Second derivative Langmuir probe measurements in Faraday dark space in Argon d.c. gas discharge at intermediate pressures

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Abstract. In a d.c. discharge tube with sectional cathodes and a common grid anode, second derivative Langmuir probe measurements were performed in the Faraday dark space in argon gas discharge at intermediate pressures. Experimental results for different radial probe positions and different distances from the cathode in axial direction are presented. It is shown that the electron energy distribution function is bi-Maxwellian. Taking into account the electron depletion caused by their sinking on the probe surface, an extension of the Druyvesteyn formula is applied for more accurate determination of the electron temperature value, \( T \), the electron density, \( n \), and the plasma potential, \( U_{pl} \), from the experimental results acquired.

1. Introduction

Gas discharges are widely used in many different contemporary technologies, such as plasma etching, plasma polymerisation, thin layer dielectric deposition, etc. General considerations, such as the minimisation of energetic bombardment of the growing film, hold true for high-quality thin film deposition, in order to avoid film resputtering and stoichiometry variation. With this aim, different approaches have been adopted in sputter deposition, such as off-axis geometry (displacing the substrate from the trajectories of the energetic particles in the plasma), and increased discharge pressure (reducing particle energy at the substrate due to thermalisation).

However, direct comparison of the working conditions in different deposition systems is difficult. In particular, for sputter deposition the potential and discharge current distribution depend upon the geometry of chamber and target assembly, the electrical bias and temperature of the substrate holder, the presence of cathode shields, shutters, etc. It is clear that detailed information on the plasma environment is of great importance for better understanding and optimisation of film growth. To optimize the parameters of the processes by numerical modeling, one needs to know the kinetics of the discharge, namely, the plasma potential, the density and the electron energy distribution functions (EEDF).

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This work is a continuation of our previous investigations [1,2,3] and aims to investigate the changes of the plasma parameters at different radial and axial positions in a multicathode – common anode discharge tube.

The classical Langmuir probe method for EEDF measurements uses the Druyvesteyn formula [4]. This method was developed for measurement at low gas pressures and small probe radii, when \( \bar{\lambda} \gg r_p + d \) (\( \bar{\lambda} \) being the mean free path of the electrons; \( r_p \), the probe radius; \( d \), the probe sheath thickness). On the other hand, for d.c. on-axis sputter deposition of thin films, a relatively high gas pressure of 100 – 400 Pa is required. For higher gas pressures, Swift [5] pointed out the effect of distortion of the results obtained by second derivative probe measurement due to plasma depletion caused by electron sink on the probe surface at a finite \( r_p/\bar{\lambda} \) ratio. In this paper, in order to take into account the effect of plasma depletion, we applied an extension of Druyvesteyn’s second derivative method to determine the plasma potential and the electron energy distribution function, respectively the electron temperature, \( T \), and the electron density, \( n \).

2. Experimental method
Langmuir probes are known for their ability to provide local measurements of the plasma parameters. The probe technique is relatively simple when all the requirements of the “classical” theory are satisfied [6]:

The EEDF, \( f(\varepsilon) \), can then be determined by using the Druyvesteyn formula [4]:

\[
f(\varepsilon) = \frac{2\sqrt{2m}}{neS} \frac{d^2I}{dU^2},
\]

where \( f(\varepsilon) \) is normalized to 1 by \( \int_0^\infty f_0(\varepsilon)\sqrt{\varepsilon}d\varepsilon = 1 \). Here \( m \), \( e \) and \( n \) are the electron mass, charge and density, \( S \) is the probe area and \( \varepsilon = eU \), where \( U \) is the probe potential with respect to the plasma potential \( U_{pl} \).

The second derivative of the probe characteristic, \( I''(U) = d^2I/dU^2 \), can be obtained in practice by superimposing a low-frequency voltage, \( \Delta U \), (differentiating signal) over the DC probe potential, \( U \), or by numerically differentiating the I-V probe characteristic. Whatever the method is, it does not give a direct measure of the value of \( I''(U) \) but, instead, a convolution of \( I''(U) \) with the instrumental function, (IF), \( \phi(U-U') \):

\[
J''(U) = \int [I''(U')\phi(U-U')]dU'
\]

(2)

So, to correctly obtain \( f(\varepsilon) \), a deconvolution procedure must be applied and knowledge of IF \( \phi(U-U') \) is required.

At differential signal \( \Delta U = V_0 \sin \omega t \) the amplitude of the probe circuit current with frequency \( 2\omega \) is proportional to \( J''(U) \). Then the normalised to 1 instrumental function is:

\[
\phi(U-U') = \begin{cases} 
\frac{8}{3\pi V_0} \left[ 1 - \left( \frac{U-U'}{V_0} \right) \right]^{3/2} & |U-U'| < V_0; \\
0 & |U-U'| \geq V_0.
\end{cases}
\]

The validity of the Druyvesteyn formula, as a means to measure the EEDF, is limited only to low gas pressures and small probe radii, when \( \bar{\lambda} \gg r_p + d \). When the pressure is raised, this inequality becomes no longer valid and distortions of the second derivative of the probe characteristic will occur due to the depletion of electrons which sink on the probe surface at a finite \( r_p/\bar{\lambda} \) ratio, as emphasized by Swift [5]. Similar results were also obtained in ref. [6,7] from a more detailed analysis of the applicability of the second derivative probe method.
To simplify our subsequent analysis, let us assume that the instrumental function is very narrow and, therefore, \( \varphi(U - U') \) can be considered a delta function, i.e. \( J''(U) = I''(U) \). Then, following [6,7], we can write an extended Druyvesteyn formula:

\[
I''(U) = Cf(eU) - C \int_{eU} K''(e,U)f(\varepsilon)d\varepsilon
\]

where \( K''(e,U) = \frac{2\psi(\varepsilon)e^2}{[e(1 + \psi) - \psi eU]^2} \), \( C = \frac{ne^2S}{2\gamma\sqrt{2m}} \) and \( \psi(\varepsilon) = \frac{r_p \ln(\sigma_p/4r_p)}{\gamma\lambda(\varepsilon)} \).

The first term in equation (3) is the well-known Druyvesteyn formula. The second term describes the effect of plasma depletion caused by charged particles sinking on the probe surface. The geometric factor \( \gamma = \frac{\gamma}{\frac{4}{3} \geq \gamma \geq 0.71} \) assumes values in the range \( 4/3 \geq \gamma \geq 0.71 \) at \( r_p/\lambda \gg 1 \), and \( \gamma = 0.71 \) at \( r_p/\lambda << 1 \). When \( r_p/\lambda \geq 1 \), the approximation for a cylindrical probe \( \gamma = 0.71 + 0.25 \lambda/r_p \) can be used.

In the case of gas pressures in the range 100 - 400 Pa, the value of the diffusion parameter is \( \psi \approx 1 \) and the EEDF is not well represented by the second derivative of the electron probe current: at small values of \( eU \), \( K''(e,U) \approx \frac{2\psi(\varepsilon)e^2}{[e(1 + \psi) - \psi eU]^2} \) and at \( e \rightarrow 0 \), \( K''(e,U) \) increases infinitely and \( I''(U) \) decreases. Thus, a significant error will result if the electron concentration is obtained by integration over the second derivative.

In addition, since \( f(\varepsilon) \) has a finite value at \( e \rightarrow 0 \), the plasma potential \( U_{pl} \) does not coincide with the potential \( U_0 \) at which the second derivative is zero.

To obviate this obstacle, a procedure was proposed [8] involving measurements with different probe radii and interpolation of the maximum position to the zero probe radius. As one can see in Figure 1, the results are satisfactory only when the width of the instrumental function is negligible and it can be considered a delta function. In the case when the influence of the IF cannot be neglected (Figure 1b), the value of the electron concentration will be overestimated.

**Figure 1a.** Model calculations with different probe radii and interpolation of the \( I''(U) \) maximum position to the zero probe radius without IF convolution.

**Figure 1b.** Model calculations with different probe radii and interpolation of the \( J''(U) \) max position to the zero probe radius with IF convolution—\( V_0=0.1 \) V.

It should be noted that the departure from the ideal \( \psi \ll 1 \) case is more noticeable in measurements in molecular gases as the values of their rotational and vibrational excitation cross-sections are one...
order of magnitude higher than the value of the elastic scattering cross-section for inert gases in the energy range of interest. As a result of the distortion of the second derivative caused by the effect mentioned above, a straightforward application of the Druyvesteyn formula for the determination of the electron concentration will yield lower values.

This is why mobility measurements are usually performed [9]. However, these experiments are complicated and give rise to substantial errors when the cross-section for elastic electron – atom collisions has a Ramsauer minimum (Ar, Kr, Xe, etc).

Bearing this in mind, we approached the problem in a different way:

In the case of a Maxwellian EEDF at non-negligible pressure and probe size, a refined procedure has been proposed and proved [1,2,3]:

a) calculations of the second derivatives using equation (3);
b) convolution of the result (equation (2)) with the instrumental function \( \phi(U - U') \);
c) obtaining the best fit with the experimental curve.

The fitting parameters are the temperature, the plasma potential and the concentration. The best fit with the experimental results was sought using the second derivative probe characteristics at a relatively high probe potential to evaluate the temperature of the electrons. The results of the model calculations with this temperature were fitted to the experimental curve using its maximum at low probe potentials. The plasma potential was evaluated by shifting the model curve along the \( U \)-axis, while the electron density was estimated by multiplying the model data by a coefficient to achieve the best fit.

When the EEDF differs from Maxwellian, a deconvolution procedure has been proposed [10].

### 3. Experimental results and discussion

The experimental set-up was described previously [11]; we will only outline here the new design of the discharge tube with radius \( R = 7 \) cm (Figure 2). It consisted of seven cathodes and a common grid anode. The system was fed by a variable high-voltage DC power supply. The measurements were conducted at a gas pressure of 220 Pa and discharge current 24 mA. To avoid gas discharge contaminants, a continuous Ar gas flow (linear velocity \( w = 0.2 \) m/s) was sustained through the discharge tube during the measurements. The Langmuir probe had a cylindrical Pt tip (radius \( 5 \times 10^{-5} \) m, length \( 4 \times 10^{-3} \) m) and was situated in the Faraday dark space at different radial and axial positions from the cathode.

![Figure 2a. Geometry of the experimental tube.](image1)

![Figure 2b. Top view of the discharge tube flange.](image2)

The differentiating with frequency \( f = \omega / 2\pi = 941 \) Hz and amplitude \( V_0 = 0.3 \) V was applied to the probe circuit. The signal with frequency \( 2\omega \) was detected by a selective amplifier, a lock-in amplifier, and measured by an ADC card in a PC.
The probe potential $U$ was changed stepwise by a potentiometer coupled to a stepping motor. The computer code allows 1 to 500 measurements of $J''(U)$ during each step of probe potential change. Recording the average value eliminated the probe circuit noise.

Figure 3a presents an experimental second derivative $J''(U)$ of the probe current (circles) for Ar gas discharge at 220 Pa at the centre ($r = 0$) of the discharge tube and at a distance $h = 2$ cm from the cathode in axial direction.

It can be clearly seen that the EEDF is bi-Maxwellian (low-energy ultimate electrons and high-energy secondary electrons), typical for the Faraday dark space in gas discharges [12]. The solid lines are the fitted results of the calculations for ultimate and secondary electrons using equation (3), convoluted with the instrumental function (equation (2)).

Figure 3a presents experimental second derivatives $J''(U)$ of the probe current at different radial probe positions $r$ and at a distance $h = 2$ cm from the cathode in axial direction.

A 5% change of the electron temperature value leads to a visible discrepancy in the best fit between the calculated and the experimental curves. We may thus accept 5% as an upper limit of the electron temperature uncertainty and (taking into account the uncertainty in the calibration of the second derivative curves) 10%, for the electron densities.

Figure 3a presents the radial distribution of the electron densities at different axial position from the cathode. It is seen that at distance 3 cm and more in axial direction from the cathode at the central part of the discharge tube the plasma parameters are uniform.

4. Conclusions
In a d.c. discharge tube with seven cathodes and a common grid anode, second derivative Langmuir probe measurements at gas pressure of 220 Pa and discharge current 24 mA were performed in the Faraday dark space at different radial and axial probe positions from the cathode.

A refinement of the second derivative Langmuir probe method at intermediate gas discharge pressures was used. Taking into account the electron depletion caused by their sinking on the probe surface, the correct values of the electron temperature, and of the electron density were evaluated.

It is shown that at distance 3 cm and more in axial direction from the cathode at the central part of the discharge tube the plasma parameters are uniform.
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