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Elastic constants of silver as a function of temperature

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The temperature dependence of the elastic constants of silver single crystals has been determined over the range 300–1173 K with the piezoelectric ultrasonic composite oscillator technique (PUCOT). From a comparison of the present results with those available from the literature, it is deduced that the PUCOT and hence other standing-wave techniques are adequate for measuring compliances, but these techniques may have complications for computations of stiffnesses.

1. Introduction

Knowledge of the elastic constants of single crystals over a wide range of temperature is of fundamental importance in characterizing many mechanical, physical, optical and thermal properties. In the case of silver single crystals, the elastic constants have been measured by Hearmon [1], Neighbours and Alers [2] and Biswas *et al.* [3] at room temperature, and by Chang and Himmel [4] at elevated temperatures. As is expected when different investigators use different techniques to measure elastic constants, discrepancies among the results exist. An examination of the literature indicates that the discrepancies are largely due to the methods used for measuring the elastic constants and the methods of interpreting the raw data.

The early stages of measuring elastic constants of solids relied entirely on quasi-static methods. The emergence of the dynamic methods in the mid 1920s led investigators to do away with static methods. Dynamic methods, such as wave propagation and resonant standing-wave techniques, are superior to quasi-static ones in terms of simplicity, relative ease of operation and reproducibility of experimental results. Unfortunately, they suffer from the fact that some materials tend to exhibit a non-linear response. This is especially true for resonant systems. This complicates the interpretation of the spectrum of resonant frequencies observed with any specimen. A further complication is the tendency of some specimens to exhibit subresonance and multiresonance of vibration during forced vibrations.

In 1976, Robinson and co-workers [5, 6] used the method known as the piezoelectric ultrasonic composite oscillator technique (PUCOT) to measure elastic constants. This technique has been used by relatively few investigators, and none has addressed the complications just mentioned. In this paper on silver single crystals, we show that the PUCOT may be used to measure the compliances of single crystals, just like other standing-wave methods, but that the success of the PUCOT, and hence of all standing-wave techniques, in measuring the compliances is not translated into success in computing the stiffnesses. In particular, it is shown that inverting compliances to obtain stiffnesses leads to error propagation. Elastic constants data are presented for silver single crystals for the temperature range 300–1173 K.

2. Experimental procedure

Single crystals of silver in the form of right circular cylinders were grown by the Bridgman technique. Spectrographic analyses of the specimens indicated that iron was the main impurity (0.02 wt %), other elements being below the resolution limits (usually 0.01 wt %). Other preparation techniques for the specimens are described elsewhere [7]. The crystallographic orientations of the specimens were determined by the Laue X-ray back-reflection method and are presented in terms of the direction cosines α, β and γ (with respect to the [1 0 0] direction) in Table I. Also in the table are values of the orientation factor κ $(=\alpha^2\beta^2+\beta^2\gamma^2+\gamma^2\alpha^2)$ and the parameter χ $(=\alpha^2\beta^2\gamma^2)$. The PUCOT was used as described elsewhere [8] to measure values of Young's modulus (E_p) and shear modulus (G_p) as a function of temperature up to 1173 K. The subscript p denotes that we are referring to perturbed deformations (as opposed to free deformations) in which coupling between lateral and longitudinal displacements occurs in the longitudinal mode, and coupling between bending and torsion occurs in the torsional mode [7].

3. Results and discussion

The elastic constants for silver single crystals were measured at temperatures in the range 300–1173 K with the PUCOT. The values of E_p and G_p for five specimens are presented in graphical form in Fig. 1. The data points for Young's modulus appear to fit smooth curves, but the data point for the shear modulus at 473 K appears to be somewhat out of line. Thus, second degree polynomial curves were fitted to the data sets for the Young's modulus, while a first degree polynomial curve was fitted to the set of data for $T \geqslant 673$ K for the shear modulus. The correlations are

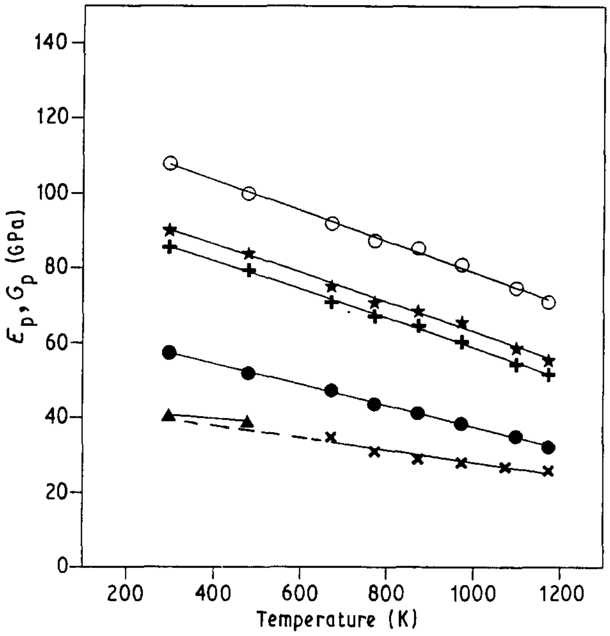

Figure 1 Plots of the measured values of Young's modulus, E_p, and shear modulus, G_p, as a function of temperature for silver single crystals, showing the apparent discontinuity for the plot of G_p at 473 K. (●) E_1, (+) E_2, (★) E_3, (○) E_4, (×) G_1, (▲) G.

$$E_1 = 65.30 - 0.026(T) - 1.38\times10^{-6}(T^2) \tag{1}$$
$$E_2 = 96.23 - 0.034(T) - 3.12\times10^{-6}(T^2) \tag{2}$$
$$E_3 = 101.19 - 0.036(T) - 2.39\times10^{-6}(T^2) \tag{3}$$
$$E_4 = 119.64 - 0.040(T) - 1.03\times10^{-6}(T^2) \tag{4}$$
$$G_1 = 44.47 - 0.016(T) \tag{5}$$

where T is in Kelvin and E_p and G_p are in GPa.

The curve for G_p shows a discontinuity between 473 and 673 K which is not associated with any physical phenomenon. Because the data points above 673 K appear to behave in a normal fashion, it was felt best to ignore the first two data points and fit the curve without them. Comparison of the extrapolated value of G_p at 300 K with the expected value showed that this treatment of the data was valid.

The eight-step method [7] of computing the elastic constants from the measured values of E_p and G_p was used to obtain the data listed in Table II. The data from the table are plotted in Figs 2 and 3 for the compliances and stiffnesses, respectively.

As can be seen from Fig. 2, the compliances increase with increasing temperature showing curvature typical of cubic materials. Fig. 3 indicates expected behaviour for the stiffnesses, i.e. they decrease with increasing temperature and show slight curvature at about 800 K. This curvature is also revealed in the measured values of Young's modulus and is associated with elastic softening of the cubic lattice [9].

TABLE I Orientation data for the silver single crystals

ID	α	β	γ	κ	χ
Ag-1	0.968147	0.174685	0.177938	0.059245	0.000906
Ag-2	0.913545	0.355348	0.175360	0.134929	0.003240
Ag-3	0.596747	0.762683	0.237854	0.260197	0.011719
Ag-4	0.738777	0.624114	0.259021	0.275347	0.014260
Ag-5	0.703336	0.535933	0.468654	0.313819	0.031207

TABLE II The compliances S_{ij} and stiffnesses C_{ij} for silver as a function of temperature compared with the data of Chang and Himmel [4]

Reference	T(K)	S_{11} (10^{-11} m^{2}N^{-1})	S_{12} (10^{-11} m^{2}N^{-1})	S_{44} (10^{-11} m^{2}N^{-1})	C_{11} (10^{-11} Nm^{-2})	C_{12} (10^{-11} Nm^{-2})	C_{44} (10^{-11} Nm^{-2})	$C_{11} - C_{12}$ (10^{-11} Nm^{-2})
Present work	300	2.3564	1.0124	2.1830	1.2032	0.9064	0.4581	0.2968
[4]		2.3291	1.0043	2.1505	1.2400	0.9400	0.4650	0.3000
Present work	400	2.4812	1.0738	2.2748	1.1869	0.9057	0.4396	0.2813
[4]		2.4487	1.0601	2.2422	1.2050	0.9200	0.4460	0.2850
Present work	500	2.6212	1.1426	2.3746	1.1694	0.9038	0.4211	0.2657
[4]		2.5814	1.1223	2.3474	1.1700	0.9000	0.4260	0.2700
Present work	600	2.7794	1.2203	2.4833	1.1506	0.9005	0.4027	0.2500
[4]		2.7743	1.1920	2.4570	1.1350	0.8800	0.4070	0.2550
Present work	700	2.9596	1.3087	2.6022	1.1302	0.8959	0.3843	0.2343
[4]		2.8960	1.2707	2.5773	1.1000	0.8600	0.3880	0.2400
Present work	800	3.1664	1.4100	2.7327	1.1082	0.8897	0.3659	0.2185
[4]		3.1513	1.3942	2.7100	1.0650	0.8450	0.3690	0.2200
Present work	900	3.4061	1.5275	2.8767	1.0845	0.8818	0.3476	0.2027
	1000	3.6869	1.6652	3.0364	1.0592	0.8724	0.3293	0.1868
	1100	4.0204	1.8287	3.2143	1.0324	0.8615	0.3111	0.1709
	1173	4.3057	1.9689	3.3575	1.0121	0.8527	0.2978	0.1594

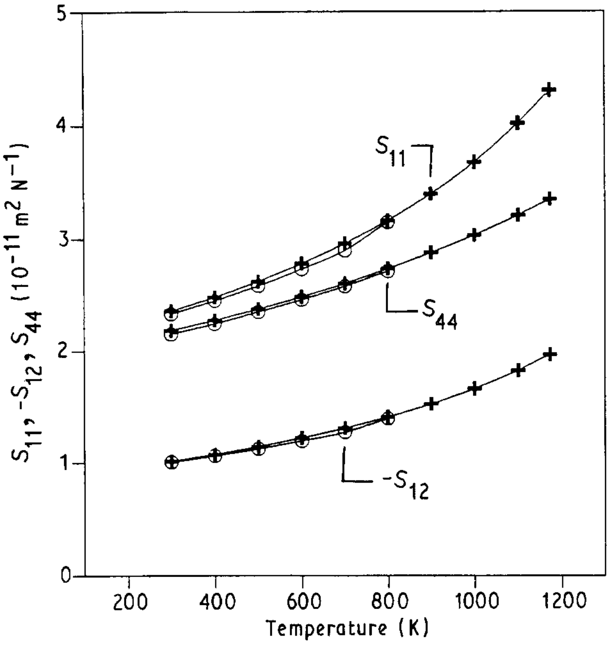

Figure 2 (+) Plots of the compliances S_{ij} for silver single crystals as a function of temperature showing curvature typical of cubic materials. (○) The results of Chang and Himmel [4] shown for comparison.

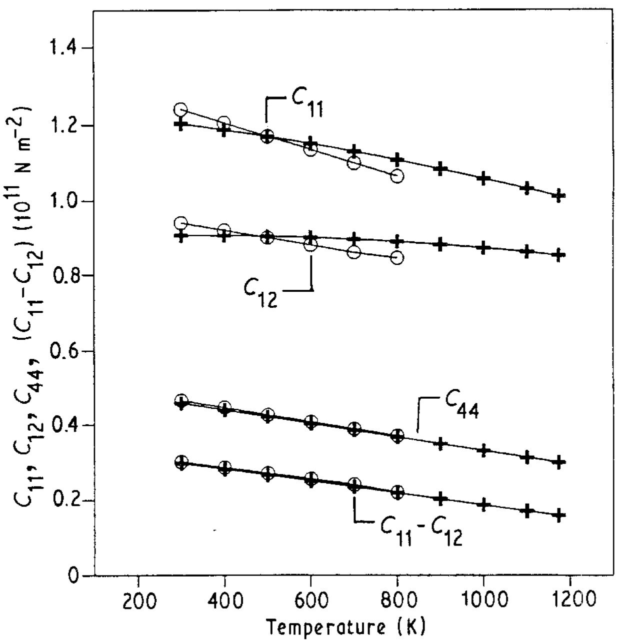

Figure 3 (+) Plots of the stiffnesses C_{ij} for silver single crystals as a function of temperature showing a small curvature near 800 K associated with elastic softening of the crystal lattice. (○) The data of Chang and Himmel [4] shown for comparison.

Comparison of the present results with those of Chang and Himmel [4] reveals that the compliances, the stiffness C_{44} and the shear modulus, $(C_{11} - C_{11})/2$, agree with one another in a satisfactory manner. The same satisfactory agreement is not apparent, however, for the stiffnesses C_{11} and C_{12}. Chang and Himmel obtained their values of stiffness directly by means of the wave-transmission method. Therefore, their results are believed to be more accurate than the present results, which were obtained by a resonance method (PUCOT). Once again, the method of using standing waves to measure elastic moduli fails to describe the elastic constants comprehensively because of error propagation in inverting compliances to stiffnesses [7].

It is useful to compare the stiffnesses computed from the compliances listed in Table II at about 300 K with those given elsewhere [7]. While the two sets of compliances are in good agreement (within 1%), the stiffnesses C_{11} and C_{12} deviate from each other by about 5%. This shows how sensitive the stiffnesses are to minor changes in the absolute values of the compliances S_{11} and S_{12}. It is also interesting to note that Biswas *et al.* [3] and Klooster *et al.* [10] employed standing waves to measure the compliance S_{44} and the quantity $(S_{11} - S_{12})/2$, and therefore the stiffness C_{44} and the modulus $(C_{11} - C_{12})$, but not the compliances S_{11} and S_{12} (from which the stiffnesses C_{11} and C_{12} are computed). Instead, they measured the stiffness C_{11} directly by a wave-transmission method for a crystal whose value of κ was zero. This indicates that even when specific orientations ($\kappa = 0$, 1/4 and 1/3) are employed, methods using standing wave are inadequate to characterize fully the elastic constants. This is one way, and perhaps the only way, to avoid inverting compliances to stiffnesses because C_{12} can be determined readily from the quantity $(C_{11} - C_{12})/2$ measured by means of standing waves.

Finally, it is mentioned that standing-wave techniques offer many advantages over wave-transmission methods because of the flexibility in dimensions and orientations of the single crystals. This is certainly true for the PUCOT. It offers flexibility in dimensions and operational procedures (frequency range 20–200 kHz, strain amplitude range 10^{-8}–10^{-4}, and temperature range 78–1700 K) unmatched by any other technique. The optimum use of the PUCOT, therefore, may be in complementing wave-transmission methods for measurements of the elastic constants of single crystals.

4. Conclusions

From these measurements with the PUCOT of the elastic constants of silver single crystals over the temperature range 300–1173 K, and a comparison of the results with data from the literature, the following conclusions can be drawn.

1. The present values of the compliances, the stiffness C_{44} and the shear modulus $(C_{11} - C_{12})/2$ agree with those of Chang and Himmel within about 2%.
2. The present values of the stiffnesses C_{11} and C_{12} agree with those of Chang and Himmel within about 5%.
3. The method of using standing waves fails to describe the elastic constants comprehensively because of error propagation in inverting compliances to stiffnesses.
4. Optimum use of the PUCOT in determining the elastic constants of single crystals may be found in supporting or complementing wave-transmission methods.

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