The Liouville equation for high harmonic generation in solid: the electromagnetic gauge dependence and the Bloch/Wannier representation

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(Dated: 25 October 2017)

Abstract

We provide a derivation of of the Liouville equation in various representations and gauges .

I. INTRODUCTION

Let $\hat{\rho}$ be the reduced density operator and $\hat{H}$ be the hamiltonian of the system, then the time development of the system is calculated by the Liouville equation, or the von Neumann equation.

$$i\hbar\frac{\partial\hat{\rho}}{\partial t} = [\hat{H}, \hat{\rho}] \tag{1}$$

The righthand side of the second line corresponds the anti-Hermite component of the $\hat{H}\hat{\rho}/\hbar$.

Let $|\mathbf{m}>$ be the time-independent orthonormal basis satisfying the complete relation $\sum_{\mathbf{m}} |\mathbf{m}><\mathbf{m}| = \mathbf{1}$. Then we have the matrix representation of the Liouville equation as follows.

$$\begin{aligned} i\hbar\frac{\partial}{\partial t} < \mathbf{m}|\hat{\rho}|\mathbf{m}' > &= < \mathbf{m}|\hat{H}\hat{\rho} - \hat{\rho}\hat{H}|\mathbf{m}' > \\ &= \sum_{\mathbf{m}''}(< \mathbf{m}|\hat{H}|\mathbf{m}'' >< \mathbf{m}''|\hat{\rho}|\mathbf{m}' > - < \mathbf{m}|\hat{\rho}|\mathbf{m}'' >< \mathbf{m}''|\hat{H}|\mathbf{m}' >). \end{aligned} \tag{2}$$

The Bloch states $\{|n\mathbf{k}>\}$ and Wannier states $\{|n\mathbf{R}>\}$ are the time-independent orthonormal basis. with the band index n and c, When we consider a system interacting with electromagnetic field, the hamiltonian of the system is

$$\begin{aligned} \hat{H} &= \frac{(\hat{\mathbf{p}} - e\mathbf{A})^2}{2m} - e\phi + \hat{V}(x) \\ &= H_0 + H_{int} \end{aligned} \tag{3}$$

where $\mathbf{A}$ is the vector potential, ϕ is the scalar potential, $\hat{H}_0 = \hat{\mathbf{p}}^2/2m + \hat{V}(x)$ is the free hamiltonian without external electric filed, and $H_{int} = (-2e\mathbf{A}\cdot\hat{\mathbf{p}} + e^2|\mathbf{A}|^2)/2m - e\phi$ is the interaction hamiltonian with electric field.

Considering the electromagnetic gauge freedom, the interaction hamiltonian is not uniqely determined (See Appendix). For convenience, we consider the length and velocity gauge from the Coulomb gauge (i.e. the divergence of the vector potential vanishes $\nabla\cdot\mathbf{A} = 0$). In the length gauge, the vector potential is set to be 0 ($\mathbf{A} = \mathbf{0}$). On the other hand, in the velocity gauge($\phi = 0$ The corresponding interaction hamiltonian of the length gauge $\hat{H}^L_{int}$ and that of the velocity gauge $\hat{H}^V_{int}$ are

$$\hat{H}^L_{int} = -e\mathbf{E}\cdot\hat{\mathbf{x}}, \tag{4}$$

$$\hat{H}^V_{int} = \frac{-2e\mathbf{A}\cdot\hat{\mathbf{p}} + e^2|\mathbf{A}|^2}{2m}. \tag{5}$$

where $\mathbf{A} = -\int \mathbf{E}dt$ is the vector potential determined by the external electric filed.

emitted field is proportional to electric current flowing through the solid The current is separated into two parts: the interband current and intraband current.[1] electric dipole is expressi

In this report, we calculated the spectra of HHG from solid by numerically solving the Liouville equation with four scheme: the Bloch representation with the length and velocity gauges (Chap. **??** and **??**, respectively), and the wannier representation with the length and velocity gauges (Chap. H and I, respectively)

II. RESULT

III. DISCUSSION

IV. CONCLUSION

Appendix A: Formulas

$$
\begin{aligned}
< n\mathbf{k}|\hat{\mathbf{x}}|n''\mathbf{k}'' > &= < n\mathbf{k}|\{-i\nabla_{\mathbf{k}''}e^{i\mathbf{k}''\cdot\hat{\mathbf{x}}}\}|u_{n''\mathbf{k}''} > \\
&= -i\nabla_{\mathbf{k}''} < n\mathbf{k}|n''\mathbf{k}'' > +i < u_{n\mathbf{k}}|e^{-i(\mathbf{k}-\mathbf{k}'')\cdot\hat{\mathbf{x}}}|\nabla_{\mathbf{k}''}u_{n''\mathbf{k}''} > \\
&= -i\nabla_{\mathbf{k}''} < n\mathbf{k}|n''\mathbf{k}'' > +\delta(\mathbf{k}-\mathbf{k}'')\mathbf{d}_{nn''}(\mathbf{k}) \qquad \text{(A1)} \\
< n''\mathbf{k}''|\hat{\mathbf{x}}|n'\mathbf{k}' > &= < u_{n''\mathbf{k}''}|\{i\nabla_{\mathbf{k}''}e^{-i\mathbf{k}''\cdot\hat{\mathbf{x}}}\}|n'\mathbf{k}' > \\
&= i\nabla_{\mathbf{k}''} < n''\mathbf{k}''|n'\mathbf{k}' > -i < \nabla_{\mathbf{k}''}u_{n''\mathbf{k}''}|e^{-i(\mathbf{k}''-\mathbf{k}')\cdot\hat{\mathbf{x}}}|u_{n'\mathbf{k}'} > \\
&= i\nabla_{\mathbf{k}''} < n''\mathbf{k}''|n'\mathbf{k}' > +\delta(\mathbf{k}''-\mathbf{k}')\mathbf{d}_{n''n'}(\mathbf{k}') \qquad \text{(A2)}
\end{aligned}
$$

$$
\begin{aligned}
i < u_{n\mathbf{k}}|e^{-i(\mathbf{k}-\mathbf{k}'')\cdot\hat{\mathbf{x}}}|\nabla_{\mathbf{k}''}u_{n''\mathbf{k}''} > &= i\int d\mathbf{x} < u_{n\mathbf{k}}|\mathbf{x}>< \mathbf{x}|\nabla_{\mathbf{k}''}u_{n''\mathbf{k}''} > e^{-i(\mathbf{k}-\mathbf{k}'')\cdot\mathbf{x}} \\
&= i\sum_{\mathbf{R}} e^{-i(\mathbf{k}-\mathbf{k}'')\cdot\mathbf{R}}\int_{BL} d\mathbf{y} < u_{n\mathbf{k}}|\mathbf{y}>< \mathbf{y}|\nabla_{\mathbf{k}''}u_{n''\mathbf{k}''} > e^{-i(\mathbf{k}-\mathbf{k}'')\cdot\mathbf{y}} \\
&= \delta(\mathbf{k}-\mathbf{k}'')(i\int_{BL} d\mathbf{y} < u_{n\mathbf{k}}|\mathbf{y}>< \mathbf{y}|\nabla_{\mathbf{k}''}u_{n''\mathbf{k}''} >) \\
&\equiv \delta(\mathbf{k}-\mathbf{k}'')\mathbf{d}_{nn''}(\mathbf{k}) \qquad \text{(A3)}
\end{aligned}
$$

$$
\begin{aligned}
i < \nabla_{\mathbf{k}''}u_{n''\mathbf{k}''}|e^{-i(\mathbf{k}''-\mathbf{k}')\cdot\hat{\mathbf{x}}}|u_{n'\mathbf{k}'} > &= i\int d\mathbf{x} < \nabla_{\mathbf{k}''}u_{n''\mathbf{k}''}|\mathbf{x}>< \mathbf{x}|u_{n'\mathbf{k}'} > e^{-i(\mathbf{k}''-\mathbf{k}')\cdot\mathbf{x}} \\
&= i\sum_{\mathbf{R}} e^{-i(\mathbf{k}''-\mathbf{k}')\cdot\mathbf{R}}\int_{BL} d\mathbf{y} < \nabla_{\mathbf{k}''}u_{n''\mathbf{k}''}|\mathbf{y}>< \mathbf{y}|u_{n'\mathbf{k}'} > e^{-i(\mathbf{k}''-\mathbf{k}')\cdot\mathbf{y}} \\
&= \delta(\mathbf{k}''-\mathbf{k}')(i\int_{BL} d\mathbf{y} < \nabla_{\mathbf{k}''}u_{n''\mathbf{k}''}|\mathbf{y}>< \mathbf{y}|u_{n'\mathbf{k}'} >) \\
&= -\delta(\mathbf{k}''-\mathbf{k}')(i\int_{BL} d\mathbf{y} < u_{n'\mathbf{k}'}|\mathbf{y}>< \mathbf{y}|\nabla_{\mathbf{k}''}u_{n''\mathbf{k}''} >)^* \\
&= -\delta(\mathbf{k}''-\mathbf{k}')\mathbf{d}_{n''n'}(\mathbf{k}') \qquad \text{(A4)}
\end{aligned}
$$

$$
\begin{aligned}
0 &= i\nabla_{\mathbf{k}} < u_{n\mathbf{k}}|u_{n'\mathbf{k}} > \\
&= i < \nabla_{\mathbf{k}}u_{n\mathbf{k}}|u_{n'\mathbf{k}} > +i < u_{n\mathbf{k}}|\nabla_{\mathbf{k}}u_{n'\mathbf{k}} > \\
&= -\mathbf{d}^*_{n'n}(\mathbf{k}) + \mathbf{d}_{nn'}(\mathbf{k}). \qquad \text{(A5)}
\end{aligned}
$$

$$
\begin{aligned}
&< nk|[\hat{x},\hat{\rho}]|n'k' > \\
&=\sum_{n''}\int dk''(< nk|\hat{x}|n''k'' >< n''k''|\hat{\rho}|n'k' > - < nk|\hat{\rho}|n''k'' >< n''k''|\hat{x}|n'k' >) \\
&=\sum_{n''}\int dk''[\{-i\nabla_{\mathbf{k}''} < n\mathbf{k}|n''\mathbf{k}'' > +\delta(\mathbf{k}-\mathbf{k}'')\mathbf{d}_{nn''}(\mathbf{k})\} < n''k''|\hat{\rho}|n'k' > \\
&\quad - < nk|\hat{\rho}|n''k'' > \{i\nabla_{\mathbf{k}''} < n''\mathbf{k}''|n'\mathbf{k}' > +\delta(\mathbf{k}''-\mathbf{k}')\mathbf{d}_{n''n'}(\mathbf{k}')\}] \\
&=i(\nabla_{\mathbf{k}}+\nabla_{\mathbf{k}'}) < nk|\hat{\rho}|n'k' > + \sum_{n''}\{\mathbf{d}_{nn''}(\mathbf{k}) < n''k|\hat{\rho}|n'k' > - < nk|\hat{\rho}|n''k' > \mathbf{d}_{n''n'}(\mathbf{k}')\}
\end{aligned}
\tag{A6}
$$

$$
\begin{aligned}
&<nk|[\hat{x},[\hat{x},\hat{\rho}]]|n'k' > \\
&=\sum_{n'''}\int dk''' < nk|\hat{x}|n'''k''' >< n'''k'''|[\hat{x},\hat{\rho}]|n'k' > - < nk|[\hat{x},\hat{\rho}]|n'''k''' >< n'''k'''|\hat{x}|n'k' >) \\
&=\sum_{n'''}\int dk'''\{-i\nabla_{k'''} < nk|n'''k''' > +\delta(\mathbf{k}-\mathbf{k}''')\mathbf{d}_{nn'''}(k)\} \\
&\quad \times[i(\nabla_{k'''}+\nabla_{k'}) < n'''k'''|\hat{\rho}|n'k' > + \sum_{n''}\mathbf{d}_{n'''n''}(\mathbf{k}''') < n''k'''|\hat{\rho}|n'k' > - < n'''k'''|\hat{\rho}|n''k' > \mathbf{d}_{n''n'}(\mathbf{k}')\}] \\
&\quad -[i(\nabla_{k}+\nabla_{k'''}) < nk|\hat{\rho}|n'''k''' > + \sum_{n''}\mathbf{d}_{nn''}() < n''k|\hat{\rho}|n'''k''' > - < nk|\hat{\rho}|n''k''' > \mathbf{d}_{n''n'''}(k''')] \\
&\quad \times\{i\nabla_{k'''} < n'''k'''|n'k' > +\delta(k'''-k')\mathbf{d}_{n'''n'}(k')\} \\
&=-(\nabla_{k}+\nabla_{k'})^2 < nk|\hat{\rho}|n'k' > +2i(\nabla_{k}+\nabla_{k'})\{\sum_{n''}\mathbf{d}_{nn''}(k) < n''k|\hat{\rho}|n'k' > - < nk|\hat{\rho}|n''k' > \mathbf{d}_{n''n'}(k')\} \\
&\quad + \sum_{n''n'''}[d_{nn''}(k)d_{n''n'''}(k) < n'''k|\hat{\rho}|n'k' > -2d_{nn''}(k) < n''k|\hat{\rho}|n'''k' > d_{n'''n'}(k') + < nk|\hat{\rho}|n''k' > d_{n''n'''}(k')d_{n'''n'}(k')]
\end{aligned}
\tag{A7}
$$

$$
[\hat{\mathbf{x}},\hat{H}_0] = [\hat{\mathbf{x}},\frac{\hat{\mathbf{p}}}{2m}] = \frac{i\hbar}{m}\hat{\mathbf{p}} \tag{A8}
$$

$$
\begin{aligned}
< n\mathbf{k}|\hat{\mathbf{p}}|n'\mathbf{k}' > &= i\frac{m(\epsilon_{n\mathbf{k}}-\epsilon_{n'\mathbf{k}'})}{\hbar} < n\mathbf{k}|\hat{\mathbf{x}}|n'\mathbf{k}' > \\
&= \begin{cases} \frac{m}{\hbar}(\epsilon_{n\mathbf{k}}-\epsilon_{n'\mathbf{k}'})\{-\nabla_{\mathbf{k}} < n\mathbf{k}|n'\mathbf{k}' > +i\delta(\mathbf{k}-\mathbf{k}')\mathbf{d}_{nn'}(\mathbf{k})\} \\ \frac{m}{\hbar}(\epsilon_{n\mathbf{k}}-\epsilon_{n'\mathbf{k}'})\{\nabla_{\mathbf{k}'} < n\mathbf{k}|n'\mathbf{k}' > +i\delta(\mathbf{k}-\mathbf{k}')\mathbf{d}_{nn'}(\mathbf{k})\} \end{cases}
\end{aligned}
\tag{A9}
$$

$$
\begin{aligned}
< n\mathbf{k}|\hat{\mathbf{p}}|n\mathbf{k} > &= \frac{m}{i\hbar} < n\mathbf{k}|[\hat{\mathbf{x}}, \hat{H}_0]|n\mathbf{k} > \\
&= \frac{m}{i\hbar}\{< u_{n\mathbf{k}}|(i\nabla_{\mathbf{k}} e^{-i\mathbf{k}\cdot\hat{\mathbf{x}}})\hat{H}_0|n\mathbf{k} > - < n\mathbf{k}|\hat{H}_0(-i\nabla_{\mathbf{k}} e^{i\mathbf{k}\cdot\hat{\mathbf{x}}})|u_{n\mathbf{k}} >\} \\
&= \frac{m}{\hbar}\{\nabla_{\mathbf{k}} < n\mathbf{k}|\hat{H}_0|n\mathbf{k} > - < \nabla_{\mathbf{k}} u_{n\mathbf{k}}|e^{-i\mathbf{k}\cdot\hat{\mathbf{x}}}\hat{H}_0|n\mathbf{k} > - < n\mathbf{k}|\hat{H}_0 e^{i\mathbf{k}\cdot\hat{\mathbf{x}}}|\nabla_{\mathbf{k}} u_{n\mathbf{k}} >\} \\
&= \frac{m}{\hbar}\{\nabla_{\mathbf{k}}\epsilon_{n\mathbf{k}} - \epsilon_{n\mathbf{k}}\nabla_{\mathbf{k}} < u_{n\mathbf{k}}|u_{n\mathbf{k}} >\} \\
&= \frac{m}{\hbar}\nabla_{\mathbf{k}}\epsilon_{n\mathbf{k}}
\end{aligned} \tag{A10}
$$

Appendix B: Bloch-basis and length gauge

$$
\begin{aligned}
& i\hbar\frac{\partial}{\partial t} < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > \\
& \quad = \sum_{n''}\int_{BZ} dk''(< n\mathbf{k}|\hat{H}_0 - e\mathbf{E}\cdot\hat{\mathbf{x}}|n''\mathbf{k}'' >< n''\mathbf{k}''|\hat{\rho}|n'\mathbf{k}' > \\
& \qquad - < n\mathbf{k}|\hat{\rho}|n''\mathbf{k}'' >< n''\mathbf{k}''|\hat{H}_0 - e\mathbf{E}\cdot\hat{\mathbf{x}}|n'\mathbf{k}' >) \\
& \quad = (\epsilon_{n\mathbf{k}} - \epsilon_{n'\mathbf{k}'}) < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > \\
& \qquad - e\mathbf{E}\cdot\sum_{n''}\int_{BZ} dk''(< n\mathbf{k}|\hat{\mathbf{x}}|n''\mathbf{k}'' >< n''\mathbf{k}''|\hat{\rho}|n'\mathbf{k}' > - < n\mathbf{k}|\hat{\rho}|n''\mathbf{k}'' >< n''\mathbf{k}''|\hat{\mathbf{x}}|n'\mathbf{k}' >) \\
& \quad = (\epsilon_{n\mathbf{k}} - \epsilon_{n'\mathbf{k}'}) < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > \\
& \qquad - e\mathbf{E}\cdot[i(\nabla_{\mathbf{k}} + \nabla_{\mathbf{k}'}) < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > + \sum_{n''}\{\mathbf{d}_{nn''}(\mathbf{k}) < n''\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > - \mathbf{d}_{n''n'}(\mathbf{k}') < n\mathbf{k}|\hat{\rho}|n''\mathbf{k}' >\}]
\end{aligned} \tag{B1}
$$

$$
\begin{aligned}
& i\hbar\frac{\partial}{\partial t} < v\mathbf{k}|\hat{\rho}|v\mathbf{k} > \\
& \quad = -e\mathbf{E}\cdot[i\nabla_{\mathbf{k}} < v\mathbf{k}|\hat{\rho}|v\mathbf{k} > + \{\mathbf{d}_{vc}(\mathbf{k}) < c\mathbf{k}|\hat{\rho}|v\mathbf{k} > - \mathbf{d}_{cv}(\mathbf{k}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} >\}] \\
& \quad = -e\mathbf{E}\cdot[i\nabla_{\mathbf{k}} < v\mathbf{k}|\hat{\rho}|v\mathbf{k} > - 2i\mathbf{Im}\{\mathbf{d}^*_{vc}(\mathbf{k}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} >\}]
\end{aligned} \tag{B2}
$$

$$
\begin{aligned}
& i\hbar\frac{\partial}{\partial t} < c\mathbf{k}|\hat{\rho}|c\mathbf{k} > \\
& \quad = -e\mathbf{E}\cdot[i\nabla_{\mathbf{k}} < c\mathbf{k}|\hat{\rho}|c\mathbf{k} > + \{\mathbf{d}_{cv}(\mathbf{k}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} > - \mathbf{d}_{vc}(\mathbf{k}) < c\mathbf{k}|\hat{\rho}|v\mathbf{k} >\}] \\
& \quad = -e\mathbf{E}\cdot[i\nabla_{\mathbf{k}} < c\mathbf{k}|\hat{\rho}|c\mathbf{k} > + 2i\mathbf{Im}\{\mathbf{d}^*_{vc}(\mathbf{k}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} >\}]
\end{aligned} \tag{B3}
$$

$$
i\hbar\frac{\partial}{\partial t} < v\mathbf{k}|\hat{\rho}|c\mathbf{k} >
$$

$$
\begin{aligned}
&=(\epsilon_{v\mathbf{k}} - \epsilon_{c\mathbf{k}}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} > \\
&\quad -e\mathbf{E}\cdot[i\nabla_{\mathbf{k}} < v\mathbf{k}|\hat{\rho}|c\mathbf{k} > + \sum_{n''}\{\mathbf{d}_{vn''}(\mathbf{k}) < n''\mathbf{k}|\hat{\rho}|c\mathbf{k} > -\mathbf{d}_{n''c}(\mathbf{k}) < v\mathbf{k}|\hat{\rho}|n''\mathbf{k} >\}] \\
&=-(\epsilon_{c\mathbf{k}} - \epsilon_{v\mathbf{k}}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} > -ie\mathbf{E}\cdot\nabla_{\mathbf{k}} < v\mathbf{k}|\hat{\rho}|c\mathbf{k} > \\
&\quad -e\mathbf{E}\cdot[-\{\mathbf{d}_{cc}(\mathbf{k}) - \mathbf{d}_{vv}(\mathbf{k})\} < v\mathbf{k}|\hat{\rho}|c\mathbf{k} > +\mathbf{d}_{vc}(\mathbf{k})\{< c\mathbf{k}|\hat{\rho}|c\mathbf{k} > - < v\mathbf{k}|\hat{\rho}|v\mathbf{k} >\}]
\end{aligned} \tag{B4}
$$

$$
\begin{aligned}
H_L &= tr\{\hat{\rho}(\hat{H}_0 - e\mathbf{E}\cdot\hat{\mathbf{x}})\} \\
&= \sum_n \int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n\mathbf{k} > \epsilon_{n\mathbf{k}} - e\mathbf{E}\cdot\{\sum_{nn'}\int d\mathbf{k}\int d\mathbf{k}' < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' >< n'\mathbf{k}'|\hat{\mathbf{x}}|n\mathbf{k} >\} \\
&= \sum_n \int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n\mathbf{k} > \epsilon_{n\mathbf{k}} \\
&\quad -e\mathbf{E}\cdot[\sum_{nn'}\int d\mathbf{k}\int d\mathbf{k}' < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > \{\frac{1}{2i}(\nabla_{\mathbf{k}} - \nabla_{\mathbf{k}'}) < n'\mathbf{k}'|n\mathbf{k} > +\delta(\mathbf{k}' - \mathbf{k})\mathbf{d}_{n'n}(\mathbf{k})\}] \\
&= \sum_n \int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n\mathbf{k} > \epsilon_{n\mathbf{k}} \\
&\quad -e\mathbf{E}\cdot\{\sum_n \int d\mathbf{k}\,\mathbf{Im}(\nabla_{\mathbf{k}} < n\mathbf{k}|\hat{\rho}|n\mathbf{k} >) + \sum_{nn'}\int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n'\mathbf{k} > \mathbf{d}_{n'n}(\mathbf{k})\} \\
&= \sum_n \int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n\mathbf{k} > \epsilon_{n\mathbf{k}} - e\mathbf{E}\cdot\{\sum_{nn'}\int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n'\mathbf{k} > \mathbf{d}_{n'n}(\mathbf{k})\}
\end{aligned} \tag{B5}
$$

$$
\begin{aligned}
H_L^2 &= \sum_n \int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n\mathbf{k} > \epsilon_{n\mathbf{k}} - e\mathbf{E}\cdot\{\sum_{nn'}\int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n'\mathbf{k} > \mathbf{d}_{n'n}(\mathbf{k})\} \\
&= \int d\mathbf{k}(< v\mathbf{k}|\hat{\rho}|v\mathbf{k} > \epsilon_{v\mathbf{k}} + < c\mathbf{k}|\hat{\rho}|c\mathbf{k} > \epsilon_{c\mathbf{k}}) \\
&\quad -e\mathbf{E}\cdot\int d\mathbf{k}[< v\mathbf{k}|\hat{\rho}|v\mathbf{k} > \mathbf{d}_{vv}(\mathbf{k}) + < c\mathbf{k}|\hat{\rho}|c\mathbf{k} > \mathbf{d}_{cc}(\mathbf{k}) + 2\mathbf{Re}\{< v\mathbf{k}|\hat{\rho}|c\mathbf{k} > \mathbf{d}^*_{vc}(\mathbf{k})\}]
\end{aligned} \tag{B6}
$$

Appendix C: Bloch-basis and velocity gauge

$$
\begin{aligned}
&i\hbar\frac{\partial}{\partial t} < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > \\
&=\sum_{n''}\int d\mathbf{k}''(< n\mathbf{k}|\hat{H}_0 - \frac{e\mathbf{A}\cdot\hat{\mathbf{p}}}{m}|n''\mathbf{k}'' >< n''\mathbf{k}''|\hat{\rho}|n'\mathbf{k}' > \\
&\quad - < n\mathbf{k}|\hat{\rho}|n''\mathbf{k}'' >< n''\mathbf{k}''|\hat{H}_0 - \frac{e\mathbf{A}\cdot\hat{\mathbf{p}}}{m}|n'\mathbf{k}' >)
\end{aligned}
$$

$$
\begin{aligned}
=&(\epsilon_{n\mathbf{k}} - \epsilon_{n'\mathbf{k}'}) < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > \\
&-\frac{e\mathbf{A}}{m} \cdot \sum_{n''} \int d\mathbf{k}'' \{< n\mathbf{k}|\hat{\mathbf{p}}|n''\mathbf{k}'' >< n''\mathbf{k}''|\hat{\rho}|n'\mathbf{k}' > - < n\mathbf{k}|\hat{\rho}|n''\mathbf{k}'' >< n''\mathbf{k}''|\hat{\mathbf{p}}|n'\mathbf{k}' >\} \\
=&(\epsilon_{n\mathbf{k}} - \epsilon_{n'\mathbf{k}'}) < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > \\
&-\frac{e\mathbf{A}}{\hbar} \cdot \sum_{n''} \int d\mathbf{k}'' [(\epsilon_{n\mathbf{k}} - \epsilon_{n''\mathbf{k}''})\{\nabla_{\mathbf{k}''} < n\mathbf{k}|n''\mathbf{k}'' > + i\delta(\mathbf{k} - \mathbf{k}'')\mathbf{d}_{nn''}(\mathbf{k})\} < n''\mathbf{k}''|\hat{\rho}|n'\mathbf{k}' > \\
&- < n\mathbf{k}|\hat{\rho}|n''\mathbf{k}'' > (\epsilon_{n''\mathbf{k}''} - \epsilon_{n'\mathbf{k}'})\{-\nabla_{\mathbf{k}''} < n''\mathbf{k}''|n'\mathbf{k}' > + i\delta(\mathbf{k}'' - \mathbf{k}')\mathbf{d}_{n''n'}(\mathbf{k}')\}] \\
=&(\epsilon_{n\mathbf{k}} - \epsilon_{n'\mathbf{k}'}) < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > \\
&-\frac{e\mathbf{A}}{\hbar} \cdot (\nabla_{\mathbf{k}}\epsilon_{n\mathbf{k}} - \nabla_{\mathbf{k}'}\epsilon_{n'\mathbf{k}'}) < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > \\
&-\frac{ie\mathbf{A}}{\hbar} \cdot \sum_{n''} [(\epsilon_{n\mathbf{k}} - \epsilon_{n''\mathbf{k}})\mathbf{d}_{nn''}(\mathbf{k}) < n''\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > - (\epsilon_{n''\mathbf{k}'} - \epsilon_{n'\mathbf{k}'}) < n\mathbf{k}|\hat{\rho}|n''\mathbf{k}' > \mathbf{d}_{n''n'}(\mathbf{k}')]
\end{aligned}
\tag{C1}
$$

$$
\begin{aligned}
i\hbar\frac{\partial}{\partial t}&< v\mathbf{k}|\hat{\rho}|v\mathbf{k} > \\
&= -\frac{ie\mathbf{A}}{\hbar} \cdot \{(\epsilon_{v\mathbf{k}} - \epsilon_{c\mathbf{k}})\mathbf{d}_{vc}(\mathbf{k}) < c\mathbf{k}|\hat{\rho}|v\mathbf{k} > - (\epsilon_{c\mathbf{k}} - \epsilon_{v\mathbf{k}}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} > \mathbf{d}_{cv}(\mathbf{k})\} \\
&= +\frac{ie\mathbf{A}}{\hbar} \cdot [2(\epsilon_{c\mathbf{k}} - \epsilon_{v\mathbf{k}})\mathbf{Re}\{\mathbf{d}^*_{vc}(\mathbf{k}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} >\}]
\end{aligned}
\tag{C2}
$$

$$
\begin{aligned}
i\hbar\frac{\partial}{\partial t}&< c\mathbf{k}|\hat{\rho}|c\mathbf{k} > \\
&= -\frac{ie\mathbf{A}}{\hbar} \cdot \{(\epsilon_{c\mathbf{k}} - \epsilon_{v\mathbf{k}})\mathbf{d}_{cv}(\mathbf{k}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} > - (\epsilon_{v\mathbf{k}} - \epsilon_{c\mathbf{k}}) < c\mathbf{k}|\hat{\rho}|v\mathbf{k} > \mathbf{d}_{vc}(\mathbf{k})\} \\
&= -\frac{ie\mathbf{A}}{\hbar} \cdot [2(\epsilon_{c\mathbf{k}} - \epsilon_{v\mathbf{k}})\mathbf{Re}\{\mathbf{d}^*_{vc}(\mathbf{k}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} >\}]
\end{aligned}
\tag{C3}
$$

$$
\begin{aligned}
i\hbar\frac{\partial}{\partial t}&< v\mathbf{k}|\hat{\rho}|c\mathbf{k} > \\
&= (\epsilon_{v\mathbf{k}} - \epsilon_{c\mathbf{k}}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} > - \frac{e\mathbf{A}}{\hbar} \cdot \{(\nabla_{\mathbf{k}}\epsilon_{v\mathbf{k}} - \nabla_{\mathbf{k}}\epsilon_{c\mathbf{k}}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} >\} \\
&\quad - \frac{ie\mathbf{A}}{\hbar} \cdot \{(\epsilon_{v\mathbf{k}} - \epsilon_{c\mathbf{k}})\mathbf{d}_{vc}(\mathbf{k}) < c\mathbf{k}|\hat{\rho}|c\mathbf{k} > - (\epsilon_{v\mathbf{k}} - \epsilon_{c\mathbf{k}}) < v\mathbf{k}|\hat{\rho}|v\mathbf{k} > \mathbf{d}_{vc}(\mathbf{k})\} \\
&= -(\epsilon_{c\mathbf{k}} - \epsilon_{v\mathbf{k}}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} > + \frac{e\mathbf{A}}{\hbar} \cdot \{(\nabla_{\mathbf{k}}\epsilon_{c\mathbf{k}} - \nabla_{\mathbf{k}}\epsilon_{v\mathbf{k}}) < v\mathbf{k}|\hat{\rho}|c\mathbf{k} >\} \\
&\quad + \frac{ie\mathbf{A}}{\hbar} \cdot \{(\epsilon_{c\mathbf{k}} - \epsilon_{v\mathbf{k}})\mathbf{d}_{vc}(\mathbf{k})(< c\mathbf{k}|\hat{\rho}|c\mathbf{k} > - < v\mathbf{k}|\hat{\rho}|v\mathbf{k} >)\}
\end{aligned}
\tag{C4}
$$

$$
\begin{aligned}
H_V &= tr\{\hat{\rho}(\hat{H}_0 - \frac{e\mathbf{A}\cdot\hat{\mathbf{p}}}{m})\} \\
&= \sum_n \int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n\mathbf{k} > \epsilon_{n\mathbf{k}} - \frac{e\mathbf{A}}{m} \cdot \{\sum_{nn'} \int d\mathbf{k} \int d\mathbf{k}' < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' >< n'\mathbf{k}'|\hat{\mathbf{p}}|n\mathbf{k} >\} \\
&= \sum_n \int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n\mathbf{k} > \epsilon_{n\mathbf{k}}
\end{aligned}
$$

$$
\begin{aligned}
&-\frac{e\mathbf{A}}{\hbar}\cdot[\sum_{nn'}\int d\mathbf{k}\int d\mathbf{k}'(\epsilon_{n'\mathbf{k}'}-\epsilon_{n\mathbf{k}}) < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > \{\nabla_{\mathbf{k}} < n'\mathbf{k}'|n\mathbf{k} > +i\delta(\mathbf{k}'-\mathbf{k})\mathbf{d}_{n'n}(\mathbf{k})\}] \\
=&\sum_{n}\int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n\mathbf{k} > \epsilon_{n\mathbf{k}} \\
&-\frac{e\mathbf{A}}{\hbar}\cdot[\sum_{n}\int d\mathbf{k}\nabla_{\mathbf{k}}\epsilon_{n\mathbf{k}} < n\mathbf{k}|\hat{\rho}|n\mathbf{k} > +i\sum_{n\neq n'}\int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n'\mathbf{k} > (\epsilon_{n'\mathbf{k}'}-\epsilon_{n\mathbf{k}})\mathbf{d}_{n'n}(\mathbf{k})]
\end{aligned}
\tag{C5}
$$

$$
\begin{aligned}
H_V^2 =& \int d\mathbf{k}(< v\mathbf{k}|\hat{\rho}|v\mathbf{k} > \epsilon_{v\mathbf{k}} + < c\mathbf{k}|\hat{\rho}|c\mathbf{k} > \epsilon_{c\mathbf{k}}) \\
&-\frac{e\mathbf{A}}{\hbar}\cdot\int d\mathbf{k}(< v\mathbf{k}|\hat{\rho}|v\mathbf{k} > \nabla_{\mathbf{k}}\epsilon_{v\mathbf{k}} + < c\mathbf{k}|\hat{\rho}|c\mathbf{k} > \nabla_{\mathbf{k}}\epsilon_{c\mathbf{k}}) \\
&+\frac{2e\mathbf{A}}{\hbar}\cdot\int d\mathbf{k}(\epsilon_{c\mathbf{k}}-\epsilon_{v\mathbf{k}})\mathbf{Im}\{< v\mathbf{k}|\hat{\rho}|c\mathbf{k} > \mathbf{d}^*_{vc}(\mathbf{k})\}
\end{aligned}
\tag{C6}
$$

Appendix D: velocity/current and position

$$
\begin{aligned}
J =& -e\frac{\partial}{\partial t}tr(\hat{\rho}\hat{\mathbf{x}}) \\
=& -e\,tr(\frac{\partial\hat{\rho}}{\partial t}\hat{\mathbf{x}}) \\
=& -\frac{e}{i\hbar}tr([\hat{H},\hat{\rho}]\hat{\mathbf{x}}) \\
=& -\frac{e}{i\hbar}tr\{\hat{\rho}(\hat{\mathbf{x}}\hat{H}-\hat{H}\hat{\mathbf{x}})\} \\
=& -\frac{e}{i\hbar}tr\{\hat{\rho}[\hat{\mathbf{x}},\hat{H}]\} \\
=& -\frac{e}{m}tr\{\hat{\rho}(\hat{\mathbf{p}}-e\mathbf{A})\} \\
=& -\frac{e}{m}\sum_{nn'}\int d\mathbf{k}\int d\mathbf{k}' < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' >< n'\mathbf{k}'|\hat{\mathbf{p}}|n\mathbf{k} > +\frac{e^2\mathbf{A}}{m}tr(\hat{\rho}) \\
=& -\frac{e}{\hbar}\sum_{nn'}\int d\mathbf{k}\int d\mathbf{k}' < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > (\epsilon_{n'\mathbf{k}'}-\epsilon_{n\mathbf{k}})\{\nabla_{\mathbf{k}} < n'\mathbf{k}'|n\mathbf{k} > +i\delta(\mathbf{k}'-\mathbf{k})\mathbf{d}_{n'n}(\mathbf{k})\} \\
&+\frac{e^2\mathbf{A}}{m}tr(\hat{\rho}) \\
=& +\frac{e}{\hbar}\sum_{nn'}\int d\mathbf{k}\int d\mathbf{k}'\nabla_{\mathbf{k}}\{< n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > (\epsilon_{n'\mathbf{k}'}-\epsilon_{n\mathbf{k}})\} < n'\mathbf{k}'|n\mathbf{k} > \\
&-\frac{e}{\hbar}\sum_{nn'}\int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n'\mathbf{k} > (\epsilon_{n'\mathbf{k}}-\epsilon_{n\mathbf{k}})\{i\mathbf{d}_{n'n}(\mathbf{k})\} + \frac{e^2\mathbf{A}}{m}tr(\hat{\rho}) \\
=& -\frac{e}{\hbar}\sum_{n}\int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n\mathbf{k} > \nabla_{\mathbf{k}}\epsilon_{n\mathbf{k}} - \frac{e}{\hbar}\sum_{nn'}\int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n'\mathbf{k} > (\epsilon_{n'\mathbf{k}}-\epsilon_{n\mathbf{k}})\{i\mathbf{d}_{n'n}(\mathbf{k})\} + \frac{e^2\mathbf{A}}{m}tr(\hat{\rho})
\end{aligned}
\tag{D1}
$$

$$
\begin{aligned}
J^2 = &-\frac{e}{\hbar}\int d\mathbf{k}(< v\mathbf{k}|\hat{\rho}|v\mathbf{k} > \nabla_{\mathbf{k}}\epsilon_{v\mathbf{k}} + < c\mathbf{k}|\hat{\rho}|c\mathbf{k} > \nabla_{\mathbf{k}}\epsilon_{c\mathbf{k}}) + \frac{e^2\mathbf{A}}{m}tr(\hat{\rho}) \\
&-\frac{e}{\hbar}\int d\mathbf{k}[< v\mathbf{k}|\hat{\rho}|c\mathbf{k} > (\epsilon_{c\mathbf{k}} - \epsilon_{v\mathbf{k}})\{i\mathbf{d}_{cv}(\mathbf{k})\} + < c\mathbf{k}|\hat{\rho}|v\mathbf{k} > (\epsilon_{v\mathbf{k}} - \epsilon_{c\mathbf{k}})\{i\mathbf{d}_{vc}(\mathbf{k})\}] \\
= &-\frac{e}{\hbar}\int d\mathbf{k}(< v\mathbf{k}|\hat{\rho}|v\mathbf{k} > \nabla_{\mathbf{k}}\epsilon_{v\mathbf{k}} + < c\mathbf{k}|\hat{\rho}|c\mathbf{k} > \nabla_{\mathbf{k}}\epsilon_{c\mathbf{k}}) + \frac{e^2\mathbf{A}}{m}tr(\hat{\rho}) \\
&+\frac{e}{\hbar}\int d\mathbf{k}(\epsilon_{c\mathbf{k}} - \epsilon_{v\mathbf{k}})\mathbf{Im}[< v\mathbf{k}|\hat{\rho}|c\mathbf{k} > \mathbf{d}_{vc}]
\end{aligned} \tag{D2}
$$

$$
\begin{aligned}
\mathbf{x} &= tr(\hat{\rho}\hat{\mathbf{x}}) \\
&= \sum_{nn'}\int d\mathbf{k}\int d\mathbf{k}' < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' >< n'\mathbf{k}'|\hat{\mathbf{x}}|n\mathbf{k} > \\
&= \sum_{nn'}\int d\mathbf{k}\int d\mathbf{k}' < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > \{\frac{1}{2i}(\nabla_{\mathbf{k}} - \nabla_{\mathbf{k}'}) < n'\mathbf{k}'|n\mathbf{k} > + \delta(\mathbf{k}' - \mathbf{k})\mathbf{d}_{n'n}(\mathbf{k})\} \\
&= \sum_{nn'}\int d\mathbf{k} < n\mathbf{k}|\hat{\rho}|n'\mathbf{k} > \mathbf{d}_{n'n}(\mathbf{k})
\end{aligned} \tag{D3}
$$

$$
\mathbf{x}^2 = \int d\mathbf{k}\{< v\mathbf{k}|\hat{\rho}|v\mathbf{k} > \mathbf{d}_{vv}(\mathbf{k}) + < c\mathbf{k}|\hat{\rho}|c\mathbf{k} > \mathbf{d}_{cc}(\mathbf{k}) + 2\mathbf{Re}\{< v\mathbf{k}|\hat{\rho}|c\mathbf{k} > \mathbf{d}^*_{vc}(\mathbf{k})\}] \tag{D4}
$$

Appendix E: The gauge transformations

$$|x> \to |x'>= U(x)|x> \tag{E1}$$

$$|k> \to |k'>= U(k)|k> \tag{E2}$$

where $U(x)$ and $U(k)$ are unitary. note: I have no idea if U(x) and U(k) are independent.

$$\phi \to \phi' = \phi + \frac{\hbar}{e}\frac{\partial \chi}{\partial t} \tag{E3}$$

$$\mathbf{A} \to \mathbf{A}' = +\frac{\hbar}{e}\nabla\chi \tag{E4}$$

note: H^V is not the hamilton of the whole system; what is does A mean?

$$\hat{O} \to U^{\dagger}\hat{O}U$$

$$e^A B e^{-A} = B + [A, B] + \frac{1}{2}[A, [A, B]] + ...$$

By Campbell-Baker-Hausdorff formula, the hamiltonian transforms as

$$\begin{aligned}
\hat{H}^V &= \frac{(\hat{\mathbf{p}} - e\mathbf{A})^2}{2m} + V(\hat{x}) \\
\to \hat{H}^L &= exp(-\frac{ie\widetilde{\mathbf{A}}\cdot\hat{\mathbf{x}}}{\hbar})\hat{H}^V exp(\frac{ie\widetilde{\mathbf{A}}\cdot\hat{\mathbf{x}}}{\hbar}) \\
&= \hat{H}^V + \frac{ie}{\hbar}[\widetilde{\mathbf{A}}\cdot\hat{\mathbf{x}}, \hat{H}^V] + o^2 \\
&= \hat{H}^V + \frac{ie}{\hbar}\{\widetilde{\mathbf{A}}\cdot[\hat{\mathbf{x}}, \hat{H}^V] + [\widetilde{\mathbf{A}}, \hat{H}^V]\cdot\hat{\mathbf{x}}\} + o^2 \\
&= \hat{H}^V + \frac{ie}{\hbar}\{\widetilde{\mathbf{A}}\cdot(i\hbar\frac{\hat{\mathbf{p}} - e\mathbf{A}}{m}) + (-i\hbar\frac{\partial\widetilde{\mathbf{A}}}{\partial t})\cdot\hat{\mathbf{x}}\} + o^2 \\
&= \frac{\hat{\mathbf{p}}^2}{2m} - e\mathbf{E}\cdot\hat{\mathbf{x}} + V(\hat{\mathbf{x}}) + (constant).
\end{aligned} \tag{E5}$$

where the vector potential $\widetilde{\mathbf{A}}$ is formally an operator. note: H^V is not the hamilton of the whole system; what does A mean?

Hereafter we consider 1-dimensional case. According to the Campbell-Baker-Hausdorff formula, the density operator transforms as

$$\begin{aligned}\hat{\rho}^V \to \hat{\rho}^L &= exp(-\frac{ie\widetilde{A}\hat{x}}{\hbar})\hat{\rho}^V exp(\frac{ie\widetilde{A}\hat{x}}{\hbar}) \\ &= \hat{\rho}^V - \frac{ie\widetilde{A}}{\hbar}[\hat{x},\hat{\rho}^V] - \frac{e^2\widetilde{A}^2}{\hbar^2}[\hat{x},[\hat{x},\hat{\rho}^V]] + O(\widetilde{A}^3)\end{aligned} \tag{E6}$$

Appendix F: variation principle and charge; the Neother's theorem

Appendix G: Wannier basis

n is the band index, $\mathbf{k}$ is the crystal momentum, $\mathbf{R}$ is the Bravais Lattice The Bloch states $\{|n\mathbf{k}>\}$ and Wannier states $\{|n\mathbf{R}>\}$ are related by the Fourier expansion.

$$|n\mathbf{R}> = \frac{1}{\sqrt{\Omega}}\int_{BZ} d\mathbf{k}\, exp(-i\mathbf{k}\cdot\mathbf{R})|n\mathbf{k}> \tag{G1}$$

$$= \frac{1}{\sqrt{\Omega}}\int_{BZ} d\mathbf{k}\, exp\{-i\mathbf{k}\cdot(\mathbf{R}-\hat{\mathbf{x}})\}|u_{n\mathbf{k}}> \tag{G2}$$

$$<\mathbf{x}|n\mathbf{R}> = \frac{1}{\sqrt{\Omega}}\int_{BZ} d\mathbf{k}\, exp\{-i\mathbf{k}\cdot(\mathbf{R}-\mathbf{x})\} <\mathbf{x}|u_{n\mathbf{k}}> \tag{G3}$$

$$|n\mathbf{k}> = \frac{1}{\sqrt{N}}\sum_{\mathbf{R}} exp(i\mathbf{k}\cdot\mathbf{R})|n\mathbf{R}> \tag{G4}$$

$$\begin{aligned}<n\mathbf{R}|n\mathbf{R}> &= \frac{1}{\Omega}\int\int_{BZ} d\mathbf{k}d\mathbf{k}' exp\{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}\} <n\mathbf{k}|n\mathbf{k}'> \\ &= \frac{1}{\Omega}\int_{BZ} dk \\ &= 1\end{aligned} \tag{G5}$$

$$\begin{aligned}<n\mathbf{k}|n\mathbf{k}> &= \frac{1}{N}\sum_{\mathbf{R}\mathbf{R}'} exp\{-i\mathbf{k}\cdot(\mathbf{R}-\mathbf{R}')\} <n\mathbf{R}|n\mathbf{R}'> \\ &= \frac{1}{\Omega}\int_{BZ} dk \\ &= 1\end{aligned} \tag{G6}$$

$$\begin{aligned}|n\mathbf{k}> &= \frac{1}{\sqrt{N}}\sum_{\mathbf{R}} exp(i\mathbf{k}\cdot\mathbf{R})|n\mathbf{R}> \\ &= \frac{1}{\sqrt{N\Omega}}\sum_{\mathbf{R}}\int_{BZ} d\mathbf{k}'\, exp\{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}\}|n\mathbf{k}'> \\ |n\mathbf{R}> &= \frac{1}{\sqrt{\Omega}}\int_{BZ} d\mathbf{k}\, exp(-i\mathbf{k}\cdot\mathbf{R})|n\mathbf{k}> \\ &= \frac{1}{\sqrt{N\Omega}}\sum_{\mathbf{R}'}\int_{BZ} d\mathbf{k}\, exp\{-i\mathbf{k}\cdot(\mathbf{R}-\mathbf{R}')\}|n\mathbf{R}'>\end{aligned} \tag{G7}$$

where N is number of Bravais lattice points, and Ω is the volume of a Brillouin Zone. The matrix element of an operator $\hat{O}$ is transferred to

$$< n\mathbf{R}|\hat{O}|n'\mathbf{R}' >\propto \int\int_{BZ} dkdk' exp[i(\mathbf{k}\cdot\mathbf{R}-\mathbf{k}'\cdot\mathbf{R}')] < n\mathbf{k}|\hat{O}|n'\mathbf{k}' > \tag{G8}$$

$$\begin{aligned} < n\mathbf{R}|\hat{H}_0|n'\mathbf{R}' >\propto & \int\int_{BZ} d\mathbf{k}\, d\mathbf{k}' exp\{i(\mathbf{k}\cdot\mathbf{R}-\mathbf{k}'\cdot\mathbf{R}')\} < n\mathbf{k}|\hat{H}_0|n'\mathbf{k}' > \\ = & \int d\mathbf{k}\, exp\{i\mathbf{k}\cdot(\mathbf{R}-\mathbf{R}')\}\delta_{nn'}\epsilon_{n\mathbf{k}} && \text{(G9)} \\ \equiv & \delta_{nn'}\widetilde{\epsilon}_n(\mathbf{R}-\mathbf{R}') && \text{(G10)} \\ & && \text{(G11)} \end{aligned}$$

$$\begin{aligned} < n\mathbf{R}|\hat{\mathbf{x}}|n'\mathbf{R}' >= & \frac{1}{\Omega}\int\int_{BZ} d\mathbf{k}\, d\mathbf{k}' exp\{i(\mathbf{k}\cdot\mathbf{R}-\mathbf{k}'\cdot\mathbf{R}')\} < n\mathbf{k}|\hat{\mathbf{x}}|n'\mathbf{k}' > \\ = & \frac{1}{\Omega}\int\int_{BZ} d\mathbf{k}\, d\mathbf{k}' exp\{i(\mathbf{k}\cdot\mathbf{R}-\mathbf{k}'\cdot\mathbf{R}')\}\{i\nabla_{\mathbf{k}} < n\mathbf{k}|n'\mathbf{k}' > +\delta(\mathbf{k}-\mathbf{k}')\mathbf{d}_{nn'}(\mathbf{k})\} \\ = & \frac{1}{\Omega}\int d\mathbf{k}\, exp\{i\mathbf{k}\cdot(\mathbf{R}-\mathbf{R}')\}\{\delta_{nn'}\mathbf{R}+\mathbf{d}_{nn'}(\mathbf{k})\} && \text{(G12)} \\ = & \delta_{nn'}\delta_{\mathbf{R}\mathbf{R}'}\mathbf{R}+\widetilde{\mathbf{d}}_{nn'}(\mathbf{R}-\mathbf{R}') && \text{(G13)} \\ & && \text{(G14)} \end{aligned}$$

here we define $\widetilde{\mathbf{d}}_{nn'}(\mathbf{R}-\mathbf{R}') \equiv \frac{1}{\Omega}\int d\mathbf{k}\, exp\{i\mathbf{k}\cdot(\mathbf{R}-\mathbf{R}')\}\mathbf{d}_{nn'}(\mathbf{k})$.

$$\begin{aligned} < n\mathbf{R}|[\hat{\mathbf{x}},\hat{\rho}]|n'\mathbf{R}' >= & \sum_{n''\mathbf{R}''}(< n\mathbf{R}|\hat{\mathbf{x}}|n''\mathbf{R}'' >< n''\mathbf{R}''|\hat{\rho}|n'\mathbf{R}' > - < n\mathbf{R}|\hat{\rho}|n''\mathbf{R}'' >< n''\mathbf{R}''|\hat{\mathbf{x}}|n'\mathbf{R}' >) \\ = & \sum_{n''\mathbf{R}''}[\{\delta_{nn''}\delta_{\mathbf{R}\mathbf{R}''}\mathbf{R}+\widetilde{\mathbf{d}}_{nn''}(\mathbf{R}-\mathbf{R}'')\} < n''\mathbf{R}''|\hat{\rho}|n'\mathbf{R}' > \\ & - < n\mathbf{R}|\hat{\rho}|n''\mathbf{R}'' > \{\delta_{n''n'}\delta_{\mathbf{R}''\mathbf{R}'}\mathbf{R}'+\widetilde{\mathbf{d}}_{n''n'}(\mathbf{R}''-\mathbf{R}')\}] \\ = & (\mathbf{R}-\mathbf{R}') < n\mathbf{R}|\hat{\rho}|n'\mathbf{R}' > \\ & + \sum_{n''\mathbf{R}''}\{\widetilde{\mathbf{d}}_{nn''}(\mathbf{R}-\mathbf{R}'')\} < n''\mathbf{R}''|\hat{\rho}|n'\mathbf{R}' > - < n\mathbf{R}|\hat{\rho}|n''\mathbf{R}'' > \widetilde{\mathbf{d}}_{n''n'}(\mathbf{R}''-\mathbf{R}')\}] \\ & && \text{(G15)} \end{aligned}$$

$$< n\mathbf{R}|[\hat{H}_0,\hat{\rho}]|n'\mathbf{R}' >= \sum_{n''\mathbf{R}''}(< n\mathbf{R}|\hat{H}_0|n''\mathbf{R}'' >< n''\mathbf{R}''|\hat{\rho}|n'\mathbf{R}' > - < n\mathbf{R}|\hat{\rho}|n''\mathbf{R}'' >< n''\mathbf{R}''|\hat{H}_0|n'\mathbf{R}' >)$$

$$
\begin{aligned}
&= \sum_{n''\mathbf{R}''} \{\delta_{nn''}\widetilde{\epsilon}_n(\mathbf{R}-\mathbf{R}'') < n''\mathbf{R}''|\hat{\rho}|n'\mathbf{R}' > - < n\mathbf{R}|\hat{\rho}|n''\mathbf{R}'' > \delta_{n''n'}\widetilde{\epsilon}_{n'}(\mathbf{R}''-\mathbf{R}')\} \\
&= \sum_{\mathbf{R}''}\{\widetilde{\epsilon}_n(\mathbf{R}-\mathbf{R}'') < n\mathbf{R}''|\hat{\rho}|n'\mathbf{R}' > - < n\mathbf{R}|\hat{\rho}|n'\mathbf{R}'' > \widetilde{\epsilon}_{n'}(\mathbf{R}''-\mathbf{R}')\}
\end{aligned}
\tag{G16}
$$

$$
\begin{aligned}
i\hbar < n\mathbf{R}|\hat{\rho}|n'\mathbf{R}' >&= \sum_{n''\mathbf{R}''} (< n\mathbf{R}||n''\mathbf{R}'' >< n''\mathbf{R}''|\hat{\rho}|n'\mathbf{R}' > - < n\mathbf{R}|\hat{\rho}|n''\mathbf{R}'' >< n''\mathbf{R}''|\hat{\mathbf{x}}|n'\mathbf{R}' >) \\
&= \sum_{n''\mathbf{R}''} (< n\mathbf{R}|\hat{\mathbf{x}}|n''\mathbf{R}'' >< n''\mathbf{R}''|\hat{\rho}|n'\mathbf{R}' > - < n\mathbf{R}|\hat{\rho}|n''\mathbf{R}'' >< n''\mathbf{R}''|\hat{\mathbf{x}}|n'\mathbf{R}' >)
\end{aligned}
\tag{G17}
$$

Appendix H: the Wannier representation -Length gauge

$$
\begin{aligned}
&\frac{\partial}{\partial t}< n\mathbf{R}|\hat{\rho}|n'\mathbf{R}' > \\
&=\frac{1}{i\hbar}\sum_{n''\mathbf{R}''}(< n\mathbf{R}|\hat{H}_0 - e\mathbf{E}\cdot\hat{\mathbf{x}}|n''\mathbf{R}'' >< n''\mathbf{R}''|\hat{\rho}|n'\mathbf{R}' > \\
&\quad - < n\mathbf{R}|\hat{\rho}|n''\mathbf{R}'' >< n''\mathbf{R}''|\hat{H}_0 - e\mathbf{E}\cdot\hat{\mathbf{x}}|n'\mathbf{R}' >) \\
&=\frac{1}{i\hbar}[(\epsilon_{n\mathbf{k}} - \epsilon_{n'\mathbf{k}'}) < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > \\
&\quad -e\mathbf{E}\cdot(< n\mathbf{k}|\hat{\mathbf{x}}|n\mathbf{k} > - < n'\mathbf{k}'|\hat{\mathbf{x}}|n'\mathbf{k}' >) < n\mathbf{k}|\hat{\rho}|n'\mathbf{k}' > \\
&\quad -e\mathbf{E}\cdot\{\sum_{n''\neq n;\mathbf{k}''} < n\mathbf{k}|\hat{\mathbf{x}}|n''\mathbf{k}'' >< n''\mathbf{k}''|\hat{\rho}|n'\mathbf{k}' > \\
&\quad - \sum_{n''\neq n';\mathbf{k}''} < n\mathbf{k}|\hat{\rho}|n''\mathbf{k}'' >< n''\mathbf{k}''|\hat{\mathbf{x}}|n'\mathbf{k}' >\}]
\end{aligned}
$$

$$
\begin{aligned}
\frac{\partial\rho_{n',n}(l',l;t)}{\partial t} &= \frac{1}{i\hbar}\sum_{n'',l''}\{[\epsilon_{n'}(l'-l'')\delta_{n'n''}] \\
-eE_0\cdot\Delta_{n'n''}(l'-l'') &+ eV_{n'n''}(l',l'')]\rho_{n'',n} \\
-[\epsilon_n(l''-l)\delta_{n'n''}] &- eE_0\cdot\Delta_{n'n''}(l'-l'') \\
&+eV_{n'n''}(l',l'')]\rho_{n'',n}\}
\end{aligned}
$$

ここで $\mathbf{m}(x;t) = \frac{1}{\sqrt{(N)}}\sum_l exp(iK\dot{l})A_n(x-l;t)$

$\epsilon_n(l'-l'')$ と $\Delta_{nn'}(l-l')$ は双極子遷移のフーリエ展開に対応

$\epsilon_n(l'-l'') = \frac{1}{N}\sum_K exp(-ik\cdot(l-l'))\epsilon_n(k-\frac{e}{\hbar c}A_0)$

$\Delta_{nn'}(l-l') = \frac{1}{N}\sum_K exp(-ik\cdot(l-l'))R_{nn'})(k)$

$V_{n'n}(l',l) = \int dx W^*_{n'}(x,l')V(x,t)W_n(x,l)$

$R_{nn'}(k) = \frac{i}{\Omega}\int dx U^*_{n'k}\nabla_k U_{nk}$

Appendix I: the Wannier representation -Velocity gauge

[3]

Appendix J: The Maximally localized Wannier function

[1] Péter Földi. Gauge invariance and interpretation of interband and intraband processes in high-order harmonic generation from bulk solids. *Phys. Rev. B*, 96:035112, Jul 2017.

[2] E.I. Blount. Formalisms of band theory. volume 13 of *Solid State Physics*, pages 305 – 373. Academic Press, 1962.

[3] Gerald J. Iafrate and Joseph B. Krieger. Quantum transport for bloch electrons in inhomogeneous electric fields. *Phys. Rev. B*, 40:6144–6148, Sep 1989.