arXiv:2303.13169

Fourier-limited attosecond pulse generation from lightwave-driven electrons in solids

光電場駆動された固体電子によるFourier限界アト秒パルス発生

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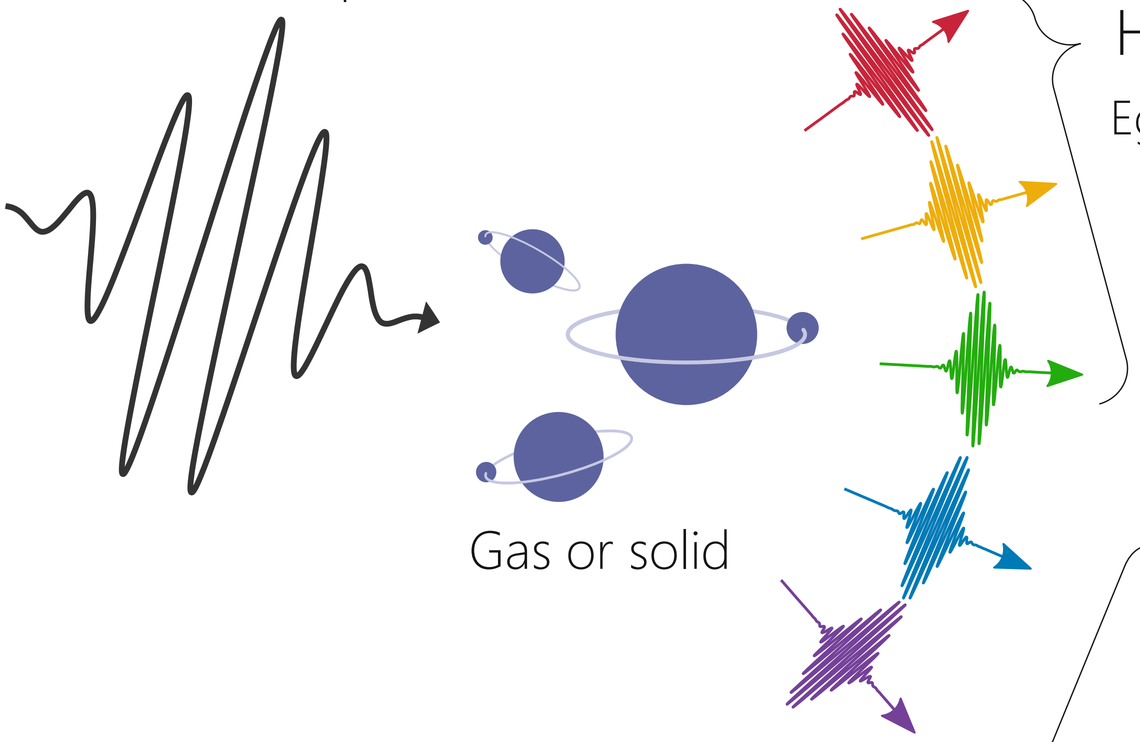
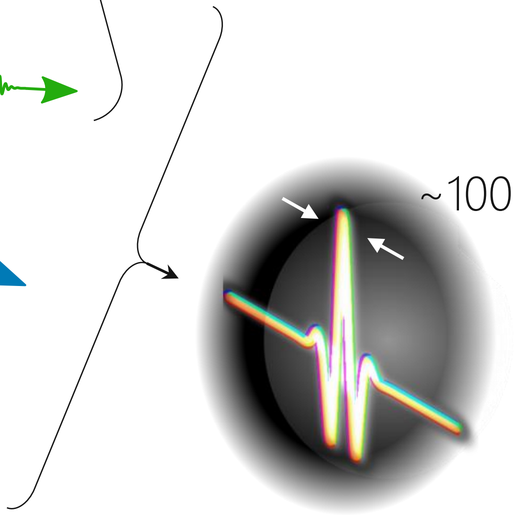
TQFT, August 28, 2023

Attosecond science and noneq. dynamics



High harmonic generation (HHG)

Eg) Ne gas: 20th to 100th harmonics
J. Macklin *et al.*, PRL **70**, 766 (1993).



M. Hentschel *et al.*, Nature **414**, 509 (2001).

Strong-field induced nonequilibrium dynamics ➡ Attosecond pulse generation

Attosecond world

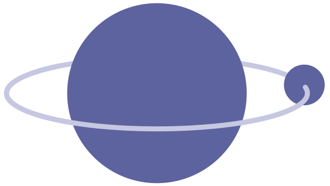
Time scale

Atto sec.

Femto sec.

Electron dynamics in atomic scale

Hydrogen atom

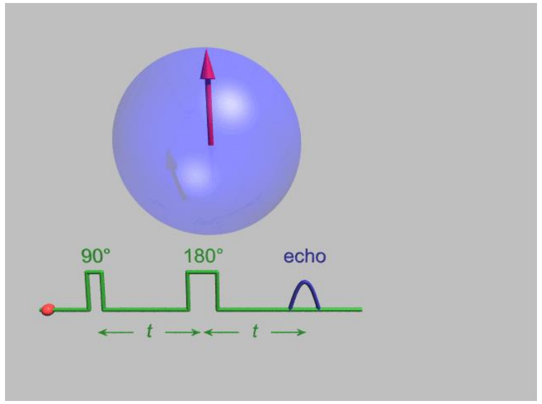


Orbital period ~ 160 as

Neutral pion decaying

$\pi^0 \to 2\gamma$ Lifetime ~ 85 as

Coherent dynamics in solids

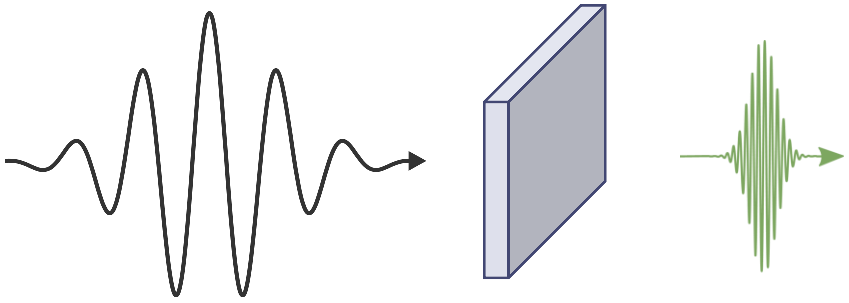


Spin echo, Wikipedia

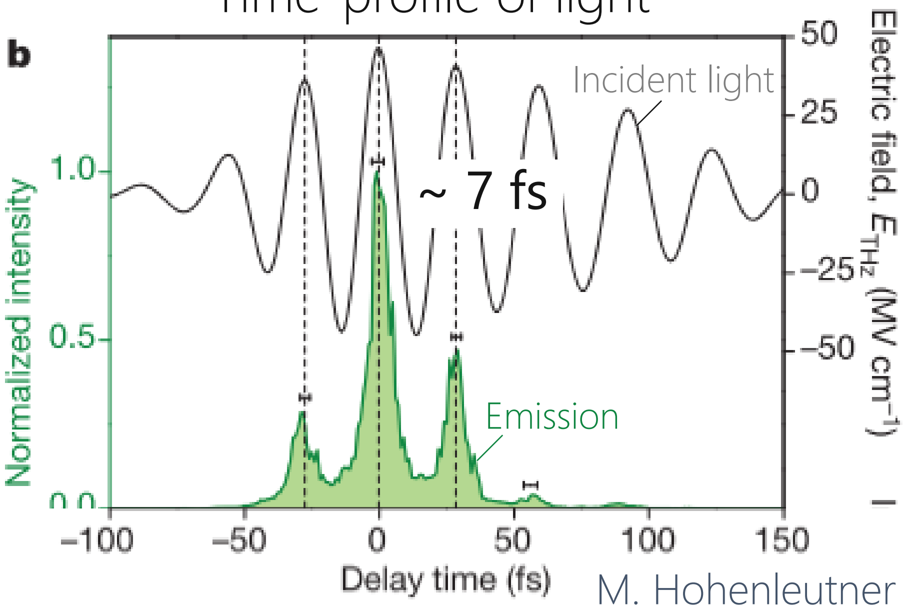
Relaxation of typical excited states ~ 10 fs

Observation of many-body dynamics before dephasing!

Attosecond science in solid materials



Time-profile of light



M. Hohenleutner *et al.*, Nature **523**, 572 (2015).

<u>Uncertainty relation</u>

$$\Delta E \cdot \Delta t \geq \hbar \approx 600 \text{ eV} \cdot \text{as}$$

~ 1 eV in typical solids

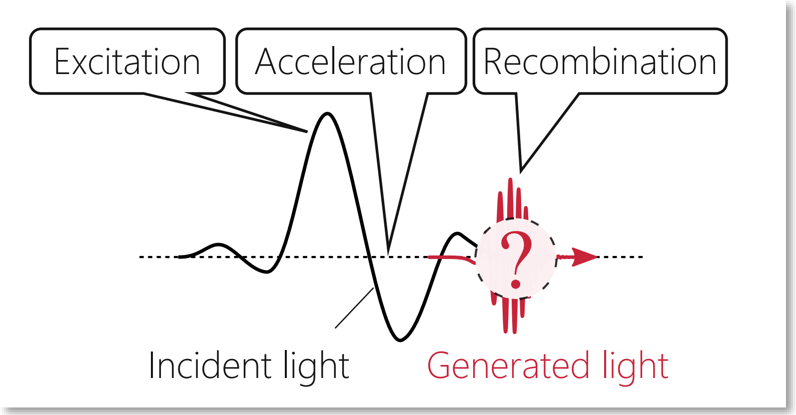
The only example: SiO_2 472 as M. Garg *et al.*, Nature **538**, 359 (2016).

➡ Need for Fourier-limited pulse generation

☐ Controlling the wave packet dynamics of electrons in solids

Emission from wave packet dynamics

3-step model P. B. Corkum, PRL **71**, 1994 (1993).

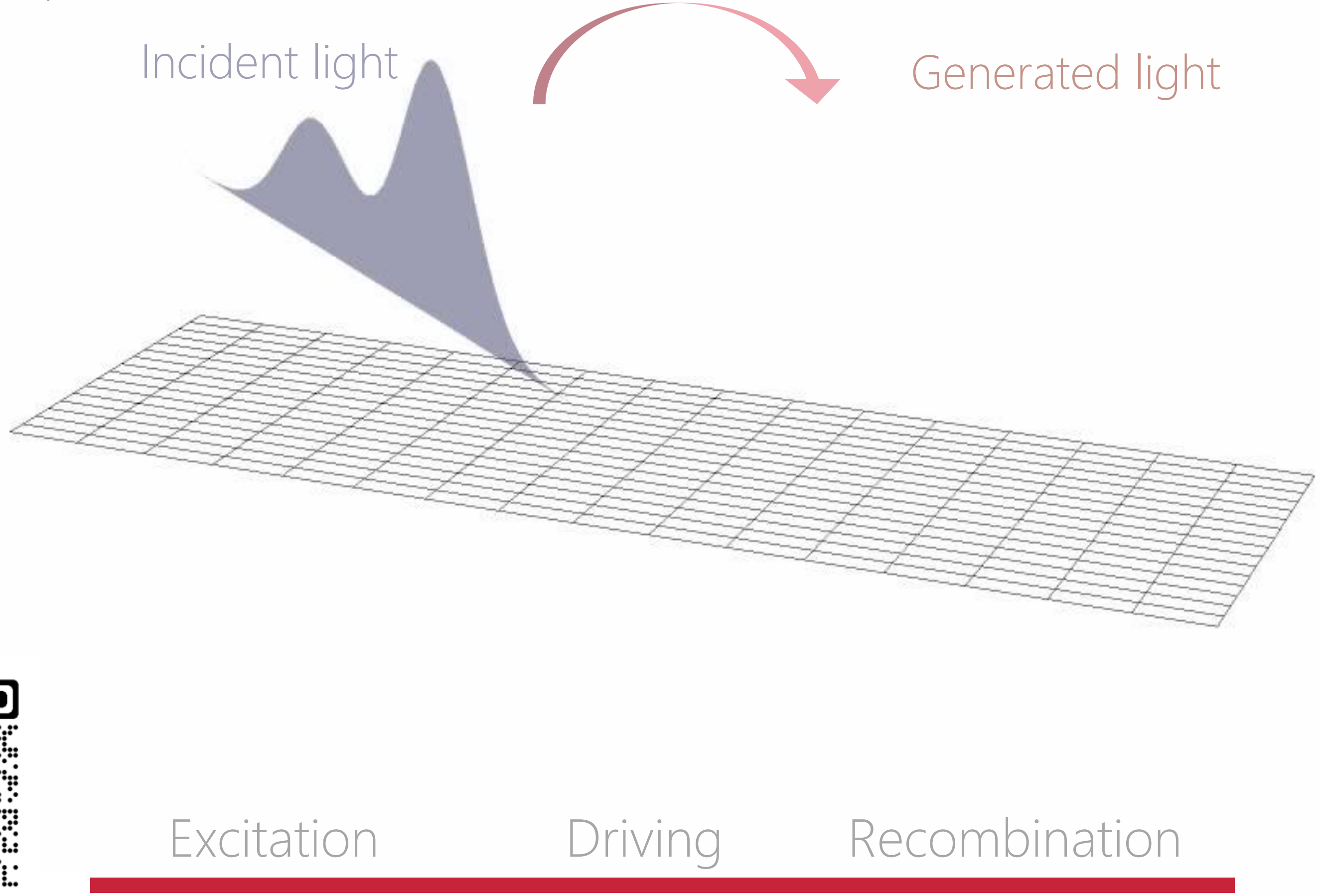


Strong-field excitation (tunneling)
→ Weak-field excitation

Echo generation from lightwave-driven electron in solids

SI, A. Ono, and S. Ishihara,
Phys. Rev. Res. **4**, 043155 (2022).

Reversed wave (echo) emission

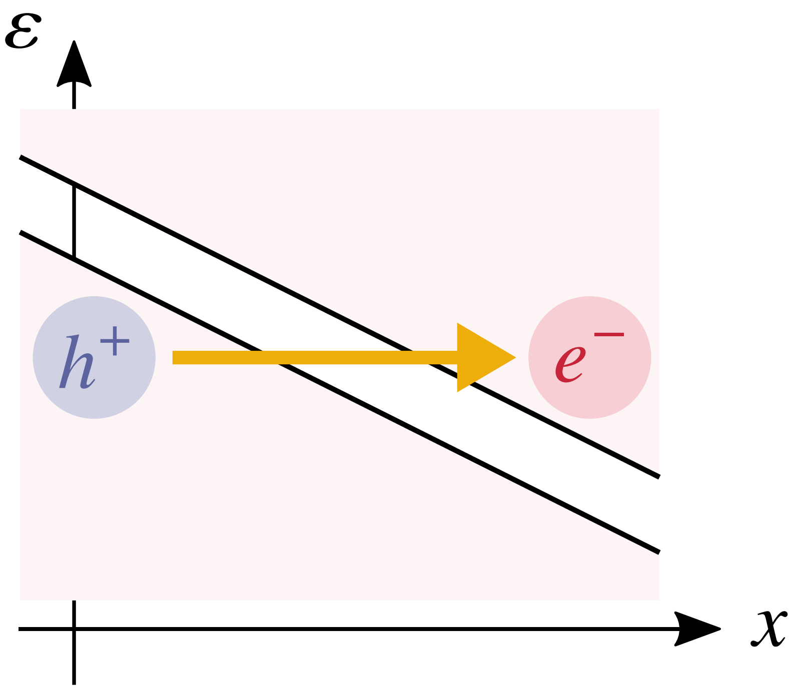


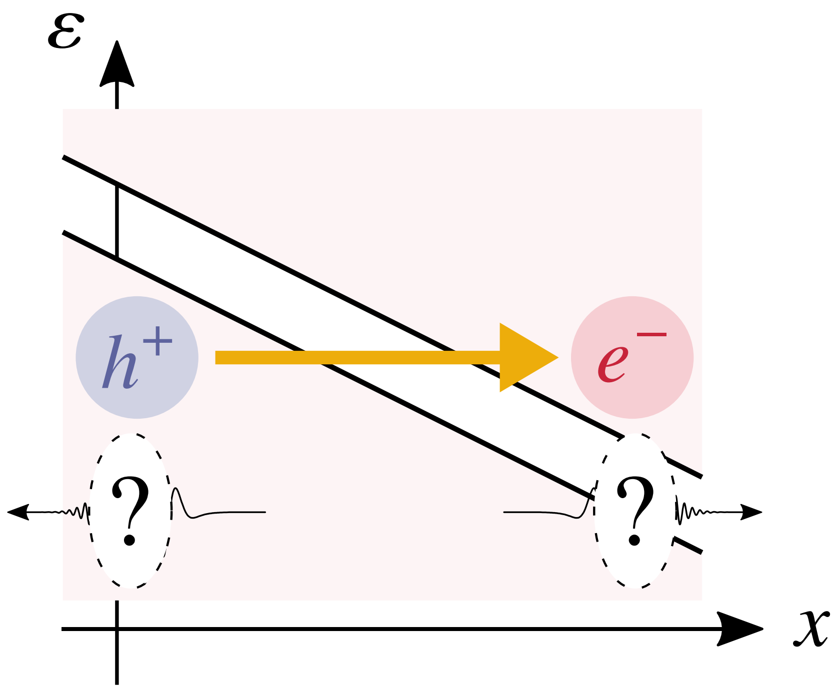
➡ Lightwave-driven dynamics of tunnel electron wave packets

Quantum tunneling by strong electric field

Landau–Zener tunneling (dielectric breakdown)

Schwinger effect





Tunneling probability $P = |\psi|^2 \sim \mathrm{e}^{-\Delta^2/v}$

➡ Need to clarify the transient dynamics of tunnel probability amplitudes!

Purpose

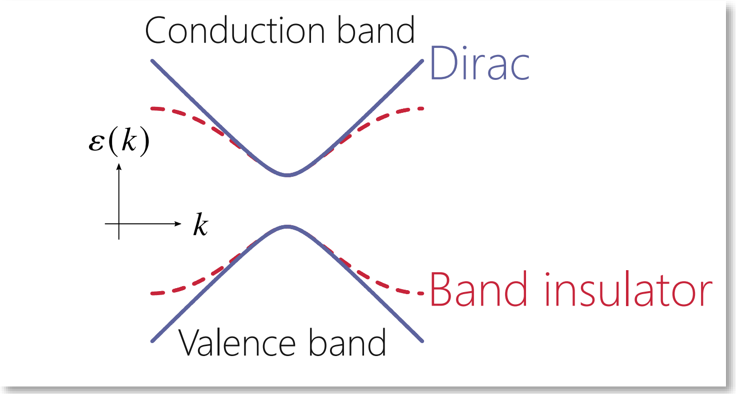
Attosecond pulse generation in solid materials

☐ Controlling the wave packet dynamics of tunnel electrons

Excitation + Driving

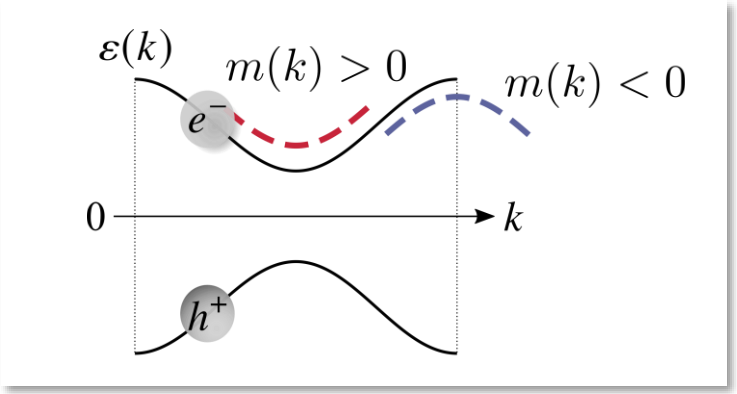
☐ What is the wave packet after tunneling?

Massive Dirac model in Sauter potential



☐ How is the wave packet driven?

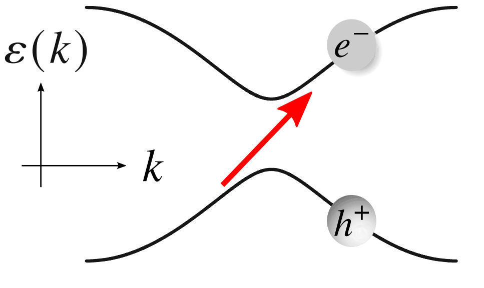
Sign change of effective mass

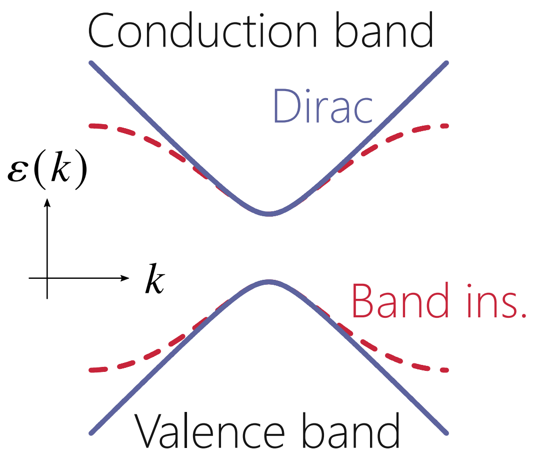


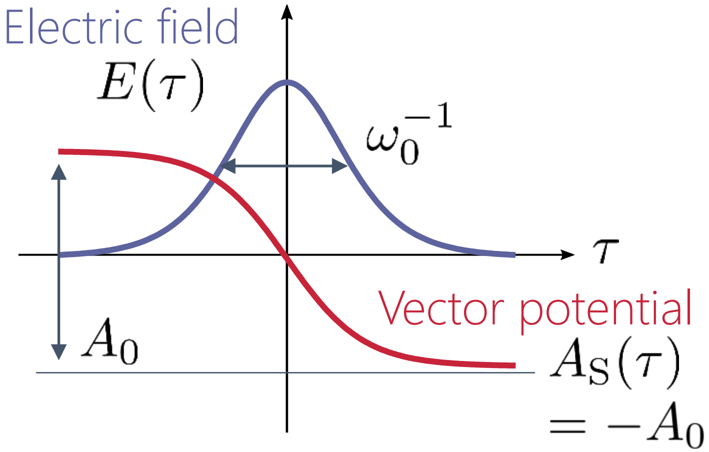
Systematic optimization of driving pulse waveforms achieves Fourier-limited pulse.

Ansatz of tunneling electron wave packets

Tunneling occurs near the gap minimum.







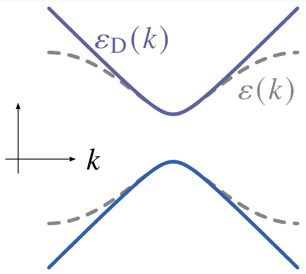
$$A_{\mathrm{S}}(\tau) = -A_0(1+\tanh(\omega_0\tau))/2$$

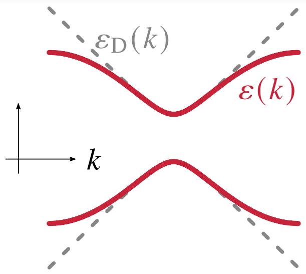
Electron–hole wavefunction

$$\overline{\psi_k^{\mathrm{v}}}\psi_k^{\mathrm{c}}(\tau \gg 0) = |\psi_k^{\mathrm{v}}||\psi_k^{\mathrm{c}}| \frac{G(\mu+\nu-\lambda)\,G(-\mu+\nu+\lambda)\,G(-\mu+\nu-\lambda)}{G(-\mu-\nu-\lambda)\,G(2\nu)^2} \mathrm{e}^{-\mathrm{i}\varepsilon_{\mathrm{D}}(k+qA_0)\tau}$$

N. B. Narozhnyi and A. I. Nikishov, Yadem. Fiz. 11: 1072-7 (1970). $G(y) = \sqrt{\frac{y\sinh(\pi y)}{\pi}}\,\Gamma(\mathrm{i}y)$

$$\to |\psi_k^{\mathrm{v}}||\psi_k^{\mathrm{c}}| \frac{G(\mu+\nu-\lambda)\,G(-\mu+\nu+\lambda)\,G(-\mu+\nu-\lambda)}{G(-\mu-\nu-\lambda)\,G(2\nu)^2} \mathrm{e}^{+\mathrm{i}\int_0^\infty \mathrm{d}\tau' \left(\varepsilon_{\mathrm{D}}(k-qA_{\mathrm{S}}(\tau'))-\varepsilon_{\mathrm{D}}(k+qA_0)\right)}\ \mathrm{e}^{-\mathrm{i}\int_0^\tau \mathrm{d}\tau' \varepsilon(k-qA(\tau'))}$$

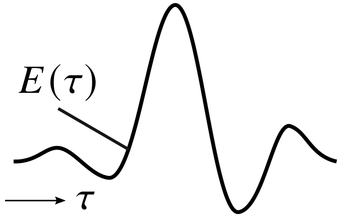




Tunneling process and dispersion relation of Bloch electrons are considered.

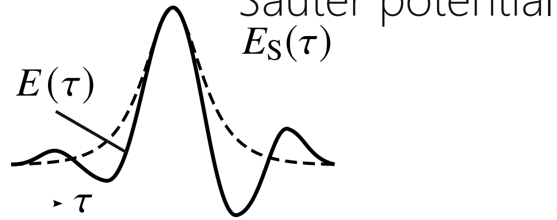
Optimization of driving-pulse waveform

1. Assume an initial waveform of an external field



⬇

2. Fit the Sauter potential to $\boldsymbol{E(\tau)}$'s peak



⬇

3. Calculate the wavefunction of tunneling electron–hole pairs $\overline{\psi_k^v}\psi_k^c(\tau)$

⬇

4. Modify the waveform to make the phase of the wavefunction constant in $\boldsymbol{k}$ by Gauss–Newton method

The analytical expression of the wavefunction facilitates the systematic optimization.

Model & Method

One-dimensional two-orbital tight-binding model

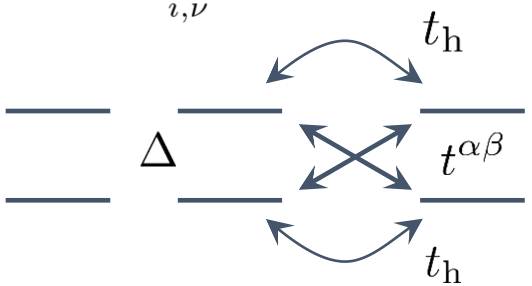
$$H = -\sum_{\langle i,j\rangle}\sum_{\nu,\nu'=\alpha,\beta} t^{\nu\nu'} c^\dagger_{\nu i} c_{\nu' j} + \sum_{i,\nu} D_\nu c^\dagger_{\nu i} c_{\nu i}$$

Transfer integral

$t_\mathrm{h} \equiv t^{\alpha\alpha} = t^{\beta\beta}$

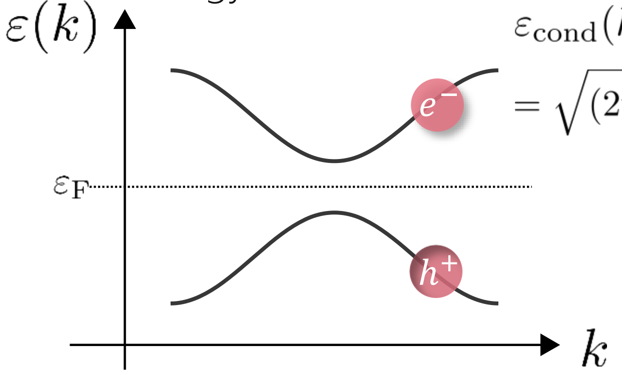
Onsite potential

$\Delta/2 \equiv D_\alpha = -D_\beta$



$t^{\alpha\beta} = 3t_\mathrm{h}$
$\Delta = 3t_\mathrm{h}$

Energy band structure



$$\varepsilon_\mathrm{cond}(k) = -\varepsilon_\mathrm{valn}(k) = \sqrt{(2t^{\alpha\beta}\cos(k))^2 + (\Delta/2)^2}$$

Dirac parameters

$mv^2 = \Delta/2$
$v = 2t^{\alpha\beta}$

Time-dependent external field

Peierls substitution $t_\mathrm{h} \to t_\mathrm{h} \mathrm{e}^{-\mathrm{i}A(\tau)}$

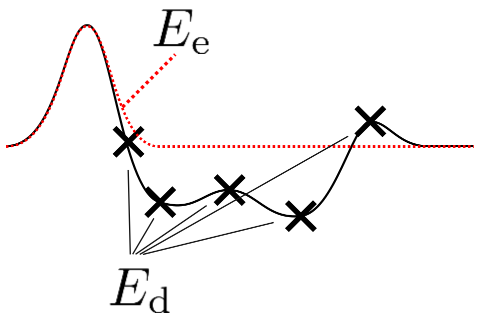
Vector potential $A(\tau) = -\int_{-\infty}^{\tau} E(\tau')d\tau'$

Electric current operator $\hat{J}(\tau) = -\frac{1}{N}\frac{\delta H(\tau)}{\delta A(\tau)}$

$$A(\tau) = A_\mathrm{e}(\tau) + A_\mathrm{d}(\tau)$$

Excitation $A_\mathrm{e}(\tau) = -A_{\mathrm{e}0}\frac{1+\mathrm{erf}(\omega_\mathrm{e}\tau/\sqrt{2})}{2}$

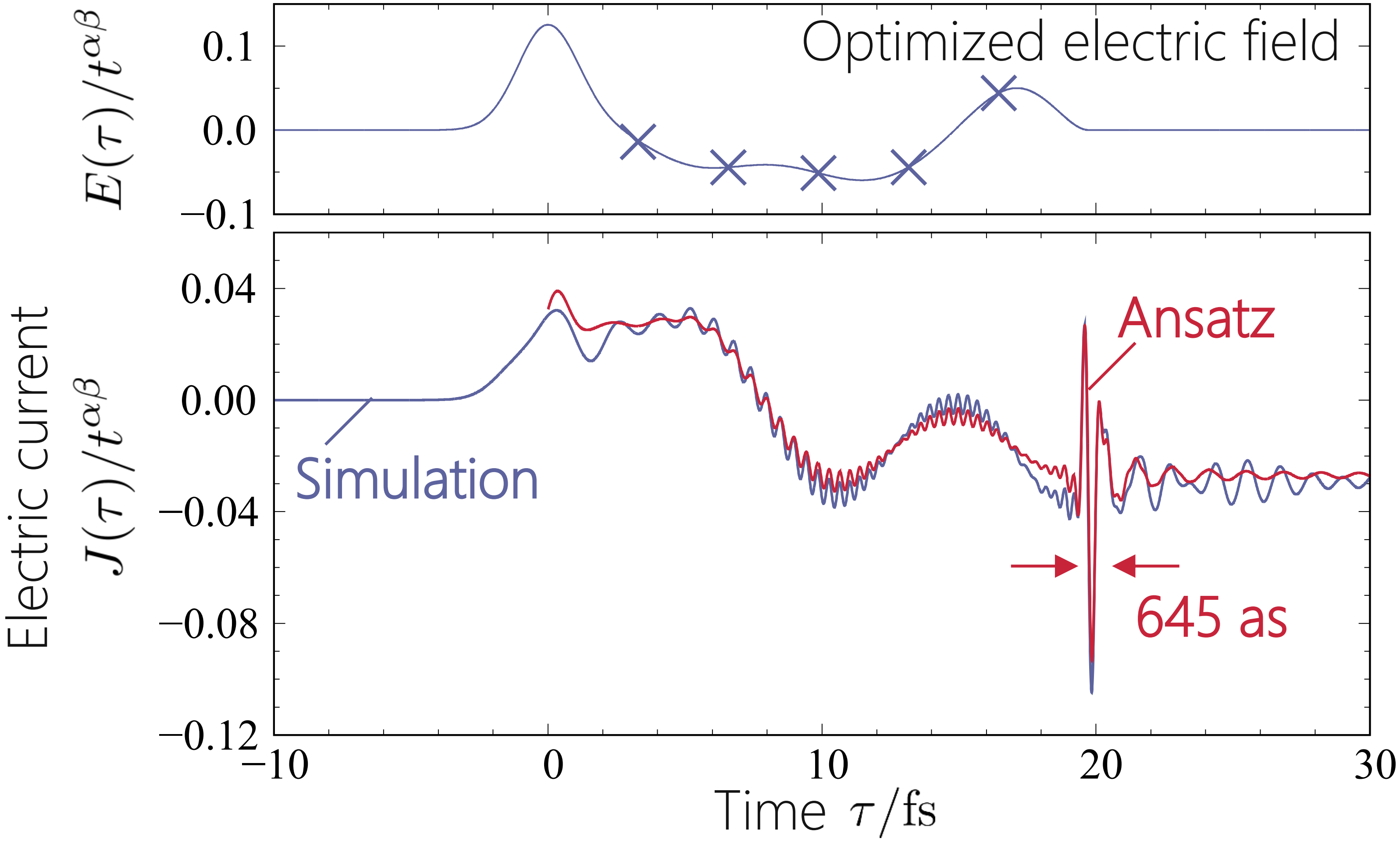
Driving $\{A_\mathrm{d}(\tau_j) \mid j = 0, 1, \cdots, M\}$



Numerical methods

Diagonalization (Wave number points $N = 1000$)

Time profile of the optimized and emitted pulses



$E_{\mathrm{peak}} \sim 9\ \mathrm{MV/cm}$

Energy gap $\sim 3\ \mathrm{eV}$

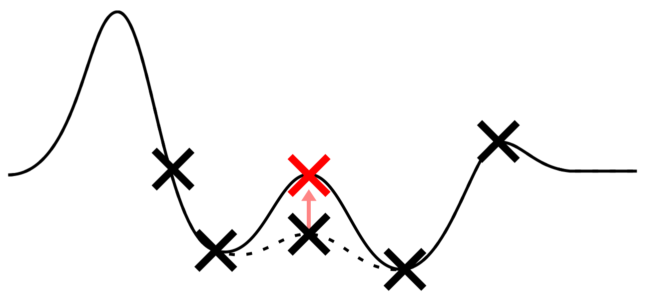
Band width $\sim 10\ \mathrm{eV}$

Our ansatz describes the transient state of tunneling electrons under optical driving.

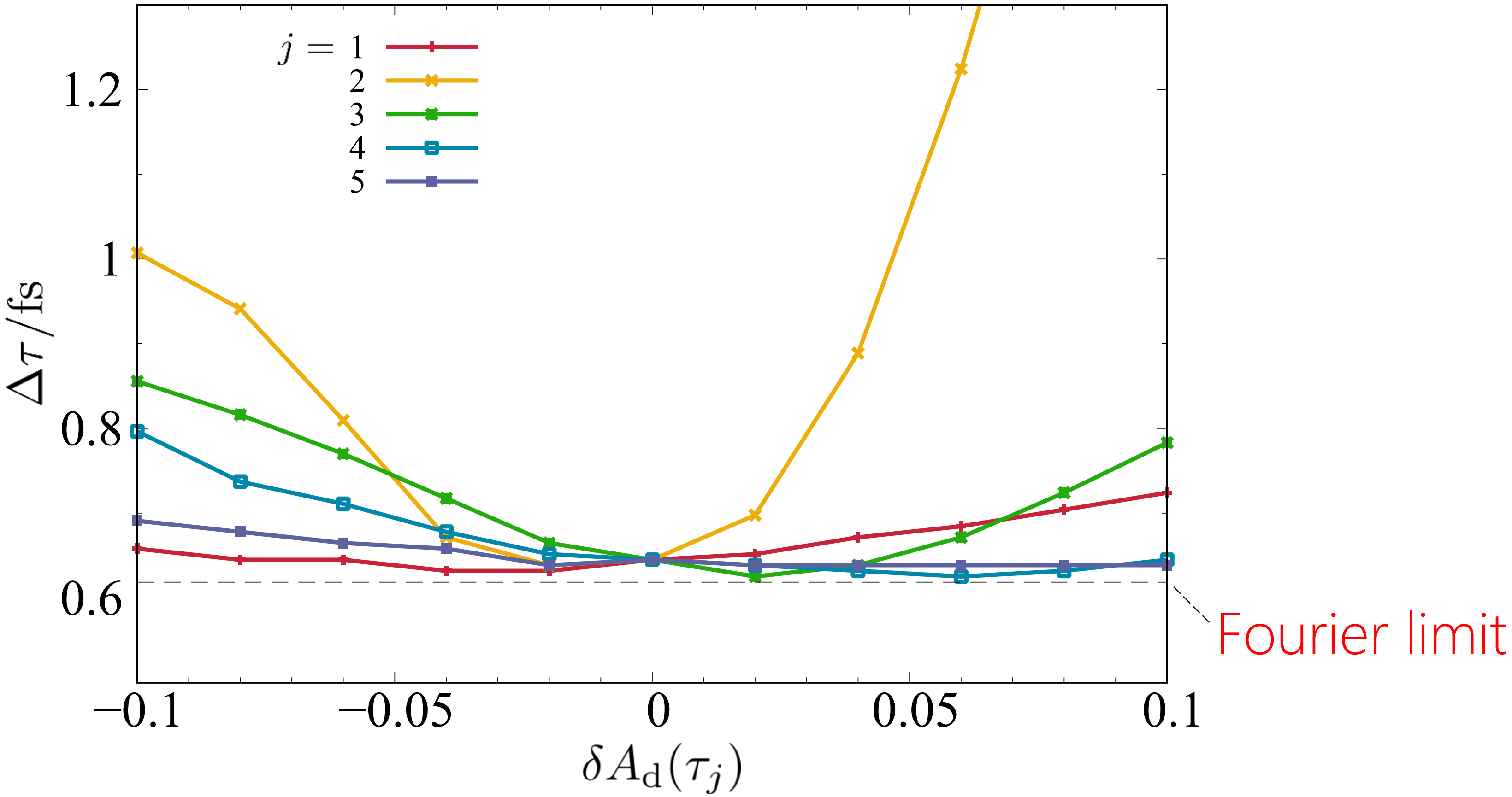
Sensitivity of pulse width

Deviation

$A_\mathrm{d}(\tau_j) \to A_\mathrm{d}(\tau_j) + \delta A_\mathrm{d}(\tau_j)$



FWHM of electric currents

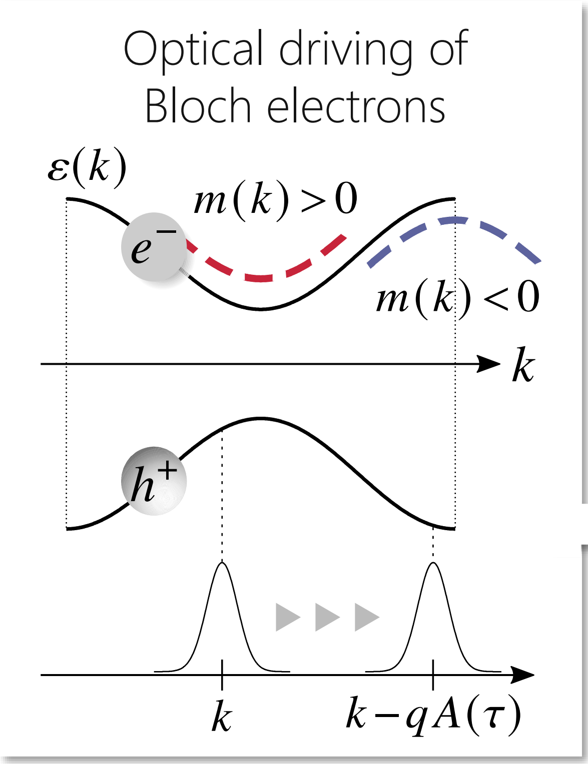


Duration approaches Fourier limit.

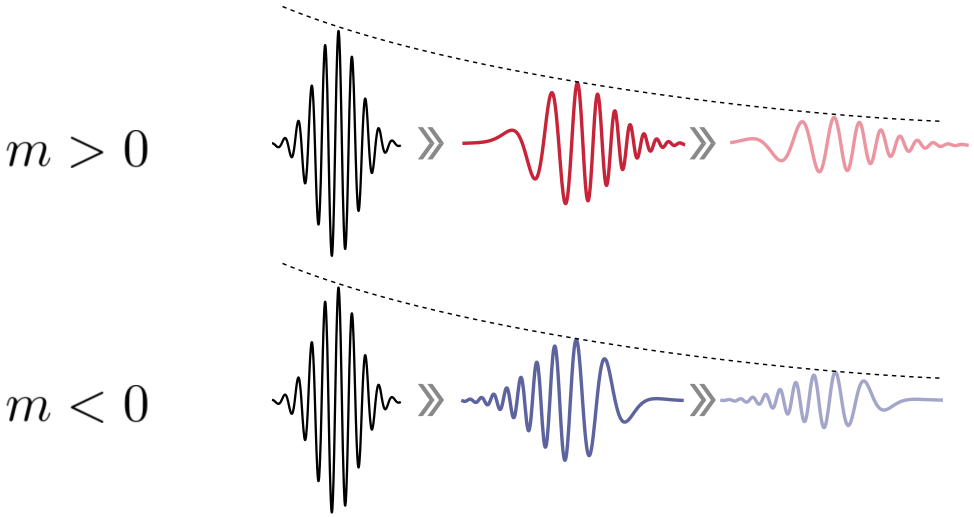
Allowable error can be estimated.

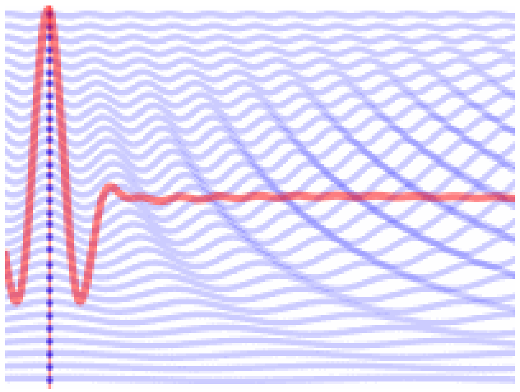
Mechanism of pulse compression

Wave packet dynamics of particles with finite mass



Dynamic sign change of mass





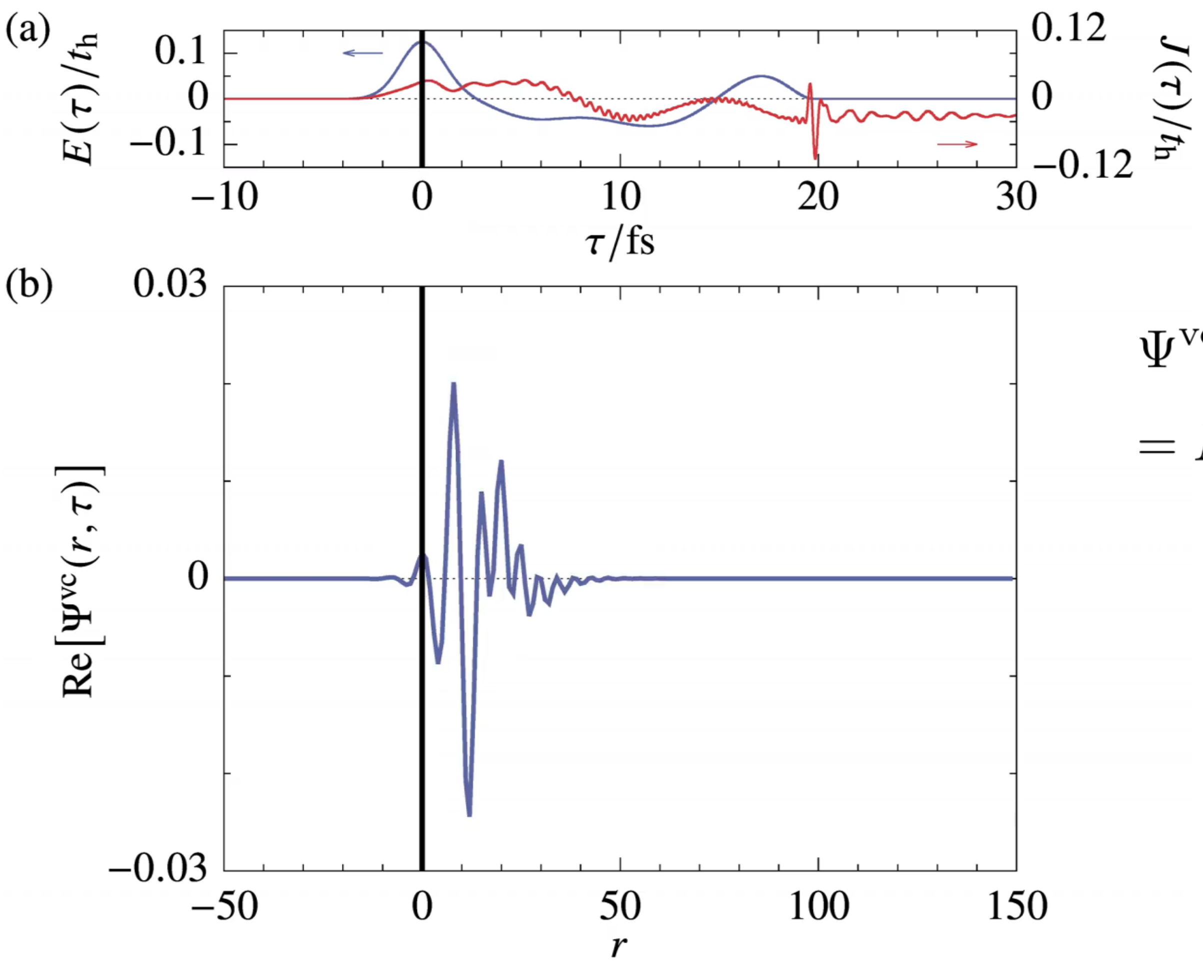
Wave packet, Wikipedia

$m(k) > 0$ $m(k) < 0$

Dynamic mass-sign change allows for the refocus of wave packets.

arXiv:2303.13169. E-mail: shohei.imai@phys.s.u-tokyo.ac.jp

Real-space wave packet dynamics



$$\Psi^{\mathrm{vc}}(r,\tau) = N^{-1}\sum_R \psi^{\mathrm{vb}}(R - r/2,\tau)^* \psi^{\mathrm{cb}}(R + r/2,\tau)$$

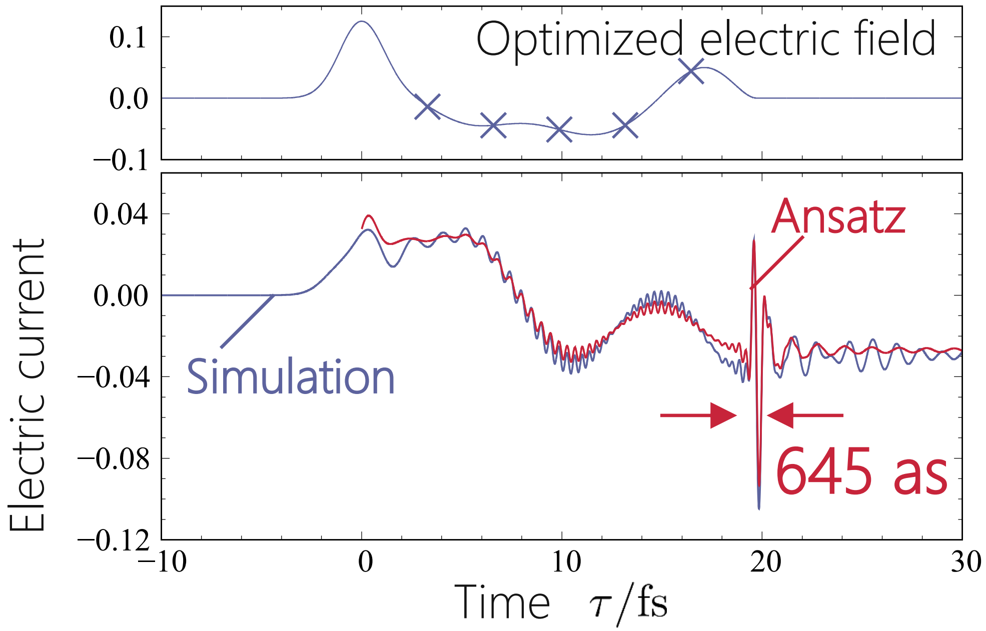
Relative displacement　Central position

Refocusing of the electron wave packet can be achieved by optimal driving.

Summary

☑ What is the wave packet of tunneling electrons?

Our ansatz reveals the waveform (wavefunction) of tunneling electrons.



☑ How is the wave packet driven?

Dynamic sign change of the mass shortens the emitted pulse duration.

☑ What is the optimal pulse waveform for attosecond pulses?

Systematic optimization of driving pulse waveforms achieves Fourier-limited pulse.

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