Plasmonic Properties of QM 2D Fractals.

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TCM

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Stained Glass

Plasmons

Definition (Plasmon)

Quanta of plasma oscillation (*think quasiparticle*).

Definition (Plasma Oscillation)

Longitudinal waves of electron charge-density.

Simple example

- 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.

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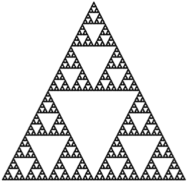
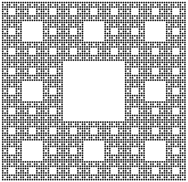
Fractals

- 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.



Problem

- Plasmonic properties are obtained from ε
  $\Longrightarrow$ Goal: calculate ε.
- There exist good methods to calculate the dielectric function in momentum space, i.e. rely on translational symmetry.
- In fractals, there is no translational symmetry!
- Calculations have to be done in real space.
  $\Longrightarrow$ New method.

System

- Electron system is described (in a 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

$$n_i = \frac{1}{\exp\left(\frac{E_i-\mu}{k_\mathrm{B}T}\right)+1} \, .$$

- Introduce a one-particle density matrix

$$\hat{\rho}_0 = \sum_i n_i \, |i\rangle\langle i| \, .$$

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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

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

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. . . a good excercise in calculus and Fourier analysis . . .

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

$$\langle \mathbf{r}|\hat{\varepsilon}(\omega)|\mathbf{r}'\rangle = \lim_{\eta\to 0+}\langle \mathbf{r}|\hat{\varepsilon}(\omega+i\eta)|\mathbf{r}'\rangle = \lim_{\eta\to 0+}\int_0^\infty \mathrm{d}\tau \; e^{i(\omega+i\eta)\tau}\,\langle \mathbf{r}|\hat{\varepsilon}(\tau)|\mathbf{r}'\rangle$$

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

$$\times \langle j|\mathbf{r}''\rangle\langle \mathbf{r}''|i\rangle\langle i|\mathbf{r}'\rangle\langle \mathbf{r}'|j\rangle$$

Tight Binding

- Electrons are "tightly bound" to atoms
  $\Longrightarrow$ atomic basis $\{|a\rangle\}$
  - $\langle a|b\rangle = \delta_{a,b}$ and $\sum_a |a\rangle\langle a| = \hat{\mathbb{1}}$,
  - $\langle \mathbf{r}|a\rangle \approx 1$.
- Example Hamiltonian of the system is

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

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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.

Parallelisation

- Use different cluster nodes (*think small supercomputers*) to calculate $\hat{\varepsilon}$ for different frequencies
  $\implies$ `MPI`, `Boost`
- Use different threads to speed up matrix operations
  $\implies$ `Intel MKL`, `OpenMP`

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.

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

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Third Iteration Sierpinski Carpet

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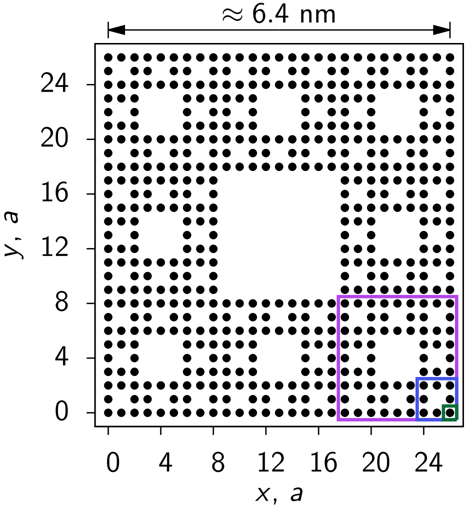
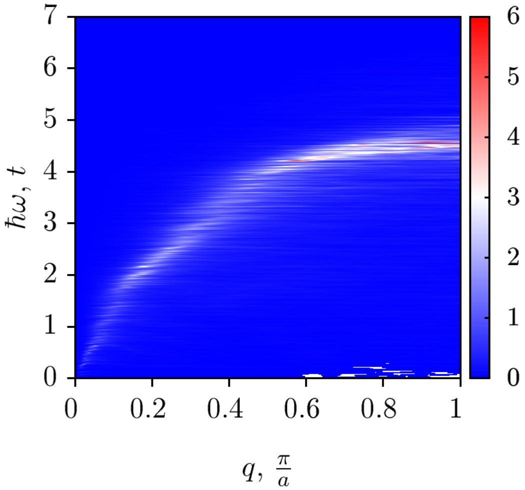


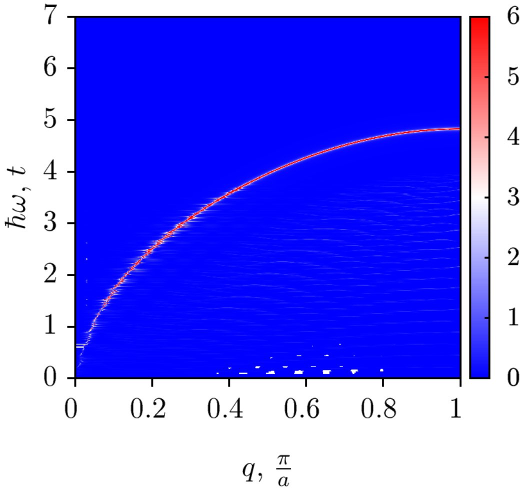
Third Iteration Sierpinski Carpet

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.

Any questions?