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Measurement of $N_2(A^3\Sigma_u^+)$ density in plasmas by cavity-ringdown absorption spectroscopy employing a diode laser

半導体レーザーを光源に用いたキャビティリングダウン吸収分光法による プラズマ中の $N_2(A^3\Sigma_u^+)$ 密度測定

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We developed a system of cavity-ringdown absorption spectroscopy employing a cw diode laser for measuring the absolute density of $N_2(A^3\Sigma_u^+)$ in plasmas. We achieved a sensitive detection limit of 10^{-7} for the absorbance. The saturation of absorption was avoided by switching off the laser beam when the cavity length was detuned slightly from the length corresponding to the perfect resonance.

1. Introduction

A problem of laser absorption spectroscopy is a narrow dynamic range and/or insufficient sensitivity for detecting species with low densities or small transition probabilities. Molecular nitrogen at the metastable $A^3\Sigma_u^+$ state is a typical species which is difficult to be detected by laser absorption spectroscopy because of the small transition probability of the first positive system, in spite of the fact that $N_2(A^3\Sigma_u^+)$ plays important roles in discharge physics as well as material processing. Our motivation for measuring the $N_2(A^3\Sigma_u^+)$ density in plasmas was to identify the most efficient species for surface nitriding of silicon [1].

Although the $N_2(A^3\Sigma_u^+)$ density is measured by laser-induced fluorescence, the calibration of the absolute density is not an easy task. Absorption spectroscopy is a better way for measuring the absolute density, but the measurement of the $N_2(A^3\Sigma_u^+)$ density by absorption spectroscopy needs a contrivance for enhancing the sensitivity. In this work, we developed cavity-ringdown absorption spectroscopy (CRDS) for measuring the absolute $N_2(A^3\Sigma_u^+)$ density [2,3].

2. CRDS system

The experimental apparatus is schematically shown in Fig. 1. Inductively-coupled plasmas (ICP) were produced in a cylindrical vacuum chamber with a diameter of 30 cm. The vacuum chamber had a quartz window at the top, and a one-turn antenna and a matching circuit were placed above the quartz window. An rf power supply at 13.56 MHz was connected to the matching circuit. Two long tubes were attached to the vacuum chamber, and the plasma was placed inside a cavity consisting of two concave mirrors with high reflectivities. The cavity length was 1 m. A piezo-electric transducer supported one of the cavity mirrors. All the optics including the mirror holders were installed inside the chamber. The mirror holders had motor-drive systems, and the angle of the mirrors were adjusted from the outside of the chamber.

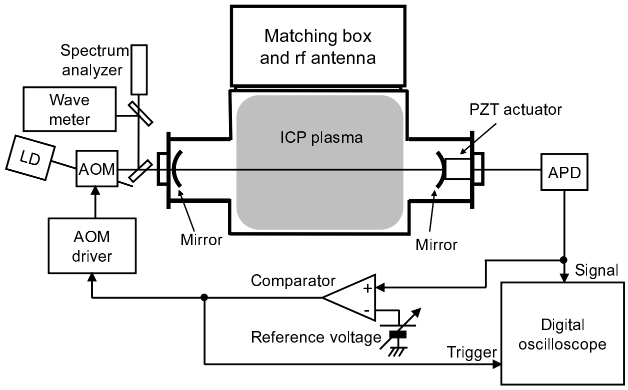

Figure 1: Experimental apparatus.

The light source was a cw diode laser. The diode laser beam was diffracted using an acoustooptic modulator (AOM). The wavelength of the diffracted laser beam was tuned around 771.1 nm, which corresponded to the $B^3\Pi_g(v' = 2) - A^3\Sigma_u^+(v'' = 0)$ absorption band of N_2 (the first positive system). The diffracted laser beam was injected into the cavity, and the intensity of the laser beam transmitted through the cavity was measured using an avalanche photo diode

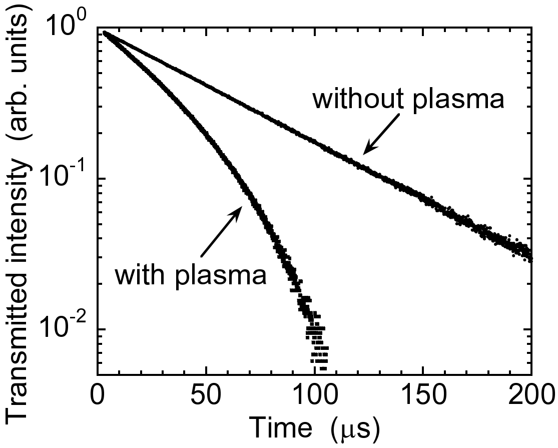

Figure 2: Decay curves of the transmitted laser intensity with and without the plasma. The laser beam was switched off when the cavity length was tuned to the almost perfect resonance.

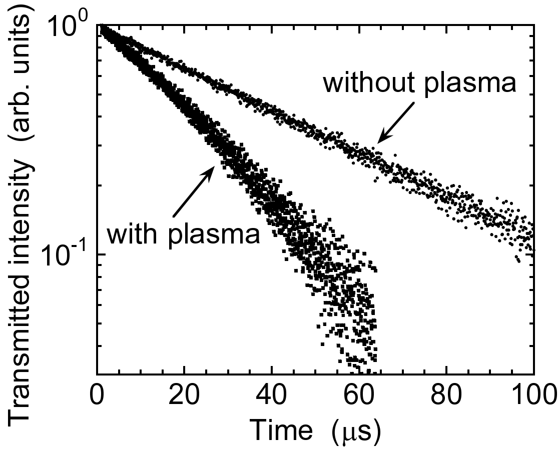

Figure 3: Decay curves of the transmitted laser intensity with and without the plasma. The laser beam was switched off when the cavity length was slightly detuned from the perfect resonance.

(APD). When the cavity length was modulated using the piezo-electric transducer, the signal from APD gave us the transmission curve of the Fabry-Perot interferometer. The resonance between the cavity length and the wavelength of the laser beam was detected using a comparator with a reference voltage. The output of the comparator was used for stopping the laser beam by switching off the AOM device. The temporal decay of the transmitted laser intensity after the truncation was recorded in a digital oscilloscope. The absorption spectrum was obtained by repeating the measurement with sweeping the laser wavelength.

3. Performance of the CRDS system

Figure 2 shows an example of the temporal decay of the transmitted laser intensity observed in the empty cavity. This decay curve was obtained by switching off the diode laser beam when the cavity length was tuned to the almost perfect resonance. The decay curve observed in the empty cavity was fitted well with an exponential function, and the decay frequency was evaluated to be $1/\tau_0 = 1.756 \times 10^4$ s^{-1}, corresponding to 99.997% for the reflectivities of the mirrors. We repeated the measurement of the decay frequency in the empty cavity for 10 times. As a result, the scattering of the decay frequency, which was evaluated by the standard deviation of the 10 measurements, was 30 s^{-1}. Therefore, the CRDS system developed in this work was compatible with a small absorbance of 10^{-7}.

We must be careful about saturation of absorption in cw laser CRDS because of the strong laser field inside the cavity. A decay curve in the presence of a nitrogen plasma is also plotted in Fig. 2. The decay curve in the presence of the plasma was not fitted with an exponential function. The decay frequency was accelerated with time. The small decay frequency in the initial part is attributed to the saturation of absorption due to the strong laser field inside the cavity. Since the laser intensity decreased with time, the saturation of absorption was moderated in the latter half of the decay curve, resulting in the acceleration of the decay frequency.

We used a simple way for avoiding the saturation of absorption in this work. Figure 3 shows decay curves with and without the plasma, when the reference voltage to the comparator was 30% of that in Fig. 2. In this case, the cavity length at the truncation was detuned slightly from the perfect resonance. In the case of the slightly detuned truncation, as shown in Fig. 3, the decay curve in the presence of the plasma was roughly fitted with an exponential function because of the weak laser intensity inside the cavity. We operated the CRDS system by adjusting the reference voltage to the comparator in order to avoid the saturation of absorption.

We measured the $N_2(A^3\Sigma_u^+)$ density successfully using the aforementioned CRDS system. The magnitude of absorbance was on the order of 10^{-5}. We also evaluated the temperature of $N_2(A^3\Sigma_u^+)$ from the Doppler broadening and the density ratio at two rotational levels. The measurement results will be shown at the conference.

References

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