Ćwiczenie 4

1. Sprawdź, czy potrafisz odpowiedzieć na następujące pytania:

   - Od jakich parametrów zależy wydajność generacji II harmonicznej w kryształach?
   - Co to jest dopasowanie fazowe? Pokaż, że dopasowanie fazowe charakteryzuje prawo zachowania pędu.
   - Co to jest dwójłomność kryształu, jakie znasz typy kryształów dwójłomnych?

2. Jak wiesz generowanie II harmonicznej jest procesem, który możemy przedstawić następująco:

$$\omega_1 + \omega_1 = 2\omega_1$$

$$\vec{k}_1 + \vec{k}_1 = \vec{k}_2; \qquad \vec{k}_2 = 2\vec{k}_1$$

czyli: $\Delta\vec{k} = \vec{k}_2 - 2\vec{k}_1 = 0$

Rozróżniamy dwa typy generowania II harmonicznej:

I typ: o+o=e (czyli oddziałują dwa fotony o polaryzacji promienia zwyczajnego, generując foton o polaryzacji promienia nadzwyczajnego)

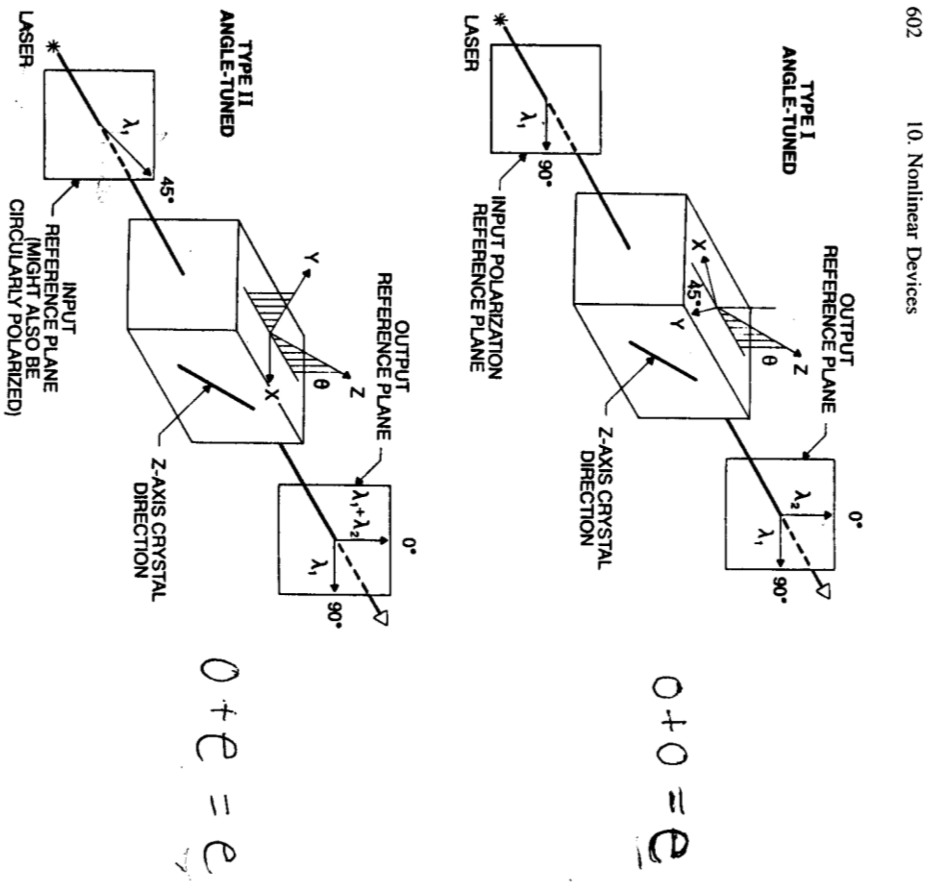
II typ: e+o=e (czyli oddziałują dwa fotony o polaryzacji:(e i o), generując foton o polaryzacji promienia nadzwyczajnego)

Na rysunku 1 zilustrowano I i II typ generowania drugiej harmonicznej. W różnych kryształach realizowany jest bądź I typ, bądź II typ, a w niektórych oba typy generacji drugiej harmonicznej.

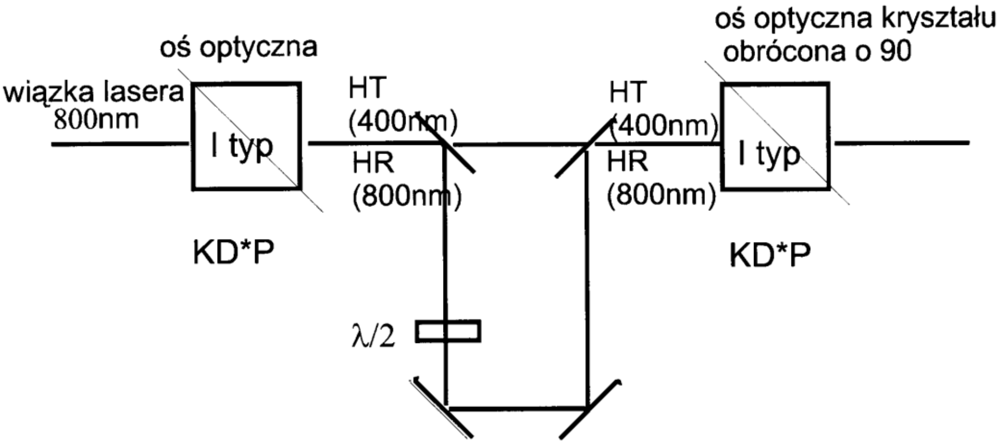
3. W pracowni laserowej spektroskopii molekularnej używamy kryształu KD*P (KD_2PO_4, czyli kwaśnego deuterowanego fosforanu potasu) oznaczanego niekiedy również $DKDPO_4$.
Sprawdź i napisz w sprawozdaniu:

   - Jaki typ drugiej harmonicznej jest generowany w KD*P?
   - Jaki jest kąt dopasowania fazowego?
   - Wymień wszystkie parametry jakich używa się do scharakteryzowania własności kryształów w optyce nieliniowej

4. Na rysunku 2 przedstawiono schemat generacji drugiej i trzeciej harmonicznej w kryształach KD*P w naszym laboratorium. Polaryzacja wiązki wejściowej jest horyzontalna (promień zwyczajny (o) względem osi optycznej pierwszego kryształu)
   Określ polaryzację drugiej i trzeciej harmonicznej.



Rys.2



Narysuj na rysunku polaryzację poszczególnych wiązek w różnych miejscach schematu.

Materialy pomocnicze z HANDBOOK OF NONLINEAR OPTICAL CRYSTALS , V.G. Dmitriev, G.G. Gurzadyan, D.N. Nikogosyan, Springer-Verlag 1991

Surface-damage thresholds I at different wavelengths and pulse durations:

λ [μm]	τ_p [ns]	I [10^9 W cm^{-2}]	Ref.
0.53	0.2	17	[3.13]
0.6943	20	0.4	[3.14]
1.06	0.01	100	[3.15]
1.06	0.2	23	[3.13]
1.06	1	> 10	[3.16]
1.065	15	14.4	[3.17]
1.06	4.7	16.6	[3.18]

Damage thresholds under focusing with a lens having a focal length F [3.19]:

λ [μm]	τ_p [ps]	F [mm]	E [μJ]
0.53	10	20	30
0.53	10	50	70
1.06	10	20	300
1.06	10	50	600

Breakdown thresholds for a high-purity KDP sample due to multiphoton ionization for bandwidth-limited light pulses (the radiation propagates along the Z axis and is polarized along the X axis) [3.20]:

λ [μm]	τ_p [ps]	I [10^{12} W cm^{-2}]
0.3547	17	2.5–5.0
0.532	21	1.1
1.064	30	2.3

Linear absorption coefficient α at various wavelengths:

λ [μm]	α [cm^{-1}]	Ref.
0.1765	1.2	[3.2]
0.35–0.53	< 0.005	[3.21]
0.78	0.024	[3.22]
0.89	0.015	[3.22]
1.06	0.03–0.05	[3.21,22]
1.315	0.3	[3.23]

Two-photon absorption coefficient β at various wavelengths [3.24]:

λ [μm]	β [cm W^{-1}]
0.266	2.7×10^{-10}
0.355	5.9×10^{-12}

Heat-conductivity coefficient κ[W cm^{-1} K^{-1}] at various temperatures T [3.25]:

$\kappa, \parallel c$	$\kappa, \perp c$	T [K]
0.0121	–	302
–	0.0134	319
0.0130	0.0176	428

3.1.2 KD_2PO_4, Deuterated Potassium Dihydrogen Phosphate (DKDP)

Negative uniaxial crystal: $n_o > n_e$
Point group: $\bar{4}2$ m
Transparency range: 0.2–2.0 μm [3.3,26]
Dispersion relations ($T = 300$ K) [3.5] (λ in μm):

$$n_o^2 = 1.661145 + \frac{0.586015\,\lambda^2}{\lambda^2 - 0.016017} + \frac{0.691194\,\lambda^2}{\lambda^2 - 30},$$

$$n_e^2 = 1.687499 + \frac{0.44751\,\lambda^2}{\lambda^2 - 0.017039} + \frac{0.596212\,\lambda^2}{\lambda^2 - 30}.$$

Other dispersion relations are given in [3.6,7].
Typical values of refractive indices:

λ [μm]	n_o	n_e
0.266	1.5546	1.5085
0.347	1.5278	1.4854
0.355	1.5263	1.4841
0.532	1.5085	1.4690
0.658	1.5032	1.4645
0.694	1.5020	1.4635
1.064	1.4928	1.4555
1.315	1.4867	1.4502

Temperature dependences of refractive indices (T in K, λ in μm) [3.5]:

$$n_o^2 = (1.55934 + 3.3935 \times 10^{-4}\,T) + \frac{(0.71098 - 4.1655 \times 10^{-4}\,T)\lambda^2}{\lambda^2 - (0.01407 + 6.4904 \times 10^{-6}\,T)} + \frac{(0.67671 + 4.8281 \times 10^{-5}\,T)\lambda^2}{\lambda^2 - 30},$$

$$n_e^2 = (1.68647 + 3.43 \times 10^{-6}\,T) + \frac{(0.46629 - 6.26 \times 10^{-5}\,T)\lambda^2}{\lambda^2 - (0.01663 + 1.3626 \times 10^{-6}\,T)} + \frac{(0.59614 + 2.41 \times 10^{-7}\,T)\lambda^2}{\lambda^2 - 30}.$$

Changes in refractive indices Δn upon cooling from room temperature (298 K) to T[K] can also be calculated by the equations [3.8]

$$\Delta n_o = n_o(T) - n_o(298) = (298 - T)(n_o^2 - 1.047) \times 0.228 \times 10^{-4},$$

$$\Delta n_e = n_e(T) - n_e(298) = (298 - T)n_e^2 \times 0.955 \times 10^{-5}.$$

Changes in refractive indices with temperature for a wavelength range 0.41–0.63 μm [3.6]:

$$\frac{\partial n_o}{\partial T} = -3.1 \times 10^{-5}\,(\mathrm{K})^{-1},$$

$$\frac{\partial n_e}{\partial T} = -2.1 \times 10^{-5}\,(\mathrm{K})^{-1}.$$

Effective nonlinearity [3.9]:

$$d_{ooe} = d_{36} \sin\theta \sin 2\varphi,$$

$$d_{eoe} = d_{oee} = d_{36} \sin 2\theta \cos 2\varphi.$$

Nonlinear coefficient [3.10,27]:

$$d_{36}(1.06\,\mu\mathrm{m}) = (0.92 \pm 0.04)\, d_{36}(\mathrm{KDP}) = (4.02 \pm 0.17) \times 10^{-13}\ \mathrm{m/V}.$$

Surface-damage thresholds:

λ [μm]	τ_p [ns]	I [10^9 W cm^{-2}]	Ref.
1.06	10	0.5	[3.28]
1.06	0.25	6	[3.29]

Linear absorption:

λ [μm]	α [cm^{-1}]	Ref.
0.53	0.005	[3.29]
1.06	0.005	[3.29]
1.315	0.025	[3.23]

Two-photon absorption:

λ [μm]	β [cm W^{-1}]	Ref.
0.266	2.7×10^{-11}	[3.30]
	2.0×10^{-11}	[3.31]
0.355	5.4×10^{-12}	[3.24]

3.1.3 $NH_4H_2PO_4$, Ammonium Dihydrogen Phosphate (ADP)

Negative uniaxial crystal: $n_o > n_e$
Point group: $\bar{4}2$ m
Transparency range: 0.184–1.5 μm [3.1,3.7]
Dispersion relations ($T = 24.8\,°$C, λ in μm) [3.32]:

$$n_o^2 = 2.302842 + \frac{0.011125165}{\lambda^2 - 0.013253659} + \frac{15.102464\,\lambda^2}{\lambda^2 - 400},$$

$$n_e^2 = 2.163510 + \frac{0.009616676}{\lambda^2 - 0.01298912} + \frac{5.919896\,\lambda^2}{\lambda^2 - 400}.$$

Other dispersion relations are given in [3.5–7].
Typical values of refractive indices ($T = 25\,°$C):

λ [μm]	n_o	n_e
0.213	1.6271	1.5684
0.266	1.5791	1.5261
0.347	1.5499	1.5004
0.355	1.5481	1.4989
0.532	1.5275	1.4815
0.694	1.5193	1.4754
1.064	1.5065	1.4681

nonlinear index and the parameters defining the tolerance to angular, wavelength, and temperature detuning from phase-matched conditions.

In the analysis mentioned above second-harmonic conversion is characterized by two parameters relating the conversion efficiency, the pump intensity and the nonlinear properties of the material. These are the nonlinear "drive" which is the source term for generation of the electric field at the second harmonic, and the detuning which is the phase mismatch between second-harmonic waves at the exit and entrance planes of the crystal. The drive is given by

$$\eta_0 = C^2 I l^2 \, , \tag{10.30}$$

where C is proportional to the nonlinear coefficient of the material, I is the pump intensity, and l is the crystal length. If d_{eff} is expressed in pm/V and λ in μm, one obtains

$$C = \frac{2.75 d_{eff}}{\lambda_2 n^{3/2}} \, .$$

The detuning is given by

$$\delta = 0.5\,\Delta k l \, , \tag{10.31}$$

where Δk is the wavevector mismatch given by (10.22).

The drive and detuning parameters determine the conversion efficiency

$$\eta = \eta_o (\sin\delta/\delta)^2 \, . \tag{10.32}$$

The use of these equations is illustrated in the following example. The theoretical conversion efficiency of a Q-switched Nd : YAG laser with an output energy of 1 Joule per pulse in a 15 ns pulse is calculated. The laser beam has a divergence of 0.3 mrad and a cross-sectional area of 0.5 cm^2 at the doubling crystal. The harmonic generator is a 2.5 cm long KD*P crystal oriented for type-II doubling. Introducing d_{eff} and n for KD*P, one obtains $C = 0.97\,GW^{-1/2}$ and from the other parameters follows a drive of $\eta_0 = 0.78$. The detuning parameter $\delta = 1.12$ is obtained from $\beta = 2.5\,(mr\text{-}cm)^{-1}$ for KD*P, $\delta\theta' = 0.3$ mrad and $l = 2.5$ cm. Introducing these two parameters into (10.32) yields a theoretical conversion efficiency of $\eta = 0.62$.

10.1.3 Properties of Nonlinear Crystals

Survey of Materials

Tables 10.1 and 10.2 list properties and phasematching parameters for several important nonlinear materials. For the design of a frequency converter the following properties of the nonlinear crystal are of key importance: value of the nonlinear coefficient, damage threshold, phase-matching and transparency range, available crystal size and optical homogeneity, and chemical and mechanical stability. The selection of a particular nonlinear material for use in a solid-state

Table 10.1. Properties of important nonlinear materials

Material	Phase-matching type	Effective nonlinear coefficient* $[10^{-12}$ m/V]	Refractive index $n_0(w)$	Damage threshold [GW/cm^2]	Absorption [cm^{-1}]
KD*P	II	0.37	1.49	0.5	0.005
KTP	II	3.18	1.74	0.5	0.010
LBO	I	1.16	1.56	2.5	0.005
BBO	I	1.94	1.65	1.5	0.005
$LiNbO_3$ (5% MgO)	I	4.7	2.23	0.10	0.002
$LiIO_3$	I	4.1	1.85	0.01	0.002

* For 1064 nm to 532 nm second-harmonic generation

Table 10.2. Phasematching parameters for 1064 to 532 nm conversion

Material	Phase matching angle	Walk-off angle	Tolerance parameters (FWHW) Angular [mr-cm]	Thermal [°C-cm]	Spectral [nm-cm]
KD*P	53.7°	1.45°	1.0	6.7	0.66
KTP	24.3°	0.26°	25	25	0.56
LBO	–	–	4.3	3.6	0.75
BBO	22.8°	3.19°	0.5	55	0.66
$LiNbO_3$ (5% MgO)	90° (1)	0	47	0.6	0.23
$LiIO_3$	30.2°	4.26°	0.7	–	–

(1) at $T = 107$°C

predicated upon high damage threshold and good optical quality, with secondary emphasis placed on the magnitude of the nonlinear coefficient.

For example, despite the lower nonlinear coefficient of KDP compared to the other materials discussed in this section, conversion efficiencies as high as 80 % have been obtained from these crystals. Since a high conversion efficiency is actually the result of material properties as well as pump source characteristics, the high peak power, narrow spectral bandwidth, small beam divergence and clean spatial and temporal beam profiles obtained from Nd : glass lasers employed in fusion research, make possible such high harmonic conversions even in crystals with modest nonlinearity.

From Table 10.1 it is clear that for high-power lasers, such as pumped pulsed, Q-switched systems, our choice of nonlinear materials is limited to crystals belonging to the KDP family, or to KTP and BBO if the small crystal size of these new materials is acceptable. The other nonlinear materials, such as $LiNbO_3$ and $LiIO_3$, have such low damage thresholds that their application has been restricted to harmonic generation of cw-pumped Nd : YAG lasers.

At the present time, the leading candidate material for nonlinear experiments with solid-state lasers is KTP, since the crystal has a large nonlinear coefficient, high damage threshold and large angular and temperature acceptance range. Its