A Note on the Throbbing of Glow Discharges in Gases at Low Pressures

Rafi M. Chaudhri and Tasneem Zahra

Centre for Solid State Physics University of Punjab, Lahore Pakistan

and M. Munawar Chaudhri

Cavendish Laboratory, Cambridge, U.K.

The phenomenon of the throbbing of glow discharges was first studied by Thomson [1] in 1931. The discharge tubes he used were probably covered with a sputtered film and the gas pressure was not controlled. He explained this phenomenon by the electrical charging of the tube walls. Since Thomson's paper there has been virtually no work on this interesting phenomenon. In this note we report on throbbing phenomena in tubes free from a sputtered film and suggest that in this situation the mechanism is the formation and decay of the negative ions in the gas rather than electrical charging of the walls of the tube.

Three types of discharge tubes (all of Pyrex glass) were investigated: in one, both electrodes were 38 mm diameter circular discs, while in the other two the anode was a hollow cylinder and the cathode either a thin co-axial wire or a circular disc placed coaxially at a short distance from one end of the cylinder. The internal diameter of the glass tube was 40 mm and its length 200 mm; another tube had an internal diameter of 120 mm, much larger than the diameter of the electrodes so as to eliminate, as much as possible, the impact of ions and electrons on the tube's walls during a run. The tube with both electrodes cylindrical had an anode of diameter 25 mm and length 100 mm, and the cathode was a 0.2 mm diameter tungsten wire placed along the axis of the anode. In the third type of tube the anode was a hollow cylinder 40 mm in diameter and 300 mm in length and the cathode was a 40 mm diameter tungsten disc placed 10 mm from one end of the anode.

The capacity of the power supply used was 0 — 5 kV d. c. at 20 mA with ripples of 50 parts per million peak to peak at full load. The discharge current was monitored on a chart recorder connected across a very small series resistance of a few ohms. The voltage drop across it was too small to affect the discharge phenomenon. The gases used were oxygen, nitrogen, carbon dioxide, and air. For the tubes with plane electrodes, the current and voltage were in the ranges 0.3 — 5 mA and 300 — 3000 V, respectively, whereas for tubes with hollow anodes these quantities were in the ranges 0.1 — 2 mA and 300 — 600 V.

The experimental procedure for tubes with plane electrodes was as follows: first the tube was evacuated down to 10^{-6} torr and then the gas to be investigated was allowed in through an adjustable leak so that a desired pressure could be maintained throughout an experiment. The discharge voltage was then gradually increased until a glow discharge appeared. At this stage the anode was gradually brought towards the cathode. Initially, this did not alter the current significantly. However, when the anode was just outside the cathode dark space the discharge was no longer steady but throbbed with a regular frequency as indicated by the luminosity from the tube and by the current-time record (when the anode was within the cathode dark space, the discharge stopped completely). If these conditions were stricktly maintained, the throbbing continued as long as the tube remained free from a sputtered film. It should also be pointed out that in a very clean and virgin tube (i. e., without any sputtered film on the tube walls) with a suitable gap between the electrodes and a proper gas pressure, the throbbing appeared at the first application of the discharge voltage without there being any continuous discharge. The throbbing was observed in all gases studied with the gas pressure being in the range 0.085 — 0.4 torr. To obtain the throbbing in the tubes with hollow anodes, the gas pressure and discharge voltage were carefully adjusted instead of the interelectrode distance.

In all the throbbing experiments the conditions and observations were very reproducible. The main features of the phenomenon were: (i) the current oscillated between zero and a maximum with a regular frequency which was dependent upon the

discharge parameters, (ii) depending upon the discharge conditions the "on" and "off" periods of a single throb were in the ranges $0.25 - 1.25$ s and $0.25 - 1.75$ s, respectively, and (iii) during a single throb the current rose sharply from zero to a maximum, decayed to $60 - 70\%$ of the maximum value and then finally dropped suddenly to zero and remained so for a certain time before rising suddenly again for the next throb cycle.

It may be emphasised that the throbbing in these experiments is not due to any charging of the walls, the impact of ions and electrons on the tubes' walls, or the external circuit. These conclusions are drawn from experiments with virgin tubes and those whose diameters were considerably larger than the electrode size. We suggest that the throbbing occurs due to the formation of negative ions (during the "on" period of the discharge) and their destruction (during the "off" period). A similar explanation, based on the formation and sweeping away of the negative ions of oxygen [2, 3], has been put forward for the negative point Trichel pulse corona in air.

In a self-sustained glow discharge it is the secondary electrons [4] from the cathode which produce sufficient ionisation between the cathode and the edge of the negative glow and thus maintain the discharge. However, during the transit of secondary electrons through the cathode dark space in an electronegative gas, negative ions will form due to the attachment processes [5]. The destruction of negative ions will also occur simultaneously, but at a different rate [6, 7]. Under suitable conditions a net density of negative ions will result. Because of their low mobility as compared with that of electrons, the electric field at the cathode will reduce to below the critical value and the discharge will stop. After the cessation of the discharge the rate of formation of negative ions will become considerably smaller than that of their destruction. The field then starts to recover and after full recovery the discharge will strike again. The whole cycle will continue to repeat as long as the discharge conditions are maitained constant and the tube walls are without a metallic film which could be formed by the deposition of neutrals and ions sputtered from the cathode [8].

We may make an approximate quantitative analysis of the phenomenon for the case of oxygen as the rate constants of attachment and detachment processes are known for this gas. The various reactions of our interest are:

(i) $O_2 + e \rightarrow O + O^-$ (dissociative attachment)
with rate constant k_1 of 1.3×10^{-12} cm^3 s$^-$ [5],

(ii) $V^- + e \rightarrow O + 2$ e (detachment by electron collision)
with rate constant k_2 of 4×10^{-12} cm^3 s^{-1} [6], and

(iii) $O^- + O \rightarrow O_2 + e$ (associative attachment)
with rate constant k_3 of 1.9×10^{-10} cm^3 s^{-1} [7].

We make the simplifying but justifiable assumptions that (a) during the "on" period the electron and the molecular oxygen densities, n cm^{-3} and n' cm^{-3}, respectively, remain constant, and (b) the density of the atomic oxygen is the same as that of the oxygen negative ions and is denoted by N cm^{-3}. The electron density in the negative glow is approximately the same as that of the positive ions, and for a current of 1 mA and the cathode area of 11.2 cm^2 this comes out to be $\sim 10^9$ cm^{-3}. The molecular oxygen density for the case being considered is 3.6×10^{15} cm^{-3}. The time variation of the oxygen negative ion density N at the cathode end of the negative glow can be given by the equation

$$\dot{N} = k_1\, n\, n' - k_2\, n\, N - k_3\, N^2 \qquad (1)$$

in which the first term on the right hand side is the rate of formation of negative ions and the other two terms give rates of their decay. On integerating and taking $N \rightarrow 0$ for $t = 0$ (i. e., the beginning of the "on" period) we obtain

$$N = 3 \times 10^{11}(1 - \exp(-60\,t))/ \\ (1 + \exp(-60\,t)) \qquad (2)$$

in which the values of the rate constants have been substituted. Equation (2) shows that the negative ion concentration increases exponentially with time at first and then approaches a constant values of 3×10^{11} cm^{-3} asymptotically. Assuming that about 99% of this density of the negative ions will be sufficient to stop the discharge, we estimate the "on" period of a throb as 0.1 s. This is roughly in agreement with the experimental values given above. It may be added that the gradual decrease of the current from the maximum value during the "on" period before its sudden drop to zero may be an indication of the build up of the negative ion concentration.

After the cessation of the discharge the formation of the negative ions will virtually stop and their concentration will start to decrease with time due to

the detachment processes and due to the sweeping away by the field. When this concentration drops to only a few percent of the maximum value, the discharge will strike again. The recovery phase may be represented by the equation

$$\dot{N} = - k_3 N^2 . \tag{3}$$

On integeration, we get

$$1/N = 1.9 \times 10^{10} t + 3.3 \times 10^{-12} , \tag{4}$$

where the initial concentration of the negative ions at the beginning of the "off" period has been taken as 3×10^{11} cm^{-3}. From (4) the time taken for the

concentration to reduce to a low value, i. e., 1% of this density is 3.3 s. This time is also in agreement with the experimental values of the "off" period.

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