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Published in:
Journal of Applied Physics

DOI:
10.1063/1.356216

Published: 01/01/1994

Document Version
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

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Download date: 20. Dec. 2018
Fabry-Pérot line shape analysis on an expanding cascaded arc plasma in argon

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(Received 23 August; accepted for publication 16 November 1993)

Fabry-Pérot line profile measurements have been used to obtain heavy particle temperatures and electron densities for an expanding cascaded arc plasma in argon. This was done for the argon 415.9 and 696.5 nm neutral lines as a function of the distance from the onset of the expansion. Temperatures in the range of 2000–12 000 K were obtained. The electron density in the beginning of the expansion appeared to be $5.6 \times 10^{21} \text{ m}^{-3}$. The 696.5 nm line profiles appeared to be asymmetric because of self-absorption by cool metastables around the plasma. The density and temperature of these metastables could be determined by fitting the measurements to a theoretical model, and appeared to be around $10^{17} \text{ m}^{-3}$ and around 3000 K, respectively.

I. INTRODUCTION: THE CASCADED ARC SETUP

Expanding cascaded arc plasmas are used for fast deposition of thin films of various kinds. Materials deposited thus far include amorphous hydrogenated carbon, graphite, diamond, as well as amorphous hydrogenated silicon. Deposition rates, obtained using this method, are far larger (i.e., on the order of 0.1 μm/s) than the ones obtained using conventional deposition techniques, such as plasma enhanced chemical vapor deposition.

To achieve a thorough understanding of the plasma deposition process, knowledge of the particle densities in the expanding plasma beam is essential. It is the aim of the present study to analyze the characteristics of the expanding plasma, sketched in Fig. 1. It consists of a cascaded arc plasma source, creating a thermal plasma (electron temperature $T_e \approx 12 \text{ 000 K}$), connected to a heavily pumped vacuum vessel (background pressure 40 Pa). The arc channel has a length of 80 mm and a diameter of 4 mm. The arc plasma expands supersonically into the vessel, creating a supersonic expansion, ending in a stationary shock, and followed by a subsonic relaxation region. The diagnostics to study this plasma jet are as follows.

**Thomson-Rayleigh scattering:** Accurate and local values for the electron density $n_e$, the electron temperature $T_e$, as well as the neutral particle density $n_0$ can be obtained as a function of axial and radial position. These measurements clearly reveal the structure of the expanding plasma jet.

**Optical emission spectroscopy:** Information about the excited level population can be obtained by means of line intensity measurements.

**Fabry-Pérot interferometry:** the set of plasma parameters $n_e$, $T_e$, and $n_0$ can be completed by measuring the neutral particles temperature $T_0$ by means of line shape analysis.

In this paper, the results of the Fabry-Pérot experiment are presented and discussed. Fabry-Pérot line shape analysis provides the opportunity to measure several important plasma parameters. To begin with, the temperature of the neutral particles in the jet can be obtained by measuring the Doppler broadening of spectral lines; it constitutes an important parameter for computer models describing the plasma jet. Further, a measurement of the Stark broadening yields the electron density: This is an independent check of the Thomson scattering data.

A third parameter, one which is more difficult to obtain, is the density of the metastable (4s) levels in, as well as around, the plasma jet. The metastable density is important for deposition experiments, as metastables may cause Penning-like ionization. Fabry-Pérot analysis of the asymmetric line profile of the self-absorbed argon 696.5 nm line gives information about both the density and the temperature of the metastables around the plasma. These data are difficult to obtain by any other (especially passive) means.

II. EXPERIMENT

A. Fabry-Pérot interferometry

The experimental setup for line shape analysis is depicted in Fig. 2. The plasma is imagined onto a monochromator entrance slit by an optical system, consisting of several lenses and mirrors. The monochromator (Bentham M300) is used as a rough wavelength selector and is set for a flat transmission bandwidth of about 0.24 nm in order to separate the line to be studied from the rest of the spectrum. After this rough selection, a parallel beam is created (lenses $L_5, f=150 \text{ mm}$ and $L_6, f=200 \text{ mm}$), in which the Fabry-Pérot interferometer is placed. The interferometer mirrors are flattened to $\lambda/100$ for the 420–500 nm wavelength region, and to $\lambda/100$ for the 650–720 nm region. Reflectivities for both wavelength regions are around 98%. Lens $L_6$ images provide the light on a 0.5 mm φ pinhole, after which it falls on a photomultiplier tube (RCA 31034). The photomultiplier is placed in a cooled housing: The working temperature is $-20 \text{ ^\circ C}$. The total (theoretical) finesse of the system is around 60 for both wavelength intervals. The free spectral ranges are $2.978 \times 10^{-2} \text{ nm}$ for the red (using a mirror spacing of 8.15 mm) and$
5.65 \times 10^{-2} \, \text{nm} \quad \text{for the blue. Apparatus profiles are measured using a low pressure argon lamp; a correction is made for the temperature of the lamp, which is known to be approximately 600 K.}

The Fabry–Pérot is a pressure-scanned type: A wavelength scan is made by varying the pressure of the gas inside the mirror cavity (in our case, neon), thus altering the refractive index of the medium between the mirrors and, hence, the wavelength which is transmitted through the system. The pressure in the Fabry–Pérot is monitored by the same software that controls the photon counting system. The software runs on a (M68000) modular laboratory computer system. A measured line profile can be processed by a least-mean-squares fit with either a Lorentzian, a Gaussian, or a Voigt profile (convoluted with the measured apparatus profile).

### III. RESULTS

#### A. Heavy particle temperatures and electron densities

The 415.9 nm line profiles all proved to be symmetric, whereas the 696.5 nm profiles showed a persistent asymmetry for measurements close to the arc (where \( n_e \) and \( T_e \) values are high). The apparatus profile, however, was always symmetric. Further, the asymmetry of the 696.5 nm emission line profile disappeared for larger \( z \) values, as well as at the edges of the plasma (measured by varying the \( x \) and \( y \) positions). The explanation for this effect will be given later on. After \( z = 30 \, \text{mm} \), the 696.5 nm profiles proved to be essentially Gaussian.

As mentioned before, one of the contributions to the line broadening is due to random motion of the atoms in the plasma, giving a Gaussian broadening in the case of a Maxwellian velocity distribution. For the half one-over-e (\( \Lambda_{1/e} \) in nm) width of the Gaussian contribution, the well-known formula

\[
\Lambda_{1/e} = \lambda_0 \sqrt{\frac{2kT_0}{m_0c^2}}
\]

holds, where \( \lambda_0 \) is the wavelength of the line (in nm), \( k \) is Boltzmann’s constant, \( T_0 \) is the temperature of the neutral particles (degrees K), \( m_0 \) is the mass of neutrals (in kg), and \( c \) is the speed of light (in m/s).

A Lorentzian contribution to the line profile is due to the Stark effect: A broadening and shift of spectral lines, caused by the random electric field of the electrons in the plasma.\(^{10}\) For most atoms (with the important exception of hydrogen) this effect is linear in the electron density. The full width at half-maximum \( \gamma_L \) (in nm) of the Lorentzian contribution is given by

\[
\gamma_L = \frac{n_e}{10^{12}} \, C_S,
\]

where \( n_e \) is the electron density in m\(^{-3}\). The constants \( C_S \) are listed by Griem\(^{10,11}\) for a large number of atomic transitions. The Stark shift of the line is proportional to the Stark width \( \gamma_L \); the proportionality constants are also listed by Griem\(^{10,11}\).

In a plasma of the kind studied here, both effects are important, whereas other effects (natural broadening, etc.)
FIG. 3. An example of a measurement (+) of the argon 451.9 nm line and its least-mean-squares Voigt fit (with residue). The horizontal axis gives only the measurement number, the interval between two neighboring points corresponds to a wavelength interval of $1.69 \times 10^{-4} \text{nm}$.

FIG. 4. The heavy particle temperature of the neutrals in the plasma jet vs the distance from the onset of the expansion (z): $\bullet$, $\circ$, $\phi$: measurements of the 696.5 nm line and reproducibility measurements; $+$: measurements of the 451.9 nm line.

FIG. 5. Electron density measurements vs axial position. Open circles denote data obtained form the Stark broadening of the argon (I) 451.9 nm spectral line, measured with the Fabry Pérot interferometer. Dots denote electron density measurements by Thomson scattering. The drawn line represents the adiabatic model by Ashkenas and Sherman (Ref. 13) for a supersonic expansion of a neutral gas through a nozzle.

The electron temperature, as determined from the width of the Lorentzian component of a measured profile, can only be accurately obtained from the 415.9 nm line profiles close to the nozzle. These line profiles show a prominent Voigt shape, and the electron densities obtained are $5.6 \times 10^{20}$ and $1.5 \times 10^{21} \text{m}^{-3}$, for $z=2 \text{mm}$ and $z=4 \text{mm}$, respectively. These values are higher than the ones obtained by Thomson scattering. However, Thomson scattering measurements close to the arc (z=2–8 mm) suffer from a large amount of stray light and are significantly less accurate than measurements with the same technique further in the expansion (for z>8 mm, the accuracy for $n_e$ is very high: about 3%–5%). Figure 5 shows the electron density measurements using both methods as a function of axial position. The solid line represents the adiabatic model by Ashkenas and Sherman; this model describes the supersonic expansion of a neutral gas through a nozzle. The values measured by Fabry–Pérot interferometry provide additional information for $n_e$ in a region where Thomson scattering becomes less accurate due to stray light. Good agreement with the adiabatic model is found.

B. Metastable densities

Until now, we have not considered the asymmetric 696.5 mm ($4p^3^3P^0 - 4s^3^S^0$) line profiles. This asymmetry of the 696.5 nm line was persistent after several new alignments, where, in each case, the apparatus profiles proved to be perfectly symmetric. Further, the 415.9 nm line profile shows no asymmetry whatsoever. Together with the fact that the 696.5 nm profiles become symmetric at larger z...
values (z > 28 mm) and at the edges of the plasma, we conclude that the asymmetry is a real, physical effect (see Fig. 6).

The following simple model gives an explanation for the observed phenomenon. We envisage the expanding plasma as consisting of two parts (see Fig. 7): a hot, fully homogeneous, central part with known $n_e$, $T_e$, $4p'$ level density, and $T_0$. This region is surrounded by a relatively cool argon gas, containing a lot of metastables. In this peripheral region, the electron density and $4p'$ level density are assumed to be negligible (Fig. 8). Both the temperature of the gas surrounding the plasma as well as the density of the $4s$ level are unknown. The distances $a$ and $b$ in Fig. 8 are determined from lateral emission scans (giving the value of $a$) and the geometry of the vessel, respectively.

Because of 696.5 nm line has a relatively large transition probability ($0.067 \times 10^8$ s$^{-1}$) and ends in a metastable $4s$ level, self absorption is to be expected. In the plasma, the self-absorption profile is the same as the emission profile: a Voigt, shifted to the red due to the Stark effect. Absorption in the cold gas surrounding the plasma, however, takes place at the unshifted wavelength, because the electron density is negligible. So the absorption profile in the periphery of the jet is a Gaussian (Doppler broadening) with its center at the unshifted wavelength.

The total intensity profile $I_{\lambda}$ is given by

$$I_{\lambda} = C[1 - \exp(-k_{p,\lambda}a)\exp(-k_{g,\lambda}b)],$$

where $k_{p,\lambda}$ and $k_{g,\lambda}$ are the absorption coefficients, embedding the $4p'$ and $4s$ densities for the plasma and the gas, respectively. The $\lambda$ dependence has the form of a Stark-shifted Voigt profile for the plasma, and the form of a Gaussian profile centered around the unshifted wavelength for the gas. $C$ is a constant.

In order to fit the measured line profiles with a least-mean squares program containing Eq. (3), one has to use data from other sources: the electron density (Thomson-Rayleigh scattering), the heavy particle temperature in the plasma (415.9 nm line profile analysis), the lengths $a$ and $b$, and the $4p'$ level density (optical emission spectroscopy). By measuring the latter, one has to in principle account for the fact that self-absorption is important (as indicated by the asymmetric 696.5 nm line profiles). In practice, however, this is not very important, as the absorption coefficients in Eq. (3) are proportional to $[(n/g)_{4s} - (n/g)_{4p'}]$ (where $g$ is the statistical weight of the level concerned). Within and outside the plasma, the $4s$ density is much larger (about a factor 100) than the $4p'$ density, whereas the statistical weights of the two levels involved are of the same order of magnitude. So, the influence of errors in $(n/g)_{4p'}$ on the line profile is small. The free parameters for the fits, then, are the $4s$ level density and the temperature of the metastable gas $T_{4s}$.

Figure 9 shows a line profile (at z = 14 mm, at the axis of the plasma jet) with its best fit. The simple model is capable of reproducing the line profiles very well. The fits thus give results for the metastable density and metastable temperature, as all the other parameters are known. The values are accurate within 30%, in view of the errors in $a$ and $b$ and the simplification by considering only two (homogeneous) zones in the vessel. In Fig. 10, the metastable temperature in the periphery is compared to the heavy particle temperature of the plasma jet. An increase of the temperature of the periphery with increasing $z$, to the temperature of the central plasma, is clearly visible. This means that the total jet is becoming more and more iso-
thermal. This is also indicated by Thomson–Rayleigh scattering measurements. The metastable densities as a function of $z$ are given in Fig. 11. Note that these reflect the $4s[4p^0]$ level densities in the periphery of the plasma (Figs. 7 and 8).

IV. DISCUSSION AND CONCLUSIONS

Fabry–Pérot measurements can extend the set of known plasma parameters ($n_e$, $T_e$, and $n_0$, Thomson–Rayleigh scattering) by adding the heavy particle temperature $T_0$. The information, however, is not spatially resolved, but always reflects a certain line of sight. This blurs some of the features so distinct in spatially resolved measurements, such as the internal structure of the (Barrel-type) shock.

The accuracy of the Fabry–Pérot measurements is estimated to be within 30% for $T_0$. This estimate was established by performing reproduceability checks on different days and by the margins of error as indicated by the fitting procedure.

The measured heavy particle temperatures are consistent with other measurements, as the fit to the heavy particle temperature at the end of the arc as well as to the electron temperature further in the vessel. Further, the background pressure in the vessel gives an extra indication of the accuracy of the measurements. If pressure equilibrium is assumed further in the vessel, the background pressure is equal to the sum of the gas pressure (which is equal to $nkT$ and is, hence, largely determined by the heavy particles in these plasmas with a low ionization degree (3%–4%)) and the stagnation pressure, which depends on the velocity of the particles. If we insert an estimated value for this speed (600 m/s after the shock) and the measured values of $T_e$ and $T_0$ further in the vessel, the sum of stagnation and gas pressures indeed matches the background pressure within 5%.

Electron density measurements using Stark broadening analysis appear to give additional information to Thomson scattering data, as the latter suffer from stray light close to the arc. The values obtained from Stark broadening of the lines are accurate within 30%, and show a good agreement with the adiabatic expansion model by Ashkenas and Sherman. This strongly supports the view that the supersonic part of the expansion very closely resembles an adiabatic expansion.

The asymmetric line profiles give a wealth of information. In order to obtain the metastable densities and temperature, we inserted all other known variables (such as the electron densities) into the fit. In principle, however, one may also use this asymmetry to determine electron densities, as the asymmetry essentially proves to be a Stark shift effect. In this case, the fitting procedure uses more free parameters. The simple model of a two-region plasma jet is capable of reproducing the asymmetric line profiles very well. The accuracies of $T_4s$ and $n_4s$ are around 30% and 50%, respectively. These margins of error are indicated by reproduceability checks and the errors given by the fitting.
procedure, as well as by uncertainties in the emission and absorption lengths $a$ and $b$ (Figs. 7 and 8).

The fitting procedures were also performed with a slightly different model, in which the metastable densities (see Fig. 8) are assumed to be constant as well, but different in regions I and II. This, however, did not significantly change the metastable densities or temperatures in the periphery.

The measured metastable densities around the plasma are substantial ($10^{17} \text{ m}^{-3}$). After a few centimeters in the expansion, these metastables are probably produced by the capture of resonant radiation from the plasma center. The temperature of the metastables around the plasma is a unique result from the line shape analysis. The metastable temperatures indicate that the plasma jet is becoming more and more isothermal, which is also indicated by Thomson scattering measurements.

ACKNOWLEDGMENTS

We would like to thank H. M. M. de Jong and M. J. F. van de Sande for their skillful technical assistance during the measurements and M. C. K. Gruijters for drawing some of the figures.