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NONLINEAR CRYSTALS FOR FREQUENCY CONVERSION

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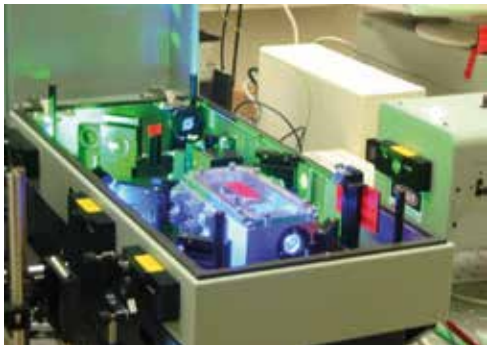

Crystals can convert wavelengths emitted by lasers using a second-order process. They allow light to be generated with smaller or higher wavelengths than those of the input beam. The issue is to select the nonlinear crystal having the best conversion efficiency at the targeted wavelengths, which requires a high non-linearity and suited transparency range as well as phase-matching conditions.

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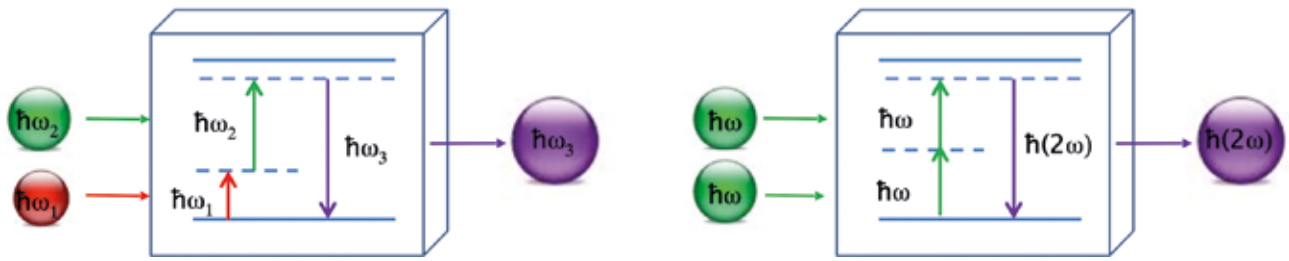
There is an ongoing need for sources able to emit a coherent radiation from the ultra-violet up to the far infrared range, but requested wavelengths are not always provided by commercial lasers. In this case, the best alternative is to use nonlinear light-matter interactions in a bulk crystal. These interactions can be described by using the dual nature of light.

QUANTUM PICTURE

Nonlinear optics can be seen as processes based on fusion or splitting of photons. The effects that are mainly considered involve three photons, which corresponds to second-order nonlinear interactions as shown in Figs. 1 and 2. The horizontal continuous lines depict the matter energy levels and the dashed lines correspond to the energy levels of the electromagnetic field. In the case of fusion, two incident photons with energy quanta $\hbar\omega_1$ and $\hbar\omega_2$, give birth to a higher energy photon at $\hbar\omega_3 = \hbar\omega_1 + \hbar\omega_2$ according to the energy conservation (see Fig. 1 left where $\omega_1 \leq \omega_2 < \omega_3$). The probability of occurrence of this elementary process is maximal when the photon momentum conservation is achieved, *i.e.* $\hbar\vec{k}_3 = \hbar\vec{k}_1 + \hbar\vec{k}_2$ where the $\vec{k}_i$ (i = 1, 2, 3) are the wave vectors. Fusion corresponds to the second-order sum-frequency

Figure 1: Quantum picture for fusion: left SFG and right SHG



generation (SFG), which is then an up-conversion process. If the two incident photons have the same quantum energy $\hbar\omega$, then a photon of twice a quantum energy $\hbar(2\omega)$ is generated, which is called second-harmonic generation (SHG) (see Fig. 1 right).

The spontaneous splitting of one incident photon with a quantum energy $\hbar\omega_3$ into two photons with energy quanta $\hbar\omega_1$ and $\hbar\omega_2$ is shown in Fig 2 left. It is the direct reverse process of fusion so that it fulfils the same photon energy and momentum conservation relations. Usually, the photon at $\hbar\omega_3$ is called the pump, and the generated photons at $\hbar\omega_2$ and $\hbar\omega_1$ the signal and idler, respectively. This elementary process is also called spontaneous parametric down conversion (SPDC). The splitting of one incident photon at $\hbar\omega_3$, can also be stimulated by a photon at $\hbar\omega_1$ or $\hbar\omega_2$ (see for example Fig. 2 right): it corresponds to both optical parametric amplification (OPA) of the stimulating photon at $\hbar\omega_2$ and to difference frequencies generation (DFG) at $\hbar\omega_1 = \hbar\omega_3 - \hbar\omega_2$ which corresponds to a "new" photon. When $\hbar\omega_2 = \hbar\omega_3$, DFG is called optical rectification (OR). In realistic situations, there is a cascading of SPDC and OPA/DFG, which is called Optical Parametric Generation (OPG). And when the nonlinear crystal is inserted in a resonant cavity, we have an Optical Parametric Oscillation (OPO) [1]. Using wavelengths, it comes $\lambda_3 < \lambda_2 \leq \lambda_1$ fulfilling $\frac{1}{\lambda_1} + \frac{1}{\lambda_2} = \frac{1}{\lambda_3}$.

WAVES PICTURE

From the waves point of view, the incident light electric field gives birth to a matter induced polarization $\vec{P}$ that radiates a light electric field $\vec{E}$ becoming a source of excitation for the volume of adjacent matter, and so on. For frequency conversion processes, the interest is the spectral distribution of the radiated electric fields $\vec{E}(\omega_i)$ where i = 1, 2 or 3. They are determined from solving the equation of propagation of matter using a perturbative approach where the matter induced polarization $\vec{P}(\omega_i)$ is expanded as a Taylor power series of the excitation electric field. The first-order is the so-called linear induced polarization $\vec{P}^{(1)}(\omega_i)$: it is radiated at the same circular frequency ω_i as the exciting electric field $\vec{E}(\omega_i)$ by involving the first-order electric susceptibility $\chi^{(1)}(\omega_i)$ of the crystal. $\chi^{(1)}(\omega_i)$ is a second-rank polar tensor described by a diagonal 3×3 matrix when it is expressed in the dielectric frame (**x**, **y**, **z**): the three coefficients allow to define the three principal refractive indices $n_x(\omega_i)$, $n_y(\omega_i)$ and $n_z(\omega_i)$. The higher orders of $\vec{P}(\omega_i)$ correspond to the nonlinear matter induced polarization. We will limit our purpose to the second-order induced polarization $\vec{P}^{(2)}(\omega_i)$, corresponding to three-wave interactions governed by the second-order electric susceptibility $\chi^{(2)}(\omega_i)$ of the crystal. In the case of SFG ($\omega_3 = \omega_1 + \omega_2$) for example, the exciting electric fields $\vec{E}(\omega_1)$ and $\vec{E}(\omega_2)$ give rise to the nonlinear induced polarization $\vec{P}^{(2)}(\omega_3 = \omega_1 + \omega_2)$ that radiates an electric field at $\vec{E}(\omega_3)$ [1]. $\chi^{(2)}(\omega_i)$ is a third-rank polar tensor described by a 3×9 matrix, or by a 3×6 matrix when using a contracted notation. Note also that the writing convention $d^{(2)}(\omega_i) = \frac{1}{2}\chi^{(2)}(\omega_i)$ is often used. The number of non-zero coefficients of the $\chi^{(2)}$ tensor depends on the point group of the crystal [1]. In this frame work, it is important to notice that the only point groups allowing non-zero $\chi^{(2)}$ coefficients are necessary non-centrosymmetric. Then a centrosymmetric crystal prohibits any second-order frequency conversion process. Let's go back to the example of SFG. The goal is to have a constructive interference between the nonlinear polarization $\vec{P}^{(2)}(\omega_3 = \omega_1 + \omega_2)$ and its radiated electric field $\vec{E}(\omega_3)$. That can be achieved if $\Delta\vec{k} = \vec{k}_3 - (\vec{k}_1 + \vec{k}_2) = \vec{0}$, which is called the phase-matching (PM) relation and corresponds to the photon momentum conservation mentioned above. In that case, the energy generated at the circular frequency ω_3 grows continuously over the crystal length, which is of prime interest for applications since very high energy conversion efficiencies close to 100% can be reached [2]. When $\Delta\vec{k} = \vec{k}_3 - (\vec{k}_1 + \vec{k}_2) \neq \vec{0}$, the interference between $\vec{P}^{(2)}(\omega_3 = \omega_1 + \omega_2)$ and $\vec{E}(\omega_3)$ is destructive so that the generated energy oscillates with a spatial period $L_c = \pi/|\Delta\vec{k}|$ that defines the parametric •••

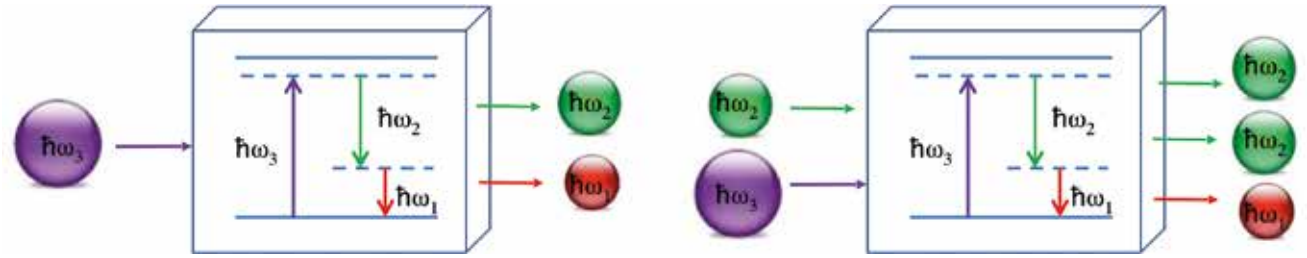


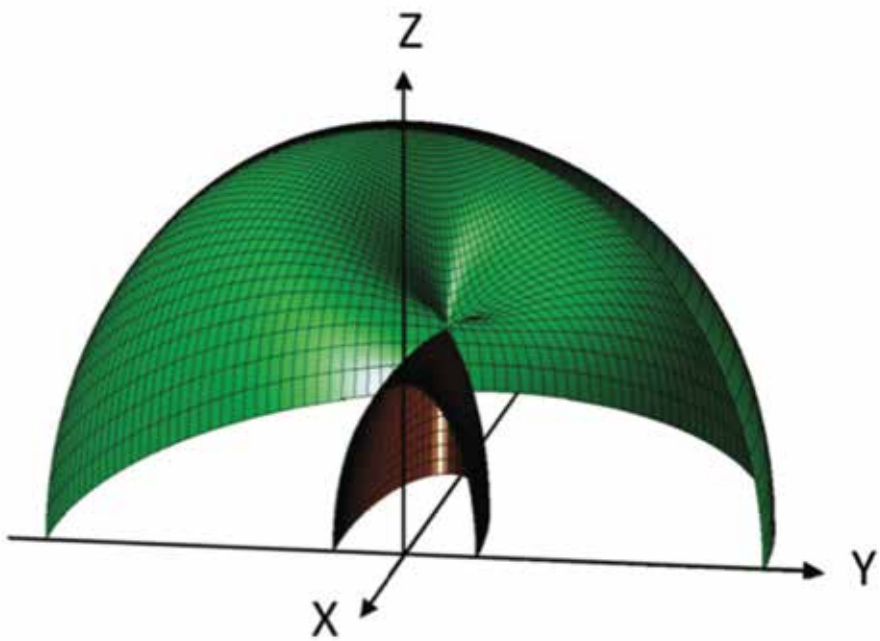
Figure 2: Quantum picture for spontaneous and stimulated splitting: Left is SPDC and right is OPA &DFG

coherent length ranging typically between 1 µm and 100 µm. However, an alternative is the technique of quasi-phase-matching (QPM) that allows the phase-mismatch $\Delta\vec{k} \neq \vec{0}$ to be compensated by the periodic modulation of the sign of the non-linearity over a period equal to 2 times L_c. This configuration is that achieved in ppKTP, ppLN or OPGaAs [3]. Another condition to perform an efficient conversion efficiency is to find a crystal with a high non-linearity. In the equations, it appears at the level of the so-called effective coefficient χ_{eff} which depends on the coefficients of the $\chi^{(2)}$ tensor and the directions of the light electric fields $\vec{E}(\omega_1)$, $\vec{E}(\omega_2)$ and $\vec{E}(\omega_3)$, each corresponding to two Eigen polarization directions $\vec{E}^{+}(\omega_i)$ and $\vec{E}^{-}(\omega_i)$ associated with the considered direction of propagation [1]. These Eigen modes are connected to the index surface described in the next section.

LINEAR CRYSTAL OPTICS

Crystals are cut oriented with the highest precision (up to 0.01°) in their crystallographic frame (**a**, **b**, **c**) by using X-rays diffraction techniques. Then it is mandatory to determine the relative orientation between the crystallographic frame and the dielectric frame in which all optical properties are described. Orthorhombic, monoclinic and triclinic crystals belong to the biaxial optical class: when the principal refractive indices verify $n_x(\lambda_i) < n_y(\lambda_i) < n_z(\lambda_i)$, the crystal is called a positive biaxial crystal; the relation of order is the reverse in the case of a negative biaxial crystal. Tetragonal, hexagonal and trigonal crystals belong to the uniaxial optical class and the same relations of order apply according to its sign, knowing that there are two independent principal refractive indices: $n_x(\lambda_i) = n_y(\lambda_i)$, called the ordinary principal index $n_o(\lambda_i)$, and $n_z(\lambda_i)$ which is the extraordinary principal index written $n_e(\lambda_i)$. Cubic crystals belong to the isotropic optical class where $n_x(\lambda_i) = n_y(\lambda_i) = n_z(\lambda_i)$. The magnitude of the principal refractive indices increases when the wavelength decreases, which is well described by the Sellmeier equation [1]. Such a behavior is classically determined by using the prism technique or from the interpolation of phase-matching curves recorded by using the sphere method for example [4]. Given a wavelength, refractive indices have an angular distribution depicted in the dielectric frame by the index surface. It is a surface with inside and outside sheets labeled (-) and (+) respectively. For a given direction of propagation, their distance from the origin corresponds to the refractive index n^- and n^+, $(n^+ - n^-)$ being called the birefringence. Figure 3 shows the index surface of a positive biaxial crystal. The two sheets are associated to two possible linear Eigen modes of the light electric field $\vec{E}^{-}(\omega_i)$ and $\vec{E}^{+}(\omega_i)$. The orientation of these two Eigen modes is a function of the direction of propagation which is an important issue to consider for calculating the value of the effective coefficient χ_{eff} [1].

Figure 3: Index surface of a positive biaxial crystal at a given wavelength using the convention $n_x < n_y < n_z$ [5].



PHASE-MATCHING CONDITIONS

For colinear interactions, the angular distribution of PM directions is found by solving:

$$\Delta k = 2\pi\left[\frac{n^{\pm}(\lambda_3, \theta_{PM}, \varphi_{PM})}{\lambda_3} - \frac{n^{\pm}(\lambda_2, \theta_{PM}, \varphi_{PM})}{\lambda_2} - \frac{n^{\pm}(\lambda_1, \theta_{PM}, \varphi_{PM})}{\lambda_1}\right] = 0$$

$(\theta_{PM}, \varphi_{PM})$ are the PM angles of spherical coordinates, expressed in the dielectric frame. In that case, one speaks of Birefringence Phase-Matching (BPM). It is easy to write the corresponding code, but there exists also the commercial code SNLO [6]. Such a calculation is mandatory before selecting a nonlinear crystal especially when the three interacting wavelengths are not classically used. Otherwise they are already provided by several compagnies for non-resonant second-order processes widely used in devices, as for example SHG of a commercial laser emitting at $\lambda_\omega = 1.064$ µm to generate $\lambda_{2\omega} = 0.532$ µm. It is necessary to have principal refractive indices that must have been determined with an accuracy better than 10^{-4} to be able to calculate the BPM angles with an accuracy better that 1°. There are three possible combinations of refractive indices leading to the three BPM types [1]:
type I $[n^-(\lambda_3), n^+(\lambda_1), n^+(\lambda_2)]$,
type II $[n^-(\lambda_3), n^-(\lambda_1), n^+(\lambda_2)]$,
type III $[n^-(\lambda_3), n^+(\lambda_1), n^-(\lambda_2)]$.

To these three types are associated three different configurations of Eigen modes polarization that it is important to know in order to be able to calculate the effective coefficient χ_{eff} as mentioned in the previous section. It is then necessary to properly polarize the incident beams. Note that BPM directions are associated to spectral, angular and thermal acceptances [1].

COMMERCIAL NONLINEAR CRYSTALS

Several commercial nonlinear crystals cut as slabs with very high optical quality and more or less large dimensions can be purchased to convert wavelengths emitted by commercial lasers from BPM or QPM second-order processes. The main suppliers are given in the table below. They can even provide PM conditions as a function of temperature. Take care of maximal values of incoming energies for not reaching the crystal optical damage threshold and Fresnel losses reducing conversion efficiencies. Losses can be decreased by a coating but the optical damage threshold might decrease too. When commercial nonlinear crystals do not correspond to your request, this article and references provide all elements to calculate BPM and QPM conditions and the associated conversion efficiencies as well as the spectral, angular and thermal acceptances.

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COMPANY	WEBSITE	MANUFACTURER OR DISTRIBUTOR	MAIN BULK NONLINEAR CRYSTALS FOR SALE
ALPHALAS	https://www.alphalas.com/products/laser-components/nonlinear-crystals.html	Custom Manufacturer	LBO, BBO, BiBO, KTP & ppKTP
ARDOP	https://www.ardop.com/details-cristaux+non+lineaires-54.html	Distributor	BBO, LBO, KDP, DKDP, KTP, KTA, AGS, AGSe, AgSe
ARTIFEX	https://artifex-engineering.com/optics/crystals/	Distributor	BBO, KTP, LBO, LN
BAE SYSTEMS	https://www.baesystems.com/en/product/nonlinear-crystal-technologies	Custom Manufacturer	ZGP, CSP, ppGaAs, ppGaP
BLUEBEAM	http://www.bbotech.com/product.php?cid=55	Custom Manufacturer	BBO, LBO, KTP, KTA, BiBO, LN
CASIX	https://www.casix.com/en/productList.html?TypeID=13966	Custom Manufacturer	BBO, LBO, KTP
CASTECH	https://www.castech.com/product/NLO-Crystals-33	Stock & Custom Manufacturer	LBO, BBO, BiBO, CLBO, KTP, RTP, KTA, LN, MgO:LN, KDP, DKDP, LiIO3
COVESION	https://covesion.com/en/	Stock & Custom Manufacturer	MgO:ppLN
CRISTAL LASER	https://www.cristal-laser.com/	Custom Manufacturer	LBO, KTP, KTA, RTP
CRYSTECH	http://www.crystech.com/product/8/	Custom Manufacturer	KTP, KTA, BBO, BiBO, LN
CRYSTRO	https://www.tggcrystal.com/supplier-nonlinear_crystals-434498.html	Distributor	KTP, LBO, BBO, LN
EDMUND OPTICS	https://www.edmundoptics.com/f/nonlinear-crystals/39487/	Distributor	BBO, LBO
EKSMA OPTICS	https://eksmaoptics.com/nonlinear-and-laser-crystals/nonlinear-crystals/	Custom Manufacturer	BBO, LBO, KDP, DKDP, KTP, CLBO, KTA, AGS, AGSe, AgSe, ZGP, LiO3, LN
FOCTECK	https://www.foctek.net/Products/list-93.html	Custom Manufacturer	BBO, KTP, LN
GAMDAN OPTICS	https://www.gamdan.com/	Stock & Custom Manufacturer	LBO, BBO, KTP
GWU	https://gwu-lasertechnik.de/crystals/	Distributor	BBO, LBO, KTP, KTA, BiBO, CLBO, RTP, MgO:LN, DKDP, KDP, LiIO3
INNOWIT	https://www.innowit.com/en/cate-1929.html	Distributor	KTP, KTA, KDP, DKDP, LBO, BBO, RTP
LASERTON	https://laserton.com/index.php?m=content&c=index&a=lists&catid=36	Custom Manufacturer	BBO, KTP, DKDP, LN, LBO, KTA, RTP
OPTOAXIS	http://www.optoaxis.com/NLO-Crystals.html	Custom Manufacturer	LN, BBO, LBO, DKDP, KTP
PRINCETON SCIENTIFIC	https://princetonscientific.com/materials/nlo-crystals/	Custom Manufacturer	KTP, KTA, KDP, BBO, LN, MgO:LN, LT
RAICOL CRYSTALS	https://raicol.com/	Stock Manufacturer	KTP, ppKTP, BBO, RTP, LBO
RAINBOW PHOTONICS	http://www.rainbowphotonics.com/	Stock Manufacturer	DAST, DSTMS, OH1, KNbO3
SHALOM EO	https://www.shalomeo.com/Laser-Crystals-and-Components/Nonlinear-Crystals	Stock & Custom Manufacturer	BBO, DKDP, KTP, LBO, BiBO, ZGP, KTA, LiIO3, LN, MgO:LN, RTP, YCOB