

Elastic constants of galena down to liquid helium temperatures

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Abstract. The elastic constants of single crystal galena have been determined from the measured ultrasonic velocities down to liquid helium temperature. A cryostat incorporating an arrangement to inject the liquid bonding material at low temperature is described. At 5 K, the values of elastic constants are $C_{11} = 14.9_0$, $C_{12} = 3.5_1$ and $C_{44} = 2.9_2 \times 10^{10}$ N/m².

Keywords. Ultrasonic velocities; elastic constants; low temperatures; galena.

1. Introduction

The experimental study of ultrasonic velocities is an important method of studying the interatomic forces in crystals. The study of the variation of the elastic constants with temperature along with the thermal expansivities and heat capacities is necessary to study the variation of Debye temperature θ_D , Gruneisen parameter γ and anisotropy parameter A^T . These parameters can be correlated to the microscopic structure and interactions in the crystal (Born and Huang 1966)

The major obstacle for the extensive work in the low temperature elastic constants is the failure of the bonding material used to bond the transducer and the sample, as the temperature is varied. We present the results of measurement of the elastic constants of single crystal galena down to liquid helium temperatures. For many geological crystals, the information of the elastic constants is available only near room temperature (Landolt-Bornstien 1966). The measurement of elastic constants of galena down to 77 K is already available (Sivaraman *et al* 1978). A cryostat suitable for use down to 4 K is described along with the bonding problems between the transducer and the sample.

2. Experimental details

A bath-type of cryostat fabricated for the purpose has been used to cool the sample. The liquid helium glass dewar is surrounded by a liquid nitrogen shield as schematically shown in figure 1. The sample chamber is hung by two thin-walled cupronickel tubes as shown in figure 2. A vacuum of better than 10^{-6} torr is maintained in the sample chamber. The specimen-transducer assembly is suspended in the sample chamber by means of thin-walled stainless steel tubes, so as to minimise the thermal

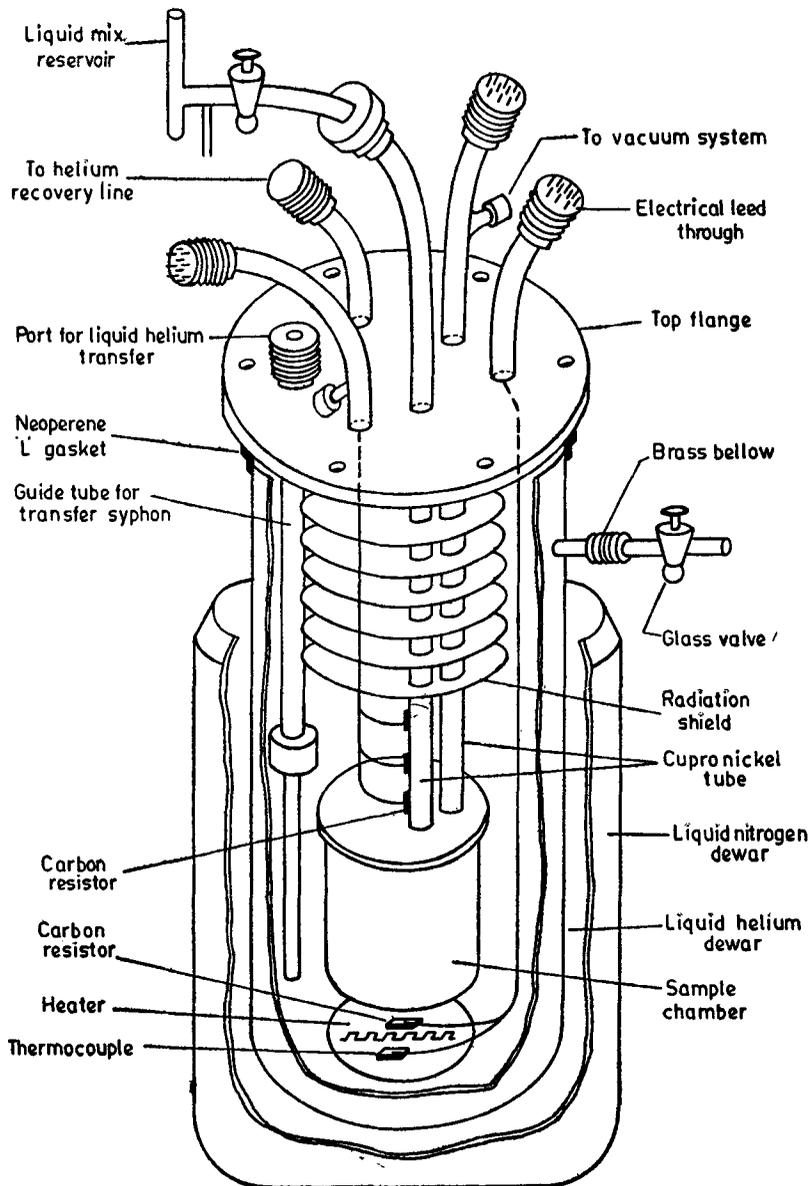


Figure 1. Schematic of the cryostat assembly.

link between the sample and the surroundings. To ensure minimum thermal gradient across the sample, an isothermal shield is provided for the sample. The temperature is measured using a Ge-resistor in the range 40-4 K and a copper-constantan thermocouple in the range 40-300 K.

An injection mechanism has been incorporated in the design, so that several liquid mixtures can be tried as bonding materials at low temperatures. For this a thin-walled stainless steel tube of small diameter (2 mm) is positioned directly above the transducer and very close to it. The other end of the tube (at room temperature)

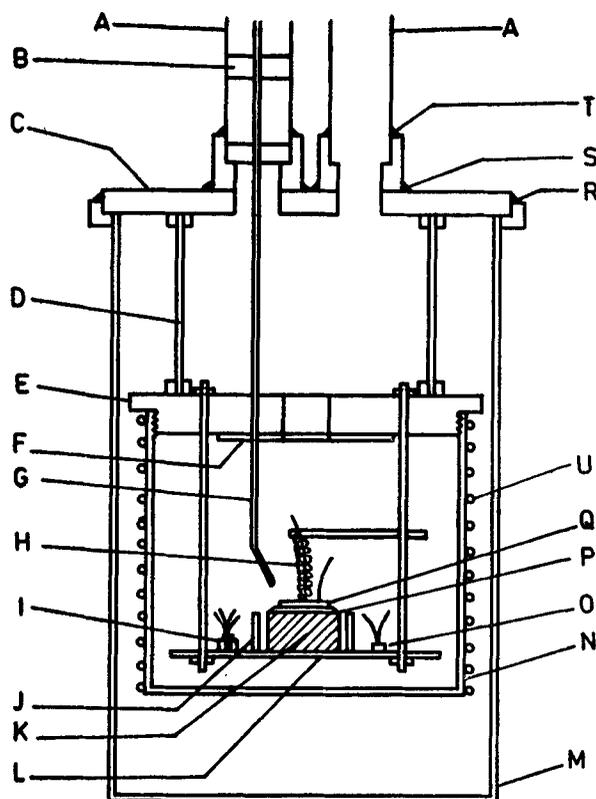


Figure 2. Sample chamber for ultrasonic studies showing the liquid injection mechanism. A—Cupronickel tube; B—Teflon spacer; C—Top flange; D—Thin walled tube; E—Threaded flange; F—PCB for anchoring electrical wires; G—Injecting tube; H—Spring; I—Germanium resistance thermometer; J—Thermal shield; K—Sample; L—Sample base; M—Outer vessel; N—Inner vessel; O—Thermocouple; P—Bond; Q—Transducer; R—Wood's metal joint; S—Braze joint; T—Solder joint; U—Heater.

is connected to a glass reservoir through a high vacuum valve. The bonding liquid to be tried at low temperature, for example EPA mixture as discussed below, petroleum ether, etc. is stored in the glass reservoir. By opening the glass valve in a controlled fashion, the liquid can be injected on the transducer-sample assembly. The part of the liquid that flows between the transducer and the sample acts as the bond.

Very good single crystals of galena obtained from the Geological Survey of India have been used in the present measurements. The samples were cut parallel to [001] and [110] directions and polished to better than 0.5μ . The orientation was confirmed by x-ray back reflection photographs and found to be correct to within $\frac{1}{2}$ degree. It is necessary that the sample diameter be larger than the transducer in order to avoid side wall effects (Truel *et al* 1969).

The bonding of a transducer such as a quartz plate or disc to a specimen is done with a variety of materials depending on the specimen and the environment where it is to be used. It may become necessary to use different bonding materials in different temperature regions. (Padaki—unpublished Ph. D. thesis for details). The failure of bonding may be due to two reasons. One is the cracking of the bond materials due

to the differential thermal contraction between the specimen and the binder and the transducer and the binder. The second reason may be that changes in the property of the bond material like embrittlement with temperature may produce mismatching in the acoustical impedance between the transducer and the specimen. In the first case a sudden vanishing of the echoes at some temperature is observed, whereas slow deterioration of echoes is observed in the second case.

In the present measurements a special low temperature araldite supplied by Thor Cryogenics Company has been used successfully down to liquid helium temperatures. Nonaq Stopcock grease (Fisher Scientific Company) works satisfactorily down to 150 K. The EPA mixture consisting of 5 parts (by volume) of ethyl ether, 6 parts of isopentane and two parts of ethyl alcohol works well from 150 to 66 K. The echoes are found to be better with nonaqueous and EPA mixture bonding in the temperature region 300–66 K in comparison with the special low temperature araldite. The measurements made with different bonding materials overlap satisfactorily. A number of other bonding materials like silicone grease failed at about 150 K.

The McSkimin's pulse superposition technique (McSkimin 1961) has been employed to measure the velocities. Co-axially gold plated X and Y cut transducers have been used to generate the longitudinal and transverse waves in the specimen. The centre frequency is 10 MHz. The resolution in the time-period measurement is better than 10 ppm. For calculating the ultrasonic velocities at temperatures other than room temperature it is necessary to correct the measured room temperature thickness of the specimen for the thermal expansion over the temperature region of interest. In the present investigation, the thermal expansion data presented by Novikova and Abrikosov (1964) were used and the variation of density was calculated, at different temperatures. The thermal expansion data was available only down to 30 K. It was extrapolated to 0 K and was used in the calculation.

3. Results and discussion

The relations between the ultrasonic velocities and elastic constants are well-known (Musgrave 1970). Galena being cubic in structure has three elastic constants namely C_{11} , C_{12} and C_{44} . The longitudinal and transverse velocities in the direction [100] yield C_{11} and C_{44} respectively. Having obtained C_{11} and C_{44} , C_{12} can be evaluated from the longitudinal velocity measured along [110] direction. Figure 3 shows the dependance of elastic constants with temperature from 300 to 5 K. It is seen that the temperature variation of C_{ij} shows the usual behaviour found for crystalline materials with each elastic constant almost approaching a constant value at low temperature, the maximum being at 0 K (extrapolated). In the range of temperature investigated, the temperature dependance is free from anomalies or evidence of phase transition. The accuracy of the data is more for C_{11} and C_{44} , which are each determined from individual velocity measurements, but is less for C_{12} which is determined from a combination of many velocities measured.

Using the elastic constant data we have calculated the Debye temperature θ_D , the Gruneisen parameter γ , the adiabatic compressibility K_S and the anisotropy parameter A^T . The various physical parameters are compared at different temperatures in table 1.

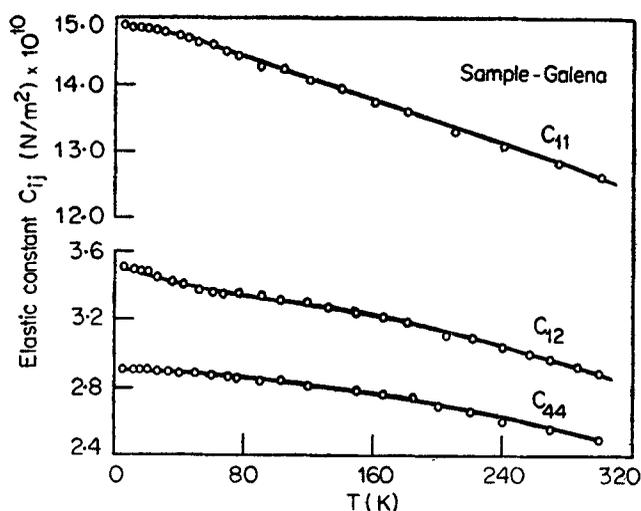


Figure 3. Temperature variation of the elastic constants of single crystal galena.

Table 1. Various physical parameters compared at 300, 77 and 5 K.

	5 K	77 K	300 K
C_{11} ($\times 10^{10}$ N/m ²)	14.9 ₀	14.4 ₄	12.6 ₅
C_{12} ($\times 10^{10}$ N/m ²)	3.5 ₁	3.3 ₅	2.9 ₁
C_{44} ($\times 10^{10}$ N/m ²)	2.9 ₂	2.8 ₆	2.5 ₀
θ_D in K	230	227	213
γ	3.3 ₁	3.0 ₇	2.4 ₁
A^T	0.51 ₄	0.51 ₆	0.51 ₈
K_S ($\times 10^{-10}$ m ² /N)	0.13 ₆	0.14 ₂	0.16 ₂

3.1 Debye temperature

Different methods are available (Alers 1965) for evaluating θ_D . In the present study Marcus and Kennedy's (1959) approach has been employed for computation of θ_D . Figure 4 shows the temperature variation of θ_D . The calorimetric determination of θ_D (Parkinson and Quarrington 1954) is also shown for comparison. Calorimetric data are available down to 20 K only. Often the calorimetric value of θ_D begins to decrease around $\theta/2$, has a minimum and then rises to attain a constant value below $\theta/50$. At temperatures less than $\theta/50$, the calorimetric and elastic values are expected to be the same. It is known that θ_D from elastic measurements is representative of the $\nu \rightarrow 0$ state in the $g(\nu)$ spectrum. This is true only for $T \rightarrow 0$. It is known that due to the existence of peaks in the $g(\nu)$ and the validity of $g(\nu) \propto \nu^2$ (Debye model) near $\nu \rightarrow 0$, the θ_D (calorimetric) shows a minimum (Gopal 1966). This is known for example in NaI (Berg and Morrison 1957), NaCl (Lundqvist *et al* 1959), Al (Walker 1956) and Na (Dixon *et al* 1963). The variation in θ_D (elastic) with temperature is quite small as expected.

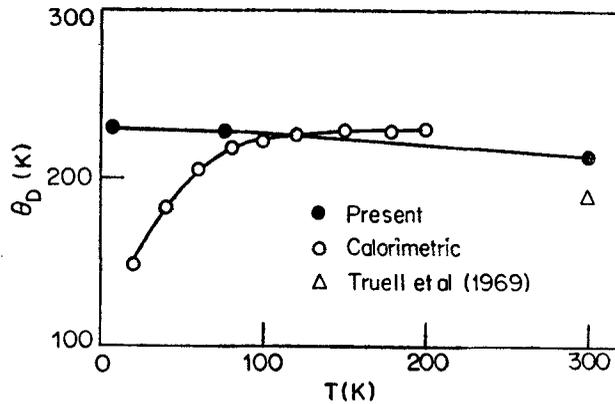


Figure 4. Temperature variation of the Debye temperature of galena.

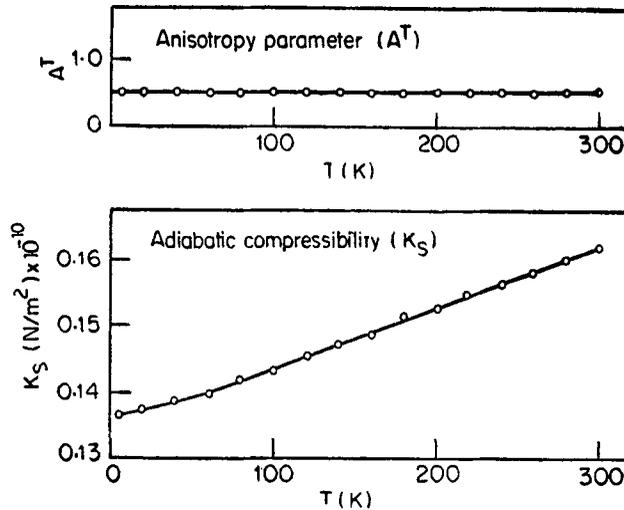


Figure 5. Adiabatic compressibility and anisotropy parameter of galena as a function of temperature.

3.2 Adiabatic compressibility (K_S) and anisotropy parameter (A^T):

In figure 5 the values of compressibilities and the anisotropy parameter are presented. The anisotropy parameter is practically constant over the complete temperature range. It is known that the temperature dependence of A^T is negative for systems like $TiCl$, $TiBr$, $CsBr$, etc. It is positive for samples like $NaCl$, LiF , KCl , etc (Gulays *et al* 1975). According to the theoretical results of Nikanorova *et al* (1965) A^T should be constant which agrees with our result.

3.3 Gruneisen parameters

The Gruneisen parameter γ is defined as $\gamma = \beta V / C_v K_T$, where β is the coefficient of volume expansion, C_v is the specific heat and K_T is the isothermal compressibility.

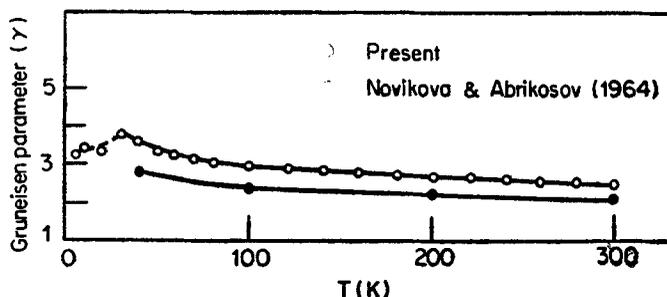


Figure 6. Gruneisen parameter of galena as a function of temperature. Data below 30 K based on extrapolated values.

The specific heat data provided by Parkinson and Quarrington (1954) have been used to determine K_T using the simple relation $C_p/C_v = K_T/K_S$. The temperature variation of γ is plotted in figure 6 along with Novikova and Abrikosov's values of γ (γ values are corrected for temperature variation of K_T). Below 30 K, the γ values are based on the extrapolation of the C_v and β data and are shown by the broken line. From the simple ideas of Gruneisen parameter it is expected that γ be a constant. But in a number of systems γ is not constant. The theory of Barron (1955) is generally used to explain the variation of γ with temperature. This theory expects a decrease in γ at around $\theta/5$. γ is generally held to vary between γ_∞ ($T \rightarrow \infty$) and γ_0 ($T \rightarrow 0$). The Barrons theory is well obeyed by materials such as Al, Cu, etc. It is generally known that the crystals of similar structure have same value for γ (Busch and Schade 1976). However LiF and NaCl both being f.c.c. crystals, LiF shows an increase in γ at low temperatures whereas NaCl shows a decrease in γ (White 1965). Vetelino *et al* (1970) have predicted a maximum in γ at low temperatures in CsCl, CsBr and CsI based on the calculation of phonon spectrum. White and Collins (1973) have observed these maxima but suggest that such maxima could be due to errors in the heat capacity data used. The maximum for γ noticed in our results could also be due to the same cause. The nature of temperature variation of γ investigated down to 40 K by Novikova and Abrikosov (1964) agrees with our result.

In conclusion the elastic constants of galena have been measured down to liquid helium temperature. Using known values of α and C_v , θ_D and γ have been calculated for this system. K_S and A^T have also been determined from the measured values of elastic constants. The results are compared with the results of some other inorganic systems.

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