Paquets d’électrons femtoseconde accélérés par laser
Application à la dynamique structurale

Jérôme Faure
Laboratoire d’Optique Appliquée
Ecole Polytechnique, France

FemtoElec
Collaborators

**Experiments: kHz laser-plasma source**
D. Gustas, I. Gonzalez, N. Zaim, B. Beaurepaire
A. Vernier, G. Gallé, D. Guénot, A. Lifschitz

**CUOS of University of Michigan:**
Z. He, A. Thomas, J. Nees, K. Krushelnick

**Laser system:**
M. Bocoum, F. Böhle, A. Jullien, J.-P. Rousseau, R. Lopez-Martens

**Beam transport**
B. van der Geer, Univ. Eindhoven, Pulsar Physics

**Sample preparation**
S. Scott, M. Lagally, Univ. Wisconsin
Outline of the talk

• Motivation / Introduction

• First laser-plasma accelerator at kHz and application to ultrafast electron diffraction

• First experiments with near-\textit{single-cycle laser pulses} \rightarrow Acceleration of \sim 5 \text{ MeV} electrons at kHz
Motivations

- Study relativistic laser-plasma interaction
- Develop alternative technology for MeV femtosecond electron beams → Laser-plasma (wakefield) acceleration
- Use electrons as a pump
  - Irradiation with femtosecond electrons, high charge
- Use electrons as a probe
  - Structural dynamics at the femtosecond level
Snapshots of cooperative atomic motions in the optical suppression of charge density waves

Maximilian Eichberger¹*, Hanjo Schäfer¹*, Marina Krumova², Markus Beyer¹, Jure Demsar¹,³, Helmuth Berger⁴, Gustavo Moriena⁵,⁶, Germán Sciaini⁵,⁶* & R. J. Dwayne Miller⁵,⁶

Electron diffraction on 1T-TaS₂, Eichberger et al., Nature 2010
A photoinduced metal-like phase of monoclinic VO$_2$ revealed by ultrafast electron diffraction

Vance R. Morrison,$^1$ Robert. P. Chatelain,$^1$ Kunal L. Tiwari,$^1$ Ali Hendaoui,$^3$ Andrew Bruhács,$^2$ Mohamed Chaker,$^3$ Bradley J. Siwick$^{1,2,*}$
Electron sources for diffraction
Compact and efficient - $10^5$ gain in elastic scatt. cross section

Photocathode + DC field: $< 100$ keV & $< fC$ charge & $> 300$ fs
Limited by space charge
Velocity dispersion

Photocathode + RF field: sub-100 fs & pC charge
RF compression at 100 keV - Van Oudheusden et al., PRL 105, 264801 (2010)
Synchronization and jitter: resolution $> 100$ fs

But many phonons with period $< 100$ fs, (graphene: 10 fs)
Parameters of ideal source

- time resolution < 10 fs
- energy 50 keV – 5 MeV
- energy spread < few %
- charge fC – pC

- emittance < 0.1 mm.mrad
- rep. rate > 100 Hz
- stability < few %

Using a laser-plasma accelerator

Relativistic intensity
$10^{18}$ W/cm$^2$

Short laser pulse
$c \tau \approx \lambda_p / 2$
Accelerating electrons in wakefields

**PROS**

- Extreme fields
  - $1 \text{ MeV} / 10 \mu m$
  - Compact accelerator
  - Mitigates space charge

- fs bunches
  - $< \lambda_p/4$

- no jitter
Laser-plasma accelerators: state of the art

**Laser:**
1 J, 30 fs, 1 Hz

**Electron beam:**
collimated (few mrad)
100 MeV – 1 GeV (1-10 % spread)
1.5 fs rms duration (O. Lundh et al., Nat. Phys. 2011)

**Our challenges**
- Operate at kHz (with only <10 mJ)
- Accelerate electrons to few MeV
- High beam quality
- Stable source
- Sub-10 fs duration

Faure et al. Nature 2004
Mangles et al. Nature 2004
Geddes et al., Nature 2004
Faure et al, Nature 2006
Leemans et al., Nat. Phys. 2006
Rechatin et al, PRL102, 164801 2009
Leemans et al., PRL 2014
Scaling laws: toward kHz lasers

\[ a_0 > (\omega_0 / \omega_p)^{2/5} \quad k_p R = 2 \sqrt{a_0} \quad \pi c = 2 R / 3 \]

Laser pulse has to be resonant with plasma wave:
\[ R \approx \lambda_p / 2, \quad c \tau \approx \lambda_p / 2 \]

Laser energy scaling \[ E_L \propto \tau^3 \propto \lambda_p^3 \]
Electron energy gain \[ \Delta E \propto \tau^2 \propto \lambda_p^2 \]

30 fs \rightarrow 1 J \rightarrow 100 \text{ MeV-1 GeV}
3 fs \rightarrow \text{mJ} \rightarrow 1-10 \text{ MeV}

**Laser pulses of 5 fs, few mJ \rightarrow possible @ kHz**!

Lu et al., PRSTAB 10, 0613001 (2007)
100 keV kHz laser-plasma accelerator (2013-15)

30 fs (too long !), 3 mJ, $I = 3 \times 10^{18}$ W/cm$^2$, 1 kHz

He et al. NJP 15, 053016 (2013)
kHz electron source @ 100 keV

Emittance from GPT best fit to data
\( \varepsilon_N = 2 \times 10^{-2} \text{ mm.mrad} \)

Coherence length
\( L_{\text{coh}} = 5 \text{ nm} \)

Stability at kHz:
pointing < 400 \( \mu \)rad
- energy: 100 keV, < det. lim.
- charge 5 fC +/− 7%

---

Z. He et al., APL 102, 064104 (2013)
Set-up for electron diffraction experiment

20 nm single crystal Gold

Phosphor screen CsI(Tl)

Diffraction pattern

Z. He et al., APL 102, 064104 (2013)
Diffraction on single crystal Gold foil

laser-plasma source 95 keV

dE/E \approx 10 \% \rightarrow «\ streaked » Bragg peaks

\sin \theta = \frac{n\lambda}{2d} \propto \frac{1}{\sqrt{E}}

\rightarrow Temporal resolution along Bragg

low energy \rightarrow late

high energy \rightarrow early
Why Silicon?

Perfect sample:

- Simple structure
- Available samples (30 nm on 100x100 µm²)
- Extremely large photoinduced signal
  (what is it ?)
Time-resolved diffraction experiment on Si
→ streaking the Bragg peaks

30 nm Si nano membrane,
S. Scott, M. Lagally, Univ. Wisconsin

LPA driver:
10 mJ, 35 fs
800 nm, 0.5 kHz
Expected scenario: lattice heating and Debye-Waller effect on Bragg peaks

Estimated absorbed fluence: \( F_{\text{abs}} = 1.5 \text{ mJ/cm}^2 \)
Density of excited carriers: \( n_{\text{exc}} = 10^{21} \text{ cm}^{-3} \) (0.5% of valence electrons)

- Direct photoexcitation @ 3 eV
- Electron thermalization \( \approx 100 \text{ fs} \)
- Electron (hole) relaxation to bottom of conduction (valence) band \( \approx \text{ps} \)
  - Emission of phonons \( \rightarrow \) lattice heating (70 %)
    \( \Delta T = 200 \text{ K} \)
- Electron-hole pair recombination
  - Auger recombination \( > 100 \text{ ps} \)
    (in principle \( \sim 2 \text{ ps} \), but screening effect)
  - Further heating of the lattice (30 %)
    \( \Delta T = 300 \text{ K} \)
Effect of photo-excitation in Si

Estimated absorbed fluence: \( F = 1.5 \text{ mJ/cm}^2 \)
Density of excited carriers: \( n_{\text{exc}} = 10^{21} \text{ cm}^{-3} \) (0.5% of valence electrons)

400 ms exposure
We observe:
- A decrease of the 220 peak on ~ 3 ps time scale (el-phonon coupling)
- Long lived metastable state > 100 ps (hot Si)

Acquisition time: 10 min (limited by charge fluctuations)

Comparison between plasma source (streaking) and DC gun (stroboscopic)

Laser-plasma electron source (100 keV)

(220) peak

- Possible to retrieve picosecond dynamics

DC electron gun (50 keV)

(220) peak

- kHz operation and source stability was key to the success of experiment

→ Laser-plasma source can resolve dynamics in Silicon

How to improve the resolution?

$$\tau_{streak} = \frac{d}{v_z} \frac{m_e^2 c^4}{(E + m_e c^2)(E + 2m_e c^2)} \frac{\delta E}{E}$$

**PROS**

- Increase spectrometer resolution $\delta E/E$
- Decrease distance to sample $d$
- Increase electron energy $E$

**CONS**

- Lower signal
- Sample damage!
- Challenging

To reach sub-10 fs resolution, we chose the challenging option → 5 MeV, few femtosecond bunches @ kHz
Laser: 5 mJ (2.5 mJ on target)  
3.5 fs, kHz laser system

R. Lopez Martens’ group:  

From laser demonstration to implementation for experiments  
→ One full year of development / debugging
Relativistic intensities with 2.5 mJ kHz laser

**Full characterization in vacuum**

- 3 µm spot FWHM
- D-scan measurement*
- 3.4 fs FWHM
- I=3x10^{18} W/cm^2

Experimental set-up

Work of D. Guénot, D. Gustas, A. Vernier

Gas target is a continuously flowing capillary $N_2$ gas jet

- kHz operation
- Pumping issues!

Capillary $N_2$ source, $\phi = 100 \, \mu m$, $P_{\text{back}} \approx 20 \, \text{bars}$, $n_{e,\text{max}} \approx 1.5-2\times10^{20} \, \text{cm}^{-3}$

FOP scintillating screen

Removable magnets, $\phi = 2 \, \text{cm}$, $B_{\text{max}} = 88 \, \text{mT}$

Removable pinhole

Lens

Side imaging interferometry

Probe pulse

Accelerating pulse, 2-2.3 mJ, 3.4 fs, $I_{\text{max}} \approx 3\times10^{18} \, \text{W/cm}^2$
First MeV beams at kHz repetition rate
D. Guénot et al., accepted in Nature Photonics

**Divergence ~40 mrad**

![Graph showing divergence](image)

**Charge ~0.5 pC/shot**

**up to 1 pC/shot**

![Graph showing charge](image)
3D PIC simulations $\Rightarrow$ 1 fs electron beams!

Trapped electrons in non linear wakefield

Simulation results at the jet exit:
- 5 MeV beams
- 400 fC of charge
- 20 mrad divergence
- 1 fs duration
3D PIC simulations reproduce the results

CALDER-Circ simulations performed using experimental Parameters and measured laser spectrum and phase

Evolution of intensity
Injected charge

slight self-focusing
accelerated electrons originate from ionization of N\textsuperscript{5+} to N\textsuperscript{6+}
very local injection at peak laser intensity
Conclusions:

• Operate at kHz (with < 10 mJ) ? ✓
• Accelerate electrons to few MeV ? ✓
• High beam quality ✓
• Stable source ✓…
• Demonstration of UED (ps resolution) ✓
• Sub-10 fs duration ?

• Optimize this source (different gases, jets, CEP…)
• Beam transport (select energy bin, recompress bunch)

POTENTIAL APPLICATIONS
• Sub-10 fs electron diffraction
• Femtolysis of water / radio-biology…
Need for collaborations

Promote applications of our sources:

- Plasma source (5 MeV, pC, 1 fs ??)
- Conventional DC gun (300 fs, few fC, 50 keV)

- SAMPLES SAMPLES SAMPLES !!
  < 100 nm, 100x100 µm²

- Irradiation applications, radiolysis…
Collaborators

**Experiments: kHz laser-plasma source**
D. Gustas, I. Gonzalez, N. Zaim, B. Beaurepaire
A. Vernier, G. Gallé, D. Guénot, A. Lifschitz

**CUOS of University of Michigan:**
Z. He, A. Thomas, J. Nees, K. Krushelnick

**Laser system:**
M. Bocoum, F. Böhle, A. Jullien, J.-P. Rousseau, R. Lopez-Martens

**Beam transport**
B. van der Geer, Univ. Eindhoven, Pulsar Physics

**Sample preparation**
S. Scott, M. Lagally, Univ. Wisconsin
Outline of the talk

• Motivation / Introduction

• First laser-plasma accelerator at kHz and application to ultrafast electron diffraction

• First experiments with near-single-cycle laser pulses → Acceleration of ~ 5 MeV electrons at kHz

• Work in progress: ultrafast dynamics in Silicon, evidence of photo-induced dynamical diffraction effects
Comparison between plasma source (streaking) and DC gun (stroboscopic)

Laser-plasma electron source (100 keV): 30 nm sample

(220) peak

\[
\frac{I_1}{I_0} - 1 = 3.1 \pm 0.8 \text{ ps}
\]

DC electron gun (50 keV): 78 nm sample

(220) peak

\[
\frac{I_1}{I_0} - 1 = 3.0 \pm 0.2 \text{ ps}
\]

- Similar dynamics
- Much larger signals on 78 nm sample

→ Use conventional DC gun to explore the physics

Collaborators

**Experiments: kHz laser-plasma source**
B. Beaurepaire, D. Gustas
A. Vernier, G. Gallé, D. Guénot, A. Lifschitz

*CUOS of University of Michigan:*
Z. He, A. Thomas, J. Nees, K. Krushelnick

**Laser system:**
M. Bocoum, F. Böhle, A. Jullien, J.-P. Rousseau, R. Lopez-Martens

**Beam transport**
B. van der Geer, Univ. Eindhoven

**Sample preparation**
S. Scott, M. Lagally, Univ. Wisconsin