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**AN ION SOURCE WITH STORAGE CAPABILITY FOR BUNCHED
BEAM RELEASE AND CONTROLLED CHEMICAL SEPARATION**

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(Submitted to Nucl. Instr. Meth. A)

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**AN ION SOURCE WITH STORAGE CAPABILITY FOR
BUNCHED BEAM RELEASE AND CONTROLLED CHEMICAL SEPARATION**

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Abstract

A discharge ion source of the FEBIAD-type is described which allows accumulation of various elements in a cooled trap and release of the stored atoms after a preselected time in a defined bunch. This bunching effect is systematically investigated for the elements Cu - Ge, Pd - Sb and Au - Bi as a function of temperature and of the trap materials graphite, zirconium, niobium, molybdenum and tantalum. The accumulation/release characteristics and the underlying adsorption/desorption mechanism are qualitatively explained in terms of the temperature and enthalpy of adsorption; the measured sequences of the elements may, with some reservation, be used to test theoretical predictions. Utilizing the differences in accumulation efficiency and release profile for neighbouring elements, a high degree of chemical selectivity may be achieved. This feature, important for mass-separation on-line, is illustrated by a test in which neutron-deficient Sn isotopes were investigated α -spectroscopically with substantially reduced isobaric contamination of In, Cd and Ag. Additionally, the bunching itself strongly increases the peak-to-noise ratio in collinear laser spectroscopy, permitting the study of isotopes hitherto inaccessible due to low production rates. The extension of this new method to other elements and to higher release speed is outlined.

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1. Introduction

Investigations of nuclear and atomic properties of nuclei with great excess of neutrons or protons in isotope separation on-line (ISOL) are often limited by background problems. This paper describes a method, which in some cases masters the two main sources of too low a signal-to-background ratio: too low rates of production and/or too high an isobaric contamination. The method is to accumulate the species of interest in a cold area, maintained for a certain time within the ion source, and to subsequently release them in a defined way, called "bunching", by heating up the cold area.

Using thermochromatographic techniques in ISOL is by no means new. They have been applied at the collector side of mass separators and are an intrinsic option of any on-line ion source. The fact that some elements are easily, and some are only badly, or not released from the catcher-ion-source-system is considered and used in practically any ISOL experiment. Also controlled application of specific temperatures and materials of the ion source to increase or decrease yields of wanted and unwanted species is common-place.

What is new in the method described here and - to our knowledge - applied for the first time is the possibility to vary the release efficiency as a function of time in a controlled way with repeated storage-release cycles. Thus the beams of interest extracted from the ion source may be bunched into time intervals short compared to accumulation periods, which are selectable over a wide range and usually only limited by the half-life of the nuclei of interest. Since both the efficiency of an accumulation/release cycle and the temporal structure of a bunch are element-specific, chemical selective and efficient release is not only reached by a more or less efficient suppression of unwanted activity, but also by setting an adjusted time window for collection of the interesting activity. In analogy to spatial separation in conventional thermochromatographic columns this technique yields "chronological thermochromatographic separation".

The bunched release of atoms, produced in a given time, leads consequently to higher beam intensities during the much shorter release interval. For experiments using the radioactive beam itself, e.g. collinear laser spectroscopy, bunching yields improved signal-to-noise ratios. This is in general not the case for experiments which use the radioactive beam to

produce point-like sources for nuclear spectroscopy and where collection and counting can be performed simultaneously. Here only the background may be considerably reduced, discriminating unwanted isobars by the controlled chemical separation mentioned above. In special cases, however, where the collection of a source prevents or limits simultaneous investigation and should therefore be kept short, bunched beam release may be advantageous, e.g. for nuclear orientation studies at low temperatures, where simultaneous implantation and measurement at very low crystal temperature may be in conflict.

In chapter 2 the principles of adsorption (the process responsible for accumulation) are discussed, followed by the description of the ion source and the cooling/heating device in chapter 3. In chapter 4 the accumulation properties of various materials for the cold area are studied off-line, using charge material and beams of stable isotopes of various elements. The implications of the results on bunching performance and chemical selectivity are compiled in chapter 5 for practical use. In chapter 6 two types of ISOL applications - collinear laser spectroscopy and α -spectroscopy of neutron-deficient isotopes in the area of Sn and below - demonstrate the possibilities opened by bunched beam release.

For the sake of clarity, the convention is used in this paper where chemical elements, which are constituents of the ion source (electrodes, adsorbing surfaces etc.), have their name fully written. Chemical symbols are used for elements, which are handled by the ion source (charge material, control gas, adsorbing atoms, ion beams, etc.).

2. Principles

A system of gaseous atoms at low pressure in presence of a metal surface of temperature T represents an equilibrium state of free particles and particles adsorbed to the surface. The interaction between gas and surface can be essentially described by two simple relations, in spite of the fact that the mechanism of adsorption is very complex involving a great number of physico-chemical parameters¹⁾.

The mean contact time t_a of a particle at the surface is given by the Frenkel-equation

$$t_a = t_0 \exp(E_D/kT), \quad (1)$$

where E_D is the heat of desorption and t_0 a surface-dependent constant of the order of 10^{-13} s. The distribution of a given number of particles between gaseous phase (N_g) and surface (N_a) is given by^{2,3)}

$$N_a/N_g = K \exp(\Delta H_a/kT) \equiv \epsilon_A \quad (2a)$$

$$\eta_A \equiv N_a/(N_a + N_g) = \epsilon_A/(1 + \epsilon_A) \quad (2b)$$

For ΔH_a , the enthalpy of adsorption, tabulated values are available, calculated on semi-empirical basis for a great number of both atoms to be adsorbed and adsorbing surfaces¹⁾. The physical meaning of η_A is that of an efficiency of adsorption, also referred to as storage or accumulation efficiency in the subsequent chapters.

Under the experimental conditions of this study, adsorption is the only relevant process for storage, which is thus solely governed by the temperature of the system and by ΔH_a , the measure of the binding strength between atom and surface. The element-specific vapour pressure plays no role for the storage efficiency, as long as the particle density is sufficiently low to approximate the situation of a zero-covered surface. This also has been found in an investigation of the desorption of lanthanides from tantalum surfaces⁴⁾ and is clearly demonstrated by the results presented in chapter 4. For example, considering the elements Cu and As which have extremely different data of ΔH_a (on a tantalum surface) and vapour pressure: Cu ($\Delta H_a = 4.0$ eV, $P_{1400K} = 10^{-3}$ mbar); As ($\Delta H_a = 6.2$ eV, $P_{1400K} > 10$ atm): At a tantalum surface of 1400 K the adsorption is nearly negligible for Cu, but practically total for As in agreement with the ΔH_a -values and in contrast to the vapour pressures.

3. Experimental

The FEBIAD-B2 ion source was chosen to test the possibilities of bunched beam release. With its high ionization efficiency ($\approx 50\%$ for Xe) and its emittance of 10 mm mrad it is well suited for collinear laser spectroscopy for which the method was primarily developed. The typical operation temperature of 1770 K is in the right range for the elements of most interest:

Ag, In, Sn, Sb, Tl, Pb and Bi. Also the rugged structure of the source withstands well the mechanical stress imposed by manyfold coupling of a cooling block. The feature of FEBIAD sources to operate with constant efficiency over a wide pressure range⁵⁾ is certainly an essential pre-requisite in a system, where fast and strong variations of partial pressures are to be expected.

Since design considerations, operating conditions and performance of the source were detailed in a preceding paper⁶⁾, the description here is restricted to the aspects relevant for the bunching system. The lay-out of the device is shown in fig. 1. Most of the design criteria are imposed by general considerations: The characteristics of accumulation and release are governed by the temporal dependence of the temperature of the cold area. The efficiency of accumulation depends on the *cold spot temperature*, i.e. on the way *cooling* is performed. The release speed depends on the *temporal increase of the temperature* when cooling is removed, i.e. on the means of *heating* the cold area.

To create the cold area, a pocket, reaching radially out from the heat-shield containment of the source, is screwed into the anode opposite to the entry for the accelerator beam. A flexible design in both heating and cooling was chosen. Two types of water-cooled, pneumatically driven cooling blocks enable considerably different *cold spot temperatures* of the pocket:

Weak cooling: The cooling block is small and just couples to the end of the pocket.

Strong cooling: A massive block with high heat capacity and good thermal contact to the cooling water embraces a large fraction of the pocket.

The *temporal increase of the pocket temperature* and its upper equilibrium (uncooled) are determined by the position of the pocket within the ion source and the power of extra heating:

Colder position (A): Here the pocket is mounted on the *Anti-cathode-side* of the anode cylinder and thus shielded from the thermal radiation of the cathode, as shown in fig. 1. Since in this configuration the accelerator beam can traverse the ion source, the target/catcher(s)

may be arranged in the pocket proper. This way the heating power of the incident beam may be used to influence the release speed with the option of controlled heating, i.e. choosing accelerator currents different during storage and release period.

Hot position (C): By reversing the mounting of the anode cylinder the pocket is brought to the *Cathode-side* of the ion source, where both entrance area and pocket are heated by the intense thermal radiation (and electron bombardment) from the cathode.

Additional heater: The small size of the weak cooler permits placing a heater concentrically around the pocket (see fig. 1), which may be powered up to 250 watts in the release period. Actually, it can also be used with any power setting between 0 and 250 watts during accumulation to reduce the storage efficiency of unwanted, weakly accumulated elements still further, thus improving chemical selectivity.

It is useful for the following to differentiate the essential thermal states of the pocket:

Hot: This is the basic state, where the pocket is neither heated nor cooled and adopts a temperature given by thermal conduction and radiation from the ion source. This temperature has been determined pyrometrically to be 1390 ± 30 K and 1530 ± 30 K at the backside of the pocket, when placed in the colder or hot position of the source, respectively.

Heated (only in case of weak cooling): When the heater is operated at 250 watts the temperature increases from 1390 K to an equilibrium temperature of 1770 ± 30 K after 12 s (measured only for position A).

Cold: This is the "cold spot" state of the pocket when cooling is coupled. These temperatures cannot be measured pyrometrically but are, under somewhat questionable assumptions, accessible indirectly (see chapter 4.1): for weak cooling 840 K and 890 K in the colder and hot position of the source, respectively, and 620 K for strong cooling.

Two thin-walled tantalum capillaries are attached radially to the ion source. The one from top is a gas line which supplies the ion source with a

constant flow of Kr or Xe from calibrated test leaks with typical outputs of 10^{-6} std.cm³/s. This is used to verify that a constant ionization efficiency exists over the total storage/release cycle, which is generally achieved within variations smaller than 5 %. The second tube from bottom is an oven line. Using the temperature gradient along the tube from room temperature to ≈ 1770 K, the temperature of the anode cylinder, defined fluxes of all elements of interest can be produced by placing small quantities of bulk material at an adjusted distance from the ion source. The right position is easily found using vapour pressure data; in contrast to what is written in chapter 2, evaporation from a macroscopic sample is of course governed by the vapour pressure. Since heating is possible by passing a DC-current through the two capillaries, positioning of the samples is less critical and allows variation of the flow into the ion source. After some experience it was possible to achieve reasonable fluxes of up to 12 elements simultaneously so that, for a given configuration, all 16 elements considered here could be tested changing the oven line only once.

The off-line experiments were made under the following standard conditions: The ion source with an orifice diameter of 1 mm was always operated with the same power settings: filament 4V/25A, electron bombardment heating of the cathode 1A/150V, anode 150V/0.8A, outlet plate 125V/0.15A. The stable elements were introduced in atomic form. No compounds were used because of the uncertainty whether dissociation happened before or after adsorption/desorption. The fluxes from the oven line into the ion source are not accessible to measurement but must, for volatile species, correspond closely to the respective ion currents leaving the source due to the high ionization efficiency. The ion currents ranged from 10 pA to 10 nA. These rates are sufficiently low to approximate the condition of zero covering, mentioned in chapter 2, for accumulation times up to 1000 s, since the pocket can store about 10^{15} atoms/cm² in a monolayer. For the tests an accumulation time of only 60 s was chosen. When weak cooling was applied the external heater was operated at zero watts during storage and at 250 watts for release, starting 3 s before the removal of the cooling block. The investigations were performed at the GSI on-line isotope separator⁷⁾.

For the measurements the ion current of a specific isotope was recorded. When cooling is switched on, the constant current I_H of the *hot* state drops to I_C , the current of the *cold* state. This (exponential) fall-off

is slightly element-specific, the corresponding half-times being 0.06 - 1 s in case of high accumulation efficiencies. When cooling is removed after 60 s, the current rises to a maximum exceeding I_H more or less substantially - the bunch - returning asymptotically to I_H . The bunching properties can be characterized by essentially three numbers:

The storage efficiency η_A is given by $(I_H - I_C)/I_H$, or by the ratio of integral bunch intensity to I_H integrated over 60 s. The bunches show a more or less steep rise to a maximum, and an exponential fall-off afterwards. The former reflects the temporal increase of the pocket temperature, a relative measure of which is given by the delay of the bunch maximum Δt_M from the moment of cooling removal. The latter is governed by the multiple adsorption/desorption processes of the released atoms in the total enclosure of the ion source (including the *hot* or *heated* pocket), before effusing through the outlet orifice: the resulting fall-off half-time is denoted τ_B . The lower limit for τ_B is the intrinsic delay of the ion source of 50 ms for medium mass atoms ($A \approx 100$). This number follows from molecular flow calculations assuming zero hold-up time in each of the approximately 3000 wall collisions before effusion.

4. Off-line Tests With Stable Isotopes

For the systematic study tantalum was chosen as the pocket material. Tantalum surfaces are present throughout the ion source (except for the boron-nitride insulators and small parts of highly purified graphite) and thus interpretation of the bunch shapes is not complicated by several surface materials. Tantalum also permits easily detachable joints if intermittent parts made from graphite are used. The less extensive tests performed with pockets made of graphite, zirconium, niobium and molybdenum are described in section 4.2.

4.1 Investigations With Tantalum Pockets

The adsorption features of the elements Cu, Zn, Ga, Ag, Cd, In, Sn, Sb, Tl, Pb and Bi on the surface of a tantalum pocket were studied for all possible configurations of heating and cooling. The relevant quantities η_A , Δt_M and τ_B are plotted in fig. 2 as a function of semi-empirical ΔH_a -values

from the systematics of Roßbach and Eichler¹⁾. Typical bunch profiles for the group Ag - Sb are presented in fig. 3. As expected the bunching properties are affected by the configuration and are strongly element-specific. The elements may be classified by several categories:

A. Elements with very low ΔH_a , where η_A is smaller than 5×10^{-5} , the experimental detection limit: The noble gases Kr or Xe, used for control of the ion source, belong naturally into this class as well as e.g. Cd under the conditions shown in fig. 3 A - C.

B. Elements, which are found in the rising part of η_A -curve above the detection limit: They show strongly increasing η_A -values with increasing ΔH_a and are released in very short but ineffective bunches. η_A is strongly affected by small differences in the **cold** state temperature. Since with a switchable cooling/heating system good temperature reproducibility from-ion-source-to-ion-source is impossible, considerable deviations in η_A have to be accepted, which leave, however, the sequence of the elements untouched. The typical scatter is indicated in fig. 2 by vertical bars for the cases where several measurements with different sources were performed. Representative for class B are e.g. Cd in fig. 3D and In, Ag in fig. 3A, B.

C. Elements which are found around and above the edge of the η_A -curve: They are bunched efficiently, the FWHM of the bunches ranging from 0.8 s upwards. Typical examples are In and Ag in fig. 3C, D and Sn, Sb in fig. 3A, B.

D. Elements with $\Delta H_a \geq 4.5$ eV for the colder and ≥ 5 eV for the hot position of the ion source: Here already the **hot** pocket starts to be a cold area which adsorbs to increasing degree. The resulting bunches contain only a fraction of the accumulated input (especially when no extra heating is or can be applied) and have surprisingly short fall-off times. The latter is due to the fact that during accumulation these elements are stored on a wider area of the pocket. When cooling is removed one part of the surface (cylindrical part) gets sufficiently hot for desorption, while the other (backside) remains cold spot, re-adsorbing a good fraction of the particles released from the hotter areas of the pocket. To this class belong e.g. Sn and Sb under the conditions of fig. 3D.

E. Elements with $\Delta H_a \geq 6$ eV: Here even the **heated** pocket and probably other parts of the ion source (outlet plate, window area) start to act as cold areas. The respective ion currents are not affected by cooling and can thus only be tiny fractions of the input. These ion currents remain for several days after the input flow has stopped ("memory effect").

The elements Hg ($\Delta H_a = 1.5$ eV), Au (5.7 eV), Ge (6.0 eV), As (6.2 eV), and Pd (6.5 eV), are not plotted in fig. 2. Their adsorption behaviour was investigated, but belonged under all conditions to the categories A (Hg), D (Ge and Au) and E (As and Pd). Hence the data can not be properly interpreted in terms of η_A , Δt_M and τ_B .

The gross trend (see fig. 2) of both $\varepsilon_A = \eta_A/(1-\eta_A)$ and the quantities characterizing the temporal dependence of the bunch, Δt_M and τ_B , is exponential with increasing ΔH_a , which is to be expected from the considerations in chapter 2. The normalized quantity η_A is plotted because of the difficulty to determine ε_A -values much higher than 10. This is partly due to the problem of peaks-at-every-mass^{8,9)} of unknown affiliation and with intensities in the range of 10^{-12} to 10^{-9} A, which often allows only the determination of a lower limit for η_A -values above 80 %. Another reason is the 2 - 5 % chance (depending on the size assumed for the cold area) of "free" particles to effuse through the outlet orifice without having contacted the cold zone, which limits the maximum value, η_A can reach, to 95 - 98 %.

The fits to the η_A -data in fig. 2 were performed using the **cold** state temperatures of the pocket, which could, however, not be determined directly. Pyrometry from the rear side is not possible due to the coupled cooler and is forbidden from the other side through the ion source, since reflected light from the substantially hotter ion source covers thermal radiation completely. Thermocouple measurements are excluded due to space problems. The following consideration permits, however, an indirect determination: Between Ge and As the point is reached where the pocket, when mounted in the colder position, is an almost perfect "cold area" even in the **hot** state. Using the known temperature of 1390 ± 30 K and assuming $\varepsilon_A = 10$ for $\Delta H_a = 6.1 \pm 0.1$ eV, the constant K of eq. (2) is calculated to $K = (7.8 \pm 2) \times 10^{-22}$. The respective ΔH_a -values for $\varepsilon_A = 10$ in the **cold** states can be traced from fig. 2. They are in case of strong cooling 2.7 ± 0.1 eV, and in case of weak cooling 3.7 ± 0.1 eV and 3.9 ± 0.1 eV

for the colder and the hot position of the ion source, respectively. The corresponding temperatures for the **cold** states are: 620 ± 30 K, 840 ± 30 K and 890 ± 30 K. These values fix the slope of the normalized exponential fits in fig. 2, which are only adjusted to the edge of the η_A -versus- ΔH_a -dependence. That the η_A -data below the edge are reproduced well, demonstrates self-consistency and gives confidence in the fitting procedure.

In contrast to the η_A -data the values for Δt_M and τ_B are measured under conditions with unknown and varying temperatures (the "appearance temperatures" of the bunches are somewhere between the **cold** and the **hot** or **heated** state temperatures of the pocket) and do therefore not permit a quantitative analysis*). They give, however, additional evidence for the relative sequence of the elements in the ΔH_a -scheme.

The data in fig. 2 indicate that some of the semi-empirical ΔH_a -values should be revised. This is most obvious for Ga, which should appear between Bi and Cu, and for Tl, which belongs just below Pb. These shifts are indicated in fig. 2 by horizontal arrows. Minor revisions concern Ag and In, which show up in reversed sequence and Sn and Sb, the ΔH_a -values of which appear to be too high by $0.3 - 0.5$ eV.

In spite of the good agreement between the experimental data and the systematics, one reservation should be kept in mind: Aim of this study was not to determine enthalpies of adsorption, especially in view of the difficulties, which experts seem to have to yield reproducible numbers (see e.g.

*) Were these temperatures known, the τ_B -values could be used to determine the mean contact times t_a per individual wall collision and possibly absolute values for the heat of desorption by equation 1. This is, however, only possible for the elements Sn and Sb, where the falling part of the bunch extends to times at which the pocket has reached its known upper equilibrium temperature. Especially simple is the case of the **heated** pocket in the colder source position, where ion source (excluding cathode) and pocket have the same well-established temperature of about 1770 K. Since the mean number of wall collisions is calculated to approximately 3000 inside the total enclosure, the measured τ_B -data yield t_a -values of 1.9 and 4.3 ms for Sn and Sb, respectively, via the relation $t_a = \tau_B / (3000 \ln 2)$.

the compilation in ref. 10) even under conditions preferable to a discharge ion source, where all kinds of impurities may prevent clean surface conditions. The measurements here only determine the sequence in which the elements desorb, but do not tell, which data points are to be shifted and which reside. For the intention of this study, the sequence in a relative scale is actually all what is needed: to compare the adsorption characteristics of different elements and on different surfaces (section 4.2). Since the semi-empirical ΔH_a -systematics existed and fitted relatively well, it was only natural to use it as a basic scale. The "experimental" ΔH_a -values thus extracted, are certainly characteristic for the used apparatus. Whether they reflect, beyond that, the properties of pure surfaces, has to be left open.

4.2 Test of Other Pocket Materials

The previous section has shown that more than half of the elements considered may be effectively bunched, but also that the others remain outside the range useful for bunching. This fact enables the possibilities to achieve chemical separation outlined in chapter 5. Nevertheless, bunching is a desirable option by itself. There are principally two ways to reach effective bunching for the elements, which are either too volatile or too refractory. Equation (2) shows that adsorption properties may be influenced by either temperature T or enthalpy of adsorption ΔH_a .

To influence adsorption via the temperature is the more difficult way. It requires lower **cold** state temperatures for elements of class A and B and higher temperature for both ion source and **hot** pocket for elements of class D and E - a requirement difficult to fulfill without considerable modification of the device.

An easier solution is to look for pocket materials that do either increase ΔH_a for elements too volatile or decrease it for too refractory elements. According to the systematics¹⁾, three other materials show promising surface properties (see fig. 4 left side). Compared to tantalum, the ΔH_a -values for a niobium surface are generally lower and for zirconium generally higher. Molybdenum shows the most remarkable feature; it seems to compress most of the elements into the range useful for bunching. To

what extent the experimental results reconcile with these expectations is subsequently described and presented on the right side of fig. 4.

The tests performed with pockets made of graphite, zirconium, niobium and molybdenum were less extensive than for tantalum. For economical reasons the source electrodes were left in tantalum (for exceptions see below under molybdenum and graphite pockets). This situation confronts the tests with a two-material problem, excluding the measured τ_B -values from analysis (the fall-off of the bunch is governed by the total enclosure, i.e. all internal surfaces contribute to it). Fortunately this is not the case for the other temporal quantity Δt_M , since the rising part of the bunch projects the increase of the partial pressure in the ion source, which is governed by the release characteristic of the pocket. The tests were restricted to the colder position of the ion source in order to i) minimize corrosion and ii) prevent coating of the pockets with layers of tantalum evaporated from the cathode (in position A the pocket is out of direct "sight" of the cathode). Among the pocket materials investigated, only tantalum, niobium and graphite were known to cause no corrosion problems at the joints. The features of zirconium were not known at all and molybdenum was known to react strongly with graphite above 1900 K. All pockets, however, turned out to be easily detachable and showed no mechanical damage except for zirconium, where deformations on the thin-walled cylindrical part of the pocket were observed.

Niobium pockets have characteristics very similar to tantalum, but with a tendency to higher ΔH_a -values instead of the opposite, shifting e.g. Sn and Sb out of the window useful for bunching.

Molybdenum pockets partly reproduce the predictions by showing an increase in ΔH_a for Cu, Ti, Pb, and Bi and a decrease for Ge and Au. Since the latter two elements have long contact times at tantalum surfaces (section 4.1), an additional test was made, lining the internal surfaces of the ion source with molybdenum foils. This way, Au was transferred into the range useful for bunching, yielding $\eta_A > 90\%$, $\Delta t_M = 10.2$ s and $\tau_B = 2.1$ s in case of weak cooling plus heating. In analogy to the footnote in section 4.1 the mean contact time t_a can be determined to be approximately 1 ms for Au, Ga and Cu at a molybdenum surface of 1770 K. To mention also the expectations which were not reproduced: Cd and Zn on the one hand and Ge, As and Pd on the other are not transferred into the

bunching window, the ΔH_a -values of Sn and Sb are not lowered and the reversal of the In-Sn-sequence is not observed.

Zirconium pockets showed the expected increase of ΔH_a and in most cases even stronger than predicted, leaving only Ti as an effectively bunching element - an easy means of selective Ti-separation. The strong increase of ΔH_a predicted for Hg, Cd and Zn turned out to be only marginal: instead of all three elements only Zn could be significantly accumulated ($\eta_A = 40\%$) and this only with reduced power settings (i.e. lower temperature) of the ion source.

Graphite pockets (totally purified, 1.9 g/cm^3 , fine-grained grade 1346 PT, Le Carbone Lorraïne) performed similar to molybdenum including the possibility of bunching Au, using the molybdenum-coated ion source. Most remarkable is the large spread in the ΔH_a -values of Ga and Cu (the latter being placed above the bunching window), yielding the best method for a separation of Ga from Cu.

The measured data of η_A and Δt_M are compiled in fig. 5. Comparison with the exponential trends of the respective data in fig. 2 permits to fit the ΔH_a -values for the additionally studied surfaces into the scale established for a tantalum surface. This comparison and the implicate revision of ΔH_a -values is presented on the right side of fig. 4. Fig. 6 illustrates the different surface properties at the example of the bunches of Ag, In, Ti, Pb and Bi in linear scale, which is better suited to demonstrate the enormous dynamics of the bunching technique.

5. Bunching, Chemical Separation and Control of Mass Separation - a Compilation

For the elements considered within this work, the results of chapter 4 may be summarized thus: **Most of the elements may be effectively bunched and can be separated efficiently and chemically selective.**

As to bunching, table 1 gives a representative survey. For all configurations Cd and Hg are found in category A or B and Ge, As and Pd in class D or E and are thus excluded from bunching. The list is intended for practical use. Therefore the bunch profiles are not characterized by

the more academical numbers Δt_M and τ_B , but by giving t_{FWHM} of the bunch and the delay Δt of this time window from the moment of cooling removal. In addition to the accumulation efficiency η_A , a bunching efficiency η_B is defined, which considers the losses caused by both (incomplete) accumulation and the setting of a time window on FWHM. Since such a window contains between 50 and 75 % (the ratio η_B/η_A) of the bunch integral, two general statements can be made for effectively bunching ($\eta_A \geq 80$ %) elements:

- Bunching costs about a factor of 2 in total intensity.
- The ratio of peak intensity of the bunch to DC-intensity is approximately given by $t_{acc}/2t_{FWHM}$.

To illustrate the latter statement: taking e.g. Ag, In, Tl or Pb with a $t_{FWHM} \approx 1$ s and choosing an accumulation time t_{acc} of half an hour, the bunch has a peak intensity which is about a factor of 1000 higher than the intensity in case of continuous (DC) operation.

As to chemical separation of adjacent elements, there is usually more than one possibility. Table 2 presents essentially the cases, which permit efficient and, if possible, selective separation. Not included are the elements Ge, As, and Pd, which belong for all configurations to category D or E and are thus out of the handling range of the ion source. The mechanism enabling separation depends on the adsorption characteristics of the element of interest:

- Elements which do not or only weakly accumulate (class A, B) can be separated from strongly adsorbing elements (C, D, E) by measuring the former continuously and retaining the latter in the **cold** pocket. The suppression of the unwanted elements is given by their ε_A -value; i.e. it is for the conditions within this work limited to a factor of 20 - 50 due to the 2-5% chance of potentially adsorbing particles not to "find" the cold area.
- Elements which bunch efficiently (class C) can be separated from those which do not or only weakly accumulate (class A, B) as well as from those which remain adsorbed even in the **hot** or **heated** pocket (D, E) by collecting only within a time window appropriate for the bunch. The

degree of chemical selectivity can be very high in this case (especially when the flow or the production of radioactive species is stopped when the separation window is open), since it scales with the ratio t_{acc}/t_{FWHM} .

- Neighbouring elements, bunching both efficiently but with major differences in the delays of their FWHM can be separated from each other by setting appropriate separation windows. The degree of chemical selectivity depends strongly on the spacing of the bunches (see e.g. fig. 3 and 6) and can be considerably enhanced by sacrificing intensity.

Besides enabling chemical separation, the method of bunched beam release is also suited to control the quality of mass separation. It actually greatly facilitates the determination of the broad range collector distribution of a certain mass or of the mutual cross-contaminations; usually a time-consuming procedure, where a radio-nuclide is used to trace a mass line down to its wings (see e.g. ref. 5 and references therein). Instead of utilizing the specific radiation as a signature of a certain isotope, the specific time profile of a bunch is used as a signature for a certain element. Then scanning of a mass line can be easily performed using a, preferably monoisotopic, element with good bunching properties. Among the elements considered here, In is the best candidate; the measured distributions are shown in fig. 7. Since a peak intensity of a bunch of up to 3 μA can be handled by the ion source without serious degradation of the ionization efficiency and since peak intensities down to 0.03 pA can be clearly identified (when impurities of Ag and Sn are small), a sensitivity of 10^{-8} can be reached; a value hardly accessible by the radioactive tracer technique, especially in view of a measuring time of less than one minute per data point.

6. On-Line Applications

The promising features of bunched beam release have been used for two types of on-line work: the production of very intense bunches of neutron-deficient isotopes of In and Sn for collinear laser spectroscopy and the chemical separation of Sn from lower-Z elements for γ -spectroscopy of very neutron-deficient Sn isotopes. When passing from the off-line to the on-line regime some additional, advantageous and disadvantageous aspects should be considered:

Off-line tests use a constant flow of atoms into the ion source; on-line the production may be easily switched on and off with the accelerator beam. This is very convenient for suppressing unwanted activity during the measuring intervals. The unknown and delayed release of the contaminants, still implanted in the catcher, make it, however, difficult to estimate the degree of chemical selectivity from off-line results.

- The accelerator beam dissipates additional power in the ion source. This may increase the release speed and possibly lower the accumulation efficiency.
- When the target/catcher foils are placed in the pocket, with different surface properties and temperatures than the pocket walls, the bunches may split up into several components.

6.1 Collinear Laser Spectroscopy of Neutron-Deficient Isotopes of In and Sn

During the last years, the hyperfine structure of neutron-deficient isotopes of In and Sn was studied with collinear laser spectroscopy by a Mainz University/GSI collaboration¹¹⁻¹⁴. One principal restriction of the extension to isotopes far-off stability was beam-independent background in the optical detection system (photo-multiplier dark current, straylight), but also background due to radioactivity built-up close to the multiplier. With a detection probability ranging from 10^{-4} to 10^{-5} photons/atom, this limited the investigations to isotopes, where mass-separated beams of approximately 10^6 ions/s were available. As outlined in chapter 1, bunched beam release opened the perspective of both reduced measuring times and extension to isotopes with lower production rates.

It was shown in ref. 6, that high yields of neutron-deficient isotopes of In and Sn are produced using an ^{16}O -beam and a target/catcher stack of thin, enriched molybdenum foils with 30 mg/cm^2 total thickness placed inside the FEBIAD-B2 ion source. The desired rates require an ^{16}O -beam of 500 particle nA. At this intensity, excessive losses of target material due to evaporation can only be avoided by using the colder position of the ion source (see fig. 1). To avoid changes on the bunching device, which costs 3 - 4 hours of beam time, a configuration was required, bunching In

and Sn simultaneously. Under these boundary conditions the off-line experience left the choice between two possibilities:

i) Using a tantalum pocket and strong cooling.

This configuration enabled improved measurements of $^{108\text{m},9}_{\text{In}}$ and of $^{108}_{\text{Sn}}$ via the reaction $^{16}\text{O}(^{97}\text{Mo},\text{xp},\gamma\text{n})$. Since the Sn-bunches are long and inefficient under these circumstances (compare fig. 3D), the target stack was placed in the pocket (see upper part of fig. 1). The beam power of 70 watts increased the release speed of the Sn-bunches (FWHM of 13 s delayed by 5 s) as determined by α -spectroscopy. One bunch with an accumulation time of 10 min sufficed to measure the hyperfine spectrum of $^{108}_{\text{Sn}}$ with good statistics¹⁵, even using the 563 nm transition for optical excitation. This transition has a lower photon/atom ratio, but the hyperfine splitting is larger and thus easier to interpret than the 453 nm line used earlier¹⁴. The beam-heating also speeds up the In-bunches ($t_{\text{FWHM}} = 0.5 \text{ s}$, $\Delta t = 0.25 \text{ s}$, see ref. 12) which showed up with a satellite peak - most probably due to the presence of two materials in the pocket. As shown in fig. 8, the loss of approximately a factor of 2 in intensity (bunching efficiency of 50 %) is by far outweighed by an improvement of the peak-to-noise ratio of ≈ 20 . That the relative intensities of the fluorescence lines are not correctly reproduced by scanning only once over a sharp bunch is in most cases a mere lack of beauty rather than disturbing: The line intensities alone are not sufficient for a spin-analysis, anyway, due to optical pumping effects.

ii) Using a molybdenum pocket and weak cooling plus heating.

This arrangement was used for spectroscopy of $^{104}_{\text{In}}$ by the reaction $^{16}\text{O}(^{92}\text{Mo},\text{p},3\text{n})^{104}_{\text{In}}$, placing the stack of ^{92}Mo -foils in a target holder on the entrance side (fig. 1 bottom). In this configuration the bunches of stable tracers and radioactivity are identical and should correspond to the values in table 1. Unfortunately, with the ^{16}O -beam on the ion source, the accumulation efficiency for In (not for Sn) dropped from 90 % to low values. After switching the accelerator beam off, the normal bunching behaviour took a long time to only partly recover. Thus, this strange effect cannot be attributed solely to beam heating and leaves only the explanation that the "oxygen-input" of $3 \times 10^{12}/\text{s}$ poisons the surface of the molybdenum pocket. Nevertheless it was possible to measure the hyperfine structure of $^{104}_{\text{In}}$ even without bunched

operation¹⁵⁾ - a procedure taking many hours instead of a measuring time of one hour expected with bunching.

6.2 Tests for α -Spectroscopy of Very Neutron-Deficient Sn Isotopes

Extending α -spectroscopy to very neutron-deficient Sn isotopes was hitherto always hindered by the fact that heavy-ion reactions like $^{16}\text{O} + ^{92}\text{Mo}$ or $^{58}\text{Ni} + ^{50}\text{Cr}$ have orders of magnitude higher yields for the lower-Z isotopes. No conventional means reduced the contamination essentially, although it has been found that niobium catchers show a slight tin-over-indium-preference. Even tests with various and heated collector foils, to reduce the amount of the more volatile contaminants, remained unsuccessful. No regime was found that retained Sn in the collector and evaporated the contaminants sufficiently (unpublished results, GSI 1981 and 1983).

Calculations with the evaporation code HIVAP¹⁶⁾ of the production rates for the isotopes from Ag to Sn in the mass range 103-105 are compiled in table 3, together with experimental yields. As to the Sn isotopes, HIVAP predicts for $A \geq 104$ higher rates via the reaction ^{16}O on enriched molybdenum targets - a reaction, however, showing yields many orders of magnitude higher for the contaminants. For $A < 104$ the substantially "cleaner" reaction ^{58}Ni on ^{50}Cr is clearly superior.

As shown in chapter 5 the method of bunched beam release should allow the chemical separation of Sn, applying a tantalum pocket and weak cooling plus heating (see fig. 3). As to the excessive yields for the contaminants, it is obvious that production (i.e. the accelerator beam) must be switched off, when the "separation window" for the Sn bunch is open. It was also clear that the beam should be stopped some time prior to the separation interval, to reduce the release of contaminants, still diffusing out of the catcher. This intermediate interval cannot be made too long due to the 21 s-half-life of ^{104}Sn ; the ratio of the time with "no beam" to the length of the storage/release cycle is a second reduction factor for the effectivity of the bunching method (besides τ_B). For clarity the run-off control diagram for the storage/release cycle in fig. 9 may be consulted. How to profit best from this intermediate interval, for which 2.5 s were selected, was, however, not obvious: to let the target/catcher stack cool down and keep the residual contamination confined or to leave it as hot as possible,

to evaporate as much of the unwanted activities as possible, before the separation window opens.

The "cold-catcher" technique was tested for the reaction with an ^{16}O -beam of 500 particle nA on a 30 mg/cm^2 ^{92}Mo target/catcher stack, mounted in a target holder on the colder side of the ion source (fig. 1 bottom), without heat-shielding applied to the window area. Under these conditions the Sn-bunches measured off-line (table 1) have a FWHM of 6.0 s delayed by 3.4 s. To improve these values the outlet hole of the ion source was increased from 1 to 1.5 mm diameter. The τ_B -value decreased indeed from 4.0 s to 1.9 s and consequently the FWHM to 4 s. The value of η_B for Sn, however, dropped from 50 % to 30 % due to the 70 watts of the accelerator beam dissipated additionally in the ion source.

The "hot catcher" technique was tested using a ^{58}Ni -beam of 40 particle nA on an external 3.0 mg/cm^2 ^{50}Cr -target. The internal catcher of thin, natural molybdenum foils was placed in the hot position of the ion source, where the foil-holder is well-heated by radiation and electron bombardment from the cathode and was heat-shielded by two carbon foils of $22 \text{ }\mu\text{g/cm}^2$ each.

The de-contamination factors at mass 104 for In, Cd and Ag were 150, 50 and 30 for the cold-catcher technique and 80, 40 with hot catchers, ^{104}Ag not being produced in the latter reaction. That they are not decisively different proves that both approaches are principally suited and that residual activity is at present the only hindrance towards even better selectivity. The different de-contamination factors cannot be explained by the different adsorption properties of gaseous atoms. Thus they indicate strongly, that the speed of diffusion out of the molybdenum catcher decreases in the sequence In-Cd-Ag.

The chemical separation for Sn thus achieved is illustrated by the α -spectra in fig. 10 for mass 105. With bunched operation the α -lines of $^{104}, ^{105}\text{Sn}$ showed up within a few minutes in singles-spectra and could be associated by the known half-lives^{17,18)}. The lines of 309.5, 423.5, 992.5, 1281.9, 1416.4 and 1457.5 keV for ^{105}Sn agree with the data recently published by the LISOL-group¹⁹⁾.

7. Conclusion and Outlook

It was shown that the method of bunched beam release can improve the experimental situation in various fields. For collinear laser spectroscopy increased signal-to-noise ratios were reached, which permit to choose transitions with a lower photon/atom ratio, to study more-exotic nuclei (possibly also ^{103}In and ^{107}Sn) and which should also facilitate application to other elements (e.g. Ag, Tl, Pb). For most of the elements studied within this work, chemically selective mass separation is now possible. The degree of chemical selectivity so far reached for e.g. Sn, gives a promising perspective for a $\bar{x}$ -spectroscopic re-investigation of $^{104,105}\text{Sn}$ under substantially better background conditions.

The extension to a higher temperature ion source such as e.g. FEBIAD-E⁹ with a typical temperature of 2100 - 2200 K, should permit "chronological thermochromatographic separation" for elements so far excluded (e.g. Ge, As and Pd), and would provide a means to speed up the relatively slow bunches of Sn and Sb. A shorter length of Sn-bunches is highly desirable for $\bar{x}$ -spectroscopy of ^{103}Sn (the 7s half-life of which is known from β -delayed protons) and is an evident prerequisite for a search for the decay of ^{102}Sn . The range of application could most likely be extended to further elements, e.g. to the transition elements below copper. Half-life measurements for neutron-rich isotopes in this area are of special interest, since recent experimental results appear to deviate systematically from predictions²⁰. These isotopes can be produced via multinucleon transfer reactions with yields sufficient for spectroscopy, as long as contamination by the higher-Z-isobars stays within reasonable limits. In spite of the detection of $^{67,69}\text{Ni}$, the semimagic-magic nucleus ^{68}Ni could so far not be identified²¹ in the high background of ^{68}Cu and ^{68}Ga . Even "wet chemistry" on separated mass-68-sources, reducing contamination by a factor of more than 25, did not allow identification of ^{68}Ni - an indication that the half-life may be too short for off-line chemical separation. Assuming the ΔH_a -value of 5.8 eV from the systematics¹⁾ for the adsorption of Ni on a tantalum surface, separation of Ni from Cu and Ga should be possible, when a temperature of ≥ 2100 K for ion source and **heated** pocket is guaranteed.

Even with the present set-up, there are further elements in the periodic table accessible. For example, the alkali metals Na ($\Delta H_a = 1.3$ eV), K (1.7 eV) and the alkaline-earths Be (3.9 eV), Ca (3.4 eV) and Ba (4.0 eV), present as parasitic beams and only randomly studied so far,

were accumulated in a weakly-cooled tantalum pocket to less or higher degree - the alkalies certainly much stronger than the low ΔH_a -values suggest (probably due to the traces of oxygen in the ion source which are known to systematically increase ΔH_a for alkalies¹⁰). Whether this might allow the separation of alkaline-earths from alkalies, an old goal of ISOL, is still to be shown.

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Table 2

Compilation of the elements, considered within this work, for which chemical separation can be performed. The capital letters in parentheses notify the category to which the elements belong in the chosen configuration as defined in section 4.1. The configurations are notified as in table 1.

Element	Configuration ^a	Separated from
Cu (C)	w, Ta, h	Zn (A), Ga (B), Ge(D), As (E)
Zn (A, B)	s, mult, c	Cu, Ga, Ge, As (all C, D, E).
Ga (C)	w, s, C, c	Cu (D), Zn (A, B), Ge (E), As (E)
Ag (C) ^b	s, Ta, h	Pd (E), Cd (A), In (C), Sn (C), Sb (D)
Cd (A, B)	s, mult, c	Pd, Ag, In, Sn, Sb (all C, D, E)
In (C)	w, Mo, c	Pd (E), Cd (A), Ag (C), Sn (C), Sb (C)
Sn, Sb (C) ^c	w, Ta, h	Pd (E), Cd (A), Ag (B), In (B)
Au (C) ^d	w, C, Mo, c	Hg (A), Tl (B), Pb (C), Bi (C)
Hg (A)	s, mult, c	Au, Tl, Pb, Bi (all C or D)
Tl (C)	s, Zr, c	Hg (A), Au, Pb, Bi (all D or E)
Tl (C) ^e	s, C, Nb, Mo, c	Hg (A), Au (D), Pb (C), Bi (C)
Pb (C) ^e	s, C, Nb, Mo, c	Hg (A), Au (D), Tl (C), Bi (C)
Bi (C)	w, Nb, c	Hg (A), Au (D), Tl (B), Pb (B)
Bi (C) ^e	s, Nb, Mo, Ta, c	Hg (A), Au (D), Tl (C), Pb (C)

^a mult: C, Zr, Nb, Mo, Ta-surfaces are all possible

^b See fig. 3

^c No configuration permits a more than marginal separation of Sn and Sb (see e.g. fig. 3)

^d Using an ion source with molybdenum-coated or graphite electrodes

^e See fig. 6

Table 1

Representative compilation of efficiently bunching elements.

The configurations are notified by e.g. s, Ta, h, which stands for strongly cooled Ta-pocket in the hot position of the ion source or by w, Mo, c for a weakly cooled Mo-pocket in the colder position (see fig. 1 for details). η_A is the accumulation efficiency, t_{FWHM} gives the characteristic width of the bunch and Δt the delay of this time window from the moment of cooling removal. η_B is the efficiency of the bunching technique, considering only the bunch integral over the FWHM.

Element	Config.	η_A	Δt [s]	t_{FWHM} [s]	η_B
Ag	s, C, c	0.95	9.0	6.7	0.67
In		0.95	8.3	5.9	0.66
Tl		0.92	3.6	1.9	0.63
Pb		0.95	7.1	3.7	0.68
Bi		0.95	11.4	7.8	0.68
Ga	w, C, c	0.82	2.7	4.3	0.53
Au ^a		0.95	6.9	4.8	0.50
Zn ^b	s, Zr, c	0.41	0.8	2.9	0.30
Tl		0.85	1.3	6.3	0.72
Cu	w, Nb, c	0.85	2.5	4.0	0.48
Bi		0.75	1.4	1.7	0.51
Cu	w, Mo, c	0.95	5.6	4.8	0.74
Ga		0.95	7.1	4.5	0.67
Ag		0.65	1.5	2.0	0.48
In		0.88	2.6	2.8	0.63
Sn		0.94	8.6	9.5	0.55
Bi		0.90	3.6	2.9	0.63
Au ^a		0.95	7.5	5.0	0.44
Ag	s, Mo, c	0.90	4.6	2.4	0.63
In		0.93	5.7	4.1	0.68
Tl		0.88	3.0	2.0	0.65
Pb		0.90	6.2	3.8	0.64
Bi		0.93	11.6	4.9	0.64
Ga	s, Ta, c	0.95	6.6	5.5	0.61
Cu		0.95	10.7	7.2	0.59
Ag		0.95	4.5	1.5	0.54
In		0.95	2.9	2.2	0.65
Tl		0.90	2.6	1.3	0.54
Pb		0.95	3.0	1.5	0.59
Bi		0.95	5.0	3.6	0.66
Ga	s, Ta, h	0.90	3.2	5.2	0.68
Cu		0.94	8.2	4.7	0.58
Ag		0.92	3.4	1.0	0.54
In		0.92	2.4	1.3	0.57
Tl		0.88	2.1	0.9	0.55
Pb		0.89	2.6	1.0	0.57
Bi		0.91	3.9	1.7	0.55
Cu	w, Ta, c	0.90	2.8	3.3	0.52
Sn		0.93	3.4	6.0	0.50
Sb		0.94	5.6	12.0	0.59
Cu	w, Ta, h	0.90	1.6	2.1	0.53
Sn		0.92	2.5	5.8	0.49
Sb		0.95	3.6	9.8	0.49

^a Using an ion source with molybdenum-coated electrodes

^b Operating the ion source with reduced power settings

Table 3

Calculations with the evaporation code HIVAP¹⁶⁾ and experimental yields for the production of mass-103-105-isotopes of Sn, In, Cd and Ag. The numbers in the upper part of the table assume 500 particle nA of 8.2 MeV/u ¹⁶O on an internal target/catcher stack of 30 mg/cm² ⁹²Mo. The bottom numbers assume 50 particle nA of 4.7 MeV/u ⁵⁸Ni on a 3.0 mg/cm² external target of ⁵⁰Cr with an internal catcher of natural molybdenum. The normalization currents of 500 and 50 particle nA are the maximum intensities, the respective targets can withstand for longer times without severe deterioration. The low values of the separation efficiency η_s for the Sn isotopes indicate that HIVAP overestimates the production rates far-off stability, although the very low η_s -value of 0.2 % for ¹⁰⁴Sn in the reaction ⁵⁸Ni on ⁵⁰Cr is partly due to a too low catcher temperature (heating of the catcher by the accelerator beam only 10 watts; an η_s -value of 2 % was measured in the same reaction with a FEBIAD-E ion source⁹⁾). The η_s -value of only 1 % for ¹⁰⁴Sn in the reaction ¹⁶O on ⁹²Mo cannot be attributed to a low temperature of the catcher (beam heating almost 70 watts), since η_s -values of 20 - 30 % were measured for ¹⁰⁶⁻¹¹¹Sn under practically identical conditions⁶⁾.

A=103			A=104			A=105		
HIVAP	HIVAP	Exp.	HIVAP	Exp.	η_s [%]	HIVAP	Exp.	η_s [%]
Sn 2x10 ³	7x10 ⁴	7.1x10 ²	1	1x10 ⁶	2.7x10 ³	20.3		
In 9x10 ⁵	8x10 ⁶	5.4x10 ⁵	7	3x10 ⁷	2.4x10 ⁶	7		
Cd 4x10 ⁷	1x10 ⁸	8.8x10 ⁶	8	1x10 ⁸	4.6x10 ⁶	4		
Ag 8x10 ⁷	6x10 ⁷	4.9x10 ⁶	8	2x10 ⁷	-	-		
Sn 9x10 ³	6x10 ⁴	1.4x10 ²	0.2	3x10 ⁵	2.3x10 ²	20.1		
In 1x10 ⁵	5x10 ⁵	2.5x10 ⁴	5	4x10 ⁵	6.3x10 ⁴	17		
Cd 6x10 ⁵	9x10 ⁵	1.5x10 ⁵	17	-	-	-		
Ag 2x10 ⁵	-	-	-	-	-	-		

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Fig. 1 FEBIAD-B2-ion source for on-line operation with the accumulation system, mounted in the colder position (A) of the ion source. By inverting the anode cylinder the ports come to the hot position (C). **Bottom:** Configuration with target/catchers in a holder on the entry side, with weak cooler (decoupled) and extra heater. **Top:** Configuration with heat-shielded window and the target/catcher stack in the pocket and with coupled strong cooler. (1) heavy-ion beam, (2) mass separator beam, (3) target/catcher stack, (4) 2 mg/cm^2 dish-shaped tungsten window, (5) window heat-shields of $2 \times 22 \text{ } \mu\text{g/cm}^2$ carbon, (6) anode, (7) cathode, (8) outlet plate, (9) accumulation pocket, (10) extra heater, (11) weak cooler, (12) strong cooler, (13) oven line, (14) gas line.

Fig. 2 Storage efficiency η_A , delay of the bunch maximum Δt_M and fall-off half-time τ_B of the bunch of various elements versus semi-empirical values of the enthalpy of adsorption ΔH_A taken from ref. 1. The typical error of measurement is of the size of the data points, whereas the bars give the typical ion-source-to-ion-source scatter for incompletely accumulated elements (only for the η_A -values of which several measurements were performed). For the normalized exponential fits to the η_A -data and the shifts indicated for Ga and Tl by horizontal arrows, see text. Points below $\eta_A = 10^{-2}$ % with downwards pointing arrow mean that the element has been looked for but η_A is below the detection limit. The lines in the Δt_M , τ_B -plots are to guide the eye.

Fig. 3 Normalized bunch profiles for stable isotopes of the elements Ag - Sb measured for all configurations after accumulation of 60 s. The unit intensity corresponds to the constant current without cooling. The cooling block is removed at $t = 0$. The profiles for weak cooling plus heating, C and D for strong cooling, the pocket being mounted in the hot and colder position of the ion source, respectively.

Fig. 4 Enthalpies of adsorption ΔH_a for various elements on different adsorbing surfaces. **Left side:** Semi-empirical values from the systematics¹⁾. **Right side:** Experimental values (for significance see end of section 4.1). Where no measurement was performed, the systematical location was taken instead. Graphite is not represented in the systematics. **Solid points:** agree with systematics or are shifted by less than 0.33 eV. Parentheses indicate that elements belong to category D and that agreement can be stated, but not in terms of η_A , Δt_M , and τ_B . **Open points:** Disagree with the systematics by more than 0.33 eV. Parentheses mean that elements are of category A or E and measurements do only confirm the position below or above the detection window but not the absolute value.

Fig. 5 Storage efficiency η_A and delay of the bunch maximum Δt_M versus semi-empirical ΔH_a -values¹⁾ for various elements at surfaces of graphite, zirconium, niobium and molybdenum. The lines are fits to the respective data for a tantalum surface taken from fig. 2. The data points for graphite (C), for which no systematical ΔH_a -values were available, are placed to give best agreement with the fitting curves for both η_A and Δt_M .

Fig. 7 Histograms of the broad range collector distribution of In, determined by bunched beam release for two beam lines of the GSI mass separator on-line 7). The full points give the distribution for the central beam line, the open points for the left beam line, where a cylindrical 45°-deflector performs additionally electrostatic "cleaning". The slit widths at the entry to the beam lines were 5 mm, which let approximately 95 % of the beam intensity pass to the Faraday cups at the end of the beam lines. The scanning steps were 0.33 mass units, corresponding closely to the 5 mm slit width. The histograms are thus integral mass scans, the data points giving directly the value of the respective cross-contamination; e.g. the contamination by mass 115 is $1-2 \times 10^{-4}$ at the masses 114 and 116 and is decreasing to 10^{-6} for more remote mass numbers. The measurements were performed with an In-beam of 20 nA (DC), giving a bunch peak intensity at mass 115 of 300 nA, using an accumulation time of 30 s. The configuration with a strongly cooled Ta-pocket in the hot position of the ion source was chosen. Due to a contamination of the oven line with Ag and Sn, yielding parasitic ion beams in excess of 100 pA, the detection limit for the In-bunches was 0.3 pA in the wings of the mass line distribution, restricting thus the sensitivity to 10^{-6} .

Fig. 6 Normalized bunch profiles for stable isotopes of Ag, In, Tl, Pb and Bi, measured for different pocket materials, accumulating for 60 s and applying strong cooling to the respective pocket in the colder position of the ion source. The unit intensity corresponds to the constant current without cooling. The cooling block is removed at $t = 0$. The accumulation efficiency η_A is higher than 88 % for all of the bunches.

Fig. 8 Fluorescence spectra for $^{108g,m1}\text{In}$ measured by collinear laser spectroscopy. Both spectra were taken under the same conditions, except measuring the upper spectrum for 70 min in DC-mode ¹¹⁾ and the spectrum at bottom in bunched operation ¹²⁾. For the latter 6 bunches with an accumulation period of 10 min and a measuring time of 0.5 s were added. The lines of the isomeric state are marked by arrows.

Fig. 9 Run-off control diagram of the storage/release cycle for the reaction $^{16}\text{O}(^{92}\text{Mo},\text{xn})^{104,105}\text{Sn}$. Both isotopes were measured simultaneously by collecting the bunches on two tape counting positions. For illustration transportation time to the respective counting positions. For illustration the bunch profiles of the stable tracers of Ag, Cd, In and Sn are given, which fix the separation window. The respective radioactive bunches differ only in so far as, instead of "constant feeding", production is switched off for 7.5 s.

Fig. 10 Part of the γ -singles spectra of mass 105 measured in the reaction $^{16}\text{O}(^{92}\text{Mo}, \text{xp}, \text{yn})$ by the conventional method (top) and with bunched beam release (bottom). For the upper spectrum 27 cycles of 6 s collection and 48 s counting time were summed. The full collection time of 48 s could not be used due to dead time problems. Although the short collection time in fact favours ^{105}Sn (31 s) against the longer lived isobars, the ^{105}Sn γ -lines are hardly visible. For the lower spectrum 25 bunches, according to the separation scheme shown in fig. 9, were summed, showing an enhancement of Sn by factors of approximately 10, 15 and 45 over Cd, Ag and In, sacrificing only 70 % of the total Sn intensity.