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Reciprocity theorem for nonlocal optics: completion of proof and application to spectroscopic analysis

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Abstract
The problem of optical reciprocity in the long wavelength limit is considered in terms of the symmetry for the scalar Green's function under the Neumann boundary conditions, for materials with nonlocal and anisotropic dielectric responses. This extends the previous work in the literature which were limited either to the Dirichlet conditions or to cases of perfect conductor and local dielectric response. Application of this symmetry principle to the analysis of surface-enhanced spectroscopy for a molecule near a metallic nanoparticle is demonstrated, accounting for the nonlocal response of the particle.

Keywords: optical reciprocity, Green's function

1. Introduction

In optics or in any phenomenon involving wave propagation, the reciprocity symmetry refers to the equivalence of the signal received by two separated parties when their roles as source and observer are interchanged [1]. Since its first introduction by Lorentz more than a century ago [2], this symmetry has found interesting applications in many areas in optics ranging from device design to the analysis of spectroscopy [1]. Hence the understanding of the validity of the reciprocity symmetry is of significance both for its fundamental importance and its various potential applications.

As can be conceived, although such symmetry is quite obvious in free space, it becomes highly nontrivial in the presence of objects with arbitrary dielectric responses in the environment. For example, in the study of optics with metallic nanostructures, we expect the dominance of nonlocal dielectric response due to the large surface-to-volume ratio for these particles. Moreover, the optical interaction dynamics here can well be described in the long wavelength approximation where electrostatics can be applied. Hence nonlocal electrostatics [3] is particularly relevant in this field of nano-optics; and it is of interest to note that recent reports have highlighted the uncertainty of the reciprocity principle in the presence of nonlocal dielectric response [4]. Note that, in spite of the apparent small magnitude of the wavevector k in the long wavelength limit which seems to imply the insignificance of the nonlocal effects, such effects are still very important for nanoparticles of sub-wavelength sizes. The reason is that the nonlocal response in this case arises mainly from the rapid variation of the electric field across the boundary (i.e. the surface) of the particle (rather than over a wavelength) and hence the relative dominance of the surface for these nanoparticles ensures the prominent significance of such nonlocal effects. In addition, the long wavelength approximation refers to the *incident* wavelength being much greater than the size of the particle, leaving the possibility of the wavelength *inside* the particle being not necessarily much larger than the characteristic dimension over which nonlocal response is significant in the medium of the particle. Hence it is justified to consider a k-dependent dielectric function in the electrostatic limit for these nanoparticles.

Mathematically, the Lorentz reciprocity principle can most conveniently be expressed through the symmetry of the Green's function (or the corresponding symmetry in the Green dyadic in electrodynamics) in the form $G(\vec{r}, \vec{r}') = G(\vec{r}', \vec{r})$,

subjected to various boundary conditions. In a previous work [5], we have established this reciprocity symmetry within nonlocal electrostatics with specification to (only) the Dirichlet boundary condition.

In the mathematical literature, it is well known that the symmetry property for $G(\vec{r}, \vec{r}')$ in its arguments in the case of Neumann boundary conditions is rather nontrivial due to the constraint from Gauss's law [6]. Unlike the case of Dirichlet boundary conditions, where such symmetry can be established as a direct consequence of the Green's theorem [5–7], a special symmetrization process must be followed in order to establish the same in the case of the Neumann conditions [8]. However, the previous treatments in the literature were all limited to either perfectly conducting media [6, 8] or finitely conducting media with local dielectric response $\varepsilon(\omega)$ [7].

It is the purpose of our present work to complete our previous proof in [5] which was limited to the case of Dirichlet boundary conditions, to establish the reciprocity symmetry for the scalar Green's function in nonlocal electrostatics under the Neumann conditions, where we shall also allow the dielectric response to be anisotropic. As recently demonstrated, new metamaterials made from composites of these metallic nanoparticles can have very strong anisotropic optical responses [9]. We shall start with a brief review of our previous work for the Dirichlet condition and then follow the approach of Kim and Jackson [8] to extend the results to the case with the Neumann condition, arriving at the establishment of the Green function (reciprocity) symmetry in this case for a situation with a much more realistic dielectric response. Following this, we shall illustrate the usefulness of our result by applying it to the analysis of surface-enhanced Raman scattering (SERS) from a molecule adsorbed at a metallic nanoparticle by calculating both the local field and the radiation enhancement factors taking into account the nonlocal response of the metal particle.

2. Mathematical formulation

As in our previous work [5], we consider the Green's function satisfying the following Poisson equation in a medium with a linear, anisotropic and nonlocal dielectric response:

$$\int \mathrm{d}^3\vec{r}_1\, \vec{\nabla}\cdot[\overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1)\cdot\vec{\nabla}_1 G(\vec{r}_1, \vec{r}')] = -4\pi\delta(\vec{r}-\vec{r}'). \quad (1)$$

We want to establish the conditions under which G will be symmetric in its arguments when the potentials are subjected to the Neumann condition. In the case of the Dirichlet boundary condition, we considered in [5] another similar equation:

$$\int \mathrm{d}^3\vec{r}_1\, \vec{\nabla}\cdot[\overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1)\cdot\vec{\nabla}_1 G(\vec{r}_1, \vec{r}'')] = -4\pi\delta(\vec{r}-\vec{r}''), \quad (2)$$

and obtain the following result by manipulating equations (1) and (2) and imposing the condition $\varepsilon_{ij}(\vec{r}, \vec{r}_1) = \varepsilon_{ji}(\vec{r}_1, \vec{r})$:

$$\begin{aligned} &-4\pi G(\vec{r}'', \vec{r}') + 4\pi G(\vec{r}', \vec{r}'') \\ &= \int_S \mathrm{d}a \int \mathrm{d}^3\vec{r}_1 \{\hat{n}\cdot[G(\vec{r}, \vec{r}')\overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1)\cdot\vec{\nabla}_1 G(\vec{r}_1, \vec{r}'') \\ &\quad - G(\vec{r}, \vec{r}'')\overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1)\cdot\vec{\nabla}_1 G(\vec{r}_1, \vec{r}')]\}. \end{aligned} \quad (3)$$

While the surface integral on the RHS of (3) can be made vanishing by applying the Dirichlet condition $G = 0$ at the boundary (thus leading to the symmetry of G), similar steps cannot be taken in the case of the Neumann condition because of the well-known fact that it is incompatible with Gauss's law by setting $\vec{\nabla}G = 0$ on the boundary [6, 8]. We thus follow the approach of Kim and Jackson [8] to establish the symmetry of G in its arguments by redefining a new scalar Green function. For clarity, we shall present our derivations in two steps.

Case (i). Medium with anisotropic local response.

Let us first consider only the anisotropic response starting with the following Poisson equations:

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

Using the following identity which is a slight generalization of the one found in [7] with $\lambda_{ij} = \lambda_{ji}$ [5]:

$$\Phi\vec{\nabla}\cdot(\overleftrightarrow{\lambda}\cdot\vec{\nabla}\Psi) - \Psi\vec{\nabla}\cdot(\overleftrightarrow{\lambda}\cdot\vec{\nabla}\Phi) = \vec{\nabla}\cdot(\Phi\overleftrightarrow{\lambda}\cdot\vec{\nabla}\Psi - \Psi\overleftrightarrow{\lambda}\cdot\vec{\nabla}\Phi), \quad (5)$$

we obtain the following result similar to (3) for the local case (with $\varepsilon_{ij} = \varepsilon_{ji}$):

$$\begin{aligned} &-4\pi G(\vec{r}'', \vec{r}') + 4\pi G(\vec{r}', \vec{r}'') = \int_S \hat{n}\cdot[G(\vec{r}, \vec{r}')\overleftrightarrow{\varepsilon}(\vec{r}) \\ &\cdot\vec{\nabla}G(\vec{r}, \vec{r}'') - G(\vec{r}, \vec{r}'')\overleftrightarrow{\varepsilon}(\vec{r})\cdot\vec{\nabla}G(\vec{r}, \vec{r}')]\,\mathrm{d}a. \end{aligned} \quad (6)$$

Now let us generalize the results in Kim and Jackson [8] to introduce the following Neumann boundary conditions for an anisotropic *local* dielectric medium:

$$\begin{aligned} \hat{n}\cdot[\overleftrightarrow{\varepsilon}(\vec{r})\cdot\vec{\nabla}G(\vec{r}, \vec{r}')]|_{\vec{r}\in S} &= -\frac{4\pi}{A} \\ \hat{n}\cdot[\overleftrightarrow{\varepsilon}(\vec{r})\cdot\vec{\nabla}G(\vec{r}, \vec{r}'')]|_{\vec{r}\in S} &= -\frac{4\pi}{A} \end{aligned} \quad (7)$$

where A is the area of the closed boundary S. Equation (6) then becomes

$$\begin{aligned} &-4\pi G(\vec{r}'', \vec{r}') + 4\pi G(\vec{r}', \vec{r}'') = -\frac{4\pi}{A}\oint_S G(\vec{r}, \vec{r}'')\,\mathrm{d}a \\ &+ \frac{4\pi}{A}\oint_S G(\vec{r}, \vec{r}')\,\mathrm{d}a. \end{aligned} \quad (8)$$

We can then follow [8] to define the following symmetrized Green function satisfying the Neumann conditions:

$$G^S(\vec{r}'', \vec{r}') = G(\vec{r}'', \vec{r}') - \frac{1}{A}\oint_S G(\vec{r}, \vec{r}')\,\mathrm{d}a, \quad (9)$$

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

Case (ii). Medium with anisotropic nonlocal response

To extend the above proof to the case with nonlocal dielectric response, we generalize equation (4) as follows:

$$\begin{aligned} &\int \mathrm{d}^3\vec{r}_1\vec{\nabla}\cdot[\overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1)\cdot\vec{\nabla}_1 G(\vec{r}_1, \vec{r}')] = -4\pi\delta(\vec{r}-\vec{r}') \\ &\int \mathrm{d}^3\vec{r}_1\vec{\nabla}\cdot[\overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1)\cdot\vec{\nabla}_1 G(\vec{r}_1, \vec{r}'')] = -4\pi\delta(\vec{r}-\vec{r}''). \end{aligned} \quad (10)$$

Using the following generalized result of the identity in equation (5):

$$\int \mathrm{d}^3\vec{r}\int \mathrm{d}^3\vec{r}_1\{\Phi(\vec{r})\vec{\nabla}\cdot[\overleftrightarrow{\lambda}(\vec{r},\vec{r}_1)\cdot\vec{\nabla}_1\Psi(\vec{r}_1)] - \Psi(\vec{r})\vec{\nabla}\cdot[\overleftrightarrow{\lambda}(\vec{r},\vec{r}_1)\cdot\vec{\nabla}_1\Phi(\vec{r}_1)]\} = \int_S \mathrm{d}a\int \mathrm{d}^3\vec{r}_1\{\hat{n}\cdot[\Phi(\vec{r})\overleftrightarrow{\lambda}(\vec{r},\vec{r}_1)\cdot\vec{\nabla}_1\Psi(\vec{r}_1) - \Psi(\vec{r})\overleftrightarrow{\lambda}(\vec{r},\vec{r}_1)\cdot\vec{\nabla}_1\Phi(\vec{r}_1)]\}. \quad (11)$$

under the condition $\lambda_{ij}(\vec{r},\vec{r}') = \lambda_{ji}(\vec{r}',\vec{r})$, we derive the following from equation (10):

$$-4\pi G(\vec{r}'',\vec{r}') + 4\pi G(\vec{r}',\vec{r}'') = \int_S \mathrm{d}a\int \mathrm{d}^3\vec{r}_1 \times\{\hat{n}\cdot[G(\vec{r},\vec{r}')\overleftrightarrow{\varepsilon}(\vec{r},\vec{r}_1)\cdot\vec{\nabla}_1 G(\vec{r}_1,\vec{r}'') - G(\vec{r},\vec{r}'')\overleftrightarrow{\varepsilon}(\vec{r},\vec{r}_1)\cdot\vec{\nabla}_1 G(\vec{r}_1,\vec{r}')]\}. \quad (12)$$

Again, introducing the following generalized Neumann conditions:

$$\hat{n}\cdot\int \mathrm{d}^3\vec{r}_1\,[\overleftrightarrow{\varepsilon}(\vec{r},\vec{r}_1)\cdot\vec{\nabla}_1 G(\vec{r}_1,\vec{r}')]|_{\vec{r}\in S} = -\frac{4\pi}{A}$$
$$\hat{n}\cdot\int \mathrm{d}^3\vec{r}_1\,[\overleftrightarrow{\varepsilon}(\vec{r},\vec{r}_1)\cdot\vec{\nabla}_1 G(\vec{r}_1,\vec{r}'')]|_{\vec{r}\in S} = -\frac{4\pi}{A}, \quad (13)$$

the Green function can be symmetrized in the form as in equation (9) 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 (see the appendix).

3. An example

Here we give an example to illustrate the above symmetrization process for a metal sphere (radius a) with an isotropic (for simplicity) but nonlocal dielectric response $\varepsilon(k,\omega)$. For the case of the Dirichlet condition, we have previously [5] applied the model of the nonlocal polarizability by Fuchs and Claro [10] to obtain the following symmetric Green function:

$$G(\vec{r},\vec{r}') = 4\pi\sum_{\ell,m}\left(\frac{1}{2\ell+1}\right)\left[\frac{r_<^\ell}{r_>^{\ell+1}} - \frac{\alpha_\ell^{\mathrm{NL}}}{(rr')^{\ell+1}}\right] \times Y_{\ell m}^*(\theta,\phi)Y_{\ell m}(\theta,\phi), \quad (14)$$

where $(r_<, r_>)$ denote the smaller or greater of (r, r') and [10]

$$\alpha_\ell^{\mathrm{NL}} = \frac{\xi_\ell - 1}{\xi_\ell + (\ell+1)/\ell}a^{2\ell+1}, \quad (15)$$

with the 'effective dielectric function' given by

$$\xi_\ell(\omega) = \left[\frac{2}{\pi}(2\ell+1)a\int_0^\infty \frac{j_\ell^2(ka)}{\varepsilon(k,\omega)}\,\mathrm{d}k\right]^{-1}. \quad (16)$$

For the same problem under the Neumann condition, we first obtain the following asymmetric Green function by solving the corresponding boundary value problem:

$$G(\vec{r},\vec{r}') = \frac{1}{r_>} - \frac{1}{r} + 4\pi\sum_{\ell=1}^\infty\sum_{m=-\ell}^{\ell}\left(\frac{1}{2\ell+1}\right) \times\left[\frac{r_<^\ell}{r_>^{\ell+1}} + \frac{\ell}{\ell+1}\frac{\alpha_\ell^{\mathrm{NL}}}{(rr')^{\ell+1}}\right]Y_{\ell m}^*(\theta',\phi')Y_{\ell m}(\theta,\phi). \quad (17)$$

To symmetrize equation (17), we apply equation (9) to calculate the surface term using only the first two terms in (17) and obtain the following:

$$\frac{1}{A}\oint_S G(\vec{r},\vec{r}')\,\mathrm{d}a = \frac{1}{r'} - \frac{1}{a}. \quad (18)$$

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

$$G^S(\vec{r},\vec{r}') = \left[\frac{1}{r_>} - \left(\frac{1}{r}+\frac{1}{r'}\right) + \frac{1}{a}\right] + 4\pi\sum_{\ell=1}^\infty\sum_{m=-\ell}^{\ell}\left(\frac{1}{2\ell+1}\right)\left[\frac{r_<^\ell}{r_>^{\ell+1}} + \frac{\ell}{\ell+1}\frac{\alpha_\ell^{\mathrm{NL}}}{(rr')^{\ell+1}}\right] \times Y_{\ell m}^*(\theta',\phi')Y_{\ell m}(\theta,\phi). \quad (19)$$

4. Physical application

To illustrate the usefulness of the results we have established, we demonstrate in this section the application of the reciprocity symmetry in the form of the Lorentz lemma [2] for two dipolar sources (in obvious notations):

$$\vec{p}_1\cdot\vec{E}_2 = \vec{p}_2\cdot\vec{E}_1, \quad (20)$$

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 [11]. Note that the symmetry in the Green function and the Lorentz lemma are two equivalent statements of the reciprocity symmetry for both the Dirichlet and Neumann boundary conditions [12]. Hence our above result together with our previous work [5] have established the general validity of equation (20) for nonlocal optics in the long wavelength limit.

As pointed out by Le Ru and Etchegoin, 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 ...' [11]. Based on this distinction, it was proposed in [11] that the more correct SERS enhancement ratio should be a product of these two enhancement factors: $M_{\mathrm{SERS}} \sim M_{\mathrm{Loc}}\cdot M_{\mathrm{Rad}}$ with the latter enhancement calculable from an application of equation (20). 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 figure 1, we have essentially reproduced the key features in the corresponding figure 1 of [11], but for a much smaller metal sphere (radius = 5 nm) so that nonlocal effects are more pronounced. Note that in this figure, equation (15) 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 M_{Loc} and M_{Rad} remain for the tangentially oriented dipoles, as was first observed in [11]. The radially oriented dipole, however, gives very similar results for both the enhancement factors in

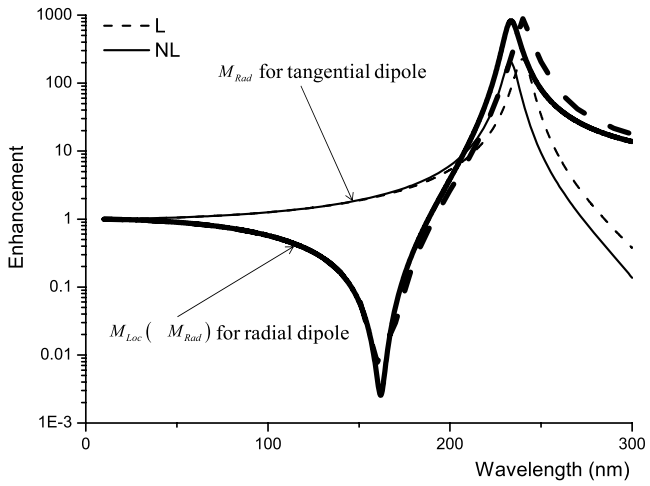

Figure 1. Spectrum of the local field and radiation enhancement factors, with the latter plotted for both radial and tangential molecular dipoles, according to both the local (dashed lines) and nonlocal (solid lines) SERS models. The molecular dipole is located at a distance of 1 nm from a silver nanosphere of 5 nm radius.

both our nonlocal calculation and the local one as reported in [11]. 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 [10].

5. Conclusion

In this work, we have demonstrated that the symmetrization process for the Neumann Green's function first introduced by Kim and Jackson [8] can indeed be generalized to the more general case of electrostatics with nonlocal and anisotropic response. This, together with our previous work [5], completes the proof of the reciprocity principle for nonlocal optics in the long wavelength limit. We have also given an example with reference to an isotropic nonlocal metal sphere to see how the construction of the symmetrized Green's function can be achieved. The result thus established should be of relevance to the study of the nano-optics involving metamaterials which are often highly anisotropic and nonlocal in their optical response. As an illustration, we have shown how the previous SERS analysis based on the reciprocity principle (Lorentz lemma) [11] can be performed within the context of nonlocal optics in the long wavelength limit.

In closing, it is of interest to note that the sharp distinction in the degree of complication between the formulations for reciprocity symmetry involving the two boundary conditions (Dirichlet versus Neumann) occurs only in electrostatics but not in electrodynamics. In the latter, the two boundary value problems can be treated more or less on an equal footing since the boundary conditions apply directly to the fields which the Green dyadics represent [12, 13].

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Appendix

Here we demonstrate explicitly that the newly constructed symmetrized Green functions in equation (9) yield the same solution for the potential with no contribution from the additional surface term. We shall only give details for the more complicated case with anisotropic nonlocal response.

To achieve this, we start with equation (11) and set $\Phi = \Phi(\vec{r})$, $\Psi(\vec{r}) = G^S(\vec{r}, \vec{r}')$ and $\overleftrightarrow{\lambda}(\vec{r}, \vec{r}_1) = \overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1)$ to obtain

$$\begin{aligned}\oint_S \mathrm{d}a \int \mathrm{d}^3\vec{r}_1 \{\hat{n} \cdot [\Phi(\vec{r})\overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1) \cdot \overrightarrow{\nabla}_1 G^S(\vec{r}_1, \vec{r}') \\ - G^S(\vec{r}, \vec{r}')\overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1) \cdot \overrightarrow{\nabla}_1 \Phi(\vec{r}_1)]\} \\ = \int \mathrm{d}^3\vec{r} \int \mathrm{d}^3\vec{r}_1 \{\Phi(\vec{r})\overrightarrow{\nabla} \cdot [\overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1) \cdot \overrightarrow{\nabla}_1 G^S(\vec{r}_1, \vec{r}')] \\ - G^S(\vec{r}, \vec{r}')\overrightarrow{\nabla} \cdot [\overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1) \cdot \overrightarrow{\nabla}_1 \Phi(\vec{r}_1)]\}. \qquad \text{(A.1)}\end{aligned}$$

With (9) into the G^S on the RHS of (A.1) and using equation (10) together with the Poisson equation for Φ, we obtain the following result:

$$\begin{aligned}\Phi(\vec{r}') = \frac{1}{A}\oint_S \Phi(\vec{r})\,\mathrm{d}a \\ + \frac{1}{4\pi}\oint_S \mathrm{d}a \int \mathrm{d}^3\vec{r}_1 G(\vec{r}, \vec{r}')\hat{n} \cdot [\overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1) \cdot \overrightarrow{\nabla}_1 \Phi(\vec{r}_1)] \\ + \int \rho(\vec{r})G(\vec{r}, \vec{r}')\,\mathrm{d}^3\vec{r} + F(\vec{r}')\Big\{\int \rho(\vec{r})\mathrm{d}^3\vec{r} \\ + \frac{1}{4\pi}\oint_S \mathrm{d}a \int \mathrm{d}^3\vec{r}_1 \hat{n} \cdot [\overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1) \cdot \overrightarrow{\nabla}_1 \Phi(\vec{r}_1)]\Big\} \\ = \frac{1}{A}\oint_S \Phi(\vec{r})\mathrm{d}a + \frac{1}{4\pi}\oint_S \mathrm{d}a \int \mathrm{d}^3\vec{r}_1 G(\vec{r}, \vec{r}')\hat{n} \\ \cdot [\overleftrightarrow{\varepsilon}(\vec{r}, \vec{r}_1) \cdot \overrightarrow{\nabla}_1 \Phi(\vec{r}_1)] + \int \rho(\vec{r})G(\vec{r}, \vec{r}')\,\mathrm{d}^3\vec{r} \qquad \text{(A.2)}\end{aligned}$$

where the surface term $F(\vec{r}') = -\frac{1}{A}\oint_S G(\vec{r}, \vec{r}')\,\mathrm{d}a$ has no contribution since the term $\{\cdots\}$ in (A.2) vanishes based on the nonlocal version of the Gauss law for Φ (cf equation (10)). Thus the symmetrized G^S leads to the same potential as the one obtained from using G.

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