Addendum to "Time-dependent variational principle with constraints for parametrized wave functions"

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The sensitivity analysis for the time-dependent variational principle (TDVP) with constraints for parametrized wave functions [K. Ohta, Phys. Rev. A **70**, 022503 (2004)] is investigated to assess the geodesic deviation caused by external parameters. The constraints for the sensitivity functions, inherited from the TDVP, are dealt with their consistency conditions. As an example of the sensitivity analysis, the geodesic deviation in the neighborhood of stationary states is investigated for general wave functions.

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Time-dependent wave functions have been used to obtain not only numerical but also vivid and intuitive understandings of various quantum-dynamical processes. As one of the most practical methods to calculate the time development of wave functions, the time-dependent variational principle (TDVP) [1–3] has become popular. Recent works using the TDVP have been reviewed in Ref. [4]. In particular, from the methodological aspects, the stationary action principle [5,6] in the TDVP has attracted much interest because it leads to pseudo-classical-mechanics of the variational parameters [7–9]. All the rich apparatus developed for the study of classical mechanics can be applied to the TDVP. If we consider constraints in the TDVP [10,11], the structure of the pseudo-classical-mechanics enables us to utilize Dirac's constrained classical mechanics [12,13]. The constraints in the TDVP are classified into the first and second classes according to Dirac's terminology.

In this study, we have investigated the sensitivity analysis [14] for the TDVP. The sensitivity analysis is a technique to assess how the solutions in nonlinear systems depend on external parameters. For example, in the Born-Oppenheimer approximation for molecules, the sensitivity analysis enables us to calculate the dynamical variation of electronic states due to nuclear coordinates. From the viewpoint of the pseudo-classical-mechanics of the TDVP, the sensitivity analysis assesses the deviation between nearby trajectories, i.e., the geodesic deviation [15] caused by external parameters. The present paper consists of three parts. In the first part, we have summarized briefly the TDVP with constraints as pseudo-classical-mechanics. In the second part, the sensitivity analysis for the TDVP has been formulated to study the geodesic deviation. As an example of the sensitivity analysis, we have given the geodesic deviation in the neighborhood of stationary states for general wave functions in the third part.

First, we assume a trial wave function of the TDVP whose time development is described through complex variational parameters $z=\{z_1,z_2,\ldots,z_M\}$ as

$$\Psi(z,\alpha,x)=\Psi(z_1(t,\alpha),z_2(t,\alpha),\ldots,z_M(t,\alpha),\alpha,x), \tag{1}$$

where α denotes the real external parameters. We consider constraints for the wave function in the TDVP [10,11]. The normalization condition for Ψ,

$$g_0(z^*,z,\alpha)=\langle\Psi|\hat{1}|\Psi\rangle-1=0, \tag{2}$$

is identified as a first-class constraint in Dirac's notation [12,13]. The second-class constraints are also considered with appropriate Hermitian operators $\hat{g}_i$ as

$$g_i(z^*,z,\alpha)=\langle\Psi|\hat{g}_i(\alpha,x)|\Psi\rangle=0 \quad (i=1,\ldots,2L). \tag{3}$$

The real Lagrangian, including the constraints with the Lagrange multipliers $\{\lambda_i\}_{i=0,2L}$, is defined by

$$L(z^*,z,\alpha)=\frac{i\hbar}{2}\sum_{i=1}^{M}\dot{z}_i\left\langle\Psi|\frac{\partial\Psi}{\partial z_i}\right\rangle-\frac{i\hbar}{2}\sum_{i=1}^{M}\dot{z}_i^*\left\langle\frac{\partial\Psi}{\partial z_i}|\Psi\right\rangle-\langle\Psi|\hat{H}|\Psi\rangle-\sum_{i=0}^{2L}\lambda_i g_i. \tag{4}$$

By requiring the action functional $S(z^*,z,\alpha)=\int_{t_0}^{t_1}L(z^*,z,\alpha)dt$ to be stationary with fixed boundary conditions [5–7], we obtain the equations of motion (EOMs) as the Euler equation

$$\dot{z}_i=\frac{1}{i\hbar}\sum_{j=1}^{M}(\boldsymbol{C}^{-1})_{ij}\frac{\partial K(z^*,z,\alpha)}{\partial z_j^*} \quad (i=1,\ldots,M), \tag{5}$$

where $\boldsymbol{C}^{-1}$ is the inverse of the overlap matrix between the local bases, $(\boldsymbol{C})_{ij}=\langle\partial\Psi/\partial z_i|\partial\Psi/\partial z_j\rangle$, and K is a new Hamiltonian defined by

$$K(z^*,z,\alpha)=\langle\Psi|\hat{H}|\Psi\rangle+\lambda_0 g_0+\sum_{j=1}^{2L}\lambda_j g_j=H+\lambda_0 g_0+\sum_{j=1}^{2L}\lambda_j g_j. \tag{6}$$

By using Eq. (5), we obtain the time development of a function $\tilde{F}(t,\alpha)=F(z^*,z,\alpha)$ as

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$$\left.\frac{\partial \tilde{F}(t,\alpha)}{\partial t}\right|_{\alpha} = \left.\frac{\partial F(z^*,z,\alpha)}{\partial t}\right|_{\alpha} = \frac{1}{i\hbar}\{F,K\}_{GP}, \tag{7}$$

where the complex generalized Poisson bracket [7] is defined by

$$\{A,B\}_{GP} = \sum_{i=1}^{M}\sum_{j=1}^{M}\left(\frac{\partial A}{\partial z_i}(\boldsymbol{C}^{-1})_{ij}\frac{\partial B}{\partial z_j^*} - \frac{\partial B}{\partial z_i}(\boldsymbol{C}^{-1})_{ij}\frac{\partial A}{\partial z_j^*}\right). \tag{8}$$

If the local bases $\{\partial\Psi/\partial z_i\}_{i=1,M}$ construct a complete system [11], we have

$$\{\langle\Psi|\hat{A}|\Psi\rangle,\langle\Psi|\hat{B}|\Psi\rangle\}_{GP} = \langle\Psi|[\hat{A},\hat{B}]_-|\Psi\rangle \tag{9}$$

for any Hermitian operators $\hat{A}$ and $\hat{B}$.

The Lagrange multipliers $\{\lambda_i\}_{i=0,2L}$ in Eq. (6) are determined so that the consistency conditions [12,13] for the constraints are satisfied as

$$\left.\frac{\partial g_i(z^*,z,\alpha)}{\partial t}\right|_{\alpha} = \frac{1}{i\hbar}\{g_i,K\}_{GP} = 0 \quad (i=0,1,\ldots,2L). \tag{10}$$

If the local bases are complete, the consistency condition (10) for the normalization condition (2) is automatically satisfied because the constraining operator for the normalization is the unit operator which is commutable with any operator. So the Lagrange multiplier λ_0 for the normalization condition is left as a gauge-fixing freedom,

$$\lambda_0(t,\alpha) = \Lambda(t,\alpha), \tag{11}$$

where the real function Λ can be selected arbitrarily. On the other hand, the Lagrange multipliers $\{\lambda_i\}_{i=1,2L}$ for the constraints (3), which are classified as second-class constraints, are determined uniquely from Eq. (10) as

$$\lambda_i(z^*,z,\alpha) = -\sum_{j=1}^{2L}\{H,g_j\}_{GP}(\boldsymbol{G}^{-1})_{ji} \quad (i=1,\ldots,2L), \tag{12}$$

where $(\boldsymbol{G})_{ji}=\{g_j,g_i\}_{GP}$. Because the Lagrange multipliers are determined to satisfy the consistency conditions (10), as mentioned above, we need to impose the constraints only on initial values of the variational parameters $\{z_i,z_i^*\}_{i=1,M}$.

Next we formulate the sensitivity analysis [14] which is a technique to assess how the solutions in nonlinear systems depend on external parameters. The sensitivity functions of the variational parameters $z=\{z_1,z_2,\ldots,z_M\}$ for real external parameters α are defined by

$$\gamma_i(t,\alpha) = \left.\frac{\partial z_i(t,\alpha)}{\partial\alpha}\right|_t \quad (i=1,\ldots,M). \tag{13}$$

The sensitivity equations, which describe the time development of the sensitivity functions, can be obtained by differentiating directly the TDVP EOM (5) with respect to external parameters α as

$$\begin{aligned}\dot{\gamma}_i = {}& \frac{1}{i\hbar}\sum_{l=1}^{M}\left\{\left[\sum_{j=1}^{M}\left(\frac{\partial(\boldsymbol{C}^{-1})_{ij}}{\partial z_l}\frac{\partial K}{\partial z_j^*} + (\boldsymbol{C}^{-1})_{ij}\frac{\partial^2 K}{\partial z_j^*\partial z_l}\right)\right]\gamma_l\right. \\ & \left.+\left[\sum_{j=1}^{M}\left(\frac{\partial(\boldsymbol{C}^{-1})_{ij}}{\partial z_l^*}\frac{\partial K}{\partial z_j^*} + (\boldsymbol{C}^{-1})_{ij}\frac{\partial^2 K}{\partial z_j^*\partial z_l^*}\right)\right]\gamma_l^*\right\} \\ & + \frac{1}{i\hbar}\sum_{j=1}^{M}\left[\left.\frac{\partial(\boldsymbol{C}^{-1})_{ij}}{\partial\alpha}\right|_{z^*,z}\frac{\partial K}{\partial z_j^*} + (\boldsymbol{C}^{-1})_{ij}\left.\frac{\partial}{\partial\alpha}\left(\frac{\partial K}{\partial z_j^*}\right)\right|_{z^*,z}\right] \\ & (i=1,\ldots,M).\end{aligned} \tag{14}$$

The sensitivity equations (14) are inhomogeneous linear differential equations with variable coefficients. The external parameters α can directly induce the deviation through the inhomogeneous terms, the third terms in the right-hand side of Eq. (14). In order to obtain the sensitivity functions by Eq. (14), we need to solve the TDVP EOM (5) beforehand. However, except for some simple cases, we will be forced to solve these equations numerically where we can solve the TDVP EOM (5) and the sensitivity equations (14) simultaneously.

Constraints for the sensitivity functions $\{\gamma_i,\gamma_i^*\}_{i=1,M}$ are also obtained by differentiating the constraints (2) and (3) with respect to α as

$$\left.\frac{\partial g_i(z^*,z,\alpha)}{\partial\alpha}\right|_t = \sum_{j=1}^{M}\left(\frac{\partial g_i}{\partial z_j}\gamma_j(t,\alpha) + \frac{\partial g_i}{\partial z_j^*}\gamma_j^*(t,\alpha)\right) + \left.\frac{\partial g_i}{\partial\alpha}\right|_{z^*,z} = 0 \quad (i=0,\ldots,2L). \tag{15}$$

Because the Lagrange multipliers $\{\lambda_i(z^*,z,\alpha)\}_{i=0,2L}$ are determined so that the consistency conditions (10) for the constraints (2) and (3) should be satisfied for every value of α, we have

$$\frac{\partial}{\partial\alpha}\left(\left.\frac{\partial g_i(z^*,z,\alpha)}{\partial t}\right|_{\alpha}\right)\Bigg|_t = \frac{\partial}{\partial t}\left(\left.\frac{\partial g_i(z^*,z,\alpha)}{\partial\alpha}\right|_t\right)\Bigg|_{\alpha} = 0 \quad (i=0,\ldots,2L). \tag{16}$$

The second equations in Eq. (16) give the consistency conditions for the constraints (15). Then, as in the case of the TDVP, we need to impose the constraints (15) only on initial values of the sensitivity functions $\{\gamma_i,\gamma_i^*\}_{i=1,M}$. If the constraints $\{g_i\}_{i=0,2L}$ have no explicit dependence on external parameters α, we adopt vanishing initial values,

$$\gamma_i(t_0,\alpha) = \gamma_i^*(t_0,\alpha) = 0 \quad (i=1,\ldots,M), \tag{17}$$

which obviously satisfy Eq. (15). The effect from the variation of the starting points of the TDVP trajectories $\{z_i,z_i^*\}_{i=1,M}$ can be excluded.

From the viewpoint of the pseudo-classical-mechanics of the TDVP, the sensitivity functions assesses the geodesic deviation caused by external parameters α. Let us consider, for example, the Born-Oppenheimer approximation for molecules. The TDVP trajectories $\{z_i,z_i^*\}_{i=1,M}$ describe the time development of electronic wave functions by regarding the adiabatic nuclear coordinates R as external parameters α. The geodesic deviation [15] caused by the variation of R is written explicitly as

$$z_i(t,R+\varepsilon) - z_i(t,R) = \gamma_i(t,R)\varepsilon + o(\varepsilon^2), \tag{18}$$

where the sensitivity functions γ_i give directly the first-order variation of the TDVP trajectories z_i. Unless the absolute values of the sensitivity functions remain small, the adiabatic separation between the electronic and nuclear modes may not be sufficient. We should consider the possibility of nonadiabatic couplings between the modes. Mathematically, the equation for the geodesic deviation is obtained as the Jacobi equation, which is a variational equation for the Euler equation [16]. The Jacobi equation, however, usually assesses the geodesic deviation caused by boundary or initial conditions. The deviation by external parameters as shown in Eq. (18) is not considered. Moreover, the TDVP Lagrangian (4), which is linear in $\dot{z}_i$ and $\dot{z}_i^*$, leads to

$$\frac{\partial^2 L}{\partial \dot{z}_i^* \partial \dot{z}_j} = \frac{\partial^2 L}{\partial \dot{z}_i^* \partial \dot{z}_j^*} = 0. \tag{19}$$

Therefore, the Jacobi equation in the TDVP is reduced to a set of linear differential equations of the first order which are exactly the same as the homogeneous part of Eq. (14). So, if we regard the initial values of the trajectories as external parameters α, we can obtain the Jacobi equation from the sensitivity equations (14) because the inhomogeneous terms will obviously vanish. Thus, the sensitivity equations can be considered as an extension of the Jacobi equation to external parameters.

For an example of the sensitivity analysis, we consider a stationary state $\{\dot{z}_i = \dot{z}_i^* = 0\}_{i=1,M}$, which is one of the special trajectories of the TDVP. The geodesic deviation in the neighborhood of the stationary state is considered for general wave functions. The stationary state can be obtained from the TDVP EOM (5) as

$$\frac{\partial K}{\partial z_i} = \frac{\partial K}{\partial z_i^*} = 0 \quad (i = 1, \ldots, M). \tag{20}$$

By using Eq. (20), we obtain the sensitivity equations (14) as the equations with constant coefficients,

$$\dot{\gamma}_i = \frac{1}{i\hbar}\sum_{l=1}^{M}\left[\left(\sum_{j=1}^{M}[(\boldsymbol{C}^{-1})_{ij}(\boldsymbol{A})_{jl}]\right)\gamma_l + \left(\sum_{j=1}^{M}[(\boldsymbol{C}^{-1})_{ij}(\boldsymbol{B})_{jl}]\right)\gamma_l^*\right] + \frac{1}{i\hbar}\sum_{j=1}^{M}[(\boldsymbol{C}^{-1})_{ij}\boldsymbol{D}_j] \quad (i = 1, \ldots, M), \tag{21}$$

where we have introduced the constant matrices $(\boldsymbol{A})_{jl} = \partial^2 K/\partial z_j^* \partial z_l$, $(\boldsymbol{B})_{jl} = \partial^2 K/\partial z_j^* \partial z_l^*$, and the constant vector $(\boldsymbol{D})_j = (\partial/\partial\alpha)(\partial K/\partial z_j^*)|_{z^*,z}$. We here consider only the normalization constraint g_0, Eq. (2). Using the freedom of gauge fixation (11) for the first-class constraint [10–13], we take the Lagrange multiplier as $\lambda_0 = -\langle\Psi|\hat{H}|\Psi\rangle/\langle\Psi|\Psi\rangle$. The Hamiltonian (6) becomes the usual expression of the energy which is normalized explicitly as

$$K(z^*, z, \alpha) = H + \lambda_0(\langle\Psi|\Psi\rangle - 1) = \frac{\langle\Psi|\hat{H}|\Psi\rangle}{\langle\Psi|\Psi\rangle}. \tag{22}$$

Then Eq. (20) can be identified as the extremal conditions of the energy in the usual variational principle for stationary states [17].

The inhomogeneous equations (21) with their complex conjugates are written simply in the matrix form,

$$\dot{\boldsymbol{\Gamma}} = \begin{pmatrix} \dot{\boldsymbol{\gamma}} \\ \dot{\boldsymbol{\gamma}}^* \end{pmatrix} = \frac{1}{i\hbar}\begin{pmatrix} \widetilde{\boldsymbol{A}} & \widetilde{\boldsymbol{B}} \\ -\widetilde{\boldsymbol{B}}^* & -\widetilde{\boldsymbol{A}}^* \end{pmatrix}\begin{pmatrix} \boldsymbol{\gamma} \\ \boldsymbol{\gamma}^* \end{pmatrix} + \frac{1}{i\hbar}\begin{pmatrix} \widetilde{\boldsymbol{D}} \\ -\widetilde{\boldsymbol{D}}^* \end{pmatrix}, \tag{23}$$

where the tilde refers to the matrices and vectors multiplied by $\boldsymbol{C}^{-1}$ from the left.

As in the usual method of variation of constants [18], let us first solve the homogeneous part of Eq. (23) by assuming the following periodic form:

$$\gamma_i(t,\alpha) = \sum_{n=1}^{M}(a_n X_{in} e^{(1/i\hbar)\omega_n t} + a_n^* Y_{in}^* e^{-(1/i\hbar)\omega_n t}), \tag{24}$$

where $\{a_n\}_{n=1,M}$ are nonvanishing constants. By substituting Eq. (24) and its complex conjugate into the homogeneous part of Eq. (23), we obtain an eigenvalue equation

$$\begin{pmatrix} \widetilde{\boldsymbol{A}} & \widetilde{\boldsymbol{B}} \\ -\widetilde{\boldsymbol{B}}^* & -\widetilde{\boldsymbol{A}}^* \end{pmatrix}\begin{pmatrix} \boldsymbol{X} & \boldsymbol{Y}^* \\ \boldsymbol{Y} & \boldsymbol{X}^* \end{pmatrix} = \begin{pmatrix} \boldsymbol{X} & \boldsymbol{Y}^* \\ \boldsymbol{Y} & \boldsymbol{X}^* \end{pmatrix}\begin{pmatrix} \boldsymbol{\omega} & \boldsymbol{0} \\ \boldsymbol{0} & -\boldsymbol{\omega} \end{pmatrix}. \tag{25}$$

This Eq. (25) from the homogeneous part of Eq. (23) is identical to the general equations-of-motion method by Rowe [19] which is an extension of the random phase approximation [17] to general wave functions. Assuming real and nonvanishing eigenvalues in $\boldsymbol{\omega}$ [17], we here normalize the eigenvectors with the $2M$-dimensional metric as

$$\begin{pmatrix} \boldsymbol{X}^\dagger & \boldsymbol{Y}^\dagger \\ \boldsymbol{Y}^{*\dagger} & \boldsymbol{X}^{*\dagger} \end{pmatrix}\begin{pmatrix} \boldsymbol{C} & \boldsymbol{0} \\ \boldsymbol{0} & -\boldsymbol{C}^* \end{pmatrix}\begin{pmatrix} \boldsymbol{X} & \boldsymbol{Y}^* \\ \boldsymbol{Y} & \boldsymbol{X}^* \end{pmatrix} = \begin{pmatrix} \boldsymbol{\sigma} & \boldsymbol{0} \\ \boldsymbol{0} & -\boldsymbol{\sigma} \end{pmatrix}, \tag{26}$$

where $(\boldsymbol{\sigma})_{nm} = \delta_{nm}$ for $\omega_n > 0$, and $(\boldsymbol{\sigma})_{nm} = -\delta_{nm}$ for $\omega_n < 0$.

Now we solve the inhomogeneous equations (23) by considering $\{a_n\}_{n=1,M}$ in Eq. (24) as functions of time,

$$a_n(t,\alpha) = -\frac{1}{\omega_n}(P_n e^{-(1/i\hbar)\omega_n t} + Q_n), \tag{27}$$

where P_n and Q_n are constants. By substituting Eq. (24) again into Eq. (23), we obtain the constants P_n in the matrix form

$$\begin{pmatrix} \boldsymbol{P} \\ -\boldsymbol{P}^* \end{pmatrix} = \begin{pmatrix} \boldsymbol{\sigma} & \boldsymbol{0} \\ \boldsymbol{0} & -\boldsymbol{\sigma} \end{pmatrix}\begin{pmatrix} \boldsymbol{X}^\dagger & \boldsymbol{Y}^\dagger \\ \boldsymbol{Y}^{*\dagger} & \boldsymbol{X}^{*\dagger} \end{pmatrix}\begin{pmatrix} \boldsymbol{D} \\ \boldsymbol{D}^* \end{pmatrix} \tag{28}$$

where Eqs. (25) and (26) have been used with simple algebraic manipulations. If we can adopt the vanishing initial values (17) for $\gamma_i(t_0,\alpha)$, we finally obtain the sensitivity functions

$$\gamma_i(t,\alpha) = \sum_{n=1}^{M} \frac{1}{\omega_n}[X_{in}(e^{(1/i\hbar)\omega_n(t-t_0)} - 1)P_n + Y_{in}^*(e^{-(1/i\hbar)\omega_n(t-t_0)} - 1)P_n^*]. \quad (29)$$

External parameters α can cause a geodesic deviation through the nonvanishing constants P_n even from vanishing initial values of the deviation. The dynamical stability of the periodic solutions (29) requires real and nonvanishing eigenvalues ω_n. This is also related to the static stability in the usual variational calculations for stationary states. That is, the dynamical instability with complex ω_n indicates the possibility of the static instability as is well known in the general equations-of-motion method by Rowe [19].

In conclusion, we have presented a sensitivity analysis for the TDVP. The obtained sensitivity equations can be considered as an extension of the Jacobi equation to the geodesic deviation which is caused by external parameters. If we consider constraints in the TDVP, the sensitivity analysis inherits them. However, as in the case of the TDVP, the consistency conditions for the constraints are also satisfied in the sensitivity analysis. We need to impose the constraints only on initial values of the sensitivity functions. For a simple example of the sensitivity analysis, we have considered a stationary state as one of the special trajectories of the TDVP. The geodesic deviation of the stationary state has led to periodic solutions which relate the dynamical and the static stabilities with each other as in the general equations-of-motion method.

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