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Introduction of the Floquet-Magnus expansion in solid-state nuclear magnetic resonance spectroscopy

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In this article, we present an alternative expansion scheme called Floquet-Magnus expansion (FME) used to solve a time-dependent linear differential equation which is a central problem in quantum physics in general and solid-state nuclear magnetic resonance (NMR) in particular. The commonly used methods to treat theoretical problems in solid-state NMR are the average Hamiltonian theory (AHT) and the Floquet theory (FT), which have been successful for designing sophisticated pulse sequences and understanding of different experiments. To the best of our knowledge, this is the first report of the FME scheme in the context of solid state NMR and we compare this approach with other series expansions. We present a modified FME scheme highlighting the importance of the (time-periodic) boundary conditions. This modified scheme greatly simplifies the calculation of higher order terms and shown to be equivalent to the Floquet theory (single or multimode time-dependence) but allows one to derive the effective Hamiltonian in the Hilbert space. Basic applications of the FME scheme are described and compared to previous treatments based on AHT, FT, and static perturbation theory. We discuss also the convergence aspects of the three schemes (AHT, FT, and FME) and present the relevant references.


I. INTRODUCTION

Much progress has been made in the application of solid-state nuclear magnetic resonance (NMR) to elucidate molecular structure and dynamics in systems not amenable to characterization by any other way. The importance of solid-state nuclear magnetic resonance stands in its ability to determine accurately intermolecular distances¹, ² and molecular torsion angles.³, ⁴ The methods have been used to systems including both microscopically ordered preparations such as membrane proteins,⁵-⁸ nanocrystalline proteins,⁹-¹¹ amyloid fibrils,¹²-¹⁶ and also disordered or amorphous systems such as glasses.¹⁷ Site-specific resolution can be obtained either by uniaxial orientation of the sample with respect to the static magnetic field¹⁸ or, through magic-angle sample spinning (MAS). Nowadays, MAS is widely used to obtain high resolution NMR spectroscopy of solids because of its effect of averaging out the orientation-dependent component of nuclear spin interactions, principally chemical shifts anisotropic and magnetic dipolar couplings. This technique can be combined with cross polarization to increase the spectral sensitivity of rare and low-gamma nuclei such as ¹³C, ¹⁵N (Ref. ¹⁹) in biopolymers or other organic solids. Therefore, MAS NMR techniques have improved to the point where complete structure determination is possible.¹⁰, ¹², ¹³

As the technique of MAS spreads to the field of solid state NMR, the concept of average Hamiltonian theory (AHT),²⁰ which is the main theoretical tool to describe the effect of time-dependent interactions, was found to be less descriptive to rotating systems, such as sample-spinning experiments.²¹ However, these types of experiments were found to be more conveniently described using Floquet theory.²²–²⁴ In this work, we introduce the fusion of AHT and FT as provided by the Floquet-Magnus expansion (FME) that can be very useful in simplifying calculations and also for providing a more intuitive understanding of spin dynamics processes.

The purpose of this article is to introduce the FME scheme to solid-state NMR, to give a general and coherent framework of the scheme, and to compare its use in solid-state NMR with other averaging approaches. Similarly to the AHT and FT theory, the primary aim of the FME approach is to provide a scheme to build an approximation of the Hamiltonian describing the stroboscopic evolution of the system over several periods. The FME approach is essentially distinguished from AHT with its function Λ(t) which provides an easy and alternative way for evaluating the spin behavior in between the stroboscopic observation points. The FME approach is in fact the fusion of the two major methods used to describe the spin dynamics in solid-state NMR: the average Hamiltonian theory based on the Magnus expansion (ME) and the Floquet theory based on the Fourier expansion. The first method (AHT) was developed by Haeberlen and Waugh in 1968²⁰ and is appropriate for stroboscopic sampling. The AHT technique does not satisfactorily describe the case of MAS spectra because in this case, the signal is usually observed continuously with a time resolution much shorter than the rotor period.²⁵, ²⁶ Nevertheless, in a variety of cases, MAS

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experiments have been successfully analyzed using AHT, which yields an effective Hamiltonian averaged over some cycle time of a periodic pulse sequence. From its natural formulation, AHT has been gradually applied to almost every kind of situation, sometimes abusively. Some examples that lend to the application of AHT include the use of a prerequisite of the approach to design a frequency-modulated analog of TPPM and explored other possibilities involving simultaneous phase and frequency modulation. Similarly, Eden and Levitt utilized symmetry arguments based on AHT to develop optimized heteronuclear decoupling sequences involving rotor-synchronized pulse sequences whose fundamental element “C” is a 2π pulse. Therefore, despite the emergence of alternative approaches such as the Floquet theory, the exact effective Hamiltonian theory, and the Floquet expansion, which have advantages in some circumstances, the average Hamiltonian theory still remains of central importance in theory of multiple-pulse NMR. All approaches are equivalent to first order. With the increase of the level of sophistication of NMR experiments, second order terms are of increasing importance, such as in diffusion experiment.

The second method (FT), which was first proposed by Shirley and introduced to NMR by Veega and Maricq, provides a more universal approach for the description of the full time dependence of the response of a periodically time-dependent system. It allows the computation of the full spinning sideband pattern that is of importance in many MAS experimental circumstances to obtain information on anisotropic sample.

The Floquet theory is based on the transformation of the N-dimensional Hilbert space of the original problem into an infinite-dimensional Floquet-Hilbert space, where the periodically time-dependent Hamiltonian becomes time-independent Floquet Hamiltonian. For numerical computations, this requires truncation of the infinite dimension of the Floquet-Hilbert space. Some aspects of a formal Floquet theory have been discussed by Sambe, Yajima and Kitada, Moore, and Dzyublik. Also, a generalized Floquet Hamiltonian based on the quasi-energy operator have been explored by Howland, Bunimovich et al., and Blekher et al. Hilbert space-Floquet theory was extended to Liouville space, using a Floquet Liouville supermatrix approach by Chu, Ho, Wang, and Jiang and applied by Kavanagh and Silbey. The Floquet theory was further simplified by the use of the multipole basis proposed by Sanctuary, which exploits the rotational invariance properties of irreducible tensor operators using a multispin basis. Providing the basis for a formal description of FT or AHT is not the goal of this paper, but, instead, the relevance and potential of the Floquet-Magnus expansion scheme in solid-state NMR over the other averaging schemes such as AHT and FT.

The AHT and FT result respectively in average and effective Hamiltonians that are expanded in a set of terms of increasing orders. These Hamiltonians are in general connected with stroboscopic detection schemes. In the case of AHT, the time evolution between the detection points is not described. In single-mode Floquet theory, the stroboscopic Hamiltonian is again connected to stroboscopic detection schemes. However, this theory provides the option to evaluate the spin evolution between the time points of detection. In contrast to common approaches of AHT and FT, the main advantage of the FME scheme is to overcome the limitations of the stroboscopic detection schemes. In the Floquet-Magnus approach, even when the first and second order $F_1$ and $F_2$ of the effective Hamiltonian are identical to their counterparts in AHT and FT, the $\Lambda_{1,2}(t)$ functions provide an easy way for evaluating the spin evolution during “the time in between” through the Magnus expansion of the operator connected to this part of the evolution. $\Lambda_1(t)$ and $\Lambda_2(t)$ are connected to the appearance of features like spinning sidebands in MAS. The evaluation of $\Lambda_1(t)$ and $\Lambda_2(t)$ is useful especially for the analysis of the non-stroboscopic evolution. For example, in the case of C7, for non-stroboscopic detection scheme, they can be used to estimate the intensity of the spinning sidebands manifold in the double-quantum dimension. Higher order effects ($F_3, \Lambda_3(t)$) can also be evaluated using the FME approach, in a way easier than in the case of AHT or FT. The FME scheme can also be extended to multimode Hamiltonians for Hilbert space analysis especially in the incommensurate case. To the best of our knowledge, we present here the first report highlighting the basics of the FME scheme and compare this approach with the other series expansions. We present a generalized FME scheme, based on the importance of the boundary conditions (at the origin of time), which provides a natural choice for the operators $\Lambda_n(0)$ to simplify calculation of higher order terms and allows FT to be managed in the Hilbert space. Applications of the FME scheme are described and compared to previous treatments based on AHT, FT, and static perturbation theory (SPT). We also discuss the convergence aspects of the three schemes (AHT, FT, and FME) and present the relevant references.

An outline of the paper is as follows. In Sec. II, we describe the FME with a brief illustration of the ME and the celebrated Floquet theorem (FT), which ensures the factorization of the solution in a periodic part and a purely exponential factor. We explicitly give the first contributions to the Floquet-Magnus expansion with the addition of higher order effects ($F_3, \Lambda_3(t)$) as an improvement to the two first-order terms $F_{1,2}$ and $\Lambda_{1,2}(t)$. In Sec. III, we discuss the convergence aspects of the three schemes (ME, FT, and FME) and present the relevant references. In Sec. IV, we compare the three schemes and we extend the comparison to the Floquet-Van Vleck approach and the static perturbation theory. Special emphasis is put on the choice of the periodic boundary conditions $\Lambda_n(0)$. The application of the approach to the case of a multimodal Hamiltonian is also briefly discussed. Section V of the paper summarizes our conclusions.

II. THEORY

A. The Floquet theorem

Floquet theory is a branch of the theory of ordinary differential equations relating to the class of solutions to linear differential equations of the form (in this paper we use a
formulation in connection with quantum mechanics but without loss of generality)
\[ i \frac{dU}{dt} = H(t)U(t), \]  
(1)

with \( U(0) = I \) as initial condition. \( H(t) \) is a complex \( n \times n \) matrix-valued function and its matrix elements are integrable periodic functions of time \( t \) with period denoted \( T \). The Floquet theorem allows one to write
\[ U(t) = P(t) \exp(-iFt), \]  
(2)

where \( F \) and \( P(t) \) are \( n \times n \) matrices. \( P(t) \) is a periodic function of time with period \( T \), i.e., \( P(t) = P(t + T) \) and \( F \) is constant. In NMR, this structure is exploited in many situations including time-dependent periodic magnetic fields or sample spinning, which is the focus of this paper. The structure of \( U(t) \) Eq. (2) was exploited in two ways by Levante et al.\(^{38}\)

The first one consists in performing a Fourier expansion of the formal solution, leading to an infinite system of linear differential equations with constant coefficients. The price of this approach is to handle an infinite dimension that can only be resolved numerically by truncation. The second approach is of perturbative nature and applied directly to the form of Eq. (2) as following:
\[ P(t) = \sum_{n=1}^{\infty} P_n(t), \]  
(3)

\[ F = \sum_{n=1}^{\infty} F_n. \]  
(4)

Each term \( F_n \) in Eq. (3) is fixed such that \( P_n(t) = P_n(t + T) \) to ensure the Floquet structure Eq. (2) at any order of approximation.

B. The Magnus expansion

The ME provides an exponential representation of the solution of a first order linear homogeneous differential equation for a linear operator. It is introduced in Eq. (1) by the following form of the propagator:
\[ U(t) = e^{-i\Omega(t)}. \]  
(5)

With the help of the Wilcox formula,\(^{57}\) Eq. (1) can be rewritten as
\[ i \frac{dU}{dt} = i \frac{d}{dt} [e^{-i\Omega(t)}] = \left\{ \int_0^1 e^{-i s \Omega(t)} \frac{d\Omega}{dt} e^{i s \Omega(t)} ds \right\} e^{-i\Omega(t)}. \]  
(6)

Introducing the adjoint operator defined in terms of the commutator as \( a\Omega Y = [\Omega, Y] \) (also known as the Liouville operator), we obtain
\[ e^{-i s \Omega(t)} \frac{d\Omega}{dt} e^{i s \Omega(t)} = \exp \{-i s a\Omega(t)\} \frac{d\Omega}{dt}. \]  
(7)

The integration in Eq. (6) can then be formally written as
\[ \int_0^1 \exp \{-i s a\Omega(t)\} ds = \frac{e^{-i s a\Omega(t)} - 1}{-i s a\Omega(t)} = \phi(-i s a\Omega(t)), \]  
(8)

where
\[ \phi(x) = \frac{e^x - 1}{x}. \]  
(9)

Inserting Eqs. (6)–(9) in Eq. (1), we arrive at
\[ \phi(-i a\Omega) \frac{d\Omega}{dt} = H(t). \]  
(10)

Or, equivalently,
\[ \frac{d\Omega}{dt} = \phi^{-1}(-i a\Omega) H(t). \]  
(11)

Equation (11) is at the basis of the ME which is simply obtained by introducing the expansion of \( \phi^{-1}(x) \)
\[ \phi^{-1}(x) = \sum_k \frac{B_k x^k}{k!}, \]  
(12)

where \( B_k \) are the Bernoulli numbers \( (B_0 = 1, B_1 = -1/2, B_2 = 1/6, B_3 = 0, B_4 = -1/30... ) \) and the perturbative expansion \( \Omega(t) = \sum_{n=1}^{\infty} \Omega_n(t) \). Then Eq. (11) reads
\[ \frac{d\Omega}{dt} = \sum_k \frac{B_k}{k!} (-i)^k a\Omega_k \{ H(t) \}. \]  
(13)

Equation (13) justifies the name of exponential perturbation theory also used for the ME in some contexts. The ME is used in NMR spectroscopy with the AHT which is built up on the basis of the conventional ME to find \( F \) in Eq. (2) by computing \( \Omega(t = T) = \exp \{-i FT\} = \exp \{-i \Omega(T)\} \). Given the relevance of the ME, criteria for the existence and the convergence have been extensively developed in the literature since Magnus proposal in 1954.\(^{58}\) In fact, Magnus was well aware that if the function \( \Omega(t) \) is assumed to be differentiable, it may not exist everywhere.

C. The Floquet-Magnus expansion

Using Eqs. (1) and (2), we obtain the following inhomogeneous first-order differential equation for \( P(t) : \)
\[ i \frac{dP}{dt} = H(t) P(t) - P(t) F, \]  
(14)

where \( P(0) = I \) can be assumed, but this is non-mandatory as will be discussed later. \( F \) is an unknown constant matrix which is determined from the time periodicity condition \( P(t + T) = P(t) \). Using the exponential ansatz \( P(t) = e^{-iT(t)} \) with \( \Lambda (t + T) = \Lambda (t) \) and the same procedure for ME derivation (see Appendix A), one can obtain the following equation:
\[
\frac{d\Lambda}{dt} = \sum_{k=0}^{\infty} \frac{B_k}{k!} (-i)^k (ad_\Lambda)^k \{ H(t) + (-1)^{k+1} F \}. \tag{15}
\]

Note the similarity between Eqs. (15) and (13), but with the additional constant term \((-1)^k F\). It is worth noticing that Eq. (15) is independent of \(\Lambda(0)\) as well as Eq. (14) (derivation is given in Appendix A). As will be shown below, the advantage of the FME approach is its ability to make a choice for \(\Lambda(0)\) different from the generally assumed \(\Lambda(0) = 0\) (i.e., \(P(0) = I\)), allowing a simplification of the perturbative calculation of \(\Lambda(t)\) and \(F\). A choice of \(\Lambda(0) \neq 0\) is equivalent to the use of a more general representation of the evolution operator (and therefore less restrictive) for Eq. (2) as

\[
U(t) = P(t)e^{-itF} P^+(0), \tag{16}
\]

which removes the constraint of a stroboscopic observation. \(P(t)\) can then be seen as the operator that introduces the frame such that the density operator is varying under the time-independent Hamiltonian \(F\). Indeed, if \(\rho(t)\) denotes the density operator of the system, we have

\[
\rho(t) = P(t)e^{-itF} P^+(0)\rho(0)P(0)e^{itF} P^+(t), \tag{17}
\]

which can be rewritten as

\[
P^+(t)\rho(t)P(t) = e^{-itF} P^+(0)\rho(0)P(0)e^{itF} \tag{18}
\]
or, introducing \(\rho_s(t) = P^+(t)\rho(t)P(t)\),

\[
\rho_s(t) = e^{-itF}\rho_s(0)e^{itF}. \tag{19}
\]

Stroboscopic observation at times \(t_0 + nT\) is governed by the effective Hamiltonian (and thus the AHT) \(P(t_0)FP^+(t_0)\) according to

\[
\rho(t_0 + nT) = P(t_0 + nT) e^{-inTF} P^+(t_0) \rho(t_0) P(t_0) e^{inTF} P^+(t_0 + nT). \tag{20}
\]

Using \(P(t_0 + nT) = P(t_0)\) this can be rewritten as

\[
\rho(t_0 + nT) = e^{-inTP(t_0)FP^+(t_0)} \rho(t_0)e^{inTP(t_0)FP^+(t_0)}. \tag{21}
\]

Thus, the representation Eq. (16) of the evolution operator will allow us to work with a more general framework dealing both with the stroboscopic observation (AHT, \(\Lambda(0) = 0\)) and the effective Hamiltonian (as described by the Floquet theory, \(\Lambda(0) \neq 0\)), from an unified point of view. Knowledge of both \(P(t)\) and \(F\) yields the stroboscopic Hamiltonian (AHT) \(H_{AHT} = P(0)FP^+(0)\) or can be obtained directly using \(P(0) = I\). As Eq. (14) is independent of \(\Lambda(0)\), Eq. (15) can serve as the basis to construct the exponential perturbation expansion for both \(\Lambda(t)\) and \(F\).

The relationship with the regular Magnus expansion can be obtained from

\[
U(T, 0) = \exp(-i\Omega(T)) = \exp\{-iT e^{-i\Lambda(0)}Fe^{i\Lambda(0)}\}, \tag{22}
\]
such that

\[
\frac{\Omega(T)}{T} = e^{-i\Lambda(0)}Fe^{i\Lambda(0)}. \tag{23}
\]

Again, Eq. (23) points out that it is only in the case \(\Lambda(0) = 0\) that the FME gives the AHT as provided by the ME. However the ME is limited to the construction of the AHT, whereas the FME also constructs the operator \(\Lambda(t)\) giving the new opportunity to obtain the evolution of the system in between the stroboscopic detection points.

At this stage, we follow the procedure described in Ref. 59 introducing the perturbation expansions \(\Lambda(t) = \sum_{n=1}^{\infty} \Lambda_n(t)\) and \(F = \sum_{n=1}^{\infty} F_n(t)\) in Eq. (15). Successive order \(F_k\) terms are determined from the boundary conditions \(\Lambda_k(t + T) = \Lambda_k(t)\). A recursive generation scheme\(^{59}\) can then be built as follows \((n \geq 1)\):

\[
\Lambda_n(t) = \sum_{j=0}^{n-1} \frac{(-i)^j B_j}{j!} \left[ W_n^{(j)}(t) + (-1)^{j+1} T_n^{(j)}(t) \right] \tag{24}
\]

The terms \(W_n^{(j)}(t)\) and \(T_n^{(j)}(t)\) are given by the same recurrence:

\[
W_n^{(j)}(t) = \sum_{m=1}^{n-j} [\Lambda_m(t), W_{n-m}^{(j-1)}(t)] (1 \leq j \leq n - 1) \tag{25}
\]

and

\[
T_n^{(j)}(t) = \sum_{m=1}^{n-j} [\Lambda_m(t), T_{n-m}^{(j-1)}(t)] (1 \leq j \leq n - 1), \tag{26}
\]

but with the initial conditions

\[
W_1^{(0)}(t) = H(t), \quad W_{n+1}^{(0)}(t) = 0, \tag{27}
\]

\[
T_n^{(0)}(t) = T_n^{(0)}, \tag{28}
\]

Taking into account Eqs. (27) and (28) for substituting the terms \(j = 0\) in Eq. (24), we can obtain the general formula

\[
\Lambda_n(t) = \Lambda_n(0) + \int_0^T G_n(t)d\tau - iT_n, \tag{29}
\]

with

\[
F_n = \frac{1}{T} \int_0^T G_n(t)d\tau. \tag{30}
\]

The first order contributions to the Floquet-Magnus expansion give (see Appendix B) explicitly

\[
G_1(t) = H(t), \tag{31}
\]

\[
G_2(t) = -\frac{i}{2}[H(t) + F_1, \Lambda_1(t)], \tag{32}
\]
\[ G_3(\tau) = \frac{i}{2} [H(\tau) + F_1, \Lambda_2(\tau)] - \frac{i}{2} [F_2, \Lambda_1(\tau)] - \frac{1}{12} [\Lambda_1(\tau), [\Lambda_1(\tau), H(\tau) - F_1]]. \] \tag{33}

If higher order terms can be computed numerically quite easily with the help of Eqs. (24)–(28), we believe that symbolic calculations software can enable formal derivation of higher order terms.

### III. DISCUSSION OF CONVERGENCE

#### A. The Magnus expansion

In general, the Magnus series does not converge unless \( H(t) \) is small in a suitable sense.\(^7\) Indeed, the convergence of the Magnus expansion is generally discussed in terms of a radius of convergence \( r_c \) defined as the number for which the following statement holds:

\[ \int_0^1 \|H(s)\|ds < r_c \quad \text{then the Magnus expansion converges.} \]

Several results on the radius of convergence \( r_c \) in terms of \( H(t) \) have been obtained in the literature. Pechukas and Light\(^6\) and Karasev and Mosolova\(^2\) obtained \( r_c = \log 2 \approx 0.693... \), whereas Chacon and Fomenko\(^6\) got \( r_c = 0.577 \). Blanes et al.\(^6\) obtained the improved bound of \( r_c = 1.086 \). Recently, a new method was developed\(^6\) to enlarge the largest domain of convergence of the Magnus expansion \( r_c \approx 1.086... \) previously obtained. An analytic estimate of the new domain of convergence found was almost twice the preceding one\(^6\) (\( r_c = 2 \)) and this new analytic bound was in agreement with the numerical estimate of the convergence radius such as no accuracy was lost in the bound. Therefore, there are more than three different convergence estimated in the literature of Magnus expansion. These convergence estimates are given with their respective proofs in the references therein.

The latest improved bound \( r_c = \pi \) was derived by Moan\(^6\) but in the context of the conventional Magnus expansion for real matrices \( A(t) = -iH(t) \). This important results was then generalized to matrices in the Hilbert space (thus for complex matrices) by Casas.\(^6\)

A new version of Magnus expansion was reported recently by Butcher et al.\(^8\) The new scheme grows on trees and forests to reorder the terms of Magnus expansion for more efficient computation. While this scheme did not provide any substantial new result to the convergence of the ME, it provides a new mean to compute Magnus expansion to the desired order.

#### B. The Floquet theory

Recoupling schemes have all been extensively treated with Floquet theory in conjunction with the Van Vleck Transformation.\(^7\) The Floquet theory approach has also been used successfully to the study of decoupling of dipolar interactions.\(^7\) The discussion of the convergence of the Floquet theory was presented by Maricq.\(^8\) In this article, an indirect method that took advantage of the periodicity of \( P(t) \) and the method of Picard approximations was used to produce a convergent sequence for the propagator \( U(t) \). This was necessary to determine the convergence of the two interdependent series \( P(t) = \sum_n P_n(t) \) and \( F = \sum_n F_n \).

#### C. The Floquet-Magnus expansion

The Floquet-Magnus scheme is the fusion of the Magnus expansion and the Floquet theory. Therefore, its solution has the required structure and evolves in the desired group (Lie group). Because the Floquet scheme is a convergent sequence, once the convergence is fulfilled in one period, it is assured for any value of time. On the contrary, in the general Magnus case, the bound always gives a running condition. The latest improved bound in the conventional Magnus expansion was found to be \( r_c(ME) = \pi \). Blanes et al.\(^6\) and Casas et al.\(^8\) showed that absolute convergence of the Floquet-Magnus series is guaranteed at least if

\[ \int_0^1 \|H(t)\|dt < r_c(FME) \equiv 0.20925. \] \tag{34}

This bound \( r_c(FME) \) in the periodic Floquet case turns out to be smaller than the corresponding bound \( r_c(ME) = \pi \) in the conventional Magnus expansion. At first glance, this could be understood as a failure of the result. Certainly, the method has been given an algorithmic formulation which already allows direct implementation.\(^9\), \(^6\), \(^6\)

### IV. COMPARISON BETWEEN FLOQUET-MAGNUS AND OTHER THEORIES

We consider the most encountered case of a time-dependent Hamiltonian \( H(t) \) which is time periodic with period \( T \). It can then be expanded in a Fourier series as

\[ H(t) = \sum_m H_m \exp(i m \omega t) = \sum_{m \neq 0} H_m \exp(i m \omega t) + H_0. \] \tag{35}

With \( \omega = 2\pi / T \). Substituting Eq. (35) into the first contributions to the Floquet-Magnus expansion Eq. (30) to Eq. (33) gives

\[ \Lambda_1(t) = \sum_{m \neq 0} \frac{H_m}{i m \omega} f_m(t) + \Lambda_1(0), \] \tag{36}

with

\[ f_m(t) = \exp(i m \omega t) - 1, \] \tag{37}

\[ F_1 = H_0, \] \tag{38}

\[ \Lambda_2(t) = -\frac{1}{2} \sum_{m \neq 0} \frac{[H_m, \Lambda_1(0)]}{m \omega} f_m(t) + i \sum_{m \neq 0} \frac{[H_0, H_m]}{m^2 \omega^2} f_m(t) + \frac{i}{2} \sum_{m \neq n, n \neq 0} (1 - \delta_{m+n}) \frac{[H_m, H_n]}{m(m + n) \omega^2} f_{m+n}(t) - \frac{i}{2} \sum_{m \neq 0, n \neq 0} \frac{[H_m, H_n]}{m n \omega^2} f_m(t) + \Lambda_2(0), \] \tag{39}
and

\[ F_2 = -i \{ H_0, \Lambda_1(0) \} + \frac{1}{2} \sum_{m \neq 0} \frac{[H_m, H_{-m}]}{m \omega} + \sum_{m \neq 0} \frac{[H_0, H_m]}{m \omega}. \]  

(40)

At this stage, the features of the FME scheme depend mainly on the choice of the initial conditions. Let us consider the following two cases corresponding to \( \Lambda(0) = 0 \) and \( \Lambda(0) \neq 0 \), respectively.

### A. Case \( \Lambda(0) = 0 \)

In this case, we obtain the following explicit results:

\[ F_1 = H_0, \]

(41)

\[ \Lambda_1(t) = \sum_{m \neq 0} \frac{H_m}{im \omega} f_m(t), \]

(42)

\[ F_2 = + \frac{1}{2} \sum_{m \neq 0} \frac{1}{m \omega} [H_m, H_{-m}] + \sum_{m \neq 0} \frac{1}{m \omega} [H_0, H_m], \]

(43)

and

\[ \Lambda_2(t) = i \sum_{m \neq 0} \sum_{n \neq 0} (1 - \delta_{n+m}) \frac{[H_m, H_n]}{m(m+n) \omega^2} f_{m+n}(t) - \frac{1}{2} \sum_{m \neq 0} \sum_{n \neq 0} \frac{[H_m, H_n]}{mn \omega^2} f_m(t) + i \sum_{m \neq 0} \frac{[H_0, H_m]}{m \omega^2} f_m(t). \]

(44)

The choice of \( \Lambda(0) = 0 \) reproduces the two first order terms of AHT, \( F_1 \) and \( F_2 \), and therefore corresponds to the stroboscopic detection scheme. However, the FME also provides the operators \( \Lambda_1(t) \) and \( \Lambda_2(t) \) allowing for the evaluation of the evolution in between the stroboscopic points. Third order terms were too cumbersome to be calculated and reproduced here.

### B. Case \( \Lambda(0) \neq 0 \)

As in the previous case, we first obtain

\[ F_1 = H_0. \]

(45)

As \( \Lambda_{n+1}(t) \) depends upon \( \Lambda_n(t) \), we seek for a suitable choice so that any increase in the number of terms should be avoided. Making the following choice of \( \Lambda_1(0) \)

\[ \Lambda_1(0) = \sum_{m \neq 0} \frac{H_m}{im \omega}, \]

(46)

seems natural from the examination of Eq. (36). It gives a time-dependent evolution operator as

\[ \Lambda_1(t) = \sum_{m \neq 0} \frac{H_m}{im \omega} e^{im \omega t}. \]

(47)

Accordingly, at each further step of the calculation, the choice of the contributions to \( \Lambda_n(0) \) is made according to the simple rule

\[ \int_0^t e^{i \omega u} du = \frac{e^{i \omega u}}{i \omega} - \frac{1}{i \omega} \frac{e^{i \omega u}}{\Lambda_n(t) - \Lambda_n(0)}. \]

(48)

Then the second order contribution \( G_2(t) \) (Eq. (32)) reads

\[ G_2(t) = -\sum_{m \neq 0} \frac{[H_m, H_n]}{m \omega} e^{i(n+m) \omega t} \]

(49)

Time integration yields much simpler expressions than Eqs. (43) and (44):

\[ \Lambda_2(t) = i \sum_{m \neq 0} \sum_{n \neq 0} (1 - \delta_{n+m}) \frac{[H_m, H_n]}{m(n+m) \omega^2} e^{i(n+m) \omega t} + i \sum_{m \neq 0} \frac{[H_0, H_m]}{m \omega^2} e^{im \omega t}, \]

(50)

\[ F_2 = \frac{1}{2} \sum_{m \neq 0} \frac{[H_m, H_{-m}]}{m \omega}. \]

(51)

It clearly shows that the application of Eq. (48) has greatly simplified the expressions of \( \Lambda_2(t) \) and \( F_2 \). In this case, this allows us to pursue with the same approach for \( \Lambda_3(t) \) (see Appendix B) yielding

\[ \Lambda_3(t) = \frac{1}{4} \sum_{m, n, k \neq 0} (1 - \delta_{m+n})(1 - \delta_{m+n+k}) \]

\[ \times \frac{[H_k, [H_m, H_n]]}{in(n+m)(k+m+n)\omega^3} e^{i(k+n+m)\omega t} \]

\[ + \frac{1}{4} \sum_{m \neq 0} (1 - \delta_{m+n}) \frac{[H_0, [H_m, H_n]]}{in(n+m)^2 \omega^5} e^{i(n+m)\omega t} \]

\[ + \frac{1}{2} \sum_{m \neq 0} (1 - \delta_{m+n}) \frac{[H_k, [H_0, H_m]]}{im^2(m+k)\omega^5} e^{i(k+m)\omega t} \]

\[ + \frac{1}{4} \sum_{m \neq 0} \frac{[H_m, [H_m, H_m]]}{im^3 \omega^3} e^{im \omega t} + \frac{1}{2} \sum_{m \neq 0} \frac{[H_0, [H_0, H_m]]}{im^2 \omega^3} e^{im \omega t} \]

\[ \times (1 - \delta_{m+n+k}) \frac{[H_m, [H_n, H_k]]}{inm(k+m+n)\omega^3} e^{i(k+n+m)\omega t}, \]

(52)
and $F_3$ as

$$F_3 = \frac{1}{3} \sum_{m,n\neq 0} \frac{[H_{m}, [H_{m-n}, H_{n}]]}{nm\omega^2} + \frac{1}{2} \sum_{m,n\neq 0} \frac{[H_{m}, [H_{0}, H_{m-n}]]}{m^2\omega^2}.$$  (53)

Equation (53) nicely reproduces the result of Ernst et al.\(^{71,77}\)

C. Similarities between theories (AHT, FT, and FME)

A quick comparison between all theories developed so far shows that the lowest-order term $F_1$ as provided by AHT,\(^{20}\) Floquet theory,\(^{35,69,71,85,86}\) or FME are all identical. This is the popular average Hamiltonian

$$H_{AHT}^{(0)} = H_{eff}^{(1)(FT)} = F_{1(FME)} = H_0.$$  (54)

Next, the first-order term in the AHT (Ref. 69) is identical to the first-order term in the stroboscopic detection.\(^{86}\) These two Hamiltonians are also identical to Eq. (43) of the FME with the stroboscopic detection condition $\Lambda(0) = 0$:

$$H_{AHT}^{(1)} = H_{FME}^{(1)(stroboscopic)} = F_{2(FME,\Lambda(0)=0)}.$$  (55)

Similarly, Maricq\(^{35}\) has shown the equivalence between the full, non-transformed, Floquet Hamiltonian, and the AHT Hamiltonian by calculating a propagator at multiples of the period of the Hamiltonian. In the article, it used a perturbative scheme to show that the two expansions (AHT, FT) are equal for each of the first two orders. For the Floquet Hamiltonian, the following second order term was found by Maricq

$$H_{FME}^{(2)}(Maricq) = \sum_{k=1}^{\infty} \frac{1}{k!\omega} [[H_k, H_{-k}] + [H_0, H_k] - [H_0, H_{-k}]]$$  (56)

which can be shown, after straightforward algebra, to be identical to Eq. (43). Higher order terms of the Floquet Hamiltonian by Maricq become very tricky to manipulate rather quickly.

The highlighted similarities of FME scheme with the Floquet theory stem here from a judicious choice of the initial conditions. As demonstrated in Subsection IV B, with $\Lambda(0) = \sum_{m\neq 0} \frac{H_m}{m\omega}$, the FME approach retrieves identical results at least for the first three orders $F_{1,2,3}$ than the FT and Van Vleck transformation.\(^{21,69,87}\) The FME approach is however unique though it provided expressions for $\Lambda_{1,2,3}(t)$. In FT,\(^{35}\) the zeroth-order term for the $P_0(t)$ is $P_0 = 1$. Next, the first order term $P_1(t)$ in the FT is similar to $\Lambda_1(t)$, Eq. (42) in the FME for the stroboscopic case ($\Lambda_1(0) = 0$). However, the direct comparison of higher order terms including $\Lambda_2(t)$ (Eq. (44)) and $P_2(t)$ becomes impossible, since $P_2(t)$ is not available easily. Obviously, the expressions of $\Lambda_{2,3}(t)$ are obtained more easily in the FME than $P_2(t)$ from the FT. Therefore the FME provides a more simplistic approach for higher orders comparatively to other averaging schemes.

D. Differences between theories (AHT, FT, FME)

Although Floquet theory, also known as secular averaging theory, and AHT are different approaches of the same Schrodinger equation, the results they provide are somewhat incompatible in the analysis of various multiple-pulse sequences in NMR. In discussing the equivalence between FT and AHT, Llo\(^{87}\) found a supplementary non-secular term in the first-order of AHT which is identical to $F_{2(FME,\Lambda(0)=0)}$, i.e., $H_{AHT}^{(1)} = H_{FME}^{(1)(stroboscopic)} = F_{2(FME,\Lambda(0)=0)}$. The answer to that is to invalidate the use of AHT in the interpretation of many NMR experiments, such as sample rotation and pulse crafting. An important point arises in the differences between AHT and the FT schemes: these approaches are respectively given in terms of time integrals and Fourier coefficient combinations, so, the type of time modulation of the Hamiltonian determines the choice between the methods to be adopted to solve a specific problem. FME gives a unified view which encompasses both the time-integral and Fourier expansion approach. One of the appealing features of the FME scheme is its expressions for $\Lambda_{1,2,3}(t)$ which are not present in other averaging approaches, and additional terms in the scheme could arise from the initial conditions ($\Lambda_1(0), \Lambda_2(0), \Lambda_3(0), \ldots$).

E. Static perturbation theory versus FME

Here, we wish to revisit the static perturbation theory\(^{21}\) which has been shown to yield the correct form of Zeeman truncated NMR interactions without the limit of stroboscopic observation of the AHT. This will give us the opportunity to shed a new light on the FME scheme and the derivation of a criterion for the two theories being compatible.

For sake of simplicity (but without loss of generality), we consider the Hamiltonian

$$H = \omega_0 I_Z + \lambda \sum_{m} (-1)^m R_{2-m} T_{2,2+m}.$$  (57)

This is a common form of Hamiltonian in solid-state NMR. $\omega_0 I_Z$ is the Zeeman interaction, $R_{2,m}$ are the lattice parts of the internal interaction which encode its orientational dependence with respect to the magnetic field, $T_{2,m}$ are second rank m-order spherical tensor describing the spin system as defined by $[I_Z, T_{2,m}] = mT_{2,m}$. The SPT in terms of the irreducible tensor operators gives the diagonal Hamiltonian (with respect to $\omega_0 I_Z$)\(^{21}\)

$$H_{SPT} = \omega_0 I_Z + \lambda R_{2,0} T_{2,0} + \frac{\lambda^2}{2\omega_0} \sum_{m \neq 0} \frac{R_{2,m} R_{2,-m}}{m} \times [T_{2,m}, T_{2,-m}].$$  (58)

As discussed in the seminal work,\(^{21}\) discrepancies between AHT and FT appear in the rotating frame representation (or more generally in the interaction frame) where the
Hamiltonian becomes time-dependent:
\[ H(t) = e^{i\omega_0 t} H e^{-i\omega_0 t} = \lambda \sum_m (-1)^m R_{2,-m} T_{2,+m} e^{im\omega_0 t}. \]  

The FME expansion Eqs. (45)–(47) yields as first order terms:
\[ F_1 = \lambda R_{2,0} T_{2,0}, \]  
\[ \Lambda_1(t) = \lambda \sum_{m \neq 0} (-1)^m \frac{R_{2,-m} T_{2,+m}}{im\omega_0} e^{im\omega_0 t}, \]

whereas the AHT (stroboscopic detection, Eqs. (41)–(43)) yields
\[ \Lambda_1(t) = \lambda \sum_{m \neq 0} (-1)^m \frac{R_{2,-m} T_{2,+m}}{im\omega_0} (e^{im\omega_0 t} - 1). \]  

The FME scheme Eq. (51) provides the same second order term as in SPT theory
\[ F_2 = \frac{\lambda^2}{2\omega_0} \sum_{m \neq 0} R_{2,m} R_{2,-m} [T_{2,m}, T_{2,-m}]. \]  

This shows that FME provides an expansion in the rotating frame which is in agreement with the static perturbation theory and Van Vleck transformations. This is not the case for the Magnus expansion. This agreement, as obtained from Eq. (46), can be easily explained by the connection that exists between the FME and SPT propagators as follows.

The propagator in SPT can be written as
\[ U(t) = e^{-iHt} = e^{-iS} e^{-iH_{\text{FME}} t} e^{iS}, \]  

where \( S \) is the diagonalizing matrix defined as \( H = e^{-iS} H_{\text{FME}} e^{iS} \). When transformed back into the laboratory frame, the FME yields the propagator
\[ U(t) = e^{-i\omega_0 I_Z t} \times e^{-i\Lambda(t)} e^{-iF t} e^{i\Lambda(0)}, \]  

which can be rewritten as
\[ U(t) = \exp(-i e^{-i\omega_0 I_Z t} \Lambda(t) e^{i\omega_0 I_Z t}) e^{-i\omega_0 I_Z t} e^{-iF t} e^{i\Lambda(0)}. \]  

Comparison between Eqs. (66) and (64) shows that the two conditions
\[ \Lambda(0) = e^{-i\omega_0 I_Z t} \Lambda(t) e^{i\omega_0 I_Z t}, \]  
\[ [I_Z, F] = 0, \]
yield the following final form of the propagator:
\[ U(t) = e^{-i\Lambda(0)} e^{-i(\omega_0 I_Z + F) t} e^{i\Lambda(0)}, \]  
which is equivalent to Eq. (64). This means that the condition
\[ S = \Lambda(0) = e^{-i\omega_0 I_Z t} \Lambda(t) e^{i\omega_0 I_Z t}, \]  
has to be fulfilled for propagators Eqs. (64) and (65) describing the same evolution at any time. The condition Eq. (67) is indeed satisfied by Eqs. (46), (47), (50), and (52), justifying a posteriori our choice for \( \Lambda(0) \). In the case \( \Lambda(0) = 0 \), Eq. (67) is only true at stroboscopic point \( \omega_0 \tau = 1 \) for Eqs. (42) and (44).

F. Extension to multimode Hamiltonian

Application of FME to multimode Hamiltonian with frequencies \( \omega = (\omega_1, \ldots, \omega_N) \) is straightforward. Considering the generalized Fourier expansion of the Hamiltonian \( (m = (m_1, \ldots, m_N) \) represents the frequency indices)
\[ H(t) = \sum_m H_m \exp(-im \cdot \omega t), \]  
we obtain
\[ \Lambda_1(t) = \sum_m \frac{H_m}{im \cdot \omega} e^{-im \cdot \omega t} \]  
and
\[ F_1 = \sum_{m \cdot \omega = 0} H_m. \]

Similarly, calculation of second order terms is straightforward using Eqs. (50) and (51). These expressions highlight the fact that the multimode Hamiltonian case can be easily treated in Hilbert space with the FME.

V. CONCLUSION

In this work, we have presented and generalized the Floquet-Magnus expansion that has been useful to shed new lights on AHT, Floquet Theory, and the static perturbation theory. The theory is based on two operators: \( \Lambda(t) \) that describes the evolution within the period and \( F \) which is the Hamiltonian governing the evolution at multiple of the period. A crucial parameter has been shown to be the periodic boundary condition \( \Lambda(0) \). The FME theory can be directly connected to the AHT method (based on Magnus expansion) for \( \Lambda(0) = 0 \) yielding \( F = H_{\text{AHT}} \). But, in contrast to the Magnus expansion, the knowledge of the operator \( \Lambda(t) \) allows the evolution in-between the stroboscopic points to be evaluated. Equivalence with the Floquet theory is obtained from a special choice of \( \Lambda(0) \neq 0 \) leading also to an expansion that is equivalent with the static perturbation theory. The general conditions for such an equivalence have been derived, and the FME provides a more concise and intuitive approach than Floquet theory.

The FME provides a quick means to calculate higher order term (here third order terms could be easily derived) allowing the disentanglement of the stroboscopic observation \( \Lambda(t) \) and effective Hamiltonian \( F \) that will be useful to describe spin dynamics in solid-state NMR and understand different synchronized or non-synchronized experiments. We made an attempt to sketch out the mean features of the technique, and hope that this paper will be helpful to describe the time evolution of the spin system at all times. The FME offers
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APPENDIX A: DERIVATION OF THE FME EXPANSION

Using the following representation of the propagator:

\[ U(t) = P(t) e^{-iFt} P^+(0), \]

in Eq. (1) we obtain

\[ i \frac{dU}{dt} = i \frac{dP}{dt} e^{-iFt} P^+(0) + P(t) F e^{-iFt} P^+(0) \]

\[ = H(t) P(t) e^{-iFt} P^+(0). \]

Multiplying all terms on the right hand side by \( P(0) \) leads to

\[ i \frac{dP}{dt} = H(t) P(t) - P(t) F. \]

Applying the Wilcox formula Eq. (6) to \( P(t) = e^{-i\Lambda(t)} \) gives

\[ i \frac{d}{dt} [e^{-i\Lambda(t)}] = \phi(-iad\Lambda(t)) \frac{d\Lambda}{dt} e^{-i\Lambda(t)}, \]

which inserted in Eq. (A3) yields

\[ \phi(-iad\Lambda(t)) \frac{d\Lambda}{dt} e^{-i\Lambda(t)} = H(t) e^{-i\Lambda(t)} - e^{-i\Lambda(t)} F, \]

which can be transformed into

\[ \frac{d\Lambda}{dt} = \phi^{-1} (-iad\Lambda)(H(t) - e^{-i\Lambda(t)} F). \]

Using \( \phi^{-1}(x)e^x = x + \phi^{-1}(x) \), we finally arrive at

\[ \frac{d\Lambda}{dt} = \sum_{k=0}^{\infty} \frac{B_k}{k!} (-iad\Lambda)^k (H(t) + (-1)^{k+1} F). \]

APPENDIX B: THE FME EXPANSION TO THIRD ORDER

From the recursive generation formula Eqs. (24)–(28), we have the following results for the second order terms:

\[ W_2^{(0)} = 0, \quad T_2^{(0)} = F_2, \]

\[ W_2^{(1)} = [\Lambda_1, H], \quad T_2^{(1)} = [\Lambda_1, F_1]. \]

The third order terms read as follows:

\[ \dot{\Lambda}_3(t) = \sum_{j=0}^{2} (-i) \frac{B_j}{j!} \]

\[ \times \left\{ W_3^{(j)} + (-1)^{j+1} T_3^{(j)} \right\} \frac{W_3^{(0)}}{dP_3^{(0)}} = F_3, \]

\[ + \frac{i}{2} \left\{ W_3^{(1)} + T_3^{(1)} \right\} - \frac{1}{12} \left\{ W_3^{(2)} - T_3^{(2)} \right\}, \]

\[ W_3^{(1)} = \sum_{m=1}^{2} [\Lambda_m, W_3^{(0)}] = \left[ \Lambda_1, W_3^{(0)} = 0 \right] + \left[ \Lambda_2, W_3^{(0)} = H_3 \right], \]

\[ = [\Lambda_2, H], \]

\[ T_3^{(1)} = \sum_{m=1}^{2} [\Lambda_m, T_3^{(0)}] = \left[ \Lambda_1, T_3^{(0)} = 0 \right] + \left[ \Lambda_2, T_3^{(0)} = F_1 \right], \]

\[ = [\Lambda_1, F_2] + [\Lambda_2, F_1], \]

\[ W_3^{(2)} = \sum_{m=1}^{2} [\Lambda_m, W_3^{(1)}] = [\Lambda_1, W_3^{(1)}] \]

\[ = [\Lambda_1, [\Lambda_1, H]], \]

\[ T_3^{(2)} = \sum_{m=1}^{2} [\Lambda_m, T_3^{(1)}] = [\Lambda_1, T_3^{(1)}] = [\Lambda_1, [\Lambda_1, F_1]], \]

\[ \dot{\Lambda}_3(t) = -F_3 + \frac{i}{2} ([\Lambda_2, H] + [\Lambda_1, F_2] + [\Lambda_2, F_1]) \]

\[ - \frac{1}{12} \left\{ [\Lambda_1, [\Lambda_1, H]] - [\Lambda_1, [\Lambda_1, F_1]] \right\}. \]

After time integration of Eq. (B7), we obtain the final expression

\[ \Lambda_3(t) = -tF_3 + \frac{i}{2} \int_0^t ([\Lambda_2, H + F_1] + [\Lambda_1, F_2]) d\tau \]

\[ - \frac{1}{12} \int_0^t ([\Lambda_1, [\Lambda_1, H - F_1]]) d\tau + \Lambda_3(0). \]
\[ F_3 = \frac{i}{2T} \int_0^T \left( \left\{ \left[ A_2, H + F_1 \right] + \left[ A_1, F_2 \right] \right\} \right) d\tau \]

\[ - \frac{1}{12T} \int_0^T \left( \left[ \left[ A_1, H - F_1 \right] \right] \right) d\tau. \]  
\hspace{2cm} (B9)

**APPENDIX C: CALCULATION OF \( \Lambda_{\mu 1,2,3}(t) \) AND \( F_{1,2,3} \) FOR A PERIODIC TIME-DEPENDENT HAMILTONIAN**

Our starting point is the Fourier expansion of the Hamiltonian as

\[ H(t) = \sum_m H_m \exp(im\omega t) = H_0 + \sum_{m \neq 0} H_m \exp(im\omega t) \]

\[ = H_0 + H, \]  
\hspace{2cm} (C1)

with \( \omega = \frac{2\pi}{T} \). Derivation of first and second order terms is straightforward and not reproduced here. \( G_3(t) \) (Eq. (33)) can be decomposed in three terms

\[ G_3(t) = -\frac{i}{2} \left[ H(t) + F_1, A_2(t) \right] - \frac{i}{2} \left[ F_2, A_1(t) \right] 
- \frac{1}{12} \left[ A_1(t), A_1(t), H(t) - F_1 \right] = A + B + C. \]  
\hspace{2cm} (C2)

After straightforward but lengthy calculations, we obtain

\[ A = \frac{1}{4} \sum_{m,n,k \neq 0} \left( \left[ H_m, [H_n, H_0] \right] \right) \frac{1 - \delta_{m+n}}{m(n+m)\omega^2} e^{i(m+n+k)\omega t} \]

\[ + \frac{1}{2} \sum_{m,k \neq 0} \left( \left[ H_m, H_0, H_k \right] \right) e^{i(n+k)\omega t} - i[H_0, A_2(t)], \]  
\hspace{2cm} (C3)

\[ B = -\frac{1}{4} \sum_{m,n \neq 0} \left( \left[ H_m, H_{-m}, H_n \right] \right) e^{im\omega t}, \]  
\hspace{2cm} (C4)

\[ C = \frac{1}{12} \sum_{m,n,k \neq 0} \left( \left[ H_m, [H_n, H_k] \right] \right) \frac{e^{i(m+n+k)\omega t}}{mn\omega^2}. \]  
\hspace{2cm} (C5)

Integration gives Eq. (52) for \( \Lambda_3(t) \). Only terms A and C contribute to \( F_3 \) as

\[ F_3 = \frac{1}{4} \sum_{m,n \neq 0} \left( 1 - \delta_{m+n} \right) \frac{\left( \left[ H_{-m+n}, [H_m, H_n] \right] \right)}{n(n+m)\omega^2} \]

\[ + \frac{1}{2} \sum_{m,n \neq 0} \frac{\left( \left[ H_{-m}, H_0, H_n \right] \right)}{m^2\omega^2} \]

\[ + \frac{1}{12} \sum_{m,n \neq 0} \left( \left[ H_m, [H_n, H_{-m+n}] \right] \right). \]  
\hspace{2cm} (C6)

Notably, summation indices can be reordered so that the first term contributes to the third term of the right-hand side of Eq. (C6). Final result is Eq. (53).


