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ENERGY, MOMENTUM, AND ANGULAR MOMENTUM OF ELECTROMAGNETIC FIELD IN A MEDIUM WITH NONLOCAL OPTICAL RESPONSE UNDER FREQUENCY-DEGENERATE NONLINEAR WAVE INTERACTION

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Abstract. The expressions for the additional terms to the electromagnetic field energy density, energy flux density, momentum density, momentum flux density, components of angular momentum density and components of anglular momentum flux density tensor in a medium with nonlocality of the *n*-th order nonlinear optical response are obtained from the Maxwell equations system for the case when the number of the interacting waves with different frequencies is less than or equal to n (frequency-degenerate processes). It is shown that the intrinsic symmetry relations between the components of both local and nonlocal nonlinear susceptibility tensors make it impossible to obtain the correct formulas for the aforementioned fundamental characteristics of the electromagnetic field as a particular case of the already known expressions for these quantities related to the nonlinear interaction of $n + 1$ waves with absolutely different frequencies if we put some frequencies equal to each other. As an example, we discuss the obtained additional terms caused by nonlocal nonlinear optical response of the medium in cases of self-focusing, second- and third-harmonic generation.

Keywords: *nonlinear optics*, *spatial dispersion*, *optical angular momentum*

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1. INTRODUCTION

The energy, momentum and angular momentum (moment of momentum) of the electromagnetic field are its most important characteristics, the analysis of which is of considerable interest both from the point of view of the electromagnetism theory and for practical applications. Being fundamental physical quantities, they obey conservation laws, which are written in the form of balance equations linking the densities of these quantities and the densities of their fluxes [1-5]. Their form depends on the spatial symmetry of the medium in which the electromagnetic field exists. In absorbing, inhomogeneous and anisotropic media [1, 3, 6-8], these equations are inhomogeneous. Other important properties of the medium, such as the frequency and spatial dispersion of the optical response and its nonlinear dependence on the external electric field strength, do not lead to inhomogeneity in the conservation laws, but can fundamentally change the formulas for the energy, momentum and angular momentum densities, as well as for their flux densities. A certain tendency can be noted: taking into account the frequency dispersion of the optical response leads, as a rule, to changes in the expressions only for the energy density, momentum density, and angular momentum density of light in the medium, without leading to significant changes in the expressions for the corresponding energy flux density, momentum flux density, and momentum flux density, while the spatial dispersion of the optical response of the medium, on the contrary, requires changes in the definitions of the energy, momentum, and angular momentum flux densities, leaving the coefficients unchanged. In turn, the nonlinearity of the optical response affects all of the above-mentioned quantities because they depend on the polarization of the medium in one way or another [12-14].

In addition to fundamental aspects of electrodynamics, the study of the influence of the optical response of the medium on the energy, momentum and angular momentum of propagating radiation is of great interest for practical applications. Thus, the energy flux density determines the intensity of radiation, which is the main characteristic used in light detection [15]. Momentum and moment of momentum determine the magnitude of mechanical effect of light on the medium [1, 16]. In addition, the angular momentum of light also has great practical potential in the tasks of information transfer, control and manipulation of microparticles and in the study of the structure of matter [17-26].

The formulas for energy, momentum and angular momentum of laser radiation propagating in media possessing nonlocal nonlinear optical response are of special interest due to the study of the peculiarities of interaction of elliptically polarized waves in them. In [27], analytical expressions for the energy density, energy flux density, momentum density, and momentum flux density of light were derived in the case when among the frequencies $n+1$ of elliptically polarized interacting waves in a medium exhibiting a nonlocal nonlinear optical response of n-th order, there are no equal to each other. For such waves in the above-mentioned media, expressions for the angular momentum component and the components of the angular momentum flux density tensor of electromagnetic field have also been derived [28]. At the same time, many of the most common processes of nonlinear optics, such as, for example, generation of second and third harmonics, self-action, and the spectroscopic scheme of Coherent anti-Stokes Raman spectroscopy (CARS), are degenerate in frequency, i.e., among the frequencies of waves interacting in a nonlinear medium there are those equal to each other. The initial opinion that the presence of degeneracy of frequencies simplifies the formulas for energy density, energy flux density, momentum density, momentum flux density, angular momentum component, and the components of the angular momentum flux density tensor and they are all easily obtained as a special case of equality of separate frequencies in the previously obtained expressions of [27, 28], turned out to be erroneous. The case of degeneracy of one or several frequencies turns out to be in fact more general and requires more complicated transformations for deriving formulas for the above-mentioned characteristics of the electromagnetic field than the limiting situation when the degeneracy coefficient of each of the frequencies of interacting waves is equal to unity. In particular, this is due to the fact that in the degenerate case the tensors describing the local and nonlocal nonlinear optical response of the medium have greater symmetry than in the nongenerated case [29, 30]. The purpose of this work is to find analytical expressions for the additions to the energy density, energy flux density, momentum density, momentum flux density, angular momentum density components, and angular momentum flux density tensor components due to the local and nonlocal nonlinear optical response of the n-th order of volume of a homogeneous nonabsorbing medium in the case when the number of interacting waves with different frequencies in it is less than or equal to n. The latter is equivalent to the fact that there are waves with the same frequencies among the $n+1$ waves formally interacting in the medium with n-order nonlinearity.

2. POLARIZATION OF MEDIUM AT FREQUENCY DEGENERATE NONLINEAR WAVE INTERACTION IN MEDIUM WITH NONLOCALITY OF OPTICAL RESPONSE

Let in a medium exhibiting nonlinearity of n-th order, of $n+1$ frequencies of the waves involved in the interaction, the first $m-1$ frequencies are different, the next $n-m+1$ frequencies are equal to ω_m, and the last

$$\omega_{n+1} = \sum_{l=1}^{m-1}\omega_l + (n-m+1)\,\omega_m.$$

The electric field strength created by them is equal to

$$\mathbf{E}(\mathbf{r},t) = \sum_{l=1}^{m}\tilde{\mathbf{E}}^{(l)}(\mathbf{r},t,\omega_l)\exp(-i\omega_l t) + \tilde{\mathbf{E}}^{(n+1)}(\mathbf{r},t,\omega_{n+1})\exp(-i\omega_{n+1}t) + \text{c.c.} = \sum_{l=1}^{m}\mathbf{E}^{(l)} + \mathbf{E}^{(n+1)} + \text{c.c.}, \quad (1)$$

where $\tilde{\mathbf{E}}^{(l)}(\mathbf{r},t,\omega_l)$ is the complex amplitude of the wave with frequency ω_l. The magnetic field induction $\mathbf{B}(\mathbf{r},t)$ (which we will consider equal to the magnetic field strength $\mathbf{H}(\mathbf{r},t)$) and the electric field induction $\mathbf{D}(\mathbf{r},t)$, similar to (2), are expressed through the complex amplitudes $\tilde{\mathbf{B}}^{(l)}(\mathbf{r},t,\omega_l)$ and $\tilde{\mathbf{D}}^{(l)}(\mathbf{r},t,\omega_l)$:

$$\begin{aligned}\mathbf{B}(\mathbf{r},t) &= \sum_{l=1}^{m}\tilde{\mathbf{B}}^{(l)}(\mathbf{r},t,\omega_l)\exp(-i\omega_l t) + \tilde{\mathbf{B}}^{(n+1)}(\mathbf{r},t,\omega_{n+1})\exp(-i\omega_{n+1}t) + \text{c.c.} = \\ &= \sum_{l=1}^{m}\mathbf{B}^{(l)} + \mathbf{B}^{(n+1)} + \text{c.c.},\end{aligned} \tag{2}$$

$$\begin{aligned}\mathbf{D}(\mathbf{r},t) &= \sum_{l=1}^{m}\tilde{\mathbf{D}}^{(l)}(\mathbf{r},t,\omega_l)\exp(-i\omega_l t) + \tilde{\mathbf{D}}^{(n+1)}(\mathbf{r},t,\omega_{n+1})\exp(-i\omega_{n+1}t) + \text{c.c.} = \\ &= \sum_{l=1}^{m}\mathbf{D}^{(l)} + \mathbf{D}^{(n+1)} + \text{c.c.}\end{aligned} \tag{3}$$

In a medium exhibiting optical nonlinearity of n-th order

$$\mathbf{D} = \mathbf{D}_L + \mathbf{P},$$

where

$$\mathbf{P} = \mathbf{P}^{loc} + \mathbf{P}^{nloc}.$$

Here $\mathbf{D}_L$ is a linearly depending on $\mathbf{E}$ part of the electric induction vector. The local $\mathbf{P}^{loc}$ and nonlocal $\mathbf{P}^{nloc}$ parts of the nonlinear polarization of the medium are also written in a form similar to $\mathbf{E}$:

$$\begin{aligned}\mathbf{P}^{loc,nloc}(\mathbf{r},t) &= \sum_{l=1}^{m}\tilde{\mathbf{P}}^{loc,nloc}(\mathbf{r},t,\omega_l)\exp(-i\omega_l t) + \tilde{\mathbf{P}}^{loc,nloc}(\mathbf{r},t,\omega_{n+1})\exp(-i\omega_{n+1}t) + \text{c.c.} = \\ &= \sum_{l=1}^{m}\mathbf{P}^{loc,nloc}(\omega_l) + \mathbf{P}^{loc,nloc}(\omega_{n+1}) + \text{c.c.}\end{aligned} \tag{4}$$

The constitutive equations for $\mathbf{P}^{loc,nloc}(\omega_l)$, where $l = 1,2,\ldots,m,n+1$, in a medium exhibiting n-order nonlinearity can be written as [30-32]

$$\begin{aligned}P_i^{loc}(\omega_l) &= \chi^{(n)}_{ii_{n+1}i_1^{l-1}i_{l+1}^{n}}\left(\omega_l;\omega_{n+1},-\bar{\omega}_1^{l-1},-\bar{\omega}_{l+1}^{m-1},-\tilde{\omega}_m^{(F(\omega_m))}\right)\times \\ &\times E_{i_{n+1}}^{(n+1)}\prod_{\substack{p=1\\ p\neq 1}}^{m-1}E_{i_p}^{(p)*}\prod_{p=m}^{n}E_{i_p}^{(m)*},\end{aligned} \tag{5}$$

if $l = 1,2,\ldots,m-1$,

$$P_i^{loc}(\omega_m) = \chi^{(n)}_{ii_{n+1}i_1^{m-1}i_{m+1}^{n}}\left(\omega_m;\omega_{n+1},-\bar{\omega}_1^{m-1},-\tilde{\omega}_m^{(F(\omega_m)-1)}\right)E_{i_{n+1}}^{(n+1)}\prod_{p=1}^{m-1}E_{i_p}^{(p)*}\prod_{p=m+1}^{n}E_{i_p}^{(m)*}, \tag{6}$$

$$P_i^{loc}(\omega_{n+1}) = \chi^{(n)}_{ii_1^n}\left(\omega_{n+1};\bar{\omega}_1^{m-1},\tilde{\omega}_m^{(F(\omega_m))}\right)\prod_{p=1}^{m-1}E_{i_p}^{(p)}\prod_{p=m}^{n}E_{i_p}^{(m)}, \tag{7}$$

$$P_i^{nloc}(\omega_l) = \Gamma_{ii_{n+1}k}(\omega_l;\omega_{n+1})\partial_k E_{i_{n+1}}^{(n+1)} + \sum_{\substack{s=1\\ s\neq 1}}^{m-1} \Gamma_{ii_sk}(\omega_l;-\omega_s)\partial_k E_{i_s}^{(s)*} + \Gamma_{ii_nk}(\omega_l;-\omega_m)\partial_k E_{i_n}^{(m)*}, \tag{8}$$

if $l = 1,2,\ldots,m-1$,

$$P_i^{nloc}(\omega_m) = \Gamma_{ii_{n+1}k}(\omega_m;\omega_{n+1})\partial_k E_{i_{n+1}}^{(n+1)} + \sum_{s=1}^{m-1} \Gamma_{ii_sk}(\omega_m;-\omega_s)\partial_k E_{i_s}^{(s)*} \tag{9}$$

and

$$P_i^{nloc}(\omega_{n+1}) = \sum_{s=1}^{m-1} \Gamma_{ii_sk}(\omega_{n+1};\omega_s)\partial_k E_{i_s}^{(s)} + \Gamma_{ii_nk}(\omega_{n+1};\omega_m)\partial_k E_{i_n}^{(m)}. \tag{10}$$

Here, the indices $i, i_1, i_2, \ldots i_{n+1}$, and k take the values x, y, and z ; the indices occurring twice are summed; the tensors $\hat{\chi}^{(n)}$ and $\hat{\gamma}^{(n)}$ of rank $n+1$ and $n+2$, respectively, determine the contributions of the local and non-local nonlinear response of n-th order to the polarization of the medium; i_s^q where $1 \leqslant s < q \leqslant n+1$, denotes the index sequence $i_s, i_{s+1}, \ldots, i_{q-1}, i_q$; and $\pm\bar{\omega}_s^q$ denotes, respectively, the frequency sequences $\omega_s, \omega_{s+1}, \ldots, \omega_{q-1}, \omega_q$; and $-\omega_s, -\omega_{s+1}, \ldots, -\omega_{q-1}, -\omega_q$; the set of s identical freqquencies ω_m is denoted in these formulas as $\tilde{\omega}_m^s$. In (5)-(7) and hereafter, $F(\omega_m)$ is the multiplicity of degeneracy of the frequency ω_m, defined as the number of occurences of ω_m after the semicolon in the arguments of the tensor component $\hat{\chi}$ or $\hat{\gamma}$, and increased by one if the frequency $-\omega_m$ is the first argument of the components of these tensors. For the convenience of writing down (8)-(10), we introduced auxiliary tensors

$$\begin{gathered}\Gamma_{ii_sk}(\omega_{n+1};\omega_s) = \partial P_i^{nloc}(\omega_{n+1}) / \partial\left(\partial_k E_{i_s}^{(s)}\right) = \\ = \gamma^{(n)}_{ii_1^{s-1}i_{s+1}^n i_s k}\left(\omega_{n+1};\bar{\omega}_1^{s-1},\bar{\omega}_{s+1}^m,\tilde{\omega}_m^{(F(\omega_m)-1)},\omega_s\right)\prod_{\substack{p=1\\ p\neq s}}^{m} E_{i_p}^{(p)} \prod_{p=m+1}^{n} E_{i_p}^{(m)},\end{gathered} \tag{11}$$

$$\begin{gathered}\Gamma_{ii_{n+1}k}(\omega_l;\omega_{n+1}) = \partial P_i^{nloc}(\omega_l) / \partial(\partial_k E_{i_{n+1}}^{(n+1)}) = \\ = \gamma^{(n)}_{ii_1^{l-1}i_{l+1}^{n+1}k}\left(\omega_l;-\bar{\omega}_1^{l-1},-\bar{\omega}_{l+1}^m,-\tilde{\omega}_m^{(F(\omega_m)-1)},\omega_{n+1}\right)\prod_{\substack{p=1\\ p\neq l}}^{m} E_{i_p}^{(p)*} \prod_{p=m+1}^{n} E_{i_p}^{(m)*},\end{gathered} \tag{12}$$

$$\begin{gathered}\Gamma_{ii_sk}(\omega_l;-\omega_s) = \partial P_i^{nloc}(\omega_l) / \partial\left(\partial_k E_{i_s}^{(s)*}\right) = \\ = \gamma^{(n)}_{ii_{n+1}i_1^{\min(l,s)-1}i_{\min(l,s)+1}^{\max(l,s)-1}i_{\max(l,s)+1}^{n}i_s k} \\ \times\left(\omega_l;\omega_{n+1},-\bar{\omega}_1^{\min(l,s)-1},-\bar{\omega}_{\min(l,s)+1}^{\max(l,s)-1},-\bar{\omega}_{\max(l,s)+1}^{m},-\tilde{\omega}_m^{(F(\omega_m)-1)},-\omega_s\right)\times \\ \times E_{i_{n+1}}^{(n+1)}\prod_{\substack{p=1\\ p\neq l,s}}^{m} E_{i_p}^{(p)*} \prod_{p=m+1}^{n} E_{i_p}^{(m)*},\end{gathered} \tag{13}$$

allowing to shorten the notation of $\mathbf{P}^{loc}(\omega_l)$ and $\mathbf{P}^{nloc}(\omega_l)$, as well as the following equations. If the upper index at i_s^q or $\overline{\omega}_s^q$ is less than the lower index, the corresponding sets are empty, and the associated products of the fields in $\mathbf{P}^{loc}(\omega_l)$,$\mathbf{P}^{nloc}(\omega_l)$, $\Gamma_{ii_sk}(\omega_{n+1};\omega_s)$, $\Gamma_{ii_{n+1}k}(\omega_l;\omega_{n+1})$, and $\Gamma_{ii_sk}(\omega_l;-\omega_s)$ are considered equal to one. The components of the tensors (11) and (13) satisfy the condition

$$\Gamma_{ii_{n+1}k}\left(\omega_l;\omega_{n+1}\right) = -\Gamma_{ii_sk}\left(-\omega_{n+1};-\omega_s\right),$$

following from the intrinsic symmetry relations of the tensor $\hat{\gamma}^{(n)}$ [30]. After substituting the explicit forms of i_s^q, $\overline{\omega}_s^q$ and $\tilde{\omega}_m^s$ into (5)-(7), the resulting expressions for $P_i^{loc}(\omega_l)$, $P_i^{loc}(\omega_m)$, and $P_i^{loc}(\omega_{n+1})$ coincide with a special case of the formulas written out in [33], and after substituting (11)-(13) into (8)-(10) and writing in them the explicit forms of i_s^q, $\overline{\omega}_s^q$, $\tilde{\omega}_m^s$, the resulting expressions for $P_i^{nloc}(\omega_l)$, $P_i^{nloc}(\omega_m)$, and $P_i^{nloc}(\omega_{n+1})$ coincide with those given in [30]. In the constitutive equations (5)-(10), the frequency ω_m has degeneracy multiplicity $F(\omega_m) = n - m + 1$, and all other frequencies have degeneracy multiplicity 1.

The absence in these formulas of several different sets of identical frequencies is due solely to the purpose of making the cumbersome formulas used shorter. All further formulas obtained in this approximation can be easily generalized to the cases of several degenerate frequencies.

3. ENERGY AND MOMENTUM OF LIGHT AT FREQUENCY DEGENERATE NONLINEAR WAVE INTERACTION

To obtain additions to the energy density

$$U^{(n)} = U^{(n,loc)} + U^{(n,nloc)}$$

to the field energy flux density vector

$$\mathbf{S}^{(n)} = \mathbf{S}^{(n,loc)} + \mathbf{S}^{(n,nloc)}$$

(also known as the Poynting vector), the momentum density component

$$g_i^{(n)} = g_i^{(n,loc)} + g_i^{(n,nloc)}$$

and the momentum flux density component

$$G_{ij}^{(n)} = G_{ij}^{(n,loc)} + G_{ij}^{(n,nloc)},$$

related to the local and nonlocal nonlinear optical response of n-th order volume of a homogeneous non-absorbing medium, it is necessary to substitute expressions (5)-(10) into the formula for $\mathbf{D}(\mathbf{r},t)$, similar to [27]. The obtained result, as well as $\mathbf{E}(\mathbf{r},t)$, $\mathbf{B}(\mathbf{r},t)$, should be further substituted into the following of Maxwell equations of energy conservation and momentum conservation laws [1-3]:

$$\frac{1}{c}\left(\mathbf{E}\cdot\partial_t\mathbf{D} + \mathbf{B}\cdot\partial_t\mathbf{B}\right) + \operatorname{div}\left[\mathbf{E}\times\mathbf{B}\right] = 0, \quad (14)$$

$$\frac{1}{c}\partial_t\left[\mathbf{D}\times\mathbf{B}\right] + \mathbf{D}\times\nabla\times\mathbf{E} + \mathbf{B}\times\nabla\times\mathbf{B} = 0, \quad (15)$$

and average the resulting expressions over time. As a result, only the derivatives of slowly changing quantities will remain in the equations. Then the obtained expressions should be transformed so that they take the form of continuity equations:

$$\frac{1}{c}\partial_t U + \operatorname{div}\mathbf{S} = 0, \quad (16)$$

$$\frac{1}{c}\partial_t g_i + \partial_j G_{ij} = 0, \tag{17}$$

relating the energy density U to the energy flux density $\mathbf{S}$ and the momentum density g_i to the momentum flux density G_{ij}. Since $\mathbf{D}$ additively includes the summands related to the linear and nonlinear components of the polarization of the medium, respectively, and each of them consists of the sum of local and nonlocal components, the entries in (16), (17) U, $\mathbf{S}$, g_i, and G_{ij} can be written in the form of

$$U = \sum_{n=1}^{\infty}\left(U^{(n,loc)} + U^{(n,nloc)}\right), \tag{18}$$

$$\mathbf{S} = \sum_{n=1}^{\infty}\left(\mathbf{S}^{(n,loc)} + S^{(n,nloc)}\right), \tag{19}$$

$$g_i = \sum_{n=1}^{\infty}\left(g_i^{(n,loc)} + g_i^{(n,nloc)}\right), \tag{20}$$

$$G_{ij} = \sum_{n=1}^{\infty}\left(G_{ij}^{(n,loc)} + G_{ij}^{(n,nloc)}\right). \tag{21}$$

To find the explicit form of the summands in the right-hand sides of equations (18)-(21), it is necessary to transform the expressions $\langle \mathbf{E}\cdot\partial_t\mathbf{P}\rangle = \partial_t\langle \mathbf{E}\cdot\mathbf{P}\rangle - \langle \mathbf{P}\cdot\partial_t\mathbf{E}\rangle$ and $\langle \mathbf{P}\cdot\partial_p\mathbf{E}\rangle$, where the angle brackets denote time averaging, and p takes the values x, y, and z. Unfortunately, this procedure turns out to be fundamentally dependent on the number of summands in the constitutive equations and, therefore, the results of [27], where m was equal to n, i.e., the frequencies of all $n+1$ interacting waves were different, cannot be directly used to find them.

Substituting constitutive equations (5)-(7) for the local nonlinear polarization into $\langle \mathbf{P}\cdot\partial_p\mathbf{E}\rangle$ and considering the property of permutation symmetry of the tensor $\hat{\chi}^{(n)}$, according to which [29]

$$\begin{gathered}\frac{1}{F(-\omega_m)}\chi^{(n)}_{i i_1^{m-1} i_{m+1}^{n} i_{n+1}}\left(\omega_m; -\bar{\omega}_1^{m-1}, -\tilde{\omega}_m^{(F(\omega_m)-1)}, \omega_{n+1}\right) = \\ = \frac{1}{F(\omega_{n+1})}\chi^{(n)}_{i_{n+1} i_1^{m-1} i_{m+1}^{n} i}\left(-\omega_{n+1}; -\bar{\omega}_1^{m-1}, -\tilde{\omega}_m^{(F(\omega_m))}\right),\end{gathered} \tag{22}$$

it is possible to check that for any of the set of frequencies $\omega_{1,2,\ldots,m,n+1}$ of interacting waves the equality is true

$$\begin{gathered}\sum_{s=1}^{m} P_{i_s}^{loc}(\omega_s)\,\partial_p E_{i_s}^{(s)*} + P_{i_{n+1}}^{loc}(\omega_{n+1})\,\partial_p E_{i_{n+1}}^{(n+1)*} + \text{c.c.} = \\ = \frac{1}{F(\omega_l)}\partial_p\left[P_{i_l}^{loc}(\omega_l)\,E_{i_l}^{(l)*}\right] + \text{c.c.}\end{gathered} \tag{23}$$

For this purpose we need to substitute formulas (5)-(7) into the left part of formula (23), then in the summand containing the derivative of the electric field strength at the frequency ω_m, to bring, applying the rules of differentiation, under the derivative of all fields at this frequency, and then, using the internal symmetry relations (22), to transform in all the resulting summands of the left part of formula (23) the components of the tensor $\hat{\chi}^{(n)}$ so that they all have the same sequences of indices and frequency arguments. If, for example, ω_l is chosen as the first frequency, the resulting expression will coincide with the right part of formula (23). The transformation of the latter and, as a consequence, finding the explicit form of the additions to the components of the energy-momentum tensor of the electromagnetic field associated with the local nonlinear optical response of the volume of a nonabsorbing isotropic medium is related to the difference between two approaches to the formal determination of the number of waves involved in the interaction.

At widespread *direct approach* it is considered that in the medium demonstrating nonlinearity of n-th order, interact $m+1$ waves with different frequencies $\omega_{1,2,\ldots,m,n+1}$, where $m < n$, for each of which Maxwell equations have the same form. It looks more natural when checking the system of equations describing the interaction of waves in a nonlinear medium for the necessary fulfillment of the laws of conservation of energy, momentum and angular momentum of light. The resulting electric field in this case is first represented as a superposition of field strengths of a given number of interacting waves with different frequencies, and then the coupling between them due to the nonlinearity of the medium is used. But within the *approach based on the limiting transition* from the case $n+1$ of waves with different frequencies to the degenerate case considered in this paper, one can consider that $n+1$ waves interact in the medium, but the equations for $n-m+1$ them having the same frequency ω_m, coincide. This case reflects the point of view, according to which in the medium having nonlinearity of n-th order, always interact exactly $n+1$ waves, even if there is a degeneracy of frequencies. In the framework of the first approach, formula (23) takes the following form

$$\sum_{s=1}^{m} P_{i_s}^{loc}(\omega_s)\partial_p E_{i_s}^{(s)*} + P_{i_{n+1}}^{loc}(\omega_{n+1})\partial_p E_{i_{n+1}}^{(n+1)*} + \text{c.c.} = \tag{24}$$
$$= \frac{1}{m+1}\partial_p\left[\sum_{l=1}^{m}\frac{1}{F(\omega_l)}P_{i_l}^{loc}(\omega_l)E_{i_l}^{(l)*} + \frac{1}{F(-\omega_{n+1})}P_{i_{n+1}}^{loc}(\omega_{n+1})E_{i_{n+1}}^{(n+1)*} + \text{c.c.}\right].$$

To obtain the right-hand side of (24) it is necessary $m+1$ times to write equality (23), successively choosing $\omega_{1,2,\ldots,m,n+1}$ as the frequency standing to the left of the semicolon in the sequence of frequency arguments of the tensor $\hat{\chi}^{(n)}$, add these expressions and divide the obtained result by $m+1$. According to the second approach, formula (23) is written in the form of

$$\sum_{s=1}^{m} P_{i_s}^{loc}(\omega_s)\partial_p E_{i_s}^{(s)*} + P_{i_{n+1}}^{loc}(\omega_{n+1})\partial_p E_{i_{n+1}}^{(n+1)*} + \text{c.c.} = \frac{1}{n+1}\partial_p\left[\sum_{l=1}^{m} P_{i_l}^{loc}(\omega_l)E_{i_l}^{(l)*} + P_{i_{n+1}}^{loc}(\omega_{n+1})E_{i_{n+1}}^{(n+1)*} + \text{c.c.}\right]. \tag{25}$$

To obtain this equality, it is necessary to multiply the right and left parts of (23), written for frequencies $\omega_{1,2,\ldots,m,n+1}$, respectively by $F(\omega_l)$, where $l = 1,2,\ldots,m,n+1$, add the resulting$m+1$ equalities and divide the result by $n+1$. Since $\sum_{l=1}^{m} F(\omega_l) + F(-\omega_{n+1}) = n+1$, the left part after this transformation remains unchanged.

Substitution of the found expressions $\langle \mathbf{P}^{loc}\cdot\partial_p\mathbf{E}\rangle$ into (15) and $\langle \mathbf{P}^{loc}\cdot\partial_t\mathbf{E}\rangle$ into (14) and comparison of the resulting formulas with (16), (17) allow us to write down the additions to the components of the energy-momentum tensor associated with the local nonlinear optical response of the non-absorbing medium volume in the following form:

$$U^{(n,loc)} = \sum_{l=1}^{m}\left[1-K(\omega_l)\right]P_i^{loc}(\omega_l)E_i^{(l)*} + \left[1-K(-\omega_{n+1})\right]P_i^{loc}(\omega_{n+1})E_i^{(n+1)*} + \text{c.c.}, \tag{26}$$

$$S_k^{(n,loc)} = 0, \tag{27}$$

$$g_p^{(n,loc)} = \sum_{l=1}^{m} e_{pij}P_i^{loc}(\omega_l)B_j^{(l)*} + e_{pij}P_i^{loc}(\omega_{n+1})B_j^{(n+1)*} + \text{c.c.} \tag{28}$$

(e_{pij} - Levy-Civita tensor),

$$G_{pk}^{(n,loc)} = \sum_{l=1}^{m}\left[\delta_{pk}K(\omega_l)P_i^{loc}(\omega_l)E_i^{(l)*} - P_k^{loc}(\omega_l)E_p^{(l)*}\right] + \\ + \delta_{pk}K(-\omega_{n+1})P_i^{loc}(\omega_{n+1})E_i^{(n+1)*} - P_k^{loc}(\omega_{n+1})E_p^{(n+1)*} + \text{c.c.} \quad (29)$$

In formulas (26)-(29), in the case of the direct approach, $K(\omega_{l,n+1}) = [(m+1)F(\omega_{l,n+1})]^{-1}$, and in the limit transition approach, $K(\omega_{l,n+1}) = (n+1)^{-1}$. At $m = n$ for all indices $F(\omega_l) = 1$, and using any of them leads to the same result.

The formulas (26) and (29) obtained within the limit transition approach and formula (28), independent of the choice of approach, differ from their analogous expressions for the energy and momentum densities and momentum flux densities due to the nonlinear local optical response of the medium volume in the case when all $n+1$ frequencies of interacting waves differ only in the number of summands in their sums. At the same time, formulas (26) and (29) in the case of the direct approach contain additional coefficients. It will be shown below that both of these approaches to finding the additions to the energy density, field energy flux density vector, momentum density component, and momentum flux density component of the propagating radiation due to the nonlinearity of the medium, when applied to obtain the contribution to the angular momentum flux density due to the nonlinearity of the medium, lead to expressions satisfying the same criterion of equivalence of the contribution of each of the interacting waves, which was demonstrated in [28]. If we substitute into $\langle \mathbf{P}\cdot\partial_p\mathbf{E}\rangle$ the constitutive equations for the nonlocal component of the nonlinear polarization of the medium (formulas (8)-(10)), then, taking into account the spatial derivatives of the electric field strength amplitudes at different frequencies contained in these formulas, we can write the following equality

$$\left\langle \mathbf{P}^{nloc}\cdot\partial_p\mathbf{E}\right\rangle = \sum_{s=1}^{m}P_{i_s}^{nloc}(\omega_s)\partial_pE_{i_s}^{(s)*} + P_{i_{n+1}}^{nloc}(\omega_{n+1})\partial_pE_{i_{n+1}}^{(n+1)*} + \text{c.c.} = \\ = \partial_p\left(\sum_{l=1}^{m-1}A_{i_1^n ik}^{(l)}\prod_{\substack{q=1\\q\neq l}}^{m-1}E_{i_q}^{(q)}\prod_{q=m}^{n}E_{i_q}^{(m)}E_i^{(n+1)*}\partial_kE_{i_l}^{(l)} + \right. \\ \left. + A_{i_1^n ik}^{(m)}\prod_{q=1}^{m-1}E_{i_q}^{(q)}\prod_{q=m}^{n-1}E_{i_q}^{(m)}E_i^{(n+1)*}\partial_kE_{i_n}^{(m)} + A_{i_1^n ik}^{(n+1)}\prod_{q=1}^{m-1}E_{i_q}^{(q)}\prod_{q=m}^{n}E_{i_q}^{(m)}\partial_kE_i^{(n+1)*}\right) - \\ - \partial_k\left(\sum_{l=1}^{m-1}A_{i_1^n ik}^{(l)}\prod_{\substack{q=1\\q\neq l}}^{m-1}E_{i_q}^{(q)}\prod_{q=m}^{n}E_{i_q}^{(m)}E_i^{(n+1)*}\partial_pE_{i_l}^{(l)} + A_{i_1^n ik}^{(m)}\prod_{q=1}^{m-1}E_{i_q}^{(q)}\prod_{q=m}^{m-1}E_{i_q}^{(m)}E_i^{(n+1)*}\partial_pE_{i_n}^{(m)} + \right. \\ \left. + A_{i_1^n ik}^{(n+1)}\prod_{q=1}^{m-1}E_{i_q}^{(q)}\prod_{q=m}^{n}E_{i_q}^{(m)}\partial_pE_i^{(n+1)*}\right) + \text{c.c.} \quad (30)$$

Here $A_{i_1^n ik}^{(l)}$ are unknown auxiliary tensors, the specific form of which determines $U^{(n,nloc)}$, $\mathbf{S}^{(n,nloc)}$, $g_i^{(n,nloc)}$ and $G_{ij}^{(n,nloc)}$. To find $A_{i_1^n ik}^{(l)}$ it is necessary to solve the system of equations, which is formed after opening the derivatives included in the right part of (30) and equating the coefficients contained in the left and right parts at the same combinations of electric field strengths and their spatial derivatives. The system of equations with respect to $A_{i_1^n ik}^{(l)}$ obtained in this way does not have a single solution due to the fact that the conservation laws (16), (17) remain unchanged when expressions with divergences and time derivatives equal to zero are added to them. Nevertheless, starting from the difference of formula (23) from the analogous expression in the case when all frequencies $\omega_{1,2,\ldots,n+1}$ are different, and from the form that formula (30) takes at $m = n$ [27], it is also possible to pick up $A_{i_1^n ik}^{(l)}$ and write (30) in the form

$$\sum_{s=1}^{m} P_{i_s}^{nloc}\left(\omega_s\right)\partial_p E_{i_s}^{(s)*} + P_{i_{n+1}}^{nloc}\left(\omega_{n+1}\right)\partial_p E_{i_{n+1}}^{(n+1)*} + \text{c.c.} =$$
$$= \frac{1}{F\left(\omega_l\right)}\left[\partial_p\left(P_i^{nloc*}\left(\omega_l\right)E_i^{(l)}\right) - \partial_k\left(E_i^{(l)}\Gamma_{ijk}\left(-\omega_l;-\omega_{n+1}\right)\partial_p E_j^{(n+1)*} + E_i^{(l)}\sum_{\substack{s=1\\ s\neq l}}^{m}\Gamma_{ijk}\left(-\omega_l;\omega_s\right)\partial_p E_j^{(s)}\right)\right] + \text{c.c.}, \tag{31}$$

if $l = 1,2,\ldots,m$, and

$$\sum_{s=1}^{m} P_{i_s}^{nloc}\left(\omega_s\right)\partial_p E_{i_s}^{(s)*} + P_{i_{n+1}}^{nloc}\left(\omega_{n+1}\right)\partial_p E_{i_{n+1}}^{(n+1)*} + \text{c.c.} =$$
$$= \frac{1}{F\left(-\omega_{n+1}\right)}\left[\partial_p\left(P_i^{nloc}\left(\omega_{n+1}\right)E_i^{(n+1)*}\right) - \partial_k\left(E_i^{(n+1)*}\sum_{s=1}^{m}\Gamma_{ijk}(\omega_{n+1};\omega_s)\,\partial_p E_j^{(s)}\right)\right] + \text{c.c.}, \tag{32}$$

if $l = n+1$. To be convinced of the validity of these expressions, it is enough to write explicitly the derivatives in their right parts and compare the coefficients at the same combinations of field strengths and their derivatives in the right and left parts of the equations. By virtue of the intrinsic symmetry relations [30], these coefficients appear to be equal to each other.

Using the equalities (31) and (32), two sets of formulas for $U^{(n,nloc)}$, $\mathbf{S}^{(n,nloc)}$, $g_i^{(n,nloc)}$, and $G_{ij}^{(n,nloc)}$, corresponding to the two different criteria of equality of frequencies of interacting waves described above, can be obtained. To realize the direct approach, it is necessary to add the sum of the expressions (31) alternately written for $\omega_l = \omega_{1,2,\ldots,m}$ with the expression (32) and divide the obtained result by $m+1$. The approach based on the limit transition requires, before adding the sum of the expressions (31) written one by one for $\omega_l = \omega_{1,2,\ldots,m}$ with expression (32), to first multiply the summands of this sum by $F(\omega_l)$, and (32) by $F(-\omega_{n+1})$, respectively. Then the result obtained by this operation should be divided by $n+1$. Due to the cumbersome nature of the resulting final expressions similar to (23) and (24), we will not give their explicit form here, but will immediately write down formulas for the additions to the components of the energy-momentum tensor of the electromagnetic field associated with the nonlocal nonlinear optical response of the non-absorbing medium volume:

$$U^{(n,nloc)} = \sum_{l=1}^{m}\left[1 - K\left(\omega_l\right)\right]P_i^{nloc}\left(\omega_l\right)E_i^{(l)*} + \left[1 - K\left(-\omega_{n+1}\right)\right]P_i^{nloc}\left(\omega_{n+1}\right)E_i^{(n+1)*} + \text{c.c.}, \tag{33}$$

$$\begin{aligned} S_k^{(n,\ nloc)} = c^{-1}\sum_{l=1}^{m}\Big[&K\left(-\omega_{n+1}\right)E_i^{(n+1)*}\times\Gamma_{ijk}\left(\omega_{n+1};\omega_l\right)\partial_t E_j^{(l)} + \\ &+K(\omega_l)E_i^{(l)}\Gamma_{ijk}\left(-\omega_l;-\omega_{n+1}\right)\partial_t E_j^{(n+1)*} + \\ &+K(\omega_l)\sum_{\substack{s=1\\ s\neq l}}^{m}E_i^{(l)}\Gamma_{ijk}\left(-\omega_l;\omega_s\right)\partial_t E_j^{(s)}\Big] + \text{c.c.}, \end{aligned} \tag{34}$$

$$g_p^{(n,nloc)} = \sum_{l=1}^{m} e_{pij}P_i^{nloc}\left(\omega_l\right)B_j^{(l)*} + e_{pij}P_i^{nloc}\left(\omega_{n+1}\right)B_j^{(n+1)*} + \text{c.c.}, \tag{35}$$

$$
\begin{aligned}
&G_{pk}^{(n,nloc)} = \\
&= \sum_{l=1}^{m} \left[\delta_{pk} K(\omega_l) P_i^{nloc}(\omega_l) E_i^{(l)*} - P_k^{nloc}(\omega_l) E_p^{(l)*} \right] - \\
&- P_k^{nloc}(\omega_{n+1}) E_p^{(n+1)*} + \delta_{pk} K(-\omega_{n+1}) P_i^{nloc}(\omega_{n+1}) E_i^{(n+1)*} - \\
&- \sum_{l=1}^{m} \Big(K(-\omega_{n+1}) E_i^{(n+1)*} \Gamma_{ijk}(\omega_{n+1};\omega_l) \partial_p E_j^{(l)} + \\
&+ K(\omega_l) E_i^{(l)} \Gamma_{ijk}(-\omega_l;-\omega_{n+1}) \partial_p E_j^{(n+1)*} + \\
&+ K(\omega_l) \sum_{\substack{s=1 \\ s\neq l}}^{m} E_i^{(l)} \Gamma_{ijk}(-\omega_l;\omega_s) \partial_p E_j^{(s)} \Big) + \text{c.c.}
\end{aligned}
\tag{36}
$$

Here the explicit form of $K(\omega_{l,n+1})$ depends on the realized approach and takes the same values as in the case of the above discussed influence of the local nonlinear optical response of the non-absorbing medium. The above comparison of formulas (26)-(29) with the analogous formulas for the energy density, energy flux density, momentum density, and momentum flux density associated with the nonlinear local optical response of the medium volume, obtained earlier for the case in which the frequencies of all $n+1$ interacting waves are different, also remains valid for formulas (33)-(36). The formula for the energy flux density associated with the nonlocal nonlinear optical response (34), using the limit transition approach, differs from the analogous formula for the nondegenerate case only by the number of terms included in it.

Despite the fact that the constitutive equations (5)-(10) for the local and nonlocal components of the nonlinear polarization of the medium were written for brevity in the form corresponding to the situation when only one frequency ω_m has a degeneracy multiplicity higher than unity, the obtained formulas (26)-(29) and (33)-(36) can be easily generalized to the case when several different frequencies of interacting waves have a degeneracy multiplicity higher than unity (up to the situation when the moduli of frequencies of all interacting waves are equal to each other). The tensor used to write the polarization of the medium

$$
\Gamma_{ijk}(\omega_l;\omega_s) = \partial P_i^{(n,nloc)}(\omega_l) / \partial\left(\partial_k E_j^{(s)}\right)
\tag{37}
$$

is in this case the product of the tensor component $\hat{\gamma}^{(n)}$, whose first frequency argument is equal to ω_l, and the last one to ω_s, by $n-1$ components of the electric field strength vector, among which the vector components at frequencies $-\omega_l$ and ω_s occur $F(-\omega_l)-1$ and $F(\omega_s)-1$ times, respectively, and the components at each of the frequencies ω_r ($\omega_r \neq \omega_s$ and $\omega_r \neq -\omega_l$) occur $F(\omega_r)$ times. Each of the sums from one to m in formulas (26)-(29) and (33)-(36) turns in this case into a sum over all possible different frequencies except for the frequency ω_{n+1} accounted for by a separate summand. In this case, in the direct approach, the explicit form of the expressions for $K(\omega_{l,n+1})$ provides a form of the record (26)-(29) and (33)-(36) containing the coefficients necessary to account for the possible degeneracy of several different frequencies. Moreover, at the approach based on the limit transition, formulas (26)-(29) and (33)-(36) do not depend at all on the multiples of degeneracy of the frequencies of the interacting waves.

4. ANGULAR MOMENTUM OF LIGHT AT FREQUENCY DEGENERATE NONLINEAR WAVE INTERACTION

From the law of conservation of momentum (15) we can obtain a formula expressing the law of conservation of angular momentum (moment of momentum) of the electromagnetic field [3, 8]:

$$
c^{-1}\partial_t\left[e_{ijp}x_j g_p\right] + \partial_k\left[e_{ijp}x_j G_{pk}\right] = e_{ikp}G_{pk}.
\tag{38}
$$

Here x_j are the Cartesian coordinates of the radius-vector. Similarly to the way the equations for energy (16) and momentum (17) relate the densities of these quantities to the densities of their fluxes, we can aim to represent (38) as a balance equation for the angular momentum density J_i and the angular momentum flux density M_{ik}:

$$c^{-1}\partial_t J_i + \partial_k M_{ik} = \tau_i. \tag{39}$$

In (39) τ_i is the component of the torque density vector due to the anisotropy of the medium. It is identically equal to zero if the axis i is the axis of symmetry of the medium of infinite order. Since g_p and G_{pk} are expressed as sums of linear and nonlinear components corresponding to different n, and each of the latter consists of two summands responsible for local and nonlocal optical responses of the medium volume to the electric field, J_i, M_{ik}, and τ_i can be naturally represented in the following form

$$J_i = \sum_{n=1}^{\infty}\left(J_i^{(n,loc)} + J_i^{(n,nloc)}\right), \tag{40}$$

$$M_{ik} = \sum_{n=1}^{\infty}\left(M_{ik}^{(n,loc)} + M_{ik}^{(n,nloc)}\right). \tag{41}$$

$$\tau_i = \sum_{n=1}^{\infty}\left(\tau_i^{(n,nloc)} + \tau_i^{(n,nloc)}\right). \tag{42}$$

The comparison of (39) and (38), taking into account (40)-(42), gives rise to the assertion that if

$$J_i^{(n,loc)} = e_{ijp} x_j g_p^{(n,loc)},$$

$$M_{ik}^{(n,loc)} = e_{ijp} x_j G_{pk}^{(n,loc)},$$

$$\tau_i^{(n,loc)} = e_{ikp} G_{pk}^{(n,loc)},$$

then these expressions for any natural n (including linear media) can indeed be considered as angular momentum density, angular momentum flux density, and torque density of light, respectively, related to the local optical response of the medium volume. If the latter is nonlocal, then, as it was shown in [27, 28], the similarly defined $\tau_i^{(n,loc)}$ components of the vector $e_{ikp}G_{pk}^{(n,nloc)}$ are not equal to zero in an isotropic medium, and therefore the latter formula cannot be considered as the torque density. In this connection, it is necessary to transform the expression $e_{ikp}G_{pk}^{(n,nloc)}$ in a way that it is represented as the sum of the full spatial derivative of some quantity $S_{ij}^{(n)}$, which will further describe the contribution of nonlocality of the nonlinear optical response to the angular momentum flux density of light, and the vector $\tilde{\tau}_i^{(n)}$, the projection of which on the symmetry axis of the medium (if present) is equal to zero [28], i.e., to find $S_{ij}^{(n)}$ and $\tilde{\tau}_i^{(n)}$, satisfying the following equality

$$e_{ikp}G_{pk}^{(n,nloc)} = \partial_j S_{ij}^{(n)} + \tilde{\tau}_i^{(n)}. \tag{43}$$

After performing such transformation, the expressions for

$$M_{ik}^{(n,nloc)} = e_{ijp} x_j G_{pk}^{(n,nloc)} - S_{ik}^{(n)}$$

and

$$\tau_i^{(n,nloc)} = \tilde{\tau}_i^{(n)}$$

will correctly describe the contribution of nonlocality of the nonlinear optical response to the angular momentum flux density of light and the torque density, respectively.

To find the explicit form of $S_{ik}^{(n)}$ and $\tilde{\tau}_i^{(n)}$ similarly [28] with the help of Kronecker delta, we redesignate in each of the summands of the formula (36) for $G_{pk}^{(n,nloc)}$ the indices at the components of the electric field strengths so that the fields at the same frequencies in each of them have the same index, and the index associated with the differentiation is always equal to j, naturally without violating the rule according to which the summation is carried out on twice occurring identical indices. The expression for $G_{pk}^{(n,nloc)}$ transformed in this way is then multiplied by e_{ikp} and we group in it all summands with identical combinations of fields and their spatial derivatives. Then in the summand containing the spatial derivative of the field at the degenerate frequency, let us introduce all the remaining field strengths at the same frequency under the differentiation operator. As a result, in further expressions this summand will contain the multiplier $[F(\omega_m)]^{-1}$. The performed transformation is specific for degenerate processes and appears to be possible solely due to the intrinsic symmetry of the tensor $\hat{\gamma}^{(n)}$ [30], due to which one can freely rearrange the indices of the fields belonging to the same frequency.

If several frequencies are degenerate, this procedure must be repeated in each of the summands containing the derivatives of the fields at these frequencies. In the remaining summands, where under the spatial derivative there are fields at nondegenerate frequencies, it is necessary to formally add a coefficient $[F(\omega_l)]^{-1}$ that equal to one. Then, in the obtained expression for $e_{ikp}G_{pk}^{(n,nloc)}$, represent the summand containing the spatial derivative of the field component at the frequency ω_l, as the difference of the summand with the derivative of the product of all electric field strengths at all frequencies and the expression equal to the product of all fields at the frequency ω_l, and the spatial derivative of the product of all other fields whose frequencies are different from ω_l. Then it is necessary to write down $m+1$ times the resulting formula for $e_{ikp}G_{pk}^{(n,nloc)}$, successively considering in each of them the value ω_l equal to $\omega_{1,2,\ldots,m,n+1}$, add them up and, realizing the above-mentioned direct approach to the number of interacting waves, divide the result by $m+1$.

In the approach based on the limiting transition from the interaction of $n+1$ waves with different frequencies to the case when among the frequencies of interacting waves there are equal to each other, it is necessary immediately after writing down the above-mentioned $m+1$ equations to multiply each of them by $F(\omega_l)$ $(l = 1,2,\ldots,m,n+1)$, and only then successively substitute for ω_l the frequencies $\omega_{1,2,\ldots,m,n+1}$. The result of summation of the resulting $m+1$ equations should be divided by $n+1$. Both of these approaches, after dividing the corresponding sums of the above $m+1$ equations by $m+1$ and $n+1$, lead to $e_{ikp}G_{pk}^{(n,nloc)}$ in the left part of the final expression. Its right part in both cases is the sum of two rather cumbersome summands.

The first of these is $\partial_j S_{ij}^{(n)}$, where

$$\begin{aligned}
S_{ij}^{(n)} = -e_{ikp}\Bigg\{\Bigg[&\sum_{s=1}^{m-1} K(\omega_s)\gamma_{ki_1^{s-1}i_{s+1}^{n}i_s j}\left(\omega_{n+1};\bar{\omega}_1^{s-1},\bar{\omega}_{s+1}^{m-1},\tilde{\omega}_m^{(F(\omega_m))},\omega_s\right) + K(\omega_m)\gamma_{ki_1^{n}j}\left(\omega_{n+1};\bar{\omega}_1^{m-1},\tilde{\omega}_m^{(F(\omega_m))}\right)\Bigg]\times \\
&\times E_p^{(n+1)*}\prod_{r=1}^{m-1}E_{i_r}^{(r)}\prod_{r=m}^{n}E_{i_r}^{(m)} + \sum_{l=1}^{m-1}\Big[K(-\omega_{n+1})\gamma_{ki_1^{l-1}i_{l+1}^{n}i_{n+1}j}\left(-\omega_l;\bar{\omega}_1^{l-1},\bar{\omega}_{l+1}^{m-1},\tilde{\omega}_m^{(F(\omega_m))},-\omega_{n+1}\right) + \\
&+\sum_{\substack{s=1\\ s\neq l}}^{m-1} K(\omega_s)\gamma_{ki_{n+1}i_1^{\min(l,s)-1}i_{\min(l,s)+1}^{\max(l,s)-1}i_{\max(l,s)+1}^{n}i_s j}\left(-\omega_l;-\omega_{n+1},\bar{\omega}_1^{\min(l,s)-1},\bar{\omega}_{\min(l,s)+1}^{\max(l,s)-1},\bar{\omega}_{\max(l,s)+1}^{m-1},\tilde{\omega}_m^{(F(\omega_m))},\omega_s\right) + \\
&+K(\omega_m)\gamma_{ki_{n+1}i_1^{l-1}i_{l+1}^{n}j}\left(-\omega_l;-\omega_{n+1},\bar{\omega}_1^{l-1},\bar{\omega}_{l+1}^{m-1},\tilde{\omega}_m^{(F(\omega_m))}\right)\Big]\times E_{i_{n+1}}^{(n+1)*}E_p^{(l)}\prod_{\substack{r=1\\ r\neq l}}^{m-1}E_{i_r}^{(r)}\prod_{r=m}^{n}E_{i_r}^{(m)} + \\
&+\Bigg[\sum_{s=1}^{m-1} K(\omega_s)\gamma_{ki_{n+1}i_1^{s-1}i_{s+1}^{m-1}i_{m+1}^{n}i_s j}\left(-\omega_m;-\omega_{n+1},\bar{\omega}_1^{s-1},\bar{\omega}_{s+1}^{m-1},\tilde{\omega}_m^{(F(\omega_m)-1)},-\omega_s\right) + \\
&+K(-\omega_{n+1})\gamma_{ki_1^{m-1}i_{m+1}^{n}i_{n+1}j}\left(-\omega_m;\bar{\omega}_1^{m-1},\tilde{\omega}_m^{(F(\omega_m)-1)},-\omega_{n+1}\right)\Bigg]\times \\
&\times E_{i_{n+1}}^{(n+1)*}E_p^{(m)}\prod_{r=1}^{m-1}E_{i_r}^{(r)}\prod_{r=m+1}^{n}E_{i_r}^{(m)}\Bigg\} + \text{c.c.}
\end{aligned} \tag{44}$$

Here

$$K\left(\omega_{l,n+1}\right) = \left[\left(m+1\right)F\left(\omega_{l,n+1}\right)\right]^{-1}$$

in the case of the direct approach, but in the limit transition approach,

$$K\left(\omega_{l,n+1}\right) = \left(n+1\right)^{-1}.$$

Given $m = n$ for all indexes $F(\omega_l) = 1$ and using either approach results in the same expression for $S_{ij}^{(n)}$. The second summand, naturally equal to

$$e_{ikp}G_{pk}^{(n,nloc)} - \partial_j S_{ij}^{(n)},$$

where $G_{pk}^{(n,nloc)}$ is given by the formula (36), goes to zero if the nonlinear medium has a symmetry axis of infinite order coinciding with x_i, i.e. is $\tilde{\tau}_i^{(n,nloc)}$. We do not give the explicit form of $\tilde{\tau}_i^{(n,nloc)}$, because differentiating the complex several times occurring products of electric field strengths in $S_{ij}^{(n)}$ makes this formula very cumbersome. Full contribution of the nonlinear optical response to the rotational momentum density

$$\tau_i^{(n)} = e_{ikp}G_{pk}^{(n,loc)} + \tilde{\tau}_i^{(n,nloc)}.$$

To conclude this section, we note that among the various modes of wave interaction caused by odd-order nonlinearities (including the linear response), there are those for the where the sums in expressions (26)-(28), (33)-(36) and (44) are real, and therefore these formulas should be written without the summand c.c. If we choose one of the waves participating in such interaction, for example, having the frequency ω_{n+1}, and write the frequency arguments in the $P_i^{(loc)}(\omega_{n+1})$ and $P_i^{(nloc)}(\omega_{n+1})$ tensors $\hat{\chi}^{(n)}$ and $\hat{\gamma}^{(n)}$ as a sequence $\omega_{n+1}, -\omega_1, -\omega_2, \ldots, -\omega_m$, it will appear that, guided by the intrinsic symmetry relations, the frequency arguments can always be rearranged so that the sum of the frequencies standing next to each other at odd and even places is equal to zero. An example of such a process is the self-interaction of light in a medium with cubic nonlinearity, which together with others will be considered in the next section.

5. SOME EXAMPLES

As examples of the use of the obtained general formulas, we give expressions for the additions to the energy density $U^{(n,nloc)}$, energy flux density vector $S_k^{(n,nloc)}$, components of the momentum density vector $g_p^{(n,nloc)}$, components of the momentum flux density tensor $G_{pk}^{(n,nloc)}$ and component of the tensor $S_{ij}^{(n)}$ associated with the nonlocal nonlinear optical response of the non-absorbing medium volume for generation of the second and third harmonics and self-focusing. The lengthy expressions for the associated with the nonlinear optical response of volume of medium components of angular momentum density

$$J_i^{(n,nloc)} = e_{ijp}x_j g_p^{(n,nloc)},$$

and angular momentum flux density

$$M_{ik}^{(n,nloc)} = e_{ijp}x_j G_{pk}^{(n,nloc)} - S_{ik}^{(n)}$$

and $\tilde{\tau}_i^{(n,nloc)}$ for these processes can be written using the expressions below for $G_{pk}^{(n,nloc)}$ and $S_{ij}^{(n)}$ if necessary.

When the second harmonic is generated in a medium with quadratic nonlinearity $n = 2$, $\omega_{1,2} = \omega$, $m = 1$, $\omega_3 = 2\omega$, and $F(\omega) = 2$, $F(-2\omega) = 1$. Therefore

$$U^{(2,nloc)} = \Big\{\big[1 - K(\omega)\big]\gamma_{ijlk}(-\omega;\omega,-2\omega) \times \\ \times E_i(\omega)\,\partial_k E_l^*(2\omega) + \\ + \big[1 - K(-2\omega)\big]\gamma_{ijlk}(2\omega;\omega,\omega) \times \\ E_i^*(2\omega)\,\partial_k E_l(\omega)\Big\} E_j(\omega) + \text{c.c.}, \tag{45}$$

$$S_k^{(2)} = c^{-1}\Big[K(-2\omega)\gamma_{ijlk}(2\omega;\omega,\omega)E_i^*(2\omega)\,\partial_t E_l(\omega) + \\ + K(\omega)\gamma_{ijlk}(-\omega;\omega,-2\omega)E_i(\omega)\,\partial_t E_l^*(2\omega)\Big]E_j(\omega) + \text{c.c.}, \tag{46}$$

$$g_p^{(2,nloc)} = e_{pij}\Big(\gamma_{ilmk}(-\omega;\omega,-2\omega)B_j(\omega)\,\partial_k E_m^*(2\omega) + \\ + \gamma_{ilmk}(2\omega;\omega,\omega)B_j^*(2\omega)\,\partial_k E_m(\omega)\Big)E_l(\omega) + \text{c.c.}, \tag{47}$$

$$G_{pk}^{(2,nloc)} = \Big\{\delta_{pk}\Big[K(\omega)\gamma_{ijlm}(-\omega;\omega,-2\omega)E_i(\omega)\,\partial_m E_l^*(2\omega) + \\ + K(-2\omega)\gamma_{ijlm}(2\omega;\omega,\omega)E_i^*(2\omega)\,\partial_m E_l(\omega)\Big] - \\ - \Big[K(-2\omega)\gamma_{ijlk}(2\omega;\omega,\omega)E_i^*(2\omega)\,\partial_p E_l(\omega) + \\ + K(\omega)\gamma_{ijlk}(-\omega;\omega,-2\omega)E_i(\omega)\,\partial_p E_l^*(2\omega)\Big] - \\ - \Big[\gamma_{kjlm}(2\omega;\omega,\omega)E_p^*(2\omega)\,\partial_m E_l(\omega) + \\ + \gamma_{kjlm}(-\omega;\omega,-2\omega)E_p(\omega)\,\partial_m E_l^*(2\omega)\Big]\Big\}E_j(\omega) + \text{c.c.}, \tag{48}$$

$$S_{ij}^{(2)} = -e_{ikp}\Big[K(\omega)\gamma_{klmj}(2\omega;\omega,\omega)E_p^*(2\omega)E_m(\omega) + \\ + K(-2\omega)\gamma_{klmj}(-\omega;\omega,-2\omega)E_p(\omega)E_m^*(2\omega)\Big]E_l(\omega) + \text{c.c.} \tag{49}$$

Here $K(\omega) = 1/4$ and $K(-2\omega) = 1/2$ for the direct approach and $K(\omega) = K(-2\omega) = 1/3$ for the approach based on the limit transition. Using the formulas of [27], obtained in the case of nondegenerate processes, we would get in formulas (45)-(49) incorrect values of $K(\omega) = 2/3$ and $K(-2\omega) = 1/3$. Besides, in the formulas (45), (47), as well as in the right-hand side of the equation (48), that not containing $K(\omega)$, the coefficient 2 not existing in the correct equality would appear at the tensor components $\hat{\gamma}$, the first frequency argument of which is equal to $-\omega$.

In the case of third harmonic generation in a medium with cubic nonlinearity, $n = 3$, $\omega_{1,2,3} = \omega$, $\omega_4 = 3\omega$i.e., $m = 1$, $F(\omega) = 3$ and $F(-3\omega) = 1$. As a result, we obtain

$$U^{(3,nloc)} = \Big\{\big[1 - K(\omega)\big]\gamma_{ijlmk}(-\omega;\omega,\omega,-3\omega)E_i(\omega)\,\partial_k E_m^*(3\omega) + \\ + \big[1 - K(-3\omega)\big]\gamma_{ijlmk}(3\omega;\omega,\omega,\omega)E_i^*(3\omega) \times \\ \times \partial_k E_m(\omega)\Big\}E_j(\omega)E_l(\omega) + \text{c.c.}, \tag{50}$$

$$S_k^{(3)} = c^{-1}\Big[K(-3\omega)\gamma_{ijlmk}(3\omega;\omega,\omega,\omega)E_i^*(3\omega)\,\partial_t E_m(\omega) + K(\omega)\gamma_{ijlmk}(-\omega;\omega,\omega,-3\omega)E_i(\omega)\,\partial_t E_m^*(3\omega)\Big] \times \\ \times E_j(\omega)E_l(\omega) + \text{c.c.}, \tag{51}$$

$$g_p^{(3,nloc)} = e_{pij}\left(\gamma_{ilmnk}\left(-\omega;\omega,\omega,-3\omega\right)B_j\left(\omega\right)\partial_k E_n^*\left(3\omega\right) + \gamma_{ilmnk}\left(3\omega;\omega,\omega,\omega\right)B_j^*\left(3\omega\right)\partial_k E_n\left(\omega\right)\right)\times E_l\left(\omega\right)E_m\left(\omega\right) + \text{c.c.}, \quad (52)$$

$$\begin{gathered} G_{pk}^{(3,nloc)} = \Big\{\delta_{pk}\Big[K\left(\omega\right)\gamma_{ijlmn}\left(-\omega;\omega,\omega,-3\omega\right)E_i\left(\omega\right)\partial_n E_m^*\left(3\omega\right) + \\ +K\left(-3\omega\right)\gamma_{ijlmn}\left(3\omega;\omega,\omega,\omega\right)E_i^*\left(3\omega\right)\partial_n E_m\left(\omega\right)\Big] - \\ -\Big[K\left(-3\omega\right)\gamma_{ijlmk}\left(3\omega;\omega,\omega,\omega\right)E_i^*\left(3\omega\right)\partial_p E_m\left(\omega\right) + \\ +K\left(\omega\right)\gamma_{ijlmk}E_i\left(\omega\right)\partial_p E_m^*\left(3\omega\right)\Big] - \\ -\Big[\gamma_{kjlmn}\left(3\omega;\omega,\omega,\omega\right)E_p^*\left(3\omega\right)\partial_n E_m\left(\omega\right) + \\ +\gamma_{kjlmn}\left(-\omega;\omega,\omega,-3\omega\right)E_p\left(\omega\right)\partial_n E_m^*\left(3\omega\right)\Big]\Big\}\times E_j(\omega)E_l(\omega) + \text{c.c.}, \end{gathered} \quad (53)$$

$$\begin{gathered} S_{ij}^{(3)} = -e_{ikp}\Big[K\left(\omega\right)\gamma_{klmnj}\left(3\omega;\omega,\omega,\omega\right)E_p^*\left(3\omega\right)E_n\left(\omega\right) + \\ +K\left(-3\omega\right)\gamma_{klmnj}\left(-\omega;\omega,\omega,-3\omega\right)E_p\left(\omega\right)E_n^*\left(3\omega\right)\Big]\times E_l\left(\omega\right)E_m\left(\omega\right) + \text{c.c.} \end{gathered} \quad (54)$$

In these formulas

$$K(\omega) = 1/6 \quad \text{and} \quad K(-3\omega) = 1/2$$

using the direct approach and

$$K\left(\omega\right) = K\left(-3\omega\right) = 1/4$$

when using the approach based on the limit transition. Using the formulas of [27], obtained in the case of non-degenerate processes, we would obtain in formulas (50)-(54) incorrect values $K(\omega) = 3/4$ and $K(-3\omega) = 1/4$. Also in the formulas (50), (52), and (53) there would appear the coefficient 3, which does not exist in the correct expression, at the summands containing the tensor components $\hat{\gamma}$, whose first frequency argument is equal to $-\omega$.

If there is self-action of an electromagnetic wave (process $\omega = -\omega + \omega + \omega$) in a medium with cubic nonlinearity ($n = 3$), then the frequency $\omega_1 = -\omega$ and $\omega_{2,3,4} = \omega$. Formally, we can consider that two waves with frequencies ω and $-\omega$ interact, and therefore $m = 2$ and $F(\omega) = F(-\omega) = 2$. In this case, the formulas take the form

$$\begin{gathered} U^{(3,nloc)} = \frac{3}{4}\Big[\gamma_{ijlmk}\left(\omega;-\omega,\omega,\omega\right)E_i^*\left(\omega\right)\partial_k E_m\left(\omega\right) + \\ +\gamma_{iljmk}\left(-\omega;\omega,-\omega,-\omega\right)E_i\left(\omega\right)\partial_k E_m^*\left(\omega\right)\Big]\times E_j^*\left(\omega\right)E_l\left(\omega\right), \end{gathered} \quad (55)$$

$$\begin{gathered} S_k^{(3)} = \frac{1}{4c}\Big[\gamma_{ijlmk}(\omega;-\omega,\omega,\omega)E_i^*(\omega)\partial_t E_m(\omega) + \\ +\gamma_{iljmk}(-\omega;\omega,-\omega,-\omega)E_i(\omega)\partial_t E_m^*(\omega)\Big]\times E_j^*\left(\omega\right)E_l\left(\omega\right), \end{gathered} \quad (56)$$

$$\begin{gathered} g_p^{(3,nloc)} = e_{pij}\,[\gamma_{ilmnk}(\omega;-\omega,\omega,\omega)B_j^*(\omega)\partial_k E_n(\omega) + \\ +\gamma_{imlnk}(-\omega;\omega,-\omega,-\omega)B_j(\omega)\partial_k E_n^*(\omega)]\times E_l^*(\omega)E_m(\omega), \end{gathered} \quad (57)$$

$$\begin{gathered} G_{pk}^{(3,nloc)} = \{\frac{1}{4}[\delta_{pk}(\gamma_{ijlmn}(\omega;-\omega,\omega,\omega)E_i^*(\omega)\partial_n E_m^*(\omega) + \\ +\gamma_{iljmn}(-\omega;\omega,-\omega,-\omega)E_i(\omega)\partial_n E_m^*(\omega)) - \\ -(\gamma_{ijlmk}(\omega;-\omega,\omega,\omega)E_i^*(\omega)\partial_p E_m(\omega) + \end{gathered}$$

$$+\gamma_{iljmk}(\omega;\omega,-\omega,-\omega)E_i(\omega)\,\partial_p E_m^*(\omega))]-$$

$$-\gamma_{kjlmn}(\omega;-\omega,\omega,\omega)E_p^*(\omega)\,\partial_n E_m(\omega)-\gamma_{kljmn}(-\omega;\omega,-\omega,-\omega)E_p(\omega)\,\partial_n E_m^*(\omega)\}\times E_j^*(\omega)E_l(\omega), \quad (58)$$

$$S_{ij}^{(3)}=-\frac{e_{ikp}}{4}[\gamma_{klmnj}(\omega;-\omega,\omega,\omega)\,E_p^*(\omega)E_n(\omega)+\gamma_{kmlnj}(-\omega;\omega,-\omega,-\omega)\,E_p(\omega)E_n^*(\omega)]\times E_l^*(\omega)E_m(\omega). \quad (59)$$

In self-focusing, both approaches lead to the same results. Note that in this process each of the expressions is automatically real; and the presence of complex conjugation in the formulas is not required. Using the formulas [27] obtained in the case of nondegenerate processes, we would obtain twice as large values as those in the right-hand sides of the equations (55)-(59).

6. CONCLUSIONS

In this work, analytical expressions for additions to the energy density, energy flux density, momentum density, momentum flux density, angular momentum density components, and components of the angular momentum flux density tensor due to the local and nonlocal nonlinear optical response of the n-th order of a homogeneous nonabsorbing medium volume in the case when the real number of interacting waves with different frequencies in it is less than or equal to n have been obtained. These additions cannot be directly determined from analogous expressions previously obtained in the case of nonlinear interaction $n+1$ of waves with different frequencies in such a medium, if in them we put some of the frequencies equal to each other, i.e. we formally consider that $n+1$ waves propagate, but the frequencies, amplitudes and wave vectors of some of them completely coincide. The formulas obtained in this paper can be used not only by those who rightly believe that the number of interacting waves with different frequencies participating in degenerate nonlinear optical processes is less than or equal to n, but also by those for whom the degenerate process is obtained as a limiting transition from the case $n+1$ of different frequencies of interacting waves, as a result of which some of them are made equal to each other, i.e., $n+1$ waves formally propagate in the medium, but some of them are completely identical to each other. The two sets of formulas obtained as a result of these two approaches differ only in the numerical coefficients, whose values are determined by the multiples of degeneracy of the frequencies of the interacting waves and the number of summands in the sums included in them. The formulas obtained in the first case have a more complicated form. Each summand in them explicitly contains the multiplicity of degeneracy of the corresponding frequency. In the second case, the analytical expressions for the fundamental characteristics of the field appear outwardly similar to the analogous formulas of the studies in which all $n+1$ frequencies of interacting waves are different. The fundamental difference of the formulas obtained in the paper is manifested in the number of summands in the expressions for the additions to the electromagnetic field characteristics due to the nonlinearity of the medium.

The found formulas for the additions to the energy density, energy flux density, momentum density, momentum flux density, components of the angular momentum density and components of the angular momentum flux density tensor due to the local and nonlocal nonlinear optical response of the volume of a homogeneous nonabsorbing medium allow us to write down their specific form for all degenerate processes of nonlinear optics.

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