# AN ANALYSIS OF FAST SPUTTERING STUDIES FOR ION CONFINEMENT TIME\*

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### Abstract

Existing heavy ion facilities such as the National Superconducting Cyclotron Laboratory at Michigan State University rely on Electron Cyclotron Resonance (ECR) ion sources as injectors of highly charged ion beams. Long ion confinement times are necessary to produce dense populations of highly charged ions due to the steadily decreasing ionization cross sections with increasing charge state. To further understand ion extraction and confinement we are using a fast sputtering technique first developed at Argonne National Laboratory (ANL) [1] to introduce a small amount of uranium metal into the plasma at a well-defined time. We present an analytical solution to the coupled ion density rate equations for using a piecewise constant neutral density to interpret the fast sputtering method.

## **INTRODUCTION**

ECR Ion Sources (ECRIS) are injectors of choice for many heavy ion accelerators worldwide. These ion sources can ionize elements to higher charge states with greater brightness than other source types, and metallic beams such as uranium or calcium are routinely produced. A pure metal or a metallic compound is vaporized into a support plasma, typically consisting of a naturally gaseous element like oxygen. Well-developed techniques for metal vaporization include laser ablation, sputtering, and neutral beam ovens. Two methods, sputtering and laser ablation, have been developed at ANL to probe ion characteristic timescales in an ECR plasma [1,2]. Repetitive short bursts of neutral metal vapor were added to the established stable gaseous plasma and observed as pulses of extracted metallic beam current. Each charge state of the metal contaminant was resolved through a dipole magnet and collected on a Faraday cup. The measured beam current was sampled with an oscilloscope. We present an analytical solution that attempts to deconvolve ion confinement from beam current waveforms. We consider the following reactions: single electron impact ionization

$$A^{i} + e = A^{i+1} + 2e, (1)$$

charge exchange

$$A^{i} + A = A^{i-1} + A^{+}, (2)$$

and electron recombination

$$A^i + e = A^{i-1} + E_\gamma \tag{3}$$

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for our analysis of ion density. In Eq. (1), (2), (3), A denotes a molecule or atom, *i* represents the charge state, *e* the electron, and  $E^{\gamma}$  the radiated photon energy.

# ANALYTICAL SOLUTION FOR ION DENSITY

A reaction rate is defined by

$$\int_{\infty} \sigma_{i \to j}(E) \nu f_e(E) dE = n_e < \sigma \nu >_{i \to j} .$$
(4)

The Electron Energy Distribution Function (EEDF) is assumed to be isotropic in velocity space and rewritten in terms of energy  $f_e(E)$ . The reaction rate is calculated by weighting the EEDF by cross section  $\sigma(E)$  and center of mass scalar velocity  $\nu$ . We assume no energy dependence on the particle location and therefore electron density  $n_e$ may be treated as a scalar and is separated from the EEDF explicitly. We may write all the source and sink rates for a single ion species as

$$dn_{i}/dt = n_{e}n_{i-1} < \sigma \nu >_{i-1 \to i} +$$

$$n_{0}(t)n_{i+1} < \sigma_{cx}\nu >_{i+1 \to i} + n_{e}n_{i+1} < \sigma_{r}\nu >_{i+1 \to i}$$

$$-n_{e}n_{i} < \sigma \nu >_{i \to i+1} - n_{0}(t)n_{i} < \sigma_{cx}\nu >_{i \to i-1}$$

$$-n_{e}n_{i} < \sigma_{r}\nu >_{i \to i-1} - n_{i}/\tau_{i}$$
(5)

wherein:  $n_i$  is the density of the  $i^{th}$  ion,  $\sigma_{cx}$  is the charge exchange cross section between ions and neutrals,  $\sigma_r$  is the recombination cross section between ions and electrons,  $\sigma$  without subscript represents an ionization cross section, and the ion confinement time for the  $i^{th}$  species is written as  $\tau_i$ . Since Eq. (5) is cumbersome, it may be simplified to

$$dn_i/dt + \gamma_i n_i = \alpha_i n_{i-1} + \beta_i n_{i+1} \tag{6}$$

by creating effective rate coefficients  $\alpha$ ,  $\beta$ , and  $\gamma$  that are multiplied to respective ion densities. These rate coefficients carry units of inverse time and are real and positive. Additionally,  $\alpha$  and  $\beta$  act as particle sources into the *i*<sup>th</sup> charge state while  $\gamma$  represents an effective sink from the *i*<sup>th</sup> charge state.

#### *Expansion in Charge State*

Due to cross coupling in ion density within Eq. (5), a complete analytical solution across all charge states is not practical. Therefore, we consider here only a small subset of charge states. We will include the effects of the neighboring charge states by taking a time derivative of Eq. (6) to produce

$$d^{2}n_{i}/dt^{2} + \gamma_{i}dn_{i}/dt = \alpha_{i}n_{i-1}/dt + \beta_{i}n_{i+1}/dt.$$
 (7)

The ion species  $n_i$  under analysis, from the beginning, was assumed to be a sufficiently small perturbation that the rate coefficients are fixed in time by the more numerous support

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plasma species. However, since charge exchange depends on the neutral density and we know  $n_0$  must change in time during a fast sputtering event we consider a piecewise constant time dependence on  $n_0$  that is introduced for a short time  $\delta$  at a starting time zero (compare Fig. 1). In addition, we assume the confinement time is fixed by the support plasma during a fast sputtering event [3,4] because the metal contaminant is a small perturbation. For example, we observe the total sum of all extracted uranium beam currents in [5] to be 0.41% of the summed supporting oxygen and nitrogen beam currents in steady state. Therefore, the recombination and ionization reaction rates are fixed within three time domains: t < 0,  $0 < t < \delta$ , and  $t > \delta$ . Then we may substitute  $dn_{i\pm 1}/dt$  with Eq. (6) to find

$$d^2n_i/dt^2 + \gamma_i dn_i/dt = (\beta_{i-1}\alpha_i + \beta_i\alpha_{i+1})n_i \qquad (8)$$

which is a second order differential equation in  $n_i$  by setting  $n_{i\pm 2} = 0$ . Making this approximation imposes that the ion density will not deviate more than one charge state away from  $n_i$ , and when coupled with the fact that the Charge State Distribution (CSD) of any real system is continuous the neglect of  $n_{i\pm 2}$  is constraining the system to have a sharp distribution around  $n_i$ .

We solve Eq. (8) with the form

$$n_i = c_1 e^{-t/\tau_{i-}} + c_2 e^{-t/\tau_{i+}} + c_3 \tag{9}$$

wherein  $\tau_{i\pm}$  may be solved in terms of rate coefficients of the form

$$1/\tau_{i\pm} = \gamma_i/2 \pm \sqrt{\gamma_i^2 + 4(\beta_{i-1}\alpha_i + \beta_i\alpha_{i+1})/2}$$
(10)

from a characteristic quadratic equation. All rate coefficients  $\alpha, \beta, \gamma$  are real and positive so therefore  $1/\tau_{i-} < 0$  and represents an effective growth rate when substituted into Eq. (9). The coefficient  $c_1$  is related to the density of  $n_{i+1}$ that feeds into the  $n_i$  charge state via stepwise ionization. However, since under the sharp CSD approximation  $n_{i-1}$ is small, it may be replaced by  $n_0(t)$  which is assumed a piecewise constant function in time. This allows for the ion density to increase exponentially up to a maximum at  $t = \delta$  wherein it must decay back down to zero in time. We estimate that  $c_1 = n_0(t)$  and  $c_2 = n_i(\delta) = n_{i0}$ . To avoid a recursive problem that requires numerical analysis we constrain  $c_2 = 0$  for  $\delta < t$  effectively turning off particle losses while the ion population is increasing. In reality, during the finite neutral pulse both  $c_1$  and  $c_2$  may be nonzero, and for analysis of the ionization times this error would need to be accounted for. The final coefficient  $c_3$  is chosen to force  $n_i(0) = 0$ .

#### Application to ECRIS

We imagine the extracted heavy ion beam current as a subset of the ion density lost from the plasma by its characteristic confinement time. Therefore the beam current follows a linear relationship with ion density [4] and should obey

$$I_i(t) = eQ\kappa \forall n_i(t) / \tau_i \tag{11}$$

with the beam line transmission  $\kappa$ , emission volume V, charge state Q, and electron charge e. Eq. (9) may be substituted into Eq. (11) for  $n_i(t)$  providing a platform to analyze

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Figure 1: Graphical representation of the piecewise constant solution for ion density.

the beam current decay curves plotted in [5]. Although  $\tau_i$ appears in Eq. (11) we compare the decay time constant to real fitted data because we do not need to sample the value of  $n_i$  and, the emission volume has a difficult geometry [6–8] to estimate. In a real system sputtering will produce a nonuniform spatial distribution of neutrals in the plasma, further convolving the prediction of a plasma emission volume.

The beam current decay after the neutral source is removed may be fitted with an exponential function of the same form as Eq. (9) and with  $c_1 = 0$ , the measured decay constant  $\tau_{im}$  may be set equal to  $\tau_{i+}$  and simplified into

$$\gamma_i = [1 - \tau_{im}^2 (\beta_{i-1} \alpha_i + \beta_i \alpha_{i+1})] / \tau_{im}.$$
(12)

Metals like uranium are not readily wall recycled by the plasma so when the sputter probe is grounded the neutral density is assumed to be small, and charge exchange is neglected. Effective rate coefficients in Eq. (12) may be expanded and solved for ion confinement time into

$$\tau_{i} = \tau_{im} [1 - \tau_{im}^{2} n_{e}^{2} (\langle \sigma_{r} \nu \rangle_{i \to i-1} \langle \sigma \nu \rangle_{i-1 \to i} + \langle \sigma_{r} \nu \rangle_{i+1 \to i} \langle \sigma \nu \rangle_{i \to i+1})$$
(13)  
$$-n_{e} \tau_{im} \langle \sigma \nu \rangle_{i \to i+1} - n_{e} \tau_{im} \langle \sigma_{r} \nu \rangle_{i \to i-1}]^{-1}.$$

In the simplest case if we neglect ionization and recombination entirely then  $\tau_i = \tau_{im}$ . Accounting for these effects decreases the denominator in Eq. (13) thereby making  $\tau_i > \tau_{im}$ .

#### SUMMARY

We demonstrate that the coupled ion density rate equation may be expanded about the  $i^{th}$  charge state and an exponential function satisfies the resulting second order differential equation. In doing so we have assumed the ionization and recombination rates are constant, the neutral density obeys a simple piecewise constant form, and ion densities beyond  $i \pm 1$  are zero. Applying this model to beam current decay curves measured from a fast sputtering method or similar [1, 2, 5], represent a lower bound for the ion confinement time of the sampled charge state in Eq. (13).

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