

Investigation of spin-polarized atomic hydrogen in solid H₂ at helium temperatures

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An investigation was made of spin-polarized atomic hydrogen in solid H₂ at helium temperatures. It was found that the recombination rate constant of H atoms in solid H₂ was independent of a magnetic field right up to $B = 7.5$ T at temperatures in the range $1.3 \text{ K} \leq T \leq 5.4 \text{ K}$. Theoretical estimates were obtained of the most probable mechanisms of recombination of polarized H atoms in solid H₂.

Studies of the properties of quantum crystals and of the behavior of quantum impurities in these crystals have become one of the basic trends of research in low-temperature physics. These properties are manifested most strikingly by helium and hydrogen crystals, which have become classical objects of such investigations. This is reflected in a large number of papers published on these crystals. The nature of motion of impurities in these crystals is particularly interesting. Atoms of ³He and hydrogen in ⁴He and H₂ crystals are the most natural objects of such investigations because of the following circumstances: a) a small mass; b) a relatively simple method for introducing them into crystals, particularly in the case of ³He in ⁴He; c) availability of well-tested methods for the observation of their behavior in crystals (NMR for ³He and ESR for H atoms). Until recently the attention of theoreticians and experimentalists has been concentrated on the system comprising ³He in ⁴He. The phenomenon of quantum diffusion of ³He atoms with an anomalous temperature dependence has been predicted¹ and confirmed experimentally,^{2,3} stimulating in turn further theoretical⁴ and experimental⁵ studies.

Obviously, the motion of H atoms in an H₂ crystal at sufficiently low temperatures should also occur by subbarrier transitions. In contrast to ³He in ⁴He, the system of H in H₂ is metastable because of the recombination process ($\text{H} + \text{H} \rightarrow \text{H}_2 + 4.4 \text{ eV}$ has no threshold), so that the decrease in the concentration of H atoms with time is a direct consequence of diffusion, and the characteristic time of this process depends on the concentration of atoms. This circumstance is the main reason why studies of the diffusion of hydrogen atoms in H₂ have become possible only recently.⁶ Katunin *et al.*⁶ established that the recombination of atoms is described by a second-order law and the temperature dependence of the recombination rate constant has two clearly distinct regions, one of which ($T \lesssim 4 \text{ K}$) corresponds to the process of quantum diffusion. A linear dependence of the recombination rate constant (and, consequently, of the diffusion coefficient, because $K \propto D$) on temperature in the range $1.3 \leq T \leq 4 \text{ K}$ was established later⁷ and was explained by using a general theory of quantum diffusion in nonregular crystals, developed in Refs. 4 and 8. The discovery of the quantum diffusion of H atoms in an H₂ crystal demonstrated that even at $T \approx 1.3 \text{ K}$ this system is metastable at concentrations $n_{\text{H}}/n_{\text{H}_2} \sim 10^{-4} - 10^{-3}$. Therefore, extension of studies

to higher concentrations of the atoms has made it necessary to find ways and methods for stabilizing them at the concentrations achieved at present. One of the possible ways of stabilizing this system is a reduction in the probability of the elementary events of the recombination of atoms when their electron spin is polarized. This idea was confirmed experimentally⁹ in investigations of spin-polarized atomic hydrogen in the gaseous phase, and a strong suppression of the recombination of atoms in the presence of high magnetic fields at low temperatures ($\mu B \gg kT$) was observed. However, various attempts to stabilize by this method the system of H atoms in an H₂ crystal have given contradictory results.¹⁰ We pointed out earlier⁷ the absence of the influence of high magnetic fields on the recombination rate constant of H atoms in H₂. We shall now give a detailed analysis of the results obtained in Refs. 6 and 7, as well as the results of direct experiments on the recombination of spin-polarized atomic hydrogen in a H₂ crystal together with a detailed discussion of these experiments.

EXPERIMENTAL METHOD

Our experiments were carried out using low-temperature apparatus based on an ESR spectrometer operating in the 2-mm range, developed by us in cooperation with the Special Design Bureau of the Donetsk Physicotechnical Institute. The apparatus is shown schematically in Fig. 1a.

A direct-amplification ESR spectrometer was used to detect hydrogen atoms. The main feature of this spectrometer was the relatively easy frequency tuning, which was achieved by using a backward-wave tube as a microwave power source and a low- Q cylindrical cavity as the working resonator. The frequency of the backward-wave tube was stabilized against a reference resonator with $Q \sim 10^4$ by an automatic frequency tuning system. A crystal of InSb cooled with liquid helium and operating as a photoresistor was employed as the detector. The spectrometer was operated in the homodyne regime, which made it possible to record both the absorption and dispersion signals. The magnetic field was modulated ($f = 25 \text{ kHz}$) and the derivatives of these signals were recorded. A method was developed for ensuring that the signals were not modulated. The working resonator was placed in the low-temperature part of the apparatus.

This part of the apparatus (Fig. 1b) contained a super-

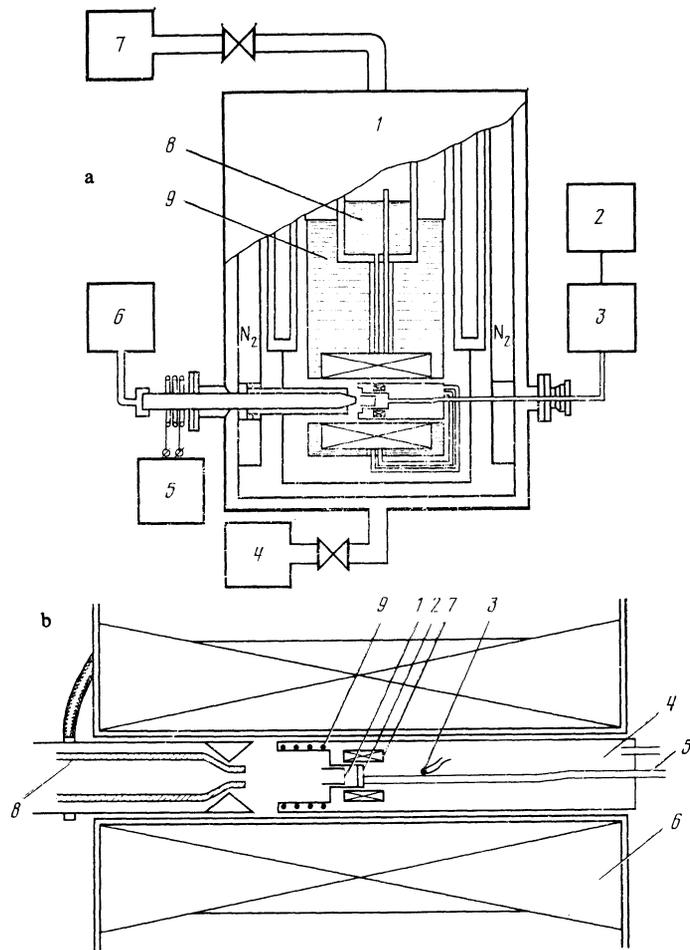


FIG. 1. a) Schematic diagram of the apparatus: 1) helium cryostat; 2) recording system of the ESR spectrometer with a block for control of the magnetic field; 3) microwave part of the spectrometer; 4) vacuum pumping system; 5) hf oscillator; 6) system for admission of H_2 ; 7) VN-4 roughing pump for removal of 4He vapor; 8) helium volume at $T = 1.3$ K; 9) helium volume at $T = 4.2$ K. b) Low-temperature part of the apparatus: 1) working resonator of the spectrometer; 2) single-crystal sapphire plate; 3) temperature sensor; 4) chamber with 4He ; 5) waveguide; 6) superconducting solenoid; 7) coil for modulation of the magnetic field; 8) channel for atoms; 9) heater.

conducting solenoid producing a field up to 8 T of $\sim 10^{-4}$ T/mm homogeneity. The working resonator of the spectrometer was located in a stainless-steel chamber and it was placed inside the solenoid. The chamber could be filled with 4He . The lowest temperature attainable in the chamber was $T = 1.3$ K and this could be maintained for 10–12 h. The chamber contained also a coil for modulation of the magnetic field, a heater, and two germanium thermometers made at the Special Design and Technological Bureau of the Institute of Semiconductors, Academy of Sciences of the Ukrainian SSR, Kiev. The cylindrical walls of the resonator and its bottom were in contact with liquid helium. A single-crystal sapphire plate, 400 μ thick, was bonded to the bottom. The cover of the cylinder was in the form of a grid with a transmission coefficient of 0.25.

Samples were prepared as follows. A mixture of hydrogen atoms and molecules arrived from a high-frequency dissociator ($f_{hf} \approx 30$ MHz) along Pyrex channels in the low-temperature zone of the apparatus and it condensed on the sapphire plate inside the working resonator. The samples were formed at $T \approx 1.3$ K. The temperature of the sapphire exposed to this hydrogen beam was somewhat higher than the temperature of helium in the chamber and it amounted to $T \lesssim 2$ K. The hydrogen-beam flux at the exit from the dissociator was $\sim 10^{18}$ particles/sec. Samples formed in 3–10 min and it was found that the ESR signal of the hydrogen

atoms increased linearly, indicating homogeneous distribution of the atoms in the sample. In our experiments the concentration of hydrogen atoms in solid H_2 was

$$n_{H1}/n_{H2} \sim 10^{-3} - 10^{-4}.$$

The ESR spectrum of H atoms in solid H_2 usually consisted of two lines separated by 5×10^{-2} T. In view of the specific features of spectroscopy in the 2-mm range and the special properties of our samples (the dispersion signal of atomic hydrogen was an order of magnitude higher than the absorption signal), the dispersion signal was recorded. The line profile in the modulation regime was then strongly distorted (Fig. 2b), whereas in the regime of recording the dispersion signal itself the line profile was classical (Fig. 2a). The amplitudes of the signals observed in the modulation regime and in the regime of recording the primary dispersion signal ($\Delta H_{pp} \approx 6 \times 10^{-4}$ T) depended linearly on the microwave power and were proportional to one another during an experiment. However, the much lower sensitivity in the modulation-free regime (with the signal/noise ratio of ~ 10 , see Fig. 2) and in the regime of recording of the derivative of the absorption signal, compared with the derivative of the dispersion signal, forced us to record the distorted line profile and to check the results against those of control experiments in which the primary signal was recorded.

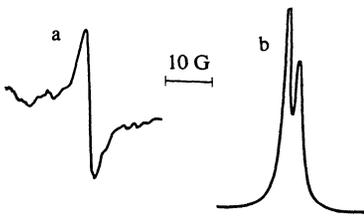


FIG. 2. One of the ESR lines for H atoms: a) dispersion signal; b) distorted dispersion signal in the modulation recording regime.

EXPERIMENTAL RESULTS

Most of the experiments on the influence of a magnetic field on the recombination rate constant of hydrogen atoms in solid H_2 were carried out at the lowest temperature attainable in our apparatus ($T \approx 1.35$ K) in fields up to 7.5 T, i.e., under the conditions of the maximum polarization of these atoms in respect of the electron spin [the degree of polarization $P = (n_{\uparrow} - n_{\downarrow}) / (n_{\uparrow} + n_{\downarrow}) = \tanh(\mu B / kT)$ at $T = 1.3$ K amounted to $P_{5T} = 0.989$ and $P_{7.5T} = 0.999$ in the fields indicated by the subscript; n_{\downarrow} is the number of H atoms with the electron spin projection $+1/2$ and n_{\uparrow} is the number with the spin projection $-1/2$]. We could expect a considerable slowing down of the recombination rate:

$$K_{B=0} / K_B = \text{ch}^2(\mu B / kT) \sim 150|_{B=5 \text{ T}}, \quad \sim 2000|_{B=7.5 \text{ T}}.$$

The experiments were carried out as follows. A sample was formed by the method described above. Then, the amplitude of the ESR signal of H atoms was measured at $T \approx 1.35$ K at equal time intervals (usually $\Delta t = 200$ sec) for 2–3 h in a field $B_0 = 5$ T. During this time the amplitude of the signal was approximately halved, but the line profile was retained. The field was then altered to B_1 ($B_1 = 0$ or $B_1 = 7.5$ T) and the sample was kept under these conditions for 2–3 h, when the initial field was restored and the amplitude of the ESR signal of hydrogen atoms was measured again. The results of two such experiments are shown in Fig. 3. Figure 3a gives the results obtained for $B_1 = 0$. Figure 3b corresponds to the case when $B_1 = 7.5$ T.

As established by us earlier,⁶ the decrease in the amplitude of the ESR signal A and, consequently, in the concentration of H atoms (n_H) in solid H_2 obeyed a quadratic law $dn_H/dt = -2Kn_H^2$, so that consequently $A^{-1}(t) = A^{-1}(0) + 2Kt$, where $K = K(T, B)$ is the recombination rate constant.

The ratio of the slopes of the latter straight line obtained for different values of the fields B_0 and B_1 represented the ratio of the recombination rate constants: $K(B_0, T_0) / K(B_1, T_0)$. Therefore, we could find the dependence of the recombination rate constants of H atoms on the applied magnetic field at a given temperature. We found that the ratios $K(B_0) / K(B_1)$ obtained at $T \approx 1.3$ K could be described by the following expressions:

$$\frac{K(5T)}{K(0)} : 1,3 \pm 0,1 \quad 1,1 \pm 0,1 \quad 0,7 \pm 0,1 \quad 0,8 \pm 0,1 \quad 1,1 \pm 0,2,$$

$$\frac{K(7,5T)}{K(5T)} : 0,9 \pm 0,1 \quad 0,7 \pm 0,1 \quad 1,0 \pm 0,1 \quad 1,1 \pm 0,2 \quad 0,9 \pm 0,2.$$

Hence, it was found that $K(B_0) / K(B_1) = 1 \pm 0.1$ for $B_1 = 0$ or 7.5 T and $B_0 = 5$ T. Several similar experiments were carried out also at $T = 4.2$ K. At this temperature there was no influence of the applied magnetic field on the recombination rate constant.

We reported earlier^{6,7} the results of an experimental determination of the temperature dependence of the recombination rate constant of H atoms in solid H_2 subjected to magnetic fields of 0.3 T (Ref. 6) and 5 T (Ref. 7). These results are plotted in the same graph in Fig. 4. In the field of 0.3 T the atoms of hydrogen were practically unpolarized in respect of the electron spin even at the lowest temperatures ($T \approx 1.3$ K) used in our experiments, whereas in the field of 5 T the degree of polarization varied very strongly: from the case of weak polarization ($n_{\uparrow} \sim n_{\downarrow}$ at $T = 4.2$ K) to the strongly polarized case when $n_{\uparrow} \gg n_{\downarrow}$ [$n_{\uparrow} / n_{\downarrow} = \exp(2\mu B / kT) \approx 150$ at $T = 1.3$ K]. A good agreement between the temperature dependences throughout the investigated temperature range also supported the absence of the influence of the magnetic field on the recombination rate constant of H atoms in solid H_2 . In view of this result and also because of the smallness of the effects expected at temperatures $T \sim 4$ –5 K, we made no special effort to determine the influence of the field at these temperatures.

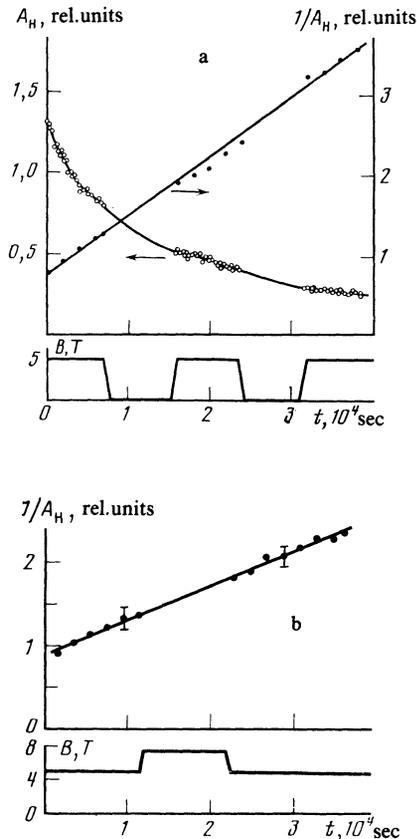


FIG. 3. a) Time dependence of the concentration of H atoms in an $H + H_2$ sample at a fixed temperature of $T = 1.3$ K: \circ) amplitude of the ESR signal of H atoms; \bullet) reciprocal of the amplitude of the ESR signal of H atoms. b) Time dependence of the reciprocal of the amplitude of the ESR signal of H atoms in a sample of $H + H_2$ at $T = 1.3$ K.

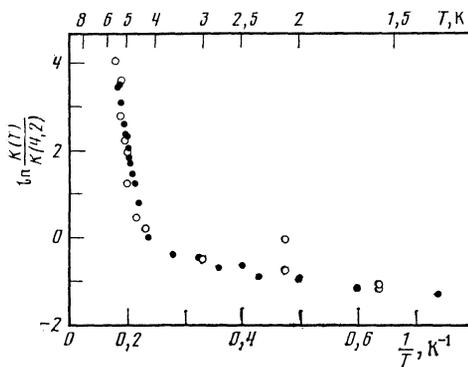


FIG. 4. Dependence of the logarithm of the ratio $K(T)/K(T_0)$ on the reciprocal of temperature: (O) results from Ref. 6 ($B = 0.3$ T); (●) results from Ref. 7 ($B \approx 5$ T).

DISCUSSION OF RESULTS

The idea of suppression of the recombination of hydrogen atoms in strong magnetic fields $\mu B \gg kT$ has been successful in the case of spin-polarized atomic hydrogen in a gaseous medium, whereas the results obtained in the present study showed that the magnetic field does not affect the recombination rate constant of H atoms in a H crystal.

The process of recombination of atoms in a solid has a number of characteristic features: there is a third body (the crystal itself) in each recombination event; diffusion of atoms in a crystal is very slow so that two atoms may be close to one another for a long time; the presence of the crystal lattice results in a rapid spin-lattice relaxation of the electron spin of atoms and this causes depolarization.

We shall bear in mind all these features and use a one-dimensional model to describe the approach and recombination of atoms. Figure 5 shows the singlet and triplet interaction potentials of two hydrogen atoms calculated allowing for the lattice potential. One atom is assumed to be at rest and located at the origin of the coordinate system. Typical values of the parameters used in this figure are $E_a = 100$ K (Ref. 6), $2\mu B \approx 10$ K, and the lattice constant $a = 3.79$ Å. The singlet and triplet potentials were taken from Ref. 11. In a strong magnetic field the electron spins of atoms are aligned (antiparallel to the field) and practically all the atoms interact in accordance with the triplet potential. If an atom is in the second or more distant coordination sphere, depolarization of the electron spin requires overcoming a barrier of $2\mu B$ (~ 10 K in our fields), so that this depolarization process is controlled by the field.

We shall now consider the processes of depolarization of the electron spin in the first coordination sphere. The likely mechanisms of depolarization from the triplet state followed by recombination were considered in Ref. 12 and the authors of that paper estimated for us the probabilities of the processes involving hydrogen atoms in an H_2 crystal (in the case where an atom of hydrogen was in the first coordination sphere).

If an H atom is in the first coordination sphere, we can see from Fig. 5 that its energy level in the triplet potential is higher than in the singlet potential (in the singlet state the energy is equal to the energy of the upper vibrational state of

the H_2 molecules). Obviously, in this situation we have to compare the lifetime of an atom in the triplet state when it is lost from the first coordination sphere τ^* with the characteristic depolarization time due to recombination in the first coordination sphere τ_g . If the condition $\tau^* \gg \tau_g$ is satisfied, the atoms which approach each other to within the first coordination sphere form a molecule. We shall now estimate τ^* and τ_g .

1. We can use the experimental results to find τ^* if we assume that the diffusion of atoms in a crystal is homogeneous. A typical recombination time of atoms in our experiments at $T \approx 1.3$ K for $n_H/n_{H_2} \sim 10^{-4} - 10^{-3}$ was 10^4 sec ($\tau_{1/2} \sim 10^4$ sec). The probability of recombination in one jump (i.e., the probability of encountering a second atom on transition from one position in a crystal to the neighboring one) is

$$p \sim n_{II}/n_{II_2} \sim 10^{-4} - 10^{-3},$$

so that we have $\tau_{1/2} \sim \tau^*/p$ or $\tau^* \sim \tau_{1/2} p \sim 1 - 10$ sec.

2. We shall now estimate τ_g .

a) In our case the main (fastest) mechanism is the exchange recombination when of the two hydrogen atoms at least one is in a "mixed" state φ_4 (we shall use the same notation as in Ref. 12) and the states φ_i are described by the following spin wave functions:

$$\begin{aligned} \varphi_1 &= \alpha(^{1/2})\beta(^{1/2}), & \varphi_2 &= \alpha(^{1/2})\beta(^{-1/2}) + \kappa\alpha(^{-1/2})\beta(^{1/2}), \\ \varphi_3 &= \alpha(^{-1/2})\beta(^{-1/2}), & \varphi_4 &= \alpha(^{-1/2})\beta(^{1/2}) - \kappa\alpha(^{1/2})\beta(^{-1/2}); \end{aligned}$$

the states φ_1 and φ_3 are "pure", whereas φ_2 and φ_4 are "mixed"; $\kappa = A/g\mu B$; A is the hyperfine interaction constant. The recombination time is then

$$\tau_{g1} \sim \frac{1}{\kappa^2} \cdot 10^{-9} \text{ s} \sim 10^{-5} \text{ sec} (B=5 \text{ T}). \quad (1)$$

b) The dipole recombination mechanism involving the interaction with the magnetic moment of a third hydrogen atom is characterized by a depolarization probability (and, consequently, a recombination probability) proportional to the concentration of H atoms. The characteristic time of this process in the case when $n_H \sim 10^{19} \text{ cm}^{-3}$ is

$$\tau_{g2} \sim 10^{-3} \text{ sec}. \quad (2)$$

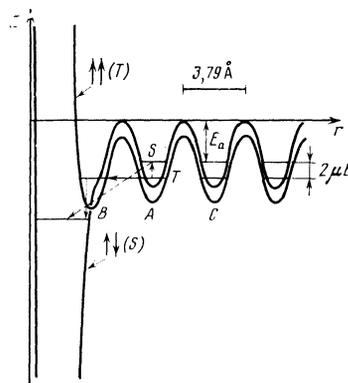


FIG. 5. Singlet and triplet interaction potentials of two H atoms in the presence of the lattice potential: $\uparrow\uparrow(T)$ triplet potential; $\uparrow\downarrow(S)$ single potential. Here, E_a is the activation energy of diffusion (~ 100 K); $2\mu B \sim 10$ K.

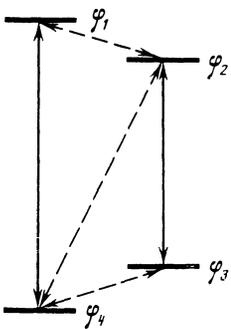


FIG. 6. Electron-nuclear transitions in a hydrogen atom subjected to a magnetic field.

c) Moreover, depolarization is possible because of the interaction of hydrogen atoms with orthomolecules in the environment. A characteristic time of this process is

$$\tau_{g3} \sim 10^{-1} \text{ sec.} \quad (3)$$

Therefore, the characteristic times of all three depolarization mechanisms in the first coordination sphere satisfy the condition

$$\tau^* \gg \tau_{gi} \quad (4)$$

and, consequently, they result in the necessary recombination of atoms if they approach each other to within the first coordination sphere. It should also be noted that the depolarization rates in cases b) and c) are independent of the magnetic field; in the case of the process a), we find that, in spite of the fact that this probability depends on the magnetic field ($W \propto \kappa^2$), it is much higher than the probability of loss of an atom from the first coordination sphere ($\tau_{g1} \ll \tau^*$) and, consequently, the corresponding recombination channel is again practically independent of the magnetic field.

In spite of the approximate nature of these estimates, a reserve of 5–6 orders of magnitude in the inequality (4) for the exchange depolarization mechanism accompanied by recombination allows us to state quite confidently that this recombination channel makes the rate constant independent of the magnetic field.

It should be pointed out that when such recombination channels are all suppressed, there still remains a channel (the $A_T-A_S-B_S$ process mentioned above) which is controlled by a magnetic field if

$$\tau_D > \tau^*, \quad (5)$$

where τ_D is the depolarization time in the second coordination sphere; we then have $\tau_D = \tau_e \exp(2\mu B/kT)$ ($\tau_e \sim 10^{-2}$ sec is given in Ref. 13). It follows from Eq. (5) that the last recombination channel can be suppressed provided the field obeys $B/T > (k/2\mu) \ln(\tau^*/\tau_e) \sim 5$. It should be quite easy to satisfy experimentally this condition. We can see from the results reported in the present paper and the above estimates that the main recombination channel is the recombination from the triplet state in the first coordination sphere, which—in the first approximation—is independent of the magnetic field. We shall consider possible ways of suppressing this recombination channel. The probability of recom-

bination in accordance with the mechanism b) depends linearly on the concentration of H atoms and, therefore, when this concentration is lowered, the probability can be made sufficiently small. The recombination mechanism associated with the presence of orthomolecules in the matrix, called channel c) above, is inapplicable to the case when a crystal consists of parahydrogen molecules. Therefore, there remains the very strong recombination channel a) associated with the exchange interaction and suppression of this channel is possible only when atoms of hydrogen in solid H_2 are in the pure state. We shall now give briefly the results of experiments designed to form a system in which the majority of hydrogen atoms is in the pure state.

It is shown in Ref. 13 that under the conditions in our system the hydrogen atoms may be polarized not only in respect of the electron spin but also in respect of the nuclear spin. This is achieved by microwave pumping of the $\varphi_3-\varphi_2$ transition (Fig. 6), as a result of which the relaxation $\varphi_2 \rightarrow \varphi_4$ and the competing $\varphi_4 \rightarrow \varphi_3$ processes can ensure that the ratio of the populations of the states φ_3 and φ_4 is $n_{\varphi_3}/n_{\varphi_4} \approx 4$. The experimentally determined typical nuclear relaxation time for the $\varphi_4 \rightarrow \varphi_3$ process is of the order of 10^3 sec. We can easily see that in the case of pumping of the $\varphi_4 \rightarrow \varphi_2$ transition the fast electron relaxation process $\varphi_2 \rightarrow \varphi_3$ transfers a hydrogen atom to the pure state φ_3 . Such pumping was produced experimentally and the ratio of the populations of the φ_3 and φ_4 states amounting to $n_{\varphi_3}/n_{\varphi_4} \approx 8$ was obtained; as in Ref. 13, the characteristic nuclear relaxation time was of the order of 10^3 sec. Therefore, periodic pumping at the frequency of the $\varphi_4-\varphi_2$ transition could be used to form a system of H in H_2 in which the majority of hydrogen atoms are in the pure state. The long nuclear relaxation time τ_n compared with τ^* could ensure suppression of the exchange recombination mechanism when two atoms approach each other in the pure state.

CONCLUSIONS

1. It was found experimentally that the recombination rate constant of hydrogen atoms in solid H_2 was independent of the applied magnetic field right up to 7.5 T at temperatures $T > 1.3$ K.

2. An analysis of the experimental results and theoretical estimates showed that the most probable mechanism of recombination of H atoms in solid H_2 occurs from the triplet state in the first coordination sphere and it depends weakly on the external magnetic field.

3. The methods for suppression of this recombination channel were suggested.

4. It was shown experimentally that microwave pumping of transitions in the electron subsystem can ensure a strong polarization of hydrogen atoms in respect of their nuclear spin.

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