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X-ray diffraction studies and thermal and elastic properties of $2H$-TaSe$_2$

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The pressure dependences of the lattice parameters a and c of $2H$-TaSe$_2$ are determined by x-ray diffraction techniques for $p \leq 4.4$ GPa. In addition, linear thermal expansions are also determined by x-ray diffraction techniques for $10 \leq T \leq 300$ K. The initial ($p = o$) linear compressibilities are obtained and combined with known (zero-sound) elastic moduli (based on neutron-diffraction measurements), primarily to determine c_{13}; resulting values of c_{13} are in the range $-2-+2$ (10^{11} dyn/cm^2). Grüneisen parameters and properties of the normal-state to incommensurate-charge-density-wave transition are considered on the basis of our results. It is also shown that, as for $2H$-NbSe$_2$, the room-temperature elastic moduli yield too small a value for the T^3 contribution to the low-temperature specific heat.

I. INTRODUCTION

Several layered transition-metal dichalcogenides, including intercalated materials, exhibit charge-density wave and/or superconducting transitions, and some attention has been given[1-6] to the interrelationships among these transitions and structural parameters, such as the interlayer distance and the lattice parameter within the layer planes. A knowledge of the second-order elastic moduli is needed to convert stress dependences of phase-transitional temperatures, of which some information is available,[1-3,7] to strain dependences. Ultrasonic measurements on these materials have not been performed, presumably because of the small crystal sizes (especially along the c axis), so it is necessary at present to consider less accurate means for determining the elastic moduli of these materials.

Previously, some of the elastic moduli of the $2H$ forms of MoS$_2$, NbSe$_2$, and TaSe$_2$ were obtained[8,9] from the acoustical branches (near $q \sim 0$) of the neutron-measured[9,10] dispersion curves. X-ray measurements of the linear compressibilities [for NbSe$_2$ and MoS$_2$ (Refs. 4 and 11)] and a vibrating-reed measurement of the compliance modulus s_{11} [for TaSe$_2$ (Ref. 12)] have been combined[2,8] with the neutron measurments to estimate c_{13} for these materials and, in the case of MoS$_2$, c_{12}. However, the use of s_{11} yields only $|c_{13}|$, and the magnitude of s_{11} is not known accurately. Furthermore, it should be noted that these analyses have ignored anharmonic and possible nonadiabatic electronic effects in the sense that no distinction has been made among zero-sound (neutron), isothermal (x-ray), and adiabatic (vibrating-reed) measurements of the elastic moduli, where it is presumed that the neutron results correspond to frequencies high enough so that phonon-lifetime effects can be ignored.

In this paper on $2H$-TaSe$_2$, x-ray data of the variation of the lattice parameters are presented as both a function of pressure at room temperature and a function of temperature at ambient pressure. In the former case, powder measurements were made for several pressures in the range 1.06–4.4 GPa (1 GPa = 10 kbars), as well as at ambient pressure. Extracted initial ($p = 0$) linear compressibilities are combined with the values of the elastic moduli c_{11}, c_{12}, and c_{33} determined from the neutron-measured dispersion curves,[9] to estimate c_{13} and to check the self-consistency of the various measurements. Our thermal-expansion data and elastic moduli are used in conjunction with previous specific heat[13,14] and other related measurements to estimate (a) anisotropic Grüneisen parameters (b) adiabatic-isothermal compliance moduli corrections, (c) Θ^{el} (equivalent to the low-temperature specific-heat Debye temperature, provided the elastic moduli are independent of temperature), (d) the jump in c_{11} at the normal-state to incommensurate-charge-density-wave (ICDW) transition, and (e) strain derivatives of the ICDW transition temperature T_0, which has been estimated[2] on the basis of a different value of c_{13} than that obtained in this work. We note that the adiabatic-isothermal corrections were considered mainly for the purpose of assessing the validity of our analysis. The fact that we have found these corrections to be negligibly small suggests that anharmonic effects are not likely to give rise to significant differences between zero-

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sound and isothermal elastic moduli, although we cannot comment on possible nonadiabatic electronic effects in this regard.

II. EXPERIMENTAL DETAILS

Powder samples of $2H$-TaSe$_2$, made by Di Salvo of Bell Labs, were used for both the linear-compressibility and thermal-expansion measurements. The latter were carried out over the range $30 < T < 300$ K. Additionally, a single 2 mm × 2 mm × ~0.5 mm crystal of $2H$-TaSe$_2$, obtained from Morris of Florida State University, was used to measure the thermal expansion normal to the layers, i.e., parallel to the c axis over the range $10 < T < 300$ K.

Hydrostatic pressures were obtained in two gasketed diamond anvil pressure cells. CsCl and NaCl, mixed with the sample, were used interchangeably as a pressure calibrant according to standard procedures.[15] Zr-filtered Mo radiation was employed and the d spacings of the following TaSe$_2$ diffraction lines were measured at most pressures: (002), (100), (101), (102), (103), (104), (110), and (112). [For several pressures the (100) and (101) lines were not considered.] The films were read on a semiautomated microdensitometer having an instrumental uncertainty of 1 μm. An (006) line was observed but, because of its reduced intensity, the uncertainty in its position was excessive, and it was not used in the analysis. At each pressure ranges of values were obtained, for both a and c, for which it was possible to fit all measured d spacings to within their estimated experimental uncertainties. [However, since the (002) reflection, which is most sensitive to "c" of the reflections considered, was not observed at $p = 3.6$ GPa, no values of c will be quoted for that pressure.] Our ambient-pressure results agree with those of Bjerkelund and Kjekshus[16] to within our estimated uncertainties.

Separate x-ray apparatus was employed in our thermal-expansion measurements. Here, Cu radiation was used after filtering through Ni, and differences (from room-temperature values) in d spacings were considered. For the same powder sample used in the pressure measurements, the diffraction lines (1 0 14), (2 0 12), (220), and (222) were examined, and pairs of values of $\Delta d/d$ corresponding to pairs of diffraction lines [(1 0 14), (220)], [(1 0 14), (222)], and [(2 0 12), (222)] were used to evaluate $\Delta c/c$ and $\Delta a/a$. Polycrystalline thermal-expansion results, given in Sec. III represent averages of these determinations. Because of the sample size and limitations of the available x-ray equipment, out study of the single-crystal sample was restricted to the (0 0 16) reflection, which yielded a determination of $\Delta c/c$ only.

III. EXPERIMENTAL RESULTS AND DISCUSSION

The measured values of $a(P)$ and $c(P)$ are shown in Fig. 1, where the solid lines, drawn appropriately through the highly accurate ambient-pressure x-ray results of Bjerkelund and Kjekshus,[16] correspond to estimated extremes of the initial compressibilities based on our data. Clearly, the possibility of a considerable nonlinear pressure dependence (in the pressure range studied) of the relatively compressible c axis was allowed for in these estimates. Values of the resulting initial compressibilities k_a and k_c are shown, in Table I, to be quite similar to those obtained for $2H$-MoS$_2$ and $2H$-NbSe$_2$. Markedly lower values of both k_a and k_c were obtained[4] in the case of NbS$_2$ (presumably the $3R$ polytype).

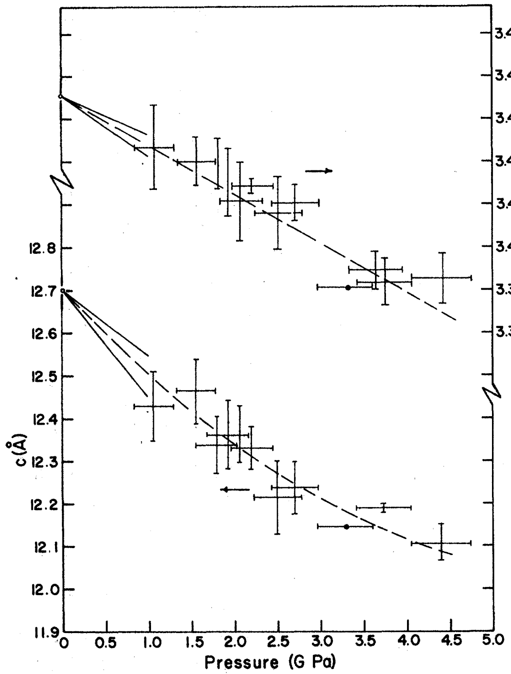

FIG. 1. Pressure dependence of lattice parameters of $2H$-TaSe$_2$. The ambient-pressure results (open circles) are those of Bjerkelund and Kjekshus (Ref. 15). The lack of vertical bars at $p = 3.3$ GPa is indicative of the marginal self-consistency of the d-spacing measurements at that pressure. The dashed lines were drawn through the data as a guide, and their slopes at low pressure correspond to the cited values of k_a and k_c in Table I. The solid lines represent estimated extreme values of the initial compressibilities.

TABLE I. Compressibilities of 2*H* transition-metal dichalcogenides.

Compound	Pressure range of experiment (GPa)	Sample type	k_a (10^{-13} cm^2/dyn)	k_c (10^{-13} cm^2/dyn)
$NbSe_2$(Ref. 4)	0–0.31	Single crystal	4.1 ± 0.5	16.2 ± 0.5
$TaSe_2$	0–4.4	Powder	3.3 ± 0.7	16 ± 4
MoS_2(Ref. 11)[a]	0–4.2	Powder	3.4 ± 0.1	16.4 ± 0.3

[a] A polynomial fit to the *c*-axis data was deemed necessary; the quoted k_c corresponds to the low-pressure limiting value of the slope of *c* vs *p*. Also evident in this work (Ref. 11) is the poor (~1%) agreement between the measured ambient-pressure lattice parameter *a* and its accepted value of 3.16 Å.

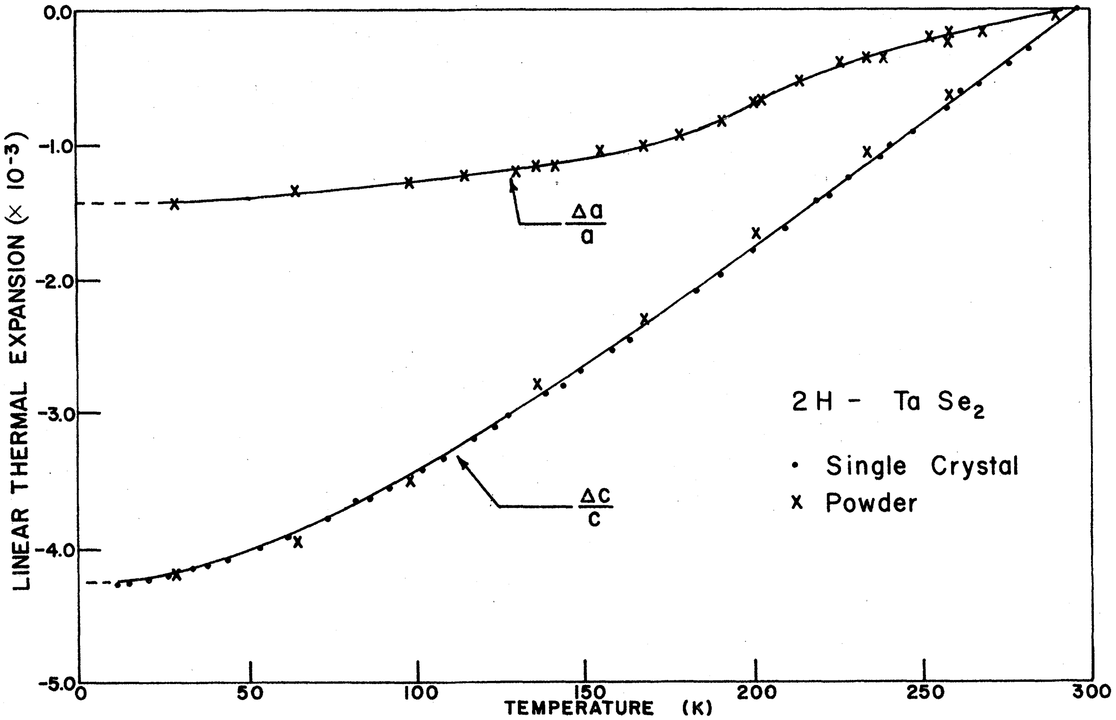

FIG. 2. Linear thermal expansions of 2*H*-$TaSe_2$.

TABLE II. Expansivities and Grüneisen parameters of 2*H*–$TaSe_2$.

T (K)	α_a (10^{-6} K^{-1})	α_c (10^{-6} K^{-1})	γ_1		γ_3	
120[a]	~3	14.7 ± 1.0	0.8[b]	0.8[c]	0.57[b]	0.59[c]
160	~5	18.3 ± 0.5	1.1[b]	1.1[c]	0.65[b]	0.70[c]
300	~6	18.3 ± 0.5	⋯	⋯	⋯	⋯

[a] At this temperature our results may be considered to be extrapolated values from beyond the ICDW phase-transitional region.

[b] Based on elastic moduli $c_{11}+c_{12}=33.6$, $c_{33}=5.5$, and $c_{13}=0.5$ in units of 10^{11} dyn/cm^2.

[c] Based on elastic moduli $c_{11}+c_{12}=29.9$, $c_{33}=5.4$, and $c_{13}=1.3$ in units of 10^{11} dyn/cm^2.

Our results, for both the polycrystalline and crystalline samples, for the thermal expansions relative to the $T = 300$ K lattice parameters are shown in Fig. 2. The two sets of data for $\Delta c/c$ agree to within combined experimental uncertainties. The more precise single-crystal results show that $\Delta c/c$ is linear in temperature for $160 < T < 300$ K, yielding an expansivity value of $\alpha_c = (18.3 \pm 0.5) \times 10^{-6}\ \mathrm{K}^{-1}$. The apparent maximum slope at $T = 250$ K of the $\Delta a/a$ vs T curve is "real" on the basis of our uncertainity estimates, but more precise measurements such as single-crystal measurements are desirable to obtain more accurate results.

Estimates of the linear thermal expansivities based on the data of Fig. 2 are shown in Table II at several temperatures. The room-temperature expansivities, in addition to other relevant thermal information, were used to estimate the expressions[17] for isothermal-adiabatic differences in the compliance moduli. Generally, these differences were found to be quite negligible, i.e., less than 0.3% of the corresponding moduli, where the latter were obtained from the succeeding portions of this paper.

IV. ESTIMATE OF c_{13}

Following the approach of Ref. 8 we write

$$c_{13} = \frac{1}{2k_a}(1 - c_{33}k_c) = \frac{1}{2 - k_c/k_a}\left(c_{11} + c_{12} - \frac{c_{33}k_c}{k_a}\right), \tag{1}$$

and we graphically check the consistency of the neutron-measured elastic moduli with the linear compressibilities obtained here. Plots of both equalities of Eq. (1) as a function of c_{33} are made for various possible values of k_a, k_c, and $c_{11} + c_{12}$. Intersections of the two expressions yield possible values of c_{13} and c_{33}. These sets of values can be tested by comparison with the c_{33} value obtained from neutron measurements.[9] Plots are made in Fig. 3 for only $k_c = 18$, $k_a = 4.0$, 3.3, and 2.6 (k_a and k_c in 10^{-13} cm^2/dyn), and for the neutron-measured values of c_{11} and c_{12}. The experimental uncertainty in $c_{11} + c_{12}$ is represented in the plot by the vertical error bars. When these latter uncertainties are taken into account it is seen that, corresponding to $k_c = 18$, all values of k_a (within the uncertainties of our x-ray measurements) are consistent with the remaining data, and that c_{13} is in the range -0.7 to 2.0 (c_{ij} in 10^{11} dyn/cm^2). In general, the results of this analysis are $-2 \leq c_{13} \leq 2$ and $k_c > 15.4$, although smaller values of k_c are consistent with our x-ray data. This result for c_{13} differs from the previous[2] result $|c_{13}| = 7.6 \pm \frac{1}{2}$ which was based in part on a vibrating-reed measurement[12] of s_{11}. However, the latter estimate of error apparently does not include the uncertainties in the neutron-measured elastic moduli; when these are included, it can be shown that the vibrating-reed measurement[12] is in accord with values of $|c_{13}|$ as small as ~1.

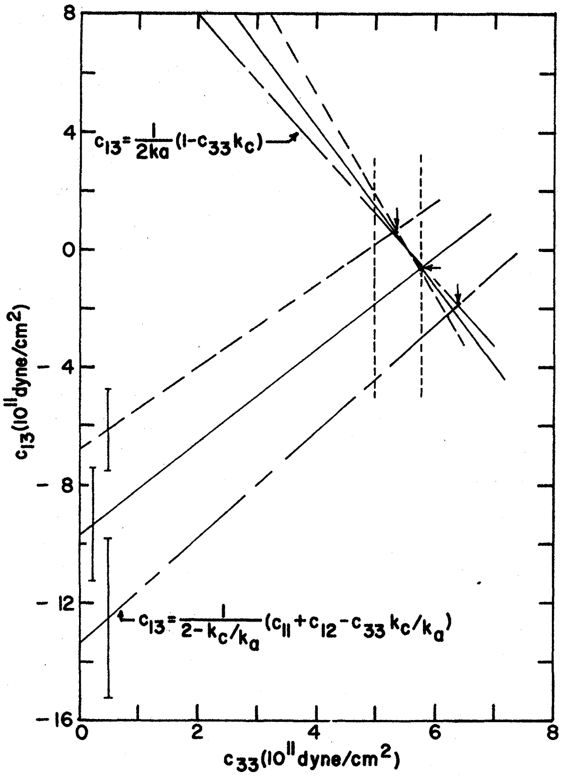

FIG. 3. Graphical solution of Eq. (1). Arrows point to simultaneous solutions of Eq. (1) with $c_{11} + c_{12} = 33.6 \times 10^{11}$ dyn/cm^2, $k_c = 18 \times 10^{-13}$ cm^2/dyn and $k_a = 2.6$ (large dashed lines), 3.3 (solid lines), 4.0 (broken lines) $\times 10^{-13}$ cm^2/dyn. The vertical bars correspond to the uncertainties in the expression for c_{13} arising from the experimental uncertainty in $c_{11} + c_{12}$. The small dashed vertical lines represent the range of c_{33} values based on neutron measurements (Ref. 9).

V. APPLICATIONS

A. Grüneisen parameters

With the availability of the thermal and elastic data presented in this paper, it is possible to obtain crude estimates of the anisotropic Grüneisen parameters of 2H-TaSe$_2$, given by the expressions[18]

$$\gamma_1 = (V/C_v)[(c_{11} + c_{12})\alpha_a + c_{13}\alpha_c], \tag{2a}$$

$$\gamma_3 = (V/C_v)(c_{33}\alpha_c + 2c_{13}\alpha_a). \tag{2b}$$

Assuming that the "electronic" contributions to the heat capacity *and* expansivities in the temperature region of interest are negligible, γ_1 and γ_3 are related to basal-plane and c-axis strain dependences of the lattice frequencies. Values of γ_1 and γ_3 for two somewhat arbitrarily chosen temperatures for which the expansivities differed are shown in Table II; the extrapolated normal-state heat capacity was used[7,13] in the $T = 120$ K estimates.

B. Debye temperature

Since elastic moduli generally decrease with increasing temperature, it is expected that Θ^{el} (300 K) $< \Theta_0^c$, where Θ^{el} and Θ_0^c are Debye temperatures appropriate to the elastic moduli and low-temperature heat capacity, respectively. In the case of $TaSe_2$, the only available information on the temperature dependence of the elastic moduli, i.e., the variation of Young's modulus[12] s_{11}^{-1}, is in accord with the idea that the lattice stiffens on cooling. We have evaluated Θ^{el} for various sets of elastic moduli that are consistent with our results for k_a and k_c as well as with the neutron-measured elastic moduli. The lowest value of Θ^{el} that we obtained was 230 K (corresponding to $c_{11} = 20.2$, $c_{12} = 9.7$, $c_{33} = 5.0$, $c_{13} = 1.0$, and $c_{44} = 1.72$). On the basis of the above discussion, our results appear to be incompatible with results for Θ_0^c of 202 K,[13] and 215 K.[19]

C. Properties of the ICDW transition

A suggestion was made recently[2] (on the basis of information on the stress dependences of T_0) that $\partial T_0/\partial \epsilon_3$ can *not* be neglected in comparison to $\partial T_0/\partial \epsilon_1$, where ϵ_3 and ϵ_1 are strains along axes perpendicular and parallel to the basal plane, respectively. It was of interest to test the validity of this suggestion in light of our improved results for c_{13}. In the course of this study, it was also found that the quoted value of dT_0/dp by Chu *et al.*[2] of 3.5 ± 0.2 K GPa^{-1} appeared to be too large on the basis of their data for T_0 vs p. Instead, the value 2.4 ± 0.3 K GPa^{-1}, which we obtained from a linear (in p) fit to their data below 0.5 GPa, is considered here, especially since this value of dT_0/dp leads to smaller values of $\partial T_0/\partial \epsilon_3$ than does the former one. The quantity $|\partial T_0/\partial \sigma_1|$ has been estimated indirectly[7] as 0.8 ± 0.3 K GPa^{-1}, where σ_1 is defined as a uniaxial stress in the basal plane ($p = -\sigma_1 = -\sigma_2 = -\sigma_3$ for hydrostatic pressures). Upon substitution into the expressions

$$\frac{\partial T_0}{\partial \epsilon_3} = \frac{\partial T_0}{\partial \sigma_1}(2c_{13} - 2c_{33}) - c_{33}\frac{dT_0}{dp}, \tag{3}$$

$$\frac{\partial T_0}{\partial \epsilon_1} = \frac{\partial T_0}{\partial \sigma_1}(2c_{11} + c_{12} - 2c_{33}) - c_{13}\frac{dT_0}{dp}, \tag{4}$$

it can be seen that, corresponding to $\partial T_0/\partial \sigma_1 = -0.8 \pm 0.3$ K GPa^{-1}, negligible values of $\partial T_0/\partial \epsilon_3$ are possible, and that $\partial T_0/\partial \epsilon_1$ is of the order of -240 K.

Barmatz *et al.*[7] have estimated the absolute magnitude of the gap $\Delta_0 \alpha_a$ in the basal-plane linear thermal expansivity at the ICDW transition on the basis of the Ehrenfest relations. However, the gap $\Delta_0 \alpha_v$ in the volumetric expansivity was not considered. From our value of dT_0/dp and from $\Delta_0 C_p$ of Ref. 7, the Ehrenfest relations[20] yield $\Delta_0 \alpha_v = 8 \times 10^{-6}$ K^{-1}, which is comparable in magnitude to $2|\Delta_0 \alpha_a| = (5 \pm 2.2) \times 10^{-6}$ K^{-1}, obtained previously,[7] and the positive sign implies that α_v increases with decreasing temperature across the transition.

Finally, it is of some interest to predict a value for $\Delta_0 c_{11}$, the jump in c_{11} at the ICDW transition from the measured value of $\Delta_0 E/E$,[12,7] where $E = s_{11}^{-1}$. A simplified expression for s_{11} in terms of the elastic moduli results if $c_{13} \sim 0$; in this approximation,

$$\frac{\Delta_0 E}{E} = [1 - 2c_{11}/(c_{11} + c_{12})]\Delta_0 c_{11}/c_{11}, \tag{5}$$

where it is also assumed that to a first approximation the gap in the shear modulus $c_{11} - c_{12}$ can be neglected.[21] Upon substitution of the appropriate quantities into Eq. (5), we obtain $\Delta_0 c_{11}/c_{11} \sim 7 \times 10^{-4}$. We estimate a possible 12% uncertainty in this value associated with the possible size of c_{13}—under the assumption of negligible jumps in c_{33} and c_{13} at T_0.

VI. SUMMARY AND CONCLUSIONS

We have presented results of linear compressibility and thermal-expansion x-ray measurements for $2H$-$TaSe_2$, and have combined the former results with neutron-measured elastic moduli to obtain a more accurate determination of c_{13} than that obtained previously, although we can specify c_{13} to only $-2 < c_{13} < 2$ (in 10^{11} dyn/cm^2). Our thermal-expansion measurements have shown that anharmonicity is probably not sufficiently important to affect the validity of our combination of zero-sound and isothermal elastic moduli, although perhaps nonadiabatic electronic effects are important in this regard. We mention here that additional support of our results has been gained by considering predictions of short-range bond stretching and valence-force-field lattice-dynamical models.[22] These considerations show that bond-stretching models with parameters appropriate for $2H$-$TaSe_2$ yield $c_{13} \sim (1-2) \times 10^{11}$ dyn/cm^2, and that valence-force-field models may yield negative values of c_{13} if ratios of intralayer bond bending to bond-stretching force constants are sufficiently large, e.g., somewhat larger than for $2H$-MoS_2.[10]

Our results have been applied to estimates of various quantities of interest such as anisotropic Grüneisen parameters, the Debye temperature, strain dependencies of T_0, and the discontinuity in c_{11} at T_0. Perhaps most significantly, we found that (a) the dependence of T_0 on c-axis strains may be negligible contrary to a previous suggestion[2] and (b) the room-temperature elastic moduli yield too low a value for the T^3 contribution to the low-temperature specific heat, i.e., $\Theta^{el} > \Theta_0^c$. Similar results to (b) were obtained for[8] $2H$-$NbSe_2$ and for[23] NbO_2; for NbO_2, accurate low-temperature measurements of elastic moduli were considered, so the discrepancy in that case is clearly meaningful. If it is presumed that the c_{ij} are only weakly dependent on temperature or that they decrease between $T=0$ and $T=300$ K, then result (b) implies the presence of low-lying energy excitations—perhaps associated with distortions of charge-density waves[23,24] in addition to the long-wavelength acoustical lattice vibrations.

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