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Reciprocity in nonlocal nano-optics

P T Leung[1,2,3] **and R Chang**[2]

[1] Department of Physics, Portland State University, PO Box 751, Portland,
OR 97207-0751, USA
[2] Institute of Optoelectronic Sciences, National Taiwan Ocean University, Keelung, Taiwan

E-mail: hopl@pdx.edu (P T Leung)

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Abstract

The principle of optical reciprocity is examined in the long wavelength limit in the presence of an isotropic nonlocal medium, by generalizing a previous result established in the literature on the symmetry of the Green's function. Our focus here is in its possible application to plasmonics, i.e. optics with metallic nanostructures. It is shown that reciprocity will still hold as long as the dielectric function is symmetric with $\varepsilon(\vec{r}, \vec{r}') = \varepsilon(\vec{r}', \vec{r})$. Hence, under this symmetric condition, anisotropic dielectric response is necessary for the breakdown of reciprocity in the long wavelength limit. We further show in the appendix that this breakdown only occurs for an asymmetric dielectric tensor, in both the local and nonlocal response cases. An example is given to illustrate this symmetry in the problem of an emitting dipole interacting with a metallic nanoparticle whose response is described by certain dielectric functions which are both frequency and wavevector dependent.

Keywords: reciprocity, nonlocal dielectric response, nano-optics

1. Introduction

The recently emerged field of plasmonics represents an exciting development of metal optics, and promises to be one significant nanotechnology which will span a large spectrum of applications from biosensing and cancer therapy using metallic nanoparticles, to the bridging of photonics and electronics using nanometallic waveguides [1]. Since most of these were achieved via the collective excitation of the free electrons at the boundary of a metallic nanostructure (known as surface plasmon excitation), the detailed theoretical understanding of the optical interaction with metallic nanostructures is crucial in order that various plasmonics applications can be implemented in a controlled fashion. While in recent years many aspects of this new field in nano-optics have been studied intensively by various researchers [2], it is our interest in this work to focus on a fundamental principle in optics—the principle of reciprocity—as applied to nano-optics with metals.

Optical reciprocity refers to the reversal symmetry between the source and the observer of the field during propagation of an electromagnetic wave in a material medium [3]. From a mathematical point of view, this symmetry arises ultimately from the symmetry of the Green's function (or dyadic) of the wave equation with respect to the observer and source locations[4]. Although the study of this symmetry has a history of over a century [3], renewed interest in its foundation and new applications of the principle have never diminished in the research into optics and spectroscopy [4]. While this symmetry is trivial when the source and observer are both located in an infinite homogeneous medium, it is rather nontrivial in the presence of boundaries created by different media of finite extents. Thus it has been constantly an interesting problem to study the possibility for the breakdown of this symmetry in the presence of a material medium [3]. Here our particular interest is in the validity of reciprocity in the presence of nonlocal optical response of the medium, since it is known that such a response is rather significant with metallic nanostructures due to the large area-to-volume ratio of these systems[5].

Although it has been established in the previous literature that this principle in general breaks down for optical phenomena involving a nonlocal medium [3, 6], most of

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[3] Author to whom any correspondence should be addressed.

[4] It is easy to see that the more popular statement of the reciprocity theorem [3] (via the 'Lorentz lemma' $\int \vec{j}_1 \cdot \vec{E}_2 \, \mathrm{d}\vec{r} = \int \vec{j}_2 \cdot \vec{E}_1 \, \mathrm{d}\vec{r}$) simply implies the symmetry for the Green dyadic $\overset{\leftrightarrow}{G}(\vec{r}, \vec{r}') = \overset{\leftrightarrow}{G}(\vec{r}', \vec{r})$ by recalling that $\vec{E}(\vec{r}) = \int \overset{\leftrightarrow}{G}(\vec{r}, \vec{r}')\vec{j}(\vec{r}') \, \mathrm{d}\vec{r}'$.

[5] As an illustration on how the nonlocal response can affect the interaction between a fluorescing molecule and a metallic nanoparticle, see, e.g. [5].

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these works revealed such non-reciprocity via consideration of various optical effects in anisotropic media (e.g. crystals), with the nonlocality arising, for example, from the response depending on the gradient of the electric field. In this work, however, we shall provide a general proof of the possible validity of reciprocity in the presence of an *isotropic* nonlocal medium, by generalizing previous works in the literature [7, 8] on the symmetry of the Green's function. This is motivated by the fact that anisotropy is rather insignificant for the description of the dielectric response from the free electrons in metals. We shall see that such validity breaks down only for materials with asymmetric dielectric response function $\varepsilon(\vec r,\vec r') \neq \varepsilon(\vec r',\vec r)$, which is unlikely for macroscopic isotropic materials.

To simplify the mathematics, we shall limit ourselves to the long-wavelength approximation so that the electrostatic approach in [7, 8] can be followed. This is justified since nonlocal effects are important only for metallic structures of dimensions <10 nm [5], which is much smaller than a wavelength in the visible spectrum. Hence we shall implement a 'quasi-static' model with a dielectric function dependent on both the wavevector and frequency, which will be valid over a large range of timescales—except perhaps the shortest ones in the range of femto- to attoseconds, as shall be discussed further in our illustrative example below. In addition, we shall only consider the simple case of the Dirichlet boundary condition with the Green's function vanishing at the boundary surface. The more complicated case with the Neumann boundary condition can presumably be treated in a similar way by redefining the Green's function, as was done previously for the simple case of a perfect conducting medium [7]. Furthermore, only linear dielectric response will be considered in this work, and we shall illustrate the results by referring to the problem of a point source interacting with a metallic nanoparticle whose dielectric response is described by certain quantum mechanical models for a homogeneous electron gas.

2. Theory

Following Kim and Jackson [7], let us consider the Green's function $G(\vec r,\vec r')$ and examine the symmetry of it under the exchange of the coordinates $\vec r$ and $\vec r'$, in the presence of nonlocal dielectric response. First we note that the proofs in [7] are limited only to vacuum and (perfect) conducting boundaries, so that under this ideal situation, the symmetry $G(\vec r,\vec r') = G(\vec r',\vec r)$ can easily be proven for the Dirichlet case with the application of Green's theorem. However, in order to extend it to the case with a nonlocal dielectric, the formulation in [7] must first be generalized to the case where a realistic (and local) dielectric medium of finite conductivity may be present. In this case, the conventional Green's theorem cannot be directly applied to establish the proof as done in [7]. It turns out that such a generalization has already been published by Schwinger *et al* [8], and we provide a brief summary of their proof in the following.

As shown in [8], in the presence of a realistic medium characterized by a *local* dielectric response function $\varepsilon(\vec r)$, the Poisson equation satisfied by $G(\vec r,\vec r')$ [7] must be generalized to the following form:

$$\vec\nabla\cdot[\varepsilon(\vec r)\vec\nabla G(\vec r,\vec r')] = -4\pi\delta(\vec r-\vec r'). \tag{1}$$

Let us also consider $G(\vec r,\vec r'')$ satisfying:

$$\vec\nabla\cdot[\varepsilon(\vec r)\vec\nabla G(\vec r,\vec r'')] = -4\pi\delta(\vec r-\vec r''). \tag{2}$$

Hence from (1) and (2), we obtain:

$$\begin{aligned}
&4\pi\delta(\vec r-\vec r'')G(\vec r,\vec r') - 4\pi\delta(\vec r-\vec r')G(\vec r,\vec r'')\\
&\quad= G(\vec r,\vec r'')\vec\nabla\cdot[\varepsilon(\vec r)\vec\nabla G(\vec r,\vec r')]\\
&\qquad - G(\vec r,\vec r')\vec\nabla\cdot[\varepsilon(\vec r)\vec\nabla G(\vec r,\vec r'')]\\
&\quad= \vec\nabla\cdot\{\varepsilon(\vec r)[G(\vec r,\vec r'')\vec\nabla G(\vec r,\vec r')\\
&\qquad - G(\vec r,\vec r')\vec\nabla G(\vec r,\vec r'')]\},
\end{aligned}\tag{3}$$

where we have used the following identity for three scalar functions [8]:

$$\vec\nabla\cdot[\lambda(\phi\vec\nabla\psi-\psi\vec\nabla\phi)] = \phi\vec\nabla\cdot(\lambda\vec\nabla\psi) - \psi\vec\nabla\cdot(\lambda\vec\nabla\phi). \tag{4}$$

Thus, integrating (3) over the $\vec r$ coordinates and applying the divergence theorem, we obtain:

$$\begin{aligned}
&4\pi\left[G\left(\vec r'',\vec r'\right) - G\left(\vec r',\vec r''\right)\right] = \oint_S \varepsilon\left(\vec r\right)\left[G\left(\vec r,\vec r''\right)\vec\nabla G\left(\vec r,\vec r'\right)\right.\\
&\qquad \left.- G\left(\vec r,\vec r'\right)\vec\nabla G\left(\vec r,\vec r''\right)\right]\cdot\hat n\,\mathrm{d}a = 0,
\end{aligned}\tag{5}$$

since the Green's function vanishes on the boundary under the Dirichlet condition. Hence (5) implies the reciprocity relation $G(\vec r',\vec r'') = G(\vec r'',\vec r')$ under this condition and in the presence of a medium with local dielectric response $\varepsilon(\vec r)$ [8].

To extend the above proof to the case with the presence of nonlocal dielectric response, we first generalize (1) and (2) to the following form:

$$\vec\nabla\cdot\left[\int\varepsilon(\vec r,\vec r_1)\vec\nabla_1 G(\vec r_1,\vec r')\mathrm{d}\vec r_1\right] = -4\pi\delta(\vec r-\vec r'), \tag{6}$$

$$\vec\nabla\cdot\left[\int\varepsilon(\vec r,\vec r_2)\vec\nabla_2 G(\vec r_2,\vec r'')\mathrm{d}\vec r_2\right] = -4\pi\delta(\vec r-\vec r''). \tag{7}$$

Now, repeating the same step as in (3) with the nonlocal expressions in (6) and (7) leads to:

$$\begin{aligned}
&4\pi\delta(\vec r-\vec r'')G(\vec r,\vec r') - 4\pi\delta(\vec r-\vec r')G(\vec r,\vec r'')\\
&\quad= \int\mathrm{d}\vec r_1\{G(\vec r,\vec r'')\vec\nabla\cdot[\varepsilon(\vec r,\vec r_1)\vec\nabla_1 G(\vec r_1,\vec r')]\\
&\qquad - G(\vec r,\vec r')\vec\nabla\cdot[\varepsilon(\vec r,\vec r_1)\vec\nabla_1 G(\vec r_1,\vec r'')]\},
\end{aligned}\tag{8}$$

where we can change the dummy coordinate $\vec r_2$ in (7) to $\vec r_1$ so that the two terms can be added under the same integral sign. From this, an integration of (8) over $\vec r$ will lead to the following result:

$$\begin{aligned}
&4\pi G(\vec r'',\vec r') - 4\pi G(\vec r',\vec r'')\\
&\quad= \int\mathrm{d}\vec r_1\int\mathrm{d}\vec r\{G(\vec r,\vec r'')\vec\nabla\cdot[\varepsilon(\vec r,\vec r_1)\vec\nabla_1 G(\vec r_1,\vec r')]\\
&\qquad - G(\vec r,\vec r')\vec\nabla\cdot[\varepsilon(\vec r,\vec r_1)\vec\nabla_1 G(\vec r_1,\vec r'')]\}\\
&\quad= \int\mathrm{d}\vec r_1\int\mathrm{d}\vec r\,\vec\nabla\cdot[G(\vec r,\vec r'')\varepsilon(\vec r,\vec r_1)\vec\nabla_1 G(\vec r_1,\vec r')\\
&\qquad - G(\vec r,\vec r')\varepsilon(\vec r,\vec r_1)\vec\nabla_1 G(\vec r_1,\vec r'')]\\
&\qquad - \int\mathrm{d}\vec r_1\int\mathrm{d}\vec r\,\varepsilon(\vec r,\vec r_1)[\vec\nabla G(\vec r,\vec r'')\cdot\vec\nabla_1 G(\vec r_1,\vec r')\\
&\qquad - \vec\nabla G(\vec r,\vec r')\cdot\vec\nabla_1 G(\vec r_1,\vec r'')].
\end{aligned}\tag{9}$$

Just like the previous local case, the first term of the last result in (9) vanishes via the conversion of the integral over $\vec{r}$ to a surface term and use of the boundary condition of the Green's function. As for the second term, we note that the term within the brackets [ ] is antisymmetric in the coordinates $\vec{r}$ and $\vec{r}_1$. Hence, if the dielectric function is symmetric in these coordinates, i.e. $\varepsilon(\vec{r},\vec{r}_1) = \varepsilon(\vec{r}_1,\vec{r})$, the double integral in the second term of (9) will also vanish and the Green's function will be symmetric, implying reciprocity holds even in the nonlocal case. This will be true, for example, for most of the well-known nonlocal quantum mechanical models for a homogeneous electron gas, such as the Linhard–Mermin function in which $\varepsilon(\vec{r},\vec{r}_1) = \varepsilon(|\vec{r}-\vec{r}_1|)$ [9]. Thus, even if it is known that this symmetry in $\varepsilon(\vec{r},\vec{r}_1)$ may not be guaranteed at the microscopic scale such as in the dielectric response on an intramolecular length scale [10], it seems that for most isotropic macroscopic and mesoscopic systems, such symmetry will be valid and hence reciprocity will hold even in the presence of an isotropic nonlocal medium.

Although anisotropic response is not significant for the free electrons in a metal, one can show that when $\varepsilon \to \overleftrightarrow{\varepsilon}(=\varepsilon_{ij})$, reciprocity will still hold in the case of both local and nonlocal response provided that the dielectric tensor is symmetric, i.e. $\varepsilon_{ij} = \varepsilon_{ji}$ (see appendix). Hence we can now clearly understand the previous results for the breakdown of reciprocity in the presence of nonlocal anisotropic response within the linear response theory [6]. The origin of such a breakdown is really from the *asymmetry* of the dielectric tensor which describes the nonlocal response from the dependence on field gradient of the displacement vector. As an example, one notes that this tensor becomes *anti*symmetric in the case when time-reversal symmetry holds, as explained in [6]. As a further example, reciprocity will also break in noncentrosymmetric media in which the dielectric function will not be symmetric even in its space arguments as explained in [3].

3. An illustrative example

To illustrate explicitly reciprocity in the presence an isotropic nonlocal metal structure, we consider the problem of a fluorescing molecule (treated as a harmonic point dipole) interacting with a metallic nanosphere. In a phenomenological model, the problem can be completely described with the knowledge of the Green's function of the electrodynamics involved[6] (see footnote 5). It is well known that the nonlocal effects are significant for metallic particles with sizes below ~10 nm. Assuming both the size (a) and the distance ($r'-a$) between the dipole and the sphere are small compared to a typical optical wavelength, electrostatics can be applied for simplicity. In addition, the lifetimes (~ ns) of the molecules are in general much longer than the characteristic time (~ inverse of the plasmon frequency) of the free electrons. Hence the spatial-temporal dispersions of the metal can be treated by a nonlocal dielectric function of the form $\varepsilon(\vec{k},\omega)$ in the Fourier space. Under these conditions, the nonlocal response of the sphere has been derived previously by Fuchs and Claro [12] in the so-called SCIB (semi-classical infinite barrier) approximation. According to [12], the nonlocal ℓth-multipole polarizability of the sphere can be obtained in the following form:

$$\alpha_\ell^{\mathrm{NL}} = \frac{\xi_\ell - 1}{\xi_\ell + (\ell+1)/\ell} a^{2\ell+1}, \tag{10}$$

where the 'effective dielectric function' is given by:

$$\xi_\ell(\omega) = \left[\frac{2}{\pi}(2\ell+1)a\int_0^\infty \frac{j_\ell^2(ka)}{\varepsilon(k,\omega)}\mathrm{d}k\right]^{-1}, \tag{11}$$

with j_ℓ the spherical Bessel function, and we have assumed an isotropic response $\varepsilon(k,\omega)$. As an example, with the simple hydrodynamic model for $\varepsilon(k,\omega)$, equation (11) can be integrated to obtain [12]:

$$\xi_\ell(\omega) = \left[\frac{1}{\varepsilon_{\mathrm{D}}} + (2\ell+1)\left(\frac{a\omega_{\mathrm{P}}}{\beta u}\right)^2 I_{\ell+1/2}(\omega)K_{\ell+1/2}(\omega)\right]^{-1}, \tag{12}$$

where ω_{P} is the bulk plasmon frequency, I, K are the modified Bessel functions, β and u are parameters as defined in [12], and the Drude dielectric function is given by: $\varepsilon_{\mathrm{D}} = 1 - \frac{\omega_{\mathrm{P}}^2}{\omega^2 + \mathrm{i}\omega\Gamma}$. With the application of the polarizability of the sphere in equation (10), the potential at a point $\vec{r} = (r,\theta,\varphi)$ due to a unit source at $\vec{r} = (r',\theta',\varphi')$ outside the sphere can be obtained, which leads to the following expression for the Green's function of the problem:

$$G(\vec{r},\vec{r}') = 4\pi\sum_{\ell,m}\left(\frac{1}{2\ell+1}\right)\left[\frac{r_<^\ell}{r_>^{\ell+1}} - \frac{\alpha_\ell^{\mathrm{NL}}}{(rr')^{\ell+1}}\right] \times Y_{\ell m}^*(\theta',\varphi')\,Y_{\ell m}(\theta,\varphi), \tag{13}$$

where $(r_<, r_>)$ denote the smaller or greater of (r, r'). Note that the derivation of the result in equation (13) is in complete analogy to the case of a charge outside a perfect conducting sphere which can be found in [13]. With the nonlocal polarizability given in (10) and (11), it is obvious from (13) that we indeed have reciprocity symmetry with $G(\vec{r},\vec{r}') = G(\vec{r}',\vec{r})$ in this case of an isotropic nonlocal metal sphere. This is due to the fact that the wavevector is integrated over in (11) which yields as a consequence an 'effective local polarizability' through (10). With this, the reciprocal symmetry of $G(\vec{r},\vec{r}')$ in (13) is just identical to that in the usual situation of electrostatics with perfect conductors [13], as can easily be checked by switching between the coordinates (r,θ,φ) and (r',θ',φ') in (13). We would like to add that in one of our previous works using this model to study reciprocity symmetry between a metallic nanosphere and a nanocavity [14], the breakdown of reciprocity was wrongly concluded due to a mistake in the transformation of (12) from inside to outside of the spherical boundary[7]. No such breakdown should have occurred for such a problem with isotropic nonlocal response as established in our present work.

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[6] For recent investigations of these effects on the emission from molecules in the vicinity of a metallic nanostructure, see, e.g. [11].

[7] Note that the transformation $\ell \to -(\ell+1)$ used in this previous paper should not have been applied to the result in our present equation (12), in going from the sphere to the cavity case. Equation (12) should just remain invariant in both the sphere and cavity case.

4. Conclusion

In conclusion, we have demonstrated, by generalizing the previous work on the symmetry of the Green's function [7, 8] and by going beyond the local response restriction, that optical reciprocity is in general valid in the presence of an isotropic nonlocal dielectric medium unless the response function is asymmetric in its coordinates, which happens only on a molecular scale [10]. We have arrived at this conclusion within the long wavelength limit in which electrostatics can be applied, and have illustrated the symmetry explicitly in the problem of a point source interacting with a nonlocal metallic nanoparticle. The validity of this symmetry principle will ensure that the recent applications [4] of it to various surface-enhanced spectroscopic analyses will remain valid, even when metallic nanostructures are considered, in which case nonlocal optical effects can be significant [5, 11]. In addition, we have also considered only the case of Dirichlet boundary conditions, and it will be of interest to verify that the same proof can be extended to the case of Neumann conditions in a similar way, by redefining a new Green's function to absorb the surface term following the approach of [7]. In principle, this reciprocity symmetry can also be demonstrated in the exact nonlocal electrodynamic theory by considering the symmetry of the Green dyadic [15]. However, the algebra involved will be more complicated and we believe that our present quasi-static approach is relatively simple and sufficient to illustrate the validity and limitation of optical reciprocity in the presence of a nonlocal response from an isotropic medium of small dimensions.

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Appendix

Here we show how the effect of anisotropy will affect the results in equation (3) for the local case and in equation (9) for the nonlocal case, respectively. It is not difficult to see that by having $\varepsilon \rightarrow \overset{\leftrightarrow}{\varepsilon}$ in (1) and (2), (3) will remain valid with the corresponding $\varepsilon \rightarrow \overset{\leftrightarrow}{\varepsilon}$ provided that (4) is still valid with $\lambda \rightarrow \overset{\leftrightarrow}{\lambda}$. To check the validity of (4) when λ becomes a tensor, one finds that both sides in (4) will equal each other if

$$\overset{\leftrightarrow}{\lambda}\cdot\vec\nabla\phi\vec\nabla\psi = \overset{\leftrightarrow}{\lambda}\cdot\vec\nabla\psi\vec\nabla\phi \tag{A.1}$$

which implies

$$\sum_{ij}\lambda_{ij}\partial_j\phi\partial_i\psi = \sum_{ij}\lambda_{ij}\partial_j\psi\partial_i\phi \tag{A.2}$$

and is true if $\lambda_{ij} = \lambda_{ji}$. Hence we conclude that (5) and hence reciprocity will be valid in the local case as long as the dielectric tensor is symmetric with $\varepsilon_{ij} = \varepsilon_{ji}$—a result well established in the literature [16].

For the nonlocal case with $\varepsilon \rightarrow \overset{\leftrightarrow}{\varepsilon}$, one can easily show that everything in equation (9) will go through just by this replacement of the dielectric function, except that the integral in the last row will change to the following form:

$$-\int \mathrm{d}\vec r_1 \int \mathrm{d}\vec r \Big\{\big[\overset{\leftrightarrow}{\varepsilon}(\vec r,\vec r_1)\cdot\vec\nabla_1 G(\vec r_1,\vec r')\big]\cdot\vec\nabla G(\vec r,\vec r'') - \Big[\overset{\leftrightarrow}{\varepsilon}(\vec r,\vec r_1)\cdot\vec\nabla_1 G(\vec r_1,\vec r'')\Big]\cdot\vec\nabla G(\vec r,\vec r')\Big\}. \tag{A.3}$$

Hence the integrand in (A.3) can be explicitly expressed as:

$$\sum_{ij}\big[\varepsilon_{ij}(\vec r,\vec r_1)\,\partial_{1j}G(\vec r_1,\vec r')\big]\,\partial_i G(\vec r,\vec r'') - \sum_{ij}\big[\varepsilon_{ij}(\vec r,\vec r_1)\,\partial_{1j}G(\vec r_1,\vec r'')\big]\,\partial_i G(\vec r,\vec r'), \tag{A.4}$$

and it will be antisymmetric in $\vec r \leftrightarrow \vec r_1$ if $\varepsilon_{ij}(\vec r,\vec r_1) = \varepsilon_{ji}(\vec r_1,\vec r)$. This will again lead to validity of reciprocity from (9) under the above symmetric condition for the dielectric function. The breakdown of this condition leading to the failure of reciprocity was discussed in [3].

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