Measurement of negative ion density in streamer discharge in air by transient cavity ringdown spectroscopy

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Atmospheric-pressure discharges generated in air are expected to be electronegative, since oxygen and water vapor are electronegative gases. However, experiments that examine negative ion densities in atmospheric-pressure discharges are limited to date. In this work, we measured the temporal variation of the negative ion density in a streamer discharge generated in air [1]. We adopted cavity ringdown spectroscopy (CRDS). An issue of CRDS for the detection of negative ions (the detection of the photon loss due to photodetachment of negative ions) in streamer discharge is the fact that the lifetime of negative ion is shorter than the ringdown time. We solved this issue by applying the transient analysis of the ringdown curve.

A streamer discharge was generated between needle (the anode) and planar (the cathode) electrodes placed inside an optical cavity in air. The optical cavity was composed of two mirrors with high reflectivities at 770-785 nm. The cavity length was 20 cm. The distance between the needle and planar electrodes was 7.5 mm, and the distance between the optical axis of the cavity and the needle electrode was 2.5 mm. The needle electrode was connected to a high-voltage pulsed power supply, and the planar electrode was electrically grounded. We employed an optical parametric oscillator (OPO) as the light source in the CRDS measurement. The wavelength and the duration of the OPO laser pulse were 777 nm (the idler output) and 10 ns, respectively. The OPO laser oscillated at 5 μs before the the trigger of the high-voltage pulse, and the repetition frequencies of the laser oscillation and the discharge were 10 Hz. The laser pulse transmitted through the cavity was detected using a photomultiplier tube (PMT).

Figure 1 shows typical ringdown curves. The ringdown curves were obtained by averaging 300 laser shots using a digital oscilloscope. The origin of the horizontal axis corresponded to the trigger to the high-voltage pulsed power supply. As shown in the figure, the ringdown curve in the absence of the discharge was approximated by an exponential function. In the presence of the discharge, we detected the superposition of the discharge noise at $0 \leq t \leq 0.3$ μs. After the noise disappeared ($0.4 \leq t \leq 1.5$ μs), we observed a steeper decrease in the transmitted laser intensity than that in the absence of the discharge. The time constant of the ringdown curve was almost the same as that in the absence of the discharge at $t \geq 1.5$ μs.

Conventional CRDS supposes a steady-state absorber, and the absorber density is deduced from the time constant of the ringdown curve. However, in the present experiment, the negative ion density had a faster time constant than the ringdown curve. In this case, we can deduce the temporal variation of the negative ion density by the transient analysis of the ringdown curve. The negative ion density is obtained by

$$n(t) = -\frac{L}{c\sigma l}\left(\frac{1}{I(t)}\frac{\mathrm{d}I(t)}{\mathrm{d}t} + \frac{1}{\tau_0}\right), \qquad (1)$$

where $I(t)$ represents the ringdown curve in the presence of the discharge, τ_0 is the ringdown time constant in the absence of the discharge, L is the cavity length, l is the length of the discharge zone along the axis of the cavity, c is the speed of light, and σ is the cross section of optical absorption or photodetachment at 777 nm. $1/\tau_0 = 1.31 \times 10^5$ s^{-1} was deduced from the ringdown curve shown in Fig. 1. We needed the moving average of the ringdown curve in the numerical calculation of $\mathrm{d}I(t)/\mathrm{d}t$ since the differential of the experimental

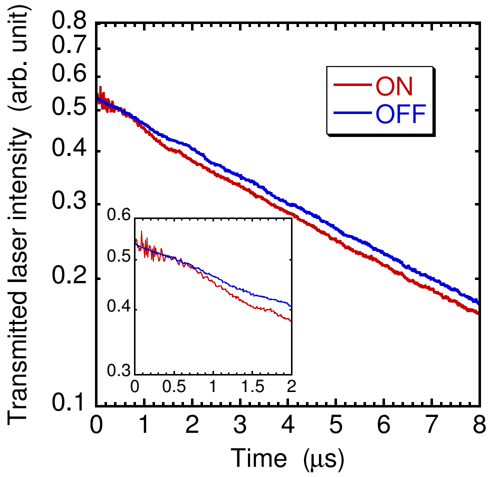

Fig. 1: Typical ringdown curves observed in the presence and absence of streamer discharge.

ringdown curve was a noisy procedure.

The temporal variation of the negative ion density was obtained as shown in Fig. 2(a) by calculating Eq. (1). We adopted the moving average with a duration of $\Delta t = 0.4$ μs. $l = 3$ mm was assumed based on the optical emission image of the streamer discharge, and we assumed $\sigma = 1 \times 10^{-22}$ m^2, which is the photodetachment cross section of O_2^- at 777 nm [2]. We cannot say anything about the phenomena at $t \leq 0.4$ μs, since we adopted moving average with $\Delta t = 0.4$ μs. The negative ion density started the increase at $t = 0.4$ μs, and the maximum negative ion density was observed at $t = 1$ μs. The maximum negative ion density was 2×10^{20} m^{-3}. Figures 2(b) and 2(c) show the waveforms of the pulsed voltage and the discharge current, respectively. It is understood from Fig. 2 that the increase in the negative ion density started after the disappearances of the discharge voltage and current. The decrease in the negative ion density was steep, and it was around the noise level at $t = 1.5$ μs.

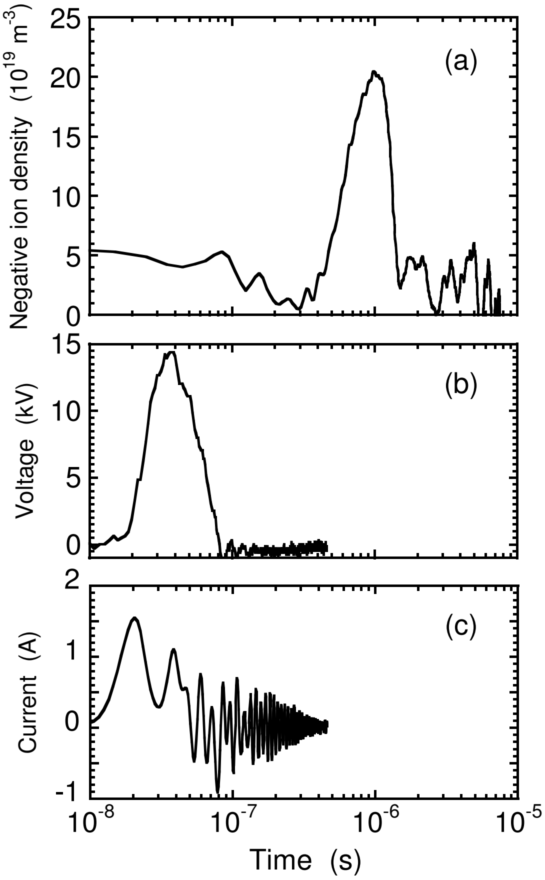

Fig. 2: Temporal variations of (a) negative ion density, (b) discharge voltage, and (c) discharge current.

It is well known that streamer discharge is composed of primary and secondary streamers [3]. The primary streamer has a high reduced electric field and a low negative ion density. It is estimated that the primary streamer arrives at the cathode at $t < 50$ ns in the present experimental condition [4]. Hence, it is considered that Fig. 2(a) represents the evolution of the negative ion density in the secondary streamer. The secondary streamer with a much lower reduced electric field enhances the electron attachment frequency to electronegative molecules, resulting in the evolution of the negative ion density at $t \geq 0.4$ μs. On the other hand, the decrease in the negative ion density at $1 \leq t \leq 1.5$ μs is consistent with the destruction of negative ions via mutual neutralization with positive ions. This is because the rate coefficient of mutual neutralization such as $O_2^- + O_2^+ \rightarrow O_2 + O_2$ is on the order of 10^{-13} m^3/s [5, 6].

In summary, we have measured the absolute negative ion density in a streamer discharge in air by transient CRDS. The present work shows the applicability of transient CRDS to the measurement of negative ion densities in atmospheric-pressure discharges.

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