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On the derivation of the Herman–Kluk propagator

Sarin A Deshpande and Gregory S Ezra

Department of Chemistry and Chemical Biology, Baker Laboratory, Cornell University, Ithaca, NY 14853, USA

E-mail: gse1@cornell.edu

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Abstract
Following an earlier derivation by Miller, we give a concise derivation of the Herman–Kluk propagator that fully exploits the symplectic structure of the problem. We also show directly that the version of the thawed Gaussian semiclassical propagator obtained by Baranger and co-workers is equivalent to the linearized Herman–Kluk propagator.

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1. Introduction

Semiclassical approximations to quantum propagators have been widely applied to introduce aspects of quantum dynamics into the motions of systems near the classical limit [1–16]. Early applications [3, 4] of semiclassical methods to problems of chemical dynamics used the Van Vleck propagator [1, 2, 7]. Direct use of the Van Vleck form, however, leads to well-known difficulties associated with the trajectory root search problem and possible divergence of the prefactor at conjugate points. It is moreover necessary to keep track of the Morse–Maslov index phase factor associated with the passage of classical trajectories through conjugate points [7, 17]. Most recent applications of semiclassical mechanics have therefore exploited initial value representations (IVR) [18, 19] of the propagator [5, 6, 9, 11–14, 16, 20–27]. Using the IVR, it is possible to propagate trajectories specified by an initial coordinate–momentum pair, rather than initial and final coordinate values. The Herman–Kluk (HK) representation of the propagator in particular has been widely applied [5, 6, 12, 13, 16, 24]. The HK propagator is a frozen Gaussian [28] IVR approximation to the propagator in the coherent state representation [10, 29, 30]. In addition to providing a semiclassical IVR for the propagator, the HK approximation has the useful property that the prefactor is always well behaved with magnitude never zero or divergent. The appropriate phase of the prefactor (branch of the square root function) is therefore easily determined through continuity [21, 23]. Generalizations of the HK prefactor have been discussed by Hu and co-workers [31, 32].

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The original derivation of the HK frozen Gaussian propagator [5] has been criticized by Baranger *et al* [14], who argue that certain deformations of integration contours in complex phase space invoked in the HK derivation are not necessarily valid. Baranger *et al* contend that this objection also holds for the derivation by Grossmann and Xavier [33]. Derivations of the HK propagator using different approaches have subsequently been given by Miller [34, 35] and by Shalashilin and Child [24], while Kay has discussed general integral representations of the semiclassical propagator [16, 21], of which the HK propagator is a particular case (see also [31, 36]).

Miller's second derivation of the HK propagator [35] is based on an expansion in terms of a time-dependent basis of Gaussian coherent states, where the members of the (continuous, overcomplete) coherent state basis are parametrized by phase points $\boldsymbol{z}_t$, with $\boldsymbol{z}_t$ evolving from $\boldsymbol{z}$ at time $t = 0$ under the classical time evolution generated by the system Hamiltonian. While this expansion is in principle exact, Miller has shown that an assumption of slow variation of the expansion coefficients with $\boldsymbol{z}_t$ combined with a local quadratic expansion of the Hamiltonian yields, upon suitable linearization of the integrand, an equation of motion for the expansion coefficients that is identical with that of the HK prefactor [35]. An elegant treatment of the HK propagator by Shalashilin and Child based on their coupled coherent state approach [25, 37, 38] employs complex combinations of (real) phase space coordinates and associated complex monodromy matrices, and makes extensive use of matrix identities resulting from the symplectic property of the monodromy matrix [25].

An intriguing feature of the HK propagator is the presence of the determinant of the complex matrix $\boldsymbol{\Lambda}^*$ (see below) in the prefactor. This prefactor presumably compensates for the fact that, in contrast to the thawed Gaussian approximation (TGA) [30, 39], the coherent states in the expansion basis are 'frozen' and do not distort or spread as they move along the guiding classical trajectory. It is therefore interesting to note that the determinant of the matrix $\boldsymbol{\Lambda}$ appears in the coherent state matrix elements of the metaplectic operator associated with a linear canonical transformation (symplectic matrix) [30]. The coherent state matrix element of the metaplectic operator associated with the monodromy matrix for the classical trajectory from $\boldsymbol{z} \to \boldsymbol{z}_t$ in turn appears in the form of the thawed Gaussian propagator proposed by Littlejohn [30] (see also [40]).

The purpose of the present paper is to point out that additional insight into the derivation and structure of the HK propagator and its relation to the TGA is obtained by exploiting the connection with the metaplectic operator and related matrix identities [40]. Specifically, the matrix $\boldsymbol{\Lambda}$, which is obtained by taking complex linear combinations of blocks of the real monodromy matrix $\boldsymbol{S}$, arises naturally without the need to explicitly invoke complex combinations of real phase space variables, essentially as a result of the interplay between the real and imaginary parts of the exponents of coherent state overlaps.

After a review of notation and definitions (section 2), we reformulate Miller's derivation of the HK propagator to fully exploit the underlying symplectic structure (section 3). Our derivation makes it clear that the HK propagator is by no means limited to Hamiltonians of the form $H = T(p) + V(q)$. We then show (section 4) that the linearized coherent state matrix element of the HK propagator is identical to the propagator in the TGA in the form given by Littlejohn [30]. This form is also identical (with a couple of modifications) to the result obtained by Baranger *et al* [14] for one-dimensional systems.

2. Notation and definitions

In this section, we briefly establish notation and essential definitions. *Real* $2N$-dimensional phase space coordinate vectors are denoted by $\boldsymbol{z} \equiv (\boldsymbol{q}, \boldsymbol{p})$, where $\boldsymbol{q}$ and $\boldsymbol{p}$ are N-dimensional

Cartesian coordinate and momentum vectors, respectively. Hamilton's equations are written as

$$\dot{\boldsymbol{z}} = \boldsymbol{J} \cdot H_{\boldsymbol{z}} \tag{1}$$

with $\boldsymbol{J}$ the $2N \times 2N$ fundamental matrix,

$$\boldsymbol{J} = \begin{bmatrix} \boldsymbol{0} & +\boldsymbol{1}_N \\ -\boldsymbol{1}_N & \boldsymbol{0} \end{bmatrix}, \tag{2}$$

and $H_{\boldsymbol{z}}$ the vector of derivatives of the classical Hamiltonian $H(\boldsymbol{z})$ with respect to phase space coordinates $\boldsymbol{z}$. The Hamiltonian $H(\boldsymbol{z})$ is not restricted to be of the form $H(\boldsymbol{z}) = T(\boldsymbol{p}) + V(\boldsymbol{q})$. Note that $\widetilde{\boldsymbol{J}} = \boldsymbol{J}^{-1} = -\boldsymbol{J}$, where $\widetilde{\boldsymbol{J}}$ is the transpose of $\boldsymbol{J}$. The symplecticity condition on a $2N \times 2N$ matrix $\boldsymbol{S}$ is

$$\widetilde{\boldsymbol{S}}\boldsymbol{J}\boldsymbol{S} = \boldsymbol{J}. \tag{3}$$

If the matrix $\boldsymbol{S}$ is symplectic then so are the following: $\boldsymbol{S}^{-1}$, $\widetilde{\boldsymbol{S}}^{-1}$, $\widetilde{\boldsymbol{S}}$. The symplectic matrices we consider here are monodromy (stability) matrices associated with classical trajectories from $\boldsymbol{z} \to \boldsymbol{z}_t$. The matrix $\boldsymbol{S}$ is partitioned as follows:

$$\boldsymbol{S} = \left[\frac{\partial \boldsymbol{z}_t}{\partial \boldsymbol{z}}\right] = \begin{bmatrix} \boldsymbol{S}_{qq} & \boldsymbol{S}_{qp} \\ \boldsymbol{S}_{pq} & \boldsymbol{S}_{pp} \end{bmatrix} \equiv \begin{bmatrix} \boldsymbol{A} & \boldsymbol{B} \\ \boldsymbol{C} & \boldsymbol{D} \end{bmatrix}. \tag{4}$$

Condition (3) implies

$$\widetilde{\boldsymbol{C}}\boldsymbol{A} = \widetilde{\boldsymbol{A}}\boldsymbol{C} \tag{5a}$$

$$\widetilde{\boldsymbol{D}}\boldsymbol{B} = \widetilde{\boldsymbol{B}}\boldsymbol{D} \tag{5b}$$

$$-\widetilde{\boldsymbol{B}}\boldsymbol{C} + \widetilde{\boldsymbol{D}}\boldsymbol{A} = \widetilde{\boldsymbol{A}}\boldsymbol{D} - \widetilde{\boldsymbol{C}}\boldsymbol{B} = \boldsymbol{1}_N, \tag{5c}$$

with a similar set of relations following from the condition $\boldsymbol{S}\boldsymbol{J}\widetilde{\boldsymbol{S}} = \boldsymbol{J}$. The inverse matrix $\boldsymbol{S}^{-1}$ is

$$\boldsymbol{S}^{-1} = \begin{bmatrix} \widetilde{\boldsymbol{D}} & -\widetilde{\boldsymbol{B}} \\ -\widetilde{\boldsymbol{C}} & \widetilde{\boldsymbol{A}} \end{bmatrix}. \tag{6}$$

The $2N \times 2N$ complex matrix

$$\boldsymbol{W} = \frac{1}{\sqrt{2}} \begin{bmatrix} \boldsymbol{1}_N & \mathrm{i}\boldsymbol{1}_N \\ \mathrm{i}\boldsymbol{1}_N & \boldsymbol{1}_N \end{bmatrix} \tag{7}$$

is both symplectic and complex symmetric. A complex symplectic matrix $\boldsymbol{S}_c$ is obtained by similarity transforming $\boldsymbol{S}$ with $\boldsymbol{W}$,

$$\boldsymbol{S}_c = \boldsymbol{W}\boldsymbol{S}\boldsymbol{W}^{-1} = \begin{bmatrix} \boldsymbol{\Lambda}^* & -\mathrm{i}\boldsymbol{\Gamma}^* \\ \mathrm{i}\boldsymbol{\Gamma} & \boldsymbol{\Lambda} \end{bmatrix}, \tag{8}$$

where we have defined the complex $N \times N$ matrices [30]

$$\boldsymbol{\Lambda} \equiv \tfrac{1}{2}[(\boldsymbol{A} + \boldsymbol{D}) + \mathrm{i}(\boldsymbol{B} - \boldsymbol{C})] \tag{9a}$$

$$\boldsymbol{\Gamma} \equiv \tfrac{1}{2}[(\boldsymbol{A} - \boldsymbol{D}) - \mathrm{i}(\boldsymbol{B} + \boldsymbol{C})]. \tag{9b}$$

Symplecticity of $\boldsymbol{S}_c$ implies

$$\boldsymbol{\Gamma}\widetilde{\boldsymbol{\Lambda}} = \boldsymbol{\Lambda}\widetilde{\boldsymbol{\Gamma}} \tag{10a}$$

$$\boldsymbol{\Lambda}^*\widetilde{\boldsymbol{\Gamma}}^* = \boldsymbol{\Gamma}^*\widetilde{\boldsymbol{\Lambda}}^* \tag{10b}$$

$$\boldsymbol{\Lambda}^*\widetilde{\boldsymbol{\Lambda}} - \boldsymbol{\Gamma}^*\widetilde{\boldsymbol{\Gamma}} = \boldsymbol{\Lambda}\widetilde{\boldsymbol{\Lambda}}^* - \boldsymbol{\Gamma}\widetilde{\boldsymbol{\Gamma}}^* = \boldsymbol{1}_N, \tag{10c}$$

while $\boldsymbol{S}_c^{-1}$ is

$$\boldsymbol{S}_c^{-1} = \boldsymbol{W}\boldsymbol{S}^{-1}\boldsymbol{W}^{-1} = \begin{bmatrix} \widetilde{\boldsymbol{\Lambda}} & \mathrm{i}\widetilde{\boldsymbol{\Gamma}}^* \\ -\mathrm{i}\widetilde{\boldsymbol{\Gamma}} & \widetilde{\boldsymbol{\Lambda}}^* \end{bmatrix}. \tag{11}$$

The coherent state centred at phase point $\boldsymbol{z} = (\boldsymbol{q}, \boldsymbol{p})$ is defined as [30]

$$\langle \boldsymbol{x}|\boldsymbol{z}\rangle = \langle \boldsymbol{x}|\hat{T}(\boldsymbol{z})|\boldsymbol{0}\rangle \tag{12a}$$

$$= \frac{1}{(\pi\hbar)^{N/4}} \exp\left\{\frac{1}{\hbar}\left[-\frac{1}{2}(\widetilde{\boldsymbol{x}} - \widetilde{\boldsymbol{q}})\cdot(\boldsymbol{x}-\boldsymbol{q}) + \mathrm{i}\widetilde{\boldsymbol{p}}\cdot\boldsymbol{x} - \frac{\mathrm{i}}{2}\widetilde{\boldsymbol{p}}\cdot\boldsymbol{q}\right]\right\}, \tag{12b}$$

where $\hat{T}(\boldsymbol{z})$ is the Heisenberg translation operator [30] and $|\boldsymbol{0}\rangle$ is the fiducial state (harmonic oscillator ground state) centred at $\boldsymbol{z} = \boldsymbol{0}$ expressed in terms of suitably scaled coordinates. Our phase convention for coherent states is determined by the action of the operators $\hat{T}(\boldsymbol{z})$ [30]. Herman–Kluk [5] and many other authors use a different convention

$$|\boldsymbol{z}\rangle_{\mathrm{HK}} = |\boldsymbol{z}\rangle\, \mathrm{e}^{-\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{q}/2\hbar} \tag{13}$$

so that the classical action $R(\boldsymbol{z}; t)$ for trajectory $\boldsymbol{z} \to \boldsymbol{z}_t$ appearing in the phase of the HK propagator [5] must be modified to have the symmetrical form

$$\Phi(\boldsymbol{z}; t) = R(\boldsymbol{z}; t) - \frac{1}{2}[\boldsymbol{p}_t\cdot\boldsymbol{q}_t - \boldsymbol{p}\cdot\boldsymbol{q}] \tag{14a}$$

$$= \int \mathrm{d}t\, L - \frac{1}{2}[\boldsymbol{p}_t\cdot\boldsymbol{q}_t - \boldsymbol{p}\cdot\boldsymbol{q}] \tag{14b}$$

$$= \int \mathrm{d}t \left[\frac{1}{2}(\boldsymbol{p}\cdot\dot{\boldsymbol{q}} - \boldsymbol{q}\cdot\dot{\boldsymbol{p}}) - H\right]. \tag{14c}$$

The overlap between coherent states $|\boldsymbol{z}\rangle$ and $|\boldsymbol{z}'\rangle$ is

$$\langle \boldsymbol{z}'|\boldsymbol{z}\rangle = \exp\left[-\frac{1}{4\hbar}(\widetilde{\boldsymbol{z}'} - \widetilde{\boldsymbol{z}})\cdot\boldsymbol{1}_{2N}\cdot(\boldsymbol{z}'-\boldsymbol{z}) + \frac{\mathrm{i}}{2\hbar}\widetilde{\boldsymbol{z}'}\cdot\boldsymbol{J}\cdot\boldsymbol{z}\right]. \tag{15}$$

Given a symplectic matrix $\boldsymbol{S}$, we define the real, symmetric and positive definite matrix

$$\boldsymbol{X} = \boldsymbol{1}_{2N} + \widetilde{\boldsymbol{S}}\boldsymbol{S}. \tag{16}$$

The matrix $\boldsymbol{X}$ factorizes as follows (cf appendix C of [26]; see also [41, 42]):

$$\boldsymbol{X} = \boldsymbol{U}\cdot\boldsymbol{V} \tag{17}$$

with

$$\boldsymbol{U} = \begin{bmatrix} \mathrm{i}\widetilde{\boldsymbol{\Lambda}} & -\mathrm{i}\widetilde{\boldsymbol{\Lambda}}^* \\ \widetilde{\boldsymbol{\Lambda}} & \widetilde{\boldsymbol{\Lambda}}^* \end{bmatrix}, \qquad \boldsymbol{V} = \begin{bmatrix} \boldsymbol{C} - \mathrm{i}\boldsymbol{A} & \boldsymbol{D} - \mathrm{i}\boldsymbol{B} \\ \boldsymbol{C} + \mathrm{i}\boldsymbol{A} & \boldsymbol{D} + \mathrm{i}\boldsymbol{B} \end{bmatrix} \tag{18}$$

and

$$\boldsymbol{X}^{-1} = \boldsymbol{V}^{-1}\cdot\boldsymbol{U}^{-1} = \frac{1}{2}\begin{bmatrix} -\widetilde{\boldsymbol{B}} + \mathrm{i}\widetilde{\boldsymbol{D}} & -\widetilde{\boldsymbol{B}} - \mathrm{i}\widetilde{\boldsymbol{D}} \\ \widetilde{\boldsymbol{A}} - \mathrm{i}\widetilde{\boldsymbol{C}} & \widetilde{\boldsymbol{A}} + \mathrm{i}\widetilde{\boldsymbol{C}} \end{bmatrix}\cdot\frac{1}{2}\begin{bmatrix} -\mathrm{i}\widetilde{\boldsymbol{\Lambda}}^{-1} & \widetilde{\boldsymbol{\Lambda}}^{-1} \\ \mathrm{i}\widetilde{\boldsymbol{\Lambda}}^{*-1} & \widetilde{\boldsymbol{\Lambda}}^{*-1} \end{bmatrix}. \tag{19}$$

Using the above results we can derive the identity

$$\boldsymbol{P} \equiv -(\boldsymbol{1} + \mathrm{i}\boldsymbol{J})\boldsymbol{S}\boldsymbol{X}^{-1}\widetilde{\boldsymbol{S}}(\boldsymbol{1} + \mathrm{i}\widetilde{\boldsymbol{J}}) = +\begin{bmatrix} -\boldsymbol{\Gamma}^*\boldsymbol{\Lambda}^{-1} & \mathrm{i}\boldsymbol{\Gamma}^*\boldsymbol{\Lambda}^{-1} \\ \mathrm{i}\boldsymbol{\Gamma}^*\boldsymbol{\Lambda}^{-1} & +\boldsymbol{\Gamma}^*\boldsymbol{\Lambda}^{-1} \end{bmatrix}. \tag{20}$$

The matrix $\boldsymbol{P}$ is manifestly symmetric, and is important in the symplectic formulation of wavepacket propagation [30, 40].

A symplectic matrix $\boldsymbol{S}$, which is associated with the linear classical canonical transformation

$$\boldsymbol{z} \mapsto \overline{\boldsymbol{z}} = \boldsymbol{S} \cdot \boldsymbol{z}, \tag{21}$$

induces a corresponding transformation of quantum mechanical operators $\hat{\boldsymbol{z}}$ by the unitary *metaplectic* operator $\hat{M}(\boldsymbol{S})$ [30],

$$\hat{\boldsymbol{z}} \mapsto \hat{M}(\boldsymbol{S})\hat{\boldsymbol{z}}\hat{M}(\boldsymbol{S})^{\dagger} = \boldsymbol{S} \cdot \hat{\boldsymbol{z}}. \tag{22}$$

General matrix elements of the operator $\hat{M}(\boldsymbol{S})$ in position, coherent state and mixed bases have been given by Littlejohn [30]. Here, we point out that the matrix element between the fiducial state and the coherent state $|\boldsymbol{z}'\rangle$ can be expressed in terms of the matrix $\boldsymbol{P}$ ([30]; [40], chapter 5)

$$\langle \boldsymbol{z}'|\hat{M}(\boldsymbol{S}, \sigma)|\boldsymbol{0}\rangle = \frac{\sigma}{\sqrt{\det[\boldsymbol{\Lambda}]}} \exp\left[-\frac{1}{4\hbar}\widetilde{\boldsymbol{z}}' \cdot (\boldsymbol{1} + \boldsymbol{P}) \cdot \boldsymbol{z}'\right]. \tag{23}$$

The (for present purposes irrelevant) sign $\sigma = \pm 1$ is associated with a 2:1 correspondence between the metaplectic and symplectic groups [30]. Note the appearance in (23) of $\sqrt{\det[\boldsymbol{\Lambda}]}$, closely related to the HK prefactor. When $\boldsymbol{S}$ is the monodromy matrix associated with a classical trajectory from $\boldsymbol{z} \to \boldsymbol{z}_t$, the state $\hat{M}(\boldsymbol{S}, \sigma)|\boldsymbol{0}\rangle$ is the fiducial Gaussian state distorted by dynamically induced spreading along the trajectory $\boldsymbol{z} \to \boldsymbol{z}_t$.

3. Miller's derivation of the HK propagator revisited

Following Miller, we consider a potentially *exact* expansion of the quantum state $|\psi_t\rangle$ evolving from the initial state $|\psi_0\rangle$ under the (time-independent) Hamiltonian $\hat{H}$ in terms of the (overcomplete) coherent state basis $|\boldsymbol{z}'_t\rangle$, where the classical phase point $\boldsymbol{z}'$ propagates to the phase point $\boldsymbol{z}'_t$ along the classical trajectory determined by the corresponding classical Hamiltonian H:

$$|\psi_t\rangle = \frac{1}{(2\pi\hbar)^N} \int \mathrm{d}^{2N}\boldsymbol{z}' |\boldsymbol{z}'_t\rangle\, \mathrm{e}^{\mathrm{i}\Phi(\boldsymbol{z}'_t;t)/\hbar} C(\boldsymbol{z}'_t)\langle \boldsymbol{z}'|\psi_0\rangle. \tag{24}$$

An exact expression for $|\psi_t\rangle$ can in principle be obtained by solving the coupled integro-differential equations for the coefficients $C(\boldsymbol{z}_t)$ that result when (24) is substituted into the time-dependent Schrödinger equation [25, 37, 38]

$$\mathrm{i}\hbar\partial_t|\psi_t\rangle = \hat{H}|\psi_t\rangle. \tag{25}$$

Time evolution under the HK propagator [5] results in an expression for $|\psi_t\rangle$ very similar to that in (24),

$$|\psi_t\rangle_{\mathrm{HK}} = \frac{1}{(2\pi\hbar)^N} \int \mathrm{d}^{2N}\boldsymbol{z}' |\boldsymbol{z}'_t\rangle\, \mathrm{e}^{\mathrm{i}\Phi(\boldsymbol{z}'_t;t)/\hbar} C_{\mathrm{HK}}(\boldsymbol{z}'; t)\langle \boldsymbol{z}'|\psi_0\rangle, \tag{26}$$

where the prefactor C_{HK} now has the explicit form

$$C_{\mathrm{HK}}(\boldsymbol{z}'; t) = \sqrt{\det[\boldsymbol{\Lambda}^*]} \tag{27}$$

with $\boldsymbol{\Lambda}$ the complex $N \times N$ matrix defined in (9*b*). (We do not explicitly include an additional phase factor (index) needed to ensure continuity of the square root.) As realized by Miller, in order to pass from (24) to (26) the coefficient $C(\boldsymbol{z}_t)$ must be considered a functional of the classical trajectory $\boldsymbol{z} \to \boldsymbol{z}_t$ only, with a weak dependence on $\boldsymbol{z}'_t$. Substituting (24) into the time-dependent Schrödinger equation and taking C and $\dot{C}$ outside the integral over $\boldsymbol{z}'$ then yields an equation of motion for C that turns out to be identical to that of the HK prefactor (27).

Here, we give a version of this derivation for general Hamiltonians $H(z)$ that explicitly and fully exploits the symplectic algebraic properties of the various matrices appearing in the calculation. Again following Miller, we set $|\psi_0\rangle = |z\rangle$. To facilitate the calculation, we consider the particular matrix element of the exact propagator

$$\langle z_t|\psi_t\rangle = \frac{1}{(2\pi\hbar)^N}\int \mathrm{d}^{2N}z'\langle z_t|z'_t\rangle\, \mathrm{e}^{\mathrm{i}\Phi(z'_t;t)/\hbar}C(z'_t)\langle z'|z\rangle. \tag{28}$$

The exact solution will satisfy

$$\mathrm{i}\hbar\int \mathrm{d}^{2N}z'\langle z_t|\frac{\mathrm{d}}{\mathrm{d}t}[|z'_t\rangle\, \mathrm{e}^{\mathrm{i}\Phi(z'_t;t)/\hbar}C(z'_t)]\langle z'|z\rangle = \int \mathrm{d}^{2N}z'\langle z_t|\hat{H}|z'_t\rangle\, \mathrm{e}^{\mathrm{i}\Phi(z'_t;t)/\hbar}C(z'_t)\langle z'|z\rangle. \tag{29}$$

The approximate equation of motion for $C(z'_t)$ is obtained by assuming that $C(z'_t)$ is a slowly varying function of z'_t, and subsequently linearizing the integrand about $z, z' = z + \Delta z$. We therefore write

$$\begin{aligned}\mathrm{i}\hbar\dot{C}(z_t)\int \mathrm{d}^{2N}z'\langle z_t|z'_t\rangle\, \mathrm{e}^{\mathrm{i}\Phi(z'_t;t)/\hbar}\langle z'|z\rangle + \mathrm{i}\hbar C(z_t)\int \mathrm{d}^{2N}z'\langle z_t|\frac{\mathrm{d}}{\mathrm{d}t}[|z'_t\rangle\, \mathrm{e}^{\mathrm{i}\Phi(z'_t;t)/\hbar}]\langle z'|z\rangle \\ \simeq C(z_t)\int \mathrm{d}^{2N}z'\langle z_t|\hat{H}|z'_t\rangle\, \mathrm{e}^{\mathrm{i}\Phi(z'_t;t)/\hbar}\langle z'|z\rangle.\end{aligned} \tag{30}$$

We now derive the equation of motion for $C(z_t)$ implied by (30), and show that it is precisely equivalent to that for the HK prefactor.

3.1. Equation of motion for the HK prefactor

The time derivative of the HK prefactor $\sqrt{\det[\mathbf{\Lambda}^*]}$ is

$$\frac{\mathrm{d}\sqrt{\det[\mathbf{\Lambda}^*]}}{\mathrm{d}t} = \frac{1}{2\sqrt{\det[\mathbf{\Lambda}^*]}}\frac{\mathrm{d}\det[\mathbf{\Lambda}^*]}{\mathrm{d}t} = \frac{1}{2}\sqrt{\det[\mathbf{\Lambda}^*]}\,\mathrm{tr}\left[\frac{\mathrm{d}\mathbf{\Lambda}^*}{\mathrm{d}t}\mathbf{\Lambda}^{*-1}\right]. \tag{31}$$

To determine $\mathrm{d}\mathbf{\Lambda}^*/\mathrm{d}t$, we note that the time evolution of the monodromy matrix S is governed by the tangent dynamics,

$$\dot{S} = J\mathfrak{H}S, \tag{32}$$

where $\mathfrak{H}$ is the $2N\times 2N$ matrix of second derivatives of the Hamiltonian, $\mathfrak{H}_{jk} \equiv \partial^2 H/\partial z_j\partial z_k$. We partition $\mathfrak{H}$ into four $N\times N$ matrices,

$$\mathfrak{H} = \begin{bmatrix}\mathfrak{H}_A & \mathfrak{H}_B\\ \mathfrak{H}_C & \mathfrak{H}_D\end{bmatrix}, \tag{33}$$

following the partitioning of S. Defining complex $N\times N$ matrices (cf equation (9))

$$\mathfrak{H}_\Lambda = \tfrac{1}{2}[(\mathfrak{H}_A + \mathfrak{H}_D) + \mathrm{i}(\mathfrak{H}_B - \mathfrak{H}_C)] \tag{34a}$$

$$\mathfrak{H}_\Gamma = \tfrac{1}{2}[(\mathfrak{H}_A - \mathfrak{H}_D) - \mathrm{i}(\mathfrak{H}_B + \mathfrak{H}_C)] \tag{34b}$$

we find

$$\dot{S}_c \equiv W\dot{S}W^{-1} \tag{35a}$$

$$= \begin{bmatrix}\dot{\mathbf{\Lambda}}^* & -\mathrm{i}\dot{\mathbf{\Gamma}}^*\\ \mathrm{i}\dot{\mathbf{\Gamma}} & \dot{\mathbf{\Lambda}}\end{bmatrix} \tag{35b}$$

$$= WJ\mathfrak{H}W^{-1}S_c \tag{35c}$$

$$= -\begin{bmatrix}\mathrm{i}\mathfrak{H}_\Lambda^*\mathbf{\Lambda}^* + \mathrm{i}\mathfrak{H}_\Gamma^*\mathbf{\Gamma} & \mathfrak{H}_\Lambda^*\mathbf{\Gamma}^* + \mathfrak{H}_\Gamma^*\mathbf{\Lambda}\\ \mathfrak{H}_\Gamma\mathbf{\Lambda}^* + \mathfrak{H}_\Lambda\mathbf{\Gamma} & -\mathrm{i}\mathfrak{H}_\Gamma\mathbf{\Gamma}^* - \mathrm{i}\mathfrak{H}_\Lambda\mathbf{\Lambda}\end{bmatrix}. \tag{35d}$$

The time evolution of $\mathbf{\Lambda}^*$ and $\mathbf{\Gamma}$ is, therefore, governed by the pair of coupled equations

$$\dot{\mathbf{\Lambda}}^* = -\mathrm{i}(\mathfrak{H}_\Lambda^* \mathbf{\Lambda}^* + \mathfrak{H}_\Gamma^* \mathbf{\Gamma}) \tag{36a}$$

$$\dot{\mathbf{\Gamma}} = +\mathrm{i}(\mathfrak{H}_\Gamma \mathbf{\Lambda}^* + \mathfrak{H}_\Lambda \mathbf{\Gamma}). \tag{36b}$$

We have

$$\dot{\mathbf{\Lambda}}^* \mathbf{\Lambda}^{*-1} = -\mathrm{i}(\mathfrak{H}_\Lambda^* + \mathfrak{H}_\Gamma^* \mathbf{\Gamma} \mathbf{\Lambda}^{*-1}) \tag{37}$$

so that the equation of motion for the HK prefactor becomes

$$\frac{\mathrm{i}}{\sqrt{\det[\mathbf{\Lambda}^*]}} \frac{\mathrm{d}\sqrt{\det[\mathbf{\Lambda}^*]}}{\mathrm{d}t} = \frac{1}{2} \operatorname{tr}[\mathfrak{H}_\Lambda^* + \mathfrak{H}_\Gamma^* \mathbf{\Gamma} \mathbf{\Lambda}^{*-1}]. \tag{38}$$

3.2. Coherent state matrix elements of the Hamiltonian

In order to derive the equation of motion for the approximate prefactor C using equation (30), we require matrix elements of the Hamiltonian $\hat{H}$ between coherent states $|\boldsymbol{z}_t\rangle$ and $|\boldsymbol{z}_t'\rangle = |\boldsymbol{z}_t + \Delta \boldsymbol{z}_t\rangle \simeq |\boldsymbol{z}_t + \boldsymbol{S}\Delta \boldsymbol{z}\rangle$. The expansion of the Hamiltonian operator $\hat{H}$ to second order about the classical phase point $\boldsymbol{z}_t$ is (ignoring possible subtleties involving operator ordering)

$$\hat{H} = H(\boldsymbol{z}_t) + H_{\boldsymbol{z}} \cdot (\hat{\boldsymbol{z}} - \boldsymbol{z}_t) + \tfrac{1}{2}(\hat{\boldsymbol{z}} - \boldsymbol{z}_t) \cdot \mathfrak{H}(\boldsymbol{z}_t) \cdot (\hat{\boldsymbol{z}} - \boldsymbol{z}_t). \tag{39}$$

We drop the linear term, as this does not survive integration over $\Delta \boldsymbol{z}$; matrix elements of the quadratic term are then

$$\begin{aligned} &\tfrac{1}{2}\langle \boldsymbol{z}_t|(\hat{\boldsymbol{z}} - \boldsymbol{z}_t) \cdot \mathfrak{H}(\boldsymbol{z}_t) \cdot (\hat{\boldsymbol{z}} - \boldsymbol{z}_t)|\boldsymbol{z}_t'\rangle \\ &\qquad = \tfrac{1}{2}\langle \boldsymbol{z}_t|\boldsymbol{z}_t'\rangle \big\{\operatorname{tr}[\mathfrak{H}_\Lambda^*] + \tfrac{1}{4}\widetilde{\Delta \boldsymbol{z}}\widetilde{\boldsymbol{S}}(\mathbf{1} + \mathrm{i}\widetilde{\boldsymbol{J}})\mathfrak{H}(\boldsymbol{z}_t)(\mathbf{1} + \mathrm{i}\boldsymbol{J})\boldsymbol{S}\Delta \boldsymbol{z}\big\}. \end{aligned} \tag{40}$$

The first term on the RHS, $\operatorname{tr}[\mathfrak{H}_\Lambda^*]$, arises from the noncommutativity of creation and annihilation operators, while the second term essentially follows from the basic matrix elements

$$\langle \boldsymbol{z}|(\hat{\boldsymbol{z}} - \boldsymbol{z})|\boldsymbol{z}'\rangle = \langle \boldsymbol{z}|\boldsymbol{z}'\rangle \tfrac{1}{2}(\mathbf{1} + \mathrm{i}\boldsymbol{J})(\boldsymbol{z}' - \boldsymbol{z}). \tag{41}$$

We therefore have (keeping in mind that this result is to be integrated over $\Delta \boldsymbol{z}$)

$$\langle \boldsymbol{z}_t|\hat{H}|\boldsymbol{z}_t'\rangle \simeq \langle \boldsymbol{z}_t|\boldsymbol{z}_t'\rangle \big\{H(\boldsymbol{z}_t) + \tfrac{1}{2}\operatorname{tr}[\mathfrak{H}_\Lambda^*] + \tfrac{1}{8}\widetilde{\Delta \boldsymbol{z}}\widetilde{\boldsymbol{S}}(\mathbf{1} + \mathrm{i}\widetilde{\boldsymbol{J}})\mathfrak{H}(\boldsymbol{z}_t)(\mathbf{1} + \mathrm{i}\boldsymbol{J})\boldsymbol{S}\Delta \boldsymbol{z}\big\}. \tag{42}$$

3.3. Time derivatives

The time derivative of the action $\Phi(\boldsymbol{z}'; t)$ is

$$\begin{aligned} \dot{\Phi}(\boldsymbol{z}_t') &= -H(\boldsymbol{z}_t') + \tfrac{1}{2}\widetilde{\boldsymbol{z}}_t' \cdot H_{\boldsymbol{z}}(\boldsymbol{z}_t') &(43a)\\ &= -H(\boldsymbol{z}_t + \boldsymbol{S}\Delta \boldsymbol{z}) + \tfrac{1}{2}(\widetilde{\boldsymbol{z}}_t + \widetilde{\Delta \boldsymbol{z}}\widetilde{\boldsymbol{S}}) \cdot H_{\boldsymbol{z}}(\boldsymbol{z}_t + \boldsymbol{S}\Delta \boldsymbol{z}). &(43b) \end{aligned}$$

Expanding $\dot{\Phi}$ about $\boldsymbol{z}$, we find that terms quadratic in $\Delta \boldsymbol{z}$ cancel, while linear terms will vanish upon integration over $\Delta \boldsymbol{z}$, so that effectively

$$\dot{\Phi}(\boldsymbol{z}_t') \simeq -H(\boldsymbol{z}_t) + \tfrac{1}{2}\widetilde{\boldsymbol{z}}_t H_{\boldsymbol{z}}(\boldsymbol{z}_t) \tag{44}$$

and

$$\mathrm{i}\hbar \partial_t\, \mathrm{e}^{\mathrm{i}\Phi(\boldsymbol{z}';t)/\hbar} \simeq +(H(\boldsymbol{z}_t) - \tfrac{1}{2}\widetilde{\boldsymbol{z}}_t H_{\boldsymbol{z}}(\boldsymbol{z}_t))\, \mathrm{e}^{\mathrm{i}\Phi(\boldsymbol{z}';t)/\hbar}. \tag{45}$$

Direct evaluation of the time derivative of $|z'_t\rangle$ gives

$$\hbar\langle z_t|\partial_t|z'_t\rangle = \langle z_t|z'_t\rangle\left[-\frac{1}{2}\widetilde{\Delta z}\widetilde{S}\cdot(JH_z(z_t)+\dot{S}\Delta z)+\frac{\mathrm{i}}{2}\widetilde{z}_t\cdot(-H_z(z_t)+J\dot{S}\Delta z)\right]. \tag{46}$$

Again discarding terms linear in Δz we obtain

$$\mathrm{i}\hbar\langle z_t|\partial_t|z'_t\rangle \simeq \langle z_t|z'_t\rangle\left[\frac{1}{2}\widetilde{z}_t\cdot H_z(z_t)+\frac{\mathrm{i}}{4}\{\widetilde{\Delta z}\widetilde{S}\widetilde{J}\mathfrak{H}S\Delta z+\widetilde{\Delta z}\widetilde{S}\mathfrak{H}JS\Delta z\}\right]. \tag{47}$$

Note that the terms involving $\frac{1}{2}\widetilde{z}_t\cdot H_z(z_t)$ cancel between (45) and (47). Moreover, the terms quadratic in Δz from (47) combine with those from the matrix element $\langle z_t|\hat{H}|z'_t\rangle$ (42) to yield the quadratic form

$$\tfrac{1}{8}\widetilde{\Delta z}\cdot\widetilde{S}(\mathbf{1}-\mathrm{i}\widetilde{J})\mathfrak{H}(z_t)(\mathbf{1}-\mathrm{i}J)S\cdot\Delta z. \tag{48}$$

3.4. Expanding the integrand

The expression

$$\mathcal{F} = \langle z_t|z'_t\rangle\,\mathrm{e}^{\mathrm{i}\Phi(z';t)/\hbar}\langle z'|z\rangle \tag{49}$$

appears as a factor in the integrand of all the integrals appearing in equation (30). We must expand $\mathcal{F}$ about the initial phase point z, with $z' = z+\Delta z$. In keeping with standard practice, we do not include any terms involving derivatives of monodromy matrix elements [11]. We have

$$\langle z'|z\rangle = \exp\left[-\frac{1}{4\hbar}\widetilde{\Delta z}\cdot\mathbf{1}\cdot\Delta z+\frac{\mathrm{i}}{2\hbar}\widetilde{\Delta z}\cdot Jz\right], \tag{50a}$$

$$\langle z_t|z'_t\rangle \simeq \exp\left[-\frac{1}{4\hbar}\widetilde{\Delta z}\cdot\widetilde{S}S\cdot\Delta z+\frac{\mathrm{i}}{2\hbar}\widetilde{z}_t\cdot JS\cdot\Delta z\right], \tag{50b}$$

$$\Phi(z';t)\simeq\Phi(z;t)-\frac{1}{2}(\widetilde{z}_tJS-\widetilde{z}J)\cdot\Delta z, \tag{50c}$$

so that the linearization of $\mathcal{F}$ is simply

$$\mathcal{F}\simeq\exp\left[\mathrm{i}\frac{\Phi(z;\mathrm{t})}{\hbar}\right]\exp\left[-\frac{1}{4\hbar}\widetilde{\Delta z}\cdot X\cdot\Delta z\right]. \tag{51}$$

3.5. Integrating the quadratic form

The integral of the quadratic form (48) is

$$\int \mathrm{d}^{2N}\Delta z\exp\left[-\frac{1}{4\hbar}\widetilde{\Delta z}\cdot X\cdot\Delta z\right]\frac{1}{8}\widetilde{\Delta z}\widetilde{S}(\mathbf{1}-\mathrm{i}\widetilde{J})\mathfrak{H}(z_t)(\mathbf{1}-\mathrm{i}J)S\Delta z$$
$$=\frac{2^N(2\pi\hbar)^N}{\sqrt{\det[X]}}\frac{1}{4}\,\mathrm{tr}[\widetilde{S}(\mathbf{1}-\mathrm{i}\widetilde{J})\mathfrak{H}(z_t)(\mathbf{1}-\mathrm{i}J)SX^{-1}]. \tag{52}$$

Cyclic invariance of the trace implies

$$\mathrm{tr}[\widetilde{S}(\mathbf{1}-\mathrm{i}\widetilde{J})\mathfrak{H}(z_t)(\mathbf{1}-\mathrm{i}J)SX^{-1}]=\mathrm{tr}[\mathfrak{H}(z_t)(\mathbf{1}-\mathrm{i}J)SX^{-1}\widetilde{S}(\mathbf{1}-\mathrm{i}\widetilde{J})]. \tag{53}$$

From (20)

$$(\mathbf{1}-\mathrm{i}J)SX^{-1}\widetilde{S}(\mathbf{1}-\mathrm{i}\widetilde{J})=-P^*, \tag{54}$$

so that the trace (53) becomes

$$\operatorname{tr}[\mathfrak{H}(z_t)(\mathbf{1}-\mathrm{i}J)SX^{-1}\widetilde{S}(\mathbf{1}-\mathrm{i}\widetilde{J})] = -\operatorname{tr}[\mathfrak{H}(z_t)P^*] \tag{55a}$$

$$= \operatorname{tr}\left[\begin{bmatrix}\mathfrak{H}_A & \mathfrak{H}_B\\ \mathfrak{H}_C & \mathfrak{H}_D\end{bmatrix}\begin{bmatrix}\Gamma\Lambda^{*-1} & \mathrm{i}\Gamma\Lambda^{*-1}\\ \mathrm{i}\Gamma\Lambda^{*-1} & -\Gamma\Lambda^{*-1}\end{bmatrix}\right] \tag{55b}$$

$$= 2\operatorname{tr}[\mathfrak{H}_\Gamma^*\Gamma\Lambda^{*-1}]. \tag{55c}$$

Using

$$\int \mathrm{d}^{2N}\Delta z \exp\left[-\frac{1}{4\hbar}\widetilde{\Delta z}\cdot X\cdot\Delta z\right] = \frac{2^N(2\pi\hbar)^N}{\sqrt{\det[X]}} \tag{56}$$

and rearranging terms in (30), we obtain the equation of motion for the approximate coefficient C,

$$\mathrm{i}\frac{\dot{C}}{C} = \frac{1}{2}\operatorname{tr}[\mathfrak{H}_\Lambda^* + \mathfrak{H}_\Gamma^*\Gamma\Lambda^{*-1}], \tag{57}$$

which is identical with that for $\sqrt{\det[\Lambda^*]}$, (38). This completes our derivation of the HK propagator.

4. Connection between the HK propagator and the TGA

We now show that linearization of the HK propagator yields directly the matrix element of the semiclassical propagator in the TGA. Consider the general coherent state matrix element of the HK propagator

$$\langle z''|\hat{U}_{\mathrm{HK}}(t)|z\rangle = \frac{1}{(2\pi\hbar)^N}\int \mathrm{d}^{2N}z'\langle z''|z'_t\rangle\sqrt{\det[\Lambda^*]}\,\mathrm{e}^{\mathrm{i}\Phi(z';t)/\hbar}\langle z'|z\rangle. \tag{58}$$

Expanding the exponent of the integrand about $z'=z$ as above, integrating over the variable $\Delta z \equiv z'-z$, and using (20), we find that the linearized matrix element (58) is identical with the Littlejohn form of the TGA matrix element [30]

$$\langle z''|\hat{U}_{\mathrm{HK}}(t)|z\rangle \simeq \langle z''|\hat{U}_{\mathrm{TGA}}(t)|z\rangle \tag{59a}$$

$$= \langle z''|\hat{T}(z_t)\hat{M}(S,\sigma)\hat{T}(z)^\dagger|z\rangle\,\mathrm{e}^{\mathrm{i}\Phi(z;t)/\hbar} \tag{59b}$$

$$= \frac{\sigma}{\sqrt{\det[\Lambda]}}\exp\left[-\frac{1}{4\hbar}(\widetilde{z}''-\widetilde{z}_t)\cdot(\mathbf{1}+P)\cdot(z''-z_t)+\frac{\mathrm{i}}{\hbar}\Phi(z;t)\right]. \tag{59c}$$

The form for the propagator (59b) in the TGA describes the wavepacket propagation in an intuitively appealing way as the following sequence of transformations: first, the initial coherent state $|z\rangle$ is translated by the operator $\hat{T}(z)^\dagger$ from the phase point z to the origin; next, the metaplectic operator $\hat{M}(S,\sigma)$ subjects the wavepacket to the transformation induced by the linear canonical transformation associated with the monodromy matrix S; finally, the distorted wavepacket is translated by the operator $\hat{T}(z_t)$ to the phase point z_t, where $z\to z_t$ under the classical time evolution. The wavepacket also accumulates the phase $\Phi(z;t)$.

The mixed semiclassical propagator derived by Baranger *et al* for one-dimensional systems [14] can be expressed very compactly in terms of the matrix elements of the metaplectic operator. For an N degree of freedom system we have

$$\langle x|\hat{U}_{\mathrm{TGA}}(t)|z\rangle = \int \mathrm{d}^N x'\langle x|\hat{T}(z_t)|x'\rangle\langle x'|\hat{M}(S,\sigma)|\mathbf{0}\rangle\,\mathrm{e}^{\mathrm{i}\Phi(z;t)/\hbar}. \tag{60}$$

Using the matrix elements [30]

$$\langle \boldsymbol{x}' | \hat{M}(\boldsymbol{S}, \sigma) | \boldsymbol{0} \rangle = \frac{1}{(\pi\hbar)^{N/4}} \frac{\sigma}{\sqrt{\det[\boldsymbol{A} + \mathrm{i}\boldsymbol{B}]}} \exp\left[-\frac{1}{2\hbar} \widetilde{\boldsymbol{x}}' \cdot \boldsymbol{Y} \cdot \boldsymbol{x}' \right], \tag{61}$$

where the complex symmetric matrix $\boldsymbol{Y} = (\boldsymbol{D} - \mathrm{i}\boldsymbol{C})(\boldsymbol{A} + \mathrm{i}\boldsymbol{B})^{-1}$, and

$$\langle \boldsymbol{x} | \hat{T}(\boldsymbol{z}_t) | \boldsymbol{x}' \rangle = \delta(\boldsymbol{x} - \boldsymbol{x}' - \boldsymbol{q}_t) \exp\left[\frac{\mathrm{i}}{2\hbar} \boldsymbol{p}_t \cdot (\boldsymbol{x} + \boldsymbol{x}') \right], \tag{62}$$

we obtain a compact expression for the TGA propagator matrix element

$$\begin{aligned} \langle \boldsymbol{x} | \hat{U}_{\mathrm{TGA}}(t) | \boldsymbol{z} \rangle &= \frac{1}{(\pi\hbar)^{N/4}} \frac{\sigma}{\sqrt{\det[\boldsymbol{A} + \mathrm{i}\boldsymbol{B}]}} \\ &\quad \times \exp\left[-\frac{1}{2\hbar} (\widetilde{\boldsymbol{x}} - \widetilde{\boldsymbol{q}}_t) \cdot \boldsymbol{Y} \cdot (\boldsymbol{x} - \boldsymbol{q}_t) + \frac{\mathrm{i}}{\hbar} \boldsymbol{p}_t \cdot \left(\boldsymbol{x} - \frac{1}{2} \boldsymbol{q}_t \right) + \mathrm{i}\Phi(\boldsymbol{z}; t) \right]. \end{aligned} \tag{63}$$

To see that (63) is essentially equivalent to the propagator obtained by Baranger *et al* for $N = 1$, note that in the one-dimensional case the ratio γ defined in [14] is

$$\gamma = \frac{\Gamma^*}{\Lambda} = \frac{(A - D) + \mathrm{i}(B - C)}{(A + D) + \mathrm{i}(B - C)}, \tag{64}$$

where Λ, Γ, A, B, C and D are now scalars, so that the quantity $(1-\gamma)/(1+\gamma)$ in the exponent of the Baranger propagator is just a one-dimensional version of $\boldsymbol{Y}$,

$$\frac{1 - \gamma}{1 + \gamma} = (D - \mathrm{i}C)(A + \mathrm{i}B)^{-1}. \tag{65}$$

As Φ is equal to the phase $S_H + (p'q' - p_r q_r)/2$ defined in [14], our result (63) is equivalent to equation (4.29) of Baranger *et al* except for two (related) differences. Thus, an extra contribution to the phase, $\mathrm{i}\mathcal{I}_r/\hbar$, with

$$\mathcal{I}_r = \frac{\hbar}{2} \int \mathrm{d}t [\mathcal{H}_{qq} + \mathcal{H}_{pp}], \tag{66}$$

appears in Baranger's propagator. The presence of this extra phase is associated with the use of the Gaussian averaged Hamiltonian $\mathcal{H}$ in the computation of the action, rather than the Weyl symbol of $\hat{H}$ (essentially the classical Hamiltonian H_C). Both of these subtle but important differences arise when the semiclassical propagator is derived by careful analysis of the coherent state path integral [14]. The properties of the Baranger version of the TGA propagator have recently been studied by Child *et al* [43].

5. Summary and conclusion

By exploiting the underlying symplectic structure of the problem, including the known expressions for matrix elements of metaplectic operators associated with the monodromy matrices of classical trajectories [30, 40], we have been able to present a concise derivation (following Miller [34]) of the semiclassical Herman–Kluk propagator [5]. Moreover, using the matrix elements of the metaplectic operator we are able to straightforwardly demonstrate that the propagator obtained by Baranger *et al* for one-dimensional systems [14] is, apart from a couple of subtle differences, identical with the expression for the propagator in the thawed Gaussian approximation given by Littlejohn [30].

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