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Developments at CERN-ISOLDE's OFFLINE 2 mass separator facility for studies of molecular ion beams

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ABSTRACT

ISOLDE's Offline 2 laboratory has been upgraded to facilitate development for the production and study of molecular ion beams. New gas injection systems have been implemented for both molecular formation in the ion source and in the radio-frequency quadrupole ion trap used for beam preparation. MagneToF detectors and time-resolved single ion counting data acquisition have been implemented for low intensity beams and studies of laser-atom or laser-molecule interactions. We present a study of the formation and ionization of BaF^+ using the upgraded facility.

1. Introduction

Molecular beams are a growing focus of research [1–3], particularly at radioactive ion beam (RIB) facilities. Systematic studies of molecular formation, extraction, and ionization are required before new techniques can be applied to online RIB facilities such as CERN-ISOLDE [4].

For molecules with low dissociation energies or molecules that are not stable at typical target and ion source temperatures (2000 °C), molecule ionization attempts may lead to dissociation instead of ionization. An alternative method of molecule formation is to induce chemical reactions with a mass-separated ion beam and a chosen reactant in an ion trap. CERN's ISOLDE offline 2 mass separator facility (YOL2) has been upgraded to enable in-source and in-trap molecular formation. Single ion counting and time-resolved data-taking capabilities have been added.

2. Facility upgrades

YOL2 features a Frontend which delivers services (power, cooling, gas) to operate an ISOLDE-compatible target and ion source unit. The YOL2 beamline includes ion optics, a mass-separator dipole magnet, and a radio-frequency-quadrupole cooler buncher (RFQ-cb) [5]. We present the addition of gas mixing and injection systems for the Frontend and the RFQ-cb. MagneToF detectors (ETP MagneToF 14925 [6]) have been added for single-ion counting and time-of-flight (ToF) measurements. A schematic of the upgrades is shown in Fig. 1.

The Frontend gas system allows the mixing of two different gases by partial pressures before injection into the target and ion source unit through a calibrated gas leak. The RFQ gas system allows injection of trace amounts of gas from a gas sample into a mixing reservoir before filling the reservoir with the helium typically used as buffer gas. The first MagneToF detector is installed in the beamline after mass separation. A drift section has been installed after ejection from the RFQ-cb, allowing longer flight path for ToF measurements or studies of bunched beams at MagneToF 2. For time-resolved data taking, ions on the MagneToF detectors are recorded with 500 ps resolution using a Cronologic TimeTagger [7].
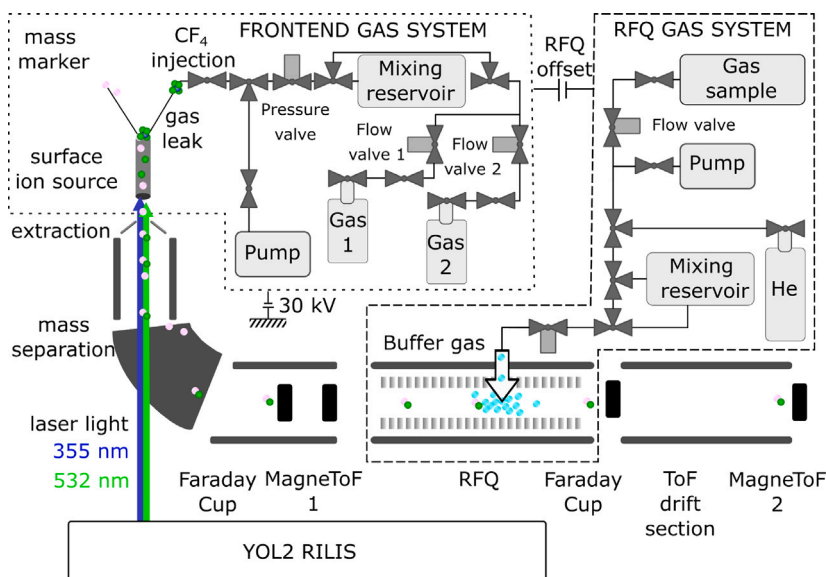
Fig. 1. Schematic of the YOL2 laboratory featuring two new gas systems, MagneToF detectors, and a ToF section. Reactants of interest are supplied from a mass marker and a gas leak. Ions are extracted as a beam from the ion source by a potential of 30 kV and separated by m/q in the separator magnet. Mass-separated beams are sent to MagneToF 1 or into the RFQ-cb, where the ions are cooled and bunched. Ion bunches can be studied on MagneToF 2 after a drift section for ToF measurements.
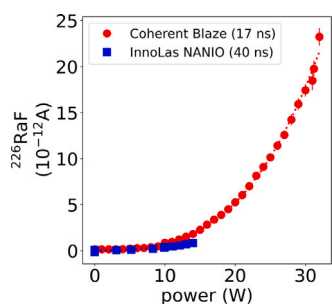

Fig. 2. Intensity of ^{226}RaF$^+$ ion beam on a Faraday Cup in response to 532 nm laser power measured before transport to the ion source for lasers of two different pulse lengths at a repetition rate of 10 kHz. Third-order polynomial fits are shown.

Table 1
Possible mechanisms of interaction between high-power laser light in the ion source and molecules, characterized by the observable dependence of ion beam intensity as a function of laser power and wavelength. See text for more details.

Laser-related effect	Laser power	Laser wavelength
Heating	Linear	None
Ablation	Threshold	Negligible
Dissociation	Linear	Threshold
Ionization	Quadratic or cubic	Threshold or resonant

3. Non-resonant laser effects in molecular beams

The ion source type most commonly employed at Isotope Separation On-Line (ISOL) facilities is a Resonance Ionization Laser Ion Source (RILIS), which uses laser light to step-wise resonantly excite an electron of the species of interest into the continuum, providing an efficient element-selective method of ionization [8,9]. For molecules, with rotational and vibrational degrees of freedom in addition to electronic states, the population may be distributed over a wide variety of states due to the high temperature environment in the target and ion source (2000 °C). In an online study, we observed dependence on laser pulse length and a third-order polynomial effect of laser power for the intensity of the radioactive molecular ion RaF$^+$ (Fig. 2). These observed dependencies motivated the systematic offline study of non-resonant laser effects using BaF$^+$ beams at YOL2.

The anticipated laser-related effects on molecular-ion production are summarized in Table 1 along with their expected power and wavelength dependencies. Each mechanism of light-molecule interaction is expected to depend differently on the total number of photons (laser power) and the energy of each photon (laser wavelength). In the case of dissociation, a linear dependence is expected for molecular bond strength less than the photon energy, and a quadratic dependence is expected otherwise. In the case of ionization, threshold steps would be in units of the ionization potential divided by the photon energy.

BaF$_x$ molecules were formed in the ion source from reactive gas injection. Excess barium was supplied as a Ba(NO$_3$)$_2$ standard solution deposited into a tantalum tube (mass marker). CF$_4$ gas was injected through a calibrated leak using the front end gas system (Fig. 1). Measurements were done with ion source heating power of 400 ± 10 W corresponding to 1700 °C, which is lower than the 2000 °C typically used in hot-cavity ion source operation. Conditions were chosen to limit the ion rate at the single-ion detector and therefore avoid saturation.

Mass scans were conducted using the YOL2 mass separator magnet with and without frequency-doubled (532 nm, 2.33 eV) and tripled (355 nm, 3.49 eV) light from an Innoslab EdgeWave Nd:YAG laser beam delivered to the ion source (Fig. 3). Ta, Ba, and BaF show increased intensity with the 355 nm light.

For Ba, with an ionization potential (IP) of 5.2 eV, and BaF (IP 4.7 eV) in a tantalum surface ion source (work function 4.3 eV), the surface ionization efficiency at the corresponding ion source temperature is expected to have an approximately linear dependence on power.

Ba$^+$ intensity was recorded while changing the power of 355 nm and 532 nm laser light in the ion source, as shown in Fig. 4. The bond dissociation energy of BaF into Ba+F has been measured as 5.90(44) eV, and BaF$_2$ into BaF+F as 6.07(44) eV [10] such that ionization is favoured over dissociation. The quadratic trends in Fig. 4 show two-photon ionization for 355 nm light and a linear trend indicating heating for 532 nm light.

TaF$_{1-2}^+$ and TaOF$_{0-2}^+$ show an increase in intensity and a laser pulse-related time structure (Fig. 5). ^{181}Ta$^+$ and ^{181}Ta$_2^+$ fragments from dissociation of TaF$_4^+$ and TaF$_5^+$ were observed and are indicated at their corresponding effective mass where identified in Fig. 3.
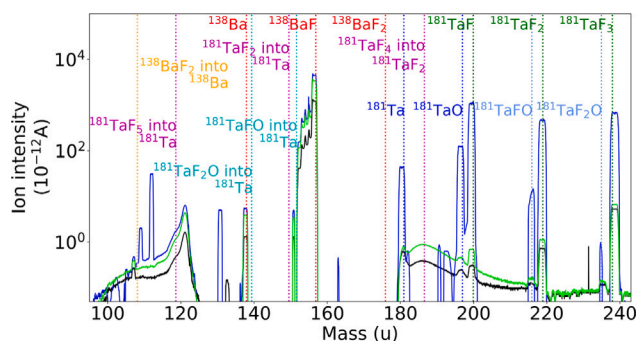
Fig. 3. A mass scan of the YOL2 mass separator magnet with 12 W of 355 nm (blue) and 532 nm (green) light in the ion source compared to the same mass scan with the laser off (black). Positions of singly-charged atomic, molecular and fragment species are shown with annotated vertical lines.
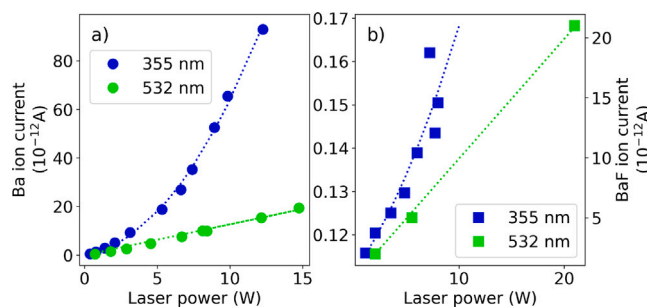

Fig. 4. The difference in laser-on and laser-off ion intensities for mass-separated beams of (a) $^{138}Ba^+$ and (b) $^{138}BaF^+$ as a function of laser power for 355 nm and 532 nm light. A second-order polynomial fit is used for 355 nm data and a linear fit is used for 532 nm data. In (b), the left vertical axis gives the value for 355 nm and the right vertical axis gives the value for 532 nm.

Fig. 5. Time spectra of mass-separated ion beams with respect to the laser trigger using 10 W of 355 nm laser light at a repetition rate of 10 kHz. The separator magnet set mass and the average number of ions per laser pulse ("bunch") are indicated on the left. Vertical axes indicate the number of ions incident on the detector per laser pulse per 100 ns ToF bin in logarithmic scale.

Species that are ionized in the extraction region between the 30 kV-ion source potential and the laboratory ground experience a fraction of the full extraction potential. Ions created in this region therefore have a different energy and are thus mass-separated with a magnetic field corresponding to this fraction of the ion source potential, resulting in peaks with narrow spread in time-of-flight as demonstrated by Heinke et al. in [11]. They then appear in the mass spectrum as broad, asymmetrical features tailing towards lower masses. In the time-domain, these ions, which are extracted immediately after creation, are therefore the first to appear at the ion detector following the laser pulse, and the bunch width is narrower than that of the ions created inside the hot cavity. The laser effect on tantalum molecules seen in Fig. 3 is observed for these extraction-ionized species on a different mass as shown in Fig. 5, indicating that some fraction are ionized outside of the ion source. Ablation can also contribute to the observed increase in ion intensity if it directly results in ions or if it increases the number of neutrals available for laser ionization.

4. Conclusions

The YOL2 facility has been upgraded to enable molecular formation with mixtures of two added gases in the ion source and with a reactive gas added to the buffer gas in the RFQ-cb. In combination with the capability to form molecules, the addition of single ion counting and time-resolved data taking enables studies of laser-related time structures and laser interaction with stable atoms and molecules as demonstrated using Ba and BaF.

High-power laser light (10s of W or more of average power) can be used for ionization of molecules in the ion source and subsequent delivery of molecular beams. The laser deposits power and can additionally increase surface ionization efficiency with an effect dependent
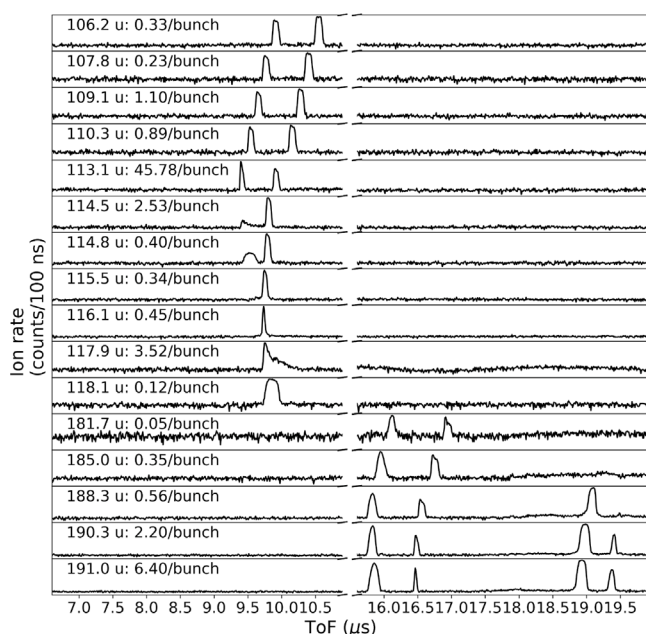
on the ionization potential of the species. In cases where ionization occurs out of the ion source in the extraction potential, a sharp time structure related to the laser pulses is visible at mass-separating fields lower than that of the expected mass as a result of mass separation at a different ion energy.

Declaration of competing interest

Acknowledgements

References

[1] G. Arrowsmith-Kron, M. Athanasakis-Kaklamanakis, M. Au, J. Ballof, R. Berger, A. Borschevsky, A.A. Breier, F. Buchinger, D. Budker, L. Caldwell, C. Charles, N. Dattani, R.P. de Groote, D. DeMille, T. Dickel, J. Dobaczewski, C.E. Düllmann, E. Eliav, J. Engel, M. Fan, V. Flambaum, K.T. Flanagan, A. Gaiser, R.G. Ruiz, K. Gaul, T.F. Giesen, J. Ginges, A. Gottberg, G. Gwinner, R. Heinke, S. Hoekstra, J.D. Holt, N.R. Hutzler, A. Jayich, J. Karthein, K.G. Leach, K. Madison, S. Malbrunot-Ettenauer, T. Miyagi, I.D. Moore, S. Moroch, P. Navrátil, W. Nazarewicz, G. Neyens, E. Norrgard, N. Nusgart, L.F. Pašteka, A.N. Petrov, W. Plass, R.A. Ready, M.P. Reiter, M. Reponen, S. Rothe, M. Safronova, C. Scheidenberger, A. Shindler, J.T. Singh, L.V. Skripnikov, A.V. Titov, S.-M. Udrescu, S.G. Wilkins, X. Yang, Opportunities for fundamental physics research with radioactive molecules, 2023, http://dx.doi.org/10.48550/ARXIV.2302.02165, URL https://arxiv.org/abs/2302.02165.

[2] M.S. Safronova, D. Budker, D. Demille, D.F. Kimball, A. Derevianko, C.W. Clark, Search for new physics with atoms and molecules, Rev. Modern Phys. 90 (2) (2018) http://dx.doi.org/10.1103/RevModPhys.90.025008, arXiv:1710.01833.

[3] L.V. Skripnikov, N.S. Mosyagin, A.V. Titov, V.V. Flambaum, Actinide and lanthanide molecules to search for strong CP-violation, Phys. Chem. Chem. Phys. 22 (33) (2020) 18374–18380, http://dx.doi.org/10.1039/d0cp01989e, arXiv:2003.10885.

[4] R. Catherall, W. Andreazza, M. Breitenfeldt, A. Dorsival, G.J. Focker, T.P. Gharsa, T.J. Giles, J.L. Grenard, F. Locci, P. Martins, S. Marzari, J. Schipper, A. Shornikov, T. Stora, The ISOLDE facility, J. Phys. G: Nucl. Part. Phys. 44 (9) (2017) http://dx.doi.org/10.1088/1361-6471/aa7eba.

[5] S. Warren, T. Giles, C.M. Pequeno, Offline 2, ISOLDE 's target, laser and beams development facility, Nucl. Instrum. Methods Phys. Res. B 463 (July 2019) (2020) 115–118, http://dx.doi.org/10.1016/j.nimb.2019.07.016.

[6] D. Stresau, K. Hunter, W. Sheils, P. Raffin, Y. Benari, A new class of robust sub-nanosecond TOF detectors with high dynamic range, in: 54th ASMS Conference on Mass Spectroscopy, Seattle, Washington, 2006, URL https://www.etp-ms.com/faqs/literature.

[7] Cronologic, TimeTagger product brief, 2022, https://download.cronologic.de/TimeTagger/TimeTagger4-product-brief.pdf. (Accessed 20 April 2022).

[8] V. Fedosseev, K. Chrysalidis, T.D. Goodacre, B. Marsh, S. Rothe, C. Seiffert, K. Wendt, Ion beam production and study of radioactive isotopes with the laser ion source at ISOLDE, J. Phys. G: Nucl. Part. Phys. 44 (8) (2017) http://dx.doi.org/10.1088/1361-6471/aa78e0.

[9] S. Rothe, B.A. Marsh, C. Mattolat, V.N. Fedosseev, K. Wendt, A complementary laser system for ISOLDE RILIS, J. Phys. Conf. Ser. 312 (SECTION 5) (2011) 1–6, http://dx.doi.org/10.1088/1742-6596/312/5/052020.

[10] National Bureau of Standards, Bond dissociation energies in simple molecules, in: National Standard Reference Data System, Vol. NSRDS-NBS 31, NIST, 1970.

[11] R. Heinke, V. Fedosseev, T. Kieck, T. Kron, B. Marsh, S. Raeder, S. Richter, S. Rothe, K. Wendt, Atom beam emersion from hot cavity laser ion sources, Nucl. Instrum. Methods Phys. Res. B 463 (March 2019) (2020) 449–454, http://dx.doi.org/10.1016/j.nimb.2019.04.026.