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CRITICAL FIELD CURVES FOR THE ThU ALLOY SYSTEM

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Critical field curves for the ThU alloy system

by

Harlan Leroy Watson

A Dissertation Submitted to the
Graduate Faculty in Partial Fulfillment of
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Critical field curves for the ThU alloy system

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The superconducting critical field curves have been measured for the ThU alloy system and have been found to be in agreement with the BCS theory critical field curves to an accuracy of 1%, thus providing additional experimental evidence that the rapid depression of the superconducting transition temperature of pure thorium by the uranium impurities is due to pair-weakening rather than to pair-breaking mechanisms. Additional experimental evidence is provided by the fact that the reduced specific heat jumps at T_c for these alloys, obtained from the slope of the superconducting critical field curves and Rutger's formula, are also in close agreement with the BCS theory. Within the accuracy of the experimental data, the electronic specific heat coefficient, γ , remains essentially constant.

I. INTRODUCTION

A. Historical Survey

Since the discovery of superconductivity by Kamerlingh Onnes (1) in 1911, the phenomenon has been the object of numerous experimental and theoretical investigations. Only a few of the important experiments and calculations which bear directly on this work will be presented here. For more extensive reviews of the subject, the reader is referred elsewhere (2-12).

One of the most easily observed properties of a superconductor is that below some transition temperature T_c , its electrical resistivity, ρ , is for all practical purposes, zero. Measurements have established an upper limit on the electrical resistivity in the superconducting state of about $10^{-23} \Omega\text{-cm}$ (13), a figure which may be compared to the lowest electrical resistivity measured in the normal state of metals of about $10^{-9} \Omega\text{-cm}$ (14). Another important property is that superconductivity may be destroyed by the application of a magnetic field at any temperature T below T_c . Experimentally, it is found for all superconductors that the temperature dependence of this critical field, $H_c(T)$, is given approximately by

$$h \equiv \frac{H_c(T)}{H_c(0)} \approx 1 - t^2 \equiv 1 - (T/T_c)^2 \quad (1)$$

In 1932 Keesom and Kok (15) found that in the absence

of a magnetic field there is a jump in the electronic specific heat at the transition temperature characteristic of a second-order phase transition to a new thermodynamic phase (16). Keesom (17), Rutgers (18), and Gorter (19) applied thermodynamics to the superconducting transition, but the validity of these treatments was questioned because the phase transition was thought to be irreversible. This objection was removed, however, when Meissner and Ochsenfeld (20) determined that the magnetic induction, B , vanishes inside a bulk superconductor regardless of the magnetic field history of the sample. This phenomenon, known as the Meissner effect, showed that the superconducting state in an external magnetic field is a single stable state to which the laws of thermodynamics apply. Soon after the discovery of the Meissner effect, Gorter and Casimir (21) presented a full thermodynamic treatment of the superconducting phase transition.

The phenomenological two-fluid model of superconductivity was introduced by Gorter and Casimir (22) in 1934. In this model it is assumed that a superconductor possesses an ordered or condensed state characterized by an order parameter which varies from zero at $T = T_c$ to unity at $T = 0$ K, thereby indicating the fraction of the total system in the superconducting state. Further, it is assumed that the entire entropy of the system is due to the disorder of the particles

in the normal state. The application of thermodynamics to the model yielded a parabolic critical field curve and an electronic specific heat varying as T^3 , both in general agreement with experiment. The electrodynamics of this model was worked out by F. and H. London (23). The London theory predicts that the magnetic field does not abruptly disappear at the surface of a bulk superconducting specimen, but falls off exponentially with distance into the specimen, with a characteristic distance called the penetration depth, λ , a quantity first measured by Shoenberg (24) in 1940. For most common superconductors $\lambda \sim 10^{-6}$ cm to 10^{-5} cm.

The London model is based on a point by point relation between the current density and the vector potential associated with the magnetic field. This implies that the wavefunctions of the superconducting electrons are governed by interactions which have a range much smaller than the penetration depth. On the basis of empirical evidence, however, Pippard (25) concluded that the current density, rather than being proportional to the vector potential at the point in question, is given by the spatial average of the vector potential over a region of extent $\xi \sim 10^{-4}$ cm. Pippard called the quantity, ξ , the coherence length and found empirically that

$$\xi = 0.15(\hbar v_F / k_B T_C) \quad , \quad (2)$$

where $\hbar$ is Planck's constant divided by 2π , v_F is the Fermi

velocity, and k_B is Boltzmann's constant. Microscopic theory confirms Equation 2 but the constant 0.15 is replaced by 0.18. The superconducting state is now characterized by a long range coherence such that substantial changes in the superconducting wavefunctions cannot occur over a distance less than about 10^{-4} cm in pure metals. In a superconductor, then, there are two important characteristic lengths, λ which determines the spatial variation of magnetic fields and ξ which determines the spatial variations of the fraction of electrons in the superconducting ground state. Thus, we may classify superconductors as either type I ($\xi > \lambda$) or type II ($\xi < \lambda$). Because only type I materials will be considered in this thesis, no more will be said about type II behavior.

The first success in understanding the origin of the superconducting state was made in 1950 when Fröhlich (26) noted that virtual emission and absorption of phonons by electrons could produce an attractive interaction between electrons. The importance of the electron-phonon interaction was soon verified experimentally by the discovery of the isotope effect (27) which showed that the transition temperature and isotopic mass, M , obeyed the approximate relation $T_C M^{1/2} = \text{constant}$. An additional breakthrough came when improved thermal conductivity (28) and specific heat measurements (29) and infrared absorption (30) measurements suggested the existence of an energy gap in the spectrum of the

electrons in superconductors. About this same time, Cooper (31) showed that any net attraction, no matter how small, between electrons can lead to a condensed state in which electron pairs are virtually excited above the Fermi surface. Finally, in 1957, a successful microscopic theory of superconductivity was presented by Bardeen, Cooper, and Schrieffer (32). This model, known as the BCS model, will be discussed in more detail later.

B. Thermodynamic Theory of the Superconducting Transition

Consider an ellipsoidal superconducting specimen of volume V with a magnetic moment $I = MV$, positioned in a solenoid such that the applied magnetic field H is parallel to the specimen's axis. All quantities will be expressed in cgs units. The relevant thermodynamic potential for the system is the Gibbs free energy

$$G = U - TS + PV - HI \quad , \quad (3)$$

where U = internal energy, T = temperature, S = entropy, and P = pressure. Differentiation of Equation 3 gives

$$dG = dU - TdS - SdT + PdV + VdP - HdI - IdH \quad . \quad (4)$$

From the second law of thermodynamics

$$dU = TdS - PdV + HdI \quad , \quad (5)$$

where HdI is the work done on the sample by the solenoid's power supply to change the sample's magnetic moment by dI . Substitution of Equation 5 into Equation 4 yields

$$dG = VdP - SdT - IdH \quad . \quad (6)$$

Integration of Equation 6 gives

$$G(T, P, H) = G(T, P, 0) - V \int_0^H M dH \quad . \quad (7)$$

At $H = H_c$, the superconducting state Gibbs energy, G_s , is equal to the normal state Gibbs free energy, G_n , so

$$G_s(T, P, H_c) = G_n(T, P, H_c) \quad . \quad (8)$$

The Gibbs free energy difference in zero field is then

$$\Delta G = G_n(T, P, 0) - G_s(T, P, 0) = -V \int_0^H (M_s - M_n) dH \quad , \quad (9)$$

where M_s and M_n are the magnetizations of the superconducting and normal states, respectively.

Electromagnetic theory gives the relation

$$B = H + 4\pi M \quad . \quad (10)$$

The Meissner effect shows that $B = 0$ inside a superconductor, hence

$$M_s = -H/4\pi \quad . \quad (11)$$

The normal state magnetization M_n is negligible compared with M_s and is taken to be zero. For a long cylindrical sample the demagnetization factor, D , is zero and the superconducting transition takes place at H_c . For an ellipsoidal sample with a different value of D , the field is distorted in the neighborhood of the sample and when the applied field reaches a value $(1-D)H_c$, the sample goes into the intermediate state in which there is coexistence of macroscopic normal and superconducting regions. Regardless of the demagnetization factor,

$$-V \int_0^H M_s dH = H_c^2 / 8\pi = \Delta G \quad , \quad (12)$$

because the magnetic work is done solely in the superconducting portion of the sample.

The differences in the entropy and specific heat are given by

$$\Delta S = S_n - S_s = -\frac{\partial}{\partial T}(G_n - G_s)_{P,H} = -\frac{VH_c}{4\pi}(\partial H_c / \partial T)_P \quad (13)$$

and

$$\begin{aligned} \Delta C = C_n - C_s &= T \frac{\partial}{\partial T}(S_n - S_s)_P \\ &= -\frac{VT}{4\pi} H_c (\partial^2 H_c / \partial T^2)_P - \frac{VT}{4\pi} (\partial H_c / \partial T)_P^2 \quad , \end{aligned} \quad (14)$$

respectively. The third law of thermodynamics requires that

$\Delta S = 0$ at $T = 0$ K, and Equation 13 shows that this requires $(\partial H/\partial T) = 0$ at $T = 0$ K. Further, $\Delta S = 0$ when $T = T_c$ since $H_c = 0$. Thus in zero field, the superconducting transition is accomplished without latent heat and is, therefore, a second-order phase transition. The specific heat jump at T_c is given by Rutger's formula (16)

$$\Delta C(T_c) = - \frac{VT_c}{4\pi} \left[\partial H_c(T_c) / \partial T \right]^2_p \quad (15)$$

On the basis of theoretical and experimental evidence the following assumptions are made concerning the entropy of a superconductor (33):

- (i) The entropies in the normal and superconducting states may be expressed as the sums of the independent lattice (S_g) and electronic (S_e) contributions (i.e., $S_n = S_{gn} + S_{en}$ and $S_s = S_{gs} + S_{es}$).
- (ii) For sufficiently low T , $S_{en} \gg S_{es}$.
- (iii) The normal electron entropy is given by the free electron model result (i.e., $S_{en}(T) = \gamma T$).
- (iv) The lattice entropy is unchanged in the superconducting transition (i.e., $S_{gn} = S_{gs}$).

Equation 13 now becomes

$$\gamma T = - \frac{VH_C}{4\pi} \left(\partial H_C / \partial T \right)_P \quad (16)$$

Integration of Equation 16 from 0 to T gives the important result

$$H_C^2 = H_0^2 - 4\pi(\gamma/V)T^2 \quad , \quad (17)$$

where $H_0 = H_C(0)$. The BCS theory predicts that Equation 17 is valid below $t = 0.25$, a result that has been experimentally verified (33). Thus critical field measurements at sufficiently low temperatures allow the determination of both H_0 and γ .

C. BCS Microscopic Theory of Superconductivity

The BCS microscopic theory of superconductivity was successful in explaining the principal experimental facts of superconductivity discussed previously: (1) a second-order phase transition at T_C ; (2) zero resistivity; (3) the Meissner effect; (4) an energy gap for single particle excitations; and (5) the isotope effect.

BCS assumes that in a metal there is an attractive force between electron pairs caused by virtual phonon exchange and a repulsive force due to the screened Coulomb interaction and when the combined effect of the two forces is attractive, the metal becomes superconducting. Because superconductivity is fairly common in metals and is not

associated with any particular crystal structure, the net attractive interaction is assumed isotropic and is replaced by a constant attractive interaction $-V'$ within a characteristic energy $\hbar\omega$ of the Fermi surface. The normal state is described by the Bloch individual-particle model while the superconducting ground state is formed by taking a linear combination of many low-lying normal state configurations in which the Bloch states are virtually occupied in pairs of opposite spin and momentum $(\vec{k}\uparrow, -\vec{k}\downarrow)$. These paired states are referred to as Cooper pairs. The transition temperature is related to the characteristic energy, $\hbar\omega$, the density of electron states for one spin direction at the Fermi energy, $N(0) = N(\epsilon_F)$, and the attractive interaction, $-V'$, in the weak coupling limit $N(0)V' \ll 1$ by the relation

$$k_B T_c = 1.14 \hbar\omega \exp[-1/N(0)V'] \quad . \quad (18)$$

Since the most attractive part of the interaction is due to high frequency phonons, $\hbar\omega$ is expected to be of the order of $k_B \theta_D$, where θ_D is the Debye temperature. In actual practice $\hbar\omega$ is assumed to be approximately $0.75 \hbar\omega$ (34), so Equation 18 is usually seen in the form

$$T_c = 0.85 \theta_D \exp[-1/N(0)V'] \quad . \quad (19)$$

The Gibbs free energy difference at $T = 0$ K is

$$\Delta G/V = 1/2 N(0) [\Delta(0)]^2 , \quad (20)$$

where $2\Delta(0)$ is the energy gap at $T = 0$ K. Using the free electron model's result for the electronic specific heat coefficient γ ,

$$\gamma = \frac{2}{3} \pi^2 k_B^2 N(0) V , \quad (21)$$

and Equation 12 we get

$$\Delta(0) = H_0 (\pi k_B^2 V / 6\gamma)^{1/2} . \quad (22)$$

Mühlschlegel (35) calculated the thermodynamic functions using the BCS theory in the weak coupling limit and expressed the results in the following form

$$(V H_c^2 / 8\pi) / \gamma T_c^2 = \frac{3}{2} t^2 (x \frac{da}{dx} - a) , \quad (23)$$

$$S_{es} / \gamma T_c = t [1 + 3(x \frac{da}{dx} - a) - \frac{3x}{2}] , \quad (24)$$

where

$$x = [\Delta(T) / \pi k_B T]^2 , \quad (25)$$

$2\Delta(T)$ = energy gap at temperature T ,

$$\Delta(0) = \frac{\pi}{\gamma_e} k_B T_c = 1.764 k_B T_c , \quad (26)$$

$\ln \gamma_e$ = Euler's constant ,

and

$$a(x) = -\frac{2}{\pi} \int_{-\infty}^{\infty} \ln(1 + e^{-\pi(x+u^2)^{1/2}}) du \\ + x(\ln \gamma_e \sqrt{x} - 1/2) + 1/3 \quad . \quad (27)$$

The BCS critical field calculations agree remarkably well with experimental data for pure superconductors. Deviations from BCS in pure superconductors are primarily due to strong-coupling effects, exhibited by Hg (33) and Pb (36), for example, and anisotropy effects, exhibited in Al (37), for example. Strong-coupling theory (38, 39), which uses the actual phonon spectrum of the superconductor to calculate the electron-phonon interaction matrix element, has improved agreement between theory and critical field data of Hg and Pb. Clem (40) modified the critical field calculation by including an anisotropic form of the electron-phonon interaction matrix element and this theory has explained a number of experiments, notably the effects of alloying on superconductivity and the deviation of low T_c superconductors from BCS.

D. Addition of Impurities to a Pure Metal

The properties of both normal and superconducting metals containing small concentrations of impurities with partially filled d- or f- electron shells have been the subject of extensive experimental and theoretical investigations. An isolated atom with unfilled d or f states generally has a spin and orbital angular momentum according to Hund's rules, and it frequently happens that the perturbations acting in a metal on an impurity are of insufficient strength to destroy the magnetic moment. A typical example of such local moment behavior occurs when atoms of 3d transition elements are dissolved in noble metals. In such a case, the scattering of the conduction electrons may depend upon the relative orientation of the spins of the impurity atoms and, in addition, both potential and spin-flip scattering may occur. These scatterings can give rise to various anomalous properties, such as the appearance of a Kondo term

$$\rho \propto -A n \ln T \quad (28)$$

in the low temperature electrical resistivity for an impurity concentration n , a giant thermoelectric power, and other related phenomena. A number of excellent review articles have treated the local moment problem for both normal (41-44) and superconducting (45-51) metals, and for more detailed discussions of the problem, the reader is

referred to these reviews and the references cited therein.

J. Friedel (52) first gave an explanation of the local moment in terms of the concept of the "virtual bound state". The basic idea of his theory is that one has an element I with a partially filled ℓ band (ℓ denotes d or f) dissolved in a normal metal matrix M, and assumes that I has an atomic energy level which lies within the bandwidth of the s conduction electrons (Figure 1). The ℓ radial wavefunction presumably does not extend very far out of the I atomic cell, so it will retain some of its atomic behavior in the alloy. Because the energy of the ℓ radial wavefunction is higher than the bottom of the conduction band, it gets broadened by mixing with nearby free electron states having approximately the same energy in the conduction band. If the broadening is not too great, the electronic charge of I remains strongly localized in a virtual ℓ -bound state in order to keep the ionic charge of I equal to that of M. Another way of looking at the problem is to reduce the interaction to an equivalent scattering problem in which I gives up all its s and ℓ electrons to the conduction band of M, thereby creating a strong localized Coulomb potential well which is almost deep enough to have bound ℓ states for every ℓ electron given up. As a result, the ℓ components of the conduction electron wave functions are strongly attracted to the atomic cell of I when their energy is near that of the vacated ℓ levels. One

Figure 1. Virtual bound ℓ -state on an impurity atom I in a normal metal matrix M.

still has a virtual bound state localized on I, which appears here as a resonance in the potential of the I atomic cell.

In order to explain the presence of a net magnetization on I, Friedel pointed out that since the virtual bound states are almost as much localized as in an atom, one would expect exchange and correlation forces to be very effective for narrow bands, with a tendency to favor spin alignment. When these forces prevail the virtual bound states have different energies for opposite spin alignment so that they accommodate unequal numbers of electrons up to the Fermi level and a localized magnetic moment exists.

In 1961, Anderson (53) put these ideas on a more quantitative basis by solving the following problem in the Hartree-Fock approximation at $T = 0$ K. Assume that a localized moment exists (Figure 2a) and can be represented by a single ℓ -orbital level whose energy is a distance $E_\ell = E$ below the Fermi level, ϵ_F , so that it is occupied by an electron of say, spin-up. A spin-down electron attempting to occupy the same orbital will see the full Coulomb repulsion, V_c , of the spin-up state because the exclusion principle allows the electrons to get relatively close to one another. The spin-down electron can only occupy an orbital with energy $E + V_c$, which must be empty if a local moment exists and, therefore, must lie above the Fermi level. Now, as before,

(a) MAGNETIC

$$\Delta = \pi \langle V_{kl}^2 \rangle N(\epsilon_F)$$

(b) NONMAGNETIC

Figure 2. Anderson model density of states distributions in (a) magnetic and (b) nonmagnetic cases.

the conduction electrons can, through the mixing interaction, $V_{k\ell}$, mix with the electron in the localized state and cause that level and the empty level above ϵ_F to be shifted and broadened with half-width Δ . The expectation value $\langle n\uparrow \rangle$ now becomes less than unity, while the expectation value $\langle n\downarrow \rangle$ becomes finite. These changes are such as to decrease the difference between the spin-up and spin-down energies: E moves up to $E + \langle n\downarrow \rangle V_C$ and $E + V_C$ moves down to $E + \langle n\uparrow \rangle V_C$. If $V_{k\ell}$ is strong enough, the situation eventually breaks down cooperatively and the system becomes unstable so that it is no longer possible to maintain a localized moment and we have a nonmagnetic state (Figure 2b). Anderson (53) formulated a Hamiltonian for this model and the solutions of this Hamiltonian allowed him to define the transition curve, shown in Figure 3, separating magnetic and nonmagnetic regions. An examination of Figure 3 reveals that in the Anderson model the magnetic regime exists for $V_C \gg \Delta$ when $(\epsilon_F - E)/V_C$ is neither too small nor too near unity, while the nonmagnetic regime exists for either $V_C \ll \Delta$ or when $(\epsilon_F - E)/V_C$ is close to 0.5 for relevant values of V_C/Δ . The Anderson model can be generalized to the case where there are $2(2\ell+1)$ degenerate ℓ levels, which fill to the Fermi level with approximately the number $\langle N \rangle$, of ℓ -electrons appropriate for the impurity being considered. Taking the Fermi level as the zero of energy, the level centers are separated from it by an energy

Figure 3. Anderson model magnetic-nonmagnetic phase diagram.

E_ℓ which depends on $\langle N \rangle$ and the level width, Δ , according to

$$E_\ell = \Delta \cot[\pi \langle N \rangle / 2(2\ell+1)] \quad , \quad (29)$$

where

$$\Delta = \pi N(0) \langle v_{k\ell}^2 \rangle \quad . \quad (30)$$

For a single level, the ℓ -electron density of states $N_\ell(0)$ is

$$N_\ell(0) = \Delta / \pi (E_\ell^2 + \Delta^2) \quad , \quad (31)$$

and the magnetic-nonmagnetic phase boundary is defined by the relation

$$\sin^2[\pi \langle N \rangle / 2(2\ell+1)] = \pi \Delta / V_c \quad . \quad (32)$$

Kondo (54) gave the first theoretical explanation of the resistance minimum which appears in the low temperature electrical resistivity of metals containing magnetic impurities (hence the name Kondo effect). He assumed that a local moment exists and characterized the impurity by assigning it a spin $\vec{S}$ which interacts with the conduction electrons through an exchange interaction represented by the interaction Hamiltonian

$$H_{\text{int.}} = - 2 J \vec{S} \cdot \vec{s} \quad , \quad (33)$$

where J is the strength of the exchange interaction and $\vec{s}$

is the average conduction electron spin density at the impurity site. A perturbation calculation of the conduction electron-impurity scattering to third order in J gave, for $J < 0$, a magnetic contribution to the electrical resistivity proportional to $-\ln T$, in agreement with experiment, but the expression diverged for negative J below a characteristic temperature of

$$T_K \sim T_F \exp[-1/N(0)|J|] \quad , \quad (34)$$

where $T_F = \varepsilon_F/k_B$ is the Fermi temperature. Subsequent applications of more sophisticated techniques to try to remove the divergence and to explain properties for temperatures both above and below the Kondo temperature, T_K , has led to the physical interpretation of T_K as a characteristic temperature below which the impurity spins tend to be compensated by the conduction electron spins, the degree of compensation increasing with decreasing temperature.

A crucial factor in all these theories is the sign and magnitude of the exchange interaction parameter, J , so it is important to understand the factors which determine these quantities. The ordinary atomic exchange J_0 is always positive, but, as Anderson and Clogston (55) first pointed out, the Anderson type mixing can lead to a negative term for J . Indeed, Schrieffer and Wolff (56) performed a canonical transformation on the Anderson Hamiltonian and found that

in the limit of small mixing between the local moment and conduction electron states, the Anderson Hamiltonian became identical to the Kondo Hamiltonian. Generally, J is given approximately by

$$J \sim J_0 + J_1, \quad (35)$$

where $J_0(>0)$ is the atomic exchange term and $J_1(<0)$ arises from the mixing between the conduction electrons and local impurity states. For transition metal solutes in noble metal matrices the dominant contribution to J is often J_1 and $J<0$, leading to Kondo type behavior, while for rare earth solutes, with the exception of Ce, J_0 is the dominant contribution to J , hence the absence of the Kondo effect. In terms of the Schrieffer-Wolff transformation J_1 is given by

$$J_1 \sim - \frac{\langle v_{k\ell}^2 \rangle V_c}{|E_\ell| (E_\ell + V_c)} \quad (36)$$

when $|E_\ell| \gg \Delta$. Here E_ℓ is the energy separating the local state and the Fermi level.

The Hartee-Fock approximation used by Anderson in his model excludes the dynamics of the problem. The operational definition of the existence of a local moment is that the excess susceptibility of the alloy over that of the pure host approximates a Curie law behavior, at least above some

temperature. This requires that the spin polarization on the impurity be long-lived, but not infinitely long-lived as in Anderson's model. Extensions of the theory to include time-dependent Hartree-Fock approximations have led to the development of a localized spin fluctuation (lsf) model (57). According to the lsf model, even when a system is nonmagnetic in the Anderson sense ($V_c < \pi\Delta$), the system can suffer large fluctuations in spin density. Physically, we might view this as an electron hopping onto the impurity state and remaining there for some characteristic lsf time, $\tau_{sf} \sim h/\Delta$, before hopping off. Associated with τ_{sf} is a characteristic temperature $T_0 \sim h/k_B\tau_{sf}$. In the limit $\tau_{sf} \gg \tau_T$, where τ_T is the thermal fluctuation lifetime ($\sim h/k_BT$), the impurities behave magnetically, while in the limit $\tau_{sf} \ll \tau_T$, the impurities behave nonmagnetically. This is equivalent to saying that when the temperature becomes of the order T_0 , the lsf becomes stable against thermally induced reorientation, and, at this temperature and above, there is no physical difference between the spin fluctuation and a genuine spin. Hence, the system behaves as if it were magnetic. For this theory the low temperature electrical resistivity is predicted to follow a $\rho_0[1 - (T/T_0)^2]$ behavior. Such behavior was first observed in AlMn and AlCr by Caplin and Rizzuto (58).

The local moment problem is considerably more complicated

if the impurity host undergoes a superconducting transition. When the impurities are nonmagnetic atoms with filled d- or f- shells, the superconducting transition temperature is changed by at most a few percent, as first found in studies using Sn (59), and Al and In (60), as superconducting hosts. For sufficiently small concentrations of all solutes these studies showed that T_c decreased linearly with increasing reciprocal electronic mean free path, l_e . For larger impurity concentrations, the curves of T_c versus concentration fell into two groups, according to the sign of the valence difference, ΔZ , between the impurity and host. T_c had a sharp upward curvature for higher valence solutes, while for those with lower valence, T_c tended to saturate for large concentration. This behavior was in sharp contrast to the rapid depression of T_c which had been observed in superconductors containing 3d-transition (61) and 4f-rare earth (62) magnetic impurities, where the addition of a few tenths of atomic percent of impurity sometimes completely suppressed superconductivity. The small depression of T_c in the nonmagnetic impurity case was difficult to understand within the framework of the BCS microscopic theory of superconductivity for the following reason. In the BCS theory it was shown that a net attractive interaction using time-reversed Bloch states led to the formation of Cooper pairs which have a binding energy of about 10^{-4} eV at $T = 0$ K. In

very "dirty" samples where $l_e \sim \xi$, the collision time τ is of the order of 10^{-14} sec, corresponding to an energy uncertainty of about 0.1 eV and all superconducting effects would be expected to change drastically, contrary to the experimental result.

Anderson's theory of dirty superconductors (63) provided an explanation of the experimental results discussed above. He suggested that the anisotropy of the energy gap present in the pure superconductor may be smoothed out by the impurity scattering. In the pure metal, the energy gap is anisotropic, depending on the momentum direction of the electrons, and T_c is primarily dependent on the large gap values. The impurity scattering mixes Bloch states of the conduction band. Therefore, the energy gap and T_c will decrease with decreasing l_e , until, when $l_e \sim \xi$, the impurity scattering becomes so large that Bloch states from all parts of the Fermi surface are mixed with more or less equal weight, thereby removing the source of anisotropy in the energy gap. Once the mixing is complete, the effect of l_e on T_c saturates. The formation of Cooper pairs then takes place between the time-reversed states which are exact one electron states of the impure metal. Anderson showed that if the scatterers are nonmagnetic, the time-reversed states are degenerate in energy and lead to the formation of Cooper pairs with infinite lifetimes as in BCS theory. If the

impurities are magnetic, however, time-reversal invariance is removed, the time-reversed scattered states are no longer degenerate in energy, and the Cooper pairs now have finite lifetimes. Such a pair-breaking interaction inhibits superconducting electron correlation and large decreases in T_c occur.

The basic features of the T_c versus concentration $[T_c(n)]$ studies of La containing rare earth impurities by Matthias et al. (62) were successfully explained by Abrikosov and Gor'kov (64) in 1960. The Abrikosov-Gor'kov (AG) theory makes three basic assumptions: (1) the paramagnetic impurity spins $\vec{S}$ are randomly distributed on the host lattice; (2) the impurity spins are unpolarized and uncorrelated; and (3) an impurity spin interacts with the average electron spin density at the impurity site via the exchange interaction given by Equation 33. The presence of the paramagnetic impurities leads to the formation of Cooper pairs with a finite lifetime τ . Within the first Born approximation, the inverse lifetime τ^{-1} (or pair-breaking parameter α) is given by

$$\alpha \equiv \tau^{-1} = \hbar^{-1} n N(0) J^2 S(S+1) \quad , \quad (37)$$

where n is the impurity concentration. For rare earth impurities where the orbital angular momentum is not quenched, $\vec{S}$ must be replaced by its projection on the total angular

momentum vector $\vec{j}$ (65). In this case, Equation 33 becomes

$$\chi_{\text{int.}} = -2J(g-1) \vec{j} \cdot \vec{s} \quad , \quad (38)$$

where g is the Landé g -factor, and Equation 37 becomes

$$\alpha \equiv \tau^{-1} = \hbar^{-1} n N(0) J(g-1)^2 j(j+1) \quad . \quad (39)$$

The theory predicts a second-order phase transition to the superconducting state in the absence of a magnetic field, and connects the reduced temperature T_c/T_{cp} to the reduced pair-breaking parameter α/α_{cr} through the relation

$$\ln(T_c/T_{cp}) = \psi(1/2) - \psi(1/2 + \frac{1}{4\gamma_e} \frac{\alpha}{\alpha_{cr}} \frac{T_{cp}}{T_c}) \quad . \quad (40)$$

Here ψ is the digamma function, T_{cp} and T_c are the superconducting transition temperatures of the pure host and alloy, respectively, and $\alpha_{cr} \equiv k_B T_{cp} / 4\gamma_e$ is the critical value of the pair-breaking parameter required to destroy superconductivity. In the limit of small α/α_{cr} , Equation 40 has the asymptotic form

$$\lim_{\frac{\alpha}{\alpha_{cr}} \rightarrow 0} T_c/T_{cp} = 1 - 0.691 (\alpha/\alpha_{cr}) \quad . \quad (41)$$

If α is varied only by changing the impurity concentration n , then α/α_{cr} in Equations 40 and 41 may be replaced by n/n_{cr} , where n_{cr} is the critical concentration at which

$$T_c = 0.$$

The AG theory also made the remarkable prediction that for an impurity concentration greater than $0.91 n_{cr}$, the energy gap in the single particle density of states disappears and the phenomenon of gapless superconductivity occurs. This results from the fact that the finite Cooper lifetime τ leads to an energy broadening $\Gamma \sim \hbar/\tau$ which introduces states into the gap and spreads out the BCS density of states. As a consequence of this energy broadening, the energy gap goes to zero faster than the superconducting order parameter. This leads to a specific heat which varies linearly with temperature rather than exponentially in the gapless region.

There exists a large body of experimental evidence which verifies the correctness of the AG theory and its extensions: Skalski et al. (66) have calculated the density of states, critical field, discontinuity in the specific heat at T_c , penetration depth, and complex conductivity; and Ambegaokar and Griffin (67) have calculated the thermal conductivity for an AG material. A few of the more relevant experiments confirming the theory are the following: tunneling measurements on PbGd by Wolff and Reif (68); specific heat measurements on bulk LaGd alloys by Finnemore et al. (69); tunneling measurements on Sn and Sn-In alloys by Millstein and Tinkham (70); critical field measurements on

ThGd by Decker et al. (71) and Decker and Finnemore (72); $T_c(n)$ measurements to $n = 0.9 n_{cr}$ on (La,Gd) Al_2 by Maple (73) which are shown in Figure 4; thermal conductivity measurements on ThGd by Cappelletti and Finnemore (74); and far-infrared measurements on PbGd by Dick and Reif (75).

Large departures from the AG $T_c(n)$ curves have been observed for $n > 0.7 n_{cr}$ for LaGd (76) and (La,Gd) $_3$ In (77), and Bennemann (78) has attributed these departures to the magnetic ordering of impurity spins. According to Bennemann's theory, the magnetic ordering of the impurity spins leads to three effects: (1) Zeeman splitting of the Fermi surface, which is destructive to superconductivity; (2) reduction of spin-flip scattering which enhances superconductivity; and (3) spin-orbit scattering, which reduces the effect of the Zeeman splitting of the Fermi surface and, therefore, enhances superconductivity. By considering the interplay of these three effects, Bennemann was able to show qualitative agreement with the $T_c(n)$ curves for LaGd and (La,Gd) $_3$ In. Additional corrections to the AG critical field curves to include additional pair-breaking effects have been given by Fulde and Maki (79) and Bennemann, Garland, and Mueller (80). Since these corrections deal with type II superconductors, the reader is referred to these papers and to the review articles by Parks (49), Bennemann (50), and Maki (51) for further discussion.

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Figure 4. Reduced transition temperature T_c/T_{cp} versus reduced concentration n/n_{cr} for $(La,Gd)Al_2$ from Maple (73), where $T_{cp} = 3.24$ K and $n_{cr} = 0.590$ at.% Gd. The solid curve represents the result of the AG theory (Equation 40).

Recently, the problem of paramagnetic impurities in superconductors has been reinvestigated by including the Kondo effect. According to Müller-Hartmann and Zittartz (81) and Ludwig and Zuckermann (82), Equation 40 still describes a superconducting Kondo system if the pair-breaking parameter is given by

$$\alpha = \frac{n}{\hbar(2\pi)^2 N(0)} \frac{\pi^2 S(S+1)}{\ln^2 T_c/T_K + \pi^2 S(S+1)}, \quad (42)$$

which has a maximum at T_K given by Equation 34. $T_c(n)$ can then be calculated by simultaneously solving Equations 40 and 42. Several examples are shown in Figure 5 from the work of Ludwig and Zuckermann (82) for $T_{cp} = 9.17$ K and $S = 1/2$. For $T_K \geq T_{cp}$ (curves A and C), $T_c(n)$ initially decreases linearly with increasing n after which it curves positively away from linearity in contrast to the AG result. A particularly interesting result can occur when $T_K \ll T_{cp}$ (curve B). For certain values of n , Equations 40 and 42 yield three solutions for $T_c(n)$. As the temperature is lowered, an alloy of the appropriate composition should first become superconducting at a temperature $T_{c1}(n)$, remain superconducting to a lower temperature $T_{c2}(n)$ at which it becomes normal, and then remain normal to yet a lower temperature $T_{c3}(n)$ at which it again becomes superconducting. The above behavior is understandable in view of the temperature

Figure 5. T_c/T_{cp} versus impurity concentration n for various values of T_{cp}/T_K from Ludwig and Zuckermann (82).

dependent pair-breaking given by Equation 42. For $T_{cp} \gg T_K$, the alloy becomes superconducting at an upper transition temperature where the pair-breaking is still small. When the temperature is lowered, pair-breaking increases strongly as T_K is approached, and superconductivity is again suppressed. Superconductivity then reappears for $T < T_K$, where the temperature is much less than the resonant temperature.

A reentrant $T_c(n)$ curve has been observed in the $(La,Ce)Al_2$ system (83, 84), where $T_K \ll T_{cp}$, but no evidence was found for a third transition temperature even though measurements were extended to 0.006 K. The extension of the Kondo effect by Maki (85) to type II superconductors is roughly in agreement with critical field curve measurements of this alloy system (86). Also, the specific heat jump at T_c determined from specific heat measurements on $(La,Ce)Al_2$ (87) are depressed below the AG value, in agreement with recent calculations by Müller-Hartmann and Zittartz (88). This effect has also been observed in the $ZnMn$ system (89), where $T_K < T_{cp}$.

Boato et al. (90) first pointed out that nonmagnetic resonant states, viewed in the sense of the Friedel-Anderson model, could affect superconductivity. When they added Fe group impurities to Al, for example, they found that T_c was depressed at a rate intermediate between the strong depression due to magnetic impurities and the weak depression observed for nontransition metal impurities. For Fe group impurities in Al, the resonant state widths are so large

($\Delta \gtrsim 1$ eV) that a local moment cannot exist. However, virtual bound states present at the impurity sites increase the density of states of electrons near each impurity, and the resultant resonance scattering can depress T_c below the value expected for a simple nonmagnetic impurity.

The first theoretical treatment of the effect of non-magnetic impurity resonant states on superconductivity was presented by Zuckermann (91). He found the initial linear decrease of T_c to be inversely proportional to Δ . In order to explain the experimental results, however, Zuckermann had to assume much too small widths for the resonant states. Ratto and Blandin (92) and Takanaka and Takano (93) extended Zuckermann's theory to include the Coulomb repulsion between d electrons of opposite spin on the impurity site. The calculation of Ratto and Blandin (RB) for the initial decrease in T_c gave fair agreement with the experimental results obtained for dilute Al-Fe group alloys by Aoki and Ohtsuka (94), the largest discrepancy between theory and experiment occurring for AlMn. Using the RB Hamiltonian, Kiwi and Zuckermann (95) calculated various thermodynamic properties for superconducting alloys, and they suggested that beyond a critical impurity concentration, gapless superconductivity might be observed. The latter was a surprising result in view of the fact that the RB Hamiltonian is time-reversal invariant and pair-weakening, in contrast to the pair-breaking result for paramagnetic impurities.

A more recent theoretical development concerning the effect of nonmagnetic localized states in superconducting alloys is due to Kaiser (96). Starting with the RB Hamiltonian, Kaiser carefully calculated the excitation spectrum and $T_c(n)$ for an isotropic BCS superconducting host. He found that Kiwi and Zuckermann's prediction of gaplessness was incorrect due to an error in their calculation of the order parameter, and showed that when the calculation was done correctly there exists no region of gapless superconductivity. According to this theory the alloys remain BCS like with

$$T_c = 1.14 \theta_D \exp(-1/f\lambda) \quad . \quad (43)$$

Here $\lambda = N(0)V'$ is the coupling constant of the pure BCS host and, for an impurity with angular momentum ℓ , the dilution factor f is given by

$$f = \frac{1 - ncd}{1 + nc} \quad , \quad (44)$$

where

$$c = \frac{(2\ell+1) N_\ell(0)}{N(0)} \quad (45)$$

and

$$d = \frac{N_\ell(0) v_c^{\text{eff}}}{\lambda} \quad . \quad (46)$$

$N_{\ell}(0)$ is given by Equation 31. The denominator in f is a dilution effect due to the participation of the ℓ -orbital electrons in superconductivity via the resonance scattering. The numerator is a pair-weakening effect due to the Coulomb repulsion between opposite spin ℓ -orbital electrons. The v_c^{eff} term in d is the effective Coulomb potential proposed by Schrieffer and Mattis (97),

$$v_c^{\text{eff}} = \frac{V_c}{1 + (V_c/\pi E_{\ell}) \tan^{-1}(E_{\ell}/\Delta)} \quad , \quad (47)$$

which arises from correlations whereby a spin-up electron hops onto an impurity site as a spin-down electron hops off, and vice versa.

The resultant dependence of $T_c(n)$ is anomalously steep in its initial descent and approximately exponential:

$$T_c = T_{cp} \exp[-An/(1 - Dn)] \quad , \quad (48)$$

where

$$A = c(1 + d)/\lambda \quad (49)$$

and

$$D = cd \quad . \quad (50)$$

A critical concentration for the complete suppression of superconductivity

$$n_{cr} = \frac{N(0)(2\ell + 1)\lambda}{N_{\ell}(0)^2 v_c^{eff}} \quad (51)$$

is also predicted by the Kaiser model.

The system which best seems to satisfy the conditions of the Kaiser theory is ThCe (98). Figure 6 shows the reduced transition temperature of ThCe alloys as a function of Ce concentration at various pressures compared with Kaiser's theory. In addition, systems exhibiting lsf behavior, such as AlMn, for example, are described by the modified exponential $T_c(n)$ proposed by Kaiser. Huber and Maple (99) have proposed that this behavior may be a general result for the superconducting-normal phase boundary when the characteristic spin fluctuation temperature $T_0 \gg T_{cp}$.

Morandi (100) and Riblet (101) have presented models to describe the effect of lsf on the superconductivity of Ir based alloys. Morandi used the exchange enhanced model of Lederer and Mills (102) and found an expression for $T_c(n)$ considerably more complicated than Kaiser's but which gave qualitative agreement with experiment. Riblet fitted his experimental $T_c(n)$ curves for IrNi, IrCo, and IrFe with the McMillan (103) expression modified by the addition of a paramagnon coupling constant proportional to the impurity concentration.

Bennemann (104) has taken a completely different

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Figure 6. Reduced transition temperature of ThCe alloys versus Ce concentration at various pressures from Huber and Maple (98). The solid lines represent the result of Kaiser's theory (Equation 48) fitted to the data by the method of least squares.

approach to this problem. He has assumed that localized spin fluctuations break pairs and obtained a $T_c(n)$ curve of the same general shape as the experimental $T_c(n)$ curves for AlMn and AlCr by assuming a pair-breaking parameter α which varied as T^2 . However, when the temperature dependence of α/α_{cr} for AlMn is calculated directly from Equation 40 and experiment, it is found to vary as $[a + b(T/T_{cp})^{1/2}]$ (46, 47). Also, the critical field curves of AlMn (105) and the specific heat jumps at T_c of AlMn (106) follow the BCS law of corresponding states, indicating that pair-breaking interactions are not present.

Rivier and MacLaughlin (107) have treated the passage from the nonmagnetic to the magnetic regime in superconducting alloys with a lsf model, and hence have bridged the gap between the AG and Kaiser theories. In their model, a single parameter, the characteristic spin fluctuation temperature, T_0 , characterizes the material and determines the degree to which a local moment picture applies. The AG result for $T_c(n)$ is obtained for temperatures $T \gtrsim T_0$ in the limit $T_0 \ll T_{cp}$, whereas an exponential dependence of T_c on n , similar to Kaiser's result, is found for $T_0 \gg T_{cp}$. The effects of lsf in both the weakly magnetic and strongly magnetic regimes have also been studied by Zuckermann (108) and Paton and Zuckermann (109).

E. Previous Measurements on ThU

Maple and coworkers (110) have presented strong evidence that the ThU alloy system shows lsf phenomena. A number of normal state properties point to this interpretation. The incremental normal state electrical resistivity, $\Delta\rho$, is described by the expression $\Delta\rho = \rho_0[1 - (T/T_0)^2]$, where $T_0 \sim 100$ K. This value of T_0 may be compared to 530 K for AlMn and 1200 K for AlCr, as determined from their low temperature electrical resistivities (58). The temperature of the resistance minimum, $T_{\min}$, obeys the relation $T_{\min} \propto n^{1/3}$, consistent with an incremental resistivity which varies as $[1 - (T/T_0)^2]$ and a lattice contribution that varies as T^5 for pure Th (111). This behavior of $\Delta\rho$ and $T_{\min}$ for ThU is quite different from the $\Delta\rho \propto -\ln T$ and $T_{\min} \propto n^{1/5}$ behavior found in Kondo systems (42, 43) where the impurities sustain well-defined local moments. Additional evidence for the presence of lsf was provided by the observation of a large peak in the thermoelectric power centered at approximately 70 K. Such "giant" thermoelectric powers have been observed in a number of impurity systems (42, 43, 112) peaking about their characteristic temperatures, whether they be Kondo or lsf systems. Finally, the effective moment determined from the incremental susceptibility, $\Delta\chi = \Delta\chi_{\text{ThU}} - \Delta\chi_{\text{Th}}$, decreased with decreasing temperature below approximately 170 K. In the temperature region from about 170 K to 300 K, $\Delta\chi$ could

be fitted to a Curie-Weiss law giving an effective moment of $3.43 \pm 0.20 \mu_B$, where μ_B is the Bohr magneton. This value of the effective moment is close to that expected for a U 5f-electron configuration of either two ($3.58 \mu_B$) or three ($3.62 \mu_B$) f electrons. All the above normal state measurements indicate that the ThU alloy system is described by a lsf model.

Superconductivity measurements of the ThU system give additional support to a lsf interpretation. Maple et al. (110) found an initially large rate of depression of T_C (7.95 K/at.% U) with $T_C(n)$ exhibiting positive curvature for increasing n, in sharp contrast to the AG theory prediction for a system with paramagnetic impurities. This $T_C(n)$ behavior is quite similar to that observed in the AlMn and AlCr systems (90) where T_C is depressed at large initial rates of 14 K/at.% Mn and 8.1 K/at.% Cr and then has a positive curvature at higher concentrations. Luengo et al. (113) have measured the low temperature specific heat of a number of ThU alloys in the normal and superconducting state and found that the jump in specific heat at T_C closely followed the BCS law of corresponding states, thereby indicating that the depression of T_C in this alloy system is due to pair-weakening, rather than pair-breaking interactions. They also found that the electronic specific heat coefficient was strongly enhanced at a rate $d\gamma/dn = 2.7 \text{ mJ/mole-K}^2 \text{ at.\% U}$

at the lowest U concentrations. If it is assumed that the initial increase in γ with U concentration is due to uranium 5f resonant states, the local density of states at the Fermi level (for one spin direction) contributed by each U atom would be 57 states/eV-U atom. Huber and Maple (114) have found that the depression of T_c for ThU alloys decreases with pressure, and proposed that this result indicates that the lifetime of the 1sf decreases with pressure.

The $T_c(n)$ data of Maple et al. (110) have been compared with a number of previously mentioned models. The pair-breaking theory of Müller-Hartmann and Zittartz (81) gives a good fit by assuming $S = 1/2$ and $T_K/T_{cp} = 32$ in Equation 42. However, in this theory, the calculated specific heat jumps at T_c in the limit $T_K/T_{cp} \gg 1$ tend towards AG (88), contrary to the specific heat results. Kaiser's theory also gives a good fit (46, 47), but the density of states obtained from the depression of T_c is small and negative, a clearly unphysical result.

Maple et al. (110) found that Morandi's model (100) gave a close fit. They also used Equation 40 and their experimental data to find the temperature dependence of the AG pair-breaking parameter α and found that $\alpha \propto (T/T_{cp})^{1/2}$, which is in disagreement with the Bennemann pair-breaking model's prediction of $\alpha \propto T^2$ (104). Riblet has also found good agreement with $T_c(n)$ for ThU using his model (101).

F. Objectives

The objective of this study has been to determine the magnetic state of U impurities in Th from their effect on the superconductivity of Th by measuring the critical field curves, $H_c(T)$, of a number of ThU alloys and comparing the experimental results with the AG and BCS theories. An additional comparison was possible because the specific heat jump at T_c calculated from the theories could be compared with the value calculated from the slope of the experimental $H_c(T)$ curve at T_c using Rutger's formula, Equation 15.

II. EXPERIMENTAL APPARATUS AND PROCEDURE

A. Sample Preparation and Analysis

The ThU and pure Th samples were provided by D. T. Peterson's group of this laboratory. The procedure in the ThU preparation was to first prepare a master alloy containing 1.88 at.% U by arc melting together the appropriate constituents, under an argon atmosphere in a water cooled copper hearth, with a tungsten tip. The master alloy was then separated into small pieces and diluted with pure thorium, by the above method, to attain the desired concentrations. The samples, which weighed approximately 11.5 g, were then swaged to dimensions of approximately 0.1 inches in diameter by 1.5 inches in length, electropolished in a perchloric acid and methanol solution, sealed in a Ta crucible and annealed at 1200°C for one week, then slow cooled to room temperature.

A chemical analysis was performed on all the ThU samples by J. S. Fritz's group of this laboratory using a spectrophotometric method which was accurate to the nearest 0.001 at.% U. The results are given in Table 1. The solid solubility of U in Th has been reported to be 1.5 at.% at 800°C (115), and so for the low U concentrations used in this study, we are assured of having good solid solutions (116).

Table 1. Atomic percent U determined by chemical analysis

Nominal at.% U	Run 1	Run 2	Average at.% U
0.025	0.030	0.030	0.030
0.050	0.051	0.053	0.052
0.050	0.060	0.057	0.059
0.075	0.076	0.077	0.077
0.100	0.101	0.102	0.102
0.125	0.126	0.127	0.127
0.150	0.149	0.150	0.150
0.150	0.161	0.161	0.161
0.200	0.212	0.217	0.215

B. Cryostat

Temperatures ranging from 1.4 K to 0.3 K were attained using the He^3 cryostat shown in Figure 7. Temperatures above 1.2 K were obtained by admitting 5 or 6 mm of He^3 gas to the He^3 system to thermally link the He^3 and He^4 evaporation chambers. Temperatures below 1.2 K were maintained by pumping on He^3 which had been condensed in the He^3 evaporation chamber.

A copper holder for three symmetrically spaced coil forms and the germanium resistance thermometer, GR-928, was bolted onto the bottom of the He^3 evaporation chamber. The surfaces in this and other pressure contacts were coated with Apiezon N grease in order to improve thermal conductivity at the contact boundary. Heat leaks into the system via

Figure 7. Cross-section of He³ cryostat.

electrical connections were minimized by cementing the leads from the germanium resistance thermometer and transition sensing coils to the coil holder, He^3 evaporation chamber, and He^4 evaporation chamber with Stycast 2850 GT, and radiation effects were minimized by enclosing the coil holder with a copper heat shield connected to the He^3 evaporation chamber.

The coils, which were used to sense the superconducting transition, were wound on linen fiber coil forms 1.8 inches long with 0.5 inch outer diameters and 0.125 inch bores. In order to make ac susceptibility measurements, each coil was wound with about 1500 turns of No. 40 copper wire on the primary, while each half of the astatic secondary contained 5000 turns of No. 40 copper wire. The turns were cemented to the coil forms with GE 7031 adhesive.

One end of the superconducting samples was coated with Apiezon N grease and placed in a 0.125 inch collar. Sufficient sample extended above the collar to fill the lower half of the transition sensing coil when the collar was held in the yoke attached to the bottom of the sample holder.

The vacuum jacket can was soldered to the can top by Wood's metal and this seal could easily be broken to change samples. The can top also contained an electrical feed-through constructed with nylon and Stycast 2850 GT to allow the electrical leads to pass from the He^4 bath to the vacuum space.

C. Thermometry and Temperature Control

Temperatures were measured using a Honeywell germanium resistance thermometer, GR-928. This thermometer had previously been calibrated by W. R. Decker (117) against the susceptibility of cerium-magnesium-nitrate (CMN) at 58 points from 0.3 K to 1.3 K, and had been calibrated against the T-58 He⁴ vapor pressure tables (118) at 12 points by W. R. Decker (117) and at 49 points by L. J. Williams (119) from 1.3 K to 4.2 K to form a complete set of data from 0.3 K to 4.2 K.

Figure 8 is a block diagram of the direct current four-lead potentiometric method used to measure the resistance of the germanium resistance thermometer. The resistance was obtained by comparing the voltage drop across the unknown resistance (GR-928) with the voltage drop across the General Radio Type 500-H 1 k Ω standard resistor for a fixed current. The germanium resistance thermometer constant current power supply is shown in Figure 9. Other components of the system included a Guideline Type 9180-B potentiometer, a constant current potentiometer power supply, an Eppley standard cell (No. 788370), and a Keithley 150B null detector. Circuit ground was at the low side of the null detector. The constant current potentiometer power supply (120) employed an Analog Devices Model 184J operational amplifier. A temperature compensated Zener diode was used as an input reference voltage. The output current drifted about 1 ppm per day.

Figure 8. Germanium resistance thermometer circuit.

Figure 9. Germanium resistance thermometer constant current power supply.

The potentiometer was standardized by adjusting the output current of the power supply.

The temperature control circuit is shown in Figure 10. The temperature control circuit used a three-lead direct current Wheatstone bridge which compared the resistance of a Speer carbon resistor (1/2W, nominal 470 Ω , grade 1002) mounted on the He³ evaporation chamber with a variable decade resistance (General Radio Type 1434-X decade resistor). The off-balance bridge voltage was amplified by a Keithley Model 150AR null detector and sent to a transistor controlled heater power supply which turned the heater current on (off) whenever the Speer carbon resistor's resistance became greater (less) than that of the decade resistor. The heater consisted of about 100 Ω of No. 36 manganin wire wound astatically and cemented to the He³ evaporation chamber with Stycast 2850 GT. The temperature could be controlled with Speer resistance variations of less than 0.1 Ω , the minimum resistance of the decade resistor. Typically, temperatures were maintained within a precision of 0.0001 K for the 1/2 hour periods during which a data point was measured.

D. Magnetic Field Sources

The applied field at the samples was generated by a liquid nitrogen cooled solenoid with an inner diameter of four inches and length of ten inches. The solenoid was

Figure 10. Wheatstone bridge used for temperature control.

wound with No. 14 square polythermalize coated copper wire with 1138 turns in eight layers and 128 turns overlaid on the ends to compensate for end effects. The layers were separated by 0.031 inch thick teflon strips placed parallel to the solenoid's axis to allow penetration of liquid nitrogen through the windings. A magnetic field homogeneity measurement using a Bell 240 Incremental Gaussmeter showed axial field homogeneity within 0.11% over a range of 3.81 cm and within 0.24% over a range of 5.72 cm from the center of the solenoid. The solenoid was calibrated at liquid nitrogen temperatures by nuclear magnetic resonance in water. Data points were taken at the center of the solenoid for six fields between 1260 Oe and 1360 Oe and yielded a value of 56.599 Oe/A.

The solenoid was powered by a Kepco Model KS 36-30M DC power supply with current regulation stable to one part in 10^4 . The output current of the power supply was programmed with external resistance provided by a Dekabox DB62 decade resistor and a General Radio Type 1432-N decade resistor connected in series. An increase in the external resistance by 0.1Ω increased the field by approximately 0.060 Oe. The resistance of the solenoid was measured by the direct current four-lead potentiometric circuit shown in Figure 11. Components of the circuit included a Leeds and Northrup K-3 potentiometer, a Dynage BSTC-2C constant voltage supply, an Eppley standard cell (No. 775042), a Leeds and Northrup

Figure 11. Magnetic field measurement circuit.

0.1 Ω , 15A shunt, and a Keithley 153 null detector.

The earth's magnetic field was compensated to approximately 0.01 Oe with a Helmholtz pair. The current and position required by the Helmholtz pair to cancel the horizontal and vertical components of the earth's magnetic field in the sample region of the cryostat were determined using the Bell 240 Incremental Gaussmeter. The Helmholtz pair power supply is shown in Figure 12.

E. Mutual Inductance Bridge

The superconducting transition temperatures of the samples were initially determined using the modified Hartshorn mutual inductance bridge shown in Figure 13. Similar bridges have been described by others (121-124).

The 32.05 Hz oscillator with a quartz crystal frequency control drove the primary circuit via a Gertsch Model CRT12AT coaxial ratio transformer. The output of the secondary of the fixed reference mutual inductance lump provided a constant voltage to the Gertsch Model 1011 AC ratio transformer. Proper adjustment of the ratio transformer balanced the inductive component of the signal from the secondary of the sample measuring coils. Quadrature phase signals due to eddy current losses and nonideal inductors in the circuit were nulled out by the resistive network. The voltage introduced into the detection circuit was proportional to

Figure 12. Helmholtz pair power supply.

Figure 13. 32.05 Hz mutual inductance bridge.

$1/(111\ 100 - R)$ where R is the setting of the variable resistors. A locally built dual phase lock-in detector (125, 126) coupled to the circuit through a 35:1 Geoformer acted as a null detector.

When the superconducting sample undergoes a transition from the normal to the superconducting state, it becomes diamagnetic and thus changes the mutual inductance between the primary and secondary of the coils surrounding the sample. This, in turn, varies the reference voltages necessary for a null condition in the secondary circuit.

F. Critical Field Measurements

The superconducting transitions in the applied magnetic fields were determined isothermally by a ballistic induction technique first reported by Cochran et al. (127). The sample, which filled the lower half of the astatically wound transition sensing coil, was placed in the cryostat at the center of the liquid nitrogen cooled solenoid with the magnetic field slightly less than H_c . The applied magnetic field was increased stepwise through H_c and changes in flux linking the sample were measured by a ballistic galvanometer in series with the transition sensing coil. The average permeability of the specimen as it made a superconducting to normal transition was determined from the galvanometer deflection accompanying each step, since each deflection

was proportional to the flux which enters the sample. The average permeability of the sample at the m^{th} step is defined as

$$\mu_m = \frac{\sum_{i=1}^m \delta_i}{\sum_{\text{all steps}} \delta_i}, \quad (52)$$

where δ_i is the deflection accompanying the i^{th} step. H_c was then determined by extrapolating a plot of μ versus the applied field to $\mu = 1$ for the temperature maintained. The procedure followed in taking data was to stabilize the temperature, wait about 15 minutes, then measure the superconducting to normal and normal to superconducting transitions.

III. RESULTS AND DISCUSSION

Critical field curve data which were obtained for seven different ThU alloys are listed in Appendix A and shown graphically in Figures 14 and 15. The data show excellent agreement with the BCS critical field curves indicated by the solid curves, but marked disagreement with the AG theory critical field curves, as illustrated for Th-0.127 at.% U by the dashed curve. The procedure used to obtain the theoretical curves will be discussed in detail later.

The primary factor limiting the accuracy of these results is the presence of hysteresis in the superconducting transitions. For our purposes we define hysteresis as the magnitude of the difference between the values of H_c obtained in increasing and decreasing magnetic fields for a given transition. The effect of annealing on the magnitude of the hysteresis is illustrated in Figure 16 by the superconducting transition obtained for Th-0.059 at.% U near $T = 0.350$ K. The transition after the sample had been annealed at 800°C for one-half hour is indicated by the open circles and the magnitude of the hysteresis is approximately $6\% H_c$. Annealing at temperatures close to the alloy phase transition tended to homogenize the samples and, in addition to reducing the hysteresis considerably, the transitions were generally sharpened. As shown in Figure 16, the hysteresis loop after the anneal tended to center on the previously

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Figure 14. Critical field data for the ThU alloys exhibiting small hysteresis plotted versus temperature. The solid curves indicate the BCS theory critical field curves and the dashed curve indicates the AG theory critical field curve for Th-0.127 at.% U. The bars on the Th-0.030 at.% U and the Th-0.059 at.% U data show the width of the superconducting transitions.

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Figure 15. Critical field data for the ThU alloys exhibiting large hysteresis plotted versus temperature. The solid curves indicate the BCS critical field curves. The bars on the data show the width of the superconducting transitions.

Figure 16. Superconducting transitions for Th-0.059 at.% U annealed at 800°C for one-half hour (open circles) and at 1200°C for one week (dark circles). H_c is an average critical field for the transition with the least hysteresis.

obtained larger hysteresis loop. Because of this behavior, the value of H_C for the smaller hysteresis loop was chosen as the midpoint of the values of H_C obtained in increasing and decreasing magnetic fields.

Generally speaking, the magnitude of the hysteresis found for the annealed samples was about a 1% effect. The Th-0.059, -0.102, -0.127, and -0.150 at.% U samples each had a maximum hysteresis of about 1% H_C , but there were deviations from this pattern. For example, the Th-0.030 at.% U sample had a maximum hysteresis of about 3% H_C at lower temperatures, and for two samples, Th-0.052 at.% U and Th-0.077 at.% U, the superconducting transitions observed at low temperatures were very broad and the maximum hysteresis was as large as 7% H_C . For some reason, the 1200°C anneal did not improve the quality of these samples. Hence, the low temperature values of H_C obtained for these two samples are much less accurate than the low temperature values of H_C obtained for the other samples and probably should be ignored. The magnitudes of the hysteresis obtained for the superconducting transitions of the alloys are listed in Appendix A.

One additional feature of the superconducting transitions obtained for the ThU alloys is illustrated in Figure 16. Each of the alloys displayed the same total galvanometer deflections if one stepped through a given

superconducting transition several times in increasing and decreasing magnetic fields, indicating that the alloys exhibited the Meissner effect.

The superconducting transition temperatures for the above alloys were determined by least squares fitting the $H_c(T)$ data near $T = T_c$ to $H_c(T) = A + BT^2$ and extrapolating to $H_c = 0$. In addition, the transition temperatures for the nominally pure Th sample, T_{cp} , and for the Th-0.161 at.% U and Th-0.215 at.% U samples were determined from a careful ac susceptibility measurement. The $T_c(n)$ data are presented in Figure 17 where the reduced transition temperature, T_c/T_{cp} , for the alloys are plotted versus uranium concentration. The solid line in the figure represents the result of the AG theory (Equation 40) with $T_{cp} = 1.360$ K and $n_{cr} = 0.157$ at.% U. The value of n_{cr} was obtained by using Equation 41 to fit the lower uranium concentration alloys. These $T_c(n)$ data are in good agreement with those obtained by Maple et al. (110). The depression of T_c is initially linear with increasing U concentration and then exhibits positive curvature for larger concentrations, in sharp disagreement with the result of the AG theory (64) for a superconductor containing paramagnetic impurities but in good agreement with the Kaiser theory (96).

The next step in the analysis of the $H_c(T)$ data was to compare the $H_c(T)$ data with the predictions of the AG and

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Figure 17. Reduced transition temperature, (T_c/T_{cp}) , of ThU versus U concentration. The solid curve represents the result of the AG theory (Equation 40) with $T_{cp} = 1.360$ K and $n_{cr} = 0.157$ at.% U. The bars on the one point indicate the uncertainty in the uranium concentration for that sample (Th-0.215 at.% U).

the BCS theories. The ratios of T_c/T_{cp} for each alloy for which $H_c(T)$ data were obtained were used to calculate the AG theory critical field curves (66) using the method explained in detail by Decker (117). When these calculations were completed, it was found that the AG theory $H_c(T)$ values were considerably smaller than the $H_c(T)$ data, as shown in detail in Figure 14 where the AG theory critical field curve for Th-0.127 at.% U is indicated by the dashed curve. Indeed, the $H_c(T)$ data lay much closer to the BCS theory critical field curves.

In order to compare the data with the BCS theory in greater detail, it was necessary to determine a value of H_0 for each alloy. The usual procedure used for obtaining H_0 is to fit experimental $H_c(T)$ data to Equation 17 for $T < 0.25 T_c$, thereby determining both H_0 and γ for a given sample, but this method was inapplicable here because the He^3 evaporation refrigerator used in this experiment was unable to attain sufficiently low temperatures. Hence, instead of using Equation 17 to determine H_0 and γ , we have used two different methods. First, we fitted the data to the simple BCS theory to verify that the theory applied rather well, and then we used a more complicated anisotropy corrected BCS expression (40) for the final fitting.

For the analysis where simple BCS was used, values of H_0 were obtained for all values of $t^2 = (T/T_c)^2 < 0.4$ for

each alloy by using the BCS critical field calculations made by Swenson (128). The average value of H_0 determined by this procedure was then taken to be the value of H_0 for a particular alloy as shown in Figures 18 and 19. H_0 derived in this way is constant within the accuracy of the data, so a simple BCS extrapolation fits the data rather well. A value of H_0 could not be obtained for Th-0.150 at.% U using this procedure because $T_c = 0.495$ K and sufficiently low reduced temperatures could not be attained with the He^3 evaporation refrigerator. The results of these H_0 calculations are given in Appendix B. A BCS critical field curve was then generated for each alloy using the values of T_c and H_0 . Figures 14 and 15 show the excellent agreement between the data and the BCS critical field curves which are indicated by the solid lines.

The critical field curves are also presented in Figures 20-25 as deviations from the fiducial parabola given by Equation 1, where the deviation function, $D(t)$, is given by

$$D(t) = h - (1 - t^2) \quad . \quad (53)$$

Data over the whole temperature range for all the alloys lay within about 0.3% of the BCS theory, indicated by the solid curves. These results can be contrasted to the behavior of $D(t)$ versus t^2 for the ThGd alloy system studied by Decker and Finnemore (72) (hereafter referred to as DF). The DF

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Figure 18. Results of the calculations of H_0 from the BCS theory for the ThU samples exhibiting small hysteresis. The bars on the Th-0.030 at.% U data show the width of the superconducting transitions. The dark lines indicate the average value of H_0 obtained.

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Figure 19. Results of the calculations of H_0 from the BCS theory for the ThU samples exhibiting large hysteresis. The bars on the data show the width of the superconducting transitions. The dark lines indicate the average value of H_0 obtained.

Figure 20. Deviation of the Th-0.030 at.% U critical field curve from the fiducial parabola compared to the BCS theory (solid curve).

Figure 21. Deviation of the Th-0.052 at.% U critical field curve from the fiducial parabola compared to the BCS theory (solid curve).

Figure 22. Deviation of the Th-0.059 at.% U critical field curve from the fiducial parabola compared to the BCS theory (solid curve).

Figure 23. Deviation of the Th-0.077 at.% U critical field curve from the fiducial parabola compared to the BCS theory (solid curve).

Figure 24. Deviation of the Th-0.102 at.% U critical field curve from the fiducial parabola compared to the BCS theory (solid curve).

Figure 25. Deviation of the Th-0.127 at.% U critical field curve from the fiducial parabola compared to the BCS theory (solid curve).

data are presented in Figure 26 and compared to the BCS and AG theories, indicated by the dashed curves and solid curves, respectively. The ThGd data lie considerably below the BCS theory curves and are in close agreement with the AG theory curves.

The electronic specific heat coefficient, γ , was also calculated from simple BCS using the relation (35)

$$\gamma = H_0^2 / 8\pi (0.2364) T_c^2 = 0.168 \frac{H_0^2}{T_c^2} V_M, \quad (54)$$

where V_M is the molar volume. Using Equation 54 with $V_M = 19.7 \text{ cm}^3/\text{mole}$ (117), $T_c = 1.390 \text{ K}$, and $H_0 = 159.22 \text{ Oe}$ for a pure Th sample, DF found that $\gamma = 4.34 \text{ mJ/mole-K}^2$, in close agreement with the calorimetric value of 4.31 mJ/mole-K^2 (129). The values of T_c and H_0 used by DF were obtained by the same method discussed above for determining T_c and H_0 for the ThU alloys. The results of the γ calculations for all the alloys are presented in Figure 27, where γ is plotted versus uranium concentration. The same data are also displayed in Figure 28 except the samples with large hysteresis have been excluded. Also included in these figures are calorimetric γ values for ThU by Luengo et al. (113) and the pure thorium γ of DF. An examination of the data indicates that γ values calculated from simple BCS are in reasonable agreement with the calorimetric γ values but

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Figure 26. Deviations of the ThGd critical field curves from the fiducial parabola compared to the BCS theory (dashed curves) and the AG theory (solid curves) from Decker and Finnemore (72).

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Figure 27. The electronic specific heat coefficient, γ , versus U concentration in ThU obtained from this study (solid squares) and from the calorimetric study (open circles) by Luengo et al. (113). The value of γ obtained for pure Th by Decker and Finnemore (72) is also shown (solid circle).

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Figure 28. The electronic specific heat coefficient, γ , versus U concentration for ThU obtained from this study for the samples exhibiting small hysteresis (solid squares) and from the calorimetric study (open circles) by Luengo et al. (113). The value of γ obtained for pure Th by Decker and Finnemore (72) is also shown (solid circle).

tend to lie somewhat lower if the samples with large hysteresis are excluded as in Figure 28. The general trend of these data indicates that γ is enhanced at a rate of about $2 \text{ mJ/mole-K}^2 \text{ at.}\% \text{ U}$. If it is assumed that the increase in γ with U concentration is due to uranium 5f resonant states, the local density of states at the Fermi level, for one spin direction, contributed by each U atom would be about 42 states/eV-atom, a rather large number.

The reduced specific heat jumps, $(\Delta C/\Delta C_p)_{T_c}$, are plotted versus the reduced transition temperatures in Figure 29. The values of $(\Delta C)_{T_c}$ were calculated by first fitting the data near T_c to $H_c = A + BT^2$ and then using Rutgers' formula (Equation 15). The value of $(\Delta C_p)_{T_c}$ was obtained by extrapolating the $(\Delta C)_{T_c}$ data to $T/T_{cp} = 1$, and this procedure yielded a value of $(\Delta C_p)_{T_c} = 8.75 \pm 0.25 \text{ mJ/mole-K}^2$, in good agreement with $(\Delta C_p)_{T_c} = 8.5 \pm 0.2 \text{ mJ/mole-K}^2$ obtained by Luengo et al. (113) and the value of $(\Delta C_p)_{T_c} = 9.0 \text{ mJ/mole-K}^2$ obtained by DF. The bars on the data points indicate the uncertainties in the slopes of the critical field curves near T_c . BCS (31) give the relation

$$\frac{(\Delta C)_{T_c}}{\gamma T_c} = 1.43 \quad , \quad (55)$$

and if γ is assumed to remain constant, then Equation 55 gives

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Figure 29. Reduced specific heat jump, $(\Delta C/\Delta C_p)_{T_c}$, versus T_c/T_{cp} . The dashed and the dot-dashed lines represent the BCS theory results, and the solid line is the AG theory result. The values obtained in this study of ThU are shown by the dark circles, while the values obtained by Luengo et al. (113) are shown by the open circles. Also included are results for ThGd (solid squares) by Decker and Finnemore (72) and for (La,Ce)Al₂ (solid triangles) by Luengo, Maple, and Fertig (87).

$$(\Delta C / \Delta C_p)_{T_c} = T_c / T_{cp} \quad (56)$$

and one obtains the dashed line shown in Figure 29. If, however, γ changes, as T_c changes, as would be expected from Figure 28, then Equation 55 becomes

$$(\Delta C / \Delta C_p)_{T_c} = \gamma / \gamma_p T_c / T_{cp} \quad (57)$$

If it is assumed that γ is enhanced at a constant rate of about 2.0 mJ/mole-K² at.% U, then the dot-dashed line shown in Figure 29 is obtained. It is seen that the ThU data from both this study and Luengo et al. (113) lie close to the predictions of BCS theory given either by Equation 56 or Equation 57. Also shown in Figure 29 are the reduced specific heat jumps for ThGd (72) which closely follow the AG theory indicated by the solid curve, and the reduced specific heat jumps for the superconducting Kondo alloy system (La,Ce)Al₂ (87), which lie considerably below the AG theory result.

Measured and calculated values (using simple BCS) of various sample characteristics for the ThU alloys used in this study are given in Table 2. An anomaly which appears in the data is that the values of T_c for Th-0.052 at.%U and Th-0.059 at.% U are almost the same, even though the chemical analysis of these alloys indicated significantly different uranium concentrations. As noted above, however, the

Table 2. Measured and calculated (from simple BCS) values for ThU alloys

Sample	T_c (K)	H_0 (Oe)	γ (mJ/mole- K^2)	$(\Delta C/\Delta C_p)_{T_c}$
Th-0.000 at.% U	1.360 $\pm$ 0.001			1.000
Th-0.030 at.% U	1.182 $\pm$ 0.001	136.600 $\pm$ 1.454	4.42 $\pm$ 0.09	0.879
Th-0.052 at.% U	1.013 $\pm$ 0.001	117.961 $\pm$ 3.213	4.49 $\pm$ 0.25	0.782
Th-0.059 at.% U	1.014 $\pm$ 0.001	116.994 $\pm$ 0.567	4.41 $\pm$ 0.04	0.752
Th-0.077 at.% U	0.863 $\pm$ 0.001	101.624 $\pm$ 3.616	4.59 $\pm$ 0.33	0.668
Th-0.102 at.% U	0.725 $\pm$ 0.001	84.518 $\pm$ 0.212	4.50 $\pm$ 0.02	0.565
Th-0.127 at.% U	0.658 $\pm$ 0.001	76.544 $\pm$ 0.412	4.48 $\pm$ 0.02	0.514
Th-0.150 at.% U	0.495 $\pm$ 0.001			0.408
Th-0.161 at.% U	0.487 $\pm$ 0.001			
Th-0.215 at.% U	0.304 $\pm$ 0.001			

lower temperature superconducting transitions for the Th-0.052 at.% U sample exhibited large hysteresis and this could imply that the uranium atoms in this sample were not homogeneous. Because only a small portion of each sample was cut off and analyzed, clustering of uranium atoms in that portion could give a higher uranium concentration than actually exists in the bulk specimen. It should also be noted that the uranium concentrations of the ThU alloys reported in the specific heat study of Luengo et al. (113) are highly suspect because the T_c values differ significantly from those found by Maple et al. (110) and from those reported here. For example, Luengo et al. reported that no superconducting transition could be observed in their Th-0.134 at.% U sample for temperatures as low as $T \approx 0.3$ K, whereas Maple et al. (110) and this work found that a nominal Th-0.2 at.% U sample undergoes a superconducting transition above 0.3 K. Also Luengo et al. reported $T_c = 0.550$ K for Th-0.110 at.% U, which is significantly lower than the value of $T_c = 0.650$ K for Th-0.127 at.% U found in this study.

Before proceeding to the details of the final fitting of the critical field curve data to take into account anisotropy effects, we shall first provide a bit of theoretical background. Markowitz and Kadanoff (130) (hereafter referred to as MK) worked out the details of the Anderson anisotropy

idea (63) discussed earlier using a model in which the constant effective electron-electron interaction V' in the simple BCS model is replaced by the effective electron-electron matrix element

$$V'_{pp'} = (1 + a_p)V'(1 + a_{p'}) \quad , \quad (58)$$

where a_p is an anisotropy function chosen to have zero average over the Fermi surface. MK calculated the change in T_c , δT_c , for a pure superconductor containing simple non-magnetic impurities and found that

$$\delta T_c(\chi) = K^1 \chi + \langle a^2 \rangle T_c I_c(\chi) \quad , \quad (59)$$

where χ is proportional to the impurity density, K^1 is a constant dependent on the impurity and the host, $\langle a^2 \rangle = \langle a_p^2 \rangle_{av.}$ is the mean-squared anisotropy, and I_c is a function obtained by numerical integration. The first term in Equation 59 is the valence effect, while the second term is the anisotropy effect. In the experimental regions of interest, MK showed that I_c could be approximated by

$$I_c \approx 0.78 \chi \ln \chi - 0.36 \chi \quad . \quad (60)$$

Both χ and the residual resistivity ratio, ρ_r , defined by

$$\rho_r = \frac{R_{4.2 \text{ K}}}{R_{273 \text{ K}} - R_{4.2 \text{ K}}} \quad , \quad (61)$$

where $R_{4.2\text{ K}}$ and $R_{273\text{ K}}$ are the resistances of the alloy at 4.2 K and 273 K, respectively, are proportional to the impurity concentration. Hence, MK write

$$\chi = \alpha^i \rho_r \quad , \quad (62)$$

where α^i is a constant of proportionality. Equation 59 can then be written in the form

$$\delta T_c = A \rho_r + B \rho_r \ln \rho_r \quad , \quad (63)$$

where A and B are arbitrary constants.

Clem (40) extended the MK model to study the changes in the critical field curves of weak-coupling superconductors due to the presence of anisotropy in the energy gap. The second paper given in reference 40 (hereafter referred to as Clem II) is of particular interest here, because Clem II describes the detailed shape of the critical field curve, $H_c(T)$, in terms of the reduction of the effect due to anisotropy when simple nonmagnetic impurities are added to a pure superconductor. Clem II shows that the reduced critical field, $h(t) = H_c(t)/H_0$, and the term $\tilde{H}_c$, defined by

$$\tilde{H}_c^2 \equiv H_c^2 / 8\pi\gamma T_c^2 \quad , \quad (64)$$

should be nearly free from valence effects and, therefore, are the relevant quantities when considering changes due to anisotropy. The results are formulated, then, in terms of

h , $\tilde{H}_c$, t , and two additional parameters, $\lambda = X/2\pi$ and the function $X_H(t, \lambda)$ used to describe the effectiveness of anisotropy corrections to $\tilde{H}_c^2$.

Two different methods have been used to evaluate $\langle a^2 \rangle$ for pure Th. Anderson et al. (131) found that $\langle a^2 \rangle = 0.021$ by measuring the depression of T_c of pure Th as carbon impurities were added. Gubser (132), on the other hand, obtained a value of $\langle a^2 \rangle = 0.019$ using the slope of the critical field curve at T_c of the pure Th sample of DF. A value of $\langle a^2 \rangle = 0.02$ will be used here.

Figure 30 shows the deviation function obtained by DF for pure Th compared to the simple BCS theory (solid curve), and to the Clem theory (dot-dashed curve) for a pure superconductor with $\langle a^2 \rangle = 0.02$. On this figure it can be seen that the $D(t)$ data lie closer to the BCS theory curve than to the Clem theory curve, but this is a bit misleading because, as noted above, the H_0 obtained by DF for pure Th was calculated from the simple BCS theory which neglects anisotropy corrections. A more accurate calculation of H_0 should include anisotropy corrections in the extrapolation. Similarly, the γ value obtained by DF also needs to include anisotropy corrections.

In view of the above arguments, we have reanalyzed the DF pure Th data. The value of λ for the DF pure Th sample is essentially zero, so the low temperature data of DF were

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Figure 30. Deviation function for pure thorium from Decker and Finnemore (72). The BCS theory result is indicated by the solid curve. The dot-dashed curve is the Clem theory result for a pure superconductor with $\langle a^2 \rangle = 0.02$.

fitted to the Clem II relation

$$h(t, \lambda) = h^0(t) \{1 - \langle a^2 \rangle [X_H(t, \lambda) - X_H(0, \lambda)]\} \quad , \quad (65)$$

with $\lambda = 0$. The $h^0(t)$ in Equation 65 are the simple BCS critical field values and may be obtained from Swenson's tables (128). $\langle a^2 \rangle$ is taken to be 0.02 and $X_H(0, 0) = 1$. Using the Clem II notation, $X_H(t, 0)$ is given by

$$X_H(t, 0) = \left[\frac{\epsilon_0(t)}{\epsilon_0(0)} \frac{H_0}{H_c(t)} \right]^2 \frac{\Lambda(0)}{\Lambda(t)} \quad , \quad (66)$$

where $\epsilon_0(t)$ and $\Lambda(t)$ are the energy gap and penetration depth, respectively, of a simple BCS superconductor. Values of $\epsilon_0(t)/\epsilon_0(0)$ and $\Lambda(0)/\Lambda(t)$ may be obtained from Mühlischelegel's tables (35). The value of H_0 calculated by this procedure is 159.73 Oe, which is about 0.5 Oe larger than the value obtained by DF. This anisotropy corrected H_0 was then used to calculate $D(t)$. The results of this calculation are shown in Figure 31, where the x's indicate the anisotropy corrected $D(t)$ values. The anisotropy corrected data now lie closer to the Clem theory curve (dot-dashed curve) than to the simple BCS theory curve (solid curve).

An anisotropy corrected γ for pure Th may now be obtained from the Clem II relation

$$\tilde{H}_0^2(\lambda) = \tilde{H}_0^2 [1 - 2\langle a^2 \rangle X_H(0, \lambda)] \quad , \quad (67)$$

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Figure 31. Deviation function for pure thorium using both the simple BCS theory H_0 (open circles) and the anisotropy corrected $H_0(x's)$. The BCS theory result is indicated by the solid curve. The dot-dashed curve is the Clem theory result for a pure superconductor with $\langle a^2 \rangle = 0.02$.

where the superscript "0" indicates the BCS value of $\tilde{H}_0^2$.

Using Equation 54, we may write Equation 67 as

$$\gamma = \frac{0.168 H_0^2 V_M}{T_c^2 [1 - 2\langle a^2 \rangle X_H(0, \lambda)]} \quad (68)$$

Substitution of $H_0 = 159.73$ Oe, $V_M = 19.7$ cm³/mole, $T_c = 1.390$ K, $\langle a^2 \rangle = 0.02$, and $X_H(0,0) = 1$ into Equation 68 gives $\gamma = 4.55$ mJ/mole-K², a value about 5% larger than the 4.34 mJ/mole-K² obtained by DF.

The study of anisotropy corrections to H_0 , $D(t)$, and γ for the ThU alloys was limited to four samples: Th-0.030, -0.059, -0.102, and -0.127 at.% U. ρ_r values, however, were obtained for all the alloys and are listed in Table 3 along with the measured values of $\delta T_c = T_c - T_{cp}$, where T_{cp} is taken to be the DF value of 1.390 K. These data are presented in Figures 32 and 33. Also included in Figure 33 are the results of Anderson et al. (131), indicated by the dashed curve, for the depression of T_c of pure thorium by carbon impurities, which have a small valence effect. It should be noted that the scattering cross section of a uranium impurity is only about 10% the cross section of a carbon impurity (133).

In the calculation of anisotropy corrections for the four ThU alloys, the value of $\alpha^i = 100$ obtained by Anderson (133) was used to determine values of χ and hence λ . The

Table 3. Residual resistivity ratios and δT_c values for ThU

Sample	ρ_r	δT_c (K)
Th-0.000 at.% U	0.034	-0.030
Th-0.030 at.% U	0.044	-0.208
Th-0.052 at.% U	0.053	-0.377
Th-0.059 at.% U	0.055	-0.376
Th-0.077 at.% U	0.057	-0.527
Th-0.102 at.% U	0.063	-0.665
Th-0.127 at.% U	0.068	-0.732
Th-0.150 at.% U	0.076	-0.896
Th-0.161 at.% U	0.077	-0.903
Th-0.215 at.% U	0.102	-1.086

value of λ for Th-0.102 at.% U was 1.003, so $X_H(\lambda, t)$ values were obtained from the $X_H(1, t)$ plot given in Figure 1 of Clem II. The values of λ obtained for Th-0.030, -0.059, and 0.127 at.% U were 0.700, 0.875, and 1.082, respectively, and $X_H(t, \lambda)$ functions had to be derived for each of these samples. For $\lambda \neq 0$, $X_H(t, \lambda)$ is defined in terms of another function $\tilde{X}_H(t, \lambda)$, where

$$X_H(t, \lambda) = \left[\frac{\epsilon_0(t)}{\epsilon_0(0)} \frac{H_c(0)}{H_c(t)} \right]^2 \tilde{X}_H(t, \lambda) \quad (69)$$

Clem II shows that the function $\tilde{X}_H(t, \lambda)$ can be expanded

Figure 32. Residual resistivity ratio, ρ_r , as a function of uranium concentration for ThU.

Figure 33. Depression of T_c for ThU as a function of ρ_r . The pure Th sample of Decker and Finnemore (72), indicated by the solid triangle, is taken as the reference T_{cp} value. The dashed curve, from the work of Anderson et al. (131), indicates the depression of T_c which would be obtained for a simple nonmagnetic impurity with a small valence effect.

about the critical temperature in terms of $(1 - t)$,

$$\tilde{X}_H(t, \lambda) = B_1(\lambda)(1 - t) + B_2(\lambda)(1 - t)^2 + \dots \quad (70)$$

where $B_1(\lambda)$ and $B_2(\lambda)$ are given by Equations 34 and 35 in Clem II. The zero-temperature result is given by

$$\tilde{X}_H(0, \lambda) = \left\{ \begin{array}{ll} \pi/2y - (1 - y^2)^{1/2} \cos^{-1} y/y - \chi(\lambda) & , y \leq 1 \\ \pi/2y + (y^2 - 1)^{1/2} \cosh^{-1} y/y - \chi(\lambda) & , y \geq 1 \end{array} \right\}, \quad (71)$$

where $y = \gamma_e \lambda$ and $\chi(\lambda)$ is defined by

$$\chi(\lambda) \equiv \psi\left(\frac{1 + \lambda}{2}\right) - \psi(1/2) \quad (72)$$

Clem II shows that Equations 70 and 71 are sufficient to give a fairly good approximation to $\tilde{X}_H(t, \lambda)$ versus t^2 as a function of λ . We have calculated $\tilde{X}_H(t, \lambda)$ for $\lambda = 0.700$, 0.875 , and 1.082 and the results are shown in Figures 34-36. The horizontal dashed line in each figure is the $\tilde{X}_H(0, \lambda)$ value obtained from Equation 71, and the dashed curve is the result of the power series expansion given by Equation 70. The solid curves are interpolated by eye between the two dashed curves and the accuracy of this method should be approximately 2%, as estimated in Clem II. Using values of $\tilde{X}_H(t, \lambda)$ and Equation 69, the function $X_H(t, \lambda)$ was calculated and the

Figure 34. $\tilde{X}_H(t, \lambda)$ versus t^2 for Th-0.030 at.% U with $\lambda = 0.700$. The horizontal dashed line is obtained from Equation 71, and the dashed curve is obtained from Equation 70. The solid curve is the result of interpolating by eye between the dashed curves.

Figure 35. $\tilde{X}_H(t, \lambda)$ versus t^2 for Th-0.059 at.% U with $\lambda = 0.875$. The horizontal dashed line is obtained from Equation 71, and the dashed curve is obtained from Equation 70. The solid curve is the result of interpolating by eye between the dashed curves.

Figure 36. $\tilde{X}_H(t, \lambda)$ versus t^2 for Th-0.127 at.% U with $\lambda = 1.082$. The horizontal dashed line is obtained from Equation 71, and the dashed curve is obtained from Equation 70. The solid curve is the result of interpolating by eye between the dashed curves.

results are shown in Figures 37-39.

An anisotropy corrected H_0 was obtained for each alloy using the $X_H(t, \lambda)$ versus t plots and Equation 65. The results of the calculations are given in Appendix C. The anisotropy corrected H_0 values were then used to calculate the deviation functions for the alloys, which are also listed in Appendix C and shown in Figures 40-43. The critical field curve data for these ThU alloys deviate a maximum of about 0.6% from the BCS theory. The anisotropy corrected H_0 values and Equation 68 were used to calculate γ values for the alloys. The results of these calculations are listed in Table 4.

Table 4. Calculated anisotropy corrected values of pure Th and ThU

Sample	λ	H_0 (Oe)	γ (mJ/mole-K ²)
Pure Th(DF)	~0	159.73	4.55
Th-0.030 at.% U	0.700	137.16 $\pm$ 1.46	4.57 $\pm$ 0.09
Th-0.059 at.% U	0.875	117.39 $\pm$ 0.57	4.54 $\pm$ 0.04
Th-0.102 at.% U	1.003	84.97 $\pm$ 0.21	4.64 $\pm$ 0.03
Th-0.127 at.% U	1.082	76.97 $\pm$ 0.41	4.62 $\pm$ 0.05

Figure 37. $X_H(t, \lambda)$, which describes the effectiveness of anisotropy corrections to $H_C^2/8\pi\gamma T_C^2$, versus t for Th-0.030 at.% U with $\lambda = 0.700$.

Figure 38. $X_H(t, \lambda)$, which describes the effectiveness of anisotropy corrections to $H_C^2/8\pi\gamma T_C^2$, versus t for Th-0.059 at.% U with $\lambda = 0.875$.

Figure 39. $X_H(t, \lambda)$, which describes the effectiveness of anisotropy corrections to $H_C^2/8\pi\gamma T_C^2$, versus t for Th-0.127 at.% U with $\lambda = 1.082$.

Figure 40. Deviation function for Th-0.030 at.% U using both the simple BCS theory H_0 (solid circles) and the anisotropy corrected H_0 (x's) compared to the BCS theory (solid curve).

Figure 41. Deviation function for Th-0.059 at.% U using both the simple BCS theory H_0 (solid circles) and the anisotropy corrected H_0 (x's) compared to the BCS theory (solid curve).

Figure 42. Deviation function for Th-0.102 at.% U using both the simple BCS theory H_0 (solid circles) and the anisotropy corrected H_0 (x's) compared to the BCS theory (solid curve).

Figure 43. Deviation function for Th-0.127 at.% U using both the simple BCS theory H_0 (solid circles) and the anisotropy corrected H_0 (x's) compared to the BCS theory (solid curve).

An examination of the anisotropy corrected γ values listed in Table 4 indicates that within the scatter of the data, the value of γ remains essentially constant. The $H_c(T)$ measurements need to be extended to lower temperatures than were reached in this experiment to obtain experimental values for H_0 and γ using Equation 17 before any definite statement can be made concerning a γ enhancement. Alternatively, a specific heat study of these samples would be useful because, as discussed above, the ThU alloys used by Luengo et al. (113) are of questionable homogeneity.

IV. SUMMARY

This research shows that the critical field curves for ThU are in agreement with the BCS theory critical field curves to an accuracy of 1%, and thus provides additional experimental evidence that the rapid depression of the superconducting transition temperature of pure thorium by the uranium impurities is due to pair-weakening rather than to pair-breaking mechanisms. The effects of spin-flip scattering, if present at all, are at least an order of magnitude smaller than the pair-weakening effects. Additional experimental evidence is provided by the fact that the reduced specific heat jumps at T_c for these alloys, obtained from the slope of the critical field at T_c and Rutger's formula (Equation 15), are in close agreement with the predictions of the BCS theory.

Using a BCS model for ThU, one would expect, from Equation 19, that the reduction in T_c would enter through $N(0)V'$. However, within the accuracy of the data, the specific heat coefficient, γ , and hence, $N(0)$, remains essentially constant. This implies that the major effect is due to a reduction in the BCS effective electron-electron interaction, V' . The Kaiser model (96) would attribute the reduction of V' to resonance scattering at the uranium sites, which increases the effective Coulomb repulsion between the mates

of the Cooper pairs. The Riblet model (100), on the other hand, would say that the Cooper pairing interaction is reduced because electron-localized spin fluctuation coupling occurs. Other complex interactions may play a role. Although no one specific mechanism can be identified as being responsible for the destruction of superconductivity in ThU, this research strongly indicates that the primary interactions are time-reversal invariant, and hence, pair-weakening rather than pair-breaking.

V. BIBLIOGRAPHY

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VII. APPENDIX A: CRITICAL FIELD DATA

Table 5. Critical field curve data for Th - 0.030 at.% U

T(K)	H _c (Oersted)	Hysteresis (Oersted)
0.3013	127.600	3.899
0.3095	125.871	2.841
0.3198	125.571	3.566
0.3293	125.183	3.481
0.3392	124.416	3.408
0.3504	123.488	2.864
0.3598	122.752	3.769
0.3700	122.214	3.713
0.3795	120.980	2.830
0.3896	120.324	3.407
0.4013	119.546	3.153
0.4254	117.301	3.113
0.4507	114.743	1.608
0.4746	112.128	2.162
0.5003	109.307	2.349
0.5238	106.717	1.868
0.5502	103.678	1.172
0.5751	100.588	1.234
0.6000	97.540	1.726
0.6249	94.076	1.104
0.6499	90.852	1.289
0.6995	83.974	0.747
0.7498	76.395	1.046
0.7988	68.593	0.809
0.8495	60.700	0.734
0.9000	52.215	0.367
0.9508	43.485	0.500
0.9996	34.664	0.182
1.0518	25.047	0.249

Table 5 (Continued)

T(K)	H _C (Oersted)	Hysteresis (Oersted)
1.0983	16.505	0.495
1.1200	12.132	0.122
1.1292	10.387	0.116
1.1377	8.756	0.058
1.1497	6.574	0.062

Table 6. Critical field curve data for Th - 0.052 at.% U

T(K)	H _C (Oersted)	Hysteresis (Oersted)
0.3013	105.897	5.139
0.3503	101.850	4.833
0.4011	97.752	5.886
0.4507	91.925	6.118
0.5003	86.404	4.268
0.5503	79.610	4.278
0.6001	72.862	4.039
0.6499	65.001	2.407
0.6995	57.194	2.169
0.7497	48.692	1.479
0.7995	40.208	0.736
0.8496	31.450	0.437
0.9000	22.128	0.493
0.9507	12.700	0.191
0.9791	6.960	0.122

Table 7. Critical field curve data for Th - 0.059 at.% U

T(K)	H _c (Oersted)	Hysteresis (Oersted)
0.3028	105.775	1.183
0.3095	105.192	1.240
0.3198	104.391	1.064
0.3296	103.559	1.053
0.3392	102.690	0.934
0.3501	101.638	0.934
0.3598	100.888	0.928
0.3701	99.682	0.872
0.3799	98.754	0.872
0.3897	97.848	1.053
0.3994	96.940	0.990
0.4251	94.203	0.985
0.4503	90.870	0.860
0.4748	88.520	0.883
0.5003	85.251	0.737
0.5238	82.406	0.957
0.5502	78.769	0.560
0.6000	72.004	0.552
0.6504	64.518	0.432
0.6998	56.620	0.252
0.7499	48.483	0.306
0.7998	39.925	0.319
0.8511	31.047	0.181
0.8992	22.113	0.062
0.9493	12.712	0.129
0.9596	10.734	0.068
0.9698	8.874	0.058
0.9768	7.380	0.180
0.9788	7.246	0.125

Table 8. Critical field curve data for Th - 0.077 at.% U

T(K)	H _c (Oersted)	Hysteresis (Oersted)
0.3014	87.983	6.244
0.3503	83.257	6.679
0.4008	77.442	5.631
0.4509	70.921	4.828
0.5002	63.954	4.138
0.5501	56.493	3.965
0.6001	48.404	2.288
0.6500	40.171	1.800
0.6996	31.543	0.623
0.7497	21.955	0.487
0.7997	12.635	0.321
0.8094	10.728	0.189
0.8193	8.717	0.127
0.8298	6.759	0.185

Table 9. Critical field curve data for Th - 0.102 at.% U

T(K)	H _c (Oersted)	Hysteresis (Oersted)
0.3013	68.739	0.305
0.3252	65.518	0.252
0.3510	62.544	0.431
0.3745	59.472	0.125
0.4004	56.236	0.441
0.4267	52.289	0.119
0.4519	48.643	0.371
0.4764	44.872	0.278
0.5005	41.107	0.123
0.5251	37.017	0.247
0.5500	32.929	0.128
0.5751	28.505	0.178
0.6009	23.955	0.121
0.6247	19.646	0.068
0.6506	14.752	0.154
0.6694	11.254	0.122
0.6885	7.237	0.0

Table 10. Critical field curve data for Th - 0.127 at.% U

T(K)	H _c (Oersted)	Hysteresis (Oersted)
0.3011	58.804	0.367
0.3252	55.918	0.565
0.3511	52.556	0.863
0.3745	49.257	0.378
0.4003	45.470	0.306
0.4253	41.778	0.387
0.4524	37.348	0.170
0.4762	33.409	0.242
0.5007	29.465	0.252
0.5251	25.311	0.127
0.5500	20.749	0.255
0.5752	16.254	0.185
0.6006	11.414	0.186
0.6204	7.524	0.059

Table 11. Critical field curve data for Th - 0.150 at.% U

T(K)	H _c (Oersted)	Hysteresis (Oersted)
0.3016	35.094	0.259
0.3251	31.066	0.241
0.3508	26.954	0.184
0.3746	22.983	0.180
0.4003	18.634	0.133
0.4262	13.821	0.061
0.4503	8.875	0.057
0.4599	7.109	0.127

VIII. APPENDIX B: H_0 AND $D(t)$ FROM BCS THEORY

Table 12. Calculation of H_0 for Th - 0.030 at.% U

H_c (Oersted)	t^2	$h(\text{BCS})$	H_0 (Oersted)
127.600±1.950	0.0650	0.92905	137.345±2.099
125.871±1.421	0.0686	0.92502	136.074±1.536
125.571±1.783	0.0732	0.91987	136.510±1.938
125.183±1.741	0.0777	0.91482	136.839±1.903
124.416±1.704	0.0824	0.90954	136.790±1.873
123.488±1.432	0.0879	0.90336	136.699±1.585
122.752±1.885	0.0927	0.89796	136.701±2.066
122.214±1.857	0.0980	0.89199	137.013±2.082
120.980±1.415	0.1031	0.88625	136.508±1.597
120.324±1.704	0.1087	0.87993	136.743±1.937
119.546±1.577	0.1153	0.87251	137.014±1.807
117.301±1.557	0.1296	0.85641	136.968±1.818
114.743±0.804	0.1455	0.83857	136.832±0.959
112.128±1.081	0.1613	0.82088	136.595±1.317
109.307±1.175	0.1792	0.80092	136.477±1.467
106.717±0.934	0.1965	0.78175	136.510±1.195
103.678±0.586	0.2168	0.75937	136.532±0.772
100.588±0.617	0.2368	0.73746	136.398±0.837
97.540±0.863	0.2578	0.71462	136.492±1.208
94.076±0.552	0.2796	0.69108	136.129±0.799
90.852±0.645	0.3025	0.66658	136.296±0.968
83.974±0.374	0.3504	0.61594	136.335±0.607
76.395±0.523	0.4026	0.56176	135.992±0.931

Table 13. Calculation of H_0 for Th - 0.052 at.% U

H_c (Oersted)	t^2	$h(\text{BCS})$	H_0 (Oersted)
105.897±2.570	0.0885	0.90268	117.314±2.847
101.850±2.417	0.1196	0.86766	117.385±2.786
97.752±2.943	0.1568	0.82592	118.355±3.563
91.925±3.059	0.1980	0.78008	117.840±3.921
86.404±2.134	0.2439	0.72972	118.407±2.924
79.610±2.139	0.2951	0.67448	118.032±3.171
72.986±2.020	0.3509	0.61542	118.394±3.282

Table 14. Calculation of H_0 for Th - 0.059 at.% U

H_c (Oersted)	t^2	$h(\text{BCS})$	H_0 (Oersted)
105.775±0.592	0.0891	0.90200	117.267±0.656
105.192±0.620	0.0931	0.89751	117.204±0.691
104.391±0.532	0.0994	0.89040	117.241±0.597
103.559±0.527	0.1056	0.88343	117.224±0.596
102.690±0.467	0.1119	0.87633	117.182±0.533
101.638±0.467	0.1192	0.86811	117.080±0.538
100.888±0.464	0.1259	0.86058	117.233±0.539
99.682±0.436	0.1332	0.85238	116.945±0.512
98.754±0.436	0.1403	0.84438	116.954±0.516
97.848±0.527	0.1476	0.83621	117.014±0.630
96.940±0.495	0.1551	0.82782	117.103±0.598
94.203±0.493	0.1757	0.80483	117.048±0.613
90.870±0.430	0.1971	0.78108	116.339±0.551
88.520±0.442	0.2192	0.75672	116.979±0.584
85.251±0.369	0.2433	0.73037	116.723±0.505
82.406±0.479	0.2667	0.70500	116.888±0.679
78.769±0.280	0.2943	0.67534	116.636±0.415
72.004±0.276	0.3500	0.61635	116.823±0.448

Table 15. Calculation of H_0 for Th - 0.077 at.% U

H_c (Oersted)	t^2	$h(\text{BCS})$	H_0 (Oersted)
87.983±3.122	0.1221	0.86485	101.732±3.610
83.257±3.340	0.1649	0.81687	101.922±4.089
77.442±2.816	0.2158	0.76047	101.834±3.703
70.921±2.414	0.2732	0.69799	101.607±3.459
63.954±2.069	0.3362	0.63087	101.374±3.280
56.493±1.983	0.4066	0.55765	101.305±3.556

Table 16. Calculation of H_0 for Th - 0.102 at.% U

H_c (Oersted)	t^2	$h(\text{BCS})$	H_0 (Oersted)
68.739±0.153	0.1725	0.80839	85.032±0.189
65.518±0.126	0.2026	0.77500	84.526±0.163
62.544±0.216	0.2341	0.74041	84.472±0.292
59.479±0.063	0.2665	0.70521	84.342±0.089
56.236±0.221	0.3047	0.66424	84.662±0.333
52.289±0.060	0.3460	0.62057	84.260±0.097
48.643±0.186	0.3880	0.57682	84.330±0.322

Table 17. Calculation of H_0 for Th - 0.127 at.% U

H_c (Oersted)	t^2	$h(\text{BCS})$	H_0 (Oersted)
58.804 $\pm$ 0.184	0.2093	0.76760	76.608 $\pm$ 0.240
55.918 $\pm$ 0.283	0.2441	0.72950	76.653 $\pm$ 0.388
52.556 $\pm$ 0.432	0.2845	0.68584	76.630 $\pm$ 0.630
49.257 $\pm$ 0.189	0.3237	0.64406	76.479 $\pm$ 0.293
45.470 $\pm$ 0.303	0.3699	0.59556	76.348 $\pm$ 0.509

Table 18. $D(t)$ for Th - 0.030 at.% U

t^2	h	$D(t)$
0.0650	0.9341	-0.0009
0.0686	0.9215	-0.0099
0.0732	0.9193	-0.0075
0.0777	0.9164	-0.0059
0.0824	0.9108	-0.0068
0.0879	0.9040	-0.0081
0.0927	0.8986	-0.0087
0.0980	0.8947	-0.0073
0.1031	0.8857	-0.0112
0.1087	0.8808	-0.0105
0.1153	0.8752	-0.0095
0.1296	0.8587	-0.0117
0.1455	0.8400	-0.0145
0.1613	0.8208	-0.0179
0.1792	0.8002	-0.0206
0.1965	0.8035	-0.0223
0.2168	0.7590	-0.0242
0.2368	0.7364	-0.0268
0.2578	0.7141	-0.0281
0.2796	0.6887	-0.0317
0.3025	0.6651	-0.0324
0.3504	0.6147	-0.0349
0.4026	0.5593	-0.0381
0.4581	0.5021	-0.0398
0.5168	0.4444	-0.0388
0.5801	0.3822	-0.0377
0.6474	0.3183	-0.0343
0.7155	0.2538	-0.0307
0.7922	0.1834	-0.0244

Table 18 (Continued)

t^2	h	$D(t)$
0.8638	0.1208	-0.0154
0.8983	0.0888	-0.0129
0.9131	0.0760	-0.0109
0.9269	0.0641	-0.0090
0.9466	0.0481	-0.0053

Table 19. $D(t)$ for Th - 0.052 at.% U

t^2	h	$D(t)$
0.0885	0.8977	-0.0138
0.1196	0.8634	-0.0170
0.1568	0.8287	-0.0145
0.1980	0.7793	-0.0227
0.2439	0.7325	-0.0236
0.2951	0.6749	-0.0300
0.3509	0.6177	-0.0314
0.4116	0.5510	-0.0374
0.4768	0.4849	-0.0383
0.5477	0.4128	-0.0395
0.6229	0.3409	-0.0362
0.7034	0.2666	-0.0300
0.7893	0.1876	-0.0231
0.8808	0.1077	-0.0115
0.9342	0.0590	-0.0068

Table 20. $D(t)$ for Th - 0.059 at.% U

t^2	h	$D(t)$
0.0891	0.9041	-0.0068
0.0931	0.8991	-0.0078
0.0994	0.8923	-0.0083
0.1056	0.8852	-0.0092
0.1119	0.8777	-0.0104
0.1192	0.8687	-0.0121
0.1259	0.8623	-0.0118
0.1332	0.8520	-0.0148
0.1403	0.8441	-0.0156
0.1476	0.8364	-0.0160
0.1551	0.8286	-0.0163
0.1757	0.8052	-0.0191
0.1971	0.7767	-0.0262
0.2192	0.7566	-0.0242
0.2433	0.7287	-0.0280
0.2667	0.7044	-0.0289
0.2943	0.6732	-0.0325
0.3500	0.6155	-0.0345
0.4113	0.5515	-0.0372
0.4761	0.4840	-0.0399
0.5467	0.4144	-0.0389
0.6219	0.3413	-0.0368
0.7042	0.2654	-0.0304
0.7861	0.1890	-0.0249
0.8761	0.1087	-0.0152
0.8952	0.0917	-0.0131
0.9144	0.0759	-0.0097
0.9276	0.0631	-0.0093
0.9314	0.0619	-0.0067

Table 21. $D(t)$ for Th - 0.077 at.% U

t^2	h	$D(t)$
0.1221	0.8657	-0.0122
0.1649	0.8192	-0.0159
0.2158	0.7620	-0.0222
0.2732	0.6978	-0.0290
0.3362	0.6293	-0.0345
0.4066	0.5559	-0.0375
0.4839	0.4763	-0.0398
0.5677	0.3953	-0.0370
0.6576	0.3104	-0.0320
0.7552	0.2160	-0.0288
0.8593	0.1243	-0.0164
0.8803	0.1056	-0.0141
0.9019	0.0858	-0.0123
0.9252	0.0665	-0.0083

Table 22. $D(t)$ for Th - 0.102 at.% U

t^2	h	$D(t)$
0.1725	0.8133	-0.0142
0.2026	0.7752	-0.0222
0.2341	0.7400	-0.0259
0.2665	0.7037	-0.0298
0.3047	0.6654	-0.0299
0.3460	0.6187	-0.0353
0.3880	0.5755	-0.0365
0.4313	0.5309	-0.0378
0.4761	0.4864	-0.0375
0.5240	0.4380	-0.0380
0.5749	0.3896	-0.0355
0.6285	0.3373	-0.0345
0.6862	0.2834	-0.0304
0.7416	0.2324	-0.0260
0.8044	0.1745	-0.0211
0.8516	0.1332	-0.0152
0.9009	0.0856	-0.0135

Table 23. $D(t)$ for Th - 0.127 at.% U

t^2	h	$D(t)$
0.2093	0.7682	-0.0225
0.2441	0.7305	-0.0254
0.2845	0.6866	-0.0289
0.3237	0.6435	-0.0328
0.3699	0.5940	-0.0361
0.4175	0.5458	-0.0367
0.4724	0.4879	-0.0397
0.5234	0.4365	-0.0401
0.5787	0.3849	-0.0364
0.6365	0.3307	-0.0328
0.6982	0.2711	-0.0307
0.7637	0.2123	-0.0240
0.8326	0.1491	-0.0183
0.8884	0.0983	-0.0133

IX. APPENDIX C: H_0 AND $D(t)$ FROM THE ANISOTROPY
CORRECTED BCS THEORY OF CLEM

Table 24. Calculation of anisotropy corrected H_0 for
Th - 0.030 at.% U

H_c (Oersted)	t^2	$h(\text{BCS-Clem})$	H_0 (Oersted)
127.600±1.950	0.0650	0.9275	137.574±2.102
125.871±1.421	0.0686	0.9234	136.313±1.539
125.571±1.783	0.0732	0.9181	136.773±1.942
125.183±1.741	0.0777	0.9129	137.127±1.907
124.416±1.704	0.0824	0.9075	137.098±1.878
123.488±1.432	0.0879	0.9013	137.011±1.589
122.752±1.885	0.0927	0.8958	137.031±2.104
122.214±1.857	0.0980	0.8898	137.350±2.087
120.980±1.415	0.1031	0.8839	136.871±1.601
120.324±1.704	0.1087	0.8775	137.121±1.942
119.546±1.577	0.1153	0.8699	137.425±1.813
117.301±1.557	0.1296	0.8536	137.419±1.824
114.743±0.804	0.1455	0.8355	137.335±0.962
112.128±1.081	0.1613	0.8174	137.176±1.322
109.307±1.175	0.1792	0.7973	137.096±1.474
106.717±0.934	0.1965	0.7779	137.186±1.201
103.678±0.586	0.2168	0.7553	137.267±0.776
100.588±0.617	0.2368	0.7332	137.190±0.842
97.540±0.863	0.2578	0.7101	137.361±1.215
94.076±0.552	0.2796	0.6864	137.057±0.804
90.852±0.645	0.3025	0.6618	137.280±0.975
83.974±0.374	0.3504	0.6111	137.414±0.612
76.395±0.523	0.4026	0.5568	137.204±0.939

Table 25. Calculation of anisotropy corrected H_0 for
Th - 0.059 at.% U

H_c (Oersted)	t^2	$h(\text{BCS-Clem})$	H_0 (Oersted)
105.775±0.592	0.0891	0.9003	117.489±0.658
105.192±0.620	0.0931	0.8958	117.428±0.692
104.391±0.532	0.0994	0.8885	117.491±0.599
103.559±0.527	0.1056	0.8815	117.480±0.598
102.690±0.467	0.1119	0.8743	117.454±0.534
101.638±0.467	0.1192	0.8660	117.365±0.539
100.888±0.464	0.1259	0.8584	117.530±0.541
99.682±0.436	0.1332	0.8501	117.259±0.513
98.754±0.436	0.1403	0.8420	117.285±0.518
97.848±0.527	0.1476	0.8337	117.366±0.632
96.940±0.495	0.1551	0.8251	117.489±0.600
94.203±0.493	0.1757	0.8020	117.460±0.615
90.870±0.430	0.1971	0.7780	116.799±0.553
88.520±0.442	0.2192	0.7535	117.478±0.587
85.251±0.369	0.2433	0.7270	117.264±0.508
82.406±0.479	0.2667	0.7013	117.505±0.683
78.769±0.280	0.2943	0.6714	117.321±0.417
72.004±0.276	0.3500	0.6122	117.615±0.451

Table 26. Calculation of anisotropy corrected H_0 for
Th - 0.102 at.% U

H_c (Oersted)	t^2	$h(\text{BCS-Clem})$	H_0 (Oersted)
68.739±0.153	0.1725	0.8055	85.337±0.190
65.518±0.126	0.2026	0.7717	84.888±0.163
62.544±0.216	0.2341	0.7369	84.874±0.293
59.479±0.063	0.2665	0.7014	84.800±0.090
56.236±0.221	0.3047	0.6603	85.167±0.335
52.289±0.060	0.3460	0.6165	84.816±0.097
48.643±0.186	0.3880	0.5727	84.936±0.325

Table 27. Calculation of anisotropy corrected H_0 for
Th - 0.127 at.% U

H_c (Oersted)	t^2	$H(\text{BCS-Clem})$	H_0 (Oersted)
58.804±0.184	0.2093	0.7642	76.948±0.241
55.918±0.283	0.2441	0.7259	77.033±0.390
52.556±0.432	0.2845	0.6820	77.062±0.633
49.257±0.189	0.3237	0.6401	76.952±0.295
45.470±0.303	0.3699	0.5915	76.872±0.512

Table 28. Anisotropy corrected $D(t)$ for Th - 0.030 at.% U

t^2	h	$D(t)$
0.0650	0.9303	-0.0047
0.0686	0.9177	-0.0137
0.0732	0.9155	-0.0113
0.0777	0.9127	-0.0096
0.0824	0.9071	-0.0105
0.0879	0.9003	-0.0118
0.0927	0.8950	-0.0123
0.0980	0.8910	-0.0110
0.1031	0.8820	-0.0149
0.1087	0.8773	-0.0140
0.1153	0.8716	-0.0131
0.1296	0.8552	-0.0152
0.1455	0.8366	-0.0179
0.1613	0.8175	-0.0212
0.1792	0.7969	-0.0239
0.1965	0.7780	-0.0255
0.2168	0.7559	-0.0273
0.2368	0.7334	-0.0298
0.2578	0.7111	-0.0311
0.2796	0.6859	-0.0345
0.3025	0.6624	-0.0351
0.3504	0.6122	-0.0374
0.4026	0.5570	-0.0404
0.4581	0.5001	-0.0418
0.5168	0.4425	-0.0407
0.5801	0.3807	-0.0392
0.6474	0.3170	-0.0356
0.7155	0.2527	-0.0318
0.7922	0.1826	-0.0252

Table 28 (Continued)

t^2	h	$D(t)$
0.8638	0.1203	-0.0159
0.8983	0.0885	-0.0132
0.9131	0.0757	-0.0112
0.9269	0.0638	-0.0093
0.9466	0.0479	-0.0055

Table 29. Anisotropy corrected $D(t)$ for Th - 0.059 at.% U

t^2	h	$D(t)$
0.0891	0.9010	-0.0099
0.0931	0.8961	-0.0108
0.0994	0.8892	-0.0114
0.1056	0.8822	-0.0122
0.1119	0.8748	-0.0133
0.1192	0.8658	-0.0150
0.1259	0.8595	-0.0146
0.1332	0.8491	-0.0177
0.1403	0.8412	-0.0185
0.1476	0.8335	-0.0189
0.1551	0.8258	-0.0191
0.1757	0.8025	-0.0218
0.1971	0.7741	-0.0288
0.2192	0.7540	-0.0268
0.2433	0.7262	-0.0305
0.2667	0.7020	-0.0313
0.2943	0.6710	-0.0347
0.3500	0.6134	-0.0366
0.4113	0.5496	-0.0391

Table 29 (Continued)

t^2	h	$D(t)$
0.4761	0.4823	-0.0416
0.5467	0.4130	-0.0403
0.6219	0.3401	-0.0380
0.7042	0.2645	-0.0313
0.7861	0.1884	-0.0255
0.8761	0.1083	-0.0156
0.8952	0.0914	-0.0134
0.9144	0.0756	-0.0100
0.9276	0.0629	-0.0095
0.9314	0.0617	-0.0069

Table 30. Anisotropy corrected $D(t)$ for Th - 0.102 at.% U

t^2	h	$D(t)$
0.1725	0.8055	-0.0220
0.2026	0.7717	-0.0257
0.2341	0.7369	-0.0290
0.2665	0.7014	-0.0321
0.3047	0.6603	-0.0350
0.3460	0.6165	-0.0375
0.3880	0.5724	-0.0396
0.4313	0.5281	-0.0406
0.4761	0.4838	-0.0401
0.5240	0.4356	-0.0404
0.5749	0.3875	-0.0376
0.6285	0.3355	-0.0360
0.6862	0.2819	-0.0319

Table 30 (Continued)

t^2	h	$D(t)$
0.7416	0.2312	-0.0272
0.8044	0.1736	-0.0220
0.8516	0.1324	-0.0160
0.9009	0.0852	-0.0139

Table 31. Anisotropy corrected $D(t)$ for Th - 0.127 at.% U

t^2	h	$D(t)$
0.2093	0.7640	-0.0267
0.2441	0.7265	-0.0294
0.2845	0.6828	-0.0327
0.3237	0.6399	-0.0364
0.3699	0.5907	-0.0394
0.4175	0.5428	-0.0397
0.4724	0.4852	-0.0424
0.5234	0.4340	-0.0426
0.5787	0.3828	-0.0385
0.6365	0.3288	-0.0347
0.6982	0.2696	-0.0322
0.7637	0.2112	-0.0251
0.8326	0.1483	-0.0191
0.8884	0.0977	-0.0139