EXPERIMENTAL STUDY OF COLD HYDROGEN ATOMS IN INTERACTION WITH GRAPHITE

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Introduction

From fundamental studies as in astrophysics to novel applications like thermonuclear fusion and energy storage, the motivation to investigate hydrogen-graphite interaction phenomena arises from various sources. Hydrogen adsorption on graphite was considered as impossible until density functional theory calculations established its feasibility. Some work [1] even demonstrated that an H atom approaching a clean basal plane of graphite has to overcome an activation barrier of 0.2 eV to adsorb on the surface. A hydrogen atom (with enough energy) weakly bonds on top of a carbon atom to form a monomer. This monomer induces a small deformation of the graphene plane. Then dimers formation is greatly facilitated because of the drop-off of the activation barrier in the vicinity of the adsorbed atom [2]. Finally clusters which are more strongly bonded can be formed and stabilize the final surface.

Experimental

The experiments were performed in an experimental set-up consisting of several interconnected ultra-high vacuum chambers with a base pressure below 2.10^{-10} torr. Two different sources were used to produce atomic hydrogen beams. The first one (Omicron EFM-H) provides hydrogen (deuterium) atoms by thermal dissociation of hydrogen (deuterium) molecules in a fine tungsten capillary heated by electronic bombardment. Released atoms are assumed to be in thermal equilibrium leading to a typical kinetic energy close to 0.250 eV ("hot" atoms). The second source is an inductively coupled plasma source tuned as atomic source (charged particles are neutralized during their diffusion in a Pyrex tube). As incoming gas is at room temperature, the mean energy of hydrogen (deuterium) atoms reaching the sample is assumed to be close to 0.025 eV ("cold" atoms).

Results and Discussion

To experimentally demonstrate the existence of the activation barrier, irradiation of the graphite sample by both atomic hydrogen sources were performed. High-Resolution Electron Energy Loss Spectroscopy (HREELS) was used to probe the vibrational modes of C-H and C-D bonds on the surface following each irradiation. The adsorption of "hot" H (and D) atoms has been extensively investigated and is commonly known to give rise to several structures in the HREEL spectrum [3]. For instance, C-H stretching vibrations exhibit features at 295, 330 and 345 meV in the presence of monomers, dimers and clusters respectively. On a clean HOPG surface the main loss feature appears at 200 meV. It corresponds to the in-plane longitudinal optical phonon mode of graphite. The surface exposed to "cold" H atoms does not exhibit any new feature (in particular no C-H related vibrations). Therefore, the similarities of both spectra readily demonstrate the absence of any adsorbed species. As a consequence, this result is an experimental evidence that the "cold" H atoms do not adsorb on HOPG surface as predicted by the adsorption model.

Subsequently, abstraction process was investigated. A clean graphite sample (figure 1.a) is exposed to "hot" D atoms. As seen in figure 1.b, HREEL spectrum presents features at 243 and 257 meV related to C-D stretching vibrations. Irradiation of this deuterated graphite sample to "cold" H atoms (figure 1.c) leads to the abstraction of the adsorbed specie. Also, no adsorption of cold H atoms is observed during the process despite the predicted drop–off of the activation barrier.

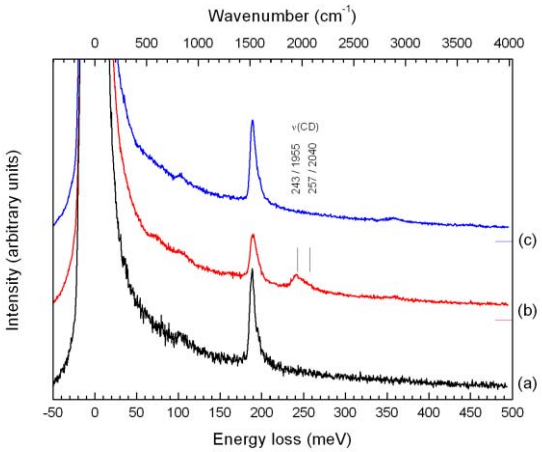

Fig. 1 HREEL spectra of (a) clean HOPG, (b) HOPG exposed "hot" H atoms and (c) subsequently exposed to "cold" H atoms.

This result suggests that the recombination of a cold atom with an adsorbed atom is a mechanism more efficient than the adsorption of a cold atom without energy barrier.

Conclusions

The present study focused on the interaction of "cold" H atoms (~0.025 eV) generated by a plasma source with a HOPG surface at room temperature. We demonstrate that H (or D) atoms do not have sufficient energy to overcome the predicted adsorption barrier of 0.2 eV therefore preventing its adsorption. Moreover, when exposing a hydrogenated HOPG surface to "cold" D atoms, a complete H abstraction is observed with no further adsorption of D atoms. Recombination of adsorbed H with "cold" D from the gas phase is favored to HD dimers and clusters formation.

References

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