

ON OPTICAL ROTATION INDUCED IN OPTICALLY INACTIVE CRYSTALS BY CROSSED ELECTRIC AND MAGNETIC FIELDS

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Optically inactive crystals of the classes $4mm$, $3m$, $6mm$ and $\bar{4}3m$ when placed in crossed electric and magnetic fields gain the ability to rotate the plane of polarization proportionally to $\chi_{ijkl}^{oem} E_k H_l$. Non-zero elements of the axial tensor χ_{ijkl}^{oem} for all crystallographical classes are derived by group theory.

Effects of optical change due to the concomitant action of fields E and H obviously require a nonlinear approximation. Already in the second approximation of variations in frequency-dependent electric permittivity tensor

$$\Delta \varepsilon_{ij}^{(2)}(\omega) = 2\pi \{ \chi_{ijkl}^{oeoe} E_k E_l + \chi_{ijkl}^{oem} E_k H_l + \chi_{ijkl}^{ome} H_k E_l + \chi_{ijkl}^{ommm} H_k H_l \}, \quad (1)$$

we have a cross-effect. The first term involving the tensor of nonlinear electro-optical susceptibility χ_{ijkl}^{oeoe} defines electrostriction and quadratic Kerr effect; the last term with the tensor of nonlinear magneto optical susceptibility χ_{ijkl}^{ommm} defines magnetostriction and quadratic Cotton-Mouton effect. The cross-effect resides in the mixed terms of Eq. (1), where the axial fourth-rank tensors χ_{ijkl}^{oem} and χ_{ijkl}^{ome} account for the optical variations induced in the crystal by the fields E and H concomitantly.

By known methods of group theory [1-3] we derive, similarly to our earlier work [4], relations between the non-zero elements of the axial tensor χ_{ijkl}^{oem} and tensor $\chi_{ijkl}^{ome} = +\chi_{ijlk}^{oem}$ (antisymmetric in their indices i and j , since one has $\chi_{ijkl}^{ome} = -\chi_{jikl}^{ome}$). Table I lists the non-zero elements of the tensor χ_{ijkl}^{oem} for all crystallographical classes.

Let the analyzing light wave propagate along the z -axis; then $\Delta \varepsilon_{xx}^{\omega} - \Delta \varepsilon_{yy}^{\omega}$ yields optical birefringence and $\Delta \varepsilon_{xy}^{\omega} - \Delta \varepsilon_{yx}^{\omega}$ optical rotation. From the antisymmetry of χ_{ijkl}^{oem} , optical birefringence cannot arise by way of the mixed terms EH but only through the

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Non-zero and independent elements of the axial tensor χ_{ijkl}^{oem} for all crystallographical classes

Class	Number of non-zero elements	Number of independent elements	Elements of χ_{ijkl}^{oem} are denoted only by their subscripts $i, j, k, l = x, y, z$
1 (C_1)	54	27	$A \equiv xyyx = -yxyx, yxxy = -xyxy, xzzx = -zxzx,$ $yzzy = -zyzy, xzxz = -zxxz, yzyz = -zyyz;$ $B \equiv xyxx = -yxxx, yxyy = -xyyy,$ $xzzy = -zxzy, yzzx = -zyzx,$ $xyzz = -yxzz, xzyz = -zxyz, yzxx = -zyxz;$ $C \equiv xzxx = -zxzx, xzzz = -zxzz,$ $xyyz = -yxyz, xyzy = -yxzy, xzyy = -zxyy,$ $yzyx = -zyyx, yzxy = -zyxy,$ $D \equiv yzyy = -zyyy, yzzz = -zyzz,$ $xyxz = -yxxz, xyzx = -yxzx,$ $xzxy = -zxxy, xzyx = -zxyx, yzxx = -zyxx,$
2 (C_2)	26	13	A, B
m (C_s)	28	14	C, D
222 (D_2)	12	6	A
$mm2$ (C_{2v})	14	7	B
4 (C_4) 6 (C_6)	26	7	$E \equiv xyyx = -yxyx = yxxy = -xyxy,$ $xzzx = -zxzx = yzzy = -zyzy,$ $xzxz = -zxxz = yzyz = -zyyz;$ $F \equiv xyxx = -yxxx = xyyy = -yxyy,$ $xyzz = -yxzz;$ $G \equiv xzzy = -zxzy = zyzx = -yzzx,$ $xzyz = -zxyz = zyxz = -yzxz.$
$\bar{4}$ (S_4)	24	6	$H \equiv xyyx = -yxyx = -yxxy = xyxy,$ $xzzx = -zxzx = -yzzy = zyzy,$ $xzxz = -zxxz = -yzyz = zyyz;$ $I \equiv xyxx = -yxxx = -xyyy = yxyy;$ $J \equiv xzzy = -zxzy = -zyzx = yzzx,$ $xzyz = -zxyz = -zyxz = yxz.$
422 (D_4) 622 (D_6)	12	3	E
4mm (C_{4v}) 6mm (C_{6v})	14	4	F, G
$\bar{4}2m$ (D_{2d})	12	3	H
3 (C_3)	42	9	$E, F, G,$ $K \equiv xzxx = -zxxx = -yzyx = zyyx =$ $= -yzxy = zyxy = -xzyy = zxyy;$

(continued) TABLE I

Class	Number of non-zero elements	Number of independent elements	Elements of χ_{ijkl}^{oem} are denoted only by their subscripts $i, j, k, l = x, y, z$
			$L \equiv yzyy = -zyyy = -xzxy = zxyx =$ $= -xzyx = zxyx = -yzxx = zyxx.$
32 (D_3)	20	4	E, L
3m (C_{3v})	22	5	F, G, K
$\bar{6}$ (C_{3h})	16	2	K, L
$\bar{6}m2$ (D_{3h})	8	1	L
23 (T)	12	2	$M \equiv xyyx = -yxyx = zxxz =$ $= -xzxz = yzzy = -zyzy,$ $N \equiv yxxy = -xyxy = xzzx =$ $= -zxzx = zyyz = -zyyz.$
$\bar{4}3m$ (T_d)	12	1	$M = -N$
432 (O) Y, K	12	1	$M = N$

square terms in E^2 and H^2 . Consequently, a cross-effect of the EH -type will cause only optical rotation, given by Eq. (1) as follows:

$$\Delta\epsilon_{xy}^{(2)}(\omega) - \Delta\epsilon_{yx}^{(2)}(\omega) = 8\pi\chi_{xykl}^{oem}E_kH_l. \quad (2)$$

(i) Crossed fields. Experimentally most favourably is the setup with fields E and H crossed, E acting along x and H along y (Voigt configuration). Table I shows the effect to be observable in the majority of crystallographical classes including, most strikingly, some of higher symmetries starting from classes $Y, K, 432, \bar{4}3m, 622, 6, 32$ etc., where Eq. (2) permits direct determinations of the element $\chi_{xyxy}^{oem} = -\chi_{yxyx}^{oem}$.

There is yet another experimental possibility, with fields E and H crossed and light propagating along H (Faraday configuration). From Table I, this effect can occur only in crystals of low symmetry, consequently in classes 1 and m .

(ii) Parallel fields. The setup with E and H parallel, though experimentally somewhat less convenient, is feasible in special conditions. In Faraday's configuration, this effect appear in many classes: 1, 2, $mm2, 4, 4mm, 3, 3m, 6$ and $6mm$ where we have also the alternative with fields E and H mutually parallel but perpendicular to light propagation (Voigt configuration).

The foregoing analysis points to the possibility of observing optical rotation induced by crossed electric and magnetic fields in crystals without natural optical activity belonging to the classes $4mm, 3m, 6mm, \bar{4}3m$, as well as Y and K . In the remaining classes, the

simultaneous action of fields E and H affects the natural optical activity, in some liquid crystals considerably. We are now attempting to detect experimentally similar cross-effects in isotropic, molecular and macromolecular [5] substances.

REFERENCES

- [1] G. Ya. Lyubarskii, *The Application of Group Theory in Physics*, Pergamon Press, Oxford 1960.
- [2] R. R. Birss, *Symmetry and Magnetism*, North-Holland Publishing Co., Amsterdam 1964; S. Bhagavantam, *Crystal Symmetry and Physical Properties*, Academic Press, London 1966.
- [3] L. L. Boyle, *Int. J. Quantum Chem.*, **3**, 231 (1969); **4**, 413 (1970) and references therein.
- [4] S. Kielich, R. Zawodny, *Optics Communications*, **4**, 132 (1971).
- [5] S. Kielich, *Appl. Phys. Letters*, **13**, 152 (1968); *J. Colloid Interface Sci.*, **30**, 159 (1969).