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Citation: AIP Conference Proceedings **1655**, 020004 (2015); doi: 10.1063/1.4916413
View online: http://dx.doi.org/10.1063/1.4916413
View Table of Contents: http://aip.scitation.org/toc/apc/1655/1
Published by the American Institute of Physics

Temperature Dependence of the Work Function of Caesiated Materials under Ion Source Conditions

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Abstract. The key parameter for the performance of negative hydrogen ion sources based on surface conversion is the work function of the converter surface. In order to enhance the negative ion yield caesium is introduced into the source which lowers the converter work function upon adsorption. However, the Cs layer is subject to background pressures of 10^{-7}–10^{-6} mbar and to H_2 or D_2 low-temperature plasmas which can have a major impact on the resultant work function. In order to determine the work function of caesiated surfaces under ion source relevant conditions and to identify influencing parameters, systematic investigations are performed at the laboratory experiment ACCesS. It is confirmed, that the work function of a sample surface in a vacuum of 10^{-6} mbar is decreased upon the evaporation and adsorption of caesium. However, the adsorbed layer is determined by the formation of Cs compounds with residual gases from the background pressure leading to an increased work function by about 0.6 eV compared to pure Cs layers. Elevated surface temperatures are beneficial for cleaning and against degradation in absence of Cs evaporation, but no further enhancement of the work function can be achieved by temperatures up to 260 °C if a Cs flux onto the surface is already present. Thus, the minimally achievable work function of a stainless steel surface in ion source relevant vacuum conditions is 2.75 eV. The influence of the substrate material and plasma surface interaction will be investigated next.

INTRODUCTION

Negative hydrogen and deuterium ion sources based on the surface conversion mechanism require a low work function converter surface. For this purpose the converter is covered with the alkali metal caesium which reduces the surface work function χ and thus significantly increases the negative ion yield [1, 2]. Quantified values for the reduction of χ for substrates covered with fractional Cs layers can be found in several publications for ultra-high vacuum conditions (background pressure below 10^{-9} mbar) depending on the substrate material and its crystallographic orientation at the surface [3–5]. Independent of the actual substrate a comprehensive trend can be observed: for Cs coverages below 0.5 monolayers χ decreases until at about 0.5–0.7 monolayers a minimum is obtained achieving an electron work function of about 1.5–1.7 eV. For covering fractions of a full monolayer and above the bulk value of a pure Cs surface of 2.14 eV [6] is attained.

In ion sources, however, the converter surface is exposed to background pressures of typically 10^{-7}–10^{-6} mbar and to H_2 or D_2 plasmas with electron temperatures of 1–2 eV, electron and positive ion densities of 10^{16}–10^{17} m^{-3} and atomic hydrogen densities of 10^{19} m^{-3}. In this case, the Cs layer and thus its work function is affected by the formation of Cs compounds upon the converter surface due to the presence of a non-negligible amount of residual gases. These compounds degrade the work function by their non-metallic nature. On the other hand, the low-temperature hydrogen plasma leads to a flux of reactive plasma species (atomic hydrogen H and positive hydrogen ions H_x^+) and UV radiation onto the surface. The plasma surface interaction most probably results in a cleaning of the caesium layer and thus in a decrease of the work function [7]. Due to this multitude of influences, values of measurements performed at ultra-high vacuum conditions cannot be applied directly to ion sources and dedicated studies have to be performed.

Some measurements of the work function of the converter surface in caesiated negative ion sources exist [8–12]. However, systematic and reproducible studies on parameters affecting the work function under ion source relevant conditions both in plasma and vacuum, like Cs evaporation rate, surface temperature or bias voltage, have not been performed to the author's knowledge. These parameters and especially their interplay mainly determine the stability and reliability of the surface work function and are thus crucial for the performance of the negative ion source and not least for the Cs consumption. Hence, investigations based on reliable absolute work function values are required, geared towards the identification of the influencing parameters.

Since in-situ measurements at ion sources are not easily performed, these fundamental investigations are carried out

Fourth International Symposium on Negative Ions, Beams and Sources (NIBS 2014)
AIP Conf. Proc. 1655, 020004-1–020004-6; doi: 10.1063/1.4916413

at the laboratory experiment ACCesS (**A**ugsburg **C**omprehensive **Ces**ium **S**etup) [13, 14] having ion source conditions in vacuum and plasma. The setup is equipped with a reliable and finely adjustable Cs source and multiple diagnostics for Cs and its environment in vacuum and plasma operation, which can be operated simultaneously in order to relate parameters from different diagnostics to the same experimental condition. Thus, ACCesS allows in particular to determine absolute values for the work function of sample surfaces depending on a variety of quantifiable parameters. This paper is dedicated to the dependence of the work function of caesiated surfaces on the Cs evaporation rate and the surface temperature in an ion source relevant vacuum, namely at background pressures of 10^{-6} mbar. Cs fluxes, the Cs density and the impurity content are monitored during the measurements.

EXPERIMENTAL SETUP

The laboratory experiment ACCesS [13, 14] consists of a stainless steel vessel with 15 cm in diameter and 10 cm in height. Background pressures of 10^{-6} mbar are obtained by a turbomolecular pump. Plasmas can be generated via inductive RF coupling (frequency 27.12 MHz, max. 600 W RF power) using a planar coil located on top of the vessel. The flexible setup allows for the attachment of a variety of peripheral equipments and diagnostics via several ports at the vessel walls and at the exchangeable bottom plate. The vessel walls can be heated by a temperature control system for the cooling water circuit of the discharge vessel, whereas the temperature was kept below 30 °C during the presented campaigns to assure reproducible conditions. Cs seeding is performed by evaporation from a Cs dispenser oven [15] placed at the bottom plate. Near the center of the vessel a sample holder is mounted electrically and thermally insulated from the grounded vessel walls. It is capable of holding samples with $3 \times 3\,\text{cm}^2$ size and the samples can be heated temperature controlled to temperatures of up to 300 °C. Owing to the insulated mounting of the sample holder the temperature of other components within the vessel, especially of the vessel wall, remains unaffected.

For the present investigations under vacuum conditions the arrangement of the diagnostics is shown in figure 1: the Cs evaporation rate is measured via a surface ionization detector (SID) mounted at the oven nozzle (denoted with 'Cs' in the figure), the volume averaged Cs density is determined by means of white light absorption spectroscopy (WABS) via the diagonal line-of-sight (LOS), the impurity content is monitored by a residual gas analyzer (RGA) and the work function is determined via the photoelectrical effect using the highly intense UV emission from a high pressure mercury lamp (100 W).

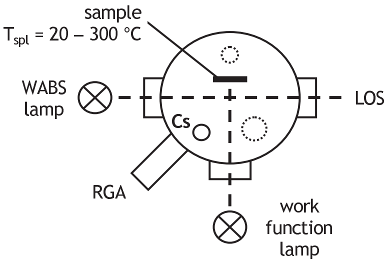

FIGURE 1. Top view of the experimental setup including the arrangement of the diagnostics and the sample surface.

A detailed view of the setup used to quantify the work function of a sample surface is shown in figure 2. The emission from the high pressure mercury lamp is led through an interference filter, two quartz lenses and the quartz window onto the sample surface mounted on the sample holder. The size of the spot on the sample is adjusted via lens 2 for maximal photocurrent (compromise between power density on the sample and absolutely absorbed power) having about 1.5 cm in diameter in the present setup. The subsequently photoemitted electrons are drawn to the vessel wall due to the bias of 30 V. The photocurrent depending on the incident wavelength is measured with a Keithley 602 Electrometer. Different wavelengths are obtained via the filter wheel that can house six interference filters and is directly mounted at the mercury lamp. Filters with central transmission wavelengths between 239 and 405 nm with a nominal FWHM of 10 nm are applied, which leads to usable photon energies between 5.3 and 3.0 eV. Due to the required bias voltage, work function measurements cannot be performed during plasma operation. An enhanced setup based on pulsing the plasma with several hertz and measuring the photocurrents during plasma-off-phases is presented in [7].

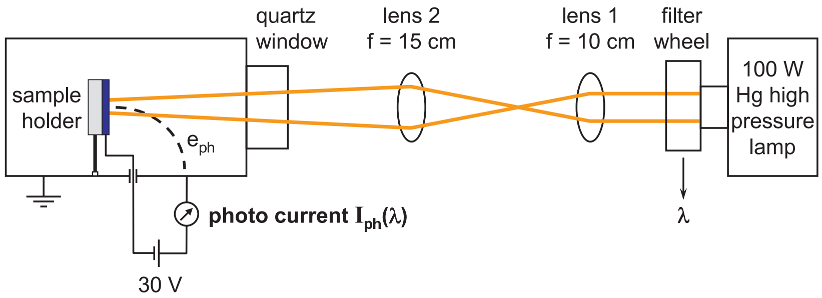

FIGURE 2. Detailed schematic of the setup for measuring the work function after the Fowler method.

Evaluation of the measured photocurrents is performed after the Fowler method [16]. Taking into account the electron energy distribution within a solid state, the amount of photoemitted electrons from a surface exposed to radiation increases with increasing photon energy. The work function χ of the surface is a parameter that describes the correlation between the measured photocurrent $I_{\mathrm{ph}}(h\nu)$ and the energy $h\nu$ of the inciding photons:

$$I_{\mathrm{ph}}(h\nu) \quad \propto \quad T_{\mathrm{spl}}^2 \, f\left(\frac{h\nu-\chi}{k_{\mathrm{B}}T_{\mathrm{spl}}}\right) \ , \quad \text{with } h\nu \geq \chi \ , \tag{1}$$

where T_{spl} is the sample temperature and k_{B} is the Boltzmann constant. Fitting the function f to the measured photocurrents for various photon energies gives the work function χ as fit parameter. For this purpose the relative intensity of the radiation inciding onto the sample has to be accounted for, which is done via calibrating the work function setup including the lenses and the quartz window of the experiment with an absolutely calibrated spectrometer positioned at the place where the sample surface would be.

The function f in equation (1) exhibits a threshold at $h\nu \approx \chi$. Hence, the closer the applied photon energies are to the work function, the steeper is f and thus, the more sensitive is the fitting routine in evaluating work functions resulting in smaller uncertainties for the determined value of χ. A thorough and elaborate analysis of the Fowler method further revealed [17], that the evaluated values of χ are always upper limits for the actual value and that this error increases with increasing difference between the actual work function and the lowest photon energy applied for the measurements. The resulting uncertainty of this effect is difficult to assess, but is typically of the order of 0.1 eV. However, to a certain extent a correction of this overestimation can be performed [17].

Hence, the presented values of χ in this paper are evaluated after the standard Fowler method [16] and corrected after [17] where possible. The indicated error bars, however, only reflect the standard error of the fit procedure.

RESULTS

Caesiation in vacuum

Figure 3 shows the effect of an increasing Cs evaporation rate on the work function of a stainless steel sample mounted on the sample holder. The sample is mechanically and chemically cleaned prior to the measurements. The evaporation rate is measured by the surface ionization detector at the oven nozzle and is directly linked to the volume averaged Cs density determined via white light absorption spectroscopy [15]. Thus, the evaporation rate can also be interpreted as the Cs flux onto the sample surface. The temperature of the sample was always below 100 °C during this campaign, determined by the passive heating due to the equipment within the experiment vessel (SID, Cs oven).

The bare stainless steel sample has a work function of above 3.5 eV, whereby this region is omitted in the graph to enhance the perception of the more relevant low work function region. Once Cs is evaporated the work function begins to decrease – at 0.25 mg/h χ is already at 3.2 eV. Increasing the evaporation rate up to slightly above 1 mg/h leads to a reduction of the work function down to about 2.8 eV. Further enhancement of the Cs evaporation to 1.6 mg/h results in the lowest measurable value for χ of a caesiated stainless steel surface under ion source relevant vacuum conditions: $\chi_{\mathrm{vac}} = 2.75$ eV. However, this value is much higher than that of a pure Cs layer of 2.14 eV, which indicates

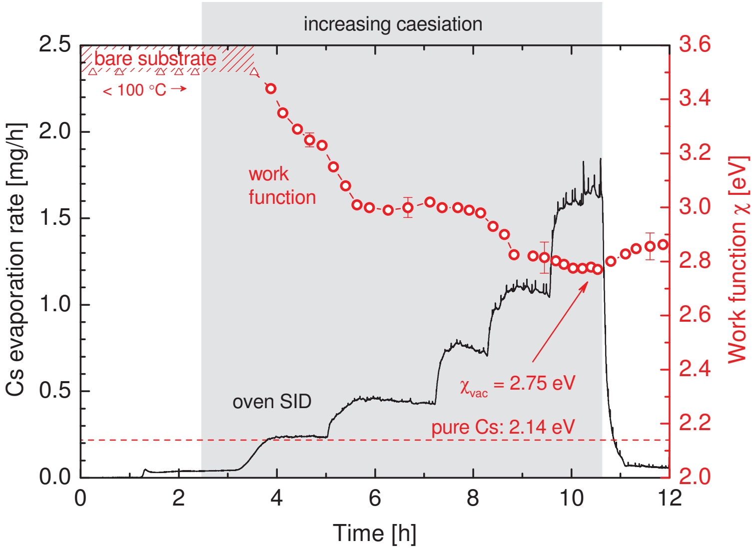

FIGURE 3. Work function of a stainless steel sample upon caesiation in vacuum with a background pressure of 10^{-6} mbar.

an involvement of Cs compounds within the surface layer. Shutting down the Cs evaporation directly leads to a deterioration of the work function due to the degradation of the Cs layer by incorporation of residual gases. This degradation proceeds far beyond the shown period attaining values of slightly above 3 eV after some days in vacuum (see figure 4).

Influence of elevated temperatures

The impact of impurities on the Cs layer should be affected by the surface temperature. Thus, the stainless steel sample is actively heated and the influence on the work function is investigated. Two different campaigns are considered, shown in figure 4: (i) heating an already caesiated sample and (ii) caesiating a bare sample at elevated surface temperatures.

After some days in vacuum the caesiated stainless steel sample from figure 3 has a degraded work function of slightly above 3 eV due to the incorporation of impurities from the residual gas. Heating the sample in the vacuum environment leads to the evaporation of the adsorbates (monitored with the residual gas analyzer and at the temporarily rising background pressure) accompanied by a decrease of the work function, whereby higher temperatures have a larger impact. At 200 °C a work function of 2.8 eV can be regained without any evaporation of Cs. This behavior indicates a cleaning effect of the Cs layer, due to which Cs compounds are cracked or evaporated from the surface. However, increasing the temperature further (not shown here) results in an increasing work function again, which can be explained by an effective evaporation of Cs from the surface (observable at a temporary but significant increase of the SID signal). Hence, the optimal temperature is around 200 °C under these vacuum conditions. Introducing Cs evaporation ($\approx$ 1.6 mg/h) and thus a flux of Cs onto the hot surface again leads to a decrease of the work function down to the minimum vacuum value of 2.75 eV.

In order to reduce the influence of impurities adsorbed at the sample surface from the beginning on, the second campaign covers caesiation of a sample at already elevated temperatures. For that purpose a bare stainless steel sample is heated to 200 °C and kept at that temperature during caesiation. Heating the sample is accompanied by temporary increasing impurity signals at the residual gas analyzer and an increasing background pressure confirming the desorption of impurities from the surface. The work function of the bare sample is hardly influenced by the heating and remains above 3.5 eV. As soon as Cs is evaporated the work function instantaneously drops to about 2.7 eV and subsequently increases slightly to a stable value of again 2.75 eV. Even increasing the evaporation rate up to more than 5 mg/h does not change the work function in that state.

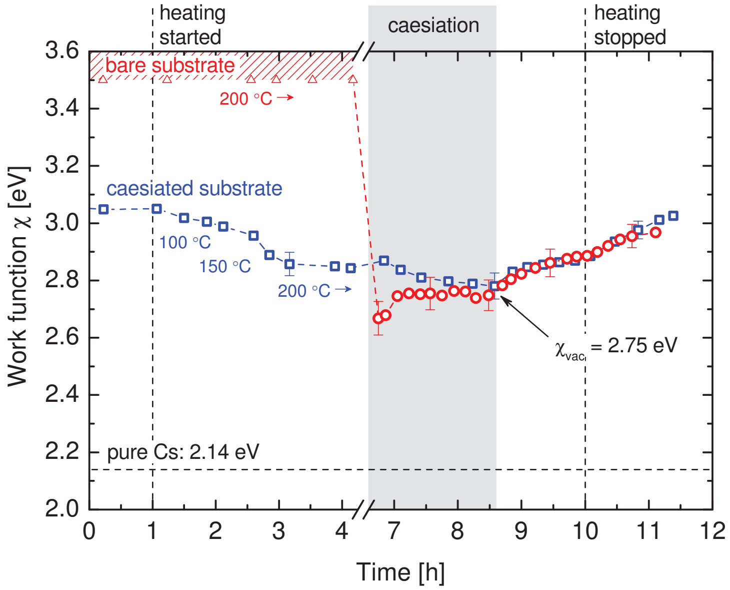

FIGURE 4. Work function of (i) a caesiated stainless steel sample and (ii) a bare stainless steel sample due to heating and caesiation in vacuum with a background pressure of 10^{-6} mbar.

After stopping the Cs flux, the work function begins to degrade, whereas the behavior is very similar for both campaigns. Moreover, it can be seen that switching off the active heating leads to an even faster degradation.

Discussion

The minimally achievable work function of a stainless steel sample caesiated under background pressures of 10^{-6} mbar is $\chi_{\text{vac}} = 2.75$ eV. This value is only determined by the Cs evaporation rate and saturates above about 1.5 mg/h. At these Cs fluxes the work function is moreover independent of the surface temperature for temperatures up to 260 °C. However, it is about 0.6 eV higher than that for pure Cs layers, which is far beyond the uncertainty of the applied method of measurement. Thus, it can be stated that Cs compounds are involved in the layer upon the substrate which increase the work function due to their non-metallic character. The ineffectiveness of surface temperatures up to 260 °C or evaporation rates of more than 5 mg/h in order to further decrease the work function indicates, that the flux of impurities from the residual gas is too high to be compensated. Hence, it is subordinate if the impurity coverage *upon* the surface is diminished by heating the sample, since the impurity flux *onto* the surface is determining. Under these conditions evaporation rates of slightly above 1 mg/h are sufficient to achieve and maintain work functions of slightly below 2.8 eV. However, in absence of a Cs flux elevated surface temperatures are beneficial for cleaning and against degradation of the Cs layer. Thus, a work function of 2.8 eV can be regained from a degraded Cs layer by heating to 200 °C without the need to evaporate caesium.

In summary, the work function of a caesiated sample under ion source relevant vacuum conditions is increased compared to pure Cs due to the co-adsorption of impurities from the background pressure. Hence, the question arises, if the substrate can have an influence on the resulting work function, or if it is mainly determined by the interplay of the fluxes of Cs and residual gases onto the surface. However, a promising possibility for an effective cleaning of the surface is plasma surface interaction. This was already shown in [7] and will be investigated in detail in comprehensive studies in the midterm.

CONCLUSION

The work function of the caesiated converter surface in sources for negative hydrogen and deuterium ions is a key parameter for the performance of the ion source and is subject to plenty of impacts during ion source operation. Background pressures of 10^{-7}–10^{-6} mbar and plasma surface interaction due to H_2 or D_2 low-temperature plasmas suggest that literature data on the evolution of the work function of fractional Cs layers obtained under ultra-high vacuum conditions cannot be applied. Thus, fundamental studies on the work function of caesiated surfaces under ion source conditions and its influencing factors are performed at the laboratory experiment ACCesS applying the Fowler method for the evaluation of reliable absolute work function values.

The investigations under ion source relevant vacuum conditions have shown, that the work function of a caesiated stainless steel sample is increased compared to a pure Cs coating, the minimum value being 2.75 eV. Elevated surface temperatures have no influence on this minimum, but are beneficial for cleaning and against degradation in absence of Cs evaporation. Thus, keeping the converter surface in negative ion sources at an elevated temperature during operation breaks can counteract the degradation of the Cs layer and ease the re-conditioning of the source. This would furthermore be beneficial for the reduction of the Cs consumption. The systematic investigations at ACCesS will be continued aiming in the next steps at the influence of the substrate material and plasma surface interaction.

ACKNOWLEDGMENTS

This work has been carried out within the framework of the EUROfusion Consortium and has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement number 633053. The views and opinions expressed herein do not necessarily reflect those of the European Commission.

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