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Rigorous forward-backward semiclassical formulation of many-body dynamics

Keiran Thompson and Nancy Makri
School of Chemical Sciences, University of Illinois, 601 South Goodwin Avenue, Urbana, Illinois 61801
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A rigorous, yet practical semiclassical formulation of time correlation functions or expectation values is presented. The main idea is to combine the forward and backward propagation steps into a single semiclassical propagator for those degrees of freedom that are not probed in a calculation, while retaining an explicit two-propagator description of the observable low-dimensional system. The combined forward-backward treatment of the environment naturally leads to extensive cancellation and results in an action that is small on the scale of Planck's constant. As a consequence, the integrand is smooth and thus amenable to Monte Carlo sampling. At the same time, important nonclassical effects terms, arising from interference among multiple bounce solutions of the system component, are fully accounted for. [S1063-651X(99)50905-7]

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Since the early days of quantum mechanics, the development of a theory that merges the gap between the wave equation and Newton's laws has attracted considerable attention. The Wentzel-Kramers-Brillouin(-Jeffreys) [WKB(J)] method and its time-dependent analog, the semiclassical propagator, offer a sufficiently satisfactory connection in the small $\hbar$ regime. In spite of well-known problems with the semiclassical wave function near caustics, the WKB(J) energy levels and the Van Vleck propagator [1,2] are exact for several model problems (see, for example, [3]). Furthermore, the semiclassical propagator has been shown to be highly accurate in nontrivial situations [4], providing a very desirable alternative to full quantum dynamics for many-body problems. However, numerical implementation of time-dependent semiclassical theory has in the past been problematic. One of the major drawbacks of the Van Vleck expression is that the relevant trajectories must satisfy double-ended boundary value conditions. Alternative formulations have been developed which use initial value representations [5], coherent states [6], or cellular dynamics [7] to avoid computationally intensive root searches. Another severe problem is the highly oscillatory structure of the semiclassical propagator, which obviates the use of importance sampling procedures and thus limits applications to low-dimensional problems. Until recently, calculations involving more than two degrees of freedom have been possible only via the use of filtering methods [8], mixed order schemes [9], or linearized approximations [10].

Recently, we introduced a rigorous semiclassical methodology for calculating influence functionals arising from many-body environments in the context of a path integral description of the system of interest [11,12]. Rather than introducing approximations to ameliorate the sign problem, our approach is based on the observation that the forward and backward time evolution operators of unprobed degrees of freedom can be combined into a single semiclassical propagation. While the forward step gives rise to an action integral that can be large, subsequent propagation in the backward time direction generally results in a net action that is small. As a consequence, the integrand is only mildly oscillatory and thus amenable to Monte Carlo sampling.

The present Rapid Communication develops a fully semiclassical forward-backward formulation of time correlation functions or expectation values. This can be thought of as arising from the stationary phase limit of the path integral expression for the observed system of interest, while retaining a forward-backward description for the environment. The resulting expression is evaluated in a coherent state representation by a combination of Monte Carlo and quadrature techniques. Numerical calculations illustrate the advantages and feasibility of the present formulation.

For concreteness we focus on correlation functions of the type

$$C(t)=\mathrm{Tr}(\rho(0)Ae^{iHt/\hbar}Be^{-iHt/\hbar}), \tag{1}$$

where $\rho(0)$ is the density operator of the initial ensemble and A and B are operators that depend only on the position r of the observable system whose conjugate momentum is denoted as p. Setting $A=B=r$, Eq. (1) becomes a position correlation function, while the choice $A=1$, $B=r$ yields the expectation value of the position operator. For simplicity of notation we treat the system as one-dimensional throughout this Rapid Communication. A generalization of the obtained expressions to the case where r is a collection of a small number of coordinates (typically, a three-dimensional vector or a component thereof) is straightforward. The remaining n degrees of freedom that are not probed in the calculation (the ''solvent'') are denoted collectively by the canonically conjugate variables represented by the n-dimensional vectors $\mathbf{R}$ and $\mathbf{P}$. Finally, we note that the present formulation can easily be generalized to describe other correlation functions or reduced density matrices.

Equation (1) is written in the following coordinate representation:

$$\begin{aligned}C(t)=&\int dr_0\int dr_t\int dr_f\int d\mathbf{R}_0\int d\mathbf{R}_t\int d\mathbf{R}_f\\&\times\langle r_0\mathbf{R}_0|\rho(0)|r_f\mathbf{R}_f\rangle A(r_f)\langle r_f\mathbf{R}_f|e^{iHt/\hbar}|r_t\mathbf{R}_t\rangle\\&\times\langle \mathbf{r}_t\mathbf{R}_t|e^{-iHt/\hbar}|r_0\mathbf{R}_0\rangle B(r_t).\end{aligned} \tag{2}$$

Using the semiclassical expression to approximate both propagators in the last equation leads to the form

$$C(t)=\int dr_0\int dr_t\int dr_f\int d\mathbf{R}_0\int d\mathbf{R}_t\int d\mathbf{R}_f\langle r_0\mathbf{R}_0|\rho(0)|r_f\mathbf{R}_f\rangle A(r_f)B(r_t)$$

$$\times\sum_{\substack{\text{forward}\\ \text{paths } r_{\rm cl}^+\mathbf{R}_{\rm cl}^+}}(2\pi i\hbar)^{-(n+1)/2}(\det\mathbf{M}_1)^{-1/2}\exp\left(\frac{i}{\hbar}S_{\rm for}[r_{\rm cl}^+,\mathbf{R}_{\rm cl}^+]\right)$$

$$\times\sum_{\substack{\text{backward}\\ \text{paths } r_{\rm cl}^-\mathbf{R}_{\rm cl}^-}}(2\pi i\hbar)^{-(n+1)/2}(\det\mathbf{M}_2)^{-1/2}\exp\left(\frac{i}{\hbar}S_{\rm back}[r_{\rm cl}^-,\mathbf{R}_{\rm cl}^-]\right). \tag{3}$$

Here $r_{\rm cl}^+(t')\mathbf{R}_{\rm cl}^+(t')$ and $r_{\rm cl}^-(t')\mathbf{R}_{\rm cl}^-(t')$ are paths in the forward and backward time direction with phase space end points $(r_0p_0\mathbf{R}_0\mathbf{P}_0,r_tp_t'\mathbf{R}_t\mathbf{P}_t')$ and $(r_tp_t\mathbf{R}_t\mathbf{P}_t,r_fp_f\mathbf{R}_f\mathbf{P}_f)$, respectively, while $S_{\rm for}$ and $S_{\rm back}$ are the corresponding action functionals. The matrices entering the Van Vleck determinants are given by the expressions

$$\mathbf{M}_1=\begin{pmatrix}\dfrac{\partial r_t}{\partial p_0} & \dfrac{\partial r_t}{\partial\mathbf{P}_0}\\ \dfrac{\partial\mathbf{R}_t}{\partial p_0} & \dfrac{\partial\mathbf{R}_t}{\partial\mathbf{P}_0}\end{pmatrix}\quad\text{and}\quad\mathbf{M}_2=\begin{pmatrix}\dfrac{\partial r_f}{\partial p_t} & \dfrac{\partial r_f}{\partial\mathbf{P}_t}\\ \dfrac{\partial\mathbf{R}_f}{\partial p_t} & \dfrac{\partial\mathbf{R}_f}{\partial\mathbf{P}_t}\end{pmatrix}. \tag{4}$$

Finally, the Maslov phase is absorbed in the square roots in order to simplify the notation. The highly oscillatory character of the integrand in Eq. (3) prevents its evaluation by Monte Carlo methods.

We now proceed to perform the integral over the intermediate solvent point $\mathbf{R}_t$ using the stationary phase approximation. By virtue of the relations

$$\frac{\partial S_{\rm for}}{\partial\mathbf{R}_t}=\mathbf{P}_t'\quad\text{and}\quad\frac{\partial S_{\rm back}}{\partial\mathbf{R}_t}=-\mathbf{P}_t, \tag{5}$$

the stationary phase condition implies $\mathbf{P}_t'-\mathbf{P}_t=0$, i.e., the solvent component of the trajectory is continuous. This feature, which is the key advantage of the present approach, arises because (by definition) the solvent degrees of freedom are not being interrogated. Note that some crossing terms, arising from multiple bounce solutions to the forward and backward propagation problems, are wiped out. This is deemed a reasonable approximation for the solvent degrees of freedom, and is fully consistent with the spirit of the semiclassical approximation to the (forward time) propagator.

To complete the stationary phase calculation we expand the action locally through quadratic terms about the stationary phase point of the solvent trajectory, keeping the system component r_t of the midpoint fixed. Integration over $\mathbf{R}_t$ introduces the factor

$$(2\pi i\hbar)^{n/2}\left(\det\frac{\partial^2S_{\text{for-back}}}{\partial\mathbf{R}_t\partial\mathbf{R}_t}\right)^{-1/2}, \tag{6}$$

where $S_{\text{for-back}}$ is the sum of forward and backward actions. Noting that this procedure maps the local dynamics of the solvent onto that of a time-dependent harmonic oscillator, one finds that the stationary phase factor is equal to

$$(2\pi i\hbar)^{n/2}\left(\det\frac{\partial^2S_{\rm for}}{\partial\mathbf{R}_0\partial\mathbf{R}_t}\right)^{1/2}\left(\det\frac{\partial^2S_{\rm back}}{\partial\mathbf{R}_t\partial\mathbf{R}_f}\right)^{1/2}$$
$$\times\left(\det\frac{\partial^2S_{\text{for-back}}}{\partial\mathbf{R}_0\partial\mathbf{R}_f}\right)^{-1/2}. \tag{7}$$

This result also arises directly from the group property of the propagator within the stationary phase approximation. It follows that the result of the stationary phase evaluation of the integral in Eq. (3) is

$$C(t)=\int dr_0\int dr_t\int dr_f\int d\mathbf{R}_0\int d\mathbf{R}_f A(r_f)B(r_t)\langle r_0\mathbf{R}_0|\rho(0)|r_f\mathbf{R}_f\rangle$$

$$\times\sum_{\substack{\text{forward system}\\ \text{paths } r_{\rm cl}^+}}\sum_{\substack{\text{backward system}\\ \text{paths } r_{\rm cl}^-}}\sum_{\substack{\text{forward-backward}\\ \text{solvent paths } \mathbf{R}_{\rm cl}}}D_{\rm for}^{\rm VV}D_{\rm back}^{\rm VV}D_{\text{for-back}}^{\rm VV}\exp\left(\frac{i}{\hbar}S_{\text{for-back}}[r_{\rm cl}^+,r_{\rm cl}^-,\mathbf{R}_{\rm cl}]\right), \tag{8}$$

where the prefactors are given by the expressions

$$D_{\rm for}^{\rm VV}=(2\pi i\hbar)^{-1/2}(\det \mathbf{M}_1)^{-1/2}\left(\det\frac{\partial \mathbf{R}_t}{\partial \mathbf{P}_0}\right)^{1/2},$$

$$D_{\rm back}^{\rm VV}=(2\pi i\hbar)^{-1/2}(\det \mathbf{M}_2)^{-1/2}\left(\det\frac{\partial \mathbf{R}_f}{\partial \mathbf{P}_t}\right)^{1/2}, \tag{9a}$$

$$D_{\rm for\text{-}back}^{\rm VV}=(2\pi i\hbar)^{-n/2}\left(\det\frac{\partial \mathbf{R}_f}{\partial \mathbf{P}_0}\right)^{-1/2}. \tag{9b}$$

Equation (8) constitutes the forward-backward semiclassical dynamics (FBSD) formulation of time correlation functions in the coordinate representation.

Apart from reducing the number of integrals for each solvent degree of freedom from three in Eq. (3) to two, the main advantage of the FBSD formulation given by Eq. (8) is that the overall action is not very large on the scale of Planck's constant and, therefore, the integrand is no longer highly oscillatory. To see this, consider evaluating the integral over r_t by the stationary phase method. The stationary phase point occurs when the final momentum p_t' of $r_{\rm cl}^{+}(t')$ is equal to the initial momentum p_t of the backward trajectory $r_{\rm cl}^{-}(t')$, in other words, when these paths combine to a single continuous forward-backward classical trajectory in the space of all variables. Note that in the present boundary condition representation such a stationary phase point exists only if the end points match exactly, i.e., if $r_f=r_0$ and $\mathbf{R}_f=\mathbf{R}_0$. However, coincidence of the forward and backward trajectories implies that the total action vanishes and the complex exponential in the integrand of Eq. (8) becomes equal to unity. On the other hand, if the above end point condition is not satisfied, the absence of stationary phase regions leads to the vanishing of the integral and thus the domains of the highly oscillatory integrand need not be sampled. (This very sharp condition on the most important regions of the integrand will be broadened in the coherent state representation to be adopted below.) Finally, note that significant quantum effects may arise from interference among classical trajectories of the observable system that do not satisfy the above stationary phase condition. For this reason we prefer to evaluate the midpoint integral in Eq. (8) by numerical integration rather than the stationary phase method. Pairs of different forward-backward system paths will give rise to nonzero action integrals; however, the fact that the solvent component of each included trajectory is still continuous leads to extensive cancellation, resulting in actions that are much smaller than these in the integrand of Eq. (3). This fact was shown to have a dramatic effect of smoothing the integrand of the forward-backward influence functional in our recent work [11,12].

Still, Eq. (8) is not in a convenient form for numerical calculations because the trajectories are specified by double-ended boundary value conditions. This problem is overcome in initial value representations. Specifically, Eq. (8) can be cast in the following coherent state representation [6]:

$$C(t)=(2\pi\hbar)^{-(n+2)}\int dr_1\int dp_1\int dr_2\int dp_2\int d\mathbf{R}_0\int d\mathbf{P}_0 D^{\rm coh}\exp\left(\frac{i}{\hbar}S_{\rm for\text{-}back}[r_{\rm cl}^{+},r_{\rm cl}^{-},\mathbf{R}_{\rm cl}]\right)$$
$$\times\langle g(r_1,p_1)G(\mathbf{R}_0,\mathbf{P}_0)|\rho(0)A|G(\mathbf{R}_f,\mathbf{P}_f)g(r_{2f},p_{2f})\rangle\langle g(r_2,p_2)|B|g(r_{1f},p_{1f})\rangle. \tag{10}$$

Here g and G are coherent states described by the wave functions

$$\langle r|g(r_0,p_0)\rangle=\left(\frac{2\gamma}{\pi}\right)^{1/4}\exp\left(-\gamma(r-r_0)^2+\frac{i}{\hbar}p_0(r-r_0)\right), \tag{11a}$$

$$\langle \mathbf{R}|G(\mathbf{R}_0,\mathbf{P}_0)\rangle=\left(\frac{2}{\pi}\right)^{n/4}(\det\Gamma)^{1/4}\exp\left(-(\mathbf{R}-\mathbf{R}_0)\cdot\Gamma\cdot(\mathbf{R}-\mathbf{R}_0)+\frac{i}{\hbar}\mathbf{P}_0\cdot(\mathbf{R}-\mathbf{R}_0)\right), \tag{11b}$$

where Γ is a diagonal matrix and $D^{\rm coh}$ is an appropriate prefactor. Note that the stationary phase condition is modified in the coherent state representation where the trajectories are specified by initial rather than boundary conditions.

Equation (10) gives the correlation function in terms of two phase space integrals for the system of interest and one phase space integral for each solvent degree of freedom. These phase space variables specify the initial conditions of the required classical trajectories. Starting at $(r_1p_1\mathbf{R}_0\mathbf{P}_0)$, a classical trajectory is launched in the forward time direction, which reaches the point $(r_{1f}p_{1f}\mathbf{R}_t\mathbf{P}_t)$ at the time t. Subsequently, the system component of the position and momentum are changed to the values (r_2p_2), while the solvent variables remain at the values $(\mathbf{R}_t\mathbf{P}_t)$ and the trajectory is continued in the backward time direction, reaching the phase space point $(r_{2f}p_{2f}\mathbf{R}_f\mathbf{P}_f)$ as the time returns to zero. The coherent state matrix element of the operator B is evaluated analytically, while the density matrix element is obtained using a high-temperature approximation, an imaginary time path integral description, or a classical path representation, as appropriate [12]. As was alluded to earlier, the propagation of trajectories along the combined forward-backward contour leads to extensive cancellation, resulting in an overall action that is generally small on the scale of Planck's constant.

To illustrate the practicality of the FBSD scheme we present below the results of propagating a Gaussian wave packet in the following Hamiltonian of three coupled anharmonic oscillators:

$$H = \tfrac{1}{2}(p^2 + P_1^2 + P_2^2) + \tfrac{1}{2}(\omega_0^2 r^2 + \omega_1^2 R_1^2 + \omega_2^2 R_2^2) - 0.1r^3 + 0.01r^4 - c_1 r R_1 - c_2 r R_2, \quad (12)$$

with $\omega_0 = \sqrt{2}$, $\omega_1 = \omega_2 = 1$, and $c_1 = c_2 = 0.1$. The initial wave packet has a width that is equal to that of the ground state of the harmonic fit about the minimum of this potential, and is displaced by the amounts $r^0 = 1$ and $R_1^0 = R_2^0 = 2$. The results presented in Fig. 1, obtained with 20 000 Monte Carlo points per integration variable, demonstrate that the FBSD scheme leads to rapid convergence over several periods of oscillation. In addition, the semiclassical results are in excellent agreement with those obtained from full quantum mechanical calculations using a split propagator method [13]. This behavior is very encouraging.

In summary, the FBSD formulation of time correlation functions presented in this Rapid Communication exploits the structure of ensemble averaged time correlation functions or expectation values to naturally eliminate the rapid oscillations of the integrand. This is achieved by observing that the time evolution operators of those degrees of freedom that are not probed directly can be combined in a single operator, which is evaluated semiclassically via trajectories that evolve along a single forward-backward time contour. The backward propagation step results in a net action that is generally small and thus the oscillations of the integrand are dramatically diminished. At the same time, the midpoint integral for the system of interest is not performed via the stationary phase method, allowing explicit treatment of the interference between the forward and backward propagators arising from multiple bounce trajectories. For these reasons, the FBSD scheme appears promising as a robust, yet rigorous, numerical tool for studying the real time dynamics of many-body systems.

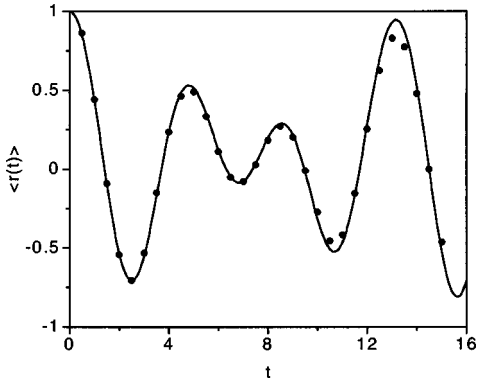

FIG. 1. Expectation value of the system position for the three-dimensional Hamiltonian given in Eq. (12). Markers: results of the FBSD scheme with 20 000 Monte Carlo samples per integration variable. The error bars are approximately equal to the size of the markers. Solid line: exact quantum results generated via the split propagator method.

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