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efficiency of an ion source**

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### **Abstract**

We report a new method of measuring the absolute ionization efficiency of an ion source, which is independent of gas flow rate fed to an ionizer and of detection efficiency of the resulting ions. The method is based on the fact that when a pulsed alternate potential is applied between the ionizer and an electrode set outside near its exit hole, ions stored inside the ionizer while the potential is repulsive for the ions are flushed out as soon as it becomes attractive. It is shown that when the duty factor and repetition rate of the applied potential are properly chosen, the magnitude of this bunching effect can simply be expressed only by the ionization efficiency. Since its magnitude can quickly be evaluated, for example, by observing the time dependence of the bunched ion current in an oscilloscope, it provides a simple and reliable way for on-line diagnostics of the source.

## 1. Introduction

An ion source is now widely used for many fields of science and technology. The absolute measurement of its efficiency is often needed for improvement of the ion source itself as well as for measurement on some related physical quantities. However, it is often not easy to determine the ionization efficiency absolutely, because it requires absolute measurement on such quantities that may fluctuate from time to time and may thus involve various difficulties in precise determination; for example, they are gas flow rate fed to an ionizer as well as efficiencies of transporting, analyzing and detecting the resulting ions supplied from the source. This is especially the case when we ionize solid material or radioactivities produced in a thick target by nuclear reactions. In this note we report a new simple method of measuring the absolute ionization efficiency on-line without such difficulties.

The present method has been discovered as a by-product of our recent beam-bunching experiment [1] using a simple ion-source technique based on the following fact: When a pulsed alternate potential is applied between an ionizer and an electrode set near its exit hole as schematically shown in the upper part of fig. 1, ions are prevented from coming out of the ionizer while the potential is repulsive for the ions inside the ionizer, whereas the stored ions will be flushed out as soon as it becomes attractive. For convenience, we call hereafter a period during which the potential is repulsive or attractive a "duty-off" or "duty-on" period, respectively. By applying this method to a surface-ionization type ion source, Shirakabe et al. [1] observed a remarkable bunching effect for alkali elements and showed experimentally that the magnitude of this bunching effect was sensitive to the ionization efficiency as well as to the duty factor and repetition rate of the applied bunching potential.

A main purpose of this paper is to prove theoretically that when the duty factor and repetition rate of the bunching potential are properly chosen, the magnitude of the above bunching effect is simply expressed only by the ionization efficiency, and that this fact can be used for its reliable on-line measurement.

In section 2 we formulate the ionization efficiency and bunching effect to derive the theoretical basis of the present method. Some experimental examples are given in section 3. Characteristic features, limitation and validity of this method are discussed in section 4, while the present result is summarized in section 5. Some numerical calculations of relevant parameters introduced in this work are given in Appendix.

## **2. Theoretical formulation**

Throughout the present work we always consider an ion source consisting of a gas supply, an ionizer and an extraction electrode as schematically shown in the upper part of fig. 1. The gas supply mentioned above does not necessarily mean that we treat here only ionization of gaseous substance at normal temperature, but it includes cases for ionization of solid material which is slowly evaporated, for example, at high temperature. The present method can also be applied to the case of an isotope separator on-line (ISOL), in which radioisotopes produced in nuclear reactions are fed to the ionizer in the similar way. The material which we want to ionize is always assumed to be supplied to the ionizer with a rate of  $n_0$  atoms per unit time.

In subsections 2.1. and 2.2., we consider the case in which only singly-charged positive ions are present. The extension of the present method to the case of presence of multiply-charged positive and negative ions will be

described in subsection 2.3. To simplify the following arguments and formulae, we also temporarily assume that both ions and neutral atoms (or molecules) inside the ionizer can be extracted or escape outside from the ionizer only through its exit hole; at the end of subsection 2.2., however, we will show that the present conclusion is correct even without this assumption.

### 2.1. Ionization efficiency

Let us express the ratio of the number of ions of interest ( $N_i^{in}$ ) to that of the corresponding neutral atoms ( $N_0^{in}$ ) existing inside the ionizer as  $\varepsilon : 1-\varepsilon$ . In other words,  $\varepsilon$  is the proportion of the ions to the sum of the ions and neutral atoms. We then write the number of the ions ( $n_i^{out}$ ) and that of the neutral atoms ( $n_0^{out}$ ) escaping out of the ionizer per unit time as

$$n_i^{out}=N_i^{in}\lambda_i \quad \text{and} \quad n_0^{out}=N_0^{in}\lambda_0, \quad (1)$$

in which  $\lambda_i$  and  $\lambda_0$  are constants determined by the type and structure of the ion source as well as by its working condition, and are called here partial escape constants for the ions and neutral atoms, respectively. Note that the dimension of  $\lambda_i$  and  $\lambda_0$  is the inverse of time. Their rough estimate in the case of a cylindrical surface ionizer will be given in Appendix. The validity of eq. (1) will be discussed in some details in section 4.

The number of total relevant atoms ( $N$ ;  $N=N_i^{in}+N_0^{in}$ ) inside the ionizer is then determined by the following differential equation including a total escape constant  $\lambda$  defined below and the gas flow rate  $n_0$  ;

$$\frac{dN(t)}{dt} = n_0 - \lambda N(t), \quad (2)$$

with

$$\lambda = \varepsilon \lambda_i + (1-\varepsilon)\lambda_0. \quad (3)$$

We here consider the case in which no bunching potential is applied. In a steady state realized when  $t \rightarrow \infty$  (or  $t \gg 1/\lambda$ ), we obtain  $N(\infty) = n_0/\lambda$  and  $n_i^{\text{out}}(\infty) = N(\infty)\varepsilon\lambda_i = n_0\varepsilon\lambda_i/\lambda$ . The ionization efficiency ( $\eta$ ) of an ion source is generally defined as the proportion of the resulting ion beam extracted from the source to the gas flow rate fed to the ionizer, and can be written as

$$\eta = \frac{n_i^{\text{out}}(\infty)}{n_0} = \frac{\varepsilon\lambda_i}{\lambda}. \quad (4)$$

It should be noted that a value of  $\eta$  depends very much on values of  $\lambda_0$  and  $\lambda_i$ , thus differing from  $\varepsilon$  generally. In fact,  $\eta$  is equal to  $\varepsilon$  only when  $\lambda_i = \lambda_0$ , and it becomes close to 1 when  $\lambda_i \gg \lambda_0$ .

Although  $\varepsilon$  has been defined as a parameter describing the ratio of the numbers of the relevant ions and neutral atoms, it can be considered to represent consequences of microscopic ionization processes occurring inside the ionizer. Since  $\varepsilon$  is equal to  $N_i^{\text{in}}/(N_i^{\text{in}}+N_0^{\text{in}})$ , it can also be regarded, at least in some cases, as the ionization efficiency of the ionizer itself. In the case of a surface ionizer described in section 3, a value of  $\varepsilon$  can be estimated from the well-known Langmuir formulae [2]. On the other hand,  $\eta$  depends on the detailed structure of the ion source and represents the ionization efficiency of the whole ion source. We would like to emphasize that the real capability of the ion source is better expressed by  $\eta$  from the experimental point of view.

## 2.2. Bunching effect and principle of the present method

Let us now consider a bunching effect realized when an alternate potential is applied to the ionizer as shown in the upper part of fig. 2. We should note that  $\lambda_i = 0$  during the duty-off period, but that a value of  $\lambda_0$  during the duty-off period as well as values of  $\lambda_i$  and  $\lambda_0$  during the duty-on period are the same as those without the bunching potential. We are interested in a solution of eq. (2) for a steady state, which is time-dependent in this case because of the periodic change of the total escape constant.

We first consider the case in which the duty-on ( $t_{on}$ ) and duty-off ( $t_{off}$ ) periods are long enough compared with the corresponding "escape time". That is,

$$t_{on} \gg \frac{1}{\lambda} , \quad (5)$$

and

$$t_{off} \gg \frac{1}{(1-\varepsilon)\lambda_0} . \quad (6)$$

in which  $\lambda$  is equal to the total escape constant realized without the bunching potential and  $\varepsilon$  is assumed to be unequal to 1.

When the numbers of total atoms inside the ionizer at the end of each duty-off and duty-on periods are denoted by  $N_{off}(\infty)$  and  $N_{on}(\infty)$  respectively, they can be written, under the conditions of (5) and (6), as  $N_{off}(\infty) = n_0 / \{(1-\varepsilon)\lambda_0\}$  and  $N_{on}(\infty) = n_0 / \lambda$  as shown in the middle part of fig. 2. A steady-state solution during the duty-on period can be obtained by solving eq. (2) with the initial condition of  $N(0) = N_{off}(\infty)$ . That is,

$$N(t) = \frac{n_0 \varepsilon \lambda_i \exp(-\lambda t)}{(1-\varepsilon)\lambda_0 \lambda} + \frac{n_0}{\lambda} , \quad (7)$$

where time  $t$  is defined in such a way that  $t=0$  in the beginning of each duty-on period. The resulting ion current can be written as  $N(t)\varepsilon\lambda_i$ .

When we express the ratio of the ion current at the beginning of the duty-on period to that at the end of the same duty-on period as  $P : 1$  as shown in the lower part of fig. 2, we obtain  $P=N_{\text{off}}(\infty)/N_{\text{on}}(\infty)=\lambda/\{(1-\varepsilon)\lambda_0\}$ . Then, it is easy to show

$$\frac{P-1}{P} = \frac{\varepsilon\lambda_i}{\lambda} = \eta. \quad (8)$$

The present method is based on eq. (8), indicating that the ionization efficiency of the ion source is equal to  $(P-1)/P$ , which is independent of  $n_0$  and of other quantities such as efficiencies of transporting, analyzing and detecting ions of interest. Moreover, it enables on-line measurement of the ionization efficiency and is therefore sensitive to possible time-dependent change of working conditions of the ion source such as temperature and degree of outgassing.

When  $\varepsilon$  is close to 1, it may become impossible to fulfill the condition (6) in an actual measurement, although the condition (5) can be satisfied easily. When the condition (6) is not satisfied,  $N_{\text{off}}(\infty)$  should be written as

$$N_{\text{off}}(\infty) = \frac{n_0(\lambda - f\varepsilon\lambda_i)}{(1-\varepsilon)\lambda_0\lambda}, \quad (9)$$

with

$$f = \exp\{-(1-\varepsilon)\lambda_0 t_{\text{off}}\}. \quad (10)$$

Then, eq. (8) should be replaced with  $\eta=(P-1)/(P-f)=(P-1)/P+f/P$ , in which the approximation is valid for  $P \gg 1$ . If  $\lambda_0$  is nearly equal to  $\lambda_i$ ,  $\varepsilon$  is also close to  $\eta$  and can be approximated by  $(P-1)/P$ . Then,  $\eta$  can approximately be written as



$$\eta \approx \frac{P-1}{P} + \frac{\exp \frac{-\lambda_i t_{off}}{P}}{P} . \quad (11)$$

We should note that  $\lambda_i$  can be measured rather easily as reported in ref. [1].

We have so far assumed for simplicity that both ions and neutral atoms inside the ionizer can escape out of it only through its exit hole. Now we would like to show that eq. (8) can be derived without this assumption. When the ionizer has the other holes or leaks through which ions and neutral atoms can also escape outside, we write the sum of total escape constants due to those holes (leaks) as  $\lambda'$ . Then,  $\lambda$  appearing in the right side of eq. (2) should be replaced with  $\lambda + \lambda'$ , and the ionization efficiency of the source is given by  $\eta = \epsilon \lambda_i / (\lambda + \lambda')$ . Because the bunching potential is usually effective only to the exit hole, values of  $N_{off}(\infty)$  and  $N_{on}(\infty)$  defined before can be written as  $N_{off}(\infty) = n_0 / \{(1-\epsilon)\lambda_0 + \lambda'\}$  and  $N_{on}(\infty) = n_0 / (\lambda + \lambda')$ , if the conditions similar to those of (5) and (6) are satisfied. Then, we can easily conclude  $(P-1)/P = \eta$  with  $P = N_{off}(\infty) / N_{on}(\infty)$  also in this case.

### 2.3. Extension to multiply-charged ions

Since the extension of the present method so far described to the case of presence of multiply-charged ions can be made in almost the same way as given in the previous subsections, only its main procedure will be described here. For convenience, the following generalized terminology is used in this subsection. That is, the  $j$ -th multiply-charged ions mean: singly-charged negative ions when  $j=-1$ , neutral atoms when  $j=0$  and singly-, doubly-, triply-charged positive ions, etc. when  $j=1, 2, 3$ , etc.

Let us denote the number (normalized proportion) of the  $j$ -th multiply-charged ions of interest existing inside the ionizer by  $N_j^{\text{in}}(\epsilon_j)$ . Note that the sum of  $\epsilon_j$  over all possible  $j$  values is equal to 1. Using the partial escape constant  $\lambda_j$  defined in the similar way as in eq. (1), the total escape constant  $\lambda$  is written as

$$\lambda = \sum_j \epsilon_j \lambda_j. \quad (12)$$

Then, eq. (2) holds also in the present case. Noting that the extracted ion current of the  $j$ -th multiply-charged ions is  $n_0 \epsilon_j \lambda_j / \lambda$  in a steady state, its ionization efficiency ( $\eta_j$ ) can be written as

$$\eta_j = \frac{\epsilon_j \lambda_j}{\lambda}. \quad (13)$$

In order to obtain the relation between the magnitude of the bunching effect and the ionization efficiency, we now consider the simplest case, in which the bunching potential is applied to positive ions in such a way that  $\lambda_j=0$  for any positive value of  $j$  during the duty-off period. Then, assuming that the conditions similar to those of (5) and (6) are satisfied, that is, if  $t_{\text{on}} \gg 1/\lambda$  and  $t_{\text{off}} \gg 1 / (\sum_{j \leq 0} \epsilon_j \lambda_j)$ , it is easy to show

$$\frac{P_j - 1}{P_j} = \sum_{j>0} \frac{\epsilon_j \lambda_j}{\lambda} = \sum_{j>0} \eta_j, \quad (14)$$

where  $P_j$  is the ratio of the current of the  $j$ -th multiply-charged positive ions in the beginning of the duty-on period to that at the end of the duty-on period. It should be noted here that eq. (14) holds for any value of  $j$  ( $j>0$ ) appearing in the left side of this equation. That is, only the sum of ionization efficiencies of all multiply-charged positive ions of interest can

be determined from the bunching effect. In order to determine each absolute value of  $\eta_j$  separately, we need to know their relative intensity distributions. It can easily be obtained, however, from the observation of their currents at the end of the duty-on period.

It is clear that when the bunching is applied to negative ions, we obtain the relation similar to eq. (14) for negative  $j$ . However, we should note that negative ions significantly produced are only singly-charged in most cases; therefore, we are often able to determine their absolute ionization efficiency uniquely from the bunching effect only.

### 3. Experimental examples

Let us now apply the present method to some experimental results, which have been re-measured for this work in the same way as described in ref. [1]. The structure of a surface-ionization type ion source used is schematically shown in the lower part of fig. 1. It has originally been designed for an isotope separator on-line (ISOL) installed at a cyclotron of the Institute for Nuclear Study, University of Tokyo [3]. An ionizer is a 4 cm long cylindrical tube made of tantalum, of which the inner diameter is 3 mm. It was heated indirectly at  $2100 \pm 100$  K. To increase the ionization efficiency, we inserted, in the present measurement, 50  $\mu\text{m}$  thick rhenium and iridium foils around the inner wall of the tantalum cylinder, because they have larger electron work functions than tantalum [4] and are expected to yield larger values of  $\epsilon$  [2]. The rhenium foil was 4 cm long and was directly attached to the inner surface of the tantalum cylinder, while the iridium foil was 2 cm long and was placed onto the rhenium foil. A 20 kV high tension was applied between the ionizer and an extraction electrode. A pulsed alternate voltage as shown in the upper part of fig. 2 was applied

between the ionizer and a bunching electrode placed at 1 mm apart from the exit hole of the ionizer. The repetition rate and duty factor of the bunching potential used are 100 Hz and 20 %, respectively; that is,  $t_{on}=2$  ms and  $t_{off}=8$  ms. Only singly-charged positive ions have been observed significantly in the present source.

The values of  $(P-1)/P$  were deduced for  $^{39}\text{K}$ ,  $^{23}\text{Na}$  and  $^7\text{Li}$  ions. For this measurement a little amount of powder of NaCl, KCl or LiF was put in a small graphite capsule placed inside a target container, which was a 14 cm long cylinder made of tantalum. In order to evaporate the above material slowly, the target container was resistively heated at 850-1100 K by feeding DC current up to 250 A. The beam intensity of each ion was measured after the mass analysis with a Faraday cup placed near the focal plane of the ISOL and was observed in an oscilloscope. More details of the experimental set-up have been described in refs. [1,3].

As an example of the results, the measured  $^{23}\text{Na}$  ion current is shown in fig. 3. A sharp rise of the current in the beginning of the duty-on period is due to sudden extraction of ions that are stored inside the ionizer during the duty-off period. The observed current then decreases monotonically with the increase of time lapse and becomes nearly constant around the end of the duty-on period, being in accordance with eq. (7). The condition (5) is therefore satisfied. This turned out to be also the case for  $^{39}\text{K}$  and  $^7\text{Li}$  ions. From the observed slope in the decrease of the ion current, we have estimated the escape time of ions ( $1/\lambda_i$ ) to be roughly 250  $\mu\text{s}$  both for  $^{39}\text{K}$  and  $^{23}\text{Na}$  ions, which is consistent with that reported in ref. [1]. (Note that the reported value of about 180  $\mu\text{s}$  in ref. [1] corresponds to the "half-life" in the fall of the ion current, though not mentioned explicitly there.) In the case of  $^7\text{Li}$ , it looks shorter than the above value although it is difficult to determine it precisely due to the small bunching effect observed. Although the escape time of neutral atoms ( $1/\lambda_0$ ) cannot be determined in this measurement, we

expect that it is somewhat longer than, but not very different from  $1/\lambda_i$ , because there is no electric nor magnetic field inside the present ionizer, except for small electric field existing only near its exit hole. If we assume this, the condition (6) is also satisfied for  $^{23}\text{Na}$  and  $^7\text{Li}$ , while it is not fulfilled for the case of  $^{39}\text{K}$ , for which the calculated values of  $\epsilon$  are close to 1 as shown in Table 1.

The values of  $\eta$  have been deduced from eq. (8) for  $^{23}\text{Na}$  and  $^7\text{Li}$ , and from Eq. (11) for  $^{39}\text{K}$ . The results are listed in Table 1. In all cases, they lie between the two values of  $\epsilon$  calculated for rhenium and iridium ionizers. However, if one looks the more details, one can note that the deduced value of  $\eta$  for  $^7\text{Li}$  is somewhat smaller than the average of the two calculated values of  $\epsilon$ , whereas in the case of  $^{23}\text{Na}$ , it is slightly larger than the corresponding average. Although the reason for this difference is not clear, it is presumably due to the difference in the degree of outgassing of the ion source, to which the actual ionization efficiency is known to be very sensitive in the case of a surface ionizer.

#### 4. Discussion

Remarkable features of the present method of measuring the ionization efficiency of an ion source can be summarized as follows: (i) The method is very simple; only an oscilloscope is needed for quick evaluation of the ionization efficiency if it is larger than, say, 1 %. Digitalizing the time dependence of the ion current during the duty-on period would make it possible to apply the present method to the case of lower efficiencies. (ii) The method is reliable because it is independent of gas flow rate fed to an ionizer and detection efficiency of the resulting ions. (iii) Because the present

method enables real-time measurement of the ionization efficiency, it can be used for on-line diagnostics of the ion source.

Let us now consider the limitation and validity of the present method. Because the essential basis of the present formulation described in section 2 is eq. (1), we first discuss the validity of this equation. For this purpose, it should be noted that gas quantity inside the ionizer generally changes to some extent by the application of the bunching potential, even if the other working conditions of the source are kept the same. Although eq. (1) can be considered as definition of  $\lambda_i$  and  $\lambda_0$ , it is meaningful only in the case, in which values of  $\lambda_i$  and  $\lambda_0$  are not sensitive to the expected change of the gas quantity. When this condition is satisfied, at least approximately, the proportionality of  $n_i^{\text{out}}$  ( $n_0^{\text{out}}$ ) to  $N_i^{\text{in}}$  ( $N_0^{\text{in}}$ ) is guaranteed within the expected change of the gas quantity. This is considered to be the case as far as we treat an ion source requiring very low gas pressure inside the ionizer, such as a surface ionizer described in this work. However, in some ion sources like a high-power plasma source, for which the relatively high gas pressure is often used, it is questionable whether the above condition is fulfilled. In such a case, careful experimental study is needed about the validity of the method. We believe that eq. (14) is useful for this purpose, because it is one of the most characteristic conclusions of the present consideration.

There are two practical problems in the application of the present method to actual ion sources. One is that electrons existing inside the ionizer near its exit hole feel some electric field penetrated from outside through the exit hole, and that they are forced to go out of the ionizer while the bunching potential is attractive for them. This may change considerably the state of possible plasma, which is important in some type ion sources. Therefore, caution should be taken to keep the magnitude of the bunching potential to

the minimum, which can easily be tuned by observing ion beam during the duty-on and duty-off periods in an oscilloscope, for instance.

Another problem is that the use of the bunching electrode may cause small change of electric field near the exit hole of the source, resulting in slightly different optical effects to the extracted ions, compared with the case of no use of the bunching electrode. Strictly speaking, this is supposedly the case in the experimental set-up described in section 3. However, because in this experiment the bunching electrode was set only 1 mm apart from the exit hole, it was considered not to change the ionization efficiency seriously. Any way, the above two facts may disturb the working condition of ion sources to some extent, and may limit the usefulness of the present method in some cases.

## **5. Conclusion**

For an ion source consisting of a gas supply, an ionizer and an extraction electrode, the ionization efficiency ( $\eta$ ) of the source can be written by use of the escape constants defined in eqs. (1) and (3). The bunching effect of ion-beam realized by a pulsed gating-potential method described in ref. [1] can simply be written only by  $\eta$ , when the duty factor and repetition rate of the applied bunching potential are properly chosen. It provides a novel way of measuring the ionization efficiency, which is independent of gas flow rate fed to the ionizer and detection efficiency of the resulting ions. This method has successfully been applied to a surface-ionization type ion source. The present method is considered to be valid when the partial escape constants are insensitive to relatively small change of the gas quantity inside the ionizer caused by the application of the bunching potential.

## Appendix

The partial escape constants defined in subsection 2.1. depend very much on the type of ionizers and the working condition of ion sources. Their rough estimate will be given here, as an example, for a cylindrical surface ionizer as shown in the lower part of fig. 1

We consider neutral atoms (or molecules) and ions homogenously distributed inside a cylindrical ionizer, of which the inner radius and length are  $a$  and  $d$ , respectively, with the condition of  $d \gg a$ . The inner radius of the exit hole is also equal to  $a$ . For simplicity, we assume here  $\lambda_i = \lambda_0$ . This is the case, at least approximately, when there is no electric field inside the ionizer.

Because  $d \gg a$ , the solid angle of the exit hole seen from those atoms (or ions) that have the same cylindrical coordinate  $z$  ( $z$  is the distance measured from the center of the exit hole along the symmetrical axis of the cylinder) is  $\omega(z) = 2\pi\{1 - z/(z^2 + a^2)^{1/2}\}$ . Then, the probability that atoms located at the coordinate  $z$  escape outside directly through the exit hole is  $p(z) = \omega(z)/4\pi$ , assuming that no collision takes place with other atoms during their flight. Its average is given by

$$p^{av} = \frac{\int_0^d p(z) dz}{d} \approx \frac{a}{2d}, \quad (A.1)$$

where the term proportional to  $(a/d)^2$  is neglected. If we put  $d=40$  mm and  $a=1.5$  mm in eq. (A.1), which are the case in ref. [1] as well as in this work, we obtain  $p \approx 0.019$ . The average flight time ( $t_z^{av}$ ) of those atoms is written as

$$t_z^{av} = \frac{\int_0^d zp(z) dz}{v_z^{av} \int_0^d p(z) dz} \approx \frac{a\{\ln(2d/a) - 0.5\}}{2v_z^{av}}, \quad (A.2)$$



where  $v_z^{av}$  is the averaged z component of velocities of the atoms of interest.

Most of the atoms moving in different directions collide with the inner wall of the ionizer and may be adsorbed for a while. Therefore, some delay time ( $t_d$ ) is needed before they become ready to try to escape again. Repeating this process many times, each atom finally escapes from the ionizer with an escape time ( $\tau$ ) given by

$$\begin{aligned}\tau &= pt_z^{av} + (1-p)p(t_d+t_z^{av}) + (1-p)^2p(2t_d+t_z^{av}) + (1-p)^3p(3t_d+t_z^{av}) + \dots \\ &= t_z^{av} + \frac{(1-p)t_d}{p} .\end{aligned}\tag{A.3}$$

The escape constant is the inverse of  $\tau$ . The delay time  $t_d$  can be expressed as the sum of the averaged flight time ( $t_r^{av}$ ) of atoms moving to the wall of the ionizer and the release time ( $t_{rel}$ ) of the adsorbed atoms at the wall; that is,

$$t_d = t_r^{av} + t_{rel}.\tag{A.4}$$

Which terms in the right side of eq. (A.4) are dominant depends very much on the properties of gas atoms (or molecules) as well as the structure, material and temperature of the ionizer.

To have a rough idea how much  $\tau$  is, let us consider  $^{39}\text{K}$  atoms as an example. Assuming that atoms inside the ionizer are heated at the same temperature as the ionizer, for example, at 2100 K as is the case in this work,  $v_z^{av} \approx 6.7 \times 10^5$  mm/s and  $v_r^{av}$  (averaged radial velocity)  $\approx 9.4 \times 10^5$  mm/s. For  $a=1.5$  and  $d=40$  mm, we obtain  $t_z^{av} \approx 3.9$   $\mu\text{s}$  and  $t_r^{av} = a/v_r^{av} \approx 1.6$   $\mu\text{s}$ . By using  $p \approx 0.019$ , it yields  $\tau \approx 87$   $\mu\text{s}$  in the case of  $t_{rel} = 0$ . This value is about three times shorter than the observed value of about 250  $\mu\text{s}$  as described in section 3. To obtain the agreement between the experimental and calculated values, we

need to assume  $t_{rel}=3.2 \mu s$ . However, it should not be taken too much serious, because the actual experimental geometry is more complicated due to the use of the rhenium and iridium foils inserted in the ionizer as well as due to a transfer tube between the ionizer and the target container as shown in the lower part of fig. 1.

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Table 1

The measured values of ionization efficiencies of the ion source ( $\eta$ ) together with values of  $\epsilon$  calculated for rhenium and iridium ionizers by the Langmuir formulae [2].

Ion	$\eta^a$ (%)	Ionization Potential <sup>b</sup> (eV)	$\epsilon^c$ (%)	
			Re	Ir
<sup>39</sup> K	95±4	4.341	93.9±0.8	98.8±0.2
<sup>23</sup> Na	38±2	5.139	15.7±0.5	50.8±0.7
<sup>7</sup> Li	8±1	5.392	4.4±0.4	20.3±0.4

<sup>a</sup> The given errors originate from uncertainties in determining the magnitude of the ion current in such photographs as shown in fig. 3.

<sup>b</sup> Taken from ref. [5].

<sup>c</sup> Calculated by using the following electron work functions for polycrystallines reported in ref. [4]; 4.96 eV for rhenium and 5.27 eV for iridium. The given errors correspond to ambiguities of the measured temperature.

## Figure Captions

Fig. 1 Simplified structure of an ion source having a bunching electrode together with essential parts of a surface ionizer used in this work. Note that heavy heat shields and various electrodes of the actual ionizer used are not shown for simplicity.

Fig. 2 An example of the applied bunching potential as well as schematic illustration of the resultant gas quantity inside the ionizer and extracted ion current realized under the conditions of (5) and (6) described in the text.

Fig. 3 The measured  $^{23}\text{Na}$  ion current observed in an oscilloscope when the bunching potential was applied as indicated in the figure.

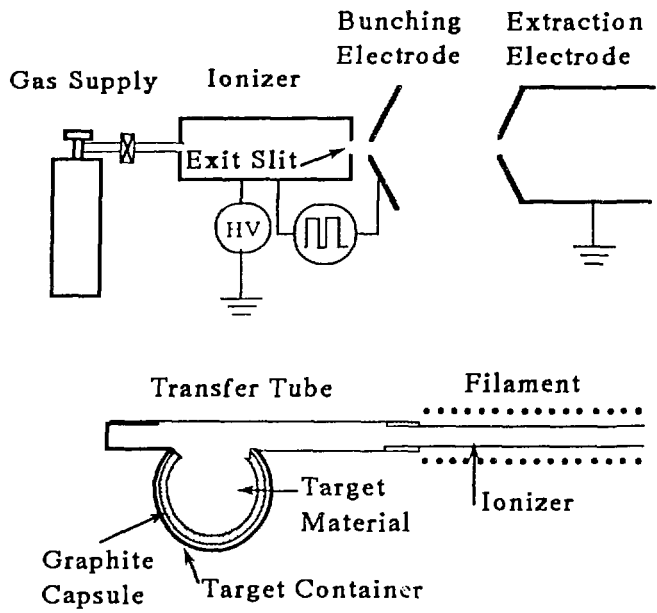


Fig. 1

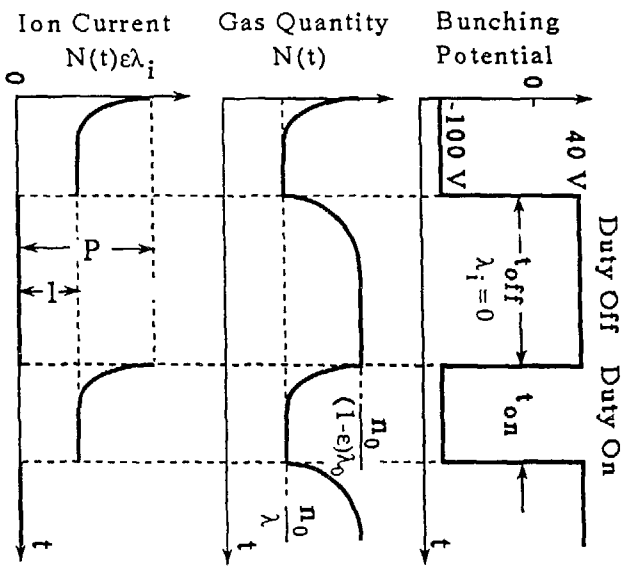


Fig. 2

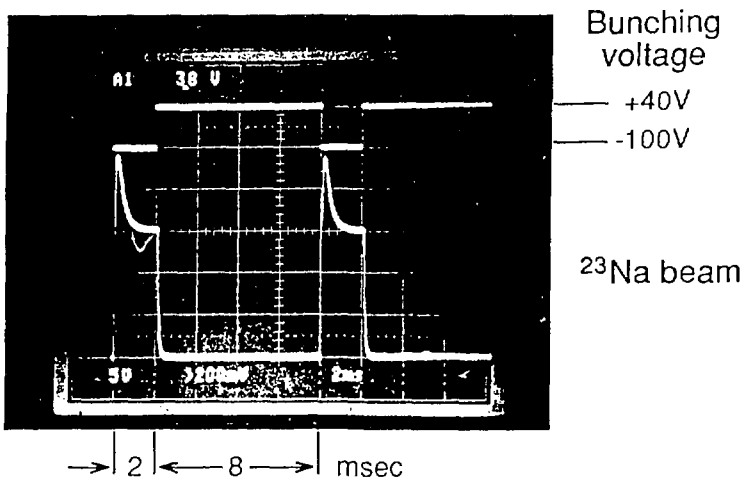


Fig. 3