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An $\hbar$-asymptotic analysis of the error in the thawed Gaussian approximation and in the corresponding initial value representation of the quantum propagator

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Abstract

An algebraic approach based on the multimode two-photon Lie algebra and its corresponding Lie group is followed to derive a formal solution to the time-dependent Schrödinger equation. This solution is written as an expansion series whose leading term corresponds to the thawed Gaussian approximation (TGA). Our scheme provides the most general expression reported so far for this approximation. By using the coherent state representation of the formal solution, the correction term to the TGA is analysed in the zero $\hbar$ asymptotic limit. The error is generally found not to vanish in this semiclassical limit. The same approach is followed to study the remainder to the TGA initial value representation (IVR) of the quantum propagator. This correction is found not to vanish either in the zero $\hbar$ limit. Hence, the TGA IVR would not be the correct semiclassical asymptotic form of the quantum propagator. The origin of this behaviour is shown to be in the existence of contributions from unphysical saddle points in the semiclassical limit. These would unveil an incorrect analytic structure of the TGA IVR propagator in that limit.

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1. Introduction

In a seminal paper published 30 years ago [1], Heller proposed an approximation for the quantum evolution of an initial coherent state (i.e. a Gaussian state in the coordinate representation) that is known as the thawed Gaussian approximation (TGA). This assumes that the initial Gaussian state remains Gaussian with time-dependent parameters. These parameters are then obtained from the system classical trajectories. The TGA provides an exact solution

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of the time-dependent Schrödinger equation (TDSE) for quantum Hamiltonians that are at most quadratic in the coordinates and momenta. In general, it just gives a local quadratic approximation to a mixed representation of the quantum propagator; this representation is defined as the propagator matrix element between a position state and a coherent state.

A decade later, Heller and coworkers [2, 3] showed that the parametrization of a Gaussian wave-packet is not unique. In other words, they found a set of possible forms known as the initial ket manifold. By a proper choice of the parameters, this generalization of the TGA, which is known as generalized Gaussian wave packet dynamics (GGWPD), was shown to be both equivalent to and derivable from a WKB approximation to the solution of the TDSE. Using this formulation and Heller's [4] extension of Miller's semiclassical theory [5], these authors also derived the semiclassical expression for the matrix elements of the propagator between coherent states. A similar approach was followed by Weissman [6, 7] to obtain the same semiclassical expression. This coherent state representation of the semiclassical propagator was known from the work of Klauder [8–10]. Its derivation from the coherent-state path integral expression of the propagator is also possible by using the stationary phase approximation. A careful realization of this approach, followed initially by Klauder [8–10], was carried out by Baranger *et al* [11]. These last authors show how different forms of the semiclassical propagator are obtained in correspondence with the symbol (Q, P, Weyl, etc) chosen for the classical Hamiltonian associated with its quantum counterpart. Recently, a derivation of this most general semiclassical expression from a WKB-like solution to the time-dependent Schrödinger equation (TDSE) in the Bargmann representation has been reported [12].

More useful than this representation of the quantum propagator as a coherent state matrix element are the so-called initial value representations (IVR). The semiclassical IVR (SCIVR) are integral representations that can be evaluated using classical trajectories and in which the integration is performed over all possible initial conditions. The well-known TGA IVR is obtained by expanding the given initial state in the overcomplete basis set of coherent sates and evolving each one of these independently with the TGA. Kay argued [13] that this form is a correct SCIVR, i.e. he showed that what has been assumed so far to be the leading semiclassical asymptotic contribution to its coordinate matrix elements provides the semiclassical Van Vleck expression [14]; we will show in this work, and anticipate here, that such an assumption is not generally correct. Later, Baranger *et al* [11], after a thorough analysis of this approximation, derived a generalized one-dimensional expression valid for choices of the classical Hamiltonian other than the Weyl symbol of its quantum counterpart. The multidimensional generalization was obtained by Pollak and Miret-Artés [15]. Recently, Parisio and de Aguiar [16] have presented a more general expression that includes the previous ones as particular cases. In a different class of SCIVR propagators we find the one obtained by Herman and Kluk [17], which is closely related to a form derived by Solari [18] for one-dimensional systems. The work carried out on the HK propagator has been reviewed by Grossmann [19], and more recently by Miller [20] and by Kay [21].

Kay [13] has developed an approach to derive all these and other more general IVR propagators; all of them are assumed to be valid in the semiclassical limit (in other words, the Van Vleck semiclassical expression [14] is obtained from what is considered to be the leading asymptotic contribution). Kay also compared numerically [22] several of these forms and found, in particular, that the TGA IVR is inferior to the HK propagator, even though propagators in the TGA class have preexponential factors decreasing with time, while those in the HK class have growing preexponential factors; furthermore, later numerical experiments [23] showed a loss of unitarity as time increases in the TGA IVR propagator. Despite these results, a recent debate [24, 25] has opened again the issue on the best choice for the semiclassical IVR of the quantum propagator, and it has been argued [25] in this context that the HK class cannot be derived from asymptotic methods. However, this conjecture has been proved to be false recently [12, 26]. Besides, a generalized form of the HK propagator that is valid for any of the possible classical representations of a given quantum Hamiltonian and for any of the possible operator ordering schemes used to quantize a classical Hamiltonian function has been obtained as a correct semiclassical asymptotic limit [12].

A similar asymptotic analysis of the TGA IVR is currently lacking. Pollak and coworkers [27–29] have used this SCIVR (as well as the HK SCIVR) as the zeroth order term of an expansion of the exact propagator. Then, this expansion is written in terms of a correction operator and allows for the evaluation of the quantum propagator up to the desired accuracy within the same SCIVR scheme. However, the $\hbar \to 0$ asymptotic analysis of this scheme has not been performed either. These tasks will become the main goal of this work. Specifically, we shall prove that, unlike the HK SCIVR, the TGA IVR is not generally a semiclassical asymptotic approximation to the quantum propagator. This is an important result that refutes previous opposite statements [11, 13, 25]. Our analysis will follow an algebraic approach that will make use of many results and properties of the multimode two-photon Lie algebra and its corresponding Lie group. As a first step, we will derive a formal expression for the exact solution of the TDSE corresponding to an initial generalized coherent state. This expression depends on a set of arbitrary parameters, a set equivalent to Heller and coworkers' initial ket manifold [2, 3]. The formal solution will be written as a perturbative expansion whose leading term corresponds to a generalized local quadratic approximation that includes the TGA as a particular case and the GGWPD as a more general one. We will also provide the explicit form of the correction term in this solution. After projecting the derived solution on a representation (the coherent state representation shall be chosen), we will show that the TGA approximation differs from the formal solution in a correction term whose perturbative expansion includes contributions proportional to all positive and negative powers of $\hbar$. Yet, for a particular choice of the arbitrary parameters, the contribution from all the negative powers vanishes; the resulting $\hbar$ expansion then becomes a true semiclassical asymptotic series whose leading term provides the well-known semiclassical expression for the coherent state matrix elements of the quantum propagator. However, this choice presupposes the use of complex trajectories defined by a two-time boundary condition, which is in contradiction with the particular scheme followed in the SCIVR. Namely, in this integral representation of the quantum propagator, trajectories must be defined by their initial conditions; thus the freedom we had in the evaluation of the propagator matrix elements now disappears. Consequently, the TGA IVR correction will generally yield terms proportional to the negative powers of $\hbar$, which prevents this particular IVR from being the asymptotically leading semiclassical representation of the quantum propagator; hence, for instance, Pollak and coworkers' correction operator to the TGA IVR [15, 27–29] would also present a bad behaviour in the zero $\hbar$ limit. In all these cases, the badly behaved terms are associated with spurious saddle points appearing in the asymptotic evaluation of generic matrix elements of the TGA IVR propagator. The existence of these saddle points have been noticed in earlier works [13]; however, their effect has been ignored so far, which has led to not always correct conclusions about the validity of the TGA IVR as a semiclassical propagator. Other inadequate behaviours of this propagator such as the fast loss of unitarity may have an explanation in this unveiled analytic structure. Despite these negative conclusions, we will show that the TGA IVR can provide a good approximation to the quantum propagator when the terms higher than quadratic in the expansion of the Hamiltonian around the classical trajectories behave as a small enough perturbation.

This paper is organized as follows. Section 2 is an introduction in which we present our algebraic approach and provide some relevant results and properties of the multimode two-photon Lie algebra and its corresponding Lie group. In section 3, we define our generalized coherent state and write its Gaussian expression in the coordinate representation. In section 4, we derive a perturbative expansion to the solution of the TDSE for an initial generalized coherent state whose leading term corresponds to a quadratic local approximation. From this leading term, we recover in section 5 the GGWPD and the TGA. The $\hbar$-asymptotic analysis of the correction term corresponding to both the TGA and the TGA IVR are performed respectively in sections 6 and 7. We end up in section 8 with the main conclusions of this work.

2. The basic formalism of the algebraic approach

2.1. The d-mode two-photon Lie algebra

In this paper, we will follow an algebraic approach and make use of group-theoretical methods to construct multimode coherent states, i.e. states that are Gaussian wave packets in the coordinate representation. The relevant algebraic structure in our case is provided by the multimode two-photon Lie algebra and its corresponding Lie group. This algebra for a system with d degrees of freedom is spanned by the set of operators

$$\left\{a_m^\dagger a_n^\dagger, a_m a_n, a_m^\dagger a_n + \frac{\delta_{mn}}{2}\hat{I}, a_n^\dagger, a_n, \hat{I};\; m,n = 1,\ldots,d\right\}, \tag{1}$$

where $\hat{I}$ is the identity operator; a_n and $a_n^\dagger$ are respectively the annihilation and creation operators of each mode. One can easily check that the algebra is indeed closed under its binary operation of commutation, with $[a_m, a_n^\dagger] = \delta_{mn}$ and $[a_m, a_n] = [a_m^\dagger, a_n^\dagger] = 0$.

The d-mode two-photon Lie group associated with this Lie algebra is obtained through the exponential map. The following four operators belonging to this group are particularly relevant for this work.

(1) The generalized displacement operator,

$$\hat{D}(\boldsymbol{\alpha}, \boldsymbol{\beta}^\star) = \exp\left[\hbar^{-\frac{1}{2}}(\boldsymbol{\alpha}\,\boldsymbol{a}^\dagger - \boldsymbol{\beta}^\star\,\boldsymbol{a})\right], \tag{2}$$

where $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}^\star$ are d-dimensional vectors with complex components α_n and $\beta_n^\star$ respectively (as usual, we use the superscript $\star$ for complex conjugation), and $\boldsymbol{a}$ and $\boldsymbol{a}^\dagger$ are d-vectors with operator components a_n and $a_n^\dagger$. The product of two of these vectors, as appears in this equation, shall be performed as a dot product; e.g., $\boldsymbol{\alpha}\,\boldsymbol{a}^\dagger = \boldsymbol{a}^\dagger\,\boldsymbol{\alpha} \equiv \sum_{n=1}^d \alpha_n a_n^\dagger$ and $\boldsymbol{\alpha}\boldsymbol{\beta}^\star = \boldsymbol{\beta}^\star\boldsymbol{\alpha} \equiv \sum_{n=1}^d \alpha_n\beta_n^\star$. The displacement operator will be unitary only if $\boldsymbol{\beta} = \boldsymbol{\alpha}$. Equation (2) shows explicitly the $\hbar$ dependence which, in other notations, is generally included in the complex $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}^\star$ parameters.

(2) The generalized squeezing operator,

$$\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star) = \exp\left[\tfrac{1}{2}(\boldsymbol{a}^\dagger\,\boldsymbol{\Xi}\,\boldsymbol{a}^\dagger - \boldsymbol{a}\,\boldsymbol{\Pi}^\star\,\boldsymbol{a})\right], \tag{3}$$

where $\boldsymbol{\Xi}$ and $\boldsymbol{\Pi}^\star$ are $d\times d$ complex symmetric matrices. The product involving two vectors and a matrix, as appears in this equation, shall be performed as a dot product of one of the vectors with the matrix linear transformation of the other; e.g., $\boldsymbol{a}^\dagger\,\mathbf{M}\,\boldsymbol{a} = \sum_{m=1}^d\sum_{n=1}^d a_m^\dagger \mathbf{M}_{mn} a_n$, $\boldsymbol{a}^\dagger\,\mathbf{M}\,\boldsymbol{\alpha} = \sum_{m=1}^d\sum_{n=1}^d a_m^\dagger \mathbf{M}_{mn}\alpha_n$, $\boldsymbol{\beta}^\star\,\mathbf{M}\,\boldsymbol{\alpha} = \sum_{m=1}^d\sum_{n=1}^d \beta_m^\star \mathbf{M}_{mn}\alpha_n$, where $\mathbf{M}$ is a general $d\times d$ complex matrix. Only if $\boldsymbol{\Pi} = \boldsymbol{\Xi}$, this squeezing operator will be unitary.

(3) The operator

$$\hat{T}(\boldsymbol{\Upsilon}) = \exp\left[\boldsymbol{a}^\dagger\,\boldsymbol{\Upsilon}\,\boldsymbol{a} - \tfrac{1}{2}(\mathrm{Tr}\,\boldsymbol{\Upsilon})\hat{I}\right], \tag{4}$$

where $\boldsymbol{\Upsilon}$ is a $d\times d$ complex matrix. This operator will be unitary only if $\boldsymbol{\Upsilon}$ is an anti-Hermitian matrix, i.e. if $\widetilde{\boldsymbol{\Upsilon}}^\star = -\boldsymbol{\Upsilon}$.

(4) The operator

$$\hat{R}(\Phi) = \exp(\mathrm{i}\Phi\hat{I}), \tag{5}$$

where Φ is a complex number; only if Φ is real, then $\hat{R}$ is unitary. This operator commutes with all the others.

In terms of these four operators, a general element, $\hat{G}$, of the Lie group may be written as

$$\hat{G}(\chi) = \hat{D}(\alpha, \beta^\star)\hat{R}(\Phi)\hat{S}(\Xi, \Pi^\star)\hat{T}(\Upsilon). \tag{6}$$

Therefore, the number of complex independent Lie group parameters is $N_G = d(2d+3)+1$; we shall denote these parameters with the N_G-dimensional vector χ, and the dependence of $\hat{G}$ on them by writing $\hat{G}(\chi)$. The two subsets formed respectively by all the operations $\{\hat{D}(\alpha, \beta^\star)\hat{R}(\Phi); \forall\alpha, \beta^\star, \Phi\}$ and $\{\hat{S}(\Xi, \Pi^\star)\hat{T}(\Upsilon); \forall\Xi, \Pi^\star, \Upsilon\}$ are subgroups of the d-mode two-photon Lie group.

2.2. *The smallest faithful matrix representation*

The smallest faithful (i.e. one-to-one) matrix representation (SFMR) of both the d-mode two-photon Lie algebra and the corresponding Lie group has dimension $(2d+2) \times (2d+2)$ [30]. By definition, its matrices preserve the algebraic and group structures; we shall write them in the following form:

$$\mathcal{M} = \begin{pmatrix} m_{11} & 0 & 0 & 0 \\ \boldsymbol{m}_{21} & \boldsymbol{M}_{22} & \boldsymbol{M}_{23} & 0 \\ \boldsymbol{m}_{31} & \boldsymbol{M}_{32} & \boldsymbol{M}_{33} & 0 \\ m_{41} & \widetilde{\boldsymbol{m}_{42}} & \widetilde{\boldsymbol{m}_{43}} & m_{44} \end{pmatrix}, \tag{7}$$

where $\boldsymbol{M}_{ij}$ represents a complex $d \times d$ matrix, $\boldsymbol{m}_{ij}$ a complex d-vector (written here as a column vector with a tilde denoting the corresponding transpose row vector), m_{ij} a complex number, and 0 is either a null d-vector or a null element (later on it will also denote a $d \times d$ null matrix). For the operators spanning the algebra, we have $m_{11} = m_{44} = 0$, while for the group elements $\hat{G}$, $m_{11} = m_{44} = 1$. Our faithful matrix representation, $\mathcal{M}$, differs from that defined in [30] (let us denote this as $\mathcal{M}^0$); however both representations are completely equivalent because they are simply related by $\mathcal{M}_{i,j} = \mathcal{M}^0{}_{f(i),f(j)}$, where f is a bijection from the set of indexes $\{i; i = 1, 2, \ldots, 2d+2\}$ to itself. Our choice is more convenient for the purpose of this work. We shall use the symbol $\boldsymbol{M}$ to denote the central $2d \times 2d$ block of $\mathcal{M}$, namely

$$\boldsymbol{M} = \begin{pmatrix} \boldsymbol{M}_{22} & \boldsymbol{M}_{23} \\ \boldsymbol{M}_{32} & \boldsymbol{M}_{33} \end{pmatrix}. \tag{8}$$

For instance, the most general linear superposition of the operators spanning the multimode two-photon algebra and its matrix representation are respectively

$$\hat{A} = \left[\boldsymbol{a}^\dagger\boldsymbol{\Psi}_{\mathrm{c}}\boldsymbol{a} - \tfrac{1}{2}(\mathrm{Tr}\,\boldsymbol{\Psi}_{\mathrm{c}})\hat{I}\right] + \tfrac{1}{2}(\boldsymbol{a}^\dagger\boldsymbol{\Psi}_{\mathrm{l}}\boldsymbol{a}^\dagger) + \tfrac{1}{2}(\boldsymbol{a}\boldsymbol{\Psi}_{\mathrm{r}}\boldsymbol{a}) + \boldsymbol{\xi}_{\mathrm{l}}\boldsymbol{a}^\dagger + \boldsymbol{\xi}_{\mathrm{r}}\boldsymbol{a} + \tfrac{1}{2}\varsigma\hat{I}$$

$$\longrightarrow \mathcal{A} = \begin{pmatrix} 0 & 0 & 0 & 0 \\ \boldsymbol{\xi}_{\mathrm{l}} & \boldsymbol{\Psi}_{\mathrm{c}} & \boldsymbol{\Psi}_{\mathrm{l}} & 0 \\ -\boldsymbol{\xi}_{\mathrm{r}} & -\boldsymbol{\Psi}_{\mathrm{r}} & -\widetilde{\boldsymbol{\Psi}}_{\mathrm{c}} & 0 \\ -\varsigma & -\widetilde{\boldsymbol{\xi}}_{\mathrm{r}} & -\widetilde{\boldsymbol{\xi}}_{\mathrm{l}} & 0 \end{pmatrix}, \tag{9}$$

where ς is a complex number, $\boldsymbol{\xi}_{\mathrm{l}}$ and $\boldsymbol{\xi}_{\mathrm{r}}$ are complex d-vectors, $\boldsymbol{\Psi}_{\mathrm{r}}$ and $\boldsymbol{\Psi}_{\mathrm{l}}$ are complex symmetric $d \times d$ matrices, and $\boldsymbol{\Psi}_{\mathrm{c}}$ is a general complex $d \times d$ matrix (the tilde over a matrix denotes its transpose). Furthermore, the exponential of this matrix provides a representation of the general element $\hat{G}$ of the Lie group. A convenient parametrization of these elements, different from that given in equation (6), and its matrix representation are written respectively in the form

$$\hat{G} = \exp\left(\tfrac{1}{2}\boldsymbol{a}^\dagger \boldsymbol{\Psi}_1 \boldsymbol{a}^\dagger + \boldsymbol{\xi}_1 \boldsymbol{a}^\dagger\right) \exp\left[\boldsymbol{a}^\dagger \boldsymbol{\Psi}_\mathrm{c} \boldsymbol{a} + \tfrac{1}{2}(\mathrm{Tr}\,\boldsymbol{\Psi}_\mathrm{c})\hat{I} + \tfrac{1}{2}\varsigma \hat{I}\right] \exp\left(\tfrac{1}{2}\boldsymbol{a}\boldsymbol{\Psi}_\mathrm{r}\boldsymbol{a} + \boldsymbol{\xi}_\mathrm{r}\boldsymbol{a}\right)$$

$$\longrightarrow \boldsymbol{\mathcal{G}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ \boldsymbol{\xi}_1 - \boldsymbol{\Psi}_1 \mathrm{e}^{-\widetilde{\boldsymbol{\Psi}}_\mathrm{c}} \boldsymbol{\xi}_\mathrm{r} & \mathrm{e}^{\boldsymbol{\Psi}_\mathrm{c}} - \boldsymbol{\Psi}_1 \mathrm{e}^{-\widetilde{\boldsymbol{\Psi}}_\mathrm{c}} \boldsymbol{\Psi}_\mathrm{r} & \boldsymbol{\Psi}_1 \mathrm{e}^{-\widetilde{\boldsymbol{\Psi}}_\mathrm{c}} & 0 \\ -\mathrm{e}^{-\widetilde{\boldsymbol{\Psi}}_\mathrm{c}} \boldsymbol{\xi}_\mathrm{r} & -\mathrm{e}^{-\widetilde{\boldsymbol{\Psi}}_\mathrm{c}} \boldsymbol{\Psi}_\mathrm{r} & \mathrm{e}^{-\widetilde{\boldsymbol{\Psi}}_\mathrm{c}} & 0 \\ -\varsigma + \widetilde{\boldsymbol{\xi}}_1 \mathrm{e}^{-\widetilde{\boldsymbol{\Psi}}_\mathrm{c}} \boldsymbol{\xi}_\mathrm{r} & -\widetilde{\boldsymbol{\xi}}_\mathrm{r} + \widetilde{\boldsymbol{\xi}}_1 \mathrm{e}^{-\widetilde{\boldsymbol{\Psi}}_\mathrm{c}} \boldsymbol{\Psi}_\mathrm{r} & -\widetilde{\boldsymbol{\xi}}_1 \mathrm{e}^{-\widetilde{\boldsymbol{\Psi}}_\mathrm{c}} & 1 \end{pmatrix}. \tag{10}$$

Many other parametrizations of the group element $\hat{G}$ are possible by changing the ordering of the exponentials or the operators included in each of the exponents [30]. Relations among them can be established by the so-called disentangling theorems, which can be directly derived with the help of the SFMR.

To conclude this section, let us write the matrix representation of the four operators defined in section 2.1.

(1) The generalized displacement operator:

$$\hat{D}(\boldsymbol{\alpha}, \boldsymbol{\beta}^\star) \longrightarrow \boldsymbol{\mathcal{D}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ \hbar^{-\frac{1}{2}}\boldsymbol{\alpha} & \mathbf{I} & 0 & 0 \\ \hbar^{-\frac{1}{2}}\boldsymbol{\beta}^\star & 0 & \mathbf{I} & 0 \\ 0 & \hbar^{-\frac{1}{2}}\widetilde{\boldsymbol{\beta}}^\star & -\hbar^{-\frac{1}{2}}\widetilde{\boldsymbol{\alpha}} & 1 \end{pmatrix}, \tag{11}$$

where $\mathbf{I}$ is the $d \times d$ identity matrix.

(2) The generalized squeezing operator:

$$\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star) \longrightarrow \boldsymbol{\mathcal{S}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \boldsymbol{\Delta} & \boldsymbol{\Omega} & 0 \\ 0 & \boldsymbol{\Lambda}^\star & \widetilde{\boldsymbol{\Delta}} & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \tag{12}$$

where

$$\boldsymbol{\Omega} = \boldsymbol{\Theta}^{-1}(\sinh\boldsymbol{\Theta})\boldsymbol{\Xi}, \tag{13a}$$

$$\boldsymbol{\Lambda}^\star = \boldsymbol{\Pi}^\star(\sinh\boldsymbol{\Theta})\boldsymbol{\Theta}^{-1}, \tag{13b}$$

$$\boldsymbol{\Delta} = \cosh\boldsymbol{\Theta}, \tag{13c}$$

$$\boldsymbol{\Theta}^2 = \boldsymbol{\Xi}\boldsymbol{\Pi}^\star. \tag{13d}$$

(3) The $\hat{T}$ operator:

$$\hat{T}(\boldsymbol{\Upsilon}) \longrightarrow \boldsymbol{\mathcal{T}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \mathrm{e}^{\boldsymbol{\Upsilon}} & 0 & 0 \\ 0 & 0 & \mathrm{e}^{-\widetilde{\boldsymbol{\Upsilon}}} & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}. \tag{14}$$

(4) The $\hat{R}$ operator:

$$\hat{R}(\Phi) \longrightarrow \boldsymbol{\mathcal{R}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \mathbf{I} & 0 & 0 \\ 0 & 0 & \mathbf{I} & 0 \\ -2\mathrm{i}\Phi & 0 & 0 & 1 \end{pmatrix}. \tag{15}$$

Since the matrix representation of a group element is unique, we must have $\mathcal{G} = \mathcal{DRST}$; then from this equality and the one-to-one property of the SFMR, we can establish the relationship between the two parametrizations given respectively by (6) and (10), and derive in such a simple way the corresponding disentangling theorem.

2.3. Some useful results derived with the help of the SFMR

With the help of the SFMR, one can readily derive the following operator transformations:

$$\hat{D}^{-1}(\boldsymbol{\alpha}, \boldsymbol{\beta}^{\star}) \begin{pmatrix} \boldsymbol{a} \\ \boldsymbol{a}^{\dagger} \end{pmatrix} \hat{D}(\boldsymbol{\alpha}, \boldsymbol{\beta}^{\star}) = \begin{pmatrix} \boldsymbol{a} + \hbar^{-\frac{1}{2}}\boldsymbol{\alpha} \\ \boldsymbol{a}^{\dagger} + \hbar^{-\frac{1}{2}}\boldsymbol{\beta}^{\star} \end{pmatrix}, \tag{16}$$

$$\hat{S}^{-1}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^{\star}) \begin{pmatrix} \boldsymbol{a} \\ \boldsymbol{a}^{\dagger} \end{pmatrix} \hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^{\star}) = \boldsymbol{S} \begin{pmatrix} \boldsymbol{a} \\ \boldsymbol{a}^{\dagger} \end{pmatrix}, \tag{17}$$

$$\hat{T}^{-1}(\boldsymbol{\Upsilon}) \begin{pmatrix} \boldsymbol{a} \\ \boldsymbol{a}^{\dagger} \end{pmatrix} \hat{T}(\boldsymbol{\Upsilon}) = \boldsymbol{T} \begin{pmatrix} \boldsymbol{a} \\ \boldsymbol{a}^{\dagger} \end{pmatrix}, \tag{18}$$

where $\hat{D}^{-1}(\boldsymbol{\alpha}, \boldsymbol{\beta}^{\star}) = \hat{D}(-\boldsymbol{\alpha}, -\boldsymbol{\beta}^{\star})$, $\hat{S}^{-1}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^{\star}) = \hat{S}(-\boldsymbol{\Xi}, -\boldsymbol{\Pi}^{\star})$, and $\hat{T}^{-1}(\boldsymbol{\Upsilon}) = \hat{T}(-\boldsymbol{\Upsilon})$ are inverse operators, and $\binom{\boldsymbol{a}}{\boldsymbol{a}^{\dagger}}$ is the notation for the $2d$-dimensional column vector built up as a direct sum of $\boldsymbol{a}$ and $\boldsymbol{a}^{\dagger}$; later on, we will use the same notation to introduce other $2d$-vectors such as $\binom{\boldsymbol{\alpha}}{\boldsymbol{\beta}^{\star}}$. We stick to the notation introduced by (7) and (8) and call $\boldsymbol{S}$ and $\boldsymbol{T}$ the central $2d \times 2d$ blocks of $\mathcal{S}$ and $\mathcal{T}$, respectively; these matrices are given by (12) and (14).

A useful property of the group element $\hat{G}$, which is a consequence of the algebraic structure, is that its derivatives with respect to the group parameters χ_n can be expressed in the following simple way:

$$\frac{\partial \hat{G}(\chi)}{\partial \chi_n} = \hat{A}_{\mathrm{l}n}(\chi)\hat{G}(\chi) = \hat{G}(\chi)\hat{A}_{\mathrm{r}n}(\chi), \tag{19}$$

where the left $\hat{A}_{\mathrm{l}n}(\chi)$ and right $\hat{A}_{\mathrm{r}n}(\chi)$ operators are elements of the algebra as those given in (9). The particular form of $\hat{A}_{\mathrm{l}n}$ and $\hat{A}_{\mathrm{r}n}$ can be obtained by writing (19) in the SFMR, namely

$$\mathcal{A}_{\mathrm{l}n} = \frac{\partial \mathcal{G}(\chi)}{\partial \chi_n}\mathcal{G}^{-1}(\chi); \qquad \mathcal{A}_{\mathrm{r}n} = \mathcal{G}^{-1}(\chi)\frac{\partial \mathcal{G}(\chi)}{\partial \chi_n}. \tag{20}$$

From these matrices and the one-to-one correspondence established by (9), we can readily obtain the operators $\hat{A}_{\mathrm{l}n}$ and $\hat{A}_{\mathrm{r}n}$.

If the (self-adjoint) Hamiltonian operator $\hat{H}(t)$ of a quantum system has the linear form given in (9), the corresponding (unitary) evolution operator $\hat{U}(t)$ will be an element of the multimode two-photon Lie group. If $\mathcal{H}(t)$ and $\mathcal{U}(t)$ are their corresponding matrix representations, then the Heisenberg equation $\mathrm{i}\hbar\, \mathrm{d}\hat{U}(t)/\mathrm{d}t = \hat{H}(t)\hat{U}(t)$, together with its usual initial condition $\hat{U}(0) = \hat{I}$, can be written in matrix form as [30]

$$\mathrm{i}\hbar \frac{\mathrm{d}\mathcal{U}(t)}{\mathrm{d}t} = \mathcal{H}(t)\mathcal{U}(t), \tag{21a}$$

$$\mathcal{U}(0) = \mathcal{I}, \tag{21b}$$

where $\mathcal{I}$ is the $(2d+2) \times (2d+2)$ identity matrix. If the expansion of $\hat{H}(t)$ in the operators spanning the algebra does not have terms proportional to $\hat{I}$, $\boldsymbol{a}$ and $\boldsymbol{a}^{\dagger}$, then $\hat{U}(t)$ will belong to the subgroup $\{\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^{\star})\hat{T}(\boldsymbol{\Upsilon}); \forall \boldsymbol{\Xi}, \boldsymbol{\Pi}^{\star}, \boldsymbol{\Upsilon}\}$; hence, the matrices in (21a) and (21b) can be restricted to their corresponding central $2d \times 2d$ blocks $\boldsymbol{H}$, $\boldsymbol{U}$, and $\boldsymbol{I}$, these being defined in accordance with (7) and (8); namely,

$$\mathrm{i}\hbar \frac{\mathrm{d}\boldsymbol{U}(t)}{\mathrm{d}t} = \boldsymbol{H}(t)\boldsymbol{U}(t), \tag{22a}$$

$$\boldsymbol{U}(0) = \boldsymbol{I}. \tag{22b}$$

The SFMR is also very useful in the evaluation of operator expectation values. For example, if $|0\rangle$ represents the normalized vacuum state (i.e. $a_n|0\rangle = 0, \forall n$, and $\langle 0|0\rangle = 1$), the vacuum expectation value of any operator belonging to the Lie group is, from (10), given by

$$\langle 0|\hat{G}|0\rangle = \exp\left[\frac{1}{2}(\mathrm{Tr}\,\boldsymbol{\Psi}_{\mathrm{c}} + \varsigma)\right] = (\det \boldsymbol{G}_{33})^{-\frac{1}{2}} \exp\left[-\frac{g_{41}}{2} + \frac{1}{2}\boldsymbol{g}_{43}\,(\boldsymbol{G}_{33})^{-1}\,\boldsymbol{g}_{31}\right], \tag{23}$$

where in the second equality we have written the expectation value, in a parametrization independent form, in terms of the matrix elements of the operator representation $\boldsymbol{\mathcal{G}}$ defined as in (7).

A remarkable property of the SFMR of the group elements $\hat{G}$ is that its central $2d \times 2d$ block, $\boldsymbol{G}$, is a symplectic matrix. Namely $\boldsymbol{G}$ satisfies

$$\widetilde{\boldsymbol{G}}\boldsymbol{J}\boldsymbol{G} = \boldsymbol{J}, \tag{24}$$

where $\boldsymbol{J}$ is the $2d \times 2d$ skew-symmetric matrix $\boldsymbol{J} = \left(\begin{smallmatrix} 0 & \mathbf{I} \\ -\mathbf{I} & 0 \end{smallmatrix}\right)$. Hence, its inverse matrix, $\boldsymbol{G}^{-1}$, which is also the central $2d \times 2d$ block of the matrix $\boldsymbol{\mathcal{G}}^{-1}$ representing the inverse operator $\hat{G}^{-1}$, is given by

$$\boldsymbol{G}^{-1} = \boldsymbol{J}^{-1}\widetilde{\boldsymbol{G}}\boldsymbol{J}. \tag{25}$$

Furthermore we have $\det \boldsymbol{G} = 1$, which implies $\det \boldsymbol{\mathcal{G}} = 1$.

On some occasions, we will have to make use of the coordinate $\hat{q}_n$ and conjugated momentum $\hat{p}_n$ operators for each mode (in vectorial notation, $\hat{\boldsymbol{q}}$ and $\hat{\boldsymbol{p}}$). As a primary choice, we shall take the following relationships:

$$\begin{pmatrix} \boldsymbol{a} \\ \boldsymbol{a}^\dagger \end{pmatrix} = \frac{1}{\sqrt{2\hbar}} \begin{pmatrix} \hat{\boldsymbol{q}} + \mathrm{i}\hat{\boldsymbol{p}} \\ \hat{\boldsymbol{q}} - \mathrm{i}\hat{\boldsymbol{p}} \end{pmatrix}. \tag{26}$$

Yet, a more general definition of the creation and annihilation operators in terms of the position and momentum operators is provided by the transformation

$$\begin{pmatrix} \boldsymbol{a} \\ \boldsymbol{a}^\dagger \end{pmatrix} \to \begin{pmatrix} \boldsymbol{a}' \\ \boldsymbol{a}'^\dagger \end{pmatrix} \equiv \hat{W}^{-1} \begin{pmatrix} \boldsymbol{a} \\ \boldsymbol{a}^\dagger \end{pmatrix} \hat{W} = \frac{1}{\sqrt{2\hbar}} \boldsymbol{W} \begin{pmatrix} \hat{\boldsymbol{q}} + \mathrm{i}\hat{\boldsymbol{p}} \\ \hat{\boldsymbol{q}} - \mathrm{i}\hat{\boldsymbol{p}} \end{pmatrix}, \tag{27}$$

where $\hat{W}$ is an operator (usually unitary) belonging to the subgroup $\{\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{T}(\boldsymbol{\Upsilon});\ \forall \boldsymbol{\Xi}, \boldsymbol{\Pi}^\star, \boldsymbol{\Upsilon}\}$ and $\boldsymbol{W}$ is its corresponding $2d \times 2d$ matrix representation. The operators in the set $\{\boldsymbol{a}', \boldsymbol{a}'^\dagger\}$ satisfy the same commutation relations as those in $\{\boldsymbol{a}, \boldsymbol{a}^\dagger\}$, thus a two-photon Lie algebra can be analogously associated with the former. Now, if $\hat{A}'(\psi')$ is a general element of this new two-photon Lie algebra and $\hat{G}'(\boldsymbol{\chi}')$ a general element of the corresponding Lie group, we have

$$\hat{A}'(\psi') = \hat{W}^{-1}\hat{A}(\psi')\hat{W}, \tag{28}$$

$$\hat{G}'(\boldsymbol{\chi}') = \hat{W}^{-1}\hat{G}(\boldsymbol{\chi}')\hat{W}, \tag{29}$$

where $\hat{A}(\psi')$ and $\hat{G}(\boldsymbol{\chi}')$ are the corresponding operators in the $\{\boldsymbol{a}, \boldsymbol{a}^\dagger\}$ two-photon Lie algebra and Lie group, respectively. Hence $\hat{A}'(\psi')$ and $\hat{G}'(\boldsymbol{\chi}')$ are obtained from $\hat{A}(\psi')$ and $\hat{G}(\boldsymbol{\chi}')$ by replacing in the latter the operators in the set $\{\boldsymbol{a}, \boldsymbol{a}^\dagger\}$ by those in the set $\{\boldsymbol{a}', \boldsymbol{a}'^\dagger\}$, respectively. Throughout the text we will keep this notation and use primed operators to represent those belonging to the two-photon algebra and group associated with the general set $\{\boldsymbol{a}', \boldsymbol{a}'^\dagger\}$. Note that the representation matrices for the operators $\hat{A}'(\psi')$ and $\hat{G}'(\chi')$ in the algebra based on $\{\boldsymbol{a}', \boldsymbol{a}'^\dagger\}$ coincide, respectively, with the representation matrices for $\hat{A}(\psi')$ and $\hat{G}(\chi')$ in the algebra associated with $\{\boldsymbol{a}, \boldsymbol{a}^\dagger\}$; we shall denote these two matrices as $\boldsymbol{\mathcal{A}}'$ and $\boldsymbol{\mathcal{G}}'$ and their $2d \times 2d$ central blocks as $\boldsymbol{A}'$ and $\boldsymbol{G}'$, respectively.

To end up this section, we should remark that all results in the following sections that will be derived without requiring an explicit choice of the expression relating the creation and annihilation operators with the coordinate and momentum operators are completely general and therefore valid for any such choice, let it be either the particular one given in (26) or the more general one given in (27).

3. The multimode coherent state

Once we have introduced the algebraic formalism, we are ready to write the expression of the most general coherent state provided by the d-mode two-photon Lie algebra, that is

$$|\chi\rangle = \hat{G}(\chi)|0\rangle = \hat{D}(\boldsymbol{\alpha}, \boldsymbol{\beta}^\star)\hat{R}(\Phi)\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{T}(\boldsymbol{\Upsilon})|0\rangle$$
$$= \exp\left(\mathrm{i}\Phi + \tfrac{1}{2}\operatorname{Tr}\boldsymbol{\Upsilon}\right)\hat{D}(\boldsymbol{\alpha}, \boldsymbol{\beta}^\star)\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)|0\rangle, \tag{30}$$

where we have used the parametrization given in (6).

Combining equations (16)–(18) and (12)–(14) with the identity $\boldsymbol{a}|0\rangle = 0$, we obtain the following set of eigenvalue equations for the generalized coherent state defined in (30):

$$\hat{G}\boldsymbol{a}\hat{G}^{-1}\hat{G}|0\rangle = \exp(-\boldsymbol{\Upsilon})\left[\boldsymbol{\Delta}\left(\boldsymbol{a} - \hbar^{-\frac{1}{2}}\boldsymbol{\alpha}\right) - \boldsymbol{\Omega}\left(\boldsymbol{a}^\dagger - \hbar^{-\frac{1}{2}}\boldsymbol{\beta}^\star\right)\right]|\chi\rangle = 0, \tag{31}$$

where the matrices $\boldsymbol{\Delta}$ and $\boldsymbol{\Omega}$ have been defined in (13*a*)–(13*d*)

Let us next derive the form of this coherent state in the coordinate representation. For this purpose, we shall use at this time the relation between the set $\{\boldsymbol{a}, \boldsymbol{a}^\dagger\}$ and the set $\{\hat{\boldsymbol{q}}, \hat{\boldsymbol{p}}\}$ given in (26). Consistently, we will also make use of the corresponding classical variables q_n and p_n (in vectorial notation $\boldsymbol{q}$ and $\boldsymbol{p}$) defined through the equation

$$\begin{pmatrix} \boldsymbol{\alpha} \\ \boldsymbol{\beta}^\star \end{pmatrix} = \frac{1}{\sqrt{2}}\begin{pmatrix} \boldsymbol{q} + \mathrm{i}\boldsymbol{p} \\ \boldsymbol{q} - \mathrm{i}\boldsymbol{p} \end{pmatrix}. \tag{32}$$

As in general we have $\boldsymbol{\alpha} \neq \boldsymbol{\beta}$, $\boldsymbol{q}$ and $\boldsymbol{p}$ may generally take complex values.

In the coordinate representation $|\boldsymbol{x}\rangle$ (i.e. $\hat{\boldsymbol{q}}\,|\boldsymbol{x}\rangle = \boldsymbol{x}\,|\boldsymbol{x}\rangle$), the generalized coherent state $\langle\boldsymbol{x}|\hat{G}|0\rangle$ will be a function of the coordinates $\boldsymbol{x}$ and of the N_G group parameters χ. By writing (31) in that representation, we arrive at the following set of differential equations:

$$\frac{\partial\langle\boldsymbol{x}|\hat{G}|0\rangle}{\partial\boldsymbol{x}} = \mathbf{f}_{\boldsymbol{x}}\langle\boldsymbol{x}|\hat{G}|0\rangle, \tag{33}$$

where $\mathbf{f}_{\boldsymbol{x}}$ is a vector field of dimension d depending on $\boldsymbol{x}$ and on the N_G group parameters χ. Furthermore, by writing down the relations given in (19) in the coordinate representation, we can obtain additional differential equations satisfied by the coherent state, namely

$$\frac{\partial\langle\boldsymbol{x}|\hat{G}|0\rangle}{\partial\chi} = \hat{\mathbf{f}}_\chi\langle\boldsymbol{x}|\hat{G}|0\rangle, \tag{34}$$

where $\hat{\mathbf{f}}_\chi$ is a differential operator vector field of dimension N_G depending on χ, $\boldsymbol{x}$, and $\hat{\boldsymbol{p}} = -\mathrm{i}\hbar\partial/\partial\boldsymbol{x}$. Since all these differential equations are linear in $\langle\boldsymbol{x}|\hat{G}|0\rangle$, they can be easily integrated. An arbitrary constant factor appears; we may fix it by requiring that the vacuum state $\langle\boldsymbol{x}|0\rangle$, be normalized and real. Then, by taking

$$\Phi = 0, \tag{35}$$

we finally obtain

$$\langle \boldsymbol{x}|\hat{G}|0\rangle = \left(\frac{1}{\pi\hbar}\right)^{d/4} \mathbb{N}_G^{-\frac{1}{2}} \exp\left[-\frac{1}{2\hbar}(\boldsymbol{x}-\boldsymbol{q})\boldsymbol{\Gamma}(\boldsymbol{x}-\boldsymbol{q}) + \frac{\mathrm{i}}{\hbar}\boldsymbol{p}\left(\boldsymbol{x}-\frac{1}{2}\boldsymbol{q}\right)\right], \tag{36}$$

where

$$\mathbb{N}_G = \det(\boldsymbol{G}_{23}+\boldsymbol{G}_{33}) = \mathrm{e}^{-\mathrm{Tr}\,\boldsymbol{\Upsilon}} \det(\boldsymbol{\Omega}+\boldsymbol{\Delta}), \tag{37}$$

and

$$\boldsymbol{\Gamma} = (\boldsymbol{G}_{33}-\boldsymbol{G}_{23})(\boldsymbol{G}_{23}+\boldsymbol{G}_{33})^{-1} = (\boldsymbol{\Omega}+\boldsymbol{\Delta})^{-1}(\boldsymbol{\Delta}-\boldsymbol{\Omega}). \tag{38}$$

The matrix $\boldsymbol{\Gamma}$ can be shown to be complex symmetric. Besides, if $\hat{G}$ is unitary, we also obtain

$$|\mathbb{N}_G| = (\det \boldsymbol{\Gamma}_{\mathrm{r}})^{-\frac{1}{2}}, \tag{39}$$

where $\boldsymbol{\Gamma}_{\mathrm{r}}$ is the real part of $\boldsymbol{\Gamma}$.

Let us proceed now to show how the previous results change if we use the two-photon Lie algebra associated with the set $\{\boldsymbol{a}', \boldsymbol{a}'^{\dagger}\}$, which was defined in (27), to set up the corresponding generalized coherent state. If $|0\rangle$ is the vacuum state for the vector operator $\boldsymbol{a}$, i.e. $\boldsymbol{a}|0\rangle = 0$, then $|0\rangle' = \hat{W}^{-1}|0\rangle$ will be the vacuum state for $\boldsymbol{a}'$; in effect, $\boldsymbol{a}'\hat{W}^{-1}|0\rangle = \hat{W}^{-1}\boldsymbol{a}|0\rangle = 0$. Therefore, the general form of the multimode coherent state with respect to the Lie group corresponding to $\{\boldsymbol{a}', \boldsymbol{a}'^{\dagger}\}$ may be defined, using (29), as

$$|\chi'\rangle' \equiv \hat{G}'(\chi')|0\rangle' = \hat{W}^{-1}\hat{G}(\chi')|0\rangle = \hat{G}(\chi)|0\rangle = |\chi\rangle\,. \tag{40}$$

Let us establish next the relationship between the parameter vectors χ' and χ. Using for both $\hat{G}(\chi)$ and $\hat{G}(\chi')$ the parametrization given in (6), the equalities in (40) then lead to the following identities:

$$\hat{W}^{-1}\hat{S}(\boldsymbol{\Xi}', \boldsymbol{\Pi}'^{\star})\hat{T}(\boldsymbol{\Upsilon}') = \hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^{\star})\hat{T}(\boldsymbol{\Upsilon}), \tag{41}$$

and

$$\hat{W}^{-1}\hat{D}(\boldsymbol{\alpha}', \boldsymbol{\beta}'^{\star})\hat{W} = \hat{D}(\boldsymbol{\alpha}, \boldsymbol{\beta}^{\star}). \tag{42}$$

These two equations provide the unprimed parameter set $\{\boldsymbol{\alpha}, \boldsymbol{\beta}^{\star}, \boldsymbol{\Xi}, \boldsymbol{\Pi}^{\star}, \boldsymbol{\Upsilon}\}$ in terms of the corresponding primed parameters. In particular, if $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}^{\star}$ are written as in (32), we have

$$\begin{pmatrix}\boldsymbol{\alpha}'\\ \boldsymbol{\beta}'^{\star}\end{pmatrix} = \frac{1}{\sqrt{2}}\boldsymbol{W}\begin{pmatrix}\boldsymbol{q}+\mathrm{i}\boldsymbol{p}\\ \boldsymbol{q}-\mathrm{i}\boldsymbol{p}\end{pmatrix}. \tag{43}$$

This equation is the classical analogue of (27). With these relations at hand, we conclude that the parameter vector χ in (40) represents precisely the set $\{\boldsymbol{\alpha}, \boldsymbol{\beta}^{\star}, \Phi, \boldsymbol{\Xi}, \boldsymbol{\Pi}^{\star}, \boldsymbol{\Upsilon}\}$ or, equivalently, the set $\{\boldsymbol{q}, \boldsymbol{p}, \Phi, \boldsymbol{G}\}$, with $\Phi = \Phi'$, and $\boldsymbol{G}$ being the $2d \times 2d$ central block of the representation matrix for $\hat{G}(\chi)$; for this block-matrix, we obtain

$$\boldsymbol{G} = \boldsymbol{W}^{-1}\boldsymbol{G}', \tag{44}$$

where $\boldsymbol{G}'$ is the $2d \times 2d$ matrix representation of $\hat{S}(\boldsymbol{\Xi}', \boldsymbol{\Pi}'^{\star})\hat{T}(\boldsymbol{\Upsilon}')$ (i.e. the $2d \times 2d$ central block of the matrix representation of $\hat{G}(\chi')$). Hence, in the coordinate representation we have $\langle \boldsymbol{x}|\chi'\rangle' = \langle \boldsymbol{x}|\chi\rangle$; thus the form of this wavefunction can be easily obtained by combining (36) and the relationship just established between the parameter vectors χ and χ'.

A useful alternative parametrization of the operator $\hat{G}(\chi)$, different from that used in (30) to define the coherent state, is given by

$$\hat{G}(\chi) = \hat{R}(\Phi)\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^{\star})\hat{T}(\boldsymbol{\Upsilon})\hat{D}(\boldsymbol{\gamma}, \boldsymbol{\delta}^{\star}), \tag{45}$$

where, with the help of the SFMR, one obtains

$$\begin{pmatrix}\boldsymbol{\alpha}\\ \boldsymbol{\beta}^{\star}\end{pmatrix} = \boldsymbol{S}\boldsymbol{T}\begin{pmatrix}\boldsymbol{\gamma}\\ \boldsymbol{\delta}^{\star}\end{pmatrix} = \boldsymbol{G}\begin{pmatrix}\boldsymbol{\gamma}\\ \boldsymbol{\delta}^{\star}\end{pmatrix}, \tag{46}$$

with $\boldsymbol{G}$ being, as usual, the $2d \times 2d$ central block of the representation matrix for $\hat{G}$. If we use the disentangling theorem

$$\hat{D}(\boldsymbol{\gamma}, \boldsymbol{\delta}^\star) = \exp\left(-\frac{\boldsymbol{\gamma}\boldsymbol{\delta}^\star}{2\hbar}\right) \exp\left(\hbar^{-\frac{1}{2}}\boldsymbol{\gamma}\boldsymbol{a}^\dagger\right) \exp\left(-\hbar^{-\frac{1}{2}}\boldsymbol{\delta}^\star\boldsymbol{a}\right), \tag{47}$$

then we obtain that the state

$$\exp\left(\frac{\boldsymbol{\gamma}\boldsymbol{\delta}^\star}{2\hbar}\right)\hat{G}(\boldsymbol{\chi})|0\rangle = \hat{G}_{\mathrm{u}}(\boldsymbol{\gamma}, \boldsymbol{\Xi}, \boldsymbol{\Pi}^\star, \boldsymbol{\Upsilon}, \Phi)|0\rangle, \tag{48}$$

where

$$\hat{G}_{\mathrm{u}}(\boldsymbol{\gamma}, \boldsymbol{\Xi}, \boldsymbol{\Pi}^\star, \boldsymbol{\Upsilon}, \Phi) = \hat{R}(\Phi)\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{T}(\boldsymbol{\Upsilon}) \exp\left(\hbar^{-\frac{1}{2}}\boldsymbol{\gamma}\boldsymbol{a}^\dagger\right), \tag{49}$$

is independent of $\boldsymbol{\delta}^\star$.[2] However, as equations (32) and (43) evidence, the choice of this parameter vector determines the values of the classical variables $\boldsymbol{q}$ and $\boldsymbol{p}$; in other words, we have many different forms to write the same state $\hat{G}_{\mathrm{u}}|0\rangle$ in the coordinate representation in terms of different values for those variables. The set of all possible $\boldsymbol{\delta}^\star$ values is equivalent to Heller and coworkers' initial ket manifold [2, 3]. This manifold can be embedded in a larger one if we make use of the general disentangling theorem

$$\begin{aligned} \hat{D}(\boldsymbol{\alpha}, \boldsymbol{\beta}^\star)\hat{R}(\Phi)\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{T}(\boldsymbol{\Upsilon}) &= \exp\left(\tfrac{1}{2}\boldsymbol{a}^\dagger\boldsymbol{\Psi}_{\mathrm{l}}\boldsymbol{a}^\dagger + \boldsymbol{\xi}_{\mathrm{l}}\boldsymbol{a}^\dagger\right) \\ &\times \exp\left[\boldsymbol{a}^\dagger\boldsymbol{\Psi}_{\mathrm{c}}\boldsymbol{a} + \tfrac{1}{2}(\mathrm{Tr}\,\boldsymbol{\Psi}_{\mathrm{c}})\hat{I} + \tfrac{1}{2}\varsigma\hat{I}\right] \exp\left(\tfrac{1}{2}\boldsymbol{a}\boldsymbol{\Psi}_{\mathrm{r}}\boldsymbol{a} + \boldsymbol{\xi}_{\mathrm{r}}\boldsymbol{a}\right). \end{aligned} \tag{50}$$

Hence, the state $\exp[-(\varsigma + \mathrm{Tr}\,\boldsymbol{\Psi}_{\mathrm{c}})/2]\hat{G}(\boldsymbol{\chi})|0\rangle$ will be independent of the parameter set $\{\boldsymbol{\xi}_{\mathrm{r}}, \boldsymbol{\Psi}_{\mathrm{r}}\}$ (the parameters ς and $\boldsymbol{\Psi}_{\mathrm{c}}$ are irrelevant since they can be always associated with complex phase-like terms). Within the TGA and GGWPD schemes, one can show that $\boldsymbol{\xi}_{\mathrm{r}}$ (or equivalently $\boldsymbol{\delta}^\star$) becomes the only relevant free parameter, but one can imagine other schemes in which the enlarged initial ket manifold could be very useful. We shall leave the analysis of this issue for a future work, and here just consider the original ket manifold corresponding to all possible $\boldsymbol{\delta}^\star$ values.

4. A perturbative solution to the time-dependent Schrödinger equation for an initial coherent state

The goal of this section is to obtain a formal solution of the time-dependent Schrödinger equation

$$\mathrm{i}\hbar\,\frac{\mathrm{d}}{\mathrm{d}t}|\Psi(t)\rangle = \hat{H}(t)|\Psi(t)\rangle, \tag{51}$$

for an initial Gaussian (i.e. coherent) state. For convenience, we shall take

$$|\Psi(0)\rangle = \exp\left(\frac{\boldsymbol{\gamma}\boldsymbol{\delta}^\star}{2\hbar}\right)\hat{G}(\boldsymbol{\chi})|0\rangle = \hat{G}_{\mathrm{u}}(\boldsymbol{\gamma}, \boldsymbol{\Xi}, \boldsymbol{\Pi}^\star, \boldsymbol{\Upsilon}, \Phi)|0\rangle, \tag{52}$$

with $\hat{G}_{\mathrm{u}}$ given in (49) and $\hat{G}$ given by any of the two equivalent forms provided by (6) and (45).

For simplicity, let us assume first that the quantum Hamiltonian operator of the multimode system, $\hat{H}(\boldsymbol{a}^\dagger, \boldsymbol{a}, t)$, is obtained as the normally ordered form of a classical Hamilton function $\mathcal{H}(\boldsymbol{\alpha}^\star, \boldsymbol{\alpha}, t)$, i.e.

$$\hat{H}(\boldsymbol{a}^\dagger, \boldsymbol{a}, t) = \hat{\mathcal{N}}\{\mathcal{H}(\boldsymbol{\alpha}^\star, \boldsymbol{\alpha}, t)\}, \tag{53}$$

[2] The state $\hat{G}_{\mathrm{u}}|0\rangle$ is always an unnormalized coherent state, even if $\hat{R}$, $\hat{S}$ and $\hat{T}$ are unitary. We use the subscript u for that reason.

where $\hat{\mathcal{N}}$ is the normal ordering operator, which replaces $\alpha_n^\star$ by $\hbar^{\frac{1}{2}} a_n^\dagger$ and α_n by $\hbar^{\frac{1}{2}} a_n$ with all $a_n^\dagger$ written to the left of all a_n. In this case, the classical Hamiltonian function is just the Q symbol (or Q phase space representation) of the quantum Hamiltonian,

$$\mathcal{H}(\boldsymbol{\alpha}^\star, \boldsymbol{\alpha}, t) = \mathcal{H}_Q(\boldsymbol{\alpha}^\star, \boldsymbol{\alpha}, t) \equiv \langle \boldsymbol{\alpha} | \hat{H}(t) | \boldsymbol{\alpha} \rangle, \tag{54}$$

where

$$|\boldsymbol{\alpha}\rangle = \hat{D}(\boldsymbol{\alpha}, \boldsymbol{\alpha}^\star)|0\rangle \tag{55}$$

is the one-photon coherent state. As the function $\mathcal{H}(\boldsymbol{\alpha}^\star, \boldsymbol{\alpha}, t)$, we can use without loss of generality its power series expansion

$$\mathcal{H}(\boldsymbol{\alpha}^\star, \boldsymbol{\alpha}, t) = \sum_{\boldsymbol{i},\boldsymbol{j}} c_{\boldsymbol{ij}}(t) \prod_{n=1}^{d} \alpha_n^{\star i_n} \alpha_n^{j_n}, \tag{56}$$

where $\boldsymbol{i}$ and $\boldsymbol{j}$ are d-vectors whose elements are indexes i_n and j_n taking values in the set of integer numbers equal or greater than zero, and $c_{\boldsymbol{ij}}(t)$ are complex coefficients. Hence, the quantum Hamiltonian becomes an expansion in the positive integer powers of the parameter $\hbar^{\frac{1}{2}}$, namely

$$\hat{H}(\boldsymbol{a}^\dagger, \boldsymbol{a}, t) = \sum_{\boldsymbol{i},\boldsymbol{j}} c_{\boldsymbol{ij}}(t) \prod_{n=1}^{d} \left(\hbar^{\frac{1}{2}} a_n^\dagger\right)^{i_n} \left(\hbar^{\frac{1}{2}} a_n\right)^{j_n}. \tag{57}$$

We write the solution of the TDSE corresponding to the initial state in (52) as $|\Psi(t)\rangle = \exp\left(\frac{\gamma\delta^\star}{2\hbar}\right)\hat{U}(t)\hat{G}|0\rangle$, where $\hat{U}(t)$ is the quantum propagator. By defining

$$\hat{U}_{\mathrm{g}}(t) = \hat{U}(t)\hat{G}, \tag{58}$$

this new propagator (we use the subscript g to remind its dependence on $\hat{G}$) is thus solution of the evolution equation

$$\mathrm{i}\hbar \frac{\mathrm{d}\hat{U}_{\mathrm{g}}(t)}{\mathrm{d}t} = \hat{H}(t)\hat{U}_{\mathrm{g}}(t), \tag{59}$$

with the initial condition

$$\hat{U}_{\mathrm{g}}(0) = \hat{G}(\chi) = \hat{R}(\Phi)\hat{D}(\boldsymbol{\alpha}, \boldsymbol{\beta}^\star)\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{T}(\boldsymbol{\Upsilon}), \tag{60}$$

where $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}^\star$ are related to $\boldsymbol{\gamma}$ and $\boldsymbol{\delta}^\star$ by (46); let us remind that while $\boldsymbol{\gamma}$ is a fixed parameter, we have total freedom to fix the arbitrary parameter vector $\boldsymbol{\delta}^\star$.

From section 2.3, we know that the quantum propagator for a system with a Hamiltonian that is at most quadratic in the creation and annihilation operators is an element of the corresponding multimode Lie group. Therefore, in systems where a local quadratic approximation might be valid, the previous result moves us to choose for $\hat{U}_{\mathrm{g}}(t)$ the ansatz

$$\hat{U}_{\mathrm{g}}(t) = \hat{G}(t)\hat{U}_{\mathrm{r}}(t), \tag{61}$$

where $\hat{G}(t)$ is a time-dependent element of the Lie group evolving from its initial value $\hat{G}(0) = \hat{G}(\chi)$. The operator $\hat{U}_{\mathrm{r}}(t)$ (the subscript r stands for remainder) is only constrained by the initial condition $\hat{U}_{\mathrm{r}}(0) = \hat{I}$, and is expected to remain close to this initial value if our previous assumption is correct.

The two main component in the time evolution of our initial coherent state will be a time-dependent overall phase and the motion of the state centre along a given path in phase space. This motion may be represented by means of a displacement operator $\hat{D}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star)$, and the operator $\hat{R}(\Phi_t)$ can account for the phase changes. Therefore, in order to accommodate these features within our scheme, and without loss of generality, we shall write

$$\hat{G}(t) = \hat{D}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star)\hat{R}(\Phi_t)\hat{G}_{\mathrm{q}}(t), \tag{62}$$

with

$$\begin{pmatrix} \boldsymbol{\mu}_0 \\ \boldsymbol{\nu}_0^\star \end{pmatrix} = \begin{pmatrix} \boldsymbol{\alpha} \\ \boldsymbol{\beta}^\star \end{pmatrix}, \tag{63}$$

and

$$\Phi_0 = \Phi, \tag{64}$$

and where $\hat{G}_{\rm q}(t)$ is another evolving Lie group element with initial value

$$\hat{G}_{\rm q}(0) = \hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{T}(\boldsymbol{\Upsilon}). \tag{65}$$

Hence, from (61) and (62)

$$\hat{U}_{\rm g}(t) = \hat{D}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star)\hat{R}(\Phi_t)\hat{G}_{\rm q}(t)\hat{U}_{\rm r}(t). \tag{66}$$

Substituting this last relation in (59), we obtain

$$\mathrm{i}\hbar\hat{G}_{\rm q}(t)\frac{\mathrm{d}\hat{U}_{\rm r}(t)}{\mathrm{d}t} = \left[\hat{D}^{-1}\hat{H}\hat{D} - \mathrm{i}\hbar\hat{D}^{-1}\frac{\mathrm{d}\hat{D}(t)}{\mathrm{d}t} + \hbar\frac{\mathrm{d}\Phi_t}{\mathrm{d}t}\right]\hat{G}_{\rm q}(t)\hat{U}_{\rm r}(t) - \mathrm{i}\hbar\frac{\mathrm{d}\hat{G}_{\rm q}(t)}{\mathrm{d}t}\hat{U}_{\rm r}(t). \tag{67}$$

From the properties of the generalized displacement operator given in (16) and (19), we have

$$\hat{D}^{-1}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star)\hat{H}(\boldsymbol{a}^\dagger, \boldsymbol{a}, t)\hat{D}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star) = \hat{H}\big(\boldsymbol{a}^\dagger + \hbar^{-\frac{1}{2}}\boldsymbol{\nu}_t^\star, \boldsymbol{a} + \hbar^{-\frac{1}{2}}\boldsymbol{\mu}_t, t\big), \tag{68}$$

and

$$\mathrm{i}\hbar\hat{D}^{-1}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star)\frac{\mathrm{d}\hat{D}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star)}{\mathrm{d}t} = \frac{1}{2}(\boldsymbol{\nu}_t^\star\dot{\boldsymbol{\mu}}_t - \dot{\boldsymbol{\nu}}_t^\star\boldsymbol{\mu}_t) + \hbar^{\frac{1}{2}}(\dot{\boldsymbol{\mu}}_t\boldsymbol{a}^\dagger - \dot{\boldsymbol{\nu}}^\star\boldsymbol{a}). \tag{69}$$

Performing now a Maclaurin expansion of the operator on the right-hand side of (68), we arrive at

$$\begin{aligned}\hat{H}\big(\boldsymbol{a}^\dagger + \hbar^{-\frac{1}{2}}\boldsymbol{\nu}_t^\star, \boldsymbol{a} + \hbar^{-\frac{1}{2}}\boldsymbol{\mu}_t, t\big) &= \mathcal{H}(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t) - \frac{\hbar}{2}\,\mathrm{Tr}\,\boldsymbol{H}_{\nu_t^\star\mu_t} \\ &\quad + \hbar^{\frac{1}{2}}\left[\frac{\partial\mathcal{H}(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t)}{\partial\boldsymbol{\nu}_t^\star}\boldsymbol{a}^\dagger + \frac{\partial\mathcal{H}(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t)}{\partial\boldsymbol{\mu}_t}\boldsymbol{a}\right] + \hat{H}_{\rm q} + \hat{H}_{\rm r},\end{aligned} \tag{70}$$

with $\hat{H}_{\rm q}$ being the quadratic symmetrically ordered Hamiltonian (thus the subscript q stands for quadratic)

$$\hat{H}_{\rm q} = \hbar\big(\boldsymbol{a}^\dagger\boldsymbol{H}_{\nu_t^\star\mu_t}\boldsymbol{a} + \tfrac{1}{2}\,\mathrm{Tr}\,\boldsymbol{H}_{\nu_t^\star\mu_t} + \tfrac{1}{2}\boldsymbol{a}^\dagger\boldsymbol{H}_{\nu_t^\star\nu_t^\star}\boldsymbol{a}^\dagger + \tfrac{1}{2}\boldsymbol{a}\boldsymbol{H}_{\mu_t\mu_t}\boldsymbol{a}\big), \tag{71}$$

where we have used $d \times d$ matrices $\boldsymbol{H}_{xy}$, with $\boldsymbol{x}, \boldsymbol{y} = \boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t$, whose elements are $(\boldsymbol{H}_{xy})_{mn} = \frac{\partial^2\mathcal{H}(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t)}{\partial x_m\partial y_n}$. In (70), the remainder $\hat{H}_{\rm r}$ has the following form:

$$\hat{H}_{\rm r}(\boldsymbol{a}^\dagger, \boldsymbol{a}, t) = {\sum_{\boldsymbol{i},\boldsymbol{j}}}' e_{\boldsymbol{i}\boldsymbol{j}}(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t)\prod_{n=1}^{d}\big(\hbar^{\frac{1}{2}}a_n^\dagger\big)^{i_n}\big(\hbar^{\frac{1}{2}}a_n\big)^{j_n}, \tag{72}$$

where $\sum_{\boldsymbol{i},\boldsymbol{j}}'$ is a restricted sum that excludes the terms with $\sum_{n=1}^{d}(i_n + j_n) < 3$. Therefore, the leading term in this $\hbar^{\frac{1}{2}}$-expansion of $\hat{H}_{\rm r}$ is $\mathrm{O}(\hbar^{3/2})$.

We now substitute equations (68) and (69) in (67) and choose the parameters $\boldsymbol{\nu}_t^\star$ and $\boldsymbol{\mu}_t$ so that the terms $\mathrm{O}(\hbar^{\frac{1}{2}})$ in this equation, which are linear in $\boldsymbol{a}^\dagger$ and $\boldsymbol{a}$, cancel out. This requirement leads to the following set of complex Hamilton equations:

$$\begin{pmatrix} \dot{\boldsymbol{\mu}}_t \\ \dot{\boldsymbol{\nu}}_t^\star \end{pmatrix} = \mathrm{i}\begin{pmatrix} -\partial\mathcal{H}(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t)/\partial\boldsymbol{\nu}_t^\star \\ \partial\mathcal{H}(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t)/\partial\boldsymbol{\mu}_t \end{pmatrix}, \tag{73}$$

which must be solved subject to the initial condition given in (63); as we know, the initial values $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}^\star$ are obtained from γ and the arbitrary $\boldsymbol{\delta}^\star$ by means of the transformation given in (46). In view of these results, we conclude that the phase space path chosen to be followed by the centre of our initial coherent state coincides with that of a generally complex classical trajectory.

The phase Φ_t in the operator $\hat{R}(\Phi_t)$ is next chosen so as to remove the operator-independent first two terms appearing in the expansion of the Hamiltonian given in (70) and the operator-independent term in (69) from the evolution equation (67); hence

$$\Phi_t = \Phi + \frac{\mathcal{F}}{2} + \frac{\mathcal{E}}{\hbar}, \tag{74}$$

with

$$\mathcal{F} = \int_0^t \mathrm{d}\tau \, \mathrm{Tr}\, \boldsymbol{H}_{\nu_\tau^\star \mu_\tau}, \tag{75}$$

and

$$\mathcal{E} = \int_0^t \mathrm{d}\tau \left[-\mathcal{H}(\boldsymbol{\nu}_\tau^\star, \boldsymbol{\mu}_\tau, \tau) + \frac{\mathrm{i}}{2}(\boldsymbol{\nu}_\tau^\star \dot{\boldsymbol{\mu}}_\tau - \dot{\boldsymbol{\nu}}_\tau^\star \boldsymbol{\mu}_\tau) \right]. \tag{76}$$

Once the O(1) and $\mathrm{O}(\hbar^{\frac{1}{2}})$ terms have been removed from (67), the operator $\hat{G}_{\mathrm{q}}(t)$ in (62) is then defined so that the remaining quadratic $\mathrm{O}(\hbar)$ terms in that equation vanish, which requires

$$\mathrm{i}\hbar \frac{\mathrm{d}\hat{G}_{\mathrm{q}}(t)}{\mathrm{d}t} = \hat{H}_{\mathrm{q}}(t)\hat{G}_{\mathrm{q}}(t). \tag{77}$$

This new evolution equation must be solved subject to the initial condition in (65). But since $\hat{H}_{\mathrm{q}}(t)$ is just quadratic in the creation and annihilation operators, the corresponding propagator, as we know from section 2.3, can also be given the form

$$\hat{G}_{\mathrm{q}}(t) = \hat{S}(\boldsymbol{\Xi}_t, \boldsymbol{\Pi}_t^\star)\hat{T}(\boldsymbol{\Upsilon}_t). \tag{78}$$

Then, by writing (77) in the SFMR, restricted to the central $2d \times 2d$ block, we obtain

$$\mathrm{i}\hbar \frac{\mathrm{d}\boldsymbol{G}_{\mathrm{q}}(t)}{\mathrm{d}t} = \boldsymbol{H}_{\mathrm{q}}(t)\boldsymbol{G}_{\mathrm{q}}(t), \tag{79}$$

with the initial condition

$$\boldsymbol{G}_{\mathrm{q}}(0) = \boldsymbol{G} = \begin{pmatrix} \boldsymbol{\Delta}\,\mathrm{e}^{\boldsymbol{\Upsilon}} & \boldsymbol{\Omega}\,\mathrm{e}^{-\widetilde{\boldsymbol{\Upsilon}}} \\ \boldsymbol{\Lambda}^\star\,\mathrm{e}^{\boldsymbol{\Upsilon}} & \widetilde{\boldsymbol{\Delta}}\,\mathrm{e}^{-\widetilde{\boldsymbol{\Upsilon}}} \end{pmatrix}. \tag{80}$$

But since, from (71),

$$\boldsymbol{H}_{\mathrm{q}}(t) = \hbar \begin{pmatrix} \boldsymbol{H}_{\nu_t^\star \mu_t} & \boldsymbol{H}_{\nu_t^\star \nu_t^\star} \\ -\boldsymbol{H}_{\mu_t \mu_t} & -\widetilde{\boldsymbol{H}}_{\nu_t^\star \mu_t} \end{pmatrix}, \tag{81}$$

i.e. $\boldsymbol{H}_{\mathrm{q}}(t)/\hbar$ is the second derivative matrix of the classical Hamiltonian function $\mathcal{H}(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t)$ along the classical trajectory $\{\boldsymbol{\nu}_\tau^\star, \boldsymbol{\mu}_\tau;\ 0 \leqslant \tau \leqslant t\}$, (79) is nothing but the classical evolution equation for the fundamental linear stability matrix associated with that trajectory. This fundamental stability matrix shall be called $\boldsymbol{U}_{\mathrm{q}}(t)$; it is known to satisfy the initial condition $\boldsymbol{U}_{\mathrm{q}}(0) = \boldsymbol{I}$, and can be written as the matrix of the coordinate transformation from the set $\{\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star\}$ to the set $\{\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star\}$, i.e.

$$\boldsymbol{U}_{\mathrm{q}}(t) = \frac{\partial(\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star)}{\partial(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star)}. \tag{82}$$

Therefore, the solution of (79), subject to the initial condition in (80), is just

$$\boldsymbol{G}_{\mathrm{q}}(t) = \boldsymbol{U}_{\mathrm{q}}(t)\boldsymbol{G}. \tag{83}$$

As comes out from the form just established for $\hat{G}_{\mathrm{q}}(t)$, this operator accounts for the changes in the quadratic Gaussian fluctuations of our evolving state.

With the choices made for $\hat{D}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^{\star})$, $\hat{R}(\Phi_t)$, and $\hat{G}_{\mathrm{q}}(t)$, equation (67) yields the following evolution equation for the remainder propagator $\hat{U}_{\mathrm{r}}(t)$,

$$\mathrm{i}\hbar \frac{\mathrm{d}\hat{U}_{\mathrm{r}}(t)}{\mathrm{d}t} = \hat{H}_{\mathrm{r}}^{\mathrm{I}}(t)\hat{U}_{\mathrm{r}}(t), \tag{84a}$$

$$\hat{U}_{\mathrm{r}}(0) = \hat{I}, \tag{84b}$$

where

$$\hat{H}_{\mathrm{r}}^{\mathrm{I}}(t) = \hat{G}_{\mathrm{q}}^{-1}(t)\hat{H}_{\mathrm{r}}(t)\hat{G}_{\mathrm{q}}(t). \tag{85}$$

For the solution of (84a) and (84b), we can write the following formal expansion:

$$\hat{U}_{\mathrm{r}}(t) = \hat{I} + \hat{C}(t), \tag{86a}$$

$$\hat{C}(t) \sim \sum_{l=1}^{\infty} \hat{U}_{\mathrm{r}}^{(l)}(t), \tag{86b}$$

where each term in this sum is given by

$$\hat{U}_{\mathrm{r}}^{(l)}(t) = \left(\frac{1}{\mathrm{i}\hbar}\right)^{l} \int_{t\geqslant\tau_n,\ldots,\tau_2\geqslant\tau_1\geqslant 0} \mathrm{d}\tau_n,\ldots,\mathrm{d}\tau_2\,\mathrm{d}\tau_1\,\hat{H}_{\mathrm{r}}^{\mathrm{I}}(\tau_n),\ldots,\hat{H}_{\mathrm{r}}^{\mathrm{I}}(\tau_2)\hat{H}_{\mathrm{r}}^{\mathrm{I}}(\tau_1). \tag{87}$$

The explicit form of $\hat{U}_{\mathrm{r}}^{(l)}(t)$ can be obtained from the expression for $\hat{H}_{\mathrm{r}}$ given in (72), the definitions of $\hat{H}_{\mathrm{r}}^{\mathrm{I}}$ and $\hat{G}_{\mathrm{q}}(t)$ given respectively in (85) and (78), and from (17) and (18); in normal order, this form should comply with the following general expression

$$\hat{U}_{\mathrm{r}}^{(l)} = \sum_{\boldsymbol{i},\boldsymbol{j}} u_{\boldsymbol{ij}}^{(l)} \prod_{n=1}^{d} a_n^{\dagger i_n} a_n^{j_n}, \tag{88}$$

with

$$u_{\boldsymbol{ij}}^{(l)} = \hbar^{N_{\boldsymbol{ij}}^{(l)}} \sum_{k\geqslant 0} u_{\boldsymbol{ij};k}^{(l)}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^{\star}, \boldsymbol{\Upsilon}, \boldsymbol{\mu}_0, \boldsymbol{\nu}_0^{\star}, t)\hbar^{k}, \tag{89}$$

where $u_{\boldsymbol{ij};k}^{(l)}$ are complex coefficients depending on time and on the initial state parameters, and

$$N_{\boldsymbol{ij}}^{(l)} = \begin{cases} (l + n_{\boldsymbol{ij}})/2 & \text{if } l_{\boldsymbol{ij}} \geqslant 3l \\ (l + n_{\boldsymbol{ij}} - 2\lfloor n_{\boldsymbol{ij}}/2\rfloor)/2 & \text{if } l_{\boldsymbol{ij}} < 3l, \end{cases} \tag{90}$$

where

$$l_{\boldsymbol{ij}} = \sum_{n=1}^{d}(i_n + j_n), \tag{91}$$

$n_{\boldsymbol{ij}} = |l_{\boldsymbol{ij}} - 3l|$, and $\lfloor n_{\boldsymbol{ij}}/2\rfloor$ gives the integer part of $n_{\boldsymbol{ij}}/2$. In light of these equations, we conclude that the expansion (86b) for the correction operator $\hat{C}(t)$ may include terms proportional to any positive integer power of $\hbar^{\frac{1}{2}}$.

The present analysis has been performed for a quantum Hamiltonian obtained from its classical counterpart following a normal-order scheme of quantization. Had we started from a Hamiltonian differently derived (e.g. by means of either the antinormal-order scheme or the Weyl–Wigner scheme), we could still write it down in normal order in a form similar to the expansion given in (57), but the coefficients c_{ij} would include now contributions proportional to integer powers of $\hbar$, namely

$$c_{ij}(t) = \sum_{k\geqslant 0} c_{ij;k}(t)\hbar^k. \tag{92}$$

This does not alter our previous analysis, and the only important detail to remark is that the classical Hamiltonian function from which the classical trajectories and all their required information are derived is, in all these cases, the smooth Q symbol, $\mathcal{H}_Q$, defined in (54).

Alternatively, we could have started from a form of a general quantum Hamiltonian written, for instance, as an expansion of either antinormally ordered or symmetrically ordered products of creation and annihilation operators (a one-parameter family of choices does indeed exist). For all these cases, (70) can then be generalized as

$$\begin{aligned}\hat{H}\big(\boldsymbol{a}^\dagger + \hbar^{-\frac{1}{2}}\boldsymbol{\nu}_t^\star, \boldsymbol{a} + \hbar^{-\frac{1}{2}}\boldsymbol{\mu}_t, t\big) &= \mathcal{H}_s(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t) - \hbar\frac{s}{2}\,\mathrm{Tr}\,\boldsymbol{H}_{s\,\nu_t^\star\mu_t} \\ &\quad + \hbar^{\frac{1}{2}}\left[\frac{\partial\mathcal{H}_s(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t)}{\partial\boldsymbol{\nu}_t^\star}\boldsymbol{a}^\dagger + \frac{\partial\mathcal{H}_s(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t)}{\partial\boldsymbol{\mu}_t}\boldsymbol{a}\right] + \hat{H}_{s\mathrm{q}} + \hat{H}_{s\mathrm{r}},\end{aligned} \tag{93}$$

in which $\mathcal{H}_s(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, t)$ is a general Hamiltonian classical symbol given by

$$\mathcal{H}_s(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}) \equiv \hat{H}\left[\left(\hbar^{-\frac{1}{2}}\boldsymbol{\beta}^\star + \frac{\hbar^{\frac{1}{2}}(s-1)}{2}\frac{\partial}{\partial\boldsymbol{\alpha}}\right), \left(\hbar^{-\frac{1}{2}}\boldsymbol{\alpha} + \frac{\hbar^{\frac{1}{2}}(s+1)}{2}\frac{\partial}{\partial\boldsymbol{\beta}^\star}\right), t\right]\cdot 1, \tag{94}$$

where s is a real parameter, $1 \geqslant s \geqslant -1$, whose value fixes the symbol or representation chosen as the classical Hamiltonian function; e.g. $s = 1$ corresponds to the Q symbol, $s = 0$ to the Weyl–Wigner symbol, and $s = -1$ to the P symbol. The differential operator on the right-hand side of this equation, which acts on the unit constant, is obtained by replacing $\boldsymbol{a}^\dagger$ and $\boldsymbol{a}$ in the original power series expansion of $\hat{H}(\boldsymbol{a}^\dagger, \boldsymbol{a}, t)$ by the specified differential operators [31]. The classical Hamiltonian $\mathcal{H}_s(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, t)$ can be generally expressed as the expansion provided in (56), but with coefficients $c_{ij}(t)$ that are $\hbar$-dependent, as those given in (92). The quadratic Hamiltonian $\hat{H}_{s\mathrm{q}}$ is like that displayed in (71), with the second derivative matrices obtained from $\mathcal{H}_s$, i.e. $(\boldsymbol{H}_{s\,xy})_{mn} = \frac{\partial^2\mathcal{H}_s(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t)}{\partial x_m \partial y_n}$ $(\boldsymbol{x}, \boldsymbol{y} = \boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t)$; finally, $\hat{H}_{s\mathrm{r}}$ is given by the general normally ordered form

$$\hat{H}_{s\mathrm{r}} = \sum_{\boldsymbol{i},\boldsymbol{j}} e_{\boldsymbol{ij}} \prod_{n=1}^{d} a_n^{\dagger i_n} a_n^{j_n}, \tag{95}$$

where

$$e_{\boldsymbol{ij}} = \hbar^{N_{\boldsymbol{ij}}} \sum_{k\geqslant 0} e_{\boldsymbol{ij};k}\hbar^k, \tag{96}$$

with

$$N_{\boldsymbol{ij}} = \begin{cases} l_{\boldsymbol{ij}}/2 & \text{if } l_{\boldsymbol{ij}} \geqslant 3 \\ 3/2 & \text{if } l_{\boldsymbol{ij}} = 1 \\ 2 & \text{if } l_{\boldsymbol{ij}} = 0, 2, \end{cases} \tag{97}$$

and $l_{\boldsymbol{ij}}$ having been defined in (91).

Our analysis can then be readily repeated for all these choices, and leads to the following general expression for the state $|\Psi(t)\rangle$ evolving from the initial state $|\Psi(0)\rangle$ given in (52),

$$|\Psi(t)\rangle = \exp\left(\frac{\gamma\delta^\star}{2\hbar}\right)\hat{G}(t)[\hat{I} + \hat{C}(t)]|0\rangle \equiv \hat{G}_{\mathrm{u}}(t)[\hat{I} + \hat{C}(t)]|0\rangle, \tag{98}$$

with

$$\hat{G}(t) = \hat{D}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star)\hat{R}(\Phi_t)\hat{G}_{\mathrm{q}}(t), \tag{99}$$

where

$$\Phi_t = \Phi + \frac{s\mathcal{F}}{2} + \frac{\mathcal{E}}{\hbar}, \tag{100}$$

with

$$\mathcal{F} = \int_0^t \mathrm{d}\tau \,\mathrm{Tr}\, \boldsymbol{H}_{s\nu_\tau^\star\mu_\tau}, \tag{101}$$

and

$$\mathcal{E} = \int_0^t \mathrm{d}\tau \left[-\mathcal{H}_s(\boldsymbol{\nu}_\tau^\star, \boldsymbol{\mu}_\tau, \tau) + \frac{\mathrm{i}}{2}(\boldsymbol{\nu}_\tau^\star\dot{\boldsymbol{\mu}}_\tau - \dot{\boldsymbol{\nu}}_\tau^\star\boldsymbol{\mu}_\tau)\right]. \tag{102}$$

The classical trajectory $\{\boldsymbol{\nu}_\tau^\star, \boldsymbol{\mu}_\tau;\ 0 \leqslant \tau \leqslant t\}$ required in these equations is a solution of the complex Hamilton equations

$$\begin{pmatrix}\dot{\boldsymbol{\mu}}_t \\ \dot{\boldsymbol{\nu}}_t^\star\end{pmatrix} = \mathrm{i}\begin{pmatrix}-\partial\mathcal{H}_s(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t)/\partial\boldsymbol{\nu}_t^\star \\ \partial\mathcal{H}_s(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t)/\partial\boldsymbol{\mu}_t\end{pmatrix}, \tag{103}$$

which must be solved subject to the initial condition

$$\begin{pmatrix}\boldsymbol{\mu}_0 \\ \boldsymbol{\nu}_0^\star\end{pmatrix} = \begin{pmatrix}\boldsymbol{\alpha} \\ \boldsymbol{\beta}^\star\end{pmatrix} = \boldsymbol{G}\begin{pmatrix}\boldsymbol{\gamma} \\ \boldsymbol{\delta}^\star\end{pmatrix}, \tag{104}$$

with $\boldsymbol{G}$ given in (80); here again, we have complete freedom to fix the value of $\boldsymbol{\delta}^\star$. The operator $\hat{G}_{\mathrm{q}}(t)$ in (99) is obtained from its restricted matrix representation, $\boldsymbol{G}_{\mathrm{q}}(t)$, which, as established by (83), is related to the fundamental stability matrix associated with the previous classical trajectory, i.e. $\boldsymbol{G}_{\mathrm{q}}(t)$ satisfies

$$\mathrm{i}\hbar\frac{\mathrm{d}\boldsymbol{G}_{\mathrm{q}}(t)}{\mathrm{d}t} = \boldsymbol{H}_{s\mathrm{q}}(t)\boldsymbol{G}_{\mathrm{q}}(t), \tag{105}$$

with $\boldsymbol{G}_{\mathrm{q}}(0) = \boldsymbol{G}$, as given in (80). The operator $\hat{C}(t)$ has always the form given in (86*b*) and (88), but its coefficients depend on the symbol s chosen for the classical Hamiltonian; i.e. the general form

$$\hat{H}_{s\mathrm{r}}^{\mathrm{I}}(t) = \hat{G}_{\mathrm{q}}^{-1}(t)\hat{H}_{s\mathrm{r}}(t)\hat{G}_{\mathrm{q}}(t) \tag{106}$$

should be used in (87) instead of $\hat{H}_{\mathrm{r}}^{\mathrm{I}}(t)$. Therefore, also in this general case, the expansion (86*b*) for $\hat{C}(t)$ includes terms proportional to any positive integer power of $\hbar^{\frac{1}{2}}$.

With the right choice for $\hat{C}(t)$, equation (86*a*) can be considered to yield an exact form for the propagator $\hat{U}_{\mathrm{r}}(t)$. However, the expansion supplied in (86*b*) and (87) for the correction operator $\hat{C}(t)$ may not generally converge, but just provide an asymptotic series for this operator. In such case, this series must be considered as a perturbative expansion for $\hat{C}(t)$; hence, it will be useful only if $\hat{H}_{s\mathrm{r}}^{\mathrm{I}}(t)$ behaves as a small enough perturbation. We will demonstrate in sections 6 and 7 that this requirement is not always satisfied in the semiclassical limit.

The scheme followed in this section can be readily extended to Hamiltonians $\hat{H}'(\boldsymbol{a}', \boldsymbol{a}'^\dagger)$ and their corresponding quantum propagators $\hat{U}'(t)$ written in terms of the more general pair of creation and annihilation vector operators $\{\boldsymbol{a}', \boldsymbol{a}'^\dagger\}$ defined in (27). In this case, our procedure leads straightforwardly to the following solution to the TDSE

$$\begin{aligned}|\boldsymbol{\chi}'\rangle_t' &= \hat{U}'(t)\hat{G}'(\boldsymbol{\chi}')|0\rangle' = \hat{U}'(t)\hat{G}(\boldsymbol{\chi})|0\rangle' \\ &= \hat{D}'(\boldsymbol{\mu}_t', \boldsymbol{\nu}_t'^\star)\hat{R}'(\Phi_t')\hat{G}_{\mathrm{q}}'(\boldsymbol{\Xi}_t', \boldsymbol{\Pi}_t'^\star, \boldsymbol{\Upsilon}_t')[\hat{I} + \hat{C}'(\boldsymbol{\mu}_t', \boldsymbol{\nu}_t'^\star, t)]|0\rangle',\end{aligned} \tag{107}$$

where we have used (40), and $|0\rangle' = \hat{W}^{-1}|0\rangle$ is the vacuum state for the set $\{\boldsymbol{a}', \boldsymbol{a}'^{\dagger}\}$, as it was introduced in section 3. As was established in section 2.3, all primed operators are functions of $\boldsymbol{a}'$ and $\boldsymbol{a}'^{\dagger}$; $\boldsymbol{\mu}'_t$ and $\boldsymbol{\nu}'^{\star}_t$ are the corresponding classical variables

$$\begin{pmatrix} \boldsymbol{\mu}'_t \\ \boldsymbol{\nu}'^{\star}_t \end{pmatrix} = \boldsymbol{W} \begin{pmatrix} \boldsymbol{\mu}_t \\ \boldsymbol{\nu}^{\star}_t \end{pmatrix} = \frac{1}{\sqrt{2}} \boldsymbol{W} \begin{pmatrix} \boldsymbol{q}_t + \mathrm{i}\boldsymbol{p}_t \\ \boldsymbol{q}_t - \mathrm{i}\boldsymbol{p}_t \end{pmatrix}. \tag{108}$$

These arise from the solution of the appropriate classical Hamilton equations corresponding to the classical Hamiltonian $\mathcal{H}'_s(\boldsymbol{\mu}', \boldsymbol{\nu}'^{\star}, t)$, which is the first term in the operator-ordering dependent expansion of $\hat{H}'(\boldsymbol{a}'^{\dagger} + \boldsymbol{\nu}'^{\star}_t/\hbar^{\frac{1}{2}}, \boldsymbol{a}' + \boldsymbol{\mu}'_t/\hbar^{\frac{1}{2}}, t)$, when such an expansion is performed in complete analogy with (93). As initial condition for $\boldsymbol{\mu}_t$ and $\boldsymbol{\nu}^{\star}_t$ we take that given in (104), so that

$$\begin{pmatrix} \boldsymbol{\mu}'_0 \\ \boldsymbol{\nu}'^{\star}_0 \end{pmatrix} = \boldsymbol{W} \begin{pmatrix} \boldsymbol{\alpha} \\ \boldsymbol{\beta}^{\star} \end{pmatrix} = \begin{pmatrix} \boldsymbol{\alpha}' \\ \boldsymbol{\beta}'^{\star} \end{pmatrix} = \boldsymbol{W}\boldsymbol{G} \begin{pmatrix} \boldsymbol{\gamma} \\ \boldsymbol{\delta}^{\star} \end{pmatrix} = \boldsymbol{G}' \begin{pmatrix} \boldsymbol{\gamma}' \\ \boldsymbol{\delta}'^{\star} \end{pmatrix}. \tag{109}$$

The operator $\hat{G}'_{\mathrm{q}}(t)$ in (107) is obtained from its $2d \times 2d$ matrix representation $\boldsymbol{G}'_{\mathrm{q}}(t)$ in the algebra associated with the set $\{\boldsymbol{a}', \boldsymbol{a}'^{\dagger}\}$; this matrix satisfies an evolution equation analogous to (105) , i.e.

$$\mathrm{i}\hbar \frac{\mathrm{d}\boldsymbol{G}'_{\mathrm{q}}(t)}{\mathrm{d}t} = \boldsymbol{H}'_{s\mathrm{q}}(t)\boldsymbol{G}'_{\mathrm{q}}(t), \tag{110}$$

with the initial condition

$$\boldsymbol{G}'_{\mathrm{q}}(0) = \boldsymbol{G}' = \begin{pmatrix} \boldsymbol{\Delta}' \,\mathrm{e}^{\boldsymbol{\Upsilon}'} & \boldsymbol{\Omega}' \,\mathrm{e}^{-\widetilde{\boldsymbol{\Upsilon}}'} \\ \boldsymbol{\Lambda}'^{\star} \,\mathrm{e}^{\boldsymbol{\Upsilon}'} & \widetilde{\boldsymbol{\Delta}}' \,\mathrm{e}^{-\widetilde{\boldsymbol{\Upsilon}}'} \end{pmatrix}. \tag{111}$$

In this evolution equation, $\boldsymbol{H}'_{s\mathrm{q}}$ is the $2d \times 2d$ matrix representation of the symmetrically ordered quadratic term $\hat{H}'_{s\mathrm{q}}$ in the above expansion of the Hamiltonian $\hat{H}'$ (i.e. the term analogous to $\hat{H}_{s\mathrm{q}}$ in (93)). The solution to (110) and (111) is

$$\boldsymbol{G}'_{\mathrm{q}}(t) = \boldsymbol{U}'_{\mathrm{q}}(t)\boldsymbol{G}', \tag{112}$$

which is written in terms of the fundamental stability matrix

$$\boldsymbol{U}'_{\mathrm{q}}(t) = \frac{\partial(\boldsymbol{\mu}'_t, \boldsymbol{\nu}'^{\star}_t)}{\partial(\boldsymbol{\mu}'_0, \boldsymbol{\nu}'^{\star}_0)} = \boldsymbol{W}\boldsymbol{U}_{\mathrm{q}}(t)\boldsymbol{W}^{-1}, \tag{113}$$

where $\boldsymbol{U}_{\mathrm{q}}(t)$ is the original stability matrix given in (82).

The operator $\hat{C}'(\boldsymbol{\mu}'_t, \boldsymbol{\nu}'^{\star}_t, t)$ in (107) is the analogue of $\hat{C}(t)$ in (98); it is associated with the operator $\hat{H}'_{s\mathrm{r}}$, which includes the non-quadratic terms in the previous expansion of the Hamiltonian. Finally, the phase Φ'_t has the form given in (100)–(102), with the substitution of $\boldsymbol{\mu}_t$ and $\boldsymbol{\nu}^{\star}_t$ by the new classical variables $\boldsymbol{\mu}'_t$ and $\boldsymbol{\nu}'^{\star}_t$, respectively; we thus have

$$\Phi'_t = \Phi + \frac{s\mathcal{F}'}{2} + \frac{\mathcal{E}'}{\hbar}, \tag{114}$$

with

$$\mathcal{F}' = \int_0^t \mathrm{d}\tau \,\mathrm{Tr}\, \boldsymbol{H}'_{s\nu'^{\star}_{\tau}\mu'_{\tau}}, \tag{115}$$

and

$$\begin{aligned} \mathcal{E}' &= \int_0^t \mathrm{d}\tau \left[-\mathcal{H}'_s(\boldsymbol{\nu}'^{\star}_{\tau}, \boldsymbol{\mu}'_{\tau}, \tau) + \frac{\mathrm{i}}{2}(\boldsymbol{\nu}'^{\star}_{\tau}\dot{\boldsymbol{\mu}}'_{\tau} - \dot{\boldsymbol{\nu}}'^{\star}_{\tau}\boldsymbol{\mu}'_{\tau}) \right] \\ &= \int_0^t \mathrm{d}\tau \left[-\mathcal{H}_s(\boldsymbol{\nu}^{\star}_{\tau}, \boldsymbol{\mu}_{\tau}, \tau) + \frac{\mathrm{i}}{2}(\boldsymbol{\nu}^{\star}_{\tau}\dot{\boldsymbol{\mu}}_{\tau} - \dot{\boldsymbol{\nu}}^{\star}_{\tau}\boldsymbol{\mu}_{\tau}) \right], \end{aligned} \tag{116}$$

where $\mathcal{H}'_s(\boldsymbol{\nu}'^{\star}_\tau, \boldsymbol{\mu}'_\tau, \tau) = \mathcal{H}_s(\boldsymbol{\nu}^{\star}_\tau, \boldsymbol{\mu}_\tau, \tau)$. The equality of the two forms for $\mathcal{E}'$ in (116) is a consequence of (108), which defines actually a canonical transformation between the variable sets $\{\boldsymbol{\mu}_t, \boldsymbol{\nu}^{\star}_t\}$ and $\{\boldsymbol{\mu}'_t, \boldsymbol{\nu}'^{\star}_t\}$. Then such an equality follows from the invariance of the Poincaré–Cartan 1-form under this kind of transformations.

Equation (108) is equivalent to the relationship

$$\hat{D}(\boldsymbol{\mu}_t, \boldsymbol{\nu}^{\star}_t) = \hat{W}^{-1}\hat{D}(\boldsymbol{\mu}'_t, \boldsymbol{\nu}'^{\star}_t)\hat{W}. \tag{117}$$

Making use of this equation and of the relation between unprimed and primed operators given in (28) and (29), equation (107) can be rewritten in the form

$$\begin{aligned} |\chi'\rangle'_t &= \hat{W}^{-1}\hat{U}(t)\hat{G}(\chi')|0\rangle \\ &= \hat{D}(\boldsymbol{\mu}_t, \boldsymbol{\nu}^{\star}_t)\hat{R}(\Phi_t)\hat{G}_{\mathrm{q}}(\boldsymbol{\Xi}_t, \boldsymbol{\Pi}^{\star}_t, \boldsymbol{\Upsilon}_t)[\hat{I} + \hat{C}(\boldsymbol{\mu}'_t, \boldsymbol{\nu}'^{\star}_t, t)]|0\rangle, \end{aligned} \tag{118}$$

where $\Phi_t = \Phi'_t$; here, we have made use of the relation

$$\hat{G}_{\mathrm{q}}(\boldsymbol{\Xi}_t, \boldsymbol{\Pi}^{\star}_t, \boldsymbol{\Upsilon}_t) = \hat{W}^{-1}\hat{G}_{\mathrm{q}}(\boldsymbol{\Xi}'_t, \boldsymbol{\Pi}'^{\star}_t, \boldsymbol{\Upsilon}'_t), \tag{119}$$

which can be established since both $\hat{W}^{-1}$ and $\hat{G}_{\mathrm{q}}$ belong to the subgroup $\{\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^{\star})\hat{T}(\boldsymbol{\Upsilon}); \forall\boldsymbol{\Xi}, \boldsymbol{\Pi}^{\star}, \boldsymbol{\Upsilon}\}$. The new parameter set $(\boldsymbol{\Xi}_t, \boldsymbol{\Pi}^{\star}_t, \boldsymbol{\Upsilon}_t)$ is obtained from the unique solution to this equation, which in $2d \times 2d$ matrix notation reads

$$\boldsymbol{G}_{\mathrm{q}}(t) = \boldsymbol{W}^{-1}\boldsymbol{G}'_{\mathrm{q}}(t). \tag{120}$$

By using (112) and (113), we then obtain from (120)

$$\boldsymbol{G}_{\mathrm{q}}(t) = \boldsymbol{W}^{-1}\boldsymbol{U}'_{\mathrm{q}}(t)\boldsymbol{G}' = \boldsymbol{U}_{\mathrm{q}}(t)\boldsymbol{W}^{-1}\boldsymbol{G}' = \boldsymbol{U}_{\mathrm{q}}(t)\boldsymbol{G}. \tag{121}$$

In summary, we have brought the general state $|\chi'\rangle'_t$ into our original form $|\chi\rangle_t = \hat{U}'(t)\hat{G}(\chi)|0\rangle = \hat{U}_{\mathrm{g}}(t)|0\rangle$, which is written in terms of the new parameter set $\chi(t) = \{\boldsymbol{\mu}_t, \boldsymbol{\nu}^{\star}_t, \Phi_t, \boldsymbol{\Xi}_t, \boldsymbol{\Pi}^{\star}_t, \boldsymbol{\Upsilon}_t\}$ or, equivalently, $\chi(t) = \{\boldsymbol{\mu}_t, \boldsymbol{\nu}^{\star}_t, \Phi_t, \boldsymbol{G}_{\mathrm{q}}(t)\}$.

5. Recovering the generalized Gaussian wave packet dynamics and the thawed Gaussian approximation

We shall show in this section how to recover both the thawed Gaussian approximation (TGA) and the generalized Gaussian wave packet dynamics (GGWPD) from the formal solution obtained in the previous section. The GGWPD, which contains the TGA as a particular case, is a local quadratic approximation that consists in truncating the perturbative expansion in (86*b*) at its leading zero order term, i.e. one assumes $\hat{C}(t) \simeq 0$. Hence, we have

$$\hat{U}_{\mathrm{g}}(t) \simeq \hat{G}(t) = \hat{D}(\boldsymbol{\mu}_t, \boldsymbol{\nu}^{\star}_t)\hat{R}(\Phi_t)\hat{G}_{\mathrm{q}}(t). \tag{122}$$

The initial ket manifold of the GGWPD would correspond to the complex space in which the arbitrary parameter vector $\boldsymbol{\delta}^{\star}$ takes all its possible values. As we know, in this general case the coordinates, $\boldsymbol{q}_t$, and momenta, $\boldsymbol{p}_t$, of the classical trajectory are defined as complex valued functions of time. On the other hand, the TGA is obtained for the particular choice $\boldsymbol{\delta} = \gamma$, which implies that the trajectory initial condition is

$$\begin{pmatrix} \boldsymbol{\mu}_0 \\ \boldsymbol{\nu}^{\star}_0 \end{pmatrix} = \begin{pmatrix} \boldsymbol{\alpha} \\ \boldsymbol{\alpha}^{\star} \end{pmatrix}, \tag{123}$$

and that $\boldsymbol{q}_t$ and $\boldsymbol{p}_t$ are real.

Therefore, in this local quadratic approximation the state evolving from the state $\hat{G}|0\rangle$ is just $\hat{U}(t)\hat{G}|0\rangle \simeq \hat{G}(t)|0\rangle$. Its form in the coordinate representation is obtained by substituting in (36) the time-dependent parameters obtained from equations (100) through (104) or the more general expressions given in (108)–(121). This yields

$$\langle \boldsymbol{x}|\hat{G}(t)|0\rangle = \left(\frac{1}{\pi\hbar}\right)^{d/4} \exp\left(\frac{\mathrm{i}s}{2}\mathcal{F}' + \frac{\mathrm{i}}{\hbar}\mathcal{A}\right)[\mathbb{N}_{G(t)}]^{-\frac{1}{2}}$$
$$\times \exp\left[-\frac{1}{2\hbar}(\boldsymbol{x}-\boldsymbol{q}_t)\boldsymbol{\Gamma}_t(\boldsymbol{x}-\boldsymbol{q}_t) + \frac{\mathrm{i}}{\hbar}\boldsymbol{p}_t\left(\boldsymbol{x}-\frac{1}{2}\boldsymbol{q}_t\right)\right], \tag{124}$$

where according to (35) we have chosen $\Phi = 0$; $\boldsymbol{q}_t$ and $\boldsymbol{p}_t$ are respectively the generally complex (real in the TGA) coordinates and momenta of the classical trajectory as generally defined in (108). Such a trajectory is a solution to the complex Hamilton equations given in (103) subject to either the general initial condition given in (104) (GGWPD choice) or the particular one given in (123) (TGA choice). The action integral, $\mathcal{A}$, in (124) is given by

$$\mathcal{A} = \int_0^t \mathrm{d}\tau \left[-\mathcal{H}_s(\boldsymbol{q}_\tau, \boldsymbol{p}_\tau, \tau) + \frac{1}{2}(\boldsymbol{p}_\tau\dot{\boldsymbol{q}}_\tau - \dot{\boldsymbol{p}}_\tau\boldsymbol{q}_\tau)\right], \tag{125}$$

where $\mathcal{H}_s(\boldsymbol{q}_\tau, \boldsymbol{p}_\tau, \tau)$ is the classical Hamiltonian written in terms of the chosen classical variables $\boldsymbol{q}_\tau$ and $\boldsymbol{p}_\tau$. The rest of the symbols in (124) are given by

$$\mathbb{N}_{G(t)} = \mathbb{N}_G \det\left[\frac{\partial(\boldsymbol{q}_t, \boldsymbol{p}_0)}{\partial(\boldsymbol{q}_0, \boldsymbol{p}_0)} + \mathrm{i}\frac{\partial(\boldsymbol{q}_t, \boldsymbol{q}_0)}{\partial(\boldsymbol{p}_0, \boldsymbol{q}_0)}\boldsymbol{\Gamma}\right], \tag{126}$$

$$\boldsymbol{\Gamma}_t = -\mathrm{i}\left[\frac{\partial(\boldsymbol{p}_t, \boldsymbol{p}_0)}{\partial(\boldsymbol{q}_0, \boldsymbol{p}_0)} + \mathrm{i}\frac{\partial(\boldsymbol{p}_t, \boldsymbol{q}_0)}{\partial(\boldsymbol{p}_0, \boldsymbol{q}_0)}\boldsymbol{\Gamma}\right]\left[\frac{\partial(\boldsymbol{q}_t, \boldsymbol{p}_0)}{\partial(\boldsymbol{q}_0, \boldsymbol{p}_0)} + \mathrm{i}\frac{\partial(\boldsymbol{q}_t, \boldsymbol{q}_0)}{\partial(\boldsymbol{p}_0, \boldsymbol{q}_0)}\boldsymbol{\Gamma}\right]^{-1}. \tag{127}$$

These last two equations have been written in terms of the $d \times d$ blocks of the $(\boldsymbol{p}, \boldsymbol{q})$-fundamental stability matrix. Finally the function $\mathcal{F}'$, which was defined in (115), can be equally written as

$$\mathcal{F}' = \frac{1}{2}\int_0^t \mathrm{d}\tau\, \mathrm{Tr}\left(\boldsymbol{W}_{qq}\boldsymbol{H}_{s_{q_\tau q_\tau}} + \boldsymbol{W}_{pp}\boldsymbol{H}_{s_{p_\tau p_\tau}} + 2\mathrm{i}\boldsymbol{W}_{qp}\boldsymbol{H}_{s_{q_\tau p_\tau}}\right), \tag{128}$$

where we have used the Hamiltonian second derivative matrices with elements $[\boldsymbol{H}_{sxy}]_{mn} = \frac{\partial^2\mathcal{H}_s}{\partial x_m \partial y_n}$ $(\boldsymbol{x}, \boldsymbol{y} = \boldsymbol{q}_t, \boldsymbol{p}_t)$, and in terms of the $d \times d$ blocks of the matrix $\boldsymbol{W}$ in (108) we have

$$\boldsymbol{W}_{qq} = \boldsymbol{I} + \widetilde{\boldsymbol{W}}_{32}\boldsymbol{W}_{23} + \widetilde{\boldsymbol{W}}_{23}\boldsymbol{W}_{32} - \widetilde{\boldsymbol{W}}_{32}\boldsymbol{W}_{22} - \widetilde{\boldsymbol{W}}_{23}\boldsymbol{W}_{33}, \tag{129a}$$

$$\boldsymbol{W}_{pp} = \boldsymbol{I} + \widetilde{\boldsymbol{W}}_{32}\boldsymbol{W}_{23} + \widetilde{\boldsymbol{W}}_{23}\boldsymbol{W}_{32} + \widetilde{\boldsymbol{W}}_{32}\boldsymbol{W}_{22} + \widetilde{\boldsymbol{W}}_{23}\boldsymbol{W}_{33}, \tag{129b}$$

$$\boldsymbol{W}_{qp} = \widetilde{\boldsymbol{W}}_{32}\boldsymbol{W}_{23} - \widetilde{\boldsymbol{W}}_{23}\boldsymbol{W}_{32} - \widetilde{\boldsymbol{W}}_{32}\boldsymbol{W}_{22} + \widetilde{\boldsymbol{W}}_{23}\boldsymbol{W}_{33}. \tag{129c}$$

Equation (124) provides the most general expression so far reported for the TGA in the coordinate representation. In Heller's original work [1] the phase $\frac{s\mathcal{F}'}{2}$ was absent (Weyl's choice). Particular values for this phase were considered by Baranger *et al* [11] in one-dimensional systems and by Pollak and Miret-Artés [15] in multidimensional ones.

In the following two sections, we will not be required to make an explicit choice for the relation between the operator set $\{\boldsymbol{a}, \boldsymbol{a}^\dagger\}$ and the set $\{\hat{\boldsymbol{q}}, \hat{\boldsymbol{p}}\}$, and thus between the classical variable sets $\{\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star\}$ and $\{\boldsymbol{q}_t, \boldsymbol{p}_t\}$. Therefore, the results and conclusions arising from our analysis shall be in this respect completely general.

6. Semiclassical asymptotic analysis of the correction term in the formal solution

In order to obtain the error in the TGA in the semiclassical limit, we have to analyse the correction operator $\hat{C}$ defined in (86*b*). Our goal consists in performing an analysis of its $\hbar$ dependence by rewriting its perturbative expansion, obtained in the previous section, as an expansion in powers of $\hbar$. This analysis is particularly simple in the generalized Bargmann representation [32]. For our state $|\Psi(t)\rangle = \hat{U}(t)|\Psi(0)\rangle$, where $|\Psi(0)\rangle = \hat{G}_{\mathrm{u}}|0\rangle = \hat{R}(\Phi)\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{T}(\boldsymbol{\Upsilon})\exp(\hbar^{-\frac{1}{2}}\boldsymbol{\zeta}\boldsymbol{a}^\dagger)|0\rangle$, we define this representation as the scalar product with the left state $\langle 0|\exp(\hbar^{-\frac{1}{2}}\boldsymbol{\eta}^\star\boldsymbol{a})\hat{T}^{-1}(\boldsymbol{\Upsilon})\hat{S}^{-1}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{R}^{-1}(\Phi)$, namely

$$\Psi(\boldsymbol{\eta}^\star, t) \equiv \langle 0|\exp\left(\hbar^{-\frac{1}{2}}\boldsymbol{\eta}^\star\boldsymbol{a}\right)\hat{T}^{-1}(\boldsymbol{\Upsilon})\hat{S}^{-1}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{U}(t)\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{T}(\boldsymbol{\Upsilon})\exp\left(\hbar^{-\frac{1}{2}}\boldsymbol{\zeta}\boldsymbol{a}^\dagger\right)|0\rangle. \tag{130}$$

Therefore, this Bargmann representation of the state $|\Psi(t)\rangle$ is a complex valued function of the complex variable $\boldsymbol{\eta}^\star$ with the parameters $\boldsymbol{\zeta}, \boldsymbol{\Xi}, \boldsymbol{\Pi}^\star$ and $\boldsymbol{\Upsilon}$ fixed by the initial state. Note that $\hat{T}^{-1}(\boldsymbol{\Upsilon})\hat{S}^{-1}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{U}(t)\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{T}(\boldsymbol{\Upsilon})$ corresponds to the evolution operator of the transformed Hamiltonian operator (not necessarily self-adjoint) $\hat{T}^{-1}(\boldsymbol{\Upsilon})\hat{S}^{-1}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{H}(t)\hat{S}(\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star)\hat{T}(\boldsymbol{\Upsilon})$, which takes, as we know from section 2, the same general form as $\hat{H}$. Hence we do not lose generality if we set to zero the parameter matrices $\boldsymbol{\Xi}, \boldsymbol{\Pi}^\star$ and $\boldsymbol{\Upsilon}$, and analyse the behaviour of the matrix element

$$\langle 0|\exp\left(\hbar^{-\frac{1}{2}}\boldsymbol{\beta}^\star\boldsymbol{a}\right)\hat{U}(t)\exp\left(\hbar^{-\frac{1}{2}}\boldsymbol{\alpha}\boldsymbol{a}^\dagger\right)|0\rangle \equiv (\boldsymbol{\beta}|\hat{U}(t)|\boldsymbol{\alpha}), \tag{131}$$

where

$$|\boldsymbol{\alpha}) = \exp\left(\hbar^{-\frac{1}{2}}\boldsymbol{\alpha}\boldsymbol{a}^\dagger\right)|0\rangle \tag{132}$$

is the standard Bargmann state, i.e., an unnormalized d-mode one-photon coherent state; the corresponding normalized state was defined in (55). From the disentangling theorem provided in (47), we obtain the relation $|\boldsymbol{\alpha}\rangle = \exp\left[-\frac{1}{2\hbar}(\boldsymbol{\alpha}^\star\boldsymbol{\alpha})\right]|\boldsymbol{\alpha})$. The magnitude given in (131) is then known as the Bargmann or coherent state representation of the quantum propagator. Substituting the formal solution of the TDSE derived in section 4 in (131), we obtain

$$(\boldsymbol{\beta}|\hat{U}(t)|\boldsymbol{\alpha}) = \langle 0|\exp\left(\hbar^{-\frac{1}{2}}\boldsymbol{\beta}^\star\boldsymbol{a}\right)\hat{G}_{\mathrm{u}}(\boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)[\hat{I} + \hat{C}(\boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)]|0\rangle, \tag{133}$$

with

$$\hat{G}_{\mathrm{u}}(\boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t) \equiv \exp\left(\frac{1}{2\hbar}\boldsymbol{\alpha}\boldsymbol{\nu}_0^\star\right)\hat{D}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star)\hat{R}(\Phi_t)\hat{G}_{\mathrm{q}}(t), \tag{134}$$

where Φ_t is defined in (100), and $\hat{G}_{\mathrm{q}}(t)$ is obtained from its restricted matrix representation, $\boldsymbol{G}_{\mathrm{q}}(t)$, which is given by (83) with $\boldsymbol{G}_{\mathrm{q}} = \boldsymbol{I}$; hence $\boldsymbol{G}_{\mathrm{q}}(t) = \boldsymbol{U}_{\mathrm{q}}(t)$. Note that $\boldsymbol{\mu}_0 = \boldsymbol{\alpha}$, and that $\boldsymbol{\nu}_0^\star$ plays now the role of the arbitrary parameter vector defining the initial ket manifold.

Whereas the operator $\hat{C}(t)$ in (133), whose perturbative expansion was given in (86*b*) and (88), acts on the vacuum state $|0\rangle$ and is written in normal order, the non-vanishing terms will be those corresponding to products of just creation operators; these correspond to null index vector $\boldsymbol{j}$ in (88), i.e.

$$\hat{C}(\boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)|0\rangle \sim \sum_{l=1}^{\infty}\sum_{\boldsymbol{i}}\hat{C}_{\boldsymbol{i}}^{(l)}(\boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)|0\rangle, \tag{135}$$

with

$$\hat{C}_{\boldsymbol{i}}^{(l)}(\boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t) = u_{\boldsymbol{i}0}^{(l)}(\boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)\prod_{n=1}^{d}a_n^{\dagger i_n}. \tag{136}$$

In light of these two equations, the operator vacuum expectation values needed in the evaluation of the right-hand side of (133) can all be derived from the generating function

$$\mathcal{G}_0(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t; \gamma) = \langle 0|\exp\left(\hbar^{-\frac{1}{2}}\boldsymbol{\beta}^\star \boldsymbol{a}\right)\hat{G}_{\mathrm{u}}(\boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)\exp(\boldsymbol{\gamma}\boldsymbol{a}^\dagger)|0\rangle. \quad (137)$$

The operator product on the right-hand side of this equation is an element of d-mode two-photon Lie group; thus its vacuum expectation value can be readily derived from its SFMR by means of (23). We obtain in this way

$$\begin{aligned}\mathcal{G}_0 = (\det \boldsymbol{U}_{\nu^\star\nu^\star})^{-\frac{1}{2}} \exp&\left\{\mathrm{i}\Phi_t + \frac{1}{2\hbar}(\boldsymbol{\nu}_0^\star\boldsymbol{\alpha} - \boldsymbol{\nu}_t^\star\boldsymbol{\mu}_t) + \frac{1}{\hbar}\boldsymbol{\beta}^\star\boldsymbol{\mu}_t\right\}\\ \times \exp&\left\{\frac{1}{2\hbar}(\boldsymbol{\nu}_t^\star - \boldsymbol{\beta}^\star)\boldsymbol{U}_{\mu\nu^\star}(\boldsymbol{U}_{\nu^\star\nu^\star})^{-1}(\boldsymbol{\nu}_t^\star - \boldsymbol{\beta}^\star)\right\}\\ \times \exp&\left\{-\frac{1}{\hbar^{\frac{1}{2}}}\gamma(\boldsymbol{U}_{\nu^\star\nu^\star})^{-1}(\boldsymbol{\nu}_t^\star - \boldsymbol{\beta}^\star) - \frac{1}{2}\gamma(\boldsymbol{U}_{\nu^\star\nu^\star})^{-1}\boldsymbol{U}_{\nu^\star\mu}\gamma\right\},\end{aligned} \quad (138)$$

in which we have denoted the blocks of the fundamental stability matrix $\boldsymbol{U}_{\mathrm{q}}(t)$ according to the expression

$$\boldsymbol{U}_{\mathrm{q}}(t) = \begin{pmatrix}\boldsymbol{U}_{\mu\mu} & \boldsymbol{U}_{\mu\nu^\star}\\ \boldsymbol{U}_{\nu^\star\mu} & \boldsymbol{U}_{\nu^\star\nu^\star}\end{pmatrix}. \quad (139)$$

Then we have, for instance,

$$\begin{aligned}&\langle 0|\exp\left(\hbar^{-\frac{1}{2}}\boldsymbol{\beta}^\star\boldsymbol{a}\right)\hat{G}_{\mathrm{u}}(\boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)\prod_{n=1}^{d} a_n^{\dagger i_n}|0\rangle\\ &\quad = \left[\frac{\partial^{i_d}}{\partial\gamma_d^{i_d}}\cdots\frac{\partial^{i_2}}{\partial\gamma_2^{i_2}}\frac{\partial^{i_1}}{\partial\gamma_1^{i_1}}\mathcal{G}_0(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t; \gamma)\right]_{\gamma=0}.\end{aligned} \quad (140)$$

Combining this equation and (136) yields

$$\langle 0|\exp\left(\hbar^{-\frac{1}{2}}\boldsymbol{\beta}^\star\boldsymbol{a}\right)\hat{G}_{\mathrm{u}}(\boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)\hat{C}_{\boldsymbol{i}}^{(l)}|0\rangle = \hbar^{N_{\boldsymbol{i}}^{(l)}} v_{\boldsymbol{i}}^{(l)}(t)\mathcal{G}_0(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t; \gamma = 0), \quad (141)$$

with

$$N_{\boldsymbol{i}}^{(l)} = \begin{cases}-l & \text{if } l_{\boldsymbol{i}} \geqslant 3l\\ -l + n_{\boldsymbol{i}} - \lfloor n_{\boldsymbol{i}}/2\rfloor & \text{if } l_{\boldsymbol{i}} < 3l,\end{cases} \quad (142)$$

and

$$v_{\boldsymbol{i}}^{(l)}(t) = \sum_{k\geqslant 0} v_{\boldsymbol{i};k}^{(l)}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)\hbar^k. \quad (143)$$

In these equations, $l_{\boldsymbol{i}} = l_{\boldsymbol{i0}} = \sum_{n=1}^{d} i_n$ (see equation (91)), $n_{\boldsymbol{i}} = |l_{\boldsymbol{i}} - 3l|$, and $\lfloor n_{\boldsymbol{i}}/2\rfloor$ gives the integer part of $n_{\boldsymbol{i}}/2$; $v_{\boldsymbol{i};k}^{(l)}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)$ are new complex coefficients, which can be written in terms of the old ones $u_{\boldsymbol{i}0;k}^{(l)}$, defined in (89). There will generally exist terms with $l_{\boldsymbol{i}} \geqslant 3l$.

Collecting all these results, we arrive finally at the following expression for the coherent state matrix element of the quantum propagator,

$$(\boldsymbol{\beta}|\hat{U}(t)|\boldsymbol{\alpha}) = \mathcal{U}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)[1 + \mathcal{C}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)], \quad (144)$$

where

$$\mathcal{U}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t) = \mathcal{G}_0(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t; \gamma = 0), \quad (145)$$

and each l-order term of the perturbative expansion for the correction term $\mathcal{C}(t)$,

$$\mathcal{C}(t) \sim \sum_{l=1}^{\infty}\mathcal{U}^{(l)}(t), \quad (146)$$

can be written in the following form:

$$\mathcal{U}^{(l)} = \hbar^{-l} \sum_{k\geqslant 0} w_k^{(l)}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)\hbar^k, \tag{147}$$

which shows explicitly its dependence on $\hbar$.

With the right choice for the correction term $\mathcal{C}$, equation (144) may be taken as an exact form for $(\boldsymbol{\beta}|\hat{U}(t)|\boldsymbol{\alpha})$. However, the $\hbar$ dependence of the perturbative expansion obtained for $\mathcal{C}$ and given by (146) and (147) implies that every term in this expansion will generally diverge in the small $\hbar$ limit; hence, even the exact form for $\mathcal{C}$ will not vanish in this limit; otherwise, the perturbative expansion, which is valid precisely for small $\mathcal{C}$, should vanish at least asymptotically as $\hbar$ goes to zero. In other words, since the remainder $\mathcal{C}$ does not generally vanish as $\hbar$ goes to zero, the expansion given by (146) and (147) does not provide an asymptotic series in the zero $\hbar$ limit; thus $\mathcal{U}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t)$ cannot be generally identified with its leading term as $\hbar$ tends to zero. However, we still have the freedom to fix the arbitrary parameter $\boldsymbol{\nu}_0^\star$. Let us consider first the TGA choice; in this case we have $\boldsymbol{\nu}_0^\star = \boldsymbol{\alpha}^\star$. Then one can readily check that, in general, the negative powers of $\hbar$ remain in our perturbative expansion of the correction term, implying that the TGA does not provides generally the leading semiclassical asymptotic contribution to the quantum time evolution of an initial coherent state. This statement does not mean that the TGA will always be a bad approximation. As discussed at the end of section 4, under the right circumstances, $\hat{H}^{\mathrm{I}}_{sr}(t)$ may behave as a small enough perturbation, so that the corresponding perturbative series becomes, at least, an asymptotic expansion in this perturbation limit. Nonetheless, as discussed earlier, the negative $\hbar$ powers in the correction term imply that this situation is not generally going to take place in the semiclassical limit, where the TGA should thus be a poor approximation.

Interestingly, there exists a particular choice of the $\boldsymbol{\nu}_0^\star$ parameter vector for which all the coefficients $w_k^{(l)}$ in (147) with $k < l + \lfloor(l+1)/2\rfloor$ vanish, so that the perturbative series for $\mathcal{C}(t)$ in (146) becomes an $\hbar$-expansion starting at first order. Namely, $\boldsymbol{\nu}_0^\star$ must be chosen such that $\boldsymbol{\nu}_t^\star = \boldsymbol{\beta}^\star$. The previous assertion then follows from the particular form of $\mathcal{G}_0(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\nu}_0^\star, t; \gamma)$ (see (138)) in this case: the derivatives of this generating function required in (140) will not provide $\hbar^{-\frac{1}{2}}$ powers, and the contribution in (141) from all terms $\hat{C}_i^{(l)}$ with l_i odd vanish. The required condition for $\boldsymbol{\nu}_t^\star$, together with that for $\boldsymbol{\mu}_0$, define the two-time boundary condition

$$\begin{pmatrix} \boldsymbol{\mu}_0 \\ \boldsymbol{\nu}_t^\star \end{pmatrix} = \begin{pmatrix} \boldsymbol{\alpha} \\ \boldsymbol{\beta}^\star \end{pmatrix}. \tag{148}$$

For this choice, which corresponds to a particular case of the GGWPD, the expansion in (144) and (147) has as its leading term

$$\mathcal{U} = \mathcal{U}_{\mathrm{c}} = \mathcal{J}^{-\frac{1}{2}} \exp\left[\frac{\mathrm{i}s}{2}\mathcal{F}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, t) + \frac{\mathrm{i}}{\hbar}\mathcal{S}(\boldsymbol{\alpha}^\star, \boldsymbol{\beta}, t)\right], \tag{149}$$

with

$$\mathcal{J}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}) = \mathcal{J}(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_0, t) = \det\left[\frac{\partial(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_0, t)}{\partial(\boldsymbol{\nu}_0^\star, \boldsymbol{\mu}_0, t)}\right] = \det\left[\frac{\partial(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_0, t)}{\partial(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t, t)}\right], \tag{150a}$$

$$\mathcal{F}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, t) = \mathcal{F}(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_0, t) = \int_0^t \mathrm{d}\tau \,\mathrm{Tr}\, \boldsymbol{H}_{s\nu^\star\mu}, \tag{150b}$$

$$\mathcal{S}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, t) = \mathcal{S}(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_0, t) = -\mathrm{i}\boldsymbol{\nu}_t^\star\boldsymbol{\mu}_t + \int_0^t [-\mathcal{H}_s(\boldsymbol{\nu}_\tau^\star, \boldsymbol{\mu}_\tau) + \mathrm{i}\dot{\boldsymbol{\mu}}_\tau\boldsymbol{\nu}_\tau^\star]\,\mathrm{d}\tau. \tag{150c}$$

The identification $(\boldsymbol{\beta}|\hat{U}(t)|\boldsymbol{\alpha}) \sim \mathcal{U}_{\mathrm{c}}$ is the well-known semiclassical approximation for the coherent states matrix elements of the quantum propagator [4, 6–12]. The correction term in this case admits therefore the following expansion:

$$\mathcal{C}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, t) = \mathcal{C}_{\mathrm{c}}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, t) \sim \sum_{l=1}^{\infty} \sum_{k \geqslant l + \lfloor (l+1)/2 \rfloor} w_k^{(l)}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, t)\hbar^{k-l}$$
$$= \sum_{k=1}^{\infty} w_k(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, t)\hbar^k, \tag{151}$$

so that we have finally

$$(\boldsymbol{\beta}|\hat{U}(t)|\boldsymbol{\alpha}) \sim \mathcal{U}_{\mathrm{c}}[1 + \mathcal{C}_{\mathrm{c}}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, t)]. \tag{152}$$

As argued in [12] the solution to the two-time boundary condition given in (148) does not have to be unique—many and even infinite physical and unphysical branches are possible. In such cases, if $\mathcal{U}_{\mathrm{c}}$ in (149) wants to represent the semiclassical transition amplitude $(\boldsymbol{\beta}|\hat{U}(t)|\boldsymbol{\alpha})$, one should sum the contributions from all the physical branches. As time t goes to zero only one of these branches turns out to produce a non negligible contribution with the right $t = 0$ limit $(\boldsymbol{\beta}|\boldsymbol{\alpha})$ for such transition amplitude. On the other hand, the factor $\mathcal{U}_{\mathrm{c}}$ in (149) and the full expansion for $\mathcal{C}_{\mathrm{c}}$ given in (151) should be evaluated for just one of these branches; this fact is a strong indication that, in general, this formal series for $(\boldsymbol{\beta}|\hat{U}(t)|\boldsymbol{\alpha})$ may not converge when $\hbar$ tends to zero. As a sensible rule then, one should choose the branch that provides the least divergent expansion for the given time t (the optimal branch may depend on time); in the semiclassical limit, this expansion will be, hopefully, asymptotically valid.

In summary, we have shown that for a right choice of the $\boldsymbol{\nu}_0^\star$ parameter vector (i.e. of the point in Heller's initial-ket manifold), the perturbative expansion of the coherent-state matrix element of the quantum propagator provides an $\hbar$-asymptotic expansion whose leading term corresponds to a branch of the well-known semiclassical approximation for that matrix element. However, for other choices of $\boldsymbol{\nu}_0^\star$, such a perturbative expansion is not in general an $\hbar$-asymptotic expansion, so that its correction term $\mathcal{C}$ does not vanish in the zero $\hbar$ limit and, consequently, its leading term $\mathcal{U}$ does not provide the correct semiclassical approximation to the propagator coherent-state matrix element.

An illustrative example of these issues is provided by the nonlinear Kerr Hamiltonian $\hat{H} = \frac{1}{2}\hbar^2 a^{\dagger 2} a^2$, for which we are going to evaluate its propagator matrix element $\mathcal{A} = \langle 0|\hat{U}(t)|\alpha\rangle$, whose exact value is $\mathcal{A} = \mathrm{e}^{-J/(2\hbar)}$, by means of the previous approximations. Let us take the Q-symbol of $\hat{H}$ as the classical Hamiltonian, i.e. $\mathcal{H} = (\alpha^\star\alpha)^2/2$, for which $J = \alpha^\star\alpha$ is a constant of the motion. Then, if we use the TGA to propagate $|\alpha\rangle$, we obtain $\mathcal{A}_{\mathrm{TGA}} = \mathrm{e}^{-J/(2\hbar)}$ for $Jt \ll 1$, and $\mathcal{A}_{\mathrm{TGA}} = (1 + \mathrm{i}Jt)^{-1/2} \exp[\mathrm{i}Jt/2 + (\mathrm{i}J^2t/2 - J)/\hbar]$ for $Jt \gg 1$. On the other hand, if we use the GGWPD asymptotic result, we obtain the exact value for all time t. Therefore, we can observe that for short time (much smaller than a period of the classical motion), where the TGA is expected to be valid, it provides indeed a good result. But for longer times, the approximation fails to provide an $\hbar$-asymptotically valid result, since the relative error $(\mathcal{A} - \mathcal{A}_{\mathrm{TGA}})/\mathcal{A}_{\mathrm{TGA}}$ does not tend to zero as $\hbar$ goes to zero, but it diverges exponentially.

Similar asymptotic analysis can be carried out for the expectation value $\langle\Psi(t)|\hat{O}|\Psi(t)\rangle/\langle\Psi(0)|\Psi(0)\rangle$ of operators, $\hat{O}$, having a well-defined classical limit. If one expands these operators as done with the Hamiltonian in (57) and (92), and makes use of the formal solution derived in section 4, then one can readily prove that, in contrast with our previous results, the correct $\hbar$-asymptotic expansion is obtained only for TGA choice, $\boldsymbol{\nu}_0^\star = \boldsymbol{\alpha}^\star$, and not for other choices of this arbitrary parameter.

Before concluding this section, we should mention that other kind of approximations different from the TGA, but which like the TGA use real classical trajectories, have been recently derived [33] for the mixed matrix element $\langle \boldsymbol{x}|\hat{U}(t)|\boldsymbol{\alpha}\rangle$. In principle, the present formalism could be used to analyse the corresponding correction terms and to determine their validity conditions.

7. Semiclassical asymptotic analysis of the correction term to the TGA IVR

In constructing the TGA IVR, one starts out from the generalized coherent state resolution of the identity operator

$$\hat{I} = \int \frac{\mathrm{d}\zeta\,\mathrm{d}\zeta^\star}{(2\pi\mathrm{i}\hbar)^d}\,\mathrm{e}^{-\zeta^\star\zeta/\hbar}\hat{G}_{\mathrm{u}}(\zeta, \Xi, \boldsymbol{\Pi}^\star, \boldsymbol{\Upsilon}, \Phi)|0\rangle\langle 0|\hat{G}_{\mathrm{u}}^\dagger(\zeta, -\Xi^\star, -\boldsymbol{\Pi}, -\Xi^\star, -\widetilde{\boldsymbol{\Upsilon}}^\star, \Phi^\star). \quad (153)$$

where according to (49)

$$\hat{G}_{\mathrm{u}}(\zeta, \Xi, \boldsymbol{\Pi}^\star, \boldsymbol{\Upsilon}, \Phi)|0\rangle = \exp\left(\mathrm{i}\Phi + \tfrac{1}{2}\operatorname{Tr}\boldsymbol{\Upsilon}\right)\hat{S}(\Xi, \boldsymbol{\Pi}^\star)\exp\left(\hbar^{-\frac{1}{2}}\zeta\boldsymbol{a}^\dagger\right)|0\rangle. \quad (154)$$

In (153), the integral measure is understood as the classical phase space measure $\mathrm{d}\zeta\,\mathrm{d}\zeta^\star \equiv \prod_{n=1}^{d}\mathrm{i}\,\mathrm{d}p_n\,\mathrm{d}q_n$, written in terms of the coordinates q_n and generalized momenta p_n of the modes. As in the previous section, we do not lose generality and gain simplicity by setting to zero the parameter matrices Ξ and $\boldsymbol{\Pi}^\star$; thus we shall just take the usual resolution

$$\hat{I} = \int \frac{\mathrm{d}\zeta\,\mathrm{d}\zeta^\star}{(2\pi\mathrm{i}\hbar)^d}\,\mathrm{e}^{-\zeta^\star\zeta/\hbar}\,|\zeta)\,(\zeta|\,, \quad (155)$$

where $|\zeta)$ is a Bargmann state as that defined in (132).

In these expressions, the integral over the phase space (q_n, p_n) of each mode can also be understood as an integral in the complex manifold defined by the two complex variables ζ_n and $\zeta_n^\star$, these being considered independent of each other. Integration takes place then in a particular two-dimensional surface $\Gamma_{\zeta_n\zeta_n^\star}$ parametrized by the phase space variables q_n and p_n; in this surface ζ_n and $\zeta_n^\star$ are complex conjugates of each other. If the function to be integrated satisfies some boundedness and analyticity criteria, $\Gamma_{\zeta_n\zeta_n^\star}$ can be conveniently deformed. Hence, under these conditions, we could also write

$$\hat{I} = \int_{\Gamma_{\zeta,\eta^\star}} \frac{\mathrm{d}\zeta\,\mathrm{d}\boldsymbol{\eta}^\star}{(2\pi\mathrm{i}\hbar)^d}\,\mathrm{e}^{-\eta^\star\zeta/\hbar}\,|\zeta)\,(\boldsymbol{\eta}|\,, \quad (156)$$

where $\Gamma_{\zeta,\eta^\star} = \prod_{n=1}^{d}\Gamma_{\zeta_n\eta_n^\star}$.

Acting now with the quantum propagator, $\hat{U}(t)$, on the left-hand side of (156) and using the formal solution obtained in section 4, we obtain

$$\hat{U}(t) = \int_{\Gamma_{\eta,\zeta^\star}} \frac{\mathrm{d}\zeta\,\mathrm{d}\boldsymbol{\eta}^\star}{(2\pi\hbar\mathrm{i})^d}\,\mathrm{e}^{-\eta^\star\zeta/\hbar}\hat{G}_{\mathrm{u}}(\boldsymbol{\eta}, \boldsymbol{\nu}_0^\star, t)[\hat{I} + \hat{C}(\boldsymbol{\eta}, \boldsymbol{\nu}_0^\star, t)]|0\rangle\,(\boldsymbol{\eta}|\,, \quad (157)$$

where $\hat{G}_{\mathrm{u}}(\zeta, \boldsymbol{\nu}_0^\star, t)$ has the form given in (134), with the substitution of $\boldsymbol{\alpha}$ by ζ; for instance, we have now

$$\boldsymbol{\mu}_0 = \zeta. \quad (158)$$

However, note that for each value of ζ and $\boldsymbol{\eta}^\star$ on the integration manifold $\Gamma_{\zeta,\eta^\star}$, we have to fix the arbitrary $\boldsymbol{\nu}_0^\star$ parameter vector, i.e.

$$\boldsymbol{\nu}_0^\star = \boldsymbol{\nu}_0^\star(\zeta, \boldsymbol{\eta}^\star) = \boldsymbol{\nu}_0^\star(\boldsymbol{\mu}_0, \boldsymbol{\eta}^\star). \quad (159)$$

Therefore, if we perform in the integral of (157) the change of variables $(\zeta, \boldsymbol{\eta}^\star) \to (\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star)$, we obtain

$$\hat{U}(t) = \int_{\Gamma_{\mu_0,\nu_0^\star}} \frac{\mathrm{d}\boldsymbol{\mu}_0\,\mathrm{d}\boldsymbol{\nu}_0^\star}{(2\pi\hbar\mathrm{i})^d}\,\mathrm{e}^{-\eta^\star\zeta/\hbar}\det\left[\frac{\partial(\boldsymbol{\nu}_0^\star, \boldsymbol{\mu}_0)}{\partial(\boldsymbol{\eta}^\star, \boldsymbol{\mu}_0)}\right]^{-1}\hat{G}_{\mathrm{u}}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t)[\hat{I} + \hat{C}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t)]|0\rangle\,(\boldsymbol{\eta}|\,, \quad (160)$$

where $\boldsymbol{\eta}^\star$ is a function of both $\boldsymbol{\mu}_0$ and $\boldsymbol{\nu}_0^\star$, i.e. $(\boldsymbol{\eta}| = \langle 0|\exp\left[\hbar^{-\frac{1}{2}}\boldsymbol{\eta}^\star(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star)\boldsymbol{a}\right]$.

We can write (160) as

$$\hat{U}(t) = \hat{U}_{\mathrm{TGA}}(t) + \hat{C}_{\mathrm{TGA}}(t), \tag{161}$$

where $\hat{U}_{\mathrm{TGA}}(t)$ and $\hat{C}_{\mathrm{TGA}}(t)$ correspond to the contributions to $\hat{U}(t)$ coming, respectively, from the terms proportional to $\hat{I}$ and $\hat{C}$ in the integrand of (160). Then $\hat{U}_{\mathrm{TGA}}(t)$ is a generalized form of the TGA IVR, and $\hat{C}_{\mathrm{TGA}}(t)$ would correspond to its correction term; the latter is related to the correction operator defined by Pollak and coworkers [15, 27–29], as will be shown later in this section.

We will not explore here the different formulations of the TGA arising from the freedom one has in the choice of the function $\boldsymbol{\nu}_0^\star(\boldsymbol{\zeta}, \boldsymbol{\eta}^\star)$ in (160), but we shall just fix it as in the standard TGA IVR, namely

$$\boldsymbol{\nu}_0^\star = \boldsymbol{\eta}^\star. \tag{162}$$

With this choice, we can write

$$\hat{U}_{\mathrm{TGA}}(t) = \int_{\Gamma_{\mu_0,\nu_0^\star}} \frac{\mathrm{d}\boldsymbol{\mu}_0\,\mathrm{d}\boldsymbol{\nu}_0^\star}{(2\pi\hbar\mathrm{i})^d}\,\mathrm{e}^{-\nu_0^\star\mu_0/\hbar}\hat{G}_{\mathrm{u}}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t)|0\rangle\,(\boldsymbol{\nu}_0|\,, \tag{163}$$

$$\hat{C}_{\mathrm{TGA}}(t) = \int_{\Gamma_{\mu_0,\nu_0^\star}} \frac{\mathrm{d}\boldsymbol{\mu}_0\,\mathrm{d}\boldsymbol{\nu}_0^\star}{(2\pi\hbar\mathrm{i})^d}\,\mathrm{e}^{-\nu_0^\star\mu_0/\hbar}\hat{G}_{\mathrm{u}}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t)\hat{C}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t)|0\rangle\,(\boldsymbol{\nu}_0|\,. \tag{164}$$

Note that (163) provides a generalized form of the TGA IVR propagator in which the initial phase space manifold $\Gamma_{\mu_0,\nu_0^\star}$ can be arbitrarily defined. The common choice for $\Gamma_{\mu_0,\nu_0^\star}$ is just the real phase space ($\boldsymbol{\mu}_0 = \boldsymbol{\nu}_0$), but (163) allows for the use of general complex trajectories and thus opens new possibilities worth exploring.

Let us now focus our attention in the semiclassical asymptotic analysis of $\hat{U}_{\mathrm{TGA}}(t)$ and $\hat{C}_{\mathrm{TGA}}(t)$ as defined in these two equations. As in the previous section, without loss of generality, we will make use of the coherent state representation. Combining (163), (164) and (144) we obtain

$$(\boldsymbol{\beta}|\hat{U}_{\mathrm{TGA}}(t)|\boldsymbol{\alpha}) = \int_{\Gamma_{\mu_0,\nu_0^\star}} \frac{\mathrm{d}\boldsymbol{\mu}_0\,\mathrm{d}\boldsymbol{\nu}_0^\star}{(2\pi\hbar\mathrm{i})^d}\mathcal{G}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t), \tag{165}$$

$$(\boldsymbol{\beta}|\hat{C}_{\mathrm{TGA}}(t)|\boldsymbol{\alpha}) = \int_{\Gamma_{\mu_0,\nu_0^\star}} \frac{\mathrm{d}\boldsymbol{\mu}_0\,\mathrm{d}\boldsymbol{\nu}_0^\star}{(2\pi\hbar\mathrm{i})^d}\mathcal{G}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t)\mathcal{C}(\boldsymbol{\beta}^\star, \boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t), \tag{166}$$

where

$$\mathcal{G}(\boldsymbol{\beta}^\star, \boldsymbol{\alpha}, \boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t) = \mathrm{e}^{-\nu_0^\star\mu_0/\hbar}\,\mathrm{e}^{\nu_0^\star\alpha/\hbar}\mathcal{G}_0(\boldsymbol{\beta}^\star, \boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t; \gamma = 0), \tag{167}$$

with the function $\mathcal{G}_0(\boldsymbol{\beta}^\star, \boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t; \gamma)$ given in (138), and $\mathcal{C}(\boldsymbol{\beta}^\star, \boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t)$ being the correction term whose perturbative expansion is provided in (146) and (147). Let us rewrite $\mathcal{G}(\boldsymbol{\beta}^\star, \boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t; \gamma)$ in the following useful form:

$$\mathcal{G} = (\det \boldsymbol{U}_{\nu^\star,\nu^\star})^{-\frac{1}{2}} \exp\left[\frac{\mathrm{i}s\mathcal{F}}{2} + \frac{\mathrm{i}}{\hbar}\mathcal{R}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_0^\star) + \frac{1}{\hbar}(\boldsymbol{\beta}^\star\boldsymbol{\mu}_t + \boldsymbol{\nu}_0^\star\boldsymbol{\alpha}) + \frac{1}{\hbar}\mathcal{Q}\right], \tag{168}$$

with

$$\mathcal{Q} = \tfrac{1}{2}(\boldsymbol{\nu}_t^\star - \boldsymbol{\beta}^\star)\boldsymbol{U}_{\mu\nu^\star}\boldsymbol{U}_{\nu^\star\nu^\star}^{-1}(\boldsymbol{\nu}_t^\star - \boldsymbol{\beta}^\star), \tag{169}$$

and a new action integral

$$\mathcal{R}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_0^\star, t) = \mathrm{i}\boldsymbol{\nu}_0^\star\boldsymbol{\mu}_0 + \int_0^t [-\mathcal{H}_s(\boldsymbol{\nu}_\tau^\star, \boldsymbol{\mu}_\tau) + \mathrm{i}\dot{\boldsymbol{\mu}}_\tau\boldsymbol{\nu}_\tau^\star]\,\mathrm{d}\tau, \tag{170}$$

which depends only on $\boldsymbol{\mu}_t$ and $\boldsymbol{\nu}_0^\star$, and satisfies

$$\mathrm{i}\,\frac{\partial \mathcal{R}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_0^\star, t)}{\partial \boldsymbol{\mu}_t} = -\boldsymbol{\nu}_t^\star, \tag{171a}$$

$$\mathrm{i}\,\frac{\partial \mathcal{R}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_0^\star, t)}{\partial \boldsymbol{\nu}_0^\star} = -\boldsymbol{\mu}_0, \tag{171b}$$

$$\frac{\partial \mathcal{R}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_0^\star, t)}{\partial t} = -\mathcal{H}_s(\boldsymbol{\nu}_t^\star, \boldsymbol{\mu}_t). \tag{171c}$$

We shall next proceed to evaluate the integrals in (165) and (166) by the saddle-point method. For convenience, before this, we shall perform the change of variables from the set $\{\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star\}$ to the set $\{\boldsymbol{\mu}_t, \boldsymbol{\nu}_0^\star\}$; thus we have $\mathrm{d}\boldsymbol{\mu}_0\,\mathrm{d}\boldsymbol{\nu}_0^\star = \mathrm{d}\boldsymbol{\mu}_t\,\mathrm{d}\boldsymbol{\nu}_0^\star(\det \boldsymbol{U}_{\mu\mu})^{-1}$. The new integration manifold $\Gamma_{\mu_t,\nu_0^\star}$ should then be deformed so as to include the stationary points of the rapidly varying exponent in $\mathcal{G}$. These stationary points determine the value of each of these two integrals (in (166) each term in the expansion of $\mathcal{C}(\boldsymbol{\beta}^\star, \boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t)$, as given in (146) and (147), is assumed to be a slowly varying function within such a manifold) . Differentiating the term proportional to $1/\hbar$ in the exponent of $\mathcal{G}$ with respect to $\boldsymbol{\mu}_t$ and $\boldsymbol{\nu}_0^\star$, and making use of (171a) and (171b), we arrive at the saddle-point conditions

$$\boldsymbol{\beta}^\star - \boldsymbol{\nu}_t^\star + \frac{\partial \mathcal{Q}}{\partial \boldsymbol{\mu}_t} = 0, \tag{172a}$$

$$\boldsymbol{\alpha} - \boldsymbol{\mu}_0 + \frac{\partial \mathcal{Q}}{\partial \boldsymbol{\nu}_0^\star} = 0. \tag{172b}$$

Since $\mathcal{Q}$ has the form given in (169), a particular solution to these equations is given by

$$\begin{pmatrix} \boldsymbol{\mu}_0 \\ \boldsymbol{\nu}_t^\star \end{pmatrix} = \begin{pmatrix} \boldsymbol{\alpha} \\ \boldsymbol{\beta}^\star \end{pmatrix}, \tag{173}$$

which is a two-time boundary condition fixing the saddle-point classical trajectory. As we know from section 6, for this trajectory, all coefficients corresponding to the zero and negative powers of $\hbar$ in the perturbative expansion of the correction term $\mathcal{C}$ vanish; hence, these terms do not contribute to the asymptotic expansion around this saddle point. We shall call this kind of stationary point a regular saddle point. However, there may exist in general other kind of solutions to (172a), (172b), in which the behaviour of the function $\mathcal{Q}$ plays a crucial role. These shall be called spurious saddle points, and they do generally exist in nonlinear systems. Although their existence had been noticed earlier [13], these spurious saddle points have been considered to be unimportant and thus disregarded in the asymptotic evaluation of the integrals in (165) and (166). However, since at these stationary points both positive and negative powers of $\hbar$ are going to appear in the perturbative expansion of $\mathcal{C}$, their contribution in the semiclassical limit is expected to be far from negligible. We shall soon verify this hypothesis.

Before going any further, we shall mention that not all the stationary points should be included in the stationary phase approximation, but only their subset called physical saddle points. This is closely related to the Stokes phenomenon [34–36]. The choice of the physical saddles is going to depend on the location of the stokes and antistokes lines (hypersurfaces in higher dimension). It is very unlikely that this location be such that all the spurious saddle points should be discarded. Furthermore, these stationary points change the whole analytic structure, which might as well affect the subset of the physical regular branches.

For a physical regular saddle point, we can easily evaluate the determinant of the second derivatives required in the saddle-point approximation; this determinant can be written in terms of determinants of the $d \times d$ blocks of the fundamental stability matrix $\boldsymbol{U}_{\mathrm{q}}$. With all these results at hand, we readily arrive at the following expressions:

$$(\beta|\hat{U}_{\mathrm{TGA}}(t)|\boldsymbol{\alpha}) = \mathcal{U}_{\mathrm{sc}}\big(1 + \mathcal{U}_{\mathrm{r}}^{(\mathrm{u})}\big), \tag{174a}$$

$$(\beta|\hat{C}_{\mathrm{TGA}}(t)|\boldsymbol{\alpha}) = \mathcal{U}_{\mathrm{sc}}\big(\mathcal{C}_{\mathrm{sc}} + \mathcal{U}_{\mathrm{r}}^{(\mathrm{c})}\big) = \mathcal{U}_{\mathrm{sc}}\mathcal{C}_{\mathrm{TGA}}, \tag{174b}$$

where $\mathcal{U}_{\mathrm{sc}}$ would correspond to $\mathcal{U}_{\mathrm{c}}$, the latter having been defined in (149), and $\mathcal{C}_{\mathrm{sc}}$, to the function $\mathcal{C}_{\mathrm{c}}$ given in (151). If the solution to the two-time boundary condition (173) is not unique, a sum over contributions $\mathcal{U}_{\mathrm{c}}$ and $\mathcal{C}_{\mathrm{c}}$ from the physical regular branches may be assumed in the determination of both $\mathcal{U}_{\mathrm{sc}}$ in (174*a*) and $\mathcal{C}_{\mathrm{sc}}$ in (174*b*); this sum is what makes $\mathcal{U}_{\mathrm{sc}}$ and $\mathcal{C}_{\mathrm{sc}}$ distinct from $\mathcal{U}_{\mathrm{c}}$ and $\mathcal{C}_{\mathrm{c}}$, since the latter are evaluated for just one of those solutions. $\mathcal{U}_{\mathrm{sc}}$ and $\mathcal{C}_{\mathrm{sc}}$ provide, respectively, the leading contribution to the integrals in (165) and (166) coming only from the physical regular saddle points; $\mathcal{U}_{\mathrm{r}}^{(\mathrm{u})}$ and $\mathcal{U}_{\mathrm{r}}^{(\mathrm{c})}$ are the remainders, which are determined by the analytic structure link to the spurious saddle points.

Let us remind at this point that $\mathcal{U}_{\mathrm{sc}}$ is the correct asymptotic semiclassical approximation to the Bargmann representation of the exact quantum propagator [12]. This asymptotic contribution is thus contained in the Bargmann representation of the TGA IVR propagator, but in this case $\mathcal{U}_{\mathrm{sc}}$ is only link to the physical regular saddle points, that is, there exists still an additional term, i.e. the remainder $\mathcal{U}_{\mathrm{r}}^{(\mathrm{u})}$ in (174*a*), whose asymptotic limit has to be carefully analysed, since this limit is going to be determined by the spurious saddle points. The effect of these stationary points is particularly dramatic in the behaviour of the other remainder $\mathcal{U}_{\mathrm{r}}^{(\mathrm{c})}$, and therefore in $\mathcal{C}_{\mathrm{TGA}}$. This conclusion arises from the analysis carried out in the previous section; namely, one can easily show that each term in the perturbative expansion of the correction term $\mathcal{C}$ given in (147) at any of the spurious saddle points will generally diverge as $\hbar$ goes to zero, which totally invalidates such an expansion in the semiclassical limit. Consequently, the remainder $\mathcal{C}_{\mathrm{TGA}}$, whose value is determined by the behaviour of $\mathcal{C}$ at these spurious saddles, cannot vanish asymptotically in the semiclassical limit whenever such stationary points do indeed exist. We shall next proceed to prove that the remainder $\mathcal{U}_{\mathrm{r}}^{(\mathrm{u})}$ is going to present as well this property, which implies that, contrarily to what has been always assumed, $\mathcal{U}_{\mathrm{sc}}$ *is not generally the correct asymptotic semiclassical expression for the Bargmann representation of the TGA IVR propagator*.

The proof is quite straightforward. Summing up equations (174*a*) and (174*b*), we obtain

$$(\beta|\hat{U}(t)|\boldsymbol{\alpha}) = (\beta|\hat{U}_{\mathrm{TGA}}(t) + \hat{C}_{\mathrm{TGA}}(t)|\boldsymbol{\alpha}) = \mathcal{U}_{\mathrm{sc}}\big(1 + \mathcal{C}_{\mathrm{TGA}} + \mathcal{U}_{\mathrm{r}}^{(\mathrm{u})}\big). \tag{175}$$

But, on the other hand, we can also write the following expression of the Bargmann transition amplitude in terms of its leading semiclassical asymptotic form:

$$(\beta|\hat{U}(t)|\boldsymbol{\alpha}) = \mathcal{U}_{\mathrm{sc}}(1 + \mathcal{C}_{\mathrm{r}}), \tag{176}$$

where $\mathcal{C}_{\mathrm{r}}$ is the corresponding remainder, which vanishes asymptotically in the semiclassical limit. Then from the last two equations we obtain

$$\mathcal{U}_{\mathrm{r}}^{(\mathrm{u})} = \mathcal{C}_{\mathrm{r}} - \mathcal{C}_{\mathrm{TGA}}. \tag{177}$$

But $\mathcal{C}_{\mathrm{r}}$ vanish asymptotically in the zero $\hbar$ limit. However, since $\mathcal{C}_{\mathrm{TGA}}$ will not generally vanish asymptotically in that limit, so will occur with $\mathcal{U}_{\mathrm{r}}^{(\mathrm{u})}$. This proves our statement, i.e. $\mathcal{U}_{\mathrm{sc}}$ cannot be generally taken as the leading semiclassical asymptotic contribution to the Bargmann representation of the TGA IVR.

From (174a), (176) and (177), we deduce that $\mathcal{C}_{\text{TGA}}$ can be identified with the error associated with the TGA IVR relative to the semiclassical value, i.e.

$$\mathcal{C}_{\text{TGA}} = \frac{(\beta|\hat{U}(t) - \hat{U}_{\text{TGA}}(t)|\alpha)}{\mathcal{U}_{\text{sc}}}, \tag{178}$$

and that the error relative to the TGA value is

$$\frac{(\beta|\hat{U}(t) - \hat{U}_{\text{TGA}}(t)|\alpha)}{(\beta|\hat{U}_{\text{TGA}}(t)|\alpha)} = \frac{\mathcal{C}_{\text{TGA}}}{1 + \mathcal{C}_{\text{r}} - \mathcal{C}_{\text{TGA}}}, \tag{179}$$

thus, these errors, as we know, do not vanish asymptotically in the semiclassical limit if the spurious saddle points contribute to $\mathcal{C}_{\text{TGA}}$; again, the TGA IVR propagator, $\hat{U}_{\text{TGA}}(t)$, is not generally a valid semiclassical propagator from a rigorous asymptotic analysis.

We will conclude this section with an extension of our analysis to the IVR scheme proposed by Pollak and coworkers [27–29], when this is applied to the particular case of the TGA IVR. These authors represent the exact quantum propagator as an expansion series in terms of a correction operator $\hat{C}_{\text{p}}(t)$, which, for the TGA IVR and in our notation, can be written as

$$\hat{C}_{\text{p}}(t) = \int_{\Gamma_{\mu_0,\nu_0^\star}} \frac{\mathrm{d}\boldsymbol{\mu}_0\,\mathrm{d}\boldsymbol{\nu}_0^\star}{(2\pi\hbar\mathrm{i})^d}\,\mathrm{e}^{-\nu_0^\star\mu_0/\hbar}\,\hat{E}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t)|0\rangle\,(\boldsymbol{\nu}_0|\,, \tag{180}$$

with

$$\begin{aligned}\hat{E}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t) &= -\hat{D}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star)\hat{H}_{sr}(t)\hat{D}^{-1}(\boldsymbol{\mu}_t, \boldsymbol{\nu}_t^\star)\hat{G}_{\text{u}}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t)\\ &= -\hat{G}_{\text{u}}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t)\hat{H}^{\text{I}}_{sr}(t),\end{aligned} \tag{181}$$

where $\hat{H}^{\text{I}}_{sr}(t)$ was defined in (106). The operator $\hat{E}$ is related to $\hat{G}_{\text{u}}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t)$ (e.g. (134)) and the Hermitian Hamiltonian $\hat{H}(t)$ through the equation

$$\mathrm{i}\hbar\frac{\mathrm{d}\hat{G}_{\text{u}}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t)}{\mathrm{d}t} = \hat{H}(t)\hat{G}_{\text{u}}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t) + \hat{E}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^\star, t). \tag{182}$$

Pollak and coworkers [27–29] provide a formal solution for the propagator $\hat{U}(t)$ in terms of $\hat{C}_{\text{p}}(t)$, which we write in slightly different but equivalent form as

$$\hat{U}(t) = \hat{U}_0(t) + \frac{\mathrm{i}}{\hbar}\int_0^t \mathrm{d}t'\,\hat{U}^\dagger(t', t)\hat{C}_{\text{p}}(t'), \tag{183}$$

where $\hat{U}_0(t) = \hat{U}_{\text{TGA}}(t)$ (as defined in (163)) is easily shown to satisfy the equation

$$\mathrm{i}\hbar\frac{\mathrm{d}\hat{U}_0(t)}{\mathrm{d}t} = \hat{H}(t)\hat{U}_0(t) + \hat{C}_{\text{p}}(t), \tag{184}$$

and $\hat{U}(t, t') = \hat{U}^\dagger(t', t)$, with $\hat{U}(t) \equiv \hat{U}(t, 0)$, is the exact propagator thus satisfying

$$\mathrm{i}\hbar\frac{\mathrm{d}\hat{U}(t, t')}{\mathrm{d}t} = \hat{H}(t)\hat{U}(t, t'), \tag{185a}$$

$$\mathrm{i}\hbar\frac{\mathrm{d}\hat{U}(t, t')}{\mathrm{d}t'} = -\hat{U}(t, t')\hat{H}(t'), \tag{185b}$$

$$\hat{U}(t, t) = \hat{I}. \tag{185c}$$

The solution to (184) is then written as the expansion [27–29]

$$\hat{U}(t) = \sum_{l=0}^{\infty}\hat{U}_l(t) \tag{186}$$

with

$$\hat{U}_{l+1}(t) = \frac{\mathrm{i}}{\hbar}\int_0^t \mathrm{d}t'\, \hat{U}_l^{\dagger}(t', t)\hat{C}_{\mathrm{p}}(t'). \tag{187}$$

Let us evaluate the first correction

$$\hat{U}_1(t) = \frac{\mathrm{i}}{\hbar}\int_0^t \mathrm{d}t'\, \hat{U}_0^{\dagger}(t', t)\hat{C}_{\mathrm{p}}(t'), \tag{188}$$

in which we will use the TGA form for $\hat{U}_0(t', t)$. This is given by an expression similar to (163) for an initial time t and final time t'. Since $t' \leqslant t$, this TGA requires back propagation of classical trajectories; the Hermitian conjugated form of this expression is then used in (188). Substituting later in this equation the expression for $\hat{C}_{\mathrm{p}}(t')$ given in (180) and the expression for $\hat{G}_{\mathrm{u}}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^{\star}, t)$ given in (134), we arrive at

$$\hat{U}_1(t) = \frac{1}{\mathrm{i}\hbar}\int_{\Gamma_{\mu_0,\nu_0^{\star}}} \frac{\mathrm{d}\boldsymbol{\mu}_0\, \mathrm{d}\boldsymbol{\nu}_0^{\star}}{(2\pi\hbar\mathrm{i})^d}\, \mathrm{e}^{-\nu_0^{\star}\mu_0/\hbar}\int_0^t \mathrm{d}t'\, \hat{U}_0^{\dagger}(t', t)\hat{G}_{\mathrm{u}}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^{\star}, t')\hat{H}_{sr}^{\mathrm{I}}(t')|0\rangle\langle\boldsymbol{\nu}_0|. \tag{189}$$

But according to (106),

$$\hat{U}_{\mathrm{r}}^{(1)}(t) = \frac{1}{\mathrm{i}\hbar}\int_0^t \mathrm{d}t'\hat{H}_{sr}^{\mathrm{I}}(t') \tag{190}$$

is just the first term in the series expansion of the correction operator $\hat{C}(t)$ (see (86*b*) and (87)). Thus performing an integration by parts of the time integral in (189), with $\hat{H}_{sr}^{\mathrm{I}}(t')$ being the easily integrable factor, we obtain

$$\hat{U}_1(t) = U^{(1)}(t) + \frac{1}{\mathrm{i}\hbar}\int_0^t \mathrm{d}t'\big[\hat{C}_{\mathrm{p}}^{\dagger}(t', t)U^{(1)}(t') - \hat{U}_0^{\dagger}(t', t)\hat{C}_{\mathrm{p}}^{(1)}(t')\big], \tag{191}$$

where $\hat{C}_{\mathrm{p}}(t', t)$ is the correction operator corresponding to $\hat{U}_0(t', t)$,

$$\hat{U}^{(1)}(t) = \int_{\Gamma_{\mu_0,\nu_0^{\star}}} \frac{\mathrm{d}\boldsymbol{\mu}_0\, \mathrm{d}\boldsymbol{\nu}_0^{\star}}{(2\pi\hbar\mathrm{i})^d}\, \mathrm{e}^{-\nu_0^{\star}\mu_0/\hbar}\hat{G}_{\mathrm{u}}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^{\star}, t)\hat{U}_{\mathrm{r}}^{(1)}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^{\star}, t)|0\rangle\, \langle\boldsymbol{\nu}_0| \tag{192}$$

is the first correction to the TGA propagator in our scheme, and

$$\hat{C}_{\mathrm{p}}^{(1)}(t) = \int_{\Gamma_{\mu_0,\nu_0^{\star}}} \frac{\mathrm{d}\boldsymbol{\mu}_0\, \mathrm{d}\boldsymbol{\nu}_0^{\star}}{(2\pi\hbar\mathrm{i})^d}\, \mathrm{e}^{-\nu_0^{\star}\mu_0/\hbar}\hat{E}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^{\star}, t)\hat{U}_{\mathrm{r}}^{(1)}(\boldsymbol{\mu}_0, \boldsymbol{\nu}_0^{\star}, t)|0\rangle\, \langle\boldsymbol{\nu}_0| \tag{193}$$

is an operator that is of second order in the non-quadratic perturbation $\hat{H}_{sr}^{\mathrm{I}}$. Hence, as one should have been expected, the first correction $\hat{U}^{(1)}(t)$ in our expansion differs from the analogous one, $\hat{U}_1(t)$, in the expansion of Pollak and coworkers only in terms of second order in the perturbative parameter. This result can be generalized to all orders, so that when both expansions are truncated at a given order l they differ only in terms of order $l + 1$ in the non-quadratic perturbation $\hat{H}_{sr}^{\mathrm{I}}$. Therefore, the wrong behaviour found in our series expansion for $\hat{U}_{\mathrm{r}}(t)$ in the semiclassical limit is also expected in the series in (186), when the TGA IVR is used for the zero order term $\hat{U}_0(t)$.

8. Conclusions

An algebraic approach based on the multimode two-photon Lie algebra and its corresponding Lie group has been followed to define the most general Gaussian state for a multidimensional system and to obtain its time evolution. This has yielded a formal solution to the time-dependent Schrödinger equation that is written as an expansion series whose leading term corresponds to the thawed Gaussian approximation (TGA). Our scheme provides the most general expression so far reported for this approximation, which includes a parameter fixing either the quantization scheme or the Hamiltonian classical symbol. The correction term to this approximation is then analysed in the zero $\hbar$ asymptotic limit, using the coherent state representation for this solution. The error was generally found not to vanish in the semiclassical limit. Only if the point in the initial ket manifold is properly chosen, the corresponding generalized Gaussian wave packet dynamics (GGWPD) provides an approximation to the exact solution that is correct in the semiclassical limit.

The same approach has been followed to analyse the error in the TGA initial value representation (IVR) of the quantum propagator, which was found not to vanish either in the zero $\hbar$ limit. Hence, this approximation to the quantum propagator does not provide the correct semiclassical asymptotic form for this operator. The origin of this behaviour is shown to be in an incorrect analytic structure of the TGA IVR which in the semiclassical limit leads to unphysical asymptotic saddle-point contributions. These contributions have been so far disregarded, which has led to the wrong conclusion that the TGA IVR provides the correct asymptotic semiclassical limit. Inadequate behaviours of this propagator such as the fast loss of unitarity may have an explanation in the unveiled analytical structure. Despite these negative results, we have shown that the TGA IVR can provide a good approximation to the quantum propagator when the terms higher than quadratic in the Hamiltonian expansion and included in the operator $\hat{H}^{\mathrm{I}}_{sr}$ can be considered a small enough perturbation.

This behaviour of TGA IVR contrasts markedly with that of the Herman–Kluk IVR [17]. The semiclassical asymptotic limit for the coherent-state matrix elements of this propagator can be easily obtained following the approach presented in [12]. All contributions in this limit come from regular saddle points so that the correct semiclassical expression is attained in this case for such matrix elements [12]. Besides, in [12, 26] it is shown that the use of a rigorous asymptotic approach always leads to the Herman–Kluk propagator as the leading semiclassical propagator. These results and our present analysis are therefore fully consistent.

Our approach has also provided a few interesting side results which are worth a closer analysis. Here we will highlight just two of them. The first one is the existence of an initial ket manifold bigger than that originally discovered by Heller and coworkers [2, 3]. This result expands significantly the space of possible forms to express a given Gaussian state by choosing different values for the parameter vector χ. The second side result is the existence of different formulations of the TGA arising from the freedom in both the choice of the function $\boldsymbol{\nu}_0^\star(\boldsymbol{\zeta}, \boldsymbol{\eta}^\star)$ in (160) and the choice of its complex integration manifold $\Gamma_{\mu_0,\nu_0^\star}$. We leave for a future work the analysis of these new results and their consequences in the search for improved numerical procedures. For this goal, the use of variational methods may be very valuable, and our algebraic approach is particularly suited for implementing these methods.

As a final comment, one should accept the fact that the TGA is the simplest of all Gaussian wavepacket approximations, since it solves the propagation problem with a single real trajectory for a known initial condition. On the other hand, IVR propagators involve the calculation of a large number of trajectories; thus one expects more accuracy from these approximations, although the physical interpretation in terms of classical processes is more difficult. Intermediate approaches that use a few real or complex trajectories have also been proposed [33]. In all these cases one would always like to measure the associated error, and the approach followed in this work can be very useful for such purpose, as we have shown for the TGA and TGA-IVR approximations.

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References

[1] Heller E J 1975 *J. Chem. Phys.* **62** 1544
[2] Huber D and Heller E J 1987 *J. Chem. Phys.* **87** 5302
[3] Huber D, Heller E J and Littlejohn R G 1988 *J. Chem. Phys.* **89** 2003
[4] Heller E J 1977 *J. Chem. Phys.* **66** 5777
[5] Miller W H 1974 *Adv. Chem. Phys.* **25** 69
[6] Weissman Y 1982 *J. Chem. Phys.* **76** 4067
[7] Weissman Y 1983 *J. Phys. A: Math. Gen.* **16** 2693
[8] Klauder J R 1978 *Path Integrals and Their Applications to Quantum, Statistical and Solid State Physic* ed G J Papadopoulos and J T Devreese (New York: Plenum) p 5
[9] Klauder J R 1979 *Phys. Rev.* D **19** 2349
[10] Klauder J R 1987 *Random Media* ed G Papanicolaou (Berlin: Springer) p 163
[11] Baranger M, de Aguiar M A M, Keck F, Korsch H J and Schellhaaß B 2001 *J. Phys. A: Math. Gen.* **34** 7227
[12] Martín-Fierro E and Gomez Llorente J M 2006 *Chem. Phys.* **322** 13
[13] Kay K G 1994 *J. Chem. Phys.* **100** 4377
[14] Van Vleck J H 1928 *Proc. Natl. Acad. Sci. USA* **14** 178
[15] Pollak E and Miret-Arté S 2004 *J. Phys. A: Math. Gen.* **37** 9669
[16] Parisio M and de Aguiar M A M 2003 *Phys. Rev.* A **68** 062112
[17] Herman M F and Kluk E 1984 *Chem. Phys.* **91** 27
[18] Solari H G 1986 *J. Math. Phys.* **27** 1351
[19] Grossmann F 1999 *Comments At. Mol. Phys.* **34** 141
[20] Miller W H 2001 *J. Phys. Chem.* A **105** 2942
[21] Kay K G 2005 *Annu. Rev. Phys. Chem.* **56** 255
[22] Kay K G 1994 *J. Chem. Phys.* **100** 4432
[23] Harabati C, Rost J M and Grossmann F 2004 *J. Chem. Phys.* **120** 26
[24] Grossmann F and Herman M F 2002 *J. Phys. A: Math. Gen.* **35** 9489
[25] Baranger M, de Aguiar M A M, Keck F, Korsch H J and Schellhaaß B 2002 *J. Phys. A: Math. Gen.* **35** 9493
[26] Kay K G 2006 *Chem. Phys.* **322** 3
[27] Ankerhold J, Saltzer M and Pollak E 2002 *J. Chem. Phys.* **116** 5925
[28] Pollak E and Shao J 2003 *J. Phys. Chem.* A **107** 7112
[29] Zhang S and Pollak E 2003 *Phys. Rev. Lett.* **91** 190201
[30] Zhang W M, Feng D H and Gilmore R 1990 *Rev. Mod. Phys.* **62** 867
[31] Louisell W H 1973 *Quantum Statistical Properties of Radiation* (New York: Wiley)
[32] Bargmann V 1961 *Commun. Pure Appl. Math.* **14** 187
[33] de Aguiar M A M, Baranger M, Jaubert L, Parisio F and Ribeiro A D 2005 *J. Phys. A: Math. Gen.* **38** 4645
[34] Rubin A and Klauder J R 1995 *Ann. Phys.* **241** 212
[35] Shudo A and Ikeda K S 1996 *Phys. Rev. Lett.* **76** 4151
[36] Parisio F and de Aguiar M A M 2005 *J. Phys. A: Math. Gen.* **38** 9317