Search for isotope effects in projectile and target ionization in
swift He$^+$ on H$_2$ / D$_2$ collisions

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Abstract

Using the COLTRIMS technique we have measured the simultaneous projectile and target ionization in collisions of He$^+$ projectiles with a mixture of gaseous H$_2$ and D$_2$ for an incident projectile energy of 650 keV. Motivated by Cooper et al. [Phys. Rev. Lett. 100, 043204 (2008)] we look for differences in the ionization cross section of the two isotopes with highest resolution and statistical significance. Contributions of the electron-electron and the electron-nucleus interactions have been clearly separated kinematically by measuring the longitudinal and transverse momentum of the recoiling ion. We find no significant isotope effect in any of our momentum distributions.

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I. Introduction

On the atomic scale, interaction such as photo ionization and charged particle impact is completely dominated by the electromagnetic force. If one neglects the tiny hyperfine structure splitting, the nucleus just provides the Coulomb potential in which the electrons move. The ionization of single atoms by charged particles is therefore not expected to depend on the isotope. Because the chemical behavior of an atom is largely determined by its electronic structure, different isotopes also exhibit very similar chemical behavior. The main exception to this is the kinetic isotope effect: due to their larger masses, heavier isotopes tend to react slightly more slowly than lighter isotopes of the same element. This effect is most pronounced for H and D, because deuterium has twice the mass of the proton. The mass effect between H and D also affects the behavior of their respective chemical bonds by changing the center of gravity (reduced mass) of the atomic system \[1, 2\]. For heavier elements the mass-difference effects on chemistry are usually negligible. In general, absolute and differential cross sections for ionization by fast charged particles are believed to be isotope independent.

In surprising contrast, Cooper and co-workers observed differences in quasi-elastic (energy transfer is small compared to the incident energy of the scattered particles) electron-scattering cross sections from gaseous H\(_2\), D\(_2\) and HD molecules \[3\]. Electron scattering with a relatively high momentum transfer was measured and experimentally determined cross sections were compared with calculated ones using Rutherford scattering theory, where the scattering cross sections depend only on the charge and not on the mass of the target. Cooper et al. found however, that the ratio I(H)/I(D) of the scattering intensities showed a much smaller value than expected from this conventional theory. For a 50:50 H\(_2\)-D\(_2\) gas mixture they found a ratio of the respective scattering intensities of \(\approx 0.7\) (a shortfall of \(\approx 30\%\)), while for the HD gas \(\approx 1\) has been seen as expected. Following these results, the absolute elastic scattering probability for H\(_2\) and the appropriate geometrical cross section seems to be smaller than expected. On the other hand, the ratio of the electron scattering intensities of H and D in the case of the HD gas agrees well with the predictions of Rutherford scattering.

Cooper et al. \[3\] speculated that entanglement between identical nuclei (in H\(_2\) and D\(_2\)) could play a crucial role. They argued that possibly short-time entanglement of the
protons with their adjacent electrons and nuclei might reduce the cross section. With this speculation they challenged the common independent scatterer model. They assumed that for fast, large momentum transfer electron scattering on a very short time scale the H\textsubscript{2} wave function can not be separated to a product of single particle wave function for electrons and nuclei and that the scattering is not effectively the sum of the scattering at any of these isolated particles. The entangled scatterer could lead to a smaller cross section in contrast to conventional theory. Then the lifetime of these quantum entanglements could be measured indirectly. They estimate that their scattering processes correspond to typical scattering times of about 500 attoseconds. If the entanglement between neighboring particles is responsible for the anomaly in cross sections, the lifetime of the entanglement must be much longer than the scattering time. In this case, the lifetime of the entanglement could be probed by varying the momentum transfer and therefore the scattering time.

Related anomalies in the cross sections of hydrogen have been observed for different scattering systems: neutron scattering from H\textsubscript{2}O and D\textsubscript{2}O molecules \[4\], electron and neutron scattering from solid polymers \[5\], neutron scattering from molecular hydrogen \[6\]. All these results represent a challenge for conventional scattering theory as well as for molecular spectroscopy. No quantitative theory so far could explain all these anomalies, but all suggested approaches today contain quantum entanglement \[6–11\]. There is also criticism of the measurements by Cooper et al., see \[12\] and references therein. Moreh suspects that the origin of the above deviations is instrumental and not due to any real deviation from the Rutherford formula. He argues that the heavier gas component in any binary gas mixture from an inlet spends on average more time in the interaction region which would cause the observed anomaly.

To shine more light on these unexpected experimental findings in electron and neutron scattering, we here use a different approach. We investigate the following reaction:

\[He^+ + H_2/D_2 \rightarrow He^{2+} + H_2^+/D_2^+ + 2e^-\] (1)

In this process simultaneously one electron is ejected from the target (H\textsubscript{2} / D\textsubscript{2}) and one from the projectile (He\textsuperscript{+}). There are two mechanisms, termed ee and ne which contribute to this reaction. In the ee process the projectile electron is knocked out by an interaction with the target electron. This violent collision between two bound electrons leads to ejection of
both of them from their respective binding. In the second process, termed \( ne \), the projectile electron is ejected by an interaction with the target nucleus and a second, independent interaction between the projectile nucleus and one of the target electrons facilitates the ejection of the target electron. Depending on the impact energy one of these two processes dominates \([13]\). The \( ee \) process occurs at rather large impact parameters. The electrons knock each other out, while the nuclei are spectators in the reaction (see Fig. 1a). Therefore the overall momentum transfer to the target nucleus is small. A projectile velocity greater than 2 a.u. is needed to initiate the process. This threshold is due to the fact that the target electron, as seen from the projectile frame, must have sufficient kinetic energy that it can ionize the projectile and simultaneously escape from the target. For \( \text{He} \) a projectile energy of 0.4 MeV is equivalent to an electron energy of 54 eV, which is the projectile binding energy \([13, 14]\). For the \( ne \) process on the contrary the impact parameters are much smaller leading to a larger transverse momentum transfer (perpendicular to the projectile beam direction) and in addition the longitudinal momentum (parallel to the projectile beam direction) is also compensated by the target nucleus \([15]\). Both electron loss processes are therefore separated in momentum space of the recoiling molecular hydrogen ion. Fig. 1c shows the recoil ion momentum distribution for a collision of \( \text{He}^+ + \text{He} \rightarrow \text{He}^{2+} + \text{He}^+ + 2e^- \) at 1 MeV. The most detailed investigations also measured the momenta of both electrons involved in the loss processes \([16–18]\).

The key idea of our experiment is, that the \( ne \) mechanism involves a violent scattering of the electron bound to the \( \text{He}^+ \) at the nucleus of the \( \text{H}_2 \) or \( \text{D}_2 \), while the \( ee \) mechanism involves only the electrons. The 30% isotope differences in the scattering of free electrons at the nuclei reported in \( 3 \) might thus also affect the scattering of bound electrons in our collision system. The strength of our experiment is, that the \( ee \) mechanism allows for robust in situ normalization of the data. While in any electron or neutron scattering experiment the comparison of cross sections for the different isotopes relies on the knowledge of the isotope ratio in the target gas mixture, this possible source of systematical error is not present in our experiment. The \( ee \) mechanism in our case is a scattering between the electrons and the nuclei are not active participants. Hence the previously reported isotope effects in electron and neutron scattering would not influence the \( ee \) contribution to the electron loss channel but only the \( ne \) contribution. We therefore search for differences in the \( ne \) versus \( ee \) contributions between the isotopes. While no standard scattering theory would predict
such an isotope effect the recent results by Cooper et al. on free electron scattering would suggest such a difference of up to 30%.

FIG. 1: (Color online) COLTRIMS allows to separate experimentally the contribution of $ee$ interaction from the $ne$ interaction to the ionization of the projectile as shown in the doubly differential cross section $d^2\sigma/(dp_\parallel dp_\perp)$. Fig. 1c for illustration shows the recoil ion momentum distribution for a collision of $He^+ + He \rightarrow He^{2+} + He^+ + 2e^-$ at 1 MeV, the horizontal axis shows the longitudinal recoil ion momentum (parallel to the projectile beam direction) and the vertical axis the transverse recoil ion momentum (perpendicular to the projectile beam direction).

II. Experiment

In our experiment we use the COLTRIMS (Cold Target Recoil Ion Momentum Spectroscopy) technique [19–21] to examine the electron loss process [22–25] in collisions of He$^+$ projectiles with a gas mixture of 50% H$_2$ and 50% D$_2$. The He$^+$ ion beam is provided by the 2.5 MV Van-de-Graaff accelerator at the Institut für Kernphysik of the Goethe-Universität in Frankfurt. The beam is collimated using three sets of adjustable slits. In front of the reaction zone an electrostatic deflector (beam cleaner) is used to separate the He$^+$ beam from charge state impurities (He$^0$ or He$^{2+}$). The reaction takes place in the overlap region of the He$^+$ beam with a supersonic gas jet providing the H$_2$ and D$_2$. An electric field projects the produced recoil ions ($H_2^+ / D_2^+$) towards a time- and position-sensitive detector [26, 27] yielding an acceptance angle of 4$\pi$ for ions up to 5 a.u. momentum. Measuring the impact position on the detector and its time-of-flight, the particle trajectories and thus the particle momenta can be determined. To optimize the ion momentum resolution, an electrostatic
lens is incorporated into the spectrometer system. Trajectory calculations including such lenses can be found in [28]. The diminishing influence of the extended interaction region on the momentum resolution can be strongly reduced by this focusing geometry. A drift tube following the acceleration part of the spectrometer yields focusing of the times-of-flight also in the third direction. The spectrometer has an electrical field of 6.5 V/cm and a length of 28 cm plus 150 cm recoil drift. Downstream of the spectrometer another electrostatic deflector is used to charge state analyze the projectile beam after the reaction. He$^+$ projectiles are dumped in a Faraday cup and only He$^{2+}$ particles from the electron loss reaction are measured with a second position sensitive detector. They serve as a trigger for our coincidence experiment. The data are acquired and stored in list mode format event-by-event. The typical times-of-flight were 12 $\mu$s for H$_2^+$ and 17 $\mu$s for D$_2^+$. The momentum resolution was determined by the simultaneously measured electron capture reaction He$^+$ + H$_2$/D$_2$ → He$^0$ + H$_2^+$/D$_2^+$ to be 0.1 a.u. Small fractions of false coincidences (< 10 %) were subtracted. The main source for this background is the single ionization with a wrongly detected projectile. In momentum space they are located around $p_\parallel \approx 0$ and in perpendicular direction constantly from 0 to maximum (time-of-flight background). Therefore we took an average of events, being left and right of the corresponding H$_2$ or D$_2$ time-of-flight peak and subtracted these from the data (although the difference between left and right side of the time-of-flight peaks was only ± 0.3 %).

III. Results and discussion

One possible source of errors in this experiment is the dissociation of D$_2^+$ which leads to a D$^+$ ion that has the same time-of-flight as H$_2^+$ and also different detection efficiency than D$_2^+$ ($\approx 3 \%$ [29]). These problems can be solved by normalization of the $ee$ process of the measured H$_2^+$ ions to the D$_2^+$ ions. Furthermore, to avoid the possible problem of regions of reduced detection efficiency on the detector, we changed the position of the detector and measured the H$_2^+$ and D$_2^+$ ion spots at different positions on our detector. To avoid background from dissociating D$^+$ and also H$_2^+$ from the residual gas and to clean the spectra we analyze only events with positive momentum in jet direction ($y$) without loss of generality. In this way we can completely separate the signal of H$_2^+$ and D$^+$.

For the main results of this paper we choose He$^+$ projectiles with an energy of 650 keV
(162.5 keV/u) colliding with H\textsubscript{2} and D\textsubscript{2}. This projectile energy had to be tuned in a way that both processes \textit{ee} and \textit{ne} can be clearly resolved. The \textit{ne} process dominates at smaller projectile energies, while the \textit{ee} process is dominant at higher energies. The ratio of both processes for H\textsubscript{2}\textsuperscript{+} depending on the projectile energy can be seen in Fig. 2 and 3.

The projectile energy dependence of the \textit{ee} process is shown in Fig. 3. In the H\textsubscript{2}\textsuperscript{+} doubly differential cross sections a circle of 0.35 a.u. radius around \(p_\parallel = 0.35\) a.u. and \(p_\perp = 0.35\) a.u. (circle line in Fig. 2) was selected which characterizes the \textit{ee} part of all counts in these spectra. The ratio of this part to all counts was plotted and shows an expected rise of the \textit{ee} process depending on the projectile energy.

After determining the \textit{ee} to \textit{ne} ratio for different projectile energies, we chose 650 keV for our comparison of H\textsubscript{2} versus D\textsubscript{2}, because both processes are similarly intense at this energy. Both molecules are separated in time-of-flight as well as in position on our detector. The recoil ion momenta perpendicular and parallel to the projectile beam direction are plotted in Fig. 4. H\textsubscript{2} is shown on the left, D\textsubscript{2} on the right. In the region of small momentum transfers
FIG. 3: Ratio of the $ee$ process to all counts in mutual projectile and target ionization in He$^+$ + H$_2$ collisions in dependence of the projectile energy. ($p_\parallel < 0.5$ a.u. and $p_\perp < 1$ a.u.) both spectra show a maximum which corresponds to the $ee$ process. The second maximum at larger total momenta corresponds to the $ne$ process.

For a more detailed comparison the longitudinal and transverse momenta of both isotopes are presented separately in Fig. 5. The data have been normalized to the integral (not peak maximum). In both directions the momentum distributions are almost identical in shape. The size of the statistical error bars is smaller than the points.

Fig. 6 shows the ratio of H$_2$ / D$_2$ for the transverse (a) and the longitudinal (b) momentum. Small deviations up to 5 % fluctuate around 0. Linear fits yield an intercept value of $-0.0077 \pm 0.0047$ and a slope of $-0.0049 \pm 0.0024$ for the case of transverse and an intercept of $0.0065 \pm 0.0058$ and a slope of $-0.0065 \pm 0.0039$ for the case of longitudinal momentum. These small numbers give a good idea of the diminutiveness of any deviation from the expectations from standard scattering theory. Most importantly however the observed deviations from unity are far smaller than 30 % deviation reported for electron or neutron impact [3–6].

As outlined in the introduction an isotope effect on electron scattering of 30 % would alter the $ne$ but not the $ee$ contribution. No such effect is observed in our data in full agreement with the expectation of standard scattering theories.
FIG. 4: (Color online) Recoil ion momenta transverse and longitudinal to the incident beam direction for mutual projectile and target ionization for 650 keV He$^+$ impact onto H$_2$ (a) and D$_2$ (b).

IV. Conclusions

The goal of the present measurement was to search for isotope differences in the ionization dynamics in a heavy particle collision with H$_2$ and D$_2$. This search is motivated by recent reports on unexpected and so far unexplained isotope differences in the elastic scattering of electrons and neutrons [3–6] of up to 30%. We have investigated electron loss which for one channel ($ne$) be thought of as a scattering of a quasi-free electron at the target nucleus (H$_2$, D$_2$) while a second channel ($ee$) provides an independent in situ normalization. In contrast to the experimental findings of Cooper et al. the present experimental results do not exhibit any significant differences above 5% between the H$_2$ and D$_2$ targets. This ‘null’ result is in line with the expectation from all standard scattering theories. A possible reason for the opposite conclusion drawn from our experiment as compared to the surprising results of the electron and neutron scattering is, that the momentum transfers are in different regimes: Cooper et al. [3] used a momentum transfer $q$ of 19.7 a.u., while in our experiment only momentum transfers of up to 5 a.u. were measured. So a rather different momentum transfer region is probed in this case. At the end of our momentum distribution in Fig. 6 we can see a tendency of an increasing normalized difference for perpendicular momenta and a decreasing normalized difference for parallel momenta. This could be a hint for
FIG. 5: (Color online) Longitudinal and transverse momenta of H$_2^+$ and D$_2^+$ for mutual projectile and target ionization by 650 keV He$^+$ impact normalized to their integral. The H$_2$ data points are shown as black squares and the D$_2$ data points as red circles. The data are projections of the data shown in Fig. 4 onto the horizontal or vertical axis.

FIG. 6: Normalized differences of the H$_2^+$ and D$_2^+$ ions (i.e. $(\sigma_{H_2} - \sigma_{D_2})/(\sigma_{H_2} + \sigma_{D_2})$, results from Fig. 5). In the case of parallel momentum events below 0 a.u. are only background and have no physical meaning, therefore they are not shown.

the reported isotope effect at higher momentum transfers, but experiments that enable these higher momentum transfers are needed to explore this possible momentum transfer dependency, as the observed effect could either depend on the momentum transfer or occur only at higher momentum transfers. Chatzidimitriou-Dreissmann suggests an increase of
the anomaly with increasing momentum transfer \[4–6,11\]. For further investigation of this question we plan to perform measurements with higher momentum transfers, but these can not be reached with our Van-de-Graaff accelerator, a storage ring is needed instead. For the sum of electron energies in the projectile frame, we can here use the approximation

$$\sum_{i=1,2} E_{\text{kin},e_i}^p \approx p_{\parallel,\text{rec}} \cdot v_p = 3.5 \text{a.u.} \cdot 2.55 \text{a.u.} = 8.93 \text{a.u.} \approx 243 \text{eV} \quad [19].$$

Although the electron energies are not presented in this paper, we consider only electrons below 243 eV due to 3.5 a.u. maximum longitudinal recoil ion momentum.

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