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General proof of optical reciprocity for nonlocal electrodynamics

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Abstract
Recent studies have established certain specific results concerning the optical reciprocity of a source and an observer in the presence of a dielectric medium with a nonlocal response (i.e. spatial dispersion). These include the case of a linear dielectric response with dependence on the electric field gradient, as well as the case of a general anisotropic nonlocal medium in the long wavelength quasistatic limit. Here we present a more rigorous study of this problem by extending the previous results to the most general case of an anisotropic nonlocal medium in the context of exact electrodynamics with reference to the symmetry of the Green dyadic. It is established that for nonmagnetic materials within linear optics, reciprocity will remain valid as long as the dielectric tensor satisfies the condition $\varepsilon_{ij}(\vec{r},\vec{r}_1) = \varepsilon_{ji}(\vec{r}_1,\vec{r})$. Possible applications of the results are briefly discussed.

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Introduction

In conventional classical optics, the reciprocity theorem is a powerful result which finds applications in many problems in optics and spectroscopy [1]. These include, for example, establishing relations between far fields and near fields from different sources [2] as well as spectroscopic analysis of surface-enhanced Raman scattering at metallic structures [3]. The theorem expresses the reversal symmetry between the source and the observer of the field during propagation of an electromagnetic wave in a material medium, and can have various mathematical forms expressed in terms of the sources and the fields [1]. Here we focus on one of its popular forms (the 'Lorentz lemma') [4]:

$$\int \vec{J}_1 \cdot \vec{E}_2 \, d^3\vec{r} = \int \vec{J}_2 \cdot \vec{E}_1 \, d^3\vec{r}, \tag{1}$$

which simply implies the symmetry for the Green dyadic $[\overleftrightarrow{G}_e(\vec{r}, \vec{r}\,')]^T = \overleftrightarrow{G}_e(\vec{r}\,', \vec{r})$ by recalling that $\vec{E}(\vec{r}) = \mathrm{i}\omega\mu_0 \int \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,') \cdot \vec{J}(\vec{r}\,')\, \mathrm{d}^3\vec{r}\,'$. This is obvious for the case of free-space Green dyadic, but the connection can also be shown for problems involving finite spatial regions. While equation (1) can be derived rigorously from Maxwell's electrodynamics with the limitation to the presence of materials with only a local dielectric response (i.e. only temporal but no spatial dispersion in the dielectric function), recent studies have raised the question of its validity in nonlocal optics [1]. In particular, it has been pointed out that a general proof of reciprocity has not yet been established in the literature for the nonlocal case [5]. Moreover, non-reciprocity has been revealed via consideration of certain 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 [6]. Nevertheless, we note that the non-reciprocity in this case referred to the effect on the polarization plane rotation rather than just the transmission of light between the source and the observer.

In a recent work, however, we have studied this problem in a more general approach without implementing any specific models for the dielectric function (except for linearity), and have concluded that while reciprocity still holds in the presence of an isotropic nonlocal medium, it breaks down only in the presence of asymmetric anisotropic materials with $\varepsilon_{ij} \neq \varepsilon_{ji}$ [7]. Moreover, such breakdown of reciprocity occurs both in the local and nonlocal response cases. Hence, nonlocality is not the main origin of the breakdown of reciprocity, but the asymmetric anisotropic response is the main course of such breakdown. This is somewhat different from what was studied in [6], where nonlocality through the field-gradient-dependent response is required to break the reciprocity symmetry for the rotation of the polarization plane of the transmitted wave.

However, our previous proof of the above results is restricted to the 'long wavelength approximation' in which electrostatics has been applied [7]. In problems with very high frequency sources (e.g. scattering between x-rays and a nanoparticle [8]) in which electrostatics breaks down and nonlocal effects can become even more significant due to the large value of the wave vector, the previous formulation [7] becomes inadequate. Hence, it will be desirable to provide a more rigorous proof of this 'nonlocal reciprocity' symmetry by implementing exact electrodynamics, as is done in the conventional establishment of the result in equation (1) for 'local optics'. In the following, we accomplish this by examining the symmetry of the Green dyadic in electrodynamics in the presence of a general anisotropic nonlocal dielectric medium.

Theory

We shall establish our results in three steps and consider both Dirichlet and Neumann boundary conditions for the Green dyadic. Only the linear response and nonmagnetic materials are studied in this work.

Case (i): isotropic local response

This is the case well established in the literature and we briefly review it here following Tai's text [9] to set the notations. Thus for the local response $\vec{D}(\vec{r}) = \varepsilon_0\varepsilon(\vec{r})\vec{E}(\vec{r})$, one can derive the following vector wave equation for fields with harmonic time dependence:

$$\nabla \times \nabla \times \vec{E}(\vec{r}) - \frac{\omega^2}{c^2}\varepsilon(\vec{r})\vec{E}(\vec{r}) = \mathrm{i}\omega\mu_0\vec{J}(\vec{r}), \tag{2}$$

which implies the following equation for the Green dyadic:

$$\nabla \times \nabla \times \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,') = \frac{\omega^2}{c^2}\,\varepsilon(\vec{r})\overleftrightarrow{G}_e(\vec{r}, \vec{r}\,') + \overleftrightarrow{I}\delta(\vec{r} - \vec{r}\,'). \tag{3}$$

Using the dyadic–dyadic Green's theorem [9],

$$\int_V \{[\nabla \times \nabla \times \overleftrightarrow{Q}]^T \cdot \overleftrightarrow{P} - [\overleftrightarrow{Q}]^T \cdot \nabla \times \nabla \times \overleftrightarrow{P}\}\,\mathrm{d}^3\vec{r} = \oint_S \{[\hat{n} \times \overleftrightarrow{Q}]^T \cdot (\nabla \times \overleftrightarrow{P}) - [\nabla \times \overleftrightarrow{Q}]^T \cdot (\hat{n} \times \overleftrightarrow{P})\}\,\mathrm{d}a, \tag{4}$$

we obtain by setting $\overleftrightarrow{P} = \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,')$ and $\overleftrightarrow{Q} = \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,'')$

$$\int_V \{[\nabla \times \nabla \times \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,'')]^T \cdot \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,') - [\overleftrightarrow{G}_e(\vec{r}, \vec{r}\,'')]^T \cdot \nabla \times \nabla \times \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,')\}\,\mathrm{d}^3\vec{r} = \oint_S \{[\hat{n} \times \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,'')]^T \cdot (\nabla \times \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,')) - [\nabla \times \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,'')]^T \cdot (\hat{n} \times \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,'))\}\,\mathrm{d}a. \tag{5}$$

By imposing on S either the dyadic Dirichlet condition [9]

$$\hat{n} \times \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,') = 0 \tag{6a}$$

or the dyadic Neumann condition [9]

$$\hat{n} \times \nabla \times \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,') = 0, \tag{6b}$$

the surface integral in equation (5) can be vanished (after manipulation using the dyadic identity $[\overleftrightarrow{c}]^T \cdot (\vec{a} \times \overleftrightarrow{b}) = -[\vec{a} \times \overleftrightarrow{c}]^T \cdot \overleftrightarrow{b}$ in the Neumann case). Hence we obtain from equation (5)

$$\int_V \{[\nabla \times \nabla \times \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,'')]^T \cdot \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,') - [\overleftrightarrow{G}_e(\vec{r}, \vec{r}\,'')]^T \cdot \nabla \times \nabla \times \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,')\}\,\mathrm{d}^3\vec{r} = 0. \tag{7}$$

Using equation (3), we obtain from (7)

$$\int_V \left\{\frac{\omega^2}{c^2}\varepsilon(\vec{r})[\overleftrightarrow{G}_e(\vec{r}, \vec{r}\,'')]^T + \overleftrightarrow{I}\delta(\vec{r} - \vec{r}\,'')\right\} \cdot \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,')\,\mathrm{d}^3\vec{r} - \int_V [\overleftrightarrow{G}_e(\vec{r}, \vec{r}\,'')]^T \cdot \left\{\frac{\omega^2}{c^2}\varepsilon(\vec{r})\overleftrightarrow{G}_e(\vec{r}, \vec{r}\,') + \overleftrightarrow{I}\delta(\vec{r} - \vec{r}\,')\right\}\mathrm{d}^3\vec{r} = 0, \tag{8}$$

which, after cancellation and integration, implies the following symmetry for the dyadic and hence reciprocity [4, 9]:

$$[\overleftrightarrow{G}_e(\vec{r}\,', \vec{r}\,'')]^T = \overleftrightarrow{G}_e(\vec{r}\,'', \vec{r}\,'). \tag{9}$$

Case (ii): isotropic nonlocal response

In this case, the linear response function has the following general form [4]:

$$\vec{D}(\vec{r}) = \int \varepsilon_0\,\varepsilon(\vec{r}, \vec{r}\,')\vec{E}(\vec{r}\,')\,\mathrm{d}^3\vec{r}\,', \tag{10}$$

and reciprocity for this case can essentially be established by repeating the above steps together with the following generalization of equation (3):

$$\nabla \times \nabla \times \overleftrightarrow{G}_e(\vec{r}, \vec{r}\,') = \frac{\omega^2}{c^2}\int \varepsilon(\vec{r}, \vec{r}_1)\overleftrightarrow{G}_e(\vec{r}_1, \vec{r}\,')\,\mathrm{d}^3\vec{r}_1 + \overleftrightarrow{I}\delta(\vec{r} - \vec{r}\,'). \tag{11}$$

The corresponding result in (8) then generalizes to

$$\int_V \left[\frac{\omega^2}{c^2}\int \varepsilon(\vec r,\vec r_2)\overleftrightarrow{G}_e(\vec r_2,\vec r'')\,\mathrm{d}^3\vec r_2 + \overleftrightarrow{I}\delta(\vec r-\vec r'')\right]^T\cdot\overleftrightarrow{G}_e(\vec r,\vec r')\,\mathrm{d}^3\vec r - \int_V [\overleftrightarrow{G}_e(\vec r,\vec r'')]^T\cdot\left\{\frac{\omega^2}{c^2}\int \varepsilon(\vec r,\vec r_1)\overleftrightarrow{G}_e(\vec r_1,\vec r')\,\mathrm{d}^3\vec r_1 + \overleftrightarrow{I}\delta(\vec r-\vec r')\right\}\mathrm{d}^3\vec r = 0, \tag{12}$$

which upon integration yields

$$\begin{aligned}\overleftrightarrow{G}_e(\vec r'',\vec r') - [\overleftrightarrow{G}_e(\vec r',\vec r'')]^T &= -\frac{\omega^2}{c^2}\int_V \mathrm{d}^3\vec r\int \mathrm{d}^3\vec r_2\,\varepsilon(\vec r,\vec r_2)[\overleftrightarrow{G}_e(\vec r_2,\vec r'')]^T\cdot\overleftrightarrow{G}_e(\vec r,\vec r')\\ &\quad+\frac{\omega^2}{c^2}\int_V \mathrm{d}^3\vec r\int \mathrm{d}^3\vec r_1\,\varepsilon(\vec r,\vec r_1)[\overleftrightarrow{G}_e(\vec r,\vec r'')]^T\cdot\overleftrightarrow{G}_e(\vec r_1,\vec r')\\ &= -\frac{\omega^2}{c^2}\int_V \mathrm{d}^3\vec r\int \mathrm{d}^3\vec r_1\,\varepsilon(\vec r,\vec r_1)\{[\overleftrightarrow{G}_e(\vec r_1,\vec r'')]^T\cdot\overleftrightarrow{G}_e(\vec r,\vec r')\\ &\quad-[\overleftrightarrow{G}_e(\vec r,\vec r'')]^T\cdot\overleftrightarrow{G}_e(\vec r_1,\vec r')\}.\end{aligned} \tag{13}$$

Hence if the nonlocal dielectric function satisfies the following symmetry condition [7]

$$\varepsilon(\vec r,\vec r_1) = \varepsilon(\vec r_1,\vec r), \tag{14}$$

then the integrand on the right-hand side is antisymmetric in $\vec r$ and $\vec r_1$, and the integral simply vanishes, leading again to reciprocity symmetry with

$$[\overleftrightarrow{G}_e(\vec r',\vec r'')]^T = \overleftrightarrow{G}_e(\vec r'',\vec r'). \tag{15}$$

Case (iii): anisotropic nonlocal response

Now we come to the most general case for a linear medium which is both anisotropic and nonlocal. With the dielectric function becoming a tensor[4] , equation (12) then generalizes to

$$\int_V \left[\frac{\omega^2}{c^2}\int \overleftrightarrow{\varepsilon}(\vec r,\vec r_2)\cdot\overleftrightarrow{G}_e(\vec r_2,\vec r'')\,\mathrm{d}^3\vec r_2 + \overleftrightarrow{I}\delta(\vec r-\vec r'')\right]^T\cdot\overleftrightarrow{G}_e(\vec r,\vec r')\,\mathrm{d}^3\vec r - \int_V [\overleftrightarrow{G}_e(\vec r,\vec r'')]^T\cdot\left\{\frac{\omega^2}{c^2}\int \overleftrightarrow{\varepsilon}(\vec r,\vec r_1)\cdot\overleftrightarrow{G}_e(\vec r_1,\vec r')\,\mathrm{d}^3\vec r_1 + \overleftrightarrow{I}\delta(\vec r-\vec r')\right\}\mathrm{d}^3\vec r = 0, \tag{16}$$

which upon integration yields the following result:

$$\begin{aligned}&\overleftrightarrow{G}_e(\vec r'',\vec r') - [\overleftrightarrow{G}_e(\vec r',\vec r'')]^T\\ &= -\frac{\omega^2}{c^2}\int_V \mathrm{d}^3\vec r\int \mathrm{d}^3\vec r_2[\overleftrightarrow{\varepsilon}(\vec r,\vec r_2)\cdot\overleftrightarrow{G}_e(\vec r_2,\vec r'')]^T\cdot\overleftrightarrow{G}_e(\vec r,\vec r')\\ &\quad+\frac{\omega^2}{c^2}\int_V \mathrm{d}^3\vec r\int \mathrm{d}^3\vec r_1[\overleftrightarrow{G}_e(\vec r,\vec r'')]^T\cdot\overleftrightarrow{\varepsilon}(\vec r,\vec r_1)\cdot\overleftrightarrow{G}_e(\vec r_1,\vec r').\\ &= -\frac{\omega^2}{c^2}\int_V \mathrm{d}^3\vec r\int \mathrm{d}^3\vec r_1\{[\overleftrightarrow{\varepsilon}(\vec r,\vec r_1)\cdot\overleftrightarrow{G}_e(\vec r_1,\vec r'')]^T\cdot\overleftrightarrow{G}_e(\vec r,\vec r')\\ &\quad-[\overleftrightarrow{G}_e(\vec r,\vec r'')]^T\cdot\overleftrightarrow{\varepsilon}(\vec r,\vec r_1)\cdot\overleftrightarrow{G}_e(\vec r_1,\vec r')\}\end{aligned} \tag{17}$$

[4] Note that with the dielectric function becoming a tensor in equation (10), one can show that the nonlocality due to field-gradient dependence studied in [6] can actually be deduced from a Taylor expansion of about in the limit of weak nonlocality.

Next we show that if the dielectric tensor satisfies the following symmetry:

$$\varepsilon_{ij}(\vec{r},\vec{r}_1) = \varepsilon_{ji}(\vec{r}_1,\vec{r}), \tag{18}$$

then the integrand in (17) is antisymmetric upon the exchange of coordinates $\vec{r} \leftrightarrow \vec{r}_1$ as follows. Under the transformation $\vec{r} \leftrightarrow \vec{r}_1$, the integrand in (17) changes to

$$[\overleftrightarrow{\varepsilon}(\vec{r}_1,\vec{r})\cdot\overleftrightarrow{G}_e(\vec{r},\vec{r}'')]^T\cdot\overleftrightarrow{G}_e(\vec{r}_1,\vec{r}') - [\overleftrightarrow{G}_e(\vec{r}_1,\vec{r}'')]^T\cdot\overleftrightarrow{\varepsilon}(\vec{r}_1,\vec{r})\cdot\overleftrightarrow{G}_e(\vec{r},\vec{r}'). \tag{19}$$

Using the following relations for the transpose of matrices:

$$[\overleftrightarrow{\varepsilon}(\vec{r}_1,\vec{r})\cdot\overleftrightarrow{G}_e(\vec{r},\vec{r}'')]^T = [\overleftrightarrow{G}_e(\vec{r},\vec{r}'')]^T\cdot\overleftrightarrow{\varepsilon}(\vec{r},\vec{r}_1), \tag{20}$$

and

$$[\overleftrightarrow{\varepsilon}(\vec{r},\vec{r}_1)\cdot\overleftrightarrow{G}_e(\vec{r}_1,\vec{r}'')]^T = [\overleftrightarrow{G}_e(\vec{r}_1,\vec{r}'')]^T\cdot\overleftrightarrow{\varepsilon}(\vec{r}_1,\vec{r}), \tag{21}$$

one sees that (19) is just the negative of the integrand in (17) and hence we have proved that (17) indeed has its integrand antisymmetric upon the exchange of coordinates $\vec{r} \leftrightarrow \vec{r}_1$, and the integral simply vanishes leading again to reciprocity symmetry with

$$[\overleftrightarrow{G}_e(\vec{r}',\vec{r}'')]^T = \overleftrightarrow{G}_e(\vec{r}'',\vec{r}'). \tag{22}$$

Discussion and conclusion

Thus we have established in this work the most general condition for optical reciprocity to hold in nonlocal optics in the presence of a linear nonmagnetic medium, which is expressed in equation (18). This reduces to the well-known local limit which requires only a symmetric local dielectric tensor for the validity of reciprocity [4, 7]. It also reduces to the isotropic nonlocal case (equation (14)) 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 $\varepsilon(\vec{r},\vec{r}_1) = \varepsilon(|\vec{r}-\vec{r}_1|)$ [7]. Thus, we have clarified here that nonlocality alone does not lead to the breakdown of the reciprocity symmetry of the Green dyadic in linear optics. It is rather the asymmetry in the anisotropic response that may lead to non-reciprocity of the Green dyadic. One possible example of this is to refer to the case studied in [6] which involved the propagation of light along a cubic axis in a crystal of a 23-point group. In this case, the nonlocality tensor γ_{ijk} may be asymmetric in the sense that $\gamma_{ijk} \neq \gamma_{jik}$, which can be shown to imply an asymmetric dielectric tensor with $\varepsilon_{ij} \neq \varepsilon_{ji}$ (see footnote 4). Hence the condition expressed in equation (18) is violated and the Green dyadic does not have to obey reciprocal symmetry. To conclude, the conditions we established for the reciprocity symmetry of the Green dyadic are consistent with the previous findings in [7] which were established only within the approximation of electrostatics. Since nonlocal effects are known to be quite significant in surface optics due to the sudden change of the fields across the boundary interface between two media, the results we established here should provide useful information to researchers in their application of the reciprocity principle in the analysis of surface spectroscopy such as those performed previously in the literature [3].

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