LASER LIGHT INTENSITY DEPENDENT NONLINEAR SUSCEPTIBILITIES IN HARMONICS GENERATION PHENOMENA

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In phenomenological as well as molecular-statistical treatments, the nonlinear suscep-tibilities of crystals and amorphous bodies determined from harmonics generation are shown to depend explicitly on the laser light intensity. This dependence leads to a self-amplification of the harmonics generated which is especially strong in statistically inhomogeneous media, where saturation of optical reorientation of the microsystems can occur.

1. Introduction

In processes of DC electric field-induced second-harmonic generation (SHG), first observed by Terhune et al. [1], the tensor of second-order nonlinear susceptibility $\chi_{ijk}^{2\omega}(E^0)$ is a function of the field strength E^0 [2, 3]. This paper is aimed at showing that nonlinear susceptibility tensors, measured in processes of harmonics generation [4], are functions of the intensity of laser light. We previously [5] performed an analysis along these lines with regard to SHG in electrically polarized dipolar gases. Here, we shall consider the problem more generally for arbitrary isotropic bodies and crystals.

2. Phenomenological treatment

In a medium acted on by a sufficiently intense, vibrating electric field $E(t) = E_\omega \cos \omega t$, the vector of induced dipole polarization is a nonlinear function of the field; in the electric dipole approximation, we have

$$P(t) = \sum_{n=1}^{\infty} \chi^{(n)}[n]E^n(t) = \sum_{n=1}^{\infty} P^{(n)}(t), \tag{1}$$

where $\chi^{(n)}$ is the tensor of n-th order nonlinear susceptibility, and $[n]$ denotes n-fold con-traction of the $(n+1)$-rank tensor $\chi^{(n)}$ and the poly-vector E^n.

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Taking into account frequency-dependence in the expansion (1), we can resolve the n-th order polarization vector $P^{(n)}$ into an even part and an odd part [6]

$$P^{(2n)}(t) = \frac{1}{2^{2n}} \left\{ \binom{2n}{n} \chi^{(2n)}(0) [2n]E_\omega^{2n} \right.$$

$$\left. + 2 \sum_{s=0}^{n-1} \binom{2n}{s} \chi^{(2n)}(2n\omega - 2s\omega) [2n]E_\omega^{2n} \cos 2(n-s)\omega t \right\}, \qquad (2)$$

$$P^{(2n-1)}(t) = \frac{1}{2^{2n-2}} \sum_{s=0}^{n-1} \binom{2n-1}{s} \chi^{(2n-1)}(2n\omega - 2s\omega - \omega) [2n-1]E_\omega^{2n-1} \cos (2n-2s-1)\omega t.$$

$$(3)$$

The even-order polarization vector (2) contains the zero-frequency contribution corresponding to a DC polarization effect within the medium [7] and the frequency-dependent polarization responsible for the generation of even $2(n-s)$ harmonics from the medium. The odd-order polarization vector (3) consists only of frequency-dependent contributions corresponding to generation of odd $2(n-s-1)$ — harmonics. To-day, second, third and higher harmonics are obtained from various isotropic and anisotropic materials [4, 8].

By Eqs. (1)–(3), we have the following expression for the i-th component of dipole polarization vibrating at the fundamental frequency ω of incident light

$$P_i^\omega(I) = \chi_{ij}^\omega(I)E_j^\omega \cos \omega t, \qquad (4)$$

where

$$\chi_{ij}^\omega(I) = \chi_{ij}^\omega(0) + \tfrac{3}{2} \chi_{ijkl}^\omega I_{kl} + \tfrac{5}{2} \chi_{ijklmn}^\omega I_{kl}I_{mn} + \dots \qquad (5)$$

is the tensor of linear dipolar susceptibility dependent on the incident light intensity, given by the tensor $I_{kl} = E_k^\omega E_l^{-\omega}/2$. From existing observations, the light intensity-dependent polarization (4) is apparent in the optical Kerr effect [9], self-induced rotation of the polarization ellipse [10], and self-focussing of laser beams [11, 12].

Since the polarization of Eq. (4) depends on the incident intensity, polarization at doubled frequency can naturally be expected to be a function of I

$$P_i^{2\omega}(I) = \tfrac{1}{2} \chi_{ijk}^{2\omega}(I)E_j^\omega E_k^\omega \cos 2\omega t, \qquad (6)$$

where, with regard to Eq. (2),

$$\chi_{ijk}^{2\omega}(I) = \chi_{ijk}^{2\omega}(0) + 2\chi_{ijklm}^{2\omega}I_{lm} + \tfrac{15}{4} \chi_{ijklmno}^{2\omega}I_{lm}I_{no} + \dots \qquad (7)$$

is the tensor of second-order nonlinear susceptibility at 2ω in the presence of a self-induced variation of highly intense light.

Likewise, by (3), we obtain for polarization at tripled vibration frequency

$$P_i^{3\omega}(I) = \tfrac{1}{4} \chi_{ijkl}^{3\omega}(I)E_j^\omega E_k^\omega E_l^\omega \cos 3\omega t, \qquad (8)$$

where the light intensity-dependence of the tensor of third-order nonlinear susceptibility is given by the expansion

$$\chi_{ijkl}^{3\omega}(I) = \chi_{ijkl}^{3\omega}(0) + \tfrac{5}{2}\chi_{ijklmn}^{3\omega}I_{mn} + \tfrac{21}{4}\chi_{ijklmnop}^{3\omega}I_{mn}I_{op} + \cdots \tag{9}$$

Similarly, we obtain with regard to (1)–(3) polarizations responsible for harmonics higher than the third.

The dependence of higher-order susceptibilities on frequency dispersion and absorption and spatial dispersion are found in general by the perturbation methods of quantum mechanics [6, 7]. Also, the symmetry relations and non-zero and mutually independent elements of polar tensors of higher ranks can be derived by the methods of group theory, as it has been done for axial tensors [13]. This makes it possible to apply nonlinear variations of the susceptibility tensors (7) and (9) for all the classes of symmetry of materials investigated for harmonics generation. Though actually only the susceptibilities $\chi_{ijk}^{2\omega}$ and $\chi_{ijkl}^{3\omega}$ have been measured hitherto, there already are available laboratory conditions for the measurement of nonlinear susceptibilities of still higher orders [14]. Such measurements will permit to carry out numerical evaluations of the various light intensity-dependent terms occurring in the expansions (7) and (9).

3. Semi-macroscopic treatment

The dependence of the tensors (5), (7) and (9) on I can be proved as well by studies of their dependence on temperature, density, and concentration, and can be made apparent in a semi-macroscopic and molecular-statistical approach [15] particularly well adapted to the analysis of isotropic bodies. But here we come up against the fact that in the isotropic optically inactive case the tensor (7) vanishes, and SHG can occur only by way of electric quadrupole and magnetic dipole polarization mechanisms [7, 8] or if a DC electric field is applied externally [1–3].

Applying perturbation calculus with Gibbs statistics, we derive in the presence of a DC electric field E^0 and light of intensity I the following distribution function [11, 15]:

$$f(\tau, E^0, I) = f(\tau, 0)\,\{1 + \beta M_i E_i^0 + \tfrac{1}{2}\beta\Delta A_{ij}^\omega I_{ij}$$

$$+ \tfrac{1}{2}\beta(B_{ijk}^\omega + \beta M_i \Delta A_{jk}^\omega)E_i^0 I_{jk} + \cdots\}, \tag{10}$$

where $f(\tau, 0)$ is the distribution function of the microsystems at the configuration τ when $E^0 = 0$ and $I = 0$, and $\beta = (kT)^{-1}$. The total electric dipole moment of the medium is M_i; its variation under the influence of light of intensity I_{jk} is given by the tensor B_{ijk}^ω of nonlinear second-order electro-optical polarizability. The quantity $\Delta A_{ij}^\omega = A_{ij}^\omega - \langle A_{ij}^\omega \rangle$ describes the fluctuation of the linear optical polarizability tensor of the medium.

By having recourse to the distribution function (10), we obtain in a good approximation for a spherical sample of volume V:

$$\chi_{ijk}^{2\omega}(E^0, I) = \frac{1}{2V}\left(\frac{n_{2\omega}^2 + 2n_0^2}{3n_0^2}\right)\left(\frac{n_\omega^2 + 2n_0^2}{3n_0^2}\right)^2\Big\{\langle C_{ijkl}^{2\omega} + \beta B_{ijk}^{2\omega}M_l\rangle$$

$$\times \left(\frac{\varepsilon+2\varepsilon_0}{3\varepsilon_0}\right) E_l^0 + \tfrac{1}{6} \left\langle E_{ijklmn}^{2\omega} + 3\beta(B_{ijk}^{2\omega}B_{lmn}^{\omega} + C_{ijkl}^{2\omega}\Delta A_{mn}^{\omega})\right.$$

$$\left. + 3\beta^2 B_{ijk}^{2\omega}M_l\Delta A_{mn}^{\omega}\right\rangle \left(\frac{\varepsilon+2\varepsilon_0}{3\varepsilon_0}\right)\left(\frac{n_{\omega}^2+2n_0^2}{3n_0^2}\right)^2 E_l^0 I_{mn} + \dots \Bigg\} . \qquad (11)$$

Above, n_{ω} and $n_{2\omega}$ are, respectively, the refractive indices for the fundamental and doubled frequency and ε the electric permittivity of the sample; n_0 and ε_0 refer to the medium surrounding the spherical sample. When deriving Eq. (11) we had recourse to the circumstance that, in the case of isotropic optically inactive media, unweighted averaging causes polar tensors of odd rank to vanish: $\langle M_i \rangle = \langle B_{ijk} \rangle = \langle B_{ijk}A_{lm} \rangle = 0$.

The first term of the expansion (10) which is linear in E^0 was discussed exhaustively in previous papers [3]. The second term, which arises by superposition of the action of the field E^0 and light intensity I on the medium, depends on temperature likewise to the DC Kerr constant [15]; however, whereas Kerr's effect is described by tensors of rank 4, the interference effect in DC SHG is given by tensors of rank 6. The sixth-rank tensors of (11) have to be averaged isotropically according to previously established transformation formulae [16].

On applying the distribution function (10) with $E^0 = 0$, we obtain for the nonlinear third-order susceptibility tensor in the presence of intense light

$$\chi_{ijkl}^{3\omega}(I) = \frac{1}{6V}\left(\frac{n_{3\omega}^2+2n_0^2}{3n_0^2}\right)\left(\frac{n_{\omega}^2+2n_0^2}{3n_0^2}\right)^3$$

$$\times \left\{\langle C_{ijkl}^{3\omega}\rangle + \tfrac{1}{8}\left\langle E_{ijklmn}^{3\omega} + 4\beta C_{ijkl}^{3\omega}\Delta A_{mn}^{\omega}\right\rangle \left(\frac{n_{\omega}^2+2n_0^2}{3n_0^2}\right)^2 I_{mn} + \dots \right\} . \qquad (12)$$

The intensity-dependent term is of a form similar to that of the fourth-rank tensor of the optical Kerr effect [11].

4. Molecular-statistical treatment

The expressions (11) and (12) derived above are accessible to an analysis on the microscopic level permitting to gain insight into the various mechanisms leading to the generation of harmonics, thus processes of electron vibration anharmonicity [7] as well as of molecular reorientation and redistribution [3, 11, 15], other statistical-fluctuational processes, and molecular motions [17]. The tensors M_i, A_{ij}, B_{ijk}, ... depend, besides the properties of the atoms and molecules, on the fields F of the surrounding electric multipoles. Thus e.g. one has for the dipole moment of N microsystems present in V

$$M_i = \sum_{p=1}^{N} \left\{ m_i^{(p)} + a_{ij}^{(p)}F_j^{(p)} + \tfrac{1}{2} b_{ijk}^{(p)}F_j^{(p)}F_k^{(p)} + \tfrac{1}{6} c_{ijkl}^{(p)}F_j^{(p)}F_k^{(p)}F_l^{(p)} + \dots \right\} \qquad (13)$$

and similar expansions for the other tensors of Eqs. (11) and (12). Expansions of this kind, in conjunction with appropriately chosen statistical distribution functions, make

it possible to investigate the influence on harmonics generation of pairwise, triple, quadruple etc. correlations of the microsystems, besides that of their intrinsic properties. In this way, the susceptibilities (11) and (12) can be expressed as functions not only of temperature but also of the density, or concentration, of the systems under study [19].

Noteworthy progress has recently been achieved with regard to the numerical determination of nonlinear optical polarizabilities for atoms and simpler molecules [2, 18–21] as well as large organic molecules [22]. These data and the knowledge of other relevant molecular parameters will permit to evaluate numerically the variations induced in the susceptibility tensors (11) and (12) by the action of sufficiently intense light beams.

5. Saturation of optical molecular reorientation

The expressions (11) and (12) cover statistical processes implying a rather weak degree of optical reorientation of the microsystems. Closer studies of the self-trapping of intense laser beams have shown that strong optical reorientation of molecules including saturation (complete alignment) can be achieved. In the latter case, the calculations can be carried out to the end only if statistical molecular correlations are neglected.

Let us assume the intense laser beam incident on a system of mutually uncorrelated molecular dipoles to propagate in the direction of the Z-axis and to be plane polarized in that of the Y-axis, along which a DC electric field E^0 acts. On omitting nonlinear polarizabilities of higher orders, we obtain for axially symmetric molecules having the electric dipole moment m

$$\chi^{2\omega}_{yyy}(E^0_y, I_y) = \frac{Nm}{2kT} \{3b^{2\omega}_{113}L_2(\pm q) + (b^{2\omega}_{333} - 3b^{2\omega}_{113})L_4(\pm q)\}E^0_y. \tag{14}$$

Above, $L_2(\pm q)$ and $L_4(\pm q)$ are Langevin functions of the second and fourth order [5] of the optical reorientation parameter of the molecule $q = \frac{\beta}{2}|a^{\omega}_{33} - a^{\omega}_{11}|\left(\frac{n^2+2}{3}\right)^2 I_y$; the sign "+" refers to "cigar-like" molecules, with a positive anisotropy of linear polarizability $(a^{\omega}_{33} - a^{\omega}_{11} > 0)$, whereas the sign "–" is for "disc-shaped" ones with negative anisotropy $(a^{\omega}_{33} - a^{\omega}_{11} < 0)$.

If the molecular system under consideration is not acted on by an external electric field $(E^0 = 0)$, the susceptibility (14) vanishes and only third-harmonic generation with susceptibility of order 3 is possible (we neglect higher-order effects)

$$\chi^{3\omega}_{yyyy}(I_y) = \frac{N}{24V} \{c^{3\omega}_{1111} + 2(3^{3\omega}_{1133} - c^{3\omega}_{1111})L_2(\pm q)$$

$$+ (c^{3\omega}_{1111} - 6c^{3\omega}_{1133} + c^{3\omega}_{3333})L_4(\pm q)\}. \tag{15}$$

Eqs. (14) and (15) hold for weak as well as strong optical reorientation of the molecules.

In particular, for $I_y = 0$, i.e. in the absence of optical reorientation, $q = 0$, they reduce to

$$\chi_{yyy}^{2\omega}(E_y^0, 0) = \frac{NmE_y^0}{10kTV}(b_{333}^{2\omega} + 2b_{113}^{2\omega}), \tag{14a}$$

$$\chi_{yyyy}^{3\omega}(0) = \frac{N}{360V}(3c_{3333}^{3\omega} + 12c_{1133}^{3\omega} + 8c_{1111}^{3\omega}). \tag{15a}$$

For the case of complete optical alignment, i.e. optical saturation, we obtain by (14) and (15) for "cigar-like" molecules

$$\chi_{yyy}^{2\omega}(E_y^0, +\infty) = \frac{Nm}{2kTV}b_{333}^{2\omega}E_y^0, \tag{14b}$$

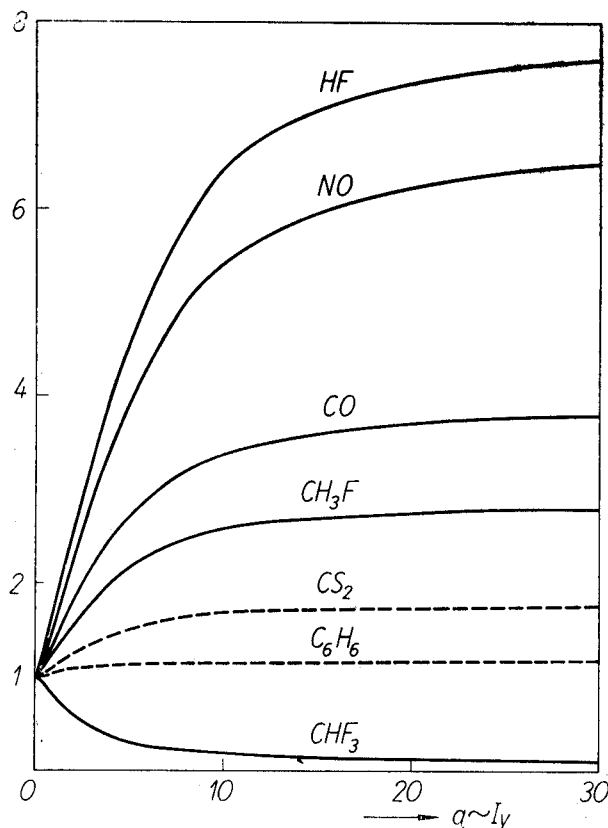$$\chi_{yyyy}^{3\omega}(+\infty) = \frac{N}{24V}c_{3333}^{3\omega}. \tag{15b}$$

Fig. 1. Relative changes in nonlinear susceptibilities due to strong optical reorientation of molecules. The full curves (HF, NO, CO, CH$_3$F, CHF$_3$) are for $\chi_{yyy}^{2\omega}(E_y^0, I_y)/\chi_{yyy}^{2\omega}(E_y^0, 0)$, the dashed ones (CS$_2$, C$_6H_6$) for $\chi_{yyyy}^{3\omega}(I_y)/\chi_{yyyy}^{3\omega}(0)$

With regard to the available literature data [8, 18–22] for the nonlinear polarizability tensor elements $b_{ijk}^{2\omega}$ and $c_{ijkl}^{3\omega}$, one is easily convinced on inspection of our formulae that in the case of "cigar-like" molecules the process of optical reorientation saturation enhances the susceptibilities (14) and (15) by at least one order of magnitude. As a consequence, the power of the harmonics generated can increase by two orders at the least.

The situation is different for "disc-like" molecules, since here the susceptibility of Eq. (14) can decrease to zero, whereas that of Eq. (15) takes the value

$$\chi_{yyyy}^{3\omega}(-\infty) = \frac{N}{24V}\,c_{1111}^{3\omega}, \tag{15c}$$

thus also leading to a considerable enhancement of the third harmonic.

On the assumption of $6c_{1133} = c_{3333} + c_{1111}$ we obtain, with regard to (15) and (15a), the relative change in nonlinear susceptibilities in the form

$$\frac{\chi_{yyyy}^{3\omega}(I_y)}{\chi_{yyyy}^{3\omega}(0)} = 1 + \frac{c_{3333}^{3\omega} - c_{1111}^{\omega}}{c_{3333}^{3\omega} + 2c_{1111}^{3\omega}}\,\{3L_2(\pm q) - 1\}, \tag{16}$$

i.e. a dependence on optical molecular reorientation identical with that of the Optical Kerr Effect [5] and self-trapping of light [23].

Fig. 1 shows the variations of Eq. (16) with increasing light intensity for CS_2 and C_6H_6 [8] and similar variations in the susceptibility ratio of Eqs. (14) and (14a) for HF, NO, CO, CH_3F and CHF_3 (the data for these molecules are taken from Ref. [24]).

6. Conclusions

We have seen that the tensors of higher nonlinear susceptibilities χ_{ijklm} and χ_{ijklmn} are accessible to determination indirectly from studies of the light intensity dependence of the 2-nd and 3-rd harmonics, respectively, and directly from generation of the 4-th and 5-th harmonics. The numbers of nonzero and mutually independent elements of these tensors for all crystallographical classes are given in Tables I and II (those of independent ones for some classes are to be found in Refs. [25, 26]). The tensor elements of χ_{ij}, χ_{ijk} and χ_{ijkl} are available from the literature [8, 27]. Those of totally symmetric tensors χ_{ijklm} and χ_{ijklmn} are given in Table II. These data permit to apply the light intensity dependent susceptibility tensors (5), (7) and (9) to the various crystallographic classes investigated experimentally in Nonlinear Optics.

Obviously, laser self-induced enhancement or abatement of the nonlinear susceptibility tensors plays a highly important part in light harmonics generation in crystals [4, 8], especially ones with structural phase transitions [28], in organic dye solutions [29], as well as in molecular, macromolecular and colloidal fluids [30]. In semiconductors, the increase in nonlinear susceptibilities is due essentially to other mechanisms [31–33].

TABLE I

The number of non-zero (N) and independent (I) elements of the nonlinear susceptibility tensor χ_{ijklm}. Symmetricity in indices is denoted by parentheses $(j...)$.

Class	χ_{ijklm} N	χ_{ijklm} I	$\chi_{i(jkl)m}$ N	$\chi_{i(jkl)m}$ I	$\chi_{i(jklm)}$ N	$\chi_{i(jklm)}$ I
$1(C_1)$	243	243	243	90	243	45
$\bar{1}(C_i)$	0	0	0	0	0	0
$m(C_s)$	122	122	122	46	122	24
$2(C_2)$	121	121	121	44	121	21
$2/m(C_{2h})$	0	0	0	0	0	0
$222(D_2)$	60	60	60	21	60	9
$mm2(C_{2v})$	61	61	61	23	61	12
$mmm(D_{2h})$	0	0	0	0	0	0
$4(C_4)$	121	61	115	22	109	11
$\bar{4}(S_4)$	120	60	120	22	114	10
$4/m(C_{4h})$	0	0	0	0	0	0
$422(D_4)$	60	30	54	10	48	4
$4mm(C_{4v})$	61	31	61	12	61	7
$\bar{4}2m(D_{2d})$	60	30	60	11	60	5
$4/mmm(D_{4h})$	0	0	0	0	0	0
$3(C_3)$	233	81	227	30	213	15
$\bar{3}(S_6)$	0	0	0	0	0	0
$32(D_3)$	116	40	110	14	96	6
$3m(C_{3v})$	117	41	117	16	117	9
$\bar{3}m(D_{3d})$	0	0	0	0	0	0
$6(C_6)$	121	51	115	16	101	7
$\bar{6}(C_{3h})$	112	30	112	14	112	8
$6/m(C_{6h})$	0	0	0	0	0	0
$622(D_6)$	60	25	54	7	40	2
$6mm(C_{6v})$	61	26	61	9	61	5
$\bar{6}m2(D_{3h})$	56	15	56	7	56	4
$6/mmm(D_{6h})$	0	0	0	0	0	0
$23(T)$	60	20	60	7	60	3
$m3(T_h)$	0	0	0	0	0	0
$432(O)$	60	10	42	3	24	1
$\bar{4}3m(T_d)$	60	10	60	4	60	2
$m3m(O_h)$	0	0	0	0	0	0
Y	60	6	42	1	0	0
Y_h	0	0	0	0	0	0
K	60	6	42	1	0	0
K_h	0	0	0	0	0	0
$\infty(C_\infty)$	121	51	115	16	101	7
$\infty m(C_{\infty v})$	61	26	61	9	61	5
$\infty/m(C_{\infty h})$	0	0	0	0	0	0
$\infty/mmm(D_{\infty h})$	0	0	0	0	0	0

TABLE II

Non-zero (N) and independent (I) elements of the tensors $\chi_{(ijklm)}$ and $\chi_{(ijklmn)}$ symmetric in all indices. The tensor elements are denoted by their subscripts x, y, z only; the number of elements resulting by permutation of indices is given in parentheses ( ). In order to reduce space, elements shared by various classes have been assembled under one capital letter

Class	$\chi_{(ijklm)}$ N	I	Elements	$\chi_{(ijklmn)}$ N	I	Elements
$1(C_1)$	243	21	$A \equiv xxxxyz(20),\ xyyyz(20),\ xyzzz(20);$ $B \equiv zzzzz,\ xxxxz(5),\ yyyyz(5),\ xxyyz(30),\ xxzzz(10),\ yyzzz(10);$ $D \equiv xxxxx,\ yyyyy,\ xzzzz(5),\ xxxxy(5),\ xyyyy(5),\ yzzzz(5),\ xxyyy(30),\ xxxzz(10),\ xxxyy(10),\ xyyzz(30),\ xxxyy(10),\ yyyzz(10);$	729	28	$A_1 \equiv zzzzzz,\ xxxxxx,\ yyyyyy,\ xxxxyy(15),\ xxxxzz(15),\ xxxyyz(15),\ xxzzzz(15),\ yyzzzz(15),\ yyyyzz(15),\ xxyyzz(90);$ $B_1 \equiv xxxxxy(6),\ xyyyyy(6),\ xxxyyy(20),\ xxxyzz(60),\ xxyyyz(60),\ xyyzzz(30);$ $D_1 \equiv xxxxxz(6),\ yyyyyz(6),\ xzzzzz(6),\ yzzzzz(6),\ xxxxyz(20),\ yyyyxz(20),\ xxxyzz(60),\ xxyyyz(60),\ xyyyzz(60),\ xxyyzz(60),\ xyyyzz(30),\ xxxxyz(30);$
$\bar{1}(C_i)$	0	0		729	28	$A_1,\ B_1$ and D_1
$m(C_s)$	122	12	D	365	16	A_1 and B_1
$2(C_2)$	121	9	A and B	365	16	A_1 and B_1
$2/m(C_{2h})$	0	0		365	16	A_1 and B_1
$222(D_2)$	60	3	A	183	10	A_1
$mm2(C_{2v})$	61	6	B	183	10	A_1
$mmm(D_{2h})$	0	0		183	10	A_1
$4(C_4)$	101	5	$E \equiv xxxyz = -yyyxz(20);$ $F \equiv zzzzz,\ xxxyz(30),\ xxxzz = yyyzz(10),\ xxxxz = yyyyz(5);$	315	8	$E_1 \equiv zzzzzz,\ xxxxxx = yyyyyy,\ xxyyzz(90),\ xxxxyy = yyyyxx(15),\ xxxxzz = yyyyzz(15),\ xxzzzz = yyzzzz(15);$ $F_1 \equiv xxxxxy = -yyyyyx(6),\ xxxyzz = -yyyxzz(60);$
$\bar{4}(S_4)$	90	4	$E' \equiv xyzzz(20),\ xxxyz = yyyxz(20),$ $F' \equiv xxxzz = -yyzzz(10),\ xxxxz = -yyyyz(5);$	315	8	E_1 and F_1

TABLE II (continued)

Class	χ$_{(ijklm)}$			χ$_{(ijklmm)}$		
	N	I	Elements	N	I	Elements
4/m(C$_{4h}$)	0	0		315	8	E_1 and F_1
422(D$_4$)	40	1	E	183	6	E_1
4mm(C$_{4v}$)	61	4	F	183	6	E_1
4̄2m(D$_{2d}$)	60	2	E'	183	6	E_1
4/mmm(D$_{4h}$)	0	0		183	6	E_1
3(C$_3$)	173	7	$H \equiv xxxxxx = -\frac{5}{3}\,xxyyyy,$ $xxxxyy = \frac{1}{3}\,xyyyy,$ $xxxxzz = -xyyzz(20);$ $J \equiv zzzzz,\ xxzzz = yyzzz = 3xxyyz(10),$ $xxxxxz = yyyyz = 3xxyyz;$ $L \equiv yyyyy = -\frac{5}{3}\,xxxxy,$ $xxyyy = \frac{1}{3}\,xxxxy,$ $yyyzz = -xxyzz(20);$	567	10	$G_1 \equiv xxxxxx = 2xxxxyy+3xxyyyy,$ $yyyyyy = 3xxxxyy+2xxyyyy,$ $zzzzzz,\ xxzzzz = yyzzzz,$ $xxxxzz = yyyyzz = 3xxyyzz;$ $H_1 \equiv xxxxxz = -\frac{5}{3}\,xyyyyz,$ $xxxyyz = \frac{1}{3}\,xyyyyz,$ $xxxzzz = -xyyzzz;$ $J_1 \equiv yyyyyz = -\frac{5}{3}\,xxxxyz,$ $xxyyyz = \frac{1}{3}\,xxxxyz,$ $yyyzzz = -xxyzzz;$ $L_1 \equiv xxxxxy = xyyyyy,$ $xxxyyy = -xyyyyy;$
3̄(S$_6$)	0	0		567	10	$G_1,\ H_1,\ J_1$ and L_1
32(D$_3$)	56	2	H	359	7	G_1 and J_1
3m(C$_{3v}$)	117	5	J and L	359	7	G_1 and J_1
3̄m(D$_{3d}$)	0	0		359	7	G_1 and J_1
6(C$_6$)	61	3	J	215	6	G_1 and L_1
6̄(C$_{3h}$)	112	4	H and L	215	6	G_1 and L_1
6/m(C$_{6h}$)	0	0		215	6	G_1 and L_1
622(D$_6$)	0	0		183	5	G_1
6mm(C$_{6v}$)	61	3	J	183	5	G_1
6̄m2(D$_{3h}$)	56	2	H	183	5	G_1
6/mmm(D$_{6h}$)	0	0		183	5	G_1
∞(C$_{\infty}$)	61	3	J	183	4	$M_1 \equiv zzzzzz,\ xxxxxz = yyyyzz = 3xxyyzz,$ $xxzzzz = yyzzzz;$ $xxxxxx = yyyyyy = 5xxxxyy = 5xxyyyy;$

TABLE II (continued)

Class	$\chi_{(ijklm)}$			$\chi_{(ijklmn)}$		
	N	I	Elements	N	I	Elements
$\infty m(C_{\infty v})$	61	3	J	183	4	M_1
$\infty\|m(C_{\infty h})$	0	0		183	4	M_1
$\infty\|mm(D_{\infty h})$				183	4	M_1
$23(T)$	60	1	$M \equiv xxxyz = yyyxz = zzzxy(20)$;	183	4	$P \equiv xxxxxx = yyyyyy = zzzzzz,\ xxyyzz(90)$; $Q \equiv xxyyyy = yyzzzz = xxxxzz(15)$; $R \equiv xxxxyy = yyyyzz = xxzzzz(15)$.
$m3(T_h)$	0	0		183	4	$P,\ Q$ and R
$432(O)$	0	0		183	3	P and $Q = R$
$m3m(O_h)$	0	0		183	3	P and $Q = R$
$\bar{4}3m(T_d)$	60	1	M	183	3	P and $Q = R$

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