Apparatus for neutron diffraction measurements on fluids up to 2 000 K and elevated pressures

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Résumé. — Cette publication décrit le principe d'un appareillage pour l'étude de la diffraction des neutrons par des systèmes fluides, à hautes températures et hautes pressions. Comme exemple, les facteurs de structure du rubidium liquide à des températures allant jusqu'à 2 000 K et près de la saturation sont présentés.

Abstract. — The paper describes the principle set-up of a high temperature-high pressure apparatus for neutron diffraction on fluids. As an example the results of the static structure factor of expanded fluid rubidium up to 2 000 K near saturation conditions are briefly presented.

There has been a great deal of interest in the past few years in the study of the interrelation between the metal-nonmetal transition and the gas-liquid critical point phase transition in fluid metals [1-4]. This interest derives not only from the demands for advanced technologies, but also from the challenging physics involved. In spite of it experimental progress is quite slow because experimentation near the critical point of a fluid metal is complicated by the fact that a combination of very high temperatures and relatively high pressures is required. The strong binding energy of the metal places the vapour-liquid critical point at temperatures T_c and pressures p_c that are, by usual standards, very high (e.g. for mercury $T_c = 1\,750$ K, $p_c = 1\,670$ bar and for rubidium $T_c = 2\,090$ K, $p_c = 140$ bar). The highly reactive nature of fluid metals at these high temperatures makes their containment in uncontaminated form very difficult and limits the number of materials suitable for sample containers. It was only through the development of a special experimental high temperature-high pressure technique that a number of experiments have been possible close to the critical point of metals.

It is the purpose of the present paper to give a brief presentation of a newly developed experimental set-up appropriate for neutron diffraction studies of fluid metals at temperatures up to 2 000 K and elevated pressures (see Fig. 1). For a detailed description of the

apparatus see reference [5]. Briefly the fluid metal sample (1) is contained in a thin-walled molybdenum cylinder of 0.2 mm wall-thickness and 28 mm length, which is closed at the top-end (2) by electron beam welding and which is connected *via* a long molybdenum capillary with a liquid reservoir (7) at the bottom end. This sample cell is mounted inside the axis of a high pressure vessel (11), made from an aluminum-alloy of high tensile strength. The pressure on the liquid metal is applied by compressed argon gas which is pressure balanced between the inside and outside of the sample cell by an opening in the liquid reservoir. The high temperatures along the measuring compartment (1) are produced by a tungsten-resistance-furnace. This is made from two concentric tungsten cylinders of 13 and 19 mm diameter, respectively, the wall-thickness of these cylinders being 20 μm . With these dimensions of the heating elements, the internal tubing is the main heater, whereas the outside tubing acts more as a current lead and as a first heat shield. Currents of 100 to 150 A necessary for high temperatures with this heater are led through a high current-high pressure plug (8) at the top end of the autoclave. The temperature along the measuring volume (1) is measured by two W-Re-thermocouples at the top (2) and bottom (3) end of the thin-walled molybdenum cylinder. The temperature profile along the cell is controlled with the aid of a second furnace (5), which

Fig. 1. — High temperature-high pressure apparatus for neutron diffraction studies on fluids at elevated pressures (for a detailed description see text).

is directly wound on the molybdenum capillary. In the height of the neutron beam heat conduction and radiation between the axis and the inside wall of the autoclave is reduced by several heat shields (6) made from 50 μm vanadium foils. With this construction, shown in figure 1, part of the gas convection around the measuring compartment is reduced, too. Thermal and electrical insulations above and below the neutron beam are made by different parts and tubings from boron-nitride (4), (5), (9) and from zirconia (10). The use of boron-nitride — a high neutron absorber — has the further advantages, that scattering contributions from above and below the fluid sample can be completely eliminated. As the high pressure neutron window must be kept at the minimum possible thickness — e.g. for 200 bar maximum pressure a 4 mm thick

window from the aluminium alloy was chosen — the temperature at the window must remain low. This is achieved by an effective water cooling (13), keeping the neutron window at room temperature even for 1 700 $^{\circ}\text{C}$ at the axis of the autoclave. All the electrical feed-throughs (12) for thermocouples, additional heaters, etc. are led through the bottom flange of the autoclave.

A selection of static structure factors $S(Q)$ of liquid Rb for different temperatures and densities obtained by neutron scattering experiments with the apparatus described in figure 1 is shown in figure 2. Two clear changes in $S(Q)$ with decreasing density or increasing temperature are apparent. The intensity of the first peak in $S(Q)$ is strongly reduced and broadened, whereas its position remains approximately constant.

Fig. 2. — Structure factor, $S(Q)$, of expanded fluid rubidium for different temperatures and densities, respectively.

Qualitatively, this reflects the fact that with decreasing density the distance of nearest neighbours does not change very much, whereas the number of nearest neighbours decreases nearly linearly with density. For lower densities or temperatures approaching the critical region ($\Delta T/T_c = T_c - T/T_c < 0.1$) $S(Q)$ is almost smeared out in the range of high wavevectors Q , whereas a strong increase in the small angle scattering range is observed. For a more detailed discussion of these results see references [6, 7].

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