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# TENSORS IN CIRCULAR-CYLINDRICAL COORDINATES FOR THE DESCRIPTION OF HARMONIC GENERATION OF CIRCULARLY POLARIZED LASER LIGHT

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A method for finding nonzero and linearly independent higher-rank tensor elements by recourse to irreducible spherical tensors, is proposed. A transformation matrix from third-rank tensors, defined in circular-cylindrical coordinates, to irreducible spherical tensors, is derived. Nonzero elements of polar third- and fourth-rank tensors, symmetric in all indices, are determined for all crystallographical classes. The results are applied to the analysis of free as well as electric field-induced second- and third-harmonic generation of laser light.

## 1. Introduction

Various physical problems are better dealt with in circular-cylindrical coordinates. This is in particular the case of studies involving circularly polarized waves. Physical quantities are represented by tensors, the elements of which depend on the choice of reference system. According to the type of the physical quantity, the tensor representing it is of some well-defined rank and possesses a given permutational symmetry of its indices. It is of practical importance to determine which of the tensor elements are nonzero and how many of them are linearly independent for physical systems with symmetry of a given point group $G$ (translational symmetry imposes no conditions on the tensor elements). The literature concerned with the analysis of the properties of tensors defined in a cartesian reference frame is very extensive [1–4].

In this paper, we shall consider the problem with regard to circular-cylindrical coordinates. With this in mind, we shall derive irreducible spherical tensors in the form of functions of tensor elements, defined in circular-cylindrical coordinates, which will serve for the determination of basis vectors of irreducible representations of point groups and hence nonzero tensor elements. The knowledge of the transformation matrix from tensors to irreducible spherical tensors is in itself a matter of high relevance since it permits one to solve many physical problems with ease and elegance. A matrix of this kind is given in table I for tensors of rank 3. Tables II and III contain the nonzero elements of tensors of ranks 3 and 4 with total permutational symmetry of indices, for all point groups.

Originally, experimental work on the generation of (difference and summation) harmonics had recourse essentially to linearly polarized light beams. In recent years, circularly polarized beams are being applied increasingly. The possibilities of generation are then more strongly dependent on the symmetry of the medium and the propagation direction of the beam than if the latter is polarized linearly. This has been applied on various occasions in research on liquid crystals, coherent-light generation in the far infrared, and measurements of picosecond light-pulse duration. A detailed discussion of selection rules for nonlinear interactions with a circularly polarized beam is to be found in the paper of Tang and Rabin [5], where references to the experimental and theoretical literature are also listed (see, moreover, the review articles [6]).

The nonzero elements of tensors of ranks 3 and 4 calculated here in a circular-cylindrical basis permit one to calculate the nonlinear electric polarisation of crystals and hence the relative intensity of light generated with a given frequency. The selection rules then result naturally from the vanishing, or not vanishing, of the appropriate electric polarisations.

## 2. The method of irreducible spherical tensors

If $x, y, z$ are versors of cartesian coordinates, the circular-cylindrical reference system is defined by the unit vectors $e_1, e_0, e_{-1}$ as follows:

$$e_1 = (-i/\sqrt{2})(x + iy), \quad e_0 = iz, \quad e_{-1} = (i/\sqrt{2}) \times (x - iy). \tag{1}$$

In this system of reference, one can define tensors $a^n_{ijk} \ldots$ of an arbitrary rank $n$, each of the $n$ indices $i, j, k, \ldots$ taking one of the values $1, 0, -1$.

For symmetric systems (molecules, crystals) of a point group $G$, the number of mutually independent tensor elements can undergo a reduction. Tensor invariance with respect to spatial symmetry operations causes some of the elements to vanish, and linear relations can also exist between some.

Tensors are characterized by permutational symmetry of their indices. The symmetry of an $n$th rank tensor is defined by a group $T^n = G \times P_n$, the direct product of a given point group $G$ and the corresponding subgroup $P_n$ of the symmetric group $S_n$, where $P_n$ describes the intrinsic symmetry of the tensor [3]. Any tensor can be written as a sum of tensors (of the same rank), each having a different property with regard to the permutation symmetry of its indices. This separation is essential in that the tensor describing a given physical quantity possesses a well defined permutational symmetry.

When determining the independent and nonzero tensor elements, we shall have recourse to the following theorem: Only those elements of a polar (axial) tensor are nonzero which belong to the symmetric (antisymmetric) representation of the point group $G$. The number of nonzero elements is equal to the number of possible combinations of tensor elements yielding basis vectors of the symmetric (antisymmetric) representation. The relation between the linearly independent and remaining elements is given by equating to zero the basis vectors of irreducible representations of the group other than the symmetric (antisymmetric) representation.

With regard to the circumstance that all point groups $G$ are subgroups of the rotation group $K_h$, the bases of irreducible representations of these groups can be determined from irreducible spherical tensors $T^l(a^n)$, which transform as irreducible representations $D^l_{g,u}$ of the group $K_h$. Spherical tensors composed of elements of a polar tensor of even rank transform as representations $D^l_g$, and odd ones, as $D^l_u$. Inversely, for axial tensors, odd ones belong to representations of the type g and even ones to the type u. This manner of proceeding has two advantages. 1) It is in itself important to have available the tensors $T^l(a^n)$ since they make many a calculation simpler by permitting one to have recourse to the well developed formalism of angular-momentum theory; and 2) one obtains a separation of the tensor into parts with strictly defined permutational properties of their indices $i, j, k \ldots$.

Irreducible spherical tensors $T^l(a^n)$ can be constructed step by step beginning by tensors of rank 1 which, by eq. (1), have the elements:

$$T^1_m(a^1) = a^1_m; \quad a^1_{\pm 1} = (\mp i/\sqrt{2})(a^1_x \pm i a^1_y), \quad a^1_0 = i a^1_z \tag{2}$$

and having recourse to the formula [7]:

$$T^l_m(a^{n_1+n_2}) = \sum_{m_1 m_2} \begin{bmatrix} l_1 & l_2 & l \\ m_1 & m_2 & m \end{bmatrix} T^{l_1}_{m_1}(a^{n_1}) \times T^{l_2}_{m_2}(a^{n_2}). \tag{3}$$

Above, the symbols in square brackets are Clebsch–Gordan coefficients, and $l$ takes the values $l_1 + l_2$, $l_1 + l_2 - l, \ldots, |l_1 - l_2|$. In this way, by multiplying two tensors of rank 1 we get three tensors $T^2(a^2)$, $T^1(a^2)$, $T^0(a^2)$. By using formula (3) we obtain tensors of a special kind only, being products of two tensors of the form $a^{n_1}_{ijk} \ldots a^{n_2}_{prs} \ldots$; this restriction, however, does not affect the generality of our considerations as the transformation laws of an arbitrary tensor are the same as for this special tensor, and this is the only property we shall be needing. The tensors of rank 2, $a^2_{ij}$, have the property that the tensors $T^2(a^2)$ and $T^0(a^2)$ are symmetric with regard to an interchange of the indices $i$ and $j$, whereas $T^1(a^2)$ is antisymmetric.

Multiplying the tensors $T^n(a^2)$ by $T^1(a^1)$ in accordance with formula (3), we obtain the spherical representation of the tensor $a^3_{ijk}$. However, in this case, not all the tensors $T^l(a^3)$ will possess a permutational symmetry so uniquely defined as was the case for the tensor of rank 2. Nevertheless it is possible to perform a symmetrization by forming appropriate linear combinations with elements of different spherical tensors of the same rank using genealogical coefficients [8, 9]. The coefficients of these combinations do not depend on the index $m$ of $T^l_m$.

From two tensors $T^1(a^3) = T^0(a^2) \times T^1(a^1)$ and $\bar{T}^1(a^3) = T^2(a^2) \times T^1(a^1)$, one can form one tensor, symmetric in all three indices, ${}^{(3)}T^1$, and one tensor symmetric in two only, ${}^{(2)}T^1$. They have the form:

$$
\begin{aligned}
{}^{(3)}T^1_m(a^3) &= \tfrac{2}{3}\,\bar{T}^1_m(a^3) + (\tfrac{5}{9})^{\frac{1}{2}}\,T^1_m(a^3),\\
{}^{(2)}T^1_m(a^3) &= -(\tfrac{5}{9})^{\frac{1}{2}}\,\bar{T}^1_m(a^3) + \tfrac{2}{3}\,T^1_m(a^3).
\end{aligned} \tag{4}
$$

We shall be denoting symmetry in $k$ indices by writing $k$ in parentheses, and antisymmetry by using square brackets.

The remaining tensors have well defined symmetry and require no further symmetrization. If a tensor is symmetric or antisymmetric in two indices, this bears on the first two indices of the tensor $a^3_{ijk}$.

The results are given in table I in the form of transformation coefficients $A^{lm\nu}_{ijk}$, relating elements of the spherical tensors with those of the tensors $a^3_{ijk}$:

$$
{}^{\nu}T^l_m(a^3) = \sum_{ijk} A^{lm\nu}_{ijk}\, a^3_{ijk}. \tag{5}
$$

Since the coefficients $A^{lm\nu}_{ijk}$ are elements of a unitary transformation matrix, the transformation inverse to (5) is easy to perform.

By a similar procedure, spherical tensors, representing the tensor $a^4_{ijkl}$ of rank 4 are readily derived. For a tensor symmetric in all its indices, the results are to be found in ref. 10; we shall use this in order to derive its nonzero elements.

Once the form of the spherical tensors as functions of the tensor elements $a^n_{ijk}\ldots$ is known, it is easy to determine the basis vectors of irreducible representations of the point groups $G$. Denoting by $\varphi^A_\alpha(n)$ an $\alpha$ vector of the irreducible representation $A$ of the group $G$ formed with elements of a tensor of rank $n$, we have:

$$
\varphi^A_\alpha(n) = \sum_m B^{lm}_{A\alpha}\, T^l_m(a^n). \tag{6}
$$

The coefficients $B^{lm}_{A\alpha}$ are tabulated [11]. Our choice of phase for spherical tensors, following Leushin [11] is such that the relation

$$
T^l_m = (-1)^{l-m}\, T^{l\,*}_{-m}, \tag{7}
$$

holds. This choice of phase is a guarantee that, on carrying out multiplication of spherical tensors by formula (3), the property (7) is unchanged. By having recourse to this method, we calculated the mutually independent and nonzero elements for tensors up to rank 4 inclusive, for all point groups. In tables II and III we list the results for polar tensors of ranks 3 and 4 symmetric in all indices. If an orientation of the reference system is required that is different from that assumed hereafter ref. 11, it can be obtained by performing a rotation given by the Euler angles $\alpha$, $\beta$, $\gamma$ and invoking the transformation law for spherical tensors:

$$
T^{l\,\prime}_m = \sum_k D^l_{mk}(\alpha, \beta, \gamma)\, T^l_k. \tag{8}
$$

The prime denotes spherical tensor elements in the new system of reference. The $D^l_{mk}(\alpha, \beta, \gamma)$ are Wigner functions.

## 3. Light-harmonics generation

A medium immersed in the field of light waves exhibits a polarisation $P(t)$, which contains in its spectrum besides the fundamental frequencies also sum, difference and harmonic frequencies. Each of the components of the spectrum of nonlinear polarisations can be expressed in terms of the intensities of the electric fields of the light waves as follows [6, 12]:

$$
P_i(\omega_3) = \chi^{(2)}_{i[j][k]}(-\omega_3, \omega_1, \omega_2)\, E^{\omega_1}_j E^{\omega_2}_k, \tag{9}
$$

$$
P_i(\omega_4) = \chi^{(3)}_{i[j][k][l]}(-\omega_4, \omega_1, \omega_2, \omega_3)\, E^{\omega_1}_j E^{\omega_2}_k E^{\omega_3}_l, \tag{10}
$$

Table I
Coefficients $A_{ijk}^{lm\nu} : {}^{\nu}T_m^l = \sum_{i,j,k} A_{ijk}^{lm\nu} a_{ijk}$

| | 0 0 0 | 0 1 1 | 1 0 1 | 1 1 0 | 0 −1 −1 | −1 0 −1 | −1 −1 0 | 0 1 −1 | 1 0 −1 | 1 −1 0 | 0 −1 1 | −1 0 1 | −1 1 0 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| ${}^{(3)}T_{10}$ | $-\sqrt{\frac{3}{2}}$ | | | | | | | $1/\sqrt{15}$ | $1/\sqrt{15}$ | $1/\sqrt{15}$ | $1/\sqrt{15}$ | $1/\sqrt{15}$ | $1/\sqrt{15}$ |
| ${}^{(3)}T_{30}$ | $\sqrt{\frac{2}{5}}$ | | | | | | | $1/\sqrt{10}$ | $1/\sqrt{10}$ | $1/\sqrt{10}$ | $1/\sqrt{10}$ | $1/\sqrt{10}$ | $1/\sqrt{10}$ |
| ${}^{(3)}T_{32}$ | | $1/\sqrt{3}$ | $1/\sqrt{3}$ | $1/\sqrt{3}$ | | | | | | | | | |
| ${}^{(3)}T_{3-2}$ | | | | | $1/\sqrt{3}$ | $1/\sqrt{3}$ | $1/\sqrt{3}$ | | | | | | |
| ${}^{(2)}T_{10}$ | | | | | | | | $-1/2\sqrt{3}$ | $-1/2\sqrt{3}$ | $1/\sqrt{3}$ | $-1/2\sqrt{3}$ | $-1/2\sqrt{3}$ | $1/\sqrt{3}$ |
| ${}^{(2)}T_{20}$ | | | | | | | | $\frac{1}{2}$ | $\frac{1}{2}$ | | $-\frac{1}{2}$ | $-\frac{1}{2}$ | |
| ${}^{(2)}T_{22}$ | | $-1/\sqrt{6}$ | $-1/\sqrt{6}$ | $2/\sqrt{6}$ | | | | | | | | | |
| ${}^{(2)}T_{2-2}$ | | | | | $1/\sqrt{6}$ | $1/\sqrt{6}$ | $-2/\sqrt{6}$ | | | | | | |
| ${}^{[2]}T_{10}$ | | | | | | | | $-\frac{1}{2}$ | $\frac{1}{2}$ | | $-\frac{1}{2}$ | $\frac{1}{2}$ | |
| ${}^{[2]}T_{20}$ | | | | | | | | $-1/2\sqrt{3}$ | $1/2\sqrt{3}$ | $1/\sqrt{3}$ | $1/2\sqrt{3}$ | $-1/2\sqrt{3}$ | $-1/\sqrt{3}$ |
| ${}^{[2]}T_{22}$ | | $-1/\sqrt{2}$ | $1/\sqrt{2}$ | | | | | | | | | | |
| ${}^{[2]}T_{2-2}$ | | | | | $1/\sqrt{2}$ | $-1/\sqrt{2}$ | | | | | | | |
| ${}^{[3]}T_{00}$ | | | | | | | | $-1/\sqrt{6}$ | $1/\sqrt{6}$ | $-1/\sqrt{6}$ | $1/\sqrt{6}$ | $-1/\sqrt{6}$ | $1/\sqrt{6}$ |

| | 1 1 1 | −1 −1 −1 | 1 1 −1 | 1 −1 1 | −1 1 1 | 1 −1 −1 | −1 1 −1 | −1 −1 1 | 0 0 1 | 0 1 0 | 1 0 0 | 0 0 −1 | 0 −1 0 | −1 0 0 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| ${}^{(3)}T_{11}$ | | | $2/\sqrt{15}$ | $2/\sqrt{15}$ | $2/\sqrt{15}$ | | | | $-1/\sqrt{15}$ | $-1/\sqrt{15}$ | $-1/\sqrt{15}$ | | | |
| ${}^{(3)}T_{1-1}$ | | | | | | $2/\sqrt{15}$ | $2/\sqrt{15}$ | $2/\sqrt{15}$ | | | | $-1/\sqrt{15}$ | $-1/\sqrt{15}$ | $-1/\sqrt{15}$ |
| ${}^{(3)}T_{33}$ | 1 | | | | | | | | | | | | | |
| ${}^{(3)}T_{3-3}$ | | 1 | | | | | | | | | | | | |
| ${}^{(3)}T_{31}$ | | | $1/\sqrt{15}$ | $1/\sqrt{15}$ | $1/\sqrt{15}$ | | | | $2/\sqrt{15}$ | $2/\sqrt{15}$ | $2/\sqrt{15}$ | | | |
| ${}^{(3)}T_{3-1}$ | | | | | | $1/\sqrt{15}$ | $1/\sqrt{15}$ | $1/\sqrt{15}$ | | | | $2/\sqrt{15}$ | $2/\sqrt{15}$ | $2/\sqrt{15}$ |
| ${}^{(2)}T_{11}$ | | | $-1/\sqrt{3}$ | $1/2\sqrt{3}$ | $1/2\sqrt{3}$ | | | | $-1/\sqrt{3}$ | $1/2\sqrt{3}$ | $1/2\sqrt{3}$ | | | |
| ${}^{(2)}T_{1-1}$ | | | | | | $1/2\sqrt{3}$ | $1/2\sqrt{3}$ | $-1/\sqrt{3}$ | | | | $-1/\sqrt{3}$ | $1/2\sqrt{3}$ | $1/2\sqrt{3}$ |
| ${}^{(2)}T_{21}$ | | | $1/\sqrt{3}$ | $-1/2\sqrt{3}$ | $-1/2\sqrt{3}$ | | | | $-1/\sqrt{3}$ | $1/2\sqrt{3}$ | $1/2\sqrt{3}$ | | | |
| ${}^{(2)}T_{2-1}$ | | | | | | $1/2\sqrt{3}$ | $1/2\sqrt{3}$ | $-1/\sqrt{3}$ | | | | $1/\sqrt{3}$ | $-1/2\sqrt{3}$ | $-1/2\sqrt{3}$ |
| ${}^{[2]}T_{11}$ | | | | $-\frac{1}{2}$ | $\frac{1}{2}$ | | | | | $-\frac{1}{2}$ | $\frac{1}{2}$ | | | |
| ${}^{[2]}T_{1-1}$ | | | | | | $\frac{1}{2}$ | $-\frac{1}{2}$ | | | | | | $-\frac{1}{2}$ | $\frac{1}{2}$ |
| ${}^{[2]}T_{21}$ | | | | $\frac{1}{2}$ | $-\frac{1}{2}$ | | | | | $-\frac{1}{2}$ | $\frac{1}{2}$ | | | |
| ${}^{[2]}T_{2-1}$ | | | | | | $\frac{1}{2}$ | $-\frac{1}{2}$ | | | | | | $\frac{1}{2}$ | $-\frac{1}{2}$ |

Table II

Nonzero elements of the polar tensor $a_{ijk}$, symmetric in all its indices. For symmetry groups having a centre of symmetry, as well as for the groups $K$ and $O$, all elements vanish

| Symmetry group | Tensor elements | Polarisation state of the second harmonic |
|---|---|---|
| $T$, $T_d$<br>$C_{4v}$, $C_{5v}$, $C_{6v}$, ..., $C_{\infty v}$ | $a_{110} = -a_{-1\,-1\,0}$ | none |
| $C_4$, $C_5$, $C_6$, ... $C_\infty$ | $a_{000}$, $a_{1\,-1\,0}$ | none |
| $D_3$, $D_{3h}$ | $a_{111} = a_{-1\,-1\,-1}$ | opp |
| $C_{3h}$ | $a_{111}$, $a_{-1-1-1}$ | opp |
| $C_{3v}$ | $a_{111} = -a_{-1\,-1\,-1}$, $a_{000}$, $a_{1\,-1\,0}$ | opp |
| $C_3$ | $a_{111}$, $a_{-1\,-1\,-1}$, $a_{000}$, $a_{1\,-1\,0}$ | opp |
| $D_2$, $D_{2d}$ | $a_{110} = -a_{-1\,-1\,0}$ | none |
| $S_4$ | $a_{110}$, $a_{-1\,-1\,0}$ | none |
| $C_{2v}$ | $a_{000}$, $a_{110} = a_{-1\,-1\,0}$, $a_{1\,-1\,0}$ | none |
| $C_2$ | $a_{000}$, $a_{110}$, $a_{-1-10}$, $a_{1-10}$ | none |
| $C_s$ | $a_{111}$, $a_{-1\,-1\,-1}$, $a_{100}$, $a_{-1\,0\,0}$, $a_{1\,-1\,-1}$, $a_{1\,1\,-1}$ | ellipt |
| $C_1$ | $a_{000}$, $a_{111}$, $a_{-1\,-1\,-1}$, $a_{100}$, $a_{-1\,0\,0}$, $a_{110}$, $a_{-1\,-1\,0}$,<br>$a_{1\,-1\,-1}$, $a_{1\,1\,-1}$, $a_{1\,-1\,0}$ | ellipt |

Table III

Nonzero elements of the polar tensor $a_{ijkl}$, symmetric in all its indices

| Symmetry group | Tensor elements | Polarisation state of the third harmonic |
|---|---|---|
| $K$, $K_h$ | $a_{1\,-1\,0\,0}$, $a_{1\,1\,-1\,-1} = 10a_{1\,-1\,0\,0}$, $a_{0000} = -21a_{1\,-1\,0\,0}$ | none |
| $O$, $O_h$, $T$, $T_h$, $T_d$ | $a_{1\,-1\,0\,0}$, $a_{1111} = a_{-1\,-1\,-1\,-1}$, $a_{1\,1\,-1\,-1} = 10a_{1\,-1\,0\,0}$<br>$- 7a_{1111}$, $a_{0000} = -21a_{1\,-1\,0\,0} + 14a_{1111}$ | opp |
| $C_5$ ... $C_\infty$<br>$C_{5v}$ ... $C_{\infty v}$<br>$C_{5h}$ ... $C_{\infty h}$<br>$D_5$ ... $D_\infty$<br>$D_{5h}$ ... $D_{\infty h}$<br>$C_{3h}$, $D_{3h}$ | $a_{0000}$, $a_{1\,1\,-1\,-1}$, $a_{1\,-1\,0\,0}$ | none |
| $C_{3v}$, $D_3$, $D_{3d}$ | $a_{0000}$, $a_{1110} = a_{-1\,-1\,-1\,0}$, $a_{1\,1\,-1\,-1}$, $a_{1\,-1\,0\,0}$ | none |
| $C_3$, $C_{3i}$ | $a_{0000}$, $a_{1110}$, $a_{-1\,-1\,-1\,0}$, $a_{1\,1\,-1\,-1}$, $a_{1\,-1\,0\,0}$ | none |
| $C_4$, $S_4$, $C_{4h}$ | $a_{0000}$, $a_{1111}$, $a_{-1\,-1\,-1\,-1}$, $a_{1\,1\,-1\,-1}$, $a_{1\,-1\,0\,0}$ | opp |
| $D_4$, $D_{4h}$, $D_{2d}$, $C_{4v}$ | $a_{0000}$, $a_{1111} = a_{-1\,-1\,-1\,-1}$, $a_{1\,1\,-1\,-1}$, $a_{1\,-1\,0\,0}$ | opp |
| $D_2$, $D_{2h}$, $C_{2v}$ | $a_{0000}$, $a_{1111} = a_{-1\,-1\,-1\,-1}$, $a_{1\,1\,1\,-1} = a_{1\,-1\,-1\,-1}$,<br>$a_{1\,1\,-1\,-1}$, $a_{1100} = a_{-1\,-1\,0\,0}$, $a_{1\,-1\,0\,0}$ | ellipt |
| $C_2$, $C_s$, $C_{2h}$ | $a_{0000}$, $a_{1111}$, $a_{-1\,-1\,-1\,-1}$, $a_{1\,1\,1\,-1}$, $a_{1\,-1\,-1\,-1}$,<br>$a_{1100}$, $a_{-1\,-1\,0\,0}$, $a_{1\,1\,-1\,-1}$, $a_{1\,-1\,0\,0}$ | ellipt |
| $C_1$, $C_i$ | $a_{0000}$, $a_{1\,1\,1\,1}$, $a_{-1\,-1\,-1\,-1}$, $a_{1000}$, $a_{-1\,0\,0\,0}$,<br>$a_{1110}$, $a_{1\,1\,1\,-1}$, $a_{1\,-1\,-1\,-1}$, $a_{-1\,-1\,-1\,0}$, $a_{1100}$<br>$a_{-1\,-1\,0\,0}$, $a_{1\,1\,-1\,-1}$, $a_{1\,1\,-1\,0}$, $a_{1\,-1\,0\,0}$, $a_{1\,-1\,-1\,0}$ | ellipt |

where the tensors* of ranks 3 and 4, $\chi^{(2)}_{ijk}$ and $\chi^{(3)}_{ijkl}$, are tensors of nonlinear susceptibility of the second and third order respectively, and the tensor indices take the values 1, 0, −1. The square brackets denote that the tensor elements are taken in the basis contra-standard to the basis (1). The transition from the standard to the contrastandard basis is the following:

$$e_{[q]} = (-1)^{1-q}\, e_{-q}. \tag{11}$$

Eqs. (10) together with tables II and III yield directly expressions for the various symmetry groups for the case of light propagating along the $z$ axis corresponding to the standard basis.

Generation by circularly polarized light presents a number of highly interesting properties and is very sensitive to the symmetry of the crystal and the direction of propagation of the exciting light beams. *E.g.*, light propagating along a threefold or sixfold inversion axis cannot generate a third harmonic, and the second harmonic is then circularly polarized oppositely to the incident wave. For the symmetry groups $D_3$, $D_{3h}$, $C_{3v}$ as well as $T$ and $T_d$, if the beam propagates along a 3 or $\bar{6}$ axis, the generation effects for right- and left-polarized light are equal to each other owing to the relation:

$$P_1(2\omega) = \chi^{(2)}_{111} E^{\omega}_{-1} E^{\omega}_{-1} = \pm P_{-1}(2\omega)$$
$$= \chi^{(2)}_{-1\,-1\,-1} E^{\omega}_{1} E^{\omega}_{1}, \tag{12}$$

but will differ in the case of the groups $C_3$ and $C_{3h}$, for which in general $\chi^{(2)}_{111} \neq \chi^{(2)}_{-1\,-1\,-1}$.

For the situation under discussion, generation of light with sum and difference frequencies will occur only if the beams, with different frequencies, are circularly polarized in the same sense; the light generated is then circularly polarized in the opposite sense.

A light beam, circularly polarized, propagating along a fourfold axis (4, $\bar{4}$, $4/m$) cannot cause second-harmonic generation; the third harmonic is then polarized oppositely. In the symmetry groups $O$, $O_h$, $T_d$, $D_4$, $D_{4h}$, $D_{2d}$ and $C_{4v}$, the third harmonic generated by right- and left-polarized light will have the same intensity (since here $\chi^{(3)}_{1111} = \chi^{(3)}_{-1\,-1\,-1\,-1}$), whereas in the groups $C_4$, $C_{4h}$ and $S_4$ these intensities can differ, since in general $\chi^{(3)}_{1111} \neq \chi^{(3)}_{-1\,-1\,-1\,-1}$.

In isotropic media with point group $K_h$, in the absence of natural or magnetic gyration, no harmonic can be generated when using circularly polarized light [5]. This effect can be put to use for controlled second-harmonic generation applying external electric static or slowly variable fields [13] having components in the plane perpendicular to the direction of propagation of the laser beam. The polarisation $P(2\omega)$ amounts to:

$$P_1(2\omega) = \chi^{(3)}_{1\,1\,-1\,-1} E^{\omega_0}_{-1} E^{\omega}_{1} E^{\omega}_{1} = P_{-1}(2\omega)$$
$$= \chi^{(3)}_{-1\,-1\,1\,1} E^{\omega_0}_{1} E^{\omega}_{-1} E^{\omega}_{-1}, \tag{13}$$

where $E^{\omega_0}$ is the static ($\omega_0 = 0$) or slowly varying (at frequency $\omega_0 \ll \omega$) electric field strength. From eq. (13), the sense of polarisation of the second-harmonic wave is always opposite to that of the exciting wave. Whereas, if the field vector $E^{\omega_0}$ is parallel to the direction of propagation of the light wave, no second harmonic will be generated.

For laser light propagating along a fourfold axis (4, $\bar{4}$, $4/m$), the polarisation of the second harmonic in the presence of a field $E^{\omega_0}$ will be elliptical, since one has the nonzero components $P_1(2\omega)$ and $P_{-1}(2\omega)$;

$$P_1(2\omega) = \chi^{(3)}_{1111} E^{\omega_0}_{-1} E^{\omega}_{-1} E^{\omega}_{-1},$$
$$P_{-1}(2\omega) = \chi^{(3)}_{-1\,-1\,11} E^{\omega_0}_{1} E^{\omega}_{-1} E^{\omega}_{-1}. \tag{14}$$

By using an AC electric field, circularly polarized, the second harmonic will be circularly polarized too, in the sense opposite to $E^{\omega_0}$.

If a light wave propagates along a threefold or sixfold axis, it can generate a second harmonic, with sense of polarisation opposite to that of the incident wave, if an electric field acts perpendicularly to the direction of propagation of the exciting beam.

In the symmetry groups $D_{3h}$, $D_{3d}$, $D_3$, $C_{3v}$, $C_{3i}$ and $C_3$ generation is possible also, if the external electric field is applied along the direction of propagation of the laser beam which is at the same time the threefold axis of the crystal.

For crystals of lower symmetry or an arbitrary direction of propagation of the exciting beam in the crystal, the light generated will in general be ellipti-

* We henceforth omit the frequency dependence of the tensors $\chi^{(n)}$.

cally polarized. The state of polarisation of the second and third harmonic waves, generated by circularly polarized light, is given accordingly in tables II and III, where "none" stands for no effect of harmonics generation, "opp" denotes that the light generated is circularly polarized in the sense opposite to that of the incident wave, and "ellipt", that the light generated in the effect is polarized elliptically.

In the preceding considerations, in accordance with eqs. (9) and (10), we applied the electric-dipole approximation in the absence of electron dispersion and absorption, for reasons of simplicity. Generation of optical harmonics in absorbing media was the subject of a discussion by Bloembergen *et al.* [12]. Highly interesting results can be expected from generation experiments in the presence of spatial dispersion and magnetic gyration [14]. The problem will be given consideration in a separate paper.

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