NEW CRYOGENIC METHODS OF USING LIQUID HYDROGEN[1]

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An apparatus has been developed for producing liquid hydrogen in amounts of 3 to 5 liters for use under conditions where portability and simplicity are important. The liquefier is of the Ahlberg type, operating from ordinary tank hydrogen, purified by passing over charcoal at 90°K. It produces about 1.5 liters of liquid hydrogen in 45 min. from three standard tanks dropped from 2000 to 1000 lb. in.$^{-2}$ A small cryostat has been designed in which a liter of liquid hydrogen will maintain temperatures from 14–20°K. over a period of 20 hr. The cryostat is 14 in. long and 6 in. in diameter. It contains an inner jacket of 1 liter of liquid nitrogen to reduce heat leak to the liquid hydrogen. It is fitted with electrical leads and a rock salt window specially designed for optical studies of superconductors.

I. INTRODUCTION

During the last half-century of physics and chemistry, the use of liquid oxygen and liquid nitrogen has progressed from the stage of a laboratory tool available only in a few specially equipped institutions to a broad field of techniques widely applied both in research laboratories and in industry. On the other hand, the availability of liquid hydrogen or liquid helium is still very restricted. There is increasing evidence, however, that these ultimate refrigerants can be of great help in many fields, especially if they can be produced by apparatus which does not require a person specially trained in low-temperature technique for its operation. Particularly in spectroscopy the possible uses of low-temperature bolometers create a need for simple portable apparatus to produce and maintain temperatures below 20°K. As a first step in filling this need, we have undertaken the development of a light portable liquefier for hydrogen, operating from standard commercial tank hydrogen, and of a small cryostat which can be used in connection with other pieces of apparatus, such as spectrographs, which will maintain temperatures of 20°K. or less for periods of 20 hr. on a single filling with liquid hydrogen. It is the purpose of this paper to describe these pieces of apparatus.

II. THE HYDROGEN LIQUEFIER

Ahlberg, Estermann, and Lundberg (1) some years ago described a small portable liquefier for producing hydrogen from tanks. Our instrument is essentially a further development of that design. It is made up of four functional parts as follows: the controls, the nitrogen-bath unit, the liquefaction unit, and the storage unit. These fit together in a single frame for convenience in transportation.

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The whole liquefier is shown schematically in figure 1. The liquid-nitrogen reservoir is shown at N. The high-pressure hydrogen gas enters the liquefier through valve V1 at room temperature, after having passed through a purifier, which will be described in a later section. Gauge G1 is a high-pressure gauge which indicates the pressure of the hydrogen gas as it is received from the purifier unit. The gas goes along the high-pressure line (HP) and enters an inter-

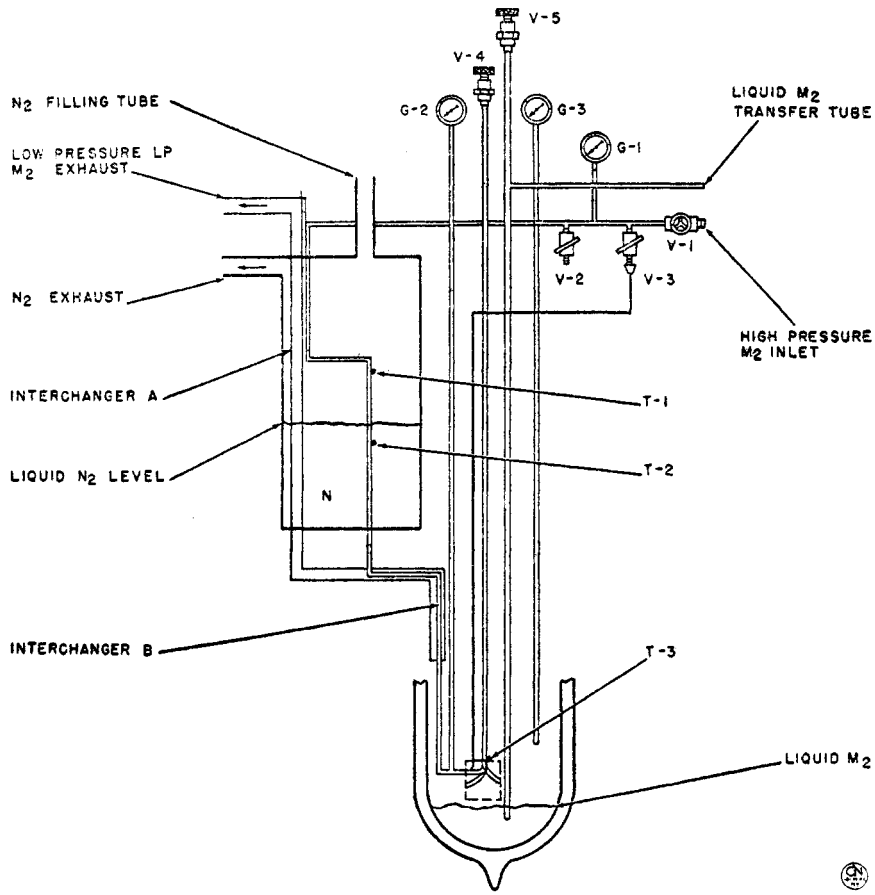

FIG. 1

changer (A), located inside the top of the liquid-nitrogen container. The high-pressure gas then goes through a copper coil within the liquid-nitrogen bath, and after that passes through interchanger B to the expansion valve T3. The pressure at this point is roughly 2000 lb. per square inch and is registered on gauge G2. In passing through the expansion valve, the gas drops to a pressure of about 5 lb. per square inch, as registered on gauge G3. There is the usual arrangement for collecting in a Dewar vessel that portion of the gas which liquefies, usually about 10 per cent. The remaining gas passes back through

the interchangers B and A. The percentage conversion to liquid decreases as the pressure falls and is about 3 per cent at 900 lb. per square inch.

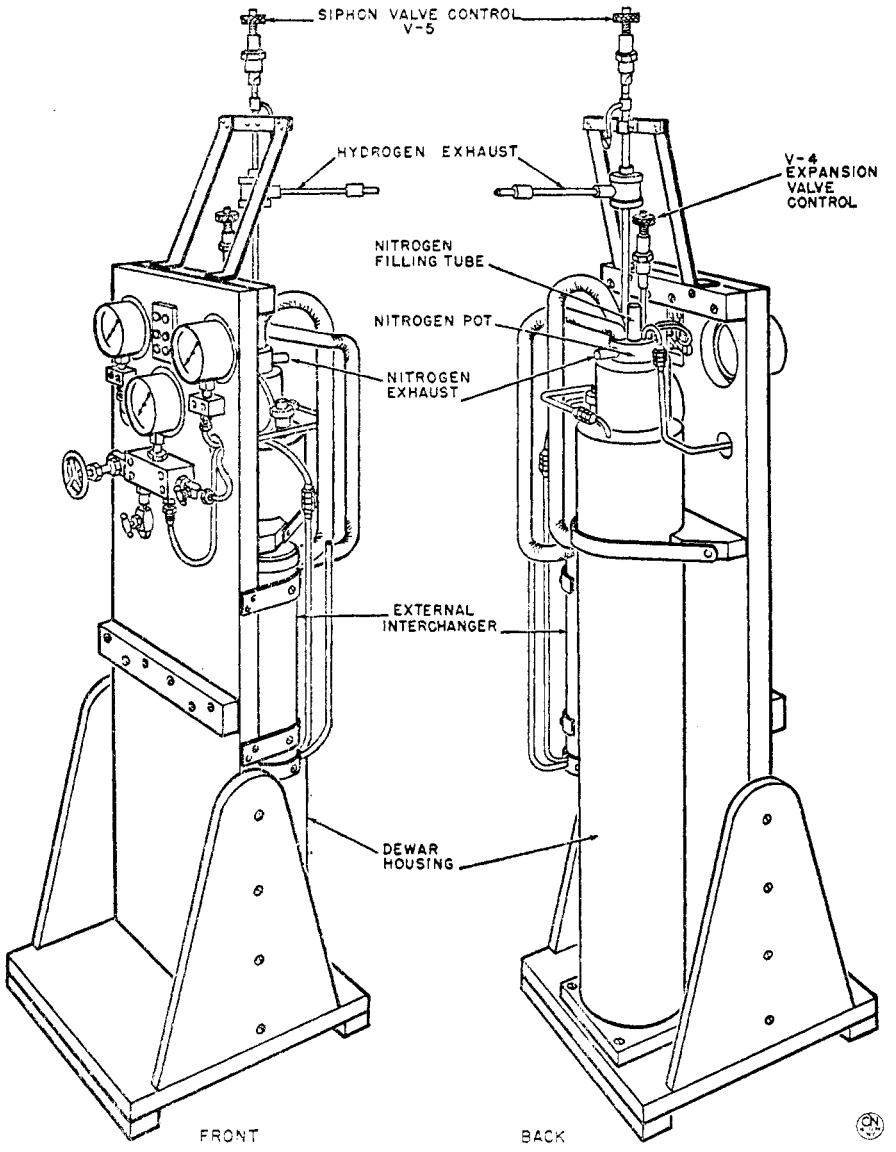

FIG. 2

The assembly of the apparatus is shown in perspective in figure 2. The controls (V1, V2, and V3) and gauges are mounted on the front of the upright wooden panel, which is about 3 ft. high. The liquefier case is on the back, with the two needle-valve controls projecting above. The lower valve (V4)

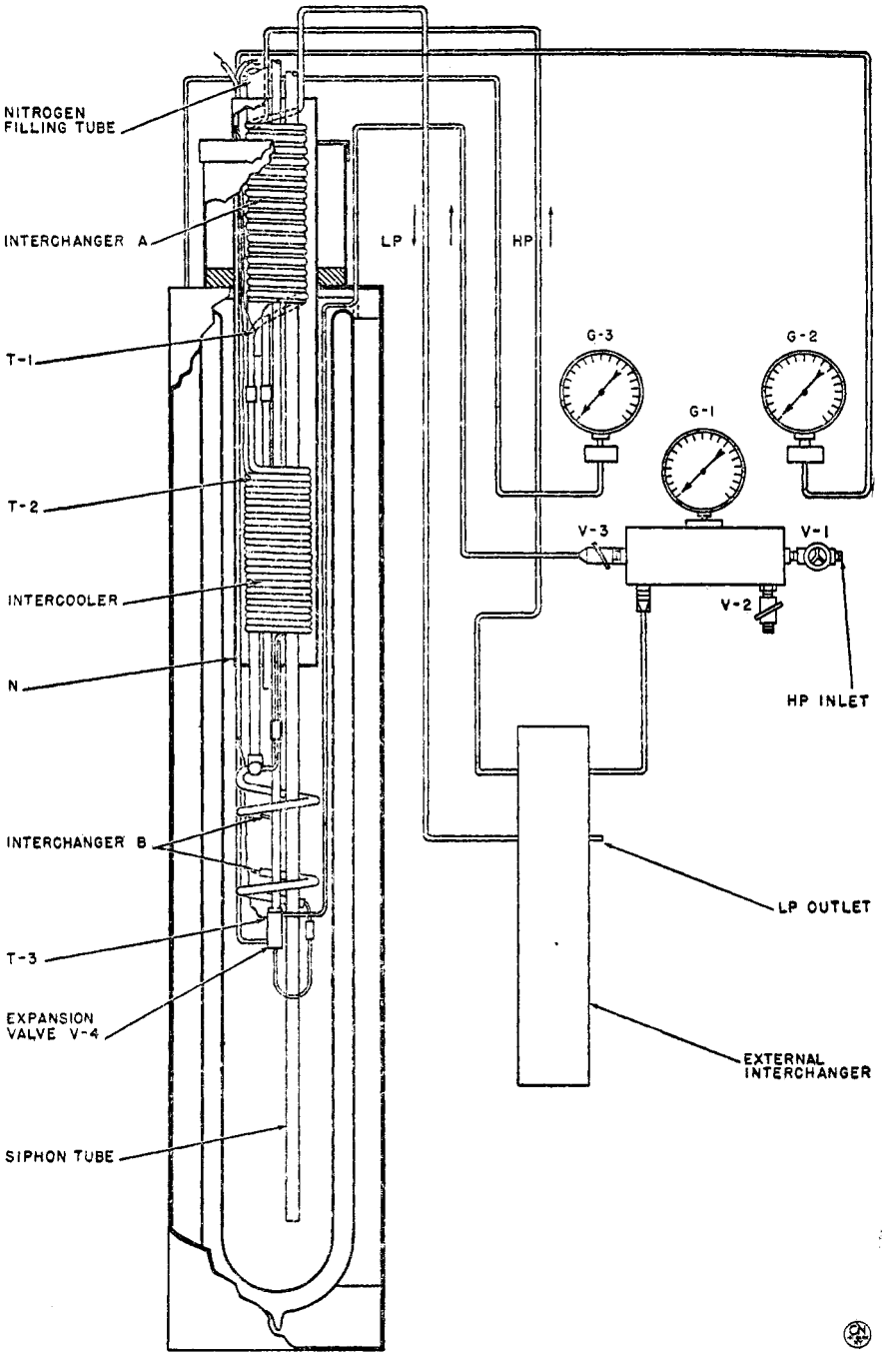
NITROGEN FILLING TUBE
INTERCHANGER A
T-1
T-2
INTERCOOLER
N
INTERCHANGER B
T-3
EXPANSION VALVE V-4
SIPHON TUBE
LP
HP
G-3
G-2
G-1
V-3
V-1
V-2
HP INLET
LP OUTLET
EXTERNAL INTERCHANGER

FIG. 3

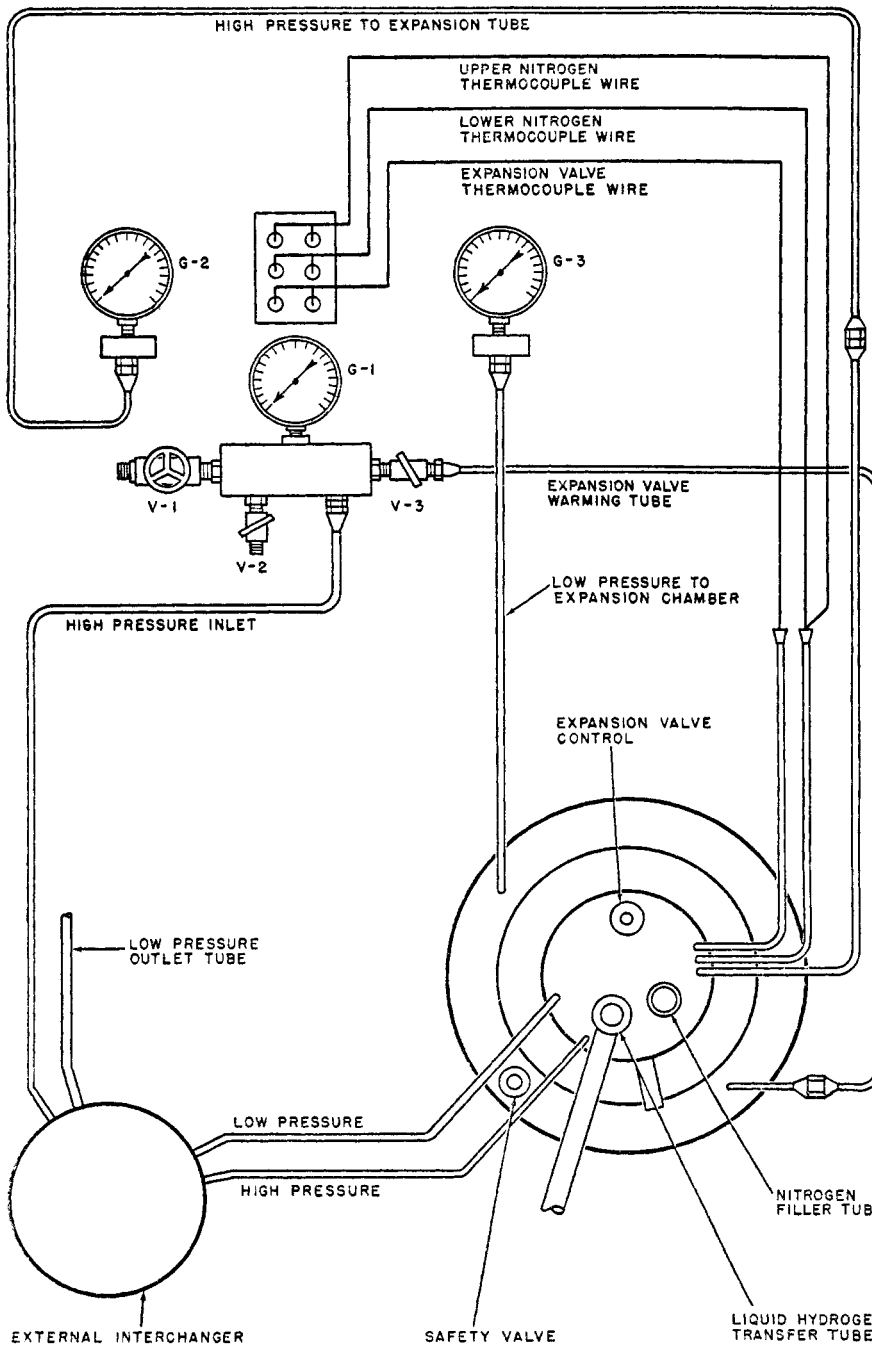
HIGH PRESSURE TO EXPANSION TUBE
UPPER NITROGEN THERMOCOUPLE WIRE
LOWER NITROGEN THERMOCOUPLE WIRE
EXPANSION VALVE THERMOCOUPLE WIRE
G-2
G-3
G-1
V-1
V-2
V-3
EXPANSION VALVE WARMING TUBE
LOW PRESSURE TO EXPANSION CHAMBER
HIGH PRESSURE INLET
EXPANSION VALVE CONTROL
LOW PRESSURE OUTLET TUBE
LOW PRESSURE
HIGH PRESSURE
NITROGEN FILLER TUBE
EXTERNAL INTERCHANGER
SAFETY VALVE
LIQUID HYDROGEN TRANSFER TUBE

FIG. 4

controls the expansion valve and the upper (V5) controls the siphoning off of liquid hydrogen.

Figure 3 shows the main case, with cover partly cut away. Figure 4 shows the distribution of inlet and outlet tubes. Figure 5 shows the detail of the needle valve controlling the siphoning off of liquid hydrogen. Figures 6 and 7 show front and rear photographs of the whole assembly.

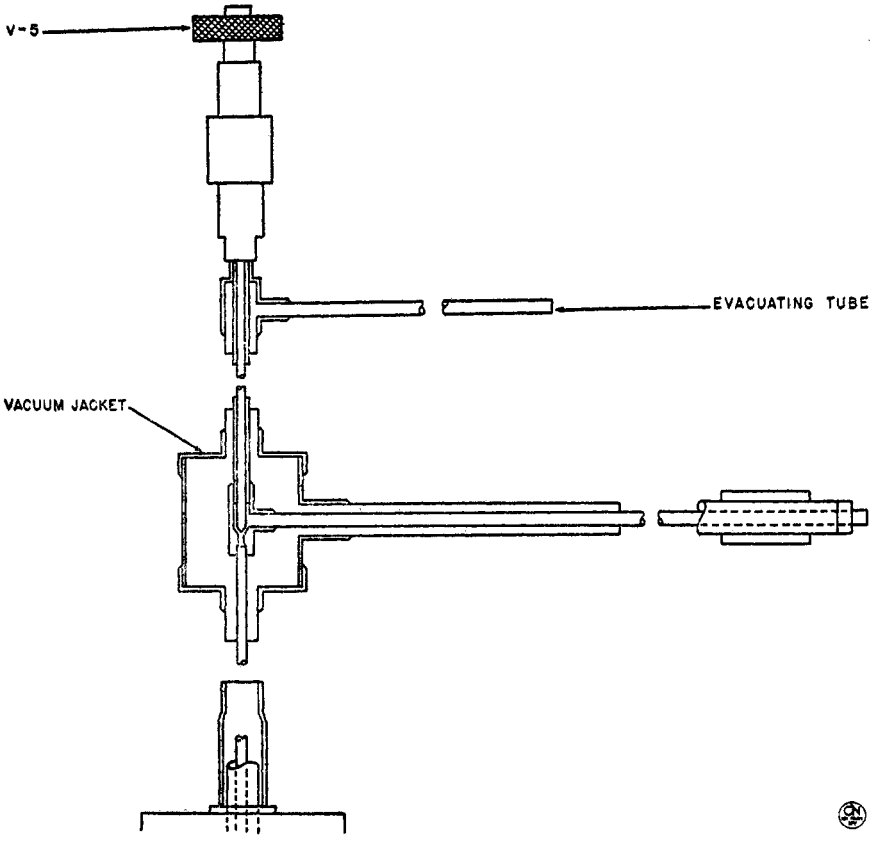

FIG. 5

As shown in figure 3, the first stage of the liquefier consists of a large supernickel tube containing both a coiled double-tube copper interchanger and a single copper tube coil in the nitrogen bath. This will be referred to as the "nitrogen-bath unit". The double-tube copper interchanger is made by soft-soldering side by side a $\frac{1}{4}$-in. copper tube and a $\frac{1}{8}$-in. copper tube. This soldered unit is then formed into a coil small enough in diameter to fit inside the large supernickel housing tube. In the unit shown in figures 6 and 7 an external interchanger was also used, coupled ahead of this interchanger.

The intercooler is made of about 20 ft. of $\frac{1}{8}$-in. diameter tubing wound into a coil so that it also will fit inside the same supernickel housing tube. The small

tube of the interchanger is coupled to the intercooler and the two are located, interchanger above intercooler, as shown in figures 2 and 3. The return gas comes from the twisted-tube interchanger and passes directly through a tube in the liquid nitrogen which covers the entire intercooler coil; the return gas tube is then joined to the large copper coil of interchanger A. Soldered to each

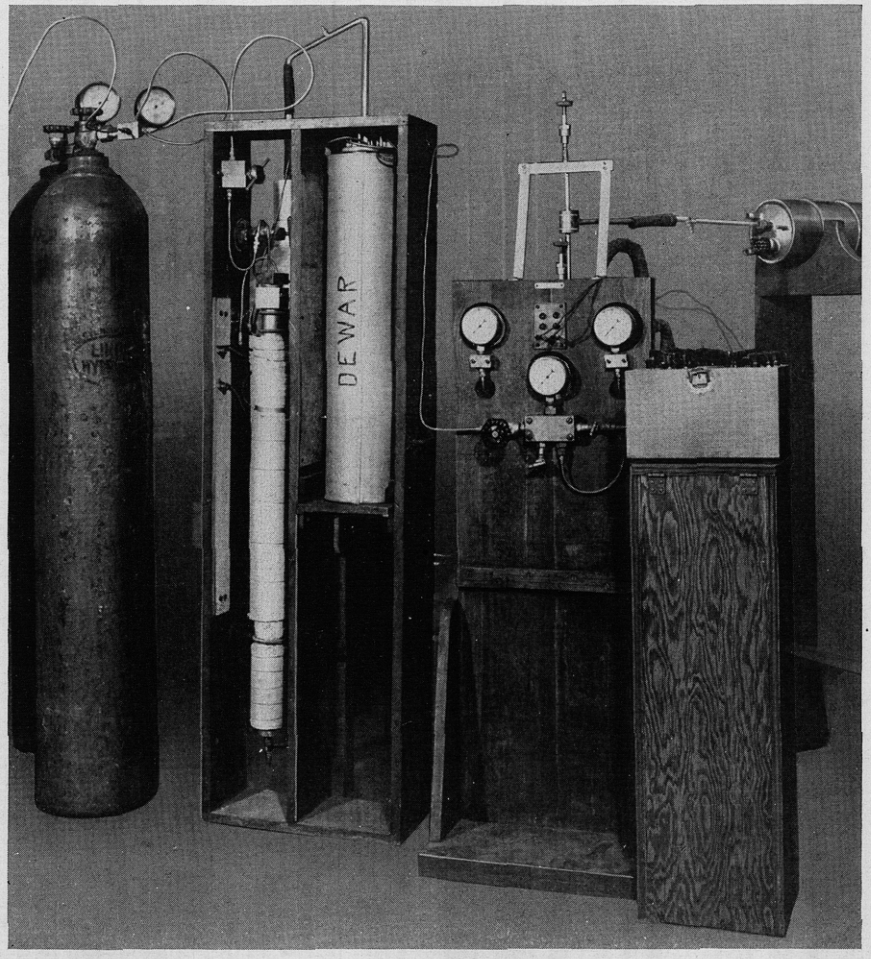

FIG. 6

end of the supernickel housing tube are heavy brass plates, through which all tubes enter and leave the nitrogen-bath unit through soldered connections. This nitrogen-bath unit is supported by a brass disc which is hard-soldered to the outside of the supernickel tube and soldered to the top edge of the main case tube. Around the upper half of the nitrogen-bath unit there is an insulated section made by soldering a brass tube around the supernickel tube and to the support plate. Into the space formed by these two concentric tubes, a filler of hair felt has been packed and a brass cap soldered to the top.

The interchanger B is soldered, directly below the nitrogen-bath unit, to the $\frac{1}{8}$-in. copper tube that comes from the lower end of the intercooler coil. This interchanger (B) is made by rolling $\frac{1}{8}$-in. O.D. x $\frac{3}{32}$-in. I.D. well-annealed supernickel tubing until a space between the walls has been reduced to 0.005 in. After rolling, this tube is then twisted until there is a lead from one twist to the next of about 1 cm. Each end of this twisted tube for a distance of about 5 in. is left unflattened and untwisted. Over the twisted tube and fitting very closely

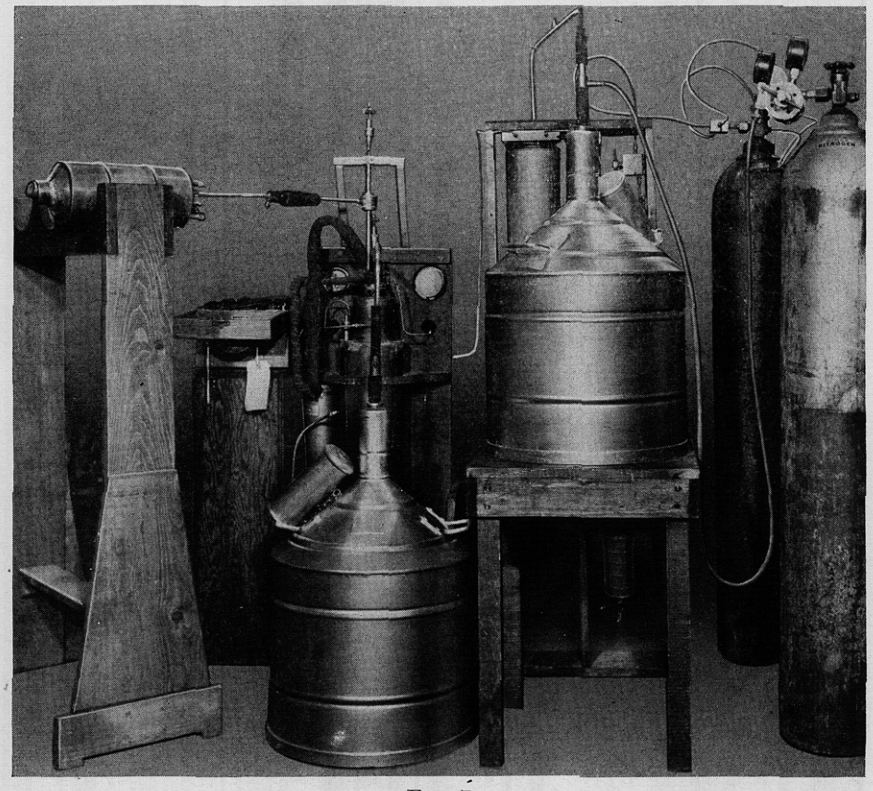

FIG. 7

to it, a lead tube is drawn. This tube is made of lead tubing having an inside diameter of $\frac{3}{16}$ in. and a wall thickness of 0.007 in. A coupling is mounted on one end of the interchanger in such a way that the high-pressure gas may pass through the supernickel twisted tube and return gas may pass around the outside of the twisted tube. The inside of the lead tube is coupled to the return tube which goes through the bottom of the nitrogen-bath unit and through liquid nitrogen, and is then coupled to the $\frac{1}{4}$-in. copper tube of interchanger A. The other end of the supernickel twisted tube then goes to the expansion valve V4, a sensitive needle valve for the control of escaping gas. Packed around the twisted-tube interchanger (B) and tight against the bottom of the nitrogen-bath unit is an insulation made of hair felt held in place by ordinary gauze bandage.

The needle valve V4 is controlled from the top of the liquefier by means of a double-walled, gas-tight, supernickel tube arrangement. This whole unit, comprising the lower portion of the nitrogen-bath unit and the twisted-tube interchanger and expansion valve, is encased in the glass Dewar vessel. The siphoning of liquid hydrogen from the Dewar is controlled by the valve shown in figure 5, vacuum jacketed as indicated. Around the Dewar vessel and insulated from it by hair felt is a brass cylindrical jacket to the top of which is soft-soldered the support flange of the nitrogen-bath unit. Into this flange is mounted a safety valve to relieve any sudden increase of pressure which may build up in the expansion chamber (see figure 2). There is also a tube leading from this flange to a pressure gauge which indicates the internal pressure of the expansion chamber. Soldered through this flange is a small-bore supernickel tube which can lead hydrogen directly from room temperature to the expansion valve seat for the purpose of warming the expansion valve in the event of the formation of a frozen plug of impurity at that point.

All gauges and valves, with the exception of the expansion valve and the transfer valve, are mounted on the wooden panel which forms the stand of the complete unit (figure 2).

III. THE PURIFIER

Our experience has been that standard commercial hydrogen, as received in the usual cylinders, is not of sufficient purity to be suitable for use in our liquefier. There is usually enough impurity of one sort or another to cause clogging after a relatively short period of operation. We therefore place a purifier unit in the line between the source tanks and the liquefier. It is shown diagramatically in figure 8a. It consists, essentially, of two parts: an activated silica gel trap, and an activated charcoal trap, the first of which removes water vapor and any other easily condensible gas, and the second, traces of oxygen and nitrogen. The tube connecting the two traps between B and C can be removed by unscrewing the compression fittings. Details of the traps are shown in figure 8b.

The silica gel trap may be activated in two ways, depending on whether or not compressed air is available. In the first method the heater is connected to a 110-volt source and a slow stream of air is passed in at the point C (figure 8a) with valve V6 open. This hot air then passes through the silica gel trap and produces a desorption of air from the silica gel surface. The air emerges at V6. About 8 hr. are required to activate the silica gel. In the second method, a vacuum pump is connected to the system at C, valve V6 being closed. Heat for desorption is again supplied by the heater. Evacuation should be continued for about 6 hr., and the vacuum pump is protected by a cold trap.

The charcoal trap is activated by evacuation through B, with valve V1 closed. Heat is supplied to the trap by means of a specially constructed heater which can be slipped into place, replacing the Dewar vessel.

Figure 6 shows, reading from left to right, a stand and tank of hydrogen, the purifier, the liquefier (front), and a cryostat (rear view). In the rack which contains the purifier traps, the heater for the charcoal is kept in a pocket at the

lower part of the rack, as is shown in figure 6. The heater is equipped with a copper–constantan thermocouple. When in operation the heater is connected to a source of electric power at 110 volts until the temperature of 150–180°C. is maintained. When the silica gel trap is being evacuated simultaneously, the heaters on the two traps are connected in parallel. After heating the charcoal trap for 6 hr., it is allowed to cool under continuous pumping. It is flushed with hydrogen gas before being used again.

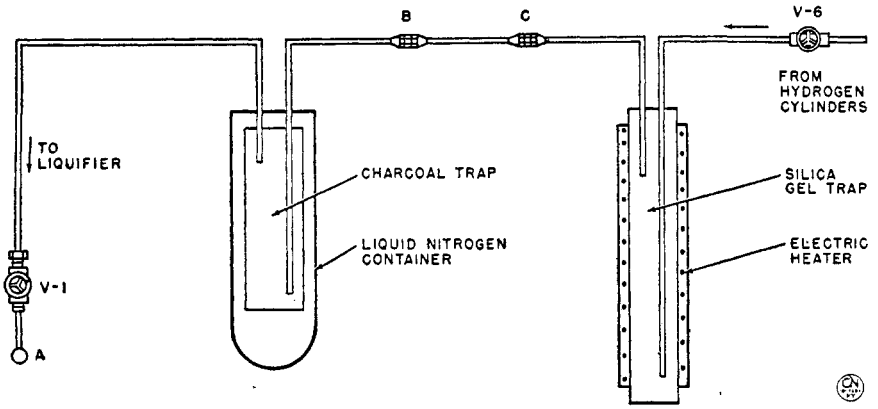

FIG. 8A

IV. CRYOSTAT

In order to provide a chamber for experiments at liquid-hydrogen temperatures with apparatus such as the superconducting bolometer, we have developed a relatively small cryostat in which temperatures between 14° and 20°K. may be maintained for periods up to 20 hr. This cryostat is shown schematically in figure 9. It is made up of a series of nested copper cans spun from copper of wall thickness about 1 mm. The outer case (A) is about 25 cm. long and 12 cm. in diameter. This serves as the vacuum jacket, the vacuum being maintained by the action of the liquid hydrogen inside the inner can, labeled N. This can has a volume of approximately 1000 cc.

A hollow tube passes through N from top to bottom at its center. Within this tube and soldered to it at the bottom there is a copper post (M) made of tubing of $\frac{3}{8}$-in. O.D. and $\frac{1}{4}$-inch. I.D., which is about 2 cm. longer than the hydrogen can and consequently extends out at the top. To the top of this post there is soldered a plate to which the bolometer base, S, is normally attached with low-melting solder. Wound around the post at M is a bolometer-sensitizing coil, made of No. 40 constantan wire, and approximately 500 ohms in resistance. A current passed through this coil raises the temperature of the top of the post and of the bolometer a few tenths of a degree above the temperature of the hydrogen in the can N.

Normally, for bolometer operation, the hydrogen in can N is kept under reduced pressure such that part of it will stay in the frozen state. Thus the can

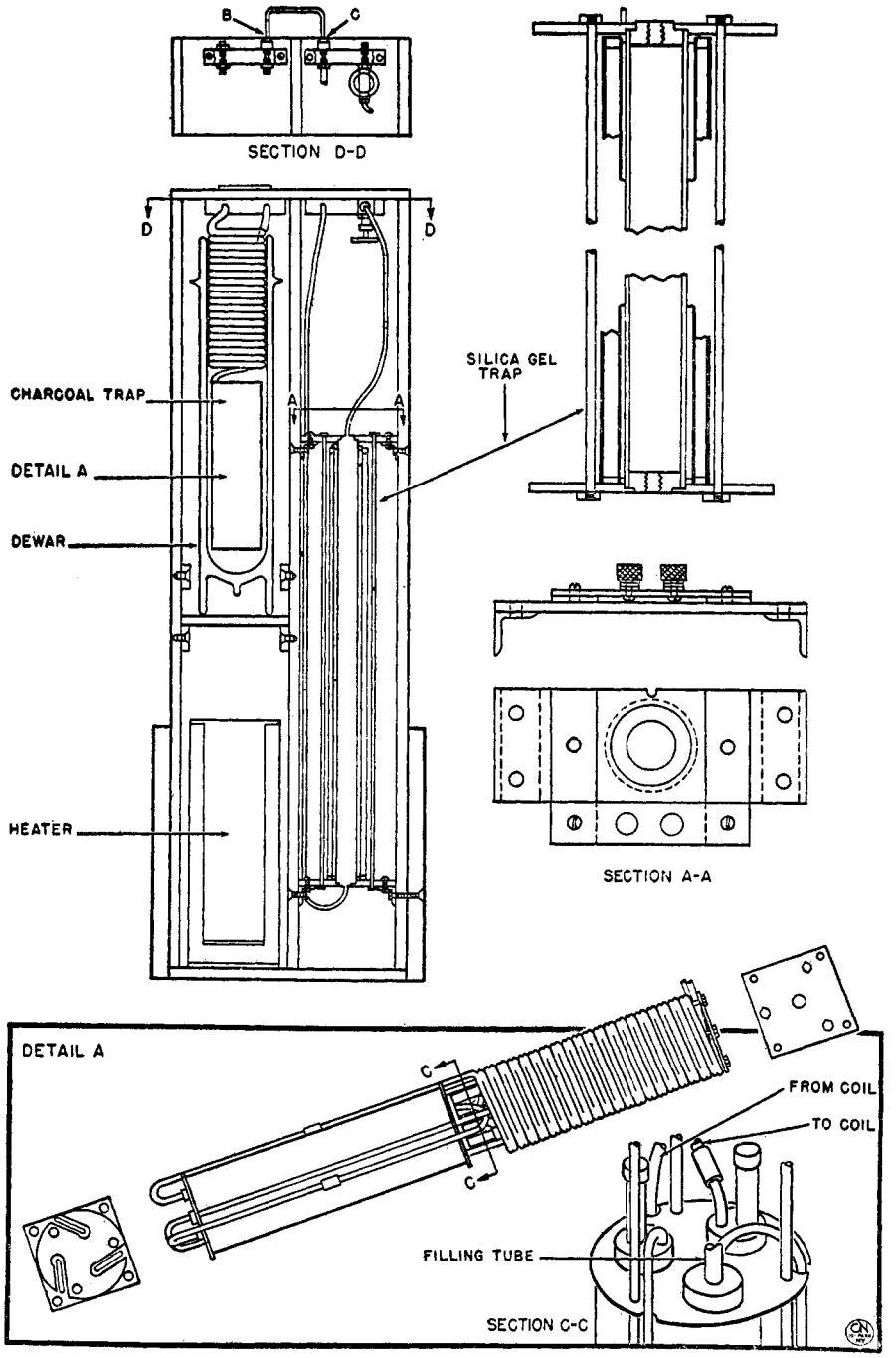
B
C
SECTION D-D
D
D
A
A
CHARCOAL TRAP
DETAIL A
DEWAR
HEATER
SILICA GEL TRAP
SECTION A-A
DETAIL A
C
C
FROM COIL
TO COIL
FILLING TUBE
SECTION C-C

FIG. 8B

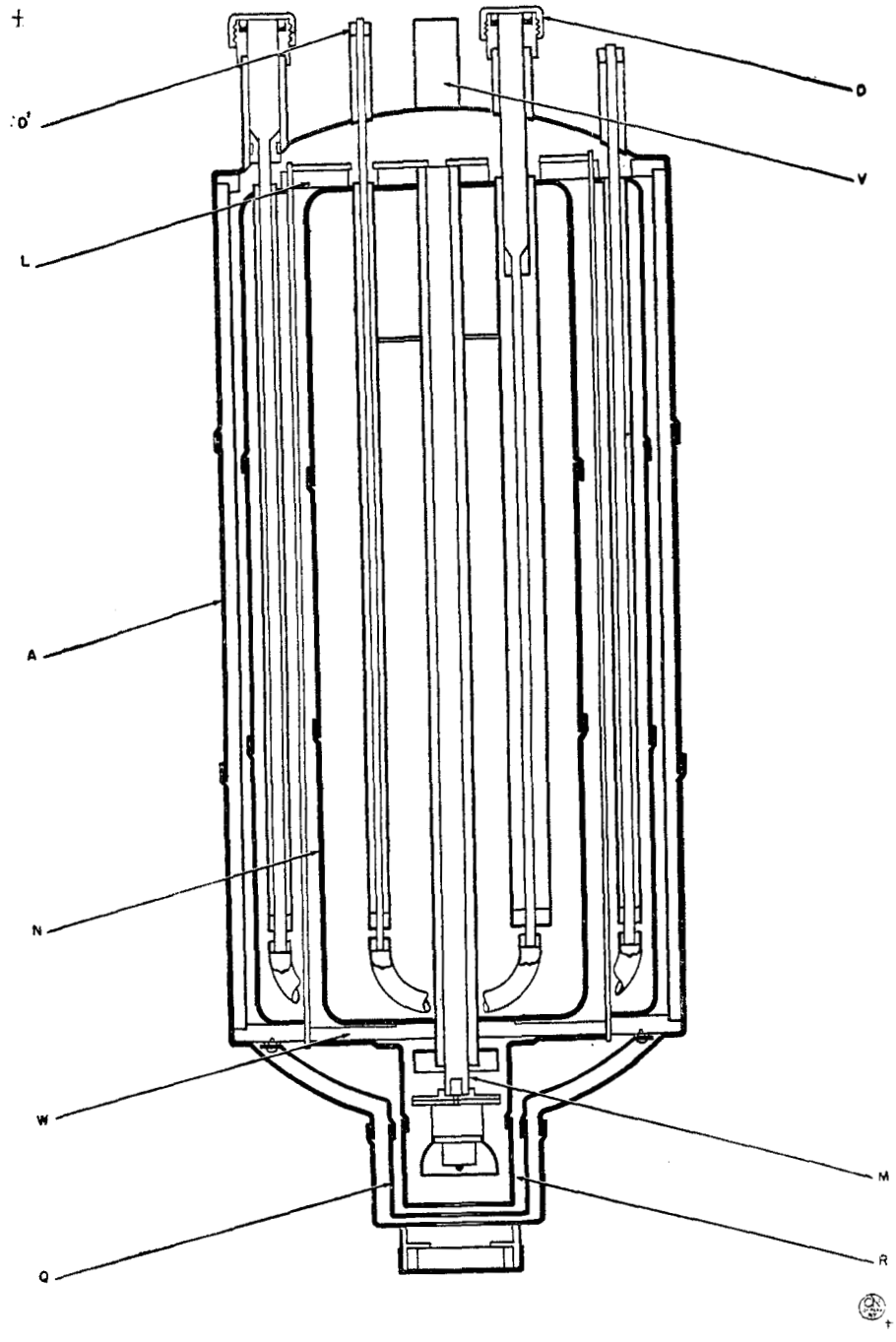

FIG. 9

will have a temperature of 14°K., and the bolometer can be raised to its proper operating temperature, usually about 14.5°K., by means of the current passed through the constantan coil. When the current through the heating coil is set at the proper value, the bolometer will remain in its sensitive state as long as the supply of hydrogen lasts in the can, with perhaps an occasional adjustment of temperature every 5 hr. or so.

The can is filled with liquid hydrogen through the tube D, which is a supernickel tube having an outside diameter of $\frac{3}{16}$ in. and a wall thickness of 0.010 in. The outer end of the tube is equipped with a screw cap which will clamp down on the rubber washer and grip tightly the glass transfer tube through which the liquid hydrogen passes into the can. A supernickel tube of $\frac{1}{4}$-in. outside diameter and 0.010-in. wall thickness, shown as D′ in the diagram, acts as a vent tube while the can is being filled. It also serves as a tube through which the space above the surface of the liquid hydrogen can be pumped to reduced pressure in order to obtain the triple-point mixture. Surrounding the liquid hydrogen can there is a double-walled can containing liquid nitrogen and having a volume of approximately 400 cc. This can provides a thermal dam which cuts down the heat leak into the liquid hydrogen from either radiation or conduction. It normally contains liquid nitrogen at the boiling point, 77.4°K. The inlet and outlet tubes ($\frac{3}{16}$-in. O.D., 0.010-in. wall thickness) for filling with liquid nitrogen are equipped similarly to the tubes on the hydrogen can.

Within these cans the tubes come up to the top facing each other, as shown in the diagram. Thus, when a can is nearly full of liquid refrigerant, it may be held either horizontally or vertically without spilling. These cans are held in place within the outer casing (A) by means of masonite spacers $\frac{3}{16}$ in. thick, marked L and W. These serve as insulator rings between the hotter and colder parts of the apparatus. It is found that when the whole apparatus is cooled, the occluded gas in the masonite is not a significant factor in damaging the vacuum. The thermal insulation is very good, as indicated by the figures on the heat leak quoted later. On the other hand, the spacers have sufficient mechanical rigidity so that the whole can will withstand vibration equivalent to a strain of 10 g. In order to provide protection from radiation, two shields are provided (Q and R). Above the radiation shields, the top of the cryostat is equipped with a rock salt window through which infrared radiation may be admitted to a bolometer.

Wires to provide the electrical leads to the bolometer and to the bolometer-sensitizing coil wound on the post enter through a vacuum-tight connector located at the bottom of the cryostat but not shown. The connector is made of an ordinary eight-prong metal radio tube, which is cut off and soldered into a hole in the cryostat. A tube (V) is provided for a preliminary evacuation of the cryostat, if desired.

In order to improve the vacuum, two cylindrical charcoal containers are placed in reëntrant tubes in the nitrogen can, so that a relatively high vacuum is produced as soon as the nitrogen can is filled. The action of these containers plus the condensing action of the liquid hydrogen in the hydrogen can is such

that a vacuum can be maintained for several days, even though the evacuating tube (V) is closed off with an ordinary piece of rubber tubing and a pinch clamp. The weight of the cryostat and its over-all dimensions are as follows:

Weight of copper cryostat, empty	10.7 lb.
Maximum diameter	5.2 in.
Over-all length, tip of window frame to end of electrical plug	15 in.
Longest external tube projection (nitrogen inlet and exhaust tube)	1.0 in.
Length of electrical plug	1¾ in.
Weight of liquid nitrogen, 900 cc. (density, 0.80 g./cc.)	1.6 lb.
Weight of liquid hydrogen, 1050 cc. (density, 0.07 g./cc.)	0.16 lb.
Total weight of cryostat when completely filled	12.5 lb.

The heat leak into the hydrogen can, measured by observing the amount of gas which evaporates from it, has a value of approximately 0.3 watt. The heat leak into the nitrogen can is about 3 watts.

V. OPERATION OF COMBINED UNIT

The liquid-nitrogen containers on the liquefier, purifier, and cryostat are first filled. The level of liquid nitrogen in the liquefier is observed by means of a thermocouple. Gaseous hydrogen at approximately 2000 lb. is then admitted to the liquefier. Approximately ½ hr. after the hydrogen has been flowing through the liquid nitrogen, significant liquefaction will begin. This is indicated when the reading of thermocouple T reaches a steady value at the appropriate amount above the value shown by the thermocouples immersed in the liquid nitrogen. It is not economical to continue liquefaction after the pressure falls from 2000 to below 900 lb.

Liquid hydrogen is transferred from the liquefier into the cryostat through a double-walled glass Dewar transfer tube. It is accomplished merely by opening the valve at the top of the liquefier to the proper amount such that the over-pressure in the liquefaction chamber will force the liquid up the siphon and into the cryostat.

If the hydrogen gas should contain impurities which get past the purifier, or if the purifier is not working properly because of a failure to bake out sufficiently, clogging may take place at the main expansion valve. This can be remedied by by-passing some of the room-temperature hydrogen through the valves provided for that purpose, past the expansion valve, thus warming it. As a rule, a few minutes of by-passing hot gas is sufficient to melt and blow out any impurities.

In order to show the length of time involved in the different steps of operation, typical data are presented in table 1.

The units described in this paper were developed over a considerable period by a large group working on a combined project during the years 1942–46. While the authors of this paper had perhaps the greatest experience with the finished

TABLE 1

TIME	REMARKS
hr. min.	
0	Cylinder manifold connected to inlet of purifier unit. One cylinder at 2000 lb. open to purifier; inlet valve to liquefier closed. Began cooling charcoal trap of purifier with liquid nitrogen.
0:05	Charcoal trap cold; opened high-pressure inlet valve V1 to liquefier. Adjusted expansion valve until low-pressure gauge (G3) showed 2 lb. in expansion chamber.
0:06	Began adding liquid nitrogen to reservoir of liquefier.
0:08	E.M.F. of thermocouple T2 steady at 6.2 millivolts.
0:10	E.M.F. of thermocouple T1 steady at 6.2 millivolts. Stopped adding liquid nitrogen. E.M.F. of T3 is now 2.0 millivolts.
0:20	T3 reads 2.5 millivolts.
0:30	T3 reads 5.7 millivolts. Liquefaction beginning. Pressure in single cylinder has now dropped to 700 lb. Three full cylinders connected to purifier unit. Total pressure 1900 lb.
1:00	G1 and G2 = 950 lb. G3 = 4.8 lb. 5-min. transfer.

Note: This was sufficient to cool and completely fill one cryostat.

units, the larger part of the credit for their construction and successful operation should be distributed among those who participated in the earlier stages of the project, not all of whom were able to share in the final successful operation. In particular, acknowledgement is made to Dr. E. R. Blanchard, Dr. W. T. Ziegler, Dr. J. W. Hickman, Dr. C. L. Christ, Mr. William Corak, and especially to members of the engineering staff, Mr. W. R. Asher and Mr. H. W. Bittner.

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(1) AHLBERG, J. E., ESTERMANN, I., AND LUNDBERG, W. O.: Rev. Sci. Instruments **8**, 422 (1937).