M.A. Kopylovsky, M.S. Brodyn, I.M. Pritula , V.Ya. Gayvoronsky

Impact of the embedded TiO_2 nanoparticles on the second harmonic generation in the KDP single crystals

KDP crystals

- low cost
- possibility of wide aperture crystals growing
- high laser damage threshold
- growth from aqueous solutions

- The relatively small values of d_{36}

ways to improve the properties

The inclusion of organic/inorganic impurities on the growth phase (KCl, Li^{3+}, xylenol orange)

The incorporation of nanoparticles **(SiO_2, CdTe)**

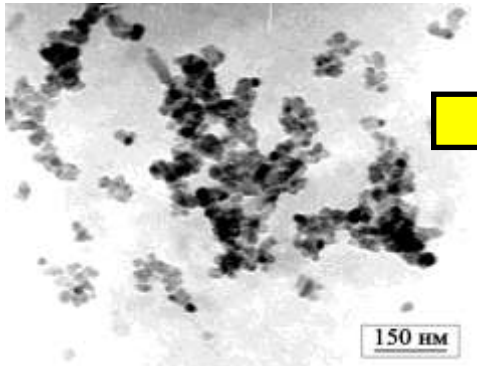
KDP crystal doped dye XO

Functional Materials 15, No.3 (2008) © 2008 — Institute for Single Crystals

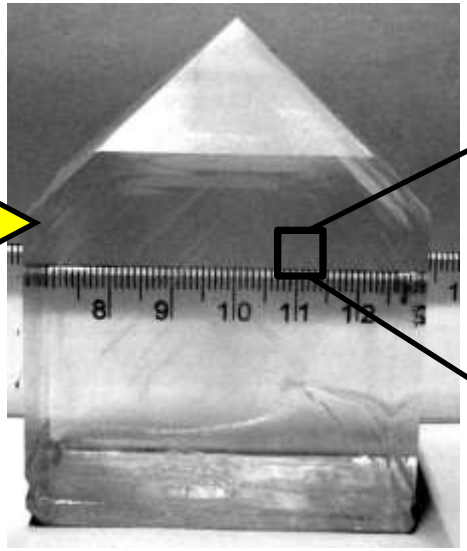
Growth and characterization of KH_2PO_4 single crystals doped with TiO_2 nanocrystals

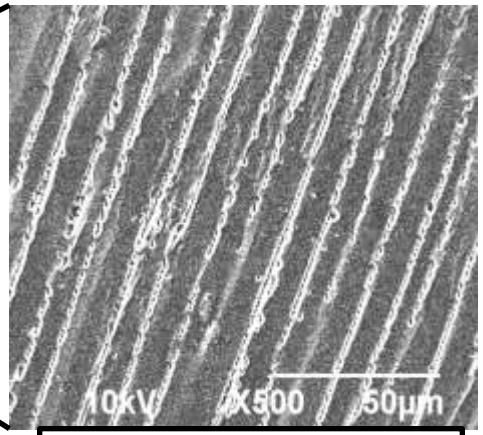
I.Pritula, V.Gayvoronsky, M.Kopylovsky*, M.Kolybaeva, V.Puzikov, A.Kosinova, V.Tkachenko, V.Tsurikov, T.Konstantiniva**, V.Pogibko****

TiO_2 nanoparticles (anatase)

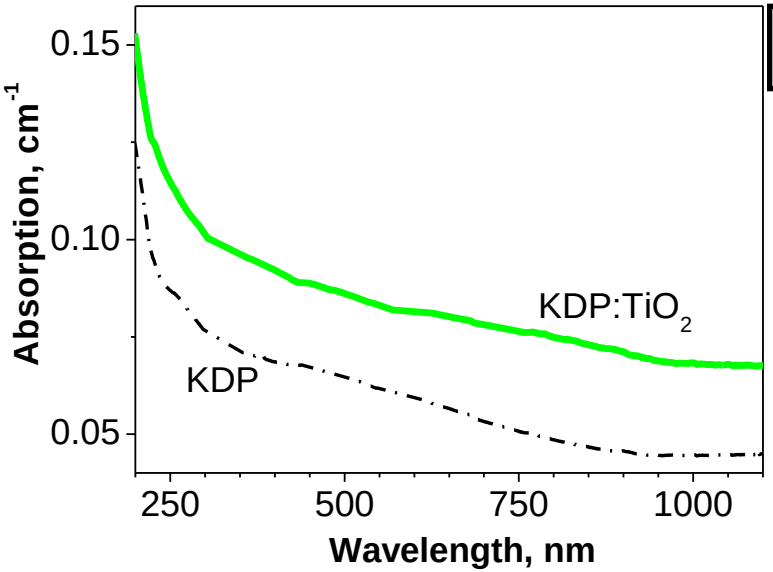


the average NPs size of 15 nm

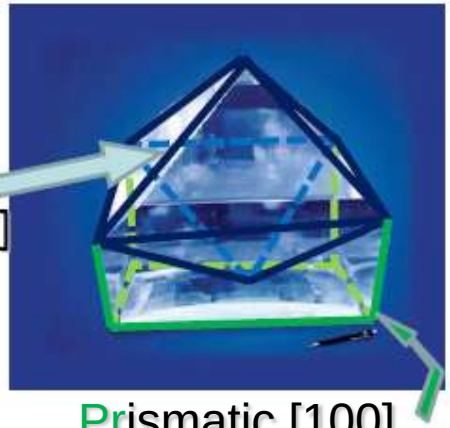


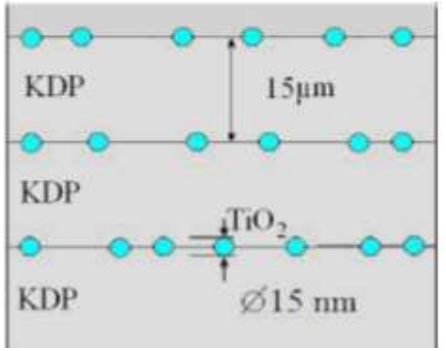


NPs form layers



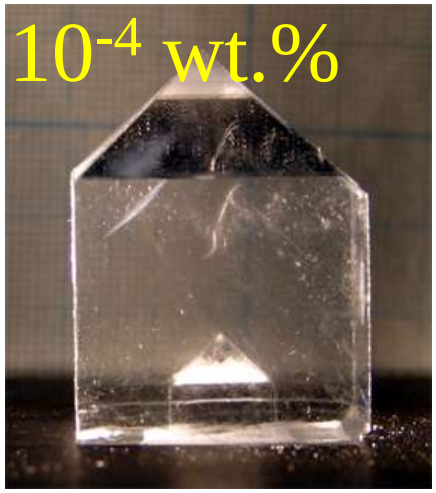
KDP:TiO_2 10^{-4} wt. %

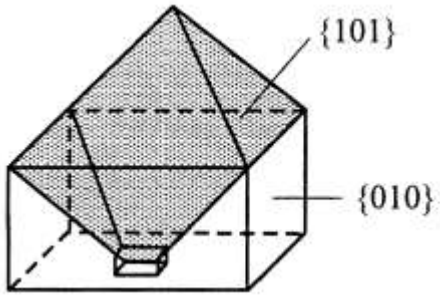


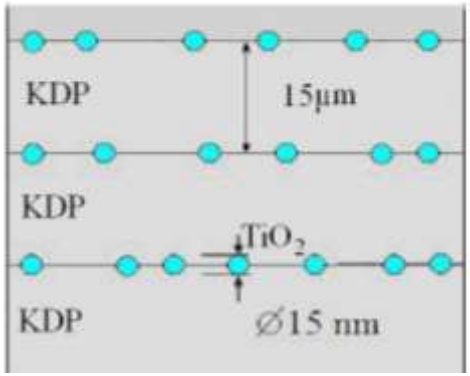


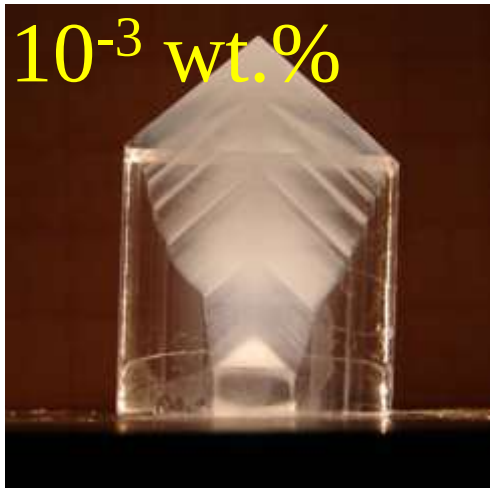
period ~15 mkm

KH_2PO_4 crystals with incorporated anatase nanocrystals - KDP:TiO_2









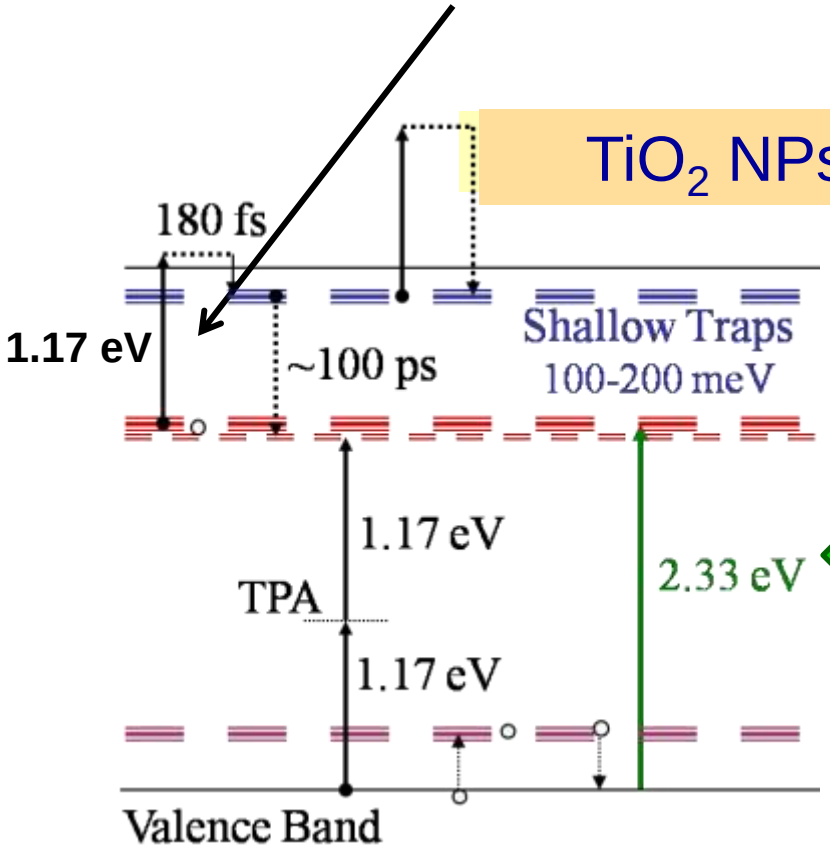
TiO_2 conc., wt. %	Sample notation	d, *mm*	Transmittance, % to inc.	
			@ 532 *nm*	@ 633 *nm*
0	P	0.81	91.3 ± 1.0	91.9 ± 0.9
	Pr	0.83	92.4 ± 0.6	93.0 ± 0.7
10^{-5}	P-5	0.75	89.8 ± 0.9	90.2 ± 1.1
	Pr-5	0.78	90.3 ± 0.9	91.0 ± 1.0
10^{-4}	P-4	0.76	89.8 ± 1.2	90.8 ± 1.0
	Pr-4	0.75	91.4 ± 1.1	92.0 ± 1.0
10^{-3}	Pr-3	0.81	89.1 ± 0.5	89.7 ± 0.5

V.Gayvoronsky, M Kopylovsky et al. Ukr. J. Phys. 55 (2010) 875

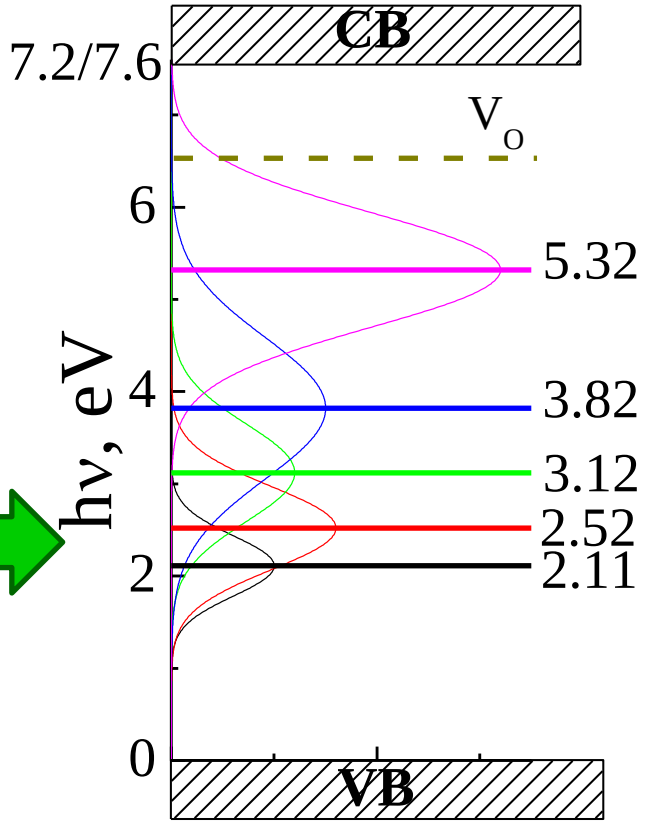
Giant nonlinear response of anatase layers

$\chi^{(3)}_{nano} \sim 10^6 \chi^{(3)}_{bulk}$ @1064 nm

TiO_2 NPs



KDP

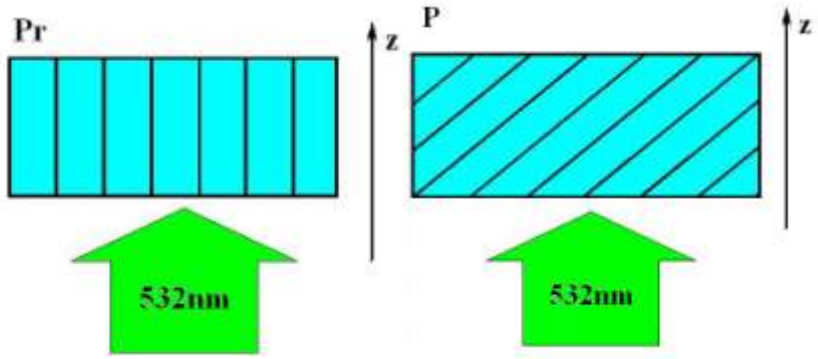


V.Gayvoronsky et al. Appl. Phys. B 80, 97 (2005)

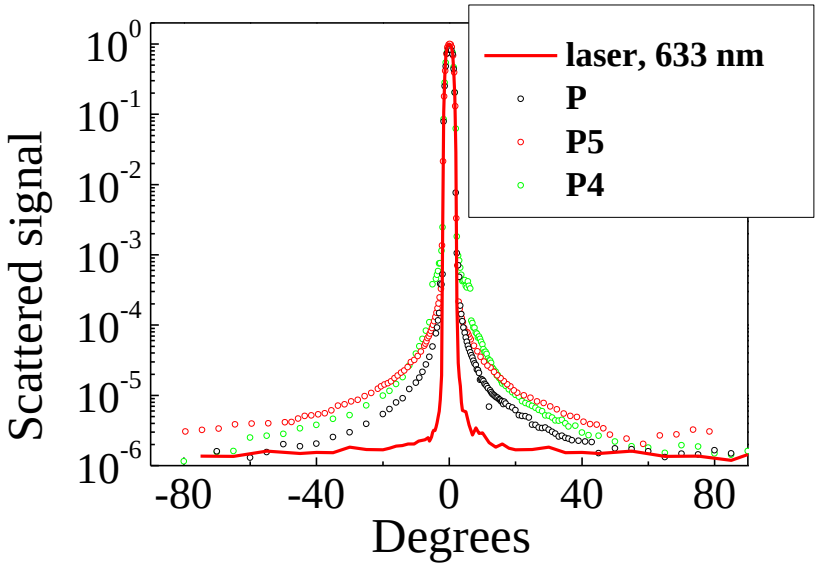
I. N. Ogorodnikov et al. Phys. Sol. State, 44, 880 (2002)

Scattering indicatrix of $KDP:TiO_2$ crystals

$$p_{\text{scat}} = \frac{2\pi}{P_0} \int_{\theta_{\min}}^{\pi/2} \frac{\Delta P(\theta)}{\Delta\Omega} \sin\theta d\theta$$



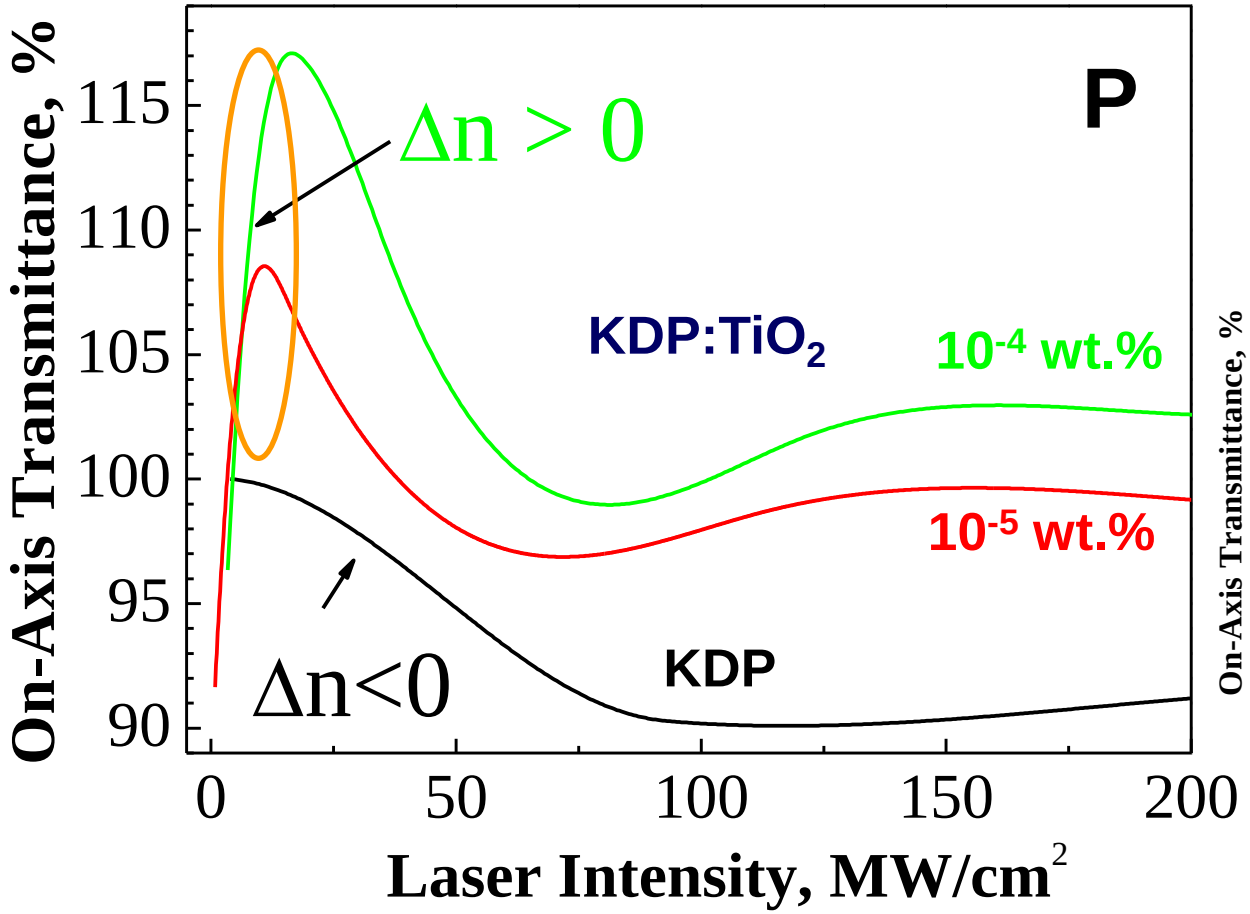
Sam-ple	Scattering losses p_{scat}, %	
	λ=532 nm	λ=633 nm
KDP (P)	1.0	1.0
P5Ti	1.7	1.7
P4Ti	2.5	1.8
KDP (Pr)	1.1	1.1
Pr5Ti	1.7	2.6
Pr4Ti	1.3	2.6
Pr3Ti	2.8	2.6

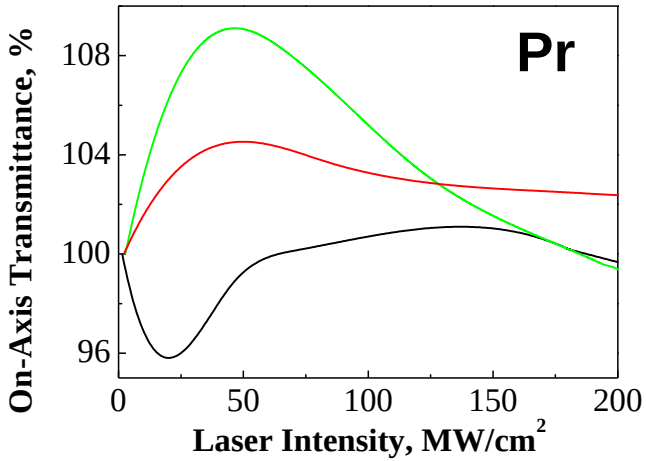


Waveguide effect in $KDP:TiO_2:KDP$ layers

V.Gayvoronsky, M Kopylovsky et al. Ukr. J. Phys. 55 (2010) 875

KDP:TiO_2 : photoinduced changes of on-axis transmittance @ 1064 nm (42 ps FWHM)





Pritula I., Kopylovsky M. et. al. Opt. Mat 33 (2011) 623

Concentration dependence of the real and imaginary parts of the cubic NLO susceptibility of the KDP:TiO_2 crystals @1064 нм

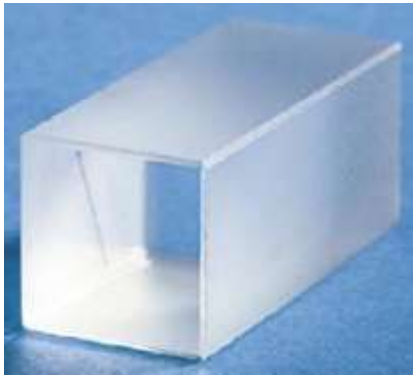
	TiO_2 conc., wt. %	Growth sector	NPs conc., 10^{10} cm^{-3}	I<I_s	
				Re($\chi^{(3)}$), 10^{-8} esu	Im($\chi^{(3)}$), 10^{-12} esu
KDP	-	P	-	-0,07	0,9
		Pr	-	-0,09	0,7
KDP:TiO_2	10^{-5}	P	3,2	2,2	1,6
		Pr	1,6	0,4	1,6
	10^{-4}	P	33	2,5	3,0
		Pr	16	0,7	0,8

Re($\chi^{(3)}$) < 0
self-defocusing

Re($\chi^{(3)}$) > 0
self-focusing

for fixed TiO_2 conc. $Re(\chi^{(3)})_P \sim (4 \div 5) \cdot Re\ (\chi^{(3)})_{Pr}$

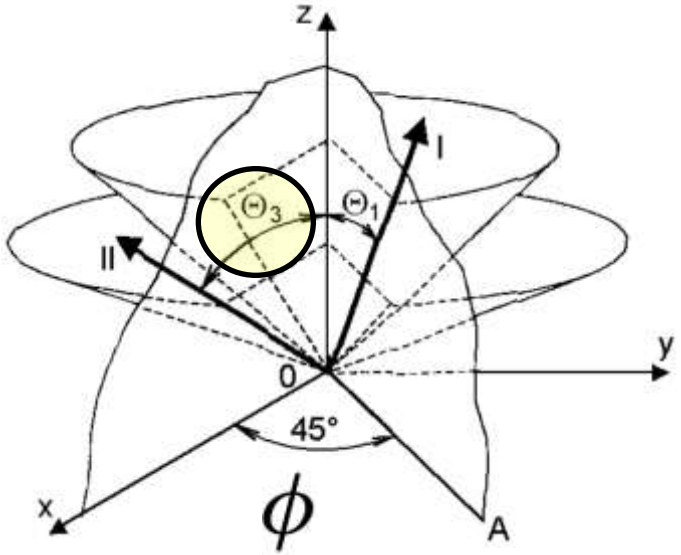
Second harmonic generation in the KDP crystals



Type 1 **oo-e**
Type 2 **oe-e**

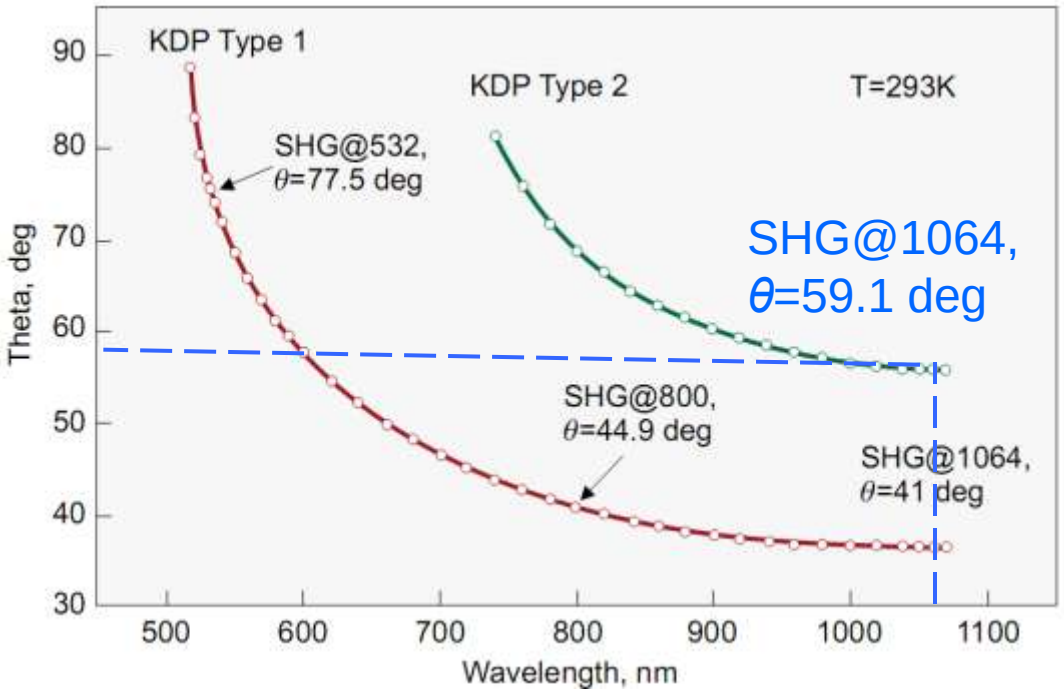
$$d_{ooe} = -d_{36} \sin\theta \sin 2\phi$$

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

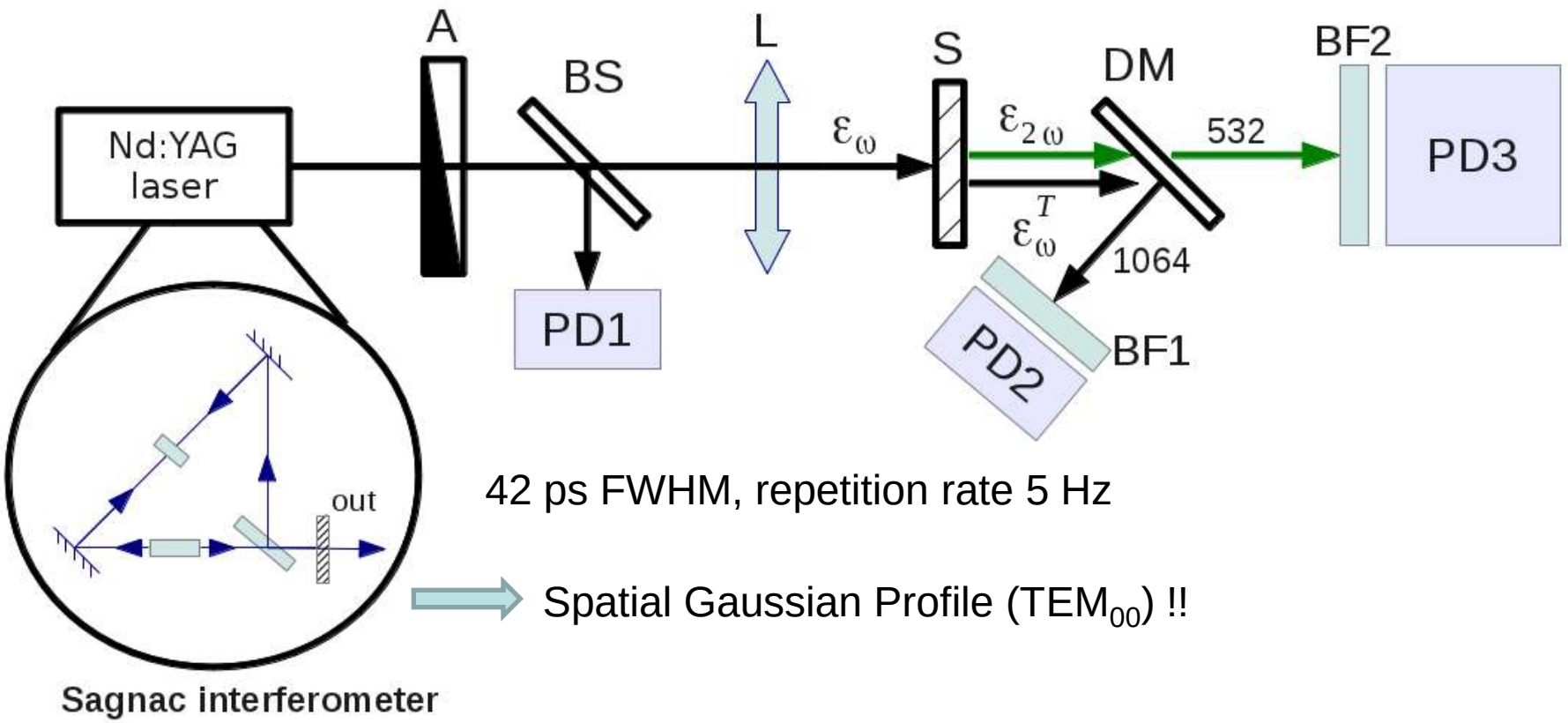


Phase-matching angles for SHG at 1064 nm

	oo-e	**oe-e**
Cut angle θ, deg	41.2	59.1
Acceptances (FWHM):		
Δθ (internal), mrad×cm	1.1	2.2
ΔT thermal, K×cm	11	13.2
Δλ spectral, nm×cm	7.25	5.57
Walk off, mrad	27	24



Investigation of second harmonic generation (SHG)



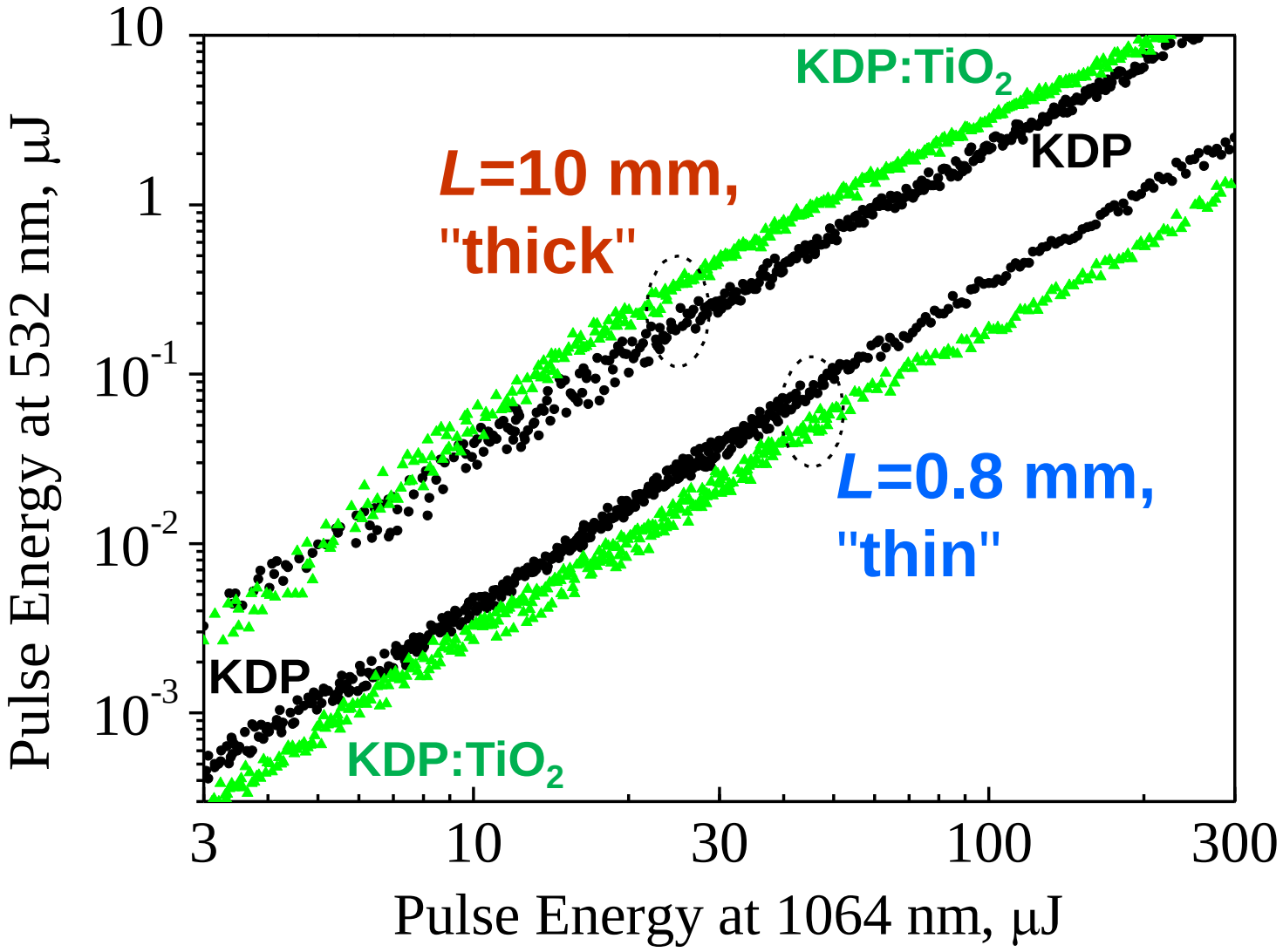
42 ps FWHM, repetition rate 5 Hz

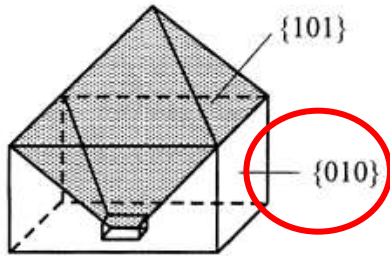
⇒ Spatial Gaussian Profile (TEM_{00}) !!

$$\eta \equiv \frac{\mathcal{E}_{2\omega}}{\mathcal{E}_\omega} = \frac{8\sqrt{2\pi}L^2|d_{eff}|^2 \exp[-L(\alpha_\omega + \alpha_{2\omega}/2)]}{c\tau\varepsilon_0[\lambda_{2\omega}a(1+n_\omega^o)(1+n_\omega^e)(1+n_{2\omega}^e)]^2}\mathcal{E}_\omega$$

given pump field approximation

SHG in KDP, KDP:TiO_2 crystals with single ps pulse @1064 nm

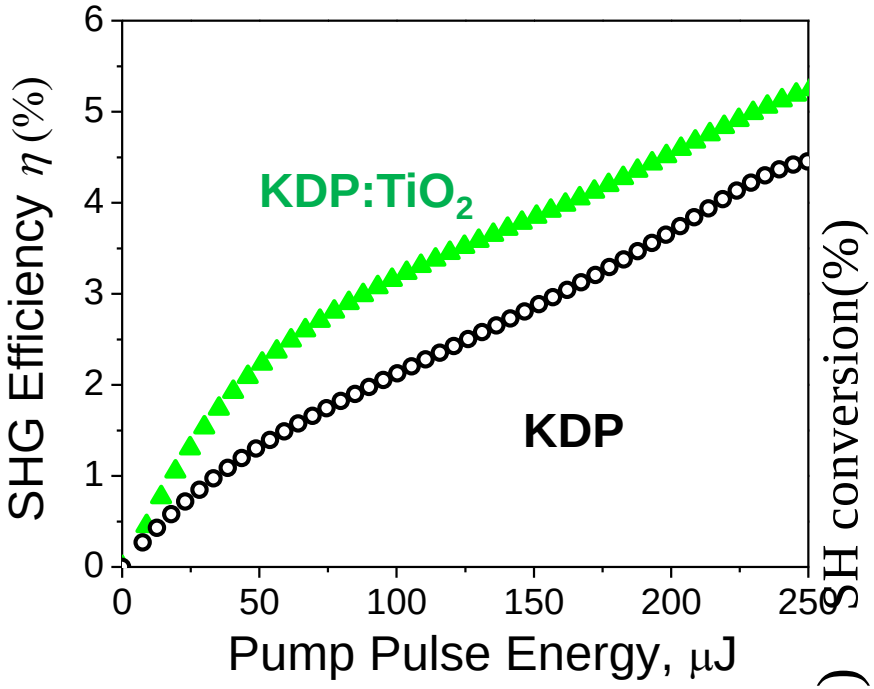




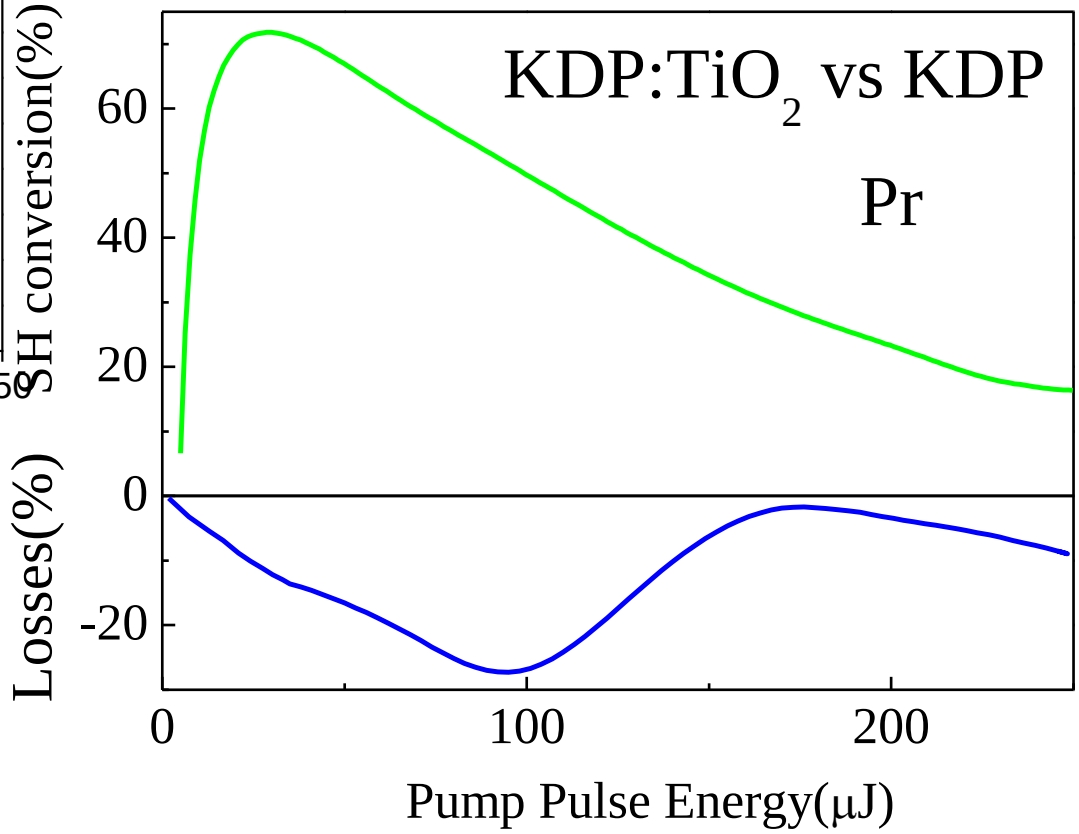
Pr

Gayvoronsky V. Ya., Kopylovsky M. A. et al. Func. Mat. 19 (2012) 54-59.

SHG efficiency in the "thick" samples



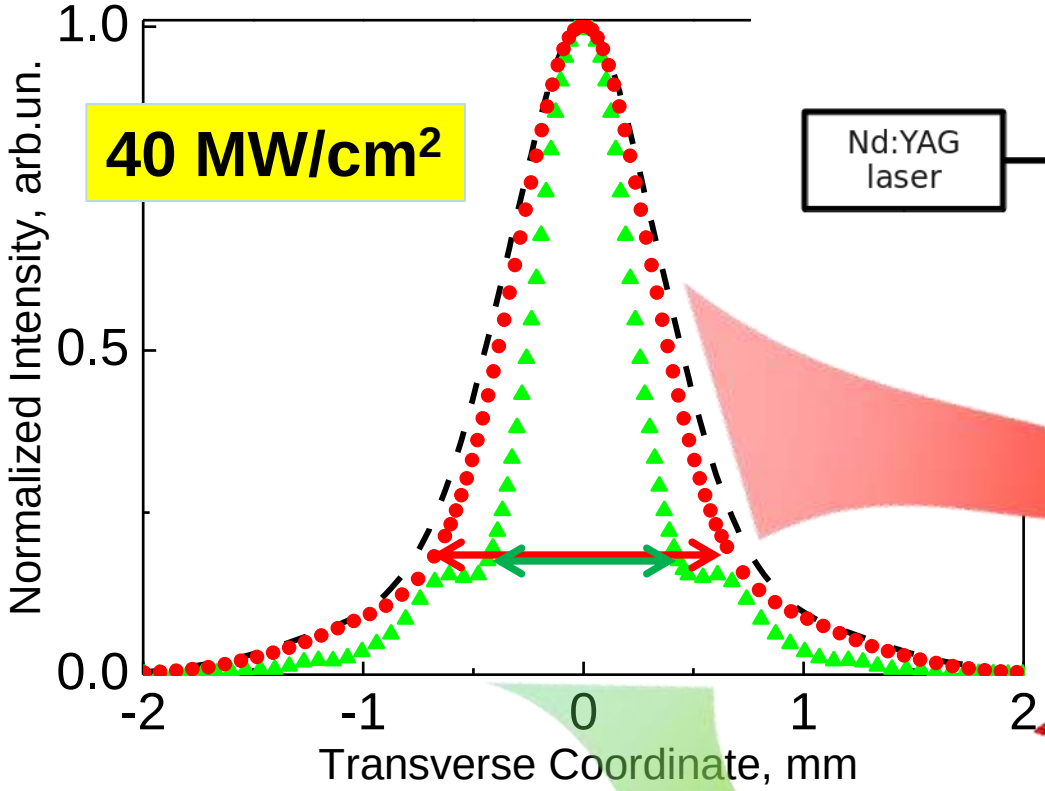
Efficiency increase by 70%

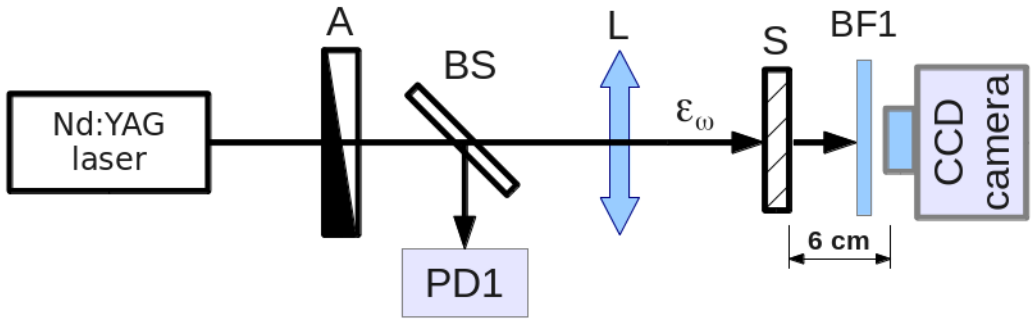


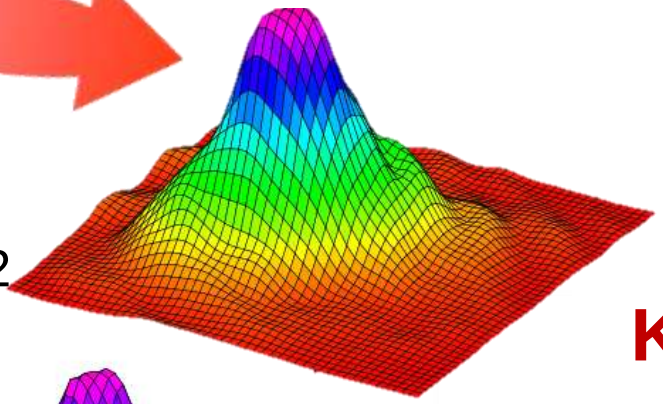
	TiO_2, wt. %	$d_{36\ eff}$, pm/V
KDP	-	0.40
KDP:TiO_2	10^{-4}	0.35

Gayvoronsky V. Ya., Kopylovsky M. A. et al. Func. Mat. 19 (2012) 54-59.

Internal self-focusing

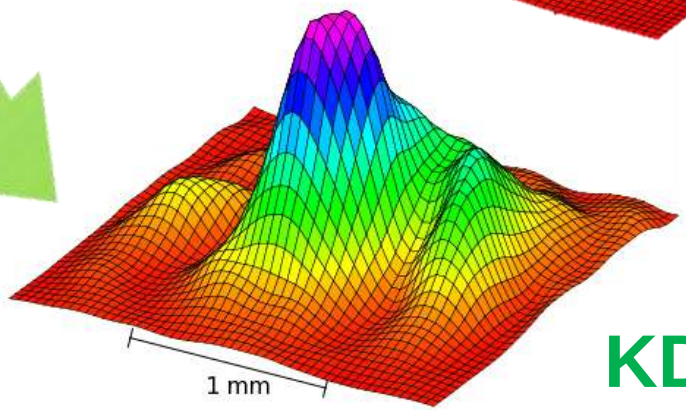






KDP

Beam narrowing to ~ 1.5 times



$KDP:TiO_2$

Gayvoronsky V. Ya., Kopylovsky M. A. et al. Func. Mat. 19 (2012) 54-59.

Contents lists available at ScienceDirect

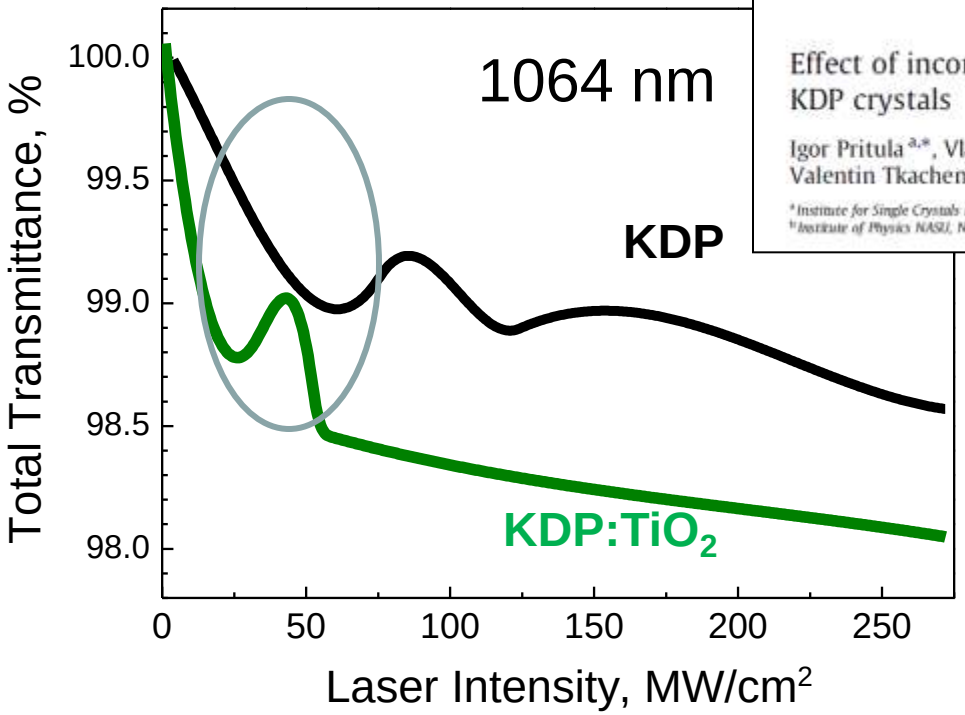
Optical Materials

journal homepage: www.elsevier.com/locate/optmat

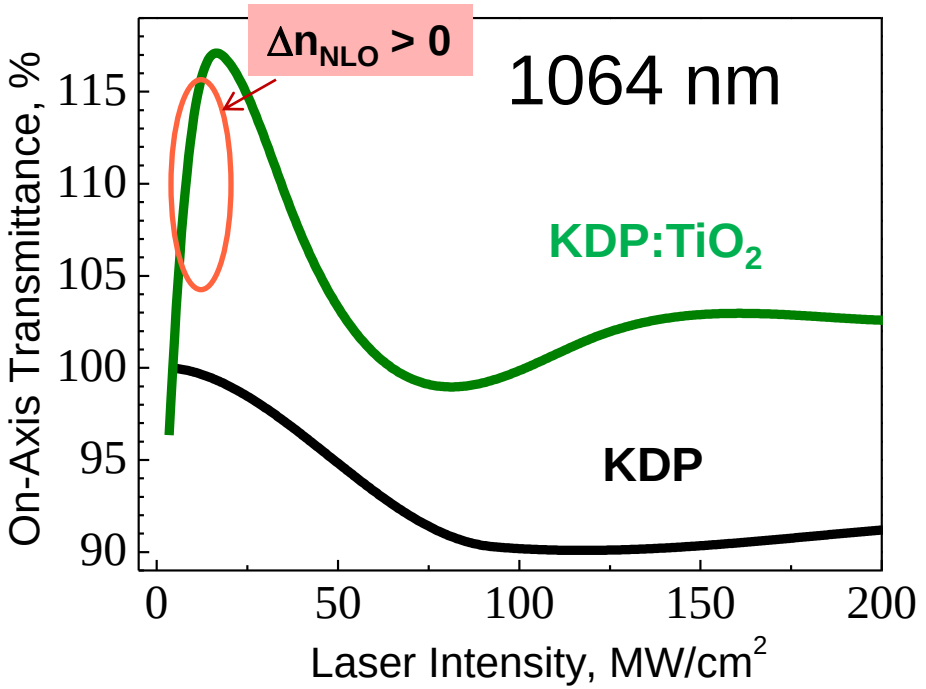
Effect of incorporation of titanium dioxide nanocrystals on bulk properties of KDP crystals

Igor Pritula [a,*], Vladimir Gayvoronsky [b], Marina Kolybaeva [a], Viacheslav Puzikov [a], Mykhaylo Brodyn [b], Valentin Tkachenko [a], Anna Kosinova [a], Maksim Kopylovsky [b], Vitaliy Tsurikov [a], Olga Bezkrovnaya [a]

[a] Institute for Single Crystals NASU, Lenin ave., 60, 61001 Kharkov, Ukraine
[b] Institute of Physics NASU, Nauki ave., 46, 03680 Kyiv, Ukraine



Sample	Re($\chi^{(3)}$), 10^{-8} esu	Im($\chi^{(3)}$), 10^{-12} esu
KDP	-0.07	0.9
KDP:TiO_2	2.2	1.6

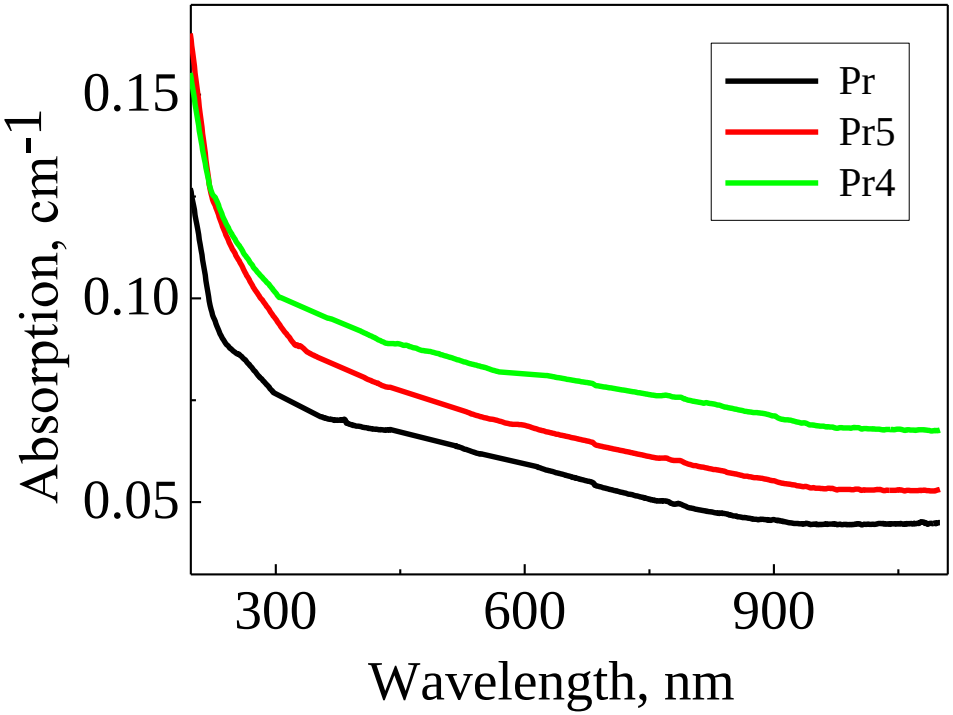


Conclusions

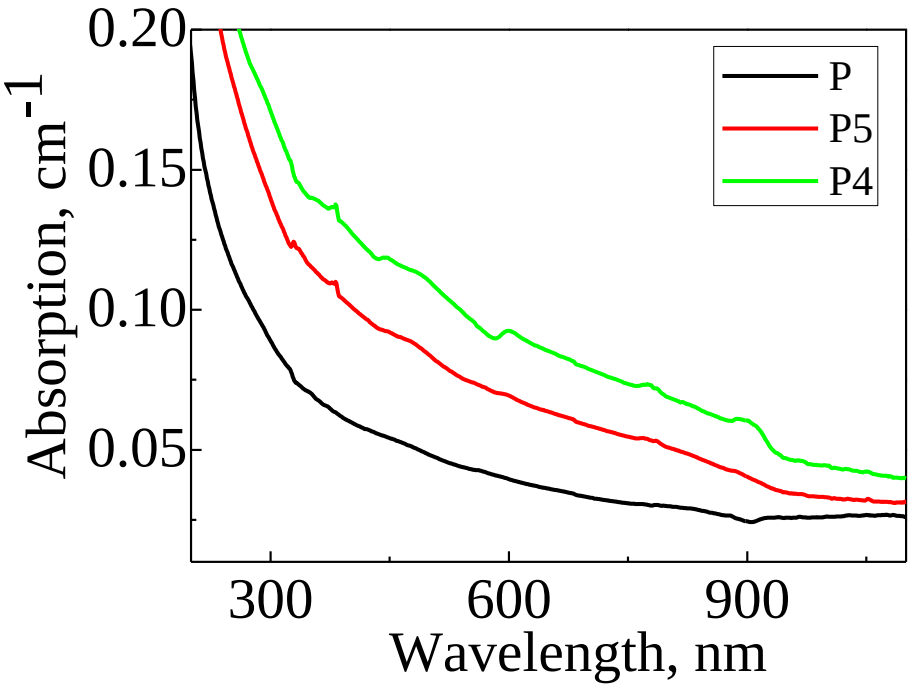
- Second harmonic generation efficiency versus the TiO_2 nanoparticles concentration incorporated in the KDP single crystals was studied within picosecond range pump pulses at 1064 nm.
- For the first time we have obtained the frequency conversion efficiency enhancement up to **70%** in the KDP:TiO_2 in comparison with the KDP crystals due to the internal self-focusing effect that was observed as the pump beam spatial profile narrowing at moderate peak intensities.

Thank you for your attention

Transmittance spectra of
KDP, KDP:TiO_2



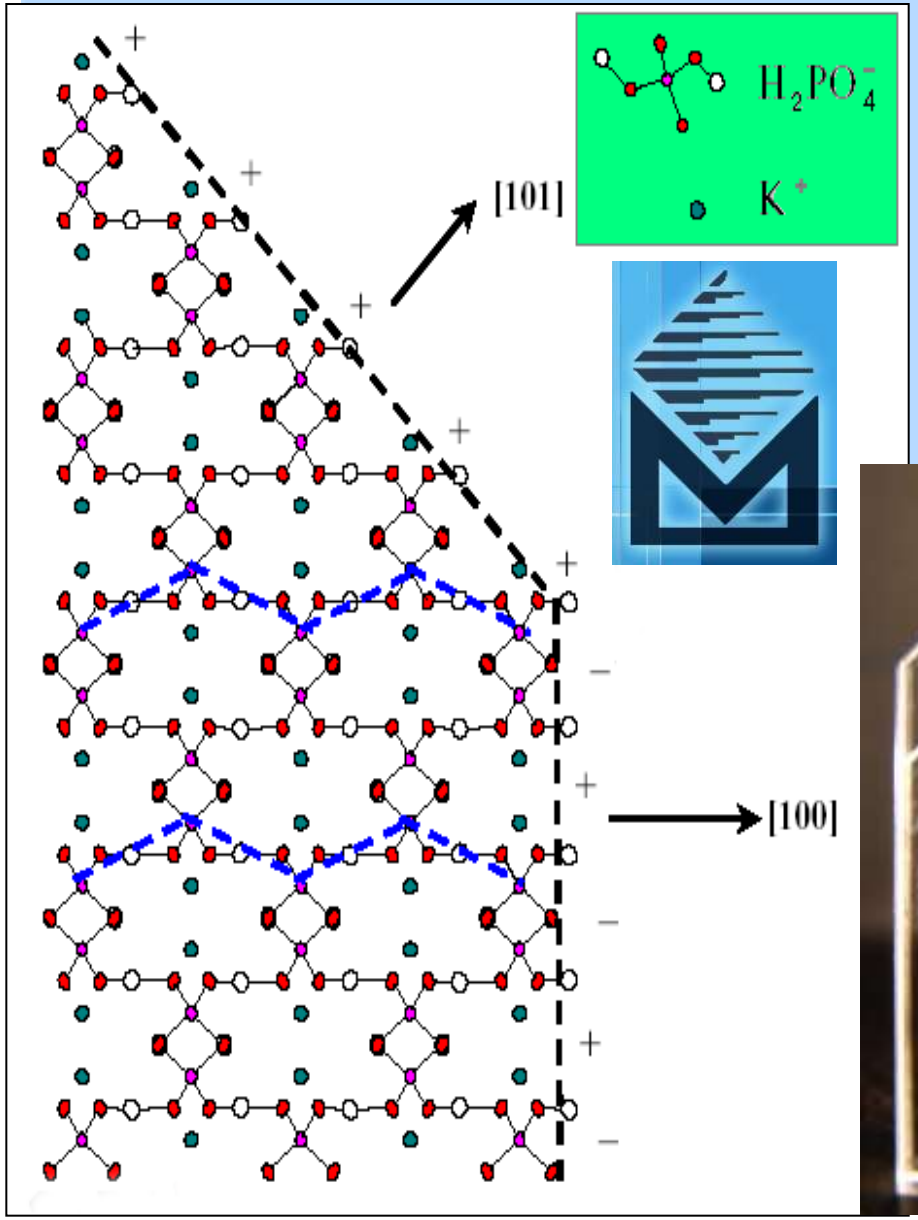
Pr



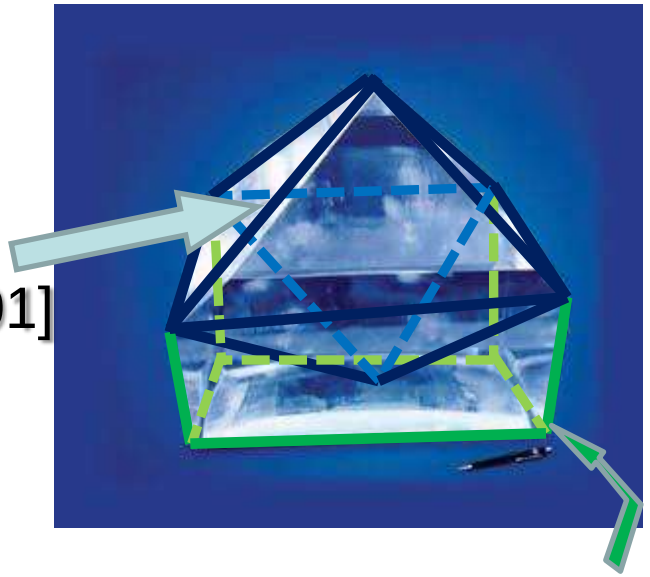
P

V.Gayvoronsky, M Kopylovsky et al. Ukr. J. Phys. 55, 875 (2010)

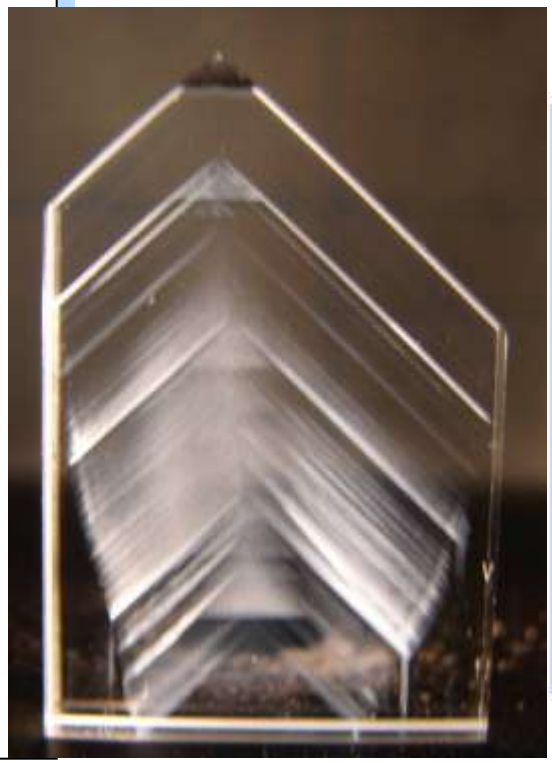
Incorporation of TiO_2 nanoparticles

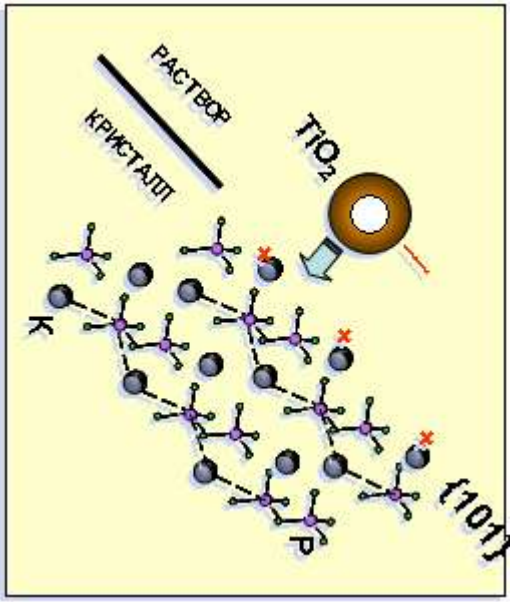


Piramidal [101]



Prismatic [100]





KDP:TiO_2 crystals

- KDP:TiO_2 – growing from an aqueous solution

Pritula I., Gayvoronsky V. et al. Func. Mat. 15 (2008) 420.

- Optical characterization

Gayvoronsky V., Starkov V. et al. Ukr. J. Phys. 55 (2010) 875.

- Cubic NLO susceptibility

Pritula I., Gayvoronsky V. et al. Opt. Mat 33 (2011) 623.

- Second harmonic generation

Gayvoronsky V., Kopylovsky M. et al. Func. Mat. 19 (2012) 54-59.