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Classification

Physics Abstracts

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Absence of continuous growth for bcc and hcp solid ^3He on Grafoil (*)

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Résumé. — Les spectres de diffraction des neutrons pris dans la région bcc et hcp du diagramme de phase de l' ^3He ne montrent pas de solidification continue de l' ^3He solide sur le substrat de Grafoil. Ce résultat est confirmé par des isopycnals mesurées à proximité de la courbe de fusion entre 1,2 K et 3,8 K.

Abstract. — Neutron diffraction patterns taken in the bcc and hcp region of the ^3He phase diagram give no evidence for a continuous nucleation of solid ^3He on a Grafoil substrate. This result is supported by isopycnals measured near the bulk melting curve between 1.2 K and 3.8 K.

Recently, the nucleation of surface solid from the liquid phase of ^3He and ^4He on a hexagonal substrate (Grafoil) and a cubic substrate (MgO) has been studied. Continuous nucleation was established for the system ^4He -Grafoil from measuring isopycnals [1, 2], taking neutron diffraction patterns [1] and by attempts to over-pressurize liquid ^4He near the phase transition curve [3], and for ^3He on MgO by recording isopycnals [4]. Thus, following the classification of adsorption of Dash [5] and Peierls [6], these two systems have been found to be of Class I (uniform deposits at all thicknesses). On the other hand, thermodynamic [4, 7] and neutron scattering investigations [7] of ^4He on the cubic structure MgO and thermodynamic measurements of ^3He on Grafoil [8] in the temperature range from 30 mK to 320 mK show no evidence for continuous nucleation of a surface solid, classifying these systems as Class II (bulk nucleation after a thin uniform deposit).

In this letter, we present neutron diffraction and thermodynamic results on the last system (^3He -Grafoil) in the bcc and hcp region of the phase diagram. The neutron scattering measurements were carried out on the DIB multidetector diffractometer at the high-flux reactor in Grenoble (ILL). We used a wavelength of $2.522 \pm 0.002 \text{ \AA}$. The sample consisted of 120 Grafoil sheets (Grade GTA, thickness 0.38 mm) with a total weight of 4.18 g. The sheets had a size of $4 \text{ cm} \times 0.2 \text{ cm}$ and were orientated parallel to the scattering plane. Assuming a specific surface of $23 \text{ m}^2/\text{g}$, one obtains a total surface of the sample of

about 100 m^2 . Because of the large neutron absorption cross-section of ^3He , the diffraction patterns were taken in reflection geometry using a rectangular Al-cell constructed for pressures up to 200 bar. The wall thickness of 2 cm prevents deformation of the cell under pressure and ensures a tight fit of the Grafoil sheets to the cell walls, guaranteeing that even at high pressures there is practically no « dead » volume in front of the substrate. This is proved by the (002) Bragg-reflection of the substrate, which is visible at all investigated points up to a pressure of 137.5 bar. Thus, we can be sure that there is a high ratio of surface solid to bulk solid and that the neutrons interact with the solid layers on the Grafoil substrate as well as with the bulk solid in the Grafoil pores. Diffraction patterns were taken at several points in the $p - T$ plane which are indicated in figure 1 (solid circles).

The thermodynamic measurements of the pressure p as a function of the temperature T in the closed sample cell were recorded at constant number of atoms (isopycnal). The temperature was determined with a calibrated carbon resistor to an accuracy of 5 mK and the pressure with a capacitive strain gauge [9] with a resolution of $5 \times 10^{-3} \text{ bar}$.

Figure 1 shows the isopycnals of the system ^3He -Grafoil (solid lines ; I-V) compared with the bulk ones (dashed lines ; A-C). One of the isopycnals in the bcc region (II) is shown with a more extended scale in figure 2. In agreement with the results of Landau and Eckstein [8] in another temperature range ($30 \text{ mK} < T < 320 \text{ mK}$), no rounding of the isopycnal in the vicinity of the melting curve, which

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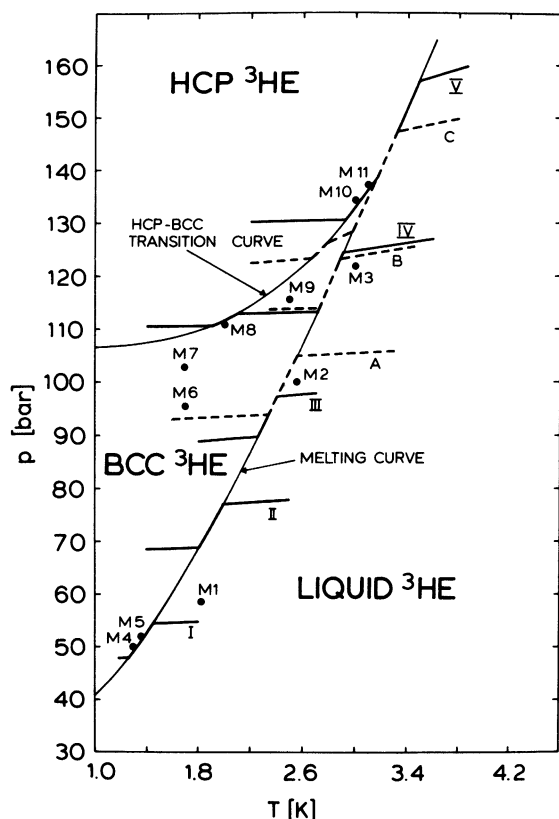


Fig. 1. — Phase diagram of ^3He with isopycnals for ^3He on Grafoil (solid lines I-V) and bulk ^3He (dashed lines A-C) at various number densities. The solid circles indicate those $p - T$ values where neutron diffraction measurements were performed.

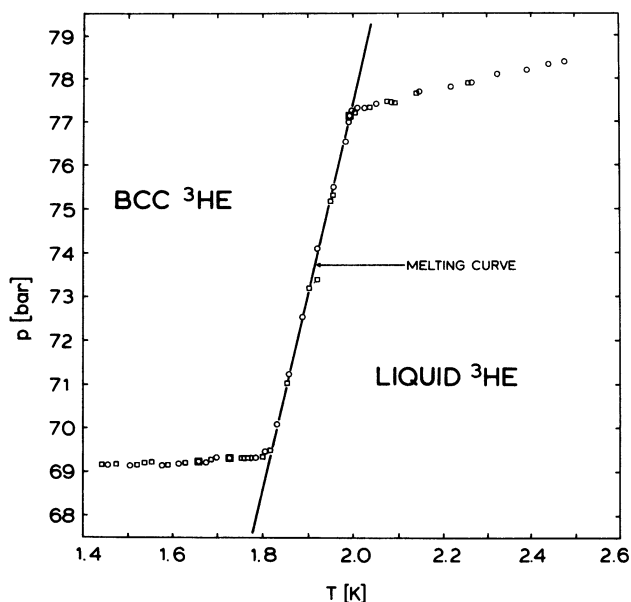


Fig. 2. — Isopycnal of ^3He on Grafoil in the bcc region of the phase diagram. The squares ($\square$) were measured at decreasing temperature and the open circles ($\circ$) at increasing temperature.

would indicate continuous nucleation, could be detected. The isopycnal has exactly the same shape as the bulk one (Fig. 1, isopycnal A).

The absence of nucleation in the bcc region is further supported by neutron scattering patterns. In contrast to our results for the system ^4He -Grafoil [1], no solid ^3He could be detected in the liquid phase near the melting curve (see Fig. 1 at points M1, M2 and M3). One of the diffraction patterns in the bcc region (M5) is shown in figure 3 (measurement M1 was subtracted since it can be regarded as a more suitable background as the scattering from the pure sample cell without ^3He). The peak in figure 3 is identified as the (110) Bragg reflection of the bulk bcc phase and, within the counting statistics, no Bragg peaks of solid hcp ^3He could be observed which are expected if continuous growth on graphite takes place [1]. Diffraction patterns taken at the points M4, M6, M7, M8 and M9 show the same result. The line widths of the Bragg peaks are entirely given by the instrumental resolution which would have allowed observation of broadening of the peak corresponding to finite crystallite sizes smaller than 600 Å. In the case of continuous nucleation, the crystallite size would have been influenced by the coherence length of the first adsorbed layer ($L \sim 150$ Å) [14].

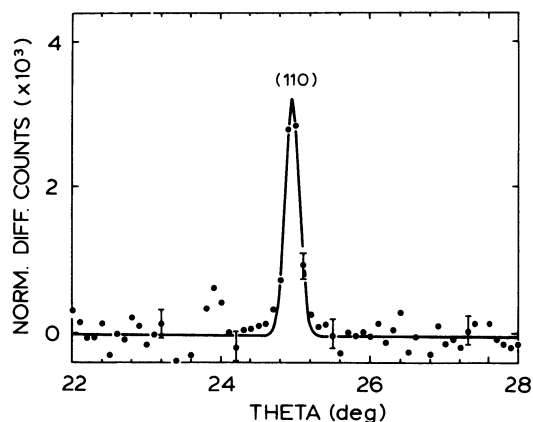


Fig. 3. — Diffraction pattern at M5 ($T = 1.36$ K, $p = 52$ bar).

Since ^3He has an extremely high neutron absorption cross-section of 3 200 barn/nucleus [10], the structure of bulk solid ^3He has not been investigated before. The lattice constants, calculated from our neutron measurements for all $p - T$ points and the molar volumina, are listed in table I. These values are almost identical with those of bulk solid ^3He , thus the properties of solid ^3He are not influenced by the presence of Grafoil surfaces. They are in very good agreement with data for bulk ^3He of Schuch and Mills [11] determined by X-ray diffraction and with thermodynamic results of Grilly and Mills [12].

For the investigation of the nucleation of ^3He on Grafoil in the hcp region, an isopycnal was measured in that region (Fig. 4). This isopycnal and the corresponding one in bulk (Fig. 1, isopycnal C) show the same behaviour. There is no rounding to be seen at the melting curve between liquid and hcp ^3He

Table I. — *Lattice parameters and molar volumina of bulk ³He.*

Invest. point	<i>T</i> [K]	<i>p</i> [bar]	Structure	Lattice constant (Å)	Molar volume cm ³ /Mol
M4	1.30	50.04	bcc	<i>a</i> ₀ = 4.225 (± 0.003)	22.71 (± 0.05)
M5	1.36	52.0	bcc	4.227	22.74
M6	1.69	95.5	bcc	4.101	20.76
M7	1.69	103.0	bcc	4.068	20.27
M8	2.00	110.5	bcc	4.046	19.94
M9	2.5	115.8	bcc	4.041	19.87
M10	3.0	134.2	hcp	<i>a</i> ₀ = 3.538 (± 0.003)	18.81 (± 0.04)
				<i>c</i> ₀ = 5.763 (± 0.003)	
M11	3.1	137.5	hcp	<i>a</i> ₀ = 3.537 (± 0.003)	18.80 (± 0.04)
				<i>c</i> ₀ = 5.762 (± 0.003)	

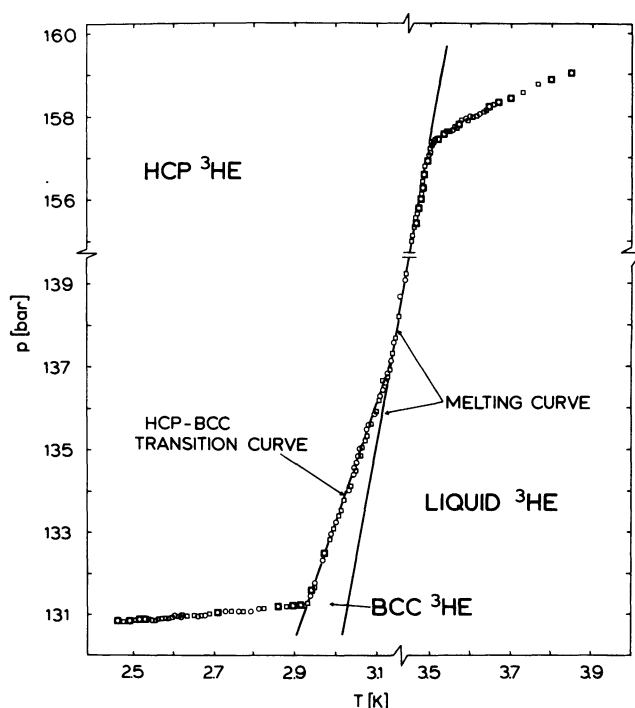


Fig. 4. — Isopycnal of ³He on Grafoil in the hcp region of the phase diagram. The squares (□) were measured at decreasing temperatures and the solid circles (○) at increasing temperature.

nor at the phase boundary between the bcc and the hcp solid. If the deviations of isopycnals from the bulk ones (Δp) were of the same order of magnitude as, for example, in the system ⁴He-Grafoil

$$(\Delta p = 0.3 \text{ bar}) \quad [1, 2]$$

or ³He — MgO ($\Delta p = 0.12 \text{ bar}$) [4], they would have been easily detected in the present experiment with an accuracy of δp of 0.005 bar. We conclude, consequently, that there is no evidence for continuous growth of a uniform surface solid near a Grafoil substrate in the hcp region also.

This conclusion is confirmed by two neutron diffraction patterns taken in the hcp region (M10, M11,

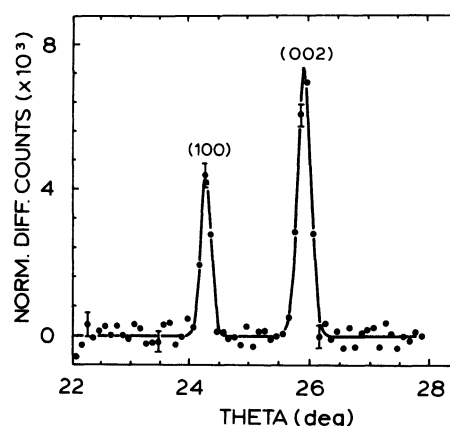


Fig. 5. — Diffraction pattern at M11 (*T* = 3.1 K, *p* = 137.5 bar).

Fig. 1) near the hcp-bcc transition curve. Figure 5 shows one of the spectra at *T* = 3.1 K and

$$p = 137.5 \text{ bar (M11)}$$

(measurement M3 was subtracted as background). The two reflections belong to the bulk hcp phase ((100), (002)). The half-width is again determined only by the instrumental resolution and therefore the same conclusion is drawn as for the bcc peak. In a hcp ³He polycrystal, one expects, in principle, three Bragg peaks ((100), (002), (101)) in the range covered by the diffraction experiment. Obviously, the (101) peak (expected at $\theta = 27.3^\circ$) is suppressed, presumably because of a highly preferred orientation of the ³He hcp solid. This preferred orientation cannot be understood by an epitaxial growth of hcp ³He on Grafoil since in that case the (002) peak should be absent, in contrast to the experimental result. This strengthens our thermodynamic result that ³He does not wet Grafoil in the hcp region.

Summarizing, our experimental data indicate the absence of continuous growth of any surface solid phase of ³He near a Grafoil substrate, both in the bcc and the hcp region of the phase diagram. This is supported by recent neutron diffraction investigations [15]

which show that no solid second layer of ^3He is formed on a UCAR ZYX graphite substrate if ^3He is adsorbed from the vapour. Thus, it seems to be clear that, even under an external pressure from the liquid phase of ^3He , there is no continuous formation of solid layers of ^3He on Grafoil. These results contradict the conclusions of Landau and Eckstein [4, 8] that continuous nucleation takes place when the symmetry of the monolayer matches the symmetry of the adjacent bulk solid lattice. Following this argument, the missing symmetry between the triangular monolayer [13, 14] and the bcc ^3He solid can prevent nucleation as

observed in our experiment. However, in the hcp region where the triangular monolayer matches the bulk hcp lattice and wetting should occur, no evidence for continuous growth can be found by thermodynamic measurements or by neutron scattering. This indicates that the symmetry between the monolayer and the bulk solid lattice cannot be the only factor which determines whether nucleation of surface solid on a substrate takes place or not.

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