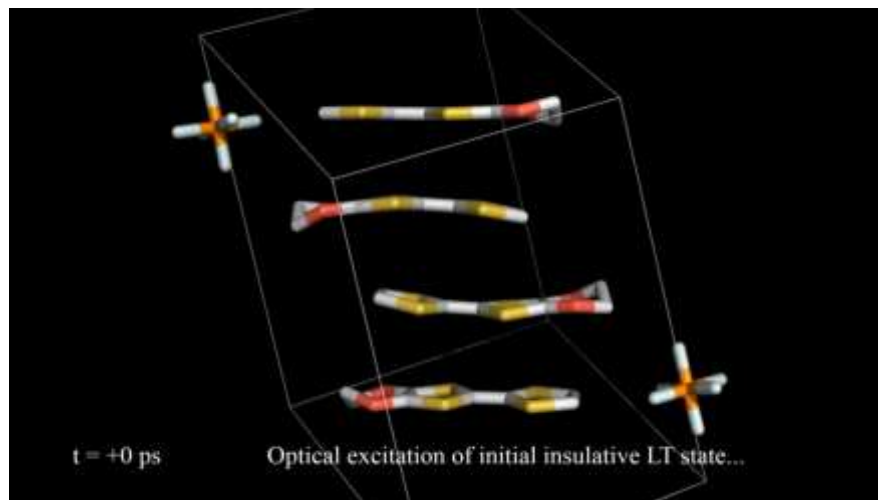
## (EDO-TTF)<sub>2</sub> PF<sub>6</sub> Diarylethene

## Phase transition

-2000 fs



M. Gao et al, Nature **496**, 343 (2013).



# Electron vs X-ray

|  | Electrons (100 keV)         | X-ray (10 keV)   |
|--|-----------------------------|------------------|
| Wavelength [Å]                                       | 0.04                        | 1.2              |
| Mechanism of radiation damage                        | Secondary electron emission | Photoelectric    |
| Ratio (inelastic/elastic) scattering events          | 3                           | 10               |
| Energy deposited relative electron per elastic event | 1                           | >1000            |
| Elastic mean free path                               | 1                           | $10^4 - 10^5$    |
| Coherence length                                     | 1 – 10 nm                   | >1 $\mu\text{m}$ |

# XFEL and Electron gun



SACLA/ Spring8



LCLS/ Stanford

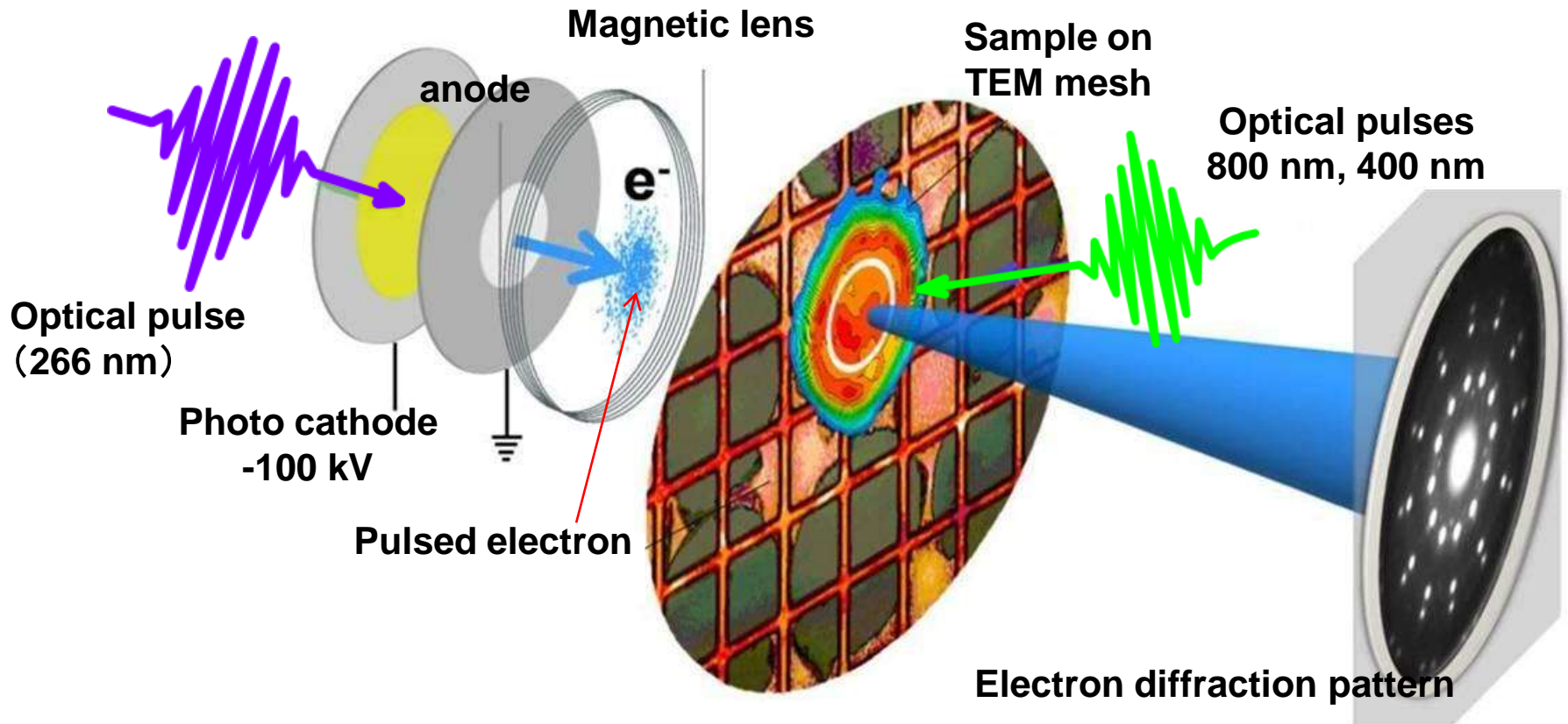


European XFEL/ DESY



~100 keV DC electron gun/ Hamburg

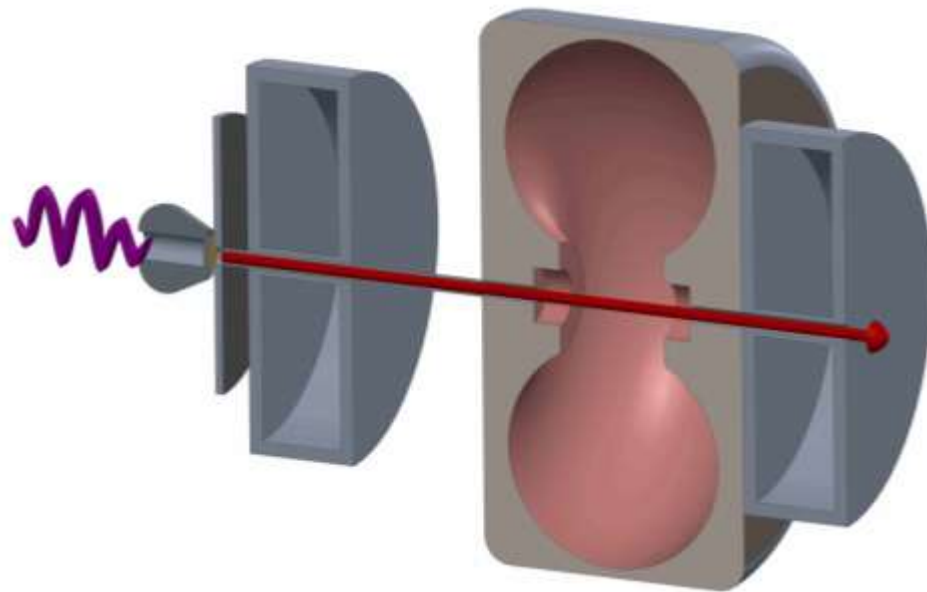
# DC Femtosecond electron diffraction



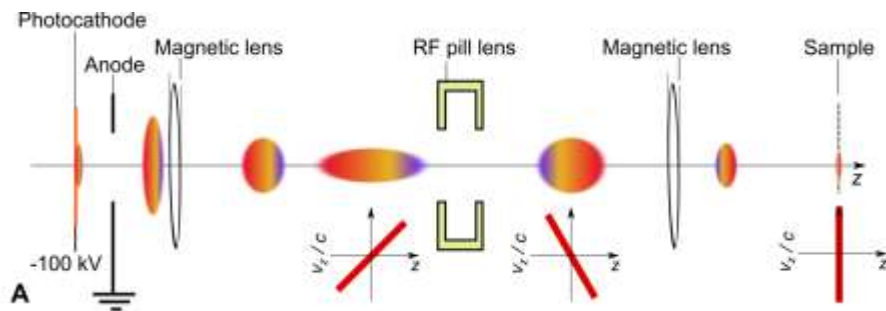
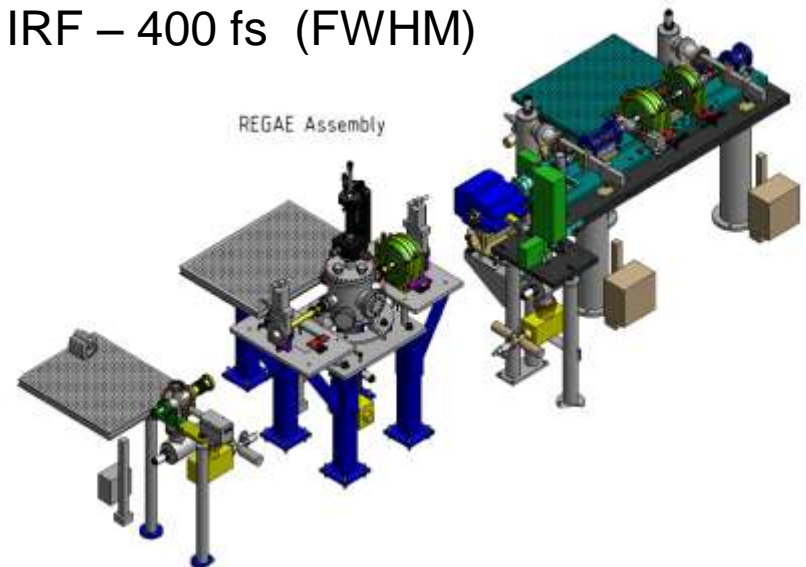
Accel voltage : 90 – 120 kV  
Pulse duration : 300 – 500 fs  
Electron density :  $10^4$  –  $10^5$  electrons/pulse



# Femtosecond Electron diffraction + RF



Buncher Machine  
100 keV  
10 Hz  
200000 e/pulse  
IRF – 400 fs (FWHM)



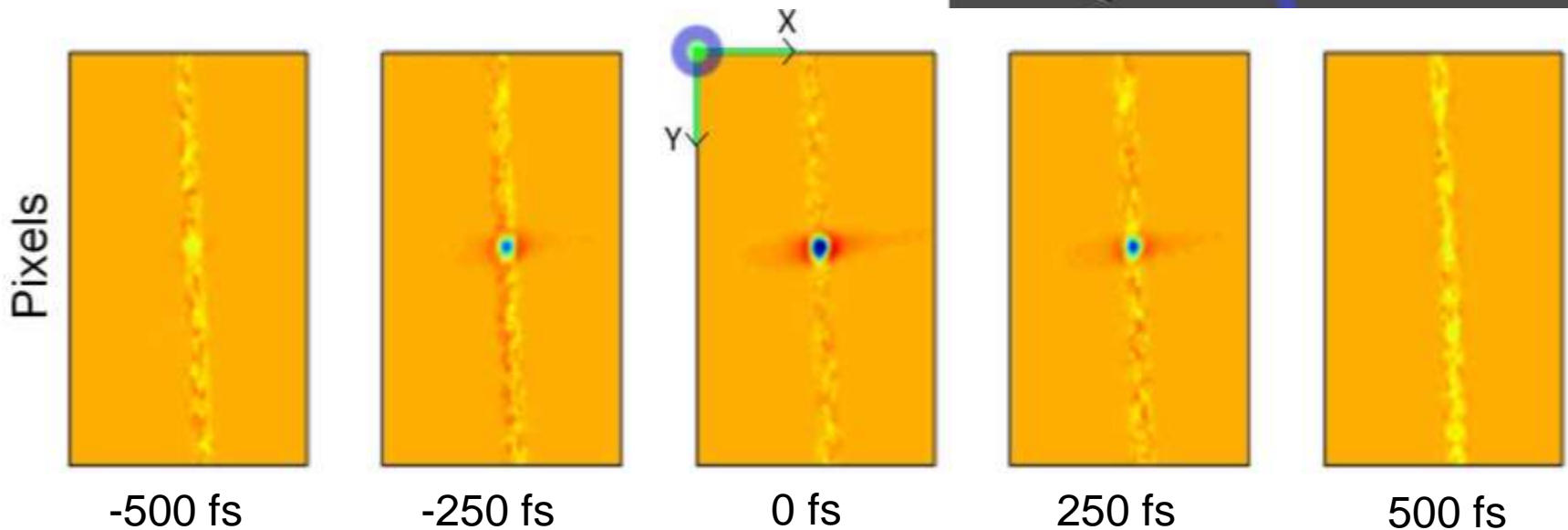
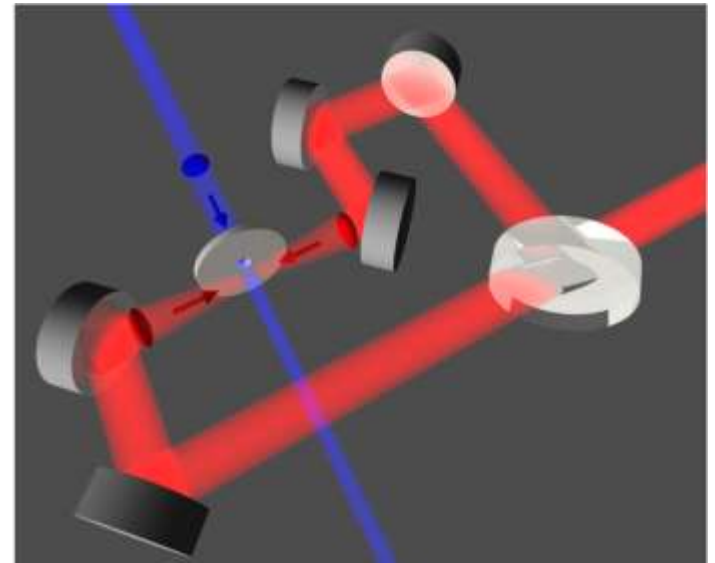
M. Gao, H. Jean-Ruel et al., Opt. Express **20**, 12048 (2012).

REGAE  
3–5 MeV  
12.5 Hz  
 $1 \times 10^7$  e/pulse  
IRF – <50 fs (FWHM)

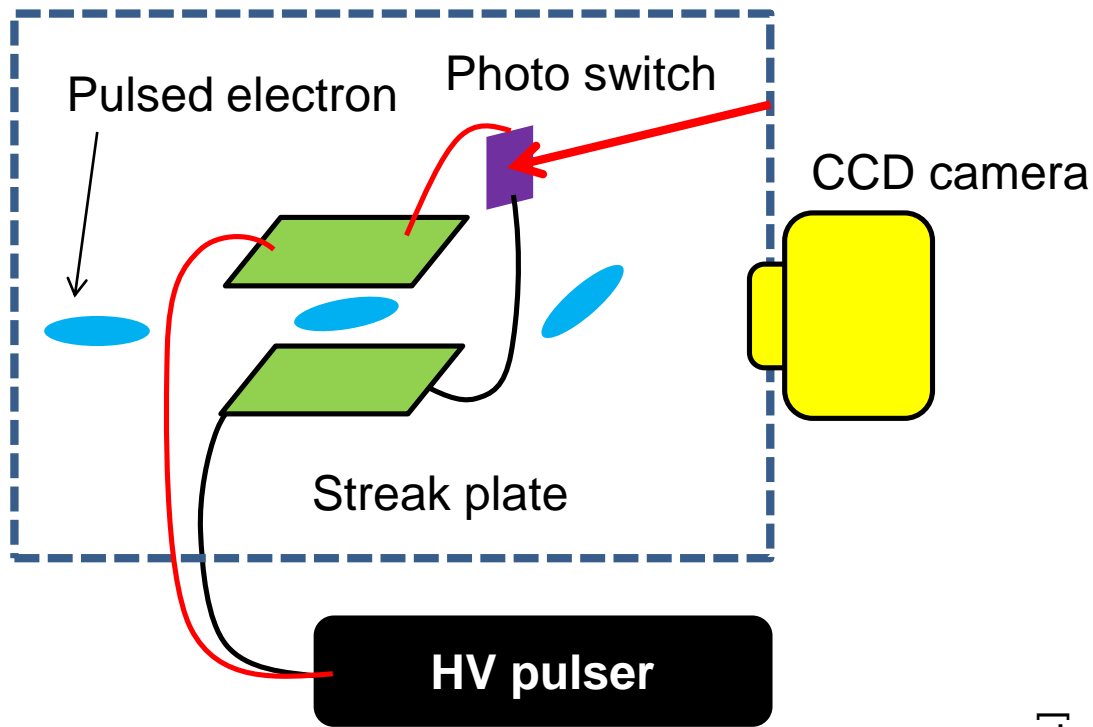
# Pulse characterization Ponderomotive

Full characterization of RF compressed femtosecond electron pulses using ponderomotive scattering

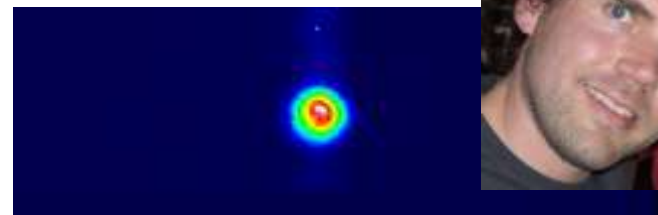
Meng Gao,<sup>1</sup> Hubert Jean-Ruel,<sup>1</sup> Ryan R. Cooney,<sup>1</sup> Jonathan Stampe,<sup>3</sup>  
Mark de Jong,<sup>3</sup> Maher Harb,<sup>1,4</sup> German Sciaini,<sup>2</sup> Gustavo Moriena,<sup>1</sup>  
and R. J. Dwayne Miller<sup>1,2,\*</sup>



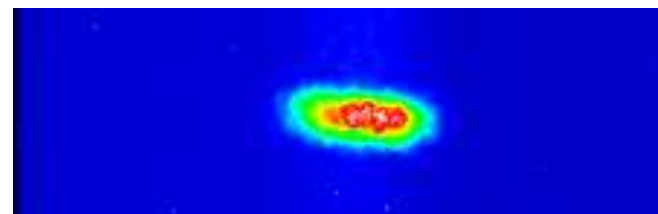
# Streak camera



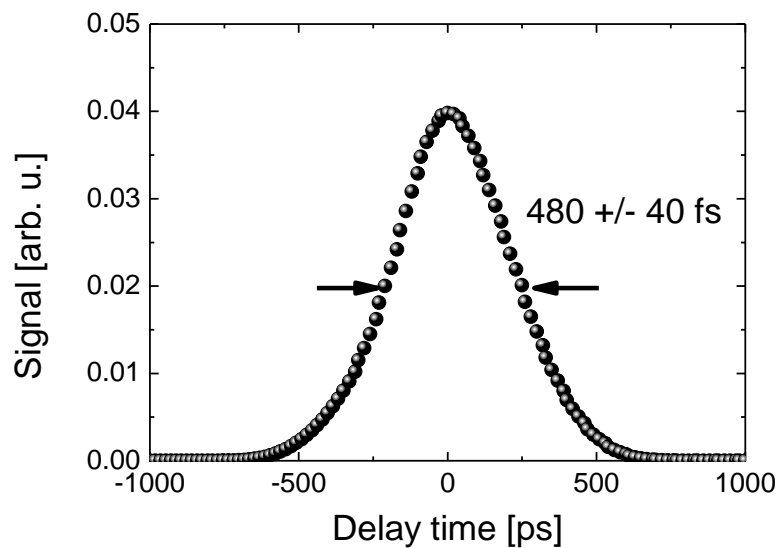
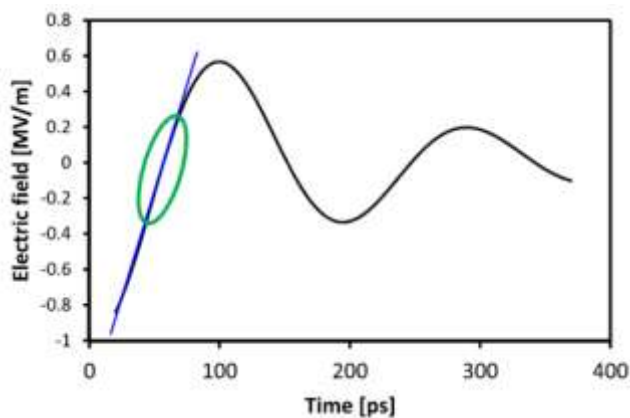
Günther Kassier



Without streak



With streak



# Outline

## ➤ Electron diffraction setup

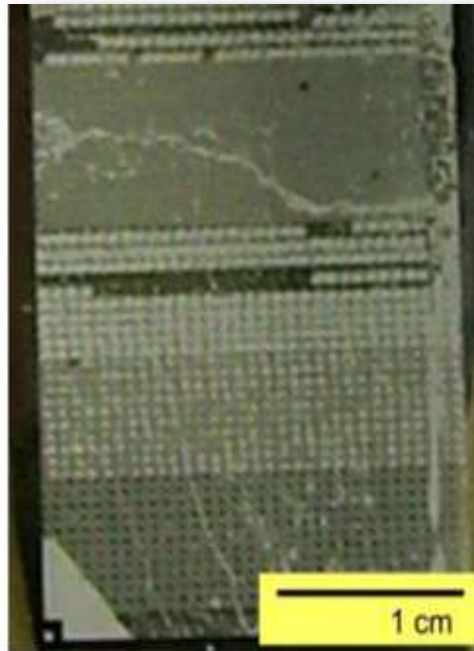
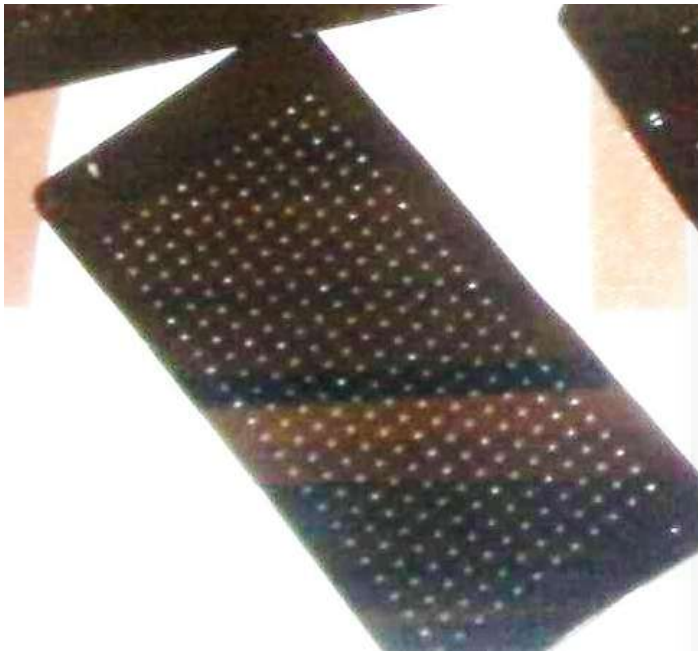
- Motivation
- DC gun, RF gun, Relativistic gun
- Pulse characterization

## ➤ Science with electron diffraction

- **Single-shot experiment**
  - KI Cold ablation driven by localized forces
- **Multiple-shot experiment**
  - Pt(dmit)<sub>2</sub> Coherent phonon in large lattice material

# Single-shot experiment

- Melting, Ablation and irreversible reactions
- Single or highly-oriented crystal sample
- Homogenous and large area sample



Julian Hirscht



Dongfang Zhang

# Si nonthermal melting

PRL **100**, 155504 (2008)

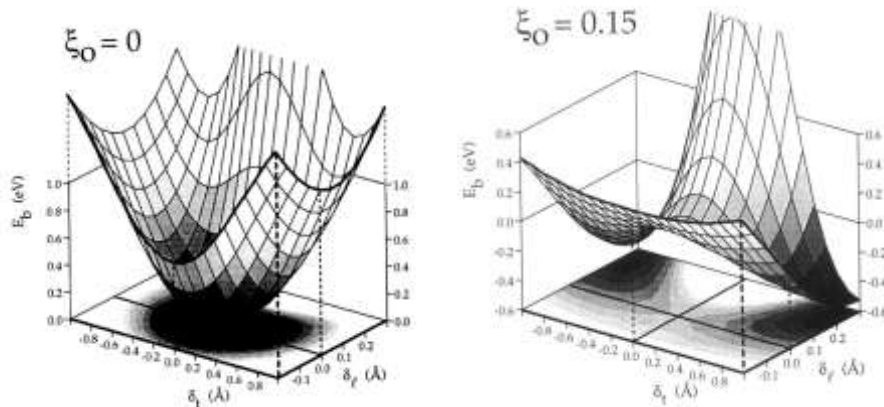
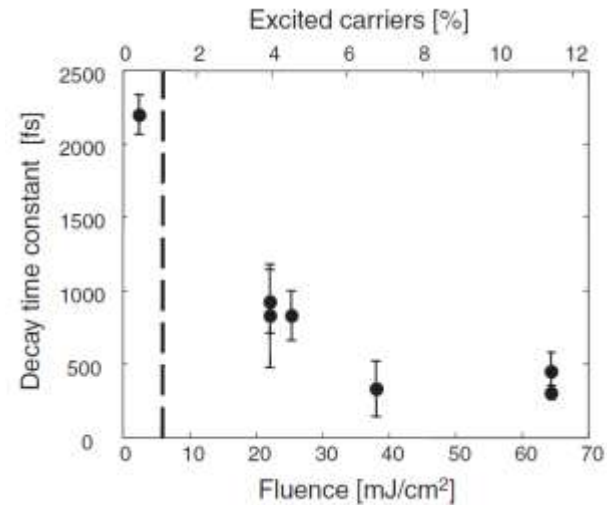
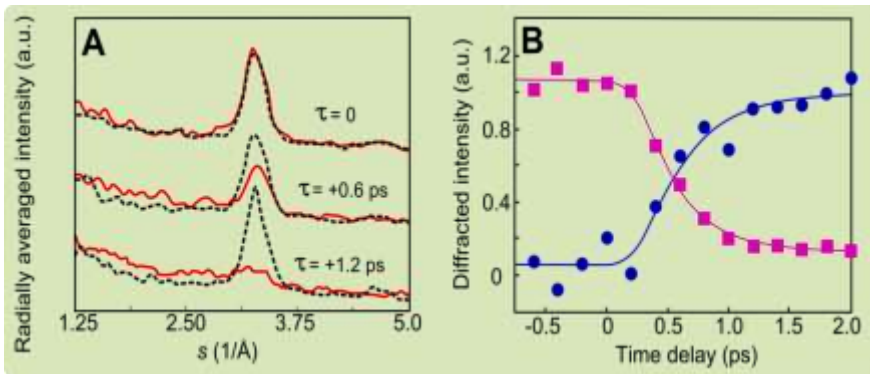
PHYSICAL REVIEW LETTERS

week ending  
18 APRIL 2008



## Electronically Driven Structure Changes of Si Captured by Femtosecond Electron Diffraction

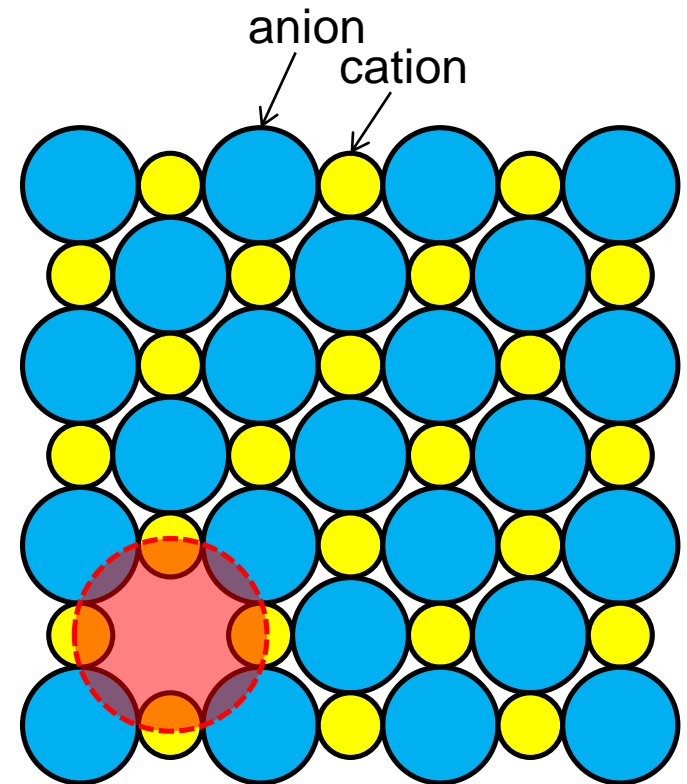
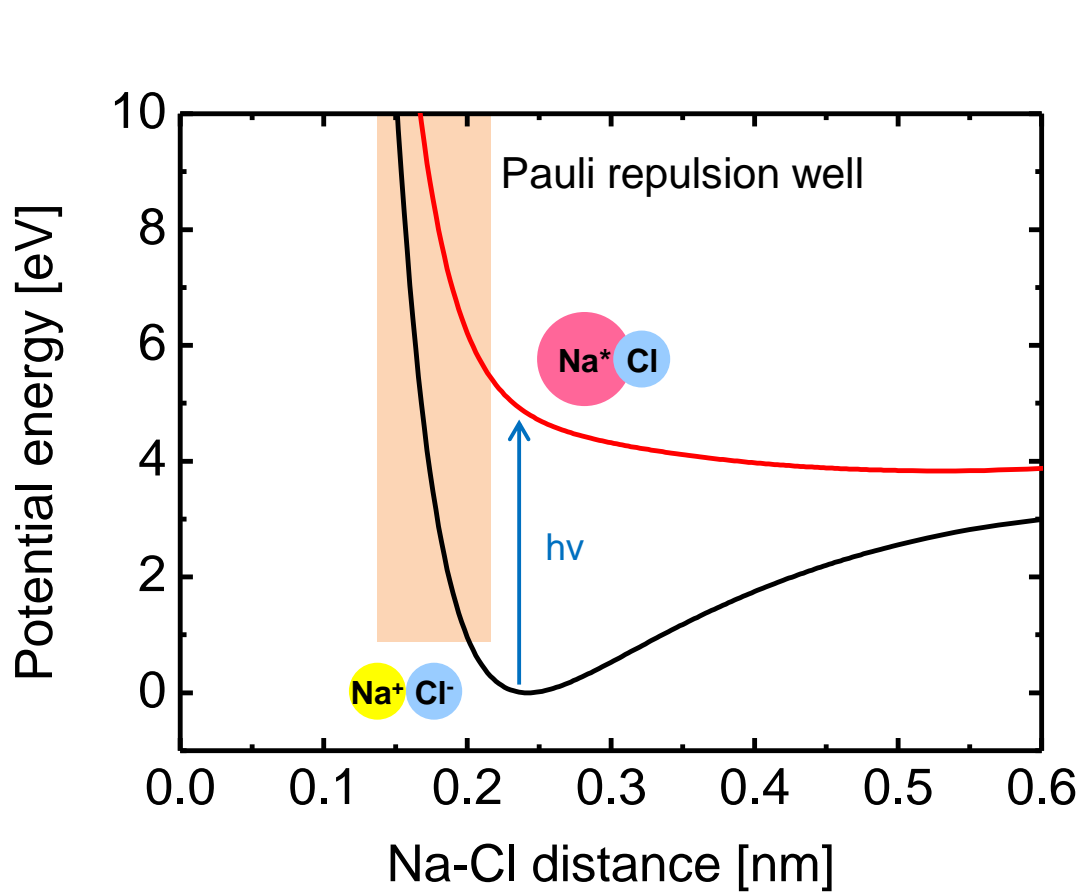
Maher Harb,<sup>1</sup> Ralph Ernstorfer,<sup>1</sup> Christoph T. Hebeisen,<sup>1</sup> Germán Sciaini,<sup>1</sup> Weina Peng,<sup>2</sup> Thibault Dartigalongue,<sup>1</sup> Mark A. Eriksson,<sup>2</sup> Max G. Lagally,<sup>2</sup> Sergei G. Kruglik,<sup>1</sup> and R. J. Dwayne Miller<sup>1,\*</sup>



**Nonthermal melting in Si !**

P. Stampfli et al. Phys. Rev. B **49**, 7299 (1994)

# Strong localized force in alkali halides



Self-trapped exciton, color center formation

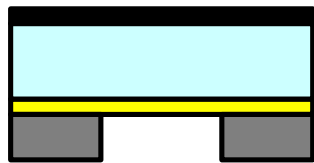
# Sample preparation – KI thin crystals

Self standing SiN (30 nm) mesh by chemical etching process

Pre-annealed (400 degrees)

Thermal deposition of KI (50 nm)

Coating with Carbon (4–5 nm)

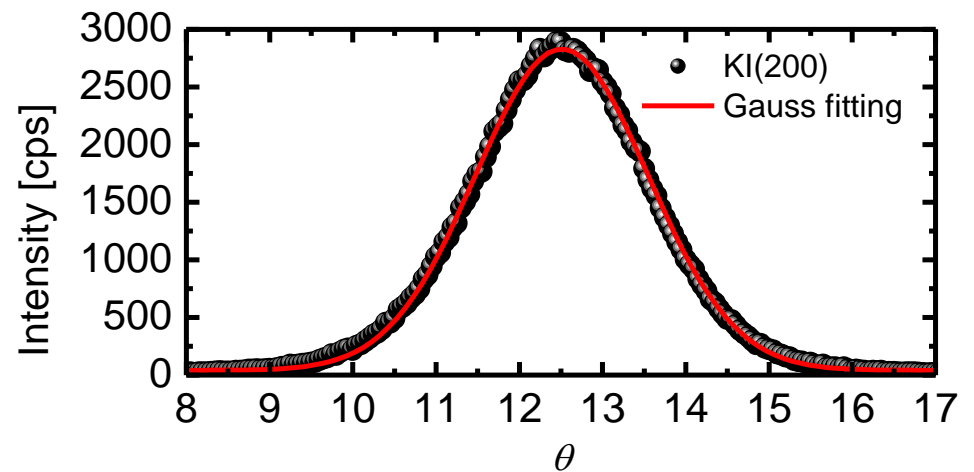
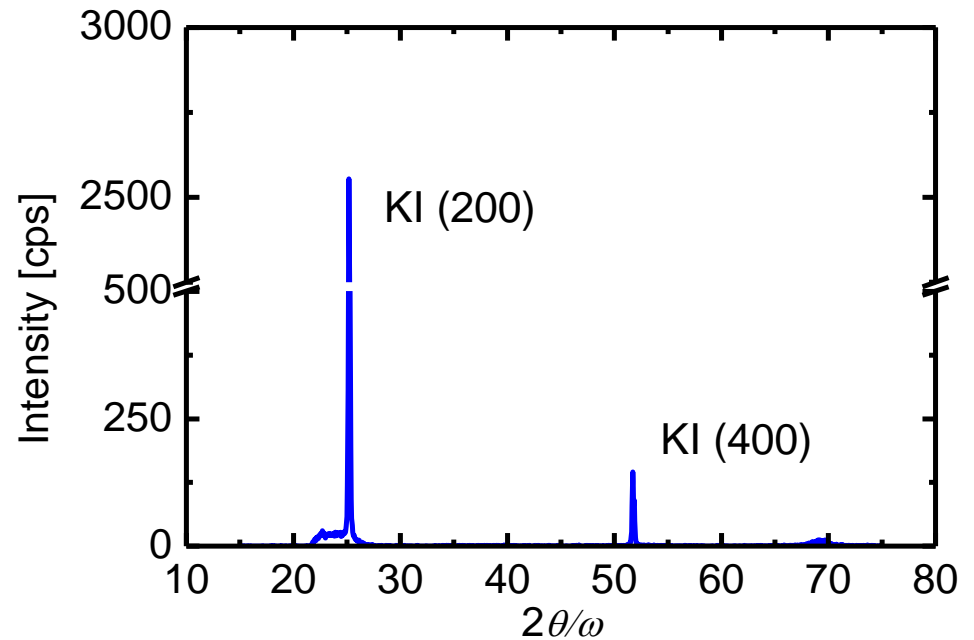


C (4–5 nm)

KI (50 nm)

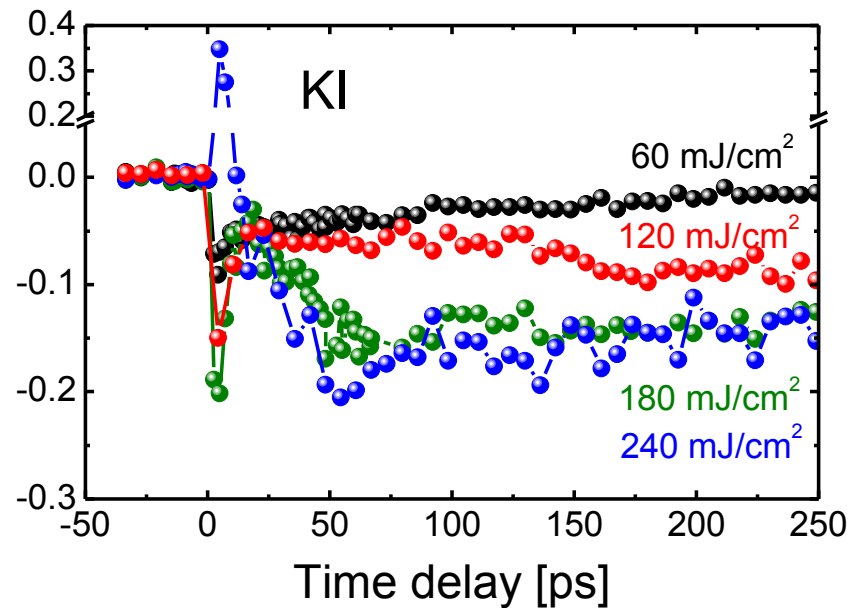
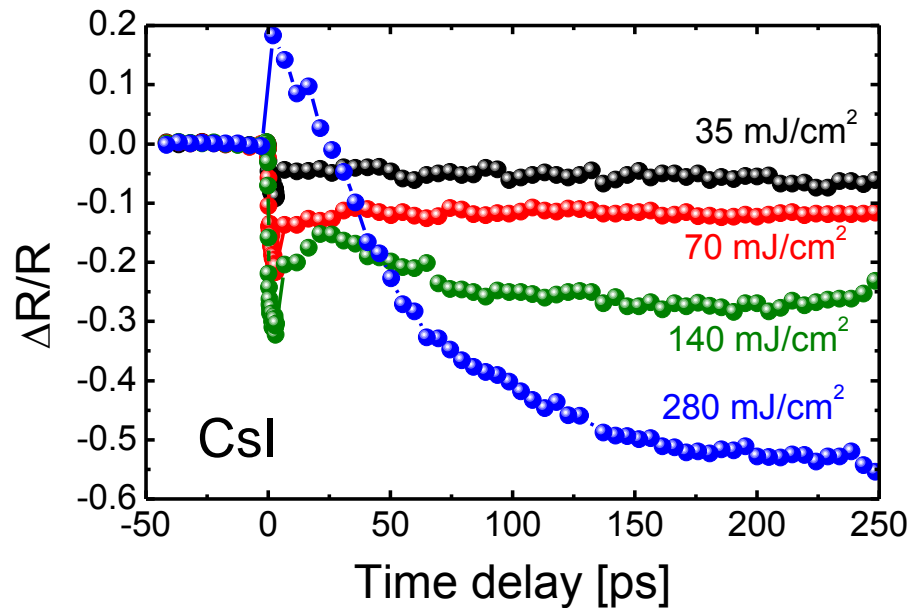
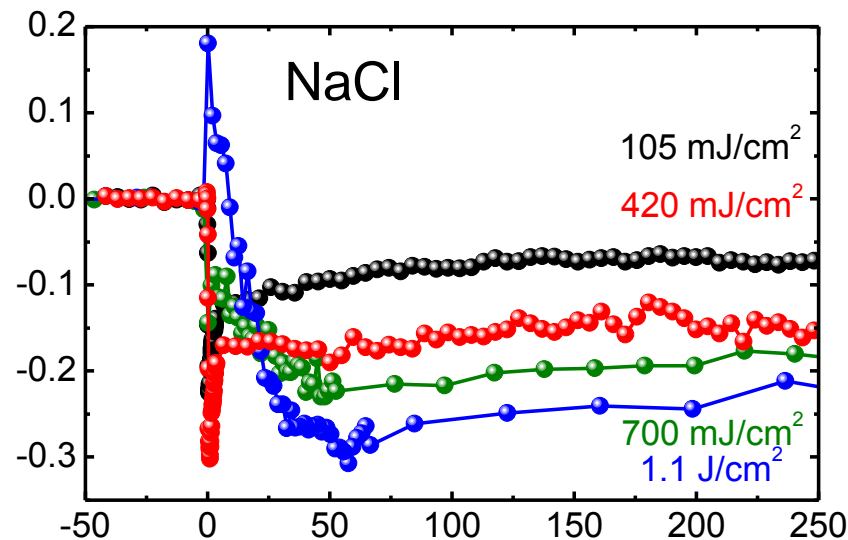
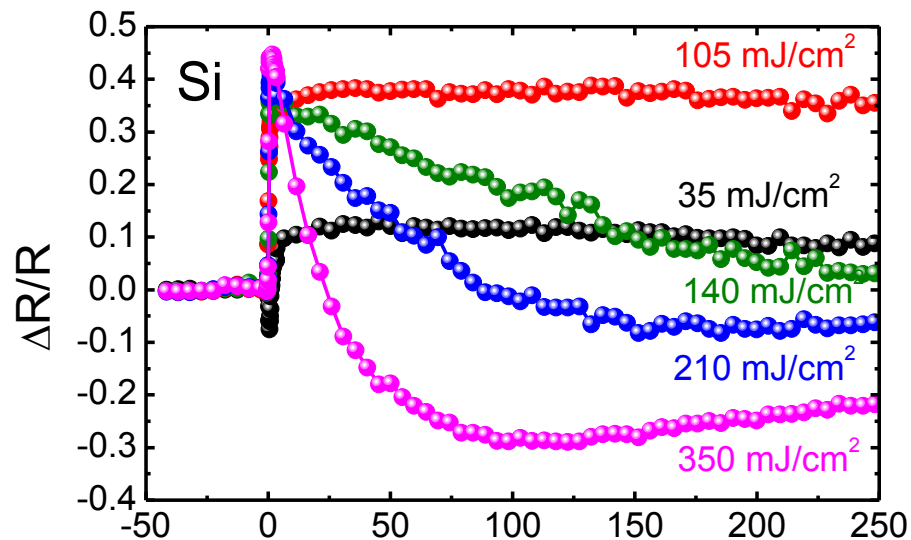
SiN (30 nm)

Si substrate

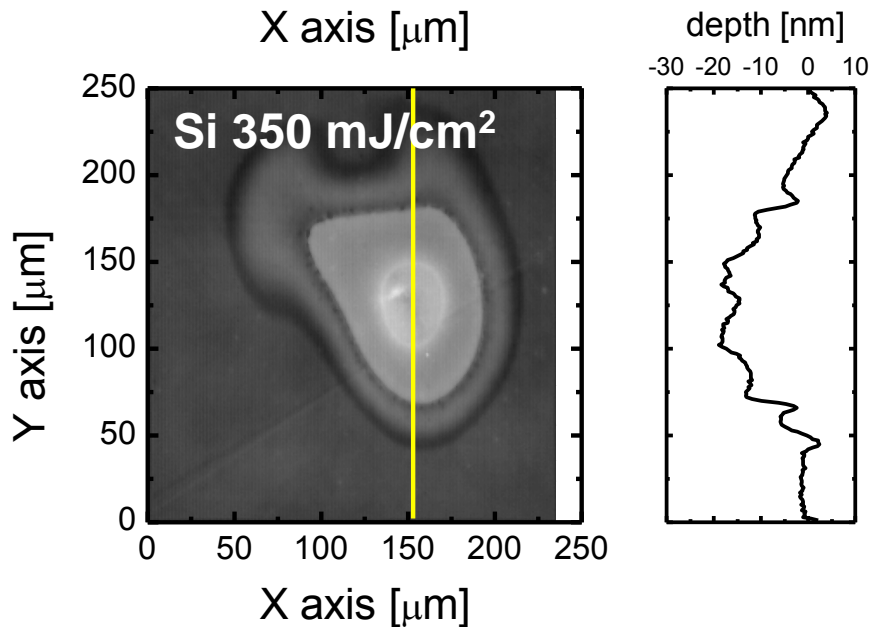
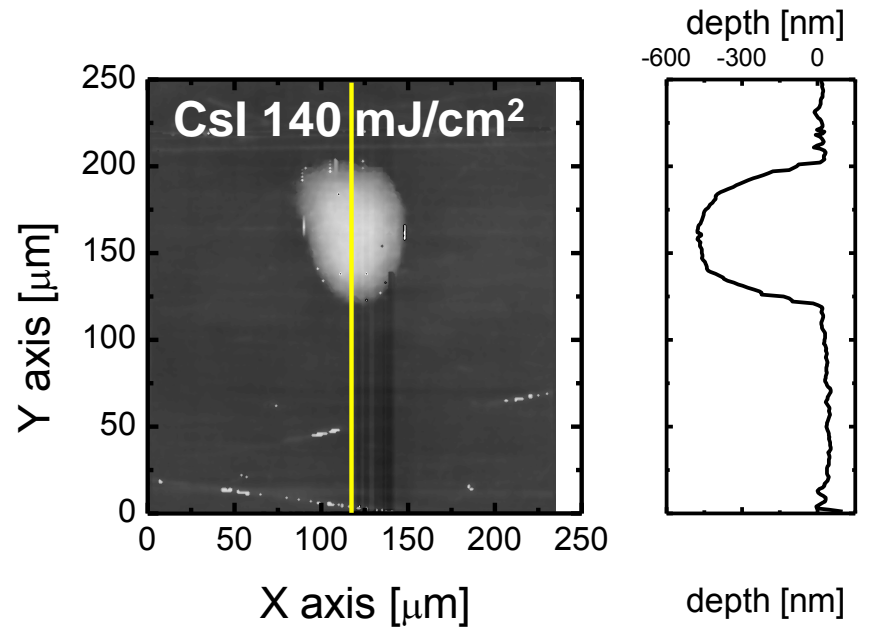




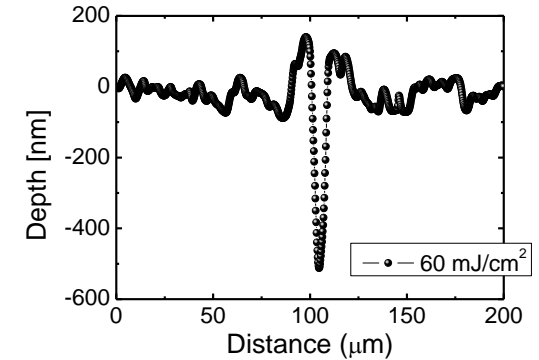
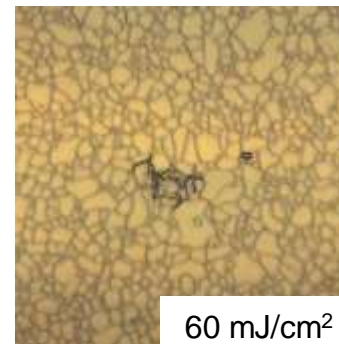
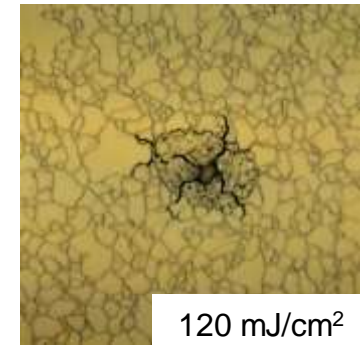
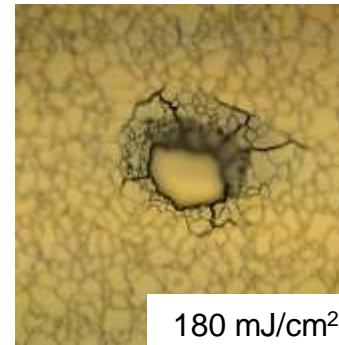
# Optical studies



# Sample surface after irradiation



KI

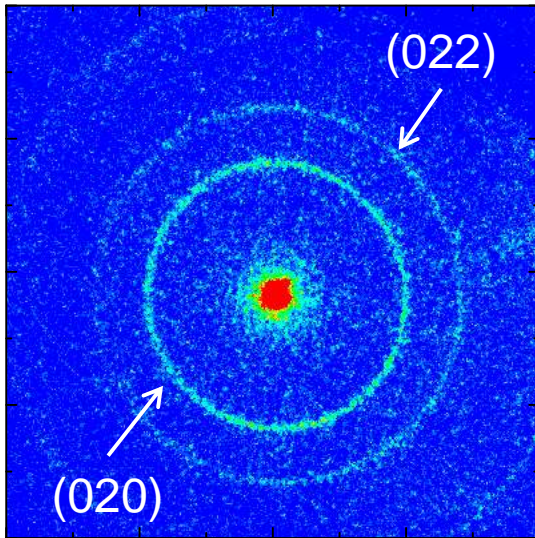


Micron deep crater formation in alkali halides with low laser fluence

# Micron deep crater in alkali halides

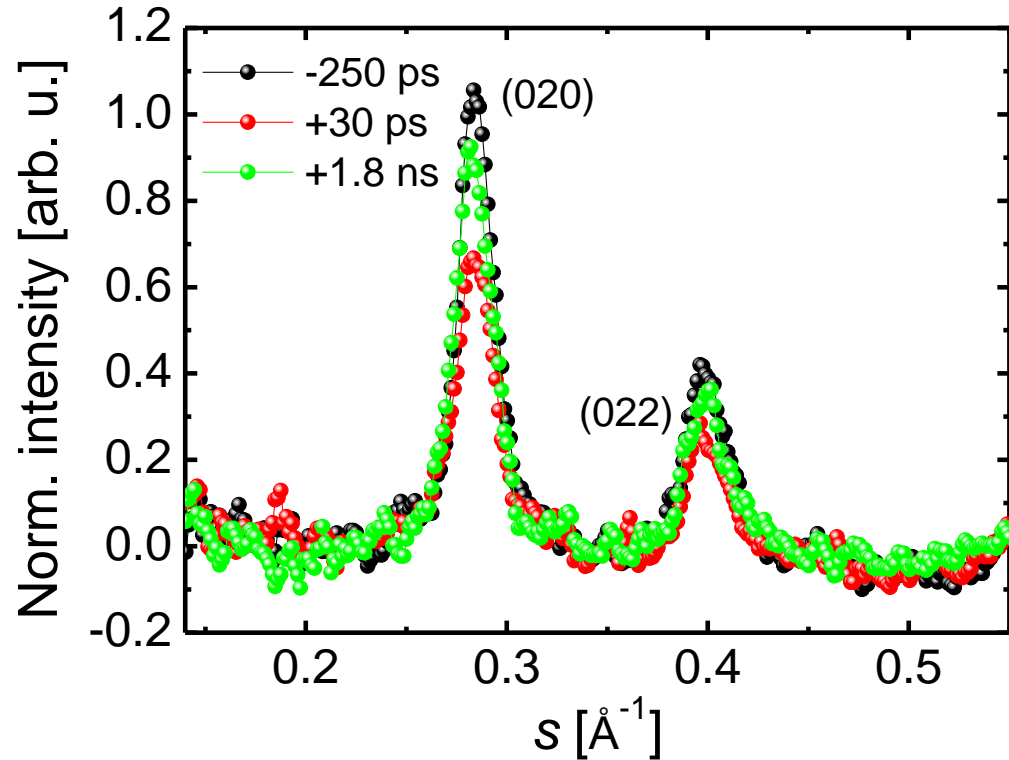
| <i>F</i>               | Depth (Si)  | <i>F</i>               | Depth (NaCl) |
|------------------------|-------------|------------------------|--------------|
| 350 mJ/cm <sup>2</sup> | 26 nm       | 420 mJ/cm <sup>2</sup> | 970 nm       |
| 210 mJ/cm <sup>2</sup> | 16 nm       | 350 mJ/cm <sup>2</sup> | 950 nm       |
| 70 mJ/cm <sup>2</sup>  | 9 nm        | <i>F</i>               | Depth (KI)   |
| <i>F</i>               | Depth (CsI) | 180 mJ/cm <sup>2</sup> | >2500 nm     |
| 280 mJ/cm <sup>2</sup> | 704 nm      | 120 mJ/cm <sup>2</sup> | 1400 nm      |
| 140 mJ/cm <sup>2</sup> | 472 nm      | 60 mJ/cm <sup>2</sup>  | 500 nm       |
| 105 mJ/cm <sup>2</sup> | 418 nm      |                        |              |

# Electron diffraction



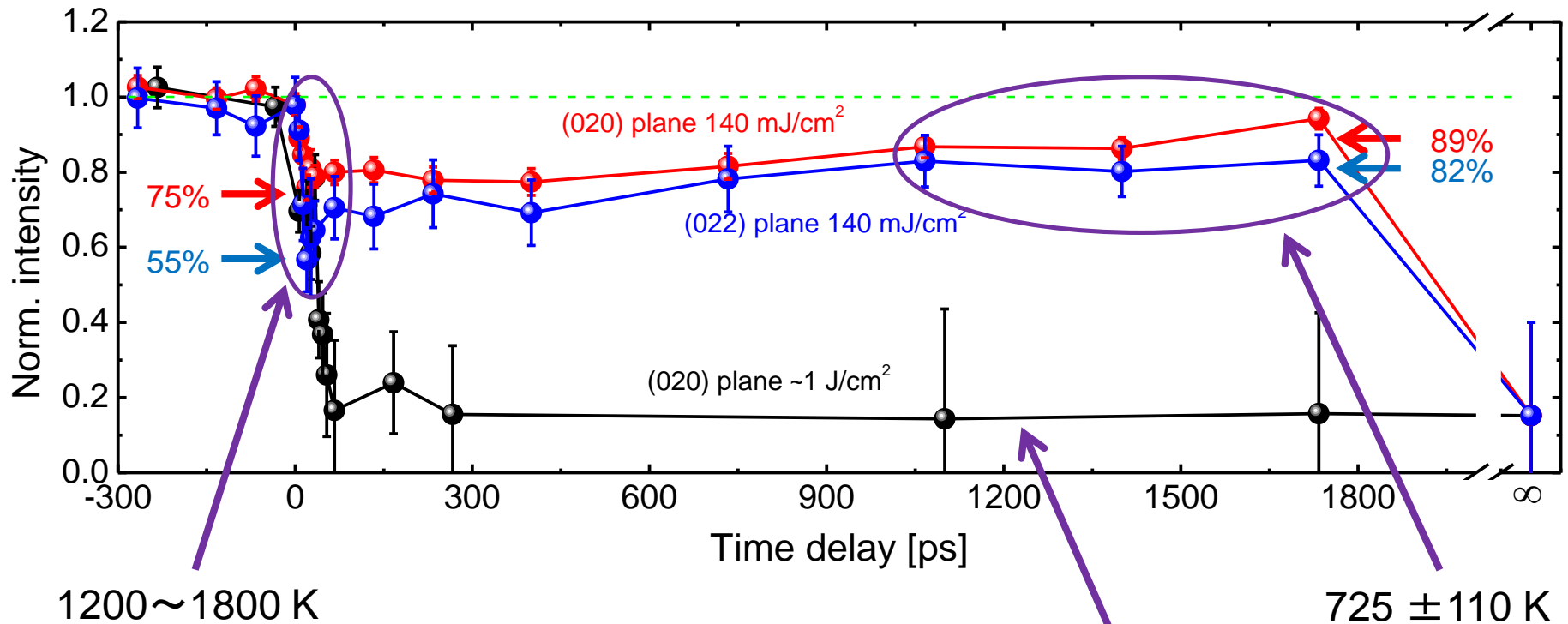
$F = 140 \text{ mJ/cm}^2$   
Absorption =  $\sim 2\%$   
Film thickness: 50 nm  
2-photon absorption

Temperature raise  
Excited halide center



$\sim 780 \text{ K}$  (melting point 954 K)  
 $\sim 4\%$

# Thermal? Plasma?



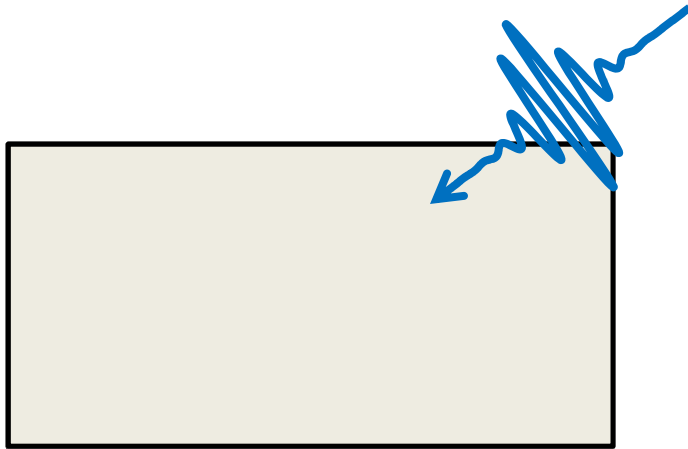
Plasma generation

$$I = |F(\vec{K})|^2$$

$$F(\vec{K}) = G(\vec{K}) \sum_{j=1}^M f_j(\vec{K}) D_j(\vec{K}) \exp(2\pi i \vec{K} \cdot \langle \vec{r}_j \rangle)$$

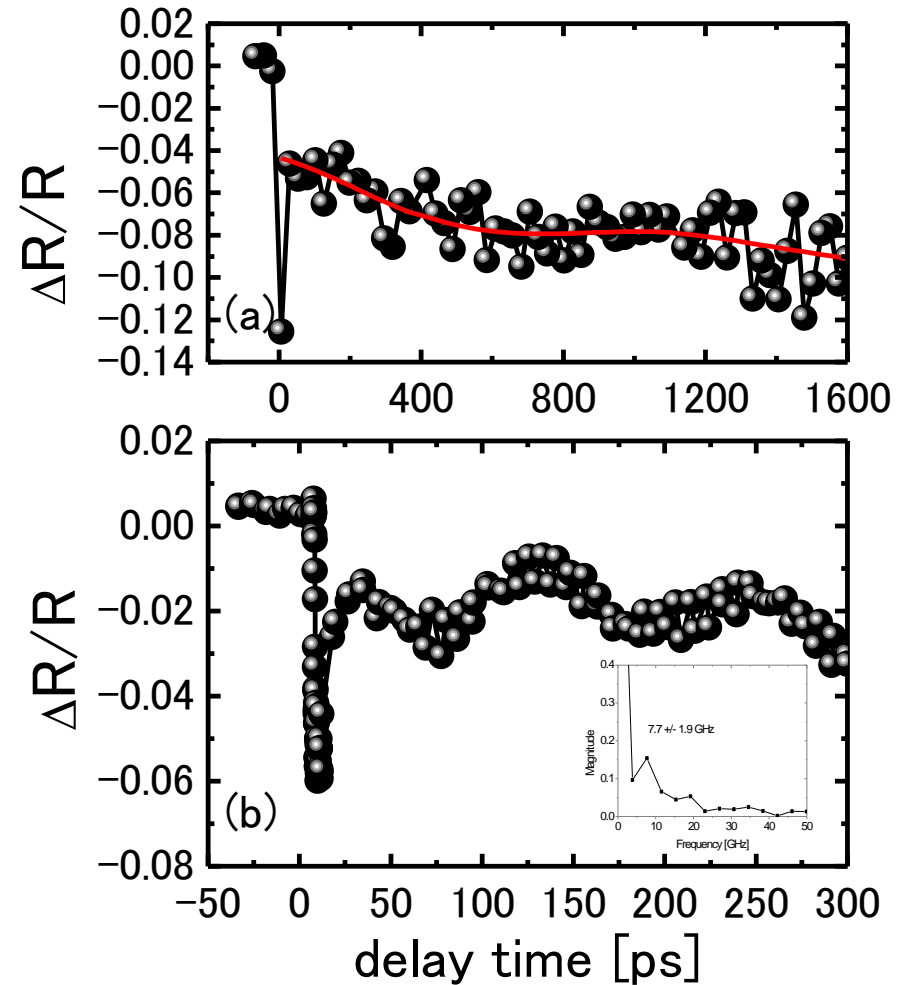
$$D_j = \exp(-K^2 B_j) = \exp(-8\pi^2 K^2 U_j) = \exp\left(\frac{8\pi^2 U_j \sin^2 \theta}{\lambda^2}\right)$$

# Localized force in alkali halides



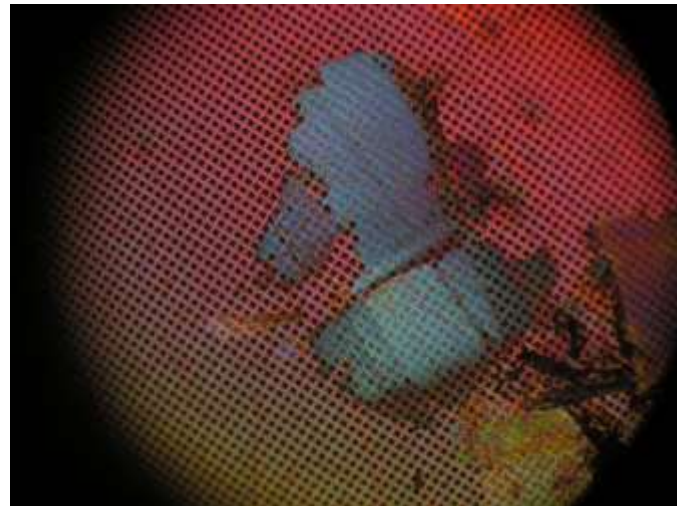
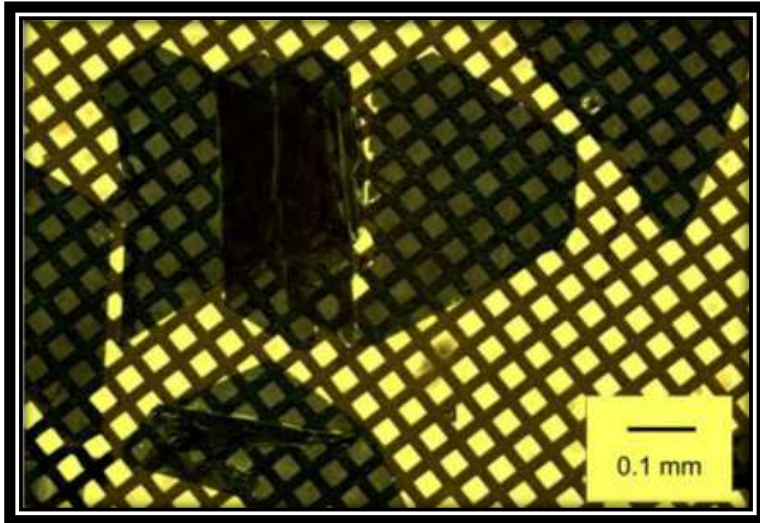
KI crystal  
photoexcitation

These defects and disordered regions introduce strong electronic stress that leads to the expulsion of material.

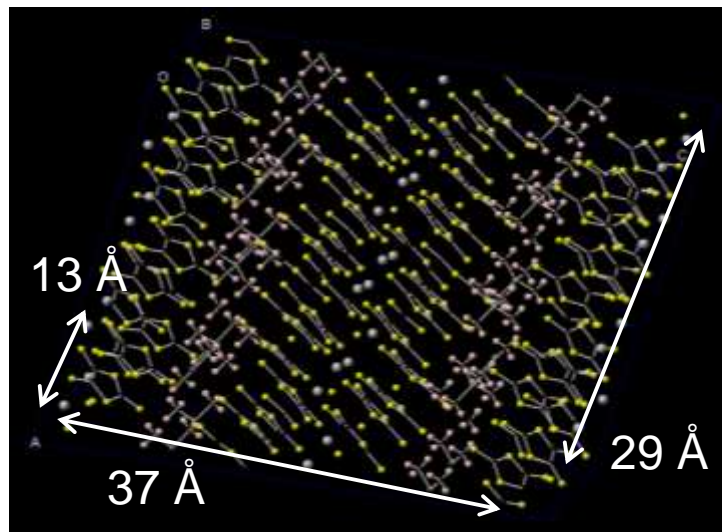


# Multiple shot experiments

- Photoinduced repetitive reactions
- Single crystal
- Large structural modification
- Find a proper material



# $\text{Me}_4\text{P}(\text{Pt}(\text{dmit})_2)_2$



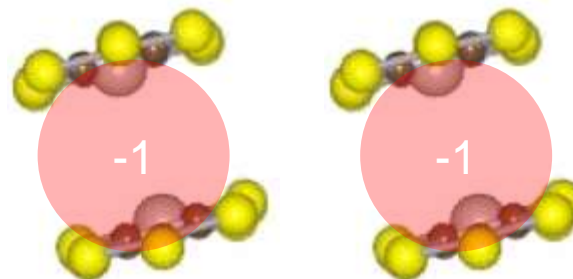
Stuart Hayes



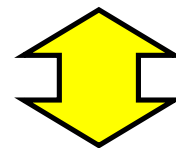
Tadahiko Ishikawa

Collaboration work  
Miller Group, Max Planck Institute  
Koshihara Group, Tokyo Institute of Technology  
Kato Group, RIKEN

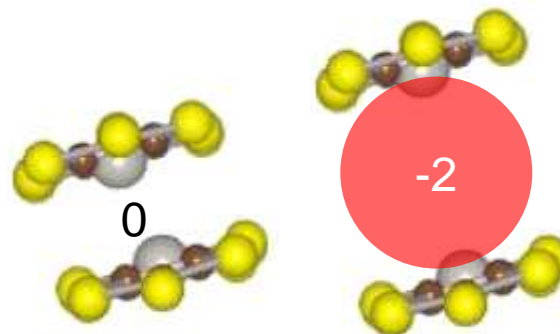
HT phase (>215 K)



Paramagnetic metal



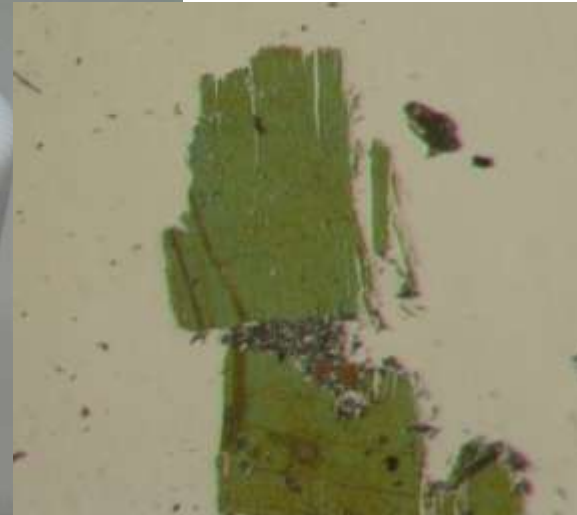
LT phase (>215 K)



Charge-ordered state  
Non magnetic insulator



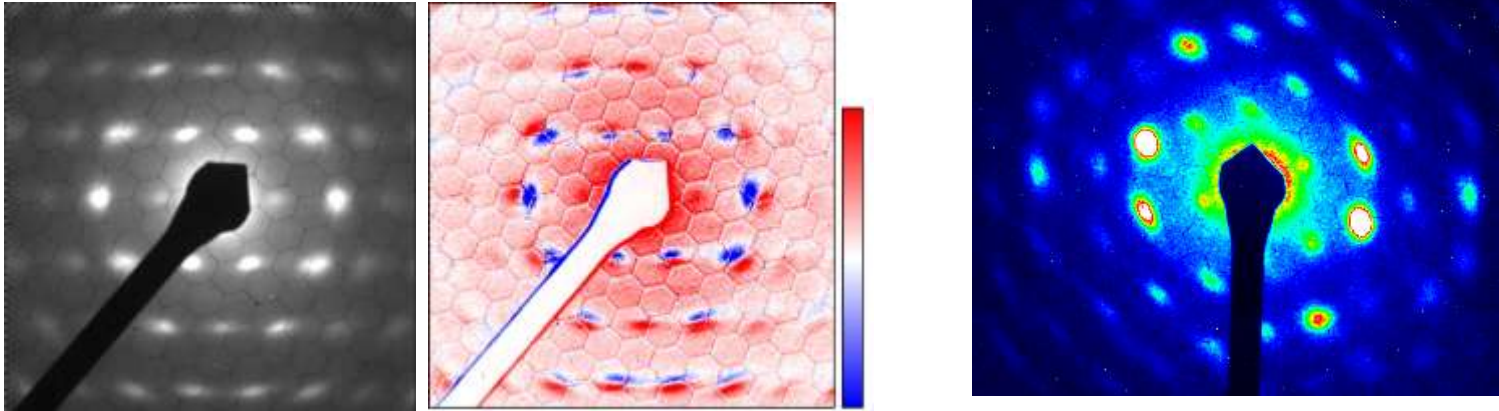
# Sample preparation



Microtome the sample down to 150 nm

Sercan Keskin

# Coherent phonon oscillation



Crystal with large lattice parameters  
→ We can go for protein crystals!?

We observed the coherent phonon oscillation...  
We have not put the index yet...

# Thank you for your attention!

