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VISUAL OBSERVATIONS OF THE SOLIDIFICATION OF HELIUM ISOTOPE SOLUTIONS

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Investigations were made of the solidification of helium isotope solutions and of pure He^4 in a glass ampoule, and a marked difference was found between these processes. The boundary between the liquid and solid phases was a finely dispersed mixture of small crystals and liquid. The ease of obtaining a solid phase of uniform composition could be explained by the special features of the crystallization of solutions.

THE results of a number of experiments [1-3] on solutions of He^3 in He^4 force one to the conclusion that the process of crystallization of helium isotope solutions should have a mechanism differing considerably from the well-known [4] simple solidification of pure He^4. Our experiments made it possible to confirm this conclusion directly by visual observations of the process of solidification of a solution with a He^3 concentration equal to 25.6%.

In these experiments, we used a glass ampoule, 45 mm long and 8 mm in diameter, joined by a Kovar junction to a capillary (1.3 mm in diameter) used to supply the He^3—He^4 mixture.

The experiments were carried out as follows. The ampoule was filled with liquid helium at the required pressure and its temperature was reduced by pumping the vapor above the helium in a Dewar flask. The pressure at the beginning of crystallization was measured with a manometer. Then, the capillary leading to the ampoule was stopped up with a solid plug of the helium under investigation so that further crystallization of the latter took place under constant volume conditions. The pressure in the ampoule was no longer measured but it was found from the phase diagram of the investigated solution, which had been studied earlier [2,1] and is shown schematically in Fig. 1.

The initial pressure in the ampoule ranged from 98 to 35 atm, which made it possible to investigate the following types of solidification of a solution (Fig. 1):

1) The whole process of crystallization of the liquid produced a hexagonal close-packed (hcp) phase (dashed curve 1).

2) Hcp crystals were precipitated first but at some temperature in the two-phase region, where the solid phase was partly formed, the hcp was transformed into the bcc (body-centered cubic) structure and further crystallization produced bcc crystals (dashed curve 2).

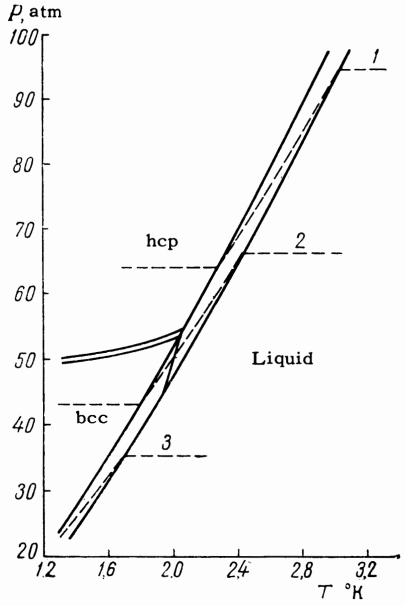

FIG. 1. Schematic phase diagram of a solution of He^3 in He^4, containing 25.6% He^3. The dashed curves represent the three types of crystallization of a solution in an ampoule under constant volume conditions.

3) Bcc crystals were produced from the beginning. In this case, it was not possible to crystallize the solution at the lowest temperature (1.3°K) of which our apparatus was capable (dashed curve 3).

In all these cases, the crystallization of the solution proceeded in the same way. At the moment the crystallization began, a light deposit of the solid phase appeared on the ampoule walls in the form of frosting. On cooling, the frosting became denser. Cooling by 0.1—0.15 deg made the mixture in the ampoule completely turbid and the turbidity persisted over a fairly wide range of temperatures (≈ 0.4 deg K). Maintenance of the solution in the two-phase region, at a constant temperature within the stated range, produced no changes in two hours. At 0.1—0.15 deg K above the temperature corresponding to the completion of solidification, the solution started to transmit light and at the end of the process it became transparent. Further cooling produced no visible changes in the solid solution. No further changes were observed at the moment of the hcp—bcc polymorphic transition (type 2) in an ampoule filled with a two-phase mixture. The crystalline phases with the hcp and bcc structures differed somewhat in their appearance: an examination, at the boundary between light and shadow, of an ampoule filled completely with the solid phase having the hcp structure revealed that it was optically somewhat nonuniform but transparent; the solid phase having the bcc structure was completely transparent.

When an ampoule was heated after the end of solidification, clearly visible cracks appeared in the transparent solid solution, their number increased, then the solution rapidly became turbid, and further heating precipitated opaque flakes. The same type of crystallization of a solution was observed also at a constant temperature when pressure was increased. The rate of increase of pressure ranged from 0.2 to 1 atm/min. The characteristic stages of the solidification of a solution are illustrated in Fig. 2.

The following description of the process of crystallization of solutions can be deduced from the observations. Many crystallization centers are produced right at the beginning and their number continues to increase. Small crystals are precipitated in the solution to form a loose aggregate, attached to the ampoule walls, which does not represent a continuous solid but has gaps filled with the liquid. Because of the strong scattering of light on the many boundaries between small crystals and the liquid, the solution becomes turbid and its transparency decreases as the number of small crystals increases. Beginning at some stage, the amount of the solid phase is so large that the further solidification of the liquid between crystals reduces the number of the liquid-crystal boundaries, which are replaced by boundaries between crystals themselves: the crystal again becomes transparent.

Our experiments showed that the nature of the crystallization of pure He^4 under the same conditions was basically different from the crystallization of solutions: the solid He^4 phase was formed by the growth of a perfectly transparent continuous layer from the walls of an ampoule. Even a sudden change in pressure (or temperature) did not disturb the growth of the crystals. The precipitation of solid He^4 in the form of hail or snow, and other features reported by Shal'nikov,[4] were not observed by us.

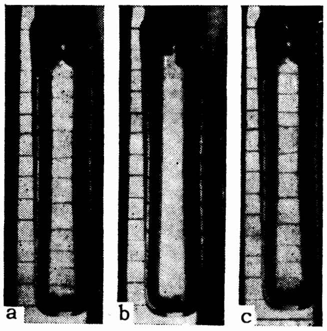

FIG. 2. Appearance of an ampoule containing a solution of He^3 in He^4 at various stages of crystallization: a) ampoule filled with liquid solution; b) ampoule containing a two-phase mixture of small crystals and the liquid; c) ampoule filled with a solid solution. The lines drawn on paper placed behind the helium Dewar are invisible when viewed through the turbid two-phase mixture.

Thus, the most important characteristic feature of the process of crystallization of He^3—He^4 solutions is the mixture of very small crystals permeated by the liquid solution, in the form of wet snow, which is observed in the region of coexistence of the liquid and solid phases. This type of crystallization naturally makes the diffusion exchange much easier at any moment in the process of solidification. This probably explains the ease of obtaining a uniform solid phase of the He^3—He^4 solutions in wide containers, which has been reported in some papers.[1-3]

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