This note is used to show how to derive linear and non-linear conductance from current-current correlation function. (References are Mahan and BAB's books)

First, we shall introduce our Hamiltonian in first-quantized form with periodic-perturbated electric field

$$H = \sum_i \frac{1}{2m}\left(p_i - \frac{e}{c}A(x_i,t)\right)^2 + \sum_{i<j} V_{ij}$$

$$= \sum_i \frac{1}{2m}{p_i}^2 + \sum_{i<j} V_{ij} - \sum_i \frac{e}{2mc}\left(p_i \cdot A(x_i,t) + A(x_i,t)\cdot p_i\right) + \sum_i \frac{e^2}{2mc^2}A^2(x_i,t)$$

$$= H_0 + H'(t)$$

According to Schrödinger equation, when choosing gauge $\nabla \cdot A = 0$, one can derive particle density and particle current density

$$\rho(x) = N\int Dx_{i\neq 1}\Psi^*(x,x_2,\dots)\Psi(x,x_2,\dots)$$

$$J(\mathrm{x,t}) = \frac{N}{2m}\int Dx_{i\neq 1}\left[\Psi^*(x,x_2,\dots)\left(p - \frac{e}{c}A(x,t)\right)\Psi(x,x_2,\dots) + c.c.\right]$$

This particle density and particle current density satisfy continuous equation

$$-\frac{\partial\rho(x,t)}{\partial t} = \nabla\cdot\frac{N}{2m}\int Dx_{i\neq 1}\left[\Psi^*(x,x_2,\dots)\left(p - \frac{e}{c}A(x,t)\right)\Psi(x,x_2,\dots) + c.c.\right]$$

Current density operator below can also be proved by directly comparing $\langle J\rangle$ and $J(\mathrm{x,t})$

$$J = \frac{1}{2m}\sum_i\left[\left(p_i - \frac{e}{c}A(x_i,t)\right)\delta(x - x_i) + \delta(x - x_i)\left(p_i - \frac{e}{c}A(x_i,t)\right)\right]$$

One can see interaction only influences wave function but not current density operator from this picture. Now let's transform it into lattice second-quantized picture, the general Hamiltonian H_0 in momentum space is

$$H_0 = \sum_k h_k c_k^\dagger c_k + \sum_{q\neq 0}\frac{V(q)}{2\Omega}\left(\sum_{k_1} c_{k_1}^\dagger c_{k_1+q}\sum_{k_2} c_{k_2}^\dagger c_{k_2-q}\right)$$

After transforming charge density operator $\rho_i \equiv c_i^\dagger c_i$ into momentum space $\rho_q = \frac{1}{\sqrt{N}}\sum_k c_{k+q}^\dagger c_k$, we can write down continuous equation in momentum space

$$i\frac{1}{\sqrt{N}}\sum_k\left(h_{k+q} - h_k\right)c_{k+q}^\dagger c_k = i\left[H_0,\rho_q\right] = \frac{\partial\rho_q}{\partial t} = iq\cdot j_q$$

When q is small, we have Fourier transformed current density operator written as

$$j_q \approx \frac{1}{\sqrt{N}}\sum_k\frac{\partial h_k}{\partial k_q}c_{k+\frac{q}{2}}^\dagger c_{k-\frac{q}{2}}$$

We can see again, interaction between particles will not influence the form of current density operator, since we have not considered vector potential for now, we use j to label this current density operator for distinguishing J for total current. According to our first-quantized result, the average of current density operator is

$$\langle J(\mathrm{x,t})\rangle = \frac{\mathrm{N}}{m\Omega}\langle p - \frac{e}{c}A(x,t)\rangle = \langle j(x,t)\rangle - \frac{n_0 e}{mc}A(x,t)$$

According to Taylor expansion, one should expect

$$\langle J_\alpha(\mathrm{x,t})\rangle = \sum_\beta \sigma_{\alpha\beta}E_\beta(x,t) + \sum_{\beta,\gamma}\chi_{\alpha\beta\gamma}E_\beta(x,t)E_\gamma(x,t)$$

Since $E(x,t) = -\frac{1}{c}\frac{\partial A(x,t)}{\partial t}$, we simply choose $A(x,t) = E\cdot e^{i(qx-wt)}$ so that $E(x,t) = \frac{iw}{c}E\cdot e^{i(qx-wt)}$. After this gauge-choosing step, term $\frac{n_0 e}{mc}A(x,t)$ in $\langle J(\mathrm{x,t})\rangle$ is just $\frac{n_0 e}{mc}E\cdot e^{i(qx-wt)}$. One only needs to compute $\langle j(x,t)\rangle$. We consider this computation below in interaction picture.

$$\langle j(x,t)\rangle = \left\langle \psi \middle| T\left[e^{i\int_{-\infty}^{t} H'(t_1)dt_1}\right] j(x,t) T\left[e^{-i\int_{-\infty}^{t} H'(t_1)dt_1}\right] \middle| \psi \right\rangle$$

Here $|\psi\rangle$ is the state without perturbation $H'(t)$. After expanding $T\left[e^{-i\int_{-\infty}^{t} H'(t_1)dt_1}\right]$ up to the second order of $H'(t)$

$$T\left[e^{-i\int_{-\infty}^{t} H'(t_1)dt_1}\right]$$
$$\approx 1 - i\int_{-\infty}^{t} H'(t_1)dt_1 - \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 H'(t_1)H'(t_2)$$

By noticing $\sum_i \frac{e^2}{2mc^2} A^2(x_i,t) = \frac{e^2}{2mc^2}\int dx\, \sum_i \delta(x-x_i)\cdot A^2(x,t) = \frac{Ne^2E^2}{2mc^2} e^{i2(qx-wt)}$ which commutes with any particle-conserved operator and the left part in $H'(t)$ is just $-\sum_i \frac{e}{2mc}(p_i\cdot A(x_i,t) + A(x_i,t)\cdot p_i) = -\frac{e}{c}\int dx\, j\cdot A(x,t) = -\frac{eE}{c}\cdot j_q e^{-iwt}$, keeping linear E and E^2 in the result, one finds for linear order

$$\langle j(x,t)\rangle_1 = i\frac{eE}{c}\int_{-\infty}^{t} e^{-iwt'}\langle\psi|[j(x,t), j(q,t')]|\psi\rangle dt'$$

For the second order

$$\langle j(x,t)\rangle_2 = -\frac{e^2E^2}{c^2}\int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 e^{-iwt_1}e^{-iwt_2}\langle\psi|j(q,t_2)j(q,t_1)j(x,t) + j(x,t)j(q,t_1)j(q,t_2)|\psi\rangle$$
$$+\frac{e^2E^2}{c^2}\int_{-\infty}^{t} dt_1 \int_{-\infty}^{t} dt_2 e^{-iwt_1}e^{-iwt_2}\langle\psi|j(q,t_1)j(x,t)j(q,t_2)|\psi\rangle$$

Compare these results with $\langle J_\alpha(\mathrm{x,t})\rangle = \sum_\beta \sigma_{\alpha\beta}E_\beta(x,t) + \sum_{\beta,\gamma}\chi_{\alpha\beta\gamma}E_\beta(x,t)E_\gamma(x,t)$, we derive linear conductance

$$\sigma_{\alpha\beta}(q,w) = \frac{e}{w\Omega}\int_0^\infty dt e^{iwt}\langle\psi|\left[j_\alpha(-q,t), j_\beta(q,0)\right]|\psi\rangle + i\frac{n_0 e}{mw}\delta_{\alpha,\beta}$$

And nonlinear conductance

$$\chi_{\alpha\beta\gamma}(q,w) = \frac{e^2}{w^2\Omega}\iint_0^\infty e^{iw(2t_1+t_2)}\langle\psi|j_\gamma(q,0)j_\beta(q,t_2)j_\alpha(-2q,t_1+t_2) + j_\alpha(-2q,t_1+t_2)j_\beta(q,t_2)j_\gamma(q,0)|\psi\rangle$$
$$- e^{iw(t_1+t_2)}\langle\psi|j_\beta(q,t_2)j_\alpha(-2q,t_1+t_2)j_\gamma(q,t_1)|\psi\rangle dt_1 dt_2$$

Now, we would like to compute $\sigma_{\alpha\beta}(q,w)$ and $\chi_{\alpha\beta\gamma}(q,w)$ for a given single-particle-production state $|\psi\rangle$.

Let's look at $\sigma_{\alpha\beta}(q,w)$ first, we would like to derive $Re\left(\sigma_{\alpha\beta}(q,w)\right)$,

$$\frac{e}{w\Omega}\int_0^\infty dt e^{iwt}\langle\psi|\left[j_\alpha(-q,t), j_\beta(q,0)\right]|\psi\rangle$$

$$= \frac{e}{w\Omega}\int_0^\infty dt e^{iwt}\sum_{k_1,k_2}\frac{\partial h_{k_1}^{m_1,n_1}}{\partial k_{1\alpha}}\frac{\partial h_{k_2}^{m_2,n_2}}{\partial k_{2\beta}}\Big(\langle c^\dagger_{k_1-\frac{q}{2},m_1}(t)c_{k_1+\frac{q}{2},n_1}(t)c^\dagger_{k_2+\frac{q}{2},m_2}c_{k_2-\frac{q}{2},n_2}\rangle$$
$$-\langle c^\dagger_{k_2+\frac{q}{2},m_2}c_{k_2-\frac{q}{2},n_2}c^\dagger_{k_1-\frac{q}{2},m_1}(t)c_{k_1+\frac{q}{2},n_1}(t)\rangle\Big)$$

$$= \frac{e}{w\Omega}\int_0^\infty dt e^{iwt}\sum_k \frac{\partial h_k^{n_2,n_1}}{\partial k_\alpha}\frac{\partial h_k^{n_1,n_2}}{\partial k_\beta}\Big(\langle c^\dagger_{k-\frac{q}{2},n_2}(t)c_{k-\frac{q}{2},n_2}\rangle\langle c_{k+\frac{q}{2},n_1}(t)c^\dagger_{k+\frac{q}{2},n_1}\rangle - \langle c^\dagger_{k+\frac{q}{2},n_1}c_{k+\frac{q}{2},n_1}(t)\rangle\langle c_{k-\frac{q}{2},n_2}c^\dagger_{k-\frac{q}{2},n_2}(t)\rangle\Big)$$

Here we chose $c^\dagger_{k_2+\frac{q}{2},m_2}$ as creating operator for m_2 band with momentum $k_2+\frac{q}{2}$ in single-particle-production basis. Now the kinetic band is not necessarily diagonalized. Take $q\to 0$ and write $\langle c_{k+\frac{q}{2},n_1}(t)c^\dagger_{k+\frac{q}{2},n_1}\rangle$ with equal time Green's function

$$\frac{e}{w\Omega}\int_0^\infty dte^{iwt}\sum_k \frac{\partial h_k^{n_2,n_1}}{\partial k_\alpha}\frac{\partial h_k^{n_1,n_2}}{\partial k_\beta}\left(e^{-i(\varepsilon_{k,n_2}+\varepsilon_{k,n_1})t}\left(I-G_{n_2,n_2}(k)\right)G_{n_1,n_1}(k)-e^{i(\varepsilon_{k,n_1}+\varepsilon_{k,n_2})t}\left(I-G_{n_1,n_1}(k)\right)G_{n_2,n_2}(k)\right)$$

Here ε_{k,n_2} is the excitation energy on band n_2 with momentum k and $G_{n_1,n_1}(k)$ is a diagonal matrix representing $\langle c_{k,n_1}c_{k,n_1}^\dagger\rangle$. Now we can compute the integral $\int_0^\infty dte^{iwt}e^{-i(\varepsilon_{k,n_2}+\varepsilon_{k,n_1})t}$ by timing a converging term $e^{-\delta t}$.

$$\frac{e}{w\Omega}Re\left[\sum_k \frac{\partial h_k^{n_2,n_1}}{\partial k_\alpha}\frac{\partial h_k^{n_1,n_2}}{\partial k_\beta}\left(\frac{1}{\delta-i\left(w-\left(\varepsilon_{k,n_2}+\varepsilon_{k,n_1}\right)\right)}\left(I-G_{n_2,n_2}(k)\right)G_{n_1,n_1}(k)\right.\right.$$

$$\left.\left.-\frac{1}{\delta-i\left(w+\left(\varepsilon_{k,n_2}+\varepsilon_{k,n_1}\right)\right)}\left(I-G_{n_1,n_1}(k)\right)G_{n_2,n_2}(k)\right)\right]$$

We would like to derive this expression with $\delta\to 0, w\to 0$ according to L'Hôpital's rule

$$\frac{e}{\Omega}Re\left[\sum_k \frac{\partial h_k^{n_2,n_1}}{\partial k_\alpha}\frac{\partial h_k^{n_1,n_2}}{\partial k_\beta}\frac{-i}{\left(\varepsilon_{k,n_2}+\varepsilon_{k,n_1}\right)^2}\left(G_{n_1,n_1}(k)-G_{n_2,n_2}(k)\right)\right]$$

$$=\frac{e}{\Omega}Im\left[\sum_k\left(\frac{\partial h_k^{n_2,n_1}}{\partial k_\alpha}\frac{\partial h_k^{n_1,n_2}}{\partial k_\beta}\frac{1}{\left(\varepsilon_{k,n_2}+\varepsilon_{k,n_1}\right)^2}\left(G_{n_1,n_1}(k)-G_{n_2,n_2}(k)\right)\right)\right]$$

Let's check the effectiveness of this expression now. It is straightforward to see there is no contribution from two occupied bands or two empty bands, or the case $\alpha=\beta$. Then only Hall conductance from occupied and empty bands is non-zero.

$$\frac{e}{\Omega}Im\left[\sum_{k,n_O,n_E}\frac{\left(\frac{\partial h_k^{n_O,n_E}}{\partial k_\alpha}\frac{\partial h_k^{n_E,n_O}}{\partial k_\beta}-\frac{\partial h_k^{n_E,n_O}}{\partial k_\alpha}\frac{\partial h_k^{n_O,n_E}}{\partial k_\beta}\right)}{\left(\varepsilon_{k,n_O}+\varepsilon_{k,n_E}\right)^2}\right]$$

Here n_O means occupied bands and n_E means empty bands. It is interesting this expression is similar with Kubo formula in Thouless's paper. If it is the tight binding model without $\sum_{i<j}V_{ij}$, we can see $\varepsilon_k=h_k$ and the expression will degenerate to the Thouless's one where conductance is quantized by integrating Berry curvature.

After we have the knowledge above, we are ready to derive the lowest order nonlinear conductance.

$$\chi_{\alpha\beta\gamma}(q,w)=\frac{e^2}{w^2\Omega}\iint_0^\infty e^{iw(2t_1+t_2)}\langle\psi|j_\gamma(q,0)j_\beta(q,t_2)j_\alpha(-2q,t_1+t_2)+j_\alpha(-2q,t_1+t_2)j_\beta(q,t_2)j_\gamma(q,0)|\psi\rangle$$
$$-e^{iw(t_1+t_2)}\langle\psi|j_\beta(q,t_2)j_\alpha(-2q,t_1+t_2)j_\gamma(q,t_1)|\psi\rangle dt_1dt_2$$