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Four-Window Cell and Cryostat for High Pressure Studies at Liquid Helium Temperature*,†

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A compact gas high pressure cell for measuring the optical properties of solids at low temperatures under pressures up to 10 kilobars is described. Using helium gas as the pressure-transmitting medium, the pressure is purely hydrostatic at 78°K and nearly hydrostatic at 7–12°K. Techniques are discussed for sealing windows of sapphire for optical studies, of beryllium for x irradiation, and dummy windows for electrical connections. A cryostat which can be accommodated in most recording spectrophotometers is also described.

INTRODUCTION

IN extending techniques for the measurement of pressure effects to low temperatures, the principal design considerations are the need to keep the mass of the pressure cell to a minimum, the requirement of effective cryogenic pressure seals, and the problem of achieving and measuring hydrostatic pressure.[1] This paper describes a versatile four-window high pressure cell which has proved quite satisfactory for this temperature range. Because of its compact size, not only are the liquid helium requirements modest, but also the cell and its cryostat are small enough to fit directly into the sample chamber of many recording spectrophotometers. By using helium gas as the pressure-transmitting medium, hydrostatic pressures are possible at temperatures down to the freezing curve of helium, while at lower temperatures the solidified helium is sufficiently soft and plastic that nearly hydrostatic conditions exist after slow cooling. Since the sample does not undergo plastic deformation or shear, it is possible to study structure-sensitive properties. The techniques for sealing high pressure gas at low temperatures, while not novel in themselves, differ in several details from conventional techniques, particularly in the use of indium metal packing rings. With its small internal volume, the pressure cell is suitable for use with a tea-cart type of pressure system.

The cell has been used principally in a study of color center formation in alkali halides at low temperatures and high pressures.[2] In this application, the color centers were produced by x irradiation through a beryllium window, and were detected by measuring the optical absorption at right angles through sapphire windows. In this case, the helium gas had the additional advantage of transparency to both visible light and x rays, as well as being chemically inert under irradiation.

The apparatus is described and illustrated as it was used in this application. In addition, techniques for introducing electrical leads are presented, and the determination of pressure and temperature at the sample discussed.

DESCRIPTION OF APPARATUS

A. Pressure Cell

The pressure cell used throughout this work is illustrated in Fig. 1. The cell was designed for and tested at pressures up to 10 kilobars,[3] and no leakage or window fracture has occurred at the maximum operating pressure of 8 kilobars.

The dimensions of the cell represent a compromise to keep both the mass to be cooled and the optical path length to a minimum and still have a reasonable aperture, in this

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[1] For a recent review of high pressure techniques and measurements, see C. A. Swenson, "Physics at High Pressure" in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1960), Vol. XI.
[2] D. B. Fitchen, Ph.D. thesis, University of Illinois, Urbana, Illinois, 1962 (results to be published).
[3] 1 bar = 10^6 dyn/cm^2 = 1.0197 kg/cm^2 = 0.9869 atm.

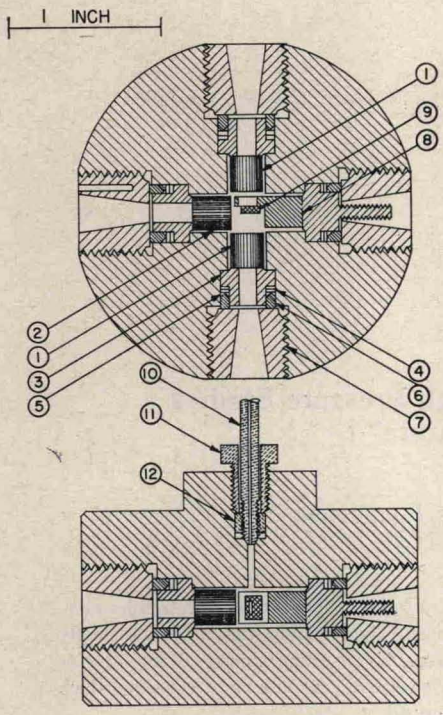

FIG. 1. The pressure cell as used for color center measurements.

case $\frac{1}{8}$-in. diameter. The outside dimensions are $2\frac{1}{4}$ in. in diameter by $1\frac{1}{2}$ in. in length, and the assembled unit weighs 23 oz.

Beryllium-copper was chosen for the cell body and various parts of the window ports because of its ease of machining and heat treating, and to avoid any problems of brittle fracture with steels at low temperatures. Beryllium-copper has been used for nonmagnetic pressure vessels, but has not been used widely in other high pressure work because of some early ruptures. The problems of selection of stock free of flaws and the performance of pressure vessels made of this alloy have been described in the literature.[4] Apparently, the trouble arises from bubbles and flaws in the hot-rolled stock used for the larger vessels. When only small-diameter vessels are needed, Paul[5] had indicated that the cold-rolled stock obtained from the supplier can probably be trusted without careful ultrasonic testing.

In the present case, the cell body was machined from the largest size of cold-rolled stock available, a $2\frac{1}{4}$-in.-diam rod of $\frac{1}{2}$-hard "Berylco 25" alloy. After machining, the piece was hardened to about Rockwell 40-C by heat treating at 600°F for 2 h, using an argon atmosphere to keep surfaces clean. Window parts were made from smaller rods of the same material.

The seats for the windows are of conventional Bridgman design[6–8] in a compact version. The optical windows (1) are oriented Linde synthetic sapphires $\frac{1}{4}$ in. in diameter by $\frac{1}{4}$ in. long, ground optically flat on the ends. The x-ray window (2) is "Berylco PX-20" extruded beryllium rod, supplied by the Beryllium Corporation of America as cylinders of the same size which were then lapped flat. The polished faces of these windows are glued with an adhesive mixture to the lapped and polished faces of the "mushroom plugs" (3). The mushroom plug rests on thin bronze, indium, and copper packing rings (4), prevented from extruding by small beryllium-copper rings of triangular cross section (5), which fit against the face of a hardened support ring (6). The support ring is held in place by the threaded plug (7) which has a 10° tapered aperture hole to reduce shadowing, and holes for a three-pin wrench so that it can be screwed in flush with the cell body.

In this experiment, the sample holder (8) is mounted on the fourth plug, a blind version of the other three, to provide easy access. The sample (9) is held on a fixed copper slit and surrounded by baffles to prevent light leaks around the sample holder.

The pressure connection is made with hard-drawn 316 stainless steel tubing (10), $\frac{1}{8}$-in. o.d. by 0.025-in. i.d., supplied by Harwood Engineering Company, and coupled to the cell with a standard Harwood gland nut (11) and collar (12).

B. Window-Sealing Techniques

The design of the window ports owes much to earlier designs of Warschauer and Paul,[7] and of Langer and Warschauer.[8] Various attempts to make a more compact window using soft metal O-ring seals proved unreliable at low temperatures, so the conventional "unsupported area" seal was used. The $\frac{1}{4}$-in.-diam stem of the mushroom plug is unsupported, thus maintaining a pressure in the packings twice that of the gas being sealed. The stem is backed up by the threaded support plug in case it should suffer "pinch-off" from the high pressures at the packing rings, but this has never occurred. The clearance between stem and plug is such that yield before rupture would probably bring them into contact and relieve the stress.

The packing rings selected were 0.035-in. bronze, 0.035-in. indium, and 0.005-in. copper. The bronze and copper rings serve mostly to contain the soft indium ring. Indium is very effective as a gas seal because it adheres well to the beryllium-copper walls to provide an excellent initial seal, and because it flows easily to maintain a seal during thermal cycling. The mushroom plug, packings, and support ring are inserted and removed as a unit, using an extractor tool which screws into the last $\frac{1}{16}$ in. of the aper-

[4] W. Paul, G. B. Benedek, and D. M. Warschauer, Rev. Sci. Instr. **30**, 874 (1959).
[5] W. Paul (private communication).
[6] P. W. Bridgman, *The Physics of High Pressures* (G. Bell and Sons, London, 1949).
[7] D. M. Warschauer and W. Paul, Rev. Sci. Instr. **28**, 62 (1957).
[8] D. Langer and D. M. Warschauer, Rev. Sci. Instr. **32**, 32 (1961).

ture hole. This preserves the packings so that they usually last for about two dozen runs.

To seal the window plugs to their seats, a modified Poulter seal is used.[9–11] The beryllium plugs and the beryllium-copper seats were lapped until matching surfaces were smooth and flat enough to adhere and support their own weight. In principle, the matching flat surfaces provide the gas-tight seal to prevent leakage out of the center hole. In practice, however, it is easier to apply a thin coat of adhesive mixture made of equal parts of G. E. No. 7031 varnish and toluene. The adhesive bond serves to hold the window securely in place during handling and also makes it possible to use parts whose faces no longer adhere because they have become lightly scratched or dented with use.

In preliminary tests, sapphire windows cracked along hexagonal cleavage planes when pressure was released too abruptly. Therefore, the pressure is always changed slowly, usually a few hundred bars per minute, and no difficulty with window fracture has been experienced.

This "unsupported area" design, using the varnish adhesive and soft indium for an initial seal, has proved quite satisfactory for withstanding the shock of cooling to low temperature and subsequent varying of pressure. Its principal shortcoming is the fact that the seal is unstable between 0 and 2000 bars at low temperatures. If the pressure is allowed to drop to this level, leaks develop through the adhesive coating.

C. Electrical Leads

When necessary, electrical leads can be introduced to the sample space in two ways. Fine wires can be led down the pressure tubing from a conventional closure at room temperature or a frozen oil seal.[12] Alternatively, one of the window ports can be used for the purpose. The sapphire window is replaced by a shorter beryllium-copper disk with a lapped face and shallow tapped holes for terminals for a single lead. The varnish coating insulates the disk from the rest of the cell, which then serves as a terminal for the other lead.

D. Cryostat

A diagram of the cryostat used in the color center experiment is shown in Fig. 2. It is a special Pyrex double Dewar system built by H. S. Martin and Son.

The pressure cell is mounted in contact with the inner Dewar filled with the appropriate refrigerant. The outer Dewar terminates in a Kovar cup which gives good thermal contact to the copper radiation shield extending down into the tail of the cryostat. The outside diameter of this

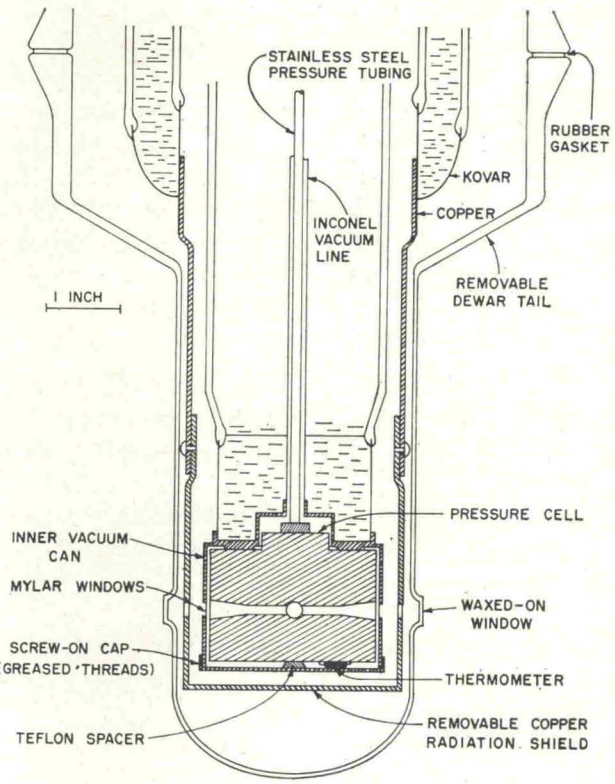

FIG. 2. Tail section of the cryostat.

tail is $3\frac{1}{2}$ in. so that it fits directly into the light proof housing on a spectrophotometer. This tail section and the lower part of the radiation shield are removable to allow easy access to the pressure cell between runs.

The Dewar volumes were chosen large enough to avoid frequent transfers and to maintain the cell at helium temperature overnight. Assuming that cooling results from the heat of vaporization alone, the 650-g cell requires $1\frac{1}{2}$ liters of liquid helium to cool from nitrogen to helium temperature. Once cooled, the helium loss rate is about 100 cc/h, so that the 2-liter inner Dewar is more than adequate. The nitrogen loss rate of the outer Dewar is such that its level drops only half as fast.

The cryostat employs a common vacuum for both Dewars, and any leakage of helium from the pressure cell is intolerable with the pressure ratio of 10^{12}. Therefore, the cell is surrounded by a close-fitting vacuum can of copper with a separate vacuum line. For runs where it is necessary to cross the region of instability of 0–2 kilobars at low temperature, the can is fitted with three 0.001-in. Mylar windows fastened to the inside surface with Epoxy resin, and the threads on the end closure of the can are coated with vacuum grease. In this way it is possible to isolate the cell and maintain a vacuum of 10^{-6} to 10^{-5} mm Hg in the main Dewar while the helium pressure in the can rises as high as one atmosphere. The disadvantage of this scheme is that the Mylar windows act as thin film interference

[9] T. C. Poulter, Phys. Rev. **35**, 297 (1930).
[10] E. Fishman and H. G. Drickamer, Anal. Chem. **28**, 804 (1956).
[11] The sapphire windows were ordered with the usual specifications that the cylinder axis be parallel to the c axis within 5° and the faces be flat to within 0.0001 in.
[12] J. S. Dugdale and J. A. Hulbert, Can. J. Phys. **35**, 720 (1957).

filters and thus complicate the background absorption correction, so they are left off whenever possible.

In cases where only a brief measurement at a single pressure is necessary, a small metal cryostat has been built in which the pressure cell itself serves as the bottom closure of the helium cold finger, and no separate vacuum jacket is provided. This is feasible because the leakage from the cell is zero when it is properly assembled and pressurized at room temperature before cooling down.

E. Gas Pressure System

Helium gas is supplied to the cell from a remote, intensifier-driven system, assembled from commercially available parts, which can compress gases to 10 kilobars. Contrary to usual practice, a needle valve is included in the high pressure gas line so that the intensifier can be recycled if necessary and the pressure cell sealed off from the system during a run.

F. Temperature and Pressure Determination

The temperature of the pressure cell is measured externally with a thermocouple or carbon resistor. It is assumed that this gives a good indication of the sample temperature, since the sample is surrounded by compressed helium and a large thermal ballast. With liquid helium in the Dewar, the temperature for a given run falls in the range 7 to 12°K, while with liquid nitrogen it is close to 78°K.

The determination of pressure at the sample is much more difficult. Above the freezing curve of helium the pressure is measured externally, using a coil of No. 40 B. & S. gauge manganin wire located in a separate pressure vessel at room temperature. The pressure is measured with an accuracy of about 0.5%, and can be held constant to within 10 bars. This gauge vessel also serves a secondary function as a pressure reservoir, its larger volume suppressing the variation of pressure with temperature in the cell.

Below the freezing curve, the manganin gauge no longer gives the sample pressure, since the pressure line is blocked with frozen helium. The final pressure in the cell at the lowest temperature can be estimated, however, using thermodynamic data available for solid helium.[13–15] Assuming cooling at constant volume from the freezing point determined by the gauge pressure, the calculated final pressure agrees within about 100 to 200 bars with that deduced indirectly from the pressure shift of the F-center absorption band. Part of this discrepancy is probably due to the uncertain correction for the change in volume on freezing, Δv_m, which affects the final pressure if the helium in the tubing freezes before that in the cell. This is apparently not the case for the cryostat described above. Several techniques for measuring the internal pressure directly and precisely at these temperatures are being investigated.

In order to change pressure during helium temperature runs, it is necessary to warm the cell and pressure tubing above the helium melting temperature, change pressure, and recool.

DISCUSSION

The performance of the apparatus described above has demonstrated the feasibility of a whole class of new measurements at high pressures and low temperatures. The cell requires little or no modification to serve in studies of the Mössbauer effect, γ- and x-ray radiation effects, photoconductivity, and fluorescence, for example. Certain limitations and difficulties have also been exposed. In particular, the problems of leakage at low pressure and accurate pressure calibration at helium temperature will require further development of techniques.

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[13] J. S. Dugdale and F. E. Simon, Proc. Roy. Soc. (London) **A218**, 291 (1953).
[14] F. A. Holland, J. A. W. Huggill, and G. O. Jones, Proc. Roy. Soc. (London) **A207**, 268 (1951).
[15] E. R. Grilly and R. L. Mills, Ann. Phys. **8**, 1 (1959).