INVESTIGATION OF π^--MESON CAPTURE BY HYDROGEN IN HYDROGENOUS SUBSTANCES

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The probability has been measured for π^- capture by hydrogen in Z_mH_n hydrides and compounds of the $Z'_kZ_mH_n$ type. A relative method is employed which is based on comparison of the π^0-meson yield from the material studied and from LiH. It is shown that the results can be satisfactorily described by a model assuming large mesic molecules. The initial capture follows a Z law. The probability for π^- capture in Z_mH_n hydrides changes markedly on transition of the atom Z between periods in the periodic table, and also depends on the type of chemical bond.

1. INTRODUCTION

NUCLEAR capture of stopped negative mesons in condensed materials is determined to a considerable extent by the mesic-atom (mesic-molecule) processes preceding it. The study of these processes, in addition to circumstantial interest, has also a large practical value, since the elementary reactions of decay and interaction of negative particles (mesons, antiprotons, hyperons) usually must be studied by stopping these particles in material with a complex chemical structure. The interpretation of these experiments requires a knowledge of the probabilities of capture of these particles by the nuclei of the atoms entering into the composition of the material.

The probability of atomic capture of μ^- mesons which have stopped in matter turns out in most cases to be roughly proportional to the nuclear charge Z, in agreement with the Z law of Fermi and Teller.[1] All observed deviations[2-4] from this law occur in the direction of a weaker Z dependence.

A different situation exists in the stopping of π^- mesons in matter. Studies of the absorption of π^- in LiF have shown that the probability of capture of a π^- meson by lithium in this compound is substantially less than follows from the Z law.[5] In studying the absorption of π^- mesons in hydrogenous materials it was observed that the probability of capture of π^- mesons by hydrogen is strongly suppressed.[6] To explain this effect the model of π^--meson interception[6] has been proposed specifically for hydrogenous materials. However, subsequent quantitative studies[7-9] have shown that the sharp suppression of π^- capture by chemically bound hydrogen nuclei is due to another mechanism, which is described by the "large-mesic-molecule" model.[10, 11] According to this model the slowed down π^- meson transfers from the continuous spectrum to the discrete spectrum into levels common to the whole molecule. The probabilities of capture of a π^- meson by a proton and a nucleus Z are determined by the mesic-molecule stage of the process, when the π^- meson transfers from the common mesic-molecule levels to the separated levels of the $p\pi^-$ and $Z\pi^-$ mesic atoms.

The Z dependence of the probability W for the cap-

ture of π^- mesons by nuclei of the hydrogen entering into the composition of binary chemical compounds of the type Z_mH_n (LiH, CH$_2$, et al.) has been studied in the region $Z \leq 8$.[7, 8] Here it was found that the reduced probability P, defined as
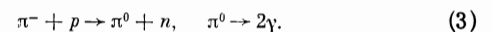
$$P = \frac{m}{n} W, \qquad (1)$$

follows a law

$$P \sim Z^{-3}. \qquad (2)$$

which subsequently found explanation in terms of the "large-mesic-molecule" model. The purpose of the present investigation (see [12]),[1] which was performed at the synchrocyclotron of the JINR Laboratory for Nuclear Problems, was to determine the dependence of the probability P on Z both for $Z < 8$ and $Z > 8$.

2. MEASUREMENTS

The experimental arrangement and procedure have been described earlier.[7-9, 13] The π^- mesons were stopped in a target of the hydrogenous material under investigation, placed between two Cerenkov total-absorption spectrometers. The spectrometers were used to record the two γ rays emitted in capture of π^- mesons by hydrogen nuclei:

$$\pi^- + p \rightarrow \pi^0 + n, \quad \pi^0 \rightarrow 2\gamma. \qquad (3)$$

The main source of background was the process of π^- charge exchange in flight in the complex nuclei entering into the composition of the target material (charge exchange of stopped π^- mesons in complex nuclei is suppressed by a factor of more than 10^4 [14, 15]). The correction associated with subtraction of this background amounted to 2% for LiH and increased up to 25% with increasing Z. The error in determination of this correction was limited by the accuracy of the measurements in the high-Z region.

Measurements of W were made for a series of hydrides Z_mH_n (including those previously studied by us[7, 8]) and more complex hydrogenous materials of the

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[1]See also Z. V. Krumsteĭn, Thesis, JINR, 1965.

Table I

Material	Z	n/m	$W \cdot 10^3$	a_L	a_L, from Eq. (7)	b_L
LiH	3	1	35 ± 4	1.26 ± 0.14	1.4	1.26 ± 0.14
$B_{10}H_{14}$	5	1,4	12.6 ± 1.5	1.44 ± 0.17	1.3	1.03 ± 0.12
CH	6	1	5.1 ± 0.6	1.28 ± 0.15	1.2	1.28 ± 0.15
N_2H_4	7	2	5.9 ± 0.7	1.30 ± 0.15	1.1	0.65 ± 0.07
H_2O	8	2	3.5 ± 0.6	1.12 ± 0.20	0.9	0.56 ± 0.10
NaH	11	1	2.4 ± 0.4	3.6 ± 0.6		
CaH_2	20	2	2.5 ± 0.3	11.0 ± 1.4		

type $Z_k' Z_m H_n$ (see Table III below). The targets had roughly the same stopping power, equivalent to 3 g/cm^2 of carbon. The chemically active hydrides $B_{10}H_{14}$, N_2H_4, NaH, and CaH_2 were enclosed in hermetically sealed aluminum containers with a wall thickness seen by the beam of about 0.5 g/cm^2. Some of the materials studied contained impurities: $B_{10}H_{14}$ (impurity of other boron hydrides 2.5%), NaH (impurity of NaOH and Na_2CO_3 5%), CaH_2 (impurity of CaO and $CaCO_3$ 10%), N_2H_4 (impurities 0.7%, including 0.36% water), KBH_4 (impurity of KOH and NaOH 7%), and $NaBH_4$ (impurity of $NaBO_2$ and $NaBH(COH_3)_3$ 3%). The remaining materials were designated as "chemically pure" or "analytically pure."

The probabilities W were determined by a relative method—by comparison of the counting rate of γ-ray pairs emitted from the target under study and from a target of LiH. Small corrections were made to the counting rate ratios obtained, which took into account the different stopping powers and the shapes of the targets. In determining the absolute values of the probabilities W the probability ratios were normalized to the quantity W_{LiH}, which was taken equal to the value $(35 \pm 4) \times 10^{-3}$ which is the average of several investigators' results.[8, 16, 17]

3. EXPERIMENTAL RESULTS

Hydrides. Probabilities W were determined for hydrides of the elements Li, B, C, N, O, Na, and Ca. The results obtained are listed in Table I. It can be seen from Fig. 1 that for $Z \leq 8$ the data obtained are well described by a dependence similar to equation (2).
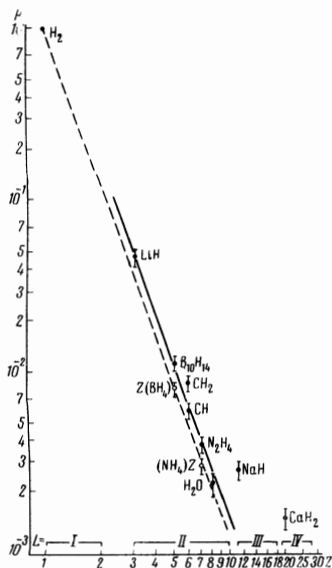

FIG. 1. Reduced probability P (formula (5)) as a function of Z. L is the number of the period in the periodic table. The solid and dashed straight lines correspond to equation (4) with $a_L = 1.28$ and $a_L = 1$. The solid points correspond to simple hydrides, and the hollow points to complex compounds.

Table II

Material	n/m	$W \cdot 10^3$	$\frac{m}{n} W / \frac{1}{2} W_{CH_2}$	a_L	b_L
CH	1.00	5.1 ± 0.6	0.77 ± 0.08	1.28 ± 0.15	1.28 ± 0.15
C_6H_6	1.00	3.5 ± 0.4	0.53 ± 0.08	1.00 ± 0.12	1.00 ± 0.12
C_7H_8	1.14	5.1 ± 0.6	0.68 ± 0.07	1.17 ± 0.13	1.03 ± 0.12
CH_2	2.00	13.2 ± 1.5	1	1.90 ± 0.22	0.95 ± 0.11
C_7H_{16}	2.29	14.6 ± 1.7	0.98 ± 0.08	1.90 ± 0.22	0.83 ± 0.09

However, in the region of higher values of Z relation (2) is violated. In the hydrides NaH and CaH_2, π^- mesons are captured by hydrogen significantly more efficiently than follows from formula (2). Possible causes of this violation are discussed below.

Hydrocarbons. The experimental values of the probability W for a series of hydrocarbons are listed in Table II. These results confirm the conclusion previously drawn by us[7] that the probability of capture of π^- mesons by bound hydrogen is nonadditive. The ratio of the values of P for CH and CH_2 was found to be 0.77 ± 0.08.[7] This result has been confirmed in recent experiments in which this ratio was found to be 0.78 ± 0.09[16] and 0.61 ± 0.08.[17]

Complex hydrogenous compounds. The materials of type $Z_k' Z_m H_n$ studied by us can be divided into two groups according to the nature of the bonds of hydrogen atoms with the atoms Z' and Z: materials in whose molecules the bonds of hydrogen atoms with the atoms Z' and Z are the same (Table III) and materials in

Table III

Material	$W \cdot 10^3$	
	Experiment	Theory, Eq. (9), $a_L = 1.28 \pm 0.15$
CH_3OH	7.5 ± 0.9	7.0 ± 0.8
C_2H_5OH	8.1 ± 0.9	7.7 ± 0.9
C_3H_6O	6.6 ± 0.8	6.7 ± 0.8
C_5H_7OOH	4.2 ± 0.5	4.9 ± 0.6

which they are different (Table IV). In compounds of the first group (Table III) the hydrogen is bound with carbon and oxygen by a covalent bond. In compounds of the second group (Table IV) the hydrogen is bound only with the light atoms of the molecule (boron and nitrogen), this bond being covalent, and the substructures $(BH_4)^-$ and $(NH_4)^+$ are bound with the heavy atoms by a bond of the ionic type, i.e., without common electrons.[18]

4. DISCUSSION OF RESULTS

In the large-mesic-molecule model the probability W for capture of a π^- meson by hydrogen in hydrides

Table IV

Material	$W \cdot 10^3$	$W_{Z'(ZH_4)}/W_{Na(BH_4)}$		a_L
		Experiment	Theory, Eq. (10)	
$Na(BH_4)$	8.4 ± 1.0	1	1	1.05 ± 1.2
$K(BH_4)$	5.9 ± 1.0	0.70 ± 0.09	0.72 ± 0.07	1.03 ± 1.2
$(NH_4)F$	4.6 ± 0.7	0.55 ± 0.07	0.51 ± 0.05	1.10 ± 0.16
$(NH_4)Cl$	3.2 ± 0.6	0.38 ± 0.06	0.36 ± 0.04	1.10 ± 0.20
$(NH_4)Br$	1.6 ± 0.2	0.19 ± 0.02	0.22 ± 0.02	0.90 ± 0.13
$(NH_4)J$	1.2 ± 0.2	0.14 ± 0.02	0.16 ± 0.02	0.94 ± 0.16

is given by

$$W = a_L \frac{n}{mZ + n} Z^{-2}. \quad (4)$$

Here the reduced probability P is defined as

$$P = \left(\frac{m}{n} + \frac{1}{Z}\right) W = a_L Z^{-3}. \quad (5)$$

The derivation of Eq. (4) is based on two assumptions.[19]

1. The probability of meson capture into different levels of the system $Z_m\pi^- H_n$ is proportional to the density of the electron cloud which existed in the vicinity of these levels before the capture of the meson. This statement is equivalent to assuming the validity of the Z law in the stage of capture into the discrete spectrum, which is reflected in the structure of the factor $n/(mZ + n)$ in (4). (Note that a comparatively small fraction of the stopped mesons ($\approx n/mZ$) are captured into the common levels. The main part of the mesons are captured immediately into levels of the heavy atom Z.)

2. A hydrogen nucleus can capture only those mesons which initially fall into a common mesic-molecule level of the molecule. The probability of this process is proportional to Z^{-2} and is determined by radiative transitions from the common levels of the system $Z_m\pi^- H_n$ to the separated levels of the mesic atoms $p\pi^-$ and $Z\pi^-$.

The coefficient a_L reflects possible deviations from the Z law, change in the conditions for π^- capture as the electron shells of the atoms build up, and features of the chemical structure of the molecule. The index L indicates the period of the periodic table to which the atom Z belongs.

The probability $W_{Z_m H}$ was defined by us so that $W_{H_2} = 1$ for reaction (3) in pure hydrogen. In this case $Z = 1$, $m = n = 1$ and the coefficient $a_I = 2$. If the influence of the effects mentioned above on the quantity a_L for the second period is small, then $a_I \approx a_{II}/2 \approx 1$. Actually, as can be seen from Table I, for the elements of the second period of the periodic table ($Z \leq 8$) the coefficient a_{II} does not depart significantly from unity.

However, in transition to the third and fourth periods (Na and Ca) the coefficient a_L changes in jumps: $a_{III} = 3.6 \pm 0.6$, $a_{IV} = 11.0 \pm 1.4$, i.e., by roughly a factor of three.[2]

We will estimate the values of a_L for hydrogen compounds with elements of the second period, taking into account the nature of the chemical bond in these compounds. These values depend on the density ρ of the distribution of the valence electron between the atoms H and Z:

$$\rho = (1 - \sigma)\rho_c + \sigma\rho_i. \quad (6)$$

Here ρ_c and ρ_i are the electron densities produced by covalent and ionic bonds, respectively; σ is the degree of ionicity of the bond,[18] which is equal to the proba-

bility of finding both electrons in that one of the atoms Z or H which is more electronegative.

The fraction of the electron density for the two electrons of a bond which is concentrated near the atom H and between the H and Z nuclei is equal to the coefficient a_L in equation (4):

$$a_L = 2\sigma + (1 - \sigma)\left(1 + \frac{s^2}{1 + s^2}\right), \quad (7)$$

where s is the overlap coefficient of the atomic wave functions. For almost all compounds $s^2/(1 + s^2) \approx 0.3$, i.e., ~ 0.15 of the electron density from each electron of the bond is contained in the region of overlap.[18] The degree of ionicity σ can be calculated with the aid of the series of electronegative elements.[18] Here, if the electronegativity of hydrogen is less than that of the atom Z (as in the water molecule H_2O), then the first term in formula (7) must be omitted. The results of calculating a_L by formula (7) are listed in Tables I and II. As a whole they agree with the experimental values.

The limiting hydrocarbons CH_2 and C_7H_{16} are exceptions. One of the causes of these deviations in the carbon series may be the increase (by 1.5 times) of the contribution of the p state to the hybrid orbital in the transition from unsaturated to saturated compounds.[18] Another possibility lies in the following: in derivation of (4) it was assumed that the mesic-molecule orbital from which a transition is possible to a level of the mesic atom $p\pi^-$ only couples the atoms H and Z. If, however, we assume that the mesic-molecule orbital belongs to all atoms of the molecule, then the factor Z^{-2} in (4) must be replaced by $n/(mZ^2 + n) \approx n/mZ^2$. Then (4) takes the form

$$W_{Z_m H_n} = b_L \frac{n}{mZ + n} \frac{n}{mZ^2 + n}. \quad (8)$$

As can be seen from Tables I and II, the coefficient $b_L \approx a_L m/n$ in the case of hydrocarbons is roughly equal to unity and depends somewhat more weakly than a_L on the form of the organic compound, but varies noticeably within the second period (except for decaborane, whose properties are close to organic). In the case of the organic compounds the dependence (8) satisfactorily describes the experimental results (Table II). Analysis of Tables I and II shows that the first hypothesis, which leads to (4), is preferable, and subsequently we will use only this hypothesis.

The dependence of the reduced probability P (formula (5)) on Z is shown in Fig. 1. The solid straight line corresponds to the value $a_{II} = 1.28$, which was obtained by averaging the coefficient a_L over the second period (see Table I). The dashed line corresponds to the value $a_L = 1$ and passes through the point $P = 1$ for hydrogen.

For complex compounds of the form $Z_k' Z_m H_n$ with a covalent bond, equation (4) takes the form[19]

$$W = \frac{a_L' v'(Z')^{-2} + a_L v Z^{-2}}{kZ' + mZ + n} \qquad v' + v = n \quad (9)$$

(v' and v are the number of bonds of the nuclei Z and Z' with hydrogen). As follows from Table III, the results of calculations on the basis of this formula agree satisfactorily with the experimental data.

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[2]One of the possible causes of this behavior of a_L may be associated with the change of the conditions for π^- capture by the molecule in the region of the valence electrons, which may depend, for example, on the number of mesic-molecule levels $N_L = 2L^2$ ($N_{II}:N_{III}:N_{IV} = 1:2.2:4$).
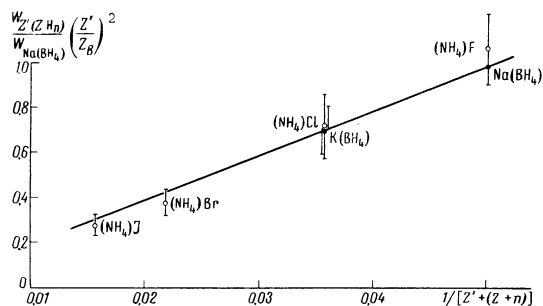
FIG. 2. Comparison of the measured probabilities W for compounds $(NH_4)Z'$ and $Z'(BH_4)$ with equation (10). The straight line is the function $20/[Z'+(Z+4)]$, which corresponds to equation (10), normalized to $W_{Na(BH_4)}$. The ammonia salts are designated by hollow circles.

In compounds of the form $Z'(ZH_4)$ (Table IV), the atom Z' is bound with the radical $(ZH_4)^-$ by an ionic bond[18] and does not participate in the interception process, i.e., $a_L' = 0$. It affects only the probability of the initial capture. In this case formula (9) takes the form ($k = m = 1$, $n = 4$)

$$W = \frac{4a_L Z^{-2}}{Z'+(Z+4)}. \tag{10}$$

It follows from this formula that the quantity $1/W$ should depend linearly on Z'. The results of the experiments (Fig. 2) confirm this conclusion. This means that in the initial capture the Z law is actually satisfied, and indicates the legitimacy of the general approach taken by us.

5. CONCLUSIONS

The analysis carried out above, on the basis of the large-mesic-molecule model, of the experimental data obtained in the present work permits us to draw the following conclusions.

1. The large-mesic-molecule model satisfactorily describes the experimental data obtained.

2. The probability of capture of π^- mesons by hydrogen in hydrides ZH changes markedly in transition between periods of the periodic table.

3. The probability of capture of π^- mesons by hydrogen in hydrogenous materials depends substantially on the type of chemical bond. This opens the possibility of using reaction (1) not only to distinguish free and bound hydrogen[20] but also for study of the nature of chemical bonds in hydrogenous materials.

4. The initial capture of π^- mesons follows a Z law.

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