Quantum Control of Hubbard excitons

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Harvard University

SPICE Workshop
Quantum materials and quantum information science
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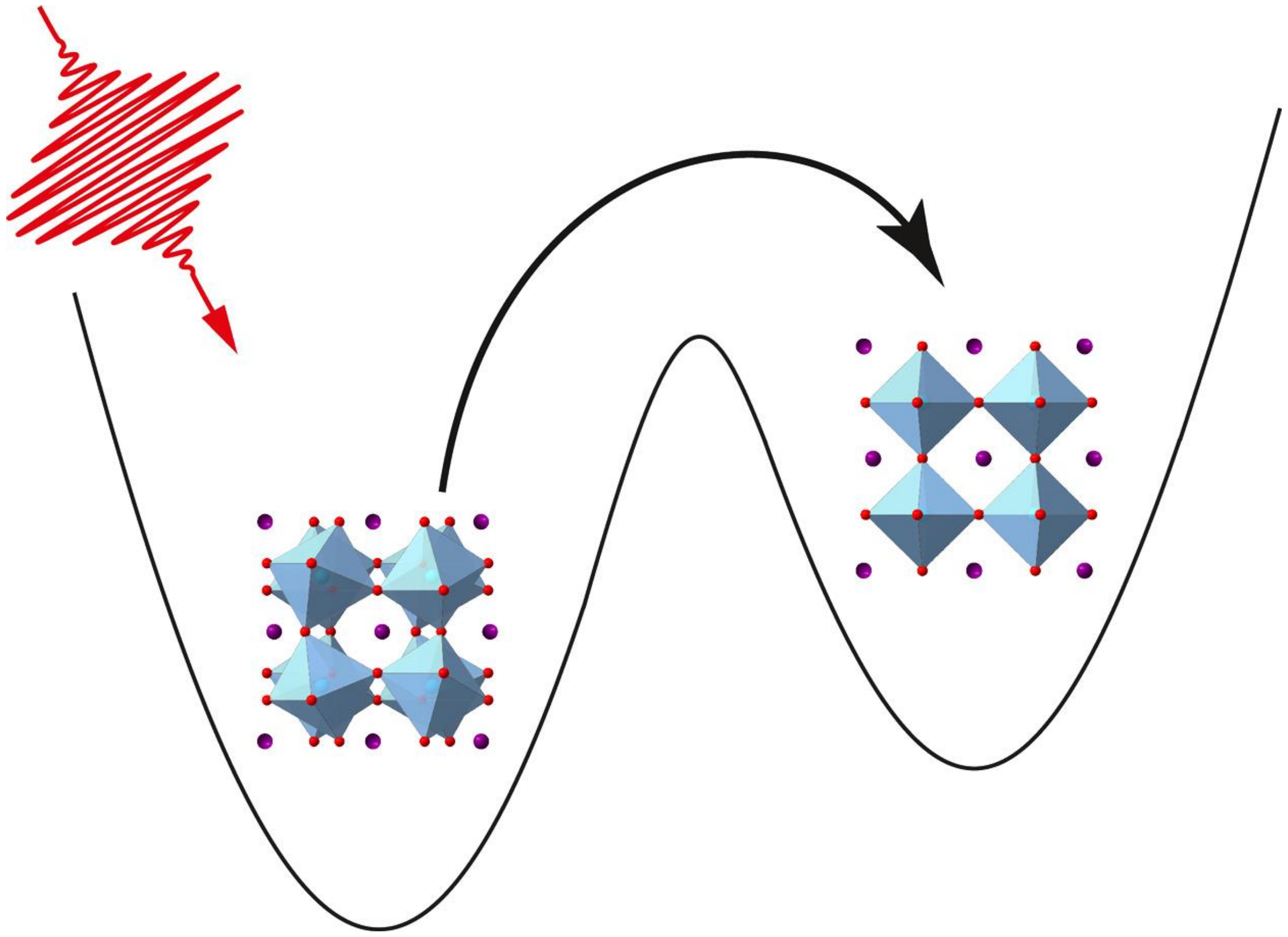
Controlling quantum materials with light

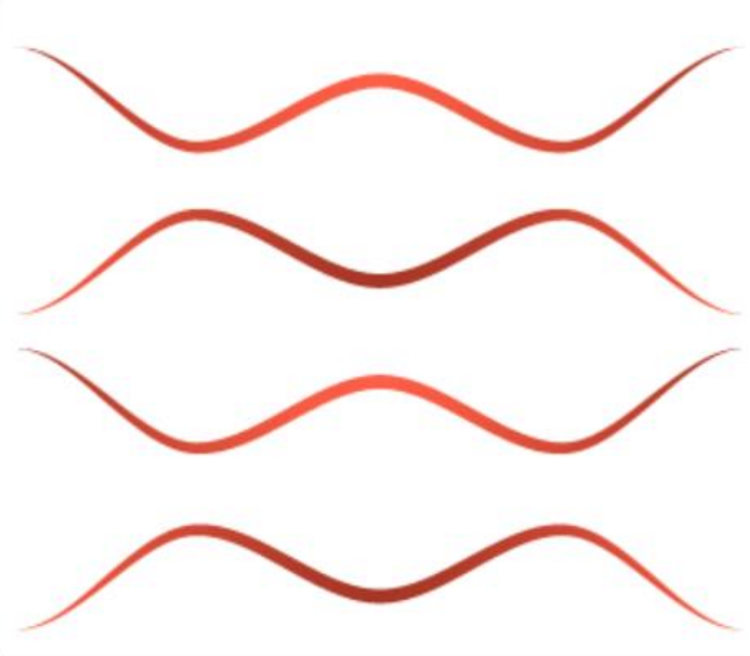


- Generate new states
- Tailor functionalities
- Tune ground state

....

Floquet engineered quantum materials

Coherent optical dressing with intense periodic fields



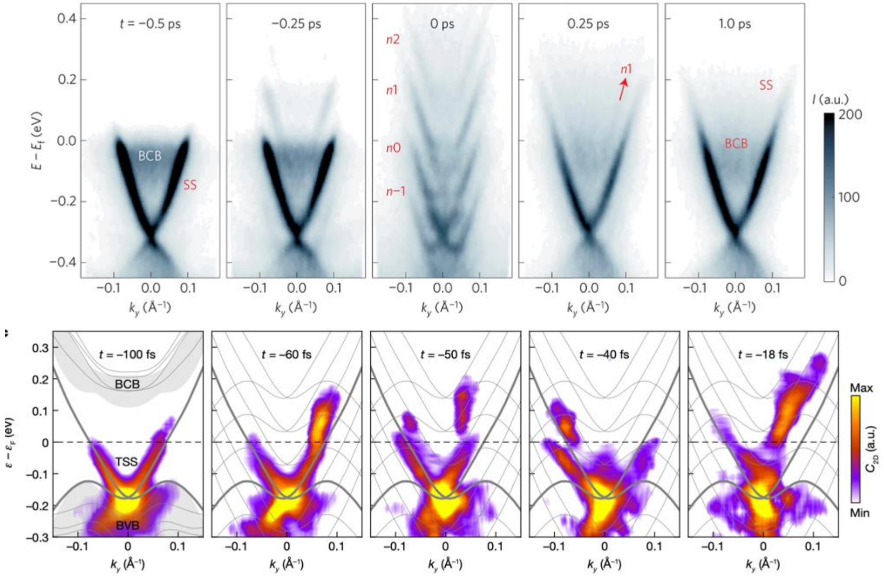
$$\mathcal{H}=\begin{pmatrix} \ddots & H^{-1} & H^{-2} & H^{-3} & H^{-4} \\ H^{1} & H^{0}-(-1)\hbar\omega & H^{-1} & H^{-2} & H^{-3} \\ H^{2} & H^{1} & H^{0}-0\hbar\omega & H^{-1} & H^{-2} \\ H^{3} & H^{2} & H^{1} & H^{0}-1\hbar\omega & H^{-1} \\ H^{4} & H^{3} & H^{2} & H^{1} & \ddots \end{pmatrix}$$

De la Torre, et al. RMP **93**, 041002 (2021)

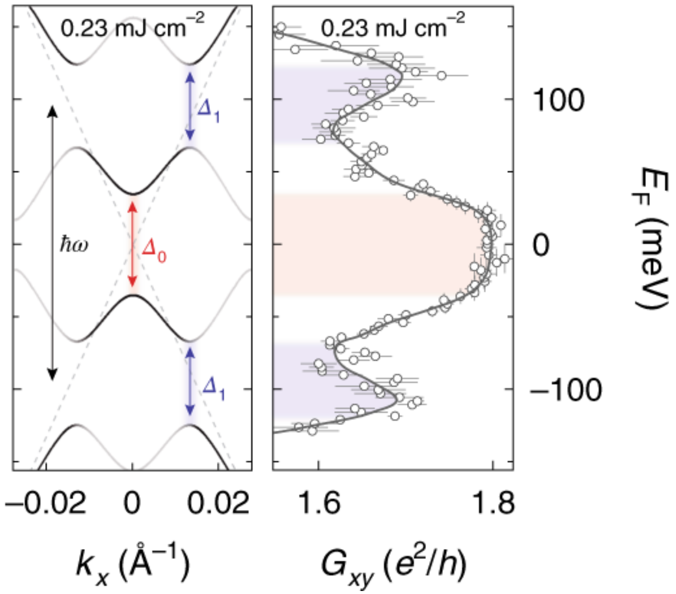
Time-dependent Hamiltonian maps to effective static Hamiltonian with mixed terms

Experimental realizations of Floquet engineering

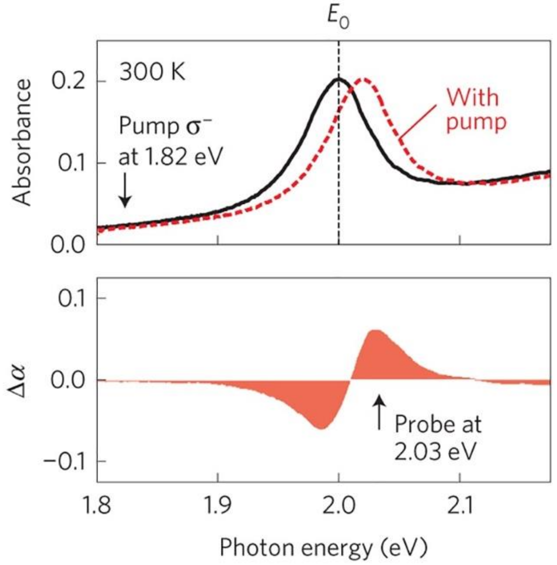
Topological insulators: Gedik, Science (2014), Nat Phys (2016); Huber Nature (2023)



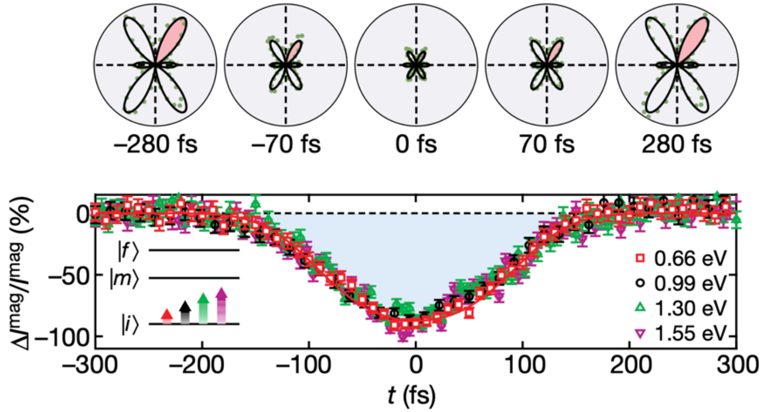
Graphene: McIver, Cavalleri Nat Phys (2020), Gedik (2025), Mathias (2025)

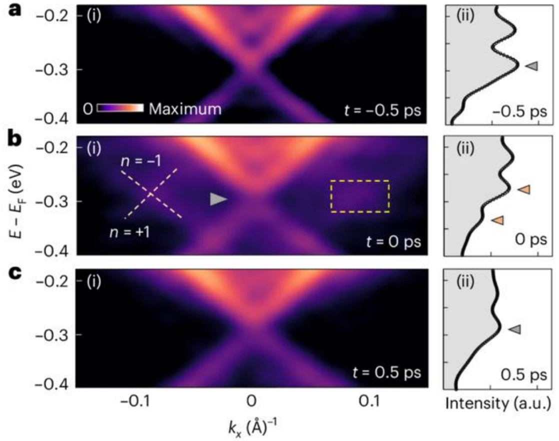


TMDs (WSe_2): Gedik Nat Mat (2014)

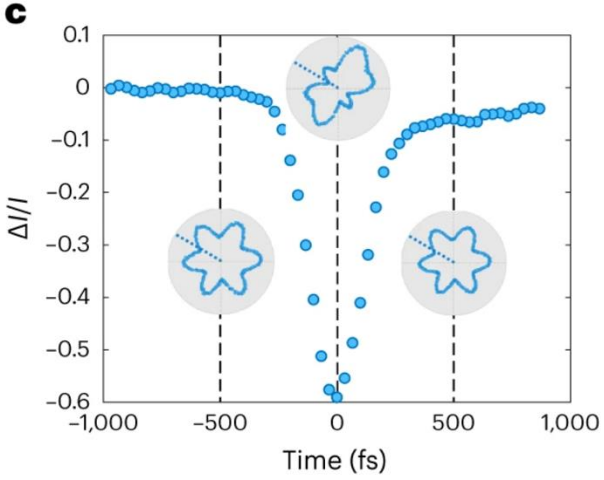


vdW magnets: Hsieh Nature (2021), Mahmood Nat Phys (2025)





Cr_2O_3: Kogar Nat Mat (2021)



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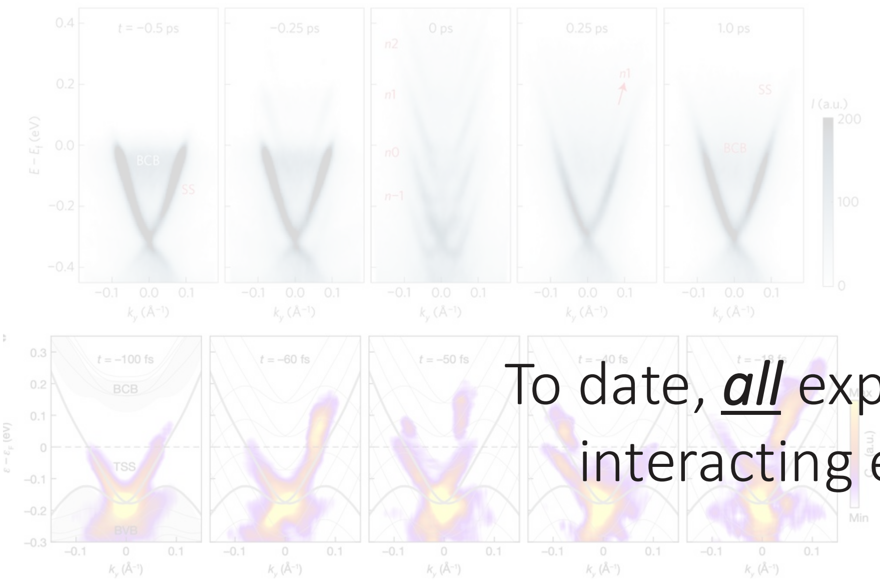
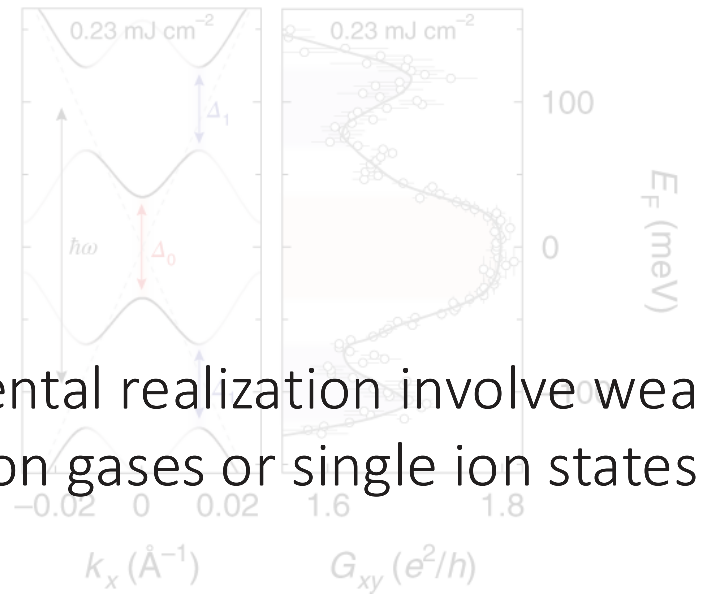


Graphene: McIver, Cavalleri Nat Phys (2020), Gedik (2025), Mathias (2025)



TMDs (WSe_2): Gedik Nature Materials (2014)

To date, ***all*** experimental realization involve weakly interacting electron gases or single ion states

vdW magnets: Hsieh Nature (2021), Mahmood Nat. Phys. (2025)

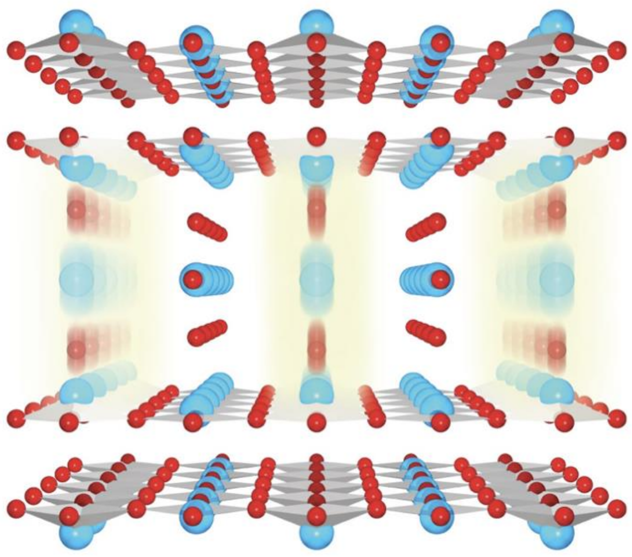
Cr_2O_3: Kogar Nat Mat (2021)

Can we achieve Floquet engineering of strongly correlated systems?

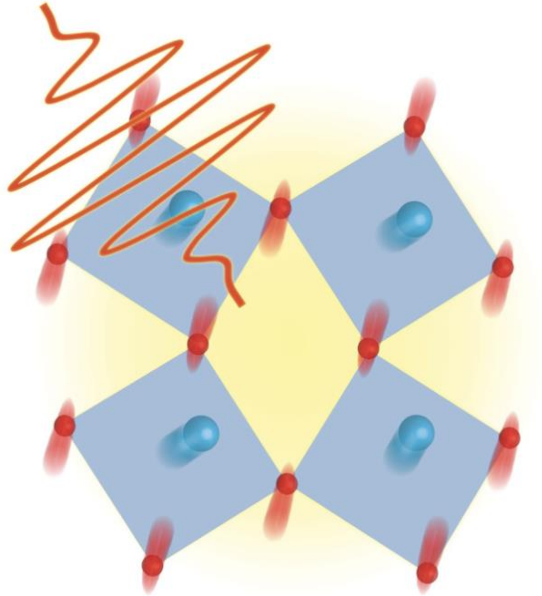
Strongly-correlated systems are extremely susceptible to external stimuli

- ➢ Striking photoinduced responses

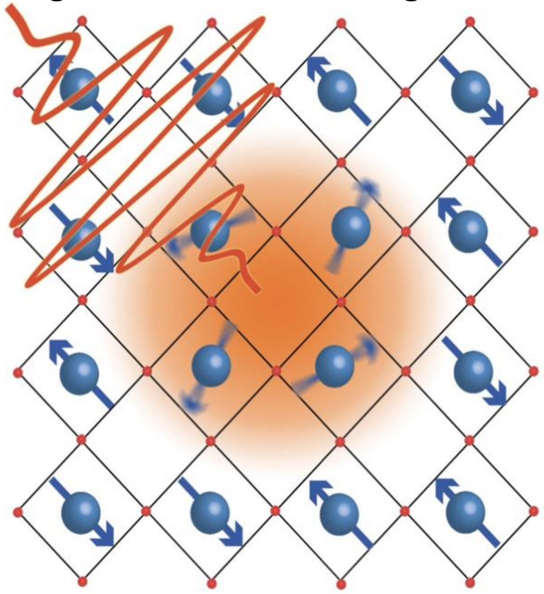
Light-induced superconductivity



Light-induced ferroelectricity



Light-induced ferromagnetism



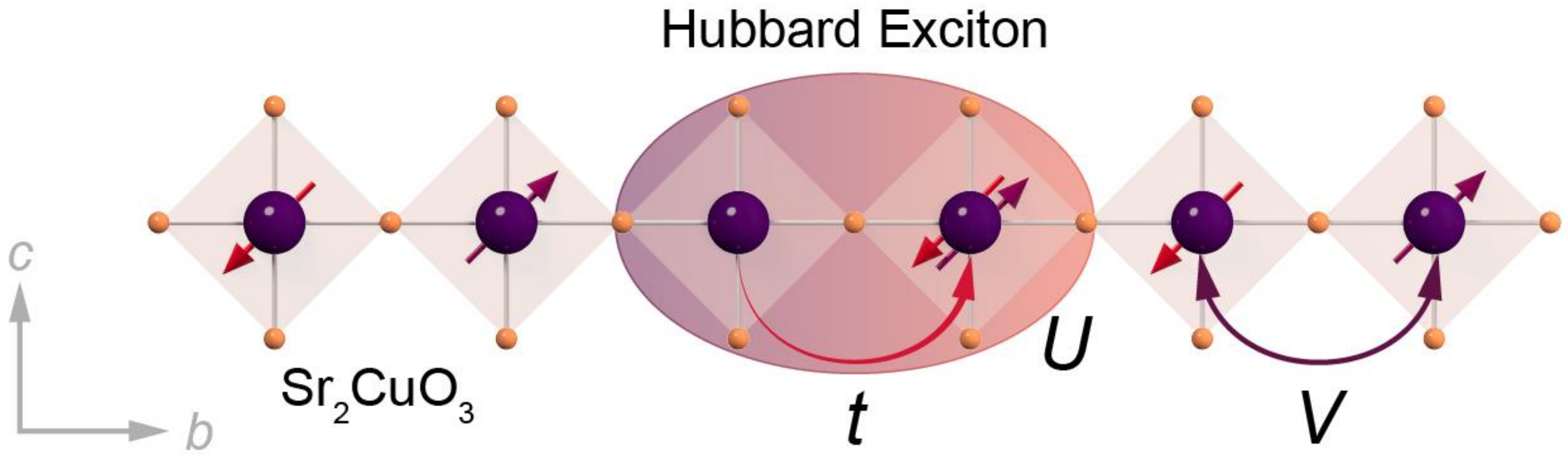
Bloch et al. Nature (2022)

Theoretical proposals for Floquet engineering in correlated systems

Itin and Katsnelson, 2015; Mentink, Balzer, and Eckstein, 2015; Claassen et al., 2017; Chaudhary, Hsieh, and Refael, 2019; Claassen and Sriram, 2021, and many more…

Concern: strong dissipation and interactions may lead to decoherence

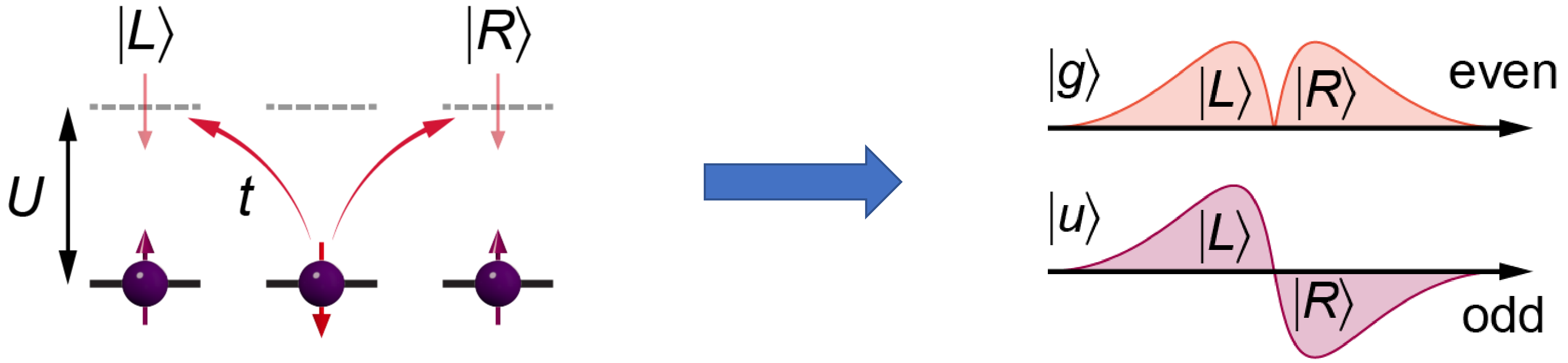
Hubbard excitons as a prototypical correlated state



- One-dimensional spin-1/2 Mott insulator
- Spin-charge separation
- Bound Hubbard exciton states
- Centrosymmetric

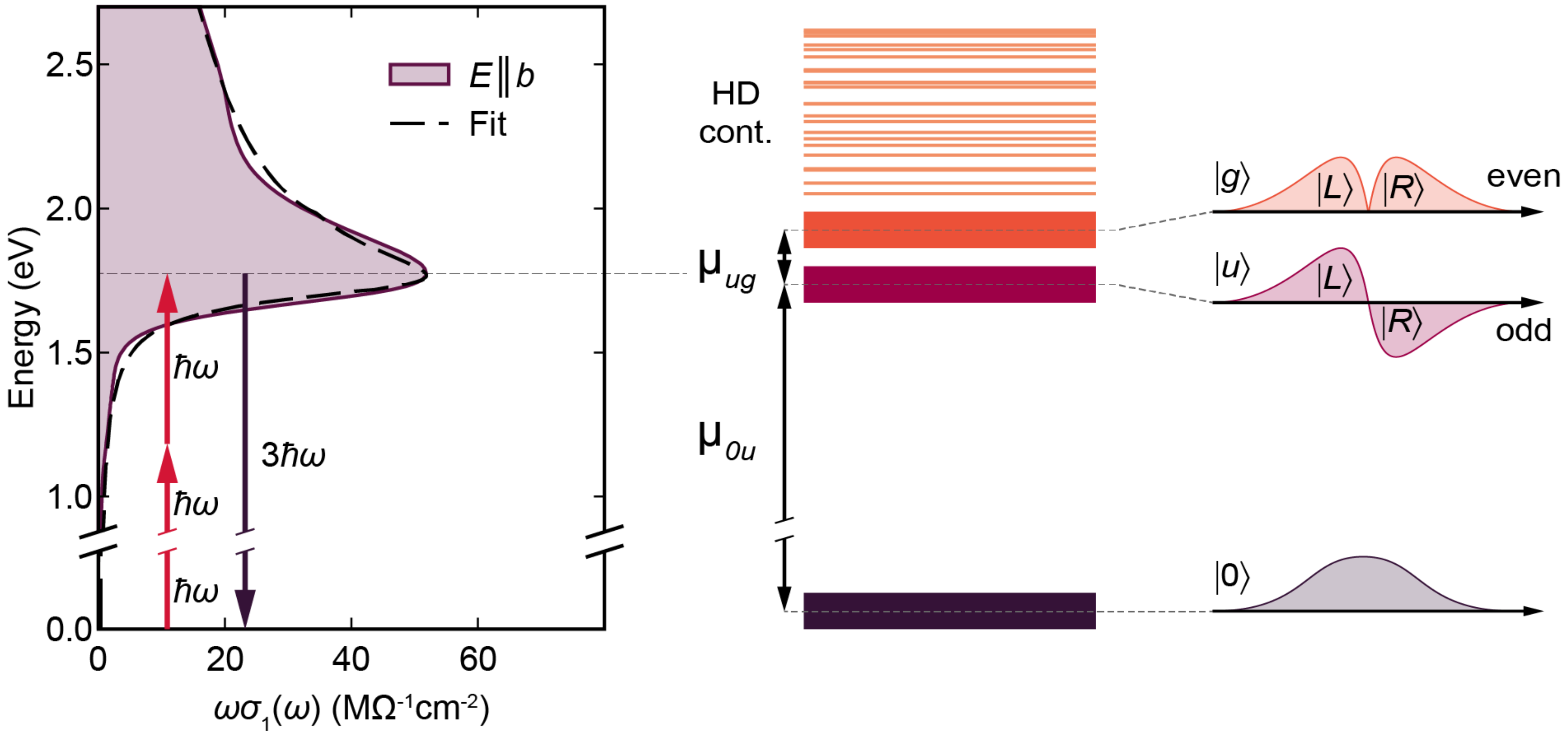
$$U = 3.96\ eV$$
$$V = 1.17\ eV$$
$$t = 0.56\ eV$$

Optical properties of the Hubbard excitons



Almost degenerate Hubbard excitons with opposite parity

Optical properties of the Hubbard excitons



Giant optical nonlinearity

$$\chi^{(3)}(-3\omega;\omega,\omega,\omega) \propto \frac{\mu_{ug}^2 \mu_{0u}^2}{(\epsilon_{HE}/\hbar - 3\omega - i\gamma)(\epsilon_{HE}/\hbar - 2\omega - i\gamma)(\epsilon_{HE}/\hbar - \omega - i\gamma)},$$

$\approx 1.4\ 10^7\ \mathrm{pm^2/V^2}$)

Kishida, et al Nature 2000
Ogasawara, et al Phys. Rev. Lett. 2000
Mizuno, et al Phys. Rev. B, 2000.
Kishida, et al Phys. Rev. Lett., 2001
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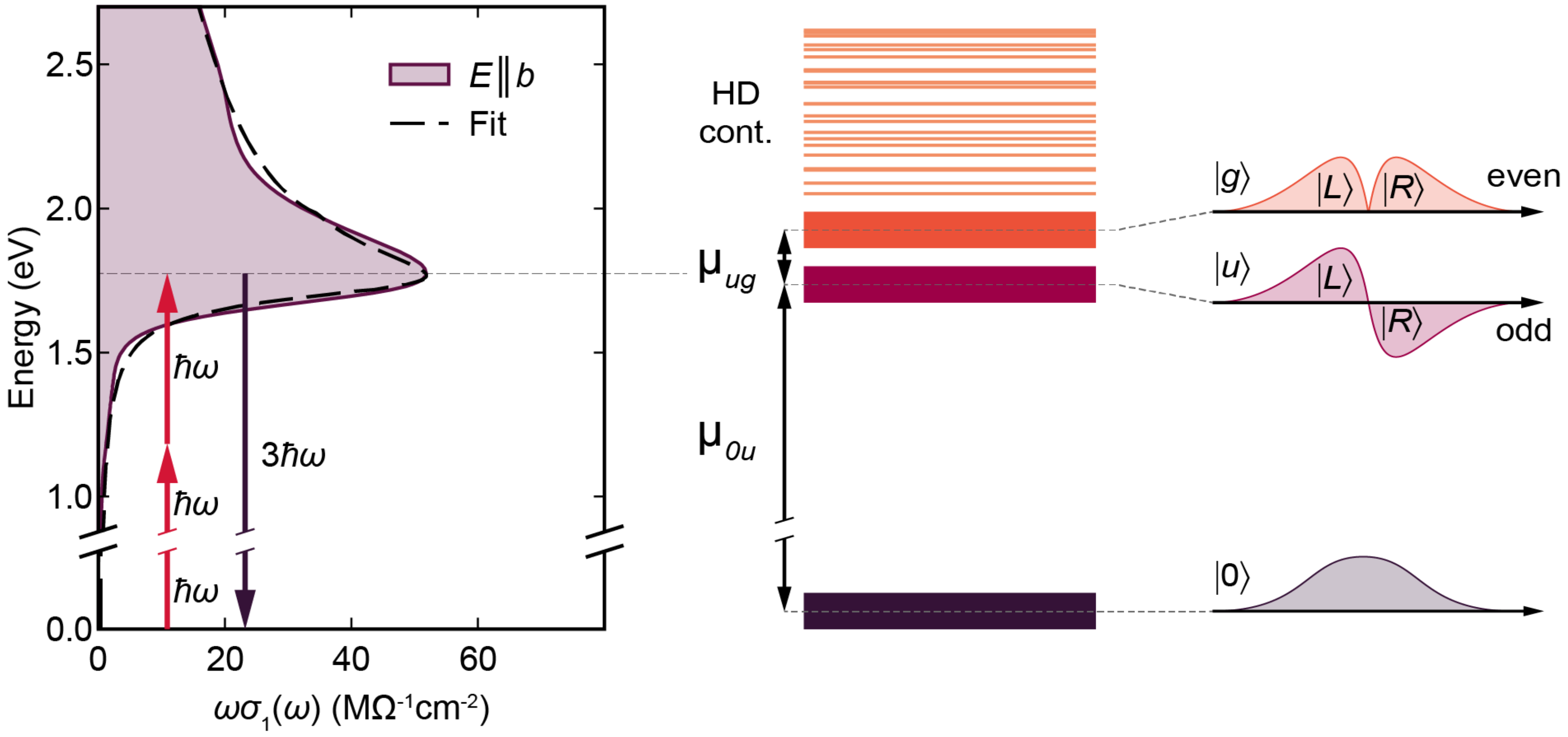

Hubbard excitons can be interrogated with third harmonic generation

Higher order response functions encode symmetry information beyond linear response

Kishida, et al Nature 2000
Ogasawara, et al Phys. Rev. Lett. 2000
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Kishida, et al Phys. Rev. Lett., 2001
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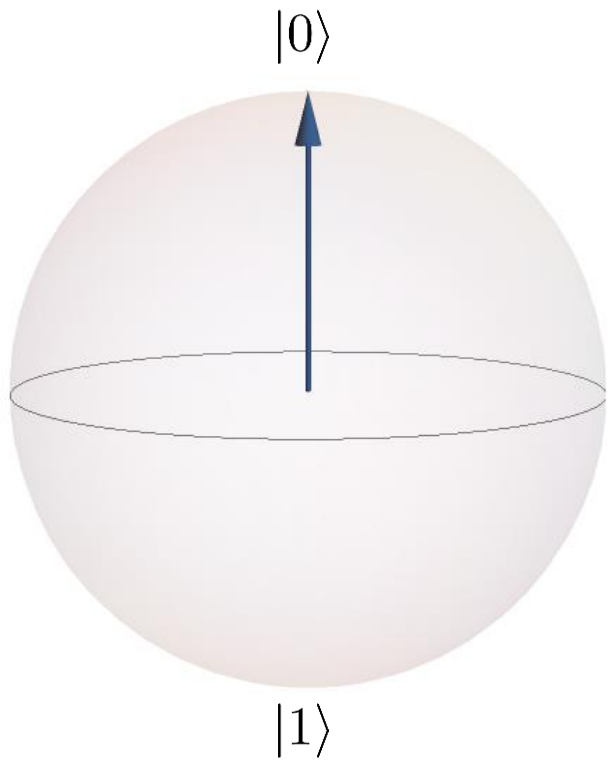
Quantum control of the Hubbard exciton

Odd and even exciton states define a Bloch sphere



Under nonresonant periodic driving (Floquet), usually:

- Energy shifts
- Wavefunction mixing

For degenerate states, mixing dominates

$$|u(t)\rangle = \cos\frac{\vartheta(t)}{2}|u\rangle + e^{i\varphi(t)}\sin\frac{\vartheta(t)}{2}|g\rangle$$
$$|g(t)\rangle = \cos\frac{\vartheta(t)}{2}|g\rangle + e^{i\varphi(t)}\sin\frac{\vartheta(t)}{2}|u\rangle$$

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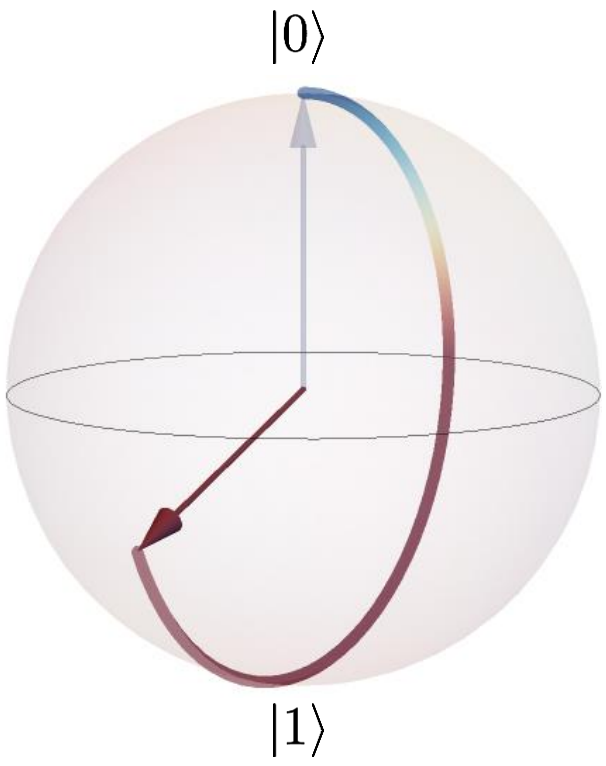
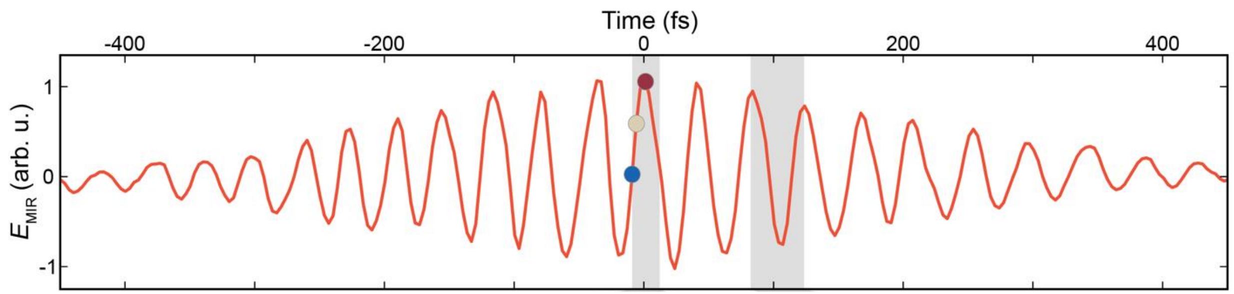


Mixing can be conceptualized as a rotation on the Bloch sphere

Rotating and probing the Hubbard exciton



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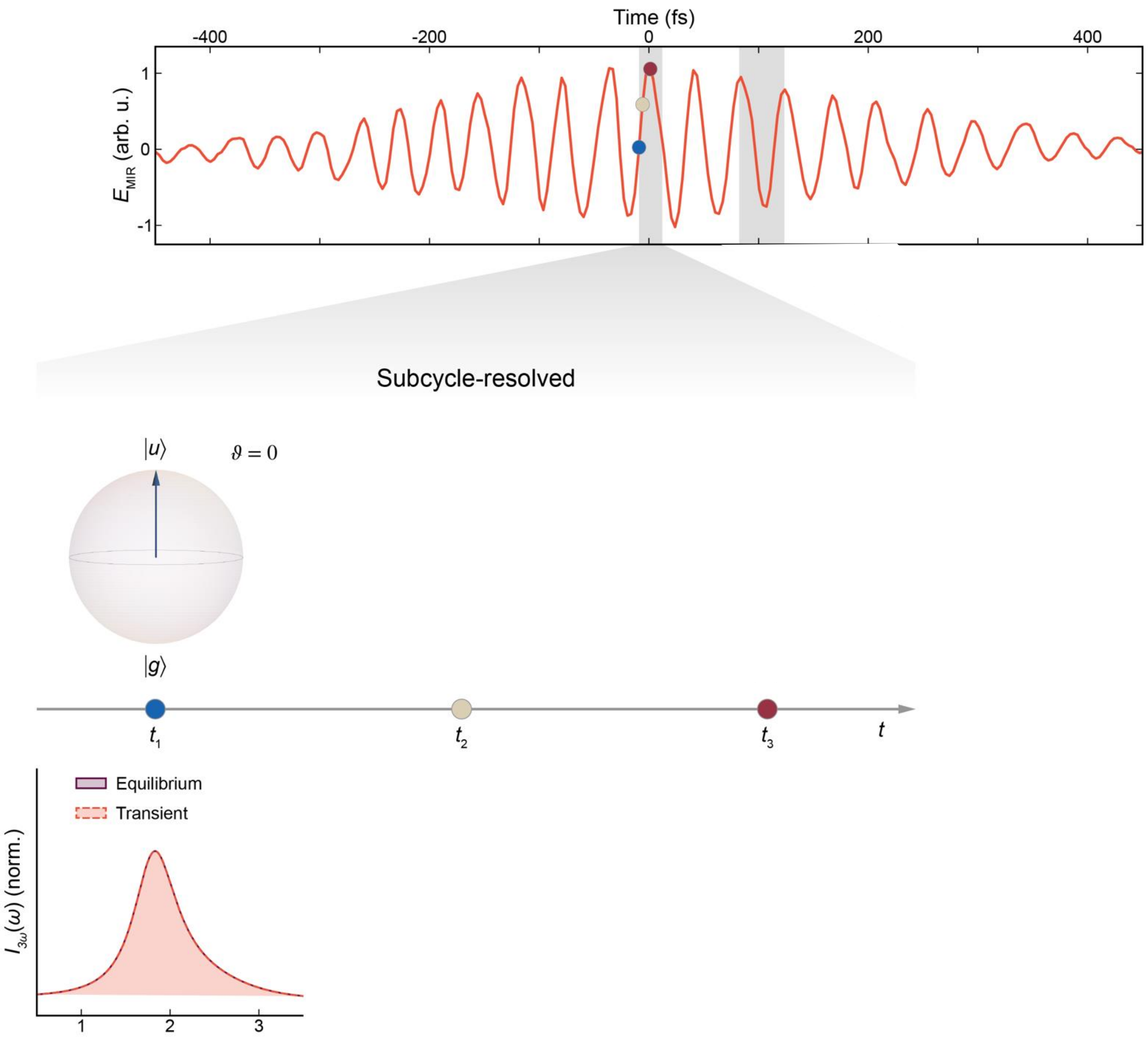

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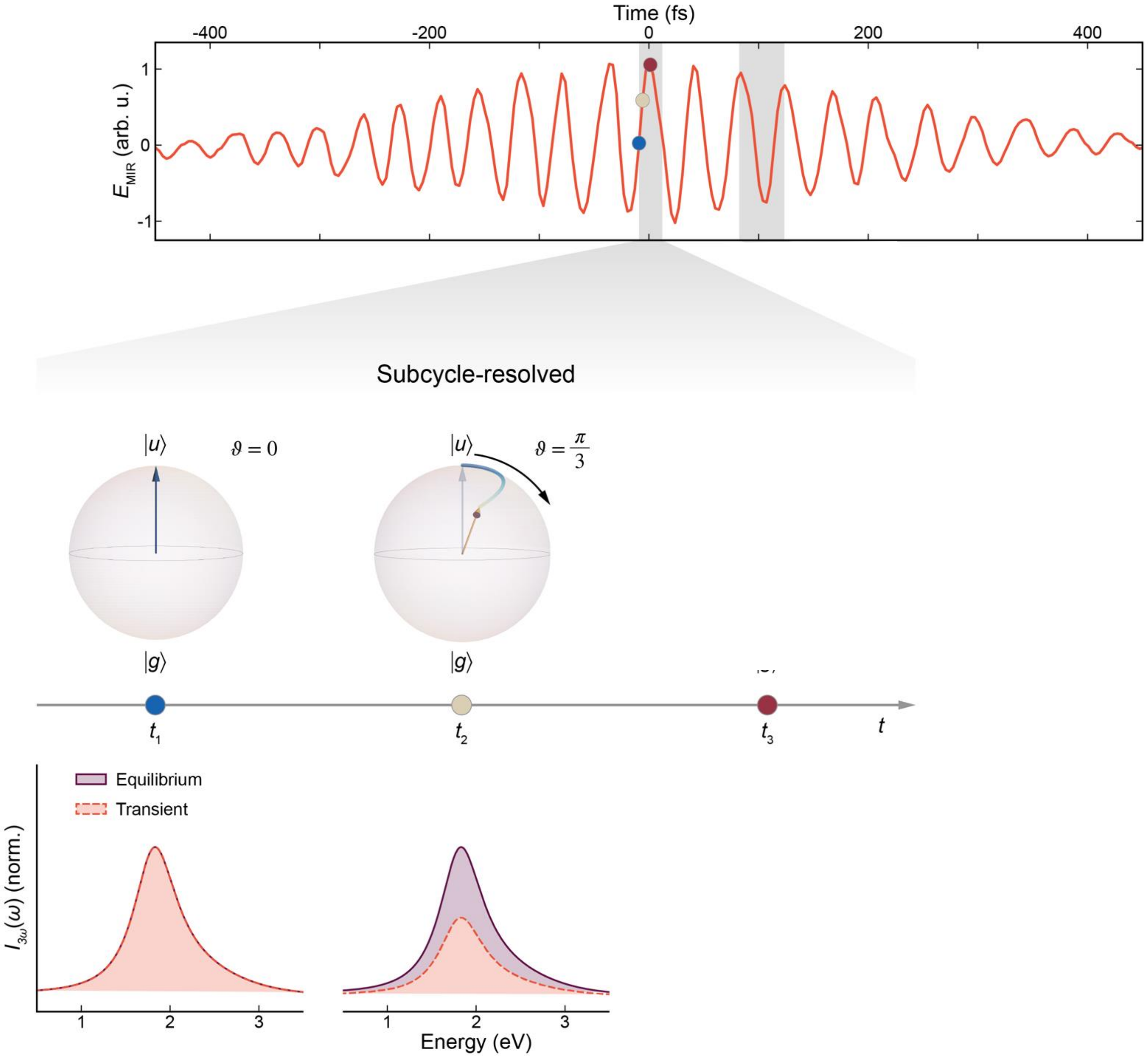

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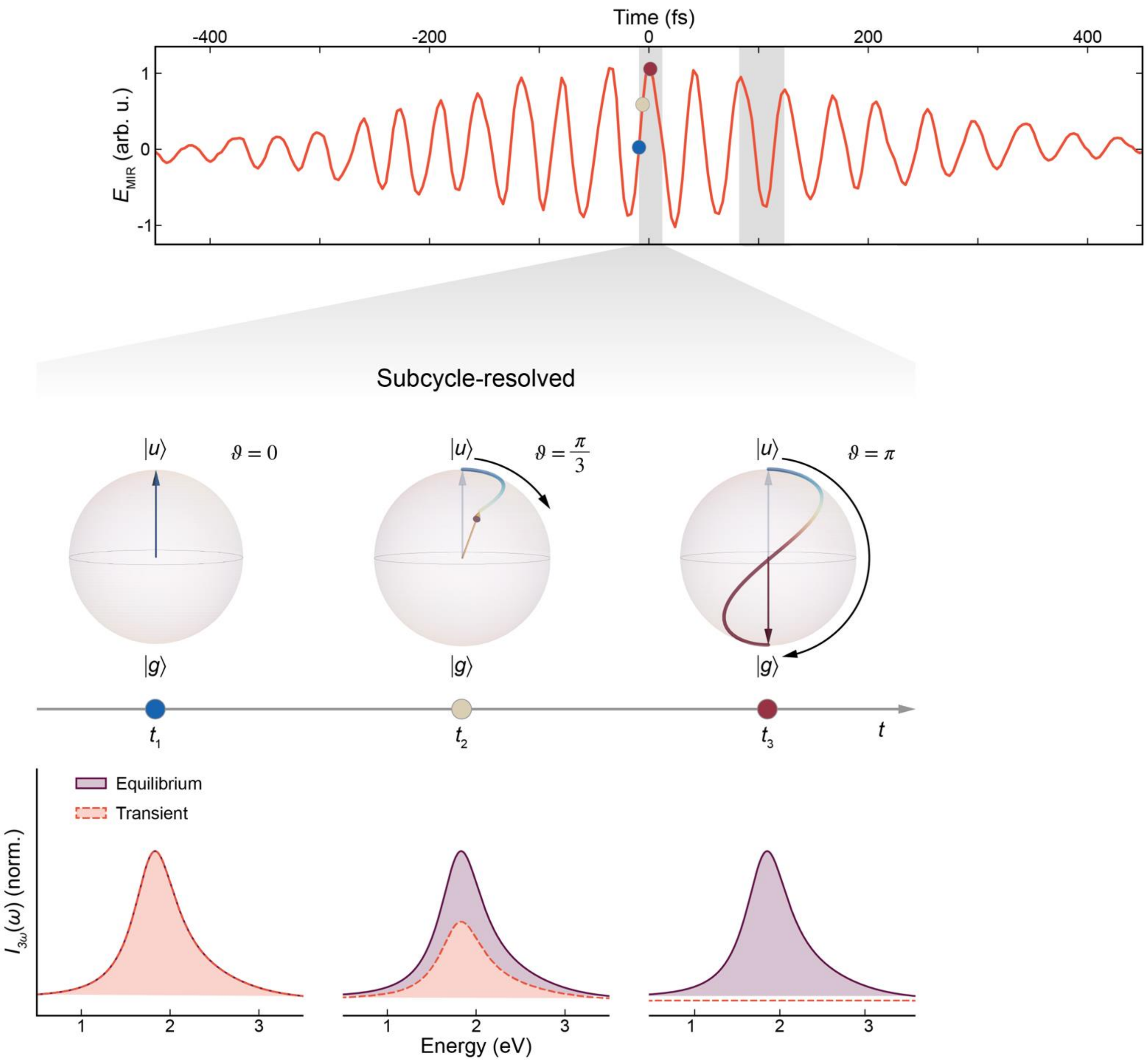

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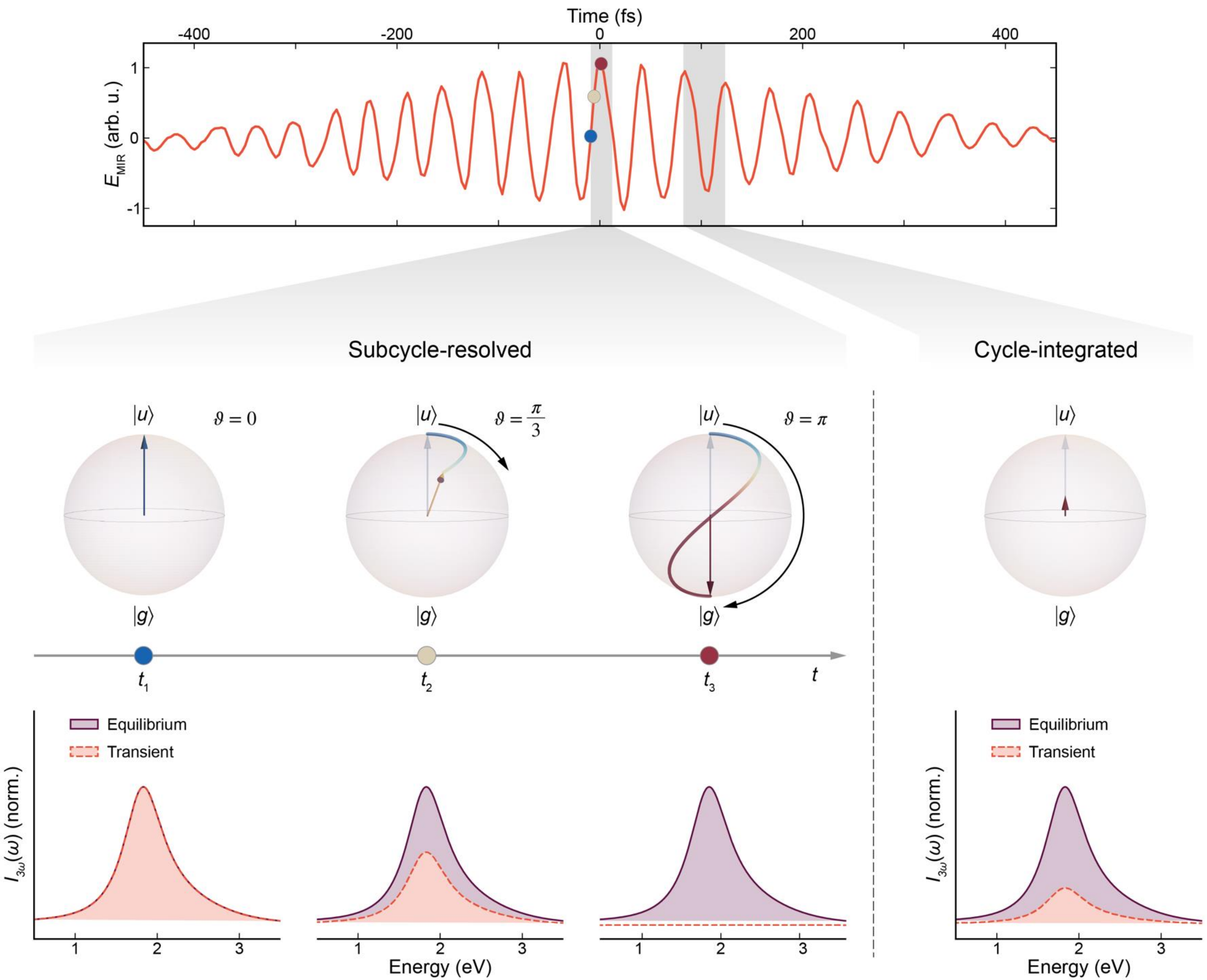
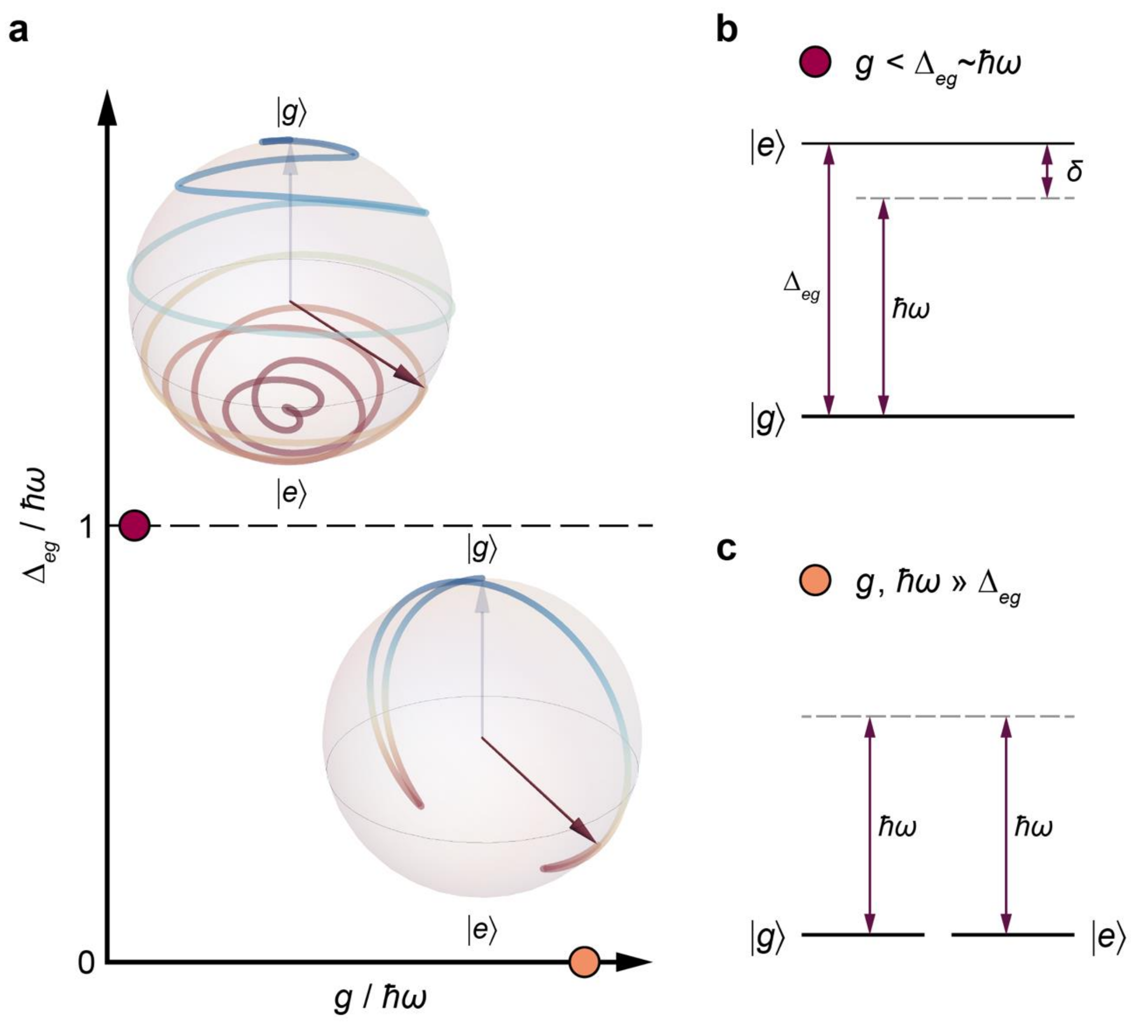


Quantum optical description of the Floquet manipulation



Conventional quantum-optical Rabi problem

Strong coupling quantum-optical Rabi problem

Distinct regime from semiconductor quantum wells and spins by Awschalom, Cundiff, Imamoglu, Li, Sherwin, …

e.g. *Phys. Rev. Lett.* **83**, 4204 (1999), *Phys. Rev. Lett.* **73**, 1178 (1994), *Nature* **410**, 60–63 (2001), *Science* **301**, 809 (2003), *Science* **310**, 651 (2005), *Nature* **453**, 1043–1049 (2008), and more…

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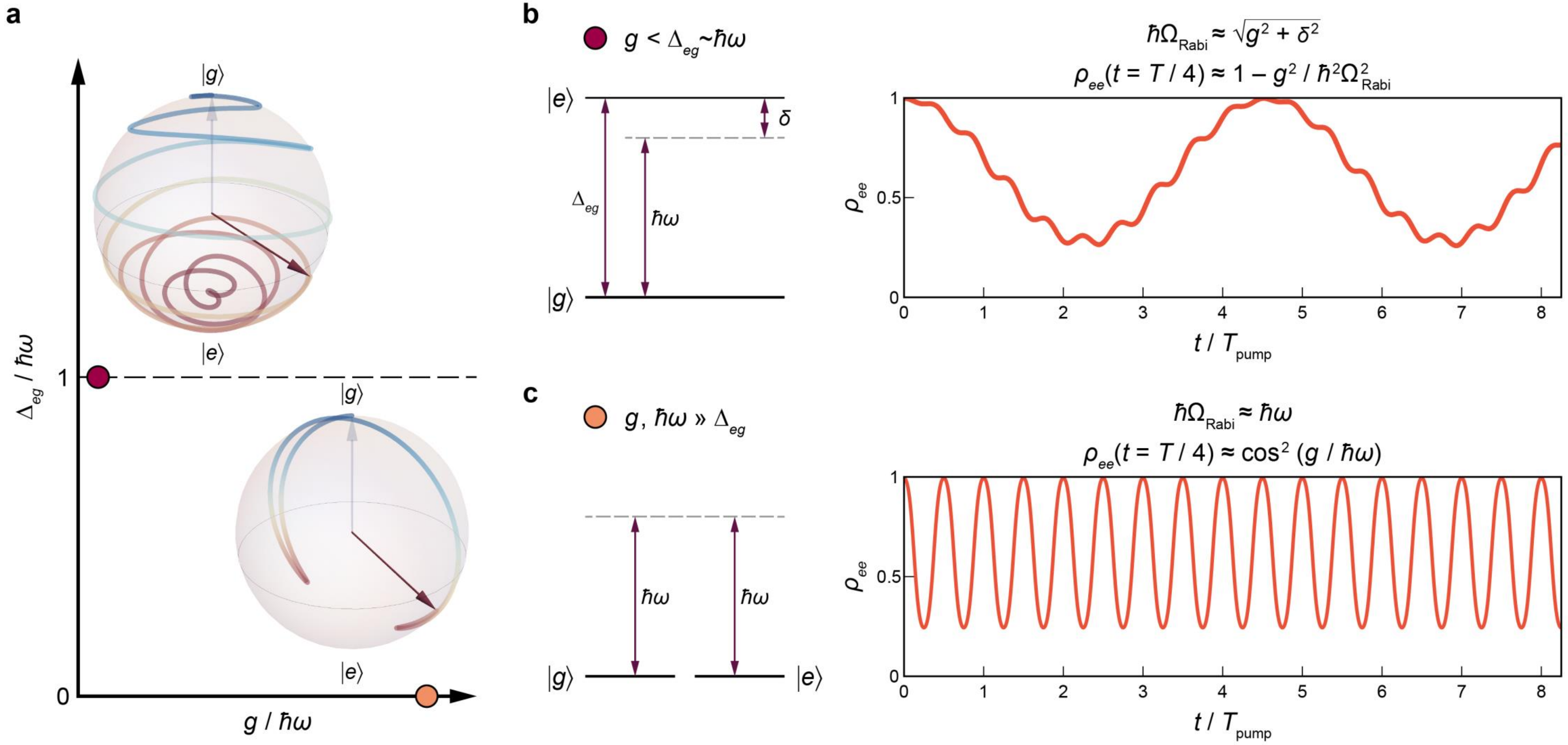

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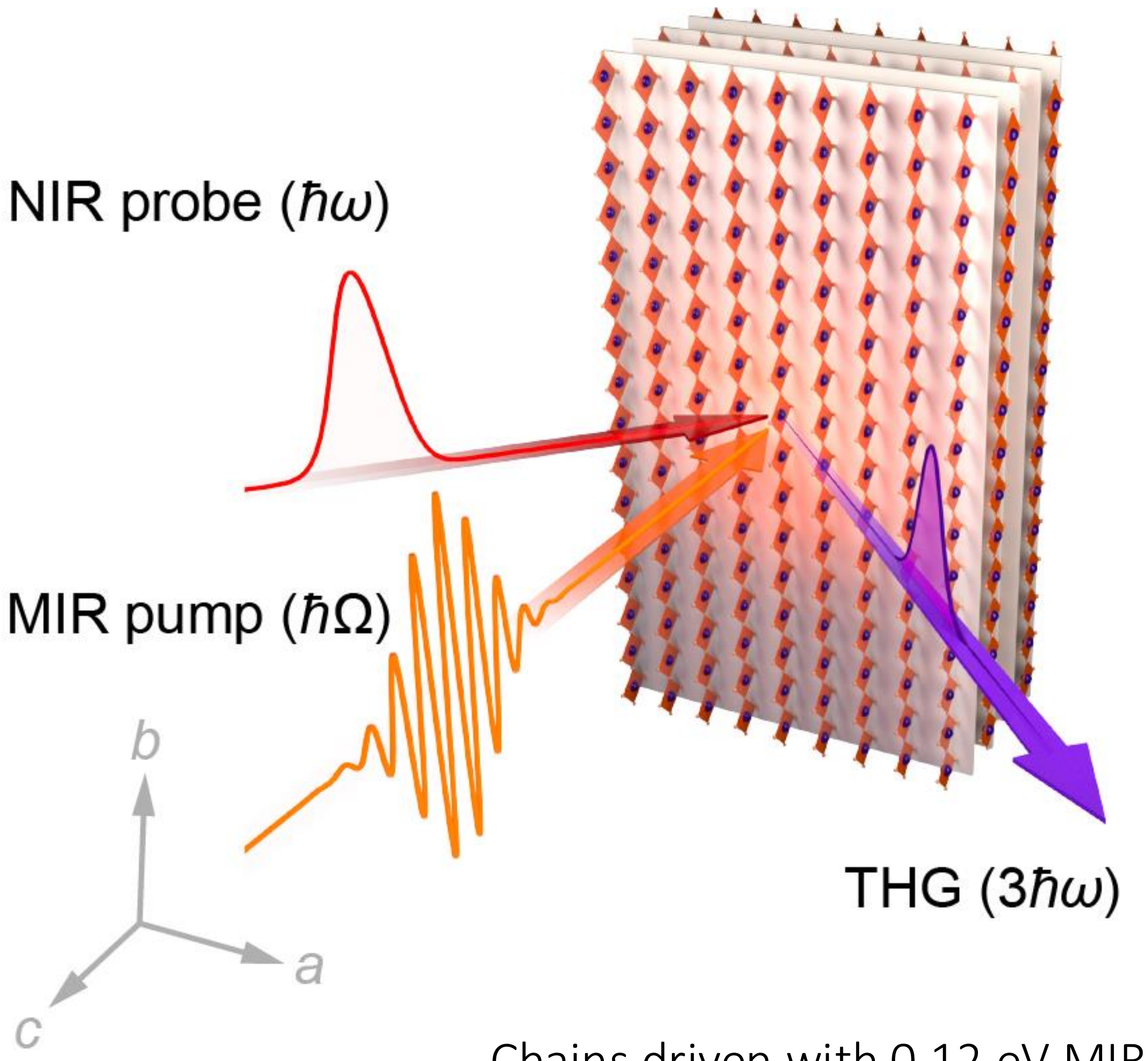
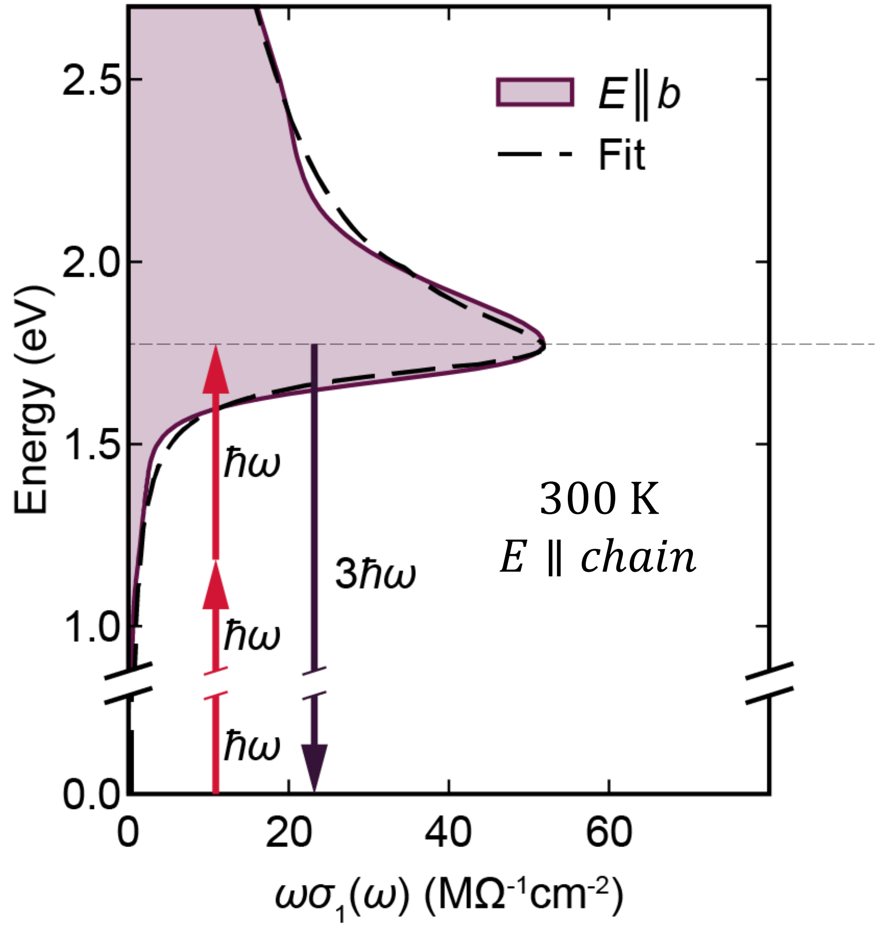
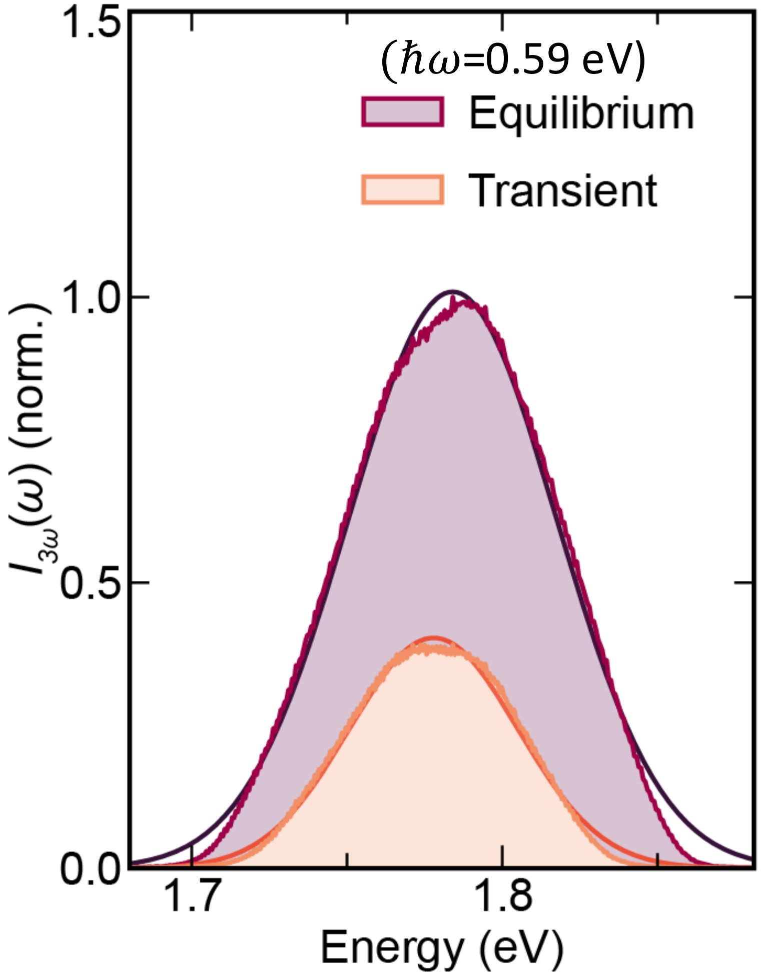
The experimental platform





Chains driven with 0.12 eV MIR pulses ($E = 1.8\ MV/cm$)

Observation of a dynamically renormalized THG



- THG response suppressed by MIR pump
- Preserved C_2 symmetry (no higher-order multipoles)
- Suppression scaling as $J_0^4(A\cos\theta)$

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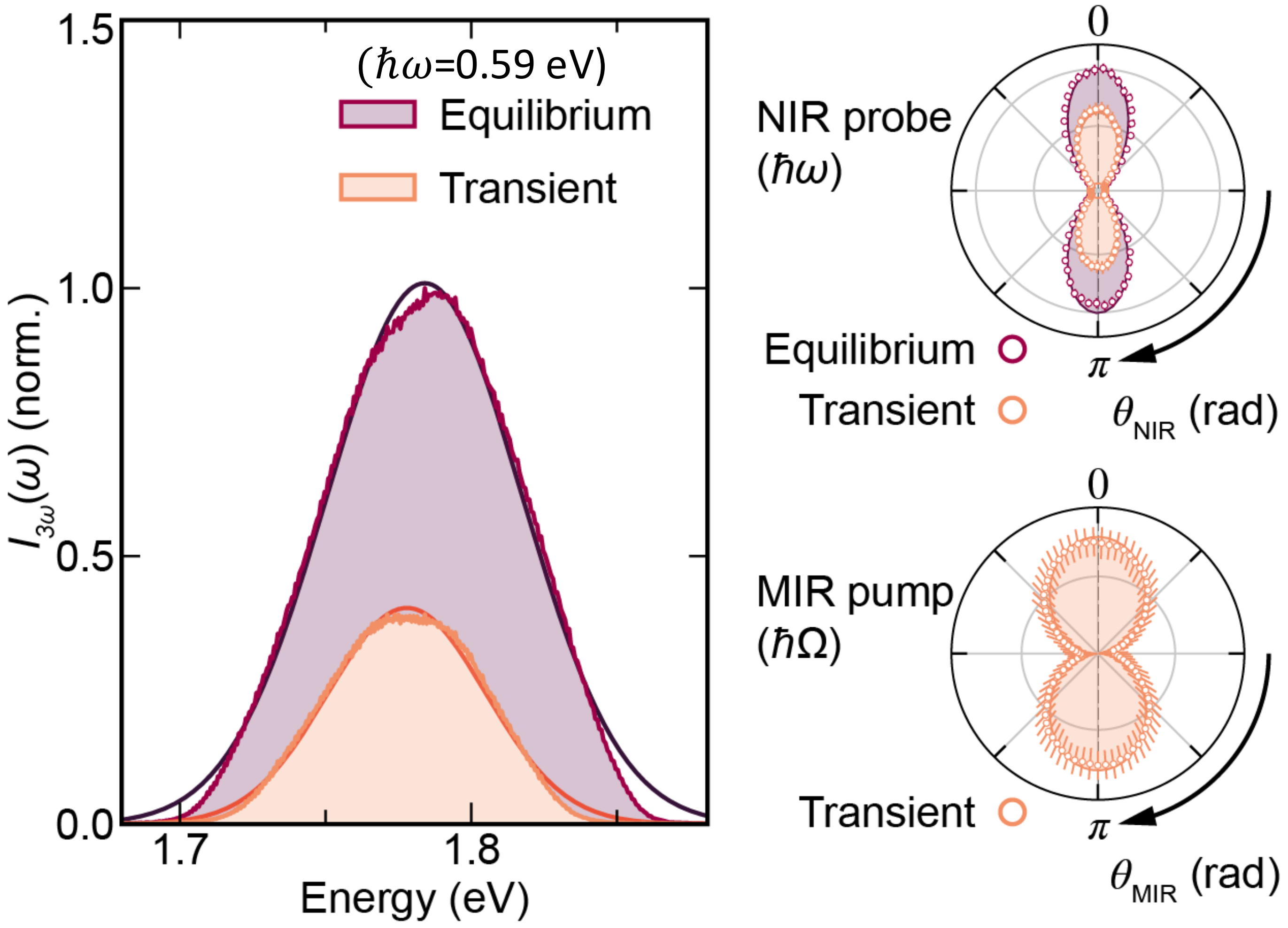

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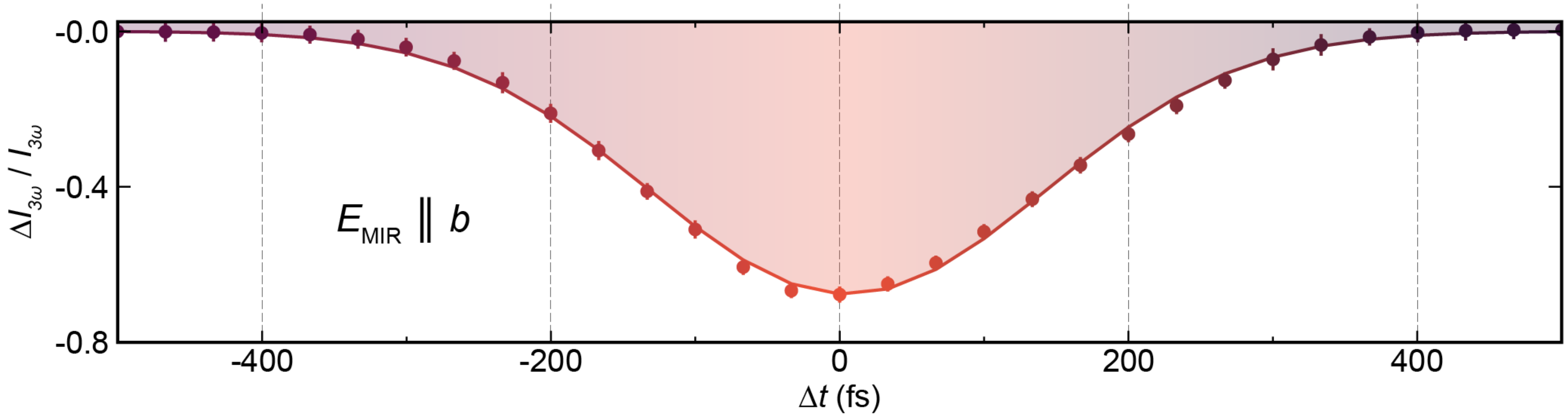
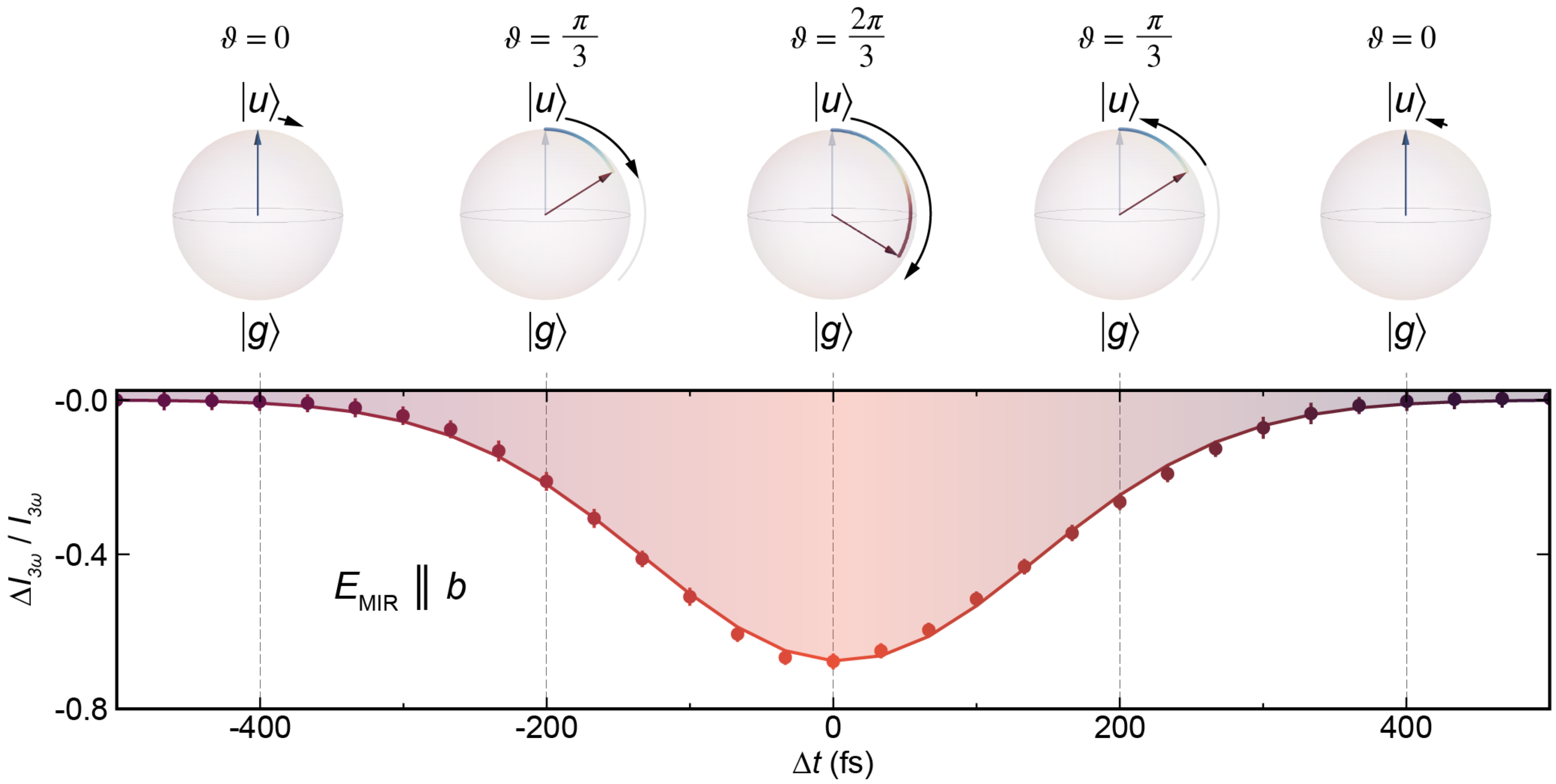
Time-dependent THG renormalization



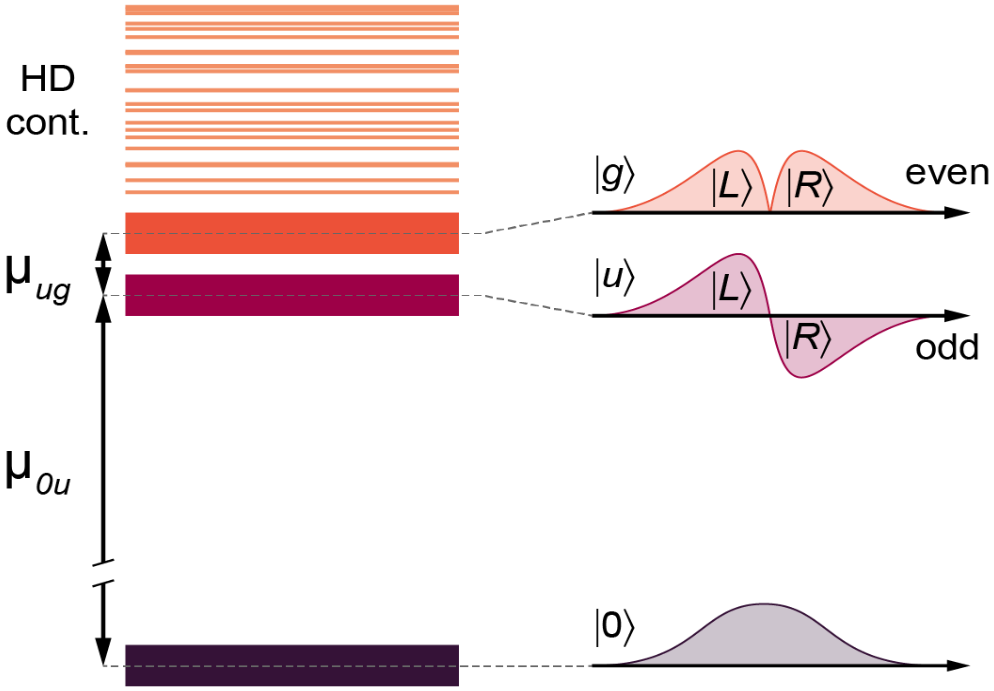
- THG suppression lasts as MIR (300 fs) and NIR (80 fs) crosscorrelation
  - ➢ Floquet dressing of Hubbard excitons

Time-dependent THG renormalization



- THG suppression lasts as MIR (300 fs) and NIR (80 fs) crosscorrelation
  - ➢ Floquet dressing of Hubbard excitons
- Three-level approximate estimation yields rotation angles above $\pi/2$

Periodic driving introduces Floquet sidebands



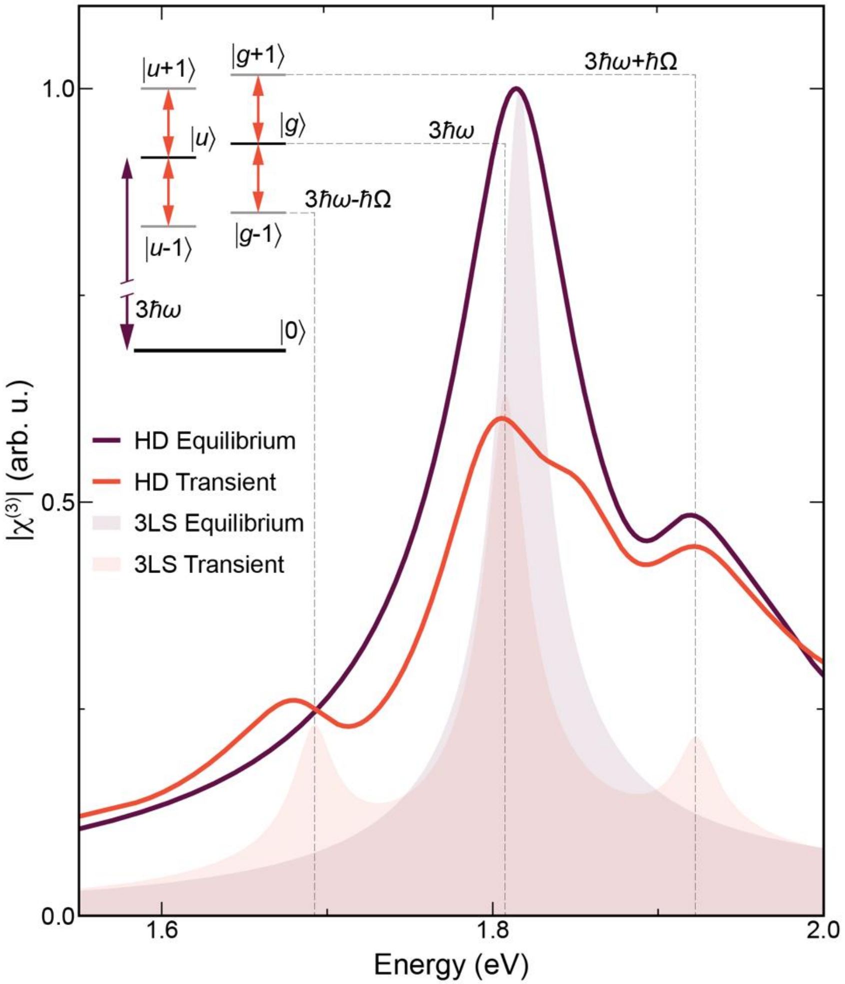
$$H(t) = H_{hd} - p \cdot E\cos(\Omega t)$$

Long MIR pulse duration enables Floquet $\chi^3(-\omega_\sigma\,;\omega_p,\omega_q,\omega_r)$ calculation for

- Three-level system ($|0\rangle$, $|u\rangle$ and $|g\rangle$)
- Full holon-doublon spectrum (including HD continuum)

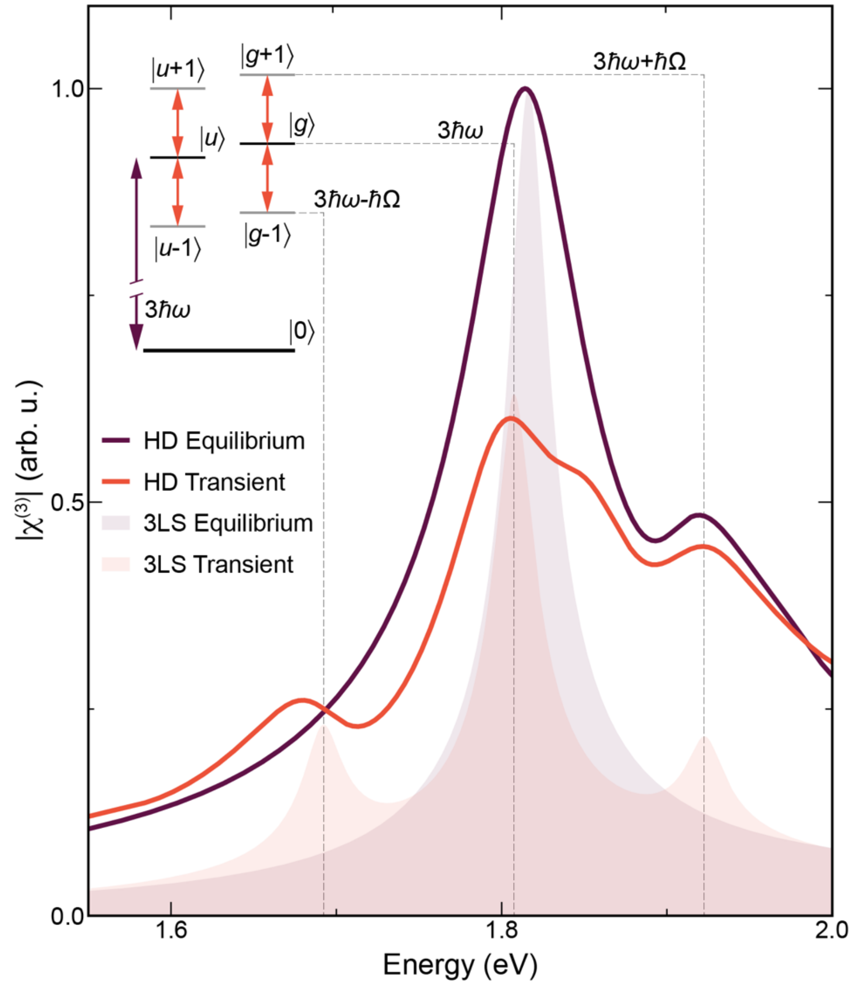
$$\chi^{(3)} \propto \sum_n \frac{X_{gu}^2 X_{u0}^2 J_n^2(\frac{\mathcal{E} X_{gu}}{\Omega})}{(E_{exc} - n\Omega - 3\omega - i\eta)(E_{exc} - n\Omega - 2\omega)(E_{exc} - n\Omega - \omega)}$$

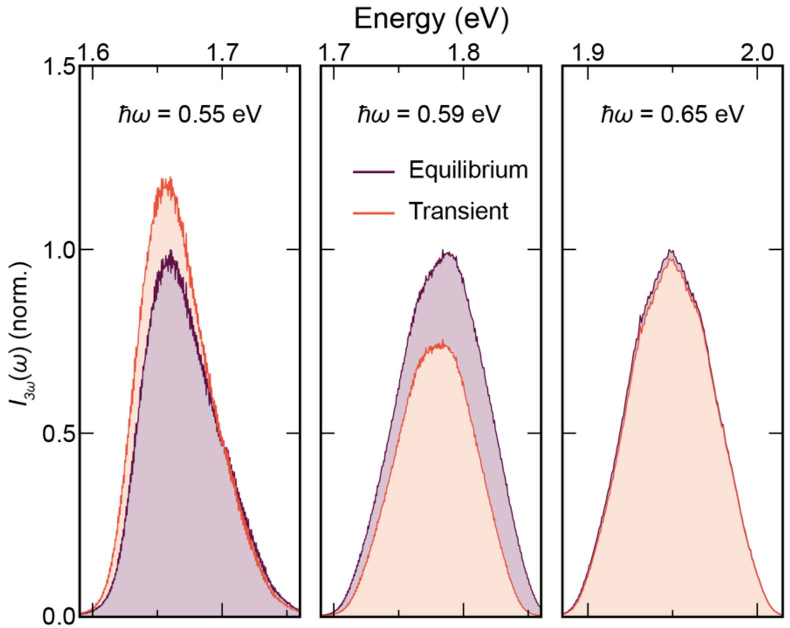
Periodic driving introduces Floquet sidebands



Periodic driving introduces **THG enhancement** upon detuning

Probing Floquet sidebands





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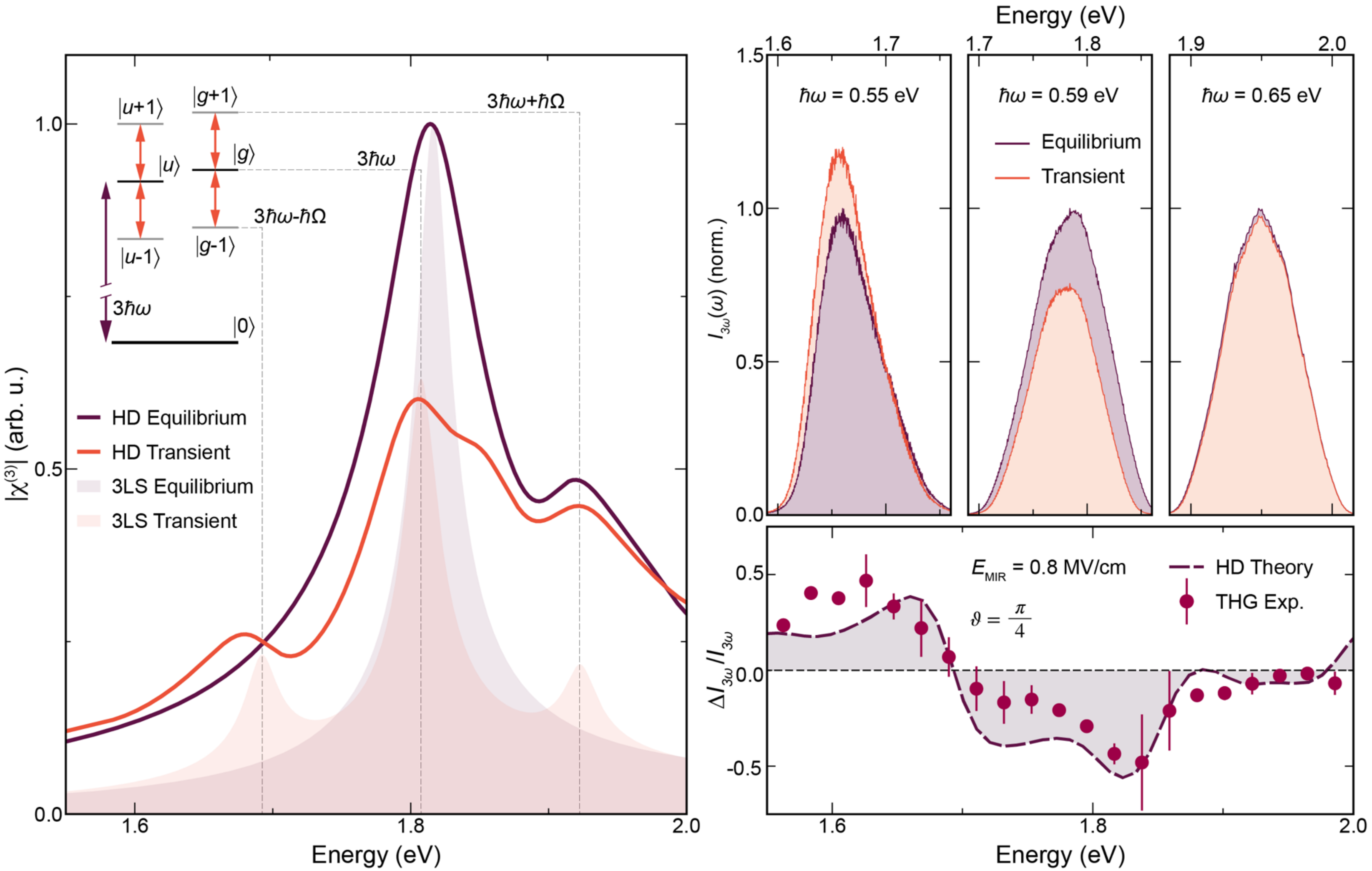
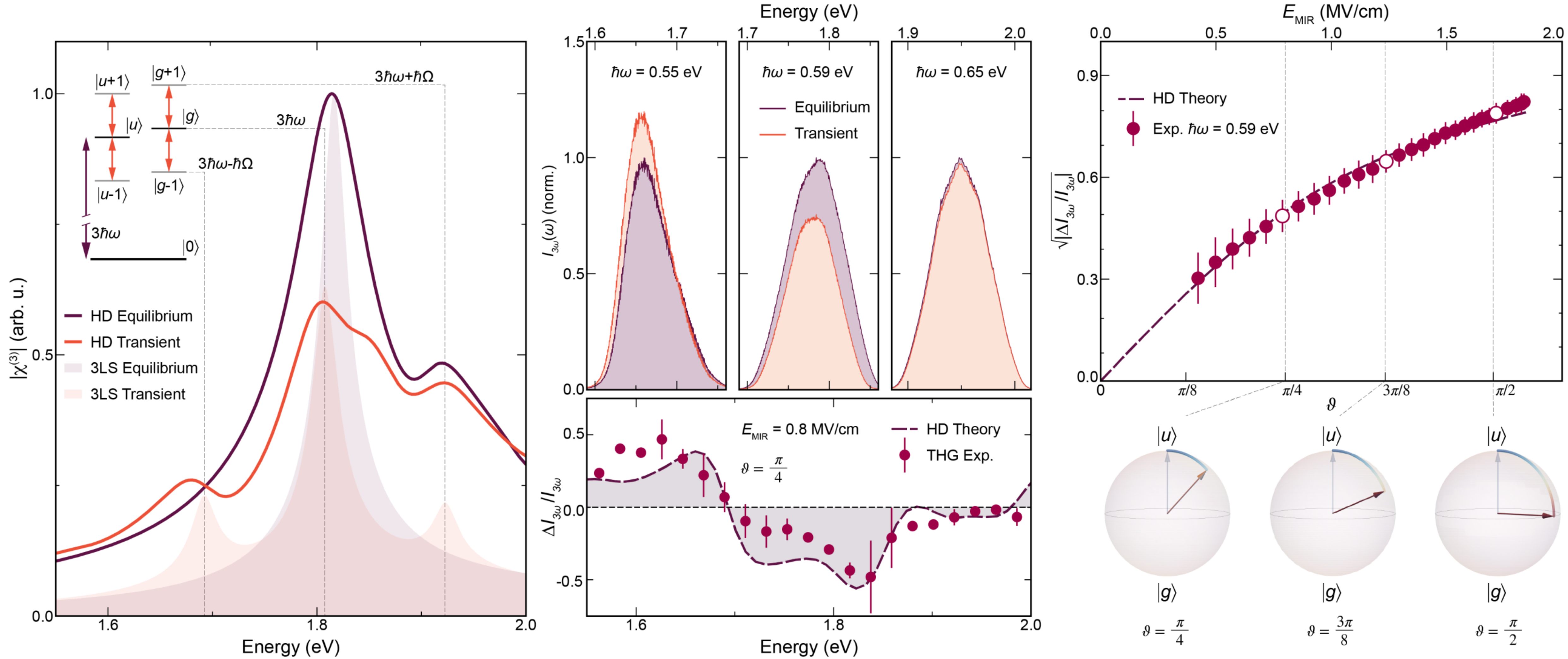


Observation of **Floquet THG enhancement**

- χ^4 peaks symmetry forbidden
- Not an exciton shift
- No tradeoff with χ^3, χ^5 processes

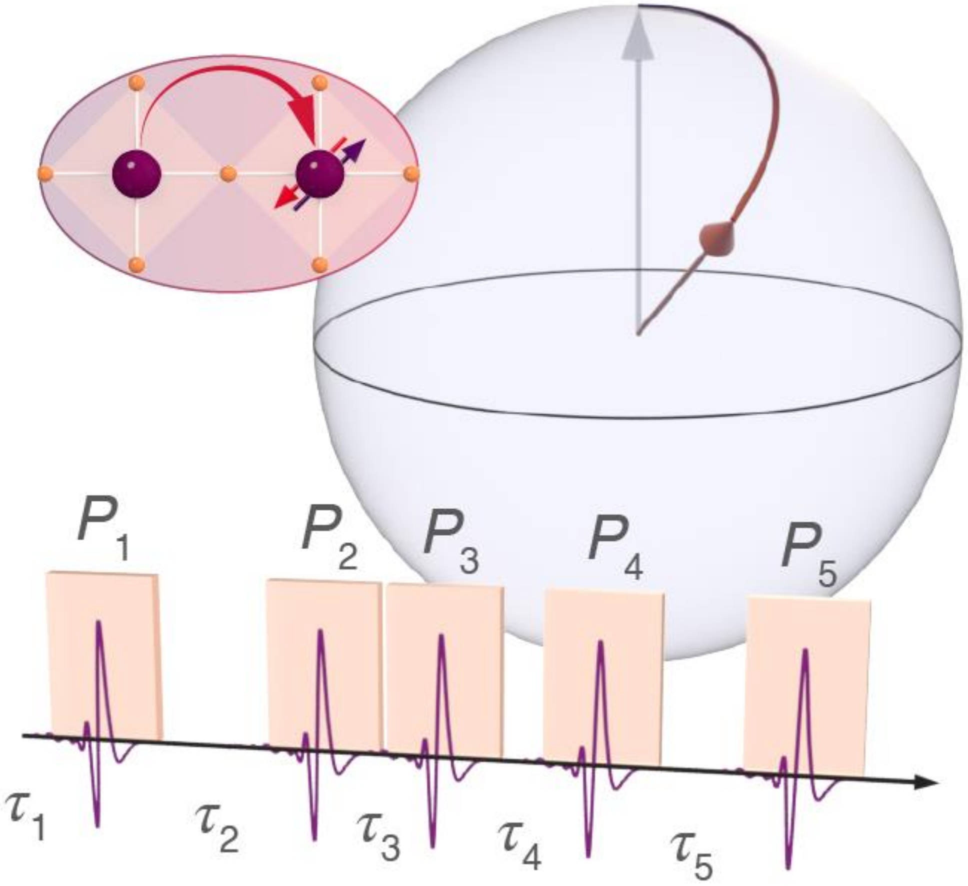
Achieving a $\pi/2$ Bloch angle rotation



Extracting Bloch angle from HD Floquet theory

- $\pi/2$ pulse rotation at 1.7 MV/cm
- Complete π-rotation at ≈ 3 MV/cm

Towards "THz NMR-like" control of correlated quantum phases



- $\pi/2$ pulses are the basic building block of any NMR-like quantum control sequence

  Ramsey, Hahn echo, WAHUHA, and more…

  Choi, Lukin et al. PRX **10**, 031002 (2020)

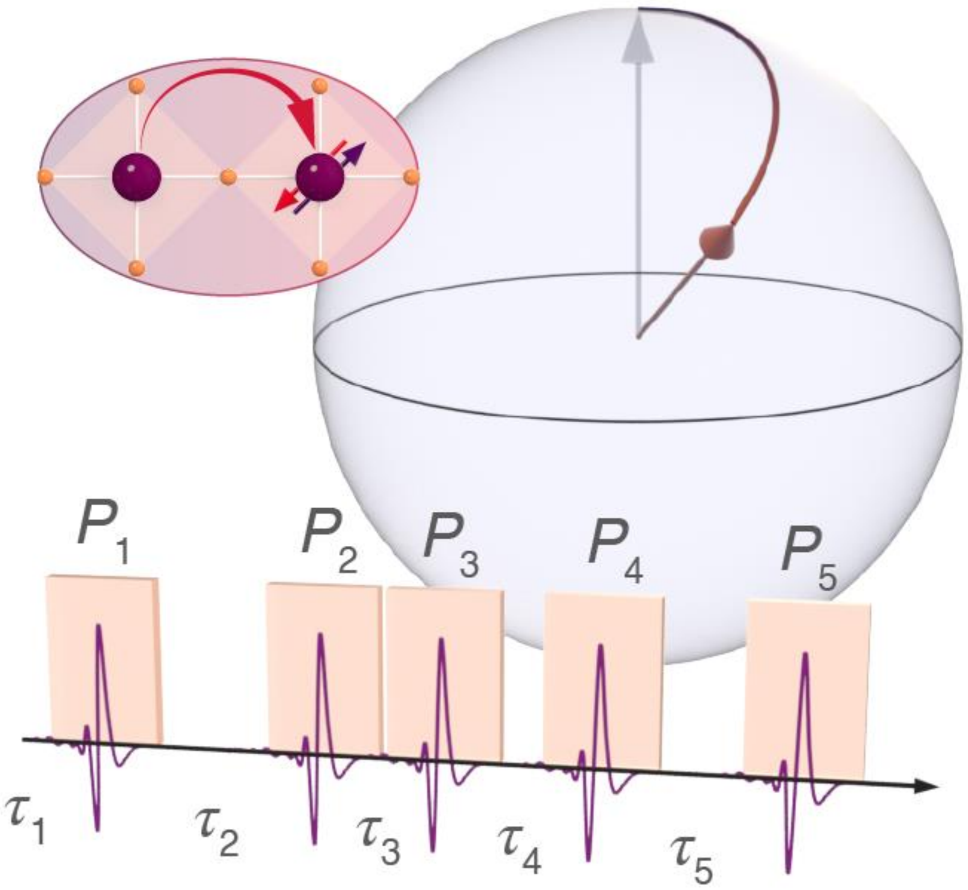
- Construction of average Floquet Hamiltonians via pulse sequences

$$H_{\mathrm{av}} = \frac{1}{T}\sum_{k=1}^{n} \tau_k \tilde{H}_k$$

- Need for pulse shaping techniques (4f-deformable mirrors and TWINS interferometers) to create sequences and reliable unitary gates

  F. Gucci et al. arXiv:2412.08318 (2024)

Towards "THz NMR-like" control of correlated quantum phases



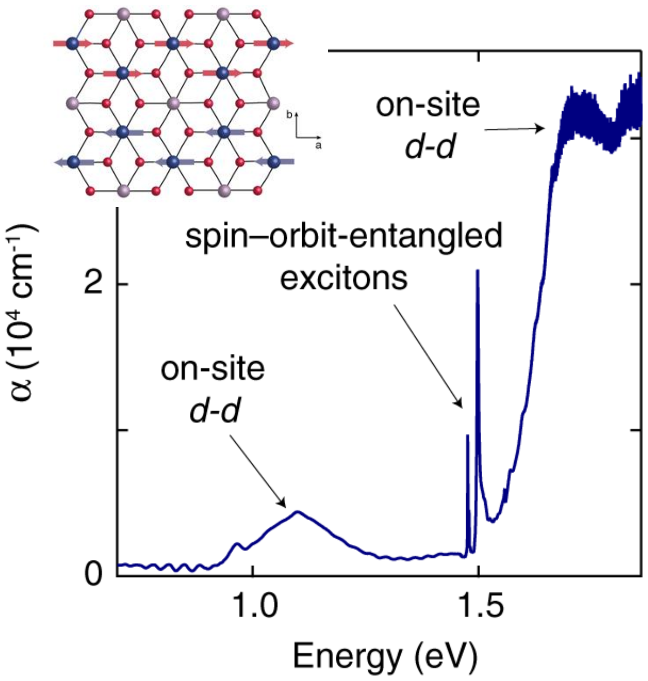
- **Synthesis of photoinduced coherent phases** (e.g., excitonic condensates, and η-pairing superconductivity)
- High-temperature **quantum sensing** with correlated excitons and polaritons
- High-repetition-rate, high-temperature **logical operations** across various excitonic platforms

Exciton-based quantum sensing in van der Waals materials

Strongly-correlated excitons can be functionalized as intrinsic and extrinsic quantum sensors

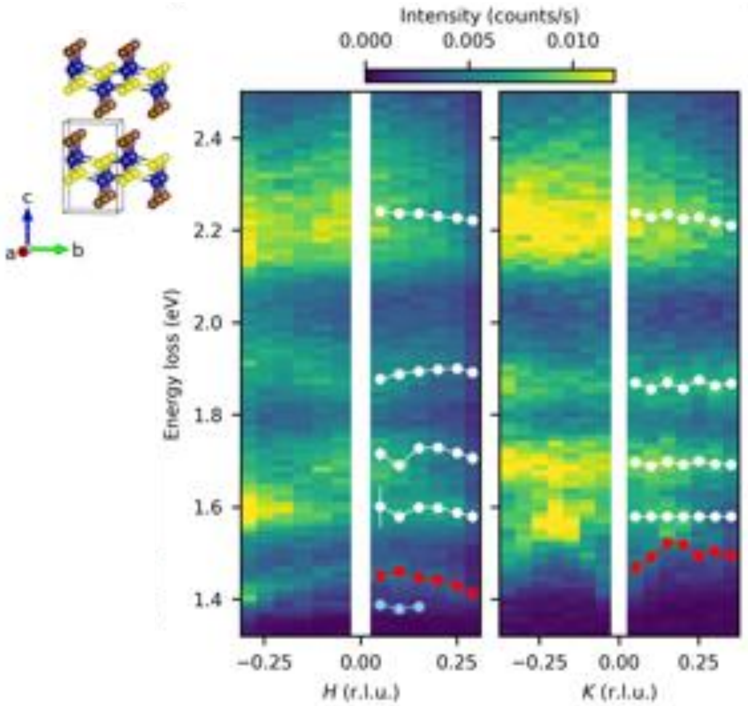
Xia et al. Nature Photonics 18, 344–349 (2024)

$NiPS_3$



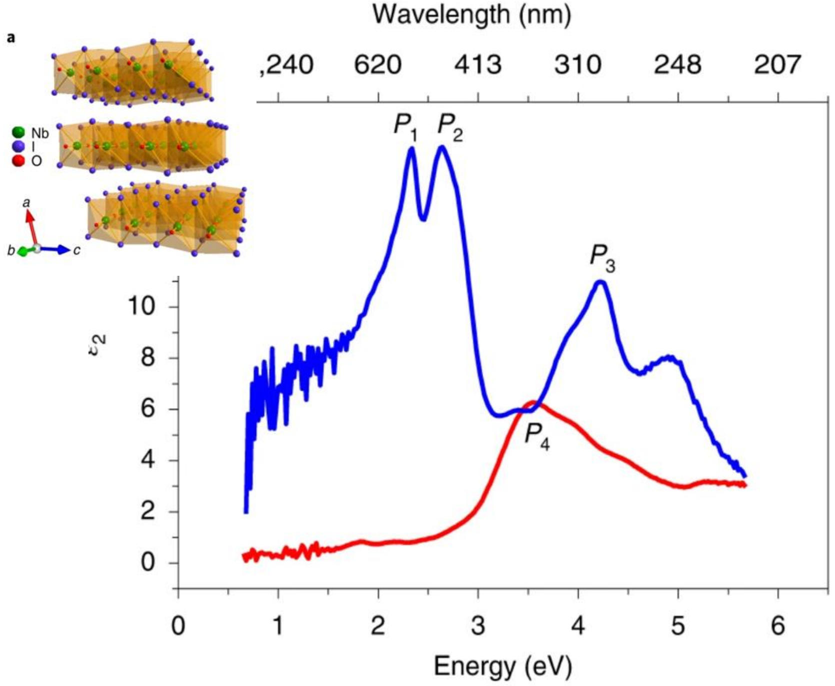
Kang et al. Nature **583**, 785–789 (2020)
Belvin et al. Nat Commun **12**, 4837 (2021)
He, MM et al. Nat Commun **15**, 3496 (2024)

CrSBr



Liebich et al. Nat. Mater. **24**, 384–390 (2025)
Shao et al. Nat. Mater. **24**, 391–398 (2025)
Sears, MM et al. under review (2025)

$NbOX_2$



Abdelwahab et al. Nat. Photon. **16**, 644 (2022)
Handa et al. Nat. Mater. (2025)
CY Huang Nat Commun **16**, 1896 (2025)

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mpsd — I-T. Lu, A. Rubio

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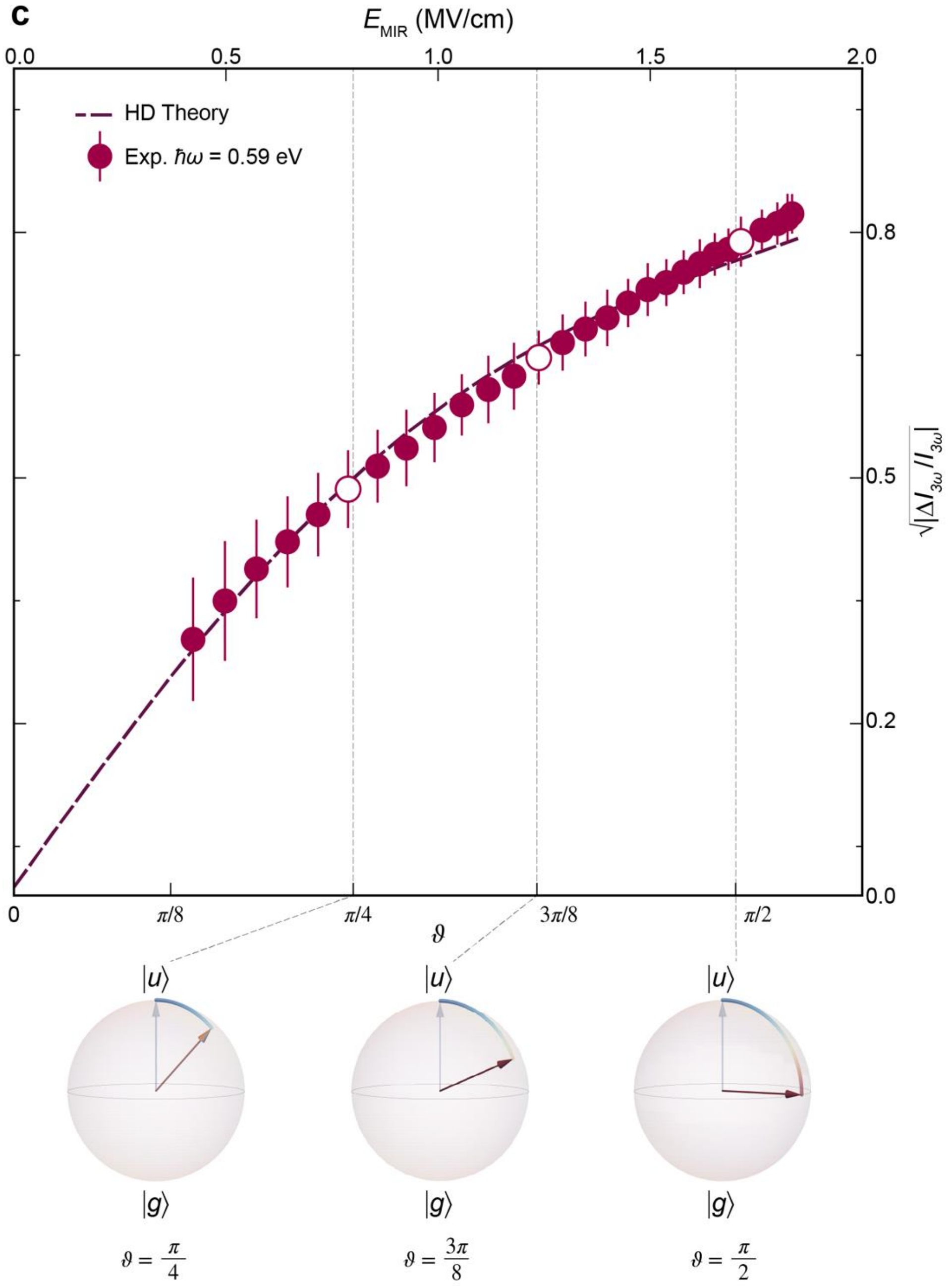
Summary



- Demonstration of quantum control of many-body wavefunction in a Mott insulator
- Realization of $\pi/2$ rotations of the many-body wavefunction for experimentally relevant fields
- Pathway for "optical NMR-like control" of correlated phases

D. R. Baykusheva, D. Carmichael, F. Glerean, T. Meng, P. B. De Oliveira, C. S. Weber, I.-T. Lu, C. C. Homes, I. A. Zaliznyak, G. D. Gu, M. P. M. Dean, A. Rubio, D. M. Kennes, M. Claassen & **M. Mitrano**, Nature Materials (2026).