Nonlinear optical response of ZnO-PMMA nanocomposites under picosecond laser excitation

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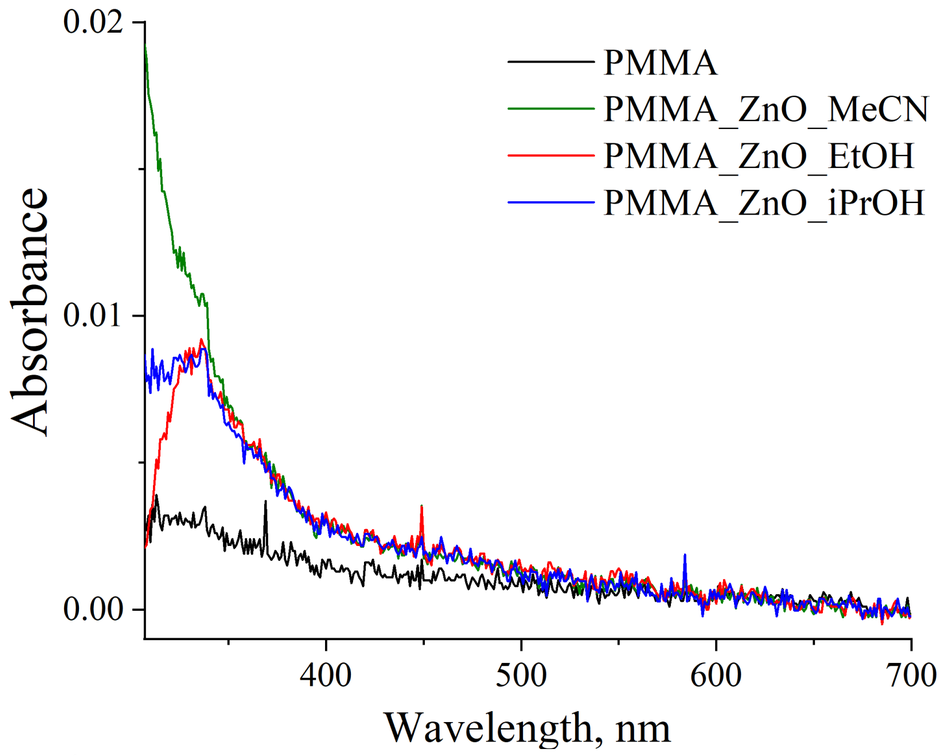
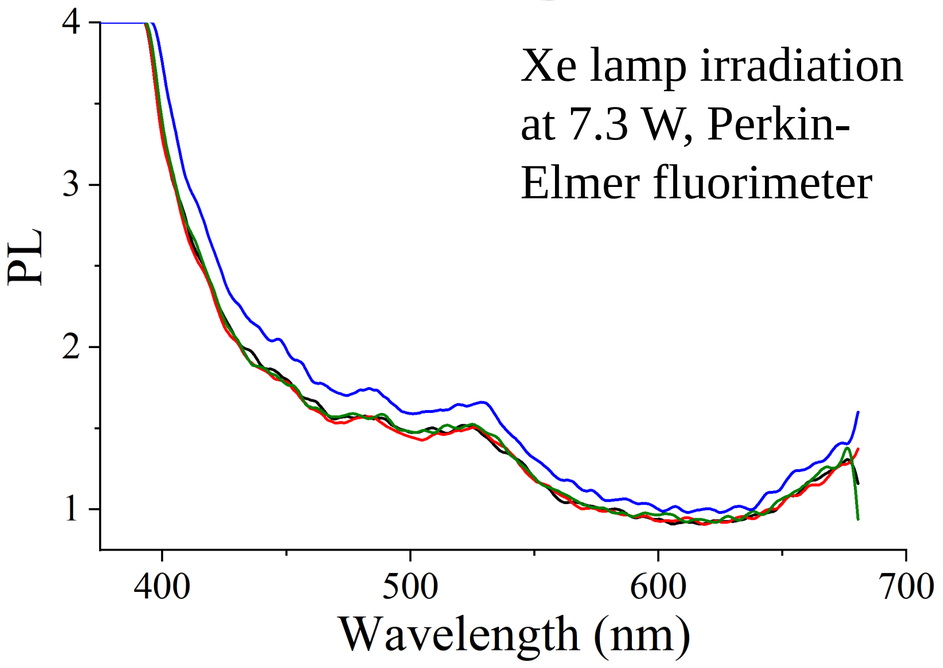
In recent times, remarkable interest has arisen in the development of organic-inorganic hybrid materials. They are based on a polymer matrix with inorganic nanoparticles (NPs) used as solar cells, field-effect transistors, optoelectronics, and nonlinear optical (NLO) devices. However, zinc oxide nanoparticles (ZnO NPs) have potential applications in various fields, including medicine, photovoltaic devices, and nonlinear optics. Ultrasmall **ZnO** NPs (d ~ 2 nm) were prepared by advanced hydrolysis routes from different solvents: i-propyl alcohol (**i-PrOH**), ethanol (**EtOH**), and acetonitrile (**MeCN**). The main idea of this work is to compare the features of ZnO/polymethylmethacrylate (PMMA) composites with the prepared ZnO nanostructures.

Samples characterization

UV-Vis and Luminescence spectra

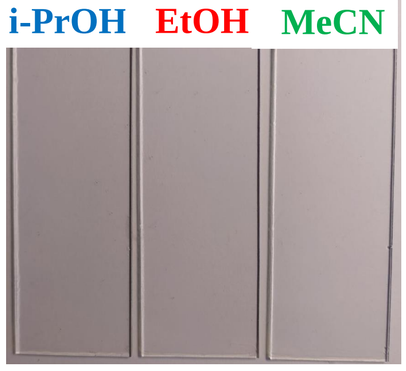




Thin polymeric films under study

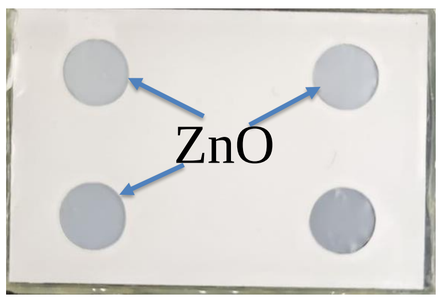
PMMA+1%ZnO

i-PrOH **EtOH** **MeCN**



Previously investigated solid ZnO NPs[1]

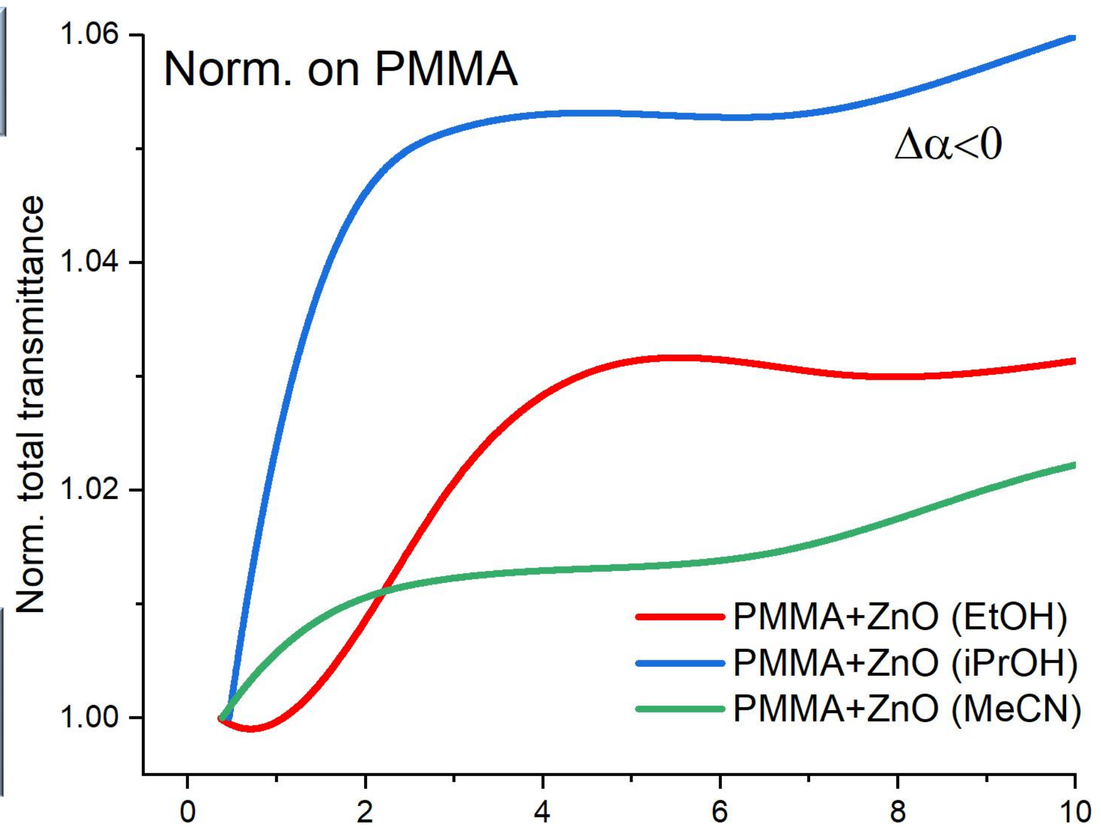
i-PrOH **EtOH**

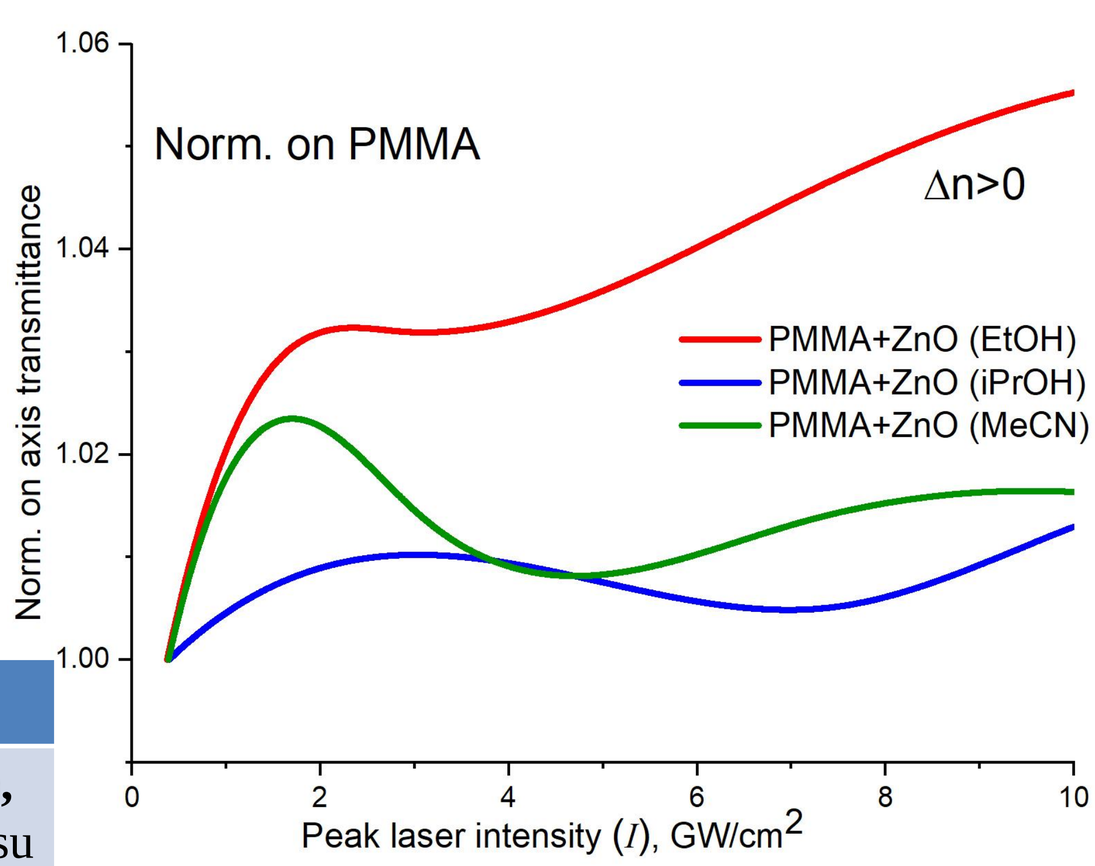


MeCN

Nonlinear Optical (NLO) Response @1064nm

Absorptive Resp. and Refractive Resp.





Solvent	ZnO/PMMA		Solid ZnO NPs[1]	
	Re($\chi^{(3)}$), $\times10^{-11}$ esu	Im($\chi^{(3)}$), $\times10^{-12}$ esu	Re($\chi^{(3)}$), $\times10^{-9}$ esu	Im($\chi^{(3)}$), $\times10^{-11}$ esu
I, GW/cm^2	1 / 8	1 / 8		
EtOH	**21.6 / 1.4**	**-1.5 / -0.6**	**2.4**	**1.4**
MeCN	**11.2 / 1.3**	**-5.9 / -0.3**	**2.9**	**0.5**
i-PrOH	**15.1 / 1.3**	**-17.0 / -2.3**	**-4.2**	**0.7**

References: **1.** V.V. Multian, A.V. Uklein, et al., Nanoscale Res. Lett. **12** (2017) 164. **2.** V.Ya. Gayvoronsky, M.A. Kopylovsky, L.A. Golovan, et al., Quantum Electronics **41** (3) (2011) 257–261.

Conclusions: Analysis of the pulsed laser radiation **@1064nm** self-action effects has shown that the **ZnO NPs** incorporated in a PMMA matrix demonstrated a self-focusing effect ($\Delta n > 0$) with refractive NLO response efficiency **Re($\chi^{(3)}$)** ~10^{-10} esu. It is sensitive to the solvent used at the synthesis stage, as it was observed for the **porous** layers of the same **ZnO NPs** with **Re($\chi^{(3)}$)** ~10^{-9} esu. The self-focusing effect saturates at the peak intensity I of about 2 GW/cm^2. Further growth of the laser intensity provides the refractive NLO response with an order of magnitude lower efficiency **Re($\chi^{(3)}$)** ≈ 1.3×10^{-11} esu, almost similarly for all samples. Obtained data can correspond to the known NLO response of porous silicon [2] with the resonant excitation of the interface electronic states of silicon nanocrystals at the initial stage, while the latter effect is due to the contribution of local fields in the nanocomposite.