

Recursive approach for non-Markovian time-convolutionless master equationsG. Gasbarri^{1,*} and L. Ferialdi^{1,2,†}¹*Department of Physics, University of Trieste, Strada costiera 11, 34151 Trieste, Italy*²*INFN, Sezione di Trieste, Strada costiera 11, 34151 Trieste, Italy*

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We consider a general open system dynamics and we provide a recursive method to derive the associated non-Markovian master equation in a perturbative series. The approach relies on a momenta expansion of the open system evolution. Unlike previous perturbative approaches of this kind, the method presented in this paper provides a recursive definition of each perturbative term. Furthermore, we give an intuitive diagrammatic description of each term of the series, which provides a useful analytical tool to build them and to derive their structure in terms of commutators and anticommutators. We eventually apply our formalism to the evolution of the observables of the reduced system, by showing how the method can be applied to the adjoint master equation, and by developing a diagrammatic description of the associated series.

DOI: [10.1103/PhysRevA.97.022114](https://doi.org/10.1103/PhysRevA.97.022114)**I. INTRODUCTION**

The investigation of open systems dynamics in quantum physics has constantly grown in recent years, pushed by the interest in developing new quantum technologies [1]. Open quantum systems are generally described by non-Markovian dynamics, which account for the memory of the interaction between the system and the environment surrounding it. Unlike approximated Markovian dynamics that are always described by a master equation of the Lindblad type [2], non-Markovian dynamics in general cannot be recast in a unique explicit structure. There is a vast literature on the formal investigation of non-Markovian dynamics [3–8], in this paper we are interested in investigating those dynamics that are derived from underlying physical models, i.e., obtained by tracing out the degrees of freedom of a physical environment, provided that the initial state is factorized. Recently, a microscopic derivation has been provided for a specific class of non-Markovian maps [9], namely those describing a system interacting with a bosonic bath that is completely characterized by its two-point correlation function. Notorious examples that fall in this category are, e.g., the non-Markovian Brownian motion [10,11] and the spin-boson model [12,13]. Moreover, it has been shown that if one considers a system described by a bosonic quadratic Hamiltonian, it is possible to derive analytically the family of Gaussian, non-Markovian, completely positive master equations [14]. However, there are many physical systems that do not fall into the Gaussian ansatz. Interesting examples are state transfer in quantum information [15], Brownian motion with nonlinear coupling [16], the donor-acceptor model [17], widely used in quantum biology, driven spin chains [18] that cover a crucial role in condensed matter, and coupled cavities in cQED [19].

General non-Markovian dynamics can be formally encoded in the Nakajima-Zwanzig master equation [20,21] that displays an integral term accounting for memory effects. This class of master equation has been thoroughly investigated [5,22], and only recently, a characterization of physically admissible integrodifferential master equations has been provided, based on a generalization of classical semi-Markov processes [7]. Since integrodifferential equations are hard to treat, a more handful tool to investigate open quantum systems are time-convolutionless (TCL) master equations [20]. We underline that the solution of a TCL master equation always satisfies a Nakajima-Zwanzig master equation [4]. Closed expressions for TCL master equations have been obtained for few analytically solvable models [10,23,24] whose dynamics fall into the family of Gaussian non-Markovian maps [9,14]. In order to derive TCL master equations in more general frameworks, a number of perturbative approaches have been developed. Among these we mention the functional integral formalism [25], the methods by Kubo and van Kampen [26–28] (originally developed in the context linear stochastic differential equations), projection operator techniques [29], hierarchical equations of motion [30], effective modes [31], stochastic Liouville–von Neumann [32], and multiple-time correlation functions [33] (for a review on the topic see [8]). These approaches allowed us to improve the theoretical description of non-Markovian dynamics, but they all suffer two drawbacks. First, in order to obtain the series up to the n th perturbative order, one has to apply repeatedly the whole formalism. This makes the derivation of higher order terms unwieldy. Second, these methods do not make clear evidence of the mathematical structure of the perturbative series in terms of commutators and anticommutators.

In this paper we tackle these issues by providing a perturbative technique that allows us to derive the master equation of a general open system in terms of a perturbative series, with the only assumption that the system and the bath are initially uncorrelated. Unlike all perturbative approaches present in the literature, our method allows us to characterize the structure

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of each expansion term through an explicit recursive formula. Such an iterative structure makes their derivation simpler. We further provide an intuitive diagrammatic description of each term of the series, which provides a useful analytical tool to build them and to infer their structure in terms of commutators and anticommutators.

We eventually apply our formalism to the evolution of the observables of the reduced system. We show how the method can be applied to the adjoint master equation, and we develop a diagrammatic description of the associated series.

II. NON-MARKOVIAN MAP AND MASTER EQUATION

We consider a system (\mathcal{S}) interacting with a generic environment (\mathcal{E}). The evolution of the open system density matrix $\hat{\rho}_{\mathcal{S}\mathcal{E}}$ in the interaction picture is described by the von Neumann equation ($\hbar = 1$)

$$i \frac{\partial \hat{\rho}_{\mathcal{S}\mathcal{E}}(t)}{\partial t} = [\hat{V}_t, \hat{\rho}_{\mathcal{S}\mathcal{E}}(t)], \quad (1)$$

where \hat{V}_t is a generic interaction Hamiltonian between the system and the environment. In order to simplify the calculations to come, we assume \hat{V}_t to be factorized, i.e.,

$$\hat{V}_t = \hat{A}_t \hat{\phi}_t, \quad (2)$$

where \hat{A}_t and $\hat{\phi}_t$, respectively, are Hermitian system and environment operators. We however stress that the formalism presented holds for the most general $\hat{V}_t = \sum_i \hat{A}_t^i \hat{\phi}_t^i$. It is convenient to introduce the left-right formalism denoting by a subscript L (R) the operators acting on $\hat{\rho}$ from the left (right) [34]. The dynamical map Φ_t for the open system is obtained by formally solving Eq. (1):

$$\Phi_t \hat{\rho}_{\mathcal{S}\mathcal{E}} = \mathcal{T} \left(e^{-i \int_0^t d\tau (\hat{V}_{\tau L} - \hat{V}_{\tau R})} \right) \hat{\rho}_{\mathcal{S}\mathcal{E}}, \quad (3)$$

where $\mathcal{T}(\cdot)$ denotes the time ordering operator. Since we are interested on the effective evolution of the system \mathcal{S} , we aim for the reduced dynamical map \mathcal{M}_t that evolves the initial state of the system $\hat{\rho}_{\mathcal{S}}$ to the state $\hat{\rho}_{\mathcal{S}}(t)$ at time t [$\hat{\rho}_{\mathcal{S}}(t) \equiv \mathcal{M}_t \hat{\rho}_{\mathcal{S}}$]. This is obtained by tracing out the environmental degrees of freedom from Φ_t . In order to do so, we assume that the open system initial state is factorized: $\hat{\rho}_{\mathcal{S}\mathcal{E}} = \hat{\rho}_{\mathcal{S}} \otimes \hat{\rho}_{\mathcal{E}}$. The dynamical map $\mathcal{M}_t(\cdot)$ is then given by

$$\mathcal{M}_t \hat{\rho}_{\mathcal{S}} = \text{Tr}_{\mathcal{E}} \left[\mathcal{T} \left(e^{-i \int_0^t d\tau V_{\tau}^-} \right) \hat{\rho}_{\mathcal{S}} \otimes \hat{\rho}_{\mathcal{E}} \right], \quad (4)$$

where $\text{Tr}_{\mathcal{E}}$ denotes the partial trace over the environment, and we have defined

$$V_{\tau}^- \equiv (\hat{V}_{\tau L} - \hat{V}_{\tau R}) \quad (5)$$

(for convenience superoperators are not denoted by a hat). We observe that Eq. (4) guarantees the complete positivity of the map, since it can be understood as the Kraus-Stinespring decomposition of \mathcal{M}_t [35]. When the environment is completely characterized by its two-point correlation function $\text{Tr}_{\mathcal{E}}[\hat{\phi}_t^i \hat{\phi}_s^j \hat{\rho}_{\mathcal{E}}]$, the trace can be performed exactly and one obtains a closed Gaussian form for \mathcal{M}_t [9]. If in addition the Hamiltonian is at most quadratic and the system operators obey linear Heisenberg equations of motion, one can exploit Wick's theorem and derive the exact master equation [14]. Unluckily, in the general case we are considering, such techniques cannot

be exploited, and one needs to tackle the problem from another perspective.

The formalism we use is based on an expansion over the map momenta that is close to the cumulant expansion introduced by van Kampen [27]. The advantage of our formalism is that it allows us to construct recursively the master equation, while this is not possible with the van Kampen approach [36]. Since the derivation is rather involved, we refer the reader to the Appendixes for mathematical details. We start by expanding Eq. (4) in Dyson's series, obtaining

$$\mathcal{M}_t = 1 + \sum_{n=1}^{\infty} (-i)^n \mu_{n,t}, \quad (6)$$

where $\mu_{n,t}$ are the integrated momenta

$$\mu_{n,t} \hat{\rho}_{\mathcal{S}} = \frac{1}{n!} \text{Tr}_{\mathcal{E}} \left[\mathcal{T} \left(\int_0^t d\tau V_{\tau}^- \right)^n \hat{\rho}_{\mathcal{S}} \otimes \hat{\rho}_{\mathcal{E}} \right]. \quad (7)$$

We observe that the subscript n of μ denotes the power of the superoperator V^- , i.e., the number of operators \hat{A} and $\hat{\phi}$ displayed by momentum. The subscript t , denoting time dependence, will be dropped in the remainder of this paper for compactness of notation. Doing so, we implicitly assume that the momenta are evaluated at time t , unless otherwise explicitly stated. In order to make this formula more transparent, we need to make explicit the dependence of μ_n over the system operators \hat{A} and on the environment n -point correlation functions. It is convenient to introduce a new pair of superoperators: $A^+ \equiv \hat{A}_L + \hat{A}_R$ and $A^- \equiv \hat{A}_L - \hat{A}_R$ (analogous definitions hold for environment operators $\hat{\phi}$). This notation is particularly convenient because one can associate to A^+ an anticommutator and to A^- a commutator. It will then be immediately evident how these building blocks contribute to the structure of the master equation. We consider the definition (7) of μ_n and we replace Eqs. (2) and (5) in it. The result in terms of A^{\pm} and ϕ^{\pm} is

$$\mu_n \hat{\rho}_{\mathcal{S}} = \frac{1}{n! 2^n} \text{Tr}_{\mathcal{E}} \left[\mathcal{T} \left(\int_0^t d\tau (A_{\tau}^+ \phi_{\tau}^- + A_{\tau}^- \phi_{\tau}^+) \right)^n \hat{\rho}_{\mathcal{S}} \otimes \hat{\rho}_{\mathcal{E}} \right]. \quad (8)$$

It is important to observe that a + superoperator for the systems is always associated to a - superoperator for the environment, and vice versa. We will shortly show that this "sign conservation rule" covers a crucial role for trace preservation of the map. We now exploit the binomial theorem, and we make explicit the time ordering simply by conditioning the time integrals. After some manipulation one finds that the result of this procedure is the mixing of the \pm superoperators (see Appendix A):

$$\mu_n = \int_0^t d\bar{\tau}_n \sum_{j=1}^n \sum_{\mathcal{P}_j} A_{\tau_1}^- \cdots A_{\tau_n}^{k_n} D_{\tau_1 \cdots \tau_n}^{+\cdots \bar{k}_n}, \quad (9)$$

where $\int_0^t d\bar{\tau}_n = \prod_{i=1}^n \int_0^t d\tau_i$, and \mathcal{P}_j denotes all the permutations of the indexes $k_i \in \{+, -\}$, with $k_1 = -$ and such that there is a j number of minus superoperators. We have also introduced the bath "ordered correlation functions," defined by

$$D_{\tau_1 \cdots \tau_n}^{+\cdots \bar{k}_n} \equiv \frac{1}{2^n} \text{Tr}_{\mathcal{E}} \left[\phi_{\tau_1}^+ \theta_{\tau_1 \tau_2} \phi_{\tau_2}^{\bar{k}_2} \cdots \theta_{\tau_{n-1} \tau_n} \phi_{\tau_n}^{\bar{k}_n} \hat{\rho}_{\mathcal{E}} \right], \quad (10)$$

where θ_{τ_i, τ_j} is one for $\tau_i > \tau_j$, zero otherwise, and provides the ordering both of the operators $\hat{\phi}$ in D , of the operators \hat{A} in Eq. (9), by conditioning the integrals limits. The 2^n prefactor represents the number of permutations of the operators $\hat{\phi}$ contained in D (provided by commutators ϕ^- and anticommutators ϕ^+). Moreover, $\bar{k}_i \equiv -k_i$ guarantees the sign conservation rule. We now exploit the cyclicity property of the trace that implies $\text{Tr}_{\mathcal{E}}[\phi^- \hat{O} \hat{\rho}_{\mathcal{E}}] = 0$ for any operator \hat{O} . According to the sign conservation rule, the contributions where A^+ is the first superoperator on the left are suppressed. As a consequence, the first system superoperator on the left of μ_n is always A^- . This is an important feature because it guarantees that the map is trace preserving (indeed $\text{Tr}_S[A^- \hat{O} \hat{\rho}_S] = 0$).

Equation (9) shows that the momenta μ_n are composed by the sum of all the permutations of the products of n superoperators A^{\pm} , where the first term on the left is A^- , and the associated environment correlation function is obtained by the sign conservation rule. By replacing Eq. (9) in Eq. (6) we obtain the explicit expression for the perturbative series of the map \mathcal{M}_t . We observe that, unlike the Gaussian case [9], one cannot sum the series and is left with the formal expression (6). However, if we consider a Gaussian bath, we can decompose higher order correlation functions in (9) by means of the Isserlis' theorem [37], and recover known results.

The dependence of the momenta on the system operators and the environment correlation function is not only important for the map structure, but plays also a relevant role for the derivation of the master equation. We look for a time local master equation of the type

$$\partial_t \hat{\rho}_S(t) = \mathbb{L}_t \hat{\rho}_S(t), \quad (11)$$

where the generator \mathbb{L}_t can be formally written as follows:

$$\mathbb{L}_t = \partial_t(\mathcal{M}_t) \mathcal{M}_t^{-1}. \quad (12)$$

Exploiting the identity $(1+x)^{-1} = \sum_{n=0}^{\infty} (-x)^n$ (under the assumption that $\|\mathcal{M}_t - 1\| < 1$) one can invert Eq. (6) obtaining

$$\mathcal{M}_t^{-1} = \sum_{n=0}^{\infty} (-i)^n M_n, \quad (13)$$

where the superoperators M_n are recursively defined as follows:

$$M_n = - \sum_{k=1}^n \mu_k M_{n-k} 0, \quad (14)$$

with $M_0 = 1$ (see Appendix B). One then sees that M_n is the sum of μ_n plus the products of momenta with order lower than n . Accordingly, M_n and μ_n contain the same number n of operators, while they differ for how the bath operators are clustered by $\text{Tr}_{\mathcal{E}}$ (and ordered by \mathcal{T}). Indeed, in μ_n the operators $\hat{\phi}$ are grouped together under the trace in Eq. (10), while in M_n one needs to consider all possible clusterings of n elements. This fact can be seen by replacing Eq. (9) in Eq. (14). For example, for $n = 2$ one finds

$$\mu_2 = \int_0^t d\bar{\tau}_2 A_{\tau_1}^- A_{\tau_2}^- D_{\tau_1 \tau_2}^{++} + A_{\tau_1}^- A_{\tau_2}^+ D_{\tau_1 \tau_2}^{+-},$$

$$M_2 = \int_0^t d\bar{\tau}_2 A_{\tau_1}^- A_{\tau_2}^- (D_{\tau_1}^+ D_{\tau_2}^+ - D_{\tau_1 \tau_2}^{++}) - A_{\tau_1}^- A_{\tau_2}^+ D_{\tau_1 \tau_2}^{+-}.$$

According to Eq. (10), while in μ_2 both bath operators ϕ are clustered together (e.g., in $D_{\tau_1 \tau_2}^{++}$), M_2 displays a term with a different clustering ($D_{\tau_1}^+ D_{\tau_2}^+$).

Replacing Eqs. (6) and (13) in Eq. (12), and after some calculations one can find (see Appendix C)

$$\mathbb{L}_t = \sum_{n=1}^{\infty} (-i)^n L_n, \quad (15)$$

with

$$L_n = \dot{\mu}_n - \sum_{k=1}^{n-1} L_{n-k} \mu_k, \quad (16)$$

and $L_0 = 0$, where we denoted the derivative with respect to time t with a dot. Equation (16) is the recursive law that allows us to build iteratively of each term of the expansion (15). Each L_n is the sum of all the possible combinations of A^{\pm} (provided that the first on the left is always A^-), suitably ordered and clustered. These terms are associated with peculiar combinations of ordered correlation functions, whose construction is elegantly described by the recursion (16). The first two terms of the series (15) can be easily obtained starting from the definition of the momenta (9):

$$L_1 = A_{\tau}^- D_{\tau}^+, \quad (17)$$

$$L_2 = A_{\tau}^- \int_0^t d\tau_1 [A_{\tau_1}^+ D_{\tau \tau_1}^{+-} + A_{\tau_1}^- (D_{\tau \tau_1}^{++} - D_{\tau}^+ D_{\tau_1}^+)]. \quad (18)$$

However, the structure of the third term is already quite complicated, and higher order terms are rather involved to compute. In order to ease the computation of the generic L_n , we provide here an intuitive diagrammatic description of how they can be built.

III. DIAGRAMMATICS

We introduce the following notation:

$$\text{---}\bigcirc\text{---} = A_{\tau}^+ \phi_{\tau}^-, \quad \text{---}\bullet\text{---} = A_{\tau}^- \phi_{\tau}^+, \quad (19)$$

and we represent the trace over the environmental degrees of freedom as linking the circles in the following way:

$$\underbrace{\text{---}\bigcirc\text{---}\text{---}\bigcirc\text{---}\text{---}\bigcirc\text{---}}_n = \int_0^t d\bar{\tau}_n A_{\tau_1}^- \cdots A_{\tau_n}^{\pm} D_{\tau_1, \dots, \tau_n}^{+\dots\mp}, \quad (20)$$

where \bigcirc denotes that in any position one can put either \bigcirc or \bullet , and D is the bath ordered correlation function defined in Eq. (10). We call the left-hand side of Eq. (20) “ n th order connected diagram,” while a “ n th order nonconnected diagram” is obtained by removing at least one connection (line connecting circles) from the respective connected diagram. Note that when we trace only a single symbol (19), this simply results in dropping the side lines, i.e., the first-order diagram reads

$$\bullet = \int_0^t d\tau_1 A_{\tau_1}^- D_{\tau_1}^+. \quad (21)$$

We stress that the role of the bath ordered correlation functions D is to link together the circles, clustering and ordering them in a specific way. Accordingly, one has that, e.g.,

$$\bullet-\bullet-\bullet = \int_0^t d\bar{\tau}_3 A_{\tau_1}^- A_{\tau_2}^- A_{\tau_3}^- D_{\tau_1 \tau_2 \tau_3}^{+++} \quad (22)$$

differs from

$$\bullet-\bullet-\bullet = \int_0^t d\bar{\tau}_3 A_{\tau_1}^- A_{\tau_2}^- A_{\tau_3}^- D_{\tau_1 \tau_2}^{++} D_{\tau_3}^+ \quad (23)$$

for how the bath operators are clustered in D . Moreover, we observe that with this notation, the fact that the first superoperator on the left is always A^- is rephrased as follows: the diagrams whose first circle on the left is white are null, i.e.,

$$\circ-\bullet-\bullet-\dots-\bullet = 0. \quad (24)$$

Having introduced the basic elements of our diagrammatics, we can move to its application. We start from the map \mathcal{M}_t (6), which is defined in terms of the momenta μ_n of Eq. (9). With the diagrams introduced above, one finds that the momentum μ_n is the sum of all possible n th order connected diagrams, i.e.,

$$\mu_n = \sum_{j=1}^n \sum_{\mathcal{P}_j} \underbrace{\bullet-\bullet-\dots-\bullet}_n, \quad (25)$$

where now \mathcal{P}_j denotes all permutations of black and white circles, such that there is a j number of black ones.

In order to build the term L_n of the generator, we denote the derivative with respect to t with a dot over a circle. With this notation, one finds that the diagrammatic version of Eqs. (17) and (18) reads

$$L_1 = \dot{\bullet}, \quad (26)$$

$$L_2 = \dot{\bullet}-\bullet-\bullet + \dot{\bullet}-\bullet-\circ. \quad (27)$$

The procedure to build Eq. (16) for a generic n with this diagrammatics is the following (we show the case $n = 3$ as explicit example):

(1) Write the n th order connected diagram composed by n black circles and put a dot on the first circle:

$$\dot{\bullet}-\bullet-\bullet. \quad (28)$$

(2) Remove a number $p \leq n - 1$ of connections from the previous connected diagram, in all possible ways. Multiply the diagrams obtained at each step by $(-1)^p$. Repeat for all p , until all $n - 1$ connections are removed, i.e., until all black circles are disconnected:

$$\dot{\bullet}-\bullet-\bullet - (\dot{\bullet}-\bullet-\bullet + \dot{\bullet}-\bullet-\bullet) + \dot{\bullet}-\bullet-\bullet. \quad (29)$$

(3) Turn a number $p \leq n - 1$ of black circles of the diagrams obtained so far into white, in all possible ways, and remembering the rule (24). Repeat for all p , until all circles

(but the first) are white:

$$\begin{aligned} L_3 = & \dot{\bullet}-\bullet-\bullet - \dot{\bullet}-\bullet-\bullet - \dot{\bullet}-\bullet-\bullet + \dot{\bullet}-\bullet-\bullet \\ & + \dot{\bullet}-\bullet-\circ - \dot{\bullet}-\bullet-\circ \\ & + \dot{\bullet}-\circ-\bullet - \dot{\bullet}-\circ-\bullet \\ & + \dot{\bullet}-\circ-\circ. \end{aligned} \quad (30)$$

(4) Exploit Eqs. (20) and (21) to translate the diagram obtained in operatorial form. Equation (30) reads

$$\begin{aligned} L_3 = & \int_0^t d\bar{\tau}_2 [A_t^- A_{\tau_1}^- A_{\tau_2}^- (D_{t \tau_1 \tau_2}^{+++} - D_t^+ D_{\tau_1 \tau_2}^{++}) \\ & - D_{t \tau_1}^{++} D_{\tau_2}^+ + D_t^+ D_{\tau_1}^+ D_{\tau_2}^+] \\ & + A_t^- A_{\tau_1}^- A_{\tau_2}^+ (D_{t \tau_1 \tau_2}^{+-+} - D_t^+ D_{\tau_1 \tau_2}^{+-}) \\ & + A_t^- A_{\tau_1}^+ A_{\tau_2}^- (D_{t \tau_1 \tau_2}^{-++} - D_{t \tau_1}^- D_{\tau_2}^+) \\ & + A_t^- A_{\tau_1}^+ A_{\tau_2}^+ D_{t \tau_1 \tau_2}^{+-+}], \end{aligned} \quad (31)$$

where the first two lines correspond to the first line of Eq. (30).

Equation (31) clearly provides insight on the mathematical structure of the master equation: L_n is the sum of all possible combinations of commutators (A^-) and anticommutators (A^+) of operators \hat{A} , multiplied by suitable combinations of bath ordered correlation functions D that encode the environment influence over the system. If one considers a Gaussian bath, one can decompose any even ordered correlation function (odd ones are zero) in terms of the two-point correlation function. Moreover, if the system Hamiltonian is bosonic and quadratic, one can exploit the operators algebra and reduce combinations of n nested (anti-)commutators to double (anti-)commutators, recovering the results of [14].

IV. ADJOINT MASTER EQUATION

We derive the adjoint master equation for the system observables that is a useful tool to investigate the evolution of physical quantities.

We define the adjoint interaction picture as the picture where the statistical operator $\hat{\rho}_{SE}$ evolves according to the free dynamics, and a generic operator \hat{O} evolves with the interaction Hamiltonian, i.e.,

$$\begin{aligned} \hat{O}_t &= \Phi_t^* \hat{O}, \\ \hat{\rho}_{SE}(t) &= e^{-i(H_S^- + H_E^-)t} \hat{\rho}_{SE}, \end{aligned} \quad (32)$$

where H_S^- and H_E^- , respectively, are the generators of the free dynamics of the system and of the environment [defined like in Eq. (5)], and Φ^* is the adjoint dynamical map defined by

$$\Phi_t^* \equiv \mathcal{T}(e^{i \int_0^t d\tau V_{\tau}^-}). \quad (33)$$

Since we are interested in the effective evolution of the system \mathcal{S} , we restrict our attention to operators of the type $\hat{O} = \hat{O}_S \otimes \hat{\mathbb{1}}_E$. Under this assumption, we obtain the reduced dynamical map \mathcal{M}_t^* by tracing out the environmental degrees of freedom from Φ_t^* . Under the further assumption that the initial state is factorized, the map \mathcal{M}_t^* is given by

$$\mathcal{M}_t^* \hat{O}_S = \text{Tr}_E [\hat{\rho}_{SE}(t) \mathcal{T}(e^{i \int_0^t d\tau V_{\tau}^-})] \hat{O}_S. \quad (34)$$

Retracing the steps that we have done deriving the master equation (11), we expand Eq. (34) in Dyson series, obtaining

$$\mathcal{M}_t^* = 1 + \sum_{n=1}^{\infty} i^n \tilde{\mu}_n, \quad (35)$$

where $\tilde{\mu}_n$ are the ‘‘adjoint integrated momenta’’

$$\tilde{\mu}_n = \int_0^t d\bar{\tau}_n \sum_{j=1}^n \sum_{\mathcal{P}_j} A_{\tau_1}^{k_1} \cdots A_{\tau_n}^{-} \tilde{D}_{\tau_1 \cdots \tau_n}^{\bar{k}_1 \cdots \bar{k}_n}, \quad (36)$$

and the ‘‘adjoint ordered correlation function’’ is defined by

$$\tilde{D}_{\tau_1 \cdots \tau_n}^{\bar{k}_1 \cdots \bar{k}_n} \equiv \frac{1}{2^n} \text{Tr}_{\mathcal{E}} [\hat{\rho}_{\mathcal{E}}(t) \phi_{\tau_1}^{\bar{k}_1} \theta_{\tau_1 \tau_2} \phi_{\tau_2}^{\bar{k}_2} \cdots \theta_{\tau_{n-1} \tau_n} \phi_{\tau_n}^{\bar{k}_n}]. \quad (37)$$

The adjoint momenta defined in Eq. (36) differ from the momenta in Eq. (9) only for the environmental contribution \tilde{D} . We now look for an adjoint master equation of the type

$$\partial_t \hat{O}_{S,t} = \mathbb{L}_t^* \hat{O}_{S,t}. \quad (38)$$

The similarities between Eqs. (6) and (35) allow us to compute the generator \mathbb{L}_t^* of the adjoint master equation that is described by the series

$$\mathbb{L}_t^* = \sum_{n=1}^{\infty} i^n \tilde{L}_n, \quad (39)$$

with

$$\tilde{L}_n = \dot{\tilde{\mu}}_n - \sum_{k=1}^{n-1} \tilde{L}_{n-k} \tilde{\mu}_k \quad (40)$$

and $\tilde{L}_0 = 0$. Because of the time dependence of the environmental state $\hat{\rho}_{\mathcal{E}}$, we cannot directly implement the diagrammatic scheme developed for the master equation (11). However, if we restrict our analysis to the steady states of the free evolution ($H_{\mathcal{E}}^- \hat{\rho}_{\mathcal{E}} = 0$), the environmental state $\hat{\rho}_{\mathcal{E}}$ will drop its time dependence. This allows us to define a diagrammatic expression for the adjoint master equation by exploiting the scheme previously developed, where rule (20) is replaced by

$$\underbrace{\bullet \cdots \bullet}_n = \int_0^t d\bar{\tau}_n A_{\tau_1}^- \cdots A_{\tau_n}^{\pm} \tilde{D}_{\tau_1 \cdots \tau_n}^{\bar{k}_1 \cdots \bar{k}_n}, \quad (41)$$

and rule (24) is replaced by

$$\bullet \cdots \bullet \cdots \circ = 0. \quad (42)$$

V. CONCLUSIONS

We have provided an iterative method that allows us to derive in a perturbative series the non-Markovian master equation for the density matrix, and its adjoint for the observables, of a generic open quantum system. The merit of our formalism is that the expansion terms are defined recursively, making their derivation easier compared to previous perturbative techniques. We have further given a diagrammatic description of the expansion terms that provides an intuitive analytical tool to build the perturbative series. Such a diagrammatics gives clear evidence of the mathematical structure of each term of the series, and explicitly shows that the environmental

effects on the dynamics are encoded on the action of a series of commutators and anticommutators of system operators, connected by the n -point environmental correlation functions.

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APPENDIX A: EXPLICIT DERIVATION OF THE EFFECTIVE MAP

In this Appendix we derive the explicit expression (9) for the ordered momenta μ_n . We consider Eq. (8) and exploiting the binomial theorem we rewrite the momentum μ_n as follows:

$$\begin{aligned} \mu_n \hat{\rho}_S &= \frac{1}{2^n n!} \sum_{k=0}^n \frac{n!}{(n-k)! k!} \text{Tr}_{\mathcal{E}} \left[\mathcal{T} \left(\int_0^t d\tau A_{\tau}^+ \phi_{\tau}^- \right)^k \right. \\ &\quad \left. \times \left(\int_0^t d\tau A_{\tau}^- \phi_{\tau}^+ \right)^{n-k} \hat{\rho}_S \otimes \hat{\rho}_{\mathcal{E}} \right]. \end{aligned} \quad (A1)$$

In order to make the time ordering explicit, we adopt the following strategy: we first resolve the time ordering for the couples $A^+ \phi^-$ and $A^- \phi^+$ independently, by conditioning the integrals with unit step functions θ :

$$\begin{aligned} \mu_n \hat{\rho}_S &= \frac{1}{2^n n!} \sum_{k=0}^n \frac{n!}{(n-k)! k!} \text{Tr}_{\mathcal{E}} \left[\mathcal{T} \left[\mathcal{T} \left(\prod_{j=k+1}^n A_{\tau_j}^+ \phi_{\tau_j}^- \right) \right. \right. \\ &\quad \left. \left. \times \mathcal{T} \left(\prod_{i=1}^k A_{\tau_i}^- \phi_{\tau_i}^+ \right) \right] \hat{\rho}_S \otimes \hat{\rho}_{\mathcal{E}} \right] \\ &= \frac{1}{2^n} \sum_{k=0}^n \text{Tr}_{\mathcal{E}} \left[\mathcal{T} \prod_{j=1}^{n-k} \int_0^t d\tau_j (A_{\tau_j}^+ \phi_{\tau_j}^-) \theta_{\tau_{j-1}, \tau_j} \right. \\ &\quad \left. \times \prod_{i=1}^k \int_0^t d\tau_i (A_{\tau_i}^- \phi_{\tau_i}^+) \theta_{\tau_{i-1}, \tau_i} \hat{\rho}_S \otimes \hat{\rho}_{\mathcal{E}} \right], \end{aligned} \quad (A2)$$

where $\tau_0 = 0$, and the arrow above the product denotes that the superoperators are ordered from the left to the right. One can see that this partial time ordering removes the factorial terms in the equation and orders in two independent blocks the integrals associated with the two couples of operators. The second step of our derivation is to order globally the two ‘‘pre-ordered’’ blocks. The result of this further ordering is the mixing of plus and minus superoperators in all the possible permutations, and the ordering of all integrals:

$$\begin{aligned} \mu_n \hat{\rho}_S &= \frac{1}{2^n} \int_0^t d\bar{\tau}_n \sum_{j=1}^n \sum_{\mathcal{P}_j} A_{\tau_1}^{k_1} \cdots A_{\tau_n}^{k_n} \\ &\quad \times \text{Tr}_{\mathcal{E}} [\phi_{\tau_1}^{\bar{k}_1} \theta_{\tau_1 \tau_2} \phi_{\tau_2}^{\bar{k}_2} \cdots \theta_{\tau_{n-1} \tau_n} \phi_{\tau_n}^{\bar{k}_n} \hat{\rho}_{\mathcal{E}}] \hat{\rho}_S, \end{aligned} \quad (A3)$$

where $d\bar{\tau}_n = \prod_{i=1}^n d\tau_i$, and \mathcal{P}_j denotes all the permutations of the indexes $k_i \in \{+, -\}$ such that there is a j number of

minus superoperators and $\bar{k}_i = -k_i$. Defining the environment ordered correlation function as

$$D_{\tau_1 \dots \tau_n}^{\bar{k}_1 \dots \bar{k}_n} \equiv \frac{1}{2^n} \text{Tr} \mathcal{E} [\phi_{\tau_1}^{\bar{k}_1} \theta_{\tau_1 \tau_2} \phi_{\tau_2}^{\bar{k}_2} \dots \theta_{\tau_{n-1} \tau_n} \phi_{\tau_n}^{\bar{k}_n} \hat{\rho} \mathcal{E}], \quad (\text{A4})$$

we recover Eq. (9):

$$\mu_n = \int_0^t d\bar{\tau}_n \sum_{j=1}^n \sum_{\mathcal{P}_j} A_{\tau_1}^{k_1} \dots A_{\tau_n}^{k_n} D_{\tau_1 \dots \tau_n}^{\bar{k}_1 \dots \bar{k}_n}. \quad (\text{A5})$$

APPENDIX B: RECURSIVE SERIES FOR THE INVERSE MAP \mathcal{M}_t^{-1}

The aim of this Appendix is to explicitly derive Eqs. (13) and (14), i.e., to express \mathcal{M}_t^{-1} as a power series of the interaction Hamiltonian \hat{V}_t of Eq. (2). We consider Eq. (7) and we formally invert it, in such a way that \mathcal{M}_t^{-1} (when it exists) can be written as

$$\mathcal{M}_t^{-1} = \left(1 + \sum_{n=1}^{\infty} (-i)^n \mu_n \right)^{-1}. \quad (\text{B1})$$

We define $\sum_{n=1}^{\infty} (-i)^n \mu_n \equiv x$ and, assuming that $|x| \leq 1$, we exploit the identity $(1+x)^{-1} = \sum_{n=0}^{\infty} (-x)^n$ obtaining

$$\mathcal{M}_t^{-1} = \sum_{n=0}^{\infty} \left(- \sum_{k=1}^{\infty} (-i)^k \mu_k \right)^n. \quad (\text{B2})$$

This equation can be rearranged by making explicit the power n :

$$\mathcal{M}_t^{-1} = \sum_{n=0}^{\infty} (-)^n \prod_{j=1}^n \left(\sum_{k_j=1}^{\infty} (-i)^{k_j} \mu_{k_j} \right). \quad (\text{B3})$$

From this expression it is clear that each term of the first series on the left is the product of series of momenta. Accordingly, such a series is not a power series of the interaction Hamiltonian \hat{V}_t because each of its terms contains all powers of momenta (and hence of the interaction \hat{V}_t). We rearrange Eq. (B3) by exploiting the Cauchy product of two series recursively (over the product of n series). The result is

$$\mathcal{M}_t^{-1} = \sum_{n=0}^{\infty} (-i)^n M_n, \quad (\text{B4})$$

with

$$M_n = \sum_{q=0}^n (-)^q \sum_{k_1 + \dots + k_q = n} \mu_{k_1} \dots \mu_{k_q}. \quad (\text{B5})$$

Equation (B4) is the correct series in power of the interaction we were looking for, as n denotes the power of the interaction Hamiltonian \hat{V}_t . The index q in Eq. (B5) instead denotes the number of partitions in which the operators are clustered (by the momenta μ_k). Since this expression for M_n is rather involved, we rewrite the terms for $n \geq 1$ as follows:

$$M_n = \sum_{k=1}^n \mu_k \sum_{q=0}^{n-k} (-)^{q+1} \sum_{k_1 + \dots + k_q = n-k} \mu_{k_1} \dots \mu_{k_q}. \quad (\text{B6})$$

One can easily check that the second series in this equation is simply Eq. (B5) for M_{n-k} . This leads us to the recursive formula of Eq. (14):

$$M_n = \sum_{k=1}^n \mu_k (-M_{n-k}), \quad \text{with } M_0 = 1. \quad (\text{B7})$$

APPENDIX C: RECURSIVE FORMULA FOR THE TIME LOCAL GENERATOR

In this Appendix we provide the technical details for the derivation of Eq. (16). We start from Eq. (12) and we substitute in it Eqs. (6) and (13), obtaining

$$\mathbb{L}_t = \sum_{n=1}^{\infty} \dot{\mu}_n \sum_{k=0}^{\infty} (-i)^{n+k} M_k. \quad (\text{C1})$$

Similarly to the previous Appendix, this one is also not a series in powers of the interaction Hamiltonian \hat{V}_t . In order to reach this goal, we exploit again the Cauchy product of two series, and we rearrange Eq. (C1) as follows:

$$\mathbb{L}_t = \sum_{n=1}^{\infty} (-i)^n L_n, \quad (\text{C2})$$

with

$$L_n = \sum_{k=1}^n \dot{\mu}_k M_{n-k}, \quad (\text{C3})$$

and M_n is determined by the recursive formula (B5). Equation (C3) is a useful expression of L_n for performing a numerical analysis. However, it is more elegant to derive a recursive relation that involves only L_n and the momenta μ . We do so by adopting the same strategy exploited in the previous Appendix. We first replace Eq. (B5) to obtain the following explicit expression:

$$L_n = \sum_{q=0}^n (-)^q \sum_{k_0 + \dots + k_q = n} \dot{\mu}_{k_0} \mu_{k_1} \dots \mu_{k_q}. \quad (\text{C4})$$

We then rearrange this sum as follows:

$$L_n = \dot{\mu}_n - \sum_{k=1}^{n-1} \left(\sum_{q=0}^{n-k} (-)^q \sum_{k_0 + \dots + k_q = n-k} \dot{\mu}_{k_0} \mu_{k_1} \dots \mu_{k_q} \right) \mu_k. \quad (\text{C5})$$

Comparing this expression for L_n with Eq. (C4), one finds the desired recursive formula:

$$L_n = \dot{\mu}_n - \sum_{k=1}^{n-1} L_{n-k} \mu_k, \quad \text{with } L_0 = 0. \quad (\text{C6})$$

APPENDIX D: VAN KAMPEN VS RECURSIVE EXPANSION

In this Appendix we compare the cost of our formalism to van Kampen formalism. We build both of the perturbative series up to the fourth term (L_4) of the expansions.

Exploiting van Kampen formalism, one finds that the generator \mathbb{L}_t can be described by the series in Eq. (15) with

$$L_n = \int_0^t d\tau_1 \cdots \int_0^{\tau_{n-1}} d\tau_n \langle V_t^- V_{\tau_1}^- \cdots V_{\tau_n}^- \rangle_{\text{oc}}, \quad (\text{D1})$$

where

$$\langle V_t^- V_{\tau_1}^- \cdots V_{\tau_n}^- \rangle_{\text{oc}} \equiv \sum (-1)^{q-1} \langle V_t^- \cdots V_{\tau_i}^- \rangle \langle V_{\tau_j}^- \cdots V_{\tau_k}^- \rangle \times \langle V_{\tau_l}^- \cdots V_{\tau_m}^- \rangle (\cdots). \quad (\text{D2})$$

While $\langle \cdots \rangle = \text{Tr}_E[\cdots \hat{\rho}_E]$ and we have introduced the new notation $\langle V_t^- V_{\tau_1}^- \cdots V_{\tau_n}^- \rangle_{\text{oc}}$, where the subscript oc stands for ‘‘ordered cumulants.’’ These are defined by the following rules:

$$L_1 = \langle V_t^- \rangle_{\text{oc}} = \langle V_t^- \rangle,$$

$$L_2 = \int_0^t d\tau_1 \langle V_t^- V_{\tau_1}^- \rangle_{\text{oc}} = \int_0^t d\tau_1 (\langle V_t^- V_{\tau_1}^- \rangle - \langle V_t^- \rangle \langle V_{\tau_1}^- \rangle), \quad (\text{D3})$$

$$\begin{aligned} L_3 &= \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \langle V_t^- V_{\tau_1}^- V_{\tau_2}^- \rangle_{\text{oc}} \\ &= \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 (\langle V_t^- V_{\tau_1}^- V_{\tau_2}^- \rangle - \langle V_t^- V_{\tau_1}^- \rangle \langle V_{\tau_2}^- \rangle - \langle V_t^- V_{\tau_2}^- \rangle \langle V_{\tau_1}^- \rangle - \langle V_t^- \rangle \langle V_{\tau_1}^- V_{\tau_2}^- \rangle + \langle V_t^- \rangle \langle V_{\tau_1}^- \rangle \langle V_{\tau_2}^- \rangle + \langle V_t^- \rangle \langle V_{\tau_2}^- \rangle \langle V_{\tau_1}^- \rangle), \end{aligned} \quad (\text{D4})$$

$$\begin{aligned} L_4 &= \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 \langle V_t^- V_{\tau_1}^- V_{\tau_2}^- V_{\tau_3}^- \rangle_{\text{oc}} \\ &= \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 (\langle V_t^- V_{\tau_1}^- V_{\tau_2}^- V_{\tau_3}^- \rangle - \langle V_t^- V_{\tau_1}^- V_{\tau_2}^- \rangle \langle V_{\tau_3}^- \rangle - \langle V_t^- V_{\tau_1}^- V_{\tau_3}^- \rangle \langle V_{\tau_2}^- \rangle - \langle V_t^- V_{\tau_2}^- V_{\tau_3}^- \rangle \langle V_{\tau_1}^- \rangle \\ &\quad - \langle V_t^- V_{\tau_1}^- \rangle \langle V_{\tau_2}^- V_{\tau_3}^- \rangle - \langle V_t^- V_{\tau_2}^- \rangle \langle V_{\tau_1}^- V_{\tau_3}^- \rangle - \langle V_t^- V_{\tau_3}^- \rangle \langle V_{\tau_1}^- V_{\tau_2}^- \rangle - \langle V_t^- \rangle \langle V_{\tau_1}^- V_{\tau_2}^- V_{\tau_3}^- \rangle \\ &\quad + \langle V_t^- V_{\tau_1}^- \rangle \langle V_{\tau_2}^- \rangle \langle V_{\tau_3}^- \rangle + \langle V_t^- V_{\tau_1}^- \rangle \langle V_{\tau_3}^- \rangle \langle V_{\tau_2}^- \rangle + \langle V_t^- V_{\tau_2}^- \rangle \langle V_{\tau_1}^- \rangle \langle V_{\tau_3}^- \rangle + \langle V_t^- V_{\tau_2}^- \rangle \langle V_{\tau_3}^- \rangle \langle V_{\tau_1}^- \rangle \\ &\quad + \langle V_t^- V_{\tau_3}^- \rangle \langle V_{\tau_1}^- \rangle \langle V_{\tau_2}^- \rangle + \langle V_t^- V_{\tau_3}^- \rangle \langle V_{\tau_2}^- \rangle \langle V_{\tau_1}^- \rangle - \langle V_t^- \rangle \langle V_{\tau_1}^- \rangle \langle V_{\tau_2}^- \rangle \langle V_{\tau_3}^- \rangle - \langle V_t^- \rangle \langle V_{\tau_1}^- \rangle \langle V_{\tau_3}^- \rangle \langle V_{\tau_2}^- \rangle \\ &\quad - \langle V_t^- \rangle \langle V_{\tau_2}^- \rangle \langle V_{\tau_1}^- \rangle \langle V_{\tau_3}^- \rangle - \langle V_t^- \rangle \langle V_{\tau_2}^- \rangle \langle V_{\tau_3}^- \rangle \langle V_{\tau_1}^- \rangle - \langle V_t^- \rangle \langle V_{\tau_3}^- \rangle \langle V_{\tau_1}^- \rangle \langle V_{\tau_2}^- \rangle - \langle V_t^- \rangle \langle V_{\tau_3}^- \rangle \langle V_{\tau_2}^- \rangle \langle V_{\tau_1}^- \rangle). \end{aligned} \quad (\text{D5})$$

These explicit expressions show that the complexity of the expansions terms grows very quickly with the expansion order. Moreover, it is not possible to obtain a recursive law, forcing one to repeatedly apply the cumbersome prescription described above to derive any expansion terms. In order to ease the comparison, we now write the same terms exploiting Eq. (C4):

$$\begin{aligned} L_1 &= \langle V_t^- \rangle, \quad L_2 = \int_0^t d\tau_1 (\langle V_t^- V_{\tau_1}^- \rangle - \langle V_t^- \rangle \langle V_{\tau_1}^- \rangle), \\ L_3 &= \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 (\langle V_t^- V_{\tau_1}^- V_{\tau_2}^- \rangle \theta_{\tau_1 \tau_2} - \langle V_t^- V_{\tau_1}^- \rangle \langle V_{\tau_2}^- \rangle - \langle V_t^- \rangle \langle V_{\tau_1}^- V_{\tau_2}^- \rangle \theta_{\tau_1 \tau_2} + \langle V_t^- \rangle \langle V_{\tau_1}^- \rangle \langle V_{\tau_2}^- \rangle), \\ L_4 &= \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 (\langle V_t^- V_{\tau_1}^- V_{\tau_2}^- V_{\tau_3}^- \rangle \theta_{\tau_1 \tau_2 \tau_3} - \langle V_t^- V_{\tau_1}^- V_{\tau_2}^- \rangle \langle V_{\tau_3}^- \rangle \theta_{\tau_1 \tau_2} - \langle V_t^- V_{\tau_1}^- \rangle \langle V_{\tau_2}^- V_{\tau_3}^- \rangle \theta_{\tau_1 \tau_2} \\ &\quad - \langle V_t^- \rangle \langle V_{\tau_1}^- V_{\tau_2}^- V_{\tau_3}^- \rangle \theta_{\tau_1 \tau_2 \tau_3} + \langle V_t^- V_{\tau_1}^- \rangle \langle V_{\tau_2}^- \rangle \langle V_{\tau_3}^- \rangle + \langle V_t^- \rangle \langle V_{\tau_1}^- V_{\tau_2}^- \rangle \langle V_{\tau_3}^- \rangle \theta_{\tau_1 \tau_2} \\ &\quad + \langle V_t^- \rangle \langle V_{\tau_1}^- \rangle \langle V_{\tau_2}^- V_{\tau_3}^- \rangle \theta_{\tau_2 \tau_3} - \langle V_t^- \rangle \langle V_{\tau_3}^- \rangle \langle V_{\tau_2}^- \rangle \langle V_{\tau_1}^- \rangle), \end{aligned} \quad (\text{D6})$$

where $\theta_{\tau_1 \cdots \tau_n}$ is one for $\tau_1 > \cdots > \tau_n$, zero otherwise. Here we has used the following expression for the momenta:

$$\mu_n = \int_0^t d\bar{\tau}_n \langle V_t^- V_{\tau_1}^- \cdots V_{\tau_n}^- \rangle \theta_{\tau_1 \cdots \tau_n}, \quad (\text{D7})$$

which is equivalent to Eq. (9) of the main text, as one can easily check by resolving the time ordering in Eq. (7). By comparing Eqs. (D3)–(D4) it is evident that when the order of the expansion grows, our method requires the computation of a lower number of terms: 4 against 6 for L_3 , and 8 against 20 for L_4 . What is even more important is that the expressions above allow for a

write a string composed by the product of n $V_{\tau_{p_i}}^-$ superoperators in between the parentheses. Partition the string into an arbitrary number of q substrings with $1 < q < n$ by inserting angular brackets in between the V^- of the original string. Multiply the resulting expression by the factor $(-1)^{q-1}$. Concerning the time arguments τ_j , they are organized as follows: The first factor of the string is always V_t^- . The remaining $V_{\tau_i}^-$ display all the permutations of the time arguments $\tau_1, \tau_2, \dots, \tau_{n-1}$ such that in each substring they are chronologically ordered. For example, in Eq. (D2) one has $t \geq \cdots \geq \tau_i, \tau_j \geq \cdots \geq \tau_k$, and $t_i \geq \cdots \geq t_m$ (see [27] for further details).

Following this prescription, we write the first four terms of the van Kampen expansion:

recursive writing [see Eq. (16)]:

$$\begin{aligned}
 L_1 &= \dot{\mu}_1, \\
 L_2 &= \dot{\mu}_2 + L_1\mu_1, \\
 L_3 &= \dot{\mu}_3 - L_2\mu_1 - L_1\mu_2, \\
 L_4 &= \dot{\mu}_4 - L_3\mu_1 - L_2\mu_2 - L_1\mu_3.
 \end{aligned}
 \tag{D8}$$

This recursion reduces even further the number of terms that need to be computed at each order, and allows for a diagrammatic description that eases their construction.

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