DC MAGNETIC FIELD-INDUCED SECOND HARMONIC GENERATION OF LASER BEAM

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Second harmonic generation (SHG) in the presence of a dc magnetic field H^0 is described by a new axial tensor of third-order non-linear susceptibility $\mathcal{H}_{ijkl}(-2\omega, \omega, \omega, 0)$. Relations between its non-zero elements are derived by group theory for all crystallographical classes. In classes $422(D_4)$, $622(D_6)$, $\bar{4}32(O)$, Y and K a magnetic field induces SHG, which at $H^0 = 0$ is absent in electric-dipole approximation.

One of us [1] pointed to the possibility of studying dc magnetic field-induced sum frequency generation of laser beams. This communication contains a detailed analysis of a particular case of frequency mixing, namely SHG, for all crystallographical classes, especially those for which field-less SHG is forbidden in electric-dipole approximation.

Phenomenologically, the process of frequency mixing of two laser beams with electric vectors $E(\omega_1, k_1)$ and $E(\omega_2, k_2)$ (frequencies ω_i, wave numbers k_i) in the presence of a dc magnetic field $H(0)$ is described by the third-order dipole polarization vector [1]:

$$P_i(\omega_3, k_3) = \mathcal{H}_{ijkl}(-\omega_3, \omega_1, \omega_2, 0)\, E_j(\omega_1, k_1) \times E_k(\omega_2, k_2) H_l(0) \;, \tag{1}$$

where $\omega_3 = \omega_1 + \omega_2$ and $k_3 = k_1 + k_2 + \Delta k$ define time and space synchronisation conditions.

The fourth-rank axial tensor $\mathcal{H}_{ijkl}(-\omega_3, \omega_1, \omega_2, 0)$ describes the variation induced in the natural non-linear susceptibility $\chi_{ijk}(-\omega_3, \omega_1, \omega_2)$ by a dc linear and uniform magnetic field.

By (1), the dc magnetic field induced SHG is accounted for by the polarization $P_i^{2\omega}$, which in simplified form is:

$$P_i^{2\omega} = \mathcal{H}_{ijkl}^{2\omega}\, E_j^{\omega} E_k^{\omega} H_l^0 \;. \tag{2}$$

By a method based on group theory [2], relations are derived between non-zero elements of the axial tensor $\mathcal{H}_{ijkl}^{2\omega}(-2\omega, \omega, \omega, 0)$ which, in the case of a dispersional medium, is symmetric only in the indices j and k and, on neglecting electronic dispersion, is symmetric in i, j, k. The number of independent tensor elements for the group $G\{g_1, g_2, \ldots, g_N\}$ is given by the formula [3]:

$$I = (1/N) \sum_{g_i \in G} \chi^{0}(g_i)\, \varkappa(g_i) \;, \tag{3}$$

where

$$\chi^{0}(g_i) = \tfrac{1}{6}\left[\chi^4(g_i) + 2\chi^2(g_i)\chi(g_i^2) + 3\chi(g_i)\chi(g_i^3)\right] \tag{4}$$

and

$$\varkappa(g_i) = 1 \text{ for proper rotation } g_i = c_\varphi \;; \quad = -1 \text{ for improper rotation } g_i = s_\varphi \;, \tag{5}$$

are characters of the element g_i respectively in tensor representation taking into consideration the permutational group of indices $P^3(i, j, k)$ and in the irreducible unit (antisymmetric) vector representation of the group G, whose character is denoted by $\chi(g_i)$. Summation extends over all N elements of the group. The linear combinations of tensor elements belonging to the various irreducible representations of group G were determined by means of Wigner's projection operator [4]:

$$\hat{P} = (f_\alpha/N) \sum_{g_i \in G} \chi^{\alpha}(g_i)\, \hat{g}_i \;, \tag{6}$$

where f_α is the dimension of the irreducible representation α of the group G and $\chi^{\alpha}(g_i)$ the character of element g_i in this representation.

The transformational properties of the tensor $\mathcal{H}_{ijkl}^{2\omega}$ were found from eqs. (3) and (6) and served for calculating relations between its non-zero elements. These are readily obtained by perform-

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Table 1

Non-zero and independent elements of the axial tensor $\mathcal{H}_{ijkl}(-2\omega, \omega, \omega, 0)$ for all crystallographical classes. The tensor elements are denoted by their subscripts x, y, z only. N signifies the number of non-zero elements, I that of independent elements. In order to reduce space, elements shared by various classes have been assembled under one capital letter

class C_1 $N = 81,\ I = 54$

$A \equiv xxxx,\ yyyy,\ zzzz,\ xxyy = xyxy,\ yyxx = yxyx,$
$xyyx,\ yxxy,\ xzzx,\ xxzz = xzxz,\ yyzz = yzyz,$
$zxxz,\ zyyz,\ yzzy,\ zzxx = zxzx,\ zzyy = zyzy,$

$B \equiv xxxy,\ yyyx,\ yxxx,\ xxyx = xyxx,\ yyxy = yxyy,$
$xyyy,\ xzzy,\ yzzx,\ zzxy = zxzy,\ zzyx = zyzx,$
$zxyz = zyxz,\ xyzz = xzyz,\ yxzz = yzxz,$

$C \equiv zzzx,\ zzzy,\ xzzz,\ zzxz = zxzz,\ zzyz = zyzz,$
$yzzz,\ xxxz,\ yyyz,\ xxzx = xzxx,\ yyzy = yzyy,$
$zxxx,\ zyyy,\ yxxz,\ xxyz = xyxz,\ xxzy = xzxy,$
$zxxy,\ xyyz,\ zyyx,\ xyzx = xzyx,\ yzxx = yxzx,$
$zyxx = zxyx,\ yyxz = yxyz,\ yyzx = yzyx,\ yxzy = yzxy,$
$xzyy = xyzy,\ zxyy = zyxy,$

class C_2	$N = 41,\ I = 28$	elements A and B
class C_s	$N = 40,\ I = 26$	elements C
class D_2	$N = 21,\ I = 15$	elements A
class C_{2v}	$N = 20,\ I = 13$	elements B
class C_4	$N = 39,\ I = 14$	elements D and E_+ and F_- a)
class S_4	$N = 40,\ I = 14$	elements E_- and F_+ and G
class D_4	$N = 21,\ I = 8$	elements D and E_+
class C_{4v}	$N = 18,\ I = 6$	elements F_-
class D_{2d}	$N = 20,\ I = 7$	elements E_-

where $D \equiv zzzz,$

$E_{\pm} \equiv xxxx = \pm yyyy,\ xxyy = xyxy = \pm yyxx = \pm yxyx,\ xyyx = \pm yxxy,$
$xzzx = \pm yzzy,\ xxzz = xzxz = \pm yyzz = \pm yzyz,\ zxxz = \pm zyyz,$
$zzxx = zxzx = \pm zzyy = \pm zyzy,$

$F_{\pm} \equiv xxyx = xyxx = \pm yyxy = \pm yxyy,\ xxxy = \pm yyyx,\ xyyy = \pm yxxx,$
$zzxy = zxzy = \pm zzyx = \pm zyzx,\ xzzy = \pm yzzx,$
$xyzz = xzyz = \pm yxzz = \pm yzxz,$

$G \equiv zxyz = zyxz,$

class C_3 $N = 71,\ I = 18$

$H \equiv zzzz,\ xxxx = yyyy = 2xxyy + xyyx,\ xxyy = xyxy = yyxx = yxyx,$
$xxzz = xzxz = yyzz = yzyz,\ zzxx = zxzx = zzyy = zyzy,$
$xyyx = yxxy,\ xzzx = yzzy,\ zxxz = zyyz,$

$K \equiv xxyx = xyxx = -yyxy = -yxyy,\ xyyy = -yxxx,$
$xxxy = -yyyx = 2yyxy + xyyy,\ zzxy = zxzy = -zzyx = -zyzx,$
$xyzz = xzyz = -yxzz = -yzxz,\ xzzy = -yzzx,$

$L \equiv xxxz = -yyxz = -xyyz = -yxyz,\ zxxx = -zxyy = -zyyx = -zyxy,$
$xxzx = xzxx = -yyzx = -yzyx = -xyzy = -xzyy = -yxzy = -yzxy,$

$M \equiv yyyz = -xxyz = -yxxz = -xyxz,\ zyyy = -zyxx = -zxxy = -zxyx,$
$yyzy = yzyy = -xxzy = -yxzx = -xyzx = -xzyx = -xzxy = -yzxx,$

class D_3	$N = 37,\ I = 10$	elements H and M
class C_{3v}	$N = 34,\ I = 8$	elements K and L
class C_6	$N = 39,\ I = 12$	elements H and K
class C_{3h}	$N = 32,\ I = 6$	elements L and M
class D_6	$N = 21,\ I = 7$	elements H
class D_{3h}	$N = 16,\ I = 3$	elements M
class C_{6v}	$N = 18,\ I = 5$	elements K
class T	$N = 21,\ I = 5$	

a) Note that a capital letter with "+" or "-" signifies that the tensor elements have to be taken positive or negative, respectively.

Table 1 (continued)

$P \equiv xxxx = yyyy = zzzz,$
$Q \equiv xxyy = xyxy = zzxx = zxzx = yyzz = yzyz,$
$R \equiv yyxx = yxyx = xxzz = xzxz = zzyy = zyzy,$
$S \equiv xyyx = zxxz = yzzy,$
$V \equiv yxxy = xzzx = zyyz,$

class O	$N = 21,\ I = 3$	elements P and $Q = R$ and $S = V$
class T_d	$N = 18,\ I = 2$	elements $Q = -R$ and $S = -V$
class Y and K	$N = 21,\ I = 2$	elements $Q = R$ and $S = V$ $zzzz = 2xxyy + xyyx$.

In the remaining crystallographical classes, all elements vanish.

ing permutations in the results derived in [2,5] for the fourth-rank polar tensor with no permutational symmetry. These relations, for all crystallographical classes, are assembled in table 1. From table 1, a dc magnetic field is found to induce SHG in linear approximation in bodies with symmetry of the classes 422(D_4), 622(D_6), $\bar{4}32$(O) as well as Y and K, since in the absence of a field such bodies fail to generate SH in electric-dipole approximation. If the laser beam propagates along z, we have with (2) and the class 422(D_4) elements of table 1:

$$P_x^{2\omega} = (\mathcal{H}_{xxxx}^{2\omega} E_x^{\omega} E_x^{\omega} + \mathcal{H}_{xyyx}^{2\omega} E_y^{\omega} E_y^{\omega}) H_x^0 + 2\mathcal{H}_{xxyy}^{2\omega} E_x^{\omega} E_y^{\omega} H_y^0 ,$$

$$P_y^{2\omega} = (\mathcal{H}_{yyyy}^{2\omega} E_y^{\omega} E_y^{\omega} + \mathcal{H}_{xyyx}^{2\omega} E_x^{\omega} E_x^{\omega}) H_y^0 + 2\mathcal{H}_{xxyy}^{2\omega} E_x^{\omega} E_y^{\omega} H_x^0 ,$$

$$P_z^{2\omega} = 2\mathcal{H}_{zxxz}^{2\omega} (E_x^{\omega} E_x^{\omega} + E_y^{\omega} E_y^{\omega}) H_z^0 . \qquad (7)$$

If the dc magnetic field is perpendicular to wave propagation, eqs. (7) become analogous to those for dc electric field-induced SHG in isotropic media [6,7]. Obviously, in the absence of dispersion, by Kleinman's symmetry conjecture the 5 independent elements in (7) reduce to 3, since

$$\mathcal{H}_{xxxx}^{2\omega} = \mathcal{H}_{yyyy}^{2\omega},\ \mathcal{H}_{xyyx}^{2\omega} = \mathcal{H}_{xxyy}^{2\omega} .$$

To class 422 belong the crystals: potassium trichloro-acetate (CCl_3, COOK·CCl_3·COOH), guanidine carbonate ($2CNH(NH_2)_2 \cdot H_2CO_3$), hydrated nickel sulphate ($NiSO_4 \cdot 6H_2O$), malite ($Al_2C_{12}O_{12} \cdot 18H_2O$) and others where predictably SHG can be induced by applying a dc magnetic field.

In the case of class 622(D_6), one has to put in (7)

$$\mathcal{H}_{xxxx}^{2\omega} = \mathcal{H}_{yyyy}^{2\omega} = 2\mathcal{H}_{xxyy}^{2\omega} + \mathcal{H}_{xyyx}^{2\omega} ,$$

reducing the problem to 3 independent elements in the presence of electron dispersion, and to 2 in its absence. Here belong the crystals: lithium iodate ($LiIO_3$), barium aluminate ($BaAl_2O_4$) etc., where SHG is predictably induced by a dc magnetic field in dipole approximation.

For class $\bar{4}32$(O), one has to put in (7)

$$\mathcal{H}_{xxxx}^{2\omega} = \mathcal{H}_{yyyy}^{2\omega} \text{ and } \mathcal{H}_{xyyx}^{2\omega} = \mathcal{H}_{zxxz}^{2\omega} .$$

It is noteworthy in this respect that, as yet, it has not been determined beyond doubt whether ammonium chloride (NH_4Cl) and cuprite (Cu_2O) crystallize in class $\bar{4}32$(O) or m3m(O_h). Now, since the tensor $\mathcal{H}_{ijkl}^{2\omega}$ possesses non-zero elements in class $\bar{4}32$ only, it should be possible to reach a decision by studying dc magnetic field-induced SHG in these compounds.

Thus, experimental studies of dc magnetic and electric field induced SHG will become an effective method of deciding the crystallographic class of bodies, the symmetry of which cannot be determined by other methods. The non-centrosymmetric classes are predicted to exhibit a considerable rise in dipole SHG intensity in function of the squared magnetic field strength H^0.

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