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A high-temperature environmental chamber for nuclear-resonant Bragg scattering studies

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A compact environmental chamber with a furnace operating in the range from room temperature to about 900 K has been built for a high-temperature nuclear-resonant Bragg scattering study. The compact size (75 mm diameter and 70 mm depth) allows an external magnetic field to be applied from outside permanent Nd–Fe–B alloy magnets; a magnetic field up to 1000 G has been obtained. The chamber can be mounted on a precise θ–2θ goniometer so that *in situ* observations of X-ray diffraction from a single crystal can be carried out. The temperature is measured by two thermocouples placed at the top and back of the sample and controlled by a PID controller. The temperature fluctuation of the chamber is less than ± 1 K.

Keywords: high-temperature chambers; magnetic field apparatus; nuclear-resonant Bragg scattering.

1. Introduction

Nuclear-resonant Bragg scattering (NBS) studies using synchrotron radiation have been carried out intensively over the past 20 years. As third-generation synchrotron light sources have been, and continue to be, commissioned, not only the conventional Mössbauer spectral studies but also new studies can be performed. Among these is the study of *in situ* NBS under versatile environmental conditions. The temperature dependence of the Lamb–Mössbauer factor was determined from α-^{57}Fe over the temperature range 9.7–1048 K by investigating its nuclear forward-scattering time evolution (Bergmann *et al.*, 1994). As shown by Zhao (1995), from the point of view of NBS, the small value of the Lamb–Mössbauer factor means that the crystal lattice contains point defects in a single crystal. The study of the diffraction phenomena from such a diffracting media with varied isotopic concentration attracted particular interest in our research. In order to study NBS of single crystals from room temperature up to high temperatures, the construction of a high-temperature chamber is required.

Several kinds of high-temperature furnace have been built for X-ray studies of crystal diffraction. For obtaining extremely high temperatures above 1400 K, gas-flame furnaces (Nukui *et al.*, 1972; Miyata *et al.*, 1979; Yamanaka *et al.*, 1981) have been used. For temperatures below 1400 K, radiative furnaces have generally been used because their simple design allows them to be built and handled easily (Kume & Kato, 1974; Hazen & Finger, 1982; Swanson & Prewitt, 1986; Jiang & Zhao, 1992). Our main interest is in the NBS study of almost perfect single crystals; a furnace that can operate from room temperature to about 900 K (below the Neel temperature of hematite, 948 K) is desirable. The construction of a radiative furnace was therefore proposed. Due to the special circumstances of the NBS study of single crystals, a chamber containing a furnace should fulfil the following requirements. (i) The total weight and volume of the chamber should be reduced to such an extent that the whole device can be mounted on a θ–2θ goniometer whose axes are horizontal. (ii) The temperature range should be from room temperature to about 900 K. It should be possible to maintain a stable temperature for several hours during the experiment and the fluctuation of the temperature should be less than several kelvin. (iii) During the experiment, as there are temperature-sensitive optical instruments (*e.g.* high-resolution crystal monochromator) around the chamber, heat from the furnace should be well shielded so that the goniometer itself is not heated and the environment around the chamber is not seriously affected by the furnace. For this reason, the furnace should be put into a vacuum chamber and the body of the chamber be cooled by running water. (iv) An external magnetic field should be applicable from the outside of the chamber. The magnetic field at the sample area (10×10 mm) should be stronger than several hundred Gauss. (v) The chamber should contain X-ray windows to allow the incident X-ray beam to enter and the diffracted beam to exit. The acceptable 2θ angle should be larger than 100°.

A high-temperature environmental chamber has been manufactured based on the above design concept. An outline of the design and the performance of the chamber are described in this paper.

2. Description of the chamber

Figs. 1 and 2 show a general view and the schematic structure of the device, respectively. The device can be separated into three parts: a furnace, a cylindrical stainless-steel vessel (75 mm diameter $\times$ 70 mm depth) and a magnetic field apparatus. The furnace is connected to the vessel by a flange. The vessel can be mounted onto a θ–2θ goniometer. An external magnetic field is applied to the chamber from its outside. In Fig. 2, the sample (*s*) sits in a groove carved on the surface of a copper block (*b*). The size of the groove is either $10 \times 10 \times 0.5$ mm or $5 \times 5 \times 0.5$ mm depending on the size of the sample in the experiment. The sample position is fixed mainly by the edge of the groove. Furthermore, the sample is loosely secured by two fine tungsten

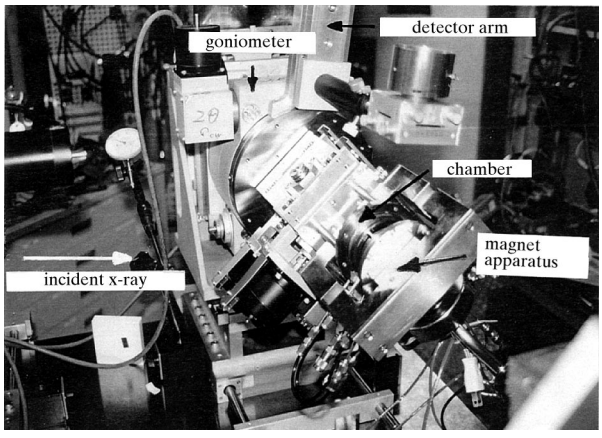

Figure 1
High-temperature environmental chamber with the magnetic field apparatus mounted on a θ–2θ goniometer.

wires which are fixed on the surface of the copper block. Two heaters (*h*) (diameter 6.35 mm and length 24.5 mm) are inserted into two column-shaped holes in the copper block. These heaters are powered by a DC power supply. The temperature of the chamber is monitored by two NiCr–NiSi thermocouples (*T*1 and *T*2); *T*1 is buried in the copper block between the heaters and the sample; *T*2 is located on the sample surface. The copper block is connected to a support (*p*), a stainless steel pipe, by a heat-isolated ceramic spacer (*c*). The support is rigidly connected to a water-cooled vacuum flange (*f*) and serves also as the vacuum evacuation outlet (*v*). The furnace is housed in a stainless-steel vessel (*Ch*) which is evacuated to about 10^{-3} torr to suppress heat loss. The flanges and the body of the chamber are cooled by running water (*w*). A magnetic apparatus made of two permanent magnets (*m*) and a magnetic yoke (*y*) is fixed at the outside of the chamber. The total weight of the chamber (including the magnetic field apparatus) is about 7.5 kg and it can be attached to a precise θ–2θ goniometer (*G*) (Kohzu RA300) with an angular precision of 0.36 arcsec. *k* is an X-ray window made from 25 μm mylar foil with an aluminium coating. The foil is sealed to the chamber by an O-ring covered by a wrapped stainless-steel band. The collectable diffraction angle (2θ) ranges from 0 to about 140° and the width of the window is 10 mm.

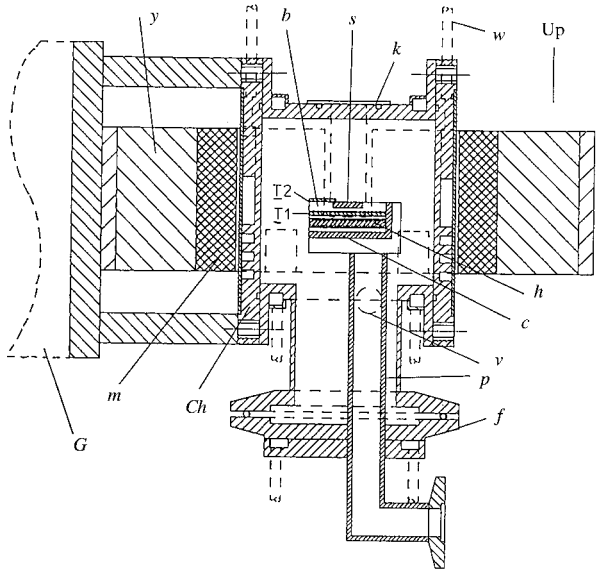

Figure 2
Vertical cross section of the chamber. *b*, copper block; *c*, ceramic spacer; *Ch*, water-cooled stainless-steel chamber; *f*, water-cooled flange; *G*, θ–2θ goniometer; *h*, heater; *k*, X-ray window; *m*, magnets; *p*, heater support; *s*, sample; *T*1 and *T*2, thermocouples; *v*, vacuum outlet; *w*, water pipe (for cooling); *y*, yoke of the magnetic circuit.

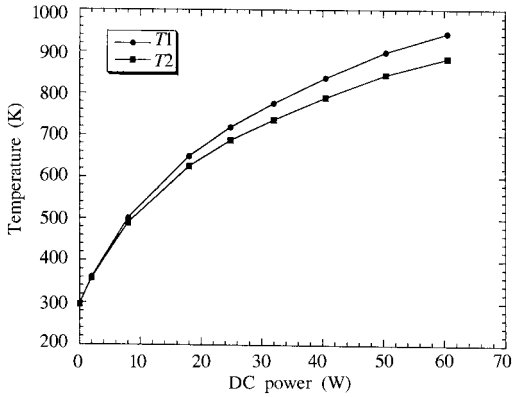

Figure 3
Temperature *versus* DC power supply measured by two thermocouples, *T*1 and *T*2. The positions of *T*1 and *T*2 are described in the text.

3. Performance of the chamber

The furnace is heated by two 150 W heaters (WATLOW Electronic Manufacturing Company) which can work under 1100 K. The heaters are powered by a standard DC power supply. Temperatures up to about 900 K have been obtained at the sample area, which typically needs about 60 W of power. Fig. 3 shows the temperature measured by two thermocouples (*T*1 and *T*2) *versus* the power supply. Owing to the different positions of the heaters, the temperatures measured by *T*1 and *T*2 were different: *T*1 was about 50 K higher than *T*2 at about 900 K. During the experiment the temperature is controlled based on *T*2, which is closer to the sample. The power supply is controlled by a PID controller and the fluctuation of the temperature at the sample area is less than ±1 K during a 4 h experimental run.

In order to reduce the leakage of the magnetic field and make its gradient small at the sample area, a magnetic circuit is constructed by connecting two Nd–Fe–B alloy magnets (40 × 40 × 13 mm) with a magnetic yoke (ss400). The structure of the magnetic circuit is shown in Fig. 4. Due to the compact size of the chamber, a relatively high magnetic field has been obtained. The magnetic field is found to be 1070 G at the centre of the apparatus (point *A*). At *B* and *C*, 10 mm away from the centre, the magnetic field obtained was 1200 and 1010 G, respectively. At the central area of the sample (10 × 10 mm), the field gradient was found to be better than 12% along the magnetic field, and 5% perpendicular to it.

To avoid inducing an external stress for fixing a single-crystal sample, especially under a high temperature, a groove (5 mm square and 0.5 mm deep) is carved at the top of the copper block (Fig. 2) and a sample of almost the same size and thickness sits in it without force. Additionally, two fine tungsten wires fixed on the copper block surface are used to loosely secure the sample. A fine

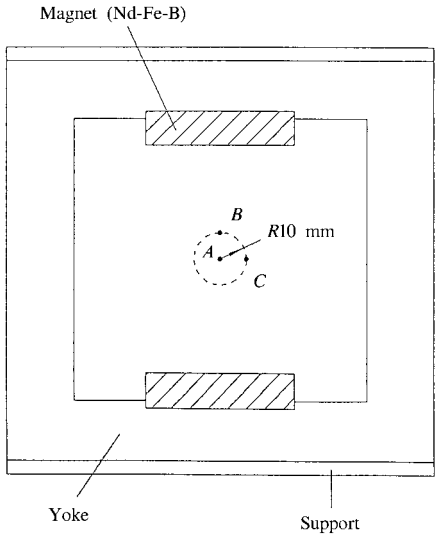

Figure 4
The magnetic field apparatus. The magnetic fields at *A*, *B* and *C* were measured as 1070, 1200 and 1010 G, respectively.

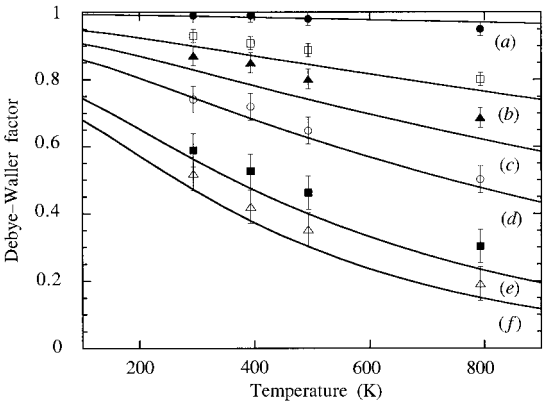

Figure 5
Debye–Waller factor of a silicon single crystal under different temperatures. The theoretical curves (solid lines) are for (*a*) 111, (*b*) 333, (*c*) 444, (*d*) 555, (*e*) 777 and (*f*) 888. Measured data: 111 (filled circles), 333 (open squares), 444 (filled triangles), 555 (open circles), 777 (filled squares) and 888 (open triangles).

θ–2θ goniometer with rotating horizontal axes was used for the experiment. When $\theta = 0^\circ$ the sample surface faced upwards. Hopefully there was no extra stress applied on the sample when θ was small. When θ became larger, but smaller than 90°, the sample was fixed by the edge of the groove and guarded by the tungsten wires. This was almost a stress-free situation.

As a preliminary test of the chamber, the integrated intensities of electronic diffraction of an almost perfect silicon single crystal were measured from room temperature to 900 K. The *hhh* diffractions, with h = 1, 3, 4, 5, 7 and 8, were measured from a silicon (111) wafer. During the experiment, Mo $K\alpha$ radiation was used and Si 111 diffraction was used for monochromation in a double-crystal diffraction set-up. Fig. 5 shows the measured Debye–Waller factor obtained from the measured integrated intensities. A theoretical curve was obtained in the frame of the standard Debye–Waller theory. During this calculation, the Debye temperature of the silicon crystal was set at 532.5 K (Aldred & Hart, 1973). It can be found from Fig. 5 that the measured data is in quite good agreement with the theoretical estimations. It shows that the present chamber could work properly for X-ray studies of single-crystal diffraction under high temperatures.

4. Summary

A compact high-temperature chamber with the temperature ranging from room temperature to 900 K has been built. A magnetic field up to 1000 G can be applied. The chamber has been tested and approved to be suitable for X-ray studies of single-crystal diffraction. It will be used for the study of nuclear-resonant Bragg scattering of single crystals.

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