

ELECTRO-OPTIC PROPERTIES of KH_2PO_4 and ISOMORPHS, KDP, KD^*P / DKDP, ADP, AD^*P

([Table of Material Properties](#) appears below)

THE POCKELS ELECTRO-OPTIC EFFECT

Crystals which belong to twenty symmetry classes which lack a center of symmetry can show a linear electro-optic effect, that is, a change in refractive indices directly proportional to an applied voltage. The symmetry conditions for the occurrence of this effect are exactly the same as for the occurrence of the piezoelectric effect. Thus, there is an exact symmetry analogy between the linear electro-optic effect (refractive index a linear function of electric field) and the converse piezoelectric effect (geometric deformation a linear function of electric field). The linear electro-optic effect has the same relation to the Kerr effect (refractive index a quadratic function of electric field) as converse piezoelectricity has to electrostriction (geometric deformation a quadratic function of electric field). The linear change in refractive index obtained at room temperature with practical electric fields (up to 20kV/cm) is only of the order of 10^{-4} . Although this is too little to change refraction angles for most practical purposes⁷⁸, it is sufficient to produce retardations of the order of one wavelength and hence lead to interference phenomena. These interference phenomena are used to modulate light phase or intensity. A one-half wavelength relative retardation can change the transmission of polarized light from 0 to 100 percent. An ac voltage producing a peak retardation of one fourth wavelength can give 100 percent modulation of the carrier.

The linear electro-optic effect may be regarded as a special case of second order (non-linear!) electric interaction in the crystal: The action of a low frequency applied field and the electric field of the optic-frequency electro-magnetic wave combine to cause electric polarization at the optic frequency.

Linear electro-optic phenomena were discovered by Roentgen in quartz and thoroughly investigated in several crystals before the turn of the century by Pockels^{1,2}, in whose honor the effect is now generally called the Pockels effect. The broader study of higher order interaction in crystals began with Franken's discovery of frequency doubling of laser beams in quartz and KH_2PO_4 ³. In many linear electro-optic devices the longitudinal effect is used, that is, the light beam and electric field are parallel. Longitudinal effect devices are particularly useful for light beams of large cross-sectional area. Other electro-optic devices use the transverse effect with the light beam perpendicular to the applied field. Transverse effect devices avoid the use of transparent electrodes in the light path. In addition, the voltage required for a given retardation can be reduced by increasing the ratio of the light path length to the electrode spacing, whereas in longitudinal effect devices the required voltage is independent of the dimensions of the crystal. It can be shown from symmetry considerations that a longitudinal effect free of background birefringence and optical activity is obtained only with crystals of two classes: the class $\bar{3}m$ of the cubic system and the class $2m$ of the tetragonal system. Class $\bar{3}m$ is represented by cubic zinc sulfide (sphalerite)



and its isomorphs including ZnSe, ZnTe, GaAs, and CuCl. For a recent collection of data on such crystals see the compilations in Landolt-Boernstein^{9,10}. Class 2m is represented by KH₂PO₄ (KDP) and its isomorphs^{7,8,9,10}. Relatively large electro-optic effects and the availability of large crystals of high perfection have given crystals of this group continuing major importance for both "longitudinal" and transverse modulators as well as frequency doubling and mixing devices. Large strain-free crystals of KDP and a number of its isomorphs are available from Cleveland Crystals, Inc. These crystals are transparent throughout the visible and ultra-violet; one of the isomorphs (KH₂PO₄) is transparent to below 0.18μm⁸⁰. The infrared cutoff is near 1.5μm for the dihydrogen phosphates and near 2.1μm for KD₂ PO₄.

The electro-optic effects are traditionally expressed by the Pockels coefficients r_{ij}, where the first subscript refers to a tensor component of the index ellipsoid and the second to the electric field vector. In crystal class 2m there are only 2 coefficients, r₆₃ and r₄₁. The first measures the effect of a field component parallel to the optic axis. This makes the crystal optically biaxial, with vibration directions of increased and decreased refraction at ±45° to the X and Y axes. The r₆₃ coefficient is controlling for all modulators with field and light beam parallel to the optic axis (longitudinal devices) as well as most transverse devices. Transverse devices based on r₆₃ have the light path (length of the modulator bar) at ±45° to X. Only the ordinary ray undergoes variation of the refractive index.

The r₄₁ effect is, in first order, a rotation of the index ellipsoid around an electric field in the X direction and leads to maximum index change for light travelling at 45° to the optic axis. In spite of the high r₄₁ coefficients of KDP type crystals such devices have found only very limited application. The Pockels coefficients r_{ij} are related to observable refractive index changes by:

$$\Delta n = f r_{ij} n^3 E \quad (1)$$

where f is a numerical factor depending on crystal symmetry and device orientation, n is the appropriate refractive index, and E is the applied field strength. For KDP type crystals in longitudinal setup f = 1 and:

$$\Delta n = f r_{63} n_o^3 E \quad (2)$$

Here n_o is the ordinary index of refraction and n the difference in index for components polarized at + and - 45° to the X axis. The path difference in fractions of a wavelength in air then becomes:

$$\Delta n x/\lambda = r_{63} n_o^3 E x/\lambda = r_{63} n_o^3 V/\lambda \quad (3)$$

where X is the crystal thickness and V the applied voltage. The voltage required to obtain one-half wavelength relative retardation then follows:

$$V_{\lambda/2} = \lambda/[2 r_{63} n_o^3] \quad (4)$$

This magnitude has been widely used to characterize crystals for modulator use and is listed in our tables; note, however, that it is inverse to the strength of the electro-optic effect and that it depends explicitly on wavelength. The quantity r₆₃n_o³ varies only a few percent over the range of transparency of these crystals.

Because KDP type crystals are uniaxial with fairly high birefringence, longitudinal effect devices made of them have a limited angular aperture. The intensity of a light beam transmitted through basal section (Z-cut) placed between crossed polarizers is:

$$I = I_o \sin^2\{[x(n_o^2 - n_e^2)\sin(2\Theta)]/[2\lambda n_o n_e^2]\} \quad (5)$$

where I_o is the intensity of the incident beam polarized at 45° to its plane of incidence, x is the plate thickness, n_e is the extraordinary index of refraction, and Θ is the angle of incidence, outside the crystal. For a KDP crystal plate 2mm thick and $\lambda = 0.589\mu\text{m}$, equation (5) shows a first maximum for the transmitted intensity at $\Theta=5.1^\circ$. For linear polarized light from a uniformly radiating point source limited by an iris to a divergence of half-angle Θ_{max} , integration of equation (5) leads to an average transmitted intensity of:

$$I_{\text{ave}} = (I_o/6) \{ [x(n_o^2 - n_e^2)_{\text{max}}] / [2 n_o n_e^2] \}^2 \text{ for } \Theta_{\text{max}} \ll 1 \quad (6)$$

If I_{ave}/I_o for a 2mm Z-cut KDP crystal in diverging $0.589\mu\text{m}$ light is to be less than 0.005, equation (6) shows that Θ_{max} must be less than 1.6° . For a prescribed transmission ratio both I_{ave}/I_o and Θ_{max} are proportional to the inverse square root of the crystal thickness. The restricted angular aperture of Pockels effect modulators can be an important limitation in certain applications, such as photographic shutters. In laser devices the angular aperture of longitudinal KDP type modulators is generally ample.

For transverse KDP type devices the factor f in eq. (2) becomes $1/2$ and the induced path difference is:

$$\Delta x = r_{63} n_o^3 x E / (2) = r_{63} n_o^3 x V / (2t) \quad (7)$$

where V is the applied voltage and t the crystal thickness between the electrodes. In a transverse modulator rod a path difference $(n_o - n_e)x$ amounting to many wavelengths is present before the application of an electric field; this may be compensated by using two exactly equal rods in tandem and rotated at 90° to each other, with appropriate phasing of the applied voltage. The allowable angular aperture is less than for a longitudinal device but this is not a serious problem in highly collimated laser devices.

The symmetry analogies of the linear electro-optic and the converse piezoelectric effects have already been mentioned. More than that, a part of the linear electro-optic effect may be regarded as secondary: The electric field causes a deformation of the crystal by the converse piezoelectric effect and this deformation in turn produces a change in the refractive indices through the elasto-optic effect. This secondary effect may be suppressed by clamping the crystal. The total (free) electro-optic coefficients, r_{63}^T and r_{41}^T are related to the clamped coefficients r_{63}^S and r_{41}^S by:

$$r_{63}^T = r_{63}^S + p_{66} d_{36}, \text{ and } r_{41}^T = r_{41}^S + p_{44} d_{14} \quad (8)$$

where p_{44} and p_{66} are elasto-optic (strain-optic) constants, and d_{14} and d_{36} the piezoelectric constants. In ADP, the the clamped effect r_{63}^S amounts to only about 60% of the free effect, while in KDP the clamped r_{63}^S is near 90% of the free r_{63}^T . For ADP and KDP X-cuts the coefficients r_{41}^S and r_{41}^T are practically identical because the piezo-electric coefficients d_{14} are very small; but this is not true for the arsenates.

The clamped condition may be realized by mechanical constraints or, more effectively, by operation at frequencies well above the elastic resonances of the device, typically about 1 MHz. Near the elastic resonance frequencies, on the other hand, elastic strain may be a multiple of $d_{36}E$, and an enhanced elasto-optic contribution to the observed optic effect results. ADP and KDP X-cut devices have been used where a flat frequency response is essential. The clamped electro-optic constant r_{63}^S as well as the clamped dielectric constant of KDP have been shown to be independent of frequency up to at least 10 GHz⁵.

The coefficients r_{63} , and to a lesser extent r_{41} , are strongly temperature dependent. Increases by a factor 1000 have been measured as the temperature is lowered close to the Curie point. This dependence for r_{63} may be expressed by a Curie-Weiss law:

$$r_{63} = r_{63}^0 \left(\frac{\epsilon_{33} - \epsilon_0}{\epsilon_{33} - \epsilon_0} \right) = r_{63}^0 [C / (T - T_c) - 1] \quad (9)$$

Here ϵ_{33} and ϵ_0 are the permittivities of the crystal and free space, C is the dielectric Curie constant, T_c is the Curie temperature, and r_{63}^0 is an electro-optic constant for dielectric polarization, an independent variable. The r_{63} coefficients are not only nearly independent of temperature but also show a much reduced variation between members of the KDP group, and between free and clamped condition. r_{63}^T is 0.059 m²/C for KH₂PO₄, 0.061 for KD₂PO₄, 0.062 for KH₂AsO₄, and 0.063 for NH₄H₂PO₄⁹.

As r_{63} coefficients measure the charge density, and hence the current required to attain a prescribed level of light modulation, the current requirement is practically independent of temperature or of the choice among KDP type crystals. The differences in room temperature electro-optic properties of KH₂PO₄ and KD₂PO₄ can be fully accounted for by the 100° difference in Curie points. The principal reason for specifying the more costly KD₂PO₄ has been the resulting higher dielectric constant and hence lower voltage requirement. In applications involving light in the 0.9 to 1.81 μm region the lower absorption of KD₂PO₄ is likely to be decisive.

The most important publications on the electro-optic properties and applications of KDP type crystals have recently been reprinted.⁵

NON-LINEAR RESPONSE TO COHERENT LIGHT

In its most general terms, second-order non-linear dielectric interaction is the generation of electric polarization proportional to the product of two electric field components. The non-linear polarization contains the sum and difference frequencies of the input signals. In the Pockels effect, one component is at optic frequency (the incident beam) while the other is at much lower frequency, well below the infrared absorption region, or even d.c. The other case of major interest is the interaction of two coherent optic-frequency signals of the same frequency, and especially the interaction of a single input component with itself. This leads to generation of an output component at doubled frequency and a d.c. electric polarization. All these effects can be expressed by non-linear optic coefficients d_{ijk} , where any two subscripts relate to the input and the third to the output. The value of the d_{ijk} depends on each of the three frequencies involved. For KDP type

crystals (class 2m) the only d_{ijk} coefficients different from zero are those with all subscripts different. For frequency doubling these are $d_{312}(2, 1, 1)$ and $d_{132} = d_{231}(2, 1, 1)$, or in contracted form, $d_{36}(2,)$ and $d_{14} = d_{25}(2,)$. If the generating and resulting frequencies are all well within the optic transmission band, the order of the subscripts affects the value of d_{ijk} to an extent that is insignificant compared to other factors determining second harmonic or sum/difference frequency intensity. Then KDP type crystals may be characterized by a single non-linear coefficient $d_{123} = d_{14} = d_{36}$ (Kleinmann relation).

The relative values of d_{ijk} for different orientation and temperature and for different substances can be obtained with much better accuracy than absolute values. Therefore the literature contains many values relative to d_{36} of KDP. Absolute values were calculated from relative values by Zernike and Midwinter⁶ and also more recently by Kurtz, Jerphagnon, and Choy⁴.

For one input or output near zero frequency (Pockels effect or rectified output) we have non-linear coefficients related to the Pockels coefficients:

$$4d_{36}(O,) = -n_o^4 r_{63}^T \quad \text{and} \quad 4d_{14}(O,) = -n_o^2 n_e^2 r_{41}^T \quad (10)$$

(ref. 5, eq. 17.,p9 59). The numerical factor with the d_{ij} is in controversy⁷⁹.

PHASE MATCHING (ref. 6, chapter 3)

For substantial generation of sum and difference frequencies, and especially for frequency doubling, it is required that the incoming and generated light are in a defined phase relation along the path of interaction. Due to normal dispersion the double-frequency light generated sees a refractive index a few hundredths higher than the fundamental frequency. This would bring the two waves out of phase over a path of about $\lambda/[n(2\lambda) - n(\lambda)]$, in the order of 10 to 50 microns. Fortunately the birefringence of many non-linear crystals, and the KDP group in particular, allows one to overcome this limitation by phase matching. The common way of achieving this (type I phase-matching) with KDP type crystals (class 2m, $n_o > n_e$) uses an ordinary wave exciting an electric vector in the basal plane at 45° to the X and Y axes. The non-linear response is electric polarization in the Z direction which builds up to an extraordinary wave. Cumulative energy transfer occurs along a common wave normal if the two waves see the same refractive index. For KDP and ADP at room temperature $n_e(2\lambda) = n_o(\lambda)$ for input wavelengths near 500 nm. Type I phase matching is obtained for the indicated wavelength with wave propagation perpendicular to the optic axis, i.e. 90° phase matching. The condition $n_e(2\lambda) = n_o(\lambda)$ occurs for longer wavelengths with KDP type rubidium and cesium salts, which have lower birefringence. This is their principal merit for optic devices. Some tuning is possible by temperature variation; $n_o - n_e$ decreases with increasing temperature.

If KDP or ADP is to be used to double red or near infrared light, angle tuning is resorted to: The wave normal is inclined to the optic axis so that the generated extraordinary ray sees a refractive index intermediate between n_o and n_e according to:

$$1/n^2(\theta, 2\lambda) = \sin^2(\theta)/n_e^2(2\lambda) + \cos^2(\theta)/n_o^2(2\lambda) = 1/n_o^2(\lambda) \quad (11)$$

Phase matching by angle tuning has three disadvantages compared to temperature tuning:

- 1) Ray directions are not parallel to the wave normal; this limits the practical interaction length.
- 2) The effective non-linear coefficient is reduced.
- 3) The angular tolerance (beam angular aperture) is reduced.

Problems 2) and 3) may be ameliorated to some extent by resorting to type II phase-matching. For KDP type crystals this employs an incoming beam in the XZ plane polarized at 45° to this plane. The interaction of the resulting ordinary and extraordinary components of the fundamental creates an extraordinary ray at doubled frequency. The effect of birefringence is reduced by one-half, and the maximum effective d coefficient now occurs for a beam at 45° to the optic axis. Phase matching of ADP or KDP for doubling of 1060nm light occurs not far from this angle.^{12,13}

TEMPERATURE LIMITATIONS

A lower temperature limit for the optic use of KDP type crystals is set by their Curie points (see table), below which optic properties are affected by domain structure which may also lead to cracking of the crystal. Operation just above the Curie point is attractive by the extremely high response to applied voltage, but this is penalized by high temperature coefficients. Most KDP type crystals convert to an alternate, inactive crystal structure at high temperatures. For all compounds except RDP, conversion does not take place below 100°C*. Chemical decomposition for the alkali metal salts does not occur below 200°C, but is noticeable for the ammonium salts at about 120°C. KDP type crystals show a small protonic conductivity with an activation energy of about 0.6 eV resulting in a positive temperature coefficient of conductivity of about 8%/°C near room temperature.^{14,15,16} Resistivity of the best available ADP and KDP crystals is from 10 to 20 x 10⁸ Ohm-meter at 25°C. Ionic impurities, such as HSO₄⁻ substituting for H₂PO₄⁻, can increase the conductivity many fold. Resistivity so far achieved with the arsenates is 1 to 3 powers of 10 lower than in ADP and KDP⁸. The negative temperature coefficient of resistance can lead to a runaway breakdown if high electric field (kV/cm) is applied continuously.

* Fully deuterated KD₂PO₄ is actually only metastable near 100°C, but crystal elements have not been observed to transform.

Properties of KDP, KD*P, ADP, and AD*P				
	KH ₂ PO ₄ (KDP)	KD ₂ PO ₄ (KD*P)	NH ₄ H ₂ PO ₄ (ADP)	ND ₄ D ₂ PO ₄ (AD*P)
Crystal Data		99 atom% D		99 atom% D
Crystal Symmetry and Class	Tetragonal, 2m	Tetragonal, 2m	Tetragonal, 2m	Tetragonal, 2m

Space Group	I 2d	I 2d	I 2d	I 2d
Lattice Constants ⁴⁷ (angstroms)	a=7.4529 c=6.9751	a=7.4697 c=6.9766	a=7.4991 c=7.5493	a=7.5193 c=7.5400
Density, g/cc	2.3325	2.3555	1.799	1.885
Cleavage	none	none	none	none
Optical Properties				
Micron region of >50% transmission (2mm thickness)	0.176 ⁽⁸⁰⁾ to 1.55 ⁽²⁵⁾	<0.2 to 2.15 ⁽²⁵⁾	0.184 ⁽⁸⁰⁾ to 1.5 ⁽⁴¹⁾	<0.2 to >2.0 *
Indices of refraction (In 22°C air)		99 atom% D		95 atom% D
0.266μm	n _o =1.5599 n _e =1.5105	n _o =1.5539 n _e =1.5071	n _o =1.5797 n _e =1.5261	n _o =1.5701 n _e =1.5191
0.3547μm	n _o =1.5318 n _e =1.4864	n _o =1.5250 n _e =1.4834	n _o =1.5485 n _e =1.49895	n _o =1.5403 n _e =1.4942
0.532μm	n _o =1.5129 n _e =1.4709	n _o =1.5071 n _e =1.4683	n _o =1.52775 n _e =1.4815	n _o =1.5211 n _e =1.4776
0.5893μm	n _o =1.5098 n _e =1.4687	n _o =1.5045 n _e =1.4661	n _o =1.52435 n _e =1.4789	n _o =1.51815 n _e =1.4751
0.6328μm	n _o =1.5079 n _e =1.4673	n _o =1.50285 n _e =1.4648	n _o =1.5222 n _e =1.4773	n _o =1.5163 n _e =1.47365
0.6943μm	n _o =1.5055 n _e =1.4658	n _o =1.50095 n _e =1.4633	n _o =1.5195 n _e =1.4754	n _o =1.5142 n _e =1.4719
1.064μm	n _o =1.4944 n _e =1.46035	n _o =1.4934 n _e =1.4583	n _o =1.5068 n _e =1.4681	n _o =1.5052 n _e =1.4658
dn/dT, 10 ⁻⁶ /°C (20 to 40°C)		99 atom% D		95 atom% D
0.266μm	dn _o /dT= -38 dn _e /dT= -29	dn _o /dT= -37 dn _e /dT= -24	dn _o /dT= -52 dn _e /dT= -2	dn _o /dT= -24 dn _e /dT= +2
0.532μm	dn _o /dT= -40 dn _e /dT= -29	dn _o /dT= -37 dn _e /dT= -24	dn _o /dT= -56 dn _e /dT= -2	dn _o /dT= -28 dn _e /dT= +2
1.064μm	dn _o /dT= -48 dn _e /dT= -31	dn _o /dT= -44 dn _e /dT= -24	dn _o /dT= -81 dn _e /dT= -3	dn _o /dT= -24 dn _e /dT= +5
Fresnel Refection Loss per surface		99 atom% D		95 atom% D
0.266μm	e-ray: 4.8% o-ray: 4.1%	e-ray: 4.7% o-ray: 4.1%	e-ray: 5.05% o-ray: 4.3%	e-ray: 4.9% o-ray: 4.25%

0.532μm	e-ray: 4.2% o-ray: 3.6%	e-ray: 4.1% o-ray: 3.6%	e-ray: 4.4% o-ray: 3.8%	e-ray: 4.3% o-ray: 3.7%
1.06μm	e-ray: 3.9% o-ray: 3.5%	e-ray: 3.9% o-ray: 3.5%	e-ray: 4.1% o-ray: 3.6%	e-ray: 4.1% o-ray: 3.6%
NLO Susceptibility d_{36}, 10^{-12}m/V				
0.6943μm	0.70 ⁽⁴⁾	0.53 ⁽⁴⁾	0.85 ⁽⁴⁾	0.77 ⁽⁴⁾
1.064μm	0.63 ⁽⁴⁾	0.42 ⁽⁴⁾	0.762 ⁽⁴⁾	---
90° Phasematching Wavelength(μm) and Temp(C)	0.518, 25° 0.527, 120°	---	0.515, -10° 0.557, 120°	---
Electrical Properties				
Resistivity (10^6 ohm-cm)	10-20	10	10-20	10
Relative dielectric constant at 25C and 1KHz		99 atom% D		99 atom% D
$_{11}^T / o$	43.2 ⁽⁸²⁾	65 ⁽⁶⁴⁾	56 ⁽⁸²⁾	70 ⁽⁸⁾
$_{11}^S / o$	42.5 ⁽⁸²⁾	62.5 ⁽⁶⁴⁾	55.5 ⁽⁸²⁾	---
$_{33}^T / o$	20.8 ⁽⁸²⁾	50 ⁽²⁵⁾	15.5 ⁽⁸²⁾	26 ⁽⁸⁾
$_{33}^S / o$	20.0 ⁽⁸²⁾	48 ⁽⁸²⁾	15.0 ⁽⁸²⁾	---
Piezoelectric Coefficients, (pC/N)				
d_{14}	1.3 ⁽⁶³⁾	3.4 ⁽⁷⁴⁾ ~98 at% D	1.8 ⁽⁸³⁾	10 ⁽⁶⁷⁾ ~98 at% D
d_{36}	21 ⁽⁶³⁾	58 ⁽²⁵⁾ 99 at% D	48.3 ⁽⁸³⁾	75 ⁽⁶⁷⁾ 99 at% D
Electromechanical Coupling Factors				
k_{14}	0.008 ⁽⁶³⁾	---	0.006	---
k_{36}	0.121 ⁽⁶³⁾	0.22 ⁽²⁵⁾	0.33	---

Unreferenced data are unpublished data of Cleveland Crystals, Inc.

Where not listed, deuteration levels are unknown.

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