Cascaded second-order contribution to the third-order nonlinear susceptibility

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Cascading of second-order nonlinear effects leads to an effective third-order nonlinearity. In addition to the macroscopic electric field at the intermediate frequencies another term has to be taken into account which is due to the locality of the intermediate polarization sources. Combining the correction terms at the three intermediate frequencies gives rise to a third-order susceptibility tensor, which exhibits the same symmetry properties as an intrinsic susceptibility. This particularly applies to the contributions from the rectified and the second-harmonic fields to the degenerate susceptibility.

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I. INTRODUCTION

It is well known that the cascading of second-order nonlinearities contributes to higher-order nonlinear effects [1,2] such as harmonic generation [3] or frequency shifting [4,5]. In the case of only weak electromagnetic fields at the intermediate frequencies the cascaded process can be described as Kerr nonlinearity by means of an equivalent third-order nonlinear susceptibility or a nonlinear refractive index. This has been shown for the self-action of electromagnetic waves by cascading via the second harmonic field [6,7], cascading via the rectified field [8–10], and also in degenerate four-wave mixing [11,12]. In the first step of the cascading process a polarization at an intermediate frequency is generated by the second-order nonlinearity from one or two fundamental waves. This polarization acts as a source for an electromagnetic field at the intermediate frequency, which is the solution of the inhomogeneous Maxwell equations. This macroscopic field mixes with another field to produce a nonlinear polarization at their sum or difference frequency in the second step. In addition to the macroscopic field another contribution exists at the intermediate frequency which is due to the nonlinear source term that has to be included in the relation between the local and the macroscopic fields at the intermediate frequency. The fact that there are two contributions to the generated polarization in the second step has been shown in Refs. [1,2]. Compared with a straightforward calculation with cascading of macroscopic susceptibilities, the second contribution can be regarded as a correction term due to cascading through the local field. Other authors consider it as the primary contribution and the resulting macroscopic electric field as a depolarization field [13]. The correction term can be neglected in many cascading situations where the macroscopic field at the intermediate frequency is strong compared to the additional field. But if both contributions are of the same order of magnitude, they both have to be regarded in calculations. This necessity can occur in cascading via an intermediate field in a situation far away from the phase-matching condition or via the rectified field. Contributions via optical rectification and the linear electro-optic effect have been considered in Ref. [10] and especially in degenerate four-wave mixing in Refs. [11–14], for solitary waves [15] and various cascading situations in Ref. [16]. In addition to the correction term in the rectified field, the contribution from the second harmonic field is taken into account in Ref. [12]. In most references, Refs. [11–15], the two contributions (macroscopic field and correction term) belonging to one intermediate field are combined at the same time in the calculations, but looked at separately for the different intermediate frequencies or wave vectors in the degenerate case. Because of the locality of the correction terms in contrast to the macroscopic geometry-dependent fields, it is reasonable to consider their contributions to a macroscopic third-order susceptibility separately from the contributions of the macroscopic fields in order to obtain a property that characterizes the material independent of the geometry. Since there are up to three different intermediate frequencies, generally three different contributions to this susceptibility have to be combined. This has been done, e.g., in Ref. [17], but only in a scalar manner.

In this paper a special emphasis is laid on the susceptibility containing the local source, especially on its tensorial nature. The main objective is to show that the resulting macroscopic susceptibility tensor has the same important properties as all susceptibility tensors have, provided that the contributing first- and second-order susceptibilities exhibit these properties, which are described in many textbooks, e.g., in Ref. [18]. As a material characteristic it can be directly added to the intrinsic susceptibility. The paper is organized as follows: in Sec. II an expression for the additional susceptibility is derived, some symmetry properties of which are examined in Sec. III. The degenerate case regarding only one frequency is considered in Sec. IV.

II. DERIVATION OF THE LOCAL CASCADING SUSCEPTIBILITY

The basic equation for describing the dependence of the macroscopic polarization on the macroscopic electric field in nonlinear materials is the well-known expansion of **P** into a Volterra series in **E**, which in the frequency domain reads as

$$\mathbf{P}(\omega) = \sum_{n=1}^{\infty} \mathbf{P}^{(n)}(\omega), \tag{1a}$$

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$$\mathbf{P}^{(n)}(\omega) = \frac{\varepsilon_0}{(2\pi)^{n-1}} \int_{-\infty}^{\infty} \cdots \int_{-\infty}^{\infty} \boldsymbol{\chi}^{(n)}(\omega;\omega_1,\ldots,\omega_{n-1},\omega_n) \vdots \mathbf{E}(\omega_1)\cdots\mathbf{E}(\omega_{n-1})\vdots\mathbf{E}(\omega_n)d\omega_1\cdots d\omega_{n-1}, \tag{1b}$$

where $\omega=\Sigma_{i=1}^{n}\ \omega_i$. $\mathbf{P}^{(n)}$ is the nth-order nonlinear polarization and is described by the convolution (1b). This expansion with the macroscopic susceptibilities $\boldsymbol{\chi}^{(n)}$ expects macroscopic fields $\mathbf{E}$ as a result of sources in a distance sufficiently far away from the observed points. We assume some monofrequent waves $\mathbf{E}_i$ with phasors $\hat{\mathbf{E}}_i$ and a time dependence of the form $\mathbf{E}_i(t)=\frac{1}{2}\hat{\mathbf{E}}_i e^{j\omega_i t}+\text{c.c.}$ The polarization vectors $\mathbf{P}_i$ also follow this notation. If, for example, two waves $\mathbf{E}_1$ and $\mathbf{E}_2$ at some frequencies ω_1 and ω_2, respectively, are coupled into a nonlinear material which lacks a center of inversion, then a polarization $\mathbf{P}_{im,0}$ at an intermediate frequency $\omega_{im}=\omega_1+\omega_2$ is generated with a phasor according to

$$\hat{\mathbf{P}}_{im,0} = \varepsilon_0 \boldsymbol{\chi}^{(2)}(\omega_{im};\omega_1,\omega_2):\hat{\mathbf{E}}_1\hat{\mathbf{E}}_2. \tag{2}$$

It is part of the second-order polarization $\mathbf{P}^{(2)}$ from Eq. (1b). It acts as a source for an electromagnetic field $\mathbf{E}_{im}$ at the frequency ω_{im} which is solution of the Maxwell equations and strongly depends on the geometry of the considered device and the exciting optical waves. At the sum frequencies, for example, at $\omega_{sum}=\omega_{im}+\omega_1$, a polarization is produced by the nonlinear interaction of $\mathbf{E}_{im}$ and $\mathbf{E}_1$. The electric field $\mathbf{E}_{im}$ is not the resulting field of sources in greater distance but of the local source $\mathbf{P}_{im,0}$ in the vicinity of the fields. Thus, the polarization $\mathbf{P}_{sum,0}$ at the sum frequency is not expressed analogous to Eq. (2) but contains another term, which has to be added to the electric field $\mathbf{E}_{im}$ and which is directly dependent on the source $\mathbf{P}_{im,0}$. A short explanation for the occurrence of this additional term is recalled as follows. Since the nonlinear polarizations basically depend on the microscopic (local) and not on the macroscopic fields, it is necessary to look at the relationship between the macroscopic polarization $\mathbf{P}_{im}$, the macroscopic electric field $\mathbf{E}_{im}$, and the local electric field $\mathbf{E}_{loc}$ at the places of the microscopic dipoles:

$$\hat{\mathbf{E}}_{loc} = \hat{\mathbf{E}}_{im} + \frac{1}{3\varepsilon_0}\hat{\mathbf{P}}_{im}. \tag{3}$$

This relationship has been derived by Lorentz [19] for cubic crystals but is assumed to be also valid for other crystals. If the macroscopic field $\hat{\mathbf{E}}_{im}$ was the result of external sources, the macroscopic polarization would be $\hat{\mathbf{P}}_{im}=\varepsilon_0\boldsymbol{\chi}^{(1)}\hat{\mathbf{E}}_{im}$:

$$\hat{\mathbf{E}}_{loc} = \left(\mathbf{I} + \tfrac{1}{3}\boldsymbol{\chi}^{(1)}\right)\hat{\mathbf{E}}_{im} = \tfrac{1}{3}(\boldsymbol{\varepsilon} + 2\mathbf{I})\hat{\mathbf{E}}_{im}. \tag{4}$$

Here, $\boldsymbol{\varepsilon}=\mathbf{I}+\boldsymbol{\chi}^{(1)}$ is used and $\mathbf{I}$ is the unity tensor. Solving for $\hat{\mathbf{E}}_{im}$ and substituting it in an expression for the sum-frequency polarization analogous to Eq. (2), i.e.,

$$\hat{\mathbf{P}}_{sum,0} = \varepsilon_0 \boldsymbol{\chi}^{(2)}(\omega_{sum};\omega_1,\omega_{im}):\hat{\mathbf{E}}_1\hat{\mathbf{E}}_{im}, \tag{5}$$

yields

$$\hat{\mathbf{P}}_{sum,0} = \varepsilon_0 \boldsymbol{\chi}^{(2)}(\omega_{sum};\omega_1,\omega_{im}):\hat{\mathbf{E}}_1[3(\boldsymbol{\varepsilon} + 2\mathbf{I})^{-1}\hat{\mathbf{E}}_{loc}]. \tag{6}$$

Similarly, the macroscopic field $\hat{\mathbf{E}}_1$ could be expressed by its respective local field. Equation (6) represents the basic relationship between the sum-frequency polarization and the intermediate-frequency electric field. Of course, if Eq. (4) is valid as a result of external sources, Eq. (5) is equivalent to Eq. (6). However, in cascading the total macroscopic polarization $\hat{\mathbf{P}}_{im}$ at the intermediate frequency consists of two parts, i.e., the term proportional to the macroscopic electric field $\boldsymbol{\varepsilon}_0\boldsymbol{\chi}^{(1)}\hat{\mathbf{E}}_{im}$ and the local source $\hat{\mathbf{P}}_{im,0}$. Thus, the local field (3) takes the form

$$\hat{\mathbf{E}}_{loc} = \left(\mathbf{I} + \frac{1}{3}\boldsymbol{\chi}^{(1)}\right)\hat{\mathbf{E}}_{im} + \frac{1}{3\varepsilon_0}\hat{\mathbf{P}}_{im,0}. \tag{7}$$

Using Eq. (7) in Eq. (6) results in

$$\hat{\mathbf{P}}_{sum,0} = \varepsilon_0 \boldsymbol{\chi}^{(2)}(\omega_{sum};\omega_1,\omega_{im}):\hat{\mathbf{E}}_1\{\hat{\mathbf{E}}_{im} + [\varepsilon_0(\boldsymbol{\varepsilon} + 2\mathbf{I})]^{(-1)}\hat{\mathbf{P}}_{im,0}\}. \tag{8}$$

In addition to Eq. (5), this equation contains a second contribution to $\hat{\mathbf{P}}_{sum,0}$ which accounts for the locality of the intermediate source and which always exists in cascading regardless of the geometry and independent of the evolution of an electromagnetic field. In many cases of cascading in integrated optics this additional term can be neglected, as the electromagnetic field $\hat{\mathbf{E}}_{im}$ at the intermediate frequency is often — intentionally or not — much larger than the contribution $[\varepsilon_0(\boldsymbol{\varepsilon}+2\mathbf{I})]^{(-1)}\hat{\mathbf{P}}_{im,0}$. But in some cases, e.g., at optical rectification, the energy transfer from the fundamental field to the intermediate field (quasistatic field) is very small. Thus, both contributions in Eq. (8) can be of the same order of magnitude and have to be regarded in calculations.

In the following we will restrict the examinations on the second contribution in Eq. (8) that explicitly contains the polarization source $\mathbf{P}_{im,0}$. Its effect on the polarization at the sum frequency $\mathbf{P}_{sum,0}$ can be regarded as the result of a third-order nonlinearity that only depends on the properties of the material and not on the geometry. It can be looked at separately from the first contribution in Eq. (8), i.e., the contribution from the macroscopic electric field at the intermediate frequency. A combination of the three contributions at the up to three intermediate frequencies yields an equivalent third-order susceptibility, a general expression for which is derived in the following. For this susceptibility $\boldsymbol{\chi}^{(3),ls}(\omega_4;\omega_1,\omega_2,\omega_3)$ (the upper index ls stands for local source), with $\omega_4=\Sigma_{i=1}^{3}\ \omega_i$, all three intermediate frequencies $\omega_{ij}=\omega_{ji}=\omega_i+\omega_j; i,j=1,2,3; i\neq j$ have to be included in the calculation. As before, $\hat{\mathbf{E}}_i$ are phasors of the electric (input) fields and $\hat{\mathbf{P}}_{ij,0}$, with $i\neq j$, are the phasors of the intermediate polarizations at the frequencies ω_{ij}. Each of $\hat{\mathbf{P}}_{ij,0}$ is given by

$$\hat{\mathbf{P}}_{ij,0} = \varepsilon_0 \boldsymbol{\chi}^{(2)}(\omega_{ij};\omega_i,\omega_j):\hat{\mathbf{E}}_i\hat{\mathbf{E}}_j. \tag{9}$$

The tensor $\boldsymbol{\eta}$ is defined in the principal-axis system of the nonlinear material as

$$\boldsymbol{\eta}(\omega) = [\boldsymbol{\varepsilon}(\omega) + 2\mathbf{I}]^{-1} \tag{10}$$

and accounts for the tensor that appears in front of the intermediate source in Eq. (8). With the three frequencies ω_{ij} three contributions for the nonlinear polarization at the sum frequency $\omega_4 = \omega_1 + \omega_2 + \omega_3$ are obtained:

$$\begin{aligned}\hat{\mathbf{P}}_{4,0} = {} & \boldsymbol{\chi}^{(2)}(\omega_4;\omega_1,\omega_{23}):\hat{\mathbf{E}}_1[\boldsymbol{\eta}(\omega_{23})\hat{\mathbf{P}}_{23,0}] \\ & + \boldsymbol{\chi}^{(2)}(\omega_4;\omega_2,\omega_{13}):\hat{\mathbf{E}}_2[\boldsymbol{\eta}(\omega_{13})\hat{\mathbf{P}}_{13,0}] \\ & + \boldsymbol{\chi}^{(2)}(\omega_4;\omega_3,\omega_{12}):\hat{\mathbf{E}}_3[\boldsymbol{\eta}(\omega_{12})\hat{\mathbf{P}}_{12,0}].\end{aligned} \tag{11}$$

The direct contribution of an intrinsic susceptibility $\boldsymbol{\chi}^{(3)}$ to the polarization $\hat{\mathbf{P}}_{4,0}$ would be

$$\hat{\mathbf{P}}_{4,0} = \tfrac{3}{2}\varepsilon_0 \boldsymbol{\chi}^{(3)}(\omega_4;\omega_1,\omega_2,\omega_3):\ \hat{\mathbf{E}}_1\hat{\mathbf{E}}_2\hat{\mathbf{E}}_3. \tag{12}$$

Using Eq. (9) in Eq. (11) and comparing the result with Eq. (12) yield the equivalent susceptibility of third order $\boldsymbol{\chi}^{(3),ls}$, the components of which are given by

$$\begin{aligned}\chi^{(3),ls}_{ijkl}(\omega_4;\omega_1,\omega_2,\omega_3) = {} & \tfrac{2}{3}\chi^{(2)}_{ijm}(\omega_4;\omega_1,\omega_{23})\,\eta_{mn}(\omega_{23}) \\ & \times\chi^{(2)}_{nkl}(\omega_{23};\omega_2,\omega_3) \\ & + \tfrac{2}{3}\chi^{(2)}_{ikm}(\omega_4;\omega_2,\omega_{13})\,\eta_{mn}(\omega_{13}) \\ & \times\chi^{(2)}_{njl}(\omega_{13};\omega_1,\omega_3) \\ & + \tfrac{2}{3}\chi^{(2)}_{ilm}(\omega_4;\omega_3,\omega_{12})\,\eta_{mn}(\omega_{12}) \\ & \times\chi^{(2)}_{nkj}(\omega_{12};\omega_2,\omega_1).\end{aligned} \tag{13}$$

Note that the sum convention is applied, i.e., Eq. (13) has to be summed over repeated indices. The total third-order susceptibility is the sum of all contributions:

$$\boldsymbol{\chi}^{(3),tot} = \boldsymbol{\chi}^{(3),ls} + \boldsymbol{\chi}^{(3),E} + \boldsymbol{\chi}^{(3),int}. \tag{14}$$

Here, $\boldsymbol{\chi}^{(3),E}$ denotes the cascaded susceptibility due to the macroscopic electric intermediate fields and $\boldsymbol{\chi}^{(3),int}$ is the intrinsic susceptibility. Maybe further contributions to the total susceptibility can be found, e.g., by cascading from higher-order nonlinearities.

III. PROPERTIES OF THE ADDITIONAL SUSCEPTIBILITY

If all three contributions are regarded for the susceptibility $\boldsymbol{\chi}^{(3),ls}$ as it is done in Eq. (13), then the resulting tensor exhibits some important symmetry properties, provided that the contributing tensors $\boldsymbol{\eta}$ and $\boldsymbol{\chi}^{(2)}$ have these properties. One important implication of these symmetries is the fact that, for example, a mathematical model for a nonlinear interaction of multiple optical waves can be energy conserving if each susceptibility involved is lossless (i.e., it has no imaginary part) and obeys the symmetry relations. The mentioned general properties are described, for instance, in Ref. [18].

The first property, the time-reversal symmetry, can be directly seen from Eq. (13) in the frequency domain:

$$\chi^{(n)}_{i;k_1,\ldots,k_n}(\omega;\omega_1,\ldots,\omega_n) = [\chi^{(n)}_{i;k_1,\ldots,k_n}(-\omega;-\omega_1,\ldots,-\omega_n)]^*. \tag{15}$$

The asterisk denotes complex conjugation. Also the intrinsic permutation symmetry

$$\begin{aligned}& \chi^{(n)}_{i;k_1,\ldots,k_p,\ldots,k_q,\ldots,k_n}(\omega;\omega_1,\ldots,\omega_p,\ldots,\omega_q,\ldots,\omega_n) \\ & \quad = \chi^{(n)}_{i;k_1,\ldots,k_q,\ldots,k_p,\ldots,k_n}(\omega;\omega_1,\ldots,\omega_q,\ldots,\omega_p,\ldots,\omega_n)\end{aligned} \tag{16}$$

is fulfilled: if any two of the indices 1, 2, 3 together with the respective indices j,k,l are interchanged in Eq. (13), then one of the expressions on the right-hand side remains unchanged while the other two change their roles mutually. The overall permutation symmetry

$$\begin{aligned}& \chi^{(n)}_{i;k_1,\ldots,k_p,\ldots,k_n}(\omega;\omega_1,\ldots,\omega_p,\ldots,\omega_n) \\ & \quad = \chi^{(n)}_{k_p;k_1,\ldots,i,\ldots,k_n}(-\omega_p;\omega_1,\ldots,-\omega,\ldots,\omega_n)\end{aligned} \tag{17}$$

is valid for $\boldsymbol{\chi}^{(3),ls}$ if it is valid for the contributing tensors: let us consider, e.g., the susceptibility

$$\begin{aligned}\chi^{(3),ls}_{jikl}(-\omega_1;-\omega_4,\omega_2,\omega_3) = {} & \tfrac{2}{3}\chi^{(2)}_{jim}(-\omega_1;-\omega_4,\omega_{23})\,\eta_{mn}(\omega_{23}) \\ & \times\chi^{(2)}_{nkl}(\omega_{23};\omega_2,\omega_3) \\ & + \tfrac{2}{3}\chi^{(2)}_{jkm}(-\omega_1;\omega_2,-\omega_{12})\,\eta_{mn}(-\omega_{12}) \\ & \times\chi^{(2)}_{nil}(-\omega_{12};-\omega_4,\omega_3) \\ & + \tfrac{2}{3}\chi^{(2)}_{jlm}(-\omega_1;\omega_3,-\omega_{13})\,\eta_{mn}(-\omega_{13}) \\ & \times\chi^{(2)}_{nki}(-\omega_{13};\omega_2,-\omega_4),\end{aligned} \tag{18}$$

based on the definition (13) and the equation $\omega_4 = \omega_1 + \omega_2 + \omega_3$. Application of Eq. (17) on Eq. (18) for each of the $\chi^{(2)}$ elements containing negative frequencies as arguments yields

$$\begin{aligned}\chi^{(3),ls}_{jikl}(-\omega_1;-\omega_4,\omega_2,\omega_3) = {} & \tfrac{2}{3}\chi^{(2)}_{ijm}(\omega_4;\omega_1,\omega_{23})\,\eta_{mn}(\omega_{23}) \\ & \times\chi^{(2)}_{nkl}(\omega_{23};\omega_2,\omega_3) \\ & + \tfrac{2}{3}\chi^{(2)}_{mkj}(\omega_{12};\omega_2,\omega_1)\,\eta_{mn}(-\omega_{12}) \\ & \times\chi^{(2)}_{iln}(\omega_4;\omega_3,\omega_{12}) \\ & + \tfrac{2}{3}\chi^{(2)}_{mjl}(\omega_{13};\omega_1,\omega_3)\,\eta_{mn}(-\omega_{13}) \\ & \times\chi^{(2)}_{ikn}(\omega_4;\omega_2,\omega_{13}).\end{aligned} \tag{19}$$

The first term on the right-hand side of Eq. (19) is identical to the first one in Eq. (13), and the other two change their roles if the order of the factors is reversed, the indices $n \leftrightarrow m$ are interchanged, and the symmetry (17) of the $\boldsymbol{\eta}$ tensor [25] is used. So the validity of the overall permutation symmetry (17) for $\boldsymbol{\chi}^{(3),ls}$ is shown. The prerequisite that $\boldsymbol{\chi}^{(2)}$ obeys Eqs. (15) and (16) is fulfilled as these are intrinsic properties of the susceptibilities; Eq. (17) applies to $\boldsymbol{\chi}^{(2)}$ in lossless materials. This is also true for $\boldsymbol{\eta}$ as long as the tensorial definition (10) or a similar expression based on the permittivity $\boldsymbol{\varepsilon}$ is valid.

A further property is the spatial symmetry of the respective crystal class. The interdependence of the tensor elements and the zero elements is determined by the symmetry operations that can be performed on a crystal of the respective class. Let s_{ij} be the elements of the matrix of such an operation. Then a tensor is transformed into the new coordinate system according to

$$\tilde{\chi}^{(n)}_{j;l_1,\ldots,l_n} = s_{ji}s_{l_1k_1}\cdots s_{l_nk_n}\chi^{(n)}_{i;k_1,\ldots,k_n}. \tag{20}$$

It can be shown that the tensor (13) exhibits the same interdependencies of the elements as an intrinsic tensor does: each of the three products on the right-hand side of Eq. (13), performed on transformed tensors, is of the form

$$\tilde{\chi}^{(2)}_{abc}\tilde{\eta}_{cd}\tilde{\chi}^{(2)}_{def} = s_{ai}s_{bj}s_{cm}\chi^{(2)}_{ijm}s_{cp}s_{dq}\eta_{pq}s_{dn}s_{ek}s_{fl}\chi^{(2)}_{nkl}. \tag{21}$$

Since $s_{cm}s_{cp}=\delta_{mp}$ and $s_{dq}s_{dn}=\delta_{qn}$ [20], where δ_{mp} equals 1 for $m=p$ and 0 for $m\neq p$, it follows that

$$\tilde{\chi}^{(2)}_{abc}\tilde{\eta}_{cd}\tilde{\chi}^{(2)}_{def} = s_{ai}s_{bj}\chi^{(2)}_{ijm}\eta_{mn}s_{ek}s_{fl}\chi^{(2)}_{nkl}. \tag{22}$$

This means that the whole tensor (13), when transformed to the new coordinate system, obeys the transformation law (20):

$$\tilde{\chi}^{(3),ls}_{abef} = s_{ai}s_{bj}s_{ek}s_{fl}\chi^{(3),ls}_{ijkl}. \tag{23}$$

Therefore it has the same spatial symmetries as an intrinsic susceptibility.

IV. DEGENERATE SUSCEPTIBILITIES

Equation (13) is deduced for different frequencies ω_i but can be equally applied to degenerate cases with common frequencies. Especially the susceptibility describing self- and cross action of waves at one frequency, i.e., $\boldsymbol{\chi}^{(3),ls}(\omega;\omega,\omega,-\omega)$, can be written according to Eq. (13) as

$$\begin{aligned}\chi^{(3),ls}_{ijkl}(\omega;\omega,\omega,-\omega) =& \tfrac{2}{3}\chi^{(2)}_{ijm}(\omega;\omega,0)\,\eta_{mn}(0)\,\chi^{(2)}_{nkl}(0;\omega,-\omega)\\ &+\tfrac{2}{3}\chi^{(2)}_{ikm}(\omega;\omega,0)\,\eta_{mn}(0)\,\chi^{(2)}_{njl}(0;\omega,-\omega)\\ &+\tfrac{2}{3}\chi^{(2)}_{ilm}(\omega;-\omega,2\omega)\,\eta_{mn}(2\omega)\\ &\times\chi^{(2)}_{nkj}(2\omega;\omega,\omega).\end{aligned} \tag{24}$$

The first two addends can be identified as cascading contributions with zero as intermediate frequency, and the third one has the second harmonic as intermediate field. It should be noted that there are no other degeneracy factors in Eq. (24) than in Eq. (13). This can either be seen when the three frequencies $(\omega_1,\omega_2,\omega_3)$ are continuously approaching $(\omega,\omega,-\omega)$, or it can be shown in a direct deduction via the rectified and the second-harmonic fields. In the latter case only one of the first two expressions in Eq. (24) would occur at first, but with a degeneracy factor of $4/3$ instead of $2/3$. The splitting of this expression into the first two ones in Eq. (24) complies with the averaging over permutations of index-frequency pairs [18] in order to fulfill the intrinsic permutation symmetry (16) for $\chi^{(3),ls}_{ijkl}$. These two terms correspond to the two contributions which are investigated, for example, in Ref. [12] in degenerate four-wave mixing due to different wave vectors in the rectified field. Equation (25) shows that by summing up these two contributions and the contribution from cascading over the second-harmonic field, the resulting tensor $\boldsymbol{\chi}^{(3),ls}$ has the properties discussed in Sec. III as a local susceptibility tensor of third order. Thus, also in the degenerate case energy conservation of the physical system can be shown in a macroscopic view when the local-field cascading is accounted for, even if no macroscopic electric rectified or second-harmonic field exists.

The susceptibility (24) can be expressed in terms of the commonly given electro-optic coefficients r_{ijk} and the nonlinear coefficients d_{ijk}. With the relation between the electro-optic coefficients r_{ijk} and the respective susceptibility

$$\chi^{(2)}_{ijk}(\omega,\omega,0) = -\tfrac{1}{2}n_i^2n_j^2r_{ijk}, \tag{25}$$

and with $\chi^{(2)}_{ijk}(2\omega,\omega,\omega)=2d_{ijk}$, Eq. (24) becomes

$$\begin{aligned}\chi^{(3),ls}_{ijkl}(\omega;\omega,\omega,-\omega) =& \tfrac{1}{6}n_i^2n_j^2n_k^2n_l^2\,\eta_{mn}(0)(r_{ijm}r_{lkn}+r_{ikm}r_{ljn})\\ &+\tfrac{8}{3}\eta_{mn}(2\omega)d_{mli}d_{nkj},\end{aligned} \tag{26}$$

provided that the susceptibilities are real quantities.

As a practical example we consider a uniaxial material in which a z-polarized optical wave exists. The effective element of the equivalent susceptibility is the 3333-element:

$$\chi^{(3),ls}_{3333}(\omega;\omega,\omega,-\omega) = \frac{1}{3}\frac{n_e^8(\omega)r_{33}^2}{\varepsilon_{33}(0)+2} + \frac{8}{3}\frac{d_{33}^2}{n_e^2(2\omega)+2}. \tag{27}$$

The second cascading contribution $\boldsymbol{\chi}^{(3),E}$ in Eq. (14) depends on the geometry of the system. The macroscopic electric field may vanish at the second-harmonic frequency in a situation far away from the phase-matching condition. Also the macroscopic rectified field can be zero or negligible: if the extension of the optical wave and the nonlinear source term in the (quasi) static field is much larger in the direction of the static polarization (i.e., the z direction) than in the directions perpendicular to it. In that case, Eq. (27) is the only cascading contribution. If the extension of the optical wave is small in the direction of the static polarization compared to the perpendicular directions, then the macroscopic rectified field reaches its maximum value of $\hat{E}_z=-\hat{P}_{z,0}/(\varepsilon_0\varepsilon_{33})$ [21], and with its negative sign, it is directed in the opposite direction of the polarization source $\hat{P}_{z,0}$. Its contribution to the nonlinear susceptibility can be derived to

$$\chi^{(3),E}_{3333}(\omega;\omega,\omega,-\omega) = -\frac{1}{3}\frac{n_e^8(\omega)r_{33}^2}{\varepsilon_{33}(0)}. \tag{28}$$

Conclusively, depending on the macroscopic rectified field — the depolarization field — the sum of the two cascading contributions ranges between

$$\chi^{(3),casc}_{3333}(\omega;\omega,\omega,-\omega) = -\frac{1}{3}\frac{2n_e^8(\omega)r_{33}^2}{\varepsilon_{33}(\varepsilon_{33}+2)} + \frac{8}{3}\frac{d_{33}^2}{n_e^2(2\omega)+2} \tag{29}$$

and the value given by Eq. (27). To assess the order of magnitude of the sum of both cascading contributions, some averaged data are taken from Refs. [22,23] for lithium niobate.

The nonlinear refractive index [24] $n_2=3Z_0\chi^{(3)}_{3333}/(4n_e^2)$ corresponding to Eq. (29) and (27) then ranges between 3.2×10^{-17} and 3.6×10^{-15} cm^2/W at a wavelength of 1 μm and low-frequency modulation. This means that in the case of a maximum macroscopic electric field, the sum of the cascading contributions is two orders of magnitude smaller than in the case of a vanishing macroscopic field. Further configurations are discussed, for instance, in Ref. [16]. Of course, the intrinsic third-order susceptibility has to be added, too. It should be mentioned that Eq. (28) is based on a static calculation, i.e., that the optical wave is continuous or slowly varying. For very short optical pulses the depolarization field leading to Eq. (28) is no longer directly proportional to the polarization source but takes a more complicated temporal shape [8,9].

V. CONCLUSIONS

Among the contributions of cascaded second-order nonlinearities to effective third-order susceptibilities, the contribution that accounts for the locality of the intermediate nonlinear polarizations is considered in this paper. This susceptibility consists of up to three different parts due to three intermediate polarizations. It is shown that this third-order susceptibility exhibits the same symmetry properties that are known from (intrinsic) susceptibilities. The elements of the tensor can be of considerable magnitude, but they can significantly be compensated by the cascading contribution via the macroscopic (depolarization) field, depending on the geometry of the configuration and the shape of the optical waves.

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[25] Application of Eq. (17) on $\boldsymbol{\eta}$ is possible, as this second-rank tensor can also be supplied with a second argument: $\eta_{nm}(\omega_{ij}) = \eta_{nm}(\omega_{ij};\omega_{ij}) = \eta_{nm}(-\omega_{ij};-\omega_{ij}) = \eta_{nm}(-\omega_{ij})$.