Abnormally Low Electron Energy and Heating-Mode Transition in a Low-Pressure Argon rf Discharge at 13.56 MHz

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The electron energy distribution function measured with improved energy resolution revealed a large number ($\approx 90\%$) of low-energy electrons having an abnormally low electron temperature ($T \approx 0.3$ V) resulting in a considerably lower mean electron energy than found in all published probe measurements in argon rf discharges at 13.56 MHz. With increasing gas pressure an abrupt transition to a high-temperature mode was found. The low-temperature mode and the observed transition are attributed to a change from stochastic to collisional electron heating enhanced by the Ramsauer effect.

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To make proper probe diagnostics and, in particular, electron energy distribution function (EEDF) measurements in rf excited plasmas, a few specific issues (see the review in Ref. 1, and references therein) require special attention. The most important of these are intensive sputtering of rf electrodes and the high rate of probe and probe holder contamination by the dielectric and conductive constituents of the rf electrodes, minimization of the rf voltage across the probe sheath due to oscillation of the plasma potential with respect to ground, and low-frequency plasma potential noise mainly due to drift and ripple in the rf supply voltage causing distortion in the probe characteristic similar to rf distortion. All the aforementioned factors lead to spreading (or flattening) of the probe characteristic and its derivatives, thus resulting in an excessively large electron energy deduced from such probe measurements performed in an rf discharge.

To address these three issues we have assembled the experimental arrangement shown in Fig. 1. This arrangement is a modification of a previously developed noise-suppression probe circuit with fast-pulse measurements of the probe characteristics followed by direct analog differentiation and digital signal processing.[2] This modification includes an rf filter, F, in the probe circuit tuned to the fundamental and second harmonic of the driving frequency and continuous probe cleaning by a highly negative bias between measuring pulses. More details on the probes, rf filter, supporting electronics, and processing procedure together with experimental data obtained over a wider range of discharge parameters will be presented in a forthcoming paper. Here we will concentrate on some essentially new results concerning the EEDF in an argon low-pressure capacitively coupled rf discharge.

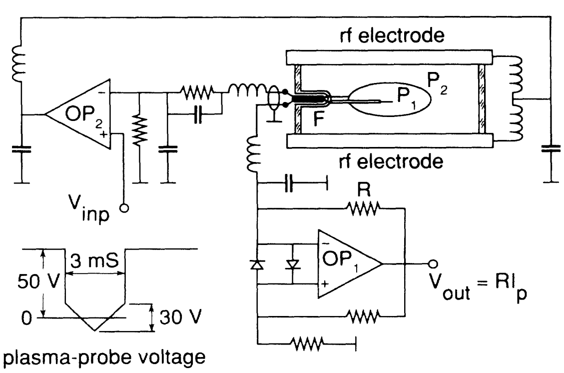

FIG. 1. Probe measurement circuit for rf-discharge diagnostics.

All measurements were made in the midplane of a parallel-plate symmetric rf discharge driven at 13.56 MHz in argon with aluminum electrodes separated by distance $L = 2$ cm and a 14.2-cm-inner-diam glass cylinder confining the discharge volume.

The absolute values of the electron energy probability function (EEPF), $f(\varepsilon)$, and the EEDF, $F(\varepsilon)$, were found directly by ensemble averaging over 1000 samples of the second derivative (d^2I_p/dV^2) of the probe current-voltage characteristics with the Druyvestein formula

$$f(\varepsilon) = F(\varepsilon)\varepsilon^{-1/2} \propto d^2I_p/dV^2 ,$$

where $\varepsilon = -eV$ is the electron energy. The mean electron energy $\langle\varepsilon\rangle$ and the plasma density n_0 were found by integrating the measured EEDF.

Typical curves of the measured EEPF and the corresponding EEDF are shown in Fig. 2 for the benchmark argon pressure of 0.1 Torr. At 0.1 Torr the measured mean electron energy, $\langle\varepsilon\rangle = 0.89$ eV, is significantly lower than $\langle\varepsilon\rangle = 3-16$ eV found from probe measurements carried out recently in argon rf discharges driven at 13.56 MHz at similar pL parameters[3-8] (p is gas pressure). The considerable difference between our results and the results of others[3-8] is presumably due to an inadequate probe system design where the issues mentioned earlier have not been properly addressed. This conclusion is supported by the fact that the dispersion in the values of $\langle\varepsilon\rangle$ obtained by other authors[3-8] is so large, and also by the observation that the largest values of $\langle\varepsilon\rangle$ were obtained in probe measurements[3,7] where no attempt was made to prevent rf probe distortion.

The EEDF in Fig. 2 differs from a Maxwellian with $\langle\varepsilon\rangle = 0.89$ eV in that it contains extra fast and slow elec-

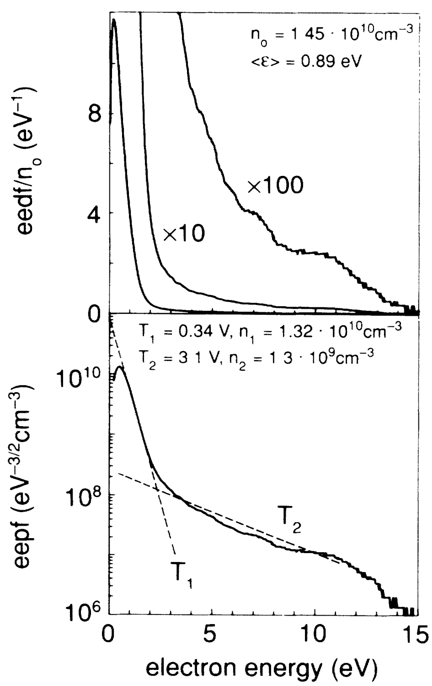

FIG. 2. The EEPF (lower) and normalized EEDF (upper), $F(\varepsilon)/n_0$, obtained for $p=0.1$ Torr and $I_d=0.3$ A rms.

trons. This feature of the EEDF in low-pressure rf discharges results from stochastic electron heating on the oscillating plasma-sheath boundary, studied theoretically[9,10] and demonstrated in rf-discharge experiments at 40.8 MHz in mercury[11] and xenon[12] and at 13.56 MHz in helium.[13] Unlike other works,[3-8] the authors[11-13] of these works minimized rf probe distortion due to the second harmonic of the plasma-potential oscillation. The difference between the EEDF obtained here and those obtained in Refs. 11–13 is the relatively large number of low-energy electrons found in argon. This makes $\langle\varepsilon\rangle$ considerably lower than that in mercury, xenon, and helium where $\langle\varepsilon\rangle$ was found to be close to those values of $\langle\varepsilon\rangle$ found in the positive column of dc discharges. Note that, in spite of the filtering of the second harmonic, the energy resolution in the second-derivative measurements[12,13] was limited to a few eV making it impossible to detect low-energy electrons.

As can be seen in Fig. 2, the EEPF obtained here can be represented as a sum of two Maxwellian distributions with two values of electron temperature, $T_1=0.34$ V and $T_2=3.1$ V, and corresponding mean electron energies, $\langle\varepsilon\rangle_1=0.51$ eV and $\langle\varepsilon\rangle_2=4.62$ eV, and plasma densities, $n_1=1.32\times10^{10}$ cm^{-3} and $n_2=1.3\times10^9$ cm^{-3}. Because of the great temperature difference and the large Ramsauer effect in argon, these two electron groups have essentially different properties and play significantly different roles in the rf discharge.

The low-energy group with its temperature close to the energy of the Ramsauer minimum has an extremely low electron-neutral (e-n) collision cross section corresponding to a low e-n collision frequency $\nu_{en1}\approx10^7$ s^{-1} and an electron mean free path $\lambda_{e1}\approx5$ cm which is much larger than the plasma half-width $d\approx0.6$ cm. These slow electrons are Maxwellian because their electron-electron (e-e) collision frequency, $\nu_{ee1}\approx5\times10^6$ s^{-1}, is much greater than the electron energy exchange frequency, $\nu_{w1}=(2m/M)\nu_{en1}\approx10^3$ s^{-1}, which is due to elastic e-n collisions only. The low-energy group originates from ionization provided by the high-energy group and oscillates collisionlessly (since $\nu_{en1}^2\ll\omega^2$) in the weak rf field, unable to gain energy either from the rf field or from the oscillating rf sheaths. Because the low-energy electrons cannot overcome the dc ambipolar potential barrier in the plasma body they cannot reach the oscillating plasma-sheath interface where stochastic heating takes place.

As a result of the Ramsauer effect the high-energy electrons ($\langle\varepsilon\rangle_2\approx10\langle\varepsilon\rangle_1$) have a large e-n collision frequency $\nu_{en2}\approx5\times10^8$ s^{-1} ($\nu_{en2}^2\gg\omega^2$) and $\lambda_{e2}\approx0.4$ cm $\approx d$. This group of electrons effectively interacts with argon atoms in elastic, excitation, and ionization collisions and compensates its energy losses through stochastic heating on the oscillating plasma-sheath boundaries. Unlike the low-energy electrons the high-energy electrons easily overcome the ambipolar potential barrier which is on the order of the mean electron energy ($\langle\varepsilon\rangle=0.89$ eV) and collide more frequently with the axial plasma boundaries, bouncing between them. During this bouncing the high-energy electrons undergo a few e-n collisions but their Ohmic heating power P_2 in the bulk plasma rf field is almost 3 orders of magnitude less than the bulk plasma power P_1 for the low-energy electrons ($P_2/P_1\approx n_2\nu_{en1}/n_1\nu_{en2}$). Although rare, elastic collisions of high-energy electrons play an important role in randomizing the phase of reflection from the sheaths, thereby providing an effective rf stochastic power transfer to the plasma electrons at the condition where ωd is too small to ensure collisionless stochastic motion in the plasma body.[10]

Compared with mercury and helium, the relatively large number of low-energy electrons in an argon rf discharge is presumably the consequence of the Ramsauer effect which may accentuate the difference between the behavior of low- and high-energy electrons. Another reason for observing so many low-energy electrons may be the use of more sophisticated probe diagnostic techniques than was done previously.[11-13]

Our measurements showed a similar shape in EEDF's for rms discharge current I_d ranging between 30 to 500 mA at a gas pressure of 0.1 Torr. For all currents we found practically unchanged values of $T_2\approx3.1$ V while the values of T_1 changed from 0.23 V for $I_d=30$ mA to $T_1=0.39$ V for $I_d=500$ mA. The change in T_1 can be explained by the effect of e-e collisions which equalize the temperatures of the two electron groups and whose importance grows with increasing plasma density.

The results found here for the EEDF are qualitatively similar to that calculated under conditions of stochastic electron heating for a 100-MHz argon discharge with $pL \approx 10^{-2}$ Torr cm but having $\langle\varepsilon\rangle \approx 5.0$ eV.[10] An excessive number of high-energy electrons has been recently found in computer simulations carried out for helium rf discharges.[14,15] The EEPF generated for a discharge[14] driven at 30 MHz and $pL \approx 0.23$ Torr cm demonstrated a rather high electron temperature with $\langle\varepsilon\rangle \approx 6$ eV, while other authors[15] found $\langle\varepsilon\rangle = 0.54$–$0.9$ eV for a discharge driven at 13.56 MHz with $pL = 0.4$ Torr cm. Although the values of $\langle\varepsilon\rangle$ differ considerably, stochastic heating was stated as the main mechanism of rf power transfer to the plasma in both simulations.

Figure 3 demonstrates the evolution of the EEPF for a fixed rf-discharge current of 0.3 A rms as the gas pressure is changed from 0.07 to 3.0 Torr. A similar evolution in the EEDF was observed at $I_d = 100$ mA. Corresponding discharge macroparameters are given in Fig. 4. The range of gas pressure was limited by probe-technique validity at the upper pressure limit and by discharge instability (prior to extinction) near 50 mTorr where the sheaths from each electrode nearly overlap.

As seen in Fig. 3, the EEPF's vary considerably in shape, being convex at high pressures and being concave at low pressures and rapidly changing from one to another at pressures about 0.4 Torr. This transition is accompanied by a corresponding sharp change in plasma density n_0 and mean electron energy $\langle\varepsilon\rangle$. For pressures $p > 0.5$ Torr, the EEPF's are Druyvesteyn-like with $\partial f/\partial\varepsilon \rightarrow 0$ as $\varepsilon \rightarrow 0$; these are typical for an argon plasma in dc or low-frequency fields ($v_{en}^2 \gg \omega^2$) with no e-e interaction.[16] At such conditions the flattening of the EEPF as $\varepsilon \rightarrow 0$ is a consequence of the Ramsauer effect when $\partial v_{en}/\partial\varepsilon > 0$ and low-energy electrons accelerate freely (no collisions) thereby escaping the low-energy region of the EEDF. An opposite picture ($\partial f/\partial\varepsilon \rightarrow \infty$) takes place at $v_{en}^2 \ll \omega^2$ when the low-energy electrons oscillate without collisions, consequently gain no energy, and thus remain in the low-energy group. Under these conditions a low-energy peak may appear in EEDF calculations that neglect e-e collisions.

In the high-pressure, or collisional-heating, regime ($p > 0.5$ Torr), Ohmic heating is the main rf power dissipation process. This heating is nonuniform along the axis due to plasma inhomogeneity and is concentrated near the plasma boundaries thus providing enhanced ionization there.[17,18] As the gas pressure decreases, stochastic heating quickly becomes the dominant electron heating process. The rf power density associated with stochastic heating P_{st} can be expressed in terms of the surface resistance R_{st}:[19] $P_{st} = J_d^2 R_{st}$, where J_d is the rms discharge current density, $R_{st} = 8\pi v_{th}/\omega_{eb}^2$, v_{th} is the electron thermal velocity, and ω_{eb} is the electron plasma frequency corresponding to the plasma boundary density $n_b = h\langle n\rangle$ with $\langle n\rangle$ being the average density over the bulk plasma. The bulk collisional plasma resistance is $R_v \approx 4\pi v_{th} 2d/\langle\omega_e\rangle^2 \lambda_e$ and the total discharge power density P transferred to the electrons can be written as fol-

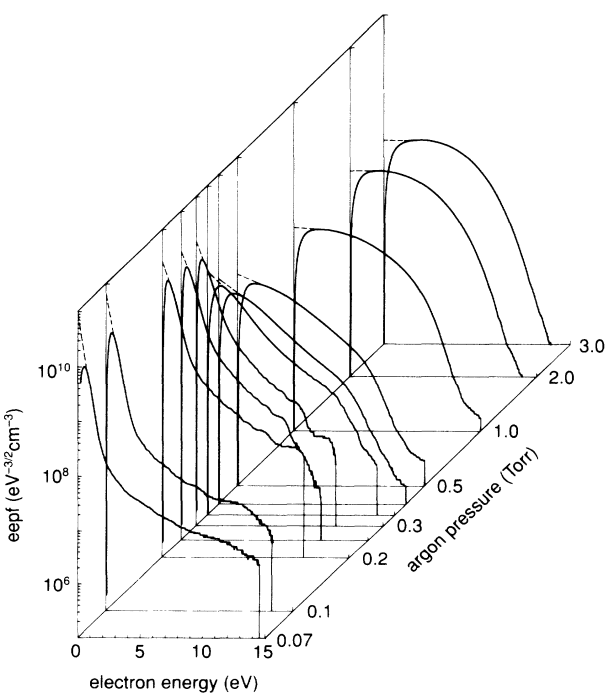

FIG. 3. The EEPF evolution with changing argon pressure, $I_d = 0.3$ A rms.

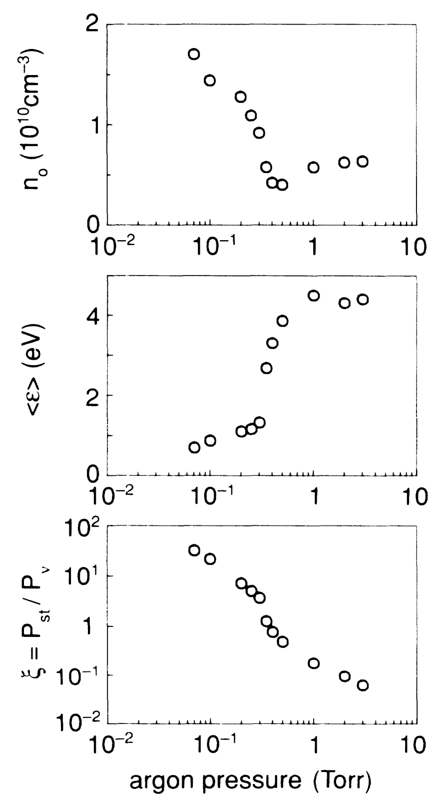

FIG. 4. Plasma density n_0, mean electron energy $\langle\varepsilon\rangle$, and ratio of stochastic to collisional power ξ vs argon pressure at fixed $I_d = 0.3$ A rms.

lows:

$$P = J_d^2(R_{st}+R_v) \approx 8\pi v_{th} J_d^2(1+hdN\sigma)/\omega_{eb}^2$$
$$= 8\pi v_{th} J_d^2(1+\xi^{-1})/\omega_{eb}^2 \,,$$

where N is the atom density, σ is the e-n elastic cross section, and $\xi = P_{st}/P_v$ is the ratio of stochastic to collisional rf power. As follows from the rf-discharge energy balance,[19] the addition of stochastic heating at the plasma boundaries leads to a decay in the bulk rf field, $E = E_v(1+\xi)^{-1/2}$, where E_v is the rf field in absence of the stochastic heating. Since $\xi \propto \sigma^{-1}$ and in argon[20] $\sigma \propto \varepsilon^{4/3}$, the drop in the bulk rf field reduces the electron energy and as a result reduces the collisional bulk electron heating even more. This kind of positive feedback provides an abrupt transition when the discharge changes from collisional to stochastic heating as the gas pressure goes down. One has to expect that the threshold pressure p_0 where this transition occurs is at $\xi \approx 1$. The parameter ξ is shown in Fig. 4 as a function of gas pressure. The values of ξ have been calculated using experimental data for d, N, and $\sigma(\langle\varepsilon\rangle)$.[20] The value of h is nearly independent of gas pressure[18] and was estimated to be $h = 0.25$. As one can see in Fig. 4, $\xi \approx 1$ at $p = 0.35$ Torr which corresponds well with p_0 at which the observed EEPF changes shape.

The transition from collisional to stochastic mode results in changes in the dependences of n_0 and $\langle\varepsilon\rangle$ on gas pressure. Thus for an rf or dc discharge controlled by collisional heating with a fixed current, plasma density grows and the effective electron temperature $T_{ef} = 2\langle\varepsilon\rangle/3e$ falls with increasing gas pressure. The opposite (although consistent with ionization balance) behavior of $n_0(p)$ and $\langle\varepsilon\rangle(p)$ can be seen in Fig. 4 for the stochastic heating mode.

Note that some decrease in T_{ef} followed by growth in T_{ef} as the gas pressure decreased monotonically was found in a xenon rf discharge[12] at a value of pL where stochastic heating was negligible. This observation seems to be confirmed by the authors[12] calculation of the EEDF for a xenon plasma in a homogeneous (no stochastic heating) rf field. Both experiment and modeling[12] showed no abrupt transition in the shape of EEDF.

A thresholdlike onset of high-energy electrons arriving at the grounded rf electrode was observed in an argon rf discharge driven at 13.56 MHz.[21] Although this effect was assumed in this work to be the result of a γ process at the powered rf electrode, the threshold value of p_0d found in this work coincides within 20% with the corresponding p_0d obtained in the present work. As to the possibility of a transition of a capacitive rf discharge into the γ regime which also may appear thresholdlike and is accompanied with the fall in electron temperature,[22-24] such transitions can be achieved only at rather large p/ω together with sufficiently high rf sheath voltages to satisfy the condition for secondary electron multiplication in the sheath.[22] The experiments reported in our work were conducted far from these conditions, although we have measured the EEDF's for discharges operating in the γ mode as well.

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