Nonlinear Optics & Materials

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The nonlinear optical processes are among the most fascinating effects that can be produced in lasers. These processes are almost magical, as they permit light of one colour (wavelength) to be converted into the light of a different colour. They are also important commercially and scientifically, as they make it possible to access parts of the spectrum that are difficult to obtain in other way. Nonlinear optics is a study that deals mainly with various new optical effects and novel phenomena arising from the interactions of intense coherent optical radiation with mater. There is a historical reason why this new branch of optical physics is termed "*nonlinear optics"*.

In the regime of conventional optics, the electric polarization vector $\boldsymbol{P}$ is simply assumed to be linearly proportional to the electric field strength $\boldsymbol{E}$ of an applied optical wave, i.e.

$$\boldsymbol{P} = \varepsilon_0 \chi \boldsymbol{E} \tag{1}$$

Where ε_0 is the free-space permittivity, χ is the susceptibility of a given medium and a plot of $\boldsymbol{P}$ versus $\boldsymbol{E}$ is a straight line.

Before 1960's, in the area of conventional optics many basic mathematical equations or formulae manifested a linear feature. The first breakthrough was achieved in 1961 when a pulsed laser beam was sent into a piezoelectric crystal sample. In this case researchers, for the first time in history of optics, observed the second-harmonic generation at an optical frequency. Shortly after that, several other optical frequency-mixing effects were sequentially demonstrated based on the use of laser radiation, which included the optical sum-frequency generation (1962), optical third-harmonic generation (1962), optical rectification, optical difference-frequency generation (1963), and the optical parametric amplification and oscillation (1965). These experimental demonstrations not only verified the validity of nonlinear polarization theories but also provided an alternative approach to generate coherent optical radiation.During the same time period, another important event was the discovery of stimulated Raman scattering (1962), which can be recognized as the second milestone in the history of nonlinear optics. The significance of this discovery is that, for the first time, the '*stimulated*' of light scattering excited by an intense laser beam was revealed. The stimulated

scattering is an alternative physical approach to generate the coherent optical radiation without the need of population inversion. Later, researchers reported the observation of another major nonlinear optical effect, stimulated Brillouin scattering (1964), which arose from the interaction of an intense monochromatic optical field with the induced hypersonic field in a scattering medium through the so-called optical electrostriction mechanism. Since then, the stimulated Brillouin scattering has become an efficient technique to generate or amplify the coherent optical radiation with a small frequency-shift or fine tunability. In principle, this effect can also be employed to generate or amplify a coherent hypersonic field in the optical medium.

The relation (1) is valid for the field strengths of conventional sources. The quantity χ is a constant only in the sense of being independent of $\boldsymbol{E}$; its magnitude is a function of the frequency. With sufficiently intense laser radiation this relation does not hold good and has to be generalized to equation (2), which can be written in the vector form, as by a power series

$$\boldsymbol{P} = \varepsilon_0[\chi^{(1)}\boldsymbol{E} + \chi^{(2)}\boldsymbol{EE} + \chi^{(3)}\boldsymbol{EEE} + \text{--------}] \qquad (2)$$

where $\chi_{ij}^{(1)}$ is a second – rank (linear) tensor, $\chi_{ijk}^{(2)}$ is a third -rank (nonlinear) tensor, and $\chi_{ijkl}^{(3)}$ is a forth-rank (nonlinear) tensor. The values of the tensor coefficients are functions of frequency and temperature.

For small field strength the polarization is proportional to the electric field $\boldsymbol{E}$ and is accounted for by the polarizatbility tensor $\chi^{(1)}$. All of the optics discussed so far has been linear optics encompassed in the term$\varepsilon_0\chi^{(1)}E$. This term represents optical phenomenon that are proportional to the electric field and are at the frequency of incoming wave.The term $\chi^{(2)}\boldsymbol{EE}$ is responsible for all of the two-wave effects. This includes second harmonic generation (SHG) (two fields at ω to make one at 2ω) and optical parametric oscillation (OPO) (one field at ω_1 and other field at ω_2 to create fields at $\omega_1 - \omega_2$ and $\omega_1 + \omega_2$). This also includes optical mixing, and the Pocals effect (change of index of refraction with applied electric field). The nonlinear polarization tensor $\chi^{(2)}$ vanishes in the crystals that have a center of symmetry (i.e. crystal symmetry). In these crystals second harmonic generation is not possible. As a result of, many of the components of $\chi^{(2)}$ will be zero or equal to other components of the tensor. Thus the second-order polarization and the corresponding monochromatic components of the optical field:

$$\boldsymbol{P}^{(2)}(\omega = \omega_1 + \omega_2) = \varepsilon_0 \chi^{(2)}(\omega_1, \omega_2)\boldsymbol{E}(\omega_1)\boldsymbol{E}(\omega_2) \qquad (3)$$

where $\chi^{(2)}$ denotes the second-order susceptibility that is a third-order tensor. The term $\chi^{(3)}$ $\boldsymbol{EEE}$ is responsible for all of the three wave effects. This includes third harmonic generation (three fields at ω to make one at 3ω) and various combinations of three fields to produce sum and difference frequencies as in four-wave mixing. This also includes the Kerr effect (where the index of refraction is a function of the input light intensity). The imaginary component of $\chi^{(3)}$ is responsible for Raman, Brillouin, and Rayleigh scattering, as well as for two-photon absorption.

Based on these comparisons described above, it is concluded that the main concern in conventional optics is the propagation and interaction with matter of the light from ordinary light sources, wherein the intensities of the light beams are so low that even a simple linear approximation is enough to give a good theoretical explanation for the related optical effects and phenomena. In this sense, the conventional optics may also be called '*linear optics*' or '*optics of weak light*'. On the other hand, '*nonlinear optics*' mainly deals with the interaction of intense laser radiation with matter. In this latter case, the intensities of laser beams can be so high that a great number of new effects and novel phenomena can be observed, and some high-order nonlinear approximations have to be employed to explain these new effects and phenomena. In this sense, nonlinear optics may also be called '*optics of intense light*'.

In the present chapter we shall discuss light propagation through crystals in the presence of an externally applied electric field. This field can in general, alter the refractive indices of the crystal and thus could induces birefringence in isotropic crystal, or could alter the birefringence property of the crystal. This effect is known as the electrooptic effect. If the changes in the refractive indices are proportional to the applied electric field, such an effect is known as Pockel's effect or the linear electrooptic effect. If the changes in indices are proportional to the square of the applied electric field, the effect is referred to as the quadratic electrooptic effect or the Kerr effect.

Nonlinear Optical Materials

For generating new frequencies from existing lasers via harmonic generation and difference generation, there has been an extensive effort in recent years to identify effective materials for such processes. In addition to having a large nonlinearity, these materials must be transparent not only at the laser frequency but also at the newly generated frequency. They must (1) be resistant to optical damage, (2) have high mechanical hardness, (3) exhibit good thermal and chemical stability, (4) be capable of being grown in useful sizes, and (5) have the appropriate phase-matching properties. The second harmonic crystals must have no inversion symmetry (i.e. non-centrosymmetric). Bulk second-order nonlinear materials are generally inorganic crystals. A number of semiconductors are useful for second harmonic generation when used in waveguides.The nonlinear crystals can be classified into two groups according to their physical properties. Crystals grown from water solutions are fragile, hygroscopic, and sensitive to thermal shock. The crystals of this group, to which KDP and its isomorphs belong, are somewhat difficult to handle because the crystals are soft, and the polished faces may be fogged if they are held with bare hands or exposed to humid atmosphere. On the other hand, the crystals are easy to grow, they are available in large sizes, and they are of excellent optical quality. Crystals grown from the melt are relatively hard, nonhygroscopic and less sensitive to thermal shock. Important members of this group crystals are $LiNbO_3$ and $Ba_2NaNb_5O_{15}$. The optical quality is usually inferior to water grown crystals because of refractive index nonuniformities associated with the crystal growth condition. Table-1, lists the primary nonlinear materials which are used in conjunction with solid state-lasers.

Some important Nonlinear Optical Crystals:-

1. Potassium dihydrogen phosphate (KDP) family

The crystals of this family have proven to be the most important group of useful second-harmonic generators. The crystals which are all negative uniaxial includes KDP, ADP, and CDA, belong to point group $\bar{4}2m$ and, thus, have a tetragonal symmetry. The crystals are grown at room temperature from a water solution and therefore are exceptionally hygroscopic. Due to the hygroscopic nature of the crystals, they are often polished in solutions of ethylene glycol rather than in water. However, the ease of crystal growth means that it is possible to obtain large crystals of very high optical quality. Most members of the KDP family have a high damage threshold for

optically induced damage. Transparency exists from 0.22 to 1.6 μm for the phosphates, and from about 0.26 to 1.6 μm for arsenates. Deuterated forms of the crystals are also available and are indicated by the notation D*. Deuteration increases the infrared limit to about 1.9 μm. KDP possesses an absorption band at 1 to 1.2 μm, so KD*P is preferred over KDP for Nd:YAG and Nd:glass lasers. The greatest attributes of this family of crystals as a nonlinear device material are their resistance to laser damage and their high optical quality. Opposing these advantages, there are several disadvantages. The materials have fairly low refractive indices, typically 1.50 to 1.55, so that they also have small nonlinear coefficients. All of the KDP isomorphs are water-soluble and have a maximum safe operating temperature of about 100 ^{0}C. The crystals are fragile and sensitive to thermal shock as well as hairline fractures, and should be heated slowly at a rate of less than about 5 ^{0}C/min.Isomorphs of these materials have similarly been used in nonlinear optics, the most widely known isomorphs being deuterated KDP, which is normally designated as KD*P. Some of the other isomorphs have been used because the temperature dependence of their refractive index allows 90^{0}phase matching for particular interactions. CDA and CD*A 90^0 phase-match the important 1.06-μm transition of Nd:YAG and Nd:glass, RDP and RDA are mainly employed to frequency-doubled the output from the ruby lasers.

2. Lithium niobate ($LiNBO_3$)

$LiNBO_3$ [194] is a 3m negative uniaxial crystal with trigonal symmetry. $LiNBO_3$ crystals are grown from the melt by a Czochralski process. This material is nonhygroscopic and hard, taking a good polish readily. In addition, they have a much higher nonlinear coefficient than members of the KDP family. However, $LiNBO_3$ has a significantly lower damage threshold than members of the KDP family. In addition, many crystals of $LiNBO_3$ are susceptible to a type of damage (often referred to a photorefractive damage) that alters the index of refraction within the crystal. The crystals of lithium niobate are transparent in the region 0.42 to 5.2 μm. Temperature sensitivity of birefringence is such that, by varying temperature, phase matching can be achieved to 90^0 to the optical axis. Doping $LiNBO_3$ with MgO reduces the photorefractive damage and permits 90-degree phase matching at a temperature of 107 ^{0}C. $LiNBO_3$ is most commonly used as a doubling crystal for internally frequency-doubled Nd:YAG lasers, and for lower-power externally frequency-doubled Nd:YAG and Nd:glass lasers.

3. Potassium titalyl phosphate (KTP)

KTP ($KTiOPO_4$) is a mm2 biaxial crystal with orthorhombic symmetry. KTP is a difficult crystal to grow, and is currently grown by hydrothermal and flux growth techniques. However, KTP possesses good optical properties, a large acceptance angle, large temperature acceptance, a large nonlinear coefficient, and high optical damage thresholds. It is a mechanically rugged and nonhygroscopic crystal. However KTP suffers from a cumulative photochemical degradation phenomena, termed *grey tracking*, caused by long-term exposure to the intense fundamental and second harmonic radiation. Although this photochemical effect can be reversed by operating the crystal at an elevated temperature, absorption in the crystal due to the grey tracking may damage the crystal beyond repair. KTP is a more recently developed crystal than KDP or $LiNbO_3$, but is emerging as one of the most popular frequency-doubling crystals for Nd:YAG and Nd:glass lasers. KTP is also finding application as an OPO material and in difference frequency application.

4. Beta-barium borate (BBO)

BBO (β-BaB_2O_4) is a 3m negative uniaxial crystal with trigonal symmetry. The crystal possesses a moderate nonlinear coefficient, high-temperature stability, and an outstanding damage threshold. It is mildly hygroscopic, typically requiring a special housing under laboratory conditions. The crystal's optical transmission also extends down to 200nm, making it possible to perform multiphoton nonlinear processes into the blue and UV. However, BBO also possesses a very low angular tolerance, meaning that good alignment and high-quality optical beams are required for efficient nonlinear conversion. The large walk-off angle also results in beams that are elliptical. Due to its broad phase-matching region. BBO finds good application for harmonic generations. It also is finding application in broadly tunable OPOs and optical parametric amplifiers (OPAs). The exceptionally high IR transmittance allows high average power OPO and OPA operation with minimal thermal heating caused by the idler radiation.

5. Barium sodium niobate ($Ba_2NaNb_5O_{15}$)

$Ba_2NaNb_5O_{15}$ is a mm2 biaxial crystal with orthorhombic symmetry. However, since $n_x \approx n_y$ to a first approximation, at least insofar as the gross properties of phase matching are concerned, the crystal can be assumed to be uniaxial. It is mechanically hard, nonhygroscopic crystal with good chemical stability and UV transparency. It

possesses a moderate nonlinear coefficient and good optical damage thresholds. This freedom from damage, combined with 90^0 phase matching, has made possible very efficient second-harmonic generation from a continuous-wave Nd:YAG laser, providing an intense source of 5300 Å radiation. However, the use of $Ba_2NaNb_5O_{15}$, which transmits light in the region 0.46 to 1.1 μm, presents other problems. Great difficulties are involved in preparing high optical quality crystals and are no longer available commercially. Commercially available crystals suffer from situation, scatter centers, and built-in strain. The most persistent defect are growth striations which are periodic variations of refractive index normal to the growth direction. Light is diffracted and refracted by these index variations, making good phase matching very difficult to achieve. $Ba_2NaNb_5O_{15}$ or "Banana" crystals were a popular nonlinear material in the mid-1970s.

6. Lithium iodate ($LiIO_3$) and Lithium formate monohydrate

$LiIO_3$ is a 6 crystal with hexagonal symmetry. The crystal is hygroscopic and significantly more fragile than $LiNbO_3$. In addition, this crystal has a rather low optical-damage threshold. However, the crystal does have a high nonlinear coefficient and does not suffer from the photorefractive damage of $LiNbO_3$. $LiIO_3$ crystals are transparent in the region 0.32 to 5.0 μm, while formate crystals are transparent in the 0.25 to 1.2μm range. Both crystals are grown from water solutions. Lithium iodate has nonlinear coefficients which are comparable to those of lithium niobate, although it does not suffer from the refractive index damage problems that plague the niobate. Also $LiIO_3$ possesses an optical quality which is far superior to $Ba_2NaNb_5O_{15}$. Consequently, lithium iodate has found application in efficient intracavity second-harmonic generation in Nd:YAG lasers. Since the refractive indices are very stable with respect to temperature, phase matching is achieved by angle tuning. Interesting features of the formate crystals are a near noncritical phase matching for Nd laser radiation and an insensitivity of the phase-match direction to temperature changes.

7. Lithium tri-borate (LBO)

LBO is a mm2 biaxial crystal with orthorhombic symmetry. It is a mechanically hard, nonhygroscopic crystal with good chemical stability and UV transparency. It possesses a moderate nonlinear coefficient and good optical damage thresholds. This crystal has a transparency range extends down to 160 nm and absorption is from 170

nm to 2.6 μm. It offers smallest walk- off angle and widest acceptance angle for both type-I and type-II phase matching for SHG and THG of Nd:YAG laser.

Table-1 Nonlinear Optical Crystals

Crystal	Crystal Class	Transparent range (μm)	d_{il} (10^{-12} m/V)	Phase matching	Damage Threshold (MW / cm^2)	Efficiency (%)
KH_2PO_4 (KDP)	$\bar{4}2m$ (a)	0.2 – 1.5	d_{36} = 0.39	φ -Tuning	~ 500	20 -30
KD_2PO_4 (KD^*P)	$\bar{4}2m$ (a)	0.2 – 1.5	d_{36} = 0.37	φ -Tuning	~ 1000	20 -40
$NH_4H_2PO_4$ (ADP)	$\bar{4}2m$ (a)	0.2 – 1.2	d_{36} = 0.47	φ -Tuning	~ 500	20 -30
$KNbO_3$ (KN)	*mm*2 (b)	0.4 - 5	d_{31} = 12.8	T-Tuning	> 500	30 ~ 60
β-BaB_2O_4 (BBO)	3*m* (a)	0.19 - 3	d_{22} = 2.3	φ -Tuning	~1500	40 ~ 60
LiB_3O_5 (LBO)	*mm*2 (b)	0.16 – 2.6	d_{31} = 0.85	φ -Tuning	~ 2000	40 ~ 60
$KTiOPO_4$ (KTP)	*mm*2 (b)	0.3 – 5.3	d_{24} = 3.3	φ -Tuning	~ 500	40 ~ 60
$LiNbO_3$ (LN)	3*m* (a)	0.33 - 5	d_{31} = -5.3 d_{22} = 2.1	T-Tuning	~ 100	30 ~ 50
$LiIO_3$	6 (a)	0.3 – 5.5	d_{31} = 5.4	φ -Tuning	~ 100	30 ~ 50
CsH_2AsO_4 (CDA)	$\bar{4}2m$ (a)	0.26 – 1.43	d_{36} = 0.40	φ -Tuning T-Tuning	~ 500	30 ~ 50
RbH_2AsO_4 (RDA)	$\bar{4}2m$ (a)	0.26 – 1.46	d_{36} = 0.39 (0.694 μm)	φ -Tuning T-Tuning	~ 350 (0.694 μm)	30 ~ 50
RbH_2PO_4 (RDP)	$\bar{4}2m$ (a)	0.22 – 1.4	d_{36} = 0.4	φ -Tuning	~ 300	30 ~ 50
$AgGaSe_2$	$\bar{4}2m$ (a)	0.7 – 18	d_{36} = 33 (10.6 μm)	φ -Tuning	~ 350 (10.6 μm)	5 ~ 10
Ag_3AsS_3	3*m* (a)	0.6 – 13	d_{15} = 11 d_{22} = 18 (10.6 μm)	φ -Tuning	~ 30 (10.6 μm)	5 ~ 10
$CdGeAs_2$	$\bar{4}2m$ (a)	2.4 – 18	d_{36} = 235 (10.6 μm)	φ -Tuning	~ 40 (10.6 μm)	10 ~ 20

(a) Negative uniaxial crystal, (b) Negative biaxial crystal, and (c) Positive uniaxial crystal.
φ -Tuning: angle tuning; **T –Tuning**: temperature tuning.

Table-2. Properties of some important nonlinear crystals

Property	KTP	BBO	LBO	CLBO
Nonlinear coefficient (pm/V)	3.1	1.94	1.16	1.11
Transmission range (μm)	0.35 - 5.5	0.19 - 3.5	0.16 – 2.6	0.16 – 2.6
Damage threshold (GW/cm^2)	> 0.5	1.5	2.5	> 2.5
Angular acceptance (mrad-cm)	20	< 1	2	1.4
Spectral acceptance (mm-cm)	0.5	0.5	0.8	1
Walk-off angle (degree)	1.3	5.5	<1	1.8
Damage resistance to moisture	High	Low	Low	Medium

Table-3. Nonlinear optical coefficients for primary materials

Material	Formula	Relative nonlinear coefficient d/d_{36} (KDP)	Nonlinear coefficient[a] [10^{-24} As/V^2]	Index of refraction n^o (1.06μm)
Ammonium dihydrogen phosphate (ADP)	$NH_4H_2PO_4$	1.2	d_{36} =5.96	1.50
Potassium dihydrogen phosphate (KDP)	KH_2PO_4	1.0	d_{36} =5.16	1.49
Potassium dideuterium phosphate	KD_2PO_4	1.06	d_{36} =5.43	1.47
Rubidium dihydrogen phosphate (KD*P)	RbH_2PO_4	0.92	d_{36} =3.81	1.49
Rubidium dihydrogen arsenate (RDA)	RbH_2AsO_4	0.64	d_{36} =2.66	1.55
Cesium dihydrogen arsenate (CDA)	CsH_2AsO_4	0.92	d_{36} =3.81	1.55
Cesium dideuterium arsenate (CD*A)	CsD_2AsO_4	0.92	d_{36} =3.81	1.55
Lithium iodate	$LiIO_3$	15.0	d_{31} =58.4	1.86
Lithium niobate	$LiNbO_3$	13.4	d_{31} =55.8	2.23
Barium sodium niobate	$Ba_2NaNb_5O_{15}$	38.0	d_{31} =159.4	2.26

[a]Absolute value of nonlinear coefficients is based on d_{36} (KDP) = 1.15×10^{-9} [esu].
Conversion: d [As/V^2] = 8.855 15 × 10^{-12} d [m/V] = 3.68×10^{-15} d [esu]

Table-4. Parameters of nonlinear crystals for critically phase matched second harmonic generation

Property	β-BaB_2O_4	$KNbO_3$	$KTiOPO_4$	LiB_3O_5	$LiNbO_3$	$LiIO_3$
Type	I	I	II	II	I	I
d_{eff} (pm / V)	2.1	11.2	3.3	0.7	5.0	1.9
Angular bandwidth (mrad-cm)*	0.7	0.5	1.7	25.0	0.9	0.5
Spectral bandwidth (cm^{-1}cm)*	35	3.9	9.6	200	4.8	9.5
Damage threshold (J cm^{-2}) at 1064 nm	13	2	15	25	10	1

Other Materials for Second Harmonic Generations

So far we have only considered inorganic piezoelectric crystals as the major second-order nonlinear materials for highly efficient SHG, OPO and OPA. Actually researchers have also paid considerable attention to exploiting organic bulk crystals for SHG. Of all the reported organic materials for second-order nonlinear optical applications, the crystals of urea, NPP [N-(4-nitrophenyl)-L-prolinol], and DAST [4-

dimethylamino-N-Methystilbazolium tosylate] can offer a superior performance for SHG of OPO. Compared to inorganic crystals, the organic compounds may have higher second-order nonlinearity, but sometimes might be limited by the available crystal size and lower optical damage threshold.

Material Parameters:

For the design of a device such as a frequency doubler or parametric oscillator, physical properties of the nonlinear material, such the value of the nonlinear coefficient, damage threshold, absorption losses, and phase-matching parameters, are of importance. For the updated OPO devices pumped with high-peak power pulsed laser beam, the conversion efficiency from the pump pulse energy to the output pulse energy can be higher than (50 ~ 70) %. The interest in OPOs, demonstrated as far back as 1965, has been revived recently due to the availability of efficient nonlinear crystals, LD pumped laser systems and high damage threshold optics. Beta Barium Borate (BBO), Lithium Triborate (LBO), Cesium Lithium borate (CLBO) and Potassium Titanyl Phosphate (KTP) have superior properties listed in Table-1 for application in the uv-near IR optical band. These include a high effective nonlinear coefficient, good optical transparency, small walk-off and a high laser damage threshold.

As seen from Table-2, KTP has a relatively large effective nonlinear coefficient and transmission in the near-infrared with moderate walk-off and damage threshold which makes it an excellent crystal for near-infrared applications. Its high resistance to moisture is most advantageous for use in the laboratory environment. KTP gives very good efficiency for OPO of Nd:YAG laser due to quasi-critical phase matching ($\theta = 90^0$; $\phi \sim 24^0$) scheme which has low walk-off, high angular acceptance and high effective nonlinear coefficient. Walk-off in acentric birefringent crystals like KTP results poor coupling between the interacting ordinary and extraordinary waves and therefore in low conversion efficiencies. Walk-off becomes zero when the pump beam propagates in a principal plane of the crystal. The values of the angular and spectral acceptance bandwidth of the crystal are decided by the particular phase matching scheme and impose a limit on the pump divergence and spectral width. The nonlinear coefficients of KDP and its isomorphs are all within a factor of 2. ADP has the largest and RDA has the smallest nonlinear coefficient shown in Table-3. Point group $\bar{4}2m$ allows three nonzero coefficients d_{14}, d_{25}, and d_{35}. Because of crystal

symmetry, all three are equal, thus for experimental purposes only one coefficient need be quoted (shown in Table-1). As a summery, Table-4, lists some experimental data for typical pulsed SHG, OPA, OPO devices. Table-1 summarizes the measured values of d_{il} and other related data of commonly used crystals for SHG. Table-4, illustrates the values of d_{eff} for uniaxial crystals of 11 point groups and for biaxial crystals of 4 point groups under light propagation in the principal planes.

4.3 Summary:

1. The nonlinear optical processes are shown by lasers, by this process light of one colour (wavelength) to be converted into the light of different colours. Nonlinear optics deals with various new optical effects arising from the interactions of intense coherent optical radiation with matter.
2. For making the intense laser, whose wavelength is unsafe for eye, safe for eye, the optical parametric oscillation (OPO) method is used. The OPO process is a second order nonlinear optical process for which the second-order susceptibility $\chi^{(2)}$ is responsible.
3. The simplest second-order process is that of second-harmonic generation (SHG). In this process, an intense laser beam of angular frequency ω_1 (= ω) is passed through a crystal having nonzero value of $\chi^{(2)}$, such that the beam emerging from the crystal contains the angular frequencies $\omega_1$of the input beam and also $\omega_2 = 2\omega_1$, twice the frequency of the input beam.
4. Generally in second-order processes two photons with different frequencies ω_1 and ω_2, when combines generates a new frequencies, the sum of the two original frequencies i.e, $\omega_1 + \omega_2$, known as *sum frequency generation* and difference of two frequencies, $\omega_1 - \omega_2$, referred to as *difference frequency generation*.
5. In the sum frequency generation, when a new photon $\omega_3 = \omega_1 + \omega_2$ is created, the frequencies ω_1 and ω_2 are destroyed. In the difference frequency generation, the photon of higher frequency ω_1 is destroyed while both ω_2 and ω_3 are created.
6. In the process of difference frequency mixing, the frequency ω_2 is amplified while the frequency ω_3 is being generated. In the process of optical parametric oscillation (OPO) the intense input laser beam at frequency ω_p is known as the

pump frequency, when passes through a nonlinear material, generates the desired frequencies ω_s (signal frequency) and the frequency ω_i (idler frequency). The amplification can be enhanced by placing the optical harmonic (nonlinear) crystal within an optical cavity in which the mirrors are specifically made reflective at either one of these two frequencies, or for both.

7. All of the frequencies $2\omega_1$, $2\omega_2$, $\omega_1 + \omega_2$ and $\omega_1 - \omega_2$ might simultaneously be generated when two input beams of frequency ω_1 and ω_2 are transmitted through the nonlinear optical material. There is a constraint known as *phase matching* that must be implemented in order to efficiently generate any of those additional frequencies.
8. There are two principal methods for achieving phase matching in birefringent optical materials, angle tuning and temperature tuning of the crystals.
9. In this chapter we study about different types of nonlinear crystals which can be used for the OPO processes, but we used here the KTP crystal as a nonlinear material, due to its favourable optical and physical properties in the near infrared.

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