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Measurement of the elastic constants of aluminium and copper using a continuous wave ultrasonic technique

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The CW technique, using a Q-meter and also a sweep frequency ultrasonic spectrometer, has been employed to measure all the elastic constants in single crystal aluminium, and C_{11} and C_{44} in single crystal copper in the temperature range, 77°K to 300°K. The details of the instrumentation, and a comparison of the results with earlier works in Al and Cu are given.

1. Introduction

Various experimental methods of increasing sophistication and versatility are being developed for the measurement of the elastic constants of solids. The now commonly used methods can be grouped into three categories. The continuous wave (CW) method, the pulse method and the CW/pulse method (Truell *et al* 1969). All the methods are widely in use. Using CW acoustic excitation, in general, there are two methods of measurement of the elastic constants. One is the Q-meter technique using which Bolef & De Klerk (1962) measured the elastic constants of several alkali halides. The other is the CW sweep frequency method. In this, one can use an acoustic probe of the reflection kind or the transmission kind (Yee & Gavenda 1968). In this paper we will describe a CW sweep frequency acoustic spectrometer using the reflection kind of acoustic probe and also present some data obtained for the temperature variation of the elastic constants of alumium and copper. We will also give some details of the Q-meter experiment done by us with one of the samples of aluminum.

2. General Principle

In order to determine the elastic constants in solids one has to measure the velocity of sound. The elastic constants can be related to the velocity of sound using the continuum elasticity theory. In cubic crystals there are three independent elastic constants. These three elastic constants can be determined by measuring the velocity of sound using appropriate propagation direction and proper polarization of the acoustic wave. The relations we will be using are

$$v_L[100] = (C_{11}/\rho)^{\frac{1}{2}}, \quad \ldots \quad (1)$$

$$v_T[100] = (C_{44}/\rho)^{\frac{1}{2}}, \quad (2)$$

and

$$v_L[111] = ((C_{11}+2C_{12}+4C_{44})/3\rho)^{\frac{1}{2}}, \quad (3)$$

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where the subscript L or T indicates longitudinal or transverse mode of excitation, the directional cosines of the propagation vector of the acoustic wave are given in the square bracket and ρ is the density of the solid.

Thus if the velocity of sound with the above mentioned direction and mode specifications is measured in a cubic crystal, all the three elastic constants can be determined. In order to measure the velocity of sound in CW experiment one has to measure the mechanical resonance frequencies of the composite resonator, consisting of the transducer, bond and the sample. The mechanical resonance frequency is the frequency at which the composite resonator contains an integral number of half wave lengths of the sound wave, and it is denoted by ν_n where n is the number of integral half wave lengths. The mechanical resonance frequency ν_n and the separation between two successive mechanicl resonance frequencies $\Delta\nu$ are related to the velocity of sound by the following relations (Bolef & De Klerk 1963)

$$v = (2l/n)[\nu_n-(m_T/m_S)(\nu_T-\nu_n)] \qquad \ldots \quad (4)$$

and

$$n = (\nu_n/\Delta\nu), \qquad \ldots \quad (5)$$

where l is the length of the sample, m_T and m_S are the specific masses (i.e., ρl, where ρ is the density) of the transducer and the sample respectively. The effect of the bond which is usually small, has been neglected.

The general method of measurement of velocity or its change with respect to temperature is as follows. First as many of the successive mechanical resonance frequencies as possible of the composite resonator are located and the average separation $\Delta\nu$ between two successive ones is estimated. Then choosing the most prominent of the mechanical resonances, the value of n is obtained from its frequency ν_n using eq. (5). Eq. (4), is then used to obtain the absolute value of v. To study the variation of v with temperature, the particular mechanical resonance is tracked as the temperature of the sample is changed.

This being the general method, there are various ways of locating mechanical resonances using CW technique. Two methods have been used by the present authors viz., the Q-meter method (Bolef & De Klerk 1963) and the CW sweep frequency spectrometer method. The former was used only to locate the mechanical resonances and to make a rough estimate of the velocity of sound. For temperature variation study, the latter method was used.

3. The Q-meter Technique

In this method the instruments required are a Q-meter and the associated variable frequency oscillator. A standard coil is used, leaving the capacitor terminals of the Q-meter open, to bring the resonance frequency into the range of the transducer response peak by tuning the capacitor control of the Q-meter. At resonance, the Q-meter will show a peak value of Q, say Q_{max}. Then the composite resonator is connected to the capacitor terminals keeping the oscillator frequency unchanged and the capacitor control is retuned for Q_{max}. The value of Q_{max} now as compared to its value without the composite resonator will be smaller. Now the frequency of the oscillator is slowly changed all the time tuning the capacitor control so that the Q-meter reads Q_{max}. Thus Q_{max} is studied as a function of frequency. When the frequency of the oscillator, is equal to one of the mechanical resonance frequencies of the composite resonator, there will be a sharp peak in the value of Q_{max}. This fact can be deduced (Bolef & De Klerk, 1963) from a simple analysis of the Q-meter circuit and the electrical equivalent circuit of the composite resonator circuit given in figure 1. Thus the mechanical resonances can be located.

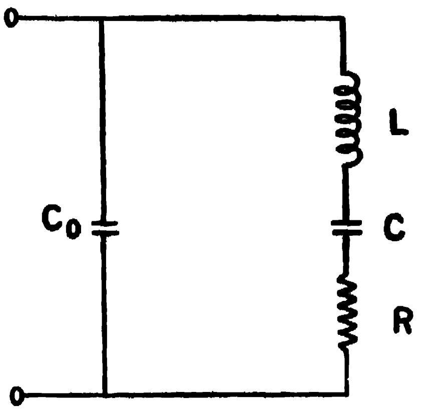

Fig. 1. Electrical equivalent circuit of the composite resonator. C_0 = static capacitance of the transducer plate and the stray capacitance.

This method was used to study a single crystal sample of aluminium. The details of the sample are given in the next section. The Q-meter used was Marconi Instruments Type TF1245. The oscillator was of model TF1246. The frequency of the mechanical resonance was measured by a frequency counter of Hewlett Packard type 524D. The experiment was done at room temperature only. Several mechanical resonances were located but as the frequency moves away from the transducer resonance peak, the measurement becomes more uncertain. So in figure 2 only three resonances are shown. The average separation between the mechanical resonances was 135 KHz. Using eq. (1), the velocity of longitudinal sound wave along [100] direction in Al, at room temperature is obtained as 6.06×10^5 cm/sec. This value is in reasonable agreement with the established results.

As described by Bolef & De Klerk (1963), from the knowledge of Q_{max} at the mechanical resonance frequency and the value Q_0 at the same frequency without the composite resonator in the circuit, the resonant resistance of the composite resonator R_S can be estimated. This R_S is a measure of the acoustic losses in the sample and hence is related to the attenuation coefficient α, and the width of the mechanical resonance line. The attenuation coefficient obtained is $\alpha = 2.35 \times 10^{-3}$ cm^{-1} and the width of the mechanical resonance line is obtained to be $\Delta\nu_S = 2.64$ KHz. Taking this value of $\Delta\nu_S$ a Lorentzian line shape is drawn in figure 2 for the mechanical resonance line of frequency 9.893 MHz

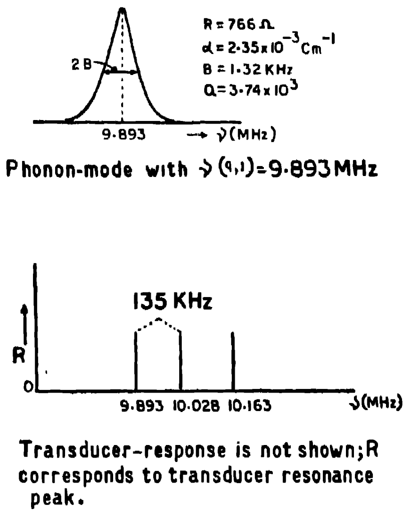

Fig. 2. Acoustic phonon modes in aluminium single crystal, q_{11} [100].

4. *CW* Sweep Frequency Spectrometer Method

The block diagram of the instrument is shown in figure 3. The sweep frequency oscillator, the r.f. amplifiers, the detector and the audio amplifier are built as a combined system and the circuit diagram is shown in figure 4. The double triode 5670 is the oscillator tube. In the grid of the tube, apart from the tuning condenser and a coil, a varicap is connected in parallel. A sawtooth voltage is applied to the varicap to have a frequency sweep. The sawtooth is taken from the Tektronix type 162 wave form generator. This gives a fixed sawtooth voltage of $+20V$ to $+130V$ at variable repetition_frequencies. In order to have a control over the range of frequency sweep an attenuator is interposed between the wave form generator and the varicap. The maximum sweep obtained was about 300 KHz around 10 MHz.

The output of the oscillator is taken from the cathode and is fed to the first r.f. amplifier, E180FI. In the plate load of this amplifier, apart from a 300Ω resistor, the matching network and the sample are included,, the details of which are shown in figure 5. This unit presents a frequency dependent impedance. When the frequency of the r.f. voltage is equal to the mechanical resonance frequency, the composite resonator presents an impedance minimum. The matching network consists of a double tuned over coupled system which inverts the impedance of the composite resonator so that the minimum of impedance in the latter is reflected as a maximum in the primary coil of the transformer which is in the plate circuit of the amplifier. If the frequency sweep covers any mechanical resonance, the output of this amplifier will exhibit a reduction in the r.f. amplitude at the mechanical resonance frequency. This is amplified by another stage r.f. amplifier, E180F II. Here a facility for auxilary output is provided for display on the oscilloscope.

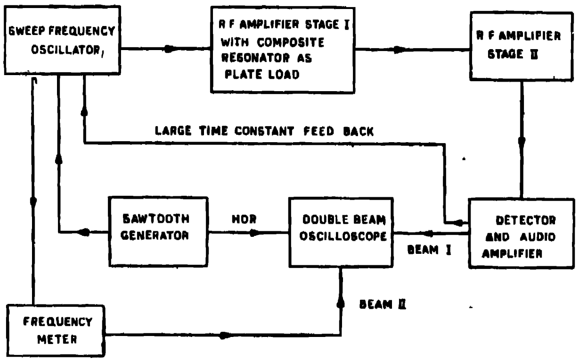

Fig. 3. Block diagram of the *CW* ultrasonic spectrometer.

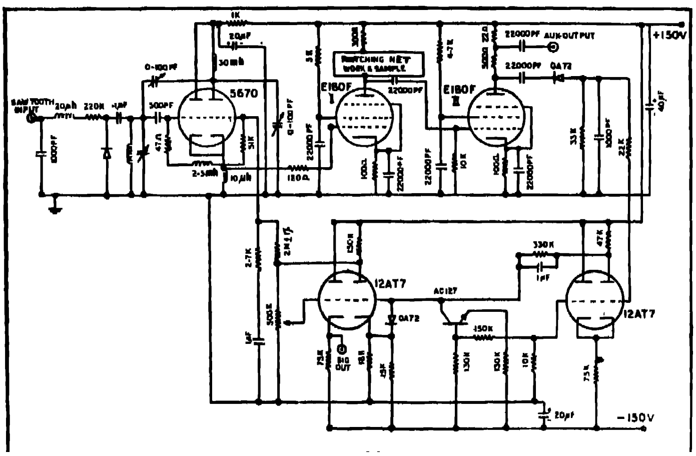

Fig. 4. Circuit diagram of the *CW* sweep frequency oscillator-detector system.

The output of the r.f. amplifier is detected by the OA72 diode and the audio output is amplified by the two 12AT7 tubes. The final signal output is taken from the cathode follower. The audio feed back provided in the circuit ensures a long time constant amplitude stability. The system in the present configuration gave a stable performance between 8-12 MHz.

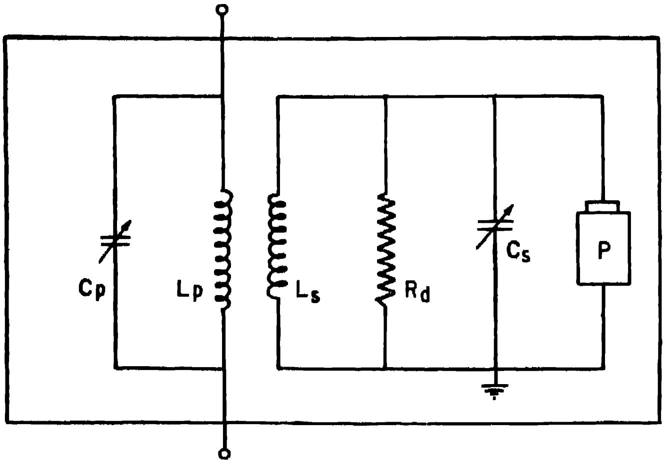

Fig. 5. Matching network. $L_p = 2.4\mu H$, $L_s = 2.17\mu H$, $R_d = 9.4K\Omega$, P-probe with the transducer and the sample.

The output of the audio amplifier is displayed on the Philips double beam oscilloscope model No. PM3230. The horizontal sweep of the oscilloscope is the same as the sweep given to the varicap so that the time base of the oscilloscope is converted into a frequency scale. The oscilloscope display is photographed and is given in figure 6. An explanation of the observed shape is given by Miller & Bolef (1969).

Fig. 6. Mechanical resonance signal in aluminium single crystal, as displayed on the oscilloscope.

To measure the frequency of the mechanical resonance, a frequency meter, Bendix radio BC221-M was used. The antenna of the frequency meter picks up the signal from the oscillator. The frequency meter has its own oscillator, the frequency of which can be varied. When the frequency of the oscillator is equal to the one the frequency meter picks up, a *peep* is emitted by the frequency meter. This *peep* is displayed on the other beam of the oscilloscope. By visual synchronisation of the *peep* from the frequency meter and the mechanical resonance signal, the frequency of the latter can be measured. The oscilloscope display of the mechanical resonance along with the synchronised *peep* is shown in figure 7.

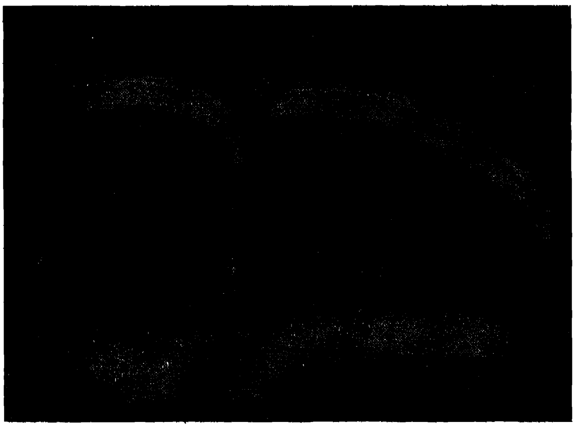

Fig. 7. Mechanical resonance signal in aluminium along with the *peep* from the frequency meter.

5. Samples and Measurement

Experiments were performed on three samples, two aluminium and one copper single crystals. The two aluminum single crystals are cylindrical in shape with the axis of the cylinder parallel to [100], (sample I), and [111], (sample II) directions respectively. The length of sample I along the cylinder axis is 2.243 cm and that of sample II is 2.473 cm at room temperature. The sample I was used in the Q-meter experiment. The copper single crystal (sample III) is in the shape of a cube. The edge of the cube is oriented along [100] direction. The accurate length of the cube in the direction in which the sound wave is passed is 1.023 cm. All the three samples are purchased in the properly cut, oriented and polished form from Semi-elements Inc. USA. The quoted purity of the samples is 99.999%.

The transducers were quartz plates of proper cut and they are suitably gold-plated (purchased from BEL, Bangalore), and bonded to the samples with DC200 silicone oil of viscosity 30,000 CS at 25°C. The sample was then mounted in a probe described earlier (Ghatak & Sinha 1974).

The method of measurement was to locate, at a definite temperature, as many mechanical resonances as possible and find out the average separation between them. Thus $\Delta\nu$ is known. The frequency of the most prominent mechanical resonance is measured. From these two data n is determined using eq. (5). The velocity of sound is determined using eq. (4). For temperature variation, only this particular mechanical resonance is tracked with the help of the *peep* from the frequency meter so that ν_n is known at all temperatures. Since n is known, the velocity of sound at all temperatures is determined. The temperature of the sample was varied from liquid nitrogen to room temperature. The temperature was measured using copper constantan thermocouple.

6. Results and Discussion

For aluminum samples, the longitudinal velocity along [100] (sample I) and along [111] (sample II) and the transverse velocity along [100] was measured. These three measurements, as can be seen from eq. (1) to eq. (3), are sufficient to determine all three independent elastic constants. In the case of copper, only the longitudinal and the transverse velocities along [100] direction were measured. So only two elastic constants could be determined. Another crystal with a different orientation would have enabled us to determine the third one. The temperature variation of the elastic constants in aluminum are plotted in figure 8 and those in copper are plotted in figure 9. For aluminium instead of C_{12} the combination $1/3(C_{11}+2C_{12}+4C_{44})$ is plotted because that is what is actually measured. The elastic constant vs. temperature curves are almost linear for all cases. In the case of aluminium, the percentage variation of elastic constants

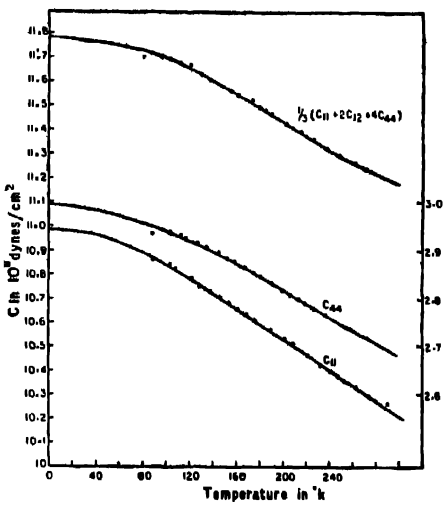

Fig. 8. Temperature vs elastic constants in single crystal aluminium. Vertical co-ordinate on the right hand side refers to C_{44}.

between 80°K and 240°K is 4.54, 3.25 and 6.43 for C_{11}, $1/3(C_{11}+2C_{12}+4C_{44})$ and C_{44} respectively. The percentage variation of C_{44} is obtained to be 1.98. In the case of copper, the percentage variation between 120°K and 240°K is obtained to be 1.96 and 3.90 for C_{11} and C_{44} respectively. Since for aluminum all the three elastic constants could be determined it is possible to determine the Debye temperature. The Debye temperature is given by (Alers 1965)

$$\theta_D = (h/k_B)(3N/4\pi V)^{1/3}(C_{11}/\rho)g,$$

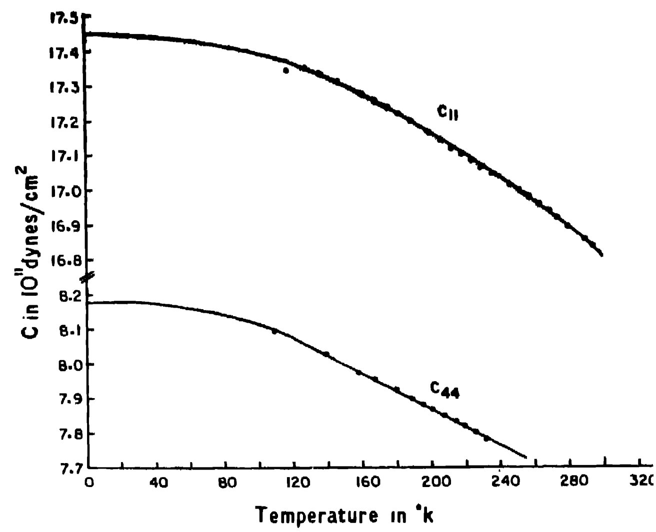

Fig. 9. Temperature vs elastic constants in single crystal copper.

where h is the Planck's constant, k_B is the Boltzmann's constant, (N/V) is the conduction electron density and ρ is the density. The function g depends on two variables $u = (C_{11}-C_{12})/2C_{11}$ and $v = (C_{44}/C_{11})$ and is plotted in the article by Alers (1965). From the knowledge of C_{11}, C_{12} and C_{44} and by interpolation from Aler's plots, the value of g is determined. The elastic constants appearing in the above formula are the zero temperature values. They have been obtained by linear extrapolation of the C vs T curves. The value thus obtained for the Debye temperature of aluminium is 433.6°K.

There are several earlier measurements of the elastic constants of aluminium and copper both for the absolute values and the variation with temperature. Sutton (1953) used a composite oscillator technique to measure the elastic constants of aluminium from 63°K to 773°K. Pulse echo technique has been used by many workers to study aluminium. Lazarus (1949) and Schmunk & Smith (1959) have studied the pressure dependence of elastic constants of aluminum at room temperature. Vallin *et al* (1964) measured the elastic constants at 4.2°K, 77°K and 300°K. Kamm & Alers (1964) measured elastic constants between 4.2°K and 300°K. Recently Ghatak & Sinha (1974) measured C_{11} and 1/3 $(C_{11}+2C_{12}+4C_{44})$ for aluminium from 77°K to 300°K using CW /pulse technique. In the literature there are theoretical calculations available. Local pseudopotential theory has been used by Wallace (1970) to calculate the phonon frequencies in aluminum. The elastic constants were then obtained by using the method of long waves. Suzuki (1971) obtained the elastic constants using the method of homogeneous deformation. The elastic constants obtained theoretically are the zero temperature values. Some of these results are given along with the values obtained in the present work for aluminum in tables 1 and 2. Table 1 contains the absolute magnitude and table 2 the percentage variation with temperature. The absolute values obtained in the present work agree resonably well with those of Ghatak & Sinha, Kamm & Alers as well as with those of Vallin *et al.* All

Table 1 : Comparison of the present results with the earlier works for aluminium (C_{ij} in units of 10^{11} dynes/cm^2)

Reference	Temperature °K	C_{11}	$\frac{1}{3}(C_{11}+2C_{12}+C_{44})$	C_{44}	C_{12}
Present	80	10.895±10%	11.725±10%	2.95 ±10%	6.228±10%
Ghatak & Sinha (1974)	80	11.395	12.110		
Kamm & Alers (1964))	80	11.373		3.128	6.191
Vallin *et al* (1964)	77	11.42		3.06	6.032
Sutton (1953)	77	12.225		3.063	7.07
Wallace (1969)	0	7.98		3.45	4.21
Suzuki (1971)	0	11.13		6.03	5.59

Table 2 : Percentage variation of the elastic constants in aluminium in the range of 80°K–240°K

Reference	C_{11}	$\frac{1}{3}(C_{11}+2C_{12}+4C_{44})$	C_{44}	C_{12}
Present	4.54±10%	3.25±10%	6.43±10%	1.98±10%
Ghatak & Sinha (1974)	4.27	4.69		
Kamm & Alers (1964)	4.45		7.09	1.15
Sutton (1953)	5.47		6.8	4.1

these values are in disagreement with those of Sutton. The percentage variaton of the elastic constants between 80°K and 240°K obtained from the present measurements is in good agreement with that of Ghatak & Sinha and Kamm & Alers for C_{11} and C_{44}. But the percentage variation of C_{12} is in between the values given by Sutton, and Kamm & Alers. The errors quoted in the present measurement take into account the inaccuracies in the measurement of frequency as well as temperature.

The elastic constants of copper were measured by Overtone & Gaffney (1955) and by Caine & Thomas (1971). Overtone & Gaffney measured the elastic constants of Cu from 4.2°K to 300°K. Caine & Thomas measured at room temperature. Lazarus (1949) measured the elastic constants at room temperature and studied their pressure dependence. Theoretical calculations were performed by Fuchs (1935), White (1958) and Sinha (1966). The experimental results of the above mentioned workers and the present results are given in table 3. Also given in table 3 are the theoretical values of Fuchs, White & Sinha. The present results agree with those of Overtone & Gaffney at 120°K. The percentage variation of the elastic constants in the range from 120°K to 240°K for the present results and the one calculated from Overtone & Gaffney are given table 4. They agree quite well.

Table 3. Comparison of the present results with earlier works for copper (C_{ij} in units of 10^{11} dynes/cm^2)

Reference	Temperature °K	C_{11}	C_{44}
Present	120	17.36±10%	8.075±10%
Overton & Gaffney (1955)	120	17.441	8.013
Lazarus (1949)	300	17.10	7.56
Fuchs (1935)	0	17.5	8.9
White (1958)	0	17.66	7.55
Sinha (1966)	—	—	7.54

Table 4. Percentage variation of the elastic constants in copper in the range of 120°K–240°K

Reference	C_{11}	C_{44}
Present	1.96	3.90
Overton & Gaffney (1955)	2.22	3.79

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