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A semiclassical hybrid approach to many particle quantum dynamics

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We analytically derive a correlated approach for a mixed semiclassical many particle dynamics, treating a fraction of the degrees of freedom by the multitrajectory semiclassical initial value method of Herman and Kluk [Chem. Phys. **91**, 27 (1984)] while approximately treating the dynamics of the remaining degrees of freedom with fixed initial phase space variables, analogously to the thawed Gaussian wave packet dynamics of Heller [J. Chem. Phys. **62**, 1544 (1975)]. A first application of this hybrid approach to the well studied Secrest-Johnson [J. Chem. Phys. **45**, 4556 (1966)] model of atom-diatomic collisions is promising. Results close to the quantum ones for correlation functions as well as scattering probabilities could be gained with considerably reduced numerical effort as compared to the full semiclassical Herman-Kluk approach. Furthermore, the harmonic nature of the different degrees of freedom can be determined *a posteriori* by comparing results with and without the additional approximation. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2213255]

I. INTRODUCTION

The semiclassical initial value representation (IVR) of the quantum mechanical propagator of Herman and Kluk,[1] (HK) has gained renewed interest due to the work of Kay more than ten years ago.[2–4] In these and in many following papers, results for a large variety of anharmonic systems have been presented, ranging from nuclear[5–10] to electronic dynamics[11–13] as well as to the dynamics on coupled electronic surfaces.[9,14] An overview of IVR methods and their applications as well as many more references are contained in two recent reviews.[15,16] As a by-product of all these studies, it turned out that for long times, IVR results are superior to the ones obtained from the single trajectory thawed Gaussian wave packet dynamics (TGWD) developed in the 1970s.[17,18] This is at the cost of having to propagate a large number of trajectories, however. Especially for systems with many degrees of freedom (DOFs), this fact becomes cumbersome. Several more or less approximative approaches to the full HK IVR expression have therefore been made. Very prominent among them are the linearization approximation and the forward-backward approach to time correlation functions. These methods have been pioneered by Miller and co-workers and have proven their usefulness for many molecular systems, as reviewed, e.g., in Ref. 9.

Recently, it has been stated that the TGWD can be derived from the HK propagator applied to a Gaussian wave packet as initial state by performing an expansion of the exponent to second order around the phase space center of the initial wave packet.[19] Based on the observation that TGWD is not complex enough to account for strong anharmonicity but is an excellent approximation for systems close to harmonic, in a system with N DOFs, one should identify those n DOFs that will behave almost harmonically in the course of time and propagate them with one single initial condition in the $2n$-dimensional phase space projection, analogously to TGWD, while retaining the full HK expression for the other $N-n$ DOFs. Thus, the central idea of this paper is to perform the transition from HK to TGWD for a selected set of DOFs analytically. In this way semiclassical hybrid wave and/or correlation functions for systems with many DOFs can be calculated consistently without any *ad hoc* assumptions on the coupling between the differently treated subsets of DOFs. As will be shown in the following, the dynamics of the trajectories as well as their stability properties are coupled and in addition they enter the final expression in a nonfactorizable way.

Other hybrid approaches, especially so-called quantum-classical methods, are frequently used in the literature. For a recent textbook on this topic, which contains many useful references, we would like to refer the reader to Ref. 20. In these quantum-classical approaches, part of the DOFs, sometimes called the "system" DOFs or the "core" DOFs, is treated fully quantum mechanically while the remaining part, the so-called "bath" DOFs or the "reservoir" DOFs, is treated classically. One can, however, also use other approximate theories to treat the reservoir.[21] Closer in spirit to the work to be presented here than quantum hybrid methods are the semiclassical hybrid approaches of Sun and Miller[22] and of Ovchinnikov and Apkarian.[23] In Ref. 22 as well as in Ref. 23 the core system is treated via the semiclassical HK IVR, while in Ref. 22 the reservoir is treated classically. In Ref. 23 the bath DOFs have been dealt with in an *ad hoc* manner by using primitive single trajectory frozen Gaussian wave packets without a time-dependent prefactor, whereas here we use TGWD. The most important aspect of the method to be developed, however, is the analytic derivation from the full HK expression of the entanglement (or correlation) between the dynamics of the different subgroups of DOFs in the exponent and in the prefactor of the final wave function. A numerical application for a two-DOFs system will then be presented in order to elucidate the potential usefulness of the methodology.

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The paper is organized in the following way. In Sec. II, after a brief introduction into the HK methodology and its connection to TGWD, the semiclassical hybrid approach will be derived. In Sec. III, we present numerical results for two degrees of freedom. As a test system we employ the Secrest-Johnson model of a harmonic DOF coupled to a translational DOF with an exponential repulsion.[24] Conclusions are given in Sec. IV. Some general formulas for N-DOFs systems needed throughout the paper are collected in the Appendices.

II. A SEMICLASSICAL HYBRID APPROACH

In order to set the stage for the derivation of the hybrid approach, we first review the HK propagator and the derivation of the more approximate TGWD from it. The proposed methodology will then be introduced and an explicit expression for the wave function will be presented.

A. The HK propagator and its relation to TGWD

The methodology to be developed in this paper is based on the multiple trajectory frozen Gaussian approach to semiclassical dynamics as pioneered by Heller[25] and improved by Herman and Kluk.[1] The latter authors derived the prefactor in the integral expression of the semiclassical propagator for an N degrees of freedom system,

$$K^{\mathrm{HK}}(\mathbf{x},t;\mathbf{x}',0)=\int \frac{d\mathbf{p}'d\mathbf{q}'}{(2\pi\hbar)^N}\sqrt{\det \mathbf{h}}\langle \mathbf{x}|g(\mathbf{p}_t,\mathbf{q}_t)\rangle \times\exp\left\{\frac{i}{\hbar}S(\mathbf{p}',\mathbf{q}',t)\right\}\langle g(\mathbf{p}',\mathbf{q}')|\mathbf{x}'\rangle. \quad (1)$$

This propagator is given in terms of Gaussian wave functions,

$$\langle \mathbf{x}|g(\mathbf{p},\mathbf{q})\rangle=\left(\frac{\det\gamma}{\pi^N}\right)^{1/4}\exp\left\{-\frac{1}{2}(\mathbf{x}-\mathbf{q})^T\gamma(\mathbf{x}-\mathbf{q}) + \frac{i}{\hbar}\mathbf{p}^T\cdot(\mathbf{x}-\mathbf{q})\right\}, \quad (2)$$

with constant width parameter matrix γ, which we here assume to be the same for both Gaussians in Eq. (1). Classical dynamics enters the HK IVR of the propagator via the classical trajectories $(\mathbf{p}_t=\mathbf{p}(\mathbf{p}',\mathbf{q}',t),\mathbf{q}_t=\mathbf{q}(\mathbf{p}',\mathbf{q}',t))$, which are initial value solutions of Hamilton's equations, and via the classical action function $S(\mathbf{p}',\mathbf{q}',t)=\int_0^t Ldt'$ with the Lagrangian $L=T-V$, denoting the difference between kinetic and potential energies.

Furthermore, and very importantly, the classical stability information plays the central role in determining the time-dependent part of the prefactor of the HK propagator. For later reference we are therefore reviewing the properties of the symplectic stability (or monodromy) matrix in Appendix A. With the quantities defined there and in a slight generalization of the original result of Herman and Kluk, the matrix $\mathbf{h}$, whose determinant appears in the HK propagator, is given by

$$\mathbf{h}=\frac{1}{2}\left(\mathbf{m}_{11}+\gamma\mathbf{m}_{22}\gamma^{-1}-i\hbar\gamma\mathbf{m}_{21}-\frac{1}{i\hbar}\mathbf{m}_{12}\gamma^{-1}\right), \quad (3)$$

for diagonal γ matrix with real and positive elements. This generalization has been derived by using the principle of stationary phase equivalence together with block matrix inversion in Appendix B. The generalized prefactor matrix above has been used previously (see, e.g., Ref. 26) and even more general versions of that matrix have been devised by Kay (see, e.g., Ref. 16). There, the only restriction on the width parameter matrix is the positive definiteness of its real part. For our purposes in this paper, the expression above is sufficient, however.

In applications to molecular and atomic quantum dynamics, the HK propagator is most frequently used to calculate wave functions and/or correlation functions. Applying the propagator expression in Eq. (1) to a Gaussian wave function, centered around $(\mathbf{p}_\alpha,\mathbf{q}_\alpha)$, as an initial state according to

$$\Psi_\alpha(\mathbf{x},t)=\int d\mathbf{x}' K^{\mathrm{HK}}(\mathbf{x},t;\mathbf{x}',0)\langle \mathbf{x}'|\Psi_\alpha(0)\rangle, \quad (4)$$

is eased by the fact that the integral over $\mathbf{x}'$ can be done analytically by using

$$\langle g(\mathbf{p}',\mathbf{q}')|\Psi_\alpha(0)\rangle=\exp\left\{-\frac{1}{4}(\mathbf{q}'-\mathbf{q}_\alpha)^T\gamma(\mathbf{q}'-\mathbf{q}_\alpha) + \frac{i}{2\hbar}(\mathbf{q}'-\mathbf{q}_\alpha)^T\cdot(\mathbf{p}'+\mathbf{p}_\alpha) - \frac{1}{4\hbar^2}(\mathbf{p}'-\mathbf{p}_\alpha)^T\gamma^{-1}(\mathbf{p}'-\mathbf{p}_\alpha)\right\}, \quad (5)$$

for the overlap between the initial wave function and the Gaussian depending on $(\mathbf{p}',\mathbf{q}')$ in the HK propagator (1). The remaining phase space integration is then usually performed by Monte Carlo integration.[27]

In order to calculate the cross correlation with another (final) Gaussian,

$$c_{\beta,\alpha}(t)=\int d\mathbf{x}\Psi_\beta^*(\mathbf{x})\Psi_\alpha(\mathbf{x},t), \quad (6)$$

also the integration over $\mathbf{x}$ will be performed analytically. The remaining numerical task is again the propagation of the trajectories and the phase space integration of Eq. (1), to be done, e.g., by the Monte Carlo method. For future use, we want to mention that also correlation functions involving eigenfunctions of the harmonic oscillator can be performed by using the helpful formula[28]

$$\int da\,\exp\{-(a-b)^2\}H_n(a)=\sqrt{\pi}b^n2^n, \quad (7)$$

where H_n denote the Hermite polynomials.

Recalling that the expression for a propagated Gaussian in Eq. (4) contains a phase space integration which can also be performed approximatively analytically, one can derive Heller's TGWD from HK. In Appendix C, we explicitly show how this is done by using a second order expansion of the exponent together with a zeroth order expansion of the

prefactor around the center of the initial Gaussian wave function. From this expression one could either directly calculate the correlation function, or alternatively one could use the full HK correlation function of Eq. (6) and derive a TGWD correlation function expression in an analogous fashion as in Appendix C. In both cases the end result reads

$$c_{\beta,\alpha}(t) = 1/\sqrt{\det(\mathbf{h}^*)}\,\exp\left\{-\frac{1}{4}(\mathbf{q}_\beta-\mathbf{q}_{\alpha,t})^T\left(\mathbf{m}_{11}+\frac{1}{i\hbar}\mathbf{m}_{12}\gamma^{-1}\right)(\mathbf{h}^*)^{-1}\gamma(\mathbf{q}_\beta-\mathbf{q}_{\alpha,t})-\frac{1}{4\hbar^2}(\mathbf{p}_\beta-\mathbf{p}_{\alpha,t})^T(\mathbf{m}_{22}\gamma^{-1}+i\hbar\mathbf{m}_{21})(\mathbf{h}^*)^{-1}\right.$$
$$\left.\times(\mathbf{p}_\beta-\mathbf{p}_{\alpha,t})+\frac{i}{2\hbar}(\mathbf{q}_\beta-\mathbf{q}_{\alpha,t})^T\left[\left(\mathbf{m}_{11}+\frac{1}{i\hbar}\mathbf{m}_{12}\gamma^{-1}\right)(\mathbf{h}^*)^{-1}\mathbf{p}_\beta+\gamma(\mathbf{m}_{22}\gamma^{-1}+i\hbar\mathbf{m}_{21})(\mathbf{h}^*)^{-1}\mathbf{p}_{\alpha,t}\right]+\frac{i}{\hbar}S(\mathbf{p}_\alpha,\mathbf{q}_\alpha,t)\right\}. \quad (8)$$

We stress that in contrast to Eq. (6) this is now a single trajectory result requiring only the propagation of one classical trajectory and its stability matrix elements.

B. Semiclassical hybrid dynamics

So far we have shown the mutual relation of results from the literature. The fact that TGWD can be derived straightforwardly (as shown in Appendix C) from the HK propagator now serves as the starting point for the derivation of our hybrid methodology. To proceed, we first divide our total number of DOFs into n that shall be dealt with on the level of TGWD and the remaining $N-n$ we want to propagate retaining the full complexity of the semiclassical HK method. An expression for the wave function in this mixed semiclassical approach, which we would like to term semiclassical hybrid dynamics, will be given in the following.

For the propagation of a Gaussian wave function with the HK propagator, we use Eq. (4) together with (1), leading to

$$\Psi_\alpha(\mathbf{x},t) = \int \frac{d\mathbf{p}'d\mathbf{q}'}{(2\pi\hbar)^N}\sqrt{\det\mathbf{h}}\langle\mathbf{x}|g(\mathbf{p}_t,\mathbf{q}_t)\rangle \times\exp\left\{\frac{i}{\hbar}S(\mathbf{p}',\mathbf{q}',t)\right\}\langle g(\mathbf{p}',\mathbf{q}')|\Psi_\alpha(0)\rangle. \quad (9)$$

Further on, we denote the momenta and coordinates of that portion of the total number of DOFs, which we want to treat with the multitrajectory approach, i.e., the core DOFs, by the vectors $\mathbf{p}_1'$ and $\mathbf{q}_1'$, respectively. For the remaining DOFs we use the phase space variables $\mathbf{p}_2'$ and $\mathbf{q}_2'$. We stress that we do not assume the factorization of the expression in Eq. (9) into a term that only depends on $(\mathbf{p}_{1,t},\mathbf{q}_{1,t})$ and one that depends only on $(\mathbf{p}_{2,t},\mathbf{q}_{2,t})$. However, as will be seen shortly, the dynamics as well as the final expression for the wave function are coupled.

The central idea in the derivation of the hybrid approach is to perform the phase space integration in Eq. (9) over those variables which carry the index 2 by using the Gaussian expansion approximation. That is, we expand the *total* exponent around the $2n$-dimensional phase space projection $(\mathbf{p}_{2,\alpha},\mathbf{q}_{2,\alpha})$ of the center of the initial wave packet up to second order, make a zero order expansion of the prefactor, and then perform the remaining Gaussian integral. In the case that all of the N DOFs would be contained in the phase space variables labeled by the index 2, this would lead to the TGWD of Heller, as sketched in Appendix C. Here, however, the integration over the remaining phase space DOFs labeled by index 1 still remains and after calculus analogous to the one described in Appendix C, we end up with the expression,

$$\Psi_\alpha(\mathbf{x},t) = \int \frac{d\mathbf{p}_1'd\mathbf{q}_1'}{(2\hbar)^N\pi^{N-n}}\sqrt{\frac{\sqrt{\det\gamma}\det\mathbf{h}}{\pi^{N/2}\det\mathbf{A}_2}} \times\exp\left\{\frac{1}{4}\mathbf{b}_2^T\mathbf{A}_2^{-1}\mathbf{b}_2+c_2\right\}, \quad (10)$$

for the wave function. In the exponent we have used the definition

$$\mathbf{A}_2 \equiv \begin{pmatrix} \gamma_2^{-1}/(4\hbar^2)+\tilde{\mathbf{r}}\tilde{\mathbf{m}}_{21}/2 & i/(4\hbar)+\tilde{\mathbf{m}}_{21}^T\tilde{\mathbf{s}}^T/2 \\ i/(4\hbar)+\tilde{\mathbf{s}}\tilde{\mathbf{m}}_{21}/2 & \gamma_2/4+\tilde{\mathbf{s}}\tilde{\mathbf{m}}_{22}/2 \end{pmatrix}, \quad (11)$$

which is a $2n\times 2n$ matrix and contains the abbreviations

$$\tilde{\mathbf{r}} = \tilde{\mathbf{m}}_{21}^T\gamma+\frac{i}{\hbar}\tilde{\mathbf{m}}_{11}^T, \quad (12)$$

$$\tilde{\mathbf{s}} = \tilde{\mathbf{m}}_{22}^T\gamma+\frac{i}{\hbar}\tilde{\mathbf{m}}_{12}^T. \quad (13)$$

In contrast to the case of TGWD which we deal with in Appendix C, the stability matrices $\tilde{\mathbf{m}}_{ij}$ are now rectangular $N\times n$ matrices, containing the derivatives with respect to the initial variables with index 2. The submatrix γ_2 of the width parameter matrix is an $n\times n$ square matrix.

The $2n$-component row vector in the exponent of Eq. (10) is defined by

$$\mathbf{b}_2^T \equiv (\mathbf{x}-\tilde{\mathbf{q}}_{\alpha,t})^T(\tilde{\mathbf{r}}^T,\tilde{\mathbf{s}}^T), \quad (14)$$

where $\tilde{\mathbf{q}}_{\alpha,t}=(\mathbf{q}_1(\tilde{\mathbf{p}}_\alpha,\tilde{\mathbf{q}}_\alpha,t),\mathbf{q}_2(\tilde{\mathbf{p}}_\alpha,\tilde{\mathbf{q}}_\alpha,t))$ with $\tilde{\mathbf{p}}_\alpha=(\mathbf{p}_1',\mathbf{p}_{2,\alpha})$ and $\tilde{\mathbf{q}}_\alpha=(\mathbf{q}_1',\mathbf{q}_{2,\alpha})$ are the trajectories which evolve according to the fully coupled classical equations of motion. The initial and final positions as well as the correspondingly defined momenta now have a tilde, because in contrast to the respective quantities without tilde, here only the initial conditions of the n DOFs are restricted to the initial wave packet center while the phase space of the remaining DOFs is being sampled. Finally

$$c_2 \equiv -\frac{1}{2}(\mathbf{x}-\tilde{\mathbf{q}}_{\alpha,t})^T\gamma(\mathbf{x}-\tilde{\mathbf{q}}_{\alpha,t})+\frac{i}{\hbar}\tilde{\mathbf{p}}_{\alpha,t}^T\cdot(\mathbf{x}-\tilde{\mathbf{q}}_{\alpha,t})$$
$$+\frac{i}{\hbar}S(\tilde{\mathbf{p}}_\alpha,\tilde{\mathbf{q}}_\alpha,t)-\frac{1}{4}(\mathbf{q}_1'-\mathbf{q}_{1,\alpha})^T\gamma_1(\mathbf{q}_1'-\mathbf{q}_{1,\alpha})$$
$$+\frac{i}{2\hbar}(\mathbf{q}_1'-\mathbf{q}_{1,\alpha})^T\cdot(\mathbf{p}_1'+\mathbf{p}_{1,\alpha})$$
$$-\frac{1}{4\hbar^2}(\mathbf{p}_1'-\mathbf{p}_{1,\alpha})^T\gamma_1^{-1}(\mathbf{p}_1'-\mathbf{p}_{1,\alpha}) \quad (15)$$

with the $(N-n)\times(N-n)$ width parameter submatrix γ_1 is the definition of the remaining scalar in Eq. (10).

Some further remarks concerning the hybrid wave function approach seem to be in order. Firstly, the way the stability information enters Eq. (10) via Eqs. (11) and (14) shows that not only the classical equations of motion are coupled but also the wave function is not factorizable and thus shows "correlation" between the different subgroups of DOFs. Secondly, in the numerics the prefactor in the expression (10) above has to be determined in such a fashion that it becomes a continuous function of time. Whenever the radicant crosses the branch cut of the square root the phase has to be adjusted accordingly, in the same way, this is done in standard HK calculations. Finally, for the calculation of correlation functions, it turns out that the **x** integration in Eq. (6) is best performed before the hybrid approximation is done. Then an expression analogous to the one for the wave function can be derived, which we do not reproduce here due to its close analogy to the wave function, but which will be used in our numerical calculations below.

III. A NUMERICAL EXAMPLE

The semiclassical hybrid dynamics outlined in the previous section allows for the calculation of wave and/or correlation functions for any system with at least two DOFs. In the following, as a first test, we will apply the method to an inelastic scattering model in which a reaction is not allowed.

A. Secrest-Johnson model

A very popular benchmark model used in many semiclassical studies so far is the Secrest-Johnson (SJ) model. It describes an inelastic scattering process and consists of a translational DOF x_1 and an internal harmonic DOF x_2. In appropriate reduced units its Hamiltonian is given by[24]

$$H(\mathbf{p},\mathbf{x})=\frac{p_1^2}{2\mu}+\frac{p_2^2}{2}+\exp\{\alpha(x_2-x_1)\}+\frac{1}{2}x_2^2. \quad (16)$$

With the parameters $\mu=2/3$ and $\alpha=0.3$ this corresponds to the physical situation of a helium atom colliding collinearly with a hydrogen molecule. In the original work of SJ, coupled channel quantum calculations of the inelastic scattering probabilities have been performed. Furthermore, in his seminal classical S-matrix work,[29] Miller pointed out the importance of interference effects in the distribution of final vibrational states.

A collection of early references on the applications of Gaussian wave packet methods to the SJ model (and also many other molecular problems) can be found in the review of Heller.[18] Out of the more recent literature on semiclassical IVR methods applied to the SJ model, we would like to mention the work of Skinner and Miller[30,31] applying, e.g., different semiclassical approximations such as the linearization approximation and the forward-backward approach and the work of Elran and Kay,[32] introducing a new semiclassical method for S-matrix extraction in the case of collinear collisions.

B. The *S* matrix

In the following we are interested in the inelastic scattering probabilities for the SJ model as a function of energy. A possible way to extract these quantities, alternative to the one presented in Ref. 32 which is applicable to semiclassical as well as full quantum dynamics, is given by the Tannor-Weeks S-matrix formalism.[33] In this approach, the S matrix, from which the corresponding probabilities can be calculated by taking the absolute square, is given by

$$S_{\beta,n';\alpha,n}(E)=\frac{(2\pi\hbar)^{-1}}{\eta^*_{\beta,n'}(E)\,\eta_{\alpha,n}(E)}\int c_{\beta,n';\alpha,n}(t)e^{iEt/\hbar}dt, \quad (17)$$

i.e., the Fourier transform of a cross-correlation function of the form

$$c_{\beta,n';\alpha,n}(t)=\langle\Psi_{\beta,n'}|\exp\{-iHt/\hbar\}|\Psi_{\alpha,n}\rangle, \quad (18)$$

where the initial wave function is given by the product,

$$\Psi_{\alpha,n}(\mathbf{x})=g_\alpha(x_1)\phi_n(x_2), \quad (19)$$

of an arbitrary incoming Gaussian translational wave function times an eigenstate of the internal Hamiltonian [for the Hamiltonian in Eq. (16) this is an eigenstate to the energy E_n of the harmonic degree of freedom]. The normalization factors in the expression for the S matrix are then given by

$$\eta_{\alpha,n}(E)=\sqrt{\frac{\mu}{2\pi\hbar k_n}}\int e^{-ik_nx_1}g_\alpha(x_1)dx_1, \quad (20)$$

with $k_n=\sqrt{2\mu(E-E_n)}/\hbar$. They "cancel" the arbitrariness of the initial wave function and can be calculated exactly analytically from the Gaussian translational part g_α of the total wave function.

In order to extract inelastic scattering probabilities for the SJ model, both the initial and the final wave functions are centered around large positive values of x_1 where the exponential part of the potential is almost constantly zero. The momentum of the initial state $p_{1,\alpha}$ is pointing inward, whereas that of the final state $p_{1,\beta}$ has to be outgoing, however.

C. HK versus full quantum mechanics

In order to get an impression of how well the full semiclassical formalism (i.e., HK for both DOFs) does, we first compare the HK results to the full quantum results for the correlation functions and the corresponding inelastic scattering probabilities $P_{n',n}(E)$, for $n=0$ and $n'=0,1,2$ in an energy range from 3 to 12 in the reduced units of Secrest and Johnson, which are used throughout the remainder of this

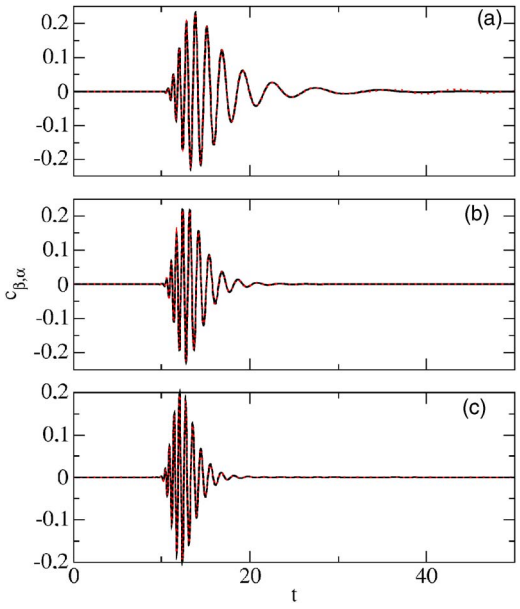

FIG. 1. Comparison of the real parts of the correlation functions $c_{\beta,n';\alpha,n}$ that enter the calculation of the S matrix for $n=0$ and (a) $n'=0$, (b) $n'=1$, and (c) $n'=2$. Full line: quantum result. Dotted line: HK result.

paper. For the quantum calculations, which were obtained by the split operator fast Fourier transform (FFT) method,[34] we have used a time step of $\Delta t=0.02$ and a (nonoptimized) grid of 2048×128 points extending from -40 to 700 in the translational and from -15 to 15 in the internal coordinate, together with absorbing boundary conditions only for the translational coordinate. For the HK calculations, we used the same time step, for which a leap frog symplectic propagation scheme for Hamilton's equations was appropriate.[35] In order to converge not only the time signals but also the probabilities derived from them, we had to propagate 10^6 trajectories.

To extract the probabilities from the corresponding S-matrix elements, we use wave packet parameters of $p_{1,\alpha}=-3$, $q_{1,\alpha}=10$ for the incoming and $p_{1,\beta}=3$, $q_{1,\beta}=20$ for the outgoing Gaussians. The width parameter was chosen to be $\gamma_1=1.5$. The wave function in the harmonic degree of freedom is again proportional to a Gaussian. For the initial α wave function it is chosen as the ground state of the harmonic oscillator, leading to $\gamma_2=1$, whereas for the final β wave function it is taken as the ground state Gaussian times the appropriate Hermite polynomial. The generalized expression for the prefactor matrix (3) in the HK expression allows us to take this choice of different γ matrix elements for the different degrees of freedom. With the choice of parameters made above, we cover the energy range in which we are interested quite well, as will be seen below.

In Fig. 1, the semiclassical full HK as well as the quantum mechanical correlation functions that enter the S-matrix expression are depicted. The HK method reproduces the quantum results very well, the only significant deviation from the quantum result can be observed for times around 50 in the elastic 0-0 case. This leads to an extremely good coincidence also between the corresponding probabilities, which is demonstrated in Fig. 2. Here, in addition to our quantum and semiclassical results we have also depicted the

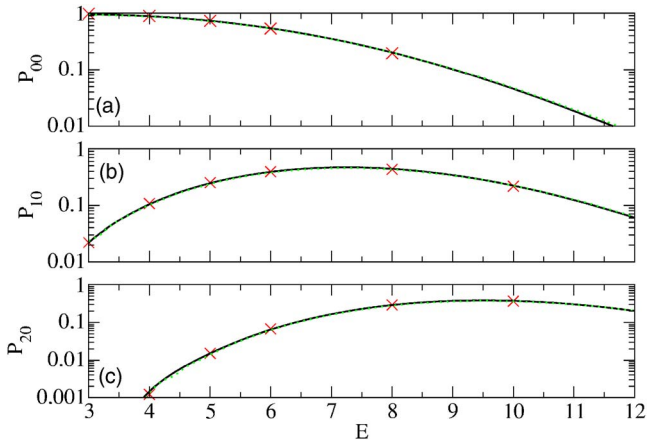

FIG. 2. Elastic and inelastic scattering probabilities for the SJ model as a function of absolute energy (a) P_{00}, (b) P_{10}, and (c) P_{20}. Full line: quantum result. Dotted line: HK result. Crosses: SJ's original results.

Secrest-Johnson values[24] for the inelastic case and in the elastic case the values inferred by Heller[17] apart from the result for $E=10$ due to the missing information on the 0-5 transition in Ref. 24. For the wave packet parameters we have chosen, the central total energy of our incoming packet is at 7.25, and thus at much lower and much higher energies, the quantum result as well as the semiclassical result are hard to converge due to the fact that these energies are not well represented in the initial Gaussian wave packet. However, the quantum result at $E=3$ is only around 1% too small compared to the inferred result for the 0-0 transition tabulated in the work of Heller[17] whereas the semiclassical result is only a bit worse. Over most of the energy range depicted, however, the agreement between the two results presented here and the ones of Secrest and Johnson[24] is excellent for the 0-0 as well as for the 0-1 and the 0-2 transitions.

D. Hybrid results versus full quantum mechanics

Now we compare hybrid results with the full quantum ones and also with the full TGWD result, where only one single trajectory is used. We did several calculations for the same wave packet parameters as in the previous subsection. In order to keep things simple we concentrate now on the elastic case and consider only the probability P_{00} and the corresponding correlation functions. We start with the discussion of the correlation functions.

In Fig. 3 we compare the quantum result (full line) for the correlation function with the one trajectory TGWD result for *both* DOFs (dotted curve). There is good agreement for short times. For longer times, however, the TGWD tends to zero well before the full quantum mechanics does and also the low frequency component in the quantum result is underestimated by full TGWD. As a first check of the hybrid method we now used HK semiclassics in the *second* (i.e., close to harmonic) DOF (dashed line), thus actually reversing the role of q_1' and q_2' in Eq. (10). For short times the amplitude of the time signal has improved a little bit. For longer times the lagging of the signal compared to the quantum one is, however, even a bit more pronounced now. In general, as could be expected, there is no dramatic change

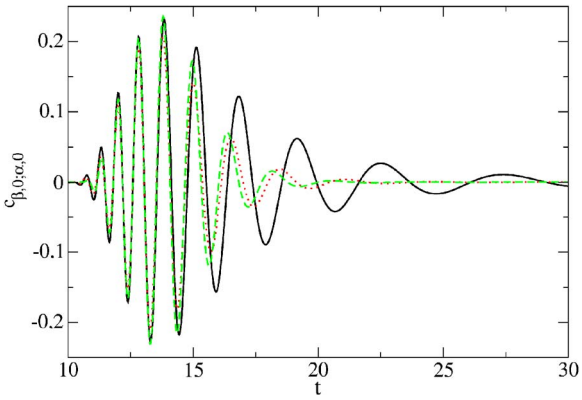

FIG. 3. Comparison of the real parts of the correlation functions $c_{\beta,0;\alpha,0}$ that enter the calculation of the S matrix. Full line: quantum result. Dotted line: full TGWD. Dashed line: hybrid result with 10^4 trajectories sampling the phase space of the "almost harmonic" DOF.

compared to the one trajectory result, although we used 10^4 initial conditions in the Monte Carlo sampling of the phase space of the harmonic DOF. This is due to the fact that the almost harmonic DOF is well described by a single initial phase space point. Trying to improve on this by using HK for this DOF is not leading to a tremendous effect.

Let us now, however, use the proposed hybrid method with 5×10^4 points sampling the phase space of the *first* DOF and TGWD for the second. The time series and the corresponding probabilities are then converged for the whole range of energies to be depicted below. For the time series, a similar comparison as above is made in Fig. 4, where we again display the quantum and the full TGWD result together with the hybrid result using the same coding for the lines as in Fig. 3. This time, due to the fact that we used HK semiclassics for the more anharmonic DOF, a tremendous improvement with respect to full TGWD is observable, leading to a result which is almost indistinguishable from the quantum one! In addition to this good numerical agreement, we also would like to mention that by using semiclassical hybrid dynamics with HK semiclassics applied to different DOFs, one can judge which ones will behave most anharmonically in the course of time.

To complete this section, we will finally compare the

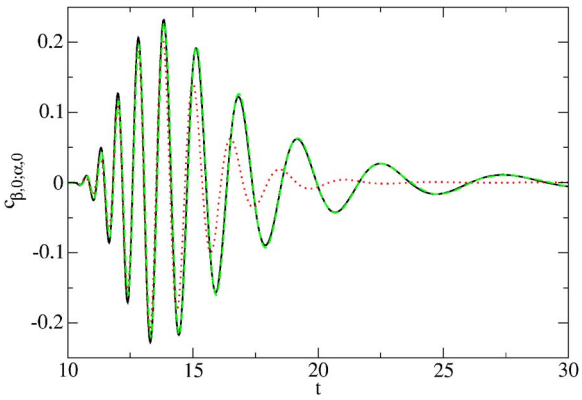

FIG. 4. Comparison of the real parts of the correlation functions $c_{\beta,0;\alpha,0}$ that enter the calculation of the S matrix. Full line: quantum result. Dotted line: full TGWD. Dashed line: hybrid result with 5×10^4 trajectories sampling the phase space of the "anharmonic" DOF.

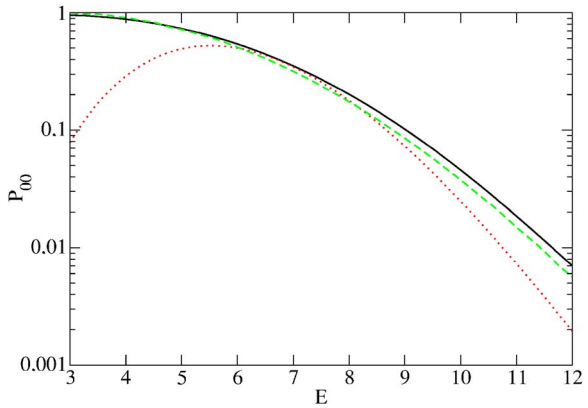

FIG. 5. Elastic scattering probability P_{00} for the SJ model as a function of absolute energy. Full line: quantum result. Dotted line: full TGWD. Dashed line: semiclassical hybrid dynamics.

probabilities P_{00} derived from the correlation functions of Fig. 4. This is done in Fig. 5, where the quantum result (full line) together with the full TGWD (dotted line) and the hybrid result sampling the phase space of the first DOF (dashed line) are displayed. The discrepancy of the full TGWD and full quantum correlation functions at long times clearly manifests itself in the low energy discrepancy of the corresponding probabilities. In addition also the high energy results are poor. Reasonable results are gained only in a narrow energy interval around the central total energy of the incoming wave packet. Compared to this, the agreement of the hybrid result with the quantum one for P_{00} is much better, but not as perfect as for the respective correlation functions, which is due to the fact that small differences of the time signal, barely visible or not depicted in the interval shown in Fig. 4, are getting magnified in the probability by the normalization procedure in extracting the S matrix. Nevertheless, the hybrid method offers a dramatic improvement as compared to the one trajectory case and helps in keeping the numerical effort down. Here we had to use a total of 5×10^4 trajectories as compared to the full semiclassical HK calculation, where 10^6 trajectories were needed to converge the results.

IV. CONCLUSIONS

We have analytically derived a semiclassical hybrid dynamics for many particle quantum systems, which allows for the selective treatment of nearly harmonic variables by a fixed initial condition in the spirit of Heller's TGWD. The remaining variables are treated on the footing of the semiclassical Herman-Kluk method. The proposed methodology is more approximate than HK applied to all N DOFs but contains correlations between the subgroups of DOFs not only in the classical dynamics but also in the resulting expression for the wave function, Eq. (10). Furthermore, it seems to be worthwhile to note that a direct route of well-defined analytic approximations from the Feynman path integral,[36] via the van Vleck–Gutzwiller propagator[37,38] and the full HK IVR expression, exists, which leads to the final expression for the hybrid wave function.

The applicability of the method has been demonstrated by using the Secrest-Johnson model as a test case. The numerical results are proving that the methodology works and gives reasonable results with reduced numerical effort, although the test case is a bit unfavorable due to the fact that Monte Carlo integration in the case of just one anharmonic variable is not the method of choice. Therefore in our test case the numerical effort compared to the full HK calculation is only a bit more than one order of magnitude lower (although we used quasirandom numbers for the sampling,[39] Monte Carlo integration becomes more favorable compared to integration using a grid for an increasing number of phase space variables). We have therefore good reason to believe that the most favorable domain of the hybrid method are systems where *several* DOFs are treated using the HK method and *many* other DOFs are dealt with by TGWD. For example, in the case of the semiclassical calculation of photodetachment spectra, which has been dealt with using a full HK approach for up to 15 coupled DOFs in Ref. 40, some of the almost harmonic cluster DOFs could possibly be dealt with by using TGWD. This special system and also systems with a general Caldeira-Leggett-type system-bath Hamiltonian[41] (with a finite number of bath oscillators) are possible candidates for further explorations of the capability of the presented methodology.

Furthermore, in the SJ test case, the DOF to be treated by the more elaborate HK method turned out to be the one with the exponential repulsion. A treatment of the almost harmonic DOF with HK led only to a marginal improvement. Therefore the methodology also allows to determine which variables behave more and which behave less harmonically. In case one would be able to distinguish the more harmonic and the more anharmonic DOFs, as mentioned above, a reduction of the numerical effort can be achieved. If this distinction is not easily possible *a priori*, one out of several similar DOFs (e.g., bath oscillators with almost the same frequency) could be tested with respect to its degree of harmonicity by applying first TGWD and then full HK to it. If there is no significant improvement of the results through HK, then all those similar DOFs can be safely treated by TGWD. In the system-bath case, one does *not* have to treat all the bath DOFs in the TGWD fashion, however. One could, e.g., add those bath oscillators to the "system" and thus treat them with HK, which are expected to be "driven resonantly" by the coupling and thus have a more anharmonic nature than those with frequencies far away from any resonances.

We thus hope that a lot of systems displaying interesting many particle physics could be better understood and/or treated in the first place by the proposed methodology.

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APPENDIX A: STABILITY MATRIX FOR *N* DOFs

In this appendix we recall the definition and some properties of the multidimensional form of the stability matrix. For small initial deviations in phase space $\delta\mathbf{p}'$, $\delta\mathbf{q}'$ one defines

$$\delta\mathbf{p}_t = \mathbf{m}_{11}\delta\mathbf{p}' + \mathbf{m}_{12}\delta\mathbf{q}', \tag{A1}$$

$$\delta\mathbf{q}_t = \mathbf{m}_{21}\delta\mathbf{p}' + \mathbf{m}_{22}\delta\mathbf{q}', \tag{A2}$$

for the final deviations, where the $N\times N$ matrices $\mathbf{m}_{ij}(i,j=1,2)$ are submatrices of the so-called stability (or monodromy) matrix

$$\mathbf{M} \equiv \begin{pmatrix} \mathbf{m}_{11} & \mathbf{m}_{12} \\ \mathbf{m}_{21} & \mathbf{m}_{22} \end{pmatrix} \equiv \begin{pmatrix} \partial\mathbf{p}_t/\partial\mathbf{p}'^T & \partial\mathbf{p}_t/\partial\mathbf{q}'^T \\ \partial\mathbf{q}_t/\partial\mathbf{p}'^T & \partial\mathbf{q}_t/\partial\mathbf{q}'^T \end{pmatrix}. \tag{A3}$$

The equation of motion for $\mathbf{M}$ is obtained by linearizing Hamilton's equations for the deviations and reads

$$\frac{d}{dt}\mathbf{M} = \begin{pmatrix} -\partial^2 H/\partial\mathbf{q}_t\partial\mathbf{p}_t^T & -\partial^2 H/\partial\mathbf{q}_t\partial\mathbf{q}_t^T \\ \partial^2 H/\partial\mathbf{p}_t\partial\mathbf{p}_t^T & \partial^2 H/\partial\mathbf{p}_t\partial\mathbf{q}_t^T \end{pmatrix}\mathbf{M} = -\mathbf{JHM}, \tag{A4}$$

where the skew symmetric matrix,

$$\mathbf{J} = \begin{pmatrix} \mathbf{0} & \mathbf{I} \\ -\mathbf{I} & \mathbf{0} \end{pmatrix}, \tag{A5}$$

and the Hessian of the Hamiltonian $H(\mathbf{p}_t,\mathbf{q}_t)$,

$$\mathbf{H} = \begin{pmatrix} \partial^2 H/\partial\mathbf{p}_t\partial\mathbf{p}_t^T & \partial^2 H/\partial\mathbf{p}_t\partial\mathbf{q}_t^T \\ \partial^2 H/\partial\mathbf{q}_t\partial\mathbf{p}_t^T & \partial^2 H/\partial\mathbf{q}_t\partial\mathbf{q}_t^T \end{pmatrix}, \tag{A6}$$

have been used. The initial conditions are following from the definition of the stability matrix to be

$$\mathbf{M}(0) = \begin{pmatrix} \mathbf{m}_{11}(0) & \mathbf{m}_{12}(0) \\ \mathbf{m}_{21}(0) & \mathbf{m}_{22}(0) \end{pmatrix} = \begin{pmatrix} \mathbf{I} & \mathbf{0} \\ \mathbf{0} & \mathbf{I} \end{pmatrix}. \tag{A7}$$

Finally, with the help of the equation of motion (A4), and the properties $\mathbf{JJ}^T=\mathbf{I}$, $\mathbf{JJ}=-\mathbf{I}$, and $\mathbf{H}^T=\mathbf{H}$, one can show that $(d/dt)\mathbf{M}^T\mathbf{JM}=\mathbf{0}$. From the initial condition, we can conclude that $\mathbf{M}^T\mathbf{JM}=\mathbf{J}$ and thus the relations,

$$\mathbf{m}_{22}^T\mathbf{m}_{11} - \mathbf{m}_{12}^T\mathbf{m}_{21} = \mathbf{I}, \tag{A8}$$

$$\mathbf{m}_{11}^T\mathbf{m}_{21} - \mathbf{m}_{21}^T\mathbf{m}_{11} = \mathbf{0}, \tag{A9}$$

$$\mathbf{m}_{22}^T\mathbf{m}_{12} - \mathbf{m}_{12}^T\mathbf{m}_{22} = \mathbf{0}, \tag{A10}$$

hold for all times.

APPENDEX B: THE HK PROPAGATOR FOR *N* DOFs

The principle of stationary phase equivalence has been used by Kay in his seminal work in the early 1990s to rederive the Herman-Kluk propagator.[2] In this appendix we show explicitly, how it can be used to allow for a more general width parameter matrix (e.g., a matrix with different elements on the diagonal to allow for appropriate widths for the different DOFs) instead of a constant times the unit matrix. To this end the HK propagator is written as

$$K^{\mathrm{HK}}(\mathbf{x},t;\mathbf{x}',0)=\int\frac{d\mathbf{p}'d\mathbf{q}'}{(2\pi\hbar)^N}\left(\frac{\det\gamma}{\pi^N}\right)^{1/2}R(\mathbf{p}',\mathbf{q}',t)\exp\left\{\frac{i}{\hbar}S(\mathbf{p}',\mathbf{q}',t)\right\}\exp\left\{-\frac{1}{2}(\mathbf{x}-\mathbf{q}_t)^T\gamma(\mathbf{x}-\mathbf{q}_t)+\frac{i}{\hbar}\mathbf{p}_t^T\cdot(\mathbf{x}-\mathbf{q}_t)\right\}$$
$$\times\exp\left\{-\frac{1}{2}(\mathbf{x}-\mathbf{q}')^T\gamma(\mathbf{x}-\mathbf{q}')-\frac{i}{\hbar}\mathbf{p}'^T\cdot(\mathbf{x}-\mathbf{q}')\right\},\tag{B1}$$

with a yet undetermined prefactor R.

For the stationary phase evaluation of the phase space integral above, we use the definition of the stability matrix from the previous appendix and the classical mechanical identities $\partial S/\partial\mathbf{p}'=\mathbf{p}_t\mathbf{m}_{21}, \partial S/\partial\mathbf{q}'=\mathbf{p}_t\mathbf{m}_{22}-\mathbf{p}'$. This leads to stationary phase conditions which are fulfilled if $\mathbf{q}_t=\mathbf{x},\mathbf{q}'=\mathbf{x}'$, that is, if the classical trajectories fulfill the boundary problem given by the propagator labels in Eq. (B1). Generally, multiple solutions to this root search problem may exist.

The Gaussian stationary phase integration using the second derivatives of the total exponent at the stationary phase points, apart from the preexponential factors, leads to a sum of expressions of the form

$$\int d\mathbf{p}'d\mathbf{q}'\,\exp\{-(\delta\mathbf{p}',\delta\mathbf{q}')^T\mathbf{A}(\delta\mathbf{p}',\delta\mathbf{q}')\}=\left(\frac{\pi^{2N}}{\det\mathbf{A}}\right)^{1/2},\tag{B2}$$

where $(\delta\mathbf{p}',\delta\mathbf{q}')$ is the $2N$-element column vector of deviations of $\mathbf{p}',\mathbf{q}'$ from the stationary phase points and with the $2N\times 2N$ matrix

$$\mathbf{A}\equiv\begin{pmatrix}\mathbf{r}\mathbf{m}_{21}/2 & \mathbf{m}_{21}^T\mathbf{s}^T/2\\ \mathbf{s}\mathbf{m}_{21}/2 & (\gamma+\mathbf{s}\mathbf{m}_{22})/2\end{pmatrix},\tag{B3}$$

where the definitions,

$$\mathbf{r}\equiv\mathbf{m}_{21}^T\gamma+\frac{i}{\hbar}\mathbf{m}_{11}^T,\tag{B4}$$

$$\mathbf{s}\equiv\mathbf{m}_{22}^T\gamma+\frac{i}{\hbar}\mathbf{m}_{12}^T,\tag{B5}$$

have been used. To proceed, we use block matrix calculus (see, e.g., Sec. 2.7 of Ref. 39), which allows us to write the determinant of the symmetric block matrix in Eq. (B3) as

$$\det\begin{pmatrix}\mathbf{a}_{11} & \mathbf{a}_{12}\\ \mathbf{a}_{12}^T & \mathbf{a}_{22}\end{pmatrix}=\det\mathbf{a}_{11}\det(\mathbf{a}_{22}-\mathbf{a}_{12}^T\mathbf{a}_{11}^{-1}\mathbf{a}_{12}).\tag{B6}$$

Using the symplecticity properties of the previous appendix, which lead to the identity

$$\mathbf{m}_{21}^T\mathbf{s}^T=\mathbf{r}\mathbf{m}_{22}-\frac{i}{\hbar}\mathbf{I},\tag{B7}$$

and by applying the rule $\det(\mathbf{ab})=\det\mathbf{a}\det\mathbf{b}$ several times, we are finally led to the expression

$$\det\begin{pmatrix}\mathbf{r}\mathbf{m}_{21} & \mathbf{m}_{21}^T\mathbf{s}^T\\ \mathbf{s}\mathbf{m}_{21} & \gamma+\mathbf{s}\mathbf{m}_{22}\end{pmatrix}=\left(\frac{2i}{\hbar}\right)^N\det\mathbf{m}_{21}\det\left\{\frac{1}{2}\left(\gamma\mathbf{m}_{11}^T+\mathbf{m}_{22}^T\gamma-i\hbar\,\gamma\mathbf{m}_{21}^T\gamma-\frac{1}{i\hbar}\mathbf{m}_{12}^T\right)\right\}.\tag{B8}$$

Combining the result of the Gaussian stationary phase integration with the remaining prefactors, using $\det\mathbf{a}=\det\mathbf{a}^T$, and by engaging the equivalence of the resulting expression with the van Vleck–Gutzwiller propagator,[37,38] the prefactor,

$$R(\mathbf{p}',\mathbf{q}',t)=\left(\det\left\{\frac{1}{2}\left(\mathbf{m}_{11}+\gamma\mathbf{m}_{22}\gamma^{-1}-i\hbar\,\gamma\mathbf{m}_{21}-\frac{1}{i\hbar}\mathbf{m}_{12}\gamma^{-1}\right)\right\}\right)^{1/2}\equiv\sqrt{\det\mathbf{h}},\tag{B9}$$

of the HK propagator emerges.

APPENDIX C: FROM HK TO TGWD FOR *N* DOFs

In order to derive TGWD from the HK propagator as applied to an initial Gaussian wave packet, we start from Eq. (4) and insert Eq. (1). In the Gaussian expansion approximation we first expand the action up to second order around the initial center $(\mathbf{p}_\alpha,\mathbf{q}_\alpha)$ of the wave packet to be propagated. This leads to

$$S(\mathbf{p}',\mathbf{q}',t)=S(\mathbf{p}_\alpha,\mathbf{q}_\alpha,t)+\mathbf{p}_{\alpha,t}^T\mathbf{m}_{21}\delta\mathbf{p}'+(\mathbf{p}_{\alpha,t}^T\mathbf{m}_{22}-\mathbf{p}_\alpha^T)\cdot\delta\mathbf{q}'+\tfrac{1}{2}\delta\mathbf{p}'^T\mathbf{m}_{11}^T\mathbf{m}_{21}\delta\mathbf{p}'+\tfrac{1}{2}\delta\mathbf{q}'^T\mathbf{m}_{12}^T\mathbf{m}_{22}\delta\mathbf{q}'+\delta\mathbf{p}'^T\mathbf{m}_{21}^T\mathbf{m}_{12}\delta\mathbf{q}',\tag{C1}$$

where $\delta\mathbf{p}'=\mathbf{p}'-\mathbf{p}_\alpha, \delta\mathbf{q}'=\mathbf{q}'-\mathbf{q}_\alpha$ now are the deviations from the initial center of the Gaussian. Then first order expansions of the final momentum and position $(\mathbf{p}_t,\mathbf{q}_t)$ are made. To this end, we use Eqs. (A1) and (A2) together with the definitions $\mathbf{p}_{\alpha,t}\equiv\mathbf{p}(\mathbf{p}_\alpha,\mathbf{q}_\alpha,t)$ and $\mathbf{q}_{\alpha,t}\equiv\mathbf{q}(\mathbf{p}_\alpha,\mathbf{q}_\alpha,t)$ for the zero order terms. To complete the calculation of the terms in the exponent of Eq. (4), the integral over $\mathbf{x}'$ has to be performed. This overlap of two (complex) Gaussians can be done exactly analytically, however, leading to the formula already given in Eq. (5). The phase space integration can thus be performed by using the general Gaussian integration formula with a linear term in the exponent

$$\int d\mathbf{p}'d\mathbf{q}'\ \exp\{-(\delta\mathbf{p}',\delta\mathbf{q}')^T\mathbf{A}(\delta\mathbf{p}',\delta\mathbf{q}')+\mathbf{b}^T\cdot(\delta\mathbf{p}',\delta\mathbf{q}')+c\}$$
$$=\left(\frac{\pi^{2N}}{\det\mathbf{A}}\right)^{1/2}\exp\left\{\frac{1}{4}\mathbf{b}^T\mathbf{A}^{-1}\mathbf{b}+c\right\}, \tag{C2}$$

following from a standard "completion of the square" argument. Using Eqs. (4) and (5) and the definitions (B4) and (B5) of the previous appendix, we make the identifications

$$\mathbf{A}=\begin{pmatrix}\mathbf{a}_{11} & \mathbf{a}_{12}\\ \mathbf{a}_{12}^T & \mathbf{a}_{22}\end{pmatrix}$$
$$\equiv\begin{pmatrix}\gamma^{-1}/(4\hbar^2)+\mathbf{r}\mathbf{m}_{21}/2 & i/(4\hbar)+\mathbf{m}_{21}^T\mathbf{s}^T/2\\ i/(4\hbar)+\mathbf{s}\mathbf{m}_{21}/2 & \gamma/4+\mathbf{s}\mathbf{m}_{22}/2\end{pmatrix}, \tag{C3}$$

for the $2N\times 2N$ matrix $\mathbf{A}$, for which we use the same symbol although it is different from the matrix of which we have calculated the determinant in the previous appendix. For the $2N$-element row vector in the above expression, we find

$$\mathbf{b}^T\equiv(\mathbf{x}-\mathbf{q}_{\alpha,t})^T(\mathbf{r}^T,\mathbf{s}^T), \tag{C4}$$

and for the remaining scalar

$$c\equiv-\frac{1}{2}(\mathbf{x}-\mathbf{q}_{\alpha,t})^T\gamma(\mathbf{x}-\mathbf{q}_{\alpha,t})+\frac{i}{\hbar}\mathbf{p}_{\alpha,t}^T\cdot(\mathbf{x}-\mathbf{q}_{\alpha,t})$$
$$+\frac{i}{\hbar}S(\mathbf{p}_\alpha,\mathbf{q}_\alpha,t) \tag{C5}$$

follows.

To proceed, we need the inverse of a block matrix, which is given by[39]

$$\begin{pmatrix}\mathbf{a}_{11} & \mathbf{a}_{12}\\ \mathbf{a}_{12}^T & \mathbf{a}_{22}\end{pmatrix}^{-1}=\begin{pmatrix}\mathbf{d}^{-1} & -\mathbf{a}_{11}^{-1}\mathbf{a}_{12}\mathbf{e}^{-1}\\ -\mathbf{e}^{-1}\mathbf{a}_{12}^T\mathbf{a}_{11}^{-1} & \mathbf{e}^{-1}\end{pmatrix}, \tag{C6}$$

with the abbreviations $\mathbf{d}\equiv\mathbf{a}_{11}-\mathbf{a}_{12}\mathbf{a}_{22}^{-1}\mathbf{a}_{12}^T$ and $\mathbf{e}\equiv\mathbf{a}_{22}-\mathbf{a}_{12}^T\mathbf{a}_{11}^{-1}\mathbf{a}_{12}$. For the calculation of the exponent it is worthwhile to note that the intermediate results,

$$\mathbf{a}_{22}\mathbf{d}=(\mathbf{s}-i\hbar\mathbf{a}_{22}\mathbf{r}\mathbf{m}_{22}\mathbf{a}_{22}^{-1}\gamma\mathbf{m}_{22}^{-1})\left(\frac{1}{8\hbar^2}\mathbf{m}_{22}\gamma^{-1}+\frac{i}{8\hbar}\mathbf{m}_{21}\right), \tag{C7}$$

$$\mathbf{a}_{11}\mathbf{e}=(-i\hbar\mathbf{r}+\mathbf{a}_{11}\mathbf{s}\mathbf{m}_{21}\mathbf{a}_{11}^{-1}\gamma^{-1}\mathbf{m}_{21}^{-1})$$
$$\times\left(\frac{1}{8\hbar^2}\mathbf{m}_{22}+\frac{i}{8\hbar}\mathbf{m}_{21}\gamma\right), \tag{C8}$$

can be employed. It then requires some algebra to show that

$$\mathbf{b}^T\mathbf{A}^{-1}\mathbf{b}=2(\mathbf{x}-\mathbf{q}_{\alpha,t})^T(\mathbf{s}^T+i\hbar\mathbf{r}^T\gamma)$$
$$\times(\mathbf{m}_{22}+i\hbar\mathbf{m}_{21}\gamma)^{-1}(\mathbf{x}-\mathbf{q}_{\alpha,t}) \tag{C9}$$

holds. Combining all the terms in the exponent and after calculating the determinant,

$$\det\mathbf{A}=(2\hbar)^{-2N}\det\mathbf{h}\ \det(\mathbf{m}_{22}+i\hbar\mathbf{m}_{21}\gamma), \tag{C10}$$

and combining it with the prefactor of Eq. (4) taken at $(\mathbf{p}_\alpha,\mathbf{q}_\alpha)$, the final expression for the wave function already given by Heller,[18]

$$\Psi_\alpha(\mathbf{x},t)=\left(\frac{\det\gamma}{\pi^N}\right)^{1/4}\det(\mathbf{m}_{22}+i\hbar\mathbf{m}_{21}\gamma)^{-1/2}$$
$$\times\exp\left\{-\frac{1}{2}(\mathbf{x}-\mathbf{q}_{\alpha,t})^T\left(\mathbf{m}_{11}\gamma+\frac{1}{i\hbar}\mathbf{m}_{12}\right)\right.$$
$$\times(\mathbf{m}_{22}+i\hbar\mathbf{m}_{21}\gamma)^{-1}(\mathbf{x}-\mathbf{q}_{\alpha,t})$$
$$\left.+\frac{i}{\hbar}\mathbf{p}_{\alpha,t}^T\cdot(\mathbf{x}-\mathbf{q}_{\alpha,t})+\frac{i}{\hbar}S(\mathbf{p}_\alpha,\mathbf{q}_\alpha,t)\right\}, \tag{C11}$$

emerges.

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