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Mesic molecular effects in the capture of negative pions stopped in gaseous hydrogen isotopes

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The influence of molecular structure on the nuclear capture probability of stopped negative pions has been observed by comparing the π^0 gamma-ray spectrum from π^- mesons stopped in HD gas to that from a mixture of equal amounts of H_2 plus D_2. The fraction of stopped pions that are captured by a proton in the H_2+D_2 mixture is $f_{\mathrm{H_2D_2}}=0.417\pm0.004$, while for HD it is $f_{\mathrm{HD}}=0.338\pm0.008$, independent of the gas pressure between 6 and 90 atm. The ratio, $f_{\mathrm{H_2D_2}}/f_{\mathrm{HD}}$, of the fractions is 1.23 ± 0.03.

I. INTRODUCTION

The absorption of pions or muons in matter is a phenomenon that is of interest in several disciplines. This is so because the stages of slowing down, stopping, and eventual capture by an atomic nucleus depend on the structure of the stopping material at the molecular, atomic, and nuclear levels. Several review articles[1–6] have treated the influence of molecular structure on the capture probability of pions on various nuclei comprising a molecule. In fact, recent measurements[7] on organic molecules of biological interest reveal that a markedly different capture schedule of π^- can occur between structural isomers. In the case of pion capture on complex nuclei experimenters[7] have relied upon the pionic x rays to determine the eventual absorption site of the pion. For hydrogen in compounds the charge-exchange reaction (1b)

$$\pi^- + p \rightarrow \begin{cases} n+\gamma_0 & \text{(1a)} \\ n+\pi^0 \rightarrow n+\gamma_1+\gamma_2 & \text{(1b)} \end{cases}$$

can be used as the signature of pion capture on protons. The π^0 production is a very clean signature since it is prominently observed only in the case of absorption on protons or ^{3}He. In deuterium, for example, the branching ratio for π^0 production from stopped π^- is about 1.45×10^{-4} (Ref. 8), and in other nuclei it is even less ($<10^{-5}$).[9]

Current experiments show that the pion is captured in regions of a molecule which have high electron density. Then, according to the model of large mesic molecules[1,2,10] a $p\pi^-$ atom is formed when the pion, captured on a molecular orbit, goes to an atomic orbit on the proton. Because the probability of Auger capture is related to the overlap between the wave function of the pion and the electron, the initial molecular orbit of the pion will be close to that of the displaced electron.[1] If the pion goes to a proton or deuteron, the $p\pi$ or $d\pi$ atom wanders through the target and through collisions either deexcites or transfers the pion to the collision partner. The $p\pi$ and $d\pi$ atoms are unique in that they are small and neutral and can come close to nuclei in a collision. This model can be parametrized by three terms P,Q,R. Let f be the fraction of pions eventually captured by protons in the target. Then

$$f=PQR\ , \tag{2}$$

where P is the probability that the pion is captured into a molecular orbit localized near the proton, Q is the probability that the pion makes a transition from the mesomolecular orbit to the $p\pi$ atomic levels, and R is the probability that the pion is retained by the proton in a collision of the $p\pi$ atom rather than transferred to a heavier collision partner. The retention probability R has been studied for several gas mixtures of the form H_2+Z (Ref. 11) and C_mH_n+Z (Ref. 12) and also H_2+D_2 (Ref. 13). In the case of H_2+D_2 mixtures it is found that only about 43% of the stopped pions are absorbed on the protons of the H_2 molecule for an equal mix of the two hydrogen isotopes. The molecular distribution factor Q is thought to have a $1/Z^2$ dependence.[10] Because of this, the dependence of Q on the reduced mass of the $p\pi$ system is rather difficult to determine if $Z\gg1$. Thus the simplest molecule possible where one can investigate the influence of

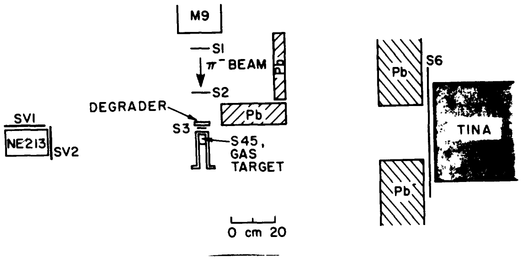

FIG. 1. Experimental setup, as described in the text.

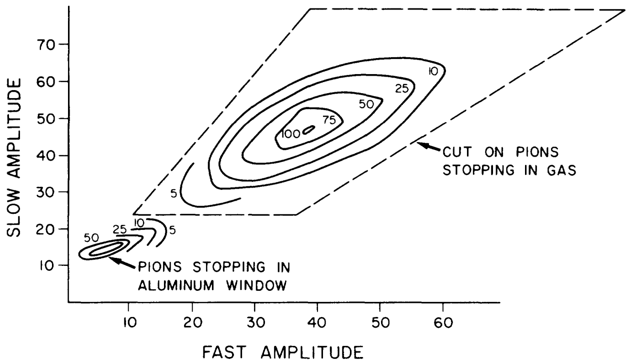

FIG. 2. Slow component (CsI) vs fast component (NE110 + CsI) for the 90-atm H_2 target. Dashed boundaries show the stop in gas cut.

the large pion mass is HD. The electron densities, assumed to determine the probability P, in H_2, D_2, and HD should be the same except for very small differences due to the small p-e and d-e reduced mass differences. In parallel with the study of stopped pion absorption in HD we can measure the effects in an equal mixture of H_2+D_2. Thus R can also be determined and in first approximation it should be independent of whether the gas is H_2+D_2 or HD because the collision is between a $p\pi$ or $d\pi$ atom and a p or d nucleus. Consequently, if $Q_{\mathrm{HD}} \neq Q_{\mathrm{H_2D_2}}$, or equivalently, $f_{\mathrm{HD}} \neq f_{\mathrm{H_2D_2}}$, then we can surmise that an intermediate large mesomolecular system exists in the capture sequence which is distinct from the three-body systems present in the collision stage. The subscript $x = \mathrm{HD}, \mathrm{H_2D_2}$ in f_x specifies the gas, either HD or the H_2+D_2 mixture. Initial indications of this effect have been seen in measurements of the gamma spectra from stopped π^- in liquid D_2,[8] and stopped π^- in liquid T_2.[14] In both cases fewer gamma rays from the hydrogen (H) contamination were observed than anticipated from the known H contamination level.

The obvious advantage of the HD system is that it should be possible to make *ab initio* calculations for the various possible transitions. In other molecules it seems too difficult, at present, to do more than make observations and link them together with a crude phenomenology.

II. EXPERIMENTAL TECHNIQUES

A. Target gases

Four targets were used in the experiment: H_2, D_2, H_2+D_2, and HD. The H_2 and D_2 were supplied at 99.99% hydrogen content. We manufactured the HD ourselves using the reaction $\mathrm{LiAlH_4+4D_2O \rightarrow LiOD + Al(OD)_3+4HD}$ as described in the article by Fookson *et al.*[15] Instead of using n-butyl ether dried over sodium, as described in Ref. 15, we used tetrahydrofuran. Analysis of the HD showed it contained (93±2)% HD and the rest of the hydrogen was H_2 and D_2. During the experiment we passed half of the gas sample through a twenty turn copper tube immersed in liquid nitrogen. By measuring the residual gas pressure in the cold trap after it warmed to room temperature we ascertained that the concentration of impurities was less than 10^{-5}.

B. Measurement technique

Pions with $T_\pi = 30$ MeV from the $M9$ channel at Tri-University Meson Facility (TRIUMF) were stopped at the $F2$ location in the gas targets. High-energy ($E_\gamma > 12$ MeV) gamma rays from pion capture on protons and deuterons were detected in the large NaI(Tl) (46-cm-diam×51-cm) detector TINA (TRIUMF iodide of natrium). The experimental setup is shown in Fig. 1. Counters $S1, S2, S3$ counted the incident beam. The Al degrader was used to slow the pions down to stop them in the high-pressure gas target. The high-pressure target has been previously described.[16] Essentially, it contains a cup made of two kinds of scintillators ($S45$ in Fig. 1), a CsI(Na) entrance counter and a plastic scintillator (Nuclear Enterprises NE110). The cup is viewed by a single 5-cm photomultiplier (RCA 8575). By exploiting the differences in the light output decay times between the slow CsI crystal and the fast plastic scintillator we can determine if a particle has penetrated the CsI and not stopped in the plastic. These stopped events then consist of particles stopping inside the cup (in the gas) or in the CsI. In Fig. 2 we present a contour plot of the slow component versus the fast component for the 90-atm H_2 run. To determine the spectrum of events that stopped in the CsI we evacuated the gas target and ran under the same conditions as with the target filled. The threshold on $S3$ was set sufficiently high so that electrons would not trigger the strobe. By examining the time-of-flight spectrum of particles coming down the channel we determined that only about 1 in 7000 events were attributable to electrons or muons. The event strobe was generated by $1\cdot2\cdot3\cdot(\mathrm{TINA})$. Charged triggers could be distinguished from neutrals by setting a bit in a coincidence buffer when $S6$ fired. The neutral triggers could be separated into neutrons and γ rays by their time of flight from the target to TINA. The time-of-flight spectrum for 88-atm D_2 is shown in Fig. 3.

A liquid scintillator (Nuclear Enterprises NE213) was used for monitoring the neutron flux from the target. It also provided an independent strobe based on $1\cdot2\cdot3\cdot(\mathrm{NE213})\cdot\overline{\mathrm{SV1}}\cdot\overline{\mathrm{SV2}}$.

With an incident flux $S1\cdot S2\cdot S3$ of $1.1\times10^5 \pi^-/\mathrm{sec}$,

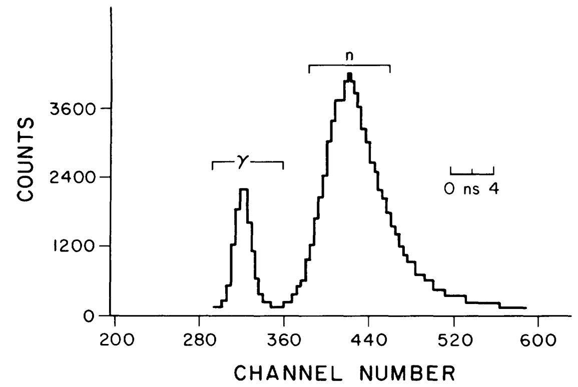

FIG. 3. Time-of-flight spectrum of neutral events for 88-atm D_2.

our pion stop rate in the cup was about 4300 per sec (3.9% stopped) for 90 atm of H_2. Gamma-ray spectra were measured for pressures between 6 and 90 atm for the four gases. Pressure measurements were made using a strain gauge transducer as described in Ref. 16.

The HD was stored at about 11 atm, and to bring it up to 100 atm we used a diaphragm compressor.[17] We also used the compressor to thoroughly mix the H_2 and D_2 for the mixed gas runs. First, one gas was introduced into the target to 50 atm. This was then transferred to a mixing bottle with about seven times the volume of the gas target plus tubing. This procedure was repeated for the second gas. After both gases were mixed in the mixing bottle, the mixed gas was pumped back into the target. The target was evacuated to a pressure less than 100 mTorr before any gas transfer took place. Thus atmospheric gases could not contaminate the mixture to more than about 10^{-6}, which was well below the 10^{-4} contamination level of the H_2 and D_2 as supplied by the manufacturer.

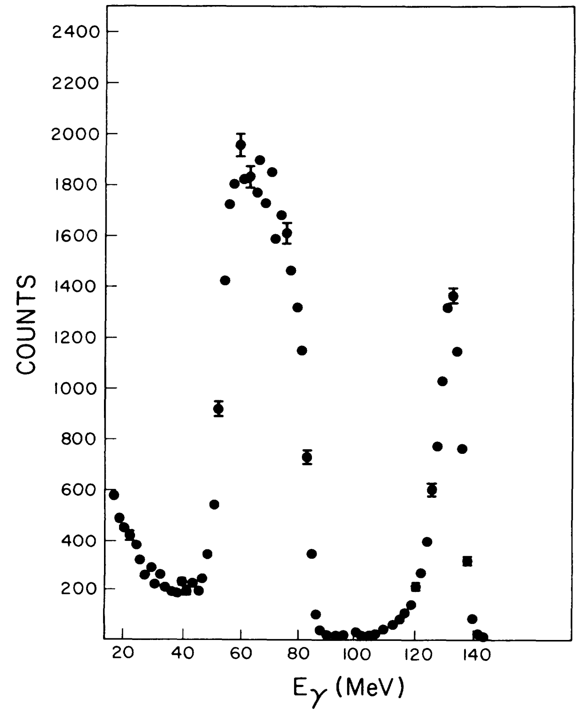

FIG. 4. Pure H_2 spectrum after background subtraction for 93 atm and $S1 \cdot S2 \cdot S3 = 6.6 \times 10^7$. Peak at 130 MeV is γ_0 from the radiative capture and the box between 50 and 85 MeV is due to γ_1 and γ_2 from the charge-exchange reaction. See Eqs. (1a) and (1b).

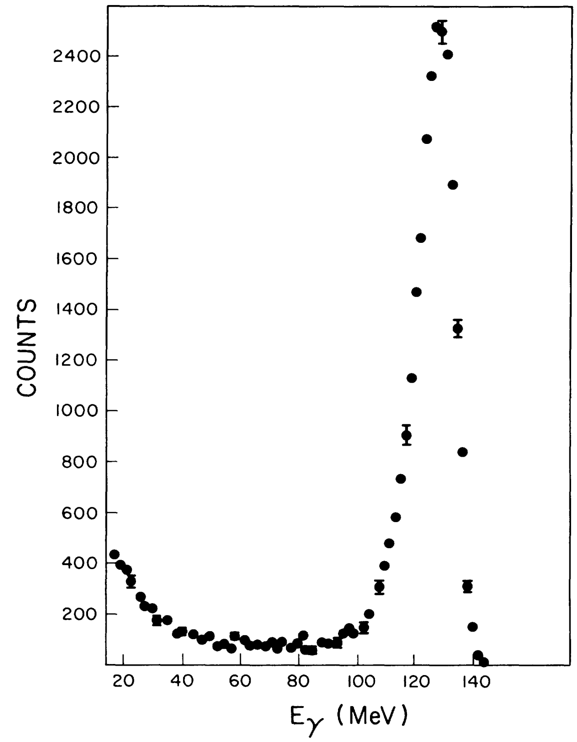

FIG. 5. Pure D_2 spectrum after background subtraction for 88 atm and $S1 \cdot S2 \cdot S3 = 29.7 \times 10^7$.

III. RESULTS

A. Analysis based on π^0 yields

In Figs. 4, 5, and 6 we present spectra taken for H_2, D_2, and the CsI background with all cuts applied, respectively. Most of background gammas contribute above the π^0 gammas in the region of γ_0. The most reliable hydrogen signal is that from the charge-exchange reaction as can be seen by the absence of this signal in the D_2 and CsI spectra.

To determine the fraction f_x of pions that are captured by protons in the gas x, we must measure the flux of gamma rays per pion stop in the gas. We could also measure the yield relative to a quantity proportional to the number of stops, such as $S1 \cdot S2 \cdot S3$. In Fig. 7 we present the yield of gammas in the π^0 region normalized to $S1 \cdot S2 \cdot S3$ versus the virially corrected density (ρ/ρ_0). The quantity ρ/ρ_0 is nearly the same as the absolute pressure in atmo-

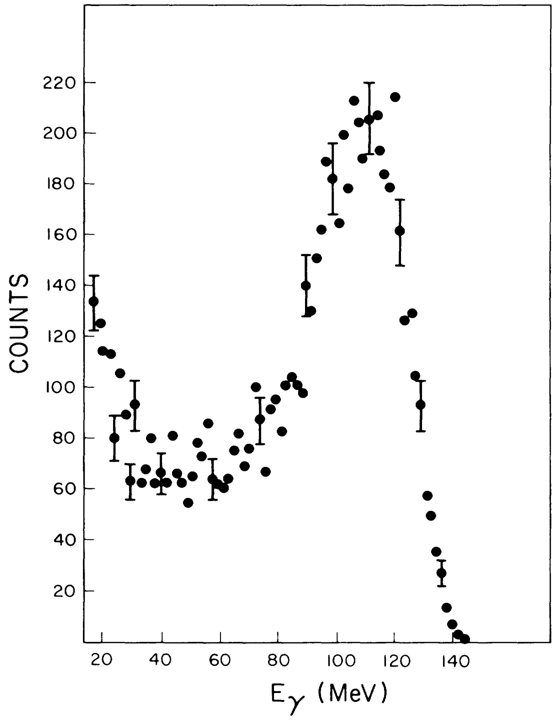

FIG. 6. Spectrum of events that stopped in the CsI scintillator for 0 atm and $S1 \cdot S2 \cdot S3 = 44.7 \times 10^7$.

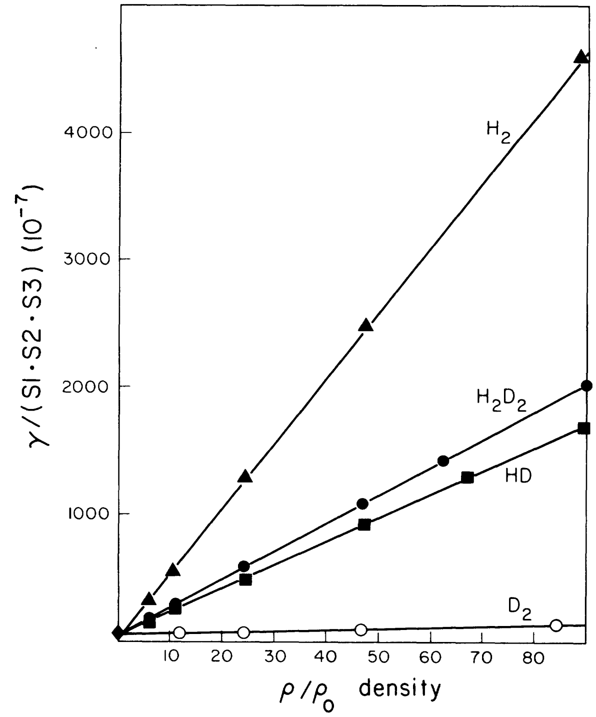

FIG. 7. Yield of γ rays for $40 < E_\gamma < 90$ normalized to $S1 \cdot S2 \cdot S3$ vs virially corrected density for the four target gases; (▲) H_2, (●) H_2+D_2, (■) HD, (○) D_2, and the background yield (◆).

spheres except for the virial corrections. This correction was made for H_2 and D_2 in the same way as in Ref. 16 and for H_2+D_2 and HD the empirical mixing rules and quantum corrections described in Refs. 18 and 19 were used. The second virial coefficients used are:

$$\bar{B} = 5.9 \times 10^{-4}/\text{atm} ,$$

$$\bar{B} = 5.6 \times 10^{-4}/\text{atm} ,$$

$$\bar{B} = 5.75 \times 10^{-4}/\text{atm} ,$$

for H_2, D_2, and HD and H_2+D_2, respectively, where

$$\rho/\rho_0 = \bar{P}(1 - \bar{B}\bar{P}) \tag{3}$$

and $\bar{P}$ is the absolute pressure. It is observed that the yield is linear with density. There is nothing significant in this other than that it is a useful means of combining the data. It is clear from Fig. 7 that the π^0 production is greater for the mixture H_2+D_2 compared to the molecular form HD. At zero density we have plotted the background value and note that all the yields extrapolate to this value. We suppose that at a given density, the same number of pions stop for all four gases. Only when the pion energy reaches some tens of electron volts might the vibrational and rotational structure of the molecules play a role in the stopping. Thus in general the gamma yield γ_x from the gas x can be written as

$$\gamma_x = f_x \gamma_{H_2} + (1 - f_x)\gamma_{D_2} . \tag{4}$$

The yields in this case are the slopes of the lines in Fig. 7. Because we are taking ratios, all efficiency and solid angle effects cancel and the yield γ_x is directly proportional to the fraction f_x of pions absorbed on the proton. From Fig. 7 a least-squares fit to the yields gives, for HD and H_2+D_2, respectively, the following values f_x:

$$f_{HD}^m = 0.355 \pm 0.005 ,$$

$$f_{H_2D_2} = 0.416 \pm 0.005 ,$$

where f_{HD}^m is our measured fraction for the HD sample before correcting for the H_2+D_2 (7%) contamination. We denote by M^m the ratio $f_{H_2D_2}/f_{HD}^m$

$$M^m = (\gamma_{H_2D_2} - \gamma_{D_2})/(\gamma_{HD} - \gamma_{D_2}) \tag{5}$$

and $M^m = 1.175 \pm 0.012$. Our value $f_{H_2D_2}$ agrees with that deduced from Ref. 13. In their experiment γ-γ coincidences from the π^0 decay were used to obtain $f_{H_2D_2} = 0.43 \pm 0.09$.

We also fitted the HD and H_2+D_2 spectra using the H_2, D_2, and background spectra as components. An example of a fitted spectrum for HD is shown in Fig. 8. The fitted hydrogen component yield is shown in Fig. 9. All three curves extrapolate to zero yield at zero density. The values f_x and M^m can be obtained by directly comparing the slopes in Fig. 9. For HD,

$$f_{HD}^m = 0.355 \pm 0.005 ,$$

for H_2+D_2,

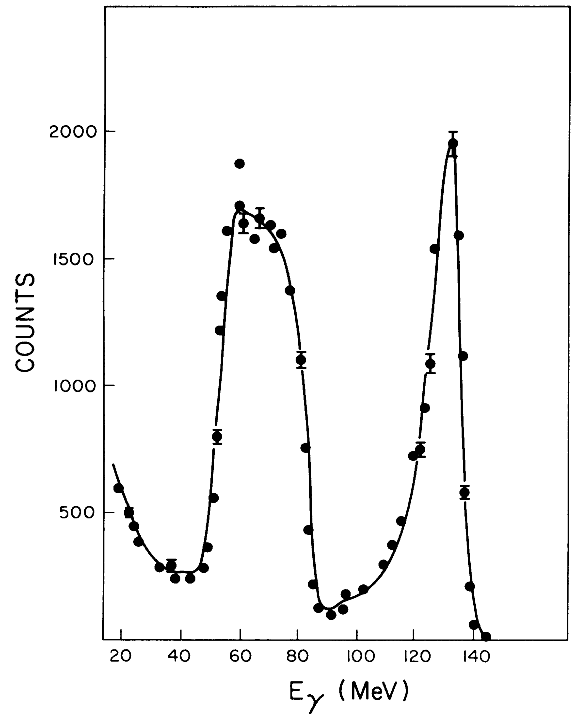

FIG. 8. Fitted spectrum for the 50-atm HD target using the measured spectral shapes shown in Figs. 4—6.

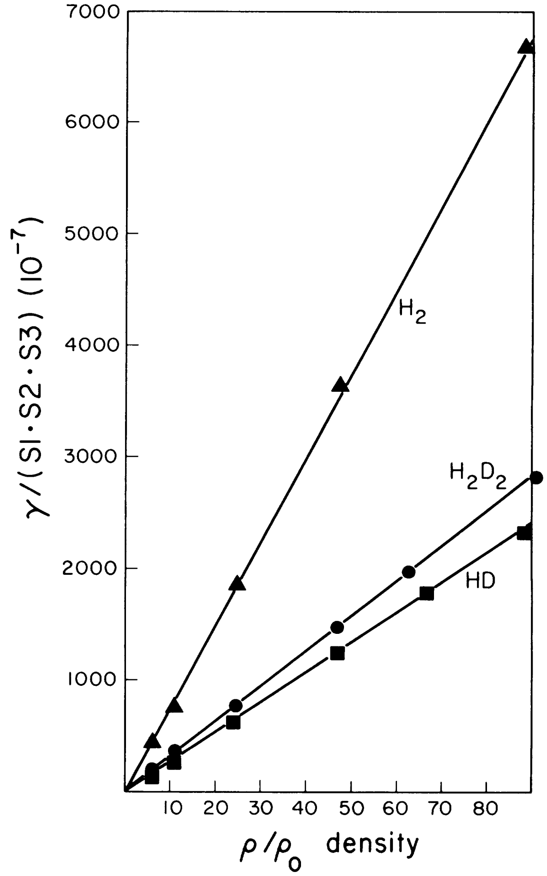

FIG. 9. Yield of $p\pi^-$ γ rays obtained by fitting the region $12 < E_\gamma < 145$ MeV. Symbols are the same as in Fig. 7.

$$f_{H_2D_2} = 0.417 \pm 0.004 ,$$

and

$$M^m = 1.175 \pm 0.008 .$$

Both the above methods rely on using the stop cut. If this cut is not used, the background becomes more troublesome, but still the unique π^0 signature allows this strong $p\pi^-$ absorption signal to be used. The yield curves for the π^0 region without the stop cut give for HD,

$$f_{HD}^m = 0.348 \pm 0.005 ,$$

and for $H_2 + D_2$,

$$f_{H_2D_2} = 0.409 \pm 0.006 ,$$

$$M^m = 1.174 \pm 0.019 .$$

Finally, we must make a correction for the H_2 and D_2 components in our HD gas. We assume that the measured f_{HD}^m values are a weighted average of the probabilities of the π^- landing on molecules HD or H_2 or D_2.

Let P_x be the fractional content of component x in the HD target. From analysis of the target we had

$$P_{HD} = 0.93 \pm 0.02 ,$$

$$P_{H_2} = \tfrac{2}{3}(1 - P_{HD}) ,$$

$$P_{D_2} = \tfrac{1}{3}(1 - P_{HD}) .$$

Using the rate diagrams from Fig. 10 (to be discussed later) we deduce that our measured value f_{HD}^m is related to the true values f_{HD} and $f_{H_2D_2}$ by

$$f_{HD}^m = P_{HD} f_{HD} + \tfrac{1}{3}(1 - P_{HD}) 2 f_{H_2D_2} + \tfrac{1}{3}(1 - P_{HD}) \left[\frac{\lambda_p^{\mathrm{mol}}}{\lambda_{p\pi} + \lambda_p^{\mathrm{mol}}} + \frac{\lambda_{p\pi}}{\lambda_{p\pi} + \lambda_p^{\mathrm{mol}}} \frac{\lambda_p^{\mathrm{at}}}{\lambda_{pd} + \lambda_p^{\mathrm{at}}} \right] . \tag{6}$$

Using the results of Ref. 13, the factor

$$\left[\frac{\lambda_p^{\mathrm{mol}}}{\lambda_{p\pi} + \lambda_p^{\mathrm{mol}}} + \frac{\lambda_{p\pi}}{\lambda_{p\pi} + \lambda_p^{\mathrm{mol}}} \frac{\lambda_p^{\mathrm{at}}}{\lambda_{pd} + \lambda_p^{\mathrm{at}}} \right]$$

lies between 0.85 and 1.0. Therefore, we obtain

$$f_{HD} = 0.338 \pm 0.008 ,$$

$$f_{H_2D_2} = 0.417 \pm 0.004 ,$$

$$M = 1.23 \pm 0.03 .$$

The errors have been added in quadrature.

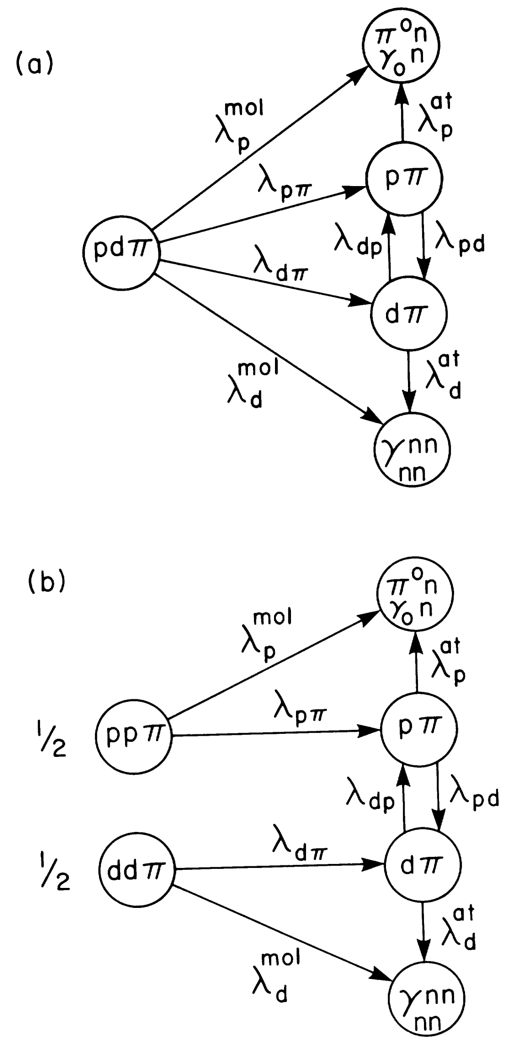

FIG. 10. Schematic rate diagrams for π^- stopping in (a) HD and (b) H_2+D_2.

B. Analysis based on γ_0 yields and neutron-neutron coincidences

We also compared the γ_0 yields, in this case defined to be the region higher in energy than π^0 gammas. Here the aim was simply to check our π^0 results for consistency. The background problem is more severe in this region. After having fitted the hydrogen component and subtracting it from the HD or H_2+D_2 spectra, we have left the D_2 plus background. Now we can compare the yield of the D_2 component in H_2+D_2 and HD to that of the pure D_2 and obtain a number g_x, where g_x is the fraction of pions absorbed on the deuteron. Clearly $f_x+g_x=1$. This procedure worked well for the H_2+D_2 mixture and came up 3% short on the $f_{HD}+g_{HD}$ sum. We attribute this shortfall to difficulty in separating the D_2 γ_0 and background. In particular the slight gain shifts we observed during the long runs affected the γ_0 region more severely than the π^0 region and broadened the γ_0 D_2 peak in a way to imitate the background.

Also by comparing the neutron-neutron coincidence rate between TINA and the NE213 counter we were able to measure the g values independently of the gamma yields. The n-n coincidence arises from the reaction $\pi^- + d \rightarrow n + n$. These values for g also agree with the γ^0 analysis within the somewhat larger errors due to poorer statistics. All the results are presented in Table I.

The 20% difference in the π^0 yield that we observe between HD and H_2+D_2 cannot be attributed to a gaseous contaminant in the HD target. The most abundant atmospheric contaminant that would not freeze or liquefy in the liquid-N_2 cold trap is Ne. Using the transfer data of Petrukhin and Suvorov[11] we would need a concentration of 7% Ne to account for a 20% reduction in the π^0 production in the HD target. However, Ne is normally present in the atmosphere at only 18 ppm. Moreover, the fact that $f_{HD}+g_{HD}$ is very nearly unity shows that those pions that do not get captured by a proton are captured by a deuteron rather than by a contaminant.

IV. DISCUSSION

The observed independence of f_x on the pressure or density of the gases is in agreement with earlier results obtained for gaseous and liquid ethane,[20] and supports the idea of the deexcitation of $p\pi^-$ atoms in collisions with other atoms.[1–5]

The explanation of the fact that $f_{HD} \neq f_{H_2D_2}$ requires an understanding of the processes involved in the earliest stages of pion capture. Detailed treatments[21–23] of the capture of mesons from the continuum into atomic orbits exist. However, except for the semiempirical model[1–3] of large mesic molecules, no theoretical treatment of the meson-molecule collision exists.[21] Day *et al.*[24] suggested that pions can be absorbed from the meso-molecular states of the $(\pi^-, H_2{}^+)$ system, and this process may reduce the pion lifetime in liquid hydrogen. They suggested this process because the nuclear capture rate due to Stark mixing was too slow to explain the observed $p\pi^-$ lifetime in liquid H_2. However, once the external Auger process is included in the $p\pi^-$ deexcitation, the calculated lifetime of the π^- in liquid H_2 is 3.5×10^{-12} sec (Ref. 25) compared to the measured value of $\sim 1.2\times10^{-12}$ sec (Refs. 26–29).

For purposes of discussion we present in Fig. 10 a schematic of the possible processes involved in the capture of π^- mesons. These diagrams are qualitative in that the transfer rates λ_{pd} or λ_{dp} and the nuclear capture rates from the atomic states $\lambda_p^{at}, \lambda_d^{at}$, depend upon the pionic atom states.[5] Unfortunately, the rates λ_{pd} and λ_{dp} have not been calculated. It is known, however, that $p\pi$ atoms formed from compounds Z_mH_n transfer their pions less readily than those formed from H_2 (Refs. 5 and 12) possibly due to a lower level of excitation. Direct formation of the $p\pi$ or $d\pi$ atoms has not been included in the diagram because of the arguments in Ref. 1. Basically, the capture calculations show that the principal quantum number for pion capture from the continuum to excited atomic states is $N \approx 15$. Since the orbits are not localized about one nucleus or the other until $N \approx 7$ in H_2, the initial orbit of the pion is a very large molecular orbit.

TABLE I. (a) Fractions f of pions captured by protons. (b) Fractions g of pions captured by deuterons. (c) Sums $f+g$ of pions captured in the gas targets.

(a)

Method	f^{m}_{HD}	$f_{\mathrm{H_2D_2}}$
π^0 yield stop cut	0.355±0.005	0.416±0.005
Fitted total hydrogen γ yield and stop cut	0.355±0.005	0.417±0.004
π^0 yield no stop cut required	0.348±0.005	0.409±0.006

(b)

Method	g^{m}_{HD}	$g_{\mathrm{H_2D_2}}$
Fitted total deuterium γ yield and stop cut	0.597±0.030	0.592±0.024
Neutron-neutron coincidences plus stop cut	0.593±0.034	0.581±0.024

(c)

Method of determining g	$f^{m}_{\mathrm{HD}}+g^{m}_{\mathrm{HD}}$	$f_{\mathrm{H_2D_2}}+g_{\mathrm{H_2D_2}}$
Fitted total deuterium γ yields and stop cut	0.952±0.030	1.009±0.024
Neutron-neutron coincidences plus stop cut	0.948±0.034	0.998±0.024

In a collision with a gas molecule the pionic atom can either deexcite or transfer the pion to the collision partner. In either case the final pionic atom will be in a lower level of excitation and thus, from the arguments in Ref. 5, a subsequent transfer is less likely. With the assumption that the pion can be transferred only once in the process $d\pi+p\leftrightarrows d+p\pi$ we get for the fractions f,

$$f_{\mathrm{HD}}=\frac{\lambda_p^{\mathrm{mol}}}{\Lambda}+\frac{\lambda_{p\pi}}{\Lambda}\frac{\lambda_p^{\mathrm{at}}}{\lambda_p^{\mathrm{at}}+\lambda_{pd}}+\frac{\lambda_{d\pi}}{\Lambda}\frac{\lambda_{dp}}{\lambda_{dp}+\lambda_d^{\mathrm{at}}}\,,$$

$$\Lambda=\lambda_p^{\mathrm{mol}}+\lambda_{p\pi}+\lambda_{d\pi}+\lambda_d^{\mathrm{mol}}\,, \tag{7}$$

and

$$f_{\mathrm{H_2D_2}}=\frac{1}{2}\frac{\lambda_p^{\mathrm{mol}}}{\lambda_p^{\mathrm{mol}}+\lambda_{p\pi}}+\frac{1}{2}\frac{\lambda_{p\pi}}{\lambda_p^{\mathrm{mol}}+\lambda_{p\pi}}\frac{\lambda_p^{\mathrm{at}}}{\lambda_p^{\mathrm{at}}+\lambda_{pd}}+\frac{1}{2}\frac{\lambda_{d\pi}}{\lambda_{d\pi}+\lambda_d^{\mathrm{mol}}}\frac{\lambda_{dp}}{\lambda_{dp}+\lambda_d^{\mathrm{at}}}\,. \tag{8}$$

At one extreme where we assume that the breakup of the mesic molecule is much faster than the nuclear capture from the molecular state ($\lambda_{d\pi}\gg\lambda_d^{\mathrm{mol}}$ and $\lambda_{p\pi}\gg\lambda_p^{\mathrm{mol}}$) we obtain a ratio,

$$\frac{f_{\mathrm{H_2D_2}}}{f_{\mathrm{HD}}}=\frac{1}{2}\frac{1+r}{P_{p\pi}+(1-P_{p\pi})_r}\,, \tag{9}$$

where

$$r=\frac{\lambda_{dp}}{\lambda_{dp}+\lambda_d^{\mathrm{at}}}\frac{\lambda_p^{\mathrm{at}}+\lambda_{pd}}{\lambda_p^{\mathrm{at}}}$$

and

$$P_{p\pi}=\lambda_{p\pi}/(\lambda_{p\pi}+\lambda_{d\pi})\,.$$

Equation (9) provides a family of curves which depend upon r and $P_{p\pi}$. Several curves are plotted in Fig. 11. The barred region corresponds to the measured M. It is clear that $r<0.71$. Moreover, from the work done on H_2+D_2 mixtures[13] we have obtained the following result:

$$\lambda_p^{\mathrm{at}}/(\lambda_p^{\mathrm{at}}+\lambda_{pd})=0.85\pm0.04\,.$$

Using their[13] value and our measurement of $f_{\mathrm{H_2D_2}}$ in the formula

$$f_{\mathrm{H_2D_2}}=\frac{1}{2}\left[\frac{\lambda_p^{\mathrm{at}}}{\lambda_p^{\mathrm{at}}+\lambda_{pd}}+\frac{\lambda_{dp}}{\lambda_{dp}+\lambda_d^{\mathrm{at}}}\right]\,, \tag{10}$$

we obtain

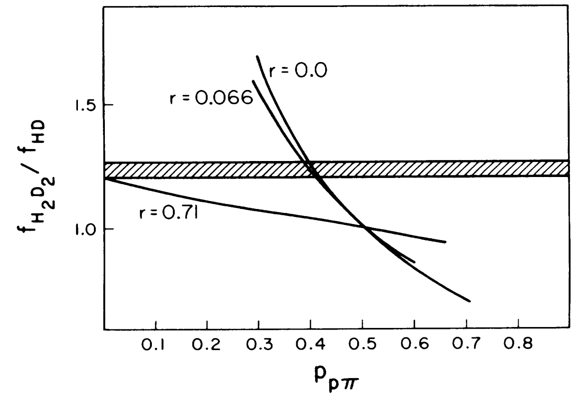

FIG. 11. Ratio $f_{\mathrm{H_2D_2}}/f_{\mathrm{HD}}$ vs P_π for different values of r.

$$\frac{\lambda_{dp}}{\lambda_{dp}+\lambda_d^{\rm at}}=-0.017\pm 0.044$$

or $\lambda_{dp}/(\lambda_{dp}+\lambda_p^{\rm at})<0.062$ at the 90% confidence level. Using this result in Eq. (9) we get

$$0.393\le P_{p\pi}\le 0.407$$

for the corresponding range

$$0.066\ge r\ge 0\ .$$

Thus if the nuclear capture from the molecular levels were negligible compared to the molecular breakup rate

$$1.46<\lambda_{d\pi}/\lambda_{p\pi}<1.54$$

for the range

$$0.066\ge r\ge 0\ .$$

This result is bigger than can be accounted for by the electromagnetic processes proposed to explain the short π^- lifetime in liquid H_2. The radiative transition rates differ by only ~7% (Ref. 25), i.e.,

$$\Gamma_{\rm rad}(\pi pd\rightarrow \pi d+p)/\Gamma_{\rm rad}(\pi pd\rightarrow d+\pi p)=M_{\pi d}/M_{\pi p}\ ,$$

where M_x is the reduced mass. Moreover, this is a slow transition. For example, the rate $(N_1L_1)\xrightarrow[\rm rad]{}(N_2L_2)$ for $(15,1)\rightarrow(1,0)$ is 1.3×10^8/sec, compared to the measured rate of 8.3×10^{11}/sec (Refs. 26–29).

The rate of the external Auger process[25] for the πx atom goes as

$$\Gamma_{\rm Auger}\sim\frac{1}{M_x^2}\frac{1}{(\Delta E_x)^{1/2}}\sim\frac{1}{M_x^{5/2}}\ .$$

If we assume the molecule $p\pi d$ breaks up into $\pi d+p$ and $p\pi+d$ by ejecting an electron from a neighboring HD molecule then

$$\Gamma_{\rm Auger}(p\pi d\rightarrow d\pi+p)/\Gamma_{\rm Auger}(p\pi d\rightarrow p\pi+d)\approx(M_{p\pi}/M_{d\pi})^{5/2}=0.85\ .$$

This effect is even in the wrong direction.

Nuclear absorption from the molecular state could also provide a means of reducing the π^0 production if $\lambda_d^{\rm mol}>\lambda_p^{\rm mol}$ but we note from Eq. (8) that it is not likely that the probability for direct absorption from the molecular states is much greater than that for molecular breakup because $f_{\rm H_2D_2}<\frac{1}{2}$. Because the pion replaces the molecular electrons it must have a good spatial overlap with them. Since the electronic molecular orbitals in HD are to first approximation a sum of atomic 1S orbitals, the amplitudes of NS state πp or πd orbitals should also be large. We can estimate the absorption rates from S states in the πp or πd atoms using the techniques of Ref. 30. The radiative absorption rates are computed using the low-energy radiative capture cross sections.

For $\pi^-+p\rightarrow\gamma_0+n$ we have

$$\lambda_{\gamma_0}(\pi^-+p\rightarrow\gamma_0+n)=V_\pi\sigma(\pi^-+p\rightarrow n+\gamma_0)\times|\phi_{\pi p}(0)|^2\ , \qquad (11)$$

and for $\pi^-+d\rightarrow 2n+\gamma$

$$\lambda_{\gamma_0}(\pi^-+d\rightarrow 2n+\gamma)=r_dV_\pi\sigma(\pi^-+p\rightarrow n+\gamma_0)\times|\phi_{\pi d}(0)|^2\ . \qquad (12)$$

Here $\sigma(\pi^-p\rightarrow n+\gamma_0)$ is the cross section for radiative capture of slow pions of velocity V_π on protons. The quantity $r_d\approx\frac{2}{3}$ accounts for the reduction of the capture cross section in deuterium due to the exclusion principle and n-n forces in the final state.[30] We assume here that nuclear capture occurs from S state.[24] The total rates of capture are, for $\pi^-+p\rightarrow x$

$$\lambda(\pi^-+p\rightarrow x)=(1+P')\lambda_{\gamma_0}(\pi^-+p\rightarrow\gamma_0+n) \qquad (13)$$

and for $\pi^-+d\rightarrow x$

$$\lambda(\pi^-+d\rightarrow x)=(1+S)\lambda_{\gamma_0}(\pi^-+d\rightarrow\lambda n+\gamma)\ , \qquad (14)$$

where P' and S are the Panofsky[31] and S ratios.[16] Using $P'=1.54$ and $S=2.83$, $|\phi_x(0)|^2=1/\pi a_x^3$, a_x is the Bohr radius of the πx atom, the ratio of total capture rates from S states of the same principal quantum number N is

$$[\lambda(\pi^-+d\rightarrow x)/\lambda(\pi^-+p\rightarrow x)]_{NS}=1.22\ . \qquad (15)$$

We can estimate the absolute rate of nuclear capture from NS states for the $\pi^-+p\rightarrow x$ reaction using the radiative capture cross sections of 131×10^{-30} cm^2 at 27.4-MeV pion kinetic energy.[32] The result is

$$\lambda(p+\pi^-\rightarrow x)=1.1\times 10^{15}/N^3\ \text{sec}^{-1}\ . \qquad (16)$$

If the nuclear capture from the molecular states $\lambda_d^{\rm mol}$ and $\lambda_p^{\rm mol}$ have the same relation to each other as the nuclear capture rates from the NS atomic states, given in Eq. (15), then $\lambda_d^{\rm mol}>\lambda_p^{\rm mol}$. Moreover, from Eq. (16) the capture rate from the $N=15$ S state is 3.3×10^{11}/sec, which is of the same order of magnitude as the collision rate of a $p\pi d$ complex of kinetic energy 1 eV with HD molecules at 100 atm pressure.

It must be mentioned, however, that the available experimental information does not seem to support a considerable role of the nuclear capture of pions from molecular orbits. Pionic x-ray transitions to the 1S state can be observed in various atoms including deuterium.[33] In He π^- atoms only 69% of the pions are absorbed by the nucleus from orbits with $N\ge 2$ (Ref. 33). And, finally, the experimental data on pion capture by protons in mixtures of hydrogen and methane with other gases show a negligible role of pion capture from molecular orbits as compared to the collisional processes.[34]

Clearly, to say something definite about this possibility a three-body calculation of the large $p\pi d$ system is needed. Such a calculation would also enable the importance of the molecular breakup rates $\lambda_{d\pi},\lambda_{p\pi}$ to be determined. The description must also explain the pressure independence of the fractions $f_{\rm H_2D_2}$ and $f_{\rm HD}$.

V. SUMMARY

The influence of molecular structure on the fraction of stopped pions that are captured by protons has been observed by comparing the π^0 production in HD gas to that in H_2+D_2 gas. A greater number of π^0 are observed in the gas mixture than in the molecular (HD) form. Neither of the electromagnetic processes used to explain the $p\pi$ atom lifetime in liquid H_2 describe the results we have obtained in HD. Nuclear capture directly from the molecular state $(p\pi d)$ may partially explain the effect, but breakup into $p\pi+d$ or $p+d\pi$ channels also is required. A multibody meson-molecule treatement needs to be done to assess the importance of the various modes involved in the nuclear absorption of the π^- in hydrogen isotopes.

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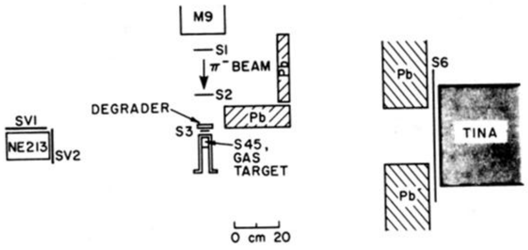

FIG. 1. Experimental setup, as described in the text.