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

$$\hat{H}(t) = -\frac{1}{2}\sum_i \nabla_i^2 + \frac{1}{2}\sum_{i,j}{}' \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} + \sum_i \Big[v(\mathbf{y}_i) - \Phi(\mathbf{y}_i)\Big] + \sum_i \hat{\omega}\Big[\mathbf{y}_i; \mathbf{A}(\mathbf{y}_i)\Big], \quad \text{(S1)}$$

where

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

$$\hat{\Omega}(\mathbf{y}) = \frac{1}{2}\Big[\boldsymbol{\nabla}\cdot\mathbf{A}(\mathbf{y}) + 2\mathbf{A}(\mathbf{y})\cdot\boldsymbol{\nabla}\Big], \quad \text{(S3)}$$

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

$$\begin{aligned} i\frac{\partial\Psi^R(t)}{\partial t} - \frac{\partial\Psi^I(t)}{\partial t} = \Big\{ -\frac{1}{2}\sum_i \nabla_i^2 + \sum_i \Big[v(\mathbf{y}_i) - \Phi(\mathbf{y}_i)\Big] \\ + \frac{1}{2}\sum_{i,j}{}' \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} + \sum_i \hat{\omega}\Big[(\mathbf{y}_i)\mathbf{A}(\mathbf{y}_i)\Big]\Big\}\Big(\Psi^R(t) + i\Psi^I(t)\Big). \end{aligned} \quad \text{(S4)}$$

Equating the real parts of Equation (S4) yields

$$\begin{aligned} \sum_i \Big[v(\mathbf{y}_i) - \Phi(\mathbf{y}_i)\Big] + \frac{1}{2}\sum_{i,j}{}' \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} + \frac{1}{2}\sum_i A^2(\mathbf{y}_i) \\ + \frac{1}{\Psi^R(t)}\frac{\partial}{\partial t}\Psi^I(t) = \frac{1}{\Psi^R(t)}\Big\{\frac{1}{2}\sum_i \Big[\nabla_i^2\Psi^R(t) - \hat{\Omega}\Psi^I(t)\Big]\Big\}. \end{aligned} \quad \text{(S5)}$$

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

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

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

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

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

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

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}\Big[\frac{\partial}{\partial r_{1\alpha}}(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|})\Big]|\Psi(t)|^2 \\
& +N\sum_{\sigma_1}\int d\mathbf{X}^{N-1}\frac{\partial}{\partial t}\Big(\Psi^R(t)\frac{\partial}{\partial r_{1\alpha}}\Psi^I(t)-\Psi^I(t)\frac{\partial}{\partial r_{1\alpha}}\Psi^R(t)\Big) \\
= \; & N\sum_{\sigma_1}\int d\mathbf{X}^{N-1}\sum_{\beta}\Big[\frac{1}{4}\frac{\partial^3|\Psi(t)|^2}{\partial r_{1\alpha}\partial^2 r_{1\beta}}-\frac{\partial}{\partial r_{1\beta}}\Big(\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}}\Big)\Big] \\
& +N\sum_{\sigma_1}\int d\mathbf{X}^{N-1}\sum_{\beta}\Big[-\Psi^R(t)\frac{\partial}{\partial r_{1\alpha}}(A_{1\beta}\frac{\partial\Psi^I(t)}{\partial r_{1\beta}})+A_{1\beta}\frac{\partial\Psi^R(t)}{\partial r_{1\alpha}}\frac{\partial\Psi^I(t)}{\partial r_{1\beta}} \\
& +\Psi^I(t)\frac{\partial}{\partial r_{1\alpha}}(A_{1\beta}\frac{\partial\Psi^R(t)}{\partial r_{1\beta}})-A_{1\beta}\frac{\partial\Psi^I(t)}{\partial r_{1\alpha}}\frac{\partial\Psi^R(t)}{\partial r_{1\beta}}\Big] \\
& +N\sum_{\sigma_1}\int d\mathbf{X}^{N-1}\sum_{\beta}\Big[-\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}}\Big] \\
& +N\sum_{\sigma_1}\int d\mathbf{X}^{N-1}\Big\{\sum_{j=2}\sum_{\beta}\Big[\frac{1}{4}\frac{\partial^3|\Psi(t)|^2}{\partial r_{1\alpha}\partial^2 r_{j\beta}}-\frac{\partial}{\partial r_{j\beta}}\Big(\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}}\Big)\Big] \\
& +\sum_{j=2}\sum_{\beta}\Big[-\Psi^R(t)\frac{\partial}{\partial r_{1\alpha}}(A_{i\beta}\frac{\partial\Psi^I(t)}{\partial r_{j\beta}})+A_{i\beta}\frac{\partial\Psi^R(t)}{\partial r_{1\alpha}}\frac{\partial\Psi^I(t)}{\partial r_{j\beta}} \\
& +\Psi^I(t)\frac{\partial}{\partial r_{1\alpha}}(A_{j\beta}\frac{\partial\Psi^R(t)}{\partial r_{j\beta}})-A_{j\beta}\frac{\partial\Psi^I(t)}{\partial r_{1\alpha}}\frac{\partial\Psi^R(t)}{\partial r_{j\beta}}\Big] \\
& +\sum_{j=2}\sum_{\beta}\Big[-\Psi^R(t)\frac{\partial A_{i\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}}\Big]\Big\}.
\end{aligned} \tag{S12}$$

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

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

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

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

where

$$\mathbf{j}_p(\mathbf{y}) = -\frac{i}{2}\left[\boldsymbol{\nabla}_{\mathbf{r}'} - \boldsymbol{\nabla}_{\mathbf{r}''}\right]\gamma(\mathbf{r}+\mathbf{r}';\mathbf{r}+\mathbf{r}'',t)\bigg|_{\mathbf{r}'=\mathbf{r}''=0}, \tag{S15}$$

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

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

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

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

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

$$\begin{aligned} t_{\alpha\beta}(\mathbf{y}) = \frac{N}{2}\sum_\sigma \int d\mathbf{X}^{N-1}\bigg[&\frac{\partial}{\partial r_\alpha}\Psi^R(\mathbf{r}\sigma,\mathbf{X}^{N-1}t)\frac{\partial}{\partial r_\beta}\Psi^R(\mathbf{r}\sigma,\mathbf{X}^{N-1}t) \\ &+\frac{\partial}{\partial r_\alpha}\Psi^I(\mathbf{r}\sigma,\mathbf{X}^{N-1}t)\frac{\partial}{\partial r_\beta}\Psi^I(\mathbf{r}\sigma,\mathbf{X}^{N-1}t)\bigg]. \end{aligned} \tag{S18}$$

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

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

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

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

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

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

Additionally, since the continuity equation is

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

we have

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

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

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

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

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'' $\boldsymbol{\ell}(\mathbf{y})$ and a contribution $\mathbf{i}(\mathbf{y})$ to the internal ''force''. The Lorentz ''force'' is

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

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

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

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

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

Hence, the sum

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

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 $\boldsymbol{\mathcal{F}}^{\mathrm{ext}}(\mathbf{y})$ 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).