Seminar: Pavia (2008. Nov.18 & 20)

Optical response theories:
from microscopic to macroscopic

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Toyota Physical and Chemical Research Institute,

1. Optical response of Nanostructures, (Springer Verlag, 2003)

2. J. Phys.: Condens. Matter, 20 (2008) 175202; Cond-mat/0611235

3. phys. stat. sol. (b), 245 (2008) 2692

Dedication to

Professor F. Bassani

Dec. 2006, Pisa

Light-matter interaction

- Light (Oscil. EM field); Matter (Charged particles)
- Light → Vibration of charges → Oscil. polarization
  → emit EM waves → further polarize matter
  → multifold influence between
  matter polarization and EM wave
- → optical response :
  Sum of all the polarization and EM field
- Methods of description:
  QED vs. Semiclassical theories (micro- & macrosc.)

Basic Theories of Light –matter Int.

- Light： Macrosc. vs. Microsc. Maxwell eqs.
  classical vs. quantized field
- Matter：Q-mechanics（Relativistic or non-rel.）

⇓

A. QED → atomic systems
B. Microsc. Nonlocal Theor. → nanostructures
C. Macrosc. Local Theor. → macro-systems

Basic structure of theories A, B, & C

- Light : (***E***, ***B***) or (vector & scalar pot.)
- Matter : (***P***, ***M***) or（***J***, charge density）

EM theory : ***J*** → induced EM field

Matter Q-theory ： EM field → induced ***J***

Self-consistent determ. of these two relations （i.e., solve simultaneous eqs.)

⇩

Optical Response ：(for a given initial condition)

→ Induced change in matter & EM field

Relation between theories "A, B, C"

Framework of B :

from general Lagrangian for matter and EM field

Its Lagrange eqs. :

⇒ { microscopic Maxwell eqs
Newton eq. for particles（Lorentz force）

⇒ microsc. Response（Q-mechanics for matter）

If we further

quantize EM field in B → A (QED)

take macrosc. average in B → C (macrosc. & local) motion of charged particles : Cl. or Q- mechanics

Motion of EM field : Maxwell eqs.

Lagrangian containing both of them :

$$L = \sum_\ell [\frac{1}{2}m_\ell v_\ell^2 - e_\ell \phi(\boldsymbol{r}_\ell) + \frac{e_\ell}{c}\boldsymbol{v}_\ell \cdot \boldsymbol{A}(\boldsymbol{r}_\ell)\ ] + \int \mathrm{d}\boldsymbol{r}\ \mathcal{L}_{EM}$$

$$\mathcal{L}_{EM} = \frac{1}{8\pi}\{(\underbrace{\frac{1}{c}\frac{\partial \boldsymbol{A}}{\partial t} + \nabla\phi}_{-\boldsymbol{E}})^2 - (\underbrace{\nabla \times \boldsymbol{A}}_{\boldsymbol{B}})^2\} = \frac{1}{8\pi}(\boldsymbol{E}^2 - \boldsymbol{B}^2)$$

→ Lagrange eqs. (Coulomb gauge)

Newton eq

$$m_\ell \frac{\mathrm{d}\boldsymbol{v}_\ell}{\mathrm{d}t} == e_\ell\left(\boldsymbol{E} + \frac{\boldsymbol{v}_\ell}{c} \times \boldsymbol{B}\right)$$

Maxwell eqs

$$\nabla^2\phi = -4\pi\rho$$

$$\frac{1}{c^2}\frac{\partial^2 \boldsymbol{A}}{\partial t^2} - \nabla^2 \boldsymbol{A} = \frac{4\pi}{c}\boldsymbol{J}(\boldsymbol{r})_{\mathbf{T}}$$

Charge & current densities

$$\rho(\boldsymbol{r}) = \sum_\ell e_\ell\ \delta(\boldsymbol{r} - \boldsymbol{r}_\ell).$$

$$\boldsymbol{J}(\boldsymbol{r}) = \sum_\ell e_\ell\ \boldsymbol{v}_\ell\ \delta(\boldsymbol{r} - \boldsymbol{r}_\ell).$$

Microscopic response theory

Lagrangian of previous slide is reliable basis.

→ Hamiltonian in EM field (Coulomb gauge)

$$H_{\rm M} = \sum_\ell \frac{1}{2m_\ell}[\boldsymbol{p}_\ell - \frac{e_\ell}{c}\boldsymbol{A}(\boldsymbol{r}_\ell)]^2 + \underline{\frac{1}{2}\sum \sum_{\ell \neq \ell '} \frac{e_\ell e_{\ell '}}{|\boldsymbol{r}_\ell - \boldsymbol{r}_{\ell '}|}}$$

Scalar pot. -related terms
(Int. energy + self-energy)

→ Hamiltonian of EM field

$$H_{\rm EM} = \int {\rm d}\boldsymbol{r}(\boldsymbol{E}_{\rm T}^2 + \boldsymbol{B}^2)$$

$$\boldsymbol{E}_{\rm T} = -\frac{1}{c}\frac{\partial \boldsymbol{A}}{\partial t}$$

Conjugate momenta for $\boldsymbol{r}_\ell$ and $\boldsymbol{A}(\boldsymbol{r})$

$$\boldsymbol{p}_\ell = m_\ell \boldsymbol{v}_\ell + \frac{e_\ell}{c}\boldsymbol{A}(\boldsymbol{r}_\ell)$$

$$\boldsymbol{\Pi}(\boldsymbol{r}) = \frac{1}{4\pi c^2}\frac{\partial \boldsymbol{A}}{\partial t} = -\frac{1}{4\pi c}\boldsymbol{E}_{\rm T}$$

Linear term of interaction

$$H'_{\rm int} = -\frac{1}{c}\int {\rm d}\boldsymbol{r}\ \boldsymbol{J}(\boldsymbol{r}) \cdot \underline{\boldsymbol{A}(\boldsymbol{r}, t)},$$

Transverse (Coulomb gauge)

Relativistic corrections (when necessary)

- spin-orbit int., Mass velocity term, Darwin term,
- spin Zeeman term

$$H_{\rm sZ} = -\int {\rm d}\boldsymbol{r}\ \boldsymbol{M}_{\rm spin}(\boldsymbol{r}) \cdot \boldsymbol{B}(\boldsymbol{r}) = -\frac{1}{c}\int {\rm d}\boldsymbol{r}\ \boldsymbol{J}_{\rm spin}(\boldsymbol{r}) \cdot \boldsymbol{A}(\boldsymbol{r})$$

$$\boldsymbol{J}_{\rm spin}(\boldsymbol{r}) = c\nabla \times \boldsymbol{M}_{\rm spin}(\boldsymbol{r})$$

$$\boldsymbol{M}_{\rm spin}(\boldsymbol{r}) = \sum_{\ell} \beta_\ell \boldsymbol{s}_\ell\ \delta(\boldsymbol{r} - \boldsymbol{r}_\ell)$$

Total current density $$\boldsymbol{I}(\boldsymbol{r}) = \boldsymbol{J}(\boldsymbol{r}) + \boldsymbol{J}_{\rm spin}(\boldsymbol{r})$$

Linear Interaction term $$H'_{\rm int} = -\frac{1}{c}\int {\rm d}\boldsymbol{r}\ \boldsymbol{J}(\boldsymbol{r}) \cdot \boldsymbol{A}(\boldsymbol{r}, t), \Big|_{\boldsymbol{J} \to \boldsymbol{I}}$$

Matter Hamiltonian = $H_{\rm M}\Big|_{\rm A=0}$ + remaining relativ. correction

Operator forms of $\{\rho,\ \boldsymbol{J},\ \boldsymbol{P},\ \boldsymbol{M}\}$

Charge density $$\rho(\boldsymbol{r}) = \sum_\ell e_\ell\, \delta(\boldsymbol{r} - \boldsymbol{r}_\ell)\ ,$$

Current density $$\boldsymbol{J}(\boldsymbol{r}) = \sum_\ell e_\ell\, \boldsymbol{v}_\ell\, \delta(\boldsymbol{r} - \boldsymbol{r}_\ell)\ ,$$

$$\nabla \cdot \boldsymbol{J} + \frac{\partial \rho}{\partial t} = 0$$

Expected relations for ***P***, ***M***

$$\nabla \cdot \boldsymbol{P} = -\rho\ ,$$
$$\boldsymbol{J} = \frac{\partial \boldsymbol{P}}{\partial t} + c\, \nabla \times \boldsymbol{M}\ .$$

Cohen-Tannoudji et al. " *Photons and Atoms*"

El. Pol. $$\boldsymbol{P}(\boldsymbol{r}) = \int_0^1 \mathrm{d}u \sum_\ell e_\ell \boldsymbol{r}_\ell\, \delta(\boldsymbol{r} - u\boldsymbol{r}_\ell)$$

Mag. Pol. $$\boldsymbol{M}(\boldsymbol{r}) = \int_0^1 u\, \mathrm{d}u \sum_\ell e_\ell \boldsymbol{r}_\ell \times \boldsymbol{v}_\ell\, \delta(\boldsymbol{r} - u\boldsymbol{r}_\ell).$$

Calculation of induced current density at time t : $< \Psi(t)|\boldsymbol{I}(\boldsymbol{r})|\Psi(t) >$

Time dep. Schrodinger eq. $i\hbar\partial\Psi/\partial t = (H_0 + H_{\rm int})\Psi$

Int. representation $\Psi = \exp(-iH_0\tau/\hbar)\tilde{\Psi}$ ⟹ $i\hbar\partial\tilde{\Psi}/\partial t = H'(t)\tilde{\Psi}$

$$H'(\tau) = \exp(iH_0\tau/\hbar)\ H_{\rm int}\ \exp(-iH_0\tau/\hbar)$$

Iterative solution $\tilde{\Psi}(t) = \tilde{\Psi}(-\infty) - \frac{i}{\hbar}\int_{-\infty}^{t} {\rm d}\tau\ H'(\tau)\tilde{\Psi}(-\infty) + \cdots$

J(**r**) contains ***A***

$$\boldsymbol{J}(\boldsymbol{r}) = \sum_\ell \frac{e_\ell}{2m_\ell}[\boldsymbol{p}_\ell\delta(\boldsymbol{r}-\boldsymbol{r}_\ell) + \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\boldsymbol{p}_\ell] - \frac{1}{c}\hat{N}(\boldsymbol{r})\boldsymbol{A}(\boldsymbol{r},t)$$

$$\hat{N}(\boldsymbol{r}) = \textstyle\sum_\ell (e_\ell^2/m_\ell)\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)$$

Separate ***A***-dep. term from total ***I(r)***

$\boldsymbol{I}(\boldsymbol{r}) = \boldsymbol{J}(\boldsymbol{r}) + \boldsymbol{J}_{\rm spin}(\boldsymbol{r})$ ⟹ $\boldsymbol{I} = \bar{\boldsymbol{I}} - \frac{1}{c}\hat{N}(\boldsymbol{r})\boldsymbol{A}(\boldsymbol{r},t)$

Evaluate the ***A***-linear term of $< \Psi(t)|\boldsymbol{I}(\boldsymbol{r})|\Psi(t) >$

Microscopic Current density (ω - Fourier comp.)

$$\tilde{\boldsymbol{I}}(\boldsymbol{r},\omega) = \bar{\chi}_0(\boldsymbol{r})\ \boldsymbol{A}(\boldsymbol{r},\omega) + \int \mathrm{d}\boldsymbol{r}' \chi(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot \boldsymbol{A}(\boldsymbol{r}',\omega),$$

separable

$$\chi_{\mathrm{em}}(\boldsymbol{r},\boldsymbol{r}',\omega) = \frac{1}{c}\sum_\nu \left[g_\nu(\omega)\bar{\boldsymbol{I}}_{0\nu}(\boldsymbol{r})\bar{\boldsymbol{I}}_{\nu 0}(\boldsymbol{r}') + h_\nu(\omega)\bar{\boldsymbol{I}}_{\nu 0}(\boldsymbol{r})\bar{\boldsymbol{I}}_{0\nu}(\boldsymbol{r}') \right]$$

$$\left(g_\nu(\omega) = \frac{1}{E_{\nu 0} - \hbar\omega - i0^+},\quad h_\nu(\omega) = \frac{1}{E_{\nu 0} + \hbar\omega + i0^+}. \right)$$

$$\bar{\chi}_0(\boldsymbol{r}) = -\frac{1}{c}\sum_\ell \frac{e_\ell^2}{m_\ell} < 0|\delta(\boldsymbol{r}-\boldsymbol{r}_\ell)|0 >$$

Separability →

simultaneous linear eqs of

$$F_{\mu\nu}(\omega) = \int \mathrm{d}\boldsymbol{r}'\ \bar{\boldsymbol{I}}_{\mu\nu}(\boldsymbol{r}')\cdot \boldsymbol{A}(\boldsymbol{r}',\omega)$$

→Unique solution of EM response

Microsc. Nonlocal Response theory, K. Cho (Springer, 2003)

No boundary condition is needed for EM field.

Rewriting of the term $\bar{\chi}_0(\boldsymbol{r}) = -\frac{1}{c}\sum_{\ell}\frac{e_\ell^2}{m_\ell} < 0|\delta(\boldsymbol{r}-\boldsymbol{r}_\ell)|0 >$

$$\hat{N}(\boldsymbol{r}) = \sum_\ell (e_\ell^2/m_\ell)\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)$$

When relativistic correction is small and LWA is valid

$$\langle 0|\hat{N}(\boldsymbol{r})|0\rangle\ \boldsymbol{A}(\boldsymbol{r}) \cong \sum_\nu \frac{1}{E_{\nu 0}}[\boldsymbol{I}_{0\nu}(\boldsymbol{r})F_{\nu 0}(\omega) + \boldsymbol{I}_{\nu 0}(\boldsymbol{r})F_{0\nu}(\omega)]$$

Then, we can renormalize this term into the “resonant” terms as

$$\widetilde{\boldsymbol{I}}(\boldsymbol{r},\omega) = \frac{1}{c}\sum_\nu[\bar{g}_\nu(\omega)\hat{\boldsymbol{I}}_{0\nu}(\boldsymbol{r})F_{\nu 0}(\omega) + \bar{h}_\nu(\omega)\hat{\boldsymbol{I}}_{\nu 0}(\boldsymbol{r})F_{0\nu}(\omega)],$$

$$\bar{g}_\nu(\omega) = g_\nu(\omega) - \frac{1}{E_{\nu 0}}, \qquad \bar{h}_\nu(\omega) = h_\nu(\omega) - \frac{1}{E_{\nu 0}}.$$

Fundamental eqs. of microsc. Nonlocal theory

Maxwell eq. and constitutive eq.

Maxwell eq. :

$$\frac{1}{c^2}\frac{\partial^2 \boldsymbol{A}}{\partial t^2} - \nabla^2 \boldsymbol{A} = \frac{4\pi}{c}\boldsymbol{I}_{\mathrm{T}}$$

Constitutive eq. :

$$\tilde{I}(\boldsymbol{r},\omega) = (1/c)\sum_{\nu}\left[g_{\nu}(\omega)\bar{\boldsymbol{I}}_{0\nu}(\boldsymbol{r})\underline{F_{\nu 0}(\omega)} + h_{\nu}(\omega)\bar{\boldsymbol{I}}_{\nu 0}(\boldsymbol{r})\underline{F_{0\nu}(\omega)}\right]$$

$$\underline{F_{\mu\nu}(\omega)} = \int \mathrm{d}\bar{\boldsymbol{r}}\ \bar{\boldsymbol{I}}_{\mu\nu}(\bar{\boldsymbol{r}}) \cdot \boldsymbol{A}(\bar{\boldsymbol{r}},\omega)$$

Selfconsistent solution is obtained as

$$\tilde{\boldsymbol{J}}(\mathbf{r},\omega) = \Sigma_\nu \{\bar{\boldsymbol{I}}_{0\nu}(\mathbf{r})\, X_{\nu 0} + \bar{\boldsymbol{I}}_{\nu 0}(\mathbf{r})\, X_{0\nu}\} + \dots$$

$$\boldsymbol{A}(\mathbf{r},\omega) = \boldsymbol{A}^{(0)}(\mathbf{r},\omega) + \mathcal{G}[\tilde{\boldsymbol{J}}]$$

Eqs. to determine X :

$$\mathbf{S}\,\mathbf{X} = \mathrm{F}^{(0)}$$

$$X_{\mu\nu} = F_{\mu\nu} / (E_{\mu\nu} - \hbar\omega - i0^+)$$

$$F_{\mu\nu}(\omega) = \int \bar{\boldsymbol{I}}_{\mu\nu}(\mathbf{r}) \cdot \boldsymbol{A}(\mathbf{r},\omega)\, d\mathbf{r}$$

For linear response in RWA :

$$\Sigma_\nu S_{\mu 0, 0\nu}\, X_{\nu 0} = F^{(0)}_{\mu 0}$$

where

$$S_{\mu 0, 0\nu} = (E_{\mu 0} - \hbar\omega)\,\delta_{\mu\nu} + \tilde{A}_{\mu 0, 0\nu}$$

$$F^{(0)}_{\mu\nu}(\omega) = \int \bar{\boldsymbol{I}}_{\mu\nu}(\mathbf{r}) \cdot \underline{\boldsymbol{A}^{(0)}(\mathbf{r},\omega)}\, d\mathbf{r}$$

Incident field

Light mediated int. between induced current densities

Interaction energy

Retarded interaction of current densities

$$\tilde{A}_{\mu 0,0\nu}(\omega) = \frac{-1}{c^2}\iint \mathrm{dr}\,\mathrm{dr}'\,\bar{I}_{\mu 0}(\mathrm{r}) \cdot \mathbf{G}^{(\mathrm{T})}(\mathrm{r}-\mathrm{r}') \cdot \bar{I}_{0\nu}(\mathrm{r}')$$

Radiative shift and width

Radiation Green's Function (transverse)

$$\mathbf{G}^{(\mathrm{T})}(\mathbf{r}-\mathbf{r}') = \frac{1}{2\pi^2}\int \mathrm{d}\boldsymbol{k}\,\frac{\mathbf{1}-\hat{\mathbf{e}}(\boldsymbol{k})\,\hat{\mathbf{e}}(\boldsymbol{k})}{\mathrm{k}^2-(q+i0^+)^2}\,\mathrm{e}^{i\boldsymbol{k}\cdot(\mathbf{r}-\mathbf{r}')}$$

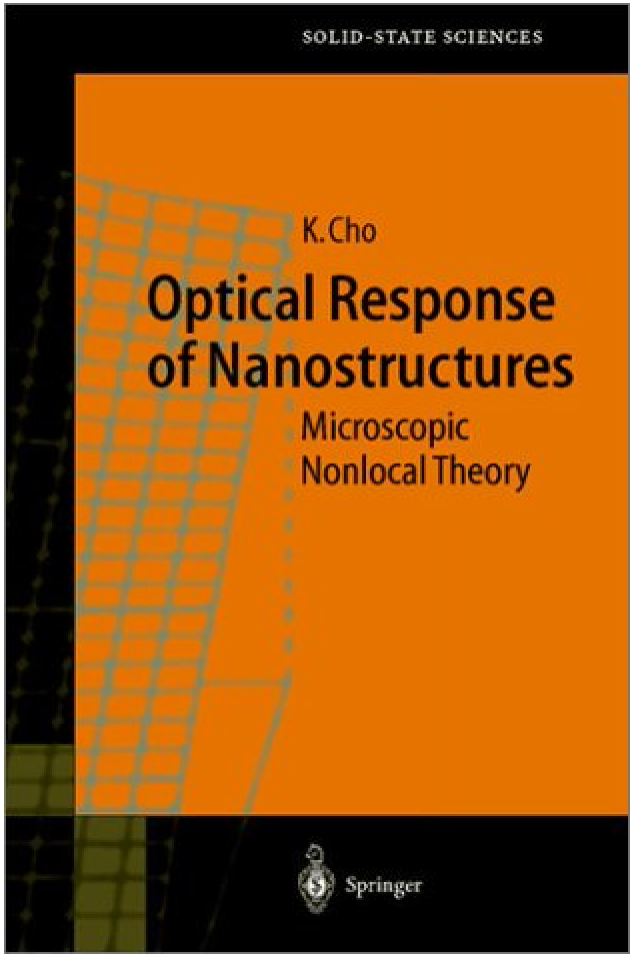
$$(\, q=\omega/c;\ \ \hat{\mathbf{e}}(\boldsymbol{k})=\boldsymbol{k}/|\boldsymbol{k}|\,)$$

Radiative width : good measure of interaction strength

(better than oscillator strength)

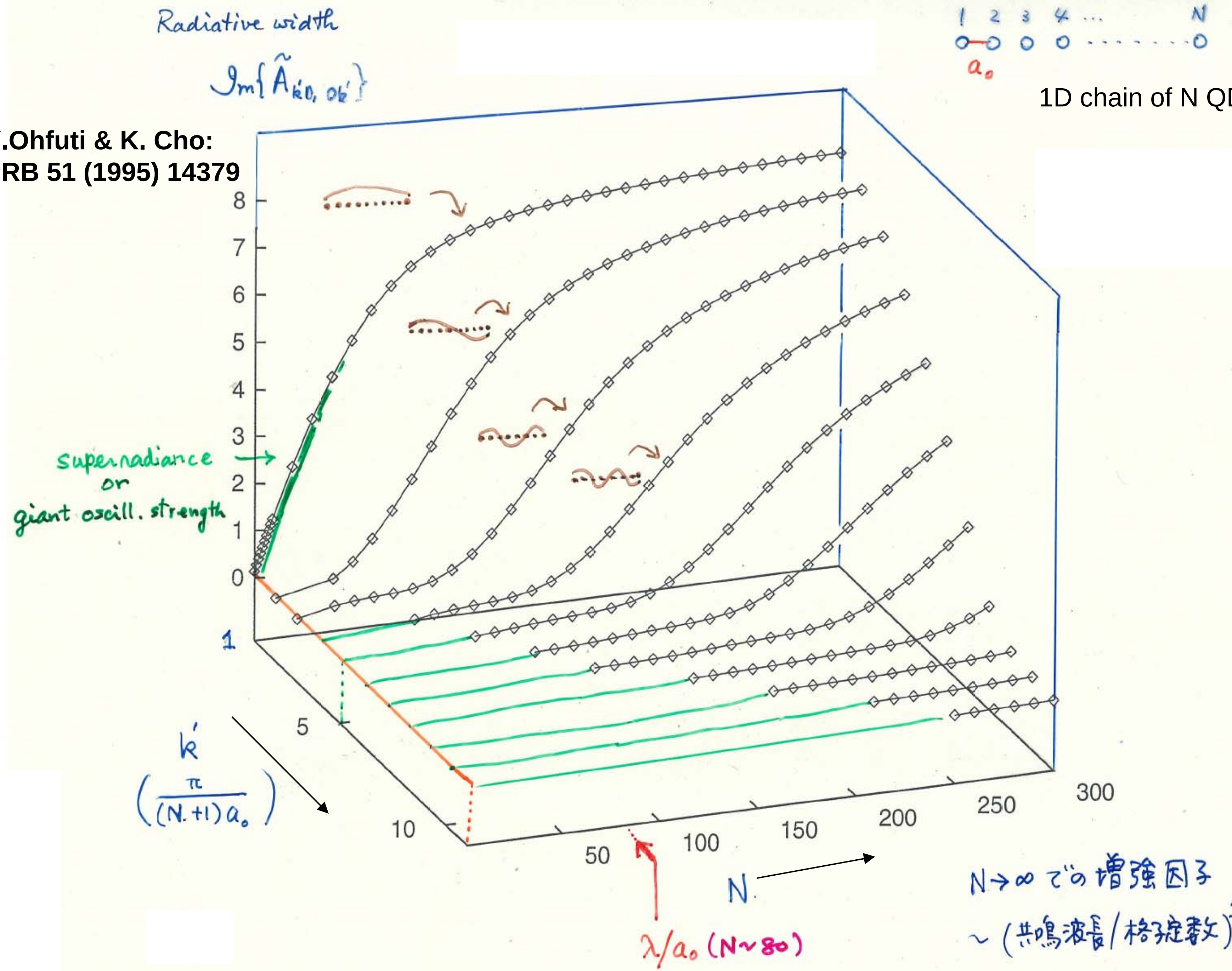
Microsc. Nonlocal Response theory

1. contains microsc. spatial variation
   (all the wavelength components).
   → appropriate for nano-studies
2. Nonlocal susceptibility, separable integral kernel: Integral eq → simultaneous linear eq.
3. Boundary condition is required, not for EM field, but for matter only.
4. Radiative width of matter excitations is included in a natural form.
5. Hierarchy of EM theories :
   A) QED
   B) Microsc. Nonlocal response
   C) macrosc. Local response
6. Natural extension to nonlinear responses
7. Derivation macrosc. Maxwell eqs. : New !



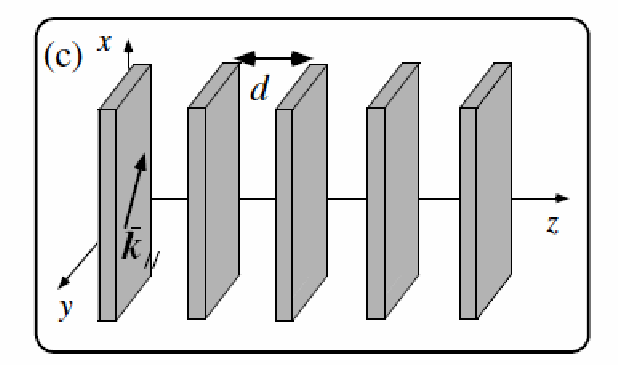
Springer Verlag 2003

Y.Ohfuti & K. Cho:
PRB 51 (1995) 14379



1D chain of N QD's

Resonant DBR structures (N layers)



T.Ikawa & K. Cho,
PRB 05,
J.Phys.Soc.Jpn. 05

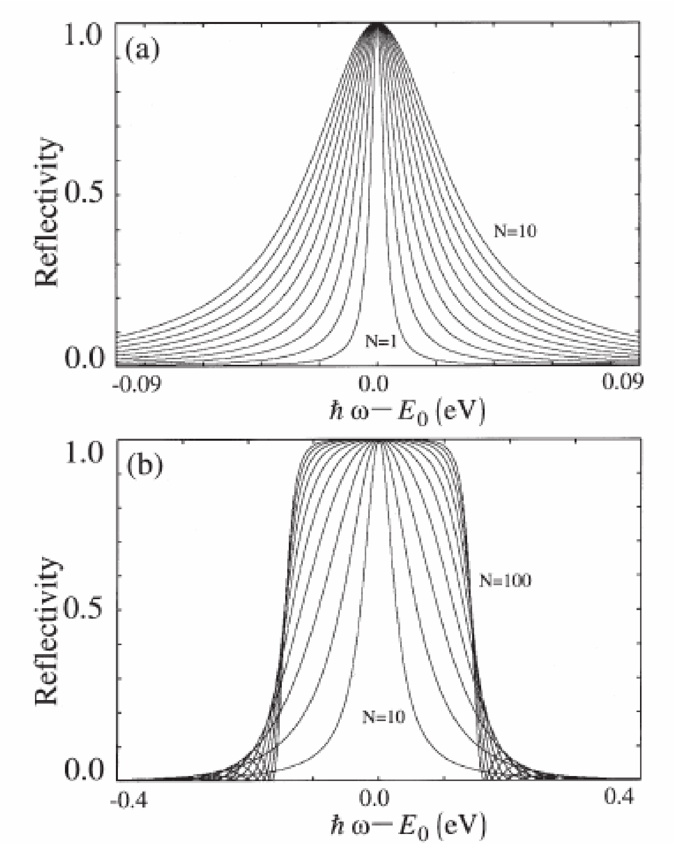
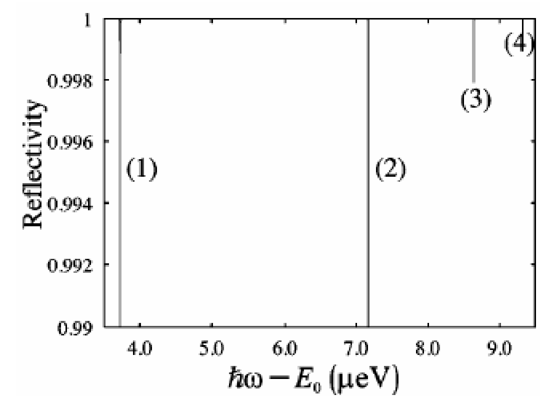
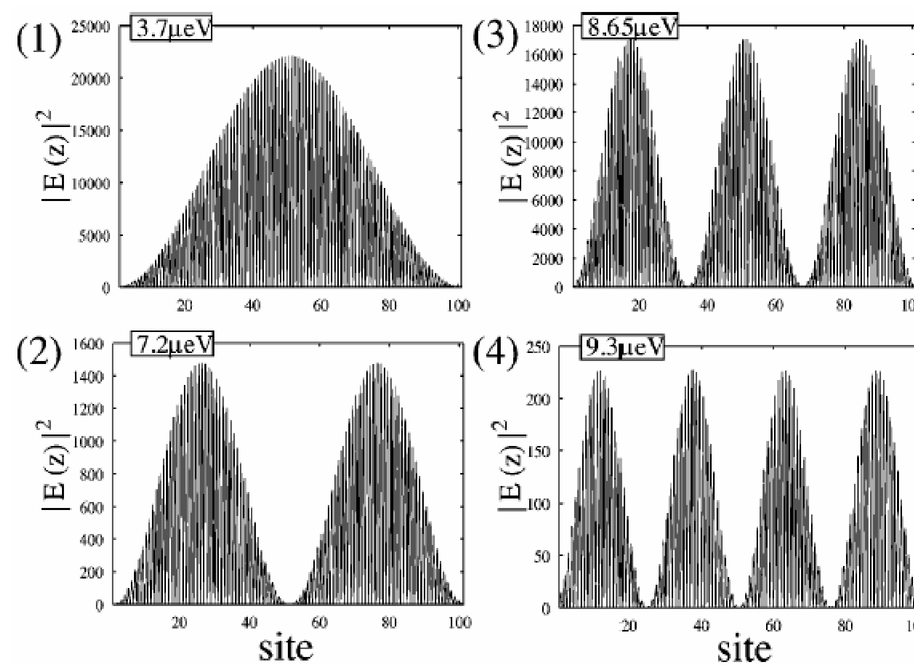


Fig. 9. Reflectivity spectrum of the array of planes for N=1–100 with $d=\lambda_r/2$.

transmission windows in total reflection region

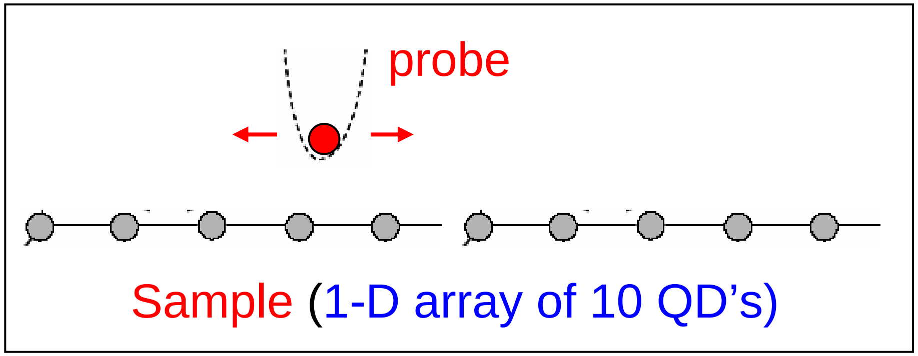


Field patterns of quantized gap modes

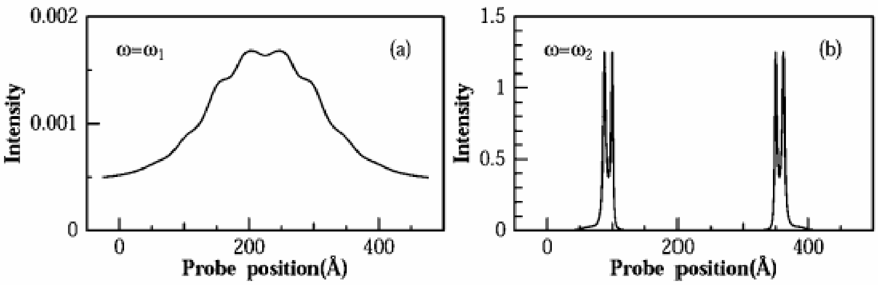


Breakdown of E1 selection rule in resonant SNOM in reflection mode

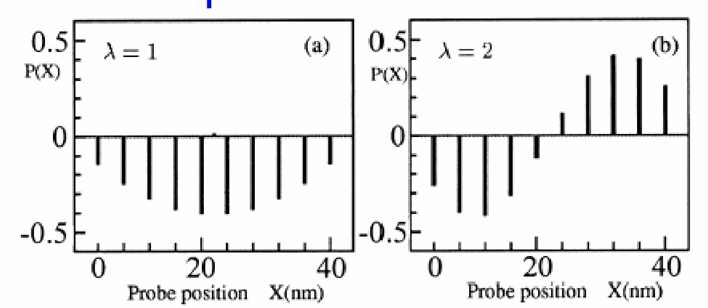
Surf. Sci.: 363 (96) 378



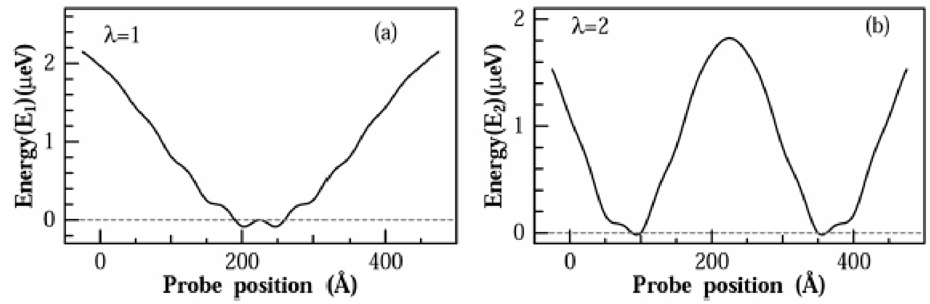
Probe position dep. of signal int.



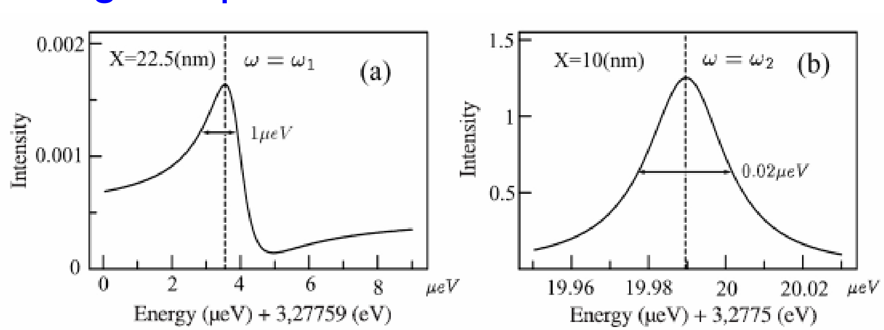
polarization pattern of two modes



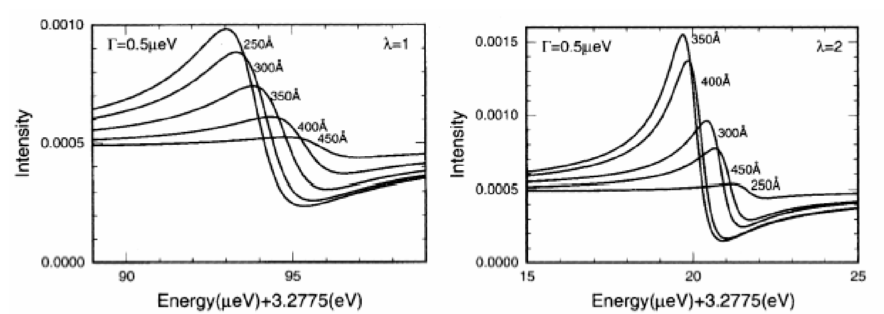
Probe position dep. of resonance freq.



Signal spectrum



Signal spectrum : comparable size !!



Reconstruction of macroscopic Maxwell eqs

Motivation:

1. Conventional form of macrosc. M-eqs. is incomplete.
   Problems exist about "uniqueness" and
   "consistency with microscopic theory"
2. macrosc. M-eqs. is still important today as
   a main tool for research (in metamaterials, photonic crystals, near-field optics, etc.)
   a fundamental subject of physics education

→ worth looking for a more complete form (after over 100 years of their birth)

Macrosc. M-eqs.

- As a phenomenology (19th C), matter = continuum, "concept of electron, q-mechanics, relativity" not existed

- Lorentz' theory of electrons : particle picture of matter phenomenology → microsc. M-eqs. for matter in vac.
  → QED, a highest accuracy theory in physics

- Efforts to derive macrosc. M-eqs. from particle picture via macrosc. average of microsc. M-eqs. in the past seem to be logically incomplete.

Microsc. Maxwell eqs.

$$\nabla \cdot \boldsymbol{E} = 4\pi\rho, \quad \nabla \cdot \boldsymbol{B} = 0,$$

$$\nabla \times \boldsymbol{B} = \frac{4\pi}{c}\boldsymbol{J} + \frac{1}{c}\frac{\partial \boldsymbol{E}}{\partial t},$$

$$\nabla \times \boldsymbol{E} = -\frac{1}{c}\frac{\partial \boldsymbol{B}}{\partial t} .$$

Charge & current densities

in particle picture

$$\rho(\boldsymbol{r}) = \sum_\ell e_\ell \, \delta(\boldsymbol{r} - \boldsymbol{r}_\ell) \ ,$$

$$\boldsymbol{J}(\boldsymbol{r}) = \sum_\ell e_\ell \, \boldsymbol{v}_\ell \, \delta(\boldsymbol{r} - \boldsymbol{r}_\ell) \ ,$$

$$\boldsymbol{v}_\ell = \frac{1}{m_\ell}[\boldsymbol{p}_\ell - \frac{e_\ell}{c}\boldsymbol{A}(\boldsymbol{r}_\ell, t)]$$

Cause & result $\rho, \boldsymbol{J} \Rightarrow \boldsymbol{E}, \boldsymbol{B}$

Charge conservation : $\nabla \cdot \boldsymbol{J} + \frac{\partial \rho}{\partial t} = 0$

Only a single constitutive eq. is needed (between J and A)

Maxwell-eqs. via scalar & vector potentials (Coulomb gauge) :

$$\nabla^2 \phi = -4\pi\rho \ , \quad \frac{1}{c^2}\frac{\partial^2 \boldsymbol{A}}{\partial t^2} - \nabla^2 \boldsymbol{A} = \frac{4\pi}{c}\boldsymbol{J}_{\mathrm{T}}$$

Macroscopic Averaging (conventional way)

$$\rho = \rho_{\mathrm{t}} + \rho_{\mathrm{p}} \ , \quad \rho_{\mathrm{p}} = -\nabla \cdot \boldsymbol{P} \ ,$$

$$\boldsymbol{J} = \boldsymbol{J}_{\mathrm{c}} + c \, \nabla \times \boldsymbol{M} + \frac{\partial \boldsymbol{P}}{\partial t} \ ,$$

Thereby, only restriction is

$$\int \mathrm{d}\boldsymbol{r} \ \rho_{\mathrm{p}} = 0$$

i.e., Polarization charge density arises
from neutral charge distribution, as a whole.

(There is no definite way to define such a distribution.)

Macrosc. Maxwell eqs.

$$\nabla \cdot \boldsymbol{D} = 4\pi\rho_{\mathrm{t}}, \quad \nabla \cdot \boldsymbol{B} = 0,$$

$$\nabla \times \boldsymbol{H} = \frac{4\pi}{c}\boldsymbol{J}_{\mathrm{c}} + \frac{1}{c}\frac{\partial \boldsymbol{D}}{\partial t},$$

$$\nabla \times \boldsymbol{E} = -\frac{1}{c}\frac{\partial \boldsymbol{B}}{\partial t}$$

P* & *M : el. & mag. pol. of matter →

$$\boldsymbol{D} = \boldsymbol{E} + 4\pi \boldsymbol{P}$$

$$\boldsymbol{H} = \boldsymbol{B} - 4\pi \boldsymbol{M}$$

Charge conservation

$$\mathbf{div}\ \boldsymbol{J}_{\mathrm{c}} + (\partial\rho_{\mathrm{t}} / \partial t) = 0$$

Materials constants (tensor)

$$\boldsymbol{P} = \chi_{\mathrm{e}} \boldsymbol{E}, \quad \boldsymbol{M} = \chi_{\mathrm{m}} \boldsymbol{H}$$

$$\boldsymbol{D} = \varepsilon \boldsymbol{E}, \quad \boldsymbol{B} = \mu \boldsymbol{H}$$

$$\varepsilon = 1 + 4\pi\chi_{\mathrm{e}}, \quad \mu = 1 + 4\pi\chi_{\mathrm{m}}$$

2 constitutive eqs.

J → P, M : unique ?

Problems in the conventional arguments

A) No criterion exists to make the unique division of

$$\rho = \rho_{\rm t} + \rho_{\rm p} \quad \text{and} \quad \boldsymbol{J} = \boldsymbol{J}_{\rm c} + c\,\nabla \times \boldsymbol{M} + \frac{\partial \boldsymbol{P}}{\partial t}$$

B) Number of necessary constitutive eqs. :

1 in microsc. M-eqs. , why 2 in macrosc. M-eqs.?

Why the dynamical variables are ***P*** & ***M*** rather than ***J***, in macrosc. M-eqs. ? Is the description in terms of (ε, μ) unconditional ?

C) k-dependence of μ : why different for spin resonance & orbital M1 transition ?

D) Appearance of (ε, μ) in dispersion eq. as a product, not a sum :

→ physically strange. In chiral symmetry, second order poles appear.

***k*-dep. of permeability**

Problem C)

M1 transitions of matter

(1) spin resonance

(2) orbital M1 transition

Usually for (1) :

$$\chi_{\mathrm{m}}(\omega) = \frac{M_0}{\omega_0 - \omega - i\gamma} \qquad \hbar\omega_0 = g\mu_{\mathrm{B}} H_0$$

→ $\mu = 1 + 4\pi\chi_{\mathrm{m}}$

Usually for (2) :

$$\boldsymbol{p}\cdot\boldsymbol{A}(\boldsymbol{r}) \sim \boldsymbol{p}\cdot\boldsymbol{A}_0(1 + i\boldsymbol{k}\cdot\boldsymbol{r}\cdots) \rightarrow i\boldsymbol{k}\cdot < \boldsymbol{r}\boldsymbol{p} > \cdot\boldsymbol{A}_0$$

→ $\mu = 1 + O(k^2)$

?

Form of dispersion equation

Problem D)

$$\frac{c^2k^2}{\omega^2} = \epsilon\mu = (1 + 4\pi\chi_{\mathrm{e}})(1 + 4\pi\chi_{\mathrm{m}})$$

Contribution of E1 & M1 transitions : why as a product ?

All the E1 transitions appear as a sum.

All the M1 transitions appear as a sum.

Why should the two groups contribute as a product.

In chiral symmetry, E1 & M1 transitions are mixed and cannot be distinguished.

→ Their contributions should appear as a sum.

Microscopic Current density (ω - Fourier comp.)

$$\tilde{\boldsymbol{I}}(\boldsymbol{r},\omega) = \bar{\chi}_0(\boldsymbol{r})\ \boldsymbol{A}(\boldsymbol{r},\omega) + \int \mathrm{d}\boldsymbol{r}'\chi(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot\boldsymbol{A}(\boldsymbol{r}',\omega),$$

separable

$$\chi_{\mathrm{em}}(\boldsymbol{r},\boldsymbol{r}',\omega) = \frac{1}{c}\sum_{\nu}\left[g_\nu(\omega)\bar{\boldsymbol{I}}_{0\nu}(\boldsymbol{r})\bar{\boldsymbol{I}}_{\nu 0}(\boldsymbol{r}') + h_\nu(\omega)\bar{\boldsymbol{I}}_{\nu 0}(\boldsymbol{r})\bar{\boldsymbol{I}}_{0\nu}(\boldsymbol{r}')\ \right]$$

$$\left(g_\nu(\omega) = \frac{1}{E_{\nu 0} - \hbar\omega - i0^+},\quad h_\nu(\omega) = \frac{1}{E_{\nu 0} + \hbar\omega + i0^+}\ . \right)$$

$$\bar{\chi}_0(\boldsymbol{r}) = -\frac{1}{c}\sum_{\ell}\frac{e_\ell^2}{m_\ell} < 0|\delta(\boldsymbol{r} - \boldsymbol{r}_\ell)|0 >$$

Separability →

simultaneous linear eqs of

$$F_{\mu\nu}(\omega) = \int \mathrm{d}\boldsymbol{r}'\ \bar{\boldsymbol{I}}_{\mu\nu}(\boldsymbol{r}')\cdot\boldsymbol{A}(\boldsymbol{r}',\omega)$$

→Unique solution of EM response

Microsc. Nonlocal Response theory, K. Cho (Springer, 2003)

No boundary condition is needed for EM field.

Rewriting of the term $\bar{\chi}_0(\boldsymbol{r}) = -\frac{1}{c}\sum_\ell \frac{e_\ell^2}{m_\ell} < 0|\delta(\boldsymbol{r}-\boldsymbol{r}_\ell)|0 >$

$$\hat{N}(\boldsymbol{r}) = \sum_\ell (e_\ell^2/m_\ell)\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)$$

When relativistic correction is small and LWA is valid

$$\langle 0|\hat{N}(\boldsymbol{r})|0\rangle\ \boldsymbol{A}(\boldsymbol{r}) \cong \sum_\nu \frac{1}{E_{\nu 0}}[\boldsymbol{I}_{0\nu}(\boldsymbol{r})F_{\nu 0}(\omega) + \boldsymbol{I}_{\nu 0}(\boldsymbol{r})F_{0\nu}(\omega)]$$

Then, we can renormalize this term into the “resonant” terms as

$$\widetilde{\boldsymbol{I}}(\boldsymbol{r},\omega) = \frac{1}{c}\sum_\nu[\bar{g}_\nu(\omega)\hat{\boldsymbol{I}}_{0\nu}(\boldsymbol{r})F_{\nu 0}(\omega) + \bar{h}_\nu(\omega)\hat{\boldsymbol{I}}_{\nu 0}(\boldsymbol{r})F_{0\nu}(\omega)],$$

$$\bar{g}_\nu(\omega) = g_\nu(\omega) - \frac{1}{E_{\nu 0}}, \qquad \bar{h}_\nu(\omega) = h_\nu(\omega) - \frac{1}{E_{\nu 0}}.$$

Comparison of different logics in macrosc. average

Conventional theories : goal of argument is to find the known form of macrosc. M-eqs.

1. Assume ***P*** & ***M*** as fundamental quantities
2. Look for susceptibilities via appropriate method

New logic：based on microsc. nonlocal response

1. Keep ***J*** as a fundamental variable
2. macrosc. Average = long wavelength approx.
   → extract the eqs for LW components of ***A*** & ***J***
3. Check if the conventional form is reproduced

New Method of derivation:
free from ”models & empirical knowledge”

general Lagrangian for matter-EM field systems

⇨ Matter Hamiltonian & interaction (include spins)

⇨ microsc. Nonlocal response (semi-classical)
(nonlocal susceptibility : q-mechanical details in a model-independent way)

⇨ Apply LWA and find the eqs. among LW components of J and A. Then, compare the result with ordinary macrosc. M-eqs.

Meaning of macrosc. Average :

LWA & statistical average ?

Statistical average has two meanings:

1. Ensemble average over matter initial states
   → included in microsc. susceptibility
2. Distribution of localized quantum states
   → nonlocal susceptibility (microsc.)
   → concentration effect (macrosc.)

→ Only 2 should be considered.

LWA of microscopic Nonlocal response

→ keep the first few terms of **Taylor** expansion of ***A & J***

Effect of LWA

Maxwell eq. (not affected)

$$\frac{1}{c^2}\frac{\partial^2 \boldsymbol{A}}{\partial t^2} - \nabla^2 \boldsymbol{A} = \frac{4\pi}{c}\boldsymbol{I}_{\mathrm{T}}$$

Constitutive eq. (affected)

$$\tilde{I}(\boldsymbol{r},\omega) = (1/c)\sum_{\nu}\left[\bar{g}_{\nu}(\omega)\bar{\boldsymbol{I}}_{0\nu}(\boldsymbol{r})\underline{F_{\nu 0}(\omega)} + \bar{h}_{\nu}(\omega)\bar{\boldsymbol{I}}_{\nu 0}(\boldsymbol{r})\underline{F_{0\nu}(\omega)}\right]$$

Contents of *F* are affected by LWA

$$\underline{F_{\mu\nu}(\omega)} = \int \mathrm{d}\bar{\boldsymbol{r}}\ \bar{\boldsymbol{I}}_{\mu\nu}(\bar{\boldsymbol{r}})\cdot\boldsymbol{A}(\bar{\boldsymbol{r}},\omega)$$

In terms of k- Fourier components,

★ $$\tilde{\boldsymbol{I}}(\boldsymbol{k},\omega) = \frac{1}{c}\sum_{\nu}\left[\bar{g}_{\nu}(\omega)\tilde{\boldsymbol{I}}_{0\nu}(\boldsymbol{k})F_{\nu 0}(\omega) + \bar{h}_{\nu}(\omega)\tilde{\boldsymbol{I}}_{\nu 0}(\boldsymbol{k})F_{0\nu}(\omega)\right]$$

$$F_{\mu\nu}(\omega) = \sum_{\boldsymbol{k}'}\tilde{\boldsymbol{I}}_{\mu\nu}(-\boldsymbol{k}')\cdot\tilde{\boldsymbol{A}}(\boldsymbol{k}',\omega)$$

Center of Taylor exp.

LWA → keep the first two terms of Taylor expansion

$$\tilde{\boldsymbol{I}}_{\mu\nu}(\boldsymbol{k}) = \frac{1}{V_{\mathrm{n}}}\int \mathrm{d}\boldsymbol{r}\exp[-i\boldsymbol{k}\cdot\boldsymbol{r}]\ \boldsymbol{I}_{\mu\nu}(\boldsymbol{r}) = \frac{\exp(-i\boldsymbol{k}\cdot\bar{\boldsymbol{r}})}{V_{\mathrm{n}}}\left(\bar{\boldsymbol{I}}_{\mu\nu} - i\boldsymbol{k}\cdot\bar{\boldsymbol{Q}}_{\mu\nu}\right)$$

$$\bar{\boldsymbol{I}}_{\mu\nu} = \int \mathrm{d}\boldsymbol{r}\ \boldsymbol{I}_{\mu\nu}(\boldsymbol{r})\ , \quad \bar{\boldsymbol{Q}}_{\mu\nu} = \int \mathrm{d}\boldsymbol{r}\ (\boldsymbol{r}-\bar{\boldsymbol{r}})\ \boldsymbol{I}_{\mu\nu}(\boldsymbol{r})$$

Substitute into the above eq. ★

$$\begin{aligned}\tilde{\boldsymbol{I}}(\boldsymbol{k},\omega) &= \frac{1}{V_n c}\sum_{\nu}\sum_{\boldsymbol{k}'} \mathrm{e}^{i(\boldsymbol{k}'-\boldsymbol{k})\cdot\bar{\boldsymbol{r}}}[\bar{g}_{\nu}(\omega)(\bar{\boldsymbol{I}}_{0\nu} - i\boldsymbol{k}\cdot\bar{\boldsymbol{Q}}_{0\nu})(\bar{\boldsymbol{I}}_{\nu 0} + i\boldsymbol{k}'\cdot\bar{\boldsymbol{Q}}_{\nu 0}) \\ &+ \bar{h}_{\nu}(\omega)(\bar{\boldsymbol{I}}_{\nu 0} - i\boldsymbol{k}\cdot\bar{\boldsymbol{Q}}_{\nu 0})(\bar{\boldsymbol{I}}_{0\nu} + i\boldsymbol{k}'\cdot\bar{\boldsymbol{Q}}_{0\nu})]\cdot\tilde{\boldsymbol{A}}(\boldsymbol{k}',\omega)\ .\end{aligned}$$

Keep the *k* = *k'* alone (Assume, averaged system is macrosc. uniform.)

LWA of constitutive eq.

$$\bar{\boldsymbol{I}}(\boldsymbol{k},\omega) = \bar{\chi}_{\rm em}\ \boldsymbol{A}(\boldsymbol{k},\omega)$$

New macrosc. susceptibility

$$\begin{aligned}\bar{\chi}_{\rm em}(\boldsymbol{k},\omega) &= \sum_\nu (n_\nu/c)\ [\bar{g}_\nu(\omega)(\bar{\boldsymbol{I}}_{0\nu} - i\boldsymbol{k}\cdot\boldsymbol{Q}_{0\nu})(\bar{\boldsymbol{I}}_{\nu 0} + i\boldsymbol{k}\cdot\boldsymbol{Q}_{\nu 0}) \\ &+ \bar{h}_\nu(\omega)(\bar{\boldsymbol{I}}_{0\nu} + i\boldsymbol{k}\cdot\boldsymbol{Q}_{0\nu})(\bar{\boldsymbol{I}}_{\nu 0} - i\boldsymbol{k}\cdot\boldsymbol{Q}_{\nu 0})]\ .\end{aligned}$$

$$\bar{\boldsymbol{I}}_{\mu\nu} = \int {\rm d}\boldsymbol{r}\ \boldsymbol{I}_{\mu\nu}(\boldsymbol{r})$$

El-dipole (E1)

$$\boldsymbol{Q}_{\mu\nu} = \int {\rm d}\boldsymbol{r}\ (\boldsymbol{r}-\bar{\boldsymbol{r}})\ \boldsymbol{I}_{\mu\nu}(\boldsymbol{r})$$

Mag-dipole (M1) + el-quadrupole

$$\bar{g}_\nu(\omega) = g_\nu(\omega) - \frac{1}{E_{\nu 0}}, \qquad \bar{h}_\nu(\omega) = h_\nu(\omega) - \frac{1}{E_{\nu 0}}.$$

Transition $(0 \rightarrow \nu)$ is (E1, M1)−active in general.

Terms of different k-dependence

$$\overline{\chi}_{\mathrm{em}} = \underline{\chi_{\mathrm{e1}}} + ik\ \underline{\chi_{\mathrm{chir}}} + k^2\ \underline{\chi_{\mathrm{m1}}}$$

$$\chi_{\mathrm{e1}} = \sum_\nu \frac{N_\nu}{c} \left[g_\nu(\omega) \bar{\boldsymbol{I}}_{0\nu} \bar{\boldsymbol{I}}_{\nu 0} + h_\nu(\omega) \bar{\boldsymbol{I}}_{\nu 0} \bar{\boldsymbol{I}}_{0\nu} \right]$$

$$\chi_{\mathrm{m1}} = \sum_\nu \frac{N_\nu}{c} \left[g_\nu(\omega) \hat{\boldsymbol{k}} \cdot \bar{\boldsymbol{Q}}_{0\nu} \hat{\boldsymbol{k}} \cdot \bar{\boldsymbol{Q}}_{\nu 0} + h_\nu(\omega) \hat{\boldsymbol{k}} \cdot \bar{\boldsymbol{Q}}_{\nu 0} \hat{\boldsymbol{k}} \cdot \bar{\boldsymbol{Q}}_{0\nu} \right]$$

$$\chi_{\mathrm{chir}} = \sum_\nu \frac{N_\nu}{c} \left[g_\nu(\omega) \{ \bar{\boldsymbol{I}}_{0\nu} \hat{\boldsymbol{k}} \cdot \bar{\boldsymbol{Q}}_{\nu 0} - \hat{\boldsymbol{k}} \cdot \bar{\boldsymbol{Q}}_{0\nu} \bar{\boldsymbol{I}}_{\nu 0} \} \right.$$

$$\left. + h_\nu(\omega) \{ \bar{\boldsymbol{I}}_{\nu 0} \hat{\boldsymbol{k}} \cdot \bar{\boldsymbol{Q}}_{0\nu} - \hat{\boldsymbol{k}} \cdot \bar{\boldsymbol{Q}}_{\nu 0} \bar{\boldsymbol{I}}_{0\nu} \} \right]$$

$$\hat{\boldsymbol{k}} = \boldsymbol{k} / |\boldsymbol{k}|$$ (unit vector along ***k***)

Poles of the three terms:

"common or different" depends on symmetry"

Dispersion equation

$$\frac{c^2k^2}{\omega^2} = 1 + \frac{4\pi c}{\omega^2}\,\bar{\chi}_{\rm em} \Longrightarrow \left(\frac{c}{\omega^2}\,\bar{\chi}_{\rm em} = \bar{\chi}_{\rm e} + \bar{\chi}_{\rm m} \,, \right)$$

in general — if E1 & M1 distinguishable

$$\bar{\chi}_{\rm e} = \frac{1}{\omega^2}\sum_{\nu\ \mathbf{E1}} N_\nu \left[g_\nu(\omega) \bar{\boldsymbol{I}}_{0\nu} \bar{\boldsymbol{I}}_{\nu 0} + h_\nu(\omega) \bar{\boldsymbol{I}}_{\nu 0} \bar{\boldsymbol{I}}_{0\nu} \right]$$

$$\bar{\chi}_{\rm m} = \frac{k^2}{\omega^2}\sum_{\nu\ \mathbf{M1}} N_\nu \left[g_\nu(\omega) (\hat{\boldsymbol{k}}\cdot\bar{\boldsymbol{Q}}_{0\nu})\ (\hat{\boldsymbol{k}}\cdot\bar{\boldsymbol{Q}}_{\nu 0}) + h_\nu(\omega) (\hat{\boldsymbol{k}}\cdot\bar{\boldsymbol{Q}}_{\nu 0})\ (\hat{\boldsymbol{k}}\cdot\bar{\boldsymbol{Q}}_{0\nu}) \right]$$

New disp. Eq. : $\frac{c^2k^2}{\omega^2} = 1 + 4\pi(\bar{\chi}_{\rm e} + \bar{\chi}_{\rm m})\,,$

Conv. Form : $\frac{c^2k^2}{\omega^2} = \epsilon\mu = (1 + 4\pi\chi_{\rm e})(1 + 4\pi\chi_{\rm m})$

Same ?

To calculate the susceptibilities via $\boldsymbol{P} = \chi_{\mathrm{e}} \boldsymbol{E}$, $\boldsymbol{M} = \chi_{\mathrm{m}} \boldsymbol{H}$

we need interaction Hamiltonian linear in ***E*** and ***H***.

We know that

"dipole approximation" gives

$$H_{\mathrm{int}} = -\boldsymbol{E} \cdot \int \boldsymbol{P}(\boldsymbol{r})\ \mathrm{d}\boldsymbol{r}$$

But no exact treatment is available in this direction.

It is worth to consider "Power-Zienau-Woolley" transformation:

"Photons & Atoms" (Cohen-Tannoudji et al)

$$L' = L + \frac{1}{c}\frac{d}{dt}\int \mathrm{d}\boldsymbol{r}\ \boldsymbol{P}(\boldsymbol{r}) \cdot \boldsymbol{A}(\boldsymbol{r}, t)$$

Then, interaction term in *L'* becomes

$$\frac{1}{c}\frac{d}{dt}\int \mathrm{d}\boldsymbol{r}\ \boldsymbol{P}(\boldsymbol{r}) \cdot \boldsymbol{A}(\boldsymbol{r}, t)\ + \frac{1}{c}\int \mathrm{d}\boldsymbol{r}\ \boldsymbol{J}(\boldsymbol{r}) \cdot \boldsymbol{A}(\boldsymbol{r}) = \int \mathrm{d}\boldsymbol{r}\ [\boldsymbol{M} \cdot \boldsymbol{B} + \boldsymbol{P} \cdot \boldsymbol{E}_{\mathrm{T}}]$$

Standard Lagrangian in Coulomb gauge:

$$L = \sum_{\ell} \frac{m_\ell \boldsymbol{v}_\ell^2}{2} - \frac{1}{2} \sum_{\ell} \sum_{\ell' \neq \ell} \frac{e_\ell e_{\ell'}}{|\boldsymbol{r}_\ell - \boldsymbol{r}_{\ell'}|} + \frac{1}{c} \int \mathrm{d}\boldsymbol{r}\ \boldsymbol{J}(\boldsymbol{r}) \cdot \boldsymbol{A}(\boldsymbol{r})$$
$$+ \frac{1}{8\pi} \int \mathrm{d}\boldsymbol{r}\ [(\frac{1}{c}\frac{\partial \boldsymbol{A}}{\partial t})^2 - (\nabla \times \boldsymbol{A})^2]$$

Conjugate momenta for $\boldsymbol{r}_\ell$ and $\boldsymbol{A}(\boldsymbol{r})$

$$\boldsymbol{p}_\ell = m_\ell \boldsymbol{v}_\ell + \frac{e_\ell}{c} \boldsymbol{A}(\boldsymbol{r}_\ell)$$
$$\boldsymbol{\Pi}(\boldsymbol{r}) = \frac{1}{4\pi c^2} \frac{\partial \boldsymbol{A}}{\partial t} = -\frac{1}{4\pi c} \boldsymbol{E}_\mathrm{T}$$

Hamiltonian

$$H_L = \sum_{\ell} \frac{1}{2m_\ell} [\boldsymbol{p}_\ell - \frac{e_\ell}{c} \boldsymbol{A}(\boldsymbol{r}_\ell)]^2 + \frac{1}{8\pi} \int \mathrm{d}\boldsymbol{r}\ (\boldsymbol{E}_\mathrm{T}^2 + \boldsymbol{B}^2) + \frac{1}{2} \sum_{\ell} \sum_{\ell' \neq \ell} \frac{e_\ell e_{\ell'}}{|\boldsymbol{r}_\ell - \boldsymbol{r}_{\ell'}|}$$

The new Lagrangian is

$$L' = \sum_\ell \frac{m_\ell \boldsymbol{v}_\ell^2}{2} - \frac{1}{2}\sum_\ell \sum_{\ell' \neq \ell} \frac{e_\ell e_{\ell'}}{|\boldsymbol{r}_\ell - \boldsymbol{r}_{\ell'}|} + \frac{1}{8\pi}\int \mathrm{d}\boldsymbol{r}\ (E_\mathrm{T}^2 - B^2)$$
$$+ \int \mathrm{d}\boldsymbol{r}\ [\boldsymbol{M}\cdot\boldsymbol{B} + \boldsymbol{P}\cdot\boldsymbol{E}_\mathrm{T}]$$

Conjugate momenta:

$$\begin{cases} \bar{\boldsymbol{p}}_\ell = m_\ell \boldsymbol{v}_\ell + \int_0^1 u\mathrm{d}u\ e_\ell\ \boldsymbol{B}(u\boldsymbol{r}_\ell)\times\boldsymbol{r}_\ell\ , \\ \bar{\boldsymbol{\Pi}} = -\frac{1}{4\pi c}(\boldsymbol{E}_\mathrm{T} + 4\pi\boldsymbol{P}_\mathrm{T}) = -\frac{1}{4\pi c}\boldsymbol{D}_\mathrm{T}\ . \end{cases}$$

The new Hamiltonian

$$H_{L'} = \sum_\ell \frac{\bar{\boldsymbol{p}}_\ell^2}{2m_\ell} + \frac{1}{2}\sum_\ell\sum_{\ell'\neq\ell}\frac{e_\ell e_{\ell'}}{|\boldsymbol{r}_\ell - \boldsymbol{r}_{\ell'}|} + 2\pi\int \mathrm{d}\boldsymbol{r}\boldsymbol{P}_\mathrm{T}(\boldsymbol{r})^2$$

matter

$$+\frac{1}{8\pi}\int \mathrm{d}\boldsymbol{r}(\boldsymbol{D}_\mathrm{T}^2 + \boldsymbol{B}^2) - \int \mathrm{d}\boldsymbol{r}\ [\boldsymbol{M}'\cdot\boldsymbol{B} + \boldsymbol{P}_\mathrm{T}\cdot\boldsymbol{D}_\mathrm{T}]$$

vacuum field

interaction

$$+\sum_\ell \frac{e_\ell^2}{2m_\ell}\left[\int_0^1 u\mathrm{d}u\ \ \boldsymbol{B}(u\boldsymbol{r}_\ell)\times\boldsymbol{r}_\ell\right]^2$$

interaction

$$\boldsymbol{M}' = \boldsymbol{M}\big|_{\boldsymbol{v}\to\boldsymbol{p}/m}$$

Both Hamiltonians are the same one

$$H = \sum_\ell \frac{m_\ell \boldsymbol{v}_\ell^2}{2} + \frac{1}{8\pi}\int \mathrm{d}\boldsymbol{r}\ (\boldsymbol{E}_\mathrm{T}^2 + \boldsymbol{B}^2) + \frac{1}{2}\sum_\ell \sum_{\ell' \neq \ell} \frac{e_\ell e_{\ell'}}{|\boldsymbol{r}_\ell - \boldsymbol{r}_{\ell'}|}$$

rewritten in terms of conjugate variables

$$H_L = H(\{\boldsymbol{r}_\ell, \boldsymbol{p}_\ell\}, \{\boldsymbol{A}, \boldsymbol{\Pi}\})$$

$$\boldsymbol{p}_\ell = m_\ell \boldsymbol{v}_\ell + \frac{e_\ell}{c}\boldsymbol{A}(\boldsymbol{r}_\ell) \quad \longleftrightarrow \quad \boldsymbol{r}_\ell$$

$$\boldsymbol{\Pi}(\boldsymbol{r}) = \frac{1}{4\pi c^2}\frac{\partial \boldsymbol{A}}{\partial t} = -\frac{1}{4\pi c}\boldsymbol{E}_\mathrm{T} \quad \longleftrightarrow \quad \boldsymbol{A}$$

$$H_{L'} = H(\{\boldsymbol{r}_\ell, \bar{\boldsymbol{p}}_\ell\}, \{\boldsymbol{A}, \bar{\boldsymbol{\Pi}}\})$$

$$\bar{\boldsymbol{p}}_\ell = m_\ell \boldsymbol{v}_\ell + \int_0^1 u \mathrm{d}u\ e_\ell\ \boldsymbol{B}(u\boldsymbol{r}_\ell) \times \boldsymbol{r}_\ell\ , \quad \longleftrightarrow \quad \boldsymbol{r}_\ell$$

$$\bar{\boldsymbol{\Pi}} = -\frac{1}{4\pi c}(\boldsymbol{E}_\mathrm{T} + 4\pi \boldsymbol{P}_\mathrm{T}) = -\frac{1}{4\pi c}\boldsymbol{D}_\mathrm{T}\ . \quad \longleftrightarrow \quad \boldsymbol{A}$$

Even if the interaction term is rewritten as $-\int \mathrm{d}\boldsymbol{r} \, [\boldsymbol{M}' \cdot \boldsymbol{B} + \boldsymbol{P}_\mathrm{T} \cdot \boldsymbol{D}_\mathrm{T}]$,

induced polarizations should be ***P***(***D, B***) and ***M***(***B, D***) , in general.

P & ***D*** are polar vectors and ***M*** & ***B*** are axial vectors, so that they are distinguishable in the presence of inversion symmetry. They are mixed in the absence of inversion symmetry (chiral symmetry).

In chiral symmetry, ***P*** = ***P***(***D***, ***B***) and **M** = **M**(**B**, ***D***). This does not fit the usual definition of electric and magnetic susceptibilties.

Only in the absence of chirality, ***P***(***D***) and ***M***(***B***). However, note that the matter Hamiltonian in this case contains an additional term

$2\pi \int \mathrm{d}\boldsymbol{r} \boldsymbol{P}_\mathrm{T}(\boldsymbol{r})^2$. This changes the excitation energies of E1 transitions, but the magnetic excitations will not be affected by this term. Thus, ***M***(***B***) can be defined with the same matter Hamiltonian.
This allows us to define a new magnetic susceptibility χ_B via

$$M = \chi_\mathrm{B} B$$

Equivalence of the dispersion eqs. (in the absence of chirality)

$$\boldsymbol{M} = \chi_\mathrm{B} \boldsymbol{B} \quad \Rightarrow \quad \mu = \frac{1}{1 - 4\pi\chi_\mathrm{B}}$$

Correct mag. Susceptibility;
Poles at mag. excitations

Then,

$$\frac{c^2 k^2}{\omega^2} = \epsilon\mu = \frac{1 + 4\pi\chi_\mathrm{e}}{1 - 4\pi\chi_\mathrm{B}}$$

$$\Rightarrow \quad \frac{c^2 k^2}{\omega^2} = 1 + 4\pi\chi_\mathrm{e} + 4\pi\frac{c^2 k^2}{\omega^2}\chi_\mathrm{B}$$

r.h.s. = sum of E1 and M1 transitions,

as in the new result

$$\frac{c^2 k^2}{\omega^2} = 1 + 4\pi(\bar{\chi}_\mathrm{e} + \bar{\chi}_\mathrm{m})$$

(non-chiral case)

Mag. tr. energies and poles of susceptibility

Important for
Left-handed systems !

conventional

$$\text{(A)} \quad \chi_{\mathrm{m}} = \frac{b}{\omega_0 - \omega - i0^+}$$

$$\text{(A)} \quad \frac{c^2 k^2}{\omega^2} = \epsilon_{\mathrm{b}} \left(1 + \frac{4\pi b}{\omega_0 - \omega - i0^+}\right)$$

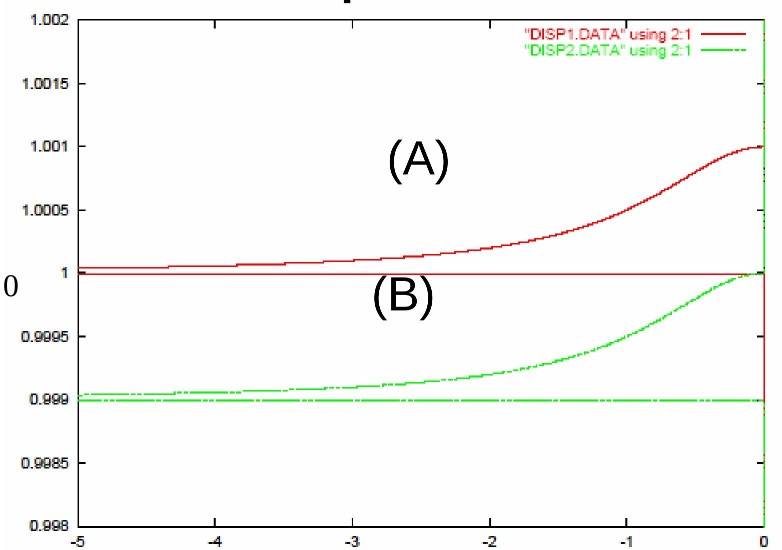
New theory

$$\text{(B)} \quad \chi_{\mathrm{B}} = \frac{b}{\omega_0 - \omega - i0^+}$$

$$\text{(B)} \quad \frac{c^2 k^2}{\omega^2} = \epsilon_{\mathrm{b}} \left(1 - \frac{4\pi b}{\omega_0 - \omega - i0^+}\right)^{-1}$$

$\varepsilon_b < 0$

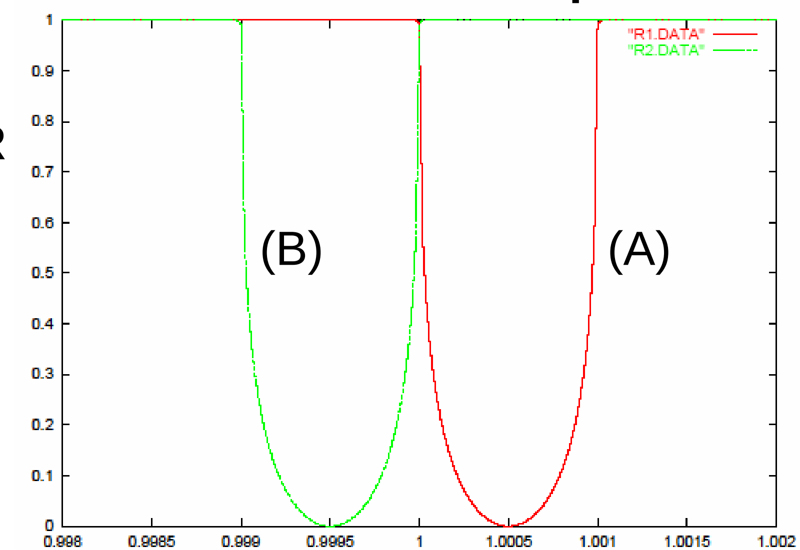
Dispersion curve



ω / ω_0

$-\ ck / \omega$

Reflectance spectrum



R

ω / ω_0

Exp. check is feasible !

Case of chiral symmetry (mixing of E1 and M1 transitions)

"Drude-Born-Fedorov (DBF)" constitutive eqs.

$$\boldsymbol{D} = \epsilon(\boldsymbol{E} + \beta\nabla\times\boldsymbol{E})$$
$$\boldsymbol{B} = \mu(\boldsymbol{H} + \beta\nabla\times\boldsymbol{H})$$

(Uniform & isotropic case)

β (chiral admittance)

Dispersion eq. : $$(\frac{ck}{\omega})^2 = \epsilon\mu\ (1 \pm \frac{\beta\omega}{c}\sqrt{\epsilon\mu})^{-2}$$

2nd order poles

essentially different from $$\frac{c^2k^2}{\omega^2} = 1 + \frac{4\pi c}{\omega^2}\bar{\chi}_{\rm em}$$

"DBF eqs" is a phenomenology, incompatible with the new macrosc. M-eq, especially near resonance.

For parametrization, one needs care about:

off-resonant cases

χ_{e1} , χ_{m1} , χ_{chir} : arbitrary parameters

resonant case

1. Chiral symmetry

   χ_{e1} , χ_{m1} , χ_{chir} have common poles

2. non-chiral symmetry

   $\chi_{\mathrm{chir}} = 0$

   χ_{e1} , χ_{m1} have different poles

Conclusion

1. New macrosc. M-eq. has the same form as the microscopic M-eq., and requires only one susceptibility tensor, $\chi_{\rm em}$

2. $\chi_{\rm em}$ contains all the effects of electric and magnetic polarizations together with their interference.

3. Dispersion eq. is $(ck)^2/\omega^2 = 1 + (4\pi c/\omega^2)\chi_{\rm em}$

4. In the absence of chiral symmetry, this result can be reduced to the conventional macrosc. M-eqs.

5. DBF eqs for chiral symmetry remains a phenomenology, not justified by the first-principles theory.
   Chiral symmetry should be treated by the new scheme.

6. Non-uniqueness problem does not arise, since we do not separate ***J*** into the contributions of ***P*** and ***M***.
7. Magnetic susceptibility should be, not χ_{m}, but χ_{B}. An experiment is proposed to check it.
8. χ_{e} and χ_{B} are two susceptibilities derived from a single microsc. susceptibility $\chi(\boldsymbol{r}, \boldsymbol{r}', \omega)$ in the case of non-chiral symmetry.
9. Better defintion of left-handed systems is $v_{\mathrm{ph}} \times v_{\mathrm{g}} < 0$. If one uses ϵ and μ, system must have non-chiral symmetry, and magnetic excitation energies should correspond to the zeros of magnetic permeability.

Rewriting $\langle 0|\hat{N}(\boldsymbol{r})|0\rangle$ term

In this appendix, we will show that the following relation

$$\langle 0|\hat{N}(\boldsymbol{r})|0\rangle\ \boldsymbol{A}(\boldsymbol{r}) = \sum_{\nu} \frac{1}{E_{\nu 0}}[\boldsymbol{I}_{0\nu}(\boldsymbol{r})F_{\nu 0}(\omega) + \boldsymbol{I}_{\nu 0}(\boldsymbol{r})F_{0\nu}(\omega)] \tag{1}$$

holds as a good approximation, when [a] the relativistic correction in $H^{(0)}$ is negligible in comparison with the main term, and [b] LWA is valid. This expression allows us to rewrite the microscopic susceptibility $\chi_{\rm cd}$ into a compact form (**??**). Though an essentially same argument is given in [**?**], we reproduce it here with some more details.

The relevant term appears as a part of induced current density arising from the $\boldsymbol{A}$ dependent term of the current density operator

$$\frac{1}{c}\ \hat{N}(\boldsymbol{r})\ \boldsymbol{A}(\boldsymbol{r})\ , \tag{2}$$

where

$$\hat{N}(\boldsymbol{r}) = \sum_{\ell} \frac{e_\ell^2}{m_\ell}\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ . \tag{3}$$

The operator $\boldsymbol{I}(\boldsymbol{r})$ is the $\boldsymbol{A}$-independent part of the current density operator,

$$\boldsymbol{I}(\boldsymbol{r}) = \sum_{\ell} \frac{e_\ell}{2m_\ell}[\boldsymbol{p}_\ell \delta(\boldsymbol{r}-\boldsymbol{r}_\ell) + \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\boldsymbol{p}_\ell]\ . \tag{4}$$

The spin dependent terms are neglected, since the relativistic correction is assumed to be small.

We introduce one more operator

$$\begin{aligned}\hat{\boldsymbol{R}}(\boldsymbol{r}) &= \sum_{\ell} e_\ell \boldsymbol{r}_\ell \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ , &(5)\\ &= \boldsymbol{r} \sum_{\ell}\ e_\ell\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ . &(6)\end{aligned}$$

Now we evaluate the commutators $[\hat{R}_\xi, H^{(0)}]$ and $[\hat{R}_\xi(\boldsymbol{r}), \hat{I}_\eta(\boldsymbol{r}')]$, where ξ, η are Cartesian coordinates. We begin with

$$[\hat{R}_\xi(\boldsymbol{r}), H^{(0)}] = r_\xi \sum_{\ell} \frac{e_\ell}{2m_\ell}[\delta(\boldsymbol{r}-\boldsymbol{r}_\ell), \boldsymbol{p}_\ell^2] \tag{7}$$

where the relativistic correction terms are neglected in $H^{(0)}$. For the evaluation of the commutators we use the relation

$$\boldsymbol{p}_\ell\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell) = -\boldsymbol{p}\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ , \tag{8}$$

which allows us to move $\boldsymbol{p}$ to the outside of the summation over ℓ.

The commutator in (7) is expanded as

$$\begin{aligned}&[\delta(\boldsymbol{r}-\boldsymbol{r}_\ell), \boldsymbol{p}_\ell^2] = \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\boldsymbol{p}_\ell^2 - \boldsymbol{p}_\ell^2\delta(\boldsymbol{r}-\boldsymbol{r}_\ell) &(9)\\ &= \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\boldsymbol{p}_\ell^2 - \boldsymbol{p}_\ell\cdot\{\boldsymbol{p}_\ell\delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\} - \boldsymbol{p}_\ell\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\cdot\boldsymbol{p}_\ell &(10)\\ &= \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\boldsymbol{p}_\ell^2 + \boldsymbol{p}_\ell\cdot\{\boldsymbol{p}\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\} - \{\boldsymbol{p}_\ell\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\}\cdot\boldsymbol{p}_\ell - \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ \boldsymbol{p}_\ell^2 &(11)\\ &= \boldsymbol{p}\cdot\{\boldsymbol{p}_\ell\delta(\boldsymbol{r}-\boldsymbol{r}_\ell) + \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ \boldsymbol{p}_\ell\} &(12)\end{aligned}$$

where (8) is used twice. Substituting this result into (7), we obtain

$$[\hat{R}_\xi(\boldsymbol{r}), H^{(0)}] = r_\xi \boldsymbol{p}\cdot \boldsymbol{I}(\boldsymbol{r}) \ . \tag{13}$$

Another commutator $[\hat{R}_\xi(\boldsymbol{r}), \hat{I}_\eta(\boldsymbol{r}')]$ is evaluated as

$$[\quad \hat{R}_\xi(\boldsymbol{r}), \hat{I}_\eta(\boldsymbol{r}')] = r_\xi \sum_\ell \frac{e_\ell^2}{2m_\ell}[\delta(\boldsymbol{r}-\boldsymbol{r}_\ell), p_{\ell\eta}\ \delta(\boldsymbol{r}'-\boldsymbol{r}_\ell) + \delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\ p_{\ell\eta}] \tag{14}$$

$$\begin{aligned} = \quad & r_\xi \sum_\ell \frac{e_\ell^2}{2m_\ell}\{\delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ p_{\ell\eta}\ \delta(\boldsymbol{r}'-\boldsymbol{r}_\ell) - p_{\ell\eta}\ \delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell) \\ & +\delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ \delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\ p_{\ell\eta} - \delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\ p_{\ell\eta}\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\} \end{aligned} \tag{15}$$

$$\begin{aligned} = \quad & r_\xi \sum_\ell \frac{e_\ell^2}{2m_\ell}[\delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ \{p_{\ell\eta}\ \delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\} + \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ \delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\ p_{\ell\eta} \\ & -\{p_{\ell\eta}\ \delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\}\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell) - \delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\{p_{\ell\eta}\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\} \\ & -\delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ p_{\ell\eta} + \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ \delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\ \boldsymbol{p}_{\ell,\eta} \\ & -\delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\{p_{\ell\eta}\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\} - \delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ p_{\ell\eta}] \end{aligned} \tag{16}$$

$$\begin{aligned} = \quad & r_\xi \sum_\ell \frac{e_\ell^2}{2m_\ell}[-p'_\eta\delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\ \delta(\boldsymbol{r}'-\boldsymbol{r}_\ell) + p'_\eta\delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell) \\ & +p_\eta\delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell) + p_\eta\delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)] \end{aligned} \tag{17}$$

$$= \quad r_\xi\ p_\eta\ \sum_\ell \frac{e_\ell^2}{m_\ell}\delta(\boldsymbol{r}'-\boldsymbol{r}_\ell)\ \delta(\boldsymbol{r}-\boldsymbol{r}_\ell) \tag{18}$$

$$= \quad r_\xi\{p_\eta\delta(\boldsymbol{r}-\boldsymbol{r}')\}\hat{N}(\boldsymbol{r}') \tag{19}$$

Let us define two operators

$$\hat{Q}(\omega) \quad = \quad \int \mathrm{d}\boldsymbol{r}\ \hat{\boldsymbol{R}}(\boldsymbol{r})\cdot \boldsymbol{A}(\boldsymbol{r},\omega) \tag{20}$$

$$\hat{F}(\omega) \quad = \quad \int \mathrm{d}\boldsymbol{r}\ \hat{\boldsymbol{I}}(\boldsymbol{r})\cdot \boldsymbol{A}(\boldsymbol{r},\omega)\ , \tag{21}$$

in terms of which eq.(13) and eq.(19) are rewritten as

$$[\hat{Q}(\omega), H^{(0)}] \quad = \quad -i\hbar\int \mathrm{d}\boldsymbol{r}\ \boldsymbol{r}\cdot\boldsymbol{A}(\boldsymbol{r},\omega)\nabla\cdot\hat{\boldsymbol{I}}(\boldsymbol{r}) \tag{22}$$

$$[\hat{Q}(\omega), \hat{I}_\eta(\boldsymbol{r}')] \quad = \quad -i\hbar\ \int \mathrm{d}\boldsymbol{r}\ \boldsymbol{r}\cdot\boldsymbol{A}(\boldsymbol{r},\omega)\ [\frac{\partial}{\partial r_\eta}\delta(\boldsymbol{r}-\boldsymbol{r}')]\hat{N}(\boldsymbol{r}')\ . \tag{23}$$

These two integrals can be rewritten via partial integration into

$$[\hat{Q}(\omega), H^{(0)}] \quad = \quad i\hbar\int \mathrm{d}\boldsymbol{r}\ \nabla\{\boldsymbol{r}\cdot\boldsymbol{A}(\boldsymbol{r},\omega)\}\ \cdot\hat{\boldsymbol{I}}(\boldsymbol{r}) \tag{24}$$

$$[\hat{Q}(\omega), \hat{I}_\eta(\boldsymbol{r}')] \quad = \quad i\hbar\ \int \mathrm{d}\boldsymbol{r}\ \frac{\partial}{\partial r_\eta}\{\boldsymbol{r}\cdot\boldsymbol{A}(\boldsymbol{r},\omega)\}\delta(\boldsymbol{r}-\boldsymbol{r}')\hat{N}(\boldsymbol{r}')\ . \tag{25}$$

Both of them contain the following factor in the integrand

$$\frac{\partial}{\partial r_\eta}\{\boldsymbol{r}\cdot\boldsymbol{A}(\boldsymbol{r},\omega)\} = A_\eta + \sum_\xi r_\xi \frac{\partial A_\xi}{\partial r_\eta}\ , \tag{26}$$

which can be approximated as $A_\eta(\boldsymbol{r},\omega)$ when LWA is a good approximation. In this case, these two commutators can be written as

$$[\hat{Q}(\omega), H^{(0)}] = i\hbar \int \mathrm{d}\boldsymbol{r}\ \boldsymbol{A}(\boldsymbol{r},\omega)\cdot\hat{\boldsymbol{I}}(\boldsymbol{r}) \tag{27}$$

$$[\hat{Q}(\omega), \hat{\boldsymbol{I}}(\boldsymbol{r}')] = i\hbar\ \boldsymbol{A}(\boldsymbol{r}',\omega)\ \hat{N}(\boldsymbol{r}')\ . \tag{28}$$

Our object of this appendix, eq.(1), is the $\langle 0|\cdots|0\rangle$ matrix element of eq.(28). Thus,

$$\langle 0|\hat{N}(\boldsymbol{r})|0\rangle\ \boldsymbol{A}(\boldsymbol{r},\omega) = \frac{-i}{\hbar}\sum_\nu [\langle 0|[\hat{Q}(\omega)|\nu\rangle\langle\nu|\hat{\boldsymbol{I}}(\boldsymbol{r})|0\rangle - \langle 0|[\hat{\boldsymbol{I}}(\boldsymbol{r})|\nu\rangle\langle\nu|\hat{Q}(\omega)|0\rangle]\ . \tag{29}$$

To evaluate $\langle\nu|\hat{Q}(\omega)|\mu\rangle$, we take the $\langle\nu|\cdots|\mu\rangle$ matrix element of eq.(27) as

$$(E_\mu - E_\nu)\ \langle\nu|\hat{Q}|\mu\rangle = i\hbar F_{\nu\mu} \tag{30}$$

Thus, we obtain the desired result

$$\langle 0|\hat{N}(\boldsymbol{r})|0\rangle\ \boldsymbol{A}(\boldsymbol{r},\omega) = \sum_\nu \frac{1}{E_{\nu 0}}\ [F_{0\nu}(\omega)\boldsymbol{I}_{\nu 0}(\boldsymbol{r}) + F_{\nu 0}(\omega)\boldsymbol{I}_{0\nu}(\boldsymbol{r})]\ , \tag{31}$$

with $E_{\nu 0} = E_\nu - E_0$.