# Equilibrium and Nonequilibrium Electrical Conductivity of a Potassium-Seeded Argon Plasma

By

# Fujihiko Sakao\* and Hiroshi Sato

Summary: Experiments have been made on the electrical conductivity of argon-potassium and helium-potassium plasmas at atmospheric pressure. The gas, its container, and electrodes are heated uniformly in an electric furnace to a temperature ranging from 1300 to 1700°C. Thermal ionization of the potassium atoms makes the gas electrically conductive. Through the ionized gas an electric current of short duration is passed, the current density ranging from 10<sup>-5</sup> to 2 A/cm<sup>2</sup>. As a function of the current density, the electric field in the plasma is determined with two electrostatic probes and effects of the sheath-voltages on the main electrodes are avoided. The gas temperature is not disturbed by the current because its duration is very short. With small current density the conductivity is independent of the current density and the plasma is in a thermal equilibrium. From the experimental value of conductivity the average cross section of a potassium atom for electron-momentum transfer is estimated to be  $2.3 \times 10^{-14}$  cm<sup>2</sup>. For intermediate current density the conductivity becomes lower than the equilibrium value. The electrons may not be in Maxwellian distribution. The decrease in the conductivity is interpreted on the bases of the non-Maxwellian distribution of the electron energy and the decrease in the mobility of electrons at high electron temperature. The apparent conductivity at high current density is large, but the current density in the plasma may not be uniform.

#### 1. Introduction

Recent developments in the magnetohydrodynamic propulsion and power generation stimulated interests in the flow of an ionized gas. Many theoretical works have been done on the electric property of high-temperature inert gas seeded with a material of low ionization potential, such as cesium, potassium, or sodium. An inert gas atom has small cross sections for both momentum and energy transfer with electrons, which lead to a high mobility of electrons and a high electrical conductivity. Alkali atoms are easily ionized and yield a large number of electrons at moderate temperature. When the ionized gas is subject to an electric field, electrons are accelerated by the field and gain energy. The energy transfer from electrons to atoms is hindered by the small cross section of an inert gas atom for energy transfer with electrons. Consequently, electrons have higher energy than neutral atoms on the average. For such a nonequilibrium condition we define the electron temperature which is higher than the gas tem-

<sup>\*</sup> Faculty of Engineering, Hiroshima University.

perature. If the ionization process is governed by electrons, we expect a higher electron number density than that corresponding to the gas temperature. Since the conductivity is proportional to the electron number density, the conductivity is higher in the nonequilibrium state. There are many theoretical investigations on the nonequilibrium conductivity, while experimental investigations are relatively few.

Harris [1, 2] measured the electrical conductivity of various inert gases seeded with cesium and potassium near thermal equilibrium. The temperature of the gas was made uniform in his experiment. The conductivity is calculated from the current and the voltage across electrodes with a small dc current. The voltage across a sheath on the electrode was ignored in the calculation of the electric field in the plasma. Although he claims that the sheath voltage is negligibly small at a small current, it is not necessarily true, as will be shown by the present experiment. The experiment by Harris was restricted to an equilibrium plasma. No investigation was made on a nonequilibrium plasma. Kerrebrock, Hoffman, and Dethlefsen [3, 4] and Zukoski, Cool, and Gibson [5, 6] made experiments on the conductivity of a plasma at high current density. The conductivity of the high-current nonequilibrium plasma was found to be much higher than that of low-current equilibrium plasma. The experimental values of conductivity are in good agreement with the calculated values based on the "two-temperature conduction theory" provided that effects of radiation [7] are taken into account properly. Further, a consideration on the electronic heat conduction [8] made the agreement more satisfactory. The electric field in the plasma was measured with probes set between main electrodes so that effects of the sheath was excluded. On the other hand, they employed a test plasma flowing in a cold passage in a marked contrast to the isothermal experiment by Harris. The temperature of the plasma is not uniform and there is some ambiguity in the value of gas temperature. Thus, at low current where the gas temperature is one of the most important parameters, their results may be subject to some uncertainty.

In the experiment by Kerrebrock and Hoffman [3] they noticed that at a certain current density the conductivity of argon-potassium mixture seemed to decrease with the increase in the current density. This relation between the conductivity and the current density can not be explained by the conventional two-temperature theory. According to the theory, the conductivity should always increase when the current density is increased. The unusual dependence of the conductivity of the conductivity on the current density was not appropriately interpreted. Moreover, the range of the current density in their experiment was not wide enough to cover the equilibrium region.

We may summarize results of previous experimental investigations on the subject as flolows:

Some of them are accompanied by effects of sheath, and others by nonuniform gas temeprature. Equilibrium and nonequilibrium conditions were not measured in the same arrangement. An interesting phenomenon appearing in an intermediate region between the equilibrium and nonequilibrium regions has not been

fully clarified.

In the present investigation an isothermal condition of plasma is realized experimentally. The temperature of the plasma and the test chamber is maintained uniform and is measured accurately. The sheath voltage on the main electrodes and the electric field in the bulk of plasma are measured separately by means of electrostatic probes. The current density in the plasma covers a wide range from the equilibrium to nonequilibrium conditions. At the lowest current, the plasma is in thermal equilibrium. At the highest current the ionization is enhanced due to the elevated electron temperature and the plasma is not in equilibrium. Between these two states there is a transient region, in which the conductivity shows a minimum for argon-potassium mixture under certain experimental conditions. Considerations on the microscopic processes in a nonequilibrium plasma led to the conclusion that the energy distribution of electrons might deviate from being Maxwellian. Based on the non-Maxwellian distribution, a plausible interpretation for the dependence of the conductivity on the electric field is obtained.

Essentials of the results obtained here have been published elsewhere [9]. However, there still remain unpublished details. This paper presents more complete informations.

# 2. Experimental Arrangements

Typical experimental conditions are as follow:

Gas composition; argon seeded with potassium vapor (in some cases helium is substituted for argon for comparison).

Gas temperature  $T_g$ ; 1400, 1500, and 1600°C.

Total pressure; 1 atm.

Mole fraction of potassium; 0.013 and 0.13% (in few cases 0.8%).

Current density; 10<sup>-5</sup>~2 A/cm<sup>2</sup>.

In order to realize such conditions, there are two methods. In some of the previous experiments [3, 4, 5, 6] the heated plasma passes through a test section where the electric properties are measured. By this arrangement it is possible to keep the temperature of the test section low. A material suitable for the electrically insulating wall is easily found. Since the plasma flows, the leak of gas through the wall causes little trouble. Thus the wall may be slotted to permit optical observation. On the other hand, the temperature is not uniform in the plasma. The temperature is low near the wall. Since there is no radiation from the cold wall, the radiation from the plasma to the wall may disturb the equilibrium condition even in the absence of the electric current. Owing to the nonuniformity in the temperature and to the presence of cooling by radiation to the wall, extreme care is needed in the measurement of the gas temperature. Possible temperature gradient in the streamwise direction might result in the number density of electrons frozen to the value at upstream section, where the gas temperature is higher [4]. Finally, the leak of electric current can not be excluded completely, because a conducting layer is formed on the wall by a reaction with the potassium vapor

in the plasma [7].

In another scheme, the plasma is contained in a opaque vessel and heated from outside, as adopted by Harris [1,2]. The temperature is uniform over the plasma, electrodes, and the wall of the vessel. Hence, the thermal equilibrium in absence of the electric current is secured. The gas temperature is easily determined. On the other hand, the high-temperature wall becomes electrically conducting. The leak of electric current through the wall is a source of error in the measured value of the conductivity. In order to minimize the leak, the material for the wall should be carefully chosen. Nevertheless, once a proper material is found, the leak is small and can be corrected for.

From the comparison between the two methods, it is clear that the isothermal experiment is preferable for a near-equilibrium experiment. Since we are to investigate a plasma in the intermediate region between the equilibrium and non-equilibrium region, we follow the isothermal scheme.

Figure 1 illustrates an outline of the experimental arrangements. Argon with impurity less than 0.1% is supplied from a 50 litre bottle at the pressure 150 kgw/cm². Through a series of pressure reducing valves, the pressure of argon is made slightly higher than the atmospheric at the entrance of the flow meter. A safety vent prevents the pressure from rising too high. A flow of argon with the flow-rate of 1 to 2 cm³/s (at 1 atm, room temperature) passes the potassium boiler and is mixed with the potassium vapor. The mixing is based on the vapor saturation at a prescribed temperature. In the potassium boiler argon is first heated up to the prescribed temperature in passing through a 50 cm-long, 4 mm-i.d. tube of stainless steel immersed in a bath of molten tin-lead mixture. Then, the argon is led into a bubble pot immersed in the same bath. The pot has an inner height of 105 mm and an inner diameter of 25 mm. At the bottom of the pot there is a pool of liquid potassium. Argon is introduced as bubbles in the potassium pool.

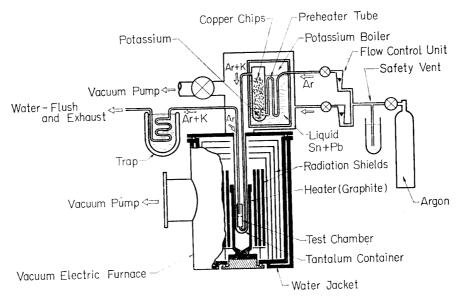


Fig. 1. Experimental arrangements.

In order to provide enough vapor the temperature of the pool is kept deliberately higher than the prescribed value by a small sheathed heater at the bottom of the pot. In the bubble pot above the potassium pool, copper chips are packed. The wide surface area of those chips wet with potassium ensures the saturation with the vapor at the prescribed temperature. The chips also screen off liquid drops of the potassium which might come out of the surface.

The tin-lead bath is heated by a sheathed electric heater immersed in it. The temperature of the bath can be kept between 250°C and 450°C, the corresponding saturation pressure of the potassium vapor being between 0.1 and 10 Torr. The temperature is measured with a chromel-alumel thermocouple and is controlled automatically to be constant. Initially, pure tin bath was used. The liquid tin severely corroded the Inconel sheath of the heater in the bath. Then pure lead was tested, but no improvement was found. The higher melting point of lead gave additional difficulties in operating at low temperatures. Then, a mixture of tin and lead was used. The corrosion of the sheath of the heater still took place. We decided to allow a moderate rate of corrosion. The corroded heater and the bath are periodically replaced by new ones. Because of its low melting point, a mixture of tin and lead is used throughout the experimental work.

Figure 2 illustrates the test chamber in detail. The mixture of argon and potassium is led into the test chamber through tantalum tubing, where the gas is heated up to the desired temperature. The test chamber is composed of a thoria tube of 23 mm-i.d. with electrodes at both ends. The electrodes are made of two parallel molybdenum plates 40 mm apart. In the test chamber ,two parallel screens of tungsten wires of 0.3 mm-diam. are inserted normal to the current at a distance of 20 mm. They are electrostatic probes. The whole test chamber is enveloped by a large tantalum container of 43 mm-i.d. The space between the test chamber and the outer container is filled with pure argon for electrical insulation. The argon-potassium mixture in the test chamber and the pure argon around

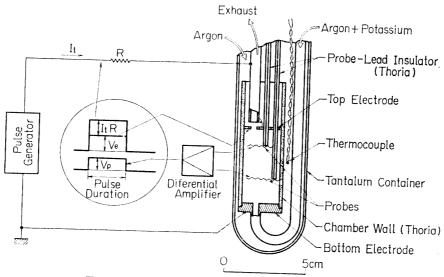


Fig. 2. Test chamber and electrical wiring.

the test chamber are gradually replaced by new gases. This replacement eliminates effects of absorption and evaporation of potassium to and from the walls of the test chamber and tubings. The mixture and the pure argon are exhausted through a common tube, so that the pressure in and around the test chamber is maintained exactly the same. In this way the leak of gas from or into the test chamber is prevented. The gases are vented out to the atmosphere through a potassium trap and a water-flush.

The test chamber and tubings are first evacuated to a pressure less than 10<sup>-3</sup> Torr. They are filled with pure argon at the atmospheric pressure and then evacuated again. This procedure is repeated a few times. The pure argon is kept flowing from that time. The temperature of the test chamber is gradually raised. Finally the temperature of the potassium boiler is raised and the whole system is ready for measurements.

The test chamber is heated in a vacuum electric furnace. In order to protect the tantalum container from any damage due to the residual gases, the furnace was kept at a pressure below 10<sup>-4</sup> Torr. A 150 mm-diam. oil diffusion pump, a 50 mm-diam. oil ejector pump, and an oil rotary pump with capacity of 300 t/min are used in series for the evacuation. The furnace uses a hollow cylinder graphite heater of 50 mm-i.d. The highest temperature of the heater is 2000°C at 20 kw-power input. The temperature of the furnace is uniform over the length of 8 cm. The test chamber and the outer container was placed in the uniform-temperature region. The gas in the test chamber, the electrodes, and the wall of the test chamber are at the same temperature. The temperature at the test chamber is measured by tungsten-tungsten rhenium thermocouples. It is found that the temperature difference between the bottom and the middle of the thoria wall of the test chamber is less than 25°C.

The gas in the test chamber becomes electrically conductive due to the thermal ionization of the potassium atoms. An electric current with a short duration is supplied to the plasma through the two disk electrodes. The instantaneous values of the electric potential at several points in and around the test chamber are recorded on an oscillogram as shown in Fig. 2. From these oscillograms the following quantities are known: (1) current  $I_t$ , (2) electrode voltage  $V_e$ , and (3) voltage difference between the two electrostatic probes,  $V_p$ . Two probes are operated at the floating condition, and only the difference of the potential between two probes is measured, so that effects of sheaths on two probes cancel out with each other. A probe together with the sheath around it blocks at most several percent of the cross sectional area of the plasma. The effect of this blocking on the overall characteristics of the plasma should be small.

The voltage between probes or electrodes may include thermoelectric voltage in the plasma. Although temperatures of the plasma and electrodes are quite uniform, it might be impossible to make the thermoelectric voltage less than 10  $\mu$ V, which amounts to a few percent of the smallest value of measured  $V_p$ . The cancellation of the thermoelectric voltage in the plasma was accomplished by measuring value of [(voltage with the pulse on) minus (voltage with the pulse off)]

instead of voltage with the pulse on itself. By this method of measurement, thermoelectric voltage in the wiring is also cancelled out.

There remains two possible sources of error. One is the change in the gas temperature due to the Joule heat. To minimize this effect, a pulsed current was used by Zukoski et al. [5]. We also use the pulse current. The duration of a pulse is  $200~\mu s$ . A pulse is triggered manually and the duty ratio is very small. A maximum temperature rise during a pulse can be estimated by the ratio (energy input)/(heat capactiy) for a unit volume of the plasma. Based on experimental data, it will be shown later (§ 3) that the temperature rise is very small. At a very small current, heating is negligible even with a continuous current. A few results of measurements with small continuous current are included in the experimental data.

The other point is concerned with the leak current through walls. At high temperature paractically any solid material does not remain to be a good insulator. In the present arrangements, the thoria wall of the test chamber is at the same temperature with the plasma, and the leak current through the wall can not be ignored. The current  $I_t$  in Fig. 2 is a sum of the leak current through the thoria wall,  $I_w$ , and the current through the plasma, I; that is,  $I_t = I_w + I$ . The value of  $I_w$  at an operating temperature was measured as a function of  $V_e$ , with pure argon in the test chamber. The results are illustrated in Fig. 3. It is worth noting that the value of  $I_w$  is not proportional to  $V_e$ . The data presented in this paper have been corrected for  $I_w$  by using the experimentally determined values of  $I_w$ .

The current pulse is supplied from a circuit shown in Fig. 4. The circuit is composed of following parts; (1) a condenser which discharges through the plasma, (2) a mercury switch to turn on the current manually, (3) a silicon-controlled-rectifier which terminates the current by short-circuitting the condenser, and (4) transistors which cut off the residual current after the pulse (such residual current could disturb measurements immediately after a pulse). The output resistance of the circuit ranges from 13.5 ohms at the highest current to 1 Meg-ohm at the lowest current. Most of the source voltage is consumed

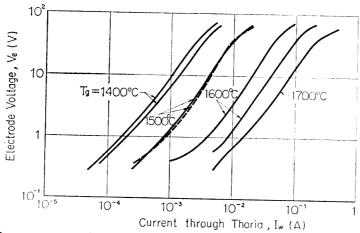


Fig. 3. Voltage-current relation of the thoria wall. Measurement with dc current.

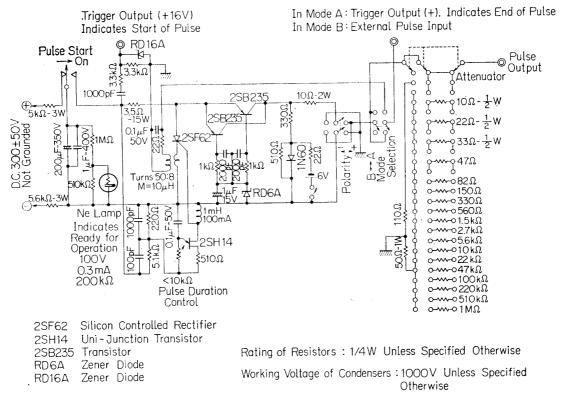


Fig. 4. Pulse current supply. The mode A is the normal mode of operation. In the mode B, only the attenuater is availed as a simple current-level regulator, with an external pulse supply.

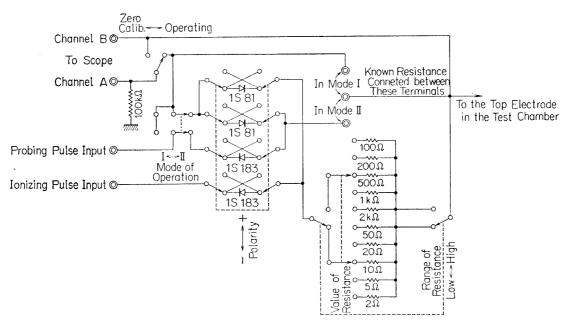


Fig. 5. Double range pulse measuring system. In the mode, I, the circuit is equivalent to a single resister. The mode II is the normal mode of operation.

in the attenuater and the current is kept constant regardless to changes in the plasma conditions during a pulse.

Attempts were made to inquire into the plasma decaying after a pulse. The plasma is brought to highly ionized state by a current pulse (main pulse) of a large amplitude and of  $200~\mu s$ . After the main pulse is turned off, electric properties of the plasma is measured with repeated pulses of a small amplitude and of very short duration (auxiliary pulses). The magnitudes of main pulse and auxiliary pulse are about 10~A and 1~mA, respectively. In order to display these two pulses of quite different magnitude in one common oscillogram, the circuit shown in Fig. 5 is used. Its essential function is to change the value of the resistance R in Fig. 2 according to whether the pulse is the main or the auxiliary. A main pulse is passed through a low resistance, and auxiliary pulses are passed through a high resistance, resulting in voltages of the same order of magnitude to be displayed on one oscillogram. With such circuit some preliminary results have been obtained. Unfortunately, there remain some difficulties and questions in results, so in this paper results on decaying plasma are not presented.

# 3. Experimental Results

Fig. 6 shows typical features of oscillograms. Traces represent the voltage  $(V_e + RI_t)$ , the electrode voltage  $V_e$ , and the probe voltage  $V_p$ , as stated in § 2. Thus the difference between the first two divided by R gives the value of the total current,  $I_t$ , which after correction for the leak current,  $I_w$  gives the current through plasma, I.

In Figs. 6(a) and 6(c) the duration of the current is  $200 \,\mu s$ . Traces in (a) show no change during the pulse period. On the other hand, in (c) a timewise change during the pulse period is remarkable. Only  $I_t$ , remains nearly constant during the pulse. The nature of the timewise change in  $V_e$  and  $V_p$  depends on the current, I. When I is small,  $V_e$  and  $V_p$  do not change during the pulse as shown in figure (a). When I is large,  $V_e$  and  $V_p$  change with a relaxation as shown in figure (c). They change rapidly at first then slowly and finally level off. When I is of an intermediate value,  $V_e$  and  $V_p$  appear to be constant as in (a); but an oscillogram with a faster sweep reveals that  $V_p$  changes as in figure (b). What is meant by those timewise

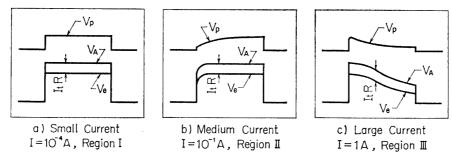


Fig. 6. Schematic voltage oscillograms. Argon-potassium. Confer Fig. 2 as for traces  $V_p$ ,  $V_A$  etc.

changes will be discussed later. Here, we plot the finall values of  $V_e$  and  $V_p$  as functions of I as shown in Figs.  $7 \sim 14$ .

Figs.  $7 \sim 12$  illustrate results for argon seeded with 0.013 and 0.13% potassium at three different temperature, 1400, 1500, and 1600°C. Those data have been corrected for the wall leak current,  $I_w$  using experimentally determined  $I_w - V_e$  curves (§ 2). In the case of potassium-seeded argon, the amount of the correction is less than 10% of the apparent value. When the test chamber is filled with argon-potassium mixture, due to the absorption of potassium on the wall the  $I_w - V_e$  relation may be different from that in pure argon. But moderate change in  $I_w$  which is usually less than 10% of I does not affect the value of I too much. Effects of the leak current through the thoria tubes around the probe-leads (Fig. 2) are estimated from the apparent resistance between two probes. It is found that the error in the measured value of electric field in the plasma caused by the leak is less than several percent. In the figures, data of  $V_p$  in different runs are marked with different symbols. Data on  $V_e$  show less run-to-run scatter.

The temperature rise of a plasma during a pulse is estimated from experimental values of I and  $V_p$ . The amount of energy given to the bulk of plasma by one pulse is at most  $0.001 \, J/\text{cm}^3$ . This leads to the change in the gas temperature of  $7^{\circ}\text{C}$ , even if whole Joule heat is used for heating the atomic species of the plasma. Thus the change in the gas temperature can be neglected.

In Figs. 13 and 14 illustrated are results for helium seeded with 0.13% potassium at temperatures of 1400 and 1500°C. Those data also have been corrected for  $I_w$ . Owing to the low conductivity of helium-potassium plasma, the value of  $I_w$ , in some

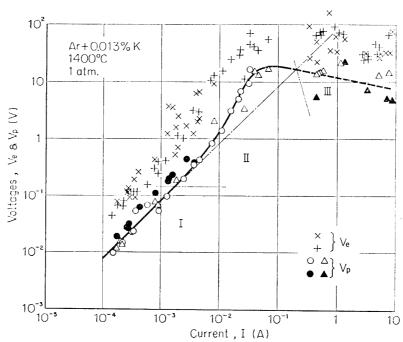


Fig. 7. Voltage-current relation of argon-potassium plasma. I.  $T_g = 1400 \,^{\circ}\text{C}$ . Potassium concentration 0.013%.  $V_e$ ; voltage between electrodes (40 mm apart), and  $V_p$ ; voltage between probes (20 mm apart).

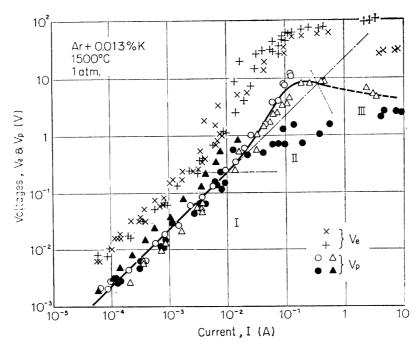


Fig. 8. Voltage-current relation of argon-potassium plasma. II  $T_q = 1500$ °C. Potassium concentration 0.013%.  $V_e$ ; voltage between electrodes (40 mm apart), and  $V_q$ ; voltage between probes (20 mm apart).

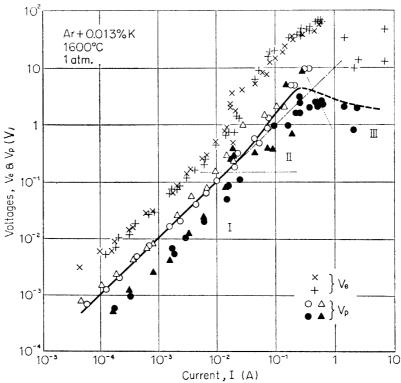


Fig. 9. Voltage-current relation of argon-potasium plasma. III.  $T_g=1600^{\circ}\text{C}$ . Potassium concentration 0.013%.  $V_e$ ; voltage between electrodes (40 mm apart), and  $V_p$ ; voltage between probes (20 mm aper).

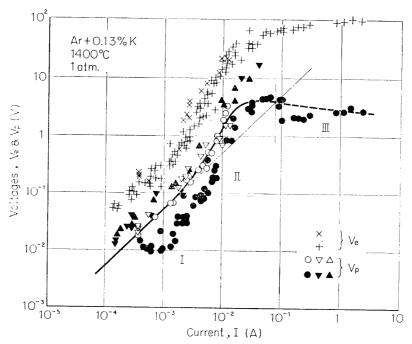


Fig. 10. Voltage-current relation of argon-potassium plasma. IV.  $T_g=1400^{\circ}\text{C}$ . Potassium concentration 0.13%.  $V_e$ ; voltage between electrodes (40 mm apart), and  $V_p$ ; voltage between probes (20 mm apart).

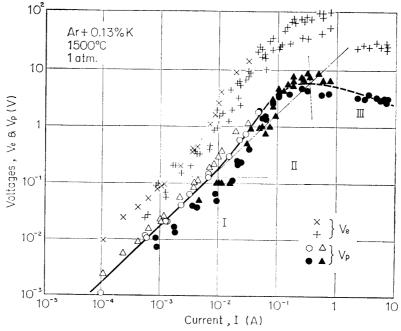


Fig. 11. Voltage-current relation of argon-potassium plasma. V.  $T_g=1500^{\circ}\mathrm{C}$ . Potassium concentration 0.13%.  $V_e$ ; voltage between electrodes (40 mm apart), and  $V_p$ ; voltage between probes (20 mm apart).

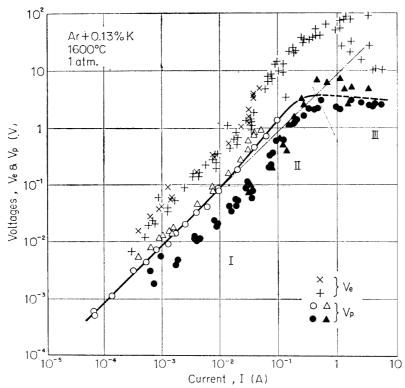


Fig. 12. Voltage-current relation of argon-potassium plasma. VI.  $T_g = 1600^{\circ}\text{C}$ . Potassium concentration 0.13%.  $V_e$ ; voltage between electrodes (40 mm apart), and  $V_p$ ; voltage between probes (20 mm apart).

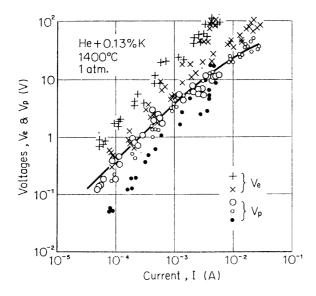


Fig. 13. Voltage-current relation of helium-potassium plasma. I.  $T_g=1400^{\circ}\text{C}$ . Potassium concentration 0.13%.  $V_e$ ; voltage between electrodes (40 mm apart), and  $V_p$ ; voltage between probes (20 mm apart).

cases, is as large as the current through the plasma, I. Since  $I = I_t - I_w$ , if  $I_w$  is of the same order of magnitude as I, then the relative error in I caused by an error or change in the  $I_w - V_e$  relation is nearly the same as the relative error in  $I_w$ . The reliability of the results is low. Here presented are only two cases of a relatively high reliability, although experiments were made for many other cases. The larger symbols in Figs. 13 and 14 denote data of higher reliability among those in the same

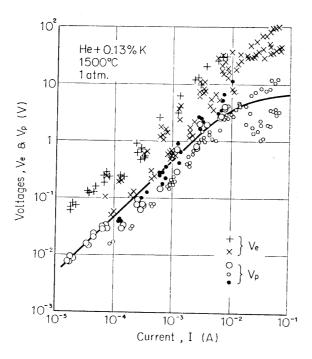


Fig. 14. Voltage-current relation of helium-potassium plasma. II.  $T_g$ =1500°C. Potassium concentration 0.13%.  $V_e$ ; voltage between electrodes (40 mm apart), and  $V_p$ ; voltage between probes (20 mm apart).

figure. The reason for the higher reliability is that for those runs the correction could be made more precisely. For an argon-potassium plasma the amount of correction is small, and the reliability is high for all data.

Hereafter discussions will be made on the prove voltage,  $V_p$ , or equivalently, the electric field in a plasma as a function of current, I, or current density, i. Other subjects e.g., the sheath voltage will be discussed later. Although there is some run-to-run scatter in the absolute values of  $V_p$ , the functional dependence of  $V_p$  on I is reproduced rather well in different runs, as is seen from figures. The result shows several interesting features.

For an argon-potassium plasma in Figs.  $7 \sim 12$ , the  $V_p - I$  relation may be divided into three regions. At small current,  $V_p$  is proportional to I, in other words, the ratio  $V_p/I$  is constant. This region is hereafter referred to as "region I". In this region, oscillograms of type of Fig. 6(a) are obtained. When I is increased beyond some critical value,  $V_p$  increases rapidly so that the ratio  $V_p/I$  becomes larger than that in region I. This second region is named "region II". In region II, oscillograms of  $V_p$  as shown in Fig. 6(b) are obtained. Inspecting data in Figs.  $7 \sim 12$ , one finds that the value of I at the boundary between regions I and II, is not constant but depends on the temperature and the composition of the gas. On the other hand, the value of  $V_p$  at the boundary is nearly constant. The value is 0.1 to 0.2V, and the corresponding value of the electric field is 0.05 to 0.1 V/cm. When I is increased further,  $V_p$  increases slowly, becomes constant, and then decreases, so that the ratio  $V_p/I$  becomes smaller than that in region I. This third region with smaller  $V_p/I$  is called "region III". In region III oscillograms as shown in Fig. 6(c) are obtained. The value of I at the boundary between regions II and III, i.e. the value of I at which the value of the ratio  $V_p/I$  is equal to that in region I, is not

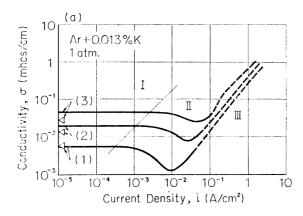
constant. The value of  $V_p$  at the boundary is not constant, but is dependent on the gas temperature and the gas composition, and ranges from 3 to 20 V.

The fact that  $V_e$  does not increase with I makes it impossible to maintain a constant current with a constant-voltage power supply. This is the reason why we use the constant-current pulse generator as shown in Fig. 4. Even with a constant-current power supply, it is difficult to maintain a stable, uniform distribution of the current in a plasma in region III, because  $V_p$  does not increase with I. The instability of the current distribution is suggested by oscillographic traces of  $V_e$  and  $V_p$ , which often show sudden jumps or irregular flactuations during a pulse.

In each of Figs.  $7 \sim 14$  a faired curve for  $V_p - I$  relation is shown. In constructing the curve the following points are considered; (1) the curve should reproduce the functional dependence of  $V_p$  on I for each run, and (2) data of a run with less scatter are weighted more.

For a helium-pottassium plasma, it is not clear whether "region II" exists or not. In Figs. 13 and 14, the value of  $V_p/I$  is constant at small value of I, and then decreases with increase in I. It does not become larger than that in region I, as far as the data in Figs. 13 and 14 are concerned. However, owing to large correction for  $I_m$ , data are less reliable, and no definite conclusions are possible.

From data on  $V_p$  versus I, the electrical conductivity of a plasma,  $\sigma$ , is calculated as a function of the current density i. Figure 15 presents the results for an argon-potassium plasma, based on the faired curves in Figs.  $7 \sim 12$ . In the  $\sigma$ -i relation the three regions appear as follows. When the current density, i is small,  $\sigma$  is inde-



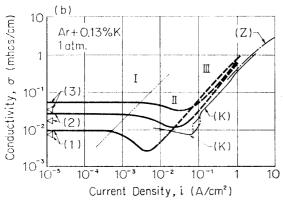
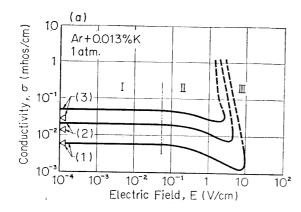


Fig. 15. Conductivity versus current density.

- (a) argon + 0.13% potassium,
- (b) argon+0.13% potassium;
- (1)  $T_g = 1400$ °C, (2)  $T_g = 1500$ °C,
- (3)  $T_a = 1600$  °C;
- (K) argon +0.16% potassium,  $T_q = 1200 \pm 30$ °C, by Kerrebrock et al. (Ref. 4),
- (Z) argon+0.6% potassium,  $T_g$ = 1730°C, by Zukoski et al. (Refs 5 and 6).

Tiangles at the ordinate show calculated values.



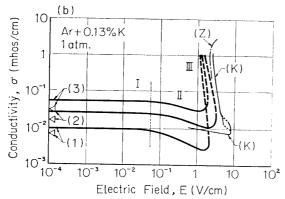
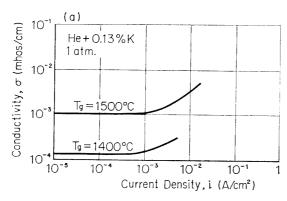


Fig. 16. Conductivity versus electric field.

- (a) argon + 0.013% potassium,
- (b) argon+0.13% potassium;
  - (1)  $T_g = 1400$ °C, (2)  $T_g$ ,=1500°C;
  - (3)  $T_g = 1600$  °C;
- (K) argon+0.16% potassium,  $T_g$ = 1200±3°C, by Kerrebrock et al. (Ref. 4),
- (Z) argon+0.6% potassium,  $T_q$ = 1730°C, by Zukoski et al. (Refs. 5 and 6).

Triangles at the ordinate show calculated values.



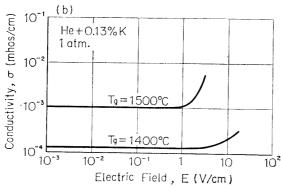
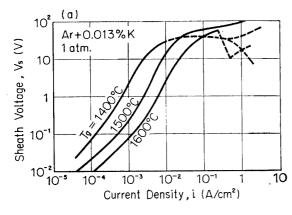


Fig. 17. Conductivity of helium-potassium plasma.

- (a) conductivity versus current density
- (b) conductivity versus electric field;

Potassium concentration 0.13%.



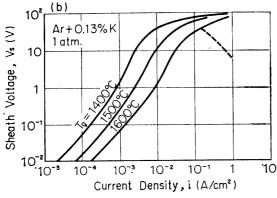


Fig. 18. Sheath voltage versus current density. Sum of the voltages across two sheaths on the two electrodes,  $V_s$  is calculated by  $V_s = V_e - 2.0V_p$ .

- (a) argon + 0.013% potassium,
- (b) argon+0.13% potassium.

pendent of i. This is the region I, and the value of  $\sigma$  in region I is denoted by  $\sigma_I$ . The value of  $\sigma_I$  is a function of the gas temperature,  $T_g$  and the potassium concentration. At somewhat larger value of i,  $\sigma$  becomes smaller than  $\sigma_I$ . This is the region II. The value of  $\sigma$  in this region is dependent on i,  $T_g$ , and the potassium concentration. At large current density  $\sigma$  becomes larger than  $\sigma_I$ . This is the region III. In this region,  $\sigma$  is dependent on i and the potassium concentration, but is weakly dependent on  $T_g$ .

In Fig. 16,  $\sigma$  is presented as a function of the electric field, E. Again three regions appear, in a similar manner as  $\sigma - i$  curve.

Fig. 17 illustrates the conductivity of a helium-potassium plasma as a function of the current density, i (Fig. (a)) and of the electric field (Fig. (b)). These data are deduced from faired curves in Figs. 13 and 14. The conductivity of a helium-potassium plasma is much lower than that of an argon-potassium plasma at the same temperature and current density.

It is well known that in a plasma adjacent to an electrode there is a thin sheath. Across the sheath usually appears a voltage drop which is called the sheath voltage. In the present experiment, the sum of sheath voltages on the two main electrodes,  $V_s$ , is obtaind by a relation  $V_s = V_e - (D_e/D_p)V_p$ . Here  $D_e$  denotes the distance between two main electrodes (=40 mm), and  $D_p$  is the distance between two probes (=20 mm). Hence  $(D_e/D_p)=2$ . The sheath voltage is obtained from experimental data as a function of I or i.

Faired curves of  $V_s - i$  relation are illustrated in Fig. 18. It is seen that  $V_s$  does

not vanish at small *i*. The value of  $V_s$  at small *i* is proportional to *i*. This fact may be seen also directly from  $V_e-I$  and  $V_p-I$  diagrams in Figs. 7-12. For instance, in Fig. 8 at  $I=10^{-4}A$ , the value of  $V_e$  is 4 to 5 times  $V_p$ , so that  $V_s$  is 1 to 1.5 times  $V_p$ . This example shows that the sheath voltage can be as large as the voltage across the bulk of plasma under some of the present experimental conditions.

# 4. Equilibrium Conductivity

In the present experiment, the temperature is uniform over the plasma and the test chamber. The wall of the test chamber is opaque to the radiation. There is no flux of energy between the plasma and the wall. The plasma is in a thermal equilibrium. When there is a small electric current, the plasma is still in equilibrium. In region I, the conductivity does not depend on the current density. The conductivity in region I should be the same as in the complete thermal equilibrium, i=0. Oscillographic observations indicate no change in  $\sigma$  due to the current. The conductivity in region I,  $\sigma_I$ , is to be compared with the value calculated by the equilibrium theory.

The fraction of ionization of atoms in a thermally equilibrium state is given by the Saha equation [10, 11] as,

$$\frac{n_e n_i}{n_0} = \frac{2g_i (2\pi m_e kT)^{3/2}}{g_0 h^3} \exp\left(-\frac{e\phi}{kT}\right).$$

Here  $n_e$ ,  $n_i$ , and  $n_0$  are number densities of electrons, ions, and neutral atoms, respectively. Factors  $g_i$  and  $g_0$  are statistical weights of ionized and neutral states of an atom. Other symbols are as follows;  $m_e$  and e denote mass and electric charge of an electron, respectively,  $\phi$  is the ionization potential of an atom, T denotes temperature, h is the Planck constant, and k denotes the Boltzmann constant. Since a plasma is electrically neutral, and usually, doubly charged ions are few, we have  $n_i = n_e$ . Then, knowing the relevant physical quantities of the atom and the temperature, one is able to determine the ratio,  $n_e^2/n_0$ . Futher, the knowledge of either of  $n_0$ ,  $n_0 + n_e$ ,  $n_e + n_0 + n_i$ , or the corresponding partial pressure enables us to determine values of  $n_e$ ,  $n_i$ , and  $n_0$ .

In the temperature range of the present experiment, i.e., up to 2000 K, only the potassium atoms are ionized significantly. For an alkali atom, the ratio  $g_i/g_0$  is equal to 1/2. The potential for double ionization is very high, i.e., 32 V, from the singly ionized state. This value should be compared with the ionization potential of 4.34 V for single ionization. Thus, the number of doubly charged ions is negligibly small. The ionization fraction of argon is extremely small.

The electrical conductivity,  $\sigma$  is given by  $\sigma = n_e e \mu$ , where  $\mu$  is the mobility of an electron in the gas. The contribution of ions in  $\sigma$  is neglected because the mobility of an ion is small. The value of  $\mu$  is calculated by the Allis formula [12].

$$\mu = \frac{4\pi e}{3m_e} \int_0^\infty F(v) \frac{d}{dv} \left( \frac{v^3}{\nu_t(v)} \right) dv,$$

where v is the speed of an electron, and  $\nu_t(v)$  is the total frequency of the diffusive momentum transfer of an electron with all atoms, ions and molecules. The function F(v) denotes the spherically symmetrical term in the expansion of the distribution function of the velocity of electrons. In region I, the distribution of electrons is Maxwellian, and F(v) is given by Ref. 13 as

$$F(v) = \left(\frac{m_e}{2\pi kT}\right)^{3/2} \exp\left(-\frac{m_e v^2}{2kT}\right).$$

The value of  $\nu_t(v)$  is calculated by (Ref. 11),

$$\nu_t(v) = v \sum_j n_j Q_j(v)$$
,

where  $n_j$  is the number density of the *j*-th species, and  $Q_j(v)$  is the diffusive momentum-transfer cross section of *j*-th species with an electron of speed v. Quantities F(v),  $\nu_t(v)$ , and  $Q_j(v)$  are written as  $F(\varepsilon)$ ,  $\nu_t(\varepsilon)$  and  $Q_j(\varepsilon)$  when they are given as functions of the electron energy  $\varepsilon = (1/2)m_ev^2$ . In the present experiment, the plasma is composed of argon atoms (indicated by a subscript "Ar"), potassium atoms (indicated by "K"), singly charged ions of potassium (indicated by "i"), and electrons (indicated by "e"). Cross sections of argon, potassium, and helium atoms for an elastic collision with an electron in references  $14 \sim 29$  are illustrated in Figs.  $19 \sim 21$ . Both the total scattering and the diffusive momentum-transfer cross sections are shown. The total scattering cross section are usually obtained from electron beam experiments and the diffusive momentum-transfer cross sections are obtained from electron swarm experiments.

In Fig. 19 a profound minimum is seen in the cross section of an argon atom. It

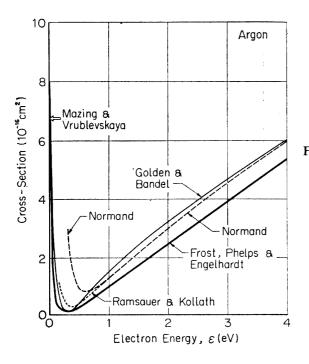


Fig. 19. Elastic collision cross section of an argon atom.

Diffusive electron momentum transfer cross section.

Frost, Phelps, and Engelhardt

(Refs 14 and 15)

Marzing and Vrublevskaya

(Ref. 16)

Total scattering cross section

Ramsauer and Kollath (Ref. 17)

Normand (Ref. 18)

Golden and Bandel (Ref. 19)

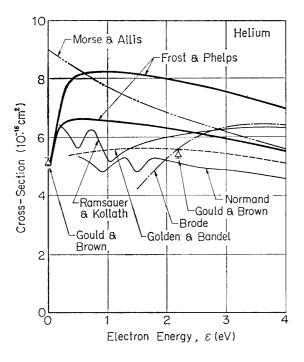


Fig. 20. Elastic collision cross section of a helium atom.

Diffusive electron momentum transfer cross section.

Frost and Phelps (Ref. 15)

Total scattering cross section

Brode (Ref. 20)

Morse and Allis; theory (Ref 21)

Ramsauer and Kollath (Ref 22)

Normand (Ref. 18)

Golden and Bandel (Ref. 23)

Averaged cross section for electrons of distributed energy

Gould and Brown (Ref. 24)

is due to the well-known Ramsauer effect. Of the several data on the cross section, we choose the data by Frost, Phelps and Engelhardt [14, 15] for the calculation of the conductivity. The reason for this choice is that only their values are the diffusive momentum-transfer cross section as a function of  $\varepsilon$ . Besides, they were obtained through an elaborate experiment and analysis. They do dot contradict with the latest results for the total scattering cross section by Golden and Bandel

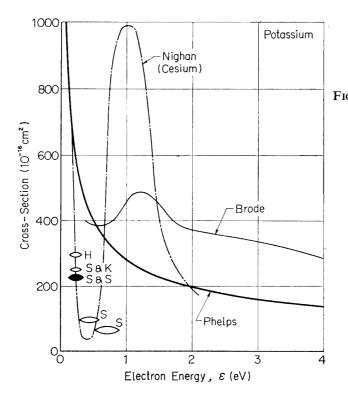


Fig. 21. Elastic collison cross section of a potassium atom. Diffusive electron momentum transfer cross section Phelps (Ref. 25) Total acattering cross section Brode (Ref. 26) Averaged cross section for electrons of distributed energy H; Harris (Ref. 2) S; Shioda (Ref. 27) S & K; Solbes and Kerrebrock (Ref. 28) S & S; Present result Diffusive electron momentum transfer cross section of a cesium Nighan (Ref. 29).

[19]. The cross section of a helium atom is almost constant in the range of electron energy from 0 to 4eV (Fig. 20).

Data on the cross section of a potassium atom are few. In Fig. 21, continuous lines are results by Brode [26] and Phelps [25] as functions of  $\varepsilon$ . Data by Harris [2], Shioda [27], and Solbes and Kerrebrock [28] are averaged cross sections for electrons with Maxwellian energy distribution at  $T_e$ . For those data  $\varepsilon$  is taken as  $(3/2)kT_e$  (mean thermal energy of an electron). In the present calculation of the conductivity a simple expression by Phelps [25] is used, which is

$$\nu_{e-K}(v) = vQ(v)n_K = 1.6 \times 10^{-6}n_K(\text{sec}^{-1})$$
  
 $(n_K; \text{ number density of potassium atoms in cm}^{-3}).$ 

This is the only data for  $Q_K(v)$  as a function of v at low electron energy as far as known to the authors.

Collisions between electrons and ions are quite different from the electron-atom collison. The Coulomb interaction is effective up to a very large distance, and the total scattering cross-section would be infinitely large. A method of connecting the ion-electron collisions with the electron-atom collision is not established. There are two schemes for this. One is "mean-free-path mixing rule". In this scheme the conductivity  $\sigma$  is put  $\sigma^{-1} = \sigma_a^{-1} + \sigma_i^{-1}$ , where  $\sigma_a$  is conductivity in the absence of electronion collisions, and  $\sigma_i$  is that in the presence of electronion collision only. The other is "integral mixing rule" to which we follow here. In this scheme electronion collisions are included in  $\nu_t(\varepsilon)$ , by suitably chosing value of  $\nu_{e-t}(\varepsilon)$ . This scheme gives better results when "mean free path" depends strongly on the electron energy as shown in references  $30 \sim 32$ . The cross section of an ion to be used in this scheme is given by Frost [11] as,

$$\nu_{e-i}(v) = vQ_i(v)n_i = 0.476K/(m_e v^2/(2kT)),$$

in which

$$K = \left[ \frac{2n_e e^2 \ln \Lambda}{(4\pi\epsilon_0)^2} \right] \left( \frac{2e}{m_e} \right)^{1/2} \left( \frac{e}{kT} \right)^{3/2}$$

$$\Lambda = \frac{3}{(2\pi n_e)^{1/2}} \left( \frac{4\pi\epsilon_0 kT}{e^2} \right)^{3/2},$$

and  $\varepsilon_0 = 8.854 \times 10^{-12} \text{Farad/} m$  (in mksa units). In reference 33, effect of the electron-electron interaction on the conductivity was shown. The effect has been included in the above expression for  $\nu_{e-i}$  by Frost.

The value of  $\sigma$  calculated by the above scheme is shown in Figs. 15 and 16 by open triangles at the ordinates. Measured values are higher than the calculated. Although there is a scatter in experimental data, the discrepancy is more than just experimental errors. The saha equation might be valid. The temperature of the plasma was measured within few degrees. The value of the ionization potential is well-established. Only possible cause is the value of the cross section.

The value of  $Q_{Ar}(\varepsilon)$  would be of high reliability. Besides, it is the lowest of all

known so far (Fig. 19). It might give the highest value of  $\sigma$ , i.e., the nearest to the experimental. The value of  $Q_i(\varepsilon)$  is not so accurate, but the frequency of electronion collisions is less than one tenth of other collision frequencies. Therefore, the contribution of  $Q_i(\varepsilon)$  in the calculated value of  $\sigma$  is small. Even if the value of  $Q_i(\varepsilon)$  is put to be zero, the calculated value of  $\sigma$  increases only by 10%. On the other hand, the value of  $Q_K(\varepsilon)$  used in the calcutation has been derived from results of experiments in the positive colum and may be correct only for electrons with moderate energy. Under the present experimental conditions, most of electrons have energy less than 1eV, that is,  $F(\varepsilon)$  has appreciable value only for  $\varepsilon < 1eV$ . The integrand in the Allis formula is large where  $\nu_t$  is small there. The value of  $\nu_t(\varepsilon)$  is determined mainly by the maximum one of  $n_{Ar}Q_{Ar}$ ,  $n_KQ_K$ , and  $n_iQ_i$ . Since  $Q_{Ar}(\varepsilon)$  has a profound minimum at  $\varepsilon = 0.3eV$ , the value of  $Q_K(\varepsilon)$  near  $\varepsilon = 0.3eV$  is important in determining the mobility in an argon-potassium mixture. It is probable that the used value of  $Q_K(\varepsilon)$  might be too large for low energy.

The value of  $Q_K(\varepsilon)$  affects also the functional dependence of  $\sigma$  on the potassium concentration,  $c_K$ . Fig. 22 illustrates the  $\sigma-c_K$  relation. Data for  $c_K=0.8\%$  are obtained with a small, continuous current through plasma instead of a pulse. Each culculated curve for a fixed temperature has a maximum. The reason for the maximum can be explained as follows. We consider a gas mixture of neutral mainbody particles (indicated by "m") and a seed (indicated by "s") to be ionized. Cross sections and the fractional concentrations of two gases are denoted by  $Q_m, Q_s$ , and  $c_m, c_s$ , respectively. For convenience, cross sections are assumed to be constant. If the ionization fraction is not too high,  $n_e$  is proportional to  $(c_s)^{1/2}$ , and the conductivity is proportional to  $(c_s)^{1/2}/(c_m Q_m + c_s Q_s)$  with neglection of electron-ion collisions. This has a maximum at  $c_s = Q_m/(Q_s - Q_m)$ . Actually, cross sections are not constant, but the qualitative conclusion remains unchanged. The larger cross

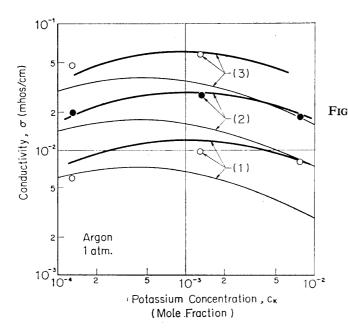


FIG. 22. Conductivity versus potassium concentration. Argon-potassium. (1)  $T_g = 1400^{\circ}\text{C}$ , (2)  $T_g = 1500^{\circ}\text{C}$ , (3)  $T_g = 1600^{\circ}\text{C}$ . Hair lines show the values calculated on the base of  $Q_K(\varepsilon)$  of Ref. 25. Full lines show the values based on assumed value,  $\overline{Q}_K = 2.3 \times 10^{-14} \, \text{cm}^2$ . Filled and open circles show the experimental values. Data for  $c_K = 0.8\%$  are obtained from measurement with small dc current.

section of the seed atom results in the smaller  $c_s$  for the maximum conductivity. Then, Fig. 22 suggests that the assumed value of the cross section of a potassium atom might be too large.

An average value of the cross section of a potassium atom may be obtained from the present results by making a best fit to measured values of  $\sigma$ . To do this, we substitute the cross section of an argon atom,  $Q_{A_T}(\varepsilon)$ , with a constant  $\overline{Q}_{A_T}$  which gives the same mobility as  $Q_{A_T}(\varepsilon)$ . The procedure of determing  $\overline{Q}_{A_T}$  is as follows. Integrating the Allis formula by part, one has,

$$\mu = \frac{4\pi e}{3m_e} \int_0^\infty \frac{v^3}{\nu_t(v)} \frac{d}{dv} F(v) dv.$$

In the present case, electrons are in a Maxwellian distribution at temperature,  $T_e$ . Introducing the expression for F(v) given by Ref. 13,

$$F(v) = \left(\frac{m_e}{2\pi kT}\right)^{3/2} \exp\left(-\frac{m_e v^2}{2kT}\right)$$

into the above formula and changing the independent variable from v to the normalized energy  $y = m_e v^2/(2kT_e)$ , one has an expression for the mobility,

$$\mu = \frac{4e}{3\sqrt{2\pi m_e k T_e} n_j} \int_0^\infty \frac{y e^{-y}}{Q_j(y)} dy$$

The value of  $\bar{Q}_{j}$  is obtained by putting

$$\mu = \frac{4e}{3\sqrt{2\pi m_e k T_e} n_j} \frac{1}{Q_j} \int_0^\infty y e^{-y} dy$$

in which the last integral is equal to 1. Thus, one has

$$\overline{Q}_{J} = \left( \int_{0}^{\infty} \frac{ye^{-y}}{Q_{J}(y)} dy \right)^{-1}$$

or,

$$\overline{Q}_{J} = \frac{4e}{3\sqrt{2\pi m_{e}kT_{e}\mu n_{J}}}.$$

Obviously,  $\overline{Q}_j$  is dependent on the functional form of F(v) and in the case of Maxwellian distribution  $\overline{Q}_j$  is a function of  $T_e$ . With  $Q_{Ar}(\varepsilon)$  by Frost et al., the value of  $\overline{Q}_{Ar}(T)$  for a temperature  $T=1500^{\circ}\mathrm{C}$  is determined to be  $2.3\times10^{-17}\,\mathrm{cm}^2$ . For ions, it is found,  $\overline{Q}_i=4\times10^{-12}\,\mathrm{cm}^2$ . Then, the value of  $\overline{Q}_K=2.3\times10^{-14}\,\mathrm{cm}^2$  gives the  $\sigma-c_K$  relation shown by the full lines in Fig. 22, which are in good agreement with the experimental results. Strictly speaking, the value of averaged cross section  $\overline{Q}_K$  can be used only for present experimental conditions; the composition of the working gas and the temperature. Nevertheless, the experimentally determined value of average cross section  $\overline{Q}_K$  gives a correct estimation for the

magnitude of  $Q_K(\varepsilon)$  for electron energy of a few tenth of 1eV. The value of  $\overline{Q}_K$  is illustrated in Fig. 21. It compares well with other experimental data on cross section.

Nighan [29] made an experiment for determining the cross section of a cesium atom with slow electrons. Since cesium and potassium atom are alike in the atomic structure, the cross section might be alike. His result also seem to support the present value on the effective cross section of a potassium atom.

# 5. Nonequilibrium Conductivity

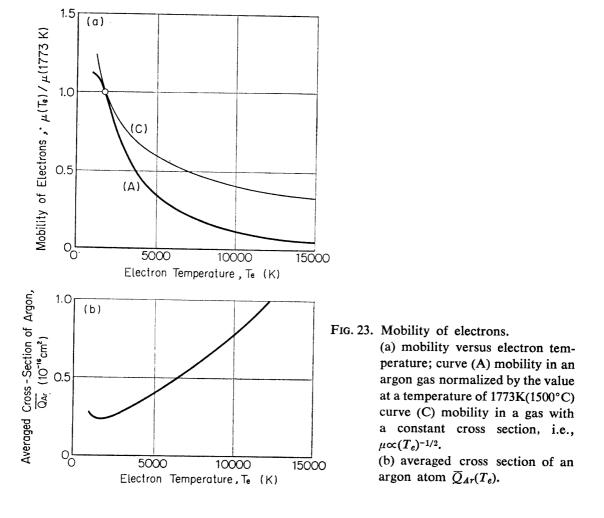
In region II the conductivity of the argon-potassium plasma is lower than that in region I. A similar result was obtained by Kerrebrock and Hoffman [3]. In their experiment on an argon-potassium plasma  $\sigma$  increased with the decrease in the current in a certain current range. Such a behavior is not expected from the conventional two-temperature conduction theory. There was no satisfactory explanation for the The equilibrium value of  $\sigma$  at small current was not measured in ther ex-Zukoski, Cool, and Gibson [5, 6] found a high value of  $\sigma$  at a periment [3]. small current. Later, by Cool and Zukoski [7] the cause of the high value was traced to the existence of a thin, poorly conducting film which buids up on the test section wall and shunts the voltage probes. (p. 758, Ref. 7). Kerrebrock and Dethlefsen [4] found that  $\sigma$  of an argon-potassium plasma decreases with the increase in the current density i, although the extent of the decrease in much less than that in The gas temperature in the experiment of Kerrebrock et al. is lower than that of Zukoski et al. Such a phenomenon may be absent at high gas temperature. Kerrebrock and Dethlefsen [4] pointed out the ambiguity in the number density of electrons in their experiment [3, 4]. However, the unexpected dependence of  $\sigma$ on i can not be explained by the number density of electrons.

In the present the dip in  $\sigma$  is clearly observed. Moreover, the conductivity data show smooth transition from region I to II, as the current is increased. As discussed in § 2, the leak current through the wall of the test chamber was measured and corrected for. Leak current between probes was also measured, and the amount was found to be small. The dip in  $\sigma$  can not be attributed to an experimental error.

The conductivity  $\sigma$  is a product,  $en_e\mu$ . A decrease in  $\sigma$  implies that  $\mu$  and/or  $n_e$  decrease. Since the current density in region II is higher than that in region I, more energy is given to electrons. The increased energy input to electrons means more heating of electrons. It is unlikely that heating of electrons results in the decrease in  $n_e$ . Accordingly, in region II the value of  $\mu$  must decrease.

In Fig. 19 the electron momentum-transfer cross section of an argon atom is shown. The cross section is minimum for an electron energy about  $\varepsilon = 0.4 eV$ , and rapidly increases as  $\varepsilon$  increases. The electrons moving in a gas under an electric field are not mono-energetic, but have distributed energies ranging from 0 to infinity. The mobility  $\mu$  is not determined by the cross section at a single value of  $\varepsilon$ , but is determined by an integral over the whole range of  $\varepsilon$ ; i.e., by the Allis formula.

In Fig. 23(a) the value of  $\mu$  in argon is illustrated as a function of the electron temperature,  $T_e$  for electrons with Maxwellian distribution. The cross section  $Q_{Ar}(\varepsilon)$  by Frost et al. (Fig. 19) has been used for calculating  $\mu$ . The averaged cross section  $\overline{Q}_{Ar}(T_e)$  determined from the value of  $\mu$  is shown in Fig. 23(b). The change in  $\mu(T_e)$  for constant value of the cross section is illustrated for comparison. This is also a decreasing function of  $T_e$  because of the relation,  $\mu \propto 1/(\overline{Q}T_e^{1/2})$ . This curve represents the dependence of  $\mu$  on  $T_e$  in a gas with an approximately constant cross section, like helium.



In Fig. 24,  $n_e$  is plotted against  $T_e$ . The value of  $n_e$  is calculated with the Saha equation at  $T_e$ . The increase in  $n_e$  is much more than the decrease in  $\mu$ , when  $T_e$  is elevated. The conductivity should not decrease if thermal equilibrium at  $T_e$  prevails, i.e., if the energy distribution for electrons is Maxwellian and the ionization fraction is the equilibrium value at  $T_e$ . In view of the experimental results showing the decrease in  $\sigma$ , there must be a deviation from equilibrium.

There are many processes in the ionization and recombination in a plasma. Important ones are;

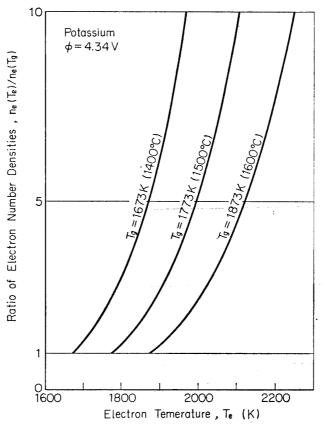


Fig. 24. Electron number density versus electron temperature, with the Saha ionization equilibrium at  $T_e$  assumed. Number density normalized by that at gas temperature.

1.	$e+M \rightleftharpoons e+e+M^+$	electronic collisional ionization (electronic three-body recombination)
2.	$M' + M \rightleftharpoons M' + e + M^+$	atomic collisional ionization (atomic three-body recombination)
3.	$M'+M \rightleftharpoons e + (M'M)^+$	associative ionization (dissociative recombination)
4.	$h\nu + M \rightleftharpoons e + M^+$	photo-ionization (radiative recombination)
5.	$M^{**} \rightleftharpoons e + M^+$	internal conversion ionization (di-electronic recombination)

Moreover, the "neutral atoms" to be ionized can be in excited states. There are not many electrons or other particles with sufficient energy for ionizing a ground state atom by a single collision. Thus the process of "ionization" is actually a multistep process of excitation, final step being the ionization. Populations of ionized and excited states in a plasma are determined by a balance among a large number of elementary processes. Rates of those processes depend on the distribution functions of electrons and atoms, which in turn depend on rates of elementary processes. Thus the problem is a very complicated one. If the whole system is

in thermal equilibrium, the distribution function of each species in the plasma is Maxwellian, and the temperature is common for all species. Moreover, each elementary process is balanced with a respective inverse process and populations in ionized and excited states are in the Saha-Boltzmann equilibrium. In region I of the present experiment the plasma is in such a state.

When a large current exists in a plasma, the Joule heat is given first to electron. If the rate of energy exchange is large among electrons themselves and among heavy particles themselves, and is small between electrons and heavy particles, there may be two temperatures in the plasma; the electron energy distribution is Maxwellian at an "electron temperature",  $T_e$  and the distribution of the translational energy of heavy particles is Maxwellian at a "gas temperature",  $T_g$ . In an electric field,  $T_e$  becomes higher than  $T_g$ . When  $n_e$  is sufficiently large, the ionization and the excitation are mainly governed by collisions with electrons, because they are very effective for ionizing and exciting atoms in comparison with collisional processes with heavy particles. In that case, the fraction is given by the Saha equation with  $T_e$ . Such a state is assumed in the conventional two-temperature conduction theory. Estimations show that the state mentioned above is realized if,  $n_e > 10^{14} \, \text{cm}^{-3}$  in an argon-potassium plasma at atmospheric pressure [34].

If  $n_e$  is low, radiative processes and collisional processes among atoms are significant. Hiramoto [35] calculated the ionization fraction of a helium-cesium plasma with consideration of six different processes of ionization and recombination. The energy distribution of the free electrons was assumed to be Maxwillian. The ionization of an atom was assumed to occur through *one* of those six processes. A combination of processes of different kinds, e.g., an excitation by a collision with an electron followed by an ionization by a collision with an atom, is not considered. His results show that  $n_e$  becomes lower than the equilibrium value at  $T_e$ . A similar procedure can be applied for the present plasma. Considerations to his results, with account to differences in gas temperature and composition in his calculation and in the present plasma, however, show that the dip in the conductivity can not exceed 10% at a most favorable estimation. In order to explain the large amount of decrease in  $\sigma$  in region II, the assumption of Maxwellian distribution of electrons should be re-examined, rather than introducing various ionization processes.

The validity of the assumption of Maxwellian energy distribution for free electrons may be examined by comparing the rate of energy exchange among electrons with that between electrons and other particles. In order to do this, we consider a reference state which is represented by;

```
number density of argon atoms, n_{Ar}=4\times10^{18}\,\mathrm{cm}^{-3} (corresponds to 1500°C, 1 atm.);
number density of potassium atoms, n_K=5\times10^{14}\sim6\times10^{15}\,\mathrm{cm}^{-3} (corresponds to potassium concentration 0.013\sim0.13\% in mole-fraction);
number density of electrons, n_e=8.5\times10^{10}\sim1.4\times10^{12}\,\mathrm{cm}^{-3} (equilibrium value at temperature of 1400\sim1600^{\circ}\mathrm{C} with the value of n_K above).
```

Of the collisional processes including electrons followings are to be considered;

electron-argon atom (e-Ar) elastic collisions,

electron-potassium atom (e-K) elastic collisions,

e-K inelastic collisions,

electron-ion (e-i) elastic collisions,

electron-electron (e-e) elastic collisions.

Inelastic collisions of electrons with argon atoms are neglected because the threshold energy is very high (11.6 eV) compared with the mean thermal energy of electrons. On the other hand, the threshold energy for the e-K inelastic collision is comparatively low (1.61 eV).

The e-e collisions tend to establish a Maxwellian distribution of electrons. Collisions between electrons and other particles tend to disturb the Maxwellian distribution if the mean energy of electrons differs from that of other particles. Among them, the e-K inelastic collisions are important. They are possible only for electrons with energy higher than the threshold value,  $1.61 \, \text{eV}$ . An electron loses most of the energy after an inelastic collision. Thus the inelastic collisions serve to decrease the number density of high-energy electrons and to increase low-energy electrons. Since the rates of those collisions are dependent on the electron energy,  $\varepsilon$ , they must be evaluated at different values of  $\varepsilon$ . We compare the rates for two groups of electrons, one of which is with relatively low energy, and the other with relatively high energy.

## Low-energy electrons

As the group of "low-energy electrons" we take electrons with energy less than 1.61 eV, which is the threshold energy for an inelastic collisions with a potassium atom in the ground state. Thus, for low-energy electrons inelastic collisions with a ground-state potassium atoms are absent. Since the number density of excited potassium atoms is low, inelastic collisions with the excited potassium atoms can be ignored. Rates of energy transfer by e-Ar, e-K, and e-i elastic collisions are compared with that of e-e collisions by estimating the following ratios for an electron;

```
(rate of energy exchange with argon atoms)/(rate of energy excange with other electrons)
```

$$= [n_{Ar}Q_{Ar}(\varepsilon)/(n_eQ_e(\varepsilon))]m_e\delta_{e-Ar}/m_{Ar},$$

(rate of energy exchange with potassium atoms)/(rate of energy exchange with other electrons)

```
= [n_{\kappa}Q_{\kappa}(\varepsilon)/(n_{e}Q_{e}(\varepsilon))]m_{e}\delta_{e-\kappa}/m_{\kappa},
```

(rate of energy exchange with ions)/(rate of energy exchange with other electrons)

```
= [n_i Q_i(\varepsilon)/(n_e Q_e(\varepsilon))] m_e \delta_{e-i}/m_i.
```

In these expressions  $m_e$ ,  $m_{Ar}$ ,  $m_K$ , and  $m_i$  are masses of an electron, an argon atom, a potassium atom, and an ion, respectively. Factors  $\delta_{e-Ar}$  etc are coefficients for

the energy transfer through an e-Ar collision etc. Values of  $\delta_{e-Ar}$  etc are dependent on the nature of the force acting between colliding particles.

As the value of  $Q_{Ar}(\varepsilon)$ , that of Frost et al. [14] is used. The cross section for e-e collisions,  $Q_e(\varepsilon)$  is assumed to be equal to Frost's value of  $Q_i(\varepsilon)$  [11] (see § 4) since e-e and e-i collisions are both due to Coulomb force. The fact that colliding electrons are faster than ions requires only minor modification in the evaluated value of  $Q_e(\varepsilon)$ , and for the purpose of an order-of-magnitude discussion the modification is not necessary. For the value of  $Q_K(\varepsilon)$  the averaged cross section of a potassium atom obtained in the present investigation (§ 4) is used. The value of  $\delta_{e-Ar}$  is nearly equal to 2, and values of  $\delta_{e-K}$  and  $\delta_{e-i}$  are taken to be equal to  $\delta_{e-Ar}$ . We put  $n_i = n_e$ , and  $m_i = m_K$ . Values of  $m_e \delta_{e-Ar}/m_{Ar}$ ,  $m_e \delta_{e-K}/m_K$ , and  $m_e \delta_{e-i}/m_i$  are nearly the same, and is equal to 1/40000 (the ratio,  $m_{Ar}/m_K$  is equal to 39.3/39.1, i.e., nearly 1).

For a typical value of energy  $\varepsilon = 0.5 \text{ eV}$ , the following results are obtained; Under all conditions of the reference state,

$$[n_{Ar}Q_{Ar}/(n_eQ_e)]m_e\delta_{e-Ar}/m_{Ar}<0.01$$
(largest for the smallest  $n_e$ )
$$[n_KQ_K/(n_eQ_e)]m_e\delta_{e-K}/m_K<0.004$$
(largest for the largest  $n_K$  and the lowest  $T_g$ )\*
$$[n_iQ_i/n_eQ_e)]m_e\delta_{e-i}/m_i<0.0005$$
(nearly constant under all the conditions)

Therefore, the distortion of electron energy distribution due to e-Ar, e-K, and e-i elastic collisons is negligible.

When an electric field is applied to a plasma, electrons are accelerated. This acceleration distorts the energy distribution. The amount of energy given to an electron from the field during a time interval between two successive e-e collisions,  $\Delta \varepsilon$  is, on the average, given by

$$\Delta \varepsilon = iE/(n_e \nu_{e-e}) = \sigma E^2/(n_e^2 \nu_e Q_e(\varepsilon)),$$

in which  $\nu_{e-e}$  is the frequency of the e-e collisions for an electron. If  $\Delta\varepsilon$  is small in comparison with the energy of the electron,  $\varepsilon$ , the distortion of the distribution function of the electron energy due to the field is small. In the above expression  $v_e$  is proportional to  $\varepsilon^{1/2}$  and  $Q_e(\varepsilon)$  is nearly proportional to  $\varepsilon^{-3/2}$  as shown in the expression for  $\nu_{e-i}$  (§ 4). Therefore,  $\Delta\varepsilon$  is proportional to  $\varepsilon$  and the ratio  $\Delta\varepsilon/\varepsilon$  is independent of  $\varepsilon$ . The ratio  $\Delta\varepsilon/\varepsilon$  is large at small  $n_e$ . For  $\varepsilon=0.5\,\mathrm{eV}$  and  $\sigma_I=0.06\,\mathrm{mho/cm}$  corresponding to the smallest value of  $n_e$ , one obtains  $\Delta\varepsilon/\varepsilon=0.8\,E^2(E\,\mathrm{in}\,V/\mathrm{cm})$ . Here  $\sigma_I$  is the conductivity in region I. At  $E=1\,V/\mathrm{cm}$ ,  $\sigma$  is 0.02 mho/cm, hence the value of  $\Delta\varepsilon/\varepsilon$  is about 0.25. Thus, it is concluded that the distortion of the distribution function due to the acceleration by a field is negligible up to  $E=1\,V/\mathrm{cm}$ . Although the electric field in region II could be higher than  $1\,V/\mathrm{cm}$ , the ratio

<sup>\*</sup> It should be remembered that  $n_e$  is proportional to  $\sqrt{n_K}$ .

 $\Delta \varepsilon/\varepsilon$  is less than 1 for most cases. Thus in regions I and II the energy distribution is Maxwellian for low-energy electron in spite of collisions with other particles and the acceleration by the electric field. We introduce an electron temperature  $T_{el}$  which characterizes the distribution of low-energy electrons.

It should be noted that the majority of electrons belong to the class of lowenergy electrons, and the mobility of electrons as a whole is determined exclusively by the mobility of low-energy electrons.

# High-energy electrons

Next, electrons with energy more than 1.61 eV are considered. Such electrons experience inelastic collisions with ground-state potassium atoms. The collisional processes to be considered are e-Ar, e-K, e-i elastic collisions and e-K inelastic collisions. The first three of them, along with the same line of argument as in the case of low-energy electrons, do not seriously distort the energy distribution of high-energy electrons. For a typical value of energy,  $\varepsilon=2$  eV, the following results are obtained;

$$(n_{Ar}Q_{Ar}/(n_eQ_e))m_e\delta_{e-Ar}/m_{Ar} < 0.7$$
  
 $(n_KQ_K/(n_eQ_e))m_e\delta_{e-K}/m_K < 0.03$   
 $(n_iQ_i/(n_eQ_e))m_e\delta_{e-i}/m_i < 0.0005$ 

The e-Ar collisions seem to be of some importance; however the e-K inelastic collisions are by far more important as shown in the following estimation.

The cross section of e-K inelastic collision,  $Q_{K,\rm inel}(\varepsilon)$  is zero for low-energy electrons and is of a finite value for electrons with energy more than 1.61 eV. The value of  $Q_{K,\rm inel}(\varepsilon)$  for  $\varepsilon > 1.61$  eV is not known. Therefore, the value of cross section by Witting and Gyftopoulos [36] for inelastic collisions with a *cesium* atom is used, and the cross section at energy  $\varepsilon = 2$  eV is taken as  $Q_{K,\rm inel}(2$  eV) =  $1 \times 10^{-14}$  cm<sup>2</sup>. Since  $n_e$  is proportional to  $(n_K)^{1/2}$  and  $n_e$  increases with the increase in temperature,  $n_K/n_e$  would be smallest (largest) for the smallest (largest)  $n_K$  at the highest (lowest) temperature. The ratio,  $n_K Q_{K,\rm inel}(2 \, {\rm eV})/(n_e Q_e(2 \, {\rm eV}))$  is estimated to be between 28 and 550 under the present experimental conditions. Since  $Q_{K,\rm inel}(\varepsilon)$  is the energy exchange cross section, this is the ratio of the rate of energy exchange.

An inelastic collision does not necessarily mean the loss of energy from free electrons. By an inelastic collision an electron loses energy and an atom is excited. The excited atom is de-excited by some process. If the excited atom is de-excited through a collision with an electron, the energy of excitation is given back to the electron (super-elastic collision). The energy lost from an electron is gained by another electron, and electrons as a whole lose no energy. On the other hand, if the excited atom is de-excited through a radiative de-excitation or through a collision with another atom, the energy lost from electrons is not recovered. It will be shown in the following that the radiative de-excitation is dominant over the super-elastic collisions.

The probability of a super-elastic collision is estimated from the cross section. Since no data are available for potassium, we again use the value for a *cesium* atom in Ref. 36. The cross section of the superelastic collision of a *secium* atom in the lowest excited level is  $50 \times 10^{-16}$  cm<sup>2</sup>. If it is multiplied by  $n_e$  and the mean thermal speed of electrons at  $T_e = 2000$  K,  $\bar{v} = 3.0 \times 10^7$  cm/s, the probability of the superelastic collision for an excited atom is estimated as  $1.5 \times 10^{-7}$   $n_e$  s<sup>-1</sup> ( $n_e$  in cm<sup>-3</sup>), which becomes  $2.1 \times 10^5$  s<sup>-1</sup> at the largest eletron number density,  $1.4 \times 10^{12}$  cm<sup>-3</sup>. The probability of the radiative de-excitation in the net account is given by [(number of atoms de-excited by the radiative de-excitation per unit time) minus (number of atoms excited by absorption of radiation per unit time)] divided by (number of atoms in the excited state). The net probability is calculated as

(radiant energy loss from unit volume of plasma)
(energy of one photon) × (number density of excited atoms)

Because the temperature is not so high, we can assume that the whole radiation comes from the transition from the first excited state to the ground state for an order-of-magnitude estimation. Then the energy of a photon is 1.61 eV. For a plasma under somewhat different conditions, Zukoski et al. [5] calculated the radiant energy loss considering effects of reabsorption with the assumption of the Boltzmann equilibrium distribution of excited atoms at the electron temperature. From their results, the radiant energy loss under the present experimental conditions can be estimated. The procedure is given in the Appendix. With estimated value for the radiant energy loss from unit volume of plasma, and the number density of excited atoms on the assumption of the Boltzmann distribution, the following results are obtained;

At  $T_e$ =2000 K, the net probability of radiative de-excitation is  $1.3 \times 10^6 \, s^{-1}$  for potassium concentration of 0.13% and  $4 \times 10^6 \, s^{-1}$  for potassium concentration of 0.013%.

These values are to be compared with the probability of super-elastic collision at the same  $T_e$ , which is smaller than  $2.1 \times 10^5 \, \mathrm{s}^{-1}$  under all conditions in the reference state. The probability of the radiative de-excitation is smaller for higher potassium concentration because the reabsorption becomes significant. The dependence of the probability on the temperature is weak. From the comparison between probabilities of two kinds of de-excitation processes it is found that the radiative de-excitation is the dominant process. Thus, most of the energy of electrons given to atoms through inelastic collisions is lost as the radiation. The ratio of collision frequencies,  $n_K Q_{K, \mathrm{inel}}(\varepsilon)/(n_e Q_e(\varepsilon))$  is the ratio of the probability of losing energy by inelastic collisions to the probability of exchanging energy among electrons. The ratio is, according to the above estimate, 28 to 550. Since the high-energy electrons are created only through e-e- collisions, the number density of high-energy electrons must be reduced very much by the inelastic collisions. As a result of this loss of high-energy electrons, the value of the

electron distribution function at high energy should be less than the Maxwellian distribution with  $T_{el}$ . The acceleration by an electric field does not serve much to restore Maxwellian distribution. As shown in the discussion on the low-energy electrons, the effect of acceleration on the distribution is smaller than that of e-e collisions. This reduction of high-energy electrons suppresses the increase in total number of electrons, when electrons are heated by the Joule heat in an electric field.

Next, the number of excited potassium atoms is considered. Fig. 25 gives a diagram of the energy levels for the valence electron of a pottasium atom. The widest gap between energy levels existing between the ground state and the first excited state is  $1.61 \, \mathrm{eV}$ . Transitions across this widest energy gap serves to absorb energy from high-energy electrons and emit it as radiation. As the ratio of energy exchang,  $n_K Q_{K,\,\mathrm{Inel}}(\varepsilon)/(n_e Q_e(\varepsilon))$  for high-energy electrons is more than 28, an electon loses its energy by inelastic collision as soon its energy reaches a value more than the threshold value,  $1.61 \, \mathrm{eV}$ , through e-e collisions. Therefore, the number of excited atoms in the first excited state is determined by the balance between the rate of creation of high-energy electrons through e-e collisions and the rate of radiative deexcitation. The number of atoms in the first excited state is thus between the equilibrium value at  $T_g$  and that at  $T_{el}$ . The number of high-energy electrons is less than the equilibrium value at  $T_{el}$ , but is more than that at  $T_g$ ;  $T_g$  is the lowest temperature of all that are present there.

Energy gaps are narrower among higher energy levels. Transitions between high energy levels are caused more easily by a collisional process with an electron or atom. On the other hand the probability of the radiative de-excitation is smaller

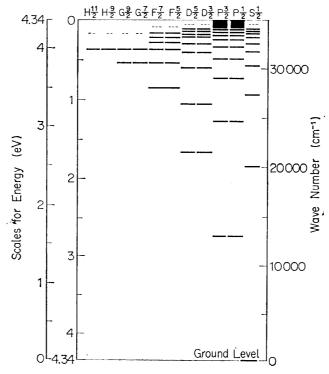


Fig. 25. Energy level structure of a potassium atom.

for the higher levels. Transitions among higher levels and the ionization from high energy levels are caused by collisions not only with high-energy electrons, but with some of low-energy electrons and atoms and by radiative transitions. As stated before, low-energy electrons are at  $T_{el}$ , atoms are at  $T_g$ , and the number of high-energy electrons and the density of radiative flux may correspond to some temperatures between  $T_{el}$  and  $T_g$ . The populations at ionized state may also lie between two equilibrium values at  $T_g$  and  $T_{el}$ .

# 6. CALCULATION OF CONDUCTIVITY OF NONEQUILIBRIUM PLASMA AND COMPARISON WITH EXPERIMENTAL RESULTS

In this section, a method is proposed for calculating the conductivity of a nonequilibrium plasma as a function of the applied electric field. The number density of electrons and the mobility are separately calculated by using two temperatures,  $T_{el}$  (defined in § 5) and  $T_{eh}$  (to be defined in this section).

In plasma in region I or II, electrons with sufficient energy for ionizing a potassium atom in the ground state by single collision, i.e.,  $\varepsilon > 4.34$  eV, are very few. Most of the ionization may occur through a multistep process, in which various processes of excitation, ionization, and their inverse processes take place. If one knows the distribution function of electrons and cross sections for these processes, one might be able to calculate the ionization fraction. However, for a nonequilibrium plasma the distribution function is to be determined simultaneously with the number of ions and excited atoms. The calculation of the distribution function is difficult because we do not have enough information on cross sections of various processes. Therefore, we use a simple method for determining the ionization fraction of a nonequilibrium plasma.

As a model to start with, we assume that numbers of atoms in the ground state, in various excited states and in the ionized state are given by the Saha-Boltzmann equilibrium at a temperature,  $T_{eh}$ , which lies between  $T_{el}$  and  $T_{g}$ . Once the value of  $T_{eh}$  is determined by some means, the number density of electrons is calculated by substituting  $T_{eh}$  in the Saha equation (§ 4). On the other hand, the mobility of electrons as a whole is determined by the Allis formula (§ 4) for Maxwellian electron distribution at  $T_{el}$ . From the number density and the mobility we calculate conductivity. The problem is how to determine  $T_{eh}$  and  $T_{el}$ .

The amount of energy input to electrons per unit volume per unit time is equal to the Joule heat, *iE*. Most of the Joule heat is given to low-energy electrons, because they constitute most of electrons. They lose a part of the energy by elastic collisions with atoms, and the rest of the energy is given to high-energy electrons through e-e collisions. High-energy electrons, which receive energy partly from the electric field and partly from low-energy electrons, in turn, consume the energy mainly in exciting potassium atoms through inelastic collisions. The energy given to potassium atoms is lost mainly by the radiation from excited atoms. A schematic flow chart of the energy is given in Fig. 26. The direct energy transfer from the electric field to high-energy electrons is small and is neglected for sim-

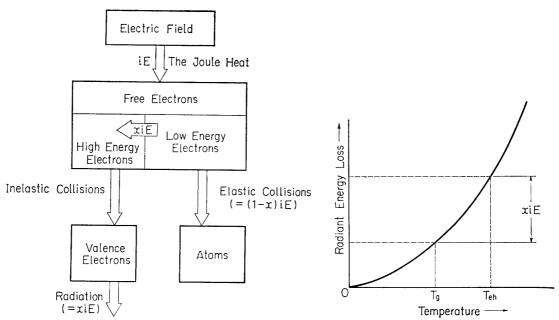


Fig. 26. Energy flow in plasma.

Fig. 27. Scheme of determing  $T_{eh}$ .

plicity. The total amount of the energy given to low-energy electrons is iE. The net energy flux from low-energy electrons to high-energy electrons is put to be xiE. This, in other words, represents the rate of the increase of high-energy electrons by collisions among low-energy electrons. Since we retain only radiant loss for high-energy electrons, the amount of the lost energy should be equal to the net energy flux from low- to high-energy electrons, xiE Therefore, x is considered to be the fraction of the radiation loss in the energy given by the Joule heat. The value of x should be between 0 and 1. The rate of energy transfer from low-energy electrons to heavy particles through elastic collisions is (1-x)iE. An extreme cases, x=0 corresponds to the situation in which no high-energy electrons are created and consequently there is no radiation from the plasma. On the other hand, x=1 means that all the input energy are converted into radiation and there is no energy loss through elastic collisions.

Since we assume that populations of excited and ionized states are given by the Saha-Boltzmann distribution at a temperature  $T_{eh}$ , the radiant energy flux from plasma is determined by  $T_{eh}$ .

A relation between the electron temparature and the radiant energy loss is derived in the Appendix. The relation is for a cold, blackbody enclosure. In the present experiment the enclosure is at the same tempearature as the gas, i.e., at  $T_g$ . When  $T_e = T_g$ , there should be no net flux of radiation from plasma to wall. Wall of the test chamber radiates the same amount of energy as that from the plasma at  $T_g$ . Thus the net energy flux from the plasma should be the difference of the flux from plasma at  $T_{eh}$  and the flux from plasma at  $T_g$ , as indicated in Fig. 27. The net flux from unit volume of plasma is  $xiE = (x\sigma)E^2$ . The value of  $T_{eh}$  is determined as a function of the radiant loss. This  $T_{eh}$  determines  $n_e$ . Thus there is one-to-one correspondence between  $n_e$  and  $x\sigma E^2$ . The value of  $\sigma$  is proportional to  $n_e$ .

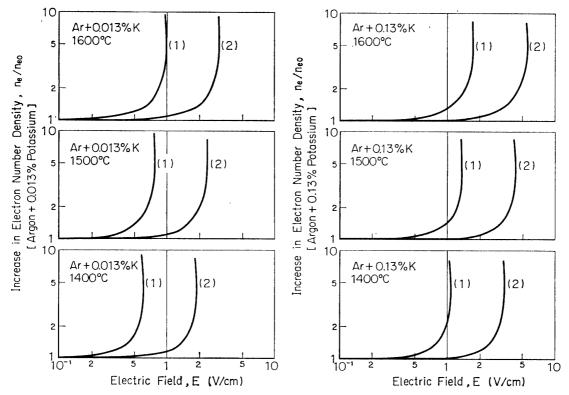


Fig. 28. Calculated electron number density versus electric field. Number density is normalized by its value at a vanishingly small electric field,  $n_{e0}$ . x; distribution parameter,  $b_0$ ; conductinity in a vanishingly small field.

(1)  $x\sigma_0=0.01 \text{ mhos/cm}$ ,

(2)  $x\sigma_0 = 0.001 \text{ mhos/cm}$ .

We consider a plasma of which conductivity is  $\sigma_0$  in a vanishingly small E, and specify a value of x. Then for each value of  $n_e$ , one value of E is determined. The  $n_e - E$  relation is illustrated in Fig. 28 with a parameter  $\sigma_0 x$ , where  $n_e$  is normalized by its value in a vanishingly small E,  $n_{e0}$ .

The mobility of electrons is calculated by the Allis formula with the assumption that low-energy electrons are in a Maxwellian distribution at  $T_{el}$ .

The ratio of frequencies of e-i and e-Ar collisions is given by,

$$n_i \overline{Q}_i / (n_{Ar} \overline{Q}_{Ar}) \le (1.4 \times 10^{12})(4 \times 10^{-12}) / [(4 \times 10^{18})(2.3 \times 10^{-17})] = 0.06$$

in which  $\overline{Q}_i$  is taken as  $4 \times 10^{-12}$  cm<sup>2</sup> at  $T_e = 2000$  K. Since in the above estimation the maximum value of  $n_i$  was used, the value of 0.06 is the maximum value of the ratio. Thus the e-i collision is negligible in comparison with the e-Ar collision. By a discussion similar to the above we have

$$n_K \overline{Q}_K/(n_{Ar} \overline{Q}_{Ar}) = 0.1,$$

for potassium concentration of 0.013%. The e-K collision is also negligible. For potassium concentration of 0.13%,  $n_K \overline{Q}_K$  and  $n_{Ar} \overline{Q}_{Ar}$  are in the same order of magnitude. As mentioned earlier, reliable data on  $Q_K(\varepsilon)$  are not available at small  $\varepsilon$ . We take as  $Q_K(\varepsilon)$  the recent result for *cesium* atom by Nighan [29]. Then, the

tatal frequency of momentum-transfer collisions of an electron with argon and potassium atoms,  $\nu_t$  is given by

$$v_t = v(\varepsilon)[n_{AT}Q_{AT}(\varepsilon) + n_KQ_K(\varepsilon)] = v(\varepsilon)n_{AT}Q_t(\varepsilon),$$

where

$$Q_t(\varepsilon) = Q_{A_T}(\varepsilon) + (n_K/n_{A_T})Q_K(\varepsilon).$$

The value of of  $Q_t(\varepsilon)$  for the case of 0.13% potassium, that is,

$$Q_t(\varepsilon) = Q_{Ar}(\varepsilon) + 0.0013Q_K(\varepsilon)$$

is illustrated in Fig. 29, together with  $Q_{Ar}(\varepsilon)$ .

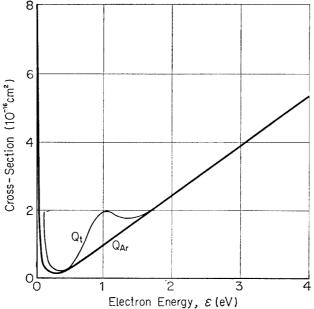


Fig. 29. Cross section of an argon-potassium mixture.  $Q_{Ar}$ ; cross section of an argon atom by Frost et. al. (Ref. 14),  $Q_t$ ; assumed cross section of argon+0.13% potassium mixture,  $Q_t = Q_{Ar} + 0.0013 Q_{Cs}$  ( $Q_{Cs}$ ; by Nighan, Ref. 29).

The temperature  $T_{el}$  is determined by an equation of energy balance in elastic collisions,

$$(1-x)iE = (1-x)\sigma E^2 = \sum_j (m_e \delta_j / m_{\alpha j}) n_e \bar{\nu}_{ej} (3/2) k (T_{el} - T_g).$$

The left-hand side of the equation is a part of the Joule heat given to electrons and the right-hand side is the energy transferred to gas atoms per unit time and volume. The right-hand side is the sum of two terms, which correspond to argon and potassium atoms, respectively. For argon and potassium, the value of  $m_a$  (mass of an atom) is nearly the same, and  $\delta$  is almost the same. Temperature  $T_g$  is assumed to be the same for all kinds of atoms and ions. So the right-hand side is equal to  $(m_e\delta/m_a)n_e(3/2)k(T_{el}-T_g)\sum \bar{\nu}_{ef}$ . Values of  $\delta$  and  $m_a$  are put equal to those for argon. The value of  $\sum \bar{\nu}_{ef}$  is given by

$$\sum_{j=1}^2 \bar{\nu}_{ej}(T_{el}) = \bar{v}(T_{el}) \overline{Q}(T_{el})$$

where  $\bar{v}$  is the mean thermal speed of electrons and  $\bar{Q}$  is the averaged cross section calculated from  $Q_t(\varepsilon)$  just like  $\bar{Q}_{Ar}$  in § 4. A simple expression for  $\sum \bar{v}_{ej}$  is derived, by the use of

$$\overline{Q} = 4e/(3\sqrt{2\pi m_e mkT_{el}}n_a\mu).$$

One can combine this equation with the relation

$$m_e \bar{v}^2 = 3kT_{el}$$

to obtain

$$\sum \bar{\nu}_{ej} = \bar{v} n_a \overline{Q}_a = \frac{4e\sqrt{3kT_{el}/me}}{3\sqrt{2\pi m_e kT_{el}}\mu} = \frac{2\sqrt{2}e}{\sqrt{3\pi}m_e \mu}.$$

Both sides of the energy balance equation is divided by

$$(1-x)\sigma = (1-x)n_e e\mu$$

and the expression for  $\sum \bar{\nu}_{ej}$  is introduced. The result is,

$$E^{2} = \frac{1}{1-x} \sqrt{\frac{6}{\pi}} \frac{\delta}{\mu^{2} m_{a}} k(T_{el} - T_{g}),$$

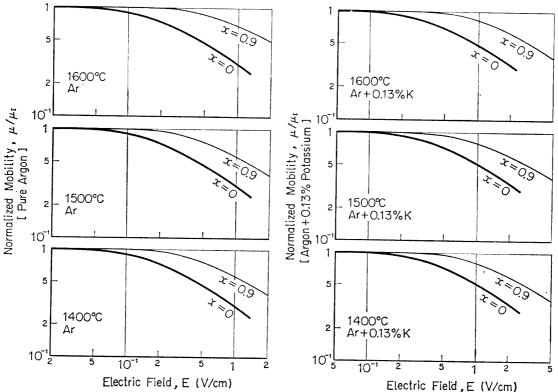


Fig. 30. Mobibity versus electric field. Mobility normalized by that in a vanishingly small field. x; distribution parameter.

in which  $\mu$  is known as a function of  $T_{el}$ . This is combined with the  $\mu-T_{el}$  relation to eliminate  $T_{el}$ , so as to obtain  $\mu-E$  relation. Results are illustrated in Fig. 30, with x as parameter. For instance, if x=0.2, low-energy electrons in an argon with 0.013% potassium at  $T_g=1773$  K (1500°C) are heated to  $T_{el}=2770$  K by an electric field E=0.25 V/cm, and  $\mu$  decreases to 70% of the equiblium value.

The last step of calculation is to combine  $\mu-E$  and  $n_e-E$  relations to obtain  $\sigma-E$  relation. For a particular gas temperature, gas composition, and value of x, we first assign a value to E. The value of  $\mu$  is given by the  $\mu-E$  curve. The value of  $n_e$  would be given by the  $n_e-E$  curve with a value of  $\sigma_0 x$ . It must be noted here that we should not put  $\sigma_0 = \sigma_I$ . The reason is as following. The  $n_e(\sigma_0, E) - E$  relation is for a constant  $\mu$ . Actually  $\mu$  decreases as E is increased. The value of  $\sigma$  with the reduced  $\mu$  is  $\sigma_I n_e/n_{e0}$  multiplied by  $\mu(E)/\mu_I$ . Thus we should put  $\sigma_0 =$ 

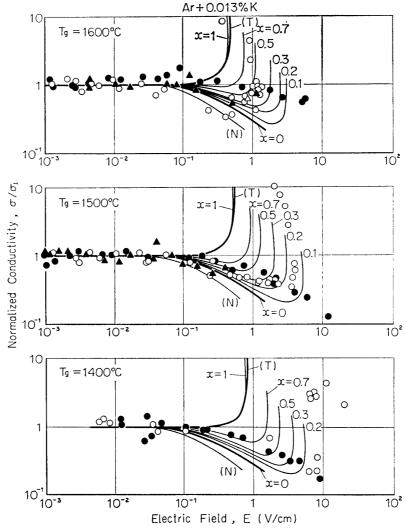


Fig. 31. Comparison between calculated and experimental results; conductivity versus electric field. I. Atmospheric argon+0.013% potassium. Conductivity is normalized by its value in region I,  $\sigma_I$  of each run. x; distribution parameter, (T) two temperature theory, (N) non-Maxwellian electron energy distribution with constant  $n_e$ .

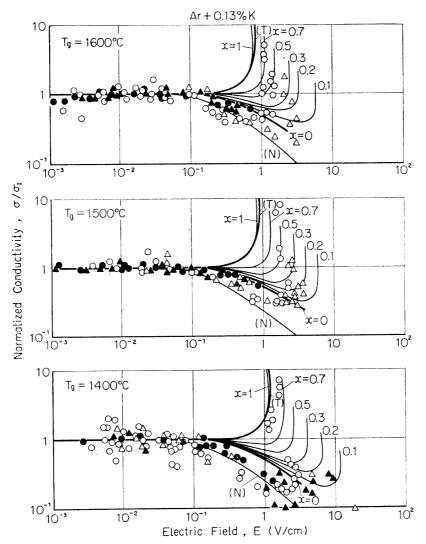


Fig. 32. Comparison between calculated and experimental results; conductivity versus electric field. II. Atmospheric argon +0.13% potassium. Conductivity is normalized by its value in region I,  $\sigma_I$  of each run. x; distribution parameter, (T) two temperature theory, (N) non-Maxwellian electron energy distribution with constant  $n_e$ .

 $\sigma_I \mu(E)/\mu_I$ , because  $\sigma_0$  is the value of  $\sigma$  the plasma would have when  $n_e = n_{e0}$  and  $\mu = \text{constant} = \mu(E)$ . Combining  $\mu$  from the  $\mu - E$  relation (with the parameter x) and  $n_e$  from the  $n_e - E$  relation (with the parameter  $\sigma_0 x = \sigma_I x \mu(E)/\mu_I$ ), we obtain  $\sigma = e \mu n_e$  for the assigned E. For a set of value of E, the corresponding values of  $\sigma$  are known. The locus of the points  $(\sigma, E)$  for a particular x is obtained.

The final result is a set of  $\sigma - E$  curves for various values of the parameter, x. Calculated results are compared with the experimental data in Figs. 31 and 32. The conductivity is normalized by the value in region I,  $\sigma_I$  of each experiment.

If x=0, heating of electrons in an electric field does not result in an increase in the number density of electrons,  $n_e$  at all, while the mobility is decreased, in other words,  $T_{eh} = T_g$  and  $T_{el}$  is raised. On the other hand, if  $T_{eh} = T_{el}$ , the result of

the present calculation is the same as that of the conventional two temperature theory [34], in which Maxwellian distribution of electrons and the Saha-Boltmann equilibrium at the electron temperature are assumed. The  $\sigma-E$  relation by the two temperature theory is shown in Fig. 31 and 32 by (T). The result is very close to the present result with x=1.

Curves indicated by (N) represent  $\sigma - E$  relation at another extreme, in which the number density of electrons is assumed to be unchanged regardless of the applied field. The e-e collision is neglected in comparison with the e-Ar collision. The distortion of the electron distribution function due to the electric field is determined by the Boltzmann equation. Frost and Phelps [15] give a solution neglecting inelastic collisions as

$$f(\varepsilon) = A \exp \left[ -\int_0^{\varepsilon} \left( \frac{m_a E^2 e^2}{6m_e n_a^2 (Q_a(\varepsilon))^2} + kT_g \right)^{-1} \right] d\varepsilon$$

where  $f(\varepsilon)$  is the spherically symmetrical term in the expansion of the energy distribution function. Suffix "a" denotes atoms, and other symbols have their usual

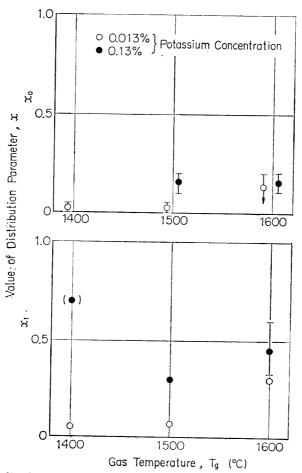


Fig. 33. Distribution parameter x determined by comparison with experimental results.  $x_0$ ; determined at the minimum of  $\sigma - E$  curve,  $x_1$ ; determined at the boundary between regions II and III.

meanings. Calculations are made by putting  $m_a = m_{A_T}$  and

$$n_a Q_a(\varepsilon) = n_K Q_K(\varepsilon) + n_{Ax} Q_{Ax}(\varepsilon)$$
.

The mobility is calculated as a function of the electric field E by substituting  $f(\varepsilon)$  into the Allis formula. The value of  $\sigma$  is obtained as a function of E. The decrease in  $\sigma$  with the increase in E is more rapid than that in the calculation with Maxwellian distribution for low-energy electrons.

It is obvious that such a calculation (N) does not agree with experimental results. The curve (T) does not agree either. Experimental data lie between these two extreme cases.

The value of x is determined from the comparison between the experimental data and the calculated  $\sigma - E$  curves with various values of x. The value of x differs under different conditions of plasma. Fig. 33 presents a correlation between x and  $T_g$ . One value of x denoted by  $x_0$  is determined from comparison with the experimental values at the minimum of  $\sigma$ . The other,  $x_1$ , is determined at the boundary between regions II and III, where  $\sigma = \sigma_I$ . Fig. 33 indicates that the value of x increases with the increase in  $T_g$  and potassium concentration. Moreover,  $x_1$  is larger than  $x_0$  at the same value of  $T_g$ . Directly from Figs. 31 and 32 one can see that x increases with the increase in  $\sigma$ . Those features of dependence of x on the plasma conditions may be explained by a model culculation, which is now being worked and will be reported later.

# 7. Nonequilibrium Conductivity at High Current Density

In region III the conductivity is higher than the equilibrium value, i.e.,  $\sigma_{III} > \sigma_I$ . An oscillogram of  $V_p$  (proportional to the electric field) shows that the conductivity increases with time during a pulse. This is because the increase in the number density of electrons overcomes the decrease in the mobility. As discussed in the preceding section, the best fit value of x increases with the increase in the current density. This means that more fraction of energy is transferred to high-energy electrons. As a result  $n_e$  increases. When the rate of the electron-electron energy transfer is much more than that of the electron-atom energy transfer throughout the whole range of electron energy, electrons are in a Maxwellian distribution and the conventional two-temperature theory is valid. If one takes electrons of energy 2eV as an example, the ratio of probabilities of electron-atom energy transfer and electron-electron energy transfer becomes 0.5 at an electron number density of  $10^{14}$  cm<sup>-3</sup>. At higher density of electrons, the two-temperature theory is certainly valid.

In the present experiment  $n_e$  is not high enough and the two-temperature theory is not applicable as shown in Figs. 31 and 32. On the other hand, the present method of calculating conductivity should give good results over the whole range of current density and  $n_e$ . In the experiment, however, there is a possibility of spatial non-uniformity of the plasma in the nonequilibrium state. When the

nonequilibrium ionization takes place the electric field in the plasma does not increase much with the increase in the current density or even decreases (see, e.g., Fig. 11). This fact is verified also by experimental results by Kerrebrock and others [3-6] in a nonequilibrium plasma. If the field decreases as the current density increases, the current in the plasma may not be evenly distributed. The current might concentrate in a narrow channel of high conductivity and high current density. An increase in the total current may not change the current density but may result in the increase of the cross sectional area of the high-conductivity channel. The nonuniformity of the current density may be caused also by the difference in the radiant energy loss. The inner part of a plasma is shielded against radiant loss by the outer part and loses energy less than the outer part. Thus the inner part may have more electrons than the outer part and has the larger current density.

In the present experiment with isothermal arrangement the gas temperature is quite uniform over the whole plasma and enclosure. The hot enclosure emits radiation back to the plasma. Nevertheless, at an elevated electron temperature a nonuniform current distribution due to the negative voltage-current characteristics may still exist. The nonuniformity may arise from the presence of a sheath near the electrode. The sheath voltage can be as high as 50% of the total electrode voltage. In addition, the ratio of length to diameter of the plasma is only 1.7, i.e., the plasma is relatively fat in shape. Consequently, the sheath voltage is important in determining the current distribution. If the sheath voltage increases as the current density increases, the sheath has a stabilizing effect and the current concentration will be prevented. In regions I and II, this is the case, as seen from Fig. 18. On the contrary, in region III the sheath voltage decreases with the current density. This enhances the nonuniformity of the current density. In view of several points discussed above, it is doubtful that the cross sectional distribution of the current density should be uniform in region III. Perhaps there are several high-current spots on the electrodes. In fact, in region III the value of probe voltage  $V_p$  (i.e., E) sometimes showed fluctuation or sudden change during a current pulse. Such behavior was never found in regions I and II. We conclude that even with the present experimental arrangements the current density is not uniform in region III. We do not think it meaningful to discuss on the details of the experimental values of the conductivity in this region.

In the experiments by Kerrebrock et al. [3, 4] and by Zukoski et al. [5, 6]  $n_e$  reaches a value high enough for applying the two-temperature theory. Good agreements between experimental and calculated results at high current density were demonstrated [3-8]. However, in those experiments the gas temperature was not uniform. The wall of the test section was cold and the amount of the radiation was larger. These conditions are unfavorable for the uniform distribution of the current. There is no experimental proof that the current density was uniform. Perhaps the situation for the current distribution is even worse than in region III of the present experiment. Detailed discussions on the conductivity does not seem to be meaningful also in their experiments.

#### 8. Conclusions

Electrical conductivity of an argon-potassium mixture at a gas temperature about 1500°C is a function of the current density. When the current is small, the plasma is in thermal equilibrium at the gas temperature and the conductivity does not depend on the current density. When the electric field in the plasma exceeds a certain value, which is about 0.04 V/cm, the conductivity starts to decrease with the increase in the current density (or electric field), shows a minimum at a certain current density and increases with further increase in the current. For a helium-potassium mixture, the presence of a minimum of the conductivity is not clear.

The sheath voltage on the electrodes is not negligible in comparison with the voltage across the bulk of plasma of 4 cm-length.

The average cross section of a potassium atom for momentum transfer with electrons is determined from equilibrium conductivity to be  $2.3\times10^{-14}$  cm<sup>2</sup>, for electrons with Maxwellian energy distribution at  $1500^{\circ}$ C.

When there is an appreciable electric field in a plasma, electrons are accelerated and thermalized by mutual collisions. This is the Joule heating. The plasma is not in equilibrium because electrons are selectively heated. From considerations on various processes taking place in a nonequilibrium argon-potassium plasma it is concluded that electrons are not in a Maxwellian distribution. Due to inelastic collisions with potassium atoms, high-energy electrons lose their energy and their number density is substantially less than that in a Maxwellian distribution. This reduction of high-energy electrons results in the ionization fraction less than expected from the "temperature" of low-energy electrons. We propose to use two temperatures for electrons, one  $T_{el}$  representing the low-energy part and the other  $T_{eh}$  for the high-energy part. The mobility of electrons is controlled by  $T_{el}$ , while the number density of electrons is determined by  $T_{eh}$ . Both temperatures are determined by the amount of energy received from the electric field and the energy flux by elastic and inelastic collisions. By introducing a parameter for the ratio of the two kinds of energy flux we can determined  $T_{el}$  and  $T_{eh}$ . Thus, the nonequibrium conductivity is calculated as a function of the electric field and the partition parameter. The decrease in conductivity with increased current density is shown plausible and functional features of experimental results are satisfactorily interpreted.

When the current through a plasma is larger, the current distribution seems to become nonuniform and unstable. No reliable data on the conductivity were obtained in that case.

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# APPENDIX: RADIANT ENERGY LOSS FROM LOW-TEMPERATURE PLASMA

In order to calculate the conductivity of a nonequilibrium plasma the amount of the radiant energy loss from unit volume of the plasma should be known as a function of the electron temperature. Calculation of the loss is complicated by the fact that the radiation from a part of plasma is absorbed again by another part of plasma. Zukoski, Cool, and Gibson [5] calculated the rate of the radiant energy loss, considering the re-absorption processes for a potassium-seeded argon plasma of a cylindrical geometry, with 1.91 cm-length and 1.29 cm-diam. The total pressure is atmospheric, potassium concentration is 0.4% in mole fraction, and the gas temperature is  $2000 \, \text{K}$ . The amount of radiation from unit volume is calculated for electron temperatures from  $2400 \, \text{to} 3500 \, \text{K}$ . Populations of excited states are assumed to be given by the Boltzmann distribution at the electron temperature  $T_e$ . Their results are shown in Fig. 34 with filled circles.

Conditions of the plasma in the present experiment are; pressure, atmospheric; potassium concentration, 0.013 or 0.13%; gas temperature  $T_g$ , 1673, 1773, and 1873 K (1400, 1500, and 1600°C respectively); geometry, a cylinder with 4.0 cm-length and 2.3 cm-diam.; range of electron temperature  $T_{eh}$  to be concerned, from 1673 to 2400 K.

Since physical conditions of plasma are different from that of Zukoski et al., an appropriate reduction rule is necessary for estimating the amount of radiant

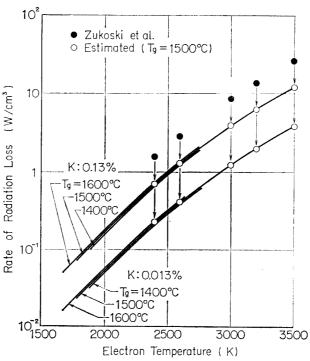


Fig. 34. Estimated rate of radiant energy loss from unit volume. Atmospheric argon+0.013 or 0.13% potassium enclosed in a cold, black-body cylinder of 23 mm-i.d. and 40 mm-height. The rate is reduced from calculated results by Zukoski et al. (Ref. 5) and extrapolated to lower temperatures.

energy loss in the present case from the values in Ref. 5. Considering several published results on the dependence of radiation on parameters of state, we conclude that the rate of radiant energy loss from unit volume of a plasma is proportional to; i) square root of the potassium concentration, ii) inverse square root of the linear dimension, (or for different geometrical shapes, inverse square root of the volume-to-surface area ratio), 3/4 power of the absolute gas temperture  $(T_q)$ . The rules i) to iii) are deduced from equations (3), (5), (6) and (7) in the paper by Hiramoto et al. [36]. Another support to the rules i) and ii) are found in equations (3) and (4) of Ref. 34, with an assumption that the line breath of a resonance line is mainly determined by collisions of a potassium atom with argon atoms. Validity of the last assumption is, for instance, shown by Lutz in a sample calculation [37].

The values of radiation loss for potassium concentrations of 0.013 and 0.13% are obtained with these rules, and presented in Fig. 34 denoted by open circles. One needs values of radiation loss at lower electron temperature for the present calculation of conductivity. We extrapolate the two curves down to 1650 K. These curves are used for determining  $T_{eh}$ .