A new high transmission infinite range mass selector for cluster and nanoparticle beams

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A new mass selection technique has been developed, which allows one to size-select charged particles from atoms to nanoparticles of almost unlimited size. It provides a mass resolution of $m/\Delta m = 20-50$ and a transmission of about 50% for the selected size, both independent of mass. The technique is based on the time-of-flight principle, but differs fundamentally from time-of-flight mass selection normally used. The basic idea is to use time-limited high voltage pulses to displace laterally a preaccelerated ion beam, without changing its direction or shape. As the movement of the ions perpendicular to their original beam direction is independent of their forward velocity, mass resolution and calibration does not depend on the ion beam energy. A mass selector of this type has been implemented successfully into a cluster deposition experiment and has proven to be reliable and simple to operate. © *1999 American Institute of Physics.* [S0034-6748(99)03712-0]

I. INTRODUCTION

The last years have seen a rapidly growing number of experiments on deposited clusters. This strong interest is due both to the wealth of fundamental physical questions which can be addressed by such experiments and to the possible technical applications of very small surface structures. Up to now, many of the experiments on deposited clusters have been done without applying mass selection. As these experiments are mostly intended to study the size dependence of cluster properties, they lack specificity due to the relatively broad cluster size distribution produced by standard cluster sources. It is thus desirable to mass select the clusters before deposition. Such a mass selection technique has to fulfill several conditions:

(1) High transmission: As the mass selection naturally reduces the overall cluster intensity, as few as possible of the desired clusters should be lost.

(2) Very broad mass range: In deposition experiments, the particles used go from small clusters containing a few atoms to rather big particles with diameters of several tens of nanometers.

(3) Tolerance of initial energy distribution: As many cluster sources produce particles with considerable kinetic energies, the performance of the mass selector (resolution, transmission, calibration) should ideally be independent of this energy.

(4) Reasonable mass resolution, independent of size: While the properties of smaller clusters depend on the exact number of atoms they are made of, larger particles (containing a few hundred atoms or more) generally exhibit a slower change of properties with size. For experiments on larger particles, mass distributions with relative widths of 1% or even 10% will often be sufficient. Thus a mass selector with a resolution between $R=10$ and $R=100$ is required. Too high a resolution of the mass selector could even be a disadvantage, as it inevitably leads to a reduction of the cluster beam intensity.

The existing mass selection techniques cannot really fulfill these conditions:

(1) Magnetic sector mass selection is good for smaller sizes, but for bigger sizes, increasingly big and unwieldy magnets are required. For example, even a very big sector magnet with a field strength of 1 T and a radius of 0.5 m at an ion energy of 500 eV still has an upper mass limit of 2.4×10^4 amu (equivalent to Ag_{222}). Furthermore, the mass resolution of such a mass selector decreases with mass, and its resolution and especially calibration depends on the initial cluster kinetic energy distribution. Since, for example, clusters produced by gas aggregation sources can have initial energies of several hundred volts, which depend sensitively on the source conditions, the mass calibration can be quite tedious.

(2) Quadrupole mass selectors could in principle meet all the conditions, if frequency tunable power supplies and impedance matching boxes were available for the required frequency range of 10^3-10^7 Hz, which up to now they are not. Tuning the voltage at a fixed frequency to scan through the masses, however, leads to a very limited mass range. Furthermore, quadrupole mass selectors require low energy ion beams, which can lead to space charge problems, and there exists the possibility that charged particles very much larger than the selected ones just break through, i.e., are not filtered out.

(3) Time-of-flight mass selectors have practically infinite mass ranges. In the usual setups, however, their duty cycle is very low, which leads to a transmission of only $10^{-5}-10^{-4}$ at best. Additionally, resolution and calibration again strongly depend on the initial energy distribution of the clus-

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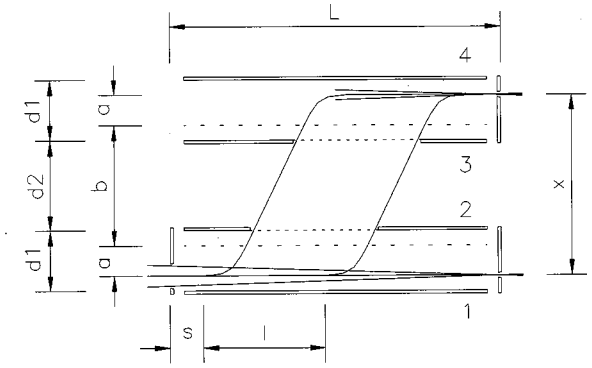

FIG. 1. Schematic of the mass selector. Short high voltage pulses applied to plates 1 and 4 are used to displace a preaccelerated convergent ion beam laterally. l: length of the ion beam package displacement, x: total displacement, a: beam offset traversed during the high voltage pulse, b: length of the field free region, d1/d2: plate separations, s: length of the portion of the ion beam which cannot be used due to field distortion, L: total length.

ters. Another disadvantage is that the standard time-of-flight technique cannot easily be used for cluster deposition experiments if low impact energy deposition is required, as it inevitably produces size-selected ion packages with broad kinetic energy distributions.

These limitations of the existing mass selection techniques motivated the development of the technique presented here. In the following, we will describe the principle of the method, give the necessary mathematical formulas, describe the actual setup, and finally present a performance example.

II. MASS SELECTION PRINCIPLE

The basic idea of the new mass selection technique is to displace an ion beam laterally by accelerating a portion of it perpendicular to its original direction by a pulsed electric field, letting it drift for some time, and then stopping the perpendicular movement by a pulsed electric field in the opposite direction. Since the magnitude of the ion beam displacement depends on the strength and timing of the high voltage pulses as well as on the ion masses, ions of different masses are dispersed into parallel beams with different displacements. With the help of an aperture one can thus select a small range out of the mass distribution of the beam.

A schematic of the setup is shown in Fig. 1. The cluster beam enters the mass selector from the left. It is focused by ion optics between the cluster source and the mass selector in such a way that its focal point is located at the small exit aperture in the end wall. The current measured behind this aperture can be used to adjust the cluster beam. By a short high voltage pulse applied to plate 1 beneath the cluster beam, the clusters are accelerated upwards. The length of the high voltage pulse is chosen such that no cluster can leave the acceleration region between plates 1 and 2 during the pulse. Therefore, all ions will gain the same momentum, and thus, all ions of the same mass will gain the same velocity perpendicular to the beam axis. The ions pass through a mesh covered opening in plate 2, and, after traversing a field free region between plates 2 and 3, enter the deceleration region between plates 3 and 4 by passing through another mesh covered opening in plate 3.

When all of the ions of the selected mass have entered this region, a high voltage pulse identical to the one used before is applied to plate 4. As a result of this pulse, the ions lose all of their velocity perpendicular to the original beam axis. They will therefore regain their original flight direction, and leave the selector through the exit aperture in the end wall.

In order to obtain a reasonable mass resolution, the ion beam should be well focused at the position of this aperture. Only then small changes in the beam displacement, due to small changes of the ion masses, can be resolved. Such a focus is obtained by the use of the time-limited high voltage pulses. As all ions of the same mass gain (and later on lose again) the same velocity, their *relative* velocities are not changed, and thus the convergence or divergence of the ion beam is conserved. An ion beam with a focal point at a certain distance from its source will still have this focal point after the lateral displacement. So if the original beam is well focused at the exit aperture position, a small aperture can be used, leading to a high mass resolution.

The time the ions need to travel from the acceleration region to the deceleration region depends on their mass. The timing of the two pulses thus defines the mass of the ions which are transmitted through the selector.

As soon as the acceleration pulse is over, the acceleration region is again filled by the cluster beam. This takes some time, which depends on the velocity of the selected ions. When the ions of the selected mass have proceeded far enough into the region, the next acceleration pulse is applied. Thereby, most of the cluster beam is used, and a very high overall transmission of 60% or higher can be reached.

It should be emphasized that the mass of the transmitted ions is independent of the ion beam energy. The only important parameter for the mass selection is the flight time needed for the lateral displacement, which depends on the strength and duration of the high voltage pulses and on the ion masses, but not on their forward energy.

The only restriction for the ion beam is that it should be reasonably well focused, which of course can become difficult if the ion energy distribution is extremely broad. Since, however, mean ion beam energies of up to a few thousand eV can be used, and a focus width of only 2–3 mm is required, this will not be a true restriction in most practical cases.

III. PARAMETER CALCULATION

Having discussed the principle of operation, we will now give the mathematical expressions describing the mass selection process.

The necessary variables are defined with the help of Fig. 1. The length l of the ion package accelerated sideways is determined by the length of the opening in plate 2. As the mass selection principal only works if the electric fields are homogeneous, the ions which experience the distorted field immediately behind the entrance aperture have to be blocked. This is achieved by the finite length of the opening. Thus, a length s of the cluster beam is not used (plus the part of the ion beam which enters the selector during the accel-

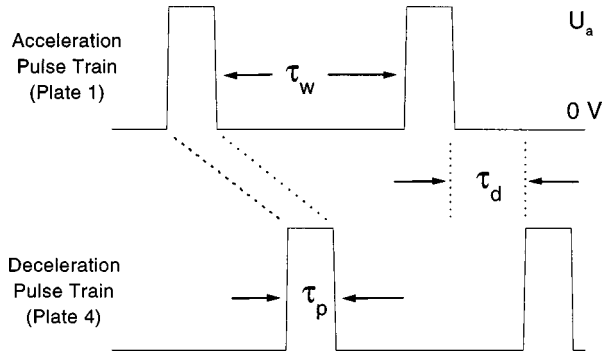

FIG. 2. Schematic of the timing of the high voltage pulses. Trains of identical pulses, displaced in time, are applied to plates 1 and 4 (see Fig. 1). τ_p: pulse length, τ_d: drift time between acceleration and deceleration pulse, τ_w: time between consecutive acceleration pulses.

eration pulse, which will also be deflected onto plate 2). The distance a, which the ions cover during the acceleration pulse (and during the deceleration pulse), should be chosen such that no ion of the selected mass can reach plate 2 during the pulse, taking the finite beam diameter into account. However, a should not be much smaller than the distance d_1 between plates 1 and 2, so that too high voltages do not have to be used for the pulses.

The timing of the pulses is shown schematically in Fig. 2. The high voltage start pulse is applied when the pulse region is filled with ions. Its duration is given by the time the selected ions need to cover the displacement a, that is,

$$\tau_p = \frac{2a}{\sqrt{2eU_p/m}} = \frac{2a}{v_p}, \tag{1}$$

where eU_p and v_p are the energy and velocity gained by the ions. If U_a is the voltage applied to plate 1, this energy is given by

$$eU_p = \frac{a}{d_1} eU_a. \tag{2}$$

The stop pulse is started when the ions have reached the second pulse region, i.e., when they have covered the drift length b. The delay time between the two pulses is thus

$$\tau_d = \frac{b}{\sqrt{2eU_p/m}} = \frac{b}{v_p}. \tag{3}$$

The waiting time between two consecutive start pulses is given by the time necessary to fill the pulse region:

$$\tau_w = \frac{s+l}{\sqrt{2eU_0/m}} = \frac{s+l}{v_0}. \tag{4}$$

Here, U_0 and v_0 are the forward energy and velocity of the ions. The time period both of the starting and stopping pulse trains is $\tau_w+\tau_p$. We can use this to determine the transmission of the mass selector. Every $\tau_w+\tau_p$, an ion package of length l is transmitted through the mass selector. During this time an ion package of total length $l_p=(\tau_w+\tau_p)v_0$ has entered the selector. The theoretical transmission efficiency for the selected mass therefore is

$$T = \frac{l}{l_p} = \frac{l}{[2a/v_p+(s+l)/v_0]v_0} = \frac{l}{s+l+2a(v_0/v_p)}. \tag{5}$$

With the right choice of parameters, this can be close to 100%. So despite the use of a pulsed technique, it is possible to obtain an almost continuous mass selected beam.

The mass resolution of the selector can be calculated from the mass dependence of the lateral displacement. For a given high voltage pulse length, all ions will obtain the same momentum. If m_0 and v_{m_0} are the mass and the perpendicular velocity of the ions which are transmitted by the selector, an ion of mass m will acquire the velocity:

$$v_m = v_{m_0}\frac{m_0}{m}. \tag{6}$$

Its lateral displacement is then given by

$$x_m = v_m\frac{\tau_p}{2} + v_m\tau_d + v_m\frac{\tau_p}{2} = \frac{m_0}{m}x, \tag{7}$$

where x is the total displacement of the transmitted ions (see Fig. 1). The derivative of the displacement with respect to the mass is

$$\left.\frac{dx_m}{dm}\right|_{m=m_0} = -\left.\frac{m_0}{m^2}x\right|_{m=m_0} = -\frac{x}{m_0}. \tag{8}$$

The exit aperture cuts a certain range of masses out of the laterally dispersed mass distribution. The width of this mass range is given by

$$\Delta m = \frac{dm}{dx_m}\Delta x = -\frac{m_0}{x}\Delta x, \tag{9}$$

where Δx is the full width half maximum (FWHM) of the convolution of the exit slit opening and the ion beam profile. The mass resolution is thus given by

$$R = \frac{m}{\Delta m} = \frac{x}{\Delta x}. \tag{10}$$

Hence, the maximum mass resolution of the mass selector is given by the ratio of the lateral displacement and the exit slit width.

IV. SETUP

The actual setup of the mass filter is shown in Fig. 3. The total length of the selector is 370 mm, while the height and the width are 150 and 90 mm, respectively. The overall lateral beam displacement is 120 mm.

In order to obtain a good performance, it is crucial that the pulsed fields are homogeneous and extremely well aligned. Thus, a rather strong construction was chosen. The plates 1–4 are mode of stainless steel, with thicknesses of 1 mm (plates 1 and 4) and 1.5 mm (plates 2 and 3). An opening is cut into plate 2 with area 12×150 mm^2, starting 80 mm behind the front end; the opening in plate 3 has dimensions 12×200 mm^2 and starts 130 mm behind the front end. Both openings are covered by mesh (Buckbee–Mears type MN-17), which is fixed to small frames made of 0.5 mm thick stainless steel. These frames are screwed onto plates 2

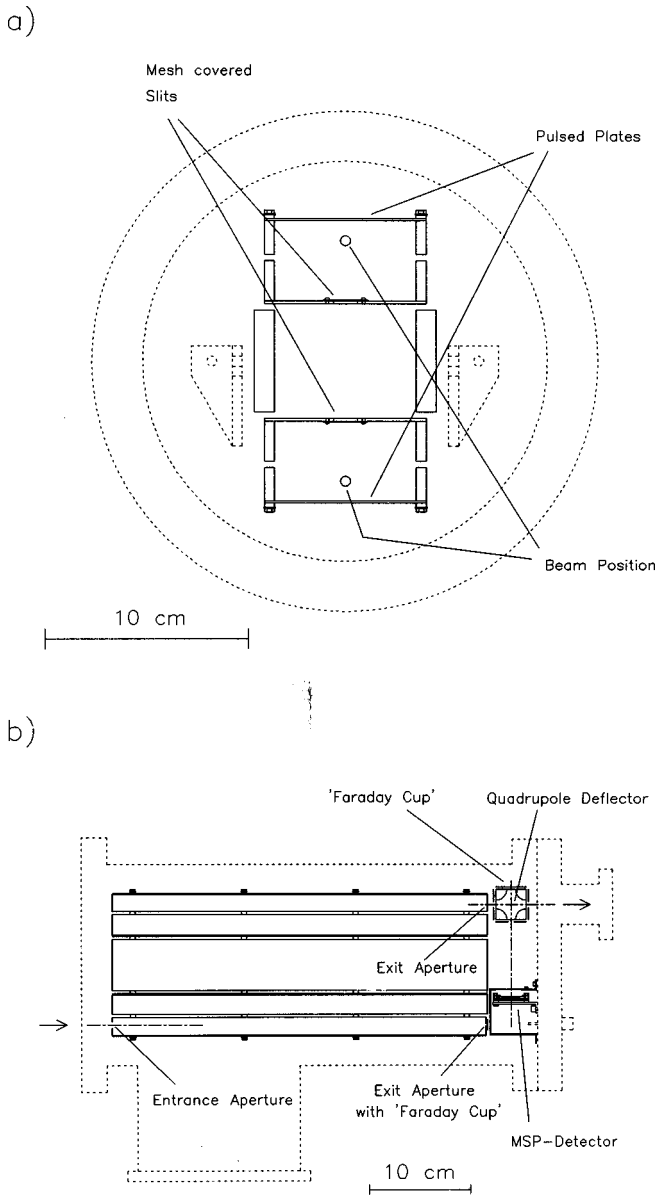

FIG. 3. Setup of the mass selector. (a) Front view; (b) side view.

and 3 by low profile M2 screws. The open sides of the acceleration, drift and deceleration regions are closed to aluminum bars, which serve both as field guards and as solid spacers which keep the plates in position. Plates and bars are held together by eight M2 threaded studdings, which are isolated by alumina tubings. The isolated studdings go right through the aluminum bars and the plates as well as through alumina rings which serve as spacers between the bars.

Care has been taken to find the right dimensions for the side plates of the acceleration and deceleration region. Ideally, one would like to have perfectly homogeneous fields inside the mass selector. Due to space limitations the side plates are positioned quite close to the selector center plane, and thus they can produce a considerable distortion of the fields. This distortion should be as small as possible, because the acceleration/deceleration scheme only works if during the stopping pulse the ions experience exactly the same electrical field as they have experienced (with opposite direction) during the accelerating pulse. With the help of SIMION[1] the

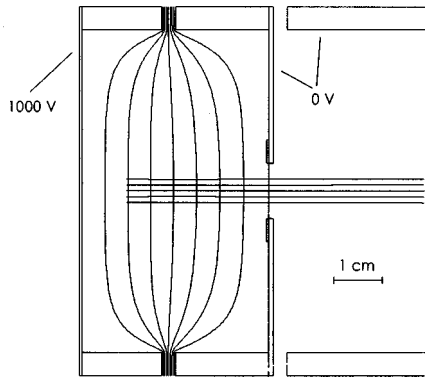

FIG. 4. Simulation (Ref. 1) of the field distribution in the acceleration region and the consequent ion trajectories. In the simulation, the field is switched off when the ions have on average gained 500 eV. The resulting trajectories deviate from perfectly horizontal trajectories by less than 0.11°.

geometry of the side plates which produces the smallest deviation of the ion trajectories from the ideal case was determined (see Fig. 4). The simulations show that with the chosen geometry the field distortion is small enough that it does not reduce the mass resolution (of up to 50) that we are aiming at. For a significantly higher mass resolution, however, a different geometry would become necessary.

The side plates of the central drift region also serve a second purpose. One potential problem of the mass selection principle presented here is a certain ambiguity in the mass selectivity. In order to obtain high transmissions, we use high voltage pulse trains with the shortest possible time delay between the pulses. All pulses are identical, so that in principle, any of the stop pulses can stop ions accelerated by one of the start pulses. Thus, if the timing is chosen such that ions of certain e/m are transmitted, there is a possibility that ions of higher masses are also transmitted. These are particles which are not decelerated by the stop pulse directly following the start pulse, but by the second, third, or even a later stop pulse. Due to their higher mass, these particles acquire a lower energy during the start pulse, and thus a smaller deflection from the beam axis, which means that they will have a lower transmission probability than the selected particles. Nevertheless, it is important to prevent such false transmissions from occurring. This is achieved by applying the stop pulse not only to plate 4, but also to one of the side plates of the field free region. As the false ions are still crossing this region when the stopping field is switched on, they will be deflected sideways and thus prevented from entering the deceleration region.

The front and back of the mass selector are closed by stainless steel plates attached to the aluminum side plates. Drilled into these plates are the apertures which define the ion beam dimensions. For the entrance aperture, a round hole of 6 mm diameter is used; for the two exit apertures either circular holes with 2 mm diameter or rectangular slits with width 3 mm are used. Behind the exit aperture at the end of the acceleration region, an electrically isolated strip of stainless steel serves as a Faraday cup to monitor the ion beam current passing the aperture, which is crucial for tuning up the selector. Behind the exit aperture of the deceleration region, a quadrupole deflector is mounted, which allows one

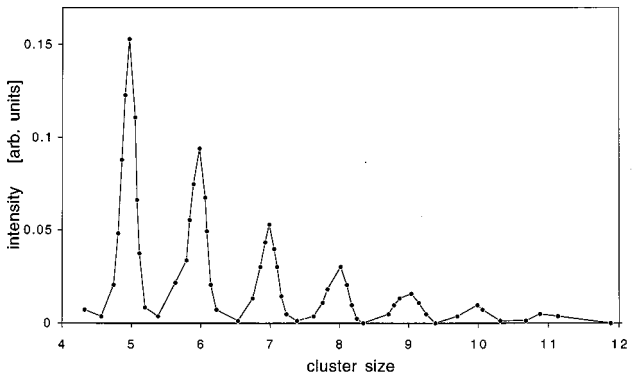

FIG. 5. Mass spectrum of ionized copper clusters Cu_n^+, $n=5-12$. The spectrum was obtained by measuring the transmitted ion current for different repetition frequencies of the high voltage pulses.

either to deflect the beam upwards onto a Faraday cup or downwards onto a microsphere plate (MSP) detector to measure the transmitted current. Without deflection the ions will go straight on and can be used for experiments.

The whole mass selector and all feedthroughs are mounted on one flange, which allows easy access to the setup. The incorporation of the mass selector into the cluster apparatus is straightforward: standard ion lenses and deflection units between the cluster source and the mass selector allow one to produce the required convergent ion beam and to align it properly. Ion optics behind the selector provide for the transport of the beam to its destination, making use of the fact that the exit aperture of the selector can be seen as a virtual ion source.

The high voltage pulses are produced by two high frequency high voltage switches (DEI, GRX-1.5k-E), which can work with repetition rates up to 600 kHz. This is more than sufficient, as in normal cases much lower frequencies are necessary. Even for a smaller cluster like Ag_{10}^+ (1080 amu) at an ion beam energy of 500 eV, one obtains pulse times [see Fig. 2 and Eqs. (1)–(4)] of $\tau_p=4.2\ \mu s$, $\tau_d=8.5\ \mu_s$, and $\tau_w=20.2\ \mu$s, which corresponds to a pulse repetition rate of about 40 kHz. Using the full 600 kHz, we could theoretically (even at 500 eV beam energy) select masses as small as 5 amu with maximum transmission.

V. PERFORMANCE

The actual mass resolution of the selector can be determined by recording a mass spectrum. This is done by measuring the transmitted current while scanning the selected mass. For a given setting of the high voltage pulse trains, the selector transmits a certain mass. This mass can be changed by scaling the three characteristic times τ_p, τ_d, τ_w by the same factor. The scaling is most easily done if one uses the same high voltage pulse train for both acceleration and deceleration. In this case, one has $\tau_d=\tau_w$, and the timing of the pulse train can be described by a parameter $\alpha=\tau_p/(\tau_p+\tau_d)$. The transmitted mass is then given by

$$m=\frac{eU_a}{d_1 x}\frac{\alpha}{f^2}. \tag{11}$$

Varying the frequency f for constant α changes the transmitted mass, but does not influence the transmission T.[2] The f^{-2} dependence was found to be very reliable over a wide range of masses.

Figure 5 shows a typical mass spectrum of small positively charged copper clusters obtained as described above. From this spectrum a mass resolution of $m/\Delta m\approx 22$ can be determined. The theoretical value of the mass resolution is given by Eq. (10): for this case, of a 3 mm wide exit slit and a lateral displacement of 120 mm, one obtains a resolution of $m/\Delta m=40$. The fact that the measured resolution does not fully meet this value is probably due to ringing of the high voltage pulses and to imperfect focussing of the ion beam.

The mass selector has now been combined with cluster sources of different types.[3,4] It is simple to operate and has been used routinely over the past two years without showing any problems.

The selector is the subject of the following patent applications: European Patent Application No: 97905297.4, Japanese Patent Application No: 530714/97, and U.S. Patent Application No: 09/125824.

ACKNOWLEDGMENTS

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[1] SIMION, D. A. Dahl and J. E. Delmore, Idaho Engineering Laboratory, EG&G Idaho Inc., P.O. Box 1625, Idaho Falls, ID 83415.
[2] When using one single high voltage pulse train instead of two, the achievable transmission is of course slightly lower than the optimum value as given by Eq. (5).
[3] I. M. Goldby, B. V. Issendorff, L. Kuipers, and R. E. Palmer, Rev. Sci. Instrum. **68**, 3327 (1997).
[4] S. J. Carroll, Ph.D. thesis, The University of Birmingham, 1999.