Supporting information

Carrier-Envelope-Phase Modulated Currents in Scanning Tunneling Microscopy

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First Order Integrated Current

For simplicity, we use the following abbreviated integration notation,

$$\int \mathrm{d}t_1 \mathrm{d}t_2 \cdots \mathrm{d}t_n \, \mathbf{A}_0\left(t, t_1\right) \mathbf{A}_1\left(t_1, t_2\right) \cdots \mathbf{A}_n\left(t_n, t\right) = \int \mathrm{d}\tau \, \mathbf{A}_0 \mathbf{A}_1 \cdots \mathbf{A}_n. \tag{S1}$$

$\mathbf{A}_i$ are general matrices, and they can also be single-variable, $\mathbf{A}_i\left(t_i\right)$. The fold of integration can be deduced from the number of $\mathbf{A}_i$.

We split the response to electric field into two parts, *i.e.*, (1) the sole response to induced bias; (2) the remaining part. The first part is

$$I_\alpha^{(1,1)}(t) = e \int \mathrm{d}\tau \, \mathrm{tr}\left[\mathbf{G}^r \mathbf{\Sigma}_1^< \mathbf{G}^a \mathbf{\Sigma}_\alpha^a + \mathbf{G}^r \mathbf{\Sigma}_{\alpha 1}^< + \mathbf{h.c.}\right]. \tag{S2}$$

Perform Fourier transform from t to ω,

$$\begin{aligned} \mathcal{F}_t\left[\int \mathrm{d}\tau \, \mathbf{G}^r \mathbf{\Sigma}_1^< \mathbf{G}^a \mathbf{\Sigma}_\alpha^a\right] &= \sum_\beta \frac{eV_\beta(\omega)}{2\pi\hbar^2\omega} \int \mathrm{d}\varepsilon \, \mathbf{G}_+^r \underset{\cdot}{\mathbf{\Sigma}}{}_\beta^< \mathbf{G}^a \mathbf{\Sigma}_\alpha^a, \\ \mathcal{F}_t\left[\int \mathrm{d}\tau \, \mathbf{G}^r \mathbf{\Sigma}_{\alpha 1}^<\right] &= \frac{eV_\alpha(\omega)}{2\pi\hbar^2\omega} \int \mathrm{d}\varepsilon \, \mathbf{G}_+^r \underset{\cdot}{\mathbf{\Sigma}}{}_\alpha^<. \end{aligned} \tag{S3}$$

Notations $\mathbf{A} = \mathbf{A}\left(\varepsilon\right)$, $\mathbf{A}_+ = \mathbf{A}\left(\varepsilon + \hbar\omega\right)$, $\underset{\cdot}{\mathbf{A}} = \mathbf{A} - \mathbf{A}_+$ are used in eq S3. Choose $V_\alpha\left(t\right) = E\left(t\right)L/2$ and $V_\beta\left(t\right) = -E\left(t\right)L/2$, we have

$$\begin{aligned} I_\alpha^{(1,1)}(\omega) &= \frac{eE\left(\omega\right)}{2\pi\hbar} \sum_\beta \int \mathrm{d}\varepsilon \, \mathrm{tr}\left[\mathcal{G}_{\alpha\beta}^{(1,1)}\left(\varepsilon, \omega\right)\right], \\ \mathcal{G}_{\alpha\beta}^{(1,1)}\left(\varepsilon, \omega\right) &= \frac{eL}{2\hbar\omega}\left[\left(2\delta_{\alpha\beta} - 1\right)\left(\mathbf{G}_+^r \underset{\cdot}{\mathbf{\Sigma}}{}_\beta^< \mathbf{G}^a \mathbf{\Sigma}_\alpha^a - \mathbf{\Sigma}_\alpha^r \mathbf{G}_+^r \underset{\cdot}{\mathbf{\Sigma}}{}_\beta^< \mathbf{G}^a\right) + \delta_{\alpha\beta}\left(\mathbf{G}_+^r \underset{\cdot}{\mathbf{\Sigma}}{}_\beta^< - \underset{\cdot}{\mathbf{\Sigma}}{}_\beta^< \mathbf{G}^a\right)\right]. \end{aligned} \tag{S4}$$

The second part is

$$I_\alpha^{(1,2)}(t) = e\int \mathrm{d}\tau\, \mathrm{tr}\left[\mathbf{G}^r\mathbf{\Delta}\mathbf{G}^<\mathbf{\Sigma}_\alpha^a + \mathbf{G}^<\mathbf{\Delta}\mathbf{G}^a\mathbf{\Sigma}_\alpha^a + \mathbf{G}^r\mathbf{\Delta}\mathbf{G}^r\mathbf{\Sigma}_\alpha^< + \mathbf{h.c.}\right]. \tag{S5}$$

Perform Fourier transform from t to ω,

$$I_\alpha^{(1,2)}(\omega) = \frac{e}{2\pi\hbar}\int \mathrm{d}\varepsilon\, \mathrm{tr}\left[\mathbf{G}_+^r\mathbf{\Delta}(\omega)\mathbf{G}^<\mathbf{\Sigma}_\alpha^a + \mathbf{G}_+^<\mathbf{\Delta}(\omega)\mathbf{G}^a\mathbf{\Sigma}_\alpha^a + \mathbf{G}_+^r\mathbf{\Delta}(\omega)\mathbf{G}^r\mathbf{\Sigma}_\alpha^< + \mathbf{h.c.}\right]. \tag{S6}$$

Set $\omega = 0$ and denote $\mathbf{\Lambda}_\alpha = \mathrm{Im}\left[\mathbf{\Sigma}_\alpha^a\right]$, we have

$$\begin{aligned} J_\alpha^{(1,2)} &= \frac{ie}{\pi\hbar}\int \mathrm{d}\varepsilon\, \mathrm{tr}\left[\mathbf{\Delta}(\omega=0)\left(\mathbf{G}^<\mathbf{\Lambda}_\alpha\mathbf{G}^r + \mathbf{G}^a\mathbf{\Lambda}_\alpha\mathbf{G}^< + F\,\mathbf{G}^r\mathbf{\Lambda}_\alpha\mathbf{G}^r - F\,\mathbf{G}^a\mathbf{\Lambda}_\alpha\mathbf{G}^a\right)\right] \\ &= 0. \end{aligned} \tag{S7}$$

$F(\varepsilon)$ is the Fermi-Dirac distribution, and $\mathbf{G}^< = -F\left[\mathbf{G}^r - \mathbf{G}^a\right]$ is used in the last line of eq S7.

The first order integrated current is

$$\begin{aligned} J_\alpha^{(1)} &= \frac{eE(\omega=0)}{2\pi\hbar}\sum_\beta \int \mathrm{d}\varepsilon\, \mathrm{tr}\left[\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(1)}(\varepsilon)\right], \\ \boldsymbol{\mathcal{G}}_{\alpha\beta}^{(1)}(\varepsilon) &= \lim_{\omega\to 0}\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(1,1)}(\varepsilon,\omega). \end{aligned} \tag{S8}$$

Second Order Integrated Current

Following the split scheme used in the first order integrated current, the first part of second order current is

$$I_\alpha^{(2,1)}(t) = e\int \mathrm{d}\tau\, \mathrm{tr}\left[\mathbf{G}^r\mathbf{\Sigma}_2^<\mathbf{G}^a\mathbf{\Sigma}_\alpha^a + \mathbf{G}^r\mathbf{\Sigma}_{\alpha 2}^< + \mathbf{h.c.}\right]. \tag{S9}$$

Perform Fourier transform from t to ω and set $\omega = 0$,

$$
\begin{aligned}
J_\alpha^{(2,1)} &= \frac{e}{4\pi^2\hbar^2}\sum_\beta \int \mathrm{d}\varepsilon_1 E\left(-\varepsilon_1/\hbar\right)E\left(\varepsilon_1/\hbar\right)\int \mathrm{d}\varepsilon\, \mathrm{tr}\left[\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(2,1)}\left(\varepsilon,\varepsilon_1\right)\right],\\
\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(2,1)}\left(\varepsilon,\varepsilon_1\right) &= -\frac{e^2L^2}{4\varepsilon_1^2}\left[\mathbf{G}^r\underline{\mathbf{\Sigma}}_\beta^<\mathbf{G}^a\mathbf{\Sigma}_\alpha^a - \mathbf{\Sigma}_\alpha^r\mathbf{G}^r\underline{\mathbf{\Sigma}}_\beta^<\mathbf{G}^a + \delta_{\alpha\beta}\left(\mathbf{G}^r\underline{\mathbf{\Sigma}}_\beta^< - \underline{\mathbf{\Sigma}}_\beta^<\mathbf{G}^a\right)\right].
\end{aligned}
\tag{S10}
$$

Notation $\underline{\mathbf{A}} = \mathbf{A} - \mathbf{A}\left(\varepsilon+\varepsilon_1\right)$ is used. Plug the relation $\mathbf{\Sigma}_\alpha^<\left(\varepsilon\right) = 2i\,\mathbf{\Lambda}_\alpha\, F\left(\varepsilon\right)$ into eq S10, it can be shown that

$$
J_\alpha^{(2,1)} = 0. \tag{S11}
$$

The second part is

$$
\begin{aligned}
I_\alpha^{(2,2)}(t) = e\int \mathrm{d}\tau\, \mathrm{tr}\Big[&\mathbf{G}^r\mathbf{\Delta}\mathbf{G}^r\mathbf{\Delta}\mathbf{G}^<\mathbf{\Sigma}_\alpha^a + \mathbf{G}^<\mathbf{\Delta}\mathbf{G}^a\mathbf{\Delta}\mathbf{G}^a\mathbf{\Sigma}_\alpha^a\\
&+\mathbf{G}^r\mathbf{\Delta}\mathbf{G}^<\mathbf{\Delta}\mathbf{G}^a\mathbf{\Sigma}_\alpha^a + \mathbf{G}^r\mathbf{\Delta}\mathbf{G}^r\mathbf{\Delta}\mathbf{G}^r\mathbf{\Sigma}_\alpha^< + \mathbf{G}^r\mathbf{\Delta}\mathbf{G}^r\mathbf{\Sigma}_{\alpha 1}^<\\
&+\mathbf{G}^r\mathbf{\Delta}\mathbf{G}^r\mathbf{\Sigma}_1^<\mathbf{G}^a\mathbf{\Sigma}_\alpha^a + \mathbf{G}^r\mathbf{\Sigma}_1^<\mathbf{G}^a\mathbf{\Delta}\mathbf{G}^a\mathbf{\Sigma}_\alpha^a + \mathbf{h.c.}\Big].
\end{aligned}
\tag{S12}
$$

Perform Fourier transform from t to ω and set $\omega = 0$,

$$
\begin{aligned}
J_\alpha^{(2,2)} &= \frac{e}{4\pi^2\hbar^2}\sum_\beta \int \mathrm{d}\varepsilon_1\, E\left(-\varepsilon_1/\hbar\right)E\left(\varepsilon_1/\hbar\right)\int \mathrm{d}\varepsilon\, \mathrm{tr}\left[\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(2,2)}\left(\varepsilon,\varepsilon_1\right)\right],\\
\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(2,2)}\left(\varepsilon,\varepsilon_1\right) &= \delta_{\alpha\beta}\Big(\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+1}^r\boldsymbol{\mu}\mathbf{G}^<\mathbf{\Sigma}_\alpha^a + \mathbf{G}^<\boldsymbol{\mu}\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\mathbf{\Sigma}_\alpha^a\\
&\quad+\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+1}^<\boldsymbol{\mu}\mathbf{G}^a\mathbf{\Sigma}_\alpha^a + \mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+1}^r\boldsymbol{\mu}\mathbf{G}^r\mathbf{\Sigma}_\alpha^< - \frac{eL}{2\varepsilon_1}\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+1}^r\underline{\mathbf{\Sigma}}_\alpha^<\Big)\\
&\quad-\frac{eL}{2\varepsilon_1}\left(2\delta_{\alpha\beta}-1\right)\left(\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+1}^r\underline{\mathbf{\Sigma}}_\beta^<\mathbf{G}^a\mathbf{\Sigma}_\alpha^a + \mathbf{G}^r\underline{\mathbf{\Sigma}}_\beta^<\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\mathbf{\Sigma}_\alpha^a\right)\\
&\quad+\mathbf{h.c.}.
\end{aligned}
\tag{S13}
$$

Notations $\mathbf{A}_{+1} = \mathbf{A}\left(\varepsilon+\varepsilon_1\right)$, $\underline{\mathbf{A}} = \mathbf{A} - \mathbf{A}_{+1}$ are used.

The second order integrated current is

$$J_{\alpha}^{(2)}=\frac{e}{4\pi^2\hbar^2}\sum_{\beta}\int \mathrm{d}\varepsilon_1\, E\left(-\varepsilon_1/\hbar\right)E\left(\varepsilon_1/\hbar\right)\int \mathrm{d}\varepsilon\,\mathrm{tr}\left[\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(2)}\left(\varepsilon,\varepsilon_1\right)\right],$$
$$\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(2)}\left(\varepsilon,\varepsilon_1\right)=\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(2,2)}\left(\varepsilon,\varepsilon_1\right). \tag{S14}$$

Third Order Integrated Current

Following the split scheme used in the first order integrated current, the first part of third order current is

$$I_{\alpha}^{(3,1)}(t)=e\int \mathrm{d}\tau\,\mathrm{tr}\left[\mathbf{G}^r\boldsymbol{\Sigma}_3^<\mathbf{G}^a\boldsymbol{\Sigma}_\alpha^a+\mathbf{G}^r\boldsymbol{\Sigma}_{\alpha 3}^<+\mathrm{h.c.}\right]. \tag{S15}$$

Perform Fourier transform from t to ω and set $\omega=0$,

$$J_{\alpha}^{(3,1)}=\frac{e}{8\pi^3\hbar^3}\sum_{\beta}\iint \mathrm{d}\varepsilon_2\mathrm{d}\varepsilon_1\, E\left(\frac{\varepsilon_1}{\hbar}\right)E\left(\frac{\varepsilon_2-\varepsilon_1}{\hbar}\right)E\left(-\frac{\varepsilon_2}{\hbar}\right)\int \mathrm{d}\varepsilon\,\mathrm{tr}\left[\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(3,1)}\left(\varepsilon,\varepsilon_1,\varepsilon_2\right)\right],$$
$$\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(3,1)}\left(\varepsilon,\varepsilon_1,\varepsilon_2\right)=\frac{e^3L^3}{16\varepsilon_1\left(\varepsilon_1-\varepsilon_2\right)\varepsilon_2}\left[\left(2\delta_{\alpha\beta}-1\right)\left(\mathbf{G}^r\underset{\sim}{\boldsymbol{\Sigma}}{}_\beta^<\mathbf{G}^a\boldsymbol{\Sigma}_\alpha^a-\boldsymbol{\Sigma}_\alpha^r\mathbf{G}^r\underset{\sim}{\boldsymbol{\Sigma}}{}_\beta^<\mathbf{G}^a\right)+\delta_{\alpha\beta}\left(\mathbf{G}^r\underset{\sim}{\boldsymbol{\Sigma}}{}_\beta^<-\underset{\sim}{\boldsymbol{\Sigma}}{}_\beta^<\mathbf{G}^a\right)\right].$$

(S16)

Notation $\underset{\sim}{\mathbf{A}}=\mathbf{A}\left(\varepsilon+\varepsilon_1\right)-\mathbf{A}\left(\varepsilon+\varepsilon_2\right)$ is used. The second part is

$$\begin{aligned}I_{\alpha}^{(3,2)}(t)=e\int \mathrm{d}\tau\,\mathrm{tr}\Big[&\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^<\boldsymbol{\Sigma}_\alpha^a+\mathbf{G}^<\boldsymbol{\Delta}\mathbf{G}^a\boldsymbol{\Delta}\mathbf{G}^a\boldsymbol{\Delta}\mathbf{G}^a\boldsymbol{\Sigma}_\alpha^a\\&+\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^<\boldsymbol{\Delta}\mathbf{G}^a\boldsymbol{\Sigma}_\alpha^a+\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^<\boldsymbol{\Delta}\mathbf{G}^a\boldsymbol{\Delta}\mathbf{G}^a\boldsymbol{\Sigma}_\alpha^a\\&+\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^r\boldsymbol{\Sigma}_\alpha^<+\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^r\boldsymbol{\Sigma}_{\alpha 1}^<\\&+\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^r\boldsymbol{\Sigma}_1^<\mathbf{G}^a\boldsymbol{\Sigma}_\alpha^a+\mathbf{G}^r\boldsymbol{\Sigma}_1^<\mathbf{G}^a\boldsymbol{\Delta}\mathbf{G}^a\boldsymbol{\Delta}\mathbf{G}^a\boldsymbol{\Sigma}_\alpha^a\\&+\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^r\boldsymbol{\Sigma}_1^<\mathbf{G}^a\boldsymbol{\Delta}\mathbf{G}^a\boldsymbol{\Sigma}_\alpha^a+\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^r\boldsymbol{\Sigma}_{\alpha_2}^<\\&+\mathbf{G}^r\boldsymbol{\Delta}\mathbf{G}^r\boldsymbol{\Sigma}_2^<\mathbf{G}^a\boldsymbol{\Sigma}_\alpha^a+\mathbf{G}^r\boldsymbol{\Sigma}_2^<\mathbf{G}^a\boldsymbol{\Delta}\mathbf{G}^a\boldsymbol{\Sigma}_\alpha^a+\mathbf{h.c.}\Big].\end{aligned} \tag{S17}$$

Perform Fourier transform from t to ω and set $\omega=0$,

$$
\begin{aligned}
J_{\alpha}^{(3,2)} &= \frac{e}{8\pi^3\hbar^3}\sum_{\beta}\iint \mathrm{d}\varepsilon_2 \mathrm{d}\varepsilon_1\, E\left(\frac{\varepsilon_1}{\hbar}\right) E\left(\frac{\varepsilon_2-\varepsilon_1}{\hbar}\right) E\left(-\frac{\varepsilon_2}{\hbar}\right)\int \mathrm{d}\varepsilon\, \mathrm{tr}\left[\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(3,2)}\left(\varepsilon,\varepsilon_1,\varepsilon_2\right)\right],\\
\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(3,2)}\left(\varepsilon,\varepsilon_1,\varepsilon_2\right) &= -\delta_{\alpha\beta}\left(\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^r\boldsymbol{\mu}\mathbf{G}_{+1}^r\boldsymbol{\mu}\mathbf{G}^<\mathbf{\Sigma}_{\alpha}^a - \mathbf{\Sigma}_{\alpha}^r\mathbf{G}^<\boldsymbol{\mu}\mathbf{G}_{+2}^a\boldsymbol{\mu}\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\right)\\
&\quad -\delta_{\alpha\beta}\left(\mathbf{G}^<\boldsymbol{\mu}\mathbf{G}_{+2}^a\boldsymbol{\mu}\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\mathbf{\Sigma}_{\alpha}^a - \mathbf{\Sigma}_{\alpha}^r\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^r\boldsymbol{\mu}\mathbf{G}_{+1}^r\boldsymbol{\mu}\mathbf{G}^<\right)\\
&\quad -\delta_{\alpha\beta}\left(\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^r\boldsymbol{\mu}\mathbf{G}_{+1}^<\boldsymbol{\mu}\mathbf{G}^a\mathbf{\Sigma}_{\alpha}^a - \mathbf{\Sigma}_{\alpha}^r\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^<\boldsymbol{\mu}\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\right)\\
&\quad -\delta_{\alpha\beta}\left(\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^<\boldsymbol{\mu}\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\mathbf{\Sigma}_{\alpha}^a - \mathbf{\Sigma}_{\alpha}^r\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^r\boldsymbol{\mu}\mathbf{G}_{+1}^<\boldsymbol{\mu}\mathbf{G}^a\right)\\
&\quad -\delta_{\alpha\beta}\left(\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^r\boldsymbol{\mu}\mathbf{G}_{+1}^r\boldsymbol{\mu}\mathbf{G}^r\mathbf{\Sigma}_{\alpha}^< - \mathbf{\Sigma}_{\alpha}^<\mathbf{G}^a\boldsymbol{\mu}\mathbf{G}_{+2}^a\boldsymbol{\mu}\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\right)\\
&\quad +\frac{eL}{2}\delta_{\alpha\beta}\left[\frac{1}{\varepsilon_1}\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^r\boldsymbol{\mu}\mathbf{G}_{+1}^r\underline{\mathbf{\Sigma}}_{\alpha}^< - \frac{1}{\varepsilon_2}\underline{\underline{\mathbf{\Sigma}}}_{\alpha}^<\mathbf{G}_{+2}^a\boldsymbol{\mu}\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\right]\\
&\quad +\frac{eL}{2}\left(2\delta_{\alpha\beta}-1\right)\left[\frac{1}{\varepsilon_1}\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^r\boldsymbol{\mu}\mathbf{G}_{+1}^r\underline{\mathbf{\Sigma}}_{\beta}^<\mathbf{G}^a\mathbf{\Sigma}_{\alpha}^a - \frac{1}{\varepsilon_2}\mathbf{\Sigma}_{\alpha}^r\mathbf{G}^r\underline{\underline{\mathbf{\Sigma}}}_{\beta}^<\mathbf{G}_{+2}^a\boldsymbol{\mu}\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\right]\\
&\quad +\frac{eL}{2}\left(2\delta_{\alpha\beta}-1\right)\left[\frac{1}{\varepsilon_2}\mathbf{G}^r\underline{\underline{\mathbf{\Sigma}}}_{\beta}^<\mathbf{G}_{+2}^a\boldsymbol{\mu}\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\mathbf{\Sigma}_{\alpha}^a - \frac{1}{\varepsilon_1}\mathbf{\Sigma}_{\alpha}^r\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^r\boldsymbol{\mu}\mathbf{G}_{+1}^r\underline{\mathbf{\Sigma}}_{\beta}^<\mathbf{G}^a\right]\\
&\quad +\frac{eL\left(2\delta_{\alpha\beta}-1\right)}{2\left(\varepsilon_2-\varepsilon_1\right)}\left[\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^r\underset{\sim}{\mathbf{\Sigma}}_{\beta}^<\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\mathbf{\Sigma}_{\alpha}^a - \mathbf{\Sigma}_{\alpha}^r\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^r\underset{\sim}{\mathbf{\Sigma}}_{\beta}^<\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\right]\\
&\quad +\frac{e^2L^2\delta_{\alpha\beta}}{8\left(\varepsilon_2-\varepsilon_1\right)}\left[\frac{1}{\varepsilon_1}\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^r\left(\underset{\sim}{\mathbf{\Sigma}}_{\alpha}^< - \underline{\mathbf{\Sigma}}_{\alpha}^<\right) - \frac{1}{\varepsilon_2}\left(\underset{\sim}{\mathbf{\Sigma}}_{\alpha}^< + \underline{\underline{\mathbf{\Sigma}}}_{\alpha}^<\right)\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\right]\\
&\quad +\frac{e^2L^2}{8\left(\varepsilon_2-\varepsilon_1\right)}\left[\frac{1}{\varepsilon_1}\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^r\left(\underset{\sim}{\mathbf{\Sigma}}_{\beta}^< - \underline{\mathbf{\Sigma}}_{\beta}^<\right)\mathbf{G}^a\mathbf{\Sigma}_{\alpha}^a - \frac{1}{\varepsilon_2}\mathbf{\Sigma}_{\alpha}^r\mathbf{G}^r\left(\underset{\sim}{\mathbf{\Sigma}}_{\beta}^< + \underline{\underline{\mathbf{\Sigma}}}_{\beta}^<\right)\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\right]\\
&\quad +\frac{e^2L^2}{8\left(\varepsilon_2-\varepsilon_1\right)}\left[\frac{1}{\varepsilon_2}\mathbf{G}^r\left(\underset{\sim}{\mathbf{\Sigma}}_{\beta}^< + \underline{\underline{\mathbf{\Sigma}}}_{\beta}^<\right)\mathbf{G}_{+1}^a\boldsymbol{\mu}\mathbf{G}^a\mathbf{\Sigma}_{\alpha}^a - \frac{1}{\varepsilon_1}\mathbf{\Sigma}_{\alpha}^r\mathbf{G}^r\boldsymbol{\mu}\mathbf{G}_{+2}^r\left(\underset{\sim}{\mathbf{\Sigma}}_{\beta}^< - \underline{\mathbf{\Sigma}}_{\beta}^<\right)\mathbf{G}^a\right].
\end{aligned}
$$

(S18)

Notations $\mathbf{A}_{+1}=\mathbf{A}\left(\varepsilon+\varepsilon_1\right)$, $\mathbf{A}_{+2}=\mathbf{A}\left(\varepsilon+\varepsilon_2\right)$, $\underline{\mathbf{A}}=\mathbf{A}-\mathbf{A}_{+1}$, $\underline{\underline{\mathbf{A}}}=\mathbf{A}-\mathbf{A}_{+2}$, $\underset{\sim}{\mathbf{A}}=\mathbf{A}_{+1}-\mathbf{A}_{+2}$ are used.

The third order integrated current is

$$J_{\alpha}^{(3)} = \frac{e}{8\pi^3\hbar^3}\sum_{\beta}\iint \mathrm{d}\varepsilon_2 \mathrm{d}\varepsilon_1\, E\left(\frac{\varepsilon_1}{\hbar}\right) E\left(\frac{\varepsilon_2-\varepsilon_1}{\hbar}\right) E\left(-\frac{\varepsilon_2}{\hbar}\right) \int \mathrm{d}\varepsilon\, \mathrm{tr}\left[\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(3)}\left(\varepsilon,\varepsilon_1,\varepsilon_2\right)\right],$$

$$\boldsymbol{\mathcal{G}}_{\alpha\beta}^{(3)}\left(\varepsilon,\varepsilon_1,\varepsilon_2\right) = \boldsymbol{\mathcal{G}}_{\alpha\beta}^{(3,1)}\left(\varepsilon,\varepsilon_1,\varepsilon_2\right) + \boldsymbol{\mathcal{G}}_{\alpha\beta}^{(3,2)}\left(\varepsilon,\varepsilon_1,\varepsilon_2\right). \tag{S19}$$