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Development of two-stage multi-anvil apparatus for low-temperature measurements

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The two-stage 6-8 multi-anvil (MA8) apparatus is an important large-volume, high-pressure technique that has been widely used in the high pressure mineralogy and material synthesis, mainly at room temperature or above. Recently, we have successfully developed a two-stage MA8 apparatus for low-temperature physical property measurements. The first-stage anvils at top and bottom sides are fabricated as a single piece in order to reduce the total size of the cylindrical module, which is put in a top-loading high pressure cryostat and compressed by a 1000 ton hydraulic press. A castable, split octahedral gasket with integrated fin was specifically designed in order to introduce the electrical leads from the inside sample container filled with a liquid pressure transmitting medium. By using tungsten carbide (WC) second-stage cubes with a truncated edge length of 3 mm and an octahedral gasket with an edge length of 6 mm, we have successfully generated pressure over 20 GPa at room temperature. Since the high pressure limit can be pushed to nearly 100 GPa by using the sintered diamond second-stage cubes, our MA8 apparatus has a great potential to expand the current pressure capacity for precise low-temperature measurements with a large sample volume.

I. Introduction

Pressure is a fundamental parameter like temperature that governs the states of matter. The application of high pressure can induce structural or electronic phase transitions or precisely tune the structural and physical properties. In condensed matter physics, the combination of high-pressure and low-temperature environments provides a very fertile ground for exploring novel quantum states of matter and exotic phenomena. For example, pres-

sure can induce a magnetic quantum critical point, near which the Landau Fermi-liquid behavior usually breaks down and unconventional superconductivity frequently takes place due to the presence of strong quantum fluctuations. Therefore, it is important to develop a high pressure apparatus for low-temperature measurements.

Despite the sophisticated low-temperature technologies existent, the high-pressure devices used in low-temperature conditions remain to be further developed due to the space constrain and other specific requirements, such as pressure homogeneity, sample volume, etc. Currently, piston-cylinder cell (PCC) [1,2] and diamond anvil cell (DAC) [3,4] are two widely used commercial high-pressure devices for in-situ physical property measurements at low temperatures. PCC offers a large sample space and relatively good hydrostaticity by employing a liquid

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pressure transmitting medium (PTM) [5], but the maximum pressure is usually limited to 4 GPa [1], which is insufficient for many studies in condensed matter. Although the DAC [3] can achieve ultra-high pressures and allow easy access for the electromagnetic radiations, the tiny sample space makes it difficult for in-situ physical property measurements requiring electrical contacts, and the solid PTM usually employed renders severe non-hydrostatic pressure conditions.

Besides the PTM, the level of pressure hydrostaticity/homogeneity also depends on the compression geometry. In comparison with DAC, multianvil-type (MA) apparatus can maintain better pressure homogeneity even if the PTM becomes solidified at low temperature and/or high pressure [6]. In addition, the MA apparatus can reach pressure above 10 GPa, much higher than PCC. The single-stage cubic anvil cell (CAC) device developed in the Institute for Solid State Physics, the University of Tokyo (ISSP, UT) [7] is one typical MA apparatus that can generate hydrostatic pressures up to 15 GPa. The design of miniature “palm”-type CAC also enabled integration with ^3He or dilution refrigerator so as to reach temperatures as low as 10 mK [8,9]. These developments of cubic-type apparatus were essential for us to discover novel quantum phenomena [10] and new superconducting materials [11] recent years.

To pursue more exotic phenomena in an extended pressure range, there is always a demand for the development of devices reaching even higher pressures. In this regard, the two-stage 6-8 multi-anvil (MA8) apparatus originally developed in 1970s by Kawai and Endo becomes an excellent option [12]. In this case, the first stage of six anvils surrounds a cubic cavity, in which it is placed the second stage, consisting of eight cubes with truncated corners forming an octahedron. After 40 years of developments, the MA8 apparatus has gain great success and has been widely used in high-pressure mineralogy and synthesis of materials. Depending on the strength of the second-stage anvils, the maximum pressure of MA8 are used for high-pressure studies at or above room temperature. In this paper, we report the development of a two-stage MA8 apparatus for precise low-temperature physical property measurements in ISSP, UT.

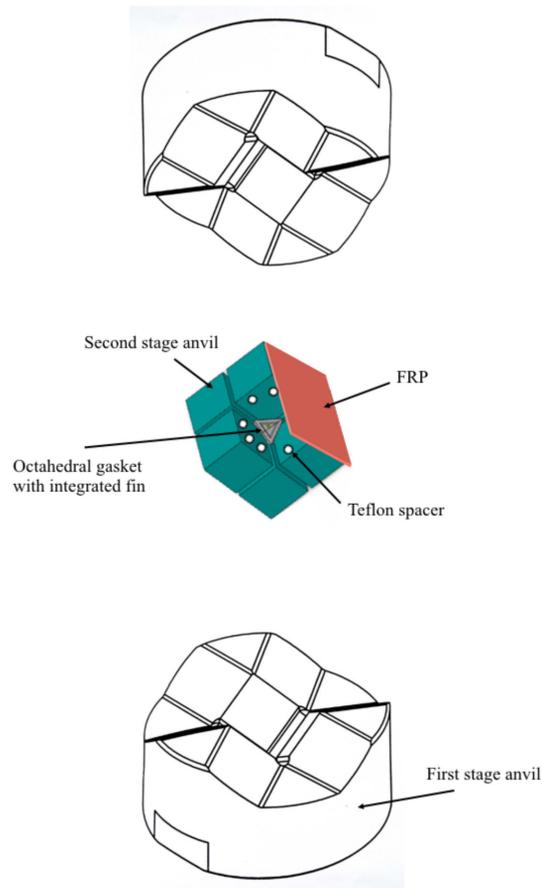


Figure 1: Schematic illustration of the first-stage and second-stage anvils.

II. Experimental setup and results

i. Two-stage MA8 device

For the commonly used two-stage MA8 apparatus, the first-stage six anvils (three on the top and three on the bottom) made of hardened steel are usually built into a thick-wall steel ring (Kawai type) or contained in a removable cylindrical module (Walker type) [13]. Such designs are not suitable for low-temperature applications because the whole MA8 device has to be inserted into a cryostat. To reduce the total size of the MA8 device, we designed the first-stage three anvils on top and bottom sides as a whole piece, as shown in Fig. 1. We have also used a nonmagnetic NiCrAl alloy to fab-

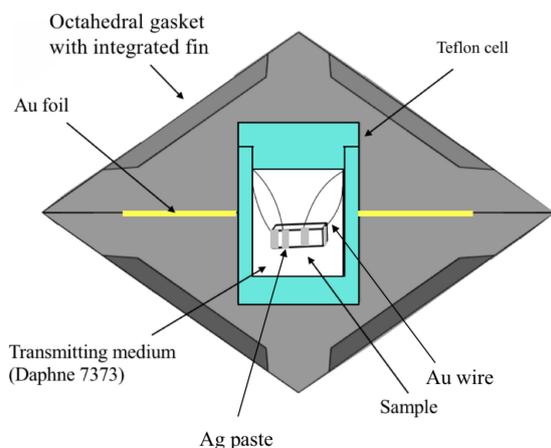


Figure 2: Cross-sectional view of the internal configuration of the gasket with teflon cell.

ricate the pair of cylindrical first-stage anvils in order to apply magnetic fields. The first-stage anvils have an outer diameter of 154 mm and form a cubic cavity with edge length of 32.3 mm. The second-stage anvils, consisting of eight cubes with truncated corners, are similar to the commonly used MA8 apparatus. Here, we employed nonmagnetic WC (TMS05/MF10 grade from Fujilloy) with an edge length of 18 mm and truncated corner of 3 mm. As a common practice, these WC cubes are held together with six pieces of Fiber-Reinforced Plastics (FRP) pads, which are 0.5 mm in thickness and 36×36 mm in area. These FRP pads also serve as an insulation to the first-stage anvils. The inside surfaces of the second-stage cubes are pasted with three 1.0 mm cubic Teflon spacers to prevent electrical contact with adjacent anvils.

ii. Gasket design and sample assembly

The adoption of a liquid PTM is essential to maintain a relatively good pressure homogeneity. However, the conventional design of octahedral gasket and sample assembly used for the MA8 apparatus also need to be modified in order to accommodate a sample container filled with liquid PTM. For this purpose, we adopt the castable, split octahedral gasket with integrated fin, which are made from Ceramacast 584-P and Ceramacast 584-L (100:28

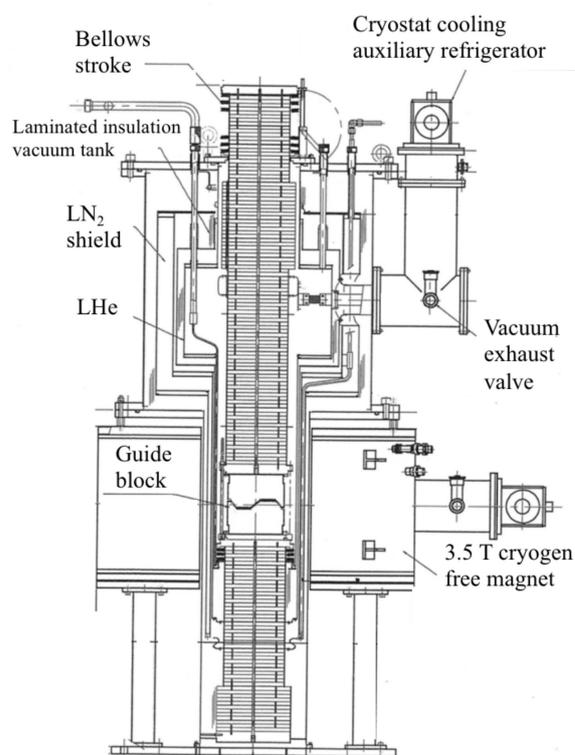


Figure 3: Cross-sectional view of the top-loading cryostat.

weight-in-weight) potting compound from Aremco Products, Inc. The half-octahedral gaskets with integrated fins are made in-house in our laboratory according to the procedures described in Ref. [14]. The edge length of the octahedron is 6 mm and the thickness of the gasket fin is 1 mm. Figure 2 depicts the internal configuration of the gasket with the sample hanging inside the Teflon capsule (I.D. 1.5 mm, O.D. 2.0 mm and length 2.5 mm), which is the same setup used in the cubic anvil cell [7]. The Teflon cell can be filled with a liquid PTM such as Daphne 7373 or Glycerol, and the electrical leads are introduced via gold foil to the surfaces of octahedral gasket, which in turn contact with the WC cubes.

iii. Top loading high-pressure cryostat

Figure 3 shows a schematic cross-sectional view of the top-loading high-pressure cryostat, in which

Table 1: Phase transitions as pressure calibrants [15].

Sample	Pressure (GPa)
Bi	2.55, 2.7, 7.7
Sn	9.4
Pb	13.4
ZnS	15.6
GaAs	18.3

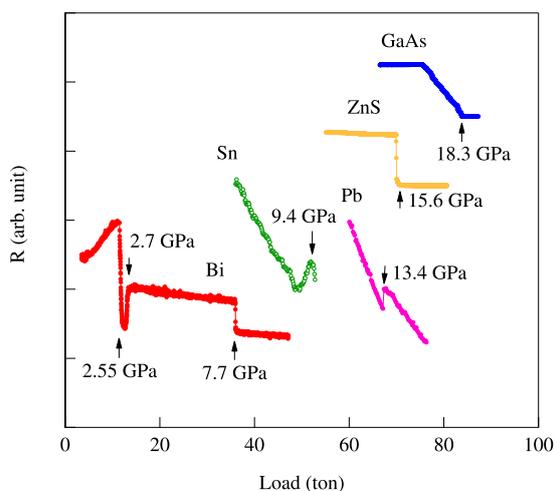


Figure 4: Electrical Resistance of Bi, Sn, Pb, ZnS and GaAs as a function of loading force.

the MA8 device is placed in between the upper and lower pushing columns. Details about the design of the high-pressure cryostat can be found in an earlier publication about the cubic anvil cell apparatus [7]. The low-temperature condition (down to 2 K) is realized by filling the cryostat with liquid nitrogen and then helium with proper pumping. Precise temperature control between 2 and 300 K was achieved by attaching a resistance heater onto the MA8 device. The pressure is generated by using a 1000-ton hydraulic press, which can maintain a constant loading force over the MA8 device over the whole temperature range. In addition, a 3.5 Tesla helium-free superconducting magnet with a large bore size is also installed and the center of the magnetic field is aligned with the sample in the MA8 device.

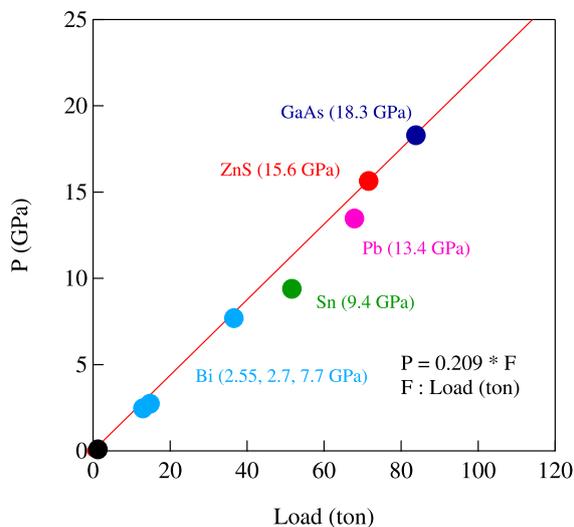


Figure 5: Pressure calibration line for a two-stage multi-anvil high pressure cell.

iv. Pressure calibration

We have performed fixed point pressure calibration at room temperature by detecting the characteristic phase transitions of Bi, Sn, Pb, ZnS and GaAs in electrical resistance. A standard four-probe method was used to measure the resistance of each sample. Table 1 summarizes the transition pressure of these materials from previous studies [15].

Figure 4 shows the electrical resistance of Bi, Sn, Pb, ZnS and GaAs as a function of loading force at room temperature. As can be seen, the characteristic phase transitions of Bi at 2.55, 2.7 and 7.7 GPa were clearly observed at loading force of 12.2, 13.7, and 36.2 tons, respectively. We defined the phase transitions which are the offset. Similarly, the resistance anomalies of Sn and Pb at 9.4 and 13.4 GPa were also observed at 51.7 and 67.9 tons, respectively. In addition, the metallization of ZnS and GaAs at 15.6 and 18.3 GPa were successfully observed at a loading force of 71.1 and 83.9 tons, respectively. Although the employed Daphne 7373 PTM becomes solid at about 2.3 GPa, these characteristic phase transitions remain very sharp, signaling an excellent pressure homogeneity up to at least 20 GPa due to the multi-anvil geometry.

Based on these measurements, we have plotted

in Fig. 5 the pressure calibration curve for our two-stage MA8 apparatus installed with WC cubes having a truncated corner of 3 mm. As can be seen, all the calibration points fall nicely on a linear curve described by P (GPa) = $0.209 \times$ Force (ton). From the extrapolation, we can reach about 25 GPa at a loading force of 120 tons, which is a much lower force than those reported in the literature employing MgO octahedron plus extra pyrophyllite gaskets. In the latter case, a large portion of loading force was dissipated on the relatively soft pyrophyllite gasket so that the calibration curve usually tends to saturate at higher loading forces. In contrast, the much improved pressure efficiency in our MA8 apparatus should be attributed to the octahedral gasket with integrated fin, which is much harder than pyrophyllite. As mentioned above, the maximum pressure at which MA8 can be pushed to is over 40 GPa by using a tapered second-stage WC anvils [16], or to nearly 100 GPa by employing much harder sintered diamond cubes [17]. It can be thus foreseen that the pressure capacity of our MA8 apparatus can be further improved.

III. Conclusions

We have successfully developed a two-stage 6-8 multi-anvil apparatus for accurate high-pressure and low-temperature measurements. By using tungsten carbide second-stage cubes with truncated corners of 3 mm and castable octahedral gasket with an edge length of 6 mm, we can generate pressures over 20 GPa at a relatively low loading force of 100 ton. An excellent pressure homogeneity/hydrostaticity up to 20 GPa has been demonstrated in our MA8 apparatus, which is expected to reach even higher pressures by employing WC anvils with smaller truncation sizes or sintered diamond anvils.

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