Hydrogen and deuterium atoms, stabilized by condensation of an atomic beam in superfluid helium

E. B. Gordon, A. A. Pel'menev, O. F. Pugachev, and V. V. Khmelenko
Institute of Chemical Physics, Academy of Sciences of the USSR

(Submitted 1 February 1983)
Pis'ma Zh. Eksp. Teor. Fiz. **37**, No. 5, 237–239 (5 March 1983)

Concentrations of H and D atoms, stabilized in a molecular matrix, up to 1×10^{20} cm^{-3} are achieved by condensation of an atomic beam in superfluid helium. Transformation of D atoms into H atoms is observed, indicating the occurrence of tunneling chemical exchange reactions between atomic and molecular hydrogen isotopes.

PACS numbers: 35.10. – d, 67.90. + z, 82.30.Hk

Collective quantum effects must be manifested most strongly for hydrogen atoms. For this reason, the problem of accumulating high concentrations of $H(n_H)$ at low temperatures is currently being persistently studied. Work on stabilizing H atoms in the gas phase, spin-oriented in strong magnetic fields ($B\cong 11T$ for $T\lesssim 0.3$ K), has been very successful.[1] However, only concentrations of 10^{17} cm^{-3} have now been achieved and for a number of reasons it is difficult to expect a large improvement.

For the more traditional method of stabilizing hydrogen atoms in a solid molecular matrix, the most important factor restricting n_H to the 10^{18} cm^{-3} level is the instability of a finite specimen relative to thermal explosion, initiated by the slow, "dark" recombination of atoms.[2] For this reason, when H atoms are frozen out of a

gas onto a cold surface, only effects such as quantum diffusion of atoms can be investigated.[3]

From this point of view, the methodical approach developed by us for nitrogen atoms, which is based on condensation of an atomic beam by introducing it into superfluid helium,[4] has great advantages: Due to preliminary cooling of the beam to $T \lesssim 10$ K, contact of "warm" atoms with the condensate is eliminated, while the high dispersity of the condensate (particle diameter $\sim 10^{-4}$ cm), together with the anomalously high thermal conductivity of He II, increase the values of n_H required for the appearance of a thermal explosion.

In the experiments described in this paper, hydrogen and deuterium atoms were directed from the zone of an hf discharge in the form of a beam onto the surface of He II, filling a specially shaped beaker. Having reached the bottom of the beaker, the atoms were detected by the EPR method using a specially built attachment.[5] The temperature of the He II at the time of condensation was 1.8 K.

The EPR spectrum of H atoms (1), shown in Fig. 1 (starting mixutre H_2: Ne:He = 1:1:40, neon was added so that the condensate containing the H atoms would sink in liquid He), is, in accordance with Ref. 6, a characteristic doublet of distorted broad lines with 508-Oe splitting and g factor of 2.002. The spectrum of the D atoms (2) had the opposite shape (splitting of 76.7 and 78.7 Oe, respectively, and line width 5.8 Oe). Characteristically, however, the spectrum of H atoms was saturated even at uhf power of 1 μW, while for D atoms saturation did not occur even at power of 100 μW.

In agreement with Ref. 3 H atoms were considerably less stable than D atoms.

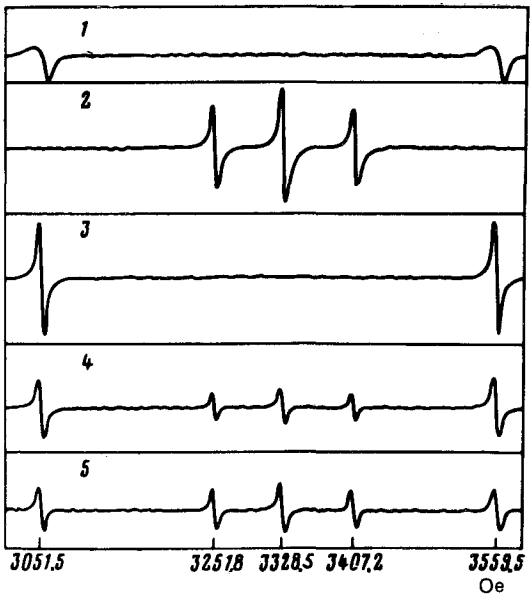

FIG. 1. EPR spectra of H and D atoms for different mixtures: 1—H_2:Ne: He = 1:1:40; 2—D_2: He = 1:20; 3—H_2:D_2:Ne: He = 1:1:1:60; 4—H_2:D_2: He = 1:4:100; 5—H_2:D_2: He = 1:10:220.

With heating (3×10^{-3} K/s H atoms were no longer observed at $T>2.55$ K; and a sharp decrease in the EPR signal was observed in a number of cases during the transition through the λ point ($T=2.19$ K), confirming our idea[2] that for systems containing atoms it is possible to have a thermal explosion with a transition through the λ point. Holding at $T=1.8$ K for 3×10^3 s decreased n_H by a factor of 2–3. On the other hand, a similar exposure of D atoms did not change the magnitude of the signal. Moreover, n_D did not change appreciably even when the specimen was held at $T=4.2$ K for 10^4 s. A neon impurity in the starting mixture did not affect any characteristics of the EPR signal of D atoms.

In view of the differences indicated above, it was of great interest to investigate mixtures containing simultaneously hydrogen and deuterium (see Fig. 1). Surprisingly, it turned out that a) for starting mixtures with composition H_2:D_2 = 1:1, the signal from D atoms is missing, while the signal from H atoms is no longer saturated, and b) even with a content we have D_2 = 1:10 $n_H = 0.6\, n_D$; c) H atoms in matrices containing deuterium were almost as stable as D atoms relative to isothermal holding and heating.

Since there is no doubt that in an hf discharge the yields of H and D atoms are approximately proportional to the partial concentrations of H_2 and D_2 in the starting mixture, respectively, the much higher concentrations of stabilized H atoms in deuterium-hydrogen mixtures indicates that even at low temperatures most of the D atoms transform into H atoms in chemical exchange reactions of the atom and molecule. Indeed, the tunneling channel in such reactions is appreciable (an estimate of the rate of tunneling reactions $H + H_2$ and $D + H_2$, according to data from the calculation in Ref. 7 and the height of the barrier for diffusion of atoms $E = 30$–100 K, gives for $T\to0$, the quantity 1–10^4 s^{-1}), but it is manifested only for reactions in which H atoms transform into H atoms, while D atoms transform into D atoms and H atoms (other reactions, due to the difference in the magnitude of the zero-point vibrations of the starting molecule and the product molecule, are endothermal by an amount 5–10 kJ/mole). As a result of these processes and the usual quantum diffusion, the starting and forming H atoms migrate along the matrix, until they reach the deuterium "coat," stabilizing them relative to displacements and, correspondingly, relative to recombination.

The fact that the characteristic lifetime of H atoms in a molecular matrix, τ_H at $T\cong2.55$ K in our experiments is less than 10^2 s (in Ref. 3, at these temperatures, $\tau_H\cong10^4$ s) indicates that the concentrations n_H obtained by us are much greater than those obtained in Ref. 3. Estimates from calorimetric measurements of the magnitude of the flux condensing in He II give the following values for mixtures 1–5 (Fig. 1): 1—$[H]/[H_2] + [Ne] = 0.1\%$; 2—$[D]/[D_2] = 0.6\%$; 3—$[H]/[H_2] + [D_2] + [Ne] = 0.4\%$; 4—$[H]/[H_2] + [D_2] = 0.15\%$; $[D]/[H_2] + [D_2] = 0.22\%$; 5—$[H]/[H_2] + [D_2] = 0.04\%$; $[D]/[H_2] + [D_2] = 0.065\%$.

We thank M. Ya. Ovchinnikova for useful discussions.

[1]I. F. Silvera and J. T. Walraven, Phys. Rev. Lett. **44**, 164 (1980).
[2]E. B. Gordon, L. P. Mezhov-Deglin, O. F. Pugachev, and V. V. Khmelenko, Zh. Eksp. Teor. Fiz. **72**, 952 (1977) [Sov. Phys. JETP **45**, 536 (1977)].

[3]A. Ya. Katunin, I. I. Lukashevich, *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **34**, 357 (1981) [JETP Lett. **34**, 357 (1981)].
[4]E. B. Gordon, L. P. Mezhov-Deglin, and O. F. Pugachev, Pis'ma Zh. Eksp. Teor. Fiz. **19**, 103 (1974) [JETP Lett. **19**, 63 (1974)].
[5]E. B. Gordon, A. A. Pel'menev, O. F. Pugachev, and V. V. Khmelenko, Fiz. Nizk. Temp. **8**, 601 (1982) [Sov. J. Low Temp. Phys. **8**, 299 (1982)].
[6]C. K. Jen, S. N. Foner, E. L. Cochran, and V. A. Bowers, Phys. Rev. **112**, 1169 (1958).
[7]T. F. George and W. H. Miller, J. Chem. Phys. **57**, 2458 (1972).

Translated by M. E. Alferieff
Edited by S. J. Amoretty