MeV Ultrafast Electron Diffraction and its Applications for Strongly Correlated Material Studies


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WORKSHOP ON ULTRAFAST ELECTRON SOURCES FOR DIFFRACTION AND MICROSCOPY APPLICATIONS
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Outline

• Advantages of MeV Ultrafast electron diffraction (UED)
  High time resolution, large scattering cross section, cost effective……

• Performance of BNL MeV UED
  High-quality diffraction – better than $10^3$ contrast
  The highest single-shot diffraction sensitivity (5 fc)
  ~100 fs time resolution

• MeV UED for Strongly Correlated Material Studies
  The ultrafast dynamics in TaSe$_2$ and La$_{0.5}$Sr$_{1.5}$MnO$_4$

• Summary
Electron diffraction vs X-ray diffraction

- Scattering cross section \( \sim 10^4 \) larger
- Less damage/scattering event
- Table-top, cost effective

- Time resolution limit
- Penetration limit

Challenges of DC UED

The energy of electrons from DC gun: <100 keV

- limited accelerating field
- Pulse broadening
- Velocity mismatch
- Limited penetration (10 nm)
Electron pulse broadening of DC UED

- Pulse broadening in accelerating field:

\[ \tau = 2.34 \times 10^{-12} (\Delta U)^{1/2} / E_{\text{acc}} \]

for \( \Delta U = 0.5 \text{ eV}, E_{\text{acc}} = 10 \text{ MV/m}, \tau \approx 160 \text{ fs} \).

- Beam expansion by space charge effect:

It is difficult to achieve <100 fs time resolution for DC UED

UED based on RF Gun

- 50-100 MV/m, 2-5 MeV
- Intense beam ($10^6$ e-, <100 fs)
- Single-shot diffraction for irreversible process
- Less (NO) velocity-mismatch
- Thicker samples
- Better sensitivity
- Timing jitter

Schematic of BNL gun based MeV UED

$$\left(\Delta t\right)^2 = \left(\Delta t_{\text{laser}}\right)^2 + \left(\Delta t_e\right)^2 + \left(\Delta t_{VM}\right)^2 + \left(\Delta t_{\text{TOA jitter}}\right)^2$$

- temporal resolution
- laser pulse length
- e- pulse length
- velocity mismatch
- TOA jitter

Timing Jitter: A challenge to MeV-UED?

RF Gun

$$\Delta t_{jitter} = \int \left( \frac{1}{V(t)} - \frac{1}{c} \right) d\ell \approx \frac{L_{\text{drift}}}{\beta c} \frac{1}{\gamma^2} \left( \frac{\Delta E}{E} \right)_{\text{jitter}}$$

For $$L_{\text{drift}} = 0.8 \text{ m}$$, $$\gamma = 6$$, and $$\frac{\Delta E}{E} = 10^{-4}$$, $$\Delta t_{jitter} < 10 \text{ fs}$$
Beam size on detector: \( \sim 200 \text{ um} \)
Synchronization of RF and laser: \(< 50 \text{ fs}\)
High Quality Diffraction: Polycrystal

Sample: polycrystalline Al

$\sigma_e \approx 30$ fs

P.F Zhu et al, unpublished
High Quality Diffraction: Crystal & Super-lattice

100 shot MeV UED in BNL

50 keV DC gun
Sample: single crystal TaS$_2$

P.F Zhu et al, unpublished
Ultrafast Study for Strongly Correlated Material

Challenges:
Coupling electronic-lattice system
→ charge, orbital and spin order

Strong interplay between charge, spins, orbital and lattice
→ complex phase diagrams, exotic material properties

One solution:
Decouple the subsystems in the time domain and then observe the dynamics of subsystems separately.

MeV UED: better time resolution & simultaneously observe diverse degrees of freedom.
TaSe$_2$: Charge Density Wave (CDW)

- Normal
- Incommensurate CDW
- Commensurate CDW (3×3)

Temperature:
- 122K
- 113K
- 92K
- 88K

Phase competition:
- Incommensurate
- Striped incommensurate
- Commensurate CDW (3×3)

Periodic lattice distortion

Layer structure and phase competition.

K Rossnagel Condens. Matter 23 (2011)
CDW Melting in TaSe$_2$

P.F Zhu et al, unpublished
La$_{0.5}$Sr$_{1.5}$MnO$_4$

Phase Diagram

Orbital and charge order

A material of Colossal Magnetoresistance

Larochelle et al. (2005)
La$_{0.5}$Sr$_{1.5}$MnO$_4$: Charge Order

Three orders are observed simultaneously

P.F Zhu et al, unpublished
kHz MeV-UED @ SJTU

- Higher rep rate (kHz vs. 10 Hz).
- Shorter bunch length (20 fs vs. 50 fs).
- Smaller time jitter (10 fs vs. 100 fs).
- Multi-mode excitations (200 nm – 3 um, THz).
- Versatile interaction chamber (cryo, gas, liquid).

1kHz, 20mJ/pulse

Layout of MeV UED

Solid-state modulator
Summary

1. MeV UED in BNL has demonstrated its capability of:
   - High-quality diffraction
   - The best sensitive detection (5-fc diffraction)
   - The best time resolution (~100 fs)

2. Ultrafast studies are performed on TaSe$_2$ and LSMO.
   - CDW melting in TaSe$_2$
   - Suppression of orbital order in LSMO

Thank you