Atomic, Molecular & Optical Physics Laboratory

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Key Sentence :

1. Atomic and molecular collisions using an electrostatic ion storage ring
2. Atomic, molecular, and photonic physics utilizing the advanced photon sources
3. Resonant coherent excitation of fast highly charged heavy ions

Key Word :

atomic and molecular physics, atomic collision, ion storage ring, electrostatic ring, ion trap, isolated single molecule, cluster, droplet, merging collision, low-temperature reaction, excited ions, meta-stable ions, radiative cooling, chemical evolution in space, bio-molecular ion reaction, laser spectroscopy, ultra-short-pulsed intense laser, time-resolved spectroscopy, coherent control, highly-charged ions, heavy ions, fast ions, channeling, resonant coherent excitation, multi-electron excitation, de-excitation x-ray, single crystal

Outline

We experimentally study wide-ranged physical processes; cold chemical reaction relevant to universe evolution, dynamics of large complex molecules, atom interaction with a ultra short-pulsed intense lasers and crystals, using new approaches and new methods in atomic, molecular and optical physics.

We constructed a compact electrostatic ion storage ring dedicated for investigating molecular collisions and dynamics. Low-energy collisions and reactions of cold molecular ions in the specific vibrational and rotational states prepared by the ring is our primary mission to be explored. Taking advantages that an electrostatic ring has no limitation of mass of stored ions, we expect that the excitation and de-excitation dynamics of large bio-molecular ions and cluster ions as well as energy-differential cross section of relevant collisions are revealed.

We have been performing merging experiments by combining atomic and molecular beams with a variety of UV/VIS/IR lasers. We have been also trying manipulation of high-energy heavy atomic ions by the crystal periodic fields. We explore fast dynamics in the ultra short time range, and spectroscopy and manipulation of heavy ions in the energy region from EUV to X-rays.

1. Atomic, and molecular collisions using a cryogenic electrostatic ion storage ring

1.1 Cryogenic electrostatic ion storage ring

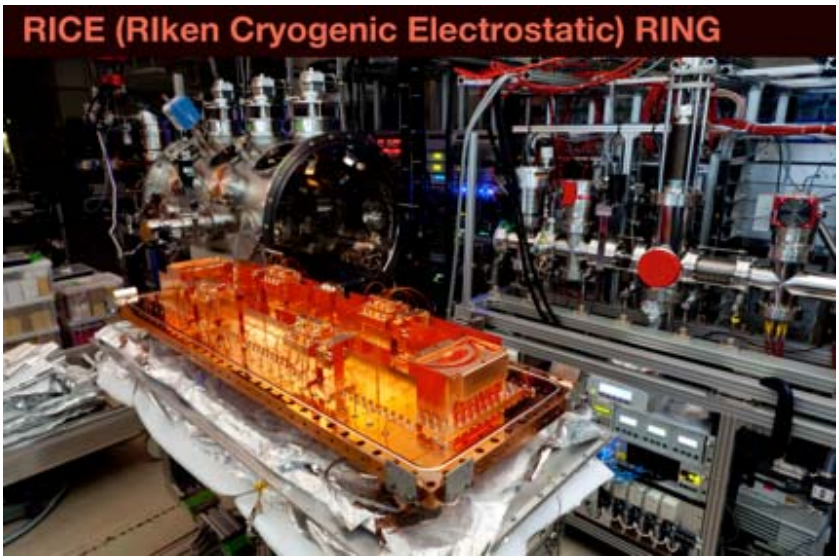

Fig.1 Photo of RICE and its electrodes on the plate

We have developed a cryogenic electrostatic storage ring (RIken Cryogenic Electrostatic Ring: RICE), which serves as a core apparatus for the production of a cold molecular ion beam in the Basic Science Interdisciplinary Research Project; Emerging Science Explored by Extreme Beam. The project aims to explore the quantum collision dynamics of the stored molecular ions in the specific vibrational and rotational states by the merging experiments with a beam of the neutral atoms. The cryogenic chamber provides an extremely high vacuum condition, which offers a much longer storage of the ion beam compared to the room-temperature apparatuses.

RICE satisfies ultra-low temperature and ultra high–vacuum environments simultaneously: the temperature of the inner vacuum chamber (IVC) reached down to 4.7K, and the vacuum was evaluated to be in the order of 10^{-12} Pa using the vacuum gauge monitoring the vacuum in the attached differential pumping chamber. In July 2014, the atomic ion storage of several 10 nA ions was attained, and experiments using molecular ions have been started recently after commissioning.

Lifetime measurement for a long period

Under the extremely high vacuum condition the neutral products resulting from collisions with residual gas is very limited. We monitored the temporal evolution of the intensity of the stored ions by extracting the ions after a specific period of the storage. From the decrease of the ion intensity during 30 min. a time constant of around 10 min. was obtained. It is noted that this storage lifetime is determined not by collisions with residual gas but by other effects like intra-beam Coulomb scattering.

Beam bunching

By keeping the bunch structure of the beam in the ring, non-destructive monitoring of the beam intensity using a Schottky detector is expected. Bunch formation of the stored ions was tried by applying the suitable sinusoidal voltage to a cavity electrode. We succeeded in detecting the bunch structure by using a 5^{th} order ellipsoidal filter to the output signal from the pickup detector, and evaluated the dumping of the stored beam intensity by the temporal change of the frequency spectra.

We now can trace the Schottky signal up to 200 ms by bunching, despite that the maximum time period was limited up to 5 ms without bunching. We plan to realize observation of the longer period by preparing a cryogenic pre-amplifier at the low-temperature area of the ring.

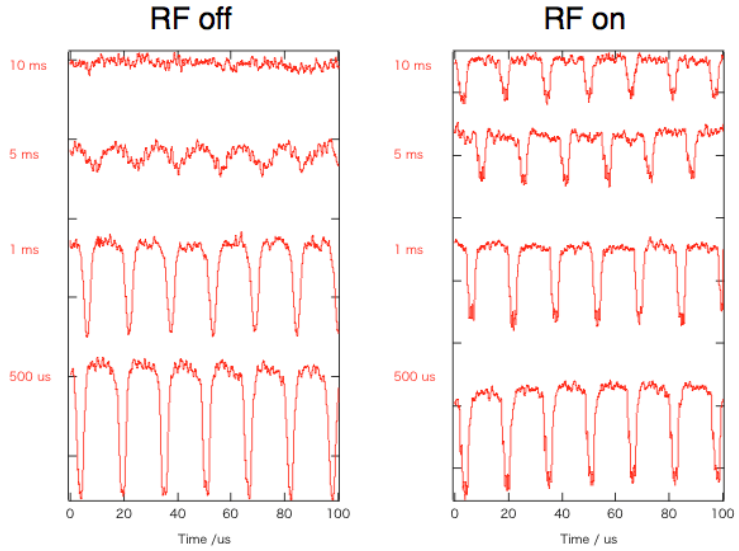

Fig.2 Schottky signals with and without RF for bunching

Laser merging experiment

Toward laser spectroscopy of molecular ions, we prepared experimental setups including a visible OPO laser and a dye laser. After evaluation of the gas pressure and the RF intensity dependences of the performance of an ECR ion source for molecular ion beam production, we succeeded in stable storage of the positive molecular ions in the ring.

We also developed an originally designed detector system to distinguish the laser light traveling co-linearly to the neutral fragments of the molecule. An Al-made mirror was placed at the angle of 45 degree with respect to the both beams. Laser light is reflected by the mirror, and detected by the photo

diode, while the energetic neutral products collide with the mirror and produce secondary electrons. They are accelerated to several keV and detected by a channeltron. After checking the proper operation, the detector system was installed to the beam line.

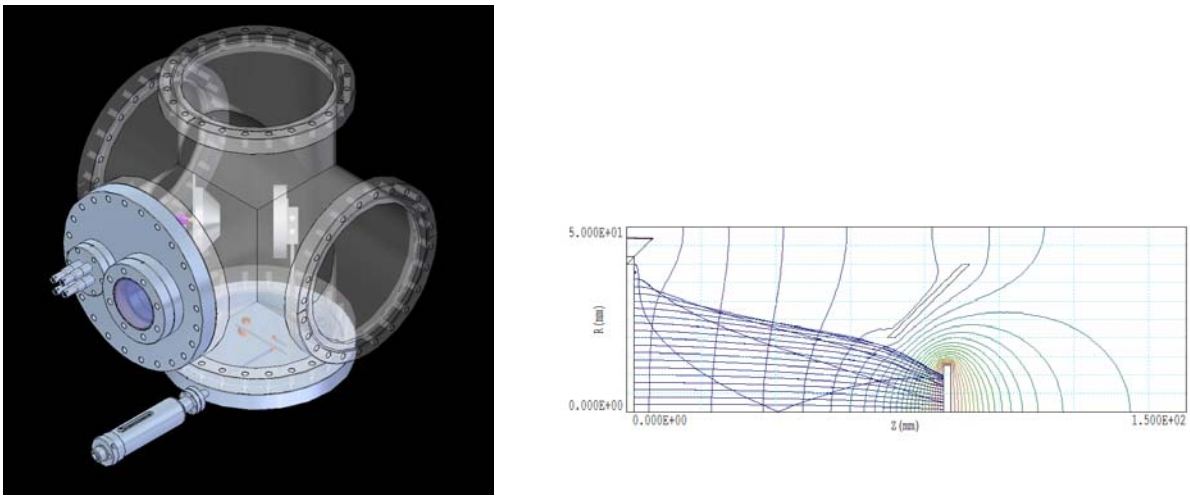

Fig.3 Schematics of the detector system (left), and potential configuration of the detectors and simulation of the accelerated secondary electrons (right).

1.2 Ion source and beam transport

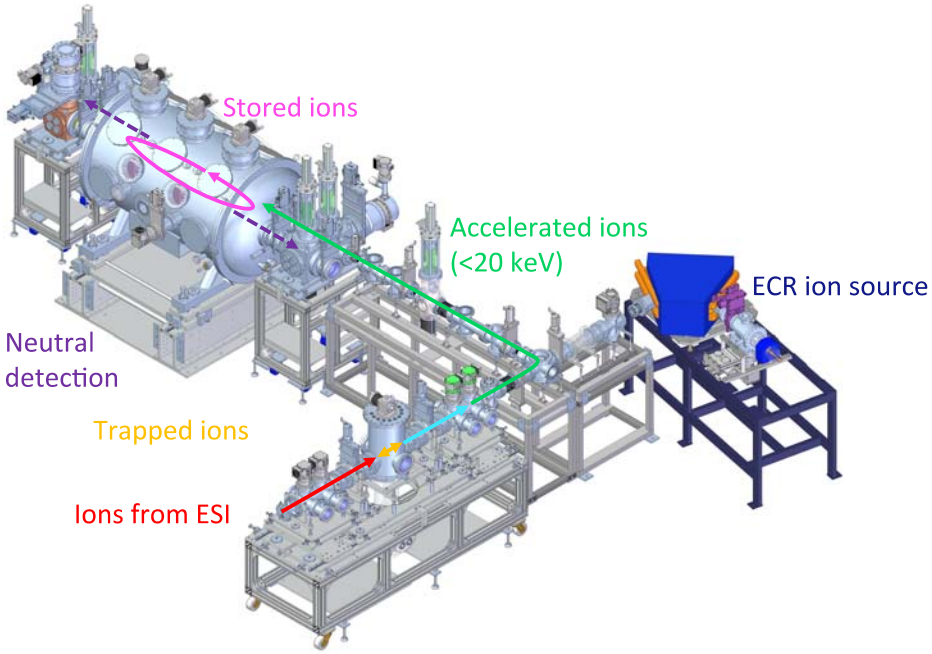

Fig.4 Ion injection system and RICE-Ring

A production system of cold ions delivered into RICE consists of an electrospray ion (ESI) source, a quadrupole mass filter, a cryogenic radiofrequency ion trap and an acceleration tube.

We aim to inject a bunched beam of cold ions into RICE. Large molecular ions are produced via the ESI source and can be disintegrated by passing them through a heated capillary. These molecular ions are guided by octupole radiofrequency fields and are mass-selected by the quadrupole mass filter. Then they are introduced into the cryogenic ion trap mounted on a 4K two-stage cold head of a GM cooler device. The stored ions are cooled by the He buffer gas collision under the environment surrounded by cold octupole electrodes.

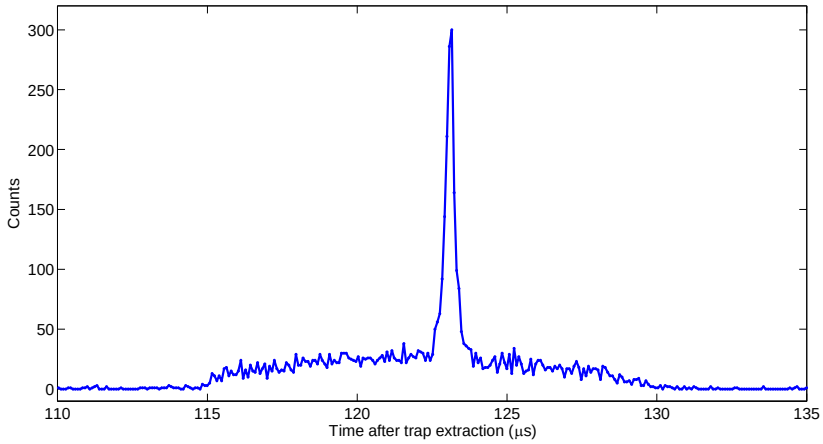

Fig.5 TOF spectrum of the neutral fragments produced by laser irradiation to extracted molecular ions $MB^{+}(H_2O)_n$ from the ion trap

To extract the stored ions from the ion trap fast and efficiently, we developed a modified version of the ion trap equipped with additional electrodes between octupole electrodes.
By applying a well-designed DC voltage to the correction electrodes in the ion trap, we add the pulsed slope-wise potential optimized for the ion extraction. Thus, the ions distributed widely in space inside the ion trap are focused in time and space at the specific point downstream of the trap, where a pulsed high voltage system is prepared. By applying a high voltage in a rapid rising time immediately after the ions bunches arrives at this point, We succeeded in producing a pulsed ion bunch of 10-20 keV in energy and a few 10 μs in width.

Then, we introduced a tunable visible OPO laser to the extracted beam at the right angle. We detected produced neutral fragments by a MCP placed downstream of the beam after expelling ion components by an electric deflector. As a typical example, a TOF (time of flight) spectrum of the neutral fragments produced by laser irradiation to the extracted hydrated methylene blue molecular ions $MB^{+}(H_2O)_n$ is shown in Fig.5. A sharp peak of less than 1μs in width corresponding to the component made by the laser irradiation is clearly observed in addition to the bunch component of about 15 μs in width resulting from collisions with residual gas of about 15 μs in width.

1.4 The neutral beam line

Taking an advantage of RICE, an ion-neutral collision experiment is planned. This study aims to investigate the dynamics of low-energy molecular ion's reaction with atomic neutrals, especially its dependence on the collision energy and molecular temperature are interesting. Merging experiments of a neutral atomic beam to a molecular ion beam stored in the ring are planned.

The neutral beam will be produced by laser photo-detachment of a negative ion beam produced by a Cesium sputter ion source. We developed a controlling system of the ion sources on the platform of the maximum 30 kV, and optimized the operation condition by monitoring and logging the Cs reservoir temperature and cathode current. We tested the vacuum condition of a photodetachment chamber utilized for neutralization of negative ions by laser, and prepared fuse-silica windows to the chamber for laser introduction.

We adopted a semiconductor solid laser for the intense photon flux, and constructed a pure water circulation system to cool the diode array as well as its interlock system. Pure water is circulated under the condition of the specific resistance of 2+/-1μS/cm and the temperature stability of +/-0.1degree. We also designed three types of focusing optics by a simulation software code ZEMAX, and prepared a test bench with multiple cylindrical lenses for monitoring a laser light profile.

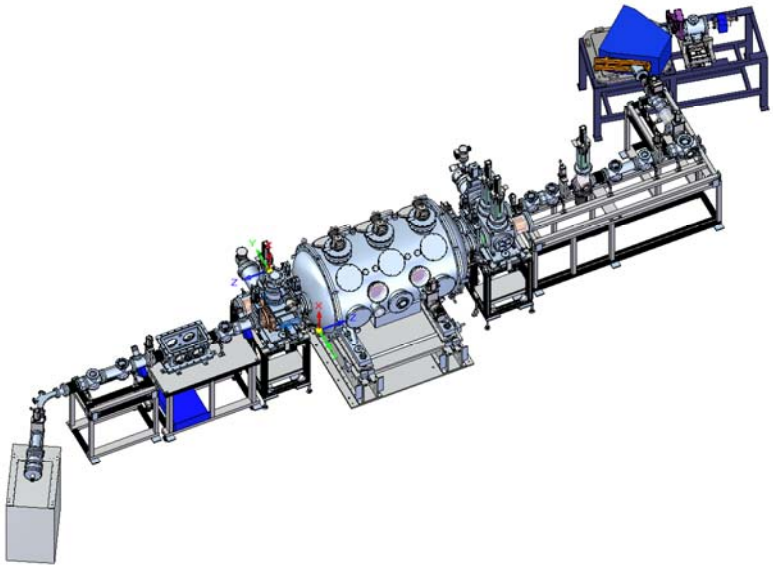

Fig.6 Schematic of ion –neutral beam merging experiments

1.5 The superconducting tunnel junction (STJ) detector operating at 1.6K

Reaction and decays of the stored ions in the electrostatic ion storage ring are generally studied by observing the neutral products by a detector placed at the extension of the straight section of the ring. The neutral product has the same velocity of the a few tens keV store ions, and often a micro channel plate (MCP) is adopted as the detector. The MCP provides the detection timing information, however, it has a disadvantage of the luck of energy information of the incident particles. Thus, assignment of a variety of the fragment from the reaction or decay is hard to achieve.

Aiming to introduce the detector equipped with enough energy resolution in the region of a few tens keV, we have been developing a superconducting tunnel junction (STJ) detector dedicated to the electrostatic ion storage ring in collaboration with Advanced Institute of Science and Technology (AIST).

In general, a STJ detector requires low temperature of around 0.3 K attained by a ^{3}He cryostat. We focused development of the STJ detector with the equivalent energy resolution even it is operated under the temperature attained by liquid ^{4}He pumping.

Specially fabricated STJ detectors with the size of 100μm x 100μm were cooled down to 1.6K obtained by liquid ^{4}He pumping, and 15q keV Ne^{q+} (q=1, 2, 3) ions extracted from the ECR ion source were injected to the STJ detector. The attained resolution was ΔE/E=15-20% at 1.6K. Subsequently, we performed the measurements at 0.3K, and reached a conclusion that the attained resolution is similar at both temperatures albeit the fact that the thermal noise is much reduced at 0.3 K. Using this detector, we can distinguish 20keV liner carbon chain C_5 and C_6 extracted from RICE. We are now developing an advanced detector element itself and modifying a detection circuit system for the higher resolution.

1.6 Molecular ions in superfluid helium droplets: exploring molecular ion dynamics at 0.4 K

Helium droplets are large liquid clusters of helium at 0.4 K. They can easily capture various molecular species and instantly cool the rotational and vibrational energies of the molecules down to 0.4 K. Additionally, the droplets are in the superfluid state, thus showing characteristic responses to the rotational motion of the captured molecules and to the relaxation process of the internal energies. This new project has started in 2014, aiming application of this helium droplet method to a wide range of molecular ions. With ions inside, the translational motion of the droplets can be controlled by conventional methods for charged particles. This project will be then focused on long-time scale molecular ion dynamics in helium droplets introduced into RICE.

Helium droplets are produced as a molecular beam from a high-pressure (> 1 MPa), low-temperature (< 20 K) nozzle of helium. After the expansion from a nozzle with a typical orifice diameter of 5 μm, helium atoms start to aggregate together to form large liquid clusters. Finally their temperature is lowered to 0.4 K by evaporation of the surface atoms. The resulting helium droplet beam is skimmed for molecular ion production at the downstream.

We developed and constructed major components of the droplet formation device, and tested the pulsed mode operation of the He droplet beam. By modifying a nozzle shape of a commercial solenoid

valve for general usage (Parker, Series 99), we succeeded in producing pulsed He droplet beam of repetition frequency of 10 Hz. For its detection, we adopted the TOF (time of flight) type mass analyzer. We also modified the design of the device for electron impact ionization, and attained an intense electron current by one order higher compared with the previous approach.

We observed multiple He-attached ions originating from He droplets as well as ions from water H_2O molecules in a TOF spectrum of the pulsed droplet beam capturing water H_2O molecules. Presently, we are preparing the experiments for observing the production of molecular ions inside He droplets as well as their dynamics using UV/VIS/IR lasers.

2 Atomic, and molecular collisions using a room-temperature electrostatic ion storage ring

2.1 Lifetime measurement and spectroscopy of negative linear carbon cluster ions

In the last few years, we have intensively studied electron detachment and radiative de-excitation processes of negative molecular ions found in the interstellar clouds. We have measured the lifetime of these hot ions stored in a room-temperature ion storage ring in Tokyo Metropolitan University (TMU) by monitoring the time-dependent neutral product yield associated with electron detachment of the negative ions. As a result, we successfully understand the whole dynamics of the delayed process of the isolated molecular ions in vacuum in the time range longer than milliseconds.

These negative molecular ions are generally in the high temperature, namely in the high vibrational states with large inner energy at the moment of their production. Electron detachment of negative ions takes place when they hold the internal energy above the electron affinity. It is known that this process proceeds statistically through the delayed process, and the rate is determined as a function of the internal energy, often termed as thermionic emission; namely, the faster rate for the higher internal energy, and vice versa. Thereby, we can obtain the information of the internal energy by observing the time dependent behavior of the electron detachment of the ions.

Generally, electronically excited states prepared at the ion source or by visible laser irradiation proceed to the vibrationally excited, but electronically ground state through the internal conversion (IC). The cooling of the molecular ions is governed by IR photon emission between these vibrational levels. Our recent experiments clarified that C_5^-, C_7^- and C_6H^- are cooled by this process in addition to electron detachment, i.e. electron depletion. On the other hand, we found that fast cooling process takes place for C_4^- and C_6^- in addition to IR emission. This is “recurrent fluorescence” via the inverse internal conversion (IIC). Namely, the ions once in the vibrationally excited but electronically ground state have a chance to go back again to the ions in the electronically excited state and emit a visible photon (recurrent fluorescence), leading to decrease of the energy to a large extent.

By virtue of a tunable visible OPO laser, we observed the change of the cooling time of C_4^- ions reflecting the initial temperature of the ions, and succeeded in extracting detailed information on the recurrent fluorescence. Thus, we concluded that the delayed processes in energy de-excitation of single molecules are explored by our ion storage ring, although they have been not well understood because of the luck of experimental method,

However, all of these findings of the rapid cooling mechanism were based on observation of the neutral product yields resulting from the electron detachment, which are, in a sense, indirect proofs.
In 2015 we directly observed recurrent fluorescence emitted from stored C_6^- ions by adopting a combination of a photomultiplier and a lens system. We detected 607nm visible lights emitted from the electronically excited state C $^2\Pi_g^+$ state (2.04 eV) by selecting a wave length with a bandpass filter.

To distinguish recurrent fluorescence from normal fluorescence, we measured the emitted light in synchronization with the circulation of the C_6^- ions. We observed that light intensity decayed with a lifetime of several 10ms. It is apparently longer than the case for normal fluorescence. Furthermore, we succeeded in observing delayed recurrent fluorescence not only from hot ions extracted from the ion source but also reheated ions by a 532 nm Nd:YAG pulse laser light.

3 Exploring a new Atomic, Molecular, and Photonic Physics utilizing the photon sources

3.1 Resonant photodetachment of the positronium negative ion (Ps^-)

The positronium negative ion (Ps^-), a bound state of two electrons and a positron with the same mass, is one of the simplest three body systems bound through Coulomb forces. Its dynamics and spectroscopy is a fascinating subject from the viewpoint of fundamental atomic physics. Recently Nagashima et. al. (Science Univ. of Tokyo) found an efficient generation scheme of Ps^- ions by injection of slow positrons onto a Na-adsorbed W surface, and he has opened up the new field of optical measurements of Ps^-.

In 2014 we performed the experiments in collaboration with the Nagashima group at the KEK Slow Positron Facility by using a tunable Dye laser system to observe a shape resonance. We tried a one-photon laser spectroscopy of the Ps^- near the Ps (n=2) threshold. Experiments were performed

utilizing a pulsed slow positron beam (pulse width 10 ns, repetition 50 Hz). Ps^- bunches were generated by injecting the beam onto the Na-adsorbed W target. They were electrostatically accelerated and then vertically crossed with probe light in a field free region. The light source was the dye laser (pulse width 5 ns, repetition 10 Hz) pumped by a Q-S.W. Nd:YAG laser. A probe light with a UV range from 226 nm to 231 nm was generated by the second harmonic generation of the light output. The fragment Ps atoms were detected by a MCP and the yield was measured as a function of the wavelength. We succeeded in observing a clear photodetachment resonance profile of Ps^-. The fragment yield increased from the Ps (n=2) threshold (5.43 eV) and formed a peak.

In 2015 we proceeded in detailed analysis of the obtained data, and we clarified that our experimental results show a good agreement with a theoretical calculation of the shape resonance of ^1P_o symmetry. This is an important milestone in the positron-related atomic physics, because that theoretical calculation of Ps^- is for the first time proved by experiments. Not only a translational-energy tunable Ps beam but also an internal-energy tunable Ps beam is also feasible in this scheme in near future.

3.2 Photoionization of rare gas systems in the intense circularly-polarized photon field

To study atomic and molecular ionization process under the intense photon field, utilizing a time of flight type photoelectron analyzer (e-TOF), we have measured photoelectron energy spectra for Xe, Kr and Ar with circularly-polarized intense 0.8 μm laser fields up to 1.9×10^{15} W/cm^2, where photoelectron originates dominantly from direct tunneling ionization.

As for the ionization under the linearly polarized intense laser light, it is well known that multiple ionization proceeds by the re-collision process where a once-released electron by ionization of a target returns back and collides with a parent target ion, leading to the further ionization. While it has been a common concept that this process does not take place for the circularly polarized light for decades, the validity of this concept is now in a debate.

The fundamental output (800nm) was generated from an amplified Ti:sapphire laser system (pulse width: 25~fs; repetition rate: 1~kHz; circular polarization lights). The laser beam was focused by an *f*=300mm lens. Target gases were introduced to a target vacuum chamber as a supersonic jet, and were crossed with the intense laser pulses. Photoelectrons emitted along the electric vector of the incident light were detected by a MCP under the field free condition. The time difference between the laser pulse (provided by a master clock of the laser system) and the arrival time of the electron was directly recorded by a digital storage oscilloscope or a fast time-to-digital converter (TDC).

In 2015 we measured detailed laser-polarization dependence of electron energy spectrum by changing from circularly polarized light to linearly polarized gradually. Under the intense laser field, a single laser shot gives rise to multiple electrons simultaneously, and it is experimentally crucial to observe all of them without loss. In addition, we discussed an experimental condition for low energy electron where careful magnetic shielding is unavoidable.

Theoretical approach has been under way in collaboration with Dr. Morishita (The University of Electro - Communications) and Dr. Tolstikhin (Kurchatov Institute, Russia). Now theoretical predictions analytically obtained based on the adiabatic approach to compare our results are available.

4. Resonant coherent excitation of fast highly charged ions

Energetic ions passing through a crystal experience periodic oscillating fields by traversing the periodic arrays of atomic planes. If one of the traversing frequency matches the electronic transition energy of the ions, they are resonantly excited: resonant coherent excitation (RCE). High-energy heavy ions enabled RCE in the x-ray energy region. We have been using 100-400MeV/u highly charged heavy ions supplied from the heavy ion accelerator at the national institute of radiological sciences (NIRS), Japan and the GSI heavy ion research institute, Germany. To control of the population in the atomic levels in the x-ray region, we have measured the charge state distribution of the ions passing through a silicon crystal, secondary electron released from the ions, and de-excitation x-rays from the excited ions while we change the angle between the incident ion beam and the crystal.

In particular, three-dimensional RCE (3D-RCE) using the periodicity of the array of the atomic planes opened up a variety of application of RCE techniques. When the ions travel in a crystal at a velocity **v**, the resonance condition for the transition energy E_{trans} is represented by $E_{trans} = h\gamma\mathbf{G}\cdot\mathbf{v}$, where h is Planck's constant, γ is the Lorentz factor and **G** represents the reciprocal lattice vector specifying the corresponding atomic planes. The reciprocal lattice vector **G** is specified by the (*k,l,m*) Miller indices. The resonance condition of 3D-RCE is satisfied by tuning two independent angles of the incident ions with respect to the atomic plane, θ and ϕ, respectively. Tilting the crystal with respect to the ion beam corresponds to scanning the oscillating frequency of the crystal electric field.

4.1 Highly excited state formation of Ar^{17+} ions by ladder-type double resonance

To prove the control of the population in the atomic levels in the x-ray region, we finally confirmed the production of the highly excited state (n=3) of H-like Ar^{17+} by a two-step resonance, that is, the ladder-type double resonance of 3D-RCE after a series of the experiments in the last few years.

In 2015 we performed a density matrix calculation using an original code developed by collaboration with the theoretical group in the Moscow State University to obtain a theoretical support to our measurements. From comparison of the transition amplitudes observed experimentally with theoretical calculation, we explicitly confirmed the production of the n=3 state via double resonance in our experiments. To discuss the x-ray emission probabilities from the highly excited states, we modified the calculation code, and estimated the Kβ X-ray emission yield theoretically. Thus, we distinguish between direct and double excitations to the highly excites states, leading to the more quantitative discussion. In June 2015, we also obtained the additional experimental data to support the data in the previous years.

Generally, excitation and ionization cross sections of high-energetic heavy ions during the collisions in solids have been well investigated, and it is well known that calculation codes like ETACHA or GLOBAL reproduced precisely the evolution of the charge-state distribution in a thin foil often used for charge exchange of the ion beam. On the other hand, as for the cross section of the excited state they differ to a large extent depending on theories, because they are not easily measured experimentally.

We can expect that the ionization cross section of the excited heavy ions state is extracted in high precision using the RCE technique by comparing the experimental data and simulation with density matrix approach by dealing the ionization cross sections as parameters. From this context, we excited H-like Ar^{17+} from the n=1state to the n=2, 3, 4 states using the identical crystal field generated by the same array of the atomic planes, and we observed enhancement of their ionization probabilities of the excited ions in the Si crystal. Through a comparison with simulation results, we are now in the process of evaluating the relative ionization cross section from each state to evaluate the validity of existing theoretical cross section data.

4.2 RCE using an extremely thin crystal

Crystal thickness is an important parameter for RCE, corresponding to the pulse width for short-pulsed lasers. In a similar manner to the energy spectrum broadening for the short-pulsed laser, the resonance broadening of RCE is predicted under a thinner crystal target because of the uncertainty principle. It is also expected to extract the intrinsic information of resonance from its width, like coherence length in the interaction with the crystal field. Previously, we have adopted crystal targets thicker than 1 micron. In 2015 we tried an extremely thin (200 nm thick) Si single crystal membrane (NORCADA Co.), and succeeded in observing the (220) planar channeling and the 1s-2p transition by RCE. The resonance width was broadened about by a factor of two. The origin of this width as well as the crystal quality is now under analysis.