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ELECTROMAGNETIC WAVE PROPAGATION IN NON-LOCAL MEDIA — NEGATIVE GROUP VELOCITY AND BEYOND

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Abstract—We study theoretically the propagation of electromagnetic waves in an infinite and homogenous medium with both temporal and spatial dispersion included. We derive a partial differential equation connecting temporal and spatial dispersion to achieve negative group velocity. Exact solutions of the equation are found and shown to lead to the possibility of exciting constant negative group velocity waves. We then investigate the effect of spatial dispersion on the power flow and derive the first-, second-, and third-order corrections of power flow due to the nonlocality in the medium. This derivation suggests a path beyond the group velocity concept.

1. INTRODUCTION

The engineering and design of new artificial media is the essence of the popular field of metamaterials. The idea is to manipulate the microscopic structure in order to produce tangible effects that can be recorded macroscopically by certain effective parameters like ϵ and μ. The main focus so far has been directed to manipulating the *temporal* dispersion of the medium.[†] However, with the steady improvement in technology, new spatial scales can be probed and manipulated, leading to interesting applications that were not possible before. One of these

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[†] Roughly speaking, temporal dispersion is captured by the functional dependence of ϵ and μ on ω.

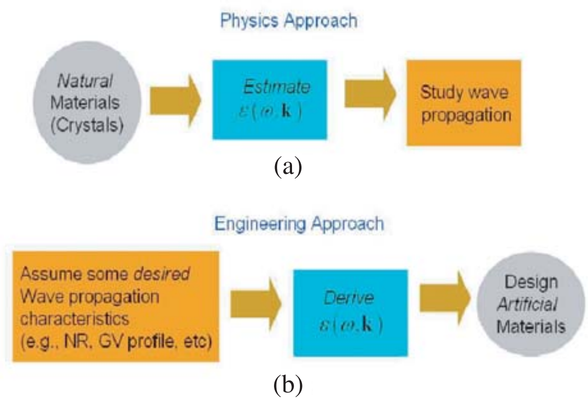

Figure 1. General philosophy of the study of electromagnetic wave propagation in dispersive media. (a) Physics approach, (b) engineering approach.

new phenomena is the nonlocal interaction between spatially separated parts of the materials, leading to what is called spatial dispersion.‡ In this case, the electromagnetic response of the medium fails to depend only on the position where we apply the external field, but depends also on the value of this field at other locations.§

It was observed that taking spatial dispersion into consideration may lead to qualitatively new phenomena not seen in conventional materials obeying classical optics (spatial dispersion is ignored). In particular, spatial dispersion can allow electromagnetic wave propagation with negative group velocity to occur, even when both the permittivity and permeability are positive [1, 2]. Such interesting behavior was originally anticipated in connection with natural materials in crystal form, where spatial dispersion is manifest, for example, in the phenomena of exciton. Recently, the same original conclusions in [1] were reinstated [3, 4]. It is still possible, however, to put the problem in a wider context by referring not only to natural crystals, but also to any type of artificial materials. To demonstrate the philosophy of the engineering approach, consider Fig. 1 where we take the medium function to be $\epsilon(\omega, \mathbf{k})$. The physics approach is illustrated in Fig. 1(a) where the starting stage is assuming certain models for the natural material under consideration (usually crystal). Then, Taylor

‡ Following the literature, we use 'spatial dispersion' and 'nonlocality' synonymously.

§ Spatial dispersion manifests itself in the functional dependence of the medium parameters on the wave vector $\mathbf{k}$. Thus, when both temporal and spatial dispersion are present, we write the permittivity and the permeability functions as $\epsilon = \epsilon(\omega, \mathbf{k})$ and $\mu = \mu(\omega, \mathbf{k})$.

series expansion of some parameters in the model (the exciton model as in [5] or the permittivity function itself as in [1]) can be applied to estimate the medium function $\epsilon(\omega, \mathbf{k})$. The next step is to apply the electromagnetic theory to study the resulted propagation. However, it is possible to invert this logic in the following way. In Fig. 1(b), we start from certain wave propagation characteristics (e.g., negative group velocity, negative-refraction propagation, etc), and then derive the medium function, $\epsilon(\omega, \mathbf{k})$, such that Maxwell's theory will allow the desired wave propagation characteristic. The future step is to find experimental methods to synthesize an artificial medium with this calculated function $\epsilon(\omega, \mathbf{k})$.

In this paper, we develop a general theoretical scheme for the *engineering* approach to electromagnetic wave propagation in dispersive materials. Our investigation is carried out through two stages. First, we focus on the special case where the group velocity is negative, which may lead (if the medium is lossless) to negative refraction. In the second stage, we go beyond the first-order approximation of the group velocity by deriving the second-, and third-order corrections of the power flow due to the spatial dispersion profile.

2. LINEAR PHENOMENOLOGICAL MODEL FOR THE MEDIUM RESPONSE

In this section, we review the basic theory of electromagnetic wave propagation in a homogenous, isotropic, and nonlocal medium described by the dielectric function $\epsilon(\omega, \mathbf{k})$. In this paper, we set $\mu = 1$.

The general relation between the electric displacement $\mathbf{D}$ and the electric field $\mathbf{E}$ is given by [7]

$$\mathbf{D}\left(\mathbf{r},\, t\right) = \int dt' \int d^3 r' \varepsilon_0 \varepsilon\left(\mathbf{r}-\mathbf{r}',\, t-t'\right) \mathbf{E}\left(\mathbf{r}',\, t'\right), \tag{1}$$

where it has been assumed that the medium is time-invariant and spatially homogeneous. The Fourier transform of the field is defined as

$$\mathbf{D}\left(\omega,\, k\right) = \int dt \int d^3 r \mathbf{D}(\mathbf{r},\, t)\, e^{j\mathbf{k}\cdot\mathbf{r}} e^{-j\omega t}, \tag{2}$$

which when applied to (1) will give

$$\mathbf{D}\left(\omega,\, \mathbf{k}\right) = \varepsilon_0 \varepsilon\left(\omega,\, \mathbf{k}\right) \mathbf{E}\left(\omega,\, \mathbf{k}\right), \tag{3}$$

where we have

$$\varepsilon\left(\omega,\, \mathbf{k}\right) = \int d\tau \int d^3 r \varepsilon\left(\mathbf{r},\, \tau\right) e^{j\mathbf{k}\cdot\mathbf{r}} e^{-j\omega\tau}. \tag{4}$$

Assume that a plane monochromatic wave is excited and propagated with fields given by

$$\mathbf{E}(\mathbf{r},t) = \mathbf{E}_0(\omega,\mathbf{k})\, e^{j\omega t - j\mathbf{k}\cdot\mathbf{r}}, \quad \mathbf{H}(\mathbf{r},t) = \mathbf{H}_0(\omega,\mathbf{k})\, e^{j\omega t - j\mathbf{k}\cdot\mathbf{r}}. \tag{5}$$

Substituting these fields into the two source-free curl Maxwell's equation, taking the curl of both sides, and using (3), we obtain

$$\mathbf{k}\times\mathbf{k}\times\mathbf{E}_0(\omega,\mathbf{k}) - \left(\omega^2/c^2\right)\varepsilon(\omega,\mathbf{k})\,\mu\mathbf{E}_0(\omega,\mathbf{k}) = 0. \tag{6}$$

From the divergence Maxwell's equation, we find

$$\varepsilon(\omega,\mathbf{k})\,\mathbf{k}\cdot\mathbf{E}_0(\omega,\mathbf{k}) = 0. \tag{7}$$

We distinguish here between the transverse (T) and longitudinal (L). Let us assume that

$$\epsilon(\omega,\mathbf{k}) \neq 0. \tag{8}$$

Then, from (7), we obtain $\mathbf{k}\cdot\mathbf{E}_0(\omega,\mathbf{k}) = 0$. This condition when applied to (6) immediately gives the dispersion relation for the transverse waves

$$\mathbf{k}\cdot\mathbf{k} = (\omega/c)^2\, n^2(\omega,\mathbf{k}), \tag{9}$$

where we have *defined* the index of refraction as

$$n(\omega,\mathbf{k})^2 \equiv \varepsilon(\omega,\mathbf{k})\,\mu. \tag{10}$$

The longitudinal modes can be obtained by setting $\epsilon(\omega,\mathbf{k}) = 0$. Therefore, (7) is satisfied with $\mathbf{k}\cdot\mathbf{E}_0(\omega,\mathbf{k}) \neq 0$. That is, contrary to the transverse wave, the wave vector here is not orthogonal to the electrical field amplitude.[‖] Strictly speaking, the dispersion relations for the L and T modes are different and should be distinguished from each other by using appropriate subscripts whenever possible. However, in this paper the main focus will be on transverse waves so these subscripts will be omitted for the simplicity of notation.

3. NEGATIVE GROUP VELOCITY MEDIA

Let us start with a very general index of refraction given by $n = n(\omega,\mathbf{k})$. The resulted dispersion relation for the transverse mode

[‖] Notice that when spatial dispersion is ignored, $\varepsilon(\omega,\mathbf{k}) = \varepsilon(\omega)$. Hence, for the L modes the equation $\varepsilon(\omega,\mathbf{k}) = 0$ can be satisfied only at discrete frequencies. In other words, the group velocity $\partial\omega_L/\partial\mathbf{k}$ is zero and no energy flow can be associated with this type of modes (an exception is some forms of plasmas [7]). Now, when spatial dispersion is considered, relation (8) is not only satisfied at *continuous* range of frequencies, but may lead also to nonzero group velocity, contributing to the power flow in the medium.

propagating in infinite, homogeneous, and isotropic medium is given by (9). The group velocity is defined as [10, 11]

$$\mathbf{v}_g = \frac{\partial \omega}{\partial \mathbf{k}} = \nabla_{\mathbf{k}} \omega = \hat{x} \frac{\partial \omega}{\partial k_x} + \hat{y} \frac{\partial \omega}{\partial k_y} + \hat{z} \frac{\partial \omega}{\partial k_z}. \tag{11}$$

Our goal now is to derive an equation connecting the spatial and temporal dispersion such that the resulted medium supports negative group velocity propagation.

Differentiate both sides of (9) with respect to k_α, where $\alpha = x, y, z$, we get

$$\frac{k_\alpha}{k} = \frac{\partial n(\omega, \mathbf{k})}{\partial k_\alpha} \frac{\omega}{c} + \frac{\partial \omega}{\partial k_\alpha} \frac{n(\omega, \mathbf{k})}{c}. \tag{12}$$

Using the following chain rule

$$\frac{\partial n(\omega, \mathbf{k})}{\partial k_\alpha} = \frac{\partial n(\omega, \mathbf{k})}{\partial \mathbf{k}} \cdot \frac{\partial \mathbf{k}}{\partial k_\alpha} + \frac{\partial n(\omega, \mathbf{k})}{\partial \omega} \frac{\partial \omega}{\partial k_\alpha}, \tag{13}$$

Equation (12) can be solved for $\partial \omega / \partial k_\alpha$ to give

$$v_{g\alpha} = \frac{\partial \omega}{\partial k_\alpha} = \frac{\frac{k_\alpha}{k} - \frac{\omega}{c} \frac{\partial n}{\partial \mathbf{k}} \cdot \frac{\partial \mathbf{k}}{\partial k_\alpha}}{\frac{n}{c} + \frac{\omega}{c} \frac{\partial n}{\partial \omega}}. \tag{14}$$

Let us now calculate the dot product between $\mathbf{v}_g$ and $\mathbf{k}$. We write

$$\mathbf{v}_g \cdot \mathbf{k} = \sum_\alpha v_{g\alpha} k_\alpha = \frac{1}{\frac{n}{c} + \frac{\omega}{c} \frac{\partial n}{\partial \omega}} \sum_\alpha \left[\frac{k_\alpha}{k} - \frac{\omega}{c} \frac{\partial n}{\partial \mathbf{k}} \cdot \frac{\partial \mathbf{k}}{\partial k_\alpha} \right] k_\alpha. \tag{15}$$

By multiplying the numerator and the denominator of (15) by k, we obtain

$$\mathbf{v}_g \cdot \mathbf{k} = \frac{k}{\frac{n}{c} + \frac{\omega}{c} \frac{\partial n}{\partial \omega}} \sum_\alpha \left[\frac{k_\alpha^2}{k^2} - \frac{\omega}{c} \frac{\partial n}{\partial \mathbf{k}} \cdot \frac{\partial \mathbf{k}}{\partial k_\alpha} \frac{k_\alpha}{k} \right]. \tag{16}$$

Notice that

$$\sum_\alpha \frac{k_\alpha^2}{k^2} = 1 \tag{17}$$

and

$$-\sum_\alpha \frac{\omega}{c} \frac{\partial n}{\partial \mathbf{k}} \cdot \frac{\partial \mathbf{k}}{\partial k_\alpha} \frac{k_\alpha}{k} = -\frac{\omega}{c} \frac{\partial n}{\partial \mathbf{k}} \cdot \frac{\mathbf{k}}{k} = -\frac{\omega}{c} \frac{\partial n}{\partial k}, \tag{18}$$

where the relation $\partial \mathbf{k}/\partial k_\alpha = \hat{a}_\alpha$ (unit vector in the α-direction) has been used. Therefore, Equation (16) reduces to

$$\mathbf{v}_g \cdot \mathbf{k} = k \frac{c - \omega \partial n/\partial k}{n + \omega \partial n/\partial \omega}. \tag{19}$$

It can be shown by the same procedure that $|\mathbf{v}_g|\,|\mathbf{k}| = |\mathbf{v}_g \cdot \mathbf{k}|$; i.e., the angle cosine $\cos\theta = \mathbf{v}_g \cdot \mathbf{k}/|\mathbf{v}_g|\,|\mathbf{k}|$ is either 1 or -1. This is because we assumed the medium to be homogenous and isotropic.¶ Therefore, we define the *negative* group velocity as the case when the angle between $\mathbf{v}_g$ and $\mathbf{k}$ is 180°. By defining $\gamma \equiv |\mathbf{v}_g|$ and assuming $k > 0$, Equation (19) can give the following result for negative group velocity

$$\frac{\omega}{c}\frac{\partial n(\omega, \mathbf{k})}{\partial k} - \frac{\gamma}{c}\left(1 + \omega \frac{\partial}{\partial \omega}\right) n(\omega, \mathbf{k}) = 1. \tag{20}$$

We call this partial differential equation the dispersion engineering equation for negative group velocity. In Sec. 4 and Sec. 5, we will study the physical and mathematical behavior of its solutions, respectively.

4. THE PHYSICAL MEANING OF NEGATIVE GROUP VELOCITY

Negative refraction must occur at the interface separating the conventional and the meta- materials if the Poynting vector $\mathbf{S}$ and the wave vector $\mathbf{k}$ in the metamaterial are oriented opposite to each other [8]. That is, if we have

$$\mathbf{S} \cdot \mathbf{k} < 0. \tag{21}$$

Here, Equation (21) is one sufficient condition for obtaining negative refraction in our metamaterial. Notice that also Snells law has to hold true in addition to condition (21) in order to obtain negative refraction. For detailed discussion of the application of boundary conditions at the interface between spatially dispersive media and a conventional medium, see [1, 3], and [4]. The question now is whether the requirement

$$\mathbf{v}_g \cdot \mathbf{k} < 0 \tag{22}$$

is equivalent to condition (21). This is identical to asking whether the Poynting vector $\mathbf{S}$ and the group velocity $\mathbf{v}_g$ are oriented in the same direction. The answer is that in general they are not [1, 9]. The two

¶ In other words, the dependence of the refraction index n on the wave vector $\mathbf{k}$ can be written identically as either $n(\mathbf{k})$ or $n(k)$.

vectors $\mathbf{v}_g$ and $\mathbf{S}$ become parallel if the medium is lossless or has small dissipation. In this case, it is possible to write [1, 9–11]

$$\mathbf{S} = W\mathbf{v}_g, \tag{23}$$

where W is the total energy density stored in the medium. Since at thermodynamic equilibrium $W > 0$ [10], it follows that $\mathbf{S}$ and $\mathbf{v}_g$ are parallel. For lossy media, the angle between these two vectors may vary considerably depending on the material parameters; no *a priori* conclusion can be stated without examining the specific dispersion and losses profile.

In some of the published literature about metamaterials, and following the original work of Veselago [12], it is common to associate negative-refraction media with the "handedness" as being left-handed, in contrast to the normal right-handedness of conventional materials. However, it has been noticed long before Veselago's work that negative refraction is a general phenomenon that should be addressed in terms of group velocities, not merely the algebraic signs of the medium parameters [8, 13]. In particular, it was predicted that negative refraction may occur even when both ϵ and μ are positive [1], a situation consistent with Equation (20), which gives the exact details of how to choose the temporal and spatial dispersion of the medium such that the resulted waves propagate possesses negative group velocity. If, furthermore, the medium has low dissipation, Equation (21) is satisfied and the medium will support negative refraction.

The correct interpretation of the group velocity is that it is the velocity of propagation of the smoothly varying wave packet's *envelope* of relatively small bandwidth (first-order approximation). This velocity is the same as the energy velocity in lossless media but in lossy materials this is not correct in general [9, 15].

From (19), we write

$$\mathbf{v}_g = \frac{c - \omega\partial n/\partial k}{n + \omega\partial n/\partial\omega}\hat{a}_{\mathbf{k}}. \tag{24}$$

The phase velocity is given by

$$\mathbf{v}_p = \frac{\omega}{k}\frac{\mathbf{k}}{k} = \frac{c}{|n|}\hat{a}_{\mathbf{k}}, \tag{25}$$

where $\hat{a}_{\mathbf{k}}$ is a unit vector in the direction of $\mathbf{k}$. Assume that a reference was chosen in the spatial direction pointing away from a given (observation) point. Thus, with respect to this direction, each of $\mathbf{v}_g$ and $\mathbf{v}_p$ can be either positive or negative. We will show now that the condition (21) is a candidate for defining metamaterials.

We accomplish this by identifying the following four distinct cases, depending on the algebraic signs of the group velocity and the wave vector[+]

I. $v_p > 0$, $v_g > 0$, $n > 0$. This is the conventional medium. Here the wave envelope and phase propagate away from the observation point. Positive refraction occurs all the time.

II. $v_p < 0$, $v_g > 0$, $n < 0$. This is the so-called Veselago medium. Here the wave envelope propagates away from the observation point while phase propagates towards the point. Negative refraction occurs in this case.

III. $v_p > 0$, $v_g < 0$, $n > 0$. This is the main interest of the present paper. Here the wave envelope propagates toward the observation point while the phase propagates away from the observation point. However, although n is positive, negative refraction may occur if the medium has small dissipation and a carefully chosen profile of the spatial dispersion is implemented.

IV. $v_p < 0$, $v_g < 0$, $n < 0$. Here, both the wave envelope and the phase propagate toward the observation point. In this case, negative refraction may occur assuming small losses, but this cannot be achieved through temporal dispersion only (see Apendix A).

To obtain better understanding of the four cases listed above, we need to resort to the important distinction between normal and anomalous dispersion.[*] We will prove now the previous statements. Cases I and II are self-evident and no further illustrations are needed here. For Case III, assume first that the medium has small losses so we can apply (23) and (24) to write

$$\mathbf{S} \cdot \hat{a}_{\mathbf{k}} = W \frac{c - \omega \partial n / \partial k}{n + \omega \partial n / \partial \omega}. \tag{26}$$

Consider first a medium exhibiting only temporal dispersion ($\partial n/\partial k = 0$). Since $n > 0$, then the only way to possibly achieve negative $\mathbf{S} \cdot \hat{a}_{\mathbf{k}}$ is to have $\partial n/\partial \omega < 0$. This is, however, the region of anomalous dispersion, which corresponds usually to high losses. This means that negative refraction is *not* guaranteed in this case. We must stress

[+] Strictly speaking, the quantities v_p and v_g appearing below are the dot products of the corresponding vectors in (24) and (25) with a unit vector in the direction of the chosen reference.

[*] Normal dispersion is characterized by a medium function, say n for example, which is monotonically increasing. Hence, $\partial n/\partial \omega > 0$. Anomalous dispersion is then defined as the opposite case when $\partial n/\partial \omega < 0$. In general, we know from experiments that anomalous dispersion is correlated with lossy media [14, 15]. For a rigorous proof that anomalous dispersion is a necessary condition for the medium to be lossy see [10].

here that a metamaterial in which the group velocity is negative is still meaningful even when there is no negative refraction. We need to refer to $\mathbf{v}_g$ as only the velocity in which a wave packet propagates without appreciable distortion [15]. Such media has been already demonstrated experimentally more than three decades ago where the group velocity was reportedly measured with supraliminal negative values in carefully designed media having anomalous dispersion [16–18].

When considering spatial dispersion, the quantity $\mathbf{S} \cdot \hat{a}_{\mathbf{k}}$ in (26) can be made negative by solutions of Equation (20) as we will show in Sec. 5. In this case, no assumption like $\partial n/\partial\omega < 0$ is necessary and condition (21) can be satisfied in low dissipation media, leading to negative refraction. Thus, spatial dispersion is the decisive factor in achieving negative refraction in such kind of metamaterials (Case III).

Finally, Case IV will be treated briefly here. Consider first the scenario when the spatial dispersion is neglected. Here, since n is already negative, Equation (24) may suggest that obtaining negative group velocity in a negative phase velocity medium is possible without operating in the region of anomalous dispersion. However, in Appendix A, we show that causality considerations does not allow this. Experimental data in [19], [20] are consistent with this conclusion as it shows that v_g and v_p become simultaneously negative in the region of anomalous dispersion. Therefore, in Case IV it is not always guaranteed to observe negative refraction even though $n < 0$. The situation again will change when spatial dispersion is considered where careful choice of the dispersion profile may lead to negative group velocity in the normal dispersion region, leading therefore to negative refraction.

5. EXACT SOLUTION FOR THE DISPERSION ENGINEERING EQUATION

5.1. Development of the Exact Solution

Before proceeding into the analytical solution of (20), it will be insightful to provide a geometrical interpretation of this solution. The relation $\omega = \omega(k)$ is nothing but the dispersion law of the medium. We may say that this equation determines a family of curves in the plane over which the general solution $n = n(\omega, k)$ will be constructed. Notice that this function is a surface in the ω-k-n 3-dimensional space. Therefore, as shown in Fig. 2, one can consider the family of curves $\omega = \omega(k)$ as base curves upon which the solution surface would be found.

Let us consider the dispersion relation $\omega = \omega(k)$ as an implicit parametrization in terms of k. Then, it is possible to write

$$\frac{d}{dk}n\left(\omega\left(k\right),k\right)=\frac{\partial n}{\partial k}+\frac{\partial n}{\partial \omega}\frac{d\omega}{dk}. \tag{27}$$

Using the definition of group velocity and the dispersion relation, we have

$$\frac{d\omega}{dk}=-\gamma\left(\omega,k\right). \tag{28}$$

Thus, from (27) and (28) we obtain

$$\begin{aligned}\frac{\omega}{c}\frac{dn}{dk}-\frac{\gamma}{c}n &= \frac{\omega}{c}\left\{\frac{\partial n}{\partial k}+\frac{\partial n}{\partial \omega}\frac{d\omega}{dk}\right\}-\frac{\gamma}{c}n\\ &= \frac{\omega}{c}\left\{\frac{\partial n}{\partial k}-\gamma\frac{\partial n}{\partial \omega}\right\}-\frac{\gamma}{c}n\\ &= \frac{\omega}{c}\frac{\partial n}{\partial k}-\frac{\omega\gamma}{c}\frac{\partial n}{\partial \omega}-\frac{\gamma}{c}n. \end{aligned} \tag{29}$$

From (27), we readily get the following *ordinary* differential equation

$$\frac{\omega\left(k\right)}{c}\frac{dn\left(\omega,k\right)}{dk}-\frac{\gamma\left(\omega,k\right)}{c}n\left(\omega,k\right)=1. \tag{30}$$

Therefore, the solution to the original partial differential Equation (20) can be thought of as solving the ordinary differential Equation (30) along the path (curve) described by the solution of the ordinary differential Equation (28). Notice that γ is in general an arbitrary positive function of both ω and k. Therefore, although the problem has been reduced into two ordinary differential equations, still no general solution is available analytically.

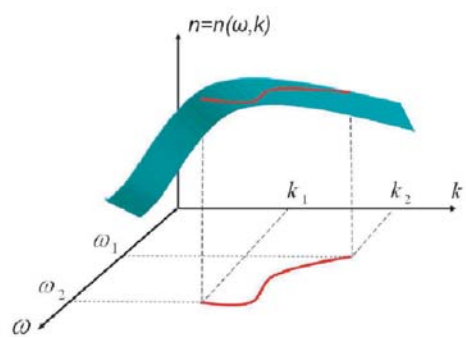

Figure 2. Geometric interpretation for solution of the dispersion engineering Equation (20) [2]. The solution $n = n(\omega, k)$ is a surface in the ω-k-n 3-dimensional space.

5.2. Solution for k-dependent Group Velocity

Consider the boundary-value problem consisting of the partial differential Equation (20) together with

$$\frac{\partial \gamma}{\partial \omega} = 0, \quad n\left(\omega, k = k_1\right) = \phi\left(\omega\right), \tag{31}$$

where $\omega_1 < \omega < \omega_2$, $\omega_1 > 0$, $k_1 > 0$. Here, $k_1 < k_2$ and $\omega_1 < \omega_2$ are positive real numbers and $\phi\left(\omega\right)$ is a general function representing the boundary condition of the problem. Since γ is function of k only, it is possible to directly integrate Equation (28) to obtain

$$\omega\left(k\right) = -\int dk\gamma + a. \tag{32}$$

Substituting (32) into (33), we find

$$\left(-\int dk\gamma + a\right)\frac{dn}{dk} - \gamma n = c. \tag{33}$$

Then we can write

$$\frac{dn}{dk} - \frac{\gamma}{-\int dk\gamma + a} n = \frac{c}{-\int dk\gamma + a}. \tag{34}$$

This equation admits the following exact solution

$$\begin{aligned} n\left(k\right) &= e^{-F}\left(c\int \frac{dk\, e^{F}}{-\int dk\gamma\left(k\right) + a} + b\right), \\ F &= \int \frac{dk\gamma\left(k\right)}{\int dk\gamma\left(k\right) - a}, \end{aligned} \tag{35}$$

where a and b are constants to be determined later. Since we are solving the ordinary differential Equation (30) along the trajectory specified by (28), then b is not independent of a, and we may write in general $b = f\left(a\right)$, where the function f is to be fixed by enforcing the boundary condition imposed on the $n = n(\omega, k)$.

5.3. Solution for Constant Group Velocity

Let us evaluate the general solution for the case when γ is constant; i.e., we want to impose the condition that the group velocity is constant but negative. In this case, (32) gives

$$\omega\left(k\right) = -\gamma k + a. \tag{36}$$

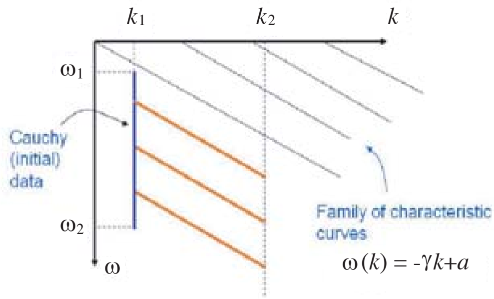

Figure 3. The characteristic curves for the problem of solving (20) under the assumption of constant negative group velocity [2].

In Fig. 3, we show the geometric structure of this case. The linear segments shown between the two lines $k = k_1$ and $k = k_2$ in the $k - \omega$ plane represent the permissible characteristic curves. Substituting (36) into the general solution (35), evaluating the integrals, we obtain

$$n = \frac{ck + f(a)}{-\gamma k + a}. \tag{37}$$

By substituting $a = \omega + \gamma k$, we arrive to

$$n(\omega, k) = \frac{ck + f(\omega + \gamma k)}{\omega}. \tag{38}$$

To find the function f, apply the boundary condition $n(\omega, k = k_1) = \phi(\omega)$ to get $f(\omega + \gamma k_1) = \omega\phi(\omega) - ck_1$. Using the transformation $x = \omega + \gamma k_1$, the function f can be expressed as

$$f(x) = (x - \gamma k_1)\,\phi(x - \gamma k_1) - ck_1. \tag{39}$$

Finally, the general solution will take the form

$$n(\omega, k) = \frac{c(k - k_1)}{\omega} + \frac{1}{\omega}\left[\omega + \gamma(k - k_1)\right]\phi(\omega + \gamma(k - k_1)). \tag{40}$$

The importance of this general expression is evident. Dispersion engineering in this case amounts to choosing the right spatial dispersion profile, starting at initial data consisting of the temporal dispersion ϕ defined at a specific value of the wavenumber k, such that the resulted wave propagation exhibits a constant negative group velocity. Therefore, while the desired anti-parallel nature of $\mathbf{S}$ and $\mathbf{k}$ is obtained in low-loss media, the group velocity does not vary with frequency, leading to minimal distortion in signal transmission.

For a medium with small losses, Kramers-Kronig relations implies

$$\frac{\partial n}{\partial \omega} = \frac{(k - k_1)(c - \gamma\phi)}{-\omega^2} + \frac{\gamma (k - k_1)}{\omega} \frac{\partial \phi}{\partial \omega} > 0. \tag{41}$$

Let us choose an initial data in the normal dispersion regime (low losses) such that $\partial\phi/\partial\omega = A\omega^2$, $A > 0$. In this case

$$\frac{\partial n}{\partial \omega} = \frac{(k - k_1)(c - \gamma\phi)}{-\omega^2} + \gamma (k - k_1) A\omega, \tag{42}$$

which can be satisfied, for example, at sufficiently high frequencies. Another possibility would be to choose $\phi(\omega + \gamma(k - k_1)) > c/\gamma > 0$.

5.4. Zero-temporal Dispersion

The consideration of spatial dispersion will lead to a new picture for the special case when temporal dispersion is ignored. We start by the following simple theorem: *Assuming constant $\gamma > 0$ (constant negative group velocity), it is impossible to guarantee achieving negative refraction when the temporal dispersion is zero.* To prove this, notice that it follows from (24) that for constant negative group velocity to be achieved and $\partial n/\partial\omega = 0$, the corresponding spatial dispersion profile is simply $n = -c/\gamma$. Thus, the refraction index is also independent of k and negative. However, as we discussed in Sec. 4, to conclude that the medium supports negative refraction we must have small losses; negative refraction *and* $n < 0$ can be guaranteed to occur only with double negative material (Case II in Sec. 4). However, we notice that to obtain negative index of refraction in such a medium, the material response *must* exhibit temporal dispersion [12]. Therefore, we conclude that there is no physical solution corresponding to n in the sense above.

Moreover, from (24) it follows that in the case of zero-temporal dispersion the group velocity takes the form

$$v_g = \frac{c}{n} - \frac{\omega}{n} \frac{\partial n}{\partial k}. \tag{43}$$

We notice two important things here. First, although n does not depend on frequency, the group velocity will have a linear dependence on frequency for nonzero spatial dispersion. Thus, it is not true to state that having a refraction index n independent of ω leads to constant negative group velocity; this statement is true only if spatial dispersion is ignored. Second, from (43), it is clear that one can achieve negative group velocity if we choose $\partial n \partial k$ large enough. In particular, if n is positive, we just need to satisfy $\partial n/\partial k > c/\omega$. Therefore, in the case of small losses, it is possible to achieve negative refraction in media that have no temporal dispersion by careful choice of the spatial dispersion profile.

6. HIGHER-ORDER CORRECTION OF THE POWER FLOW — BEYOND GROUP VELOCITY

In this section, we will re-examine the problem of electromagnetic wave propagation in nonlocal media through the more general perspective of power flow. There are several reasons for that. First, the group velocity concept developed in the previous parts is inherently a first-order approximation, limiting its applicability to a certain form of the field (narrow-band signals). Second, the consideration of power and energy quantities should lead naturally to a deeper understanding of wave propagation in nonlocal media since comparison with microscopic energy analysis can guide the interpretation of the results derived here using the macroscopic field theory. Finally, newer phenomena appear when spatial dispersion is considered in the problem. As we will show by the end of this section, higher-order corrections to the power flow will be generally dependent on the structure of the field assumed throughout the discussion, in this case the quasi-monochromatic field shown in (45) below.

To summarize, we consider here the effect of spatial dispersion on the direction of power flow in dissipation-free media. The consideration of higher-order corrections of the power flow, resulting from taking into account the effect of nonlocality in the medium, leads to important corrections, which can not be described within the group velocity paradigm developed in the previous parts of this paper.

Our starting point will be the Poynting's theorem

$$\nabla \cdot \left(\bar{\mathcal{E}} \times \bar{\mathcal{H}}\right) = -\left(\bar{\mathcal{E}} \cdot \frac{\partial \bar{\mathcal{D}}}{\partial t} + \bar{\mathcal{H}} \cdot \frac{\partial \bar{\mathcal{B}}}{\partial t}\right). \tag{44}$$

The field is assumed to be in the form

$$\begin{aligned} \bar{\mathcal{E}}\left(\mathbf{r},t\right) &= \mathrm{Re}\left\{\mathbf{E}\left(\mathbf{r},t\right)\right\} = \mathrm{Re}\left\{\mathbf{E}_0\left(\mathbf{r},t\right) e^{-j\left(\mathbf{k}_0\cdot\mathbf{r}-\omega_0 t\right)}\right\} \\ &= \frac{1}{2}\left\{\mathbf{E}_0\left(\mathbf{r},t\right) e^{-j\left(\mathbf{k}_0\cdot\mathbf{r}-\omega_0 t\right)} + c.c.\right\}, \end{aligned} \tag{45}$$

where $\mathbf{E}_0$ is the (lowpass) complex amplitude and and $c.c.$ denotes the complex conjugate term. We assume that the temporal spectrum of the field, centered around ω_0, is narrowband. However, the spatial spectrum, centered around $\mathbf{k}_0$, could be wideband. By applying a simple change of variables in the Fourier transform of the field, we can write the following

$$\mathbf{E}(\omega, \mathbf{k}) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dt dr^3 \, \mathbf{E}(\mathbf{r}, t) \, e^{j(\mathbf{k}\cdot\mathbf{r} - \omega t)} = \mathbf{E}_0 (\omega - \omega_0, \, \mathbf{k} - \mathbf{k}_0), \quad (46)$$

where $\mathbf{E}_0(\omega, \mathbf{k})$ is the Fourier transform of $\mathbf{E}_0(\mathbf{r}, t)$, from which we can see the motivation for calling $\mathbf{E}_0$ the low-pass equivalent of the field.

By applying the inverse Fourier transform to write $\mathbf{D}$ and $\mathbf{E}$ in (1), differentiating the result with respect to time, we obtain

$$\frac{\partial \mathbf{D}}{\partial t} = \frac{1}{2\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} d\omega \, dk^3 j\omega\varepsilon_0 \varepsilon(\omega, \mathbf{k}) \, \mathbf{E}(\omega, \mathbf{k}) \, e^{-j(\mathbf{k}\cdot\mathbf{r} - \omega t)}. \quad (47)$$

Next, the dielectric function is expanded using Taylor series but we retain only the first-order approximation for the temporal dispersion while we keep the third-order approximation for the spatial dispersion[♯]. Therefore, we obtain

$$\begin{aligned} \omega\varepsilon(\omega, \mathbf{k}) &= \omega_0 \varepsilon(\omega_0, \mathbf{k}_0) + \partial(\omega\varepsilon)/\partial\omega\tilde{\omega} \\ &\quad + \omega_0 \left(\tilde{\mathbf{k}} \cdot \nabla_{\mathbf{k}}\right) \varepsilon + (1/2)\omega_0 \left(\tilde{\mathbf{k}} \cdot \nabla_{\mathbf{k}}\right)^2 \varepsilon \\ &\quad + (1/6)\omega_0 \left(\tilde{\mathbf{k}} \cdot \nabla_{\mathbf{k}}\right)^3 \varepsilon, \end{aligned} \quad (48)$$

where $\mathbf{k} = \mathbf{k}_0 + \tilde{\mathbf{k}}$ and $\omega = \omega_0 + \tilde{\omega}$ and all derivatives are evaluated at ω_0 and $\mathbf{k}_0$. By substituting (48) into (47), we obtain

$$\begin{aligned} \frac{\partial \mathbf{D}}{\partial t} &= \frac{j\omega_0\varepsilon_0}{2\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} d\omega \, dk^3 \varepsilon(\omega_0, \mathbf{k}_0) \, \mathbf{E}(\omega, \mathbf{k}) \, e^{-j(\mathbf{k}\cdot\mathbf{r} - \omega t)} \\ &\quad + \frac{\varepsilon_0}{2\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} d\omega \, dk^3 \frac{\partial(\omega\varepsilon)}{\partial\omega} (j\tilde{\omega}) \, \mathbf{E}(\omega, \mathbf{k}) \, e^{-j(\mathbf{k}\cdot\mathbf{r} - \omega t)} \\ &\quad + j\frac{\omega_0\varepsilon_0}{2\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} d\omega \, dk^3 \, \left(\tilde{\mathbf{k}} \cdot \nabla_{\mathbf{k}}\right) \varepsilon \mathbf{E}(\omega, \mathbf{k}) \, e^{-j(\mathbf{k}\cdot\mathbf{r} - \omega t)} \end{aligned}$$

[♯] For the case of wideband signals, i.e., fields that vary rapidly in time, the time average integration can not be performed without further information about the specific mathematical function under study. To keep the derivations at the most general level, we restricted our presentation in this paper to signals that are slow enough in time.

$$+\frac{j\varepsilon_0\omega_0}{2}\frac{1}{2\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}d\omega\,dk^3\left(\tilde{\mathbf{k}}\cdot\nabla_{\mathbf{k}}\right)^2\varepsilon\mathbf{E}\left(\omega,\mathbf{k}\right)e^{-j(\mathbf{k}\cdot\mathbf{r}-\omega t)}$$
$$+\frac{j\varepsilon_0\omega_0}{6}\frac{1}{2\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}d\omega\,dk^3\left(\tilde{\mathbf{k}}\cdot\nabla_{\mathbf{k}}\right)^3\varepsilon\mathbf{E}\left(\omega,\mathbf{k}\right)e^{-j(\mathbf{k}\cdot\mathbf{r}-\omega t)}. \quad (49)$$

By applying the change of integration variables $\mathbf{k} = \mathbf{k}_0 + \tilde{\mathbf{k}}$ and $\omega = \omega_0 + \tilde{\omega}$ and employing (46), Equation (49) can be reduced to

$$\begin{aligned}\frac{\partial\mathbf{D}}{\partial t} =\ & j\omega_0 A\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}d\tilde{\omega}\,d\tilde{k}^3\varepsilon\left(\omega_0,\mathbf{k}_0\right)\mathbf{E}_0\left(\tilde{\omega},\tilde{\mathbf{k}}\right)e^{-j\left(\tilde{\mathbf{k}}\cdot\mathbf{r}-\tilde{\omega}t\right)}\\ &+A\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}d\tilde{\omega}\,d\tilde{k}^3\frac{\partial\left(\omega\varepsilon\right)}{\partial\omega}\left(j\tilde{\omega}\right)\mathbf{E}_0\left(\tilde{\omega},\tilde{\mathbf{k}}\right)e^{-j\left(\tilde{\mathbf{k}}\cdot\mathbf{r}-\tilde{\omega}t\right)}\\ &+j\omega_0 A\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}d\tilde{\omega}\,d\tilde{k}^3\left(\tilde{\mathbf{k}}\cdot\nabla_{\mathbf{k}}\right)\varepsilon\mathbf{E}_0\left(\tilde{\omega},\tilde{\mathbf{k}}\right)e^{-j\left(\tilde{\mathbf{k}}\cdot\mathbf{r}-\tilde{\omega}t\right)}\\ &+\frac{1}{2}j\omega_0 A\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}d\tilde{\omega}\,d\tilde{k}^3\left(\tilde{\mathbf{k}}\cdot\nabla_{\mathbf{k}}\right)^2\varepsilon\mathbf{E}_0\left(\tilde{\omega},\tilde{\mathbf{k}}\right)e^{-j\left(\tilde{\mathbf{k}}\cdot\mathbf{r}-\tilde{\omega}t\right)}\\ &+\frac{1}{6}j\omega_0 A\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}d\tilde{\omega}\,d\tilde{k}^3\left(\tilde{\mathbf{k}}\cdot\nabla_{\mathbf{k}}\right)^3\varepsilon\mathbf{E}_0\left(\tilde{\omega},\tilde{\mathbf{k}}\right)e^{-j\left(\tilde{\mathbf{k}}\cdot\mathbf{r}-\tilde{\omega}t\right)}. \quad (50)\end{aligned}$$

where $A = (\varepsilon_0/2\pi)\exp\left(-j\mathbf{k}_0\cdot\mathbf{r}+j\omega_0 t\right)$. Using the Fourier transform pair $\nabla_{\mathbf{r}} \to -j\tilde{\mathbf{k}}$ and then applying to the result the inverse Fourier transform definition of $\mathbf{E}_0\left(\mathbf{r},\,t\right)$, Equation (50) can be simplified as follows

$$\begin{aligned}\frac{\partial\mathbf{D}}{\partial t} =\ & j\omega_0\varepsilon_0\varepsilon e^{-j(\mathbf{k}_0\cdot\mathbf{r}-\omega_0 t)}\mathbf{E}_0\left(\mathbf{r},\,t\right)\\ &+\varepsilon_0 e^{-j(\mathbf{k}_0\cdot\mathbf{r}-\omega_0 t)}\frac{\partial\left(\omega\varepsilon\right)}{\partial\omega}\frac{\partial}{\partial t}\mathbf{E}_0\left(\mathbf{r},\,t\right)\\ &-\varepsilon_0\omega_0 e^{-j(\mathbf{k}_0\cdot\mathbf{r}-\omega_0 t)}\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\right)\varepsilon\mathbf{E}_0\left(\mathbf{r},\,t\right)\\ &+\frac{j\omega_0\varepsilon_0}{2}e^{-j(\mathbf{k}_0\cdot\mathbf{r}-\omega_0 t)}\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\right)^2\varepsilon\mathbf{E}_0\left(\mathbf{r},\,t\right)\\ &+\frac{\omega_0\varepsilon_0}{6}e^{-j(\mathbf{k}_0\cdot\mathbf{r}-\omega_0 t)}\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\right)^3\varepsilon\mathbf{E}_0\left(\mathbf{r},\,t\right), \quad (51)\end{aligned}$$

which contains only the complex low-pass equivalent representation of the field, i.e., $\mathbf{E}_0$. Since the field was assumed to be slow enough in time (narrowband temporal frequency spectrum), we can compute the time-average as $\left\langle \bar{\mathcal{E}} \cdot \frac{\partial \bar{\mathcal{D}}}{\partial t} \right\rangle = (1/T) \int_0^T dt\, \bar{\mathcal{E}} \cdot \partial \bar{\mathcal{D}}/\partial t \simeq (1/4)\, \mathbf{E}^* (\mathbf{r}, t) \cdot \partial \mathbf{D} (\mathbf{r}, t)/\partial t + c.c.$, where T is the period [7, 11]. Since the medium is taken to be lossless ($\varepsilon^* = \varepsilon$, $\mathbf{k} = \mathbf{k}^*$), it follows from (51) that

$$\begin{aligned}\left\langle \bar{\mathcal{E}} \cdot \partial \bar{\mathcal{D}}/\partial t \right\rangle = & \frac{\varepsilon_0}{4} \frac{\partial (\omega \varepsilon)}{\partial \omega} \frac{\partial}{\partial t} (\mathbf{E}_0 \cdot \mathbf{E}_0^*) - \frac{\varepsilon_0 \omega}{2} \mathrm{Re} \{\mathbf{E}_0^* \cdot (\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}})\, \varepsilon \mathbf{E}_0\} \\ & - \frac{\varepsilon_0 \omega}{4} \mathrm{Im} \left\{\mathbf{E}_0^* \cdot (\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}})^2 \varepsilon \mathbf{E}_0\right\} + \frac{\varepsilon_0 \omega}{12} \mathrm{Re} \left\{\mathbf{E}_0^* \cdot (\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}})^3 \varepsilon \mathbf{E}_0\right\}, \quad (52)\end{aligned}$$

where we replaced the arbitrary frequency ω_0 by ω. In writing the first term in the RHS of (52), the identity $\partial/\partial t\, (\mathbf{E}_0^* \cdot \mathbf{E}_0) = \partial \mathbf{E}_0^*/\partial t \cdot \mathbf{E}_0 + \mathbf{E}_0^* \cdot \partial \mathbf{E}_0/\partial t$ was used. Strictly speaking, if the medium is local, only this term will survive, the rest becoming zero.

In order to obtain meaningful results about the power flow in the medium, we must combine the second, third, and fourth terms in the RHS of (52), which are due to spatial dispersion, with the divergence of the Poynting vector appearing in the LHS of (44). Therefore, we should be able to obtain

$$\nabla_{\mathbf{r}} \cdot \mathbf{S} = -\, (\partial/\partial t)\, (W_e + W_m)\,, \quad (53)$$

where the electric and magnetic energies W_e and W_m are given by $W_e = (\varepsilon_0/4) \partial\, (\omega \varepsilon_0 \varepsilon)/\partial \omega \mathbf{E}_0 \cdot \mathbf{E}_0^*$ and $W_m = (1/4) \partial\, (\omega \mu_0 \mu)/\partial \omega \mathbf{H}_0 \cdot \mathbf{H}_0^*$, respectively. Therefore, it is seen that the problem now is a question of operator algebra in which the goal is to move the operator $\nabla_{\mathbf{r}}$ outside the terms in the RHS of (52) that are contributing to the power flow due to spatial dispersion.

For the second term in the RHS of (52), i.e., the first-order correction of spatial dispersion, this can be easily achieved by the following identity

$$2\mathrm{Re} \{\mathbf{E}_0^* \cdot (\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}})\, \varepsilon \mathbf{E}_0\} = \nabla_{\mathbf{r}} \cdot \{\nabla_{\mathbf{k}} \varepsilon \mathbf{E}_0^* \cdot \mathbf{E}_0\}\,. \quad (54)$$

The third and fourth terms in the RHS of (52), i.e., the second- and third-order corrections, are rather more difficult to obtain. Special identities were derived by the authors and presented in Appendix B and Appendix C for the second- and third-order corrections, respectively. Employing the identities (54), (72), and (80) to simplify (52), plugging the results into the Poynting theorem (44), and after straightforward manipulations, we finally obtain (53) with

$$\mathbf{S} = \mathbf{S}_0 + \mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3, \quad (55)$$

which represents the total power density vector. The conventional Poynting vector contribution is given by $\mathbf{S}_0 = (1/2)\mathrm{Re}\{\mathbf{E}^* \times \mathbf{H}\}$. The first-order correction is given by

$$\mathbf{S}_1 = -\frac{\varepsilon_0}{4}\omega\nabla_{\mathbf{k}}\varepsilon\mathbf{E}_0^* \cdot \mathbf{E}_0. \tag{56}$$

The second-order correction is written as

$$\mathbf{S}_2 = -\frac{\varepsilon_0\omega}{4}\sum_{m=1}^{3}\mathrm{Im}\left\{E_{0m}^*\left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\right)\varepsilon E_{0m}\right\}. \tag{57}$$

The third-order correction is given by

$$\begin{aligned}\mathbf{S}_3 \;=\; & \frac{\varepsilon_0\omega}{24}\sum_{m=1}^{3}E_{0m}^*\left[\nabla_{\mathbf{r}} \cdot \left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\right)\right]\varepsilon E_{0m} \\ & -\frac{\varepsilon_0\omega}{24}\sum_{m=1}^{3}\left(\nabla_{\mathbf{r}}E_{0m}^* \cdot \nabla_{\mathbf{k}}\right)\nabla_{\mathbf{k}}\left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}}\right)\varepsilon E_{0m} \\ & +\frac{\varepsilon_0\omega}{24}\sum_{m=1}^{3}E_{0m}\left[\nabla_{\mathbf{r}} \cdot \left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\right)\right]\varepsilon E_{0m}^*. \end{aligned} \tag{58}$$

We remind the reader that the formulation of the present section is more general than the results related to negative group velocity, which ere presented in the previous sections, for two reasons. First, the final description of the power flow obtained in (55) is valid for any kind of wave propagation, being excited by a source or merely normal (eigenmodes) waves propagating in a source-free medium. Second, in the present section we retained the *vector* dependence of the medium on the wave vector, indicating that the spatial dispersion may introduce preferred directions in the medium response. Thus, the final solution is still able to capture this feature since we don't replace $\mathbf{k}$ by k in expressions like (56), (57), and (58).

On the qualitative level, the first-order vector correction $\mathbf{S}_1$ obviously explains the possibility to obtain negative group velocity although both ϵ and μ are positive. Even though $\mathbf{S}_0$ is directed along $\mathbf{k}$ (because both ϵ and μ are positive [12, 15]), the vector corrections $\mathbf{S}_1$, $\mathbf{S}_2$ and $\mathbf{S}_3$ can reverse the direction of the total vector $\mathbf{S}$, leading to negative refraction.

On the quantitative level, the results of the first-order correction in (56) and those obtained in Sec. 3 are in agreement with each other. To see that, let us consider the case of isotropic non-magnetic material with positive dielectric constant and focus on quasi-monochromatic waves. In this case, it is easy to see form Maxwell's equations, applied to each Fourier mode of the propagating wave packet, that $|E_0| = |\eta H_0|$, where $\eta = \sqrt{(\mu_0\mu)/(\varepsilon_0\varepsilon)}$ [1, 7, 11]. Moreover, it also follows from Maxwell's equations that $\mathbf{S}_0$ is directed along $\mathbf{k}$ and the same conclusion applies to $\mathbf{S}$ since the medium is isotropic. Therefore, by ignoring the contribution of terms higher than the first-order approximation, we can write

$$\mathbf{S}=\mathbf{S}_0+\mathbf{S}_1=\frac{\varepsilon_0 |E_0|^2}{4}\left(2nc-\omega\frac{\partial}{\partial k}n^2\right)\hat{a}_{\mathbf{k}}=\frac{n\varepsilon_0 |E_0|^2}{2}\left(c-\omega\frac{\partial n}{\partial k}\right)\hat{a}_{\mathbf{k}}. \quad (59)$$

where the definition (10) was used. The total energy stored in the medium is given by

$$\begin{aligned} W &= W_e + W_m = \frac{1}{4}\left[\varepsilon_0\frac{\partial(\omega\varepsilon)}{\partial\omega}|\mathbf{E}_0|^2+\mu_0\frac{\partial(\omega\mu)}{\partial\omega}|\mathbf{H}_0|^2\right] \\ &= \frac{|\mathbf{E}_0|^2}{4}\left[\varepsilon_0\frac{\partial(\omega\varepsilon)}{\partial\omega}+\mu_0\frac{\partial(\omega\mu)}{\partial\omega}\frac{\varepsilon\varepsilon_0}{\mu_0}\right]=\frac{\varepsilon_0|\mathbf{E}_0|^2}{4}\left[2\varepsilon+\omega\frac{\partial\varepsilon}{\partial\omega}\right] \\ &= \frac{n\varepsilon_0|\mathbf{E}_0|^2}{2}\left(n+\omega\frac{\partial n}{\partial\omega}\right). \end{aligned} \quad (60)$$

Therefore, using (59), (60), and (23), relation (19) follows immediately. This provides a physical interpretation for the new quantity $-\omega\partial n/\partial k$ in the numerator of (19). This term reflects the contribution of the first-order vector correction of the power flow due to spatial dispersion.

We mention also that it is possible theoretically to have $\nabla_{\mathbf{k}}\varepsilon = 0$ at a frequency and wavelength in which $\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\varepsilon \neq 0$. In this case, the second-order term $\mathbf{S}_2$ becomes dominant and may lead to important corrections even when the overall effect of spatial dispersion is weak. A similar argument applies to the third-order term.

Finally, consider the higher-order corrections in (57) and (58). We can see that $\mathbf{S}_2$ and $\mathbf{S}_3$ depend not only on the spatial dispersion profile, i.e., the functional dependence of ϵ and μ on $\mathbf{k}$, but also on the rate of the spatial variation of the field, the functional dependence of $\mathbf{E}$ and $\mathbf{H}$ on $\mathbf{r}$. For example, if ω and $\mathbf{k}$ are chosen such that $\mathbf{S}_1$ vanishes while $\mathbf{S}_2$ is nonzero (the GV concept fails then to apply), then the derivations above indicate that it is possible to increase the effect of $\mathbf{S}_2$, while choosing its direction to be opposite to $\mathbf{S}_0$, in order to achieve negative group velocity, by tuning the rate of the spatial *field* variation, not just the dielectric and permeability dispersion profiles. In a nutshell, the higher-order contributions of spatial dispersion to the power flow are dependent on the structure of the field itself.

7. SOME GENERAL REMARKS

We end this paper by some general remarks. The ultimate origin of nonlocality is the non-vanishing finite spatial extension of the wavefunctions of the particles constituting the medium under interest [21]. This means that a self-consistent approach, at least in the semi-classical sense, should directly provide expressions for the response functions that include both temporal and spatial dispersion. While many such methods are available in literature, e.g., see [1] and [21], the computational complexity of a realistic problem comprising, say, periodic arrangements of unit cells engineered to achieve desired electromagnetic performance, makes the method very difficult to apply in iterated design procedures. Instead, one may develop a suitable effective-field theory, taking into consideration some of the physical mechanisms that generate nonlocality in the electromagnetic response. Then, this theory, once tested and refined, can be used in an iterative optimization algorithm to achieve the required goals. Moreover, it may be possible to achieve nonlocal effects even within the regime of *classical* electrodynamics by carefully exploiting near-field interactions at the nanoscale [22].

The reader should notice that, as mentioned in Section 5, our design methodology is restricted to certain finite frequency and wavenumber range of interest (i.e., $\omega_1 < \omega < \omega_2$ and $k_1 < k < k_2$.) This limitation relaxes considerably the restrictions imposed by causality, as explicated in Kramers-Kronig relations. For example, the requirement that the medium is lossless in the band of interest can be achieved at the expense of permitting higher losses *outside* this band. In other words, within the $\omega - k$ space, our method is meant to be applied *locally* in order to preserve the consistency with the global restrictions imposed by causality. On the other hand, within the the spatio-temporal space, the conservation of energy is always satisfied locally by the Poyntings theorem in Equation (44).

Some complications in the actual design may arise from the phenomenon of additional waves in nonlocal media. There exist various strategies to deal with this problem [1, 21]. In this case, there could be other modes excited in the structure of interest in addition to the transverse modes studied in this paper. Such waves may affect the performance of the device/medium, for example by carrying part of the energy of the incident beam. Further study of such modes is beyond the scope of the present paper.

Finally, the mathematical results of our paper apply to the simplest type of media, i.e., isotropic and homogenous media. It is expected however, that actual designs, for example using periodic structure, will require relaxing one or both of these conditions. Our work then aims to establish some conceptual understanding of what is new in the physics of nonlocal medium in terms of results that can be derived rigorously from (1) Maxwells equations, (2) energy conservation, and (3) causality. It is still needed to investigate more complicated media in order to demonstrate how the basic theory developed in this paper should be modified.

8. CONCLUSION

A theoretical framework for synthesizing metamaterials exhibiting negative group velocity was proposed. It was found that by carefully exploiting the interplay between temporal and spatial dispersion new phenomena can be observed. An exact solution consisting of a medium profile supporting constant negative group velocity propagation was obtained. The effect of higher-order terms, up to the third-order approximation of the power flow correction, was also derived.

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APPENDIX A. PROOF FOR CASE IV IN SEC. 4

From (24) and (25) we can write

$$\mathbf{v}_g = \frac{n}{n + \omega \partial n/\partial \omega} \mathbf{v}_p. \tag{A1}$$

For negative phase velocity media, we have $n = -\sqrt{\mu\varepsilon}$. Therefore, it is possible to write

$$\begin{aligned} n + \omega \partial n/\partial \omega &= -\sqrt{\varepsilon\mu} + \omega \partial \left(-\sqrt{\varepsilon\mu}\right)/\partial \omega \\ &= -\sqrt{\varepsilon\mu} - \frac{\omega}{2\sqrt{\varepsilon\mu}} \left[\varepsilon \frac{\partial \mu}{\partial \omega} + \mu \frac{\partial \varepsilon}{\partial \omega} \right] \\ &= -\sqrt{\varepsilon\mu} \left\{ 1 + \frac{\omega}{2\varepsilon\mu} \left[\varepsilon \frac{\partial \mu}{\partial \omega} + \mu \frac{\partial \varepsilon}{\partial \omega} \right] \right\}. \end{aligned} \tag{A2}$$

From Kramers-Kronig relations, we know that in a medium with small losses the identities $\partial/\partial\omega\,(\omega\varepsilon) > 0$ and $\partial/\partial\omega\,(\omega\mu) > 0$ hold [10]. This in turn yields $\partial\varepsilon/\partial\omega > -\varepsilon/\omega$ and $\partial\mu/\partial\omega > -\mu/\omega$. Noticing that both

ϵ and μ are negative, the previous two inequalities, when combined together, will give $\mu\partial\varepsilon/\partial\omega + \varepsilon\partial\mu/\partial\omega < -2\varepsilon\mu/\omega$, or

$$1 + \frac{\omega}{2\varepsilon\mu}\left[\varepsilon\frac{\partial\mu}{\partial\omega} + \mu\frac{\partial\varepsilon}{\partial\omega}\right] < 0. \tag{A3}$$

Therefore, (A1), (A2), and (A3) lead to group and phase velocities with signs opposite to each other.

APPENDIX B. DERIVATION OF THE SECOND-ORDER TERM IDENTITY

Our goal is to write the third term in the RHS (52) as a divergence of vector. We expand

$$\mathbf{E}_0^* \cdot (\nabla_\mathbf{r} \cdot \nabla_\mathbf{k})^2 \, \varepsilon \mathbf{E}_0 - c.c. = \sum_{m=1}^{3} E_{0m}^* \, (\nabla_\mathbf{r} \cdot \nabla_\mathbf{k})^2 \, \varepsilon E_{0m} - c.c., \tag{B1}$$

where the index $m = 1, 2, 3$ enumerates three Cartesian directions. We introduce now the following identity

$$E_{0m}^* (\nabla_\mathbf{r} \cdot \nabla_\mathbf{k})^2 \varepsilon E_{0m} - c.c. = \nabla_\mathbf{r} \cdot \{E_{0m}^* \, (\nabla_\mathbf{r} \cdot \nabla_\mathbf{k} \nabla_\mathbf{k}) \varepsilon E_{0m} - c.c.\} \, . \tag{B2}$$

Proof. Using the vector identity

$$\nabla \cdot (\psi \mathbf{A}) = \nabla\psi \cdot \mathbf{A} + \psi \nabla \cdot \mathbf{A}, \tag{B3}$$

we find

$$\begin{aligned} & \nabla_\mathbf{r} \cdot \{E_{0m}^* \, (\nabla_\mathbf{r} \cdot \nabla_\mathbf{k} \nabla_\mathbf{k}) \, \varepsilon E_{0m} - c.c.\} \\ = \; & \nabla_\mathbf{r} E_{0m}^* \cdot (\nabla_\mathbf{r} \cdot \nabla_\mathbf{k} \nabla_\mathbf{k}) \, \varepsilon E_{0m} + E_{0m}^* \, [\nabla_\mathbf{r} \cdot (\nabla_\mathbf{r} \cdot \nabla_\mathbf{k} \nabla_\mathbf{k})] \, \varepsilon E_{0m} \\ & - \nabla_\mathbf{r} E_{0m} \cdot (\nabla_\mathbf{r} \cdot \nabla_\mathbf{k} \nabla_\mathbf{k}) \, \varepsilon E_{0m}^* - E_{0m} \, [\nabla_\mathbf{r} \cdot (\nabla_\mathbf{r} \cdot \nabla_\mathbf{k} \nabla_\mathbf{k})] \, \varepsilon E_{0m}^*. \end{aligned} \tag{B4}$$

By applying the dyadic identities

$$\mathbf{ab} \cdot \mathbf{c} = \mathbf{a} \, (\mathbf{b} \cdot \mathbf{c}) \, , \quad \mathbf{c} \cdot \mathbf{ab} = (\mathbf{c} \cdot \mathbf{a}) \, \mathbf{b}, \tag{B5}$$

$$\mathbf{A} \cdot \left(\bar{\mathbf{C}} \cdot \mathbf{B}\right) = \left(\mathbf{A} \cdot \bar{\mathbf{C}}\right) \cdot \mathbf{B}, \tag{B6}$$

we find

$$\begin{aligned} & \nabla_\mathbf{r} E_{0m}^* \cdot (\nabla_\mathbf{r} \cdot \nabla_\mathbf{k} \nabla_\mathbf{k}) \, \varepsilon E_{0m} \\ & = \nabla_\mathbf{r} E_{0m}^* \cdot (\nabla_\mathbf{k} \nabla_\mathbf{k} \cdot \nabla_\mathbf{r}) \, \varepsilon E_{0m} = (\nabla_\mathbf{r} E_{0m}^* \cdot \nabla_\mathbf{k} \nabla_\mathbf{k} \varepsilon) \cdot \nabla_\mathbf{r} E_{0m} \\ & = \nabla_\mathbf{r} E_{0m} \cdot (\nabla_\mathbf{r} E_{0m}^* \cdot \nabla_\mathbf{k} \nabla_\mathbf{k} \varepsilon) = \nabla_\mathbf{r} E_{0m} \cdot ((\nabla_\mathbf{r} E_{0m}^* \cdot \nabla_\mathbf{k}) \, \nabla_\mathbf{k} \varepsilon) \\ & = \nabla_\mathbf{r} E_{0m} \cdot ((\nabla_\mathbf{r} \cdot \nabla_\mathbf{k}) \, \nabla_\mathbf{k} \varepsilon E_{0m}^*) = \nabla_\mathbf{r} E_{0m} \cdot (\nabla_\mathbf{r} \cdot \nabla_\mathbf{k} \nabla_\mathbf{k}) \, \varepsilon E_{0m}^*. \end{aligned} \tag{B7}$$

Next, using (B5) we find

$$\begin{aligned}\left(\nabla_{\mathbf{k}}\cdot\nabla_{\mathbf{r}}\right)^2 &= \left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\right)\left(\nabla_{\mathbf{k}}\cdot\nabla_{\mathbf{r}}\right)=\nabla_{\mathbf{r}}\cdot\left(\nabla_{\mathbf{k}}\left(\nabla_{\mathbf{k}}\cdot\nabla_{\mathbf{r}}\right)\right)\\ &= \nabla_{\mathbf{r}}\cdot\left(\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\cdot\nabla_{\mathbf{r}}\right)=\nabla_{\mathbf{r}}\cdot\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\right).\end{aligned}\tag{B8}$$

Finally, be employing (B7) and (B8) in (B4), the required identity (B2) is obtained.

By using the identity (B2) in (B1), we obtain

$$-\left(\varepsilon_0\omega/4\right)\operatorname{Im}\left\{\mathbf{E}_0^*\cdot\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\right)^2\varepsilon\mathbf{E}_0\right\}=\nabla_{\mathbf{r}}\cdot\mathbf{S}_2,\tag{B9}$$

where where $\mathbf{S}_2$ is given by (57).

APPENDIX C. DERIVATION OF THE THIRD-ORDER TERM IDENTITY

Our goal is to write the fourth term in the RHS (52) as a divergence of vector. We expand

$$\mathbf{E}_0^*\cdot\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\right)^3\varepsilon\mathbf{E}_0+c.c.=\sum_{m=1}^{3}\left\{E_{0m}^*\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\right)^3\varepsilon E_{0m}+c.c.\right\}.\tag{C1}$$

We state now the following identity

$$\begin{aligned}&E_{0m}^*\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\right)^3\varepsilon E_{0m}+E_{0m}\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\right)^3\varepsilon E_{0m}^*\\ =\;&\nabla_{\mathbf{r}}\cdot\left\{E_{0m}^*\left[\nabla_{\mathbf{r}}\cdot\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\right)\right]\varepsilon E_{0m}\right.\\ &-\left(\nabla_{\mathbf{r}}E_{0m}^*\cdot\nabla_{\mathbf{k}}\right)\nabla_{\mathbf{k}}\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\right)\varepsilon E_{0m}\\ &\left.+E_{0m}\left[\nabla_{\mathbf{r}}\cdot\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\right)\right]\varepsilon E_{0m}^*\right\}.\end{aligned}\tag{C2}$$

Proof. By applying the identity (B3), the RHS of (C2) can be written as

$$\begin{aligned}\text{RHS} =\;& \nabla_{\mathbf{r}}E_{0m}^*\cdot\left[\nabla_{\mathbf{r}}\cdot\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\right)\right]\varepsilon E_{0m}\\ &+E_{0m}^*\nabla_{\mathbf{r}}\cdot\left[\nabla_{\mathbf{r}}\cdot\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\right)\right]\varepsilon E_{0m}\\ &-\nabla_{\mathbf{r}}\left(\nabla_{\mathbf{r}}E_{0m}^*\cdot\nabla_{\mathbf{k}}\right)\cdot\left[\nabla_{\mathbf{k}}\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\right)\varepsilon E_{0m}\right]\\ &-\left(\nabla_{\mathbf{r}}E_{0m}^*\cdot\nabla_{\mathbf{k}}\right)\nabla_{\mathbf{r}}\cdot\left[\nabla_{\mathbf{k}}\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\right)\varepsilon E_{0m}\right]\\ &+\nabla_{\mathbf{r}}E_{0m}\cdot\left[\nabla_{\mathbf{r}}\cdot\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\right)\right]\varepsilon E_{0m}^*\\ &+E_{0m}\nabla_{\mathbf{r}}\cdot\left[\nabla_{\mathbf{r}}\cdot\left(\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\nabla_{\mathbf{k}}\right)\right]\varepsilon E_{0m}^*.\end{aligned}\tag{C3}$$

By applying the triadic identities

$$\mathbf{abc}\cdot\mathbf{d}=\mathbf{ab}\left(\mathbf{c}\cdot\mathbf{d}\right),\quad \mathbf{d}\cdot\mathbf{abc}=\left(\mathbf{d}\cdot\mathbf{a}\right)\mathbf{bc},\tag{C4}$$

together with (B6), the third term in (C3) can be manipulated as

$$\begin{aligned} & \nabla_{\mathbf{r}} \left(\nabla_{\mathbf{r}} E_{0m}^{*} \cdot \nabla_{\mathbf{k}} \right) \cdot \left[\nabla_{\mathbf{k}} \left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}} \right) \varepsilon E_{0m} \right] \\ = \ & \nabla_{\mathbf{r}} \left(\nabla_{\mathbf{r}} E_{0m}^{*} \cdot \nabla_{\mathbf{k}} \right) \cdot \left[\left(\nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \cdot \nabla_{\mathbf{r}} \right) \varepsilon E_{0m} \right] \\ = \ & \left[\nabla_{\mathbf{r}} \left(\nabla_{\mathbf{r}} E_{0m}^{*} \cdot \nabla_{\mathbf{k}} \right) \cdot \nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \varepsilon \right] \cdot \nabla_{\mathbf{r}} E_{0m} \\ = \ & \nabla_{\mathbf{r}} E_{0m} \cdot \left[\nabla_{\mathbf{r}} \left(\nabla_{\mathbf{r}} E_{0m}^{*} \cdot \nabla_{\mathbf{k}} \right) \cdot \nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \varepsilon \right] \\ = \ & \nabla_{\mathbf{r}} E_{0m} \cdot \left[\nabla_{\mathbf{r}} \left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}} \right) \cdot \nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \varepsilon E_{0m}^{*} \right] \\ = \ & \nabla_{\mathbf{r}} E_{0m} \cdot \left[\nabla_{\mathbf{r}} \cdot \left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \right) \varepsilon E_{0m}^{*} \right]. \end{aligned} \tag{C5}$$

Also, the fourth term in (C3) can be put in the form

$$\begin{aligned} & \left(\nabla_{\mathbf{r}} E_{0m}^{*} \cdot \nabla_{\mathbf{k}} \right) \nabla_{\mathbf{r}} \cdot \left[\nabla_{\mathbf{k}} \left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}} \right) \right] \varepsilon E_{0m} \\ = \ & \left(\nabla_{\mathbf{r}} E_{0m}^{*} \cdot \nabla_{\mathbf{k}} \right) \left[\nabla_{\mathbf{r}} \cdot \left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \right) \right] \varepsilon E_{0m} \\ = \ & \left(\nabla_{\mathbf{r}} E_{0m}^{*} \cdot \nabla_{\mathbf{k}} \right) \left[\left(\nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \cdot \nabla_{\mathbf{r}} \right) \cdot \nabla_{\mathbf{r}} \right] \varepsilon E_{0m} \\ = \ & \nabla_{\mathbf{r}} E_{0m}^{*} \cdot \left[\left(\nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \cdot \nabla_{\mathbf{r}} \right) \cdot \nabla_{\mathbf{r}} \right] \varepsilon E_{0m} \\ = \ & \nabla_{\mathbf{r}} E_{0m}^{*} \cdot \left[\nabla_{\mathbf{r}} \cdot \left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \right) \right] \varepsilon E_{0m}. \end{aligned} \tag{C6}$$

Next, using the identities (C4), we find

$$\begin{aligned} \left(\nabla_{\mathbf{k}} \cdot \nabla_{\mathbf{r}} \right)^3 &= \left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}} \right) \left(\nabla_{\mathbf{k}} \cdot \nabla_{\mathbf{r}} \right) \left(\nabla_{\mathbf{k}} \cdot \nabla_{\mathbf{r}} \right) \\ &= \nabla_{\mathbf{r}} \cdot \left\{ \nabla_{\mathbf{k}} \left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}} \right) \left(\nabla_{\mathbf{k}} \cdot \nabla_{\mathbf{r}} \right) \right\} = \nabla_{\mathbf{r}} \cdot \left\{ \nabla_{\mathbf{k}} \left[\nabla_{\mathbf{r}} \cdot \left(\nabla_{\mathbf{k}} \left(\nabla_{\mathbf{k}} \cdot \nabla_{\mathbf{r}} \right) \right) \right] \right\} \\ &= \nabla_{\mathbf{r}} \cdot \left\{ \nabla_{\mathbf{k}} \left[\nabla_{\mathbf{r}} \cdot \left(\nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \cdot \nabla_{\mathbf{r}} \right) \right] \right\} = \nabla_{\mathbf{r}} \cdot \left\{ \left[\nabla_{\mathbf{k}} \left(\nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \cdot \nabla_{\mathbf{r}} \right) \right] \cdot \nabla_{\mathbf{r}} \right\} \\ &= \nabla_{\mathbf{r}} \cdot \left\{ \left[\nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \cdot \nabla_{\mathbf{r}} \right] \cdot \nabla_{\mathbf{r}} \right\} = \nabla_{\mathbf{r}} \cdot \left\{ \nabla_{\mathbf{r}} \cdot \left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \nabla_{\mathbf{k}} \right) \right\}. \end{aligned} \tag{C7}$$

Finally, by substituting (C5), (C6), and (C7) into (C3), the identity (C2) is obtained.

Applying the identity (C2) to (C1), we obtain the desired relation

$$\left(\varepsilon_0 \omega / 12 \right) \mathrm{Re} \left\{ \mathbf{E}_0^{*} \cdot \left(\nabla_{\mathbf{r}} \cdot \nabla_{\mathbf{k}} \right)^3 \varepsilon \mathbf{E}_0 \right\} = \nabla_{\mathbf{r}} \cdot \mathbf{S}_3, \tag{C8}$$

where $\mathbf{S}_3$ is given by (58).

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