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Laser-induced Fluorescence Spectroscopy with Sub-nanosecond Laser Pulse toward Observation of Excitation Transfer Processes of Helium Atoms in an Atmospheric-pressure Plasma

大気圧プラズマ中でのヘリウム原子励起移行過程の観測に向けたサブナノ秒レーザー誘起蛍光分光

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In a glow discharge helium plasma at pressures from 0.001 to 0.1 atm and in a micro-hollow cathode plasma from 0.05 to 1.0 atm, a laser light pulse of 620 ps width excites helium atoms from the 2^1S level to 3^1P level. Temporal developments of the intensities of the subsequent fluorescence from n=3 levels are observed. From the coupled rate equation for the populations in the 3^1P and 3^1D levels with the excitation-transfer and quenching rates of these levels, the temporal developments are examined.

1. Introduction

It is known that excitation transfer processes between energetically close excited levels take place in a plasma due to atom collisions together with collisional quenching. Such processes may become substantial in high pressure plasmas like an atmospheric-pressure plasma, which gathers much notice recently for many kinds of applications because of their freedom from vacuum enclosure and high particle density [1]. For the plasma diagnostics, quantitative evaluations of the collision rates and of the limit of binary collision approximation are necessary.

For a helium plasma at a gas pressure less than 0.01 atm, excitation from an n=2 level to an n=3 level with a several ns laser pulse has been done and subsequent fluorescence from several n=3 levels have been observed, where n is a principle quantum number [2-5]. From the analysis of the temporal development of the fluorescence intensities, the excitation-transfer and quenching rate coefficients have been determined [2-5].

At the gas pressures larger than 0.01 atm, the time constant evaluated from the rates become comparable to the laser pulse-width, and then the quantitative analysis of the temporal development becomes difficult. Furthermore, a conventional glow discharge cannot generate a stable plasma at the gas pressures larger than 0.1 atm.

In this work, we use a laser light source having a sub-nanosecond pulse-width and observe laser induced fluorescence (LIF) for a pure helium plasma generated in a glow discharge at pressures from 0.001 to 0.1 atm or in a micro-hollow cathode discharge from 0.05 to 1.0 atm.

2. Experiment

Figure 1 shows a schematic diagram of the experimental set-up. A 620 ps laser pulse from a dye laser excited by a sub-nanosecond N_2 laser (Usho, KEN-X) excites helium atoms from the 2^1S level to 3^1P level. We observed LIF from n=3 levels with a monochromator (Nikon, G-250), a photo-multiplier (Hamamatsu, H6780-2, 780 ps rise time) and a digital oscilloscope (Agilent, DSO5054A, 800 ps rise time) for a glow discharge plasma (5 mm diameter and 100 mm length, 1 mA discharge current) or for a micro-hollow cathode plasma (1 mm diameter and 1 mm anode-cathode distance, 15 mA discharge current) [6].

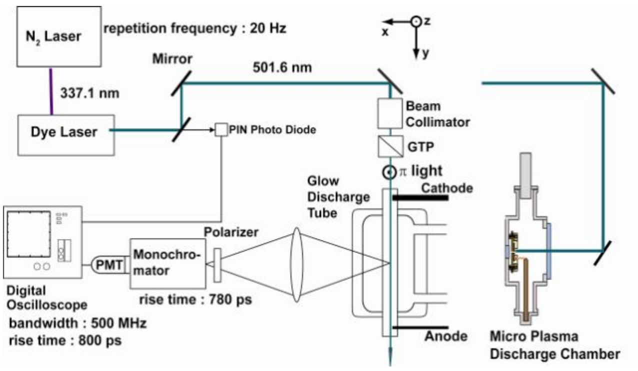

Fig. 1. A schematic illustration of the experimental set-up with a glow discharge or a micro-hollow cathode discharge

3. Results and discussion

Figure 2 shows an example of the temporal developments of the observed relative fluorescence intensities at 0.05 atm for (a) 2^1S-3^1P transition and for (b) 2^1P-3^1D, 2^1P-3^1S and 2^3P-3^3D transitions, divided by respective Einstein's A coefficients. The intensities of the 2^1S-3^1P and 2^1P-3^1D transitions are dominant, and then populations of respective upper levels are dominant.

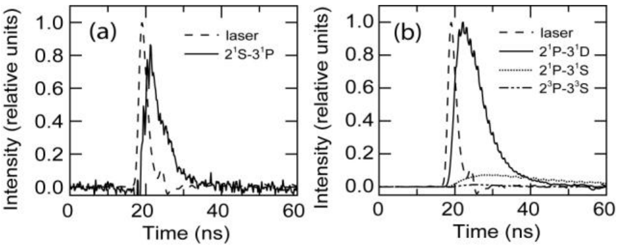

Fig. 2. Temporal developments of the fluorescence intensities of the (a) 2^1S-3^1P transition and (b) 2^1P-3^1D, 2^1P-3^1S and 2^3P-3^3D transitions.

Figures 3 (a-e) show temporal developments of the fluorescence intensities observed for the glow discharge plasma. Figs. 3 (f-h) show the results for the micro-hollow cathode plasma. In the case of the gas pressures beyond 0.1 atm we can show only the 2^1P-3^1D intensity because the 2^1S-3^1P intensity becomes too weak to examine the temporal development. In Fig. 3, each of the temporal developments is normalized to its peak intensity for all the cases. It is noted that the temporal developments of the 2^1P-3^1D intensity at 0.1 atm in the glow discharge and in the micro-hollow cathode discharge are similar to each other.

Considered with the excitation transfer and quenching of the 3^1P and 3^1D levels, the coupled rate equation for the populations in these level, $N(3^1P)$ and $N(3^1D)$, are given as

$$\begin{cases} \dfrac{\mathrm{d}}{\mathrm{d}t} N(3^1\mathrm{P}) = K(3^1\mathrm{D} \to 3^1\mathrm{P}) N(3^1\mathrm{D}) - K(3^1\mathrm{P}) N(3^1\mathrm{P}) \\ \dfrac{\mathrm{d}}{\mathrm{d}t} N(3^1\mathrm{D}) = K(3^1\mathrm{P} \to 3^1\mathrm{D}) N(3^1\mathrm{P}) - K(3^1\mathrm{D}) N(3^1\mathrm{D}) \end{cases}$$

where $K(3^1P \rightarrow 3^1D)$ and $K(3^1D \rightarrow 3^1P)$ are the excitation transfer rates from 3^1D to 3^1P and from 3^1P to 3^1D, respectively, and $K(3^1P)$ and $K(3^1D)$ are the quenching rates of the 3^1P and 3^1D levels, respectively.

The coupled rate equation can be solved analytically. With the solution and the rate coefficients due to atom collisions reported in ref. [7], we try to reproduce the temporal developments of the fluorescence intensities. The result is shown by the solid line in Fig. 3, where convolution of the instrumental time response is done. It is found that the temporal development of the 2^1S-3^1P intensity is well reproduced while that of the 2^1P-3^1D intensity is not for the gas pressures between 0.005 and 0.1 atm. The reason is not clear at present.

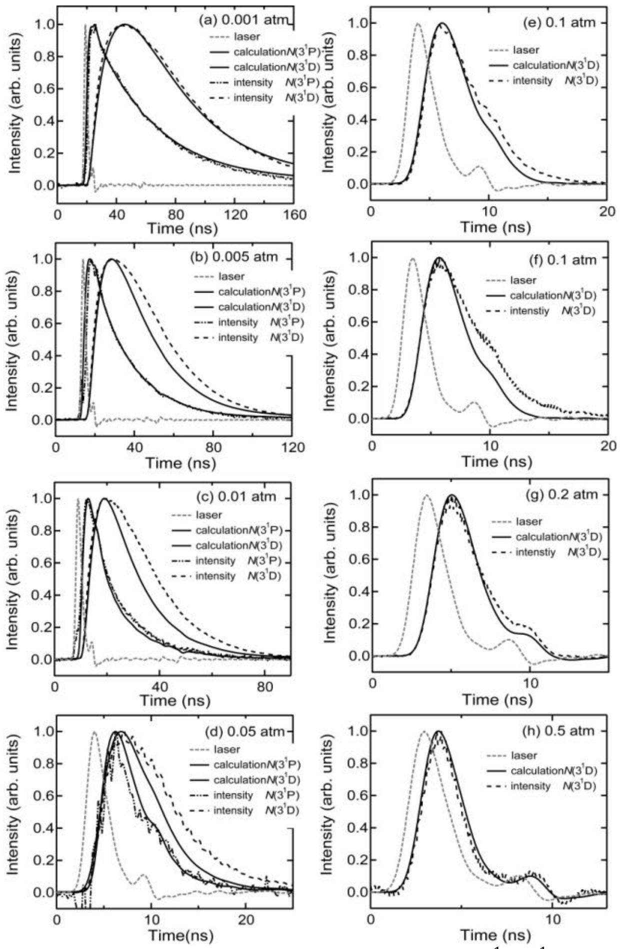

Fig. 3. Temporal developments of the 2^1S-3^1P and 2^1P-3^1D fluorescence intensities and the results of the reproduction by the solution of the coupled rate equation.

Acknowledgments

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