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Pion transfer from hydrogen to deuterium in H_2O+D_2O mixtures

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The transfer of negative pions captured by hydrogen to deuterium has been studied in H_2O+D_2O mixtures. The D_2O-concentration dependence of the transfer rate was analyzed on the basis of a phenomenological model in order to determine the transfer parameters. The fit gives an asymptotic transfer of about 42%, which is smaller than the previous results, but slightly larger than the value for H_2+D_2 gas mixtures. A mesomolecular effect of HOD molecules was discussed on the basis of the concentration dependence of the transfer rate.

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The process involved in the slowing down and capture of negative pions in matter has been discussed in detail by Ponomarev [1] and Horvath [2]. In hydrogen-containing molecules or mixtures, a pion initially captured in the hydrogen is often transferred to one of the neighboring higher-Z atoms and then absorbed by the nucleus. On the other hand, the pionic hydrogen, π^-p, undergoing no transfer is signaled by a π^0 from the charge-exchange reaction $\pi^-+p\rightarrow\pi^0+n$ (branching ratio=60%). In simple gaseous systems, such as H_2+Z, the transfer occurs through the following collision of the π^-p atom with a higher-Z atom:

$$\pi^-p+Z\rightarrow\pi^-Z+p\ .$$

Regarding hydrogenic compounds, Jackson *et al.* [3,4] have proposed an additional mechanism, in which the pion tunnels along a Z—H bond, as well as the above-mentioned external transfer. However, no complete understanding has yet been attained for the entire mechanism of the atomic capture and successive transfer of pions.

The external transfer phenomenon has been systematically studied in gas mixtures of H_2+Z by measuring the Z-concentration dependence of the pion charge-exchange probabilities [5–8]. The experimental results have shown that, except for deuterium, the transfer probability exceeds 90% at large concentrations. The H_2+D_2 system is a special case because both the pionic hydrogen (π^-p) and pionic deuterium (π^-d) are neutral and, hence, move freely through other atoms and molecules. The transfer process, $\pi^-p+d\rightarrow\pi^-d+p$, is expected to be much smaller than for heavier-Z atoms, and the inverse transfer reaction, $\pi^-d+p\rightarrow\pi^-p+d$, is also thought to occur. Experimentally, the pion transfer in H_2+D_2 gas mixtures was first measured by Petrukhin and Prokoshkin [6]. A recent measurement performed at TRIUMF [9] showed that the transfer probability was (32±3 %) at large D_2 concentrations.

In H_2O+D_2O mixtures, most of the pions stopping in the mixture are captured in the oxygen atom due to its large Z. If a pion is initially captured by the hydrogen atom, one of the following processes takes place: (i) transfer to an oxygen atom, (ii) transfer to a deuterium atom, or (iii) nuclear absorption on the proton. Process (i) has the largest probability among them. The pion transfer of hydrogen to deuterium [process (ii)] can still be observed due to the D_2O-concentration dependence of π^0 decays, since the concentration of oxygen remains unchanged.

Petrukhin, Risin, and Suvorov [7] studied this system, finding little difference between H_2O+D_2O and H_2+D_2 mixtures. This led to the conclusion that the difference in the π^-p and π^-d, which results in the transfer of interest, does not appear in collisions of π^-p and π^-d with oxygen atoms. A recent measurement by Stanislaus *et al.* [10], however, showed that the transfer in H_2O+D_2O mixtures is slightly larger than that in H_2+D_2 gas mixtures. This is inconsistent with the fact that a transfer from hydrogen bound with a heavier atom to Z [7,11] has a smaller probability compared to that of H_2+Z mixtures. Based on the assumption that the hydrogen would mostly be in a HOD molecule at large D_2O concentrations, Stanislaus *et al.* stated that the pion preferably selecting the deuterium from the HOD molecule in the breakup results in an enhancement of the transfer probability in H_2O+D_2O mixtures.

This type of study will provide important information concerning the relation between the transfer process and a large mesomolecular state. Unfortunately, the previous data had relatively large uncertainties in determining the transfer mechanism. Further precise measurements are necessary to further elaborate the discussion given above. We therefore measured the D_2O-concentration dependence of the charge-exchange reaction in the H_2O+D_2O

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system in order to determine more precise parameters of the transfer. In our measurement, improvements were made in the counting statistics and in concentration range; further, the capture rates in the mixtures were determined by the Balmer series pionic x-ray intensities from the pionic oxygen in addition to stop events from a counter telescope.

The experiment was performed in the $\pi\mu$ channel of 12-GeV-proton synchrotron at the National Laboratory for High Energy Physics (KEK) of Japan [12]. Since the construction and performance of the experimental setup are described elsewhere [13,14] in detail, only an outline is presented here. The momentum, its bite, and intensity of the negative pion beam were 140 MeV/c, $\Delta P/P=\pm 2.5\%$, and typically $1\times 10^4\ \pi^-$/sec, respectively. The pions were decelerated with a graphite degrader in order to stop them in the sample; the incident and stopped pions were counted with a conventional counter telescope. The π^0 decay ($\rightarrow 2\gamma$), which indicates the charge-exchange reaction in $\pi^- p$, was detected using a pair of lead-glass Cherenkov counters. The pionic x rays, which indicate the capture by atoms other than hydrogen and deuterium, were measured with two Ge low-energy photon spectrometers.

The measured samples were H_2O, D_2O, and mixtures of H_2O+D_2O at various concentrations. The sample was contained in a target holder of rectangular shape (50×65 mm^2 in area and 5 or 7.5 mm in thickness) made of beryllium or aluminum, and was mounted in the measuring chamber at an angle of 40° to the beam axis. The chamber was filled with helium gas to reject the interference of air with pionic spectrum for oxygen [14]. Each measurement was continued for 3–8 h at 20 °C and the number of stopped pions was $(3-7)\times 10^7 \pi^-$ per run.

The event of a pion capture by hydrogen in a sample was assigned based on the energy (about 70 MeV) and on the γ-ray trajectory reproduced from the detection positions of the detectors as described elsewhere [13]. The background two-γ-ray signals, which originated from fast π^- reactions and stop events in the plastic scintillation counter set at the upstream of the target, were evaluated based on measurements for a blank sample and the dependence of the two-γ-ray event on the degrader thickness.

The charge-exchange probability ($W_{\rm H}$) can be obtained from the number of the true two-γ-ray events and stopped pions in the sample, taking account of the detection efficiency of the Cherenkov counters. Here, we determined the relative charge-exchange probabilities (W), normalized to the $W_{\rm H}$ value for H_2O, for each concentration in order to reduce any systematic errors and to be subjected to the successive analysis for the transfer phenomenon.

The obtained W values are listed for the concentration ratio (C) in Table I. Here, C is defined as $C=n_d/n_p$, where n_d and n_p are the numbers of deuterium and hydrogen atoms, respectively. Even when the concentration of D_2O changes, the transfer rate from hydrogen to oxygen is unaltered, since the oxygen concentration remains constant. It follows that the observed concentration dependence of W is ascribed only to the pion transfer from hydrogen to deuterium. Figure 1 shows the dependence of W on the D_2O concentration [$C_{\rm D}$, where $C_{\rm D}=C/(1+C)$].

TABLE I. Relative charge-exchange probabilities (W) and transfer probabilities $[1-(1+C)W]$ as a function of the concentration ratio (C).

C	W	$1-(1+C)W$
0.0827	0.881±0.014	0.046±0.015
0.0989	0.864±0.013	0.050±0.014
0.232	0.751±0.012	0.075±0.014
0.395	0.617±0.011	0.140±0.015
0.595	0.512±0.010	0.183±0.015
0.793	0.446±0.008	0.201±0.015
0.992	0.385±0.008	0.233±0.015
1.239	0.340±0.007	0.238±0.016
1.639	0.264±0.006	0.305±0.016
2.460	0.193±0.005	0.332±0.017
3.264	0.156±0.005	0.336±0.020
4.236	0.122±0.004	0.359±0.023
7.850	0.075±0.003	0.342±0.025

We made reasonable assumptions that the capture rate of H_2O is the same as that of D_2O and that the small binding-energy difference between the $\pi^- p$ and $\pi^- d$ atoms does not appear in collisions with the oxygen atoms. According to a phenomenological model [6], the W can be expressed by the formula $W=PQR$, where P, Q, and R are the probabilities that the pion is captured into a molecular orbit localized near the proton, that the pion makes a transition from the mesomolecular orbit to the $\pi^- p$ atomic level, and that the pion is retained by the proton in a collision, rather than being transferred to heavier atoms, respectively. We can treat this formula as including the parameters concerning only hydrogen, since the contribution of oxygen is canceled by normalizing to W_H for H_2O. Here, if there is no difference in the transition from a mesomolecular to the atomic orbit

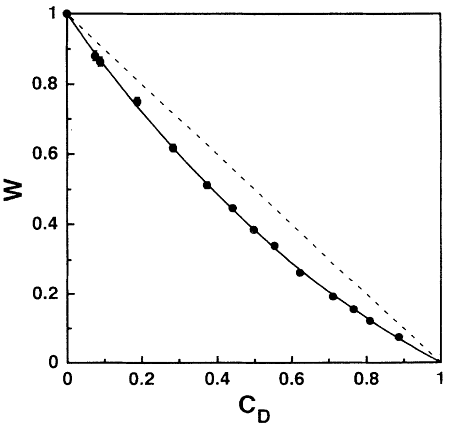

FIG. 1. Relative charge-exchange probabilities as a function of the D_2O concentration ($C_{\rm D}$) in a H_2O+D_2O mixture. The dashed line presents the expected relative charge-exchange probabilities if there was no transfer. The solid curve is the fit of the phenomenological model [Eq. (1)].

and/or in the isolation process of $\pi^- p$ among the H_2O, D_2O, and HOD systems, the probability (Q) should be set to unity. Then, the W could be expressed by

$$W=\frac{1}{1+C}\frac{1+\kappa C}{1+\kappa C+\Lambda C}\ , \tag{1}$$

where κ is the ratio of nuclear capture probability in the $\pi^- p+d$ to $\pi^- p+p$ collisions, and Λ is the ratio of the transfer probability in the $\pi^- p+d$ collision to the nuclear capture probability in the $\pi^- p+p$ collision.

By fitting Eq. (1) to the experimental results, we determined the parameters as $\Lambda=0.52\pm0.04$ and $\kappa=0.73\pm0.11$ (reduced $\chi^2=0.84$). In Table II the results are compared with the previous results for H_2O+D_2O mixtures [10] and H_2+D_2 gas mixtures [9]. Both the Λ and κ values are different from the previous results for the H_2O+D_2O system [10]. An asymptotic transfer probability at large concentrations is given as the ratio $\Lambda/(\Lambda+\kappa)$. Our asymptotic transfer probability ($42\pm4\,\%$), lies between the two previous values for H_2O+D_2O mixtures and H_2+D_2 gas mixtures. We believe that this result is more reliable than the other data, since our experiment was improved in counting statistics and in the range of the concentration ratio ($C=0.083-7.85$). Figure 2 presents the transfer probabilities, $1-(1+C)W$, as a function of C together with the results by Stanislaus *et al.* [10].

The $\pi^- p$ from H—Z is created in a more tightly bound orbit than that from H_2. The transfer rate of the former is therefore expected to be slower than the latter. The present result is consistent with the above-mentioned consideration, since the obtained relative transfer rate ($\Lambda=0.52$) is smaller than $\Lambda=0.65$ for the H_2+D_2 system. The large asymptotic transfer probability obtained is due to the smaller κ value compared to that for the H_2+D_2 system [9].

Then, we will consider the mesomolecular effect of HOD molecules as a cause of discrepancy between the parameters discussed above. For the H_2+D_2 and HD systems, Aniol *et al.* [15] introduced a parameter f [$=W_H(H_2+D_2)/W_H(HD)$], which means the ratio of the charge-exchange probability in the H_2+D_2 system to that in the HD system, so as to include such molecular effects into their model analysis. They obtained experimentally that $f=1.23\pm0.03$, and discussed the molecular effects on isolating the $\pi^- p$ or $\pi^- d$ atoms from the pionic HD system. For the H_2O+D_2O system, the molecular effect has been proposed by Stanislaus *et al.* [10] as a possible mechanism to explain the observed enhancement of transfer in the H_2O+D_2O system compared to that in the H_2+D_2 system.

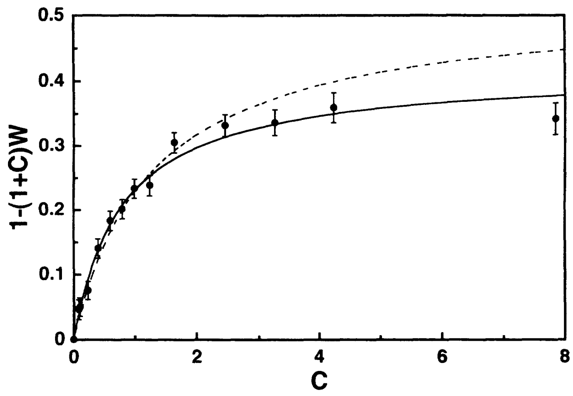

FIG. 2. Transfer probabilities $[1-(1+C)W]$ as a function of C. The solid curve is the fit for the present result, and the dashed curve is the estimate with the previous parameters by Stanislaus *et al.* [10].

Here, we introduce a similar parameter f [$=W_H(H_2O+D_2O)/W_H(HOD)$] in order to discuss the existence of the effect. If molecular effects also exist in the HOD system, the f value would become larger than unity. Assuming that the isotope exchange reaction, $H_2O+D_2O \rightleftarrows 2HOD$, completely attains equilibrium, we can express the concentrations of individual components as $[H_2O]=C_H^2$, $[D_2O]=C_D^2$, and $[HOD]=2C_HC_D$, where $C_H=1-C_D=1/(1+C)$. Providing that only pions captured by hydrogen in HOD molecules are influenced by the transition from the pionic molecular orbit to the atomic orbit of deuterium, we obtain the following simple expression for Q using the parameter f:

$$Q=\frac{2[H_2O]+[HOD]f^{-1}}{2[H_2O]+[HOD]}=\frac{1+Cf^{-1}}{1+C}\ . \tag{2}$$

Equation (1) is then rewritten as

$$W=\frac{1+Cf^{-1}}{(1+C)^2}\frac{1+\kappa C}{1+\kappa C+\Lambda C}\ . \tag{3}$$

Since the similarity of C dependence of the parameters f and κ makes it difficult to obtain convergent results for three parameters, f, κ, and Λ, by fitting Eq. (3) to the experimental results, we can only obtain the preliminary results as follows: $0.7<f<1.6$, $0.12<\Lambda<0.56$, and $0<\kappa<1.6$. In order to test the influence of $f\neq1$ (indicating the existence of the mesomolecular effect), we assumed that the difference between the asymptotic transfer probabilities of the H_2O+D_2O and H_2+D_2 systems is attributed only to the parameter f, and obtained $f=1.17$. The fit of Eq. (3) using the fixed parameter f leads to $\Lambda=0.37\pm0.05$ and $\kappa=0.80\pm0.16$ (reduced $\chi^2=0.83$). The above discussion, however, cannot pro-

TABLE II. Comparison of the parameters from the fit to the phenomenological model.

	System	Λ	κ	$\Lambda/(\Lambda+\kappa)$
Present work	H_2O+D_2O	0.52 ± 0.04	0.73 ± 0.11	0.42 ± 0.04
Stanislaus *et al.* [10]	H_2O+D_2O	0.41 ± 0.05	0.38 ± 0.14	0.52 ± 0.10
Weber *et al.* [9]	H_2+D_2	0.65 ± 0.07	1.40 ± 0.22	0.32 ± 0.03
"World fit" [9]	H_2+D_2	0.45 ± 0.04	0.93 ± 0.14	0.33 ± 0.03

vide evidence that the condition of $f=1.17$ can explain the present results better than that of $f=1$, because both of the cases provided reasonable results in the statistical consideration.

In conclusion, we have obtained improved data for the pion transfer from hydrogen to deuterium in mixtures of H_2O+D_2O. There was no clear evidence that the larger asymptotic transfer probability compared to the H_2+D_2 system was attributed to the molecular effect of the HOD molecule.

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