1 Theory of Linear Response in a Nutshell

Classically Consider an electron system subject to small external perturbation $V_{\text{ext}}(\mathbf{r}, t)$. By definition of the *inverse dielectric function* the total potential $V(\mathbf{r}, t)$ is given by

$$V(\mathbf{r}, t) = \int \mathrm{d}^3 r' \int_{-\infty}^{t} \mathrm{d}t' \; \varepsilon^{-1}(\mathbf{r}, \mathbf{r}', t - t') V_{\text{ext}}(\mathbf{r}', t') \; . \tag{1}$$

The upper bound of the integral over t' is t in place of ∞ due to causality: the "effect" cannot precede the "cause". This is equivalent to saying that $\varepsilon^{-1}(\mathbf{r}, \mathbf{r}', \tau) = 0$ whenever $\tau < 0$. The Fourier transform of $\varepsilon^{-1}(\mathbf{r}, \mathbf{r}', \tau)$ is

$$\varepsilon^{-1}(\mathbf{r}, \mathbf{r}', \omega) = \int_{0}^{\infty} \mathrm{d}\tau \; e^{i\omega\tau} \, \varepsilon^{-1}(\mathbf{r}, \mathbf{r}', \tau) \; . \tag{2}$$

Applying Titchmarsh's theorem to ε^{-1}, we get that $\varepsilon^{-1}(\mathbf{r}, \mathbf{r}', \omega)$ is the limit $\eta \to 0+$ of $\varepsilon^{-1}(\mathbf{r}, \mathbf{r}', \omega + i\eta)$ which is holomorphic in the upper complex plane. Taking Fourier transform of eq. (1), we obtain[1]

$$V(\mathbf{r}, \omega) = \int \mathrm{d}^3 r' \; \varepsilon^{-1}(\mathbf{r}, \mathbf{r}', \omega) V_{\text{ext}}(\mathbf{r}', \omega) \; . \tag{3}$$

Eqs. (2) and (3) may equivalently be formulated as

$$\begin{aligned} V_{\text{ext}}(\mathbf{r}, \omega) &= \int \mathrm{d}^3 r' \varepsilon(\mathbf{r}, \mathbf{r}', \omega) V(\mathbf{r}') \; , \text{ where} \\ \varepsilon(\mathbf{r}, \mathbf{r}', \omega) &= \lim_{\eta \to 0+} \varepsilon(\mathbf{r}, \mathbf{r}', \omega + i\eta) = \lim_{\eta \to 0+} \int_{0}^{\infty} \mathrm{d}\tau \; e^{i(\omega + i\eta)\tau} \, \varepsilon(\mathbf{r}, \mathbf{r}', \tau) \; . \end{aligned} \tag{4}$$

Quantum Mechanically Now consider a system of non-interacting electrons described in a single-particle approximation by a Hamiltonian $\hat{H}_0$. Let E_i denote single-particle energy levels with corresponding eigenstates $|i\rangle$. One-particle density matrix is then

$$\hat{\rho}_0 = \sum_i n_i \, |i\rangle\langle i| \; , \tag{5}$$

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[1]

$$\begin{aligned} V(\mathbf{r}, \omega) &= \int_{-\infty}^{\infty} \mathrm{d}t \; e^{i\omega t} \, V(\mathbf{r}, t) \\ &= \int \mathrm{d}^3 r' \int_{-\infty}^{\infty} \mathrm{d}t \int_{-\infty}^{t} \mathrm{d}t' \; e^{i\omega t} \, \varepsilon^{-1}(\mathbf{r}, \mathbf{r}', t - t') V_{\text{ext}}(\mathbf{r}', t') \\ &= \int \mathrm{d}^3 r' \int_{-\infty}^{\infty} \frac{\mathrm{d}\omega'}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}t \int_{-\infty}^{t} \mathrm{d}t' \; e^{i\omega t} e^{-i\omega' t'} \varepsilon^{-1}(\mathbf{r}, \mathbf{r}', t - t') V_{\text{ext}}(\mathbf{r}', \omega') \\ &= \int \mathrm{d}^3 r' \int_{-\infty}^{\infty} \frac{\mathrm{d}\omega'}{2\pi} V_{\text{ext}}(\mathbf{r}', \omega') \int_{-\infty}^{\infty} \mathrm{d}t \int_{-\infty}^{t} \mathrm{d}t' \; e^{i(\omega - \omega')t} e^{i\omega'(t - t')} \, \varepsilon^{-1}(\mathbf{r}, \mathbf{r}', t - t') \\ &= \int \mathrm{d}^3 r' \int_{-\infty}^{\infty} \frac{\mathrm{d}\omega'}{2\pi} \; V_{\text{ext}}(\mathbf{r}', \omega') \int_{-\infty}^{\infty} \mathrm{d}t \, e^{i(\omega - \omega')t} \int_{\infty}^{0} (-\mathrm{d}\tau) \; e^{i\omega'\tau} \, \varepsilon^{-1}(\mathbf{r}, \mathbf{r}', \tau) \\ &= \int \mathrm{d}^3 r' \int_{-\infty}^{\infty} \mathrm{d}\omega' \, \varepsilon^{-1}(\mathbf{r}, \mathbf{r}', \omega') V_{\text{ext}}(\mathbf{r}', \omega') \cdot \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}t \; e^{i(\omega - \omega')t} \\ &= \int \mathrm{d}^3 r' \; \varepsilon^{-1}(\mathbf{r}, \mathbf{r}', \omega) V_{\text{ext}}(\mathbf{r}', \omega) \; . \end{aligned}$$

where n_i denotes the occupational number at energy E_i which, in equilibrium, is given by the Fermi-Dirac distribution. Electron density operator is $\hat{N}(\mathbf{r}) = |\mathbf{r}\rangle\langle\mathbf{r}|$. Equation of motion reads

$$i\hbar\frac{\mathrm{d}\hat{\rho}_0}{\mathrm{d}t} = [\hat{H}_0, \hat{\rho}_0] = 0 \ .$$

Within RPA (Random Phase Approximation) we are interested in the reponse of the system to the perturbation of the form $\hat{V}e^{-i(\omega+i\eta)t}$ (**TODO:** explain η). In the first order approximation, $\hat{\rho} = \hat{\rho}_0 + \hat{\rho}' + \mathcal{O}(\hat{V}^2)$, where $\hat{\rho}_0$ is defined by eq. (5) and $\hat{\rho}' \propto \hat{V}$. We thus have

$$\left.\begin{aligned} i\hbar\frac{\mathrm{d}\hat{\rho}}{\mathrm{d}t} &= i\hbar\frac{\mathrm{d}\hat{\rho}_0}{\mathrm{d}t} + i\hbar\frac{\mathrm{d}\hat{\rho}'}{\mathrm{d}t} \\ [\hat{H}, \hat{\rho}] &= [\hat{H}_0, \hat{\rho}_0] + [\hat{H}_0, \hat{\rho}'] + [\hat{V}, \hat{\rho}_0]e^{-i(\omega+i\eta)t} + \mathcal{O}(\hat{V}^2) \end{aligned}\right\} \implies i\hbar\frac{\mathrm{d}\hat{\rho}'}{\mathrm{d}t} = [\hat{H}_0, \hat{\rho}'] + [\hat{V}, \hat{\rho}_0]e^{-i(\omega+i\eta)t} \ . \tag{6}$$

Using an ansatz $\hat{\rho}' = \widetilde{G}\hat{V}e^{-i(\omega+i\eta)t}$, where $\widetilde{G}$ is some time-independent operator, we obtain[2]

$$\begin{aligned} \langle i|\widetilde{G}\hat{V}|j\rangle &= \frac{n_i - n_j}{E_i - E_j - \hbar(\omega + i\eta)}\langle i|\hat{V}|j\rangle \\ &\equiv \langle i|\hat{G}|j\rangle \, \langle i|\hat{V}|j\rangle \ , \end{aligned}$$

where we have defined a new operator $\hat{G}$ by

$$\langle i|\hat{G}|j\rangle = \frac{n_i - n_j}{E_i - E_j - \hbar(\omega + i\eta)} \ . \tag{7}$$

We can now calculate the induced electron density $\delta\hat{N}(t)$:

$$\begin{aligned} \langle \mathbf{r}|\delta\hat{N}(t)|\mathbf{r}\rangle &= \mathrm{Tr}(\hat{N}(\mathbf{r})\hat{\rho}) - \mathrm{Tr}(\hat{N}(\mathbf{r})\hat{\rho}_0) = \mathrm{Tr}(\hat{N}(\mathbf{r})\hat{\rho}') \\ &= \sum_{i,j}\langle j|\mathbf{r}\rangle\langle\mathbf{r}|i\rangle\langle i|\widetilde{G}\hat{V}|j\rangle e^{-i(\omega+i\eta)t} \\ &= \sum_{i,j}\langle i|\hat{G}|j\rangle\langle j|\mathbf{r}\rangle\langle\mathbf{r}|i\rangle\langle i|\hat{V}|j\rangle e^{-i(\omega+i\eta)t} \ . \end{aligned} \tag{8}$$

The total potential $\hat{V}$ is the sum of external potential $\hat{V}_{\mathrm{ext}}$ and the potential induced by the variation of the charge density, i.e.

$$\langle \mathbf{r}|\hat{V}_{\mathrm{tot}}(t)|\mathbf{r}\rangle = \langle \mathbf{r}|\hat{V}_{\mathrm{ext}}(t)|\mathbf{r}\rangle + \int \mathrm{d}^3r' \ \langle \mathbf{r}|\hat{V}_{\mathrm{Coulomb}}|\mathbf{r}'\rangle\langle\mathbf{r}'|\delta\hat{N}(t)|\mathbf{r}'\rangle \ , \tag{9}$$

where $\hat{V}_{\mathrm{Coulomb}}$ is the Coulomb interaction potential. We have also used the fact that $\hat{V}$ is

[2]Calculating matrix elements:

$$\begin{aligned} \langle i|i\hbar\frac{\mathrm{d}\hat{\rho}'}{\mathrm{d}t}|j\rangle &= \hbar(\omega + i\eta)\langle i|\hat{\rho}'|j\rangle \ , \\ \langle i|[\hat{H}_0, \hat{\rho}']|j\rangle &= (E_i - E_j)\langle i|\hat{\rho}'|j\rangle \ , \\ \langle i|[\hat{V}, \hat{\rho}_0]|j\rangle &= (n_j - n_i)\langle i|\hat{V}|j\rangle \ . \end{aligned}$$

Eq. (6) now reads

$$\langle i|\hat{\rho}'|j\rangle = \frac{(n_i - n_j)\, e^{-i\omega t + \eta t}}{E_i - E_j - \hbar(\omega + i\eta)}\langle i|\hat{V}|j\rangle \ .$$

diagonal in position representation. Using eq. (8) we get[3]

$$\langle \mathbf{r}|\hat{V}_{\text{ext}}(t)|\mathbf{r}\rangle = \int \mathrm{d}^3 r' \ \langle \mathbf{r}|\hat{\varepsilon}(t)|\mathbf{r}'\rangle\langle \mathbf{r}'|\hat{V}|\mathbf{r}'\rangle \ , \text{ where}$$

$$\langle \mathbf{r}|\hat{\varepsilon}(\tau)|\mathbf{r}'\rangle = \left(\langle \mathbf{r}|\mathbf{r}'\rangle - \sum_{i,j} \langle i|\hat{G}|j\rangle \int \mathrm{d}^3 r'' \ \frac{e^2}{\|\mathbf{r}-\mathbf{r}''\|} \ \langle j|\mathbf{r}''\rangle\langle \mathbf{r}''|i\rangle\langle i|\mathbf{r}'\rangle\langle \mathbf{r}'|j\rangle \right) \ e^{-i(\omega+i\eta)\tau} \ .$$

Using Titchmarsh's theorem once again, we obtain

$$\begin{aligned}
\langle \mathbf{r}|\hat{V}_{\text{ext}}(\omega)|\mathbf{r}\rangle &= \int \mathrm{d}^3 r' \ \langle \mathbf{r}|\hat{\varepsilon}(\omega)|\mathbf{r}'\rangle\langle \mathbf{r}'|\hat{V}|\mathbf{r}'\rangle \ , \text{ where} \\
\langle \mathbf{r}|\hat{\varepsilon}(\omega)|\mathbf{r}'\rangle &= \lim_{\eta\to 0+} \langle \mathbf{r}|\hat{\varepsilon}(\omega+i\eta)|\mathbf{r}'\rangle = \lim_{\eta\to 0+} \int_0^\infty \mathrm{d}\tau \ e^{i(\omega+i\eta)\tau} \ \langle \mathbf{r}|\varepsilon(\tau)|\mathbf{r}'\rangle \\
&= \langle \mathbf{r}|\mathbf{r}'\rangle - \lim_{\eta\to 0+} \sum_{i,j} \langle i|\hat{G}|j\rangle \int \mathrm{d}^3 r'' \ \frac{e^2}{\|\mathbf{r}-\mathbf{r}''\|} \ \langle j|\mathbf{r}''\rangle\langle \mathbf{r}''|i\rangle\langle i|\mathbf{r}'\rangle\langle \mathbf{r}'|j\rangle \\
&= \langle \mathbf{r}|\mathbf{r}'\rangle - \int \mathrm{d}^3 r'' \ \langle \mathbf{r}|\hat{V}_{\text{Coulomb}}|\mathbf{r}''\rangle\langle \mathbf{r}''|\hat{\chi}(\omega)|\mathbf{r}'\rangle \ , \\
\langle \mathbf{r}''|\hat{\chi}(\omega)|\mathbf{r}'\rangle &= \lim_{\eta\to 0+} \sum_{i,j} \langle i|\hat{G}|j\rangle\langle j|\mathbf{r}''\rangle\langle \mathbf{r}''|i\rangle\langle i|\mathbf{r}'\rangle\langle \mathbf{r}'|j\rangle \ , \\
\langle \mathbf{r}|\hat{V}_{\text{Coulomb}}|\mathbf{r}''\rangle &= \frac{e^2}{\|\mathbf{r}-\mathbf{r}''\|} \ ,
\end{aligned} \tag{10}$$

which is the exact equivalent of eq. (4) in the classical description. $\hat{\chi}$ is called the *polarizability matrix*.

[3] At point $\mathbf{r}$ we have

$$\begin{aligned}
\langle \mathbf{r}|\hat{V}_{\text{ext}}(t)|\mathbf{r}\rangle &= \langle \mathbf{r}|\hat{V}_{\text{tot}}(t)|\mathbf{r}\rangle - \int \mathrm{d}^3 r' \ \frac{e^2}{\|\mathbf{r}-\mathbf{r}'\|} \ \langle \mathbf{r}'|\delta\hat{N}(t)|\mathbf{r}'\rangle \\
&\overset{(8)}{=} \langle \mathbf{r}|\hat{V}|\mathbf{r}\rangle e^{-i(\omega+i\eta)t} - \int \mathrm{d}^3 r' \ \frac{e^2}{\|\mathbf{r}-\mathbf{r}'\|} \sum_{i,j} \langle i|\hat{G}|j\rangle \ e^{-i(\omega+i\eta)t} \ \langle j|\mathbf{r}'\rangle\langle \mathbf{r}'|i\rangle\langle i|\hat{V}|j\rangle \\
&= \left(\langle \mathbf{r}|\hat{V}|\mathbf{r}\rangle - \sum_{i,j} \langle i|\hat{G}|j\rangle \int \mathrm{d}^3 r' \int \mathrm{d}^3 r'' \int \mathrm{d}^3 r''' \ \frac{e^2}{\|\mathbf{r}-\mathbf{r}'\|} \ \langle j|\mathbf{r}'\rangle\langle \mathbf{r}'|i\rangle\langle i|\mathbf{r}''\rangle\langle \mathbf{r}'''|j\rangle\langle \mathbf{r}''|\hat{V}|\mathbf{r}'''\rangle \right) e^{-i(\omega+i\eta)t} \\
&= \left(\langle \mathbf{r}|\hat{V}|\mathbf{r}\rangle - \sum_{i,j} \langle i|\hat{G}|j\rangle \int \mathrm{d}^3 r' \int \mathrm{d}^3 r'' \ \frac{e^2}{\|\mathbf{r}-\mathbf{r}'\|} \ \langle j|\mathbf{r}'\rangle\langle \mathbf{r}'|i\rangle\langle i|\mathbf{r}''\rangle\langle \mathbf{r}''|j\rangle\langle \mathbf{r}''|\hat{V}|\mathbf{r}''\rangle \right) e^{-i(\omega+i\eta)t} \ .
\end{aligned}$$

2 Application

We now apply the results of sec. 1 to a finite 2D lattice described in the tight binding approximation. We do the calculation in *atomic basis*, i.e. the basis of local site wave functions $|a\rangle$ ($a \in \{0, N-1\}$, where N is the number of sites). The following assumptions are made:

- $\langle a|b\rangle = \delta_{a,b}$,
- atomic basis is complete, i.e. $\sum_a |a\rangle\langle a| = \hat{\mathbb{1}}$,
- $|a\rangle$'s are localized around the corresponding sites, i.e. $\langle \mathbf{r}|a\rangle \approx \delta(\mathbf{r} - \mathbf{r_a})$, where $\mathbf{r_a}$ is the position of a'th site. This is the key to making a step from analytical formulas to numerical calculations.

These assumptions essentially mean that the step from position representation to atomic basis is performed by $|\mathbf{r}\rangle \to |a\rangle$, $\mathbf{r} \to \mathbf{r_a}$ and $\int \mathrm{d}^3 r \to \sum_a$. Eq. (10) now reads[4]

$$\begin{aligned}
\langle a|\hat{V}_{\text{ext}}(\omega)|a\rangle &= \sum_b \langle a|\hat{\varepsilon}(\omega)|b\rangle\langle b|\hat{V}|b\rangle \ , \text{ where} \\
\langle a|\hat{\varepsilon}(\omega)|b\rangle &= \langle a|b\rangle - \sum_c \langle a|\hat{V}_{\text{Coulomb}}|c\rangle\langle c|\hat{\chi}(\omega)|b\rangle \ , \\
\langle a|\hat{\chi}(\omega)|b\rangle &= \lim_{\eta\to 0+} \sum_{i,j} \langle i|\hat{G}|j\rangle\langle j|a\rangle\langle a|i\rangle\langle i|b\rangle\langle b|j\rangle \ , \\
\langle a|\hat{V}_{\text{Coulomb}}|b\rangle &= \begin{cases} \dfrac{1}{4\pi\epsilon_0}\dfrac{e}{\|\mathbf{r_a} - \mathbf{r_b}\|} & \text{, if } a \neq b, \\ V_0 & \text{, if } a = b, \end{cases}
\end{aligned} \tag{11}$$

The trick that allows the calculations of $\hat{\chi}(\omega)$ in a reasonable time is to rewrite it in terms of matrices:

$$\begin{aligned}
\langle a|\hat{\chi}(\omega)|b\rangle &= \overbrace{A(a,b)}^{\text{row vector}} \underbrace{\hat{G}}_{\text{square matrix}} \overbrace{A(a,b)^\dagger}^{\text{column vector}} = \overbrace{\Big(\underbrace{\hat{G}^{\mathrm{T}} A(a,b)^{\mathrm{T}}}_{\texttt{GEMV}}\Big)^{\mathrm{T}} A(a,b)^\dagger}^{\texttt{DOT}} \ , \text{ where} \\
G_{i,j} &= \langle i|\hat{G}|j\rangle \overset{(7)}{=} \frac{n_i - n_j}{E_i - E_j - \hbar(\omega + i\eta)} \quad \text{with } \eta \text{ small, and} \\
A(a,b)_i &= \langle a|i\rangle\langle i|b\rangle = \langle a|i\rangle\langle b|i\rangle^* \ .
\end{aligned} \tag{12}$$

The second form of $\langle a|\hat{\chi}(\omega)|b\rangle$ with a lot of transposes may seem strange, but it is of utmost importance. It allows us to calculate matrix elements of $\hat{\chi}(\omega)$ using just two `BLAS` operations: matrix-vector product (`GEMV`) and dot-product (`DOT`). It is now straightforward to write a highly parallel implementation of eq. (12) and it will not be discussed here any further.

With eqs. (11) and (12) implemented, we can obtain $\hat{\varepsilon}(\omega)$ for any system, given its tight-binding Hamiltonian $\hat{H}$ and sites positions $\big\{\mathbf{r_a}|\, a \in \{0, \ldots, N-1\}\big\}$. The measurable quantity (**TODO:** give links to articles about EELS experiments) is the *loss function* $-\mathrm{Im}\left[\langle \mathbf{q}|\hat{\varepsilon}^{-1}(\omega)|\mathbf{q}\rangle\right]$, where $|\mathbf{q}\rangle$'s denote momentum eigenfunctions. Let $\epsilon_n(\omega)$'s be eigevalues of

[4] Eq. (10) was written in Gauss system. For the calculations it is, however, easier to use electron-volts. We thus replace e^2 by $\frac{e}{4\pi\epsilon_0}$. We also introduce the *self-interaction potential* V_0 to prevent degeneracies in $\langle a|\hat{V}_{\text{Coulomb}}|a\rangle$.

$\hat{\varepsilon}(\omega)$ with the corresponding eigenfunctions $|\phi_n(\omega)\rangle$'s. We can now express the loss function as[5]

$$-\operatorname{Im}\left[\langle \mathbf{q}|\hat{\varepsilon}^{-1}(\omega)|\mathbf{q}\rangle\right] = -\sum_n |\langle \mathbf{q}|\phi_n(\omega)\rangle|^2 \cdot \operatorname{Im}\left[\frac{1}{\epsilon_n(\omega)}\right] .$$

Within the tight-binding approximation $\langle \mathbf{q}|\phi_n(\omega)\rangle$ can be calculated as follows

$$\langle \mathbf{q}|\phi_n(\omega)\rangle = \sum_a \int \mathrm{d}^3 r \ \langle \mathbf{q}|\mathbf{r}\rangle\langle \mathbf{r}|a\rangle\langle a|\phi_n(\omega)\rangle = \sum_a \langle \mathbf{q}|\mathbf{r_a}\rangle\langle a|\phi_n(\omega)\rangle = \frac{1}{(2\pi)^{3/2}} \sum_a e^{-i\mathbf{q}\cdot\mathbf{r_a}}\langle a|\phi_n(\omega)\rangle .$$

[5]Obviously, if $\epsilon_n(\omega)$ is an eigenvalue of $\hat{\varepsilon}(\omega)$ with an eigenvector $|\phi_n(\omega)\rangle$, then $1/\epsilon_n(\omega)$ is an eigenvalue of $\hat{\varepsilon}^{-1}(\omega)$ with the same eigenvector. We thus have

$$\begin{aligned} -\operatorname{Im}\left[\langle \mathbf{q}|\hat{\varepsilon}^{-1}(\omega)|\mathbf{q}\rangle\right] &= -\operatorname{Im}\left[\sum_{n,m}\langle \mathbf{q}|\phi_n\rangle\langle\phi_n|\hat{\varepsilon}^{-1}(\omega)|\phi_m\rangle\langle\phi_m|\mathbf{q}\rangle\right] \\ &= -\operatorname{Im}\left[\sum_n |\langle \mathbf{q}|\phi_n(\omega)\rangle|^2 \frac{1}{\epsilon_n(\omega)}\right] = -\sum_n |\langle \mathbf{q}|\phi_n(\omega)\rangle|^2 \cdot \operatorname{Im}\left[\frac{1}{\epsilon_n(\omega)}\right] . \end{aligned}$$

3 Experiments

Let's now apply the aquired tools to examine plasmonic properties of some systems. For this we need a way of visualising the results. We will follow the approach taken in [Wang Weihua et al., 2015].

Classically, plasmons are identified by $\varepsilon(\omega) = 0$. In reality, there is also loss due to the parameter $\eta \neq 0$ in eq. (11). We therefore seek frequencies ω such that

$$\hat{\varepsilon}(\omega)|\phi_n(\omega)\rangle = \epsilon_n(\omega)|\phi_n(\omega)\rangle \ , \text{ with } \epsilon_n(\omega) \in \mathbb{C} \setminus \mathbb{R}, \text{ i.e. purely imaginary.} \tag{13}$$

As noted in [Andersen et al., 2012], "when the imaginary part of eigenvalue $\epsilon_n(\omega)$ does not vary too much around the plasmon frequency ω'", condition (13) is equivalent to the condition that

$$-\operatorname{Im}[\epsilon_n^{-1}(\omega)] \text{ has a local maximum at } \omega' \ .$$

This implies that $\epsilon_n(\omega)$ should be a continuous in the neighbourhood of ω'. The problem is to choose n. In which of all the eigenvalues are we interested in? And does n vary with ω or remain constant? The approach taken in [Wang Weihua et al., 2015] is to let n be a function of ω defined as the index of the eigenvalue with the highest $-\operatorname{Im}[\epsilon_j^{-1}(\omega)]$. We then seek the maxima of $-\operatorname{Im}[\epsilon_{n(\omega)}^{-1}(\omega)]$ and call them *plasmon frequencies*. Although it is not obvious whether such an approach is mathematically valid, the fact that $-\operatorname{Im}[\epsilon_{n(\omega)}^{-1}(\omega)]$ turns out to be continuous in the calculations seems like a good physical justification.

Finally, for each (ω, n) pair, $\operatorname{Re}[\langle \mathbf{r}|\phi_{n(\omega)}(\omega)\rangle]$ gives a good insight in how the plasmon modes "look like". We now discuss a couple of systems for which we've performed the outlined calculations.

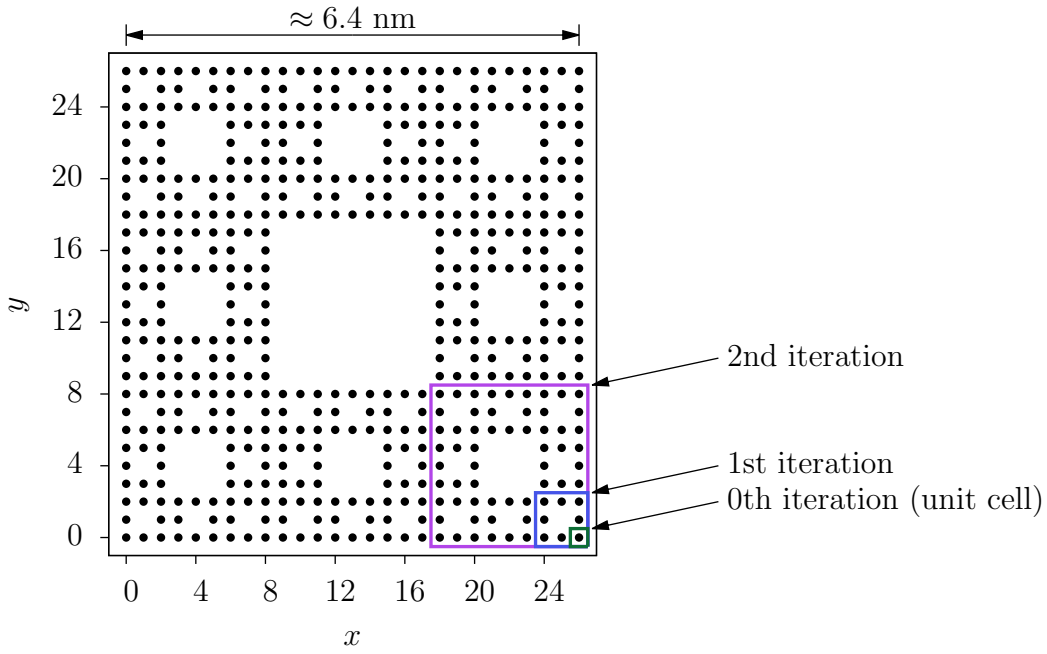

Figure 1: Third iteration Sierpinski carpet. x and y coordinates are given in terms of lattice constant. Width of the sample is $3^3 = 27$ unit cells. In this case we chose the lattice constant of graphene $a \approx 0.246$ nm, which results in the total width of about six and a half nanometers.

3.1 Third iteration Sierpinski carpet

We start by examining a small system — third iteration Siepinski carpet. The sample is shown in fig. 1. Using TIPSI Python package we can construct a tight-binding Hamiltonian for the

problem. Hopping value of 2.8 eV is used. As we need eigenenergies and eigenstates anyway to make use of eq. (11), we can have a look at the density of states (fig. 2). Although the

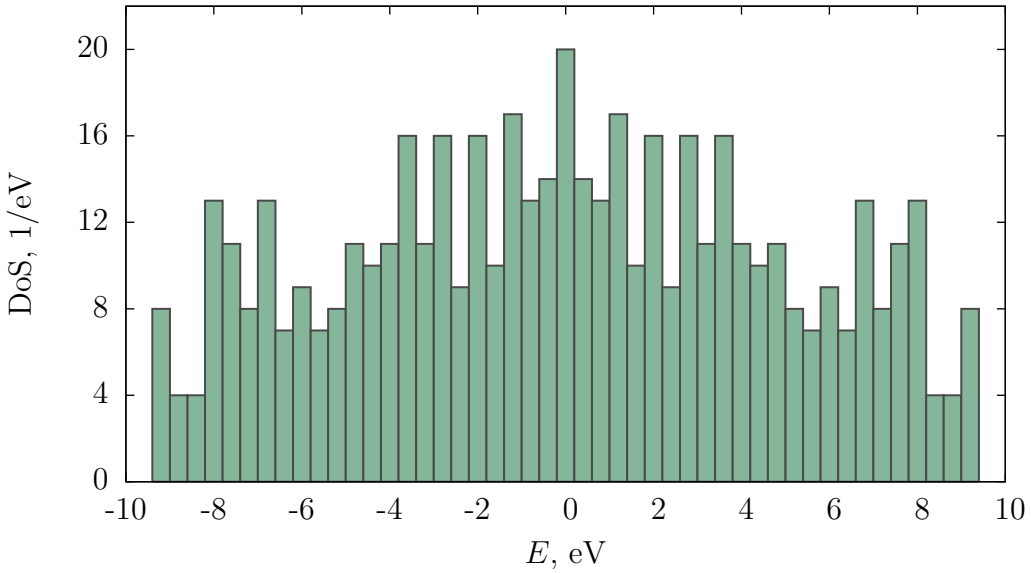

Figure 2: Density of states for the 3rd iteration SC. Due to the small size of the system, we can't say much about the distribution of energy eigenvalues. Except that it's highly fluctuating — as one would expect for a fractal system.

number of points is quite small (512 atomic sites), we can still extract some information. For example, all possible energies lie within the $(-9.5\text{ eV}, 9.5\text{ eV})$ range. Hence, $E_i - E_j < 19$ eV for any i, j. $G_{i,j}$ has $(E_i - E_j - \hbar(\omega + i\eta))$ in the denominator, thus for ω's greater than ≈ 20 eV, $-\operatorname{Im}[\epsilon^{-1}_{n(\omega)}(\omega)]$ will asymptotically approach zero — no need to search for plasmons there.

Calculating the loss function $-\operatorname{Im}[\epsilon^{-1}_{n(\omega)}(\omega)]$ for frequencies up to ≈ 22 eV results in the spectrum shown on fig. 3. High number of sharp peaks suggests either that there are plenty of plasmons in the sample or that our definition of the loss function is ill formed. A higher resolution plot shows that the loss function is smooth around its maxima. Its derivative experiences a discontinuity at *almost every* minima, which suggests that our definition it not perfect after all. We will discuss alternatives later.

References

[Andersen et al., 2012] Andersen, K., Jacobsen, K. W., and Thygesen, K. S. (2012). Spatially resolved quantum plasmon modes in metallic nano-films from first-principles. *Physical Review B*, 86(24):245129.

[Wang Weihua et al., 2015] Wang Weihua, Christensen Thomas, Jauho Antti-Pekka, Thygesen Kristian S., Wubs Martijn, and Mortensen N. Asger (2015). Plasmonic eigenmodes in individual and bow-tie graphene nanotriangles. *Scientific Reports*, 5:9535.

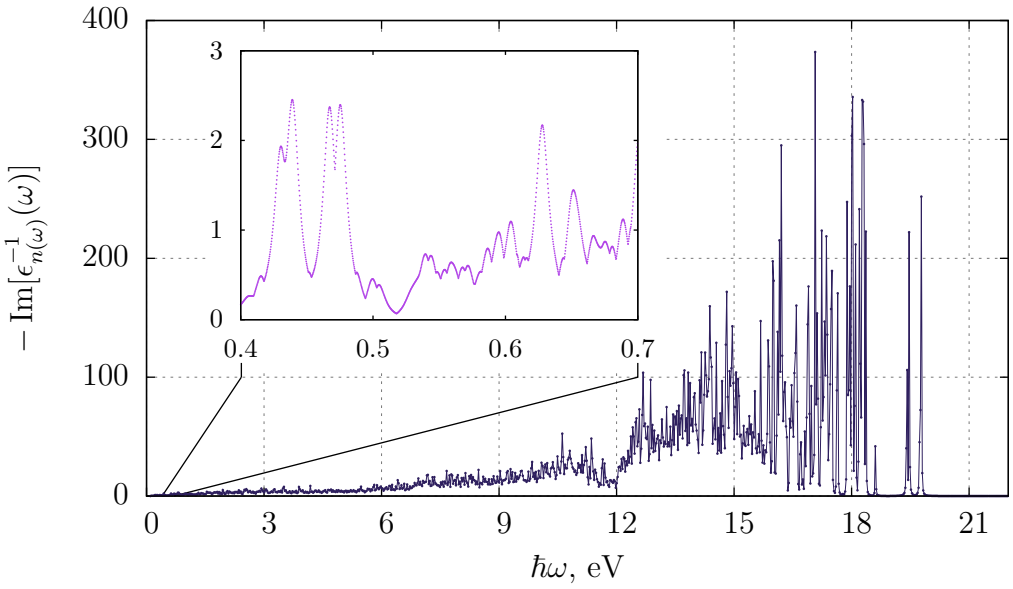

Figure 3: Loss function spectrum. The spectrum is highly fluctuating (but not self-similar). This suggests that there are many plasmons.