

# Simultaneous two and three photon resonant enhancement of third-order NLO susceptibility in an azo-dye functionalized polymer film

ESI for communication in PCCP

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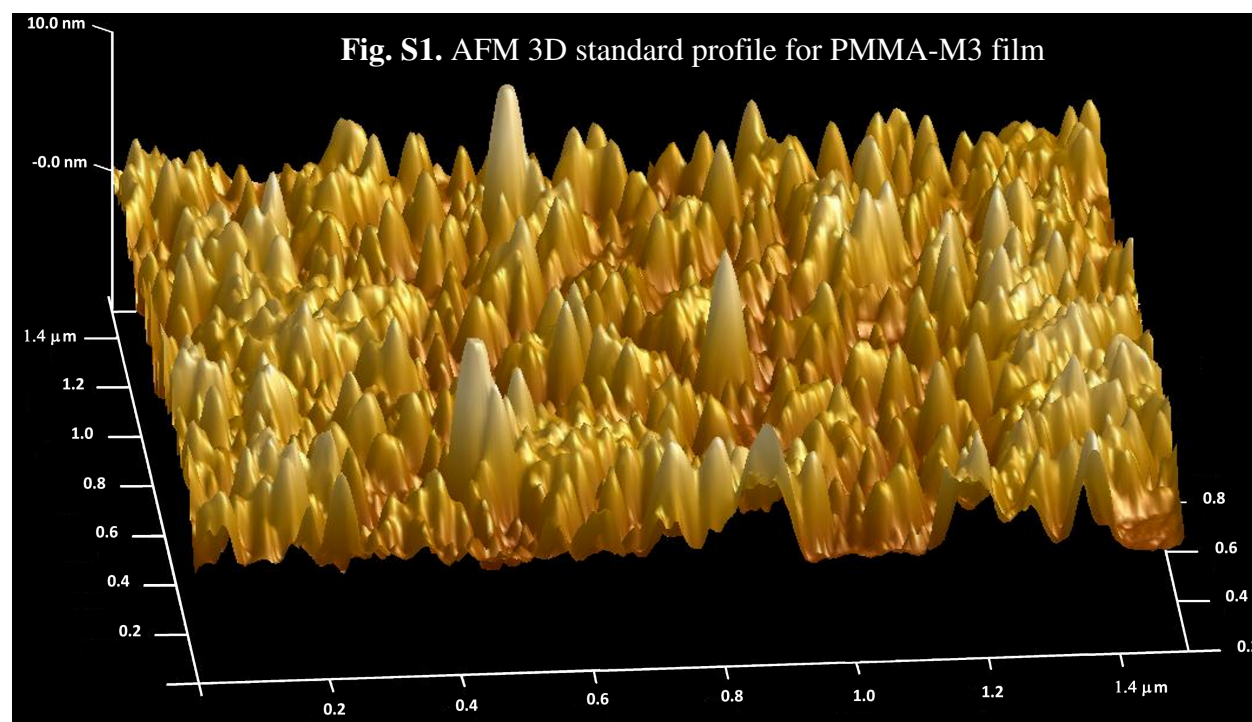
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## Atomic force microscopy (AFM) characterization of the PMMA-M3 film

**Instrument:** MultiMode 8 Atomic Force Microscope (Bruker), equipped with a Nanoscope V controller, operated in tapping mode, using a cantilever length of 225  $\mu\text{m}$ , with etched silicon tip (nominal radius 8 nm) and a resonance frequency of about 75 kHz. AFM measurements were performed at room temperature with a scan rate of 1 Hz.



### Results Roughness PMMA-M3

Image Z Range	13.8 nm
Image Surface Area	1.05 $\mu\text{m}^2$
Image Projected Surface Area	1.04 $\mu\text{m}^2$
Image Surface Area Difference	0.613 %
Image Rq	1.38 nm
Image Ra	0.944 nm
Image Rmax	13.8 nm

### Results Depth PMMA-M3

Peak to Peak Distance	1.076 nm
Minimum Peak Depth	9.088 nm
Maximum Peak Depth	10.2 nm
Depth at Histogram Maximum	10.16 nm

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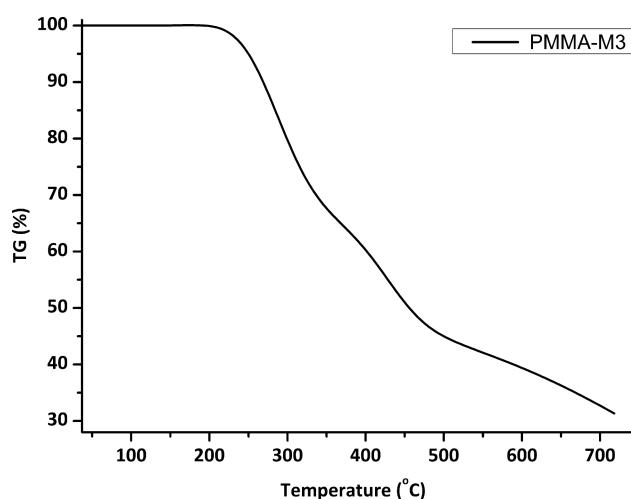
## Thermal Analysis of the PMMA-M3 polymer

### *Instrument:*

NETZSCH STA 449C Jupiter system;  
simultaneous TGA-DSC was carried out from  
ambient temperature up to 700°C at a heating  
rate of 5°C/min under helium gas flow.

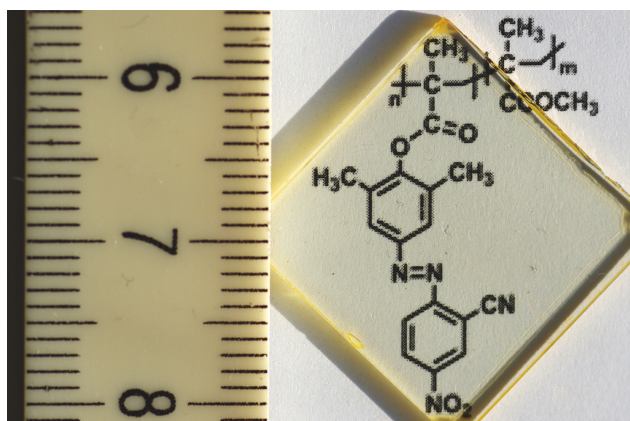
### *Characteristic values:*

Glass transition temp.	$T_g=148^\circ\text{C}$
Onset of degradation temp.	$T_{on}=246^\circ\text{C}$
Temp. at 5% weight loss	$T_{5\%}=271.5^\circ\text{C}$



**Fig. S2.** Thermogravimetry curve of PMMA-M3

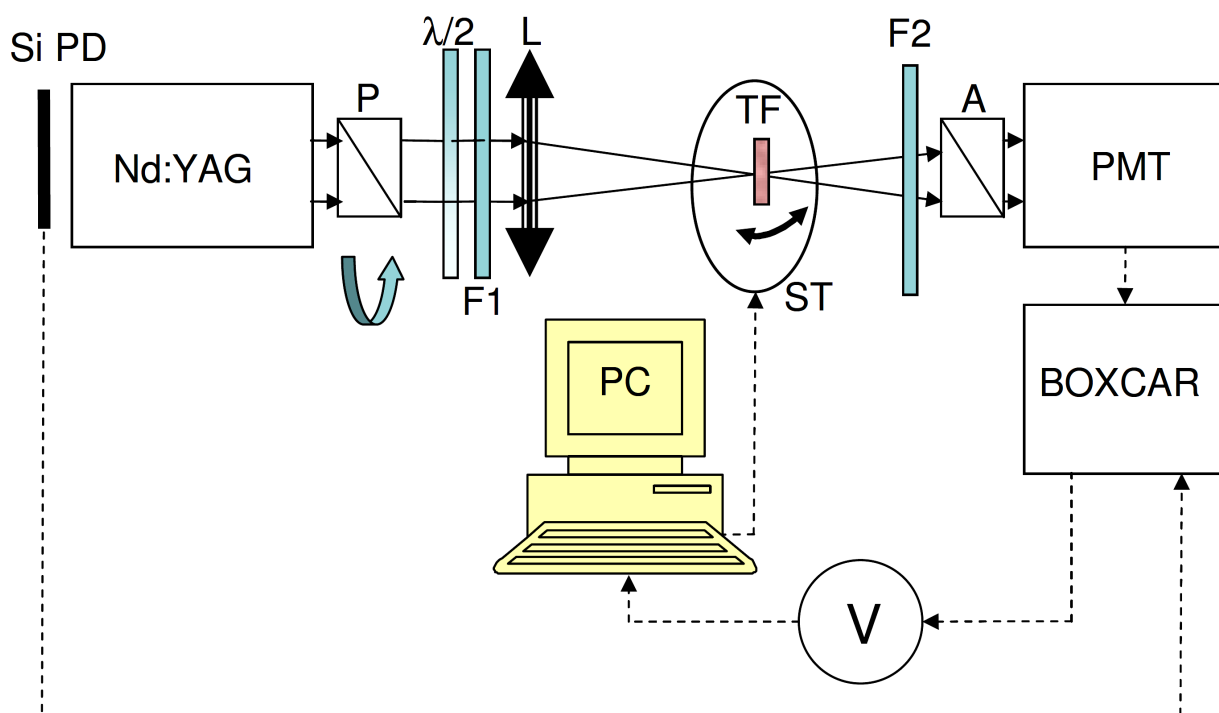
## Optical transparency of the PMMA-M3 film



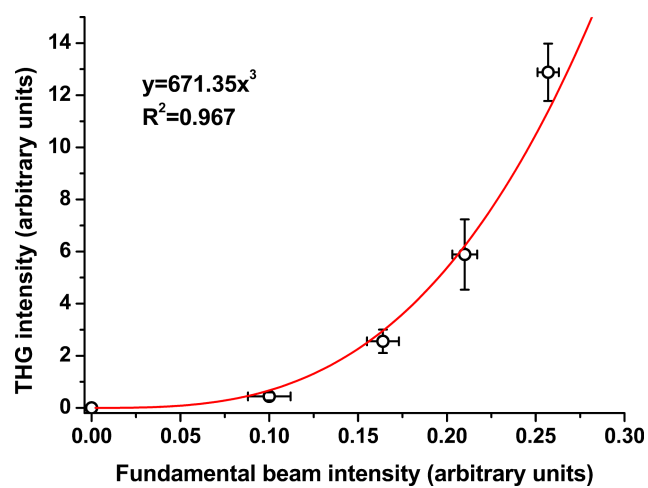
The optical photography reveals the good quality of the film, the paper roughness and printer fading can be seen with equal sharpness directly and through the film

**Fig. S3.** Optical photography of the PMMA-M3 film

### Optical bench for THG measurements

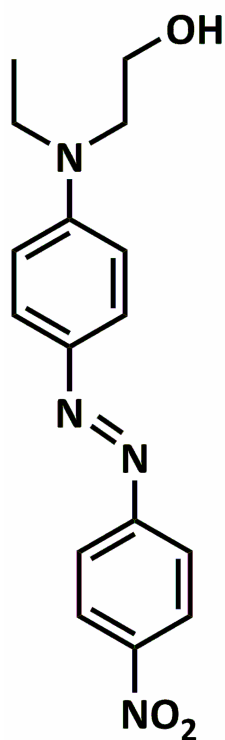


**Fig. S4.** Experimental setup used for THG measurements: *Nd:YAG* - 1064.2 nm pulsed laser; *P* - polarizer for checking polarization of fundamental beam;  $\lambda/2$  - half wave plate; *F1* - attenuation filters; *L* - converging lens; *TF* - studied film; *ST* - computer controlled rotating sample stage; *F2* - 3rd harmonic filters: 355 nm narrow pass interference filter and 1064.2 nm cutting filter; *A* - analyzer for harmonic beam; *PMT* - photomultiplier tube; *BOXCAR* - triggered amplifier; *V* - digital voltmeter; *Si PD* - fast Si photodiode for laser power control and signal triggering; *PC* - computer for system control and data recording.



**Fig. S5.** THG intensity dependence on excitation power

**Fig.S6.** Chemical formula for the reference dye - the Disperse Red 1



N-Ethyl-N-(2-hydroxyethyl)-4-(4'-nitrophenylazo) aniline