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THE TEMPERATURE DEPENDENCE OF THE ELASTIC CONSTANTS
OF Cu, Ag AND Au ABOVE ROOM TEMPERATURE

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ABSTRACT

The adiabatic elastic constants C_{44}, $1/2 (C_{11} - C_{12})$ and $1/2 (C_{11} + C_{12} + 2C_{44})$ have been measured for copper, silver and gold over the temperature range from 300° to about 800°K using the conventional ultrasonic pulse-echo technique. The room temperature values of the stiffness coefficients are shown to be in acceptable agreement with previously published data for the noble metals. Over the entire range from 300° to 800°K it is found that, to a remarkably good approximation, the elastic constants for all three metals decrease linearly with temperature. Additional evidence is presented to show that the linear temperature dependence of the elastic constants for silver extends to at least 1000°K, i.e. to within 0.8 of the absolute melting temperature. The isothermal compressibilities calculated from the elastic constant data are used to evaluate the dilational term in the specific heat, $C_{dil} = C_p - C_v$, and it is established that the approximate Nernst-Lindemann relation for estimating C_{dil} is valid for Cu, Ag, and Au at least up to 800°K.

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INTRODUCTION:

Although generally lacking, knowledge of the elastic constants of solids at temperatures approaching their melting points is of interest in several connections. For example, the isothermal compressibilities, derived from such data, make it possible to obtain an accurate estimate of the dilational or thermal expansion contribution to the specific heat. This dilational term, which increases rapidly above the Debye temperature, must be subtracted, along with the electronic specific heat, from the calorimetrically determined values of the specific heat at constant pressure in order to separate out the contribution due solely to lattice vibrations. Moreover, the high temperature elastic properties also enter in an important way into the theory of diffusion in cubic metals as Zener[1] and LeClaire[2] have shown, the magnitude of the pre-exponential factor, D_o, in the usual Arrhenius equation for the self diffusion coefficient is explicitly related to the temperature dependence of the stiffness moduli.

Since the elastic constants of the monovalent noble metals, copper, silver, and gold, have previously been determined only over the temperature range from 4.2° to 300°K, we felt it would be desirable to extend the measurements to substantially higher temperatures. In the present investigation, the adiabatic stiffness coefficients for all three metals have been determined from 300° to about 800°K using the conventional ultrasonic pulse-echo technique. Unfortunately, it was not possible to carry out measurements above 800°K because of deterioration of the epoxy resin which was used to cement the quartz transducers to the single crystal specimens. Even if a better high temperature bonding agent were available, the measurements would still have been limited to about 846°K - the temperature of the α-β transformation in quartz - using the techniques currently employed.

The elastic constant data obtained for Cu, Ag and Au between 300° and 800°K have subsequently been employed to evaluate the dilational contribution to the specific heat, i.e., $C_p - C_v$. The latter calculations, in turn, have permitted a fairly stringent test to be made of the semi-empirical Nernst-Lindemann relation[3], by means of which the dilational term is frequently approximated in the absence of the required compressibility data[4].

EXPERIMENTAL DETAILS

Copper, silver and gold single crystals in the form of 2.5 cm diameter spheres were grown under vacuum ($\sim 5 \times 10^{-6}$ Torr) in previously outgassed reactor grade graphite crucibles using a modified Bridgman technique. The nominal purities and sources of the starting materials were as follows: Cu, 99.999%, American Smelting and Refining Company; Ag, 99.9999%, oxygen-free grade, COMINCO Electronic Products, Inc.; Au, 99.99% Western Gold and Platinum Company. Specimens with faces normal to the <110> direction were then cut from each crystal by spark erosion. The orientations of these surfaces were determined by the standard back reflection Laue method and were adjusted to within 0.25 degree of <110>.

Inasmuch as the Laue reflections obtained directly from the spark eroded surfaces were found to be somewhat broad or diffuse, the crystals were first etched to remove the damaged surface layer resulting from the spark cutting operation; after etching (dilute HNO_3 for Cu and Ag, aqua regia for Au) sharp Laue reflections were observed. Each crystal was then mounted inside a surface ground hardened steel ring of appropriate thickness and polished with 3μ diamond paste on a vibratory lap (Syntron) until the two {110} faces were flat and parallel to within $\pm$ 0.0002 cm. Lapping in this manner preserved the sharpness of the Laue reflections, thus indicating that the specimen surfaces

were comparatively free of distortion. Two copper specimens of different
lengths were prepared as well as two of silver and one of gold. The lengths
of the resulting crystals, measured at 24°C were: Cu, 1.2040 and 1.6939 cm;
Ag, 1.2037 and 1.5786 cm; Au, 0.6175 cm.

For cubic crystals, three independent combinations of the elastic
constants C_{11}, C_{12} and C_{44} may be obtained by measuring the velocities of
plane longitudinal waves and appropriately polarized transverse waves propa-
gating the <110> direction. The three directly measured quantities are, in
the notation introduced by Zener[5], $C'_{11} = 1/2 (C_{11} + C_{12} + 2C_{44}) = \rho V_\ell^2$, $C = C_{44} = \rho V_{t_1}^2$ and $C' = 1/2 (C_{11} - C_{12}) = \rho V_{t_2}^2$, where ρ is the crystal density, V_ℓ is the
longitudinal wave velocity and V_{t_1} and V_{t_2} are the velocities of transverse
shear waves polarized in the <100> and <110> directions, respectively.
We have measured these sound wave velocities with a Sperry Products ultra-
sonic attenuation comparator[6]. Longitudinal waves were generated by 1/2-in.
dia. X-cut quartz crystal transducers and transverse waves by Y-cut crystals,
both resonant at 10 Mc/sec. The quartz transducers were bonded to the speci-
men surfaces with a special high-temperature epoxy resin consisting of Epon
1031 (Shell Development Co.) and nadic methyl anhydride.[*]

Experience showed that the most satisfactory results were obtained
when the epoxy resin was applied without the use of a supplementary catalyst.
After curing for about 2 hours at 150°C, the epoxy resin bond provided
excellent acoustic coupling between the quartz transducer and all three metals
at temperatures up to roughly 550°K. Above this temperature, the quality of
the bond varied depending upon the particular metal and, to some extent, on
the mode of propagation. The coupling, in general, was best for Cu and poorest

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[*]This material was kindly supplied by Dr. H. Chen, formerly of the Shell
Development Company, Emeryville, Calif. The exact proportions of the two
ingredients are unknown.

for Au, with silver being intermediate. A complete loss of coupling was frequently observed above about 550°K on initial warmup, as evidenced by a sudden drop in echo intensity. However, it was found that by raising the temperature to about 700°K and then slowly cooling back to 500° - 550°K, sufficient coupling could be restored to permit additional measurements to be made on reheating.

With Cu and Ag, the longitudinal and the two shear modes could all be propagated successfully up to 800°K. With gold, on the other hand, only the shear wave velocity, V_{t1}, corresponding to particle displacement in the $<100>$ direction could be measured to temperatures as high as 800°K; despite repeated efforts, measurements of the other two wave velocities could not extend above about 550°K because of the lack of sufficient acoustic coupling.

Measurements were made with the specimen mounted in a suitable holder inside a resistance heated furnace in which an atmosphere of dry argon was maintained. A flat copper plate, against which the crystal was held in good thermal and electrical contact, served as the specimen support. Furnace temperatures were controlled to $\pm 2°C$ and the temperature of the specimen was measured to within $\pm 1°C$ with a calibrated copper-constantan thermocouple inserted in a small hole in the copper plate. A more detailed description of the equipment and measuring techniques has been given elsewhere[7].

To estimate the magnitude of any possible transit-time corrections due to the finite thickness of the acoustic bond, a series of experiments were carried out at room temperature using specimens of different length as well as different bonding agents (e.g., "Nonaq" stopcock grease vs. epoxy resin). In all cases, the measured wave velocities agreed to within 0.1 %. This is well within the scatter of the elastic constant data as a function of temperature (See Figures 1 - 3).

The room temperature densities used in computing the elastic constants, viz., $\rho = 8.937$ g/cm^3 for Cu, 10.50 g/cm^3 for Ag and 19.30 g/cm^3 for Au were derived from the x-ray lattice spacings given by Pearson[8]. These densities, together with the measured sound velocities, were corrected for thermal expansion using the data of Nix and McNair[9] and Balluffi and Simmons[10].

RESULTS

The directly measured elastic constants $C = \rho V^2$ for Cu, Ag and Au are plotted as a function of temperature in Figures 1 - 3. The solid lines shown in these figures represent, in each case, the best least squares fit to the observed values. In Tables I-III we have listed the smoothed values of the adiabatic elastic constants C_{11}', C' and C taken from Figures 1 - 3 as well as the calculated stiffness coefficients C_{11} and C_{12}. Also included in these tables are the smoothed values of the adiabatic bulk moduli $B_s = 1/3 (C_{11} + 2C_{12}) = C_{11}' - C - C' 1/3$ and the isothermal compressibilities, χ_T, which have been calculated from B_s in the standard manner[3].

The number of significant figures displayed in Tables I-III is greater than is warranted by the absolute accuracy of the measurements, but an extra figure has been retained both to indicate the smoothness of the data and to permit a valid determination of the temperature coefficients; this is justified, to some extent, because the relative variation of the elastic constants with temperature has been established with somewhat better accuracy than have the absolute values. From the statistical spread or scatter in the experimental data shown in Figures 1 - 3, the uncertainties in the directly measured constants C_{11}', C and C' are found to be $\pm 0.5\%$. Using the standard propagation-of-error treatment,[11] the corresponding uncertainties in the calculated quantities are as follows: C_{11}, $\pm 0.7\%$, C_{12}, $\pm 0.9\%$; B_s, $\pm 0.9\%$.

Comparison with Previous Room Temperature Measurements

A detailed comparison between the results obtained at 300°K and room temperature elastic constant data for the noble metals previously reported in the literature is given in Table IV. The most accurate measurements for Cu are those of Overton and Gaffney[12] who also employed the ultrasonic pulse-echo technique. Agreement is seen to be quite good — within 0.3% for the shear constants C and C', 0.5% for C'_{11} and 0.7% for B_s. The present experimental values for Ag are likewise in good agreement with earlier measurements by Bacon and Smith[13], Neighbours and Alers[14] and Corll[15], the maximum percentage difference between any of the values listed in Table IV being only about 0.6%. For gold, however, the agreement is somewhat less good, especially for C' which is about 2% higher than Neighbours and Alers' value. The isothermal compressibilities computed from the adiabatic bulk moduli also compare favorably with previously published data for these metals and are within 2% of Bridgman's static room temperature values. Chang and Hultgren[7] have recently measured the longitudinal and transverse sound velocities in polycrystalline Cu over the temperature range 77° to 800°K; they report virtually the same temperature dependence of the compressibility as that from the present single crystal measurements.

Temperature Dependence of the Elastic Constants

The most noteworthy feature of the data recorded in Figs. 1 - 3 is that, within the uncertainty of the measurements, the elastic stiffness coefficients for all three metals decrease linearly with temperature over the entire range investigated. For further reference we have also plotted in Fig. 1 the elastic constants of Cu between 4.2° and 300°K taken from the work of Overton and Gaffney; similarly, the low temperature data of Neighbours and Alers for Ag

and Au are reproduced in Figs. 2 and 3, respectively. In general, by making

only slight relative shifts along the vertical axes, the least-squares lines which

describe the present high temperature measurements may be joined, without

any appreciable discontinuity in slope, to the corresponding low temperature

curves. This offers further support for the consistency of the data and also

indicates that the linear variation of the elastic constants holds to good approxi-

mation down to about 50°K for Au and Ag and nearly 100°K for Cu. These

temperatures roughly correspond to about one-third the Debye temperature

in each case.

Accurate modulus data which confirm and extend the observed linear

temperature dependence for silver have recently been obtained by Guinan[16].

Using the resonant bar technique, Guinan has measured the elastic moduli of

< 100 > - and < 111 > - oriented silver single crystals of both 6 - 9's pure and

oxygen saturated (~ 40 ppm) specimens over the range from 200° to 1000°K.

His data, to which small corrections have been applied for lateral inertia

effects are plotted as a function of temperature in Fig. 4. They demonstrate

that Young's moduli for silver continue to decrease with temperature in strictly

linear fashion at least up to 1000°K, and perhaps well beyond.

To provide a more succinct comparison with Guinan's measurements,

we have calculated the Young's moduli, E_{100} and E_{111}, for silver from our data

(Table II) as well as from the low temperature data of Neighbours and Alers.

The following standard relations between the moduli and the elastic constants

have been employed[17]:

$$\frac{1}{E_{100}} = \frac{1}{3} \quad \frac{1}{3B_s} + \frac{1}{C'}$$

(1)

and

$$\frac{1}{E_{111}} = \frac{1}{3} \quad \frac{1}{3B_s} + \frac{1}{C}$$

The resulting values are plotted in Fig. 4. It is clear from this figure that the calculated values of E_{100} and E_{111} and the temperature dependence of E_{100} and E_{111} from our data as well as those of Neighbours and Alers agree reasonably well with the directly measured values of Ag by Guinan within the experimental error.

We should also mention here that because of experimental limitations, most of the constants for gold above 550°K could only be arrived at by linear extrapolation. However, in view of the behavior exhibited by Cu and Ag, and the observed linear temperature dependence of C_{44} for Au over the full temperature range, this extrapolation can be carried out with complete confidence. Although we feel that these extrapolated values are probably reliable to within $\pm 1\%$, they are enclosed in parentheses in Table III to distinguish them from the experimentally measured quantities.

The thermal variation of the elastic constants near room temperature is summarized in Table V. Following the procedure adopted in the review by Huntington[18], we have listed in Table V the values of $1/C_{ij} (dC_{ij}/dT)$ at 300°K computed from the data given in Figs. 1 - 3 and Tables I - III. These temperature coefficients are expressed in parts per/10^6°K. The corresponding quantities deduced from the measurements of Overton and Gaffney and Neighbours and Alers are also presented in this table, as are the values of $1/E_{100}(dE_{100}/dT)$ and $1/E_{111} (dE_{111}/dT)$ for silver. The temperature coefficients

for the directly measured constants, C, C' and C_{11}' for Cu and Au are seen
to be in good accord with earlier results, but the thermal variation of C_{11}' and
C' for Ag is 10 - 15% greater than that found by Neighbours and Alers. This
accounts for the somewhat smaller temperature dependence of E_{100} calculated
from Neighbours and Alers' data. The excellent correspondence previously
noted between Guinan's measurements on silver and the temperature coefficients
of E_{100} and E_{111} derived from the present experiments is also demonstrated in
Table V.

Dilational Contribution to the Specific Heat

Knowing the isothermal compressibilities of Cu, Ag and Au between
room temperature and 800°K, (see Tables I - III) the dilational term in the
specific heat, C_{dil}, can be computed from the well-known thermodynamic
relation[3]

$$C_{dil} = C_p - C_v = \frac{9\beta^2 TV}{\chi_T} \tag{3}$$

where C_p and C_v are the specific heats at constant pressure and volume,
respectively, β is the linear coefficient of thermal expansion and V is the atomic
volume. In making these calculations, the necessary values of β have been
obtained from the thermal expansion data of Nix and MMcNair[9] and Balluffi
and Simmons[10], and the atomic volumes, corrected for thermal expansion,
have been computed from the lattice constants given by Pearson[8].

When compressibility data at high temperatures are lacking, as is
generally the case, C_{dil} is frequently approximated using the Nernst-Lindemann
relation

$$C_{dil} = AC_p^2 T \tag{4}$$

and taking the Nernst-Lindemann parameter

$$A = 9\beta^2 V / C_P^2 \chi_T \qquad (5)$$

to be constant independent of temperature. The validity of this assumption
may be tested with the aid of the data recorded in Tables I - III and the values
of Cp (T) for Cu, Ag and Au given in the recent compilation by Hultgren, et.al.[19]
We listed in Table VI, for example, the values of A for all three metals between
300° and 800°K calculated from Eq. (5) solely on the basis of experimentally
determined quantities. The essential constancy of A for Cu and Au over this
relatively wide temperature range is indeed apparent. For silver, the value
of A decreases roughly 8% between room temperature and 800°K. However,
in terms of specific heat, the error introduced using the value of A obtained
at room temperature amounts to only 1% of the total specific heat at 800°K.

DISCUSSION

The data presented above demonstrate that the linear temperature
dependence of the elastic constants for Ag holds remarkably well to tempera-
tures of at least 1000°K, or to within 0.8 of the absolute melting temperature.
The thermal variation of the elastic constants of Cu and Au appears qualitatively
similar in all aspects. Although the linear variation with temperature has been
established experimentally for these two metals only up to about 800°K, the
fact that they have higher melting points than Ag would seem to suggest that
the existing data for Cu and Au could safely be extrapolated in a linear fashion
to 0.8 Tm ≃ 1100°K. The strickly linear decrease in the stiffness coefficients
of Cu, Ag and Au with temperature indicates that the elastic stiffness coefficients
decrease in essentially a linear manner with thermal energy. Moreover, it

suggests that relaxation processes could have exerted but little influence on the measured moduli. If this were not so, a departure from linearity would have been observed, especially at high temperatures, and associated with the lowering of the true modulus.

Turning now to a consideration of the dilational specific heats, Gschneidner[4] has recently indicated that the constancy of the parameter A in Eq. (4) has been demonstrated experimentally over a wide temperature range for many metals. The basis for this statement was not documented, but the calculations summarized in Table VI demonstrate that it applies quite well to the noble metals at temperatures up to 800°K. In fact, the assumption that A is a constant independent of temperature is well enough supported experimentally to justify the use of the approximate Nernst-Lindemann relationship in estimating C_{dil} at temperatures considerably beyond those covered in this investigation. For this purpose it appears that a slightly more reliable estimate of C_{dil} between say 800°K and the melting point might be obtained by using the values of A at 800°K listed in Table VI instead of the room temperature values.

Some additional calculations not recorded here have been carried out. Using the approximate Nernst-Lindemann relation, Eq. (4), and assuming that the electronic specific heat varies linearly with temperature, we have evaluated the lattice contribution to the specific heats from room temperature to the respective melting points for all three metals. As might be anticipated, the lattice specific heats so obtained are in excellent agreement with the values predicted by the simple Debye continium model, and the Dulong and Petit value, $C_v = 3R = 5.96$ cal/deg-g-atom is approached within 0.04 cal/deg-g-atom at the melting point of Cu and Ag. For gold, the agreement is less good, but it

appears that either the dilational or the electronic contribution may have been overestimated since the calculated values of C_v decrease with temperature above about 900°K and fall about 0.20 cal/deg.g-atom below the Dulong-Petit value at the melting point.

Table I. Adiabatic Elastic Constants in Units of 10^{12} dyne/cm^2
and Isothermal Compressibility in Units of 10^{-13} cm^2/dyne
for Copper

T°K	C	C'	C_{11}'	C_{11}	C_{12}	B_s	χ_T
300	0.758	0.2355	2.220	1.700	1.225	1.385	7.44
350	0.745	0.2305	2.190	1.675	1.215	1.370	7.56
400	0.731	0.2250	2.165	1.655	1.205	1.355	7.69
450	0.718	0.2200	2.135	1.635	1.195	1.345	7.80
500	0.704	0.2145	2.105	1.615	1.185	1.330	7.93
550	0.691	0.2095	2.075	1.595	1.175	1.315	8.06
600	0.677	0.2045	2.045	1.575	1.165	1.300	8.19
650	0.663	0.1990	2.020	1.555	1.155	1.290	8.33
700	0.650	0.1940	1.990	1.535	1.145	1.275	8.47
750	0.636	0.1885	1.960	1.515	1.135	1.260	8.63
800	0.623	0.1835	1.930	1.495	1.125	1.250	8.77

Table II. Adiabatic Elastic Constants in Units of 10^{12} dyne/cm^2 and Isothermal Compressibility in Units of 10^{-13} cm^2/dyne for Silver

T°K	C	C'	C'_{11}	C_{11}	C_{12}	B_s	χ_T
300	0.465	0.1525	1.555	1.240	0.940	1.040	10.00
350	0.455	0.1480	1.530	1.225	0.930	1.025	10.20
400	0.446	0.1440	1.510	1.205	0.920	1.015	10.40
450	0.436	0.1400	1.485	1.190	0.910	1.005	10.60
500	0.426	0.1360	1.460	1.170	0.900	0.990	10.80
550	0.417	0.1315	1.440	1.155	0.890	0.980	11.05
600	0.407	0.1275	1.415	1.135	0.880	0.965	11.25
650	0.398	0.1235	1.395	1.120	0.870	0.955	11.50
700	0.388	0.1195	1.370	1.100	0.860	0.940	11.75
750	0.379	0.1155	1.345	1.085	0.855	0.930	12.00
800	0.369	0.1110	1.325	1.065	0.845	0.915	12.25

Table III. Adiabatic Elastic Constants in Units of 10^{12} dyne/cm^2 and Isothermal Compressibility in Units of 10^{-13} cm^2/dyne for Gold

T°K	C	C'	C_{11}'	C_{11}	C_{12}	B_s	χ_T
300	0.424	0.1485	2.200	1.925	1.630	1.725	6.01
350	0.417	0.1460	2.180	1.910	1.615	1.705	6.10
400	0.410	0.1435	2.155	1.890	1.605	1.700	6.19
450	0.403	0.1410	2.135	1.875	1.590	1.685	6.28
500	0.397	0.1385	2.115	1.850	1.580	1.670	6.38
550	0.390	0.1360	2.090	1.840	1.565	1.655	6.48
600	0.383	0.1335	2.070	1.820	1.555	1.645	6.58
650	0.376	0.1310	2.050	1.805	1.540	1.630	6.69
700	0.369	0.1285	2.025	1.785	1.530	1.620	6.80
750	0.362	0.1260	2.005	1.770	1.520	1.600	6.90
800	0.355	0.1235	1.985	1.755	1.505	1.580	7.06

Table IV. Comparison of Present Results with Literature Data at 300°K

Metal	Investigators	$G \times 10^{-12}$	$G' \times 10^{-12}$	$B_S \times 10^{-12}$	$\chi_T \times 10^{13}$
Cu	This Study	0.756	0.235	1.38	7.49
	Schmunk & Smith[20]	0.7489	0.2338	1.395	
	Overton & Gaffney[12]	0.7539	0.2348	1.371	
	Goens & Weerts[21]	0.753	0.263	1.38	
	Lazarus[22]	0.756	0.235	1.40	
	Bridgman[4]				7.64
	Chang & Hultgren[7]				7.42
Ag	This Study	0.464	0.152	1.04	9.98
	Bacon & Smith[13]	0.4613	0.1528	1.036	
	Neighbours & Alers[14]	0.4612	0.1516	1.038	
	Corll[15]	0.4632	0.1527	1.038	
	Bridgman[4]				9.93
Au	This Study	0.424	0.149	1.73	6.01
	Neighbours & Alers[14]	0.4195	0.1460	1.729	
	Bridgman[4]				5.78

Table V. Thermal Variation of Elastic Constants of Cu, Ag, and Au

Expressed by $-\dfrac{1}{C_{ij}}\dfrac{dC_{ij}}{d_T}$ in parts $/10^{6}$ °C at 300°K.

Metal	Investigator	$-TC$	$-TC'$	$-TC'_{11}$	$-TE_{100}$	$-TE_{111}$
Cu	This Study	360	440	260	-	-
	Overton & Gaffney	370	425	240	-	-
Ag	This Study	410	540	295	515	380
	Neighbours & Alers	400	470	260	455	375
	Guinan, oxygen free	-	-	-	540	390
	Guinan, oxygen sat'd	-	-	-	-	380
Au	This Study	325	340	195	-	-
	Neighbours & Alers	300	340	190	-	-

Table VI. Values of Lindemann Parameter

| T, °K	$A \times 10^{-5}$ in g-atom/cal		
	Cu	Ag	Au
300	1.67	2.21	1.97
400	1.67	2.16	1.97
500	1.67	2.15	1.97
600	1.67	2.08	1.95
700	1.64	2.06	1.95
800	1.61	2.05	1.92

FIGURE CAPTIONS

Figure 1.   Elastic Constants of Cu as a Function of Temperature

Figure 2.   Elastic Constants of Ag as a Function of Temperature

Figure 3.   Elastic Constants of Au as a Function of Temperature

Figure 4.   The Young's Moduli in the Directions $<111>$ and $<100>$ for
Ag as a Function of Temperature.

Fig. I

Fig. 2

Fig 3

Fig. 4.
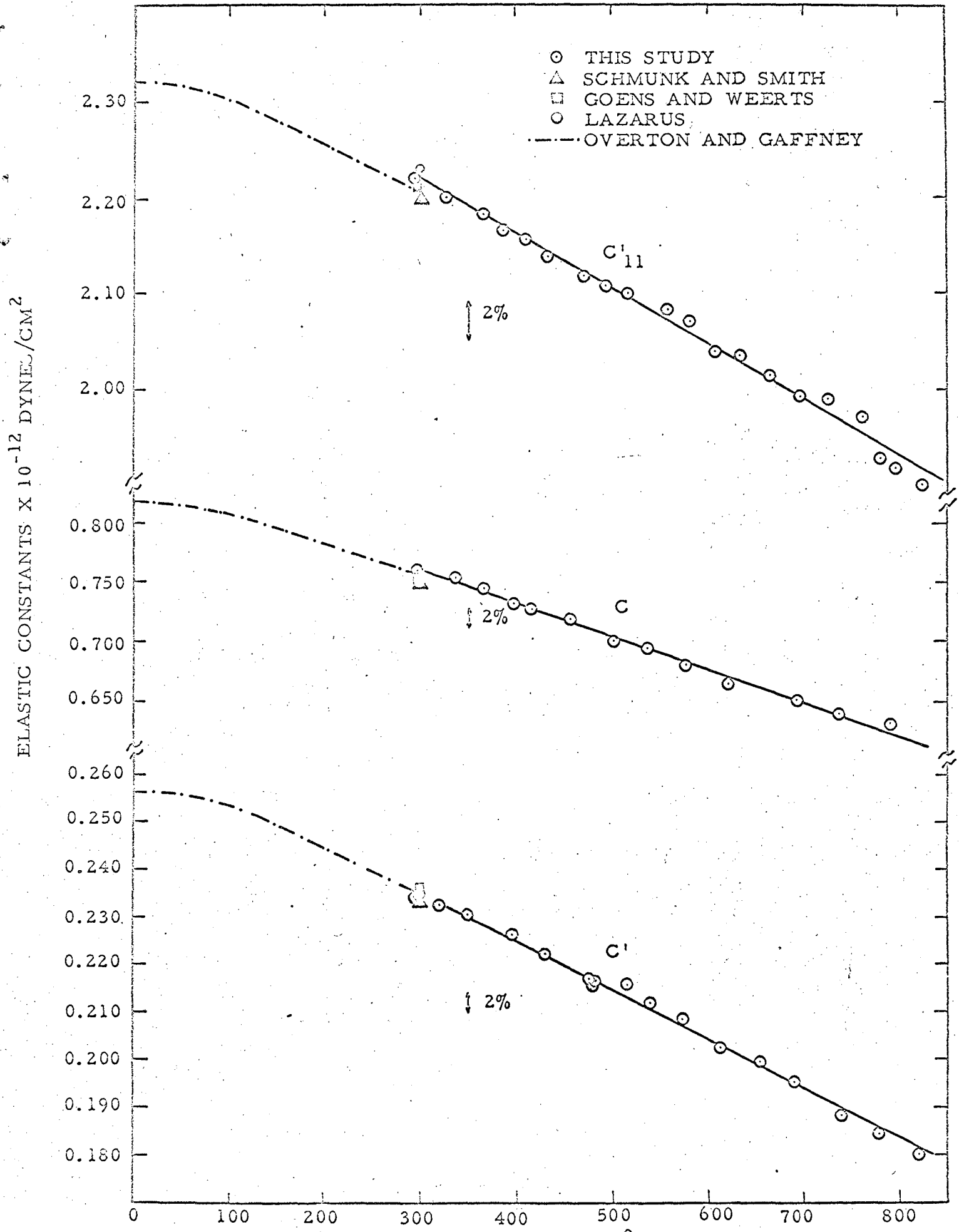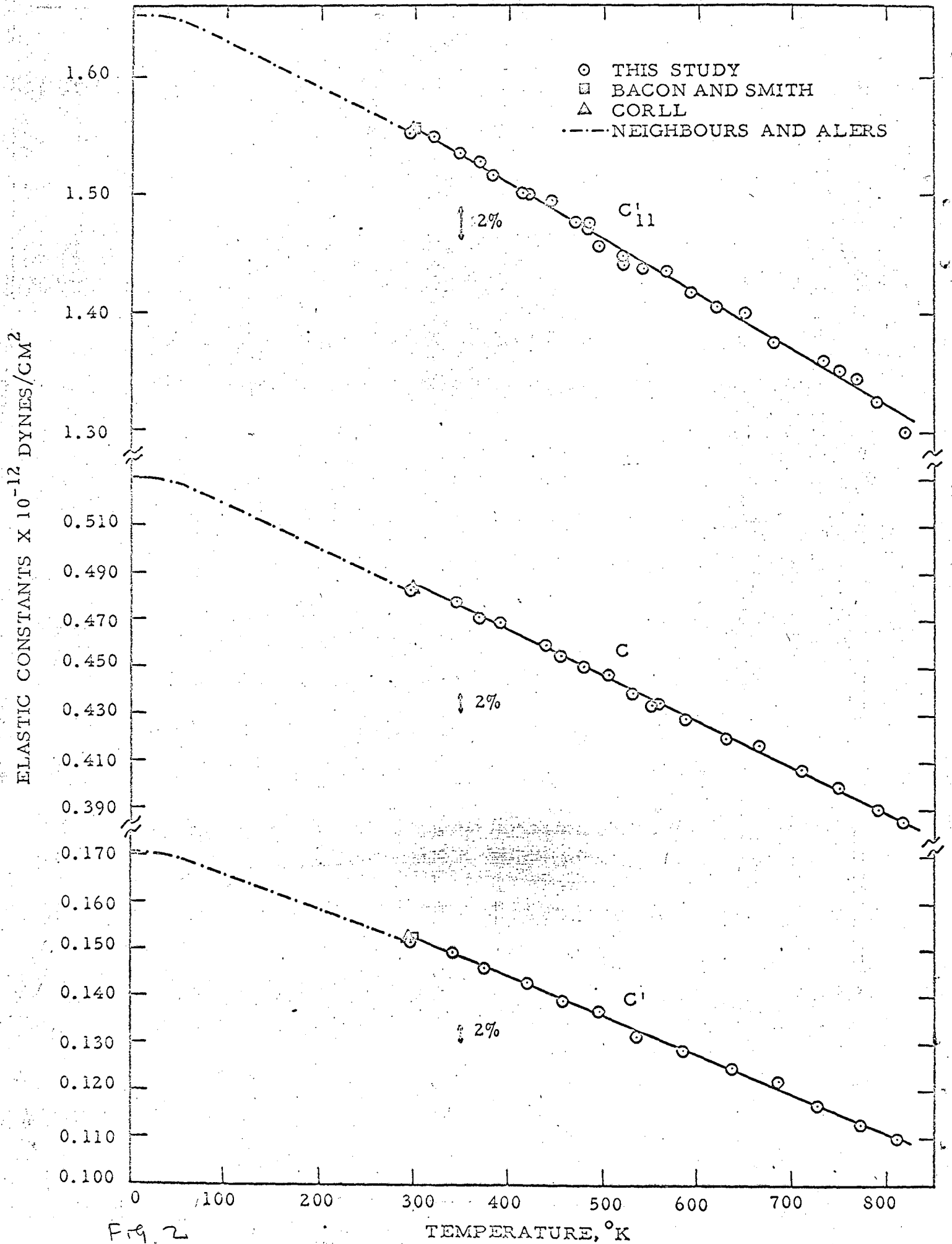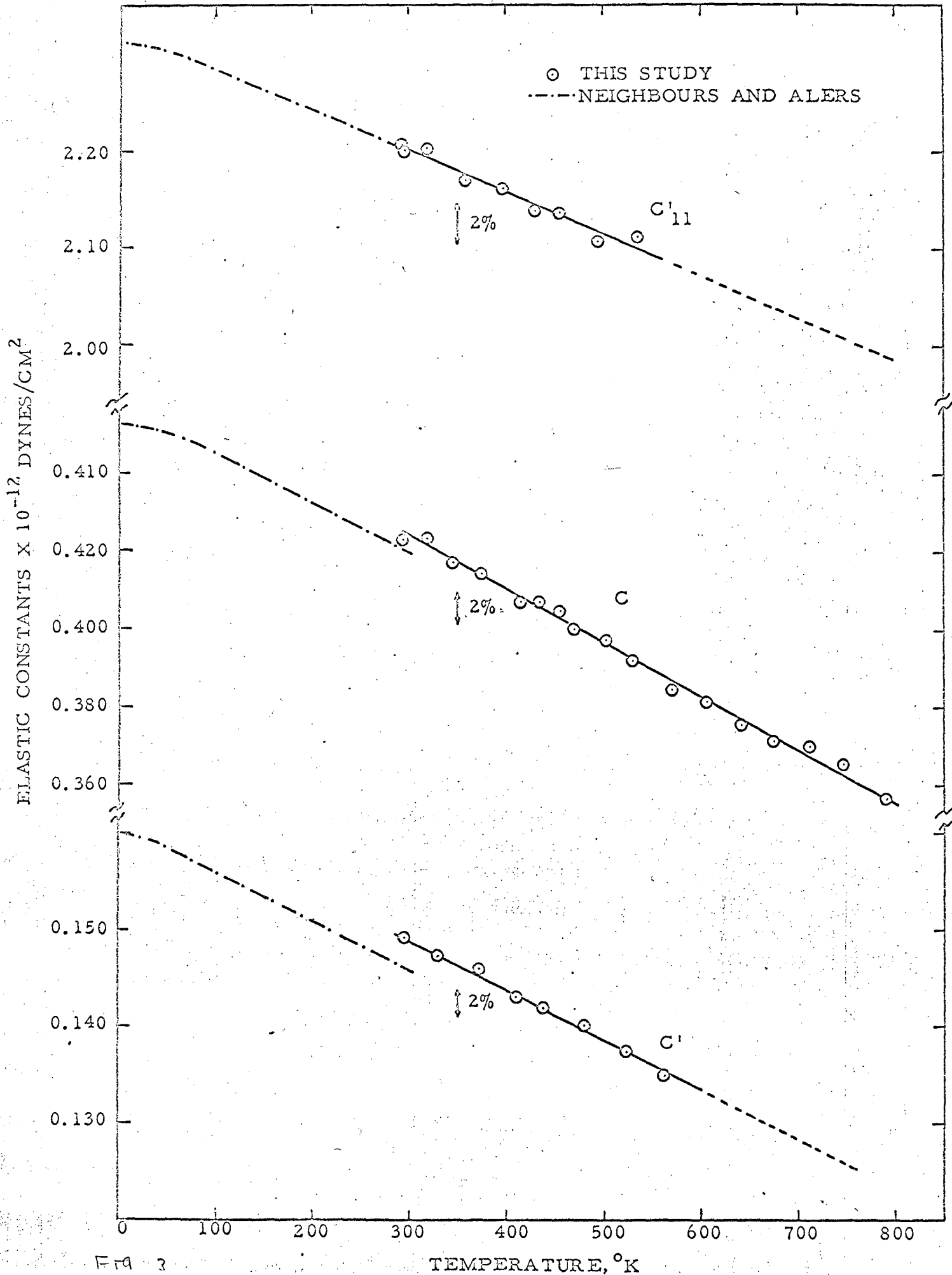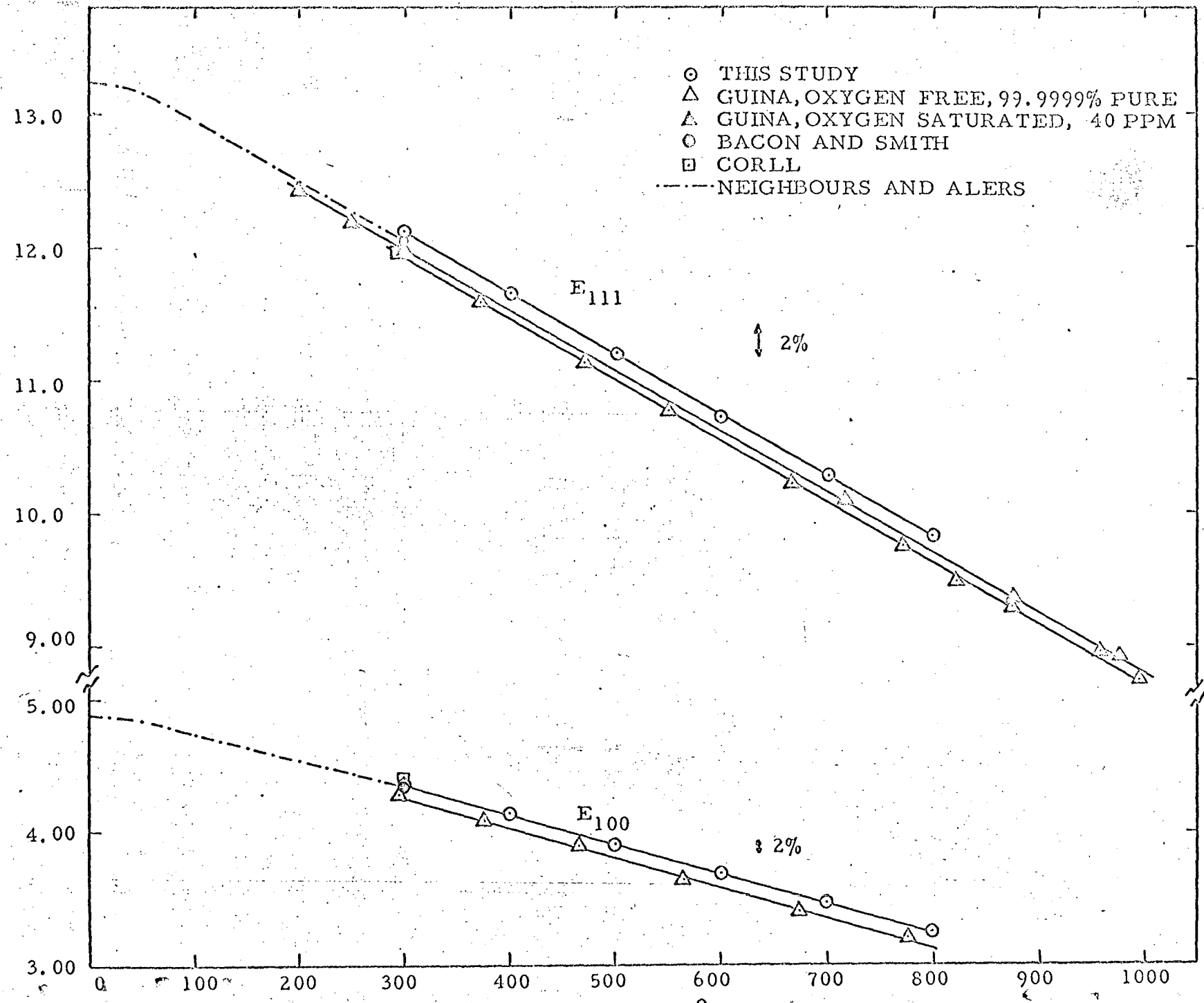
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