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Reciprocity in Nonlocal Optics and Spectroscopy

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1. Introduction
In any wave propagation, the wave can go through various scattering processes through interaction with target in the environment from a source to a detector. In such a process, reciprocity refers to the equality in the signal received when the source and the detector are reversed, that is; their respective positions switched (Potton, 2004). We can find many interesting applications which based on either its validity or its breakdown, in the large number of areas involving transmission of signals ranging of classical optical problems. In classical optics, reciprocity is a powerful result which finds applications in many problems in optics (Potton, 2004) and spectroscopy (Hill et al., 1997). For example, we can establish relations between far fields and near fields from different sources as well as spectroscopic analysis of surface enhanced Raman scattering (SERS) at metallic structure (Kahl & Voges 2000; Ru & Etchegoin, 2006). However, in the previous literature, the optical reciprocity always has been discussed under local optics (Potton, 2004). Now we try to describe the optical reciprocity from electrostatics to electrodynamics under nonlocal optics in order to consider some quantum effects of the particles. Our goal is that the general conditions to determine that the optical reciprocity remains or breaks down will be constructed under nonlocal optics. Some examples and applications will also be discussed.

2. Reciprocity in electrostatics (Green reciprocity)
If we consider an object whose size is much smaller than the wavelength of the incident light, then the effect of retardation can be neglected. Hence we can simply use electrostatics to discuss the interaction between the light and the material. In mathematics, we usually use two popular forms to describe optical reciprocity. One is the Lorentz lemma in electrostatics and the other is the symmetry of the scalar Green function.

2.1 Lorentz lemma in electrostatics
Lorentz lemma in electrostatics form is well-known with local optical response of the medium in the literature. We will extend to consider nonlocal optical response of the medium, since it is known that such response is rather significant with metallic nanostructures due to the large surface-to-volume ratio of these systems. First we write the mathematical form of the Lorentz lemma in electrostatics as follows (Griffiths 1999; Jackson, 1999):
where $\Phi_1 (\Phi_2)$ is the electric potential resulting from the total charge density $\rho_1 (\rho_2)$. Here we will derive this lemma in two different kinds of circumstances.

### 2.1.1 Anisotropic local response

In the beginning, we start from the Poisson equations with two different distributions of charge density $\rho_1$ and $\rho_2$:

$$
\nabla \cdot \left[ \varepsilon (\vec{r}) \cdot \nabla \Phi_1 (\vec{r}) \right] = -4\pi \rho_1
$$

$$
\nabla \cdot \left[ \varepsilon (\vec{r}) \cdot \nabla \Phi_2 (\vec{r}) \right] = -4\pi \rho_2
$$

where $\varepsilon (\vec{r})$ is a dielectric tenser. Next we use Eq. (A1) and put the tensor $\chi = \varepsilon$, the value $\Phi = \Phi_1$ and $\Psi = \Phi_2$. Thus we have the following equality:

$$
\int \left[ \Phi_1 \nabla \cdot \left( \varepsilon \cdot \nabla \Phi_2 \right) - \Phi_2 \nabla \cdot \left( \varepsilon \cdot \nabla \Phi_1 \right) \right] d^3 \vec{r} = \oint_S \left[ \nabla \cdot \left( \varepsilon \cdot \nabla \Phi_2 \right) \right] d a + \oint_S \left[ \nabla \cdot \left( \varepsilon \cdot \nabla \Phi_1 \right) \right] d a,
$$

under the symmetric condition of a dielectric tensor $\varepsilon_{ij} = \varepsilon_{ji}$. Combining with Eq. (2) and extending the finite volume to all space ($\mathbb{R}^3$), we can remove the surface integral in Eq. (3) and obtain Eq. (1). Hence we prove the Lorentz lemma in electrostatics under the symmetry condition of the dielectric tensor, that is; the optical reciprocity does not break down under the symmetry condition of the dielectric tensor ($\varepsilon_{ij} = \varepsilon_{ji}$) in the case of anisotropic local response of the medium.

### 2.1.2 Anisotropic nonlocal response

In this case, we will extend to consider the nonlocal response. Here we write the Poisson equations with two different charge densities $\rho_1$ and $\rho_2$:

$$
\nabla \cdot \left[ \int \varepsilon (\vec{r}, \vec{r}') \cdot \nabla \Phi_1 (\vec{r}') d^3 \vec{r}' \right] = -4\pi \rho_1 (\vec{r})
$$

$$
\nabla \cdot \left[ \int \varepsilon (\vec{r}, \vec{r}') \cdot \nabla \Phi_2 (\vec{r}') d^3 \vec{r}' \right] = -4\pi \rho_2 (\vec{r})
$$

and we use Eq. (A5) with $\chi (\vec{r}, \vec{r}_1) = \varepsilon (\vec{r}, \vec{r}_1)$, the value $\Phi = \Phi_1$ and $\Psi = \Phi_2$. Thus we get the following identity:

$$
\int d^3 \vec{r} \int d^3 \vec{r}_1 \left\{ \Phi_1 (\vec{r}) \nabla \cdot \left[ \varepsilon (\vec{r}, \vec{r}_1) \cdot \nabla \Phi_2 (\vec{r}_1) \right] - \Phi_2 (\vec{r}) \nabla \cdot \left[ \varepsilon (\vec{r}, \vec{r}_1) \cdot \nabla \Phi_1 (\vec{r}_1) \right] \right\}
$$

$$
= \oint_S d a \oint_s d a \left\{ \nabla \cdot \left[ \varepsilon \cdot \nabla \Phi_2 \right] \right\}
$$

under the condition $\varepsilon_{ij} (\vec{r}, \vec{r}') = \varepsilon_{ji} (\vec{r}', \vec{r})$. Next we combine Eq. (4) and extend the finite volume to $\mathbb{R}^3$; thus we can remove the surface integral in Eq. (5) and get Eq. (1) again. Hence we prove the Lorentz lemma in electrostatics under the symmetry condition of the dielectric tensor $\varepsilon_{ij} (\vec{r}, \vec{r}') = \varepsilon_{ji} (\vec{r}', \vec{r})$. Thus the optical reciprocity does not break down under the symmetry condition of the dielectric tensor in the case of anisotropic nonlocal response of the medium.
2.2 Scalar Green function

Another method to describe the optical reciprocity is the symmetry of the scalar Green function. The mathematical form is (Jackson, 1999):

$$ G(\vec{r},\vec{r}') = G(\vec{r}',\vec{r}) $$

(6)

We divide into two cases, and consider two kinds of boundary conditions to discuss the symmetry of the scalar Green function. One is the Dirichlet boundary condition and the other is the Neumann boundary condition.

2.2.1 Anisotropic local response

Referring to Eq. (2), the corresponding two equations of scalar Green function are in the following forms:

$$ \nabla \cdot \left[ \vec{e}(\vec{r}) \cdot \nabla G(\vec{r},\vec{r}') \right] = -4\pi \delta(\vec{r} - \vec{r}') $$

$$ \nabla \cdot \left[ \vec{e}(\vec{r}) \cdot \nabla G(\vec{r},\vec{r}^*) \right] = -4\pi \delta(\vec{r} - \vec{r}^*) $$

(7)

where $\vec{r}$ and $\vec{r}'$ ($\vec{r}^*$) are the positions of the field and source, respectively. $\delta$ denotes the Dirac delta function. Let us apply Eq. (A1) and put $\lambda = \vec{e}(\vec{r})$, $\Phi = G(\vec{r},\vec{r}')$ and $\Psi = G(\vec{r},\vec{r}^*)$.

Hence we have the following equality:

$$ \left\{ \left[ G(\vec{r},\vec{r}') \vec{\nabla} \cdot \left[ \vec{e}(\vec{r}) \cdot \nabla G(\vec{r},\vec{r}') \right] - G(\vec{r},\vec{r}') \vec{\nabla} \cdot \left[ \vec{e}(\vec{r}) \cdot \nabla G(\vec{r},\vec{r}^*) \right] \right] d^3\vec{r} \right\} $$

$$ = \oint_S \vec{n} \cdot \left[ G(\vec{r},\vec{r}') \vec{e}(\vec{r}) \cdot \nabla G(\vec{r},\vec{r}') - G(\vec{r},\vec{r}^*) \vec{e}(\vec{r}) \cdot \nabla G(\vec{r},\vec{r}^*) \right] da $$

(8)

under the condition $\epsilon_{ij}(\vec{r}) = \epsilon_{ji}(\vec{r})$ and we also combine Eq. (6) to get the following result:

$$ -4\pi G(\vec{r},\vec{r}') + 4\pi G(\vec{r}',\vec{r}^*) $$

$$ = \oint_S \vec{n} \cdot \left[ G(\vec{r},\vec{r}') \vec{e}(\vec{r}) \cdot \nabla G(\vec{r},\vec{r}') - G(\vec{r},\vec{r}^*) \vec{e}(\vec{r}) \cdot \nabla G(\vec{r},\vec{r}^*) \right] da $$

(9)

Next we will divide into two different boundary conditions to discuss. In the case of the Dirichlet boundary condition, we have:

$$ G(\vec{r},\vec{r}) = G(\vec{r},\vec{r}^*) = 0 $$

(10)

with $\vec{r} \in S$. Substitute this into Eq. (9) and we obtain Eq. (6), establishing the symmetry of the scalar Green function with the Dirichlet boundary condition under the symmetry condition of the dielectric tensor.

In the case of the Neumann boundary condition, let us generalize the results in Kim et al (Kim, 1993) to introduce the following Neumann boundary conditions (Xie, 2010):

$$ \vec{n} \cdot \left[ \vec{e}(\vec{r}) \cdot \nabla G_N(\vec{r},\vec{r}') \right] \bigg|_{\vec{r} \in S} = \frac{4\pi}{A} $$

$$ \vec{n} \cdot \left[ \vec{e}(\vec{r}) \cdot \nabla G_N(\vec{r},\vec{r}^*) \right] \bigg|_{\vec{r} \in S} = \frac{4\pi}{A} $$

(11)

where $A$ is the area of the closed boundary $S$. Eq. (9) then becomes:
We can then follow our previous work (Xie, 2010) to define the following symmetrized Green function:

\[ G_N^S (\vec{r}', \vec{r}) = G_N (\vec{r}', \vec{r}) - \frac{1}{A} \oint_S G_N (\vec{r}, \vec{r}') \, da , \]  
(13)

which can be shown explicitly to lead to the same solution for the potential with no contributions from the additional surface term.

### 2.2.2 Anisotropic nonlocal response

In this case, we will consider the anisotropic nonlocal response in the material. The Poisson equations with two different distributions of charge density as given in Eq. (4) will have the corresponding scalar Green functions satisfying:

\[
\begin{align*}
\int d^3 \vec{r}_1 \nabla \left[ \varepsilon (\vec{r}, \vec{r}_1) \cdot \nabla_i G (\vec{r}_1, \vec{r}') \right] &= -4\pi \delta (\vec{r} - \vec{r}') \\
\int d^3 \vec{r}_1 \nabla \left[ \varepsilon (\vec{r}, \vec{r}_1) \cdot \nabla_i G (\vec{r}_1, \vec{r}') \right] &= -4\pi \delta (\vec{r} - \vec{r}') .
\end{align*}
\]  
(14)

Next we use Eq. (A5) and let \( \tilde{\chi} = \varepsilon , \Phi (\vec{r}) = G (\vec{r}, \vec{r}') \) and \( \Psi (\vec{r}) = G (\vec{r}, \vec{r}') \), leading to:

\[
\begin{align*}
\oint_S \int d^3 \vec{r}_1 \left\{ \tilde{\chi} (\vec{r}, \vec{r}_1) \cdot \nabla_i G (\vec{r}_1, \vec{r}') - G (\vec{r}, \vec{r}') \tilde{\chi} (\vec{r}, \vec{r}_1) \cdot \nabla_i G (\vec{r}_1, \vec{r}') \right\} \\
= \oint_S \int d^3 \vec{r}_1 \left\{ \tilde{\chi} (\vec{r}, \vec{r}_1) \cdot \nabla_i G (\vec{r}_1, \vec{r}') - G (\vec{r}, \vec{r}') \tilde{\chi} (\vec{r}, \vec{r}_1) \cdot \nabla_i G (\vec{r}_1, \vec{r}') \right\} ,
\end{align*}
\]  
(15)

under the condition \( \varepsilon_{ij} (\vec{r}, \vec{r}_1) = \varepsilon_{ji} (\vec{r}, \vec{r}_1) \), combining Eqs. (14) and (15) leads to:

\[
\begin{align*}
-4\pi \chi (\vec{r}, \vec{r}') + 4\pi G (\vec{r}, \vec{r}') \\
= \oint_S \int d^3 \vec{r}_1 \left\{ \tilde{\chi} (\vec{r}, \vec{r}_1) \cdot \nabla_i G (\vec{r}_1, \vec{r}') - G (\vec{r}, \vec{r}') \tilde{\chi} (\vec{r}, \vec{r}_1) \cdot \nabla_i G (\vec{r}_1, \vec{r}') \right\} .
\end{align*}
\]  
(16)

Now we will divide into two different kinds of the boundary conditions. First we consider the Dirichlet boundary condition (Eq. (10)). The RHS of Eq. (16) becomes zero and thus we obtain the optical reciprocity under the symmetry of the dielectric tensor. Next we consider the Neumann boundary condition \( G_N \). Again, introducing the following generalized Neumann conditions (Xie, 2010):

\[
\begin{align*}
\hat{n} \cdot \int d^3 \vec{r}_1 \left[ \varepsilon (\vec{r}, \vec{r}_1) \cdot \nabla_i G_N (\vec{r}_1, \vec{r}') \right] |_{r 
abla} &= \frac{4\pi}{A} \\
\hat{n} \cdot \int d^3 \vec{r}_1 \left[ \varepsilon (\vec{r}, \vec{r}_1) \cdot \nabla_i G_N (\vec{r}_1, \vec{r}') \right] |_{r 
abla} &= -\frac{4\pi}{A} ,
\end{align*}
\]  
(17)

the Green function can be symmetrized in the form as in Eq. (13) under the following symmetric condition for the dielectric tensor: \( \varepsilon_{ij} (\vec{r}, \vec{r}') = \varepsilon_{ji} (\vec{r}, \vec{r}') \). Note further that just like the local case, this symmetrized Green function can be shown to lead to the same solution for the potential. Now we demonstrate explicitly that the newly-constructed symmeterized...
Green functions in Eq. (13) yield the same solution for the potential with no contribution from the additional surface term. To achieve this, we start with Eq. (A5) and set $\Phi = \Phi(\vec{r})$, $\Psi(\vec{r}) = G_N^S(\vec{r},\vec{r}')$, and $\vec{A}(\vec{r},\vec{r}_i) = \vec{E}(\vec{r},\vec{r}_i)$ to obtain

$$
\oint_S d\vec{F}_i \{ \vec{h} \cdot [\Phi(\vec{r})\tilde{E}(\vec{r},\vec{r}_i) \cdot \nabla, G_N^S(\vec{r},\vec{r}') - G_N^S(\vec{r},\vec{r}_i) \cdot \nabla, \Phi(\vec{r}_i)] \}
= \oint d^3\vec{r} \{ \Phi(\vec{r}) \tilde{O} \cdot [\tilde{E}(\vec{r},\vec{r}_i) \cdot \nabla, G_N^S(\vec{r}_i,\vec{r})] - G_N^S(\vec{r},\vec{r}_i) \tilde{O} \cdot [\tilde{E}(\vec{r},\vec{r}_i) \cdot \nabla, \Phi(\vec{r}_i)] \}.
$$

(18)

With Eq. (13) into the $G_N^S$ of Eq. (18) and using Eq. (14) together with the Poisson equation for $\Phi$, we obtain the following result:

$$
\Phi(\vec{r}') = \frac{1}{A} \oint d^3\vec{F}_s \Phi(\vec{r}) da + \frac{1}{4\pi} \oint d^3\vec{F}_s G_N(\vec{r},\vec{r}') \tilde{h} \cdot \tilde{E}(\vec{r},\vec{r}_i) \cdot \nabla, \Phi(\vec{r}_i)
+ \oint \rho(\vec{r}) G_N(\vec{r},\vec{r}') d^3\vec{r}' + F(\vec{r}') \left\{ \frac{1}{4\pi} \oint d^3\vec{F}_s \tilde{h} \cdot \tilde{E}(\vec{r},\vec{r}_i) \cdot \nabla, \Phi(\vec{r}_i) \right\},
$$

(19)

$$
= \frac{1}{A} \oint_d^3\vec{F}_s \Phi(\vec{r}) da + \frac{1}{4\pi} \oint_d^3\vec{F}_s \oint d^3\vec{F}_s G_N(\vec{r},\vec{r}') \tilde{h} \cdot \tilde{E}(\vec{r},\vec{r}_i) \cdot \nabla, \Phi(\vec{r}_i)
+ \oint \rho(\vec{r}) G_N(\vec{r},\vec{r}') d^3\vec{r}'
$$

where the surface term $F(\vec{r}')$ has no contribution since the term $\{\ldots\}$ in Eq. (18) vanishes based on the nonlocal version of the Gauss law for $\Phi$ [See Eq. (4)]. Thus the symmetrized $G_N^S$ leads to the same potential as the one obtained from using $G_N$.

### 2.3 Some examples for scalar Green functions

We will give some examples to display the explicit forms of the Green function in both Dirichlet and Neumann boundary conditions (Chang 2008; Xie, 2010). Consider a metal sphere (radius $a$) with an isotropic (for simplicity) but nonlocal dielectric response $\varepsilon(k,\omega)$.

For the case of Dirichlet condition, we have previously applied the model of the nonlocal polarizability by Fuchs and Claro (Fuchs & Claro, 1987) to obtain the following symmetric Green function:

$$
G(\vec{r},\vec{r}') = 4\pi \sum_{\ell=0}^{\infty} \sum_{m=-\infty}^{\infty} \frac{1}{2\ell+1} \left[ \frac{r_\ell'}{r_\ell} - \frac{\alpha_{\ell\ell}^{NL}}{(2\ell+1)^{\ell+1}} \right] Y_{\ell m}(\theta',\varphi') Y_{\ell m}(\theta,\varphi),
$$

(20)

where $(r_\ell, r_\ell')$ denote the smaller or greater of $(r, r')$, and

$$
\alpha_{\ell\ell}^{NL} = \frac{\xi_{\ell+1} - 1}{\xi_{\ell} + (\ell+1)/(\ell+1)} a^{2\ell+1},
$$

(21)

with the “effective dielectric function” given by:

$$
\xi_{\ell}(\omega) = \left[ \frac{2(2\ell+1)a}{\pi} \int_0^\infty j_{\ell+1}^2(ka) \frac{1}{\varepsilon(k,\omega)} dk \right]^{-1}.
$$

(22)

For the same problem under the Neumann condition, first we get the following Green function by solving the corresponding boundary value problem:
which does not satisfy Eq. (6). However, we can use the previous methods to symmetrize the above asymmetric Green function. Applying Eq. (13) to calculate the surface term using only the first two terms in Eq. (23) and obtain the following:

$$\frac{1}{\mathcal{A}} \oint_{S} G_{N}(\bar{r}, \bar{r}') \, da = \frac{1}{r} - \frac{1}{a}.$$  \hfill (24)

We thus obtain the final symmetrized Neumann Green function for the region outside a nonlocal metal sphere in the following form:

$$G_{N}^{S}(\bar{r}, \bar{r}') = \left[ \frac{1}{r_{\infty}} - \left( \frac{1}{r} + \frac{1}{r_{\infty}} \right) + \frac{1}{a} \right]$$

$$+ 4 \pi \sum_{\ell=1}^{\infty} \sum_{m=-\ell}^{\ell} \frac{1}{2 \ell + 1} \left[ \frac{r_{\infty}^{\ell}}{r_{\infty}^{\ell+1}} + \frac{\alpha_{NL}^{\infty}}{\ell + 1 (r_{\infty}^{\ell+1})^{\ell+1}} \right] Y_{\ell m}^{*}(\theta', \varphi') Y_{\ell m}(\theta, \varphi).$$ \hfill (25)

In our examples, we can find that the optical reciprocity does not break down since the dielectric function satisfies $\varepsilon (\bar{r}, \bar{r}') = \varepsilon (|\bar{r} - \bar{r}'|)$.

### 3. Reciprocity in electrodynamics

In the previous discussion, we have restricted our problem to the “long wavelength approximation” in which electrostatics has been applied (Chang, 2008). In problems with very high frequency source (e.g. scattering between X-rays and a nanoparticle (Ruppin, 1975)) in which electrostatics breaks down and nonlocal effects can become even more significant due to the large value of the wavevector, the previous formulation (Chang, 2008) becomes inadequate. Here we use the method of exact electrodynamics to study the optical reciprocity. Again, we shall refer to both the Lorentz lemma in electrodynamics and the symmetry of dyadic Green function.

#### 3.1 Lorentz lemma in electrodynamics

The mathematical form of the Lorentz lemma in electrodynamics is as follows (Landau et al., 1984):

$$\int \bar{J}_{1}(\bar{r}) \cdot \bar{E}_{2}(\bar{r}) \, d^{3}\bar{r} = \int \bar{J}_{2}(\bar{r}) \cdot \bar{E}_{1}(\bar{r}) \, d^{3}\bar{r},$$ \hfill (26)

where $\bar{E}_{1}$ ($\bar{E}_{2}$) is the electric field resulting from the current density $\bar{J}_{1}$ ($\bar{J}_{2}$). Next we will derive this formula in two different cases.

#### 3.1.1 Anisotropic local response

In the beginning, let us consider time harmonic Maxwell’s equations ($-e^{-i\omega t}$):
Now we consider two different sources $\mathbf{J}_1$ and $\mathbf{J}_2$ which correspond to two electric fields $\mathbf{E}_1$ and $\mathbf{E}_2$ and two magnetic fields $\mathbf{H}_1$ and $\mathbf{H}_2$, respectively. We obtain from Eq. (27) the following result:

$$
\mathbf{H}_2 \cdot \nabla \times \mathbf{E}_1 - \mathbf{E}_1 \cdot \nabla \mathbf{H}_2 + \mathbf{E}_2 \cdot \nabla \times \mathbf{H}_1 - \mathbf{H}_1 \cdot \nabla \times \mathbf{E}_2 = \frac{i\omega}{c} \left[ (\mathbf{B}_1 \cdot \mathbf{H}_2 - \mathbf{H}_1 \cdot \mathbf{B}_2) + (\mathbf{E}_1 \cdot \mathbf{D}_2 - \mathbf{D}_1 \cdot \mathbf{E}_2) \right] + \frac{4\pi}{c} \left( \mathbf{J}_1 \cdot \mathbf{E}_2 - \mathbf{J}_2 \cdot \mathbf{E}_1 \right),
$$

(28)

which can be simplified to the following form:

$$
\nabla \cdot \left( \mathbf{H}_1 \times \mathbf{E}_2 - \mathbf{E}_1 \times \mathbf{H}_2 \right) = \frac{4\pi}{c} \left( \mathbf{J}_1 \cdot \mathbf{E}_2 - \mathbf{J}_2 \cdot \mathbf{E}_1 \right),
$$

(29)

if the dielectric tensor satisfies the symmetry condition $\epsilon_{ij} = \epsilon_{ji}$ and the permeability satisfies the symmetry condition $\mu_{ij} = \mu_{ji}$. Furthermore, let us integrate over $\mathbb{R}^3$ in Eq. (29) and use the divergence theorem to convert the left side of Eq. (29) into a surface integral which can be removed. Hence we obtain the Lorentz lemma in electrodynamics (i.e. Eq. (26)).

### 3.1.2 Anisotropic nonlocal response

Next we will extend the local response to the nonlocal response where the relation between the auxiliary field and the magnetic field is described by

$$
\mathbf{H}(\mathbf{r}) = \int d^3\mathbf{r}' \hat{\mathbf{\mu}}^{-1}(\mathbf{r}, \mathbf{r}') \cdot \mathbf{B}(\mathbf{r}').
$$

(30)

Thus Eq. (28) will become:

$$
\nabla \cdot \left( \mathbf{H}_1 \times \mathbf{E}_2 - \mathbf{E}_1 \times \mathbf{H}_2 \right) = \frac{4\pi}{c} \int d^3\mathbf{r}' \left\{ \hat{\mathbf{B}}_1(\mathbf{r}) \cdot \hat{\mathbf{\mu}}^{-1}(\mathbf{r}, \mathbf{r}') \cdot \hat{\mathbf{B}}_2(\mathbf{r}') - \hat{\mathbf{\mu}}^{-1}(\mathbf{r}, \mathbf{r}') \cdot \hat{\mathbf{B}}_1(\mathbf{r}') \cdot \hat{\mathbf{B}}_2(\mathbf{r}) \right\} + \frac{4\pi}{c} \left( \mathbf{J}_1 \cdot \mathbf{E}_2 - \mathbf{J}_2 \cdot \mathbf{E}_1 \right),
$$

(31)

We integrate over $\mathbb{R}^3$ and use the divergence theorem to remove the surface term again. Thus Eq. (31) becomes:

$$
\frac{4\pi}{c} \int (\mathbf{J}_1 \cdot \mathbf{E}_2 - \mathbf{J}_2 \cdot \mathbf{E}_1) d^3\mathbf{r}
$$

$$
= \frac{i\omega}{c} \int d^3\mathbf{r} \int d^3\mathbf{r}' \left\{ \hat{\mathbf{B}}_1(\mathbf{r}) \cdot \hat{\mathbf{\mu}}^{-1}(\mathbf{r}, \mathbf{r}') \cdot \hat{\mathbf{B}}_2(\mathbf{r}') - \hat{\mathbf{\mu}}^{-1}(\mathbf{r}, \mathbf{r}') \cdot \hat{\mathbf{B}}_1(\mathbf{r}') \cdot \hat{\mathbf{B}}_2(\mathbf{r}) \right\},
$$

(32)

$$
+ \frac{i\omega}{c} \int d^3\mathbf{r} \int d^3\mathbf{r}' \left\{ \hat{\mathbf{E}}_1(\mathbf{r}) \cdot \hat{\mathbf{\varepsilon}}(\mathbf{r}, \mathbf{r}') \cdot \hat{\mathbf{E}}_2(\mathbf{r}') - \hat{\mathbf{\varepsilon}}(\mathbf{r}, \mathbf{r}') \cdot \hat{\mathbf{E}}_1(\mathbf{r}') \cdot \hat{\mathbf{E}}_2(\mathbf{r}) \right\},
$$
which can be simplified to get the Lorentz lemma in electrodynamics under the symmetry condition of the dielectric tensor $\epsilon_\mu (\vec{r}, \vec{r}') = \epsilon_{\mu'} (\vec{r}', \vec{r})$ and the permeability tensor $\mu_\mu (\vec{r}, \vec{r}') = \mu_{\mu'} (\vec{r}', \vec{r})$.

### 3.2 Dyadic Green function

The other method to describe the optical reciprocity is the symmetry of dyadic Green function and the mathematical form is the following form (Tai, 1993; Xie, 2009a, 2009b):

\[
\left[ \tilde{C}_e (\vec{r'}, \vec{r}) \right]^T = \tilde{C}_e (\vec{r}, \vec{r'}),
\]  

where $T$ is the transport. We shall establish our results in two steps and restrict ourselves to the case of boundary conditions for the dyadic Green function.

#### 3.2.1 Anisotropic local response

First we consider only local response which is simpler and sets the framework for the treatment of the more complicated nonlocal case. Thus we assume the following constitutive relations:

\[
\vec{D}(\vec{r}) = \vec{\varepsilon}(\vec{r}) \cdot \vec{E}(\vec{r}) \quad \text{and} \quad \vec{B}(\vec{r}) = \vec{\mu}(\vec{r}) \cdot \vec{H}(\vec{r}).
\]

For fields with time harmonic dependence, we have the following vector identity:

\[
\nabla \times \vec{\mu}^{-1}(\vec{r}) \cdot \nabla \times \vec{E}(\vec{r}) - \frac{\alpha^2}{c^2} \vec{E}(\vec{r}) \cdot \vec{E}(\vec{r}) = i\omega \frac{4\pi}{c} \vec{J}(\vec{r}),
\]

which implies the following differential equation for the electric dyadic Green function of the problem:

\[
\nabla \times \vec{\mu}^{-1}(\vec{r}) \cdot \nabla \times \tilde{C}_e (\vec{r}, \vec{r'}) - \frac{\alpha^2}{c^2} \tilde{C}_e (\vec{r}, \vec{r'}) \cdot \tilde{C}_e (\vec{r}, \vec{r'}) = \frac{4\pi}{c} \delta(\vec{r} - \vec{r'}).
\]

Using Eq. (A9), we set $\tilde{A} = \tilde{C}_e (\vec{r}, \vec{r'})$, $\tilde{B} = \tilde{C}_e (\vec{r}, \vec{r})$ and $\tilde{A} = \vec{\mu}^{-1}(\vec{r})$ to obtain the following identity:

\[
\int \left[ \nabla \times \vec{\mu}^{-1} \cdot \nabla \times \tilde{C}_e (\vec{r}, \vec{r'}) \right]^T \cdot \tilde{C}_e (\vec{r}, \vec{r'}) - \left[ \tilde{C}_e (\vec{r}, \vec{r'}) \right]^T \cdot \nabla \times \vec{\mu}^{-1} \cdot \nabla \times \tilde{C}_e (\vec{r}, \vec{r'}) \right] d^3\vec{r}
= \oint \left[ \nabla \times \vec{\mu}^{-1} \cdot \nabla \times \tilde{C}_e (\vec{r}, \vec{r'}) \right]^T \cdot \tilde{C}_e (\vec{r}, \vec{r'}) - \left[ \tilde{C}_e (\vec{r}, \vec{r'}) \right]^T \cdot \nabla \times \vec{\mu}^{-1} \cdot \nabla \times \tilde{C}_e (\vec{r}, \vec{r'}) \right] d\vec{a}
\]

Hence from either the dyadic Dirichlet condition:

\[
\hat{n} \times \tilde{C}_e (\vec{r}, \vec{r'}) = 0,
\]

or the dyadic Neumann condition\(^1\):

\[
\hat{n} \times \left[ \vec{\mu}^{-1} \cdot \nabla \times \tilde{C}_e (\vec{r}, \vec{r'}) \right] = 0,
\]

\(^1\) Note that the condition introduced in Eq. (38) is a generalization of $\hat{n} \times \left[ \nabla \times \tilde{C}_e (\vec{r}, \vec{r'}) \right] = 0$ with the explicit inclusion of the magnetic permeability tensor.
Eq. (36) leads to:
\[
\int \left[ \nabla \times \bar{\mu}^{-1} \cdot \nabla \times \bar{G}_e(\bar{r}, \bar{r}^\prime) \right]^T \cdot \bar{G}_e(\bar{r}, \bar{r}^\prime) - \left[ \bar{G}_e(\bar{r}, \bar{r}^\prime) \right]^T \cdot \nabla \times \bar{\mu}^{-1} \cdot \nabla \times \bar{G}_e(\bar{r}, \bar{r}^\prime) \right] d^3\bar{r} = 0. \tag{39}
\]
Substituting Eq. (35) into Eq. (39), we have:
\[
\int \left\{ \frac{\omega^2}{c^2} \left[ \bar{e}(\bar{r}) \cdot \bar{G}_e(\bar{r}, \bar{r}^\prime) \right]^T + \frac{4\pi}{c} \bar{I} \delta(\bar{r} - \bar{r}^\prime) \right\} \cdot \bar{G}_e(\bar{r}, \bar{r}^\prime) d^3\bar{r} - \int \left[ \bar{G}_e(\bar{r}, \bar{r}^\prime) \right]^T \cdot \left\{ \frac{\omega^2}{c^2} \bar{e}(\bar{r}) \cdot \bar{G}_e(\bar{r}, \bar{r}^\prime) + \frac{4\pi}{c} \bar{I} \delta(\bar{r} - \bar{r}^\prime) \right\} d^3\bar{r} = 0
\]
which implies the symmetry of the dyadic Green function. Eq. (39) provides that the dielectric tensor is symmetric: \( \epsilon_{ij}(\bar{r}) = \epsilon_{ji}(\bar{r}) \). We remark that the validity of Eq. (36) has already required \( \mu_{ij}(\bar{r}) = \mu_{ji}(\bar{r}) \) as explained before.

### 3.2.2 Anisotropic nonlocal response

Now we consider both electric and magnetic nonlocal responses and thus generalize the dyadic differential equation in Eq. (35) to:
\[
\nabla \times \int \bar{\mu}^{-1}(\bar{r}, \bar{r}_1) \cdot \nabla \times \bar{G}_e(\bar{r}_1, \bar{r}^\prime) d^3\bar{r}_1 - \frac{\omega^2}{c^2} \int \bar{e}(\bar{r}_1) \cdot \bar{G}_e(\bar{r}_1, \bar{r}^\prime) d^3\bar{r}_1 = \frac{4\pi}{c} \bar{I} \delta(\bar{r} - \bar{r}^\prime). \tag{41}
\]
Using Eq. (A15), we set
\[
\bar{A}(\bar{r}) = \bar{G}_e(\bar{r}, \bar{r}^\prime), \quad \bar{B}(\bar{r}) = \bar{G}_e(\bar{r}, \bar{r}^\prime), \quad \text{and} \quad \bar{\lambda} = \bar{\mu}^{-1}
\]
and obtain the following form:
\[
\int d^3\bar{r} \int d^3\bar{r}_1 \left\{ \nabla \times \bar{\mu}^{-1}(\bar{r}, \bar{r}_1) \cdot \nabla \times \bar{G}_e(\bar{r}_1, \bar{r}^\prime) \right\}^T \cdot \bar{G}_e(\bar{r}, \bar{r}^\prime)
- \int d^3\bar{r} \int d^3\bar{r}_1 \left\{ \bar{G}_e(\bar{r}, \bar{r}^\prime) \right\}^T \cdot \nabla \times \bar{\mu}^{-1}(\bar{r}, \bar{r}_1) \cdot \nabla \times \bar{G}_e(\bar{r}_1, \bar{r}^\prime)
= \frac{4\pi}{c} \bar{I} \delta(\bar{r} - \bar{r}^\prime). \tag{42}
\]
Again, with either Dirichlet or Neumann conditions\(^2\), we obtain from Eq. (42):
\[
\int d^3\bar{r} \int d^3\bar{r}_1 \left\{ \nabla \times \bar{\mu}^{-1}(\bar{r}, \bar{r}_1) \cdot \nabla \times \bar{G}_e(\bar{r}_1, \bar{r}^\prime) \right\}^T \cdot \bar{G}_e(\bar{r}, \bar{r}^\prime)
- \int d^3\bar{r} \int d^3\bar{r}_1 \left\{ \bar{G}_e(\bar{r}, \bar{r}^\prime) \right\}^T \cdot \nabla \times \bar{\mu}^{-1}(\bar{r}, \bar{r}_1) \cdot \nabla \times \bar{G}_e(\bar{r}_1, \bar{r}^\prime)
= 0. \tag{43}
\]
\(^2\) The Neumann condition in Eq. (38) in the nonlocal case has also to be generalized to the following form: \( \int \hat{n} \times \bar{\mu}^{-1}(\bar{r}, \bar{r}_1) \cdot \nabla \times \bar{G}_e(\bar{r}_1, \bar{r}^\prime) d^3\bar{r} = 0 \)
Substituting Eq. (41) into Eq. (43), we have the following identity:

$$\begin{align*}
\int \left[ \frac{\alpha^2}{c^2} \int \tilde{\varepsilon}(\vec{r}, \vec{r}_1) \cdot \tilde{G}_c(\vec{r}, \vec{r}_1, \vec{r}^*) d^3 \vec{r}_1 + \frac{4\pi i}{c} \delta(\vec{r} - \vec{r}^*) \right] \cdot \tilde{G}_c(\vec{r}, \vec{r}) d^3 \vec{r} \\
= \int \left[ \tilde{G}_c(\vec{r}, \vec{r}^*) \right]^T \cdot \left\{ \frac{\alpha^2}{c^2} \int \tilde{\varepsilon}(\vec{r}, \vec{r}_1) \cdot \tilde{G}_c(\vec{r}, \vec{r}_1, \vec{r}^*) d^3 \vec{r}_1 + \frac{4\pi i}{c} \delta(\vec{r} - \vec{r}^*) \right\} d^3 \vec{r}
\end{align*}$$

which again implies Eq. (33) in a way that is similar to the above case for local response.

4. The equivalence of Lorentz lemma and Green function formulation

So far, we have shown two different mathematical formulations for discussing the optical reciprocity. Now the question is: are these two statements equivalent? Now we give a proof.

4.1 Electrostatics

First we demonstrate the equivalence between Lorentz lemma and the symmetry of the scalar Green function in electrostatics, by starting with a slightly more general form of Eq. (1) with the surface terms retained:

$$\int \left( \rho_1 \Phi_2 - \rho_2 \Phi_1 \right) d^3 \vec{r} = \frac{1}{4\pi} \oint_S \hat{n} \cdot \left( \Phi_1 \tilde{\varepsilon} \cdot \nabla \Phi_2 - \Phi_2 \tilde{\varepsilon} \cdot \nabla \Phi_1 \right) da .$$

Note that the above can be applied to the finite boundary region. To demonstrate the equivalence between Eq. (1) and Eq. (6), let us consider two unit point charge distribution as follows:

$$\rho_1 = \delta(\vec{r} - \vec{r}^*), \quad \rho_2 = \delta(\vec{r} - \vec{r}') ,$$

and the potentials at each of their locations are then given by the scalar Green function:

$$\Phi_1 = G(\vec{r}, \vec{r}^*) , \quad \Phi_2 = G(\vec{r}, \vec{r}') .$$

Substituting Eqs. (46) and (47) into Eq. (45) leads to the following result:

$$G(\vec{r}^*, \vec{r}') - G(\vec{r}', \vec{r}^*) = \frac{1}{4\pi} \oint_S \hat{n} \cdot \left[ G(\vec{r}, \vec{r}') \tilde{\varepsilon} \cdot \nabla G(\vec{r}, \vec{r}') - G(\vec{r}, \vec{r}') \tilde{\varepsilon} \cdot \nabla G(\vec{r}, \vec{r}) \right] da .$$

Note that the proof of the equivalence between the two versions of the reciprocity principle in the previous section remains valid for the case with nonlocal response, with Eq. (48) generalized to the following form:

$$G(\vec{r}^*, \vec{r}') - G(\vec{r}', \vec{r}^*) = \frac{1}{4\pi} \oint_S da \int d^3 \vec{r}_1 \left\{ \hat{n} \cdot \left[ G(\vec{r}, \vec{r}^*) \tilde{\varepsilon}(\vec{r}, \vec{r}_1) \cdot \nabla_1 G(\vec{r}_1, \vec{r}') - G(\vec{r}, \vec{r}') \tilde{\varepsilon}(\vec{r}, \vec{r}_1) \cdot \nabla_1 G(\vec{r}_1, \vec{r}^*) \right] \right\} .$$
Here we separate into two different kinds of the boundary conditions to discuss:
First, with the Dirichlet boundary condition given in Eq. (10) substituted into Eq. (48), we obtain Eq. (6). Thus we have demonstrated the equivalence between the Lorentz lemma in electrostatics and the scalar Green function under the Dirichlet boundary condition.
Second, the Neumann boundary condition is given by Eq. (11) and thus Eq. (48) becomes the following form:

\[ G_N(\vec{r},\vec{r}') - G_N(\vec{r}',\vec{r}) = -\frac{1}{A} \oint_S G_N(\vec{r},\vec{r}') da + \frac{1}{A} \oint_S G_N(\vec{r},\vec{r}') da. \]  

(49)

By the pervious method, we can establish the symmetry of the scalar Green function as shown in Eq. (6).

4.2 Electrodynamics
Next we will show that the equivalence between these two statements which are the optical reciprocity in the form of Lorentz lemma in electrodynamics and of the symmetry of the dyadic Green function. To demonstrate this equivalence, first we start from Lorentz lemma in electrodynamics by retaining the surface terms (Xie, 2009b):

\[ \frac{4\pi}{c} \int \left( \vec{J}_1 \cdot \vec{E}_2 - \vec{J}_2 \cdot \vec{E}_1 \right) d^2\vec{r} = \oint_S \hat{n} \cdot \left( \vec{E}_1 \cdot \vec{H}_2 - \vec{E}_2 \cdot \vec{H}_1 \right) da. \]  

(50)

Note that Eq. (50) is a direct consequence from Maxwell’s equations and the surface terms are kept to allow for the presence of finite boundaries and nontrivial material with both permittivity and permeability. Although these surface terms are usually discarded (Kahl & Voges, 2000; Ru & Etchegoin, 2006; Landau et al., 1984; Iwanaga et al., 2007), they have also been considered in some studies in the literatures (Xie et al., 2009; Porto et al., 2000; Joe et al., 2008). Hence we must keep them to demonstrate the exact equivalence between the two versions of optical reciprocity.

In the beginning, let us consider two unit point current sources due to electric dipole (with moment \( p \)) as follows:

\[ \vec{J}_1 = -i\omega p \delta(\vec{r} - \vec{r}') \hat{e}_i, \]
\[ \vec{J}_2 = -i\omega p \delta(\vec{r} - \vec{r}') \hat{e}_j. \]

(51)

and the electric fields at each of their locations are given in terms of the column component of the dyad as follows:

\[ \vec{E}_1 = \frac{\omega^2 p}{c} \vec{G}_a(\vec{r},\vec{r}') \], \quad \vec{E}_2 = \frac{\omega^2 p}{c} \vec{G}_a(\vec{r},\vec{r}') . \]

(52)

Substituting Eqs. (51) and (52) into Eq. (50) leads to the following result:

\[ -\frac{4\pi\omega}{c} \left[ \hat{e}_i \cdot \vec{G}_a(\vec{r},\vec{r}') - \hat{e}_j \cdot \vec{G}_a(\vec{r}',\vec{r}) \right] \]
\[ = \oint_S \hat{n} \cdot \left[ \vec{G}_a(\vec{r},\vec{r}') \times \vec{H}_2(\vec{r}) - \vec{G}_a(\vec{r}',\vec{r}) \times \vec{H}_1(\vec{r}) \right] da. \]  

(53)
Hence using Maxwell’s equations and the vector triple product, we obtain:

\[
-\frac{4\pi i\omega}{c} \left[ \left[ \bar{G}_r(\bar{r}',\bar{r}) \right]_{ij} - \left[ \bar{G}_r(\bar{r'},\bar{r}) \right]_{ji} \right] = \oint_S \left[ \hat{n} \times \bar{G}_{ai}(\bar{r},\bar{r}') \right] - \left[ \hat{n} \times \bar{G}_{aj}(\bar{r},\bar{r}') \right] d\mathbf{a} 
\]

\[
\frac{\partial}{\partial t} \oint_S \left[ \hat{n} \times \bar{G}_{ai}(\bar{r},\bar{r}') \right] - \left[ \hat{n} \times \bar{G}_{aj}(\bar{r},\bar{r}') \right] d\mathbf{a} = \frac{\omega}{c} \oint_S \left[ \hat{n} \times \bar{G}_{ai}(\bar{r},\bar{r}') \right] - \left[ \hat{n} \times \bar{G}_{aj}(\bar{r},\bar{r}') \right] d\mathbf{a} 
\]

\[
= \frac{\omega}{c} \oint_S \left[ \left[ \hat{n} \times \bar{G}_{ai}(\bar{r},\bar{r}') \right] - \left[ \hat{n} \times \bar{G}_{aj}(\bar{r},\bar{r}') \right] \right] d\mathbf{a} 
\]

Hence we have:

\[
\frac{4\pi}{c} \left[ \left[ \bar{G}_r(\bar{r}',\bar{r}) \right]_{ij} - \left[ \bar{G}_r(\bar{r'},\bar{r}) \right]_{ji} \right] = \oint_S \left[ \hat{n} \times \bar{G}_{i}(\bar{r},\bar{r}') \right] - \left[ \hat{n} \times \bar{G}_{j}(\bar{r},\bar{r}') \right] \left[ \bar{G}_r(\bar{r},\bar{r}') \right] \left[ \bar{G}_r(\bar{r},\bar{r}') \right] d\mathbf{a} 
\]

We can rewrite Eq. (55) in dyadic form as follows:

\[
\frac{4\pi}{c} \left[ \left[ \bar{G}_r(\bar{r}',\bar{r}) \right] - \left[ \bar{G}_r(\bar{r'},\bar{r}) \right] \right] = \oint_S \left[ \left[ \hat{n} \times \bar{G}_{i}(\bar{r},\bar{r}') \right] - \left[ \hat{n} \times \bar{G}_{j}(\bar{r},\bar{r}') \right] \right] \left[ \bar{G}_r(\bar{r},\bar{r}') \right] \left[ \bar{G}_r(\bar{r},\bar{r}') \right] d\mathbf{a} 
\]

By imposing on \( S \) either the dyadic Dirichlet condition (Eq. (37)) or the dyadic Neumann condition (Eq. (38)), the surface integral in Eq. (56) can be made vanished by applying the dyadic triple product in the Neumann case. Hence under either one of these boundary conditions, Eq. (56) will lead to the symmetric property of the dyadic Green function in Eq. (33).

5. Some examples

We have established the general conditions for optical reciprocity to hold in nonlocal optics from the method of electrostatics to electrodynamics. The general conditions are:

\[\text{Note that the proof of the equivalence between the two versions of the reciprocity principle in the previous section remains valid for the case with nonlocal response, with Eq. (54) generalized to the following form:}\]

\[
-\frac{4\pi i\omega}{c} \left[ \left[ \bar{G}_r(\bar{r}',\bar{r}) \right]_{ij} - \left[ \bar{G}_r(\bar{r'},\bar{r}) \right]_{ji} \right] 
\]

\[
= \frac{\omega}{c} \oint_S \left[ \hat{n} \times \bar{G}_{ai}(\bar{r},\bar{r}') \right] - \left[ \hat{n} \times \bar{G}_{aj}(\bar{r},\bar{r}') \right] d\mathbf{a} 
\]

\[
-\frac{\omega}{c} \oint_S \left[ \hat{n} \times \bar{G}_{ai}(\bar{r},\bar{r}') \right] - \left[ \hat{n} \times \bar{G}_{aj}(\bar{r},\bar{r}') \right] d\mathbf{a} 
\]

\[
\frac{\omega}{c} \oint_S \left[ \hat{n} \times \bar{G}_{ai}(\bar{r},\bar{r}') \right] - \left[ \hat{n} \times \bar{G}_{aj}(\bar{r},\bar{r}') \right] d\mathbf{a} 
\]
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\[
\varepsilon_{ij}(\vec{r}, \vec{r}') = \varepsilon_{ji}(\vec{r}', \vec{r}) \\
\mu_{ij}(\vec{r}, \vec{r}') = \mu_{ji}(\vec{r}', \vec{r})
\]

which are the extension conditions of local optics. This reduces to the well-known local limit which requires only a symmetric local dielectric tensor for the validity of reciprocity (Chang, 2008; Iwanaga, 2007). It also reduces to the isotropic nonlocal case which is known to be valid for most of the well-known nonlocal quantum mechanical models for a homogeneous electron gas, such as the Linhard-Mermin function in which \( \varepsilon(\vec{r}, \vec{r}') = \varepsilon(\vec{|r - r|}) \) (Chang, 2008). Moreover, we also give two interesting examples that may lead to the breakdown of the reciprocity in linear optics. One example is that the following dielectric tensor:

\[
\varepsilon = \begin{pmatrix}
\varepsilon_x & -ig & 0 \\
ig & \varepsilon_y & 0 \\
0 & 0 & \varepsilon_z
\end{pmatrix},
\]

which is hermitian but not symmetric (Vlokh & Adamenko, 2008). Another example is to refer to the case studied in the literature (Malinowski et al., 1996) which involved the propagation of light along a cubic axis in a crystal of 23 point group. In this case, the nonlocality tensor \( \gamma_{ij} \) may be asymmetric in the sense that \( \gamma_{ij} \neq \gamma_{ji} \), which can be shown to imply an asymmetric dielectric tensor \( \varepsilon_{ij} \neq \varepsilon_{ji} \). Here we give a proof. With the dielectric function becoming a tensor, we have:

\[
D_i(\vec{r}) = \int \varepsilon_{ij}(\vec{r}, \vec{r}') \cdot E_j(\vec{r}') d^3\vec{r}'.
\]

Next we change the variable \( \vec{r}' = \vec{r} + \vec{a} \) and use a Taylor series for the electric field to obtain the following form:

\[
D_i(\vec{r}) = \int \varepsilon_{ij}(\vec{r}, \vec{r} + \vec{a}) E_j(\vec{r} + \vec{a}) d^3\vec{a}.
\]

For case of weak nonlocality, where \( \varepsilon_{ij}(\vec{r}, \vec{r} + \vec{a}) \neq 0 \) only for \( \vec{a} \) within a small neighborhood of \( \vec{r} \), higher order terms in Eq. (60) can be neglected, and we recover the identity which has occurred in Eq. (1) of the literature (Malinowski et al., 1996):

\[
D_i(\vec{r}) = E_i(\vec{r}) \int \varepsilon_{ij}(\vec{r}, \vec{r} + \vec{a}) d^3\vec{a} + \partial_x E_j(\vec{r}) \int \varepsilon_{ij}(\vec{r}, \vec{r} + \vec{a}) a_x d^3\vec{a}
\]

where the first term and second term of the above equation denote the contribution of locality and nonlocality, respectively. Since the nonlocality tensor \( \gamma_{ij} \) satisfies the relation \( \gamma_{ij} \neq \gamma_{ji} \), we conclude that the electric tensor \( \varepsilon_{ij} \) satisfies the relation \( \varepsilon_{ij} \neq \varepsilon_{ji} \). However, this is the same with what was studied in the literature (Malinowski et al., 1996), where
nonlocality through the field gradient dependent response is required to break reciprocity symmetry for the rotation of the polarization plane of the transmitted wave. In their statement, if the nonlocality $\gamma_{ij}$ satisfies the relation $\gamma_{ij} = \gamma_{ji}$, optical reciprocity breaks down. In our viewpoint, From Eq. (61), the relation $\gamma_{ij} = \gamma_{ji}$ implies the relation $\epsilon_{ij}(\vec{r}, \vec{r} + \vec{a}) = \epsilon_{ji}(\vec{r}, \vec{r} + \vec{a})$ where violates Eq. (57). Thus the reciprocity may break down. Hence our mathematical formulations provide a general examination to determine if the optical reciprocity remain or break down initially.

6. Application to spectroscopic analysis

In this section, we demonstrate the application of the reciprocity symmetry in the form of the Lorentz lemma for two dipolar sources (in obvious notations):

$$\vec{p}_1 \cdot \vec{E}_2 = \vec{p}_2 \cdot \vec{E}_1, \quad (62)$$

Fig. 1. Spectrum of the local field and radiation enhancement factors, with the latter plotted for both radial and tangential molecular dipoles, according to both the local (dashed lines) and nonlocal (solid lines) SERS models. The molecular dipole is located at a distance of 1 nm from a silver nanosphere of 5 nm radius to the calculation of the various surface-enhanced Raman scattering (SERS) enhancement factors from a molecule adsorbed on a metallic nanoparticle following the recent work of Le Ru and Etchegoin. As pointed out by Le Ru and Etchegoin (Ru & Etchegoin, 2006), in any SERS analysis, one must distinguish carefully between the local field and the radiation enhancement since ‘... the induced molecular Raman dipole is not necessarily aligned
parallel to the electric field of the pump beam . . . . Based on this distinction, it was proposed in the literature (Ru & Etchegoin, 2006) that the more correct SERS enhancement ratio should be a product of these two enhancement factors: \( \tilde{M}_{SERS} = \tilde{M}_{Loc} \cdot \tilde{M}_{Rad} \) with the latter enhancement calculable from an application of Eq. (62). This formulation has then corrected a conventional misconception in the literature of SERS theory with models exclusively based on the fourth power dependence of the local field.

In Fig. 1, we have essentially reproduced the key features in the corresponding Fig. 1 of the literature (Ru & Etchegoin, 2006), but for a much smaller metal sphere (radius = 5 nm) so that nonlocal effects are more pronounced. Note that in this figure, Eq. (21) has been used to calculate the various quantities represented by solid lines and we note that, with the nonlocal response of the metal particle, the sharp differences between \( \tilde{M}_{Loc} \) and \( \tilde{M}_{Rad} \) remain for the tangentially oriented dipoles, as was first observed in the literature (Ru & Etchegoin, 2006). The radially oriented dipole, however, gives very similar results for both the enhancement factors in both our nonlocal calculation and the local one as reported in the literature (Ru & Etchegoin, 2006). Note that the nonlocal effects are most significant in the vicinity of the plasmon resonance frequency, with the peaks slightly blueshifted due mainly to the semiclassical infinite barrier (SCIB) approximation adopted in this model (Fuchs & Claro, 1987).

7. Conclusion

Fig. 2. The description of optical reciprocity in four different distributions of the material media

We have constructed the conditions for optical reciprocity in the case with a nonlocal anisotropic magnetic permeability and electric permittivity, motivated by the recent explosion in the research with metamaterials according to two different mathematical viewpoints (Lorentz lemma and Green function formulation) furthermore that are
equivalent. These results reduce to the well-known conditions in the case of local response. Note that while the symmetry in $\vec{r}$ and $\vec{r}'$ will be valid for most materials on a macroscopic scale (Jenkins & Hunt, 2003), that in the tensorial indices will not be valid in general for complex materials such as bianisotropic or chiral materials (Kong, 2003). Importantly, our mathematical formulations provide a general examination to determine if the optical reciprocity remain or break down initially. However, it will be of interest to design some optical experiment to observe the breakdown of reciprocity symmetry with these systems in the study of metamaterials. One possible way is to observe transmission asymmetry in the light propagating through these materials as shown in Fig. 2 which shows this interesting process and lists four different distributions of the material media. According to our pervious mathematical prediction, we will have optical reciprocity still remains valid in (a), (b) and (c); but it may break down in (d).

8. Appendix Give a proof of some useful mathematical identities (Chang, 2008; Xie, 2009a, 2009b)

\[ \int_V \left[ \Phi \nabla \cdot \left( \vec{\lambda} \cdot \nabla \Psi \right) - \Psi \nabla \cdot \left( \vec{\lambda} \cdot \nabla \Phi \right) \right] d^3\vec{r} = \int_S \left( \Phi \vec{\lambda} \cdot \nabla \Psi - \Psi \vec{\lambda} \cdot \nabla \Phi \right) da, \]  
\( \text{(A1)} \)

under the condition $\lambda_j = \lambda_i$.

To prove Eq. (A1), we will first prove the following identity:

\[ \nabla \cdot \left[ \vec{\lambda} \cdot \left( \Phi \nabla \Psi - \Psi \nabla \Phi \right) \right] = \Phi \nabla \cdot \left( \vec{\lambda} \cdot \nabla \Psi \right) - \Psi \nabla \cdot \left( \vec{\lambda} \cdot \nabla \Phi \right), \]  
\( \text{(A2)} \)

under the condition $\lambda_j = \lambda_i$. Using the Einstein notation to express Eq. (A2), we have for the LHS:

\[ \nabla \cdot \left[ \vec{\lambda} \cdot \left( \Phi \nabla \Psi - \Psi \nabla \Phi \right) \right] = \Phi \partial_{\lambda j} \partial_i \Psi - \Psi \partial_{\lambda j} \partial_i \Phi 
+ \lambda_j \left( \partial_\lambda \Phi \right) \left( \partial_i \Psi \right) - \left( \partial_\lambda \Psi \right) \left( \partial_i \Phi \right), \]  
\( \text{(A3)} \)

and for the RHS of Eq. (A2):

\[ \Phi \nabla \cdot \left( \vec{\lambda} \cdot \nabla \Psi \right) - \Psi \nabla \cdot \left( \vec{\lambda} \cdot \nabla \Phi \right) = \Phi \partial_{\lambda j} \partial_i \Psi - \Psi \partial_{\lambda j} \partial_i \Phi. \]  
\( \text{(A4)} \)

Thus Eqs. (A3) and (A4) are equal under the condition $\lambda_j = \lambda_i$ and hence we prove Eq. (A1).

\[ \int d^3\vec{r} \int d^3\vec{r}_1 \left\{ \Phi(\vec{r}) \nabla \cdot \left[ \vec{\lambda}(\vec{r}, \vec{r}_1) \cdot \nabla \Psi(\vec{r}_1) \right] - \Psi(\vec{r}) \nabla \cdot \left[ \vec{\lambda}(\vec{r}, \vec{r}_1) \cdot \nabla \Phi(\vec{r}_1) \right] \right\} = \int_S da \int d^3\vec{r}_1 \left\{ \hat{\nu}_1 \cdot \left[ \Phi(\vec{r}) \vec{\lambda}(\vec{r}, \vec{r}_1) \cdot \nabla_1 \Psi(\vec{r}_1) - \Psi(\vec{r}) \vec{\lambda}(\vec{r}, \vec{r}_1) \cdot \nabla_1 \Phi(\vec{r}_1) \right] \right\}, \]  
\( \text{(A5)} \)

under the condition $\lambda_j(\vec{r}, \vec{r}') = \lambda_i(\vec{r}', \vec{r})$.

First we prove the following identity:

\[ \int d^3\vec{r} \int d^3\vec{r}_1 \left\{ \Phi(\vec{r}) \nabla \cdot \left[ \vec{\lambda}(\vec{r}, \vec{r}_1) \cdot \nabla \Psi(\vec{r}_1) \right] - \Psi(\vec{r}) \nabla \cdot \left[ \vec{\lambda}(\vec{r}, \vec{r}_1) \cdot \nabla \Phi(\vec{r}_1) \right] \right\} = \int d^3\vec{r} \int d^3\vec{r}_1 \left\{ \nabla \cdot \left[ \Phi(\vec{r}) \vec{\lambda}(\vec{r}, \vec{r}_1) \cdot \nabla \Psi(\vec{r}_1) - \Psi(\vec{r}) \vec{\lambda}(\vec{r}, \vec{r}_1) \cdot \nabla \Phi(\vec{r}_1) \right] \right\}, \]  
\( \text{(A6)} \)
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under the condition \( \lambda_j(\vec{r}, \vec{r}') = \lambda_j(\vec{r}', \vec{r}) \). Again we express the left side as:

\[
\int d^3\vec{r} \int d^3\vec{r}_i \left\{ \Phi(\vec{r}) \nabla \left[ \tilde{A}(\vec{r}, \vec{r}_i) \cdot \nabla \Psi(\vec{r}_i) \right] - \Psi(\vec{r}) \nabla \left[ \tilde{A}(\vec{r}, \vec{r}_i) \cdot \nabla \Phi(\vec{r}_i) \right] \right\} 
\]

and the right side as:

\[
\int d^3\vec{r} \int d^3\vec{r}_i \left\{ \Phi(\vec{r}) \partial_j \lambda_j(\vec{r}, \vec{r}_i) \partial_i \Psi(\vec{r}_i) - \Psi(\vec{r}) \partial_j \lambda_j(\vec{r}, \vec{r}_i) \partial_i \Phi(\vec{r}_i) \right\} 
\]

Thus Eqs. (A7) and (A8) are equal under the condition \( \lambda_j(\vec{r}, \vec{r}') = \lambda_j(\vec{r}', \vec{r}) \) and hence Eq. (A6) is established. We can again use the divergence theorem to establish Eq. (A5).

\[
\int \left\{ \nabla \times \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{B}} \right\} \cdot \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{A}} - \left[ \tilde{\vec{B}} \cdot \nabla \times \tilde{\vec{B}} \right] \cdot \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{A}} 
\]

under the condition \( \lambda_j = \lambda_j \).

Let us first establish the following simpler vector identity:

\[
\nabla \cdot \left[ \tilde{\vec{B}} \times \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{A}} - \tilde{\vec{A}} \times \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{B}} \right] = \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{B}} - \tilde{\vec{B}} \cdot \nabla \times \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{A}} 
\]

under the condition \( \lambda_j = \lambda_j \). In explicit Einstein’s summation convention, we have for the LHS of Eq. (A10):

\[
\nabla \cdot \left[ \tilde{\vec{B}} \times \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{A}} - \tilde{\vec{A}} \times \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{B}} \right] = \epsilon_{ijk} \epsilon_{lmn} \left( \tilde{B}_i \partial_j \lambda_{lm} \partial_m A_n - A_i \partial_j \lambda_{lm} \partial_m \tilde{B}_n \right) + \epsilon_{ijk} \epsilon_{lmn} \lambda_{ij} \left[ (\partial_i B_j)(\partial_m A_n) - (\partial_i A_j)(\partial_m B_n) \right]
\]

and the RHS of Eq. (A10):

\[
\tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{B}} = \epsilon_{ijk} \epsilon_{lmn} \left( A_i \partial_j \lambda_{lm} \partial_m B_n - B_i \partial_j \lambda_{lm} \partial_m A_n \right)
\]

Thus Eqs. (A11) and (A12) are equal under the condition \( \lambda_j = \lambda_j \) and hence Eq. (A10) is established.

From Eq. (A10) application of the divergence theorem leads to:

\[
\int \left[ \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{B}} - (\nabla \times \tilde{\vec{A}} \cdot \nabla \times \tilde{\vec{B}}) \cdot \tilde{\vec{B}} \right] d^3\vec{r} 
\]

and thus we get the following form by generalizing \( \tilde{\vec{B}} \) to a second rank tensor \( \tilde{\vec{B}} \):
\[
\begin{align*}
\int \left[ \left( \nabla \times \hat{\lambda} \cdot \nabla \times \hat{E} \right) \cdot \hat{A} - \left[ \hat{B} \right]^T \cdot \nabla \times \hat{A} \cdot \nabla \times \hat{A} \right] d^3 \vec{r} \\
= \oint \left[ \hat{n} \times \hat{B} \right] \cdot \left( \hat{A} \cdot \nabla \times \hat{A} \right) - \left[ \hat{A} \cdot \nabla \times \hat{B} \right]^T \cdot \hat{n} \times \hat{A} \right] da.
\end{align*}
\] (A14)

Hence we repeat this step for \( \hat{A} \) leads to the result in Eq. (A9).

\[
\begin{align*}
\int d^3 \vec{r} \int d^3 \vec{r}_1 \left[ \nabla \times \hat{A} (\vec{r}, \vec{r}_1) \cdot \nabla \times \hat{B} (\vec{r}_1) \right] \\
- \left[ \hat{B} (\vec{r}) \right]^T \cdot \nabla \times \hat{A} (\vec{r}, \vec{r}_1) \cdot \nabla \times \hat{A} (\vec{r}_1) \\
= \oint \left[ \hat{n} \times \hat{B} (\vec{r}) \right] \cdot \left( \hat{A} (\vec{r}, \vec{r}_1) \cdot \nabla \times \hat{A} (\vec{r}_1) \right) - \left[ \hat{A} (\vec{r}, \vec{r}_1) \cdot \nabla \times \hat{B} (\vec{r}_1) \right]^T \cdot \hat{n} \times \hat{A} (\vec{r}) \right] da.
\end{align*}
\] (A15)

under the condition \( \lambda_{ij} (\vec{r}, \vec{r}') = \lambda_{ji} (\vec{r}', \vec{r}) \).

Let us first establish the following identity:

\[
\begin{align*}
\int d^3 \vec{r} \int d^3 \vec{r}_1 \nabla \left[ \hat{B} (\vec{r}) \cdot \nabla \times \hat{A} (\vec{r}, \vec{r}_1) \right] \\
- \hat{A} (\vec{r}) \cdot \nabla \times \hat{A} (\vec{r}_1) \cdot \nabla \times \hat{B} (\vec{r}_1) \\
= \int d^3 \vec{r} \int d^3 \vec{r}_1 \left[ \hat{A} (\vec{r}, \vec{r}_1) \cdot \nabla \times \hat{A} (\vec{r}_1) \right. \\
- \hat{A} (\vec{r}_1) \cdot \nabla \times \hat{A} (\vec{r}, \vec{r}_1) \cdot \nabla \times \hat{B} (\vec{r}) \right].
\end{align*}
\] (A16)

Again we express the left side as:

\[
\begin{align*}
\int d^3 \vec{r} \int d^3 \vec{r}_1 \nabla \left[ \hat{B} (\vec{r}) \cdot \nabla \times \hat{A} (\vec{r}, \vec{r}_1) \right] \\
- \hat{A} (\vec{r}) \cdot \nabla \times \hat{A} (\vec{r}_1) \cdot \nabla \times \hat{B} (\vec{r}_1) \\
= \epsilon_{ij} \epsilon_{lmn} \int d^3 \vec{r} \int d^3 \vec{r}_1 \left[ B_i (\vec{r}) \partial_j A_{lij} (\vec{r}, \vec{r}_1) \partial_m A_k (\vec{r}_1) - A_{ijk} (\vec{r}) \partial_l A_{ijl} (\vec{r}, \vec{r}_1) \partial_m B_k (\vec{r}_1) \right],
\end{align*}
\] (A17)

and the right side as:

\[
\begin{align*}
\int d^3 \vec{r} \int d^3 \vec{r}_1 \left[ \hat{A} (\vec{r}, \vec{r}_1) \cdot \nabla \times \hat{A} (\vec{r}_1) \right. \\
- \hat{A} (\vec{r}_1) \cdot \nabla \times \hat{A} (\vec{r}, \vec{r}_1) \cdot \nabla \times \hat{B} (\vec{r}) \right] \\
= \epsilon_{ij} \epsilon_{lmn} \int d^3 \vec{r} \int d^3 \vec{r}_1 \left[ A_{ijk} (\vec{r}) \partial_l A_{ijl} (\vec{r}, \vec{r}_1) \partial_m B_k (\vec{r}_1) - B_i (\vec{r}) \partial_j A_{lij} (\vec{r}, \vec{r}_1) \partial_m A_k (\vec{r}_1) \right].
\end{align*}
\] (A18)

Hence Eq. (A17) is equal to Eq. (A18) by imposing \( \lambda_{ij} (\vec{r}, \vec{r}') = \lambda_{ji} (\vec{r}', \vec{r}) \) and the result in Eq. (A15) can again be obtained by the same method as that in proving Eq. (A9).

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10. References


This comprehensive volume thoroughly covers wave propagation behaviors and computational techniques for electromagnetic waves in different complex media. The chapter authors describe powerful and sophisticated analytic and numerical methods to solve their specific electromagnetic problems for complex media and geometries as well. This book will be of interest to electromagnetics and microwave engineers, physicists and scientists.

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