Plasmonic Properties of QM 2D Fractals.

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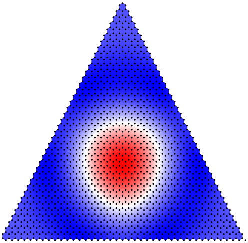
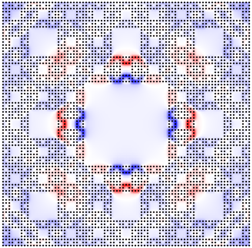
TCM





Stained Glass

Plasmons

Definition (Plasmon)

Quantum of plasma oscillation (*think quasiparticle*).

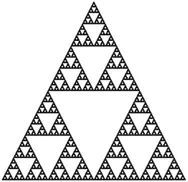
Definition (Plasma Oscillation)

Longitudinal waves of electron charge-density.

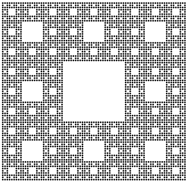
Fractals

- Features: nowhere differentiable, have fractal dimensions, self-similar, etc.
- Ramification: minimum number of links that one needs to remove to separate a macroscopic part of infinite fractal.

Finite ramification: Solved analytically by L.P. Kadanoff (1982).



Infinite ramification: Unsolvable analytically (?). Numerical calculations exist (*done at Radboud*), but not of plasmonic properties.



Tight Binding

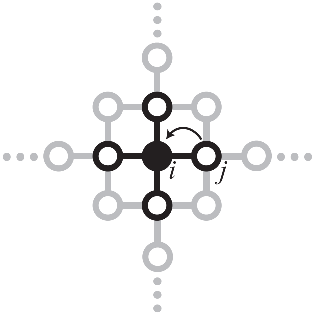
- atomic basis $\{|a\rangle\}$
- Hamiltonian of the system is

$$\hat{H} = -t \sum_{\langle a,b \rangle} (\hat{c}_a^\dagger \hat{c}_b + \text{h.c.}) \; ,$$

$\hat{c}_a^\dagger$ and $\hat{c}_a$ are creation and annihilation operators, t is the hopping value.

- Hopping:

$$\underbrace{\hat{c}_a^\dagger \underbrace{\hat{c}_b |\psi\rangle}_{\text{“take” from } b}}_{\text{“move” to } a}$$



System

- Electron system is described (in one-particle approximation) by
  - eigenenergies $\{E_i\}$ and
  - eigenstates $\{|i\rangle\}$.
- Grand-canonical ensemble, i.e. temperature T and chemical potential μ are fixed.
- Occupational numbers are given by the Fermi-Dirac distribution.

Problem

- All plasmonic properties are obtained from the dielectric function $\hat{\varepsilon}$.
  $\Longrightarrow$ Goal: calculate $\hat{\varepsilon}$.
- There exist good methods for calculation of the dielectric function in momentum space, i.e. they rely on translational symmetry of the system.
- But in fractals, there is no translational symmetry!
- Calculations have to be done in real space.
  $\Longrightarrow$ New method.

Dielectric Function in Real Space

Within the Random Phase Approximation (RPA) we obtain:

$$\langle \mathbf{r}|\hat{V}_{\text{ext}}(\omega)|\mathbf{r}\rangle = \int \mathrm{d}^3 r' \, \langle \mathbf{r}|\hat{\varepsilon}(\omega)|\mathbf{r}'\rangle\langle \mathbf{r}'|\hat{V}|\mathbf{r}'\rangle \ , \ \text{where}$$

$$\langle \mathbf{r}|\hat{\varepsilon}(\omega)|\mathbf{r}'\rangle = \langle \mathbf{r}|\mathbf{r}'\rangle - \lim_{\eta\to 0+} \sum_{i,j} \frac{n_i - n_j}{E_i - E_j - \hbar(\omega + i\eta)} \int \mathrm{d}^3 r'' \frac{e^2}{\|\mathbf{r} - \mathbf{r}''\|} \times \langle j|\mathbf{r}''\rangle\langle \mathbf{r}''|i\rangle\langle i|\mathbf{r}'\rangle\langle \mathbf{r}'|j\rangle \ .$$

Algorithm

Calculating $\hat{\varepsilon}$ for a single ω is an $\mathcal{O}(N^4)$ problem, where N is the number of particles.

- Hack: rewrite everything in terms of matrix operations and use BLAS (*Basic Linear Algebra Subroutines*).
- Use a supercomputer for calculations.
- Hack: use symmetries of the system.

Code

- Written C++ package to calculate $\hat{\varepsilon}$ for a general system described by TB.
- $\approx$ 8.5 thousands lines of code (*excluding* data analysis scripts).
- ≈ 400000 computational hours spent, that is $\approx$ 45 years of calculations.

Electron Energy Loss Spectroscopy (EELS)

- Shoot electrons on the sample.
  - Electron beam with a well-defined wavevector $\mathbf{k}$.
  - Inelastic scattering results in energy loss $E = \hbar\omega$ and momentum transfer $\hbar\mathbf{q}$.
- Look at (usually) transmitted electrons.

$$\overbrace{\frac{d^2\sigma}{d\Omega dE}}^{\text{double-differential cross section}} \propto \underbrace{-\operatorname{Im}\left[\frac{1}{\langle \mathbf{q}|\hat{\varepsilon}(\omega)|\mathbf{q}\rangle}\right]}_{\text{loss function}} .$$

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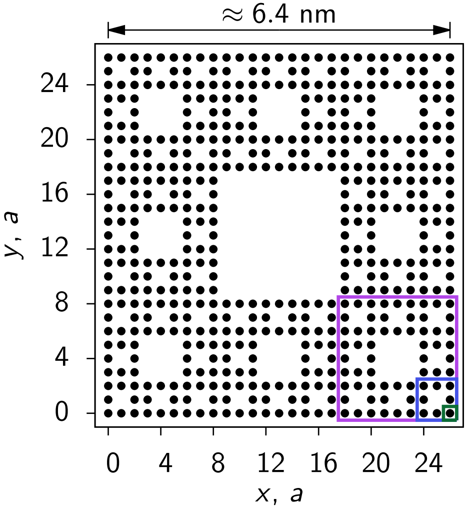
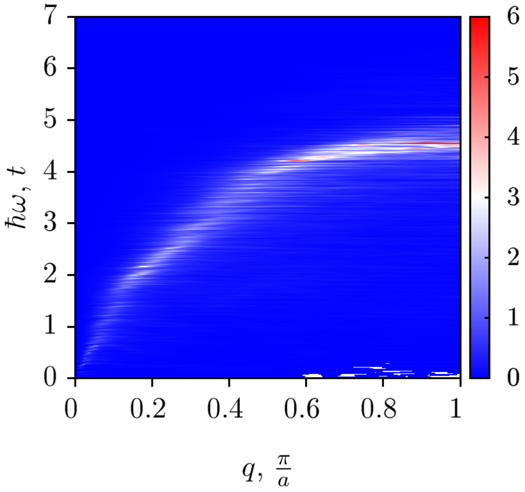
Third Iteration Sierpinski Carpet

System



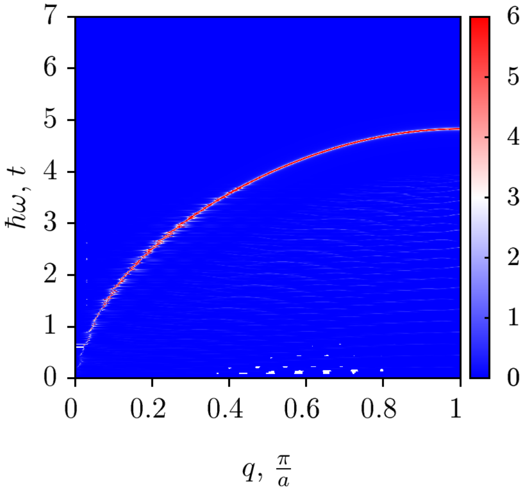
Third Iteration Sierpinski Carpet

Results



Square Lattice

Results



Summary

- It *is* possible to do calculations of $\hat{\varepsilon}$ in real space.
- There *are* well defined plasmons in Sierpinski carpets, which is *unexpected* for a system without translational invariance.
- The created package can be *reused* to study plasmonic properties of other systems.

Outlook

- Publish the results, i.e. write a paper.
- Look at *other* systems with no translational symmetry.

Any questions?

Simple example of plasma oscillation

- Free electron gas with positively charged atom cores in the background.
- Oscillating electric field $\mathbf{E}(t) = \mathbf{E_0} \exp(-i\omega t)$.
- Equation of motion:

$$\ddot{\mathbf{x}} + \gamma\dot{\mathbf{x}} = -\frac{e}{m}\mathbf{E}(t) \ .$$

- Displaced electrons generate a polarization $\mathbf{P} = -Ne\mathbf{x}$ (N is the electron density).
- From $\epsilon_0\mathbf{E} + \mathbf{P} = \epsilon_0\varepsilon(\omega)\mathbf{E}$ we get $\varepsilon(\omega)$.
- In high frequency limit $\varepsilon(\omega) \approx 1 - \frac{\omega_p^2}{\omega^2}$, ω_p — plasma frequency.
- If $\omega = \omega_p$, we get longitudinal oscillation of electron-charge density.

Random Phase Approximation

- Perturbation of the form

$$\hat{V} e^{-i(\omega + i\eta)t}\,, \;\text{with}\; \eta \to +0\,.$$

($\hat{V}$ is diagonal in $\mathbf{r}$-basis, i.e. $\langle \mathbf{r} | \hat{V} | \mathbf{r}' \rangle = V(\mathbf{r}) \delta^3(\mathbf{r} - \mathbf{r}')$)

- Results in an induced charge density variation $\delta \hat{N}(t)$
- Self-consistency equation

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