Experimental report

26/06/2024

Proposal: 7-05-550 **Council:** 10/2022

Title: Proving the existence of high temperature, intermolecular nuclear magnetic ordering in nanoconfined H2

Research area: Physics

This proposal is a new proposal

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Samples: activated carbon

Instrument	Requested days	Allocated days	From	To
WASP	5	5	13/09/2023	18/09/2023

Abstract:

Magnetic nuclear spin correlations between nuclei are very rare in materials, typically only occurring at very low temperatures (nK-mK). These coherent quantum phases of matter are of particular interest as quantum sensors or even as neutron polarizer materials. We have collected preliminary data indicating long-range nuclear spin coupling may occur for nanoconfined hydrogen molecules (H2) at temperatures 3-4 orders of magnitude higher than the current record. Using temperature-dependent XYZ spin polarised neutron scattering on WASP, these experiments will decouple nuclear spin-incoherent, nuclear coherent and magnetic scattering of nanoconfined H2 to experimentally determine if previously observed scattering deviations are indeed caused by intermolecular (H2-H2) nuclear-spin correlations or alternatively originate from nuclear short-range order between H2 and C or electronic magnetic interactions with the carbon network. If nuclear spin correlations between nanoconfined H2 molecules are proven, this work will demonstrate how coherent quantum phases of matter can be generated at temperatures far higher than the current state of the art using a simple methodology ¿ nanoconfinement.

EXP 7-05-550 : Intermolecular Nuclear Magnetic Ordering in Nanoconfined H_2 at "high" temperatures.

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Abstract

Motivated by previous anomalous neutron scattering and magnetisation data we perform temperature-dependent XYZ spin polarised neutron scattering on WASP (diffraction mode) below the Bragg limit on H_2 and D_2 confined in nanoporous carbon. The results reveal a nuclear coherent scattering factor contributes to the intrinsic scattering from nanoporous carbon, likely reflecting carbon nanoplatelet sizes. Adsorption of H_2 and D_2 dampens and enhances this scattering factor, respectively. This may be a feature of contrasting caused by differing coherent scattering cross sections filling the pores/voids in the carbon framework. Nuclear spin incoherent scattering is seen to have a general increase upon adsorption of H2 and D2. A very broad plateau is observed in the NSI H2 at 2 K, however, due to increased measurement uncertainties observed for this particular temperature, it is unsure if the observation is real. A time dependence is also observed in the NSI scattering for H2, indicating a link to ortho-to-para conversion within the pore network. No changes were observed in the magnetic scattering. The results remain inconclusive as to proving the source of the previously observed Q-dependence in the elastic line of nanoconfined H2, further experimentation on a higher-resolution instrument is required to resolve this quandary.

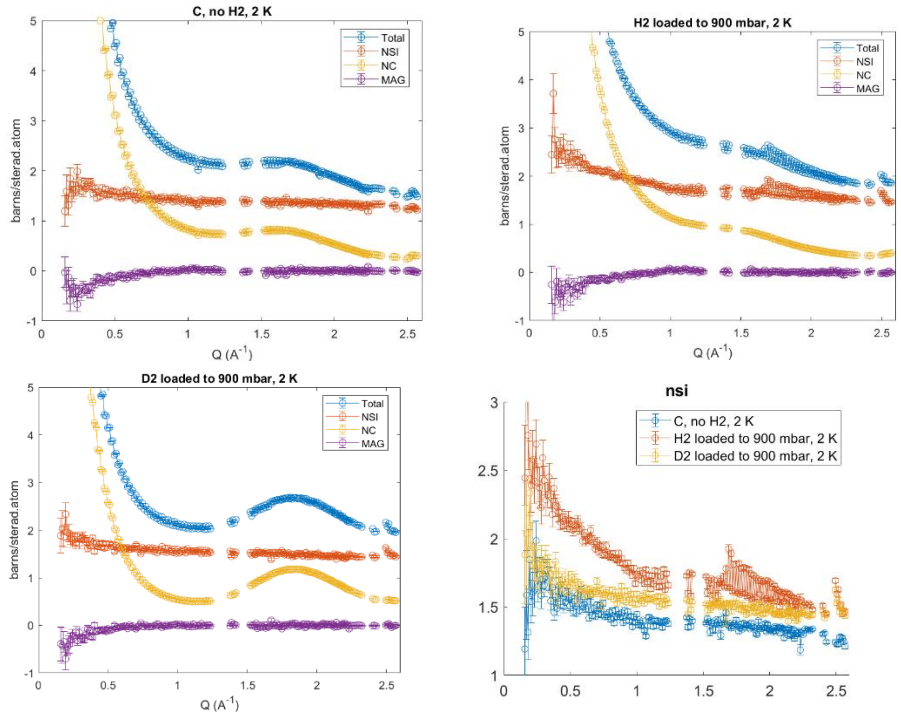

Figure 4. Comparison of XYZ neutron scattering from carbon under vacuum, Carbon+H_2 and Carbon+D_2 all at 2 K.

Discussion

In comparison (**Figure 4**), there are clear changes to the neutron scattering upon adsorption and nanoconfinement of the adsorptive gases, H_2 and D_2, this is most pronounced at the lowest temperatures when the confined species solidifies in the pores, however, at the lowest temperature 2 K, the largest experimental error in data is observed for H_2, chiefly from the NSI scattering, why this occurred is unclear. 10 K has been used for direct comparison means in this discussion (see **Figure 5**) although some of the 2 K measurements are considered.

In separating the spin-polarised XYZ neutron scattering into its components, it is evident that there are no changes to the magnetic scattering across the samples, this is most clearly seen by the direct comparison in **Figure 5**, ruling out an electronic magnetic component from the carbon causing Q-dependent features in the neutron scattering in the timescale of this experiment and in the previous INS experiment (**Figure 1**).

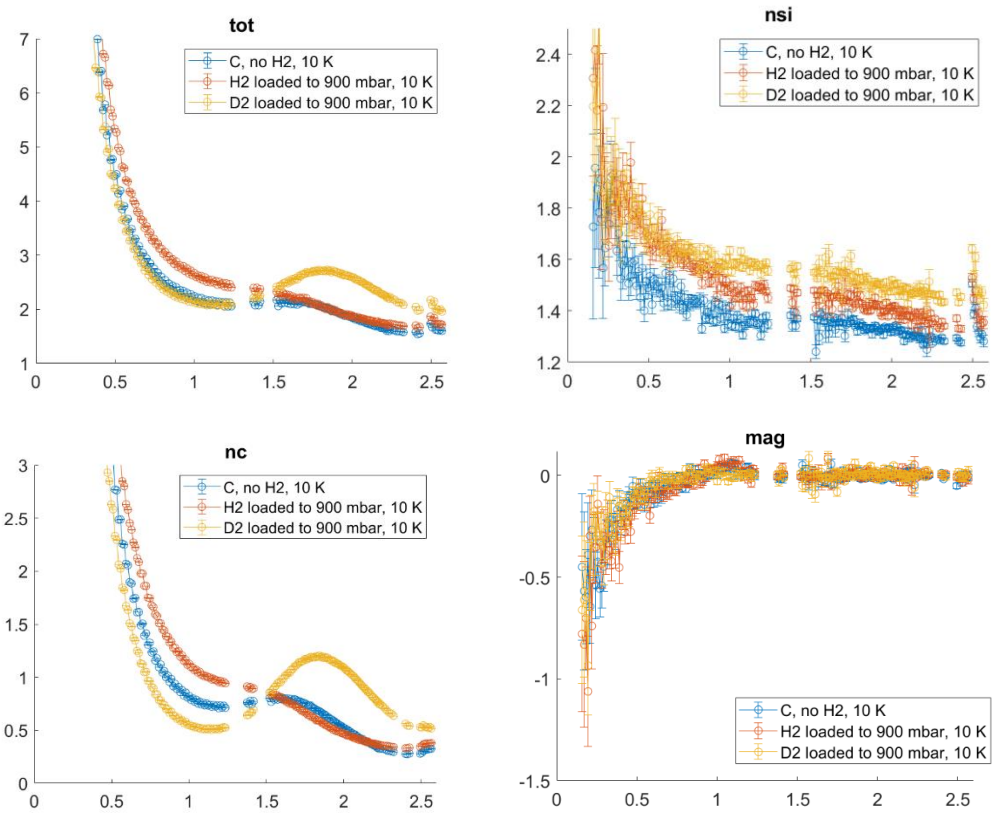

Figure 5. Direct Comparison of all scattering between carbon, H_2 & D_2 experiments at 10 K.

There are clear features and changes to the nuclear coherent contributions to the total scattering upon adsorption and nanoconfinement of the gases, and these make the majority of the changes to the total scattering. The key observed coherent scattering feature is inherent to the carbon itself, however, the addition of the H_2 appears to flatten this feature. This is seen in the coherent difference spectra displayed in **Figure 6.** Across the Q-values that the feature is observed in carbon, a clear loss is observed in the difference spectra (H_2-carbon). Below these values (<1.5 A^{-1}) an approximate exponential increase in coherent scattering is observed as Q decreases. Likewise, there is a small increase in the region 2.25-2.6 A^{-1}, similar to the Q value of the feature previously observed in **Figure 1**. This suggests that the q-dependence may be caused by nuclear short-range order of the H2 molecules with the carbon, however, the very small amplitude is not significant enough to analytically attribute nuclear short-range order to the cause of the previous observation. Longer experimental time, investigation of higher Q-values, or the use of a higher resolution instrument such as D-007 would help confirm this.

In direct comparison, Nuclear spin incoherent scattering increases upon adsorption of H_2 species, but marginal amount. Inspection of the difference spectra at 10 K (**Figure 6**) fails to resolve any features in the increase. Inspection of the 2 K difference spectra (**Figure 7**) does resolve a very broad plateau in the data at ~1.4 A^{-1}**,** however**,** the increased error in the rebinned data around the detector crossover points (~1.6 A^{-1}) makes the interpretation of this feature questionable and would require re-experimentation on WASP or a higher resolution diffraction instrument such as D-007.

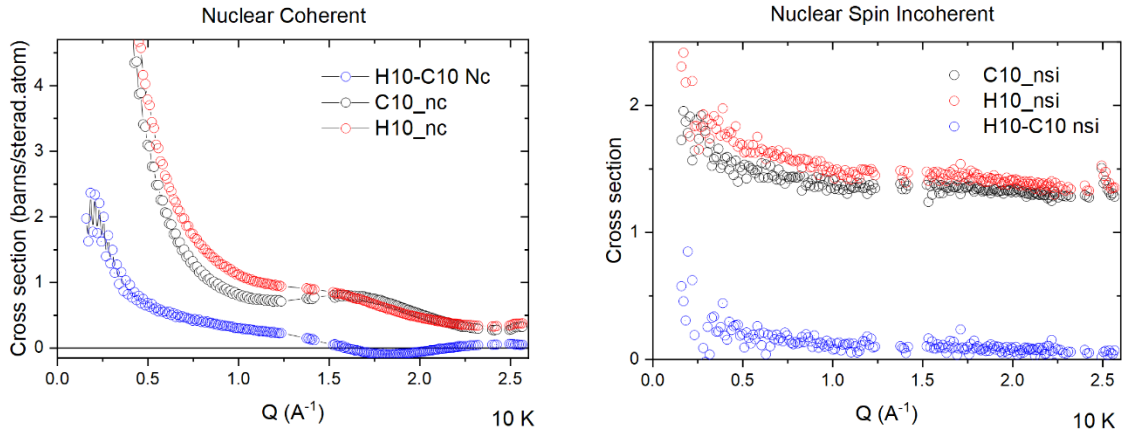

Figure 6. Nuclear coherent and nuclear spin incoherent difference spectra of carbon and H2 experiments at 10 K.

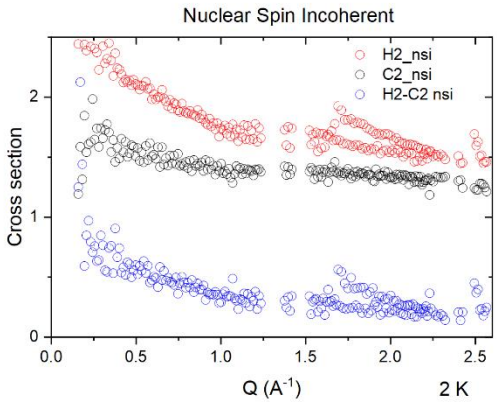

Figure 7. Nuclear spin incoherent difference spectra of carbon and H_2 experiments at 2K.

Conclusions

From the experiments conducted, the source of the previous Q-dependence in elastic scattering for nanoconfined H_2 remains elusive. The experiments are inconclusive, in that they do not exhibit the same q-dependence as previously observed, with the most qualitative indications of both Coherent and Nuclear spin incoherent scattering causing the feature. It is suggested that the experiment be repeated on a higher resolution instrument, such as D-007, for a longer collection time and with a sample with lower C-H bonds in it, this will require a much weaker/shorter acid-washing step, in preparation of the samples.