

Canonical Perturbation Theory and Lie Transforms: Application to Nonlinear Resonant Wave-Particle Interaction

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Resonant wave-particle interactions has been one of the main paradigms, on which the modern theory of complex dynamics and chaos has been applied. In this work, these interactions are studied in the context of the Hamiltonian formalism and the Canonical Perturbation Theory (CPT) with utilization of Lie Transform techniques (LT). The canonical perturbation method for single particle motion is reviewed and extended in order to provide results for the collective particle behavior under interaction with wave fields of either localized or periodic profiles. Analytical expressions for the calculation of phase-averaged quantities of physical interest as well as diffusion equations are derived.

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1. Introduction

Resonant wave-particle interactions form the underlying mechanism of a variety of phenomena occurring in nature as well as in technological applications and devices, where a plasma or a particle beam interacts with electromagnetic waves. The fundamental importance and the complex character of such interactions have result in extensive research studies and, consequently, in considerable progress in understanding the features of wave and particle dynamics under resonant interactions. Their complicated features, resulting from the nonlinear character of the interaction, have motivated the field of chaotic dynamics for which the wave-particle interactions have been used as a main paradigm (chap. 2, Ref. [1]). On the other hand, resonant wave-particle interactions constitute the operation principle of several devices of major technological interest, and as such they have to be understood and optimized for efficient device design and performance; consequently there exist a large set of still open issues. Among the most important applications is the interaction of rf radiation with tokamak plasmas in fusion devices, for the electron cyclotron resonant heating (ECRH) and current drive (CD) [2, 3], and the electron beam interaction with electromagnetic waves in gyrodevices, for the high-power, high-frequency microwave generation [4–7].

The presence of an electromagnetic (or electrostatic) wave results in perturbation of the free particle motion, so that a test particle can either gain or lose energy, depending drastically on its initial position and momentum. Its motion can become chaotic under certain conditions where resonance overlap occur in the phase space of the system [8–19]. The collective dynamical behavior of a large ensemble of such particles determines the state of the system as well as the energy exchange between the wave and the particles and its study utilizes a kinetic theory description [20–22]. The complete picture of the

plasma state is described by the self-consistent model, consisting of the kinetic (Vlasov) equation coupled with the Maxwell equations for the wave fields. It is in the first part of the self-consistent model, i.e. the Vlasov equation for a given wave field, that we are focusing in this work in order to reduce the original Vlasov equation to an equation having lower number of dimensions; namely, in terms of action-angle variables, an action diffusion equation, where the angle-dependence has been eliminated. This equation, within its respective domain of validity, can replace the Vlasov equation in the fully self-consistent model, and the corresponding action distribution function can be used for the calculation of the source terms of the Maxwell equations, namely charge and current densities.

In most cases the kinetic equation governing the evolution of the particle distribution function is simplified, under certain assumptions, to a quasilinear diffusion equation (QDE) of the Fokker-Planck type [22–24]. The quasilinear diffusion equation, describing an irreversible process corresponding to slow time diffusion of particles and respective wave absorption, is currently the main model for studying the interaction of electromagnetic waves with plasmas. The standard derivation procedure [22, 23] of the QDE utilizes a rather heuristic approach, under which several assumptions come into play. However, the lack of a rigorous method for deriving the QDE as a low order approximation of the original kinetic equation, results to the difficulty of proceeding to a higher order approximation in a unified context and defining a hierarchy of approximating equations, with corresponding domains of validity. The latter is of particular importance, since a number of previous studies [25–27] have shown that nonquasilinear diffusion can take place under the presence of a set of waves with relatively broad spectrum. Thus, several works have studied the origin of the breakdown and the controversies of the quasi-linear theory [28–30] and have considered respective generalizations [29, 31, 32].

In addition, the domain of validity of the quasi-linear theory can be investigated in terms of the nonlinearity parameter [3, 19] defined as $\epsilon_{NL} = t_f/\tau_{bE}$, where t_f and τ_{bE} are the particle flight time through a wavepacket and the oscillation period of a particle trapped inside the wave, respectively. The limiting cases $\epsilon_{NL} \ll 1$ and $\epsilon_{NL} \gg 1$ correspond to the quasilinear and the adiabatic [19] case, respectively. Considering that $\epsilon_{NL} \sim \tan \chi$, [3] where $\chi = \tan^{-1}(v_{\perp}/v_{\parallel})$ is the pitch angle of particles, there is always a cone in the velocity space that falls into the nonlinear regime.

Apart from the kinetic description and the approximate QDE, the collective behavior of a particle beam has been studied analytically with the application of perturbation methods to the particle equations of motion. It has been shown that first order perturbation analysis for the single particle motion can result in second order accurate calculations of phase (or position) averaged quantities, a result that it is known as Madey's theorem. The latter has been mostly applied for the calculation of gain (efficiency) in microwave sources [33].

The main aim of this work is to review and extend the canonical perturbation theory with utilization of Lie transform techniques in order to provide a unified context under which the collective particle behavior interacting with an electromagnetic wave can be studied in terms of rigorously obtained analytical approximations of phase averaged quantities and approximate diffusion equations. The ordering of the respective perturbation scheme is related to the aforementioned nonlinearity parameter, providing thus a direct measure of the domain of validity of the results in the parameter and phase space. Our approach utilizes the canonical perturbation method and the Lie transforms [34–37] as applied to the Hamiltonian system describing the single particle motion and relates the single particle dynamics to the collective particle behavior. In this context, an alternative and more general derivation procedure of the Fokker-Planck QDE is provided and its relation to the Madey's theorem as a quasilinear approximation, is shown. *More importantly, the adopted method allows for extending these results to higher order: It is shown that a third order canonical perturbation analysis allows for fourth order accurate calculations of phase averaged quantities, in analogy with the Madey's theorem, and can also be used in the derivation of a higher order diffusion equation. The latter includes higher order derivatives of the distribution function (than the QDE) and can be considered as a deterministic analogue of a higher order expansion of the master equation of a stochastic process (chap. 9, Ref. [38]).*

Although, the method utilized in this work is quite generic and applicable in a large variety of systems describing resonant wave-particle interactions, the specific paradigm under consideration consists of a Hamiltonian describing the wave-particle interaction close to a cyclotron resonance and it is derived from the fully relativistic Hamiltonian under a set of assumptions. These

simplifications allow for focusing on the consequences of considering perturbations beyond the quasilinear approximation, while there is no loss of generality since most of the essential features of the nonlinear cyclotron resonant wave-particle interactions are taken into account. The corresponding assumptions can be easily removed and the respective effects can be taken into account, generalizing the results for more complex cases. Concerning the form of the wave, the theory is applied in two cases: periodic waves with discrete spectrum, commonly occurring in toroidal configurations and localized waves having continuous spectrum related to the ponderomotive effect in plasmas [39–44] and to wave-particle interactions of finite length in microwave devices [4–7].

The paper is organized as follows. A specific Hamiltonian system is derived from the generic Hamiltonian describing the wave-particle interaction, in the second section. The canonical perturbation method with the utilization of the Lie transforms technique is applied to the Hamiltonian system under consideration, in the third section. The fourth section utilizes the results of the perturbation theory in order to provide high order analytical calculations of phase averaged quantities, while in the fifth section, a higher order diffusion equation is derived. Finally, the results and conclusions are summarized in the last section.

2. Hamiltonian system

In the following we formulate the Hamiltonian system describing the wave-particle interactions. A quite standard derivation procedure of a simplified Hamiltonian is adopted (similar to that of Ref. [3]); however it is briefly given in Appendix A due to some modifications regarding the consideration of a many-waves field. Therefore, we consider a wave electric field consisting of multiple wavepackets and having the form

$$\mathbf{E} = \sum_i E_0^{(i)}(\mathbf{r}) \Re e \left[\mathbf{f}^{(i)} F^{(i)}(\mathbf{r}) e^{i(\mathbf{k}_i \cdot \mathbf{r} - \omega_i t)} \right] \quad (1)$$

where $E_0^{(i)}(\mathbf{r})$ is the amplitude which is constant along the magnetic field (assumed to be uniform), $\mathbf{f}^{(i)} \equiv \mathbf{E}^{(i)}/|\mathbf{E}^{(i)}|$ is the complex polarization vector, \mathbf{k}_i is the wave vector, ω_i is the wave frequency, and the function $F^{(i)}(\mathbf{r})$ describes the electric field profile. Each wave may correspond to a mode given by a specific and, in general, different branch of the plasma dispersion relation. A Cartesian coordinate system (x, y, z) is used so that $\mathbf{B} = \mathbf{e}_z B_0$ and $\mathbf{k}_i = \mathbf{e}_x k_{\perp,i} + \mathbf{e}_z k_{\parallel,i}$, where $(\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z)$ are the corresponding unit vectors. In the following, it is assumed that the perpendicular scale of $E_0^{(i)}$, $\mathbf{f}^{(i)}$ and $F^{(i)}$ is large compared to the particle gyration radius and the variation of the polarization vector along the magnetic field is considered negligible, resulting to $E_0^{(i)} = \text{const}$, $\mathbf{k}_i = \text{const}$ and $F^{(i)}(\mathbf{r}) = F^{(i)}(z)$. As shown in Appendix A, the simplified Hamiltonian describing the particle

motion under interaction with the waves has the following form:

$$H = H_0(J) + \epsilon H_1(J, \theta, t) \quad (2)$$

where

$$H_0(J) = J^2, \quad H_1(J, \theta, t) = -\frac{E}{2} (2J)^{k_0/2} e^{ik_0\theta} g(t) + c.c. \quad (3)$$

The parameter ϵ is a dimensionless parameter, which denotes the fact that $H_1(J, \theta)$ is treated as a small perturbation; it will be used for counting the order of the expansion in the following perturbation method and it can be set equal to unity in the final results. The effective strength of the perturbation introduced by each wave is directly related to the nonlinearity parameter ϵ_{NL} as given in [3]. (J, θ) are the action-angle variables of the unperturbed system $H_0(J)$ describing the free particle motion (under the absence of the wave). The function $g(t)$ provides the total wave field determined by the profile $F^{(i)}(z)$ and the frequency mismatch (Ω_i) with respect to the k_0 harmonic of the gyrofrequency, of each wavepacket:

$$g(t) = \sum_i w_E^{(i)} F^{(i)}(t) e^{i(k_0\theta - \Omega_i t)} \quad (4)$$

It is worth mentioning that the Hamiltonian (2) appears in a wide range of applications where wave-particle interactions occur such as ECRH and ECCD in fusion plasmas [3] as well as in gyrotron cavities ([5–7, 18]). This model is capable of describing the basic underlying mechanism of cyclotron resonance which is of physical and technological interest in configurations based on wave-particle interactions. Moreover, the generic form of the wave profiles, considered in the model, allows for the study of particle interactions with a variety of waves such as periodic waves having discrete spectra and/or solitary waves having continuous spectra, as well as pulses ranging from ordinary (adiabatic) wavepackets to ultrashort (fewcycle) impulses.

3. Lie transform perturbation theory

In order to extend the application of the Canonical Perturbation Theory to higher order, the utilization of the Lie transform theory [34, 35] is necessary for treating the complexity of the expansions. Although the method of Lie transform is, in spirit, identical to the Poincare-Von Zeipel method [36], which is based on the classical mixed variable generating functions (discussed in text books on Classical Mechanics, such as [37]), there are at least two important advantages, in favor of this method: (i) the transformations as expressed in terms of Lie operators are significantly simpler, and (ii) the Lie operators commute with functions, a property that in

the following will be shown very useful for calculating the evolution of phase space functions and their phase averages.

Before considering our specific case, let us briefly summarize some of the essential concepts of the Lie transform perturbation theory. Without loss of generality we consider the case of a nonautonomous system with one degree of freedom such as the one considered in the following. The evolution of a function of the phase space variables $\mathbf{z}(t)$ (and time) $f(\mathbf{z}, t)$ from time t_0 to time t can be provided by the time development operator $S_H(t; t_0)$:

$$f(\mathbf{z}(t; t_0), t) = S_H(t; t_0) f(\mathbf{z}_0, t_0) \quad (5)$$

with $\mathbf{z}(t; t_0)$ satisfying the Hamilton equations of motion (with Hamiltonian $H(\mathbf{z})$) under the initial condition $\mathbf{z}(t_0; t_0) = \mathbf{z}_0$. The derivation of the operator $S_H(t; t_0)$ is equivalent with solving the equations of motion, which is not possible for most cases. Instead, a change of variables under the transform $\mathbf{z}' = T(\mathbf{z}, t)\mathbf{z}$ can lead to a new system with Hamiltonian $K(\mathbf{z}')$, in which the time development operator $S_K(t; t_0)$ can be easily computed. These are the cases for which the new system is, either integrable with \mathbf{z}' corresponding to the action-angle variables the new Hamiltonian, or, more generally, when the new Hamiltonian does not depend on the phases and the action of the operator $S_K(t; t_0)$ to a function $f(\mathbf{z}', t)$ leaves the actions unchanged and evolves the phases and time according to:

$$f(\mathbf{z}'(t; t_0), t) = S_K(t; t_0) f(\mathbf{z}'_0, t_0) = f(J'_0, \theta'_0 + \theta') \quad (6)$$

where

$$\theta' = \int_{t_0}^t \omega_K(J'_0, s) ds, \quad \omega_K(J'_0, t) = \frac{\partial K(J'_0, t)}{\partial J'_0} \quad (7)$$

In that sense, the solution of the system can be given if the appropriate transformation T is constructed. According to Lie transform theory, the operator T can be represented as

$$T = e^{-L} \quad (8)$$

where $Lf = [w, f]$, for any function $f(\mathbf{z}, t)$, with $[\cdot, \cdot]$ denoting the Poisson bracket. The function $w(\mathbf{z})$ is defined as the Lie generator and the operator of the inverse transformation is $T^{-1} = e^L$. The Lie transform operator has the important properties that generates canonical transformations and commutes with functions. The latter implies directly that the evolution of a function $f(\mathbf{z}, t)$ can be calculated by subsequently transforming to the new variable set \mathbf{z}' , applying the time development operator $S_K(t; t_0)$ and transforming back to the original variables \mathbf{z} , according to

$$f(\mathbf{z}(t; t_0), t) = T(\mathbf{z}_0, t_0) S_K(t; t_0) T^{-1}(\mathbf{z}_0, t_0) f(\mathbf{z}_0, t_0) \quad (9)$$

The aforementioned procedure apart from being applicable to integrable system, it also provides a perturbation method for solving approximately near-integrable systems, in which the Hamiltonian has a small nonintegrable part of order ϵ . In such cases the canonical transform T can be constructed as a power series in ϵ , by utilizing the method of Deprit, according to which the old Hamiltonian H , the new Hamiltonian K and the transformation T along with the Lie generator w are expanded in power series of ϵ (Appendix B).

Notice that, although we need the transformation expansion for T up to fourth order, we will need only to derive the Lie generator w up to third order. As it will be shown in the following, the knowledge of the Lie generator up to third order (actually w_1 , w_2 and only a part of w_3 is needed) allows for calculations of phase averaged functions, describing collective particle characteristics, which are accurate up to fourth order. This result corresponds to a higher order extension of the Madey's theorem and it will also be crucial for the derivation of the high order diffusion equation governing the evolution of the particle momentum (action) distribution.

Within our approach, equations (B.5)-(B.7), providing w_1 , w_2 and w_3 respectively, will be solved in the finite time interval $[t_0, t]$. This approach will be proved appropriate for our purposes for the following reasons: (i) the operator governing the evolution of phase space functions shown in (9) is greatly simplified, (ii) the problem of small denominators, appearing in the case of infinite time intervals, is avoided, [45] and (iii) time-infinitesimal canonical transformations (from t to $t + \Delta t$) related to the derivation of high order diffusion equations can be directly considered.

For our specific Hamiltonian (2) we consider only first order perturbations ($H_n = 0$, for $n > 1$). The equation for w_1 is

$$\frac{\partial w_1}{\partial t} + \omega_0(J) \frac{\partial w_1}{\partial \theta} = K_1 + (P_{1,1} e^{ik_0 \theta} + c.c.) \quad (10)$$

where

$$P_{1,1} = \frac{1}{2} E (2J)^{k_0/2} g(t) \quad (11)$$

and $\omega_0(J) = \partial H_0(J) / \partial J = 2J$ is the unperturbed frequency. Since there is no θ -independent term in the r.h.s. we can set $K_1 \equiv 0$. The solution in the interval $[t_0, t]$ is

$$w_1(J, \theta, t; t_0) = F_{1,1} e^{ik_0 \theta} + c.c.$$

with

$$F_{1,1} = \frac{1}{2} E (2J)^{k_0/2} e^{-ik_0 \omega_0 t} \int_{t_0}^t g(s) e^{ik_0 \omega_0 s} ds \quad (12)$$

as obtained from (B.10).

Proceeding to second order, the equation for w_2 is

$$\frac{\partial w_2}{\partial t} + \omega_0(J) \frac{\partial w_2}{\partial \theta} = 2K_2 + (P_{2,0} + P_{2,2} e^{i2k_0 \theta} + c.c.) \quad (13)$$

with

$$P_{2,0} = ik_0 \frac{\partial}{\partial J} (F_{1,1} \bar{P}_{1,1}) \quad (14)$$

$$P_{2,2} = -ik_0 P_{1,1}^2 \frac{\partial}{\partial J} \left(\frac{F_{1,1}}{P_{1,1}} \right) \quad (15)$$

By defining

$$2K_2 = -(P_{2,0} + c.c.) \quad (16)$$

or

$$K_2 = \frac{1}{4} E^2 k_0^2 (2J)^{k_0-1} \bar{g}(t) \int_{t_0}^t [2J(s-t) - i] \times g(s) e^{ik_0 \omega_0 (s-t)} ds + c.c. \quad (17)$$

the second order generating function w_2 is obtained as

$$w_2 = F_{2,2} e^{i2k_0 \theta} + c.c.$$

$$F_{2,2} = \frac{1}{2} E^2 k_0^2 (2J)^{k_0} e^{-i2k_0 \omega_0 t} \int_{t_0}^t g(s) e^{ik_0 \omega_0 s} \times \left(\int_{t_0}^s (s'-s) g(s') e^{ik_0 \omega_0 s'} ds' \right) ds \quad (18)$$

The third order equation is

$$\frac{\partial w_3}{\partial t} + \omega_0(J) \frac{\partial w_3}{\partial \theta} = 3K_3 + (P_{3,1} e^{ik_0 \theta} + P_{3,3} e^{i3k_0 \theta} + c.c.) \quad (19)$$

with

$$P_{3,1} = \frac{ik_0}{2} \left[2F_{1,1} \frac{\partial}{\partial J} (P_{2,0} + \bar{P}_{2,0}) + \frac{1}{P_{1,1}} \frac{\partial}{\partial J} (F_{2,2} \bar{P}_{1,1}^2) - \frac{1}{\bar{F}_{1,1}} \frac{\partial}{\partial J} (P_{2,2} \bar{F}_{1,1}^2) \right], \quad (20)$$

$$P_{3,3} = \frac{ik_0}{2} \left[F_{1,1}^3 \frac{\partial}{\partial J} \left(\frac{P_{2,2}}{F_{1,1}^2} \right) - P_{1,1}^3 \frac{\partial}{\partial J} \left(\frac{F_{2,2}}{P_{1,1}^2} \right) \right] \quad (21)$$

Since there is no θ -independent term we can set $K_3 \equiv 0$, and the third order generating function is obtained as

$$w_3 = F_{3,1} e^{ik_0 \theta} + F_{3,3} e^{i3k_0 \theta} + c.c. \quad (22)$$

with $F_{3,1}, F_{3,3}$ provided implicitly through eqs. (B.10), (11), (14)-(15) and (20)-(21) (their explicit forms are too lengthy to be presented). However, only $F_{3,1}$ will be needed for the calculation of phase averaged quantities and the derivation of the action diffusion equation up to fourth order, as will be shown in the following section.

Once we have calculated the Lie generating functions we can define the canonical transformation from the action angle variables of the unperturbed system $\mathbf{z} = (J, \theta)$ to the new variables $\mathbf{z}' = (J', \theta')$, up to third order. This transformation allows for construction of approximate invariants of the motion which contain all the essential information of particle dynamics and can be used for providing approximately the phase space topology of the system. On the other hand, knowledge of the Lie generator allows for the definition of a symplectic (canonical) mapping which can be utilized for accurate particle trajectory calculations [45, 46]. In comparison with standard (non-canonical) integration schemes, this mapping has the advantage of being directly related to the specific Hamiltonian system, thus preserving the phase space volume and all the invariants of the motion.

4. Calculation of averaged quantities

In the previous section we have calculated the canonical transformation to the new variable set, up to third order; the corresponding invariant of the motion provides information for the single particle dynamics which are accurate up to third order. However, in most cases where wave-particle interactions occur, we are interested in the collective particle behavior, which is usually expressed through phase-averaged quantities of an ensemble of particles, having different initial conditions. In the following, we will show that knowledge of the Lie generators up to second order and partial knowledge of the third order Lie generator is capable of determining such phase-averaged quantities up to fourth order ($O(\epsilon^4)$). This result can be considered as a higher-order extension of the Madey's theorem [33], which shows that we can calculate phase-averaged quantities with ($O(\epsilon^2)$)-accuracy by utilizing first order ($O(\epsilon^1)$) perturbation theory.

The evolution of any function of the phase space variables $G(\mathbf{z}) = G(J, \theta)$ is determined through equation (9). As a result of solving the equations providing the Lie generators in the finite time interval $[t_0, t]$ one can easily show that $w_n(\mathbf{z}_0, t_0) = 0$ and

consequently $T(\mathbf{z}_0, t_0) = I$. On the other hand, we have $S_K(t; t_0)T^{-1}(\mathbf{z}_0, t_0) = T^{-1}(J_0, \theta_0 + \theta', t)$, so that

$$G(J(t), \theta(t)) = T^{-1}(J_0, \theta_0 + \theta', t)G(J_0, \theta_0) \quad (23)$$

The case where G is a function of the action only is the most interesting since it is related to energy exchange between the particles and the wave, electric current calculations and kinetic energy distribution of the particles. The average of such a function $G(J)$ over an ensemble of particles having a uniform initial phase distribution and an initial action distribution $F(J_0)$, is

$$\begin{aligned} & \langle G(J(t)) \rangle_{(J_0, \theta_0)} \\ &= \langle T^{-1}(J_0, \theta_0 + \theta', t)G(J_0)F(J_0) \rangle_{(J_0, \theta_0)} \\ &= \left\langle \left\langle T^{-1}(J_0, \theta_0 + \theta', t)G(J_0) \right\rangle_{\theta_0} F(J_0) \right\rangle_{J_0} \end{aligned} \quad (24)$$

where $\langle \cdot \rangle_x$ denotes averaging with respect to x .

It can be easily shown that for the phase-averaging of the term $T^{-1}G$ in (24) the following hold for $m, n = 1, 2$:

$$\langle L_n^1 G \rangle_{\theta_0} = \langle L_n^3 G \rangle_{\theta_0} = 0, \quad \langle L_n L_m G \rangle_{\theta_0} = 0, \quad m \neq n \quad (25)$$

while

$$\langle L_1 L_3 G \rangle_{\theta_0} = \langle L_1 L_{3,1} G \rangle_{\theta_0}, \quad \langle L_3 L_1 G \rangle_{\theta_0} = \langle L_{3,1} L_1 G \rangle_{\theta_0} \quad (26)$$

where, using the linearity of the Poisson bracket, we write

$$L_x G(J) = \sum_m L_{x,m} G(J) + c.c. \quad (27)$$

with

$$L_{x,m} = [F_{x,m} e^{imk_0 \theta}, \cdot] + c.c. \quad (28)$$

These relations show that only the term $F_{3,1} \exp(ik_0 \theta)$ results in nonzero phase-averaged contribution, from the third order Lie generating function w_3 , and that, from the sixteen terms of the operator T^{-1} , as obtained through fourth order (B.3), only half of them remain nonzero after phase-averaging. More importantly, *the phase-averaged operator T^{-1} considered up to fourth order $O(\epsilon^4)$ contains only terms involving lower order Lie generating functions, namely w_1, w_2 and part of w_3 .* Therefore we have

$$\begin{aligned} \langle T^{-1}G \rangle_{\theta_0} &= \langle G \rangle_{\theta_0} + \frac{1}{2} \langle L_1^2 G \rangle_{\theta_0} + \frac{1}{8} \langle L_2^2 G \rangle_{\theta_0} + \frac{1}{24} \langle L_1^4 G \rangle_{\theta_0} + \frac{1}{24} \langle L_1^2 L_2 G \rangle_{\theta_0} \\ &+ \frac{1}{12} \langle L_1 L_2 L_1 G \rangle_{\theta_0} + \frac{1}{8} \langle L_2 L_1^2 G \rangle_{\theta_0} + \frac{1}{12} \langle L_1 L_{3,1} G \rangle_{\theta_0} + \frac{1}{3} \langle L_{3,1} L_1 G \rangle_{\theta_0} \end{aligned} \quad (29)$$

with

$$\frac{1}{2} \langle L_1^2 G \rangle_{\theta_0} = k_0^2 (G' |F_{1,1}|^2)' \tag{30}$$

$$\frac{1}{8} \langle L_2^2 G \rangle_{\theta_0} = k_0^2 (G' |F_{2,2}|^2)' \tag{31}$$

$$\frac{1}{24} \langle L_1^4 G \rangle_{\theta_0} = \frac{k_0^4}{12} \left[\left(3 (G' |F_{1,1}|^2)'' - G' (|F_{1,1}|^2)'' \right) |F_{1,1}|^2 \right]' \tag{32}$$

and

$$\frac{\langle L_1^2 L_2 G \rangle_{\theta_0}}{24} + \frac{\langle L_1 L_2 L_1 G \rangle_{\theta_0}}{12} + \frac{\langle L_2 L_1^2 G \rangle_{\theta_0}}{8} = -\frac{k_0^3}{6} \left[4 \Im m (F_{1,1}^2 \bar{F}_{2,2}) G'' + 2 (\Im m (F_{1,1}^2 \bar{F}_{2,2}) G')' \right]' \tag{33}$$

$$\begin{aligned} \frac{1}{12} \langle L_1 L_{3,1} G \rangle_{\theta_0} + \frac{1}{3} \langle L_{3,1} L_1 G \rangle_{\theta_0} \\ = \frac{k_0^2}{3} [\Re e (\bar{F}_{1,1} F_{3,1}) G']' \end{aligned} \tag{34}$$

with the prime denoting differentiation with respect to the action. Note that if we keep only the $O(\epsilon^2)$ term (30) we have the well-known result of the Madey's

theorem. [33] Also, it is worth mentioning that there are no terms are of order $O(\epsilon^3)$, meaning that it is necessary to proceed to next order for increasing the calculation accuracy. The remaining terms are all of order $O(\epsilon^4)$, so that they all have to be taken into account in order to have consistent calculation of the averaged quantities with error of the order $O(\epsilon^5)$. Therefore the variation of a function $G(J)$ can be written as

$$\begin{aligned} \langle \Delta G \rangle_{(J_0, \theta_0)} = \left\langle \left\{ k_0^2 [G' (|F_{1,1}|^2 + |F_{2,2}|^2)]' + \frac{k_0^2}{3} [\Re e (\bar{F}_{1,1} F_{3,1}) G']' - \frac{k_0^3}{6} [4 \Im m (F_{1,1}^2 \bar{F}_{2,2}) G'' \right. \right. \\ \left. \left. + 2 (\Im m (F_{1,1}^2 \bar{F}_{2,2}) G')' \right]' + \frac{k_0^4}{12} \left[\left(3 (G' |F_{1,1}|^2)'' - G' (|F_{1,1}|^2)'' \right) |F_{1,1}|^2 \right]' \right\} F(J_0) \right\rangle_{J_0} \end{aligned} \tag{35}$$

Of particular interest is the calculation of functions of the form $G(J) = J^n, n = 1, 2, 3, ..$ which are related to standard quantities describing the collective behavior of the particles under the presence

of a wave. Therefore, for $G(J) = J$ we can obtain the mean value of the action variation corresponding to momentum / energy exchange between the the wave and an ensemble of particles:

$$\langle \Delta J \rangle_{(J_0, \theta_0)} = k_0^2 (|F_{1,1}|^2 + |F_{2,2}|^2)' + \frac{k_0^2}{3} \Re e (\bar{F}_{1,1} F_{3,1})' - \frac{k_0^3}{3} \Im m (F_{1,1}^2 \bar{F}_{2,2})'' + \frac{k_0^4}{6} \left[(|F_{1,1}|^2)'' |F_{1,1}|^2 \right]' \tag{36}$$

where an initial action distribution $F(J_0) = \delta(J - J_0)$ has been considered, for simplicity. Similarly, we can obtain $\langle \Delta J^2 \rangle_{\theta_0}$, related to the standard deviation

and the effective width of the action distribution as well as $\langle \Delta J^3 \rangle_{\theta_0}$ related to the skewness which is a measure of the asymmetry of the action distribution induced due to the interaction with a wave. Finally, if

we consider the phase-averaged distribution function

itself $G(J) = \langle F(J, \theta) \rangle_\theta$, we have

$$\begin{aligned} \langle \Delta F(J) \rangle_{\theta_0} &= \left\{ k_0^2 (G'(|F_{1,1}|^2 + |F_{2,2}|^2))' + \frac{k_0^2}{3} [\Re e(\bar{F}_{1,1} F_{3,1}) G']' - \frac{k_0^3}{3} \right. \\ &\times \left[2\Im m(F_{1,1}^2 \bar{F}_{2,2}) G'' + (\Im m(F_{1,1}^2 \bar{F}_{2,2}) G')' \right] + \frac{k_0^4}{12} \left[(3(G|F_{1,1}|^2)'' - G'(|F_{1,1}|^2)'')|F_{1,1}|^2 \right]' \left. \right\} F(J_0) \end{aligned} \quad (37)$$

which relates the initial action distribution function at t_0 with its form after evolution for a finite time interval.

of the original Liouville equation (38) to the same order with the order of the operator T^{-1} .

For the phase-averaged distribution $F(J)$ we have

5. Higher order diffusion equation

In this section we consider the evolution of the action distribution function and derive a fourth order diffusion equation which at the second order reduces to the Fokker-Planck equation corresponding to the quasilinear approximation. In order to derive the diffusion equation, along the lines of the previous section, we consider that the function of the action $G(J)$ is the phase-averaged distribution function $F(J) = \langle f(J, \theta) \rangle_\theta$, where $f(J, \theta)$ is the phase-space distribution function, the evolution of which is governed by the Liouville equation (chap. 9, Ref. [37])

$$\frac{\partial f}{\partial t} + [f, H] = 0 \quad (38)$$

By considering an infinitesimal transformation in the interval $[t, t + \Delta t]$, the evolution of the distribution function f , according to (23) is given by

$$f(J, \theta)_{t+\Delta t} - f(J, \theta)_t = \tilde{T}^{-1}(J, \theta + \omega \Delta t, t + \Delta t) f(J, \theta)_t \quad (39)$$

where $f(J, \theta)_t = f(J(t), \theta(t))$ and $\tilde{T}^{-1} \equiv T^{-1} - I$. Note that since \tilde{T}^{-1} is a canonical transformation both the sign and the normalization (number of particles) are invariants under the evolution (chap.1, Ref. [47]). By dividing both parts with Δt and considering the limit $\Delta t \rightarrow 0$ we obtain

$$\frac{\partial f(J, \theta, t)}{\partial t} = \frac{\partial \tilde{T}^{-1}(J, \theta, t)}{\partial t} f(J, \theta, t) \quad (40)$$

This equation can be considered as an approximation

$$\frac{\partial F(J, t)}{\partial t} = \frac{\partial \langle \tilde{T}^{-1}(J, \theta, t) \rangle_\theta F(J, t)}{\partial t} \quad (41)$$

Equation (41) can be considered as a high order diffusion equation with the highest order of the derivatives of F with respect to the action J being equal to the order of the operator T^{-1} . At the second order $O(\epsilon^2)$, according to (30), the well known Fokker-Planck equation is derived

$$\frac{\partial F}{\partial t} = \frac{\partial}{\partial J} \left(D(J, t) \frac{\partial F}{\partial J} \right) \quad (42)$$

corresponding to the quasilinear approximation, with

$$D(J, t) = k_0^2 \frac{\partial |F_{1,1}|^2}{\partial t} \quad (43)$$

and F_1 obtained from (12). It can be easily shown that

$$\langle (\Delta J_1)^2 \rangle_\theta = 2k_0^2 |F_{1,1}|^2 \quad (44)$$

where $\Delta J_1 = L_1 J$ is the first order action variation so that D can be written as

$$D(J, t) = \lim_{\Delta t \rightarrow 0} \frac{\langle (\Delta J_1)^2 \rangle_\theta}{2\Delta t} \quad (45)$$

which corresponds to the definition of the quasilinear diffusion coefficient (chap. 8, Ref. [38]).

The fourth-order diffusion equation is directly derived from equations (29)-(33) and (41)

$$\begin{aligned} \frac{\partial F}{\partial t} = & k_0^2 \frac{\partial}{\partial J} \left[(|F_{1,1}|^2 + |F_{2,2}|^2)_t \frac{\partial F}{\partial J} \right] + \frac{k_0^2}{3} \frac{\partial}{\partial J} \left[\Re e(\bar{F}_{1,1} F_{3,1})_t \frac{\partial F}{\partial J} \right] - \frac{k_0^3}{6} \frac{\partial}{\partial J} \left[4 \Im m(F_{1,1}^2 \bar{F}_{2,2})_t \frac{\partial^2 F}{\partial J^2} \right. \\ & \left. + 2 \frac{\partial}{\partial J} \left(\Im m(F_{1,1}^2 \bar{F}_2)_t \frac{\partial F}{\partial J} \right) \right] + \frac{k_0^4}{12} \frac{\partial}{\partial J} \left[\left(3 \frac{\partial^2}{\partial J^2} \left(|F_{1,1}|^2 \frac{\partial F}{\partial J} \right) - \frac{\partial^2 (|F_{1,1}|^2)}{\partial J^2} \frac{\partial F}{\partial J} \right) |F_{1,1}|^2 \right]_t \end{aligned} \quad (46)$$

where the operator $(\cdot)_t$ denotes the partial derivative with respect to t acting only to $F_{x,m}$.

Equation (46) can be considered as a deterministic analogue of a higher order expansion of the master equation of a stochastic process (chap. 9, Ref. [38]). It is important to emphasize the addition of higher order derivatives of the distribution function. In fact, it is inconsistent to write to second order (quasilinear) Fokker-Planck equation in which the diffusion coefficient (45) has been calculated to higher than first order accuracy, without retaining the higher-order derivatives of the distribution function (chap. 9, sec. 6, [38]). An other point which needs to be noticed for the higher order diffusion equations is the conservation of the sign (positivity) of an evolving distribution function and of the normalization (number of particles) as well. The preservation of these two properties is ensured for the case of exact canonical transformation, such as the infinitesimal transformation used in eq. (39) and leading to eq. (40), which contains derivatives of infinite order if all terms of the series expansion of the canonical transformation are kept. When the corresponding series of the transformation is truncated at some order, the corresponding transformation is no longer exactly canonical; however it does converge to a canonical perturbation in the limit of small perturbation strength ($\epsilon \rightarrow 0$). In the same spirit, the positivity and the normalization of the distribution functions, are preserved for small perturbations. A similar feature has also been considered in the context of higher order expansions of the master equation for the case of stochastic processes [38], where it is shown [48] that higher order diffusion equations, obtained through the Kramers-Moyal expansion do not preserve the positivity of the distribution function, in general. In order to ensure this property, the Kramers-Moyal expansion has to be truncated either after the second term, resulting to the Fokker-Planck equation (42), or an infinite number of terms has to be retained. However, in spite of the fact that loss of the positivity contradicts our intuition for a distribution function, higher order approximations of the distribution function are useful. They provide better approximations of the actual distribution functions in terms of any integral norm L^p , and this is not just a mathematical issue: In almost all cases of physical interest, calculations of specific integrals and moments of the distribution function is the focal issue. Thus, although the higher order approximate distribution function can become slightly negative (usually in the tails) for strong perturbations, it

is capable of providing excellent approximations of quantities of physical interest. [38, 49]

From a physical point of view, higher-order terms are proportional to the third and fourth power of the wave amplitudes, and are related to nonlinear cyclotron resonances between particles and the beats of more than one spectral components of the waves. These terms describe the effect of nonlinear coupling between the different wave components on the evolution of the particle distribution function. Although the effect of such mode coupling has been extensively studied with respect to the evolution of the wave components, there are only a few works on the topic of such nonlinear corrections on the particle distribution function, as also mentioned in [32]. These higher-order corrections are significant in cases where the linear (quasilinear) growth rate is small because only a few particles can resonate with the wave, or in cases of super-thermal particles with very high velocities which can interact resonantly with the beating of two or more spectral components of the waves. Also, higher-order terms and nonlinear resonant wave coupling have been considered as responsible for the breakdown of quasilinear theory [28–30] and the numerical observations of non-quasilinear diffusion [25–27] in one-dimensional Langmuir turbulence where they have been related to nonlinear Landau damping.

6. Summary and conclusions

Resonant wave-particle interactions have been studied within the context of canonical perturbation method and Lie transforms. The aim of this work is to provide a theoretical approach, under which the perturbation theory of single particle motion is related to two aspects of the collective particle behavior: the calculation of phase-averaged quantities of physical interest and the derivation of a diffusion equation. In the first order of perturbation the method provides a formal context for the derivation of two well-known results, namely Madey's theorem and quasilinear diffusion equation. More importantly, this approach reveals a formal procedure for the extension to higher order perturbations, related to stronger wave fields, and provides novel analytical results. Therefore, it is shown that third order perturbation theory for the single particle motion allows for the calculation of phase-averaged quantities and the derivation of a diffusion equation with fourth order accuracy.

A simplified Hamiltonian system, describing

resonant wave-particle interactions, has been considered, in order to clearly introduce, without loss of generality, the consequences of considering perturbations beyond the quasilinear approximation. However, there is no inherent restriction of this approach, preventing its applicability to more complex cases, as well as Hamiltonian systems occurring in other areas of physics. It is expected that these results can bring to light new physical aspects beyond the limits of validity of the traditional quasilinear theory

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Appendix A: Simplified Hamiltonian system

By utilizing the generalized momentum \mathbf{P} and the vector potentials of the main magnetic field \mathbf{A}_0 and the wave field $\tilde{\mathbf{A}}$

$$\mathbf{P} = \mathbf{p} + q\mathbf{A}, \quad \mathbf{A} = \mathbf{A}_0 + \tilde{\mathbf{A}} \quad (\text{A.1})$$

$$\mathbf{A}_0 = \mathbf{e}_y B_0 x,$$

$$\tilde{\mathbf{A}} = \sum_i \frac{E_0^{(i)} c}{\omega_i} \Im \left[\mathbf{f}^{(i)} F^{(i)}(z) e^{i(k_{\perp, i} x + k_{\parallel, i} z - \omega_i t)} \right] \quad (\text{A.2})$$

the Hamiltonian of the system can be written in the following form

$$H(x, z, \mathbf{P}, t) = m_0 c^2 \gamma, \quad \gamma = \sqrt{1 + \frac{1}{m_0^2 c^2} (\mathbf{P} - q\mathbf{A})^2} \quad (\text{A.3})$$

where e, m_0, c and \mathbf{p} are the particle charge, the rest mass, the speed of light and the kinematic momentum, respectively. By expanding up to linear order with respect to the perturbed vector potential, we have

$$H \approx m_0 c^2 \gamma_0 - q\mathbf{v} \cdot \tilde{\mathbf{A}} \quad (\text{A.4})$$

where

$$\gamma_0 = \sqrt{1 + \frac{1}{m_0^2 c^2} (\mathbf{P} - q\mathbf{A}_0)^2}, \quad \mathbf{v} = \frac{1}{m_0 \gamma_0} (\mathbf{P} - q\mathbf{A}_0) \quad (\text{A.5})$$

The ratio of the omitted quadratic term over the retained linear term is of the order $v_E^{(i)}/v_{\perp} \sim cE_0^{(i)}/(v_{\perp} B_0) \ll 1$ where $v_E^{(i)} \equiv eE_0^{(i)}/(m_0 \omega_i)$. Next we apply a canonical transformation $(x, y, z, P_x, P_y, P_z) \mapsto (\phi, Y, Z, J_{\perp}, P_Y, P_Z)$ with the generating function

$$F_3(P_x, P_y, P_z, \phi, Y, Z) = \frac{1}{m_0 \omega_{c0}} \times \left(\frac{P_x^2 \tan \phi}{2} - P_x P_y \right) - P_y Y - P_z Z \quad (\text{A.6})$$

defining, in the place of x and P_x the new pair of canonical conjugate variables

$$\phi = \tan^{-1} \left(\frac{P_y - m_0 \omega_{c0} x}{P_x} \right), \quad J_{\perp} = -\frac{P_x^2}{2m_0 \omega_{c0} \cos^2 \phi} \quad (\text{A.7})$$

where $\omega_{c0} = qB/m_0$ is the nonrelativistic cyclotron frequency, which is negative for electrons (which is the case considered in the following). Therefore, we can obtain

$$\mathbf{k}_i \cdot \mathbf{r} = k_{\parallel, i} Z + \alpha_i \sin \phi + \psi_0^{(i)}, \quad \alpha_i \equiv -\frac{k_{\perp, i} v_{\perp}}{\omega_c},$$

$$\psi_0^{(i)} = \frac{k_{\perp, i} P_Y}{m_0 \omega_{c0}} = \text{const} \quad (\text{A.8})$$

with

$$v_{\perp} = \frac{\sqrt{-2m_0 \omega_{c0} J_{\perp}}}{m_0 \gamma_0}, \quad \omega_c \equiv \frac{\omega_{c0}}{\gamma_0},$$

$$\gamma_0 = \sqrt{1 + \frac{1}{m_0^2 c^2} (P_Z^2 - 2m_0 \omega_{c0} J_{\perp})} \quad (\text{A.9})$$

For the Hamiltonian, as expressed in the new variable set, the variable Y is cyclic and consequently its conjugate momentum P_Y (and ψ_0) is conserved. Assuming that the carrier frequencies of all wavepackets are close to the k_0 -th harmonic of the gyrofrequency $\omega_i \approx k_0 |\omega_c|$, the second term of the Hamiltonian (A.4) can be expanded into Bessel functions and by keeping only the resonant terms we obtain

$$H = m_0 c^2 \gamma_0 - \sum_i \frac{e E_0^{(i)}}{\omega_i} F^{(i)}(z) \times \Im m \left[\left(v_{\parallel} f_{\parallel, i} J_{k_0}(\alpha_i) + \frac{v_{\perp}}{2} (J_{k_0-1}(\alpha_i) f_i^- + J_{k_0+1}(\alpha_i) f_i^+) \right) e^{i(k_{\parallel, i} z + k_0 \phi - \omega_i t + \psi_0^{(i)})} \right] \quad (\text{A.10})$$

Furthermore, assuming the particles to be weakly relativistic ($v/c \ll 1$), and since $\alpha_i \sim v_{\perp}/c$ we can use take into account only the lowest terms from the small argument expansion of the Bessel functions, so that

$$H = m_0 c^2 \gamma_0 - \frac{J_{\perp}^{k_0/2}}{2} \sum_i w_E^{(i)} F^{(i)}(z) e^{i(k_0 \phi + k_{\parallel, i} z - \omega_i t)} + c.c. \quad (\text{A.11})$$

where

$$\omega_E^{(i)} = \frac{v_E^{(i)} |f_i^-| |2m_0 \omega_{c0}|^{k_0/2}}{2(k_0 - 1)!} \left(\frac{k_{\perp, i}}{2m_0 \omega_{c0}} \right)^{k_0-1} e^{i(\psi_0^{(i)} + \arg f_i^- - \pi/2)} \quad (\text{A.12})$$

Also, expanding γ_0 up to fourth order with respect to v/c (and keeping only terms containing J_{\perp}) we obtain the Hamiltonian

$$H = -\omega_{c0} \left(1 - \frac{v_{\parallel}^2}{2c^2} \right) J_{\perp} - \frac{k_0 \omega_{c0}^2}{2m_0 c^2} J_{\perp}^2 - \frac{J_{\perp}^{k_0/2}}{2} \sum_i w_E^{(i)} F^{(i)}(z) e^{i(k_0 \phi + k_{\parallel, i} z - \omega_i t)} + c.c. \quad (\text{A.13})$$

and using the canonical transformation with generating function $F_2 = [\phi + \omega_{c0}(1 - v_{\parallel}^2/2c^2)t]J$ we obtain

$$H = -\frac{k_0 \omega_{c0}^2}{2m_0 c^2} J^2 - \frac{J^{k_0/2}}{2} \sum_i w_E^{(i)} F^{(i)}(z) e^{i(k_0 \theta + k_{\parallel, i} z - \omega_i t)} + c.c. \quad (\text{A.14})$$

with $J = J_{\perp}$ and $\theta = \phi + \omega_{c0}(1 - v_{\parallel}^2/2c^2)t$.

Based on physical arguments, [3] in certain cases we can consider that the canonical momentum $P_z = v_{\parallel} m_0$ is constant so that we can replace the variable z by $v_{\parallel} t$. Thus, we introduce the new time variable $|v_{\parallel}|t$ and use the scaling transformation

$$\theta \longmapsto -\theta, \quad J \longmapsto sJ \quad (\text{A.15})$$

with $s = 2|v_{\parallel}|m_0 c^2 / (k_0 \omega_{c0}^2)$ in order to obtain the reduced Hamiltonian

$$H = J^2 - E \frac{(2J)^{k_0/2}}{2} e^{ik_0 \theta} \sum_i w_E^{(i)} F^{(i)}(t) e^{i(k_0 \theta - \Omega_i t)} + c.c. \quad (\text{A.16})$$

where

$$E = \frac{1}{2v_{\parallel}} \left(\frac{m_0 c^2 v_{\parallel}}{k_0 \omega_{c0}^2} \right)^{k_0/2-1}, \quad \Omega_i = k_{\parallel, i} - \frac{k_0 \omega_{c0}}{v_{\parallel}} \left(1 - \frac{v_{\parallel}^2}{2c^2} \right) - \frac{\omega_i}{v_{\parallel}} \quad (\text{A.17})$$

Appendix B: Deprit's perturbation series

transformation T along with the Lie generator w are

According to the method of Deprit [34, 35], the old Hamiltonian H , the new Hamiltonian K and the

expanded in power series of ϵ (Appendix B).

$$\begin{aligned} H(\mathbf{z}, t, \epsilon) &= \sum_{n=0}^{\infty} \epsilon^n H_n(\mathbf{z}, t), \\ K(\mathbf{z}, t, \epsilon) &= \sum_{n=0}^{\infty} \epsilon^n K_n(\mathbf{z}, t), \\ T(\mathbf{z}, t, \epsilon) &= \sum_{n=0}^{\infty} \epsilon^n T_n(\mathbf{z}, t), \\ w(\mathbf{z}, t, \epsilon) &= \sum_{n=0}^{\infty} \epsilon^n w_{n+1}(\mathbf{z}, t) \end{aligned} \quad (\text{B.1})$$

where the expansion of w has been appropriately chosen in order to generate the identity transformation $T_0 = I$ to the lowest order. The transformations T and T^{-1} which will be used in the following are given below, through fourth order:

$$T_0 = I, \quad (\text{B.2a})$$

$$T_1 = -L_1, \quad (\text{B.2b})$$

$$T_2 = -\frac{1}{2}L_2 + \frac{1}{2}L_1^2, \quad (\text{B.2c})$$

$$T_3 = -\frac{1}{3}L_3 + \frac{1}{6}L_2L_1 + \frac{1}{3}L_1L_2 - \frac{1}{6}L_1^3, \quad (\text{B.2d})$$

$$\begin{aligned} T_4 = & -\frac{L_4}{4} + \frac{L_3L_1}{12} + \frac{L_2^2}{8} + \frac{L_1L_3}{4} - \frac{L_2L_1^2}{24} \\ & - \frac{1}{12}L_1L_2L_1 - \frac{1}{8}L_1^2L_2 + \frac{1}{24}L_1^4; \end{aligned} \quad (\text{B.2e})$$

$$T_0^{-1} = I \quad (\text{B.3a})$$

$$T_1^{-1} = L_1 \quad (\text{B.3b})$$

$$T_2^{-1} = \frac{1}{2}L_2 + \frac{1}{2}L_1^2 \quad (\text{B.3c})$$

$$T_3^{-1} = \frac{1}{3}L_3 + \frac{1}{6}L_1L_2 + \frac{1}{3}L_2L_1 + \frac{1}{6}L_1^3 \quad (\text{B.3d})$$

$$\begin{aligned} T_4^{-1} = & \frac{L_4}{4} + \frac{1}{12}L_1L_3 + \frac{L_2^2}{8} + \frac{1}{4}L_3L_1 + \frac{L_1^2L_2}{24} \\ & + \frac{1}{12}L_1L_2L_1 + \frac{1}{8}L_2L_1^2 + \frac{1}{24}L_1^4 \end{aligned} \quad (\text{B.3e})$$

The equations providing the Lie generator w and the new Hamiltonian K , to fourth order are:

$$K_0 = H_0 \quad (\text{B.4})$$

$$\frac{\partial w_1}{\partial t} + [w_1, H_0] = K_1 - H_1 \quad (\text{B.5})$$

$$\begin{aligned} \frac{\partial w_2}{\partial t} + [w_2, H_0] &= 2(K_2 - H_2) \\ &- L_1(K_1 + H_1) \end{aligned} \quad (\text{B.6})$$

$$\begin{aligned} \frac{\partial w_3}{\partial t} + [w_3, H_0] &= 3(K_3 - H_3) - L_1(K_2 \\ &+ 2H_2) - L_2(K_1 + \frac{1}{2}H_1) - \frac{1}{2}L_1^2H_1 \end{aligned} \quad (\text{B.7})$$

$$\begin{aligned} \frac{\partial w_4}{\partial t} + [w_4, H_0] &= 4(K_4 - H_4) - L_1(K_3 + 3H_3) \\ &- L_2(K_2 + H_2) - L_1^2H_2 - L_3(K_1 + \frac{1}{3}H_1) \\ &- \frac{1}{6}(L_1L_2 + 2L_2L_1 + L_1^3)H_1 \end{aligned} \quad (\text{B.8})$$

By selecting the arbitrary functions K_n so that the θ -independent part of the r.h.s. is eliminated, one can show that these equations can be written in the general form

$$\frac{\partial w_n}{\partial t} + [w_n, H_0] = \sum_{m \neq 0} P_{n,m}(J, t) e^{imk_0\theta} \quad (\text{B.9})$$

where n is the order of perturbation and m is the harmonic number of the corresponding term. Their solutions are given as

$$w_n = \sum_{m \neq 0} F_{n,m} e^{imk_0\theta},$$

$$F_{n,m} = \int_{t_0}^t P_{n,m}(J, s) e^{imk_0\omega_0(s-t)} ds \quad (\text{B.10})$$

with $\omega_0 = \partial H_0 / \partial J$.

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