Optical properties of turbid colloids: An effective-medium approach

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In collaboration with:

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… and interesting discussions with:

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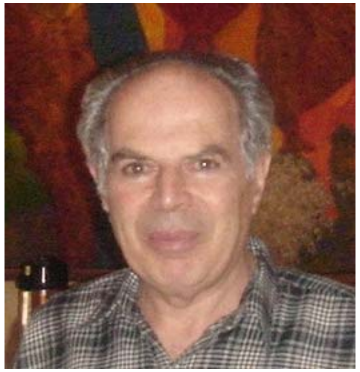


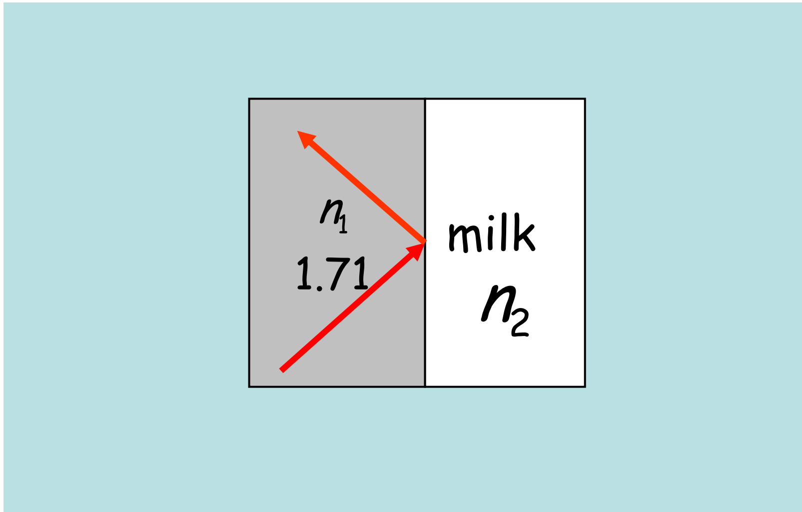
Peter Halevi

Motivation

refractive index of milk

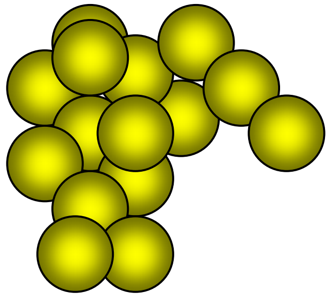
critical angle

$$\sin\theta_c = \frac{n_2}{n_1}$$



δn_2 → states of aggregation

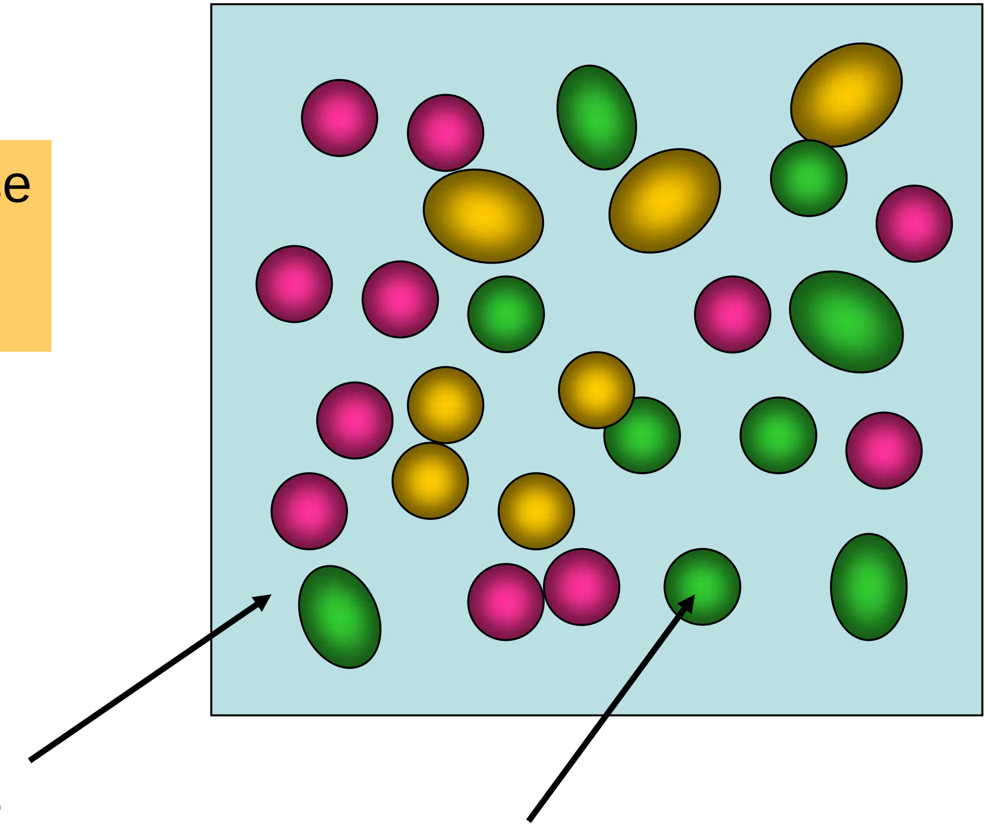
Real time



…but is white…and turbid…

Colloid

Inhomogeneous phase dispersed within a homogeneous one



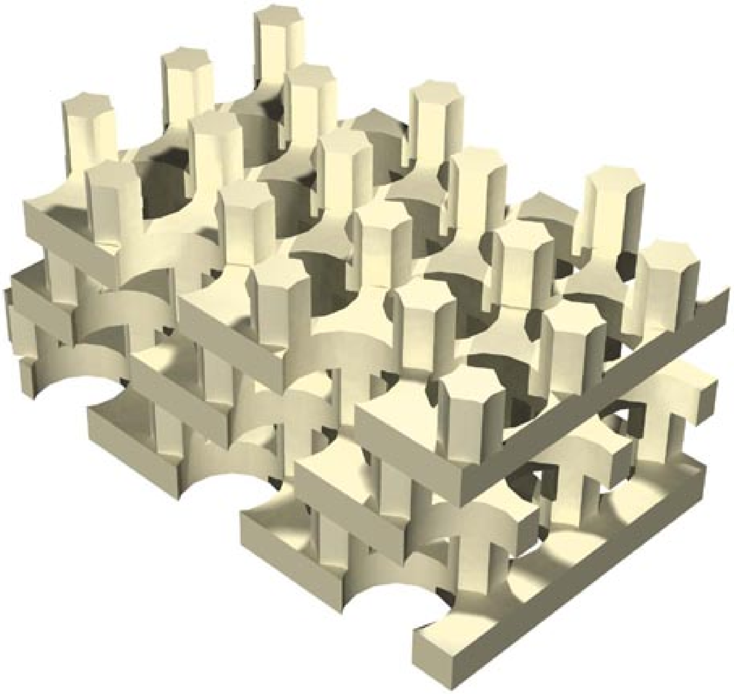
homogeneous phase

colloidal particles

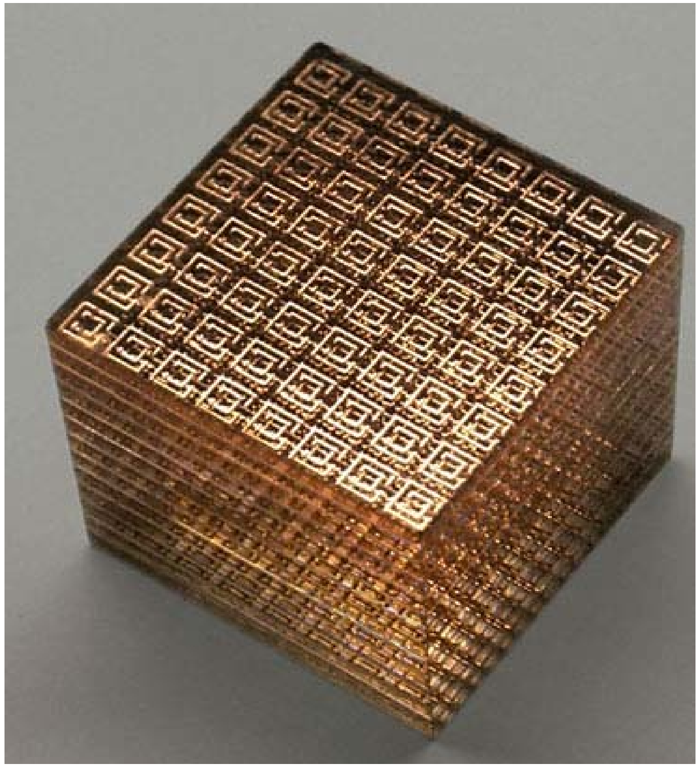
Colloidal systems

continuous phase	disperse phase	name	examples
liquid	solid	sol	Milk, paints, blood, tissues
liquid	liquid	emulsion	milk, water in benzene
liquid	gas	foam	foam, whipped cream
solid	solid	solid sol	composites, policrystals, rubys
solid	liquid	solid emulsion	milky quatz, opals
solid	gas	solid foam	porous media
gas	solid	solid aereosol	smoke, powder
gas	liquid	liquid aereosol	fog

"Ordered" colloids



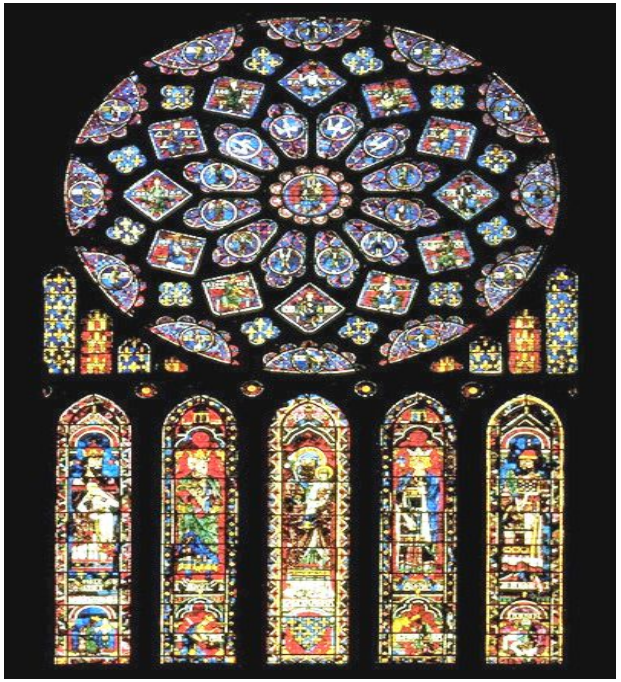
Photonic crystals



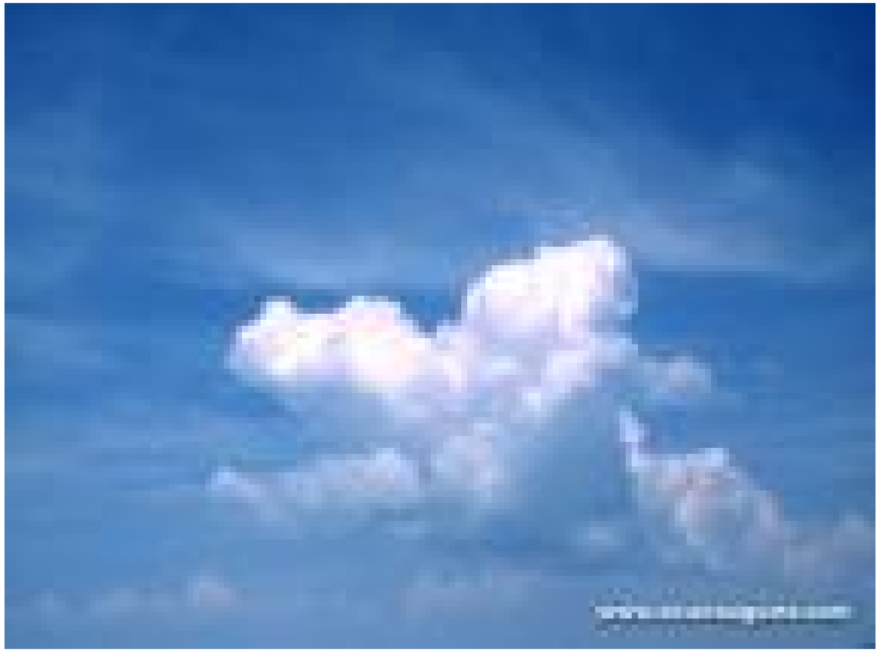
Metamaterials

Optical properties

Propagation of light



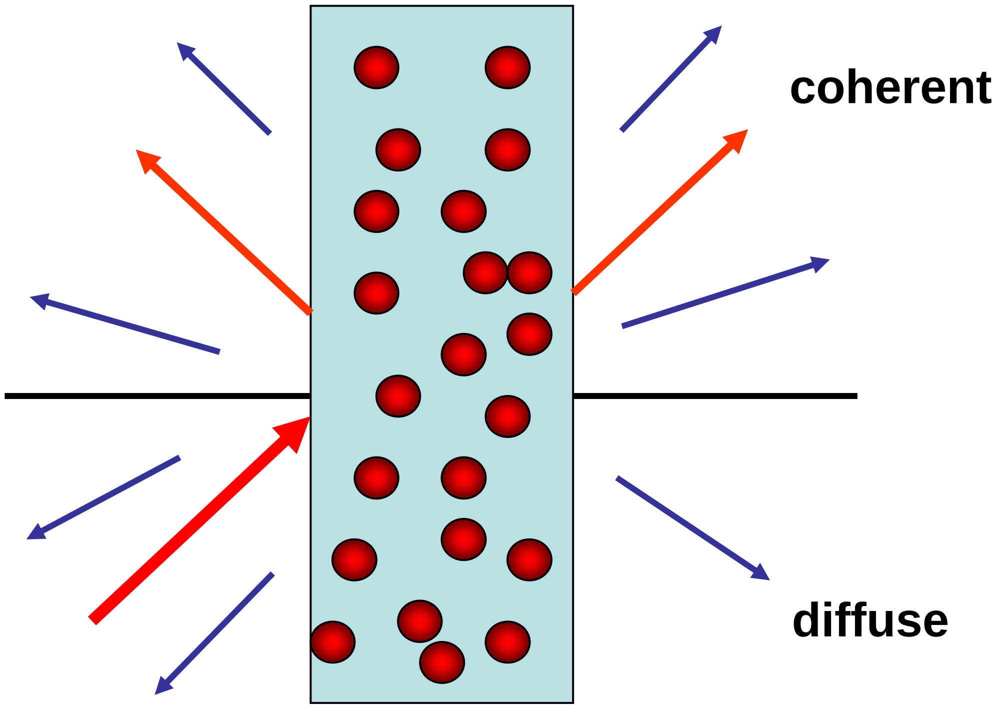
transmission



reflection

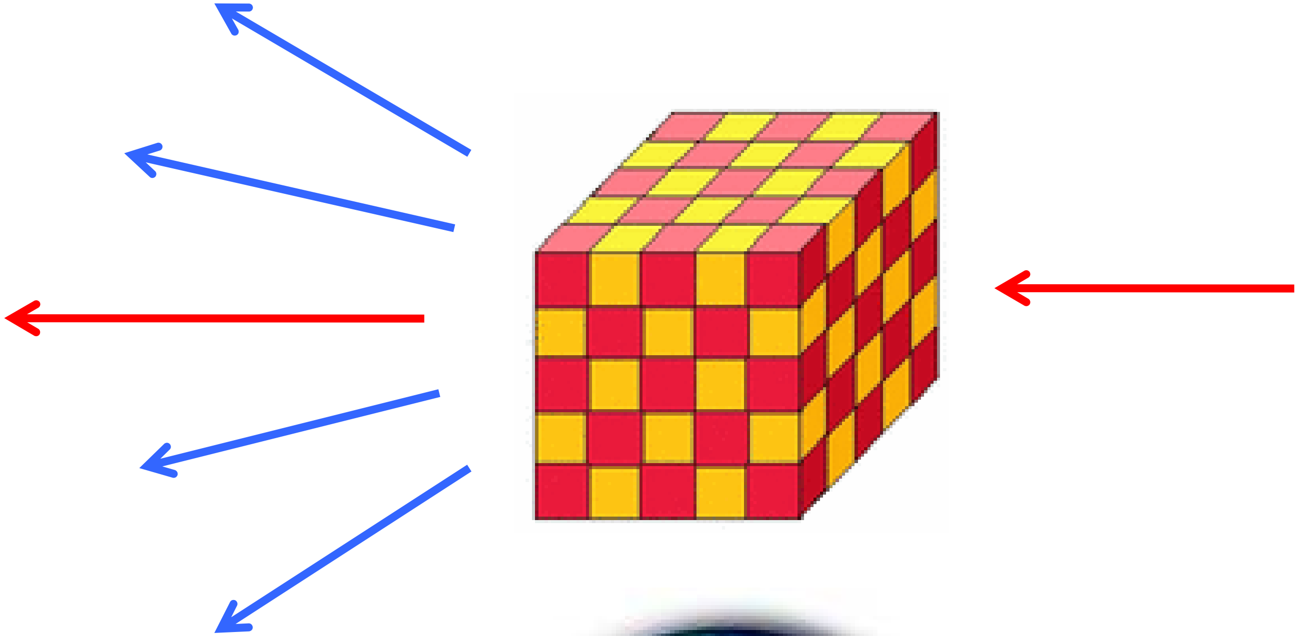
Light scattering

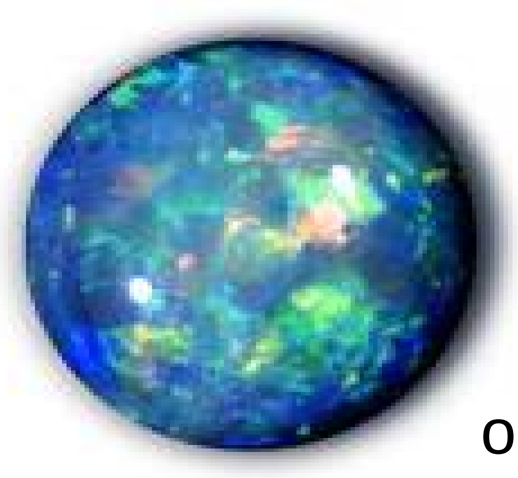
$400 \leq \lambda \leq 800$ nm



turbidity

Light diffraction





opal

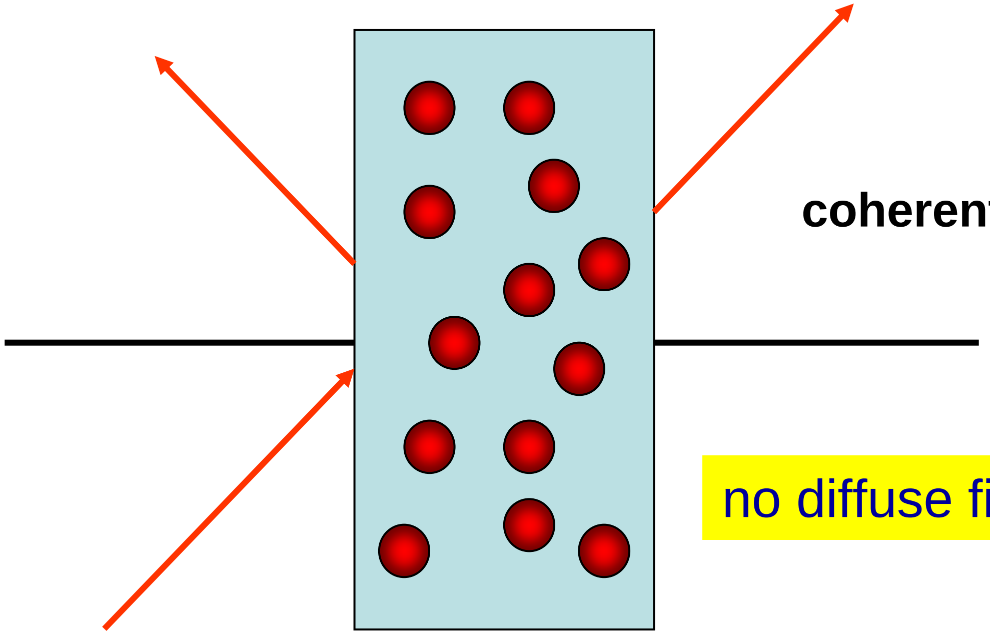
Small colloidal particles

size parameter

$$ka = \frac{2\pi a}{\lambda} \ll 1$$

$$a \ll \frac{\lambda}{2\pi} \approx 0.1\,\mu m$$

scattering can be neglected

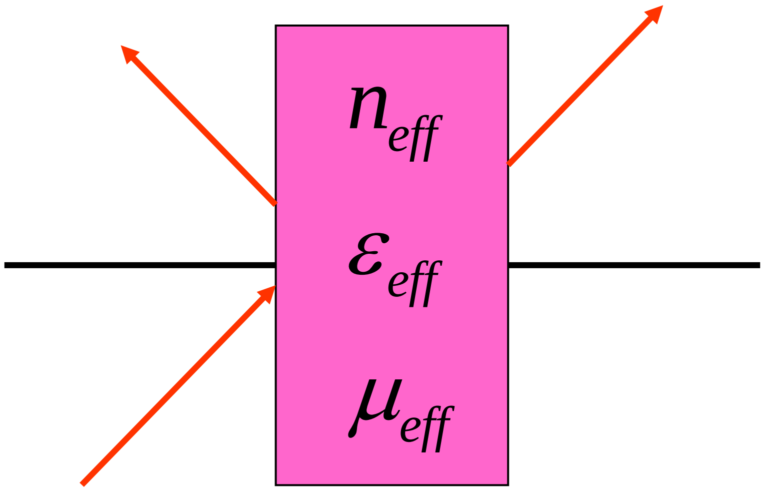


i.e. macroscopic electrodynamics

macroscopic field

Effective medium

effective medium



effective properties

$$n_{eff}\left[\{optical\},\{structural\}\right]$$

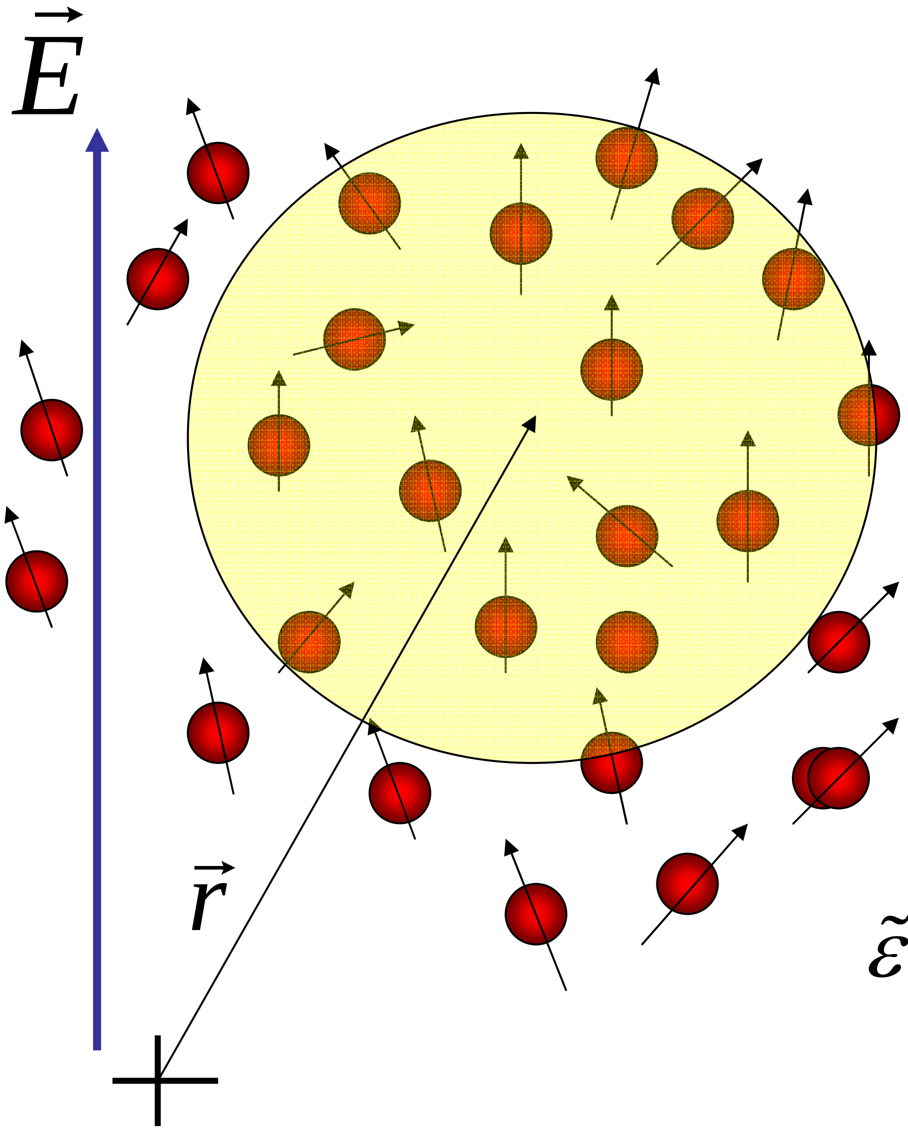
↓

Effective-medium theories

↓ *"unrestricted"*

Continuum Electrodynamics

Averaging procedure



spatial average

$$\lambda \gg a \qquad R \gg a$$

$$\left\langle \vec{P} \right\rangle = \varepsilon_0 \chi_{eff} \left\langle \vec{E} \right\rangle$$

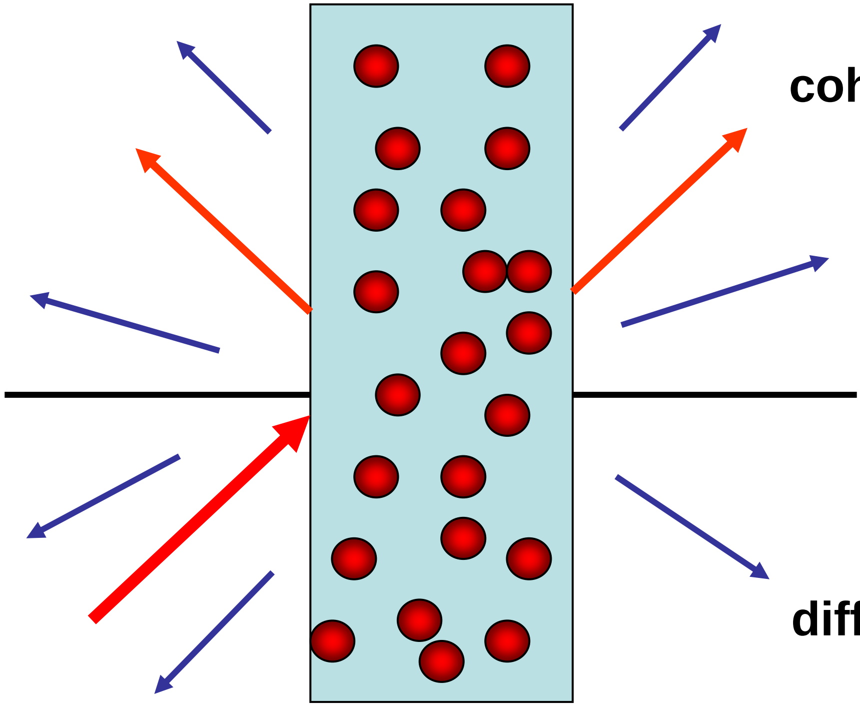
i.e.

$$\tilde{\varepsilon}_{eff} = 1 + \chi_{eff} = \frac{1 + 2f\tilde{\alpha}}{1 - f\tilde{\alpha}}$$

Turbid colloids

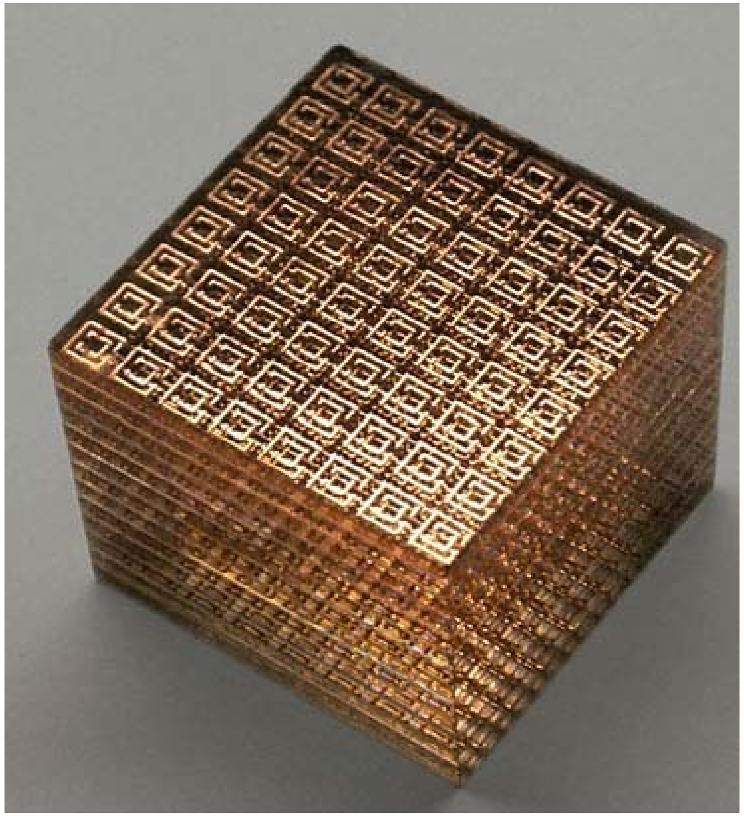
$$ka = \frac{2\pi a}{\lambda} \approx 1$$

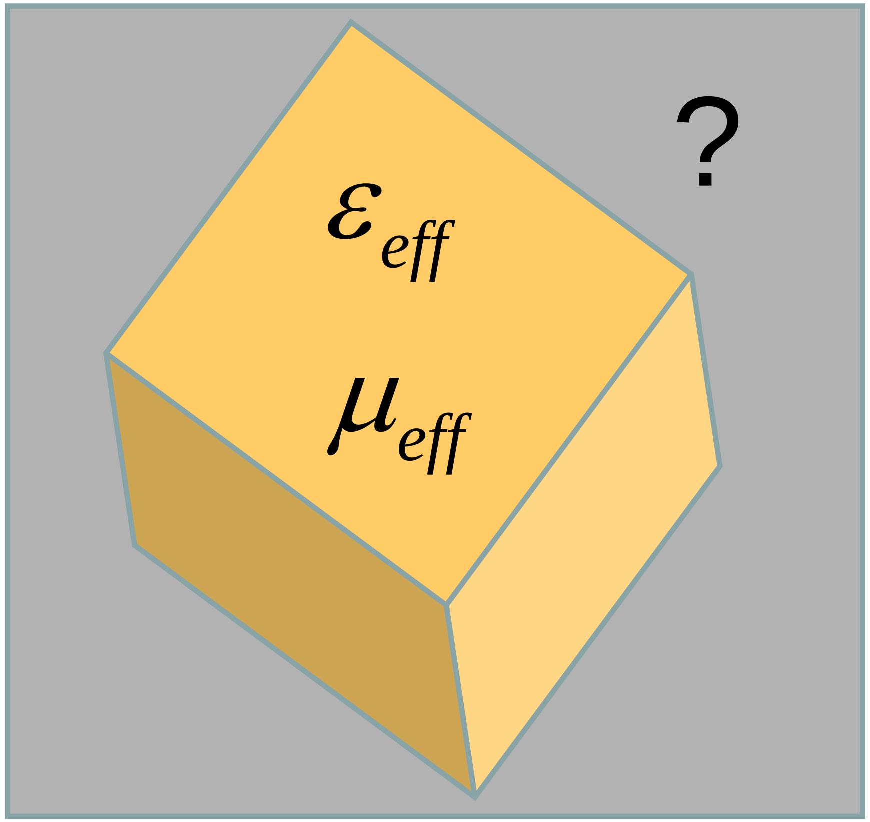
Is it possible to define an effective medium?



Is there an effective index of refraction?

Homogenization





$$\lambda \sim 10a$$

$$n_{eff} = \sqrt{\varepsilon_{eff}\,\mu_{eff}}$$

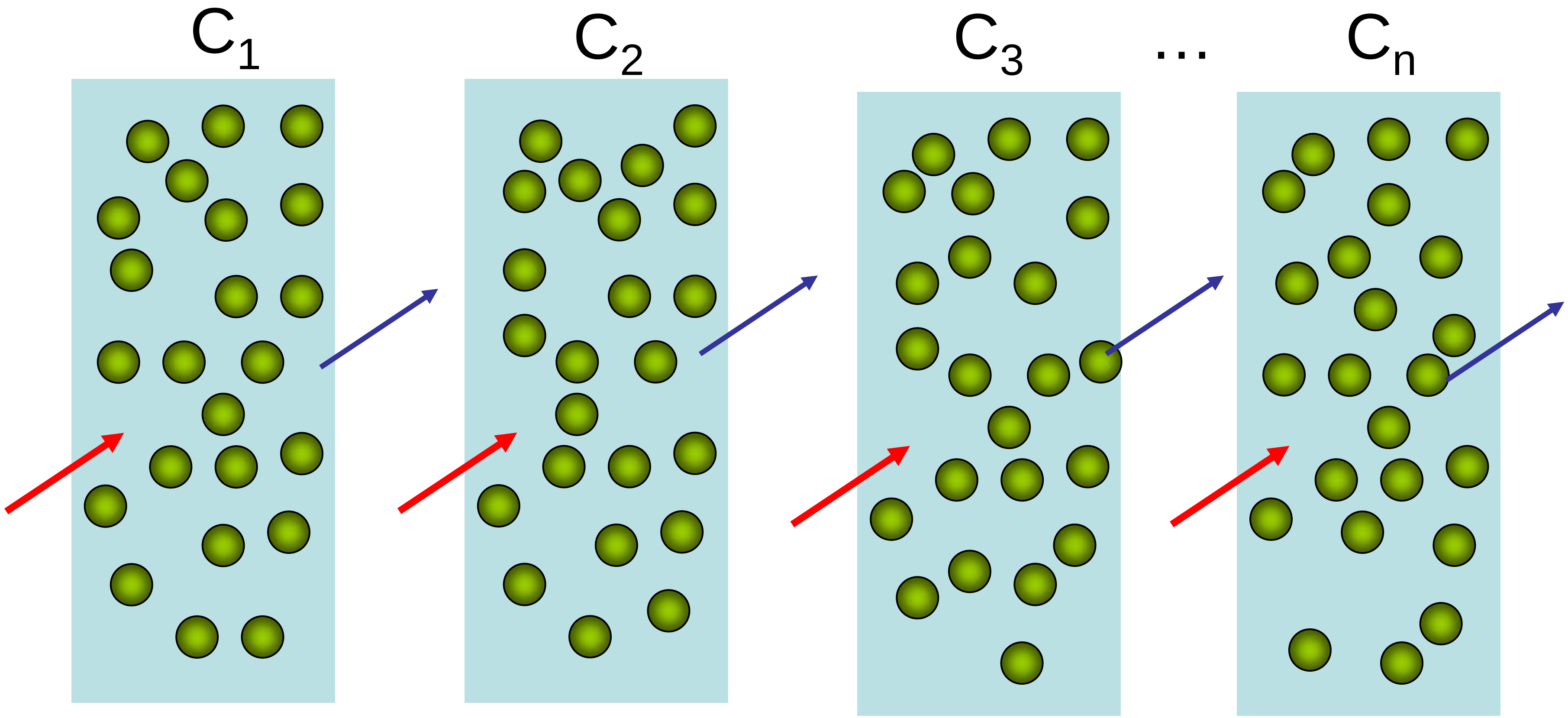
Effective medium for turbid colloids

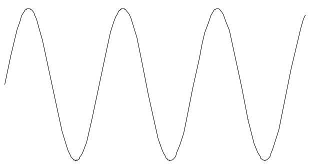
Coherent beam

$$\left\langle \vec{E} \right\rangle = \frac{1}{N} \sum_{\{C_n\}} \vec{E}\left(\vec{r} ; \left\{ \vec{R}_1 , \vec{R}_2 , \ldots \vec{R}_N \right\}_{C_n} \right)$$

configurational average

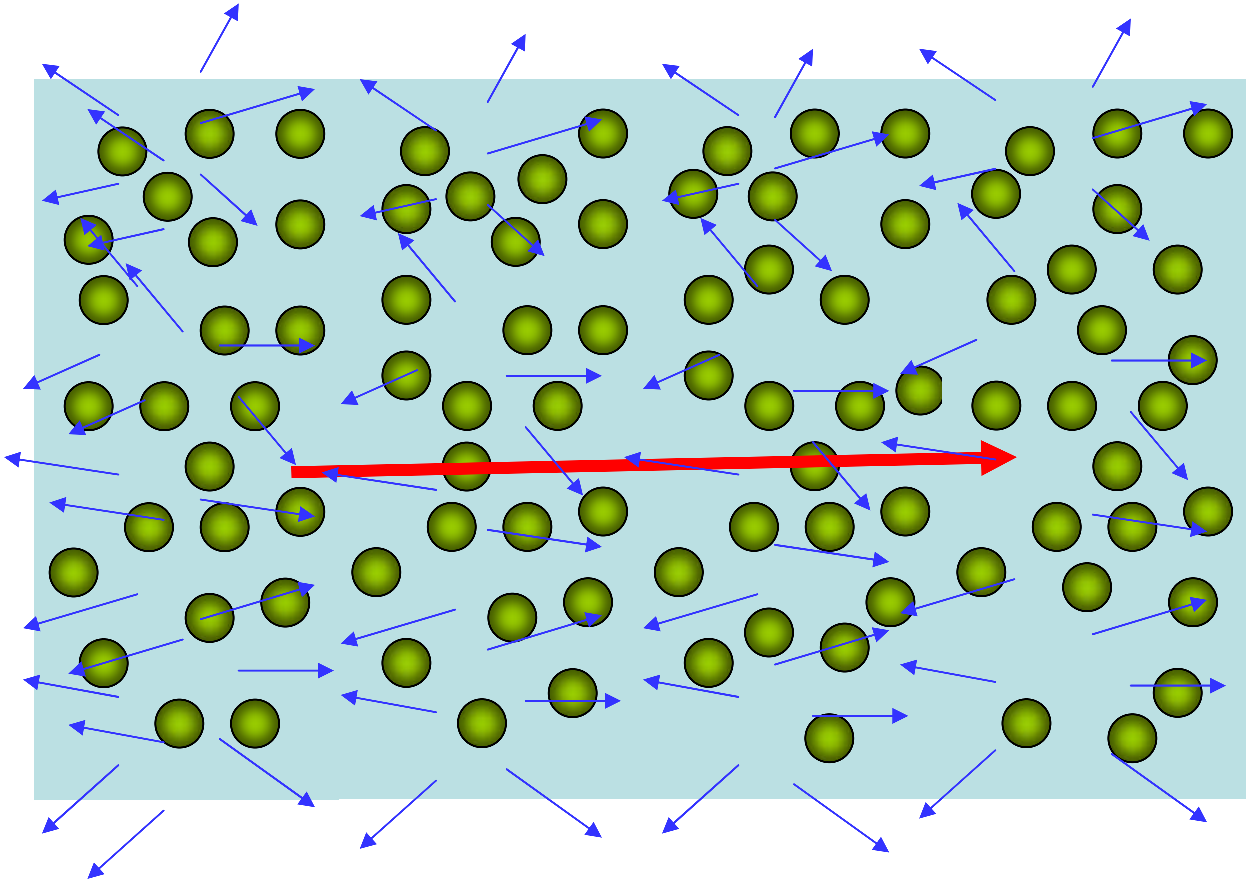
Coherent beam

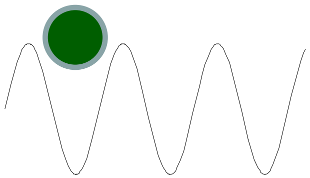




$$\left\langle \vec{E} \right\rangle = \frac{1}{N} \sum_{\{C_n\}} \vec{E}\left(\vec{r}; \left\{ \vec{R}_1, \vec{R}_2, \ldots \vec{R}_N \right\}_{C_n} \right)$$

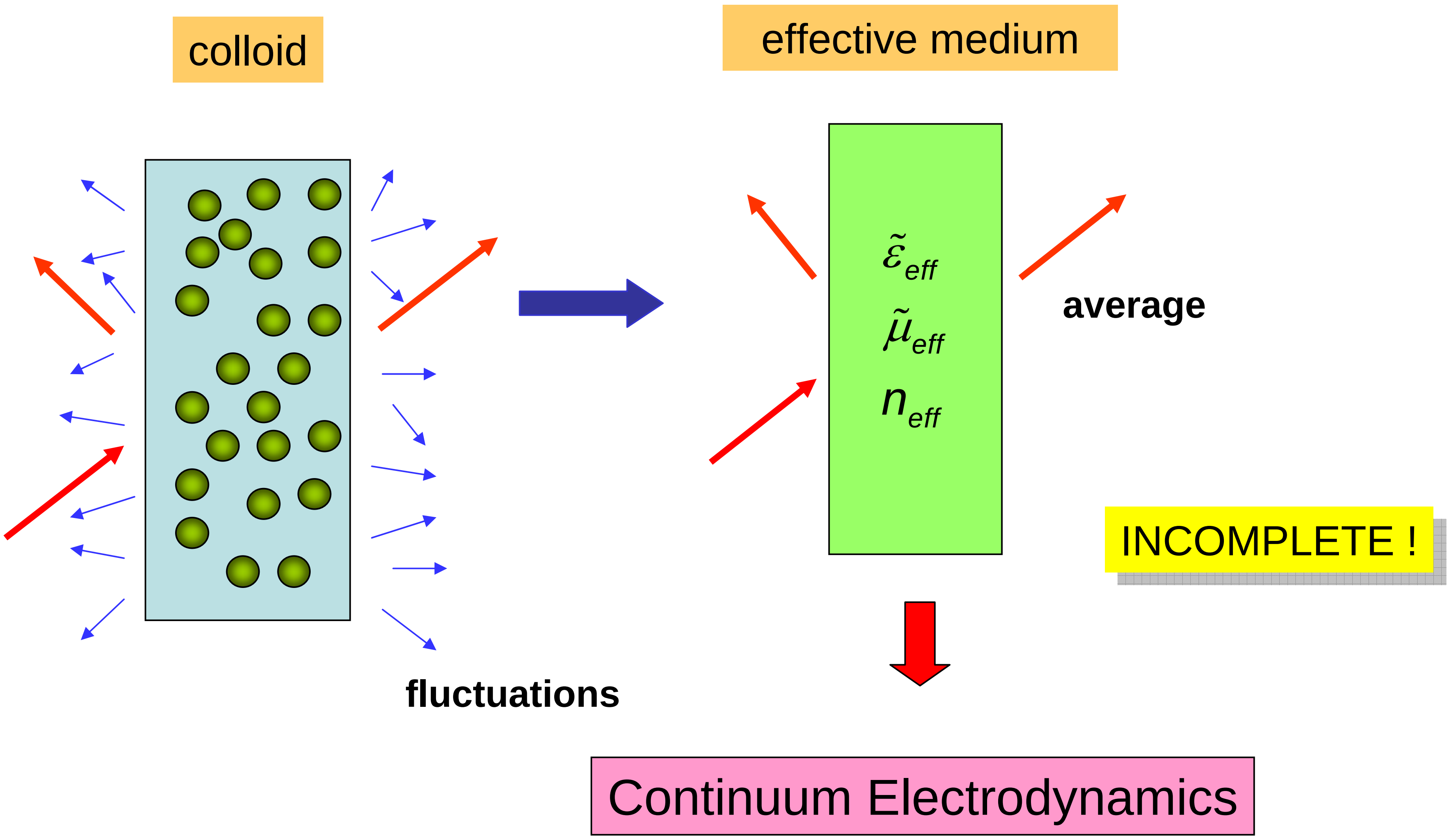
Total field




λ_0

$$\vec{E} = \langle \vec{E} \rangle + \delta \vec{E}$$

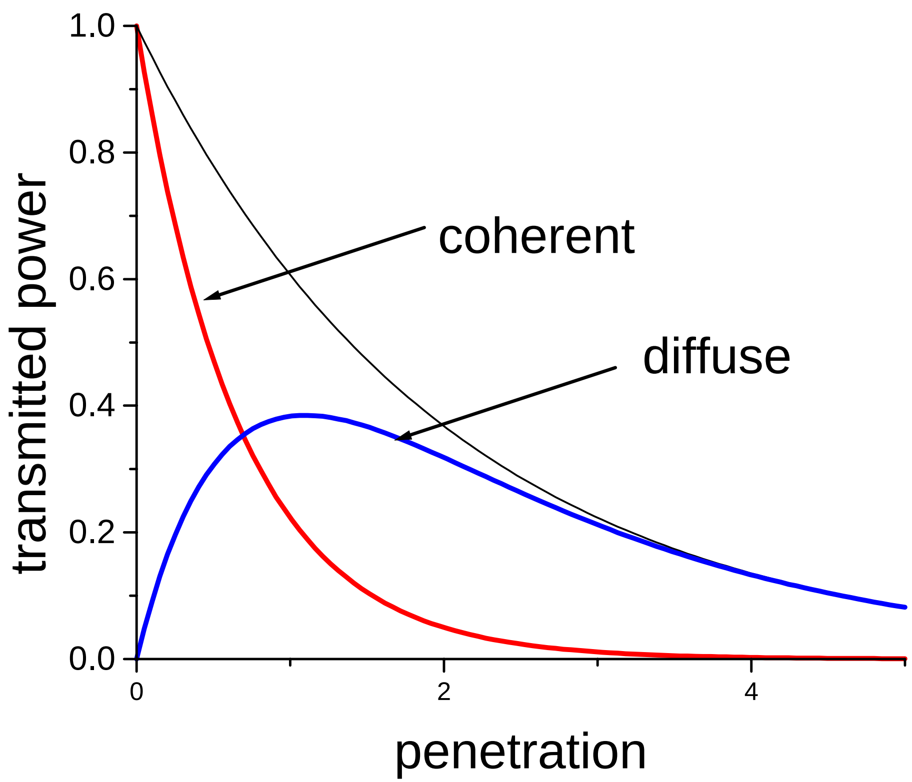
Extended effective medium



Energy conservation

$$\langle Power \rangle \propto \langle E^2 \rangle = \langle E \rangle^2 + \langle \delta E^2 \rangle$$

$$ka \sim 1$$



scattering

dissipation

Metamaterials

Homogenization of metamaterials by field averaging (invited paper)

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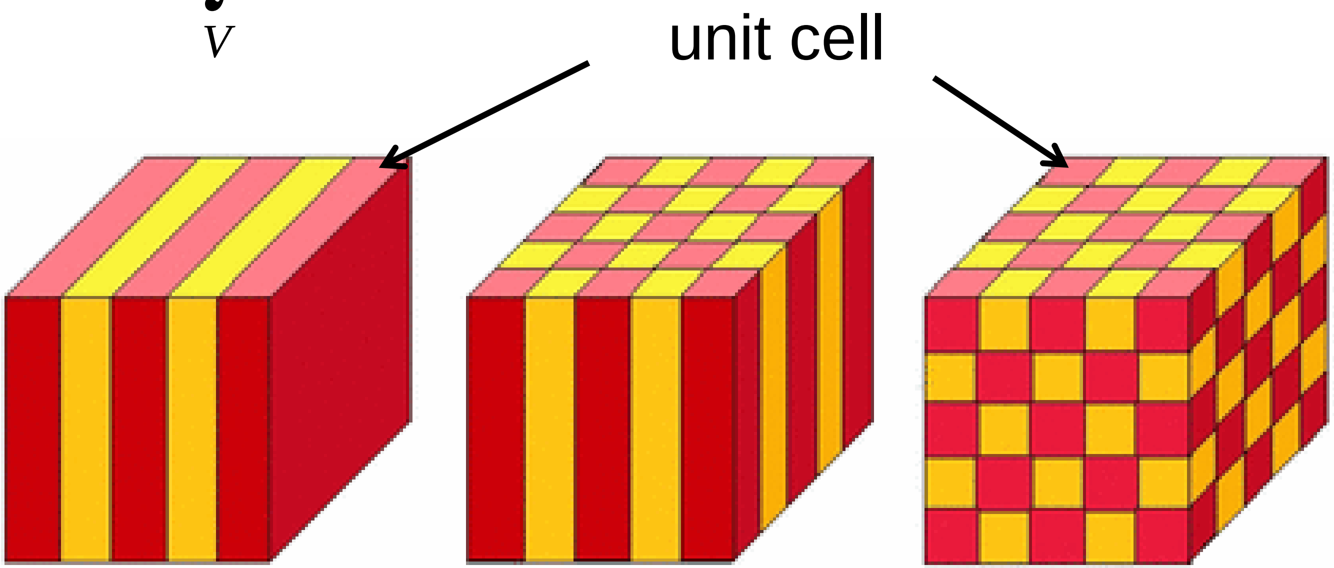
$$\left\langle \vec{E} \right\rangle (\vec{r}) = \int_V \vec{E}(\vec{r})\, d^3 r$$



Comments

static regime. However, for most of the metamaterials demonstrated to date, d is not negligible with respect to the wavelength; yet, in these cases, the description of the structure as a homogeneous medium has produced sensible and useful results, implying that it should be possible to modify or extend the homogenization concepts embodied in Eqs. (1). In 2000, such a modification was suggested and shown to produce self-consistent results for the permittivity and permeability of wire and SRR media.[23]

Our intent here is to justify a numerically based homogenization scheme based on Eqs. (1), in which the local fields computed for one unit cell of a periodic structure are averaged to yield a set of macroscopic fields. Once

Coherent beam

$$\vec{E}(\vec{r}) = \exp[i\vec{k}\cdot\vec{r}]\,\vec{e}(\vec{r})$$

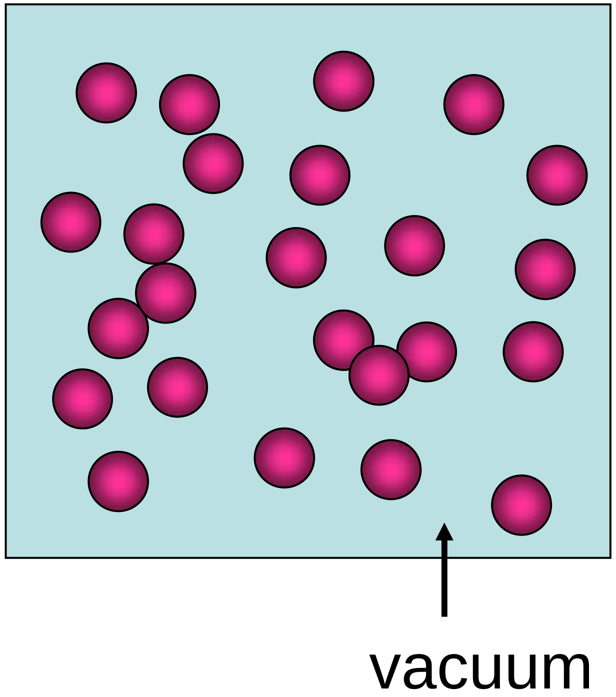
$$\vec{e}(\vec{r}) = \sum_{\vec{G}} \vec{e}_{\vec{G}} \exp[i\vec{G}\cdot\vec{r}]$$

$$e_{\vec{G}} = \int_V \vec{e}(\vec{r}) \exp[-i\vec{G}\cdot\vec{r}]\,d^3r$$

$$\left\langle \vec{E} \right\rangle(\vec{r}) = \vec{e}_{\vec{G}=0} \exp[i\vec{k}\cdot\vec{r}]$$

$$e_{\vec{G}=0} = \int_V \vec{e}(\vec{r})\,d^3r$$

Previous Attempts



MODEL: Random system of identical spheres

a identical

$\varepsilon = \varepsilon_p(\omega)$ local

$\mu = \mu_0$ nonmagnetic

$$n_p = \sqrt{\varepsilon_p(\omega)/\varepsilon_0}$$

$$k_0 = \frac{\omega}{c}$$

van de Hulst

Light scattering by small particles (1957)

$f \ll 1$ dilute limit

complex

$$n_{eff} = 1 + \underbrace{i\gamma S(0)}_{\delta n_{eff}}$$

δn_{eff}

$$\gamma = \frac{3}{2}\frac{f}{(k_0 a)^3}$$

volume filling fraction

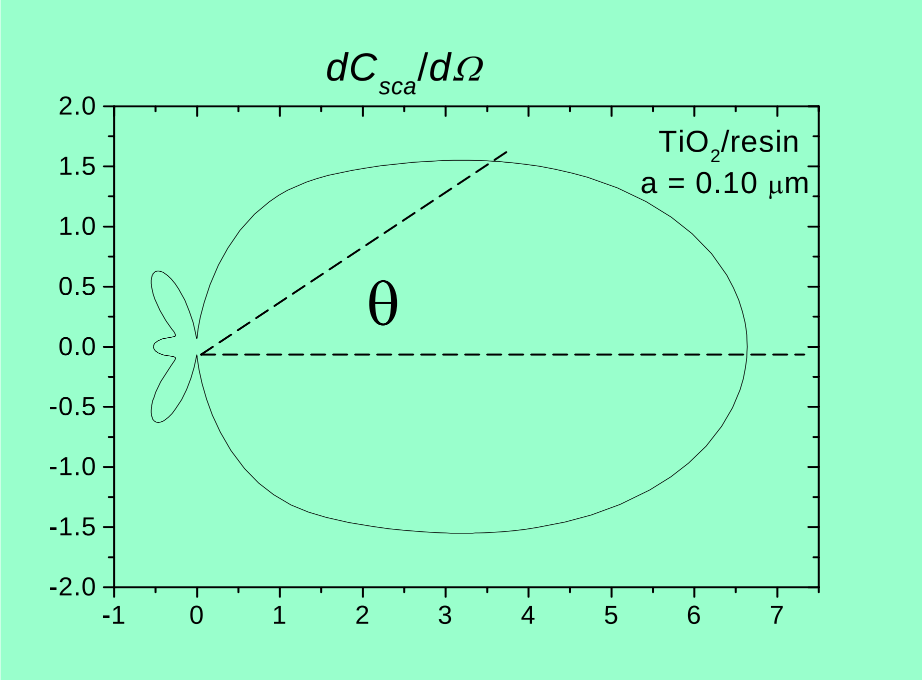
Scattering matrix

$$\begin{pmatrix} E_{\parallel}^{s} \\ E_{\perp}^{s} \end{pmatrix} = \frac{e^{ikr}}{-ikr} \begin{pmatrix} S_2 & 0 \\ 0 & S_1 \end{pmatrix} \begin{pmatrix} E_{\parallel}^{inc} \\ E_{\perp}^{inc} \end{pmatrix}$$

sphere

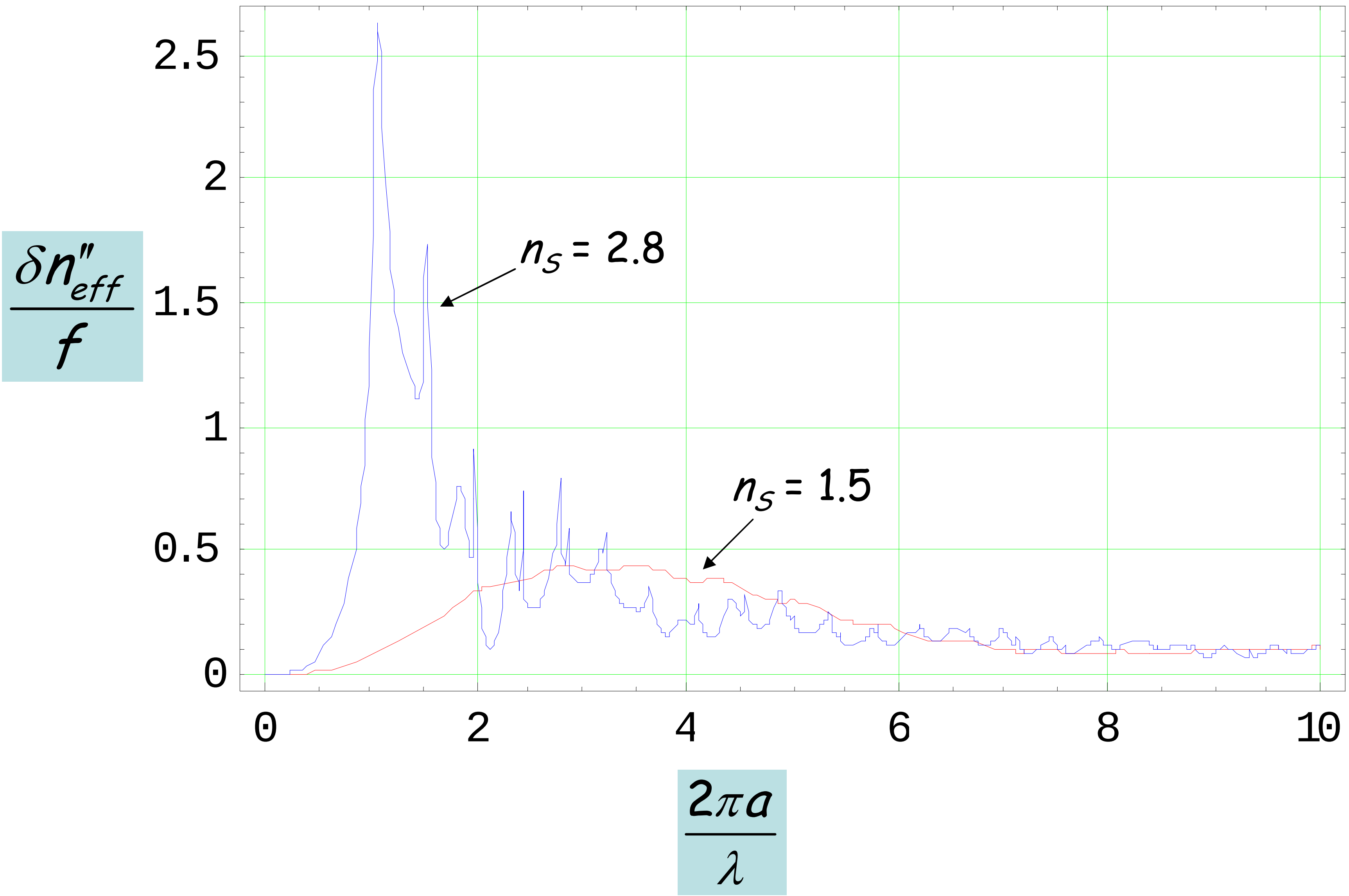
$$S_1(0) = S_2(0) = S(0)$$

MIE



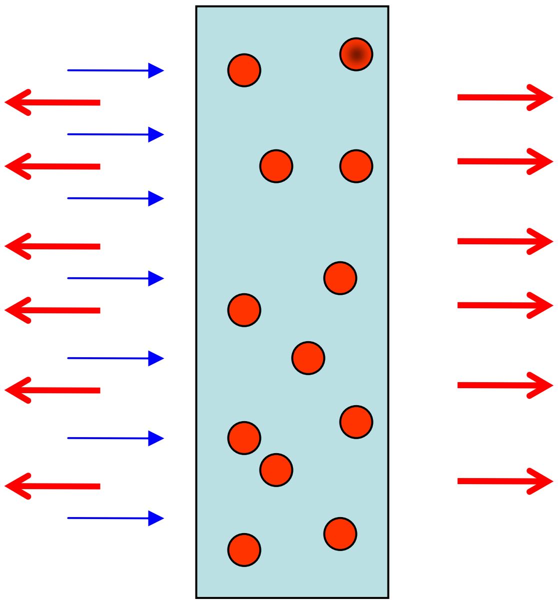
van de Hulst

scattering



Craig Bohren

J. Atmos Sci. 43, 468 (85)



transmission $n_{eff} = 1 + i\gamma S(0)$

reflection $n_{eff} = 1 + i\gamma S_1(\pi)$

Proposition

$$\varepsilon_{eff} = 1 + i\gamma\left[S(0) + S_1(\pi)\right]$$

$$\mu_{eff} = 1 + i\gamma\left[S(0) - S_1(\pi)\right]$$

MAGNETIC ?

$$r = \frac{\sqrt{\mu} - \sqrt{\varepsilon}}{\sqrt{\mu} + \sqrt{\varepsilon}}$$

...It might be expected that a composite medium is nonmagnetic if its components are, but this is not correct... which was recognized as long ago as 1909 by Gans and Happel...

Attempts

RG Barrera & A García-Valenzuela

JOSA A ***20****, 296 (2003)*

$$\mu_{eff}^{TE}(\theta_i) = 1 + \frac{i\gamma S_-^{(1)}(\theta_i)}{\cos^2\theta_i}$$

$$\varepsilon_{eff}^{TE}(\theta_i) = 1 + i\gamma\left(2S_+^{(1)}(\theta_i) - S_-^{(1)}(\theta_i)\tan^2\theta_i\right)$$

MAGNETIC

$$S_+^{(1)}(\theta_i) = \frac{1}{2}\left[S(0) + S_1(\pi - 2\theta_i)\right]$$

$$S_-^{(1)}(\theta_i) = S(0) - S_1(\pi - 2\theta_i)$$

Normal incidence

$$\varepsilon_{eff} = 1 + i\gamma\left[S(0) + S_1(\pi)\right]$$

$$\mu_{eff} = 1 + i\gamma\left[S(0) - S_1(\pi)\right]$$

Comment:

…is a quite uncomfortable result…

….to say the least…

Small particles

$$S(0) = S_1(\pi - 2\theta_i)$$

Our new result

IN TURBID COLLOIDAL SYSTEMSTHE EFFECTIVE MEDIUM ***EXISTS***
BUT IT IS ***NONLOCAL***

ELECTROMAGNETIC RESPONSE

GENERALIZED EFFECTIVE CONDUCTIVITY

$$\left\langle \vec{J}_{ind} \right\rangle = \hat{\sigma}_{eff} \left\langle \vec{E} \right\rangle$$

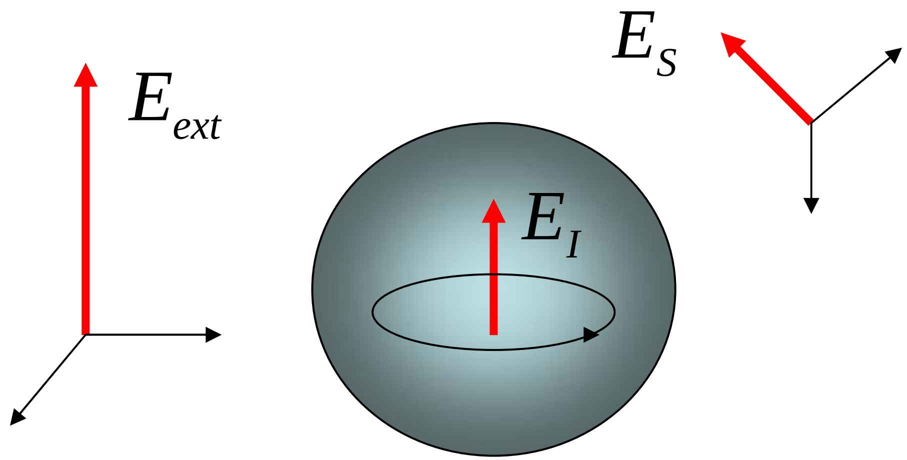
TOTAL

LINEAR OPERATOR

Local vs nonlocal

Is it a matter of taste?...

ISOLATED SPHERE



$$\sigma_S(\vec{r};\omega) = \begin{cases} \sigma_S(\omega) & \vec{r} \in V_S \\ 0 & \vec{r} \notin V_S \end{cases}$$

$$\vec{J}_{ind}(\vec{r};\omega) = \sigma_S(\vec{r};\omega)\vec{E}_I(\vec{r};\omega)$$

$$= \int \underbrace{\overleftrightarrow{\sigma}_S^{NL}(\vec{r},\vec{r}\,';\omega)} \cdot \vec{E}_{ext}(\vec{r}\,';\omega)\, d^3r'$$

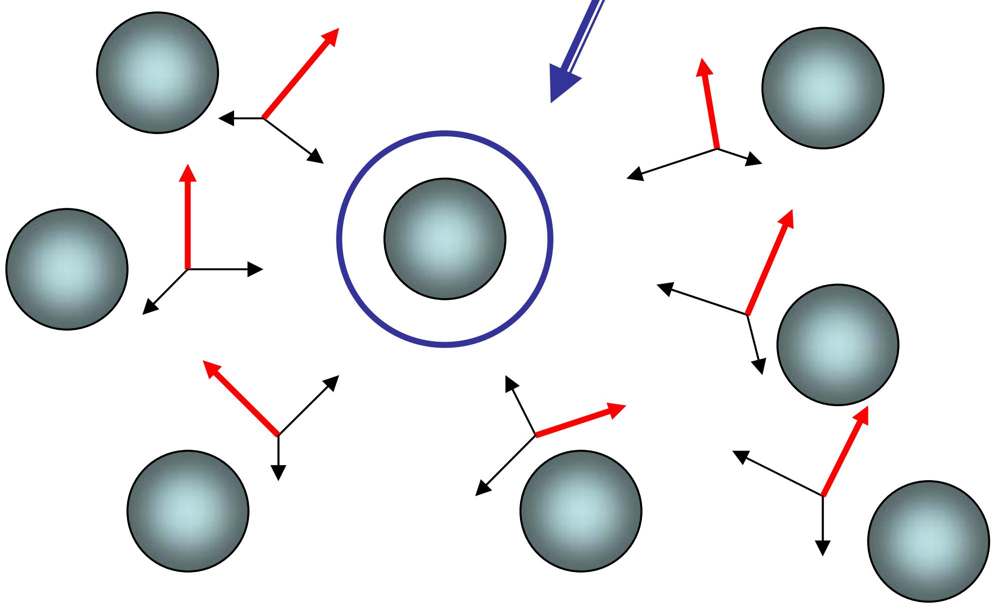
Total induced current

$$\vec{J}_{ind}(\vec{r};\omega) = \sum_i \vec{J}_{ind,i}(\vec{r};\omega)$$

$$= \sum_i \int \overleftrightarrow{\sigma}_S^{NL} \vec{r} - \vec{r}_i, \vec{r}\,' - \vec{r}_i;\omega) \cdot \vec{E}_{exc,i}(\vec{r}\,';\vec{r}_1,\vec{r}_2,\ldots\vec{r}_{i-1},\vec{r}_{i+1},\ldots\vec{r}_N;\omega)\, d^3r'$$

NONLOCAL

EXCITING FIELD



Effective-Field Approximation

$$\vec{E}_{exc,i}(\vec{r}';\vec{r}_1,\vec{r}_2,\ldots\vec{r}_{i-1},\vec{r}_{i+1},\ldots\vec{r}_N;\omega) \approx \left\langle \vec{E}(\vec{r}',\omega) \right\rangle$$

…valid in the dilute regime

$$\vec{J}_{ind}(\vec{r};\omega) = \sum_i \int \overleftrightarrow{\sigma}_S^{NL}(\vec{r}-\vec{r}_i,\vec{r}'-\vec{r}_i;\omega)\cdot\left\langle \vec{E}(\vec{r}',\omega) \right\rangle d^3r'$$

$$\left\langle \vec{J}_{ind}(\vec{r};\omega) \right\rangle = \int \underbrace{\left\langle \sum_i \overleftrightarrow{\sigma}_S^{NL}(\vec{r}-\vec{r}_i,\vec{r}'-\vec{r}_i;\omega) \right\rangle}_{\overleftrightarrow{\sigma}_{eff}(|\vec{r}-\vec{r}'|;\omega)} \left\langle \vec{E}(\vec{r}',\omega) \right\rangle d^3r'$$

the probability density
Is homogeneous

$\overleftrightarrow{\sigma}_{eff}(|\vec{r}-\vec{r}'|;\omega) \longleftarrow$ GENERALIZED NONLOCAL CONDUCTIVITY

GENERALIZED NONLOCAL OHM'S LAW

Momentum representation

$$\left\langle \vec{J} \right\rangle^{ind} (\vec{p},\omega) = \overleftrightarrow{\sigma}_{eff}(\vec{p},\omega) \cdot \left\langle \vec{E} \right\rangle (\vec{p},\omega) \qquad n_0 = \frac{N}{V}$$

$$\boxed{\overleftrightarrow{\sigma}_{eff}(\vec{p},\omega) = n_0 \overleftrightarrow{\sigma}_S^{NL}(\vec{p}' = \vec{p},\vec{p};\omega)}$$

FT

$$\overleftrightarrow{\sigma}_S^{NL}(\vec{r},\vec{r}';\omega) \rightarrow \overleftrightarrow{\sigma}_S^{NL}(\vec{p},\vec{p}';\omega) \rightarrow \overleftrightarrow{\sigma}_S^{NL}(\vec{p},\underline{\vec{p}' = \vec{p}};\omega)$$

3 X 3 = 9

LT scheme

Probability density is homogeneous and isotropic

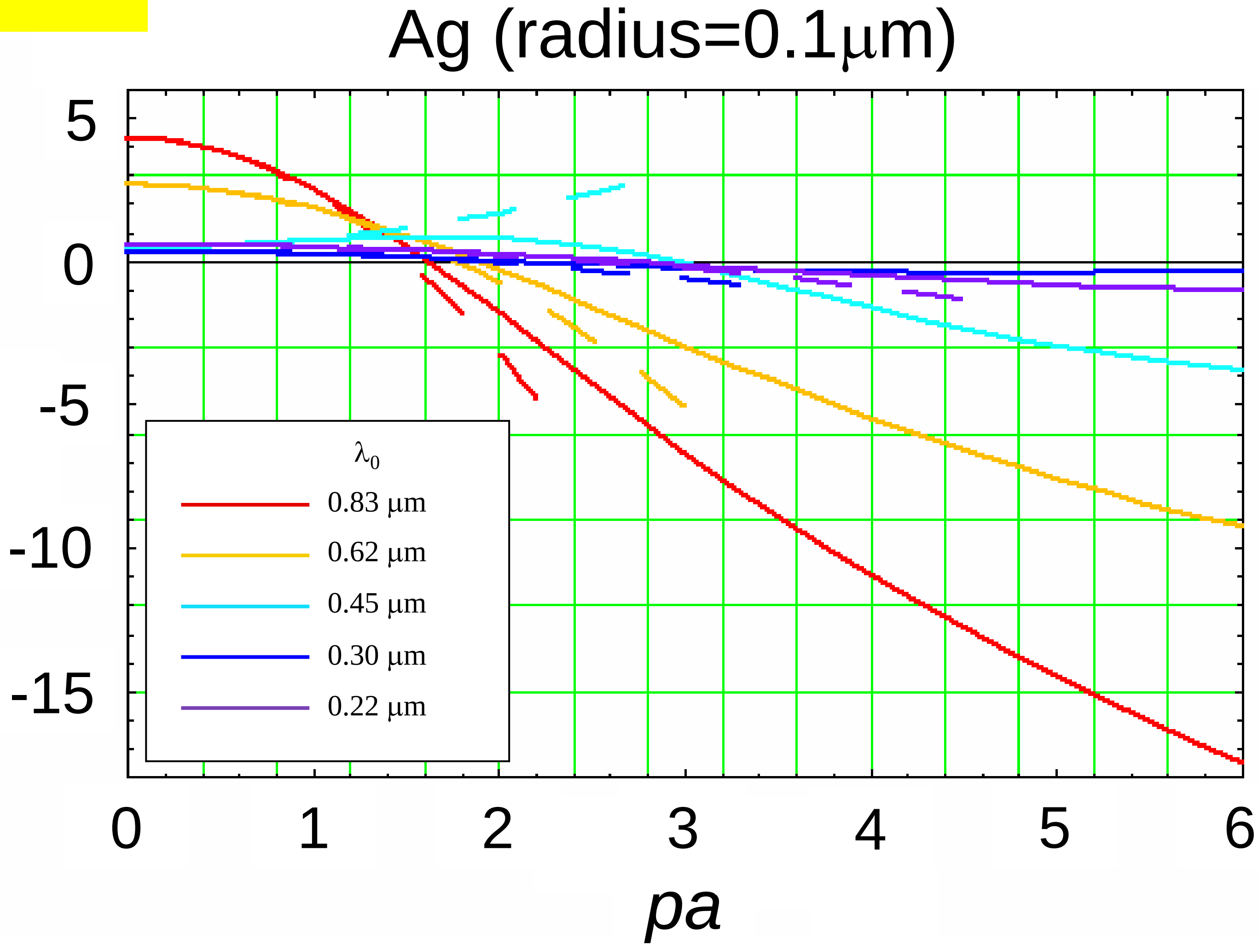
$$\overleftrightarrow{\sigma}_{eff}(\vec{p};\omega) = \sigma^{L}_{eff}(p,\omega)\hat{p}\hat{p} + \sigma^{T}_{eff}(p,\omega)(\overline{\overline{1}} - \hat{p}\hat{p})$$

generalized effective nonlocal dielectric function

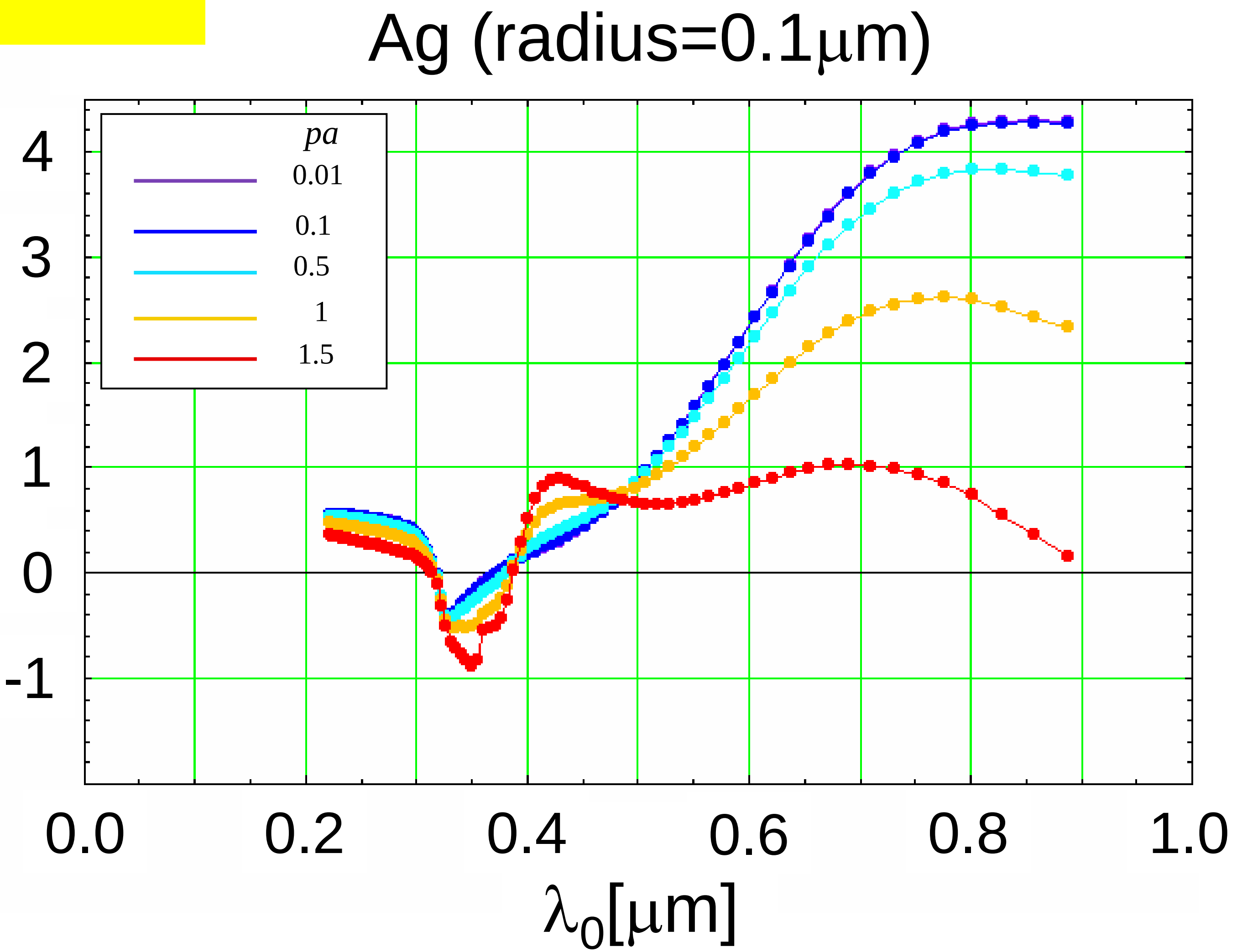
$$\overleftrightarrow{\varepsilon}_{eff}(\vec{p};\omega) = \overleftarrow{1}\varepsilon_0 + \frac{i}{\omega}\overleftrightarrow{\sigma}_{eff}(\vec{p};\omega)$$

$$\varepsilon^{L}_{eff}(p,\omega) \qquad \varepsilon^{T}_{eff}(p,\omega)$$

$$\frac{\mathrm{Re}[\varepsilon^T(p;\omega)-1]}{f}$$

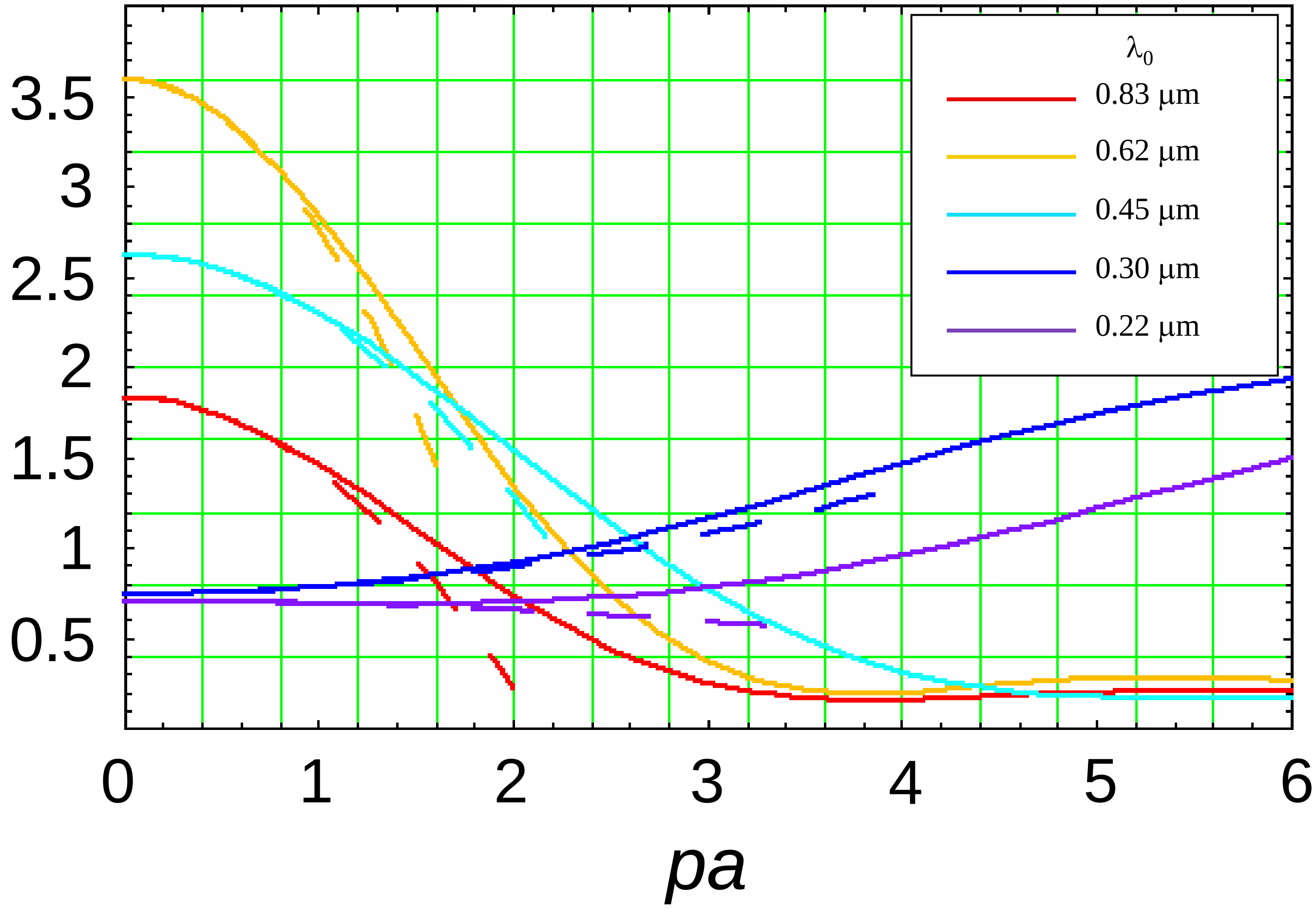


$$\frac{\mathrm{Re}[\varepsilon^T(p;\omega)]-1}{f}$$

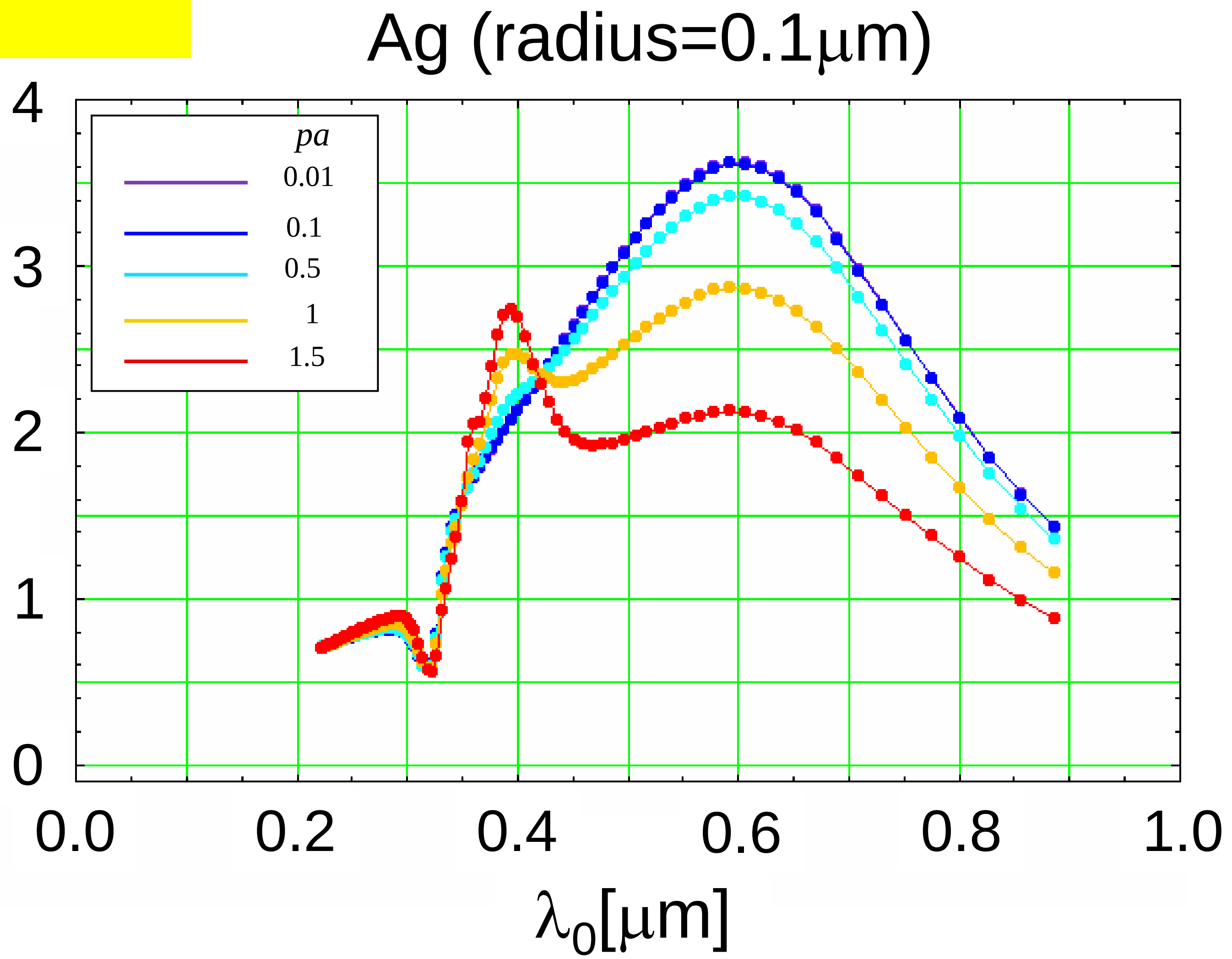


$$\frac{\mathrm{Im}[\varepsilon^{T}(p;\omega)]-1}{f}$$

Ag (radius=0.1μm)



$$\frac{\mathrm{Im}[\varepsilon^T(p;\omega)]-1}{f}$$



Electromagnetic modes

dispersion relation

$$p = p' + ip''$$

longitudinal $$\tilde{\varepsilon}^{L}_{eff}(p,\omega) = 0$$

transverse $$p = k_0\sqrt{\tilde{\varepsilon}^{T}_{eff}(p,\omega)}$$

nonlocal

$$p^L(\omega)$$

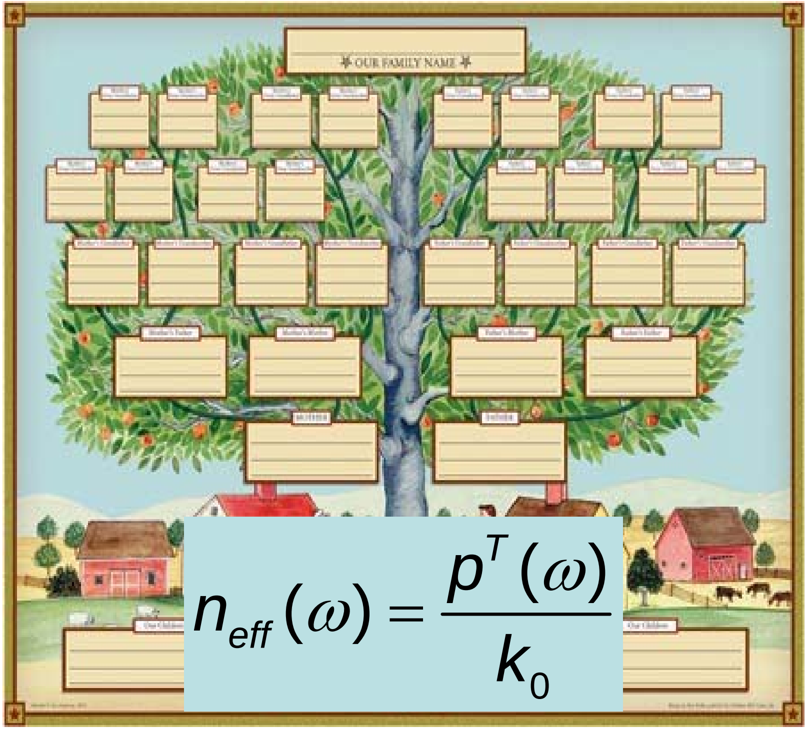
$$p^T(\omega)$$

effective index of refraction

$$p^T(\omega) = k_0 n_{eff}(\omega)$$

$$n_{eff}(\omega) = \frac{p^T(\omega)}{k_0}$$

Genealogy



$$n_{eff}(\omega) = \frac{p^T(\omega)}{k_0}$$

$$p = k_0 \sqrt{\tilde{\varepsilon}^T_{eff}(p \to 0;\, \omega)}$$ local

$$p = k_0 \sqrt{\tilde{\varepsilon}^T_{eff}(p, \omega)}$$ nonlocal

Comparisons

Exact

$$p = k_0 \sqrt{\tilde{\varepsilon}^{T}_{eff}(p,\omega)} = k_0 \sqrt{\tilde{\varepsilon}^{[0]}_{eff}(\omega) + \tilde{\varepsilon}^{L(T)[2]}_{eff}(\omega)(pa)^2 + \ldots}$$

Long wavelength

local

$$p = k_0 \sqrt{\tilde{\varepsilon}^{[0]}_{eff}(\omega)} \Rightarrow n_{eff} = \sqrt{\tilde{\varepsilon}^{[0]}(\omega)}$$

Quadratic

nonlocal

$$p = k_0 \sqrt{\tilde{\varepsilon}^{[0]}(\omega) + \tilde{\varepsilon}^{T[2]}(\omega)(pa)^2 + \ldots} \Rightarrow n_{eff} = \sqrt{\frac{\varepsilon^{[0]}(\omega)}{1-(k_0 a)^2\, \tilde{\varepsilon}^{T[2]}(\omega)}}$$

Light-cone approximation

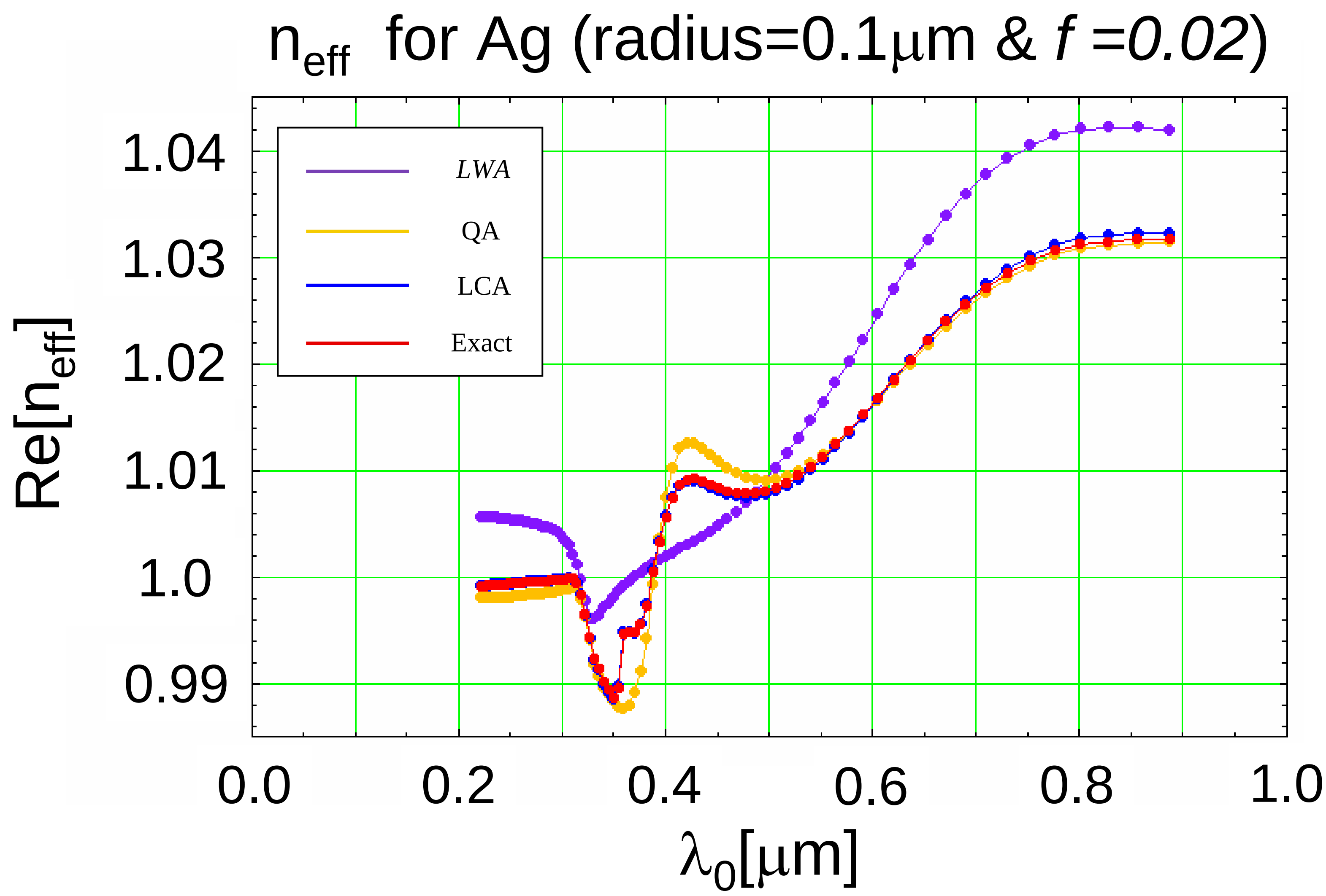
nonlocal

$$p^2 = k_0^2\, \tilde{\varepsilon}^T (p = k_0, \omega)$$

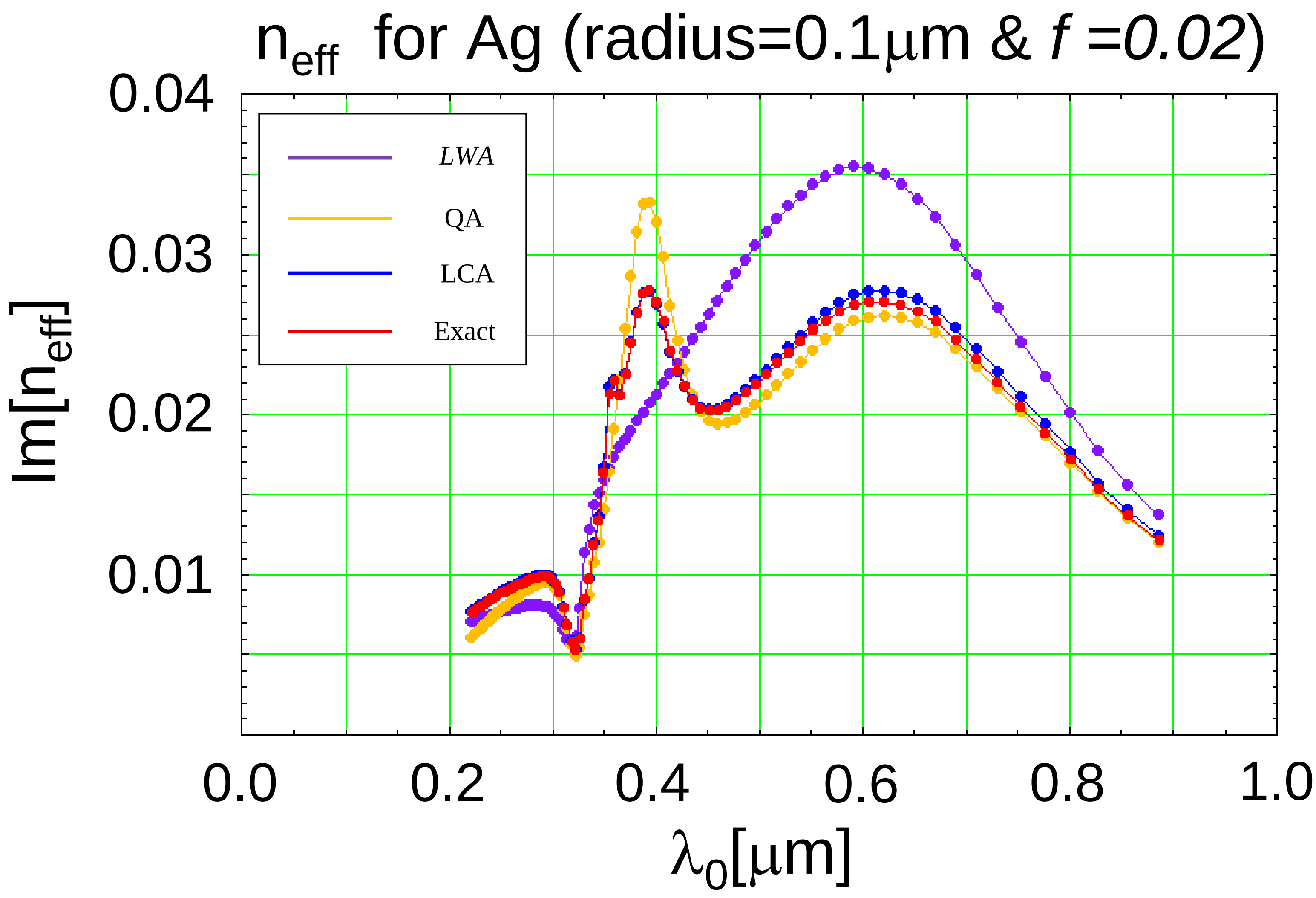
nonlocal ancestry

$$n_{eff} = 1 + i\gamma S(0)$$

van de Hulst



n_{eff} for Ag (radius=0.1μm & *f =0.02*)
Re[n_{eff}]
1.04
1.03
1.02
1.01
1.0
0.99
0.0
0.2
0.4
0.6
0.8
1.0
λ_0[μm]
LWA
QA
LCA
Exact



n_eff for Ag (radius=0.1μm & f =0.02)
0.04
0.03
0.02
0.01
Im[n_eff]
LWA
QA
LCA
Exact
0.0
0.2
0.4
0.6
0.8
1.0
λ_0[μm]

Metamaterials

Homogenization of metamaterials by field averaging (invited paper)

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$$\bar{\varepsilon}_x = \varepsilon_0 \frac{\sin(q_y d)}{q_y d}. \quad (19)$$

A similar calculation for the permeability yields

$$\bar{\mu}_z = \mu_0 \frac{\sin(q_y d)}{q_y d}. \quad (20)$$

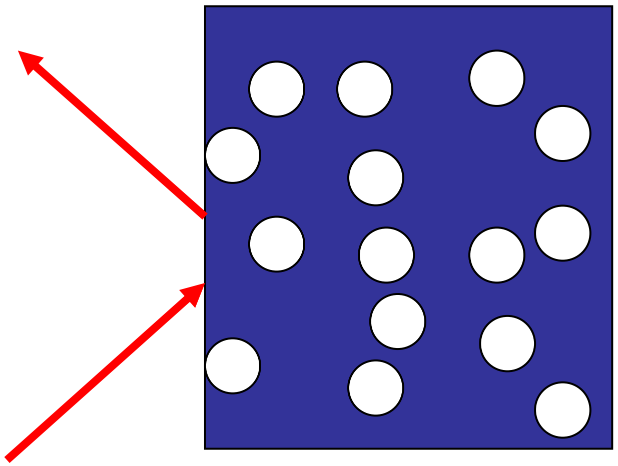
Equations (19) and (20) reveal that the material parameters found from the field-averaging method exhibit spatial dispersion; that is, the material parameters are functions of the propagation vector. Inserting Eqs. (19) and (20) into Eq. (17), we recover the correct dispersion relation for free space, or

$$\omega = q_y \sqrt{\varepsilon_0 \mu_0}. \quad (21)$$

Spatially dispersive medium parameters are less intuitive and less convenient to apply. However, the terms dependent on $\mathbf{q}$ in Eqs. (19) and (20) are seen to be a result of the finite differencing or discretization of Maxwell's equations and have no other physical origin. Since we recover the correct free-space dispersion relation, Eq. (21), via this scheme there is the hint that the averaged material parameters may have validity if we simply remove the term $\sin(q_y d)/q_y d$. When the unit cell is not empty, this procedure will obviously not lead to an exact result but may still be a reasonable approximation.

Consequences

If $n_{eff}(\omega)$ has a nonlocal ancestry it cannot be used in local CE (Fresnel's relations)



$$n_{eff} = 1 + i\gamma S(0)$$

ABUSE

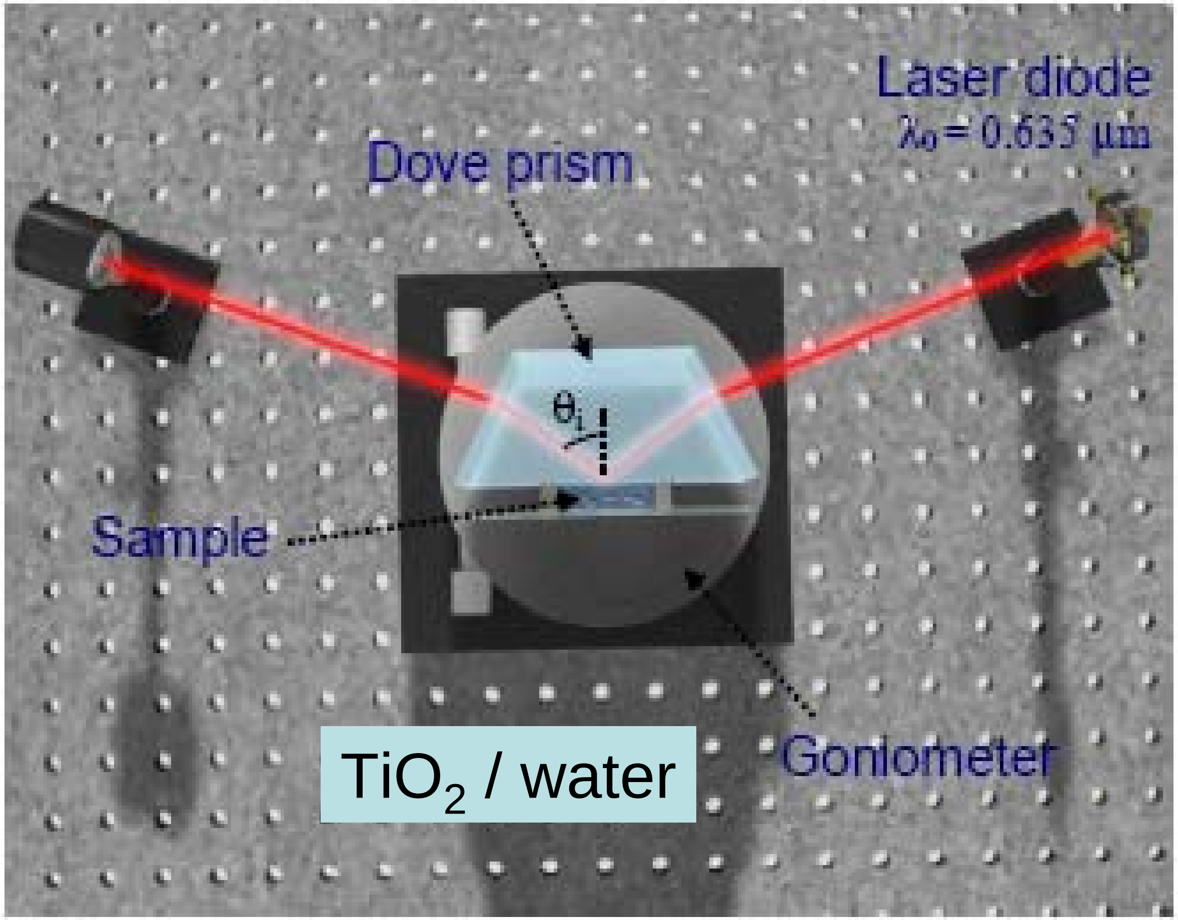
Internal Reflection configuration

Internal reflection configuration

great sensitivity

IEMM

$$R^{Fresnel}(\theta_i; n_{eff}^{vdH})$$



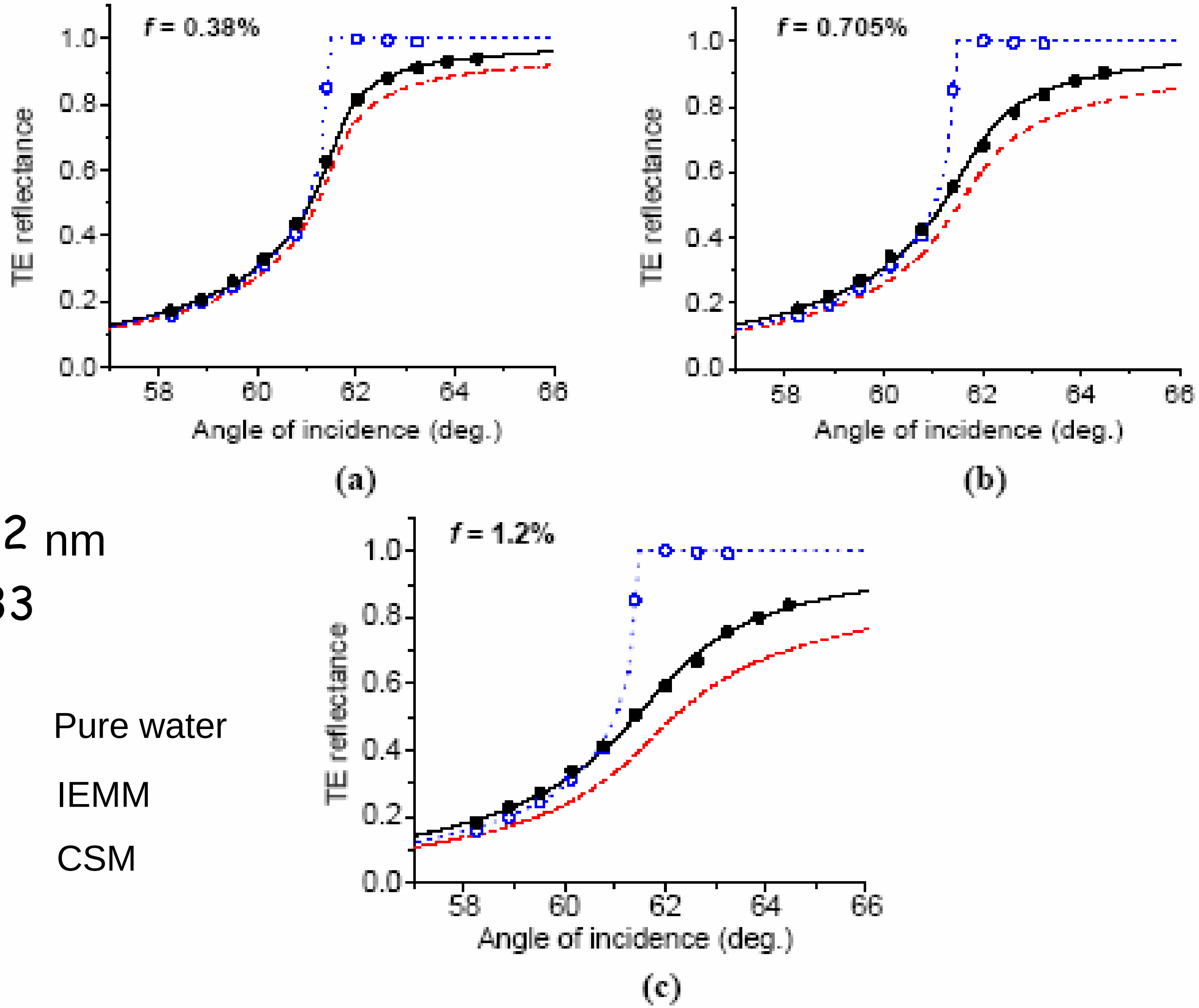
TiO_2 / water

$$R(\theta_i)$$

A García-Valenzuela, RG Barrera,
C. Sánchez-Pérez, A. Reyes-Coronado,
E Méndez, Optics Express, **13**, 6723 (2005)

Comparison

TiO$_2$ / water



$a_0 = 112$ nm

$\sigma = 1.33$

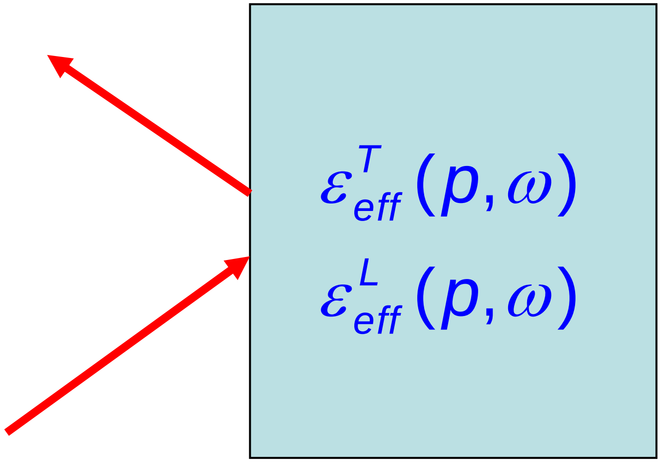
Pure water

IEMM

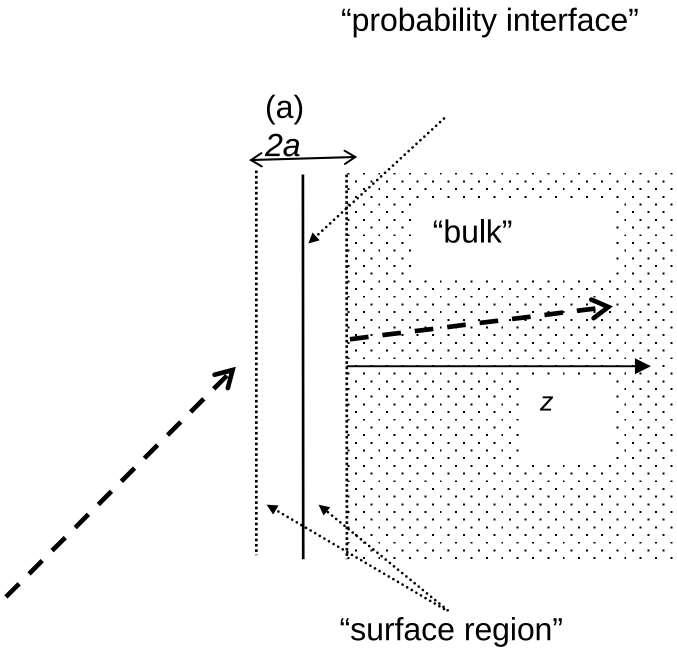
CSM

Nonlocal optics

not possible



translational invariance



$$\varepsilon^{T}_{eff}(p,\omega) \rightarrow \varepsilon^{T}_{eff}(\vec{p}_{||};p_z,p'_z;\omega)$$

$$\varepsilon^{L}_{eff}(p,\omega) \rightarrow \varepsilon^{L}_{eff}(\vec{p}_{||};p_z,p'_z;\omega)$$

Information about the surface

$\epsilon\ \mu$ scheme

material fields

$$\rho^{ind} = -\nabla \cdot \vec{P}$$

$$\vec{J}_M = \nabla \times \vec{M}$$

$$\left\langle \vec{J}_{ind} \right\rangle = \vec{J}_P + \vec{J}_M$$

$$\tilde{\varepsilon}_{eff}(p,\omega) = \varepsilon^L(p,\omega)$$

$$\tilde{\mu}_{eff}(p,\omega) = \frac{1}{1 - \frac{k_0^2}{p^2}\left(\varepsilon_{eff}^T(p,\omega) - \varepsilon_{eff}^L(p,\omega)\right)}$$

magnetic response !

$$p = k_0 \sqrt{\tilde{\varepsilon}_{eff}(p;\omega)\tilde{\mu}_{eff}(p;\omega)}$$

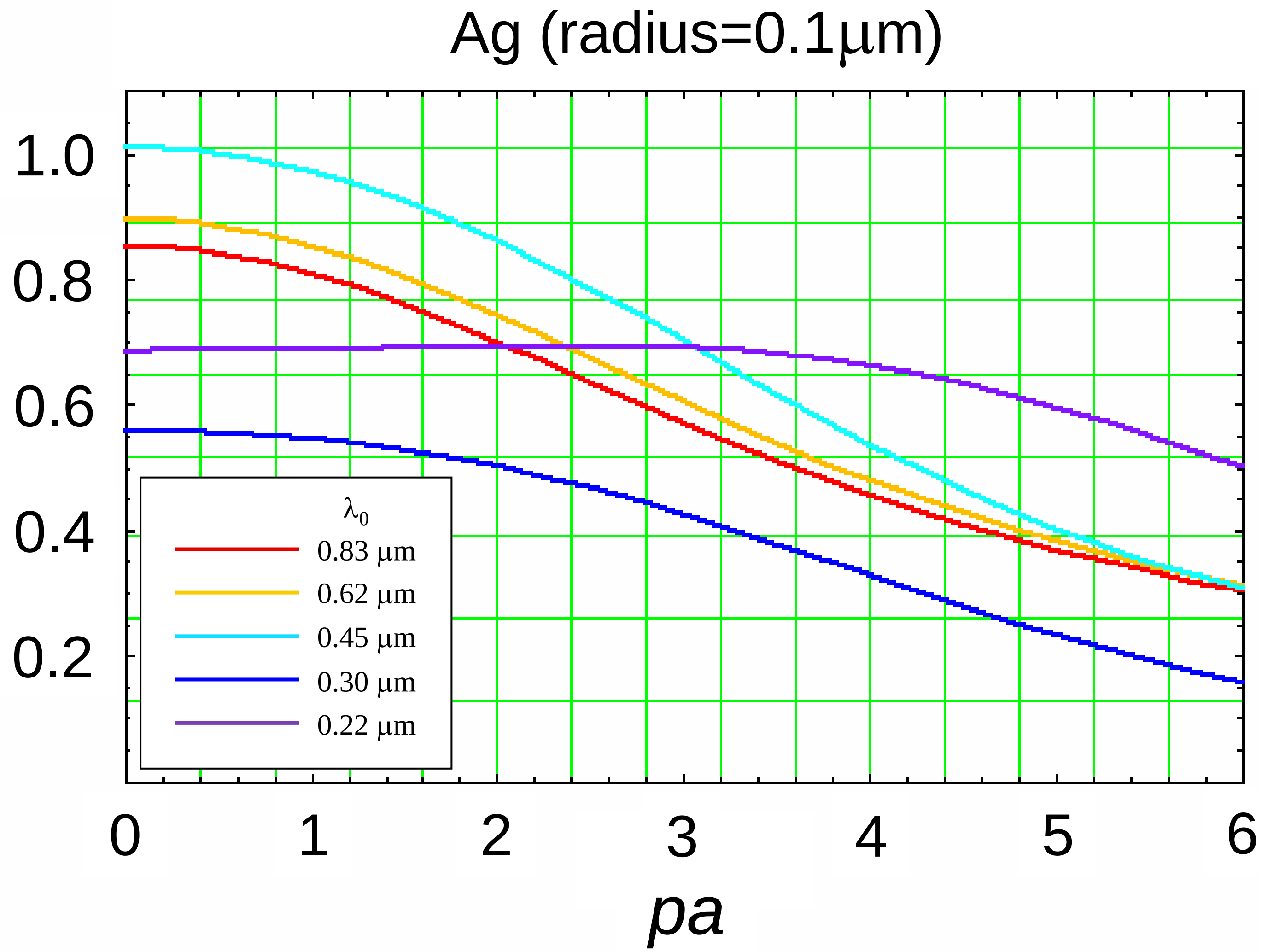
dispersion relation

$$p = k_0 \sqrt{\tilde{\varepsilon}_{eff}^T(p,\omega)}$$

dispersion relation

$$\frac{1}{f}\mathrm{Re}\left[\frac{\mu_0}{\mu(\vec{p},\omega)}-1\right]$$

MAGNETIC RESPONSE



Conclusions

In turbid colloidal systems the effective index of refraction, due to its nonlocal character, is able to describe the *propagation* of light, but it cannot describe its *reflection*

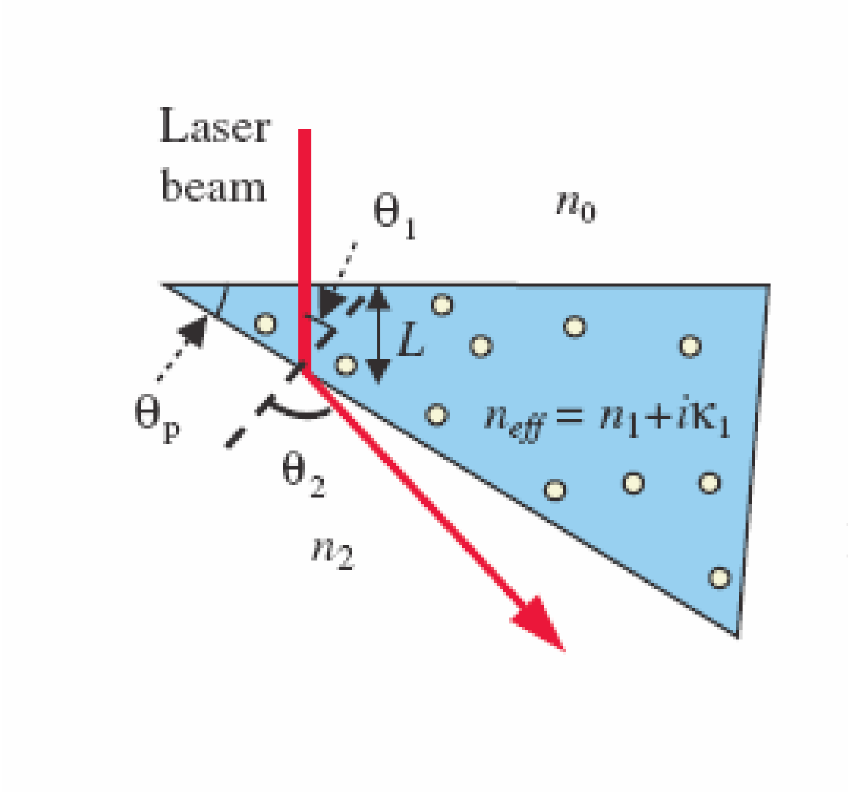
This is important because the naïve use of the effective index of refraction in the calculation of reflection amplitudes has been done many times without too much (intellectual) reflection

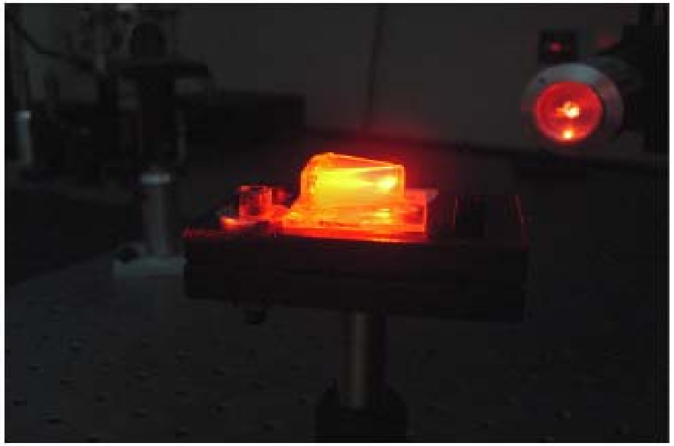
There is a magnetic response in colloidal systems with non magnetic components (optical magnetism)

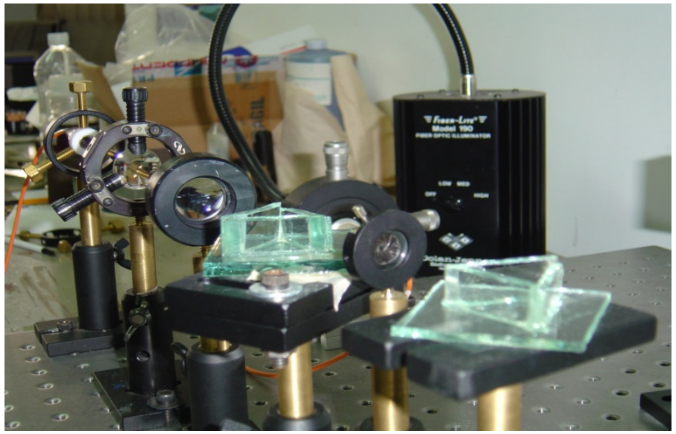
How to measure n_{eff}?

Latex spheres / water

Refraction Spectroscopy







A. Reyes-Coronado, A García-Valenzuela,
C. Sánchez-Pérez, RG Barrera
New Journal of Physics **7** (2005) 89 [1-22]

Refraction spectroscopy

Firm theoretical grounds for particle sizing in turbid colloids using light refraction

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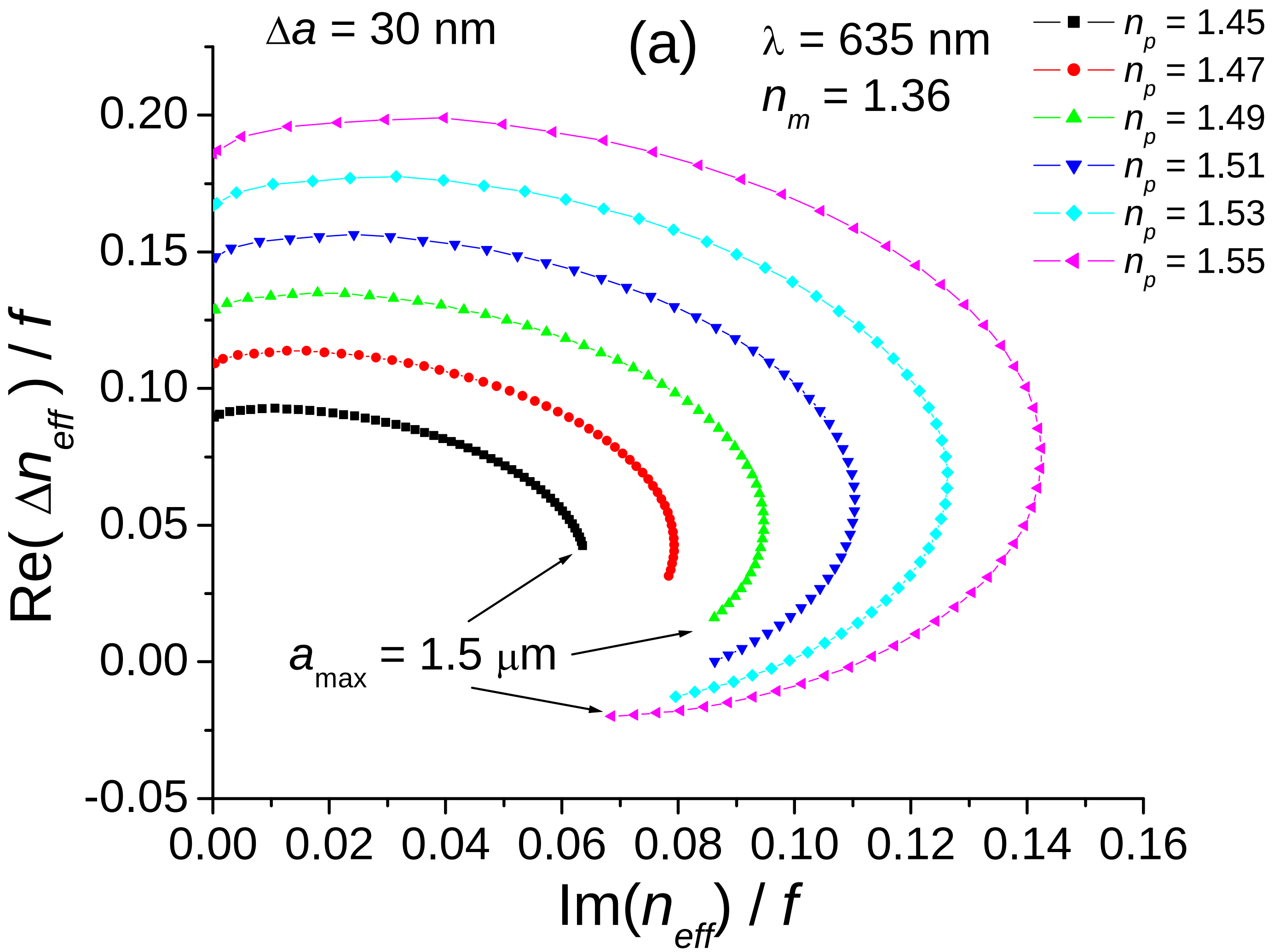
Abstract. Using a non-local effective-medium approach we analyze the refraction of light in a colloidal medium. We discuss the theoretical grounds and all the necessary precautions to design and perform experiments to measure the effective refractive index in dilute colloids. As an application, we show that it is possible to size small dielectric particles in a colloid from the measurement of the complex effective refractive index and the volume fraction occupied by the particles.

1. Introduction.

Particle sizing



Perspectives

BULK

HIGHER DENSITY

Quasicrystalline approximation

LONGITUDINAL MODES

DO THEY EXIST ?

$$\varepsilon^L(\vec{p},\omega) = 0$$

ENERGY TRANSFER

$$\vec{S} \cdot \hat{p} = ?$$

$$\vec{S} = \vec{E} \times \vec{H} = \vec{E} \times \hat{\mu}^{-1}\vec{B}$$

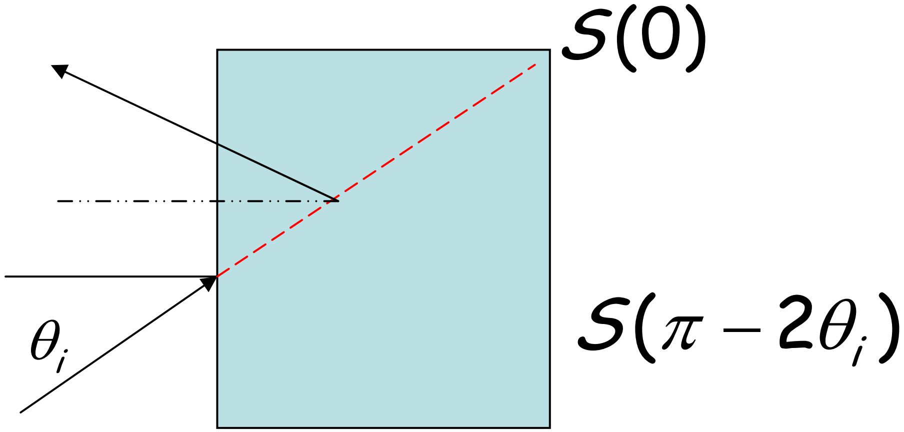
LEFTHANDED ?

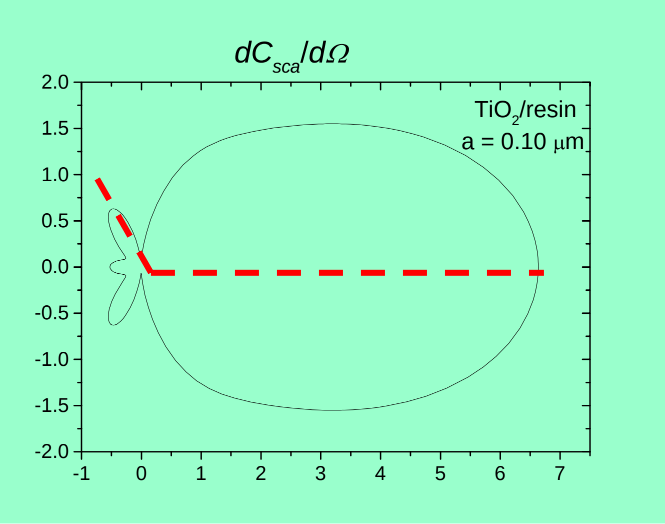
$$\vec{p}(\omega) \times \vec{E} = k_0 n_{eff}(\omega)\hat{p} \times \vec{B}$$

Coherent scattering model

$$\gamma = \frac{3f}{2x^3}$$

$$r_{hs}^{TE} = \frac{\gamma S_1(\pi - 2\theta_i)/\cos\theta_i}{i(\cos\theta_i + [\cos^2\theta_i + 2i\gamma S(0)]^{1/2}) - \gamma S(0)/\cos\theta_i}$$





Comparison

Fit with $n_{eff}^{van\,de\,Hulst}$

Table 1. Retrieved and nominal values of experimental parameters.

Particle size	Retrieved values	Nominal values
Small spheres	$a = 0.1076\ \mu m$	$a = 0.111 \pm 0.005\ \mu m$
	$n_{sphere} = 1.566$	$n_{sphere} = 1.588$
	$\theta_1 = 47.955°$	$\theta_1 = 48.1 \pm 0.22°$
	$L = 2.039$ mm	$L = 1.9 \pm 0.25$ mm
Medium spheres	$a = 0.155\ \mu m$	$a = 0.155 \pm 0.007\ \mu m$
	$n_{sphere} = 1.588$	$n_{sphere} = 1.588$
	$\theta_1 = 48.175°$	$\theta_1 = 48.1 \pm 0.22°$
	$L = 2.05$ mm	$L = 2 \pm 0.25$ mm
Large spheres	$a = 0.247\ \mu m$	$a = 0.24 \pm 0.01\ \mu m$
	$n_{sphere} = 1.55$	$n_{sphere} = 1.588$
	$\theta_1 = 48.337°$	$\theta_1 = 48.1 \pm 0.22°$
	$L = 1.65$ mm	$L = 1.9 \pm 0.25$ mm

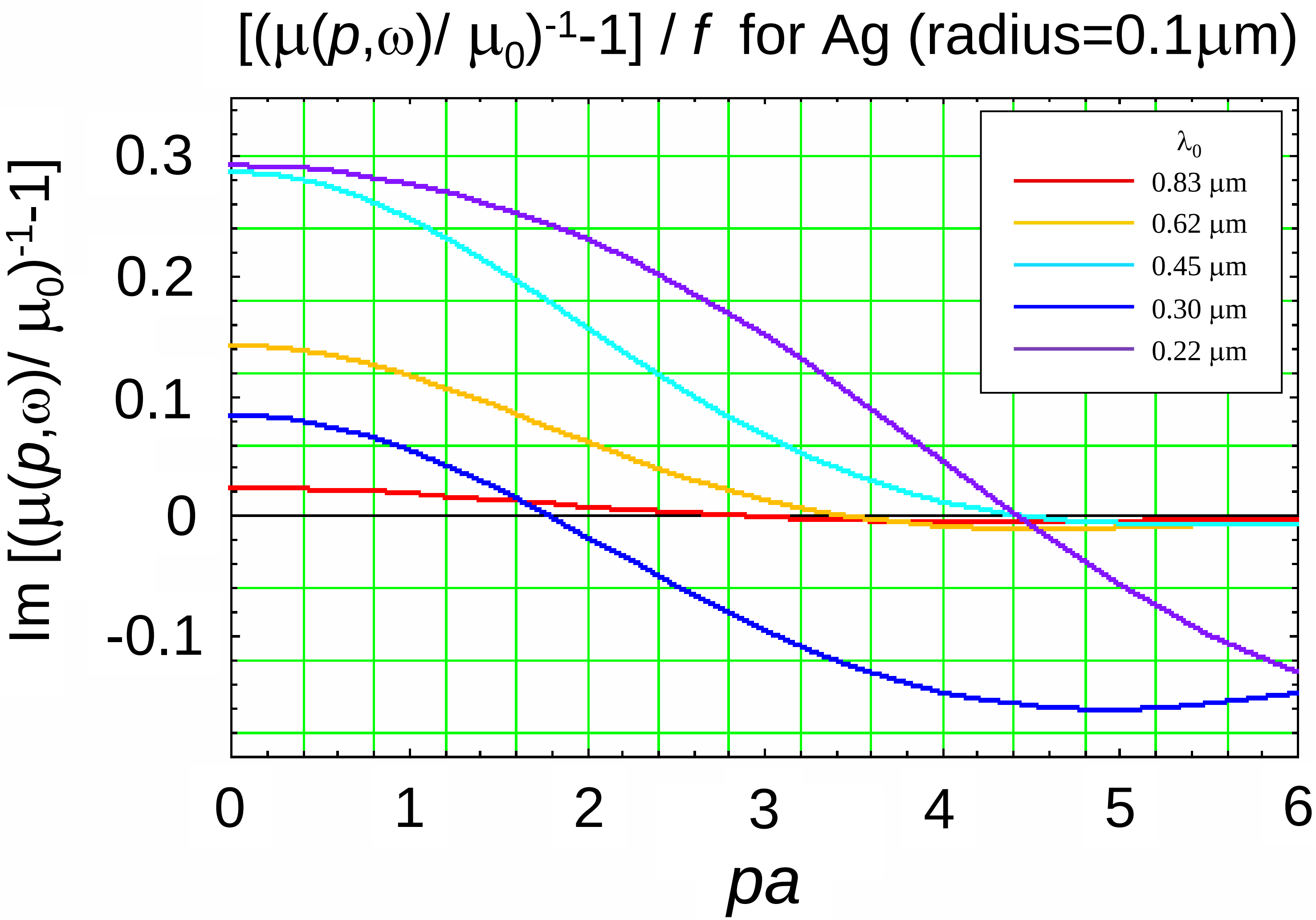
NEXT STEP → CBS

Results

$$\frac{\mu_{eff}(p,\omega)}{\mu_0} = \frac{1}{1 - \frac{k_0^2}{p^2}\left(\underline{\varepsilon_{eff}^T(p,\omega) - \varepsilon_{eff}^L(p,\omega)}\right)}$$

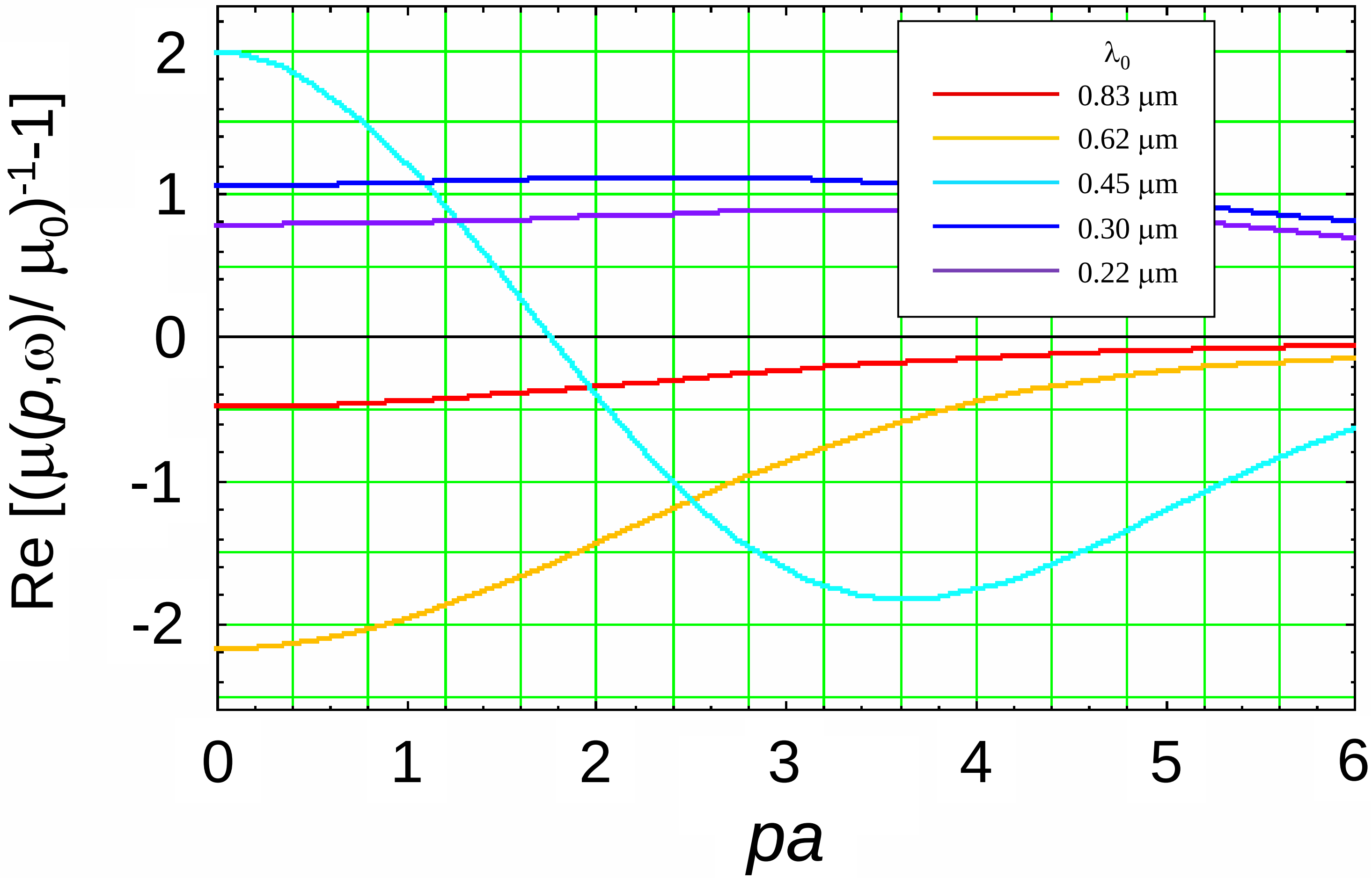
$$\frac{\mu_0}{\mu_{eff}(p,\omega)} - 1 = -\frac{k_0^2}{p^2}\left(\underline{\varepsilon_{eff}^T(p,\omega) - \varepsilon_{eff}^L(p,\omega)}\right)$$

MAGNETIC RESPONSE



MAGNETIC RESPONSE

$[(\mu(p,\omega)/\mu_0)^{-1}-1]/f$ for TiO_2 (radius=0.1μm)

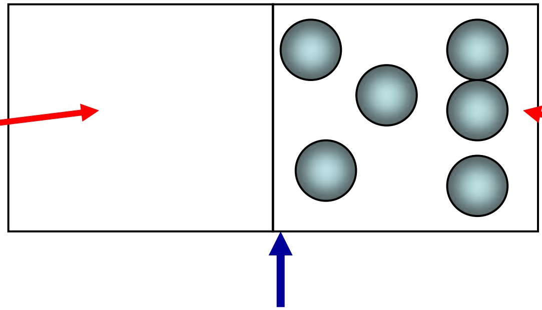


Perspectives

GENERALIZED NONLOCAL OHM'S LAW

$$\left\langle \vec{J} \right\rangle^{ind}(\vec{p},\omega) = n_0 \overleftrightarrow{\sigma}_S^{NL}(\vec{p}'=\vec{p},\vec{p};\omega)\cdot\left\langle\vec{E}\right\rangle(\vec{p},\omega)$$

SURFACE

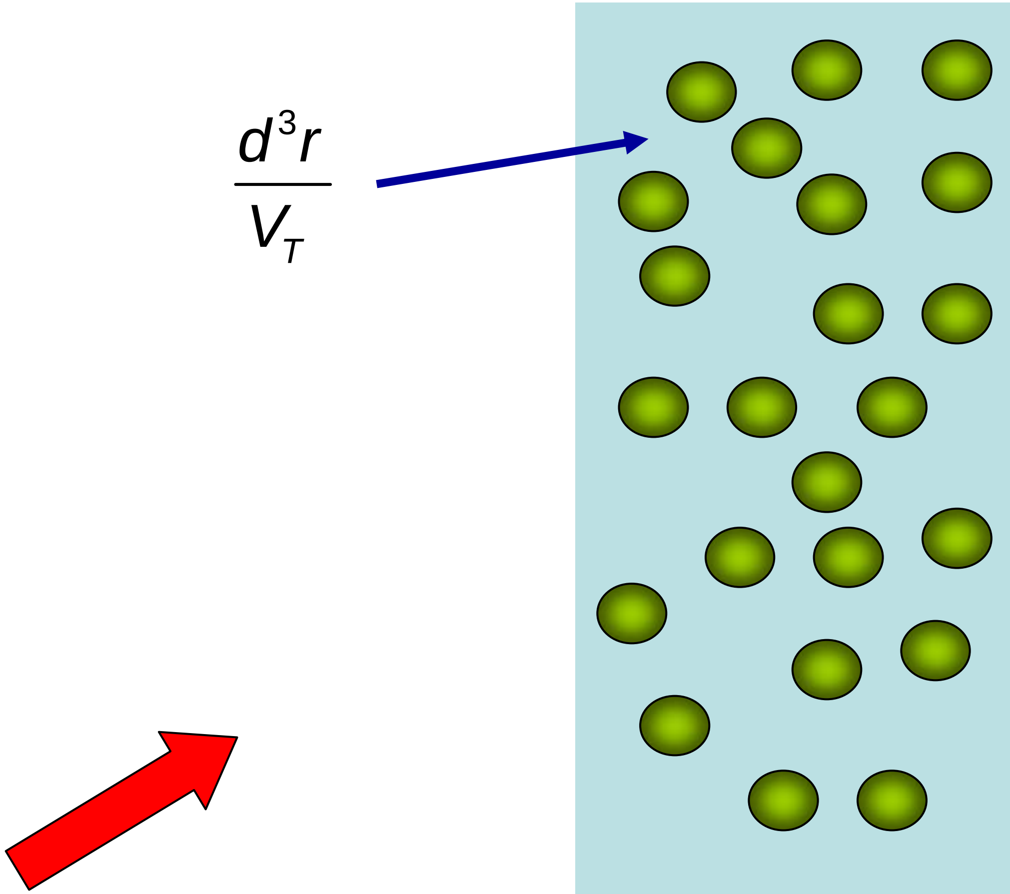


$$\left\langle \vec{J}_{ind}\right\rangle(\vec{p};\omega) = n_0\int \frac{d^3p'}{(2\pi)^3}\,\sigma_{HS}(\vec{p},\vec{p}';\omega)\left\langle\vec{E}\right\rangle(\vec{p}';\omega)$$

$$\overleftrightarrow{\sigma}_S^{NL}(\vec{p},\vec{p}';\omega)\,\Delta(\vec{p}-\vec{p}') \longleftarrow \int_{z_i>0}\exp[-i(\vec{p}-\vec{p}')]\cdot\vec{r}_i\, d^3r_i$$

SURFACE

Configurational average



CONFIGURATION

HOMOGENEOUS
AND
ISOTROPIC

"ON THE AVERAGE"

Generalized NL conductivity

$$\underbrace{i\omega\mu_0\overleftrightarrow{\sigma}_S^{NL}(\vec{r},\vec{r}';\omega)}_{\overleftrightarrow{T}(\vec{r},\vec{r}';\omega)} = U(\vec{r};\omega)\left[\delta(\vec{r}-\vec{r}')\overleftrightarrow{I} + \int \overleftrightarrow{G}_0(\vec{r},\vec{r}';\omega)\cdot \underbrace{i\omega\mu_0\overleftrightarrow{\sigma}_S^{NL}(\vec{r}'',\vec{r}';\omega)}_{\overleftrightarrow{T}(\vec{r},\vec{r}';\omega)} d^3r''\right]$$

$$U(\vec{r};\omega) = i\omega\mu_0\sigma_S(\vec{r};\omega)$$

$$\overleftrightarrow{T}(\vec{r},\vec{r}';\omega) \longrightarrow i\omega\mu_0\overleftrightarrow{\sigma}_S^{NL}(\vec{r},\vec{r}';\omega)$$

T matrix

Small pa

$$\tilde{\varepsilon}_{eff}^{L(T)}(p,\omega) = \tilde{\varepsilon}_{eff}^{[0]}(\omega) + \tilde{\varepsilon}_{eff}^{L(T)[2]}(\omega)\left(\underline{pa}\right)^2 + \dots$$

$$\tilde{\varepsilon} \equiv \frac{\varepsilon}{\varepsilon_0}$$

$p \to 0$ LOCAL LIMIT

NONLOCAL DEPENDENCE

CALCULATION PROCEDURE

INDEPENDENT

Scattering from an *isolated* sphere

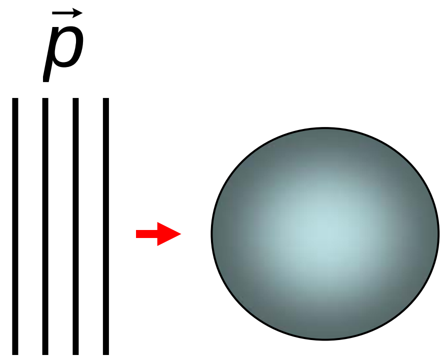
$$\vec{E}_0 \exp[i\left(\vec{p}' \cdot \vec{r} - \omega t\right)]$$

$$(p', \omega)$$

$$\vec{J}^{ind}(\vec{p},\omega) = \vec{\sigma}_S^{NL}(\vec{p},\vec{p}';\omega) \cdot \vec{E}_0$$

$$\vec{p}' = \vec{p}$$

Internal field



$$\vec{J}^{ind}(\vec{r};\omega) = \sigma_S(\omega)\vec{E}^{\mathrm{int}}(\vec{r};\omega)$$

$$J_{ind}(\vec{p}.\omega) = \sigma_S(\omega)\int d^3r\, \vec{E}^{\mathrm{int}}(\vec{r};\omega)\exp[-i\vec{p}\cdot\vec{r}]$$

$$\xi\sum_{n=1}^{\infty} E_n\left(c_n\vec{M}^{(1)}_{o1n} - id_n\vec{N}^{(1)}_{e1n}\right) + (1-\xi)\vec{E}^{ext}$$

$$\sum_{l=0}^{\infty}(2l+1)(-i)^l\, j_l(pr)P_l(\cos\theta)$$

Results

$$\frac{\varepsilon^L(p;\omega)-1}{f}$$

longitudinal

$$\varepsilon^L(p,\omega)=1+f\varsigma\left[1+\chi_S\sum_{n=1}^{\infty}3n(n+1)(2n+1)d_n^L\frac{j_n(x_S)}{x_S}\frac{j_n(x_i)}{x_i}\right]$$

$$\varsigma\equiv 1-\frac{1}{\tilde{\varepsilon}_S}$$

$$x_i=pa$$

$$x_S=k_S a$$

$$k_S=\omega\sqrt{\varepsilon_S\mu_0}$$

Results

transverse

$$\xi = \frac{k_S^2 - k_0^2}{k_S^2 - p^2}$$

$$\frac{\varepsilon^T(p;\omega) - 1}{f}$$

$$\varepsilon^T(p,\omega) = 1 + f\chi_S(1-\xi)$$

$$+ 2\pi x_0^2 a \chi_S \xi \sum_{n=1}^{\infty} (2n+1) \left\{ c_n I_2(n,n) + d_n \left[\frac{n+1}{x_i} I_1(n,n-1) + \frac{n}{x_i} I_1(n+1,n) - I_2(n+1,n-1) \right] \right\}$$

$$I_2(n,m) \equiv \int_0^1 x^2 j_n(x_i x) j_m(x_S x) dx$$

$$I_1(n,m) \equiv \int_0^1 x j_n(x_i x) j_m(x_S x) dx$$

$$I_2(n,n) \equiv \frac{1}{x_i^2 - x_S^2} \left[x_S j_n(x_i) j_{n-1}(x_S) - x_i j_{n-1}(x_i) j_n(x_S) \right]$$

$$x_i = pa$$

$$x_S = k_S a$$

$$k_S = \omega\sqrt{\varepsilon_S \mu_0}$$

$$k_0 = \omega\sqrt{\varepsilon_0 \mu_0}$$