Development of High Sensitive Laser Absorption Spectroscopy for Arc-Heater flow Diagnostics

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The Cavity enhanced absorption spectroscopy system using a diode laser was applied to the plasma plume diagnostics. An absorption line of ArI at 842.46 nm in an argon plasma torch plume (750W, 3slm) was targeted. Using cavity mirrors with more than 99.95% reflectance, the sensitivity was successfully enhanced by three orders of magnitude compared with the conventional single-pass laser absorption spectroscopy. The number density of 6.6 x $10^{13}$$m^{-3}$ of absorbing particles, argon $4s^2$[3/2] was detected.

Nomenclature

A	=	Einstein coefficient
c	=	velocity of light
d_0	=	absorption length
$d_{\rm eff}$	=	effective absorption length
g	=	statistical weight
I	=	absorbing state
J	=	excited state
I	=	transmitted laser intensity
I_0	=	probe laser intensity
k	=	Boltzmann constant
K	=	integrated absorption coefficient
k_ν	=	absorption coefficient
M_A	=	atomic mass
n	=	number of reflections
n_i	=	number density of absorbing state
R	=	reflectivity of mirrors
T	=	transmittance of mirrors
$T_{\rm tr}$	=	translational temperature
ν	=	laser frequency
ν_0	=	center absorption frequency
$\Delta\nu_{\rm D}$	=	full width at half maximum of Doppler profile
$\Delta\nu_{\rm L}$	=	full width at half maximum of Lorentz profile

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I. Introduction

HIGH enthalpy flow generators have been developed to simulate re-entry conditions to test the thermal protection system (TPS) since the late 1950s.[1] However, their exact plume conditions are mostly unknown because they are usually in strong thermo-chemical non-equilibrium. Therefore, it is useful to measure the chemical composition and enthalpy in these plumes for the evaluation of TPS, as well as for the validation of CFD models of non-equilibrium flows.

In our previous study, laser absorption spectroscopy (LAS) has been applied to the plumes of the arc-heaters and the inductively coupled plasma generator, and the number density of meta-stable atomic oxygen and translational temperature are measured. [2,3] In these measurement, strong absorption signals of the OI at 777.19 nm line were observed in oxygen or argon/oxygen flows. Then the number density OI at absorbing state $3s^5S$ was estimated as 3 x 10^{15} – 3 x 10^{17} m^{-3}. However, in a nitrogen/oxygen flow, the OI ($3s^5S$) number density was lower than the detectable limit of 3 x 10^{15} (fractional absorption of 1%).[4] This would be because large part of input energy was consumed in nitrogen dissociation, resulting in lower electron temperature.

In another attempt of LAS in the NASA Ames IHF (60MW) arcjet facility, [5] the absorption signal of OI was not detected in the plume of nitrogen/oxygen flows, though it could be detected inside the arc discharge region. Then, to detect the OI absorption in nitrogen/oxygen plumes, sensitivity of LAS has to be enhanced by more than two orders of magnitude.

Recently, cavity ring down spectroscopy (CRDS) has been employed as one of the high sensitive laser absorption spectroscopy.[6,7] In this method, the decay rate of light intensity inside a stable optical resonator, often called the ring-down cavity (RDC), is measured. Laser is first injected into the RDC, and is then interrupted in below μs by an acoustooptic modulator[8] or a mechanical chopper.[9] The transmitted laser decays exponentially in time. The decay rate is proportional to the total optical losses inside the RDC as a function of the laser frequency. The lower detection limits of the number density for many species have been demonstrated to be from 10^{13} to 10^{16} m^{-3}.[10]

However, for the data acquisition schemes of CRDS, the complexity and cost of digitization is high, especially in high-performance systems where a large number of bits (e.g., 12 to 14) and high speed (e.g., 100MHz) is required simultaneously.[11] To simplify CRDS and eliminate the requirement for digitization of a time-domain signal, cavity-enhanced spectroscopy (CES) was developed. CES has many different implementations.[12,13] In this study, Cavity Enhanced Absorption Spectroscopy (CEAS)[14,15], which is the simplest method in the CES methods, was applied for sensitivity enhancement. In this method, a diode laser can be used without pulsation. Moreover, it enables us to have enough amplified power, while providing detection sensitivity comparable to CRDS.

In this research, the CEAS system was constructed to diagnose an argon plasma torch flow.

II. Theory

A. Laser Absorption Spectroscopy

The intensity variation dI of a laser beam with intensity I propagating in the x direction through an absorbing medium, is expressed as follows,

$$\mathrm{d}I = -k_\nu I_0 \mathrm{d}x. \tag{1}$$

The k_ν is the function of the laser frequency ν and expressed in the following Voigt profile

$$k_\nu = K \frac{2}{\Delta\nu_\mathrm{D}} \sqrt{\frac{\ln 2}{\pi}} \int_{-\infty}^{\infty} \frac{\exp(-t^2)}{y^2 + (x-t)^2} \mathrm{d}t$$
$$x = \frac{\sqrt{\ln 2}(\nu - \nu_0)}{\Delta\nu_\mathrm{D}} \tag{2}$$
$$y = \frac{\sqrt{\ln 2}\Delta\nu_\mathrm{L}}{\Delta\nu_\mathrm{D}}$$

Number density of absorption state n_i is related to the integrated absorption coefficient K as,[16]

$$K \equiv \int_{-\infty}^{\infty} k_\nu d\nu \approx \frac{\lambda_0^{\ 2}}{8\pi} \frac{g_j}{g_i} A_{ji} n_i. \tag{3}$$

And the translational temperature is related to the Doppler width as follows.

$$T_{tr} = \frac{c^2 \cdot (\Delta\nu_\mathrm{D})^2 M_\mathrm{A}}{8 \ln 2 \cdot k\nu_0^{\ 2}}. \tag{4}$$

B. Cavity Enhanced Absorption Spectroscopy

In CEAS, only a laser beam whose frequency meets the resonance condition can transmit the RDC. Then, the absorption signal is obtained through integration of the total signal transmitted through an RDC. The integrated transmitted signal as the summation as follows,

$$I = I_0 T^2 \sum R^{2n} \exp\{-(2n+1)k_\nu d_0\} \tag{5}$$

The summation can be approximated by an integral over n (for large n) resulting in as follows

$$I = I_0 T^2 \exp(-k_\nu d_0) \int_0^{\infty} \{R\exp(-k_\nu d_0)\}^{2n} \mathrm{d}n. \tag{6}$$

Solving this expression, the following equation is obtained.

$$\frac{I}{I_0} \approx (1-R)^2 \frac{\exp(-k_\nu d_0)}{-2\ln\{R\exp(-k_\nu d_0)\}}. \tag{7}$$

Here, the mirror loss is neglected. When an absorber is placed in the cavity, the transmitted signal for frequency sweep can be obtained.

The lower measurement limit in the single-pass LAS depends on the signal-to-noise ratio. In our measurement system, it was about 1%. In this range, the following approximation is valid.

$$1 - \exp(-k_\nu d_0) \approx k_\nu d_0. \tag{8}$$

Then, the effective absorption length d_eff is expressed as follows.

$$d_\mathrm{eff} = \frac{d_0}{1-R} \tag{9}$$

The theoretical ratio d_eff/d_0 is plotted in Fig.1. The magnification by two orders of magnitude will be obtained when the transmittance of the mirrors is 1%, or the reflectance is 99%.

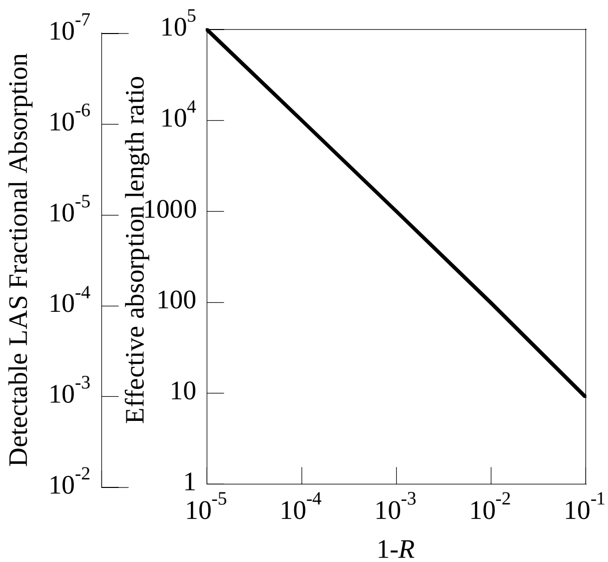

Fig.1 Theoretical effective absorption length ratio d_eff/d_0 with corresponding detectable LAS fractional absorption to R, reflectance of the mirrors.

III. Experimental Setup

The schematic of measurement system for CEAS is shown in Fig.2. A tunable diode-laser with an external cavity (EOSI DMD845) was employed which is not accompanied with mode-hops in wide modulation width. The laser frequency was scanned over about 30GHz interval at a repetition frequency of 1Hz using a function generator. High reflection concave mirrors (Layertec) whose reflectance is higher than 99.95% and curvature radius is 1m were used for constructing a RDC. A faraday optical isolator was inserted close behind the laser to reduce the optical feedback. An etalon was used whose free spectra range is 0.75 GHz to measure the frequency modulation. The emission from the plasma was shielded using a band-pass filter whose center wavelength is 840 nm and full width at half maximum is 10 nm. The transmitted laser intensity was detected by a photo detector (Thorlabs INC, DET110).

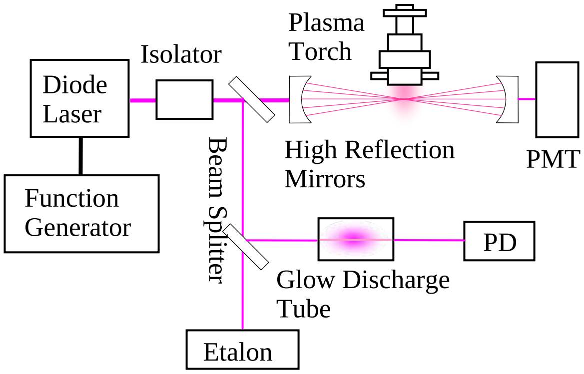

Fig.2 The schematic of measurement system for cavity enhanced absorption spectroscopy

In CEAS, if the losses in a RDC due to optical windows etc. are larger than absorption, the absorption cannot be detected. Therefore, in this research, an atmospheric plasma torch is used. The schematic of the plasma torch is shown in Fig. 3. Its exit diameter, input power, discharge voltage and argon flow rate were 2 mm, 750 W, 15 V and 3 slm, respectively.

In this research, the absorption line of ArI at 842.46 nm was targeted. An argon glow discharge tube was used to know the center absorption frequency. Its input power, discharge voltage and pressure were 2.0 W, 400 V and 80 Pa, respectively.

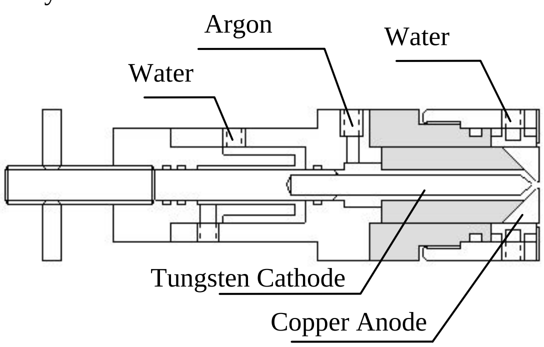

Fig.3 Configuration of plasma torch (left) and its photo in operation

IV. Result and Discussion

A. Measurement Result by single-pass Laser Absorption Spectroscopy

First, the plasma torch was diagnosed by single-pass LAS. Typical LAS signals of the plasma torch plume and the glow discharge tube plasma along with the etalon signal are shown in Fig.4. Under the atmospheric pressure, the Von del Waals (pressure) broadening, due to non-radiative interactions with other neutral species, may be significant, while under low pressure such as in a glow discharge tube, this broadening is negligibly small. Then, the absorption profile in a plasma torch is broader than that in a glow discharge tube.

The absorption profile at each distance from the exit of the plasma torch was fitted by the Voigt profile, while that of the glow discharge tube was fitted by the Gauss profile. From the fitting, translational temperature and argon ($4s^2[3/2]$) number density at 0mm were obtained as 15,700K, 6 x $10^{16}m^{-3}$ for the plasma torch plume and 610K, 2 x $10^{17}m^{-3}$ for the glow discharge tube plasma, respectively. The number density was lower in the plasma torch than that in the glow discharge tube due to quenching.

The fractional absorption distribution on the plume axis is shown in Fig.5. As mentioned above, the measurement limit on the fractional absorption by single-pass LAS is about 1%. Therefore, the absorption was not detected at the location further than 4.5mm.

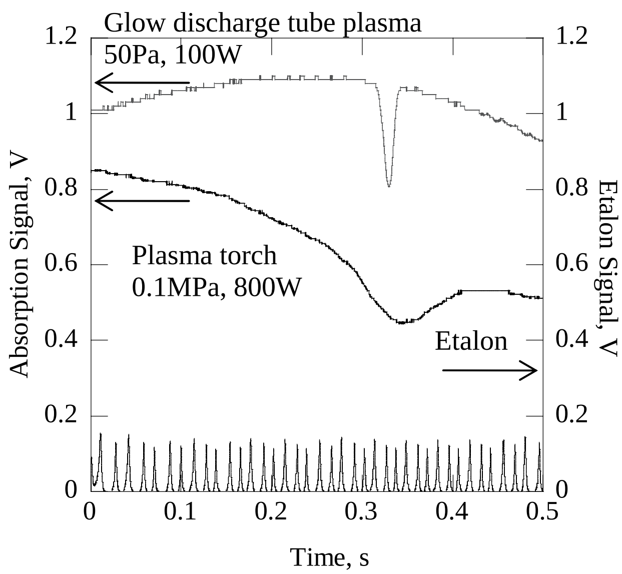

Fig.4 Single-pass laser absorption signals of plasma torch flows at 0mm distance from the exit and of the glow discharge tube plasma

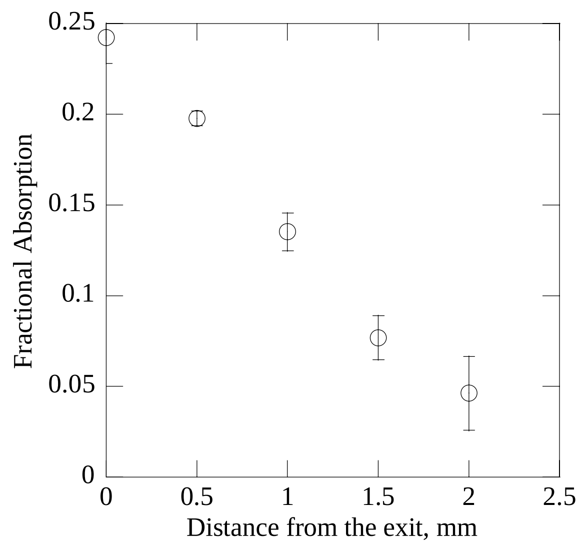

Fig.5 Fractional Absorption distribution in the plasma torch plume

B. Measurement Result by Cavity Enhanced Absorption Spectroscopy

Typical CEAS signal of the plasma torch plumes, LAS signal of the glow discharge tube plasma along with the etalon signal are shown in Fig.6. In CEAS, the transmitted laser intensity is not always constant for the mechanical vibration of mirrors. In order to improve the measurement accuracy, CEAS signal was averaged for 16 sweeps. Then, the dots in Fig.7 were obtained. A line is the theoretical curve assuming effective absorption length ratio d_{eff}/d_0 of 2550. There was no transmission around the center absorption frequency because of the large magnification of sensitivity. Because the reflectivity of the mirrors is more than 99.95%, the theoretical effective absorption length ratio of this cavity is estimated higher than 2000. Therefore measured d_{eff}/d_0 is consistent with the theory.

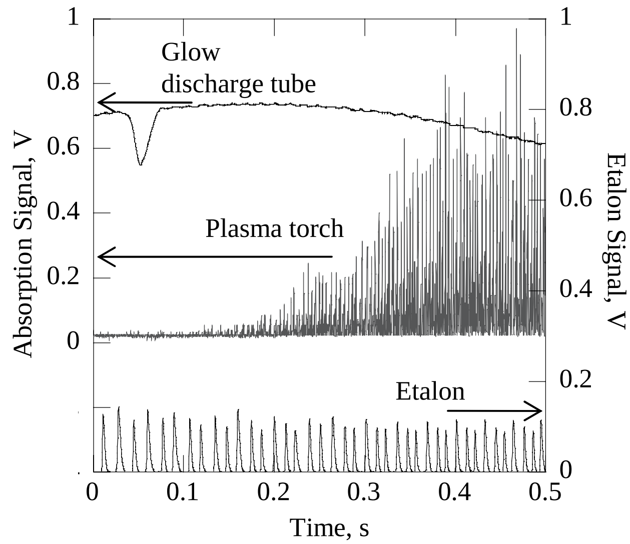

Fig.6 CEAS signal in plasma torch plumes at 2mm from the exit and single-pass LAS signal in the glow discharge tube

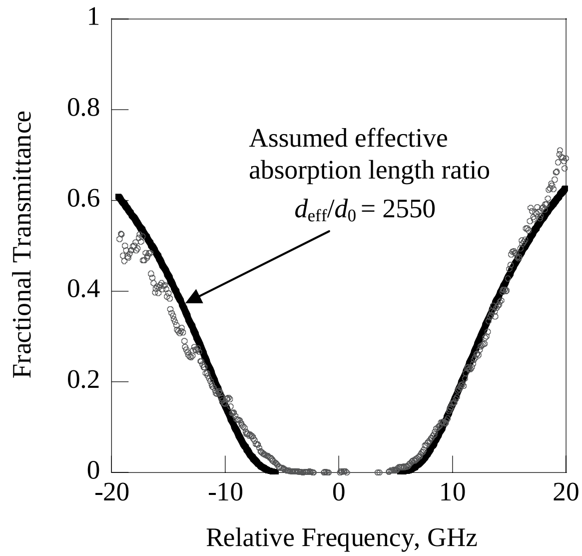

Fig.7 Averaged CEAS profile in plasma torch flows at 2mm from the exit and estimated profile with assumed effective absorption length ratio

Although the absorption signal was not detected by single-pass LAS further than 2mm from the exit of the plasma torch, it was detected by CEAS from 0 mm to 4.5 mm. However, as shown in Fig.7, the whole absorption profile from 0 mm to 3 mm was not detected. When d_{eff}/d_0 was assumed to be unchanged with the distance, the absorbance was estimated as shown in Fig.8. The fractional absorption from 0mm to 2mm calculated by single-pass LAS was also shown in this figure. By fitting the absorption profiles by the Voigt profile, the argon ($4s^2$[3/2]) number density distribution on the plume axis was calculated as shown in Fig.9. The number density from 0 mm to 2 mm calculated by single-pass LAS was also shown in the figure. The number density was found exponentially decreasing up to 4.5 mm.

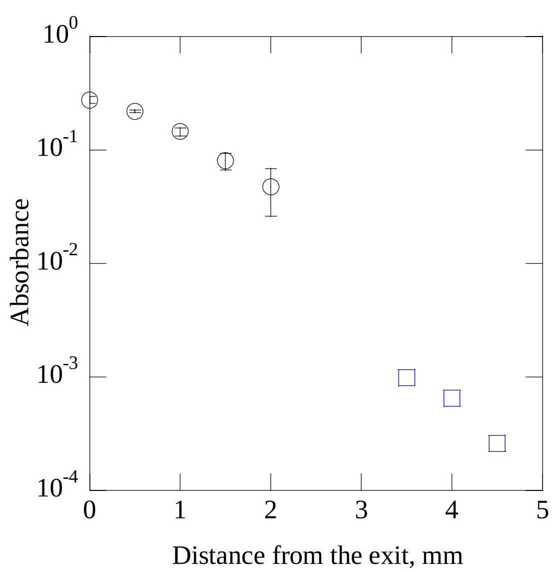

Fig.8 Absorbance distribution of the plasma torch obtained by single-pass LAS from 0mm to 2mm (open circle) and by CEAS (open square).

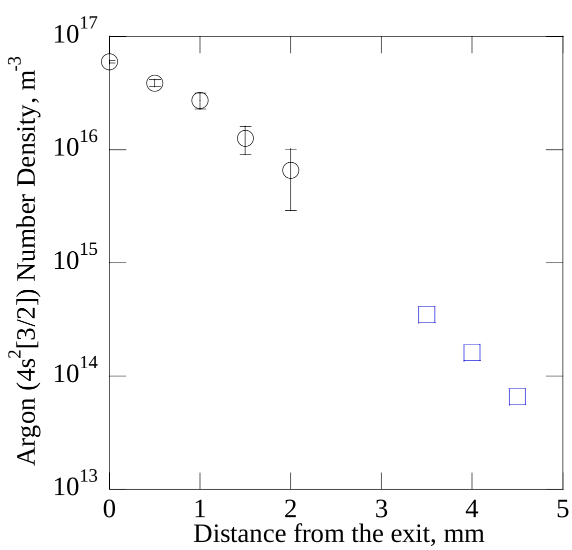

Fig.9 Argon ($4s^2$[3/2]) number density distribution in the plasma torch plume obtained by single-pass LAS (open circle) and by CEAS (open squre).

V. Conclusion

i. The Cavity Enhanced Absorption Spectroscopy system was developed using a pair of high reflection mirrors added to the conventional LAS system and applied to the diagnostics of atmospheric plasma torch flow.
ii. Compared with the conventional single-pass LAS, the sensitivity was successfully enhanced by the factor of 2550. As a result, the number density of $6.6 \times 10^{13} m^{-3}$ of absorbing particles, argon $4s^2[3/2]$ was detected.

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