उच्च आवेशित आयन किरणपुंजों का उपयोग करते हुए आयन-परमाणु टकराव

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N_2O के आयनीकरण प्रेरित विघटन गतिकी

एच. सिंह, जे. मुखर्जी, के. कुमार, मो. ए.के.ए. सिद्दीकी, एम. दास, और. डी. मिश्रा
टाटा मूलभूत अनुसंधान संस्थान, होमी भाभा रोड, मुंबई-400005, भारत

की आण्विक संरचना।

सारांश

उच्च वेग व्यवस्था (v > 1 a.u.) में N_2O के आयनीकरण प्रेरित क्षय गतिकी का अध्ययन करने के लिए एक नया COLTRIMS (कोल्ड टारगेट रिकॉइल आयन मोमेंटम स्पेक्ट्रोमीटर) विकसित किया गया। यह प्रयोग 30 MeV पर F^{9+} आयन किरण का उपयोग करके किया गया। N_2O परा-ध्वनिक गैस जेट के साथ अंतःक्रिया पर, ZN_2O का बहु-गुना आयनीकरण होता है। यह कुलम्ब विस्फोट के कारण अणु के वियोजन की ओर ले जाता है। वियोजन चैनल N_2O^{3+} N^++N^+O^+ विश्लेषण किया और गतिज ऊर्जा निस्सरण (KER) वितरण की गणना की गई। खंडों के बीच गति संबंध का भी अध्ययन किया गया है। तब गतिशीलता की तुलना कम वेग (v <1 a.u.) पर पहले से मौजूद डेटा ईसीआरआईए (इलेक्ट्रॉन साइक्लोट्रॉन अनुनाद आयन त्वरक) से की गई और यह समझने की कोशिश की गई कि अणुओं के वियोजन गतिकी में प्रक्षेप्य के वेग की भूमिका क्या रहती है। एक प्रतिक्रिया सूक्ष्मदर्शी भी अभिकल्पित की गई।

Ion–Atom Collisions Using Highly Charged Ion Beams

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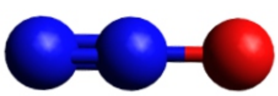
Ionization Induced Dissociation Dynamics of N_2O

H. Singh*, J. Mukherjee*, K. Kumar , Md A.K.A. Siddiki, M. Das, and D. Misra*
Tata Institute of Fundamental Research, Homi Bhabha Road, Mumbai-400005, INDIA



Molecular Structure of N_2O

ABSTRACT

We have developed a new COLTRIMS (Cold Target Recoil Ion Momentum Spectrometer) to study the ionization induced decay dynamics of N_2O in the high velocity regime (v > 1 a.u.). The experiment was performed using F^{9+} ion beam at 30 MeV. On interaction with the N_2O supersonic gas jet, multi-fold ionization of N_2O takes place. This leads to the dissociation of the molecule due to the coulomb explosion. We have analysed the dissociation channel $N_2O^{3+} \rightarrow N^+ + N^+ + O^+$ and calculated the Kinetic Energy Release (KER) distribution. Momentum correlation between the fragments has also been studied. We have then compared the dynamics with the already existing data at low velocity (v < 1 au.) from the ECRIA (Electron Cyclotron Resonance Ion Accelerator) and tried to understand how velocity of projectile plays a role in the dissociation dynamics of molecules. We have also designed a reaction microscope.

KEYWORDS: *COLTRIMS, Ion beam, Coulomb ionization, Kinetic energy release, Dissociation dynamics*

**Authors for Correspondence: H. Singh, J. Mukherjee, and D. Misra*
E-mail: harpreet.singh@tifr.res.in, jibak.mukherjee@tifr.res.in, and dmisra@tifr.res.in

Introduction

The ionization of atoms or molecules using Highly charged ions (HCI) is of importance for fundamental research as well as their applications in molecular physics and biology to study radiation-induced damage. In molecules, the complexity increases being a many-body system. On interaction with the highly charged projectile, elastic and inelastic processes can take place such as electron capture, transfer excitation, ionization, etc. In the high-velocity regime (v > 1 a.u.) Ionization dominates compared to other processes [1]. In atoms, interaction with HCI may lead to excitation or electron removal from outer shells or core shells. In the case of core-shell excitation or ionization, the atom relaxes either by radiative decay or auger decay. In the case of molecules, the ionization usually leads to dissociation due to Coulomb repulsion.

However, in the case of polyatomic molecules, after interaction we may get multiply ionized molecular ions which are rather unstable and tend to dissociate into constituent ions due to the Coulomb force coming into action (Coulomb Explosion). There can be many dissociation pathways such as two-body, three body etc. All the bonds can break simultaneously (concerted decay) or they can break sequentially (Sequential Decay). The interaction time scale is of the order of a hundred attoseconds which is small compared to the rotational and vibrational timescale due to which the nuclear motion freezes during the interaction. This leads to the excitation of the molecule in the Frank Cordon region. Also, since the axial-recoil approximation is valid, one can obtain the angular distributions of the fragment ions.

Experimental Setup

The experiment employed a novel momentum imaging technique known as COLTRIMS (Cold Target Recoil Ion Momentum Spectroscopy) [2]. F^{9+} ion projectile beam was provided by the PELLETRON-LINAC FACILITY at 30 MeV energy (v = 7.98 a.u.) The projectile beam was made to interact orthogonally with the supersonically cooled target jet of N_2O gas. The jet was produced by the isentropic expansion of gas through a 30 μm into the source chamber. The center portion of the jet is extracted with the help of a 400 μm skimmer in the skimmer chamber and further collimated with a 500 μm skimmer. The pressure in the main chamber at the time of the experiment was of the order of 10^{-7} mbar. After the interaction the ionized molecular ion fragments and the recoil ions are detected with the help of two-stage Wiley-McLaren type spectrometer [3]. The schematic of COLTRIMS is shown in Fig.1(a) and the actual image of the setup is shown in Fig. 1(b). The length of the extraction, acceleration and drift region are 15mm, 90mm and 520mm respectively. The field in the extraction and acceleration region was set to 173.33 and 250.67 V/cm, respectively which ensures 4π sr collection efficiency for singly charged ion with energy up to 13 eV. After the drift region the recoil ions are detected with the help of a position sensitive MCP (Micro Channel Plate-DLD (Delay Line Anode) detector.

The electron produced from the ionization of the molecules is detected with the help a CEM (Channel Electron Multiplier) which gives the start signal and the last recoil ion coming from the fragmentation gives the stop signal to complete the detection of all ions produced in a single event. Despite that, ions from other events may also come. To filter that out we have a momentum sum gate of about 15 a.u. as all the ions produced in a single event will follow momentum conservation. The momentum for the recoil ions can be calculated from the hit position x, y and the time of flight T. CoboldPC Software was used for the data acquisition and each event information is stored in a lmf (list mode file) format which can be later used for offline analysis.

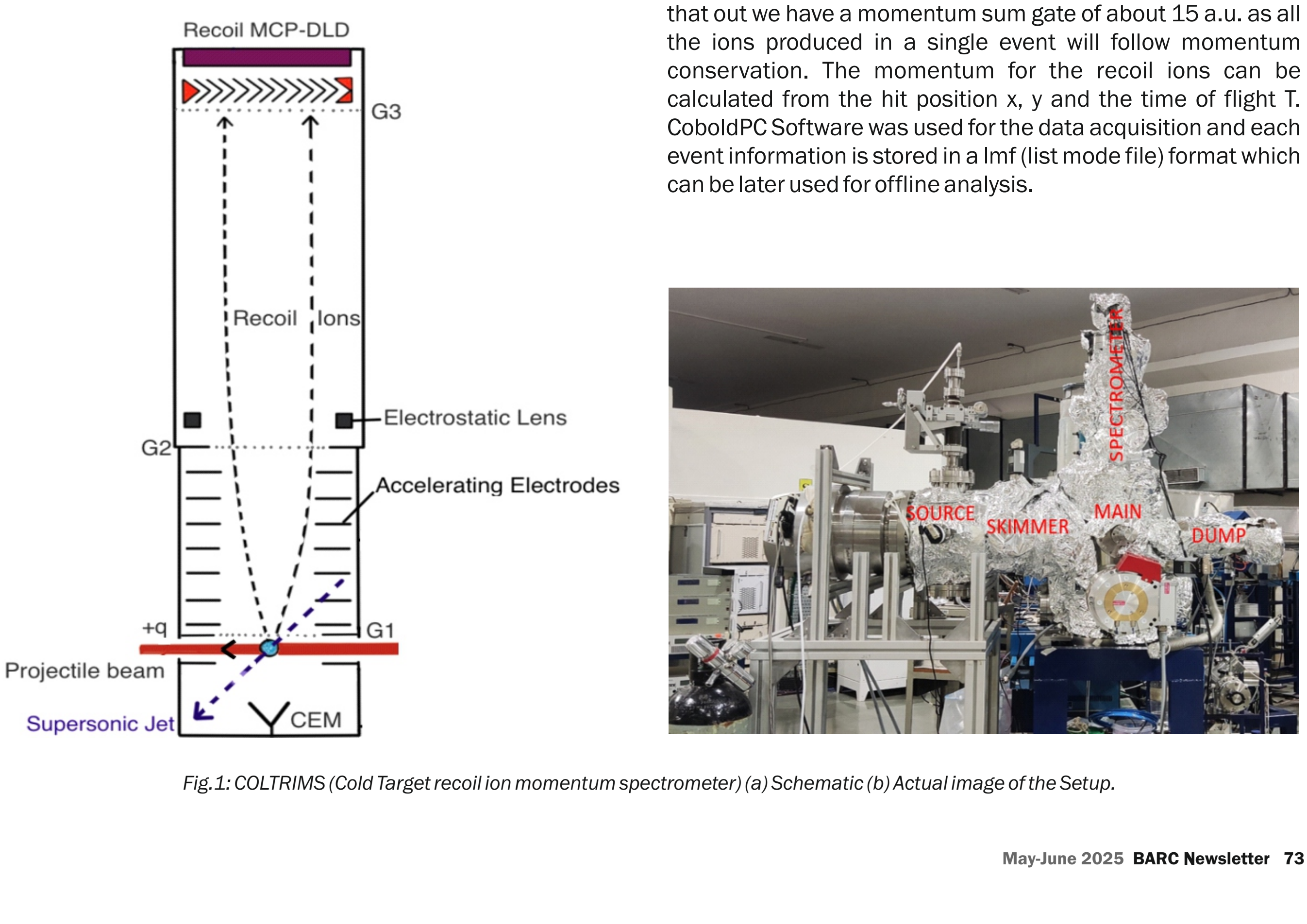


Fig.1: COLTRIMS (Cold Target recoil ion momentum spectrometer) (a) Schematic (b) Actual image of the Setup.

Results

TOF (Time of Flight) Spectrum

Time of flight spectrum in Fig.2 shows the various recoil ions recorded by the detector. This TOF corresponds to the first ion in the event recorded. We can see some prominent peaks such as N_2O^+, N^+_2, N^+, O^+ and, NO^+ which are characteristic of the target N_2O. Except for N_2O^+ all other ions are coming due to the Coulomb explosion of the parent ion. The time of flight separates each ion on the basis of the m/q ratio. There can be many possible channels of dissociation for two-body or three body dissociation depending on the charge state of the molecular ion. To filter out a particular channel of interest we plot the TOF coincidence maps which are Time of flight of first ion vs second ion and Time of flight of second ion vs third ion. We then select the region corresponding to channel of our interest which filters the ions coming from the single event.

KER (Kinetic Energy Release)

From the momentum we can calculate the KER which is the sum of the Kinetic energies of all the fragments. We have compared the KER for the two different projectile: F^{9+} at v = 7.98 a.u from the PLF and Ar^{8+} at v = 0.44 a.u from ECRIA. The KER peaks at 23 and 24 eV for F^{9+} and Ar^{8+}, respectively which is similar. From the KER we also get an idea about the states being populated as it is difference between the frank-cordon excitation point and the dissociation limit. High velocity projectile on interaction may lead to more population in the higher excited states thereby giving more intensity for higher KER values. This is slightly visible in KER range (30-40 eV) in Fig.2.

Comparison

We have done the analysis for Ar^{8+} at v = 0.44 a.u from ECRIA with the same target gas N_2O and tried to compare the two results. The results only differ in the statistics as the effective data obtained in case of Pelletron experiment was less, but in terms of the dynamics it is mostly similar. We have already seen the KER comparison and the peak value is about same though we expected the counts for higher KER values to increase. Similarly, the branching ratio for the sequential and concerted we got in case of Ar^{8+} was 15.2:84.8. Again, we expected the ratio to be less in case of F^{9+} as the higher excited states are mostly repulsive leading to a concerted breakup.

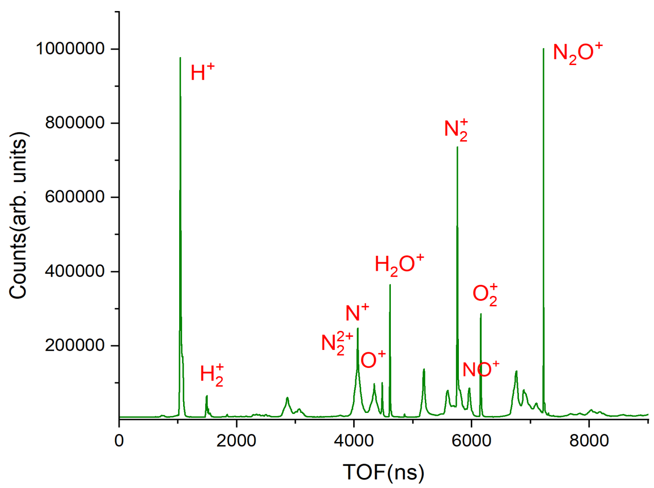


Fig.2: Time of Flight (TOF) Spectrum for first ion in event.

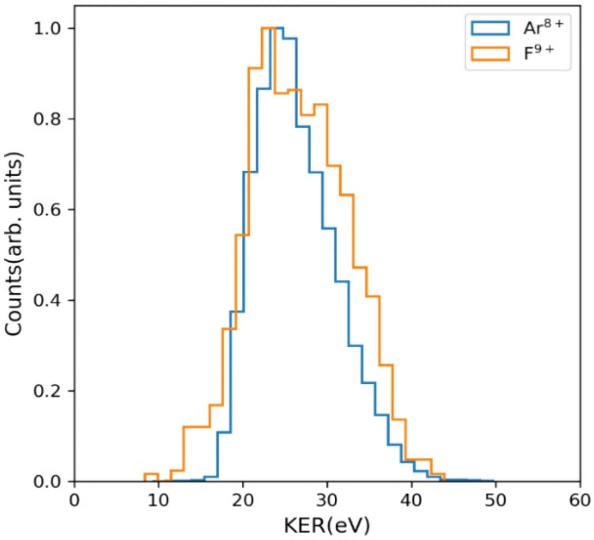


Fig.3: KER (Kinetic Energy Release) distribution.

Future Plan

Development of new COLTRIMS reaction microscope setup for kinematically complete ion atom/molecule collision studies

We have designed a new COLTRIMS reaction microscope [5] which is capable of measuring momentum vector of individual reaction products, the recoil ion and the electron, with subatomic scale precision. This will enable us to perform kinematically complete experiments involving ion atom collision. Once we measure the momentum vector of individual ions we can deduce quantities like kinetic energy, electron emission angle, orientation of molecule before fragmentation, etc.

Design of the spectrometer

The spectrometer consists of three regions. The extraction region for extraction of reaction products, the recoil ion and electrons. The field free drift region of the recoil ion to separate time of flight in m/q. The deceleration region for electron to achieve time focusing. The region between drift tube and extraction region will act as electrostatic lens for time and space focusing of the recoil ion. The extraction region has length 99 mm. The drift tube part has length 221 mm. The deceleration region of electron has length 60 mm. For confinement of electron within 80 mm diameter helical path, an external uniform magnetic field will be applied by a pair of Helmholtz coils.

Cold target: Cold target will be prepared by supersonic expansion through micro meter size nozzle. A geometrically cooled supersonic molecular jet will be formed by passing the beam through two skimmers.

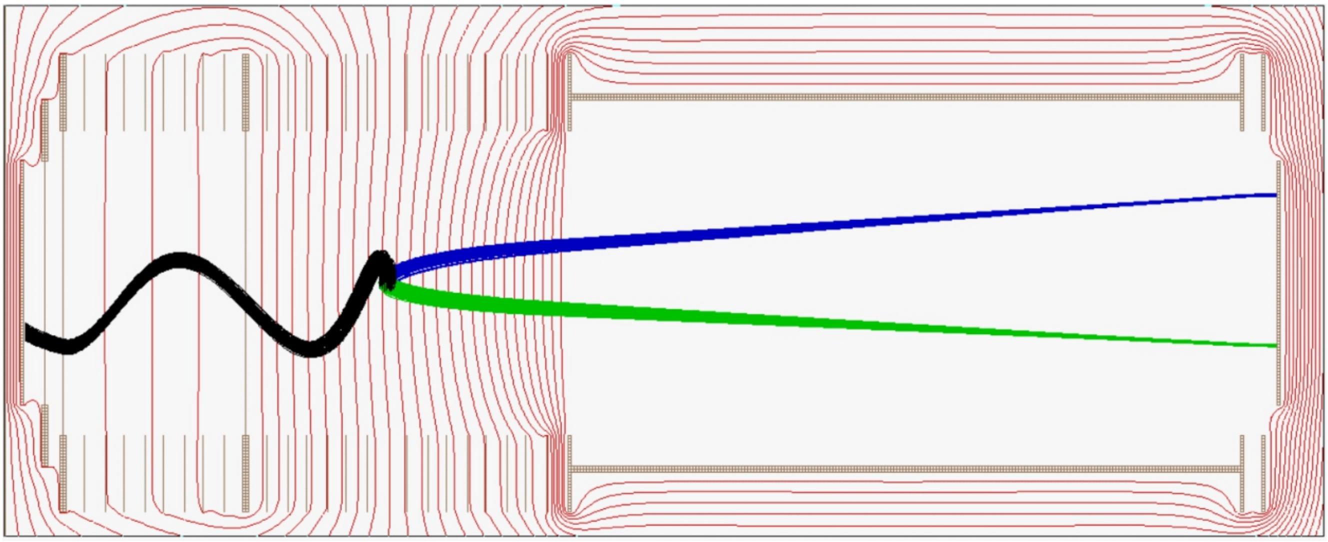


Fig.4: Design of the COLTRIMS reaction microscope. The equipotential surfaces are shown in red curves. Recoil on trajectories are shown in blue and green color. Electron trajectory is shown in black colour. The left most region is the deceleration region for electron, the middle part is extraction region for recoil ion and electron, the rightmost part is the field free drift region for recoil ion. Magnetic field lines are not show in this figure.

Simion simulation for momentum component of the recoil ion and the electron: We will record hit coordinates in the respective recoil ion and electron detectors and time of hitting as raw data for every event. From the hit position and time of flight we will calculate the momentum components.

The transverse momentum component of the recoil ion follows linear relationship with hit radius and the parallel component follows linear relationship with time of flight if kinetic energy gain during electrostatic extraction if much higher than initial energy of the recoil ion. The transverse component of the electron follows $p_{e\perp} = r_0 \sin(\omega t/2)$, where $\omega = B/m_e$ is the angular velocity in applied magnetic field. Since electron initial energy will be comparable to energy gain during extraction, parallel momentum component will follow a nonlinear relationship with time of flight.

Some possible Experiments with the Reaction Microscope

Fully differential cross section measurement of ionization followed by fragmentation of diatomic molecules

In earlier studies people have observed double slit interference pattern in electron emission from H_2 [6], N_2 [7], O_2 [8] upon heavy ion collision. They have measured double differential cross section (DDCS) of electron. But in those studies, information about fragmentation of parent diatomic molecules are missing. With the reaction microscope by measuring all reaction products momentum vectors we can study the orientational effect during collision. Measurement of Kinetic Energy Release (KER) spectrum and orientation during fragmentation will reveal complete information about ionization processes.

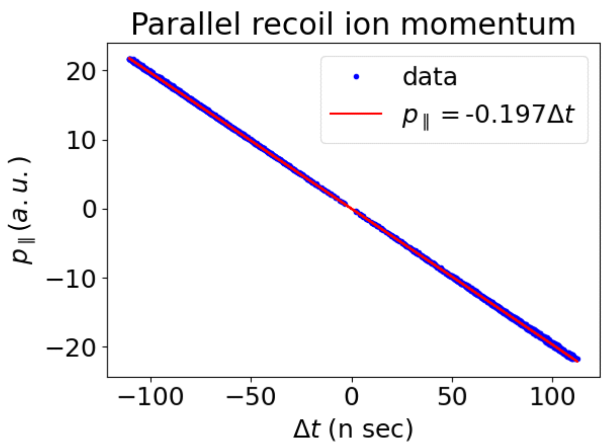


Fig.5: The recoil ion momentum parallel to spectrometer axis mapping function with time of flight. Here $\Delta t = t - t_0$, where t_0 is time of flight for zero momentum component.

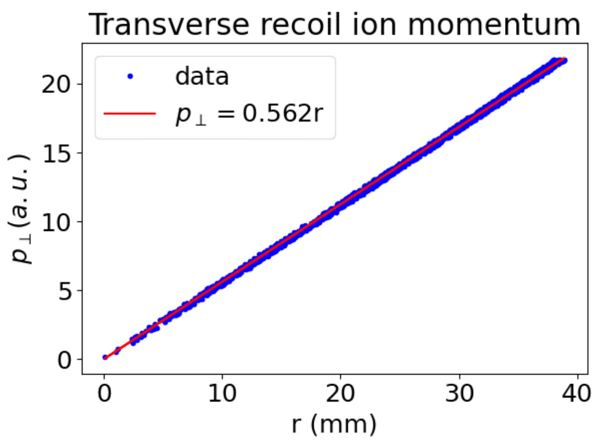


Fig.6: The recoil ion momentum component transverse to spectrometer axis mapping function with hit radius in the recoil ion detector.

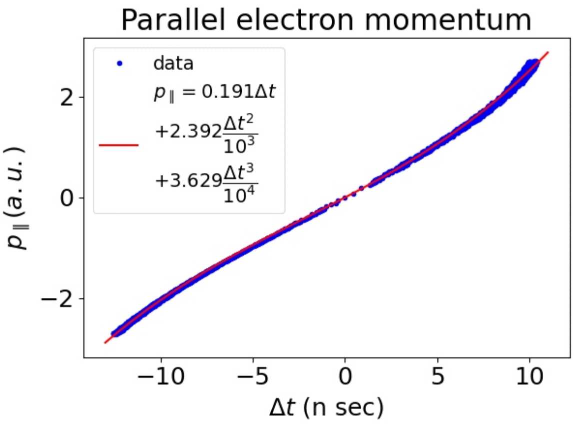


Fig.7: The electron momentum component parallel to the spectrometer axis mapping function with time of flight of the electron. Here $\Delta t = t - t_0$, where t_0 is time of flight for zero momentum component.

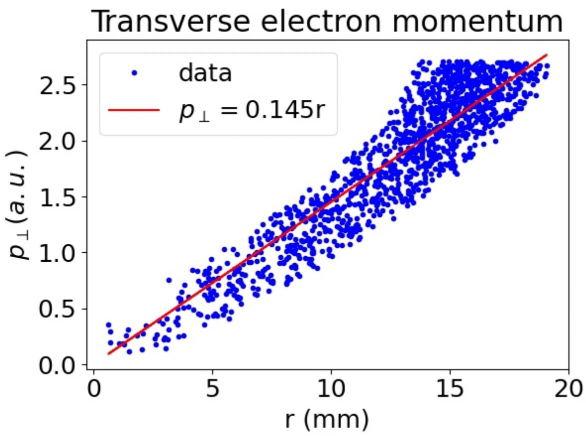


Fig.8: The electron momentum component transverse to spectrometer axis mapping function with hit radius at the electron detector.

Fragmentation dynamics of dimer

Measuring all momentum vectors of the fragmented dimer atoms or molecules we can measure kinetic energy release (KER) and the orientation during collision. Also measuring momentum vector of ejected electrons, we can distinguish between different phenomenon like Coulombic explosion (CE), radiative charge transfer (RCT), inter molecular Coulombic decay (ICD). As we know, charge transfer occurs differently in weakly bound system than molecules with strong bonding [5], [6], [7]. It is important to know role of the neighbor atom/molecule in energy transfer, electron emission. This helps us in understanding fundamental laws of nature as well as help in modelling radiation damage. Thus, it is important to study interaction of this kind of system.

Conclusion

We have developed a new COLTRIMS Setup to study ionization induced dissociation dynamics of N_2O on interaction with F^{9+} in the Pelletron LINAC Facility. We have studied the KER distribution coming the breakup of the ionized molecule. Using momentum Correlation plots we have tried to understand the different types of decay and found the concerted mechanism to be the dominant one. Finally, we have compared these dynamics at high velocity to a low velocity projectile. We have found the dynamics mostly remains the same. We have designed a new reaction microscope for complete kinematic study of molecular fragmentation. The simulation has been done and will be tested in the future.

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