

# Formation of Nuclear-Polarized Phases of H Atoms Embedded in Solid H<sub>2</sub> Films

S. Sheludiakov,\* P. T. McColgan, D. M. Lee, and V. V. Khmelenko

*Institute for Quantum Science and Engineering, Department of Physics and Astronomy, Texas A&M University, College Station, Texas 77843, USA*

J. Järvinen, J. Ahokas, and S. Vasiliev

*Department of Physics and Astronomy, University of Turku, 20014 Turku, Finland*

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We report on an experimental observation of two phases of hydrogen atoms in solid H<sub>2</sub> films at temperatures of 0.1–0.8 K, characterized by a large enhancement of the nuclear spin polarization compared to that given by Boltzmann statistics ( $p = 0.15$  at  $T = 0.15$  K). The first phase with  $p = 0.35(5)$  is formed spontaneously during sample storage in a high magnetic field ( $B = 4.6$  T). The second phase with an even higher nuclear polarization,  $p = 0.75(7)$ , can be achieved at  $T \leq 0.55$  K by repeating sequences of dynamic nuclear polarization followed by a system relaxation. Upon warming through the range 0.55–0.65 K, the highly nuclear-polarized phase undergoes a phase transition to the spontaneously polarized phase which breaks down at  $T \simeq 0.8$  K, and the nuclear polarization gradually converges to the Boltzmann distribution. We discuss possible scenarios for explaining the nature of the observed phenomena.

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The system of hydrogen atoms embedded in an H<sub>2</sub> lattice represents a very special class of quantum solids. Even though the H<sub>2</sub> matrix is less quantum than solid <sup>3</sup>He and <sup>4</sup>He in terms of the de Boer parameter, for H atoms it allows the largest degree of delocalization. Considering impurities in quantum crystals, H in solid H<sub>2</sub> becomes the best candidate for studying delocalization and exchange phenomena between impurities as well as supersolidlike behavior [1,2]. A spectacular feature of H atoms in solid molecular hydrogen is a migration through the H<sub>2</sub> crystal by a repetition of the exchange tunneling reaction  $H + H_2 \rightarrow H_2 + H$  [3,4]. As an advantage over <sup>4</sup>He atoms, the accumulation of H atoms in the ground state may be sensed directly by the methods of magnetic resonance. Moreover, a temperature dependence of the nuclear spin polarization of H atoms should be different for the Bose and Boltzmann statistics [5], which may reveal a possible Bose-Einstein condensation (BEC) of H atoms in solid H<sub>2</sub>.

Ahokas *et al.* [6] observed a spectacularly large deviation of the nuclear spin polarization  $p = (n_a - n_b)/(n_a + n_b)$  of H atoms embedded in solid H<sub>2</sub> from the Boltzmann distribution, with  $n_a$  and  $n_b$  being the populations of the two lower hyperfine levels [Fig. 1(a)] determined from the H atom electron-spin resonance (ESR) line areas. They found  $p = 0.5$  at 0.15 K, which greatly exceeds  $p = 0.15$  as given by the Boltzmann statistics at this temperature. This observation was verified by subsequent experiments [7,8]. Analyzing these results, Hazzard and Mueller [5] considered the formation of a local BEC in the region of a high concentration of H atoms in solid H<sub>2</sub> as a possible explanation for an overpopulation of the ground  $|a\rangle$  state.

Nevertheless, no further studies of this phenomenon were carried out, and its origin remained unclear.

In the present work, we report on an ESR study of the temperature and time dependence of nuclear polarization of H atoms embedded in a solid H<sub>2</sub> matrix at temperatures 0.1–1.1 K in a high magnetic field. We observed the formation of two phases, characterized by a different deviation of the H atom nuclear polarization from the Boltzmann distribution. The first phase, with  $p = 0.35(5)$ , which we defined as the spontaneously nuclear-polarized phase (SNPP), is formed spontaneously in the course of sample storage in a high magnetic field (4.6 T). The second

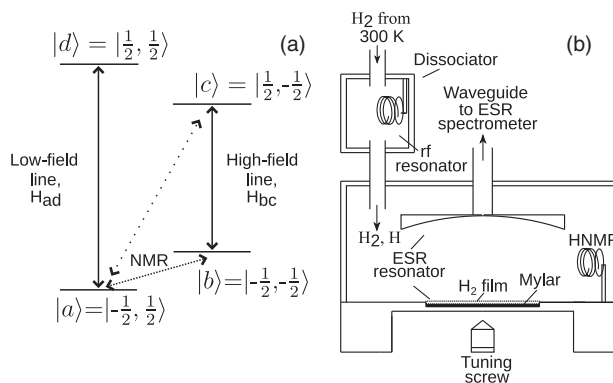


FIG. 1. (a) Energy level diagram for H atoms in a high magnetic field. The electronic and nuclear spin states are labeled as  $|m_s, m_I\rangle$ , respectively. The transitions  $H_{ad}$ ,  $H_{bc}$ , and  $H_{ab}$  are all allowed. The ESR  $H_{ca}$  forbidden transition is shown by the dotted line. (b) The sample cell schematic.

phase, which we defined as the highly nuclear-polarized phase (HNPP), is characterized by an exceptionally high nuclear polarization of H atoms,  $p = 0.75(7)$ . The formation of the HNPP is stimulated by dynamic nuclear polarization (DNP) via the Overhauser effect (OE) [9]. We observed that the HNPP is stable at temperatures up to  $T \simeq 0.55$  K, where the transition to the spontaneously nuclear-polarized phase takes place. The SNPP breaks down at  $T \simeq 0.8$  K, and the nuclear polarization approaches the Boltzmann distribution at higher temperatures.

The experimental setup is based on an Oxford 200 dilution refrigerator which accommodates a sample cell (SC) and a 128 GHz cryogenic ESR spectrometer [8,10]. The sample cell is thermally anchored to the mixing chamber of the dilution refrigerator, which makes it possible to stabilize the SC temperature in the range  $\simeq 0.09$ – $2$  K. The SC is placed at the center of a 4.6 T superconducting magnet and contains an open-design ESR Fabry-Perot resonator [Fig. 1(b)] and an auxiliary helical resonator (HNMR) tuned to the frequency of the NMR  $H_{ab}$  transition ( $f \simeq 910$  MHz) [Fig. 1(a)].

The sample deposition consisted of several consecutive steps (see [11], Sec. I). First, we admitted several millimoles of  $H_2$  gas into the sample cell kept at  $T \simeq 15$ – $20$  K, which led to accumulating  $H_2$  in the liquid state. After that, we slowly cooled the sample cell in order to solidify the hydrogen film with a thickness of  $\simeq 100$   $\mu\text{m}$ . Finally, we cooled the sample cell down to 0.5–0.8 K and started an rf discharge [14,15] using the HNMR coil [Fig. 1(b)]. A small amount of helium was condensed into the SC, sufficient to form a saturated superfluid film, which was necessary to run the discharge in helium vapor. The electrons created during the discharge have energies of the order of 100 eV and were able to dissociate  $H_2$  molecules in the film only in a rather thin surface layer,  $\simeq 100$  nm [16]. The rf discharge in the SC was run for 5–7 days, and the accumulation of H atoms in solid  $H_2$  was controlled by registration of their ESR spectra. The discharge was stopped when the H atom concentration achieved the value of  $10^{19}$   $\text{cm}^{-3}$  (see [11], Sec. II).

After stopping the rf discharge and cooling the sample cell to  $T = 0.15$  K, from ESR spectra we observed a clear deviation of the nuclear polarization ( $p = 0.25$ ) from the value corresponding to the Boltzmann occupation,  $p = 0.15$ , of the lower two hyperfine levels  $|a\rangle$  and  $|b\rangle$  [Fig. 1(a)]. During sample storage at  $T = 0.15$  K, the polarization spontaneously slowly increased with time and eventually saturated at  $p = 0.35$  after 5 days [green circles in Fig. 2(a)], achieving the SNPP. Similar measurements were also performed at  $T = 0.47$  K, which provided the same polarization behavior.

To enhance the process of nuclear polarization by transferring the  $|b\rangle$  atoms to the ground  $|a\rangle$  state, we used the OE DNP followed by polarization relaxation to a certain steady state. This DNP effect is based on saturation

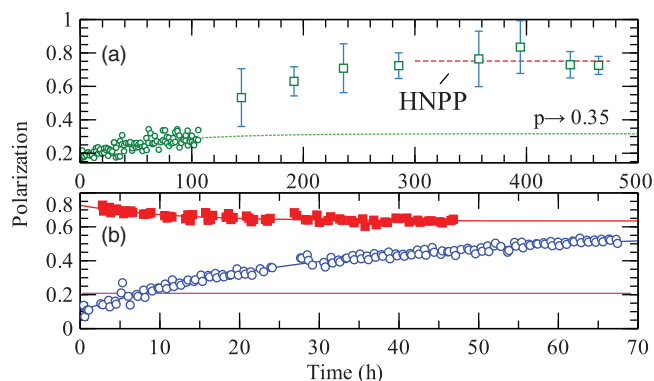


FIG. 2. (a) Buildup of the nuclear polarization of H atoms after stopping the rf discharge and storing the sample at  $T = 0.15$  K (green circles) followed by the application of SRSs (green squares). The polarization  $p = 0.75$ , corresponding to the HNPP, is designated by a dashed red line. The asymptotic polarization for the SNPP is  $p \simeq 0.35$ . (b) Relaxation of H atom nuclear polarization after applying the OE (red squares) and after saturation of the  $H_{ab}$  NMR transition (blue circles) measured at  $T = 0.1$  K. Polarization corresponding to Boltzmann occupation of the  $|a\rangle$  and  $|b\rangle$  levels at  $T = 0.1$  K ( $p = 0.21$ ) is shown by a solid magenta horizontal line.

of the allowed ESR  $H_{bc}$  transition with a subsequent cross relaxation through the forbidden  $H_{ac}$  transition [see Fig. 1(a)]. By using saturation-relaxation sequences (SRSs), we were able to gradually increase the polarization up to the value  $p \simeq 0.75$ , which eventually ceased to change with time. This steplike growth of polarization after applying SRSs at  $T = 0.15$  K is presented in Fig. 2(a) by open green squares. The HNPP was achieved after about 4.5 days of sample storage and application of the SRSs during the following 5.5 days. An ESR spectrum of H atoms in solid  $H_2$  at 0.1 K at a nuclear polarization of  $p = 0.75$  is presented in Fig. 3. The measurement of the polarization relaxation after OE saturation is shown by the upper trace in Fig. 2(b) [17]. The nuclear spin-lattice relaxation times of H atoms in the  $H_2$  matrix at temperatures below 1 K are extremely long,  $\sim 10^5$  s [18]. Thus, a measurement of the polarization recovery for each

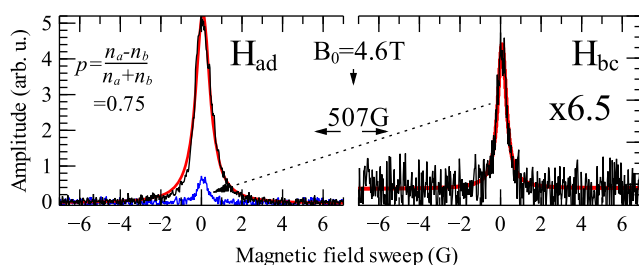


FIG. 3. ESR spectra of H atoms in the  $H_2$  matrix measured at  $T = 0.1$  K after reaching the HNPP. On the left-hand side, we show for comparison the signals obtained for  $H_{ad}$  and  $H_{bc}$  lines on the same scale. The hyperfine level populations  $n_a$  and  $n_b$  are proportional to the  $H_{ad}$  and  $H_{bc}$  ESR line areas.

temperature point was done for a time interval of 3 days. The asymptotic polarization  $p_0$  was obtained from an exponential fit:  $p(t) = p_0 + a \exp(-t/T_{1N})$ , where  $a$  is a free fitting parameter and  $T_{1N}$  is the nuclear spin-lattice relaxation time (see [11], Sec. III).

In order to verify that the polarization indeed relaxes to this steady state value after applying the OE saturation, we carried out an alternative measurement. We equalized the populations of  $|a\rangle$  and  $|b\rangle$  states by applying an rf excitation at the NMR  $H_{ab}$  transition frequency (910 MHz), and after that we recorded the process of polarization recovery [see the lower curve in Fig. 2(b)]. We found that the polarization approached nearly the same high value as those obtained by applying the SRSs [Fig. 2(b)].

We also studied the possibility of reaching the HNPP by applying the SRSs right after creating the sample and cooling it to  $T = 0.15$  K, *without waiting* for the SNPP to be established [Fig. 4(a)]. The characteristic time for the formation of HNPP in experiments presented in Figs. 2(a) and 4(a) appeared to be nearly the same even though the numbers of the SRSs applied were different. Growth of the asymptotic value of polarization after each measurement was also accompanied by an increase of  $T_{1N}$ , which rose by a factor of 3–5 after 1 week of sample storage [Fig. 4(b)]. These observations allow us to conclude that both factors, the sample storage time and the use of SRSs, are essential for the HNPP formation. We also found that the characteristic time of the polarization buildup for reaching the HNPP does not depend on temperature in the range 0.09–0.55 K, while the HNPP could not be achieved at temperatures higher than  $T = 0.55$  K.

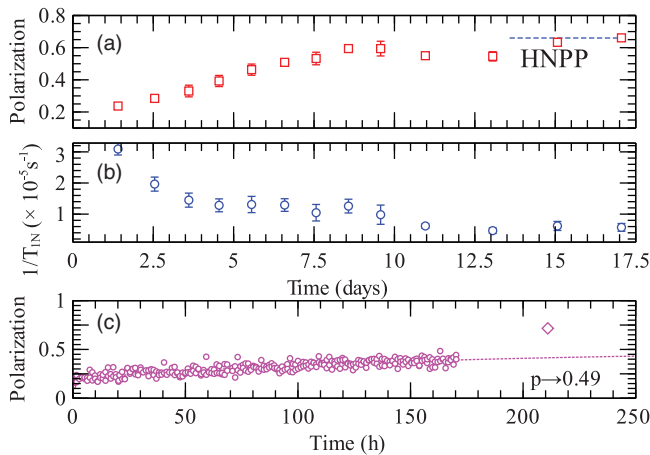


FIG. 4. (a) Buildup of the nuclear spin polarization of H atoms after the application of the SRSs while storing the sample at 0.15 K. (b) Evolution of  $T_{1N}$  for the measurement (a). The polarization corresponding to the HNPP is designated by a dashed blue line. (c) Polarization recovery after keeping a magnetic field at zero for 24 h. The HNPP of  $p \simeq 0.75$  had been reached prior to discharging the magnet. The HNPP can be rebuilt by a single SRS (magenta diamond) after a waiting period of 9 days. The asymptotic polarization for (c) is  $p \simeq 0.49$ .

We also made a special effort to study the influence of the magnetic field on the formation and stability of the HNPP of H atoms. Initially, we retrieved several spectra of both ESR transitions after finishing the OE saturation and then *completely discharged* the magnet and kept the magnetic field at zero for 24 h. Then the magnet was charged back to  $B = 4.6$  T. After storage in a zero magnetic field, the nuclear polarization decreased to  $p = 0.07$ , which corresponds to the Boltzmann occupation of the  $|a\rangle$  and  $|b\rangle$  levels in a zero magnetic field. We did not observe any decrease of the H atom concentration during manipulation of the magnetic field in this experiment. After the magnetic field,  $B = 4.6$  T, was restored, we measured the polarization recovery, which approached  $p \simeq 0.49$ . The characteristic time of this process  $\simeq 7$  days [Fig. 4(c)] appeared to be greater than that for the process of polarization recovery shown in the lower curve in Fig. 2(b), about 3 days. On the other hand, the polarization recovers to  $p \simeq 0.49$ , which clearly exceeds the value  $p \simeq 0.35$  corresponding to the SNPP [Fig. 2(a)], indicating a possible memory effect. We also verified that the HNPP could be easily achieved again by a *single* SRS as depicted by an open magenta diamond in Fig. 4(c).

After the HNPP was formed, we performed a measurement of the asymptotic polarization as a function of the temperature (Fig. 5). Each point was acquired in the same manner, by using the SRSs. The points were measured upwards starting from  $T = 0.09$  K. We observed that the steady state polarization  $p \simeq 0.75$  does not change significantly at temperatures of 0.09–0.55 K. However, it decreased to the value of  $\simeq 0.35$  rather abruptly between 0.55 and 0.65 K. The values of the asymptotic nuclear polarization ( $p \simeq 0.35$ ) formed a well-defined plateau in the range of 0.65–0.8 K, where they remained independent of the temperature. These values are very close to the nuclear polarization obtained spontaneously (SNPP), by storing

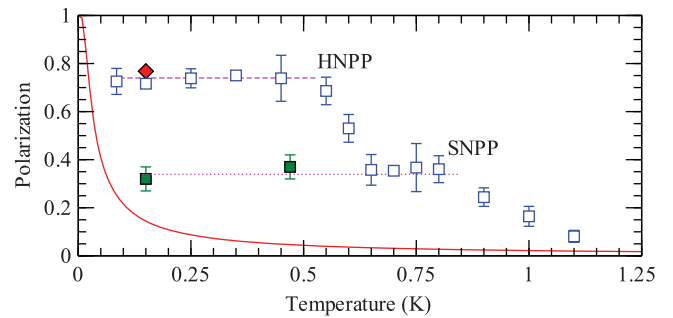


FIG. 5. The steady state polarization of H atoms in solid  $H_2$  after reaching the HNPP as a function of the temperature (blue squares). The red line depicts the values of polarization based on the Boltzmann statistics. The red diamond was measured right after acquiring points at  $T = 0.65$ –0.8 K, cooling back to 0.15 K, and using the SRS. The green squares ( $p \simeq 0.35$ ) show the SNPP values measured at  $T = 0.15$  K [Fig. 2(a)] and  $T = 0.47$  K. The dashed and dotted magenta lines are guides for the eye.



fresh samples at  $T = 0.15$  and  $0.47$  K ( $p \simeq 0.35$ ). The asymptotic polarization decreased further at higher temperatures ( $T > 0.8$  K) and approached the Boltzmann statistics at  $T > 1$  K. We also verified that the HNPP can be restored by cooling atoms being in SNPP at  $T \sim 0.55$ – $0.8$  K to  $T < 0.55$  K and applying a single SRS as shown in Fig. 5 by a red diamond.

We now discuss the formation and possible nature of the spontaneous and highly nuclear-polarized phases. The SNPP forms spontaneously during the course of sample storage in a high magnetic field at  $T < 0.8$  K for 5–7 days. The HNPP requires the use of SRSs after establishing the SNPP. Once the HNPP is formed, its high steady state polarization recovered in only 2–3 days after the sample was intentionally depolarized by saturating the  $H_{ab}$  NMR transition [see the lower trace in Fig. 2(b)]. The nuclear spin ordering was easier to restore after a disruption of the magnetic field if the HNPP had been established prior to that, demonstrating a memory effect [Fig. 4(c)].

We anticipate that the buildup of nuclear polarization of H atoms in solid  $H_2$  crystals is a consequence of a long-range interaction between hydrogen atoms mediated by the quantum properties of the  $H_2$  matrix. Solid para- $H_2$  is composed of the lightest nonmagnetic molecules with a strong delocalization. The H atoms are delocalized to an even larger extent due to their tunneling motion through the  $H_2$  matrix. Previously, we observed [17] that the strong interaction effects are responsible for anomalous DNP of the pairs of H atoms in H-D<sub>2</sub>-H complexes residing at the distance of two lattice constants in solid D<sub>2</sub>:H<sub>2</sub> mixtures. Assuming that similar long-range interactions also exist in the quantum crystals of H in  $H_2$ , we may suggest several possible mechanisms leading to the buildup of nuclear polarization observed in this work.

A BEC-like behavior in the regions of a large local concentration of H atoms was considered as a possible explanation of the observed overpopulation of the  $|a\rangle$  state [5]. This would require much larger densities,  $\sim 5 \times 10^{20}$  cm<sup>-3</sup> at  $T = 0.2$  K [6], and would have led to additional dipolar ESR line broadening not observed in our experiment. However, the observed memory effect for nuclear polarization in the ensembles of H atoms might result from a spatial arrangement of H atoms in the  $H_2$  matrix. Although BEC of H atoms in solid  $H_2$  is unlikely to take place at our temperatures and H atom concentrations, the statistical attraction of the identical bosonic atoms may lead to their accumulation in the lowest energy state in the case of a sufficiently strong interaction between them.

As a second possibility, we suggest that the buildup of a high nuclear polarization of H atoms may be due to the formation of a cooperative spin-aligned state in regions of the  $H_2$  matrix where the H atom interaction becomes sufficiently strong. The magnetic ordering in solids may be established if the interaction energy exceeds the thermal energy. A delocalization of H atoms in the  $H_2$  matrix

leading to the strong interaction may favor the emergence of such a spin-aligned state.

After creating the sample, it takes time for the H atoms to migrate to the regions where the crystalline fields are most homogeneous and the interaction effects are the strongest. This may explain a slow buildup of the SNPP. By performing the OE, we decrease the effective temperature of the nuclear spin system,  $T_{\text{eff}}$ , which may facilitate further nuclear ordering in the regions where the interaction energy exceeds  $k_B T_{\text{eff}}$ , thus forming the HNPP.

Finally, we put forward a mechanism for the increased nuclear polarization of H atoms in the  $H_2$  matrix due to the formation of triplet  $H_2$  molecules. The electron spins of H atoms for high magnetic fields and low temperatures of our experiment are completely polarized, and the H atoms interact via the  ${}^3\Sigma_u^+$  potential [19]. The  ${}^3\Sigma_u^+$  triplet potential has a very shallow minimum ( $\epsilon_0 = 6.46$  K [19]) with no bound state in the gas phase [20]. The potential minimum becomes deeper in a high magnetic field ( $\epsilon_0 \simeq 13$  K at  $B = 4.6$  T [19,21]), which under the influence of the surrounding  $H_2$  lattice may become deep enough to allow the formation of weakly bound  $H_2$  molecules in the  ${}^3\Sigma_u^+$  state. The molecular wave function symmetry in the case of the odd electronic  ${}^3\Sigma_u^+$  wave function requires the alignment of H atom nuclear spins for the lowest energy state of such an  $H_2$  molecule [22,23] (see [11], Sec. IV). We suggest that the  $H_a$ - $H_a$  pair would correspond to the ground state for a  ${}^3\Sigma_u^+$   $H_2$  molecule, while the formation of the  $H_b$ - $H_b$  pairs would be energetically less favorable. When an  $H_b$  atom approaches an  $H_a$  atom, it should reverse the nuclear spin due to their interaction to form the bound  ${}^3\Sigma_u^+$  state. It is also possible that a nuclear spin can be reversed due to the wave function symmetry restrictions even without the formation of the bound  ${}^3\Sigma_u^+$  state if the mutual H-H atom interaction energy is sufficient to trigger the spin flip.

The weakly bound  ${}^3\Sigma_u^+$  state can be easily destroyed by zero-point fluctuations or phonons, thus providing two  $H_a$  atoms for tunneling through the  $H_2$  matrix. They can then meet other  $H_b$  atoms to form  ${}^3\Sigma_u^+$  molecules. This way, more and more  $H_a$  atoms move through the  $H_2$  matrix, leading to the buildup of nuclear polarization in the system. The nuclear spin orientation of the  $H_a$  atoms may persist due to the very long  $T_{1N}$ , thus leading to the buildup of nuclear polarization. The above process might correspond to the formation of SNPP. The SNPP breakdown at  $T > 0.8$  K may be triggered by a gradual enhancement of nuclear-spin lattice relaxation (see [11], Sec. III) or a breakdown of the  ${}^3\Sigma_u^+$  bound state if the binding energy of this state is of the order of 0.8 K.

In the case of the HNPP, the  $|a\rangle$ -state occupation and, thus, formation of the  $H_a$ - $H_a$  pairs are additionally stimulated by the OE DNP. It is possible that the HNPP is associated with a spatial aggregation of  $H_2$  molecules in the  ${}^3\Sigma_u^+$  state or, alternatively, a different arrangement of increased population of individual  $H_a$  atoms. This spatial

arrangement may break up for  $T > 0.55$  K. The proposed model is tentative, and a rigorous quantitative theoretical analysis is required for understanding the observed values of nuclear polarization for the SNPP ( $p = 0.35$ ) and HNPP ( $p = 0.75$ ).

In summary, in this work we observed the formation of two robust phases characterized by the H atom nuclear polarization substantially exceeding that corresponding to the Boltzmann distribution. The first phase,  $p \simeq 0.35$ , is formed spontaneously during sample storage in a high magnetic field. The formation of the second, highly nuclear-polarized phase ( $p \simeq 0.75$ ) is stimulated by use of the OE. Further experimental and theoretical work will be required for a full understanding of the nature and origin of these two phases. To ultimately observe a possible onset of H atom BEC in solid H<sub>2</sub>, lower temperatures and higher concentrations than those achieved in this work would be required.

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\*seshel@physics.tamu.edu

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