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Do the ion velocity distribution functions depend on the ion charge state?

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Abstract

There are divergent results in the literature on the (in)dependence of the ion velocity distribution functions on the ion charge states. Apparently, most time-of-flight methods of measurements indicate independence whereas most measurements with electrostatic analyzers state the opposite. It is shown here that this grouping is coincidental with investigations of pulsed and continuous arcs. All results can be consolidated by taking ion-neutral interaction into account, especially charge exchange collisions with the metal neutrals produced by the arc itself. The velocity distribution functions are independent of charge state when produced at cathode spots but become charge-state dependent when the plasma interacts with neutrals.

It is well known that the ion velocities in cathodic vacuum arcs are supersonic with respect to the ion sound velocity. Extensive measurements have been done by many authors using a range of methods. Focusing on the methods that can resolve charge and/or mass and provide information on distribution functions, one usually employs either (i) electrostatic field methods,[1-4] or (ii) time-of-flight (TOF) techniques.[5-9] There is considerable divergence in the published data. Generally, researchers using electrostatic methods came to the conclusion that the energy distribution functions, at least in part, depend on the charge state of the ion species under consideration, whereas TOF methods stated that the velocity distribution functions are independent of the charge state.

In this contribution, we briefly consider the differences of the methods, report about an experiment using an instrument with an electrostatic energy analyzer, and offer a new explanation consolidating the divergent results.

All of the techniques and experimental circumstances should be examined to determine the reasons for the variance in the results. Let us start with the TOF methods. In the simplest case, no charge or mass resolution is used, and one can simply measure the drift time of the plasma flow over a given macroscopic distance, giving us an *average velocity of ions in the flow direction*. To create time markers, the plasma production can be either kept short,[5] or modulated by oscillating the arc current,[9] or by superimposing short current spikes,[10] or by a rapid (crow-bar) switch-off.[11] In the latter case, it was shown that the derivative of the ion current contains information on the *charge-state-averaged velocity distribution function in the flow direction*. Using a 2-m-long plasma drift TOF section, a comprehensive collection of charge-state-averaged velocities was created.[9] By combining a short plasma drift section and an ion beam time-of-flight

system, mass/charge resolution was added to determine the *average velocity of ions in the flow direction.*[8,12] It was shown that for this pulsed system with short plasma drift section, ions drifted from the cathode to the ion extractor with about the same speed, regardless of their charge.

Electrostatic techniques come in various embodiments such as the retarding field analyzer[13] (RFA), or curved sector analyzer (like the EQP by HIDEN Ltd), or cylindrical mirror analyzer (CMA, like the PPM422 by Pfeiffer Vacuum). The RFAs have the disadvantage that they cannot discriminate between ions of different masses and charges. The so-called plasma monitors combine the electrostatic energy analyzer with a mass spectrometer, which, strictly speaking, does not measure mass but the mass-to-charge ratio. Typically, there is an entrance mesh (for RFAs) or a single, small entrance aperture (for plasma analyzers). The potential difference between plasma potential and the entrance plane produces a sheath in which positive ions are accelerated ($V_{pl} > V_{entrance}$) or decelerated ($V_{pl} < V_{entrance}$). For $V_{pl} > V_{entrance}$ the energy gain is

$$\Delta E_{kin}(Q) = Qe\left(V_{pl} - V_{entrance}\right) \tag{1}$$

provided that there are no collisions in the sheath; Q is the charge state number and e is the elementary charge. Often, $V_{entrance}$ is the ground potential and set to zero. If we want to infer about the kinetic energy distribution functions *in the plasma* in the direction of measurement, one needs to subtract this energy gain. The situation $V_{pl} < V_{entrance}$ is not as simple because only the energetic fraction of the distribution, i.e. ions with $E_{kin} > \left|Qe\left(V_{pl} - V_{entrance}\right)\right|$, can reach the analyzer entrance, and furthermore the sheath

may not be stable.[14,15] This also applies to the situation when the entrance is grounded and the plasma potential is negative.

We will focus now on the fact that TOF experiments generally gave results at variance to those obtained by electrostatic experiments. Therefore, we executed an ion energy experiment using an electrostatic method on a plasma similar to one previously investigated with a TOF technique.

The plasma was produced by a cathodic arc plasma source of the "minigun" type[16] operating in repetitively pulsed mode. Arcs were "self-triggered" by applying the full charging voltage of the pulse-forming-network (400 V) between cathode and anode.[17] The arc pulses were about 350 μs long with 200 A of discharge current fed by a pulse-forming-network and switched by a high current thyristor. Aluminum is a popular cathode material and so we decided to use this material, allowing us to compare our results with the literature.

The cathodic arc plasma was streaming towards the grounded entrance of a differentially pumped plasma analyzer (EQP by HIDEN Ltd., operating in positive ion mode), placed about 25 cm away, see Fig. 1. The diameter of the entrance aperture was 100 μm. The vacuum in the discharge chamber was about 10^{-4} Pa and about 10^{-5} Pa inside the analyzer.

Data acquisition with the EQP analyzer was synchronized with the pulsed arcs (1 pulse per second) by using the analyzer's external synchronization feature. In this mode, the analyzer is on standby when the plasma is not present (the synchronization port sees 0 V) and resuming work when the plasma is on (synchronization port sees 5 V). The data acquisition was done over the entire arc pulse lengths.

Each individual measurement of the energy distribution function was characterized by a large noise on the signal, which is quite common for cathodic arc plasmas. In order to obtain more a representative distribution curve, each measurement was repeated five times and the raw data curves were averaged. Figure 2 shows the averaged curves for mass (actually mass/charge) setting 27, 13.5, and 9, corresponding to Al^+, Al^{2+}, and Al^{3+}, respectively. The analyzer's software uses "energy" on the abscissa but it is the internal voltage used to set the ion energy for passing the energy filter, and therefore the scale corresponds to energy/charge. Figure 3 shows the same data but multiplied with the charge state number, indicating that the energy distribution functions (in the direction of observation) have roughly the same shape and the same average value for the different charge states. A more careful consideration indicates that the Al^+-curve is approximately Gaussian while the other (Al^{2+} and Al^{3+}) can be approximated by a shifted Maxwellian distribution as observed with other experiments[3,4,18],

$$f_i(E) = C_s\left[E - \Delta E_{kin}\right]\exp\left[-\left(\sqrt{E - \Delta E_{kin}} - \sqrt{E_{dir}}\right)^2 \Big/ kT\right] \qquad (2)$$

where C_s is a scaling constant, ΔE_{kin} is the kinetic energy gain given by Eq.(1), and E_{dir} is the average directed ion energy measured on the axis of the measuring instrument. We assumed that the plasma potential is very close to ground and therefore no attempt was made to correct for ion acceleration in the sheath, i.e., $\Delta E_{kin} \approx 0$.

The present results obtained with an electrostatic analyzer support earlier findings obtained with the time-of-flight technique,[5,8,12] namely that there are no significant differences in the drift velocity (or equivalent kinetic energy) for the different charge states. Therefore, it is not the kind of the measuring technique that leads to the divergent

results, rather, the grouping of results with the measuring techniques is coincidental with other conditions.

At first one would think of different plasma potentials, which would lead to different acceleration in the sheath before the analyzer entrance. This, however, turns out not to be the main culprit. As we will show, the main issues are the ion-neutral interactions and the time-dependent presence of neutrals in the path of the plasma flow.

Let us start with a curious TOF observation by Tsuruta and coworkers[5] who found that the ion velocities are independent of charge states for the 15 μs long arc pulse unless one measures at a later time when the plasma already decays. Again using a TOF technique, Yushkov[12] observed that neutral gas (air, N_2, Ar, Xe) with a pressure of 50 mPa ($4x10^{-4}$ Torr) can change the velocity distribution function of copper ions in a *charge state dependent manner*: lower charge states suffered greater velocity reduction than higher charge states. Similar results were recently seen with aluminum ions in oxygen and argon background gas.[19] Most of the velocity studies in the literature were done at much lower pressure than 50 mPa. Pulsed arcs are usually in high vacuum of 10^{-4} Pa (~10^{-6} Torr) and DC arcs either also in high vacuum or with low background pressure of Ar to stabilize the arc at low current. Therefore, one would not readily assume that the neutral background is sufficient to create large effects.

However, neutrals are produced by the condensing arc plasma itself. In a work on energetic condensation it was shown that the cathodic arc ions have a significant rate of self-sputtering.[20] Furthermore, the sticking coefficient is much smaller than unity if energetic ions impact the surface at large angles with respect to the surface normal. Therefore, self-sputtering and limiting sticking are large sources of *metal* neutrals that

lead to a time-dependent filling of the volume in which the plasma flows. This time-dependent neutral background was already identified as responsible for the evolution of ion charge state distributions.[21] Depending on the geometry of the chamber and electrode arrangement, it takes about 100 μs and more to establish a (noisy) steady-state density, a time that coincides well with the filling time of the chamber with metal neutrals.[21] Additional sources of metal neutrals are evaporating macroparticles and formerly active, still-hot locations of cathode spots.

With this idea at hand, let us re-visit the literature. It turns out that all reports on the independence of the velocity on the charge state were obtained with pulsed plasmas and short drift sections, i.e. under conditions when the plasma coming from the cathode spot does not yet encounter many neutrals. All investigations using continuous arcs reported some sort of charge state dependence, but often not quite proportional to the charge state. In continuous system, the neutral density is steady-state but fluctuating because there is a balance of supply and removal.

Some experiments with pulsed systems also resulted in charge-state-dependent yet less-than-charge-proportional results. The data of Davis and Miller[2] where obtained with a detector placed at the end of a long, relatively narrow tube through which the plasma had to flow, i.e., conditions typical for oblique angle condensation having a low sticking coefficient. Therefore, one can expect that large amounts of neutrals were present despite careful backing of the apparatus.

Ion-neutral collisions reduce the average charge state[22,23] which is particularly effective for charge-exchange collisions,[21]

$$M^{Q+} + M \rightarrow M^{(Q-1)+} + M^{+} \tag{3}$$

where “M” stands for “metal.” Neutrals produced from ions by self-sputtering or low sticking effects are less energetic that the original ions. Charge-exchange collisions produce ions of reduced charge states that have lower energy than ions of the same charge state coming directly from the cathode spot. This mechanism leads to a charge-state dependent shift towards lower energy as the interaction with neutrals proceeds. The highest charge state, Al^{3+} in our experiment, is strongly affected in the sense that those ion suffering collisions are removed from the population. What we detect are the remaining ions, and therefore we see only little change in terms of energy for Al^{3+} but a reduction of kinetic energy for the lower charge states.

In summary, we can conclude that the divergence of data on ion energies obtained by time-of-flight versus electrostatic techniques is coincidental with the choice of pulse versus continuous arcs. It is not the method of measurements but the presence of neutrals in the plasma flow path that creates the conditions for diverging results. In the presence of neutrals, which in large parts are created by the plasma itself, charge exchange collisions supply lower charged ions with reduced energy. The velocity of ion in the “original” plasma near cathode spots does not depend on the charge state, which points to a gas-dynamic mechanism of ion acceleration which is based on pressure gradients and electron-ion coupling. The charge state dependence observed with continuous arcs is not the result of charge state dependent acceleration, as widely assumed, but due to charge state dependent deceleration caused by charge exchange collisions.

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Figure Captions

FIG. 1 Experimental setup with a pulsed cathodic arc plasma source of the "minigun" type, ejecting aluminum plasma towards the entrance of a differentially pumped plasma analyzer.

FIG. 2 Raw data of distribution function for "mass" settings of 27, 13.5, and 9, versus analyzing voltage, corresponding to Al^{+}, Al^{2+}, and Al^{3+} ions, respectively. Each curve represents the average of five individual measurements.

FIG. 3 Data of Fig. 2 but multiplied with the charge state number, indicating that the ion energies are approximately independent of the charge state.

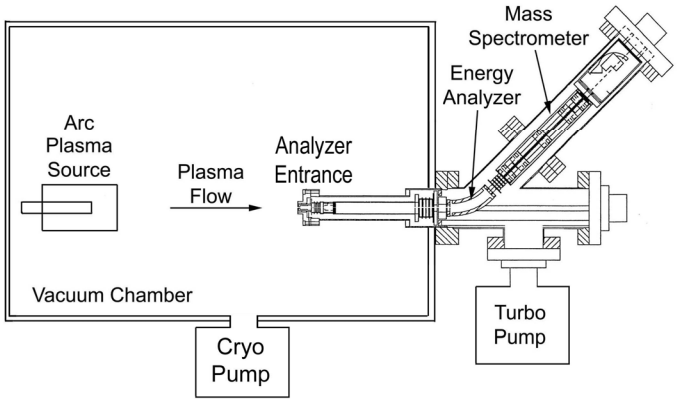
Mass
Spectrometer
Energy
Analyzer
Arc
Plasma
Source
Plasma
Flow
Analyzer
Entrance
Vacuum Chamber
Cryo
Pump
Turbo
Pump

FIG. 1

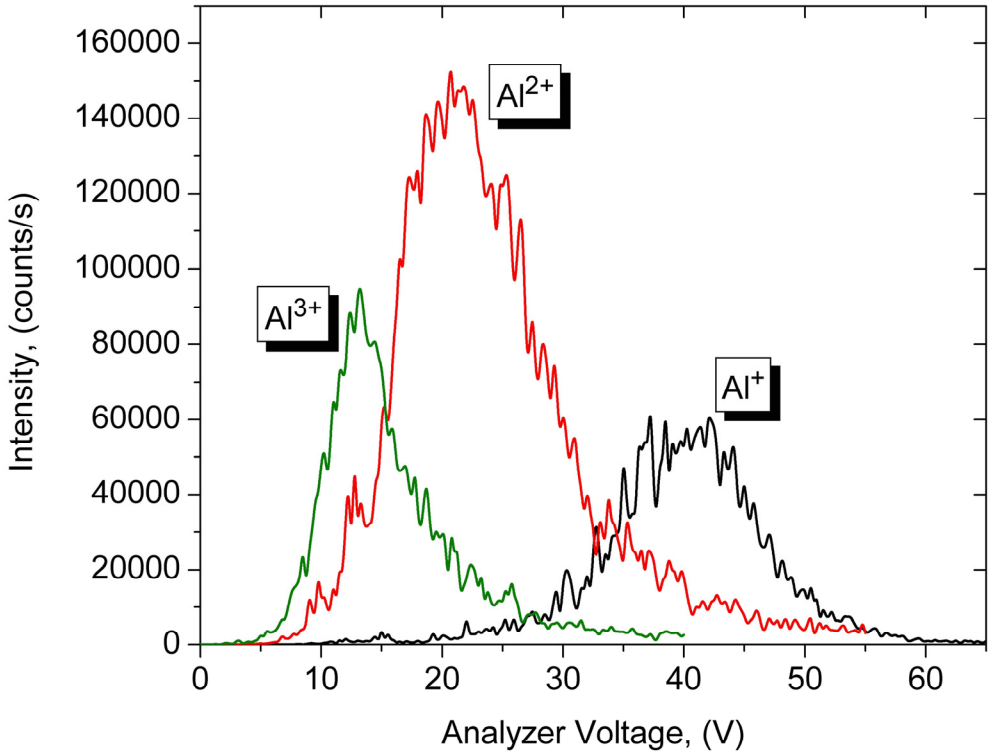
160000
140000
120000
100000
80000
60000
40000
20000
0
Intensity, (counts/s)
0
10
20
30
40
50
60
Analyzer Voltage, (V)
Al2+
Al3+
Al+

FIG.2

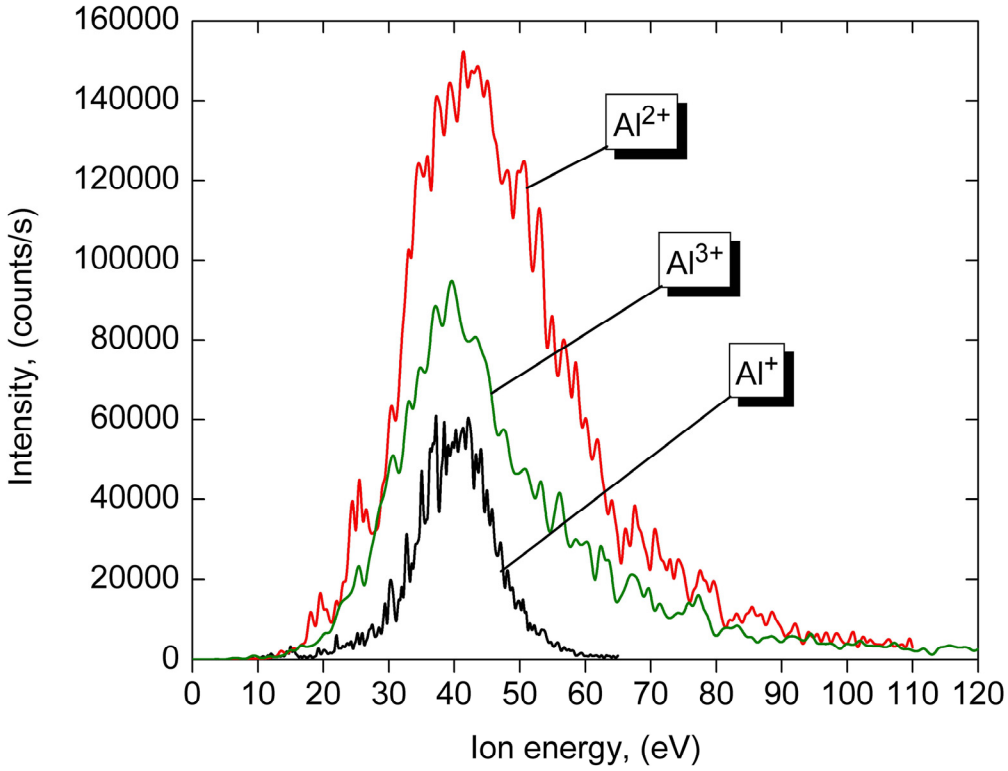
160000
140000
120000
100000
80000
60000
40000
20000
0
Intensity, (counts/s)
0
10
20
30
40
50
60
70
80
90
100
110
120
Ion energy, (eV)
Al2+
Al3+
Al+

FIG. 3