Exploring two dimensional coherent spectroscopy

arXiv: 2401.17266 & arXiv: 2405.14954 Yoshito & Ciarán

Motivation: Probe the non-linear response of quantum magnets (using THz spectroscopy) to access their intrinsic excitations.

broad, featureless continuum in lin. spec.

- fractional excitations (spinons, Majorana fermions, ...)

"hidden" excitations in linear spectroscopy

- multipolar excitations (no restriction to dipolar selection rules)
- interplay of excitations

$\langle S_i^\alpha(t_1) S_j^\beta(0)\rangle$

$\langle S_i^\alpha(t_2) S_j^\beta(t_1) S_k^\gamma(0)\rangle$

- non-linear susceptibilities & higher-order dynamical correlation fct.

Quadrupolar excitations (as a principal example):

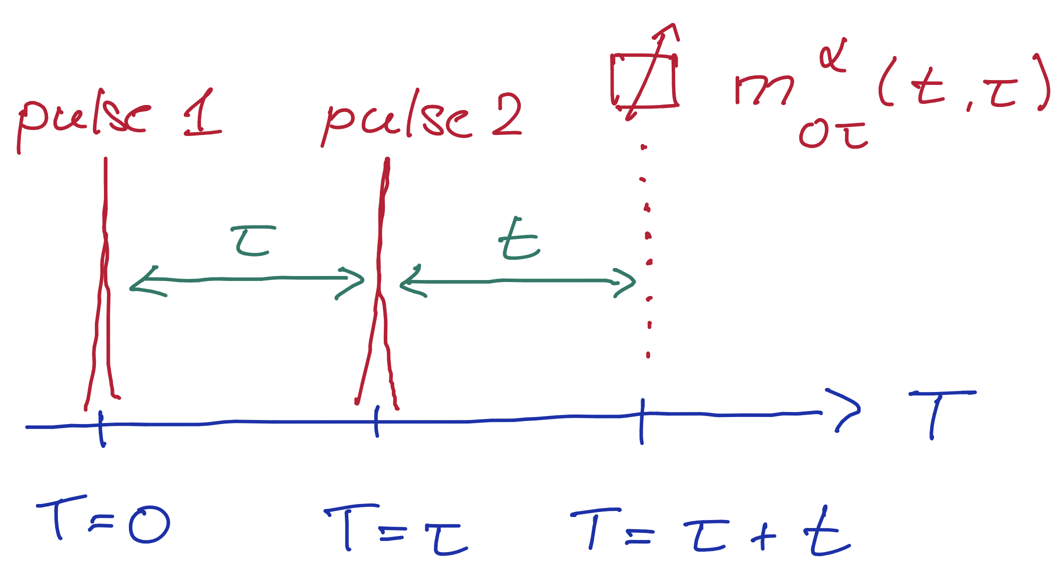
Let's go beyond conventional dipolar excitations such as magnons, acknowledging that most experimental probes obey dipolar selection rules.

Spin-1 systems exhibit multipolar excitations with $\Delta m^2 = 2$ on-site. For a spin-½ system this is not possible, but $\Delta m^2 = 2$ can arise only for two excitations (magnons) at different sites.

Experimentally, multipolar excitations are therefore almost invisible. But some recent progress

- hybridize quadrupolar + dipolar excitations
- higher-order processes (RIXS experiments)
- 2D coherent spectroscopy.

2d coherent spectroscopy (2DCS)



THz pulse (K. Nelson 2017, P. Armitage 2019)
$\Rightarrow$ proper energy scale for (quantum) magnets

$$m^{\alpha}_{NL}(t,\tau) = m^{\alpha}_{0\tau}(t,\tau) - \overbrace{m^{\alpha}_{0}(t,\tau) - m^{\alpha}_{\tau}(t,\tau)}^{\text{single-pulse magnetization}}$$

2nd order non-linear susceptibility

$$= \underline{B_0}\, B_\tau\, \chi^{(2)}_{\alpha\alpha\alpha}(t,\tau)$$

only cross-terms

$$\left.\begin{aligned} &+ \underline{(B_0)^2 B_\tau}\; \chi^{(3,1)}_{\alpha\alpha\alpha\alpha}(t,\tau,0) \\ &+ \underline{B_0 (B_\tau)^2}\; \chi^{(3,2)}_{\alpha\alpha\alpha\alpha}(t,0,\tau) \end{aligned}\right\} \text{objects of interest}$$

$$+ \ \ldots$$

$\chi^{(3,1)} \rightarrow$ quadrupolar excitations

$\chi^{(3,2)} \rightarrow$ fractional excitations

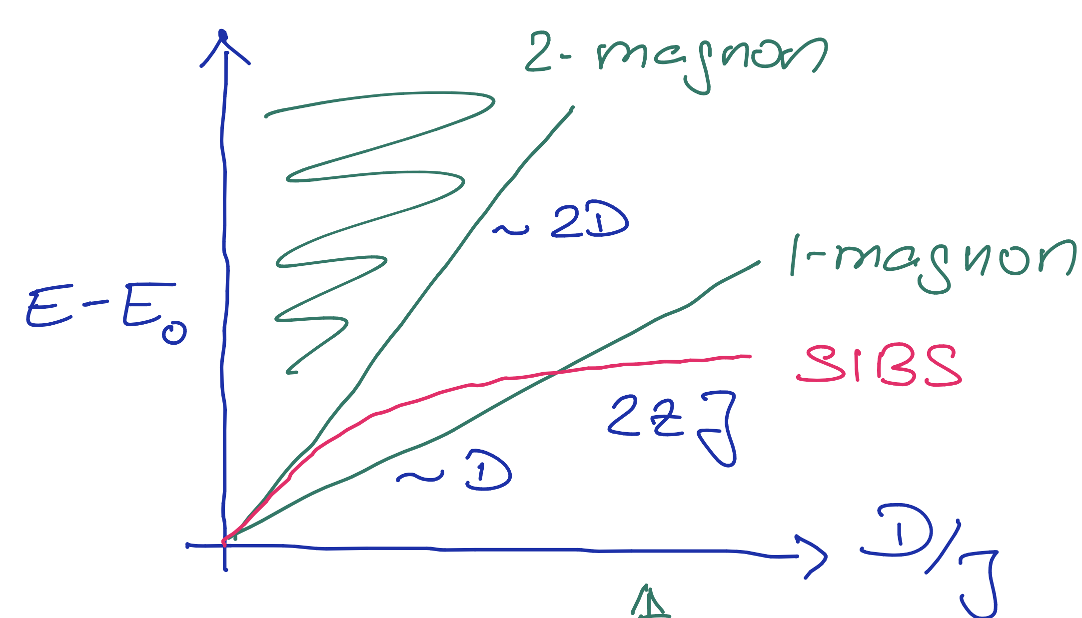
Models Spin-1 FM Heisenberg model w/ single-ion anisotropy

$$\mathcal{H} = -\underbrace{J}_{J>0 \text{ FM}} \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j - \underbrace{D}_{D>0 \text{ single-ion anisotropy}} \sum_i (S_i^z)^2$$

$U(1) \times \mathbb{Z}_2$ symmetry

$\Rightarrow$ ground state is a fully polarized ferromagnet

↑↑ • ↑↑	↑ • ↑ • ↑	↑↑↓↑↑
1-magnon	2-magnon	single-ion bound state SIBS



z coordination number of lattice

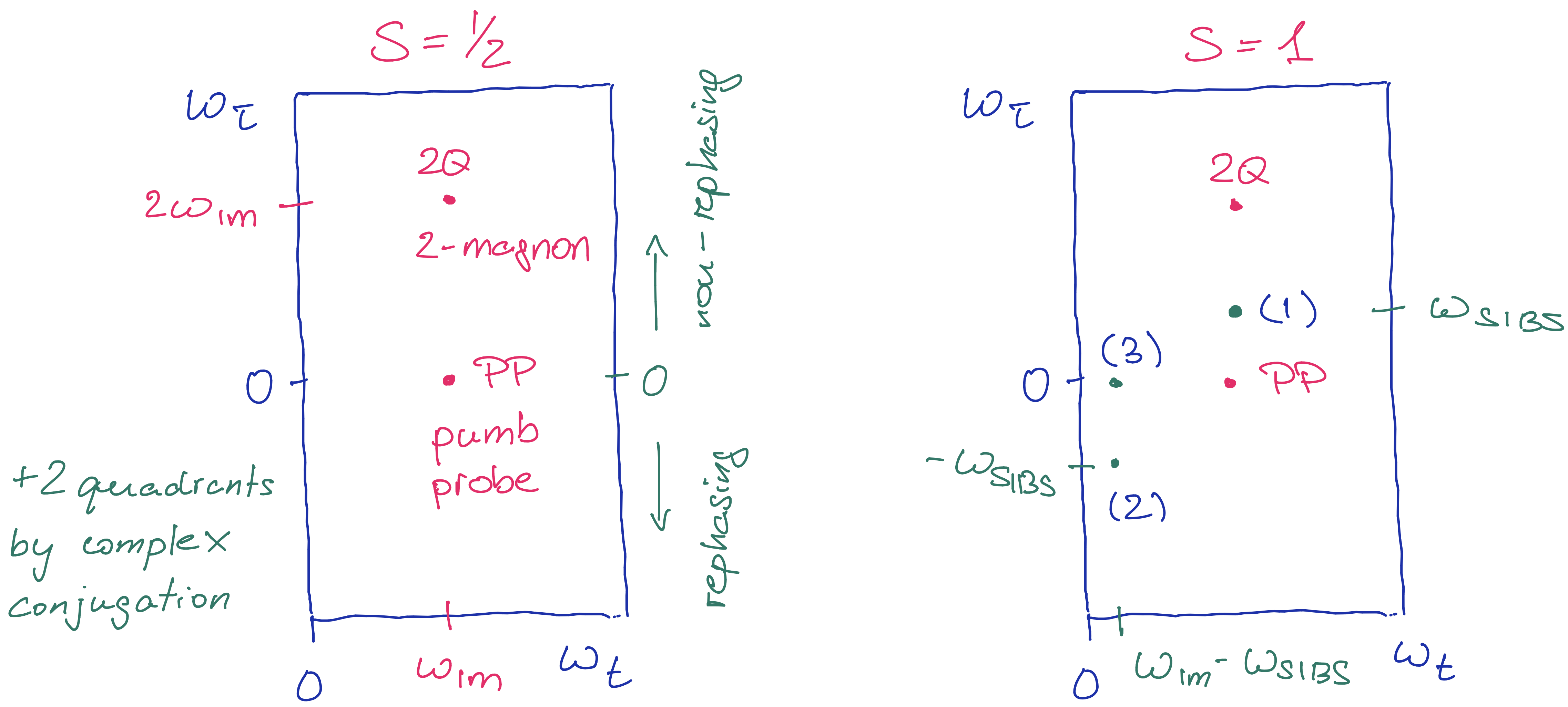
linear response sees only one excitation

Spin-½ XXZ model with longitudinal field

$$\mathcal{H} = -J \sum_{\langle ij \rangle} \underbrace{\gamma}_{\gamma/J = 1.1} (S_i^x S_j^x + S_i^y S_j^y) + S_i^z S_j^z - \underbrace{h_z}_{h_z/J = 5} \sum_i S_i^z$$

$\Rightarrow$ ground state is also a fully polarized ferromagnet with 1-magnon ($\omega_{1m} = h_z$) and 2-magnon excitations.

2DCS signatures $\chi^{(3,1)}_{xxxx}$



$\Rightarrow$ 3 additional peaks!

microscopic origin of peak (1)

$T = 0$ 2x field $M^x(0)\,M^x(0)$ $\rightarrow$ SIBS excitation

$\downarrow$ $\omega_\tau = \omega_{SIBS}$

$T = \tau$ 1x field $M^x(\tau)$ $\rightarrow$ SIBS to 1-magnon

$\downarrow$ $\omega_t = \omega_{1m}$

$T = \tau + t$ measurement $M^x(\tau + t)$ $\rightarrow$ back to ground state

$$\langle GS|\, \underbrace{M^x(\tau+t)}_{GS}\, \underbrace{M^x(\tau)}_{\substack{\text{1-magnon} \\ \omega_t = \omega_{1m}}}\, \underbrace{M^x(0)\,M^x(0)}_{\substack{\text{SIBS} \\ \omega_\tau = \omega_{SIBS}}}\, |GS\rangle$$

peak (2)

$$\langle GS|\, \underbrace{M^x(0)\,M^x(0)}_{\substack{\text{SIBS} \\ \omega_\tau = -\omega_{SIBS}}}\, M^x(\tau+t)\, \underbrace{M^x(\tau)}_{\substack{\text{1-magnon} \\ \omega_t = \omega_{1m} - \omega_{SIBS}}}\, |GS\rangle$$

peak (3)

$$\langle GS | \underbrace{M^x(0)}_{\text{1-magnon}} \underbrace{M^x(\tau)}_{\text{SIBS}} M^x(\tau+t) \underbrace{M^x(0)}_{\text{1-magnon}} | GS$$

$\omega_t = \omega_{1m} - \omega_{SIBS}$ $\qquad$ $\omega_\tau = \omega_{1m} - \omega_{1m} = 0$

pumb-probe peak (dipolar)

+ 2 more processes

$$\langle GS | \underbrace{M^x(\tau+t)}_{\text{1-magnon}} M^x(\tau) \underbrace{M^x(0) M^x(0)}_{\text{0-magnon}} | GS \rangle$$

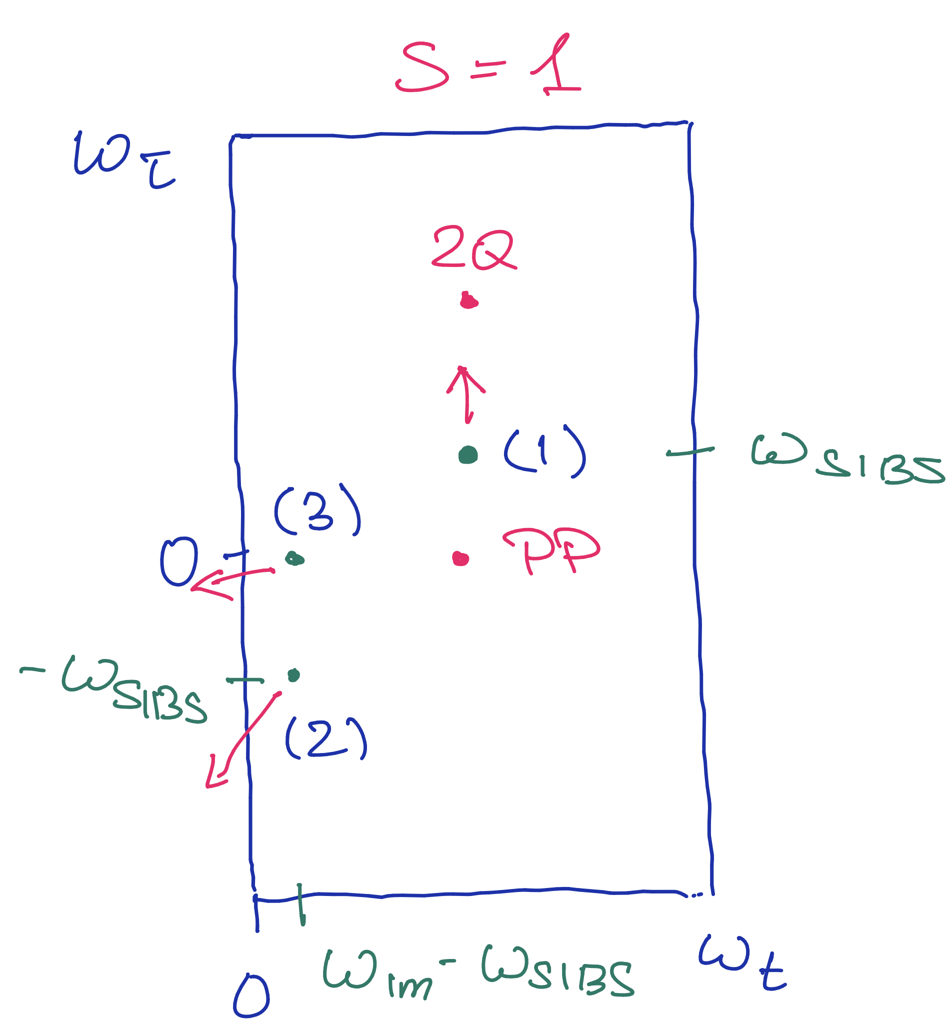
$\omega_t = \omega_{1m}$ $\qquad$ $\omega_\tau = 0$

2Q peak (dipolar)

$$\langle GS | \underbrace{M^x(\tau+t)}_{\text{1-magnon}} M^x(\tau) \underbrace{M^x(0) M^x(0)}_{\text{2-magnon (independent)}} | GS \rangle$$

$\omega_t = \omega_{1m}$ $\qquad$ $\omega_\tau \approx 2\omega_{1m}$

As we vary the single-ion anisotropy these peaks will start to shift



What about the other 3rd order susceptibility ?

$\chi^{(3,2)}$ $\longrightarrow$ access to quadrupolar weight.

How did you do these calculations ?

- numerical exact diagonalization for $L=100$
  - finite-size effects, comparison to MPS/DMRG arXiv: 2401.17266
- analytical generalized spin-wave theory
- semi-analytical 2-kink and 4-kink calculations
  - spinons in 1D TFIM ($\rightarrow$ Johannes' work)

What about materials & experiment ?

$NiNb_2O_6$ spin-1 chain

$D/J = 0.3$ SIBS $\approx$ 2-magnon
hard to discern

FeI_2 easy-axis triangular magnet

neutron scattering (hybridized SIBS)

SIBS is well separated from 2-magnon continuum

$\rightarrow$ identify quadrupolar nature via 2DCS

1-2-20 compounds
intermetallics

non-Kramers doublets in $Pr(Ti,V,Ir)_2(Al,Zn)_{20}$

quadrupolar & octupolar doublets

$\rightarrow$ no dipolar excitations $\rightarrow$ 2DCS

Outlook & afternoon discussions

- generalized spin-wave theory & 4-kink calculations — Cristian, David & Yoshito, Johannes

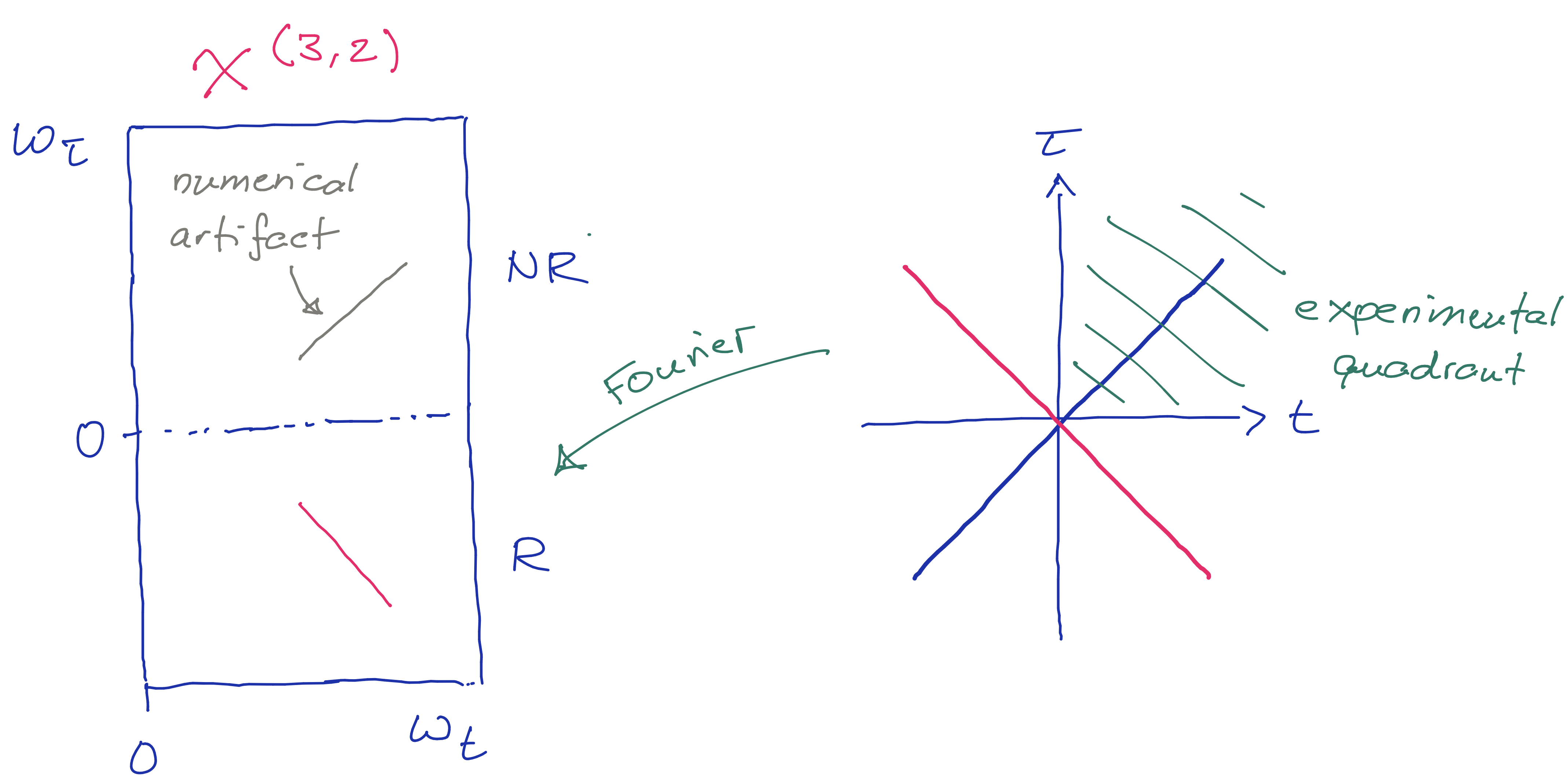
- computational approaches

  ED vs. DMRG vs. SUNNY

  finite-temperature extensions

- fractionalized excitations — Peter & Johannes, Nandini & Wolfram

  spinons ⊕ Majoranas



- materials

  \+ theory-experiment collaboration — Peter & Ciarán