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TIME-DEPENDENT HARTREE-FOCK PHASE: UNIQUE IMPLICATION
OF VARIATIONAL PRINCIPLE

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ABSTRACT

The Time-Dependent Hartree-Fock (TDHF) variational principle is shown to yield a unique solution with no indeterminacy in its time-dependent phase. The long-standing contrary opinion arises from an impermissible restriction to norm-preserving variations.

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"Customary" TDHF is Phase Ambiguous

We take as the fundamental variational principle for the time-dependent Schrodinger theory the "Action-integral" form

$$\delta \int_{t_1}^{t_2} \langle \Phi(t) | H - i\hbar \partial/\partial t | \Phi(t) \rangle dt = 0. \quad (1)$$

Arbitrary variations of (1) which vanish at (t_1, t_2) are allowed. Equation (1) then implies the exact time-dependent Schrödinger equation [2-3].

For Time-Dependent Hartree-Fock theory only determinants are to be allowed for the solution Φ and $\Phi + \delta\Phi$. But the additional constraint that the variation be norm-preserving, i.e.,

$$\langle \Phi | \delta\Phi \rangle = 0, \quad (2)$$

has customarily[†] been imposed upon the variational process. The most general determinantal variation so restricted is

$$\delta\Phi = \sum_{\substack{\mu \in F \\ \sigma \notin F}} \epsilon_{\mu}^{\sigma}(t) a_{\sigma}^{\dagger} a_{\mu} \Phi, \quad (3)$$

where F denotes the set of single-particle states filled in Φ , and the coefficients, $\epsilon_{\mu}^{\sigma}(t)$, are arbitrary functions of time which vanish at t_1, t_2 .

The variational principle, eq. (1), then yields the customary conditions

$$\langle \Phi | a_{\mu}^{\dagger} a_{\sigma} (H - i\hbar \partial/\partial t) | \Phi \rangle = 0, \quad (\mu \in F, \sigma \notin F), \quad (4)$$

which are fulfilled for any determinantal solution to the TDHF equation,

$$\mathcal{H}_{\text{HF}}[\Phi]\Phi = i\hbar \dot{\Phi} \quad (5)$$

because the TDHF Hamiltonian, $\mathcal{H}_{\text{HF}}$, is a functional of H and Φ so constructed as to have matrix elements identical with those of the exact Hamiltonian, H , between Φ and its one-particle one-hole states:

[†]By "customary" we mean the apparently generally accepted derivation of the TDHF variational conditions (4), which as we shall show does not yield the variational conditions (12), because assumption (2) is invoked, either explicitly (refs. [4-7]) or implicitly (as in ref. [8]).

$$\langle \Phi | a_{\mu}^{\dagger} a_{\sigma} (H - \mathcal{H}_{\text{HF}}) | \Phi \rangle = 0, \quad (\mu \in F, \sigma \notin F). \quad (6)$$

We label the customary choice for $\mathcal{H}_{\text{HF}}$ as $\mathcal{H}_{\text{HF}}^0$. It is defined by

$$\mathcal{H}_{\text{HF}}^0[\Phi] = \sum_{i=1}^A h(\vec{r}_i, \vec{p}_i, t) \quad (7a)$$

where the matrix elements of $h(\vec{r}, \vec{p}, t)$ are given by

$$h_{\alpha\beta} = t_{\alpha\beta} + \mathcal{V}_{\alpha\beta} \quad (7b)$$

with t equal to the kinetic energy, and the Hartree-Fock potential given by

$$\mathcal{V}_{\alpha\beta} = \sum_{\lambda \in F} \iint d\vec{r}_1 d\vec{r}_2 \psi_{\alpha}^*(\vec{r}_1, t) \psi_{\lambda}^*(\vec{r}_2, t) v(\vec{r}_1 - \vec{r}_2) \{ \psi_{\beta}(\vec{r}_1, t) \psi_{\lambda}(\vec{r}_2, t) - \psi_{\beta}(\vec{r}_2, t) \psi_{\lambda}(\vec{r}_1, t) \} \quad (7c)$$

and where $v(\vec{r}_i - \vec{r}_j)$ is the exact two-body interaction.

We note that a distinct arbitrary, real, space-independent, time-dependent functional of Φ_i , $B_i([\Phi_i(t)]; t)$, may be added to $\mathcal{H}_{\text{HF}}^0$ for each separate solution,[†] Φ_i , without violating conditions (4) and (6). Thus, the most general TDHF Hamiltonian consistent with (6) is

$$\mathcal{H}_{\text{HF}}^B[\Phi_i(t)] = \mathcal{H}_{\text{HF}}^0[\Phi_i(t)] + B_i([\Phi_i(t)]; t). \quad (8)$$

Solutions $\Phi_i^{(1)}, \Phi_i^{(2)}$ corresponding to different choices of $B_i^{(1)}(t), B_i^{(2)}(t)$ in (8), and to the same initial condition, $\Phi_i(t_0)$, are related by a phase factor,

$$\Phi_i^{(2)}(t) = \exp\left\{-\frac{i}{\hbar} \int_{t_0}^t [B_i^{(1)}(t') - B_i^{(2)}(t')] dt'\right\} \Phi_i^{(1)}(t). \quad (9)$$

We thus see that the customary treatment, based on the variational form (3), leaves each separate TDHF solution indeterminate up to its own arbitrary multiplicative phase factor.

We emphasize the qualitative difference between this indeterminacy and the freedom to add a single time-dependent "gauge" function to $\mathcal{H}_{\text{HF}}^0$ (or in the Schrödinger case, to H). The "gauge transformation alters *every* solution by the *same* phase factor, and leaves the *relative* phase of any two solutions *unchanged*. In contrast, the indeterminacy (8) provides the

[†]Each solution is defined by a specified initial determinant, $\Phi_i(\vec{x}; t_0)$, and is labelled by the index i .

freedom to select the phase of each solution independently, and therefore to make arbitrary alterations in the relative phases.

Norm-Violating Variations Uniquely Determine Phase

Note that the restriction, (2), to norm-preserving variations, (3), violates the specification that arbitrary variations are allowed in (1). More specifically, the fact that the solutions must a posteriori have constant norm is not a valid reason for such a restriction. To the contrary, we shall see that the norm-violating variations provide the very variational condition which guarantees a posteriori the solution's constant norm (eq. (12b) below).

Therefore, in contrast with the customary derivation we allow instead of (3) the most general determinantal variation, of the form

$$\delta\Phi = \epsilon_0(t)\Phi + \sum_{\mu} \epsilon_{\mu}^{\sigma}(t) a_{\sigma}^{+} a_{\mu} \Phi. \quad (10)$$

The additional variation,

$$\delta\Phi_0 = \epsilon_0(t)\Phi, \quad (11)$$

included in eq. (10) yields two conditions in addition to (4). The first is the "phase condition",

$$\langle\Phi|H - i\hbar\partial/\partial t|\Phi\rangle = 0, \quad (12a)$$

which is implied immediately by the variation, $\langle\delta\Phi_0|$. The second is the constant norm condition,

$$\frac{d}{dt}\langle\Phi|\Phi\rangle = 0, \quad (12b)$$

which is obtained by taking the difference of (12a) and the corresponding condition from the partially time-integrated variation, $|\delta\Phi_0\rangle$.

We call (12a) the "phase condition" because it uniquely determines the function $B_i(t)$ of (8), (9) as follows

$$B_i(t) = \langle\Phi_i|H - \mathcal{H}_{HF}^0|\Phi_i\rangle = -\frac{1}{2} \sum_{\mu\nu \in F} \bar{v}_{\mu\nu, \mu\nu}. \quad (13)$$

(Substitute eq. (5) for ϕ_1 , with $\mathcal{H}_{HF}$ given by $\mathcal{H}_{HF}^{B_1}$ of eq. (8), into eq. (12a).) We refer to (13) as the "Constant- $\langle \mathcal{H} \rangle$ " value of $B_1(t)$, because it implies for the expectation value of $\mathcal{H}_{HF}^{B_1}$, the result

$$\langle \phi_1 | \mathcal{H}_{HF}^{B_1} | \phi_1 \rangle = \langle \phi_1 | H | \phi_1 \rangle = E_1, \quad (14)$$

which value remains constant under the TDHF evolution in time [1]. Thus the variational principle (1) uniquely determines the TDHF equation of motion when the most general determinantal variations, including norm-violating variations, are exploited. The resulting unique equation of motion determines its TDHF solutions uniquely, including their phase factors.

Thus we find the alleged "arbitrariness" of the phases of TDHF solutions to be a false inference which arises not from the restriction to determinantal-preserving variations, but instead from the unjustifiable restriction to norm-preserving variations. In fact, the variational principle (1) implies a unique solution, including a unique phase, for each initial condition.

Schrödinger Phase Indeterminacy Parallels TDHF

Also, if this norm-conserving restriction were invoked in the derivation of the exact Schrödinger equation, the result would be a phase-indeterminate Schrödinger theory. For variations $\delta\Psi$ satisfying

$$\langle \delta\Psi | \Psi \rangle = 0 \quad (15)$$

the variational principle, eq. (1), yields

$$\langle \delta\Psi | H - i\hbar \partial/\partial t | \Psi \rangle = 0 \quad (16)$$

which under the restriction (15) implies only the equation,

$$(H - i\hbar \partial/\partial t) | \Psi_1(t) \rangle = B_1(t) | \Psi_1(t) \rangle, \quad (17)$$

where $B_1(t)$ may be an arbitrary real function, chosen separately for each solution Ψ_1 . (Cf., remarks following eq. (9).) For any solution of (17) clearly honors (16) if $\langle \delta\Psi |$ is restricted by (15).

Advantages of Constant- $\langle \mathcal{H} \rangle$; Minimal Error

Besides the fact that it is the unique implication of the variational principle, several practical advantages accrue to the Constant- $\langle \mathcal{H} \rangle$ version of the TDHF theory.

First, the norm of the error

$$\chi_{B_i}(t) = (H - i\hbar\partial/\partial t)\phi_i^{B_i}(t), \quad (18)$$

of the TDHF solution, ϕ_i , is minimal when $\phi_i^{B_i}(t)$ solves the Constant- $\langle \mathcal{H} \rangle$ problem defined by (8) and (13). Substitution of eq. (5) into (18) gives:

$$\begin{aligned} \langle \chi_{B_i} | \chi_{B_i} \rangle &= \langle \phi_i^{B_i} | (H - \mathcal{H}_{HF}^0 - B)^2 | \phi_i^{B_i} \rangle \\ &= (\langle \phi_i^{B_i} | H - \mathcal{H}_{HF}^0 | \phi_i^{B_i} \rangle - B_i)^2 + \frac{1}{4} \sum_{\substack{\mu\nu \in F \\ \mu\nu \notin F}} \bar{v}_{\mu\nu, \sigma\tau}^2, \end{aligned} \quad (19)$$

where the last equality is obtained by inserting a complete set of np-nh determinants into the first line, and invoking (6) for the one-particle, one-hole matrix elements. Clearly the norm (19) is minimal when B_i satisfies the Constant- $\langle \mathcal{H} \rangle$ condition, (13).

Constant- $\langle \mathcal{H} \rangle$ Reduces to Exact Determinantal Result

Secondly, Constant- $\langle \mathcal{H} \rangle$ TDHF solutions are identical with determinantal exact Schrödinger eigensolutions. Consider χ_0 , which is a determinantal ground state eigenfunction of the exact Hamiltonian, H ,

$$H\chi_0 = E_0\chi_0. \quad (20)$$

Then the exact time-dependent solution, Ψ , which reduces to χ_0 at $t = t_0$ is

$$\Psi(\vec{x}, t) = \{\exp -iE_0(t-t_0)/\hbar\} \chi_0. \quad (21)$$

Since the ground state energy is stationary with respect to *arbitrary* variations of the wave function, χ_0 must be the least energy solution also of the stationary Hartree-Fock problem, and therefore an eigenfunction of the Hartree-Fock Hamiltonian; thus,

$$\mathcal{H}_{HF}^0 \chi_0 = \epsilon_0 \chi_0. \quad (22)$$

The time-evolution of χ_0 according to the TDHF equation, (4), (7), (8), is

$$\Phi(t) = \exp\left\{-\frac{i}{\hbar}[\epsilon_0(t-t_0) + \int_{t_0}^t B_1(t')dt']\right\}\chi_0 \quad (23)$$

and is identical with the exact Schrödinger wave function $\Psi(t)$ only if $B_1(t)$ satisfies the Constant- $\langle \mathcal{H} \rangle$ condition, eq. (13). A non-trivial examples of just this situation is given in ref. [1].

Overlap Maximized for Short Time Intervals

Thirdly, Constant- $\langle \mathcal{H} \rangle$ TDHF maximizes, to leading order $(\Delta t)^2$, the overlap with exact Schrödinger solutions evolved from initial determinants. Such an exact solution is given by

$$\Psi(t_0+\Delta t) = \Phi(t_0) + \frac{\Delta t}{i\hbar} H \Phi(t_0) + O[(\Delta t)^2]. \quad (24)$$

To the same order in Δt , the corresponding TDHF solution is

$$\Phi(t_0+\Delta t) = \Phi(t_0) + \frac{\Delta t}{i\hbar}[\mathcal{H}_{HF}^0(t_0) + B_1(t_0)] \Phi(t_0) + O[(\Delta t)^2]. \quad (25)$$

Hence,

$$(\Phi(t_0+\Delta t), \Psi(t_0+\Delta t)) = 1 + \frac{\Delta t}{i\hbar} \langle \Phi(t_0) | H - \mathcal{H}_{HF}^0 - B_1 | \Phi(t_0) \rangle + O[(\Delta t)^2]. \quad (24)$$

Then for the Constant- $\langle \mathcal{H} \rangle$ B_1 -value, (13), the second term on the r.h.s. vanishes, leading to the asserted maximal (unit) overlap, to order $(\Delta t)^2$.

Practical Importance of Phase in Time-Averaged TD- $\mathcal{H}$ -HF Theory

The indeterminacy of the phase in TDHF has, until recently, been of no consequence *in practice* due to the fact that TDHF applications heretofore have involved only a single TDHF solution and have assumed all the physical content of the description to reside in the corresponding expectation values of physical operators. Since there arise only expectation values for one wave function, its time-dependent phase can affect no physical prediction of the theory. In this limited context of such "closed" applications of TDHF, the claim that the time-dependent phase of the TDHF solution is arbitrary is valid at the practical level, although erroneous and misleading in principle.

A recent generalization of the TDHF reaction theory has been advanced [9] (the "TD- $\mathcal{S}$ -HF" method) which calculates reaction amplitudes, using a separate TDHF solution for each ingoing and outgoing reaction channel, as time-averages of overlap amplitudes analogous to the Schrödinger S-matrix elements:

$$\bar{\mathcal{S}}_{fi} = (T_2 - T_1)^{-1} \int_{T_1}^{T_2} \langle \phi_f^{(-)}(\vec{x}, t') | \phi_i^{(+)}(\vec{x}, t') \rangle dt'. \quad (27)$$

Here the numerical value of every amplitude $\bar{\mathcal{S}}_{fi}$ will clearly depend upon the relative time-dependent phase of the states ϕ_f and ϕ_i , just as the analogous S-matrix of Schrödinger theory depends upon the relative phases of the exact solutions.

Determinantal Phase Condition and Indeterminacy of Single Particle TDHF States

The customary variational conditions (4) are satisfied by any determinant built out of solutions to any set of single-particle TDHF equations of the form,

$$(\hbar - i\hbar \partial/\partial t)\psi_\mu = \sum_{\nu \in F} \psi_\nu b_{\nu\mu}(t) \quad (\mu \in F), \quad (28)$$

where $\hbar$ is defined in eq. (7) and $b_{\nu\mu}(t)$ is an arbitrary Hermitean matrix.

(Practical calculations customarily assume that the matrix $b_{\nu\mu}$ vanishes.)

We now show that the total effect on the determinantal solution of altering the arbitrary matrix $b_{\nu\mu}$ is solely to induce an overall phase change.

Consider a time-dependent unitary transformation, $U(t)$, among the filled TDHF single-particle states, ψ_μ , which satisfy (28) for $b_{\nu\mu} = 0$,

$$(\hbar - i\hbar \partial/\partial t)\psi_\mu = 0. \quad (29)$$

Then the transformed states,

$$\tilde{\psi}_\mu = \sum_{\nu} U_{\mu\nu} \psi_\nu \quad (30)$$

satisfy

$$(\hbar - i\hbar \partial/\partial t)\tilde{\psi}_\mu = -i\hbar \sum_{\nu} \tilde{\psi}_\nu (\dot{U}U^\dagger)_{\mu\nu}. \quad (31)$$

Comparison with eq. (28) shows that such a transformation is equivalent to the following choice for $b_{\nu\mu}$:

$$b_{\nu\mu} = -i\hbar(\dot{U}U^\dagger)_{\nu\mu}. \quad (32)$$

Since U is unitary, $b_{\nu\mu}$ is Hermitean. Thus, for each non-zero Hermitean matrix b , eq. (32) specifies the unitary transformation U which brings eq. (28) to the form (29). Furthermore, the determinants $\tilde{\Phi}$ and Φ built from the corresponding single-particle states $\tilde{\psi}_\mu$ and ψ_μ are related by a phase factor

$$\tilde{\Phi} = (\det U)\Phi, \quad (33)$$

since $\det U$ is of unit magnitude (U is unitary).

The phase factor can be determined explicitly by use of the identity

$$\det A(t) = [\det A(0)] \exp \int_0^t \text{Tr}(\dot{A}A^{-1})dt' \quad (34)$$

which holds for any non-singular differentiable matrix.[†] Then for $A = U$,

(32) and (34) imply

$$\det U(t) = \det U(0) e^{+\frac{i}{\hbar} \int_0^t \text{Tr} b(t')dt'} \quad (35)$$

Equation (35) relates the Constant- $\langle \mathcal{H} \rangle$ phase condition (13) to the (trace of) matrix $b_{\nu\mu}$: In eq. (9) select $B_i^{(2)} = 0$ and choose $B_i^{(1)} = B$ to satisfy the Constant- $\langle \mathcal{H} \rangle$ condition (13). Then by identifying the phase factor in (9) with that in (33), one obtains the equality

$$\text{Tr} b = -\tilde{B} \quad (36)$$

as the specification of the matrix b in (28) which honors the Constant- $\langle \mathcal{H} \rangle$ phase condition.

This single condition (36) on an otherwise arbitrary $A \times A$ Hermitean matrix, $b_{\nu\mu}$, constitutes a very weak restriction. Thus, although the variational principle implies a unique Constant- $\langle \mathcal{H} \rangle$ TDHF equation and a unique A -particle determinantal solution to that equation, much freedom remains in the choice of eqs. (28) for the single-particle state, ψ_μ . However, since this freedom allows

[†]Identity (34) is the solution to the first order differential equation obtained directly from the definition of the derivative of a determinant in terms of its cofactor expansion.

no alteration whatsoever of the determinantal wave function it can have no physical significance.

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