# Experimental Evidence for the Existence of the $2p\sigma$ Bound State of $HeH^{2+}$ and Its Decay Mechanism

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Experimental evidence for the existence of a bound state of $HeH^{2+}$ is presented. Bates and Carson predicted a bound $2p\sigma$ state of this three-body system in spite of the strong repulsion between the nuclei. Decay is expected by electronic transition to the $1s\sigma$ repulsive ground state with a mean lifetime of $\sim 1$ nsec. A novel coincidence technique was developed for measurements of long lived molecular ions which dissociate in flight to the detector using a small powerful magnet to analyze the ions within a few nanoseconds. Coincidences between the fragments detected at the deflection angle associated with $HeH^{2+}$ indicate clearly that a bound state was formed during the collision and decayed after the analyzer.

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Three body problems have been of increasing interest in the last few years especially for three charged particles where the interaction between them is well known [1–9]. These systems provide a good test ground for improving our understanding of a few body problems. One classic example is the $H_2^+$ molecular ion which is separable in confocal elliptical coordinates within the Born-Oppenheimer approximation. Another example is the He atom which is more complicated because of the masses of the particles involved. This effect of the masses of the three interacting particles was studied from the He atomic limit (i.e., one heavy particle) to the $H_2^+$ molecular limit (i.e., one light particle) by Chen and Lin [1]. The effect of increasing strength of the attractive potential in the Hamiltonian can be studied using heliumlike ions with increasing $Z$. On the other hand, increasing the repulsive potential term in the Hamiltonian will generally result in an unbound system which can be studied by scattering methods only [10]. One exception from this trend is the asymmetric $HeH^{2+}$ molecular ion for which one bound state has been predicted by Bates and Carson [2]. This special system of three charged interacting particles has been solved analytically within the Born-Oppenheimer approximation and the energy curves of its low lying states are shown in Fig. 1. For this molecular ion the nuclear repulsion term, $Z_1Z_2/R$, is much larger than for the $H_2^+$ molecular ion. As a result the $1s\sigma$ ground state of $HeH^{2+}$ is repulsive, and so are most of the other electronic states. Winter, Duncan, and Lane [3] have shown that out of the lowest 20 states of $HeH^{2+}$ only the $2p\sigma$, $4f\sigma$, and $4f\pi$ states are bound. The lowest bound state is the $2p\sigma$ first excited electronic state. It has a minimum 0.849 eV deep at $R_0=3.89$ a.u. which can sustain a significant number of vibrational states. The $HeH^{2+}$ is the only system of three charged particles with a strong repulsive potential term in the Hamiltonian (i.e., larger than $e^2/R$) that are expected to be bound by their Coulomb interactions.

The mean lifetime of the $2p\sigma$ state is expected to be short because it can decay by an electronic transition to the repulsive $1s\sigma$ ground state. The oscillator strength of this transition, as well as other transitions between $HeH^{2+}$ states, have been calculated by Arthurs and coworkers [4]. The mean lifetime of the $2p\sigma$ bound state can be calculated using the well known spontaneous decay rate formula [11]

$$\tau^{-1}=W_{ka}^{s}=\frac{2\omega_{ka}^{2}}{mc^{3}}|f_{ka}|F^{2}, \tag{1}$$

where $F=\int\psi_{v'}^{*}\psi_{v}\,dR$ is the Franck-Condon factor. Us-

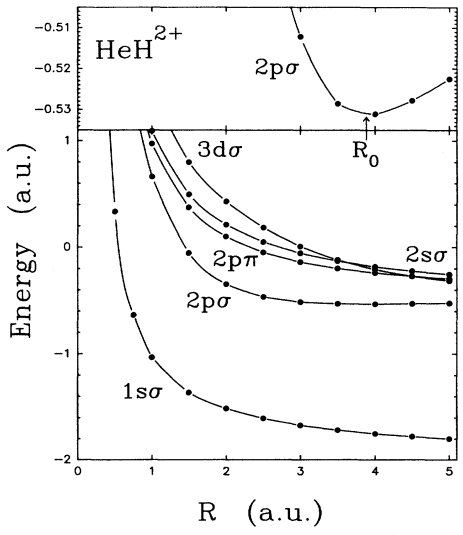


FIG. 1. Potential energy curves for $HeH^{2+}$ from Ref. [2] (zero corresponds to $He^{2+}+H^{+}+e$). The lines are a cubic spline fit to the calculated values at certain $R$ values indicated by the symbols.

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ing the transition frequency $\omega_{ka}(R_0) \sim 1.22$ a.u. and oscillator strength $f_{ka}(R_0) = 0.0250$ given by Arthurs and co-workers [4] and assuming that the Franck-Condon factor is approximately 1, the mean lifetime is estimated to be about 1 nsec.

Even though the $HeH^{2+}$ molecular ion was predicted to have a bound state by Bates and Carson [2] in the 1950s, no experimental evidence for its existence has been reported so far. The major problems making the experimental search for the $2p\sigma$ bound state of $HeH^{2+}$ difficult are its short mean lifetime and the low production probability. The short mean lifetime ($\sim 1$ nsec) limits us to methods in which the $HeH^{2+}$ ions are analyzed within a few nanoseconds of formation, which is clearly an experimental challenge. To make things even worse, the production probability of the $HeH^{2+}$ bound state by stripping an electron from $HeH^+$ is expected to be small because of the large difference between the equilibrium internuclear distances of the $HeH^{2+}(2p\sigma)$ bound state, $R_0 = 3.89$ a.u., and the $HeH^+(^1\Sigma^+)$ ground state, $R_0 = 1.46$ a.u. As a result of this large difference the Franck-Condon factor for these transitions is negligible for the lowest vibration level. This factor increases for the highly excited vibrational states of $HeH^+(^1\Sigma^+)$ because their wave functions extend further away from $R_0$. Thus, it is expected to be very small for the $2p\sigma$ state and negligible for the highly excited $4f\sigma$ and $4f\pi$ states for which the equilibrium internuclear distances are much larger ($R_0 = 11.766$ and 16.452, respectively).

We have developed an experimental method which enabled us to determine the existence of this short lived bound state of $HeH^{2+}$. In order to increase the chances for production and detection of the $HeH^{2+}(2p\sigma)$ bound state a fast $HeH^+$ beam from the 1 MV Technion Van de Graaff accelerator was used. The rf source of this accelerator is known to produce singly charged molecular ions in the electronic ground state with a significant fraction in highly excited vibrational states, thus making it possible to populate the $HeH^+(2p\sigma)$ state by a vertical stripping transition from the $HeH^+(^1\Sigma^+)$ electronic ground state. A beam energy of 900 keV, the highest available on this accelerator, was used. At this beam energy the ions' speed is about 6 mm/nsec. Thus, the target cell and analyzer have to be less than $\sim 20$ mm long if some reasonable number of $HeH^{2+}$ molecular ions are expected to survive through the system. In order to reduce contaminants in the $HeH^+$ beam, a velocity selector (Wien filter) was used between the 15° analyzing magnet and the target cell.

The experimental setup used for this experiment, shown in Fig. 2, consisted of a short target cell (6 mm), with small entrance and exit collimators (0.5 and 1.0 mm diameters, respectively), in which the $HeH^+$ projectiles collided with a thin Ar target ($P \sim 2$ mTorr). The ions produced in these collisions were analyzed according to their mass to charge ratio by a strong magnetic field ($\sim 0.5$ T) formed in the narrow gap between two small permanent magnets (each having a 12.7 mm diameter). The trajectory of the ions after the analyzer is determined within 2 to 3 nsec from the moment of their production. During this short time the number of $HeH^{2+}(2p\sigma)$ molecular ions is expected to be reduced by about an order of magnitude.

The analyzed ions are then detected by a surface barrier detector 830 mm downstream. A vertical slit, 2 mm wide, in front of the detector defines the angular resolution of the system to be about 0.1°. The position of this detector can be changed relative to the beam axis such that the yield of the different ions can be measured as a function of the deflection angle. Another surface barrier detector, with a 0.5 mm diam collimator, is placed on the beam axis for normalization. The trajectories of the

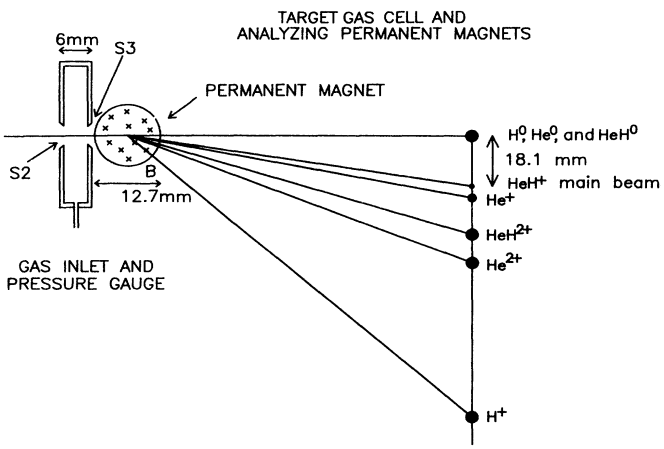


FIG. 2. Experimental setup and schematic trajectories of the different ions after the analyzing magnet.

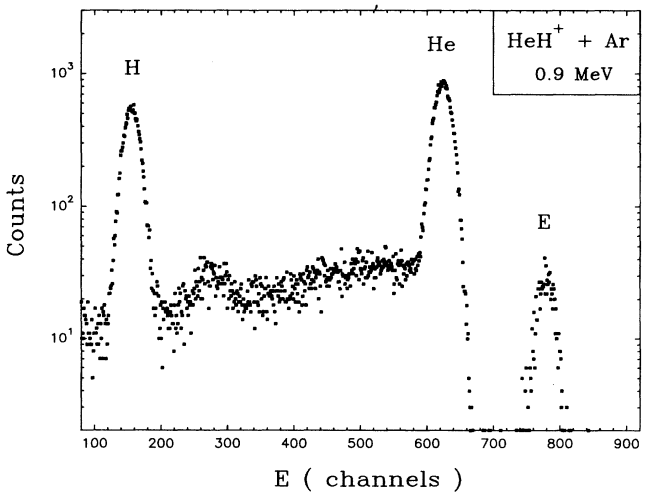


FIG. 3. Energy spectrum of the ions hitting the detector placed on the $HeH^{2+}$ trajectory. The full energy peak labeled by $E$ is associated with both the H and He fragments hitting the detector simultaneously.

different ions are well separated as shown in Fig. 2. The $HeH^{2+}$ molecular ions which have passed through the magnetic field before they have dissociated, are deflected by an angle of 2.5°. The flight time to the detector is of the order of 140 nsec, thus no $HeH^{2+}(2p\sigma)$ molecular ions are expected to reach it.

The ions that hit the detector produce a signal proportional to their energy. Hydrogen fragments should therefore peak at 0.2 of the beam energy while the He fragments should peak at 0.8 of the beam energy. (The fragments have approximately the beam velocity, thus the energy is proportional to their masses.) These peaks are clearly seen in the energy spectrum shown in Fig. 3. Furthermore, in cases where both the hydrogen and helium fragments hit the detector simultaneously the full energy of the beam will be deposited in the detector. These events contribute to the full energy peak, also shown in the figure.

The best signature for the formation of a bound state of $HeH^{2+}$ is the simultaneous detection of $H^+$ and $He^+$ fragments from its dissociation after the analyzing magnet. These fragments follow approximately the trajectory of the $HeH^{2+}$ molecular ions if the dissociation happens after the magnetic field. This trajectory is well resolved from the trajectories of the other ions which were produced before the magnet, as can be seen from Fig. 2. The deviations from the $HeH^{2+}$ trajectory caused by the "Coulomb explosion" of the molecule are small because the beam energy is orders of magnitude larger than the energy released in the dissociation. These $H^+ + He^+$ coincidence events are recorded under the full energy peak shown in Fig. 3.

The number of hydrogen, helium, and $H^+ + He^+$ coincidence events normalized to a constant number of hydrogen atoms is plotted as a function of the deflection angle in Fig. 4. The $He^{2+}$ and $He^+$ peaks appear at the expected deflection angles 3.2° and 1.6°, respectively, while the $H^+$ peaks at much larger angles. The small yield of $H^+$ and $He^+$ at all other angles is due to interactions with the residual gas between the pole faces of the magnet which is located at the exit of the target cell. The $H^+ + He^+$ coincidence rate peaks at the deflection angle where the $HeH^{2+}$ molecular ion is expected, whereas the random coincidence rate is practically zero at deflection angles far from the helium peaks. This indicates clearly that a bound state of the $HeH^{2+}$ molecular ion is formed in these collisions, as reported recently [12], and that it did not decay before passing through the magnetic field.

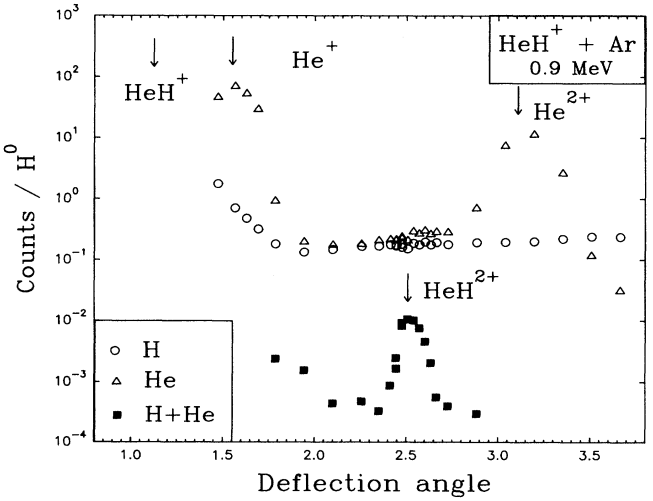


FIG. 4. Normalized number of counts of hydrogen ions, helium ions, and $H^+ + He^+$ coincidence events as a function of the deflection angle. The deflection angle of the main $HeH^+$ beam is marked for reference.

The apparent production rate of bound $HeH^{2+}(2p\sigma)$ molecular ions was determined to be about $2\times10^{-20}$ cm$^2$ by measuring their yield relation to the neutral He fragment production as a function of target pressure. The He fragment production relative to the main $HeH^+$ beam was determined in a similar way. The true production rate can then be calculated if the mean lifetime is known. The ions travel about 3 nsec through the system; assuming that the mean lifetime is of the order of 1 nsec, as estimated using Eq. (1), only ~5% of them are analyzed. Thus the true production cross section is of the order of $4\times10^{-19}$ cm$^2$. This cross section is only a rough estimate because the mean lifetime is not really known.

The decay of the $HeH^{2+}(2p\sigma)$ molecular ion is predicted theoretically to follow *via* an electronic transition to the $HeH^{2+}(1s\sigma)$ repulsive ground state, around $R_0 = 3.89$ a.u., which then dissociates rapidly into $H^+ + He^+$ releasing about 7 eV as kinetic energy. For this energy the radius of the spot on the detector is expected to be ~4.6 mm for the $H^+$ and ~1.1 mm for the $He^+$. The $H^+$ spot is larger than the vertical slit width used, thus only a small fraction of $H^+ + He^+$ pairs from this decay channel can be measured in coincidence, i.e., only the ones aligned approximately parallel to the vertical slit. On the other hand, for the $4f\sigma$ and $4f\pi$ highly excited bound states the energy released will be much smaller, and 2.4 and 1.7 eV, respectively, if they decay to the ground state and even less if they decay to a highly excited repulsive state. The radius of the spot on the detector for these states will be of the order of the slit size because of the low energy and most of these ions will be detected, if they are produced. The kinetic energy of the fragments can be evaluated by measuring the radial distribution of the $HeH^{2+}$ fragments, i.e., the $H^+ + He^+$ coincidence rate as a function of the aperture radius in front of the detector, thus determining which of the bound states of $HeH^{2+}$ was formed in the collision. This measurement was performed using a wider vertical slit of 4 mm (angular resolution of 0.2°) and an iris aperture in front of the detector placed on the $HeH^{2+}$ molecular ion trajectory. Furthermore, the distance between the detector and the magnet was increased to 1050 mm, to improve resolution.

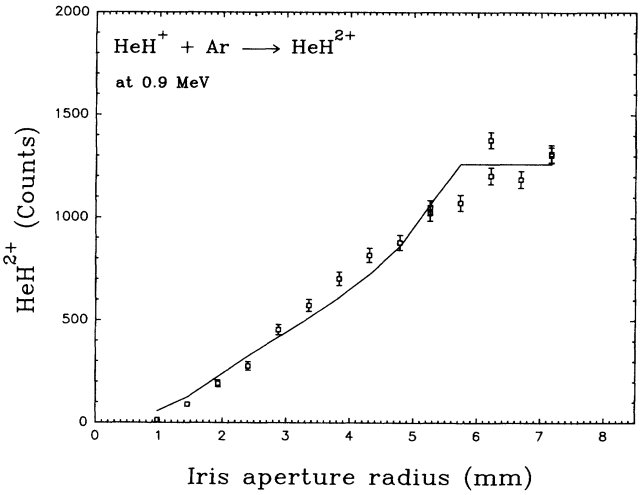


FIG. 5. Normalized number of coincidence events as a function of the aperture size in front of the detector placed on the $HeH^{2+}$ trajectory. The line is the expected distribution for an energy release of 6.0 eV.

The normalized number of counts under the full energy peak is plotted as a function of the iris aperture radius in Fig. 5. The $H^+ + He^+$ coincidence rate was negligible at the smallest aperture, indicating that no atomic or molecular contaminants (with $m/q = 2.5$) were present in the beam. This rate increases with increasing radius up to a constant value as expected for a dissociating molecule. In the figure we also show the radial distribution expected for a single value of energy release, $E_k$. Fitting this curve to the data the energy released was determined to be $6.0 \pm 1.7$ eV. This approximate value of $E_k$ is consistent with the theoretical value of $\sim 7$ eV expected for a $2p\sigma$ to $1s\sigma$ transition at $R = R_0$. On the other hand, this value is much larger than the expected energy release in the decay of the highly excited states. Thus, we may conclude that the populations of $4f\sigma$ and $4f\pi$ or even higher bound states of $HeH^{2+}$ are negligible as expected. The fragments' radial distribution thus provides experimental evidence that the bound state of $HeH^{2+}$ formed in these collisions is the lowest available one, and that it decays as expected by a $2p\sigma$ to $1s\sigma$ transition.

In summary, clear experimental evidence for the formation of a bound state of the $HeH^{2+}$ molecular ion in stripping collisions of 900 keV $HeH^+$ with Ar has been presented. The bound state of the doubly charged $HeH^{2+}$ molecular ion was identified by detecting its $H^+ + He^+$ fragments in coincidence along the trajectory where this molecular ion was expected. This novel method can be used even if no molecular ions reach the detector. The radial distribution of the fragments on the detector was also measured and it is consistent with the calculated one for the $2p\sigma$ to $1s\sigma$ transition, and inconsistent with the $4f\sigma$ and $4f\pi$ states, thus giving additional support to the discovery of the $HeH^{2+}(2p\sigma)$, and to its decay mechanism by electronic transitions to the repulsive $1s\sigma$ ground state. The $HeH^{2+}$ molecular ions survived at least 2–3 nsec in order to pass through the analyzing system, and all of them decayed before reaching the detector, suggesting a mean lifetime of the order of a few nanoseconds. The measurement of such a short mean lifetime with reasonable precision is difficult, but we plan to measure it directly in the near future using a new apparatus in which the distance between the target cell and the analyzing magnet can be varied. We hope that our measurements of the $HeH^{2+}(2p\sigma)$ bound state will initiate more experimental and theoretical studies of this bound system of three charged particles. In particular, better calculations of the mean lifetimes of the $2p\sigma$ state and more detailed measurements in which nonadiabatic effects might be seen are desirable.

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