Quantum Electronics **43** (8) 731 – 734 (2013)

INTERACTION OF LASER RADIATION WITH MATTER

PACS numbers: 42.70.Ce; 79.20.Ds
DOI: 10.1070/QE2013v043n08ABEH015142

Effect of laser pulse repetition frequency on the optical breakdown threshold of quartz glass

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Abstract. **The thresholds of optical breakdown in the volume of quartz glass were measured in relation to the number of pulses under irradiation by ultrashort laser pulses with different pulse repetition frequencies (1 – 400 kHz). Increasing this frequency from 10 to 400 kHz was found to substantially lower the breakdown threshold for 500-fs long pulses (at a wavelength of 1030 nm) and to lower to a smaller degree for 5-ps long pulses (515 nm). A strong frequency dependence of the breakdown threshold is observed under the same conditions as a manifold decrease of the breakdown threshold with increase in the number of pulses in a pulse train. The dependence of the optical breakdown on the number of pulses is attributable to the accumulation of point defects under multiple subthreshold irradiation, which affects the mechanism of collisional ionisation. In this case, the frequency dependence of the breakdown threshold of quartz glass is determined by the engagement of short-lived defects in the ionisation mechanism.**

Keywords: *ultrashort pulses, optical breakdown, quartz glass.*

1. Introduction

The increasingly active technological use of ultrashort-pulse laser systems, including the microprocessing of transparent materials [1], generates the need for improving their productivity by raising the pulse repetition frequency f along with an increase in average power. The major part of research on glass micromodification by femtosecond pulses is executed for $f > 100$ kHz [2] and in some cases for $f = 25$ MHz [3–5]. Under these conditions the heat released in the focal volume has no time to completely disperse into the ambient space during the laser interpulse time. The accumulation of heat results in the gradual heating, melting, and eventually material modification outside of the region of laser radiation absorption [4]. Structural difference was discovered in quartz modified by exposure to pulses with $f = 1$ MHz and 1 kHz; supposedly this is also attributable to a stronger and longer heating of the material irradiated by pulses with a higher pulse repetition frequency [6].

Can a high repetition frequency affect the very mechanism of radiation absorption in quartz and, as a consequence, change the optical breakdown threshold? This possibility was experimentally borne out in Ref. [7] by comparing the optical breakdown thresholds of quartz for a single pulse of length $\tau = 25$ fs and a pair of the same pulses with a varied delay between them. For a minimal delay (70 fs), the double-pulse breakdown threshold was 2.3 times lower in comparison with a single pulse, but increased (~1.8-fold) rapidly again when the delay was lengthened to 200 fs to finally return to the initial value when the delay increased to 2.5 ns. The lowering of the breakdown threshold for double pulses may be explained by the fact that the residual photoexcitation of the material after the first pulse favours the development of an electron avalanche during the course of the second pulse. The greatest threshold drop is observed when the remaining free electrons, whose lifetime in quartz amounts to about 200 fs, serve as a seed for the avalanche development. A weaker effect with increasing the interpulse delay to several nanoseconds is supposedly due to self-trapped excitons [8], which produce defect levels with a lowered ionisation energy in the forbidden band. It is pertinent to note that the longest interpulse delay (2.5 ns) whereby the lowering of the breakdown threshold was observed is equivalent to a very high pulse repetition frequency (400 MHz), which is unattainable for modern high-power laser systems.

The experimental data outlined in our work demonstrate that the relative lowering of the breakdown threshold in quartz glass under the action of ultrashort pulses may also be observed at substantially lower repetition frequencies – 400 kHz. The key factors for the realisation of this effect are a large number of pulses in the train and the parameters of laser radiation (the pulse duration and wavelength).

2. Experiment

Two laser systems were employed in our experiments. A VaryDisk50 (Dausinger + Giesen) multimode laser system generated pulses at a wavelength 1030 nm with a duration $\tau = 500$ fs and a maximum frequency $f = 400$ kHz. The highest average power was equal to 45 W. A TruMicro 5050 (TRUMPF) laser generated pulses with $\tau = 5$ ps at wavelengths 1030 and 515 nm with the same maximum repetition frequency. The highest average power was equal to 42 W at the fundamental frequency and to 10 W at the second harmonic frequency. Both laser systems were equipped with a built-in optical pulse selector, which permitted lowering the pulse repetition frequency to 1 – 10 kHz and also forming pulse trains of prescribed length. A lens ($F = 100$ mm) integrated with a two-coordinate angular scanner focused the laser radiation to a distance of about 1 mm from the front surface inside a KU1 quartz glass brick (10 × 10 × 20 mm)

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Received 1 February 2013; revision received 6 March 2013
Kvantovaya Elektronika **43** (8) 731 – 734 (2013)
Translated by E.N. Ragozin

polished on all sides. The Gaussian spot diameter (20–30 μm) was determined from the size of imprints on the surface of a SiC ceramic for different pulse energies. The sample was irradiated by pulse trains of different length, which were each time directed to a new place in order to determine their minimal number that caused damage in the glass volume for a given pulse repetition frequency and a given pulse energy.

The main criterion of the optical breakdown in quartz glass was the emergence of plasma glow in the focal region, which was visually detected in the observation of the irradiation zone with a microscope. To prevent the scattered laser radiation from entering the microscope, use was made of a narrow-band dielectric mirror (λ = 515 nm). A similar approach was employed in Refs [7, 9], except that the glow was recorded with a photodetector. For τ = 5 ps this criterion correlated strictly with the detection of microscopic defects in the sample volume with the help of optical microscopy. For τ = 500 fs, the damage visible with the microscope emerged for a substantially greater number of pulses than that required for the emergence of the glow. This difference is supposedly attributable to the difference in nature of the emerging material damage: a microcrack and/or a bubble for τ = 5 ps and a small change of the refractive index for τ = 500 fs [2].

3. Results of measurements and their discussion

Figures 1–3 show the experimental dependences of the breakdown threshold of quartz glass on the number of pulses for the three laser parameter combinations tested (wavelength + pulse duration) and different repetition frequencies. The breakdown thresholds are expressed in terms of the maximum energy density in the focal plane, which were calculated proceeding from the size of the focal waist in the air, i.e. neglecting the laser beam transformation in glass owing to the effect of self-focusing, although the peak power at the breakdown threshold in our experiments (5–200 MW for τ = 500 fs and 3–20 MW for τ = 5 ps) far exceeded the power of self-focusing in quartz glass equal to 5.2 MW at a wavelength of 1.06 μm [10]. This simplified approach is obviously inapplicable in the determination of the absolute value of the breakdown threshold, but seems to be quite acceptable when following the relative lowering of the breakdown threshold with increase in pulse repetition frequency. We note that our resultant values are comparable with the data of other authors [9, 11, 12], which diverge by factors of two–three for the pulse duration range of interest to us.

For τ =500 fs (λ = 1030 nm) (Fig. 1) we observed a manifold lowering of the breakdown threshold of optical glass with increase in the number of pulses (a so-called cumulative effect) for all pulse repetition frequencies tested in our work. The breakdown thresholds for frequencies of 1 and 10 kHz lie in the same curve corresponding to a 'low-frequency' cumulative effect. However, increasing f to 400 kHz is attended with an appreciable lowering of breakdown thresholds for trains of more than three-four pulses. By contrast, for τ = 5 ps and the same wavelength (Fig. 2), not only did we see no frequency effect on the breakdown threshold whatsoever, but the 'low-frequency' cumulative effect turned out to be quite moderate in magnitude. In this case, the main lowering of the breakdown threshold (by only 20 %) is observed even with the second pulse; its further lowering in the multiple irradiation amounts to only 5 % of the initial level. The data obtained for the second harmonic and a pulse duration of 5 ps (Fig. 3) occupy an intermediate position between these two extreme cases. A small threshold lowering for a high repetition frequency is observed only for very long pulse trains ($N > 100$), and the difference between the breakdown thresholds under single and multiple irradiation is greater than in the case of picosecond IR pulses, but smaller than for femtosecond

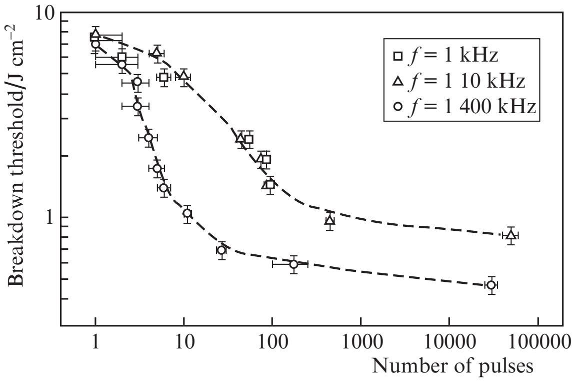

Figure 1. Dependences of the breakdown threshold of quartz glass on the number of pulses with different pulse repetition frequencies for τ = 500 fs (λ = 1030 nm).

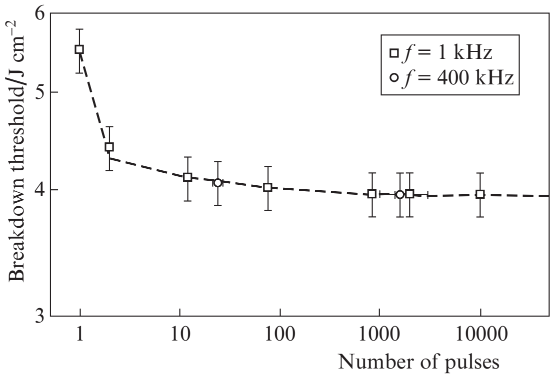

Figure 2. Dependences of the breakdown threshold of quartz glass on the number of pulses with different pulse repetition frequencies for τ = 5 ps (λ = 1030 nm).

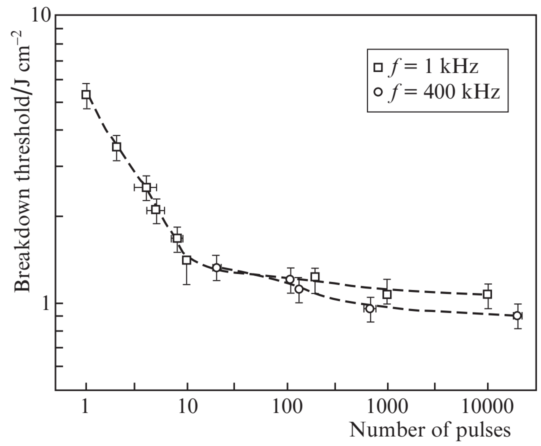

Figure 3. Dependences of the breakdown threshold of quartz glass on the number of pulses with different pulse repetition frequencies for τ = 5 ps (λ = 515 nm).

pulses. From the data obtained one may draw the following conclusion: the stronger the manifestation of the 'low-frequency' cumulative effect, i.e. the greater the lowering of the breakdown threshold under multiple irradiation with $f = 1$ kHz, the stronger the dependence of the breakdown threshold on the repetition frequency.

It is well known that one of the results of the multistage relaxation of photoexcitation in quartz is the production of point structural defects, some of which have virtually infinite lifetimes and are capable of accumulating under multiple irradiation [13]. There is an opinion that it precisely these long-lived defects with a lowered ionisation energy which are responsible for the lowering of the breakdown threshold under multipulse irradiation by ultrashort pulses [9, 11, 14, 15]. The hypothesised mechanism responsible for the lowering of the breakdown threshold consists in the generation of additional free electrons in the multiphoton ionisation with participation of defect levels, which fosters the enhanced development of collisional avalanche ionisation.

Collisional ionisation is believed to be the key mechanism for the production of absorbing plasma in the irradiation of quartz and other wide-gap dielectrics by pico- and nanosecond pulses [12, 16]. With a decrease in pulse duration to several picoseconds and below, i.e. in going over to the femtosecond range, the efficiency of an electron avalanche lowers appreciably because of its limited development time, while the contribution of nonlinear photoionisation (tunnel or multiphoton ionisation) increases. The electron avalanche nevertheless remains an important component of ionisation mechanism, without which the critical plasma density cannot be attained. According to the calculations of different authors [12, 16], the tunnel ionisation from the valence band in quartz can independently provide the production of a strongly absorbing plasma only for very short pulses (below 10 fs), so that the contribution of collisional ionisation may be neglected.

The published data of numerical simulations of electron avalanche development in quartz suggest that the breakdown (damage) threshold weakly depends on the initial, albeit rather low, free-electron density. According to Ref. [17], the peak intensity required to heat quartz to a temperature of 1500 K and higher by pulses with $\tau = 100$ ps ($\lambda = 1064$ nm) decreases from $\sim$240 to $\sim$130 GW cm^{-2} with increasing initial electron density from 10^{15} to 1.5×10^{18} cm^{-3}. In Ref. [12] the quartz breakdown threshold was determined proceeding from the criterion that the plasma density reached the critical density under irradiation by laser pulses of different duration (20 fs – 100 ps) at a wavelength of 800 nm. These simulations included the mechanisms of collisional ionisation and photoionisation – both separately and simultaneously. According to the data presented in Ref. [12], an increase in the initial electron density from 10^8 to 10^{15} cm^{-3} entails a (1.5–2)-fold lowering of the breakdown threshold for 0.2–100 ps long pulses. For pulses shorter than 70 fs, the breakdown threshold is independent of the initial electron density because of the massive contribution from photoionisation. According to some estimates [18, 19], in quartz the efficiency of point defect production amounts to about 10^{-3} of the number of generated electron-hole pairs. Proceeding from this figure, the maximum defect density produced under multiple subthreshold irradiation may amount to $\rho_{\rm crit}10^{-3} = 10^{18}$ cm^{-3}, where $\rho_{\rm crit} \approx 10^{21}$ cm^{-3} is the critical plasma density. In view of the simulation data of Refs [18, 19] outlined above, this rise of the initial electron density (from 10^8 to 10^{18} cm^{-3}) must result in a three- or four-fold lowering of the breakdown threshold, which is in reasonable agreement with our experimental data.

The breakdown threshold sensitivity to the initial free-electron density throughout the pulse duration range under investigation is an important ground for attributing the observable lowering of the breakdown threshold to the accumulation of easily ionisable point defects in quartz. Another important factor is, in our opinion, the rate of point defect production in the multiple subthreshold irradiation. According to the data of numerical simulations of avalanche ionisation in quartz exposed to picosecond IR pulses [17], in the purely avalanche ionisation with a low initial free-electron density the peak plasma density exhibits a very rapid (abrupt) growth as the laser intensity approaches the breakdown threshold. Even a small intensity lowering in comparison with the breakdown threshold results in a sharp decrease in the peak plasma density, making hardly possible the accumulation of point defects under subthreshold irradiation, because their production rate (per pulse) is proportional to the peak plasma density, as discussed in the foregoing. However, the intensity dependence of the peak plasma density becomes gently sloping on radiation frequency doubling [17] owing to a rise of multiphoton ionisation in efficiency and a substantial growth of the number of seed electrons at the initial stage of avalanche ionisation. The more gently sloping dependence permits point defects to be faster produced and accumulated even for a significant lowering of the intensity below the breakdown threshold. A similar effect would also be expected on pulse shortening from 5 ps to 500 fs, which is also accompanied with an enhancement of nonlinear photoionisation and therefore makes flatter the dependence of the peak plasma density on the laser intensity. Therefore, we hypothesise that it is precisely the dependence of the point defect production rate on the pulse parameters which underlies the experimentally discovered features of the 'low-frequency' cumulative effect.

It is also well known that relatively short-lived point defects with lifetimes of the order of several microseconds are also generated in the course of electron–hole plasma relaxation [20]. For a sufficiently high pulse repetition frequency (hundreds of kiloherts), these defects may also be involved in the ionisation mechanism by inducing an additional lowering of the breakdown threshold. The degree of this lowering depends on the density of short-lived defects at the instant of arrival of the next pulse, which must increase with pulse repetition frequency and the number of pulses in the train. The frequency-dependent lowering of the breakdown threshold must manifest itself under the same conditions as the 'low-frequency' cumulative effect, i.e. when the ionisation mechanism is sensitive to the presence of point defects and the dependence of the peak plasma density on the laser intensity is flat.

Can the frequency dependence of the breakdown threshold be to a certain extent related to material heating? Proceeding from the thermal diffusivity of quartz glass $\chi = 0.01$ cm^2 s^{-1}, the characteristic time of heat diffusion from a domain of radius $R = 10$ μm is $\tau_{\rm d} = R^2/\chi = 0.1$ ms. It is evident that the prerequisites for heat accumulation in our experiments emerge even for $f = 10$ kHz. Unfortunately, our attempts to find reliable data on the absorption of laser energy in quartz under subthreshold irradiation did not meet with success, and so we cannot estimate the material heating in the focal region by either a single pulse, or a pulse train with a high repetition frequency. The following experimental observations testify to the possibility in principle that the material temperature may affect the breakdown threshold. Direct measurements of the damage threshold of a quartz film induced by 100-fs and 2-ps long pulses ($\lambda = 800$ nm) (the film was externally heated) [21] revealed that the threshold variation in the 300–500 K range did not exceed the uncertainty of experiment of $\sim 10\%$. However, on raising the temperature to 1300–1750 K, Bude et al. [22] observed, in the irradiation by 7-ns pulses ($\lambda = 355$ nm), a lowering of the surface damage threshold by 30%–50%. These temperatures are close to the melting point of quartz glass and may supposedly be reached only in the immediate vicinity of the optical breakdown threshold. Therefore, at present we cannot completely rule out the possibility that material heating affects the breakdown threshold under laser irradiation with a high pulse repetition frequency; however, this factor alone can hardly be responsible for the threshold lowering observed in our experiment.

4. Conclusions

We observed a significant lowering of the multipulse breakdown of quartz glass for pulses with $\tau = 500$ fs ($\lambda = 1030$ nm) with increasing their repetition frequency from 10 to 400 kHz. The frequency effect, at least in the 1–400 kHz range, becomes weaker in going over to longer (5 ps) half wavelength ($\lambda = 515$ nm) pulses and vanishes completely for pulses with $\tau = $ 5 ps and $\lambda = 1030$ nm. The cumulative effect in quartz glass – the lowering of the threshold for long pulse trains – simultaneously becomes weaker. The dependence of the breakdown threshold on the number of pulses may be qualitatively explained by the accumulation of point defects under multiple subthreshold irradiation, which affects the mechanism of collisional ionisation. In this case, the frequency dependence of the breakdown threshold of quartz glass is due to the involvement of short-lived defects in the ionisation mechanism. Furthermore, the effect of material heating on the breakdown threshold under high-frequency laser irradiation cannot be ruled out.

Acknowledgements. The authors express their appreciation to V.V. Kononenko for granting samples for the investigations and his interested discussion of results, and to M.A. Larionov for his invaluable assistance in experiments. This work was supported by the Russian Foundation for Basic Research (Grant No. 11-02-12242).

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