

Refraction and light transport in turbid colloids: Is the Poynting vector ill defined?

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ETOPIM 8

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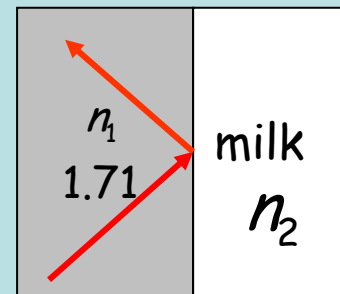
Motivation 1



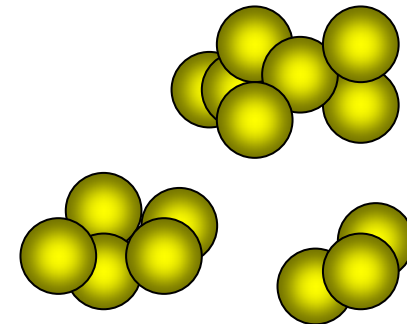
refractive index of milk

critical angle

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



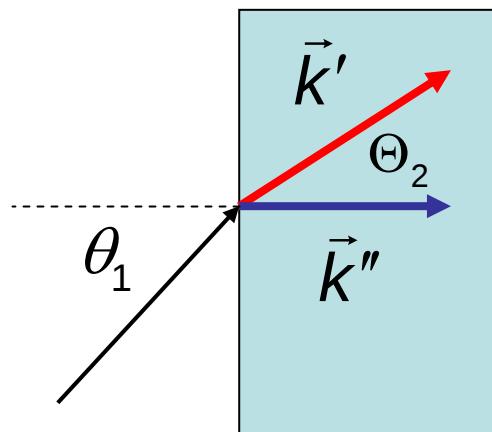
...but is white...and turbid...



Motivation 2

Absorption

$$\vec{k} = \vec{k}' + i\vec{k}''$$



Inhomogeneous wave

$$\vec{S} = \vec{E} \times \vec{H}$$

$$\vec{H} = \frac{\vec{B}}{\mu}$$

$$\vec{S} = \vec{E} \times \frac{\vec{B}}{\mu_0} \quad \rightarrow \quad \frac{\vec{E} \cdot \vec{J}^{ind}}{Q} > 0$$

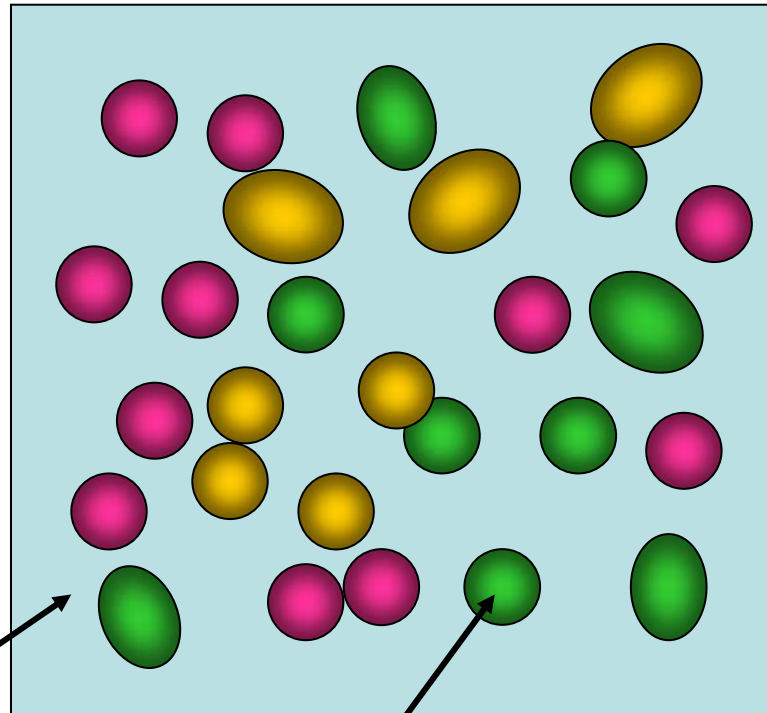
$$\vec{k}' \cdot \vec{k}'' > 0$$

**negative refraction
impossible**

Colloid



Inhomogeneous phase
dispersed within a
homogeneous one



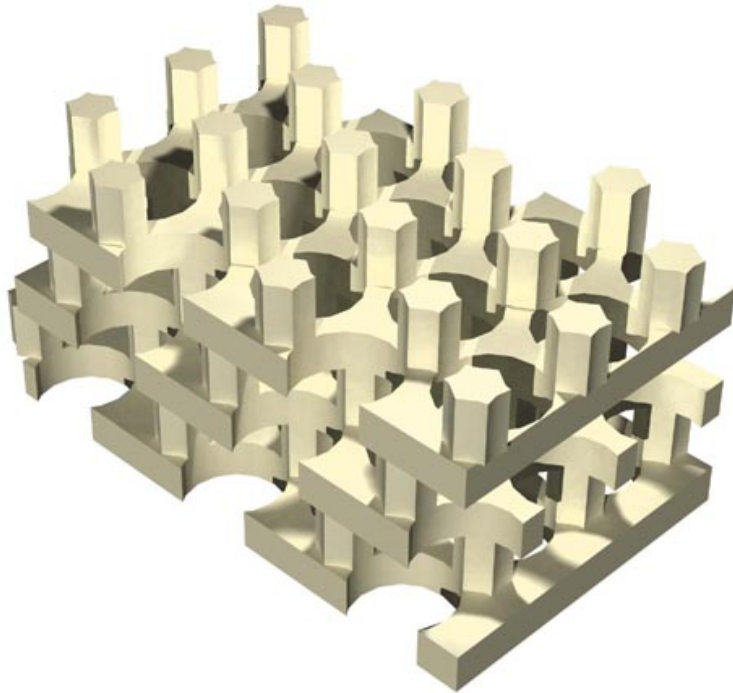
homogeneous phase

colloidal particles

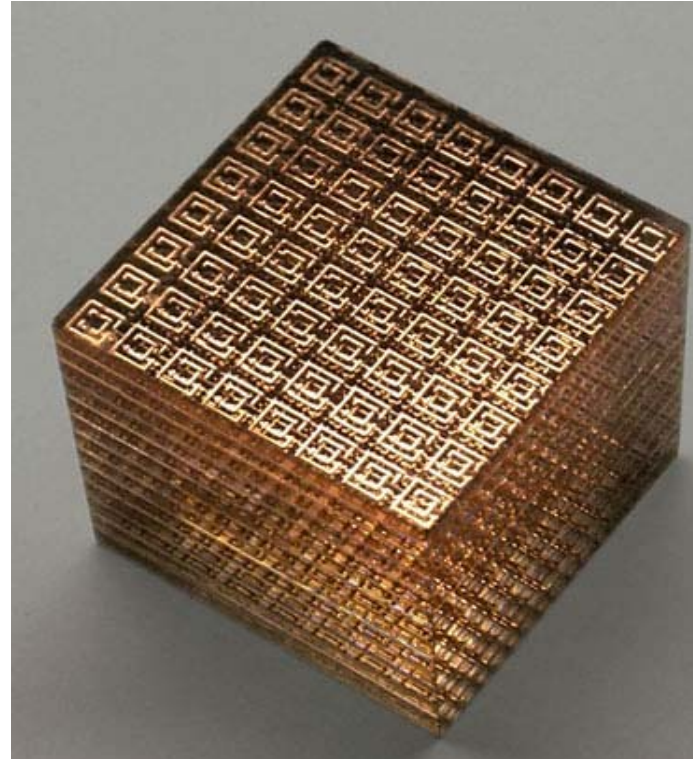
Colloidal systems

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

“Ordered” colloids

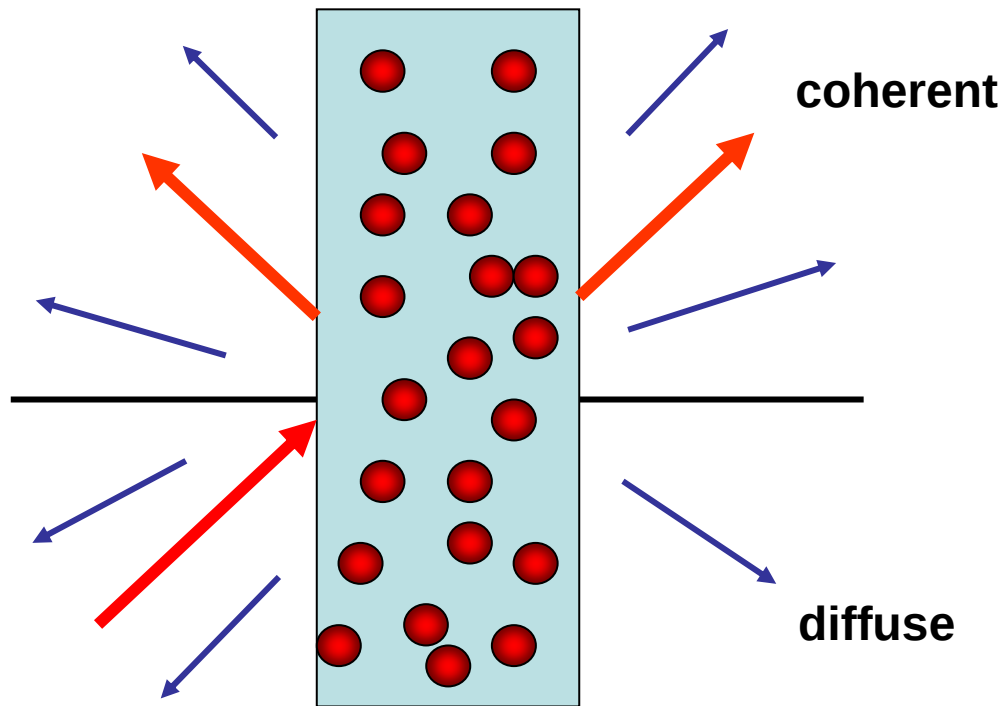


Photonic crystals



Metamaterials

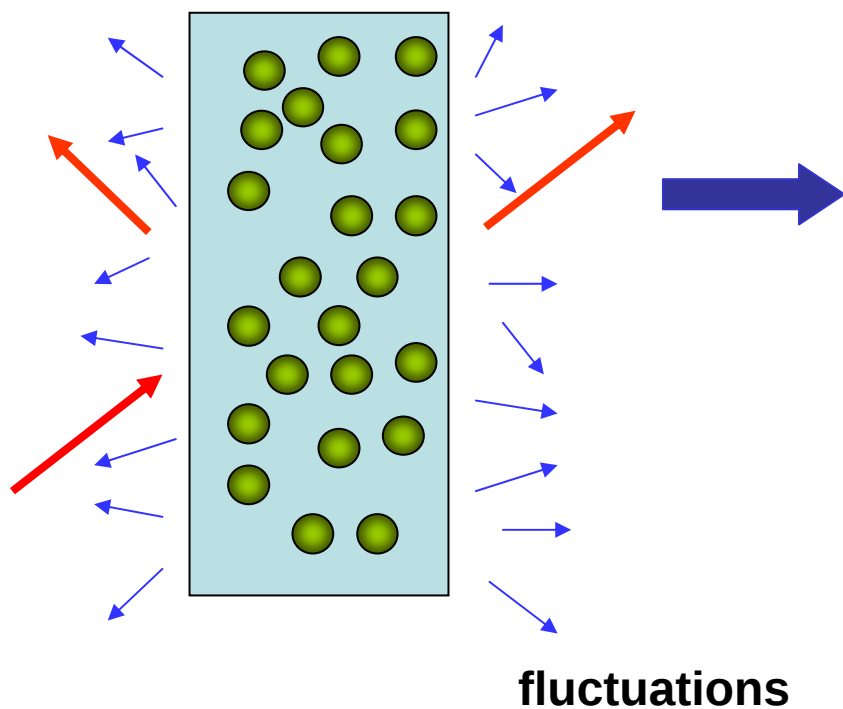
Light scattering



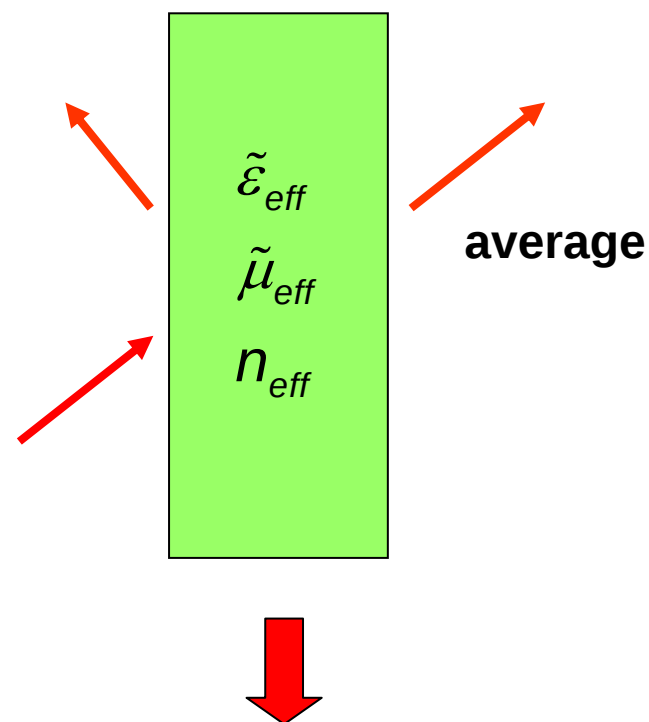
Effective medium



colloid



effective medium ?



Continuum Electrodynamics

Our result

IN TURBID COLLOIDAL SYSTEMS THE EFFECTIVE MEDIUM **EXISTS**
BUT IT IS **NONLOCAL**

the probability density is homogeneous

$$\langle \vec{J}_{ind}(\vec{r}; \omega) \rangle = \int \vec{\sigma}_{eff}(|\vec{r} - \vec{r}'|; \omega) \langle \vec{E}(\vec{r}', \omega) \rangle d^3r'$$

↑
total

Spatial dispersion

$$\langle \vec{J} \rangle^{ind}(\vec{k}, \omega) = \vec{\sigma}_{eff}(\vec{k}, \omega) \cdot \langle \vec{E} \rangle(\vec{k}, \omega)$$

↑

. Phys. Rev. 75 (2007) 184202 [1-19].

LT scheme



Probability density is homogeneous and isotropic

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

generalized effective nonlocal dielectric function

$$\hat{k} \equiv \frac{\vec{k}}{k}$$

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

$\varepsilon_{eff}^L(k, \omega)$	$\varepsilon_{eff}^T(k, \omega)$
----------------------------------	----------------------------------

$\varepsilon \mu$

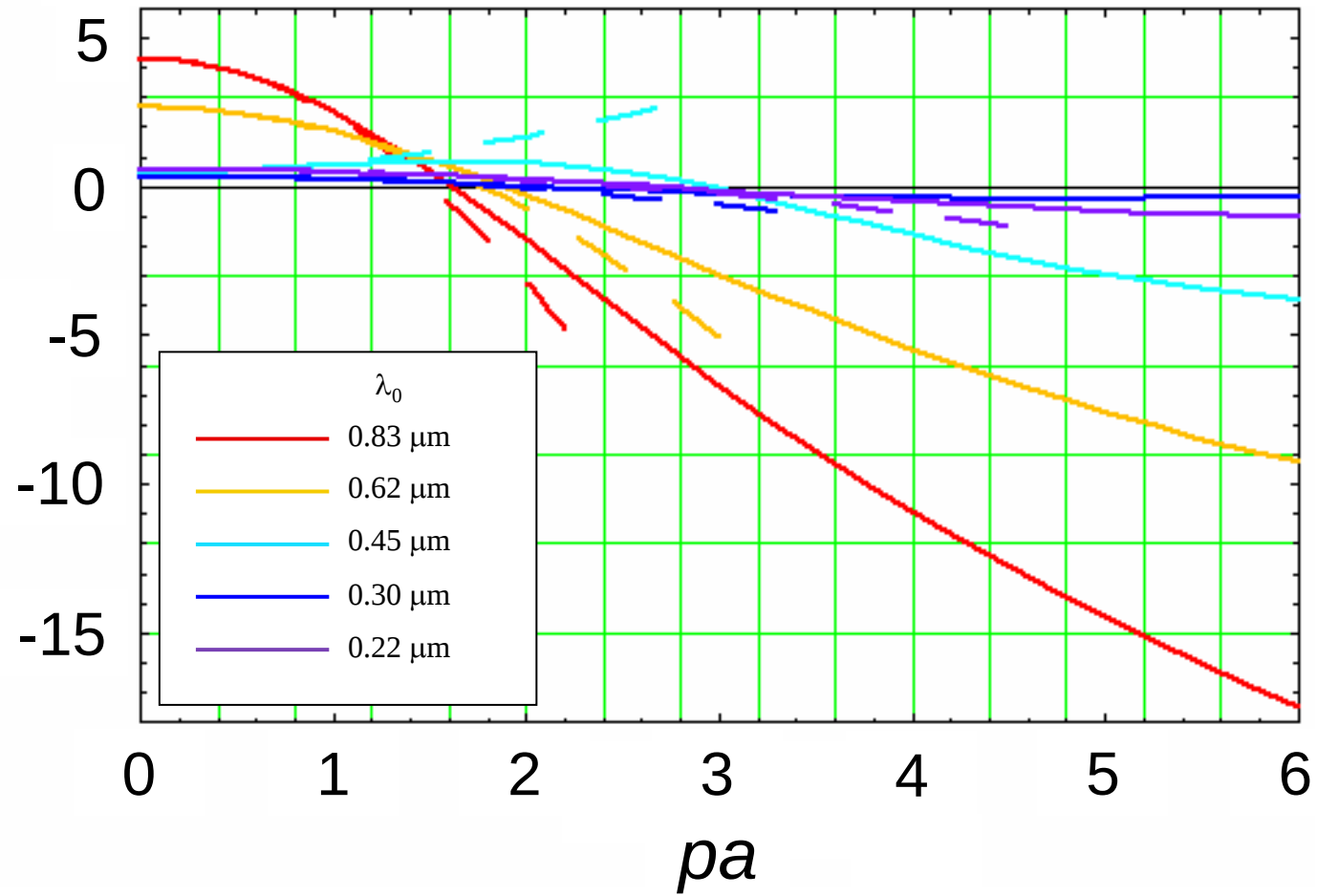
Long wavelength ($ka \rightarrow 0$)

“local limit”

$$\varepsilon^L(k, \omega) = \varepsilon^{[0]}(\omega) + \varepsilon^{L[2]}(\omega) \frac{k^2}{k_0^2} + \dots \quad \varepsilon^T(k, \omega) = \varepsilon^{[0]}(\omega) + \varepsilon^{T[2]}(\omega) \frac{k^2}{k_0^2} + \dots$$

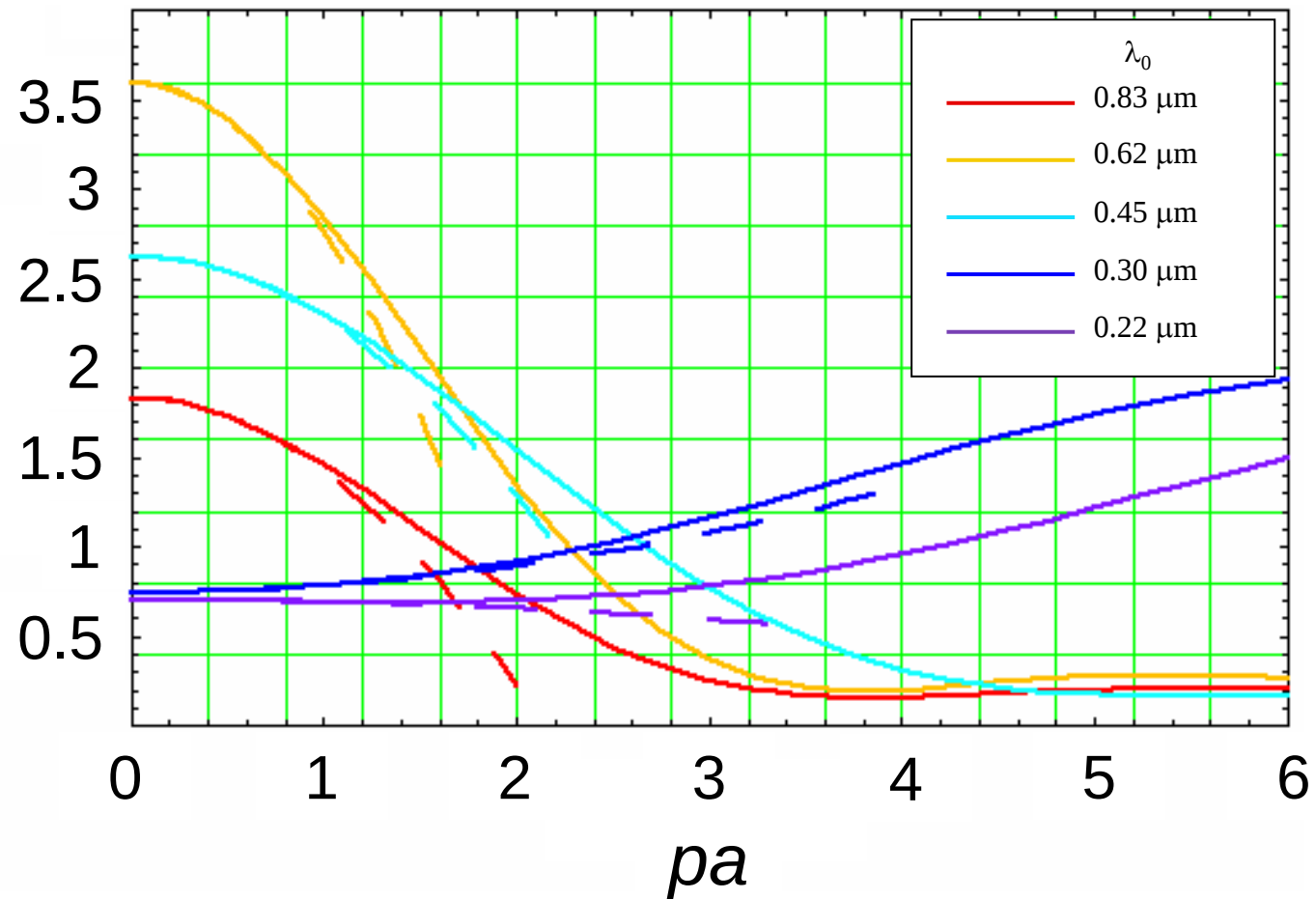
$$\frac{\text{Re}[\varepsilon^T(p; \omega)] - 1}{f}$$

Ag (radius=0.1 μm)



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

Ag (radius=0.1 μm)



$$n_{\text{eff}}(\omega)$$

Phys. Rev. 75 (2007) 184202 [1-19].

Poynting Theorem

$$\nabla \cdot \frac{1}{2} \text{Re} \left(\vec{E} \times \frac{\vec{B}^*}{\mu_0} \right) = \frac{1}{2} \text{Re} \left(\frac{\vec{B}}{\mu_0} \cdot (\nabla \times \vec{E}^*) - \vec{E} \cdot \left(\nabla \times \frac{\vec{B}^*}{\mu_0} \right) \right) \quad \text{Math}$$

$$\text{Maxwell} \quad = \frac{1}{2} \text{Re} \left(-\frac{\vec{B}}{\mu_0} \cdot \frac{\partial \vec{B}^*}{\partial t} - \vec{E} \cdot (\vec{J}^{ext*} + \vec{J}^{ind*}) - \epsilon_0 \vec{E} \cdot \frac{\partial \vec{E}^*}{\partial t} \right)$$

$$\nabla \cdot \frac{1}{2} \text{Re} \left(\vec{E} \times \frac{\vec{B}^*}{\mu_0} \right) + \text{Re} \frac{\partial}{\partial t} \left(\frac{\epsilon_0}{2} |\vec{E}|^2 + \frac{|\vec{B}|^2}{2\mu_0} \right) = \underbrace{-\frac{1}{2} \text{Re}(\vec{J}^{ext} \cdot \vec{E}^*)}_{W_{ext}} - \underbrace{\frac{1}{2} \text{Re}(\vec{J}^{ind} \cdot \vec{E}^*)}_Q$$

Plane wave $W_{ext} \rightarrow Q$

WORK

$$\vec{J}^{ind} = i\omega\epsilon_0(n^2 - 1)\vec{E} \quad \text{free-propagating mode}$$

$$W_{ind} = \frac{1}{2}\text{Re}(\vec{J}^{ind} \cdot \vec{E}^*) = \frac{1}{2}\text{Re}[i\omega\epsilon_0(n^2 - 1)\vec{E} \cdot \vec{E}^*]$$

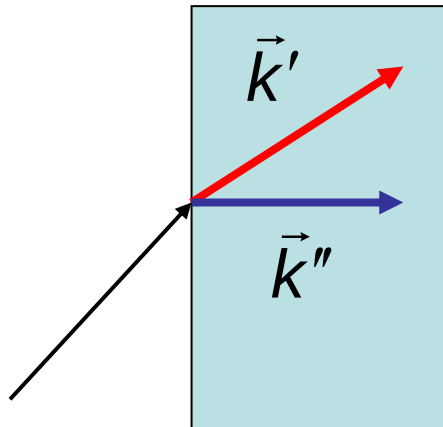
$$= \frac{\omega}{2}\epsilon_0 \underbrace{\text{Im} n^2}_Q |\vec{E}|^2 \quad \longrightarrow \quad \text{Im} n^2 > 0$$

dispersion relation

$$k^2 = \vec{k} \cdot \vec{k} = (\vec{k}' + i\vec{k}'')^2 = k_0^2 n^2$$

$$k'^2 - k''^2 = k_0^2 \text{Re} n^2$$

$$2\vec{k}' \cdot \vec{k}'' = k_0^2 \text{Im} n^2 > 0$$



Inhomogeneous wave

Negative refraction is impossible



Science **316**, 430 (2007)

Negative Refraction at Visible Frequencies

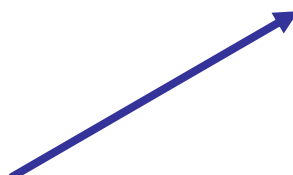
Henri J. Lezec,^{1,2*†} Jennifer A. Dionne,^{1*} Harry A. Atwater¹

Nanofabricated photonic materials offer opportunities for crafting the propagation and dispersion of light in matter. We demonstrate an experimental realization of a two-dimensional negative-index material in the blue-green region of the visible spectrum, substantiated by direct geometric visualization of negative refraction. Negative indices were achieved with the use of an ultrathin Au-Si₃N₄-Ag waveguide sustaining a surface plasmon polariton mode with antiparallel group and phase velocities. All-angle negative refraction was observed at the interface between this bimetal waveguide and a conventional Ag-Si₃N₄-Ag slot waveguide. The results may enable the development of practical negative index optical designs in the visible regime.

Nonlocal calculation

$$\frac{1}{2} \text{Re}(\vec{J}^{ind} \cdot \vec{E}^*)$$


$$\nabla \cdot \frac{1}{2} \text{Re} \left(\langle \vec{E} \rangle \times \frac{\langle \vec{B} \rangle^*}{\mu_0} \right) + \frac{\partial}{\partial t} \left(\frac{\epsilon_0}{2} |\langle E \rangle|^2 + \frac{1}{2\mu_0} |\langle B \rangle|^2 \right) = -W_{ext} - W_{ind}$$

$$\vec{J}^{ind}(\vec{r}; t) = \int dt' \int d^3r' \vec{\sigma}(\vec{r} - \vec{r}'; t - t') \cdot \vec{E}(\vec{r}'; t')$$


quasi-monochromatic wave

$$\vec{E}(\vec{r}, t) = \text{Re} \vec{E}_0(\vec{r}, t) \exp[i\vec{k} \cdot \vec{r} - \omega t] \quad |\nabla E_{0\alpha}| \ll k E_0 \quad \left| \frac{\partial E_{0\alpha}}{\partial t} \right| \ll \omega E_0$$


$$\vec{E}_0(\vec{r}', t') = \vec{E}_0(\vec{r}, t) + [(\vec{r}' - \vec{r}) \cdot \nabla] \vec{E}_0 + (t' - t) \frac{\partial \vec{E}_0}{\partial t} + \dots$$

Induced current

$$J_{\alpha}^{ind}(\vec{r}, t) = \exp[i(\vec{k} \cdot \vec{r} - \omega t)] \left\{ -i\omega \left(\varepsilon_{\alpha\beta}(\vec{k}, \omega) - \varepsilon_0 \delta_{\alpha\beta} \right) E_{0\beta}(r, t) \right. \\ \left. - \omega \frac{\partial \varepsilon_{\alpha\beta}(\vec{k}, \omega)}{\partial k_{\gamma}} \frac{\partial E_{0\beta}(\vec{r}, t)}{\partial x_{\gamma}} + \left[(\varepsilon_{\alpha\beta} - \varepsilon_0 \delta_{\alpha\beta}) + \omega \frac{\partial \varepsilon_{\alpha\beta}(\vec{k}, \omega)}{\partial \omega} \right] \frac{\partial E_{0\beta}(\vec{r}, t)}{\partial t} \right\}$$

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

$$\frac{1}{2} \text{Re}(\vec{J}^{ind} \cdot \vec{E}^*)$$

long wavelength $ka \rightarrow 0$

“local”

$$\varepsilon^L(k, \omega) = \underline{\varepsilon^{[0]}(\omega)} + \varepsilon^{L[2]}(\omega) \frac{k^2}{k_0^2} + \dots$$

$$\varepsilon^T(k, \omega) = \underline{\varepsilon^{[0]}(\omega)} + \varepsilon^{T[2]}(\omega) \frac{k^2}{k_0^2} + \dots$$

$$\text{Im } \varepsilon^L \ll \text{Re } \varepsilon^L$$

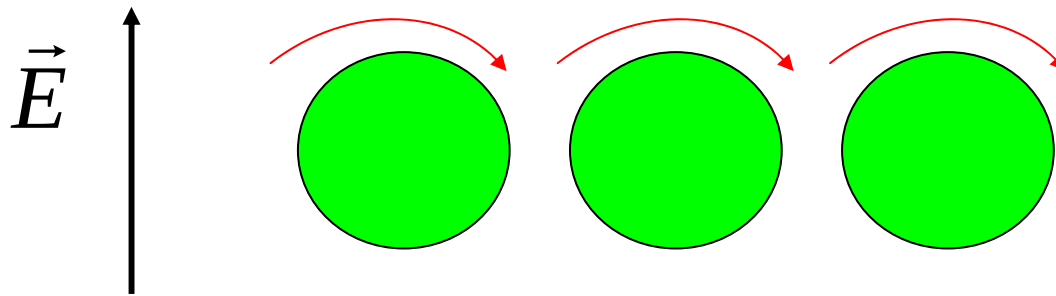
$$\text{Im } \varepsilon^T \ll \text{Re } \varepsilon^T$$

Work on the induced currents

$$W_{ind} = \frac{\omega}{2} \left[\text{Im } \varepsilon^L(k, \omega) |E_0^L|^2 + \text{Im } \varepsilon^T(k, \omega) |E_0^T|^2 \right]$$

$$- \nabla \cdot \frac{1}{2} \text{Re} \left[\frac{1}{\varepsilon_0 \mu_0} (\vec{E}_0 \times \vec{B}_0^*) \varepsilon^{T[2]*}(\omega) + \frac{\varepsilon^{L[2]}(\omega)}{\omega \varepsilon_0 \mu_0} (\vec{k} |E_0^L|^2 + k E_0^{L*} \vec{E}_0^T) \right]$$

$$+ \frac{1}{2} \frac{\partial}{\partial t} \text{Re} \left[\frac{\partial \omega \varepsilon^{[0]}(\omega)}{\partial \omega} |E_0|^2 + \frac{k^2}{\varepsilon_0 \mu_0} \left(\frac{\partial}{\partial \omega} \frac{\varepsilon^{L[2]}(\omega)}{\omega} |E_0^L|^2 + \frac{\partial}{\partial \omega} \frac{\varepsilon^{T[2]}(\omega)}{\omega} |E_0^T|^2 \right) - \varepsilon_0 |E_0|^2 \right]$$



$$\nabla \cdot \frac{1}{2} \text{Re} \left(\langle \vec{E} \rangle \times \frac{\langle \vec{B} \rangle^*}{\mu_0} \right) + \frac{\partial}{\partial t} \left(\frac{\varepsilon_0}{2} |\langle E \rangle|^2 + \frac{1}{2\mu_0} |\langle B \rangle|^2 \right) = -W_{ext} - W_{ind}$$

Energy theorem

Transverse waves

$$\nabla \cdot \frac{1}{2} \text{Re} \left[\frac{1}{\mu_0} (\vec{E}_0 \times \vec{B}_0^*) (1 - \varepsilon^{T[2]*}(\omega) / \varepsilon_0) \right] + \frac{1}{2} \frac{\partial}{\partial \omega} \text{Re} \left[\frac{1}{\mu_0} |B_0|^2 + \frac{\partial \omega \varepsilon^{[0]}(\omega)}{\partial \omega} |E_0|^2 \right. \\ \left. + \frac{k^2}{\varepsilon_0 \mu_0} \frac{\partial}{\partial \omega} \frac{\varepsilon^{T[2]}(\omega)}{\omega} |E_0|^2 \right] = -W_{ext} - \frac{\omega}{2} \text{Im} \left[\varepsilon^{[0]}(\omega) + \frac{k^2 \varepsilon^{T[2]}(\omega)}{\omega^2 \varepsilon_0 \mu_0} \right] |E_0|^2$$

$$\vec{S}_{trans} = \frac{1}{2} \text{Re} \left[\frac{1}{\mu_0} (\vec{E}_0 \times \vec{B}_0^*) (1 - \varepsilon^{T[2]*}(\omega) / \varepsilon_0) \right]$$

$\epsilon \mu$ scheme

$$\vec{J}^{ind}(\vec{r}, t) = \frac{\partial \vec{P}(\vec{r}, t)}{\partial t} + \nabla \times \vec{M}(\vec{r}, t)$$

NOT UNIQUE

quasi-monochromatic

$$\vec{J}^{ind}(\vec{r}, t) = \vec{J}_0^{ind}(\vec{r}, t) \exp \left[i(\vec{k} \cdot \vec{r} - \omega t) \right]$$

$$\vec{P}(\vec{r}, t) = \vec{P}_0(\vec{r}, t) \exp \left[i(\vec{k} \cdot \vec{r} - \omega t) \right]$$

$$\vec{M}(\vec{r}, t) = \vec{M}_0(\vec{r}, t) \exp \left[i(\vec{k} \cdot \vec{r} - \omega t) \right]$$

Response

$$\vec{J}^{ind}(\vec{r}, t) = \frac{\partial \vec{P}(\vec{r}, t)}{\partial t} + \nabla \times \vec{M}(\vec{r}, t)$$

$$\vec{P}_0(\vec{r}, t) = \left[\left(\varepsilon^l(k, \omega) - \varepsilon_0 \right) \vec{P}^L + \left(\varepsilon^t(k, \omega) - \varepsilon_0 \right) \vec{P}^T \right] \cdot \vec{E}_0(\vec{r}, t)$$

$$\vec{M}_0(\vec{r}, t) = \left[\frac{1}{\mu_0} - \frac{1}{\mu(k, \omega)} \right] \vec{B}_0(\vec{r}, t)$$

CHOICE I

$$\varepsilon^t(k, \omega) = \varepsilon^t(0, \omega)$$

$$\mu_I(k, \omega)$$

CHOICE II

$$\varepsilon^t(k, \omega) = \varepsilon^l(k, \omega)$$

$$\mu_{II}(k, \omega)$$

Poynting vector in $\epsilon\mu$

long wavelength limit

$$\left[\frac{1}{\mu_0} - \frac{1}{\mu(0, \omega)} \right] \vec{B}_0 = \left[\frac{1}{\mu_0} - \frac{1}{\mu(0, \omega)} \right] \frac{1}{\omega} \vec{k} \times \vec{E}_0$$

$$\vec{S}_{trans} = \frac{1}{2} \text{Re} \left[\vec{E}_0 \times \left(\underbrace{\frac{\vec{B}_0^*}{\mu_0} - \vec{M}_0^*}_{\vec{H}_0} \right) - \vec{E}_0 \times \frac{\vec{B}_0^*}{\mu_0} \frac{\epsilon^{t[2]*}(\omega)}{\epsilon_0} \right]$$

CHOICE I

$$\epsilon^{t[2]*}(\omega) = 0$$

$$\mu_I(0, \omega)$$

CHOICE II

$$\epsilon^{t[2]*}(\omega) = \epsilon^{l[2]*}(\omega) = \epsilon^{L[2]}(\omega)$$

$$\mu_{II}(0, \omega)$$

Poynting vector in LT

The correct expression of the Poynting vector

$$\vec{S}_{trans} = \frac{1}{2} \text{Re} \left[\frac{1}{\mu_0} \left(\vec{E}_0 \times \vec{B}_0^* \right) \left(1 - \varepsilon^{T[2]*}(\omega) / \varepsilon_0 \right) \right]$$


$\varepsilon^{T[2]*}(\omega)$ is NOT a non-local correction

This is the LOCAL limit

Conclusions

- (i) We used the formalism developed for the treatment of the non-local effective medium associated to turbid colloids to find a general expression for the energy theorem, in the long wavelength limit, in terms of parameters associated to the non-local response of the system.
- (ii) We found an explicit expression for the Poynting vector in terms of these non-local parameters and show that there are many ways of writing it in terms of the electric permittivity ϵ and the magnetic permeability μ , depending on the choice taken to define them.
- (iii) This approach can be extended to finite wave vectors and non negligible dissipation as well as energy transport for free-propagating modes