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Nonlocal distribution of the recombination energy in spin-polarized atomic hydrogen

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One of the important but little studied characteristics of spin-polarized hydrogen is the enormous energy release due to surface recombination into molecular H_2. We have studied the relaxation and distribution of this energy throughout a sample. A phenomenological parameter, F, defined as the fraction of total recombination energy released locally at the point of recombination has been measured. Our upper bound of $F<0.04$ demonstrates that very little energy is directly coupled to the surface at this point.

The release of recombination energy in a gas sample of spin-polarized hydrogen can lead to significant heating of the sample. Recombination processes have presented a severe barrier to achieving Bose-Einstein condensation (BEC) in high-density spin-polarized hydrogen ($H\downarrow$). The immediate effect of recombinative decay is a loss of atoms and thus a loss of density. However, the primary limitation is the substantial heat release from an $H+H+X\rightarrow H_2+X$ recombination event, the most energetic chemical reaction per unit mass known, that has so far kept high-density $H\downarrow$ experiments well away from temperatures required for BEC. In fact, when these conditions are approached, the recombinative heating often becomes a runaway, positive feedback process, ending spectacularly in an explosive destruction of the sample.[1–3]

In the 14 years since the stabilization of $H\downarrow$, there have been numerous studies of the rates at which various recombination and relaxation processes proceed. However, only recently have there been any studies of how the energy released during a recombination event is distributed throughout the sample.[4,5] In various models of the decay of a high-density $H\downarrow$ gas, the total energy release of $D_0=51\,967$ K (Ref. 6) has been assumed to be distributed either uniformly throughout the sample or deposited entirely at the point of recombination.[3,7] Knowledge of the details of the process is important not only for our fundamental understanding of this system, but also for the design of experiments to produce BEC. In this paper we study the energy distribution for a particular process, two-body recombination on a helium surface. We find that only a very small fraction of the energy is deposited locally at the point of recombination; F, defined as the fraction of the recombination energy which is deposited locally, is less than 0.04. The process by which the energy is distributed is discussed below.

Initially, when two $H\downarrow$ atoms recombine, a transition is made from the $^3\Sigma_u^+$ potential, which has no bound states, to the $^1\Sigma_g^+$ potential. A highly excited H_2^* molecule is formed. Because of the momentum mismatch between the initial and final states, cold ($T<1$ K) H atoms are not likely to recombine into deeply bound molecular states. Figure 1 shows some of the bound, rovibrational levels of the H-H singlet potential closest to the continuum. Recombination takes place with the highest probability for the smallest energy change between initial and final states. With a high probability, recombination produces H_2^* in the $(v,J)=(14,4)$, (14,3), or (14,2) rovibrational states with a binding energy of 0.26, 72, and 137 K, respectively.[8] Thus, only of order 100 K or less ($<2\times10^{-3}D_0$) is released as kinetic energy in the initial recombination step. The rest of the energy is released later in a series of steps via collisional relaxation as the H_2^* cascades down the ladder of over 350 possible states to the ground level. These steps must also be of order a few hundred degrees kelvin, the smallest quantized energy step, in order to minimize once again the momentum mismatch between initial and final states.

Our technique for determining F can be understood from basic knowledge of the hydrogen-helium-surface system.[9] Atomic hydrogen adsorbs on the helium surface as a two-dimensional gas; in equilibrium σ the surface coverage of H, is related to the bulk gas density n by $\sigma=n\lambda\exp(E_a/k_BT)$, where T is the temperature of the surface and the $H\downarrow$ gas, $\lambda=(2\pi\hbar^2/m_Hk_BT)^{1/2}$ is the

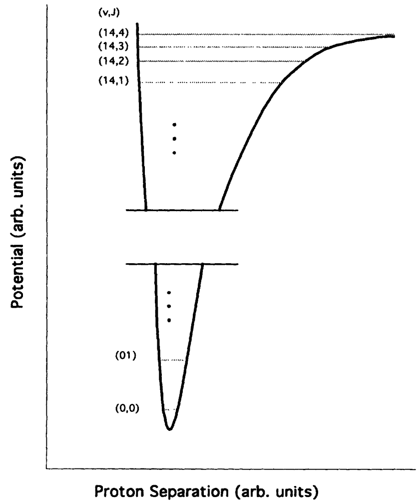

FIG. 1. The singlet H-H potential, showing some of the rovibrational states near the continuum. There are other states near the continuum besides those shown here, but they have high angular momentum J and are not likely final states after a recombination event, due to a very large centrifugal barrier.

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thermal de Broglie wavelength, m_H is the mass of an H atom, and $E_a \approx 0.96$ K is the adsorption energy of an H atom on liquid ^{4}He.[10] At low surface coverage, recombination takes place at a quadratic rate, $d\sigma_i/dt = -K_{ij}\sigma_i\sigma_j$, where i,j designates the hyperfine spin states of the participating hydrogen atoms (for electron spin-down hydrogen there are two spin states designated $|a\rangle$ and $|b\rangle$, with $|a\rangle$ being the lowest in energy). Here, K_{ij} is indexed because the recombination rate constants depend sensitively on the hyperfine states.

In our experiment, we use an element with a small surface area which has a weak thermal coupling to a large volume, large surface area cell filled with H↓ and covered with a film of liquid helium. The small surface can be heated to change its temperature and thus the hydrogen surface coverage without affecting the temperature of the large cell or the gas. An ideal surface is a bolometer, shown in Fig. 2. The composite bolometer, which has been described elsewhere[11] is a $0.7 \times 0.7 \times 0.25$ mm^3 chip neutron-transmutation-doped (NTD) Ge bolometer, glued to one side of a $1 \times 1.5 \times 0.025$ cm^3 sapphire plate, with the main thermal contact to the cell being two 25-μm-diam copper wires. The bolometer was carefully calibrated as a sensitive thermometer. It has a total surface area of about 3.0 cm^2. The H↓ sample cell is a copper cylindrical can, with a volume of 387 cm^3 and a surface area of 400 cm^2. It is immersed in a 4-T magnetic field and cooled to a temperature $T_0 = 225$ mK inside a dilution refrigerator. ^{4}He was added to the cell to form a thick (≈ 140 Å) superfluid film covering all surfaces including the bolometer. Atomic hydrogen, formed in a room-temperature microwave discharge, is thermalized to the cell temperature before entering the cell, at a constant rate.

The bolometer temperature could be increased by simply increasing its bias current, which increases the ohmic heating. When the bolometer is in contact with hydrogen it receives additional heating from a fraction F of energy from atoms recombining on its surface and a fraction $(1-F)$ of energy from atoms recombining on cell walls elsewhere, depositing their energy nonlocally on the bolometer. Electrically warming the bolometer surface by a few tens of millikelvin has a dramatic effect on the hydrogen coverage and thus the local recombination rate, assuming all other conditions are held fixed. The local recombination rate, depends on the square of the coverage, and thus exponentially on the local temperature as $\exp(2E_a/k_B T)$.[9,10] This rate can be reduced by more than an order of magnitude so that recombination heating is dominated by the nonlocal source. From a differential analysis of these two regimes, F can be determined. In the analysis that follows, we were able to determine only an upper bound on F rather than its value. This was primarily due to the small value of F and uncertainties in some of the system parameters. The small upper bound finally settles the long standing question of where the recombination energy goes and has important implications for future attempts to achieve BEC in H↓.

Data are taken by biasing the bolometer at several different currents and measuring the resulting voltage drops across the Ge chip, so that a power versus temperature characteristic can be determined for the bolometer at a given hydrogen density. This characteristic represents the amount of electrical power which must be dissipated in the bolometer in order to raise it to a certain temperature. When H↓ is present, it gives information about the net power deposited on the bolometer surface by the H↓ gas as a function of the bolometer surface temperature, T_b. We made measurements with H↓ densities between 2.0×10^{13}/cm^3 and 9.0×10^{13}/cm^3, and bolometer temperatures between 225 and 290 mK. The raw data, acquired at ambient conditions of $T_0 = 225$ mK and $B_0 = 4$ T, are shown in Fig. 3. Clearly from Fig. 3, as the bolometer temperature is raised, there is a crossover from a net heating of the bolometer by the H↓ (due to recombination) to a net cooling of the bolometer by the H↓ (due to the thermal coupling it provides to the cell walls). The

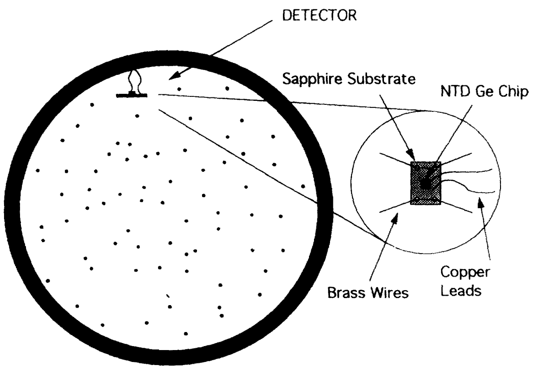

FIG. 2. Schematic of the spin-polarized hydrogen cell with the composite NTD Ge bolometer. The fine copper leads and brass support wires provide very weak thermal coupling to the cell, as well as a path by which superfluid ^{4}He can flow onto the bolometer substrate.

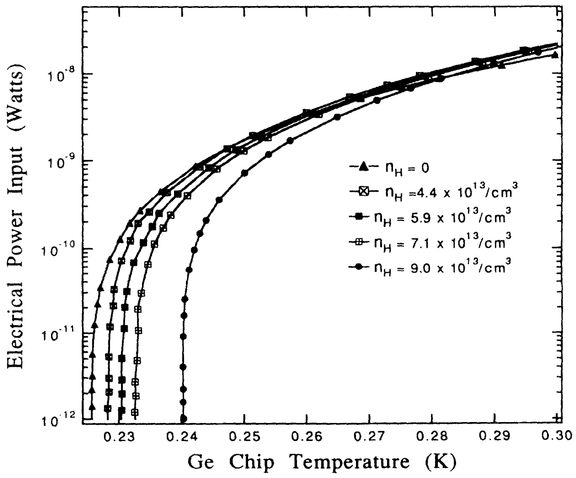

FIG. 3. Electrical power vs bolometer temperature data, taken at several different H↓ densities: $n = 0; 4.4 \times 10^{13}$/cm^3; 5.9×10^{13}/cm^3; 7.1×10^{13}/cm^3; and 9.0×10^{13}/cm^3.

cooling provided by the H↓ gas is characterized by an accommodation coefficient α which has been measured previously.[12,13] As the H↓ density is increased, the crossover point moves to higher bolometer temperatures, since the recombination heating goes as n^2, and the accommodation cooling effect goes as n.

In the conditions used for our measurements, over 99% of the decay of H↓ is due to the two-body surface recombination process. The densities are too low for three-body or higher-order recombination to be important, and the temperature is too low for resonant bulk recombination to be appreciable.[1–3,14,15] The density is low enough that the mean free path for relaxation of H_2^* by collision with atoms in the gas phase is much larger than the cell dimensions.[16] Though the cell is filled continuously during a measurement, the H↓ sample may still develop a small steady-state nuclear polarization, $p=(n_b-n_a)/(n_a+n_b)$, due to the preferential recombination of the $|a\rangle$-state atoms.[17] Here, n_b is the density of $|b\rangle$-state atoms in the sample, and n_a is the density of $|a\rangle$-state atoms. Nuclear-spin polarization of the sample can become a problem in the analysis because the H↓ decay rate depends sensitively on p and decreases quickly as p increases. By measuring density decay curves, we determined experimentally that $p\approx 0$.

In order to analyze our data we fit the power vs temperature curves with cubic splines and then subtracted the curves with $n>0$ from the one with $n=0$. We call this the difference power, ΔP,

$$\Delta P(T_b)\equiv P_{\text{loc heat}}+P_{\text{nonloc heat}}-P_{\text{cool}}\ ; \tag{1}$$

the three terms will be discussed below. ΔP, as a function of bolometer temperature T_b is shown in Fig. 4 for $n=5.9\times10^{13}/\text{cm}^3$. ΔP is due entirely to the presence of the H↓ gas, which affects the power curve in only two ways: by providing recombination energy, which heats the bolometer; and by providing a conduction path to the cell, which cools the bolometer.

The additional power deposited on the bolometer by the H↓ comes from two terms, one for local recombination heating and one for nonlocal recombination heating.

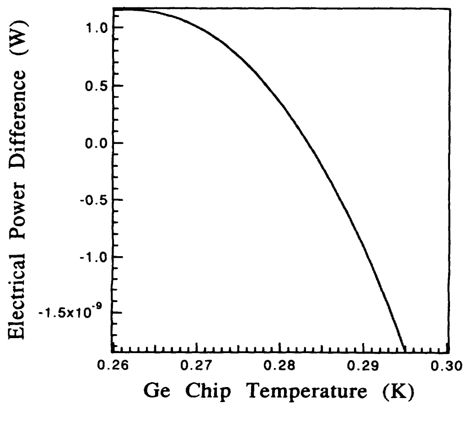

FIG. 4. The difference power ΔP between the electrical power vs temperature curve for $n=0$ and $n=5.9\times10^{13}/\text{cm}^3$.

The local term is given by

$$P_{\text{loc heat}}=0.5D_0F\,dN_{\text{bolo}}/dt\ , \tag{2}$$

where

$$dN_{\text{bolo}}/dt=A_{\text{bolo}}\bar{K}\lambda^2(T_0)\exp(2E_a/k_BT_b)n^2/2\ . \tag{3}$$

Here, dN_{bolo}/dt is the number of H↓ atoms recombining on the bolometer surface per second, and $A_{\text{bolo}}\approx3.0\ \text{cm}^2$ is the area of the bolometer, and $\bar{K}=(1-p)^2K_{aa}+(1-p^2)K_{ab}$. $K_{aa}=8.3\times10^{-8}\ \text{cm}^2/\text{s}$ and $K_{ab}=3.3\times10^{-8}\ \text{cm}^2/\text{s}$ are the second-order surface recombination rate coefficients.[10]

The nonlocal term is given by

$$P_{\text{nonloc heat}}=0.5D_0(1-F)\frac{A_{\text{bolo}}}{A_{\text{wall}}+A_{\text{bolo}}}\frac{dN_{\text{cell}}}{dt}\ , \tag{4}$$

where

$$dN_{\text{cell}}/dt=A_{\text{wall}}\bar{K}\lambda^2(T_0)\exp(2E_a/k_BT_0)n^2/2 + dN_{\text{bolo}}/dt\ . \tag{5}$$

Here, A_{wall} is the area of the cell. The nonlocal term is due to recombination occurring on all surfaces of the cell (including the bolometer itself) with a fraction $A_{\text{bolo}}/(A_{\text{wall}}+A_{\text{bolo}})$ of the nonlocally distributed heat entering the bolometer. We have assumed that all of the energy released during a recombination event which is not deposited locally is distributed uniformly throughout the area of the entire cell. This is reasonable since a very large number of wall collisions are required to relax a molecule to its ground state, and wall collisions should be the dominant relaxation mechanism at the low gas densities in our experiment.[4,16]

The power taken away from the bolometer via accommodation cooling by the H↓ gas is given by

$$P_{\text{cool}}=\alpha 2k_B(T_b-T_0)nv_H(T_0)A_{\text{bolo}}/4\ , \tag{6}$$

where $v_{\text{H}}(T)=(8k_BT/\pi m_{\text{H}})^{1/2}$ is the mean thermal velocity of a H atom in the gas. The literature value for α at 225 mK is approximately 0.11.[12,13]

In principle, the value of F can be determined by using Eq. (1) to analyze our data. However, such a determination requires precise knowledge of the effective accommodation coefficient which depends upon the geometry of the cell. Since our geometry differed from that in which α had been measured, we could not determine a reliable absolute value for F. Alternatively, it is possible to find an experimental regime where the cooling is unimportant, so that it is not necessary to know the value of the accommodation coefficient in order to determine an upper bound for F. This is how we analyzed the data.

The key to obtaining an upper bound on F is to examine the rate of decrease of ΔP with T_b, characterized by

$$\partial(\Delta P)=\Delta P(T_{b1})-\Delta P(T_{b2})>0,\quad T_{b2}>T_{b1}\ . \tag{7}$$

$P_{\text{nonloc heat}}$ is independent of T_b, except for the last term in Eq. (5), which is small by the ratio $A_{\text{bolo}}/A_{\text{wall}}$. Dropping the cooling term, Eq. (6), since experimentally we observe it to be small in the range where we evaluate $\partial(\Delta P)$, we find

$$\partial(\Delta P) > P_{\mathrm{loc\ heat}}(T_{b1}) - P_{\mathrm{loc\ heat}}(T_{b2}) = \frac{D_0}{4}\bar{K} A_{\mathrm{bolo}} F n^2 \lambda^2(T_0)[\exp(2E_a/k_B T_{b1}) - \exp(2E_a/k_B T_{b2})] \ . \quad (8)$$

Because the cooling term is positive, the effect of neglecting it is to make the upper bound on F even more conservative. The smallest value of the upper bound determined from Eq. (8) is found for the lower values of T_b and with small temperature differences. Using the data for $n = 5.9\times 10^{13}/\mathrm{cm}^3$ in Fig. 4, and taking $T_{b1} = 0.250$ K and $T_{b2} = 0.254$ K, we find $F \leq 0.040$, for $p \approx 0$. Other density data are consistent with this result. In an earlier analysis, before having experimentally determined the nuclear polarization p, Zhao *et al.*[4] used the value of $p = 0.5$ and found that $F \leq 0.10$.

The method just described neglects the composite nature of the bolometer and assumes that the bolometer film temperature remains equal to the Ge chip temperature at all levels of power input and all H↓ gas densities. This turns out to be a good assumption, as we have shown by a numerical simulation of the thermal response of each element of the bolometer.[18]

On physical grounds, we might expect that F is very small, since the initial recombination event places the H_2 molecule into a highly excited state only of order 100 K below the continuum (less than 0.2% of the total energy). Each possible trajectory for the H_2^* can be quantified with a probability p_i and local heating fraction F_i. For example, the H_2^* might dissolve into the liquid ^{4}He film with probability p_1, and such an event might give an effective local heating fraction, $F_1 \approx 1$, as an excited molecule will probably relax very quickly once inside the liquid where it interacts constantly with the ^{4}He atoms surrounding it. The H_2^* might take a completely different path, elastically scattering off of H atoms near the surface, and then finally leaving the vicinity of the position of recombination with probability p_2. In this case, F_2 would be less than about 0.002. The upper bound we have measured is for $F = \Sigma_i p_i F_i$.

The low upper bound for F that we have found means that it is not easy for the recombination heat to leave a high-density H↓ sample in a closed system. For such a system it would have been advantageous if it were found that recombination energy was deposited locally into the helium surface, where it can be easily removed. For example, if the sample dimensions are smaller than the mean free path for relaxation, the H_2 may possibly enter the liquid helium and deposit all of its energy there. The low upper bound for F is a propitious result if one is able to prepare a high-density H↓ sample in an open configuration or study the two-dimensional hydrogen gas on the helium surface. In this case, the H_2^* has the possibility of leaving the system altogether, taking as much as $(1-F) > 96\%$ of the total recombination energy away with it. Experiments designed to take advantage of the small value of F will suppress one of the principal obstacles to observing degenerate behavior in spin-polarized hydrogen.

In summary and comparison of our results, we have found $F < 0.04$ with negligible uncertainty for this upper bound. Vasilyev *et al.*[5] have used a different technique for the same density regime (two-body recombination on helium surfaces) and find values of F between 0 and 0.2 and give a value of 0.01 with an uncertainty of 0.11 for an average over several experiments. Matsubara *et al.*[4] give a value of 0.014 for F but the details of this experiment and the regime of measurement (two- or three-body recombination on helium surfaces) are not yet available.

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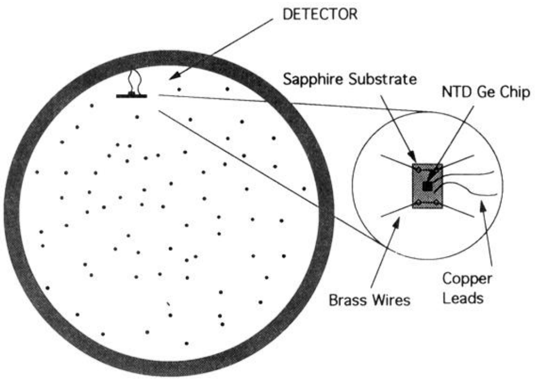

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