Investigation of laser-induced damage and related multiphoton absorption changes in lithium niobate crystals at high repetition rate femtosecond pump

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Abstract. An R-on-1 laser-induced damage-threshold (LIDT) testing method was applied to test coated and uncoated magnesium oxide-doped periodically poled lithium niobate (MgO:PPLN) samples pumped by femtosecond Yb:KGW laser pulses at kHz and MHz pulse repetition rates. LIDT values decreased by ∼1.5 and ∼38 times when increasing repetition rate from 100 kHz to 571 kHz and to 76 MHz, respectively. We also investigated nonlinear absorption changes in $LiNbO_3$ crystals at the pulse repetition range from 60 kHz to 600 kHz with trains consisting of 100 identical femtosecond pulses. Laser beam transmission in the crystal experienced the drop of ∼18% from initial pulses of train to the next 40 pulses at intensities 40 - 15% lower than LIDT due to nonlinear absorption of 220 fs duration pulses at 1.03 µm.

Keywords: laser-induced damage threshold, nonlinear absorption, femtosecond pulses, MgO:PPLN, $LiNbO_3$.

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1 Introduction

Tunable femtosecond laser radiation is usually generated in optical parametric amplifiers (OPA) and synchronously pumped optical parametric oscillators (SPOPO) utilizing nonlinear crystals, pumped by the tightly focused laser beams of high intensity. The nonlinear crystal is the weak link since its laser-induced damage threshold (LIDT) limits pump intensity, as well as the maximum output power of OPAs and SPOPOs. The LIDT values of similar optical components (for example mirrors or nonlinear crystals) depend on such parameters as pulse duration,[1–4] wavelength,[5,6] beam diameter,[7,8] pulse repetition rate,[9] measurement methodology (1 on 1, S on 1, R on 1)[10–13] and on such less documented parameters of the laser system used in LIDT measurements such as pulse contrast and pulse energy stability. This complicated dependence of the LIDT on so many parameters makes evaluation of the applicability of different nonlinear crystals in femtosecond

OPAs and SPOPOs possible only in the case when the LIDT values are obtained for the same pump laser parameters such as wavelength, pulse duration, beam diameter and pulse repetition rate. Characteristic values of the last one are 1-1000 kHz in OPAs and ≈80 MHz in SPOPOs .

The part of the absorbed ultrashort pulse energy depends on the nonlinear absorptivity which includes not only multiphoton absorption (MPA) but also avalanche ionization and absorption due to avalanche ionization-excited carriers at high intensities close to the damage threshold.[14–18] Absorptivity for femtosecond and picosecond pulses in cases of two-photon, three-photon and even four-photon absorptions in crystals and glasses can reach values exceeding 60-95%. For fluence (F) values exceeding LIDT the absorption causes transparent media's damage. Significant incubation was observed to occur in many cases already during the first ten laser pulses reducing the LIDT from the single-pulse value by 60% in the multi-pulse case.[19] Periodically poled lithium niobate (PPLN) is a commonly used nonlinear medium for the ultrafast high repetition rate OPAs and SPOPOs in the mid-IR range due to the high nonlinear coefficient and wide transmission interval. Until today, most of the research and the commercially available PPLN based SPOPOs have been pumped by femtosecond Ti:sapphire lasers,[20–22] but currently the increasing use of femtosecond laser systems based on Yb doped gain media, operating at 1.03 μm wavelength can be observed.[23,24] Despite the fact that lithium niobate ($LiNbO_3$ or LN) crystals have been widely used for the development of OPAs and SPOPOs for a long time, there is a lack of data about their LIDT values. Only few experimental LIDT values of lithium niobate exposed by femtosecond radiation were reported by other authors (Table 1). The results clearly show a large difference between the LIDT values obtained for the same material, but for different laser radiation parameters. In the Ref.,[25] authors obtained a significant difference of LIDT values for difference repetition rates while over parameters keeping the same. They got more than 8 times lower LIDT of lithium niobate at

1000 kHz comparing to the LIDT obtained at 20 kHz. An important role of the high repetition rate difference for the spread of LIDT results at femtosecond regime can be related to heat accumulation effects taking place even in a femtosecond regime, because the time interval between pulses becomes insufficient for heat to diffuse.[26,27] In metal and dielectric mirrors, the decrease in LIDT value when increasing the repetition rate from a kHz to a MHz regime was observed by B. J. Nagy, et al.[28] All tests showed that LIDT values strongly depended on the repetition rate and the $F_{1kHz}/F_{4.3MHz}$ was obtained from 2.7 to 4.8, depending on the material (F - fluence of LIDT). Decrease of LIDT value of MgO:PPLN with the increase of the pulse repetition rate was also theoretically evaluated by the use of the classical two-temperature model by Zhuolin Su, et al.[29] Heat accumulation also affects the increase of the nonlinear absorption efficiency due to decreasing of material bandgap (E_g) as the temperature increases.[30] In literature, the value of the lithium niobate energy bandgap varies from 3.7 eV to 4.7 eV.[31] Thus, the four-photon nonlinear absorption should dominate at the pump wavelength of 1.03 µm as the photon energy is ~ 1.2 eV. However, when the heat accumulation takes place, the shift of the UV edge to the longer wavelengths must be included and therefore the number of photons in MPA decreases from 4 to 3 or even 2 when temperature of MgO:PPLN increases and approaches the melting temperature. The efficiency of multiphoton absorption drops with the increase of the MPA order: according to results from Ref.[32,33] the ratio of lithium niobate nonlinear absorption coefficients are in the order of $\frac{\sigma_4}{\sigma_3} \approx 10^{-14}\ \mathrm{m}^2/\mathrm{W}$ and $\frac{\sigma_3}{\sigma_2} \approx 10^{-16}\ \mathrm{m}^2/\mathrm{W}$ (here indexes of σ mark the order of MPA). Considering these relations, the efficiency of nonlinear absorption, caused by the decrease of the number of photons taking part in MPA, must increases with crystal temperature rise, too.

In this paper, we report laser-induced damage tests on coated and uncoated MgO:PPLN crystals using different pulse repetition rates of Yb:KGW laser systems. In contrast to the previously

Table 1 Experimental parameters and results of lithium niobate LIDT values. Here: LN - lithium niobate, MgO:LN - lithium niobate doped by magnesium oxide, Nd:MgO:LN - lithium niobate doped by neodymium and magnesium oxide.

Wavelength, nm	Pulse duration, fs	Repetition rate, kHz	LIDT, $\mathrm{mJ/cm^2}$	Material	Ref.
800	300	0.067	1000	LN	[34]
800	120	1	1350	Nd:MgO:LN	[35]
800	80	1	520	LN	[10]
800	50	1	800	LN	[11]
800	150	1	1400	LN	[36]
1040	61	52000	0.8	MgO:LN	[26]
1030	330	20	803	MgO:LN	[25]
1030	330	50	487	MgO:LN	[25]
1030	330	100	175	MgO:LN	[25]
1030	330	500	116	MgO:LN	[25]
1030	330	1000	98	MgO:LN	[25]

reported experiment[37]where different radiation parameters were varied, here we focused on the variation of single parameter: repetition rate. Results show that the damage threshold at kHz and MHz repetition rates differ by tens of times. Moreover, in the section 3 we study the transmission dependence on the pump fluence and repetition rate in the uncoated lithium niobate crystal. The study elucidated that the LIDT dependence on the repetition rate is related to the MPA and its sensitivity to the incubation effects.

2 Laser-induced damage-threshold of MgO:PPLN crystals

2.1 *Materials and method*

In LIDT studies we used two 5 % doped MgO:PPLN crystal samples: one with broadband $\mathrm{Nb_2O_5/SiO_2}$ AR coatings for $\sim 1.4 - 1.8$ μm spectral range and for a pump wavelength (1.03 μm) and another

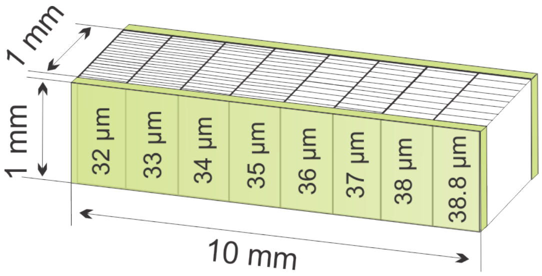

Fig 1 Principal view of MgO:PPLN geometry. The green layer marks the AR coating. Crystals have eight grating periods which was ranging from 32 µm to 38.8 µm. The geometry of uncoated crystal was identical.

without coatings (Fig.1). Coatings on crystals were deposited using the ion beam sputtering (IBS) technique. Experiments were performed using the R-on-1 test method: a single site of the sample was exposed for a fixed time equal to 300 s by multiple pulse trains with step by step increased energy fluencies from small values at the level of $\sim$0.3 $\mathrm{mJ/cm^2}$ at MHz and 17-50 $\mathrm{mJ/cm^2}$ at kHz cases up to LIDT. The experimental setup for the laser-induced surface damage threshold measurements is presented in Fig.2a). The energy of the pump pulses was changed by the use of a half-wave plate and a polarizer. LIDT was indicated by a sharp drop in the power transmission (Fig.2c)). In order to embrace the damage threshold at the femtosecond pump in kHz and MHz ranges we used two different Yb:KGW laser sources. The first source was a Yb:KGW oscillator (Flint, Light Conversion Co., Ltd) providing pulses of 89 fs duration at 76 MHz repetition rate. The second source was a Yb:KGW laser (Pharos, Light Conversion Co., Ltd) providing pulses of 298 fs duration in 10 - 571 kHz range. Both laser sources emitted radiation at a wavelength of 1.03 µm. In order to compare results obtained only at different pulse repetition rates while keeping the other parameters of both lasers equal, we expanded the duration of first source from 89 fs to 297 fs in the BK7 glass slab. In both cases, the laser beam was focused by a 75 mm focal length lens on the front surface of the crystal. The detection of the focused beam was realized with a CCD camera and the effective focused beam diameter (16 µm) was estimated according to the ISO 21254:2011

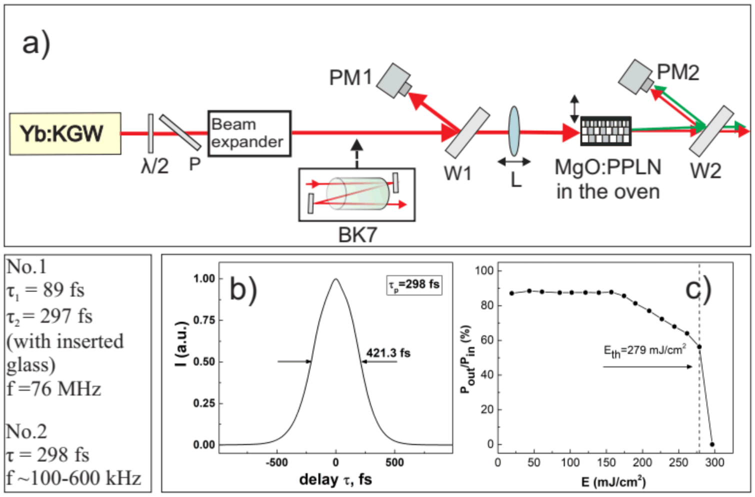

Fig 2 Experiment setup of LIDT measurements: λ/2 - half wavelength phase plate, P - polarizer, W - wedge, L - lens, PM - power meter. Here: red arrow - pump radiation (1.03 µm), green arrow - second harmonic of the pump (0.515 µm); b) Autocorrelation trace of Yb:KGW (Pharos) radiation, measured by an autocorrelator (Geco, Light Conversion Co, Ltd) and c) LIDT measurement at 571 kHZ and 298 fs (P*out* - output power, P*in* - incident power, *E* - pump fluence).

standard.

2.2 Results of LIDT experiment and discussion

Tests of laser-induced damage with the kHz laser source at 298 fs were performed at six different repetition rates in 100-571 kHz range (Table 2). At 100 kHz, 200 kHz and 300 kHz, LIDT values obtained for coated samples were roughly similar, showing that incubation effects did not play an important role. However, further increase of the repetition rate led to a decrease in LIDT values. A particularly significant difference could be seen in comparing the results obtained at kHz and MHz repetition rate regimes. LIDT values decreased by ∼1.5 and ∼ 38 times increasing repetition rate from 100 kHz to 571 kHz and to 76 MHz, respectively.

The damage morphology and LIDT values for MgO:PPLN with AR coatings obtained at three kHz and one MHz repetition rates are shown in Fig.3. The LIDT for fixed kHz and MHz repetition rates were estimated at least three times and variation of LIDT values was less than 5%. In all cases

Table 2 Experimental LIDT values for MgO:PPLN crystal at different repetition rates

f, kHz	LIDT of coated MgO:PPLN F_{th}, $\mathrm{mJ/cm^2}$	LIDT of uncoated MgO:PPLN F_{th}, $\mathrm{mJ/cm^2}$
100	436.6	575.2
200	438.6	517.7
300	438.8	485.3
400	407.7	431.4
571	278.6	283.1
76000	11.8	$>$12.4 *

* Damage to the uncoated crystal was not observed at all because of the irradiance being too low (the maximum energy fluence was equal to 12.4 $\mathrm{mJ/cm^2}$ for 297 fs long pulses due to the 37% loss in the expansion glass).

the formed pit on the MgO:PPLN surface was 1.9 - 3.4 times smaller than the effective focused beam diameter (d_{eff}=16 µm), and it corresponds to the reduction of the affected zone by $\sqrt{4} = 2$ times for four-photon processes. The small rim surrounding the pit formed by laser ablation can also be noticed in all images of the damage morphology. Its formation the most probably was caused by melting and re-solidification of melted material after exposure.[34,38] Visible dots around the damage area in Fig.3(a-d) are probably splashed out material particles during the gasification or phase explosion.[39,40]

The uncoated crystal was damaged at a higher energy value than the crystal with AR coatings, as can be seen from the comparison of the experimental results in Table2. According to the Ref.,[41] the extinction coefficient of $\mathrm{Nb_2O_5}$ thin films was zero when the light wavelength was in the range of 370-345 nm depending on the film deposition conditions. It means that for high intensity femtosecond pump pulses at 1.03 µm propagating in such optical thin-film, not four, but three-photon absorption must appear. This leads the more efficient nonlinear absorption at lower fluencies in the case of coated samples. In the case of uncoated crystal the losses of laser radiation on the entrance surface, which is of the order of 13%, also must be included.

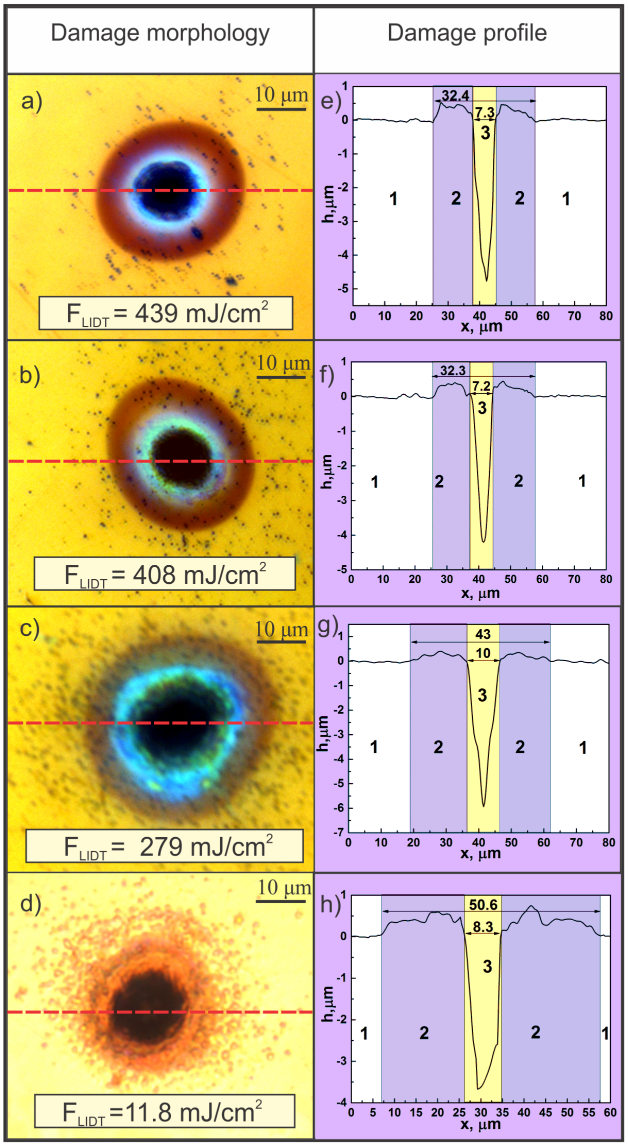

Fig 3 Images of laser-induced damage sites on MgO:PPLN with AR coatings for 298 fs pulses at 200 kHz (a, e), 400 kHz (b,f) 571 kHz (c, g), and 76 MHz (d, h) repetition rates made by optical microscopy (a-d) and profilometer (e-h). Numbered areas (e-f) show non-affected areas (1), surface swelling zones (2) and formed pits (3).

3 The change of nonlinear absorption in the lithium niobate

3.1 Experimental setup

The simple method for nonlinear absoption estimation is a measurement of pulses transmission with increasing their intensity.[42,43] In the ref.,[43] the strong influence of two-photons absorption to the propagation of high-power femtosecond pulses in lithium niobate at 388 nm was observed.

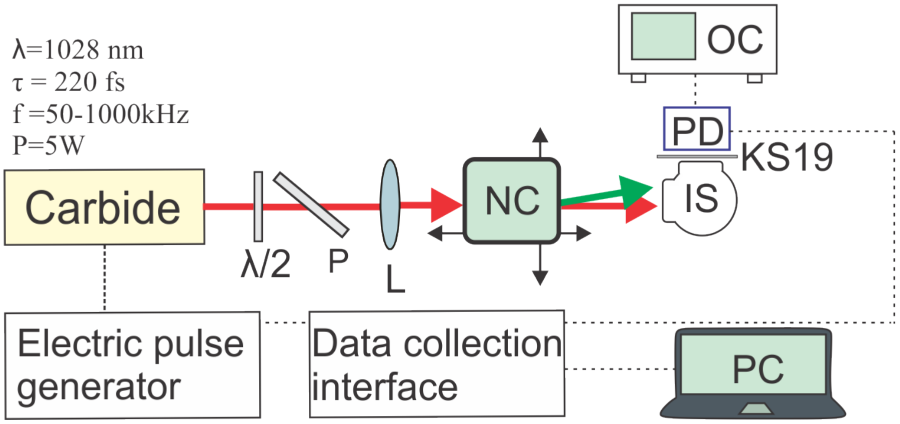

Fig 4 Experimental setup for nonlinear absorption measurements: λ/2 - half-wavelength phase plate, P - polarizer, L - lens (f=50 mm), NC-nonlinear crystal, IS - integrate sphere, KS19 - optical filter, PD - photodiode, OC - oscilloscope. Here: red arrow - pump radiation, green arrow - second harmonic of the pump.

They obtained the decrease of transmission more than 60% when pump intensity reached 200 $\mathrm{GW/cm^2}$. We investigated the nonlinear absorption by mentioned transmission measurement of femtosecond laser pulses at various repetition rates and energy fluencies (Fig.4). Laser source was Carbide laser (Light Conversion, Ltd), emitting 220 fs duration pulses at 1028 nm wavelength with posibility to change the repetition rate from 60 kHz to 1000 kHz. The pulse duration, their number and repetition rate were controlled by the laser software. Nonlinear crystal was mounted on a translation stage, allowing precise movement of the crystal longitudinally and transversally with respect to laser radiation which was focused with a lens of 50 mm focal length at the crystal surface. The radiation passed though the nonlinear crystal was collected by integrated sphere. The KS19 filter was used to block the second harmonic generation, which occurs in nonlinear crystal. Multifunctional data collection device (National Instruments), electric pulse generator, laser, photodiode, oscilloscope and computer were combined together to properly match, synchronize and collect experiment data, which was finally processed by the software. Tests were performed by irradiating crystal surface point with femtosecond pulse trains, consisted of 100 pulses. After first pass of such train of pulses through the crystal, the process was repeated (Fig.5). If no change in transmission was recorded (Fig.5(a,b)), the whole process was repeated with higher power. If lower transmittance of pulses was observed during the second train, the laser damage of the crystal was indicated (Fig.5(c,d)). In this case, the position of the crystal was changed for folowing measurements. As nonlinear materials we used the uncoated 4 mm thick $LiNbO_3$ crystal. The theoretically calculated transmission of the LN for 1028 nm wavelength radiation was 75% (the real transmission was by 8% lower due to reflections, surface scatering and absorption in the crystal). As mentioned in the Introduction, the energy bandgap of LN is in the range of 3.7 eV - 4.7 eV means that 4 photons of 1028 nm radiation are required at initial stage of nonlinear absorption.

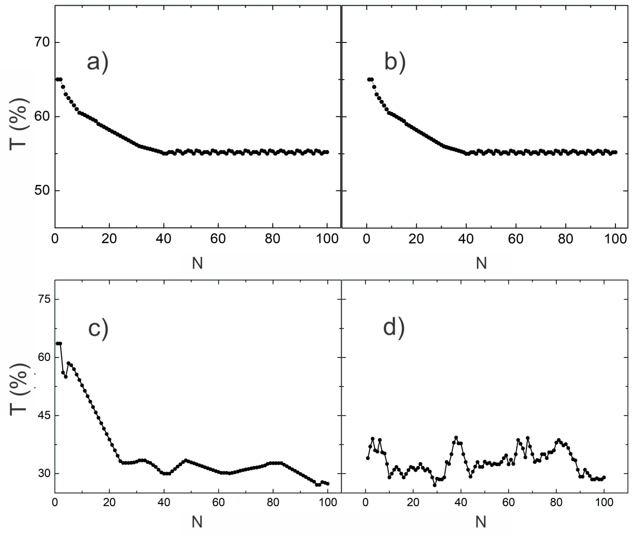

Fig 5 Transmission (T) of separate femtosecond pulses in lithium niobate in first and second trains of pulses at 200 kHz: 0.016 $\mathrm{J/cm^2}$ (a,b) and 0.29 $\mathrm{J/cm^2}$ (c,d). Here: N-number of the pulse.

3.2 Results and discussion

Firstly, measurements were performed using different energy fluencies at fixed repetition rate Fig.6 and the strong dependency of pulse train transmission on energy fluence was obtained. At low energy fluences, no change in crystal transmission was observed (black curves in Fig. 6). At 200 kHz repetition rate, the transmission of pulse train at energy density 0.016 $\mathrm{J/cm^2}$ fell from

65% to 55% in 40 pulses and from 64% to 33% in 24 pulses at 0.29 $\mathrm{J/cm^2}$. The decrease in the recorded transmission with increasing pulse energy density indicated the presence of multiphoton absorption. The faster reduction of transmission at larger fluencies was caused by higher efficiency of MPA. The last grew due to the higher flow of photons and the decreasing order of multiphoton ionization caused by the heat accumulation.

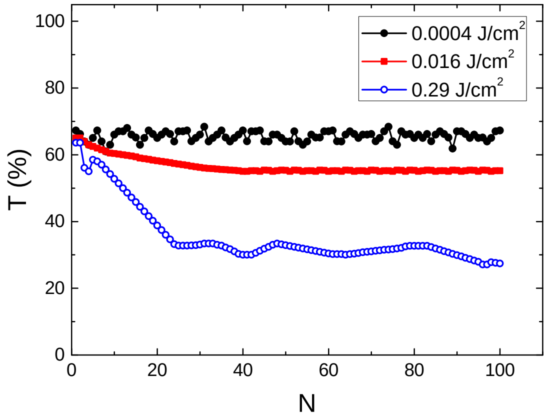

Fig 6 Lithium niobate transmission measurements results at certain energy densities at *f*=200 kHz.

Secondly, we performed investigation of crystal transmission at four different repetition rates of pulses: 60 kHz, 200 kHz, 400 kHz and 600 kHz. Summarized results are depicted in Fig.7. The strong laser beam transmission dependence from pulse repetition rate were observed. From the comparison of both curves in the Fig.7 it can be noticed that nonlinear absorption makes strong influence at 15-40% (depending on the pulse repetition rate) lower energy density than observed LIDT values, making conditions for the significant (18%) drop of transmission in first 40 pulses of the femtosecond pulses train.

4 Conclusions

Laser-induced damage threshold measurements of the MgO:PPLN crystal with and without coatings were performed at kHz and MHz repetition rate regimes of femtosecond Yb:KGW laser

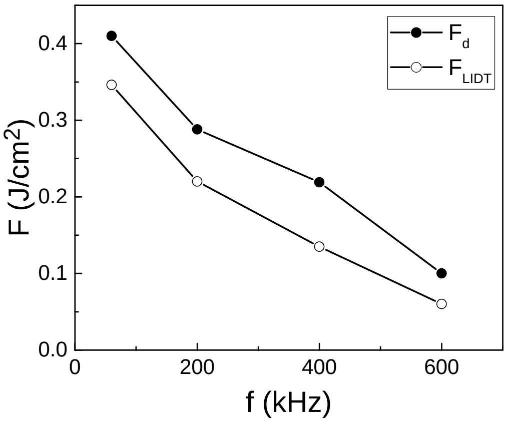

Fig 7 Dependencies of energy fluence which caused the ∼18% drop of of transmission from initial value in next 40 pulses (F_d) and LIDT fluence (F_{LIDT}) on pulse repetition rate (f).

sources. LID thresholds of kHz repetition rate differ from those of MHz rate by more than factor of 20. Due to these results, scaling laws for LIDT estimation is an unreliable tool and the difference between repetition rates should be taken into account. At 100-300 kHz repetition rate regime, no decrease of LIDT value with increasing repetition rate for the coated MgO:PPLN was observed and the LIDT value was ∼439 $\mathrm{mJ/cm^2}$. We also achieved a good congruence by comparing our observed decline of LIDT values in kHz pulse repetition rate regime with obtained by F. Bach et al.[25] They obtained $F_{1\mathrm{kHz}}/F_{500\mathrm{kHz}} = 1.5$ while we got $F_{1\mathrm{kHz}}/F_{571\mathrm{kHz}} = 2$. Additionally we performed the observation of the nonlinear absorption change for lithium niobate under irradiation with trains of high-repetition rate femtosecond pulses at several pulse repetition rates in the range from 60 kHz to 600 kHz. We indicated the increase of the MPA efficiency as the pulse repetition rate or energy density increases. The significant MPA efficiency at 15-40% lower fluencies than crystal LIDT is the cause of fast transmission drop in the nonlinear crystal and such transmission observation could help roughly predict material's LIDT without damaging it.

Acknowledgments

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