Study of nonequilibrium ionization-recombination kinetics
with a shock tube method by rarefaction wave cooling

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Abstract. The abstract A shock tube method is introduced to study the ionization-recombination kinetics of high temperature gas, in which a test gas is heated and ionized by a reflected shock wave and subsequently quenched by a strong rarefaction wave reflected on the end wall of the driver section as the main cooling wave associated with a rarefaction wave incident back into region 5 when the reflected shock wave interacts with the contact surface. As the quenching rate of the strong rarefaction wave reaches 10^6K/s, a nonequilibrium ionization-recombination process occurs, during which the ion recombination with electrons dominates.

1 Introduction

Ionization-recombination kinetics of gas at high temperatures is closely related with enhancement and attenuation of plasma sheath and ionization wake arising from hypersonic flight, and plays an important role in arc discharge, plasma chemical industry and plasma chemical vapor deposition. It is significant to develop detailed study methods, master the law of ionized gas kinetics at high temperatures, and provide ionization and recombination mechanisms as well as elementary kinetic parameters such as rate coefficients of ionization and recombination.

The previous studies on ionization-recombination processes were performed in arc discharge and flame. In arc discharge, the energy of incident electrons is usually changed while gas molecules keep at the low temperatures. It is hardly used to measure the kinetic data at the state of thermally excited atoms and molecules. However, the excitation of internal state of atoms and molecules is important for nonequilibrium kinetic processes. On the other hand, the deficiency of the experiments in flame is the severe restriction by the flame condition. All the experiments are restricted within the narrow limits of flame temperature. In addition, complicated composition and chemical reactions in flame interfere with the determination of ionization-recombination processes.

Shock tube is another effective method to study the ionization-recombination process. Its advantage is that the gas state parameters can be fairly well controlled and determined by use of aerodynamic methods, and that the ionization-recombination process occurs in an unadulterated environment. In the conventional shock tube method to study the ionization-recombination kinetics, a pressure driven shock tube is used to produce a reservoir of high temperature gas, which is subsequently expanded in a nozzle to attain a cooling state. Then the electron density, electron temperature and radiation intensity are measured along the nozzle. The typical examples are the experimental determinations of the dissociative recombination rates of NO^+, N_2^+ and O_2^+ with electrons by measuring the excessive electron number density along the nozzle in a high enthalpy shock tunnel, made by Dunn and Lordi[1,2,3].

In this work, another shock tube method is introduced to study the ionization-recombination kinetics of high temperature gas, in which a test gas is heated and ionized by a reflected shock wave and subsequently quenched by a strong rarefaction wave reflected on the end wall of the driver section of the shock tube. As a result, a nonequilibrium ionization-recombination process occurs, during which the ion recombination with electrons dominates. Thus, the variation of electron number density in the cooling process of the strong rarefaction wave can be monitored by using such Langmuir probe technique to study the ionization-recombination process.

By comparison, the way to obtain the ionized gas is same as Dunn and Lordi's by the reflected shock wave heating, but the subsequent cooling process is quite different. Here we use an incident unsteady strong rarefaction wave produced as the expansion fan reflects on the end wall of the driver section.

In order to obtain the optimum reflection time of the rarefaction wave, a piston adjustable in position is fixed in the driver section, so that the head-on collision of the reflected shock with the oncoming contact surface occurs when the reflected rarefaction head overtakes the contact surface. Since another rarefaction wave is reflected back into region 5 when the reflected shock passes through the contact surface under the present condition, thus the two rarefaction waves come simultaneously into region 5 to enhance the cooling strength of rarefaction wave. Using a piezo-electric transducer to monitor the pressure evolution of the whole process, the cooling rate of the rarefaction wave is measured as K/s, which is comparable to the strength of Dunn and Lordi's nozzle cooling.

The advantage of the present method is that the whole process of the rarefaction cooling and the variation of the electron density can be monitored simultaneously. The variation of ionized gas property can be studied under a quasi-stationary condition. In comparison with Dunn and Lordi's method, as the non-ideal effects of flow are excluded, the present method is conducive to accurately determining and analyzing kinetic process.

2 Experimental

The experiments are conducted in a single pulse shock tube made of stainless steel at the Institute of Mechanics, Chinese Academy of Sciences. The driver section has a length of 1.2m, and the driven section is 1.8m in length. Both have circular cross sections with an inner diameter of 44mm. A 20L dump tank is connected to the driven section through a branch tube just ahead of the diaphragm, which is utilized for absorbing the reflected shock wave and enhancing the cooling rate. The pure aluminum foil diaphragm is used and handle control of a needle to rupture the diaphragm is adopted to ensure a good reproducibility for experiments. Before every run, the driven section is evacuated down to a minimum pressure of 10Pa. The leakage rate is smaller than 10 Pa/min. The time interval from all valves closing to the diaphragm rupture is shorter than 3min. Hydrogen is used as the driver gas. The test gas is argon of purity greater than 99.8% as diluent, mixed with a small quantity of the substance investigated. A piston adjustable in position is fixed in the driver section in order to obtain the optimum reflection time of the rarefaction wave. Both the experiment and the calculation give that the optimum position of the pistol is 0.83m apart from the diaphragm in the present experiments. The shock tube is run at a fix initial temperature of $T_1 = 343.2$K, and initial pressures of $P_1 = 4$KPa.

A typical pressure measurement record is shown in Fig.1, when the length of the driver section is 0.83m. The wave diagram in an ideal gas calculated for a typical case of $M_S = 3.5$ is shown in Fig.2, where S, S', S_R, C and R denote the incident shock, the second shock, the reflected shock, the contact surface and the rarefaction wave head, respectively; S_T, R' denote the shock wave transmitted into region 3 and the rarefaction wave reflected back into region 5 after the collision of the reflected shock with the contact surface, respectively; and O denotes the position of the diaphragm. The existence of the dump tank is equivalent to an expanding section at the entrance of the driven section and can be described equivalently with one-dimensional steady expansion. The experiment gives that the equivalent steady expansion section ratio is 1.8. The calculation indicates that the critical speed of sound is reached at the diaphragm and subsequently a shock wave S' is created to adjust gas flow speed to be matched with that in region 3.

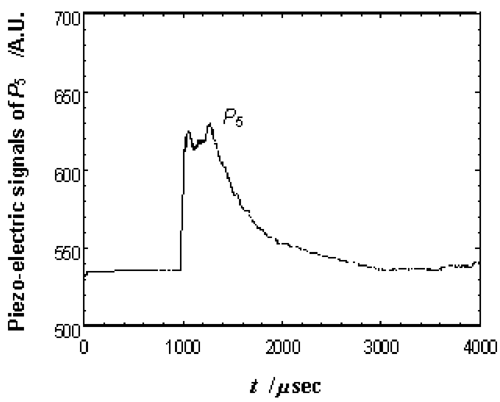

Fig. 1. Measurement record of the piezo-electric signal of P_5 at the length of the driver section being 0.83m

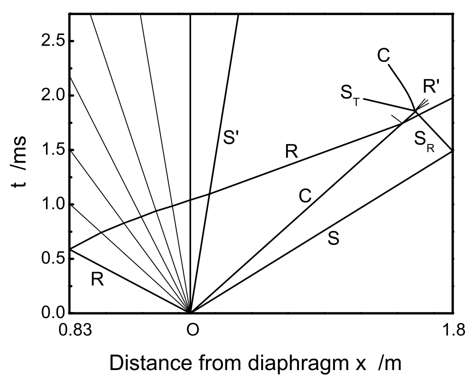

Fig. 2. Wave diagram of the single pulse shock tube with a dump tank ($M_S = 3.5$)

3 Cooling rate of the rarefaction wave

A piezo-electric transducer mounted on the end wall of the driven section is used to monitor P5 and the pressure evolution during the rarefaction wave arriving. The inert gas argon is chosen as diluent in the present experiments so as to neglect the heat effect of the substance investigated. Furthermore argon atoms do not ionize and maintain to be at the ground state in this case, and then the cooling stage could be treated as an adiabatic process. The varying history of density and temperature of the test gas in the cooling process could simply be derived from that of pressure by use of the adiabatic relations:

$$\left(\frac{\rho}{\rho_{50}}\right) = \left(\frac{P}{P_{50}}\right)^{\frac{1}{\gamma}}, \quad \left(\frac{T}{T_{50}}\right) = \left(\frac{P}{P_{50}}\right)^{\frac{\gamma-1}{\gamma}} \tag{1}$$

where $\gamma = \frac{5}{3}$ is the adiabatic index of argon and the subscript 50 refers to the corresponding initial state just behind the reflected shock wave.

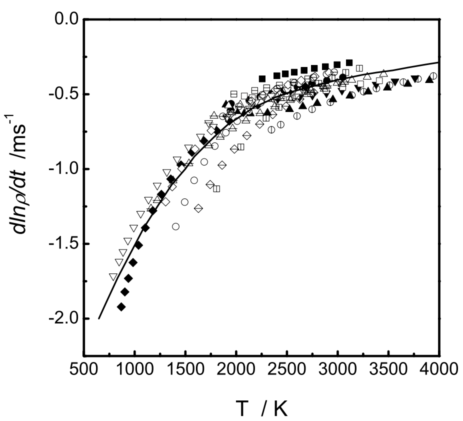

Fig. 3. Variation of $\frac{dln\rho}{dt}$ with temperature

The variation of $\frac{dln\rho}{dt}$ with temperature in the cooling process can be obtained according to the adiabatic relations. Fig.3 shows the measurement result of 21 runs with the Mach number ranging from 3.1 to 3.8. The different symbols on the graph correspond to different running Mach number. Furthermore, the plot of $lg\left(-\frac{dln\rho}{dt}\right)$ versus lgT exhibits a liner property, as shown in Fig. 4. Using least square analysis of all the data points in Fig. 4, we obtain

$$lg\left(-\frac{dln\rho}{dt}\right) = -1.068lgT + (3.371 \pm 0.114) \tag{2}$$

Using the adiabatic relations again, the cooling rate can be derived from Eq.(2)

$$\frac{dT}{dt} = (-1.62 \pm 0.42)\times 10^{6} T^{-0.068} \text{ K/s} \tag{3}$$

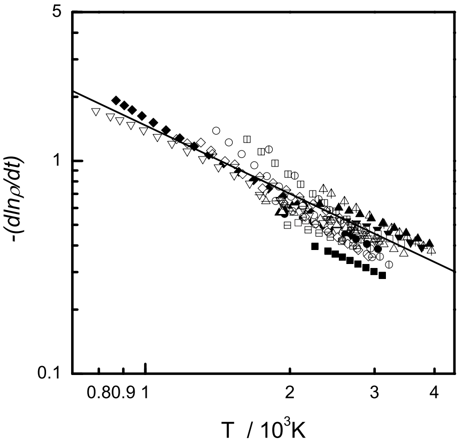

Fig. 4. Variation of $lg\left(-\frac{dln\rho}{dt}\right)$ with lgT

The exponent of temperature in Eq.(3) indicates that $\frac{dT}{dt}$ is almost independent of temperature and constant in the cooling process of the present experiments. Using this relation, the derivative with respect to temperature can be transformed into the derivative with respect to time.

From Eq.(3), we know that the cooling rate of the rarefaction wave reaches the magnitude order of 10^6K/s in the present experiments, which is comparable to the strength of Dunn and Lordi's nozzle cooling. Within the experimental time of about 3ms in the present experiments, the temperature in the reflection shock wave region could decrease from 3000K to 600K.

Strictly speaking, there exist three cooling processes in the present single-pulse shock tube. The first cooling wave is the rarefaction wave reflected on the end wall of the driver section; the second is the rarefaction wave reflected back into region 5 when the reflected shock passes through the non-tailored interface; and the third is the rarefaction wave incident in the dump tank when the reflected shock passes through it. The cooling effect of the third rarefaction is negligible, and the strong rarefaction reflected on the end wall is the main cooling wave. This strong reflected rarefaction as the main cooling wave is more effective than the rarefaction reflected back into region 5 under the condition for a non-tailored interface.

4 Conclusion

A shock tube method is introduced to study the ionization-recombination kinetics of high temperature gas. The feature of this method is that a test gas is heated and ionized by a reflected shock wave and subsequently quenched by a strong rarefaction wave reflected on the end wall of the driver section as the main cooling wave associated with a rarefaction wave incident back into region 5 when the reflected shock wave interacts with the contact surface. In order to obtain the optimum reflection time of the rarefaction wave, a piston adjustable in position is fixed in the driver section to change the length ratio of the two sections, so that the reflected shock and the reflected rarefaction head collide simultaneously with the contact surface to produce a strong convergent rarefaction wave into region 5. By both calculating the wave system under the typical condition and the experimental observation, this method is verified feasible. Furthermore, the optimum position of the pistol is 0.83m apart from the diaphragm, and the cooling rate of the rarefaction wave in region 5 reaches the magnitude order of 10^6K/s, which is comparable to the strength of Dunn and Lordi's nozzle cooling. Using the present method, by comparison, the requisite device is simple, and it is possible to monitor the whole evolution process of a given particle of test gas at the same observation point and to realize a study of ionization-recombination process of high temperature gas at an almost static condition. Since the non-ideal effects of flow are excluded, the present method is conducive to accurately determining and analyzing kinetic processes. We have determined the recombination rate coefficients of NO^+ ion, Na^+ ion, and F atom with electrons at the temperature range of 1000 $\sim$ 3000K by using the present method[5,6,7].

References

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