Spontaneous emission at 193 nm and gain measurements in F_2 containing excimer gas mixtures

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ABSTRACT

The net gain and spontaneous emission at 193 nm have been measured in X-ray preionized discharges excited by a single pulse charge transfer scheme in Ar and F_2 containing mixtures with He and Ne as a buffer gas. With a pumping pulse of ~ 100 ns (FWHM) and a specific peak power deposition of ~ 1 MW cm^{-3} bar^{-1} in a gas mixture containing F_2 : Ar : He (0.1% : 5% : 94.9%) at 2 bar total pressure the spontaneous emission with a peak intensity of ~ 200 W $srad^{-1}$ and a pulse width (FWHM) of ~ 60 ns was measured. The net gain profile with a peak value ~ 20% cm^{-1} and a pulse width FWHM ~ 60 ns has been obtained under the same pumping conditions.

Keywords: excimer laser, high-pressure gas discharge, gain measurements, spontaneous emission.

1. INTRODUCTION

Excimer lasers are powerful sources of coherent ultraviolet (UV) and vacuum ultraviolet (VUV) radiation and they are used extensively in industry and scientific research. One of our research topics is the molecular fluorine excimer laser emitting radiation at 157 nm. A number of experiments on producing a long (~ 100 ns and longer) discharge pulse in F_2 : He and F_2 : He : Ne gas mixtures have been performed. We have achieved stable and homogeneous discharges in such mixtures during 100 - 150 ns with a peak specific power deposition up to 1 - 2 MW cm^{-3} bar^{-1}.

Important things to know are how strong and how long the spontaneous emission pulse and the spatial and temporal profile of the laser gain in the discharge are. Knowledge of these values allows estimation of the possible laser parameters and rates of kinetic processes as formation and decay of the laser levels.

Radiation of molecular fluorine at 157 nm is in the VUV region and this wavelength region complicates the measurements. Thus, as a first step in our experiments, we decided to carry out spontaneous emission and gain measurements in excimer laser gas mixtures containing in Ar and F_2 producing ArF radiation at a wavelength of 193 nm. Addition of several percent of Ar into a He : F_2 mixture does not change very significantly the electrical parameters of the discharge in terms of power deposition and pulse duration. The emission of these mixtures is at 193 which is in the UV and not in the VUV region and this fact makes the experiments much easier. The results of our experiments with F_2 : Ar : He and F_2 : He : Ar : Ne mixtures are presented in this work.

2. EXPERIMENTAL SETUP AND METHOD.

The experimental setup for spontaneous emission and laser gain measurements is shown in Fig. 1. The discharge chamber (1) consists of a cylindrical quartz tube with inner diameter of 74 mm, two Al flanges and two Al electrodes attached to the flanges. The upper electrode is 50 mm in diameter and serves as anode. The lower electrode is 60 mm in diameter and serves as cathode. Both electrodes have rounded edges to prevent enhancing of the electric field and are covered with a thin layer of Ni. The lower flange has a round opening 1 cm in diameter in the middle, and the lower electrode has a cavity 1 cm in diameter, which forms the input window for the X-ray preionization pulse. The discharge gap between electrodes is adjustable and is 0.5 cm in these experiments.

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The X-ray preionization pulse is produced by a homemade vacuum X-ray source (2). We utilize a vacuum corona-plasma cathode similar to the devices described in Ref. 1 and Ref. 2 to create an electron beam and then convert its energy into X-ray radiation when the e-beam is stopped by a thin Ta foil. The output window of the source is shielded by Pb plates to form a collimated X-ray beam 1 cm in diameter. The X-ray source is powered by a homemade 6-stage mini-Marx generator (3), which is charged by a HV DC power supply (Hypotronics R 60A) (4) and triggered by a homemade triggering unit (5).

A HV pulse-forming unit (7) includes a TGI1-1000/25 thyratron as the main switch, 4 MOhm protective resistances and a 3 nF storage capacitance. An additional 47 kOhm resistance (not shown in Fig. 1) is connected parallel to the discharge chamber to close the circuit and allows charging of the storage capacitor. The pulse forming unit is charged by a HV DC power supply (Hypotronics 8100-10) (8) and the thyratron is triggered by a home-made triggering unit (9). A home-made current transformer (Rogovsky coil) (10) and a home-made high voltage probe (11) are used to measure a discharge current and voltage waveform.

As probe laser for the gain measurements (14) we use an ExciStar XS-200 ArF excimer laser made by TuiLaser (Germany) with a 3 x 6 mm^2 beam cross-section. The laser pulse passes through an aperture (15), which limits the vertical dimension of the laser beam to 5 mm to fit the discharge gap length. The neutral density filter (16) attenuates the pulse energy approximately 500 times. A semi-transparent mirror (17) splits the laser beam. One part comes through a 193 nm filter LC-193 BP 20 (18) to a control photo-diode FND 100Q (19). The other part goes through the middle of the discharge chamber along the diameter, then through an aperture (20) and another 193 nm filter LC-193 BP 20 (21) to the reference photodiode UV 444BQ (22). The second aperture (20) allows us to change the amount of the gathered spontaneous emission due to change of the solid angle. The diameter of the aperture is greater than 5 mm in these experiments.

The signals from both photo-diodes as well as the current and voltage waveforms are traced by a digital oscilloscope Tektronix TDS 640A (12) and processed by a PC (13).

Two pulse generators (Hewlett-Packard 8003A and Farnell PG102) are coupled and are used as a primary generator (6) to trigger the mini-Marx generator, the discharge and the probe laser with the required delays to provide a homogeneous discharge initiation and temporal scanning of the discharge by the probe laser.

Prior to the experiments the discharge chamber is evacuated and then filled with a 5% F_2 : He gas mixture at 2 bar and left overnight to passivate the chamber. Just before the experiments the chamber is evacuated again and filled with the desired F_2 : Ar : He or F_2 : He : Ar : Ne gas mixture at the required gas pressure.

The charging voltage of the discharge pulse forming unit is adjustable and is usually less than 16 kV. Different charging voltages provide different input energies of the discharge pumping pulse.

The photo-diodes have been calibrated and checked for linearity by means of the probe laser and a Molectron EnergyMax500 energy meter prior to the experiments. The transmittances of the neutral density filter, both 193 nm filters, the quartz chamber wall and the beam-splitting mirror, as well as its reflectance, have been measured by means of the probe laser and this energy meter as well.

We record all measurements at the oscilloscope with a bandwidth of 100 MHz. The home-made voltage probe and current transformer are proven to have a wider bandwidth, however, with the 400 MHz bandwidth of the oscilloscope the current and voltage waveforms have the same shape and width but are much more noisy. On the other hand, the 100 MHz bandwidth of oscilloscope provides a high enough temporal resolution.

In the gain measurements we firstly determined the ratio of the amplitudes of the signals from the control (S_c) and reference (S_r) photo-diodes averaged over several (10-20) shots with the discharge chamber filled with the working mixture but with no discharge. Then we measured the amplitudes S_c^* and S_r^* during the discharge pulse with different delays between the initiation of the discharge and the laser pulse. The ratio S_c/S_r (without a discharge) refers to the splitting ratio of the beam, the difference in absorption of two beams along their paths and the different sensitivities of the photo-diodes. The second ratio S_c^*/S_r^* shows how the first one changes due to amplification of the laser pulse or its absorption in the chamber during a discharge. Thus, we determine the net gain along the path L, spatially averaged, as follows:

$$(g-a)*L = ln(\frac{S_c^*/S_r^*}{S_c/S_r}) = ln(\frac{S_c^*/S_c}{S_r^*/S_r}). \quad (1)$$

In other words, we look for a change of the control photo-diode signal due to amplification or absorption of the laser pulse in gas during a discharge S_c^*/S_c and correct this ratio by a factor S_r^*/S_r which describes the fluctuations of the laser pulse energy and intensity.

3. EXPERIMENTAL RESULTS AND DISCUSSION

In Fig. 2 typical measured discharge voltage (a) and current (b) waveforms, the calculated power deposition (c) and the spontaneous emission (d) measured by the control photo-diode are shown. In this case a discharge in F_2 : Ar : He (0.1% : 5% : 94.9%) mixture at total pressure 2 bar and charging voltage 14 kV is used. The voltage increases until breakdown of the gap, then the voltage collapses to approx. 4 kV as the discharge starts and then slowly decreases. The current changes slower, and it has a half of sine shape (in Fig. 2b only a part of the current waveform is shown). In this particular case the current reaches the maximum value at approximately 100 ns after the initiation of the discharge. The first sharp peak of the calculated power deposition waveform has no meaning since the current is very low during this period. Thus we look for the amplitude of the second long part of the power deposition waveform, which in this case is approximately 750 kW, and the half-width of the pulse is approximately 100 ns. The half-width of the spontaneous emission signal is approximately 60 ns, and is shorter than the duration of the pumping pulse. Roughly, the spontaneous emission follows the pumping density, and the maximum of the emission appears at the moment of the maximum of the pumping density. Since we have calibrated the photodiodes by means of the probe laser and have measured the solid angle, within which the spontaneous emission is gathered, it is possible to calculate the light intensity in terms of power emitted in a unit solid angle. In this case the peak value is approximately 240 W srad^{-1}.The discharge appears in the middle of the preionized area. At low charging voltages it occupies only the small central part of it, several millimeters in diameter. As the charging voltage increases the current rises almost linearly, and so does the visible diameter of the discharge. In this region the quasi-steady-state voltage is constant, so the power deposition also linearly rises. This behavior resembles the well-know normal current density effect for the DC discharges. At higher charging voltages the discharge occupies the entire preionized volume, and the cross-section of the discharge is then approximately 1 cm or slightly greater in diameter. At even higher power deposition the discharge appears as an arc from the beginning.

It is also worth mentioning that at low power deposition (< 200 kW) the FWHM of the pumping pulse is slightly longer: approximately 140-160 ns. With an increase of the charging voltage the FWHM decreases and at a pumping power deposition higher than 600 kW it stays constant at about 100 ns. The FWHM of the spontaneous emission is almost constant at any pumping power density and it is about 60 ns with 10 ns accuracy.

The dependence of the peak spontaneous emission intensity on the peak power deposition is shown in Fig. 3 for different gas mixtures. In Fig. 3a the total gas pressure of a F_2 : Ar : He mixture is 2 bar, the concentration of Ar is 5% and the concentration of F_2 is 0.075% (1), 0.1% (2) and 0.15% (3). In Fig. 3b the total gas pressure of the F_2 : Ar : He mixture is 2 bar, the F_2 concentration is 0.1% and the Ar concentration is 2.5% (1), 5% (2) and 10% (3). In Fig. 3c the results for a F_2: Ar : He (0.1% : 5% : 94.9%) gas mixture at total pressure 1.5 (1), 2 (2) and 3 bar (3) are shown. Finally, in Fig. 3d the results for F_2 : Ar : He (0.1% : 5% : 94.9%) and F_2 : He : Ar : Ne (0.1% : 1.9% : 5% : 93%) are shown. In all cases the peak spontaneous emission intensity grows with the peak pumping power deposition and then reaches saturation. The saturation appears roughly when the discharge reaches its maximum cross-section, defined by the preionization area. However, the saturated light intensity and the power deposition, at which the saturation is reached, depend on the gas mixture composition and total pressure. For example, in Fig. 3c it is shown that at lower total gas pressure the saturation appears at lower pumping power and the saturated spontaneous emission intensity is lower. A higher concentration of F_2 leads to a higher saturation pumping power and a higher saturated light output, as in Fig. 3a. The spontaneous emission from the discharge in excimer laser gas mixtures with Ne as a buffer gas is slightly stronger than from discharges in He-based mixtures, as shown in Fig. 3d. For various Ar concentrations (Fig. 3b) the situation is more complicated: the emission is the weakest in the case of 5% Ar concentration (Fig. 3b (2)) and the strongest in the case of 2.5% Ar concentration (Fig. 3b (1)), while the discharge in a mixture with 10% of Ar produces an intermediate light intensity.

During the gain measurements we reduced the second aperture diameter thus decreasing the amount of the gathered spontaneous emission radiation. We chose the aperture size such that the control photo-diode response to the peak

spontaneous emission intensity is comparable to its response to the laser pulse with a specific energy ~ 500 nJ cm^{-2}. Then we observed the laser pulse passed through a discharge on a background of the spontaneous radiation of the discharge, as shown in Fig. 4a. After that we subtracted the over 10 shots averaged spontaneous emission waveform, measured under the same discharge excitation conditions, but without firing the probe laser. The restored signal is shown in Fig. 4b. In Fig. 4c the corresponding signal from the reference photo-diode is shown. Thus we defined S_c^* (Fig. 4b) and S_r^* (Fig. 4c) in Eq. 1 as the amplitudes of these signals. S_c and S_r are measured 10-20 times with the same alignment of the experimental setup, but without discharge initiation, and then their ratio is averaged over these shots to produce the S_c/S_r term of the Eq. 1.

In Fig. 5 the temporal evolution of the net gain (g-a)*L is shown. The discharge is excited in a F_2 : Ar : He (0.1% : 5% : 94.9%) gas mixture at 2 bar total pressure and at a charging voltage of 14 kV. The net gain is determined by Eq. 1, where S_c, S_r, S_c^* and S_r^* are measured in the experiments as described above. The solid squares (1) are the experimental points, the open circles with error bars (2) are the averaged values. The horizontal error bar represents the time interval of averaging for a group of the experimental points, the vertical error bar shows the standard deviation. The dashed line (3) fits the results. The FWHM of the gain profile is approximately 60 ns, the same as the FWHM of the spontaneous emission waveform. The peak net gain is approximately 18 ± 8%.

The dependence of the peak (maximum) net gain (g-a)*L on the peak pumping power deposition is shown in Fig. 6a for discharges in F_2 : Ar : He (0.1% : 5% : 94.9%) (1) and F_2 : He : Ar : Ne (0.1% : 1.9% : 5% : 93%) (2) gas mixtures at 2 bar total pressure. The corresponding peak intensities of the spontaneous emission, measured together with the gain, are shown in Fig. 6b. The net gain and the spontaneous emission intensity grows with the increase of the pumping power in both cases. When Ne is used as a buffer gas, the net gain and the spontaneous emission are stronger than in the case of He as a buffer gas. Also, in the case of Ne the net gain and the emission intensity reach saturation at a pumping power of approximately 650 kW. In the case of He the saturation has not been observed. The maximum obtained gain is about 20 ± 8% in both cases of Ne and He as buffer gas.

The shot-to-shot deviation of the measured net gain is high within every single group of points close to each other in the time domain. Due to this fact, the experimental error is large. First of all, the procedure of subtracting the background radiation is not very accurate. Secondly, the discharge itself is not very stable from shot to shot, in fact, the spontaneous emission intensity also fluctuates, as it is shown in Fig. 3. Thus, we believe that the population of the upper laser level fluctuates as well, and hence, should do the net gain. And the last and the most important source of errors is fluctuation of the probe laser beam polarization. Actually, according to the manufacturer, the TuiLaser company, the polarization of the laser radiation is random in every shot. Due to the fact that part of the beam is reflected by a dielectric semi-transparent mirror and another part is transmitted by the mirror, the ratio of the signal amplitudes after the beam-splitting is determined by the transmission and reflection coefficient of the mirror. These coefficients depend on the wavelength, angle of incidence of the beam and the polarization of the beam, which is not constant. Indeed, we observe that the photo-diodes signal amplitudes ratio S_c/S_r fluctuates up to 5% of its value even without a discharge ignition. The same fluctuation occurs during gain measurements, so the error becomes larger, as we take ratio of two fluctuating values. And, of course, the logarithm function makes situation worse:

$$ln(x+\delta) = ln(x(1+\delta/x)) = ln(x) + ln(1+\delta/x) \approx ln(x) + \delta/x, \qquad (2)$$

where $x \sim 1$, $\delta < x$.

4. CONCLUSION

With a very simple single pulse charge transfer excitation scheme we obtained a stable discharge in different Ar and F_2 containing excimer laser gas mixtures across a 0.5 cm gap in a plane-to-plane electrode configuration with a 1 cm diameter X-ray preionized volume. The discharge current and pumping power deposition, the spontaneous emission intensity and net gain increased with the charging voltage. As the charging voltage increases the discharge cross-section grows until the discharge occupies the entire preionized volume. After the discharge cross-section has become constant, the spontaneous emission and gain saturates, but the current and power deposition keep growing with the increase of the charging voltage.

The observed spontaneous emission intensity and gain depend on the gas mixture composition and total gas pressure. For example, in a discharge in F_2 : Ar : He (0.1% : 5% : 94.9%) at 2 bar and 16 kV charging voltage, a pumping pulse with peak power ~ 1 MW (specific peak power deposition ~ 1 MW cm^{-3} bar^{-1}) and a FWHM of ~ 100 ns is obtained. This pumping pulse leads to an observed spontaneous emission pulse with peak intensity ~ 200 W $srad^{-1}$ and a FWHM ~ 60 ns. During this pulse we measured the net gain with a peak value ~ 20 ± 8% and a FHWM of ~ 60 ns, which roughly follows the spontaneous emission pulse waveform.

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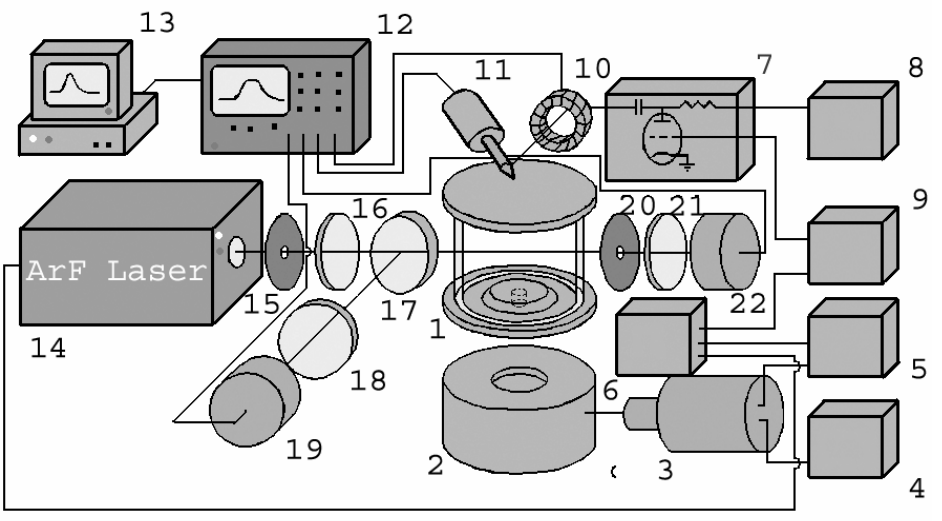

Fig. 1. Experimental setup for spontaneous emission and laser gain measurements. (1) discharge chamber; (2) – vacuum X-rays source; (3) 6-stage mini-Marx generator; (4) HV DC power supply; (5) triggering unit; (6) primary pulse generator; (7) HV pulse forming unit; (8) HV DC power supply; (9) triggering unit; (10) current transformer (Rogovsky coil); (11) HV probe; (12) oscilloscope; (13) PC; (14) probe laser; (15) aperture; (16) neutral density filter; (17) beam-splitting mirror; (18) 193 nm filter; (19) reference photo-diode; (20) aperture; (21) 193 nm filter; (22) control photo-diode.

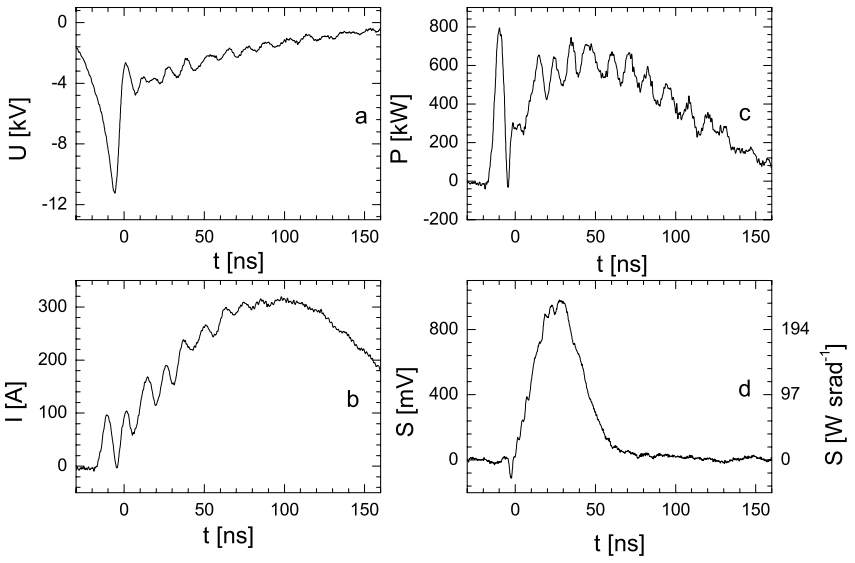

Fig. 2. (a) Measured discharge voltage waveform. (b) Measured discharge current waveform. (c) Calculated power deposition waveform. (d) Measured waveform of the spontaneous emission from the discharge: left hand side vertical axis – measured signal from the control photo-diode in mV, right hand side axis – calculated light intensity in W srad^{-1}. Discharge in F_2 : Ar : He (0.1% : 5% : 94.9%) at a total pressure 2 bar and charging voltage 14 kV.

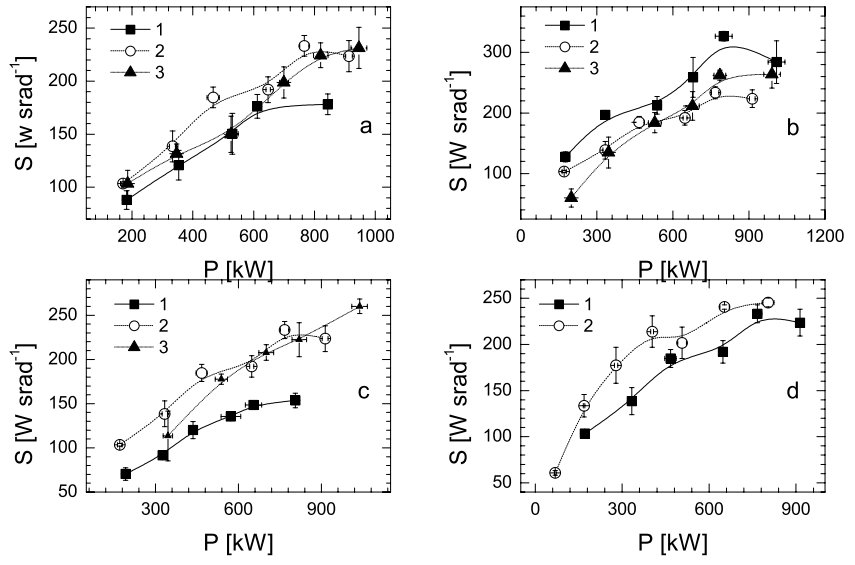

Fig. 3. Dependence of the maximum spontaneous emission light intensity S on the maximum power deposition into the discharge. (a) Mixture with 0.075% (1), 0.1% (2), 0.15% (3) of F_2 and 5% of Ar in He as a buffer gas at a total pressure 2 bar. (b) Mixture with 0.1% of F_2 and 2.5% (1), 5% (2), 10% (3) of Ar in He as a buffer gas at a total pressure 2 bar. (c) F_2 : Ar : He (0.1% : 5% : 94.9%) mixture at total pressure 1.5 (1), 2 (2) and 3 bar (3). (d) F_2 : Ar : He (0.1% : 5% : 94.9%) and F_2 : He : Ar : Ne (0.1% : 1.9% : 5% : 93%) mixtures at a total pressure of 2 bar.

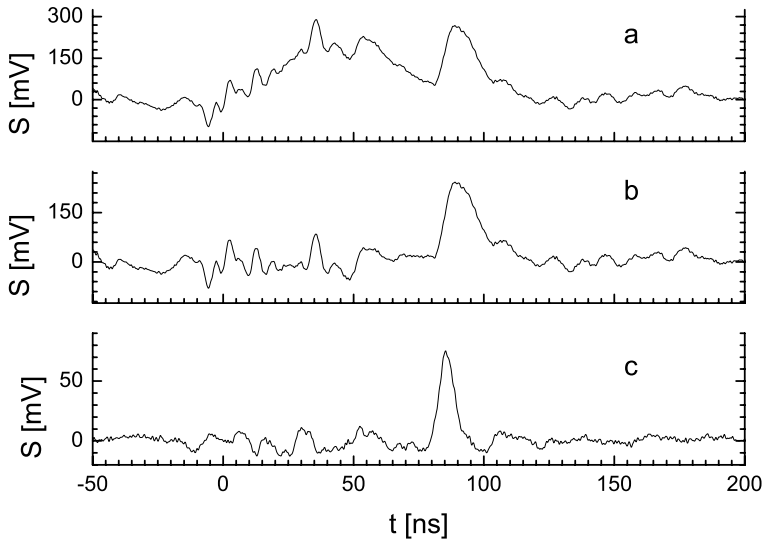

Fig. 4. (a) Measured control photo-diode signal (spontaneous emission and amplified laser pulse). Discharge in a F_2 : Ar : He (0.1% : 5% : 94.9%) mixture at a total pressure of 2 bar and charging voltage 14 kV. (b) Restored amplified laser pulse signal from the control photo-diode. (c) Measured reference photo-diode signal during the same discharge pulse.

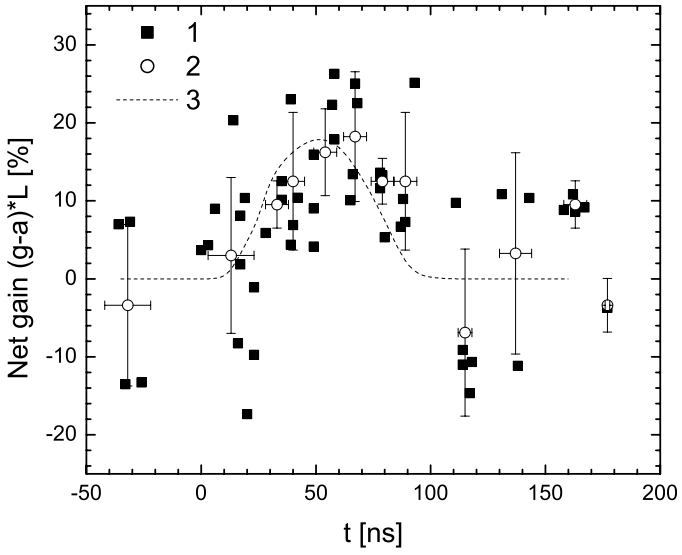

Fig. 5. Temporal evolution of the net gain (g-a)*L measured in a F_2 : Ar : He (0.1% : 5% : 94.9%) mixture at a total pressure of 2 bar and a charging voltage 14 kV. (1) Experimental points, (2) averaged values, (3) fit line.

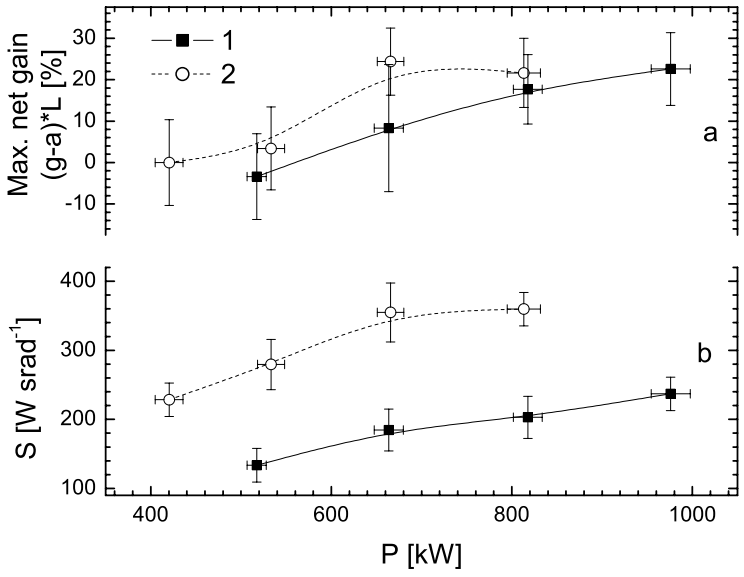

Fig. 6. Dependence of the peak net gain (g-a)*L (a) and peak spontaneous emission light intensity (b) on the peak power deposition in F_2 : Ar : He (0.1% : 5% : 94.9%) (1) and F_2: He : Ar : Ne (0.1% : 1.9% : 5% : 93%) mixture (2) at total pressure of 2 bar.