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# Photo-Assisted $O^-$ and $Al^-$ Production with a Cesium Sputter Ion Source

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**Abstract.** It has been recently proposed that the production of negative ions with cesium sputter ion sources could be enhanced by laser-assisted resonant ion pair production. We have tested this hypothesis by measuring the effect of pulsed diode lasers at various wavelengths on the $O^-$ and $Al^-$ beam current produced from $Al_2O_3$ cathode of a cesium sputter ion source. The experimental results provide evidence for the existence of a wavelength-dependent photo-assisted enhancement of negative ion currents but cast doubt on its alleged resonant nature as the effect is observed for both $O^-$ and $Al^-$ ions at laser energies above a certain threshold. The beam current transients observed during the laser pulses suggest that the magnitude and longevity of the beam current enhancement depends on the cesium balance on the cathode surface. It is shown that the ions produced by the laser exposure originate from slightly different potential than the surface produced ions, which allows us to constrain the underlying physical mechanisms. It is concluded that the photo-assisted negative ion production could be of practical importance as it can more than double the extracted beam current under certain operational settings of the cesium sputter ion source. We discuss experiments designed to confirm or dispute the relevance of the ion pair production for negative ion production with cesium sputter ion sources and the possibility of ion pair production explaining the beneficial effect of xenon admixture on the negative ion yield of an RF-driven $H^-$ ion source.

## INTRODUCTION

The negative ion production in cesium sputter ion sources is traditionally attributed to surface ionization (see e.g. [1] and original references therein). The ionization efficiency depends on the work function of the surface [2], the sputtering yield and the escape velocity of the negative ions [3]. Despite extensive research on the physics of cesium sputter ion sources, there is a persistent discrepancy between the yields of negative ions deduced with reasonable estimates of the electron affinities and work functions involved, and the measured negative ion currents as noted already in 1960s [4]. The extracted currents often exceed even the optimistic predictions, which motivates the search of the underlying physical mechanism. To our knowledge, there are no systematic experiments thus far attempting to explain the difference between the theory and experiments, which is probably due to the lack of a testable hypothesis on the mechanism governing the negative ion production through an alternative path.

In a recent publication, Vogel [5] argues that resonant ion-pair production – first noted in thermal alkali vapors [6] – could explain the enhanced yield of negative ions in cesium sputter sources. The ion pair-production is described by the (chemical) reaction

$$A^* + B \rightarrow A^+ + B^-, \tag{1}$$

which depicts the interaction between a neutral atom in an excited state ($A^*$) and a ground state neutral atom B with positive electron affinity, resulting to the formation of a positive ($A^+$) and negative ($B^-$) ion pair.

The ion pair production cross section $\sigma$ is a function of the ionization potential of the donor (A) and the electron affinity of the acceptor (B) and can be calculated by multiplying the area of an excited electron's orbit by the total

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probability of the electron exchange, integrated over the impact parameter $b$ from 0 to the orbit radius $R_c$, i.e.

$$\sigma(\nu,\gamma) = 2\pi R_c^2 \int_0^1 p(x)(1-p(x))xdx, \tag{2}$$

where $\nu^2 = I_{p,A}/13.6$, $\gamma^2 = E_{A,B}/13.6$, $R_c = 2/(\nu^2-\gamma^2)$ and $x = b/R_c$ (with the ionization potential and electron affinity given in eV units). The electron exchange probability $p(x)$ obtained from Landau-Zener-Stückelberg (LZS) formalism is available in the literature [7, 8, 9]. The electron transfer is considered resonant when the effective ionization potential, $I_{p,\mathrm{eff}}$, of the electronically excited donor atom equals the sum the acceptor atom's electron affinity $E_A$ and the reaction endothermicity $\Delta E$ i.e.

$$I_{p,\mathrm{eff}} = E_A + \Delta E. \tag{3}$$

The effective ionization potential corresponding to the above resonance condition can be estimated from a power fit to experimental data [7], i.e.

$$I_{p,\mathrm{eff}} = E_A \left(\frac{4.72}{E_A}\right)^{0.275}. \tag{4}$$

The approximation is based on experiments with molecular negative ions but it has been shown to be applicable also for atomic species (see Fig. 4 in Ref. [5] for details).

In Ref. [5] it is argued that – in the cesium sputter negative ion source – electronically excited Cs atoms can contribute to the negative ion yield via the described ion pair production mechanism. The Cs atoms can be excited in inelastic collisions with electrons emitted from the cathode, presumably explaining the "blue cathode glow" noted in the literature [10], or by absorption of photons from an external source. The latter offers a practical means to enhance the negative ion currents on the condition that Cs atoms can be excited to electronic states, which are in resonance with the affinity states of the anions. Ref. [5] describes an experiment where the $C^-$ beam current of a cesium sputter ion source increased 10% when the volume in front of the cathode was exposed to a 450 nm / 5 W laser beam from the radial direction. It is argued that some of the laser power was absorbed by the excitation of the Cs(7p)-states, which are allegedly in resonance with the effective ionization potential in the reaction $Cs(7p)+C \rightarrow Cs^+ + C^-$.

We have recently reported experimental results on photo-assisted $O^-$ ion production with a cesium sputter ion source [11]. In this paper, we expand on that work e.g. by reporting results on photo-assisted production of $Al^-$ ions from $Al_2O_3$ cathode. Measuring the response of both negative ion species on the laser exposure allows testing of the ion pair production hypothesis. This is because the excited states of Cs which are in resonance with the $O^-$ and $Al^-$ pair production differ as shown in Fig. 1.

## EXPERIMENTAL SETUP

The experimental data were taken at the JYFL accelerator laboratory on a Multi-Cathode Source of Negative Ions by Cesium Sputtering (MC-SNICS, see Ref. [13] for mechanical details) by National Electrostatics Corporation (NEC). In the following, we only give an overview of the source operation with a schematic drawing in Fig. 2 and refer the reader to the literature [1, 13] for further details. Cesium is evaporated from an external oven into the ionization chamber, where some of it condenses on the surface of the cathode and lowers the effective work function of the cathode material. Some of the cesium vapor is surface ionized on the hot surface of the resistively heated ionizer. The $Cs^+$ ions are then accelerated toward the cathode by biasing it to a kV order of magnitude negative potential. The ion flux is focused with a "cesium focus lens" placed 5 mm from the cathode and biased to ~1 kV potential with respect to the cathode. The negative ions are liberated from the surface by the $Cs^+$ bombardment and self-extracted by the cathode potential, propelling them through the extraction channel into the adjacent beamline. The primary 'control knobs' of the negative ion current are the the cesium oven temperature, ionizer temperature (heating power) and cathode bias affecting the cathode cesium coverage, $Cs^+$ flux impinging the cathode and the sputtering yield, respectively.

In our experiment, the cathode was prepared by pressing $Al_2O_3$ powder into a cylindrical notch 1 mm in diameter on the surface of a copper housing. The photo-assisted production of $O^-$ and $Al^-$ ions was investigated by illuminating the $Al_2O_3$ sample with various diode lasers through a viewport in a 30 degree dipole magnet, allowing a direct line-of-sight to the cathode. It was confirmed by a direct measurement with a thermal power sensor that 5–20% of the

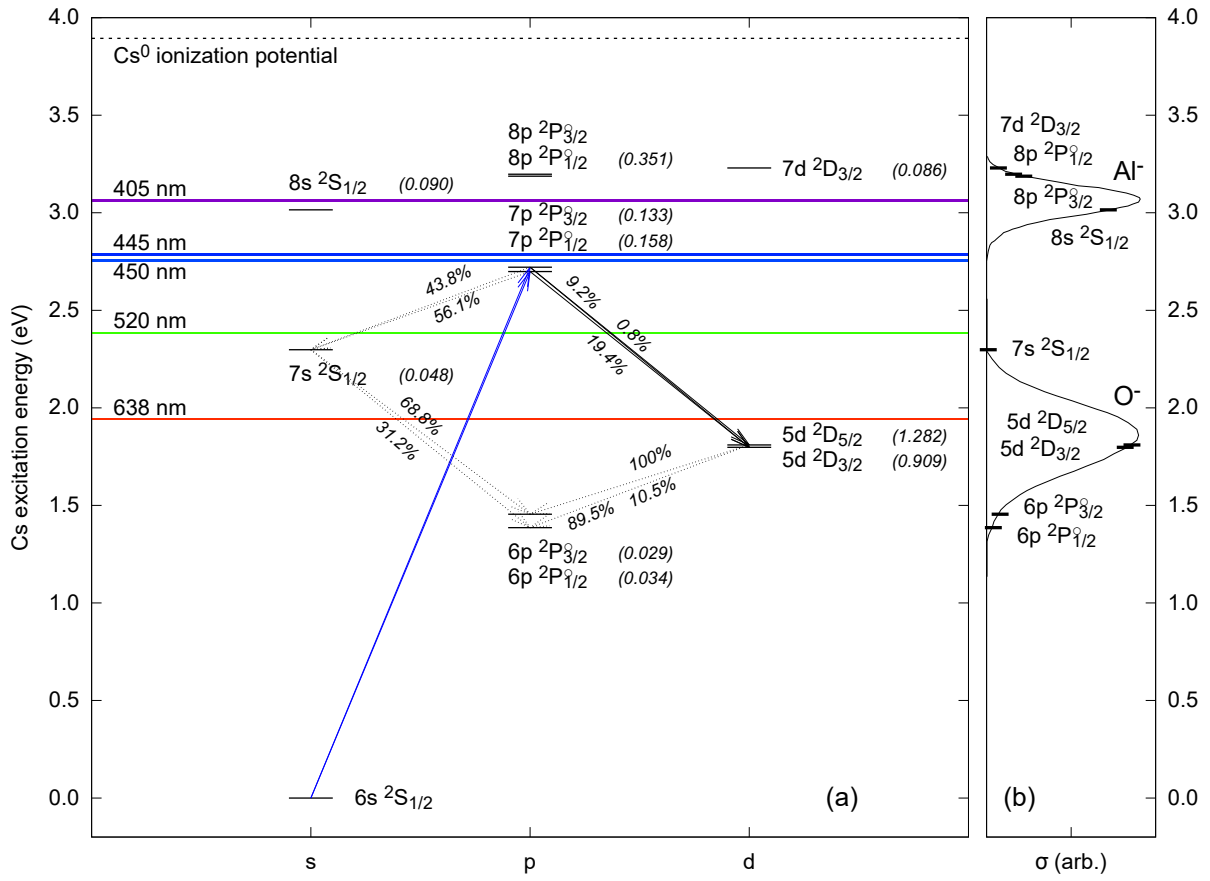


**FIGURE 1.** A partial Grotrian diagram of neutral cesium. The diagram (a) shows the putative excitations to 7p-states by the 450 nm laser and the wavelengths of all diode lasers used in our experiment with the linewidths corresponding to the FWHM of each diode. The radiative decays populating the 7s, 5d and 6p states that overlap in energy with the normalized $O^-$ ion pair production cross section [5] $\sigma$ (b) are indicated by black arrows. The most likely excited state contributing to the $O^-$ pair production is the metastable 5d (populated by the transitions marked with solid downward arrows) almost matching the peak of the cross section shown by the black bars projected onto the cross section curve. The pair production cross section of $Al^-$ overlaps with the 8s, 8p and 7d excited states of neutral cesium, which are either optically forbidden or energetically inaccessible from the ground state with laser wavelengths ≥ 405 nm. The spontaneous lifetime of each excited state in μs is shown in parentheses. The branching ratios (of >5%) of the de-excitations calculated from the reported oscillator strengths [12] are displayed next to each downward transition.

laser power was delivered to the cathode. The power is limited by the mismatch between the beam spot size and the geometrical apertures i.e. extraction channel and sample diameter. The effect of the laser exposure on the $O^-$ and $Al^-$ beam current was probed by modulating the output of the laser(s) by pulsing the drive current (on/off) at various frequencies and measuring the extracted negative ion beam currents from a Faraday cup downstream from the dipole magnet separating different ion species and ion energies. Figure 3 is a photograph of the experimental setup.

## EXPERIMENTAL RESULTS

The experiments were started with a 450 nm (2.76 eV), 2 nm FWHM laser presumably exciting the 7p-states of neutral Cs as argued in Ref. [5], which then populate the long-lived 5d-states that are expected to contribute to $O^-$ pair production (see Fig. 1). At the same time, transitions from the ground state to the 8s, 8p and 7d-states – which could contribute to $Al^-$ ion yield via pair production – are optically forbidden (8s) or remain energetically inaccessible (8p and 7d) by single-photon excitation. Figure 4 shows the extracted $O^-$ and $Al^-$ beam currents as a function of

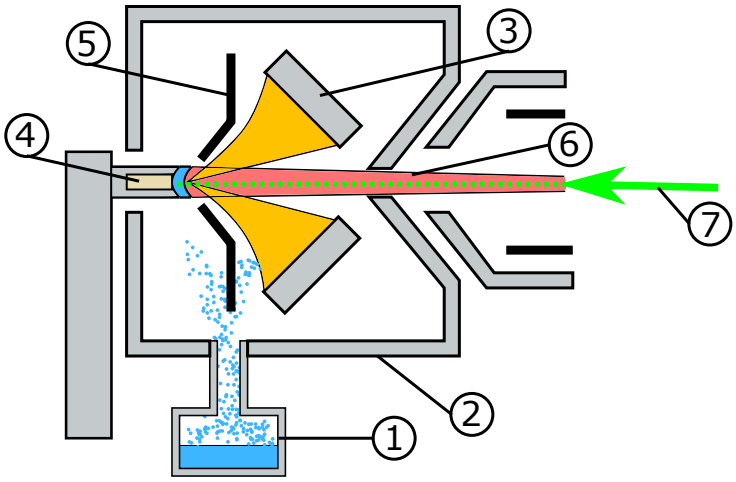


**FIGURE 2.** Schematic drawing of the SNICS ion source. (1) Cesium oven and transfer line, (2) ionization chamber, (3) ionizer, (4) cathode with $Al_2O_3$ powder target, (5) $Cs^+$ focusing electrode (immersion lens) and (6) extraction channel and electrodes with the negative ion beam depicted with red color. In our experiments the laser beam was illuminating the cathode face through the extraction channel as indicated by the green arrow (7).

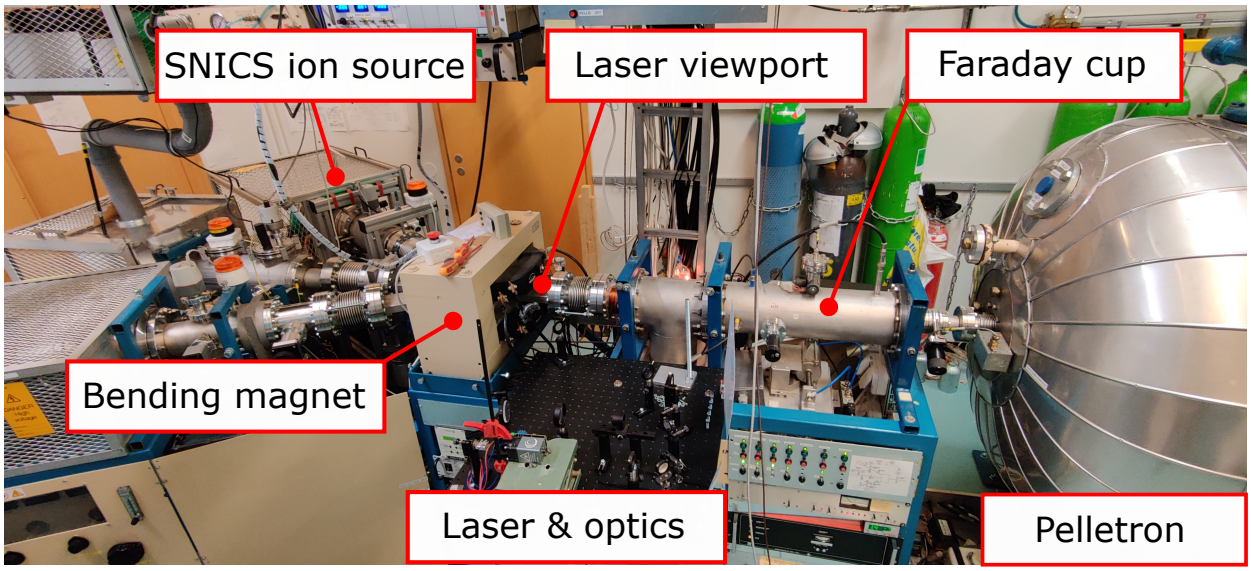


**FIGURE 3.** The experimental setup at JYFL accelerator laboratory. The laser beam was focused to the SNICS cathode through the viewport of the bending magnet.

time when the $Al_2O_3$-cathode was exposed to short (100 ms, 50% duty factor) laser pulses. Both, $O^-$ and $Al^-$ beams exhibit a prompt enhancement reacting to the laser pulse contrary to the expectation. It was confirmed that the effect is indeed a prompt one by comparing the signal rise time to the combined time constant of the Faraday Cup and the transimpedance amplifier. The fact that both beam currents increase by 5–6% during the laser pulse implies that the laser exposure amplifies an already existing negative ion production channel. The periodic ripple visible especially in the case of the $Al^-$ beam is due to the 50 Hz mains signal. The extracted beam currents are only modest as the ionizer temperature controlling the $Cs^+$ ion flux incident on the cathode was deliberately maintained low to observe the photo-assisted contribution superimposed on the continuous beam current. The magnitude of the prompt effect was observed to depend linearly on the laser power in the 320–1600 mW range accessible with the 450 nm diode [11].

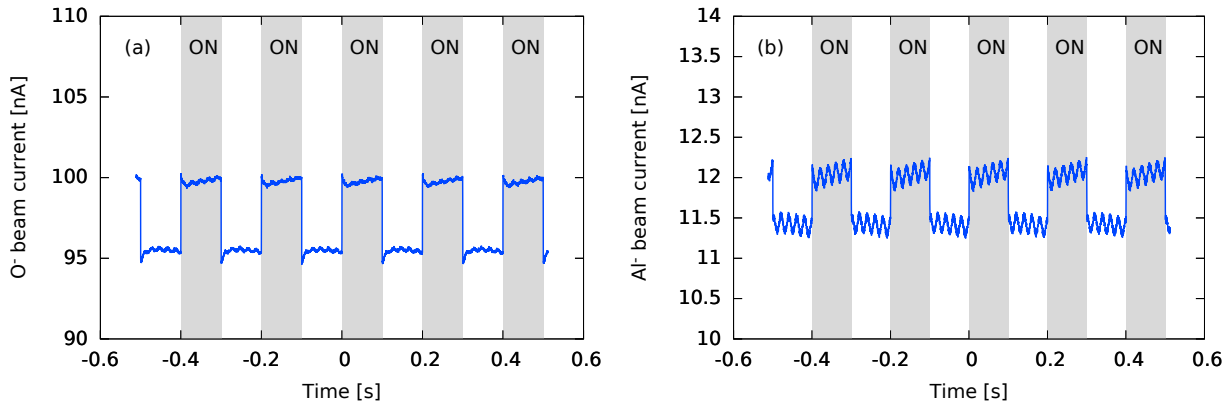


**FIGURE 4.** The prompt effect of the 450 nm laser with 1 W power on (a) $O^-$ (5.3%) and (b) $Al^-$ (5.9%) beam currents.

In order to gain understanding on the observed photo-assisted negative ion production, we measured the $O^-$ beam current gain by the 450 nm laser exposure as a function of the beamline dipole magnet field strength at $B < B_{optimum}$. Here $B_{optimum}$ refers to the magnetic field corresponding to maximum beam current without the laser, i.e. the mean energy of the sputtered ions. The result of the sweep is shown in Figure 5. The prompt effect of the laser exposure is more significant when $B < B_{optimum}$. This means that energy of the ions created due to the laser exposure is below the mean energy of the ions produced by cesium sputtering and therefore extracted with an energy corresponding to the cathode potential and the energy of the sputtered particles ejected from the surface (with a certain energy spread).

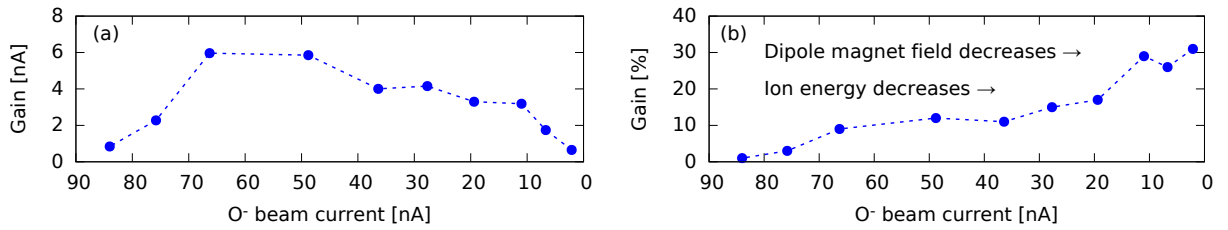


**FIGURE 5.** The prompt gain in nA-units (a) and %-units (b) induced by the 450 nm / 1 W laser on the $O^-$ beam current when the dipole magnet field was decreased from the optimum (optimum setting determined without the laser exposure).

Figure 6(a) shows the response of the $O^-$ beam current to four different diode lasers at 405, 450, 520 and 638 nm wavelengths, i.e. 3.06, 2.76, 2.38 and 1.94 eV. The 5d excited states of Cs that are expected to be in pair production of $O^-$ ions are not populated with the 405, 520 and 638 nm lasers (see Ref. [11] for further details). Nevertheless, a prompt enhancement of the $O^-$ beam current is observed with the 405 nm laser, which together with Fig. 4 leads us to conclude that, rather than a resonance process, the photo-assisted contribution is a threshold effect. The effect of the 405 nm laser is larger than the effect of the 450 nm laser, which could imply a wavelength dependence at photon energies above the threshold. However it must be emphasized that the two data in question were recorded under slightly different ion source settings. Figures 6(b)&(c) demonstrate that (i) the photo-assisted effect induced by the 405 nm laser was observed for both $O^-$ and $Al^-$, (ii) the effect is observed also at elevated beam current, making it relevant for the ion source operation for Ion Beam Analysis and (iii) the prompt effect is followed by a slow transient, which is believed to be due to varying Cs balance on the cathode surface.

The observations of the photo-assisted effect not being wavelength sensitive; its magnitude depending on the laser power, and the indication of the importance of the cesium balance on the cathode surface, motivated us to conduct further experiments with a 6 W, 445 nm laser. This was done in order to study the effect of the laser power and wavelength at elevated beam currents; to assess the practicality of the photo-assisted ion production to boost the beam currents extracted from the SNICS ion source under "normal" operating conditions. Fig. 7(a) shows an example measured at source settings where the laser exposure approximately doubles the extracted $O^-$ current. In addition to

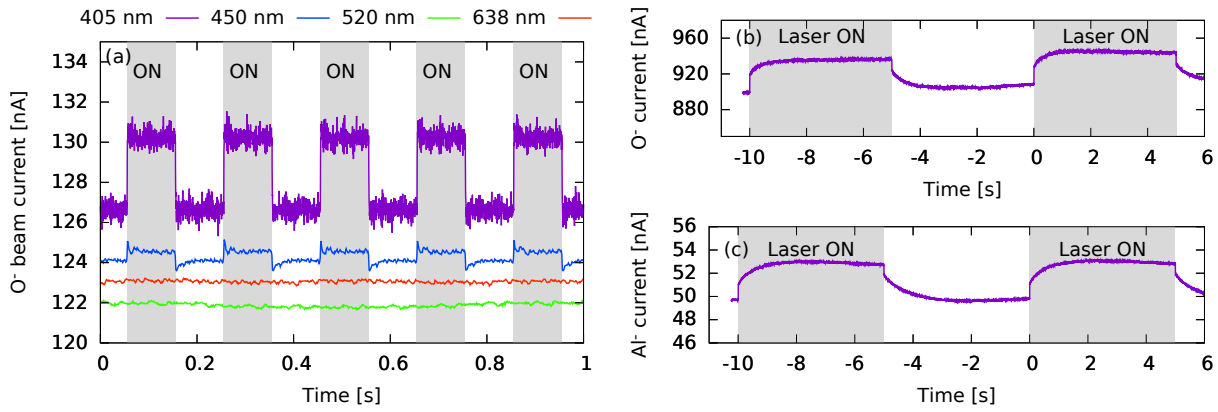


**FIGURE 6.** The effect of (a) 100 ms laser pulses at 405 nm / 1 W, 450 nm / 1.6 W, 520 nm / 1.0 W and 638 nm / 0.7 W wavelengths / powers on the $O^-$ beam current. The effect of 5 s laser pulses at 405 nm / 1 W on $O^-$ (b) and $Al^-$ (c) at elevated beam current. The beam current was adjusted by changing the cesium oven and ionizer temperatures.

the prompt effect there is a long-term transient, which is believed to be caused by a change of the cathode surface cesium coverage and, hence, its work function. The importance of the cesium coverage is highlighted by Fig. 7(b) showing the effect of the 445 nm laser power on the $O^-$ beam recorded with source settings differing from those in Fig. 7(a). In this case, at very low powers a small prompt effect is observed but it is then overpowered by a gradual decrease of the beam current. Only at 6 W power the enhancement of the beam current by the laser-induced (prompt) effect is larger than the effect caused by the removal of cesium from the surface.

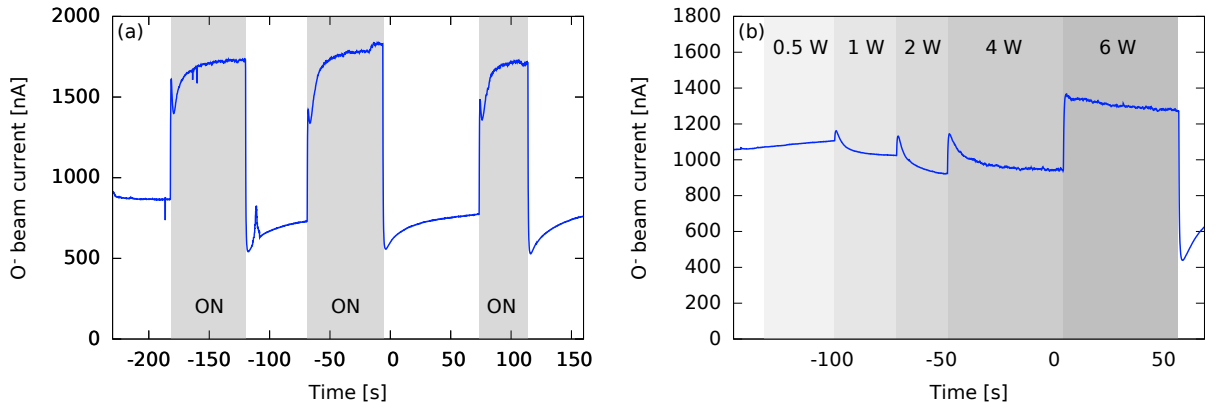


**FIGURE 7.** (a) The effect of the 445 nm, 6 W laser on the $O^-$ yield at elevated beam current. (b) The effect of the 445 nm laser power on the $O^-$ beam current at different cesium oven and ionizer temperature.

## DISCUSSION

The experiments described above and in Ref. [11] have confirmed the existence of a photo-assisted enhancement of negative ion production in cesium sputter ion sources, first noted by Vogel [5]. The insensitivity of the prompt effect on the laser wavelength and the fact that both, $O^-$ and $Al^-$ exhibit a similar enhancement, questions the hypothesis of resonant ion pair production explaining the observed photo-assisted beam current gain. We therefore suggest that the observed effect is probably due to laser-induced variation of the Cs density on the cathode surface, which affects the surface work function. In this case the laser-induced prompt effect would be best explained by photoelectrons ejected from the low work function surface, and contributing to the negative ion yield by (i) direct electron attachment, (ii)

increased volumetric ionization of neutral Cs and subsequent change of the $Cs^+$ flux bombarding the cathode or (iii) by promoting the ion pair production through enhanced electron impact excitation to the relevant excited states of neutral Cs in the close proximity of the cathode surface [14]. The photoelectron emission from cesiated surfaces has been demonstrated to depend significantly on the Cs coverage [15], which would explain the sensitivity of the photo-assisted negative ion production on the Cs balance. It is also worth pointing out that varying the (unknown) Cs-coverage of the cathode and its work function can be expected to affect the survival probability of the negative ions. According to so-called probability model [16, 17], the negative ion survival probability, $p_s$, is proportional to the escape velocity of the negative ion normal to the surface, $v_\perp$, and the surface work function, $\phi$ as

$$p_s \propto v_\perp(\phi - E_A - \Delta E_A), \tag{5}$$

where $E_A$ is the electron affinity of the negative ion far from the surface and $\Delta E_A$ the shift of the electron affinity level near the cathode surface. The data in Fig. 5 implies that the escape velocity of the ions formed through the prompt effect differs slightly from the escape velocity of those ions forming the bulk of the extracted beam, which affects their survival probability. Thus, the measurement of the photo-assisted gain of the beam current as a function of the cathode bias – which determines the energy of the sputtering $Cs^+$ ions and affects the escape velocity of the ejected negative ions – could potentially shine light to the mechanism explaining the laser-induced (prompt) effect.

The photo-assisted negative ion production could be of practical importance for the operation of cesium sputter ion sources as demonstrated by the factor of >2 increase of the $O^-$ current achieved with the 6 W laser as well as the temporal control of the beam current. The method could be applied for reducing the erosion rate of the cathode and, thus, increasing its lifetime by enabling to reach the same beam current (as without the laser) at reduced $Cs^+$ flux.

It is emphasized that our experiments are not sufficient to dispute the ion pair production contributing to the negative ion yield. This is because the emission wavelength of the diode laser(s) does not correspond to the resonant excitation energy of the either 7p state of cesium. The possible contribution of ion pair production on the negative ion currents extracted from cesium sputter ion sources should instead be confirmed or disputed with an adjustable wavelength laser scanning across the relevant wavelengths corresponding to both 7p excitations of neutral Cs at 455.7 and 459.5 nm. Thus, the experiments will be continued with a 130 mW CW laser (Toptica DLC DL pro HP) with a scanning range of 455–460 nm: enhancing the available power at the resonant wavelength by several orders of magnitude. Such an experiment is partly motivated by the fact that surface effects cannot explain the increase of $C^-$ ion in Ref. [5], where the cathode surface was not directly illuminated by the radially injected laser beam. A complete assessment of the photo-assisted negative ion production also requires experiments with other negative ions and cathode materials. The role of Cs could be best studied with cathode materials made of Cs compounds, such as CsCl typically used for the production of $Cl^-$ ions. This is because the fluctuation of the cathode Cs coverage could be suppressed for these materials, which would then allow assessing the effect of the laser without suffering from long term transients and sensitivity to source parameters.

## ON THE POSSIBILITY OF ION PAIR PRODUCTION IN PLASMA ION SOURCES

It is expected that in cesiated plasma ion sources, the benefits of exposing the negative ion production surface to a photon flux from an external source are limited, as plasmas naturally radiate up to several tens of percent of the discharge power in the UV/VUV-range [18, 19]. Nevertheless, ion pair production could presumably enhance the volume production of negative ions in two-component discharges if the resonant excited states are populated by inelastic electron impact of photo-excitation. Kalvas et al. [20] have reported that the negative ion current density extracted from an RF-driven multicusp $H^-$ ion source increased noticeably when xenon gas was introduced into the hydrogen discharge as shown in Fig. 8(a).

In Ref. [20] the authors concluded that: “The volume production of $H^-$ is enhanced by gas mixing because the cold electron density near the extraction increases as heavier ions bring cold electrons through the magnetic filter to the low-energy plasma chamber”. The given explanation contradicts the conclusion by Komppula et al. arguing that volume production of $D^-$ ions in a multicusp ion source is limited by the diffusion of the heavier isotope across the transverse filter field [21]. Thus, alternative explanations for the $H^-$ beam current enhancement by the Xe admixture should be sought. A possible mechanism is the excitation of neutral Xe atoms to 6p, 7p and 6d states, which are in resonance with the pair production of $H^-$ ions as shown in Fig. 8(b). The above states can be populated from the ground state by inelastic electron impact (6p and 7p) or by the absorption of Lyman- and Werner-band photons emitted by the decay of the $B^1\Sigma_u^+$ and $C^1\Pi_u$ states of the $H_2$ molecules (6d) radiating several watts of power in the correct

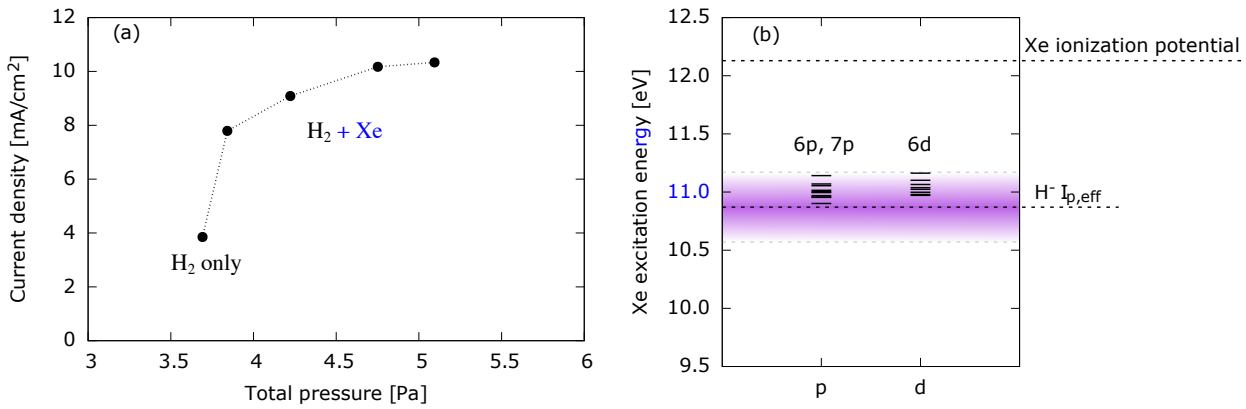


**FIGURE 8.** (a) The negative ion beam current density extracted from an RF-driven multicusp $H^-$ ion source as a function of total hydrogen and xenon pressure (the figure is reproduced from Ref. [20]). (b) Electronically excited states of neutral xenon overlapping with the $H^-$ pair production cross section (marked with the gradient color).

wavelength range [18]. Regardless of the actual mechanism the experimental observation warrants further studies with gas admixtures in volume production $H^-$ ion sources.

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