SOME FEATURES OF π^--MESON CAPTURE BY HYDROGEN IN CHEMICAL COMPOUNDS

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The probability has been measured for capture of π^- mesons by hydrogen in different chemical compounds (bases, salts, and pseudohydrides). The experimental results obtained are satisfactorily explained in terms of the large-mesic-molecule model. In the case of a compound with q identical groups containing hydrogen, the probability for π^--meson capture by bound hydrogen is suppressed by a factor q as the result of quantum-mechanical resonance phenomena. The probability of π^--meson capture in pseudohydrides is considerably less than in normal hydrides, which indicates a complex structure of the pseudohydrides.

1. INTRODUCTION

IN our previous papers[1-3] it was shown that capture of π^- mesons by hydrogen in compounds of the type Z_mH_n and other hydrogen-containing materials is determined to major extent by the formation of a large mesic molecule preceding the capture, when the π^- meson transfers from the continuous spectrum to a discrete spectrum in the common levels of the system $Z_m\pi^- H_n$. Consideration of this preceding stage permits explanation of the sharp decrease in the probability of π^--meson capture by hydrogen with increasing Z in materials of the type Z_mH_n and $Z'_kZ_mH_n$, and other features of the capture process.

In the present paper we report the results of further studies of π^--meson capture by chemically bound hydrogen for various classes of compounds: bases, salts, and hydrides of the transition metals (pseudohydrides). This study reveals new features of the capture mechanism for compounds whose composition includes several identical groups containing hydrogen. The experiments were performed with the synchrocyclotron of the Laboratory for Nuclear Problems, Joint Institute for Nuclear Research, by the technique described previously.[4-7]

2. EXPERIMENTAL RESULTS

Measurements of the probability W for capture of π^- mesons by hydrogen in hydrogen-containing materials were made by a relative method— by comparison of the yields of γ-ray pairs from the reaction

$$\pi^- + p \rightarrow \pi^0 + n, \quad \pi^0 \rightarrow 2\gamma \tag{1}$$
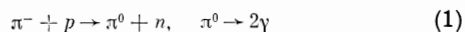
from the target being studied and from a target of lithium hydride. The ratios obtained were then normalized to the value $W_{LiH} = (35 \pm 4) \times 10^{-3}$.[1] The intensity of reaction (1) for the materials studied turned out to be extremely small ($W = 10^{-3}$–10^{-4}), and the contribution from the background process of π^--meson charge exchange in flight in the complex nuclei entering into the target composition[5] was as high as 50% in some cases. The accuracy of the values of W was determined mainly by the uncertainty in measurement of this contribution.

The values of W obtained are presented in Tables I–III.

3. DISCUSSION

In Table I the measured probabilities W for bases $Z'(OH)_q$ are compared with values computed from the formula

$$W_{Z'(ZH_n)_q} = a_L \frac{qnZ^{-2}}{Z' + q(Z + n)}, \tag{2}$$

which follows from the large-mesic-molecule model.[2] In an earlier paper[1] it was shown that this formula is accurately satisfied for a broad class of compounds. In our case Z = 8, the number of the period in the periodic table of the elements is L = II, $a_{II} = 1.28 \pm 0.15$,[1] and n = 1. As can be seen from Table I, for materials of type $Z'(OH)_q$ which contain q identical hydrogen-containing groups, formula (2) gives values which are too high in comparison with experiment by q times, i.e., the π^--meson capture process occurs as if the "removal" of a π^- meson at a hydrogen atom involved not only the oxygen atom directly bound with it in the hydroxyl group OH but also the oxygen atoms from the q − 1 other hydroxyl groups.

This explanation is a natural one in terms of the quantum-mechanical theory of resonance. Actually, all q hydroxyl groups in the alkalis $Z'(OH)_q$ are identical, and therefore a meson captured initially by one of them is subsequently distributed among all the groups as the result of quantum-mechanical exchange effects (like the π electrons in the benzene molecule). Consequently, the probability of finding a π^- meson in each of the hydroxyl groups is 1/q. However, the OH group in which

Table I   Probabilities W for bases $Z'(OH)_q$

Material	$10^4 W_{exp}$	q	$\dfrac{W_{Eq.\ (2)}}{W_{exp}}$	$10^4 W_{Eq.\ (3)}$
KOH	6.7±2.1	1	1.1±0.3	7.2±0.8
Ca(OH)$_2$	5.7±1.4	2	1.9±0.4	5.3±0.6
Cd(OH)$_2$	3.3±1.2	2	1.8±0.6	3.1±0.4
Ba(OH)$_2$	2.5±0.7	2	2.2±0.6	2.7±0.3
Ba(OH)$_2$·8H$_2$O	18±3	2	—	
	0±3 *			1.3±0.2
Al(OH)$_3$	6.3±1.8	3	2.4±0.6	5.0±0.6

*After subtraction of the contribution from water of crystallization (see Eq. (5)).

Table II Probabilities W for salts $RC(Z'_h Z_m H_n)_g$

Material	$10^4 W_{exp}$	$10^4 W^*_{exp}$	q	$\dfrac{W_{Eq.\,(2)}}{W^*_{exp}}$	$10^4 W_{Eq.\,(4)}$
$(NH_4)_2Cr_2O_7$	6.0 ± 1.1	6.0 ± 1.1	2	2.2 ± 0.4	6.5 ± 0.7
$(NH_4)_2TiF_6$	7.0 ± 1.4	7.0 ± 1.4	2	2.4 ± 0.5	8.3 ± 0.9
$Cd(CH_3COO)_2$	8.7 ± 1.3	8.7 ± 1.3	2	2.2 ± 0.3	9.7 ± 1.1
$Ba(CH_3COO)_2$	10.5 ± 1.4	10.5 ± 1.4	2	1.7 ± 0.3	9.0 ± 1.0
$Pb(CH_3COO)_2\cdot3H_2O$	12 ± 2	6 ± 2	2	2.0 ± 0.6	6.1 ± 0.8
$Ca(CH_3COO)_2\cdot H_2O$	9.3 ± 1.4	5.5 ± 1.6	2	4.2 ± 1.3	11.5 ± 1.8
$Zn(CH_3COO)_2\cdot2H_2O$	7.5 ± 1.2	1.3 ± 1.4	2	15 ± 8	9.5 ± 1.2
$C_2H_4Cl_2$	13 ± 2	13 ± 2	2	2.2 ± 0.3	14 ± 1
CH_3J	10 ± 2	10 ± 2	1	1.7 ± 0.4	17 ± 3
C_4H_9J	33 ± 4	33 ± 4	1	1.1 ± 0.3	37 ± 4

In calculation of W in the compounds $R(NH_4)_q$ we assumed $a_{II} = 1.0 \pm 0.1$ according to the measurements for ammonia salts.[1] For all the remaining compounds we assumed $a_{II} = 1.28 \pm 0.15$.[1] W^* is the probability W after subtraction of the water of crystallization according to Eq. (5).

the meson was initially captured is distinguished, since in capture of the meson the hydrogen lost its electron. The loss by the OH group of the hydrogen electron does not destroy the condition of identity of the groups, since the shape of the potential wells is determined mainly by the oxygen atom. Therefore the oxygen atoms from all q groups participate uniformly in the capture of a π^- meson which initially enters one of the groups. Capture by the hydrogen atom can occur only in the initial group, since in the remaining OH groups this is prevented by the electron remaining in the hydrogen atom. Thus, in systems with q identical groups the quantum-mechanical resonance effect should lead to suppression of the probability of π^--meson capture by bound hydrogen by q times in comparison with formula (2),[1] which for symmetric systems of the type $R(ZH_n)_q$ takes the form

$$W_{R(ZH_n)_q} = a_L \frac{nZ^{-2}}{Z'' + q(Z+n)}. \qquad (3)$$

Here R is a group of atoms with total nuclear charge Z''. As can be seen from Table I, the values of W computed from formula (3) are in good agreement with the experimentally determined values.

For the case of complex salts having a structure $R(Z'_k Z_m H_n)$, formula (3) is generalized as follows:

$$W_{R(Z'_k Z_m H_n)_q} = a_L \frac{nZ^{-2}}{Z'' + q(kZ'+mZ+n)}, \qquad (4)$$

where Z is the nuclear charge of the atom bound with hydrogen. The values of W calculated with this formula are listed in Table II.

As can be seen from Tables I and II, the regularity which we have noted has a general nature and does not depend on the form of the identical groups, and formula (4) accurately describes the experimental results for a broad class of materials: bases, acid and basic salts, and even organic halides.

Some of the materials investigated contained water of crystallization, i.e., had the structure $R(Z'_k Z_m H_n)_q \cdot Q(H_2O)$. In determination of the probability $W^* = W_{R(Z'_k Z_m H_n)_q}$ in this case it is necessary to subtract the contribution from capture of π^- mesons by hydrogen in the molecules of the water of crystallization:

$$W^* = W_{R(Z'_k Z_m H_n)_q \cdot Q(H_2O)} - \frac{QZ_{H_2O}W_{H_2O}}{Z'' + QZ_{H_2O} + q(kZ_{k'} + mZ_m + n)}. \qquad (5)$$

Here $W_{H_2O} = (35 \times 6) \times 10^{-4}$,[1] and the number of electrons in the molecule of water of crystallization is $Z_{H_2O} = 10$. Use of formula (5) is justified, since the Z law is satisfied in the initial "capture" of the π^- mesons.[1] In order to check the correctness of Eq. (5) we performed a control experiment: the probability W was measured for $Ba(OH)_2 \cdot 8H_2O$ and $Ba(OH)_2$ (after removal of the water of crystallization). As can be seen from Table I, the result of this experiment is consistent with formula (5).

The effect revealed in the study of bases is also observed, as can be seen from Table II, in the case of salts: the probability of capture of a π^- meson by hydrogen atoms does not depend on the total number of hydrogen atoms in the compound but only on the number in a group, if all the groups are identical. We emphasize that this effect occurs both for the basic radical $(NH_4)^+$ and the acid radical $(CH_3COO)^-$. Apparently it always appears when there are identical groups in the compound. For example, the compound $C_2H_4Cl_2$ also is included in this rule: the capture probability in it is a factor of two smaller than the value predicted by formula (2). The experimental results for $Ca(CH_3COO)_2 \cdot H_2O$ and $Zn(CH_3COO)_2 \cdot H_2O$ do not follow the general rule. The cause of this deviation is not yet clear.[2] For the remaining materials, if we take into account the uncertainty in the value of the coefficient a_L,[3] the measured and calculated values agree.

As was noted previously,[1] the measured values of 1/W for compounds of the type $Z''(ZH_n)$ differing only in the atoms Z'' follow a linear dependence on Z''.

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[1] For clarity we have simplified the discussion. In reality the assumption of the distinct nature of the initial hydroxyl group is unnecessary, since the exchange interaction between all of the groups involves not only the π^- meson but also q - 1 electrons from the hydrogen atoms of the other groups. These electrons will inhibit the capture of mesons by hydrogen, and as a result the total probability of finding a meson near all of the hydrogen atoms is $\rho_\pi/(\rho_\pi + \rho_e) = 1/q$, since in the subsystem $(ZH_n)_q$ there is one meson ($\rho_\pi = 1$) and q - 1 electrons inhibiting the capture ($\rho_e = q - 1$). Thus, the probability W will be suppressed in these compounds by q times in comparison with formula (2).

[2] It is possible that the acetic acid salts of Ca and Zn used in the measurements lost their water crystallization. In this case the values of W_{exp} agree with the theoretical values (see Table II).

[3] For the hydrides $Z_m H_n$ of the second period $a_L = a_{II} = 1.28$, while for the salts $(NH_4)Z$ and $Z(BH_4)$ we have $a_{II} = 1$.[1]
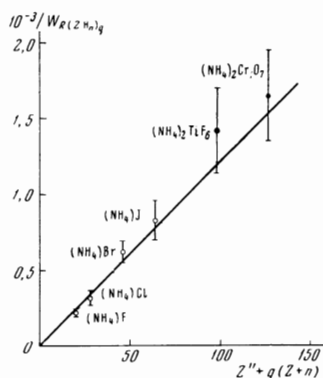
Comparison of the measured probabilities $W_{R(NH_4)_q}$ with Eq. (4). Z'' is the total nuclear charge of the atom group R. The straight line is Eq. (4) calculated for $a_{II} = 1$. The solid circles are the data of the present work, and the hollow circles are those obtained previously.[1]

The similar dependence

$$1/W_{R(ZH_n)_q} \sim Z'' + \text{const}, \tag{6}$$

following from formula (4) is satisfied for the complex ammonia salts $R(NH_4)_q$ studied in the present work (see the figure).

In the case of titanium and zirconium hydrides, formula (3) goes over to the following form[2]:

$$W_{Z_mH_n} = a_L \frac{nZ^{-2}}{mZ + n}. \tag{7}$$

Comparison of the measured probabilities $W_{Z_mH_n}$ with those calculated from formula (7) is shown in Table III. In calculating W for the hydrides of Ti we used the value $a_{IV} = 11.0 \pm 1.4$.[1]

The coefficient a_V was not measured in the previous studies. If we use the empirical relation $a_{L+1}/a_L \approx 3$ to determine it,[1] the value $a_V = 32$ obtained turns out to be larger than the maximum permissible value $a_V = 21$ which follows from the condition $a(Z) = a_L n/(mZ + n) \geq 1$.[2] in calculating W for ZrH_2 the latter value of a_V was assumed.

As can be seen from Table III the measured values of W for the titanium hydrides are considerably smaller than those obtained from formula (7). It is extremely likely that this discrepancy is due to the different nature of the chemical bond in the hydrides of Ti from that in normal hydrides. These compounds, in contrast to the true hydrides (LiH, CaH_2, and so forth) studied by us previously,[5,1] are pseudohydrides.[8] The nature of the hydrogen-metal bond in such compounds is not clear at this time. According to qualitative ideas,[8] the hydrogen in pseudohydrides

is partially bound chemically and partially ionized and existing in the form of a proton gas. Since the π^- meson cannot be captured by a free proton, only that part of the hydrogen in pseudohydrides which is chemically bound participates in the capture of π^- mesons, which leads to a reduction in the probability W. According to the estimates which can be made on the basis of our measurements in terms of the scheme described, roughly half of the hydrogen in these compounds is ionized.

In conclusion we take this occasion to thank S. S. Gershteĭn for helpful discussions and for assistance in the work.

Probabilities W for hydrides Z_mH_n

Material	$10^4 W_{exp}$	$10^4 W_{Eq. (7)}$
TiH_2	2.5 ± 1.2	19 ± 3
$TiH_{1.7}$	6.5 ± 1.4	16 ± 2
ZrH_2	6.2 ± 1.4	6.1

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183