Studies of hydrogen atom recombination in solid hydrogen deuteride

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Abstract

We used the method of electron spin resonance (ESR) to investigate the temperature dependent recombination rate of H atoms in solid molecular hydrogen deuteride (HD). A 1.5 μm thick solid molecular HD film was deposited at a rate of 2 monolayer/s, onto a gold surface maintained at T=1.5 K. H and D atoms were accumulated in the film by maintaining radio-frequency electric discharge above the film for 19 days. After further storage of the sample for 30 hours, at T $<$ 1 K, the D atom signal vanished. The concentration of H atoms was monitored as the sample was warmed stepwise from 1.1 K to 2.8 K. The recombination rate of H atoms in solid HD was found to be proportional to temperature in this range.

Keywords: Hydrogen atoms, Hydrogen deuteride, Electron spin resonance, Tunneling chemical reactions

1 Introduction

Solid matricies of molecular hydrogen isotopes are fascinating objects to study quantum tunneling reactions with participation of atomic hydrogen isotopes [1–7]. The recombination rates of H atoms in solid H_2 and D atoms in solid D_2 were measured

at low temperatures by using the method of electron spin resonance (ESR) [2, 3, 8, 9]. The exchange tunneling reactions D+H_2 → HD+H and D+HD → D_2+H, resulting in increasing number of H atoms and decreasing the number D atoms, were studied in mixed H_2-D_2 solids [4–6, 10–12] and in solid hydrogen deuteride (HD) [13]. Measurements described in Ref. [13] were performed just after finishing the process of accumulation of H and D atoms in the HD film. The rate constants for the exchange tunneling reaction D+HD → D_2+H were determined by recording the decay of D atom concentration shortly after finishing the accumulation process. Usually the exchange tunneling reactions were studied in the conditions when the total number of atomic species (H and D) in the sample was constant.

Previously attempts were made to measure the exchange tunneling reaction rate in D_2+0.5% H_2 solids at temperatures 2.5 and 3.5 K [13]. In these experiments in addition to the conversion of D atoms into H atoms, the total density of atomic species in the sample decreased due to their recombination. Due to this complication the qualitative data were not obtained from these measurements.

In this work we present the first studies of H atom recombination in solid HD matrix. After finishing accumulation of H and D atoms in solid HD matrix we stored the sample at temperature 350 mK for ∼ 48 hours, after which the signal from D atoms disappeared due to exchange tunneling reactions. We then monitored decay of H atoms in the HD matrix at different temperatures. The decrease of H atoms in the sample was due to the second order reaction of H atom recombination. The measured recombination rate constants for H atoms in HD solids, in the temperature range from 1.1 K to 2.8 K, are presented in this article.

2 Experimental setup

This experiment was conducted using a commercial Oxford 200 dilution refrigerator. Electron spin resonance (ESR) method was used to obtain the data presented in this work. For the ESR measurements, a cryogenic mm-wave (∼ 128 GHz) ESR spectrometer[14] was used. The HD sample was prepared and studied in the sample cell (SC), located in the center of a 4.6 T superconducting magnet, which provided the biasing magnetic field for the ESR measurements. In addition to the main 4.6 T magnet, two additional coaxial superconducting coils were present in the experimental assembly. A smaller solenoid wound on the same coil form as the main magnet, used to vary the magnetic field (±500 G) for cw-ESR measurements. And an anti-helmholtz coil wound around the vacuum can of the dilution unit, used to apply or remove an axial magnetic field gradient (±20 G/cm) to the sample.

The SC assembly used in this experiment was identical to the cell used in our previous work[15, 16]. A more detailed description of the sample cell may be also found in Ref.[17].

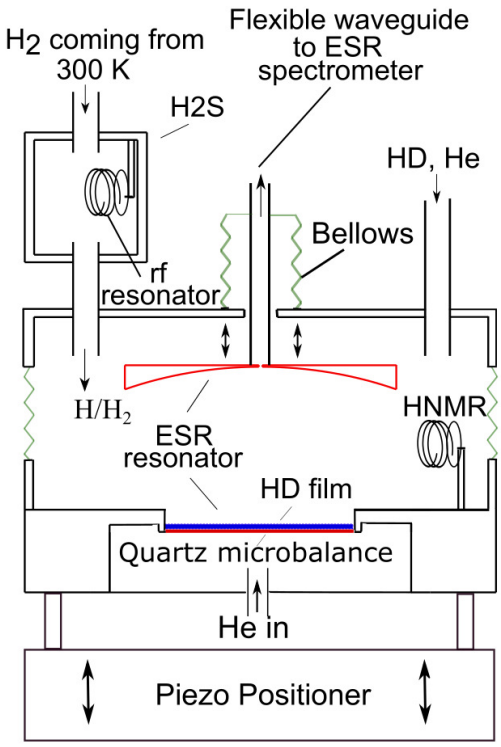

Fig. 1 Simplified schematic of the sample cell.

The central feature of the SC is the mm-wave Fabry-Pérot resonator used in the ESR measurements. The Fabry-Pérot cavity consists of an upper spherical mirror, made of polycrystalline copper, and a lower flat mirror, gold plated quartz disc. In addition to acting as the flat mirror of the resonator, the gold plating doubles an electrode for measuring the resonant frequency of the quartz disc, allowing the lower mirror to be operated as a quartz microbalance (QM). By monitoring the resonance frequency of the QM, the thickness of the deposited film on the bottom mirror is measured.

In addition to the mm-wave resonator, the SC contains a 950 MHz helical resonator and 2 capillaries connecting the room temperature gas handling system to the cell. The helical resonator was used to produce radio-frequency (RF) electric discharge in the SC, by driving high-power RF pulses. The electrons generated by the RF discharge, with energies $\sim$ 100 eV, bombard the HD film producing atomic species within the film by, $\mathrm{HD} + h\nu_e \rightarrow \mathrm{H} + \mathrm{D}$. The two capillaries were both directed to the center of the lower flat mirror of the FP resonator, they are nearly identical but serve different purposes. The first capillary provides a direct connection from the room temperature gas handling system to the SC, and is used to supply gasses which form the sample under study. The second capillary has a small volume, containing a second helical resonator, along it's path the SC. At the beginning of the experiment H_2 gas was condensed in this volume, and driving the helical resonator produces spin polarized

H↓ gas flux directed into the SC. This volume is the H_2 source (H2S), which can also be used as a source of H_2 molecules upon increasing temperature to 4 K.

2.1 Sample preparation

At the beginning of the experiment, the solid HD film was condensed on the flat bottom mirror of the Fabry-Pérot resonator, by supplying HD gas to the SC from a fixed volume, with pressure ~ 5 torr, integrated in the room temperature gas handling system. The film deposition rate was maintained at ≈ 2 monolayer/s and the SC temperature was maintained at 1.5 K, throughout the film deposition process. Most of the HD molecules condensed on the flat mirror, cooled from the other side by superfluid ^{4}He, which allowed for efficient removal of the heat released during condensation. The HD film was grown to an overall thickness of 1.5 μm, controlled by the QM.

After formation of the solid HD film, the SC was first evacuated, then a small quantity of He was condensed in the cell. Spin polarized H↓ gas, supplied to the cell by the H2S, was then used for initial tuning of the ESR spectrometer. After registration of the H↓ atoms in the gas phase, the discharge in H2S was stopped.

Discharge was then started in the SC by using the HNMR coil, and maintained for the following 19 days, for accumulation of H and D atoms in the HD film. The sample cell temperature was maintained at 650 mK throughout the duration of the accumulation stage, which corresponds to a saturated vapor pressure of ≈ 1 mtorr for ^{4}He [18].

After finishing of the accumulation stage of the experiment, discharge in the SC was stopped and then discharge was started in the H2S to supply H↓ into the SC and perform absolute calibration of the ESR method. A calorimetric method, described in detail in Ref.[17], involving measurement of the heat release from H↓ atom recombination, was used to calibrate the number of H and D atoms stabilized in the HD film.

As a next step we performed measurements of recombination rates of hydrogen atoms in solid HD films at different temperatures, in the range from 1.1 to 2.8 K. Measurements of the recombination rates were conducted by stabilizing the SC at fixed temperature and continuously measuring the decrease of H atom ESR signals during period of a few hours.

3 Experimental Results

3.1 ESR spectrum

The full absorption ESR spectra taken at various stages of the experiment are presented in Fig.2. The ESR spectrum of H in solid HD is a doublet signal consisting of the low field line (a-line; H_a) and the high field line (b-line; H_b) separated by ~ 508 G. In addition to the two H lines four other lines were observed in this experiment, all near the center of the H spectrum. Three of these lines (low-field; D_{LF})(center-field; D_{CF})(high-field; D_{HF}), separated by ~ 78 G, are attributed to the triplet signal of D atoms in the sample. The remaining line was a broad (~ 100 G) resonance located near the center of the H atom spectrum, labeled the broad central line (BCL)[19]. The

broad central line in the spectrum was observed in all stages of the experiment. The integrated area of the BCL was on the same order of magnitude as the sum of the hydrogen a and b line integrals. However, the linewidth was more than an order of magnitude larger. The shift ($\sim$ 20 G) of the BCL center relative to the center of the H atom spectrum, observed between the green and red curves, correlates to removing the current from gradient coil which maintained high field homogeneity throughout the experiment. Therefore, we consider the spin-containing species responsible for the BCL was not located in the solid HD sample, but rather on the metallic surfaces of the top spherical mirror of the FP resonator. The BCL can be associated with either H–H radical pairs with a strong exchange interaction, or caused by OH radical adsorbed on the spherical mirror of the FP resonator [19].

The upper trace in Fig. 2 shows that, in the ESR spectrum, rather small D atom lines were present during the accumulation process, when RF discharge was running in the SC. It was found that D atom lines also can be seen for some period (2–3 hours) after stopping discharge. D atoms participated in exchange tunneling reactions with HD molecules: D+HD $\rightarrow$ D_2+H, which result in disappearance of D atom ESR spectra, and increasing of intensity of H atom ESR spectra, as seen in the center trace of Fig. 2. We started measurements of H atom recombination in solid HD after storage of the sample for 30 hours at T $<$ 1 K without running discharge. The lower trace of Fig. 2 shows the ESR spectrum at the end of H atom recombination measurements. One can see that the signal of H atoms in this ESR spectrum was much smaller than at the beginning of measurements.

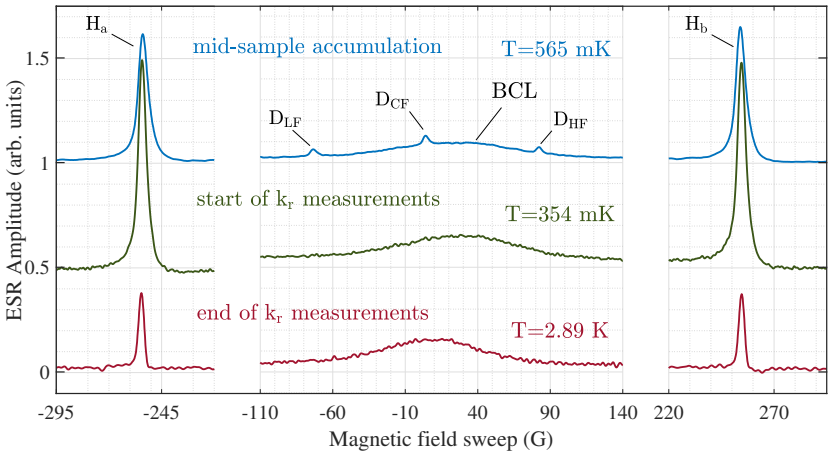

Fig. 2 ESR absorption spectra of the sample, centered about the H-atom resonances, at various stages the experiment. Upper trace (blue curve) ESR spectrum after 10 days of accumulation, $\approx 1/2$ total sample accumulation time. Central trace (green curve) spectrum measured at the beginning of the recombination measurements. Lower trace (red curve) final spectrum measured at the end of the recombination measurements.

3.2 Determination of concentration of H atoms, [H], in solid HD

The concentration of H atoms, [H] (atoms/cm^{-3}), in the HD sample was determined by measuring the broadening of the ESR linewidth due to dipole-dipole interaction of neighboring H atoms. The formula[20] relating ESR linewidth broadening to local concentration of H atoms is provided by Eq. 1,

$$\Gamma = 1.06 \times 10^{-19}\,[\mathrm{H}] + \Gamma_0 \tag{1}$$

where Γ is the measured full width at half maximum (FWMH) linewidth in Gauss of the H atom ESR signal. And Γ_0 is the FWHM contribution due to nuclear dipole moments of the HD molecules in the HD crystal, taken as the lowest measured FWHM of H atoms in HD during the experiment, 2.6(1) G, in close agreement with [21].

3.3 Measurement of H atom recombination rate (k_r) in solid HD at fixed temperature

Recombination rates of H atoms in solid HD were measured by stabilizing the sample temperature, then measuring the decay of the concentration of H atoms over time scales on the order of hours. As an example, Fig. 3a shows the dependence of H atom concentration on time at temperature 2.8 K. The decay is well described by the second order recombination reaction H+H$\rightarrow$H$_2$. Fig. 3b shows that time dependence of the reciprocal of H atom concentration is linear, with slope equal to $2k_r$ according to Eq. 2.

$$\frac{1}{[\mathrm{H}]} = \frac{1}{[\mathrm{H}]_0} + 2k_r t \tag{2}$$

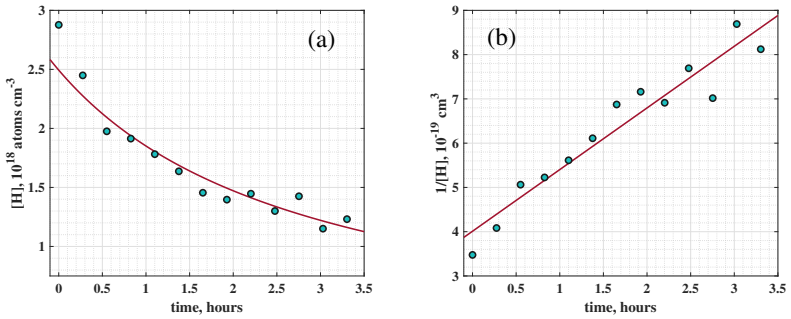

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Fig. 3 H atom concentration evolution in time, at constant temperature, T= 2.8 K. (a) dependence on time of local concentration of H atoms, [H], (b) dependence of reciprocal of H atom concentration, $\frac{1}{[\mathrm{H}]}$ on time. Measured local concentration-(teal circles), fit curve-(red line).

3.4 Temperature dependence of H atom recombination rate in solid HD

Results for the temperature dependence of the H atom recombination rate k_r, for the temperature range 1.1–2.8 K, are presented in Fig 4, and Table 1. Fig. 4 shows the dependence of the logarithm of the recombination rate constants on reciprocal temperature. Changing the temperature of the solid HD sample from 1.1–2.8 K led to a substantial increase, more than three orders of magnitude, of the recombination rate of H atoms in solid HD, from 2.87×10^{-27} to 1.18×10^{-23} cm^3/s.

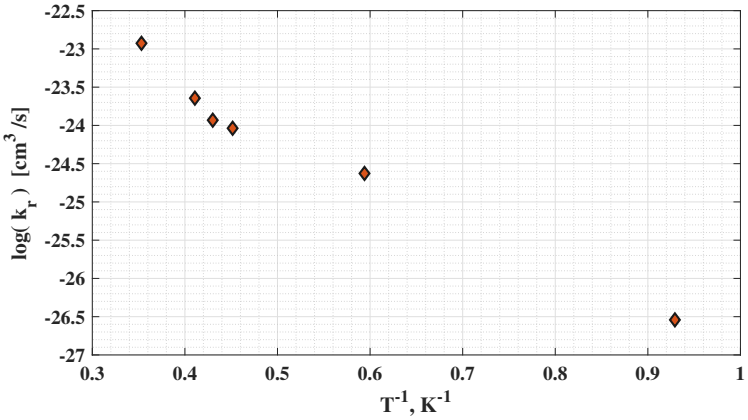

Fig. 4 Temperature dependence of H atom recombination rate in HD matrix.

Table 1 Temperatures of measurements, recombination rates of H atoms in HD matrix, initial concentrations of H atoms and time intervals of measurements.

T, K	k_r, cm^{3}s^{-1}	$[H]_{initial}$, atoms cm^{-3}	$\Delta t_{measured}$, hours
1.1	2.87×10^{-27}	7.80×10^{18}	6.6
1.7	2.36×10^{-25}	1.62×10^{19}	2.3
2.2	9.17×10^{-25}	1.11×10^{19}	1.2
2.3	1.17×10^{-24}	1.01×10^{19}	0.57
2.4	2.27×10^{-24}	9.05×10^{18}	0.41
2.8	1.18×10^{-23}	5.12×10^{18}	3.6

4 Discussion

In this work we have presented the first ever measurements of the temperature dependence of H atom recombination rate for H in solid HD matrix. Previous measurements have been performed for the similar systems of H in solid H_2 and D in solid D_2[3]. Figure 5 shows a comparison of the temperature dependences of recombination rates on temperatures, of atoms in different quantum solids: H in solid H_2[3, 20], D in solid

D_2 [3], D in D_2-He nanoclusters, and H in solid HD. Only one measurement, at 1.35 K, was taken for the recombination rate of D atoms in D_2-He nanoclusters[22]. The measured rates of the exchange tunneling reaction[13] D+HD→ D_2+H, which exhibits no temperature dependence from 1–4 K, are also shown in Fig. 5.

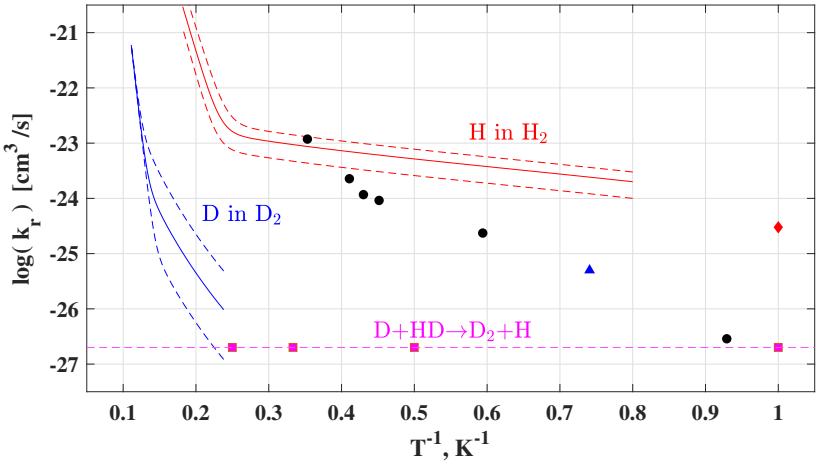

Fig. 5 Measurements presented in this work compared with previous results. Temperature dependence of recombination rates of H in HD (black circles), D in D_2 [3] (blue curve), H in H_2 (red curve[3] and red diamond[20]), D in D_2-He nanoclusters [22] (blue triangle), and rate of tunneling reaction D+HD → H+D_2 [13] (magenta curve and magnenta squares).

Both systems, H in H_2 and D in D_2 exhibit a clear transition from a high temperature regime, in which $k_r(T) \sim \exp(-\mathrm{E_a/T})$ is governed by classical diffusion processes, to a low temperature regime $k_r(T) \sim BT^n$ governed by quantum tunneling diffusion, at temperatures 4 and 7.5 K respectively [3]. Although we did not observe such a clear transition in our experiment, it is reasonable to expect that for the temperature range of this experiment, the recombination rate is in the regime governed by quantum diffusion processes.

The first argument supporting our claim that the presented measurements were likely governed by the quantum tunneling regime, is the relatively weak temperature dependence of $k_r(T)$. Fitting the experimental data using the Arrhenius formula, Eq. 3 with $B \to 0$, we calculate an activation energy $E_a \approx 10$ K, for the H in HD recombination reaction. In contrast, the measured activation energies for the reactions H in H_2 and D in D_2 were 103(5) K and 270(30) K, respectively, more than an order of magnitude larger than in this experiment. Additionally the calculated value for A, the $T \to \infty$ asymptote of k_r, using this fit model gives log(A) = -21.5, which is substantially smaller than the previously reported values log(k_D^0)=-8.2 for D in D_2, and log(k_H^0)=-12.4 for H in H_2.

$$k_r(T) = Ae^{-E_a/T} + BT^n \tag{3}$$

In contrast, if we fit the experimental data using only the second term in Eq. 3, we calculate log(B) = -26.7, n = 8.1. Which is in much better agreement with the values obtained for D in D_2; log(B_{D_2}) = -31.5, n = 8.8, and H in H_2, log(B_H) = -23.7, n=1. Additionally, comparing the fit results using the residual sum of squares metric, we find that the fit obtained from the quantum tunneling model $k_r(T) \sim BT^n$ is better by a factor of ≈ 2.

The expected mechanism of quantum diffusion is similar for the case of H in HD compared to that of D in D_2 and H in H_2. In all cases we deal with the tunneling exchange reaction, e.g. H+H_2 $\rightarrow$ H_2+H. In these formulae, one of the atoms of the molecule tunnels (to the left) through the barrier, leaving a free atom in the right. If the lattice is without defects and there is no other impurity atom nearby, such exchange occurs without change of energy. For the H+DH$\rightarrow$HD+H reaction, the D atom moves to the left. Or the atom should physically exchange with the molecule, like it happens with the diffusion of ^{3}He in solid ^{4}He[23, 24]. For low temperatures, when the overbarrier (Arrhenius) diffusion is frozen, the rate of H in HD tunneling should be much smaller because the D atom is heavier than H, and HD is heavier than H_2. We observe this: the difference from H in H_2 is two orders of magnitude at 1 K.

Our results show that the H atom recombination rate in solid HD exhibits a stronger temperature dependence than the recombination rate of H atoms in solid H_2. However, the recombination rate measured for D in solid D_2-He nanoclusters, at 1.35 K, is close to that obtained for H in solid HD.

5 Conclusion

1. The observed temperature dependent recombination rate for H in solid HD is monotonically increasing with temperature.
2. All measured recombination rates in this experiment were at least an order of magnitude greater than the rate of the tunneling reaction D+HD $\rightarrow$ H + D_2 measured previously[13].
3. The temperature dependence of H atom recombination rate k_r for H in HD is much stronger than for previously measurements of H in H_2, over comparable temperature range.

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