TRIUMF

Syd Kreitzman Physical Sciences Division … MuSR 2025 St. John's Newfoundland

The Strong Collision Model using a Projector Operator Formalism

The strong collision is formulated using a projection operator formalism. The formalism allows for a compact formal "solution" to the problem. It also has a direct connection with cumulant stochastic averaging theory. Using the projector operator ¶ = $|p_e\rangle\langle 1|$ the formal solution to the strong collision model can be written as

$$|\rho_{sc}\rangle = e^{(i[\omega]-\nu+\nu\P)t}|p_e\rangle = \mathcal{T}_L exp[\int_0^t e^{(-\nu+\nu\P)(t-\tau)} i[\omega] d\tau]|p_e\rangle \rightarrow \rho_{sc}(t) = \langle 1|\rho_{sc}\rangle \qquad \text{(1a b} \rightarrow \text{c)}$$

Some Definitions:

$|\ \rangle$, a ket (column) vector in the "frequency site" space; the $|p_e\rangle$ ket is filled with the normalized equilibrium site occupation probabilities: $|p_e^n\rangle$ has the equilibrium probability in the n'th location, else 0. $|1^n\rangle$ has 1 in the n'th location, else 0.

$\langle\ |$, a bra (row) vector: $\langle 1|$ bra is filled with 1's: $\langle 1^n| = 1$ in the n'th location, else 0

¶ = $|p_e\rangle\langle 1|$ a projection operator in the site space

i.e. ¶ ¶ = $|p_e\rangle\langle 1|p_e\rangle\langle 1| = |p_e\rangle\langle 1|$ since $\langle 1|p_e\rangle = 1$

$[\omega]$ is a diagonal matrix with entries ω^n associated with the probability $|p_e^n\rangle$

Strong Collision (SC) Site Migration Dynamics can be then represented by the DE

$$|\dot{p}(t)\rangle = (-\nu+\nu\P)|p(t)\rangle \rightarrow |p(t)\rangle = e^{(-\nu+\nu\P)t}|p(0)\rangle$$

$$\text{using } e^{\nu\P t} = 1 + (e^{\nu t}-1)\P \text{ then } |p(t)\rangle = e^{-\nu t}|p(0)\rangle + (1-e^{-\nu t})|p_e\rangle$$

i.e. The initial distribution decays with rate ν and evolves into the equilibrium distribution independent of its initial values. $|p_e\rangle$ is an eigenket of $\nu(-1+\P)$ and $\exp((-\nu+\nu\P)t)$ with eigenvalues 0 and 1, independent of ν.

Adding Spin Polarization Evolution to the Equation of Motion

We adopt the perspective that there are a set frequencies and equilibrium probabilities that define the signal we see experimentally. When the test spin probe makes a transition into a new site $|1^n\rangle$ it starts to accumulate phase at the rate ω^n which defines that site. The 2d set of $\{\omega^n, p_e^n\}$ defines the problem … obvious for TF less so for ZF or LF. Since phase can be represented as a complex number, letting the occupation probabilities $|p\rangle$ transition into complex $|\rho\rangle$ kets allows one to immediately write down the DE for $|\rho\rangle$ as below.

$$|\dot{\rho}(t)\rangle = (i[\omega]-\nu+\nu\P)|\rho(t)\rangle \rightarrow |\ddot{\rho}(t)\rangle = (i[\omega]-\nu+\nu\P)|\dot{\rho}(t)\rangle \qquad \text{(2a} \rightarrow \text{b)}$$

Solving the Equation of Motion …. first thoughts

The obvious formal solution is (1a)of the abstract. Looks deceptively simple at first sight except for the fact that $[\omega]$ and ¶ don't commute. There is a vast literature on how to handle such DE's both theoretically and numerically, but they do not in account for the unique nature of the ¶ operator. For ν=0, equations (1) collapse to the initial static frequency resolved polarization function defined by the set of $|p_e\rangle$.

Equivalence to the Classical SC and beyond

There are several approaches with the easiest to treat the diagonal $i[\omega]$-ν term in (2) as the homogeneous one … to give the very recognizable solution

$$|\rho(t)\rangle = e^{(i[\omega]-\nu)t}|p_e\rangle + \nu\int_0^t e^{(i[\omega]-\nu)(t-\tau)}\P|\rho(\tau)\rangle d\tau \rightarrow \langle 1|\rho(t)\rangle = \rho(t) = \rho_s(t)e^{-\nu t} + \nu\int_0^t \rho_s(t-\tau)e^{-\nu(t-\tau)}\rho(\tau)d\tau \qquad (3)$$

However, in (2) there is in fact no special status to the $i[\omega]$-ν term, and one can choose to call the ¶ term as the "homogeneous" one and therefore write an "equally valid" alternate integral equation

$$|\rho(t)\rangle = e^{(-\nu+\nu\P)t}|p_e\rangle + \int_0^t e^{(-\nu+\nu\P)(t-\tau)} i[\omega]|\rho(\tau)\rangle d\tau = |p_e\rangle + \int_0^t (e^{-\nu(t-\tau)} + (1-e^{-\nu(t-\tau)})\P) i[\omega]|\rho(\tau)\rangle d\tau \qquad (4)$$

It will be seen that equations that share this kernel form the mathematical basis, for example, of the averaging prescription that is required when calculating the stochastic cumulants, if that's one's fancy, to approximate the dynamics. However, before delving into the details of that endeavour, the author has observed (and most likely re-observed) that one can straightforwardly formulate, and numerically execute, robust calculations for the dynamics of these systems as follows.

A Robust Standard Approach … i.e making sure the "first jump is done right"!

In the same fashion as one can iterate the standard integral equation (3) for the average polarization to higher orders, the shared structure of (2a and b) indicates that this just as easily be done for $\dot{\rho}(t)$, … i.e. one finds

$$\dot{\rho}(t) = \dot{\rho}_s(t)e^{-\nu t} + \nu e^{-\nu t}\int_0^t \dot{\rho}_s(t-\tau)\rho_s(\tau)d\tau + \nu^2\int_0^t \rho(t-\tau)e^{-\nu\tau}\int_0^\tau \dot{\rho}_s(\tau')\rho_s(\tau-\tau')d\tau' d\tau$$

The first observation is that the last term is very localized in the vicinity of the peak of $\dot{\rho}(\tau)$, so it should be valid to expand $\rho(t-\tau)$ into $\rho(t)-\tau\dot{\rho}(t)$. This results in the following 1st order DE (called approx$_5$ or a$_5$) is;

$$\rightarrow \dot{\rho}_{a_5}(t) = \frac{\dot{\rho}_s(t)e^{-\nu t} + \nu e^{-\nu t}\int_0^t \dot{\rho}_s(t-\tau)\rho_s(\tau)d\tau + \nu^2\rho_{a_5}(t)\int_0^t e^{-\nu\tau}\int_0^\tau \dot{\rho}_s(\tau')\rho_s(\tau-\tau')d\tau' d\tau}{1+\nu^2\int_0^t \tau e^{-\nu\tau}\int_0^\tau \dot{\rho}_s(\tau')\rho_s(\tau-\tau')d\tau' d\tau} \qquad (5)$$

Solving this DE numerically for the K-T function[1] is reasonably straight-forward and leads to the set of curves called Approx 5 below. If the denominator is set to 1, (i.e. ignoring the τ term in the expansion of ρ(t-τ)) the numerical solution given in Approx4, marginally less accurate than Approx5. Approx's 1→3 were not satisfying.

The SC Prescription for Stochastic Averaging …. "Abragam" and beyond

Eqn (4) can be iteratively expanded and one recognizes $e^{(-\nu+\nu\P)t} = e^{-\nu t} + (1-e^{-\nu t})\P$ as the SC stochastic propagation operator responsible for the time-ordered-probability weighted averaging of the phase accumulation of the individual components of $\rho(t)$, in terms of the moments of the frequency distribution. The functional forms are frequency independent and only need to be calculated once for all line shapes. The first three terms (i.e. short enough to display) are shown below. The use of a symbolic math package is required for higher orders.

$$|\rho(t)\rangle = (1+\sum f^k([\omega],\nu,t))|p_e\rangle \text{ with each } f^k \text{being } \mathcal{O}\omega^k \qquad (6)$$

$$f^1(t) = i\omega\frac{(1-e^{-\nu t})}{\nu} \qquad f^2(t) = -\omega^2\frac{1-e^{-\nu t}(1+\nu t)}{\nu^2} - \langle\omega^2\rangle\frac{2e^{-\nu t}+\nu(t+te^{-\nu t})-2}{\nu^2} \qquad (7)$$

$$f^3(t) = i\omega^3\left(\frac{e^{-\nu t}-1}{\nu^3} + \frac{t\,e^{-\nu t}}{\nu^3} + \frac{t^2e^{-\nu t}}{2\nu}\right) + \frac{i\langle\omega^2\rangle\omega e^{-\nu t}(\nu^2t^2 - 12\,e^{\nu t} + 6\nu t - \nu^2t^2e^{\nu t} + 6\nu te^{\nu t} + 12)}{2\nu^3}$$

Note that $\langle\omega\rangle = \langle 1|[\omega]|p_e\rangle = 0$ is assumed. At this stage one could simply be bold and claim that the series in (6) should be recast into the form of a time ordered stochastic cumulant expansion to be more "physically" relevant.

$$\rho^n(t) = \rho_e^n\mathcal{T}_L e^{<\int_0^t i\omega_n(\tau)d\tau>_{sc}} \qquad (8a)$$

$$\stackrel{def?}{=} \rho_e^n e^{f_n^1(t)+f_n^2(t)-(1/2)(f_n^1(t))^2+f_n^3(t)-f_n^1(t)f_n^2(t)+(1/3)(f_n^1(t))^3+\mathcal{O}^c(\{f\}_n^{>3})}\ldots \qquad (8b)$$

With this perspective one can refer to the results below when taking the f^1+f^2 terms only into the cumulant … and then attempting to improve the result by adding in f^3 ➛ a "judicious" selection of the # of terms is needed.

Mathematical "Formalities"

One can formalize/interpret 8(b) . First apply the "interaction" picture identity, valid for non-commuting O_1 O_2

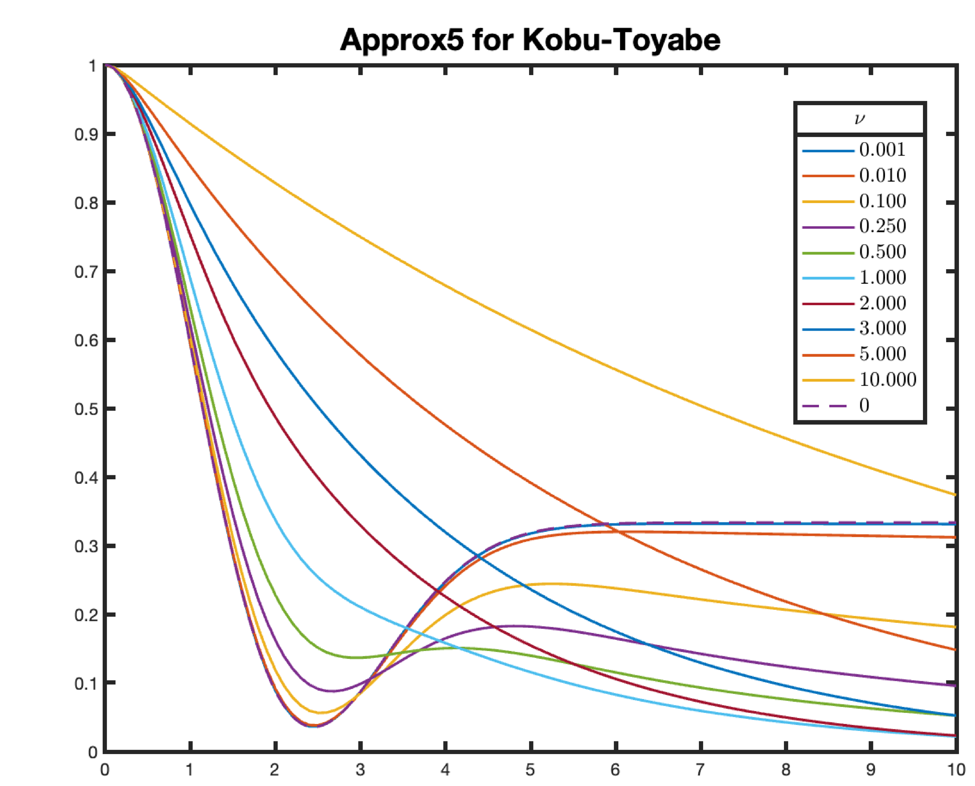
$$e^{(O_1+O_2)t} = e^{O_1t}\mathcal{T}_L exp[e^{\int_0^t e^{-O_1\tau}O_2e^{O_1\tau}d\tau}] \qquad (= \mathcal{T}_R exp[e^{\int_0^t e^{O_1\tau}O_2e^{-O_1\tau}d\tau}]e^{O_1t}) \qquad (9)$$

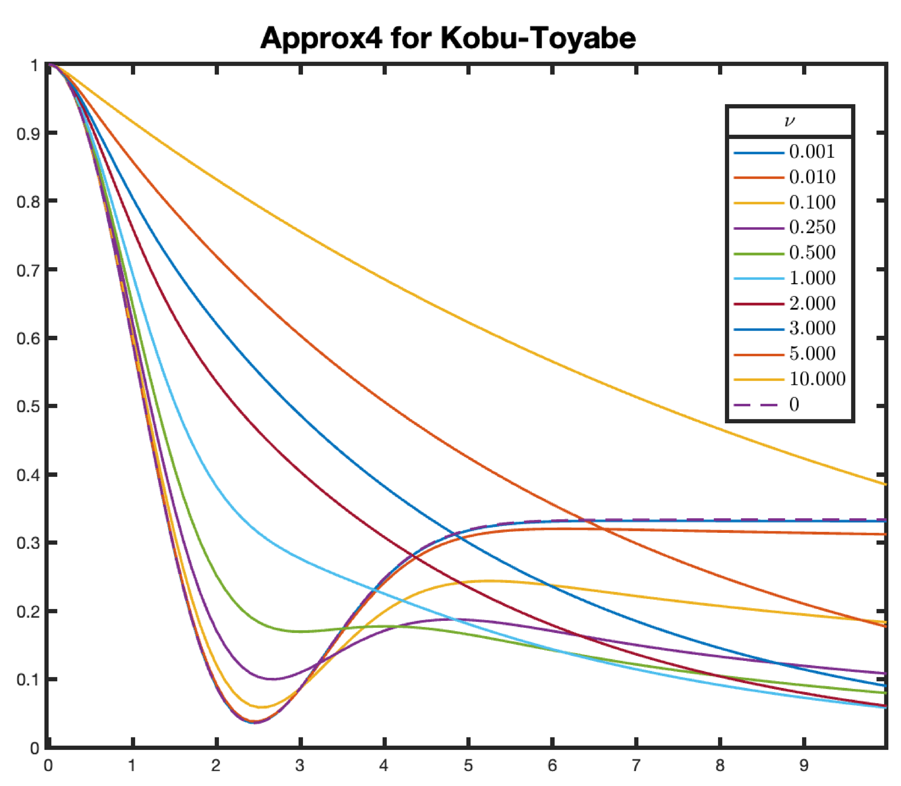
with $O_1 = \nu(-1+\P)$ and $O_2 = i[\omega]$ … then using the T_L version

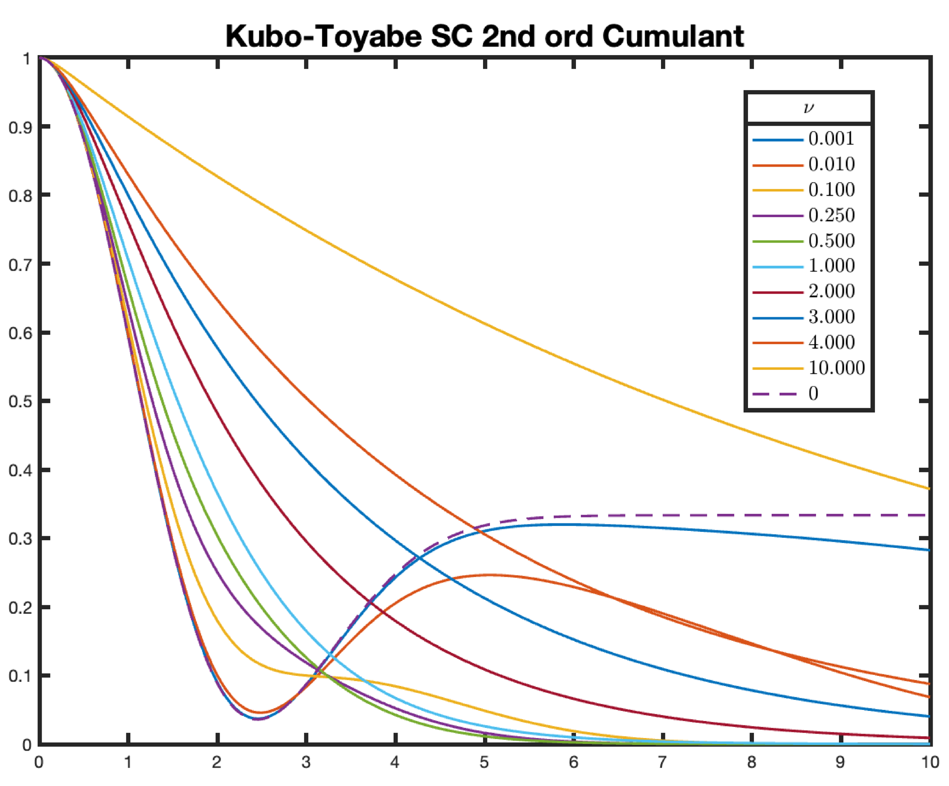
$$e^{(i[\omega]-\nu+\nu\P)t}|p_e\rangle = e^{(-\nu+\nu\P)t}\mathcal{T}_L exp[e^{\int_0^t e^{(\nu-\nu\P)\tau}i[\omega]e^{(-\nu+\nu\P)\tau}d\tau}]|p_e\rangle$$

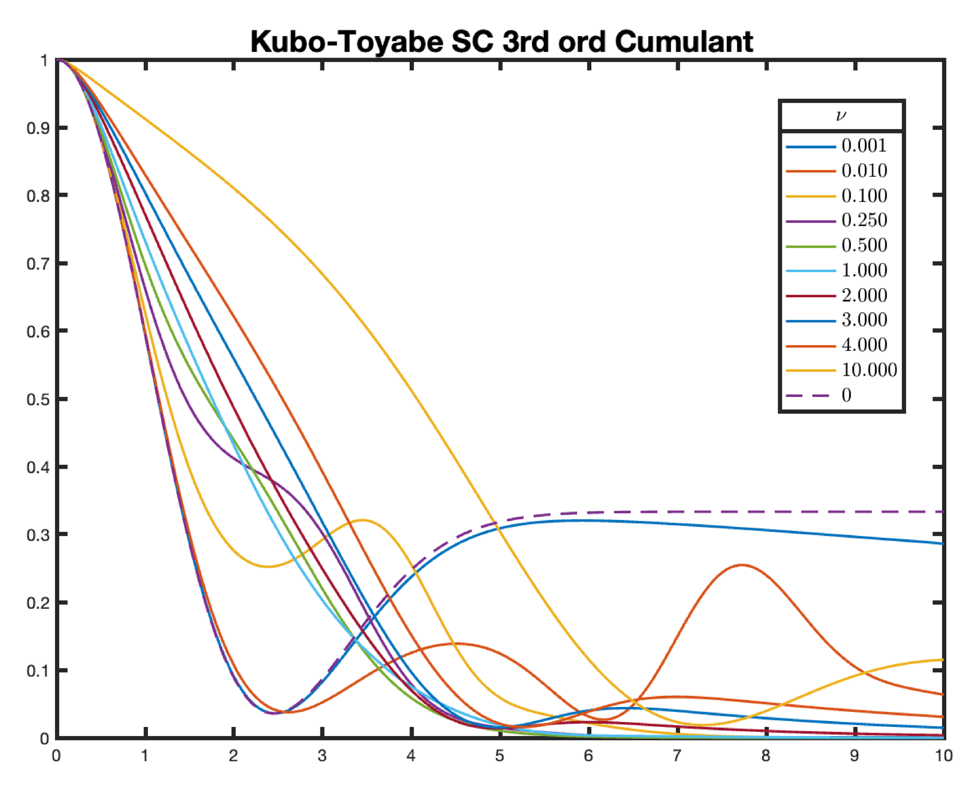
Adopting the physicist's short-cut (which can be substantiated by examining the detailed structure of the of the time-ordered infinite series) that $|p_e\rangle$ is time independent, and can therefore be "moved" into the T_L argument, use the fact that $\exp((-\nu+\nu\P)\tau)|p_e\rangle = |p_e\rangle$, and then move it back out of the T_L argument, gives

$$|\rho(t)\rangle = \mathcal{T}_L exp[\int_0^t e^{(-\nu+\nu\P)(t-\tau)}i[\omega]d\tau]|p_e\rangle = \mathcal{T}_L exp[\int_0^t (e^{-\nu(t-\tau)} + (1-e^{-\nu(t-\tau)})\P)i[\omega]d\tau]|p_e\rangle \qquad (10)$$









[1] R. Kubo and T. Toyabe, in Magnetic Resonance and Relaxation, edited by R. Blinc (North-Holland, Amsterdam, 1967),

Discovery Accelerated