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Application of Floquet-Magnus Expansion During the Phase Modulated Lee-Goldburg Radiation in Solid State NMR

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Abstract

This work uses the Floquet-Magnus expansion approach to investigate the spin system evolution during the phase modulate Lee-Goldburg radiation experiment. Until now, the Frequency switched Lee-Goldburg and its variant called the Phase module Lee-Goldburg have been treated by only the average Hamiltonian theory and the bimodal Floquet approach. In this article, we use the expansion schemes of the Floquet-Magnus expansion to calculate the effective Hamiltonian and propagator during the spin dynamics. We present a remarkable iterative approach for the Floquet-Magnus expansion. Our work unifies and generalizes existing results of the Floquet-Magnus expansion and delivers illustrations of novel springs that boost previous applications that are based on the classical information. The method presented could plays a major role in the interpretation of a number of fine NMR experiments in solids, which provide significant new insight in spin physics. The generality of the work points to potential applications in problems related in solid-state NMR and theoretical developments of spectroscopy as well as interdisciplinary research areas whenever they include spin dynamics concepts. The considered method of Floquet-Magnus expansion has recently found new major areas of applications such as in topological materials..

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Introduction

Over the past four decades, the theory in solid-state nuclear magnetic resonance (SSNMR) has emerged into a standard tool for spin dynamics calculations. In most instances, time-dependent linear differential equation towards obtaining propagators can be obtained with the average Hamiltonian theory (AHT) [1-22] and the Floquet theory (FLT) [23-26]. For more complex SSNMR experiments requiring more than four frequency modulations, the combination of AHT and FLT called Floquet-Magnus expansion (FME) [27-29] has proven to be particularly reliable. The control of spin systems using the FME approach can provide a more intuitive understanding of spin dynamics processes. In this manuscript, the main topics concern the application of FME to the Phase Modulated Lee-Goldburg (PMLG) radiation experiment in Solid State NMR [30]. Vinogradov and co-workers introduced the PMLG to achieve line narrowing not by frequency switching (FSLG), but by a series of pulses with well-defined phases [30]. Unlike the FSLG that uses only one step in the jumping of frequency together with a phase shift, the PMLG uses a large number of steps given by the number of pulses. The PMLG has the ability to adapt in many different situations such as handling the amplitudes and phases of the pulses in a way the FSLG experiment may not be capable of executing. This pulse sequence built-up several possibilities to improve the efficiency of line narrowing in related experiments such as hetero-correlation investigations [31,32]. In recent years, Vinogradov et al. presented a description of PMLG experiments using a bimodal Floquet theory treatment [33,34]. This bimodal is a version of Shirley's Floquet theory approach that yielded an appropriate theoretical framework for the description of the PMLG under magic-angle (MAS) influence [24]. The current work consists on evaluating the FME terms for the PMLG radiation experiment. The topic of FME is well known in mathematics and the scientific community and its investigation is important and useful in various pulse sequences such as PMLG. In spin dynamics, obtaining the propagators is of major interest in solving a time-dependent linear differential equation. The quantum propagator U (t) generally satisfies the time-dependent Schrodinger equation, which is difficult to solve except if the Hamiltonian H

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(t) is time independent (H) or it commutes with itself at two different times. The dynamics of a quantum system under a time-dependent Hamiltonian is an important general problem in various areas of chemical and physical chemistry [35]. In several situations such as the cases of the adiabatic and strong-collision limits [36,37], some approaches on the time dependence of the Hamiltonian allow approximate solutions to be obtained as perturbation expansions. Llor [38] studied the equivalence between dynamical averaging methods of the Schrodinger equation, with for example, the AHT and the FLT. But it was found that the results provided by the two methods are incompatible in the analysis of various multiple-pulse sequences in NMR [17-22,26]. Several authors such as Feldman and co-workers [39], Goldman and co-workers [40], Buishvilli and co-workers [41], and Maricq [26] reported remarkable contributions to unifying the AHT and FLT. One of the successful attempts was made by Llor [38] who derived a clear relationship between the two perturbative approaches (AHT and FLT) by reformulating the FLT in terms of a static block-diagonalization procedure known as van Vleck transformation. This restructuration proved that the effective Hamiltonians of both perturbative methods are equivalent. One important physical example in which the FME have been applied successfully is the periodically driven harmonic oscillator. Also, Klarsfeld and Oteo [42] applied the FME approach to a periodically driven harmonic oscillator to obtain a remarkable feature consisting in the infinite Floquet-Magnus expansion solved with the expression F determining the spinning sideband intensity and the function useful to evaluate the spin behavior during or in between the RF pulses.

In the following section II, we present a remarkable iterative approach for the Floquet-Magnus expansion based on the formulation described in the appendix. In Section III, we effectively calculated the first two terms of the FME applied to the Phase Modulated Lee-Goldburg radiation experiment. In section V, we presented the work in a prospect of spin physics.

Theoretical analysis

The Floquet-Magnus expansion expands its propagator in the form of a more general representation of the evolution operator as [28]

$$U(t) = P(t)e^{-itF}P^{+}(0) \qquad (1)$$

which eradicates the limitation of a strobe's inspection. The function *P(t)* expresses the operator related to the time independent Hamiltonian F and the variable density operator. The FME propagator looks quite different than the Fer expansion propagator, which is also an alternative developing approach to control the spin dynamics in solid-state NMR [29]. In this article, we probed the spin-locking radiation experiment under sample spinning using the FME approach. Setting an infinite sequence (*u1, u2, u3*,...), the n^{th} partial sum σ_n is the sum of the first *n* terms of the sequence,

$$\sigma_n = \sum_{l=1}^{n} u_l \qquad (2)$$

Mathematically speaking, a series converges if there exists a number *p* such that for any arbitrarily small positive number ȝ, there is a large integer *N* such that for all

$$n \geq N \qquad (3)$$

$$|\sigma_n - p| \leq \xi \qquad (4)$$

Convergence of the Magnus expansion has come into question in various applications. In general, the Magnus series does not converge unless the Hamiltonian is small in a proper sense. The divergence of the Magnus expansion guides to inconsistencies in the spin dynamics of solid-state NMR [40,46-49]. The FME can be reformulated in the sense of an iterative approach, which can be observed in the following picture. Let us start with the expansion of the propagator of the FME in a more general representation of the evolution operators expressed by the Eq. (1), where the time-independent Hamiltonian F is a series defined as

$$F = \sum_{n} F_n \qquad (5)$$

Using the exponential ansatz,

$$P(t) = e^{-i\Lambda(t)} \qquad (6)$$

where the function

$$\Lambda(t) = \sum_{n} \Lambda_n(t) \qquad (7)$$

Is the argument of the operator *P(t)*. The first term of the integrand, $G_1(t)$,

$$G_1(t) = \tilde{H}(t) \qquad (8)$$

allows to compute the first term of the time-independent Hamiltonian, F_1,

$$F_1 = \frac{1}{\tau_C}\int_0^{\tau_C} G_1(u)du \qquad (9a)$$

and the combination of the terms $G_1(t)$ and F_1 generates the first term of the width of the spinning sidebands in MAS, Λ_1 (t),

$$\Lambda_1(t) = \Lambda_1(0) + \int_0^t G_1(u)du - tF_1 \qquad (9b)$$

Next, the combination of Λ_1 (t) and F_1 allows to obtain the second term of the integrand, $G_2(t)$,

$$G_2(t) = -\frac{i}{2}[H(t) + F_1, \Lambda_1(t)] \qquad (10)$$

Subsequently, the second term of the integrand $G_2(t)$, allows to compute the second term of the time-independent Hamiltonian F_2, such as

$$F_2 = \frac{1}{\tau_C}\int_0^{\tau_C} G_2(u)du \qquad (11)$$

and so on. Higher order terms can be computed numerically quite easily and symbolic calculations software can enable formal derivation of higher order terms. The above iteration can be summarized in the following picture or diagram,

$$\left\{\begin{array}{c} G_1(Eq.(8)) \rightarrow F_1(Eq.(9-a)) \downarrow \rightarrow (G_1 + F_1)(Eq.(9-b)) \rightarrow \Lambda_1; \\ (\Lambda_1 + F_1)(Eq.(10)) \rightarrow G_2; \\ G_2(Eq.(10)) \rightarrow F_2(Eq.(11)) \downarrow \ldots \\ \ldots\ldots \\ \ldots\ldots \end{array}\right\} \qquad (12)$$

The initial condition $\Lambda(0)$ does actually influence the average Hamiltonian use in the interpretation of data. The general functions F_n and $G_n(t)$ are defined as [28]

$$F_n = \frac{1}{\tau_C}\int_0^{\tau_C} G_n(u)du \qquad (13)$$

$$\Lambda_n(t) = \Lambda_n(0) + \int_0^t G_n(u)du - tF_n \qquad (14)$$

Application of the FME to Phase Modulated Lee-Goldburg radiation

TThe Hamiltonian describing the Phase Modulated Lee-Goldburg (PMLG) radiation is giving by Vinogradov et al. [30,31],

$$H(t) = H_{CS}(t) + H_D(t) + H_{RF}(t) \quad (15)$$

where $H_{CS}(t)$, $H_{CS}(t)$, and $H_{CS}(t)$ are the chemical shift terms, dipole-dipole interaction terms, and the rf field term, respectively. As described by Bennet, et al., the dipolar interaction Hamiltonian has the form [43]

$$H_D(t) = \sum_{i<j}\sum_{n=-2}^{2} \omega_D^{ij} G_n^{ij} e^{in\omega_r t}\left[2I_{Zi}I_{Zj} - (I_i^+ I_j^- + I_i^- I_j^+)\right] \quad (16)$$

and the chemical shift (CS) interaction

$$H_{CS}(t) = \sum_i \left\{ \Delta\omega_i I_{Zi} + \sum_{n=-2}^{2} \omega_{CS}^i g_n^i e^{in\omega_r t} I_{Zi} \right\} \quad (17)$$

The dipolar interaction ω_D^{ij} and the complex geometric coefficients G_n^{ij}, g_n^i, are depending on the polar angles $(\theta^{ij},\varphi^{ij})$ of the distance vector r_{ij} between the protons I and j in the rotor frame. The CS Hamiltonian contains a sum of isotropic chemical shift terms and chemical shift anisotropy terms. Let us write the RF Hamiltonian in the form [31],

$$H_{RF}(t) = \omega_1(t)\left(I_X \cos\phi(t) + I_Y \sin\phi(t)\right) \quad (18)$$

where $\omega_1(t)$ is the RF amplitude and $\varphi(t)$ is a time dependent phase. For convenience, as suggested by Mehring and Waugh [44], the Hamiltonian describing the PMLG radiation can be assessed by its spin system transformation to the interaction frame described by the RF [30]. For simplicity reasons, the spin operators are represented by their irreducible tensor components and their transformation can be written as

$$U_{RF}^{-1}(t) T_{lo} U_{RF}(t) = \sum_{m=-l}^{l} \alpha_{lm}(t) T_{lm} \quad (19)$$

where $U_{RF}(t)$ is the operator transforming the spin system to the interaction frame. For the cyclic transformation (time constant,), the above equation can be developed with, $\omega_C = \frac{2\pi}{t_C}$ as

$$\alpha_{lm}(t) = \sum \alpha_{lmk} e^{\{ik\omega_C t\}} \quad (20)$$

The Hamiltonian in the interaction frame is the sum of the two terms,

$$\tilde{H}(t) = \tilde{H}_{CS}(t) + \tilde{H}_D(t) \quad (21)$$

where

$$\tilde{H}_{CS}(t) = \sum_i \sum_{m=-1}^{1} \left(\Delta\omega^i + \sum_{n=-2}^{2} \omega_{CS}^i g_n^i e^{in\omega_r t} \right) \sum_{k=-\infty}^{\infty} \alpha_{1mk} e^{ik\omega_C t} T_{1m}^i \quad (22)$$

and

$$\tilde{H}_D(t) = \sum_{i<j}\sum_{m=-2}^{2} \left\{ \sum_{n=-2}^{2}\sum_{k=-\infty}^{\infty} \sqrt{6}\omega_D^{ij} G_n^{ij} e^{in\omega_r t} \alpha_{2mk} e^{ik\omega_C t} T_{2m}^{ij} \right\} \quad (23)$$

Other interaction Hamiltonians of decoupling multiple-pulse schemes can have similar treatments as above if they are periodic in time

Calculation of the first and second order terms of FME

Considering the RF decoupling cycle time τ_c and the rotor period τ_r such as, $\tau_r = N\tau_c$ or $\omega_c = N\omega_r$. The first order contribution to the FME, for $k \in [-\infty,\infty]$ and n = {-1,-2,0,1,2} is -2

$$F_1 = \frac{1}{\tau_C}\int_0^{\tau_C} \tilde{H}(\tau)d\tau = \sum_i \Delta\omega^i \alpha_{1mk} T_{1m}^i + \sum_i \omega_{CS}^i g_n^i \alpha_{1mk} T_{1m}^i + \sum_{i<j} \sqrt{6}\omega_D^{ij} G_n^{ij} \alpha_{2mk} T_{2m}^{ij} \quad (24)$$

This result is equivalent to the time-independent terms with the coefficients for $k = 0$ and $n + kN = 0$. We can generate uniquely the isotropic chemical shift terms if we set the conditions, $N > 2$ and $n + kN \neq 0$. Hence, we have

$$F_1 = \sum_i \Delta\omega^i \alpha_{1mk} T_{1m}^i \quad (25)$$

The function, $\Lambda_1(t)$, which provides a way for evaluating the spin behavior in between the stroboscopic observation points can be calculated as

$$\begin{aligned}\Lambda_1(t) &= \int_0^t \tilde{H}(\tau)d\tau - tF_1 \\ &= \sum_i \sum_{m=-1}^{1} \Delta\omega^i \alpha_{1mk} T_{1m}^i \sum_{k=-\infty}^{\infty} \int_0^t e^{ik\omega_C\tau} d\tau + \sum_i \sum_{m=-1}^{1}\sum_{n=-2}^{2} \omega_{CS}^i g_n^i \alpha_{1mk} T_{1m}^i \sum_{k=-\infty}^{\infty} \int_0^t e^{i\omega r(n+kN)\tau} d\tau \\ &+ \sum_{i<j}\sum_{m=-2}^{2}\sum_{n=-2}^{2}\sum_{k=-\infty}^{\infty} \sqrt{6}\omega_D^{ij} G_n^{ij} \alpha_{2mk} T_{2m}^{ij} \int_0^t e^{i\omega r(n+kN)\tau} d\tau - tF_1\end{aligned} \quad (26)$$

where we have set $\Lambda_1(0) = 0$ and used the condition, $\omega_c = N\omega_r$. Using both conditions

$$\begin{cases} k = 0, n + kN = 0 \\ N > 2, n + kN \neq 0 \end{cases} \quad (27)$$

we obtain,

$$\begin{aligned}\Lambda_1(t) &= \sum_i \sum_{m=-1}^{1} \Delta\omega^i \alpha_{1mk} T_{1m}^i \sum_{\substack{k=-\infty \\ k\neq 0}}^{\infty} \frac{1}{ikN\omega_r}\left(e^{ikN\omega_r t} - 1\right) + \\ &\sum_i \sum_{m=-1}^{1}\sum_{n=-2}^{2}\sum_k \omega_{CS}^i g_n^i \alpha_{1mk} T_{1m}^i \frac{1}{i(n+kN)\omega_r}\left(e^{i(n+kN)\omega_r t} - 1\right) + \\ &\sum_{i<j}\sum_{m=-2}^{2}\sum_{n=-2}^{2}\sum_k \sqrt{6}\omega_D^{ij} G_n^{ij} \alpha_{2mk} T_{2m}^{ij} \frac{1}{i(n+kN)\omega_r}\left(e^{i(n+kN)\omega_r t} - 1\right)\end{aligned} \quad (28)$$

The second-order contributions to the FME can be calculated as the following,

$$\begin{aligned}F_2 &= \frac{1}{2i\tau_C}\int_0^{\tau_C}\left[\tilde{H}(t) + F_1, \Lambda_1(t)\right]dt \\ &= \frac{1}{2i\tau_C}\int_0^{\tau_C}\left\{\left[\tilde{H}_{CS}(t), \Lambda_1(t)\right] + \left[\tilde{H}_D(t), \Lambda_1(t)\right] + \left[F_1, \Lambda_1(t)\right]\right\}dt \\ &= \frac{1}{2i\tau_C}(I_1 + I_2 + I_3)\end{aligned} \quad (29)$$

where

$$I_1 = \int_0^{\tau_C}\left\{\left[\tilde{H}_{CS}(t), \Lambda_1(t)\right]\right\}dt \quad (30)$$

$$I_2 = \int_0^{\tau_C}\left\{\left[\tilde{H}_D(t), \Lambda_1(t)\right]\right\}dt \quad (31)$$

and

$$I_3 = \int_0^{\tau_C}\left\{\left[F_1, \Lambda_1(t)\right]\right\}dt \quad (32)$$

Let us use the following notation and description,

$$\tilde{H}_{CS}(t) = \left(a_1 e^{ik\omega_C t} + a_2 e^{i(n+kN)\omega_r t}\right) T_{1m}^i \quad (33)$$

$$\tilde{H}_D(t) = a_3 e^{i(n+kN)\omega_r t} T_{2m}^{ij} \quad (34)$$

$$\Lambda_1(t) = a_4\left(e^{ikN\omega_r t} - 1\right)T_{1m}^i + a_5\left(e^{i(n+kN)\omega_r t} - 1\right)T_{1m}^i + a_6\left(e^{i(n+kN)\omega_r t} - 1\right)T_{2m}^{ij} \quad (35)$$

$$F_1 = a_7 T_{1m}^i \quad (36)$$

where the expressions of the coefficients a_1, a_2, a_3,... are given in the appendix. The above integrals I_1, I_2 *and* I_3 can be evaluate as

$$I_1 = \left\{ \begin{aligned} &\int_0^{\tau_C}\left(a_1 e^{ik\omega_C t} + a_2 e^{i(n+kN)\omega_r t}\right) a_4\left(e^{ikN\omega_r t} - 1\right)dt + \\ &\int_0^{\tau_C}\left(a_1 e^{ik\omega_C t} + a_2 e^{i(n+kN)\omega_r t}\right) a_5\left(e^{i(n+kN)\omega_r t} - 1\right)dt \end{aligned} \right\}\left[T_{1m}^i, T_{1m}^i\right] + \cdot\int_0^{\tau_C}\left(a_1 e^{ik\omega_C t} + a_2 e^{i(n+kN)\omega_r t}\right) a_6\left(e^{i(n+kN)\omega_r t} - 1\right)dt\left[T_{1m}^i, T_{2m}^{ij}\right] \quad (37)$$

$$I_2 = \left\{ \begin{array}{l} \int_0^{\tau_C} a_3 e^{i(n+kN)\omega_r t} a_4 \left(e^{ikN\omega_r t} - 1\right) dt + \\ \int_0^{\tau_C} a_3 e^{i(n+kN)\omega_r t} a_5 \left(e^{i(n+kN)\omega_r t} - 1\right) dt \end{array} \right\} \left[T_{2m}^{ij}, T_{1m}^{i}\right] + \int_0^{\tau_C} a_3 e^{i(n+kN)\omega_r t} a_6 \left(e^{i(n+kN)\omega_r t} - 1\right) dt \left[T_{2m}^{ij}, T_{2m}^{ij}\right] \quad (38)$$

$$I_3 = \left\{ \begin{array}{l} \int_0^{\tau_C} a_{10} a_4 \left(e^{ikN\omega_r t} - 1\right) dt + \\ \int_0^{\tau_C} a_{10} a_5 \left(e^{i(n+kN)\omega_r t} - 1\right) dt \end{array} \right\} \left[T_{1m}^{i}, T_{1m}^{i}\right] + \int_0^{\tau_C} a_{10} a_6 \left(e^{i(n+kN)\omega_r t} - 1\right) dt \left[T_{1m}^{i}, T_{2m}^{ij}\right] \quad (39)$$

As discussed above, obtaining the FME terms will allow calculating the propagators with the 0th-and 1rst - order average Hamiltonians,

$$U(\tau_C) \approx \exp\left\{-i\tau_C(\overline{H}_{FME}^{(0)} + \overline{H}_{FME}^{(1)}) + \ldots\right\} \quad (40)$$

The basic objective of decoupling methods is to produce an effective Hamiltonian that is exclusively influenced by the isotropic chemical shift interaction. The decoupling techniques have been described originally by the average Hamiltonian method [4]. In this light, the Lee-Goldburg pulse eliminates the dipolar interaction to zeroth order [31] and the FSLG to first order [44] in AHT. These methods were studied with the assumption of static conditions and only recently that the inclusion of magic-angle spinning during the experiment yield the theoretical treatment with a bimodal Floquet theory [33,34].

Spin Physics

It is important to mention that the physics of a single-particle system is different from the one of a spin ensemble. The ambiguity arises because of the adoption of simple matrix representation like Pauli or equivalent representation for operator in describing the spin system in both synopsis. In principle, for a many-body system, one would have to arrogate a rank-n spin tensor or a 2n-th dimensional matrix representation. But, this is unwieldy as the individual spin packets are identical and assumed to be non-interacting with other spin packets, i.e. the off-diagonal elements in the giant matrix representation are zero. Hence, it is possible to block diagonalize the giant matrix, which is sufficient to use the simple Pauli matrices along with density operator to describe the entire spin ensemble. One important ambivalence in the theory of solid-state NMR is that the act of measurement causes the spin state to collapse to the eigenstate of the measurement operator, i.e. the FID cannot be recorded continuously, and quadrature detection is not possible. The flaw lies in the fact that the previous statement is only applicable to the case of single particle, while solid-state NMR is dealing with a spin ensemble [45].

Appendix

A1. The average Hamiltonian and the propagator of the FME

Starting with the Schrodinger Equation, and introducing the expansions [46-50].

$$\Lambda(t) = \sum_n \Lambda_n(t) \quad (41)$$

and

$$F = \sum_n F_n \quad (42)$$

the FME expansion can be summarized as

$$F_n = \frac{1}{\tau c}\int_0^{\tau c} G_n(u)du, \quad (43)$$

$$\Lambda_n(t) = \Lambda_n(0) + \int_0^t G_n(u)du - tF_n \quad (44)$$

The first functions $G_n(t)$ are defined as

$$G_1(t) = H(t) \quad (45)$$

$$G_2(t) = -\frac{i}{2}[H(t)+F_1, \Lambda_1(t)] \quad (46)$$

$$G_3(t) = -\frac{i}{2}[H(t)+F_1, \Lambda_2(t)] - \frac{i}{2}[F_2, \Lambda_1(t)] - \frac{i}{12}[\Lambda_1(t), H(t)-F_1] \quad (47)$$

τ_c is the period of the modulation such as

$$\Lambda_n(\tau_c) = \Lambda_n(0) \quad (48)$$

$$H(\tau_c + t) = H(t) \quad (49)$$

A much better choice is given by the general rule

$$\int_0^{\tau c} \Lambda_n(u)du = 0 \quad (50)$$

The second order, F_2, can be written in its general form as

$$F2 = \frac{1}{2}\int_0^{\tau c} G_2(u)du = -\frac{i}{2\tau c}\int_0^{\tau c}[H(u), \Lambda_1(u)]du - \frac{i}{2\tau c}\int_0^{\tau c}[F1, \Lambda_1(u)]du \quad (51)$$

The second term cancels, thus yielding

$$F_2 = -\frac{i}{2\tau c}\int_0^{\tau c}[H(u), \Lambda_1(u)]du \quad (52)$$

Introducing,

$$H(t) = \sum_m H_m e^{im\omega t} \quad (53)$$

with

$$\omega = 2\pi/\tau_c \quad (54)$$

we obtained the well know results [28,29]

$$F_1 = H_0 \quad (55)$$

$$A_1(t) = \Lambda 1(0) + \int_0^t \sum_{m\neq 0} H_m e^{im\omega t} dt = \int_0^t \sum_{m\neq 0} \frac{H_m}{im\omega} e^{im\omega t} \quad (56)$$

$$F_2 = \frac{1}{2}\sum_{m\neq 0}\frac{[H_m, H_{-m}]}{m\omega} \quad (57)$$

whereas the choice

$$\Lambda_n(0) = 0 \quad (58)$$

gives the Magnus expansion

$$\Lambda_1(t) = \int_0^{\tau} \sum_{m\neq 0} H_m e^{im\omega t} dt = \sum \frac{H_m}{im\omega}(e^{im\omega t} - 1) \quad (59)$$

$$F_2 = \frac{1}{2}\sum_{m\neq 0}\frac{[H_m, H_{-m}]}{m\omega} + \sum_{m\neq 0}\frac{[H_0, H_m]}{m\omega} \quad (60)$$

A3. Coefficients in the integrals I_1, I_2 and I_3

$$a_1 = \sum_i \sum_{m=-1}^{1} \sum_{k=-\infty}^{\infty} \Delta\omega^i \alpha_{1mk} \quad (61)$$

$$a_2 = \sum_i \sum_{m=-1}^{1} \sum_{n=-2}^{2} \sum_{k=-\infty}^{\infty} \omega_{CS}^i g_n^i \alpha_{1mk} \quad (62)$$

$$a_3 = \sum_{i<j} \sum_{m=-2}^{2} \sum_{n=-2}^{2} \sum_{k=-\infty}^{\infty} \sqrt{6}\omega_D^{ij} G_n^{ij} \alpha_{2mk} \quad (63)$$

$$a_4 = \sum_i \sum_{m=-1}^{1} \sum_{k=-\infty}^{\infty} \Delta\omega^i \alpha_{1mk} \frac{1}{ikN\omega_r} \quad (64)$$

$$a_5 = \sum_i \sum_{m=-1}^{1} \sum_{n=-2}^{2} \sum_{k=-\infty}^{\infty} \omega_{CS}^i g_n^i \alpha_{1mk} \frac{1}{i(n+kN)\omega_r} \quad (65)$$

$$a_6 = \sum_{i<j} \sum_{m=-2}^{2} \sum_{n=-2}^{2} \sum_{k=-\infty}^{\infty} \sqrt{6}\omega_D^{ij} G_n^{ij} \alpha_{2mk} \frac{1}{i(n+kN)\omega_r} \quad (66)$$

$$a_7 = \sum_i \sum_{m=-1}^{1} \Delta\omega^i \alpha_{1m0} \quad (67)$$

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Conflicts of Interest

All authors declare that they have no conflicts of interest.

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