Nonlinear absorption of femtosecond laser pulses in PMMA

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Owing to its excellent physical and chemical properties, polymethylmethacrylate (PMMA) remains the basic soft-matter polymeric material platform for multiple photonic applications [1], [2]. In dielectric materials, including PMMA, multiphoton absorption is the key initial process of the energy deposition at low and moderate intensities of ultrashort laser pulses [3]. Experimental measurement of the main parameters of this process, such as multiphoton absorption coefficients and nonlinear refractive index, is of great interest. Based on the experimental measurement of the nonlinear transmission coefficient of 1-mm thick plane-parallel plate made of PMMA irradiated with tightly focused (NA = 0.25, focal length f' = 16 mm, $1/e^2$ -intensity focal spot radius $w_0 \approx 3.5 \pm 0.5 \mu m$). 1030-nm laser pulses with 250 fs duration, the nonlinear dependence of this parameter on the pulse energy was established by the I-scan method. It was found that in the range of intensities not exceeding the threshold value of $I_{\rm th} \approx 10~{\rm TW/cm^2}$ the main mechanism of laser pulses attenuation is 8-photon absorption (excited energy level $8 \times 1.2 \text{ eV} \approx 9.6 \text{ eV}$, coefficient $\beta_8 = 3 \times 10^{-5} \text{ cm}^{13}/\text{TW}^7$) implying photoionization of the PMMA chains. The nonlinear transmission measurement of weakly focused laser pulses (f' = 200 mm, $1/e^2$ intensity focal spot radius $w_0 \approx 30.5 \pm 1 \ \mu \text{m}$) based on the z-scan method made it possible to estimate the value of the nonlinear refractive index of PMMA $n_2 = 5.5 \times 10^{-4} \text{cm}^2/\text{TW}$. The obtained results were confirmed by near-IR femtosecond laser induced permanent refractive-index modifications and MMA-related changes in Raman spectra of the studied PMMA sample.

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