Time and momentum resolved scattering approaches for non-equilibrium spectroscopies in correlated systems

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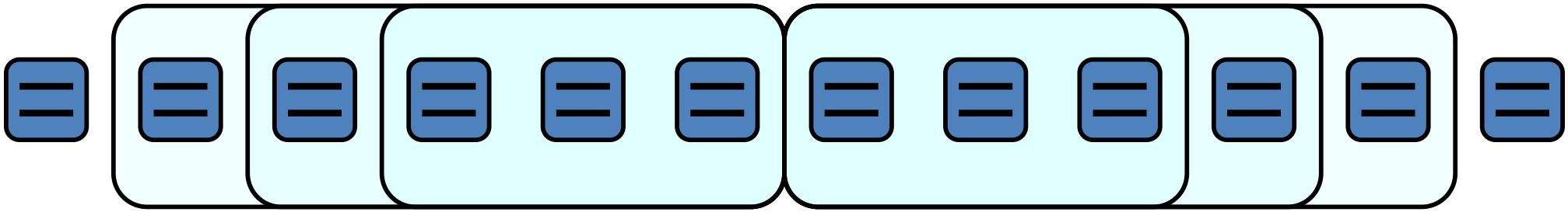

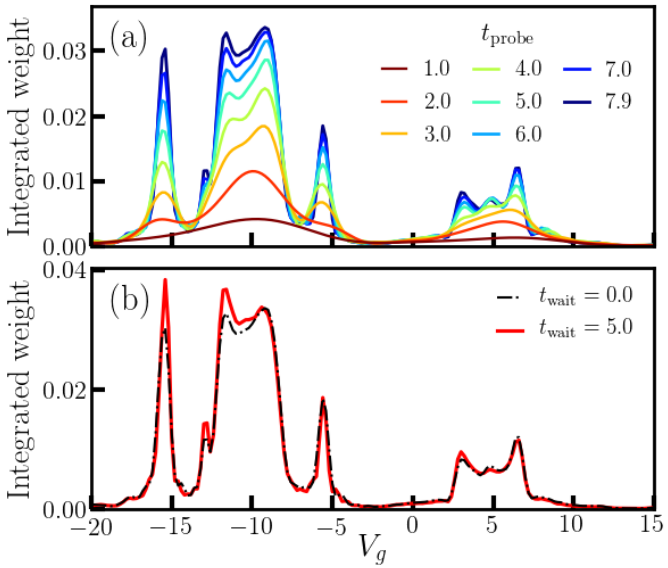

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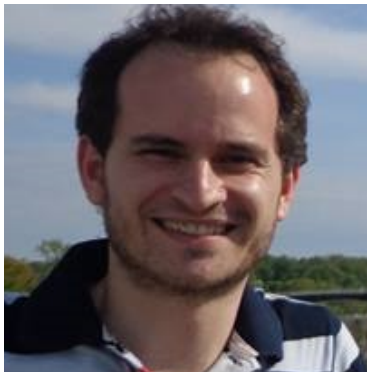
Krissia Zawadzki
(ICTP-Sao Paulo, Brazil)



Alberto Nocera
(now @ UBC)

Luhang Yang
(Northeastern)

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References:

K. Zawadzki and AEF, Phys. Rev. B 100, 195124 (2019)

K. Zawadzki , A. Nocera and AEF, arXiv:2002.04243

K. Zawadzki and AEF, Phys. Rev. B, 102, 235141 (2020)

What is this talk about?

Arguably, the two most pressing challenges in computational CMT are (IMHO):

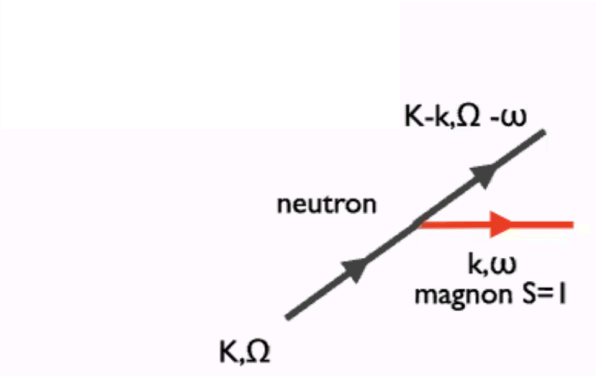
- (unbiased) Methods to study d-dimensional (d>1) strongly correlated systems (spin-liquids, High-Tc). QMC except when it suffers from sign problem. PEPS (see Corboz's talk) is a very promising route, as well as ML inspired variational wave functions.

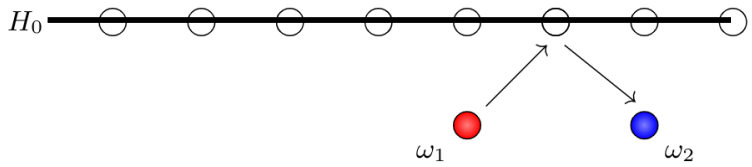
- Methods to calculate non-equilibrium spectra (e.g. after a pump or quench: transient regimes, metastable states, non-equilibrium phase transitions, hidden physics at high energies, Floquet physics).

This talk is about introducing a new general approach to study non-equilibrium spectroscopies based on directly solving the time-dependent Schrödinger Eq.

Energy-loss spectroscopies

”particle-in, particle-out” spectroscopy. E.g., neutron scattering, Compton, EELS





$$H_d = \omega_1 \sum_\ell n_{1\ell} + \omega_2 \sum_\ell n_{2\ell} \qquad \omega = k^2/2m$$

$$V = J' \sum_\ell O_\ell (c^\dagger_{1\ell} c_{2\ell} + H.c.) = \frac{J'}{L^2} \sum_\ell O_\ell \sum_{q,p} \left(e^{i(p-q)\ell} c^\dagger_{2p} c_{1q} + H.c \right)$$

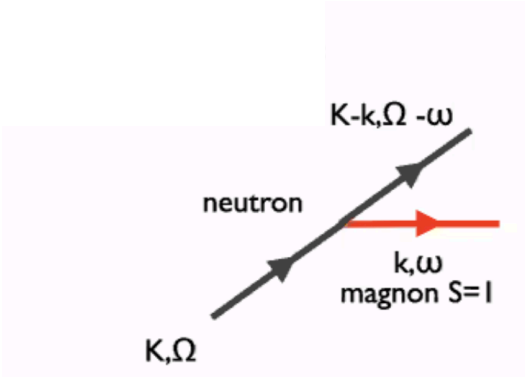
Initial state: $|\Psi(t=0)\rangle = |gs\rangle\,|k_0, 0\rangle$

Detection: $n_{2k}(t) = \frac{1}{L} \sum_{\ell,\ell'} e^{ik(\ell-\ell)} \langle c^\dagger_{2\ell} c_{2\ell'}(t) \rangle$

We measure the energy and momentum “lost” by the particle (energy and momentum transferred to the sample)

See, e.g., I. A. Zaliznyak and J. M. Tranquada, “Neutron scattering and its application to strongly correlated systems,”

In equilibrium



$$H_d = \omega_1 \sum_\ell n_{1\ell} + \omega_2 \sum_\ell n_{2\ell}$$

$$V = J' \sum_\ell O_\ell (c^\dagger_{1\ell} c_{2\ell} + H.c.) = \frac{J'}{L^2} \sum_\ell O_\ell \sum_{q,p} \left(e^{i(p-q)\ell} c^\dagger_{2p} c_{1q} + H.c \right)$$

Initial state: $|\Psi(t=0)\rangle = |gs\rangle\,|k_0, 0\rangle$

Detection: $n_{2k}(t) = \frac{1}{L} \sum_{\ell,\ell'} e^{ik(\ell-\ell)} \langle c^\dagger_{2\ell} c_{2\ell'}(t) \rangle$

$$\langle n_d(k,t) \rangle = \int_0^t dt_1 \int_0^t dt_2 \langle V_k(t_1) n_d(k,t) V_k(t_2) \rangle$$

$$\sim \sum |\langle m|O_k|gs\rangle|^2 \delta(\omega - E_m + E_0)$$ Fermi's Golden Rule

Usual formulation in terms of equilibrium Green's functions
-> Dynamic structure factors/spectral functions

Efficient methods for large systems: Dynamical DMRG, time-dependent DMRG, NRG, QMC+MaxEnt…

Out of equilibrium

Initial state: $\rho_0 = \rho \otimes |0\rangle\langle 0|$ with $\rho = \sum_n a_m^* a_n |m\rangle\langle n|$

$$S_O(\mathbf{k}, \omega, t) = 4\pi^2 \sum_m \left| \sum_n a_n \langle m|O_\mathbf{k}|n\rangle \delta^t(\omega - \omega_{mn}) \right|^2 ,$$

where we have introduced the time dependence in the definition

$$\delta^t(\omega) = \frac{1}{\pi} \frac{\sin(\omega t/2)}{\omega} \underset{t\to\infty}{\to} \delta(\omega)$$

We need all the eigenstates and eigenvalues to evaluate the allowed transition energies and matrix elements!

J. K. Freericks, H. R. Krishnamurthy, and T. Pruschke, Phys. Rev. Lett. 102, 136401 (2009).
C. Shao, T. Tohyama, H.-G. Luo, and H. Lu, Phys. Rev. B 93, 195144 (2016).

Idea:

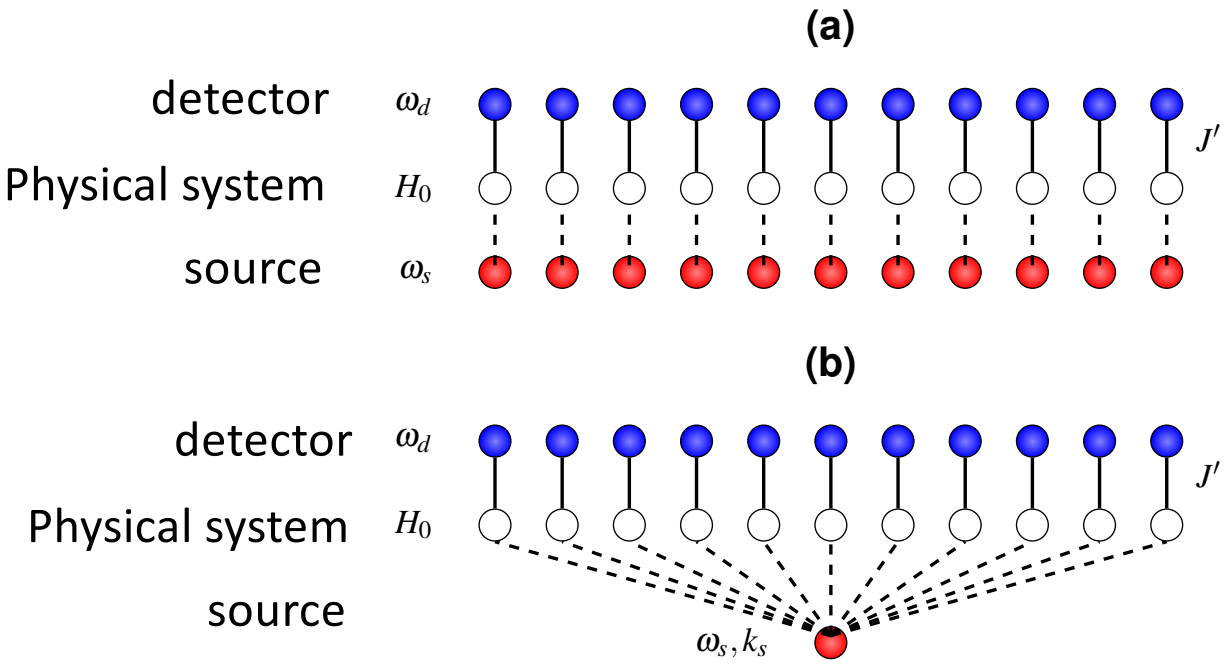
Simulate the actual scattering event, including scattered particles, interactions terms, and solve the Schrödinger Eq.

Results are obtained by measuring energy and momentum of the outgoing particle.

Momentum conservation

Initial state: $|\Psi(t=0)\rangle = |gs\rangle\,|k_0, 0\rangle$

Detection: $n_{2k}(t) = \frac{1}{L}\sum_{\ell,\ell'} e^{ik(\ell-\ell)}\langle c^{\dagger}_{2\ell} c_{2\ell'}(t)\rangle$



"chain-chain" geometry

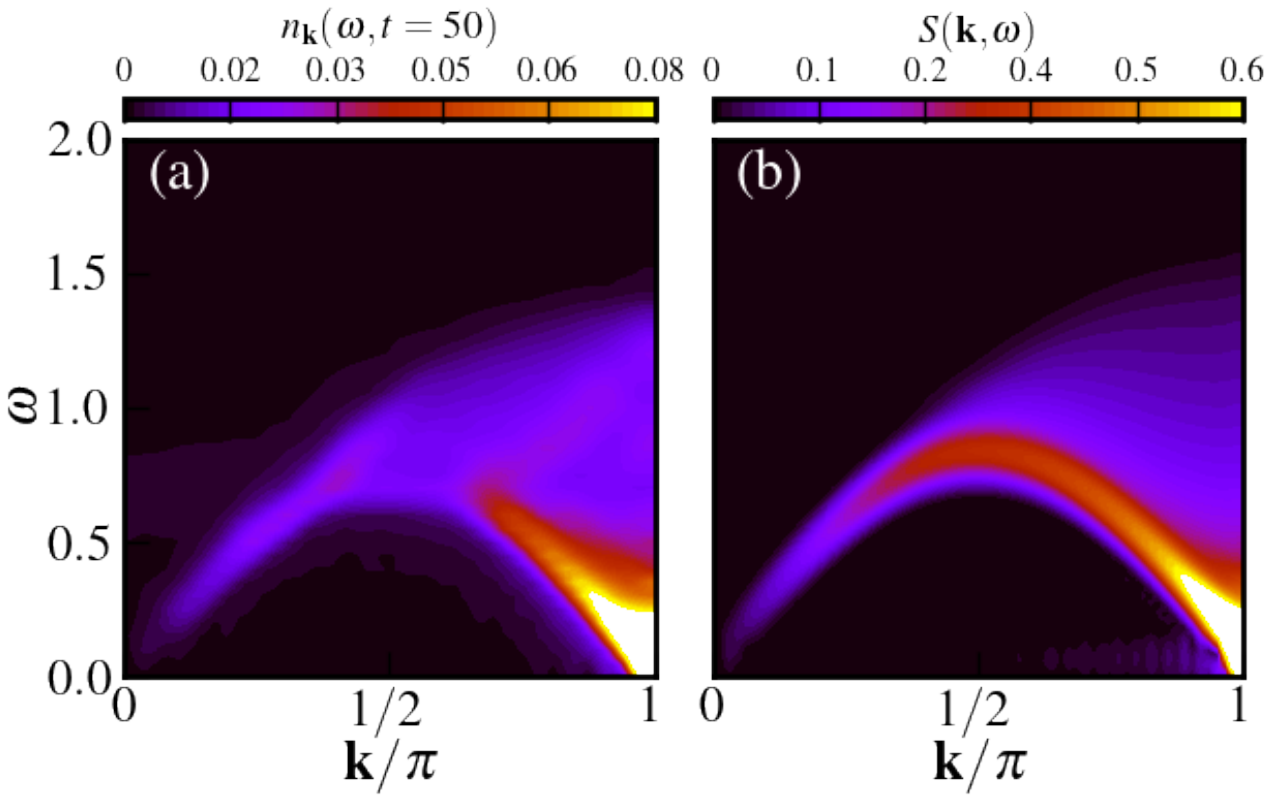
"star-chain" geometry

INS: Heisenberg chain

Time-dependent Density Matrix Renormalization Group solver

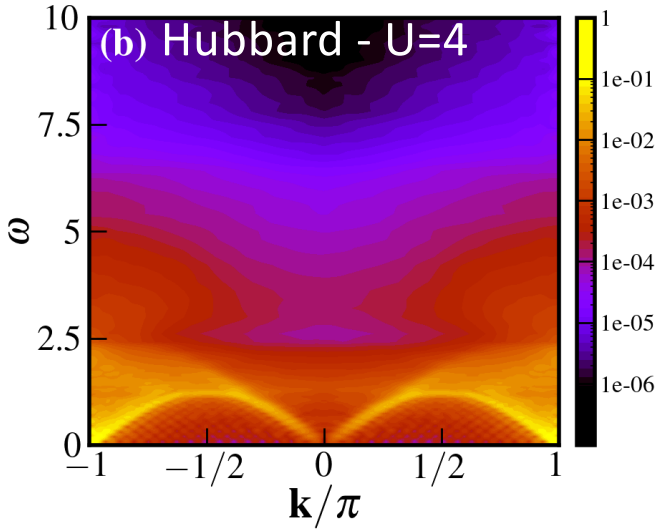
Time-dep. scattering approach

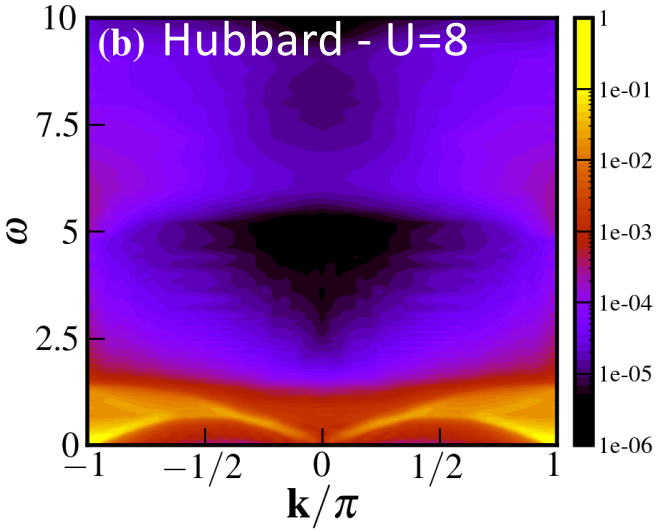
Green's function (perturbation theory)

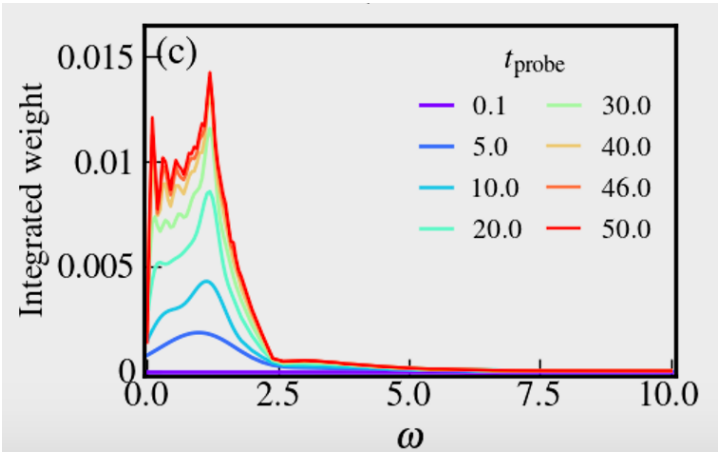


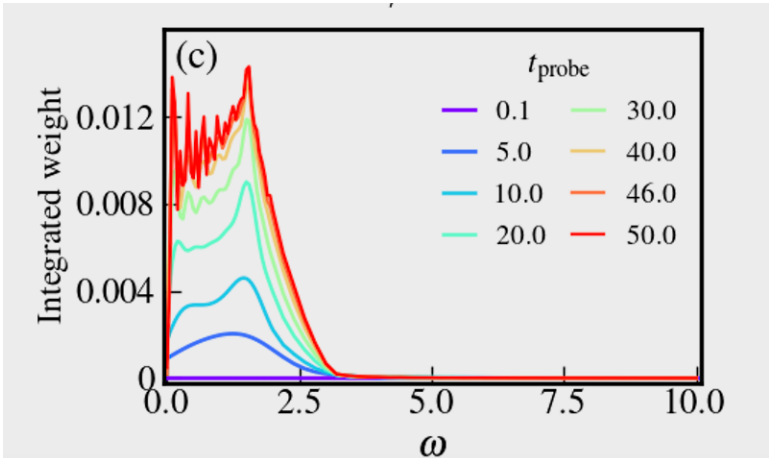
INS: Hubbard chain

Higher order processes couple charge density and spin!









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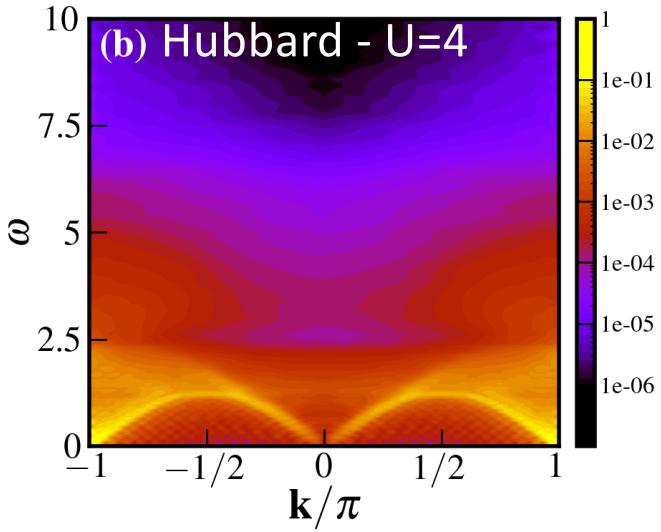
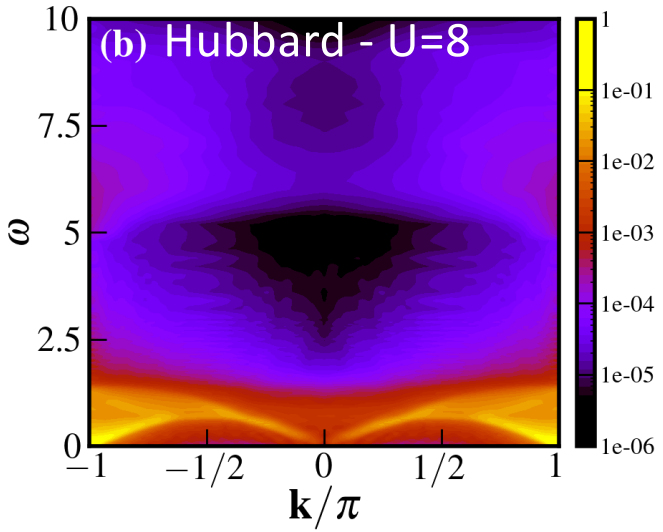
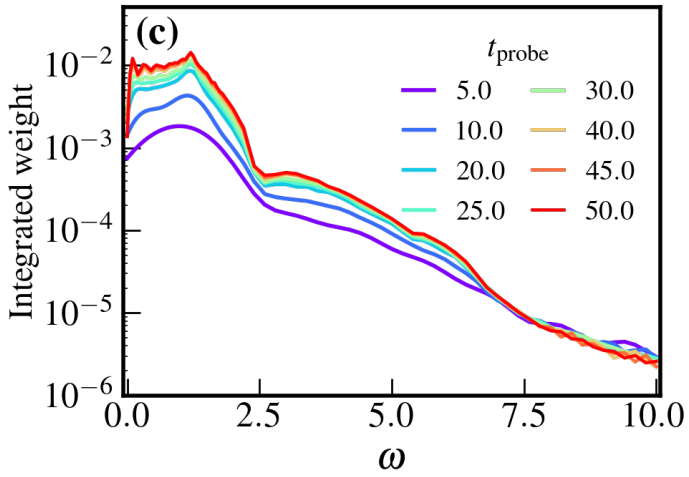
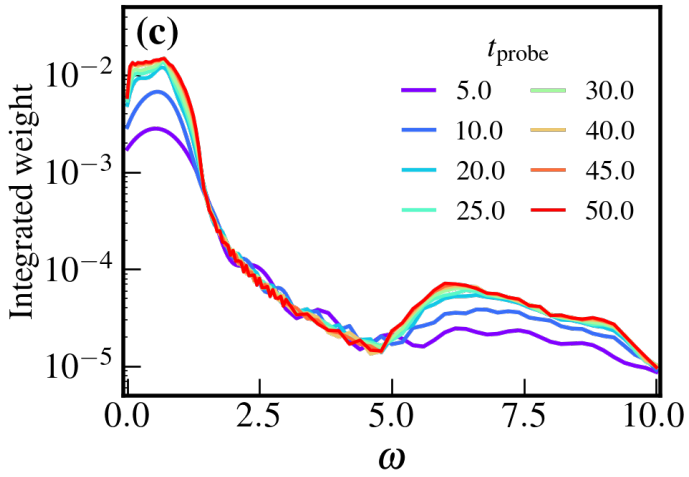


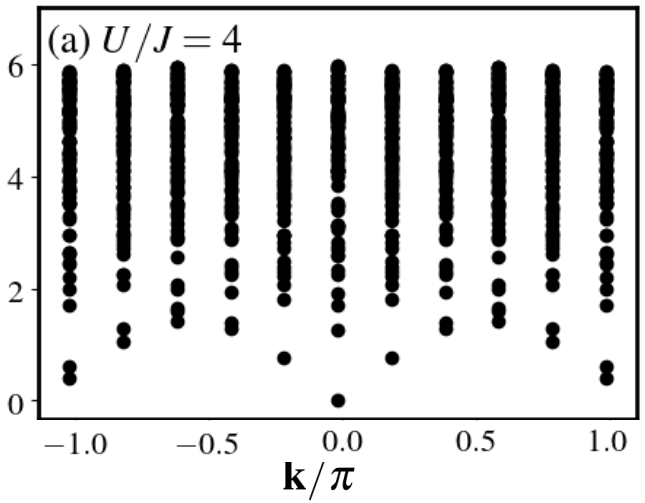


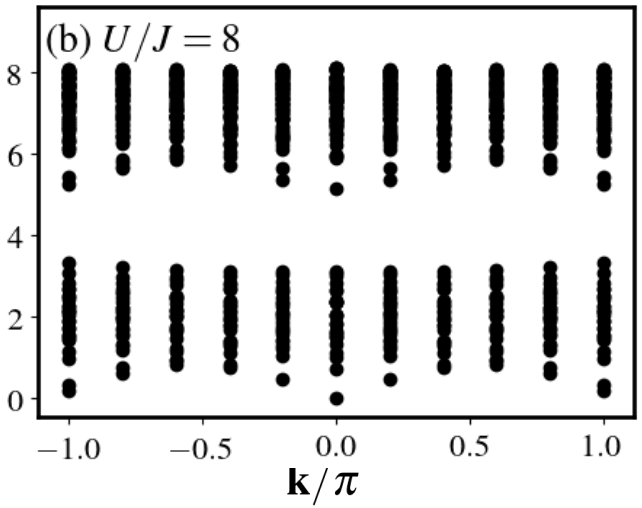


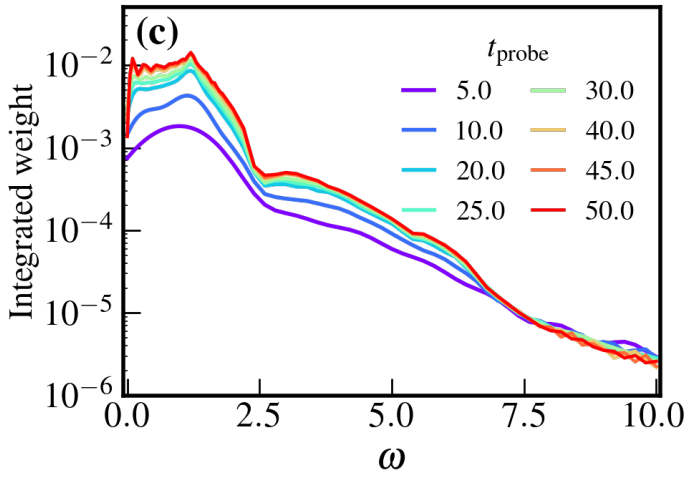


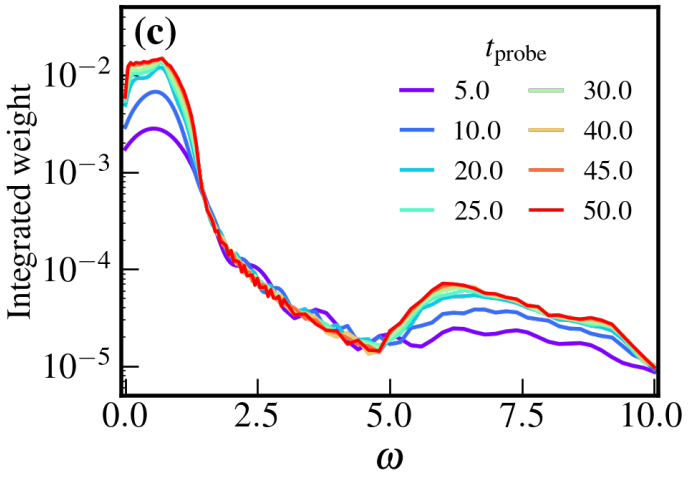
Inelastic neutron scattering

Higher order processes couple charge density and spin!

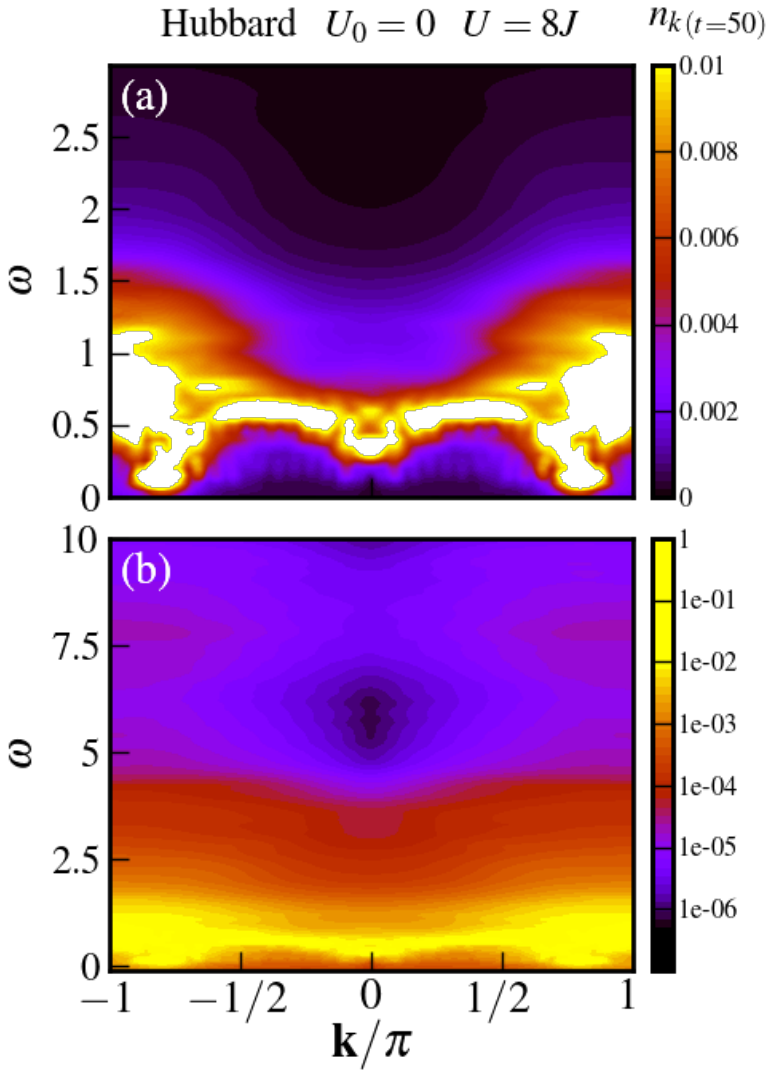






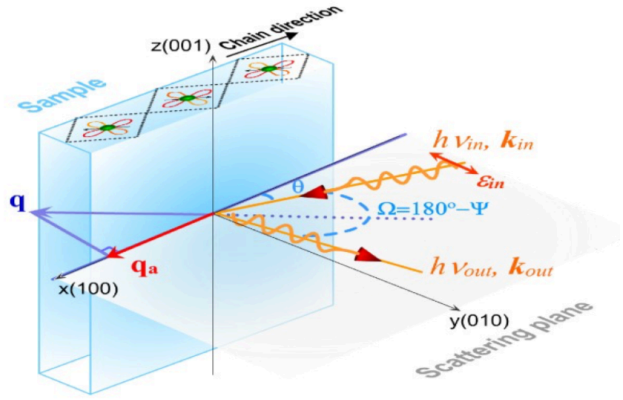


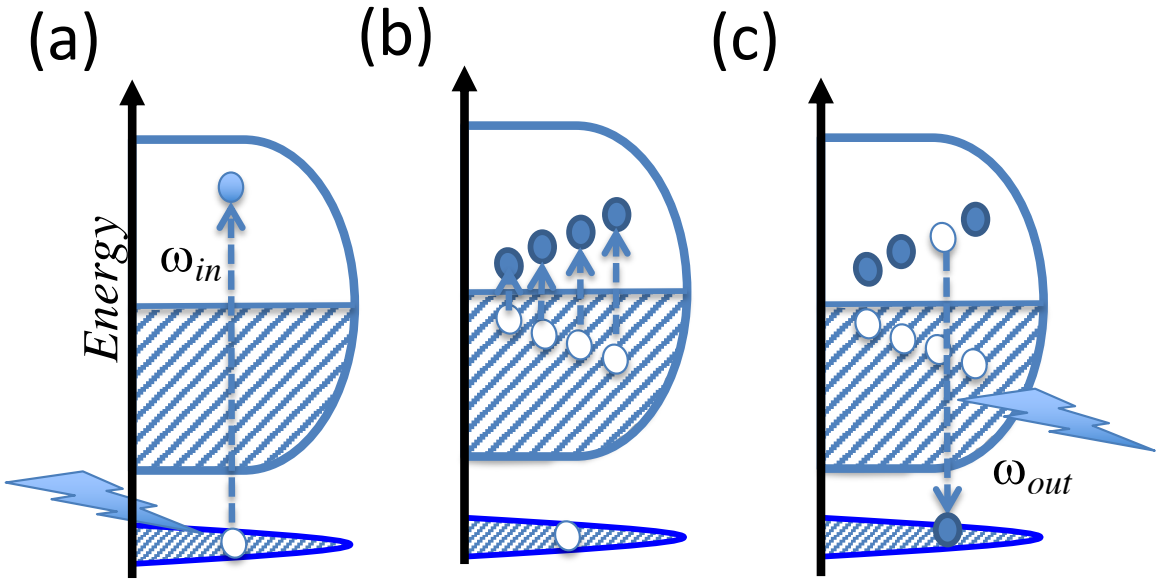
Hubbard chain after a quench



Momentum shift due to “photo-doping”

Core-hole spectroscopies





Resonant inelastic X-ray scattering (RIXS)

- F. de Groot and A. Kotani, Core Level Spectroscopy of Solids, Advances in Condensed Matter Science (CRC Press, 2008).
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- Y. Wang, M. Claassen, C. D. Pemmaraju, C. Jia, B. Moritz, and T. P. Devereaux, Nature Reviews Materials 3, 312 (2018)
- L. J. P. Ament, M. van Veenendaal, T. P. Devereaux, J. P. Hill, and J. van den Brink, Rev. Mod. Phys. 83, 705 (2011).
- J.-P. Rueff and A. Shukla, Journal of Electron Spectroscopy and Related Phenomena 188, 10 (2013), progress in Resonant Inelastic X-Ray Scattering.
- Y. Chen, Y. Wang, C. Jia, B. Moritz, A. M. Shvaika, J. K. Freericks, and T. P. Devereaux, Phys. Rev. B 99, 104306 (2019).
- C. Jia, K. Wohlfeld, Y. Wang, B. Moritz, and T. P. Devereaux, Phys. Rev. X 6, 021020 (2016).

RIXS spectrum

$$H = H_0 + H_c + H_{ph}, \quad H_c = -U_c \sum_i n_{di}(1 - n_{pi}), \quad H_{ph} = \sum_{\mathbf{k},\lambda} \omega_{\mathbf{k}} (b^\dagger_{\mathbf{k},\lambda} b_{\mathbf{k},\lambda} + 1/2)$$

$$V = V_{in} + V_{out}, \quad V_{out} = V_{in}^\dagger$$

$$V_{in} = \sum_{\mathbf{k},\lambda} b_{\mathbf{k},\lambda} D_{\mathbf{k},\lambda} = \sum_{\mathbf{k},\lambda} b_{\mathbf{k},\lambda} \sum_{i,\alpha,\sigma} \left(e^{i\mathbf{k}\cdot\mathbf{R}_i} D_i^\dagger + \text{h.c.} \right) \qquad D^\dagger_{i,\lambda} = \sum_{\alpha,\sigma} \Gamma^\lambda_\alpha d^\dagger_{i,\sigma} p_{i\alpha,\sigma}$$

$$I_{RIXS}(\omega', t) = \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^t dt_1' \int_0^{t_1'} dt_2' \langle \psi | V_{out}(t_1') V_{in}(t_2') b^\dagger_{\mathbf{k}'} b_{\mathbf{k}'} V_{out}(t_2) V_{in}(t_1) | \psi \rangle$$

$$= \sum_f \left| \int_0^t dt_1 \int_0^{t_1} dt_2 e^{-i(\omega + E_0)t_1} e^{-i(\omega' + E_f)t_2} \langle f | D_{\mathbf{k}'} e^{-iH_e(t_2 - t_1)} D^\dagger_{\mathbf{k}} | 0 \rangle \right|^2$$

$$= \sum_f \left| \sum_n \int_0^t dt_1 \int_0^{t_1} dt_2 e^{-i(\omega + E_0 - E_n)t_1} e^{-i(\omega' + E_f + E_n)t_2} \langle f | D_{\mathbf{k}'} | n \rangle \langle n | D_{\mathbf{k}} | 0 \rangle \right|^2$$

$$I_{RIXS}(\omega') = 4\pi^2 \sum_f \left| \sum_n \frac{\langle f | D_{\mathbf{k}'} | n \rangle \langle n | D^\dagger_{\mathbf{k}} | 0 \rangle}{(\omega + E_0 - E_n + i\eta)} \right|^2 \delta(\omega - \omega' + E_0 - E_f)$$

- F. de Groot and A. Kotani, Core Level Spectroscopy of Solids, Advances in Condensed Matter Science (CRC Press, 2008).
- T. P. Devereaux and R. Hackl, Rev. Mod. Phys. 79, 175 (2007).
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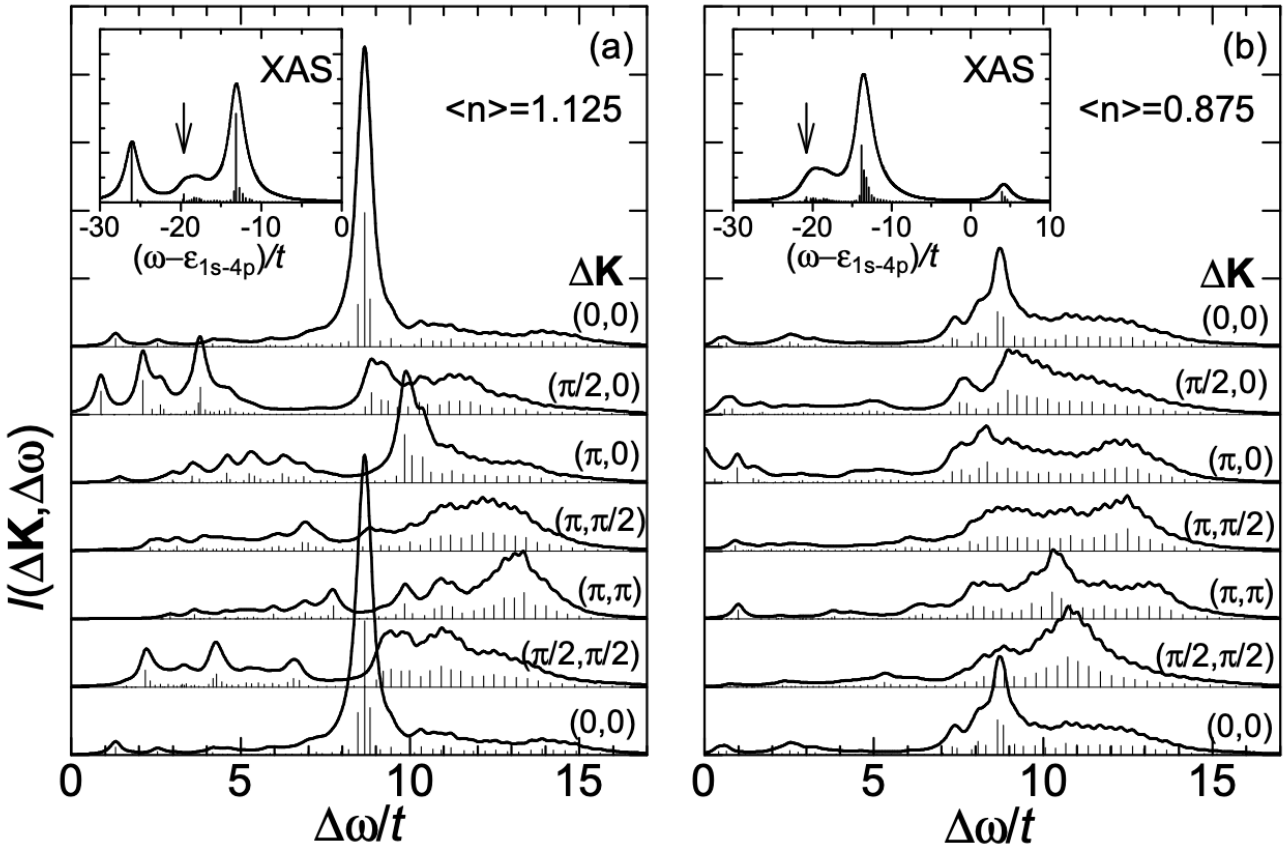
- Y. Chen, Y. Wang, C. Jia, B. Moritz, A. M. Shvaika, J. K. Freericks, and T. P. Devereaux, Phys. Rev. B 99, 104306 (2019).

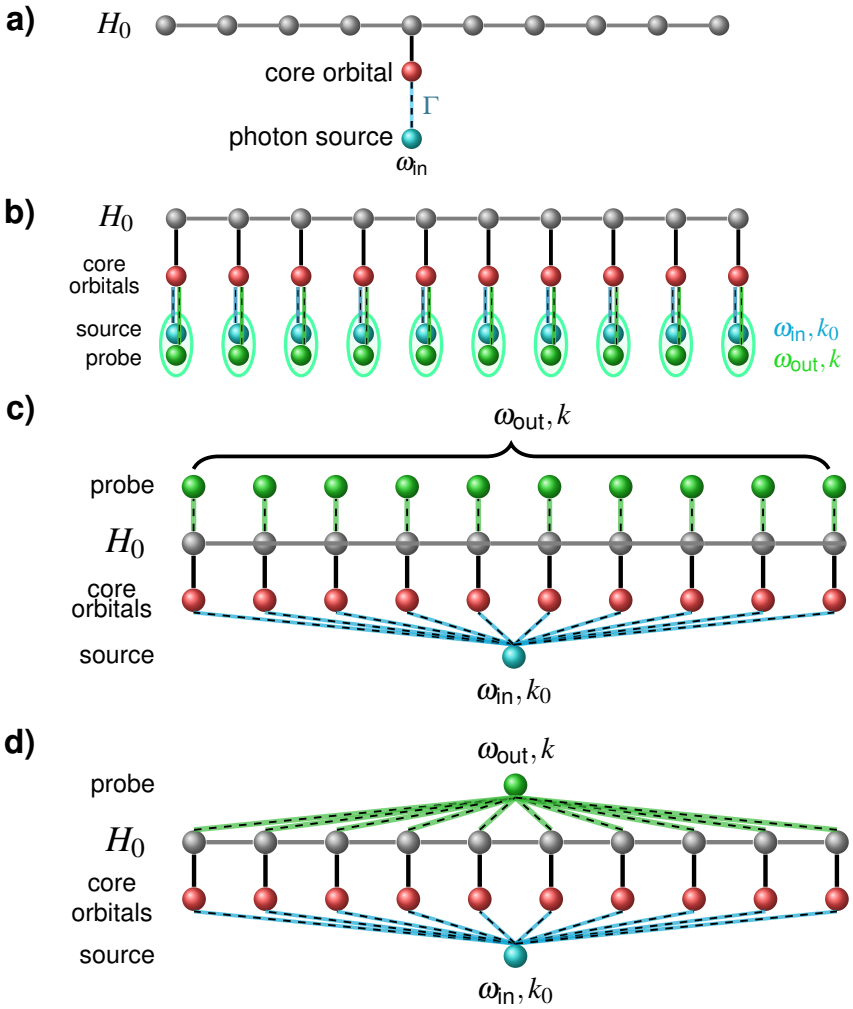
Typical results with ED

2D Hubbard (Indirect RIXS; Cu K edge)



Kenji Tsutsui, Takami Tohyama, and Sadamichi Maekawa
Phys. Rev. Lett. **91**, 117001

Numerical protocols

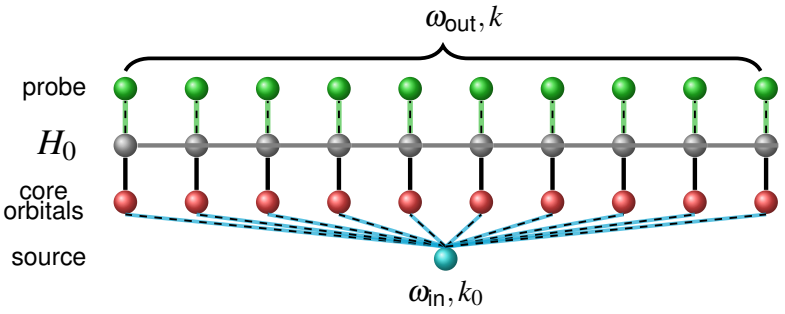


$$H_{source} = |\mathbf{k}_{in}\rangle\langle\mathbf{k}_{in}| + \lambda \sum_{ij} n_{b,s,i} n_{b,s,j}$$

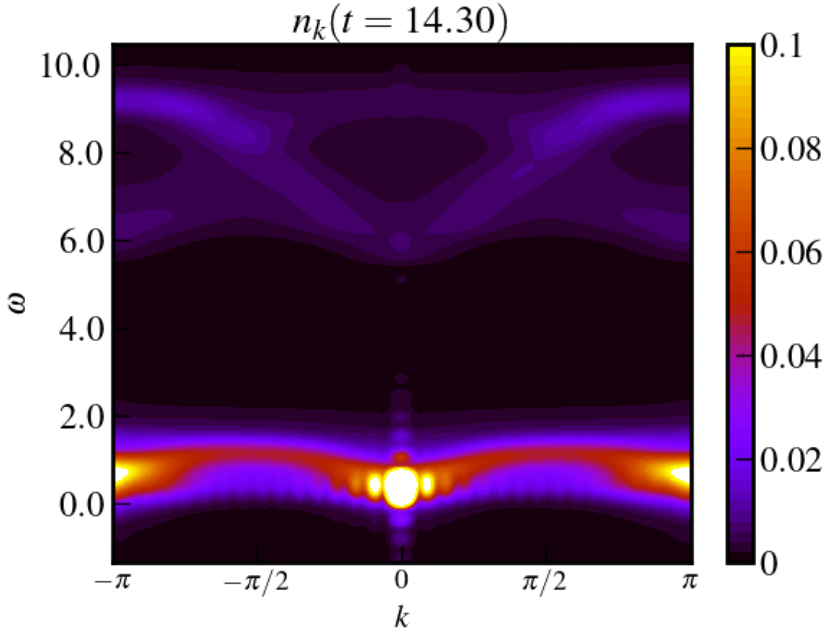
$$V_{in}^{\sigma\sigma'} = \sum_{n} (\Gamma_s^{\sigma\sigma'} \frac{e^{i\mathbf{k}_{in}\cdot\mathbf{R}_n}}{\sqrt{N}} b_s + \Gamma_d^{\sigma\sigma'} b_{d,n}) d_{n\sigma'}^{\dagger} p_{n\sigma}$$

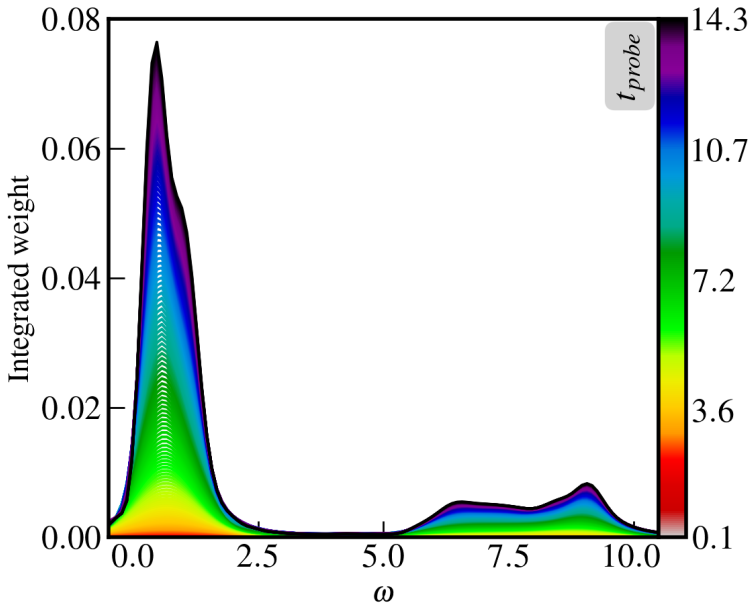
Momentum resolution

L=32, U=8, U_c=1.5, Γ=0.1, ω_0=4.47

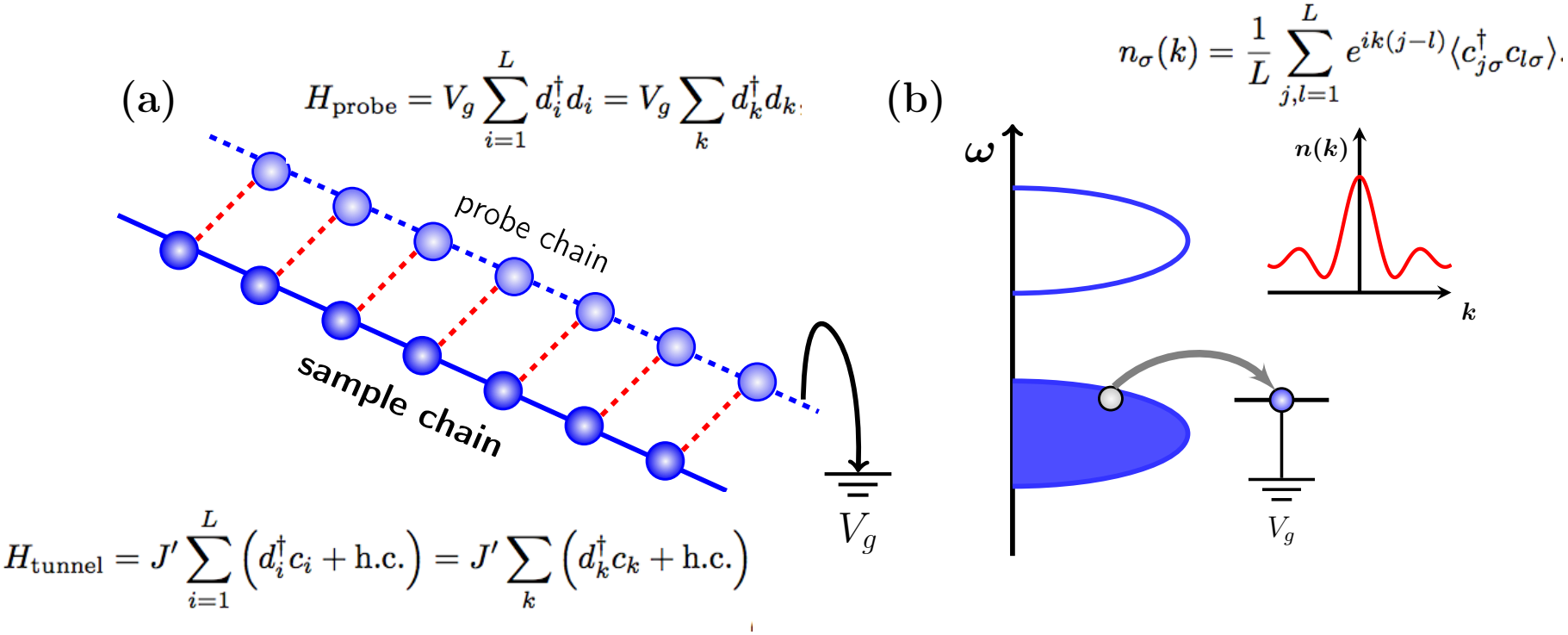


In the DMRG context this means one time-dependent run for each value ω (dozens, if not hundreds)





Time dependent ARPES



(a) Proposed tunneling setup: a sample chain is connected to a probe chain via a tunneling barrier. The probe chain is set at a target gate voltage Vg. (b) Particles tunnel for a short period of time to the probe chain, where their momentum distribution n(k) is measured.

See also: G. Cohen, E. Gull, D. R. Reichman, and A. J. Millis, Phys. Rev. Lett. 112, 146802 (2014); A. Kantian, U. Schollwoeck, and T. Giamarchi, Phys. Rev. Lett. 115, 165301 (2015).

Non-interacting toy example

$$H_0 = -J \sum_{i=1,\sigma}^{L-1} \left(c_i^\dagger c_{i+1} + \text{h.c.} \right) = \sum_k \omega_k c_k^\dagger c_k$$

$$H_{\text{tunnel}} = J' \sum_{i=1}^{L} \left(d_i^\dagger c_i + \text{h.c.} \right) = J' \sum_k \left(d_k^\dagger c_k + \text{h.c.} \right)$$

$$H_{\text{probe}} = V_g \sum_{i=1}^{L} d_i^\dagger d_i = V_g \sum_k d_k^\dagger d_k$$

$$H = \begin{bmatrix} \omega_k & -J \\ -J & V_g \end{bmatrix}$$

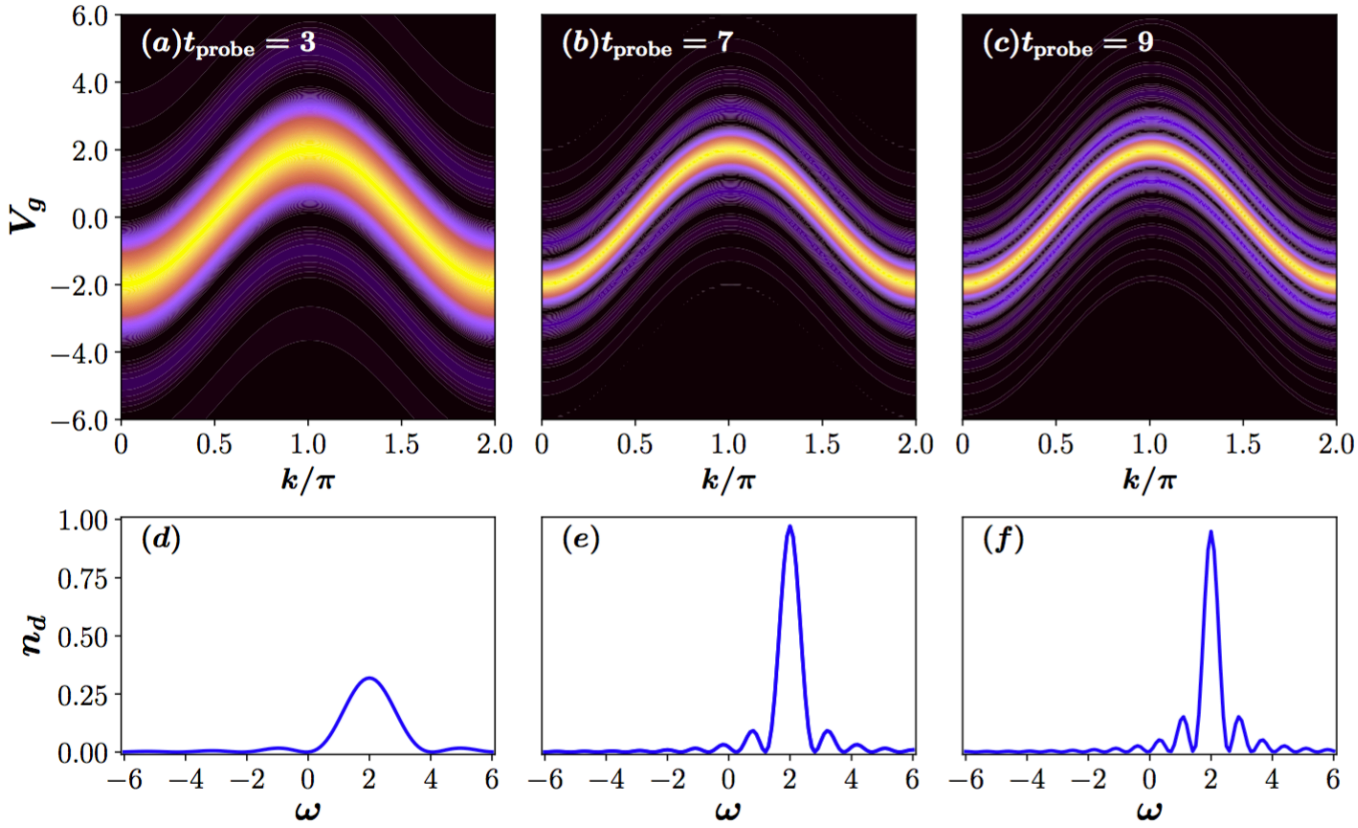
Solution in any QM book

$$\begin{aligned} \langle 01|t\rangle &= -i\langle 01|V|10\rangle \int_0^t dt'\, e^{i(E_f - E_i)t'} \\ &= -iJ \left[\frac{e^{i(E_f - E_i)t} - 1}{i(E_f - E_i)} \right] \\ &= -iJ e^{i(E_f - E_i)t/2} \frac{2\sin\left((E_f - E_i)t/2\right)}{i(E_f - E_i)}, \end{aligned}$$

$$n_d(t) = J^2 \left[\frac{4\sin^2\left((V_g - \omega)t/2\right)}{(V_g - \omega)^2} \right],$$

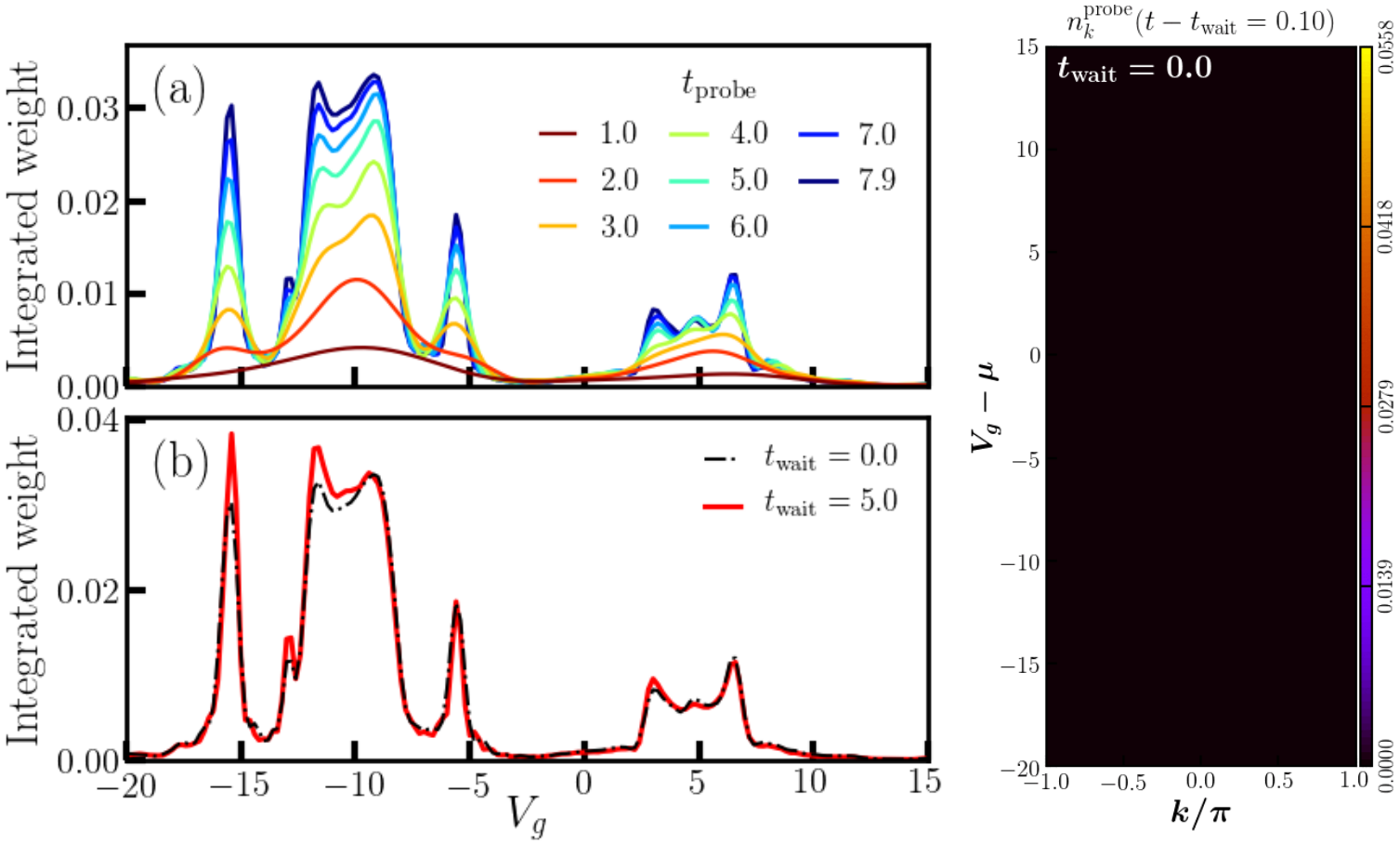
Non-interacting toy example

$$H = \begin{bmatrix} \omega_k & -J \\ -J & V_g \end{bmatrix}$$



Evolution with probing time

Hubbard model, n=1. Quench from U=2, V=0 to U=20, V=5

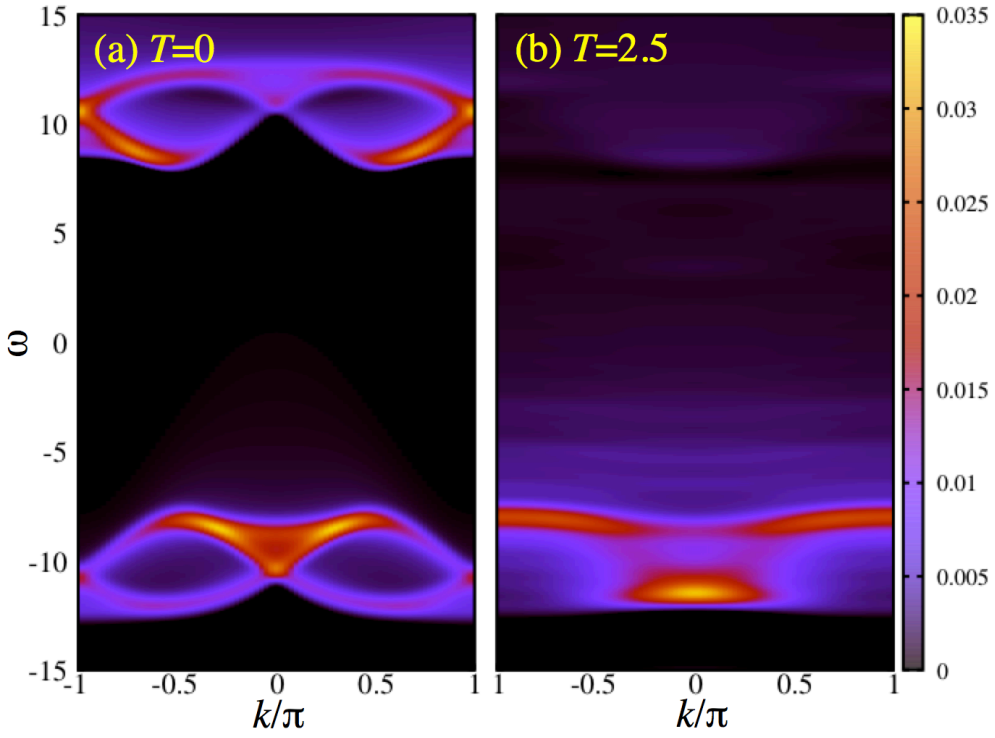


(a) Integrated spectral weight as a function of Vg for twait = 0 and different probe times, demonstrating the resolution improvement.
(b) Same as (a) but for twait = 0 and 5, at the final tprobe = 7.9.

Equilibrium and non-equilibrium spectra:

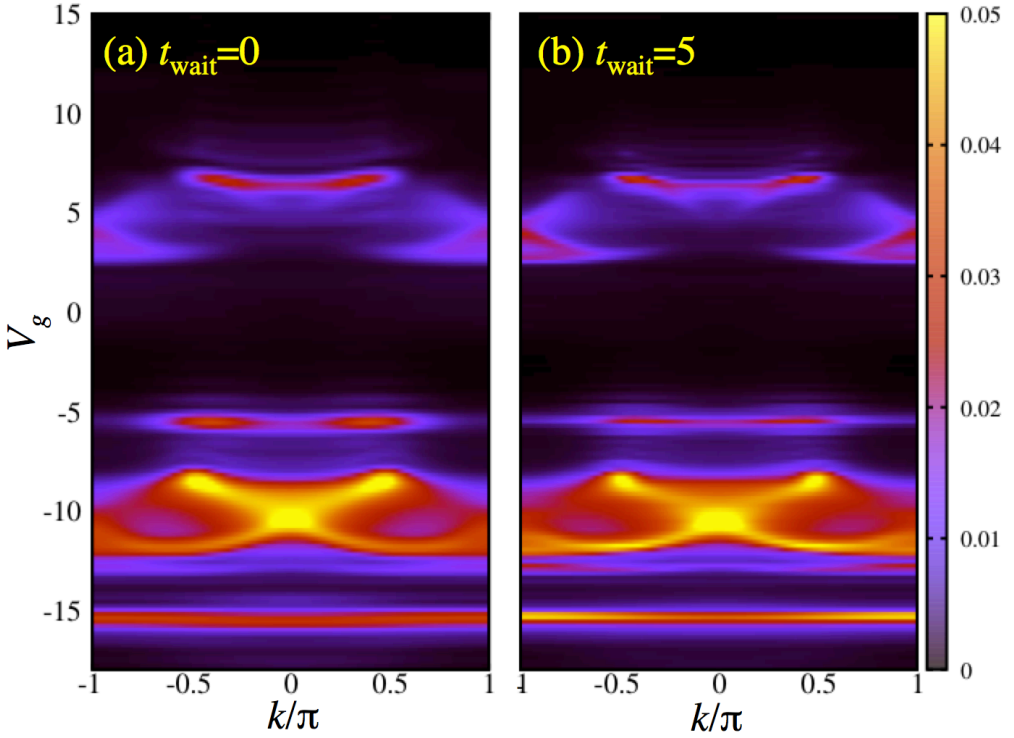
Hubbard model (n=1, U=20, V=5)

Finite temperature



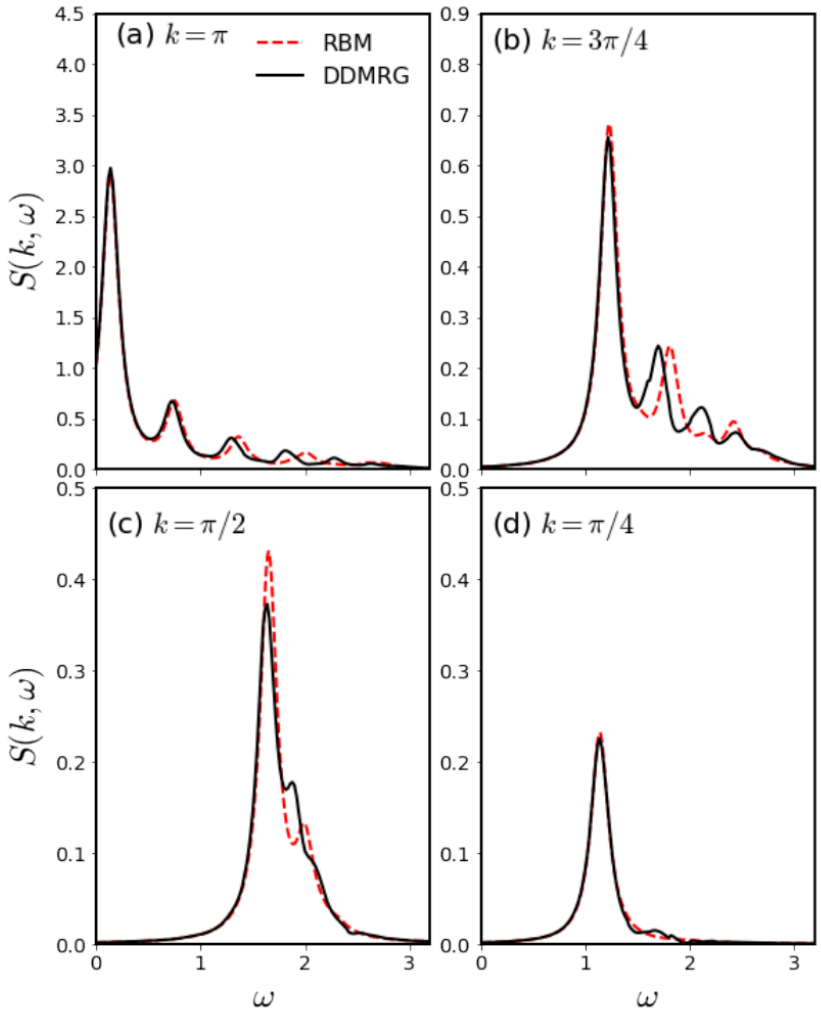
Momentum resolved spectrum of the 1D extended Hubbard model at half-filling at (a) zero temperature, where negative(positive) frequencies correspond to occupied(empty) states, and (b) T = 2.5J, obtained with the tDMRG method for a chain of length L = 40, and interaction U = 20, V = 5.

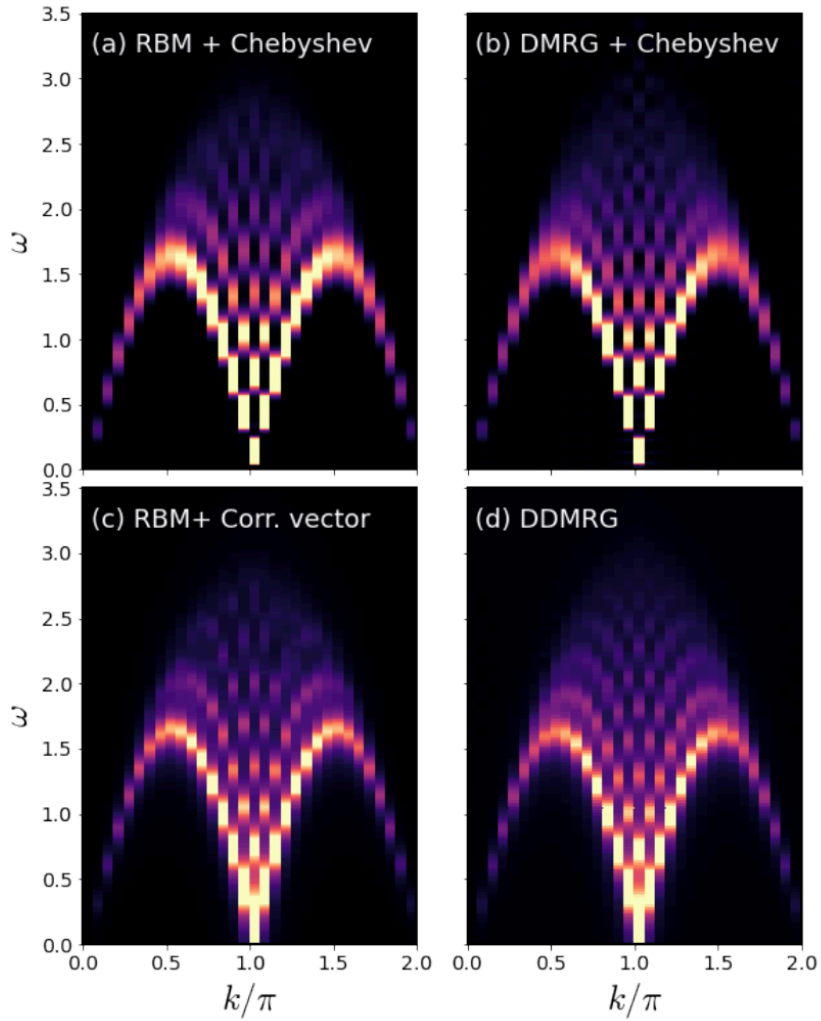
Quench from U=2, V=0



Momentum resolved tunneling spectrum of the 1D extended Hubbard model at half-filling after a sudden quench in the interactions from U0 = 2, V0 = 0 to U = 20, V = 5, obtained with the tDMRG method for a chain of length L = 32 a time (a) twait = 0 and (b) twait = 5 after the quench, and a probe time tprobe = 7.9

Chebyshev expansion of spectral functions using restricted Boltzmann machines





Douglas Hendry, Hongwei Chen, Phillip Weinberg, AEF, **arXiv:2103.08804**

Conclusions

- We have a devised a time-dependent scattering approach to calculate spectral information at and away from equilibrium, overcoming all the limitations of ED.
- We extended these ideas to core-hole spectroscopies. It requires to include the core orbitals, and can be used to obtain all scattering channels, with and without spin-orbit.
- The method can readily be extended to obtain momentum resolution and to multi-band problems.
- It can also be applied to study Raman scattering and non-linear optical processes such as in two/three-photon spectroscopies.
- In the context of inelastic scattering, higher order processes yield information about the charge fluctuations.
- It can be implemented within a DMFT framework.

K. Zawadzki and AEF, Phys. Rev. B 100, 195124 (2019)
K. Zawadzki , A. Nocera and AEF, arXiv:2002.04243
K. Zawadzki and AEF, Phys. Rev. B, 102, 235141 (2020)