Focusing of charged particle beams with various glass-made optics

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Abstract.
We have developed methods to focus slow highly charged ions, MeV ions, and muon beams with various glass-made beam optics. (1) A focusing effect for an Ar⁸⁺ beam of 8 keV through a cm-long tapered capillary was obtained with a density enhancement of the transmitted beam compared with that of the input beam, which increases from 1 to 6 as the input current decreases from 30 pA to 0.8 pA. To study the stability of the transmitted beams through a glass capillary, we have measured the transmission of an 104 keV Ar⁸⁺ beam through a gap between a pair of parallel glass plates, and observed a precisely vibrational output current. (2) For 4 MeV He⁵⁺ beam, a 100 times density enhanced beam by a cm-long tapered capillary with a closed outlet was utilized to irradiate a cell in liquid. The range of the beam was controlled by the closed outlet with accuracy of ∼1 µm. (3) Using 40 cm-long tapered glass tubes, a density enhancement of a factor of ≃2 was observed for both positive and negative muon beams with an energy of 13 MeV.

1. Introduction
Interaction of slow highly charged ions (HCI) with multi-capillaries has recently been intensively studied experimentally [1-11] as well as theoretically [12-14]. One of the most prominent features is a so-called guiding effect, a transportation of slow HCIs along the capillary axis keeping their initial charge states even when the capillary axis was tilted against the beam direction. The fact that most of the guided ions keep their initial charge suggests that the ions do not touch the inner wall of the capillaries during transportation, i.e., an amazingly well-tuned electric field is automatically formed in each capillary. In other words, a self-organized charge-up of the inner wall plays an important role in realizing the guiding effect. A somewhat similar but qualitatively different phenomenon has been reported for MeV ion beams by Nebiki et al. [15], where a 2 MeV He⁺ beam was transmitted through a cm-long single tapered glass capillary. In this case, multiple small angle scatterings are expected to be the major player to induce the phenomena, and the beam density at the capillary outlet was found to be much higher than that at the inlet, which is called a focusing effect. It is then an interesting question whether various...
charged particles are guided and focused by a tapered single capillary. We briefly describe here how slow HCI's [16-18], MeV ions [19, 20], and positive/negative muons [21] are really focused by a tapered single glass capillary (tube).

In section 2, the focusing effect as well as the guiding effect are discussed for 8 and 64 keV Ar\(^{8+}\) beams transmitted through a cm-long glass capillary, which could produce a nanobeam of slow HCIs. Further, a transmission experiment with a 104 keV Ar\(^{8+}\) beam through a gap of a paired parallel glass plates is described, which shows a new “in-plane” guiding effect ([22]). In section 3, the focusing effect of ∼MeV ion beams is described as well as a possible application to a cell irradiation. In section 4, the focusing effect observed for 54 MeV/c (13 MeV) positive/negative muon beams transmitted through 40 cm-long tapered glass tubes is discussed.

2. Transmission experiment with HCI beams

A beam with the diameter of less than 1 micron (hereafter, nanobeam) of slow HCIs is not yet practically available because HCI beams are sometimes so weak that the beam intensity is reduced drastically by a collimator or slit. In addition, collisions of the beam with the inner wall of the collimator or the slit induce contamination due to charge-changed ions. When magnetic and/or electrostatic lenses are combined, good emittance is required. Single tapered glass capillary has the following advantages. (1) Slow HCI can be reflected from the inner wall without a close collision, keeping its initial charge state. (2) The taper can enhance the density of the output beam. (3) The size of the output beam is the same size of the outlet inner diameter. Considering these points, single tapered glass capillaries are one of the feasible techniques to produce a nanobeam of slow HCI.

The first experiment of the transmission of slow HCI beam through insulator capillaries was performed by Stolterfoht et al. [1]. They found that most of the transmitted ions kept their initial charge state for a 3 keV Ne\(^{7+}\) beam through a multi-capillary foil of polyethylene terephthalate (PET). In subsequent experiments employing PET [2-7], SiO\(_2\) [8, 9] and Al\(_2\)O\(_3\) [10, 11] foils, the transmission of beams without charge-state changing was observed. The results of simulations have explained the slow HCI transmission without touching the inner wall in terms of the dynamics of the charge-up distribution induced by the beam irradiation. The guiding of electron beams was also reported recently [23, 24]. The phenomenon can be explained as follows: The incident ions entering the capillary hit the inner wall and consequently cause it to become charged. When the accumulated charge becomes large enough to prevent the following incident ions from touching the inner wall, the ions travel more or less parallel to the wall, and then the ions can exit from the outlet. If the diameter of the outlet is smaller than that of the inlet, and is in µm/nm order, a density enhanced micro-/nanobeam of slow HCI is realized.

2.1. Experimental setup

![Figure 1. A schematic view of the experimental setup][16].

An ion beam of 8 keV Ar\(^{8+}\) was extracted from a 14.5 GHz Caprice ECR (Electron
Cyclotron Resonance) ion source at RIKEN, and then transported to an experimental chamber, where the vacuum level was \( \sim 10^{-5} \) Pa, via a mass/charge selecting magnet. The ion beam was then cut by a 2 mm \( \phi \) aperture (see Fig. 1), injected in a tapered glass capillary made of borosilicate, and finally detected by a PSD (Position Sensitive Detector) set at 125 mm downstream from the capillary inlet via a deflector. The divergence of the incoming beam after the aperture was at most \( \pm 3.3 \) mrad. The deflector was used for the charge-state analysis of the transmitted ions. The tapered capillary was prepared by heating a straight glass tube, and then stretched by pulling both ends with a constant force. The taper angle can be controlled by tuning the temperature and the force \[17\]. The outer and inner diameters of the capillary at the inlet were 2 mm and 0.8 mm, respectively, and those at the outlet were 55 \( \mu \)m and 24 \( \mu \)m, respectively. (Hereafter, \( D_{\text{in}} \) for the inlet inner diameter and \( D_{\text{out}} \) for the outlet inner diameter.) A typical length of the capillary was 50 mm. To avoid macroscopic charge-up of the entrance surface of the capillary it was covered by Al foil with a 0.8 mm hole. The foil was used to monitor the incoming ion current of 0.1 - 100 pA.

2.2. Density enhancement of the output beam

For a capillary of \( D_{\text{out}} = 24 \) \( \mu \)m, the transmitted beam was guided within as large as \( \pm 87 \) mrad \( (= \pm 5^\circ) \), and had a circular spot at the PSD and the divergence of \( \pm 5 \) mrad without the charge transfer inside the capillary, where the transmission rate was at most 1% \[16\]. Without tilting the capillary with respect to the beam axis, we have observed stable transmission for more than 1200 sec as seen in Fig. 2, where the maximum counts at the PSD was about 1600 cps \[16\].

![Figure 2. Time dependence of the number of transmitted ions. The maximum counts at the PSD was about 1600 cps [16].](image)

![Figure 3. Input current dependence of the enhancement factor for 8 keV Ar\(^{8+}\) beam.](image)

The density enhancement factor, \( \sigma \), was introduced and defined by the ratio of \( N_t/S_o \) to \( N_i/S_i \), where \( N_t \) is the number of transmitted ions, \( N_i \) the number of injected ions into the capillary, and \( S_o \) and \( S_i \) the geometrical outlet and inlet cross sections of the capillary, respectively. For this measurement, the density of the output beam was larger than that of the input beam, where the factor, \( \sigma \), was approximately 10 \[16\]. However, the density enhancement could depend on the beam energy, the charge state of the ion, the ion mass, the input current intensity, outlet size and surface resistivity of the capillary and so on. We examined the input current dependence of the density enhancement. In the experiment, to reduce the individual characteristics of each glass capillary, three capillaries with \( D_{\text{out}} = 20 \) \( \mu \)m, which were made under the same conditions, were used. Figure 3 shows the enhancement factor as a function of the input current without tilting the capillary. The horizontal and vertical errors were estimated to be about 10 % due to the input beam fluctuation. The enhancement factor was found to decrease from 6 to 1 with the input current varying from 0.8 pA to 30 pA. To

\[ \text{Counts at PSD [cps]} \]

\[ \text{Time since beam started [s]} \]

\[ \text{Enhancement factor} \sigma \]

\[ \text{Input current [pA]} \]
confirm the upper limit of the enhancement, measurements with input currents less than 0.8 pA are needed.

2.3. Nano-meter sized beam

Slow HCIs have high ability to modify surfaces and cause efficient sputtering without damaging the substrate very much. For example, a single HCI induces a nanometer sized dot on graphite [25-27] and Al$_2$O$_3$ surfaces [25]. It was also found that the F-Si bond direction of a Si(001)-F surface can be reconstructed from the 3D momentum distribution of F$^+$ ions desorbed by slow HCIs, i.e., a stereo-chemical analysis can be done with slow HCIs [28]. Once a nanobeam is available, these functions specific to slow HCIs can be used to realize various nano-fabrications, e.g., nano-patterning of nanodots and element-sensitive nano-imaging.

![Figure 4](image)

**Figure 4.** A manufactured outlet ($D_{out}=500$ nm) of a glass capillary by FIB.

The transmitted beam through a capillary with $D_{out}=900$ nm was successfully obtained for Ar$^{8+}$ ions with an energy of 64 keV. The spot at the PSD had widths (FWHM: full width at half maximum) of 0.30 mm and 0.40 mm in the vertical and horizontal directions, respectively. The corresponding divergences were ±2.0 mrad and ±2.7 mrad, respectively [17]. An outlet with $D_{out}$ more than 900 nm has been obtained by cutting around the outlet under an optical microscope. Cutting is performed by gripping the edge of the capillary at the required $D_{out}$, then bending and breaking the capillary, since the outlet diameter is ~100 nm just after the shaping by the heating and pulling described above. However, it is impossible to manufacture the outlet with $D_{out}$ smaller than several hundreds of nm by this method. Therefore a focused ion beam (FIB: Hitachi FB-2100) of 40-keV Ga$^+$ was employed for this purpose. Figure 4 shows a thus manufactured outlet of a glass capillary with $D_{out}=500$ nm. The scanning parameters of the Ga$^+$ beam, e.g., scanning directions, interlace rate, scanned region, etc., were tuned to prevent the Ga$^+$ beam from being bent due to the charge up around the capillary outlet.

2.4. Vibrational transmission of HCI beam through a gap of a paired parallel glass plates

In order to provide a nanobeam as a tool, stability of the transmission is required. The stability is expected to be sensitive to the balance between the charge-up and the discharge on the inner wall. To make the wall shape simple, we have observed slow HCI transmission through a gap between paired parallel glass plates (See Fig. 5(a)), instead of a tapered glass capillary.

![Figure 5](image)

**Figure 5.** (a) A schematic drawing of the setup, (b) The transmitted current through the gap as a function of the irradiation time for the entering current of 120 pA.

The glass plate used in this study was 26 mm in length, 20 mm in width and 0.9 mm in thickness, which was a piece of a microscope slide made of soda lime glass. The gap in the
paired plates was set to 0.1 mm. To avoid macroscopic charge-up of the entrance surfaces of the slides, they were covered with conductive paste. An aluminum vertical slit of 0.17 mm was installed at the entrance of the gap. The transmitted current through the gap was measured by an aluminum plate at ∼120 mm downstream from the exit of the gap. Figure 5(b) shows the transmitted current as a function of the irradiation time for an incident current of 120 pA of 104 keV Ar8+ beam. As seen in Fig. 5(b), repetition of the transmission was observed, as well as a report of a transmission experiment for 240 keV proton beam with a glass tube [29]. It is worth noting that the precise structure was almost kept in the repetition. It means that the reconstruction and the decay of the charge-up on insulator surface can be well-arranged. Further analysis is in progress.

3. Transmission experiments with MeV-energy ion beams

An application of µm-/nm-sized He ion beams in a MeV region with glass capillaries has already been performed for the analysis of materials [15, 30]. In the work of [15], transmission of 2 MeV He2+ ions without significant energy loss was reported. We have developed another technique using MeV-energy ions for biological applications with closed outlet capillaries.

3.1. Range control with a closed outlet

If the outlet of a glass capillary is closed by a thin lid for a beam injection into liquid, it is possible to control the range of the beam in the liquid, because the energy-loss of ions within the lid can be controlled by adjusting the thickness of the lid. Recently, glass capillary with a sub-micron-thick glass lid was introduced to realize the irradiation of wet samples, especially living cells [19, 20]. In the experiment, 4 MeV He2+ ions were injected into liquid scintillator and the scintillation volume, which corresponds to the range and struggling of the ions, showed good agreement with a simulation by SRIM-2006 [31]. The enhancement factor was observed to be more than 100. Figure 6(a) shows how to fabricate a capillary outlet with a thin lid. FIB is used to cut the thick lid, which is already made by filling with melted glass. Figures 6(b) and (c) are the photographs of the fabricated outlet taken by an FIB device and an optical microscope, respectively. The thickness was set to 1 µm. The arrows in Figs. 6(a) and (b) are the trace of Ga+ beam.

![Figure 6](image)

**Figure 6.** (a) FIB is used to cut the thick lid, which is already made by filling with melted glass. (b) and (c) are photographs of the fabricated outlet taken by an FIB device and an optical microscope, respectively. Arrows in (a) and (b) are the trace of the Ga+ beam. (d) ion irradiation of a cell in liquid.

3.2. Application to cell irradiation

Figure 6(d) explains how to use the capillary for ion irradiation of a cell in liquid without breaking the vacuum of the beam transport or the wet environment of the cell. A typical animal cell is about 50 µm in diameter, composed of complex intracellular organelles and assembled
macromolecules with various shapes and sizes. Selective inactivation or disruption of appropriate cellular structures, ranging in size from several µm down to 100 nm or less, would be an attractive approach to investigate their function in living cells.

When an energetic ion is injected in a cell, chemical bonds of biological molecules are broken via electronic (excitation, ionization) and nuclear recoil processes as well as secondary processes due to various radicals formed in the cell. It is then naturally expected that a micro- or nanobeam of ions has a high potential to induce space-selective inactivation or disruption. For this purpose, many research groups have been intensively working on the preparation of microbeams of energetic ions. In a conventional and straightforward scheme, a well-focused energetic ion beam prepared in vacuum is extracted in air via a vacuum isolation window and then shot into a biological cell in water. In most of those cases, the ions penetrate through the sample, thus the irradiation is made along the one dimensional ion path. The glass capillary with a lid has a remarkable advantage to control not only two dimensional spatial resolution but also the range in the material. In other words, the distance from the outlet to the Bragg peak can be set inside a cell with the accuracy of ∼1 µm for 2 ∼ 4 MeV He2+ ions [19]. Moreover, according to simulations by SRIM-2006, the bombardment region can further be reduced down to ∼100 nm when a 20 keV He2+ beam is injected in a capillary with a lid thickness of 100 nm and an outlet diameter of 100 nm.

In order to demonstrate further that the microbeam so prepared can induce deactivation of a biological cell, a HeLa cell whose nucleus was labeled with a fusion protein of histone H2B and green fluorescent protein (GFP) was bombarded by a He2+ beam (100 pA) prepared by a capillary with a lid thickness of 7.3 µm and Dout of 9.6 µm. With a beam energy of 4 MeV at the capillary entrance, photobleaching of only part of the cell nucleus was successfully observed [20]. By inserting a capillary into the cell just like a microinjection method, “surgery” within an arbitrary region of a cell becomes possible. This provides a new technique to study the function of intracellular structures.

4. Transmission experiments with muon beams

Considering the focusing of MeV ion beams due to multiple small angle scatterings as seen in the previous section, a muon beam with an energy of more than MeV could be focused by a tapered glass tube. Consequently experiments investigating the focusing of muon beams were performed.

4.1. Utilizing muon beam as a probe

Muons implanted into a sample are quickly thermalized and then localized at characteristic sites. These muons interact with the local magnetic field, resulting in muon spin precession and/or depolarization. This makes spin polarized muons very sensitive and versatile microscopic probes to study magnetic properties in various materials. One of the obstacles of this technique is that the size of the polarized muon beam from accelerator facilities is relatively large; typically a few tens of mm FWHM at the focal point. However, most of intriguing samples, such as newly synthesized chemical compounds and biological samples, are often much smaller in size than the beam diameter. Therefore, density enhancement of muon beams by capillary focusing should have many potential applications.

4.2. Experiments of focusing the muon beams

The experiment was performed at Port 2 of RIKEN-RAL Muon Facility at Rutherford Appleton Laboratory, U.K. [32]. Figure 7 schematically shows the experimental setup [21]. Muons with specified momentum are transported to the beam port, and extracted into atmosphere through a thin Mylar foil and an aluminum collimator of inner diameter 40 mm and length 85 mm. Beam divergence is ∼2° in standard deviation. The beam has a pulsed
structure with a repetition rate of 50 Hz, each pulse consists of two bunches of 70 ns FWHM separated by 320 ns. Typical beam intensity is about $10^4$ muons per pulse for the momentum of 54 MeV/$c$. Positron/electron contamination is negligible. Considering the size of the muon beam, we used tapered glass tubes with $D_{in} = 46$ mm. The tubes were made of Pyrex, whose components are similar to those of borosilicate. In order to make variations of taper angles, the length of the tubes was 400 mm, $D_{out} = 3$-20 mm, and the wall thickness of the tube was 2 mm. A beam momentum of 54 MeV/$c$ (13 MeV) was used because muon loss in air of 400 mm is negligible, while muons stop completely in a 2 mm glass wall, so that it does not interfere with the muon intensity observation. We prepared plastic scintillation counters (EJ212) with thickness of 0.5 mm and diameters of 20 mm. The scintillator was placed 3 mm downstream of the tube to monitor the muon intensity. The pulsed muon signal from the photomultiplier tube was averaged over 128 beam pulses with a digital storage oscilloscope.

Figure 8 shows the enhancement factor $\sigma$ as a function of $D_{out}$ together with the results of the numerical simulation, where $\sigma$ has the same meaning as other sections but is defined as the ratio of the yields of scintillation with the glass tube to that without the tube. In the simulation; 1) the energy loss is scaled from proton data using a semi-empirical formula, 2) the energy straggling is given by the Vavilov distribution, and 3) the angular distribution is given by the Moliere expression for multiple scattering [33]. As seen in Fig. 8, beam density enhancements were observed for all the conditions studied, and furthermore the simulation successfully reproduced the over-all behavior of the experimental results. This enhancement occurs because a certain fraction of muons incident on the inner wall is reflected via small angle scattering and transported to the outlet of the tube. The enhancement factor $\sigma$ increases from $\sim1.5$ to $\sim2.3$ as $D_{out}$ decreases from 20 to 3 mm. It is worth noting that the enhancement for negative muons was almost identical to that of positive muons [21]. The enhancement factor of $\sim2$ can reduce beam time by half. Moreover this focusing method with the glass tubes is free from magnetic field, which might interfere with the magnetic field for $\mu$SR measurements. The simulation also suggests that the density enhancement factor improves further when the tube is made of heavy materials like copper, lead, or gold. Preparations in this direction are in progress.

5. Summary and outlook

For HCl beams, a focusing effect for 8 keV Ar$^{8+}$ beams through a cm-long tapered capillary ($D_{out} = 20 \, \mu m$) was observed with a density enhancement, which increases from 1 to 6 as the input current decreases from 30 pA to 0.8 pA. A beam size of 900 nm in diameter was obtained for Ar$^{8+}$ beams of 64 keV. In order to investigate the stability of the transmission, paired parallel glass plates have been used. Further detailed study of the focusing effect and the stability of the
transmission enables us to provide a stable, density-enhanced HCI nanobeam for, e.g., nano-patterning and element-sensitive nano-imaging.

Using MeV ions, a glass capillary with a lid has a remarkable advantage to control not only two-dimensional spatial resolution but also the range in the material. It means that the distance from the outlet to the Bragg peak can be set inside a cell with an accuracy of ∼1 µm for 2 ∼ 4 MeV He^{2+} ions. This method is one of the feasible techniques for selective inactivation or disruption of appropriate cellular structures, ranging in size from several µm down to 100 nm or less, to investigate their function in living cells.

We observed a density enhancement of a factor of ≃2 for both positive and negative muon beams with an energy of 13 MeV using 40 cm-long tapered glass tubes. In order to obtain further density enhancement, tubes made of heavy materials like copper, lead, or gold will be examined.

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