

## Quantum jumps and rate operators in open quantum system dynamics





### **Turku Centre for Quantum Physics** Non-Markovian Processes and Complex Systems Group



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Dalibard, Castin, Molmer PRL 1992

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Piilo, Maniscalco, Härkönen, Suominen: PRL 2008

## 3. Unified framework: ROQJ - Rate operator quantum jumps

Smirne, Caiaffa, Piilo PRL 2020 See also Chruscinski, Luoma, Piilo, Smirne arXiv:2009.11312



## Preliminaries: problem setting



 Any realistic quantum system S is coupled to its environment E

Master equation description:

$$\frac{d\rho(t)}{dt} = -i[H,\rho_S] + \sum_k \gamma_k(t) \left( A_k \rho_S(t) A_k^{\dagger} - \frac{1}{2} A_k^{\dagger} A_k \rho_S(t) - \frac{1}{2} \rho_S A_k^{\dagger} A_k \right)$$

• Decomposition of the density matrix  $\rho(t) = \sum_{i} P_i(t) |\psi_i(t)\rangle \langle \psi_i(t)| \longrightarrow \begin{array}{c} \text{Stochastic} \\ \text{descriptions} \end{array}$ 

## Simple classification of Monte Carlo/stochastic methods

	Markovian	non-Markovian
Jump methods:	MCWF (Dalibard, Castin, Molmer) Quantum Trajectories (Zoller, Carmichael)	Fictitious modes (Imamoglu) Pseudo modes (Garraway) Doubled H-space (Breuer, Petruccione) Triple H-space (Breuer) Non-Markovian Quantum Jump (Piilo et al)
Diffusion methods:	QSD (Diosi, Gisin, Percival)	Non-Markovian QSD (Strunz, Diosi, Gisin,Yu) Stochastic Schrödinger equations (Barchielli)
Jump	Diffusion	•
GUILLE GUILL		Plus:Wiseman, Gambetta, Budini, Gaspard, Lacroix, Donvil and Muratore-Ginanneschi (not comprehensive list, apologies for any omissions)

## Simple classification of Monte Carlo/stochastic methods



## Simple classification of Monte Carlo/stochastic methods





## Markovian Monte Carlo Wave Function method

### Density matrix and state vector ensemble

Suppose now we want to solve the semigroup, Markovian GKSL equation

$$\frac{d\rho(t)}{dt} = -i[H,\rho_S] + \sum_k \gamma_k \left( A_k \rho_S A_k^{\dagger} - \frac{1}{2} A_k^{\dagger} A_k \rho_S - \frac{1}{2} \rho_S A_k^{\dagger} A_k \right)$$

Q: How to solve the master equation?

Few exact models and analytical solutions
 Can we find the solution by evolving an ensemble of state vectors instead of directly solving the density matrix?

Generally, we can decompose the density matrix as

$$\rho(t) = \sum_{i} P_i(t) |\psi_i(t)\rangle \langle \psi_i(t)|$$

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## Basics of stochastic state vector evolution

Monte Carlo wave function method (Markovian) (Dalibard, Castin, Molmer, PRL 1992) Ensemble of N state vectors  $\begin{bmatrix}
\psi_1(t_0) \longrightarrow \psi_1(t_1) & \dots & \psi_1(t_n) \\
\psi_2(t_0) \longrightarrow \psi_2(t_1) & \dots & \psi_2(t_n) \\
\vdots \\
\psi_N(t_0) \longrightarrow \psi_N(t_1) & \dots & \psi_N(t_n)
\end{bmatrix}$ Time

At each point of time, density matrix  $\rho$  as average of state vectors  $\Psi_i$ :

$$\rho(t) = \frac{1}{N} \sum_{i=1}^{N} |\psi_i(t)\rangle \langle \psi_i(t)|$$

The time-evolution of each  $\Psi_i$  contains stochastic element due to random quantum jumps.



Time-evolution of state vector  $\Psi_i$ :

At each point of time: decide if quantum jump happened.

 $P_j$ : probability that a quantum jump occurs in a given time interval  $\delta t$ :



E

For example: 2-level atom Probability for atom being transferred from the excited to the ground state and photon emitted.



## Example: driven 2-state system, Markovian

Quantum jump: Discontinuous stochastic change of the state vector. Excited state probability P for a driven 2-level atom

**Excited** state



Ground state

$$\frac{d\varrho}{dt} = -i[H,\varrho] + \Gamma \left[ \sigma_{-}\varrho\sigma_{+} - \frac{1}{2} \{\sigma_{+}\sigma_{-},\varrho\} \right]$$
$$\rho(t) = \frac{1}{N} \sum_{i=1}^{N} |\psi_{i}(t)\rangle \langle \psi_{i}(t)|$$



## Example: driven 2-state system, Markovian





## Markovian Monte Carlo wave function method

Master equation to be solved:

$$\frac{d\rho(t)}{dt} = \frac{1}{i\hbar} \left[ H_S, \rho \right] + \sum_m \Gamma_m C_m \rho C_m^{\dagger} - \frac{1}{2} \sum_m \Gamma_m \left( C_m^{\dagger} C_m \rho + \rho C_m^{\dagger} C_m \right)$$

#### For each ensemble member $\psi$ :

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$$i\hbar \frac{d}{dt} |\Psi(t)\rangle = H |\Psi(t)\rangle$$

$$H = H_s + H_{dec}$$

$$H_{dec} = -\frac{i\hbar}{2} \sum_{m} \Gamma_{m} C_{m}^{\dagger} C_{m}$$

$$\delta p_m = \delta t \Gamma_m \langle \Psi | C_m^{\dagger} C_m | \Psi \rangle$$

Solve the time dependent Schrödinger equation.

Use non-Hermitian Hamiltonian H which includes the decay part  $H_{\rm dec.}$ 

Key for non-Hermitian Hamiltonian: Jump operators  $C_m$  can be found from the dissipative part of the master equation.

For each channel m the jump probability is given by the time step size, decay rate, and decaying state occupation probability.



4. Ensemble average over  $\psi$  :s gives the density matrix and the expectation value of any operator A

$$\langle A \rangle(t) = \frac{1}{N} \sum_{i} \langle \psi_i(t) | A | \psi_i(t) \rangle$$



## **Measurement scheme interpretation**

Two-level atom in vacuum

Two-level atom MC evolution by

Total system evolution



 $H_{dec} = -\frac{i\hbar\Gamma}{2} |e\rangle\langle e|$ 

Jump operator

Non-Hermitian Hamiltonian

 $\left| P = \delta t \Gamma \left| c_{\rho} \right|^2 \right|$ 

Jump probability

Measurement scheme: continuous measurement of photons in the environment.

 $(c_g|g\rangle + c_e|e\rangle) \otimes |0\rangle \rightarrow$  $(c'_{g}|g\rangle + c'_{e}|e\rangle) \otimes |0\rangle + \sum_{\lambda} c_{\lambda}|g\rangle \otimes |1_{\lambda}\rangle$ 

Ontinuous measurement of the environmental state gives conditional pure state realizations for the open system The open system evolution is average of these realizations





## Questions: • What happens when the decay rates depend on time?

What happens when the decay rates turn temporarily negative?

$$\frac{d\rho(t)}{dt} = -i[H,\rho_S] + \sum_k \gamma_k(t) \left( A_k \rho_S(t) A_k^{\dagger} - \frac{1}{2} A_k^{\dagger} A_k \rho_S(t) - \frac{1}{2} \rho_S A_k^{\dagger} A_k \right)$$



## 2. Non-Markovian Quantum Jumps

Piilo, Maniscalco, Härkönen, Suominen: PRL 2008



## Markovian vs. non-Markovian evolution (1)

Markovian dynamics: Decay rate constant in time. Non-Markovian dynamics: Decay rate depends on time, obtains temporarily negative values.



## Markovian vs. non-Markovian evolution (1)

Markovian dynamics: Decay rate constant in time. Non-Markovian dynamics: Decay rate depends on time, obtains temporarily negative values.

Example: 2-level atom in photonic band gap.



Markovian description of quantum jumps fails, since gives negative jump probability.

For example: negative probability that atom emits a photon.



## Starting point:

General non-Markovian master equation local-in-time:

$$\frac{d\rho(t)}{dt} = \frac{1}{i\hbar} \left[ H_S, \rho \right] + \sum_m \Delta_m(t) C_m \rho C_m^{\dagger} - \frac{1}{2} \sum_m \Delta_m(t) \left( C_m^{\dagger} C_m \rho + \rho C_m^{\dagger} C_m \right)$$

- Jump operators C<sub>m</sub>
- Time dependent decay rates  $\Delta_m(t)$ .
- Decay rates have temporarily negative values.



## Starting point:

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- Jump operators C<sub>m</sub>
- Time dependent decay rates  $\Delta_m(t)$ .
- Decay rates have temporarily negative values.

Example: 2-level atom in photonic band gap. Jump operator C for positive decay:  $\sigma_-=|g
angle\langle e|$ 



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## Non-Markovian quantum jump (NMQJ) method

Quantum jump in negative decay region: The direction of the jump process reversed

$$\begin{split} |\psi\rangle \overleftrightarrow{} |\psi'\rangle &= \frac{C_m |\psi\rangle}{||C_m |\psi\rangle||}, \quad \Delta_m(t) > 0\\ |\psi\rangle \overleftrightarrow{} |\psi'\rangle &= \frac{C_m |\psi\rangle}{||C_m |\psi\rangle||}, \quad \Delta_m(t) < 0 \end{split}$$

Negative rate process creates coherences

Jump probability:

$$P = \frac{N}{N'} \delta t |\Delta_m(t)| \langle \psi | C_m^{\dagger} C_m | \psi(t) \rangle$$

N: number of ensemble members in the target state N': number of ensemble members in the source state

The probability proportional to the target state!



## NMQJ example

For example: two-level atom  $\sigma_{-}=|g
angle\langle e|$ 



Jump probability: 
$$P = \frac{N_0}{N_g} \delta t |\Gamma(t)| |\langle \psi_0 | e \rangle|^2$$

The essential ingredient of non-Markovian system: memory. Recreation of lost superpositions.



## Non-Markovian quantum jumps

## In terms of probability flow in Hilbert space:

**Positive rate** 





## Non-Markovian quantum jumps

## In terms of probability flow in Hilbert space:

**Positive rate** 





## Non-Markovian quantum jumps

## In terms of probability flow in Hilbert space:

**Positive rate** 



Memory in the ensemble: no jump realization carries memory of the I jump realization; I jump realization carries the memory of 2 jumps realization...

#### Negative rate: earlier occurred random events get undone.





$$\frac{d}{dt}\rho = -i[H(t),\rho]$$

$$+ \sum_{k} \Delta_{k}^{+}(t) \left[ C_{k}(t)\rho C_{k}^{\dagger}(t) - \frac{1}{2} \left\{ C_{k}^{\dagger}(t)C_{k}(t),\rho \right\} \right]$$

$$- \sum_{l} \Delta_{l}^{-}(t) \left[ C_{l}(t)\rho C_{l}^{\dagger}(t) - \frac{1}{2} \left\{ C_{l}^{\dagger}(t)C_{l}(t),\rho \right\} \right]$$

 $\rho(t) = \sum_{\alpha} \frac{N_{\alpha}(t)}{N} |\psi_{\alpha}(t)\rangle \langle \psi_{\alpha}(t)| \qquad \text{ensemble}$ 

Deterministic evolution and positive channel jumps as before... Negative channel with jumps

 $D^{j_{-}}_{\alpha \to \alpha'}(t) = |\psi_{\alpha'}(t)\rangle \langle \psi_{\alpha}(t)|$ 

where the source state of the jump is

 $|\psi_{\alpha}(t)\rangle = C_{j_{-}}(t)|\psi_{\alpha'}(t)\rangle/||C_{j_{-}}(t)|\psi_{\alpha'}(t)\rangle||$ 

...and jump probability for the corresponding channel

$$P_{\alpha \to \alpha'}^{\mathcal{I}_{-}}(t) = \frac{N_{\alpha'}(t)}{N^{\mathcal{I}_{-}}(t)} |\Delta_{j_{-}}(t)| \delta t \langle \psi_{\alpha'}(t) | C_{j_{-}}^{\dagger}(t) C_{j_{-}}(t) | \psi_{\alpha'}(t) \rangle.$$





Basic idea:

Weighting jump path with jump probability and deterministic path with no-jump probability gives the master equation (as in MCWF)

The ensemble averaged state over dt is



Here, other quantities are similar as in original MCWF except: P's: jump probabilities D's: jump operators

By plugging in the appropriate quantities gives the match with the master equation !









 Rebentrost, Chakraborty, Aspuru-Guzik:
 "Non-Markovian quantum jump in excitonic energy transfer" The Journal of Chemical Physics 2009

### • Ai, Fan, Jin, Cheng:

"An efficient quantum jump method for coherent energy transfer dynamics in photosynthetic systems under the influence of laser fields" (includes comparison to HOM) New Journal of Physics 2014

#### Renaud, Grozema:

"Intermolecular Vibrational Modes Speed Up Singlet Fission in Perylenediimide Crystals"

The Journal of Physical Chemistry Letters 2015



## 3. Unifying framework: Rate operator quantum jumps (ROQJ)

Smirne, Caiaffa, Piilo PRL 2020 Earlier work with QSD: Caiaffa, Smirne, Bassi: PRA 2017



## What is the problem? Example

## "Eternal" non-Markovian master equation $\dot{\rho} = \frac{1}{2} \sum_{k=1}^{3} \gamma_k(t) [\sigma_k \rho \sigma_k - \rho], \quad \text{Hall et al PRA 2014}$

- $\odot$  Pauli matrices  $\sigma_k$
- Decoherence rates  $\gamma_1(t) = \gamma_2(t) = 1$ ,  $\gamma_3(t) = -\tanh t < 0$  for all t > 0
- Map CP but breaks CP-divisibility for all t > 0
- "Eternal" non-Markovian according to RHP criteria

## **•**...however, P-divisible for all t > 0



Why Markovian MCWF does not work? • Rate for  $\sigma_z$  jump  $\gamma_3(t) = -\tanh t_1 < 0$  for all t>0 gives negative jump probability  $P_j = \delta t \Gamma p_e < 0$ Why non-Markovian NMQ does not work? • Reverse jump probility  $P_{\alpha \to \alpha'}^{j_-}(t) = \left(\frac{N_{\alpha'}(t)}{N_{\alpha}(t)}\right) \Delta_{j_-}(t) |\delta t \langle \psi_{\alpha'}(t) | C_{j_-}^{\dagger}(t) C_{j_-}(t) | \psi_{\alpha'}(t) \rangle.$ singularity in the jump probability (can not cancel something which never happened) Note: however, fully classical Markovian

description with ancillas exists



- Processes exists which always break CP-divisibility and always preserve P-divisibility
- "In-between" Markovian and non-Markovian
- No known jump descriptions without ancillas- exists

## What is the most general stochastic jump description valid in all regimes?

Reminder about maps  $\Phi_{t,0} = \Phi_{t,s} \Phi_{s,0}$ : CP-divisibility:  $\Phi_{t,s}$  is CP P-divisibility:  $\Phi_{t,s}$  is P



## ROQJ

# ROQJ - Rate operator quantum jumpsMaster equation

$$\mathscr{L}[\rho(t)] = -\frac{i}{\hbar}[H_s,\rho(t)] + \sum_{\alpha=1}^{n^2-1} c_\alpha(t) \left( L_\alpha(t)\rho(t)L_\alpha(t)^{\dagger} - \frac{1}{2} \left\{ L_\alpha^{\dagger}(t)L_\alpha(t),\rho(t) \right\} \right)$$

• At this stage, consider P-divisible dynamics.

• Negative rates allowed, as long as **transition rate operator** positive semi-definite (non-negative eigenvalues) for any pure state  $|\psi(t)\rangle$ 

$$W_{\psi(t)}^{J} = \sum_{\alpha=1}^{n^{2}-1} c_{\alpha}(t) (L_{\alpha}(t) - \ell_{\psi(t),\alpha}) |\psi(t)\rangle \langle \psi(t)| (L_{\alpha}(t) - \ell_{\psi(t),\alpha})^{\dagger}$$
  
$$\ell_{\psi(t),\alpha} = \langle \psi(t)| L_{\alpha}(t) |\psi(t)\rangle$$
  
$$H_{\psi(t)} = H_{S}(t) - \frac{i\hbar}{2} \sum_{\alpha=1}^{n^{2}-1} c_{\alpha}(t) \times \left(L_{\alpha}^{\dagger}(t)L_{\alpha}(t) - 2\ell_{\psi(t),\alpha}^{*}L_{\alpha}(t) + |\ell_{\psi(t),\alpha}|^{2}\right)$$

deterministic evolution



## ROQJ

## • We can diagonalize and write with eigenvalues

$$W_{\psi(t)}^{J} = \sum_{j=1}^{n-1} \lambda_{j}(t) \left| \varphi_{\psi(t),j} \right\rangle \left\langle \varphi_{\psi(t),j} \right|$$
$$= \sum_{j=1}^{n-1} V_{\psi(t),j} \left| \psi(t) \right\rangle \left\langle \psi(t) \right| V_{\psi(t),j}^{\dagger},$$

## Here defined

$$V_{\psi(t),j} = \sqrt{\lambda_j(t)} \left| \varphi_{\psi(t),j} \right\rangle \langle \psi(t)$$

Transfers from current state to eigenstate of rate operator with a rate given by the eigenvalue

## • Therefore deterministic evolution interrupted by jumps

 $|\psi(t)\rangle \rightarrow \frac{V_{\psi(t),j} |\psi(t)\rangle}{\|V_{\psi(t),j} |\psi(t)\rangle\|}$ 

## which occur with probability

 $p_j(t) = \|V_{\psi(t),j} |\psi(t)\rangle \|^2 \mathrm{d}t.$ 





## **ROQJ** - example



Simulation produces analytical results
 In general possible to prove match with master equation



- Markovian MCWP has measurement scheme interpretation
- No known measurement schemes in non-Markovian regime (Diosi PRL 2008; Gambetta, Wiseman PRL 2008)

Where is the border between the two? How do we lose measurement scheme interpretation?



- It is possible to show in mathematically rigorous manner that the method has continuous measurement scheme interpretation following Barchielli and Belavkin JPhysA 1991
- Therefore measurement scheme exists for master equations with negative rates as long as P-divisible
- Operations for the count (jump) defined by  $\mathcal{I}_{\omega_t,j}\rho = V_{\omega_t,j}\rho V_{\omega_t,j}^{\dagger}t \qquad j = 1, \dots n$   $V_{\psi(t),j} = \sqrt{\lambda_j(t)} |\varphi_{\psi(t),j}\rangle\langle\psi(t)|$
- Trajectory upto time t  $\omega_t = (t_1, j_1; t_2, j_2; \dots t_m, j_m)$
- Orresponding state transformation

$$\rho \mapsto \frac{\mathcal{I}_{\omega_t,j}\rho}{\operatorname{Tr}\left\{\mathcal{I}_{\omega_t,j}\rho\right\}}$$



• Operations for the count (jump) defined by

 $\mathscr{I}_{\omega_t,j}\rho = V_{\omega_t,j}\rho V_{\omega_t,j}^{\dagger} \mathrm{d}t \qquad j = 1, \dots n - 1, \qquad \qquad V_{\psi(t),j} = \sqrt{\lambda_j(t)} \left|\varphi_{\psi(t),j}\right\rangle \langle \psi(t)|$ 

## Important points:

- The operations are conditioned on the whole trajectory
- For example: the past jumps influence what is the current state and may influence the diagonalization and the construction of the corresponding jump operator
- Measurement scheme: In addition of measurement record requires also computational resources and time dependent basis for the measurement depending on the trajectory

Does this describe memory effects or not?



Schematics with "2D" Bloch sphere Type of stochastic realizations

Markovian MCWF



The jumps take the realizations to same state no matter where located prior to jump



In-between region with ROQJ



• The jumps take the realizations to states which depend on the current state and therefore also on prior sequence

Non-Markovian quantum jumps



The jumps take the realization to state which the realization had in the past (recovery of lost info)







## No memoryAlways to same state

- "Dependence" from the past for single realization
- Where you go next depends where you are at the moment
- On the level of density matrix monotonic loss of coherence

Does this describe memory or not?

## non-Markovian



- Backflow of info for single realizations
- Backflow of info for density operator
- Going back where you were before







## No memoryAlways to same state

- "Dependence" from the past for single realization
- Where you go next depends where you are at the moment
- On the level of density matrix monotonic loss of coherence

Note that also in Markovian case dependence from the past in terms of jump probability via

Does this describe memory or not?

non-Markovian



- Backflow of info for single realizations
- Backflow of info for density operator
- Going back where you were before

The source of NM?

Also classical Markovian rate equation solution exists (Megier et al SciRep 2017)



General scheme including non-Markovian regime

 Divide transition rate operator to positive and negative eigenvalue parts

$$\begin{split} W_{\psi_k(t)}^J &= \sum_{j=1}^{n-1} \lambda_j(t) \left| \varphi_{\psi_k(t),j} \right\rangle \left\langle \varphi_{\psi_k(t),j} \right| \\ &= \sum_{j^+} \lambda_{j^+}(t) \left| \varphi_{\psi_k(t),j^+} \right\rangle \left\langle \varphi_{\psi_k(t),j^+} \right| - \sum_{j^-} \left| \lambda_{j^-}(t) \right| \left| \varphi_{\psi_k(t),j^-} \right\rangle \left\langle \varphi_{\psi_k(t),j^-} \right| \right\rangle \end{split}$$

- For positive part, use the earlier scheme
- For negative part, calculate the jump probabilities in similar manner as for NMQJ and use in the reverse jumps as a source the eigenstates of the transition rate operator



## ROQJ - in non-P-div region

Reverse jumps, corresponding to negative eigenvalues of W, are

$$B_{\psi_k(t),\psi_{k'}(t),j^-} = \sqrt{\left|\lambda_{\psi_{k'}(t),j^-}\right|} \left|\psi_{k'}(t)\rangle\langle\psi_k(t)\right|$$

• Source of reverse jump is the eigenstate of W:  $|\psi_k(t)\rangle = |\varphi_{\psi_{k'}(t),j^-}\rangle$ 

• Probability given by  $p_{j^-}^{(k \to k')}(t) = \frac{N_{k'}(t)}{N_k(t)} \left| \lambda_{\psi_{k'}(t), j^-} \right| dt$ 

## **One general framework for all regimes:**

When P-div: jumps to eig. states of rate operator W
When P-div: broken: jumps out of the eig. states of W
ROQJ works also when neither MCWF nor NMQJ works (when P-div. with negative rates)



## ROQJ - in non-Markovian (non-P-div) region

• 7-site driven system Unitary part:  $H_S = \sum_{i \neq j} \Omega_{i,j} |i\rangle \langle j|$ Jump (Lindblad) operators):  $L_{i,j} = |i\rangle \langle j|$  (49 of them) Jump rates (contain negative regions):  $c(t) = 0.5[(1 - e^{-0.5t})0.3 + e^{-0.3t} \sin(4.5t)]$ 





## Is the rate operator unique? Can we have a family of rate operators?

Chruscinski, Luoma, Piilo, Smirne arXiv:2009:11312 (work still ongoing and developed...)



• Master equation  $\dot{\rho} = \frac{1}{2} \sum_{k=1}^{3} \gamma_k(t) [\sigma_k \rho \sigma_k - \rho],$ 

## For example

Rate operator RI  $\mathbf{R1} = \sum_{k=1} \gamma_k(t) \sigma_k |\psi\rangle \langle \psi | \sigma_k + \sum_k \gamma_k(t) |\psi\rangle \langle \psi |$ k=1 $K1(t) = \frac{i}{2}\gamma(t)1$  deterministic evolution Rate operator R2  $\mathbf{R2} = \sum \gamma_k(t)\sigma_k |\psi\rangle \langle \psi | \sigma_k + (\gamma_1(t) + \gamma_2(t)) |\psi\rangle \langle \psi |$  $K2(t) = \frac{i}{2} [\gamma_1(t) + \gamma_2(t)] 1$  deterministic evolution Chruscinski, Luoma, Piilo, S Chruscinski, Luoma, Piilo, Smirne

arXiv.2009.11312



### With RI



Chruscinski, Luoma, Piilo, Smirne



With R2



Chruscinski, Luoma, Piilo, Smirne arXiv:2009:11312



## CONCLUSIONS

- MCWF Monte Carlo Wave Function (1992)
- NMQJ Non-Markovian Quantum Jumps (2008)
- **ROQJ** Rate Operator Quantum Jumps (2020)
  - General starting point for any regime
  - Unifies the framework for using quantum jumps to describe open system dynamics
  - Measurement scheme for master equations with negative rates (P-div, no ancillas used)
  - For the direction of having families of rate operators...



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