

Microscopic-macroscopic connection

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Outline

1. Introduction: which quantities do we need
 2. Macroscopic average
 - Definition
 - Examples
 - Dielectric tensor for cubic symmetries
 - Dielectric tensor for non-cubic symmetries
 - Properties
 - Principal axis
 - Summary
-

Linear response

Perturbation theory

- For a sufficiently small perturbation, the response of the system can be extended into a Taylor series, with respect to the perturbation.
- We will consider only the first order (linear) response, proportional to the perturbation.
- The linear coefficient linking the response to the perturbation is called a response function. It is independent of the perturbation and depends only on the system.
- ≠ Strong field interaction (laser field for instance)

Examples

Density response function

$$n_1(\vec{r}, t) = \int dt' \int d\vec{r}' \chi(\vec{r}, t, \vec{r}', t') V(\vec{r}', t')$$

Dielectric tensor

$$\vec{D}(\vec{r}, t) = \int dt' \int d\vec{r}' \vec{\epsilon}(\vec{r}, t, \vec{r}', t') \vec{E}(\vec{r}', t')$$

Which quantities do we need?

Absorption coefficient

The general solution of the Maxwell's equations for the electric field is

$$\vec{E}(\vec{r}, t) = \vec{E}_0 e^{i(kx - \omega t)}$$

$$k = \frac{\omega}{c} \sqrt{\epsilon}$$

Defining the complex refractive index as $n = \sqrt{\epsilon} = \nu + i\kappa$, the electric field inside a medium is the damped wave:

$$\vec{E}(\vec{r}, t) = \vec{E}_0 e^{i\frac{\omega}{c}x n} e^{-i\omega t} = \vec{E}_0 e^{i\frac{\omega}{c}\nu x} e^{-\frac{\omega}{c}\kappa x} e^{-i\omega t}$$

ν and κ are the **refraction index** and the **extinction coefficient** and they are related to the dielectric constant ($\epsilon = \epsilon_1 + i\epsilon_2$) as

$$\epsilon_1 = \nu^2 - \kappa^2 \quad \epsilon_2 = 2\nu\kappa$$

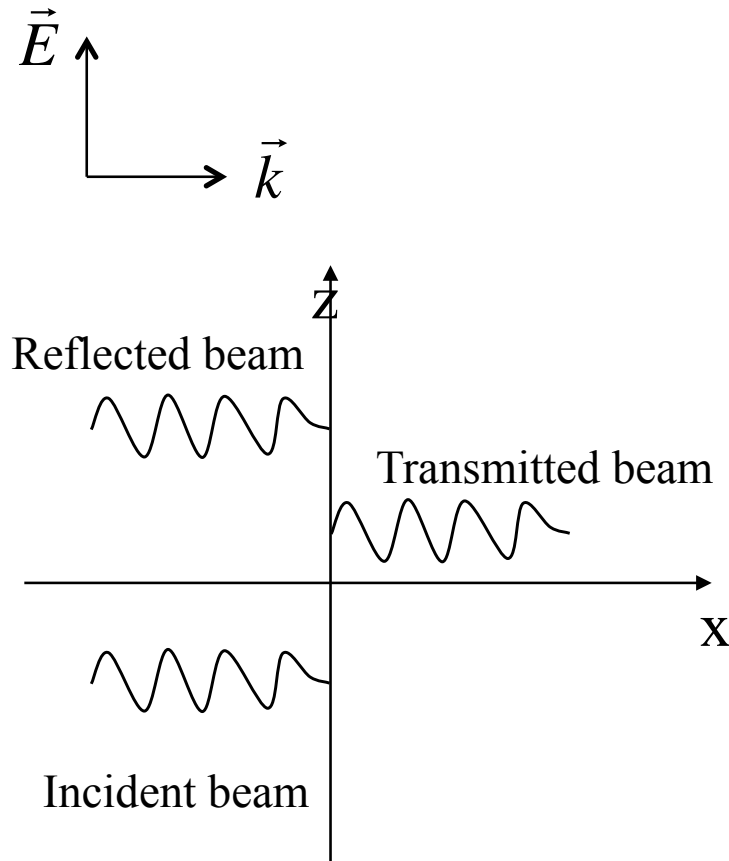
The absorption coefficient α is the inverse distance where the intensity of the field is reduced by 1/e

$$\delta = \frac{c}{\omega\kappa}$$

$$\alpha = \frac{2\omega\kappa}{c} = \frac{\omega\epsilon_2}{\nu c}$$

(related to the optical skin depth δ).

Which quantities do we need?



Reflectivity

Normal incidence reflectivity

$$R = \left| \frac{E_T}{E_i} \right|^2 < 1$$

Using the continuity of the tangential component of the electric field at the surface

$$R = \frac{(1 - \nu)^2 + \kappa^2}{(1 + \nu)^2 + \kappa^2}$$

The knowledge of the optical constant implies the knowledge of the absorption and of the reflectivity, which can be compared with the experiment.

Which quantities do we need?

Energy loss by a fast particle

Given an external charge density ρ_{ext} , one can obtain the external potential V_{ext}

$$k^2 V_{\text{ext}}(\vec{k}, \omega) = 4\pi \rho_{\text{ext}}(\vec{k}, \omega) \quad (\text{Poisson equation})$$

The response of the system is an induced density, defined by the response function χ

$$\rho_{\text{ind}}(\vec{k}, \omega) = \chi(\vec{k}, \omega) V_{\text{ext}}(\vec{k}, \omega)$$

and the total (induced + external) potential acting on the system is

$$V_{\text{tot}}(\vec{k}, \omega) = \left[1 + \frac{4\pi}{k^2} \chi(\vec{k}, \omega) \right] V_{\text{ext}}(\vec{k}, \omega) = \varepsilon^{-1}(\vec{k}, \omega) V_{\text{ext}}(\vec{k}, \omega)$$

Dielectric function:

$$V_{\text{ext}}(\vec{k}, \omega) = \varepsilon(\vec{k}, \omega) V_{\text{tot}}(\vec{k}, \omega)$$

Which quantities do we need?

Energy loss by a fast particle

Charge density of a particle (e^-) with velocity v : $\rho_{ext} = e\delta(\vec{r} - \vec{v}t)$

The total electric field is $\vec{E}_{tot} = -\vec{\nabla}_{\vec{r}} V_{tot}(\vec{r}, t)$
and the energy lost by the electron in unit time is

$$\frac{dW}{dt} = \int d\vec{r} \vec{j} \cdot \vec{E}_{tot}$$

with the current density

$$\vec{j} = e\vec{v}\delta(\vec{r} - \vec{v}t)$$

We get

$$\frac{dW}{dt} = -\frac{e^2}{\pi^2} \int \frac{d\vec{r}}{k^2} \text{Im} \left\{ \frac{\omega}{\epsilon(\vec{k}, \omega)} \right\}$$

Electron Energy Loss Spectroscopy:

$$-\text{Im} \left\{ \frac{1}{\epsilon(\vec{k}, \omega)} \right\}$$

is called the loss function

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Maxwell's equations in vacuum

$$\vec{\nabla} \cdot \vec{E}_{ext}(\vec{r}, t) = 4\pi\rho_{ext}(\vec{r}, t)$$

$$\vec{\nabla} \cdot \vec{B}_{ext}(\vec{r}, t) = 0$$

$$\vec{\nabla} \times \vec{E}_{ext}(\vec{r}, t) = -\frac{1}{c} \frac{\partial \vec{B}_{ext}}{\partial t}$$

$$\vec{\nabla} \times \vec{B}_{ext}(\vec{r}, t) = \frac{4\pi}{c} \vec{j}_{ext}(\vec{r}, t) + \frac{1}{c} \frac{\partial \vec{E}_{ext}}{\partial t}$$

E: electric field

B: magnetic field
(induction)

ρ_{ext} and \vec{j}_{ext} are the (external) free charge and current density

They can be arbitrary, but they have to fulfill the continuity equation

$$\vec{\nabla} \cdot \vec{j}_{ext}(\vec{r}, t) + \frac{\partial \rho_{ext}}{\partial t} = 0$$

Maxwell's equations in a medium

$$\begin{aligned}\vec{\nabla} \cdot \vec{E}_{tot}(\vec{r}, t) &= 4\pi\rho_{tot}(\vec{r}, t) \quad \text{or} \quad \vec{\nabla} \cdot \vec{D}(\vec{r}, t) = 4\pi\rho_{ext}(\vec{r}, t) \\ \vec{\nabla} \cdot \vec{B}_{tot}(\vec{r}, t) &= 0 \\ \vec{\nabla} \times \vec{E}_{tot}(\vec{r}, t) &= -\frac{1}{c} \frac{\partial \vec{B}_{tot}}{\partial t} \\ \vec{\nabla} \times \vec{B}_{tot}(\vec{r}, t) &= \frac{4\pi}{c} \vec{j}_{tot}(\vec{r}, t) + \frac{1}{c} \frac{\partial \vec{E}_{tot}}{\partial t}\end{aligned}$$

It is often more convenient to use

$$\vec{D} = \vec{E}_{tot} + 4\pi\vec{P}$$

instead of E_{tot} , as D can be very close to the external field

$$\vec{\nabla} \cdot \vec{D} = \vec{\nabla} \cdot \vec{E}_{ext}$$

(see the previous slide)

With $\rho_{tot} = \rho_{ext} + \rho_{ind}$ and $\vec{j}_{tot} = \vec{j}_{ext} + \vec{j}_{ind}$,

ρ_{ind} and \vec{j}_{ind} are the **induced charge** and **current density**:

they are not arbitrary, but reflect the spatial structure of the medium on a microscopic level and the motion of the particle in it, including also the response to an external field.

Microscopic spatial fluctuations



- Infinite crystals → microscopic inhomogeneities (atomic scale)
- Semi-infinite crystals → presence of the surface
- Desordered medium → liquid
- Rough surfaces

Macroscopic average

We consider spatial fluctuations whose characteristic length scale is much smaller than the wavelength of light

Macroscopic quantities

Quantities that are slowly varying over the unit cells.
where V is the volume per unit cell of the crystal.

$$\lambda \gg V^{1/3}$$

Examples

$$E_{\text{ext}}(r, t), A_{\text{ext}}(r, t), V_{\text{ext}}(r, t), \dots$$

Typical values:

- dimension of the unit cell for silicon $a_{\text{cell}} \approx 0.357 \text{ nm}$
- Visible radiation $400 \text{ nm} < \lambda < 800 \text{ nm}$

Macroscopic average



Microscopic quantities

Total and induced fields are rapidly varying. They include the contribution from electrons in all regions of the cell.

The contribution of electrons close to or far from the nuclei will be very different.

⇒ Large and irregular fluctuations over the atomic scale.



Examples

$$E_{\text{tot}}(\mathbf{r}, t), j_{\text{ind}}(\mathbf{r}, t), \rho_{\text{ind}}(\mathbf{r}, t), \dots$$

Macroscopic average

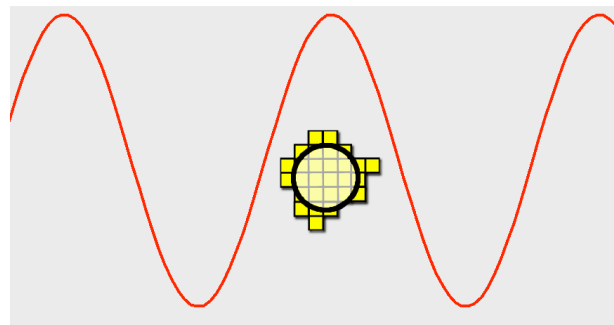
Measurable quantities

One measures quantities that vary on a macroscopic scale.

In the **long wavelength** limit,
the macroscopic neighbourhood contains many particles

We have to average over distances :

- ✓ large compared to the cell diameter
- ✓ small compared to the wavelength of the external perturbation



Macroscopic average

General definition:

We have to define two operators \hat{P}_a and \hat{P}_f which extract the average component and the fluctuation component of any function F

$$\hat{P}_f = \hat{1} - \hat{P}_a \quad \begin{array}{l} F_a = \hat{P}_a F \\ F_f = \hat{P}_f F \end{array}$$

\hat{P}_a and \hat{P}_f have the following properties:

\hat{P}_f

1) $\hat{P}_a^2 = \hat{P}_a \quad \hat{P}_f^2 = \hat{P}_f \quad \text{and} \quad \hat{P}_a \hat{P}_f = \hat{P}_f \hat{P}_a = 0 \quad \Rightarrow \text{Projectors}$

2) \hat{P}_a commutes with the time and space differential operators

The average part of the field must obey the macroscopic Maxwell's equations

Macroscopic average

The differences between the microscopic fields and the averaged (macroscopic) fields are called the local fields

Complexity of the problem:

- Macroscopic external field \Rightarrow induced fields
- The macroscopic procedure must take into account the fact that all the components of the induced fields will create the response.

Procedure:

- model for the system expressed in terms of an hamiltonian
 - microscopic response of the system (linear-response theory for instance)
- \Rightarrow Definition of the microscopic dielectric tensor $\vec{D}(\vec{r}, \omega) = \int d\vec{r}' \epsilon(\vec{r}, \vec{r}', \omega) E(\vec{r}', \omega)$
- Averaging: definition of ϵ_M which relates the average parts of D and E

Macroscopic average

Infinite crystals

Functions having the crystal symmetries $V(\mathbf{r} + \mathbf{R}) = V(\mathbf{r})$, where \mathbf{R} is any vector of the **Bravais** lattice, can be represented by the Fourier series

$$V(\vec{r}, \omega) = \sum_{\vec{q}, \vec{G}} V(\vec{q} + \vec{G}, \omega) e^{i(\vec{q} + \vec{G})\vec{r}}$$

It can be also written as

$$V(\vec{r}, \omega) = \sum_{\vec{q}} V(r; \vec{q}, \omega) e^{i\vec{q}\vec{r}}$$

where $V(\vec{r}; \vec{q}, \omega) = \sum_{\vec{G}} V(\vec{q} + \vec{G}, \omega) e^{i\vec{G}\vec{r}}$

is a **periodic function**, with respect to the Bravais lattice.

Varies strongly even if the original wave is a long wave and nearly constant within each cell (contains all the **G-harmonics** of the field).

Macroscopic average

Infinite crystals

Spatial average over a cell of the periodic part

$$\begin{aligned} V(\vec{R}, \omega) &= \langle V(\vec{r}; \vec{q}, \omega) \rangle_R \\ &= \frac{1}{\Omega} \int d\vec{r} \sum_{\vec{G}} V(\vec{q} + \vec{G}, \omega) e^{i\vec{G}\vec{r}} \\ &= V(\vec{q} + \vec{0}, \omega) \end{aligned}$$

The macroscopic average corresponds to the $G=0$ component.

⇔ Truncation that eliminates all wave vectors outside the first Brillouin zone
(wave-vector truncation)

Macroscopic quantities have all their G components equal to 0, except the $G=0$ component.

→ Satisfies the two criteria previously defined

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Macroscopic average

A simple example: the longitudinal case

All the fields can be expressed in terms of potentials ($\mathbf{E} = -\nabla V$)

The longitudinal **dielectric function** is defined as

$$V_{ext}(\vec{r}, \omega) = \int d\vec{r}' \epsilon(\vec{r}, \vec{r}', \omega) V_{tot}(\vec{r}', \omega) \quad (\text{Real space})$$

$$V_{ext}(\vec{q} + \vec{G}, \omega) = \sum_{G'} \epsilon(\vec{q} + \vec{G}, \vec{q} + \vec{G}', \omega) V_{tot}(\vec{q} + \vec{G}', \omega)$$

(Reciprocal space)

V_{ext} is a macroscopic quantity : $V_{ext}(\vec{q} + \vec{G}, \omega) = V_{ext}(q, \omega) \delta_{G0}$

This is not the case for $V_{tot}(\vec{q} + \vec{G}, \omega)$

Macroscopic average of V_{ext} :

$$V_{ext}(\vec{q}, \omega) = \sum_{G'} \epsilon_{0G'}(\vec{q}, \omega) V_{tot}(q + G', \omega) \neq \epsilon_{00}(\vec{q}, \omega) V_{tot}(q, \omega)$$

The average of the product is not the product of the averages

Macroscopic average

A simple example: the longitudinal case

We have also
$$V_{tot}(\vec{q} + \vec{G}, \omega) = \sum_{G'} \epsilon_{GG'}^{-1}(\vec{q}, \omega) V_{ext}(q + G', \omega)$$

where is ϵ_{GG}^{-1} , the inverse dielectric function :

$$\sum_{G''} \epsilon_{GG''}(\vec{q}, \omega) \epsilon_{G''G'}^{-1}(\vec{q}, \omega) = \delta_{GG'}$$

Macroscopic average of V_{tot} :

V_{ext} is macroscopic $\Rightarrow V_{tot}(\vec{q} + \vec{G}, \omega) = \epsilon_{G0}^{-1}(\vec{q}, \omega) V_{ext}(q, \omega)$

$$V_{tot}(\vec{q}, \omega) = \epsilon_{00}^{-1}(\vec{q}, \omega) V_{ext}(q, \omega)$$

Macroscopic average

A simple example: the longitudinal case

Macroscopic dielectric constant:

$$V_{ext}(\vec{q}, \omega) = \epsilon_M(\vec{q}, \omega) V_{tot}(\vec{q}, \omega) \Rightarrow \epsilon_M(\vec{q}, \omega) = \frac{1}{\epsilon_{00}^{-1}(\vec{q}, \omega)}$$

- Inversion of the full dielectric matrix $\epsilon_{GG'}(\vec{q}, \omega) \rightarrow \epsilon_{GG'}^{-1}(\vec{q}, \omega)$
- We take the $G=G'=0$ component of $\epsilon_{GG'}^{-1}(\vec{q}, \omega)$

Interpretation

All the microscopic components of the induced field will couple together to produce the macroscopic response

Macroscopic average

Another example: the Lorentz model

For non interacting dipoles (dilute media)

$$\vec{P} = N\alpha \vec{E}_{loc}$$

α : polarisability of the dipole (atoms or molecules)

N : number of dipoles per unit volume (density)

\vec{E}_{loc} : local field acting on a given dipole

If $\vec{E}_{loc} = \vec{E}_{tot}$, we have
$$\vec{D} = \vec{E}_{tot} + 4\pi \vec{P} = \vec{E}_{tot} + 4\pi N\alpha \vec{E}_{tot}$$

$$\vec{D} = \epsilon_M \vec{E}_{tot} \Rightarrow \epsilon_M = 1 + 4\pi N\alpha$$

Mosotti (1850)
Claiusius (1879)
Lorentz (1909)

Macroscopic average

The Lorentz model

In fact, $\mathbf{E}_{loc} \neq \mathbf{E}_{tot}$ $\vec{E}_{loc} = \vec{E}_{tot} + \frac{4}{3}\pi \vec{P}$ in the Lorentz model
(for isotropic or cubic media)

$$\vec{P} = \frac{N\alpha}{1 - \frac{4\pi}{3}N\alpha} \vec{E}_{tot}$$

$$\epsilon_M = 1 + \frac{4\pi N\alpha}{1 - \frac{4\pi}{3}N\alpha}$$

- ✓ Based on classical physics, does not include microscopic (quantum-mechanical) effects
- ✓ Increase of the static dielectric function
- ✓ Red shift of the transition frequency
- ✓ Not the case, when including microscopic effects


$$\alpha(\omega) = \frac{f_{01}^2}{\omega_{01}^2 - \omega^2} \Rightarrow$$

$$\epsilon_M = 1 + \frac{4\pi N f_{01}^2}{\omega_{01}^2 - \omega^2 - \frac{4\pi}{3} N f_{01}^2}$$

Macroscopic average



Summary

- We have defined microscopic and macroscopic fields
 - Microscopic quantities have to be averaged to be compared to experiments
 - The dielectric function has
 - a microscopic expression (related to quantum mechanics)
 - a macroscopic expression (classical scheme - Maxwell's equations)
 - Absorption $\leftrightarrow \text{Im} \{ \epsilon_M \}$ and EELS $\leftrightarrow -\text{Im} \{ 1/\epsilon_M \}$
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Dielectric tensor for cubic symmetries

Useful definitions

Longitudinal fields

$$\vec{\nabla} \times \vec{E}(\vec{r}) = 0 \quad \text{or} \quad \vec{k} \times \vec{E}(\vec{k}) = 0$$

E(k) propagates along k

Examples:

- plasmon oscillations
- scattering
- electron energy loss

Transverse fields

$$\vec{\nabla} \cdot \vec{E}(\vec{r}) = 0 \quad \text{or} \quad \vec{k} \cdot \vec{E}(\vec{k}) = 0$$

E(k) propagates perpendicular to k

Examples:

- photons
- optical properties of solids

Some definitions: $\vec{E}(\vec{r}) \leftrightarrow \vec{E}(\vec{k})$ (Fourier transform)

with $\vec{k} = \vec{q} + \vec{G}$ for crystals

(q is in the first Brillouin zone and G is a reciprocal lattice vector)

Dielectric tensor for cubic symmetries

Transverse-longitudinal decomposition:

Any vector field can be split into **longitudinal** and **transverse** components

$$\vec{E} = \vec{E}^L + \vec{E}^T \quad \text{with} \quad \vec{\nabla} \times \vec{E}^L = 0 \quad \text{and} \quad \vec{\nabla} \cdot \vec{E}^T = 0$$

$$\vec{E}^L(\vec{k}) = \hat{k} \left[\hat{k} \cdot \vec{E}(\vec{k}) \right] \quad \text{and} \quad \hat{k} = \frac{\vec{k}}{k} \quad (\text{In real space, the relations are nonlocal})$$

Macroscopic dielectric tensor

The relation $\mathbf{D}(\mathbf{q}, \omega) = \boldsymbol{\varepsilon}_M(\mathbf{q}, \omega) \mathbf{E}_{\text{tot}}(\mathbf{q}, \omega)$ can be written in terms of the **longitudinal** and **transverse** components

$$\begin{pmatrix} D^L \\ D^T \end{pmatrix} = \begin{pmatrix} \varepsilon_M^{LL} & \varepsilon_M^{LT} \\ \varepsilon_M^{TL} & \varepsilon_M^{TT} \end{pmatrix} \begin{pmatrix} E_{\text{tot}}^L \\ E_{\text{tot}}^T \end{pmatrix}$$

Dielectric tensor for cubic symmetries

Question:

How can we make the link between

- the microscopic dielectric tensor

$$\vec{D}(\vec{q} + \vec{G}, \omega) = \sum_{\vec{G}'} \vec{\epsilon}(\vec{q} + \vec{G}, \vec{q} + \vec{G}', \omega) E_{tot}(\vec{q} + \vec{G}', \omega)$$

Microscopic
components
of \mathbf{D} and \mathbf{E}_{tot}

- the macroscopic dielectric tensor

$$\vec{D}(\vec{q}, \omega) = \vec{\epsilon}_M(\vec{q}, \omega) E_{tot}(\vec{q}, \omega)$$

Macroscopic
components
of \mathbf{D} and \mathbf{E}_{tot}

Dielectric tensor for cubic symmetries

No symmetry

$$\vec{\epsilon}_M(\vec{q}, \omega) = \begin{pmatrix} \epsilon_M^{LL} & \epsilon_M^{LT} \\ \epsilon_M^{TL} & \epsilon_M^{TT} \end{pmatrix}$$

A longitudinal (**transverse**) perturbation induces longitudinal and **transverse** responses.

Cubic symmetry with $q \rightarrow 0$

$$\vec{\epsilon}_M(\vec{q}, \omega) = \begin{pmatrix} \epsilon_M^{LL} & 0 \\ 0 & \epsilon_M^{TT} \end{pmatrix}$$

- A longitudinal perturbation induces a longitudinal response only.
- A transverse perturbation induces a transverse response only.
- Independent of the direction of \mathbf{q}

This holds only for macroscopic quantities

The microscopic dielectric tensor has off-diagonal elements ϵ^{LT} and ϵ^{TL}

Cubic symmetries with $q \rightarrow 0$

Longitudinal dielectric function

$$\epsilon_M^{LL}(\omega) = \lim_{q \rightarrow 0} \frac{1}{1 + \frac{4\pi}{q^2} \chi_{\rho\rho}(\vec{q}, \omega)}$$

where $\chi_{\rho\rho}(\mathbf{q}, \omega)$ is the density-density response function (TDDFT), defined as

$$\rho_{ind}(\vec{q}, \omega) = \chi_{\rho\rho}(\vec{q}, \omega) V_{ext}(\vec{q}, \omega)$$

Transverse dielectric function

$$\lim_{q \rightarrow 0} \epsilon_M^{TT}(\vec{q}, \omega) = \epsilon_M^{LL}(\omega)$$

Dielectric tensor

The tensor is diagonal and contains only one quantity $\epsilon_M^{LL}(\omega)$

Cubic symmetries with $q \neq 0$

Longitudinal dielectric function

One can show that the relation

$$\epsilon_M^{LL}(\vec{q}, \omega) = \frac{1}{1 + \frac{4\pi}{q^2} \chi_{\rho\rho}(\vec{q}, \omega)}$$

holds also when $\vec{q} \neq 0$

depends on \vec{q}

Transverse dielectric function

$$\epsilon_M^{TT}(\vec{q}, \omega) \neq \epsilon_M^{LL}(\vec{q}, \omega)$$

We have also $\epsilon_M^{TL}(\vec{q}, \omega) \neq 0$ $\epsilon_M^{LT}(\vec{q}, \omega) \neq 0$

These quantities are much more complicated and need further approximation to be computed (cannot be expressed in terms of TDDFT).

Cubic symmetries

Summary

- We have defined the longitudinal and transverse components of the dielectric tensor.
- In the long wavelength limit $q \rightarrow 0$, only one quantity is needed (optical isotropy)


$$\varepsilon_M^{LL}(\omega) = \varepsilon_M^{TT}(\omega) = \lim_{q \rightarrow 0} \frac{1}{1 + \frac{4\pi}{q^2} \chi_{\rho\rho}(\vec{q}, \omega)}$$

- For $q \neq 0$, only ε_M^{LL} has a simple expression in terms of the response function.

Cubic symmetries



Some references

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Non-cubic symmetries



Properties of the macroscopic quantities

$$\vec{\epsilon}_M(\vec{q}, \omega) = \begin{pmatrix} \epsilon_M^{LL}(\vec{q}, \omega) & \epsilon_M^{LT}(\vec{q}, \omega) \\ \epsilon_M^{TL}(\vec{q}, \omega) & \epsilon_M^{TT}(\vec{q}, \omega) \end{pmatrix}$$



Non-cubic symmetries

Dielectric tensor - General case

$$\vec{\epsilon}_M(\vec{q}, \omega) = 1 + 4\pi\vec{\alpha}(\vec{q}, \vec{q}, \omega) \left[1 + 4\pi\hat{q} \frac{\hat{q}\vec{\alpha}(\vec{q}, \vec{q}, \omega)}{1 - 4\pi\alpha^{LL}(\vec{q}, \vec{q}, \omega)} \right]$$

Quasipolarisability α : $\vec{j}_{ind}(\vec{q} + \vec{G}, \omega) = \sum_{\vec{G}'} \underline{\vec{\alpha}(\vec{q} + \vec{G}, \vec{q} + \vec{G}', \omega)} \vec{E}_{pert}(\vec{q} + \vec{G}', \omega)$

But one can show that the relation :
holds also for the non-cubic symmetries.

$$\epsilon_M^{LL}(\vec{q}, \omega) = \frac{1}{1 - 4\pi\alpha^{LL}(\vec{q}, \vec{q}, \omega)}$$

and we have $\alpha^{LL}(\vec{q}, \vec{q}, \omega) = -\frac{1}{q^2} \chi_{\rho\rho}(\vec{q}, \vec{q}, \omega)$

Longitudinal-longitudinal
dielectric function

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Non-cubic symmetries $q \rightarrow 0$

Main general result concerning ϵ_M : ϵ_M is an analytic function of \vec{q}

The limit $q \rightarrow 0$ does not depend on the direction of q .

\Rightarrow We can define $\epsilon_M(\omega)$ as $\epsilon_M(\omega) = \lim_{\vec{q} \rightarrow 0} \epsilon_M(\vec{q}, \omega)$

ϵ_M^{LL} is not analytic in the general case

Depending on the **symmetry** of the system, one can define the **3 principal axis, if they exist**, $(\mathbf{n}_1, \mathbf{n}_2, \mathbf{n}_3)$ defining a frame in which $\epsilon_M(\omega)$ is diagonal.

If \vec{E}_{tot} is parallel to one of these axis \mathbf{n}_i $\vec{\epsilon}_M(\omega) \vec{E}_{tot}(\omega) = \epsilon_i(\omega) \vec{E}_{tot}(\omega)$

$\epsilon_i(\omega)$ can be calculated as a **longitudinal dielectric function** $\epsilon_i(\omega) = \epsilon_M^{LL}(\vec{n}_i, \omega)$

Non-cubic symmetries $q \rightarrow 0$

The distinction between longitudinal and transverse is not meaningful

The only important direction is the direction of the electric field

If $q \rightarrow 0$, the fields do not propagate

Existence of the principal frame ? ϵ_M is symmetric but complex ...

No general answer

Use of geometrical arguments

Symmetries

- Cubic
- Hexagonal
- Orthorombic

- Monoclinic
- Triclinic

Shorter wavelength $q \neq 0$

Alternative:

We can use $\vec{\epsilon}_M(\vec{q}, \omega) = 1 + 4\pi\vec{\alpha}(\vec{q}, \vec{q}, \omega) \left[1 + 4\pi\hat{q} \frac{\hat{q}\vec{\alpha}(\vec{q}, \vec{q}, \omega)}{1 - 4\pi\alpha^{LL}(\vec{q}, \vec{q}, \omega)} \right]$

where $\vec{j}_{ind}(\vec{q} + \vec{G}, \omega) = \sum_{\vec{G}'} \vec{\alpha}(\vec{q} + \vec{G}, \vec{q} + \vec{G}', \omega) \vec{E}_{pert}(\vec{q} + \vec{G}', \omega)$

The induced current can be evaluated through the (Time-Dependent)-Density-Current Functional Theory (TD-DCFT)

Which quantities do we need?

Electron Energy Loss Spectroscopy: $-\text{Im} \left\{ \frac{1}{\epsilon(\vec{q}, \omega)} \right\}$

In that case, $\epsilon(\vec{q}, \omega) = \epsilon_M^{LL}(\vec{q}, \omega)$

with $V_{ext}(\vec{q}, \omega) = \epsilon(\vec{q}, \omega) V_{tot}(\vec{q}, \omega)$

Is this correct? The perturbation is longitudinal.
What about the transverse response?

One can show that

$$\vec{E}^T(\vec{q}, \omega) = \frac{\omega^2}{c^2 q^2} \vec{D}^T(\vec{q}, \omega) \quad \text{and} \quad \frac{\omega^2}{c^2 q^2} \approx \frac{v^2}{c^2}$$

In the nonrelativistic approximation, the transverse fields are negligible and the LL component of the dielectric tensor describes the energy loss of charged particles

Outline

1. Introduction: which quantities do we need

2. Macroscopic average

- Definition

- Examples

- Dielectric tensor for cubic symmetries

- Dielectric tensor for non-cubic symmetries

- Properties

- Principal axis

- Summary

Summary



- ✓ The key quantity is the dielectric tensor.
 - ✓ Relation between microscopic and macroscopic fields.
 - ✓ For cubic crystals, the longitudinal dielectric function $\epsilon_M(\omega)$ defines entirely the optical response in the long wavelength limit ($q \rightarrow 0$).
 - ✓ For non-cubic crystals, the dielectric functions calculated along the principal axis can be used to define entirely the optical response in the long wavelength limit.
 - ✓ For non-vanishing momentum, the situation is not so simple: $\epsilon_M^{LL}(\mathbf{q}, \omega)$ only can be defined in a simple way.
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