Apparatus Modification Simulations for the Genesis Project

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Abstract

The Genesis Project, conducted by the Savin Group of Columbia University out of the Astrophysics labs at Nevis, aims to gain a better understanding of the origins of organic chemistry in the cosmos. A novel merged beams apparatus is used to measure cross section data for the reaction of neutral C with H$_3^+$ to form CH$_2^+$ and H$_2$ - the cross section measurements can be used to derive thermal rate coefficients for this reaction for a wide range of temperatures. Possible future modifications to the apparatus include a linear ion trap to store cold H$_3^+$ molecules for interaction, and the results from the simulation of that ion trap are shown here. In addition, to study reactions with deuterium based (instead of hydrogen based) chemistry, the apparatus’ chicane will have to be modified, and the modifications and simulations for that updated chicane will also be shown here.

Introduction

Interstellar molecular gas-phase chemistry is believed to be responsible for the production of the first organic molecules in the universe. Molecular clouds act as chemical factories, which have mostly hydrogen and helium (and, in much smaller quantities, heavier atoms like carbon, oxygen, nitrogen, iron, etc.) as their raw materials, and produce a host of different molecules. Molecular clouds are low density (10 - 10$^3$ particles cm$^{-3}$)$^{[1]}$, so the collisions between molecules can be assumed to be binary, however the low temperatures of molecular clouds (10-300 K) all but rule out neutral-neutral chemistry, as the molecules’ thermal energies are too low to overcome reaction activation barriers. For reactions to take place at these temperatures, neutrals must be polarized by ions. Though molecular clouds are generally low density, they can be large enough that UV light cannot penetrate deep enough to ionize atoms/molecules - cosmic rays however, have high enough energy to ionize neutral atoms and molecules in even large molecular clouds.

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There are two reactions pathways available in which ionized hydrogen atoms/molecules contribute to the formation of CH$^+$. (1) H$^+$ (H$_2^+$) + CO $\rightarrow$ C$^+$ + OH (H$_2$O) followed by C$^+$ + H$_2$ $\rightarrow$ CH$^+$ + H or C$^+$ + H$_2$ $\rightarrow$ CH$_2^+$. This reaction pathway has been well studied at molecular cloud temperatures. (2) H$^+$ (H$_2^+$) + H$_2$ $\rightarrow$ H$_3^+$ (H) followed by H$_3^+$ + C $\rightarrow$ CH$^+$ + H$_2$ or H$_3^+$ + C $\rightarrow$ CH$_2^+$ + H. There is very little research on this molecular pathway at molecular cloud temperatures. This experiment focuses in particular on the proton transfer process C + H$_3^+$ $\rightarrow$ CH$^+$ + H$_2$. From CH$^+$, there is only one reaction with molecular hydrogen gas making CH$_3^+$, and the possible molecular network radiates out to a sprawling tree of precursors to organic molecules and hydrocarbons (Figure 1).

![Chemical reaction network leading to the formation of precursors to organic molecules and hydrocarbons](image)

Fig. 1.— Chemical reaction network leading to the formation of precursors to organic molecules and hydrocarbons

Previous research has studied the reaction C + H$_3^+$ $\rightarrow$ CH$_3^+$ + H$_2$. A classical calculation of the rate coefficient for this reaction, using Langevin theory, has predicted a constant rate coefficient across all relevant temperatures, however two different semi-classical calculations of the rate coefficient, using different quantum-mechanical potential energy surfaces, show temperature dependence in the temperature range 10-300 K $^{[2]}$, $^{[3]}$. Fully quantum-mechanical calculations for this reaction are outside of the range of computation at the moment. Measurement on an analogous reaction (C + D$_3^+$ $\rightarrow$ CD$_3^+$ + D$_2$) have been performed using heated graphite rods that effuse a gas of C$_n$ in excited states in to an ion trap containing D$_3^+$, however these measurement are at an effective temperature of 3000 K, and one temperature only, and there is debate as to how to convert rate coefficients between isotopologues of a reaction; some say there is no difference, while others say the rate coefficient scales by the

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$^4$ Savić, I., Cerm´ak, I., & Gerlich, D. 2005, Int. J. Mass Spectrom., 240, 139
square root of the ratio of the reduced masses. Regardless, theory and experiment have not converged significantly on either the magnitude or temperature dependence of this reaction in any temperature range relevant to molecular clouds. A novel merged fast beams apparatus is needed to study this reaction at relevant effective temperatures with C in its ground-term, and the apparatus design and built by the Savin group accomplishes this.

**Apparatus**

*Carbon Source*

The apparatus begins with a carbon *sputter source* - the source works by ionizing cesium vapor between electrodes that accelerate and focus the ions toward a target of pulverized and compacted graphite. Carbon anions of many kinds are ejected from the surface of the target and accelerated down the beam line by a potential; the velocity of the ions at the exit of the sputter source is proportional to the square root of their charge to mass ratio.

*Ion Optics*

After the beam exits the source it is shaped and steered using *XY steerers, Einzel lenses* and apertures. The XY steerers are two horizontally and vertically aligned, oppositely charged electrodes in series that can be used to steer the beam in the respective dimensions. The Einzel lens is a series of three cylindrical voltage plates, with the middle plate biased to a different potential to the other plates; these plates act as a radially symmetric lens that can either focus or defocus the ion beam depending on the needs of the the experiment. The beam then travels through a *Wien filter* which is a velocity selector that works by balancing the forces from orthogonal electric and magnetic fields; selecting the velocity of the ions flowing through allows the Wien filter to select for the C- specifically from the rest of the beam.

*Beam Profile Monitor*

After more steering, the beam flies through a *Beam Profile Monitor (BPM)*, which rotates a thin metal wire through the beam path in both the x and y directions. When the wire rotates through the beam, electrons are expelled from the wire and collected by a electron collection shroud. By analyzing the current from the collection shroud with respect to the rotation of the wire, an indirect profiles of the beam can be examined (Figure 2, 3).

![Fig 2.— Diagram of a Beam Profile Monitor (BPM). Seeley et al., 2008.](image1)

![Fig. 3.— Profile reading from a BPM signal. Seeley et al., 2008](image2)
The beam is deflected around a 90° bend by a cylindrical deflector, and more ion optics before it enters the photodetachment region.

**Photodetachment Region**

The photodetachment region contains a floating cell, biased to a potential which speeds up or slows down the ions as they flow toward the center of the cell, and returns them to their original velocity when they exit. A 1.8 kW laser at 808-nm (1.57-eV) is directed so as to intercept the beam in the center of the floating cell. The laser photodetaches carbon anions into ground term triplet-P neutral carbon (C(3P)) with ~4% efficiency. Biasing the floating cell to different potentials allows for variation in the energy of the neutral carbon beam (Figure 4).

![Photodetachment Region](image)

**Fig. 4.— Variable Potential $U_f$ in the floating cell allows for fine-tuning C beam energy**

What remains of the C⁻ beam is diverted out of the beam line by voltage plates immediately following the photodetachment region.

**Hydrogen Source**

The source for the H₃⁺ is a duoplasmatron source with works by heating a cathode filament which ejects electrons and ionizes hydrogen gas, forming a plasma. Hydrogen ions of many kinds, positive and negative, are formed, and the positive ions are accelerated out of the source with a potential in a similar fashion to the sputter source. It should be noted that the ions coming out of this source have internal energies on the order of thousands of Kelvin, but this will be discussed in a later section. The H₃⁺ is selected for using the Wien filter, and the beam is shaped and its profile characterized using the same ion optics and BPMs as the carbon beam line. The end of the hydrogen beam line, at least as it’s own separate entity, is the merger, which steers the H₃⁺ beam in to the C beam, so they co-propagate.
Interaction Region

The interaction region begins and ends with a BPM to measure the profiles of the two beams, and determine their overlap factor. The C beam energy is adjusted to adjust the relative energy between the two beams. The beams can be compared to fast cars on a highway - though they are both moving very fast, but their relative velocities (and therefor relative energies) are low if they are moving at close to the same velocity. At the exit of the interaction region is the chicane.

Chicane

“Chicane” is a racing term for a segment of road containing an extra turn, or bend. The chicane in this apparatus uses four sets of oppositely charged electrodes to send the CH$^+$ beam “around a bend” - sending it out of, and then back in to the beam path, with the C beam (Figure 5). H$_3^+$ is much lighter than CH$^+$ ($M_{CH^+}/M_{H3^+} = ~4.3$), so the H$_3^+$ beam is sent in to a faraday cup above the second set of electrodes instead of around the bend with the CH$^+$ beam.

After the chicane, the neutral carbon beam is sent in to a faraday cup at the end of the beam line, and the CH$^+$ beam is sent in to the detection region.

Detection Region

The CH$^+$ beam is sent through two cylindrical deflectors, one bending the beam 90° straight upwards, and the other bending it 90° back parallel to the beam bath, and in to a Channel Electron Multiplier (CEM) which measures the signal.

Chopping

To isolate the signal from the CH$^+$ from background and the noise from the other beams, a method called chopping is used. There are 4 beam configurations possible from turning the C
and H$_3^+$ beams off and on, and the noise can be isolated by adding and subtracting the signals from each in a clever way. (R1) - when both the C beam and H$_3^+$ beams flow through the interaction region the signal from the CH$^+$ is present, along with the noise form the C beam, H$_3^+$ beam, and background are present. (R2) - the noise from the C beam (along with background) can be isolated by chopping the H$_3^+$ beam off (deflecting the H$_3^+$ beam from the beam path with the voltage plate before the interaction region). (R3) - the noise from the H$_3^+$ beam (and background) can be isolated by chopping the C beam (turning off the laser, and therefore production of neutral C). (R4) - by chopping both beams off, the background noise can be isolated. Adding the signals R1 and R4 and subtracting the sum of the signals R2 and R3, the background noise is canceled, along with the individually isolated C and H$_3^+$ noise signals, and the CH$^+$ signal is all that is left.

Table 1.— the signal (S), present in the configuration highlighted in green (R$_1$), is isolated by subtracting the isolated noise from the C beam (R$_2$), H$_3^+$ beam (R$_3$), and background (R$_4$) - present in all.

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$S = (R$

After the signal is isolated, the signal, S (after some transmission efficiencies, T, are accounted for), the velocities of the neutral and ion beams, $v_n$ and $v_i$ respectively, the beam currents of the neutral and ion beams, $I_n$ and $I_i$ respectively, the length of the interaction region, L, and the overlap factor $\langle\Omega(z)\rangle$ - so notated because it is calculated by assuming the overlap between the beginning and the end of the interaction region is the average overlap at the ends - can be used to determine the merged beams rate coefficient - the cross section, $\sigma$, times the velocity, $v$, averaged over the velocity spread of the experiment - can be

$$\langle\sigma v\rangle_v = \left(\frac{S}{T}\right) \left(\frac{e^2 v_n v_i}{I_n I_i}\right) \left(\frac{1}{L \langle\Omega(z)\rangle}\right)$$

Fig. 6.— Formula for the merged beams rate coefficient, calculated using the signal (S), transmission efficiencies (T), elementary charge (e), velocity of the neutral and ion beams ($v_n$, $v_i$), beam current of the ion and neutral beams ($I_i$, $I_n$), length of the interaction region (L) and overlap factor ($\langle\Omega(z)\rangle$).
These results can be manipulated, with knowledge of the relative velocities of the beam, and the velocity spread of the experiment, to generate the cross section for the reaction. This cross section, when convolved with a Maxwell-Boltzmann distribution (the distribution most likely for molecular clouds) will produce a thermal rate coefficient for this reaction as a function of temperature.

**My Contribution**

*Lab Work*

During my time in the lab I assisted in the cleaning and maintenance of a scroll pump, the alignment of the beam path, the maintenance of the laser, the maintenance of the source cooling systems, the removal, examination, and reattachment of many of the segments of the apparatus, the collection of data, and the identification of possible sources of noise in the apparatus.

*Ion Trap Simulation*

In its current state, the apparatus uses hot H$_3^+$, with an internal temperature on the order of 5000 K. Clearly this is not ideal if we wish for our results to be analogous to the chemistry of molecular clouds. The H$_3^+$ gas can be cooled using a pulsed ion source system that uses adiabatic expansion to cool the gas as it exist the source. However, as its name suggest, this source is pulsed, and a continuous beam is required for useful interaction data.

The pulsed ion source can be used if an ion trap is utilized. Storage ring ion traps are not desirable for this experiment as the interaction region with the linear neutral C beam would be all but negligible. A linear ion trap is needed. Once the hydrogen is trapped in the linear ion trap, the neutral carbon beam can be sent through the middle of the trap to interact with the H$_3^+$ beam, and the product of the reaction can be detected down the beam path. I assisted in the simulation and modification of an existing design for a linear ion trap - a cylindrically-symmetric cross section of which is shown below (Figure 7).

![Ion Trap Cross Section](image)

**Fig. 7.— Cylindrically symmetric linear ion trap cross section**

The ions would float in from the left, at which point the electrodes on the right side would be brought up to voltage so they repel the ion in the opposite direction. The trap acts like an optical resonator, or a concave mirror focusing the beam to the midpoint of the trap. As the ions are sent back to the left, the left set of electrodes would be brought up to voltage so that the
ions are trapped between the two electrode “mirrors”. The grounds surrounding the electrode ensure that there is minimal potential acting on the beam in the middle portion of the trap. This trap could be timed so that it can trap multiple pulses from the ion source at a time, as the time for the electrodes to be brought up to voltage and brought back down is far lower than the time-length of the pulsed source ion-bundles.

It is of interest whether or not the ion trap would have any effect on the profile of the beam. Using the simulation program SIMION™, data was recorded for 10 simulated particles using a circular-profile beam with a gaussian energy distribution at a mean of 7.05 keV and a FWHM of 10 eV (predicted energy spread for the beam) and coulomb repulsion, and the position of the particles was recorded every time any of them passed through the center of the trap. Scatter plots were made to show the position in the particles in the plane perpendicular to the direction of travel and get an idea of the profile of the beam. Histograms of the particles per unit area for these profiles made using bins of radial annuli scaled by their respective relative areas to get an idea of the radial distribution of the beam from the center of the beam line were also plotted. The results of these beam profile measurements will be shown in the next section (Figures 8, 9, 10, 11). There is a slight but significant focusing effect of the beam, but this effect may only be as noticeable as it is in these plots because they were made using data taken in the center of the trap, where the electrode “mirrors” focus the beam.

It is also of interest what percentage of the particles will have the same energy and velocity as they came in with, as the energy of the C beam will still be fine-tuned by the potential in the floating cell to vary the relative energy between the beams. Thankfully, the cross section for the reaction is very small when the beams are traveling in the opposite direction, as the relative energy of the beams is much higher. However, a significant portion of the beam will still have energy less than it’s initial energy as it is slowed by the electrodes in the two end caps. For this reason it was necessary to take data of the particles at every time step in the simulation (using the same set of simulated ions as before) and calculate a time-averaged energy probability distribution for the particles over a period of 10 µs (Figure 12). The distribution shows roughly 80% of the time the beam has an energy of 7.03 keV or higher (in the range of the initial energy distribution), meaning that 40% of the time the particles have an energy and velocity in the desired direction, but this is sufficient for the needs of the experiment - more importantly, only 20% of the time will the particles have energies in the ranges between 0 and 7.07 keV, where the cross section for the reaction varies widely with relative energy.
Chicane Modification

Future experiments with the apparatus may study reaction with deuterium instead of hydrogen - i.e. $\text{C} + \text{D}_3^+ \rightarrow \text{CD}^+ + \text{D}_2$. Deuterium is heavier than hydrogen, and the chicane for the CH$^+$ reaction only works due to the high ratio between the mass of the CH$^+$ beam and the H$_3^+$ beam (~4.3). The ratio of the mass of CD$^+$ to D$_3^+$ is about 2.3, and simulation of the chicane suggest that it (in its current form) would not be able to separate the D$_3^+$ beam from the CD$^+$ beam (Figure 13) - the electric force from the first electrode no longer provides significantly different acceleration between the two ion beams to separate them so that one beam flows in to the faraday cup, and the other stays below the top electrode of the second electrode pair. To work around this problem, I removed the upper electrode from the second electrode pair, and moved the second and third electrodes upward ~2 cm, so that the D$_3^+$ beam could be sent up in to the faraday cup, and the CD$^+$ beam could be steered back in to the beam path (Figure 14). Simulations suggest that this solution would work with minimal effect to the velocity and energy of the beam. Simulations for the chicane were also performed using SIMION™.

Simulations (especially those in which data was recorded at every time step) were quite time-intensive (simulations of 10 particles over 1 msec taking around over an hour and a half) and generated massive files of data. To handle input files of this size, Matplotlib/Pyplots software packages were use (plotting tools using the Python language and environment).
Fig. 8.— Beam profile from ion beam simulation of 10 particles over 658 μsec. Point recorded every time simulated particle passes through y-z-plane at x = 500 mm in ion trap 1000 mm units in length. Ion flight begins in positive x-direction with randomly selected starting points in circular beam profile radius r = 2.5 mm.
Fig. 9.— Beam profile from ion beam simulation of 10 particles from 4687 to 5463 μsec. Point recorded every time simulated particle passes through y-z-plane at x = 500 mm in ion trap 1000 mm units in length. Ion flight begins in positive x-direction with randomly selected starting points in circular beam profile radius $r = 2.5$ mm.
Fig. 10.— Ion trap beam profile radial distribution from 0 to 658 μsec. Bins are annuli of equal width, starting with circle in center and moving outward. Bins weighted by area to give particles per unit area. Plot generated with Matplotlib/Pyplots.
Fig. 11.— Ion trap beam profile radial distribution from 0 to 658 μsec. Bins are annuli of equal width, starting with circle in center and moving outward. Bins weighted by area to give particles per unit area. Plot generated with Matplotlib/Pyplots.
Fig. 12.— Time averaged energy probability distribution from simulation of 10 particles with Gaussian energy distribution with mean at 7.03 keV and FWHM of 10 eV over 10 µsec simulated time. 80% of the time, particles have energy greater than 7.03 keV, meaning 40% of the time, particles co-propagate with energy and velocity useful for experimentation. Plot generated with Matplotlib / Pyplots.
Fig. 13.— Cross section of simulated chicane with D$_3^+$ beam (red), CD$^+$ beam (green), and C beam (black). With the apparatus' current setup, the first electrode-pair cannot sufficiently separate the CD$^+$ beam from the D$_3^+$ beam without the CD$^+$ beam striking the top of the second electrode-pair.

Fig. 14.— Chicane modification: the top of the second electrode pair is removed, and the tops of the second and third electrode pair are moved ~2 cm upward to allow D$_3^+$ beam (red) to enter faraday cup, and CD$^+$ beam (green) to be steered back in to beam path with C beam (black)
Conclusion

The novel merged beam apparatus constructed by the Savin group allows for fine-tuned measurement of the rate coefficients of reactions crucial to our understanding and modeling of the origins of organic chemistry in the cosmos. The measurements made using this apparatus will significantly lower our uncertainty in the rates at which these reactions take place. In my time at Nevis laboratories, I was able to simulate an ion trap that will possibly allow future versions of the apparatus to study reaction of C with H$_3^+$ at internal temperatures similar to those of interstellar gas-phase molecular clouds. I also simulated and modified the chicane for the apparatus to prove that studies can be performed studying these reaction complexes with deuterium instead of hydrogen.

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