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Calorimetric determination of the electron-phonon mass enhancement of α-uranium*

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A conduction-electron mass enhancement λ for the 2-K superconducting α phase of uranium was obtained from a comparison of the linear coefficient of the electronic heat capacity at high temperature, γ_b, with that at low temperature, γ_0. A λ of 0.9 ± 0.2 was found. The γ_0 value used was obtained in a high-pressure experiment reported in the literature. The γ_b value was extracted from published heat-capacity measurements below 500 K by adjustment of the data to the volume of the low-temperature experiment, subtraction of an appropriate Debye function, correcting for anharmonicity, and graphically fitting the resultant electronic contribution. The uncertainty in λ is dominated by the uncertainty in the characterization of the constant-volume lattice anharmonicity, which was estimated using Wallace's recent empirical correlation based on Grüneisen constants. Using the observed magnitude of λ and T_c, McMillan's equation for the superconducting transition temperature indicates that large Cooper-pair-weakening interactions are probably present in α-uranium. It is known that the γ_0 value of the 2-K superconducting α phase of uranium is $\sim 30\%$ higher than the γ_0 value of the lower-T_c single-crystal uranium at zero pressure. A combined analysis of the electronic heat capacity and magnetic susceptibility below room temperature indicates that the different values of γ_0 reflect not only changes in the unenhanced electronic density of states at the Fermi energy, but also substantial changes in λ. Furthermore, the low-pressure depression of T_c relative to the α-phase T_c value of 2 K is in large part due to a depression in λ.

I. INTRODUCTION

The superconductivity of uranium metal has been a subject of controversy since first detected[1] in 1942. Many ambiguities associated with the experimental situation concerning uranium metal have been clarified in recent years.[2] Broad bulk superconducting transitions, which are sensitive to grain size, occur well below 1 K in polycrystals at zero pressure.[3] No bulk transitions have been conclusively detected in single crystals[3] above ~0.1 K. A maximum of ~2 K occurs in the superconducting transition temperature T_c near 12 kbar.[4,5] Furthermore, a clear relationship exists between the initial enhancement of T_c and the suppression of a low-temperature volume minimum under pressure.[5-7] The volume minimum occurs[8] at 43 K at zero pressure and is accompanied, most dramatically, by sharp minima in the elastic moduli.[9,10] The anomalies in the elastic moduli are linearly depressed in temperature by application of pressures[6] less than 4 kbar. Hence uranium metal is a strongly pressure-enhanced superconductor until the volume minimum is suppressed. In addition, at 11 kbar uranium metal has a large positive superconducting isotope effect.[11] The purpose of the investigation reported in the present paper was to obtain the electron-phonon mass enhancement for α-uranium to help clarify the dominant interactions that influence the low-temperature physical properties of uranium metal.

Four distinct categories of proposals have been advanced to explain the nature of the superconductivity of uranium metal and, in particular, the role that f electrons play in the low-temperature physical properties.

(i) *New mechanisms* were proposed whereby f electrons facilitate the electron pairing necessary for the formation of the superconducting state. Such pairing would occur either by exchange of virtual f excitations above the Fermi energy,[12] or by exchange of virtual polarons if the f-like electrons are below the Fermi energy.[11] It was subsequently found that both models predict the isotope effect would be small, and, in the case of virtual-polaron coupling, the isotope effect would be positive.[13] The proximity of f-like states to the Fermi energy in α-uranium would be responsible for the higher T_c value compared with that of thorium metal.

(ii) *Cooper-pair-breaking interactions* (arising from magnetic scattering processes) were proposed whereby either local moments[7] or spin-density excitations[5] suppress the formation of the usual BCS superconducting state. The low-temperature volume minimum would be associated with the appearance of, or ordering of f electrons.

(iii) *Unusually large Cooper-pair-weakening interactions* were proposed whereby strong Coulomb repulsions associated with a narrow f-like band suppresses the superconducting state.[14] Such interactions could give rise to a positive isotope effect due to the mass dependence of the phonon cutoff frequency.

(iv) *Phonon instabilities* were proposed whereby the electron-phonon interaction is modified by phonon softening and stiffening.[15] The phonon frequency shifts would be caused by the narrow-band f-electronic screening effects. The low-temperature volume minimum and, hence, the resultant negative Grüneisen parameter would be a reflection of low-energy acoustic-phonon softening.[16]

Before invoking new mechanisms for superconductivity, it should be clear that the existing theoretical framework cannot describe the physical properties of the system. Lanthanum, similar to uranium, is a highly pressure-sensitive superconductor[17] that exhibits a volume minimum at low temperatures.[18] Superconducting tunneling experiments on the double-hexagonal-close-packed phase of lanthanum provided information on the phonon spectrum which clearly indicates that the electron-phonon interaction strength increases with pressure.[15] Hence, specifically in the case of uranium, it is not required to invoke new superconductive mechanisms to describe these related low-temperature physical properties. Attempts also have been made to explain the positive isotope effect within the usual electron-phonon framework.[14,19] These attempts will be considered in Sec. III.

Proposals (ii)–(iv) invoke conventional pairing interactions. The second, Cooper-pair-breaking effects, would be evident in calorimetric studies as a rapid reduction in the magnitude of the heat capacity anomaly at T_c relative to that at the 2-K maximum in T_c. In the absence of Cooper-pair-breaking effects, the heat-capacity anomaly at T_c might be expected to follow the BCS law of corresponding states. Recent zero-pressure heat-capacity measurements at the superconducting transition for one polycrystalline sample were clearly inconsistent with pair-breaking effects.[3]

A determination of the magnitude of the electron-phonon mass enhancement, λ, *can provide the necessary information to determine whether proposal* (iii) or (iv) *dominates the low-temperature anomalies in uranium metal*. If, for instance, unusually large Cooper-pair-weakening interactions are involved, then for a given T_c, λ also would be unusually large when compared with that calculated from the usual BCS-type expressions.

The most useful expression for calculating T_c within the BCS framework is McMillan's semiempirical strong-coupled expression[20]

$$T_c = \frac{\Theta}{1.45} \exp\left(- \frac{1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)} \right) , \qquad (1)$$

where Θ is a characteristic phonon temperature and can be taken as the Debye temperature, μ^* is a strength of the Cooper-pair-weakening interaction, and λ, the electron-phonon mass enhancement, is the attractive pairing interaction. The value of λ can, in principle, be obtained from heat-capacity data. The linear coefficient γ_0 of the electronic heat capacity C_E obtained experimentally at low temperatures can be expressed as

$$\gamma_0 = \lim_{T \to 0} (C_E/T) = (1+\lambda)\gamma_b \ , \qquad (2)$$

where γ_b is proportional to $N_b(0)$, the noninteracting, one-electron density of states at the Fermi energy for both spin directions, and

$$\gamma_b = \tfrac{2}{3}\pi^2 k_B^2 N_b(0) \ . \qquad (3)$$

$N_b(0)$ is the density of states obtained from band-structure calculations. According to theoretical calculations, above a temperature Θ the electron-phonon many-body alteration of the electronic heat capacity tends to zero monotonically.[21] At high temperatures, the many-body alterations of the one-electron density of states become unimportant. If the measured electronic heat capacity can be characterized above Θ and extrapolated to $T=0$ K, γ_b can be obtained. Recently, Knapp *et al.*[22] obtained, from analyses of heat-capacity data, λ values for Sc, Y, V, Pt, Pd, Nb, and Ta. Their results are generally consistent with the known T_c values, band-structure calculations, or other experimental determinations of γ_b for these materials.

II. DATA ANALYSIS AND RESULTS

The analysis to obtain the λ value pertains to the α phase. The α phase is assumed to exist between 43 and 934 K at zero pressure, and at all temperatures below 934 K for pressures greater than ~10 kbar. [The α phase could also sometimes be retained at zero pressure by quenching from above 43 K (Ref. 23).] Ho *et al.*[4] measured γ_0 at ~10 kbar and obtained a value of 12.2 mJ/°K^2 mole. The smoothed data by Flotow and Lohr[24] between 5 and 350 K and by Ginnings and Corruccini[25] between 273 and 1173 were used for the high-temperature heat-capacity analysis. (The analysis was performed below 500 K to avoid the effects associated with the 934-K $\alpha \to \beta$ crystallographic phase transition.) Accuracies of ± 0.2% were reported for the Flotow and Lohr heat-pulse data, and approximately ± 0.3% for the Ginnings and Corruccini data, which were obtained by the "drop" method using an ice calorimeter. The two sets of data interface smoothly, and pertain to high-purity (>99.96-at.% U) polycrystalline samples.

The published smoothed tabulations of C_p, the constant-pressure heat capacity, were corrected to constant volume (C_v) using the thermodynamic equality

$$C_p - C_v = \alpha_v^2 V T \beta^{-1} \ ,$$

where α_v is the volume thermal-expansion coefficient, V is the molar volume, and β is the volume compressibility. The necessary information to correct C_p to C_v appears in the literature for all α-phase temperatures.[26,27] At room temperature

C_p and C_v differ by 3%. The correction of C_v to the volume of the low-temperature experiment at 10 kbar was made using the thermodynamic relation

$$\left(\frac{\partial C_v}{\partial V}\right)_T = T\left(\frac{\partial(\alpha/\beta)}{\partial T}\right)_v .$$

C_v can be represented as the sum $C_L + C_E$, where the lattice heat capacity, C_L, consists of harmonic (C_{Lh}) and anharmonic (C_{La}) contributions, and C_E consists of noninteracting one-electron contributions C_{Eb} plus the many-body enhancement contributions C_{Ee},

$$C_v = C_L + C_E = (C_{Lh} + C_{La}) + (C_{Eb} + C_{Ee}) . \qquad (4)$$

Throughout the temperature region of the analysis, the dominant term in Eq. (4) is C_{Lh}, which can be represented by a Debye function. The term C_{La} is defined relative to the choice of the Debye temperature. To obtain a Debye temperature that is characteristic of all phonon modes (rather than the lowest energy modes obtained from elastic constant and low-temperature heat-capacity measurements), it is necessary to analyze the heat-capacity data at temperatures at which C_{Lh} is greater than one-half the classical limit of $3R$. The harmonic lattice heat capacity approaches the classical limit according to the expression

$$C_{Lh} = 3R\left(1 - \frac{1}{12}\frac{\hbar^2\langle\omega^2\rangle}{k_B^2 T^2} - \cdots\right) ,$$

where $\langle\omega^2\rangle$ is the second moment of the phonon spectrum.[28] In the Debye model, $\langle\omega^2\rangle = [(3k_B^2)/(5\hbar^2)]\,\Theta_\infty^2$, where Θ_∞ is the high-temperature Debye temperature. Θ_∞ is obtained graphically in Fig. 1 from the slope of a $(C_v - 3R)/T$-vs-T^{-3} plot in the region $0.7 \le T/\Theta_\infty \le 1.3$. (This procedure is valid assuming C_{La} and C_E are predominantly linear.) A value of $\Theta_\infty = 175$ K is obtained. Using this value of Θ_∞ and the room-temperature Grüneisen constant[26] of 2.35, the high-temperature linear coefficient of C_{La} is obtained from Wallace's recent empirical correlation.[29] (Although the correlation was derived for cubic solids, Wallace found that it also described C_{La} for some hexagonal-close-packed metals, even with nonideal c/a ratios.) A high-temperature value of C_{La}/T of − 0.8 mJ/°K² mole was obtained. The error limits for C_{La}/T will be considered at the end of this section.

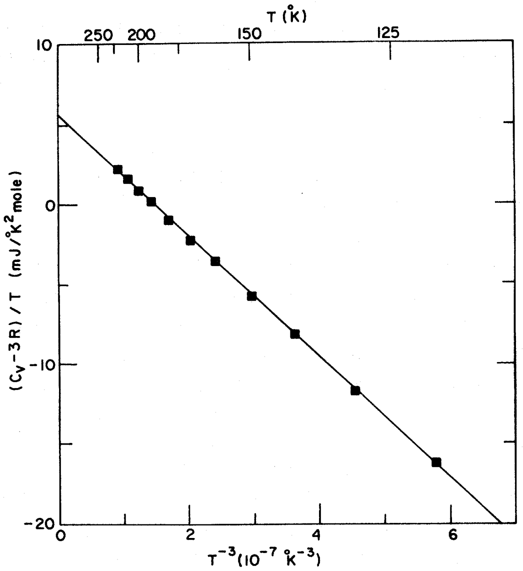

FIG. 1. Heat capacity of α-uranium plotted as $(C_v - 3R)/T$ vs T^{-3} using the smoothed data of Ref. 24. The slope of the straight line indicates that Θ_∞ is 175 K.

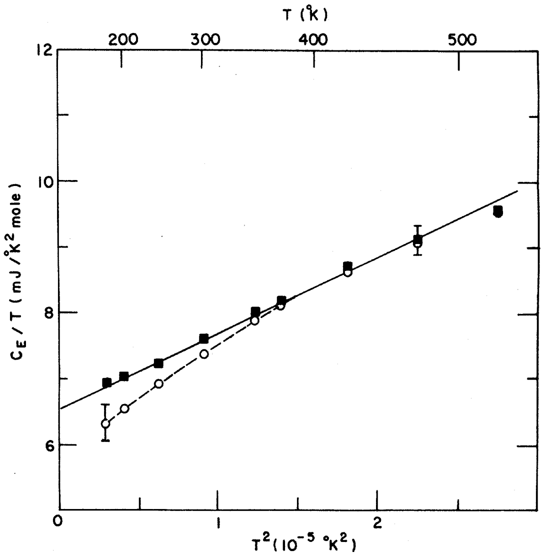

FIG. 2. Electronic heat capacity of α-uranium divided by temperature (circles and dashed curve) plotted vs T^2 using the smoothed data of Refs. 24 and 25. The error bars represent the reported accuracies of ± 0.2% below 350 K and ± 0.3% above 350 K. Also plotted are the same results corrected for the electron-phonon renormalization contribution C_{Ee}, based on Grimvall's calculation (Ref. 21) adjusted to a Debye phonon spectrum (squares). Note that C_{Ee} is negative in this temperature range. The straight-line extrapolation to $T = 0$ K yields a γ_b value of 6.5 mJ/°K² mole.

The electronic heat capacity divided by temperature versus T^2 for $T > \Theta_\infty$ is plotted in Fig. 2. (The lowest-order correction to C_{Eb}/T is, in general, proportional to T^2.) The dashed curve and circles include the electron-phonon renormalization contribution C_{Ee}, which is negative in this temperature region. At the highest temperatures, the plot is essentially linear and gives a $T = 0$ K intercept of $\gamma_b = 6.5$ mJ/°K² mole; hence $\lambda = \gamma_0/\gamma_b - 1 = 0.9$. [A value of $(C_V - C_L)/T$ can be obtained from the $T=\infty$ intercept of Fig. 1, however, Fig. 2 provides a firmer characterization of C_{Eb}/T since the value of Θ_∞ and electron-phonon renormalization effects become unimportant in a higher-temperature analysis.] Using these values of λ and γ_b, C_{Ee} can be obtained from Grimvall's model calculation,[21] adjusted to a Debye phonon spectrum. (Grimvall assumed that the noninteracting one-electron density of states is constant but, for the present purpose, C_{Ee} is not sensitive to that assumption.) Subtracting C_{Ee} from C_E, all data points in Fig. 2 become consistent with the linear extrapolation of C_{Eb}/T (the solid line). The equation

$$C_{Eb}=6.5\,T+1.2\times10^{-5}T^3\ \text{mJ/°K mole} \tag{5}$$

is obtained. At room temperature C_E is 8% of the measured heat capacity.

To check for any obvious thermodynamic discrepancy, the individual contributions to the measured C_p are represented in terms of reasonable extrapolation functions, and the entropy is calculated and compared with the measured entropy at high temperatures. The harmonic lattice is represented by a Debye function with $\Theta_\infty = 175$ K; the anharmonic terms C_{La} and $C_p - C_v$ are represented by AC_p^2T functions, where the A values are constants determined at room temperature from the C_{La} and $C_p - C_v$ values; and C_E is represented by Eq. (5) and Grimvall's enhancement contribution[21] adjusted to a Debye phonon spectrum. At all temperatures above Θ_∞, the calculated entropy agrees with the measured entropy to well within the 0.2% uncertainty in the measured entropy. The agreement is excellent but, of course, the extrapolation functions do not represent the heat-capacity contributions at low temperatures, primarily because the metal does not remain in the α phase.

Three sources of error are present in the analysis: (a) the experimental data errors in γ_0; (b) the uncertainty of the extrapolation used to obtain γ_b; and (c) the uncertainty of the high-temperature anharmonic contribution C_{La}. This last uncertainty is the major source of error in λ. (For $T>\Theta_\infty$ and for T removed from any phase transformation, C_{La} is generally linear to a good approximation—according to both experimental observation and theoretical expectation.[28,29] An absolute error in C_{La}/T produces the same absolute error in γ_b.) Based on the observed deviations from Wallace's empirical expression for constant-volume anharmonicity[29] (for the 18 solids he used in establishing a correlation), C_{La} is reliable to within a factor of 2 for α-uranium. Hence, the errors for γ_b are essentially $\pm C_{La}/T$ and $\lambda=0.9\pm0.2$.

III. DISCUSSION

As emphasized earlier, a determination of the magnitude of λ provides a criterion for choosing between proposals iii and iv for the dominant mechanism underlying the superconductivity of uranium metal. A μ^* of $0.27^{+0.07}_{-0.08}$ is calculated from Eq. (1) using $T_c=2$ K, $\Theta=175$ K, and $\lambda=0.9\pm0.2$. This is greater than the value of μ^* of 0.2 obtained from Bennemann and Garland's empirical correlation with $N_b(0)$.[30] (It is also greater than the generally accepted μ^* value of 0.10 for simple metals and of 0.13 for transition metals.[20]) This indicates that large Cooper-pair-weakening interactions are probably present in α-uranium. The upper limit of μ^* would suggest that short-lived spin fluctuations play a role in modifying the superconductivity of uranium.

The BCS theory, as originally formulated,[31] predicted that T_c is proportional to M^ξ, where $\xi=-\frac{1}{2}$. This mass dependence of T_c arises from the mass dependence of the phonon frequencies (i.e., $\Theta \propto M^{-1/2}$). Morel and Anderson[32] first demonstrated that, for low-T_c superconductors, deviations from the BCS isotope effect generally arise from the mass dependence of μ^*. This is due to the cutoff in the phonon spectrum since

$$\mu^*=\mu\,(1+\mu\ln E_B/\omega_c)^{-1}\ ,$$

where the screening of the Coulomb interaction μ depends on the ratio of E_B, the electronic bandwidth, to ω_c, the phonon cutoff frequency. McMillan obtained the isotope-effect coefficient[20]

$$\xi=-\frac{1}{2}\left[1-\left(\mu^*\ln\frac{\Theta}{1.45\,T_c}\right)^2\frac{1+0.62\lambda}{1+\lambda}\right]\ . \tag{6}$$

A value of ξ equal to $0.0^{+0.3}_{-0.2}$ is calculated from Eq. (6) using $T_c=2$ K, $\Theta=175$ K, $\lambda=0.9\pm0.2$, and $\mu^*=0.27^{+0.07}_{-0.08}$. This value of ξ is substantially different than the experimentally[11] deduced value of 2 obtained at 11 kbar; however, both the measured and calculated values of ξ differ markedly from a value of $-\frac{1}{2}$.

The volume change associated with isotopic substitution can be estimated from the zero-point motion and the compressibility. Since ^{235}U has a larger volume than ^{238}U and the more recent experiments[33] indicate that $\partial T_c/\partial P>0$ at 11 kbar, small positive deviations from the calculated isotope effect are expected. Also, it has been argued that if a sharp pressure-sensitive peak in the electronic density of states occurs within $\hbar\omega_c$ of the Fermi energy, even the magnitude of the measured isotope effect could be explained.[14,19] However, such a feature in the electronic density of states would produce pronounced variations in C_{Eb}/T below 500 K, which are not present. Realistic band-structure calculations for α-uranium would be quite useful for further clarification.

To understand the dominant mechanism responsible for the low-pressure depression of T_c relative to the 2-K superconducting α-phase T_c, we consider the influence of the volume minimum on the single-electron density of states. It is known that the zero-pressure γ_0 value for uranium metal is significantly less than the α-phase value[4] of 12.2 mJ/°K^2 mole. At zero pressure, the γ_0 value depends on grain size.[3] Single crystals have lower γ_0 values than polycrystals, and a single-crystal γ_0 value of 9.14 mJ/°K^2 mole has been obtained in two independent studies.[3,34] Based on Eqs. (2) and (3), this reduction of γ_0 relative to the α-phase γ_0 value is due to a reduction of λ, $N_b(0)$, or both. Ross and Lam[35] reported that at zero pressure χ is large, paramagnetic, and decreases 5% on cooling from 300 to 4.2 K. They interpreted the measured χ as being dominated by Pauli and orbital (χ_0) contributions from d or f electrons. The ratio of the electronic density of states obtained from χ and γ_b (N_χ/N_{γ_b}) is ~4. The deviation from a ratio of unity indicates that the χ_0 contribution to χ is large, that χ_p is exchange enhanced, or both. Since the $T = 0$ K intercept of the measured[35] χ is ~3% less than that extrapolated on a χ-vs-T^2 plot from $T > \Theta_\infty$, it can be concluded that $N_b(0)$ is reduced ~3 to ~12% because of the low-temperature phase transition. (The 3% reduction assumes that the factor of four obtained as the ratio of N_χ/N_{γ_b} is due to exchange enhancement of χ_p, and the 12% reduction assumes that χ_0 is $\frac{3}{4}$ of χ.) To estimate $\lambda' = \gamma_0'/\gamma_b' - 1$, where the primes are the single-crystal zero-pressure values, it is sufficient (a) to obtain the value of γ_b from a high-temperature heat-capacity analysis, using the $T = 0$-K volume of quenched-in α phase at zero pressure, and (b) to reduce this value of γ_b by (3–12)%, which will yield γ_b', to take into account the change in $N_b(0)$ associated with the low-temperature phase transition. Proceeding in the manner outlined above, a λ' of ~0.4 is obtained. A combination of this λ' value with reasonable values of Θ and μ^* is consistent with the single-crystal zero-pressure T_c $\lesssim 0.1$ K inferred from bulk measurements.

From the analysis presented above, it is clear that large Cooper-pair-weakening interactions are probably present in uranium *and* that λ is pressure sensitive. The changes in the electron-phonon mass enhancement are primarily responsible for the initial pressure-dependent T_c of uranium metal. These shifts in λ reflect phonon-mode shifting that occurs under pressure. Additional manifestations of phonon-mode shifting are the pronounced anomalies in the elastic moduli[9,10] at the low-temperature volume minimum, the negative Grüneisen parameters[16] at low temperatures, and the 22- and 37-K phase transitions found only for single-crystal uranium.[10,34,36] Experimental information (i.e., inelastic neutron scattering or superconductive tunneling), which is unavailable at present, would be useful in determining which phonon modes are most important in influencing the superconductivity of uranium.

IV. CONCLUSIONS

Using both the high- and low-temperature electronic heat-capacity coefficients, a λ of 0.9 ± 0.2 was found for the 2-K superconducting α phase of uranium. Using McMillan's equation,[20] a μ^* of $0.27^{+0.08}_{-0.07}$ and an isotope effect $T_c \propto M^\xi$, where $\xi = 0.0^{+0.3}_{-0.2}$, were calculated for α-uranium. The reduction in the γ_0 value and T_c of single-crystal uranium at zero pressure, relative to that of the pressure-stabilized α phase, is largely due to a substantial reduction in λ. In general, shifts in λ are a consequence of phonon-mode shifting. Specifically, such shifts in uranium are also evident in the pronounced anomalies in the elastic moduli,[9,10] at the low-temperature volume minimum, and in the negative Grüneisen parameters at low temperatures.[16]

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