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A design of simple clamp type pressure apparatus utilized for measurements of magnetic susceptibility and electrical resistivity at low temperatures is presented. The cell consists of WC piston and Be–Cu cylinder which was autofrettage processed, and sample cavity consists of a teflon bucket and an electrode plug. In a temperature range from 300 K down to 77 K, pressure was determined by manganin gauge which was calibrated by Bi I→II transition pressure at room temperature and also the temperature dependence of pressure coefficient of manganin resistance was taken into account. As a result, the cell was capable of generating hydrostatic pressures up to 30 kbar at room temperature and at least up to 25 kbar at 4.2 K.

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INTRODUCTION

The investigation of the magnetic properties at high pressure and low temperature is undoubtedly one of the important projects for the basic understanding of magnetism. Along the lines of this adjudication, we reported the effect of pressure on the crystallographic as well as magnetic phase transitions of some ferromagnetic alloys and compounds in a temperature range down to 4.2 K under hydrostatic pressures up to 8 kbar.[1,2] There, both the piston and cylinder in a clamp type pressure cell were made of Be–Cu, and the cylinder was used within an elastic limit.

However, there should remain many phenomena, which will be uncovered and understood magnetically when the measurements are made under much higher hydrostatic pressures. Recently, we have designed a simple clamp cell which consists of the WC piston and the autofrettage processed Be–Cu cylinder. A teflon bucket, which has been generally used in the high pressure and low temperature measurements,[3,4,5] was employed for a sample cavity in the cell. In the present paper, the details of the design of the cell and the determination of pressure are presented. The apparatus has been practically utilized for the measurements of magnetic susceptibility and of electrical resistivity at low temperatures.

I. PRESSURE CELL, SAMPLE CAVITY, AND MEASURING SYSTEM

Pressure cell. The pressure generating system, the pressure cell, consists of an assembly of shaft, piston and cylinder. They are made of hardened (HR_C 57) alloy steel (JIS SUJ-2), WC (GTi 10, MITSUBISHI METAL Ltd.), and hardened (HR_C 44) Be–Cu (BeA-25, NGK INSULATORS Co., Ltd.), respectively. The hardening was made in our laboratory in accordance with prescription. Figure 1 shows a cross section of the pressure cell. The dimensions of cylinder are 35, 27 and 6 mm, in length, o.d. and i.d., respectively. The assembly is inserted inside the holder (JIS SCM-4) together with the WC backing plate bound with a Be-Cu binding ring. The holder in our device also serves as a protector against the pressure cell. The cylinder was not press fitted through the holder, so that it could be removed. Instead, it was tightened with brass supporting nut and polyester tape was tightly wound over both the bottom part of the cylinder and the binding ring. A thermocouple was attached to the cylinder installed through the hole in the holder as indicated in the left-hand wall of the holder in Fig. 1.

For the purpose of elevating the working pressure, we tried to perform hydraulic autofrettage process to the cylinder during actual pressurizations. When the cylinder was subjected to maximum pressure of 30 kbar the expansion inside the cylinder hole, over which the pressure is actually exerted, was measured. The amount of expansion was about 3% of i.d. Then it was confirmed that further expansion was not found in subsequent pressurizations, indicating the accomplishment of the autofrettage. As a result, it has been found that a maximum working pressure could be elevated constantly up to 30 kbar without any trouble. On the other hand, when the Be–Cu cylinder has been used as an elastic cylinder without autofrettage process, the maximum working pressure would be reduced to about 10 kbar. For the measurements at lower temperatures, the whole assembly has been transferred and placed in the thermal insulating chamber.

Sample cavity. In Fig. 2(a) are shown the details of a sample cavity, which consists of a teflon bucket, 1 mm in wall thickness, and a Be–Cu electrode plug. The seal of the bucket against the piston or the plug is provided by unhardened Be–Cu sealing rings with triangle cross section. Varnish (GE 7031) was applied to the part of bucket in Fig. 2(a) to which the plug touches. The cylinder was completed with fine finishing so as to fit the piston inside the cylinder hole without any play. The seal of the electrode plug was provided as follows by means of single cone sealing. A cone is of stainless steel and the cone angle is 60°. The plug has a hole, 1.4 mm in diameter, and two 60° cone openings at both

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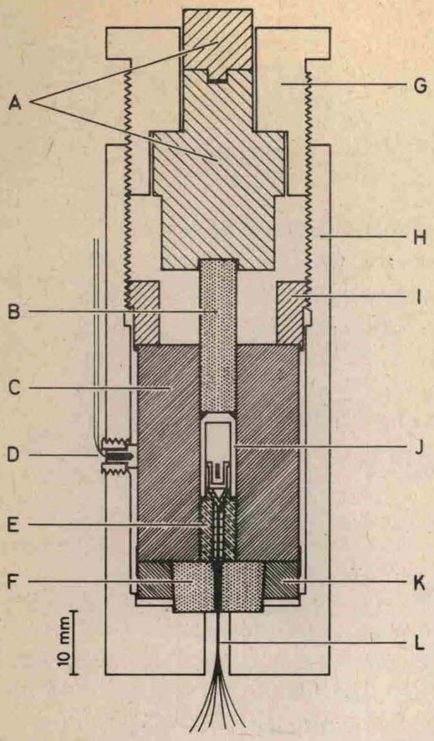

FIG. 1. Cross section of pressure cell. A: Shaft, B: Piston, C: Cylinder, D: Thermocouple, E: Electrode plug, F: Backing plate, G: Lock nut, H: Holder, I: Supporting ring, J: Teflon bucket, K: Binding ring, L: Leads.

the upper and lower ends of the hole, where the upper opening is much deeper. Hereafter, the expression like upper or lower will simply be used concerning the configuration drawn in the figure.

The embedments of the Formvar-insulated copper wires 0.14 mm in diameter, and of the cone into the plug hole were completed with epoxy resin (STYCAST 2850 GT, Emerson & Cuming Inc.), in the following order. (i) The epoxy resin is degassed in vacuum for a minute. (ii) After passing the lead wires through the plug hole, the fused resin is poured into the plug hole from the upper opening. The steel cone is plugged at this step and the resin is cured. (iii) The fused resin is poured from the lower opening, so as not to fill up to the opening. Care must be taken so that the lead wires should not touch both plug and cone. Therefore, the pour was made very carefully by holding the lead wires taut.

Measuring system. The measuring system of the electrical resistance is as follows. A bakelite bobbin was on the top surface of the cone as in Fig. 2(a) and a rectangular or cylindrical sample with lead wires was placed in the bobbin. A manganin gauge wire, referred to as manganin hereafter, for the pressure determination was loosely wound on the bobbin and is connected in series to the current leads of the sample. The resistance has been measured by a conventional dc four-leads method and the total number of lead wires embedded through the plug hole, therefore, was six. The configuration of the coil system for the measurement of the susceptibility is illustrated in Fig. 2(b). The primary coil, 40 turns, was made by winding the Formvar copper wire, 0.06 mm in diameter, on a bobbin. The bobbin in Fig. 2(a) has the same size. A search coil consists of two coils, 10 turns each and with Formvar wire of the same diameter, connected in series but in opposite sense. They were wound on another bobbin which is capable of inserting firmly into the primary coil bobbin. The manganin was wound noninductively and placed around the primary bobbin.

II. PRESSURE CALIBRATION

Among standard fluid transmitting media, referred to as medium hereafter, spindle oil, isopropanol[6] and a mixture of 1 : 1 n pentane and isoamyl alcohol[7] were adopted in the first place. Spindle oil was adopted, since it is relatively viscous so that the possible leak in the initial stage of pressurization could be restrained.

For the purpose of examining the solidifying process of the medium under pressure, the manganin resistance was measured at room temperature. Figure 3 shows the relative change in resistance $\Delta R/R$ of the manganin at room temperature as a function of load to the piston, where the reference was taken at normal pressure. The manganin wire used was obtained from AKABANE YAKIN Ltd., and of 0.1 mm in diameter. The data presented in the figure are those in up-load stroke and the load was applied using a hydraulic press. In Fig. 3, the circles and squares are the experimental points of isopropanol and a mixture of 1 : 1 n pentane and isoamyl alcohol, respectively, and triangles are of spindle oil. As might be expected, the result for the spindle oil shows the indication of solidification at 13 kbar and the oil was not used in practice. Since the results for different runs

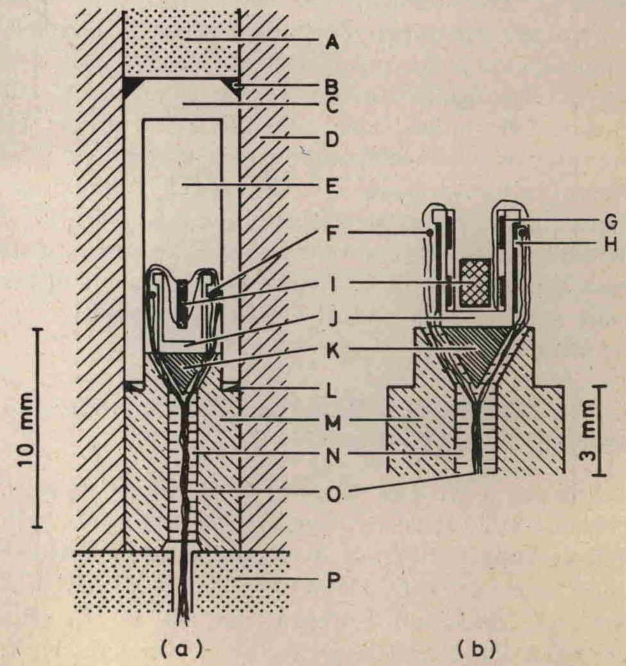

FIG. 2. (a) Cross section of sample cavity for electrical measurement. A: Piston, B: Sealing ring, C: Teflon bucket, D: Cylinder, E: Pressure transmitting liquid, F: Manganin gauge, I: Specimen, J: Coil bobbin, K: Cone, L: Sealing ring, M: Plug shaft, N: Plug hole, O: Lead wires, P: Backing plate. (b) Coil assembly for magnetic measurement. G: Secondary coil, H: Primary coil.

were numerically the same for all the media concerned, the reproducibility of the pressurization was regarded as satisfactory. Furthermore, the corrections due to frictions caused by the various sources could be considered to be constant during different runs.

In the first place, the manganin resistance was calibrated at room temperature using the Bi I → II transition pressure. In the measurement of the resistance of Bi as a function of the change in resistance of the manganin, the transitions of Bi I → II (25.4 kbar) and II → III (27.0 kbar) were clearly observed with the same characteristics as have been observed in general. Since the resistance of the manganin at room temperature had been confirmed to vary linearly with pressure in a pressure range presently concerned,[8] the result at Bi I → II transition was sufficient for the correct determination of the pressure coefficient of the manganin resistance at room temperature. The mean value thus determined was 0.24%/kbar, while the results are 0.23%/kbar by Nomura *et al.*,[8] 0.238%/kbar by Zeto and Vanfleet[9] and 0.23%–0.24%/kbar by Yamamoto.[10]

The pressures at lower temperatures except near 4.2 K were determined by the manganin resistance. For that purpose, the temperature dependence of the manganin resistance was observed first from 300 down to 77 K. Figure 4(a) exemplifies the results under different starting pressures at 300 K. The curves A, B and C are under pressures and curve D is at atmospheric pressure p = 0 kbar. The medium was a mixture of 1 : 1 n pentane and isoamyl alcohol. The cooling of the apparatus was made very slowly with every possible care.[11] All curves in Fig. 4(a) vary monotonically, except small anomalies locating between 150 and 200 K in curves A, B and C.

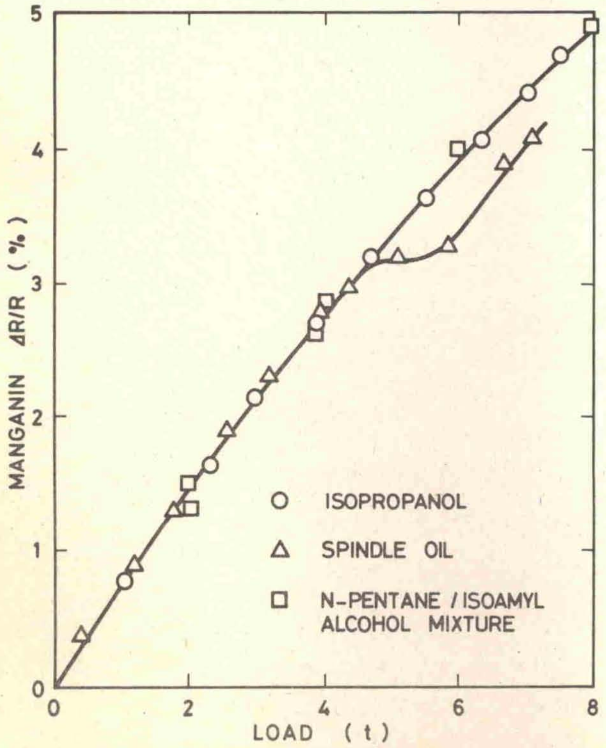

FIG. 3. Relative change in resistance $\Delta R/R$ of manganin wire as a function of load. Measurement was made at room temperature and the reference resistance was taken at room temperature.

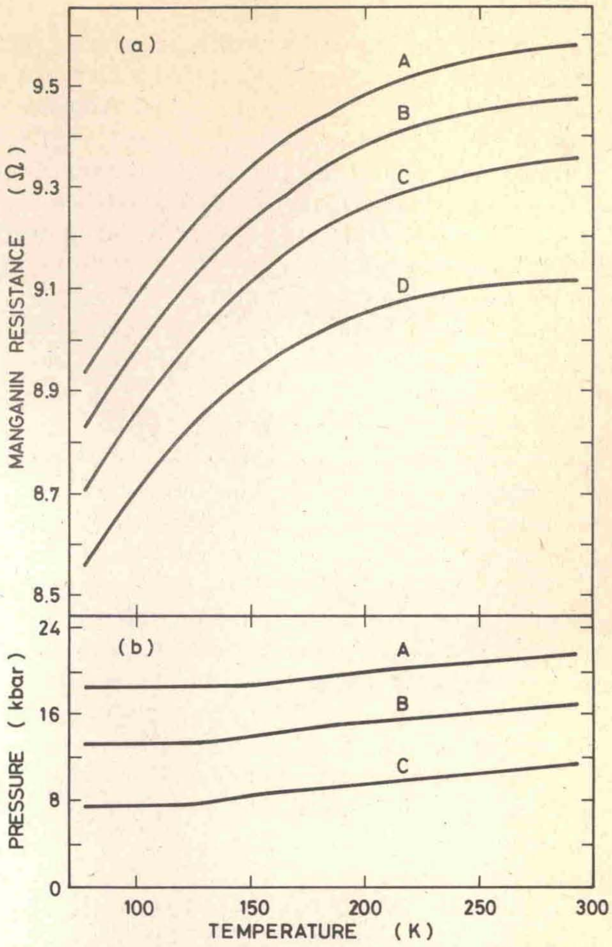

FIG. 4. (a) Temperature dependence of manganin resistance from 300 K down to 77 K. Curves A, B and C are under high pressures and D at atmospheric pressure. (b) Pressure drop as a function of temperature. Different curves correspond to different initial pressures at room temperature. See Fig. 4(a).

In order to determine the pressure, the temperature dependence of the pressure coefficient should be known. Yamamoto[10] has made thorough investigations under various conditions of manganin wires from different manufacturers. The ranges he measured were 123 K to 373 K in temperature and hydrostatic pressures up to 10 kbar. On the other hand, Itskevich[11] has measured the resistance at 20.4, 77 K and at room temperature up to 7 kbar. According to their results, the pressure coefficient of manganin resistance might be regarded as constant independently of temperature between 77 and 300 K, with a difference of ±2%. In the present work, therefore, the pressure coefficient at room temperature, 0.24%/kbar, for the manganin wire presently employed was used as a coefficient for all temperatures down to 77 K.

The real pressure in the sample cavity at any given temperature was then evaluated from the temperature dependence of the relative change in resistance shown in Fig. 4(a), using the value of the constant pressure coefficient given above. The results obtained down to

77 K are shown in Fig. 4(b) for different starting pressures at 300 K. The curves A, B and C correspond respectively to those in Fig. 4(a). The tendency of change in pressure in the figure is arranged as follows. (i) The pressure reduces linearly as temperature lowers, (ii) the reduction becomes relatively highpitched in the intermediate temperature range, possibly due to the onset of freezing, and (iii) the pressure turns to become almost constant after the completion of freezing. The reduction rates in step (i) were almost the same for exemplified cases A, B and C, and the mean value was −0.015 kbar/deg.

From various runs with different initial starting pressures at room temperature, which could be predetermined from the load pressure using the results in Fig. 3, it has been found that the maximum pressure drop, that is the difference between the initial and almost constant final values, was about 4 kbar for 1 : 1 n pentane and isoamyl alcohol mixture, so far as the initial pressures were higher or around those quoted in Fig. 4(b). As a result, it has been confirmed that the pressure cell presently designed is capable of generating pressures at least up to about 25 kbar at lower temperature like 4.2 K. Here it should be mentioned that the data on the temperature dependence of manganin resistance such as exemplified in Fig. 4(a) were completely reversible and reproducible. Therefore, together with the experimental fact mentioned above that the reduction in pressure was not practically found after freezing, it might safely be concluded that the pressure was almost hydrostatic through and after freezing.

Although the extrapolated pressure of the curve in Fig. 4(b) could be used near 4.2 K, the pressure was also determined by the use of a superconducting manometer.

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