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Observation of a Phase Transition in Adsorbed Hydrogen at 101 Kelvin

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Abstract : While adsorbent surfaces such as graphite are known to increase the melting temperature of solid H2, this effect is normally rather small, increasing to 20 Kelvin (K) relative to 14 K in the bulk. An as-yet unidentified phase transition has been observed in a system of H2 adsorbed in a porous, locally graphitic, Saran carbon with sub-nanometer sized pores at temperatures (74-101 K) and pressures (> 76 bar) well above the critical point of bulk H2 using hydrogen adsorption and neutron scattering experiments. Adsorption data shows a discontinuous pressure jump in the kinetics at 76 bar after nearly an hour of equilibration time, which is identified as an exothermic phase transition. This discontinuity is observed in the 87 K isotherm, but not the 77 K isotherm. At higher pressures, the measured isotherms show greater excess adsorption at 87 K than 77 K. Inelastic neutron scattering measurements also show a striking phase transition, with the amount of high angle scattering (corresponding to large momentum transfer/ large effective mass) increasing by up to a factor of 5 in the novel phase. During the course of the neutron scattering experiment, three of these reversible spectral phase transitions were observed to occur in response to only changes in sample temperature. The novel phase was observed by neutron scattering only at high H2 pressure (123 bar and 187 bar) and temperatures between 74-101 K in the sample of interest, but not at low pressure (30 bar), or in a control activated carbon at 186 bar of H2 pressure. Based on several of the more unusual observations, such as the slow equilibration and the presence of both an upper and lower temperature bound, a reasonable hypothesis is that this phase forms only in the presence of a high concentration of ortho-H2 (nuclear spin S=1). The increase in adsorption with temperature, temperatures which cross the lower temperature bound observed by neutron scattering, indicates that this novel phase is denser. Structural characterization data on the adsorbent shows that it may support a commensurate solid phase denser than those known to exist on graphite at much lower temperatures. Whatever this phase is eventually proven to be, these results show that surfaces can have a more striking effect on hydrogen phases than previously thought.

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