DENSITY MEASUREMENTS IN SOLID He^4*

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AF 49(638)-690

An apparatus has been constructed for measuring the density of solid helium as a function of temperature and pressure by measuring the dielectric constant at microwave frequencies. The density of a non-polar substance can be calculated from the dielectric constant using the Clausius-Mossotti relation:

$$\left(\frac{\epsilon - 1}{\epsilon + 2} \right) \frac{M}{\rho} = \frac{4\pi}{3} A,$$

where ϵ is the dielectric constant, M is the molecular weight, ρ is the density, and A is the molar polarization. For a microwave cavity, ϵ is given by

$$\epsilon = \left(\frac{f_o}{f} \right)^2 ,$$

where f_o is the resonant frequency when evacuated and f is the frequency when filled with the sample.

The measurements reported here are on solid He^4 in the region of the γ (body-centered cubic) phase discovered by Vignos and Fairbank.[1] Goldstein has predicted[2] that solid He^4

No DDC limit

should have anomalous thermal properties, including a negative isobaric thermal expansion coefficient ($\alpha = 1/V(\partial V/\partial T)_p < 0$) near the melting curve. More recently Grilly and Mills[3] have measured the molar volume of liquid He^4, the volume change upon melting of the solid, and the volume change of the transition between the α (hexagonal close packed) and γ phases. They have also made estimates of the isothermal compressibility and the thermal expansion coefficient of the solid, with an indication that the latter was negative in the ranges 1.3 - 1.5°K and 1.8 - 1.9°K near the melting curve.

In this apparatus the microwave signal is generated at 10 Kmc and multiplied to 20 Kmc by a crystal multiplier. The 20 Kmc signal is fed to the cavity through a matched magic tee, and the reflected signal from the cavity is received at a crystal detector on another arm of this same tee. Part of the 10 Kmc signal is tapped off with a directional coupler, reflected from an accurately calibrated wavemeter, and detected at a crystal. Attenuators reduce the microwave power input to the cavity to about 10^{-5} watts.

The cylindrical sample cavity, which operated in the TE_{111} mode, was machined in two parts from electrolytically pure copper. It is contained in a thick walled copper mounting block and is completely surrounded by the pressurized helium

sample to eliminate distortions due to pressure changes. The waveguide leading down into the helium bath is of thin walled stainless steel.

In the waveguide, just above the cavity coupling hole, is a high pressure seal matched to transmit the microwave signal. The seal was formed between two flanges with a Teflon gasket and an indium 'O' ring. The waveguide section at each flange was filled with a casting of epoxy resin to provide a mechanically strong backing for the Teflon gasket.

Commercial bottled helium purified in a liquid nitrogen cooled charcoal trap was used for the sample. Compressed nitrogen gas applied across a mercury filled stainless steel U-tube and a stainless steel Toepler system were used to pressurize the sample. The sample fill line was insulated from the cryostat bath by a vacuum jacket, and inside the jacket near the base a wire heater was wound on the fill line. The uninsulated section of fill line wes 3 mm i.d. and 4 cm long. The insulated line was 0.7 mm i.d. stainless steel capillary. It is assumed that the vacuum jacket and the heater insure that the smaller diameter section of the fill line is not plugged with solid.

Pressures were measured with a 0 - 50 atm Heise bourdon tube gauge accurate to 0.05 atm. The gauge used was calibrated

against a similar model which in turn had been recently calibrated against standard gauges by the manufacturer. The temperatures were calculated from the vapor pressure of the cryostat bath, which was held constant to 0.002°K by a Walker pressure regulator.[4]

The klystron was frequency modulated at 1 kc over a mode, and the resonant absorption curves of the sample cavity and the calibrated wavemeter were observed simultaneously on a dual beam oscilloscope. When the absorption curves coincided the cavity frequency was twice the frequency of the wavemeter.

A major difficulty in measurements on solid helium is establishing pressure equilibrium within the sample. The low surface-to-volume ratio of the cylindrical microwave cavity allows equilibrium to be reached easily within the cavity itself. However, solid plugged the uninsulated section of the fill line and the cavity coupling hole. It was found that for increasing pressures the equilibrium times were very long, on the order of hours, whereas for decreasing pressures they were of the order of 15 minutes. The technique that gave the best results was to force the solid in with an over-pressure of up to 3 atm and then to decrease the pressure gradually until the sample resonant absorption curve moved readily and consistently with further decreases of pressure. For a given change in pressure the sample was assumed to be in equilibrium

when no measurable additional shift in the frequency occurred in a time long compared to the time for the original observed change.

Points were taken at constant temperature for six temperatures between 1.45°K and 1.73°K. The results for pressures near the melting and transition curves are shown in Fig. 1. The lower density γ phase is clearly seen as the first step in the curve. The measured volume changes for melting and for the solid transition are listed in Table I. The volume changes for melting agree with those of Grilly and Mills[3] to within 2-3%, while the solid transition volume changes are slightly higher than theirs but in agreement within the experimental error. Their data indicated a minimum in the solid transition volume change near the center of the γ phase. No such effect is seen here.

TABLE I

Measured Volume Changes in Cm^3 per Mole at the Melting Point and at the Solid Transition Point

Temp. °K	1.457	1.488	1.567	1.601	1.651	1.724
ΔV_m	1.82	1.81	1.74	1.70	1.64	1.50
ΔV_{tr}	0.19	0.20	0.21	0.21	0.19	0.21

No attempt was made to resolve the pressures at melting and at the transition accurately. In general the measured pressures are 0.05 - 0.14 atm higher than those measured by Grilly and Mills[3] and about the same amount lower than those measured by Vignos and Fairbank.[1] The isothermal compressibility of the α phase was calculated from points taken at pressures of 30 - 34 atm and was $(3.9 \pm 0.2) \times 10^{-3}$ per atm. The compressibility of the γ phase can only be estimated from the points shown in Fig. 1. It appears no greater than that of the α phase, and possibly less. Grilly and Mills[3] estimated the compressibility of the α and γ phases to be 2.6×10^{-3} and 3.3×10^{-3} per atm, respectively.

Of particular interest is the isobaric thermal expansion coefficient of the solid. The points taken at 1.488°K in the α phase indicate a slightly higher density than at 1.457°K, but the difference is less than the experimental error. From 1.57°K to 1.72°K the expansion coefficient is positive and less than 10^{-2} per °K. One set of points was taken at a constant pressure of 28.60 atm between 1.1°K and 1.7°K. The thermal expansion coefficient was everywhere positive and less than 10^{-2} per °K. The thermal expansion coefficient of the γ phase can be estimated by extrapolating the lines through the points in Fig. 1. Again, the density appears to decrease with

increasing temperature except at 1.488°K, and again the differences are less than the experimental error.

The sources of error in this technique are: i) non-equilibrium between the pressure in the sample and the external system, ii) changes in the zero density frequency of the experimental cavity, iii) errors in the apparent frequency due to mismatches in the coupling circuit, iv) uncertainty in the limit of accuracy of the Clausius-Mossotti relation and the value of the helium polarization.

By comparing the data on the liquid with that of Grilly and Mills[3] the error in the absolute density values due to ii) and iii) was estimated to be less than $\pm$ 0.0005 gm/cm^3, or 0.3%. The error for small variations in density during a particular run is about $\pm$ 0.0001 gm/cm^3. The molar polarization of He^4 from 1.4 - 1.7°K as calculated from the volume of the liquid at the melting curve[3] is 0.1238 $\pm$ 0001 at 20 Kmc, as compared to the value of 0.1228 at 9.1 Kmc and over the same temperature range measured by Grebenkemper and Hagen[5] for the liquid at vapor pressure.

The cavity used in the present measurements was calibrated between 1.8 and 1.1°K and no measurable change ($\pm$ 0.2 mc) in the zero density resonant frequency was observed. It has been noted that between experimental runs (i.e., when the cavity

is warmed to room temperature and exposed to air) changes in the zero density frequency of 0.03% can occur.

A capacitance-type transducer pressure gauge has been designed and built to be mounted directly on the sample cavity and will be used to measure the pressure in future work. It is hoped that a more exact measure of the thermal expansion coefficient will be possible. Later measurements will be extended to He^3 and He^3-He^4 mixtures at pressures up to 130 atm and temperatures down to $0.3^{\circ}K$.

REFERENCES

*This work was supported by an Air Force University Postdoctoral Research Associateship granted by the Air Force Office of Scientific Research.

1. J.H. Vignos and H.A. Fairbank, Phys. Rev. Letters 6, 265 (1961); 6, 646 (1961).

2. L. Goldstein, Phys. Rev. 122, 726 (1961).

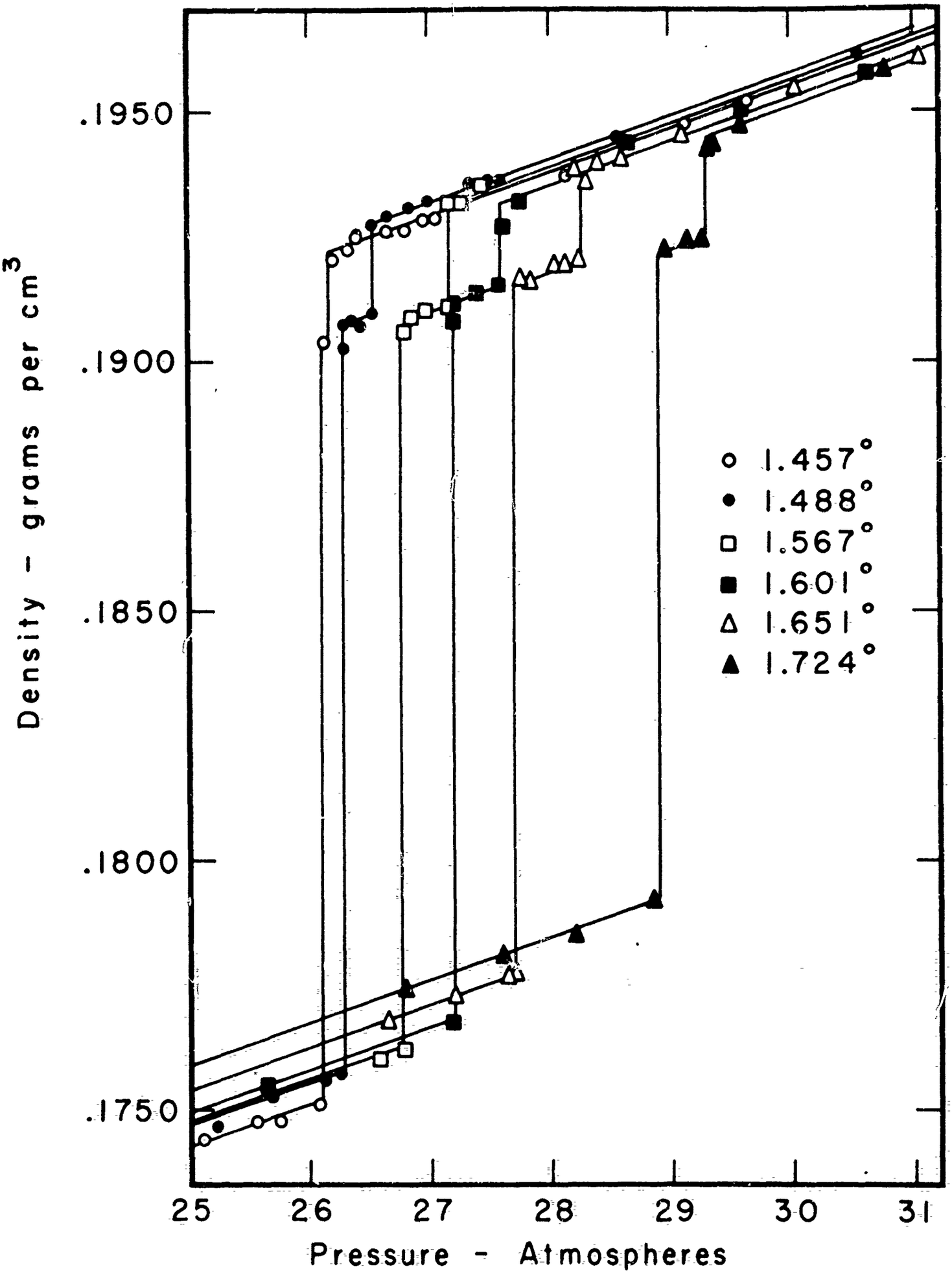
3. E.R. Grilly and R.L. Mills, Ann. Phys. (NY) 18, 250 (1962).

4. E.J. Walker, Rev. Sci. Inst. 30, 834 (1959).

5. C.J. Grebenkemper and John P. Hagen, Phys. Rev. 80, 89 (1950).

FIGURE CAPTION

Fig. 1: The density of solid He^4 as a function of pressure at six temperatures, showing the melting point and solid phase transition.


.1950
.1900
.1850
.1800
.1750
Density - grams per cm3
25
26
27
28
29
30
31
Pressure - Atmospheres
1.457°
1.488°
1.567°
1.601°
1.651°
1.724°