中国科学院物理研究所
Institute Of Physics Chinese Academy Of Sciences

超快物質科學中心
Center for Ultrafast Physics

First-principles Studies of Ultrafast Dynamics in the Condensed Matter Systems

Advanced Attosecond Laser Infrastructure (AALI)

汇报人：廉超

时间：2026-5-24

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含时薛定谔方程

量子力学中，系统的演化由含时薛定谔方程描述：

$$i\hbar\frac{\partial}{\partial t}\Psi(\mathbf{r},t)=\hat{H}\Psi(\mathbf{r},t)$$

对于单粒子系统，哈密顿量通常形式为：

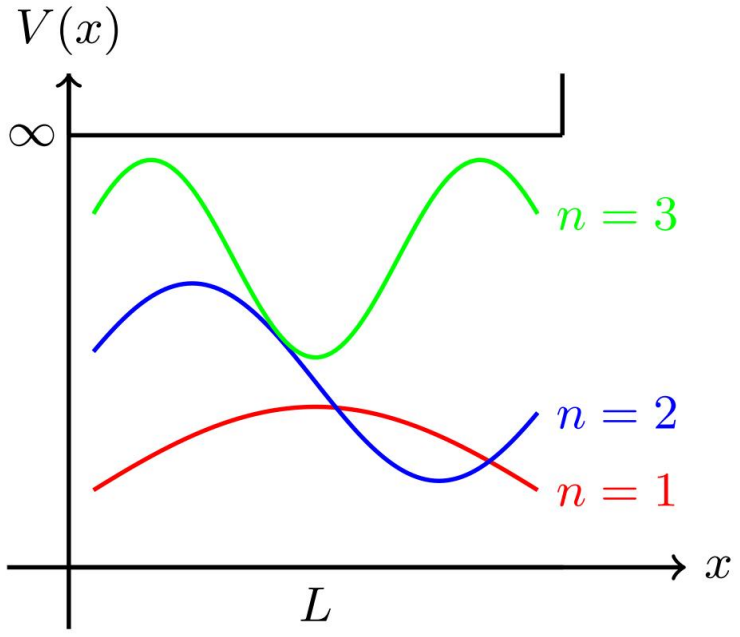
$$\hat{H}=-\frac{\hbar^2}{2m}\nabla^2+V(\mathbf{r},t)$$

势场：

$$V(x)=\begin{cases}0, & 0<x<L\\ \infty, & \text{其他}\end{cases}$$

定态薛定谔方程：

$$-\frac{\hbar^2}{2m}\frac{d^2}{dx^2}\psi_n(x)=E_n\psi_n(x)$$



本征函数：

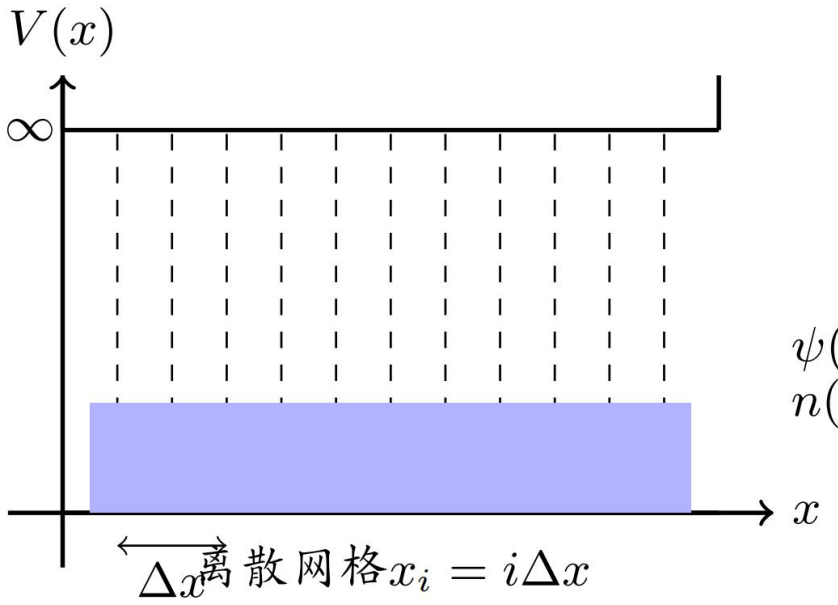
$$\psi_n(x)=\sqrt{\frac{2}{L}}\sin\left(\frac{n\pi x}{L}\right),\quad n=1,2,3,\ldots$$

本征值：

$$E_n=\frac{n^2\pi^2\hbar^2}{2mL^2}$$

从解析求解到数值计算

数值离散化：无限深势井的有限差分法



$$\psi(x_i) \approx \psi_i$$
$$n(x_i) \approx n_i$$

```
import numpy as np
import matplotlib.pyplot as plt

L = 1.0
N = 100
x = np.linspace(0, L, N)
dx = x[1] - x[0]

# Kinetic energy matrix
T = np.diag(-2*np.ones(N))
T += np.diag(np.ones(N-1), 1)
T += np.diag(np.ones(N-1), -1)
T = -0.5 * T / dx**2

# Boundary conditions
T[0, :] = 0
T[-1, :] = 0
T[0, 0] = 1
T[-1, -1] = 1

# Solve eigenvalues
E, psi = np.linalg.eigh(T)
psi = psi / np.sqrt(dx)  # Normalize

# First three states
for i in range(3):
    plt.plot(x, psi[:, i], label=f'n={i+1}')
plt.legend()
plt.show()
```

从无限深势井推广到多粒子

考虑N个相互作用粒子的系统：

$$i\hbar\frac{\partial}{\partial t}\Psi(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N,t)=\hat{H}\Psi(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N,t)$$

多粒子哈密顿量：

$$\hat{H}=\sum_{i=1}^{N}\left[-\frac{\hbar^2}{2m}\nabla_i^2+V_{\text{ext}}(\mathbf{r}_i)\right]+\frac{1}{2}\sum_{i\neq j}V_{\text{ee}}(\mathbf{r}_i-\mathbf{r}_j)$$

关键特点

- 波函数Ψ依赖于所有粒子的坐标
- 电子-电子相互作用V_{ee}使问题复杂化
- 势场不再是简单的外势，而是依赖于系统的波函数

耦合方程的自洽求解

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Kohn-Sham方程与自洽求解

Kohn-Sham方程：

$$\left[-\frac{\hbar^2}{2m}\nabla^2 + V_{\mathrm{KS}}[n](\mathbf{r})\right]\psi_i(\mathbf{r}) = \varepsilon_i\psi_i(\mathbf{r})$$

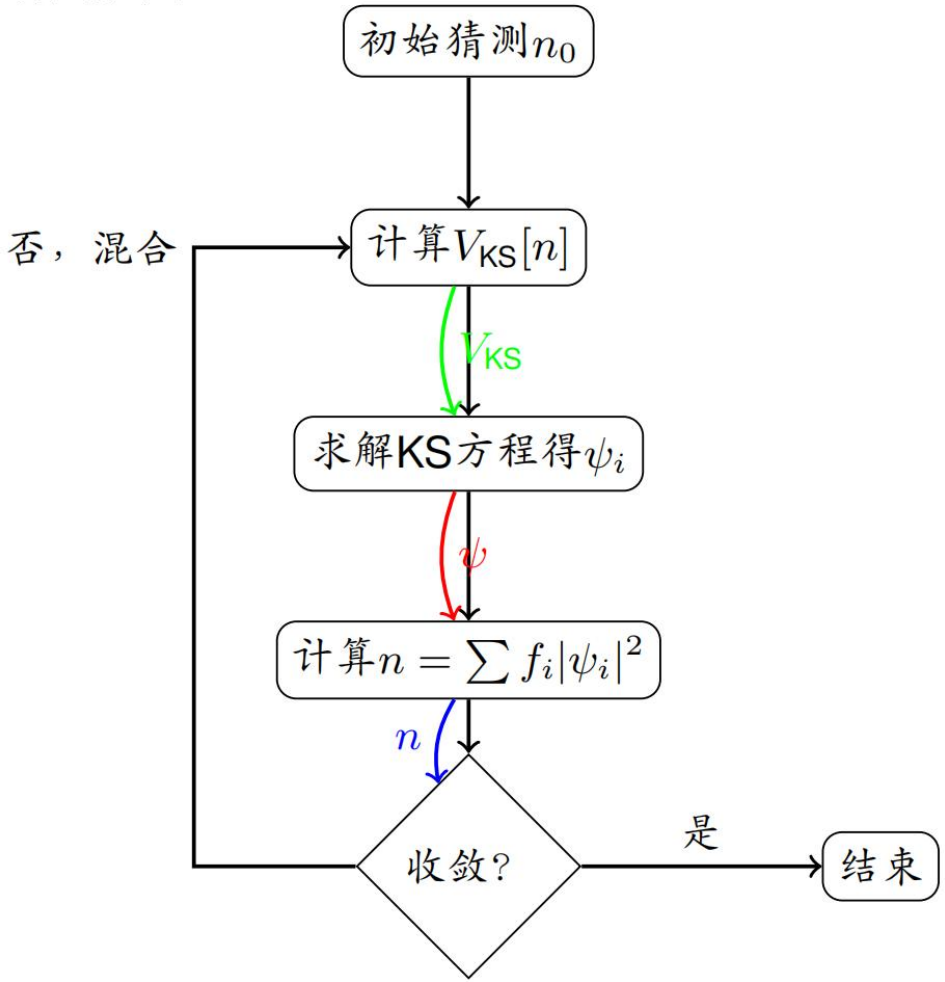
电荷密度：

$$n(\mathbf{r}) = \sum_i f_i|\psi_i(\mathbf{r})|^2$$

Kohn-Sham势：

$$V_{\mathrm{KS}}[n](\mathbf{r}) = V_{\mathrm{ext}}(\mathbf{r}) + V_{\mathrm{H}}[n](\mathbf{r}) + V_{\mathrm{xc}}[n](\mathbf{r})$$

自洽循环：



考虑到多电子体系的复杂性，实际计算量可能增加10^3– 10^6倍！

高性能计算语言与并行方案

编译型语言：

- **Fortran**
  - 历史悠久，数值计算传统
  - 高效的数组操作
  - 常见DFT软件：VASP, Quantum ESPRESSO, Abinit
- **C/C++**
  - 现代软件工程
  - 灵活的内存管理
  - 常见DFT软件：OpenMX, ABACUS

并行编程模型：

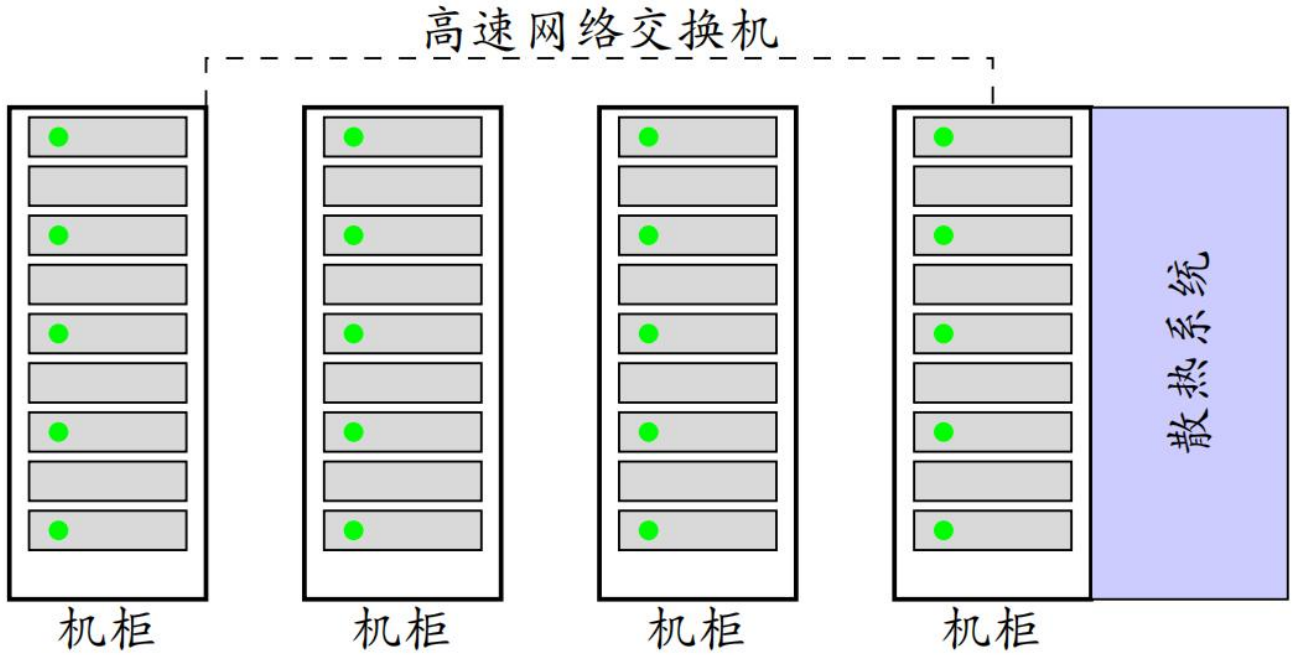
- **MPI** (Message Passing Interface)
  - 多节点分布式内存并行
  - 跨机器通信
- **OpenMP**
  - 单节点共享内存并行
  - 多线程
- **CUDA/OpenCL**
  - GPU加速
  - 大规模并行计算

新算法开发需求：

- 选择一个大规模、模块化、可扩展的基本框架
- 理解新算法所涉及的模块
- 设计、实现、调试验证新算法
- 应用于实际计算体系上，探索新物理

高性能计算平台与远程连接

高性能计算机房：



- Linux操作系统
- 专业散热设施
- 24/7稳定运行

SSH客户端命令行界面：

```
+-----------------------------------------+
| user@local:~$ ssh user@hpc.example.com  |
| Password: ********                      |
| Last login: Wed May 15 09:30:00 2024    |
|                                         |
|      Welcome to Supercomputer Cluster!  |
|                                         |
| [user@hpc-login ~]$ pwd                 |
| /home/user                              |
| [user@hpc-login ~]$ ls                  |
| project1  project2  scripts             |
| [user@hpc-login ~]$ cd project1         |
| [user@hpc-login project1]$ ls           |
| input.in  OUTCAR  POSCAR                |
| [user@hpc-login project1]$ sbatch job.sh |
| Submitted batch job 12345               |
| [user@hpc-login project1]$ squeue       |
|  JOBID PARTITION NAME USER ST TIME NODES |
|  12345 compute   job  user R  0:05 4    |
+-----------------------------------------+
```

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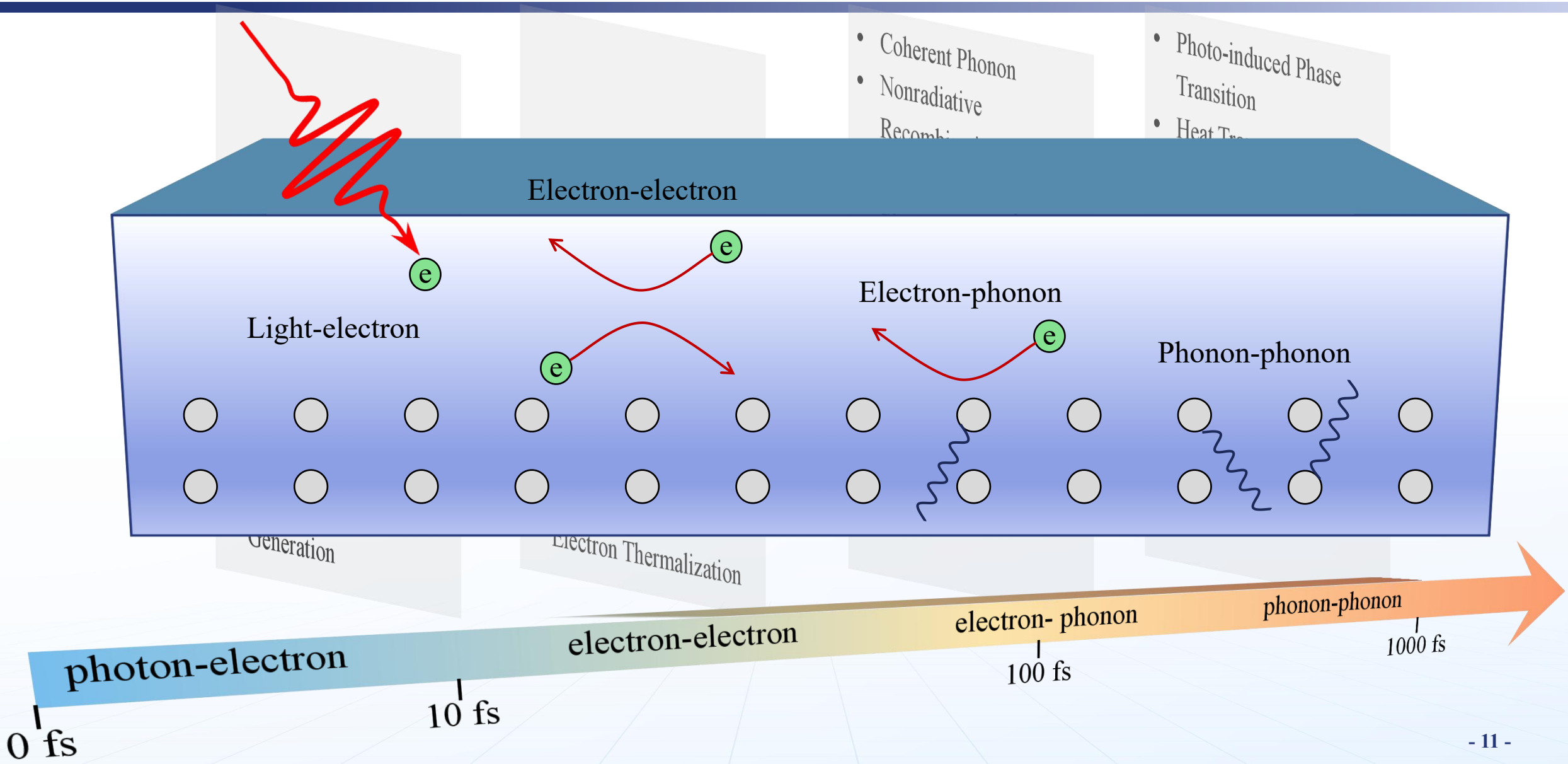
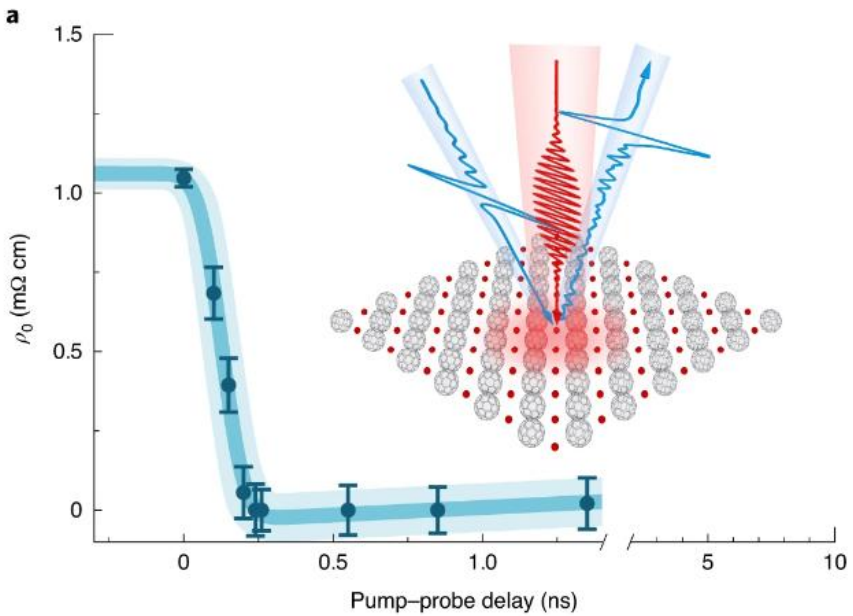
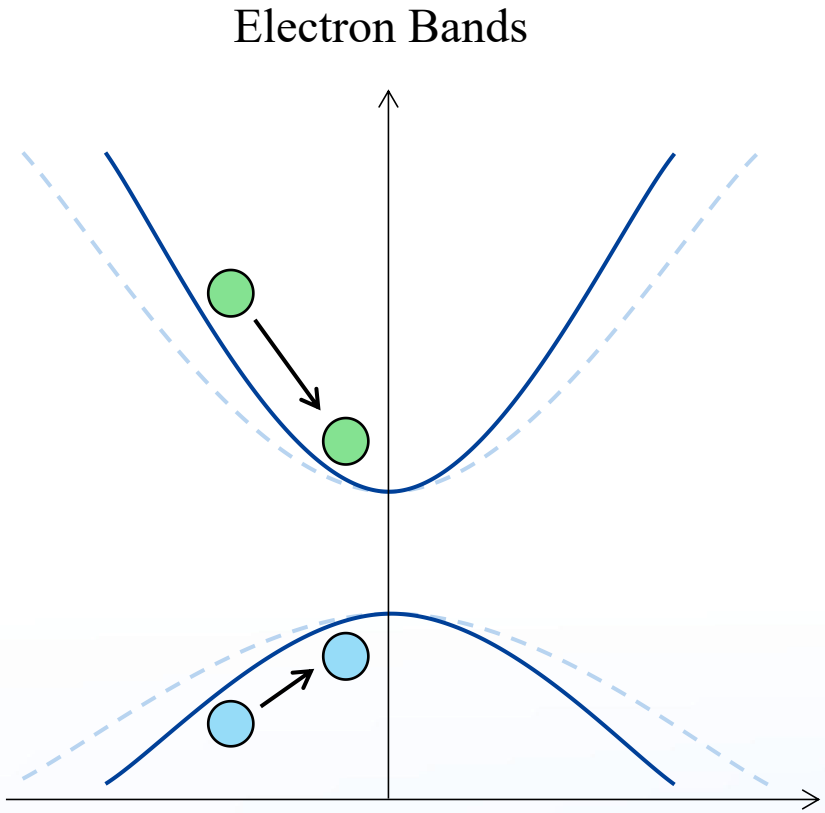
Fundemental Interactions

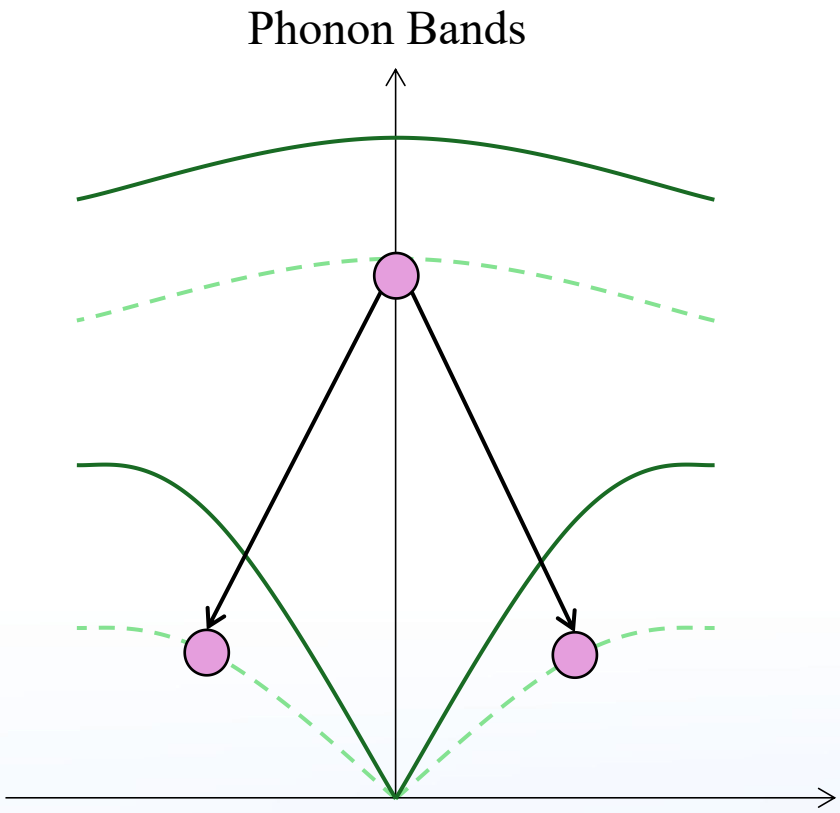


Why TDDFT, not something else



M. Budden et al., Nat. Phys. 17, 611 (2021).





Pump-probe Experiments Must Have:

- **Strong Field** **(0.1 - 10 V/nm)**
- **Dynamics** **(as - ps)**
- **Multi-Properties** **(Electron, Spin, Phonon)**

Simulations Must Have:

- **First Principles Base** **(Atomistic Picture)** ~~**Emperical models**~~
- **Time-dependence** **(Non-equilibrium)** ~~**Frequency-domain**~~
- **Molecular Dynamics** **(Structural change)** ~~**Perturbation Methods**~~
- **Band structure evolution** **(Density/Wavefunction-Based)** ~~**Green-Function Methods**~~

Unified Framework for Photon-electron-phonon

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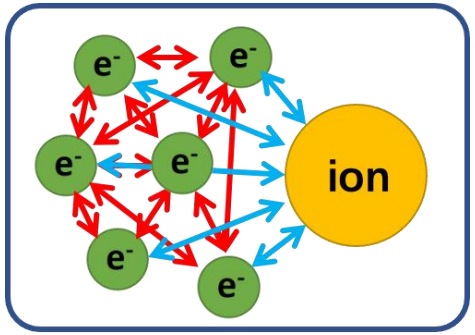
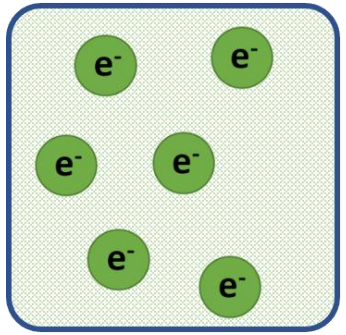
Electron-light Interaction	Real-time Propagation	Electron-phonon Coupling
$H = H_{\mathrm{el}} + H_{\mathrm{el-ph}} + H_{\mathrm{el-light}}$	$H\psi = E\psi$ ↓ $H\psi(t) = -i\dot{\psi}(t)$	$\mathbf{F}_I = \sum_j \langle \psi_j \vert \nabla_{\mathbf{R}_I} H \vert \psi_j \rangle$ $\mathbf{F}_I = m_I \ddot{\mathbf{R}}_I$
Electric fields in solid will destroy translational symmetry	The integration time step of the time-dependent equation < 1 as	Ion dynamics under the ground-state potential energy surface
Introduce a velocity-gauge field	**Step size > 200 as with adiabatic eigenstates**	**coherent oscillation and phase transitions in excited-state potential energy surface**

Time Dependent Density Functional Theory



$$\rho(\boldsymbol{r},t) = \rho^{\mathrm{KS}}(\boldsymbol{r},t)$$

$$\rho^{KS}(\boldsymbol{r},t) = \sum_i \psi_i^*(\boldsymbol{r},t)\psi_i(\boldsymbol{r},t)$$



Interacting System

Non-interacting (Kohn-Sham)

Runge-Gross Theorem: one-to-one correspondence between the time-dependent electron density ρ(r, t) and the time-dependent external potential.

Van Leeuwen Theorem: There exists a non-interacting system whose ρ_KS(r, t) is identical to the ρ(r, t) of the interacting system.

$$\mathrm{i}\hbar\frac{\partial\psi_i(t)}{\partial t} = \widehat{H}_{\mathrm{KS}}(t)\psi_i(t)$$

$$\psi_i(t_2) = \widehat{U}\,(t_2,t_1)\psi_i(t_1)$$

(Courtesy of Daqiang Chen)

E. Runge and E. K. U. Gross, Rev. Lett. **52**, 997 (1984).
R. van Leeuwen, Phys. Rev. Lett. **82**, 3863 (1999).

Velocity-Gauge Field

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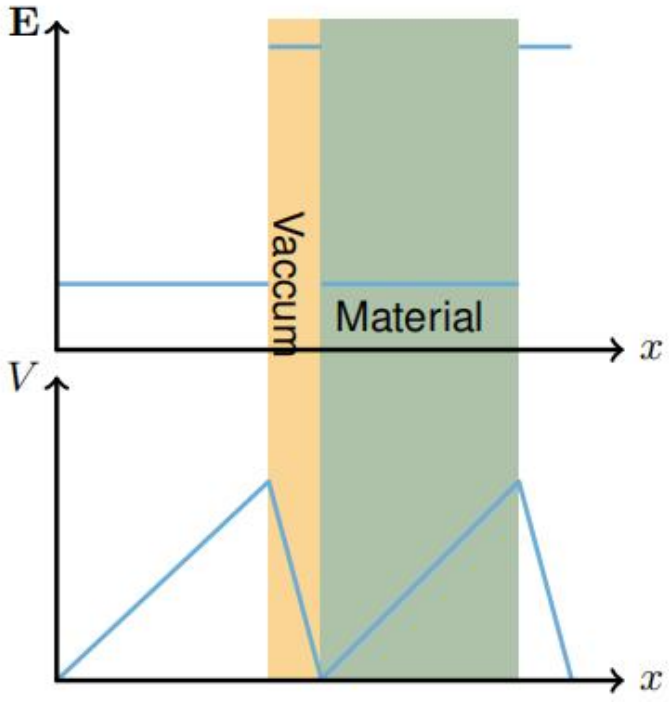
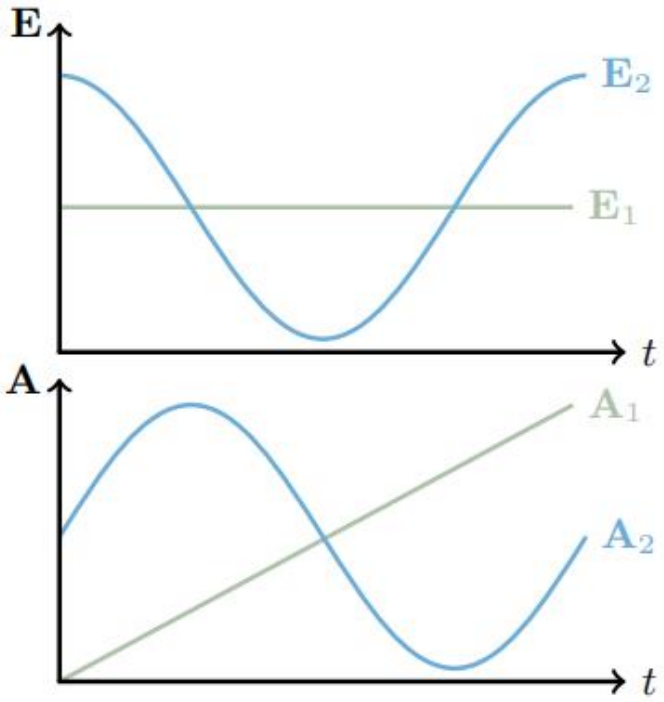
Length gauge

1. For $x \in (0, L-\delta)$, $V_{\mathrm{H}}(x) = V_{\mathrm{H}}(x) + eE_0 x$
2. To ensure periodicity, require narrow $\delta \to 0$ vacuum layer $[L-\delta, L]$ with unphysical field $E(x) = E_0/\delta \to \infty$.



Velocity gauge

1. $\mathbf{A}_{\mathrm{ext}}(t) = -c\int_0^t \mathbf{E}_{\mathrm{ext}}(t')dt'$
2. The periodicity is sustained with uniform $\mathbf{A}(t)$ in a unit cell.
3. Only physical in a time-dependent case.



YABANA Kazuhiro et al, Phys. Rev. B **54**, 4484 (1996). Phys. Rev. B **62**, 7998 (2000).

Efficient Evolution

TDKS equation $n \to \gamma\mathbf{k}$

$$i\hbar\frac{\partial}{\partial t}\psi_{\gamma\mathbf{k}}(\mathbf{r},t) = H(\mathbf{r},t)\psi_{\gamma\mathbf{k}}(\mathbf{r},t)$$

where Hamiltonian is

$$H(\mathbf{r},t) = \frac{1}{2m}\left[-i\hbar\nabla + e\mathbf{A}(t)\right]^2 + V(\mathbf{r})$$

Here, i, j, h is adiabatic, H, γ is TDKS. Within $t - t_1 < \Delta t$, expand with adiabatic basis

$$\psi_{\gamma\mathbf{k}}(\mathbf{r},t) = \sum_{i\mathbf{k}'} c_{i\mathbf{k}'\gamma\mathbf{k}}(t)\phi_{i\mathbf{k}'}(\mathbf{r},t_1)\exp\left[-i\frac{\epsilon_{i\mathbf{k}'}(t_1)}{\hbar}(t-t_1)\right]$$

The adiabatic basis is **stationary** eigenvector of Hamiltonian

$$H(\mathbf{r},t_1)\phi_{i\mathbf{k}}(\mathbf{r},t_1) = \epsilon_{i\mathbf{k}}(t_1)\phi_{i\mathbf{k}}(\mathbf{r},t_1),$$

Initial condition

$$\psi_{i\mathbf{k}}(\mathbf{r},t=0) := \phi_{i\mathbf{k}}(\mathbf{r},t=0),$$
$$c_{i\mathbf{k}'\gamma\mathbf{k}}(t=0) := \delta_{i\gamma}\delta_{\mathbf{k}\mathbf{k}'},$$

Z. Wang, S.-S. Li, and L.-W. Wang, Phys. Rev. Lett. **114**, 063004 (2015)
C. Lian, S.-J. Zhang, S.-Q. Hu, M.-X. Guan, and S. Meng, Nat Commun 11, 43 (2020).

Efficient Evolution in Adiabatic Basis

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Approximation: $h_{ij,\mathbf{k}}(t)$ changes linearly between $[t_1, t_2]$ $(t_2 = t_1 + \Delta t)$:

$$h_{ij,\mathbf{k}}(t) = h_{ij,\mathbf{k}}(t_1) + \frac{t - t_1}{t_2 - t_1}\left[h_{ij,\mathbf{k}}(t_2) - h_{ij,\mathbf{k}}(t_1)\right],$$

$\phi_{l,\mathbf{k}}(\mathbf{r}, t_2)$ and $\epsilon_{l\mathbf{k}}(t_2)$ are the adiabatic basis and the eigenvalue at time t_2, which are solved in a self-consistent process.

Propagator operator $U_{\mathbf{k}}(t_2, t_1)$ is calculated with the knowledge of $H_{\mathbf{k}}(t)$ in the Crank-Nicholson scheme

$$\begin{aligned} U_{\mathbf{k}}(t_2, t_1) &= \prod_{s=0}^{N_t} \frac{\exp[-i/\hbar H_{\mathbf{k}}(t_2 - sdt)dt/2]}{\exp[i/\hbar h_{\mathbf{k}}(t_2 - sdt)dt/2]} \\ &= \prod_{s=0}^{N_t} \frac{1 - i/\hbar h_{\mathbf{k}}(t_2 - sdt)dt/2}{1 + i/\hbar H_{\mathbf{k}}(t_2 - sdt)dt/2}, \end{aligned}$$

where $dt = \Delta t/N_t \sim 0.1$ as is the integration time step. The coefficients $c_{i\gamma,\mathbf{k}}$ are thus propagated as

$$C_{\mathbf{k}}(t_2) = U_{\mathbf{k}}(t_2, t_1)C_{\mathbf{k}}(t_1).$$

Z. Wang, S.-S. Li, and L.-W. Wang, Phys. Rev. Lett. **114**, 063004 (2015)
C. Lian, S.-J. Zhang, S.-Q. Hu, M.-X. Guan, and S. Meng, Nat Commun 11, 43 (2020).

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第一性原理算法开发

含时密度泛函理论

凝聚态超快动力学模拟

TDAP Accomplishments

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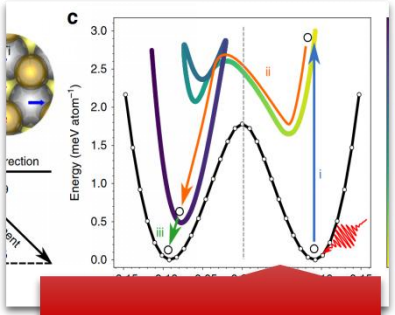
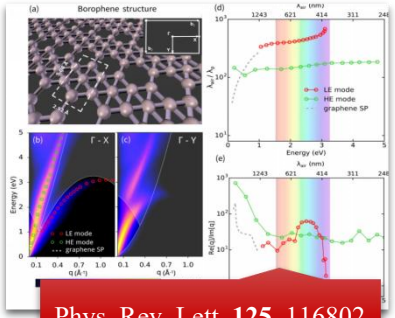
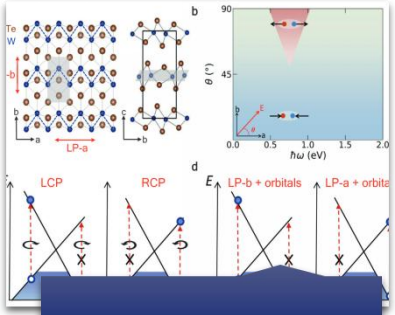
- Full *ab initio* method for coupled **light-electron-spin-phonon** dynamics
- x200 acceleration with efficient evolution
- Calculation-only papers: **Nat. Mater**. x1 , **PRL** x 12, **Nat. Commun**. x5, **Sci. Adv**. x4



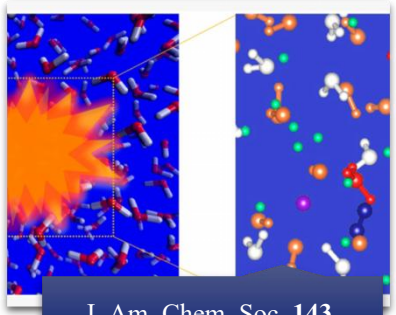
Nat Commun **11**, 43 (2020).



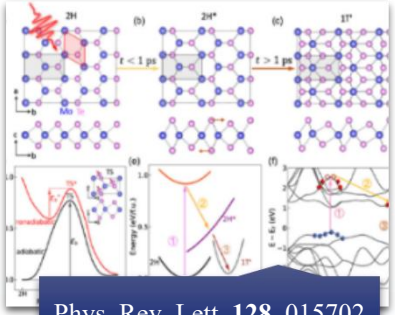
Phys. Rev. Lett. **125**, 116802 (2020).



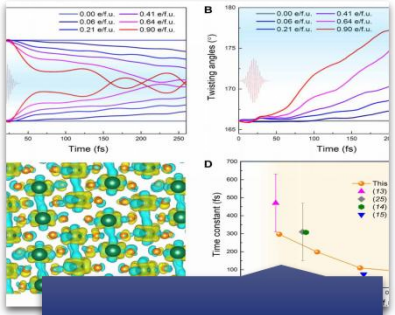
Nat Commun **12**, 1885 (2021).



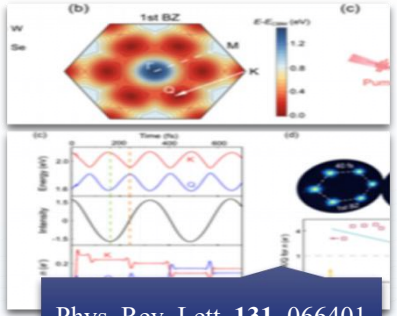
J. Am. Chem. Soc. **143**, 10382 (2021).



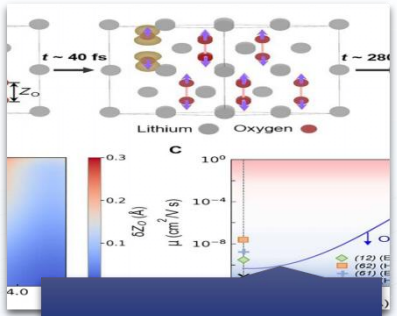
Phys. Rev. Lett. **128**, 015702 (2022).



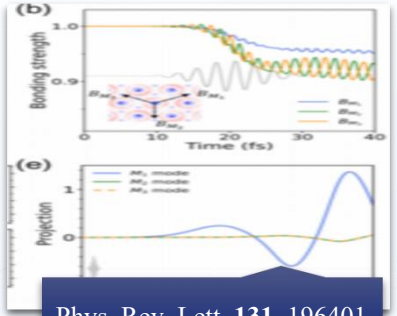
Sci. Adv. **8**, eadd2392 (2022).



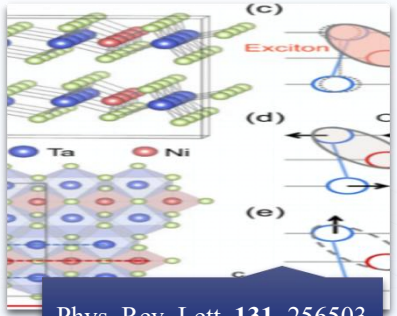
Phys. Rev. Lett. **131**, 066401 (2023).



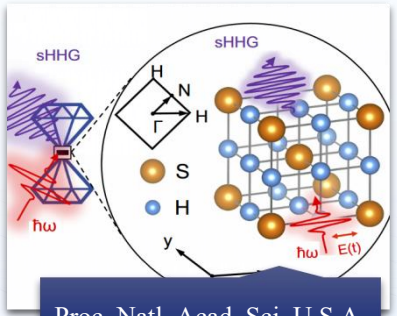
Sci. Adv. **9**, eadg3833 (2023).



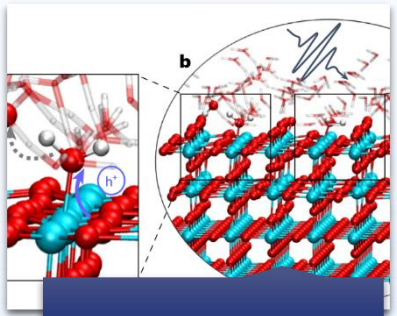
Phys. Rev. Lett. **131**, 196401 (2023).



Phys. Rev. Lett. **131**, 256503 (2023).

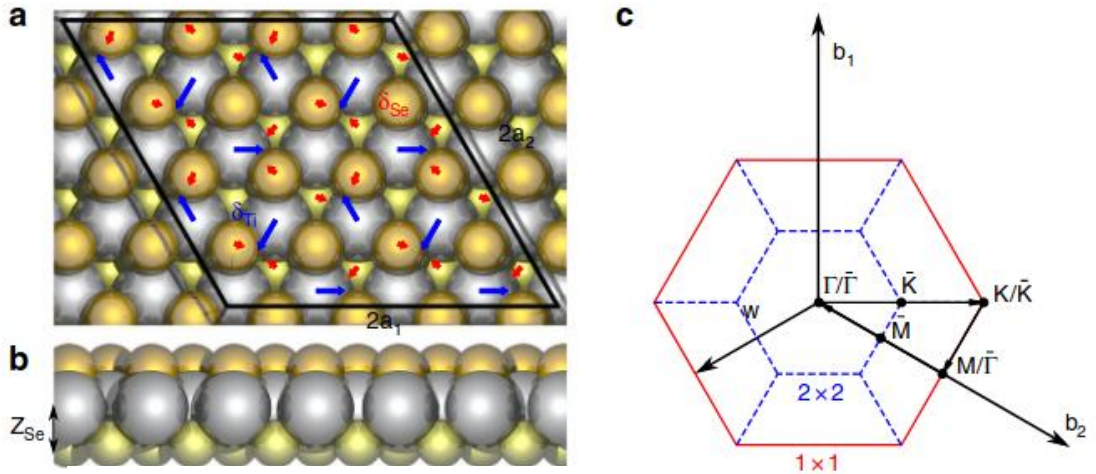


Proc. Natl. Acad. Sci. U.S.A. **121**, e2316775121 (2024).

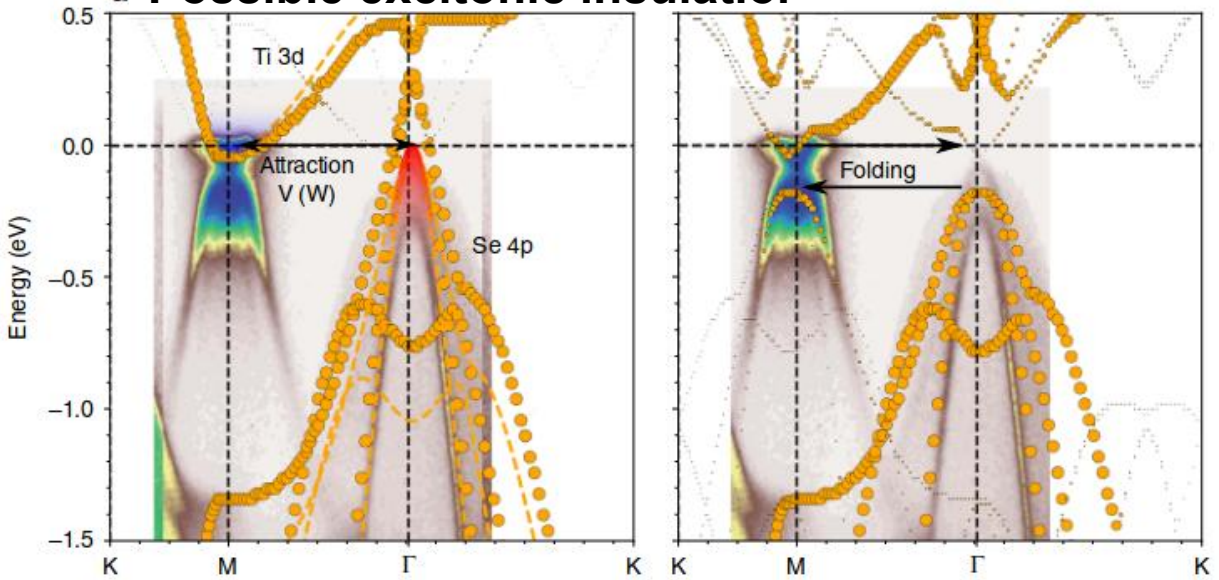


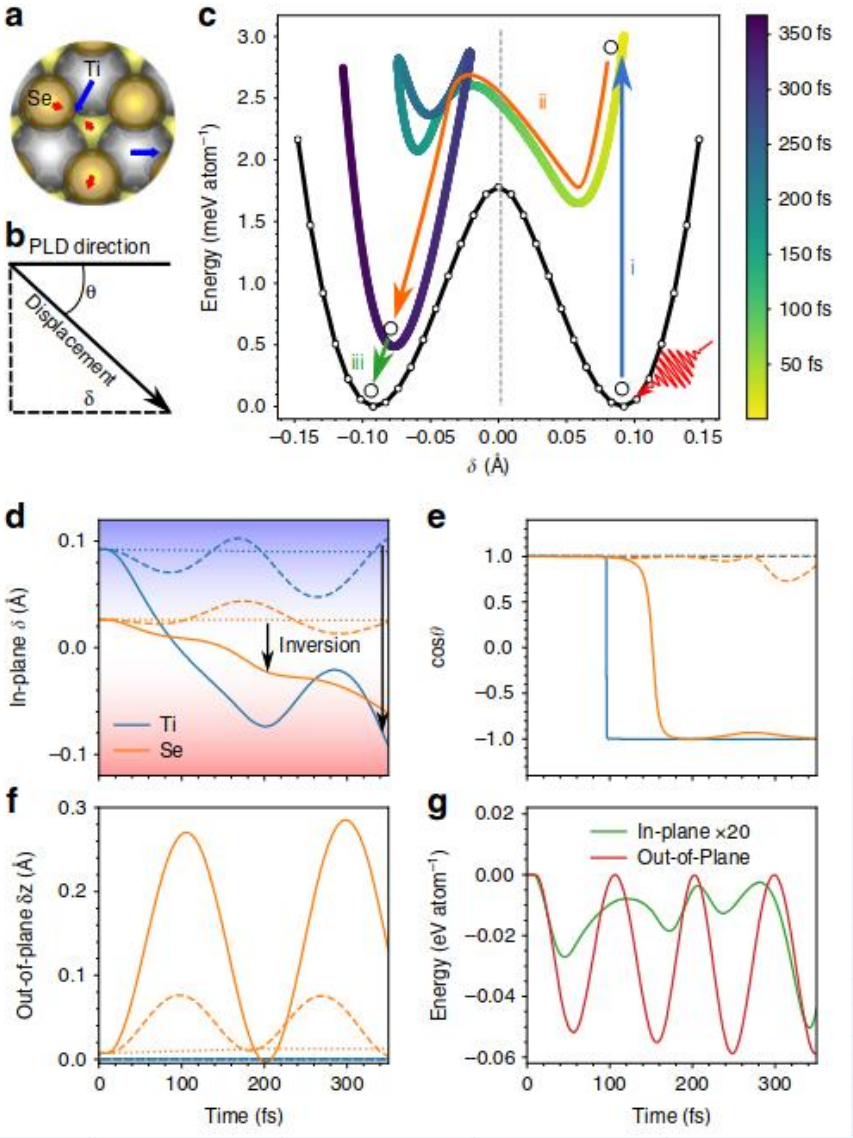
Nat. Mater. **23**, 1100(2024).

Ultrafast CDW dynamics in $TiSe_2$



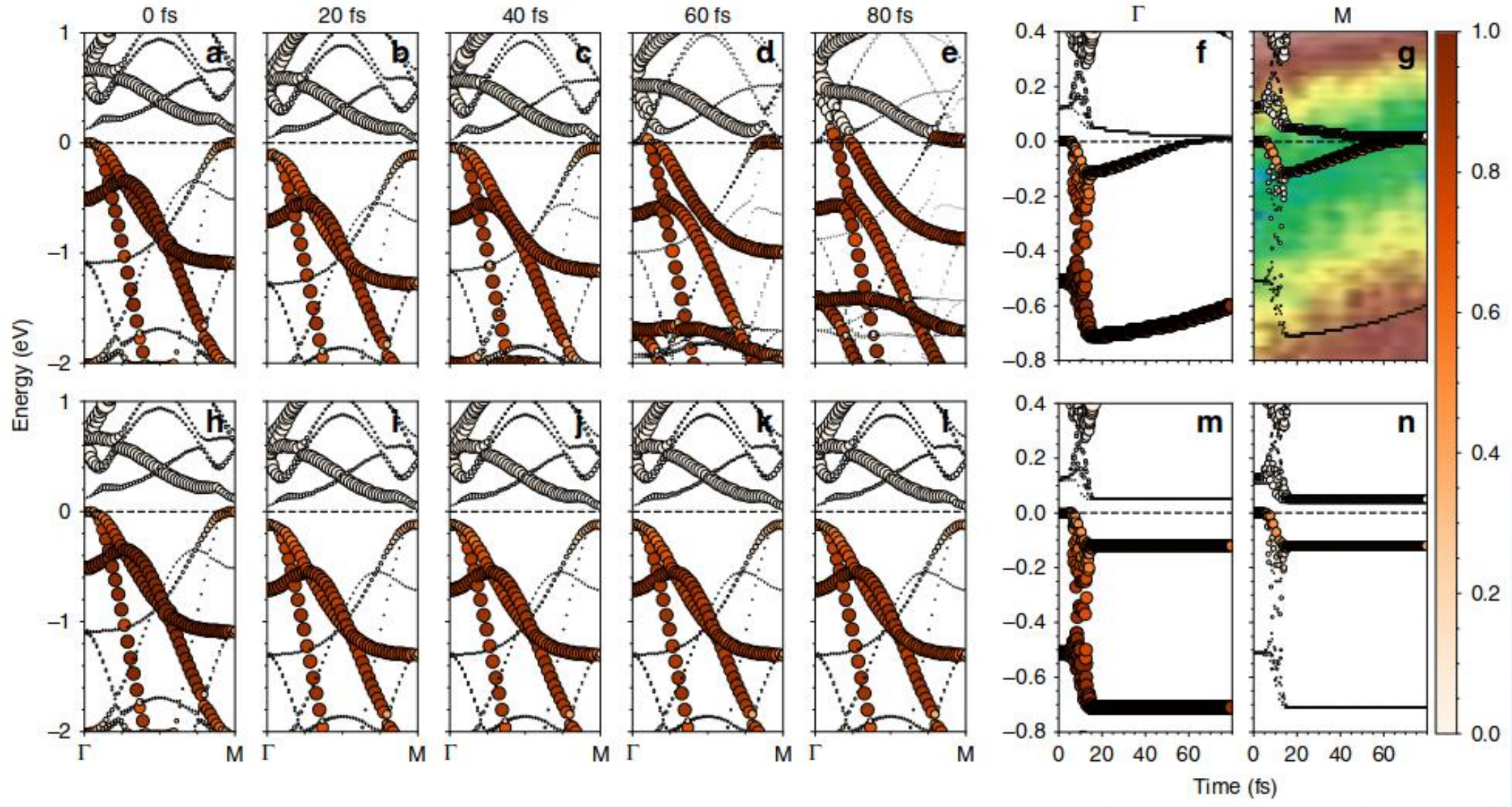
d Possible excitonic insulatior





C. Lian, S.-J. Zhang, S.-Q. Hu, M.-X. Guan, and S. Meng, Nat Commun **11**, 43 (2020).

TR-ARPES Simulations



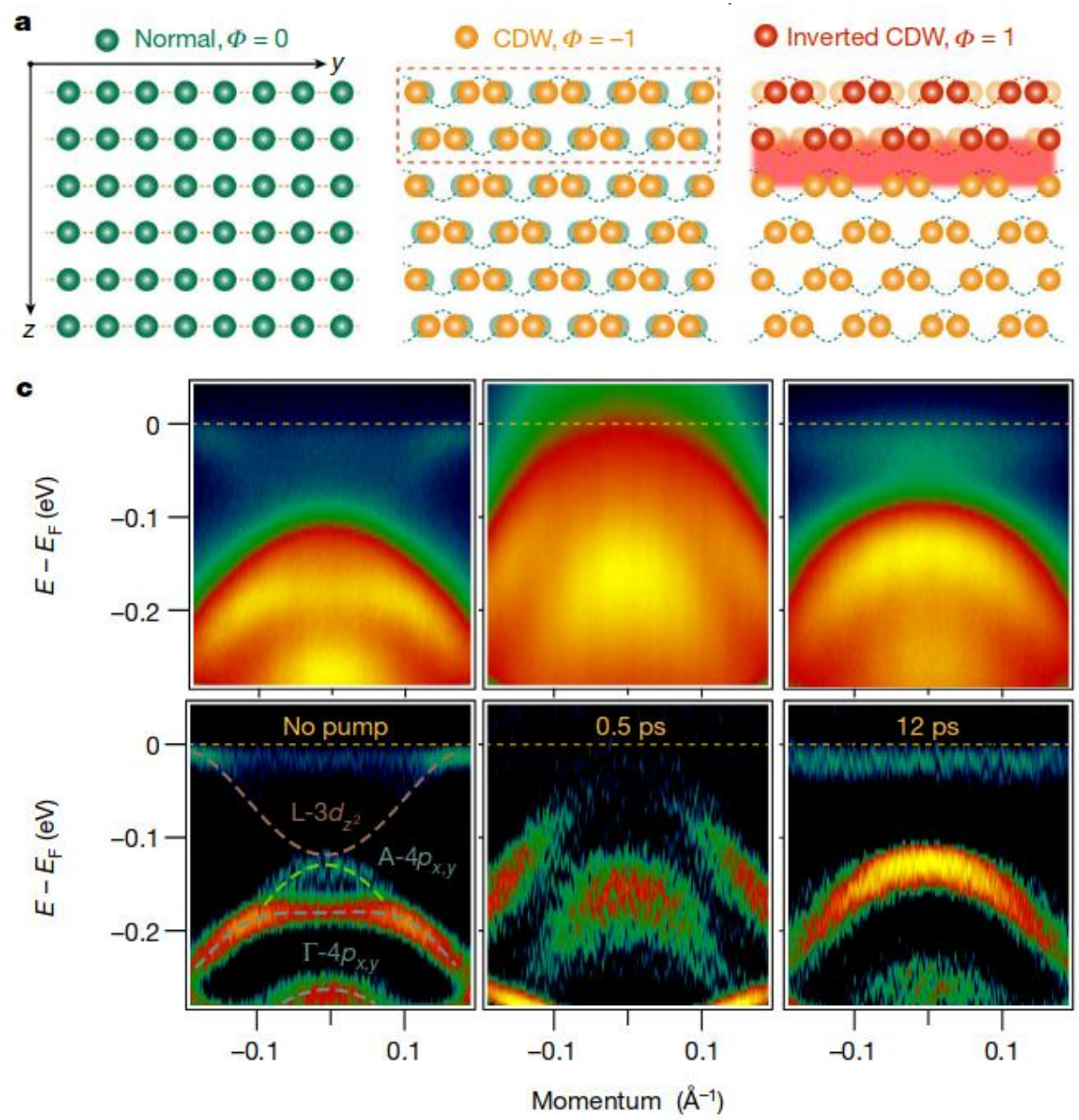
C. Lian, S.-J. Zhang, S.-Q. Hu, M.-X. Guan, and S. Meng, Nat Commun **11**, 43 (2020).

Experimental Validation

Nature 595, 239 (2021) observed CDW Inversion (Dao Xiang and Wentao Zhang et al)

*Ultrafast photon excitation in a charge density wave (CDW) material can induce a complete inversion of the phase order via an electron–phonon scattering process, and the symmetry of the inverted phase (Φ = 1) is equivalent to the original phase (Φ = −1)**[19]**.*

In summary, we detected photoinduced macroscopic CDW domain walls in 1T-TiSe2. The found domain walls existed parallel to the surface in the interval between the photo-inverted phase and the original phase and could be placed near the surface by finely tuning the excitation energy.



Experimental Validation

Urs Staub et al [Phys. Rev. Research 3, 013128 (2021)]
supported the structurally assisted melting mechanism

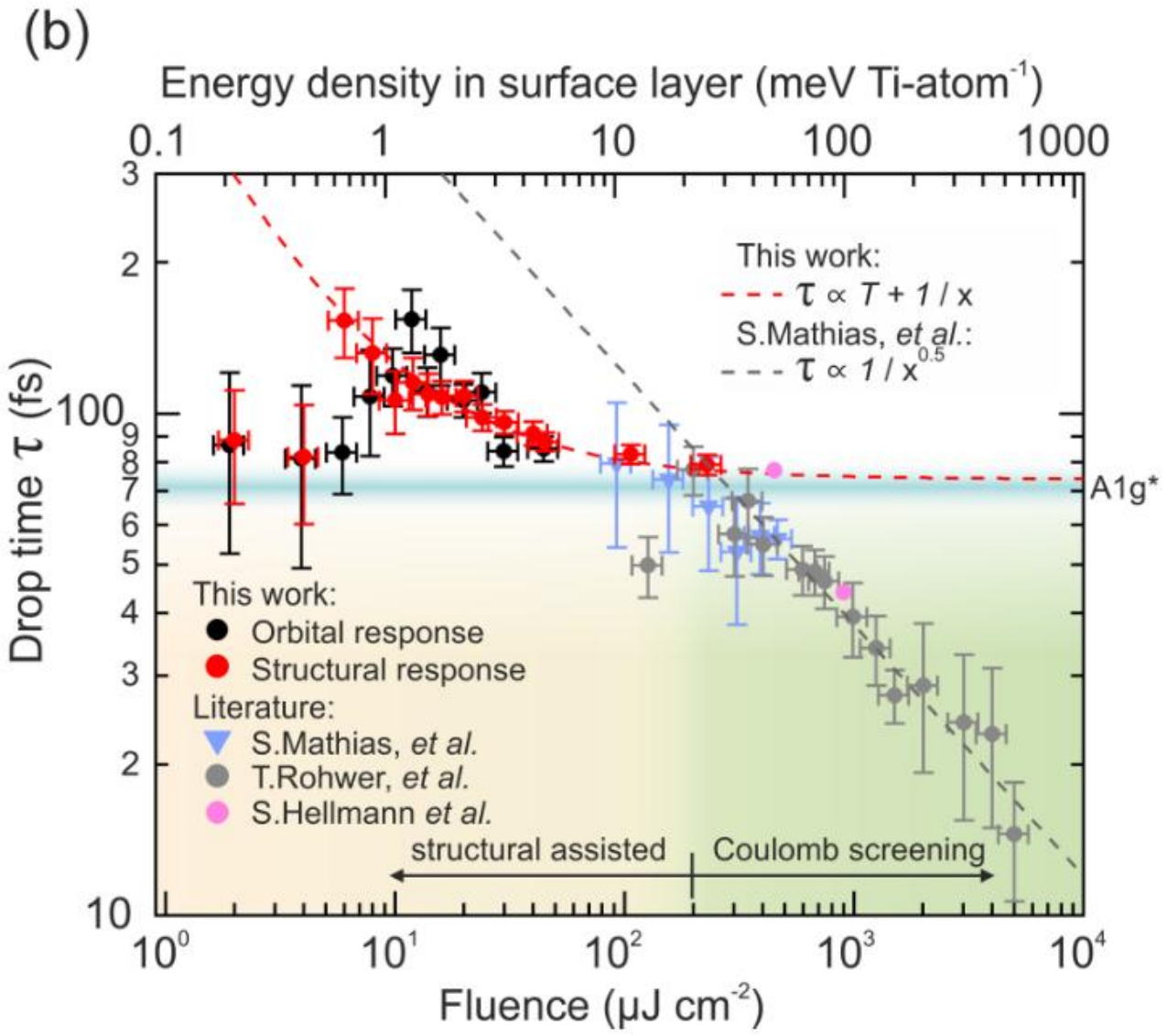
*These results point toward a structurally assisted melting of carrier correlations. Hot-carrier and hole injection distorts the local energetic landscape **[9]**, causing a structural rearrangement along the A1g trajectory towards the RT phase, resulting in coherent atomic oscillations around the new, shifted potential minimum.*

Reviewed by Rev. Mod. Phys. 93, 041002 (2021):

*TDDFT has been successfully applied to a host of systems in weak and strong external driving fields, ranging from atoms and molecules to periodic solids (De Giovannini et al., 2013; **Lian et al., 2020**).*

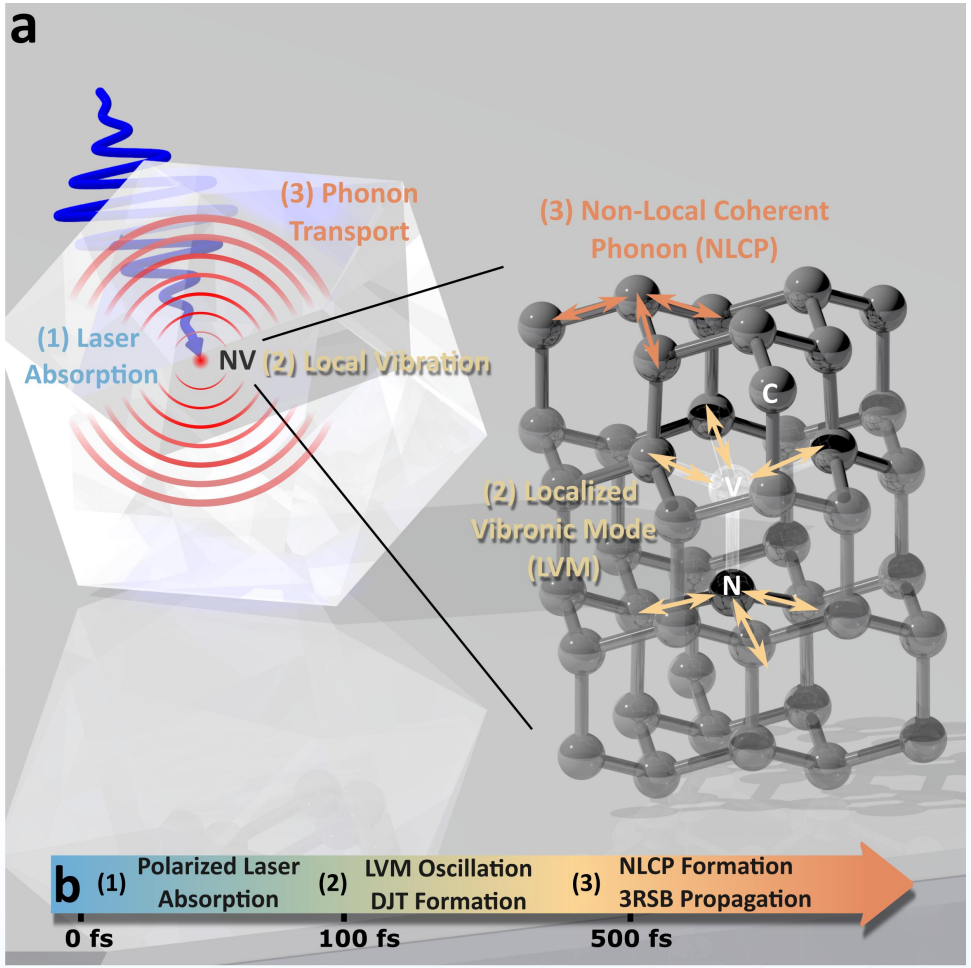
Similar mechanism:

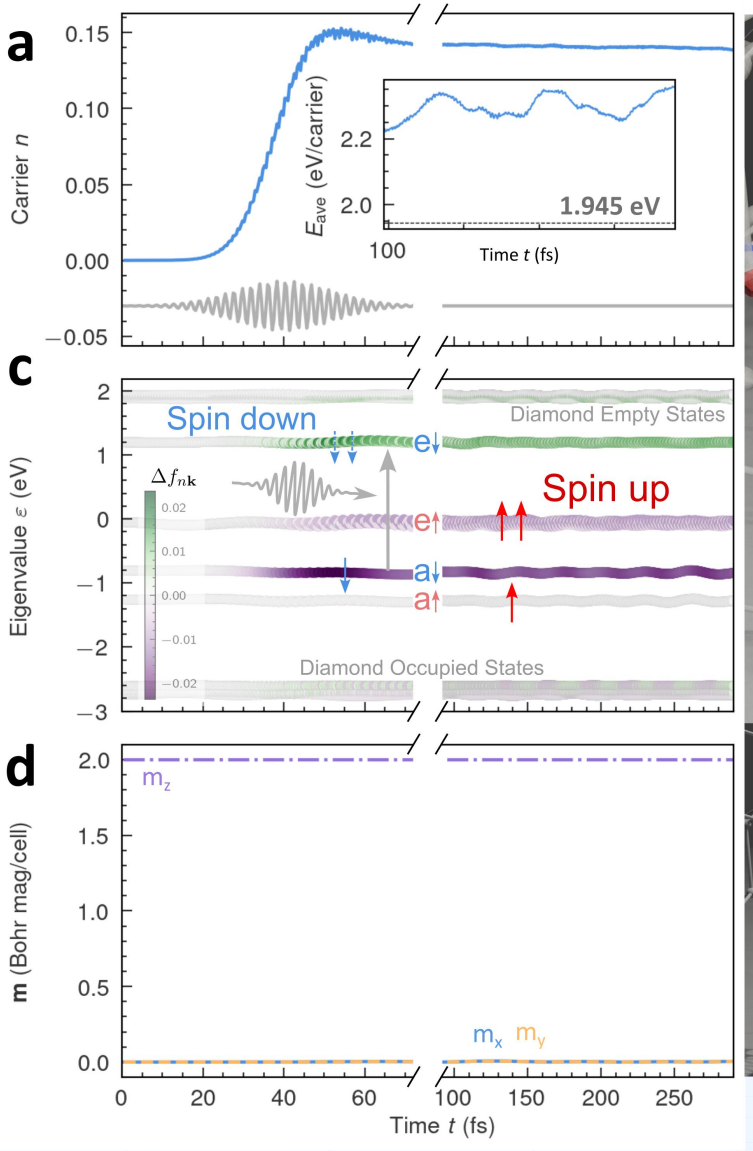
C. Lian, Z. A. Ali, H. Kwon, and B. M. Wong, J. Phys. Chem. Lett. 10, 3402 (2019).
H. Ning*, O. Mehio*, **C. Lian***, et al, and D. Hsieh, Phys. Rev. B 106, 205118 (2022).

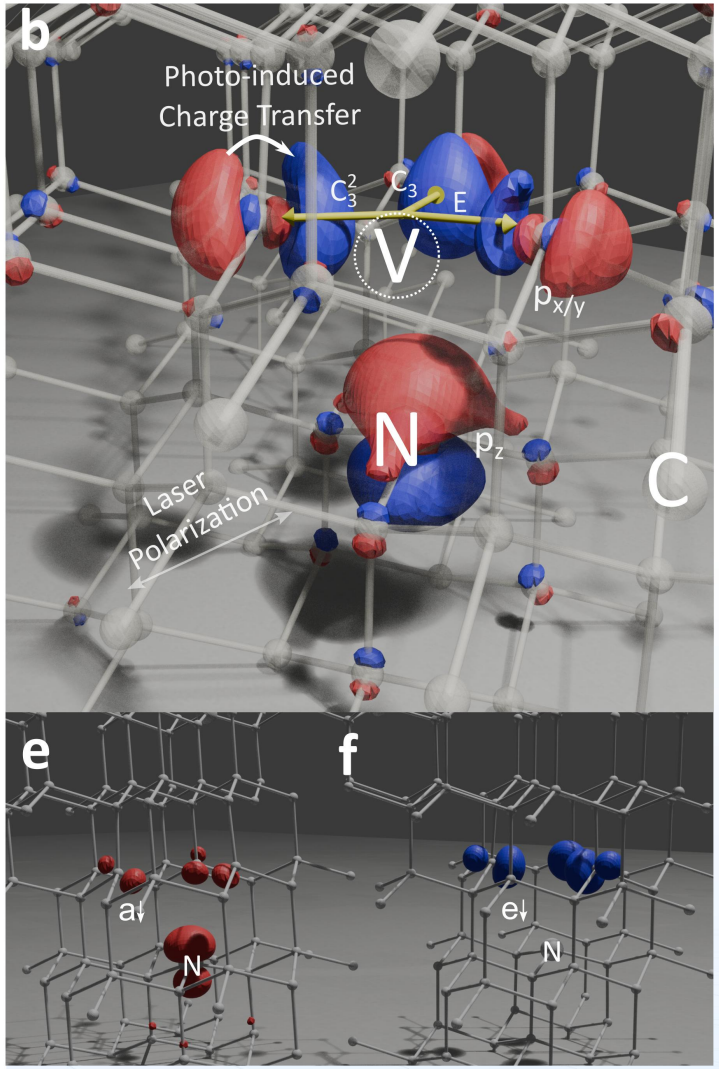


Macroscopic Symmetry-Breaking in NV Centers

超快物質科學中心
Center for Ultrafast Physics



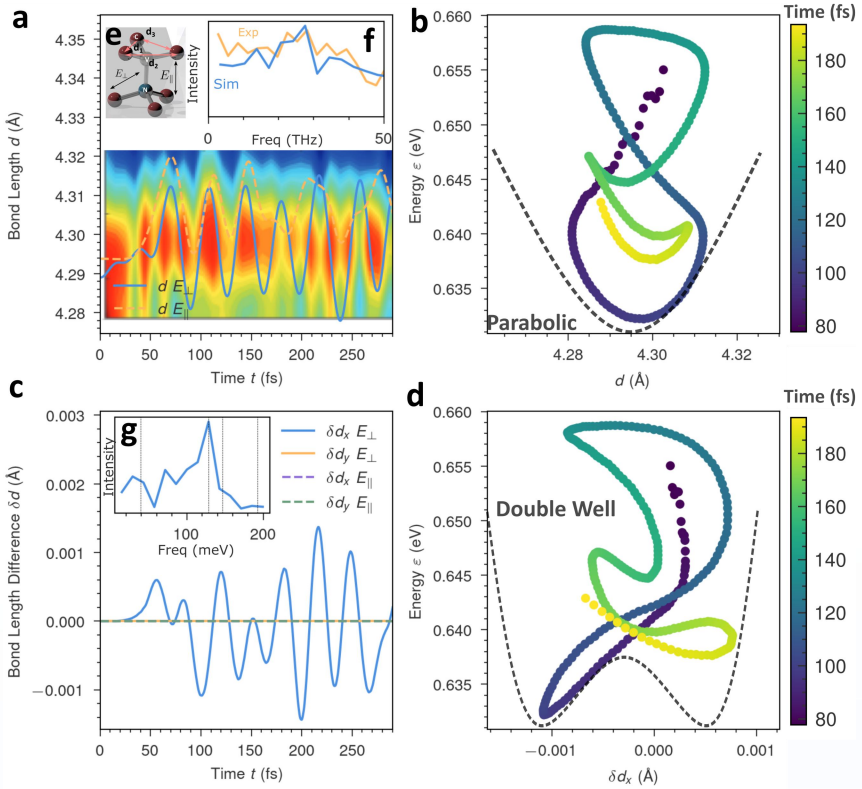


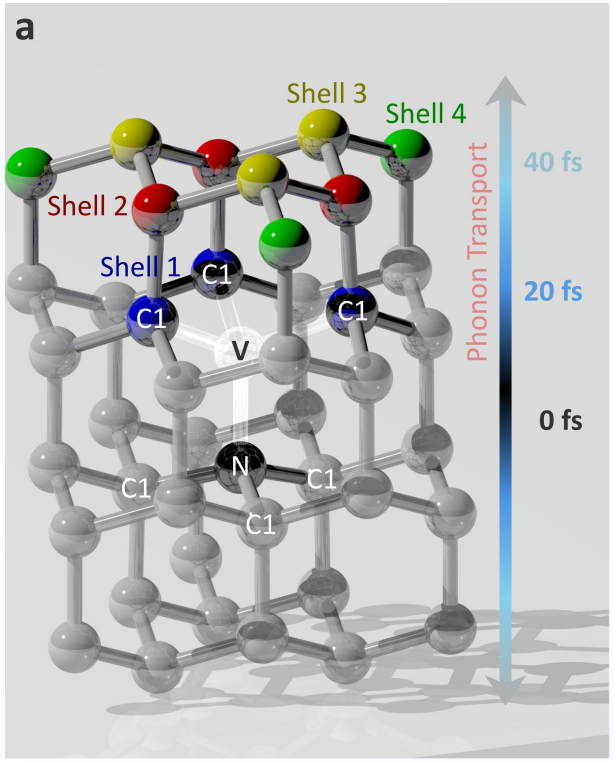


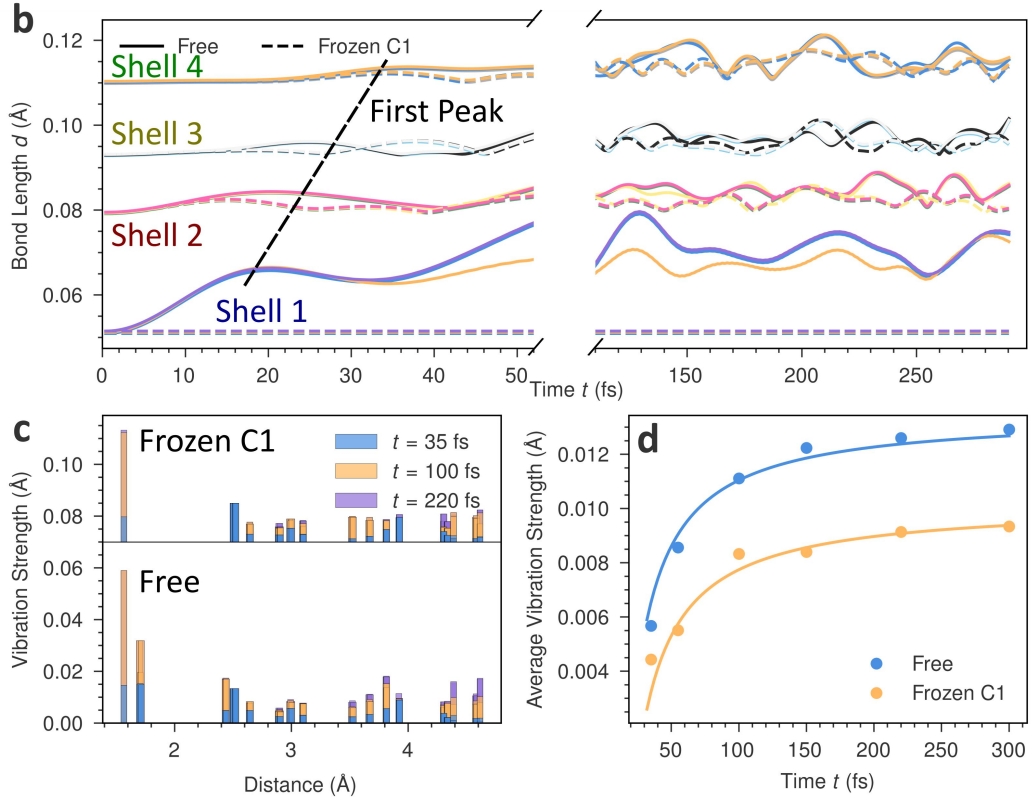
Yang Gao. ..., and Chao Lian*. Ultrafast laser induces macroscopic symmetry-breaking of diamond color centers. *Nat Commun* (2026).

Macroscopic Symmetry-Breaking in NV Centers

超快物質科學中心
Center for Ultrafast Physics







Yang Gao. ..., and Chao Lian*. Ultrafast laser induces macroscopic symmetry-breaking of diamond color centers. *Nat Commun* (2026).

总结与致谢

超快物質科學中心
Center for Ultrafast Physics

- 含时密度泛函算法开发 = 高效程序 + 多体相互作用 + 含时薛定谔方程
- 基于自主研发的第一性原理成功模拟实际体系的超快动力学

中国科学院
CHINESE ACADEMY OF SCIENCES

稳定支持青年团队

Stable Support for Youth Group in Basic Research Field, CAS

中华人民共和国科学技术部
Ministry of Science and Technology of the People's Republic of China

重点研发计划

National Key R&D Program of China

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Tellrium: Dr. H. Ning, Prof. D. Heish

Graphene: Dr. J. Li, Prof. X. Zhu, Prof. J. Guo

NV center: Prof. A. Gali

欢迎加入

中国科学院物理研究所超快物质科学中心

聚焦超快科学与物质科学交叉发展的最前沿，以超快科学与物质科学的结合为牵引，开展新一代先进超快激光技术、超快物性测量和超快科学研究，旨在建设以超快时间分辨的物质科学研究为突出特点的超快科学与技术研究高地。

> 中心承担“先进阿秒激光设施”建设任务，其科学目标是建设当前最先进的，波段、性能以及应用终端覆盖最全的，以阿秒时间分辨能力和高度时空相干性为主要特点的综合性超快电子动力学研究设施。

中心长期面向海内外招聘超快科学领域的学科带头人及科研骨干人才，提供优厚的科研条件及待遇，诚邀有志之士加盟！

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- 阿秒液相动力学
- 阿秒关联物态
- 阿秒磁学
- 阿秒理论

招聘岗位

杰出研究员、特聘研究员I类、特聘研究员II类、关键技术人才

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