Investigation of electrooptical breakdown threshold in gas mixtures of complex chemical composition

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Abstract. The results of combined electrooptical breakdown (DC $E$~0–13.2 kV/cm + laser $\lambda$~213, 266, 355, 532, 1064 nm, $t_{0.5}$~18 ns, $I_0$~$10^9$–$10^{11}$ W/cm$^2$) of gases (He, Ne, Ar, Kr) and their binary and triple mixtures at subatmospheric pressure ($10^1$–$10^5$ Pa) experimental investigation are presented for the first time. Thresholds of optical, electrical and combined electrooptical breakdown for those were experimentally determined. The possibility of breakdown threshold components decrease at simultaneous optical and electrical impact on the gas, and the possibility of lowering the thresholds of the combined breakdown of doped gases have been investigated.

1. Introduction

Ignition of pulsed breakdown of gases has been investigated for a long time, and spark discharge in DC or low frequency AC electric field is perhaps the most studied and frequently used. Despite this, studies of electrical discharges are ongoing and are aimed primarily on finding ways to improve energy efficiency (e.g. for lighting and lightning protection), also due to combined impact. So, to date, various combinations of DC, AC, RF [1] and microwave electric fields and light fields [2], [3] have been investigated, including combinations with various sources of seed electrons (thermal [4], emission [5], formed by chemical reactions or the preionization gas gap [6]). And in most cases the vector fields for the combined effect were set to collinear (radiation going along the electric field).

Interest in the study of optical and electrooptical breakdown is associated with the ignition of breakdown in hydrocarbon fuels. The advantages of laser ignition of fuel mixtures are that the concentration of harmful compounds (NO$_x$, CO, SN) in the exhaust is proportional to the temperature in the combustion chamber, which can be reduced when lean mixtures are burnt. The ignition of such mixtures by an electric spark requires a substantial increase of the ignition system power, which reduces spark plug’s lifetime. The specific power of the engine is proportional to the degree of compression in the combustion chamber, pressure increase within reasonable limits leads to an increase in the required voltage at the spark ignition, while the laser ignition threshold is reduced. Laser ignition can be effectively used at low pressures in the combustion chamber also, at pulsed periodic ignition of laser plasma (optical plasmatron) in the engines with continuous fuel supply.

The aim of this work is to study the possibility of breakdown thresholds components mutual reduction at simultaneous optical and electrical impact on the gas, and the possibility of breakdown threshold decrease at doping by other gas. Combined impacts could be considered as those with one of...
components domination or with their equal effect [7]. At electrical and optical breakdown of gases minimum thresholds occur in the regions of \(~10^{2}-10^{3}\) Pa and \(~10^{6}-10^{7}\) Pa, respectively [8]. In addition to the optimum for each mode of breakdown, there are also regions where combined impact is the most energy efficient. Analysis of mechanisms of electric and optical breakdown suggests the existence of synergistic effects, when formation of seed electrons and their acceleration is combined [9].

2. Experimental setup
ISO 40-KF vacuum cross had been pumped down to \(10^{-2}\) Pa and than filled with gas mixtures (>99.99 %, Linde gas) with pressure varied from \(10^{5}\) to \(10^{6}\) Pa. (Fig. 1). Radiation of five harmonics (\(\lambda\sim1064\) nm, 532 nm, 355 nm, 266 nm and 213 nm) of nanosecond (\(\tau\sim11-18\) ns) Nd:YAG lasers (LS-2147, Lotis TII and LQ929, Solar LS) with maximum pulse energy \(E_{1064,max}\sim0.91\) J, \(E_{532,max}\sim0.32\) J, \(E_{355,max}\sim0.074\) J, \(E_{266,max}\sim0.046\) J and \(E_{213,max}\sim0.009\) J have been used in the experiments, exposure was carried out in a repetitively pulsed mode (10 Hz).

During the experiments radiation was focused by a quartz lens \((F\sim150\) mm) in a spot diameter of \(~0.12\) mm in the gap \((d\sim3–10\) mm) between two movable copper electrodes, one of which had a positive potential up to \(U\sim5\) kV (second electrode was grounded), giving electric field strength up to \(E\sim13.2\) kV/cm (current limited \(5.25\) mA) and radiation limiting power densities \(I_{1064}\sim1.2\cdot10^{11}\) W/cm\(^2\), \(I_{532}\sim4.3\cdot10^{10}\) W/cm\(^2\), \(I_{355}\sim9.2\cdot10^{9}\) W/cm\(^2\), \(I_{266}\sim6.1\cdot10^{9}\) W/cm\(^2\) and \(I_{213}\sim1.2\cdot10^{9}\) W/cm\(^2\).

Plasma broadband radiation and electric grid shortening were considered as breakdown evidence.

For breakdown dynamics experiments Nomarski interferometer with 20 ns exposure time was used.

3. Results
Optical and electrical thresholds at different pressures were evaluated initially, the results of these reference experiments correspond to published ones [10], and measured the threshold of the combined breakdown: at fixed voltage radiation intensity was increased until spark observation. Threshold value was recorded for breakdown probability of 50% [11].

Electric field influence on combined breakdown threshold becomes more obvious with pressure and wavelength decrease (Fig. 2). This trend might be explained by multi-photon ionization probability increase at short wavelength irradiation, seeding electron recombination decrease during their number doubling period (ca. 20 ps) and electron energy increase due to acceleration by electric field in between of collisions.
Figure 2. Dependence of electrooptical threshold breakdown Ar at different pressures on the electric field, laser intensity and wavelength $\lambda = 532$ nm (1 – $p \approx 1 \times 10^5$ Pa, 2 – $p \approx 4.9 \times 10^4$ Pa, 3 – $p \approx 2.3 \times 10^3$ Pa, 4 – $p \approx 1.1 \times 10^4$ Pa, 5 – $p \approx 5.5 \times 10^3$ Pa, 6 – $p \approx 2.8 \times 10^3$ Pa, 7 – $p \approx 1.4 \times 10^3$ Pa, 8 – $p \approx 6.8 \times 10^2$ Pa).

The next experiments were conducted to determine the required concentration (partial pressure) of doping gas, which would considerably reduce optical breakdown threshold. For this doping was changed as 50 %, 40 %, 10 %, 5 %, 1 %. As can be seen from Fig. 3, if optical breakdown threshold decrease occurred at all, it happened even at small amounts of additives. For Ne – Ar and Ne – Kr mixtures doping rates 1% and 5% of Ar and Kr, respectively, were enough for optical breakdown threshold decrease. No anomalous effect mentioned in [12] for 1% of Ne in Ar (Fig. 3b) was detected. The optical breakdown threshold decrease, as compared to a net background gas was accompanied by an increase of the electrical breakdown threshold, opposite cases were not detected in our experiments. For a mixture of 90% He – 10% Ne ($p \approx 9 \times 10^4$ Pa) we observed an increase of the electrical breakdown threshold (fig. 4a), without reducing the optical one, as pressure decreases, this effect is less pronounced (fig. 4b).

Figure 3. Dependence of electrooptical breakdown threshold of Ne mixtures with Kr (a) and Ar (b) on the electric field and laser intensity ($p \approx 9 \times 10^4$ Pa); a: 1 – 50% Ne – 50% Kr, 2 – 60% Ne – 40% Kr, 3 – 90% Ne – 10% Kr, 4 – 95% Ne – 5% Kr, 5 – 99% Ne – 1% Kr, 6 – 100% Kr, 7 – 100% Ne; b: 1 – 100% Ne, 2 – 100% Ar, 3 – 99% Ne – 1% Ar, 4 – 99% Ar – 1% Ne.
Unlike pure gases and binary mixtures, we could not find in the literature the results of any study of breakdown phenomena in triple mixtures. Performing experiments with gases doped with small amounts of two additives we have found the effect of those additives mutual suppression (negative interference). This phenomena for argon and krypton (both are Penning additives – Fig.2) was observed in the neon (Fig.5).

4. Conclusions
We experimentally determined the thresholds of optical, electrical and combined electrooptical breakdown for noble gases and their binary and triple mixtures with different ratio of components. It was found that the decrease of the threshold for optical breakdown in the presence of the electric field is particularly evident at low pressures. For some binary mixtures the observed decrease of the optical breakdown threshold (the threshold of electrical breakdown in relation to the electrical breakdown threshold of the buffer gas increases), while this effect is already apparent at low concentrations of additives ~1-5%. For triple mixtures we found mutual suppression effects (negative interference) of the additives on breakdown thresholds.
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