Amplitude-free correlation function based on an algebra for coordinate transformation in semiclassical integrals

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We present an algebra that facilitates a systematic coordinate transformation in semiclassical integrals such as those between the initial and final value representations. Applying this algebra to Maslov-type semiclassical wave packet theory [A. Inoue-Ushiyama and K. Takatsuka, Phys. Rev. A **59**, 3256 (1999)], a semiclassical correlation function is extracted, which is free of the amplitude factor that suffers an exponential divergence in a chaotic system.

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I. INTRODUCTION

Semiclassical mechanics [1–5] has been regarded for a long time as a promising alternative to full quantum mechanics for systems composed of heavy particles, such as, e.g., molecular vibration and chemical reaction systems. Nevertheless, it is still extremely difficult to quantize vibrational states in a chaotic system, due to divergence in the amplitude factor (prefactor) of the semiclassical wave functions. This rather short paper is concerned with a systematic method of handling coordinate transformations in semiclassical theory, and as its consequence we find an approximate form of the quantum correlation function, which is particularly useful in a calculation of the spectra of chaotic systems. We first show a theoretical scheme to construct a semiclassical correlation function, which is free of the amplitude factor. In a companion paper [6] we will present numerical results for a quantization of a strongly chaotic cluster which is composed of seven identical atoms as an application. The latter paper mainly treats more numerical aspects in practical applications, including the permutation symmetry of identical particles, and also shows a spectrum arising from a straightforward application of a semiclassical theory based on an action decomposed function [7]; thereby it exhibits the numerical difficulty one encounters in quantizing chaos. It also presents a spectrum improved by careful use of both the initial and final value presentations, along with a spectrum based on an amplitude-free correlation function.

We begin with the current progress [8–14] made in the semiclassical Feynman kernel [15] K, into which a coordinate transformation is introduced which is relevant to the present paper. Suppose a transition amplitude in a propagated system is written in a coordinate representation as

$$\langle\Phi|\phi(t)\rangle=\left\langle \Phi\left|\exp\left(-\frac{i}{\hbar}Ht\right)\right|\phi\right\rangle = \int\int \Phi^*(q_t)K(q_t,q_0,t)\phi(q_0)dq_t dq_0. \quad (1)$$

This representation demands quite a tedious approach, because it requires a double-ended root search problem. It was Miller and co-workers [8,9] who introduced a coordinate transformation in such a way that

$$\langle\Phi|\phi(t)\rangle=\int\int \Phi^*(q_t)K(q_t,q_0,t)\phi(q_0)\left|\frac{\partial q_t}{\partial p_0}\right|dp_0dq_0, \quad (2)$$

in which the classical paths representing the semiclassical kernel are specified in terms of their initial conditions (q_0,p_0). To be more specific, the standard form of a semiclassical kernel is usually written as

$$K_{sc}(q,q_0,t)=(2\pi i\hbar)^{-N/2}\int \delta(q-q_t)\left|\frac{\partial q_t}{\partial p_0}\right|^{-1/2} \times\exp\left(\frac{i}{\hbar}S_1(q_t,q_0,t)-i\pi\frac{\lambda}{2}\right)dq_t, \quad (3)$$

where S_1 and λ are the classical action integral and the Maslov index, respectively. We tentatively call this form a final value representation (FVR). The toughest problem inherent in the FVR is that the amplitude factor $|\partial q_t/\partial p_0|^{-1/2}$ diverges at every caustic point. On the other hand, in the transformed expression called the initial value representation (IVR),

$$K_{sc}(q,q_0,t)=(2\pi i\hbar)^{-N/2}\int \delta(q-q_t)\left|\frac{\partial q_t}{\partial p_0}\right|^{1/2} \times\exp\left(\frac{i}{\hbar}S_1(q_t,q_0,t)-i\pi\frac{\lambda}{2}\right)dp_0, \quad (4)$$

the diverging factor at the caustic has been inverted to $|\partial q_t/\partial p_0|^{1/2}$, which simply becomes zero. This excellent property was rediscovered in the early 1990s by other authors in various variants of the representation [10–14], leading to the current situation in which semiclassical theories are widely applied to molecular dynamical studies. As a price for this divergence-free nature, the absolute value of the new amplitude factor $|\partial q_t/\partial p_0|^{1/2}$ grows almost exponentially with time in the case of classical chaos, which

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causes another difficulty in the calculation of quantum spectra. This is the central problem we consider in this paper and companion papers.

The aim of the present paper is twofold: First we formulate a simple and systematic algebra of the relevant coordinate transformation by introducing the square root of the volume element as $dq_0 = dq_0^{1/2} dq_0^{1/2*}$. Then we present an approximate semiclassical correlation function, based on our action decomposed function (the Maslov type semiclassical wave function) [7], which is free from the diverging amplitude factor $|\partial q_t/\partial p_0|^{1/2}$. Since the presence of this factor makes a calculation of energy spectra in a chaotic system prohibitively difficult, our correlation function will provide a useful alternative to study quantum spectra in chaos [16,17].

The idea of the amplitude-free correlation function was first launched by Miller [2]. Recently, Shao and Makri developed a theory that led to a correlation function free of the amplitude factor (prefactor) [18]. Our study was developed in an independent context, and in fact our resultant correlation function [Eq. (52)] is different from that of Ref. [18] in that ours should be applied under a limited condition.

This paper is organized as follows. Section II reviews selected semiclassical theories, which are particularly relevant to the coordinate transformation. We then propose a practical method of handling the coordinate transformation in semiclassical theory. Various representations of the correlation function are shown in Sec. IV, by which we derive a correlation function free of the amplitude factor.

II. SEMICLASSICAL THEORIES

Here we make a brief review of two different kinds of semiclassical theory, which are particularly relevant to the main issue of this article. One is a phase-space path integral, which is typically invariant with respect to the coordinate transformation. The other is a wave packet type theory, with which we propose a representation of the correlation function that is free from the annoying amplitude factor.

A. Phase-space path integral

Although the above transformation from Eqs. (3) and (4) should keep the kernel and relevant semiclassical integrals such as the correlation function mathematically invariant, they change the values in practice. As an illustrative example, let us consider the behavior of the integrands at a caustic point. In the FVR the $|\partial q_t/\partial p_0|^{-1/2}$ diverges, but $|\partial q_t/\partial p_0|^{1/2}$ becomes zero in the IVR. The divergence reflects the physical fact that the density of the trajectories happens to be enormous at caustics (it is still finite in full quantum mechanics, however). It is therefore implied that very many trajectories are required to compensate for the ''zero'' brought about by the IVR. However, a transformation problem does not necessarily arise in all semiclassical theories. A semiclassical theory based on the phase-space path integral [19] is such an example. The transition amplitude in Eq. (1) is written as

$$\left\langle \Phi \left| \exp\left(-\frac{i}{\hbar}Ht\right) \right| \phi \right\rangle = (2\pi\hbar)^{-N} \times \exp\left(-i\frac{N\pi}{2}\right) \int dZ_i a(\phi Z_i;\Phi Z_f) \times \left(\frac{\partial(Z_f - Z_i)}{\partial Z_i}\right)^{1/2} \exp\left(\frac{i}{\hbar}S(Z_f,Z_i,t)\right) \tag{5}$$

in terms of what we call the dynamical characteristic function [19]

$$a(\phi Z_i;\Phi Z_f) = a(\phi q_i p_i;\Phi q_f p_f) = \int dx\, \phi(x+q_i)\Phi^*(x+q_f) \times \exp\left(\frac{i}{\hbar}x(p_f - p_i)\right), \tag{6}$$

where $Z_i = (q_i, p_i)$ and $Z_f = (q_f, p_f)$ specify the initial and final points in phase space, respectively. The Jacobian matrix, whose determinant appears in Eq. (5), is

$$\left[\frac{\partial(Z_f - Z_i)}{\partial Z_i}\right] = \left[\frac{\partial Z_f}{\partial Z_i}\right] - I, \tag{7}$$

where I is a $2N\times 2N$ unit matrix, and $[\partial Z_f/\partial Z_i]$ is the well-known stability matrix:

$$\left[\frac{\partial Z_f}{\partial Z_i}\right] = \left[\frac{\partial(q_f,p_f)}{\partial(q_i,p_i)}\right]. \tag{8}$$

The Liouville theorem ensures that the determinant $\partial Z_f/\partial Z_i$ is kept to unity, resulting in $dZ_i = dZ_f$. For Eq. (7), we have

$$\frac{\partial(Z_f - Z_i)}{\partial Z_i} = \prod_{k=1}^{N} [1-\exp(ib_k)][1-\exp(-ib_k)] = \prod_{k=1}^{N} \left[2\sin\left(\frac{b_k}{2}\right)\right]^2, \tag{9}$$

where $\exp(ib_k)$ are the eigenvalues of the matrix $[\partial Z_f/\partial Z_i]$, with b_k a complex number in general.

We note that Eq. (5) is represented in terms of the initial value representation from the outset [compare with Eq. (1)]. Furthermore, the coordinate transformation is straightforward as

$$\left\langle \Phi \left| \exp\left(-\frac{i}{\hbar} Ht \right) \right| \phi \right\rangle$$
$$= \left\langle \phi \left| \exp\left(\frac{i}{\hbar} Ht \right) \right| \Phi \right\rangle^*$$
$$= (2\pi\hbar)^{-N} \exp\left(-i\frac{N\pi}{2} \right) \int dZ_f a^*(\Phi Z_f; \phi Z_i)$$
$$\times \left[\frac{\partial(Z_i - Z_f)}{\partial Z_f} \right]^{1/2} \exp\left(-\frac{i}{\hbar} S(Z_i, Z_f, -t) \right), \quad (10)$$

which is readily proved with the help of the following two facts:

$$a(\phi Z_i; \Phi Z_f)^* = a(\Phi Z_f; \phi Z_i) \quad (11)$$

and

$$\frac{\partial(Z_f - Z_i)}{\partial Z_i} = \frac{\partial Z_f}{\partial Z_i} \frac{\partial(Z_f - Z_i)}{\partial Z_f} = \frac{\partial(Z_i - Z_f)}{\partial Z_f}. \quad (12)$$

This transparent property is a clear advantage of phase-space quantum theory.

B. Maslov-type semiclassical wave function

The Maslov-type semiclassical theory [20] begins with the wave function

$$\Psi(q,t) = F(q,t) \exp\left[\frac{i}{\hbar} S_{cl} \right], \quad (13)$$

which is to be propagated in terms of the equation of motion of the lowest order approximation in $\hbar$ to the Schrödinger equation. The higher order effects are taken into account in different ways by the Bohm and Maslov theories: the so-called quantum potential is considered in the Bohm theory, while the Maslov theory takes account of the hierarchical series of quantum transport [20]. S_{cl} is the classical action that satisfies the Hamilton-Jacobi equation. Note that picking up S_{cl} among the various possible forms [21] is equivalent to specifying an initial condition that is imposed on the trajectories generated by S_{cl}. For later convenience, we choose S_{cl} to be an F_2-type generating function [21], such that $S_{cl}(q,p_0,t) = F_2(q,p_0,t) = F_1(q,q_0,t) + q_0 p_0$. [Neither F_2 nor F_1 should be confused with $F(q,t)$ of Eq. (13).] The function is determined by the equation of motion

$$\frac{\partial F}{\partial t} + v \cdot \nabla F = -\frac{1}{2}(\nabla \cdot v) F, \quad (14)$$

where $v = \partial S_2 / \partial q$ is the classical velocity. We use mass-weighted coordinates throughout, so that all the masses are scaled to unity. Equation (14) is integrated as follows. We start from the following observation:

$$\frac{\partial F^2}{\partial t} + \nabla \cdot (v F^2) = 0. \quad (15)$$

Note that F^2 rather than $|F|^2$ is considered in this ''equation of continuity'' (note that F^2 can be complex), although both satisfy Eq. (15). F^2 can be readily integrated locally along classical paths in terms of a Jacobian determinant $\partial q_t / \partial q_0$, which is a minor determinant of the so-called stability matrix [Eq. (8)]. $(\partial q_t / \partial q_0)^{-1} = \partial^2 S_2(q_t, p_0, t) / \partial q_t \partial p_0$ is interpreted as the density of families of classical trajectories which are labeled by p_0. It is not difficult to derive, from the Hamilton-Jacobi equation for $S_{cl}(q_t, p_0, t)$, that

$$\frac{\partial}{\partial t} \left(\frac{\partial q_t}{\partial q_0} \right)^{-1} + \nabla \cdot \left[v \left(\frac{\partial q_t}{\partial q_0} \right)^{-1} \right] = 0. \quad (16)$$

Furthermore, one has the initial condition $(\partial q_t / \partial q_0)^{-1} = 1$, since $q_t = q_0$ at $t = 0$. Thus $(\partial q_t / \partial q_0)^{-1}$ can be regarded as a local representation of the Green function for the propagator of Eq. (15). On comparing Eqs. (15) and (16), together with the initial conditions above, one immediately has [7]

$$F(q_t, t) = F(q_0, 0) \left(\frac{\partial q_t}{\partial q_0} \right)^{-1/2}$$
$$= F(q_0, 0) \left| \frac{\partial q_t}{\partial q_0} \right|^{-1/2} \exp\left[\mp \frac{i\pi M(q_0 \to q_t)}{2} \right], \quad (17)$$

where the derivative $\partial q_t / \partial q_0$ is taken under the fixed initial momentum p_0, and $M(q_0 \to q_t)$ is the Maslov index in this representation that counts the number of zeros of $\partial q_t / \partial q_0$ up to the degeneracy. Thus the local solution, denoted by $\Psi^{p_0}_{local}(q_t, t)$, has been obtained, which is to be propagated along a trajectory of a fixed initial momentum p_0. The final expression for the wave function is then written as

$$\Psi_{p_0}(q,t) = \int \delta(q - q_t) \Psi^{p_0}_{local}(q_t, t) dq_t$$
$$= \int \delta(q - q_t) \Psi^{p_0}_{local}(q_t, t) \left| \frac{\partial q_t}{\partial q_0} \right| dq_0$$
$$= \int \delta(q - q_t(q_0, p_0)) F(q_0, 0) \left| \frac{\partial q_t}{\partial q_0} \right|^{1/2}$$
$$\times \exp\left[\frac{i}{\hbar} S_2(q_t, p_0, t) \mp \frac{i\pi M(q_0 \to q_t)}{2} \right] dq_0. \quad (18)$$

A transformation from the final value to the initial value representation has been adopted. We note that there still remains the problem of which sign in Eqs. (17) and (18) should be taken from the possible choices $\mp$.

Equation (17) implies norm conservation, namely,

$$|F(q_t, t)|^2 = |F(q_0, 0)|^2 \left| \frac{\partial q_t}{\partial q_0} \right|^{-1}, \quad (19)$$

or, more algebraically,

$$|F(q_t, t)|^2 dq_t = |F(q_0, 0)|^2 dq_0. \quad (20)$$

However, the square of Eq. (17) simply brings about a different relation

$$F(q_t,t)^2 dq_t = F(q_0,0)^2 \exp[\mp i\pi M(q_0 \rightarrow q_t)] dq_0. \tag{21}$$

Therefore, we need an algebra to describe both of these in a consistent manner, which is to be applied to a correlation function,

$$\begin{aligned} C(t) &= \langle \Psi_{p_0}(0) | \Psi_{p_0}(t) \rangle \\ &= \int F^*(q_t,0) F(q_0,0) \left| \frac{\partial q_t}{\partial q_0} \right|^{1/2} \exp\left[-\frac{i}{\hbar} p_0 q_t \right. \\ &\quad \left. - \frac{i}{\hbar} S_2(q_t,p_0,t) \mp \frac{i\pi M(q_0 \rightarrow q_t)}{2} \right] dq_0, \end{aligned} \tag{22}$$

where $q_t = q_t(q_0,p_0)$ is the end point of a trajectory at time t with the initial condition (q_0,p_0). We stress that an outstanding feature of this correlation function is its low dimensionality in the integral only over q_0 coordinates. Equation (22) should be compared with Eqs. (2) and (5), where the integrals are to be carried out over the (q_0,p_0) coordinates.

C. Difficulty in semiclassical eigenvalue calculation

For quantization of integrable systems, the EBK condition has established itself both theoretically and numerically [17]. Quantization of chaotic systems is by far more difficult. The periodic orbit theory [16] has been a key theory to locate the eigenvalues with use of the trace

$$\begin{aligned} \mathrm{Tr}\,\delta(E-H) &= (2\pi\hbar)^{-1} \int dt \int dq \left\langle q \left| \exp\left(-\frac{i}{\hbar} Ht \right) \right| q \right\rangle \exp\left(\frac{i}{\hbar} Et \right) \\ &= (2\pi i\hbar)^{-N/2} \int dt \int dq \left| \frac{\partial q}{\partial p_0} \right|^{-1/2} \\ &\quad \times \exp\left(\frac{i}{\hbar} S_1(q,q,t) - i\pi \frac{\lambda}{2} + \frac{i}{\hbar} Et \right). \end{aligned} \tag{23}$$

This integral is well known to be asymptotically dominated only by the so-called periodic orbits, the number of which increases almost exponentially with the length of period. However, theory encounters the following difficulties. First, it follows $|\partial q_t/\partial p_0| = 0$ at caustics [15], which in turn brings about a divergence in the integral. This is not peculiar to chaos, however. The other difficulty is that a chaotic system is associated (by definition) with a fact that $|\partial q_t/\partial p_0|^{-1/2}$ becomes exponentially smaller as time passes in the final value representation, or equivalently $|\partial q_t/\partial p_0|^{1/2}$, becomes exponentially larger in the initial value representation. This comes from the imaginary part of b_k of Eq. (9)[22]. A Fourier transform of a function whose amplitude grows (or diminishes) exponentially necessarily results in a Lorentzian. We will show this more clearly along with numerical data in a companion paper [6]. (Note that b_k are purely real in integrable systems, and the Lorentzian does not follow in the EBK quantization.) Thus the trace formula [16] is expressed as the sum of a huge number of Lorentzians, each of which comes from different periodic orbits. Thus the δ-function spikes are not usually realized in practice in the periodic orbit sum or its variants.

Aurich *et al.* [23] detoured this approach by aiming at zeros of

$$\mathrm{Det}(E-H), \tag{24}$$

instead of searching poles of the resolvent. Unfortunately, however, it turned out that a variety of sophisticated (somewhat artificial) techniques to locate the zeros at correct positions on the E coordinate are required, including a Borel sum over the periodic orbit expansion [23]. It seems that this difficulty could not be avoided as long as a semiclassical expression of the spectrum is associated with the amplitude that comes from the stability matrix $[\partial Z_f/\partial Z_{i.}]$.

III. SQUARE ROOT OF VOLUME ELEMENTS IN SEMICLASSICAL INTEGRALS

To treat the transformation among the semiclassical integrals more systematically, we begin with a rather general multidimensional integral:

$$I = \int f(q_0) dq_0 = \int f(q_t) \left| \frac{\partial q_0}{\partial q_t} \right| dq_t. \tag{25}$$

In this expression, one should note that a small volume element dq_t can be an oriented volume with respect to dq_0, the sign of which is represented in terms of that of the Jacobian determinant $\partial q_t/\partial q_0$, since the volume can be inverted in many ways in addition to the change of its shape and volume. Let us define the square root of the volume elements of the integral [24], such that

$$I = \int f(q_0) dq_0^{1/2} dq_0^{1/2*}, \tag{26}$$

where $dq_0^{1/2*}$ is the complex conjugate of $dq_0^{1/2}$. The positive semidefinite quantity dq_0 (dq_t) is understood to be a product:

$$dq_0 \equiv |dq_0| = dq_0^{1/2} dq_0^{1/2*} \quad \text{and} \quad dq_t \equiv |dq_t| = dq_t^{1/2} dq_t^{1/2*}. \tag{27}$$

As in ordinary complex numbers, the square of $dq_t^{1/2}$ is not equal to dq_t unless the former is real valued. In this sense, the related confusion can be avoided if we note $dq_t \equiv |dq_t|$. We now define $dq_t^{1/2}$ as

$$dq_t^{1/2} = \exp\left[i\frac{\pi}{2} N(q_t) \right] |dq_t|^{1/2}, \tag{28}$$

where $N(q_t)$ is the sum of zeros up to the degeneracy of the following determinant picked up by a Jacobian determinant,

$$\frac{\partial q_t}{\partial q_{t=X}}, \tag{29}$$

along a path. It is convenient to set the reference of time $t=X$ at far remote past, symbolically denoted as $X=-\infty$.

At each time the determinant of Eq. (29) changes its sign, $N(q_t)$ acquires π (if the degeneracy of zero happens to be M, it acquires $M\pi$). In view of the physical continuity on the Riemann surface, $N(q_t)$ should proceed as $0\rightarrow\pi\rightarrow 2\pi\rightarrow 3\pi\rightarrow 4\pi\rightarrow\cdots$. This number is should be counted as a positive number in the positive time direction ($t>0$), and a negative number in the negative direction ($t<0$). Then, in order to comprehend Eqs. (17), (20), and (21), it is natural to use the following expression as our theoretical basis:

$$F(q_t,t)dq_t^{1/2}=F(q_0,0)dq_0^{1/2}, \tag{30}$$

which is valid only when the two points are connected by a trajectory. This represents a conservation law including the phase. In fact, Eq. (30) comes back to

$$F(q_t,t)\exp\left[i\frac{\pi}{2}N(q_t)\right]|dq_t|^{1/2} = F(q_0,0)\exp\left[i\frac{\pi}{2}N(q_0)\right]|dq_0|^{1/2}, \tag{31}$$

which in turn gives

$$F(q_t,t)=F(q_0,0)\exp\left(-i\frac{\pi}{2}[N(q_t)-N(q_0)]\right)\left|\frac{\partial q_t}{\partial q_0}\right|^{-1/2}, \tag{32}$$

where we understand the absolute value of the square of the Jacobian as

$$\left|\frac{\partial q_t}{\partial q_0}\right|^{-1/2}=\left|\frac{\partial q_0}{\partial q_t}\right|^{1/2}=\frac{|dq_0|^{1/2}}{|dq_t|^{1/2}}. \tag{33}$$

Since the Maslov index is defined as

$$M(q_0\rightarrow q_t)=N(q_t)-N(q_0) \tag{34}$$

we should have the sign in Eq. (17) fixed as

$$F(q_t,t)=F(q_0,0)\exp\left(-i\frac{\pi}{2}M(q_0\rightarrow q_t)\right)\left|\frac{\partial q_t}{\partial q_0}\right|^{-1/2}. \tag{35}$$

Also, both Eqs. (20) and (21) are naturally derived from Eq. (31). Incidentally, one can readily prove a useful identity

$$\begin{aligned} dq_t^{1/2*}dq_0^{1/2} &= \exp\left[-i\frac{\pi}{2}[N(q_t)-N(q_0)]\right]|dq_t|^{1/2}|dq_0|^{1/2} \\ &= \exp\left[-i\frac{\pi}{2}[N(q_t)-N(q_0)]\right]\left|\frac{\partial q_t}{\partial q_0}\right|^{1/2}dq_0 \\ &= \exp\left[-i\frac{\pi}{2}[N(q_t)-N(q_0)]\right]\left|\frac{\partial q_0}{\partial q_t}\right|^{1/2}dq_t. \end{aligned} \tag{36}$$

IV. CORRELATION FUNCTION

The quantum correlation function provides information from which the energy spectrum can be extracted through its Fourier transform

$$S(E)=\int \langle\Psi(0)|\Psi(t)\rangle\exp\left(\frac{i}{\hbar}Et\right)dt. \tag{37}$$

There can be a variety of ways of representing the correlation function in semiclassical mechanics, some of which can be particularly useful in quantizing classical chaos.

A. Various representations of the correlation function

Insert a formal equation $F(q_t,t)=F(q_0,0)dq_t^{-1/2}dq_0^{1/2}$ into Eq. (18), and we have

$$\begin{aligned} \Psi_{p_0}(q,t) &= \int \delta(q-q_t)\Psi_{local}^{p_0}(q_t,t)dq_t \\ &= \int \delta(q-q_t)\exp\left[\frac{i}{\hbar}S_2(q_t,p_0,t)\right] \\ &\quad \times F(q_t,t)dq_t^{1/2}dq_t^{1/2*} \\ &= \int \delta(q-q_t)\exp\left[\frac{i}{\hbar}S_2(q_t,p_0,t)\right] \\ &\quad \times F(q_0,0)dq_0^{1/2}dq_t^{1/2*}. \end{aligned} \tag{38}$$

Different representations of a wave function result in different expressions of the correlation function. An example is

$$\begin{aligned} C_{p_0}(s,t) &= \langle\Psi_{p_0}(s)|\Psi_{p_0}(t)\rangle \\ &= \int \exp\left[\frac{i}{\hbar}S_2(q_t,p_0,t)-\frac{i}{\hbar}S_2(q_t,p_0,s)\right] \\ &\quad \times F^*(q_t,s)F(q_t,t)dq_t \\ &= \int\int \delta(q_s-q_t)F^*(q_{01},0)F(q_{02},0) \\ &\quad \times\exp\left[\frac{i}{\hbar}S_1(q_t,q_{02},t)+\frac{i}{\hbar}p_0q_{02}\right. \\ &\quad -\frac{i}{\hbar}S_1(q_s,q_{01},s) \\ &\quad \left.-\frac{i}{\hbar}p_0q_{01}\right]dq_{01}^{1/2*}dq_{02}^{1/2}dq_s^{1/2}dq_t^{1/2*}. \end{aligned} \tag{39}$$

The last equality makes use of the last term of Eq. (38). Therefore we have a basic property

$$C_{p_0}(s,t)^* = \langle \Psi_{p_0}(s)|\Psi_{p_0}(t)\rangle^* = \int\int \delta(q_s - q_t)F(q_{01},0)F^*(q_{02},0) \times \exp\left[-\frac{i}{\hbar}S_2(q_t,q_{02},t) - \frac{i}{\hbar}p_0 q_{02} + \frac{i}{\hbar}S_2(q_s,q_{01},s) + \frac{i}{\hbar}p_0 q_{01}\right] dq_{01}^{1/2} dq_{02}^{1/2*} dq_s^{1/2*} dq_t^{1/2} \quad (40)$$

Thus the relations $C_{p_0}(s,t)^* = C_{p_0}(t,s)$ and $C_{p_0}(0,t)^* = C_{p_0}(0,-t)$ naturally result.

The standard form of the correlation function used in practice is of course

$$C_{p_0}(0,t) = \int F^*(q_t,0)\exp\left[-\frac{i}{\hbar}p_0 q_t\right] \times \exp\left[\frac{i}{\hbar}S_2(q_t,p_0,t)\right]F(q_0,0)\,dq_0^{1/2}dq_t^{1/2*} = \int F^*(q_t,0)F(q_0,0)\exp\left[-\frac{i}{\hbar}p_0 q_t + \frac{i}{\hbar}S_1(q_t,q_0,t) - i\frac{\pi}{2}(N(q_t)-N(q_0))\right]|dq_t|^{1/2}|dq_0|^{1/2}, \quad (41)$$

which looks beautifully symmetric. In the second equality in this equation, Eq. (36) was used. The conversion into the form of Eq. (22) is easy with the use of the identity

$$|dq_t|^{1/2}|dq_0|^{1/2} = \left|\frac{\partial q_t}{\partial q_0}\right|^{1/2} dq_0, \quad (42)$$

and with the help of Eq. (34).

B. Evaluation of a correlation function

The spectrum we are going to numerically evaluate [6] is a Fourier integral of the form

$$S(E) = \mathrm{Re}\lim_{T\to\infty}\frac{2}{T}\int_0^{T/2} C_{p_0}(-t,t)\exp\left(2\frac{i}{\hbar}Et\right)dt. \quad (43)$$

Then Eq. (39) requires two trajectories that end up at the same point q_t by different routes, namely,

$$q_t(q_{02},p_0) = q_{-t}(q_{01},p_0), \quad (44)$$

where $q_t(q_{02},p_0)$, for instance, indicates a position q_t reached by a trajectory at time t, that starts with an initial condition (q_{02},p_0) at time $t=0$. [Note that $q_{-t}(q_{01},p_0) = q_t(q_{01},-p_0)$.] If it happens that $q_{01}=q_{02}$, Eq. (44) is readily satisfied. However, these paths are not connected smoothly at $t=0$ unless $p_0=0$, since a momentum jump from $-p_0$ to p_0 is necessary.

To see what kind of trajectories make major contributions, let us take an average of $S(E)$ of Eq. (43) over the initial momentum p_0. It safices to see an average of $C_{p_0}(-t,t)$ over p_0, and focus on

$$\int dp_0 C_{p_0}(-t,t) = 2\pi\hbar\int\int \delta(q_{-t}-q_t)F^*(q_{01},0)F(q_{02},0)\delta(q_{01}-q_{02}) \times \exp\left[\frac{i}{\hbar}S_1(q_t,q_{02},t) - \frac{i}{\hbar}S_1(q_s,q_{01},-t)\right] dq_{01}^{1/2*}dq_{02}^{1/2}dq_{-t}^{1/2}dq_t^{1/2*}. \quad (45)$$

The trajectories contributing to this integral should satisfy

$$q_{01} = q_{02} \quad (46)$$

and

$$q_t(q_{01},p_0) = q_{-t}(q_{01},p_0) = q_t(q_{01},-p_0) \quad (47)$$

due to the newly appearing δ function. The stationary phase argument applied to Eq. (45) requires

$$p_t(q_{01},p_0) = -p_t(q_{01},-p_0). \quad (48)$$

However, general trajectories do not satisfy Eqs. (47) and (48) simultaneously. There are two important exceptions. One is the case of a periodic orbit in phase space. Only for periodic orbits whose periods T happen to be the same as time t, for which $p_T(q_{01},p_0)=p_0$ and $p_T(q_{01},-p_0)=-p_0$ are realized, the correlation function can have significant values. This would give an alternative expression to periodic orbit theory [16], to which we do not intend to return in this paper.

The other special case arises from the trajectories of

$$p_0 = 0. \quad (49)$$

This condition is stronger in that $p_0=0$ ensures the equalities in both Eqs. (47) and (48) for any q_{01}, q_t, and t. Furthermore, trajectories having $p_0=0$ at $t=0$ must be appropriate to represent a motion corresponding to standing waves, since they are generally formed in a fixed boundary. In view of the well-known difficulty inherent in periodic orbit theory, we hereafter focus on the correlation function composed of the trajectories of $p_0=0$.

C. A correlation function free of the amplitude factor

It was suggested from the above analysis that a potentially useful representation of the correlation function can be generated in terms of $p_0=0$ and $q_{02}=q_{01}$ to quantize classically chaotic systems. We resume from Eq. (39), with $p_0=0$ and $s=-t$. Even if we set $p_0=0$, those trajectories satisfying $q_t(q_{01},p_0)=q_{-t}(q_{02},p_0)$, with $q_{02}\neq q_{01}$, remain in the integration of Eq. (39). However, since they are disconnected in configuration space, and because of Eq. (46), their contributions must be very small. We thus deliberately disregard these trajectories and keep only those trajectories satisfying $p_0=0$ and $q_{02}=q_{01}$, leaving an ''essential part'' of the correlation function as

$$\tilde{C}_0(-t,t)=\int\int \delta(q_{-t}-q_t)F^*(q_{01},0)F(q_{01},0)\times\exp\left[\frac{i}{\hbar}S_1(q_t,q_{01},t)-\frac{i}{\hbar}S_1(q_{-t},q_{01},-t)\right]\times dq_{01}^{1/2*}dq_{01}^{1/2}dq_{-t}^{1/2}dq_t^{1/2*}, \tag{50}$$

in which both the geometry and the action integrals along the trajectories are now smoothly connected, and

$$q_{-t}(q_{01},p_0=0)=q_t(q_{01},p_0=0). \tag{51}$$

We have no way to estimate exactly how small are the contributions made by all the neglected trajectoies. Also, so far there is no ''practical method'' to overcome the divergence problem described above in quantizing chaotic systems. (By a practical method, we mean a method that can be applied to molecular vibrational states of more than three dimensions.) We will therefore numerically test the extracted correlation function by applying it to a vibrational problem of a seven atomic cluster [6], since Eq. (50) has a distinguished advantage in quantizing chaos, as shown below.

$\tilde{C}_0(-t,t)$ of Eq. (50) is rewritten to appear as

$$\begin{aligned}\tilde{C}_0(-t,t)=&\int\int \delta(q_{-t}-q_t)|F(q_{01},0)|^2\exp\left[\frac{i}{\hbar}S_1(q_t,q_{01},t)-\frac{i}{\hbar}S_1(q_{-t},q_{01},-t)\right]dq_{01}\\&\times\exp\left[i\frac{\pi}{2}N(q_{-t})\right]|dq_{-t}|^{1/2}\\&\times\exp\left[-i\frac{\pi}{2}N(q_t)\right]|dq_t|^{1/2}\\=&\int dq_{01}|F(q_{01},0)|^2\exp\left[2\frac{i}{\hbar}S_1(q_t,q_{01},t)-i\frac{\pi}{2}M(q_{-t}\to q_t)\right],\end{aligned} \tag{52}$$

where the Maslov index is defined as usual: $M(q_{-t}\to q_t)=N(q_t)-N(q_{-t})$. It is remarkable that an annoying amplitude factor, such as $|\partial q_t/\partial p_0|^{-1/2}$ or $|\partial q_t/\partial q_{01}|^{1/2}$, is missing in this expression. Recall that $|\partial q_t/\partial p_0|^{-1/2}$ diminishes exponentially for a chaotic system, while $|\partial q_t/\partial q_{01}|^{1/2}$ diverges exponentially; both thereby causing numerical difficulties in the Fourier transform. Such difficulties will be numerically shown very clearly in our following paper [6]. On the other hand, the Maslov index remains in the correlation function, which constitutes an essential part of the quantum phase. Thus a calculation of the Maslov index, through the computation of the matrix $[\partial q_t/\partial q_{01}]$, or by a some other technique, is unavoidable.

To identify the property of Eq. (52), let us investigate the element $dq_{-t}^{1/2}dq_t^{1/2*}$ a little further. Owing to the characteristics of paths adopted in relation (52), $\delta(q_{-t}-q_t)dq_{-t}^{1/2}dq_t^{1/2*}$ can be rewritten as [see Eq. (36), replacing $dq_0^{1/2}$ with $dq_{-t}^{1/2}$]

$$\begin{aligned}\delta(q_{-t}-q_t)dq_{-t}^{1/2}dq_t^{1/2*}&=\delta(q_{-t}-q_t)\exp\left[-i\frac{\pi}{2}(N(q_t)-N(q_{-t}))\right]|dq_t|^{1/2}|dq_t|^{1/2}\\&=\delta(q_{-t}-q_t)\times\exp[-i\pi N(q_t)]\left|\frac{\partial q_t}{\partial q_{-t}}\right|^{1/2}dq_{-t}\\&=\delta(q_{-t}-q_t)\times\exp[-i\pi N(q_t)]\left|\frac{\partial q_{-t}}{\partial q_t}\right|^{1/2}dq_t.\end{aligned} \tag{53}$$

Here we see the Jacobian determinants that characterize either the initial and final representations. However, both $\partial q_t/\partial q_{-t}$ and $\partial q_{-t}/\partial q_t$ are unity, or

$$\left.\frac{\partial q_t}{\partial q_{-t}}\right|_{p_0=0}=\left.\frac{\partial q_{-t}}{\partial q_t}\right|_{p_0=0}=1, \tag{54}$$

under Eq. (51). This is rather obvious, since the stability matrix should be considered only in the limited manifold of trajectories that satisfies $p_0=0$ at $t=0$, and hence comes back exactly to the same points in such a way that

$$q_{-t}+\Delta q\to q_t+\Delta q \quad \text{as time passes as } -t\to 0\to t \tag{55}$$

for any arbitrary displacement of Δq. Therefore, the amplitude factor for this particular case is canceled out.

The energy spectra in integrable systems are quantized in terms of the information of the action integral and the Maslov index alone, as typically realized in the EBK condition. The energy spectra extracted from the correlation function of Eq. (52),

$$S(E)=\frac{2}{T}\mathrm{Re}\lim_{T\to\infty}\int_0^{T/2}\tilde{C}_0(-t,t)\exp\left(2\frac{i}{\hbar}Et\right)dt$$
$$=\frac{2}{T}\lim_{T\to\infty}\int_0^{T/2}dt\int dq_{01}|F(q_{01},0)|^2\cos\left[\frac{2}{\hbar}S_1(q_t,q_{01},t)-\frac{\pi}{2}M(q_{-t}\to q_t)+\frac{2}{\hbar}Et\right], \quad (56)$$

does not make a formal distinction between chaotic and integrable systems. On the other hand, it is never trivial that the spectra arising from Eq. (52) can cover the entire spectrum, since $\tilde{C}_0(-t,t)$ is an extraction from the full correlation function. In other words, there can exist a possibility that some spectral peaks may be missing from those spectra. Nevertheless, this expression is quite promising, and deservers numerical tests [6].

V. CONCLUDING REMARKS

We have developed a systematic algebra for coordinate transformation in semiclassical integrals, which can provide a convenient perspective and practice in the actual performance of transformations among the initial and final value representations. Various variants for representations of quantum correlation functions have been shown.

As a harvest of this algebraic approach, we have found a representation of a semiclassical correlation function based on an action decomposed function, [Eq. (52)], which is free from amplitude factors such as $|\partial q_f/\partial q_i|^{-1/2}$. Since this factor quite often causes fatal troubles in numerical calculations of the energy spectra for a chaotic system, it can serve as a useful alternative to existing methods for quantizing classically chaotic systems. We will show a practical application of the thus obtained amplitude-free correlation function in a companion paper [6], in which a systematic numerical study is made of the vibrational spectrum of an Ar_7-like cluster with various methods.

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