

Apparatus for high-pressure, low-temperature, neutron-scattering measurements*

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We describe an apparatus which is designed for neutron-scattering studies of single crystals under hydrostatic pressures up to 5 kilobar and at temperatures between 77 and ~ 300 K. The safety features of the system are emphasized.

I. INTRODUCTION

Much has been learned in recent years about the interatomic forces in solids from the analysis of phonon-dispersion curves obtained in inelastic neutron-scattering experiments. If it is assumed that the forces are derived from an effective two-body potential, then a neutron experiment at a single temperature and pressure yields information about the first and second derivatives of that potential at certain values of the interatomic separation. In order to obtain further information it is necessary to vary the temperature and/or the pressure. Useful information about the potential may also be extracted from neutron experiments on liquids.

Many experiments have been performed to measure changes in phonon spectra with temperature, but the effects of temperature (at constant pressure) are two-fold. The material changes in volume, and the amplitudes of vibration increase. It is highly desirable to separate these effects experimentally by performing (a) constant temperature measurements as a function of volume by applying pressure, and (b) constant volume measurements as a function of temperature by adjusting the applied pressure to counteract the effect of the temperature change. The first type of experiment has been performed on several materials,¹ and the second type of experiment has been performed on neon² and rubidium.³ In this paper we describe the experimental

apparatus which was used in a recent experiment to measure changes in phonon frequencies in Rb as a function of temperature for several constant values of the atomic volume.³

Since Rb melts at 312 K (at atmospheric pressure) the measurements were made at and below room temperature; because hydrostatic pressures of several kilobars were required, helium gas was used to transmit the pressure to the single crystal of Rb. In order to avoid the complication of solidifying the He,⁴ and in order to simplify the experimental design, measurements were restricted to temperatures above the boiling point of liquid nitrogen (77 K). At this temperature He becomes solid under a pressure of about 15 kilobar.⁵

In the following section we describe the main features of the apparatus; an earlier report⁶ contains more detailed information on the components of the system. In Sec. III we discuss the safety features of the system, and the final section contains some conclusions.

II. DESCRIPTION OF THE APPARATUS

In Fig. 1 we show a simplified diagram of the high-pressure generating system. Helium gas from a high-purity bottle (at ~ 150 bar) is first compressed to about 1.4 kilobar using a motor-driven gas compressor. The highest available purity should be used. Furthermore, since the time required to pump up the system depends strongly on the bottle pressure, a high bottle pressure is most desirable. The gas is further compressed to pressures up to 5 kilobar using a gas intensifier⁷ driven by a hand-operated hydraulic pump.

A length of 0.159-cm o.d., 0.018-cm i.d., capillary line connects the high-pressure generating system to the sample vessel. At each end the line is silver soldered to a standard blind plug, drilled to accept the line.⁸ The pressure vessel was designed by Professor W. B. Daniels of the University of Delaware, and will be described elsewhere.⁹ It is fitted with a standard cone fitting to which the capillary line is attached.

The vessel is attached to a copper cold block inside the cryostat (Fig. 2). A 2.5-liter nitrogen tank, stainless steel, communicates with the cold block via a stainless-steel tube. The temperature of the block is regulated by adjusting the mass flow of N through it. This is achieved by controlling the back pressure in the exhaust line using a solenoid valve which is driven by a simple temperature controller. At temperatures close to but below room temperature the block may be heated using two 50-W, 110-V cartridge heaters

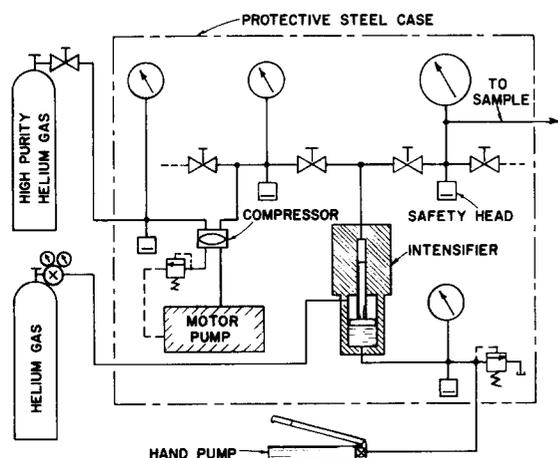
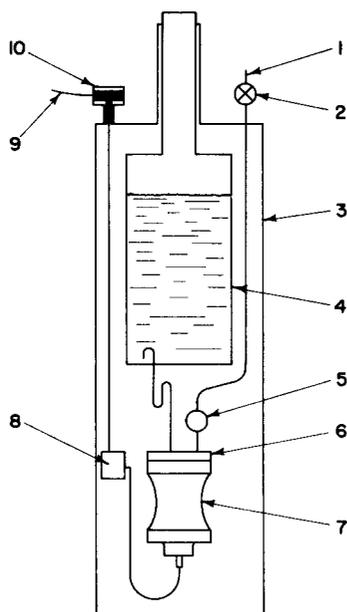


FIG. 1. Schematic of the high-pressure generating system. Only the main components are shown. A detailed description is contained in Ref. 6. The hand pump and the components on the low-pressure side of the intensifier are charged with hydraulic oil. The electric motor pump is also filled with oil. The compressor contains a diaphragm which isolates the oil from the high-purity He gas.

FIG. 2. Variable temperature cryostat. 1—Exhaust line; 2—solenoid valve; 3—vacuum chamber; 4—liquid nitrogen tank; 5—phase separator; 6—copper cold block; 7—pressure vessel; 8—small valve; 9—high-pressure line from generating system; 10—compressed rubber seal.



wired in parallel. The outer jacket of the cryostat is a 57.2 cm length of 6061 aluminum alloy pipe, 15.2 cm in diameter, with 0.7-cm wall thickness. 1.3-cm thick plates are attached at either end. The top plate contains a compressed rubber feedthrough seal for the high-pressure line, heater wires, and thermocouple wires.

In order to isolate the Rb crystal from the atmosphere during assembly of the cryostat, a small valve is placed in the capillary line, about 20 cm from the pressure vessel. This valve is shown in Fig. 3. Port A communicates with the vessel, and port C communicates with the high-pressure generating system. An 0-80 steel screw, ~ 1.3 cm long, with its end machined to a 60° cone, is inserted through port B and screwed into the valve body. When it is fully screwed in, it prevents communication between port A and the other two ports. It thus operates as a valve at pressures up to a few bar. In normal operation the screw is backed off two turns, and port B is closed with a standard high-pressure plug and gland nut.⁶

III. SAFETY CONSIDERATIONS

The high-pressure generating system is largely constructed of manufactured items which are rated for pressures considerably higher than the pressures realized in operation. Each section of the system is protected using a safety head/rupture disk assembly. In addition the hydraulic lines are fitted with relief valves to protect both the system and the pumps from overpressurization. The bulk of the high-pressure system is enclosed in a six-sided box, $60 \times 152 \times 120$ cm high, constructed of 0.63-cm thick mild steel.

The high-purity gas bottle is protected from overpressurization by a check valve and from sudden depressurization by a surge check valve. The high-pressure valves are operated in a remote fashion¹⁰ (Fig. 4). The valve is mounted with its stem facing away from the front panel. The handle is replaced with a spur gear which engages a similar gear on a separate shaft. This shaft penetrates the front plate and is fitted with a handle. By turning the handle

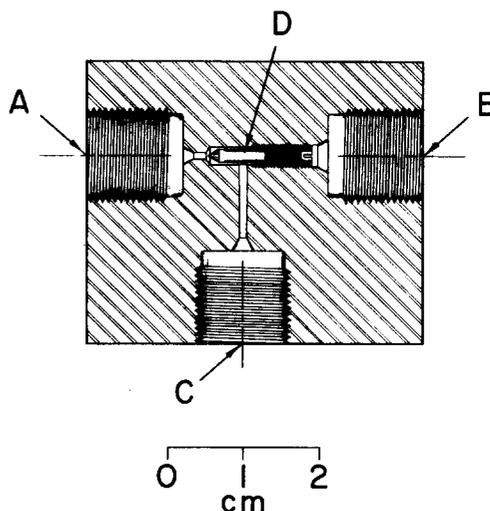


FIG. 3. Small valve. Ports A, B, and C are each tapped to accept a $\frac{1}{4}$ -20 gland nut. The 0-80 steel screw is identified as D.

clockwise, the valve is closed, as is the normal convention. Should a valve stem blow, it will travel away from the front plate, protecting the operator from serious injury. The high-pressure gauges are fitted with solid fronts so that expanding gas will not blow out the front of the case and injure the operator.

Owing to the very small internal diameter of the capillary line, any whipping action, should the line rupture, is easily contained. (The force on the end of the line is about 1.2×10^6 dyn, or about 1.2 kg weight, if the inside pressure is 5 kilobar.) On the other hand the gas jet is potentially dangerous. We chose to enclose the capillary line in thick rubber hose, but we believe this precaution is unnecessary.

The cryostat is fitted with a ~ 0.06 bar popoff valve, which prevents pressurization of the cryostat. If the pressure vessel, under 5 kilobar pressure, were to rupture and release gas into the vacuum space of the cryostat, the final pressure (in the absence of any release mechanism) would be of the order of 1–10 bar. Since most of the gas would have to travel through the long capillary line, which presents a considerable resistance to gas flow, the pressure in the vacuum space would be unlikely to rise above 1 or 2 bar. Furthermore the 15.2-cm diam Al pipe is rated for commercial use at 10 bars.

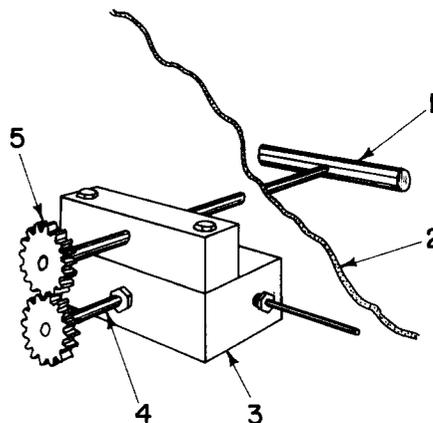


FIG. 4. The remotely operated high-pressure valve. 1—Valve handle, situated outside the protective steel plate; 2—protective steel plate; 3—high-pressure valve body; 4—valve stem; 5—spur gear.

Several pressure tests have been performed to confirm the safety of the apparatus. The capillary line, the small valve, and the pressure vessel were pressurized to >7 kilobar using a 10 kilobar hand-operated oil pump.¹¹ During this test and subsequent tests the vessel was loaded with an Al dummy to minimize the free volume in the vessel. Gas tests were also performed using the high-pressure generating system. High-purity gas was used for the tests as well as during the later experiments, and precautions were taken to maintain the purity of the gas. The gas from the bottle was passed through a line filter, and then through a liquid nitrogen trap. Exhaustive purging procedures were adopted at all times and in addition the intensifier, a potential source of contamination with oil, was fitted with a second He supply attached to the space between the two pistons (Fig. 1). If oil were to contaminate the gas in the capillary line, a block might form at a specific point. Such a block could cause very high pressures to accumulate in the pressure vessel. We tested this possibility by lowering a dummy end piece, which replaced the pressure vessel, under pressure into a large Dewar of liquid nitrogen. In several attempts we were unable to produce any evidence of a blockage.

IV. DISCUSSION

There are indications that more and more neutron-scattering investigations in the future will be conducted under some sort of elevated pressure. Experiments on powders have been performed up to pressures of the order of 30 kilobars using opposed pistons.¹² Investigations of liquids and gases have also been reported at temperatures both below and above room temperature.¹³ Measurements on single crystals yield more information than measurements on polycrystalline materials, but they suffer the additional requirement that hydrostatic pressure must be applied. This is generally possible using a hydraulic fluid, unless the experiment must be made at low temperatures, in which case He gas is the only answer. It is sometimes necessary to go into the region of temperatures and pressures where solid He must be employed.^{4,14}

An important constraint on the design of a high-pressure neutron-scattering experiment is the requirement that the neutrons be able to penetrate the pressure vessel walls. The vessel must be fitted with neutron windows which are sufficiently strong (at all temperatures of interest) but have reasonable transmission. In the case of isotropic samples it is sometimes possible to make the measurements at just one or two scattering angles,¹² but in single-crystal studies it is generally necessary for the window to cover a wide range of angles with respect to both the incident and scattered beams. In such cases a 360° window is usually the most sensible

choice. Thus one is immediately limited to more modest pressures than in the case of isotropic samples.

With these thoughts in mind we believe the apparatus described in the present paper is a useful system for studying single crystals at low temperatures and high pressures using neutron-scattering techniques. The high-pressure generating station is very largely independent of the particular experiment, whereas the design of the pressure vessel is determined by several considerations, including temperature range, pressure range, and the nature of the sample to be examined. Wherever possible we would recommend the use of thinner tubing (e.g., 0.08-cm o.d., 0.015-cm i.d. tubing) to connect the pressure vessel to the generating station (assuming this is considered necessary), because it is often advantageous to make measurements on a triple-axis spectrometer with variable incident energy, in which case the sample crystal will not only rotate but translate during the course of the experiment.

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⁸The method of silver soldering a capillary line to a blind plug is due to Professor W. B. Daniels, who has used this procedure for the past 11 years in his own laboratory.

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