





Spectral multipole theory for nano-antennas

(Directivity, Ideal absorption and unitarity)

Spectral Theory of Novel Materials : April 2016

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Maxwell's equations in inhomogeneous media

Time-harmonic Maxwell's equations : Inhomogeneous medium

$$\nabla \times \frac{1}{\mu(\vec{x})} \nabla \times \vec{\mathbf{E}}(\vec{x}) - \varepsilon(\vec{x}) \left(\frac{\omega}{c}\right)^2 \vec{\mathbf{E}}(\vec{x}) = i\mu_0 \omega \vec{\mathbf{j}}_s(\vec{x})$$

Green's function : Inhomogeneous medium

$$\nabla \times \frac{1}{\mu(\vec{x})} \nabla \times \vec{\mathbf{G}}(\vec{x}, \vec{x}') - \varepsilon(\vec{x}) \left(\frac{\omega}{c}\right)^2 \vec{\mathbf{G}}(\vec{x}, \vec{x}') = \vec{\mathbf{I}}\delta(\vec{x} - \vec{x}')$$

Total solution to the wave equation !

$$\vec{\mathbf{E}}(\vec{\mathbf{x}}) = i\mu_0\omega \int \vec{\mathbf{G}}(\vec{\mathbf{x}},\vec{\mathbf{x}}') \cdot \vec{\mathbf{j}}_s(\vec{\mathbf{x}}')$$

Physical problems can often be formulated as a multiple-scattering problem

$$\vec{\mathbf{E}}_{\text{scat}}$$

$$\vec{\mathbf{E}}_{\text{scat}}$$

$$\vec{\mathbf{E}}_{\text{scat}}$$

$$\vec{\mathbf{E}}_{\text{inc}}(\vec{\mathbf{x}}) = i\mu_0\omega \int \vec{\mathbf{G}}_0(\vec{\mathbf{x}}, \vec{\mathbf{x}}') \cdot \vec{\mathbf{j}}_s(\vec{\mathbf{x}}')$$

Meso-scopic approximation : permittivity, $\varepsilon(\vec{x})$ is piecewise constant and $\mu(\vec{x})=1$

$$\nabla \times \frac{1}{\mu(\vec{x})} \nabla \times \vec{\mathbf{G}}(\vec{x}, \vec{x}') - \varepsilon(\vec{x}) \left(\frac{\omega}{c}\right)^2 \vec{\mathbf{G}}(\vec{x}, \vec{x}') = \vec{\mathbf{I}}\delta(\vec{x} - \vec{x}')$$

T-matrix simplifies the formulation of the multiple-scattering problem

$$\vec{\mathbf{G}}(\vec{x},\vec{x}') = \vec{\mathbf{G}}_{\mathbf{0}}(\vec{x}-\vec{x}') + \vec{\mathbf{G}}_{\mathbf{0}}(\vec{x}-\vec{x}'') \cdot \vec{\mathbf{T}}(\vec{x}'',\vec{x}''') \cdot \vec{\mathbf{G}}_{\mathbf{0}}(\vec{x}'''-\vec{x}')$$

$$\vec{\mathbf{E}}_{inc}(\vec{x}) = i\mu_0 \omega \int \vec{\mathbf{G}}_0(\vec{x}, \vec{x}') \cdot \vec{J}_5(\vec{x}')$$

$$\vec{\mathbf{E}}_{inc}(\vec{x}) = \vec{\mathbf{E}}_{inc}(\vec{x}) + \vec{\mathbf{G}}_0(\vec{x} - \vec{x}'') \cdot \vec{\mathbf{T}}(\vec{x}'', \vec{x}''') \cdot \vec{\mathbf{E}}_{inc}(\vec{x}''')$$

$$\underbrace{\vec{\mathbf{E}}_{scat}(\vec{x})}_{\vec{\mathbf{E}}_{scat}(\vec{x})}$$

Green's function of a homogeneous media

Scalar case

$$\Delta G_0(\vec{x} - \vec{x}') + k^2 G_0(\vec{x} - \vec{x}') = -\delta(\vec{x} - \vec{x}')$$

$$G_0(\vec{x} - \vec{x}') = \frac{e^{ik|\vec{x} - \vec{x}'|}}{4\pi |\vec{x} - \vec{x}'|} = \frac{i}{4\pi} h_0(|\vec{x} - \vec{x}'|)$$

Vector case

$$\nabla \times \nabla \times \vec{\mathbf{G}}(\vec{x} - \vec{x}') - k^2 \vec{\mathbf{G}}(\vec{x} - \vec{x}') = \vec{\mathbf{I}}\delta(\vec{x} - \vec{x}')$$
$$\vec{\mathbf{G}}_0(\vec{r}) = -\frac{e^{ikr}}{4\pi k^2 r^3} \text{P.V.}\left\{(1 - ikr - k^2 r^2)(\vec{\mathbf{I}} - \hat{r}\hat{r}) - 2(1 - ikr)\hat{r}\hat{r}\right\} - \frac{\vec{\mathbf{I}}}{3k^2}\delta(\vec{r})$$
$$\vec{r} \equiv \vec{x} - \vec{x}'$$

Homogenous scalar Helmholtz equation with constant permittivity



$$\Delta \psi + k^2 \psi = 0$$

$$\underbrace{\frac{1}{r} \frac{\partial^2 (r\psi)}{\partial r^2} - \frac{\overline{L}^2}{r^2} \psi - k^2 \psi = 0}_{k^2 = \varepsilon \left(\frac{\omega}{c}\right)^2}$$

$$\underline{r}(r, \theta, \phi) = \psi_r(r) Y_{n,m}(\theta, \phi) \qquad \overline{L}^2 Y_{n,m}(\theta, \phi) = n(n+1) Y_{n,m}(\theta, \phi)$$

$$Y_{n,m}(\theta, \phi) = \overline{P}_n^m(\cos \theta) \exp(im\phi) \qquad \begin{cases} n = 0, 1, 2...\infty \\ m = -n, ..., n \end{cases}$$

Spherical Bessel function equation

Change of variables:

$$j_n(kr) \equiv \psi_r(r) \equiv \frac{Z(r)}{(kr)^{1/2}}$$

$$r^{2} \frac{d^{2}Z}{dr^{2}} + r \frac{dZ}{dr} + \left[k^{2}r^{2} - \left(n + \frac{1}{2}\right)^{2}\right]Z = 0$$

Linearly independent solutions

$$\Delta \psi + k^2 \psi = 0$$





Outgoing spherical Hankel functions (+) $h_n^{(+)}(x) = j_n(x) + iy_n(x)$ $h_0^{(+)}(x) = -\frac{i}{x}e^{ix}$ $h_1^{(+)}(x) = -e^{ix}\left(\frac{1}{x} + \frac{i}{x^2}\right)$ Spherical Neumann functions (2) $y_n(x)$



Incoming spherical Hankel functions (-) $h_n^{(-)}(x) = j_n(x) - iy_n(x)$ $h_0^{(-)}(x) = \frac{i}{x}e^{-ix}$ $h_1^{(-)}(x) = -e^{-ix}\left(\frac{1}{x} - \frac{i}{x^2}\right)$

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Partial wave basis (scalar waves)





Outgoing "partial" wave :

$$\varphi_{n,m}^{(+)}(k\mathbf{r}) = h_n^{(+)}(kr)Y_{n,m}(\theta,\phi)$$

Vector partial waves

$$\nabla \times \nabla \times \vec{\mathbf{E}} - k^2 \, \vec{\mathbf{E}} = 0$$

$$\vec{\mathbf{M}}_{n,m}^{(1)}(k\mathbf{r}) = j_n(kr)\vec{\mathbf{X}}_{n,m}(\theta,\phi)$$
Ricatti-Bessel functions
$$\vec{\mathbf{N}}_{n,m}^{(1)}(k\mathbf{r}) = \frac{1}{kr} \Big[j_n(kr)\sqrt{n(n+1)}\vec{\mathbf{Y}}_{n,m}(\theta,\phi) \\ + \psi'_n(x)\vec{\mathbf{Z}}_{n,m}(\theta,\phi) \Big]$$

$$n=1,2,...,\infty \quad -n < m < n$$

$$\nabla \cdot \vec{\mathbf{M}}_{n,m} = \nabla \cdot \vec{\mathbf{N}}_{n,m} = 0 \qquad \nabla \times \vec{\mathbf{M}}_{n,m} = k \vec{\mathbf{N}}_{n,m} \quad \nabla \times \vec{\mathbf{N}}_{n,m} = k \vec{\mathbf{M}}_{n,m}$$

Vector spherical harmonics

$$\vec{\mathbf{Y}}_{n,m}(\theta,\phi) = \hat{\mathbf{r}}Y_{n,m} \qquad n = 0,...,\infty \qquad m = -n,...,n$$
$$\vec{\mathbf{X}}_{n,m}(\theta,\phi) = e^{im\phi} \begin{bmatrix} \hat{\mathbf{\theta}}i\overline{u}_n^m - \hat{\mathbf{\phi}}\overline{s}_n^m \end{bmatrix} \qquad n = 1,...,\infty \qquad m = -n,...,n$$
$$\vec{\mathbf{Z}}_{n,m}(\theta,\phi) = e^{im\phi} \begin{bmatrix} \hat{\mathbf{\theta}}\overline{s}_n^m + \hat{\mathbf{\phi}}i\overline{u}_n^m \end{bmatrix} \qquad n = 1,...,\infty \qquad m = -n,...,n$$

$$\overline{u}_{n,m}(\theta,\phi) = \frac{1}{\sqrt{n(n+1)}} \frac{m}{\sin\theta} \overline{P}_n^m(\cos\theta)$$
$$\overline{s}_{n,m}(\theta,\phi) = \frac{1}{\sqrt{n(n+1)}} \frac{d}{d\theta} \overline{P}_n^m(\cos\theta)$$

Partial wave expansions for Green's functions

$$G_0(\vec{x} - \vec{x}') = \frac{e^{ik|\vec{x} - \vec{x}'|}}{4\pi|\vec{x} - \vec{x}'|} = ik \sum_{n=0}^{\infty} (-)^m \varphi_{n,m}^{(+)}(k\vec{r}_{>})\varphi_{n,-m}^{(1)}(k\vec{r}_{<})$$

$$\begin{aligned} \vec{\mathbf{G}}_{0}(\vec{r}) &= -\frac{e^{ikr}}{4\pi k^{2}r^{3}} \operatorname{P.V.}\left\{ (1 - ikr - k^{2}r^{2})(\vec{\mathbf{I}} - \hat{r}\hat{r}) - 2(1 - ikr)\hat{r}\hat{r}\right\} - \frac{\vec{\mathbf{I}}}{3k^{2}}\delta(\vec{r}) \\ &= ik\sum_{n=0}^{\infty} (-)^{m} \left\{ \overrightarrow{\mathbf{M}}_{n,m}^{(+)}(k\vec{r}_{>})\overrightarrow{\mathbf{M}}_{n,-m}^{(1)}(k\vec{r}_{<}) + \overrightarrow{\mathbf{N}}_{n,m}^{(+)}(k\vec{r}_{>})\mathbf{N}_{n,-m}^{(1)}(k\vec{r}_{<}) \right\} - \frac{\hat{r}\hat{r}}{k^{2}}\delta(\vec{r}) \end{aligned}$$

T-matrix of a scatterer (multipole representation) "Seeing is believing and all we see is scattered light" J.C. Stover



Scattering by a sphere Lorenz(1890)-Mie(1908)-Debye(1909) theory





Gustav Mie (1868-1957)



Ludvig Lorenz (1829–91) **"Light scattering and reflection by a transparent sphere (surface)"** in Oeuvres scientifiques de **L. Lorenz**. revues et annotées par H. Valentiner. Tome Premier, Libraire Lehmann & Stage, Copenhague, 1898, p 403-529.

T-matrix of a sphere

$$f_{n,m}^{(e)} = T_n^{(e)} e_{n,m}^{(e)}$$

$$f_p^{(h)} = T_n^{(h)} e_{n,m}^{(h)}$$

$$\begin{bmatrix} f_p^{(h)} \\ f_p^{(e)} \end{bmatrix} = \begin{bmatrix} T_1^{(h)} & \cdots & 0 \\ 0 & \cdots & T_n^{(h)} \end{bmatrix} \begin{bmatrix} 0 & \cdots & 0 \\ 0 & \cdots & 0 \\ 0 & \cdots & 0 \end{bmatrix} \begin{bmatrix} e_p^{(h)} \\ 0 & \cdots & 0 \\ 0 & \cdots & 0 \end{bmatrix} \begin{bmatrix} e_p^{(h)} \\ e_p^{(e)} \end{bmatrix} = \begin{bmatrix} T_1^{(h)} \\ e_p^{(e)} \end{bmatrix} = \begin{bmatrix} T_1^{(h)} \\ e_p^{(e)} \end{bmatrix}$$

Lorenz-Mie Coefficients : a_n , b_n $T_n^{(e)} = -a_n$ $T_n^{(h)} = -b_n$

Traditional "Mie" form

$$a_{n} = \frac{\rho_{s}\psi_{n}(k_{s}R)\psi_{n}'(kR) - (\mu_{s}/\mu)\psi_{n}(kR)\psi_{n}'(k_{s}R)}{\rho_{s}\psi_{n}(k_{s}R)\xi_{n}'(kR) - (\mu_{s}/\mu)\xi_{n}(kR)\psi_{n}'(k_{s}R)}$$
$$b_{n} = \frac{\rho_{s}\psi_{n}(kR)\psi_{n}'(k_{s}R) - (\mu_{s}/\mu)\psi_{n}(k_{s}R)\psi_{n}'(kR)}{\rho_{s}\xi_{n}(kR)\psi_{n}'(k_{s}R) - (\mu_{s}/\mu)\psi_{n}(k_{s}R)\xi_{n}'(kR)}$$

 $\rho_s = \frac{k_s}{k} = \frac{N_s}{N}$

The way we write things is important !



Aggregate of *N* objects subject to an incident field :

$$e^{(j)} = a^{(j)} + \sum_{l=1, l \neq j}^{N} H^{(j,l)} f^{(l)}$$
$$f^{(j)} = T^{(j)} e^{(j)}$$

$$\vec{E}_{inc}$$

$$\begin{bmatrix} T^{(1)} \end{bmatrix}^{-1} & -H^{(1,2)} \dots & -H^{(1,N)} \\ -H^{(2,1)} & \begin{bmatrix} T^{(2)} \end{bmatrix}^{-1} \dots & -H^{(2,N)} \\ \vdots & \ddots & \vdots \\ -H^{(N,1)} & -H^{(N,1)} \dots & \begin{bmatrix} T^{(N)} \end{bmatrix}^{-1} \end{bmatrix} \begin{bmatrix} f^{(1)} \\ f^{(2)} \\ \vdots \\ f^{(N)} \end{bmatrix} = \begin{bmatrix} a^{(1)} \\ a^{(2)} \\ \vdots \\ a^{(N)} \end{bmatrix}$$

Matrix balancing



Matrix balanced equations are well conditioned for numerical inversion

$$\begin{bmatrix} \bar{T}^{(1)} \end{bmatrix}^{-1} & -\bar{H}^{(1,2)} & \dots & -\bar{H}^{(1,N)} \\ -\bar{H}^{(2,1)} & \left[\bar{T}^{(2)} \right]^{-1} & \dots & -\bar{H}^{(2,N)} \\ \vdots & & \ddots & \vdots \\ -\bar{H}^{(N,1)} & -\bar{H}^{(N,1)} & \dots & \left[\bar{T}^{(N)} \right]^{-1} \end{bmatrix} \begin{bmatrix} f^{(1)} \\ f^{(2)} \\ \vdots \\ f^{(N)} \end{bmatrix} = \begin{bmatrix} a^{(1)} \\ a^{(2)} \\ \vdots \\ f^{(N)} \end{bmatrix}$$

$$T_n^{(e)} = -\frac{j_n(kR)}{h_n^{(+)}(kR)} \frac{\frac{\varepsilon_s}{\varepsilon}\varphi_n(kR) - \varphi_n(k_sR)}{\frac{\varepsilon_s}{\varepsilon}\varphi_n^{(+)}(kR) - \varphi_n(k_sR)} \equiv \frac{j_n(kR)}{h_n^{(+)}(kR)} \overline{T}_n^{(e)}$$

$$\varphi_n = \frac{[zj_n(z)]'}{j_n(z)} \qquad \varphi_n^{(+)}(x) = \frac{\left[xh_n^{(+)}(x)\right]'}{h_n^{(+)}(x)}$$

B. Stout et al., J. Opt. Soc. Am. A, 25 (2008)



Decay rates and directivity of quantum emitters near nano-antennas

$$\vec{E}(\vec{r}) = \omega^2 \mu_0 \overline{\vec{G}}(\vec{r}, \vec{0}) \cdot \vec{p}$$
$$\langle P_e \rangle = \frac{\omega^3}{2} \mu_0 \text{Im}[\vec{p}^* \cdot \overline{\vec{G}}(\vec{0}, \vec{0}; \boldsymbol{\omega}) \cdot \vec{p}] \propto \Gamma_e \qquad \tau \propto \frac{1}{\Gamma_e}$$

Quasi-analytic partial wave formulation :

$$\frac{\Gamma_e}{\Gamma_0} = \frac{\langle P_e \rangle}{\langle P_{e,0} \rangle} = 1 + \frac{6\pi}{\operatorname{Re}\{k_b\}} \operatorname{Re}\left\{k_b \sum_{j,l=1}^N f^{\dagger} \cdot H^{(0,j)} \cdot T^{(j,l)} \cdot H^{(l,0)} \cdot f\right\}$$

$$\frac{\Gamma_r}{\Gamma_0} = \frac{\langle P_r \rangle}{\langle P_{r,0} \rangle} = 1 + 6\pi \operatorname{Re} \left\{ k_b \sum_{i,j,k,l=1}^N \left[T^{(j,i)} \cdot H^{(i,0)} \cdot f \right]^{\dagger} \cdot J^{(j,k)} \cdot T^{(k,l)} \cdot H^{(l,0)} \cdot f \right\} + 12\pi \operatorname{Re} \left[\sum_{j,k=1}^N \left[J^{(k,0)} \cdot f \right]^{\dagger} \cdot T^{(k,j)} \cdot H^{(j,0)} \cdot f \right]$$

Method : B.Stout, et al. J. Opt. Soc. Am. B, 28, doi:10.1364/JOSAB.28.001213 (2011)

Multipole theory can simulate (Optical) Antenna properties

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 Subwavelength field enhancements to efficiently excite quantum emitters





• Lifetime modification of quantum emitters



Nature Communications, 3, 962 (2012) Angewandte Chemie, 51, (2012) NANO Letters 11, (2011)

Directive emissions



Optical receiving antenna?

No equivalent in optics for co-axial to cables to extract energy from the antenna !

Need a sub-wavelength receiving element to convert light energy to another form !

(Receiving element can also be described via T-matrix)



T-matrix coefficients are constrained by the underlying physics ! (spherically symmetric case)



Cross sections in Mie theory

Valid for any scatterer

Extinction:
$$\sigma_e = -\frac{1}{k^2} \operatorname{Re}(f^{\dagger}.p) = -\frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1) \operatorname{Re}(t_n^{(e)} + t_n^{(h)}) = +\frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1) \operatorname{Re}(a_n + b_n)$$

Scattering:
$$\sigma_s = \frac{1}{k^2} f^{\dagger} f = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1) \left(\left| t_n^{(e)} \right|^2 + \left| t_n^{(h)} \right|^2 \right) = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1) \left(\left| a_n \right|^2 + \left| b_n \right|^2 \right)$$

Absorption : $\sigma_a = \sigma_e - \sigma_s$

Exact electric dipole polarizability can be deduced from Mie theory !

$$\alpha(\omega) = 6\pi T_1^{(e)}/ik^3$$

Quasi-static limit – electric dipole

$$\lim_{x \to 0} j_n(x) \to \frac{x^n}{(2n+1)!!} \qquad \qquad \lim_{x \to 0} \varphi_n^{(+)}(x) \to -n$$

$$x = kR$$

$$\lim_{x \to 0} h_n(x) \to \frac{(2n-1)!!}{ix^{n+1}} \qquad \qquad \lim_{x \to 0} \varphi_n(x) \to n+1$$

$$\lim_{\boldsymbol{x}\to 0} T_n^{(e)}(\boldsymbol{x}) = \lim_{\boldsymbol{x}\to 0} -\frac{j_n(\boldsymbol{x})}{h_n^{(+)}(\boldsymbol{x})} \frac{\frac{\varepsilon_s}{\varepsilon} \varphi_n(\boldsymbol{x}) - \varphi_n(\rho_s \boldsymbol{x})}{\frac{\varepsilon_s}{\varepsilon} \varphi_n^{(+)}(\boldsymbol{x}) - \varphi_n(\rho_s \boldsymbol{x})} \to \frac{i\boldsymbol{x}^{2n+1}(n+1)}{(2n-1)!!(2n+1)!!} \frac{\frac{\varepsilon_s}{\varepsilon} - 1}{\frac{\varepsilon_s}{\varepsilon} n + (n+1)}$$

Electric dipole term (n=1) dominates for small particle sizes :

$$\lim_{\boldsymbol{k} R \to 0} T_1^{(\boldsymbol{e})}(\boldsymbol{k} R) \to i \boldsymbol{k} R^3 \frac{2}{3} \frac{\varepsilon_s - \varepsilon}{\varepsilon_s + 2\varepsilon}$$

A particle much smaller than the wavelength generally interacts with incident light via its electric dipole moment $\vec{\mathbf{p}} = \epsilon_0 \varepsilon_b \alpha(\omega) \vec{\mathbf{E}}_{exc}$

 ε_b ε_s

$$\lim_{\omega \to 0} \alpha(\omega) = 4\pi R^3 \frac{\varepsilon_s - \varepsilon}{2\varepsilon + \varepsilon_s} \equiv \alpha_0$$

 $\alpha(\omega) = 6\pi T_{\cdot}^{(e)} / ik^3$



S-Matrix (absorption/lasing and energy conservation/unitary limit)

The S-matrix relates the outgoing part of the total field to the incoming part

Spherically symmetric particles :

$$S_n^{(e)}(kR) = -\frac{h_n^{(-)}(kR)}{h_n^{(+)}(kR)} \frac{\frac{\varepsilon_s}{\varepsilon}\varphi_n^{(-)}(kR) - \varphi_n(k_sR)}{\frac{\varepsilon_s}{\varepsilon}\varphi_n^{(+)}(kR) - \varphi_n(k_sR)}$$

Limit behaviors correspond to cross section bounds

$$\sigma_{\text{scat,}n}^{(e,h)} = \frac{\lambda^2 (2n+1)}{8\pi} \left(\left| 1 - S_n^{(e,h)} \right|^2 \right) \qquad \sigma_{\text{ext,}n}^{(e,h)} = \frac{\lambda^2 (2n+1)}{4\pi} \operatorname{Re} \left\{ 1 - S_n^{(e,h)} \right\} \qquad \sigma_{\text{abs,}n}^{(e,h)} = \frac{\lambda^2 (2n+1)}{8\pi} \left(1 - \left| S_n^{(e,h)} \right|^2 \right)$$

$$\text{Ideal absorption : } S_n^{(e,h)} = 0 \qquad \text{Im} \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad S_n^{(e,h)} = e^{i2\delta_n^{(e)}} , \quad S_n^{(h)} = e^{i2\delta_n^{(h)}} \qquad S_n^{(e)} = e^{i2\delta_n^{(e)}} , \quad S_n^{(h)} = e^{i2\delta_n^{(h)}} \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad S_n^{(e,h)} = e^{i2\delta_n^{(e)}} , \quad S_n^{(h)} = e^{i2\delta_n^{(h)}} \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad S_n^{(e,h)} = e^{i2\delta_n^{(e)}} , \quad S_n^{(h)} = e^{i2\delta_n^{(h)}} \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad S_n^{(e,h)} = e^{i2\delta_n^{(e)}} , \quad S_n^{(h)} = e^{i2\delta_n^{(h)}} \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad S_n^{(e,h)} = e^{i2\delta_n^{(e)}} , \quad S_n^{(h)} = e^{i2\delta_n^{(h)}} \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad S_n^{(e,h)} = e^{i2\delta_n^{(e)}} , \quad S_n^{(e,h)} = 1 \qquad S_n^{(e,h)} = 0 \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad \sigma_{\text{ext},n}^{(e,h)} = \sigma_{\text{scat},n}^{(e,h)} = \sigma_{\text{abs},n}^{(e,h)} = 0 \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad S_n^{(e,h)} = 0 \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad S_n^{(e,h)} = e^{i2\delta_n^{(e)}} , \quad S_n^{(e)} = 1 \qquad S_n^{(e,h)} = 0 \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad S_n^{(e,h)} = 0 \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad S_n^{(e,h)} = 0 \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad S_n^{(e,h)} = 0 \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad S_n^{(e,h)} = 0 \qquad \text{Energy conservation circle } \left| S_n^{(e,h)} \right| = 1 \qquad S_n^{(e,h)} = 0 \qquad$$

Simplicity of analytic structure (Weierstrass factorization)



Ideal absorption – 'Coherent perfect absorption' $S_n^{(e,h)} = 0$ Ultimate absorption ?

$$s_n^{(e)}(kR) = -\frac{h_n^{(-)}(kR)}{h_n^{(+)}(kR)} \frac{\frac{\varepsilon_s}{\varepsilon}\varphi_n^{(-)}(kR) - \varphi_n(k_sR)}{\frac{\varepsilon_s}{\varepsilon}\varphi_n^{(+)}(kR) - \varphi_n(k_sR)}$$



IA can be found by solving a complex transcendental equation

$$\frac{\varepsilon_s}{\varepsilon}\varphi_n^{(-)}(kR) - \varphi_n(k_sR) = 0$$

V.Grigoriev, et al ACS Photonics 10.1021/ph500456w ACS Photonics (2015)

Ideal absorption is possible for both electric and magnetic $\overline{\varepsilon} \equiv \frac{\varepsilon_s}{\varepsilon}$ modes in high index materials



IA in homogeneous particles and realistic materials ? Yes, but only for certain sizes and frequencies

IA electric dipole - exact calculation

IA – electric dipole point-like approximation

Silver – Experimental dispersion curves : Johnson & Christy Gold –



Ideal absorption can occur in subwavelength particles (Using realistic materials like gold and silver)

in a N_b =1.5 background medium



Silicon particles should also exhibit Ideal absorption



Electric near-field enhancements



Electric near-field enhancements



Magnetic near-field enhancements



Ideal Absorption limit

$$\sigma_{\rm abs} = \sigma_{\rm scat} \sim \lambda^2/8$$

Unitary limit

$$\sigma_{\rm scat} \sim \lambda^2/2$$



$$\sigma_{\rm abs} = \sigma_{\rm scat} \sim \lambda^2/8$$

 $\sigma_{\rm scat} \sim \lambda^2/2$


- Multipole theory helps understand sub-wavelength dielectric antennas
- The S matrix is well adapted to studying fundamental limits of light-matter interactions like ideal absorption, unitarity, invisibility, and lasing.
- Limit behaviors can occur in either electric and magnetic modes of particles.
- Ideal Absorption with realistic materials at arbitrary frequencies will often require coated particle designs in order to introduce additional parameters.

The K-matrix relates the regular part of the total field to its diverging part

$$\vec{\mathbf{E}}_{\text{tot}}(k\vec{r}) = \vec{\mathbf{E}}_{\text{exc}}(k\vec{r}) + \vec{\mathbf{E}}_{\text{scat}}(k\vec{r})$$

$$= \sum_{n,m}^{\infty} \left\{ \left[r_{n,m}^{(h)} \vec{\mathbf{M}}_{n,m}^{(1)}(k\vec{r}) + r_{n,m}^{(e)} \vec{\mathbf{N}}_{n,m}^{(1)}(k\vec{r}) \right] + \left[d_{n,m}^{(h)} \vec{\mathbf{M}}_{n,m}^{(2)}(k\vec{r}) + d_{n,m}^{(e)} \vec{\mathbf{N}}_{n,m}^{(2)}(k\vec{r}) \right] \right\}$$

$$d \equiv \overline{K}. r$$

$$\vec{\mathbf{k}} = -i\overline{K}. (\overline{\mathbf{T}} + \overline{\mathbf{I}}) \qquad \overline{\mathbf{T}}^{-1} = i\overline{K}^{-1} - \overline{\mathbf{I}}$$

$$\overline{K} = i(\overline{\mathbf{S}} - \overline{\mathbf{I}}). (\overline{\mathbf{I}} + \overline{\mathbf{S}})^{-1} \qquad \overline{\mathbf{S}} = (\overline{\mathbf{I}} - i\overline{K}). (\overline{\mathbf{I}} + i\overline{K})^{-1}$$

$$Caley Transform$$

$$K_{n}^{(e)} = -\tan\delta_{n}^{(e)},$$

$$K_{n}^{(h)} = -\tan\delta_{n}^{(h)}$$

K-Matrix is Hermitian for a lossless scatterer (adapted to studying energy conservation and limit behaviors)

 $\overline{\overline{K}}^{\dagger} = \overline{\overline{K}}$: Energy conservation



Simplicity of analytic structure (Weierstrass factorization)

$$S_{n}(x) = A \exp(-2iBx) \prod_{\alpha} \frac{x - x_{z,\alpha}}{x - x_{p,\alpha}} = A \exp(-2iBx) \prod_{\alpha=1}^{\infty} \frac{(x - x_{z,\alpha})(x + x_{z,\alpha}^{*})}{(x - x_{p,\alpha})(x + x_{p,\alpha}^{*})} \qquad x \equiv kR$$

$$Im\{kR\}$$

$$= \frac{1}{2} + \frac{1}{$$

K-Matrix (Reaction matrix) Gives access to all limit behaviors of light-matter interactions

Spherically symmetric particles :

$$K_{n}^{(e,h)} \rightarrow \infty \Rightarrow S_{n}^{(e,h)} = T_{n}^{(e,h)} = -1 \qquad \longrightarrow \qquad \text{Unitary limit} \\ K_{n}^{(e,h)} = 0 \Rightarrow S_{n}^{(e,h)} \rightarrow 1 \quad , \ T_{n}^{(e,h)} \rightarrow 0 \qquad \longrightarrow \qquad \text{Invisible} \end{cases} \qquad \text{lossless} \\ K_{n}^{(e,h)} = i \Rightarrow S_{n}^{(e,h)} \rightarrow \infty \quad , \ T_{n}^{(e,h)} \rightarrow \infty \qquad \longrightarrow \qquad \text{Emission - lasing} \\ K_{n}^{(e,h)} = -i \Rightarrow S_{n}^{(e,h)} \rightarrow 0 \quad , \ T_{n}^{(e,h)} \rightarrow -\frac{1}{2} \qquad \longrightarrow \qquad \text{Ideal absorption} \end{cases} \qquad \text{gain} \\ \text{loss}$$

Other formulations of physical limits

$$\sigma_{\text{ext}} = k \text{Im}\{\alpha(\omega)\}$$

$$\sigma_{\text{scat}} = \frac{k^4 |\alpha(\omega)|^2}{6\pi}$$

$$\alpha(\omega) = 6\pi T_1^{(e)} / ik^3$$

Unitary limit : $\sigma_{\text{ext}} \le \frac{3\lambda^2}{2\pi}$
Unitarity : $\text{Im}\{\alpha\} \ge \frac{k^3 |\alpha|^2}{6\pi}$

$$\sigma_{\text{ext}} \le \frac{3\lambda^2}{2\pi} = \frac{k|\alpha|^2}{\text{Im}\{\alpha\}}$$

Electric near-field enhancements



z (µm)

kR = 0.8

60

1.5

0.5

0.5

z (µm)

Translation-addition theorem (scalar wave case)

Scalar wave translation-addition matrices

$$\alpha_{\nu,\mu;n,m}\left(k\vec{\mathbf{r}}_{0}\right) = 4\pi i^{\nu-m} \sum_{q=|n-\nu|}^{q=n+\nu} i^{q} \, 3Y\left(n,m,\nu,\mu;q\right) h_{q}\left(kr_{0}\right) Y_{q,m-\mu}\left(\theta_{0},\phi_{0}\right)$$
$$\beta_{\nu,\mu;n,m}\left(k\vec{\mathbf{r}}_{0}\right) = Rg\left\{\alpha_{\nu,\mu;n,m}\left(k\vec{\mathbf{r}}_{0}\right)\right\} = 4\pi i^{\nu-m} \sum_{q=|n-\nu|}^{q=n+\nu} i^{q} \, 3Y\left(n,m,\nu,\mu;q\right) j_{q}\left(kr_{0}\right) Y_{q,m-\mu}\left(\theta_{0},\phi_{0}\right)$$

Looks horrible but relatively easy to derive in k-space !

$$3Y(\nu,\mu;n,m;q) \equiv \int_0^{\pi} \sin\theta d\theta \int_0^{2\pi} d\phi Y_{n,m}(\theta,\phi) Y_{\nu,\mu}^*(\theta,\phi) Y_{q,m-\mu}^*(\theta,\phi)$$
$$= \left(-\right)^m \left[\frac{(2n+1)(2\nu+1)[2q+1]}{4\pi}\right]^{1/2} \binom{n \quad \nu \quad q}{0 \quad 0 \quad 0} \binom{n \quad \nu \quad q}{-m \quad \mu \quad \mu-m}$$

Translation-addition theorem (vector wave case)

$$\vec{\Psi}^{t}(k\vec{\mathbf{r}}) = \vec{\Psi}^{t}(k\vec{\mathbf{r}}').J(k\vec{\mathbf{r}}_{0}) \qquad r' > r_{0}$$
$$\vec{\Psi}^{t}(k\vec{\mathbf{r}}) = Rg\left\{\vec{\Psi}^{t}(k\vec{\mathbf{r}}')\right\}.H(k\vec{\mathbf{r}}_{0}) \qquad r' < r_{0}$$
$$Rg\left\{\vec{\Psi}^{t}(k\vec{\mathbf{r}})\right\} = Rg\left\{\vec{\Psi}^{t}(k\vec{\mathbf{r}}')\right\}.J(k\vec{\mathbf{r}}_{0}) \qquad \forall |\vec{\mathbf{r}}_{0}|$$

$$H\left(k\vec{\mathbf{r}}_{0}\right) = \begin{bmatrix} A_{\nu\mu,nm} & B_{\nu\mu,nm} \\ B_{\nu\mu,nm} & A_{\nu\mu,nm} \end{bmatrix} \qquad A_{\nu\mu,nm} = \frac{1}{2}\sqrt{\frac{1}{\nu(\nu+1)n(n+1)}} \begin{bmatrix} 2\mu m\alpha_{\nu,\mu;n,m} \\ +\sqrt{(n-m)(n+m+1)}\sqrt{(\nu-\mu)(\nu+\mu+1)\alpha_{\nu,\mu+1;n,m+1}} \\ +\sqrt{(n-m)(n-m+1)}\sqrt{(\nu-\mu)(\nu-\mu+1)\alpha_{\nu,\mu-1;n,m-1}} \end{bmatrix}$$

$$J\left(k\vec{\mathbf{r}}_{0}\right) = Rg\left\{ \begin{bmatrix} A_{\nu\mu,nm} & B_{\nu\mu,nm} \\ B_{\nu\mu,nm} & A_{\nu\mu,nm} \end{bmatrix} \right\} \qquad B_{\nu\mu,nm} = -i\frac{1}{2}\sqrt{\frac{2\nu+1}{2\nu-1}\frac{1}{\nu(\nu+1)n(n+1)}} \begin{bmatrix} 2m\sqrt{(\nu-\mu)(\nu+\mu)\alpha_{\nu-1,\mu;n,m+1}} \\ +\sqrt{(n-m)(n+m+1)}\sqrt{(\nu-\mu)(\nu+\mu-1)\alpha_{\nu-1,\mu+1;n,m+1}} \\ -\sqrt{(n+m)(n-m+1)}\sqrt{(\nu+\mu)(\nu-\mu-1)\alpha_{\nu-1,\mu-1;n,m-1}} \end{bmatrix}$$

Possible values of the Mie coefficients limited by the underlying physics !

Mie Coefficients $a_n(\omega)$ and $b_n(\omega)$



Analogue Antennas in the micro-wave regime (low index : $N \cong 2$)

Nature - Scientific Reports, 3 3063 (2013)

Multimode interference in a dielectric sphere can yield directive emissions



$$I_{r}(\theta,0) = I_{0} \left[\left(e^{i\varphi} + \gamma_{1}^{(a)} \alpha_{1}^{(e)} + \gamma_{2}^{(b)} \alpha_{2}^{(h)} \right) \cos\theta + 2\gamma_{2}^{(a)} \alpha_{2}^{(e)} \cos^{2}\theta + \gamma_{1}^{(b)} \alpha_{1}^{(h)} - \gamma_{2}^{[a]} \alpha_{2}^{(e)} \right]^{2}$$

Analogue Antennas in the micro-wave regime (low index : $N \cong 2$)

Nature - Scientific Reports, 3 3063 (2013)

KCCRM





Measured emission and theory



8,7 GHz

9,4 GHz



Changing antenna directivity with displacement



Benedicto; Bonod, Stout "Optimization of dielectric Antenna directivity" : in preparation

Suppressing forward scattering with high index dielectric antennas

- Isolated resonances
- Simpler interferences
- Fully suppressed forward emissions

- (a) Actual emission
- (b) Electric dipole suppressed

Modifying quantum decay rates with nano-antennas

Quasi-analytic multipole formalism : (radiative and non-radiative decay rates)

$$\frac{\Gamma_e}{\Gamma_0} = \frac{\langle P_e \rangle}{\langle P_{e,0} \rangle} = 1 + \frac{6\pi}{\operatorname{Re}\{k_b\}} \operatorname{Re}\left\{k_b \sum_{j,l=1}^N f^{\dagger} \cdot H^{(0,j)} \cdot T^{(j,l)} \cdot H^{(l,0)} \cdot f\right\}$$

$$\frac{\Gamma_{r}}{\Gamma_{0}} = \frac{\langle P_{r} \rangle}{\langle P_{r,0} \rangle} = 1 + 6\pi \sum_{j,l,k,l=1}^{N} \left[T^{(j,i)} \cdot H^{(i,0)} \cdot f \right]^{\dagger} \cdot J^{(j,k)} \cdot T^{(k,l)} \cdot H^{(i,0)} \cdot f + 12\pi \operatorname{Re} \left\{ \sum_{j,l=1}^{N} \left[J^{(k,0)} f \right]^{\dagger} \cdot T^{(k,j)} \cdot H^{(j,0)} \cdot f \right\}$$

K-Matrix (Reaction matrix)

The K-matrix relates the regular part of the total field to its diverging part

$$\vec{\mathbf{E}}_{tot}(k\vec{r}) = \vec{\mathbf{E}}_{exc}(k\vec{r}) + \vec{\mathbf{E}}_{scat}(k\vec{r}) = \sum_{n,m}^{\infty} \left\{ \left[r_{n,m}^{(h)} \vec{\mathbf{M}}_{n,m}^{(1)}(k\vec{r}) + r_{n,m}^{(e)} \vec{\mathbf{N}}_{n,m}^{(1)}(k\vec{r}) \right] + \left[d_{n,m}^{(h)} \vec{\mathbf{M}}_{n,m}^{(2)}(k\vec{r}) + d_{n,m}^{(e)} \vec{\mathbf{N}}_{n,m}^{(2)}(k\vec{r}) \right] \right\}$$

$$d \equiv \overline{\overline{K}}.r \qquad \Longrightarrow \qquad \left\{ \qquad \overline{\overline{T}} = -i\overline{\overline{K}}.\left(\overline{\overline{T}} + \overline{\overline{I}}\right) \qquad \overline{\overline{T}}^{-1} = i\overline{\overline{K}}^{-1} - \overline{\overline{I}} \right\}$$

$$K_n^{(e)} = -\frac{j_n(kR)}{y_n(kR)} \frac{\frac{\varepsilon_s}{\varepsilon} \varphi_n(kR) - \varphi_n(k_sR)}{\frac{\varepsilon_s}{\varepsilon} \varphi_n^{(2)}(kR) - \varphi_n(k_sR)}$$

 $K_n^{(e)} = -\tan \delta_n^{(e)},$ $K_n^{(h)} = -\tan \delta_n^{(h)}$ Reaction matrix is Hermitian for lossless scatterers

$$i\overline{\overline{T}}^{-1} = -\overline{\overline{K}}^{-1} - i\overline{\overline{I}}$$

Unitary limit
$$\longrightarrow K_n \to \infty \longrightarrow \frac{\varepsilon_s}{\varepsilon} \varphi_n^{(2)}(kR) - \varphi_n(k_sR) = 0$$

K-matrix is useful for deriving approximate non-transcendental approximations – to appear !

Rigorous formulation of "radiative corrections" - other "optical theorem"

$$\alpha(\omega) = 6\pi T_1^{(e)}/ik^3 \qquad \alpha^{-1} = \alpha_{n.r.}^{-1} - i\frac{k^3}{6\pi} \implies \alpha_{n.r.} = \frac{6\pi}{k^3}K_1^{(e)}$$

- Use multipole methods to determine quasi-normal modes of systems
- Quasi-normal modes should help optimize the performance of photonic systems.
- Quasi-normal modes should help formulating theories of strong coupling between a quantum emitter and its nano-photonic environment.

Vector "partial" waves

Maxwell equation :

 $\nabla \times \nabla \times \vec{\mathbf{E}} - k^2 \vec{\mathbf{E}} = \vec{\mathbf{0}}$

 $\nabla \cdot \vec{\mathbf{M}} = 0$ $\nabla \cdot \vec{\mathbf{N}} = 0$

 $\psi_n(x) \equiv x j_n(x)$ $\psi'_n(x) = \left[j_n(x) + x j'_n(x) \right]$

 $\vec{\mathbf{M}}^{(1)}{}_{n,m}(k\vec{r}) = j_n(kr)\vec{\mathbf{X}}_{n,m}(\theta,\phi)$ $\vec{\mathbf{N}}^{(1)}{}_{n,m}(k\vec{r}) = \frac{1}{kr} \Big[j_n(kr)\sqrt{n(n+1)}\vec{\mathbf{Y}}_{n,m}(\theta,\phi) + \frac{\psi'_n(kr)\vec{\mathbf{Z}}_{n,m}(\theta,\phi) \Big]$

Electric and Magnetic types :

Solutions to Maxwell's propagation equations in spherical coordinates

 $\nabla \times \nabla \times \vec{\mathbf{E}} - k^2 \, \vec{\mathbf{E}} = 0$

<u>Transverse</u> vector "partial" waves : $\nabla \cdot \vec{\mathbf{M}}_{n,m} = \nabla \cdot \vec{\mathbf{N}}_{n,m} = 0$

$$\vec{M}_{n,m}(k\vec{r}) = \frac{\nabla \times \left[\vec{r}\varphi_{n,m}(k\vec{r})\right]}{\sqrt{n(n+1)}}$$
$$\vec{N}_{n,m}(k\vec{r}) = \frac{\nabla \times \vec{M}_{n,m}(k\vec{r})}{k}$$

magnetic modes

electric modes

$$\Delta \varphi + k^2 \varphi = 0$$

$$\varphi_{n,m}(k\vec{r}) = j_n(kr)Y_{n,m}(\theta,\phi)$$

Linearly independent solutions $\nabla \cdot \vec{\mathbf{M}} = 0$ $\nabla \cdot \vec{\mathbf{N}} = 0$

 $\vec{\mathbf{M}}^{(1)}{}_{n,m}(k\vec{r}) = j_n(kr)\vec{\mathbf{X}}_{n,m}(\theta,\phi)$

 $\vec{\mathbf{N}}^{(1)}{}_{n,m}(k\vec{r}) = \frac{1}{kr} \Big[j_n(kr) \sqrt{n(n+1)} \vec{\mathbf{Y}}_{n,m}(\theta,\phi) \\ + \psi'_n(kr) \vec{\mathbf{Z}}_{n,m}(\theta,\phi) \Big]$

 $\psi_n'(kr) = [xj_n(x)]'$

Spherical Bessel functions (1)

 $\vec{\mathbf{M}}^{(2)}_{n,m}(k\vec{r}) = y_n(kr)\vec{\mathbf{X}}_{n,m}(\theta,\phi)$

 $\vec{\mathbf{N}}^{(2)}{}_{n,m}(k\vec{r}) = \frac{1}{kr} \Big[y_n(kr) \sqrt{n(n+1)} \vec{\mathbf{Y}}_{n,m}(\theta,\phi) + \chi'_n(kr) \vec{\mathbf{Z}}_{n,m}(\theta,\phi) \Big]$

 $\chi_n'(x) = [xy_n(x)]'$

Spherical Neumann functions (2)

Outgoing and incoming boundary conditions

$$\vec{\mathbf{M}}^{(+)}{}_{n,m}(k\vec{r}) = h_n^{(+)}(kr)\vec{\mathbf{X}}_{n,m}(\theta,\phi)$$

$$\vec{\mathbf{N}}^{(+)}{}_{n,m}(k\vec{r}) = \frac{1}{kr} \Big[h_n^{(+)}(kr)\sqrt{n(n+1)}\vec{\mathbf{Y}}_{n,m}(\theta,\phi) + \xi_n^{(+)\prime}(kr)\vec{\mathbf{Z}}_{n,m}(\theta,\phi) \Big]$$

$$h_n^{(+)}(x) = j_n(x) + iy_n(x)$$

$$\xi_n^{(+)\prime}(x) = \Big[h_n^{(+)}(x) \Big]'$$

$$h_{0}^{(+)}(x) = -\frac{i}{x}e^{ix}$$
$$h_{1}^{(+)}(x) = -e^{ix}\left(\frac{1}{x} + \frac{i}{x^{2}}\right)$$
$$\vdots$$

$$\vec{\mathbf{M}}^{(-)}{}_{n,m}(k\vec{r}) = h_n^{(-)}(kr)\vec{\mathbf{X}}_{n,m}(\theta,\phi)$$

$$\vec{\mathbf{N}}^{(-)}{}_{n,m}(k\vec{r}) = \frac{1}{kr} \Big[h_n^{(-)}(kr) \sqrt{n(n+1)} \vec{\mathbf{Y}}_{n,m}(\theta,\phi) + \xi_n^{(-)\prime}(kr) \vec{\mathbf{Z}}_{n,m}(\theta,\phi) \Big]$$

$$h_n^{(-)}(x) = j_n(x) - iy_n(x)$$

$$\xi_n^{(-)'}(x) = \left[h_n^{(-)}(x)\right]'$$

Incoming spherical Hankel functions (-)

$$h_0^{(-)}(x) = \frac{i}{x}e^{-ix}$$
$$h_1^{(-)}(x) = -e^{-ix}\left(\frac{1}{x} - \frac{i}{x^2}\right)$$
$$\vdots$$

The K-matrix relates the regular part of the total field to its diverging part

Simplicity of analytic structure (Weierstrass factorization)

$$S_{n}(x) = A \exp(-2iBx) \prod_{\alpha} \frac{x - x_{z,\alpha}}{x - x_{p,\alpha}} = A \exp(-2iBx) \prod_{\alpha=1}^{\infty} \frac{(x - x_{z,\alpha})(x + x_{z,\alpha}^{*})}{(x - x_{p,\alpha})(x + x_{p,\alpha}^{*})} \qquad x \equiv kR$$

$$Im\{kR\}$$

$$s_{n}^{(e)}(kR) = -\frac{h_{n}^{(-)}(kR)}{h_{n}^{(+)}(kR)} \frac{\varepsilon_{s}}{\varepsilon} \varphi_{n}^{(-)}(kR) - \varphi_{n}(k_{s}R)}{kR}$$

$$s_{n}^{(e)}(kR) = -\frac{h_{n}^{(-)}(kR)}{h_{n}^{(+)}(kR)} \frac{\varepsilon_{s}}{\varepsilon} \varphi_{n}^{(+)}(kR) - \varphi_{n}(k_{s}R)}{kR}$$

$$\kappa_{1}^{(e)}$$

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$$s_{n}^{(e)}(kR) = -\frac{h_{n}^{(-)}(kR)}{h_{n}^{(+)}(kR)} \frac{\varepsilon_{s}}{\varepsilon} \varphi_{n}^{(+)}(kR) - \varphi_{n}(k_{s}R)}{kR}$$

$$\kappa_{1}^{(e)}$$

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$$s_{n}^{(e)}(kR) = -\frac{h_{n}^{(-)}(kR)}{kR} \frac{\varepsilon_{s}}{\varepsilon} \varphi_{n}^{(+)}(kR) - \varphi_{n}(k_{s}R)}{kR}$$

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$$\kappa_{1}^{(e)}$$

$$s_{n}^{(e)}(kR) = -\frac{h_{n}^{(e)}(kR)}{kR} \frac{\varepsilon_{s}}{\varepsilon} \varphi_{n}^{(+)}(kR) - \varphi_{n}(k_{s}R)}{kR}$$

$$\kappa_{1}^{(e)}$$

$$s_{n}^{(e)}(kR) = -\frac{h_{n}^{(e)}(kR)}{kR} \frac{\varepsilon_{s}}{\varepsilon} \varphi_{n}^{(+)}(kR) - \varphi_{n}(k_{s}R)}{kR}$$

Ideal absorption given by the solution of a transcendental equation solving for of $\varepsilon_s/\varepsilon$ as a function of kR

Determining the Unitary limit condition

$$s_{n}^{(e,h)} = -1 \quad , \qquad t_{n}^{(e,h)} = -1 \quad , \qquad \kappa_{n}^{(e,h)} \to \infty$$

$$s_{n}^{(e)}(kR) = -\frac{h_{n}^{(-)}(kR)}{h_{n}^{(+)}(kR)} \frac{\varepsilon_{s}}{\varepsilon} \varphi_{n}^{(-)}(kR) - \varphi_{n}(k_{s}R)}{h_{n}^{(+)}(kR)} \qquad t_{n}^{(e)} = -\frac{j_{n}(kR)}{h_{n}^{(+)}(kR)} \frac{\varepsilon_{s}}{\varepsilon} \varphi_{n}(kR) - \varphi_{n}(k_{s}R)}{h_{n}^{(+)}(kR)} \frac{\varepsilon_{s}}{\varepsilon} \varphi_{n}^{(+)}(kR) - \varphi_{n}(k_{s}R)}$$

$$\kappa_n^{(e)} = -\frac{j_n(kR)}{y_n(kR)} \frac{\frac{\varepsilon_s}{\varepsilon} \varphi_n(kR) - \varphi_n(k_sR)}{\frac{\varepsilon_s}{\varepsilon} \varphi_n^{(2)}(kR) - \varphi_n(k_sR)}$$

The unitary limit can be found by solving a transcendental equation

$$\frac{\varepsilon_s}{\varepsilon}\varphi_n^{(2)}(kR) - \varphi_n(k_sR) = 0$$

Light interactions with a particle much smaller than the wavelength

electric dipole description (polarizability)

$$\mathbf{\hat{p}} = \epsilon_0 \varepsilon_b \alpha(\omega) \mathbf{\vec{E}}_{exc}$$

$$\vec{\mathbf{p}} = \mathcal{E}_0 \mathcal{E} \alpha \left(\boldsymbol{\omega} \right) \vec{\mathbf{E}}_{\text{exc}} \qquad \qquad \boldsymbol{\alpha}_0 = \lim_{k \to 0} \alpha \left(\boldsymbol{\omega} \right) = 4\pi R^3 \frac{\mathcal{E}_s - \mathcal{E}}{2\mathcal{E} + \mathcal{E}_s}$$

Radiative correction model :
$$\lim_{\omega \to 0} \alpha(\omega) = \frac{A(\omega)}{1 - i \frac{k^3}{6\pi} A(\omega)} \cong \frac{\alpha_0}{1 - i \frac{k^3}{6\pi} \alpha_0}$$

Green function point-like model :
$$\lim_{\omega \to 0} \alpha(\omega) \cong \frac{\alpha_0}{1 - \frac{1}{6\pi} \frac{k^2}{R} \alpha_0 - i \frac{k^3}{6\pi} \alpha_0}$$

Mie point-like model :

$$\lim_{\omega \to 0} \alpha(\omega) \cong \frac{\alpha_0}{1 - \frac{3}{20\pi} \left(\frac{\varepsilon_s - 2\varepsilon}{\varepsilon_s - \varepsilon}\right) \frac{k^2}{R} \alpha_0 - i \frac{k^3}{6\pi} \alpha_0}$$

$$\alpha^{-1} = \alpha_{q.s.}^{-1} - i \frac{k^3}{6\pi}$$

 \mathcal{E}_{s}

'Point-like' models – single resonant model $k = \frac{\omega}{c} \sqrt{\varepsilon}$

Fundamental limits (Dipole scattering)

$$\alpha(\omega) = t_1^{(e)} \frac{6\pi}{ik^3} \qquad \qquad \sigma_{\text{ext}} = k \text{Im}\{\alpha(\omega)\} \\ \sigma_{\text{scat}} = \frac{k^4 |\alpha(\omega)|^2}{6\pi}$$

Unitary limit (lossless scattering)

$$\alpha_{u.l} = 6\pi i/k^3 \implies \sigma_{ext} = \sigma_{scat} \sim \lambda^2/2$$

Ideal absorption – Optical sink

$$\alpha_{\text{I.A}} = 3\pi i/k^3 \implies \sigma_{\text{abs}} = \sigma_{\text{scat}} \sim \lambda^2/8$$

K-Matrix (Reaction matrix) Gives access to all limit behaviors of light-matter interactions

Spherically symmetric particles :

$$K_{n}^{(e,h)} \rightarrow \infty \Rightarrow S_{n}^{(e,h)} = T_{n}^{(e,h)} = -1 \qquad \longrightarrow \qquad \text{Unitary limit} \\ K_{n}^{(e,h)} = 0 \Rightarrow S_{n}^{(e,h)} \rightarrow 1 \quad , \ T_{n}^{(e,h)} \rightarrow 0 \qquad \longrightarrow \qquad \text{Invisible} \end{cases} \qquad \text{lossless} \\ K_{n}^{(e,h)} = i \Rightarrow S_{n}^{(e,h)} \rightarrow \infty \quad , \ T_{n}^{(e,h)} \rightarrow \infty \qquad \longrightarrow \qquad \text{Emission - lasing} \\ K_{n}^{(e,h)} = -i \Rightarrow S_{n}^{(e,h)} \rightarrow 0 \quad , \ T_{n}^{(e,h)} \rightarrow -\frac{1}{2} \qquad \longrightarrow \qquad \text{Ideal absorption} \end{cases} \qquad \text{gain} \\ \text{loss}$$

Mie theory Highlights the symmetry of the different matrices

$$T_n^{(e)} = -\frac{j_n(kR)}{h_n^{(+)}(kR)} \frac{\frac{\varepsilon_s}{\varepsilon}\varphi_n(kR) - \varphi_n(k_sR)}{\frac{\varepsilon_s}{\varepsilon}\varphi_n^{(+)}(kR) - \varphi_n(k_sR)}$$

$$T_n^{(h)} = -\frac{j_n(kR)}{h_n^{(+)}(kR)} \frac{\frac{\mu_s}{\mu}\varphi_n(kR) - \varphi_n(k_sR)}{\frac{\mu_s}{\mu}\varphi_n^{(+)}(kR) - \varphi_n(k_sR)}$$

$$S_n^{(e)} = -\frac{h_n^{(-)}(kR)}{h_n^{(+)}(kR)} \frac{\frac{\varepsilon_s}{\varepsilon}\varphi_n^{(-)}(kR) - \varphi_n(k_sR)}{\frac{\varepsilon_s}{\varepsilon}\varphi_n^{(+)}(kR) - \varphi_n(k_sR)}$$

$$S_{n}^{(h)} = -\frac{h_{n}^{(-)}(kR)}{h_{n}^{(+)}(kR)} \frac{\frac{\mu_{s}}{\mu} \varphi_{n}^{(-)}(kR) - \varphi_{n}(k_{s}R)}{\frac{\mu_{s}}{\mu} \varphi_{n}^{(+)}(kR) - \varphi_{n}(k_{s}R)}$$

$$K_n^{(e)} = -\frac{j_n(kR)}{y_n(kR)} \frac{\frac{\varepsilon_s}{\varepsilon} \varphi_n(kR) - \varphi_n(k_sR)}{\frac{\varepsilon_s}{\varepsilon} \varphi_n^{(2)}(kR) - \varphi_n(k_sR)} \qquad \qquad K_n^{(h)} = -\frac{j_n(kR)}{y_n(kR)} \frac{\frac{\mu_s}{\mu} \varphi_n(kR) - \varphi_n(k_sR)}{\frac{\mu_s}{\mu} \varphi_n^{(2)}(kR) - \varphi_n(k_sR)}$$

K-Matrix is Hermitian for a lossless scatterer (adapted to studying energy conservation and limit behaviors) $\overline{\overline{K}}^{\dagger} = \overline{\overline{K}} \quad : \text{Energy conservation}$

Weierstrass factorization (useful for solving transcendental equations)

Electric Ideal absorption

$$\frac{\varepsilon}{\varepsilon_s}\varphi_n(k_s R) = \varphi_n^{(-)}(kR)$$

Magnetic Ideal absorption

$$\varphi_n(k_s R) = \varphi_n^{(-)}(kR)$$

Electric mode unitary limit

 $\frac{\varepsilon_s}{\varepsilon}\varphi_n^{(2)}(kR) = \varphi_n(k_sR)$

Magnetic mode unitary limit

$$\varphi_n^{(2)}(kR) = \varphi_n(k_s R)$$

$$\varphi_n(z) = \frac{[zj_n(z)]'}{j_n(z)} = 2 + \sum_{\alpha=1}^{\infty} \left(\frac{2z^2}{z^2 - a_{n,\alpha}^2}\right)$$

$$\varphi_1^{(-)}(x) = \frac{\left[xh_1^{(-)}(x)\right]'}{h_1^{(-)}(x)}$$

Vector spherical harmonics

Scalar spherical harmonics : $Y_{nm}(\theta,\phi) = \left| \frac{2n}{4\pi} \right|$

$$Y_{nm}(\theta,\phi) \equiv \left[\frac{2n+1}{4\pi} \frac{(n-m)!}{(n+m)!}\right] e^{im\phi} P_n^m(\cos\theta)$$

3 types of **vector** spherical harmonics :

$$\begin{split} \overrightarrow{W}_{n,m}^{(1)}(\theta,\phi) &\equiv \overrightarrow{Y}_{n,m}(\theta,\phi) = \widehat{r}Y_{n,m}(\theta,\phi) & n = 0,\dots\infty \qquad m = -n,\dots n \\ \overrightarrow{W}_{n,m}^{(2)}(\theta,\phi) &\equiv \overrightarrow{X}_{n,m}(\theta,\phi) = \overrightarrow{Z}_{n,m}(\theta,\phi) \times \widehat{r} & n = 1,\dots,\infty \qquad m = -n,\dots,n \\ \overrightarrow{W}_{n,m}^{(3)}(\theta,\phi) &\equiv \overrightarrow{Z}_{n,m}(\theta,\phi) = \frac{r\nabla Y_{n,m}}{\sqrt{n(n+1)}} = \widehat{r} \times \overrightarrow{X}_{n,m}(\theta,\phi) \end{split}$$

$$\int_{0}^{4\pi} d\Omega \, \overrightarrow{W}_{n,m}^{(j),*}(\theta,\phi) \cdot \overrightarrow{W}_{n,m}^{(k)}(\theta,\phi) = \delta_{j,k} \delta_{n,\nu} \delta_{m,\mu}$$

IA in homogeneous particles and realistic materials ? Yes, but only for certain sizes and frequencies

IA electric dipole - exact calculation

IA – electric dipole point-like approximation

Silver – Experimental dispersion curves : Johnson & Christy Gold –

Point-like models and their extensions can yield analytic formulas for approximating the IA conditions

Point-like model :
$$\alpha^{-1} = \alpha_0^{-1} - \frac{k^2}{6\pi R} - i\frac{k^3}{6\pi}$$
 IA condition : $\alpha_{0.S} = 3\pi i/k^3$

$$= \frac{1}{\epsilon_{0.S}^{(e)}(kR)} = -\frac{2 + \frac{2}{3}(kR)^2(1 - ikR)}{1 - \frac{2}{3}(kR)^2(1 - ikR)}$$

$$= \frac{1}{\epsilon_{0.S}^{(e)}(kR)} = \frac{$$
S-matrix symmetrizes the limit behaviors



Ideal Absorption (optimizes ?) near field enhancements





$$C_{\rm abs} = C_{\rm scat} \sim \lambda^2/8$$

Sub-wavelength IA particles produce large field enhancements