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Excitations in the Soliton Phases of D_2 on Graphite

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The transverse phonon at the zone boundary has been studied in the commensurate(C)-phase of D_2 adsorbed on graphite with inelastic neutron scattering. This excitation looses intensity and shows a measurable lifetime in the higher density striped domain wall phase adjacent to the C-phase. In addition an excitation at lower energy appears. These modes resist also above 7K in the reentrant liquid phase showing that this phase is in reality a solid phase with domain walls which possess however only short range order as can be concluded from elastic neutron scattering studies.

The phase diagram of monolayers of deuterium (D_2) on graphite has been studied by elastic neutron scattering[1] and recently by heat capacity measurements[2]. D_2 on graphite presents a ($\sqrt{3}$ x $\sqrt{3}$) 30° commensurate phase (C) at temperatures lower than 18K and, when the coverage (ρ) is increased it changes to an incommensurate phase (IC) up to the monolayer completion (see Fig.1). The C-IC transition is not an abrupt one, but goes through several other intermediate phases, depending on the path taken in the coverage- temperature diagram. The phase diagrams of quantum gases on graphite show all the commensurate phase and are very similar if the temperature is scaled. This common aspect arises because the zero point motion gives a repulsive part to the adatom-adatom interaction so that the monolayer expands till the lock-in distance of the $\sqrt{3}$ structure is reached. The study of the intermediate phases is of particular interest firstly to test the influence of the different isotopes of the quantum gases and secondly to probe theories of the C-IC transition[3-7] which involve different domain wall structures. Studies of the structure of the adsorbed D_2 layer using neutron diffraction[8] and LEED[9] revealed a striped domain wall array in the α-phase adjacent to the $\sqrt{3}$ phase. Other neutron diffraction pattern showing satellites could be measured in the intermediate phase near to the IC-phase. The β-phase looks like a reentrant liquid[7] but the structure factor[8] and the specific heat peak[2], even if being small and broad between the β and fluid phase, indicate a separate phase with a longer range order than in a liquid.

It is the aim of the inelastic neutron scattering studies to learn more about the dynamics of the adsorbed layer and some results have already been published[1,10]. In the present report we concentrate on the C-phase and the adjacent α and β-phases. We have studied the dynamics of D_2 adsorbed on papyex using the triple axis spectrometer IN3[11] in a Be filter after the sample configuration, a fixed final neutron energy (E_F =1.22THz) and a horizontally focusing analyser. The sample cell was filled with papyex[12] sheets oriented parallel to the neutron scattering plane and was positioned on the spectrometer in a focusing geometry to improve the signal. At all coverages the elastic peak was also measured to insure the determination of the coverage. The papyex cell was measured without any D_2 inside and this was taken as the background to be subtracted from each scan. The points of the phase diagram where inelastic scans have been performed are shown in fig.1.

The peak corresponding to the rotational transition for the D_2 molecule[13], $E_{J=0}-E_{J=1}$ = 1.808 THz, is present in all the scans as a small peak and is coverage and temperature independent (figs. 2-3).

The spectra taken at T = 4K for different coverages are shown in fig. 2. Around ρ = 1 (point of completion of the C phase) at ρ = 0.94 and ρ = 1.04 there is a well defined peak for E = 0.91 THz which corresponds to the zone boundary transverse phonon of the C adsorbate. Since the papyex sheets are a 2-dimensional powder, an orientationally averaged spectra is measured which favors strongly the signal from the zone boundary[14]. The intensity of this peak increases with ρ until ρ = 1 and then starts to diminish because the further adsorbed molecules cause local defects.

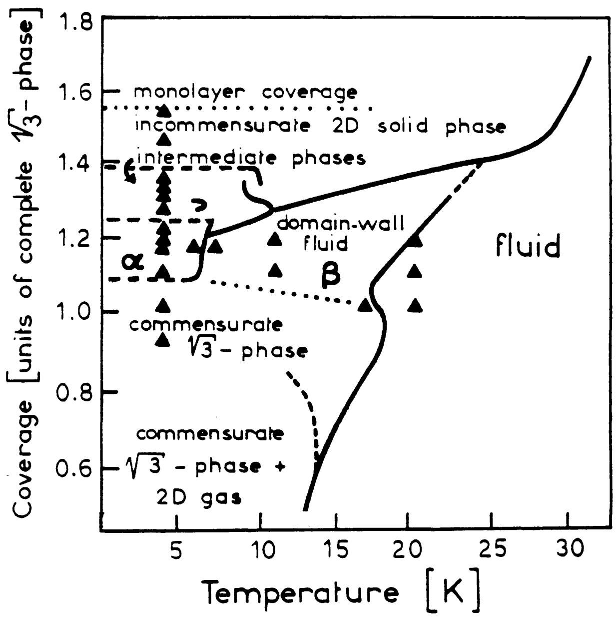

FIG.1 Phase diagram of D_2 adsorbed on graphite determined by heat capacity measurements (taken from reference 2). The black triangles indicate where inelastic neutron spectra have been measured.

The α-phase extends from ρ = 1.10 to 1.25 (fig.1). In this region one finds two peaks (fig.2). The new peak appears at a lower energy, E = 0.6 THz. The C peak begins to broaden due to lifetime effects, since the striped domain wall phase at this densities is formed by a non periodic alternance of superheavy walls and C structure[8,15]. It shifts also slightly to higher energies indicating a stiffening of the adsorbate lattice. The sum of the integrated intensity for both peaks remains nearly constant in this range of coverages (163 for ρ = 1.14 and 156 for ρ = 1.20). This and the fact that the energy of the lower mode doesn't change suggests that it is an excitation of a single domain wall. If it were due to the interaction of domain walls it should shift its energy when the distance between them changes. As the number of domain walls is increased the intensity of the low energy peak increases and that of the C-peak decreases. Unfortunately there exists still no theoretical model for the excitations in a soliton structure in 2D. A very rude estimate of a unidimensional model in the direction normal to the walls gives an effective mass of two times that of the C structure for each domain wall. It is quite suggestive that the relation of the energies of this two peaks is nearly $\sqrt{2}$ which would be the case for two harmonic oscillators with the mentioned mass ratio and unchanged force constants.

On the other hand the structure of the β-phase is still not completely clarified. It is puzzling that a fluid should exist at temperatures as low as 6K for this system. The scans taken at ρ = 1.14 (fig.3) show that the transverse

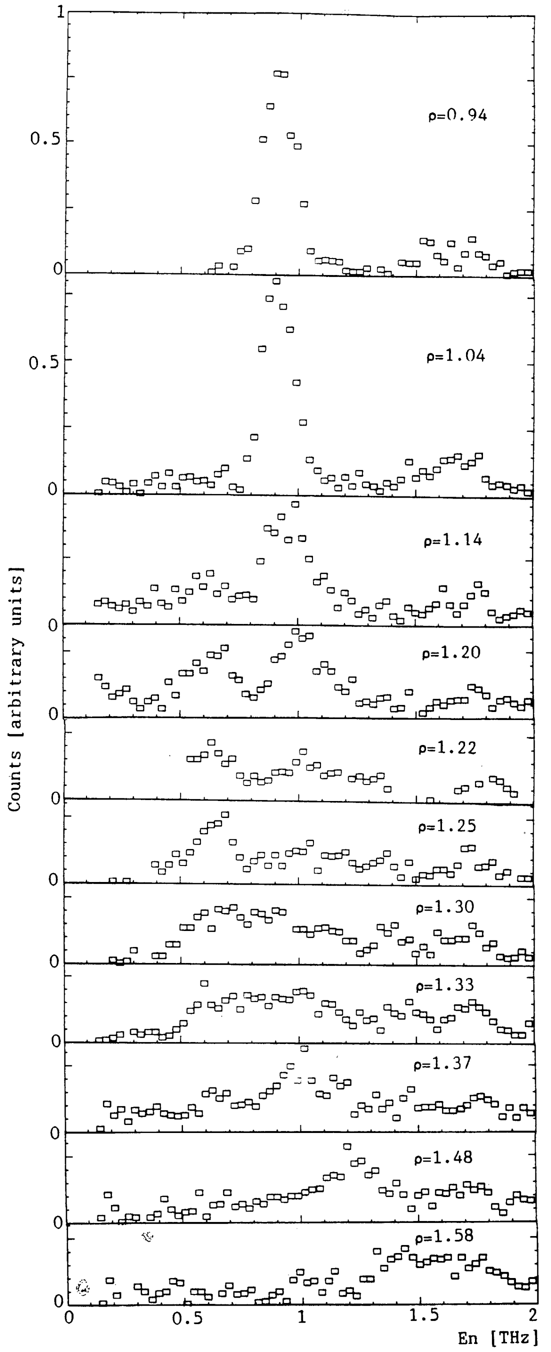

FIG.2 Measured neutron groups for the transverse phonons in D_2 on graphite at T = 4K. ρ = 1 stands for the complete commensurate monolayer. (Q = 1.7 $Å^{-1}$)

C peak still can be easily recognized at temperatures as high as 11K. The last spectra at 20K shows that only at this temperature this mode disappears. To be able to support this transverse mode this "fluid" must show a longer range correlation than what is assumed for a normal fluid. No broadening of the peak can be seen between 6 and 11K.

For higher coverages ($\rho > 1.3$) there exists only a very broad feature indicating the existence of a different structure than the striped domain wall. This is confirmed also by the elastic neutron diffraction experiments[8]. It is possible that the interaction between walls (e.g. intersection energy) is changing[3b]. Two excitations reappear at ρ=1.37 as before, but possibly arising from a different domain wall structure.

The excitation increases strongly its energy for still higher density ($\rho > 1.37$) showing that the lattice continues to get stiffer.

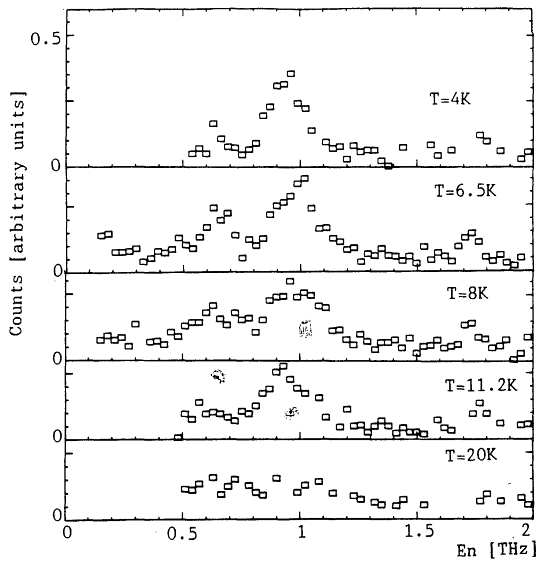

FIG.3 Measured neutron groups for D_2 on graphite for different temperatures at ρ = 1.14.(Q =1.7 $Å^{-1}$)

For the first time it has been shown that in the α-phase an excitation of a single domain wall coexists with the mode which corresponds to the C-structure. In this phase the energy of this excitation doesn't depend on the number of walls and the total integrated intensity remains constant. The domain wall fluid (β-phase) is capable of sustaining a transverse phonon up to high temperatures implying that it has a longer correlation that it was supposed before. It is suggested that it is in reality not a fluid at all, but a solid in a more disordered state.

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