

Generation of Huygens' dipoles for any spherical nanoparticle excited by counter-propagating plane waves: study of scattered helicity: supplement

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Supplement DOI: <https://doi.org/10.6084/m9.figshare.17185877>

Parent Article DOI: <https://doi.org/10.1364/OE.447827>

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1. SCATTERED FIELD

Let us consider two counter-propagating plane waves along the Z axis (incident on a spherical particle of radius a , located at the coordinates origin) whose electric and magnetic field are given by:

$$\mathbf{E}_1 = E_0 \exp(ikz)(\hat{x} + i\zeta\hat{y}) \quad (\text{S1})$$

$$\mathbf{E}_2 = -E_0 q \exp(-ikz) \exp(i\delta)(\hat{x} + i\zeta\hat{y}) \quad (\text{S2})$$

$$\mathbf{H}_1 = \frac{1}{i\omega\mu} \nabla \times \mathbf{E}_1 \quad (\text{S3})$$

$$\mathbf{H}_2 = \frac{1}{i\omega\mu} \nabla \times \mathbf{E}_2 \quad (\text{S4})$$

being E_0 the amplitude of the counter-propagating fields, q the beam ratio between amplitudes of the counter-propagating waves, $\zeta \in [-1, 1]$ is a parameter that determines the polarization state (we will study the cases $\zeta = 0$ and $\zeta = 1$ that describes linearly and circularly polarized incident waves respectively), δ shows the dephase between the two plane waves and $k^2 = \omega^2 \epsilon \mu$ is the wave number appropriate to the surrounding medium. According to Bohren [1] (and using his notation), the scattered $\mathbf{E}_s$ field by the spherical particle can be expanded in vector spherical harmonics as:

$$\mathbf{E}_s = \mathbf{E}_{s1} + \mathbf{E}_{s2} \quad (\text{S5})$$

where

$$\mathbf{E}_{s1} = \mathbf{E}_f^+(\theta, \phi) + i\zeta \mathbf{R}[\mathbf{E}_f^+(\theta, \phi)] \quad (\text{S6})$$

$$\mathbf{E}_{s2} = \mathbf{E}_f^-(\theta, \phi) + i\zeta \mathbf{R}[\mathbf{E}_f^-(\theta, \phi)] \quad (\text{S7})$$

being $\mathbf{R}[\mathbf{X}(\theta, \phi)] = \mathbf{X}(\theta, \phi - \pi/2)$ (for any vectorial field $\mathbf{X}$), the first term in equations S6-S7 corresponds to the scattered field generated by the $\hat{x}$ component of the incident field and the second term to the scattered field generated by the $\hat{y}$ component of the incident wave [1]. The fields $\mathbf{E}_f^+$ and $\mathbf{E}_f^-$ are given by:

$$\mathbf{E}_f^+ = \sum_{n=1}^{\infty} E_n \left(i a_n \mathbf{N}_{e,01n}^{(3)} - b_n \mathbf{M}_{o,01n}^{(3)} \right) \quad (\text{S8})$$

$$\mathbf{E}_f^- = q \exp(i\delta) \sum_{n=1}^{\infty} E_n \left(i a_n \mathbf{N}_{e,01n}^{(3)-} - b_n \mathbf{M}_{o,01n}^{(3)-} \right) \quad (\text{S9})$$

where $E_n = E_0 i^n \frac{2n+1}{n(n+1)}$. Furthermore, $\mathbf{N}_{e,01n}^{(3)}, \mathbf{M}_{e,01n}^{(3)}$ are the vector spherical harmonics and a_n, b_n are the Mie coefficients, all of them defined according to [1].

For next calculations it is interesting to introduce the explicit form of $\mathbf{N}_{e,01n}^{(3)}, \mathbf{M}_{e,01n}^{(3)}$ that are given by:

$$\mathbf{M}_{e,01n}^{(3)} = \nabla \times [r \psi_{e,o}(r, \theta, \phi) \mathbf{u}_r] \quad (\text{S10})$$

$$\mathbf{N}_{e,01n}^{(3)} = \nabla \times [\mathbf{M}_{e,01n}^{(3)}] / k \quad (\text{S11})$$

where the curl operator is assumed in spherical coordinates, $\mathbf{u}_r$ is the radial unitary vector and the functions $\psi_{e,o}(r, \theta, \phi)$ are given by:

$$\psi_e(r, \theta, \phi) = \cos(\phi) P_n^1(\cos(\theta)) H_1^n(kr) \quad (\text{S12})$$

$$\psi_o(r, \theta, \phi) = \sin(\phi) P_n^1(\cos(\theta)) H_1^n(kr) \quad (\text{S13})$$

being $P_n^1(\cos(\theta))$ and $H_1^n(kr)$ the associated Legendre polynomials and the spherical Hankel function of first kind respectively.

$\mathbf{N}_{e,o1n}^{(3)-}, \mathbf{M}_{e,o1n}^{(3)-}$ are also vector spherical harmonics that are obtained from $\mathbf{N}_{e,o1n}^{(3)}, \mathbf{M}_{e,o1n}^{(3)}$ by applying a rotation matrix M_y of an angle π about the Y axis.

If we consider a generic vectorial field $\mathbf{U}(r, \theta, \phi)$ in spherical coordinates given by:

$$\mathbf{U}(r, \theta, \phi) = R(r, \theta, \phi) \hat{u}_r + \Theta(r, \theta, \phi) \hat{u}_\theta + \Phi(r, \theta, \phi) \hat{u}_\phi \quad (\text{S14})$$

where R, Θ and Φ are arbitrary functions, the rotated field $\mathbf{U}^-(r, \theta, \phi)$ is given by:

$$\mathbf{U}^-(r, \theta, \phi) = R(r, \pi - \theta, \pi - \phi) \hat{u}_r - \Theta(r, \pi - \theta, \pi - \phi) \hat{u}_\theta - \Phi(r, \pi - \theta, \pi - \phi) \hat{u}_\phi \quad (\text{S15})$$

so according to equation S15 and taking into account equations S10-S13 it can be deduced that:

$$\mathbf{N}_{e1n}^{(3)-} = (-1)^n \mathbf{N}_{e1n}^{(3)} \quad (\text{S16})$$

$$\mathbf{N}_{o1n}^{(3)-} = (-1)^{n+1} \mathbf{N}_{o1n}^{(3)} \quad (\text{S17})$$

$$\mathbf{M}_{e1n}^{(3)-} = (-1)^n \mathbf{M}_{e1n}^{(3)} \quad (\text{S18})$$

$$\mathbf{M}_{o1n}^{(3)-} = (-1)^{n+1} \mathbf{M}_{o1n}^{(3)} \quad (\text{S19})$$

Taking into account that:

$$\mathbf{R}[\mathbf{N}_{e1n}^{(3)}] = \mathbf{N}_{o1n}^{(3)}, \mathbf{R}[\mathbf{N}_{o1n}^{(3)}] = -\mathbf{N}_{e1n}^{(3)} \quad (\text{S20})$$

$$\mathbf{R}[\mathbf{M}_{e1n}^{(3)}] = \mathbf{M}_{o1n}^{(3)}, \mathbf{R}[\mathbf{M}_{o1n}^{(3)}] = -\mathbf{M}_{e1n}^{(3)} \quad (\text{S21})$$

and introducing equations S6-S9 and S16-S21 into equation S5 and adequately regrouping the terms, the scattered field field will be given by:

$$\mathbf{E}_s = \sum_{n=1}^{\infty} E_n \left(i A_n \mathbf{N}_{e1n}^{(3)} - B_n \mathbf{M}_{o1n}^{(3)} + i \zeta \left(i A_n \mathbf{N}_{o1n}^{(3)} + B_n \mathbf{M}_{e1n}^{(3)} \right) \right) \quad (\text{S22})$$

A_n and B_n being the parameters related to Mie coefficients a_n, b_n by equations:

$$A_n = \left(1 + (-1)^n q \exp(i\delta) \right) a_n \quad (\text{S23})$$

$$B_n = \left(1 - (-1)^n q \exp(i\delta) \right) b_n \quad (\text{S24})$$

Taking into account the relations of vector spherical harmonic respect the operator $\nabla \times$ given by equation S11 and $\mathbf{M}_{e,o1n}^{(3)} = \nabla \times [\mathbf{N}_{e,o1n}^{(3)}] / k$ [1], and using the Maxwell equation: $\mathbf{H}_s = \frac{1}{i\omega\mu} \nabla \times \mathbf{E}_s$, we obtain:

$$\mathbf{H}_s = \frac{k}{\omega\mu} \sum_{n=1}^{\infty} E_n \left(A_n \mathbf{M}_{e1n}^{(3)} + i B_n \mathbf{N}_{o1n}^{(3)} + i \zeta \left(A_n \mathbf{M}_{o1n}^{(3)} - i B_n \mathbf{N}_{e1n}^{(3)} \right) \right) \quad (\text{S25})$$

2. HELICITY

The eigenstates of helicity operator $\Lambda = \frac{\nabla \times}{k}$ are given by the Riemann-Silberstein linear combinations as it was mentioned in the main text:

$$\mathbf{G}_{\pm} = \frac{1}{\sqrt{2}} (\mathbf{E}_s \pm i\eta \mathbf{H}_s) \quad (\text{S26})$$

with $\eta = \sqrt{\mu/\epsilon}$ the medium impedance, so that:

$$\Lambda \mathbf{G}_{\pm} = \pm \mathbf{G}_{\pm} \quad (\text{S27})$$

which implies that $\mathbf{G}_\pm$ has a well defined helicity of ± 1 .
By introducing S22 and S25 into equation S26, we obtain:

$$\mathbf{G}_+^s = \frac{1}{\sqrt{2}} \sum_{n=1}^{\infty} E_n (iA_n \mathbf{F}_e^+ - B_n \mathbf{F}_o^+ + \xi(iB_n \mathbf{F}_e^+ - A_n \mathbf{F}_o^+)) \quad (\text{S28})$$

$$\mathbf{G}_-^s = \frac{1}{\sqrt{2}} \sum_{n=1}^{\infty} E_n (-iA_n \mathbf{F}_e^- - B_n \mathbf{F}_o^- + \xi(iB_n \mathbf{F}_e^- + A_n \mathbf{F}_o^-)) \quad (\text{S29})$$

where for simplicity we have used the functions $\mathbf{F}_e^+ = \mathbf{M}_{e1n}^{(3)} + \mathbf{N}_{e1n}^{(3)}$, $\mathbf{F}_o^+ = \mathbf{M}_{o1n}^{(3)} + \mathbf{N}_{o1n}^{(3)}$, $\mathbf{F}_e^- = \mathbf{M}_{e1n}^{(3)} - \mathbf{N}_{e1n}^{(3)}$ and $\mathbf{F}_o^- = \mathbf{M}_{o1n}^{(3)} - \mathbf{N}_{o1n}^{(3)}$.

A. Helicity of scattered fields for linearly polarized incident fields

If we assume that incident fields are linearly polarized ($\xi = 0$) equations S28 and S29 becomes:

$$\mathbf{G}_+^s = \frac{1}{\sqrt{2}} \sum_{n=1}^{\infty} E_n (iA_n \mathbf{F}_e^+ - B_n \mathbf{F}_o^+) \quad (\text{S30})$$

$$\mathbf{G}_-^s = \frac{1}{\sqrt{2}} \sum_{n=1}^{\infty} E_n (-iA_n \mathbf{F}_e^- - B_n \mathbf{F}_o^-) \quad (\text{S31})$$

It is evident from previous equations that $\mathbf{G}_\pm^s = 0$ for any spatial position if and only if $A_n = B_n = 0$ but then $\mathbf{G}_\pm^s = 0$. Therefore, it is demonstrated that when the counterpropagating incident fields are linearly polarized, the scattered field is not an eigenstate of the helicity.

B. Helicity of scattered fields for circularly polarized incident fields

If we assume that incident fields are circularly polarized ($\xi = 1$) equations S28 and S29 becomes:

$$\mathbf{G}_+^s = \frac{1}{\sqrt{2}} \sum_{n=1}^{\infty} E_n (iA_n \mathbf{F}_e^+ - B_n \mathbf{F}_o^+ + (iB_n \mathbf{F}_e^+ - A_n \mathbf{F}_o^+)) \quad (\text{S32})$$

$$\mathbf{G}_-^s = \frac{1}{\sqrt{2}} \sum_{n=1}^{\infty} E_n (-iA_n \mathbf{F}_e^- - B_n \mathbf{F}_o^- + (iB_n \mathbf{F}_e^- + A_n \mathbf{F}_o^-)) \quad (\text{S33})$$

this equations can be rewritten as:

$$\mathbf{G}_+^s = \frac{1}{\sqrt{2}} \sum_{n=1}^{\infty} E_n (A_n + B_n)(i\mathbf{F}_e^+ - \mathbf{F}_o^+) \quad (\text{S34})$$

$$\mathbf{G}_-^s = \frac{1}{\sqrt{2}} \sum_{n=1}^{\infty} E_n (A_n - B_n)(-i\mathbf{F}_e^- + \mathbf{F}_o^-) \quad (\text{S35})$$

From previous equations and, as it is discussed in the main text, it is possible to obtain a well defined scattered helicity if $A_n = B_n$ or $A_n = -B_n$.

3. DEDUCTION OF EQUATION 24

Using the well known trigonometric identity:

$$\tan(2x) = \frac{2\tan(x)}{1 - \tan(x)^2} \quad (\text{S36})$$

it is easy to obtain from equation 24 ($\delta = \arctan(\pm 2 \frac{|b_1|}{|a_1|})$) that:

$$\tan(\delta) = \tan(\pm 2 \arctan(|b_1|/|a_1|)) = \pm \frac{2|a_1||b_1|}{|a_1|^2 - |b_1|^2} \quad (\text{S37})$$

If $\Delta = 0$ then by equation (23) $\delta = \arctan\left(\pm \frac{2|a_1||b_1|}{|a_1|^2 - |b_1|^2}\right)$, then

$$\tan(\delta) = \pm \frac{2|a_1||b_1|}{|a_1|^2 - |b_1|^2} \quad (\text{S38})$$

so equations 23 and 24 are equivalent.
An alternative deduction is that equation 23:

$$\delta = \arg \left[\frac{(a_1 \mp b_1)}{(a_1 \pm b_1)} \right] \quad (\text{S39})$$

can be rewritten as:

$$\delta = \arg \left[\frac{(1 \mp b_1/a_1)}{(1 \pm b_1/a_1)} \right] = \arg \left[\frac{(1 \mp |b_1| \exp(-i\Delta)/|a_1|)}{(1 \pm |b_1| \exp(-i\Delta)/|a_1|)} \right] \quad (\text{S40})$$

Being $\Delta = \phi_a - \phi_b$. If $\Delta = \pm \frac{\pi}{2}$ then $\exp(-i\Delta) = \mp i$ and then:

$$\delta = \arg \left[\frac{(1 \pm i|b_1|/|a_1|)}{(1 \mp i|b_1|/|a_1|)} \right] \quad (\text{S41})$$

Taking into account that numerator and denominator are conjugate complex of each other, and the fact that $\arg(Z/Z^*) = 2\arg(Z)$ we finally obtain that:

$$\delta_{\pm} = \pm 2 \arctan \left(\frac{|b_1|}{|a_1|} \right) \quad (\text{S42})$$

REFERENCES

1. C. F. Bohren and D. Huffman, *Absorption and Scattering of Light by Small Particles* (Jonh Wiley & Sons, 1998).