

CAPTURE AND TRANSFER OF PIONS IN HYDROGENOUS MATERIALS

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ABSTRACT

Pionic hydrogen is a short-lived exotic hydrogen isotope in which a negative pion replaces the atomic electron. The formation and subsequent interactions of pionic hydrogen are discussed, with emphasis on the process of pion transfer. Recent results using the pion charge-exchange reaction (π^- , π^0) obtained at TRIUMF are reviewed.

1. Introduction

The study of the processes involved when a negative pion stops in matter has been a topic of some interest for the past 30 years. Detecting and understanding these processes is a truly interdisciplinary effort, involving features of atomic, molecular and nuclear physics as well as elements of organic and physical chemistry. Of particular interest is the situation when the pion stops in a material containing hydrogen. In this case a pionic hydrogen atom can be formed, which is then able to wander throughout the material and can end up transferring the pion to a neighbouring atom. A series of experiments have been performed in the last few years at TRIUMF to elucidate the mechanisms governing the formation and transfer of pionic hydrogen. The classic review of the field is that of Ponomarev [1]; more recent work has been reviewed by Schneuwly [2] and Horváth [3].

Negatively charged particles such as pions, muons, kaons etc. are believed to be very similar in the manner in which they slow down and stop in matter. At sufficiently low kinetic energies, they will be captured into molecular or atomic orbitals, typically of high excitation. Being different than the atomic electrons, they do not feel the Pauli exclusion principle, and will cascade down to lower orbitals by radiative transitions or via the emission of Auger electrons. It is only in these lower orbitals where the difference between the different particles manifests itself. For strongly interacting particles like the π^- the interaction is no longer dominantly electromagnetic, and the probability of absorption by the nucleus becomes appreciable. Thus, while it is not usually possible to directly apply knowledge gained from pionic atoms to, for example, muonic atoms, the similarities are enough to prove useful. In fact, as discussed below, the study of pion transfer sheds light on aspects of muonic transfer that are not otherwise

readily accessible, in particular, transfer from excited states of the exotic atom. This is of considerable potential interest in the realm of muon-catalyzed fusion. Generally speaking, data on pion capture and transfer provide a valuable "testing ground" for models of similar muonic processes; a successful model should be able to account for the behaviour of pions, muons, kaons etc.

The typical exotic atom is an order of magnitude smaller (10^{-11} - 10^{-10} cm) than the corresponding normal atom, due to the reduced mass effect. One might therefore expect that molecular and chemical-bond effects could largely be ignored in studying the atomic and nuclear capture process, since the mesonic orbitals are well within those of the valence electrons. In fact, it turns out that chemical effects are rather important in pionic and muonic systems. This has led to the hope of using these exotic atoms as a probe of the electronic structure of molecules. In particular, pionic hydrogen appears to be a useful tool for studying the bonding of hydrogen in a molecule, and perhaps also the nature of the hydrogen bond between molecules.

Studies of atomic and molecular effects for stopped pions and muons are of interest for other reasons as well. Transfer effects need to be understood and accounted for in various classes of nuclear and particle physics experiments, especially those using hydrogen targets (e.g. fundamental weak interaction measurements such as muon capture in hydrogen, where muonic transfer processes are a major concern [4]). Finally, the ultimate fate of a pion stopping in a chemical system is important for bio-medical purposes, especially for application to pion radiotherapy which is being developed at TRIUMF and PSI.

Of the pionic atoms, pionic hydrogen is unique in two ways. Like other exotic hydrogen atoms (μ^-p , K^-p), it is electrically neutral. This, along with its small size, enables it to penetrate freely the electron shells of neighbouring atoms. This allows the π^-p to de-excite by giving energy to electrons of another atom; this is known as the external Auger effect. It also allows the pions to be transferred to atoms of higher nuclear charge Z , i.e. a reaction of the form



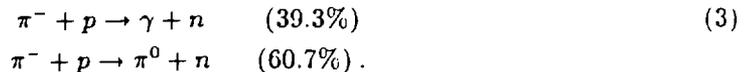
can take place. This reaction is irreversible if $Z > 1$, due to the increased binding energy of the π^- in the π^-Z atom. The similarity of reaction (1) to the muon transfer reaction



is obvious. There is, however, an important difference. Unlike the μ^- which reaches the ground state before undergoing nuclear absorption, the π^- can be absorbed from nS orbits where $n \leq 5$. The fact that the π^-p system is so ephemeral relative to the comparatively long-lived μ^-p atom means that transfer becomes more difficult to observe. For similar transfer rates, the fraction of π^- 's that will transfer is much smaller than for μ^- 's, simply because the competing π^- nuclear absorption rate is so large. The capture and transfer processes are all typically much more rapid than the pion decay rate (the free pion lifetime is 26×10^{-9} s), and so pion decay can be ignored. (There may be exceptions to this; a recent measurement at KEK

found that during the atomic cascade process, 2% of negative kaons stopped in liquid helium were "trapped" in metastable states with an overall lifetime of about 40 ns [5]. It would be very interesting to verify if a similar effect exists for π^- 's).

The second unique aspect of pionic hydrogen is the nature of its demise. If the π^- is not transferred to another atom, it will eventually be absorbed by the proton. This nuclear capture proceeds through one of two branches [6]:



The observation of a π^0 from the charge-exchange reaction is a very clean signature of nuclear capture on hydrogen. For other nuclei the production of π^0 's from π^- 's captured at rest is either kinematically forbidden, or strongly suppressed ($\leq 10^{-5}$ branching ratio [7]). Pion absorption for these nuclei leads instead to emission of energetic neutrons, charged particles and occasionally high-energy (~ 100 MeV) γ rays. The two exceptions to the rule are ${}^3\text{He}$ and the deuteron, with branching ratios for the charge-exchange reaction at rest of $(12.8 \pm 1.2)\%$ [9,10] and $(1.45 \pm 0.19) \times 10^{-4}$ [8] respectively. The potential experimental problem of background from in-flight (π^-, π^0) reactions can be eliminated by using an incident pion beam of low energy (< 25 MeV). This experimental advantage of being able to select π^- 's that have been absorbed by hydrogen is very useful tool, and is another reason that pionic hydrogen is an attractive system to study.

Molecular and chemical bond effects in pion capture can be quite large. As an example, the capture probability on hydrogen in hydrazine (N_2H_4) is 30 times less than in a gaseous mixture of $\text{N}_2 + 2\text{H}_2$ [11]. These effects are usually understood in terms of the model of large mesic molecules, introduced by Ponomarev [1]. In this picture, the π^- (or μ^-) initially captures into a molecular orbital by ejecting an Auger electron. These molecular orbitals are similar to those of the ejected electron, hence the sensitivity to electronic wavefunctions and chemical bonds. Imanishi *et al.* [12] have recently extended this model to account for Auger effects in the subsequent de-excitation.

Despite a considerable body of data on pion capture in various hydrogenous materials and mixtures, there are still a considerable number of unanswered questions. In condensed systems, there is even controversy about the basic mechanism of pion transfer. Part of the difficulty lies in the fact that the available models typically have a sufficient number of undetermined parameters to fit a large range of data. Also contributing to the problem is the fact that there are relatively few quantities that are experimentally accessible; typically these are pionic x-ray yields and the charge-exchange probability. One is therefore obliged to select carefully systems that can shed light on particular aspects of the problem. The high-quality low-momentum π^- beams available at TRIUMF, along with an excellent detection system for π^0 's produced at rest, have enabled us to improve on some of the older measurements and to begin to answer some of the outstanding questions in the field.

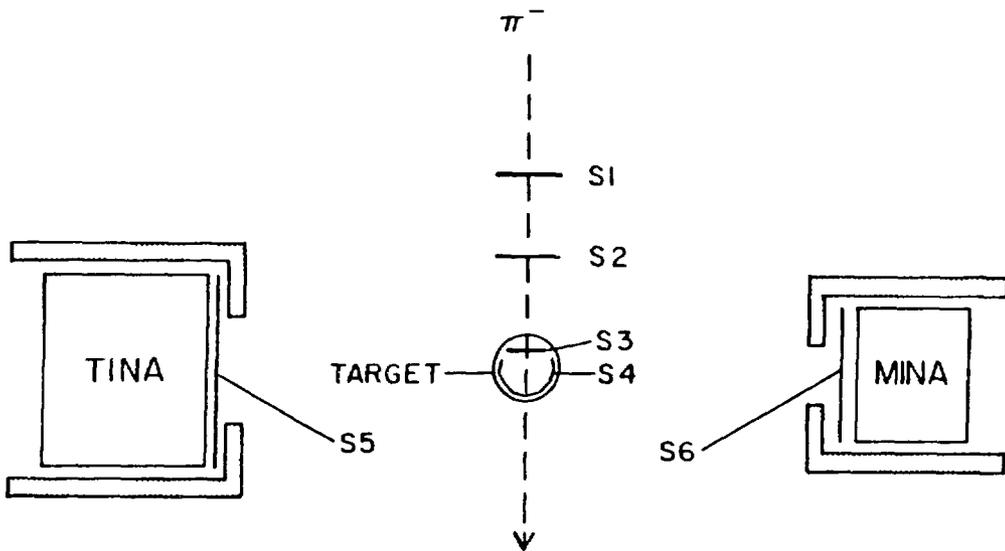


Figure 1: Experimental layout. TINA and MINA are the two large NaI(Tl) gamma-ray detectors. S_1 - S_6 are plastic scintillators.

2. Gaseous Mixtures

Perhaps the simplest possible system to consider is a gaseous mixture of hydrogen with other elements. The pion transfer reaction has been extensively studied, largely by the Dubna group, for mixtures of hydrogen with noble gases (He, Ne, Ar, Kr, Xe) [13,14], and simple molecules such as D_2 , CO, N_2 , and CO_2 [14,15,16]. We have chosen to study the $H_2 + D_2$ system [17], which is particularly interesting because of the relation to similar processes in muon-catalyzed fusion.

The pion transfer is studied by measuring the variation of the π^0 yield as the relative concentration of H_2 and D_2 is varied. Figure 1 shows the experimental setup, which is essentially the same for all our measurements. The production of π^0 's is signalled by the decay $\pi^0 \rightarrow \gamma\gamma$ (98.8% branching ratio). The two γ 's are produced back-to-back and with equal energies in the rest frame of the π^0 ; however the π^0 has a small kinetic energy of 2.9 MeV. Therefore the photons are Doppler-shifted in the lab frame and so have a small distribution about 180° and energies of between 55 MeV and 83 MeV. The γ 's are detected by two large NaI(Tl) scintillators (TINA ϕ 46 cm \times 51 cm; MINA ϕ 36 cm \times 36 cm) at 180° to each other. Plastic scintillators (S_5, S_6) in front of TINA and MINA enable us to reject signals due to charged particles.

The incident π^- beam was at a momentum of 78 MeV/c (kinetic energy of 20 MeV) with

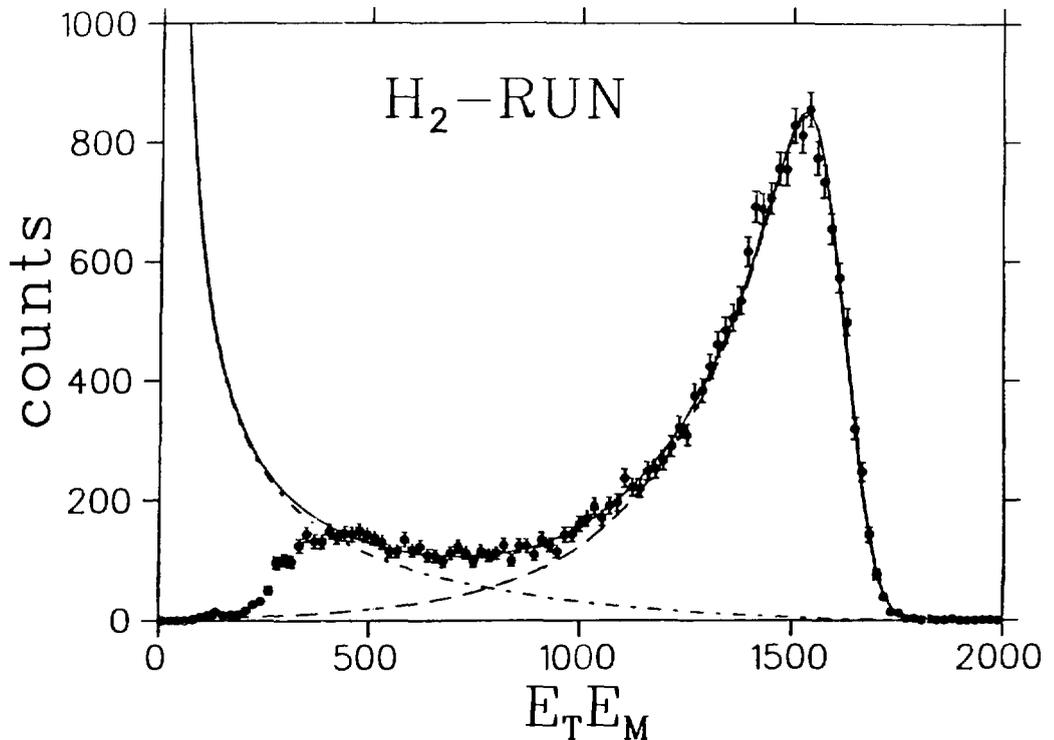


Figure 2: Spectrum of the product of the γ -ray energies $E_T E_M$ from a hydrogen target. The dashed line is a fit to the π^0 peak, and the dot-dashed line is the fitted background.

a typical flux of 10^5 s^{-1} . The arrival and stopping of an incident π^- is signalled by a series of thin plastic scintillators S_1 - S_4 (see Fig. 1). S_3 was made of a very thin (0.5 mm) deuterated plastic $(\text{CD})_n$ to reduce the number π^0 's produced from pions stopping in the scintillator.

The essentially continuous time structure of the TRIUMF beam (99% duty factor) is an advantage here. The time between the arrival of the π^- in the target, and its eventual nuclear capture is $< 10^{-9} \text{ s}$; the π^0 decays practically instantaneously (10^{-16} s), and so the two γ 's must be detected in time coincidence with the arrival of the π^- . This requirement allows the suppression of various backgrounds, notably that from energetic neutrons.

The kinematics of the π^0 decay require that

$$E_T E_M = \frac{M_{\pi^0}^2}{4\sin^2(\psi/2)}, \quad (4)$$

where E_T and E_M are the γ energies deposited in the two detectors, M_{π^0} is the π^0 mass and ψ is the angle between the two γ rays. Therefore the π^0 events will appear as a peak in a histogram of $E_T E_M$, as illustrated in Fig. 2. The width of the peak is primarily due to the energy resolutions of the NaI detectors which were each $\sim 7\%$ at 100 MeV. The number of π^0 events is easily extracted by fitting to a function representing the peak superimposed on a smooth background.

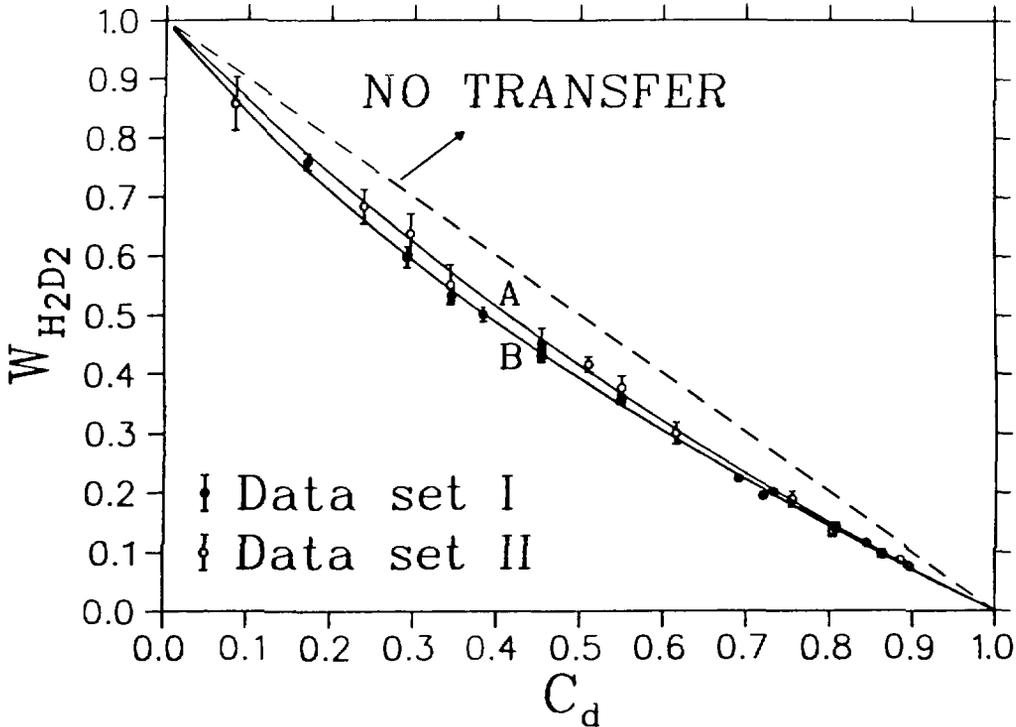


Figure 3: Relative π^0 production versus deuterium concentration for $H_2 + D_2$ mixtures. The dashed line is the expectation if transfer did not occur. The curves marked A and B are the fits to Eq. (6) for the two data sets.

Figure 3 shows the relative π^0 production as the concentration of deuterium (C_d) is increased. If there were no transfer, then the data should fall along the straight line indicated, with the reasonable assumption that the molecular stopping probability is the same for H_2 and D_2 . Clearly, significant transfer from the hydrogen to the deuterium does take place. For example, in a 50:50 mixture fewer than half the pions are absorbed on the hydrogen.

To further interpret the data, we adopt a phenomenological approach, inspired by the work of Petrukhin and Prokoshkin [15]. We assume that nuclear capture occurs only in collision, due to Stark mixing. In this approach, the probability of pion capture on hydrogen is viewed as being dependent on a competition between collisionally-induced capture and collisionally-induced transfer. In the simplest case, where the inverse transfer reaction $\pi^- d + p \rightarrow \pi^- p + d$ is negligible, one has that the capture probability on hydrogen can be written

$$W_H = \frac{C_p}{C_p + C_d} \frac{\beta_{pp}C_p + \beta_{pd}C_d}{\beta_{pp}C_p + \beta_{pd}C_d + \lambda_{pd}C_d} \quad (5)$$

Here we have defined β_{pp} as the probability of nuclear capture on the proton in a $\pi^- p + p$ collision, β_{pd} as the probability of nuclear capture on the proton in a $\pi^- p + d$ collision, and λ_{pd} as the probability of transfer from p to d in a $\pi^- p + d$ collision. C_p and C_d are the

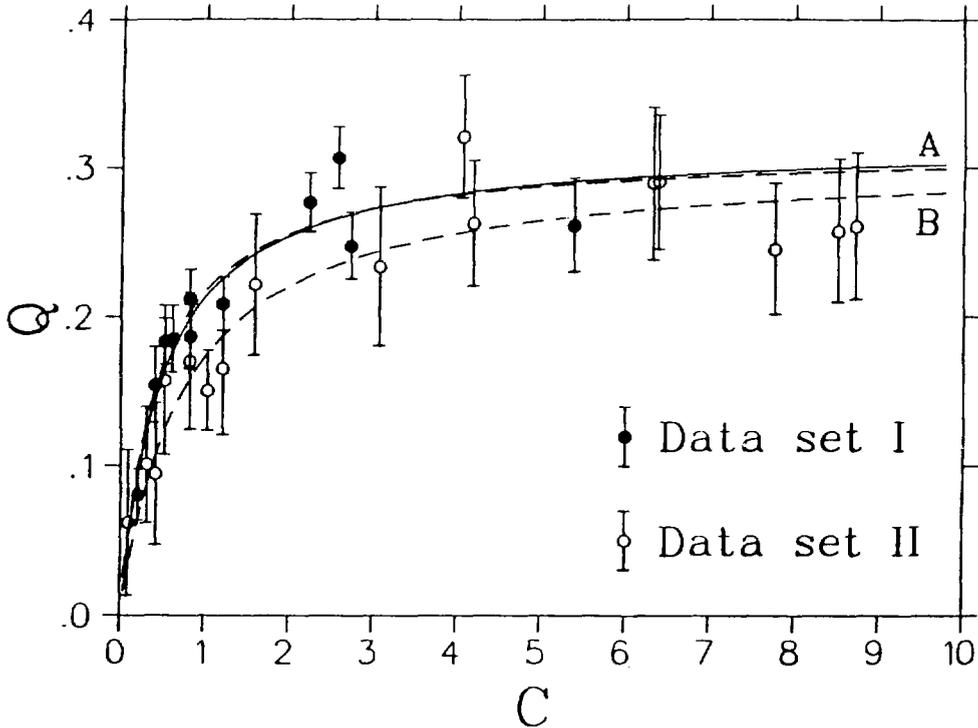


Figure 4: Transfer probability Q versus relative concentration for $H_2 + D_2$ mixtures. The dashed curves are the fits to Eq. (7) for the two data sets, and the solid curve is the common fit, which yielded the parameters listed in Table 1.

relative concentrations of hydrogen and deuterium respectively. The model can be expressed in terms of only two parameters $\kappa = \beta_{pd}/\beta_{pp}$ and $\Lambda = \lambda_{pd}/\beta_{pp}$. Defining the concentration ratio $C = C_d/C_p$, one has

$$W_H = \frac{1 + \kappa C}{(1 + C)(1 + \kappa C + \Lambda C)} \quad (6)$$

Similarly, the net transfer probability Q can be written

$$Q = \frac{\beta_{pd}C_d}{\beta_{pp}C_p + \beta_{pd}C_d + \lambda_{pd}C_d} = \frac{\Lambda C}{1 + \kappa C + \Lambda C} \quad (7)$$

from which it is easy to see that the transfer probability is simply related to W_H :

$$W_H = \frac{1 - Q}{1 + C} \quad (8)$$

This transfer probability is plotted as a function of C in Fig. 4. The data were obtained during two separate running periods which were analyzed independently to determine our systematic errors. The results of the two fits are shown, as well as a common fit to all our data. Table 1

Table 1: The parameters of Eq. (7), extracted from the present data, compared to those obtained by Petrukhin and Prokoshkin [15], and those obtained by a fit to our data as well as the data from Aniol *et al.* [18] and Kravtsov *et al.* [19] (“World fit”).

Parameter	Present result	Petrukhin and Prokoshkin [15]	World fit
Λ	0.65 ± 0.07	0.4 ± 0.1	0.45 ± 0.04
κ	1.40 ± 0.22	1.3 ± 0.4	0.93 ± 0.14
κ/Λ	2.2 ± 0.4	3.3 ± 1.3	2.1 ± 0.4
$Q(C \rightarrow \infty)$	$(32 \pm 3)\%$	$(23 \pm 4)\%$	$(33 \pm 3)\%$

displays the parameters extracted from the latter fit, and compares them to those obtained by Petrukhin and Prokoshkin [15]. The agreement is reasonable, although our data point to a somewhat larger asymptotic value for the transfer. Two other measurements for W_H are available [18,19]; both are more precise than our data, but both were only taken at one value of the concentration. Inclusion of these two datum (“World fit” in Table 1) changes the extracted values for Λ and κ somewhat. This reflects the fact that these two parameters are fairly highly correlated in the fit. The values for κ/Λ and $Q(C \rightarrow \infty)$ are essentially unchanged however. The result $\kappa/\Lambda > 1$ indicates that in $\pi^-p + d$ collisions, the probability for capture is higher than that for transfer. Since $\kappa \sim 1$, the probability of pion capture in hydrogen is similar for $\pi^-p + p$ collisions and $\pi^-p + d$ collisions.

In an ancillary experiment, we have searched for a pressure dependence of the transfer, and found none, in agreement with previous observations [18]. In fact, the asymptotic transfer is in good agreement with the value $Q = (28 \pm 13)\%$ at $C = 0.84$ measured at liquid hydrogen densities [20]. As emphasized by Horváth [21], this is important because any non-collisional processes would induce a virtual density (pressure) dependence for W_H . The absence of such a dependence is strong evidence that collisional processes dominate the capture and transfer as assumed in our analysis.

3. Alcohols

Since the nuclear capture rate of a π^- on a hydrogen atom is the net result of atomic capture followed by any transfer processes (if they occur), it is often difficult to disentangle the two effects. The analysis of data on pion charge-exchange probabilities to extract transfer rates, often depends on assumptions about the initial pion capture on the different atoms within the molecule. In a complex molecule, the pion charge-exchange probability is an average over the

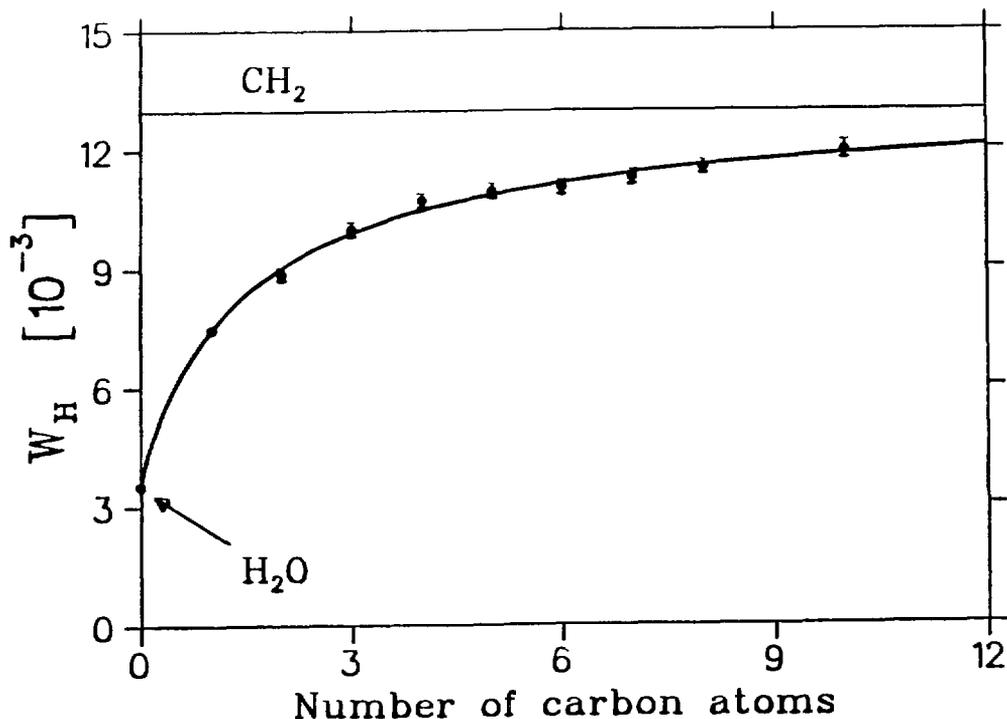


Figure 5: Capture probability in hydrogen, W_H , of alcohols and water. The solid curve is a spline fit through the values calculated using Eq. (9).

chemically different hydrogen atoms in the molecule. Therefore, systematic studies of charge exchange in closely related chemical species could prove desirable. The hope is to determine an empirical scale of *relative* capture rates on atoms in specific chemical environments. Such a scale could then be useful in isolating effects due to transfer reactions.

With this idea in mind, we proceeded to measure pion charge-exchange probabilities for a series of alcohols. To analyze the results, we postulate that the pion capture on a given hydrogen atom in the molecule depends on its position in the molecule. We define the probability for capture on the hydrogen in the OH group as R_H^O , that for a hydrogen attached to the neighbouring carbon atom as R_H^α (in the chemist's language, this is a hydrogen in the α position with respect to the OH group), and that for all other hydrogens as R_H^β . R_C and R_O are the pion capture rates on carbon and oxygen respectively. The simple model is that W_H is given by

$$W_H = \frac{n_H^\alpha R_H^\alpha + R_H^O + n_H^\beta R_H^\beta}{n_H^\alpha R_H^\alpha + R_H^O + n_H^\beta R_H^\beta + n_C R_C + R_O}, \quad (9)$$

where the n 's are the appropriate number of atoms in the particular molecule. It is important to note that the R factors parameterize both the initial selection of the atom by the pion and any transfer. Figure 5 depicts the experimental results for the primary alcohols, plotted as a function of n_C , the number of carbon atoms in the molecule. Also included is the W_H value

Table 2: Pion capture probabilities in hydrogen for alcohols. In the last column the values calculated using Eq. (9) with the parameters given in Table 3 are presented. The fit was determined using the W_H values for water and the primary alcohols only (the first 10 entries).

Molecule	Formula	W_H (10^{-3})	Calculated (10^{-3})
Water	H ₂ O	3.52 ± 0.07	3.52 ± 0.05
Methanol	CH ₃ OH	7.47 ± 0.09	7.47 ± 0.10
Ethanol	CH ₃ CH ₂ OH	8.88 ± 0.16	9.04 ± 0.08
Propan-1-ol	CH ₃ (CH ₂) ₂ OH	9.98 ± 0.17	9.90 ± 0.07
Butan-1-ol	CH ₃ (CH ₂) ₃ OH	10.70 ± 0.17	10.47 ± 0.08
Pentan-1-ol	CH ₃ (CH ₂) ₄ OH	10.95 ± 0.18	10.87 ± 0.08
Hexan-1-ol	CH ₃ (CH ₂) ₅ OH	11.03 ± 0.18	11.17 ± 0.08
Heptan-1-ol	CH ₃ (CH ₂) ₆ OH	11.29 ± 0.19	11.41 ± 0.08
Octan-1-ol	CH ₃ (CH ₂) ₇ OH	11.54 ± 0.17	11.59 ± 0.08
Decan-1-ol	CH ₃ (CH ₂) ₉ OH	11.96 ± 0.22	11.87 ± 0.08
Deuterated Methanol	CD ₃ OH	0.845 ± 0.031	1.17 ± 0.02
Deuterated Methanol	CH ₃ OD	5.55 ± 0.11	6.31 ± 0.10
Butan-2-ol	CH ₃ CH ₂ CH(OH)CH ₃	10.85 ± 0.17	10.54 ± 0.08
2-Methyl-propan-2-ol	(CH ₃) ₃ COH	10.66 ± 0.18	10.61 ± 0.09
Cyclohexanol	(CH ₂) ₅ CHOH	8.23 ± 0.18	9.57 ± 0.07

for water. The data clearly show a monotonic increase in the charge-exchange probabilities with n_C which is easily reproduced by the model. The W_H value for polyethylene is indicated on the figure, and seems to be a plausible asymptotic limit for the fit, as might be expected.

The experimental values are given in Table 2, along with the W_H values calculated using the fit. Also listed are the values for deuterated methanols (to be discussed below) as well as several other alcohols. The fitted capture ratios are given in Table 3. The value for R_H^a/R_C is slightly lower than R_H^b/R_C . This is consistent with the lower electron density expected around hydrogen atoms in the methylene group adjacent to the hydroxyl group, due to the strongly electronegative nature of the oxygen atom. Since the atomic capture process is expected to be dominated by Auger transitions, the capture rate on an atom is expected to depend on the electron density around that atom. Similarly, the value for R_H^o/R_C is much less than either R_H^a/R_C or R_H^b/R_C . This is consistent with the large reduction in the electron density around a hydrogen atom attached directly to the very electronegative oxygen atom. The location of the hydroxyl group does not appear to effect the charge-exchange probabilities significantly (compare the results for Butan-1-ol and Butan-2-ol). The calculation for 2-Methyl-propan-2-

Table 3: Best fit capture ratios determined from the primary alcohols.

Capture Ratio	Fitted value
R_H^β/R_C	$(6.72 \pm 0.05) \times 10^{-3}$
R_H^α/R_C	$(6.28 \pm 0.08) \times 10^{-3}$
R_H^O/R_C	$(3.48 \pm 0.07) \times 10^{-3}$
R_O/R_C	1.97 ± 0.02

ol, which has a very different structure than the primary alcohols, also reproduces the data well. However, the calculation does rather poorly for cyclohexanol. Whether this is related to its ring structure or is a problem with the measurement is not yet clear. We have applied a similar model to the existing data on alkanes [22,23] and found no effects due to ring structures [24].

The results for the deuterated methanols CD_3OH and CH_3OD are particularly interesting. The sum of the capture probabilities in CD_3OH and CH_3OD is considerably less than the probability for CH_3OH . This is also seen by comparing the individual W_H values of the deuterated methanols with the predictions of our model (c.f. Table 2). This is clearly evidence of transfer. However, we can correct for the effect of transfer using the $\pi^-p + d \rightarrow \pi^-d + p$ transfer parameters determined from our H_2/D_2 measurements, or perhaps more applicably, the H_2O/D_2O data discussed below. After this correction the W_H values are 1.33×10^{-3} and 6.24×10^{-3} for CD_3OH and CH_3OD respectively, in quite reasonable agreement with the predictions of our model.

Finally, we have attempted to apply the model parameters deduced from the alcohols to existing data on a very different system, a series of sugar isomers. Earlier work at TRIUMF measured charge-exchange probabilities W_H for several hexose and pentose isomers of sugars, and found evidence of an inverse correlation between W_H and the melting point [25,26]. It was suggested that this correlation was indicative of hydrogen bonding causing a higher melting point and reducing the electron density around the hydrogen atoms and hence reducing the pion capture probability. The present model applied to the sugar data overestimates the W_H values by about a factor of two. This could be considered as additional evidence of the effect of hydrogen bonding on pion capture.

4. Dibromoalkanes

Charge-exchange probabilities were also measured for the first five primary dibromoalkanes (chemicals of the form $Br(CH_2)_nBr$). The results are shown in Fig. 6; the curve is simply a

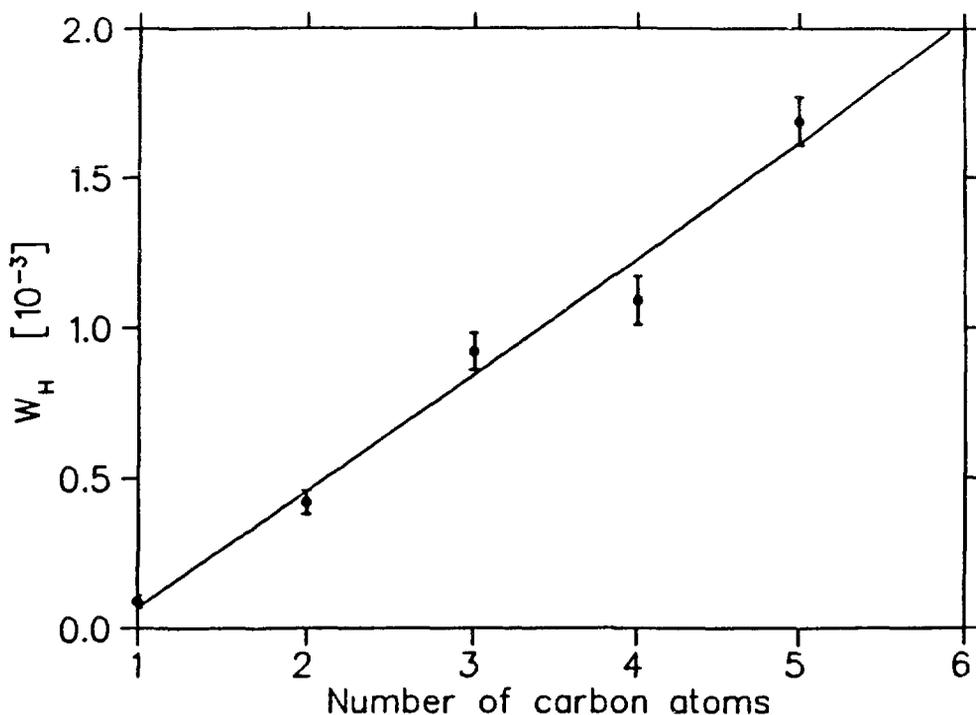


Figure 6: Capture probability in hydrogen for dibromoalkanes. The line is a guide to the only.

guide to the eye. The W_H values increase dramatically as the chain length increases. This again shows the important effect of a very electronegative group (the Br atoms in this case) on the probability of the π^- capturing on a hydrogen atom. As the chain lengthens, less of the molecule is affected by the distortions in the electron density due to the Br atoms on the ends. Unlike the alcohols series, the curve shows no signs of flattening out, probably indicating that the Br atoms have considerable influence throughout the molecule, even in the case of dibromopentane ($\text{Br}(\text{CH}_2)_5\text{Br}$). Notice that the W_H value for dibromopentane is still far below the value for $(\text{CH}_2)_n$ (12.9×10^{-3}). Unfortunately, it appears that there is not sufficient data with only 5 species investigated to apply a model similar to the one used for the alcohols (Eq. (9)) and get meaningful results.

5. Transfer in $\text{H}_2\text{O}/\text{D}_2\text{O}$ Mixtures

The results on pion charge-exchange probabilities in mixtures of gases, for example the H_2/D_2 mixtures discussed above, clearly indicate that the π^- is transferred from one molecule to another. This can be labelled an "external" transfer process. The data are quite consistent with the collisional picture described above, in which the small, neutral π^-p atom breaks away from the molecule and is transferred in a subsequent collision, *i.e.* $\pi^-p + Z \rightarrow \pi^-Z + p$. In more complicated molecules than H_2 , the collisional transfer could conceivably also precede

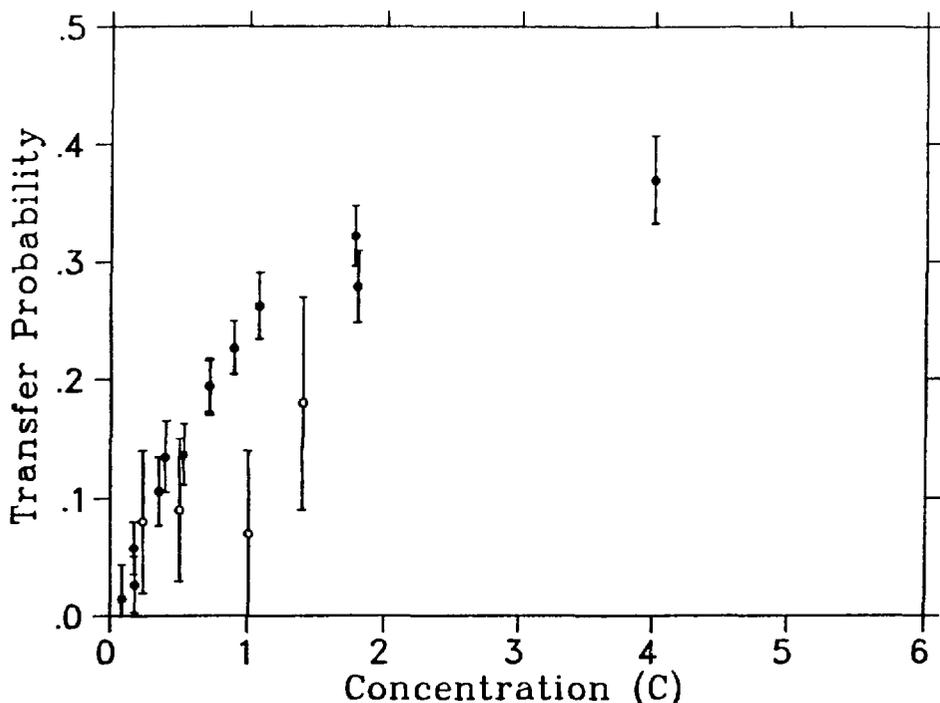


Figure 7: Transfer probability Q as a function of relative concentration for mixtures of liquid H_2O+D_2O . The open circles are the data of Petrukhin *et al.* [31] and the closed circles are the present data.

via an "internal" route, that is, the π^-p could move to other atoms in the same molecule.

Such an alternative mechanism for pion transfer in complex molecules has been proposed by D.F. Jackson and co-workers [27,28,29]. In this model, transfer occurs only to the neighbouring atom to which the hydrogen is bonded. The transfer occurs by a barrier penetration method: the π^- tunnels along the Z-H bond. This mechanism is clearly quite different in nature than collisional transfer; it is purely an "internal" transfer process, and so it cannot explain the data on pion and muon transfer in gas mixtures. However, a wide range of both pion charge-exchange and pionic x-ray data on hydrogenous molecules can be fit with this approach [27,30].

Clearly it is important to separate the two proposed mechanisms for pion transfer in condensed systems. An obvious question is whether there is any evidence of a need to invoke an "external" transfer process in condensed systems? To answer this, one approach is to adopt a method similar to the measurements on gas mixtures. The idea is to investigate the suppression of W_H as one varies the relative concentration of two liquids, only one of which is hydrogenous.

An appealing possibility, obviously analogous to the H_2/D_2 gas mixtures, is to investigate mixtures of liquid H_2O and D_2O . Under the assumption that the initial π^- stopping probability is the same for the D_2O molecule as for the H_2O , the observation of a non-linearity in the concentration dependence of W_H would be evidence for "external" transfer. This measurement

was first attempted by Petrukhin *et al.* [31], and while the data were sketchy, they indicated a non-linearity similar to that seen in the gas mixtures. We have repeated the experiment with higher precision and over a broader range of concentrations, and confirm the effect [32]. Figure 7 illustrates our results and compares them with the previous data. The behaviour is similar to that observed in the gas mixtures, but not identical - in particular, the asymptotic value of the transfer is $52 \pm 10\%$ compared to $32 \pm 3\%$ for the gases, using the same phenomenological model to fit the data. Unfortunately, the result is not unambiguous. At high concentrations of D_2O , the hydrogen would be found primarily in a HOD molecule. Therefore the concentration dependence could be interpreted in terms of molecular effects, such as the deuteron preferentially carrying away the π^- in the breakup of the π^- -HOD molecule. The lingering possibility also remains that the initial pion stopping probability on the H_2O molecule depends upon the D_2O concentration (such effects have been observed in some gas mixtures [33,34]). If this were the case, W_H would have a non-linear concentration dependence even without any pion transfer.

6. Organic Mixtures

In addition to the H_2O/D_2O system, Petrukhin *et al.* [31] investigated two other systems, mixtures of $C_{10}H_{22}$ and CCl_4 as well as mixtures of C_6H_6 and CCl_4 . Again, non-linearities in the concentration dependence of W_H were observed, qualitatively similar to the behaviour seen in the H_2O/D_2O case. However, two assumptions had to be made in the analysis. The first, as discussed above for the H_2O/D_2O mixtures, was that the molecular stopping probabilities do not depend on the concentration. The second assumption was that these stopping probabilities scale with the pion stopping power. Molecular stopping occurs at much lower energies than those at which stopping powers are measured, and situations have been observed where the molecular capture does not scale with the stopping power [9]. Thus the results could not be considered as unequivocal evidence of "external" transfer.

We have recently adopted a similar approach, with one important modification [35]. We used mixtures of bromodecane ($C_{10}H_{21}Br$) and carbon tetrachloride CCl_4 . The selection of these two materials allows us to "tag" the π^- stops on each molecule using pionic x-rays. Stopping on the bromodecane is signalled by the 223.5 keV $Br\ 4f \rightarrow 3d$ x-ray and stopping on the CCl_4 is signalled by the 150.7 keV $Cl\ 3d \rightarrow 2p$ x-ray. The x rays were detected using an intrinsic germanium detector. Simultaneous measurement of the charge-exchange probability and the pionic x-rays allows us to avoid the assumptions discussed above, and to examine the transfer process unambiguously.

If the molecular stopping distributions are independent of concentration, then the ratio $(N_{3d \rightarrow 2p}^{Cl} / N_{4f \rightarrow 3d}^{Br})/C$, should be independent of concentration. Here $N_{3d \rightarrow 2p}^{Cl}$ is the number of Cl x-rays detected, $N_{4f \rightarrow 3d}^{Br}$ is the number of Br x-rays detected and C is the relative concentration. Figure 8 shows the results for this ratio plotted against the concentration; no significant dependence is seen. This enables us to rule out variations in the molecular stopping probability

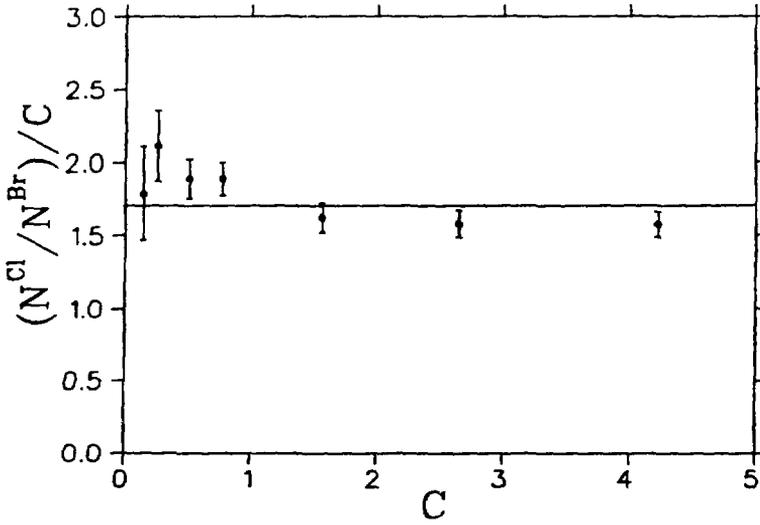


Figure 8: The ratio of the number of pionic x-rays from Cl and Br divided by the relative concentration C , vs. C , for mixtures of bromodecane and carbon tetrachloride. The line is a guide to the eye.

as an explanation for effects seen in the charge-exchange yield. It also allows us to correct the charge-exchange data for the relative stopping probabilities in the two types of molecules in the mixture, without having to make any assumptions. With Eq. (3), the number of π^0 events detected is given simply by

$$N_{\pi^0} = 0.607 \epsilon_{\pi^0} P_{\alpha} P_H (1 - T_T) , \quad (10)$$

where ϵ_{π^0} is the π^0 detection efficiency, P_{α} is the probability of a π^- stopping on the hydrogenous molecule (bromodecane), P_H is the probability that a π^- stopping on the bromodecane will go to a hydrogen atomic orbital and T_T is the total transfer probability to other (higher- Z) atoms. Similarly, the number of bromine $4f \rightarrow 3d$ x-ray transitions observed is given by

$$N_{4f \rightarrow 3d}^{Br} = \epsilon_{Br} P_{\alpha} P_{Br} P_{4f \rightarrow 3d} , \quad (11)$$

where ϵ_{Br} is the detection efficiency for the Br x-ray, P_{Br} is the probability, given a π^- stopping on the bromodecane molecule, of the π^- going to a bromine atomic orbital and $P_{4f \rightarrow 3d}$ is the probability of the $4f \rightarrow 3d$ transition in the cascade in Br. By considering the ratio of $N_{\pi^0} / N_{4f \rightarrow 3d}^{Br}$, the dependence on the molecular stopping probability P_{α} cancels out.

The transfer probability T_T can be decomposed into three terms:

$$T_T = T_i + T_{\alpha} + T_{\beta} . \quad (12)$$

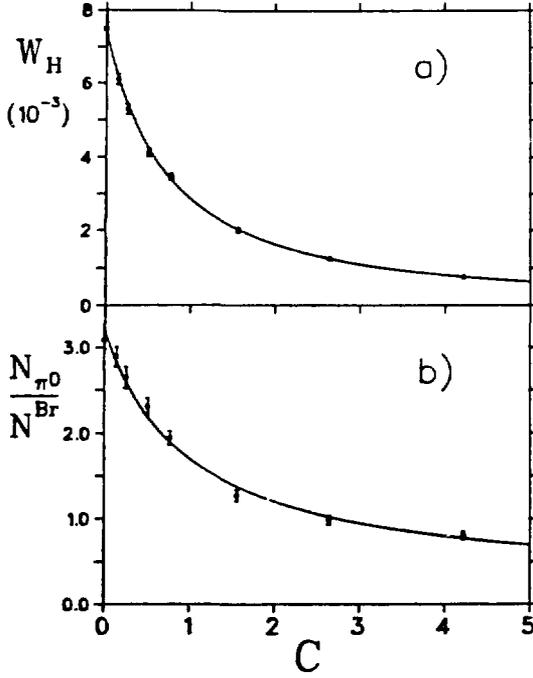


Figure 9: a) The pion capture probability in hydrogen W_H vs. the relative concentration C , and b) the ratio of the number of π^0 's to the number of Br x-rays, for mixtures of bromodecane and carbon tetrachloride. The lines are to guide the eye.

Here T_i , the "internal" transfer term, is the probability for the π^- to transfer to different atoms in the same molecule (*i.e.* the bromodecane). Note that this could represent the barrier penetration mechanism of Jackson *et al.*, the collisional process, a combination of the two, or possibly other unspecified mechanisms. The terms T_α and T_β represent "external" transfer, to atoms in different bromodecane molecules (T_α) or to atoms in a CCl_4 molecule (T_β). The internal transfer probability T_i should be independent of the molecular ratio $C = n_\beta/n_\alpha$, 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 $T_\alpha = \frac{A_\alpha n_\alpha}{n_\alpha + n_\beta}$ and $T_\beta = \frac{A_\beta n_\beta}{n_\alpha + n_\beta}$, where A_α and A_β are transfer constants for α (bromodecane) and β (carbon tetrachloride) molecules respectively. We make no assumption here as to the mechanism for external transfer. Together with (10)-(12) this gives

$$\frac{N_{\pi^0}}{N_{\text{Br}}^{4f \rightarrow 3d}} = \epsilon \left[1 - T_i - \frac{A_\alpha}{1+C} - \frac{A_\beta}{1+(1/C)} \right], \quad (13)$$

where the factor ϵ is defined by

$$\epsilon = 0.607 \frac{\epsilon_{\pi^0} P_H}{\epsilon_{\text{Br}} P_{\text{Br}} P_{4f \rightarrow 3d}}, \quad (14)$$

and is independent of C . The only concentration dependent terms are those involving the two external transfer terms A_α and A_β . Thus any concentration dependence observed in $N_{\pi^0}/N_{4f \rightarrow 3d}^{\text{Br}}$ is evidence that these terms must be non-zero.

The concentration dependences of W_H and $N_{\pi^0}/N_{4f \rightarrow 3d}^{\text{Br}}$ are shown in Fig. 9. Figure 9(b) clearly shows that a significant amount of external transfer is taking place. Unfortunately, there are too many parameters in the model to extract the transfer rates A_β , A_α and T_i from the data independently [35]. Despite this, two conclusions are inescapable: the external transfer terms A_α and A_β cannot both be zero, and $A_\alpha < A_\beta$. The second conclusion comes directly from the fact that the ratio $N_{\pi^0}/N_{4f \rightarrow 3d}^{\text{Br}}$ decreases as C increases, and implies that the CCl_4 "scavenger" is more efficient at accepting the transferred pions than the bromodecane. We have repeated the measurement using a different molecule, trifluoro-trichloro-ethane (CF_3CCl_3) instead of CCl_4 . The results are qualitatively the same. The conclusion is clear: no model describing pion transfer in condensed systems can be considered complete unless it encompasses "external" transfer.

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