Atomic, Molecular & Optical Physics Laboratory（2021）
Chief Scientist: Toshiyuki Azuma (D.Eng.)

(0) Research fields

CPR Subcommittee**:** Physics

Keywords: ion storage ring, molecular cooling dynamics, molecular spectroscopy, helium droplet, highly-charged heavy ions, muonic atom, resonant coherent excitation

(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 femtosecond. Our research subjects are also full of variety; cold large molecular and cluster ions circulating in an ion storage ring or embedded in helium droplet, high-energy highly-charged heavy atomic ions like uranium ions passing through a single crystal, exotic atoms like muonic atom (μ^-, Z^+). Our long-term goal is to find common and fundamental processes or interactions in unusual, peculiar, or sometimes exotic systems and manipulate their dynamics.

(2) Current research activities (FY2021) and plan (until Mar. 2025)

(A-1) Dynamics of molecular ions stored in an electrostatic ion storage ring

The dynamics of an isolated single molecule in a vacuum covering a long-timescale and very low energy reactions involving cold molecules are unexplored but full of fascinating subjects in quantum and statistical physics. We have investigated deexcitation dynamics for years, taking advantage of electrostatic ion storage rings, which enable us to store large molecular ions for a long time in a vacuum. In FY2021, we challenged to study the dynamics of relatively large molecular anions in addition to small molecular ions, using TMU E-ring. We selected a pentacene anion ($C_{22}H_{14}^-$), which is a prototypical member of the polycyclic aromatic hydrocarbon (PAH) family of molecules. As an important constituent of interstellar clouds, this molecule is of particular interest to the fields of astrophysics and astrochemistry. Spontaneous electron detachment and photo-induced delayed electron detachment by introducing a light pulse from a tunable OPO laser at 890 nm wavelength under merging conditions was measured in the electrostatic ion storage ring. The dissipation mechanisms of the internal energy of the stored hot ions were inferred both from the temporal profile of the spontaneous detachment and from the laser-firing time dependence of the photoinduced temporal profile. Simulations based on statistical emission processes reproduced the observed behaviors, providing the value for the radiative cooling rates in the time range of a few tens of milliseconds. We also performed laser-induced electron detachment spectroscopy of C_2^- ions, produced by a cesium-sputter ion source and stored in the electrostatic ion storage ring RICE, to study highly excited states and their temporal evolution in the longer time region than in TMU E-ring. We found that the overall spectra showed a striking difference from previously reported spectroscopic measurements using other ion source like a laser ablation source. Most of the peaks we observed could not ascribed to doublet–doublet transitions, and expected to be originated in the highly-excited states. We studied the lifetimes of them by varying the ion storage time prior to laser irradiation and discovered shorter- and longer-lived components of 1.5–3 ms and more than 10 ms, respectively. The former lifetime is in good agreement with the autodetachment component, suggesting a possible detachment channel from a metastable quartet state.

(A-2) Dynamics of molecular ions embedded in superfluid helium droplets

Helium droplets are nano-size clusters of a superfluid at a temperature of 0.4 K. They easily capture foreign atomic and molecular species by collision, and their weak perturbation and transparency enable the optical access to the captured species to study the molecular-scale interaction of matter and superfluid. Helium droplets are produced in the form of a molecular beam, where laser spectroscopy and mass spectrometry are applicable. In FY2021, We performed the beam control of charged helium droplets by static electric fields. The electron bombardment was employed as the ionization method. Using a beam deflector at the downstream, we observed the size distribution of charged pure droplets, which is consistent with the distributions of neutral droplets previously observed. This means that the electron ionization is a promising way to generate the charged droplets, enabling the slow dynamics observation of the electrostatically controlled droplet beam. This method will be useful to generate molecular ions inside the droplets and study the ion-superfluid interaction.

Future plan, 1) We will observe lifetime change due to Fermi resonance due to interference of different vibrational modes in triatomic molecular ions like N_2O^+ . 2) Generation of molecular ions embedded in helium droplets by electron impact is now planned. 3) The detection of neutral product species by a superconducting transition-edge sensor (TES) calorimeter is now under development; direct identification of neutral products with different mass is highly expected.

(B-1) Spectroscopy of exotic atoms; highly-charged atomic heavy ion

Hyperfine structures of highly charged ions (HCIs) are favorable spectroscopic targets for exploring fundamental physics along with nuclear properties. Recent proposals of HCI atomic clocks highlight their importance, especially for many-electron HCIs, and they have been theoretically investigated by refining atomic-structure calculations. However, developments in hyperfine spectroscopy of many-electron HCIs have not proceeded due to experimental difficulty. In FY2021 we demonstrated hyperfine-structure-resolved laser spectroscopy of HCIs in an electron beam ion trap plasma, employing the magnetic dipole transition in the 4d95s state of 127 I7+. Ion-state manipulation by controlled electron collisions in the well-defined laboratory plasma enables laser-induced fluorescence spectroscopy of trapped HCIs. The observed spectrum of evaporatively cooled ions under low magnetic fields shows characteristic features reflecting the hyper- fine structures. The present demonstration using combined optical and plasma approaches provides a new benchmark for state-of-the-art atomic calculations of hyperfine structures in many-electron HCIs and offers possibilities for a variety of un- exploited experiments.

(B-2) Spectroscopy of exotic atoms; muonic atom

A muonic atom is composed of a negative muon and a positive nucleus. During FY2019-2020, using the superconducting transition-edge-sensor (TES) detector with an excellent energy resolution (better than 6.0 eV FWHM for the x-ray peak less than 10 keV) combined with the very slow and intense negative muon beam, we successfully observed X-rays from muonic atoms under two different extreme conditions; μNe in a vacuum and μFe close to the surface in metal. In FY2021, we measured electronic characteristic X-rays from μAr isolated in a vacuum. The involved dynamics are categorized as those between the cases of μNe and μFe. Through muon induced Auger process in the muon deexcitation cascade, initially-bound electrons of Ar atoms are stripped off; however, one or a few electrons are expected to remain, which was predicted by the simulation, but no direct experimental evidence has been obtained up to now. We succeeded in observing several electronic characteristic K X-ray peaks in the region from 2.7 to 3.0~keV. Comparison with theoretical calculations indicates that these peaks are ascribed to exotic atomic systems, which consist of one, two, or three electrons with a specific electronic configuration at the *K*- and *L*-shells in addition to the muon and nucleus, i.e., H-, He-, and Li-like μAr. They are very different from the previously-observed broad electronic characteristic X-ray spectra from μFe. Thus, we conclude that we have newly established a powerful approach to studying structures and dynamics of muonic atoms by observing with electronic characteristic x-rays.

Future plan, 1) The precise measurement of muonic x rays from muonic molecular systems like ddμ (d: deuteron) is planned by preparing a solid D_2 target. These x rays are expected to possess rich information on the vibrational state of muonic molecules, which has never been detected before. 2) By introducing the new TES calorimeter covering higher X-ray energy, we plan to perform 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 traveling with the energy of several tens GeV using the periodic crystal field instead of the light field, which is called resonant coherent excitation (RCE). The high-precision spectroscopy of highly-charged heavy ions in the x-ray energy region is another approach to check QED under the strong field. The RCE technique developed by our group is one of the promising realistic methods. At the GSI/FAIR, Germany, we planned 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. In March 2022, we had a beam time for a week. Detectors and measurement systems were transferred from Japan to GSI, and the utmost efforts were made to prepare the setup. Unfortunately, however, due to problems with the accelerator, especially in the final stage of UNILAC, the beam was only available for a few hours and the experiment could not proceed. Once again, beamtime is scheduled for FY2024.

Future plan, 1) At the GSI/FAIR, the experiments are scheduled for FY2024. 2) The 1*s*-2*p* transition of H-like U^{91+} by the RCE technique for testing QED is our primary goal at the future facility of FAIR.

(3) Members

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(Senior Research Scientist)
Susumu Kuma

(Research Scientist)
Naoki Kimura

(Postdoctoral Researcher)
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(4) Representative research achievements

1. S. Iida, S. Kuma, M. Kuriyama, T. Furukawa, M. Kusunoki, H. Tanuma, K. Hansen, H. Shiromaru, and T. Azuma, "IR-photon quenching of delayed electron detachment from hot pentacene anions", **Phys. Rev. A** 104, 043114 (2021) . *selected as Editors' Suggestions.*
2. Hatsuki Otani, Hiroko Nakahara, Haruka Goto, Susumu Kuma, and Takamasa Momose, **"**Electronic spectroscopy of Mg-phthalocyanine embedded in cold hydrogen clusters produced by a pulsed nozzle**",** Hatsuki Otani, Hiroko Nakahara, Haruka Goto, Susumu Kuma, and Takamasa Momose, **J. Chem. Phys.** 155, 044309 (2021).
3. T. Okumura, T. Azuma, D. A. Bennett, P. Caradonna, H. I. Chiu, W. B. Doriese, M. S. Durkin, J. W. Fowler, J. D. Gard, T. Hashimoto, R. Hayakawa, G. C. Hilton, Y. Ichinohe, P. Indelicato, T. Isobe, S. Kanda, D. Kato, M. Katsuragawa, N. Kawamura, Y. Kino, K. Mine, Y. Miyake, K.M. Morgan, K. Ninomiya, H. Noda, G. C. O'Neil, S. Okada, K. Okutsu, T. Osawa, N. Paul, C. D. Reintsema, D. R. Schmidt, K. Shimomura, P. Strasser, H. Suda, D. S. Swetz, T. Takahashi, S. Takeda, S. Takeshita, H. Tatsuno, X. M. Tong, Y. Ueno, J. N. Ullom, S. Watanabe, S. Yamada, "De-excitation dynamics of muonic atoms revealed by high precision spectroscopy of electronic K X-rays using a superconducting TES detector", **Phys. Rev. Lett.** 127, 053001(2021). *selected as Editors' Suggestions*.
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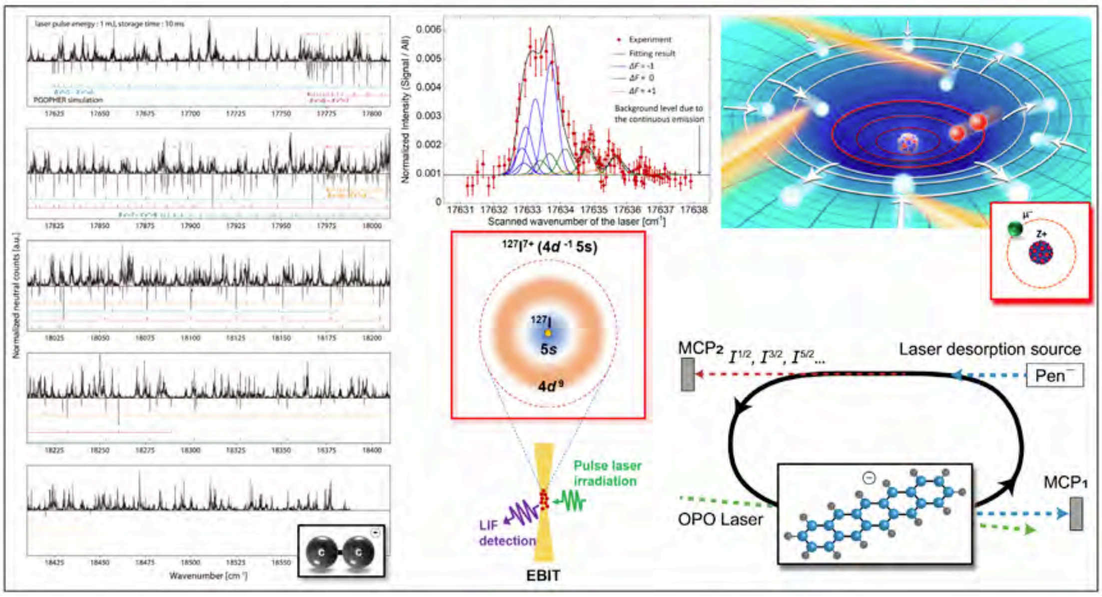
5. T. Azuma, “De-excitation is exciting”, 36th Symposium on Chemical Kinetics and Dynamics, Web-conference, Jun. 3, 2021.
6. N. Kimura, "Laser spectroscopy of forbidden transitions between metastable excited states of I^{7+} in an electron beam ion trap", The 32nd International Conference on Photonic, Electronic and Atomic Collisions (ViCPEAC 2021), Web-conference, Jul.22, 2021.

Supplementary



Overview of activities of RIKEN AMO Laboratory in FY2021

Laboratory Homepage

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