Extended Field Solver

6/25/20

- Goal: Extend the scope of auxiliary-field Poisson solver [Car & Parrinello, SSC 62, 403 ('87)] to include wave equations for vector potential, in the framework of Maxwell-TDDFT (time-dependent density functional theory) approach [Yabana, PRB 85, 045134 ('12)] in the Lorenz gauge [Gabay, PRB 101, 235101 ('20)].

- Maxwell equations

$$\begin{cases} \nabla \times \mathbb{E} + \dfrac{1}{c}\dfrac{\partial \mathbb{B}}{\partial t} = 0 & (1) \\ \nabla \times \mathbb{B} - \dfrac{1}{c}\dfrac{\partial \mathbb{E}}{\partial t} = \dfrac{4\pi}{c}\mathbb{J} & (2) \\ \nabla \cdot \mathbb{E} = 4\pi\rho & (3) \\ \nabla \cdot \mathbb{B} = 0 & (4) \end{cases}$$

where $\mathbb{E}$ & $\mathbb{B}$ are electric & magnetic fields, while charge (ρ) & current ($\mathbb{J}$) densities satisfies continuity equation,

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \mathbb{J} = 0 \tag{5}$$

– Vector & scalar potentials

Since the magnetic field is divergence-free (Eq.(4)), it can be represented as the curl of a vector field. Thus, we define vector potential $\mathbb{A}$ through

$$\mathbb{B} = \nabla \times \mathbb{A} \tag{6}$$

Substituting Eq.(6) in (1), we obtain

$$\nabla \times \left(\mathbb{E} + \frac{1}{c} \frac{\partial \mathbb{A}}{\partial t} \right) = 0 \tag{7}$$

Since $\mathbb{E} + c^{-1} \partial \mathbb{A} / \partial t$ is curl-free, it can be represented as the gradient of a scalar field. Thus, we define scalar potential ϕ through

$$\mathbb{E} + \frac{1}{c} \frac{\partial \mathbb{A}}{\partial t} = -\nabla \phi \tag{8}$$

or

$$\mathbb{E} = -\frac{1}{c} \frac{\partial \mathbb{A}}{\partial t} - \nabla \phi \tag{9}$$

– Wave equations for potentials

We have used the source-free equations, (1) & (4), to define vector & scalar potentials. We now use the rest, (2) & (3), to derive partial differential equations for vector & scalar potentials.

Substituting Eqs. (6) & (9) to (2),

$$\underbrace{\nabla\times\nabla\times}_{-\nabla^2+\nabla\nabla\cdot}\mathbb{A} - \frac{1}{c}\frac{\partial}{\partial t}\left[-\frac{1}{c}\frac{\partial\mathbb{A}}{\partial t} - \nabla\phi\right] = \frac{4\pi}{c}\mathbb{J}$$

$$-\nabla^2\mathbb{A} + \nabla\nabla\cdot\mathbb{A} + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathbb{A} + \frac{1}{c}\nabla\frac{\partial\phi}{\partial t} = \frac{4\pi}{c}\mathbb{J}$$

$$\therefore \left[\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \nabla^2 + \nabla\nabla\cdot\right]\mathbb{A} + \frac{1}{c}\nabla\frac{\partial\phi}{\partial t} = \frac{4\pi}{c}\mathbb{J} \qquad (10)$$

Substituting Eq. (9) in (3),

$$\nabla\cdot\left[-\frac{1}{c}\frac{\partial\mathbb{A}}{\partial t} - \nabla\phi\right] = 4\pi\rho$$

$$-\nabla^2\phi - \frac{1}{c}\frac{\partial}{\partial t}\nabla\cdot\mathbb{A} = 4\pi\rho \qquad (11)$$

– Lorenz gauge

Equations (10) & (11) constitute four equations to determine four unknown quantities (A_x, A_y, A_z, ϕ) from four known quantities (J_x, J_y, J_z, ρ). However, $\mathbb{J}$ & ρ are not independent but are related by continuity equation (5). Accordingly, we need to introduce one more condition (i.e., gauge condition) to uniquely determine $\mathbb{A}$ & ϕ.

We here adopt the Lorenz gauge

$$\frac{1}{c}\frac{\partial}{\partial t}\phi + \nabla\cdot\mathbb{A} = 0 \tag{12}$$

Taking gradient of Eq. (12),

$$\frac{1}{c}\nabla\frac{\partial\phi}{\partial t} + \nabla\nabla\cdot\mathbb{A} = 0 \tag{13}$$

Using Eq. (13) to eliminate ϕ from Eq. (10), we obtain

$$\left[\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \nabla^2 + \cancel{\nabla\nabla\cdot}\right]\mathbb{A} - \cancel{\nabla\nabla\cdot\mathbb{A}} = \frac{4\pi}{c}\mathbb{J}$$

$$\therefore \quad \left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \nabla^2\right)\mathbb{A} = \frac{4\pi}{c}\mathbb{J} \tag{14}$$

Also, using Eq. (12) to eliminate $\mathbb{A}$ in (11), we obtain

$$-\nabla^2\phi - \frac{1}{c}\frac{\partial}{\partial t}\left(-\frac{1}{c}\frac{\partial}{\partial t}\phi\right) = 4\pi\rho$$

$$\therefore \quad \left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \nabla^2\right)\phi = 4\pi\rho \tag{15}$$

In summary, both vector & scalar potentials follow wave equations in Lorenz gauge.

$$\boxed{\begin{cases} \left(\dfrac{1}{c^2}\dfrac{\partial^2}{\partial t^2} - \nabla^2\right)\mathbb{A} = \dfrac{4\pi}{c}\mathbb{J} & (16) \\ \left(\dfrac{1}{c^2}\dfrac{\partial^2}{\partial t^2} - \nabla^2\right)\phi = 4\pi\rho & (17) \end{cases}}$$

– Single-electron Hamiltonian

(Classical Hamiltonian)

$$H(t) = \frac{1}{2m}\left(\mathbb{P} + \frac{e}{c}\mathbb{A}(\mathbb{r},t)\right)^2 - e\phi(\mathbb{r},t) \qquad (18)$$

where m & e are mass & (absolute) charge of electron, and $\mathbb{r}$ & $\mathbb{P}$ are its position & momentum.

(Hamiltonian operator)

$$\hat{H}(t) = \frac{\hat{\mathbb{P}}^2}{2m} + \frac{e}{2mc}\left[\hat{\mathbb{P}}\cdot\mathbb{A}(\mathbb{r},t) + \mathbb{A}(\mathbb{r},t)\cdot\hat{\mathbb{P}}\right] + \frac{e^2}{2mc^2}\mathbb{A}^2(\mathbb{r},t) - e\phi(\mathbb{r},t) \qquad (19)$$

- Current operator

In second-quantization,

$$\hat{H}(t) = \sum_\sigma \int d\mathbf{r}\, \hat{\psi}^\dagger_\sigma(\mathbf{r}) \Big\{ -\frac{\hbar^2}{2m}\nabla^2 + \frac{e}{2mc}\Big[\underset{\uparrow \text{ integration by parts}}{\frac{\hbar}{i}\nabla \mathbf{A}(\mathbf{r},t)} + \mathbf{A}(\mathbf{r},t)\frac{\hbar}{i}\nabla\Big]$$

$$+ \frac{e^2}{2mc^2}\mathbf{A}^2(\mathbf{r},t) - e\phi(\mathbf{r},t)\Big\}\hat{\psi}_\sigma(\mathbf{r}) \qquad (20)$$

$$= \sum_\sigma \int d\mathbf{r}\, \hat{\psi}^\dagger_\sigma(\mathbf{r}) \Big(-\frac{\hbar^2}{2m}\nabla^2\Big)\hat{\psi}_\sigma(\mathbf{r})$$

$$+ \frac{e}{2mc}\sum_\sigma \int d\mathbf{r}\, \mathbf{A}(\mathbf{r},t)\Big\{\hat{\psi}^\dagger_\sigma(\mathbf{r})\frac{\hbar}{i}\nabla\hat{\psi}_\sigma(\mathbf{r}) - \Big(\frac{\hbar}{i}\nabla\hat{\psi}^\dagger_\sigma(\mathbf{r})\Big)\hat{\psi}_\sigma(\mathbf{r})$$

$$+ \frac{e}{c}\mathbf{A}(\mathbf{r},t)\hat{\psi}^\dagger_\sigma(\mathbf{r})\hat{\psi}_\sigma(\mathbf{r})\Big\}$$

$$- e\sum_\sigma \int d\mathbf{r}\, \hat{\psi}^\dagger_\sigma(\mathbf{r})\phi(\mathbf{r},t)\hat{\psi}_\sigma(\mathbf{r})$$

$$= \sum_\sigma \int d\mathbf{r}\, \hat{\psi}^\dagger_\sigma(\mathbf{r}) \Big(-\frac{\hbar^2}{2m}\nabla^2\Big)\hat{\psi}_\sigma(\mathbf{r})$$

$$- \frac{1}{c}\sum_\sigma \int d\mathbf{r}\, \mathbf{A}(\mathbf{r},t) \times \Big(-\frac{e}{2m}\Big)\Big\{\hat{\psi}^\dagger_\sigma(\mathbf{r})\frac{\hbar}{i}\nabla\hat{\psi}_\sigma(\mathbf{r}) - \Big(\frac{\hbar}{i}\nabla\hat{\psi}^\dagger_\sigma(\mathbf{r})\Big)\hat{\psi}_\sigma(\mathbf{r})$$

$$+ \frac{e}{c}\mathbf{A}(\mathbf{r},t)\hat{\psi}^\dagger_\sigma(\mathbf{r})\hat{\psi}_\sigma(\mathbf{r})\Big\}$$

$$- e\int d\mathbf{r}\, \phi(\mathbf{r},t)\sum_\sigma \hat{\psi}^\dagger_\sigma(\mathbf{r})\hat{\psi}_\sigma(\mathbf{r})$$

In summary,

$$\hat{H}(t) = \sum_\sigma \int d\mathbf{r}\, \hat{\psi}^\dagger_\sigma(\mathbf{r}) \left(-\frac{\hbar^2}{2m}\nabla^2\right) \hat{\psi}_\sigma(\mathbf{r})$$

$$- \frac{1}{c}\int d\mathbf{r}\, \mathbf{A}(\mathbf{r},t)\left[\hat{\mathbf{j}}_p(\mathbf{r}) + \frac{1}{2}\hat{\mathbf{j}}_d(\mathbf{r})\right]$$

$$+ \int d\mathbf{r}\, \phi(\mathbf{r},t)\, \hat{\rho}(\mathbf{r}) \qquad (21)$$

Here, the current operator $\hat{\mathbf{j}}(\mathbf{r})$ is

$$\hat{\mathbf{j}}(\mathbf{r}) = -\frac{e}{2m}\sum_\sigma \left[\hat{\psi}^\dagger_\sigma(\mathbf{r})\frac{\hbar}{i}\nabla\hat{\psi}_\sigma(\mathbf{r}) - \left(\frac{\hbar}{i}\nabla\hat{\psi}^\dagger_\sigma(\mathbf{r})\right)\hat{\psi}_\sigma(\mathbf{r})\right]$$

$$+ \frac{e^2}{mc}\mathbf{A}(\mathbf{r},t)\,\hat{\rho}(\mathbf{r}) \qquad (22)$$

$$= \hat{\mathbf{j}}_p(\mathbf{r}) + \hat{\mathbf{j}}_d(\mathbf{r}) \qquad (23)$$

and the charge density operator $\hat{\rho}(\mathbf{r})$ is

$$\hat{\rho}(\mathbf{r}) = -e\sum_\sigma \hat{\psi}^\dagger_\sigma(\mathbf{r})\,\hat{\psi}_\sigma(\mathbf{r}) \qquad (24)$$

In Eq. (23), $\hat{\mathbf{j}}_p$ & $\hat{\mathbf{j}}_d$ are paramagnetic & diamagnetic current operators, respectively.

- Continuity equation

In Heisenberg picture, equation of motion of charge density operator is

$$i\hbar \frac{\partial}{\partial t} \hat{\rho}_H(\mathbf{r},t) = [\hat{\rho}_H(\mathbf{r}), \hat{H}] \tag{25}$$

Note the Hamiltonian terms involving diamagnetic current & scalar potential are proportional to the density operator, thus their commutator with $\hat{\rho}$ vanishes. The remaining terms containing kinetic operator & paramagnetic current give rise to:

$$i\hbar \frac{\partial}{\partial t} \hat{\rho}_H(\mathbf{r}) = -e \sum_\sigma \int dx \, [\hat{\psi}^\dagger_\sigma(\mathbf{r}) \hat{\psi}_\sigma(\mathbf{r}), \hat{\psi}^\dagger_\sigma(x) \left(-\frac{\hbar^2}{2m}\nabla_x^2\right) \hat{\psi}_\sigma(x)]$$

$$+ \frac{e}{c} \sum_\sigma \int dx \, \mathbb{A}(x) \, [\hat{\psi}^\dagger_\sigma(\mathbf{r}) \hat{\psi}_\sigma(\mathbf{r}), \hat{\mathbf{j}}_p(x)] \tag{26}$$

In the first term in rhs, note

$$[\hat{\psi}_\sigma^+(\mathbf{r})\hat{\psi}_\sigma(\mathbf{r}), \hat{\psi}_\sigma^+(x)\left(-\frac{\nabla_x^2}{2m}\right)\hat{\psi}_\sigma(x)]$$

$$= \underbrace{r^+ r x^+}_{\delta(x-r) - x^+ r}\left(-\frac{\nabla_x^2}{2}\right)x - x^+\left(-\frac{\nabla_x^2}{2}\right)\underbrace{x r^+}_{\delta(x-r) - r^+ x} r$$

$$= \delta(x-r)\, r^+\left(-\frac{\nabla_r^2}{2}\right)r - \cancel{r^+ x^+ r\left(-\frac{\nabla_x^2}{2}\right)x}$$

$$- x^+\left(-\frac{\nabla_x^2}{2}\right)\delta(x-r)\, r + \cancel{\underbrace{x^+ r^+}_{-r^+ x^+}\left(-\frac{\nabla_x^2}{2}\right)\underbrace{x r}_{-r x}} \sim \mathcal{a}$$

Here, note the integration by parts:

$$\int dx\, x^+ \nabla_x^2 \delta(x-r)\, r$$

$$= -\int dx\, \nabla_x x^+ \cdot \nabla_x \delta(x-r)\, r$$

$$= +\int dx \left(\nabla_x^2 x^+\right)\delta(x-r)\, r$$

$$= \left(\nabla_r^2 r^+\right) r$$

Thus,

$$\mathcal{a} \sim \delta(x-r)\, r^+\left(-\frac{\nabla_r^2}{2}\right)r - \delta(x-r)\left(-\frac{\nabla_r^2}{2}r^+\right)r$$

Namely, the first term of Eq.(26) yields

$$-e\sum_\sigma\left[\hat{\psi}^+_\sigma(\mathbf{r})\left(-\frac{\hbar^2}{2m}\nabla^2\right)\hat{\psi}_\sigma(\mathbf{r})-\left(-\frac{\hbar^2}{2m}\nabla^2\hat{\psi}^+_\sigma(\mathbf{r})\right)\hat{\psi}_\sigma(\mathbf{r})\right]$$

$$=\frac{e\hbar^2}{2m}\sum_\sigma\underbrace{\left[\hat{\psi}^+_\sigma(\mathbf{r})\nabla^2\hat{\psi}_\sigma(\mathbf{r})-(\nabla^2\hat{\psi}^+_\sigma(\mathbf{r}))\hat{\psi}_\sigma(\mathbf{r})\right]}_{\nabla\cdot(r^+\nabla r)-\cancel{\nabla r^+\cdot\nabla r}-\{\nabla\cdot[(\nabla r^+)r]-\cancel{\nabla r^+\cdot\nabla r}\}}$$

$$=\frac{e\hbar^2}{2m}\sum_\sigma\nabla\cdot\left[\hat{\psi}^+_\sigma(\mathbf{r})\nabla\hat{\psi}_\sigma(\mathbf{r})-(\nabla\hat{\psi}^+_\sigma(\mathbf{r}))\hat{\psi}_\sigma(\mathbf{r})\right] \quad -\beta$$

On the other hand, the second term of Eq.(26) is

$$-\frac{e^2}{2mc}\sum_\sigma\int dx\,\mathbb{A}(x)\left[\hat{\psi}^+_\sigma(\mathbf{r})\hat{\psi}_\sigma(\mathbf{r}),\ \hat{\psi}^+_\sigma(x)\frac{\hbar}{i}\nabla_x\hat{\psi}_\sigma(x)\right.$$
$$\left.-\left(\frac{\hbar}{i}\nabla_x\hat{\psi}^+_\sigma(x)\right)\hat{\psi}_\sigma(x)\right] \quad -\gamma$$

Note

$$[r^+r,\ x^+\tfrac{\hbar}{i}\nabla_x x]-[r^+r,\ (\tfrac{\hbar}{i}\nabla_x x^+)x]$$

$$=r^+\underbrace{r\,x^+}_{\delta(x-r)}\tfrac{\hbar}{i}\nabla_x x-x^+\tfrac{\hbar}{i}\nabla_x\underbrace{x\,r^+}_{\delta(x-r)}r-r^+\underbrace{r(\tfrac{\hbar}{i}\nabla_x x^+)}_{\delta(x-r)}x+(\tfrac{\hbar}{i}\nabla_x x^+)\underbrace{x\,r^+}_{\delta(x-r)}r$$

$$\therefore \gamma = -\frac{e^2\hbar}{2mci}\sum_\sigma \Big\{ \overbrace{A(r)\psi_\sigma^\dagger(r)\nabla\psi_\sigma(r) + \nabla[A(r)\psi_\sigma^\dagger(r)]\psi_\sigma(r)}^{\nabla\cdot(A\psi^\dagger\psi)}$$

$$+ \underbrace{\psi_\sigma^\dagger(r)\nabla[A(r)\psi_\sigma(r)] + A(r)(\nabla\psi^\dagger(r))\psi(r)}_{\nabla\cdot(A\psi^\dagger\psi)}$$

$$= -\frac{e^2\hbar}{mci}\sum_\sigma \nabla\cdot\left[\mathbb{A}(\mathbb{r})\hat{\psi}_\sigma^\dagger(\mathbb{r})\hat{\psi}_\sigma(\mathbb{r})\right] \qquad \sim \delta$$

Using expressions β & δ in Eq. (26),

$$i\cancel{\hbar}\frac{\partial}{\partial t}\hat{\rho}_H(\mathbb{r}) = \nabla\cdot\Big\{\frac{e\hbar^{\cancel{2}}}{2m}\sum_\sigma\left[\hat{\psi}_\sigma^\dagger(\mathbb{r})\nabla\hat{\psi}_\sigma(\mathbb{r}) - (\nabla\hat{\psi}_\sigma^\dagger(\mathbb{r}))\psi_\sigma(\mathbb{r})\right]$$

$$-\frac{e^2\cancel{\hbar}}{mci}\sum_\sigma\left[\mathbb{A}(\mathbb{r})\hat{\psi}_\sigma^\dagger(\mathbb{r})\hat{\psi}_\sigma(\mathbb{r})\right]\Big\}$$

$$\therefore \frac{\partial}{\partial t}\hat{\rho}_H(\mathbb{r}) = \nabla\cdot\Big\{\overbrace{\frac{e}{2m}\sum_\sigma\left[\hat{\psi}_\sigma^\dagger(\mathbb{r})\frac{\hbar}{i}\nabla\hat{\psi}_\sigma(\mathbb{r}) - \left(\frac{\hbar}{i}\nabla\hat{\psi}_\sigma^\dagger(\mathbb{r})\right)\hat{\psi}_\sigma(\mathbb{r})\right]}^{-\hat{\mathbb{J}}_p(\mathbb{r})}$$

$$+ \underbrace{\frac{e^2}{mc}\mathbb{A}(\mathbb{r})\sum_\sigma\hat{\psi}_\sigma^\dagger(\mathbb{r})\hat{\psi}_\sigma(\mathbb{r})}_{-\frac{e}{mc}\mathbb{A}(\mathbb{r})\hat{\rho}(\mathbb{r}) = -\hat{\mathbb{J}}_d(\mathbb{r})}\Big\}$$

Namely,

$$\frac{\partial}{\partial t}\hat{\rho}_H(\mathbb{r},t) + \nabla\cdot\Big[\underbrace{\hat{\mathbb{J}}_p(\mathbb{r}) + \hat{\mathbb{J}}_d(\mathbb{r})}_{\hat{\mathbb{J}}(\mathbb{r})}\Big] = 0$$

In summary

$$\boxed{\begin{aligned}
&\frac{\partial}{\partial t}\hat{\rho}_H(\mathbf{r},t) + \nabla\cdot\hat{\mathbf{J}}_H(\mathbf{r},t) = 0 && (27)\\
&\begin{cases}
\hat{\rho}(\mathbf{r}) = -e\sum_\sigma \hat{\psi}^\dagger_\sigma(\mathbf{r})\,\hat{\psi}_\sigma(\mathbf{r}) & (24)\\
\hat{\mathbf{J}}(\mathbf{r}) = -\frac{e}{2m}\sum_\sigma\left[\hat{\psi}^\dagger_\sigma(\mathbf{r})\,\frac{\hbar}{i}\nabla\hat{\psi}_\sigma(\mathbf{r}) - \left(\frac{\hbar}{i}\nabla\hat{\psi}^\dagger_\sigma(\mathbf{r})\right)\hat{\psi}_\sigma(\mathbf{r})\right] & \\
\qquad\qquad + \frac{e}{mc}\mathbf{A}(\mathbf{r},t)\,\hat{\rho}(\mathbf{r}) & (22)\\
\qquad = \hat{\mathbf{J}}_p(\mathbf{r}) + \hat{\mathbf{J}}_d(\mathbf{r}) & (23)
\end{cases}
\end{aligned}}$$

— Multiscale field solvers

We consider a spatially-uniform laser pulse & embedding electric field as an external vector potential [6/18/20]

$$\tilde{\mathbb{A}}_{ext}(t) = -\frac{1}{c}\mathbb{A}_{ext}(t)$$

$$= \frac{\mathbb{D}_{ext}}{\omega_{ext}}\sin(\omega_{ext}t - \varphi_{ext}) \times \sin^2\left(\frac{\pi t}{T_{0ext}}\right)\Theta(0<t<T_0)$$

$$+ \mathbb{D}_{emb}t \tag{28}$$

The total vector potential $\mathbb{A}(t)$ is a sum of external & induced terms:

$$\mathbb{A}(t) = \mathbb{A}_{ext}(t) + \mathbb{A}_{ind}(t) \tag{29}$$

Following the multiscale Maxwell-TDDFT approach, we take $\mathbb{A}_{ind}(t)$ to be a smoothly varying function in space. We consider it to be constant, at least within a divide-&-conquer (DC) domain.

Therefore, Eq. (16) becomes

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \cancel{\nabla^2}\right) \mathbb{A}_{ind} = \frac{4\pi}{c} \mathbb{J}_{avg} \tag{30}$$

Here, $\mathbb{J}_{avg}$ is spatially-averaged current,

$$\mathbb{J}_{avg} = \frac{1}{\Omega}\int_{\Omega} d\mathbb{r}\, \mathbb{j}(\mathbb{r}) \tag{31}$$

and the current density in the Kohn-Sham (KS) scheme is

$$\mathbb{j}(\mathbb{r}) = -\frac{e}{2m}\sum_{n\sigma}\left[\psi^*_{n\sigma}(\mathbb{r})\frac{\hbar}{i}\nabla\psi_{n\sigma}(\mathbb{r}) - \left(\frac{\hbar}{i}\nabla\psi^*_{n\sigma}(\mathbb{r})\right)\psi_{n\sigma}(\mathbb{r})\right] f_{n\sigma}$$

$$+ \frac{e}{mc}\sum_{n\sigma} \mathbb{A}(\mathbb{r},t)\left\{-e\sum_{n\sigma}|\psi_{n\sigma}(\mathbb{r})|^2 f_{n\sigma}\right\}$$

$$\therefore\ \mathbb{j}(\mathbb{r}) = -\frac{e}{m}\sum_{n\sigma} \mathrm{Re}\left[\psi^*_{n\sigma}(\mathbb{r})\frac{\hbar}{i}\nabla\psi_{n\sigma}(\mathbb{r})\right] f_{n\sigma}$$

$$-\frac{e^2}{mc}\mathbb{A}(\mathbb{r},t)\underbrace{\sum_{n\sigma}|\psi_{n\sigma}(\mathbb{r})|^2 f_{n\sigma}}_{\text{electron number density } n(\mathbb{r})} \tag{32}$$

where $f_{n\sigma}$ is the occupation number.

We introduce

$$\tilde{\mathbb{A}}_{ind}(t) = -\frac{1}{c}\mathbb{A}_{ind}(t) \qquad (33)$$

and its field equation becomes

$$\boxed{\begin{aligned} &\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\tilde{\mathbb{A}}_{ind} = -\frac{4\pi}{c^2}\mathbb{J}_{avg} \qquad (34) \\ &\text{where} \\ &\mathbb{J}_{avg} = \frac{1}{\Omega}\int_{\Omega} d\mathbb{r}\, \mathbb{j}(\mathbb{r}) \qquad (31) \\ &\mathbb{j}(\mathbb{r}) = -\frac{e}{m}\sum_{n\sigma} \mathrm{Re}\left[\psi^*_{n\sigma}(\mathbb{r})\frac{\hbar}{i}\nabla\psi_{n\sigma}(\mathbb{r})\right] f_{n\sigma} \\ &\qquad\qquad + \frac{e^2}{m}\tilde{\mathbb{A}}(\mathbb{r},t)\, n(\mathbb{r}) \qquad (35) \\ &n(\mathbb{r}) = \sum_{n\sigma} |\psi_{n\sigma}(\mathbb{r})|^2 f_{n\sigma} \qquad (36) \end{aligned}}$$

For induced scalar potential, we instead consider Hartree potential

$$v_H(\mathbf{r},t) = -e\,\phi(\mathbf{r},t) \tag{37}$$

and it follows, instead of Eq. (17),

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \nabla^2\right) v_H = -4\pi e\rho = 4\pi e^2 n$$

$$\boxed{\therefore \left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \nabla^2\right) v_H(\mathbf{r},t) = 4\pi e^2 n(\mathbf{r},t) \qquad (38)}$$

※ In Eqs. (35) & (38), electron number density in LFD is interpreted as the deviation of density from time $t=0$ [6/13/20].