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DEVIATION FROM LOCAL THERMAL EQUILIBRIUM IN AN ATMOSPHERIC ARGON ARC PLASMA

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Abstract : The electron temperature T_e is obtained from five optically non-thin Ar I line-transitions. It is possible to measure T_e without any knowledge of transition probabilities. The electron density N_e can be calculated from measured excited level densities using the *partial* LTE-model. It appears that there is a functional dependence of N_e on T_e, suggesting that transport processes are of only minor importance. The deviations from LTE can be described by assuming a N_e-dependent overpopulation of the ground level of Ar I. This overpopulation factor varies from 1,9 for $N_e = 2 \times 10^{16} cm^{-3}$ to 1,25 for $N_e = 2 \times 10^{17} cm^{-3}$.

Introduction : During the past twenty years several criteria are developed by which it can be decided whether or not a specific plasma is in local thermal equilibrium (LTE). From theoretical considerations it appears that in laboratory-plasmas, which are considered to be in LTE, the value of the electron density N_e often is the determining factor. However, theoretical values for this minimum electron density $N_{e,LTE}$ show a large spread. (Griem /1/ : $4.10^{15} cm^{-3}$; Deutsch /2/ : $10^{18} cm^{-3}$). Recent calculations /2,3/ - with more accurate expressions for cross-sections and taking into account of radiation-absorption - yield values for $N_{e,LTE}$, which are roughly ten times larger than earlier calculations /1/. Also the experimental values for $N_{e,LTE}$ show a large scatter (Neumann /4/ : $3.10^{14} cm^{-3}$; Bober and Tankin /5/ : $10^{18} cm^{-3}$). It is our aim to give further quantitative information in this matter.

Method : In an atmospheric argon plasma, with an electron temperature T_e of about 12000 K and $N_e \simeq 10^{16} cm^{-3}$, deviations from LTE can occur due to the large energy-gap (11,5 eV) between the ground level and the first excited levels (4s-group). The coupling between the ground level and the excited levels is too small to ensure a LTE-distribution : the ground level will be overpopulated with respect to the excited levels. Further it can be argumented that, if N_e is larger than $10^{15} cm^{-3}$, the occupation of the excited levels (from the 4s-group upwards) will be in equilibrium with the free electrons. This means that for the plasma of interest ($T_e \simeq$ 1-1,5 eV, $N_e \simeq 10^{17} cm^{-3}$) the model of *partial* LTE (PLTE) holds. The radiation, originating from transitions between levels of the 4p and 4s group of Ar I (700-800 nm), is partially absorbed in the plasma. This self-absorption is a measure for the density of the 4s-level involved. The method used to determine the density of the excited atoms is based on the equation of radiative transfer. The (cylindrically symmetric) plasma is observed end-on, i.e. in a direction parallel to the axis of symmetry at a distance r from this axis. Let the axis of symmetry be the z-axis of a co-ordinate system in which the plasma extends between z=0 and z=L. The solution of the equation of radiative transfer - which gives the relation between the spectral emission coefficient $\varepsilon_\lambda(\lambda,r)$, the spectral absorption coefficient $\kappa(\lambda,r)$ and the spectral intensity of radiation in the direction of z-axis $I_\lambda(\lambda,r,z)$ - becomes with the boundary condition z=0, $I_\lambda=0$:

$$I_\lambda(\lambda,r,L) = \frac{\varepsilon_\lambda(\lambda,r)}{\kappa(\lambda,r)} \left[1-\exp\left(-\kappa(\lambda,r)L\right)\right] \qquad (1)$$

If corrections are made for the continuum emission and absorption in the vicinity of the spectral line ($q \rightleftarrows p$, $q<p$), measurements of the spectral radiance I_λ and of the spectral absorption-factor exp $(-\kappa L)$ yield the source function :

$$S_{\lambda,pq} = \frac{\varepsilon_{\lambda,pq}}{\kappa_{pq}} \qquad (2)$$

Here $\varepsilon_{\lambda,pq}$ and κ_{pq} are the spectral line emission coefficient and the spectral line absorption coefficient respectively. When $\varepsilon_{\lambda,pq}$ and κ_{pq} can be described by the same profile one obtains :

$$S_{\lambda,pq} = \frac{2hc^2}{\lambda^5} \left[\frac{g_p n_q}{g_q n_p} - 1\right]^{-1} \qquad (3)$$

Replacement of the density fraction $g_p n_q / g_q n_p$ by the Boltzmann-factor exp $(hc/\lambda kT)$ defines an excitation temperature T, which in case of PLTE equals T_e. The electron density N_e can be determined from the measured level densities using the PLTE model.

Experimental arrangement : The plasma was generated in a cascade-arc (diameter discharge-channel 5 mm; the discharge-length was 65 and 85 mm, respectively). To avoid an Abel-transformation of measured data the cylindrically symmetric arc was viewed end-on. A telecentric arrangement was used to achieve that the optical system selects a parallel beam (0,5 mm x 0,5 mm) from the radiation, emitted by the arc discharge. The opening angle in our experiment was 0,002 rad. In order to obtain the radial distribution of radiance, the arc could be displaced perpendicular to the optical axis. The spectral resolution of the radiance was achieved with a 1.0 meter monochromator. If necessary, the spectral measurements were corrected for the apparatus function (halfwidth 0,025 nm) of the monochromator. The self-sbsorption of the radiation was determined by placing a spherical mirror behind the arc in such a way that the plasma was imaged in itself. A slowly rotating chopper was placed between the arc and the spherical mirror in order to obtain the spectral emission and spectral absorption simultaneously.

Results and conclusions : Spectral emission and absorption measurements are performed for 5 Ar I lines (696,5; 727,3; 750,3; 763,5 and 794,8 nm) as a function of both the arc current (40, 80, 140 and 200A) and the relative distance r/R from the discharge-axis (r/R varies till 0,80; R is the channel radius). The plasma pressure was 1 atm. By investigation of the wavelength-dependence of the ratio of spectral emission and spectral absorption it could be verified that in all cases the emission and absorption line-profiles are identical. For the same current and position each of the 5 line-transitions yielded the same value of the temperature. It turned out to be possible to determine a temperature of 15000 K with an accuracy of 100 K without any knowledge of transition-probabilities. Fig. 1 shows the electron density N_e as a function of T_e. N_e was calculated from the 696,5 nm result, using the PLTE-model and a transition probability of 0,067 x $10^8 sec^{-1}$ (full line). The broken line of fig. 1 represents the LTE-relation between N_e and T_e for a pressure of 1 atm.

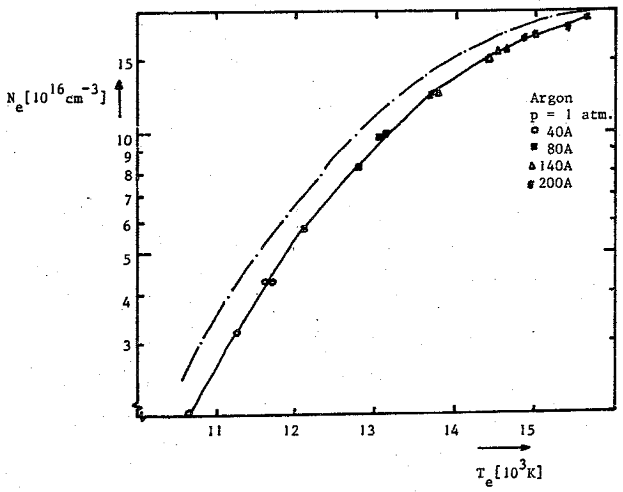

Fig. 1 Electron density N_e versus electron temperature T_e
—— the measured relation $N_e(T_e)$ for p=1 atm.
-.- the calculated relation $N_e(T_e)$ for p=1 atm. assuming LTE.

From fig. 1 it can be seen that N_e (and consequently the ground level density n_1, which can be determined from the full curve $N_e(T_e)$ of fig. 1 and the equation of state) is a function of T_e only, irrespective of the specific conditions (radius and current) at which the quantities are measured. The relationship differs from that of LTE and yields an overpopulation factor of the ground level n_1/n_1*, which varies from 1.9 at N_e = 2 x $10^{16}cm^{-3}$ to 1,25 at N_e = 2 x $10^{17}cm^{-3}$ (cf. fig. 2). Here the quantity n_1* denotes the ground level density in LTE, obtained from T_e and N_e.
This result suggests the existence of PLTE in which the overpopulation of the ground level is caused by radiative recombination processes; the transport processes are apparently only of minor importance. This interpretation of the data is supported by the results of model calculations of the Ar I-system for the case of a homogeneous and stationary plasma with optically thick resonance lines /6/. The model yields overpopulations close to and even higher than the experimentally obtained values (cf. fig. 2).

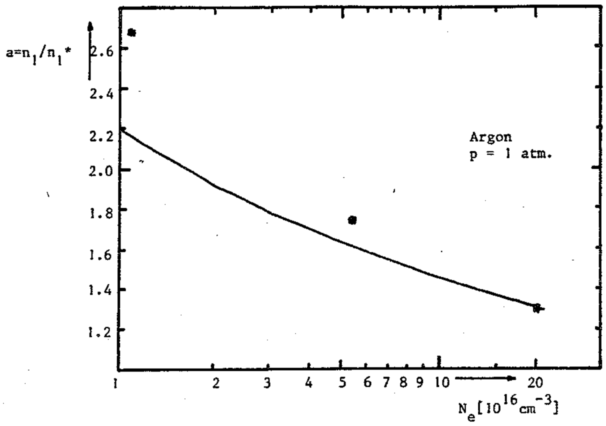

Fig. 2 The overpopulation of the ground level n_1/n_1* versus N_e
– the measured relation
** model of Katsonis*

The line measurements are carefully corrected for the continuum radiation. For the higher arc currents (140, 200A) the continuum radiation is partially reabsorbed in the plasma. The continuum absorption coefficient for λ=696,5 nm amounts to 8,5 x $10^{-3}cm^{-1}$. In the vicinity of λ=700 nm the contribution of the free-bound radiation (fb) is about of the same magnitude as the free-free radiation (ff). Assuming that the Schlüter-factor ξ^{ff} is only a very weak function of both wavelength and temperature (ξ^{ff}=1,7) the factor ξ^{fb} can be obtained. We have found ξ^{fb} = 1,65 ± 10%, which is in good agreement with the results given in ref. /7/.
In order to investigate the effect of the cold layers near the electrodes the measurements for the 696,5 nm line are done for two arc lengths (85 and 65 nm). No systematic deviations in the results of the two measurements are detected. Furthermore it appeared that the effective arc length can be given by the geometrical distance between anode and cathode. /8/.

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