

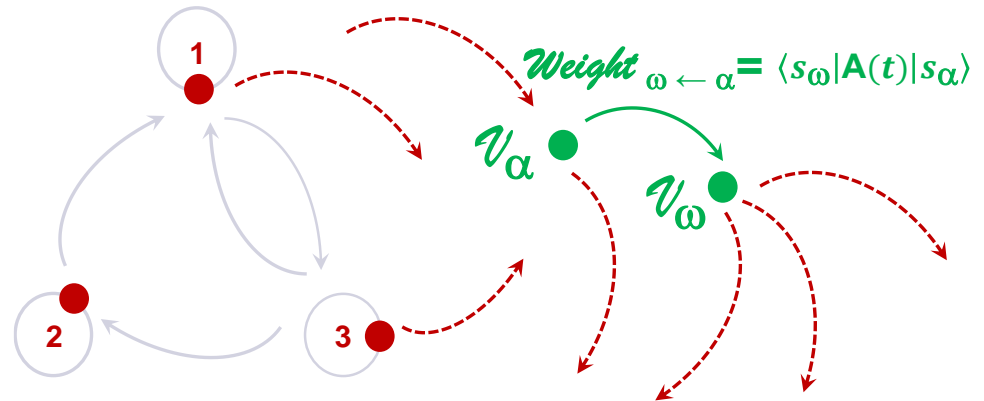
A New Approach of Ordered Exponential in NMR: the Path-Sum

C. Bonhomme¹, P.-L. Giscard²

¹ Laboratoire de Chimie de la Matière Condensée de Paris, Sorbonne Université, Paris, France

² Laboratoire Joseph Liouville, Université du Littoral Côte d'Opale, Calais, France

christian.bonhomme@upmc.fr



60th Experimental Nuclear Magnetic Resonance Conference

April 7 - 12, 2019

Asilomar Conference Center, Pacific Grove, California

General context – The evolution operator $U(t)$

Dyson time-ordering operator

$$U(t', t) = \text{OE}[-i H(t', t)] = \mathbf{T} \exp\left(-i \int_t^{t'} H(\tau) d\tau\right)$$

$$U(\tau_c) = \exp\left(-i\tau_c \sum_{n=0}^{\infty} \overline{H^{(n)}}\right)$$

Magnus

$$\overline{\hat{H}} = {}^{(0)}\hat{H} - \frac{1}{2} \sum_{n \neq 0} \frac{[{}^{(-n)}\hat{H}, {}^{(n)}\hat{H}]}{n\omega_m} + \frac{1}{2} \sum_{n \neq 0} \frac{[[{}^{(n)}\hat{H}, {}^{(0)}\hat{H}], {}^{(-n)}\hat{H}]}{(n\omega_m)^2}$$

Floquet

$$+ \frac{1}{3} \sum_{k, n \neq 0} \frac{[{}^{(n)}\hat{H}, [\hat{H}, {}^{(-n-k)}\hat{H}]]}{kn\omega_m^2} + \dots$$

G. Floquet, *Ann. Sci. Ecole Norm. Sup.*, 1883

F.J. Dyson, *Phys. Rev.*, 1949

W. Magnus, *Pure Appl. Math.*, 1954

F. Fer, *Bull. Classe Sci. Acad. Roy. Bel.*, 1958

J.H. Shirley, *Phys. Rev.*, 1965

U. Haebleren, J.S. Waugh, *Phys. Rev.*, 1968

M.M. Maricq, *Phys. Rev.*, 1982

S. Vega, E.T. Olejniczak, R.G. Griffin, *J. Chem. Phys.*, 1984

I. Scholz, B.H. Meier, M. Ernst, *J. Chem. Phys.*, 2007

M. Leskes, P.K. Madhu, S. Vega, *Progress in NMR Spect.*, 2010

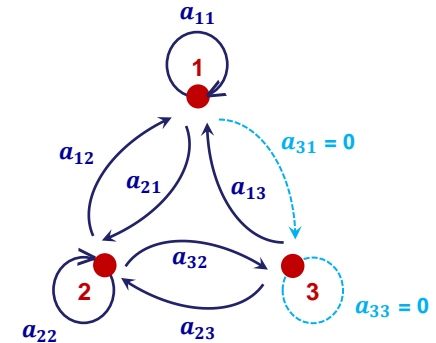
M. Goldman, P. J. Grandinetti, A. Llor *et al.*, *J. Chem. Phys.* 1992

E.S. Mananga, *Solid State NMR*, 2013

K. Takegoshi, N. Miyazawa, K. Sharma, P. K. Madhu, *J. Chem. Phys.*, 2015

...

■ Basic results of algebraic graph theory

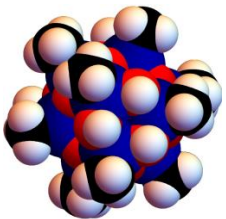


■ Path-Sum applied to Ordered Exponential (OE)

$$\text{OE}[\mathbf{A}](t', t) = \begin{pmatrix} \int_t^{t'} G_{K_2,11}(t', \tau) d\tau & \text{OE}_{12}(t', t) \\ \text{OE}_{21}(t', t) & \int_t^{t'} G_{K_2,22}(t', \tau) d\tau \end{pmatrix}$$

■ Applications:

- ▶ Circularly polarized excitation
- ▶ Linearly polarized excitation, Bloch-Siegert (BS) effect
- ▶ N spins: homonuclear dipolar Hamiltonian, H_D



Basic results of algebraic graph theory

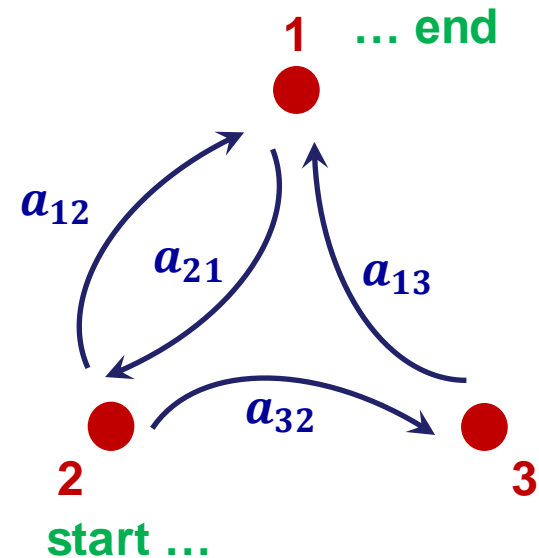
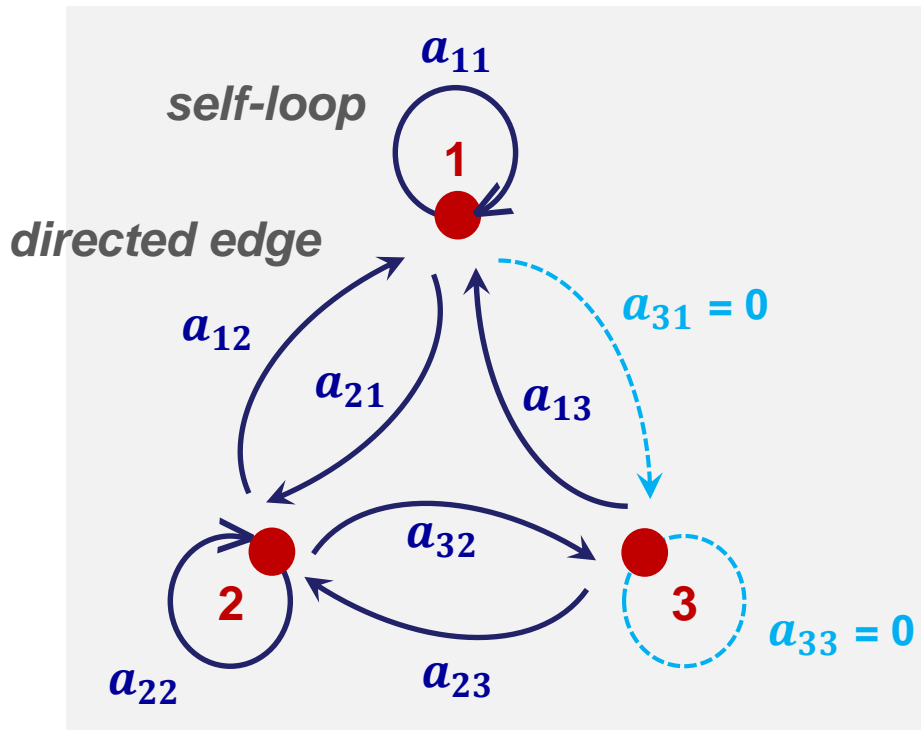
$$\mathcal{G} = (\mathcal{V} \text{ vertex set}, \mathcal{E} \text{ edge set})$$

Adjacency *finite* matrix $A_{\mathcal{G}}$

$$A_{\mathcal{G}} = \begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ 0 & a_{32} & 0 \end{pmatrix}$$



entry: *weight* on a *directed edge*



ex.: walk $\mathcal{W}_{1 \leftarrow 2}$ (from \mathcal{V}_2 to \mathcal{V}_1) of length 4

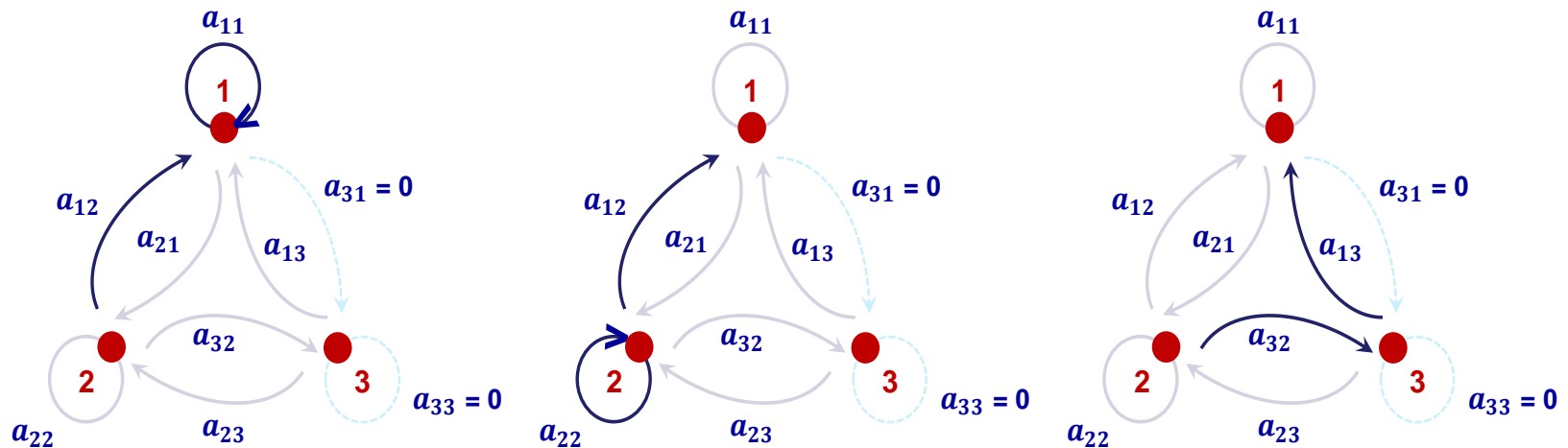
Basic results of algebraic graph theory

the **powers** of the **Adjacency matrix** A_G on a graph G generate
ALL weighted WALKS \mathcal{W} on G

$$A_G^2 = \begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ 0 & a_{32} & 0 \end{pmatrix}^2 = \begin{pmatrix} \blacksquare & \vdots & \vdots \\ \vdots & \ddots & \vdots \\ \vdots & \dots & \vdots \end{pmatrix}$$

\mathcal{W} of length 2 from v_2 to v_1 ($1 \leftarrow 2$)

$a_{11} \times a_{12} + a_{12} \times a_{22} + a_{13} \times a_{32}$



$$\Sigma = a_{11}a_{12} + a_{12}a_{22} + a_{13}a_{32}$$

Basic results of algebraic graph theory

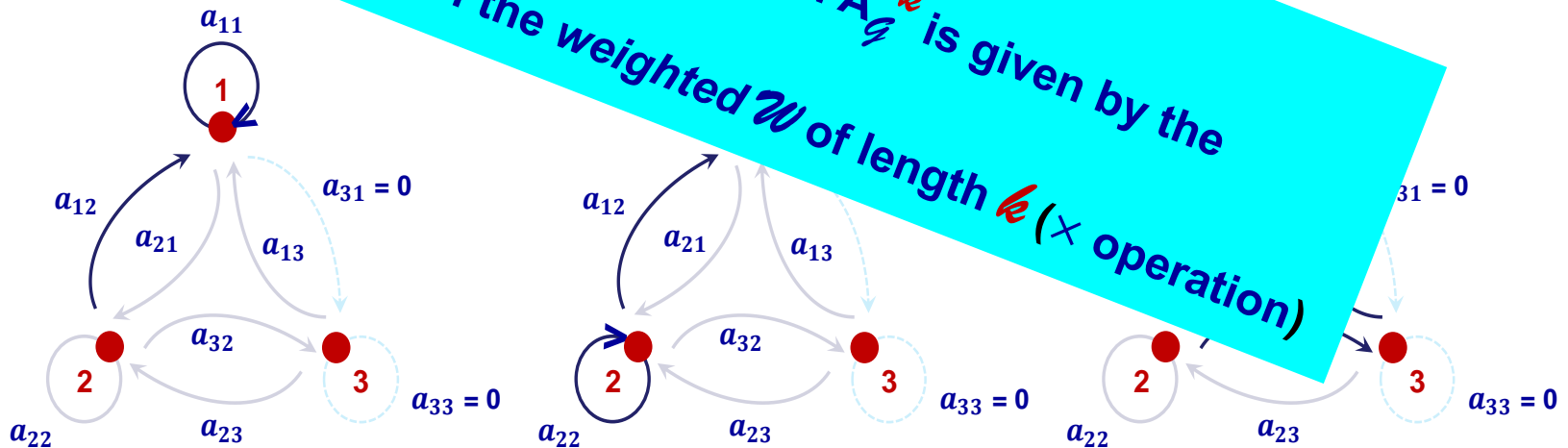
the **powers** of the **Adjacency matrix** A_G on a graph G generate
ALL weighted WALKS \mathcal{W} on G

$$A_G^2 = \begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \\ 0 & a_{31} \end{pmatrix}$$

$$a_{11} \times a_{12} + a_{12} \times a_{22} + a_{13} \times a_{32}$$

\mathcal{W} of length 2 from v_2 to v_1 ($1 \leftarrow 2$)

to keep in mind:
 each element of A_G^k is given by the
 sum of the weighted \mathcal{W} of length k (\times operation)

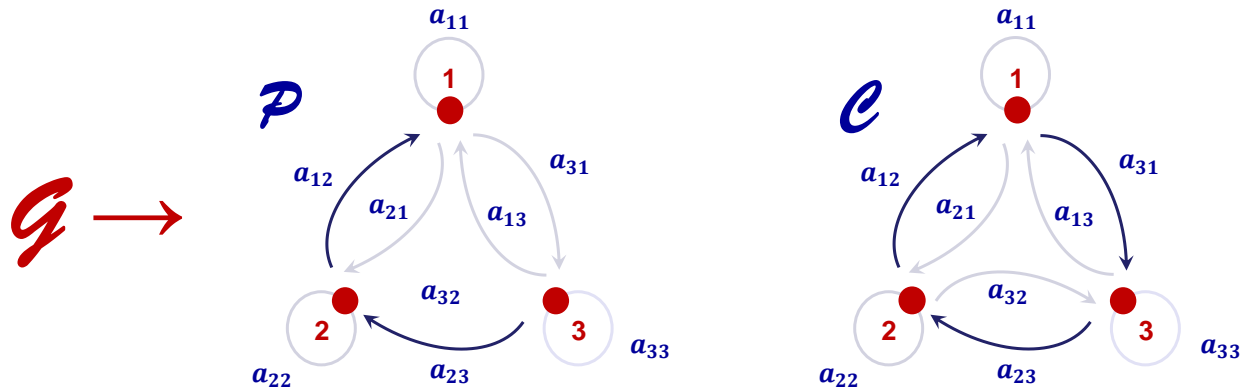


$$\Sigma = a_{11}a_{12} + a_{12}a_{22} + a_{13}a_{32}$$

Path-Sum

◇ *simple path* \mathcal{P} (self avoiding walk): \mathcal{W} whose \mathcal{V} are all **distinct**

◇ *simple cycle* \mathcal{C} (self avoiding polygon): \mathcal{W} whose **endpoints** are **identical** and **intermediate** \mathcal{V} are all **distinct** and different from the endpoints



« **Fundamental Theorem of Arithmetic** » on \mathcal{G} (P.-L. Giscard, 2012)

▶ \mathcal{W} factor **uniquely** into **prime** elements, i.e. **simple paths** and **simple cycles**

▶ if \mathcal{G} is **finite** the number of primes is **finite**

▶ resummation of all \mathcal{W} involves a **finite** number of operations: **sum on simple paths** and **continuous fraction of simple cycles** with vertex removal

Power series of A_q

$$\text{ex.: } \exp[A_q] = \sum_{k=0}^{\infty} \frac{1}{k!} A_q^k$$

$$(A_q)^k = \begin{pmatrix} \dots & & \\ \vdots & (A_q)^k_{\omega\alpha} & \vdots \\ \dots & & \end{pmatrix}$$

to keep in mind:
each element of A_q^k is given by the
sum of the weighted \mathcal{W} of length k (standard \times operation)

Power series of A_G

ex.: $\exp[A_G] = \sum_{k=0}^{\infty} \frac{1}{k!} A_G^k$

$$(A_G)^k = \begin{pmatrix} \dots & & \dots \\ \vdots & (A_G)^k_{\omega\alpha} & \vdots \\ \dots & & \dots \end{pmatrix}$$

$$F(A_G)_{\omega\alpha} = \sum_{k=0}^{\infty} c_k \sum_{\mathcal{W}_{G, \alpha\omega; k}} a_{\omega h_k} \cdots \times a_{h_3 h_2} \times a_{h_2 \alpha}$$

power series of A_G

all weighted walks \mathcal{W} from v_α to v_ω of length k

Power series of $A_{\mathcal{G}}$

ex.: $\exp[A_{\mathcal{G}}] = \sum_{k=0}^{\infty} \frac{1}{k!} A_{\mathcal{G}}^k$

$$(A_{\mathcal{G}})^k = \begin{pmatrix} \dots & & \dots \\ \vdots & (A_{\mathcal{G}})^k_{\omega\alpha} & \vdots \\ \dots & & \dots \end{pmatrix}$$

$$F(A_{\mathcal{G}})_{\omega\alpha} = \sum_{k=0}^{\infty} c_k \sum_{\mathcal{W}_{\mathcal{G}, \alpha\omega; k}} a_{\omega h_k} \cdots \times a_{h_3 h_2} \times a_{h_2 \alpha}$$

power series of $A_{\mathcal{G}}$

all weighted walks \mathcal{W} from v_{α} to v_{ω} of length k

Path-Sum

« Fundamental Theorem of Arithmetic » on \mathcal{G} (P.-L. Giscard, 2012)

- ▶ \mathcal{W} factor *uniquely* into *prime* elements, i.e. *simple paths* and *simple cycles*
- ▶ if \mathcal{G} is *finite* the number of primes is *finite*
- ▶ resummation of all \mathcal{W} involves a *finite* number of operations: *sum on simple paths* and *continuous fraction of simple cycles* with vertex removal

Power series of A_G

ex.: $\exp[A_G] = \sum_{k=0}^{\infty} \frac{1}{k!} A_G^k$

$$(A_G)^k = \begin{pmatrix} & & \dots & \\ & & (A_G)^k_{\omega\alpha} & \\ & & \dots & \\ & & & \dots \end{pmatrix}$$

$$F(A_G)_{\omega\alpha} = \sum_{k=0}^{\infty} c_k \sum \mathcal{W}_{G, \alpha\omega; k} a_{\omega h_k} \cdots \times a_{h_3 h_2} \times a_{h_2 \alpha}$$

power series of A_G

all weighted walks \mathcal{W} from v_α to v_ω of length k

Path-Sum

$$F(A_G)_{\omega\alpha} = \sum_{\mathcal{P}_{G, \alpha\omega; \ell}} f(a_{\omega\omega}) \times a_{\omega\mu_\ell} \cdots f(a_{\mu_2\mu_2}) a_{\mu_2\alpha} \times f(a_{\alpha\alpha})$$

sum on the finite set of *simple paths* \mathcal{P} of length ℓ

edge weight

effective v weight

sum over the finite set of *simple cycles* \mathcal{C}
(continued fraction of finite breadth)

$$\mathbf{A}_g(t) = \begin{pmatrix} \dots \\ \langle s_\omega | \mathbf{A}(t) | s_\alpha \rangle \\ \dots \end{pmatrix}$$

$$\text{OE}[\mathbf{A}_g](t', t) = \begin{pmatrix} \dots \\ \langle s_\omega | \text{OE}[\mathbf{A}_g](t', t) | s_\alpha \rangle \\ \dots \end{pmatrix}$$

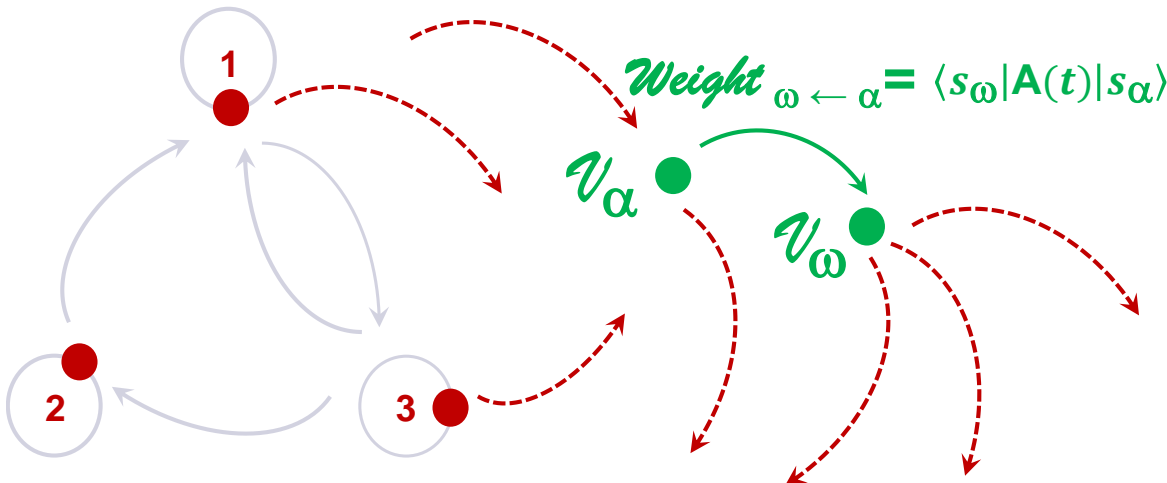
Σ ALL weighted walks $\omega \leftarrow \alpha$ on \mathbf{A}_g

but using \star -product

$$(f \star g) = \int_t^{t'} f(t', \tau) g(\tau, t) d\tau$$

instead of \times

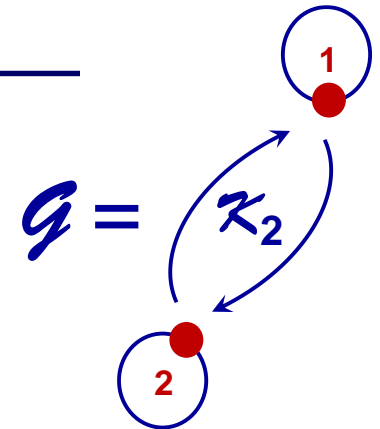
\mathbf{A}_g



Path-Sum

An example: 2×2 matrix

$$\mathbf{A}(t) = \begin{pmatrix} a_{11}(t) & a_{12}(t) \\ a_{21}(t) & a_{22}(t) \end{pmatrix}$$



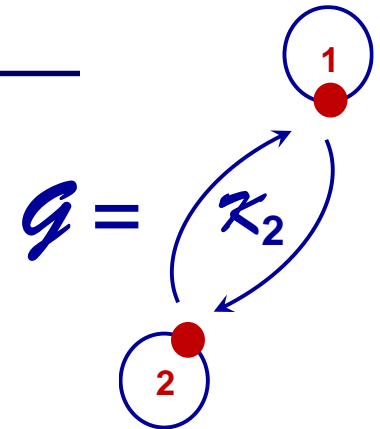
Path-Sum

$$\text{OE}[\mathbf{A}](t', t) = \begin{pmatrix} \int_t^{t'} G_{K_2, 11}(t', \tau) d\tau & OE_{12}(t', t) \\ OE_{21}(t', t) & \int_t^{t'} G_{K_2, 22}(t', \tau) d\tau \end{pmatrix}$$

- ▶ **entry** → solving an equation with *analytical tools*
- ▶ **finite** number of operations → *unconditional convergence*
- ▶ **non perturbative** formulation of OE
- ▶ **scalability**

An example: 2×2 matrix

$$\mathbf{A}(t) = \begin{pmatrix} a_{11}(t) & a_{12}(t) \\ a_{21}(t) & a_{22}(t) \end{pmatrix}$$



Path-Sum

$$\mathbf{OE}[\mathbf{A}](t', t) = \begin{pmatrix} \int_t^{t'} G_{K_2, 11}(t', \tau) d\tau & OE_{12}(t', t) \\ OE_{21}(t', t) & \int_t^{t'} G_{K_2, 22}(t', \tau) d\tau \end{pmatrix}$$

« **Fundamental Theorem of Arithmetic** » on \mathcal{G} (P.-L. Giscard, 2012)

► \mathcal{W} factor **uniquely** into **prime** elements, i.e. **simple paths** and **simple cycles**

► if \mathcal{G} is **finite** the number of primes is **finite**

► resummation of all \mathcal{W} involves a **finite** number of operations: **sum on simple paths** and **continuous fraction of simple cycles** with vertex removal



An example: 2×2 matrix

$$(f * g) = \int_t^{t'} f(t', \tau) g(\tau, t) d\tau$$

$$\text{OE}[A](t', t) = \begin{pmatrix} \int_t^{t'} G_{K_2,11}(t', \tau) d\tau & OE_{12}(t', t) \\ OE_{21}(t', t) & \int_t^{t'} G_{K_2,22}(t', \tau) d\tau \end{pmatrix}$$

$a_{ij}(t)$

$$[\mathbf{1}_* - (* * * \dots)]^{*-1} = \sum_{n \geq 0} (* * * \dots)^{*n}$$

Neumann series (analytical)
linear Volterra (2nd kind) (numerical)

An example: 2×2 matrix

$$(f * g) = \int_t^{t'} f(t', \tau) g(\tau, t) d\tau$$

$$OE[A](t', t) = \begin{pmatrix} \int_t^{t'} G_{K_2,11}(t', \tau) d\tau & OE_{12}(t', t) \\ OE_{21}(t', t) & \int_t^{t'} G_{K_2,22}(t', \tau) d\tau \end{pmatrix}$$

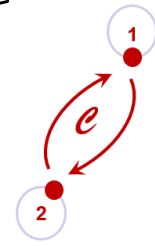
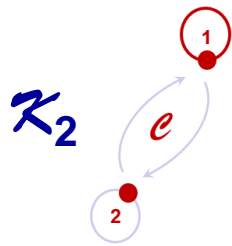
$a_{ij}(t)$

$$[1_* - (* * * \dots)]^{*-1} = \sum_{n \geq 0} (* * * \dots)^{*n}$$

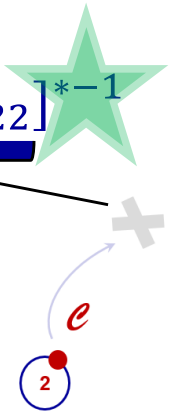
Neumann series (analytical)
linear Volterra (2nd kind) (numerical)

sum on simple cycles

$$G_{K_2,11} = [1_* - a_{11} - a_{12} * G_{K_2 \setminus \{1\},22} * a_{21}]^{*-1}$$

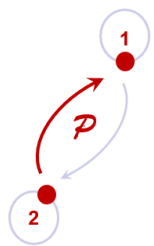


$$G_{K_2 \setminus \{1\},22} = [1_* - a_{22}]^{*-1}$$



$$OE_{12}(t', t) \equiv \int_t^{t'} G_{K_2 \setminus \{2\},11} * a_{12} * G_{K_2,22}(t', \tau) d\tau$$

$$G_{K_2 \setminus \{2\},11} * a_{12} * G_{K_2,22}(t', \tau)$$



- ▶ END !
- ▶ finite sum on simple \mathcal{P}

- ▶ END of the *continued fraction* !
- ▶ finite sum on e

sum on simple paths

Summary (partial)

- ▶ ... take a **finite** matrix $\mathbf{A}_g(\mathbf{t})$ associated to g (Hermitian or not, periodic or not...)
- ▶ each entry of \mathbf{A}_g^k is given is given by a **finite** number of operations by using Path-Sum (with \times product)
- ▶ each entry of $\mathbf{OE}[\mathbf{A}_g](t', t)$ is given is given by a **finite** number of operations by using Path-Sum (with $*$ -product and $[\mathbf{1}_* - (* * * \dots)]^{*-1}$)

Summary (partial)

▶ ... take a **finite** matrix $\mathbf{A}_g(\mathbf{t})$ associated to g (Hermitian or not, periodic or not...)

▶ each entry of \mathbf{A}_g^k is given by a **finite** number of operations by using Path-Sum (with \times product)

▶ each entry of $\mathbf{OE}[\mathbf{A}_g](t', t)$ is given by a **finite** number of operations by using Path-Sum (with $*$ -product and $[\mathbf{1}_* - (* * * \dots)]_*^{-1}$)

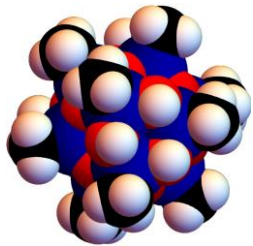
■ the **matrix** nature of the problem is **fully replaced** when working on **entries**

■ or, one can keep it partially... → **PARTITIONS** (**scalability**)

■ the **convergence** of the **Neumann** series (**analytical**) is **superexponential**

■ a convenient (**numerical**) approach: linear **Volterra** equations (**2nd kind**)

- Basic results of algebraic graph theory
- Path-Sum applied to the ordered exponential (OE)
- Applications:
 - ▶ Circularly polarized excitation
 - ▶ Linearly polarized excitation, Bloch-Siegert (BS) effect
 - ▶ N spins homonuclear dipolar Hamiltonian, H_D



Applications – Circularly polarized excitation (test model)

$$\mathbf{H}(t) = \begin{pmatrix} \frac{\omega_0}{2} & \beta e^{-i\omega t} \\ \beta e^{i\omega t} & -\frac{\omega_0}{2} \end{pmatrix}, [\mathbf{H}(t'), \mathbf{H}(t)] \neq 0$$

$$\mathbf{H}(t) = \frac{1}{2} \omega_0 \boldsymbol{\sigma}_z + \beta [\boldsymbol{\sigma}_x \cos(\omega t) + \boldsymbol{\sigma}_y \sin(\omega t)]$$

$$[1_* - (* * * \dots)]^{*-1}$$

Path-Sum

$$G_{K_2,11}(t) = \left(1_* - \frac{\omega_0}{2i} + \frac{i\beta^2}{\Delta} (e^{-i\Delta(t'-t)} - 1) \right)^{*-1}$$

OE entry

Neumann series

$$OE[-i\mathbf{H}](t)_{11} = 1 + \sum_{n=0}^{\infty} \frac{(-it\beta^2/\Delta)^{n+1}}{(n+1)!} \sum_{k=0}^{n+1} \binom{n+1}{k} \left(\frac{\Delta\omega_0}{2\beta^2} - 1 \right)^k {}_2F_1 \left(-k, -k+n+1; -n-1; \frac{\Delta^2}{\frac{\Delta\omega_0}{2} - \beta^2} \right)$$

Gauss hypergeometric



OE[-iH](t)

$$\begin{pmatrix} e^{-\frac{1}{2}it(\Delta + \frac{\omega_0}{2})} \left(\cos(\alpha t/2) + \frac{i}{\alpha} \left(\Delta - \frac{\omega_0}{2} \right) \sin(\alpha t/2) \right) & -\frac{2i\beta}{\alpha} e^{-\frac{1}{2}it(\Delta + \frac{\omega_0}{2})} \sin(\alpha t/2) \\ -\frac{2i\beta}{\alpha} e^{\frac{1}{2}it(\Delta + \frac{\omega_0}{2})} \sin(\alpha t/2) & e^{\frac{1}{2}it(\Delta + \frac{\omega_0}{2})} \left(\cos(\alpha t/2) - \frac{i}{\alpha} \left(\Delta - \frac{\omega_0}{2} \right) \sin(\alpha t/2) \right) \end{pmatrix}$$

$$\mathbf{U}(t) = \exp\left(-\frac{1}{2}i\omega t \boldsymbol{\sigma}_z\right) \exp\left(-it \left(\frac{1}{2}(\omega_0 - \omega) \boldsymbol{\sigma}_z + \beta \boldsymbol{\sigma}_x\right)\right)$$

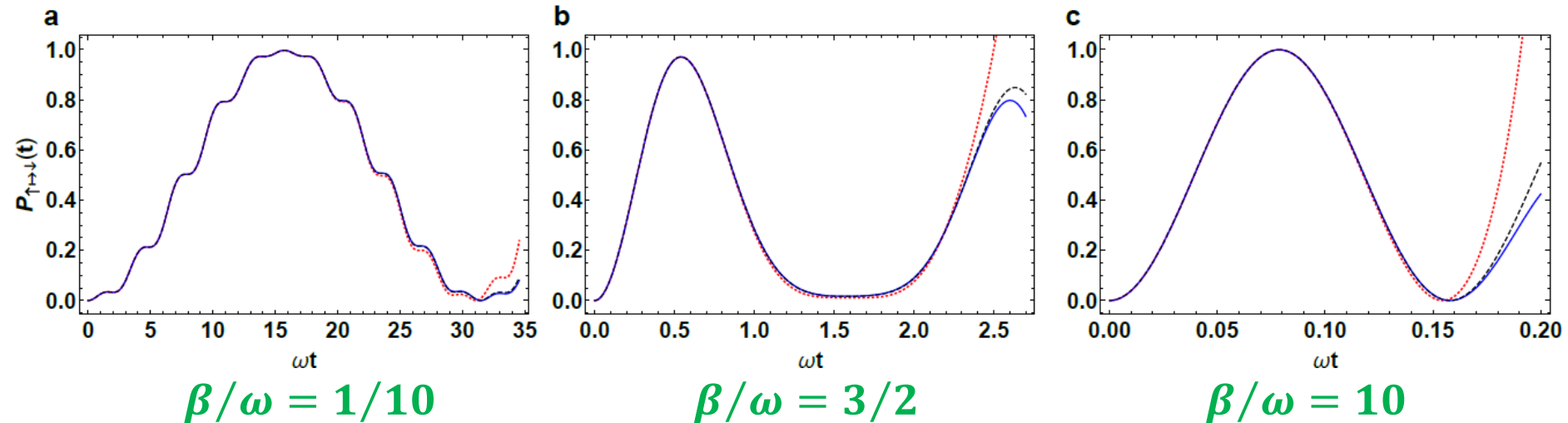
Applications – Linearly polarized excitation, Bloch-Siegert (BS) effect

$$\mathbf{H}(t) = \frac{1}{2}\omega_0\boldsymbol{\sigma}_z + 2\beta\boldsymbol{\sigma}_x\cos(\omega t)$$

$$\mathbf{H}(t) = \begin{pmatrix} \frac{\omega_0}{2} & 2\beta\cos(\omega t) \\ 2\beta\cos(\omega t) & -\frac{\omega_0}{2} \end{pmatrix}$$

P(t) transition probability

$\omega = \omega_0$ or $\omega \neq \omega_0$



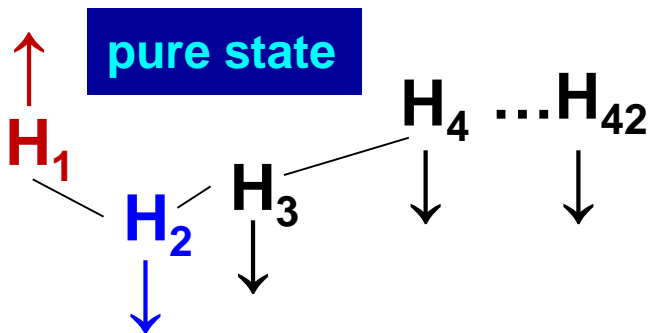
► analytical expression with few orders of the Neumann series

Applications – N spin systems, homonuclear dipolar Hamiltonian, H_D

$t = 0$

Coll.: F. Ribot, France

$(\text{CH}_3)_{12}(\text{OH})_6\text{Sn}_{12}$

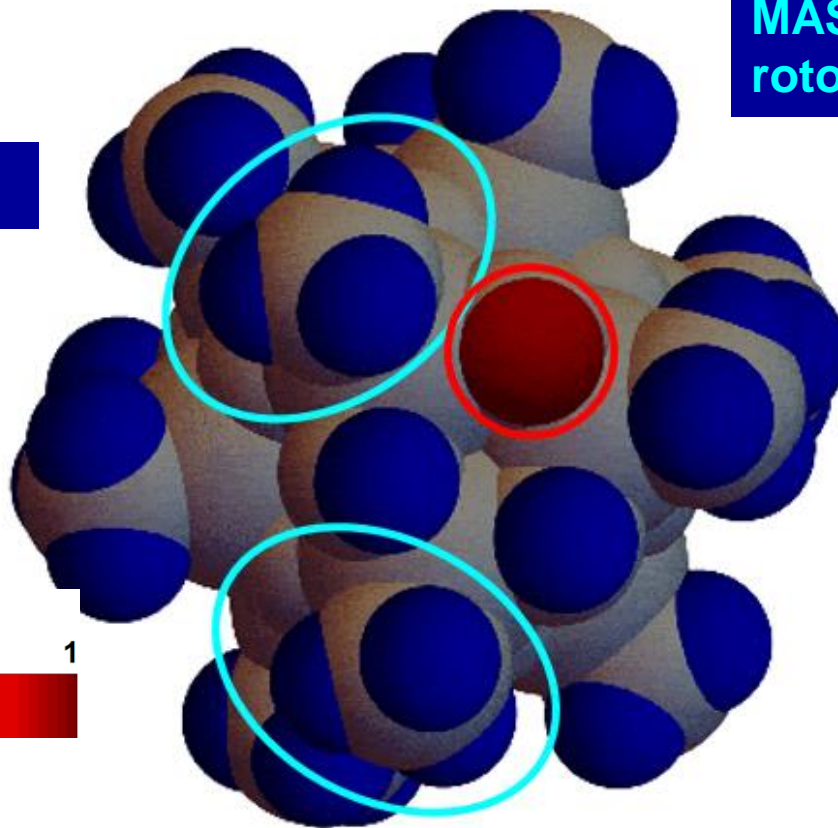
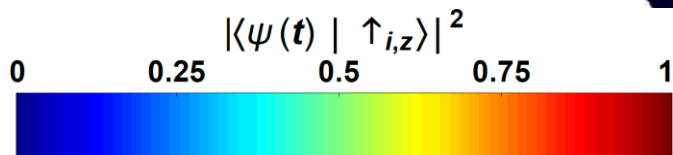


$t \text{ (ms)} = 0.$

42 protons
« rigid » CH_3

MAS 10 kHz
rotor period 0.1 ms

analytical expression

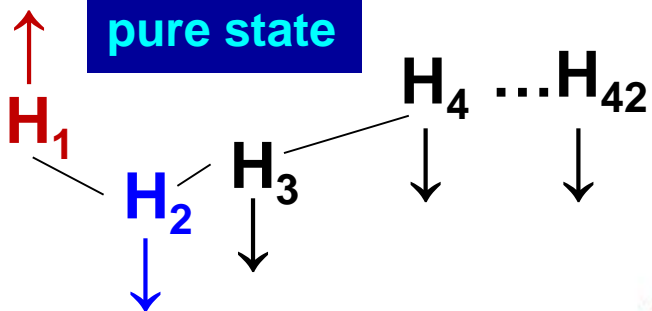


Applications – N spin systems, homonuclear dipolar Hamiltonian, H_D

$t = 0$

Coll.: F. Ribot, France

$(\text{CH}_3)_{12}(\text{OH})_6\text{Sn}_{12}$

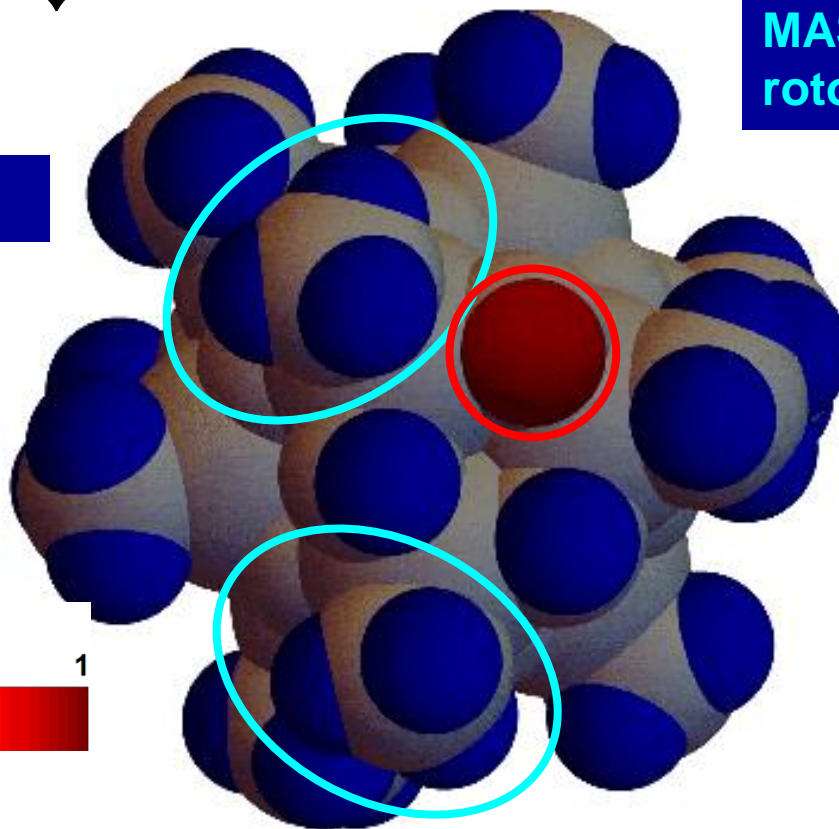
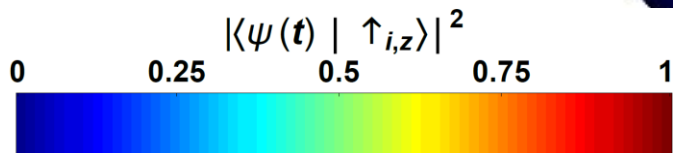


$t \text{ (ms)} = 0.$

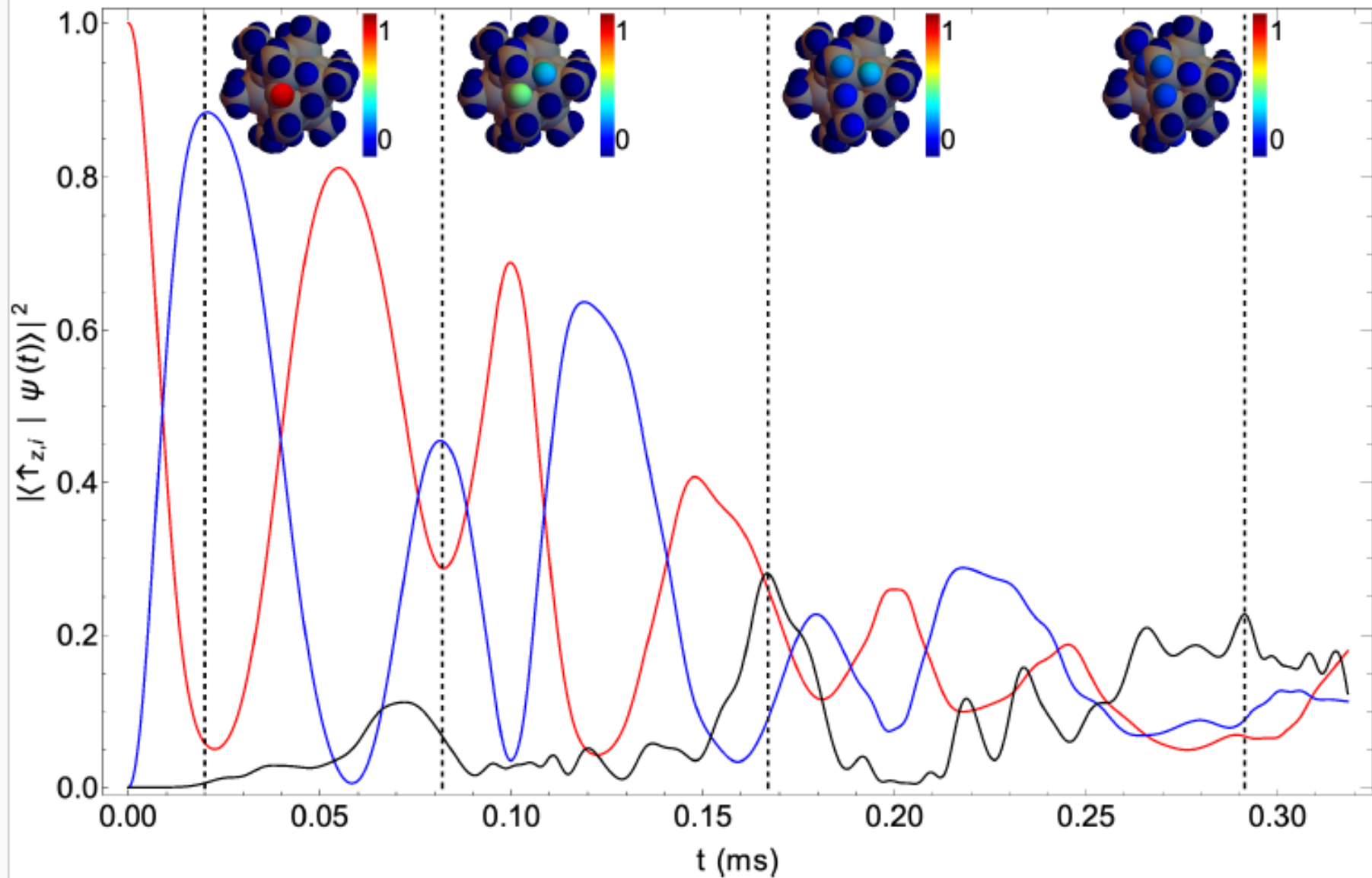
42 protons
« rigid » CH_3

MAS 10 kHz
rotor period 0.1 ms

analytical expression



Applications – N spin systems, homonuclear dipolar Hamiltonian, H_D



Path-Sum



- ▶ a new approach
- ▶ analytical expression for $U(t)$
- ▶ unconditional convergence
- ▶ non perturbative formulation
- ▶ scalable to large spin systems
- ▶ other theory/applications to come...

(very) warm thanks to P.-L. Giscard

Ass. Pr. in Calais, France

Liouville laboratory

Algebraic Combinatorials

giscard@univ-littoral.fr



Post doctoral position available in Paris: on NMR instrumentation & DNP

To go further – Path-Sum vs other methods

- ▶ main goal → get an **exact** form for $U(t)$



- ▶ FLOQUET

- ▶ ZASSENHAUS

- ▶ FER/TROTTER-SUZUKI

- ▶ MAGNUS

- ▶ **PATH-SUM**

- ▶ usually:  on $H(t)$ → choice in

- ▶ FLOQUET

- ▶ ZASSENHAUS

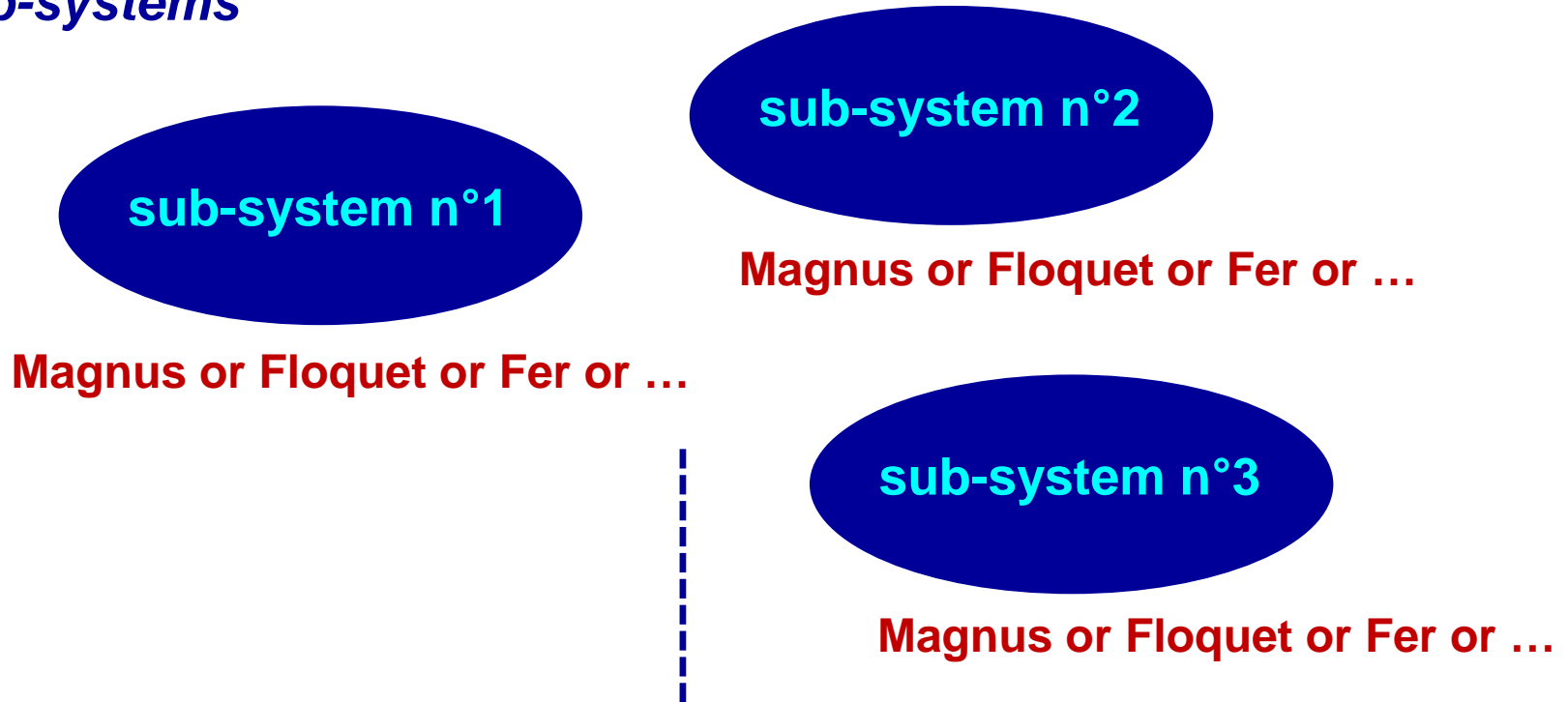
- ▶ FER/TROTTER-SUZUKI

- ▶ MAGNUS

- ▶ **PATH-SUM** is **exact** and **PARTITIONS** allow to **choose the dimension** of the of the working space from $H(t)$ to $U(t)$

To go further – Scale invariance

Take a **partition** of a spin system in a set of (*smaller, independent*) *sub-systems*



the **exact** evolution of the **entire** spin system as functions of the evolutions of the **isolated sub-systems** is given by **Path-Sum**

(though **non contiguous blocks** in $H(t)$ matrix!)

To go further – WHY does Path-Sum work?

- ▶ the **EXACT** result is given by a **FINITE** number of terms
- ▶ the **matrix** nature of the problem is **fully replaced** when working on **entries**
- ▶ or, one can keep it partially... → **PARTITIONS**

- ▶ **hard** work → $[\mathbf{1}_* - (* * * \dots)]^{*-1}$



- ▶ hopefully: the **Neumann series** give the analytical solution at any order with unconditional convergence (not to be “found” ... just apply a “recipe”)
- ▶ the **convergence** of the Neumann series is **superexponential**
- ▶ a **convenient** numerical approach: linear **Volterra** equations (**2nd kind**)

$$D_x^2 u + \left[\frac{\gamma}{x} + \frac{\delta}{x-1} + \frac{\epsilon}{x-a} \right] D_x u + \left[\frac{\alpha\beta x - q}{x(x-1)(x-a)} \right] u = 0$$

ex.: the best obtainable solution for the **general 2×2 matrix** (closed form for the **confluent Heun's special functions**) (see Q. Xie, 2018)

To go further – Exponential explosions

- ▶ 1st explosion: related to the **size of $H(t)$** with **many-body** systems (Q nature)
- ▶ 2nd explosion: related to the **time** needed to isolate the **primes** (\mathcal{G} nature)

Lanczos–Path-Sum (numerical) fixes the 2nd explosion:

Idea behind: initial $H(t)$ → time dependent **tridiagonal matrix**

expectations: to reach **excellent convergence** with the breadth of the continued fraction and why not ?... "Circumvent" the 1st explosion

P.-L. Giscard et al., 2019, in preparation

To go further – Complexity theory

▶ for *finite* \mathcal{G} : the decomposition of \mathcal{W} in *primes* (e.g. *simple paths* & *cycles*) for the \blacksquare (*nested*) operation *exists and is unique*



▶ to determine the existence of a prime of *length* L is *NP-complete* (no(?) algorithm with polynomial complexity)

▶ to *count* them is *#P-complete* (the same but for counting problems)

▶ to count them for a fixed *length* L is *#W[1]-complete* (same as *#P-complete* but with parameters, such as L , taken into account)

▶ **BUT:** for *sparse* \mathcal{G} : counting becomes *polynomial* in the max degree of \mathcal{G} !

see: P. L. Giscard et al., *Algorithmica*, 2019

► fundamentally: $\mathcal{R}_{\text{resolvent}}[A(t)]_{*} \text{ product} = \frac{d}{dt} \text{OE}[A(t)] \rightarrow \text{Path-Sum}$

- each **entry** of $A(t)$ must be **bounded** on $[0,t]$, a **bounded** interval of time
- if the entries are **not bounded**, Path-Sum still work ... but perhaps the Neumann series will **not converge**
- **continuity** is **not** necessary
- **if continuity**: Volterra equations are much **easier** to handle
- $A(t)$ can be Hermitian **or not**, periodic **or not** ... and entries can be: matrices, quaternions, octonions, division rings...

- ▶ **finite** $A(t)$: **sufficient** condition for **finite breadth** of the continued fraction
- ▶ **NOT** a **necessary** condition: ex. a **finite** number of **simple cycles** in an **infinite** matrix
- ▶ in some cases, Path-Sum can still be applied on **infinite matrices**: **strong symmetry**, e.g. invariance by translation (soluble **non-linear** Volterra equations)

In other words:

- **infinity** of cycles ... but **self-similar** like in a **fractal**
- the corresponding continued fraction is of **finite breadth**

To go further – Taylor... or Neumann series?

▶ take one entry: $f(t) = \text{OE}[A(t)]_{ij}$

▶ **Taylor** series: expansion in t^n i.e. $f(t) = \sum_{n=0}^{\infty} \frac{f^{(n)}(0)}{n!} t^n$

ex.: $\frac{1}{1-t} = \sum_{n=0}^{\infty} t^n = 1 + t + t^2 + \dots + t^n + \dots$ with **$r = 1$** (*radius of CV*)

▶ **Neumann** series: uses the -product, i.e. $f(t) = \sum_{n=0}^{\infty} f^{*n}$

each order contains functions represented by infinite Taylor series

$r = \infty$ (!) with *uniform & superexponential CV*

To go further – N spins starting with a pure state

- ▶ starting with a **pure state** with **1** up-spin (total: **N**, **any geometry**)

Path-sum contains all ***N-order correlations***

→ **if $\omega_{rot} = 0$**

all terms of the Neumann series are ***explicitly*** known

→ **if $\omega_{rot} \neq 0$**

still ***analytical*** up to the CV of the series to the solution

- ▶ starting with a **pure state** with **4 or 5** up-spin is still tractable
(*i.e.* no exponential explosion)

To go further – Pure state vs partial polarization

▶ **Pure state:** if k up-spins over N and $k \ll N \rightarrow$ space of states dim. $\approx N^k$
(suppression of the exponential explosion)

▶ **Partial polarization:** a **cut-off** is needed \rightarrow if $\left| \frac{int_{i,j}}{intV} \leq \frac{1}{\text{cut-off}} \right|$ then
 $int_{i,j} = 0$

cut-off: « high » for **chains** but decreases for more « **dense** » spin systems

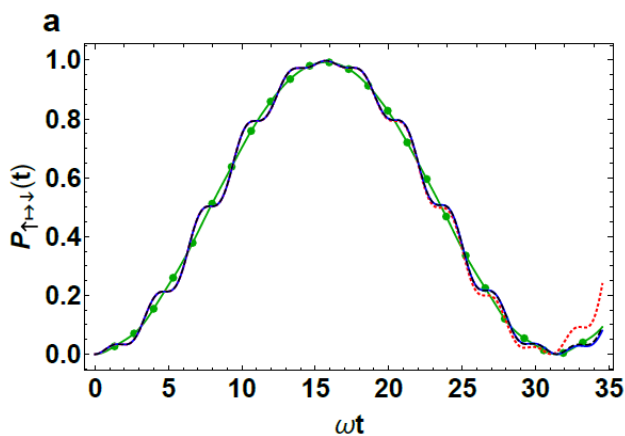


next target: to extend **Path-Sum** to **mixed states** *via* a **decomposition on pure states**

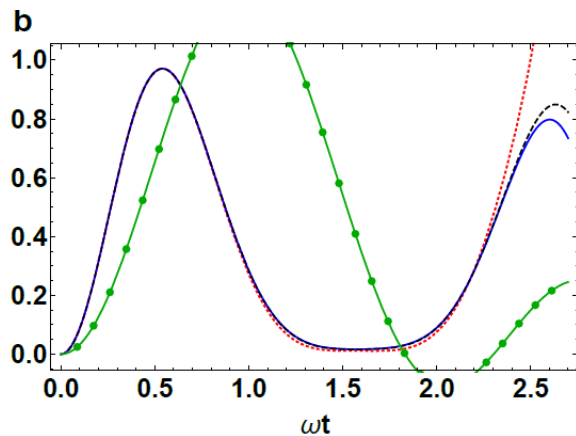
To go further – Path-Sum vs Floquet theory for Bloch-Siegert effect

P(t) transition probability

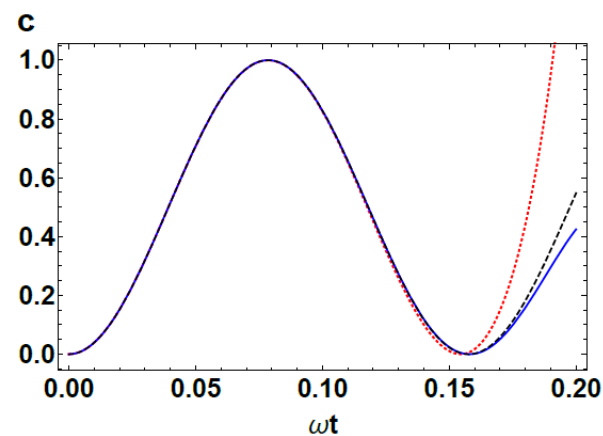
$\omega = \omega_0$ OR $\omega \neq \omega_0$



$\beta/\omega = 1/10$

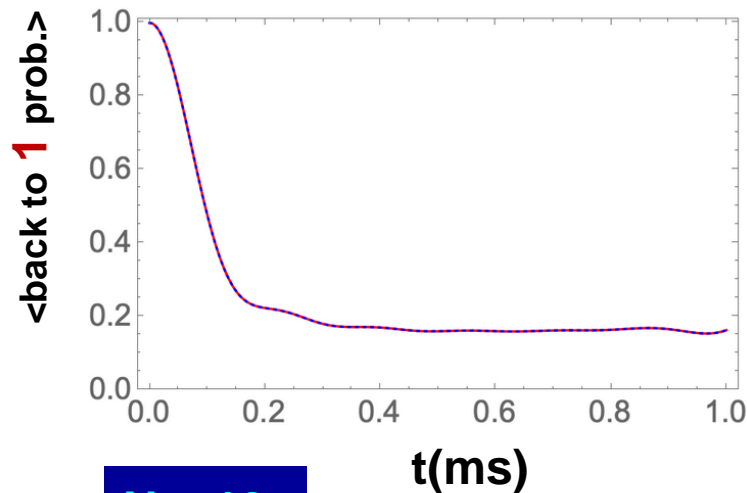
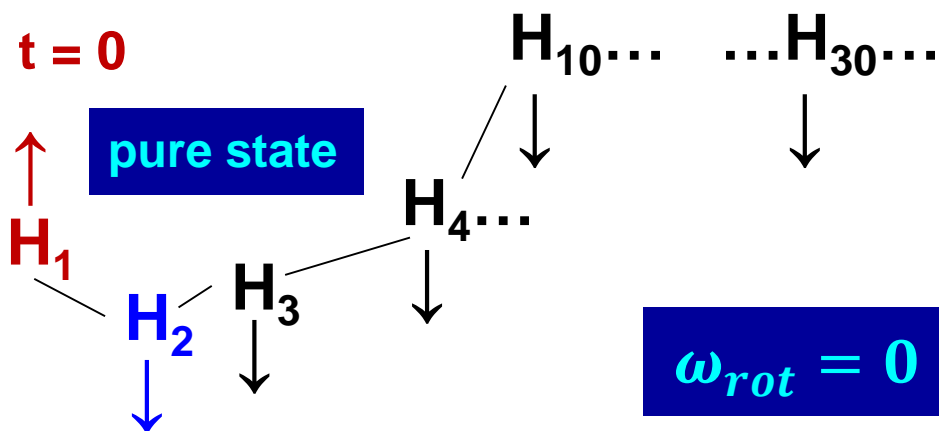


$\beta/\omega = 3/2$

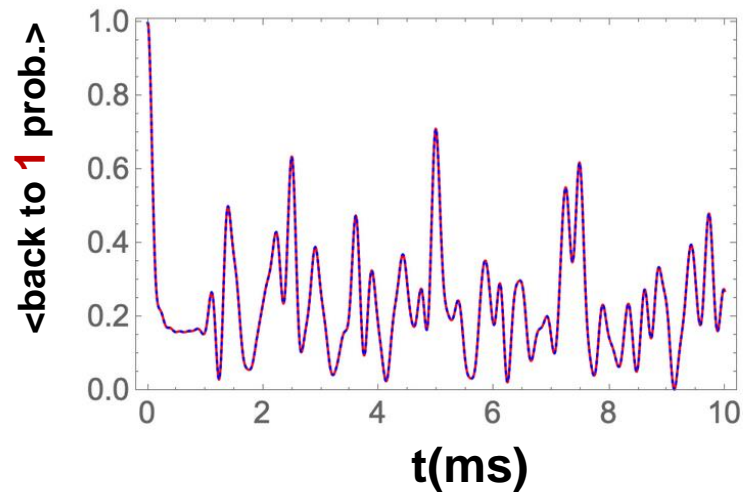
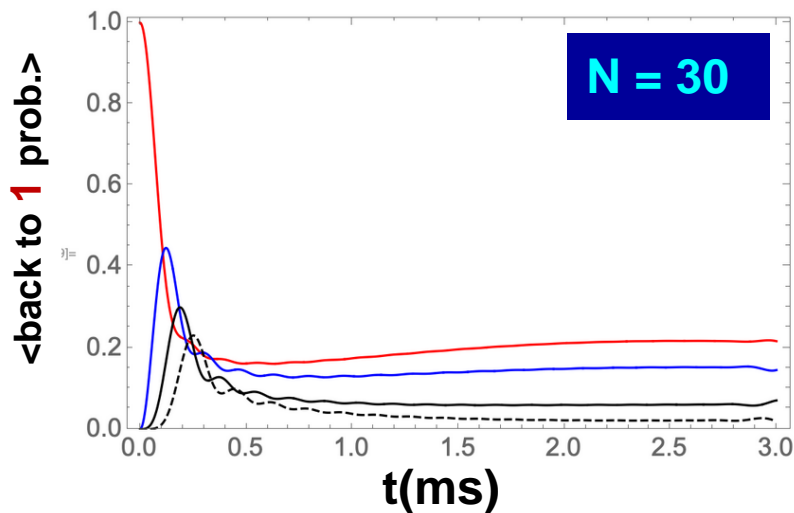


$\beta/\omega = 10$

To go further – N spin chains and H_D

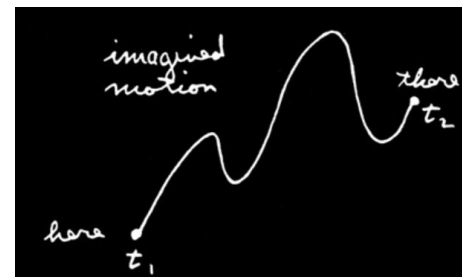


$N = 10$



To go further – Liouvillian space, Feynman paths and diagrams

- ▶ extension of Path-Sum in the Liouvillian space is possible using the **adjoint operator** of $H(t)$



« With application to quantum mechanics, path integrals suffer most grievously from a serious defect. They do not permit a discussion of spin operators or other such operators in a simple and lucid way » (R.P. Feynman)

- ▶ Path-sum can be used starting from the **Lagrangian** with **action** as **weight** on a given \mathcal{W}
- ▶ Path-sum can be used starting from the **Hamiltonian** with **energy** as **weight** on a given \mathcal{W}
- ▶ **Feynman diagrams**: \mathcal{W} of \mathcal{G} in the state space (but **continuous**)
- ▶ Path-sum performs a formal **re-summation** of an infinite number of \mathcal{W} , *i.e.* Feynman diagrams !