Applications of electrooptic materials in laser research*

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Abstract. In the present communication, a comprehensive and systematic survey of the need of the electrooptic devices, its underlying physics and dependence on the material aspects is presented. Principles are outlined for the development of materials having no centre of symmetry, and for second harmonic generation as a tool for testing such an accentricity. After grouping some important electrooptic crystals, the material properties are related to the electrooptic modulator design parameters and some typical crystals are discussed in the light of these relations. In conclusion, the problem areas in electrooptics are indicated and the relevance of such investigations to the national scene is discussed.

1. Introduction

The rapid development of laser technology has hastened the possibility of using laser systems for a variety of applications. For sometime, due to nonavailability of peripheral materials and devices like modulators, deflectors, Q-switches and harmonic generators, significant progress could not be achieved. In recent years, a host of investigators, particularly from USA, USSR and Japan have initiated the preparation, characterisation and subsequent utilisation of these materials for device applications. These studies were on such a scale that numerous industries have commercialised many of these devices. In India, though laser technology has been taken up even on commercial level, the technology of peripheral materials and devices is meagre, however.

Most of the devices mentioned above work on the principle of modulation of laser beam. At optical frequencies the modulation can be achieved by rapidly altering the propagation characteristics, i.e., the velocity through the medium. One way of doing this is by changing the index of propagation of a material at a rapid rate as the light passes through it. The refractive index of a material can be changed by applying an electric, acoustic or magnetic field (electrooptic, acoustooptic or magnetooptic effects respectively). In the present discussion, the authors confine themselves to the electrooptic effects only. The other effects

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will be presented elsewhere. In the past a few review articles and books (Kaminow 1966, Milek and Neuberger 1972, Wemple and DiDomenico 1972, Kaminow 1974, Hartfield and Thompson 1978 and Narasimhamurty 1981) have appeared on the subject, each one dealing with a particular aspect like physics and description of the effect, material properties or device fabrication. Other books include the application of electrooptics to characterisation of some typical materials like biopolymers (O'Konski 1978). In the present article, a brief reference is made to the physics and characteristics of electrooptic materials followed by material property and design parameter relations. Finally, the relevance of these investigations to the national scene and important problem areas in electrooptic material research are highlighted. The entire discussion is limited to the bulk materials and no attempt is made to include the theoretical aspects.

2. Electrooptic (EO) effects and characterisation techniques

The optical properties of crystals can be described by optical index ellipsoid or indicatrix (Nye 1960) given by

$$\sum_{ij} S_{ij} x_i x_j = 1 \tag{1}$$

where $S_{ij} = \left(\dfrac{1}{\epsilon}\right)_{ij} \equiv \left(\dfrac{1}{n^2}\right)_{ij}$. Here ϵ is the electronic (relative) dielectric constant and n the refractive index. With the application of an electric field the ellipsoid gets deformed and these distortions are simply related to changes in the coefficients S_{ij} in equation (1). Thus on application of an electric field, the changes in the phenomenological tensor quantities which relate the changes $\Delta(1/n^2)_{ij}$ to crystal perturbation can be given by

$$\Delta \left(\frac{1}{n^2}\right)_{ij} = \sum_{k} r_{ij,k} E_k + \sum_{kl} R_{ij,kl} E_k E_l + \ldots \tag{2}$$

Here the first term on right hand side represents the linear electrooptic (LEO) effect and $r_{ij,k}$ (third rank tensor components in units of m/V) are called the Pockels or linear electrooptic constants. The second term represents the quadratic electrooptic (QEO) effect and $R_{ij,kl}$ (fourth rank tensor components in units of m/V²) are called the Kerr or quadratic electrooptic constants. It can be shown by classical or tensorial methods that LEO effect exists only in crystals which are acentric. An alternative to equation (2), but more fundamental in nature has the field induced crystal polarisation P as the driving term. Thus

$$\Delta \left(\frac{1}{n^2}\right)_{ij} = \sum_{k} f_{ij,k} P_k + \sum_{kl} g_{ij,kl} P_k P_l + \ldots \tag{3}$$

Here f-coefficients (in units of m^2/C) and g-coefficients (in units of m^4/C^2) are called the polarisation optic coefficients and are similar to Pockels and Kerr constants respectively. Between r and f coefficients, the f coefficients are not only remarkably constant from material to material, but also do not change substantially over the frequency range $0–10^{15}$ Hz and hence the magnitude of linear susceptibility is of great importance in the study of electrooptics. In order to understand the entire electrooptic behaviour one has to apply corrections for the indirect effects caused in the material due to converse piezoelectric effect for LEO effect and electrostriction for QEO effect. The strain optical constants play a vital role in these studies as also in the study of acoustooptic devices (Veerabhadra Rao and Narasimhamurty 1975), the other important laser peripheral devices.

It is clear from the above discussion that in the final analysis, the determination of EO constants reduces to the determination of changes in optical parameters S_{ij} under the influence of an electric field. There are three parameters to determine these changes :

    (i) the field induced birefringence
    (ii) the angle of rotation of the optical indicatrix, and
    (iii) the absolute changes in the index.

Different methods are employed depending on the nature of change in the optical index ellipsoid which in turn depends on the point group symmetry. For example, the first method is used for $\bar{4}3m$ point group, second for $\bar{4}2m$ and third for 4 mm for some typical configurations. Field induced birefringence has been measured by standard techniques employing a calibrated compensator (Veerabhadra Rao and Narasimhamurty 1972, Kaschema *et al* 1975, Fox and Bruton 1975 and Baumann 1976) in the usual optical alignment. In some cases, the birefringence measurements alone cannot yield the coefficients and absolute change in n (or phase change) are required. Such measurements can be made by either using an interferometer (Kaminow 1966, Zook *et al* 1967) or through the use of a prism in a light deflection experiment (Ippen 1966). Other techniques include the optical heterodyne methods (De Lang 1968). Some of the methods can be used only for isotropic materials while some others are universal in nature and some are useful for LEO or QEO studies, one in the presence of the other. Kaminow (1974) has dealt with some of the fundamental techniques to study EO behaviour.

3. Characteristics of EO materials

Materials with large EO coefficients appear to have a potentiality as harmonic generation materials and hence an objective in the search of EO materials may be to find materials that make good modulators and harmonic generators. A comparision (Hulme 1972) of the materials for EO and harmonic generating

devices indicates that the optical transparancy, availability of the materials in larger sizes and durability under ambient conditions are common requirements for both of them. For a selection of good EO material and modulator design, the material properties and parameters (Milek and Neuberger 1972) given in Table 1 are important in addition to the requirements indicated above.

Table 1. Material parameters and their importance in EO studies

Parameter	Importance
1. Refractive index (n)	1. Figure of merit ($n^3 r$), reflection losses
2. Transmission	2. Spectral region for modulation
3. Relative dielectric constant (ϵ)	3. Capacitance, speed of electric fields in the crystal
4. Loss tangent (tan δ)	4. Electrical loss or modulation power
5. Electrical resistivity (ρ)	5. Space charge effects, heating of crystal
6. Crystal growing technique	6. Laboratory, commercial availability of large, good optical quality single crystals
7. Hardness	7. Resistance to strains
8. Solubility	8. Ambient environment applications
9. Photoelastic and piezoelectric properties	9. Clamped and unclamped effects
10. Thermal conductivity (k)	10. Deleterious birefringence
11. Radiation damage	11. High laser power suitability

Intuitive arguments lead to two types of materials with large polarisability : (a) with large electronic polarisability involving higher atomic numbered elements and closed shells, and (b) with large ionic polarisability like ferroelectrics and piezoelectrics. Since LEO effect is the one which finds more use in applications, one tries to develop materials with large LEO effect to be used in devices. The main condition for any material to show LEO effect is acentricity and hence in the following, the generation of acentricity in materials followed by an experimental test for the same is discussed briefly.

Bergman *et al* (1969) developed a general principle involving the nonbonded electron pairs leading to acentricity in materials. Considering the general requirements of high index of refraction, transparancy in the visible and infrared for nonlinear optical materials, one arrives at a system of the type ABL_x where A is the cation, B the highly polarisable component and L is of lesser polarisability with x amount. Usually the studies are confined to the system where $L_x = O_3$, making the material ABO_3 (Ex. $LiNbO_3$, $Ba_2NaNb_5O_{15}$ etc.) with B atoms situated at the centre of a distorted octahedron of oxygens. Accepting the stereochemical activity hypothesis, it is found on calculating the B–O distances that the nonbonded pair of electrons on B atoms (like Xe, I) definitely

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increases the distortion of the octahedron as compared to B atoms not containing the unbounded electron pairs (like Nb, Ta). Additional considerations like molecular geometry, radius ratio, ion size also ensure the accentricity of cation site. On these principles Bergman *et al* (1969) developed a variety of materials like XeO_3, MIO_3 (M = H, Li, Na, K etc.), and $MTeO_3$ (M = Ca, Sr, Ba, Zn etc.) which are found to be acentric. A second harmonic analyser has been designed and fabricated by Dougherty and Kurtz (1976) to test the existence of crystalline accentricity. The system (Figure 1) can be used even for powdered crystalline
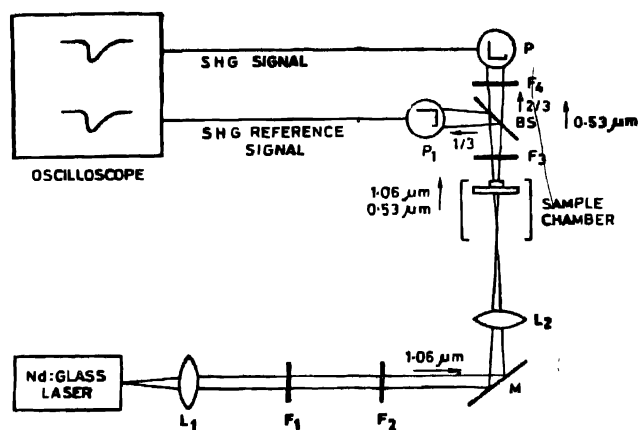

Figure 1. Second harmonic analyser

samples of milligram quantities. The experimental set up consists of a laser source (Nd : glass), and lenses L_1 and L_2 to collimate and focus the laser beam respectively. Various filters are used, to avoid flash lamp light (F_1), to control the intensity of fundamental laser light (F_2), to eliminate the laser wavelength (F_3) and a 50% second harmonic filter (F_4). M is a front surface mirror and BS the beam splitter. P_1 and P_2 are detectors of the radiation whose signals are fed to an oscilloscope. The sample chamber contains the sample and the temperature can be varied. The entire system should be made light tight so that the unit can be operated in normal laboratory illumination. The well balanced harmonic signals on oscilloscope indicate the presence of acentricity in the sample. The test can normally be done in about 10 to 30 minutes. One should take care to see that the material is free from acentric contaminants including the second phases of nominal material being studied.

4. Some important EO materials

Usually the EO single crystals are divided into four groups (Kaminow 1974) viz., isomorphs of ferroelectric KDP type and antiferroelectric ADP type, ferroelectric or pyroelectric ABO_3 type, tetrahedrally coordinated binary AB type semiconducting compounds and miscellaneous which do not fit into above three groups. In Table 2 are given some of the representative materials with their point groups and typical values of $r_{ij} \times 10^{20}$ m/V (in brackets).

Table 2. Some important single crystal materials

KDP Type $\bar{4}2m$ (r_{63})

KDP (10.5)	RDP (15.5)	RDA (13.0)
DKDP (26.4)	DRDP (21.4)	ADA (9.2)
KDA (10.9)	ADP (8.5)	CDA (18.6)
DKDA (18.2)	DADP (11.9)	DCDA (36.6)

ABO_3 Type compounds (perovskite family) (r_{33})

$LiNbO_3$, 3m (32.2)	$K_3Li_2Nb_5O_{15}$, 4mm (78)
$LiTaO_3$, 3m (30.3)	$Sr_{0.75}Ba_{0.25}Nb_2O_6$, 4mm (1340)
$BaTiO_3$, 4mm (28)	$Sr_{0.5}Ba_{0.5}Nb_2O_6$, 4mm (218)
$LiIO_3$, 4mm (6.4)	$Sr_{0.25}Ba_{0.75}Nb_2O_6$, 4mm (45)
$Ba_2NaNb_5O_{15}$, mm2(48)	$KTa_x Nb_{1-x} O_3$, 4mm(450)

AB Type compounds

ZnO, 6mm $(r_{41}=1.03)$	CuCl, $\bar{4}3m$ $(r_{41}=6.1)$
ZnS, $\bar{4}3m$ $(r_{11}=2.1)$	CuBr, $\bar{4}3m$ $(r_{11}=0.85)$
ZnSe, $\bar{4}3m$ $(r_{41}=2)$	GaP. $\bar{4}3m$ $(r_{41}=5)$
ZnTe, $\bar{4}3m$ $(r_{41}=4.55)$	GaAs, $\bar{4}3m$ $(r_{41}=1.6)$

Miscellaneous crystals

$Bi(GeO_4)_3$, $\bar{4}3m$ $(r_{41}=1.3)$	$NaNO_2$, mm2 $(r_{63}=3.0)$
Sodium uranyl acetate, 23, $(r_{41}=8.7)$	$K_3Mg_3(SO_4)_3$, 23 $(r_{41}=0.04)$
Quartz, 32 $(r_{41}=0.47)$	$(NH_4)_2Cd_2(SO_4)_3$, 23 $(r_{41}=0.8)$
Richelle salt, 222 $(r_{41}=2)$	$(NH_4) Mn_2(SO_4)_3$, 23 $(r_{41}=0.6)$

Among these crystals particular mention may be made of KDP, its isomorphs and deuterated versions. It is found that in most cases the deuteration increases the EO constants, thereby decreasing the half-wave voltages. Other important EO crystals are $LiTaO_3$, strontium barium niobate etc., the main advantage being nonhygroscopic nature and higher transmission ranges. Some work also has been initiated to observe LEO effect in doped alkali halides (Verlan and Tsyashchehenko 1975, Tada 1976). A survey of present day EO modulators and

indicates that through KDP and its isomorphs are hygroscopic, they are the most widely used modulator materials.

EO ceramics (Land 1967, Haertling and McCampbell 1972, Land *et al* 1974) are another class of important EO materials. An EO ceramic is a transparent or partially transparent polycrystalline ferroelectric material, i.e., it is an aggregate of the ferroelectric single crystal grains of size 0·5 to 4μ or more. Most of the important EO ceramics developed are based on lead zirconate titanate (PZT) modified by lanthanum, barium and strontium resulting in rhomboeedral slim loop or tetragonal PLZT, ferroelectric and slim loop PBZT, and ferroelectric PSZT respectively. Compared to single crystals, EO ceramics are less expensive, can be hot pressed to any size or shape, and optic axis orientation can be controlled. Important property unknown in single crystals and possible with ceramics is the variation of the effective birefringence and coarse grained EO effects. Some important applications of EO ceramics are light valves, and shutters, spectral filters, image storage and display devices. A variety of optical glasses (Pailletto 1975, 1976), particularly the neodymium, niobate and colored glasses, are also ot importance in EO studies. Other popular EO materials attracting the attention of the materials scientists are organic materials (Yamamoto and Ishihara 1976, Dazai *et al* 1977, Maogonum and Miller 1977).

5. Design parameters and material properties

With the nature and properties of different materials established, one may try to obtain a relation between the design parameters and material properties in order to 'cook' a material to the specification of an EO device. As an example, the design of an EO modulator is considered here. The important design equations[*] for high frequency small aperture modulation for use in wideband (> 100 MHz) communication system (DiDomenico and Wemple 1969) are :

(a) Temperature sensitivity,

$$\delta T = 5 \times 10^{-12} \left(\frac{\alpha}{b^2} \right) \left(\frac{V\pi}{\eta^3} \right) \cdot \delta T, \, °C \tag{4}$$

(b) Bandwidth,

$$\Delta f_m = 0.4 \, \frac{KQ}{\lambda^2} \, \text{MHz} \tag{5}$$

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[*] Varilus parameters used are : α : Diffraction parameter which limits aperture to length ratio ; b : Breadth of the modulator crystal in mm ; U_π : Stored energy parameter ; η : Packing density ; δT_π : Phase retardation in radians ; k : Thermal conductivity in w/°C.m ; Q : Dielectric quality factor at frequency f_m ; λ : Light wavelength in microns ; P_R : Reactive power in mW : Δf_m : Bandwidth in MH$_2$: I$_\pi$: Modulation current in amperes ; P : Polarisation in C/m^2 ; $V\pi$: Half-rave voltage in volts ; K_c : Dielectric constant along polar

(c) Power requirement,

$$(P_R/f_m)_{Min} = 2 \cdot 5 \times 10^3 \, b^2 \lambda \eta^3 \left(\frac{\delta T}{\delta T_s} \right) \cdot \frac{mw}{MHz} \tag{6}$$

(d) Current requirement,

$$(I\pi/f_m) = 2\lambda b(\eta^3/P) \cdot \frac{mA}{MHz} \tag{7}$$

(e) Voltage requirement,

$$V\pi = \frac{8\alpha\lambda^2}{b} \; (\eta^3/K_e\rho). \; V \tag{8}$$

(f) Modulator capacitance,

$$C = (160/V_\pi)(I \; /f_\pi) \; pF. \tag{9}$$

The following conclusions can be drawn from above equations for typical values of different parameters in the design of the EO modulator.

1. Smaller the stored energy parameter U_x, more precise should be the temperature control δT (Eq. 4). Modulator frontal area b^2 also should be made smaller to maximise δT.

2. The thermal conductivity k and quality factor Q should be higher to obtain a higher banewith (Eq. 5). For example, for $KTa_{0.65}Nb_{0.35}O_3$ $k = 5$ w/°Cm and Δf_m becomes 5 Q MHz at 0·633 μ and 2 Q at 1·06 μ. If the banawidth is to be more than 300 MHz, Q should be greater than 60 and 150 for 0·633 μ and 1·06 μ respectively. Hence the loss factor effects the bandwidth seriously and should be measured in all prospective EO materials.

3. In order to keep the power requirments (Eq. 6) a minimum U_π and b should be reduced for a fixed value of $(\delta T/\delta T)$ or U and $(\delta T/\delta T_s)$ should be reduced for a fixed value of b. Thus the temperature control places a lower limit on the drive power and thermal considerations place an upper limit on the bandwidth.

4. Smaller polarisation P requires higher modulator currents (Eq. 7). For materials with $P < 0·1$ C/m², and bandwidth greater than 300 MHz, current required will be of the order of 0·5 A. For example, for $LiTaO_3$, $P = 0·5$ C/m² and $I = 160$ mA.

5. The voltage requirements (Eq. 8) are controlled by the dielectric quantity K_iP. If V_x is to be less than about 25 V, the dielectric quality K_eP should be greater than 50.

6. The modulation capacitance (Eq. 9) will be greater than 5 pF if $V_\pi = 25$ V, $I > 240$ mA for a bandwidth of 300 MHz.

Generally one finds that the crystals of 4 mm symmetry ($\eta = 1$) are inherently superior both in power and bandwith to those of 6 mm ($\eta = 1\cdot2$) as can seen from Equations (6) and (7). In practice, since it is not possible to get the best EO material, satisfying all the design conditions, one tries to obtain the nearest material for a particular design. In Table 3 the characteristics for "BEST" EO material are compared with some well known oxygen octahedra ferroelectrics, for $0\cdot63\,\mu$ and $1\cdot06\,\mu$. The calculations are for $V \leqslant 25$ V, $\delta T_s = 0\cdot35$ rad., and $\alpha = 8$.

A perusal of Table 3 indicates that $LiTaO_3$ requires maximum drive power compared to other crystals and hence the circuitry developed for $LiTaO_3$ can be employed for other crystals listed. If the bandwidth is to be extended to 1000 MHz it requires higher values of Q which is difficult to achieve or in other words the loss properties should be clearly understood. Also, the operation at $1\cdot06\,\mu$ is far less practical than $0\cdot63\,\mu$ since the currents required are larger and powers are also large with the exception of KTN. Materials with low dielectric constant require unreasonably long crystals, and temperature control also is more stringent. Materials like KTN, strontium barium niobate and strontium potassium niobate appear as suitable EO materials, provided that the large power and current requirements are met. $BaTiO_3$ looks attractive if half-wave voltage can be reduced. It can also be noted from Table 3 that by suitable cooking of $LiNbO_3$ and KTN an optimal material with low, drive power and better EO properties may be tailored. Ultimately, in practice, the available temperature controller places a lower bound on modulator power and, dielectric Q and thermal conductivity place an upper bound on bandwidth and drive current becomes large in materials having smaller values of polarisation P.

6. Problem areas and conclusion

A comprehensive survey of the literature indicates that the problem areas in electrooptics can be broadly calassified into three major groups, viz., (a) material development, (b) EO characterisation of material, and (c) device making and other relevant applications.

In the first group, the importance of the search for newer materials with very low half-wave voltages and other attractive properties need not be over-emphasised. These can be crystals belonging to any of the groups mentioned earlier or others like organic materials, ceramics and glasses. In the second group, a necessity for developing new experimental techniques for characterisation is felt. This is for the determination of the EO constants and their variation with parameters like temperature, wavelength and impurities. Further, the properties like loss factor, thermal conductivity and others discussed in section 3

Table 3. Comparison of design parameters of some typical materials with best EO materials

Material	$P_R/f_m \frac{mW}{MHz}$		$I_\pi/f_m \frac{mA}{MHz}$		$\delta T°C$		5mm		1mm	
	0.63μ	1.06μ	0.63μ	1.06μ	0.63μ	1.06μ	0.63μ	1.06μ	1.06μ	1.06μ
1. Best	0·35	6	0·26	0·44	0·05	0·05	0·125	0·125	7·0	4·2
2. $LiTaO_3$	10·00	49	0·5	3·6	0·04	0·015	0·18	0·50	15·0	66·0
3. $LiNbO_3$	7·5	36	0·6	2·6	0·03	0·01	0·19	0·52	16	72
4. $Ba_2NaNb_5O_{15}$	7	33	0·6	2·6	0·06	0·02	0·16	0·44	12	52
5. $KTa_{0.65}Nb_{0.35}O_3$	2	9	3·2	5·3	0·03	0·08	0·125	0·125	7	4
7. $Sr_{0.5}Ba_{0.5}Nb_2O_6$	3	15	0·7	1·3	0·04	0·10	0·125	0·125	7	4
8. $Sr_2KWb_5O_{15}$	2	9	0·7	1·3	0·01	0·07	0·125	0·125	7	4

should bo investigated for the prospective EO materials. In the third gioup, now devices may bo tried in addition to the existing ones and other application areas (liko biopolymor electrooptics) may be attempted. Theoretical models working satisfactorily for EO effects in different materials also are not yet available.

In the national scone, the investigations on oloctrooptics is moagro. Somo work has been dono in the development of oxporimental techniquos and studies on materials (Veciabhadia Rao and Narasimhamurty 1972, 1976 and 1978) for the determination of EO constants   Work has also boon done m the country on ceramics of PLZT systems with respect to their dioloctric properties (Gururaja and Subbarao 1979, Gururaja et al 1979). With a little effort tho technology can bo extended to materials which will be of importance as EO ceramics. With heavy funding and importance to laser technology it is but natural that comparable activity should bo initiated in the laser peripheral devices and their technology also.

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