

# Supplementary Materials: Electron Correlations in Local Effective Potential Theory

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## 1. Supplementary Material: Derivation of the General “Quantal Newtonian” Second Law

We provide here the derivation of the “Quantal Newtonian” second law of Equation (16) corresponding to the time-dependent Schrödinger equation of Equation (15). We rewrite the Hamiltonian as

$$\hat{H}(t) = -\frac{1}{2} \sum_i \nabla_i^2 + \frac{1}{2} \sum'_{ij} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} + \sum_i \left[ v(\mathbf{y}_i) - \Phi(\mathbf{y}_i) \right] + \sum_i \hat{\omega} \left[ \mathbf{y}_i; \mathbf{A}(\mathbf{y}_i) \right], \tag{S1}$$

where

$$\hat{\omega} \left[ \mathbf{y}; \mathbf{A}(\mathbf{y}) \right] = \frac{1}{2} A^2(\mathbf{y}) - i \hat{\Omega}(\mathbf{y}), \tag{S2}$$

$$\hat{\Omega}(\mathbf{y}) = \frac{1}{2} \left[ \nabla \cdot \mathbf{A}(\mathbf{y}) + 2\mathbf{A}(\mathbf{y}) \cdot \nabla \right], \tag{S3}$$

with  $\mathbf{y} = \mathbf{r}t; \mathbf{y}_i = \mathbf{r}_i t$ . In general, the wave function may be written as  $\Psi(\mathbf{X}t) = \Psi^R(\mathbf{X}t) + i\Psi^I(\mathbf{X}t)$ , where  $\Psi^R(\mathbf{X}t) \equiv \Psi^R(t)$  and  $\Psi^I(\mathbf{X}t) \equiv \Psi^I(t)$  are its real and imaginary parts. On substituting the wave function into the Schrödinger equation of Equation (15), we have

$$\begin{aligned} i \frac{\partial \Psi^R(t)}{\partial t} - \frac{\partial \Psi^I(t)}{\partial t} &= \left\{ -\frac{1}{2} \sum_i \nabla_i^2 + \sum_i \left[ v(\mathbf{y}_i) - \Phi(\mathbf{y}_i) \right] \right. \\ &\left. + \frac{1}{2} \sum'_{ij} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} + \sum_i \hat{\omega} \left[ \mathbf{y}_i; \mathbf{A}(\mathbf{y}_i) \right] \right\} \left( \Psi^R(t) + i\Psi^I(t) \right). \end{aligned} \tag{S4}$$

Equating the real parts of Equation (S4) yields

$$\begin{aligned} &\sum_i \left[ v(\mathbf{y}_i) - \Phi(\mathbf{y}_i) \right] + \frac{1}{2} \sum'_{ij} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} + \frac{1}{2} \sum_i A^2(\mathbf{y}_i) \\ &+ \frac{1}{\Psi^R(t)} \frac{\partial}{\partial t} \Psi^I(t) = \frac{1}{\Psi^R(t)} \left\{ \frac{1}{2} \sum_i \left[ \nabla_i^2 \Psi^R(t) - \hat{\Omega} \Psi^I(t) \right] \right\}. \end{aligned} \tag{S5}$$

On differentiating Equation (S5) with respect  $r_{1\alpha}$ , where  $r_{1\alpha}$  is the  $\alpha$  coordinate of  $\mathbf{r}_1$ , and then multiplying both sides by  $[\Psi^R(t)]^2$ , we obtain

$$\begin{aligned} &\frac{\partial}{\partial r_{1\alpha}} \left[ v(\mathbf{y}_i) - \Phi(\mathbf{y}_i) + \frac{1}{2} \sum_{j=2} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} + \frac{1}{2} A^2(\mathbf{y}_i) \right. \\ &\quad \left. + \frac{1}{\Psi^R(t)} \frac{\partial}{\partial t} \Psi^I(t) \right] [\Psi^R(t)]^2 \\ &= \frac{1}{2} \sum_i \sum_{\beta} \left[ \Psi^R(t) \frac{\partial^3 \Psi^R(t)}{\partial r_{1\alpha} \partial r_{i\beta}^2} - \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \frac{\partial^2 \Psi^R(t)}{\partial r_{i\beta}^2} \right] \\ &\quad - \sum_i \sum_{\beta} \left[ \Psi^R(t) \frac{\partial}{\partial r_{1\alpha}} \left( A_{i\beta} \frac{\partial \Psi^I(t)}{\partial r_{i\beta}} \right) - A_{i\beta} \frac{\partial \Psi^I(t)}{\partial r_{i\beta}} \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \right] \\ &\quad - \frac{1}{2} \sum_i \sum_{\beta} \left[ \Psi^R(t) \frac{\partial}{\partial r_{1\alpha}} \left( \Psi^I(t) \frac{\partial A_{i\beta}}{\partial r_{i\beta}} \right) - \Psi^I(t) \frac{\partial A_{i\beta}}{\partial r_{i\beta}} \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \right]. \end{aligned} \tag{S6}$$

Now since

$$\begin{aligned} & \frac{1}{4} \frac{\partial^3 \Psi^R(t) \Psi^R(t)}{\partial r_{i\beta} \partial r_{i\beta} \partial r_{1\alpha}} \\ = & \frac{1}{2} \frac{\partial^2 \Psi^R(t)}{\partial r_{i\beta}^2} \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} + \frac{\partial \Psi^R(t)}{\partial r_{i\beta}} \frac{\partial^2 \Psi^R(t)}{\partial r_{i\beta} \partial r_{1\alpha}} + \frac{1}{2} \Psi^R(t) \frac{\partial^3 \Psi^R(t)}{\partial r_{i\beta} \partial r_{i\beta} \partial r_{1\alpha}}, \end{aligned} \tag{S7}$$

the first term of the right side in Equation (S6) can be written as

$$\begin{aligned} & \frac{1}{4} \frac{\partial^3 \Psi^R(t) \Psi^R(t)}{\partial r_{i\beta} \partial r_{i\beta} \partial r_{1\alpha}} - \frac{\partial}{\partial r_{i\beta}} \left[ \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \frac{\partial \Psi^R(t)}{\partial r_{i\beta}} \right] \\ = & \frac{1}{2} \Psi^R(t) \frac{\partial^3 \Psi^R(t)}{\partial r_{i\beta} \partial r_{i\beta} \partial r_{1\alpha}} - \frac{1}{2} \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \frac{\partial^2 \Psi^R(t)}{\partial r_{i\beta}^2}. \end{aligned} \tag{S8}$$

Thus, Equation (S6) is

$$\begin{aligned} & \frac{\partial}{\partial r_{1\alpha}} \left[ v(\mathbf{y}_1) - \Phi(\mathbf{y}_1) + \frac{1}{2} A^2(\mathbf{y}_1) + \frac{1}{2} \sum_{j=2} \frac{1}{|\mathbf{r}_1 - \mathbf{r}_j|} \right. \\ & \left. + \frac{1}{\Psi^R(t)} \frac{\partial}{\partial t} \Psi^I(t) \right] [\Psi^R(t)]^2 \\ = & \sum_i \sum_\beta \left[ \frac{1}{4} \frac{\partial^3 \Psi^R(t) \Psi^R(t)}{\partial r_{1\alpha} \partial^2 r_{i\beta}} - \frac{\partial}{\partial r_{i\beta}} \left( \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \frac{\partial \Psi^R(t)}{\partial r_{i\beta}} \right) \right] \\ & - \sum_i \sum_\beta \left[ \Psi^R(t) \frac{\partial}{\partial r_{1\alpha}} \left( A_{i\beta} \frac{\partial \Psi^I(t)}{\partial r_{i\beta}} \right) - A_{i\beta} \frac{\partial \Psi^I(t)}{\partial r_{i\beta}} \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \right] \\ & - \frac{1}{2} \sum_i \sum_\beta \left[ \Psi^R(t) \frac{\partial}{\partial r_{1\alpha}} \left( \Psi^I(t) \frac{\partial A_{i\beta}}{\partial r_{i\beta}} \right) - \Psi^I(t) \frac{\partial A_{i\beta}}{\partial r_{i\beta}} \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \right]. \end{aligned} \tag{S9}$$

Similarly we have the following equation for the imaginary part of the wave function  $\Psi^I(t)$ :

$$\begin{aligned} & \frac{\partial}{\partial r_{1\alpha}} \left[ v(\mathbf{y}_1) - \Phi(\mathbf{y}_1) + \frac{1}{2} A^2(\mathbf{y}_1) + \frac{1}{2} \sum_{j=2} \frac{1}{|\mathbf{r}_1 - \mathbf{r}_j|} \right. \\ & \left. + \frac{1}{\Psi^I(t)} \frac{\partial}{\partial t} \Psi^R(t) \right] [\Psi^I(t)]^2 \\ = & \sum_i \sum_\beta \left[ \frac{1}{4} \frac{\partial^3 \Psi^I(t) \Psi^I(t)}{\partial r_{1\alpha} \partial^2 r_{i\beta}} - \frac{\partial}{\partial r_{i\beta}} \left( \frac{\partial \Psi^I(t)}{\partial r_{1\alpha}} \frac{\partial \Psi^I(t)}{\partial r_{i\beta}} \right) \right] \\ & - \sum_i \sum_\beta \left[ \Psi^I(t) \frac{\partial}{\partial r_{1\alpha}} \left( A_{i\beta} \frac{\partial \Psi^R(t)}{\partial r_{i\beta}} \right) - A_{i\beta} \frac{\partial \Psi^R(t)}{\partial r_{i\beta}} \frac{\partial \Psi^I(t)}{\partial r_{1\alpha}} \right] \\ & - \frac{1}{2} \sum_i \sum_\beta \left[ \Psi^I(t) \frac{\partial}{\partial r_{1\alpha}} \left( \Psi^R(t) \frac{\partial A_{i\beta}}{\partial r_{i\beta}} \right) - \Psi^R(t) \frac{\partial A_{i\beta}}{\partial r_{i\beta}} \frac{\partial \Psi^I(t)}{\partial r_{1\alpha}} \right]. \end{aligned} \tag{S10}$$

Using the fact that

$$\begin{aligned} & \frac{\partial}{\partial r_{1\alpha}} \left( \frac{1}{\Psi^R(t)} \frac{\partial}{\partial t} \Psi^I(t) \right) [\Psi^R(t)]^2 - \frac{\partial}{\partial r_{1\alpha}} \left( \frac{1}{\Psi^I(t)} \frac{\partial}{\partial t} \Psi^R(t) \right) [\Psi^I(t)]^2 \\ = & \frac{\partial}{\partial t} \left( \Psi^R(t) \frac{\partial}{\partial r_{1\alpha}} \Psi^R(t) - \Psi^I(t) \frac{\partial}{\partial r_{1\alpha}} \Psi^I(t) \right), \end{aligned} \tag{S11}$$

Then on summing Equations (S9) and (S10) and operating by  $N \sum_{\sigma_1} \int d\mathbf{X}^{N-1}$  on the resulting equation, we have that

$$\begin{aligned}
 & N \sum_{\sigma_1} \int d\mathbf{X}^{N-1} \left[ \frac{\partial}{\partial r_{1\alpha}} (v(\mathbf{y}_1) - \Phi(\mathbf{y}_1)) \right. \\
 & \left. + \frac{1}{2} A^2(\mathbf{y}_1) + \frac{1}{2} \sum_{j=2} \frac{1}{|\mathbf{r}_1 - \mathbf{r}_j|} \right] |\Psi(t)|^2 \\
 & + N \sum_{\sigma_1} \int d\mathbf{X}^{N-1} \frac{\partial}{\partial t} \left( \Psi^R(t) \frac{\partial}{\partial r_{1\alpha}} \Psi^I(t) - \Psi^I(t) \frac{\partial}{\partial r_{1\alpha}} \Psi^R(t) \right) \\
 = & N \sum_{\sigma_1} \int d\mathbf{X}^{N-1} \sum_{\beta} \left[ \frac{1}{4} \frac{\partial^3 |\Psi(t)|^2}{\partial r_{1\alpha} \partial^2 r_{1\beta}} - \frac{\partial}{\partial r_{1\beta}} \left( \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \frac{\partial \Psi^R(t)}{\partial r_{1\beta}} + \frac{\partial \Psi^I(t)}{\partial r_{1\alpha}} \frac{\partial \Psi^I(t)}{\partial r_{1\beta}} \right) \right] \\
 & + N \sum_{\sigma_1} \int d\mathbf{X}^{N-1} \sum_{\beta} \left[ -\Psi^R(t) \frac{\partial}{\partial r_{1\alpha}} \left( A_{1\beta} \frac{\partial \Psi^I(t)}{\partial r_{1\beta}} \right) + A_{1\beta} \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \frac{\partial \Psi^I(t)}{\partial r_{1\beta}} \right. \\
 & \left. + \Psi^I(t) \frac{\partial}{\partial r_{1\alpha}} \left( A_{1\beta} \frac{\partial \Psi^R(t)}{\partial r_{1\beta}} \right) - A_{1\beta} \frac{\partial \Psi^I(t)}{\partial r_{1\alpha}} \frac{\partial \Psi^R(t)}{\partial r_{1\beta}} \right] \tag{S12} \\
 & + N \sum_{\sigma_1} \int d\mathbf{X}^{N-1} \sum_{\beta} \left[ -\Psi^R(t) \frac{\partial A_{1\beta}}{\partial r_{1\beta}} \frac{\partial \Psi^I(t)}{\partial r_{1\alpha}} + \Psi^I(t) \frac{\partial A_{1\beta}}{\partial r_{1\beta}} \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \right] \\
 & + N \sum_{\sigma_1} \int d\mathbf{X}^{N-1} \left\{ \sum_{j=2} \sum_{\beta} \left[ \frac{1}{4} \frac{\partial^3 |\Psi(t)|^2}{\partial r_{1\alpha} \partial^2 r_{j\beta}} - \frac{\partial}{\partial r_{j\beta}} \left( \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \frac{\partial \Psi^R(t)}{\partial r_{j\beta}} + \frac{\partial \Psi^I(t)}{\partial r_{1\alpha}} \frac{\partial \Psi^I(t)}{\partial r_{j\beta}} \right) \right] \right. \\
 & + \sum_{j=2} \sum_{\beta} \left[ -\Psi^R(t) \frac{\partial}{\partial r_{1\alpha}} \left( A_{j\beta} \frac{\partial \Psi^I(t)}{\partial r_{j\beta}} \right) + A_{j\beta} \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \frac{\partial \Psi^I(t)}{\partial r_{j\beta}} \right. \\
 & \left. + \Psi^I(t) \frac{\partial}{\partial r_{1\alpha}} \left( A_{j\beta} \frac{\partial \Psi^R(t)}{\partial r_{j\beta}} \right) - A_{j\beta} \frac{\partial \Psi^I(t)}{\partial r_{1\alpha}} \frac{\partial \Psi^R(t)}{\partial r_{j\beta}} \right] \\
 & \left. + \sum_{j=2} \sum_{\beta} \left[ -\Psi^R(t) \frac{\partial A_{j\beta}}{\partial r_{j\beta}} \frac{\partial \Psi^I(t)}{\partial r_{1\alpha}} + \Psi^I(t) \frac{\partial A_{j\beta}}{\partial r_{j\beta}} \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \right] \right\}.
 \end{aligned}$$

As the wave function and its derivative vanish at infinity, the last term of Equation (S12) vanishes on integration over  $dr_{j\beta}$ . The second and third terms of Equation (S12) may be rewritten as

$$\begin{aligned}
 & \sum_{\beta} \left[ -\Psi^R(t) \frac{\partial}{\partial r_{1\alpha}} \left( A_{1\beta} \frac{\partial \Psi^I(t)}{\partial r_{1\beta}} \right) + \Psi^I(t) \frac{\partial}{\partial r_{1\alpha}} \left( A_{1\beta} \frac{\partial \Psi^R(t)}{\partial r_{1\beta}} \right) \right. \\
 & + A_{1\beta} \left( \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \frac{\partial \Psi^I(t)}{\partial r_{1\beta}} - \frac{\partial \Psi^I(t)}{\partial r_{1\alpha}} \frac{\partial \Psi^R(t)}{\partial r_{1\beta}} \right) \\
 & \left. - \Psi^R(t) \frac{\partial A_{1\beta}}{\partial r_{1\beta}} \frac{\partial \Psi^I(t)}{\partial r_{1\alpha}} + \Psi^I(t) \frac{\partial A_{1\beta}}{\partial r_{1\beta}} \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} \right] \tag{S13} \\
 = & \sum_{\beta} \left\{ \frac{\partial A_{1\beta}}{\partial r_{1\alpha}} \left[ \Psi^I(t) \frac{\partial \Psi^R(t)}{\partial r_{1\beta}} - \Psi^R(t) \frac{\partial \Psi^I(t)}{\partial r_{1\beta}} \right] \right. \\
 & \left. + \frac{\partial}{\partial r_{1\beta}} \left[ A_{1\beta} \left( \Psi^I(t) \frac{\partial \Psi^R(t)}{\partial r_{1\alpha}} - \Psi^R(t) \frac{\partial \Psi^I(t)}{\partial r_{1\alpha}} \right) \right] \right\}.
 \end{aligned}$$

In turn Equation (S13) can be expressed in terms of the paramagnetic current density  $\mathbf{j}_p(\mathbf{y})$  as

$$k_{\alpha}(\mathbf{y}_j \mathbf{p} \mathbf{A}) = \sum_{\beta} \left[ \{ \nabla_{\alpha}(\mathbf{r}) A_{\beta}(\mathbf{y}) \} \mathbf{j}_{p\beta}(\mathbf{y}) + \nabla_{\beta} \{ A_{\beta}(\mathbf{y}) \mathbf{j}_{p\alpha}(\mathbf{y}) \} \right], \tag{S14}$$

where

$$\mathbf{j}_p(\mathbf{y}) = -\frac{i}{2} [\nabla_{\mathbf{r}'} - \nabla_{\mathbf{r}''}] \gamma(\mathbf{r} + \mathbf{r}'; \mathbf{r} + \mathbf{r}'', t) \Big|_{\mathbf{r}'=\mathbf{r}''=\mathbf{r}}, \quad (\text{S15})$$

and  $\gamma(\mathbf{r}\mathbf{r}'t)$  is the single-particle density matrix.

Finally, to address the first term of Equation (S12), we note that the kinetic “force”  $\mathbf{z}(\mathbf{y})$  is defined by its component as

$$z_\alpha(\mathbf{y}) = 2 \sum_\beta \frac{\partial}{\partial r_\beta} t_{\alpha\beta}(\mathbf{y}), \quad (\text{S16})$$

where  $t_{\alpha\beta}(\mathbf{y})$  is the kinetic energy density tensor defined in terms of the single-particle density matrix  $\gamma(\mathbf{r}\mathbf{r}'t)$  as

$$t_{\alpha\beta}(\mathbf{y}) = \frac{1}{4} \left[ \frac{\partial^2}{\partial r'_\alpha \partial r''_\beta} + \frac{\partial^2}{\partial r'_\beta \partial r''_\alpha} \right] \gamma(\mathbf{r}'\mathbf{r}''t) \Big|_{\mathbf{r}'=\mathbf{r}''=\mathbf{r}}. \quad (\text{S17})$$

On substituting the complex form of the wave function into the expression for  $\gamma(\mathbf{r}\mathbf{r}'t)$ , the tensor may be written as

$$t_{\alpha\beta}(\mathbf{y}) = \frac{N}{2} \sum_\sigma \int d\mathbf{X}^{N-1} \left[ \frac{\partial}{\partial r_\alpha} \Psi^R(\mathbf{r}\sigma, \mathbf{X}^{N-1}t) \frac{\partial}{\partial r_\beta} \Psi^R(\mathbf{r}\sigma, \mathbf{X}^{N-1}t) + \frac{\partial}{\partial r_\alpha} \Psi^I(\mathbf{r}\sigma, \mathbf{X}^{N-1}t) \frac{\partial}{\partial r_\beta} \Psi^I(\mathbf{r}\sigma, \mathbf{X}^{N-1}t) \right]. \quad (\text{S18})$$

Employing Equations (S14)–(S16), we obtain Equation (S12) in terms of the various “forces” to be

$$\begin{aligned} \rho(\mathbf{y}) \nabla [v(\mathbf{y}) + \frac{1}{2} A^2(\mathbf{y}) - \Phi(\mathbf{y})] - \mathbf{e}_{ee}(\mathbf{y}) + \mathbf{z}(\mathbf{y}) + \mathbf{d}(\mathbf{y}) \\ + \mathbf{k}(\mathbf{y}) \mathbf{j}_p \mathbf{A} + \frac{\partial}{\partial t} \mathbf{j}_p(\mathbf{y}) = 0, \end{aligned} \quad (\text{S19})$$

where the “forces”  $\mathbf{e}_{ee}(\mathbf{y})$ ,  $\mathbf{d}(\mathbf{y})$  are defined in the text.

In terms of the physical current density  $\mathbf{j}(\mathbf{y})$  which is

$$\mathbf{j}(\mathbf{y}) = \mathbf{j}_p(\mathbf{y}) + \rho(\mathbf{y}) \mathbf{A}(\mathbf{y}), \quad (\text{S20})$$

the terms  $k_\alpha(\mathbf{y}) \mathbf{j}_p \mathbf{A}$  may be written as

$$\begin{aligned} k_\alpha(\mathbf{y}) \mathbf{j}_p \mathbf{A} = k_\alpha(\mathbf{y}) \mathbf{j} \mathbf{A} - \sum_\beta \left[ \rho(\mathbf{y}) A_\beta \nabla_\alpha A_\beta + A_\beta \nabla_\beta (\rho(\mathbf{y}) A_\alpha) \right. \\ \left. + \rho(\mathbf{y}) A_\alpha \nabla_\beta A_\beta \right]. \end{aligned} \quad (\text{S21})$$

Additionally, since the continuity equation is

$$\nabla \cdot \mathbf{j}(\mathbf{y}) + \frac{\partial \rho(\mathbf{y})}{\partial t} = 0, \quad (\text{S22})$$

we have

$$\begin{aligned} \frac{\partial}{\partial t} \mathbf{j}_p(\mathbf{y}) &= \frac{\partial}{\partial t} [\mathbf{j}(\mathbf{y}) - \rho(\mathbf{y}) \mathbf{A}(\mathbf{y})] \\ &= \frac{\partial}{\partial t} \mathbf{j}(\mathbf{y}) + \mathbf{A}(\mathbf{y}) \nabla \cdot \mathbf{j}(\mathbf{y}) - \rho(\mathbf{y}) \frac{\partial \mathbf{A}(\mathbf{y})}{\partial t}. \end{aligned} \quad (\text{S23})$$

Employing Equations (S21) and (S23), Equation (S19) is

$$\begin{aligned} \rho(\mathbf{y}) [\nabla v(\mathbf{y}) + \mathbf{E}(\mathbf{y})] - \mathbf{e}_{ee}(\mathbf{y}) + \mathbf{z}(\mathbf{y}) + \mathbf{d}(\mathbf{y}) + \mathbf{A}(\mathbf{y}) \nabla \cdot \mathbf{j}(\mathbf{y}) \\ + \mathbf{k}(\mathbf{y} \mathbf{j} \mathbf{A}) - \sum_{\beta} \nabla_{\beta} [\rho(\mathbf{y}) \mathbf{A}(\mathbf{y}) A_{\beta}(\mathbf{y})] = 0, \end{aligned} \tag{S24}$$

with  $\mathbf{E}(\mathbf{y}) = -\nabla \Phi(\mathbf{y}) - \frac{\partial \mathbf{A}(\mathbf{y})}{\partial t}$ .

The last three terms of Equation (S24) contain the vector potential  $\mathbf{A}(\mathbf{r})$  and can be afforded a physical interpretation as the sum of the external Lorentz “force”  $\ell(\mathbf{y})$  and a contribution  $\mathbf{i}(\mathbf{y})$  to the internal “force”. The Lorentz “force” is

$$\ell(\mathbf{y}) = \mathbf{j}(\mathbf{y}) \times \mathbf{B}(\mathbf{y}), \tag{S25}$$

so that with  $\mathbf{B}(\mathbf{y}) = \nabla \times \mathbf{A}(\mathbf{y})$ , we have

$$\ell_{\alpha}(\mathbf{r}) = \sum_{\beta} [j_{\beta}(\mathbf{y}) \nabla_{\alpha} A_{\beta} - j_{\beta} \nabla_{\beta} A_{\alpha}]. \tag{S26}$$

The contribution of the magnetic field to the internal “force”  $\mathbf{i}(\mathbf{y})$  is defined by its components as

$$i_{\alpha}(\mathbf{y}) = \sum_{\beta} \nabla_{\beta} I_{\alpha\beta}(\mathbf{y}), \tag{S27}$$

where

$$I_{\alpha\beta}(\mathbf{y}) = [j_{\alpha}(\mathbf{y}) A_{\beta}(\mathbf{y}) + j_{\beta}(\mathbf{y}) A_{\alpha}(\mathbf{y})] - \rho(\mathbf{y}) A_{\alpha}(\mathbf{y}) A_{\beta}(\mathbf{y}). \tag{S28}$$

Hence, the sum

$$\begin{aligned} \ell_{\alpha}(\mathbf{y}) + i_{\alpha}(\mathbf{y}) = k_{\alpha}(\mathbf{y} \mathbf{j} \mathbf{A}) - \sum_{\beta} \nabla_{\beta} [\rho(\mathbf{y}) A_{\alpha}(\mathbf{y}) A_{\beta}(\mathbf{y})] \\ + \sum_{\beta} A_{\alpha}(\mathbf{y}) \nabla_{\beta} j_{\beta}(\mathbf{y}). \end{aligned} \tag{S29}$$

Thus, Equation (S24) may be written as the “Quantal Newtonian” second law of Equations (16)–(19).

For the model noninteracting fermion system in the same external field  $\mathcal{F}^{\text{ext}}(\mathbf{y})$  and possessing the same basic variables  $\{\rho(\mathbf{y}), \mathbf{j}(\mathbf{y})\}$ , a similar proof obtained by writing the orbitals as  $\phi_j(\mathbf{y}) = \phi_j^R(\mathbf{y}) + i\phi_j^I(\mathbf{y})$  then leads to the corresponding ‘Quantal Newtonian’ second law of Equations (22) and (23).