PRESENT AND FUTURE PROJECTS OF TMU ATOMIC PHYSICS GROUP

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Abstract. The experimental atomic physics group of Tokyo Metropolitan University now intensively promotes three projects; 1) research of huge molecular ions by a liquid-nitrogen-cooled electrostatic ion storage ring, 2) research of highly-charged ion interaction with a crystal using high-energy heavy ion storage ring, and 3) research of very low-energy ion-atom interaction through ion swarms in helium gas cooled down to 4 K. In this report, these projects are briefly introduced in a comprehensive manner.

INTRODUCTION

For more than a decade, we have been mainly involved in the low-energy highly-charged ion (HCl)-collision experiments using two compact liquid nitrogen (LN₂)-cooled EBIS [1], and a 14 GHz ECR [2]. We have been investigating HCl interaction with electrons, atomic ions, molecular ions and cluster ions as well as their reaction dynamics in low energy collisions. Without restricting our research in this field, we now focus on several new projects expanding our activities.

The first project is an electrostatic ion storage ring. We are now under development of a new electrostatic ion storage ring, which is free from the limit in mass of circulating ions, and is able to store heavy molecular/cluster ions. This ring is considered to be a promising tool to open up application to the nano-particle and biological sciences. We specially designed a compact-sized ring to be kept at LN₂ temperature in order to cool down stored molecular/cluster ions to the vibrationally ground state via infrared radiation. We have already succeeded in storage of heavy atomic ions, and we are now trying to store various types of ions.

The second one is investigation of resonant coherent excitation of high-energy heavy ions induced by the periodic crystal field. Several tens GeV heavy ions are supplied by a new Japanese synchrotron storage ring dedicated to medical treatment. Owing to excellent coherence due to high energy of projectiles, it is possible to perform high-resolution atomic spectroscopy of highly-charged heavy ions with a few bound electrons. Recently we have also succeeded in exciting the ions into specific m-states (magnetic sub level) utilizing polarization of the periodic crystal field.

The third one is an ion swarm equipment in helium gas cooled down to 4 K. We observed that the ions injected into the He gas medium at very low temperature form helium clusters, i.e., ions attached with several He atoms. In addition, we have recently found exceptionally low ion mobility characteristic to molecular ions due to transient
rotational excitation.
Thus, we now deal with the wide range of incident ion beam energy, i.e., from several tens GeV down to less than meV, and handle a variety of targets from atomic targets to molecules, clusters, up to crystals. In this report, we briefly introduce these topics.

LIQUID-NITROGEN-COOLED ELECTROSTATIC ION STORAGE RING

A new electrostatic ion storage ring is now under construction by collaboration of atomic physics and cluster chemistry groups in Tokyo Metropolitan University (TMU) [3]. Recently electrostatic ion storage rings have been successfully applied to research in atomic physics by the Aarhus group [5] and the KEK group [5]. These rings are, in principle, free from the limit in mass of circulating ions, because the electrostatic rigidity is proportional to $E/q$, and is independent of the ion mass, and are able to store heavy polyatomic molecular/cluster ions. The ring has another unique feature that the vibrationally excited states of these molecular ions are deexcited to the environmental temperature through infrared radiation in the case that molecular ions circulate in the ring. In addition, the ring control is straightforward because of absence of hysteresis, and is designed in a compact table-top size.

We have started its construction in 2003. Our new ring is basically designed under a common concept with the preceding rings, however, has some unique characteristics. We specially designed our ring so that it can be cooled down to the LN$_2$ temperature. A fraction of large molecular or cluster ions still remain in the excited states at room temperature, however, our new ring enables them to vibrationally de-excite completely down to the ground state.

The outline of the TMU electrostatic ion storage ring is shown in Fig.1. The compact-sized ring for the maximum ion energy of 30q keV has a race-track-type shape of 7.7 meter in circumference with two straight regions of 1.2 m in length for collision experiments. It is equipped with two 160$^\circ$ cylindrical electrostatic deflectors, four 10$^\circ$ parallel plate electrostatic deflectors which allow injection and extraction of ions, and four focusing and defocusing quadruple doublets. A period of ion circulation in the ring is 3.2 $\mu$s for 30 keV proton, and it increases in proportion to the square root of mass. All parts and structures were designed taking shrinking due to cooling into accounts in advance, and they were machined and constructed with precision of $\pm$ 0.5 mm. A good vacuum is crucial for storage of low-energy, large molecular ions. We attained the pressure of $5 \times 10^{-9}$ Pa in the ring by prebaking at 900 $^\circ$C of all instruments before assembling, together with baking of the whole ring for about 5 days at 200~300$^\circ$C.

Concerning beam diagnosis, four non-destructive beam position monitors (BPM) are installed to pick up induced charges resulting from the passage of a charged beam nearby. They provide information of the beam center and the circulation period. The BPM is a cylindrical electrode of 50 mm in diameter, 100 mm in length with field clamps at both sides. It is divided into four parts, and the beam center is derived from the ratio of charge induced in each divided electrode. We expect that highly-resolved mass spectra are obtained from this BPM signal by applying the fast Fourier transformation (FFT)
taking advantage of the constant circulating frequency. Another detector for neutral particles produced by fragmentation of the primary ions is placed on the extension of the straight region. A micro channel plate (MCP) and a phosphor screen, whose image is recorded by a CCD camera.

In order to cool down the temperature of the electrodes surrounding the ion trajectories, all of them are covered with thermal shields, in which LN$_2$ circulates, and are cooled radiatively. The electrodes are floated by insulators of alumina with good thermal conductivity, and the thermal shields are sustained by four rods of titanium alloy with low thermal conductivity. In addition, thin stainless reflectors are inserted for heat insulation between the shield and the chamber wall. Thus, the 80% region of the beam trajectory is surrounded by these cooled thermal shields. The temperature of the thermal shields reached about 80 K after 24 h of cooling, and that of the electrodes approached the equilibrium.

We constructed the ion injection system so that several different ion sources can be switched by an electrostatic quadruple selector. For a benchmark test of the ring operation, we prepare a duoplasma-type ion source, which produces both of positive and negative ions, and a laser-ablation type ion source for production of carbon cluster ions. Simultaneous injection of various types of ions is possible in principle, which is one of advantages of the electrostatic control of the ion trajectory. On the other hand, mass selection is in need for the injection of single species of ions. In the setup using the duoplasma-type ion source, extracted ions are accelerated to 30q keV, chopped in a pulse mode by combination of apertures and an electrostatic deflector, and charge/mass selected by an ExB Wien filter. After such mass selection, they are injected into the ring by operation of the 10° deflector in the synchronized pulse mode. We prepare a doubly differential pumping system between the ring and the ion sources because of the relatively poor vacuums in these ion sources compared with that of the ring.

We have already succeeded in storage of rare gas ions like Ne, Ar, Kr, Xe singly charged ions, and now under optimization of a variety of operation parameters and checking of beam diagnosis detectors. We plan researches concerning temperature dependence for metastable structure and isomerization process of isolated ions, lifetime measurements of doubly charged ions, as well as low energy collision dynamics between ions and neutral particles by using merging beam techniques.
RESONANT COHERENT EXCITATION OF HIGHLY CHARGED ION CHANNELING THROUGH SILICON CRYSTALS

When energetic ions of more than hundreds keV/u penetrate a single crystal parallel to a crystal axis or plane, the ions travel in an open space without colliding with atoms in atomic strings or planes guided by the static crystal field, which is generally called "channeling". The channeling ions feel a time-dependent oscillating field also in the projectile flame, since they pass across a periodic array of the atomic strings or ordered planes. If an energy, corresponding to the frequency of this oscillating field matches with a transition energy of an internal degree of freedom of the ions, excitation called Resonant Coherent Excitation (RCE) takes place. In order to excite an innershell electron of highly charged heavy ions, a spacing between atoms is of the order of several Å. Accordingly, heavy ions of several 100 MeV/u is in need. In history, RCE was theoretically proposed by Okorokov in 1965 [8], and Datz in ORNL has for the first time succeeded in observation of RCE in 1978 [9]. Recently we have started the experiments of RCE at the Heavy Ion Medical Accelerator in Chiba (HIMAC) [10][11]. The accelerator covers up to several hundreds MeV/u for the maximum attainable velocity, and provides up to Kr as the heaviest ions at present. The advantages of the large velocity of heavy ions are not only the requirement to fulfill the resonance condition, but also high coherence, resulting from smaller probability of incoherent collision process of the ions with target electrons like excitation, ionization, and capture.

We have succeeded in observing resonant excitation spectra of highly-charged ions in high precision by three different methods which relate to different signals accompanying RCE between the ground and excited states. After the resonant excitation, the excited state proceeds to one of three channels; resonant de-excitation to the ground state, de-excitation by emitting X-rays to the ground state, and ionization by the collision with the target electrons. 1) The ions in the excited state are more easily ionized through the collision with the target electrons compared with those in the ground state. Therefore, the fraction of the higher charge state in the charge-state distribution of the emerging ions from the crystal increases under the resonance condition. 2) At the same time, the yield of electrons stripped off the ions (convoy electron) also increases under the resonance condition. 3) When the ions escape from ionization, X-rays are emitted. Increase of X-ray yield under resonance condition confirms RCE. The methods 1 and 2 are based on the same process, which compete with the process in the method 3. As the atomic number, \( Z \), of the heavy ion becomes larger, the ionization cross section is reduced, and the lifetime of the excited state is shortened. Therefore, the method 3 becomes efficient. We have observed RCE through all of these three methods.

For instance, we observed RCE of the 1s electron to the \( n = 2 \) state of 390 MeV/u hydrogen-like \( \text{Ar}^{17+} \) ions in planar channeling in a 21 \( \mu \text{m} \)-thick Si crystal. In order to fulfill the resonance condition, we varied the angle of the crystal with respect to the beam direction in the \((2\bar{2}0)\) plane from the [110] axis, \( \theta \), where the ions travel through the periodic array of atomic strings whose direction is defined by the 2D reciprocal vector of \((k/A,l/B)\), \((k,l:\text{integer})\). By scanning the incident angle, \( \theta \), the resonant excitation corresponding to the electronic transition from the 1s state to the \( n=2 \) level of \( \text{Ar}^{17+} \) ions were observed in several conditions of the incident angle, \( \theta \), corresponding to
combinations of \((k,l)\) as increases of the ionized \(\text{Ar}^{18+}\) fraction in the transmitted \(\text{Ar}\) ions. Figure 2 shows the resonance spectrum for \((k,l) = (1,1)\).

This resonance spectrum has complicated structures and basically consists of two peaks; a single peak at the higher energy side and a doublet peak with a dip at the lower side. Each peak is not symmetric; the single peak skews to the higher energy side, and the doublet peak has a tail to the lower energy side. These features are explained by the spin-orbit \((l \cdot s)\) interaction and Stark effect for the excited levels due to the crystal electric field. In order to obtain the detailed information of these complicated structures, we measured the deposited energy of the channeling ions into the crystal adopting a 21 \(\mu\)m thick silicon surface barrier detector (SSD) as a target crystal. By detection of the deposited energy by the SSD, it is possible to extract the information of the ion trajectory, because this deposited energy reflects the ion trajectory in the crystal. We simultaneously measured the deposited energy of 390 MeV/u \(\text{Ar}^{17+}\) ions by the SSD and the charge-state distribution of the emerging ions, which revealed such dependence more clearly.

We noticed that this technique has a possibility to determine the precise energy levels of the 1s ground state of heavy ions. We are able to determine the Lamb shift of the 1s state electron of \(\text{Ar}^{17+}\) in precision of several percents. This RCE technique is a completely new method of spectroscopy in principle compared with traditional wave length measurements with a Bragg crystal spectrometer, or energy measurements with a semiconductor detector.

We have also observed resonance profiles for the de-excitation X-ray from RCE heavy ions, which was clearly observed owing to the large X-ray yield \([12][13]\). As shown in Fig.3, resonance of the electronic transition from 1s to \(n=2\) states in \(\text{Ar}^{17+}\) ions corresponding to \((k,l) = (1,1)\) was clearly observed as an increase in X-ray emission yields for Si(Li) detectors as well as an increase in the ionized \(\text{Ar}^{18+}\) fraction in transmitted Ar ions, when we vary the tilt angle. The main difference of the resonance
profile between the X-ray yield and the charge state fraction is suppression of the peak intensity at the lower energy side, and is explained by the large fraction of 2s component of the excited level, and the de-excitation to the ground state by one photon is not allowed.

In addition, we also obtained the resonance profiles for the convoy electron yield [14]. We detected the electrons traveling in the same direction and at the same velocity as the projectiles. They originate in ionization of the projectiles, and often called as convoy electron. In the present condition, the energy of the convoy electron amounts to several hundred keV. We detected the convoy electron using a combination of a magnetic electron analyzer and a 5 mm-thick Si-detector. The resonance spectra quite similar to those obtained by the charge state measurement were successfully measured.

Finally we point out that we measured other heavy ions like Fe, Kr ions, and the transitions to the higher excited states like n=3,4. And we also observed not only hydrogen-like ions but also He-like ions both by the charge state measurement and the X-ray emission measurement. Furthermore, we recently found azimuthal anisotropy in X-ray emission of helium-like Fe$^{24+}$ ions due to alignment of the excited states resulting from the polarization of the periodic crystal field.

**ION SWARMS IN COOLED HELIUM GAS**

We have been interested in the ion transport properties in very low temperature gases for a long period. The ion swarm, which is a cloud of ions moving in a buffer gas under influence of an electric field, is one of research subjects in our group. We have developed a selected-ion-injected drift tube mass spectrometer for ion swarm experiments at very low temperature [15]. Helium gas temperatures of 77.4 K and 4.3 K in the drift tube are
achieved with cooling by liquid nitrogen and liquid helium, respectively.

Primary ions are produced with an electron-impact ion source, selected by a Wien filter, and injected into the drift tube through an aperture with a kinetic energy lower than 10 eV. The injected ions quickly thermalize in elastic collisions with helium gas and begin to drift towards the end of the tube under the influence of a weak uniform electric field produced by guard ring electrodes. Ions elected by an aperture of the end plate are analyzed by a quadrupole mass filter and detected by a secondary electron multiplier. We have two electrical shutters for drifting ions in the tube and measure drift time spectrum between each shutter.

By using this apparatus, we have obtained mobilities of various ions in cooled helium gas. The reduced mobility \( K_0 \) is defined by the following formula:

\[
K_0 = \frac{v_d}{N_0 E},
\]

where \( v_d \) is the drift velocity obtained from the arrival spectra, \( N \) is the number density of gas molecules, \( E \) is the electric field strength in the drift region, and \( N_0 \) is Loschmidt’s number \( 2.68676 \times 10^{25} \text{ cm}^{-3} \). The ion mobility in the gas is determined by the interaction potential between the ion and the gas molecules. If only the polarization potential \( V_{\text{pol}} = -\frac{\alpha q^2}{2\epsilon} \) is considered, the ion mobility has a finite constant value, which is called the polarization limit or Langevin limit:

\[
K_{\text{pol}} = 13.853/\sqrt{\alpha \mu} \quad \text{(in cm}^2\text{V}^{-1}\text{s}^{-1}),}
\]

where \( \alpha \) is the polarizability of the gas molecule in Å\(^3\) and \( \mu \) is the reduced mass of the ion and the gas molecule in amu. In the very low collision energy, the ion-neutral collision is governed by the polarization potential. Therefore the ion mobility should approach the polarization limit as the electric field and gas temperature are lowered. However, measured mobilities of diatomic molecular ions, \( \text{O}_2^+, \text{CO}^+, \text{and NO}^+ \), are lower than \( K_{\text{pol}} \) in the low electric field region at 4.3 K [16, 17, 18]. On the other hand, the mobility of \( \text{N}_2^+ \) in helium gas at 4.3 K shows ordinary behavior and approaches \( K_{\text{pol}} \) at very low field [16]. Recently the reason of this phenomenon has been revealed to be the transient rotational excitation, which is very similar to the Feshbach resonance from molecular dynamical calculations using accurate interaction potentials between the molecular ions and the helium atom [19].

Mobilities of atomic ions in helium gas have also been measured at 4.3 K. In the case of \( \text{He}^+ \), the mobility shows the quantum effect in very low electric field region [20]. This effect has been reproduced fairly well by the full quantum calculation [21].

Not only the ion mobility, but also the helium cluster formation has been investigated with the same apparatus. The helium cluster ions are considered to be formed through the following three-body ion-atom association reactions:

\[
X^+ + \text{He} + \text{He} \leftrightarrow X\text{He}^+ + \text{He}
\]

\[
X\text{He}_{n-1}^+ + \text{He} + \text{He} \leftrightarrow X\text{He}_n^+ + \text{He}
\]

(3)
where $X^+$ denotes the injected ion into the drift tube. We have observed magic numbers for several cluster ions. A common magic number $n = 12$ for the clusters which has $N_2^+$, $CO^+$, and $O_2^+$ as the ionic core indicates the icosahedral structure [22].

ACKNOWLEDGMENTS

We thank the collaboration of K. Komaki and Y. Yamazaki for the RCE experiment, which is one of research projects with heavy ions at NIRS-HIMAC. All of the works are in part supported by Grant-in-Aid for Scientific Research (no.13440126, no.14204062, no.14340124, no.16204030) from Japan Society for the Promotion of Science.

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