Divide-&-Conquer Maxwell-Ehrenfest-Surface Hopping (DC-MESH)

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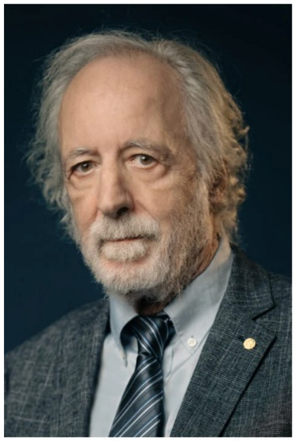
Goal: To describe multiscale light-matter interaction ranging from atto-to-nano seconds & pico-to-micro meters

Linker *et al*., *Science Adv*. **8**, eabk2625 ('22)
Razakh *et al*., *PDSEC* (IEEE, '24)

Dawn of Attosecond Physics

The Nobel Prize in Physics 2023



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Pierre Agostini

Prize share: 1/3

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Ferenc Krausz

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Anne L'Huillier

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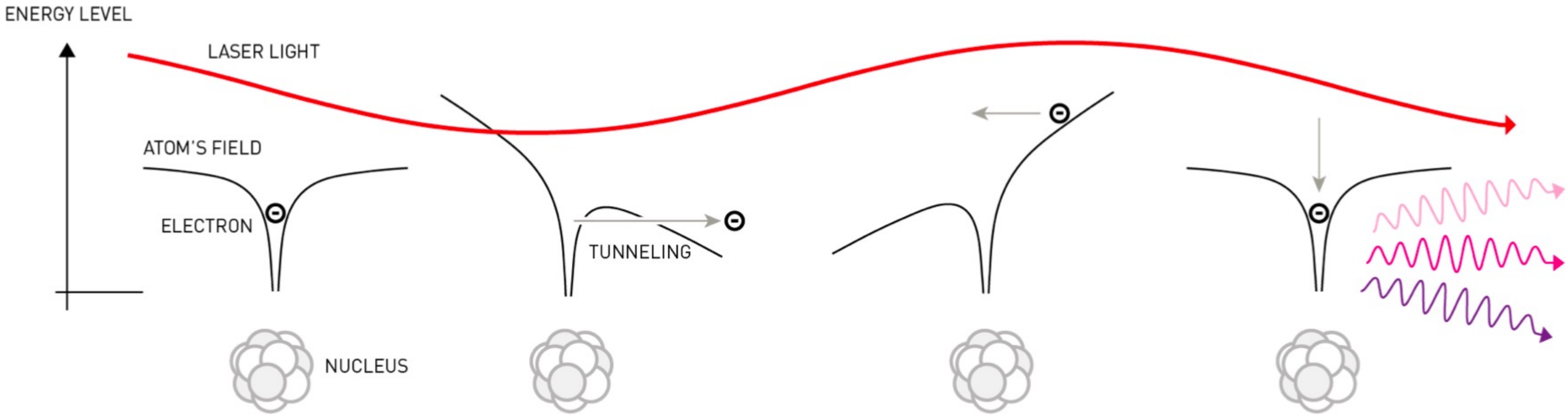
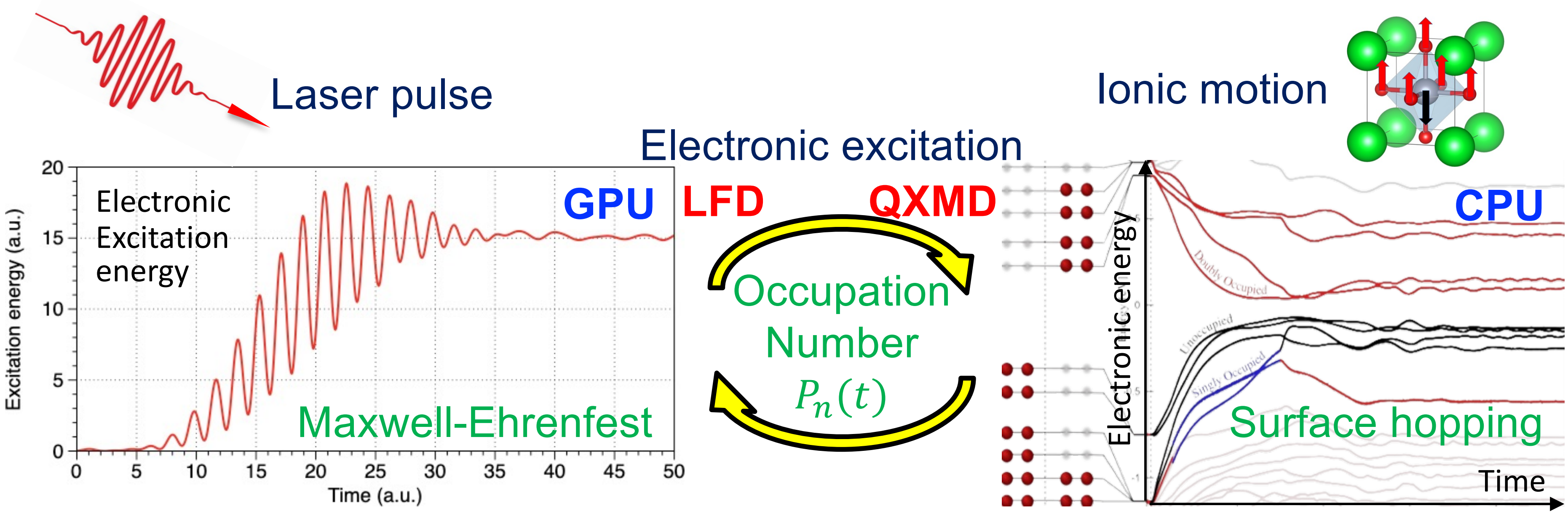
The Nobel Prize in Physics 2023 was awarded to Pierre Agostini, Ferenc Krausz and Anne L'Huillier "for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter"



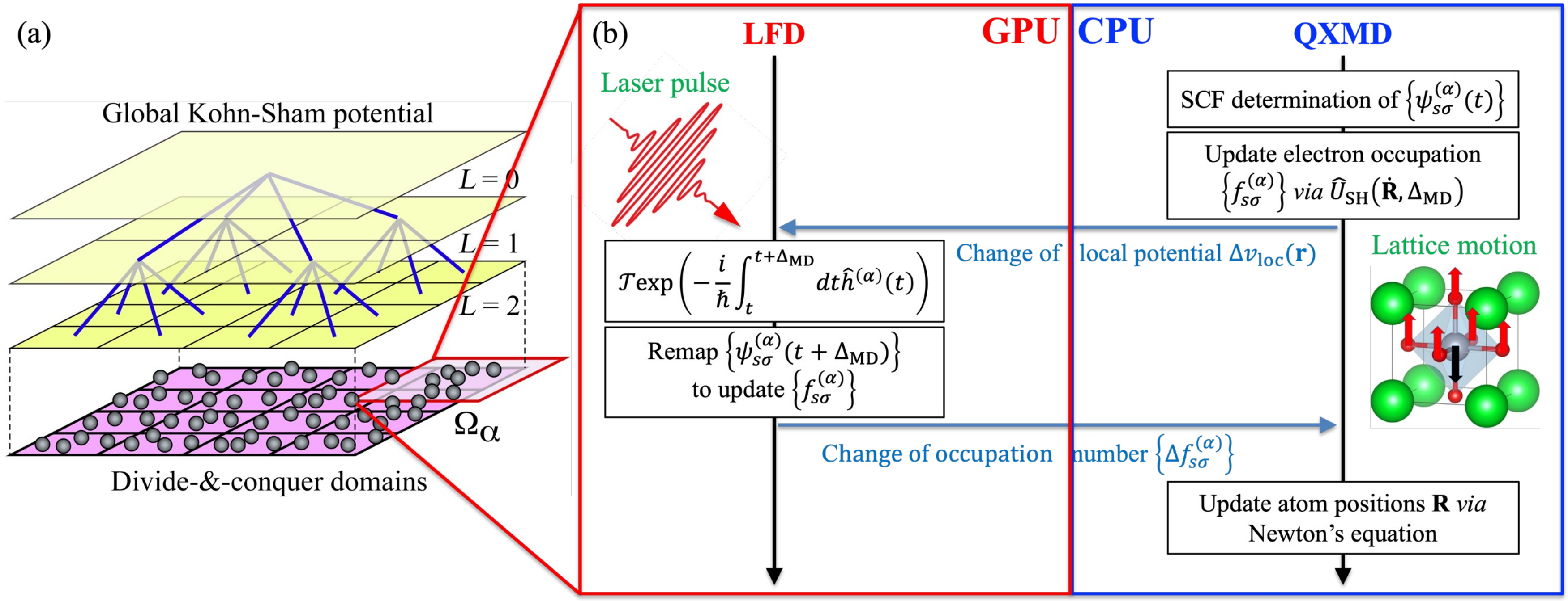
Nonadiabatic Quantum MD: DC-MESH

- **DC-MESH (divide-&-conquer Maxwell + Ehrenfest + surface-hopping): *O*(*N*) algorithm to simulate photo-induced quantum materials dynamics**
- **LFD (local field dynamics): Maxwell equations for light & real-time time-dependent density functional theory equations for electrons to describe light-matter interaction**
- **QXMD (quantum molecular dynamics with excitation): Nonadiabatic coupling of excited electrons & ionic motions based on surface-hopping approach**
- **"Shadow" LFD (GPU)-QXMD (CPU) handshaking *via* electronic occupation numbers with minimal CPU-GPU data transfer**
- **GSLD: Globally sparse (interdomain Hartree coupling *via* multigrid) & locally dense (intradomain nonlocal exchange-correlation computation *via* BLAS) solver**



Linker *et al*., *Science Adv*. **8**, eabk2625 (2022); Razakh *et al*., *PDSEC* (IEEE, '24)

Divide-Conquer-Recombine (DCR)



- **Treat multi-physics at appropriate scales & levels of approximation**
- **Hartree potential & electromagnetic field are computed globally using the scalable *O*(*N*) multigrid method & macroscopic grid, respectively**
- **Higher-order correlations represented by the exchange-correlation (XC) kernel in time-dependent density functional theory (TDDFT) are treated locally within each divide-&-conquer (DC) domain since they are known to be short-ranged** [Nakano & Ichimaru, *Phys. Rev. B* **39**, 4930 ('89)]

- **See notes on** [dynamic correlation], [DCR-NAQMD], [embedded TDDFT], and [Ehrenfest-hopping dynamics (EHD)]

LFD Algorithm

- **Hamiltonian in the α-th domain** [Yabana, *Phys. Rev. B* **85**, 045134 ('12); Jestadt, *Adv. Phys*. **68**, 225 ('19)]

$$\hat{h}(t,\mathbf{R}(t)) = \overbrace{\frac{1}{2}\left(\frac{\nabla}{i}+\frac{1}{c}\mathbf{A}(\mathbf{r}_\alpha,t)\right)^2 - \phi(\mathbf{r}_\alpha,t) + \hat{v}_{\mathrm{xc}} + v_{\mathrm{ion}}(\mathbf{r},\mathbf{R})}^{\hat{h}_{\mathrm{el}}(t)} + \overbrace{\Delta\dot{\mathbf{R}}\bullet\frac{\partial}{\partial\mathbf{R}}v_{\mathrm{ion}}}^{\hat{h}_{\mathrm{el-ion}}}$$

Electromagnetic vector & scalar potentials at the α-th domain

Nonadiabatic coupling

- **Trotter expansion of time propagator**

$$\exp(-i\hat{h}\Delta_{\mathrm{MD}}) \cong \underbrace{\exp(-i\hat{h}_{\mathrm{el-ion}}\Delta_{\mathrm{MD}}/2)}_{\text{QXMD}}\,\underbrace{\mathcal{T}\exp\left(-i\int_t^{t+\Delta_{\mathrm{MD}}} dt\hat{h}_{\mathrm{el}}(t)dt\right)}_{\text{LFD}}\,\underbrace{\exp(-i\hat{h}_{\mathrm{el-ion}}\Delta_{\mathrm{MD}}/2)}_{\text{QXMD}}$$

- **Self-consistent propagator** [Sato, *J. Chem. Phys*. **143**, 224116 ('15); Lian, *Adv. Theo. Sim*. **1**, 1800055 ('18)]

$$\mathcal{T}\exp\left(-i\int_t^{t+\Delta_{\mathrm{MD}}} dt\hat{h}_{\mathrm{el}}(t)\right) \cong \prod_{n=1}^{N_{\mathrm{QD}}=\Delta_{\mathrm{MD}}/\Delta_{\mathrm{QD}}} \exp\left(-i\Delta_{\mathrm{QD}}\hat{h}_{\mathrm{el}}\left(t+\left(n-\frac{1}{2}\right)\Delta_{\mathrm{QD}}\right)\right)$$

- **Nonlocal exchange-correlation propagator** [Vlcek, *J. Chem. Phys*. **150**, 184118 ('19)]

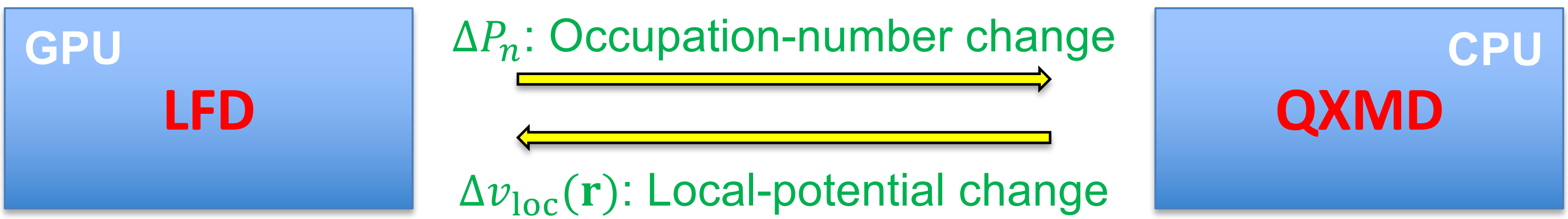
$$\exp(-i\Delta_{\mathrm{QD}}\hat{h}_{\mathrm{el}}) \cong \frac{1-i\hat{v}_{\mathrm{nl}}\Delta_{\mathrm{QD}}/2}{\left\|\left(1-i\hat{v}_{\mathrm{nl}}\Delta_{\mathrm{QD}}/2\right)|\psi_n(t)\rangle\right\|}\exp(-i\Delta_{\mathrm{QD}}\hat{h}_{\mathrm{loc}})\frac{1-i\hat{v}_{\mathrm{nl}}\Delta_{\mathrm{QD}}/2}{\left\|\left(1-i\hat{v}_{\mathrm{nl}}\Delta_{\mathrm{QD}}/2\right)|\psi_n(t)\rangle\right\|}$$

$$\hat{v}_{\mathrm{xc}} = \underbrace{\hat{v}_{\mathrm{loc}}}_{\text{local}} + \underbrace{\hat{v}_{\mathrm{nl}}}_{\text{nonlocal}};\quad \hat{h}_{\mathrm{el}} = \hat{h}_{\mathrm{loc}} + \hat{v}_{\mathrm{nl}}$$

- **See note on** self-consistent time propagator

Reduced-Communication Shadow Dynamics

- **Fundamental physics equations are local at the finest spatiotemporal scales, *i.e.*, simple partial differential equations with differential operators acting locally in a data-parallel fashion — LFD fits naturally to GPU; on the other hand, coarse-grained schemes to approximately describe complex chemical interactions often come with an excessive computational cost of nonlocal operations in space and time — QXMD takes advantage of complex instruction sets in CPU**
- **At each molecular-dynamics step, LFD informs QXMD of occupation-number change due to light-electron & electron-electron interactions**
- **QXMD performs excited-state quantum molecular dynamics & informs LFD of local-potential change for the next N_{QD} $(= \Delta_{\mathrm{MD}}/\Delta_{\mathrm{QD}})$ quantum-dynamics steps**
- **"Shadow" electronic wave functions in LFD are resident on GPU, while QXMD wave functions on CPU, to minimize CPU-GPU data transfers**



Shadow dynamics: real-time time-dependent density functional theory (RT-TDDFT)

Pseudopotentials: projector augmented wave (PAW), ultrasoft (Vanderbilt)

Exchange-correlation functionals: metaGGA (SCAN), hybrid exact-exchange (HSE *etc.*), DFT+U, DFT-D, nonlocal vdW

- **See note on** shadow EHD

Data-Parallel & BLASified LFD

- **Data-parallel local LFD: Auxiliary-field electronic time propagator for local potential** [Car & Parrinello, *Solid State Commun*. 62, 403 ('87); Nakano *et al*., *Comput. Phys. Commun*. **83**, 181 ('94)] **on real-space mesh achieves high performance on GPU**

- **BLASified nonlocal LFD: Operation of nonlocal potential is projected onto a vector space spanned by Kohn-Sham orbitals at time 0 within the real-time scissor approximation** [Wang *et al*., *J. Phys. Condens. Mat*. **31**, 214002 ('19)]**, making it dense matrix operations implemented with highly optimized level3 (or matrix-matrix) BLAS (basic linear algebra subprogram) library on GPU**

$$\hat{v}_{\mathrm{nl}}|\psi_n(t)\rangle \cong \Delta_{\mathrm{sci}} \sum_{m \geq \mathrm{LUMO}} |\psi_m\rangle\langle\psi_m|\psi_n(t)\rangle$$

Razakh *et al*., *PDSEC* (IEEE, '24); Nariman *et al*., *PMBS* (IEEE, '24)

- **See notes on** [auxiliary-field electron propagator] and [real-time scissor]

Global Maxwell's Equations

- **Global Maxwell's equations are solved on a macroscopic grid**
  Yabana, *Phys. Rev. B* **85**, 045134 ('12); *cf.* Gabay, *Phys. Rev. B* **101**, 235101 ('20)

$$\mathbf{A} = \mathbf{A}_{\text{ext}} + \mathbf{A}_{\text{ind}} + \mathbf{A}_{\text{xc}}$$

$$\begin{cases} \left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \frac{\partial^2}{\partial \mathbf{R}^2}\right)\mathbf{A}_{\text{ind}} = \frac{4\pi}{c}\mathbf{J} & \text{Induced vector potential} \\ \left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \frac{\partial^2}{\partial \mathbf{R}^2}\right)\phi = \frac{4\pi}{c}\rho & \text{Scalar potential} \end{cases}$$

- **Local domain-averaged current & charge densities**

$$\mathbf{J}(\mathbf{R}_\alpha, t) = \frac{1}{\Omega_\alpha}\int_{\Omega_\alpha} d\mathbf{r}\mathbf{j}(\mathbf{r}, t)$$

$$\rho(\mathbf{R}_\alpha, t) = -\frac{1}{\Omega_\alpha}\int_{\Omega_\alpha} d\mathbf{r}n(\mathbf{r}, t)$$

$$\mathbf{j}(\mathbf{r}, t) = -\sum_{n\sigma} \text{Re}\left[\psi_{n\sigma}^*(\mathbf{r}, t)\frac{\nabla}{i}\psi_{n\sigma}(\mathbf{r}, t)\right] f_{n\sigma} - \frac{1}{c}\mathbf{A}(\mathbf{r}, t)n(\mathbf{r}, t)$$

$$n(\mathbf{r}, t) = \sum_{n\sigma}\left|\psi_{n\sigma}(\mathbf{r}, t)\right|^2 f_{n\sigma}$$

- **Long-range correction in time-dependent current density functional theory (TDCDFT)** [Vignale, *Phys. Rev. Lett.* **77**, 2037 ('96); Maitra, *Phys. Rev. B* **68**, 045109 ('03); Sun, *Phys. Rev. Lett.* **127**, 077401 ('21)]

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \frac{\partial^2}{\partial \mathbf{R}^2}\right)\mathbf{A}_{\text{xc}} = -\frac{\alpha}{c}\mathbf{J} \quad \text{Exchange–correlation vector potential}$$

- **See note on** Maxwell solver and exciton dynamics