Transfer Mechanisms for Pionic Hydrogen in Organic Liquids

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The transfer of negative pions in mixtures of bromodecane and carbon tetrachloride has been investigated by combined measurements of π^0's produced from nuclear capture on hydrogen and pionic x rays following Coulomb capture on bromine and chlorine. A significant fraction of pion transfer occurs via an external-transfer process. The results represent the first direct observation of transfer in condensed matter.

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In recent years a considerable quantity of data has been acquired on the capture of negative pions in molecular systems. Despite this, a number of important questions regarding the molecular interactions of pions remain unanswered. Chief among these are the distribution of pions within the molecule immediately following molecular capture, and the nature and significance of subsequent processes involving transfer of pions from hydrogen to atoms of higher nuclear charge. The study of transfer processes involving pionic hydrogen is useful in understanding similar processes for muonic atoms. This is especially significant because pion transfer occurs only from excited states of the exotic atom, the analogous process of muon transfer from excited states being critical in muon-catalyzed fusion.

Experiments which measure charge-exchange (π^-,π^0) probabilities in simple systems such as mixtures of hydrogen with noble or other gases suggest that a significant proportion of pions which are initially captured on hydrogen are subsequently transferred to higher-Z atoms before nuclear capture occurs.[1,2] Such a transfer is believed to take place by a process in which the small neutral $p\pi^-$ atom first breaks away from the hydrogen molecule (in which "atomic" capture initially occurs) and then transfers the pion to a higher-Z atom in a subsequent collision,[3,4] i.e., $p\pi^- + Z \rightarrow Z\pi^- + p$. The large increase in binding energy once the π^- is transferred to a higher-Z atom ensures that the transfer is essentially one-way. This process is called an "external-transfer" process if the pion is transferred outside of the molecule onto which it was originally captured.

In more complex molecular systems (e.g., organic compounds) more involved mechanisms come into play. Whether the pion undergoes atomic capture on hydrogen instead of on other atoms in the molecule depends on the local electronic wave functions, but only the broad principles are understood. The details of the subsequent pion transfer process are even less well understood and there is dispute about the nature of the transfer mechanism itself. It has been suggested[5,6] that an alternative pion transfer mechanism is important in these systems. In this mechanism the π^- is transferred to a higher-Z atom within the same molecule, essentially by tunneling of the pion along a Z-H bond. Alternatively, the neutral $\pi^- p$ system may move to other atoms in the same molecule. Such processes may be said to be "internal-transfer" processes. Unfortunately, the very large amount of data on charge-exchange probabilities in hydrogen-containing molecules, present in the solid or liquid state, do not allow one to determine the mechanism of pion transfer unambiguously because of the lack of knowledge about the relative initial capture probabilities of pions onto the different atoms within the molecule.

So far only one experiment[7] has attempted to study pion transfer in molecular systems in the condensed phase in a direct way, by measuring pion charge-exchange probabilities in mixtures of hydrogenous compounds with nonhydrogenous ones. The observation of a π^0 is a clean signature that the pion underwent atomic, and subsequently nuclear, capture on hydrogen, as the charge-exchange probability for all other nuclei (except ^{3}He) is strongly suppressed. However, in order to extract estimates of pion transfer parameters, an assumption had to be made relating the pion capture probabilities in the two kinds of molecules (in this case benzene and carbon tetrachloride) to the stopping power of the two molecules. Molecular capture occurs at much lower pion energies than the energies at which stopping powers are normally measured, and it has been observed[8] that capture probabilities do not always scale with the stopping power. Thus the analysis based on this assumption is questionable, and it is conceivable that the effect observed was due to a concentration dependence of the partitioning of the pion among the components of the mixture, and not due to pion transfer at all. Furthermore, the possibility of internal transfer was not allowed for in the analysis.

In the present work the aim is to remove the ambiguity concerning the π^- stopping probabilities, by monitoring π^- captures on high-Z atoms using pionic x-ray signals at the same time as measuring pion capture on hydrogen using the standard method of detecting π^0's. We

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have thus chosen to study pion capture in mixtures of bromodecane [$CH_3(CH_2)_9Br$] and carbon tetrachloride (CCl_4), at different molecular concentrations. Pionic x rays from bromine and chlorine provide a signal of stopping pions on the two kinds of molecules. Thus knowing the molecular capture probabilities, the data on charge exchange on hydrogen allow the internal- and external-transfer processes to be examined in more detail.

The experiment was performed in the M13 low-energy pion channel at TRIUMF. The π^- beam with an incident momentum of 78 MeV/c passed through three beam scintillation counters S_1, S_2, and S_3 and stopped in the target. The π^0 detection technique was very similar to the one described in detail in Ref. 9. Two large NaI detectors TINA (46 cm diam×51 cm) and MINA (36 cm diam×36 cm) were used to detect, in coincidence, the two γ rays from the π^0 decay. The π^0 detection system had an efficiency of $\sim 10^{-3}$. A π^0-related event was signaled by a coincidence of $S_1 \cdot S_2 \cdot S_3 \cdot$TINA$\cdot$MINA. The absolute pion capture probability in hydrogen, $W_{\rm H}$, for each of the targets was calculated by normalizing to that for CH_2, which was taken to be $(12.9 \pm 1.8) \times 10^{-3}$, the weighted mean of results from three independent measurements.[10-12] Pionic x rays were detected using an intrinsic germanium detector of 50-cm^3 active volume, kept at a distance of 17.5 cm from the target, covering a solid angle of $\sim 7 \times 10^{-3}$. The detector had an intrinsic resolution of 2.5 keV at 1.3 MeV (1.9 keV at 200 keV). All the targets used in this experiment were mixtures of bromodecane and carbon tetrachloride, at different relative concentrations. They were contained in stainless-steel target holders, 13.3 cm diam×1.55 cm, with 0.025-mm-thick windows. The fraction of incident pions stopping in the target windows was determined by measuring the pion stops as a function of target thickness; less than 0.5% of the beam stopped in the front or back windows.

We shall now consider the capture of pions in a target containing a mixture of n_α molecules of a hydrogen-containing molecule denoted α (bromodecane) and n_β molecules of a molecule β (CCl_4) which contains no hydrogen. To settle the problem of relative capture probabilities in the two different molecules, the ratio of bromine $4f \to 3d$ to chlorine $3d \to 2p$ x-ray transition intensity was measured for several CCl_4 concentrations. The Cl $3d \to 2p$ peak was centered at 150.7 ± 0.2 keV in agreement with previous measurements. The Br $4f \to 3d$ peak was centered at 223.5 ± 0.2 keV, which is reported here for the first time in the literature. In Fig. 1 the ratio $(N^{\rm Cl}_{3d\to 2p}/N^{\rm Br}_{4f\to 3d})/C$, where $C = n_\beta/n_\alpha$, is plotted as a function of C, and is seen to be constant. This behavior has been observed in solids,[13] but in mixtures of noble gases nonlinear effects have been observed.[14] The x-ray intensity ratio can be written as

$$\frac{N^{\rm Cl}_{3d\to 2p}}{N^{\rm Br}_{4f\to 3d}} = \frac{\epsilon_{\rm Cl}P_\beta P_{\rm Cl}P_{3d\to 2p}}{\epsilon_{\rm Br}P_\alpha P_{\rm Br}P_{4f\to 3d}} = M(\beta,\alpha)\frac{\epsilon_{\rm Cl}P_{\rm Cl}P_{3d\to 2p}}{\epsilon_{\rm Br}P_{\rm Br}P_{4f\to 3d}}C\,, \qquad (1)$$

where P_β is the capture probability on a β molecule, $P_{\rm Cl}$ is the probability of a π^- going to an atomic orbital of chlorine (given that a π^- is captured on a β molecule), $P_{3d\to 2p}$ is the probability of a $3d \to 2p$ pionic x-ray transition (given that a π^- is captured on chlorine), and $\epsilon_{\rm Cl}$ is the efficiency of the detector for $3d \to 2p$ x rays. The terms P_α, $P_{\rm Br}$, $P_{4f\to 3d}$, and $\epsilon_{\rm Br}$ are defined in a similar manner. We make the assumption that the pionic x-ray transition probabilities $P_{4f\to 3d}$ and $P_{3d\to 2p}$ are the same for pions initially captured on Br and Cl as for pions transferred to Br and Cl. $M(\beta,\alpha)$ is defined by $P_\beta/P_\alpha = M(\beta,\alpha)C$, i.e., it is the capture ratio on β molecules to α molecules in a 1:1 mixture. From Fig. 1 we can conclude that $M(\beta,\alpha)$, $P_{\rm Br}$, and $P_{4f\to 3d}$ (and likewise $P_{\rm Cl}$ and $P_{3d\to 2p}$) are concentration independent. Thus the ratio of pion captures on molecules of different kinds in a mixture is proportional to the relative numbers of these molecules. This is critical information for the interpretation of the π^- transfer from hydrogen to higher-Z atoms, inferred from the concentration dependence of the π^0 yield.

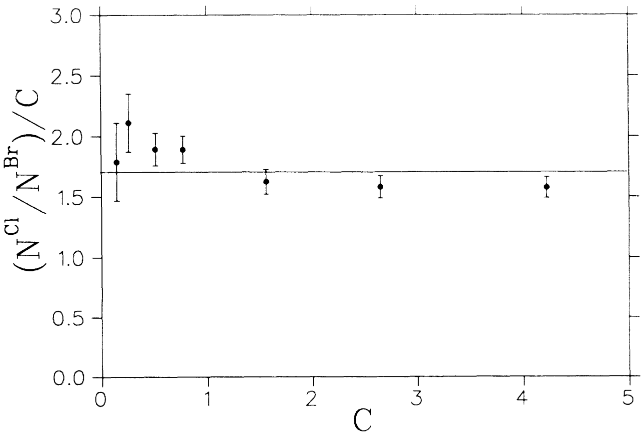

FIG. 1. X-ray ratio $(N^{\rm Cl}_{3d\to 2p}/N^{\rm Br}_{4f\to 3d})/C$ as a function of the concentration C.

The pion capture probability for the mixture can be written as

$$W_{\rm H} = P_\alpha P_{\rm H}(1 - T_T)\,, \qquad (2)$$

where $P_{\rm H}$ is the probability, given that capture occurs on an α molecule, that the π^- goes to a hydrogen atomic orbital, and T_T is the total transfer probability of a π^- from hydrogen to other (higher-Z) atoms. The concentration independence of $M(\beta,\alpha)$ means that the capture probability on molecule α can be expressed as

$$P_\alpha = \frac{1}{1 + (n_\beta/n_\alpha)M(\beta,\alpha)}\,. \qquad (3)$$

In principle, pions can transfer from hydrogen to higher-Z atoms in several different ways. They may transfer to atoms within the same molecule (internal transfer), the probability for which will be denoted T_i. In addition, they may be transferred to atoms in different α molecules (transfer probability T_α) or to atoms in β molecules (transfer probability T_β). Therefore T_T can be written $T_T=T_i+T_\alpha+T_\beta$. The internal transfer probability T_i should be independent of the molecular ratio C, whereas T_α and T_β are expected to depend (in different ways) on the molecular ratio. In the simplest approximation, the external-transfer probability to a molecule of a particular kind is proportional to the mole fraction for that molecule in the mixture. Therefore we have $T_\alpha=A_\alpha n_\alpha/(n_\alpha+n_\beta)$ and $T_\beta=A_\beta n_\beta/(n_\alpha+n_\beta)$, where A_α and A_β are transfer constants for α and β molecules, respectively. Together with (2) and (3), W_H becomes

$$W_H=\frac{P_H}{1+M(\beta,\alpha)C}\left[1-T_i-\frac{A_\alpha}{1+C}-\frac{A_\beta}{1+1/C}\right], \quad (4)$$

where P_H is assumed to be independent of the molecular ratio C.

We can consider two limiting cases of (4). In the limit of $C\to 0$, (4) reduces to $W_H=P_H(1-T_i-A_\alpha)$, which is the required expression for the charge-exchange probability in a system composed purely of hydrogenous α molecules. In the limit that $C\to\infty$, (4) shows that $W_H\to 0$, as required for the charge-exchange probability for a target composed only of β molecules. For arbitrary concentrations C, the formula for W_H depends on five parameters, three of which (T_i,A_α,A_β) describe the different transfer processes from hydrogen while the remaining two parameters $[M(\beta,\alpha),P_H]$ describe the initial capture process: first onto α molecules and then onto hydrogen atoms.

The concentration dependence of the W_H values is shown in Fig. 2(a) and exhibits a steady decrease with increasing concentration. However, since the concentration dependence of W_H is present not only in the transfer term [cf. Eq. (4)], the observed concentration dependence cannot be interpreted unambiguously as evidence for external transfer.

By using combined information from the π^0 intensity and pionic x-ray intensity the number of unknown parameters can be reduced. Using the above notation, the ratio of the pion charge-exchange probability (W_H) to the number of bromine $4f\to 3d$ transitions ($N^{Br}_{4f\to 3d}$) can be written as

$$\frac{W_H}{N^{Br}_{4f\to 3d}}=\frac{P_\alpha P_H(1-T_i-T_\alpha-T_\beta)}{\epsilon_{Br}P_\alpha P_{Br}P_{4f\to 3d}}. \quad (5)$$

Using the same approach for the charge-exchange probability as for the derivation of (4) leads to

$$\frac{N_{\pi^0}}{N^{Br}_{4f\to 3d}}=\epsilon\left[1-T_i-\frac{A_\alpha}{1+C}-\frac{A_\beta}{1+1/C}\right], \quad (6)$$

where the factor ϵ is defined by $\epsilon=\epsilon_{\pi^0}P_H/\epsilon_{Br}P_{Br}P_{4f\to 3d}$, and ϵ_{π^0} is the π^0 detection efficiency ($N_{\pi^0}=\epsilon_{\pi^0}W_H N_{\pi^-}$).

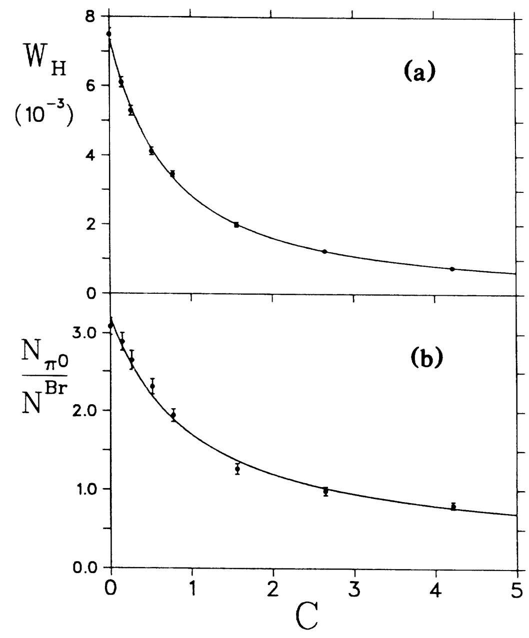

FIG. 2. (a) Pion capture probability on hydrogen W_H as a function of the concentration C. (b) $N_{\pi^0}/N^{Br}_{4f\to 3d}$ as a function of the concentration C. In each case the line is a guide to the eye.

By inference from the probabilities P_{Br} and $P_{4f\to 3d}$ being independent of the concentration, the factor ϵ is also concentration independent. The only concentration-dependent terms are those involving the two external-transfer terms A_α and A_β. The observable $N_{\pi^0}/N^{Br}_{4f\to 3d}$ is thus expressed in terms of four unknowns: ϵ, and the three transfer parameters T_i, A_α, and A_β. Supposing the external-transfer processes to have negligible probability, then $A_\alpha\simeq A_\beta\simeq 0$ and the ratio $N_{\pi^0}/N^{Br}_{4f\to 3d}$ would be concentration independent. On the other hand, if the terms involving external transfer are significant, then the ratio $N_{\pi^0}/N^{Br}_{4f\to 3d}$ will be concentration dependent. A plot of the $N_{\pi^0}/N^{Br}_{4f\to 3d}$ values is shown in Fig. 2(b) and exhibits a steady decrease with increasing concentration, similar to Fig. 2(a). As discussed above, the observation of a concentration dependence of this ratio provides a clear indication that a significant amount of external transfer is taking place.

Equation (6) can be rearranged to give

$$\frac{N_{\pi^0}}{N^{Br}_{4f\to 3d}}=\epsilon\left[(1-T_i-A_\beta)+(A_\beta-A_\alpha)\frac{1}{1+C}\right], \quad (7)$$

where $N_{\pi^0}/N^{Br}_{4f\to 3d}$ has a linear dependence on $1/(1+C)$ with slope $\epsilon(A_\beta-A_\alpha)$ and vertical intercept $\epsilon(1-T_i-A_\beta)$. A plot of $N_{\pi^0}/N^{Br}_{4f\to 3d}$ vs $1/(1+C)$ is shown in Fig. 3. A least-squares fit yields $\epsilon(A_\beta-A_\alpha)=3.01\pm 0.10$ and $\epsilon(1-T_i-A_\beta)=0.196\pm 0.048$. Clearly there are still too many parameters in the model for the transfer rates A_α, A_β, and T_i to be extracted from the data. If P_H were known then ϵ could be calculated from its definition and absolute x-ray intensity measurements

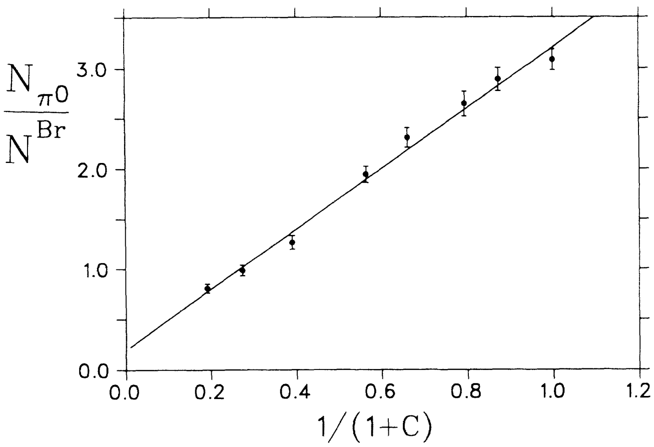

FIG. 3. $N_{\pi^0}/N^{\rm Br}_{4f\to 3d}$ as a function of $1/(1+C)$. The line is a linear best fit with parameters given in the text.

for bromine $4f\to 3d$ transitions. For $C=0$ the number of bromine $4f\to 3d$ transitions is given by $N^{\rm Br}_{4f\to 3d}=\epsilon_{\rm Br}P_{\rm Br}P_{4f\to 3d}N_{\pi^-}$. As ϵ_{π^0} is known from the measurement with H_2 in a high-pressure gas target[9] and verified in a Monte Carlo calculation it becomes obvious that $P_{\rm H}$ is the only unknown in ϵ. However, $P_{\rm H}$ is not *a priori* known. Even using particular models to predict $P_{\rm H}$, Eq. (7) only affords certain allowed ranges for the transfer parameters. Despite this, two conclusions are inescapable: The external-transfer terms A_α and A_β cannot both be zero, and $A_\alpha < A_\beta$. In condensed systems involving *muonic* hydrogen, Daniel *et al.*[15] also concluded that external transfer was indicated, by studying muonic x-ray enhancements in aqueous solutions. However, similar x-ray enhancements have been observed in systems containing no hydrogen,[16] so that muonic transfer need not be the only explanation for the effects seen, although it is the most probable one.

In summary, π^- transfer has been studied for the first time with pionic x-ray measurements in tandem with π^0 detection. The pions were stopped in liquid targets of a hydrogenous molecule mixed with an impurity molecule at different concentrations. The pionic x-ray information labeled the atomic capture of a π^- in different atoms (Br,Cl). The π^0 detection, following the basic nuclear-capture reaction $\pi^- p \to \pi^0 n$, was an unambiguous tag for stopping π^- in hydrogen. The x-ray measurements show that the per-atom capture ratio on atoms of different nuclear charge (Br,Cl) is independent of the relative molecular concentrations. In contrast, the ratios of π^- captures on hydrogen to bromine in the bromodecane molecule was found to depend strongly on the concentration of the impurity-molecule species (CCl_4). These results are not consistent with any model which only considers "internal" transfer of the pions from hydrogen to heavier elements. Instead, the results require at least some contribution from an "external"-transfer process, such as has been identified in measurements in the gaseous state. We suggest that both internal and external transfer must occur.

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