Atomic, Molecular & Optical Physics Laboratory（2019）
Chief Scientist: Toshiyuki Azuma (D.Eng.)

(0) Research field

CPR Subcommittee: Physics

Keywords: ion storage ring, molecular cooling dynamics and spectroscopy, highly-charged heavy ions, resonant coherent excitation, exotic atom

(1) Long-term goal of laboratory and research background

We have explored experimental atomic, molecular, and optical (AMO) physics by taking unique and original approaches covering the wide energy scale from GeV down to meV, and the time region from hour to femto-second. Our research subjects are also full of variety; cold large molecular and cluster ions circulating in an ion storage ring, high-energy highly-charged heavy atomic ions like uranium ions passing through a single crystal, exotic atoms like muonic atom (μ^-, Z^+) and negative positronium (e^+, e^-, e^-). Our long-term goal is to find common and fundamental processes or interaction taking place in such unusual, peculiar or sometimes exotic systems, and to manipulate their dynamics freely.

(2) Current research activities (FY2019) and plan (until Mar. 2025)

(A) Molecular dynamics using an electrostatic ion storage ring

Dynamics of an isolated single molecule in vacuum covering a long-time scale, and very low energy reactions involving cold molecules are unexplored, but full of fascinating subjects in quantum and statistical physics, and they are also keys to understand the evolution of chemical species in space. Taking advantages of electrostatic ion storage rings, which enable us to store large molecular ions for a long time in vacuum, we have been concentrated on observing delayed electron emission process for years and found new electronic radiative cooling process referred to as recurrent fluorescence. In 2019, we observed laser-induced delayed electron detachment from isolated Si_2^- stored in an electrostatic ion storage ring on the ten microseconds time scale. This molecule is homo-diatomic and no rovibrational cooling is expected because of the absence of the dipole moment. We found that the excitation spectra for photon energies near the threshold show well-resolved multi-peak structures, which are attributed to rovibronic transitions to the electronic excited state. This structure only appears in the signal measured with the delay. The occurrence of delayed detachment on such a long timescale is unusual for diatomic molecules. We have shown that the existence of excited electronic states of an anion, which are otherwise overshadowed by the strong direct detachment process, is revealed by measurements of delayed electron emission in an ion storage ring.

To access the cold reactions and cold molecules, we need a specific cryogenic ion storage ring. We had developed a new cryogenic (4K) compact electrostatic ring (RICE), and we succeeded in storing ions for more than 1 hour under the extremely good vacuum condition. In 2019 we observed rotational and vibrational bands of a small molecular ion N_2O^+ to investigate the population evolution of these quantum levels which is governed by the state-selective dynamics. The temporal evolution of the internal state populations of the N_2O^+ ions after being stored in the ring was state-selectively measured by action spectroscopy via a predissociation process using a wavelength-tunable pulsed laser. We found both vibrational cooling within 5~s and unchanged rotational behavior in the same molecule. This sharp unbalance between vibrational and rotational cooling, which has been well known both in theory and experiment, was clearly observed by the combination of long-time storage and high-precision spectroscopy. Furthermore, the cooling dynamics were traced in real-time and revealed the relevant characteristic multi-vibrational-mode dynamics inside. Such dynamics in a simple system like triatomic molecules are essential as a fundamental aspect of understanding non-thermal and non-equilibrium behavior.

Future plan, 1) We will challenge the energy-differential measurement of the low-energy collisions of ion-neutral species by the merging experiments. 2) We are in preparation of cold (0.4 K) molecular ions embedded in a helium droplet for spectroscopy. 3) The detection of neutral product species by a superconducting transition-edge sensor (TES) calorimeter is scheduled for direct identification of neutral products with different mass.

(B) Spectroscopy of exotic atoms; muonic atom and positronium negative ion

A muonic atom is composed of a negative muon and a positive nucleus. Because of the large mass of negative muon compared with electron, the orbital of a negative muon is located very close to, sometimes even inside, the nucleus of a muonic atom. Measurements of muonic X-rays emitted from muonic atoms are thus an ideal probe to explore quantum electromagnetic dynamics (QED) under extremely strong electric fields. QED is one of the most successful theories in Physics, and our measurement is the critical approach to check the validity of the bound state QED. In 2019 we started the high-precision spectroscopy of muonic X-rays from muonic atoms isolated in vacuum at J-PARC, Japan by introducing the superconducting TES calorimeter. We have succeeded in observing 6.3 keV muonic X-rays of the n=5-4 transition of muonic neon (μNe) with the resolution of about 5 eV. This transition includes the 2-5 eV QED effect and no contribution of the nuclear size ambiguity is expected. We determined it with a precision better than 0.3 eV, and the result agreed with the theoretical prediction. This success will provide a new research field to explore the QED effect of high-Z muonic atoms.

We also have been involved in laser spectroscopy of positronium negative ion, Ps^- ($e^+e^-e^-$). It is one of the simplest three-body systems interacting via Coulomb force, and provides an excellent testing ground for the three-body problem. We succeeded in determining the electron affinity of about 0.3 eV i.e., binding energy of Ps^-, and the obtained result with an accuracy within 0.1meV again agreed with the stringent theory of three-body calculations including relativistic effects.

Future plan, 1) The electronic X-rays emitted from muonic atoms provide a rich information of the deexcitation process of muonic atoms associated the electron-hole production with Auger process and electron refilling from the surrounding. We plan to measure these lines intensively. 2) By introducing the new TES calorimeter covering higher X-ray energy, we plan to extend precision spectroscopy of heavier-Z muonic atoms.

(C) Resonant coherent excitation of highly-charged atomic heavy ions using a single crystal

We have been investigating resonant excitation of heavy atomic ions travelling with the energy of several tens GeV using the periodic crystal field instead of the light field, which is called resonant coherent excitation (RCE). This resonant excitation has been established as a new tool for manipulation of heavy ions without using photons in the energy region from EUV to X-rays. In 2019 at HIMAC, Chiba, Japan, we measured an interference effect of the two oscillating electric fields with different polarization and phase in three-dimensional (3D)-RCE. We can select the 3D-RCE conditions from multiple atomic planes simultaneously. We observed the constructive interference for the $1s^2$-1s2p transition of 465 MeV/u He-like Ar^{16+} ions, while the destructive one for 1s-2p transition of 501 MeV/u H-like Fe^{25+} ions. By comparing theoretical simulation we could understand these features depending on their different polarization and phase.

The high-precision spectroscopy of highly-charged very heavy ions in the x-ray energy region is another approach to check QED (quantum electromagnetic dynamics) under the strong field. The RCE technique developed by our group is one of the promising realistic methods. At the GSI/FAIR, Germany, we had already observed the 4.46 keV electronic transition from the $2s_{1/2}$ to $2p_{3/2}$ state of Li-like U^{89+} ions accompanying three electrons. In 2019 we were engaged in upgrading of the experimental device aiming at precise spectroscopy for determining the absolute transition energy.

Future plan, 1) At the GSI/FAIR, we plan to measure the absolute transition energy of 4.46 keV from the $2s_{1/2}$ to $2p_{1/2}$ and $2p_{3/2}$ states of Li-like U^{89+} ions assisted by simultaneous high-precision measurement of the applied voltage to the electrode of the electron cooler at the ion storage ring ESR. 2) The 1s-2p transition of H-like U^{91+} by the RCE technique for testing QED is our main goal at the future facility of FAIR.

(3) Members

(Chief Scientist)
Toshiyuki Azuma
(Senior Research Scientist)
Susumu Kuma
(Research Scientist)
Naoki Kimura
(Contract Researcher)
Shinji Okada
(Special Postdoctoral Researcher)
Preeti Mishra, Yasuhiro Ueno
(Postdoctral Researcher)
Kiattichart Chartkunchand, Takuma Okumura

(4) Representative research achievements

1. "Impact of a hydrophobic molecular ion in the early stage of cloud formation", Linda Feketeová, Paul Bertier, Thibaud Salbaing, Toshiyuki Azuma, Florent Calvo, Bernadette Farizon, Michel Farizon and Tilman D. Märk, **PNAS 116**, (45) 22540-22544 (2019).

2. "Roadmap on photonic, electronic and atomic collision physics: III. Heavy particles: with zero to relativistic speeds", Friedrich Aumayr, Kiyoshi Ueda, Emma Sokell, Stefan Schippers, Hossein Sadeghpour, Frédéric Merkt, Thomas F Gallagher, F Barry Dunning, Paul Scheier, Olof Echt, Tom Kirchner, Stephan Fritzsche, Andrey Surzhykov, Xinwen Ma, Roberto Rivarola, Omar Fojon, Lokesh Tribedi, Emily Lamour, José R Crespo López-Urrutia, Yuri A Litvinov, Vladimir Shabaev, Henrik Cederquist, Henning Zettergren, Marika Schleberger, Richard A Wilhelm, Toshiyuki Azuma, Philippe Boduch, Henning T Schmidt and Thomas Stöhlker, **J. Phys. B52**, 171003 (2019).

3. "Radiative cooling of cationic carbon clusters, C_N^+ , N = 8, 10, 13 and 16", FQ. Chen, N. Kono, R. Suzuki, T. Furukawa, H. Tanuma, P. Ferrari, T. Azuma, J. Matsumoto, H. Shiromaru, V. Zhaunerchyk, K. Hansen, **Phys. Chem. Chem. Phys. 21**, 1587 (2019).

4. "RIKEN cryogenic storage ring RICE", 8th International Workshop on Electrostatic Storage Devices (ESD8), August 26, 2019, Tianjin, China.

5. 「10 GeVからmeVまでのエネルギースケールで原子分子の量子状態を制御し観測する」, 第19 回多元研研究発表会, 2019年12月12日, 東北大学.

Supplementary

Group photo of RIKEN AMO Laboratory

Laboratory Homepage
https://www.riken.jp/en/research/labs/chief/atom_mol_opt_phys/index.html
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