

It is simpler to calculate the coefficients  $C_{i_1 i_2}^i$  with the aid of the recurrence relation

$$C_{i_1 i_2}^i = C_{i_1 i_2-1}^{i-1} + b_{i+1} C_{i_1 i_2-1}^{i+1} - b_{i-1} C_{i_1 i_2-2}^i, \quad (\text{A. 10})$$

which follows from formulas (A. 1), (A. 4) and (A. 7); to this one should add the initial condition  $C_{i_0}^i = 1$ . In particular we obtain from this the following expressions

$$\begin{aligned} C_{i_1}^{i+1} &= 1, \quad C_{i_1}^{i-1} = b_i; \\ C_{i_2}^{i+2} &= 1, \quad C_{i_2}^{i+1} = b_i + b_{i+1} - b_i, \quad C_{i_2}^{i-2} = b_i b_{i-1}; \\ C_{i_3}^{i+3} &= 1, \quad C_{i_3}^{i+1} = b_i + b_{i+1} + b_{i+2} - b_i - b_2, \\ C_{i_3}^{i-1} &= b_i (b_{i-1} + b_i + b_{i+1} - b_i - b_2), \quad C_{i_3}^{i-3} = b_i b_{i-1} b_{i-2} \end{aligned} \quad (\text{A. 11})$$

etc.

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## Mobility and chemical bond of hydrogen in titanium and palladium hydrides

A. K. Kachalkin, Z. V. Krumshtĕin, V. I. Petrukhin, V. M. Suvorov, D. Horvath, and I. A. Yutlandov

Joint Institute of Nuclear Research

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The probabilities for  $\pi^-$  meson capture by hydrogen are measured at 25, 155 and 200°C in the hydride  $\text{TiH}_{1.65}$  at 25-120 and -196°C in the hydride  $\text{PdH}_{0.67}$ . An analysis of the results shows that, within the accuracy of the measurement ( $\sim 10\%$ ), a sharp change (up to  $10^{12}$ ) in the mobility of hydrogen in the hydrides, induced by temperature changes in the ranges indicated, does not noticeably affect the probabilities for  $\pi^-$  meson capture by bound hydrogen, i.e., does not lead to appreciable changes in the Me-He bond. A comparison of the capture probabilities for palladium hydride and the hydrides of neighboring transition metals shows that there are no pronounced anomalies in the Pd-H bond.

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At the present time there are two opposing hypotheses for explanation of the character of the hydrogen bond in transition metal hydrides<sup>[1,2]</sup>; the proton hypothesis (the hydrogen in the hydride is principally in the form of the proton  $\text{H}^+$ ) and the hydride hypothesis (existence of hydrogen in the form of the hydride ion  $\text{H}^-$ ). The hydride hypothesis is successfully used for the calculation of crystalline lattices and interionic distances. The proton hypothesis in turn allows us to explain the behavior of

the hydrogen in diffusion and relaxation processes.<sup>[1]</sup> There is no single experiment at the present time which would disprove one hypothesis or the other.<sup>[2]</sup>

Earlier, it was shown experimentally that the probability of capture of pions by bound hydrogen is sensitive to the features of the chemical bond of the hydrogen in the molecules.<sup>[3-5]</sup> In the present research, we made an attempt to discover the effect of a change in the hydrogen

TABLE I.

Hydride	T, °C	f	$\nu_T/\nu_{25}$
TiH <sub>1.65</sub>	155	0.16±0.15	124
	200	0.15±0.20	355
PdH <sub>0.67</sub>	-120	0.27±0.54	1.3·10 <sup>-4</sup>
	-196	0.01±0.11	1.6·10 <sup>-12</sup>

mobility on the probability  $W$  of capture of  $\pi^-$  mesons by bound hydrogen. According to our premises, if the increase in the mobility is due to a change in the character of the chemical metal-hydrogen bond (in the electron density at the proton), then the probability  $W$  should also change.<sup>[3-5]</sup> In correspondence with this, we investigated the capture of pions by hydrogen in titanium hydride TiH<sub>1.65</sub> in which an exponential increase in the hydrogen mobility sets in at a temperature  $\geq 100^\circ\text{C}$ ,<sup>[6,7]</sup> and in palladium hydride PdH<sub>0.67</sub>, where the anomalously high room-temperature mobility of the hydrogen falls off exponentially upon lowering the temperature to  $-200^\circ\text{C}$ .<sup>[7]</sup> In both cases, the change in the hydrogen mobility was established by the method of proton magnetic resonance.<sup>[6,7]</sup>

The research was carried out on the  $\pi^-$  meson beam of the synchrocyclotron of the Joint Institute for Nuclear Research. The experimental setup and data reduction procedure were similar to those described earlier.<sup>[8]</sup> Analysis of the results was carried out on the basis of concepts of the model of large mesomolecules, according to which the probability of capture of  $\pi^-$  mesons by the hydrogen in the hydrides ZH<sub>*n*</sub> is described by the formula

$$W = \frac{nZ^{-\alpha_Z}}{Z+n} \alpha_Z \quad (1)$$

where the empirical coefficient  $\alpha_Z$  reflects the features of the chemical Z-H bond and is connected with the electron density at the proton.<sup>[3-5]</sup>

For the hydride<sup>1)</sup> TiH<sub>1.65</sub>, the measurements were made at the temperatures 25, 155, 200 °C, for the hydride PdH<sub>0.67</sub>—at 25, -120 and -196 °C. The palladium hydride was prepared by the method of interaction of metallic palladium (in the form of a plate of thickness ~ 1 mm) with molecular hydrogen at room temperature and atmospheric pressure. The palladium was first placed in an airtight vessel of stainless steel, was activated by heating to 800 °C with evacuation to a pressure of 10<sup>-6</sup> Torr. Hydridization took place over a period of 8 hours. We used technical-grade hydrogen passed through a carbon trap cooled to liquid nitrogen temperatures. The composition of the hydride was determined by weighing the vessel with the palladium before and after hydridization; the result corresponded to the formula PdH<sub>0.67±0.01</sub>. Heating of TiH<sub>1.65</sub> target was accomplished by forced hot air. The PdH<sub>0.67</sub> target was cooled by liquid-nitrogen vapor or submerged in liquid nitrogen. The temperature of the samples was measured with a copper-constantan thermocouple.

The temperature dependence of the probability of capture of  $\pi^-$  mesons by the hydrogen in the hydrides can be characterized by the quantity  $f = (W_T - W_{25})/W_{25}$ , where

$W_{25}$  and  $W_T$  are the capture probabilities for the temperatures 25 °C and  $T^\circ\text{C}$ , respectively. The obtained values of  $f$  are given in Table I. The values of the relative change in the diffusion correlation frequency  $\nu_T$ , which characterizes the mobility of the hydrogen, are also given in the table. They are determined from the formula<sup>[6]</sup>

$$\nu_T = \nu_0 \exp \{-E_d/RT\}. \quad (2)$$

The activation energy  $E_d$  is equal to 9.4 kcal/mole for TiH<sub>1.65</sub><sup>[6]</sup> and 5.6 kcal/mole for PdH<sub>0.67</sub>.<sup>[10]</sup> It is seen from the table that, within the limits of accuracy of the experiment, sharp changes in the mobility of hydrogen in metallic hydrides do not lead to changes in the character of the metal-hydrogen bond.

Among hydrides of the transition metals, the system Pd/H in certain respects is not typical.<sup>[11]</sup> The ability of palladium to absorb large quantities of hydrogen was discovered more than 150 years ago. The extraordinarily rapid diffusion of hydrogen through metallic palladium has long been used for obtaining superpure hydrogen. The physical properties of palladium are extremal<sup>[11]</sup> among its neighboring elements (Ru, Rh, Pd, Os, Ir, Pt) of the eighth group of the periodic table; among them, only palladium has a stable hydride. According to the theory of Pauling, the Pd-H chemical bond should be completely covalent, since the hydride of palladium is not covalent but a metallic hydride. The unique ability of palladium (and especially of its alloys with sulfur) to preserve its elasticity and mechanical strength in the hydridized state allows us to use it as a thin membrane for purification of the hydrogen. It is known that metallic palladium which possesses paramagnetic properties loses its anomalously high magnetic susceptibility when saturated with hydrogen, so that the hydride of composition PdH<sub>0.5</sub> is already diamagnetic and this diamagnetism can be preserved after complete removal of the hydrogen.<sup>[11]</sup> Therefore, it can be expected that the hydride of palladium can occupy a special position also with respect to the character of the Me-H bond.<sup>[11]</sup> The value  $\alpha_{Pd} = 41.3 \pm 3.6$ , obtained from the probability of  $\pi^-$ -meson capture in the hydride PdH<sub>0.67</sub>,  $W = (2.8 \pm 0.3) \cdot 10^{-4}$ , fits well the  $\alpha_Z(Z)$  line obtained in Ref. 3 for metallic hydrides (see Fig. 1). This means that with respect to the distribution of the electron density the Pd-H bond does not differ essentially from the bond in the hydrides of other metals. It is possible that the specific properties of the system

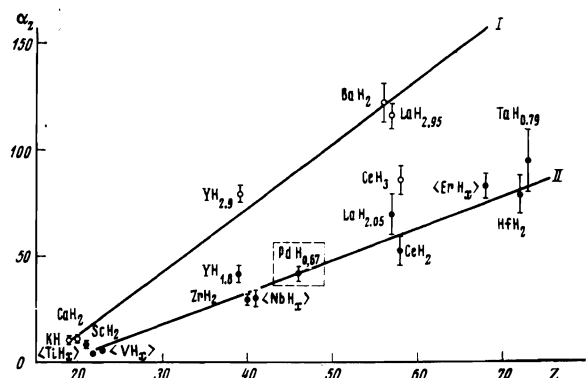


FIG. 1.

palladium-hydrogen are explained not by the feature of the chemical bond Pd-H, but, for example, by the structural changes of the crystal lattice. It is known that in palladium hydride, in contrast to the other hydrides of the transition metals, except CrH, the hydrogen fills octahedral vacancies without first filling tetrahedral vacancies.<sup>[1]</sup>

Since the capture probability of  $\pi^-$  mesons by hydrogen  $W \approx 0$  for strong OH-acids ( $\text{HNO}_3$ ,  $\text{H}_2\text{SO}_4$ ) with a long ionic bond O-H, the value of  $W$  obtained by us for the hydride  $\text{PdH}_{0.67}$  refutes the hypothesis that hydrogen in it, as also in other metallic hydrides, is predominantly in the form of the proton  $\text{H}^+$ . Preference should apparently be given to the idea of Gibbs<sup>[12]</sup> that  $\text{H}^+$  is  $\text{H}^-$  in an excited state and exists in an insignificant amount in equilibrium with  $\text{H}^-$ . From this point of view, all the steric and chemical characteristics of palladium hydride are determined by the properties of the hydride ion  $\text{H}^-$ , while the high mobility—by the specific properties of the proton  $\text{H}^+$ .

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## Measurement of the polarization correlation coefficient $C_{nn}$ of elastic $pp$ scattering at energies of 550 and 630 MeV

N. S. Borisov, L. N. Glonti,<sup>1)</sup> M. Yu. Kazarinov, Yu. M. Kazarinov, Yu. F. Kiselev, V. S. Kiselev, G. G. Macharashvili,<sup>1)</sup> V. N. Matafonov, B. S. Neganov, I. Strakhota, V. N. Trofimov, Yu. A. Usov, B. A. Khachaturov, and M. R. Khayatov<sup>2)</sup>

*Joint Institute for Nuclear Research, Dubna*

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The polarization correlation coefficient  $C_{nn}$  was determined for the elastic  $pp$  scattering at energies of 550 and 630 MeV for four scattering angles. The coefficient  $C_{nn}$  was determined by scattering a beam of protons of 0.34-0.36 polarization by a "frozen" polarized proton target with maximum polarization of  $0.98 \pm 0.03$ . The results of the measurements indicated that, in the investigated energy range,  $C_{nn}$  depended weakly on the angle and energy.

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The energy and angular dependences of the polarization correlation coefficient  $C_{nn}$  were determined in the range 550-630 MeV by measuring  $C_{nn}$  for  $550 \pm 15$  and  $630 \pm 10$  MeV and four scattering angles  $\vartheta = 41^\circ, 69^\circ, 77^\circ, 91^\circ$  and  $\vartheta = 40.6^\circ, 69.6^\circ, 78^\circ, 92^\circ$ , respectively (center-of-mass system). The experimental method and the features of the apparatus were described in detail earlier.<sup>[1]</sup> A polarized proton beam of 0.34-0.36 polarization and a polarized proton target of the "frozen" type with maximum polarization  $0.98 \pm 0.03$  were used. The coefficient  $C_{nn}$  was determined by measuring the intensity of the scat-

tered proton beam produced as a result of scattering of the polarized beam by the target and this was done for various combinations of the beam and target polarizations:  $I_{++}, I_{+-}, I_{-+}$ , and  $I_{--}$ , where the first index identifies the direction of polarization of the target and the second that of the beam relative to the normal of the left-hand scattering plane.

The measurements were carried out during five-minute exposures. The sign of the target polarization changed after 8-12 h operation and the sign of the beam