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ELASTIC CONSTANTS OF CADMIUM FROM 300° TO 575°K

Y. A. Chang and L. Himmel

January 1966
ELASTIC CONSTANTS OF CADMIUM FROM 300° TO 575°K.

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ABSTRACT

The adiabatic elastic constants of single crystal cadmium have been determined by means of the ultrasonic pulse-echo technique over the temperature interval from 300° to 575°K. The values obtained at 300°K are: \( C_{11} = 11.45 \), \( C_{33} = 5.035 \), \( C_{66} = 3.750 \), \( C_{44} = 1.985 \) and \( C_{13} = 3.99 \) in units of \( 10^{11} \) dynes/cm\(^2\). The temperature dependence of the linear compressibilities, the Grüneisen parameter and the lattice contribution to the specific heat have been calculated using the elastic constants obtained in the present study and other necessary thermal data available in the literature.

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INTRODUCTION

Among metals crystallizing in the hexagonal close-packed structure, cadmium and zinc are the only two exhibiting anomalously large values of c/a. Although the occurrence of these large axial ratios is not entirely understood in terms of theory, it is clear that the highly anisotropic properties, such as elastic constants, coefficients of thermal expansion and electrical conductivities are associated with these large axial ratios. The elastic constants of cadmium over the temperature interval from 4.2º to 300ºK have been determined by Garland and Silverman(1,2). However, the elastic properties of cadmium from 300ºK to its melting point are of special interest, since there is a reversal of the temperature coefficient of the axial ratio within this temperature range(3,4). Moreover, the elastic properties of zinc from 4.2ºK to its melting point obtained by Alora and Neighbours(5) show interesting behavior with respect to compression in the directions parallel and perpendicular to the c-axis. In view of the structural similarities between cadmium and zinc and the reversal of the temperature coefficient of the axial ratio for cadmium, it was felt desirable to extend the measurement of the elastic constants of cadmium to temperature near its melting point.

Besides being valuable in the understanding of metallic binding, elastic constants are necessary in computing the Grüneisen parameter and the dilational contribution to the specific heat. This dilational term, which becomes appreciable at high temperatures, must be subtracted along with the electronic term from the experimentally determined specific heat at constant pressure in order to obtain the contribution due solely to lattice vibrations.

In the present study, the five elastic constants of cadmium have been
determined from 300° to 575°K using the ultrasonic pulse-echo technique. The compressibilities derived from the elastic constants were used to calculate the Grüneisen parameter and the dilational specific heat. The values of the lattice specific heat at constant volume obtained by subtracting the dilational and electronic terms from the experimentally determined values of $C_p$ were then compared with the classical value of $3R$ at high temperatures.

EXPERIMENTAL PROCEDURE

For a hexagonal crystal, four of the five independent elastic constants, namely $C_{11}, C_{33}, C_{44}$ and $C_{66}$, may be obtained by measuring the velocities of plane longitudinal waves and appropriately polarized transverse wave propagating in the directions parallel and perpendicular to the c-axis. These velocities are related to the elastic constants according to the following relationship (6,7):

$$C_{ij} = \rho v^2$$

where $\rho$ is the crystal density and $v$ is the appropriate wave velocity in question. The fifth elastic constant, $C_{13}$ must be obtained by measuring the velocities propagating in the direction at an angle $\phi$ to the c-axis. The velocities of acoustic waves propagating in this direction are related to the elastic constants by the following equations (6,7):

$$\rho v^2 = C_{66} \sin^2 \phi + C_{44} \cos^2 \phi$$

$$2 \rho v^2 = C_{11} \sin^2 \phi + C_{33} \cos^2 \phi + C_{44} \pm \left\{ \left[C_{11} - C_{44}\right] \sin^2 \phi \right\}^{1/2}$$

The term $v_t$ in equation (2) is the velocity of pure transverse waves propagating in this direction with plane waves polarized perpendicular to the c-axis. The other two velocities as expressed by equation (3) are of mixed character. The plus sign refers to a quasi-longitudinal wave velocity, $v_{qL}$ and the
minus sign refers to a quasi-transverse velocity, \( v_q \). Among the three wave velocities propagating in this direction, the measurement of either \( v_q \) or \( v_t \) will yield the fifth elastic constant, \( C_{13} \). The other two velocities served to have a check of internal consistency of the data. It is evident from equation (3) that there are two possible mathematical roots for \( C_{13} \). The correct one is determined by the lattice stability condition (8).

In the present study, the growing of three single crystals of 99.999% pure cadmium in the form of 2.5 cm diameter spheres was carried out in an inert atmosphere of high-purity helium in previously outgassed reactor-grade graphite crucibles using a modified Bridgman technique. One specimen with faces normal to the c-axis, a second with faces parallel to the c-axis and a third with faces at an angle of 55° to the c-axis were then cut from each of the three spherical single crystals by electric discharge machining. The orientations of these crystals were determined by back reflection Laue method to within 0.25°. To remove the cold-worked surface layer of these specimens due to electric discharge machining, the crystals were first etched in a solution of nitric acid and methyl alcohol (50% acid by volume). After etching, each crystal was then mounted inside a surface ground hardened steel ring of appropriate thickness and polished with 3 micron diamond paste on a vibratory lap (Syntron) until the two faces were flat and parallel to within \( \pm 0.0002 \) cm.

The velocities of acoustic waves were measured by means of a Sperry Products ultrasonic attenuation comparator (9) in a manner described previously (10, 11). X-cut and Y-cut quartz crystals, 1/2-in diameter and resonant at 10 kc/sec, were used to generate either the pure longitudinal or transverse waves. A special high-temperature epoxy resin consisting of Epon 1031 (Shell Development Co.) and nadic methyl anhydride served as the
bonding agent between the quartz transducer and the cadmium crystals from ambient temperature to about 575°C.

The density of cadmium at 500°C, 8.652 g/cc, used in computing the elastic constants was calculated from x-ray lattice parameters (3). To correct for the density and the measured sound velocities as a function of temperature, the thermal expansion data of Owen and Roberts (5) were used.

RESULTS

The smoothed values of C_{11}, C_{33}, C_{44} and \( \rho v_{q1}^2 (\phi = 22^\circ) \) taken from Fig. 1 are listed in Table I. Also included in this table are the smoothed values of C_{12}, C_{15} and the adiabatic compressibilities parallel and perpendicular to the c-axis: \( \chi_{||} \) and \( \chi_\perp \). The elastic constant C_{12} is not an independent modulus, but related to C_{66} by the equation, C_{66} = (C_{11} - C_{12})/2. The values of C_{15} were calculated from the directly measured quantities \( \rho v_{q1}^2 \) by means of equation (5). The adiabatic compressibilities parallel and perpendicular to the c-axis were derived from the smoothed values of the elastic constants. The appropriate equations are:

\[
\chi_{||} = \frac{(C_{11} + C_{12} - 2C_{15})}{C} \quad (4)
\]

\[
\chi_\perp = \frac{(C_{33} - C_{15})}{C} \quad (5)
\]

where C = C_{33}(C_{11} + C_{12}) - 2C_{15}. The values of \( \chi_{||} \) and \( \chi_\perp \) are plotted as a function of temperature in Fig. 2.

The number of significant figure displayed in Table I is greater than is warranted by the absolute accuracy of the measurements, but an extra figure has been retained to indicate the trend of the data with temperature. From the statistical spread or scatter in the experimental data as shown in Fig. 1, the uncertainties in the directly measured quantities: C_{11}, C_{33}, C_{44}, C_{66} and \( \rho v_{q1}^2 (\phi = 22^\circ) \) are found to be \% 0.5%. Using the standard propagation-of-error treatment (12), the uncertainties in the calculated quantities are
as follows: \( C_{12} = 1.7\% \); \( C_{13} = 3.26\% \); \( \gamma_{//} = 1.17\% \); and \( \gamma_{\perp} = 7.5\% \).

Two checks of internal consistency of the data were made at 300°K. The measured values of \( \rho v^2 (\phi = 22^\circ) \) was found to be 0.4\% higher than the computed value using equation (2). The second check of internal consistency of the data was made by computing the measured values of \( \rho v^2_{q1} + \rho v^2_{q2} \) at 300°K with the calculated value according to the relationship \( \rho v^2_{q1} + \rho v^2_{q2} = C_{11} \sin^2 \phi + C_{33} \cos^2 \phi + C_{44} \) (equation 3). Again the difference between the measured and calculated values is about 0.4\%.

As shown in Fig. 1, the elastic constants obtained in the present study join smoothly with the low-temperature data of Garland and Silverman\(^{(1,2)}\). The values of \( C_{11} \), \( C_{33} \), \( C_{44} \) and \( C_{66} \) reported by Abowitz\(^{(13)}\) at 300°K are in good agreement with our results. Since Garland and Silverman, as well as Abowitz, used a wrong value of \( \rho \) for cadmium at room temperature, all of their reported values of the elastic constants were corrected using the new values calculated from x-ray lattice parameters. The older values of the elastic constants obtained by Grüneisen and Goens\(^{(14)}\) as well as Bridgman\(^{(15)}\) are also included in Fig. 1 for comparison.

A comparison is also made for the isothermal volume compressibility, \( \chi^T \), derived from the elastic constants, and the directly measured values. The value of \( \chi^T \) obtained in the present study at 300°K is \( 2.21 \times 10^{-12} \text{cm}^2/\text{dyne} \), using the well-known thermodynamic relationship:

\[
\chi^T = \chi + \alpha^T v / \rho_p
\]

where \( \chi^T \) is the isothermal volume compressibility, \( \chi \) is the adiabatic volume compressibility, \( \alpha = \text{isotropic-adiabatic-volume-compressibility} \), is the volume coefficient of thermal expansion, \( T \) is the absolute temperature, and \( V \) is the specific volume. The values of \( \alpha \) and \( \rho_p \) are available in the literature\(^{(3,16)}\). Bridgman\(^{(17)}\), Richards\(^{(18)}\) and Adams et. al.\(^{(19)}\) have
reported the following values of $\chi^T$: $2.10 \times 10^{-11}$, $2.10 \times 10^{-11}$ and $2.25 \times 10^{-11}$ cm$^2$/dyne, which are in reasonable agreement with the value obtained in the present study.

DISCUSSION

For the purpose of comparison, the ratios: $C_{11}/C_{12}$, $C_{11}/C_{33}$ and $\chi_{11}/\chi_{12}$ at 300°K for eleven metals with hexagonal close-packed structure are listed in Table II together with the values of $c/a$ taken from the compilation of Pearson(27). With the exception of thallium, the elastic anisotropy of those metals correlates qualitatively with the $c/a$ ratio. For cadmium and zinc, the values of $C_{11}/C_{12}$, $C_{11}/C_{33}$ and $\chi_{11}/\chi_{12}$ are much greater than unity, presumably due to the anomalously large values of $c/a$.

As shown in Fig. 1, all the elastic constants decrease with increasing temperature. However, the shear anisotropic factor, $A = C_{11}/C_{12}$, at first increases with temperature to about 460°K and then decreases above this temperature (see Table I). This unusual temperature dependence of $A$ may be associated with the reversal of the temperature coefficient of the $c/a$ axial ratio(3,4).

Due to departure from the ideal $c/a$ ratio for cadmium, one expects the lattice to be more compressible along the $c$-axis than perpendicular to the $c$-axis. This indeed is the case. However, as shown in Fig. 2, $\chi_{11}$ increases with temperature continuously up to 575°K, while $\chi_{12}$ decreases with temperature. This is somewhat different from the temperature dependence of the linear compressibilities for zinc(6); for which $\chi_{12}$ remains constant. Only close to the melting point does $\chi_{12}$ decrease. Since $\chi_{12}$ involves the difference of two large quantities, the discrepancy between the temperature dependences of $\chi_{11}$ for cadmium and zinc may not be real. In any case, the fact that the temperature dependence for $\chi_{11}$ and $\chi_{12}$ are different suggests that the bonding between the atoms in the basal plane is certainly different from
that between the atoms of adjacent planes. As pointed out previously\((2,5)\),
these phenomena are consistent with the model proposed for zinc and cadmium
by Wallace\((26)\), who assumes a system of resonating covalent bonds between
atoms in the basal plane.

Having the values of \(\kappa_1\) and \(\kappa_\perp\), the values of \(\gamma\), the Grüneisen
parameter, were computed according to the following formula for hexagonal
solids:

\[
\gamma = \left(\frac{2 \beta_\perp + \beta_\parallel}{(2 \kappa_\perp + \kappa_\parallel)C_p}\right)^\frac{1}{4}
\]  

(7)

are plotted in Fig. 5. The terms \(\beta_\perp\) and \(\beta_\parallel\) in equation (7) are the
coefficients of thermal expansion perpendicular and parallel to the c-axis.
The values of \(\gamma\) in Fig. 5 from 300\(^\circ\)K down to 10\(^\circ\)K are taken from the work
of McCann and White\((29)\). The Grüneisen parameter \(\gamma\) decreases from 3.4
at 10\(^2\)K down to 2.17 at about 350\(^0\)K, and then increases above this temperature.
Since there is no theoretical model for predicting the values of \(\gamma\) for
anisotropic solids, not much can be said about the temperature dependence
of \(\gamma\) for cadmium.

Finally, we have evaluated the lattice contribution to the specific
heat from room temperature to 575\(^\circ\)K by substracting the dilational and
electronic specific heats from the experimentally determined values of \(C_p\).
The dilational term was evaluated by means of the thermodynamic relationship:

\[
C_p - C_v = \alpha \frac{2TV}{\gamma \gamma}
\]  

(8)

where \(\alpha = 2 \beta_\perp + \beta_\parallel\) is the volume coefficient of thermal expansion. The
electronic contribution, which is a small quantity, was assumed to vary
linearly with temperature. The value of the electronic coefficient,
\(\gamma_{el} = 1.62 \times 10^{-4}\) cal/deg\(^2\)g-atom was taken from the recent compilation
of Hultgren et. al.\((16)\). The values of the lattice specific heat so obtained
at high temperatures approach the classical value of 3\(R = 5.96\) cal/deg g-atom
within the uncertainties of all the experimental quantities involved.
Table I. Elastic Properties of Cadmium. The Adiabatic Elastic Constants and

Values of \( \rho v^2_{q1}(\phi = 22^\circ) \) are in the units of \( 10^{11} \text{ dyne/cm}^2; \)

The Shear Anisotropic Ratio, \( A = C_{66}/C_{44} \) is dimensionless and

Adiabatic Linear Compressibilities are in Units of \( 10^{-13} \text{ cm/dyne}. \)

<table>
<thead>
<tr>
<th>T, K</th>
<th>( C_{11} )</th>
<th>( C_{33} )</th>
<th>( C_{66} )</th>
<th>( C_{44} )</th>
<th>( \rho v^2_{q1} )</th>
<th>( C_{12} )</th>
<th>( C_{13} )</th>
<th>A</th>
<th>( \psi_{11} )</th>
<th>( \psi_{12} )</th>
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<td>300</td>
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<td>5.085</td>
<td>3.750</td>
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<td>4.960</td>
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<td>1.818</td>
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<td>1.94</td>
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<td>380</td>
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<td>1.95</td>
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<td>1.97</td>
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<td>3.96</td>
<td>1.92</td>
<td>19.2</td>
<td>1.92</td>
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Table II. Comparison of the Axial Ratio, c/a, the Adiabatic Elastic
Constant Ratios, C_{66}/C_{44} and C_{11}/C_{33}, and the Adiabatic
Linear Compressibility Ratio, \gamma_N/\gamma_L for Cadmium with Those
of Other Hexagonal Close-packed Metals

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<tr>
<th>Metals</th>
<th>c/a</th>
<th>C_{66}/C_{44}</th>
<th>C_{11}/C_{33}</th>
<th>\gamma_N/\gamma_L</th>
<th>Reference</th>
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<td>1.89</td>
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<td>6.3</td>
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<td>0.66</td>
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Figure Captions:

Fig. 1. The Adiabatic Elastic Constants of Cadmium as a Function of Temperature.

Fig. 2. The Adiabatic Linear Compressibilities of Cadmium as a Function of Temperature.

Fig. 3. The Grüneisen Parameter of Cadmium as a Function of Temperature.
Fig. 1

Elastic Constants x 10^-11 Dyne/cm²

- GARLAND AND SILVERMAN
- ABOWITZ
- GRÜNEISEN
- BRIDGMAN
- THIS STUDY

\[ \rho v^2 \]

\[ C_{11} \]

\[ C_{33} \]

\[ C_{66} \]

\[ C_{44} \]
Fig. 2

TEMPERATURE, °K

LIGBRAL AND SILVERMAN
THIS STUDY

\( \chi_{11} \)

\( \chi_1 \)
VALUES OF $\gamma$ BELOW 300°K - Mc CAMMON AND WHITE
VALUES OF $\gamma$ ABOVE 300°K - THIS STUDY

Fig. 3
REFERENCES


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