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Ultrasound Measurements of Cerium under High Pressure in a Large Volume Press Combined with Energy Dispersive X-ray Scattering and Radiography

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We performed ultrasonic velocity measurements of polycrystalline cerium under pressure across the γ-α phase transition at room temperature beyond 3 GPa. In contrast to previous methods all quantities were directly measured and no assumptions made in our approach: The length dimension of the sample was determined by *in situ* radiography, the pressure by energy dispersive X-ray diffraction from the sample and the travel time of the ultrasound pulses by the pulse overlap method. We give values for the longitudinal and transverse speeds of sound, c_L and c_T, respectively, as well as for the adiabatic bulk modulus B_S and shear modulus $G = \rho c_T^2$. Our Debye temperature values are in good agreement with recent results derived from phonon dispersion relation measurements on single crystals. The ratio of the Debye temperature values bracketing the transition, however, indicates a lattice contribution to the volume collapse of ~ (0.68 ± 0.06) k_B, in line with previous results based on neutron scattering, but larger than determined previously by the classical piston-cylinder apparatus. Possible reasons are discussed.

4f and 5f electron behavior in the lanthanides and actinides continues to be one of the most interesting aspects of modern condensed matter physics, in particular with regard to the electronic contributions to bonding and screening and the resulting structural behavior of the elemental solids under pressure. The most famous example for the physics of electron correlation is that of cerium which experiences a large iso-structural volume collapse of 14.5 % from the γ- to the α-phase at room temperature once it is compressed to 0.75 GPa. The Clausius-Clapeyron relation of the *thermodynamic equilibrium* line for $dP/dT = (S_\alpha - S_\gamma) / (V_\alpha - V_\gamma) = 5.14\cdot10^6$ J/m^{3}K (using 0.55 GPa at 295 K as one point to define this line and the critical point of 1.50 GPa at 480 K as the other [1,2]) permits one to calculate the change in entropy across the room temperature transition, ΔS (295 K) = $S_\alpha - S_\gamma = 1.77\ k_B$ (= .152 meV/K) with $k_B = 1.38\cdot10^{-23}$ J/K as the Boltzmann constant. $T\cdot\Delta S$ therefore amounts to 44.9 meV at room temperature and $P\cdot\Delta V$ = 22.2 meV (V_γ = 32.80 Å^3 and V_α = 28.06 Å^3 at 0.75 GPa and 295 K). Based on the volume changes at different temperatures Amadon *et al*. [3] concluded that the term $T\cdot\Delta S$ dominates ΔE and $P\Delta V$ in the relation $\Delta E = T\cdot\Delta S - P\cdot\Delta V$ all the way to the critical point and that the transition is mainly entropy driven. Since cerium displays Curie-Weiss type magnetic behavior in the γ-phase but only paramagnetic behavior in the high-pressure α-phase [4] it was in the past surmised that the change in entropy was caused by the loss in the magnetic moment (J = 5/2) resulting in $\Delta S = \ln(2J+1)\cdot k_B = \ln 6\ k_B = 1.79\ k_B$ which is virtually the same value found above for ΔS (295 K) based on *dP/dT*. Jeong *et al.* [5], however, pointed out that not only the electrons or their moments contribute to the transition but also the lattice, namely by $(0.75\pm 0.15)\ k_B$, slightly less than half, based on Debye temperature fits to isotropic thermal displacements versus temperature obtained by neutron scattering. Subsequently, this result has been the subject of intense scrutiny. Another technique, the ultrasonic investigation of macroscopic samples under pressure, can also provide Debye temperature values for the different phases in cerium. Moreover, ultrasonic measurements are of great importance since they also allow the determination of other macroscopic sample properties such as all the moduli (bulk, shear, Young's,..) as well as the Poisson coefficient. One very recent [6] and one much older [7] ultrasound experiment reported values for the lattice contribution that were smaller by more than a factor 2 than those of ref. [5]. Recently, two types of Kondo volume collapse models were found to be capable of fitting to the experimentally measured equation of state data [2], one offering only a 2 % lattice contribution to the VC whereas the other was consistent with roughly 50 % [2], largely because the lattice Grüneisen γ was much bigger in the two-phase region. Both were consistent with the data, in fact not only the Kondo volume collapse model appeared to replicate the data well but also the older Hubbard-Mott model [8], an enduring controversy over the last couple of decades. A recent experimental effort measuring the phonon dispersion relations of single crystals of cerium under pressure found indeed large changes in the mode Grüneisen parameters [9]. Nevertheless, the contribution of the lattice appeared to veer towards the smaller value, $(0.33 \pm 0.03)\ k_B$ [9], apparently settling the controversy. Here we report new data obtained with a new suite of experimental techniques. We find a value for ΔS that approximately confirms Jeong *et al.'s* data [5] while, surprisingly, is not being in conflict with the measurements from ref.[9].

The older ultrasound measurements from Voronov *et al.* [7] indicated a value of ~ $0.33\ k_B$ (θ_γ = 136 K, θ_α = 152 K). Doubts were cast on the data of ref. [7] by Decremps *et al.* [6], mainly because the material used by ref. [7] was only 98.5 % pure. Another problematic issue was the method of finding the length of the sample indirectly by iterations starting from the ambient value and then using 8% [7] for the volume change from the γ- to the α-phase at room temperature which conflicts with the actual 14.5%. In fact this issue plagues all ultrasound measurements that do not allow a direct length measurement and have to make assumptions about the volume change [6, 7, 10]. Yet another possible source of error is the fact that the authors of ref. [7] only determined the changes in the travel times with pressure compared to ambient, not the absolute travel times. The ultrasonic travel time at zero pressure was apparently calculated from published literature data [11] to which the changes determined at higher pressure were added. Nevertheless, the modern ultrasound experiment taken on high purity material in a high pressure intensifier via classical pulse overlap method found an even smaller value for the entropy change based on the Debye temperatures bracketing the VC transition: 0.22 k_B (θ_γ = 132.8 K, θ_α = 142.9 K) [6], in good agreement with a theoretical calculation [12].

The big difference between the previous ultrasound measurements and ours is that all the important quantities in the present experiment are measured directly without any assumptions: The sample is observed and measured by X-ray radiography *in situ*, the pressure is measured via energy dispersive X-ray scattering of the sample using the previously established equation of state [2], and the travel time of the ultrasound pulses is determined using the pulse-overlap method.

Cerium disks were cut and cored from 1 mm thick foil purchased from AlfaAesar. Nominal purity was 99.9%. Diameter was 1.5 mm. The appropriate height was achieved by polishing the puck down to about 0.65 mm. No further sample treatment was applied, such as heating to 500 C and annealing for 24 hours under a vacuum of 10^{-4} Torr [7], or melting/recrystallization under high-purity argon [6]. Figure 1 shows the sample assembly that would allow heating to temperatures of ~ 2000 K [13, 14] but was used here only at room temperature. The pressure environment is not completely hydrostatic as in the high pressure intensifier experiments to 1 GPa [6,7] but cerium is quite soft and the transition does take place at the correct pressure over a certain range as previously reported for polycrystalline cerium compressed in a diamond anvil cell [2] . Due to the diameter of the sample the energy dispersive X-ray diffraction is taken close to the outer edge of the cerium disk which also provides a check of the sample quality, especially important for higher temperature experiments later on. We did not observe signs of oxidation during our measurements. Radiography delivered a shadow-image that got focused onto a CCD camera by a microscope objective (see Fig. 2). The length per pixel was calibrated previously. The pulse overlap method determined the travel time of the ultrasound pulses taken between 15 and 30 MHz [13, 14]. The transverse waves coupled better at the lower frequency range, the longitudinal ones at the higher end. Our data start at about 0.25 GPa since there was no ultrasound coupling to the sample for lower pressures (see figure 3 for transverse waves). Figure 3 also shows clearly the change in the ultrasound response at the volume collapse.

Figure 4 displays the values for the ultrasonic velocities and compares them with previous results obtained by refs. [6, 7, 10]. The data of ref. [6] were extracted from their published data for B_S

and ρc_T^2 using our EOS [2]. The present data for the longitudinal wave velocity are practically indistinguishable from the previous data for pressures below the volume collapse at 0.75 GPa. After the volume collapse the new and previous data diverge, which is not surprising since the previous experimenters had to make assumptions about the size of the volume collapse (17% ref. [6] and 8% for ref. [7]), as mentioned before. The discrepancy again emphasizes the need for an independent length measurement especially for pressures higher than the transition.

While all the data for the longitudinal sound speed are in agreement in the γ-phase, the data for the transverse wave speeds are quite different. While the data of ref. [6, 7] and [10] are very close to within 1300 m/s (ref. [6] slightly below and ref. [7, 10] slightly above), our transverse wave speed hovers at about 1100 m/s and rises very slowly with pressure towards the transition (verified on 3 samples). Strangely enough, after the VC, there is much better agreement between the sets of data. Our measured values quickly climb during the VC to about 1400 m/s and from then on rise very slowly under compression. At 3.0 GPa the value is 1430 m/s. The data of ref. [7] jump across the VC to almost 1500 m/s and stop at 0.9 GPa, those of ref. [6] also climb to ~1400 m/s and stop at 1.03 GPa. The data of both ref. 6 and 7 climb quite more rapidly than ours, possibly a consequence of the assumption made about the size of the volume change. Based on these data, Fig. 5 exhibits our values for the shear modulus $G = \rho v_T^2$ and the bulk modulus $B_S = \rho\ (c_L^2 - 4/3\ c_T^2)$.

Since the longitudinal sound speeds are almost identical for all experiments below the VC but the transverse ones are quite different, one has to focus onto the different sample treatment as origin of the discrepancy. One of the key requirements when using polycrystalline samples is to ensure the right meso-scale environment, namely the existence of a sufficient number of grains in random orientation so that the sample can be considered elastically isotropic. Heating to high temperatures over a long period of time, annealing and even more so melting and recrystallization will likely grow large crystallites, possibly generate preferred orientations, and/or make the sample deviate from a sufficient number of grains in random orientation. This randomized sample texture is apparently recovered after pressurization across the volume collapse because then the transverse sound speeds are in good agreement with each other.

Fig. 6 shows a comparison of the adiabatic and isothermal bulk moduli, B_S and B_T, which are related by the ratio of the isobaric and isochoric specific heats: $B_S / B_T = C_P / C_V$. The value for B_T was evaluated by fitting a 3rd order polynomial to our P(V) data from 2008 [2] and then find $B_T = -V\ (dP/dV)_T$.

Since $C_P > C_V$, then $B_S > B_T$ always, as is the case in Fig. 6 except for the very low pressure data. Knowing either one, C_P or C_V, one could determine the other using B_S / B_T.

We now turn our attention to the effective (averaged) sound speed which is used in the calculation of the Debye temperature. C_{eff} is defined as follows [e.g. 15]:

$$c_{eff} = \left(\frac{1}{3}(\frac{2}{c_T^3} + \frac{1}{c_L^3})\right)^{-1/3}$$

Fig. 7 displays the behavior of the effective sound speed versus pressure. It starts at approximately 1250 m/s and climbs to beyond 1600 m/s at 3 GPa, an increase of more than 25 %. The transition region itself clearly ranges from ~ 0.6 to 1.1 GPa (1270 to 1540 m/s), accounting for the largest part of the Debye temperature change, about 22 %.

The effective sound speed in turn affects the Debye temperature which is evaluated by calculating

$$\theta_D = \frac{h}{2k_B} \cdot c_{eff} \cdot \sqrt[3]{\frac{6}{\pi\, V_{at}}} \sim c_{eff}/a$$

[15] with h as Planck's constant, k_B as the Boltzmann constant, V_{at} the atomic volume of a cerium atom in the lattice, c_{eff} the effective speed of sound and a as the lattice constant. The change in the lattice constant accounts for only 5.1 % across the transition.

Fig. 8 shows the Debye temperatures θ_D for our experiments. Our values at the edges of the transition region are 118 and 151 K. Using the approximation $\Delta S \sim 3\, k_B \ln(\theta_\alpha/\theta\gamma)$ one finds $\Delta S = (0.74 \pm 0.05)\, k_B$ assuming ± 1 K uncertainty. If one narrows the region to right beneath and above the VC (121 and 149 K) one still finds $\Delta S = (0.62 \pm 0.05)\, k_B$. Since the choice of the transition region is somewhat subjective we will use from now on the average of the two values: $\Delta S = (0.68 \pm 0.06)\, k_B$.

Also plotted in Fig. 8 are Debye temperature data reported by ref. [9]: 117 K for 0.0001 GPa (based on the neutron scattering data of [16], 122 K for 0.4 GPa, 123 K for 0.6 GPa, 138 K for 0.8 GPa and 157 K for 2.5 GPa. Evidently, our present data and those of ref. [9] agree very well with each other. The value for ΔS of ref. [9] comes from the use of 138 and 122 K as the Debye temperatures bracketing the VC at 0.6 and 0.8 GPa: $\Delta S = 0.37\, k_B$ (from the phonon density of states). Ref. [9] also reports 0.31 kB from the values for the mean square atomic displacements in the two phases. However, as shown in Fig.

7, our data (and implicitly those of ref. [9]) indicate that the transition is not finished at 0.8 GPa but continues on to ~ 1.1 GPa, even though cerium is reported to be crystallographically already in the α-phase at 0.8 GPa for single crystals according to Krisch et al. [9]. While the lattice has obviously transformed, the elastic properties appear to continue to evolve.

Our value of 0.68 k_B confirms the previously reported one by Jeong et al. [5], namely (0.75±0.15) k_B, which was found by measuring the slope of the isotropic thermal displacements obtained by neutron scattering at 0.527 and 0.412 GPa. The corresponding Debye temperatures in the α- and γ-phase at the transition were 133 and 104 K [5].

The present data point to inconsistencies in the literature values. Previous, by neutron scattering determined, values of the elastic constants, C_{ij}, for cerium at zero pressure are: C_{11} = 24.1 GPa, C_{12} = 10.2 GPa and C_{44} =19.4 GPa [16]. This would make the adiabatic bulk modulus $B_S = (C_{11} + 2 C_{12})/3$ = 14.83 GPa, significantly less than most measurements which put it at around or slightly more than 20 GPa as shown in Fig. 5 and Table 1 [1,2,5,6,7,17]. Therefore the published C_{ij}'s from ref. [16] appear too low. Similarly, it is well known that the Voigt and Reuss estimates for the shear modulus give upper and lower bounds for its value. With the C_{ij} above we find $G_V = 1/5*(C_{11} – C_{12} +3 C_{44})$ = 14.4 GPa and $G_R = 5*(C_{11} – C_{12})* C_{44} /[4* C_{44} + 3* (C_{11} – C_{12})]$ = 11.3 GPa, and our value for G at low pressure, 8.5 GPa (see Fig.5), would lie outside of this range (see Table 1). Moreover, using B_S = 14.83 GPa and the arithmetic Hill average, $G_{Hill} = (G_V + G_R)/2$ = 12.85 GPa, and ρ = 6773 kg/m^3 for the zero pressure density, the longitudinal sound velocity should be ~ 2100 m/s to 2170 m/s, not the ~ 2300 m/s observed by all experiments. Again, it is likely that the C_{ij} values for cerium at zero pressure ought to be modified.

This is reflected in estimates of the Debye temperature based on the ultrasonic velocities. The best way to achieve this goal is accomplished by averaging the C_{ij} according to a method recommended by Ledbetter [18], which involves taking the Reuss and geometric Hill estimates of the shear modulus of 11.30 and 12.77 GPa (see above) to calculate c_T and c_L (see Table 1). This procedure gives the Debye temperature at zero pressure using the equation for θ_D given above, 138.4 K (Hill) and 130.7 K (Reuss) – see Table 1. According to Ledbetter these two values should bracket the real one - with the Hill value somewhat closer to the exact result but too high. The error, however, would be less than 3% for cerium's elastic anisotropy $2C_{44}/(C_{11} – C_{12})$ = 2.79 [18]. Clearly, the Debye temperatures of [9] as well as ours in the γ-phase are much lower. This despite the fact, that the Debye temperature value of ref. [9] was obtained from the same phonon dispersion relation that led to the C_{ij} reported by [16].

In summary we have measured the ultrasonic longitudinal and transverse wave velocities under pressure with a method that is not relying on any assumptions but on direct measurements only. This makes the technique ideally suited for the study of phase transitions with volume discontinuities such as cerium. While our values for the longitudinal wave velocity in cerium under pressure are in good agreement with all previous reports, we find that the previously reported transverse wave velocities are higher - one has to conclude that the heating, melting and recrystallization treatment of refs. [6, 7, 10] resulted in a texture that was far from a random aggregate of crystallites and the reported Debye temperatures for the γ-phase are too high which in turn made the reported entropy changes too small. Our result for $\Delta S = (0.68 \pm 0.06)\ k_B$ confirms approximately the intensely scrutinized value of $0.75\ k_B$ [5] for the lattice contribution to the entropy change across the VC in cerium. Inconsistencies with the published values for the elastic constants C_{ij} call for their renewed evaluation even at ambient pressure. Since the measurements of the transverse sound speed of ref. [7] are based on zero pressure measurements performed on lanthanide samples in cast form [10] this also asks for another measurement of the transverse sound speeds in all the lanthanides at zero pressure.

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Table 1

Zero pressure adiabatic bulk modulus B_S, shear modulus G, longitudinal and transverse sound speeds c_L and c_T, Debye temperature Θ_D

P = 0	B_S (GPa)	G (GPa)	C_T (m/s)	C_L (m/s)	Θ_D (K)
Present [@ 0.25 GPa]	23.4	8.5	1110	2250	115 (1246)
Exp. Ref. [9][4]					117
Exp. Ref. [6][1]	20.5	11.17	1284	2295	
Exp. Ref. [7],	20.28	12.35	1330	2300	135 (1476)
[10][2]	18.85		1350	2283	137
Exp. Ref. [11]	20.2	12.0	1332	2300	
Calc. based on	14.83	14.42 (G_V)[3a]			
C_{ij} of Ref. [15]		11.30 (G_R)[3b]		2101	130.7 (Reuss)
C_{11} = 24.1 GPa,		12.86 (G_{Hill} arithm.)[3c]			
C_{12} = 10.2 GPa,		12.77 (G_{Hill} geom.)[3d]		2169	138.4 (Hill geom.)
C_{44} = 19.4 GPa					
Theory [12]	30.2	14.84 (G_V)[3a]			
C_{11} = 37.9 GPa,		10.18 (G_R)[3b]	1226	2542	126.3 (1378)
C_{12} = 26.4 GPa,		12.51 (G_{Hill} arithm.)[3c]			
C_{44} = 20.9 GPa		12.29 (G_{Hill} geom.)[3d]	1347	2623	138.3 (1509)

[1]: c_L and c_T were calculated from their published values for $B_S = \varrho(c_L^2 - 4/3\ c_T^2)$ and $G = \varrho c_T^2$ and the equation of state from ref. 2.

[2]: Ref. 7 and 10 used the ultrasonic velocities at zero pressure from ref. 11 as starting point.

[3a]: $G_V = 1/5*(C_{11} - C_{12} + 3\ C_{44})$

[3b]: $G_R = 5*(C_{11} - C_{12})*\ C_{44} / [4*\ C_{44} + 3*(C_{11} - C_{12})]$

[3c]: G_{Hill} (arithm.) = $½(G_V + G_R)$

[3d]: G_{Hill} (geom.) = $(G_V \cdot G_R)^{½}$

[4]: Using the same neutron scattering data as ref. 16 for the basis of their evaluation.

Figures

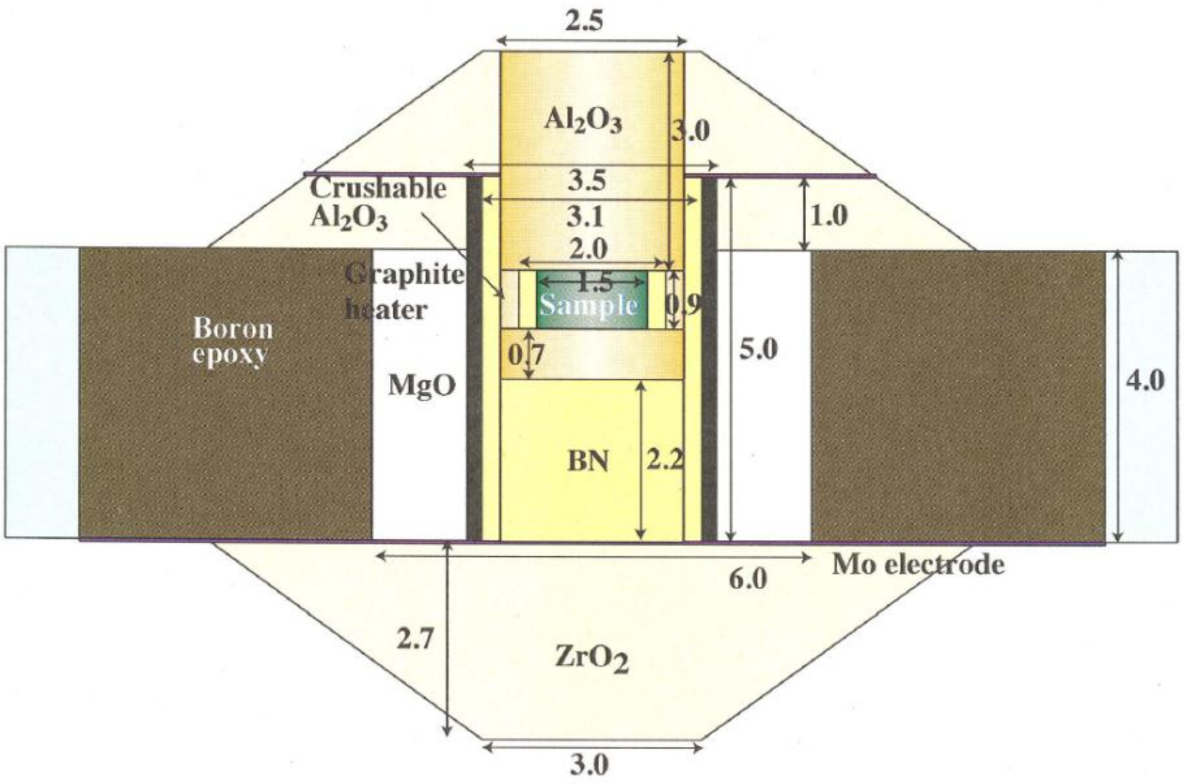

Fig. 1: Schematic of the sample assembly with dimensions (refs. [13, 14]).

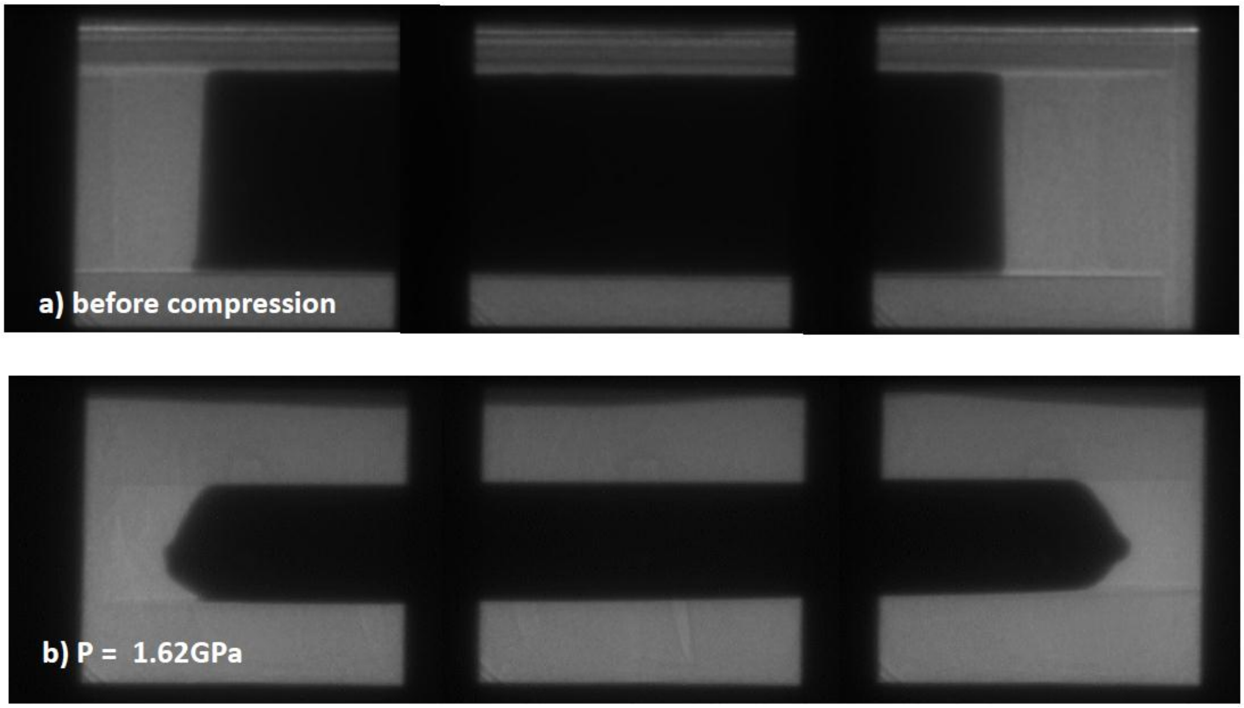

Fig.2 : Composites of three radiography images showing the left edge, center and right edge – each window approximately 0.9 mm wide, slightly less than the white beam dimension.

a) before compression - The 1.5 mm wide round cerium disk of 0.630 mm thickness is sitting between the two Al_2O_3 plugs and surrounded on the side by soft BN and crushable Al_2O_3 .

b) after completion of the volume collapse at 1.62 GPa. The thickness of the sample is an average across the center image.

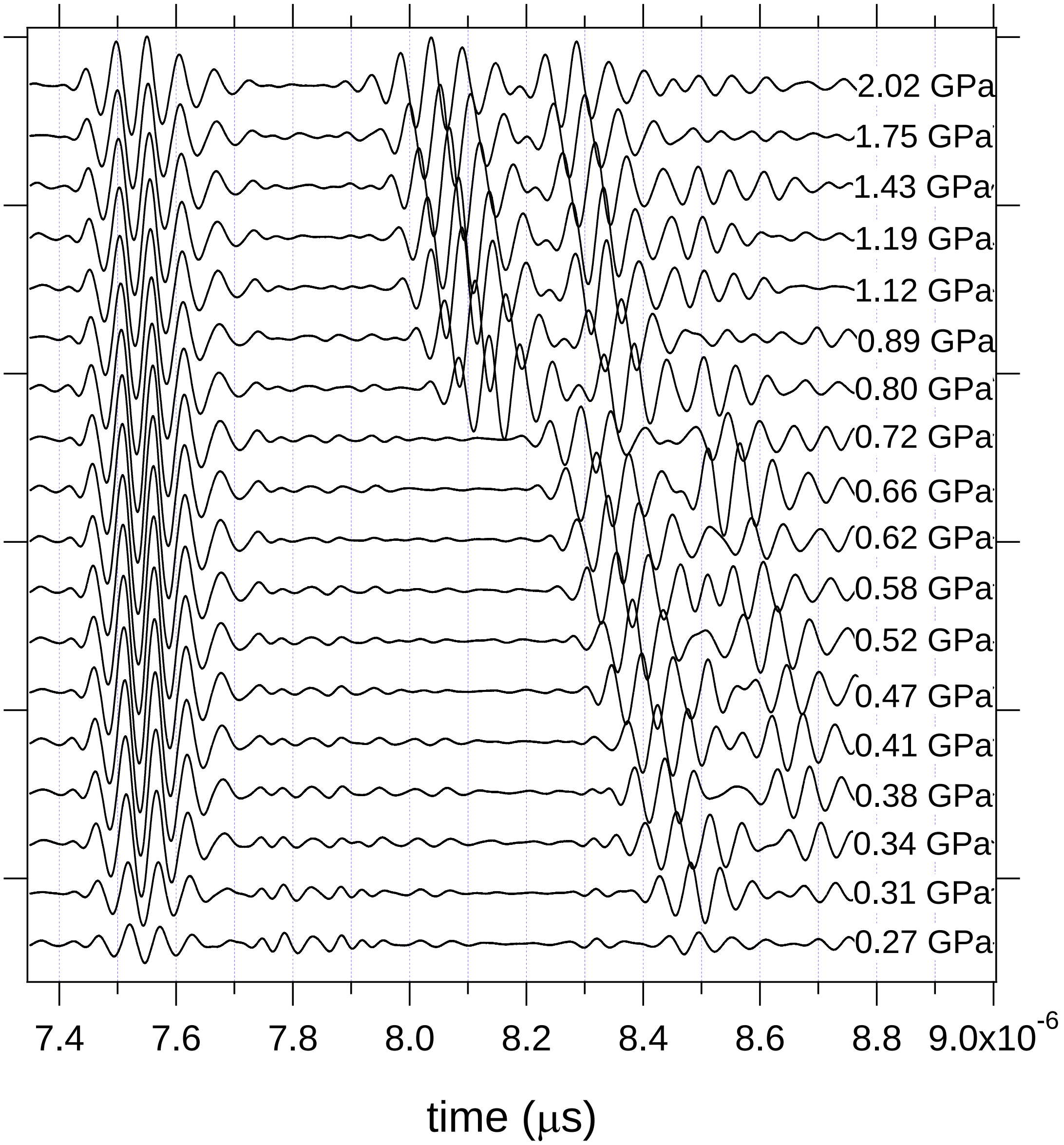

Fig. 3: Traces of transverse waves at 20 MHz. Lowest pressure is at the bottom where almost no reflection of the wave from either the top of the sample (at ~ 7.45 μs) or from the bottom (starting at ~ 8.4 μs) is observed. Clearly visible is the volume collapse between 0.72 and 0.80 GPa.

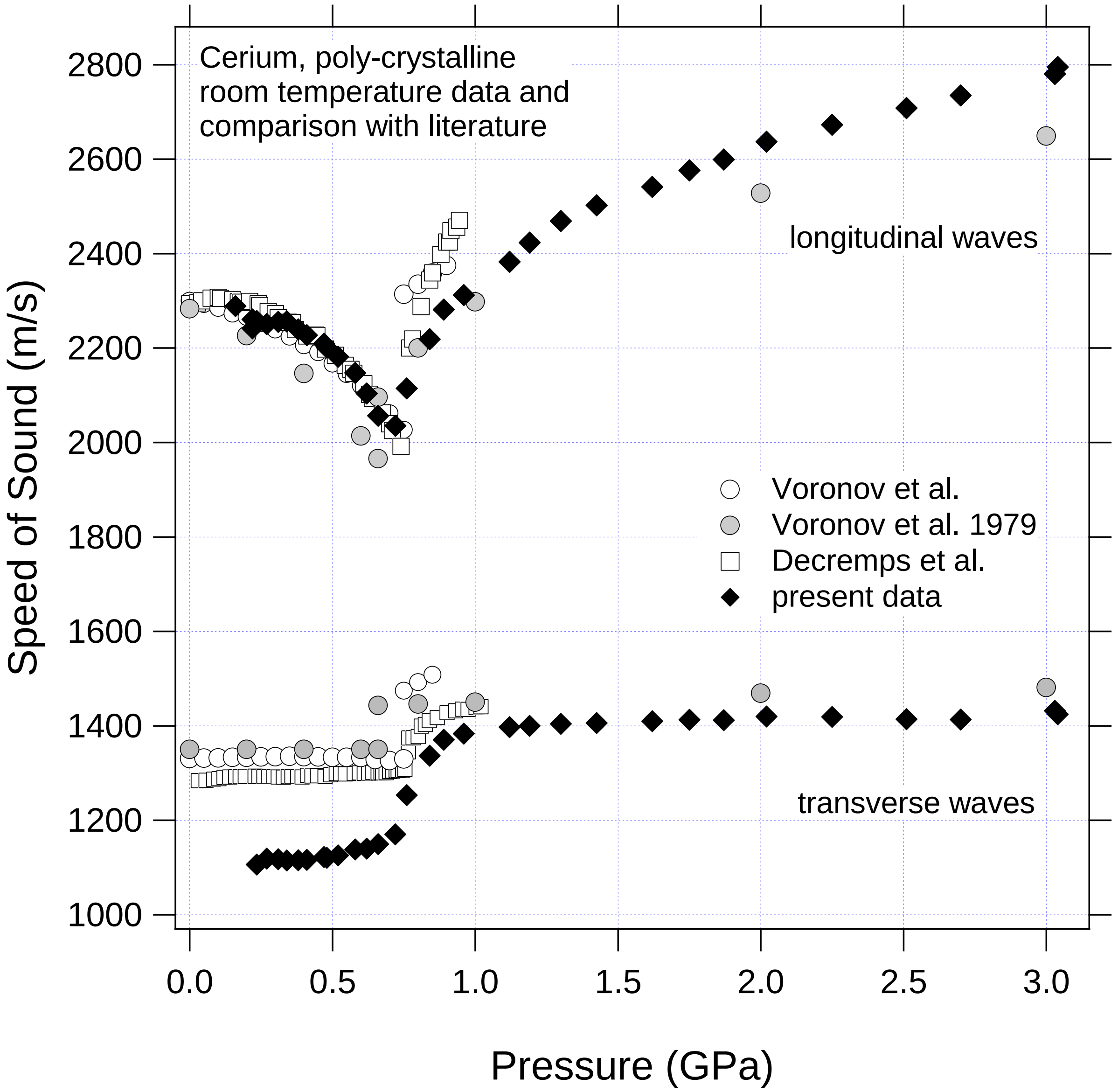

Fig. 4: Ultrasonic speeds (longitudinal, transverse) and comparison with previously reported data. For sake of clarity only one set of data from our samples is shown. The 1960 data of ref. 7 extend to 0.9 GPa, the later ones from 1979 [10] are more sparse but reach beyond 8 GPa.

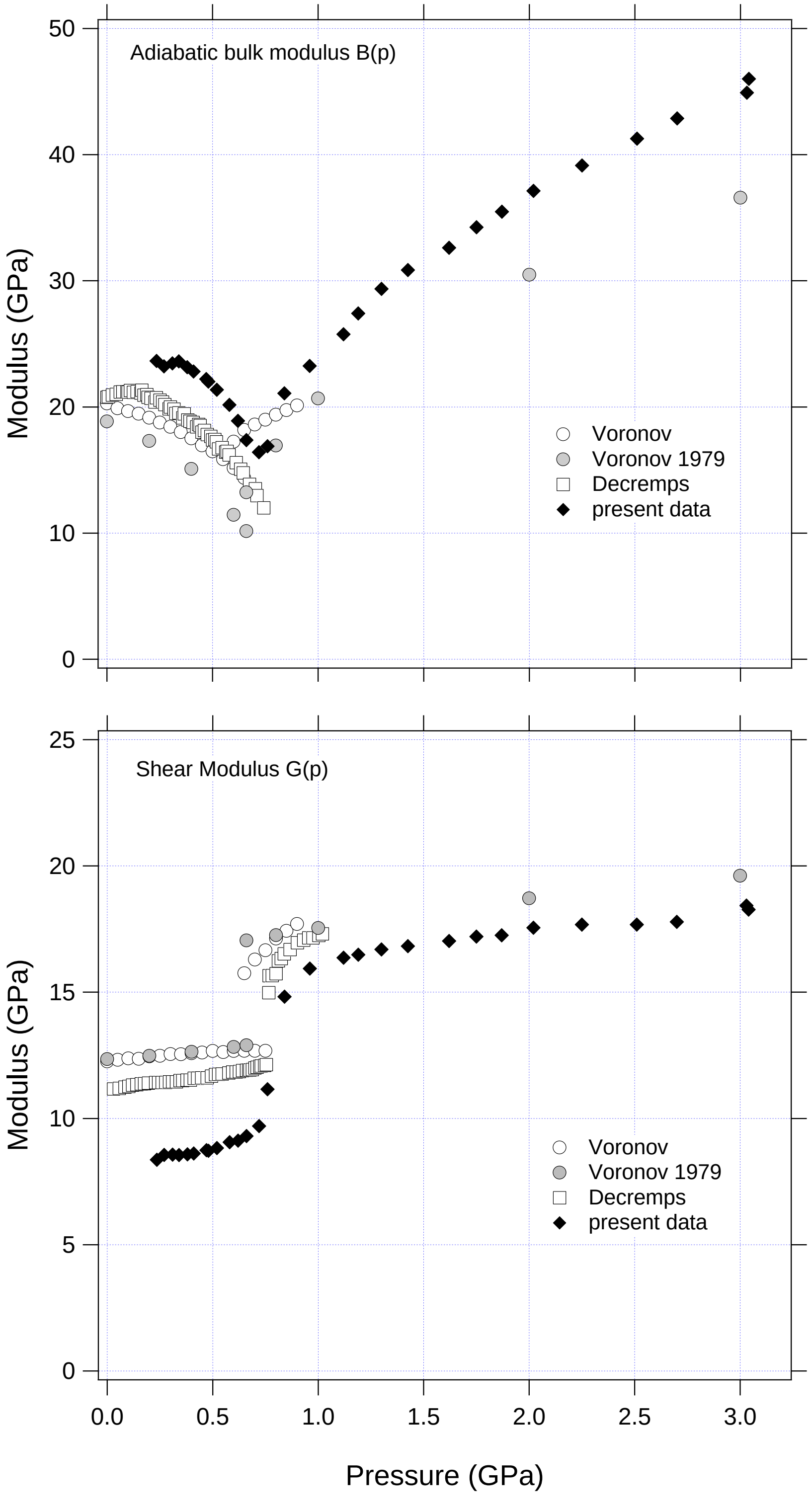

Fig. 5: Values for the the adiabatic bulk modulus B = $\rho(c_L^2-4/3\rho c_T^2)$ and the shear modulus ρc_T^2.

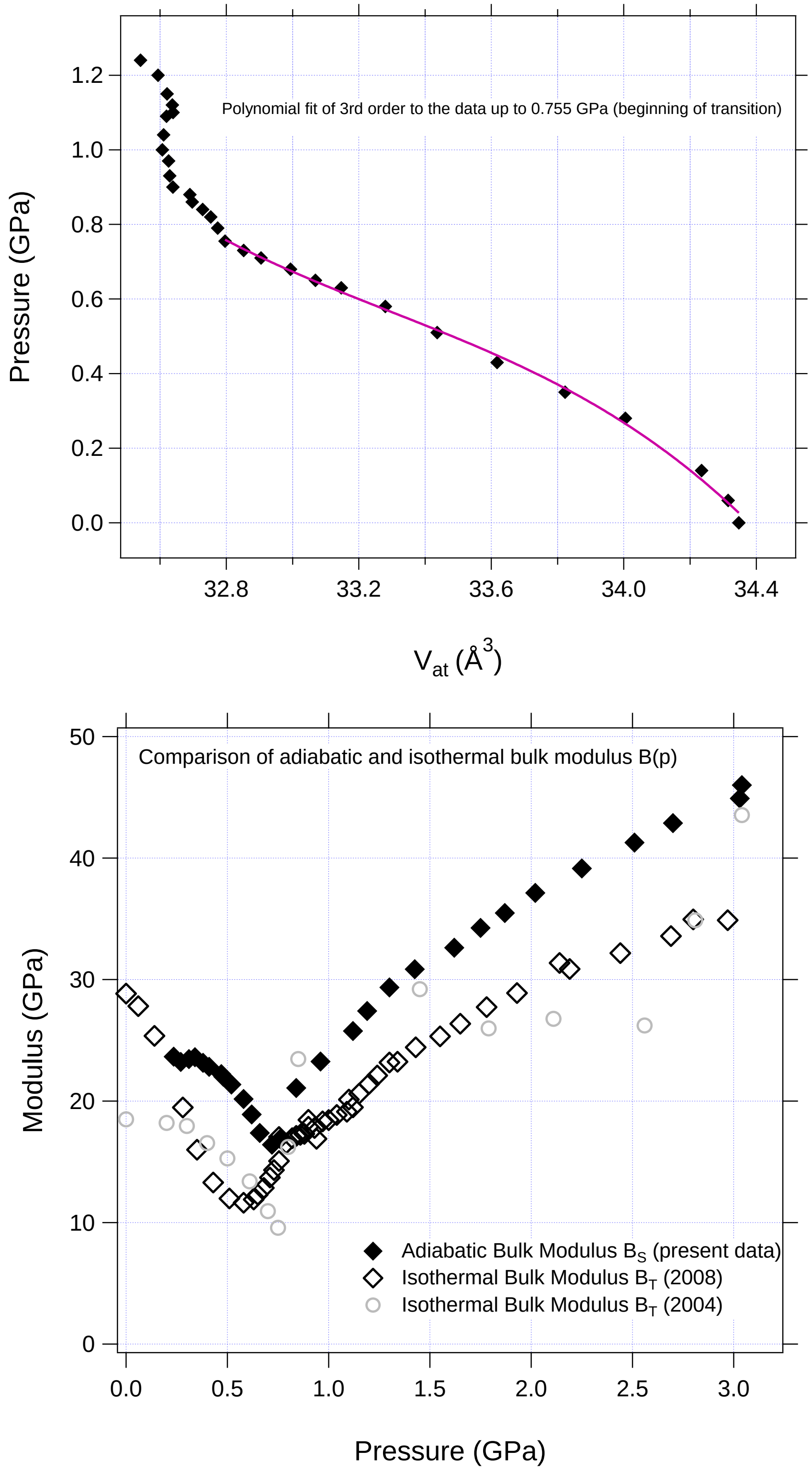

Fig. 6: Comparison of B_S and B_T which are related by the thermodynamic equation $B_S/B_T = C_P/C_V$ (meaning B_S should always be bigger than B_T). A third order polynomial was fitted to the room temperature isothermal P(V) data up to the transition pressure of 0.75 GPa (ref. [2], top panel) and the value of B_T evaluated by $B_T = -V\,(dP/dV)_T$. Other B_T data (2004) are taken from ref. [5].

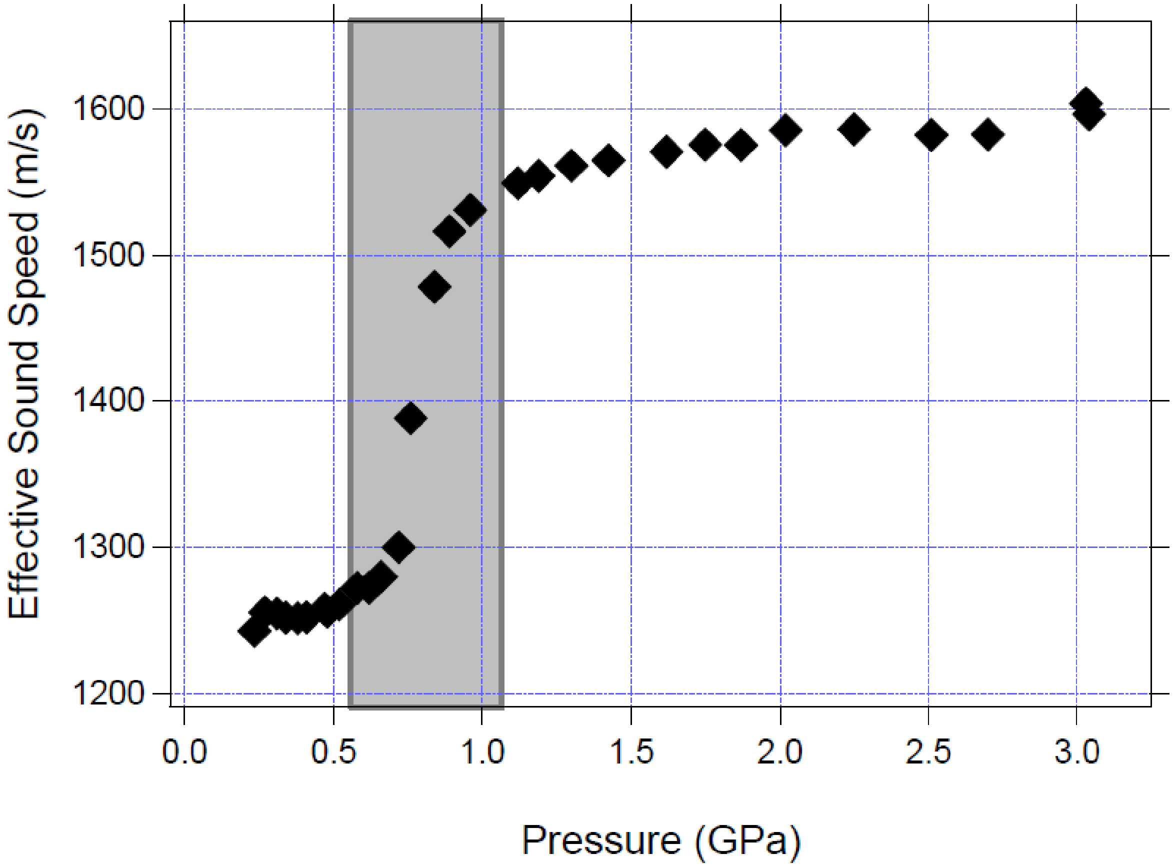

Fig. 7: Effective sound speed in cerium versus pressure at room temperature. The shaded region emphasizes the pressure range (mixed phase regime) over which the changes take place.

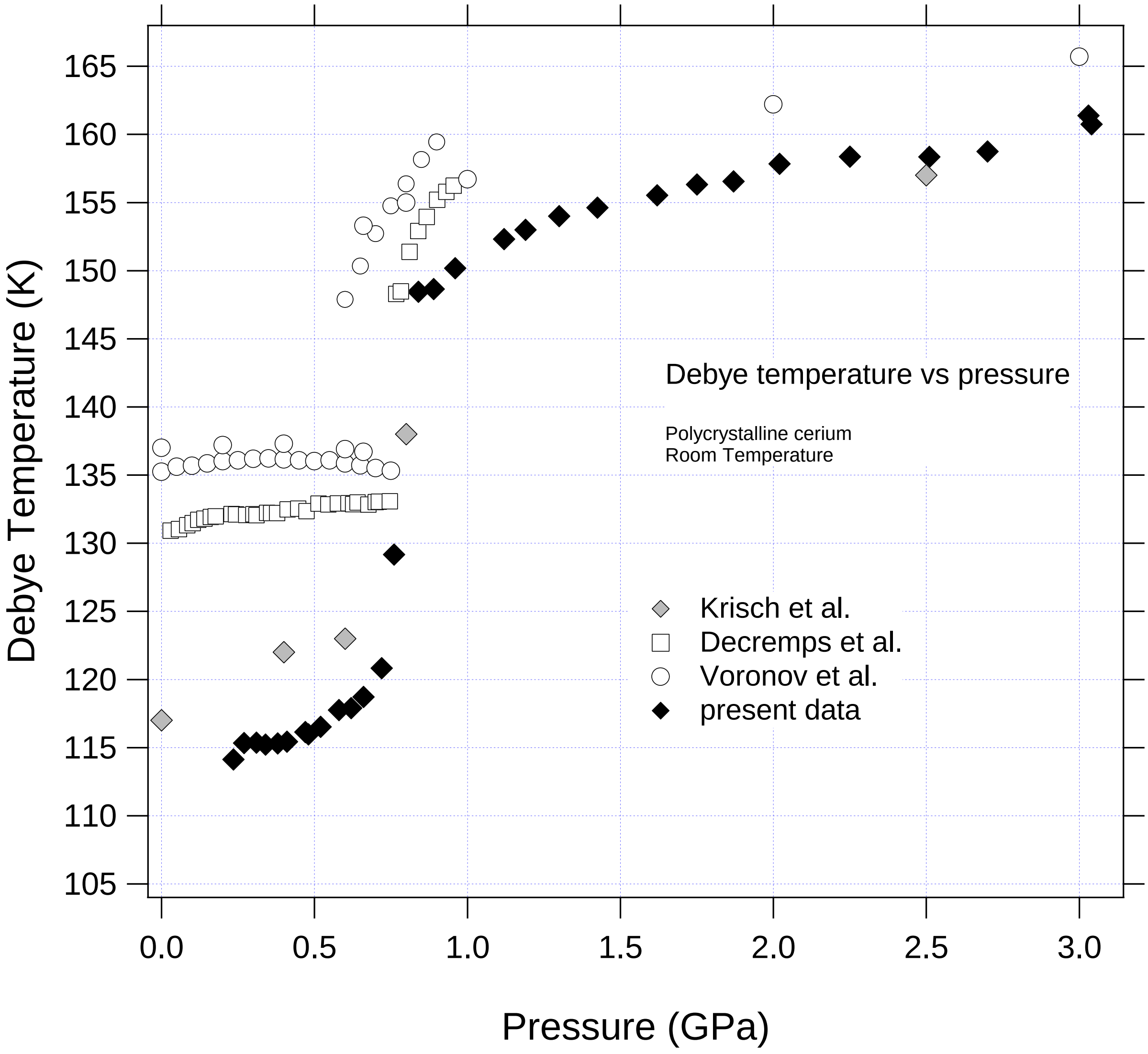

Fig. 8: Debye temperatures of cerium versus pressure. The zero pressure data point from ref. [9] is based on the neutron scattering data from ref. [16]. The other values at 0.4, 0.6, 0.8 and 2.5 GPa are in good agreement with ours.