

THE DYNAMICS OF HYDROGEN IN SOLID C₆₀

Carbon is the most studied element in the entire periodic table. For decades it was believed that pure carbon existed in two forms, diamond and graphite. Thus it was quite surprising when a third form of pure carbon, called fullerenes, was discovered in the mid 1980's. The most well-known of this new-class of molecular materials is the one consisting of 60 carbon atoms which takes the shape of an atomic-scale soccer ball. These beautifully symmetric molecules are commonly called “buckyballs”.

In the solid state, C₆₀ molecules form a structure with large interstitial spaces between the molecules (Fig. 1). These spaces are easily large enough to accommodate a wide variety of atomic and molecular species which can significantly influence the properties of the resulting compound. Most notable are the alkali doped C₆₀ compounds which display superconductivity at reasonably high temperatures. However, it has also been shown that various gases including hydrogen, nitrogen, carbon monoxide and oxygen can be absorbed into the spaces between C₆₀ molecules. This suggests that solid C₆₀ might be useful as a medium for the safe storage of hydrogen or as a molecular sieve

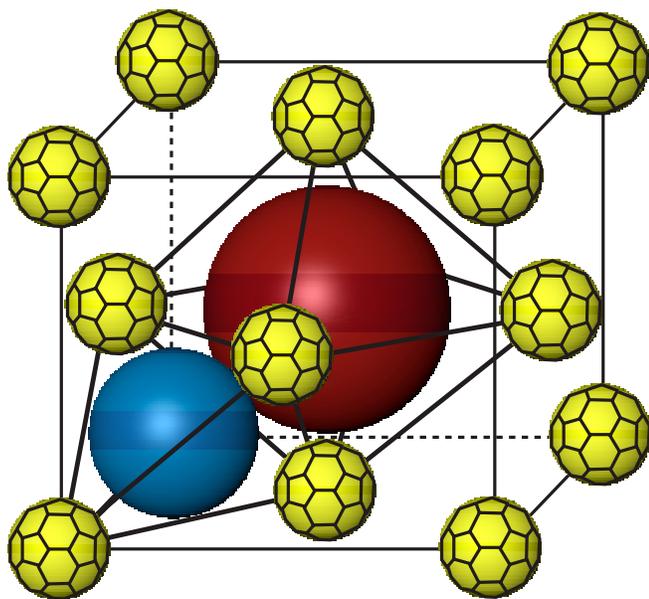


FIGURE 1. The yellow balls indicate the positions of the C₆₀ molecules while the red ball resides in the larger octahedral site and the blue ball sits on one of the tetrahedral sites. The hydrogen molecules must go into one of these spaces between the “buckyballs”.

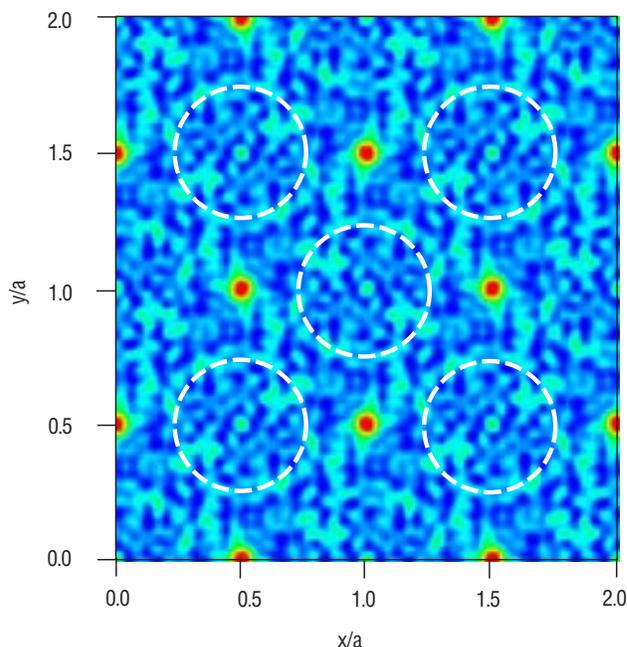


FIGURE 2. A Fourier difference map of the [100] crystallographic plane which shows the locations of the C₆₀ molecules using dashed white lines and the location of the deuterium molecules by the red and yellow contours at the octahedral position.

to separate these gases. In spite of this, relatively little effort has been made to understand the interactions which govern these potentially useful properties, namely the interaction of the trapped species with the C₆₀ host. Hydrogen in C₆₀ is also of fundamental interest because it is a nearly perfect example of a simple quantum object (hydrogen) trapped in a classical matrix (C₆₀). The location and quantity of the trapped hydrogen is determined using neutron diffraction. The data show a 40% occupancy of the octahedral site (as illustrated in Fig. 2) in contrast to a zero occupancy of the tetrahedral sites.

Neutron spectroscopy is a direct way of probing the bonding in materials and it is a particularly powerful tool with which to study the dynamics of hydrogen because of the large incoherent scattering cross-section of hydrogen. Inelastic scattering spectra obtained using the Fermi Chopper Spectrometer (Fig. 3) show well-defined peaks, which can be attributed to the quantized rotational levels of the hydrogen and deuterium molecules. These peaks are only slightly shifted from the positions expected for the free rotations of these molecules. The simplicity of this behavior allows us to probe the effects of relatively weak

interactions with the host lattice, which lead to the small departure from the free rotor case.

To explore this more quantitatively we have performed model calculations using a simple Born-Meyer type of potential. The rotational potentials felt by hydrogen and deuterium molecules within this model are shown in Fig. 4. The panel on the left shows the calculated potential for the case of complete orientational disorder of the C_{60} molecules while that on the right shows the potential for ordered C_{60} . The difference in energy between the red and blue regions is only ~ 1 meV, in agreement with the experimental observation that the rotations are nearly free. The diagrams below the calculated potentials show the change in the quantum energy levels as the strength of the orientational potential is varied. For the disordered case, no change is seen over this limited range of potential, whereas for the ordered C_{60} case, the level at ~ 14.7 meV splits into 2 states, as is observed experimentally. Thus the degree of splitting is extremely sensitive to the symmetry and the strength of the interactions.

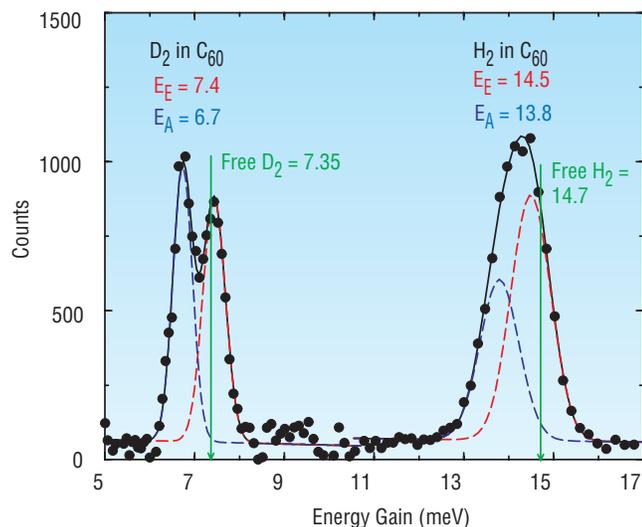


FIGURE 3. Neutron spectra showing the rotational transitions for hydrogen and deuterium absorbed in solid C_{60} . Note that the transition is a doublet in both cases with a splitting of 0.7 meV. The center of the doublet is slightly shifted from the value expected for the free molecules, which is indicated by the green arrow.

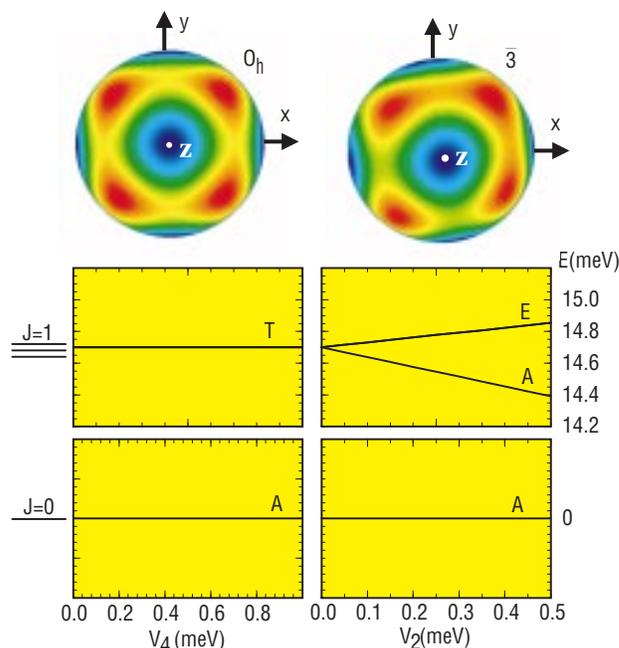


FIGURE 4. The top diagrams show contour plots of the orientational potential felt by a hydrogen molecule when C_{60} molecules are orientationally disordered (left) and orientationally ordered (right). The total variation in the potential (from red to blue) is about 1 meV indicating that the rotation is only weakly hindered. The bottom diagrams show the quantum rotational levels as a function of the orientational potential when C_{60} molecules are orientationally disordered (left) and orientationally ordered (right).

Neutron scattering combined with self-consistent phonon calculations demonstrates that the symmetry of the interstitial site is $\bar{3}$. Furthermore, the magnitude of the splitting (0.7 meV) is comparable to the size of the rotational barrier arising from the intermolecular interactions between the hydrogen and the C_{60} molecules. Additional studies are underway which will probe how these interactions are changed by the application of pressure and the co-intercalation of charged species into the C_{60} host.