

INTERNATIONAL CONFERENCE ON INELASTIC X-RAY SCATTERING,
STONY BROOK, NY
JUNE 28, 2019

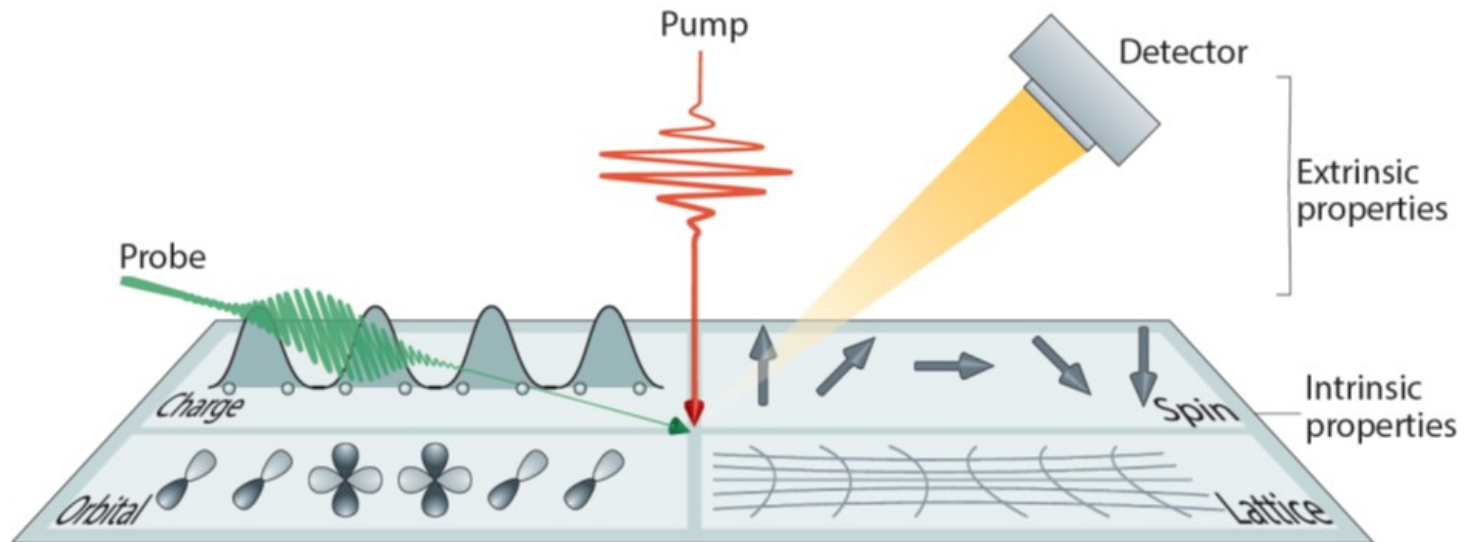
Tracking Collective Spin and Charge Excitations through Time-Resolved Raman and RIXS Spectra

Yao Wang

Department of Physics
Harvard University



Ultrafast Technique

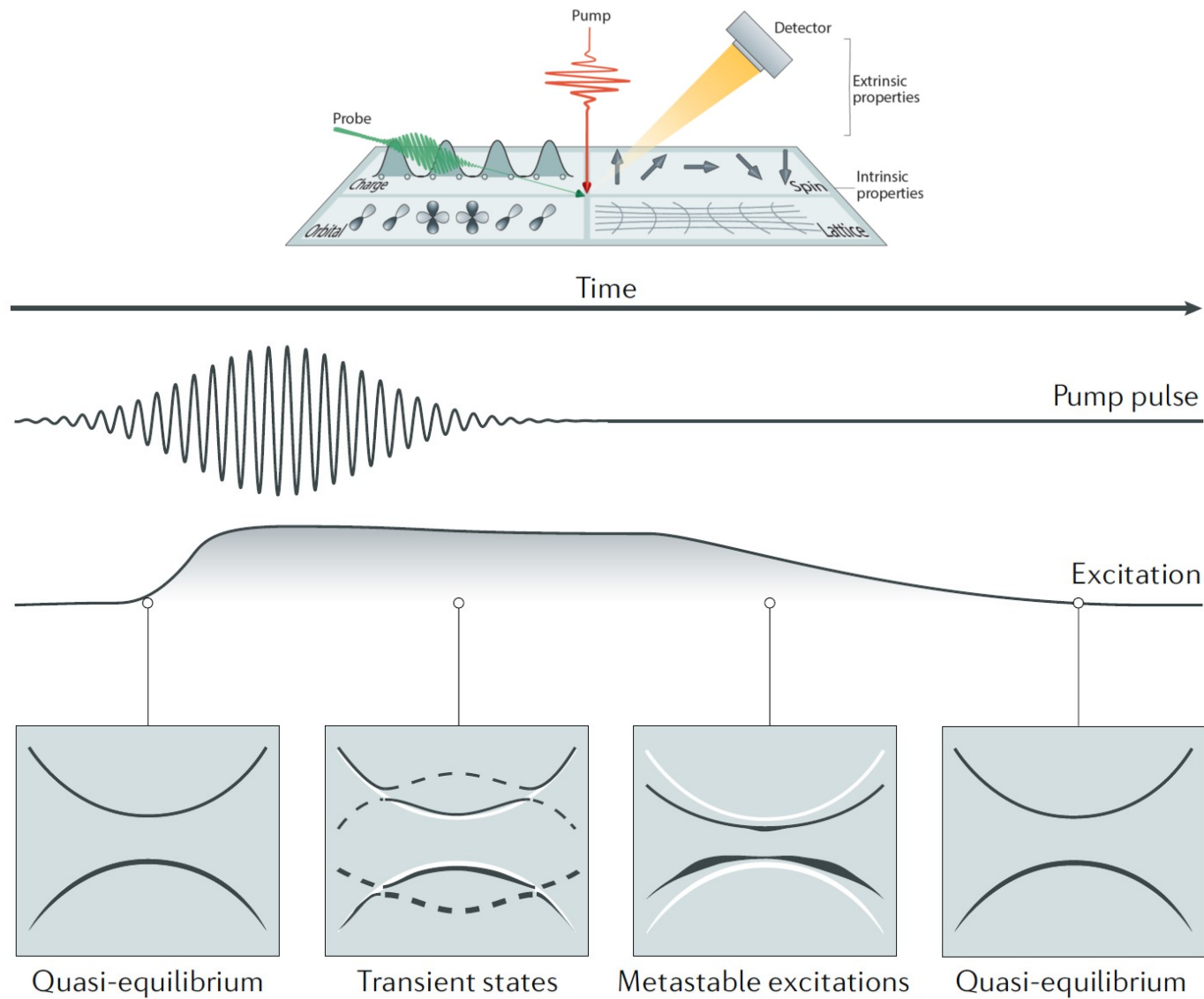


Wang *et al.* *Nat. Rev. Mater.* **3**, 312 (2018)



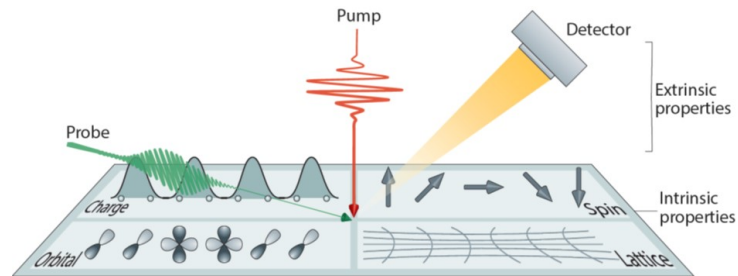
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Ultrafast Technique





Ultrafast Technique



time scale

quasi-equilibrium

during pump

after pump

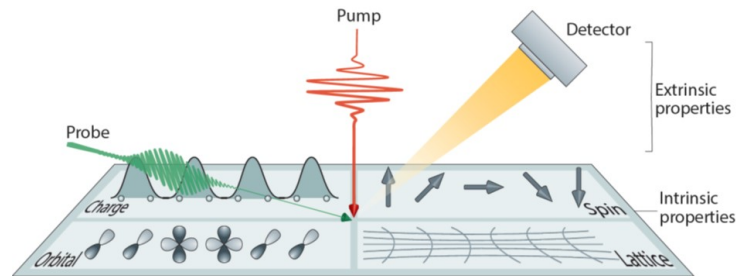
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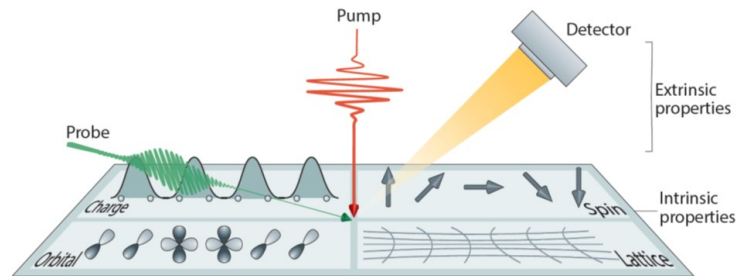


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time scale / purposes	quasi-equilibrium	during pump	after pump
control effective doping or temperature			
control single-particle band structure			
control effective interaction			

Ultrafast Technique



time scale purposes	quasi-equilibrium	during pump	after pump
control effective doping or temperature	 Yang <i>et al.</i>	 D'Alessio <i>et al.</i>	 Boschini <i>et al.</i>
control single- particle band structure	 Gerber <i>et al.</i>	 Wang <i>et al.</i>	 Schmitt <i>et al.</i>
control effective interaction	 De Conte <i>et al.</i>	 Mentink <i>et al.</i>	 Fausti <i>et al.</i>

Theory for Pump-Probe Spectroscopy

- **Pump and probe** $\mathcal{H}(t) = \mathcal{H}_{\text{pump}}(t) + \mathcal{H}_{\text{probe}}(t)$

- **Expansion of evolution operator**
non-perturbative perturbative

$$\begin{aligned} \mathcal{T} e^{-i \int_{-\infty}^t \mathcal{H}(\tau) d\tau} &\approx \mathcal{U}_0(t, -\infty) - i \int_{-\infty}^t \hat{\mathcal{H}}_{\text{probe}}(\tau) d\tau - i \int_{-\infty}^t dt_2 \int_{-\infty}^{t_2} dt_1 \hat{\mathcal{H}}_{\text{probe}}(t_2) \hat{\mathcal{H}}_{\text{probe}}(t_1) \\ &= \mathcal{U}_0(t, -\infty) - i \int_{-\infty}^t \hat{\mathcal{H}}_{\text{pr}}^{(\text{ab})}(\tau) d\tau - i \int_{-\infty}^t dt_2 \int_{-\infty}^{t_2} dt_1 \hat{\mathcal{H}}_{\text{pr}}^{(\text{ab})}(t_2) \hat{\mathcal{H}}_{\text{pr}}^{(\text{ab})}(t_1) \\ &\quad - i \int_{-\infty}^t dt_2 \int_{-\infty}^{t_2} dt_1 \hat{\mathcal{H}}_{\text{pr}}^{(\text{ab})\dagger}(t_2) \hat{\mathcal{H}}_{\text{pr}}^{(\text{ab})}(t_1) - i \int_{-\infty}^t \hat{\mathcal{H}}_{\text{pr}}^{(\text{ab}2)}(\tau) d\tau - i \int_{-\infty}^t \hat{\mathcal{H}}_{\text{pr}}^{(\text{sc})}(\tau) d\tau. \end{aligned}$$

$$\mathcal{H}_{\text{pr}}^{(\text{ab})}(t) = - \sum_{\alpha, \beta} \sum_{\mathbf{q} \varepsilon} \sum_{\mathbf{k} \sigma} M_{\alpha \beta}^{(1)}(\mathbf{q}, \mathbf{k}, t) c_{\mathbf{k}+\mathbf{q}, \sigma}^{(\alpha)\dagger} c_{\mathbf{k} \sigma}^{(\beta)} a_{\mathbf{q} \varepsilon}$$

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- **Semi-classical photon field**

$$|N\rangle_{\text{ph}} = e^{-N/2} e^{\sqrt{N} a_{\mathbf{q}_i \varepsilon_i}^\dagger} |0\rangle \quad \Rightarrow \quad g(\tau; t) = \sqrt{N(\tau)} = \frac{1}{\sqrt{2\pi\sigma_{\text{pr}}}} e^{-(\tau-t)^2/2\sigma_{\text{pr}}^2}$$

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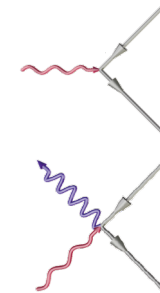
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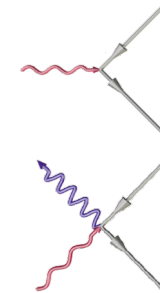
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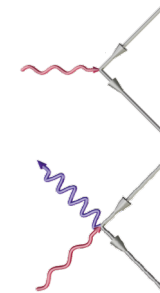
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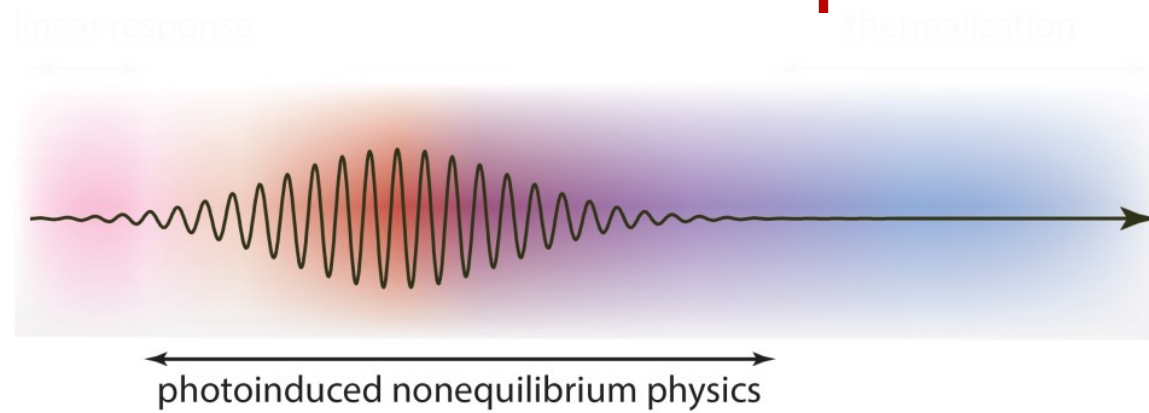
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Puzzle in a Realistic Pump Process

equilibrium
state



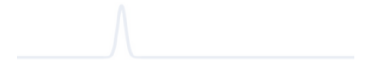
nonequilibrium
state

- Simple pictures

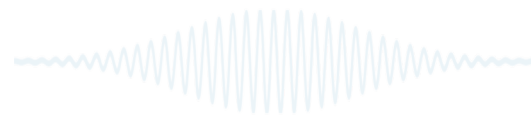
- Periodic drive



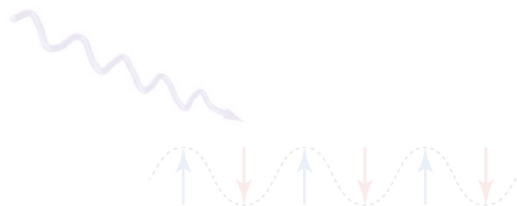
- instantaneous quench



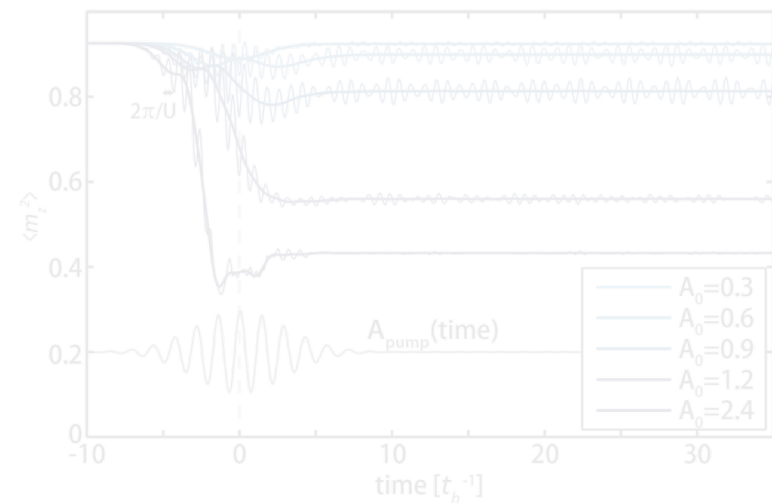
- Realistic pump



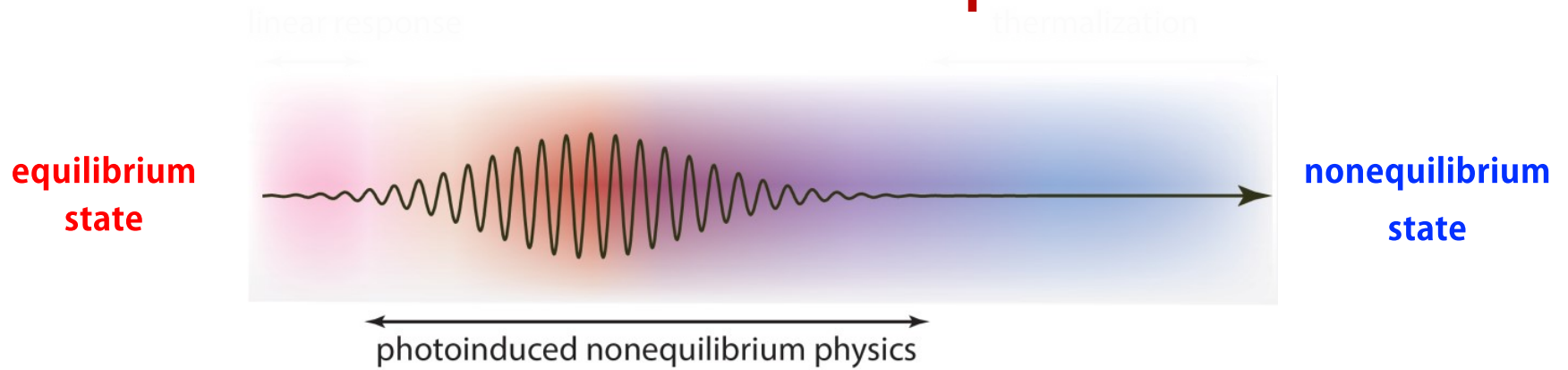
- Transient dynamics of a pumped Mott insulator



$$\mathcal{H} = -t_h \sum_{i,\sigma} \left(c_{i\sigma}^\dagger c_{i+1\sigma} + h.c. \right) + U \sum_i n_{i\uparrow} n_{i\downarrow}$$



Puzzle in a Realistic Pump Process

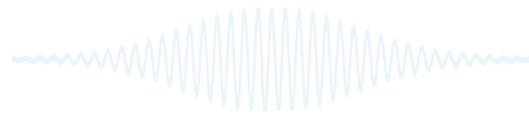


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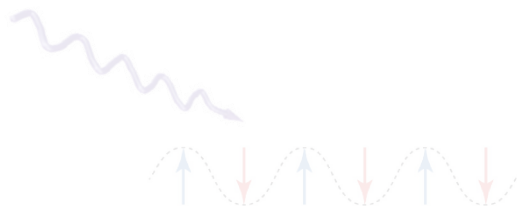
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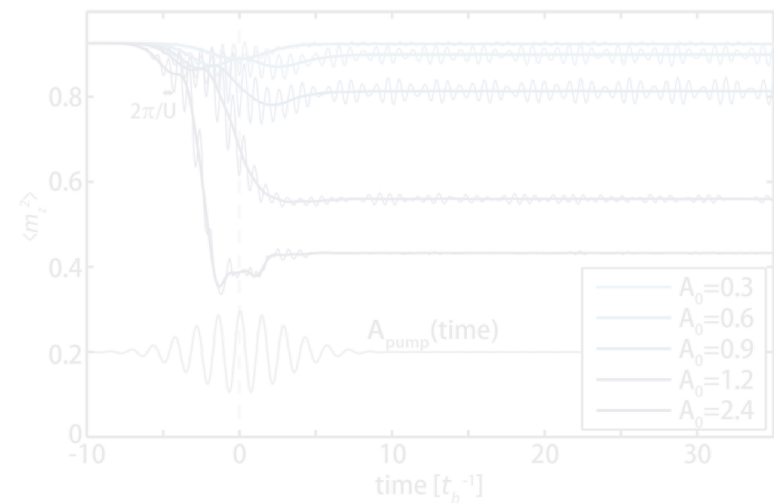
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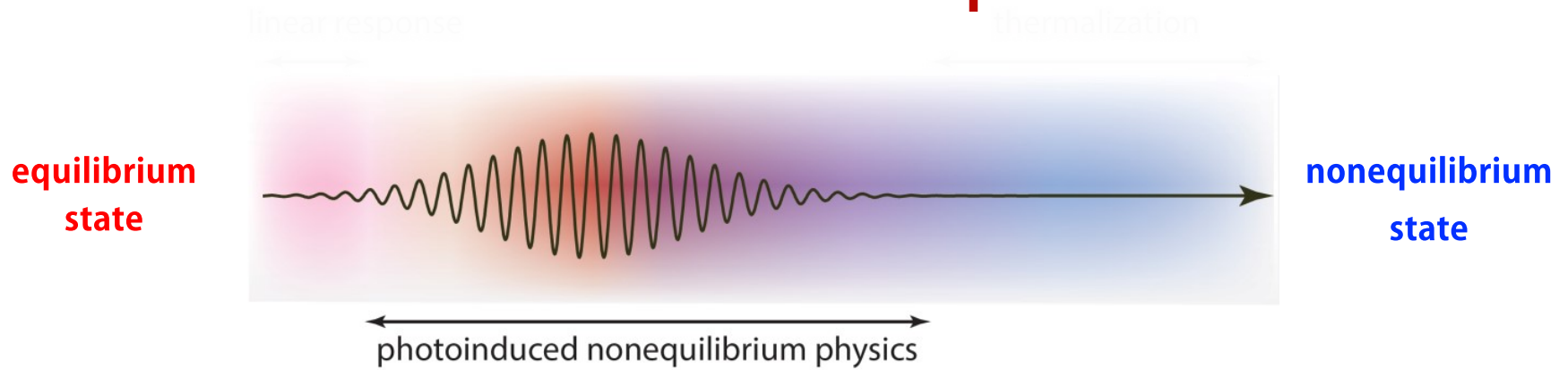
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Puzzle in a Realistic Pump Process



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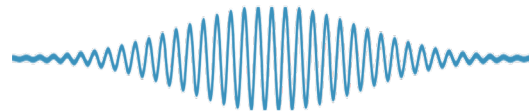
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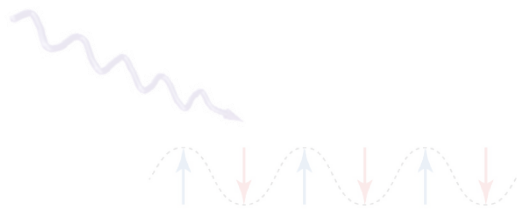
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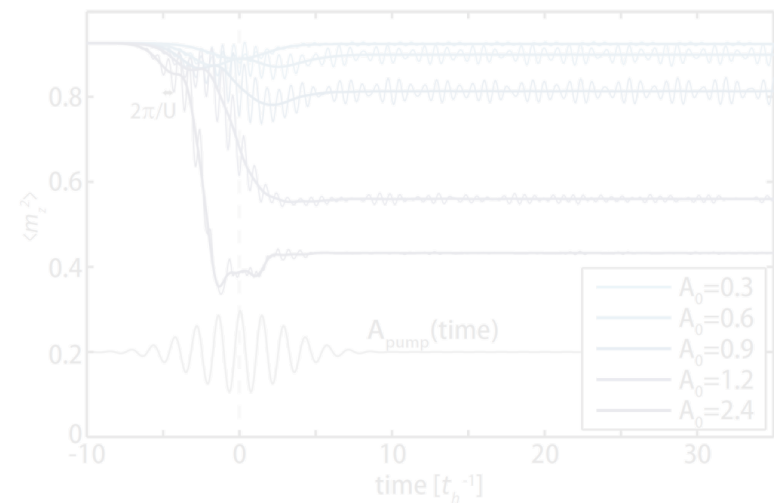
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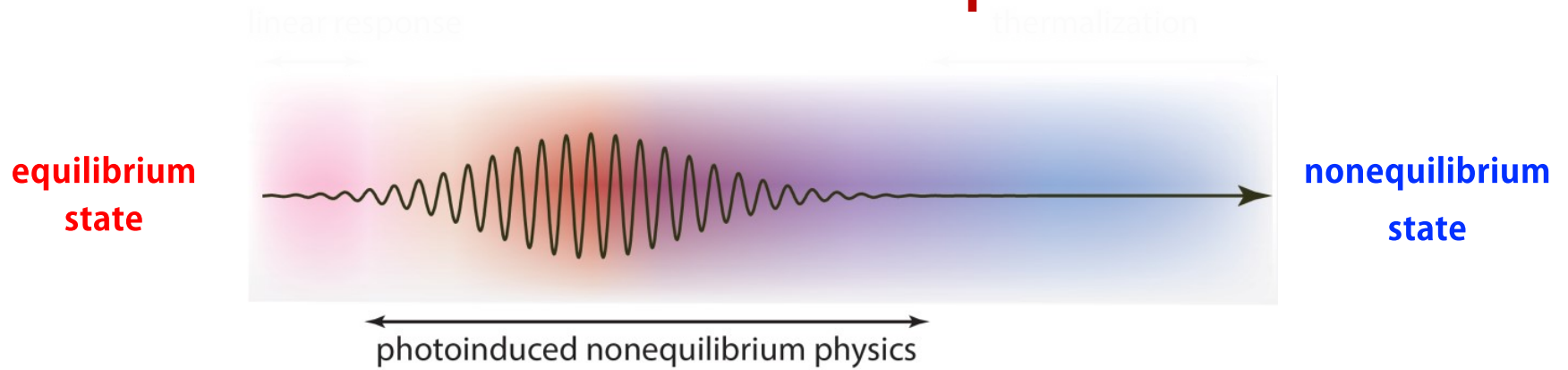
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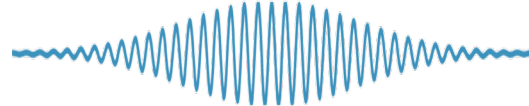


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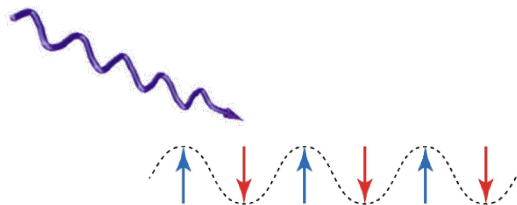
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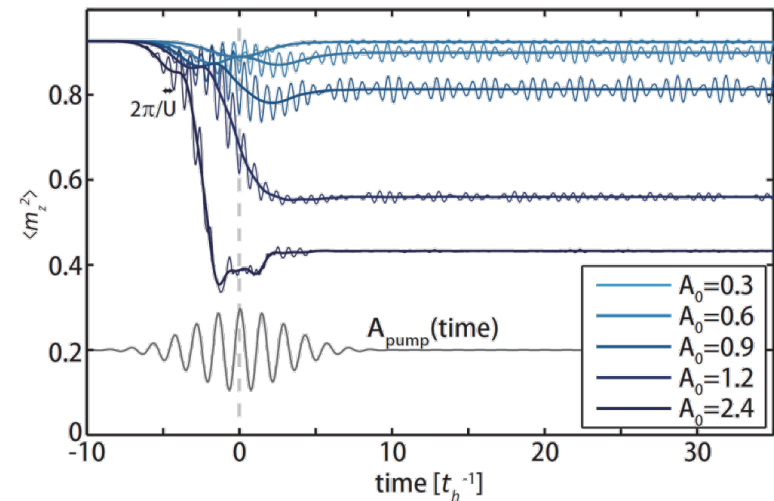
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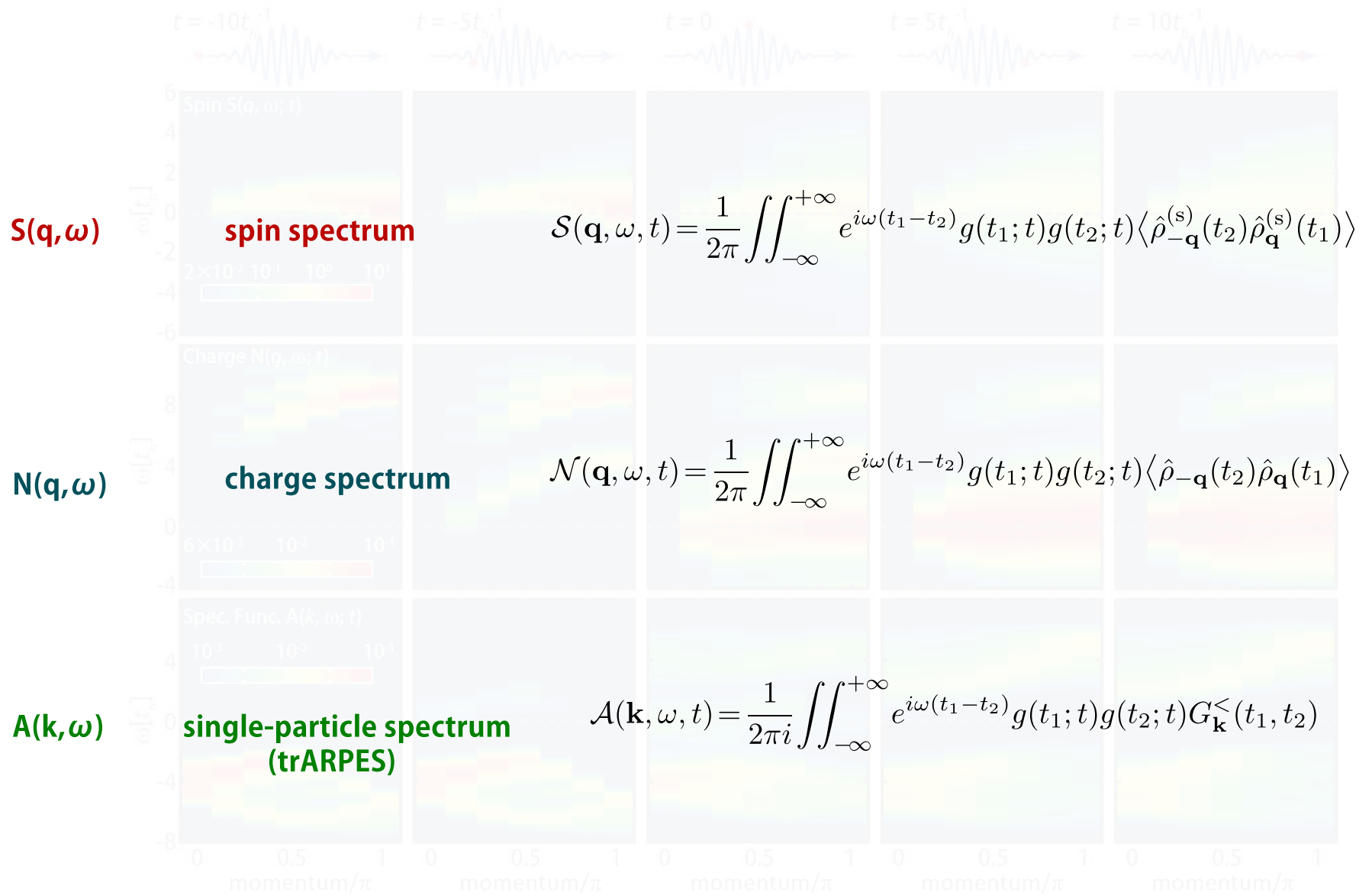


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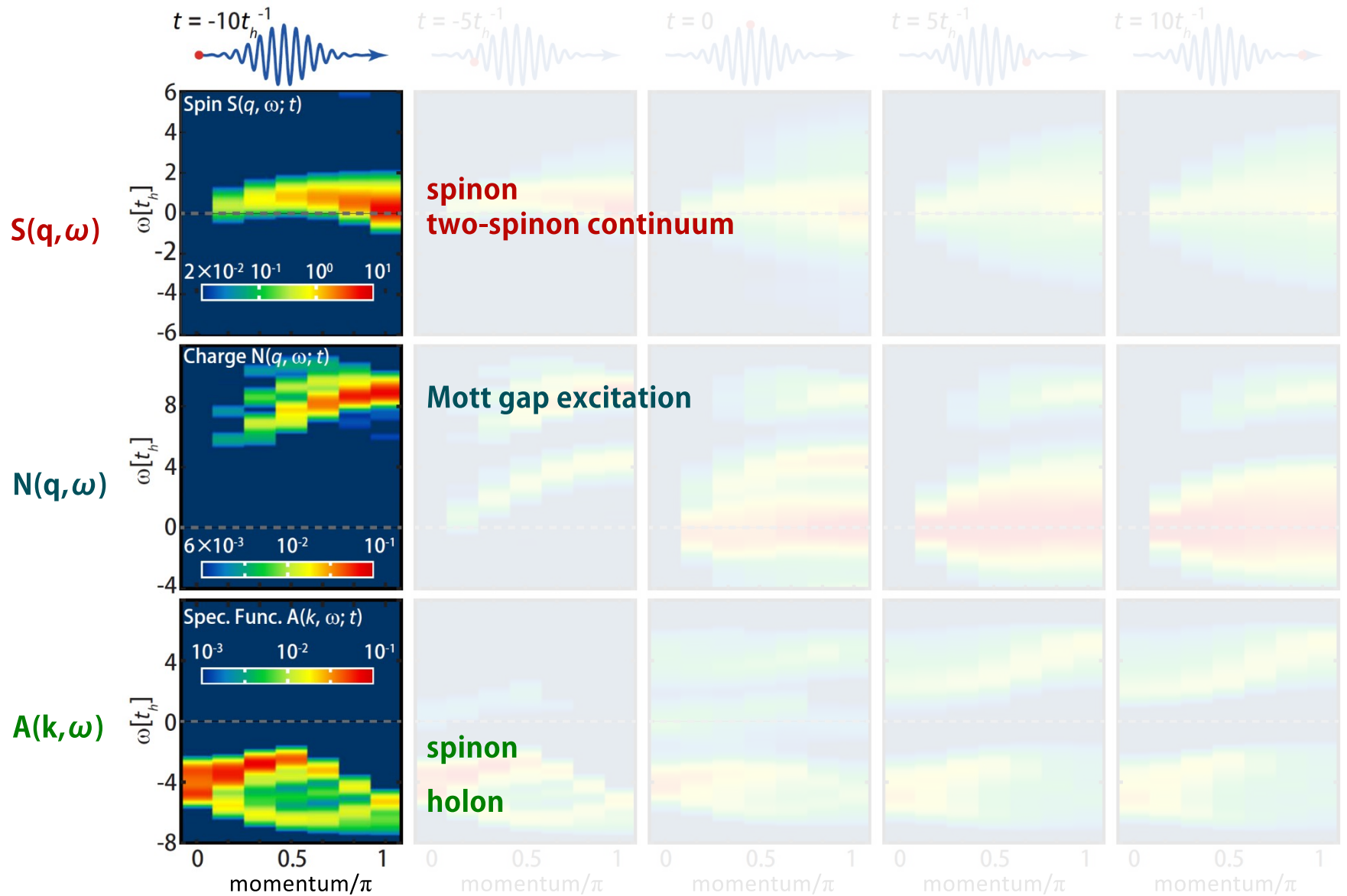




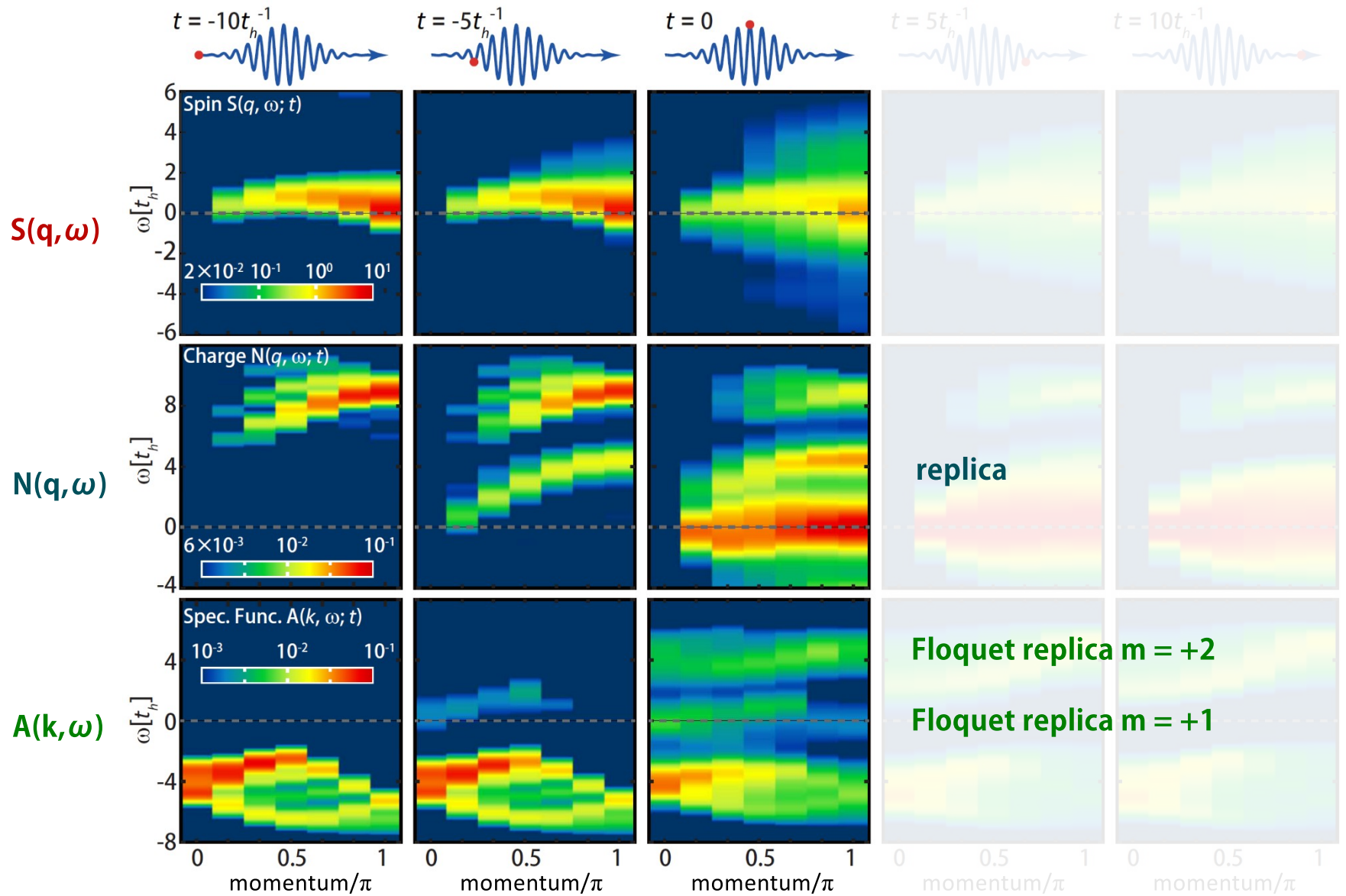
Transient Spin, Charge and Single-Particle Spectra



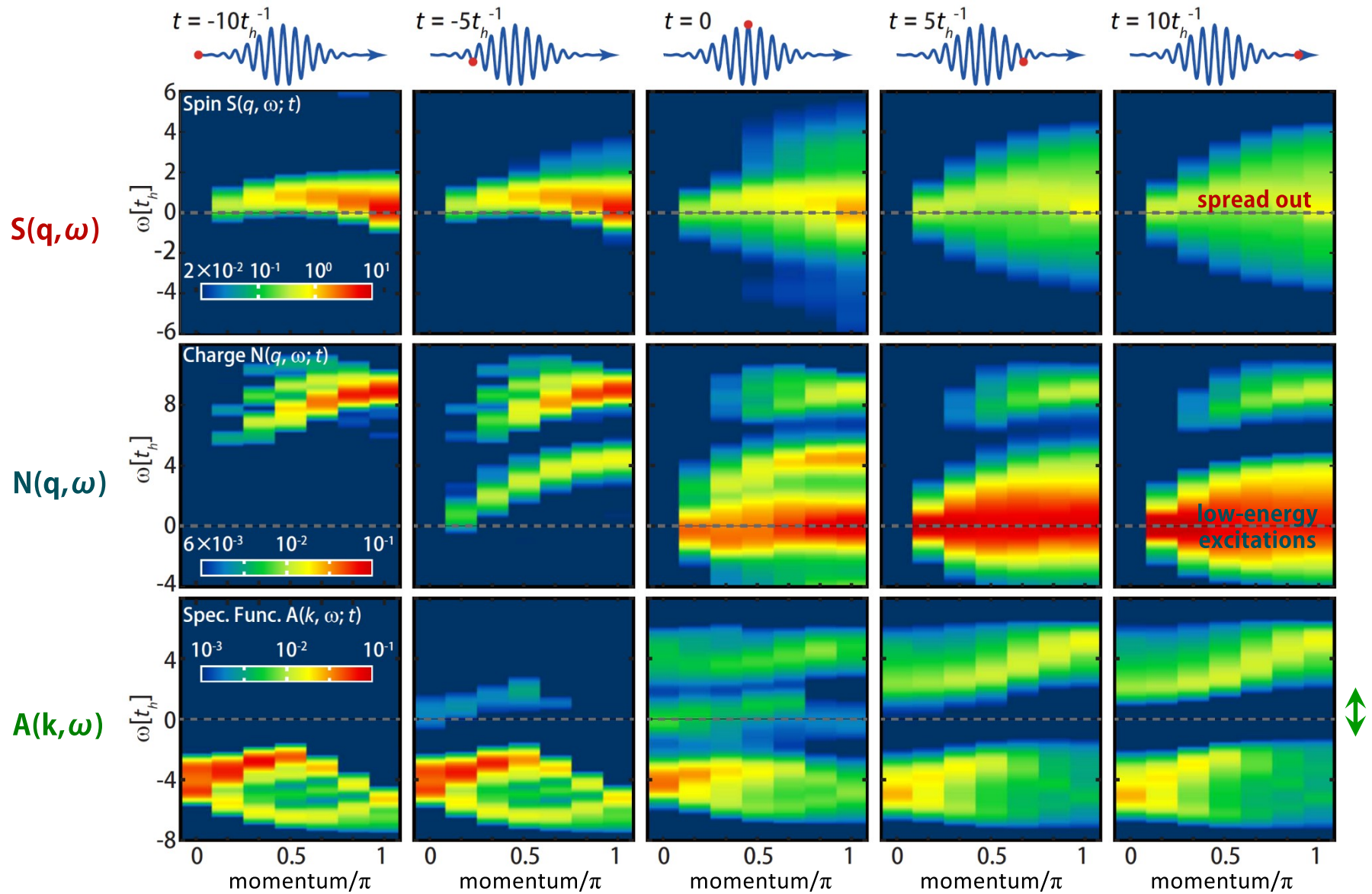
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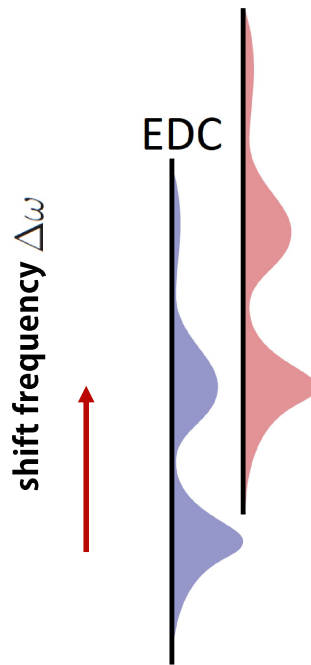




Resonance of Transient States and Remnant Excitations

- **Auto-correlation**

$$C(k, \Delta\omega; t) = \int_{-\infty}^{\infty} A(k, \omega; t) A(k, \omega + \Delta\omega; t) d\omega$$

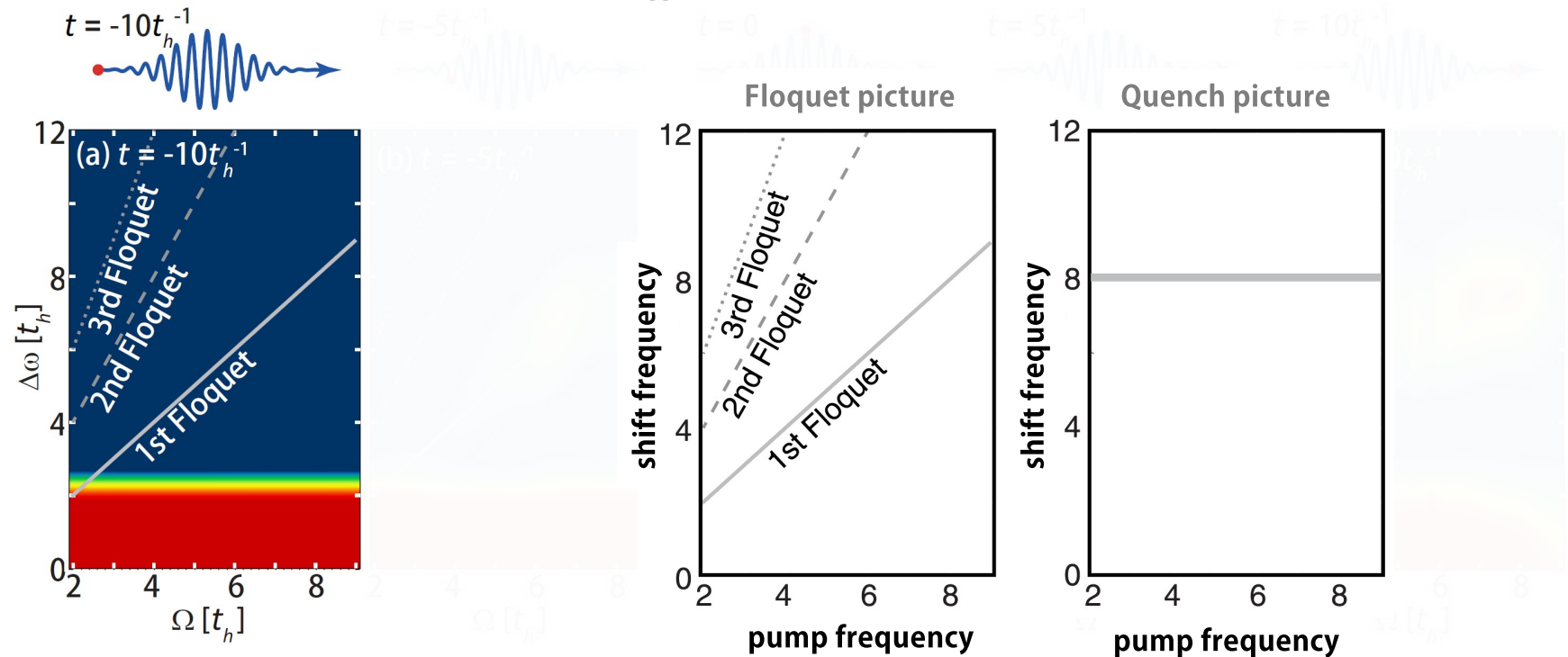




Resonance of Transient States and Remnant Excitations

- Auto-correlation for different pump frequencies

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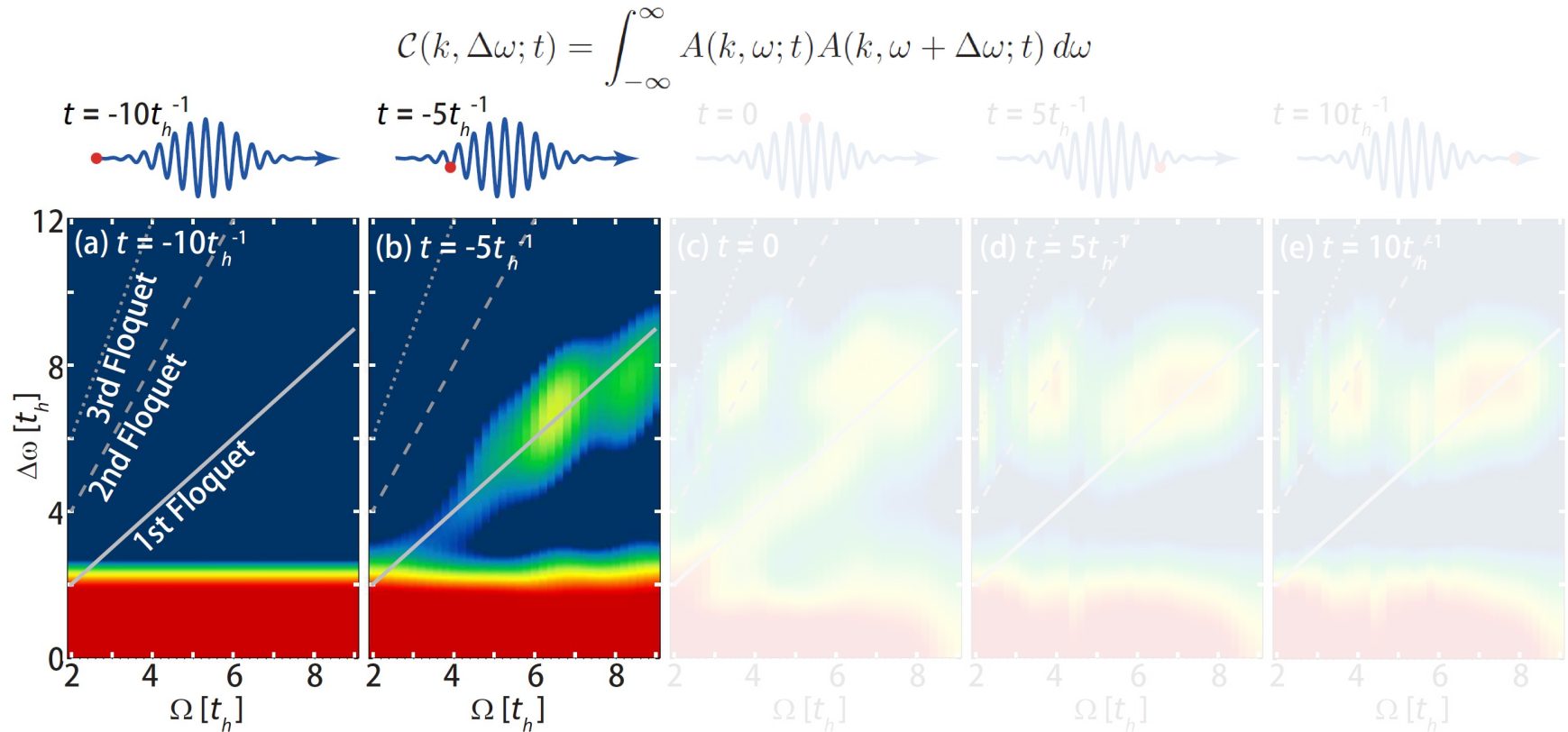
- Real excitations are developed through this resonance
- Post-pump excitations are coherently engineered





Resonance of Transient States and Remnant Excitations

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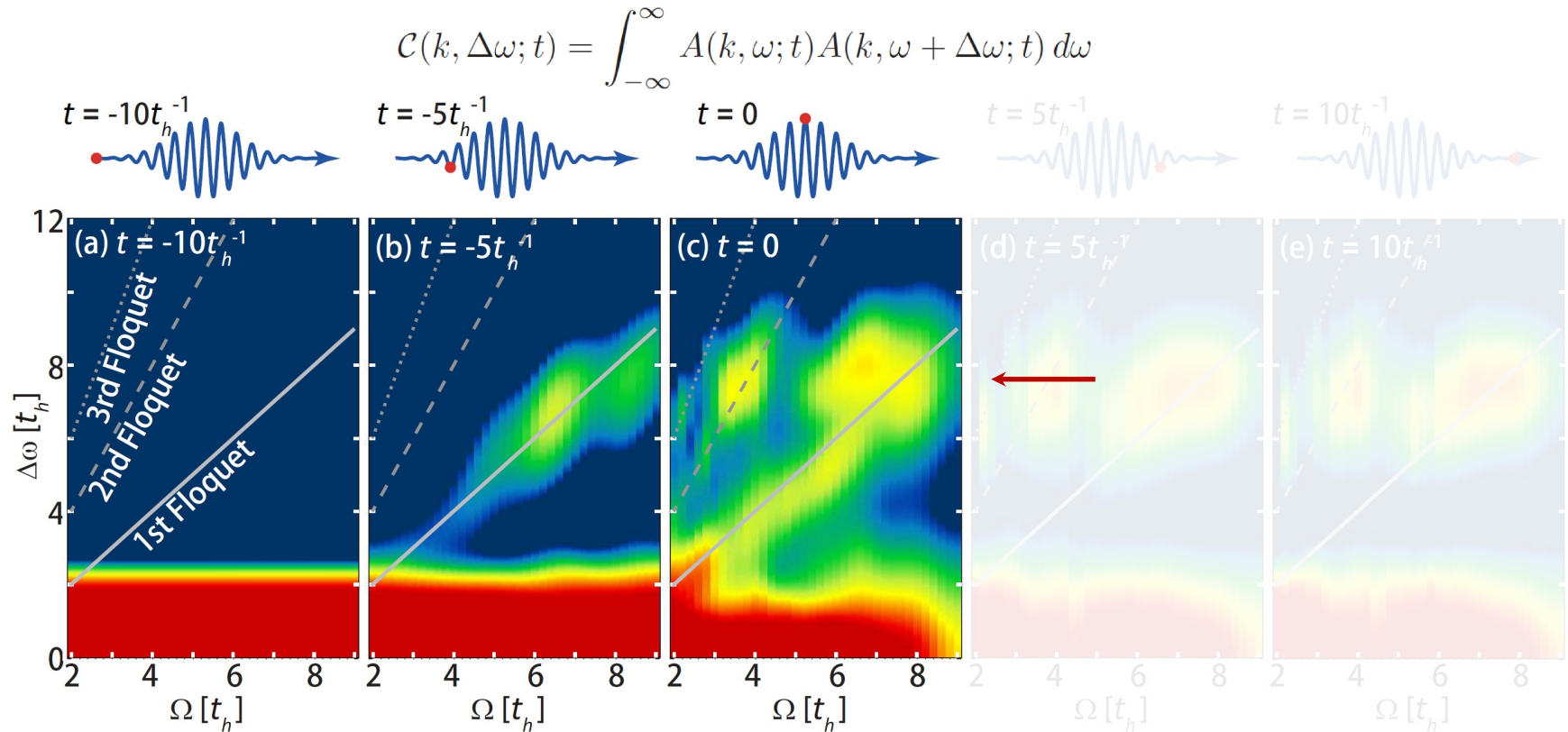
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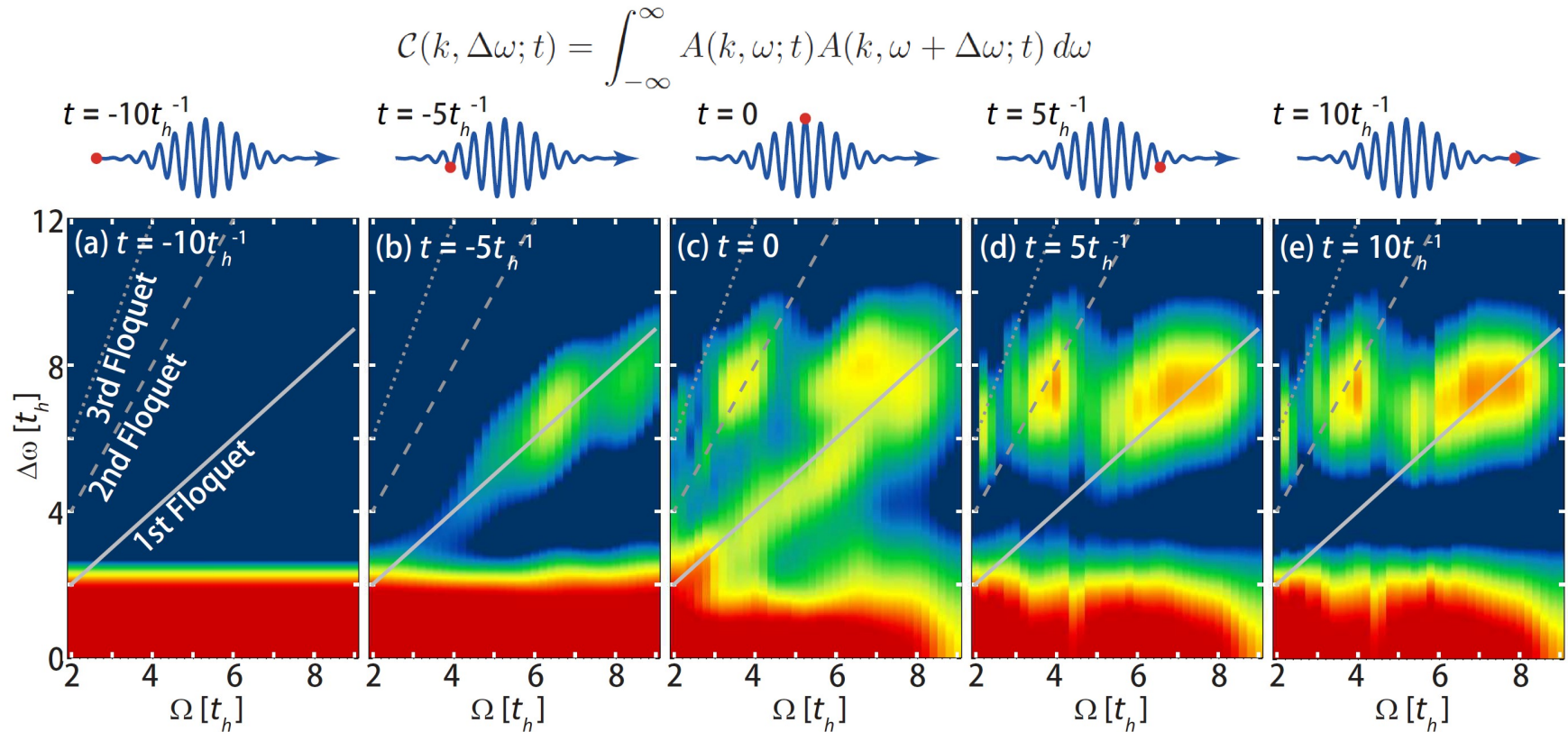
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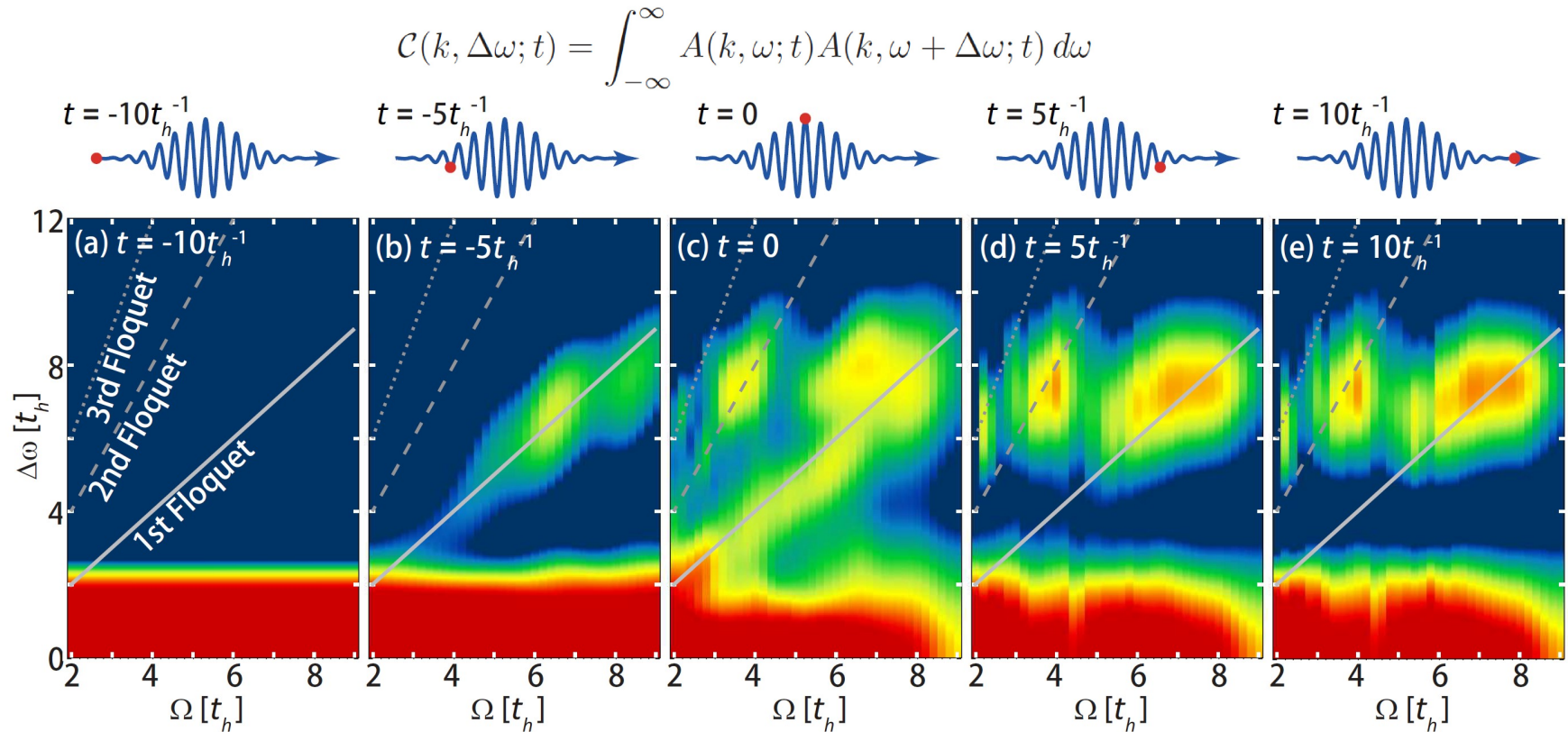
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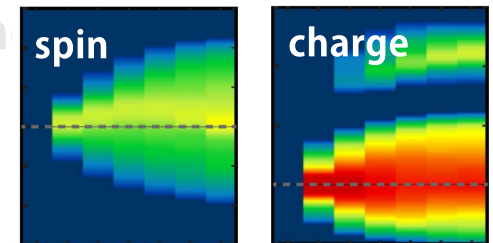


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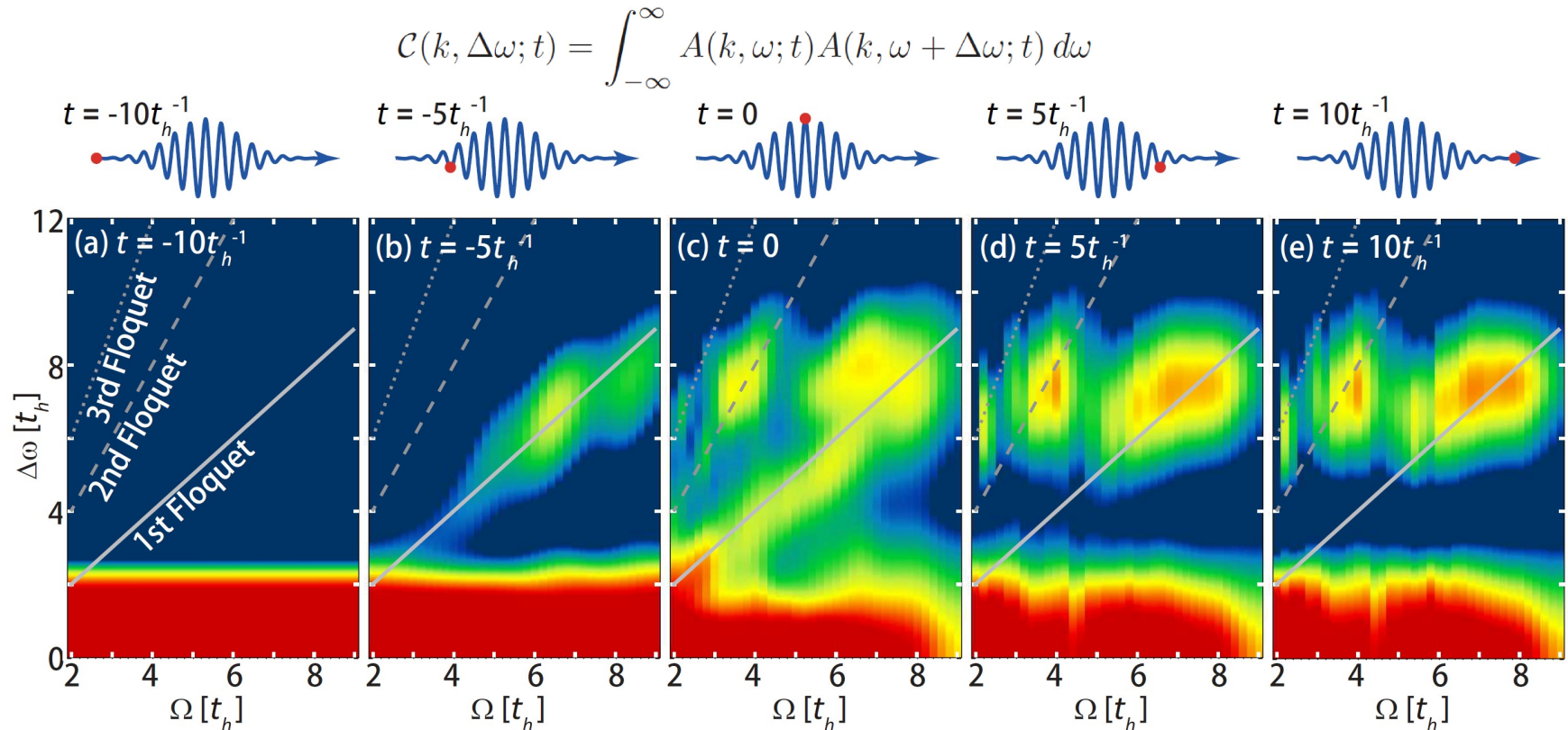
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- Post-pump excitations are coherently engineered





Resonance of Transient States and Remnant Excitations

- Auto-correlation for different pump frequencies



- Real excitations are developed through this **resonance**
- Post-pump excitations are **coherently engineered**

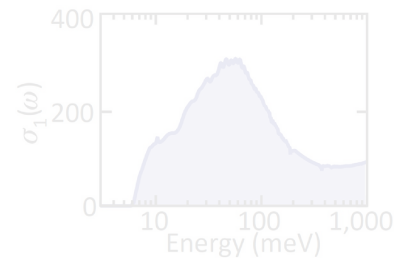


From Equilibrium to Nonequilibrium Spectrum

Conductivity

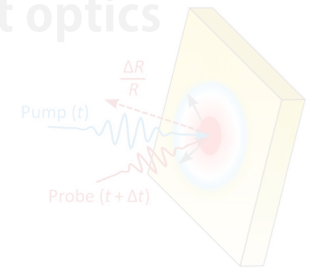
Equilibrium Spectrum

• Optics



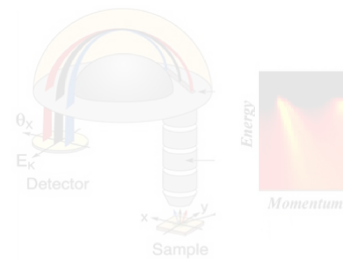
Pump-Probe Spectrum

• Ultrafast optics



Electronic structure

• ARPES

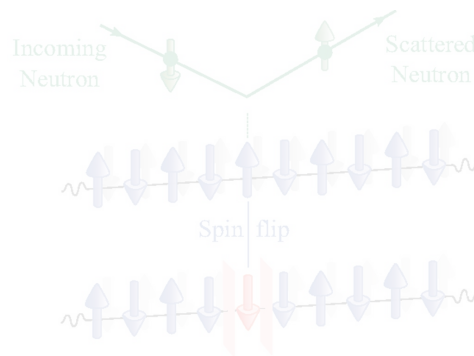


• Time-resolved ARPES



Dynamical spin structure factor

• Inelastic neutron scattering

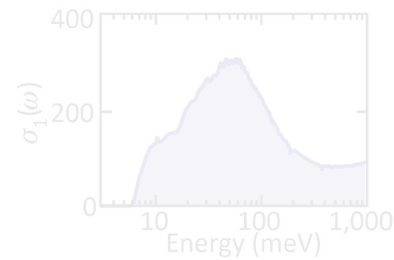


From Equilibrium to Nonequilibrium Spectrum

Conductivity

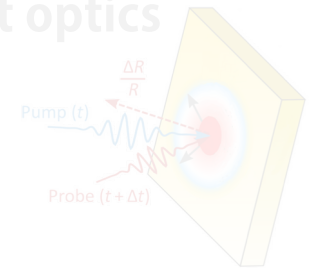
Equilibrium Spectrum

• Optics



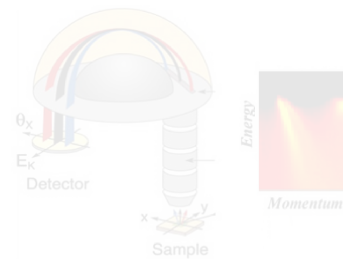
Pump-Probe Spectrum

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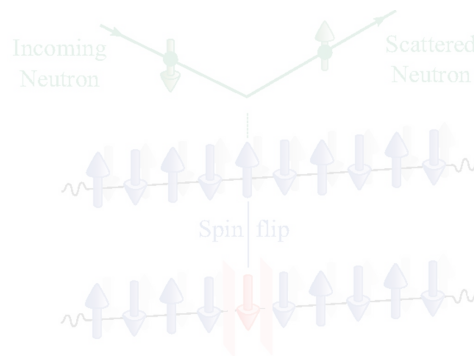


• Time-resolved ARPES



Dynamical spin structure factor

• Inelastic neutron scattering

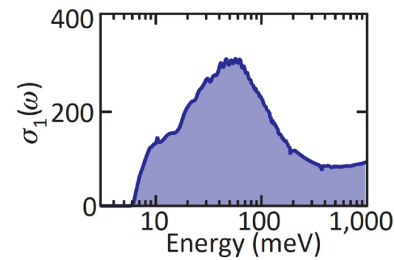


From Equilibrium to Nonequilibrium Spectrum

Conductivity

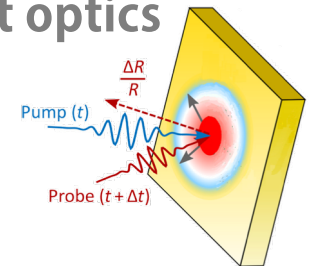
Equilibrium Spectrum

- Optics



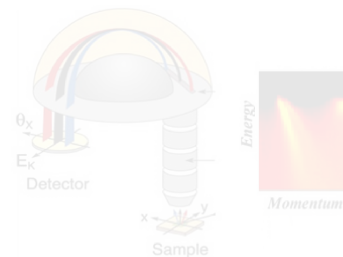
Pump-Probe Spectrum

- Ultrafast optics



Electronic structure

- ARPES

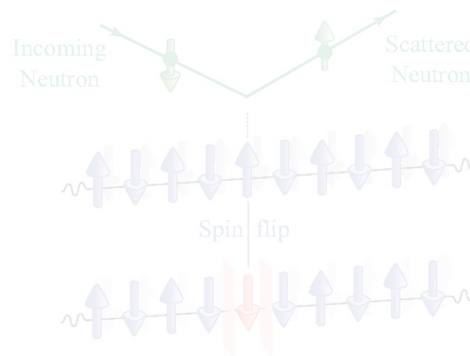


- Time-resolved ARPES



Dynamical spin structure factor

- Inelastic neutron scattering

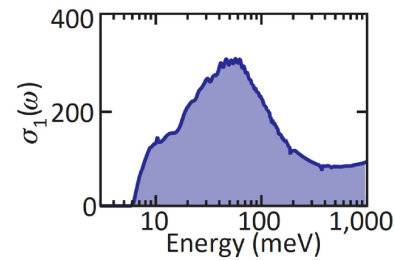


From Equilibrium to Nonequilibrium Spectrum

Conductivity

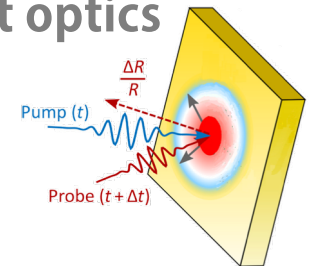
Equilibrium Spectrum

- Optics



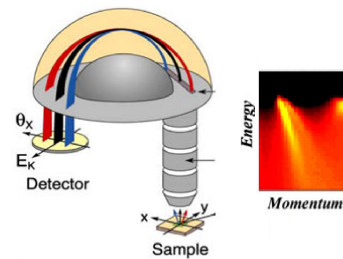
Pump-Probe Spectrum

- Ultrafast optics

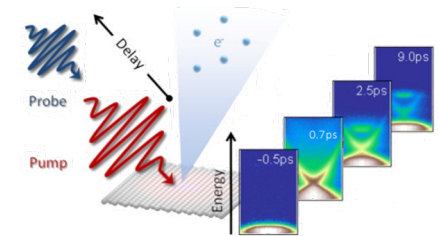


Electronic structure

- ARPES

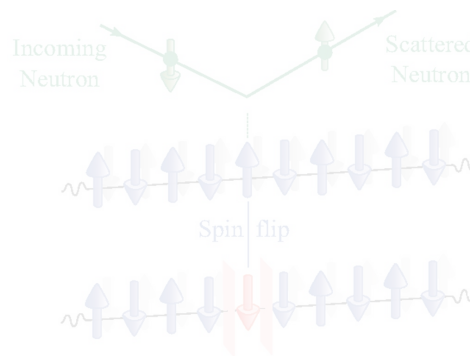


- Time-resolved ARPES



Dynamical spin structure factor

- Inelastic neutron scattering

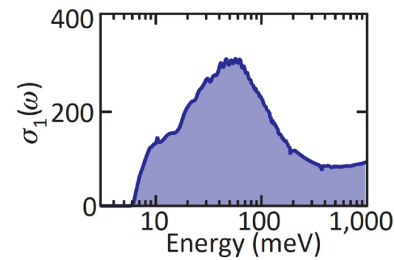


From Equilibrium to Nonequilibrium Spectrum

Conductivity

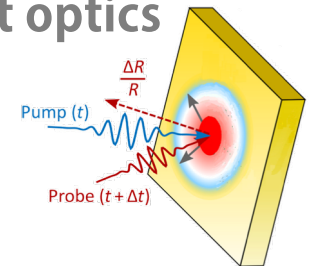
Equilibrium Spectrum

- Optics



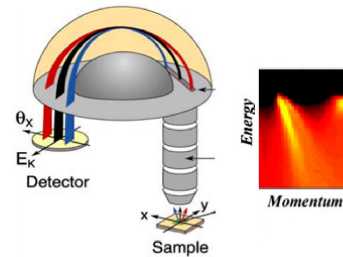
Pump-Probe Spectrum

- Ultrafast optics

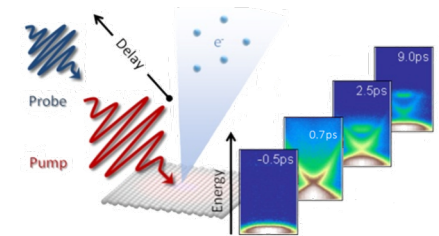


Electronic structure

- ARPES

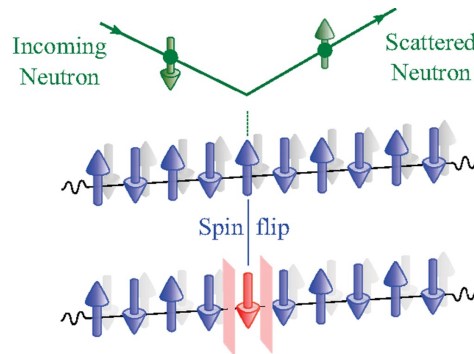


- Time-resolved ARPES



Dynamical spin structure factor

- Inelastic neutron scattering

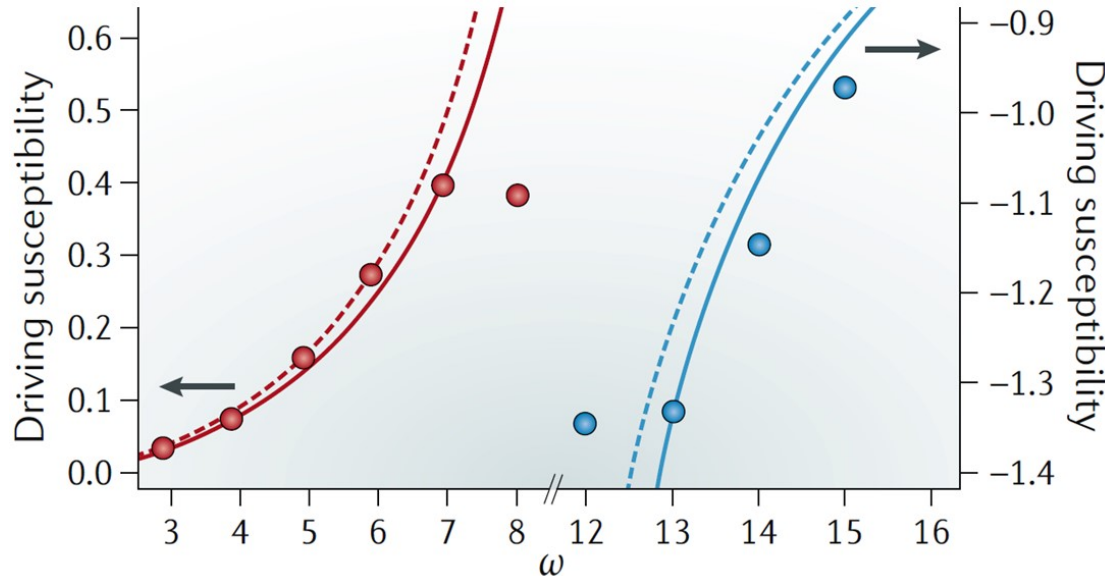


Ultrafast Spin and Charge Excitations

- Replica side bands: bridging quantum quench and Floquet states
- Parameter renormalization
- High-order correlations induced by symmetry breaking

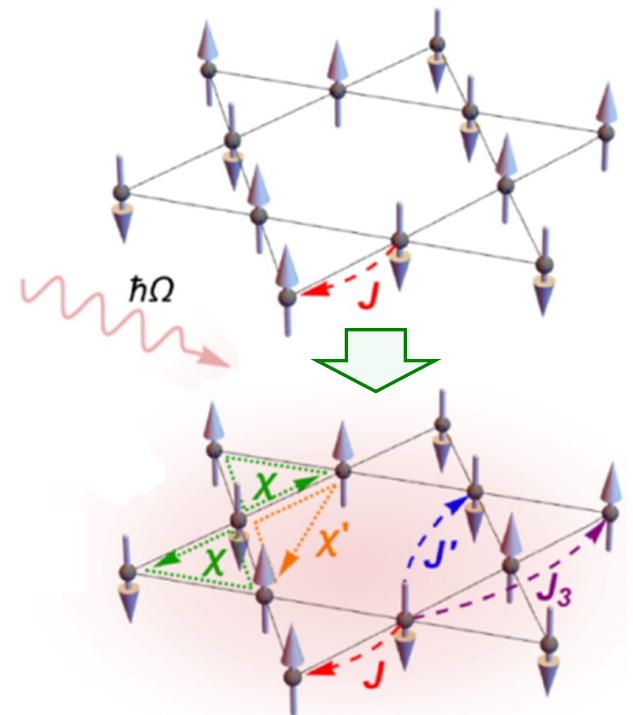
engineering spin interaction

$$\frac{J(A_0)}{J(A_0 = 0)} = \sum_{m=-\infty}^{+\infty} \frac{\mathcal{J}_{|m|}(A_0)^2}{1 + m\Omega/U}$$



Mentink *et al.* *Nat. Commun.* **6**, 6708 (2015)

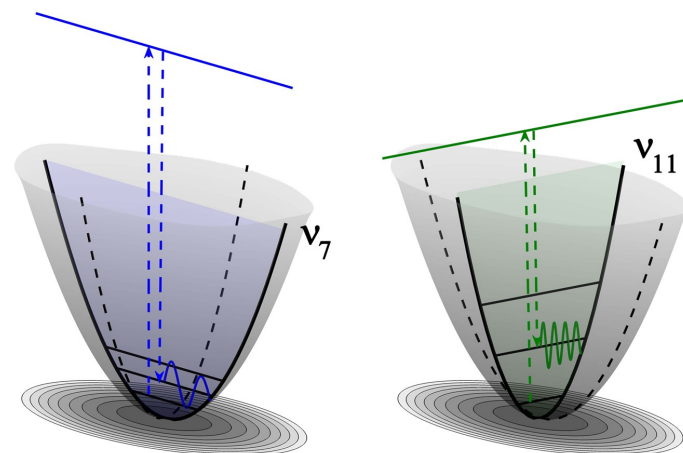
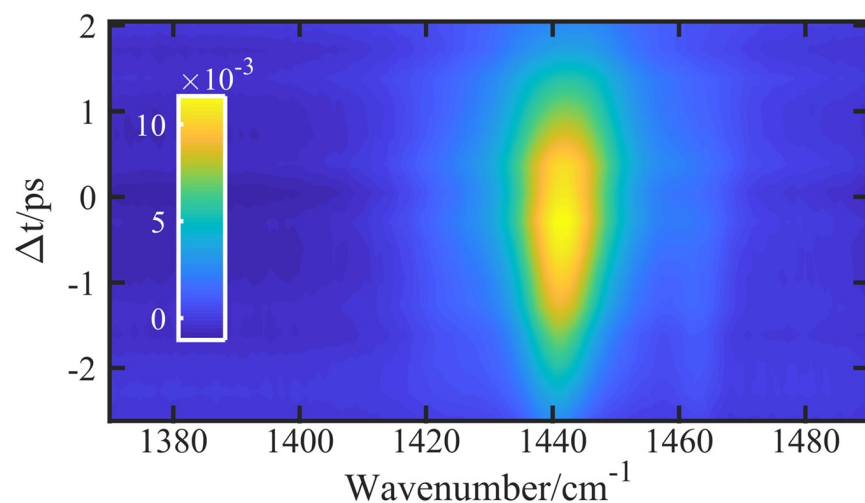
photo-induced chiral spin liquids



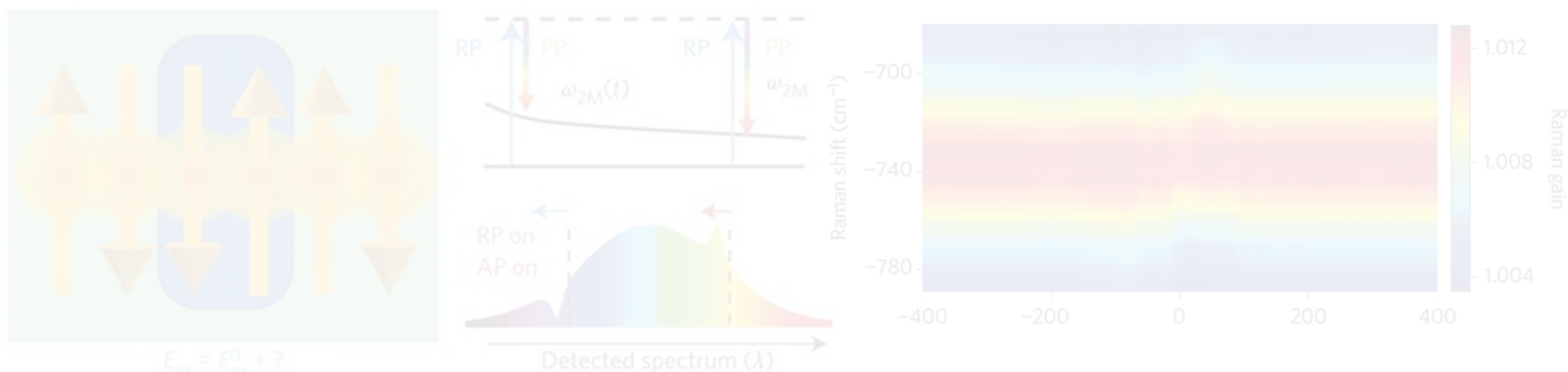
Claassen *et al.* *Nat. Commun.* **8**, 1192 (2017)

Time-Resolved Raman Scattering Technique

- Chemistry: tracking vibrational modes

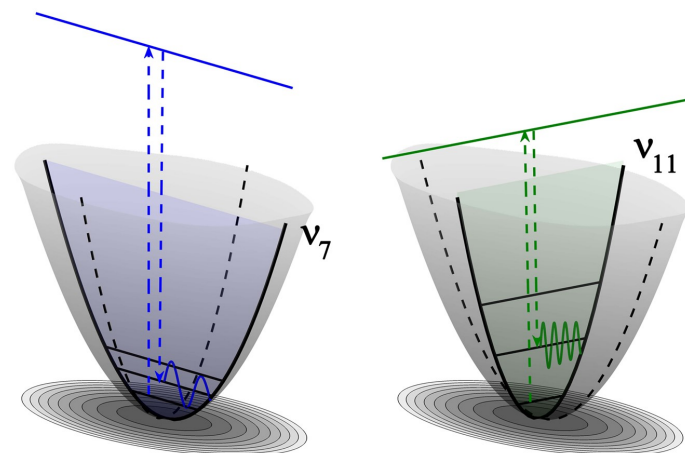
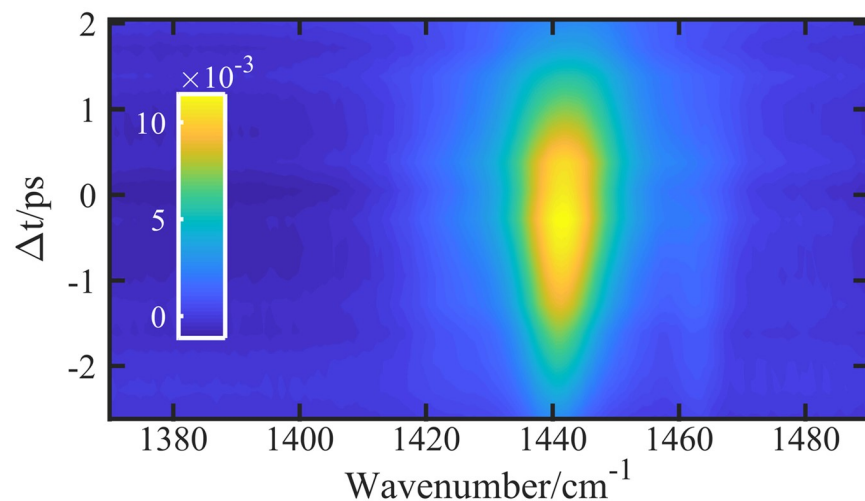


- Quantum materials: tracking collective excitations

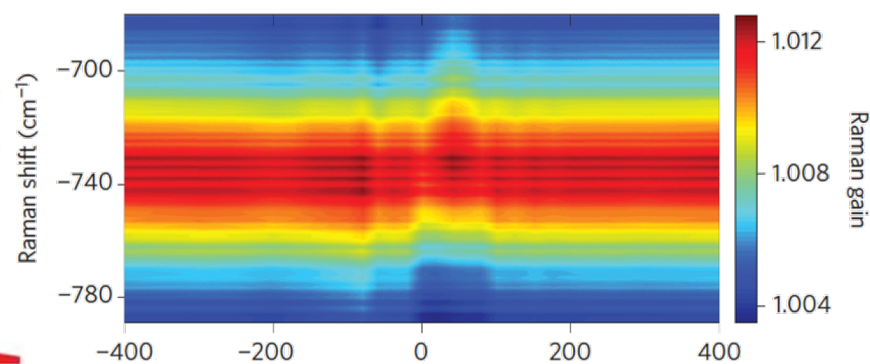
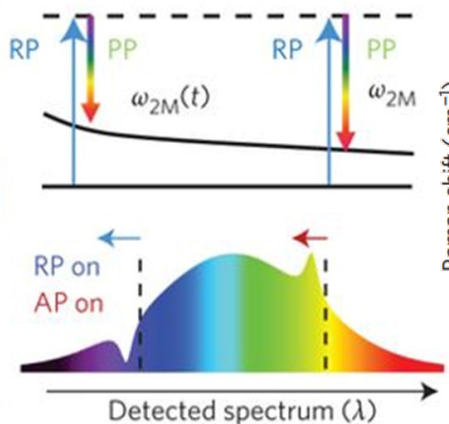
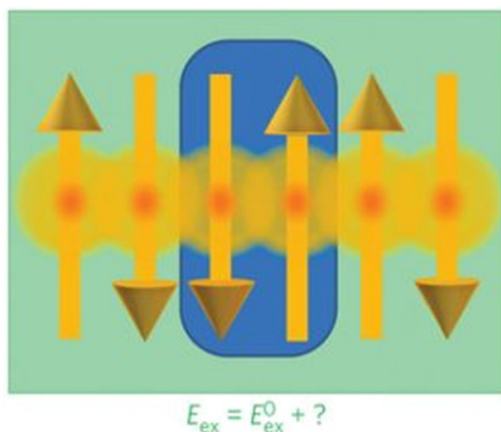


Time-Resolved Raman Scattering Technique

- Chemistry: tracking vibrational modes



- Quantum materials: tracking collective excitations





Time-Resolved Raman Scattering: Tracking Spin Exchange

- Time-resolved (non-resonant) Raman cross-section

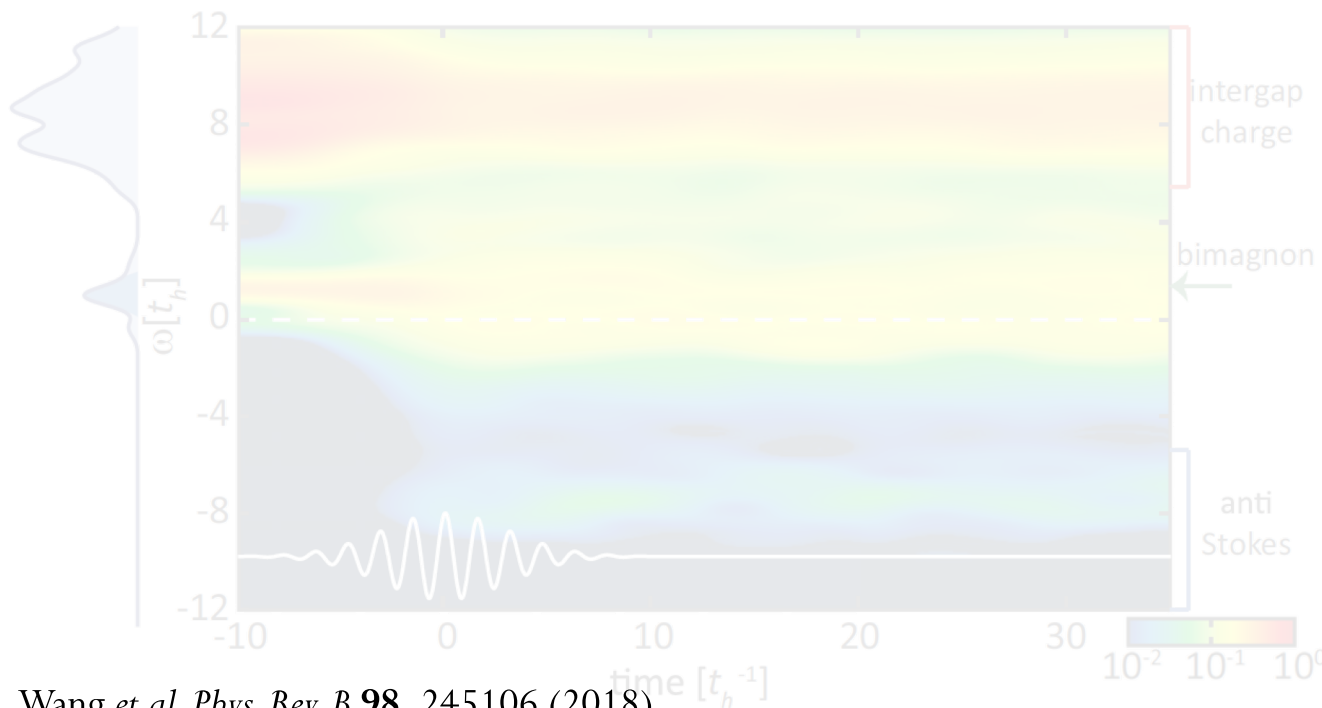
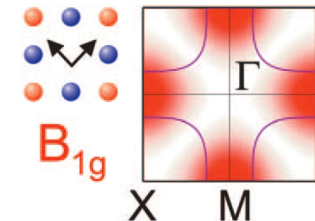
$$\mathcal{R}_{\varepsilon_s \varepsilon_i}(\omega, t) = \iint_{-\infty}^{\infty} dt_1 dt_2 e^{i\omega(t_2 - t_1)} g(t_1; t) g(t_2; t) \chi_{\varepsilon_s \varepsilon_i}(t_1, t_2)$$

$$\chi_{\varepsilon_s \varepsilon_i}(t_1, t_2) = i \langle \psi(t_2) | \hat{\gamma}_{\varepsilon_s \varepsilon_i}^\dagger(t_2) \mathcal{U}_0(t_2, t_1) \hat{\gamma}_{\varepsilon_s \varepsilon_i}(t_1) | \psi(t_1) \rangle$$

probe shape function $g(\tau; t)$

scattering vertex

$$\hat{\gamma}_{B_{1g}}(t) = \sum_{\mathbf{k}} \left[\frac{\partial^2}{\partial k_x^2} - \frac{\partial^2}{\partial k_y^2} \right] \varepsilon_{\mathbf{k} - \mathbf{A}(t)} n_{\mathbf{k}}$$



bimagnon: double spin flip
 $\sim 3J$ (spin exchange)



Wang *et al.* *Phys. Rev. B* **98**, 245106 (2018)

Shvaika *et al.*, *Cond. Mat. Phys.* **21**, 33707 (2018)



HARVARD
UNIVERSITY

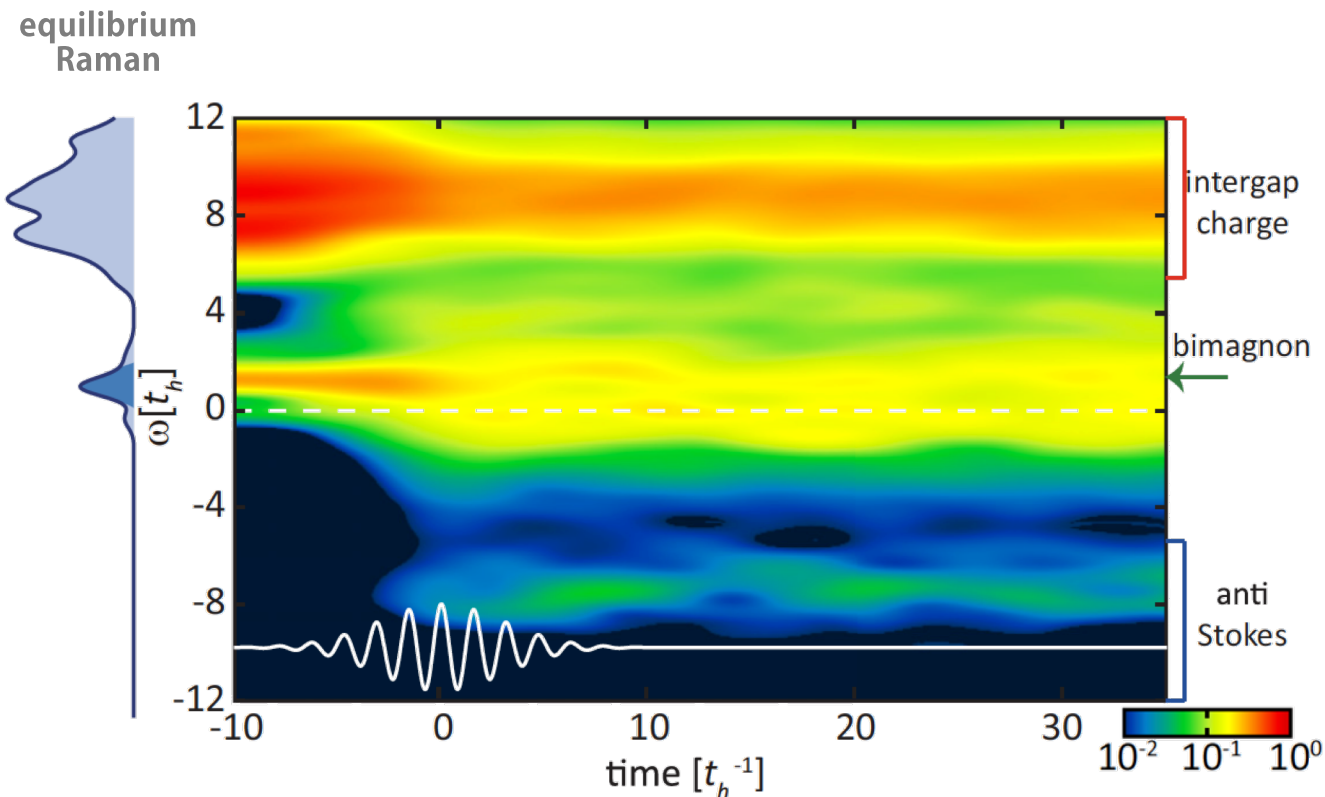
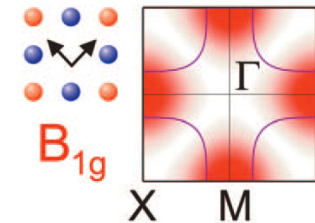


Time-Resolved Raman Scattering: Tracking Spin Exchange

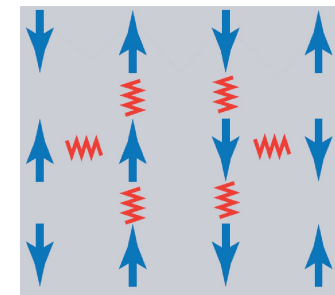
- Time-resolved (non-resonant) Raman cross-section

$$\mathcal{R}_{\varepsilon_s \varepsilon_i}(\omega, t) = \iint_{-\infty}^{\infty} dt_1 dt_2 e^{i\omega(t_2 - t_1)} g(t_1; t) g(t_2; t) \chi_{\varepsilon_s \varepsilon_i}(t_1, t_2)$$

$$\chi_{\varepsilon_s \varepsilon_i}(t_1, t_2) = i \langle \psi(t_2) | \hat{\gamma}_{\varepsilon_s \varepsilon_i}^\dagger(t_2) \mathcal{U}_0(t_2, t_1) \hat{\gamma}_{\varepsilon_s \varepsilon_i}(t_1) | \psi(t_1) \rangle$$



bimagnon: double spin flip
 $\sim 3J$ (spin exchange)



Wang *et al.* *Phys. Rev. B* **98**, 245106 (2018)

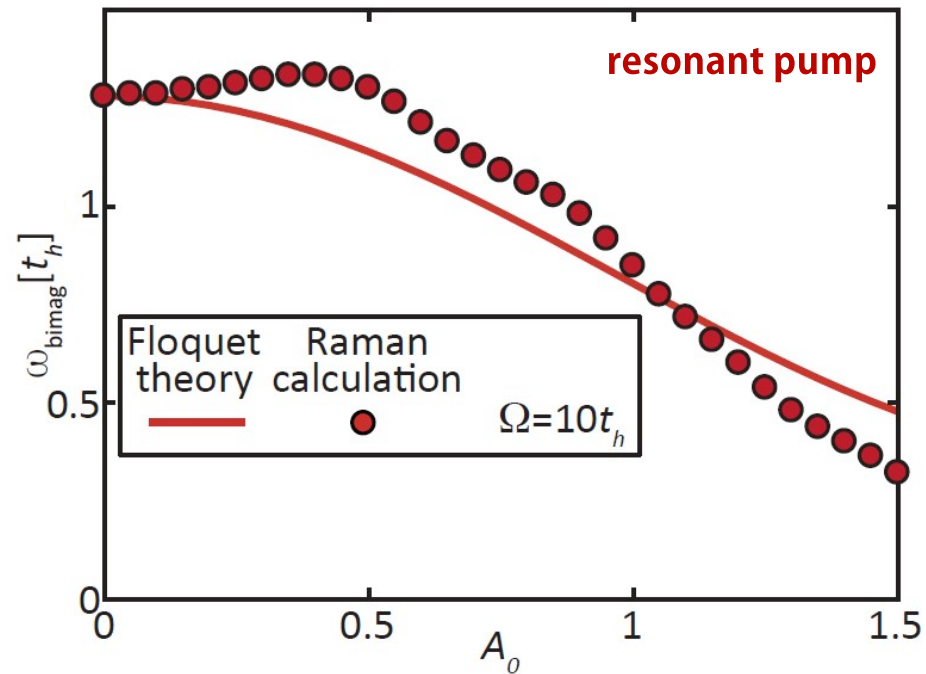
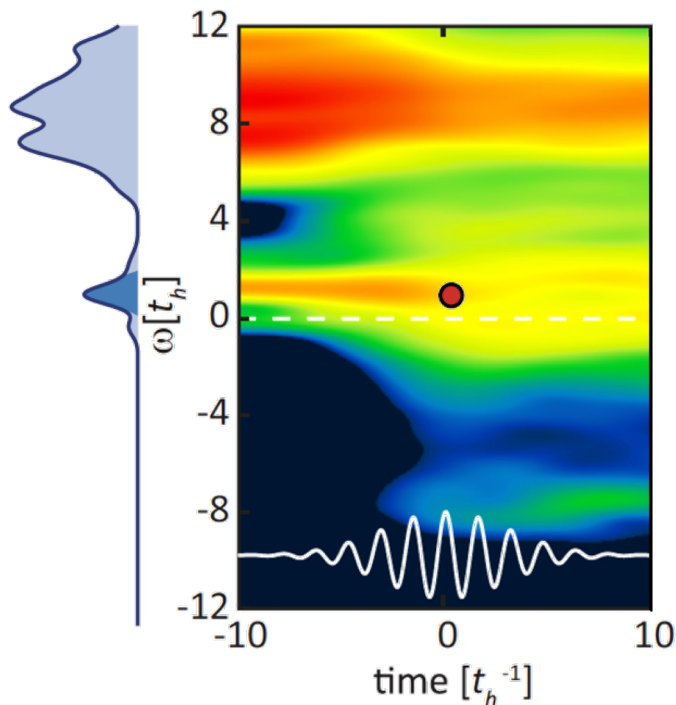
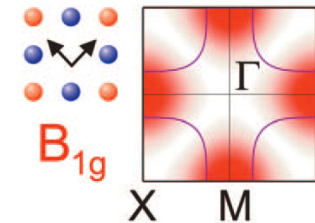
2D Hubbard $U=8t_h$ | pump: $A_0=0.6$, $\Omega = 4t_h$, $\sigma_t = 3.0t_h^{-1}$ | probe: $\sigma = 2.0t_h^{-1}$

Time-Resolved Raman Scattering: Tracking Spin Exchange

- Time-resolved (non-resonant) Raman cross-section

$$\mathcal{R}_{\varepsilon_s \varepsilon_i}(\omega, t) = \iint_{-\infty}^{\infty} dt_1 dt_2 e^{i\omega(t_2 - t_1)} g(t_1; t) g(t_2; t) \chi_{\varepsilon_s \varepsilon_i}(t_1, t_2)$$

$$\chi_{\varepsilon_s \varepsilon_i}(t_1, t_2) = i \langle \psi(t_2) | \hat{\gamma}_{\varepsilon_s \varepsilon_i}^\dagger(t_2) \mathcal{U}_0(t_2, t_1) \hat{\gamma}_{\varepsilon_s \varepsilon_i}(t_1) | \psi(t_1) \rangle$$



Wang *et al.* *Phys. Rev. B* **98**, 245106 (2018)

2D Hubbard $U=8t_h$ | pump: $\Omega = 10t_h, \sigma_t = 3.0t_h^{-1}$ | probe: $\sigma = 2.0t_h^{-1}$

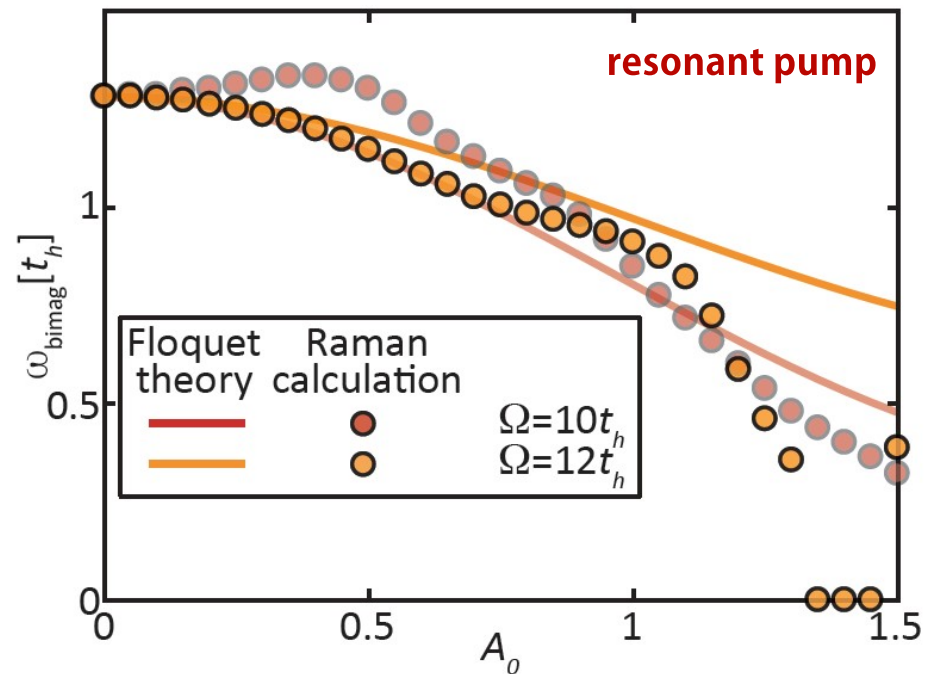
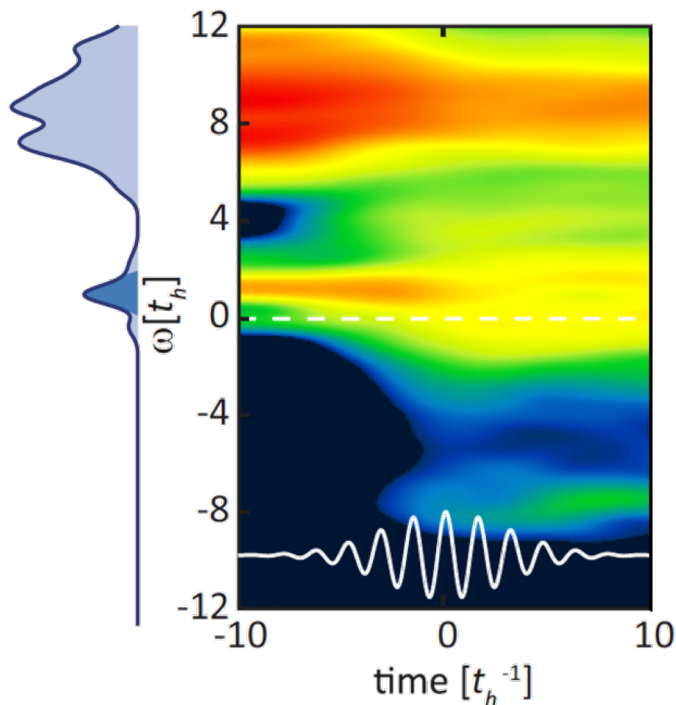
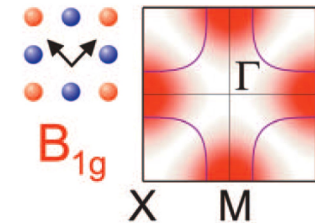


Time-Resolved Raman Scattering: Tracking Spin Exchange

- Time-resolved (non-resonant) Raman cross-section

$$\mathcal{R}_{\varepsilon_s \varepsilon_i}(\omega, t) = \iint_{-\infty}^{\infty} dt_1 dt_2 e^{i\omega(t_2 - t_1)} g(t_1; t) g(t_2; t) \chi_{\varepsilon_s \varepsilon_i}(t_1, t_2)$$

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Wang *et al.* *Phys. Rev. B* **98**, 245106 (2018)

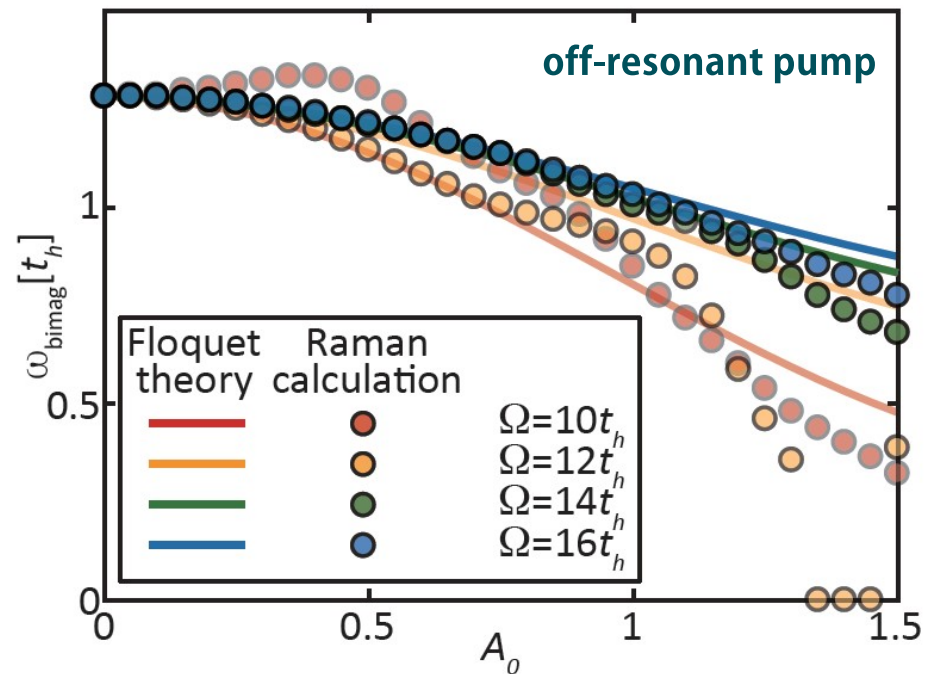
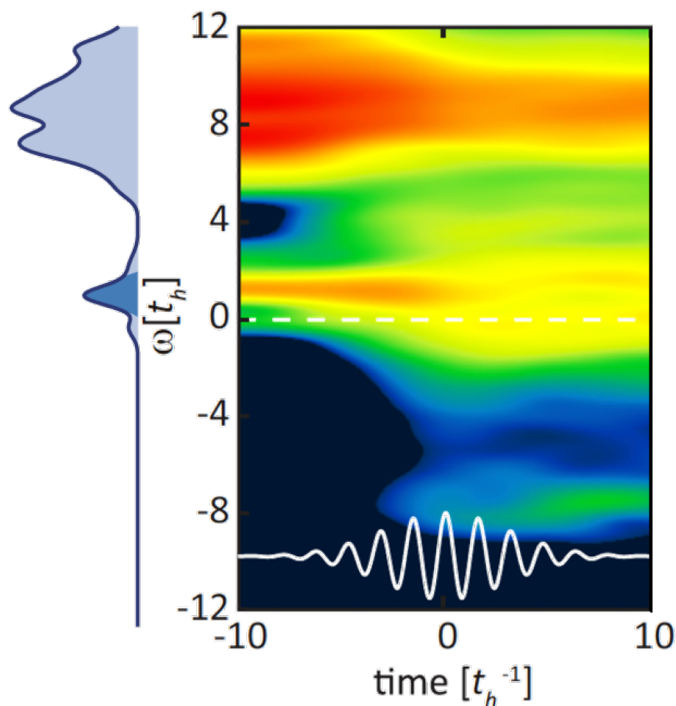
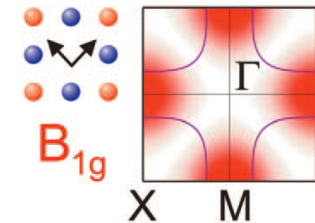
2D Hubbard $U = 8 t_h$ | pump: $\Omega = 10-12 t_h, \sigma_t = 3.0 t_h^{-1}$ | probe: $\sigma = 2.0 t_h^{-1}$

Time-Resolved Raman Scattering: Tracking Spin Exchange

- Time-resolved (non-resonant) Raman cross-section

$$\mathcal{R}_{\varepsilon_s \varepsilon_i}(\omega, t) = \iint_{-\infty}^{\infty} dt_1 dt_2 e^{i\omega(t_2 - t_1)} g(t_1; t) g(t_2; t) \chi_{\varepsilon_s \varepsilon_i}(t_1, t_2)$$

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Wang *et al.* *Phys. Rev. B* **98**, 245106 (2018)

2D Hubbard $U=8t_h$ | pump: $\Omega = 10-16t_h$, $\sigma_t = 3.0t_h^{-1}$ | probe: $\sigma = 2.0t_h^{-1}$

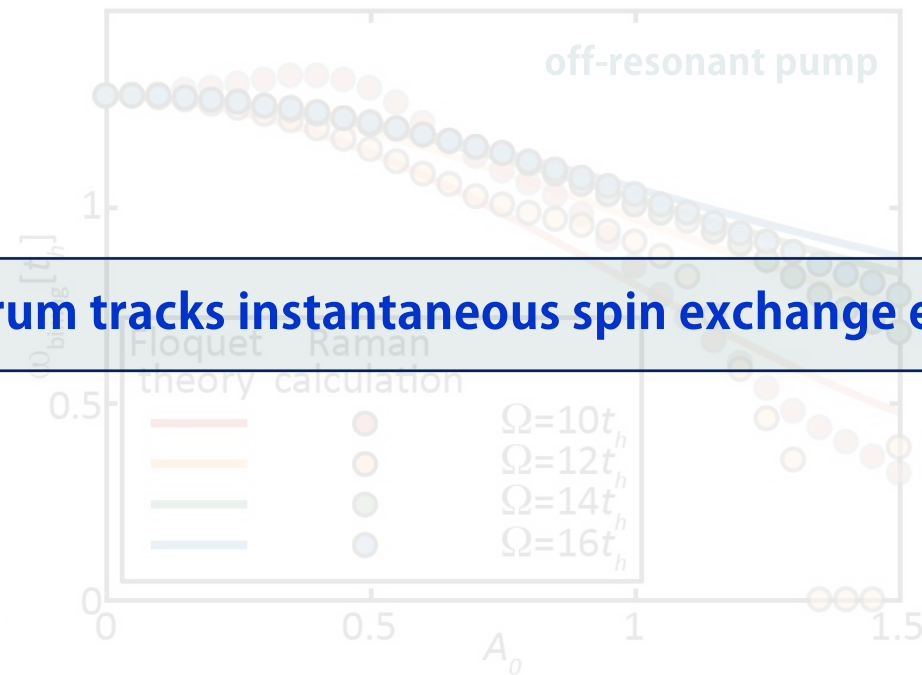
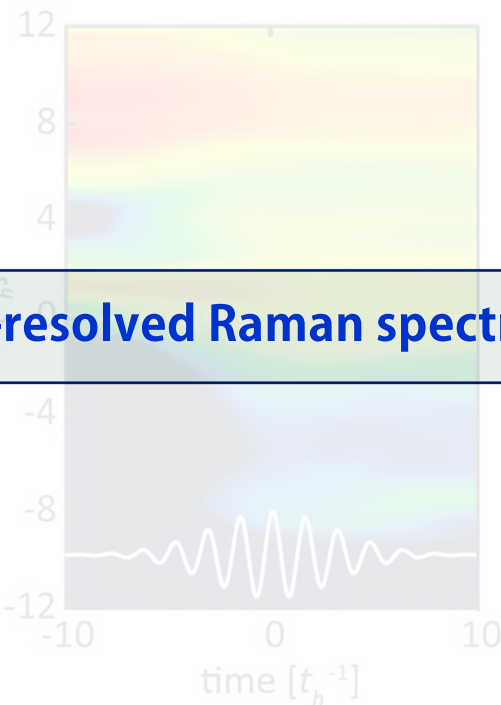
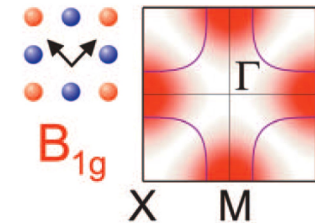


Time-Resolved Raman Scattering: Tracking Spin Exchange

- Time-resolved (non-resonant) Raman cross-section

$$\mathcal{R}_{\varepsilon_s \varepsilon_i}(\omega, t) = \iint_{-\infty}^{\infty} dt_1 dt_2 e^{i\omega(t_2 - t_1)} g(t_1; t) g(t_2; t) \chi_{\varepsilon_s \varepsilon_i}(t_1, t_2)$$

$$\chi_{\varepsilon_s \varepsilon_i}(t_1, t_2) = i \langle \psi(t_2) | \hat{\gamma}_{\varepsilon_s \varepsilon_i}^\dagger(t_2) \mathcal{U}_0(t_2, t_1) \hat{\gamma}_{\varepsilon_s \varepsilon_i}(t_1) | \psi(t_1) \rangle$$



Time-resolved Raman spectrum tracks instantaneous spin exchange energy

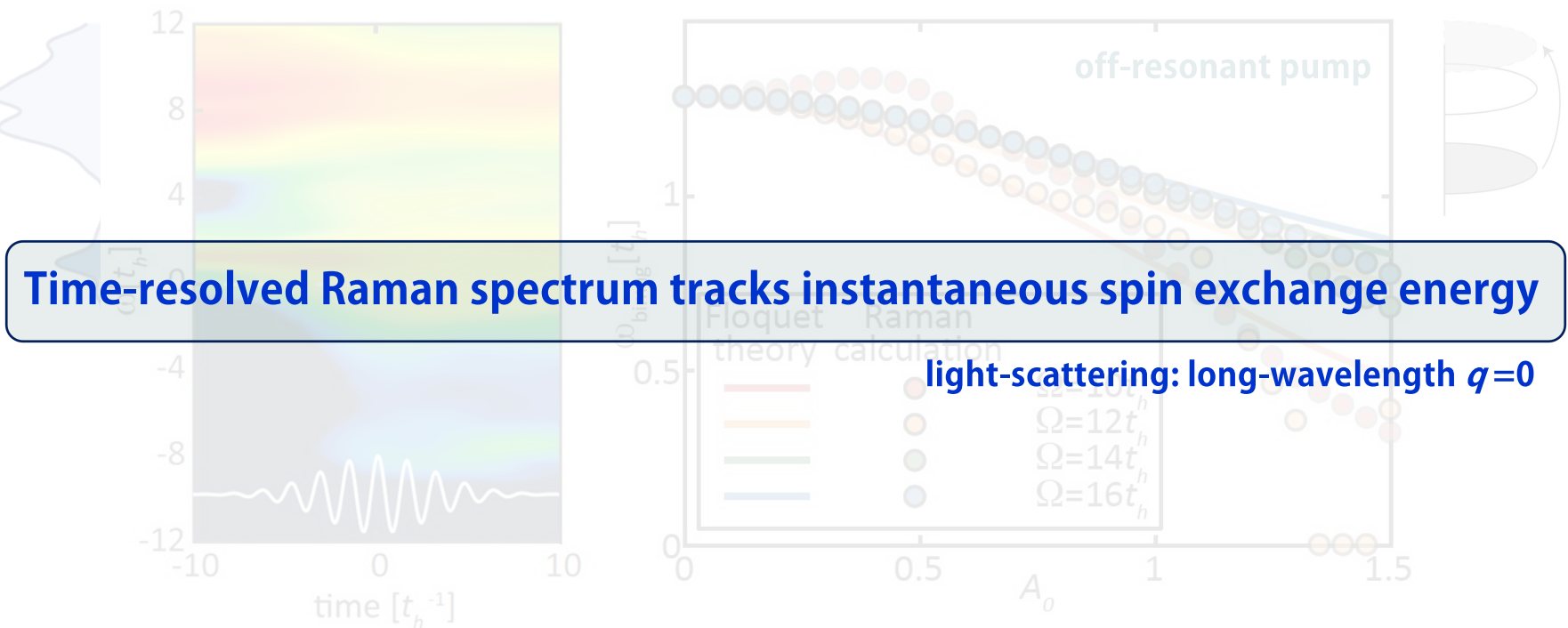
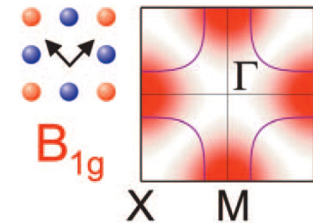


Time-Resolved Raman Scattering: Tracking Spin Exchange

- Time-resolved (non-resonant) Raman cross-section

$$\mathcal{R}_{\varepsilon_s \varepsilon_i}(\omega, t) = \iint_{-\infty}^{\infty} dt_1 dt_2 e^{i\omega(t_2 - t_1)} g(t_1; t) g(t_2; t) \chi_{\varepsilon_s \varepsilon_i}(t_1, t_2)$$

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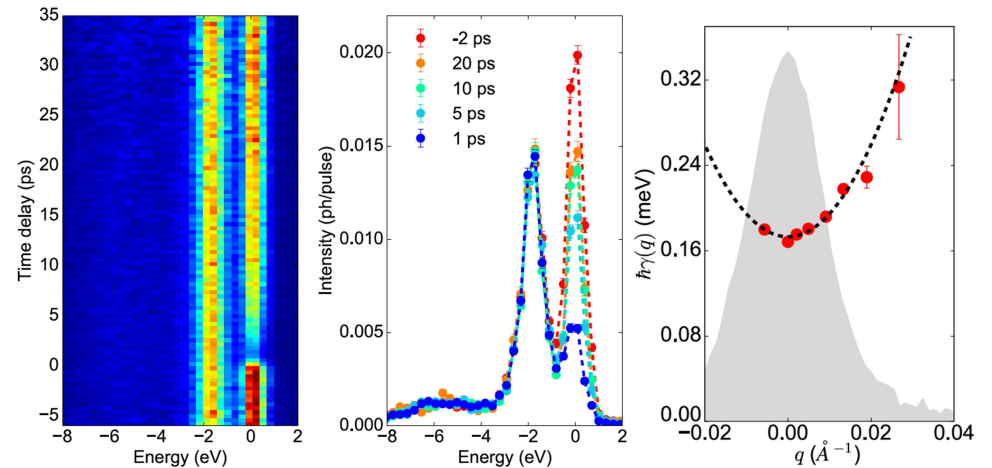
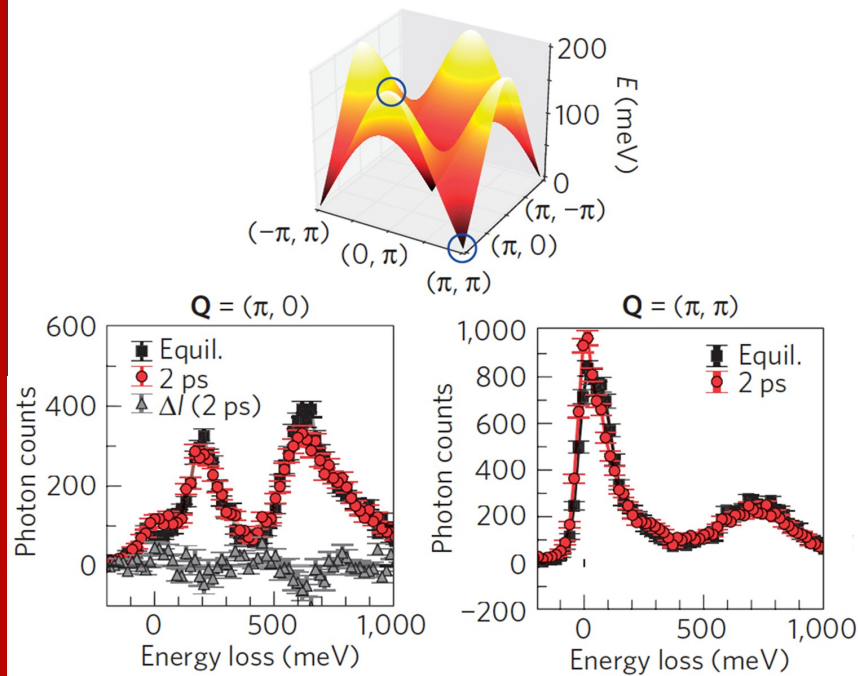
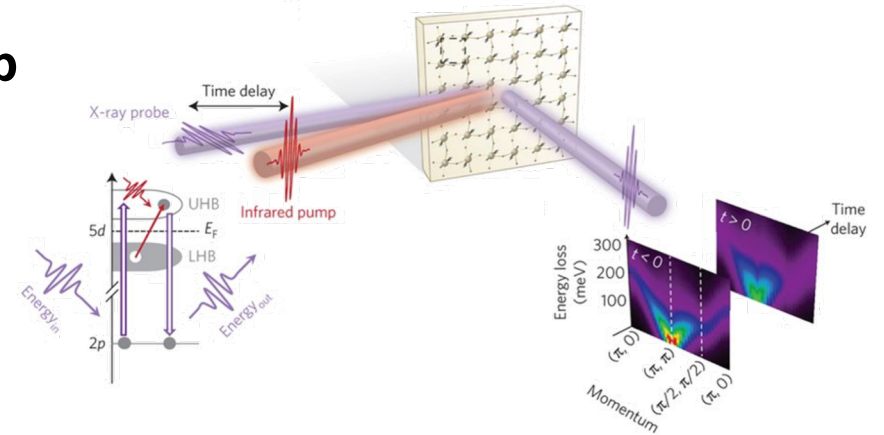


Wang *et al.* *Phys. Rev. B* **98**, 245106 (2018)

2D Hubbard $U=8t_h$ | pump: $\Omega = 10-16t_h, \sigma_t = 3.0t_h^{-1}$ | probe: $\sigma = 2.0t_h^{-1}$

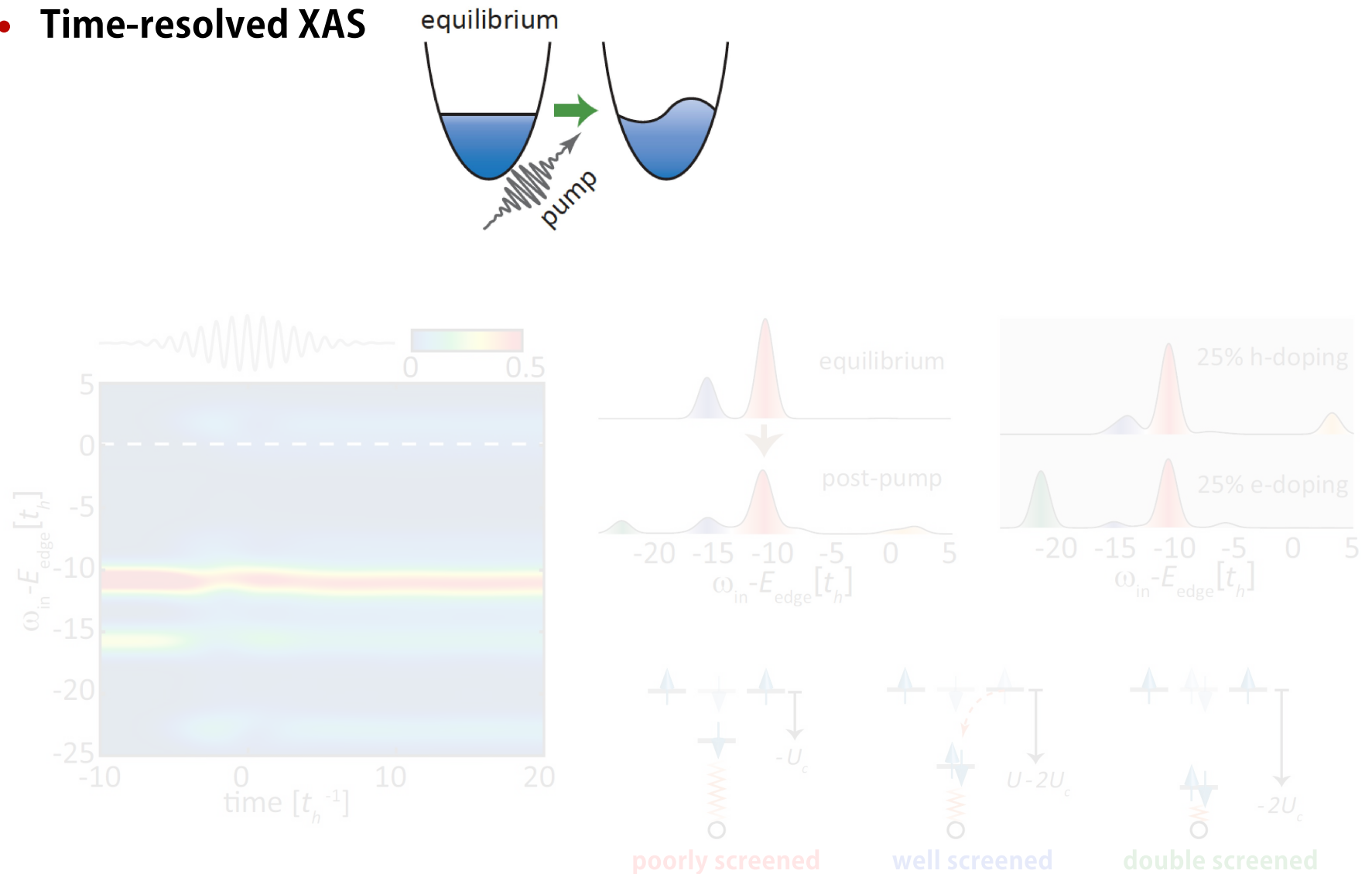
Time-Resolved RIXS Technique

- Time-resolved RIXS experimental setup
- trRIXS in spin-orbital system (iridate)
- trRIXS in stripe ordered cuprate



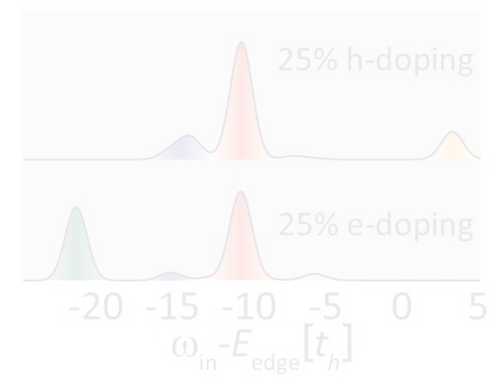
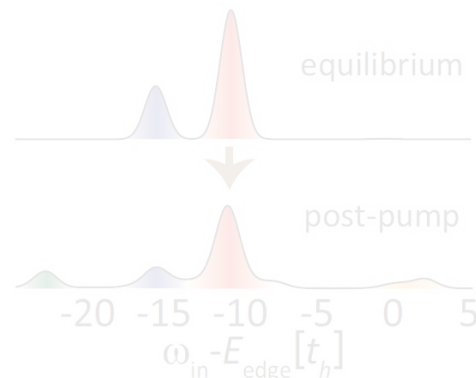
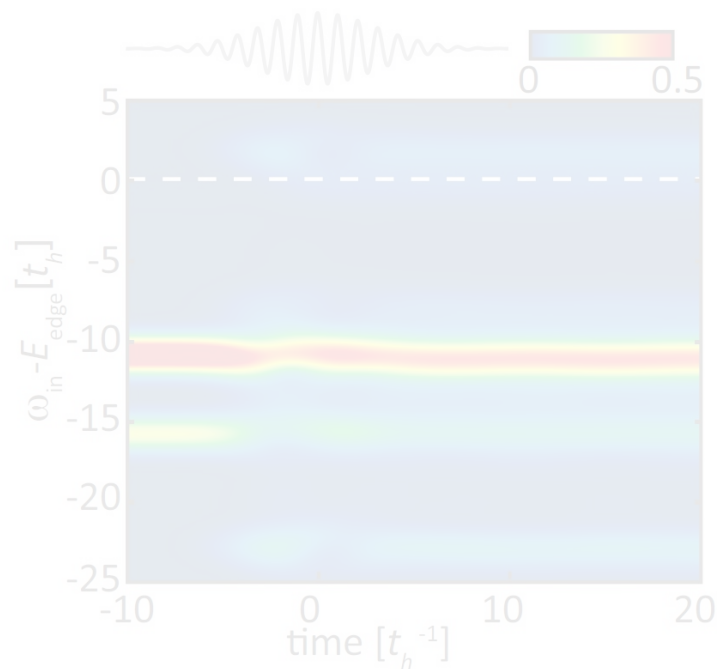
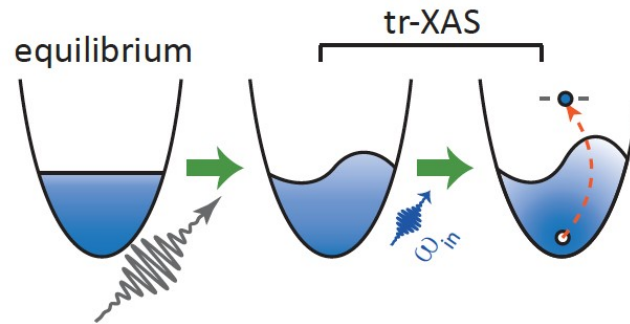
Time-Resolved X-Ray Absorption: Tracking Configuration

- Time-resolved XAS



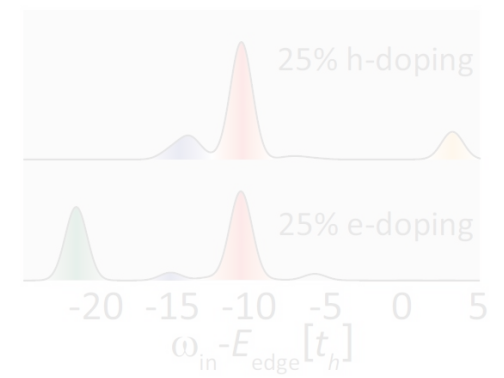
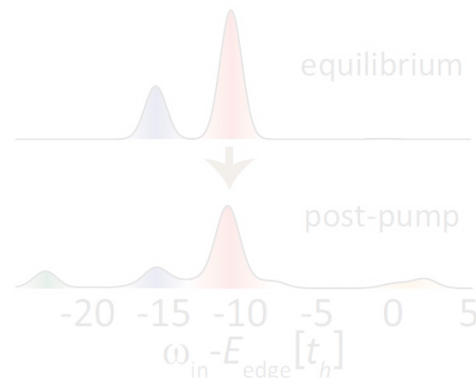
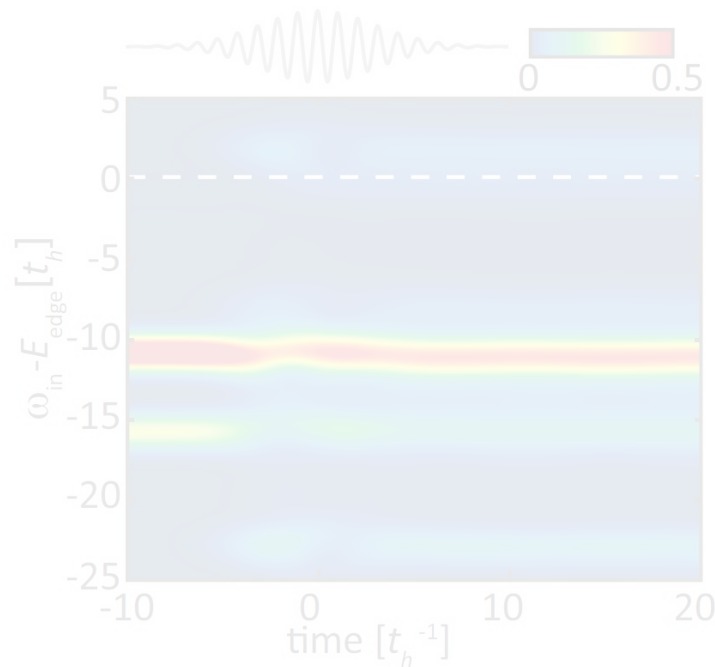
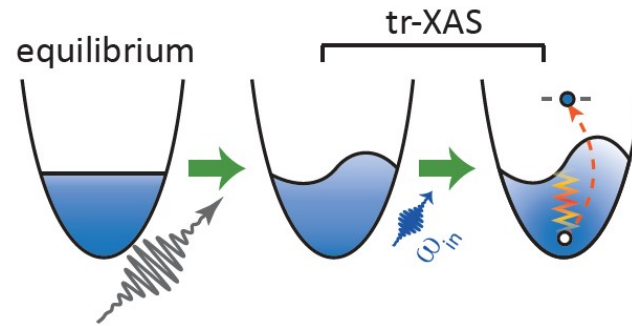
Time-Resolved X-Ray Absorption: Tracking Configuration

- Time-resolved XAS



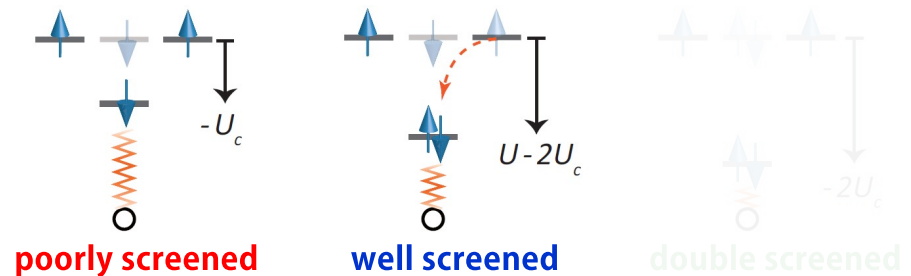
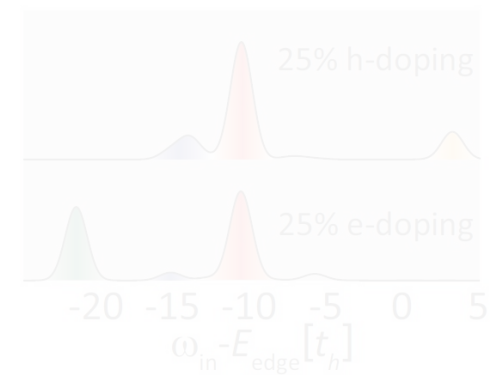
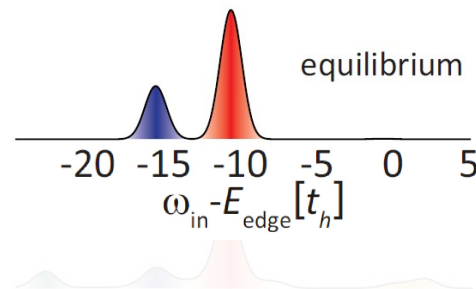
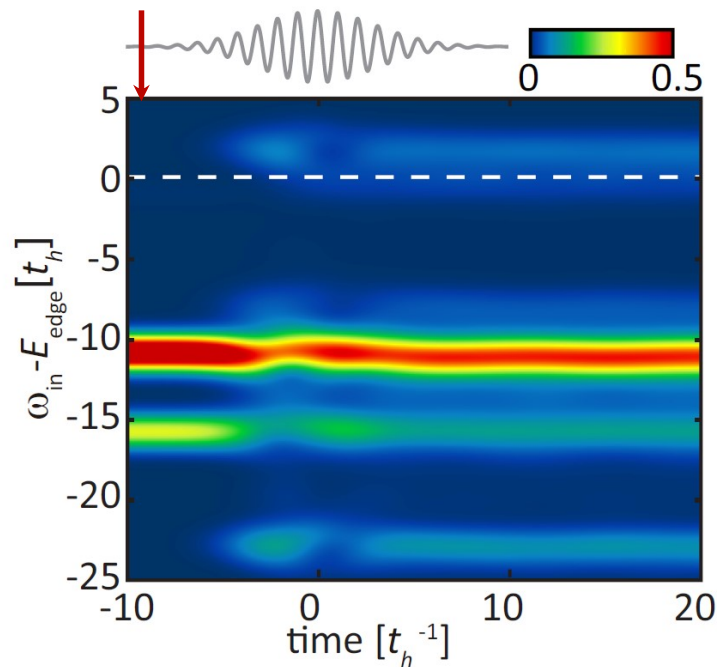
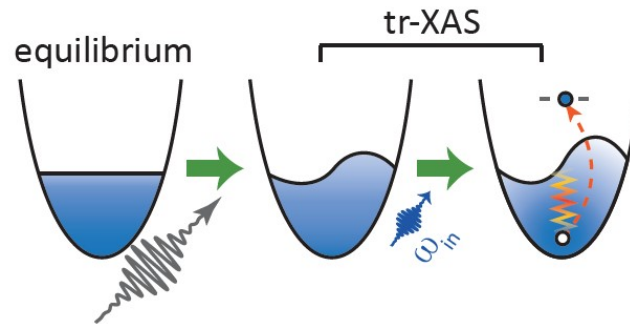
Time-Resolved X-Ray Absorption: Tracking Configuration

- Time-resolved XAS



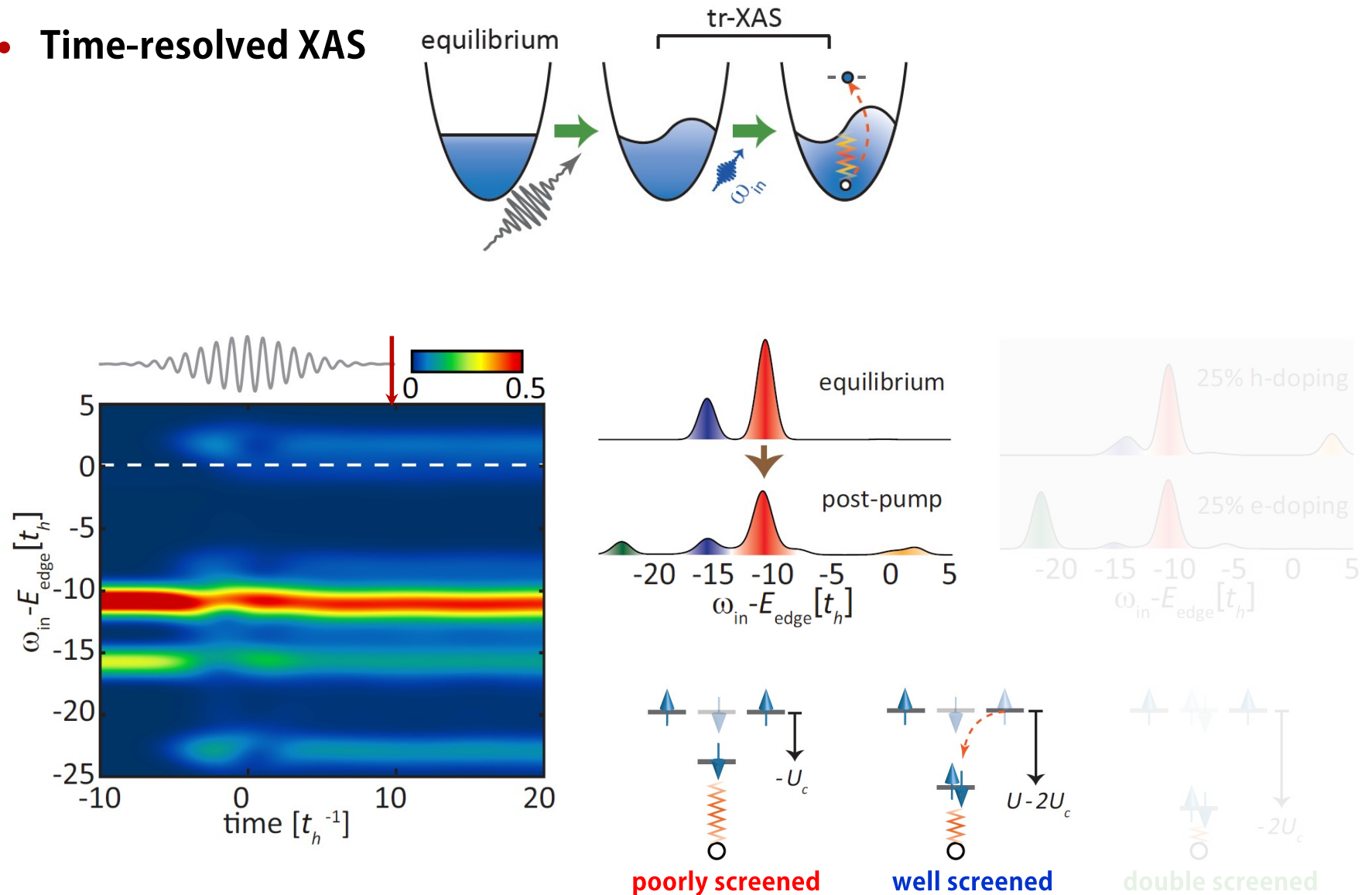
Time-Resolved X-Ray Absorption: Tracking Configuration

- Time-resolved XAS



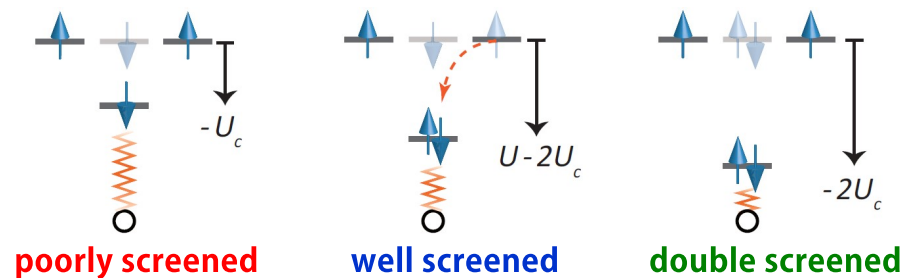
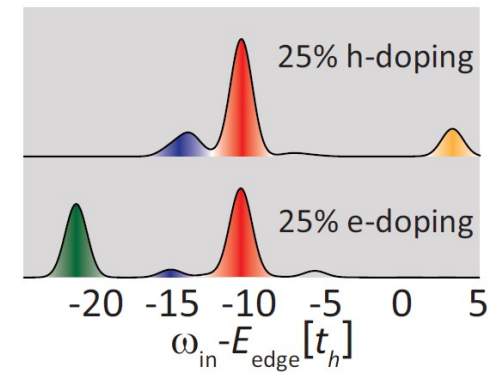
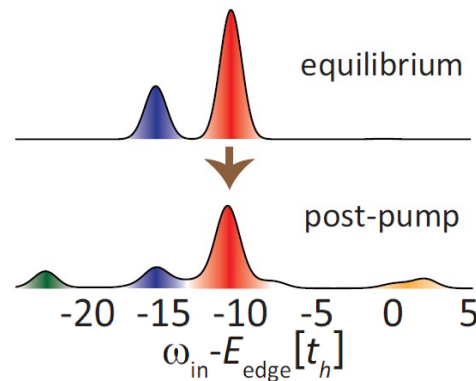
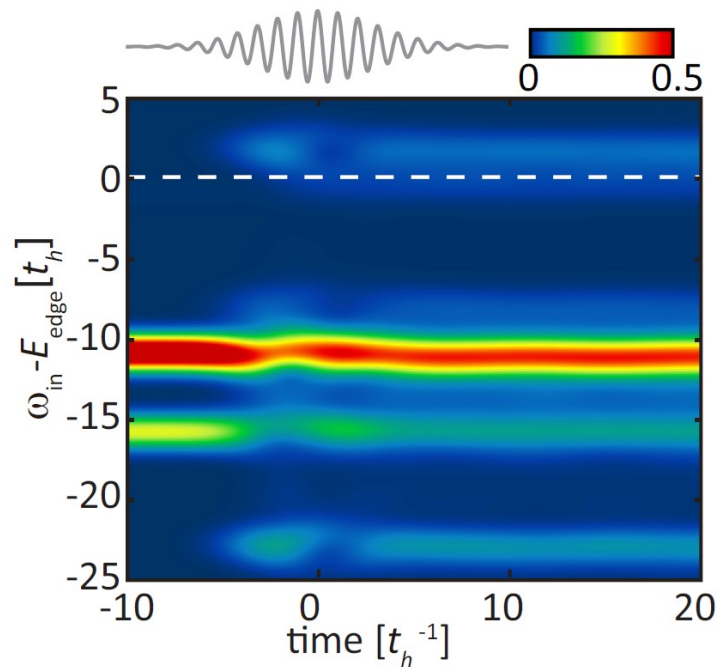
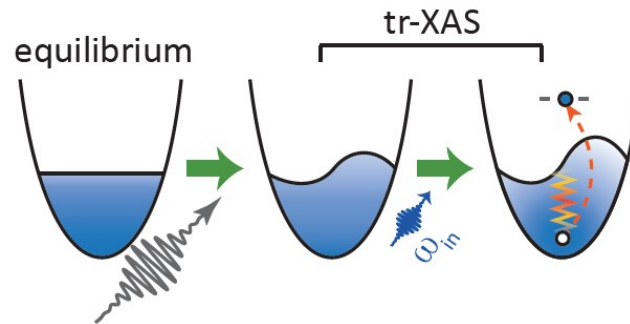
Time-Resolved X-Ray Absorption: Tracking Configuration

- Time-resolved XAS



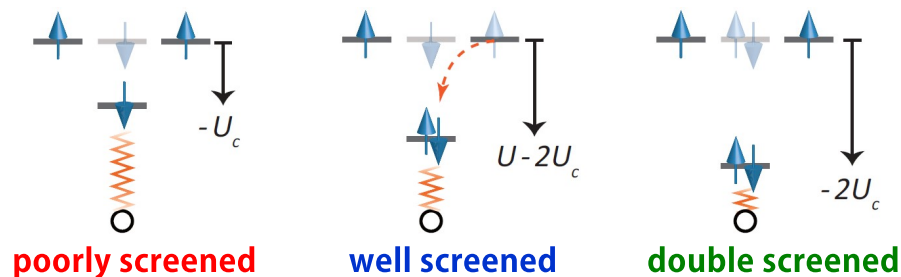
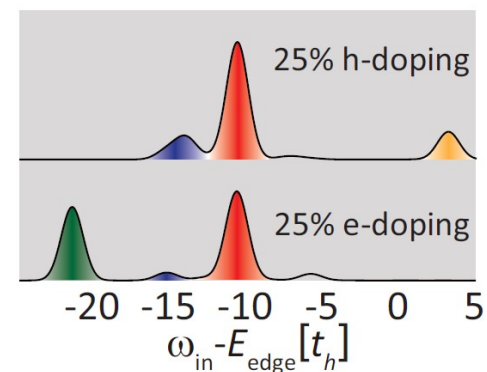
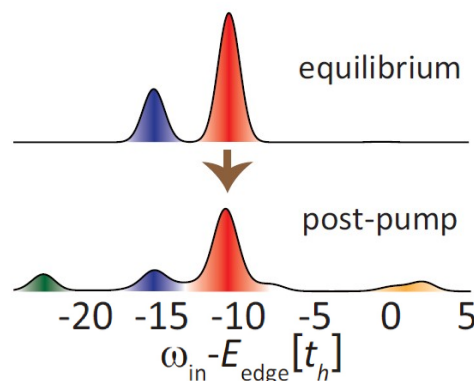
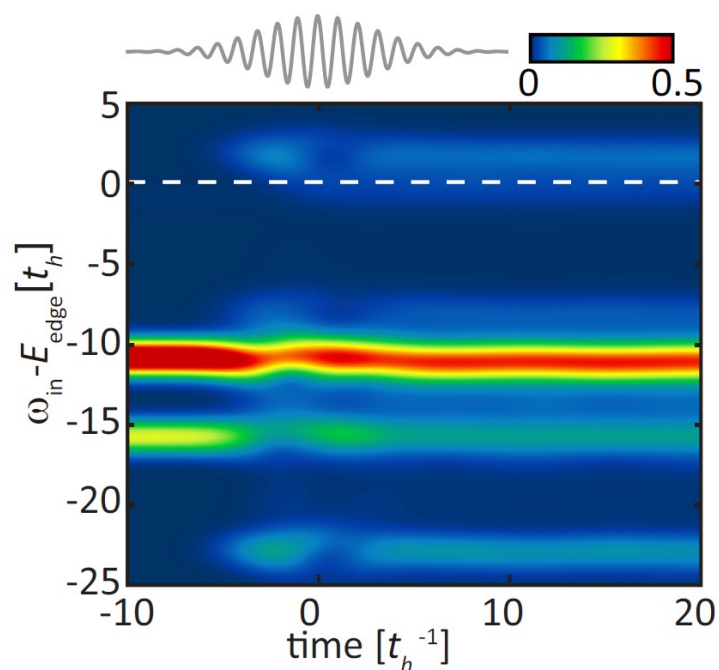
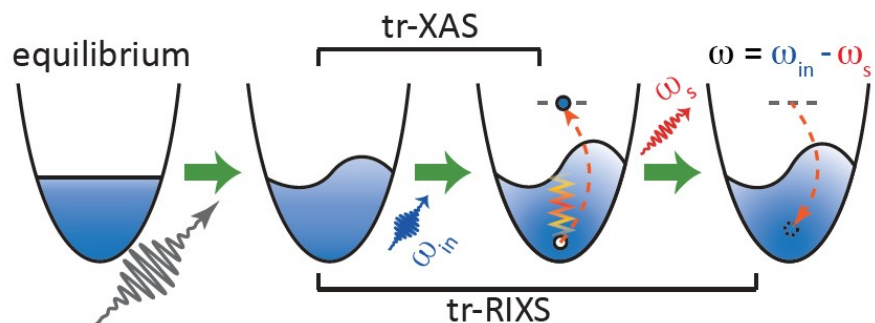
Time-Resolved X-Ray Absorption: Tracking Configuration

- Time-resolved XAS



Time-Resolved X-Ray Absorption: Tracking Configuration

- Time-resolved XAS
RIXS



Time-Resolved RIXS: Dissecting Charge Excitations

- Time-resolved RIXS cross-section [indirect *e.g.* Cu *K*-edge; $q = (\pi, 0)$]

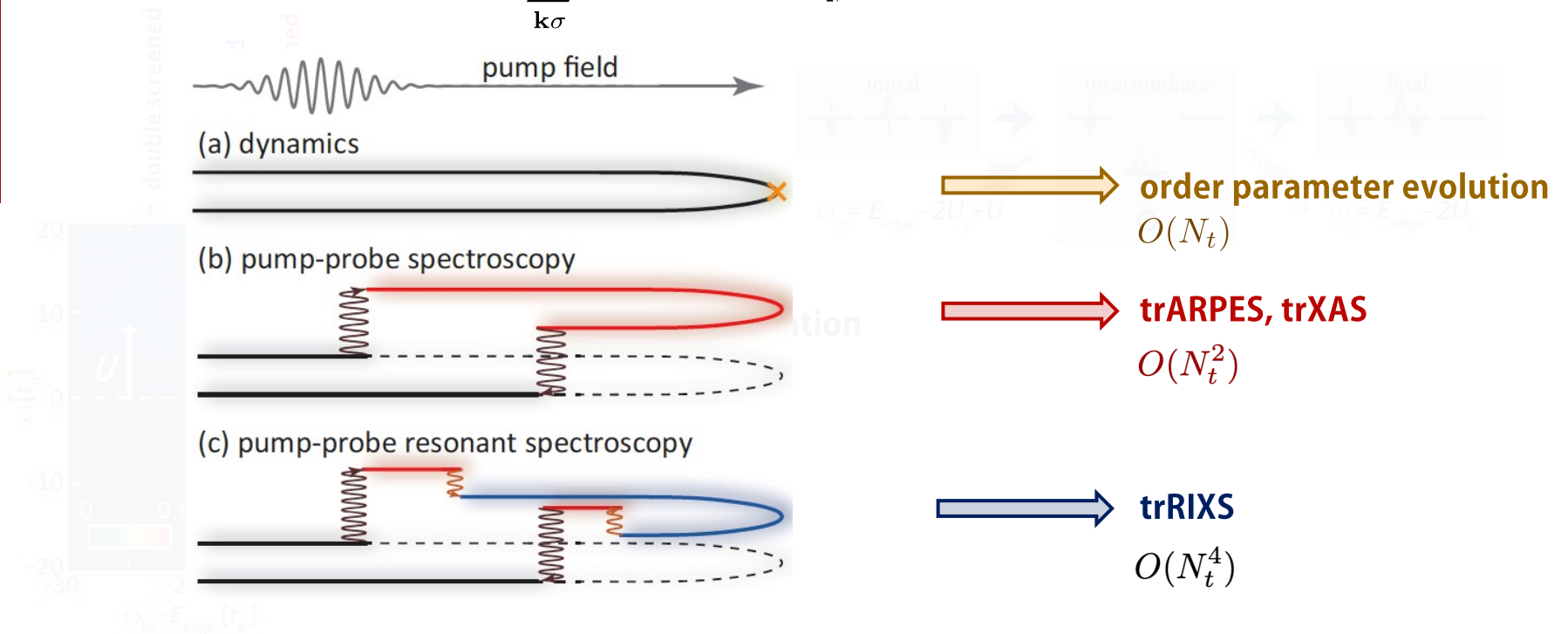
$$\mathcal{I}(\mathbf{q}, \omega_i, \omega_s)(t) = \iint dt_2 dt'_2 \int_{-\infty}^{t'_2} dt_2 \int_{-\infty}^{t'_1} dt_1 e^{i\omega_i(t_2-t_1) - i\omega_s(t'_2-t'_1)} g(t_1, t) g(t_2, t) l(t_1, t'_1) l(t_2, t'_2) \\ \times \langle \hat{D}_{\mathbf{q}_i \varepsilon_i}^\dagger(t_2) \hat{D}_{\mathbf{q}_s \varepsilon_s}(t'_2) \hat{D}_{\mathbf{q}_s \varepsilon_s}^\dagger(t'_1) \hat{D}_{\mathbf{q}_i \varepsilon_i}(t_1) \rangle$$

probe shape function $g(\tau; t)$

core-hole lifetime $l(t_1, t'_1) = e^{-|t'_1-t_1|/\tau_{\text{core}}}$

dipole transition $\mathcal{D}_{\mathbf{q}\varepsilon}(t) = \sum_{\mathbf{k}\sigma} M_{\alpha\beta}(\mathbf{q}, \mathbf{k}, t) c_{\mathbf{k}+\mathbf{q}, \sigma}^{(\alpha)\dagger} c_{\mathbf{k}\sigma}^{(\beta)}$

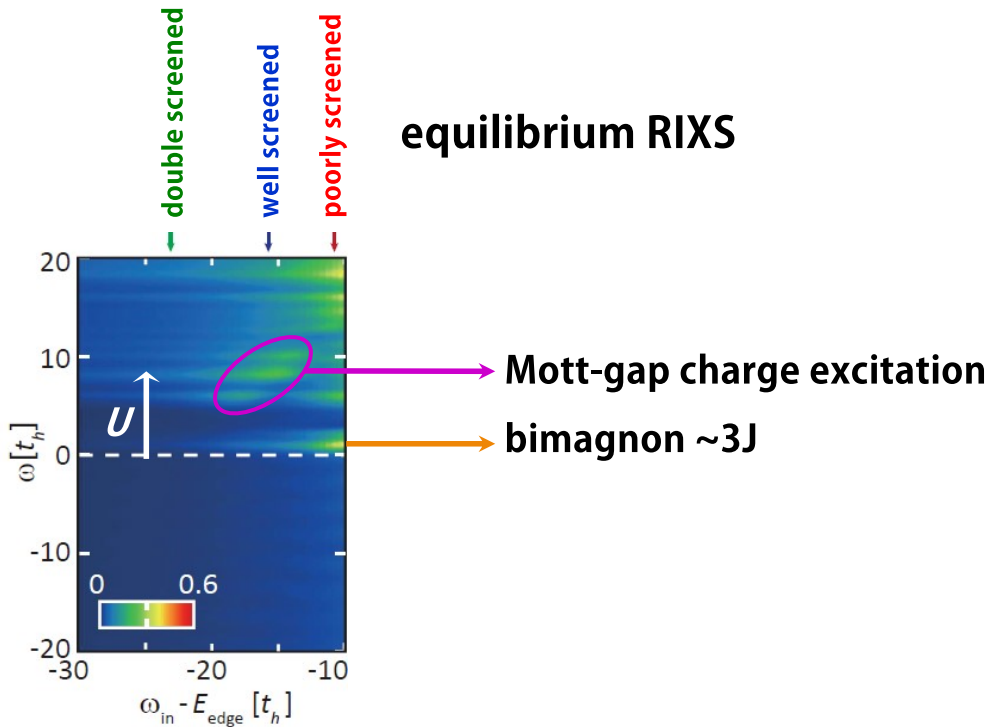
matrix element $M_{\alpha\beta}(\mathbf{q}, \mathbf{k}, t)$



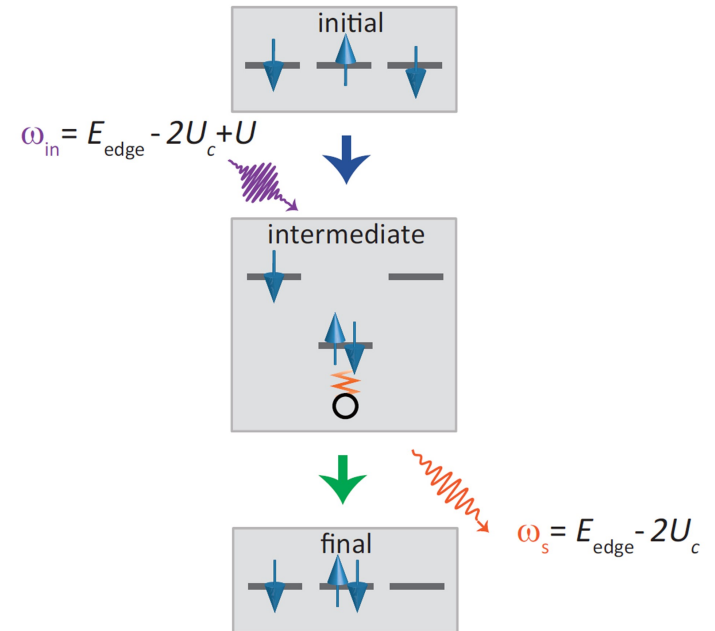
Time-Resolved RIXS: Dissecting Charge Excitations

- Time-resolved RIXS cross-section [indirect *e.g.* Cu *K*-edge; $q = (\pi, 0)$]

$$\mathcal{I}(\mathbf{q}, \omega_i, \omega_s)(t) = \iint dt_2 dt'_2 \int_{-\infty}^{t'_2} dt_2 \int_{-\infty}^{t'_1} dt_1 e^{i\omega_i(t_2-t_1) - i\omega_s(t'_2-t'_1)} g(t_1, t) g(t_2, t) l(t_1, t'_1) l(t_2, t'_2) \\ \times \langle \hat{\mathcal{D}}_{\mathbf{q}_i \varepsilon_i}^\dagger(t_2) \hat{\mathcal{D}}_{\mathbf{q}_s \varepsilon_s}(t'_2) \hat{\mathcal{D}}_{\mathbf{q}_s \varepsilon_s}^\dagger(t'_1) \hat{\mathcal{D}}_{\mathbf{q}_i \varepsilon_i}(t_1) \rangle$$



generation of Mott-gap excitation

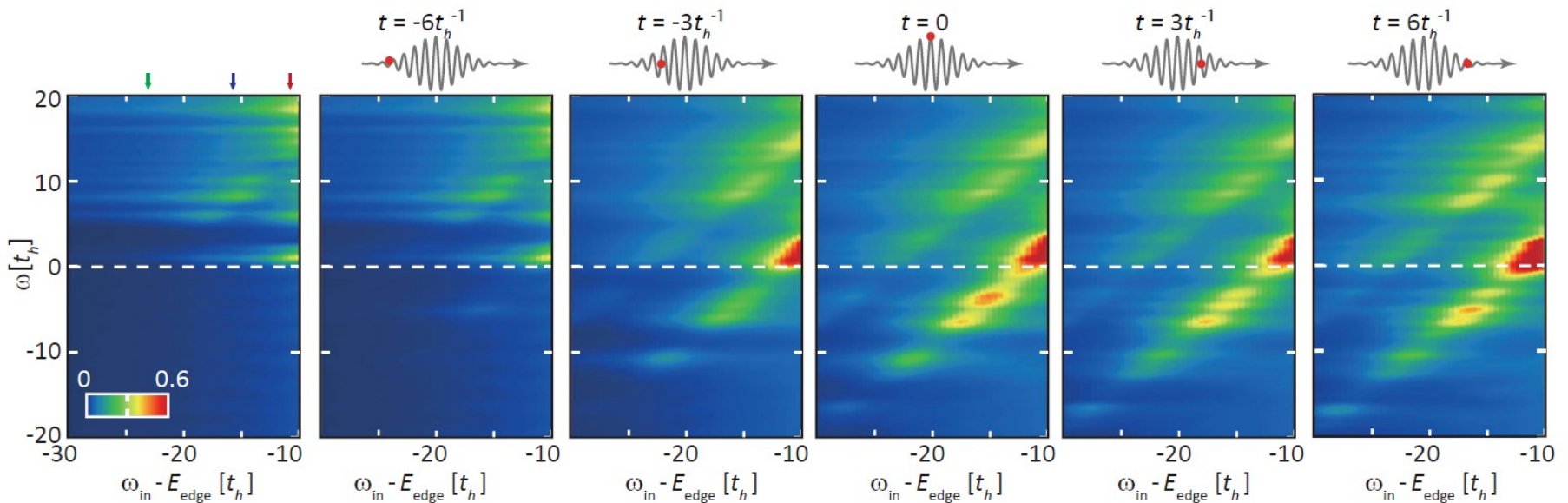


Time-Resolved RIXS: Dissecting Charge Excitations

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- Post-pump trRIXS
 - Tracking magnetic and charge excitation: bimagnon and in-gap charge

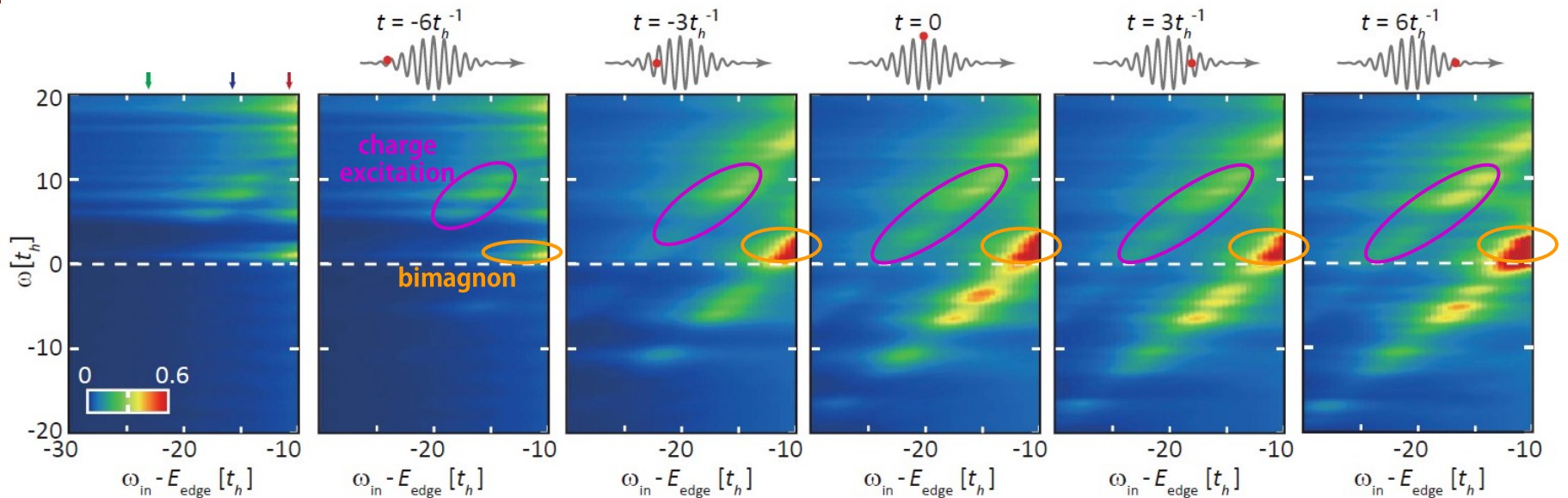


Time-Resolved RIXS: Dissecting Charge Excitations

- Time-resolved RIXS cross-section [indirect *e.g.* Cu *K*-edge; $q = (\pi, 0)$]

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- Post-pump trRIXS
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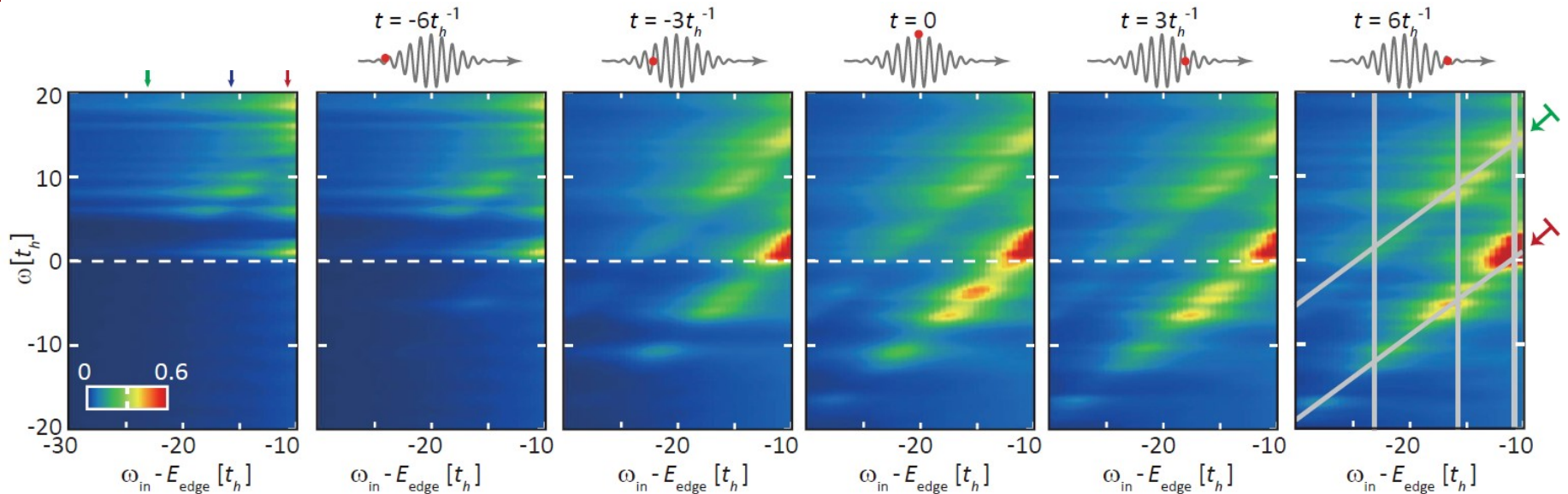


Time-Resolved RIXS: Dissecting Charge Excitations

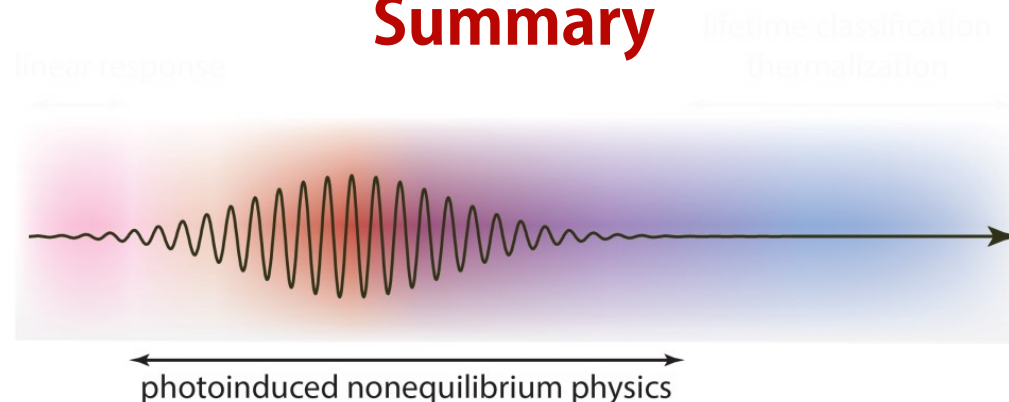
- Time-resolved RIXS cross-section [indirect *e.g.* Cu *K*-edge; $q = (\pi, 0)$]

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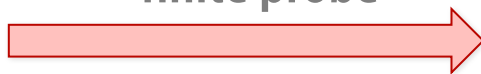
- Post-pump trRIXS
 - Tracking magnetic and charge excitation: bimagnon and in-gap charge



Summary

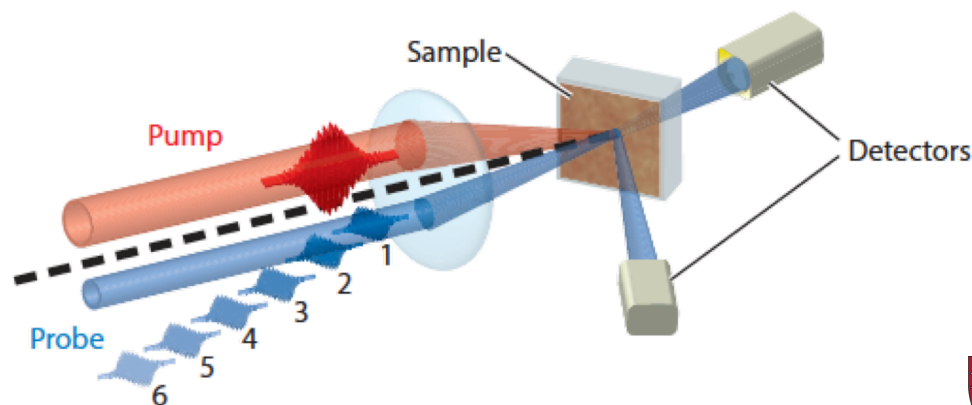


- How are excitations engineered by a pulsed pump?

- Transient virtual states  remnant excitations

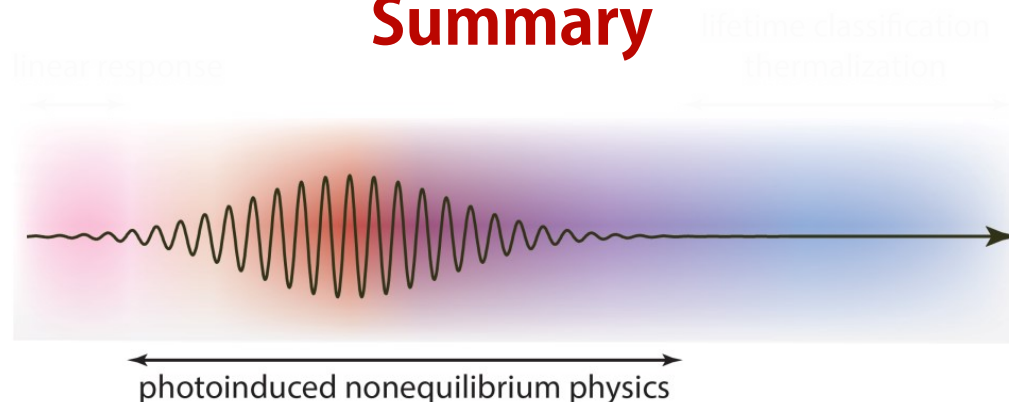
- How to detect transient collective excitations?

- **trRaman**: bimagnon **trXAS**: global photodoping **trRIXS**: charge and spin

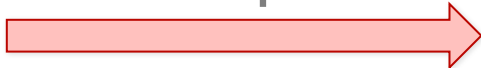




Summary



- **How are excitations engineered by a pulsed pump?**

- Transient virtual states  remnant excitations
correlations

- **How to detect transient collective excitations?**

- **trRaman**: bimagnon **trXAS**: global photodoping **trRIXS**: charge and spin



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Cheng-Chien Chen
UAB



Brian Moritz
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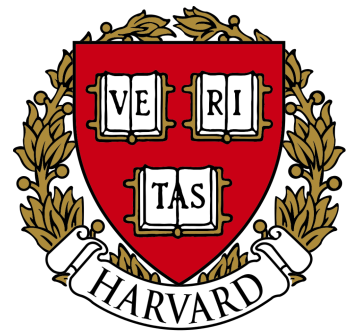


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HARVARD
UNIVERSITY

Thanks for your attention!



Thanks for your attention!

