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Optimal control of quantum dynamics: a new theoretical approach

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Abstract. A new theoretical formalism for optimal quantum control has been presented. The approach stems from the consideration of describing the time-dependent quantum systems in terms of the real physical observables, namely the probability density $\rho(x, t)$ and the quantum current $j(x, t)$ which is well documented in Bohm's hydrodynamical formulation of quantum mechanics. The approach has been applied for manipulating the vibrational motion of HBr in its ground electronic state under an external electric field.

1. Introduction

Manipulating the outcome of chemical dynamics by properly tailoring an external field has been an active field of research in recent times [1–14]. Problems where the external field is an electromagnetic field have received the most attention [2–14], although other applications may arise as well. Theoretically, there are two basic paradigms for such a method of control: a static control scheme [13, 14] and a dynamic control scheme [2–12]. In the static scheme [13, 14] one uses two or more CW light fields (optical coherence) and the superposition of two or more eigenstates (molecular coherence) to cause interference between different plausible pathways to a final quantum state, and the outcome is controlled by tailoring different parameters of the optical and molecular coherences. Whereas the dynamic scheme [2–12] creates non-stationary states of one's choice, by optimally designing the electric field. This comes under the domain of the optimal control theory [15], a mathematical tool commonly used in engineering. A basic difficulty in attaining the control designs is the computational effort called for in solving the time-dependent Schrödinger equation, often repeatedly in an iterative fashion over an extended spatial region.

In this paper, we introduce a new formulation aimed at reducing the effort required for quantum optimal control (QOC). Our recent work [16, 17] has shown that Bohmian quantum hydrodynamics (BQH) is capable of being much more efficient than the conventional method (e.g. FFT propagation) and this should carry over to the optimal control task. This paper will show how the BQH can be utilized in QOC. The formulation is based on the hydrodynamic description of the quantum mechanics emerging mainly from the work of Bohm [18, 19] where the dynamics is described by two equations, namely the equation of motion for the probability density, $\rho(r, t)$ and that for the quantum current, $j(r, t)$ which are defined as $\rho(r, t) = \Psi^*(r, t)\Psi(r, t)$ and $j(r, t) = \frac{1}{2}\frac{\hbar}{m}\,\mathrm{Im}[\Psi^*\nabla\Psi - \Psi\nabla\Psi^*]$, where Ψ is the complex wavefunction in the time-dependent Schrödinger equation (TDSE) and Im refers to the imaginary value. Thus one bypasses the explicit use of the time-dependent Schrödinger equation and hence the typically oscillatory nature of the complex wavefunction. This seems beneficial in the first place because (a) one deals with the real quantum mechanical variables and (b) density and quantum current possess a smooth spatial variation as opposed to the wavefunction. Recent illustrations [16, 17] have demonstrated the smooth spatial and temporal nature of the variables and the ability to discretize them on a relatively small number of grid points. This paper is organized as follows. In section 2 we give a brief account of the BQH. In section 3 we provide the QOC formulation based on the BQH. In section 4 we apply the method to manipulate the vibrational motion of a HBr molecule in its ground electronic state. Section 5 concludes the paper.

2. Bhomian quantum hydrodynamics

Despite its extraordinary success, quantum mechanics has, since its inception some 70 years ago, been plagued by conceptual difficulties. According to orthodox quantum theory, the complete description of a system of particles is provided by its wavefunction Ψ which obeys the time-dependent Schrödinger equation,

$$\mathrm{i}\hbar\frac{\partial\Psi(q,t)}{\partial t} = H\psi(q,t). \tag{1}$$

According to Bohm [18], the complete description of a quantum system is provided by its wavefunction $\Psi(q,t)$, $q \in R^3$, and its configuration $Q \in R^3$ where Q is the position of the particle. The wavefunction, which evolves according to Schrödinger's equation (equation (1)) choreographs the motion of the particle which evolves according to the equation

$$\frac{\mathrm{d}Q}{\mathrm{d}t} = \frac{\hbar}{m}\frac{\mathrm{Im}(\Psi^*\nabla\Psi)}{\Psi^*\Psi} \tag{2}$$

where $\nabla = \frac{\partial}{\partial q}$. In the above equation H is the usual non-relativistic Hamiltonian for spinless particle given as

$$H = -\frac{\hbar^2}{2m}\nabla^2 + V. \tag{3}$$

Equations (1) and (2) give a complete specification of the quantum theory describing the behaviour of any observables or the effects of measurement. Note that Bohm's formulation incorporates Schrödinger's equation into a rational theory, describing the motion of particles, merely by adding a single equation, the guiding equation (equation (2)). In so doing it provides a precise role for the wavefunction in sharp contrast with its rather obscure status in orthodox quantum theory. The additional equation (equation (2)) emerges in an almost inevitable manner. Bell's preference is to observe that the probability current j^Ψ and the probability density $\rho = \Psi^*\Psi$ would classically be related by $j = \rho v$ obviously suggests that

$$\frac{\mathrm{d}Q}{\mathrm{d}t} = v = j/\rho. \tag{4}$$

Bohm, in his seminal hidden-variable paper wrote the wavefunction Ψ in the polar form $\Psi = R\mathrm{e}^{\mathrm{i}S/\hbar}$ where S is real and $R \geqslant 0$, and then rewrote the Schrödinger's equation in terms of these new variables, obtaining a pair of coupled evolution equations, the continuity equation for $\rho = R^2$ as

$$\frac{\partial\rho}{\partial t} = -\nabla\cdot(\rho v) \tag{5}$$

which suggests that ρ should be interpreted as a probability density, and a modified Hamilton–Jacobi equation for S,

$$\frac{\partial S}{\partial t} + H(\nabla S, q) + V_q = 0 \tag{6}$$

where $H = H(p, q)$ is the classical Hamiltonian function corresponding to equation (3), and

$$\begin{aligned} V_q &= -\frac{\hbar^2}{2m}\frac{\nabla^2 R}{R} \\ &= -\frac{\hbar^2}{2m}\nabla^2 \ln \rho^{1/2} - \frac{\hbar^2}{2m}(\nabla \ln \rho^{1/2})^2. \end{aligned} \tag{7}$$

Equation (6) differs from the classical Hamilton–Jacobi equation only by the appearance of an extra term, the quantum potential V_q. Similar to the classical Hamilton–Jacobi equation, Bohm defined the quantum particle trajectories, by identifying ∇S with mv, by

$$\frac{\mathrm{d}Q}{\mathrm{d}t} = \frac{\nabla S}{m} \tag{8}$$

which is equivalent to equation (4). This is precisely what would have been obtained classically if the particles were acted upon by the force generated by a quantum potential in addition to the usual forces. Although an interpretation in classical terms is beautifully laid down in the above equations, one should keep in mind that in so doing, the linear Schrödinger equation is transformed into a highly nonlinear equations (equations (5) and (6)). By taking the gradient on both sides of equation (6) we obtain

$$\frac{\partial}{\partial t}\boldsymbol{v} = -(\boldsymbol{v} \cdot \nabla)\boldsymbol{v} - \boldsymbol{v} \times (\nabla \times \boldsymbol{v}) - \frac{1}{m}\nabla(V + V_q). \tag{9}$$

Defining the quantum current as

$$\boldsymbol{j}(q, t) = \frac{1}{2}\frac{\hbar}{m}\,\mathrm{Im}[\Psi^*(q, t)\nabla\Psi(q, t) - \Psi(q, t)\nabla\Psi^*(q, t)] = \rho(q, t)\boldsymbol{v}(q, t)$$

and using the equation $\nabla \times \boldsymbol{v} = 0$ we readily obtain the expression for the motion of the quantum current as

$$\frac{\partial}{\partial t}\boldsymbol{j} = -\boldsymbol{v}(\nabla \cdot \boldsymbol{j}) - (\boldsymbol{j} \cdot \nabla)\boldsymbol{v} - \frac{\rho}{m}\nabla(V + V_q). \tag{10}$$

Equations (5), (6), (9) and (10) describe the motion of a quantum particle in the hydrodynamical representation of TDSE. However, the many-particle description of the BQH can be found elsewhere [24]. It may be noted that density alone cannot sufficiently describe a quantum system, one requires both density and the quantum current for the purpose. As is evident, the motion of a quantum particle is governed by the quantum current vector $\boldsymbol{j}$ unlike the TDSE where the time propagator $\mathrm{e}^{\mathrm{i}Ht}$ plays a key role in the particle's motion. The difficulties arising out of the evaluation of the exponential of an operator in more than one dimension is completely bypassed in the hydrodynamical equations. Although the hydrodynamical equations resemble the classical fluid dynamical equations, the quantum identity prevails because of the fact that the quantum current evolves with respect to a potential V_q which has no classical analogue [19]. It should be noted that the term V_q was inherently present in the expression to stabilize the hydrodynamical approach to the TDSE. The numerical instability in the hydrodynamical approach to the TDSE without the presence of the V_q term may be related to 'shock' formation in classical hydrodynamics (cf the Navier–Stokes equation) without some fictitious smoothing potential. In the numerical solution we shall work with the equations governing the motion of density (equation (5)) and the quantum current (equation (10)). The motivation for considering the above equations lies in the fact that (a) the density ρ and quantum current J are uniquely defined for a given potential in a many-body system, whereas the phase S can be multivalued ($S = S \pm n\pi$, $n =$ even); and (b) they are quantum mechanical observables. Equations (5) and (6) suggest that one can obtain density and the quantum current directly for $t > 0$ provided the values were known at $t = 0$. Thus, the scheme bypasses the evaluation of the wavefunction during the occurrence of the dynamics. However, at $t = 0$, one has to solve the time-independent Schrödinger equation for the wavefunction and calculate $\rho(q, 0)$ and $j(q, 0)$.

3. Quantum optimal control and Bhomian quantum hydrodynamics

Quantum optimal control theory seeks to design an external field to fulfil a particular objective. This section will provide the rigorous mathematical formulation of the hydrodynamic method to design an optimal time-dependent field that drives a quantum wavepacket to a desired objective at the target time $t = T$. For this purpose, consider a general target expectation value defined as $\Theta_T = \int_0^T \Theta\rho(x, T)\,\mathrm{d}x$, where Θ is an observable operator and $\rho(x, T)$ is the probability density which obeys the hydrodynamical equations, namely equations (5) and (10). The goal is to steer Θ_T as close as possible to a desired value Θ^d. We define a quadratic cost functional as

$$J_q = \tfrac{1}{2}\omega_a(\Theta_T - \Theta^d)^2. \tag{11}$$

Minimization of J_q amounts to the equalization of Θ_T to Θ^d. However, ρ in the above equation must obey the hydrodynamical equations, namely equations (5) and (10). Thus, we have to fulfil this constraint whereby we obtain the unconstrained cost functional as

$$\begin{aligned}\bar{J} = J_q &- \iint \lambda_1(x,t)\left[\frac{\partial\rho(x,t)}{\partial t} + \frac{\partial j(x,t)}{\partial x}\right]\mathrm{d}x\,\mathrm{d}t \\ &- \iint \lambda_2(x,t)\left[\frac{\partial j(x,t)}{\partial t} + \frac{\partial}{\partial x}\left(\frac{j^2}{\rho}\right) + \rho\frac{\partial}{\partial x}(V + V_q + V_{ext}(t))\right]\mathrm{d}x\,\mathrm{d}t \end{aligned} \tag{12}$$

where $V_{ext}(t)$ represents the external potential due to the interaction between the particle and the electric field, $E(t)$ to be designed.

Thus, in the above equations, we have introduced two Lagrange multipliers $\lambda_1(x, t)$ and $\lambda_2(x, t)$. There exists another constraint involving the total energy in the field which must be imposed on the optimization procedure. This constraint takes the form

$$\tfrac{1}{2}\omega_e\left[\int_0^T E^2(t)\,\mathrm{d}t - E_p\right] = 0 \tag{13}$$

where E_p is the energy of the pulse and $E(t)$ is the field to be designed. The parameters ω_a and ω_e are the positive weights balancing the significance of the two terms, namely J_q and $J_e = \frac{1}{2}\omega_e\int_0^T E^2(t)\,\mathrm{d}t$, respectively. The term $J_e = \frac{1}{2}\omega_e\int_0^T E^2(t)\,\mathrm{d}t$ represents the penalty due to the fluence of the external field. So the full unconstrained cost functional takes the form

$$\begin{aligned}\bar{J} = J_q &- \iint \lambda_1(x,t)\left[\frac{\partial\rho(x,t)}{\partial t} + \frac{\partial j(x,t)}{\partial x}\right]\mathrm{d}x\,\mathrm{d}t \\ &- \iint \lambda_2(x,t)\left[\frac{\partial j(x,t)}{\partial t} + \frac{\partial}{\partial x}\left(\frac{j^2}{\rho}\right) + \rho\frac{\partial}{\partial x}(V + V_q + V_{ext}(t))\right]\mathrm{d}x\,\mathrm{d}t \\ &+ \tfrac{1}{2}\omega_e\left[\int_0^T E^2(t)\,\mathrm{d}t - E_p\right]. \end{aligned} \tag{14}$$

In this equation $\bar{J}$ is seen to be a functional of five functions, namely $\rho(x,t)$, $j(x,t)$, $\lambda_1(x,t)$, $\lambda_2(x,t)$ and $E(t)$, all of which are real, unlike in the conventional method [6–8]. In the above equations J is the cost functional and j is the quantum current. The total variation of $\bar{J}$ can be written as

$$\delta\bar{J} = \iint \frac{\delta\bar{J}}{\delta\rho(x,t)}\delta\rho(x,t)\,\mathrm{d}x\,\mathrm{d}t + \iint \frac{\delta\bar{J}}{\delta j(x,t)}\delta j(x,t)\,\mathrm{d}x\,\mathrm{d}t$$
$$+ \iint \frac{\delta\bar{J}}{\delta\lambda_1(x,t)}\delta\lambda_1(x,t)\,\mathrm{d}x\,\mathrm{d}t + \iint \frac{\delta\bar{J}}{\delta\lambda_2(x,t)}\delta\lambda_2(x,t)\,\mathrm{d}x\,\mathrm{d}t$$
$$+ \iint \frac{\delta\bar{J}}{\delta E(t)}\delta E(t)\,\mathrm{d}x\,\mathrm{d}t. \tag{15}$$

For any optimal solution $\delta\bar{J} = 0$, which gives

$$\frac{\delta\bar{J}}{\delta\rho(x,t)} = \frac{\delta\bar{J}}{\delta j(x,t)} = \frac{\delta\bar{J}}{\delta\lambda_1(x,t)} = \frac{\delta\bar{J}}{\delta\lambda_2(x,t)} = \frac{\delta\bar{J}}{\delta E(t)} = 0. \tag{16}$$

We have provided in the appendix the full expression for $\delta\bar{J}$. Comparing equation (16) with equation (A14) we obtain

$$\frac{\delta\bar{J}}{\delta\lambda_1(x,t)} = -\frac{\partial\rho}{\partial t} - \frac{\partial j}{\partial x} = 0 \tag{17}$$

$$\frac{\delta\bar{J}}{\delta\lambda_2(x,t)} = -\frac{\partial j}{\partial t} - \frac{\partial}{\partial x}\left(\frac{j^2}{\rho}\right) - \rho\frac{\partial}{\partial x}(V + V_q + V_{ext}(t)) = 0 \tag{18}$$

$$\frac{\delta\bar{J}}{\delta j(x,t)} = \frac{\partial\lambda_2}{\partial t} + \frac{\partial\lambda_1}{\partial x} + 2\frac{\partial}{\partial x}\left(\lambda_2\frac{j}{\rho}\right) - 2\frac{\lambda_2}{\rho}\frac{\partial j}{\partial x} + 2\frac{\lambda_2 j}{\rho^2}\frac{\partial\rho}{\partial x} = 0 \tag{19}$$

$$\frac{\delta\bar{J}}{\delta\rho(x,t)} = \frac{\partial\lambda_1}{\partial t} + 2\frac{\lambda_2 j}{\rho^2}\frac{\partial j}{\partial x} - \frac{\partial}{\partial x}\left(\lambda_2\frac{j^2}{\rho^2}\right) - 2\frac{\lambda_2 j^2}{\rho^3}\frac{\partial\rho}{\partial x} - \lambda_2\frac{\partial}{\partial x}(V + V_q + V_{ext}(t))$$
$$-\frac{1}{2\mu\rho^{1/2}}\frac{\partial^2}{\partial x^2}\left(\frac{1}{\rho^{1/2}}\frac{\partial}{\partial x}(\lambda_2\rho)\right) + \frac{1}{4\mu\rho^{3/2}}\frac{\partial^2}{\partial x^2}\rho^{1/2}\frac{\partial}{\partial x}(\lambda_2\rho) = 0 \tag{20}$$

$$\frac{\delta\bar{J}}{\delta\rho(x,T)} = \omega_a[\Theta_T - \Theta^d]x - \lambda_1(x,T) = 0 \tag{21}$$

$$\frac{\delta\bar{J}}{\delta j(x,T)} = -\lambda_2(x,T) = 0 \tag{22}$$

$$\frac{\delta\bar{J}}{\delta E(t)} = \int \lambda_2(x,t)\rho(x,t)\frac{\partial}{\partial x}\mu(x)\,\mathrm{d}x + \omega_e E(t) = 0. \tag{23}$$

Equations (19) and (20) can be rewritten in a simple form as

$$\frac{\partial\lambda_2}{\partial t} + \frac{\partial}{\partial x}(\lambda_2 v_\lambda) + S_1[\rho, j, \lambda_2] = 0 \tag{24}$$

and

$$\frac{\partial\lambda_1}{\partial t} + \frac{\partial}{\partial x}(\lambda_1 v_\lambda) - \lambda_2\frac{\partial}{\partial x}(V + V_q(\lambda_2) + V_{ext}) + S_2[\rho, j, \lambda_2] = 0 \tag{25}$$

where

$$S_1 = -2\frac{\lambda_2}{\rho}\frac{\partial j}{\partial x} \tag{26}$$

and

$$S_2 = -\lambda_2\frac{\partial}{\partial x}(V_q(\rho) - V_q(\lambda_2)) - \frac{j^2}{\rho^2}\frac{\partial \lambda_2}{\partial x} - \frac{1}{4\rho^{1/2}}\frac{\partial^2}{\partial x^2}\left[\frac{1}{\rho^{1/2}}\frac{\partial}{\partial x}(\lambda_2\rho)\right]$$
$$+\frac{1}{4\rho^{3/2}}\frac{\partial^2}{\partial x^2}\rho^{1/2}\frac{\partial}{\partial x}(\lambda_2\rho). \tag{27}$$

Note that the above expression for Θ_T restricts the operator Θ to being only a multiplicative operator, for example, the distant $\hat{x}$ which we have used in the subsequent numerical calculations. However, other forms of operator can also be considered in the BQH QOC formulation with the different constraint expressions, for example, if Θ is the momentum operator ($\hat{p}$) we would require the constraint equations (5) and (9) since $p_T = m\int_0^T \rho(x,T)\nabla S(x,T)\,\mathrm{d}x$.

The equations for λ_1 and λ_2 resemble those of ρ and j, with the only difference being the extra source terms S_1 and S_2. The source terms depend on ρ and j. v_λ in the above equations is the velocity associated with the Lagrange multiplier and is given as $v_\lambda = \frac{\lambda_1}{\lambda_2}$ and $V_q(\lambda_2)$ is given by

$$V_q(\lambda_2) = -\frac{\hbar^2}{2\mu}\frac{\nabla^2\lambda_2^{1/2}}{\lambda_2^{1/2}}.$$

Note that equations (17) and (18) are the equations of motion for the probability density and the quantum current density, respectively, obtained in section 2. Whereas equations (24) and (25) are the equations of motion for the two Lagrange multipliers λ_2 and λ_1, respectively. It should be noted that in obtaining the above equations (see the appendix) we have assumed no variation on either $\rho(x,0)$ or $j(x,0)$. Thus, we start from an initial ($t=0$) $\rho(x,0)$ and $j(x,0)$ to solve equations (17) and (18) for $\rho(x,t)$ and $j(x,t)$, respectively. Equations (24) and (25) can be solved for $\lambda_1(x,t)$ and $\lambda_2(x,t)$ provided starting values $\lambda_1(x,t_s)$ and $\lambda_2(x,t_s)$ are known. These have been obtained from equations (21) and (22), respectively, as

$$\lambda_1(x,t_s) = \omega_a[\Theta_T - \Theta^d]x \qquad \text{and} \qquad \lambda_2(x,t_s) = 0 \tag{28}$$

where $t_s = T$, i.e. the final time. Thus, one has to perform backward propagation to solve both the equations of motion involving $\lambda_1(x,t)$ and $\lambda_2(x,t)$. Having calculated $\rho(x,t)$, $j(x,t)$, $\lambda_1(x,t)$ and $\lambda_2(x,t)$ as described above, one has to carry out an optimization of the quadratic cost functional (equation (11)) with respect to the electric field $E(t)$ which, according to equation (23), takes the form

$$E(t) = -\frac{1}{\omega_e}\int \lambda_2(x,t)\rho(x,t)\frac{\partial}{\partial x}\mu(x)\,\mathrm{d}x. \tag{29}$$

This constitute the details of the BQH QOC method.

4. Application to the HBr molecule

We have mentioned in the preceding section that we need the initial density $\rho(x,0)$ and the quantum current $j(x,0)$ in the present method. These have been evaluated by solving the time-independent Schrödinger equation for the HBr molecule in the ${}^1\Sigma^+$ state where the potential energy is assumed Morse type of the form [20]

$$V = D_e(1 - \exp(-\beta(x - x_e)))^2 \tag{30}$$

where $\beta = \omega_e\left(\frac{\mu}{2D_e}\right)^{1/2}$, $D_e = \frac{\omega_e^2}{4\omega_e x_e}$ with $\omega_e = 2648.975$ cm^{-1}, $\omega_e x_e = 45.217$ cm^{-1}, $x_e = 1.414\,43$ Å and μ is the reduced mass of HBr.

Having obtained $\rho(x, 0)$ and $j(x, 0)$ we carry out the control by the present method. The following are the necessary steps for the computer implementation of the present method:

4.1. Present method

Step 1. Make an initial guess for the electric field $E(t)$, which is zero in our calculation.

Step 2. Solve the coupled equations, namely equations (17) and (18) for $\rho(x, t)$ and $j(x, t)$, respectively, starting from $\rho(x, 0)$ and $j(x, 0)$. The solution is calculated by using the flux-corrected transport (FCT) algorithm [21] modified by us for the purpose of solving the quantum hydrodynamical equations [16, 17]. In so doing, we adopt the Eulerian scheme

Step 3. Evaluate the final values for $\lambda_1(x, T)$ and $\lambda_2(x, T)$ given by equation (28).

Step 4. Use $\lambda_1(x, T)$ and $\lambda_2(x, T)$ to solve equations (24) and (25) for $\lambda_1(x, t)$ and $\lambda_2(x, t)$, respectively. This is done by backward propagation, by putting $\mathrm{d}t = -\mathrm{d}t$ (see [16]). We follow the same method as in step 2 to solve these equations. It should be noted that equations (24) and (25) have source terms which depend on $\rho(x, t)$ and $j(x, t)$ calculated in step 2.

Step 5. Calculate the quadratic cost functional given by equation (11).

Step 6. Optimize the function in equation (11) with respect to the electric field, $E(t)$ given by equation (29). Here we use the conjugate direction search method [22] for the optimization.

Step 7. Iterate steps 2–6 until a convergence criterion is satisfied.

The external potential is of the form $V_{ext}(x, t) = -\mu(x)E(t)$, where $\mu(x)$ is the dipole function for HBr and is given by [23] $\mu(x) = A_0 + A_1(x - x_e) + A_2(x - x_e)^2$, where $A_0 = 0.788$, $A_1 = 0.315$ and $A_2 = 0.575$. In our calculation the range of spatial dimensions is $0 \leqslant x \leqslant 12$ au and that of time is $0 \leqslant t \leqslant 2000$ au. The total number of spatial mesh points is 60, which gives $\Delta x = 0.2$ au. Similarly, the total number of time steps is 2000, which corresponds to $\Delta t = 1.0$ au, ω_e in equation (29) is taken as 0.5, and ω_a as 1000. The target operator is $\Theta = x$ and $\Theta^d = 3.0$ and 3.5 au.

Figure 1 shows the electric fields corresponding to two different values of Θ^d namely 3.0 (full curves) and 3.5 (dotted curves). These pulses excite several vibrational states (not shown here) mainly by a sequence of single quantum transitions. The peak value of the field is ≈ 0.08 au (the corresponding intensity is $\approx 10^{14}$ W cm^{-2}) for $\Theta^d = 3.5$ and ≈ 0.02 au (the corresponding intensity is $\approx 10^{13}$ W cm^{-2}) for $\Theta^d = 3.0$ au. A detailed characterization of the optimal field can, however, be made by Fourier transforming the field. Figure 2 shows the average distance $\langle x\rangle$ as a function of time. Notice the desired control of $\langle x\rangle = 3.0$ and 3.5 au at $T = 2000$ au is obtained through the oscillatory motion of the packed induced by the optimal electric pulse (figure 1). Figure 3 shows the initial and final densities for the two values of Θ^d.

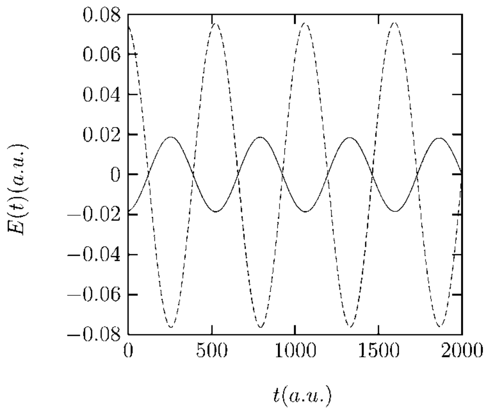

Figure 1. Optimal electric field shown as a function of time for $\Theta_T = 3.0$ au (full curve) and $\Theta_T = 3.5$ au (dotted curve).

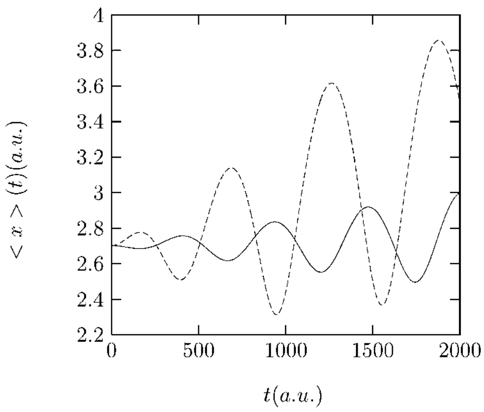

Figure 2. The expectation values $\langle x \rangle$ shown as a function of time for $\Theta_T = 3.0$ au (full curve) and $\Theta_T = 3.5$ au (dotted curve).

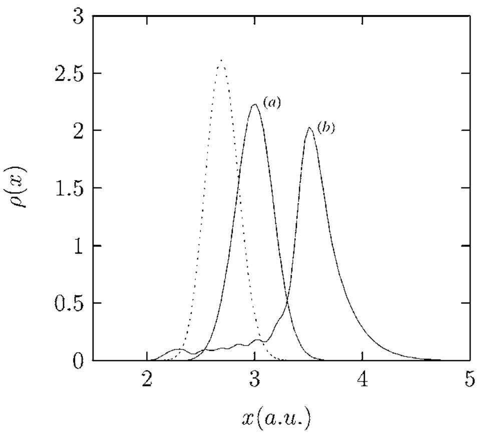

Figure 3. Initial ($t = 0$) (dotted curve) and final ($t = T$) (full curve) density corresponding to $\Theta_T = 3.0$ au (a) and $\Theta_T = 3.5$ au (b).

The packet is distorted in shape, while approximately retaining its original variance during the evolution. During the optimization process the total integrated probability density remained at unity up to a deviation of 10^{-7}. The number of iterations in the optimization to achieve the results is five and it takes only 3 min (real) on an IRIX IP30 machine with a R4400 6.0 CPU. As a test for the acceptability of the present method we have carried out the following experiment: the electric fields (figure 1) so obtained have been plugged into the TDSE and then solved for the wavefunction. The results for the density and the expectation value of $\langle x \rangle$ resemble closely those given in figure 2.

5. Conclusion

In the present paper we have presented a new scheme for carrying out the optimal design based on BQH. We have derived the control equations to obtain a time-dependent external field with an illustration for the manipulation of the vibrational motion of the HBr molecule in the $^1\Sigma^+$ state. The working dynamical variables in the BQH, namely ρ (figure 3), j, λ_1 and λ_2 are relatively slowly varying spatial functions (figure 4) compared with the wavefunction

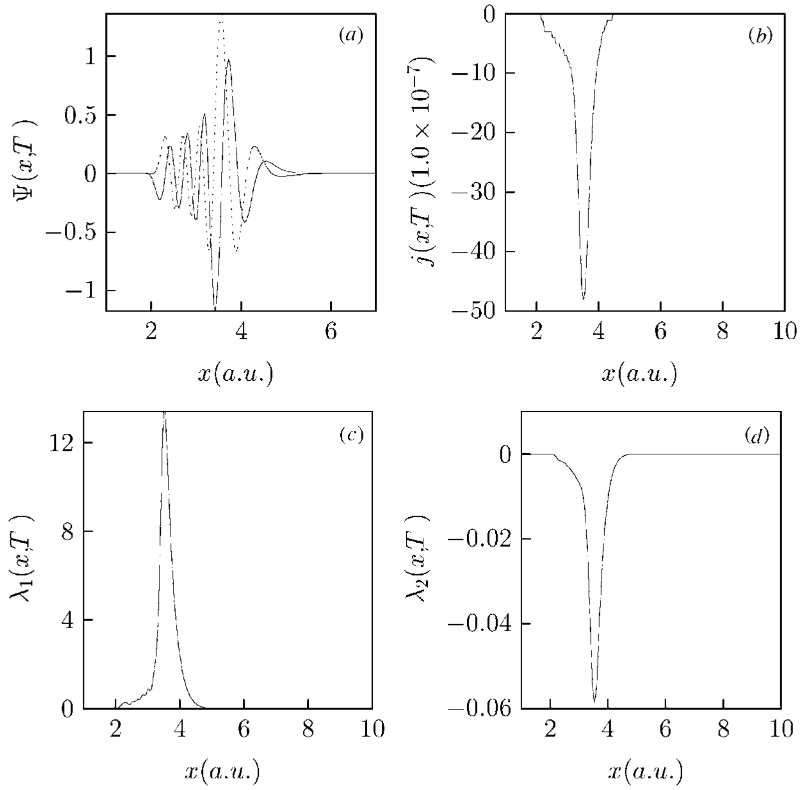

Figure 4. Hydrodynamical variables, namely $j(x, T)$ (*b*), $\lambda_1(x, T)$ (*c*), $\lambda_2(x, T)$ (*d*) and the real (full) and imaginary (dotted) values of the wavefunction (*a*) plotted as a function of x. Note that the hydrodynamical variables are smooth spatial function, unlike the wavefunction.

(figure 4, curve (*a*)) which apparently enhances the efficiency and the numerical saving of the BQH QOC method for controlling dynamics.

Although we have illustrated our new method in one spatial dimension, the approach is general and is directly extendible to higher dimensions and a wavepacket dynamics in four dimensions has already been performed [16] within our method. The use of the alternating direction implicit (ADI) [16, 17] method in the present method makes the quantum control calculation much easier compared with the conventional method, especially for the multidimensional problem. In conventional optimal control theory, the role of the complex Lagrange multiplier is to provide feedback [6] for designing the electric field and to guide the dynamics to an acceptable solution. The BQH QOC method, on the other hand, introduces two such Lagrange multipliers, λ_1 and λ_2 both of which are real variables. The first Lagrange multiplier λ_1, which corresponds to the quantum current j (cf equations (18) and (25)) has, however, no direct role to provide feedback for designing the electric field (equation (29)) and only guides the dynamics in conjunction with the second Lagrange multiplier λ_2. It may be worth mentioning that since the quadratic cost functional (equation (11)) is a functional of density, the Lagrange multiplier λ_2 (equivalent to the density ρ, cf equations (17) and (24)) enters into the expression for the optimal electric field (equation (29)). However, cases where one desires to manipulate the quantum flux (which is directly related to the quantum current

j) by constructing a quadratic cost functional dependent on j, the Lagrange multiplier λ_1 will appear explicitly into the expression for the external field.

It should be pointed out that the present method could prove to be difficult in cases where the dynamics may lead to the creation of nodes in the density profile since the quantum potential appearing in the constraint equation blows up on the occurrence of such an event. However, such an occurrence of nodes can be countered by fixing a lower limit to the density of the order of the machine precision. In other words, this means that one never encounters an absolute nodal point where the density is exactly zero. Future studies need to explore the other area of control within the BQH QOC method, for example, controlling the quantum flux.

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Appendix

The variation of $\bar{J}$ given by equation (14) has to be taken with respect to $\rho(x,t)$, $j(x,t)$, $\lambda_1(x,t)$, $\lambda_2(x,t)$ and $E(t)$. Any variation $\delta\rho(x,t)$, $\delta j(x,t)$, $\delta\lambda_1(x,t)$, $\delta\lambda_2(x,t)$ and $\delta E(t)$ will lead to the variation $\delta\bar{J}$ given as

$$\begin{aligned}\delta\bar{J} = \delta J_q &- \iint\left[\frac{\partial\rho}{\partial t} + \frac{\partial j}{\partial x}\right]\delta\lambda_1\,\mathrm{d}x\,\mathrm{d}t \\ &- \iint\left[\frac{\partial j}{\partial t} + \frac{\partial}{\partial x}\left(\frac{j^2}{\rho}\right) + \rho\frac{\partial}{\partial x}(V + V_q + V_{ext})\right]\delta\lambda_2\,\mathrm{d}x\,\mathrm{d}t \\ &- \iint\lambda_1\left[\frac{\partial}{\partial t}\delta\rho + \frac{\partial}{\partial x}\delta j\right]\mathrm{d}x\,\mathrm{d}t \\ &- \iint\lambda_2\left[\frac{\partial}{\partial t}\delta j + \frac{\partial}{\partial x}\left(\frac{2j}{\rho}\delta j - \frac{j^2}{\rho^2}\delta\rho\right) + \delta\rho\frac{\partial}{\partial x}(V + V_q + V_{ext})\right. \\ &\left. + \rho\frac{\partial}{\partial x}(\delta V + \delta V_q + \delta V_{ext})\right]\mathrm{d}x\,\mathrm{d}t + \omega_e\int E(t)\delta E(t)\,\mathrm{d}t.\end{aligned} \tag{A1}$$

Now, we have $\delta V = 0$ and δV_{ext} can be written as $\delta V_{ext} = -\delta(\mu(x)E(t)) = -\mu(x)\delta E(t) - E(t)\delta\mu(x)$. Since $\mu(x)$ is kept fixed, we obtain $\delta V_{ext} = -\mu(x)\delta E(t)$. J_q in the above equation is given by

$$J_q = \tfrac{1}{2}\omega_a\left[\int\rho(x,T)x\,\mathrm{d}x - x_{cm}^d\right]^2. \tag{A2}$$

Hence

$$\delta J_q = \omega_a[\langle x\rangle(T) - x_{cm}^d]\int x\delta\rho(x,T)\,\mathrm{d}x. \tag{A3}$$

Substituting equations (A3) into equation (A1) we obtain

$$\begin{aligned}\delta\bar{J} = \omega_a[\langle x\rangle(T) - x_{cm}^d]\int x\delta\rho(x,T)\,\mathrm{d}x &- \iint\left[\frac{\partial\rho}{\partial t} + \frac{\partial j}{\partial x}\right]\delta\lambda_1\,\mathrm{d}x\,\mathrm{d}t \\ &- \iint\left[\frac{\partial j}{\partial t} + \frac{\partial}{\partial x}\left(\frac{j^2}{\rho}\right) + \rho\frac{\partial}{\partial x}(V + V_q + V_{ext})\right]\delta\lambda_2\,\mathrm{d}x\,\mathrm{d}t\end{aligned}$$

$$
\begin{aligned}
&-\iint \lambda_1 \frac{\partial}{\partial t}\delta\rho\,\mathrm{d}x\,\mathrm{d}t - \iint \lambda_1 \frac{\partial}{\partial x}\delta j\,\mathrm{d}x\,\mathrm{d}t - \iint \lambda_2 \frac{\partial}{\partial t}\delta j\,\mathrm{d}x\,\mathrm{d}t \\
&-2\iint \lambda_2 \frac{j}{\rho}\frac{\partial}{\partial x}\delta j\,\mathrm{d}x\,\mathrm{d}t - 2\iint \lambda_2 \frac{1}{\rho}\frac{\partial j}{\partial x}\delta j\,\mathrm{d}x\,\mathrm{d}t + 2\iint \lambda_2 \frac{j}{\rho^2}\frac{\partial \rho}{\partial x}\delta j\,\mathrm{d}x\,\mathrm{d}t \\
&+\iint \lambda_2 \frac{j^2}{\rho^2}\frac{\partial}{\partial x}\delta\rho\,\mathrm{d}x\,\mathrm{d}t + 2\iint \lambda_2 \frac{j}{\rho^2}\frac{\partial j}{\partial x}\delta\rho\,\mathrm{d}x\,\mathrm{d}t - 2\iint \lambda_2 \frac{j^2}{\rho^3}\frac{\rho}{\partial x}\delta\rho\,\mathrm{d}x\,\mathrm{d}t \\
&-\iint \lambda_2 \frac{\partial}{\partial x}(V + V_q + V_{ext})\delta\rho\,\mathrm{d}x\,\mathrm{d}t - \iint \lambda_2 \rho \frac{\partial}{\partial x}\delta V_q\,\mathrm{d}x\,\mathrm{d}t \\
&+\iint \lambda_2 \rho \frac{\partial}{\partial x}(\mu(x)\delta E(t))\,\mathrm{d}x\,\mathrm{d}t + \omega_e \int E(t)\delta E(t)\,\mathrm{d}t. \qquad \text{(A4)}
\end{aligned}
$$

The fourth and sixth terms in the above equation can be simplified by integration by parts as follows:

$$
\begin{aligned}
\iint \lambda_1 \frac{\partial}{\partial t}\delta\rho\,\mathrm{d}x\,\mathrm{d}t &= \int \lambda_1(x,T)\delta\rho(x,T)\,\mathrm{d}x - \int \lambda_1(x,0)\delta\rho(x,0)\,\mathrm{d}x \\
&\quad - \iint \frac{\partial \lambda_1}{\partial t}\delta\rho(x,t)\,\mathrm{d}x\,\mathrm{d}t \qquad \text{(A5)}
\end{aligned}
$$

$$
\begin{aligned}
\iint \lambda_2 \frac{\partial}{\partial t}\delta j\,\mathrm{d}x\,\mathrm{d}t &= \int \lambda_2(x,T)\delta j(x,T)\,\mathrm{d}x - \int \lambda_2(x,0)\delta j(x,0)\,\mathrm{d}x \\
&\quad - \iint \frac{\partial \lambda_2}{\partial t}\delta j(x,t)\,\mathrm{d}x\,\mathrm{d}t. \qquad \text{(A6)}
\end{aligned}
$$

The fifth, seventh and tenth terms can similarly be expressed by integration by parts as follows:

$$
\iint \frac{\partial}{\partial x}\delta j\,\mathrm{d}x\,\mathrm{d}t = \int \lambda_1(x_r,t)\delta j(x_r,t)\,\mathrm{d}t - \int \lambda_1(x_l,t)\delta j(x_l,t)\,\mathrm{d}t - \iint \frac{\partial \lambda_1}{\partial x}\delta j(x,t)\,\mathrm{d}x\,\mathrm{d}t \qquad \text{(A7)}
$$

$$
\begin{aligned}
\iint \lambda_2 \frac{j}{\rho}\frac{\partial}{\partial x}\delta j\,\mathrm{d}x\,\mathrm{d}t &= \int \frac{\lambda_2(x_r,t)j(x_r,t)}{\rho(x_r,t)}\delta j(x_r,t)\,\mathrm{d}t - \int \frac{\lambda_2(x_l,t)j(x_l,t)}{\rho(x_l,t)}\delta j(x_l,t)\,\mathrm{d}t \\
&\quad - \iint \frac{\partial}{\partial x}\left(\lambda \frac{j}{\rho}\right)\delta j\,\mathrm{d}x\,\mathrm{d}t \qquad \text{(A8)}
\end{aligned}
$$

$$
\begin{aligned}
\iint \lambda_2 \frac{j^2}{\rho^2}\frac{\partial}{\partial x}\delta\rho\,\mathrm{d}x\,\mathrm{d}t &= \int \frac{\lambda_2(x_r,t)j^2(x_r,t)}{\rho^2(x_r,t)}\delta\rho(x_r,t)\,\mathrm{d}t - \int \frac{\lambda_2(x_l,t)j^2(x_l,t)}{\rho^2(x_l,t)}\delta\rho(x_l,t)\,\mathrm{d}t \\
&\quad - \iint \frac{\partial}{\partial x}\left(\lambda_2 \frac{j^2}{\rho^2}\right)\delta\rho\,\mathrm{d}x\,\mathrm{d}t. \qquad \text{(A9)}
\end{aligned}
$$

The 15th term is

$$
\iint \lambda_2 \rho \frac{\partial}{\partial x}(\mu(x)\delta E(t))\,\mathrm{d}x\,\mathrm{d}t = \iint \lambda_2 \rho \frac{\partial}{\partial x}\mu(x)\delta E(t)\,\mathrm{d}x\,\mathrm{d}t. \qquad \text{(A10)}
$$

The 14th term involves the variation in $\bar{J}$ due to the change in the quantum potential δV_q, where V_q is given by $V_q = -\frac{\hbar^2}{2\mu}\frac{\nabla^2\rho^{1/2}}{\rho^{1/2}}$. This gives

$$
\delta V_q = -\frac{\hbar^2}{4\mu\rho^{1/2}}\frac{\partial^2}{\partial x^2}\left(\frac{1}{\rho^{1/2}}\delta\rho\right) + \frac{\hbar^2}{4\mu\rho^{3/2}}\frac{\partial^2}{\partial x^2}\rho^{1/2}\delta\rho. \qquad \text{(A11)}
$$

By integration by parts we simplify the 14th term as follows:

$$\iint \lambda_2\rho\frac{\partial}{\partial x}\delta V_q\,\mathrm{d}x\,\mathrm{d}t = \int \lambda_2(x_r,t)\rho(x_r,t)\delta V_q(x_r,t)\,\mathrm{d}t - \int \lambda_2(x_l,t)\rho(x_l,t)\delta V_q(x_l,t)\,\mathrm{d}t$$
$$+\int \frac{\partial}{\partial x}(\lambda_2\rho)\frac{1}{2\rho^{1/2}}\frac{\partial}{\partial x}\left(\frac{1}{2\rho^{1/2}}\delta\rho\right)\bigg|_{x_l}^{x_r}\mathrm{d}t$$
$$-\int \frac{\partial}{\partial x}\left(\frac{\partial}{\partial x}(\lambda_2\rho)\frac{1}{2\rho^{1/2}}\right)\frac{1}{2\rho^{1/2}}\bigg|_{x_r}\delta\rho(x_r,t)\,\mathrm{d}t$$
$$+\int \frac{\partial}{\partial x}\left(\frac{\partial}{\partial x}(\lambda_2\rho)\frac{1}{2\rho^{1/2}}\right)\frac{1}{2\rho^{1/2}}\bigg|_{x_l}\delta\rho(x_l,t)\,\mathrm{d}t$$
$$+\iint \frac{\partial^2}{\partial x^2}\left[\frac{\partial}{\partial x}(\lambda_2\rho)\frac{1}{2\rho^{1/2}}\right]\frac{1}{2\rho^{1/2}}\delta\rho(x,t)\,\mathrm{d}x\,\mathrm{d}t$$
$$-\iint \frac{\partial}{\partial x}(\lambda_2\rho)\frac{1}{4\rho^{3/2}}\frac{\partial^2}{\partial x^2}\rho^{1/2}\delta\rho(x,t)\,\mathrm{d}x\,\mathrm{d}t, \tag{A12}$$

where x_r and x_l are the right and left ends of the one-dimensional grid, and $F(x)|_{x_l}^{x_r} = F(x_r) - F(x_l)$ where $F(x)$ is any function. The first and the second terms in equation (A12) are the contributions due to the change in the quantum potential at the two ends of the boundary only. Since, we take a large grid, ρ at the two ends of the grid is very small and can be assumed to be constant. This leads to $V_q(x_r,t)$ and $V_q(x_l,t)$ being very high constant values at any time and hence $\delta V_q(x_r,t) = \delta V_q(x_l,t) = 0$. By the same argument we can also neglect the contributions due to the third, fourth and fifth terms. Combining all the terms we obtain the full variation in $\bar{J}$ as

$$\delta\bar{J} = \omega_a[\langle x\rangle(T) - x_{cm}^d]\int x\delta\rho(x,T)\,\mathrm{d}x - \iint\left[\frac{\partial\rho}{\partial t} + \frac{\partial j}{\partial x}\right]\delta\lambda_1\,\mathrm{d}x\,\mathrm{d}t$$
$$-\iint\left[\frac{\partial j}{\partial t} + \frac{\partial}{\partial x}\left(\frac{j^2}{\rho}\right) + \rho\frac{\partial}{\partial x}(V + V_q + V_{ext})\right]\delta\lambda_2\,\mathrm{d}x\,\mathrm{d}t$$
$$-\int \lambda_1(x,T)\delta\rho(x,T)\,\mathrm{d}x + \int \lambda_1(x,0)\delta\rho(x,0)\,\mathrm{d}x$$
$$+\iint \frac{\partial\lambda_1}{\partial t}\delta\rho\,\mathrm{d}x\,\mathrm{d}t - \int \lambda_2(x,T)\delta j(x,T)\,\mathrm{d}x + \int \lambda_2(x,0)\delta j(x,0)\,\mathrm{d}x$$
$$+\iint \frac{\partial\lambda_2}{\partial t}\delta j\,\mathrm{d}x\,\mathrm{d}t - \int \lambda_1(x_r,t)\delta j(x_r,t)\,\mathrm{d}t + \int \lambda_1(x_l,t)\delta j(x_l,t)\,\mathrm{d}t$$
$$+\iint \frac{\partial\lambda_1}{\partial x}\delta j(x,t)\,\mathrm{d}x\,\mathrm{d}t - 2\int \frac{\lambda_2(x_r,t)j(x_r,t)}{\rho(x_r,t)}\delta j(x_r,t)\,\mathrm{d}t$$
$$+2\int \frac{\lambda_2(x_l,t)j(x_l,t)}{\rho(x_l,t)}\delta j(x_l,t)\,\mathrm{d}t + \iint \frac{\partial}{\partial x}\left(\lambda_2\frac{j}{\rho}\right)\delta j(x,t)\,\mathrm{d}x\,\mathrm{d}t$$
$$+\int \frac{\lambda_2(x_r,t)j^2(x_r,t)}{\rho^2(x_r,t)}\delta\rho(x_r,t)\,\mathrm{d}t - \int \frac{\lambda_2(x_r,t)j^2(x_r,t)}{\rho^2(x_r,t)}\delta\rho(x_r,t)\,\mathrm{d}t$$
$$-\iint \frac{\partial}{\partial x}\left(\lambda_2\frac{j^2}{\rho^2}\right)\delta\rho\,\mathrm{d}x\,\mathrm{d}t - 2\iint \lambda_2\frac{1}{\rho}\frac{\partial j}{\partial x}\delta j\,\mathrm{d}x\,\mathrm{d}t$$
$$+2\iint \lambda_2\frac{j}{\rho^2}\frac{\partial\rho}{\partial x}\delta j\,\mathrm{d}x\,\mathrm{d}t + \iint \lambda_2\frac{2j}{\rho^2}\frac{\partial j}{\partial x}\delta\rho\,\mathrm{d}x\,\mathrm{d}t$$

$$
-2\iint \lambda_2 \frac{j^2}{\rho^3}\frac{\partial \rho}{\partial x}\delta\rho\,\mathrm{d}x\,\mathrm{d}t - \iint \lambda_2 \frac{\partial}{\partial x}(V + V_q + V_{ext})\delta\rho\,\mathrm{d}x\,\mathrm{d}t
$$
$$
-\frac{1}{\mu}\iint \frac{\partial^2}{\partial x^2}\left[\frac{\partial}{\partial x}(\lambda_2\rho)\frac{1}{2\rho^{1/2}}\right]\frac{1}{2\rho^{1/2}}\delta\rho\,\mathrm{d}x\,\mathrm{d}t
$$
$$
+\frac{1}{\mu}\iint \frac{\partial}{\partial x}(\lambda_2\rho)\frac{1}{4\rho^{3/2}}\frac{\partial^2}{\partial x^2}\rho^{1/2}\delta\rho\,\mathrm{d}x\,\mathrm{d}t + \iint \lambda_2\rho\frac{\partial}{\partial x}\mu(x)\delta E(t)\,\mathrm{d}x\,\mathrm{d}t. \qquad \text{(A13)}
$$

This expression has 26 terms. Out of which, the fifth and eighth terms can be dropped because we do not vary the initial density and quantum current. Again, the 10th, 11th, 13th, 14th, 16th and 17th terms can also be dropped with the assumption that $\rho(x,t)$ and $j(x,t)$ are very small at the boundary. Thus, the actual full variation in $\bar{J}$ becomes

$$
\delta\bar{J} = \omega_a[\langle x\rangle(T) - x^d_{cm}]\int x\delta\rho(x,T)\,\mathrm{d}x - \iint\left[\frac{\partial\rho}{\partial t} + \frac{\partial j}{\partial x}\right]\delta\lambda_1\,\mathrm{d}x\,\mathrm{d}t
$$
$$
-\iint\left[\frac{\partial j}{\partial t} + \frac{\partial}{\partial x}\left(\frac{j^2}{\rho}\right) + \rho\frac{\partial}{\partial x}(V + V_q + V_{ext})\right]\delta\lambda_2\,\mathrm{d}x\,\mathrm{d}t
$$
$$
-\int \lambda_1(x,T)\delta\rho(x,T)\,\mathrm{d}x + \iint \frac{\partial\lambda_1}{\partial t}\delta\rho\,\mathrm{d}x\,\mathrm{d}t
$$
$$
-\int \lambda_2(x,T)\delta j(x,T)\,\mathrm{d}x + \iint \frac{\partial\lambda_2}{\partial t}\delta j\,\mathrm{d}x\,\mathrm{d}t
$$
$$
+\iint \frac{\partial\lambda_1}{\partial x}\delta j(x,t)\,\mathrm{d}x\,\mathrm{d}t + \iint \frac{\partial}{\partial x}\left(\lambda_2\frac{j}{\rho}\right)\delta j(x,t)\,\mathrm{d}x\,\mathrm{d}t
$$
$$
-\iint \frac{\partial}{\partial x}\left(\lambda_2\frac{j^2}{\rho^2}\right)\delta\rho\,\mathrm{d}x\,\mathrm{d}t - 2\iint \lambda_2\frac{1}{\rho}\frac{\partial j}{\partial x}\delta j\,\mathrm{d}x\,\mathrm{d}t
$$
$$
+2\iint \lambda_2\frac{j}{\rho^2}\frac{\partial\rho}{\partial x}\delta j\,\mathrm{d}x\,\mathrm{d}t + \iint \lambda_2\frac{2j}{\rho^2}\frac{\partial j}{\partial x}\delta\rho\,\mathrm{d}x\,\mathrm{d}t
$$
$$
-2\iint \lambda_2 \frac{j^2}{\rho^3}\frac{\partial \rho}{\partial x}\delta\rho\,\mathrm{d}x\,\mathrm{d}t - \iint \lambda_2 \frac{\partial}{\partial x}(V + V_q + V_{ext})\delta\rho\,\mathrm{d}x\,\mathrm{d}t
$$
$$
-\frac{1}{\mu}\iint \frac{\partial^2}{\partial x^2}\left[\frac{\partial}{\partial x}(\lambda_2\rho)\frac{1}{2\rho^{1/2}}\right]\frac{1}{2\rho^{1/2}}\delta\rho\,\mathrm{d}x\,\mathrm{d}t
$$
$$
+\frac{1}{\mu}\iint \frac{\partial}{\partial x}(\lambda_2\rho)\frac{1}{4\rho^{3/2}}\frac{\partial^2}{\partial x^2}\rho^{1/2}\delta\rho\,\mathrm{d}x\,\mathrm{d}t + \iint \lambda_2\rho\frac{\partial}{\partial x}\mu(x)\delta E(t)\,\mathrm{d}x\,\mathrm{d}t. \qquad \text{(A14)}
$$

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