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The many-body wave function of a system of interacting particles confined by a time-dependent harmonic potential and perturbed by a time-dependent spatially homogeneous electric field is derived via the Feynman path-integral method. The wave function is comprised of a phase factor times the solution to the unperturbed time-dependent Schrödinger equation with the latter being translated by a time-dependent value that satisfies the classical driven equation of motion. The wave function reduces to that of the Harmonic Potential Theorem wave function for the case of the time-independent harmonic confining potential. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4820245]

I. INTRODUCTION

The harmonic oscillator with constant mass and frequency, whose analytical solution was determined by Schrödinger,[1] has played a significant role in physics. Since then, there has been considerable literature on the harmonic oscillator with time-dependent (TD) frequencies, TD masses, or both, or with TD perturbative potentials (see, e.g., Refs. 2–14). These are exactly solvable models that have applications in quantum chemistry, quantum optics, plasma physics, and quantum field theory. As an example,[8] the Hamiltonian of a charged particle in the presence of a particular axially symmetric TD electromagnetic field can be recast as that of a harmonic oscillator with TD frequencies. The case of a TD harmonic oscillator with a time-independent inverse harmonic potential has also been considered.[15] These are all single particle models with exact solutions and they play a significant role in physics.

The extension of a TD harmonic oscillator to a particle interaction that is inverse harmonic and many-dimensional has been studied.[16] For the case of two (Coulombically interacting) electrons confined by a time-independent harmonic well – the Hooke's atom – closed-form analytical solutions[17–20] for specific ground and excited states can be derived. Hence, how the two electrons are correlated can be studied. Other examples, of harmonically confined two-electron systems are the Hooke's molecule and molecular ions,[20–22] the Hooke's atom with an additional contribution linear in the relative coordinate,[23] and the Hooke's atom in a magnetostatic field.[24] As a consequence, properties such as the structure of the corresponding nonlocal (dynamic) Fermi-Coulomb hole charge distribution as a function of electron position, and the correlation contribution to the kinetic energy, for both a ground and an excited state have been studied.[20,21,25–27] The Hooke's atom also describes spherical nuclear models[28] and quantum dots.[27–31] The Hooke's species[20–22] lead to a corollary to the Hohenberg-Kohn[32] theorem of time-independent density functional theory. In turn, the Hooke's species in which the positions of the nuclei are TD,[20–22] leads to a corollary to the Runge-Gross theorem[33] of TD density functional theory.

In this paper, we derive the many-body wave function of a system of N particles interacting with a force dependent on the interparticle distance that are confined by a harmonic potential whose frequencies are time-dependent and which are perturbed by a TD spatially homogeneous electric field. Thus, we extend the work on the harmonic oscillator with TD frequencies and TD perturbations to the many-body domain. A "first principles" derivation employing the Feynman path-integral method[34,35] is used to obtain the wave function. The principal attribute of the method is that the many-body wave function is naturally revealed as a result of the derivation. No assumptions as to its structure need be made. The wave function is comprised of a TD phase factor times the solution of the interacting unperturbed TD Schrödinger equation, the latter being translated by a TD value which in turn is the solution of the classical driven harmonic oscillator equation. Thus, the evolution of observables such as the density that are represented by non-differential Hermitian operators, is that of the unperturbed property but moving rigidly. As a consequence of the phase factor, this is not the case for observables that are represented by differential operators such as the physical current density.

For the case when the harmonic frequencies are time-independent, the wave function derived reduces to that of the Harmonic Potential Theorem (HPT) wave function[36] which has a similar structure. (For other proofs of the theorem, see Refs. 37 and 38.) The HPT has played an important role in the understanding of the evolution of states within TD Schrödinger theory, the description of Schrödinger theory in terms of "classical" fields and quantal sources via the "quantal Newtonian" second law,[20,39] and in its manifestations as TD density functional[33,40–42] and quantal density functional[20,43–46] theories, and in quantum fluid dynamics.[20,47,48] The HPT wave function constitutes a special case of the wave function derived in this work. This means

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that the HPT wave function can also be derived via the Feynman path-integral method. Finally, we note that the properties of the HPT such as those of the time evolution of the density as noted above, can be shown to be intrinsic to the recently developed quantum continuum mechanics.[49–53] In this formalism, the collective variables of the density and paramagnetic current density are obtained directly. This contrasts with the "quantal Newtonian" first and second laws that are in terms of fields acting on the individual particles.

We next derive the wave function.

II. DERIVATION OF WAVE FUNCTION

Consider a system of N particles with arbitrary interaction $u(\mathbf{r}_i - \mathbf{r}_j)$ in an external TD harmonic potential $v(\mathbf{r}) = \frac{1}{2}\mathbf{r}\cdot\mathbf{K}(t)\cdot\mathbf{r}$, with $\mathbf{K}(t)$ the spring constant. A spatially homogeneous TD driving electric field $\mathbf{E}(t) = -\mathbf{f}(t)/e$ is turned on at time $t = 0$. We assume $f(t) = 0$ for $t \le 0$. For example, the field $\mathbf{E}$ could correspond to the electric field of a high intensity laser pulse employed in the study of atoms and molecules. The Hamiltonian of the system is

$$\hat{H}(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N; t) = \hat{H}_0(t) - \mathbf{f}(t)\cdot\sum_j \mathbf{r}_j, \tag{1}$$

where the unperturbed component is

$$\hat{H}_0(\mathbf{r}_1, \ldots, \mathbf{r}_N; t) = \sum_i \frac{1}{2m}\hat{\mathbf{p}}_i^2 + \sum_i \frac{1}{2}\mathbf{r}_i\cdot\mathbf{K}(t)\cdot\mathbf{r}_i + \sum_{i\neq j} u(\mathbf{r}_i - \mathbf{r}_j). \tag{2}$$

We restrict our discussion to the case such that $\mathbf{K}$ is diagonal: $\mathbf{K} = \mathrm{diag}(k_x, k_y, k_z)$ so that

$$\frac{1}{2}\mathbf{r}_i\cdot\mathbf{K}(t)\cdot\mathbf{r}_i = \frac{1}{2}m\left[\omega_x(t)x_i^2 + \omega_y(t)y_i^2 + \omega_z(t)z_i^2\right]. \tag{3}$$

The wave function $\Psi(\mathbf{r}_1, \ldots, \mathbf{r}_N; t)$ satisfies the TD Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\Psi(\mathbf{r}_1, \ldots, \mathbf{r}_N; t) = \hat{H}(\mathbf{r}_1, \ldots, \mathbf{r}_N; t)\Psi(\mathbf{r}_1, \ldots, \mathbf{r}_N; t). \tag{4}$$

We next derive the wave function via the Feynman path-integral method.

Define the center of mass and relative coordinates and momentum[29,54,55] as

$$\mathbf{R}^{(1)} = \mathbf{R} = \frac{1}{N}\sum_i \mathbf{r}_i, \quad \hat{\mathbf{P}}^{(1)} = \sum_i \hat{\mathbf{p}}_i \tag{5}$$

and

$$\begin{aligned} X^{(2)} &= x_1 - x_2, \\ X^{(3)} &= x_1 + x_2 - 2x_3, \ldots, \\ X^{(N)} &= x_1 + x_2 + \ldots + x_{N-1} - (N-1)x_N, \end{aligned} \tag{6}$$

and similarly for $Y^{(2)}, \ldots, Y^{(N)}$, $Z^{(2)}, \ldots, Z^{(N)}$, and $P^{(2)}, \ldots, P^{(N)}$. The Hamiltonian of Eq. (1) can be rewritten as (see the Appendix for the derivation)

$$\hat{H} = \hat{H}_{\mathrm{cm}}(t) + \hat{H}_{\mathrm{rel}}(t), \tag{7}$$

where

$$\hat{H}_{\mathrm{cm}}(t) = \frac{\hat{\mathbf{P}}^2}{2M} + \frac{M}{2}\left[\left(\omega_X^2(t)X^2 + \omega_Y^2(t)Y^2 + \omega_Z^2(t)Z^2\right)\right] - \mathbf{F}(t)\cdot\mathbf{R}, \tag{8}$$

with $M = Nm$ and $\mathbf{F}(t) = N\mathbf{f}(t)$. Here, $\hat{H}_{\mathrm{cm}}(t)$ is the Hamiltonian describing the motion of the center of mass only. Note that the frequencies of the harmonic potential trapping the mass center are the same as those of that trapping each individual particles, i.e., $\omega_X(t) = \omega_x(t)$, $\omega_Y(t) = \omega_y(t)$, and $\omega_Z(t) = \omega_z(t)$. $\hat{H}_{\mathrm{rel}}(t)$ is a function of only the relative coordinates and contains the effects of the interaction (see the Appendix). It can be readily shown that $[\hat{H}_{\mathrm{cm}}(t), \hat{H}_{\mathrm{rel}}(t)] = 0$. Thus, the center-of-mass motion and the relative motion are separable. Therefore, the total wave function of the Hamiltonian is the product of the wave functions of the center-of-mass motion and relative motion:

$$\Psi(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N; t) = \Phi(\mathbf{R}, t)\varphi_{\mathrm{rel}}(\mathbf{R}^{(2)}, \ldots, \mathbf{R}^{(N)}; t). \tag{9}$$

The relative motion wave function $\varphi_{\mathrm{rel}}(\mathbf{R}^{(2)}, \ldots, \mathbf{R}^{(N)}; t)$ satisfies the Schrödinger equation

$$i\hbar\frac{\partial\varphi_{\mathrm{rel}}(\mathbf{R}^{(2)}, \ldots, \mathbf{R}^{(N)}; t)}{\partial t} = \hat{H}_{\mathrm{rel}}(t)\varphi_{\mathrm{rel}}(\mathbf{R}^{(2)}, \ldots, \mathbf{R}^{(N)}; t), \tag{10}$$

and the center-of-mass motion wave function $\Phi(\mathbf{R}, t)$ satisfies the Schrödinger equation

$$i\hbar\frac{\partial\Phi(\mathbf{R}, t)}{\partial t} = \hat{H}_{\mathrm{cm}}(t)\Phi(\mathbf{R}, t), \tag{11}$$

with the initial condition

$$\Phi(\mathbf{R}, 0) = \Phi_n^0(\mathbf{R}) = N H_{n_1}(\alpha_X X)H_{n_2}(\alpha_Y Y)H_{n_3}(\alpha_Z Z) \times\exp\left[-\frac{1}{2}\left(\alpha_X^2X^2 + \alpha_Y^2Y^2 + \alpha_Z^2Z^2\right)\right] \tag{12}$$

being the $n = \{n_1, n_2, n_3\}$th state of a three-dimensional harmonic oscillator with H_n the corresponding Hermite polynomial, and where $\alpha_i = (\frac{M\omega_i(0)}{\hbar})^{\frac{1}{2}}$, $i = X, Y, Z$, and $N = N_1N_2N_3$, with $N_i = [\frac{\alpha_i}{\sqrt{\pi}2^{n_i}n_i!}]^{\frac{1}{2}}$, $i = 1, 2, 3$, the normalization constant.

It is clear that the Hamiltonian $\hat{H}_{\mathrm{cm}}(t)$ can be separated into its X, Y, Z components with each component being independent of the others. The X-component of the wave function is

$$\Phi(X, t) = \int_{-\infty}^{+\infty} K_X(X, t; X_0, 0)\Phi(X_0, 0)dX_0, \tag{13}$$

where the propagator for the X-component is[56]

$$K_X(X_b, t_b; X_a, t_a) = Ae^{\frac{i}{\hbar}[B+C+D+E]}, \tag{14}$$

and where

$$A = \left[\frac{M\omega_X(0)}{2\pi i\hbar\rho_X(t_b)\rho_X(t_a)\sin\phi_X(t_b, t_a)}\right]^{\frac{1}{2}}, \tag{15}$$

$$B = \frac{M}{2}\left[\frac{\dot{\rho_X}(t_b)X_b^2}{\rho_X(t_b)} - \frac{\dot{\rho_X}(t_a)X_a^2}{\rho_X(t_a)}\right], \tag{16}$$

$$C = \frac{M\omega_X(0)}{2\sin\phi_X(t_b,t_a)}\left[\left(\frac{X_b^2}{\rho_X(t_b)^2}+\frac{X_a^2}{\rho_X(t_a)^2}\right)\cos\phi_X(t_b,t_a) - 2\frac{X_bX_a}{\rho_X(t_b)\rho_X(t_a)}\right], \quad (17)$$

$$D = \frac{1}{\sin\phi_X(t_b,t_a)}\left[\frac{X_b\int_{t_a}^{t_b}dt\,F_X(t)\rho_X(t)\sin\phi_X(t,t_a)}{\rho_X(t_b)} + \frac{X_a\int_{t_a}^{t_b}dt\,F_X(t)\rho_X(t)\sin\phi_X(t_b,t)}{\rho_X(t_a)}\right], \quad (18)$$

$$E = -\frac{2\int_{t_a}^{t_b}\int_{t_a}^{t}F_X(t)F_X(\tau)\sin\phi_X(t_b,t)\sin\phi_X(\tau,t_a)d\tau dt}{M\omega_X(0)\sin\phi_X(t_b,t_a)}, \quad (19)$$

where ρ_X satisfies the auxiliary equation

$$\ddot{\rho}_X + \omega_X^2(t)\rho_X = \omega_X(0)^2/\rho_X^3, \quad (20)$$

and

$$\phi_X(\tau,t) = \mu_X(\tau) - \mu_X(t), \quad (21)$$

with

$$\mu_X(t) = \omega_X(0)\int_0^t\frac{dt}{\rho_X^2(t)}. \quad (22)$$

Substituting the propagator into Eq. (13), one obtains the X-component wave function to be[57,58]

$$\Phi(X,t) = e^{i\gamma_{n_1}^X(t)}\phi_{n_1}(X,t) = e^{i\gamma_{n_1}^X(t)}\left(\frac{1}{2^{n_1}n_1!}\sqrt{\frac{M\omega_X(0)}{\pi\hbar\rho_X^2}}\right)^{\frac{1}{2}} \times \exp\left(-\frac{P_X^2}{2}+\frac{iR_X}{2}\right)\times H_{n_1}(P_X), \quad (23)$$

where

$$P_X = P_X^0 + V_X(t) = \sqrt{\frac{M\omega_x(0)}{\hbar}}\frac{X}{\rho_X(t)} + V_X(t), \quad (24)$$

with

$$V_X(t) = \frac{1}{\sqrt{M\omega_X(0)\hbar}}\int_0^t\rho_X(\tau)F_X(\tau)\sin\phi_X(\tau,t)d\tau. \quad (25)$$

$$R_X = R_X^0 + \sqrt{\frac{M\omega_X(0)}{\hbar}}\frac{2XU_X(t)}{\rho_X(t)}, \quad (26)$$

with

$$R_X^0 = \frac{M}{\hbar}X^2\left(\frac{\dot{\rho_X}}{\rho_X}\right) \quad (27)$$

and

$$U_X(t) = \frac{1}{\sqrt{M\omega_X(0)\hbar}}\int_0^t\rho_X(\tau)F_X(\tau)\cos\phi_X(\tau,t)d\tau, \quad (28)$$

and the X-component phase

$$\gamma_{n_1}^X(t) = -\left(n_1+\frac{1}{2}\right)\mu_X(t) - S_X(t), \quad (29)$$

with

$$S_X(t) = \int_0^t\frac{\omega_X(0)\left[U_X{}^2(\tau) - V_X{}^2(\tau)\right]}{2\rho_X^2(\tau)}d\tau. \quad (30)$$

Note that from Eqs. (23) and (24) we can see that under the influence of an external electrical field, the wave function is shifted by

$$\Lambda_X(t) = -\frac{\rho_X(t)}{M\omega_X(0)}\int_0^t\rho_X(\tau)F_X(\tau)\sin\phi_X(\tau,t)d\tau. \quad (31)$$

Using Eq. (31), one can verify that $\Lambda_X(t)$ satisfies the X-component TD equation of motion

$$\frac{d^2\Lambda_X(t)}{dt^2} + \omega_X(t)^2\Lambda_X(t) - F_X(t)/M = 0. \quad (32)$$

Moreover, making use of Eqs. (23)– (30), and the equation

$$iX\sqrt{\frac{M\omega_X(0)}{\hbar}}\frac{U_X}{\rho_X} = \frac{i}{\hbar}XM\left[\dot{\Lambda}_X(t) - \frac{\dot{\rho}_X}{\rho_X}\Lambda_X(t)\right] = \frac{i}{\hbar}X\left[P_{\Lambda_X} - M\frac{\dot{\rho}_X}{\rho_X}\Lambda_X(t)\right], \quad (33)$$

the final wave function can be rewritten as

$$\Phi(X,t) = e^{-\frac{i}{\hbar}\{\tilde{S}[\Lambda_X(t)] - P_{\Lambda_X}X\}}\Phi_{n_1}^0(X-\Lambda_X(t),t), \quad (34)$$

where

$$\tilde{S}[\Lambda_X(t)] = \int_0^t d\tau\left[\frac{M\dot{\Lambda_X}{}^2(\tau)}{2} - \frac{M\omega_X^2(\tau)\Lambda_X^2(\tau)}{2}\right] \quad (35)$$

is the classical action without the coupling to the external electric field term and

$$\Phi_{n_1}^0(X-\Lambda_X(t),t) = \left(\frac{1}{2^{n_1}n_1!}\sqrt{\frac{M\omega_X(0)}{\pi\hbar\rho_X^2}}\right)^{\frac{1}{2}} \times \exp\left\{-i\left(n_1+\frac{1}{2}\right)\mu_X(t) + \frac{i}{2}R_X^0 - \frac{\left(P_X^0\right)^2}{2}\right\}H_{n_1}\left(P_X^0\right) \quad (36)$$

is the time-dependent wave function in the absence of the external perturbing electric field. Generalizing Eq. (34) to the 3D case, we obtain

$$\Phi(\mathbf{R},t) = e^{-\frac{i}{\hbar}\{\tilde{S}[\mathbf{\Lambda}(t)] - \mathbf{P}_{\mathbf{\Lambda}(t)}\cdot\mathbf{R}\}}\Phi_{n_1,n_2,n_3}^0(\mathbf{R}-\mathbf{\Lambda}(t),t). \quad (37)$$

Employing Eq. (9), the total wave function is then

$$\Psi(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N;t) = e^{-\frac{i}{\hbar}\{\tilde{S}[\mathbf{\Lambda}(t)] - \mathbf{P}_{\mathbf{\Lambda}(t)}\cdot\mathbf{R}\}}\varphi_{\rm rel}(\mathbf{R}^{(2)},\ldots,\mathbf{R}^{(N)};t) \times \Phi_{n_1,n_2,n_3}^0(\mathbf{R}-\mathbf{\Lambda}(t),t). \quad (38)$$

The wave function is thus comprised of the product of a phase factor, the relative coordinate function, and the unperturbed wave function translated by a value that satisfies the driven classical equation of motion. In the case when the frequencies are time-independent, the wave function reduces to that of the HPT wave function.[36]

III. CONCLUDING REMARKS

Via the Feynman path-integral method, we have derived the many-body wave function for particles such as electrons confined by a TD harmonic well and in the presence of a TD spatially homogeneous electric field. This extends prior work on such systems to the many-body realm. The wave function possesses the characteristic that the time evolution of observables represented by non-differential hermitian operators is that of the property translated rigidly by a TD value that satisfies the classical equation of motion. Thus, we envisage it being of value in theories such as TD density functional theory in which the density is a basic variable. (A basic variable of quantum mechanics is a gauge invariant property knowledge of which uniquely determines the Hamiltonian.[32,59]) Approximate action functionals of the density can then be tested against the exact structure. It is noted that as a consequence of the phase factor, the above characteristic is not exhibited by properties such as the paramagnetic and thereby the physical current density. Nonetheless, the exact evolution of such properties is also known. This is of value in TD current density functional theories in which the basic variables are the density and physical current density.[60]

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APPENDIX: THE RELATIVE COORDINATE HAMILTONIAN

In this appendix we reduce the Hamiltonian $\hat{H}$ of Eqs. (1)–(3) to its center of mass $\hat{H}_{\rm cm}$ and relative $\hat{H}_{\rm rel}$ components, thereby providing an expression for the latter. With the center of mass and relative coordinates defined by Eqs. (5) and (6), the transformed coordinates may be expressed as

$$\begin{pmatrix} \mathbf{R^{(1)}} \\ \mathbf{R}^{(2)} \\ \mathbf{R}^{(3)} \\ \vdots \\ \mathbf{R}^{(N)} \end{pmatrix} = T \begin{pmatrix} \mathbf{r_1} \\ \mathbf{r}_2 \\ \mathbf{r}_3 \\ \vdots \\ \mathbf{r}_N \end{pmatrix}, \tag{A1}$$

where the transformation matrix T is

$$\begin{pmatrix} \frac{1}{N} & \frac{1}{N} & \frac{1}{N} & \cdots & \frac{1}{N} \\ 1 & -1 & 0 & \ldots & 0 \\ 1 & 1 & -2 & \ldots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 1 & 1 & 1 & \ldots & -(N-1) \end{pmatrix}. \tag{A2}$$

Since

$$\nabla_{\mathbf{r}_i} = \frac{\partial \mathbf{R}^{(1)}}{\partial \mathbf{r}_i} \nabla_{\mathbf{R}^{(1)}} + \frac{\partial \mathbf{R}^{(2)}}{\partial \mathbf{r}_i} \nabla_{\mathbf{R}^{(2)}} + \ldots + \frac{\partial \mathbf{R}^{(N)}}{\partial \mathbf{r}_i} \nabla_{\mathbf{R}^{(N)}}, \tag{A3}$$

we have

$$\begin{pmatrix} \nabla_{\mathbf{r}_1} \\ \nabla_{\mathbf{r}_2} \\ \nabla_{\mathbf{r}_3} \\ \vdots \\ \nabla_{\mathbf{r}_N} \end{pmatrix} = \tilde{T} \begin{pmatrix} \nabla_{\mathbf{R}^{(1)}} \\ \nabla_{\mathbf{R}^{(2)}} \\ \nabla_{\mathbf{R}^{(3)}} \\ \vdots \\ \nabla_{\mathbf{R}^{(N)}} \end{pmatrix}, \tag{A4}$$

where $\tilde{T}$ is the transpose of T. Thus,

$$\frac{1}{2m} \sum_i \hat{\mathbf{p}}_i^2 = -\frac{\hbar^2}{2m} \sum_i \nabla_{\mathbf{r}_i}^2 = -\frac{\hbar^2}{2m} (\nabla_{\mathbf{r}_1} \nabla_{\mathbf{r}_2} \ldots \nabla_{\mathbf{r}_N}) \begin{pmatrix} \nabla_{\mathbf{r}_1} \\ \nabla_{\mathbf{r}_2} \\ \vdots \\ \nabla_{\mathbf{r}_N} \end{pmatrix}$$

$$= -\frac{\hbar^2}{2m} (\nabla_{\mathbf{R}^{(1)}} \nabla_{\mathbf{R}^{(2)}} \ldots \nabla_{\mathbf{R}^{(N)}}) T \tilde{T} \begin{pmatrix} \nabla_{\mathbf{R}^{(1)}} \\ \nabla_{\mathbf{R}^{(2)}} \\ \vdots \\ \nabla_{\mathbf{R}^{(N)}} \end{pmatrix}. \tag{A5}$$

The product of the matrices

$$T\tilde{T} = \begin{pmatrix} \frac{1}{N} & 0 & 0 & 0 & \ldots & 0 \\ 0 & 2 & 0 & 0 & \ldots & 0 \\ 0 & 0 & 6 & 0 & \ldots & 0 \\ 0 & 0 & 0 & 12 & \ldots & 0 \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & 0 & 0 & \ldots & N(N-1) \end{pmatrix} \tag{A6}$$

is a diagonal matrix. This means that for each transformed coordinate $\mathbf{R}^{(i)}$ there corresponds an effective mass m_i. Thus, $m_1 = Nm = M$, $m_2 = m/2$, $m_3 = m/6$, …, $m_N = m/N(N-1)$. Thus, the kinetic energy operator is

$$\hat{T} = -\frac{\hbar^2}{2m} \sum_i \nabla_{\mathbf{r}_i}^2 = -\sum_{i=1}^{N} \frac{\hbar^2}{2m_i} \nabla_{\mathbf{R}^{(i)}}^2 = \frac{\hat{\mathbf{P}}^2}{2M} - \sum_{i=2}^{N} \frac{\hbar^2}{2m_i} \nabla_{\mathbf{R}^{(i)}}^2. \tag{A7}$$

From Eq. (A1)

$$\begin{pmatrix} \mathbf{r_1} \\ \mathbf{r}_2 \\ \vdots \\ \mathbf{r}_N \end{pmatrix} = T^{-1} \begin{pmatrix} \mathbf{R^{(1)}} \\ \mathbf{R}^{(2)} \\ \vdots \\ \mathbf{R}^{(N)} \end{pmatrix} \quad \text{or} \quad \mathbf{r}_i = \sum_{\ell=1}^{N} (T^{-1})_{i\ell} \mathbf{R}^{(\ell)}, \tag{A8}$$

where the inverse matrix is

$$T^{-1} = \begin{pmatrix} 1 & \frac{1}{2} & \frac{1}{6} & \frac{1}{12} & \cdots & \frac{1}{N(N-1)} \\ 1 & -\frac{1}{2} & \frac{1}{6} & \frac{1}{12} & \cdots & \frac{1}{N(N-1)} \\ 1 & 0 & \frac{1}{3} & \frac{1}{12} & \cdots & \frac{1}{N(N-1)} \\ 1 & 0 & 0 & -\frac{1}{4} & \cdots & \frac{1}{N(N-1)} \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots \\ 1 & 0 & 0 & 0 & \ldots & -\frac{1}{N} \end{pmatrix}. \tag{A9}$$

With the harmonic force constant matrix

$$\mathbf{K}(t) = \begin{pmatrix} \omega_x^2(t) & 0 & 0 \\ 0 & \omega_y^2(t) & 0 \\ 0 & 0 & \omega_z^2(t) \end{pmatrix}, \tag{A10}$$

the external potential energy operator which is

$$\begin{aligned} \hat{V} &= \frac{1}{2} m \sum_{i=1}^{N} \tilde{\mathbf{r}}_i \cdot \mathbf{K}(t) \cdot \mathbf{r}_i \\ &= \frac{1}{2} m \big[\omega_x^2(t)\big(x_1^2 + x_2^2 + \cdots + x_N^2\big) \\ &\quad + \omega_y^2(t)\big(y_1^2 + y_2^2 + \cdots + y_N^2\big) \\ &\quad + \omega_z^2(t)\big(z_1^2 + z_2^2 + \cdots + z_N^2\big)\big], \end{aligned} \tag{A11}$$

may be rewritten employing Eq. (A8) as

$$\begin{aligned} \hat{V} &= \frac{1}{2} m (\mathbf{R}^{(1)}\mathbf{R}^{(2)} \ldots \mathbf{R}^{(N)}) \tilde{T}^{-1} \mathbf{K}(t) T^{-1} \begin{pmatrix} \mathbf{R^{(1)}} \\ \mathbf{R}^{(2)} \\ \vdots \\ \mathbf{R}^{(N)} \end{pmatrix} \\ &= \frac{1}{2} m (\mathbf{R}^{(1)}\mathbf{R}^{(2)} \ldots \mathbf{R}^{(N)}) \begin{pmatrix} N & 0 & 0 & \ldots & 0 \\ 0 & \frac{1}{2} & 0 & \ldots & 0 \\ 0 & 0 & \frac{1}{6} & \ldots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & 0 & \ldots & \frac{1}{N(N-1)} \end{pmatrix} \\ &\quad \times \mathbf{K}(t) \begin{pmatrix} \mathbf{R^{(1)}} \\ \mathbf{R}^{(2)} \\ \vdots \\ \mathbf{R}^{(N)} \end{pmatrix} \\ &= \sum_{i=1}^{N} \frac{1}{2} m_i \tilde{\mathbf{R}}^{(i)} \cdot \mathbf{K}(t) \cdot \mathbf{R}^{(i)} \\ &= \frac{1}{2} N m \big[\omega_X^2(t) R_X^{(1)2} + \omega_Y^2(t) R_Y^{(1)2} + \omega_Z^2(t) R_Z^{(1)2}\big] \\ &\quad + \frac{1}{2}\left(\frac{m}{2}\right)\big[\omega_X^2(t) R_X^{(2)2} + \omega_Y^2(t) R_Y^{(2)2} + \omega_Z^2(t) R_Z^{(2)2}\big] + \cdots. \end{aligned} \tag{A12}$$

Observe that the transformed operators $\hat{T}$ and $\hat{V}$ are of the same form as in the original Hamiltonian, but with effective masses m_i replacing the electron mass m. From Eq. (A8),

$$\begin{aligned} \mathbf{r}_i - \mathbf{r}_j &= \sum_{\ell=1}^{N} [(T^{-1})_{i\ell} - (T^{-1})_{j\ell}] \mathbf{R}^{(\ell)} \\ &= \sum_{\ell=2}^{N} [(T^{-1})_{i\ell} - (T^{-1})_{j\ell}] \mathbf{R}^{(\ell)}, \end{aligned} \tag{A13}$$

since $(T^{-1})_{i1} = 1$. Thus, the transformed electron-interaction operator is

$$\hat{U} = \sum_{i \neq j} u \left(\sum_{\ell=2}^{N} [(T^{-1})_{i\ell} - (T^{-1})_{j\ell}] \mathbf{R}^{(\ell)} \right). \tag{A14}$$

Observe that the transformed operator depends only on the relative coordinates.

The relative coordinates Hamiltonian is then

$$\begin{aligned} \hat{H}_{\rm rel}(t) &= \sum_{i=2}^{N} \left[-\frac{\hbar^2}{2m_i} \nabla_{\mathbf{R}^{(i)}}^2 + \frac{m_i}{2} \tilde{\mathbf{R}}^{(i)} \cdot \mathbf{K}(t) \cdot \mathbf{R}^{(i)} \right] \\ &\quad + \sum_{i \neq j} u \left(\sum_{\ell=2}^{N} [(T^{-1})_{i\ell} - (T^{-1})_{j\ell}] \mathbf{R}^{(\ell)} \right), \end{aligned} \tag{A15}$$

which is a function of time. The center of mass Hamiltonian is given by Eq. (8).

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