Phonon Spectrum of Thorium

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Inelastic-neutron-scattering measurements of the phonon dispersion relation in thorium at room temperature are reported. The measurements were made along the $(0, 0, \zeta)$, $(0, \zeta, \zeta)$, (ζ, ζ, ζ) , and $(0, \zeta, 1)$ symmetry directions. The dispersion curves are dominated by nearest-neighbor interactions but indicate a residual long-range interaction to at least seventh nearest neighbors. A seventh-nearest-neighbor force-constant model was fitted to the data and used to obtain a frequency distribution function and the temperature dependence of the Debye temperature. The transverse modes along the $(0, \zeta, \zeta)$ direction appear to show structure indicative of strong electron-phonon interaction effects.

I. INTRODUCTION

Metallic thorium has a conduction band formed from the 6d and 7s atomic states, while the 5fshells are empty but are believed to lie not too far above the Fermi level.^{1,2} The band-structure calculations of Gupta and Loucks¹ show that the 6d and 7s levels hybridize to form a conduction band similar to that of a transition metal. Thorium is a superconductor and thus might be expected to have a large electron-phonon interaction. The present experiment was motivated by an interest in determining the possible effects of any low-lying unfilled f bands on the phonon spectrum and possible effects due to the electron-phonon interaction. These measurements are the first inelastic-neutron-scattering measurements to be reported on an element in the actinide series, due to the unavailability hitherto of large single crystals.

II. MEASUREMENTS AND RESULTS

The crystal used for these measurements was approximately 1 cm in diameter and 2.0 cm long. It was grown using a variation of the strain-annealing technique.³ A rod of polycrystalline thorium was sealed into a tantalum capsule under purified argon and heated to 1450 °C. The temperature was lowered to 1300 °C and raised again to 1450 °C rather quickly. This temperature cycle was repeated three times and the specimen was then annealed at 1320 °C for 48 h and slowly cooled. A typical analysis of the thorium used in the preparation showed the following impurities: oxygen 80 ppm, carbon 55 ppm, and nitrogen 30 ppm. The neutron measurements were done using the tripleaxis spectrometer at the Ames Laboratory Research Reactor in the constant-Q mode of operation with fixed incident energy. Incident wavelengths of 2.09 and 1.49 Å were used for the measurements, the former for measurement of the lower-

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frequency phonons and for branches where greater instrumental resolution was necessary. All measurements were made at room temperature. Thorium has a coherent scattering cross section of 12.1 b, but the measurements are made more difficult by the extremely large atomic mass and by a thermal absorption cross section of 7.5 b which together act to reduce the intensities of the onephonon peaks.

The measurements were made along the three principal symmetry directions of the reciprocal lattice, namely, the $(0, 0, \zeta)$, $(0, \zeta, \zeta)$, and (ζ, ζ, ζ) directions. In addition, measurements were made along the $(0, \zeta, 1)$ direction, which although not a symmetry axis nevertheless has the property that the dynamical matrix factorizes for \vec{q} along this direction so that a Fourier analysis of ω^2 yields linear combinations of the interatomic force constants. The frequencies were estimated in the usual manner, namely, by fitting the observed neutron groups and the background to a series of Gaussian peaks and taking the phonon frequency to be the centroid of the corresponding neutron group minus the background. Errors in the phonon frequencies were estimated to be 0.015 THz, except for those cases where the statistical errors were greater than this. For these phonons the estimated error is given as the statistical error. Table I lists the measured phonon frequencies and errors. The experimental dispersion curves are shown in Fig. 1, together with a seventh-nearest-neighbor forceconstant model fit which is discussed in Sec. III. It should be noted that experimental results are displayed for the Π' branch only along $(0, \zeta, 1)$, since the Π branch is simply its mirror image about W. The fitted curves are constrained to fit the room-temperature elastic constants for thorium as measured by Armstrong *et al.*⁴ It may be seen that the initial slopes of the dispersion curves obtained from these elastic constants are compatible

TABLE I. Normal-mode frequencies (in THz) for the symmetry branches in thorium at room temperature.

<u></u>	1)		1)
<u>ه</u>	<u>ν</u> (0 ζ ζ) <i>Τ</i> .	5	$\frac{\nu}{(0 \ \xi \ \xi) T_{-}}$
0 1	(∨, ∍, ≥/▲1 0 377±0 015	0 1	(v, v, b, 5/42) 0 545 ± 0 015
0.1	0.377 ± 0.015 0.657 ± 0.015	0.1	0.545 ± 0.015 1 071 + 0 015
0.3	0.031 ± 0.015 0.998 ± 0.015	0.3	1.553 ± 0.015
0.35	1.168 ± 0.015	0.35	$1,744 \pm 0.015$
0.4	1.302 ± 0.015	0.40	1.934 ± 0.015
0.45	1.407 ± 0.015	0.45	2.080 ± 0.015
0.50	1.500 ± 0.015	0.50	2.241 ± 0.015
0.55	1.558 ± 0.015	0.55	2.463 ± 0.015
0.6	1.631 ± 0.015	0.60	2.693 ± 0.015
0.65	$\textbf{1.720} \pm \textbf{0.015}$	0.7	3.083 ± 0.015
0.7	1.808 ± 0.015	0.8	$\textbf{3.32}\pm\textbf{0.03}$
0.75	1.883 ± 0.015	0.9	3.45 ± 0.02
0.80	2.002 ± 0.015	1.0	3.474 ± 0.015
0.85	2.113 ± 0.015		
0.9	2.202 ± 0.015		
0.95	2.246 ± 0.015		
1.0	2.259 ± 0.015		
	(0, ζ, ζ) <i>L</i>		
0.1	0.825 ± 0.01		
0.2	1.549 ± 0.015		
0.3	2.06 ± 0.02		
0.4	2.630 ± 0.015		
0.5	3.056 ± 0.015		
0.0	3.110 ± 0.013 2.80 ± 0.04		
0.7	2.80 ± 0.04 2.666 ± 0.015		
0.9	2.355 ± 0.015		
1.0	2.259 ± 0.015		
	$(0, 0, \zeta)L$		(0, 0, ζ) T
0 2	1 013+0 015	0.2	0.75 ± 0.015
0.2	1.015 ± 0.015 1.48 ± 0.01	0.3	$1,071 \pm 0,015$
0.4	1.94 ± 0.02	0.4	1.364 ± 0.015
0.5	2.417 ± 0.015	0.5	1.585 ± 0.015
0.6	2.81 ± 0.01	0.6	1.836 ± 0.015
0.7	3.08 ± 0.02	0.7	1.977 ± 0.015
0.8	3.347 ± 0.015	0.8	2.135 ± 0.015
0.9	3.45 ± 0.03	0.9	2.243 ± 0.015
1.0	$\textbf{3.474} \pm \textbf{0.015}$	1.0	$\textbf{2.259} \pm \textbf{0.015}$
	(٤, ٤, ٤)L		(ζ, ζ, ζ) <i>T</i>
0 1	1.055 ± 0.015	0 1	0.465 ± 0.03
0.15	1.502 ± 0.015	0.2	0.85 ± 0.015
0.2	1.864 ± 0.015	0.25	0.98 ± 0.02
0.25	2.25 ± 0.04	0.3	1.11 ± 0.015
0.3	2.542 ± 0.015	0.35	1.215 ± 0.015
0.35	2.841 ± 0.015	0.4	1.23 ± 0.03
0.4	3.054 ± 0.015	0.45	$\textbf{1.25} \pm \textbf{0.02}$
0.425	3.163 ± 0.015	0.5	1.28 ± 0.02
0.45	3.160 ± 0.015		
0.475	$\textbf{3.3}\pm\textbf{0.1}$		
0.5	3.24 ± 0.08		
	(0, ζ, 1)Λ		(0, ζ, 1)Π'
0.0	2.259 ± 0.015	0.0	2.259 ± 0.015
0.2	2.20 ± 0.015	0.2	2.358 ± 0.015
0.4	2.21 ± 0.05	0.4	2.60 ± 0.015
0.6	2.22 ± 0.05	0.6	3.15 ± 0.015
U.8 1 0	2.213 ± 0.015	U.8 1 0	3.42 ± 0.015
T. 0	4.209 ± 0.015	T. 0	3.470 ± 0.015

with our measured points within experimental error except for the $T_1(0, \zeta, \zeta)$ branch, where the measured frequency for the phonon at (0.1, 0.1, 0) $2\pi/a$ is about 20% greater than that expected from the elastic constants. It may also be seen from Fig. 1 that the transverse modes along $(0, \zeta, \zeta)$ exhibit pronounced structure in the sense that the curvature of these branches becomes positive for a certain region of wave-vector space. This behavior is discussed in Sec. IV in connection with effects arising from the electron-phonon interaction in thorium.

III. ANALYSIS

In order to determine the range of the interatomic forces and to obtain a force-constant model which would serve as a convenient interpolation formula for the phonon frequencies throughout the zone, a Fourier analysis was made of the squares of the frequencies of the different branches along the symmetry directions. We shall refer to the Fourier coefficients thus obtained as the interplanar force constants for these branches,⁵ although strictly speaking the coefficients for the $(0, \zeta, 1)$ direction are not interplanar force constants but simply linear combinations of interatomic force constants. Figure 2 illustrates the variance ratio or goodness of fit versus number of Fourier coefficients for some of the branches analyzed in this way. Table II lists the interplanar force constants obtained for all the branches by including as many coefficients as were needed to obtain convergence. Also listed are the interplanar force constants obtained from the fitted seventh-nearest-neighbor force-constant model. It may be noted both from Fig. 2 and Table II that the transverse branches did not require more Fourier coefficients for convergence than were contained in the seventh-nearest-neighbor model, but the longitudinal branches and the Λ branch along $(0, \zeta, 1)$ required at least one more coefficient for convergence indicating that these branches involve interactions beyond seventh neighbors. Nevertheless, to restrict the number of parameters we did not consider models involving more than seven neighbors. Following the method of Squires,⁶ we may relate the interplanar force constants to the interatomic force constants through a series of linear relations. The measured elastic constants may also be expressed in terms of linear combinations of the interatomic force constants. In addition, since a general tensorforce-constant model extending out to seven neighbors cannot be determined from the symmetrydirection data and elastic constants alone,⁷ the fifth- and seventh-neighbor forces were restricted to be axially symmetric⁸ thus imposing four extra linear constraints on the parameters. Thus the total of 24 force-constant parameters were deter-



FIG. 1. Phonon dispersion curves for thorium at room temperature along the principal symmetry directions. The experimental errors lie within the circles representing the measured points unless indicated by an error bar. The smooth curves represent the seventh-nearest-neighbor force-constant model referred to in the text.

mined from 40 linear relations involving the interplanar force constants, elastic constants, and 6 axially symmetric conditions for the fifth and seventh neighbors. The interatomic force constants obtained by a linear least-squares analysis are listed in Table III. Figure 1 shows that the fit to the measured dispersion curves is quite good except for the Λ branch along the $(0, \zeta, 1)$ direction. The Fourier analysis for this branch (Fig. 2) indicates that an extra coefficient involving at least eighth-neighbor force constants is necessary to obtain a good fit. However, we do not believe that the over-all dispersion curves are sensitive to these higher neighbor interactions. It may be noted that the force constants imply dominant nearest-neighbor interactions, these being more than an order of magnitude larger than any further neighbor interactions. The nearest-neighbor force constants were tested for axial symmetry, and were found to obey the axially symmetric condition to within only 20%.

The above model was used to generate a frequency distribution function $g(\nu)$ using the method of Gilat and Raubenheimer.⁹ This is shown in Fig. 3. Also indicated are the positions of the critical points obtained from the symmetry directions alone. The frequency distribution function was used to obtain a Debye temperature for thorium as a function of temperature. This is shown in Fig. 4. It may be seen that above 90 K, Θ_D stays remarkably constant at a value of about 143 K. There have been a number of measurements of the specific heat of thorium. The analysis of Clusius and Franzosini¹⁰ yields a Θ_D which levels off at about 155 K at the higher temperatures. However,

this analysis was based on a value of 19×10^{-4} cal mole⁻¹ K⁻² for the linear coefficient γ of the electronic specific heat. The low-temperature data of Gordon et al.¹¹ and of Smith and Wolcott, ¹² on the other hand, yield a value for γ of about 11.2 $\times 10^{-4}$ cal mole⁻¹ K⁻² which would reduce the value for Θ_D obtained from the experimental specific heat considerably at the higher temperatures. The high-temperature data of Wallace¹³ yields an increasing contribution to the specific heat which he ascribes to thermal expansion effects and electronic level population effects, although this excess heat is based on a lattice specific heat obtained from Θ_p of 155 K. We believe that the actual lattice specific heat may be obtained more accurately from our calculation since it is fitted to the phonon



FIG. 2. Variance ratio for the Fourier fit to ω^2 vs \vec{q} for certain branches plotted against the number of coefficients used.

 0.061 ± 0.023 the Fourier analysis had converged; the lower line refers to the interplanar force constants calculated from the seventh-nearest-neighbor model : : : : പ്പ : : : : : ď, : 0.066±0.028 0.105 - 0, 004 ± 0, 006 0, 001 0.111 ± 0.056 : പ് : : : $\begin{array}{c} 0.\ 012 \pm 0.\ 010 \\ 0.\ 008 \end{array}$ 0.072 ± 0.075 -0.112 ± 0.018 പ്പ : 0.216 : : -0.084 0.0354 ± 0.023 -0.111 ± 0.030 -0.0610.070±0.006 0.075 -0.099 ± 0.013 -0.105 -0.161 ± 0.086 0.245 ± 0.032 357 ± 0.043 : 4 0.191 -0.186 0. 0583 ± 0. 015 0. 065 -0.0969 ± 0.034 -0.369±0.078 -0.339 -0.007±0.039 -0.083 0.204±0.013 0.166 0.629 ±0.123 0.525 : : ជុំ : -0.058 -0.0942 ± 0.036 -0.0187 ± 0.021 2.487±0.052 2.400 0.387±0.123 0.189 0.299±0.096 0.957 -0.300 ± 0.047 -0.137 ± 0.014 3.887 ± 0.025 0.618 ± 0.008 ፈ -0.365 -0.141 3.915 0.621 -0.080 -0.036 1.758±0.014 1.721 **4.** 699 ± 0. 045 1.919 ± 0.021 1.611 ± 0.056 -5.700 ± 0.126 2.169 ±0.06 2.227 : : đ 4.672 1.977 ÷ 1.646 -5.615 13.005 ± 0.094 13.072 7.594±0.06 7.214 : : ሳ 1: (0, ⁵, 1)II' (0, ξ, ζ)T₂ ξ, ξ)L $(0, \xi, \xi)T_1$ (0, ^t, 1)A (0, 0, *t*)L (0, 0, *ξ*)T (č, č, č)L (č, č, č)T é

dispersion curves and together with the experimental low-temperature value for the electronic specific heat coefficient may be used to accurately identify the excess contributions considered by Wallace. Gordon *et al.* also obtain a Θ_D at 0 K of 163.3 K which is, not surprisingly, higher than ours since the latter are based on room-temperature phonon frequencies.

IV. DISCUSSION

We have compared the dispersion curves for thorium with those of other fcc metals in order to examine the role the electronic structure plays in determining the vibrational spectra of metals. Lead is the closest metal to thorium in the Periodic Table with the fcc structure, and like thorium it has four electrons outside the core. The ratio of the core radius to nearest-neighbor distance is roughly the same for the two metals. The dispersion curves for lead, however, show much more pronounced structure and sharper anomalies¹⁴ than those of thorium. The real-space counterpart of this behavior is that the force constants in lead drop off very slowly with increasing neighbor distance. Thus, for example, the second-neighbor force constants in lead are about a third as large as the first-neighbor force constants. In thorium, on the other hand, the first-neighbor force constants are over ten times as large as any other force constants, although from the second neighbor onwards they too drop off very slowly. It is this dominant first-neighbor interaction which makes the dispersion curves of thorium look simpler than those of many other fcc metals. The only other fcc metals with such dominant first-neighbor interactions are the noble metals and nickel. In these metals the large first-neighbor interactions are due to the spatial extent of the *d*-electron wave functions around each atom. Although as mentioned above the energy bands of thorium near the Fermi level do have d-like character, the number of occupied d-like states is relatively small compared to those in the noble metals or nickel.¹ However since there are a large number of d-like states just above the Fermi level in thorium, the virtual excitations induced by the electron-phonon interaction cause the electrons to spend part of their time in spatially extended d-electron-like states thus contributing to the large nearest-neighbor interaction. It should be noted that lead does not have any d-like states close to the Fermi level, and thus does not show this effect. In order to see mathematically how this "virtual hard core" comes about, we may examine a typical term in the microscopic expression for the electronic contribution to the dynamical matrix, ¹⁵

$$E_{\alpha\beta}^{(1)}(q) = \frac{1}{2\Omega} \sum_{k,k'}^{\prime} \frac{n(k) - n(k')}{E_k - E_{k'}} |V_{k'k}|^2 , \qquad (1)$$

TABLE II. Interplanar force constants for thorium (units of 10⁴ dyn/cm). The top line for each mode refers to the interplanar force constants obtained after

m temperature (units in 10° dyn/cm).				
α ¹¹⁰	1.181±0.025			
a ¹¹⁰	-0.218 ± 0.049			
β_3^{110}	$\textbf{1.176} \pm \textbf{0.037}$			
α_1^{200}	-0.090 ± 0.044			
α_3^{200}	-0.093 ± 0.025			
α_1^{211}	-0.035 ± 0.028			
α_2^{211}	0.015 ± 0.015			
eta_1^{211}	0.060 ± 0.017			
eta_2^{211}	-0.072 ± 0.011			
$lpha_1^{220}$	-0.013 ± 0.013			
$lpha_3^{220}$	$\textbf{0.096} \pm \textbf{0.022}$			
β_{3}^{220}	0.054 ± 0.039			
α_1^{310}	-0.041 ± 0.025			
$lpha_2^{310}$	-0.008 ± 0.010			
α_{3}^{310}	-0.003 ± 0.014			
β_{3}^{310}	-0.013 ± 0.012			
$lpha_1^{222}$	-0.034 ± 0.011			
β_{1}^{222}	-0.061 ± 0.018			
α_1^{321}	0.013 ± 0.012			
$lpha_2^{321}$	$\textbf{0.043} \pm \textbf{0.012}$			
$lpha_3^{321}$	-0.021 ± 0.006			
β_1^{321}	0.009 ± 0.002			
eta_2^{321}	0.013 ± 0.002			
β_{3}^{321}	0.026 ± 0.005			

TABLE III. Interatomic force constants for thorium at room temperature (units in 10^4 dyn/cm).^a

^aThe notation is the same as in Ref. 6.

where n(k) and E_k are, respectively, the occupation number and energies of the Bloch states k, and $V_{k'k}$ is an effective electron-phonon matrix element between states k, k'. It may be seen from this expression that if one of the states k, k' is d-like, the contribution $E_{\alpha\beta}(\mathbf{q})$ is symmetric with regard to whether it is occupied and the excited state is freeelectron-like or whether the d-like state is the excited state and the occupied state is free-electronlike. Thus a band of unoccupied d-like states just above the Fermi level will provide the same kind of large near-neighbor interactions as a band of similar states just below the Fermi level. In thorium, moreover, recent energy-band calculations have estimated the 5f bands to be roughly 0.4 Ry above the Fermi level. Although these are much further away from the Fermi level than the d bands, the 5f wave functions will be even more spread out and so could contribute somewhat to enhancing the nearest-neighbor interaction.



FIG. 3. Phonon density-of-states function $g(\nu)$ calculated from the seventh-nearest-neighbor force-constant model. Critical points obtained from the symmetry points in the zone are indicated.

We now turn to a discussion of the anomalous behavior of the T_1 and T_2 branches along the $(0, \zeta, \zeta)$ direction. If we regard the positions of the anomalies as being the wave vectors at which the dip from a smooth curve is a maximum then these occur at $(2\pi/a)(0, 0.7, 0.7)$ for the T_1 branch and $(2\pi/a)(0, 0.5, 0.5)$ for the T_2 branch. Our measurements also do not preclude an anomaly at $(2\pi/a)(0, 0.7, 0.7)$ for the L branch along $(0, \zeta, \zeta)$, since the phonons in this region were hard to measure accurately and also the dispersion curve is changing sharply in this region. The anomalies are too broad to be ascribed to the Kohn effect, ¹⁶ namely, the logarithmic singularities in the derivative of the dispersion curve as the wave vector passes through an external diameter of the Fermi surface. Such anomalies generally manifest themselves in materials with strong electron-phonon coupling, as in lead¹⁴ and in these too the effect is rather small. On the other hand, it has been recognized for some time now¹⁷ that large peaks in the generalized susceptibility function $\chi(\mathbf{Q})$ of the met-



FIG. 4. Calculated Debye temperature for thorium vs temperature.

though qualit

al (rather than abrupt changes in the derivative) also produce local depressions in the dispersion curves in the vicinity of the wave vectors $(\vec{Q}_m - \vec{H})$ where \vec{Q}_m is the value of \vec{Q} at which $\chi(\vec{Q})$ peaks and \vec{H} is the reciprocal-lattice vector required to translate \vec{Q} back into the first zone. The diagonal elements of the dielectric function $\epsilon(\vec{Q}, \vec{Q})$ are determined by $\chi(\vec{Q})$, and thus pronounced structure in $\chi(\vec{Q})$ will lead, in general, to structure in the phonon dispersion curves. The general expression for $\chi(\vec{Q})$ is given by

$$\chi(\vec{\mathbf{Q}}) = \sum_{k,k'} \frac{n(k) - n(k')}{E_k - E'_k} \langle \psi_k | e^{-i\vec{\mathbf{Q}}\cdot\vec{\mathbf{r}}} | \psi_{k'} \rangle \langle \psi_{k'} | e^{i\vec{\mathbf{Q}}\cdot\vec{\mathbf{r}}} | \psi_k \rangle .$$
(2)

It should be noted that even the transverse modes will be affected for $\vec{q} = \vec{Q}_m$ (i.e., $\vec{H} = 0$) if \vec{Q}_m occurs in the first zone if the metal is non-free-electronlike, since if the states are not plane-wave states, the electron-phonon matrix element can still couple states \vec{k} and $\vec{k} + \vec{q}$ via a transverse phonon.¹⁵

Our results indicate that the dominant peaks in $\chi(\vec{Q})$ for thorium probably occur along the $(0, \xi, \xi)$ direction. From Eq. (2) it may be seen that these peaks would be likely to occur for a \vec{Q} which connects two nested regions of the Fermi surface.¹⁸ The Fermi-surface calculations of Gupta and Loucks¹ indicate that along the $(0, \xi, \xi)$ direction, the opposite faces of the rounded cube-type hole surface are nested, and so are diametrically opposite flat portions of the so-called lung electron surfaces. However the \vec{Q} vectors corresponding to these separations in their calculations are $(2\pi/a)(0, 0.43, 0.43)$ and $(2\pi/a)(0, 0.82, 0.82)$, respectively, which do not agree with the positions of the observed anomalies. It should be borne in

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mind that the calculations although qualitatively correct are in quantitative disagreement with some of the de Haas-van Alphen periods as measured by Boyle and Gold, ¹⁹ and the peaks in $\chi(\vec{\mathbf{Q}})$ may well be very sensitive to the details of the band structure. Without detailed calculation of the electronphonon matrix elements, it is not possible to predict which peak in $\chi(\vec{\mathbf{Q}})$ will give rise to structure in a particular branch of the dispersion curve. It is also somewhat disturbing that the nested region along the $(0, 0, \zeta)$ axis also present in their Fermisurface calculations does not seem to give rise to any structure in the $(0, 0, \zeta)$ dispersion curves.

V. SUMMARY

Our measurements indicate that the dispersion curves in thorium at room temperature are dominated by a large non-axially-symmetric nearestneighbor interaction, which is probably due to virtual excitations involving the d and possibly the flevels close to the Fermi level. There is also a residual long-range interaction extending to at least seven nearest-neighbor shells. The anomalous behavior of the T_1 and T_2 branches along $(0, \zeta, \zeta)$ has been ascribed to structure in the generalized susceptibility function $\chi(\mathbf{Q})$ along this direction, although the positions of the anomalies cannot be quantitatively accounted for by the calculated band structure.

It would be interesting to confirm the peaks we have determined in $g(\nu)$ by means of superconducting tunneling measurements on thorium. To date, no experimental results on the phonon density of states by this method are available for comparison with our results.

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