Ultrasonics and X-ray diffraction under pressure in the Paris–Edinburgh cell

D. Lheureux *, F. Decemps, M. Fischer, A. Polian, J.P. Itié, G. Syfosse, A. Zarembowitch

Physique des Milieux Condensés, UMR 7602, 4 place Jussieu Tour 18, 4ème étage B 77, 75252 Paris cedex 05, France

Abstract

Our objective consists in validating a new set-up which will permit us to carry out simultaneously ultrasonic and X-ray diffraction measurements under pressure. To validate the results obtained by this new set-up, the elastic properties of a single crystal of germanium were studied. Our results are in good agreement with those of Goncharova et al. and McSkimin and Andreatch. The results of the present study are compared with those of Menoni et al. and obtained by X-ray diffraction in a diamond anvil cell. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Elastic constants; Equation of state; Germanium; High pressure; Paris–Edinburgh cell; Ultrasonics; X-ray diffraction

1. Introduction

The bulk modulus \(B_0\) and its first pressure derivative \(B'_0\) are fundamental parameters which are commonly determined by ultrasonic studies at low pressure \((P < 1 \text{ GPa})\) with high accuracy. High pressure X-ray diffraction is also used to determine the equation of state \(V(P, T)\). The results of such measurements are then compared with theoretical models, using ultrasonic determination of \(B_0\) and \(B'_0\).

Generally, ultrasonic experiments are carried out independently of X-ray diffraction experiments with a completely different experimental environment: pressure transmitting medium, pressure gauge and hydrostatic conditions are quite different for the respective techniques.

The development of a new pressure device called the 'Paris–Edinburgh cell' makes it possible to carry out simultaneously ultrasonic and X-ray diffraction experiments at much higher pressures. Indeed, samples of millimetric dimensions, which are necessary for ultrasonic measurements, can be compressed in this cell to at least 6 GPa. Moreover, the dimensions and weight of this cell are small enough to enable its easy use for diffraction in synchrotron facility stations, like the DW11 station of the LURE facility (Orsay, France).

The general purpose of this experiment is the simultaneous determination of the equation of state and elastic constants of materials under pressure. From this determination, the volume dependence of the second order elastic constants and Gruneisen parameters can be deduced with high accuracy.

In the present paper, we describe the new experimental set-up. The experimental procedure is validated by studying the pressure dependence of the elastic constants of germanium single crystal up to 6 GPa. This validation is obtained by comparison of the present results with those obtained previously by McSkimin and Andreatch [1] and Goncharova et al. [2].

2. Experiments and calculation

2.1. Samples

In the present experiments, small cylindrical samples were cut from the same piece of germanium. These cylinders (3 or 2.4 mm in diameter and 3 mm in length) were oriented by Laue back reflection X-ray diffraction method with their axis along the [100] and [110] direc-
tions. The parallelism of the two flat opposite faces was better than 1°.

2.2. Experimental set-up

Originally the design of the Paris-Edinburgh cell was developed for high pressure neutron diffraction measurements [3]. The samples are compressed between two opposite toroidal tungsten carbide anvils. The hydraulic press which generates the necessary thrust is connected by a piston to the lower part of the cell.

In order to adapt this cell for ultrasonic measurements, the anvil profiles were modified: the rear face of the upper anvil was flattened and polished in order to stick the ultrasonic transducer. The experimental cell and the sample surroundings are shown in Figs. 1 and 2.

The Ge sample is encased by a boron nitride (BN) cylinder which is used as a pressure transmitting medium. Compressed NaCl powder is located below and around the cylindrical Ge sample. This set-up is put into a boron epoxy gasket (5:1 in mass) which has a low X-ray absorption.

The pressure in the cell is determined by measuring the sodium chloride lattice parameter and by using Decker’s equation of state [4]. The accuracy is ±0.1 GPa (Fig. 3). The uniaxial stress components are evaluated using Singh’s theory [5] and the real hydrostatic pressure is deduced.

Due to the fact that there is no control on the vertical axis of the sample, the Bragg conditions may not be satisfied under pressure, and hence X-ray diffraction measurements are not reliable.

2.3. Ultrasonic measurements

The longitudinal ultrasonic waves are generated by a 15 MHz composite transducer and the transverse waves...
Fig. 3. Transit times with the associated elastic constants.

The pressure dependence of the transit time has been measured up to 5 GPa for $t_1(P)$, 6.3 GPa for $t_2(P)$, and 50 GPa for $t_3(P)$. A linear fit gives an account of this evolution. In order to evaluate the second order elastic constants under pressure, the Cook method [6] has been used. In this way, for cubic crystals, a new parameter $s(P)$ is introduced. It is given by [6]:

$$s(P) = 1 + \frac{1}{4d^2(\rho_0)} \left[ \frac{1}{\rho_1} \frac{1}{t_1} \frac{1}{t_1(P)} \right] - d^2(\rho_0) s(P) = \frac{3}{t_1} - \frac{4}{t_1(P)}$$

where $A$ is a thermodynamic correction between the adiabatic and isothermal elastic moduli [6]. The equation of state can be expressed by the following relation:

$$V(P) = \frac{1}{0} = \frac{1}{s(P)}$$

3. Results

The pressure dependence of the transit time has been measured up to 5 GPa for $t_1(P)$, 6.3 GPa for $t_2(P)$, and 50 GPa for $t_3(P)$. A linear fit gives an account of this evolution. In this way, for cubic crystals, a new parameter $s(P)$ is introduced. It is given by [6]:

$$s(P) = 1 + \frac{1}{4d^2(\rho_0)} \left[ \frac{1}{\rho_1} \frac{1}{t_1} \frac{1}{t_1(P)} \right] - d^2(\rho_0) s(P) = \frac{3}{t_1} - \frac{4}{t_1(P)}$$

where $A$ is a thermodynamic correction between the adiabatic and isothermal elastic moduli [6]. The equation of state can be expressed by the following relation:

$$V(P) = \frac{1}{0} = \frac{1}{s(P)}$$

3. Results

The pressure dependence of the transit time has been measured up to 5 GPa for $t_1(P)$, 6.3 GPa for $t_2(P)$, and 50 GPa for $t_3(P)$. A linear fit gives an account of this evolution.
order to compare our experiments, each transit time was normalized to an ambient sample length of 3 mm. The normalized transit times are shown in Fig. 3. Each experiment was done twice with different sample dimensions, with good reproducibility.

The pressure dependence of the elastic constants is shown in Fig. 4. The good agreement between our results and those published by Goncharova et al. [2] validate the new ultrasonic measurement set-up under pressure.

Moreover, the equation of state of Ge was calculated from the ultrasonic measurements and by using Eqs. (7) and (8). For comparison, the equation of state obtained by Menoni et al. [8] by X-ray diffraction in a diamond anvil cell are reported in Fig. 5. The values of $B_0$ and $B'_0$ were obtained by a fit of both sets of results using Murnaghan’s equation of state. The $B_0$ value (74.09 GPa) is consistent with the Menoni determination (74.9 GPa), but there is a large discrepancy (~35%) for the $B'_0$ determination (respectively 4.4 and 3). This can be explained by different techniques, the different nature of the sample (powder vs. single crystal) and the different experimental environments between the two experiments.

4. Conclusions

The good agreement between our ultrasonic results and those obtained previously by other methods validates this new ultrasonic measurement set-up under pressure. The pressure measurement using the NaCl Decker equation of state is a first step towards the direct and simultaneous determination of the sample equation of state.

This experiment will be improved to enable simultaneous ultrasonic and X-ray diffraction measurements on the studied material at higher pressure.
References