

Reinterpretation of pion-transfer probabilities in mixtures of H₂O and D₂O

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A model of the capture and transfer of negative pions in mixtures of H₂O and D₂O is described. It is shown that the probability of pion transfer from hydrogen to deuterium that has been extracted from data on charge-exchange probabilities in several recent experiments should be reinterpreted as a combination of the hydrogen-to-deuterium and hydrogen-to-oxygen transfer probabilities. This explains a discrepancy between the results for the pion-transfer probabilities in mixtures of H₂O and D₂O and the transfer probabilities extracted from data on pion capture in mixtures of H₂ and D₂.

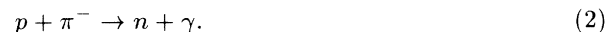
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I. INTRODUCTION

The capture and transfer of negative pions from hydrogen to other atoms have been the subject of many studies in order to elucidate the mechanisms of these processes [1, 2]. Although the initial stages of the capture process have never been observed directly, it is generally believed that when a low energy π^- encounters a molecule it may undergo capture by an Auger process into a pionic molecular orbital [1]. After a series of Auger or radiative transitions the π^- eventually reaches an atomic orbital and then undergoes an atomic cascade before finally reaching an orbital near the nucleus from which nuclear capture can occur. Transfer of the π^- may also occur during the collision of the pionic atom with another atom or molecule. In compounds containing hydrogen these processes can be studied experimentally by means of the charge-exchange reaction that occurs in approximately 60% of nuclear captures of a π^- by a proton [3]:



The π^0 that is produced rapidly decays to two photons which can be conveniently detected in a coincidence measurement. In the remaining 40% of nuclear captures the following reaction occurs:



Apart from the special case of ³He, the cross section for π^0 production on nuclei other than hydrogen is negligible and the observation of a π^0 is thus a unique signal of π^- nuclear capture on a proton.

An especially simple and important system that has been studied in detail is that consisting of mixtures of H₂ and D₂ [4]. At high deuterium concentration it has been shown that there is a significant probability, equal to $(32 \pm 3)\%$, that the π^- will transfer from a pionic hydrogen atom to a deuterium atom. Recently there have been several studies of a related, but more complex system, consisting of mixtures of H₂O and D₂O [5, 6]. In this system there is the possibility of pion transfer from hydrogen to both deuterium and oxygen:



and

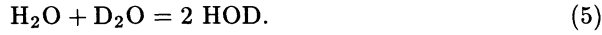


In [5] the pion-transfer probability from hydrogen to deuterium in this system was found to be $(52 \pm 10)\%$ at high concentrations of D₂O and this result appears to have been confirmed by a recent repetition of the same experiment with better statistics which found a transfer probability of $(42 \pm 4)\%$ [6]. Both these results are surprisingly large given that pion-transfer probabilities are expected to be an increasing function of the nuclear charge of the atom to which transfer occurs [2, 7, 8]. Since the nuclear charge for oxygen is greater than that for deuterium, the transfer probability to oxygen should be larger than that to deuterium and one would therefore expect the transfer probability from hydrogen to deuterium in mixtures of H₂O and D₂O to be smaller than that in mixtures of H₂ and D₂, in conflict with the above observations. In both [5] and [6] it was suggested that the high transfer probability to deuterium might be due to the π^- in a HOD molecule preferentially selecting the deuteron in the molecular breakup. The analysis in both experiments, however, was based on the assumption that pion transfer to oxygen could be ignored since the measured charge-exchange probabilities were normalized to the charge-exchange probability for pure water. The relation between the charge-exchange probability and the transfer probability from hydrogen to deuterium in the H₂O-D₂O system was therefore assumed to be the same as that in the H₂-D₂ system. In this article we present a derivation of the relation between the charge-exchange probability and the transfer probabilities in this system which shows that, even after normalizing to pure water, the transfer probability to oxygen enters directly into the relation between the measured charge-exchange probability and the transfer probability to deuterium. This leads to a fundamental reinterpretation of the the transfer probability extracted from the experimental data which resolves the apparent discrepancy between the results for the H₂-D₂ system and the H₂O-D₂O system.

II. THEORY

We present below a derivation of the relation between the charge-exchange probability and pion-transfer probabilities in a mixture of H₂O and D₂O, based on the simple

model of pion capture and transfer outlined in [1]. Initially, the molecular composition of the mixture will be assumed to be in the ratio C molecules of D_2O to one molecule of H_2O . The H_2O and D_2O molecules can react to produce HOD according to



At equilibrium, the relative numbers of H_2O , HOD, and D_2O molecules will be in the ratio $1:2C:C^2$. According to [1], the probability of nuclear capture on a hydrogen nucleus in such a mixture is given by

$$W(C) = P_{H_2O}(C)A_{H_2O}B(C) + P_{HOD}(C)A_{HOD}B(C), \quad (6)$$

where $P_{H_2O}(C)$ and $P_{HOD}(C)$ are the probabilities that the pion undergoes molecular capture on a H_2O molecule or HOD molecule, A_{H_2O} and A_{HOD} are the conditional probabilities that the pion undergoes atomic capture on hydrogen, given that it is captured into a H_2O molecule or HOD molecule, and $B(C)$ is the conditional probability that the pion undergoes nuclear capture on the hydrogen nucleus, given that it is captured into an atomic orbital on hydrogen. The above equation for $W(C)$ is based on the assumption that pion transfer from deuterium to hydrogen is negligible in comparison to transfer from hydrogen to deuterium and the same assumption was also made in the analysis of the data on pion capture in both mixtures of H_2 and D_2 [4] and mixtures of H_2O and D_2O [5, 6]. The excess binding energy released by the formation of a $d\pi$ atom in reaction (3) tends to drive the transfer from left to right [9, 4]. If it is also assumed that H_2O , HOD, and D_2O behave identically as far as molecular capture is concerned, then the initial molecular capture probabilities $P_{H_2O}(C)$ and P_{HOD} are given by

$$P_{H_2O}(C) = \frac{1}{(1+C)^2} \quad (7)$$

and

$$P_{HOD}(C) = \frac{2C}{(1+C)^2}. \quad (8)$$

The probabilities A_{H_2O} and A_{HOD} depend only on factors that are internal to the H_2O or HOD molecules and therefore do not depend on the concentration C . It seems reasonable, as a first approximation, to assume that $A_{H_2O} = 2A_{HOD}$ since there are two hydrogen atoms in the H_2O molecule and only one in the HOD molecule. Once a pion is in an atomic orbital on a $p\pi$ atom, it can do one of four things before undergoing nuclear capture: transfer to another hydrogen atom, leaving the system essentially unchanged, transfer to a deuterium atom, transfer to an oxygen atom, or decay. The decay rate is very slow relative to the rates for pion transfer or nuclear absorption and this process can therefore be ignored. Hence the transfer probability can be written as

$$B(C) = 1 - Q_D(C) - Q_O(C), \quad (9)$$

where $Q_D(C)$ is the conditional probability that the pion undergoes transfer to a deuterium atom, given that the pion is in a $p\pi$ atom, and $Q_O(C)$ is the conditional probability that the pion undergoes transfer to an oxygen

atom, given that the pion is in a $p\pi$ atom. The quantity $Q_O(C)$ is nearly independent of C since the oxygen atom concentration remains the same, independent of the deuterium concentration. However, a weak C dependence of $Q_O(C)$ arises through the processes of nuclear capture and transfer of pions in collisions of $p\pi$ atoms with deuterium which modify $Q_O(C)$. This can be seen more clearly from the expression for $Q_O(C)$ in terms of the rates for the fundamental processes that the $p\pi$ atom can undergo

$$Q_O(C) = \frac{\lambda_{pO}c_O}{\beta_{pp}c_p + \beta_{pd}c_d + \beta_{pO}c_O + \lambda_{pd}c_d + \lambda_{pO}c_O}, \quad (10)$$

where, in a similar notation to that used in [4] for the H_2 - D_2 system, $\beta_{pp}c_p$ is the rate for nuclear absorption in $p\pi + p$ collisions, $\beta_{pd}c_d$ is the rate for nuclear absorption in $p\pi + d$ collisions, $\beta_{pO}c_O$ is the rate for nuclear absorption in $p\pi + O$ collisions, $\lambda_{pd}c_d$ is the rate for pion transfer in $p\pi + d$ collisions, and $\lambda_{pO}c_O$ is the rate for pion transfer in $p\pi + O$ collisions. The quantities c_p , c_d , and c_O are the atomic concentrations of hydrogen, deuterium, and oxygen. The quantity c_O is independent of C and the C dependence of $Q_O(C)$ thus arises solely from the terms involving c_d in the denominator of Eq. (10). Substituting Eqs. (7), (8), and (9) into Eq. (6) gives the result

$$W(C) = \frac{1}{(1+C)} A_{H_2O} [1 - Q_D(C) - Q_O(C)]. \quad (11)$$

In pure water, $C=0$ and $Q_D(0) = 0$. Hence the nuclear capture probability in pure water is

$$W(C=0) = A_{H_2O}[1 - Q_O(0)]. \quad (12)$$

In recent experiments on pion transfer in mixtures of H_2O and D_2O [5, 6] the principal observable was the quantity W_{expt} , the charge-exchange probability at concentration C , normalized to the charge-exchange probability in pure water. Since the nuclear capture branching ratio for reactions (1) and (2) is independent of concentration, W_{expt} is equal to the ratio of the nuclear capture probabilities at concentration C to that at $C = 0$. From Eqs. (11) and (12), W_{expt} is given by

$$W_{\text{expt}} = \frac{W(C)}{W(C=0)} = \frac{1}{(1+C)} \frac{[1 - Q_D(C) - Q_O(C)]}{[1 - Q_O(0)]}. \quad (13)$$

This can be rewritten as

$$W_{\text{expt}} = \frac{1}{(1+C)} [1 - Q(C)], \quad (14)$$

where

$$Q(C) = \frac{Q_D(C) + [Q_O(C) - Q_O(0)]}{[1 - Q_O(0)]}. \quad (15)$$

III. CONCLUSION

Equation (14) is the same relation that was used in the analysis of the results reported in [5, 6] in order to extract the transfer probability $Q(C)$ from the experi-

mental charge-exchange probability normalized to pure water. The quantity $Q(C)$ was interpreted as representing the transfer probability to deuterium in the mixture of H_2O and D_2O and therefore it was assumed to be comparable with the analogous transfer probability measured in mixtures of H_2 and D_2 . The above analysis has shown, however, that $Q(C)$ is a function of both the transfer probability from a $p\pi$ atom to deuterium and the transfer probability to oxygen. If the small term $[Q_{\text{O}}(C) - Q_{\text{O}}(0)]$ is ignored, then $Q(C)$ is just the transfer probability to deuterium divided by the quantity $[1 - Q_{\text{O}}(0)]$ where this latter quantity represents the probability that the pion does not transfer to oxygen in pure water. As discussed above, it is expected that $Q_{\text{O}}(0) \geq Q_{\text{D}}(C)$ at large deuterium concentrations. Hence the quantity $[1 - Q_{\text{O}}(0)]$

should be significantly less than unity and the probability $Q(C)$ is thus expected to be significantly greater than the transfer probability to deuterium, $Q_{\text{D}}(C)$. The apparent anomaly, reported in [5, 6], that $Q(C)$ is greater than the transfer probability, $Q_{\text{D}}^{\text{H}_2-\text{D}_2}(C)$, measured in mixtures of H_2 and D_2 [4], can therefore be explained simply by the fact that $Q(C)$ and $Q_{\text{D}}^{\text{H}_2-\text{D}_2}(C)$ represent quite different physical quantities. This conclusion holds irrespective of whether or not the difference $[Q_{\text{O}}(C) - Q_{\text{O}}(0)]$ is negligibly small in relation to $Q_{\text{D}}(C)$.

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