The goals of this part of the JRML program are to study the different mechanisms leading to molecular dissociation and charge exchange following fast collisions, slow collisions, or interactions with an intense short laser pulse.

Molecular dissociation imaging of collision-induced dissociation and dissociative capture in slow $\text{H}_2^+$ + Ar (He) collisions, D. Hathiramani, I. Ben-Itzhak, J.W. Maseberg, A.M. Sayler, M.A. Smith, and K.D. Carnes. The dissociation of hydrogen molecular ions following a slow collision (keV) is studied by 3D momentum imaging of the fragments. The two main processes at this collision energy, collision-induced dissociation (CID, e.g. $\text{H}_2^+$ + Ar $\rightarrow$ $\text{H}^+$ + $\text{H}$ + Ar) and dissociative capture (DC, e.g. $\text{H}_2^+$ + Ar $\rightarrow$ $\text{H}$ + $\text{H}$ + Ar$^+$), are experimentally separated in the method we recently developed. Using an electric field in the target region followed by a field free region, we managed to separate the CID from the DC in time. Thus, it is possible to evaluate the relative importance of these two processes, because both CID and DC are measured simultaneously. The same is true for the ratio of the two possible CID channels for heteronuclear molecules, i.e. $\text{A}^+$+$\text{B}$ or $\text{A}$+$\text{B}^+$. Moreover, this method allows one to distinguish experimentally between two different mechanisms of CID as shown in the figure, and thus study each one of them in detail. The two mechanisms are (i) CID caused by an electronic excitation to a repulsive state, and (ii) CID caused by a vibrational/rotational excitation. These mechanisms differ in the kinetic energy release upon dissociation (KER) and the momentum transfer to the projectile (labeled as $P_{\perp}$ in the figure). For the first the KER is relatively large while the momentum transfer to the projectile is very small. In contrast the latter is associated with small KER and very large momentum transfer. This distinguishes the present work from previous studies [1], in which no such separation was possible.

The electronic excitation to a repulsive state (mostly the first excited state of $\text{H}_2^+$) is the dominant CID mechanism for 3 keV $\text{H}_2^+$ + Ar collisions. The angular dependence of this CID mechanism indicates that the dissociating fragments align along the beam direction for “short” molecular ions and perpendicular to the beam for “long” ones, as predicted by Green and Peek [2].
The vibrational/rotational dissociation mechanism shows very strong alignment effects, as shown in the figure. First, molecular ions aligned perpendicular to the beam velocity (i.e. ? = 90°) are much more likely to dissociate by this mechanism. Second, the dissociation velocity is preferentially aligned along the momentum transfer (i.e. a = 0°, where a is the angle between the dissociation velocity in the x-y plane and the direction of the transverse momentum transfer). The angular distribution in ? is much narrower than in a, as indicated by the much higher power of the \( \cos^n \) fit to the data. Further analysis of the DC and CID processes as well as model calculations of the vibrational CID mechanism are underway.

Recently we have conducted studies of CID of HD\(^+\) in similar collisions in search of the isotopic effects observed previously for this molecular ion [see, for example, Refs. 3,4], namely, that the dissociation into \( H + D^+ \) was favored over \( H^+ + D \). Preliminary results indicate that such effects, if they exist, are much smaller than those reported in previous work. These results will be presented as an invited talk in the CAARI 2002 meeting.

**Future plans:** Systematic studies of DC and CID caused by either electronic or vibrational excitation will be conducted focusing on the effect of the target species and the collision energy. Our preliminary results indicate significant differences in vibrational CID between Ar and He targets. Furthermore, we plan to investigate both these processes for a few additional simple molecular ions, such as HeH\(^+\), He\(_2^+\) and H\(_3^+\). While conducting these studies on the existing system, an improved setup, which includes a cold jet target, will be assembled and tested. This will enable the simultaneous measurement of the recoil ion momentum for DC reactions, thus providing kinematically complete information about the process.


**Isotopic effects and asymmetries in bond-rearrangement and bond-breaking processes in water ionized by fast proton impact.** A.M. Sayler, Z.S. Casey, J.W. Maseberg, D. Hathiramani, K.D. Carnes, B.D. Esry, and I. Ben-Itzhak. Studies of ionization and fragmentation of water molecules by fast protons and highly charged ions have revealed an interesting isotopic preference for H-H bond rearrangement. Specifically, the dissociation of H\(_2\)O\(^+\)\(\rightarrow\)H\(_2^+\)+O is about twice as likely as D\(_2\)O\(^+\)\(\rightarrow\)D\(_2^+\)+O, with HDO\(^+\)\(\rightarrow\)HD\(^+\)+O in between. Further investigations of this isotopic effect lead us to discover a similar isotopic effect following double ionization of water,
i.e. in H$_2$O$^2+$→H$_2^++$O$^+$, although these results are preliminary. In addition, we have observed large asymmetries in bond breaking in the HDO isotope. For example, HDO$^+→$H$^+$+OD is more likely than HDO$^+→$D$^+$+OH by about a factor of 1.5, suggesting that it is easier to break the O-H bond than the O-D bond. This preference is even larger in the dissociation of HDO$^{2+}$. Calculations are underway to determine the relative production rates for the different isotopes from the overlap of the initial and final vibrational wave functions and the time evolution of the final wave function. In addition, we determined the relative cross sections of all dissociation channels including H$^+$ + H$^+$ + O which typically are not measured by TOF techniques. These results were presented by Max Sayler as an invited talk in the undergraduate research session of DAMOP 2002.

**Future plans:** The isotopic enhancement in the H$_2$O$^{2+}→$H$_2^++$O$^+$ dissociative double ionization channel requires further investigation to determine if it is similar in magnitude to that found in single ionization. We also intend to study ionization and fragmentation of water by fast highly charged ions and compare them to fast proton impact.

**High intensity laser interactions**

1. **Evidence for pondermotive-gradient field-ionization in an intense focused laser beam,** E. Wells (UVA), I. Ben-Itzhak, and R.R. Jones (UVA). We have measured the Rydberg ion population produced during intense laser ionization in Xe, Kr, and Ar. The branching ratio for production of Rydberg ions, A$^{(q-1)+*}$, to ions A$^q+$ has been measured as a function of laser intensity and polarization. Using 100 fs, 790 nm laser pulses, singly and double charged Rydberg ions are observed in Xe, but in Kr and Ar only singly charged Rydberg ions are seen. Model calculations of ionization by the gradient of the pondermotive potential (∇U$_p$) of the focused laser beam suggest that the Rydberg population is inversely proportional to the ionization potential of the ion species. These calculations are in agreement with our data except for the Xe$^{2+*}$/Xe$^{3+}$ ratio, which is anomalously large, approximately a factor of two higher than the Xe$^+$/Xe$^{2+}$ ratio.

2. **Photo ionization and photo dissociation of H$_2^+$ by an intense short pulse laser,** I. Ben-Itzhak, J.W. Maseberg, A.M. Sayler, M.A. Smith K.D. Carnes, Z. Chang, C. Fehrenbach, and C.L. Cocke. We have recently begun measurements of ionization and dissociation of a few keV H$_2^+$ beam crossed by an intense short-pulse laser beam using 3D molecular dissociation imaging on a newly installed apparatus employing techniques similar to those described in 3.1. Only a couple of previous measurements have been reported so far [1,2].

**Future plans:** We plan to measure the dependence of ionization and dissociation of H$_2^+$, and other simple molecular ions, on the duration of the intense laser pulse. It has been predicted that the ratio of ionization to dissociation will change with increasing number of laser cycles, i.e. pulse width [3].

Formation and decay mechanisms of $^4\text{He}_2^{2+}$ dimers, I. Ben-Itzhak, and A. Bar-David, I. Gertner, and B. Rosner (Technion). We have measured the decay rate and kinetic energy release upon dissociation of $^4\text{He}_2^{2+}$ dimers formed in $\text{He}_2^{2+} + \text{Ar}$ charge stripping collisions. The measured mean lifetime indicates that $\text{He}_2^{2+}$ molecular ions are preferentially formed with high angular momentum, i.e. high $l$ values, as suggested by our previous measurements of $^3\text{He}^+\text{He}^{2+}$. For the $^4\text{He}_2^{2+}$ isotope low $l$-states as well as high $l$-states are within the high sensitivity range of our experimental setup.

Publications:

1. "A comparative study of the ground state dissociation of $\text{H}_2^+$ and $\text{D}_2^+$ induced by ionizing and electron capture collisions with $\text{He}^+$ at velocities of 0.25 and 0.5 a.u.", W. Wolff, I. Ben-Itzhak, H.E. Wolf, C.L. Cocke, M.A. Abdallah, and M. Stöckli, Phys. Rev. A 65, 042710 (2002).