

# Thermodynamics Of S-Wave Pairing In Uranium And Cerium Based Heavy Fermion Compounds

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**Abstract**— Superconductivity with simple *s*-wave pairing symmetry is another intriguing phenomenon in heavy-fermion systems. Heavy-fermion superconductors favor the nodal pairing states, such as the *d*-wave and *p*-wave states, rather than the *s*-wave state. The *s*-wave Cooper pairing in Uranium and Cerium based HF systems has been studied by analyzing the periodic Anderson model by means of the Bogoliubov-Valatin approach (BVT) while focusing on the interorbital Cooper pairing between a conduction electron (*c* electron) and an *f* electron. The ground state energy ( $E_0$ ) specific heat ( $C_V$ ) and electronic specific heat coefficient ( $\gamma$ ) of this heavy superconductors have been determined in the framework of the *s*-wave model. The critical temperature for Uranium and Cerium based compounds is  $T_c=1.74\text{K}$  and  $T_c=0.24\text{K}$  respectively which are in agreement with known experimental values. Moreover, the BVT study shows that the interorbital *c*-*f* pairing is essential for the appearance of the *s*-wave superconductivity in Cerium based compounds.

**Keywords**—Heavy fermions, Specific heat, Transition temperature.

## I. INTRODUCTION

In usual heavy-fermion superconductors, the strong Coulomb repulsion between *f* electrons favors the nodal *d*-wave symmetry, which has been the subject of a number of theoretical studies [1]. The nuclear magnetic and quadrupole resonances (NMR and NQR) in typical heavy-fermion compounds show a power-law temperature dependence of the spin-lattice relaxation rate and the lack of the Hebel-Slichter peak [2] which indicates the existence of line nodes. Moreover, the phase diagram has the same feature as that of high- $T_c$  cuprates with *d*-wave symmetry; superconductivity appears near the antiferromagnetic phase [3]. However, conventional *s*-wave superconductivity has also been found in some compounds. In NQR measurements on  $\text{CeRu}_2$  and  $\text{CeCo}_2$ , the spin-lattice relaxation rate exhibits an exponential decay at low temperatures and shows the Hebel-Slichter peak. Moreover, the recent photoemission spectroscopy (PES) experiment on  $\text{CeRu}_2$  has shown that the density of states (DOS) has a clear superconducting gap at the Fermi level. All these results were interpreted as evidence for the fully gapped pairing state with *s*-wave symmetry. Usually, this type of

simple pairing symmetry can be understood within the framework of the conventional electron-phonon mechanism. However, it is unclear whether the electron-phonon attraction can be dominant since the Coulomb repulsion is rather strong in heavy-fermion systems [4]. The multi-orbital nature is one of the characteristic features of heavy-fermion systems, which are composed of itinerant electrons in the conduction orbitals (*c* electrons) and localized electrons in the *f* orbitals (*f* electrons) [5]. The correlation between *c* and *f* electrons leads to various intriguing phenomena, such as the Kondo effect [3] quantum critical behavior [6] and magnetic orderings due to the Ruderman Kittel-Kasuya-Yosida interaction [7]. Recently, the importance of such orbital degrees of freedom has also been recognized in the studies of superconductivity in the other strongly correlated electron systems. For example, the material dependence in the critical temperature of cuprates has been explained by using the multi-orbital Hubbard models [8]. Moreover, the multi-orbital nature is considered to be the key for understanding the high- $T_c$  superconducting properties in iron pnictides [9]. Previous studies [10] suggested that the multi-orbital nature can be a source of *s*-wave superconductivity in heavy-fermion systems. Hanzawa and Yosida and Spalek [11] discussed the interorbital Cooper pairing between *c* and *f* electrons, which we call the “*c*-*f* pairing,” as a possible mechanism for *s*-wave superconductivity. More recently, the present authors [10] also studied the *c*-*f* pairing for finite Coulomb repulsion, and presented a mean field phase diagram of the *s*-wave superconducting state. Note, however, that the mean-field approximation cannot properly describe local charge, spin, and orbital fluctuation effects, which are crucial in heavy-fermion systems. Thus more sophisticated treatment is required to achieve a deeper understanding of the nature of the interorbital pairing [5]. The advent of heavy-fermion (HF) superconductivity (SC) in  $\text{CeCu}_2\text{Si}_2$  followed the discoveries of super fluidity in  $^3\text{He}$  and HF phenomena in  $\text{CeAl}_3$  [12]. Given the antagonistic nature of SC and magnetism, the observation of SC in  $\text{CeCu}_2\text{Si}_2$  came as big surprise: All superconductors known at that time lose their SC when doped with a tiny amount ( $\sim 1\%$ ) of magnetic impurities – whereas in  $\text{CeCu}_2\text{Si}_2$  a dense, periodic lattice of (100 at%) magnetic  $\text{Ce}^{3+}$  ions is necessary to generate the superconducting state. This was inferred from the observation that the non-magnetic reference compound  $\text{LaCu}_2\text{Si}_2$  is not a superconductor and that SC in  $\text{CeCu}_2\text{Si}_2$

is fully suppressed by doping with a tiny amount of non-magnetic impurities [13]. For  $\text{CeCu}_2\text{Si}_2$ , the ( $T \rightarrow 0$ ) Sommerfeld coefficient  $\gamma \approx 1 \text{ J/K}^2\text{-mole}$  exceeds the  $\gamma$  value of a simple metal like Cu by about three orders of magnitude. From the observation that the jump  $\Delta C(T)/T$  at  $T_c \approx 0.6 \text{ K}$  is of the same gigantic order as the normal-state value of  $C/T$  at  $T_c$ , it was concluded that the heavy-mass charge carriers make up the Cooper pairs [12]. Similar conclusions were drawn for a few U-based HF superconductors discovered in the mid 1980ies, i.e.,  $\text{UBe}_{13}$ ,  $\text{UPt}_3$  and  $\text{URu}_2\text{Si}_2$  [13].

Superconductivity in HF metals involves pairing order parameters which are distinct from the BCS  $s$ -wave type. Strong support for this is lent by the existence of multiple superconducting phases, similar to what was observed for superfluid  $^3\text{He}$ . Multiphase superconductivity was found for  $\text{UPt}_3$ ,  $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$  and  $\text{PrOs}_4\text{Sb}_{12}$  [14]; the latter compound is unique here, since electric quadrupole fluctuations rather than magnetic dipole fluctuations, as commonly assumed for HF superconductors, are believed to mediate the Cooper pairing. Quite generally, the microscopic pairing mechanism in HF superconductors is driven by electronic interactions, contrasting SC in classical (BCS) superconductors mediated by electron-phonon coupling.

The coexistence of superconductivity and long range magnetic order was observed in several ferromagnets ( $\text{UGe}_2$ ,  $\text{URhGe}$  etc) as well as antiferromagnets ( $\text{UPd}_2\text{Al}_3$ ,  $\text{UNi}_2\text{Al}_3$  etc) [3].

On the other hand some heavy fermion compounds such as  $\text{CeRu}_2$ ,  $\text{CeCo}_2$  and  $\text{CeCu}_2\text{Si}_2$  are known to exhibit  $s$ -wave superconductivity [15].

The pairing mechanism in HFSC at finite temperature is not established. Extensive experimental and theoretical studies on  $\text{UPt}_3$ , a typical HF superconductor, have been devoted in order to elucidate the pairing symmetry realized in this particular material [16].  $\text{UPt}_3$  discovered in 1983 [17] is rather special in this class of materials because it is found in 1988 [18] that the superconducting transition temperature is split ( $T_{c1} = 0.58 \text{ K}$  and  $T_{c2} = 0.53 \text{ K}$ ) self evidently suggesting a rich internal structure of the Cooper pair, definitely not a simple  $s$ -wave pairing. From the experimental data it follows that the physics in the heavy-fermion systems is rather complex. The localized character of the  $f$ -electrons at high temperatures crosses over to a delocalized one at low temperatures. The high  $\gamma$ -values point to pronounced  $f$ -density of states, and, hence, to a strongly interacting electron liquid, assuring the presence of many-body effects. Historically, “on  $s$ -wave pairing” began with the publication “Generalized Bardeen-Cooper-Schrieffer States and the Proposed Low-Temperature Phase of  $^3\text{He}$ ” by Anderson and Morel. They considered the possibility of BCS pairing with non-zero angular momentum, and studied its physical consequences. When super fluidity was discovered in 1972 by Osheroff et al, 1972 [19] in  $^3\text{He}$  it was immediately clear that this was not a conventional  $s$ -wave BCS superfluid because there was more than one superfluid phase. Increasing evidence for non- $s$ -wave pairing came from many experimental results and within about a year after their discovery the three superfluid phases of  $^3\text{He}$  were undisputedly identified as  $p$ -wave (as proposed by Anderson and Morel) spin-triplet superfluids. The search for “non- $s$ -wave superconductivity”, the

metallic analog to super fluidity in  $^3\text{He}$ , was unsuccessful for more than a decade. In 1979 Steglich discovered superconductivity in  $\text{CeCu}_2\text{Si}_2$  [12]. This was the first in a new class of heavy-fermion superconductors, which now include the U-based compounds  $\text{UBe}_{13}$ ,  $\text{UPt}_3$ ,  $\text{URu}_2\text{Si}_2$ ,  $\text{UNi}_2\text{Al}_3$ , and  $\text{UPd}_2\text{Al}_3$ . Unusual temperature dependences of heat capacity, penetration depth, and sound absorption led to conjectures that these materials were non- $s$  wave superconductors.

## II. FORMALISM

### A. Model Hamiltonian for $s$ -wave superconductivity in heavy fermion systems

A typical HF system composed of itinerant  $c$  electrons and nearly localized  $f$  electrons, which hybridize with each other is considered. Usually such a system is modeled by the periodic Anderson Hamiltonian  $H_{\text{PAM}} = H_0 + H_V$ , [10]

$$H = H_{\text{dir}} + H_{\text{on}} + H_{c-f} + H_{\text{ee}} + H_{\text{rep}} \quad (1)$$

where  $H_{\text{dir}}$  represents the hopping electrons,  $H_{\text{on}}$  represents the on-site energy of  $f$  electrons,  $H_{c-f}$  represents the hybridization between the  $c$  and  $f$  states,  $H_{\text{ee}}$  represent the band occupation while  $H_{\text{rep}}$  represents the on-site coulomb repulsion in the  $f$  orbital.

$$H = \sum_k \varepsilon_k (c_k^\dagger c_k + c_{-k}^\dagger c_{-k}) + \sum_f \varepsilon_f n_f^\dagger - \sum_{k\sigma} t_k (c_{k\sigma}^\dagger f_{-k\sigma} - c_{-k\sigma}^\dagger f_{k\sigma} + H.c) - \mu \sum_{i\sigma} (n_{i\sigma}^c + n_{i\sigma}^f) + \sum_k U_k (n_{i\uparrow}^f n_{i\downarrow}^f) \quad (2)$$

where  $\varepsilon_k$  is the on-site energy of  $c$  electrons  $n_{i\sigma}^f = f_{i\sigma}^\dagger f_{k\sigma}$  and  $\varepsilon_f$  is the on-site energy of  $f$  electrons,  $c_{i\sigma}^\dagger$  ( $f_{i\sigma}^\dagger$ ) creates an itinerant  $c$  electron (a localized  $f$  electron) with spin  $\sigma$  at site  $i$  and  $t_k$  is the hybridization energy between the conduction and  $f$  electron states  $\mu$  is the chemical potential that determines the occupation of the band and  $U_k$  is the on-site Coulomb repulsion.

Equation (2) which is modified form of the PAM Hamiltonian is expressed in terms of creation and annihilation operators and then diagonalized to obtain the elements of the of the Hamiltonian that correspond to stationary states where the system is in equilibrium. Bogoliubov-Valatin Transformations are used to transform equation (2). This is achieved by defining two new operators related to the fermion creation and annihilation operators as follows

$$\gamma_k = u_k c_k - v_k c_{-k}^+ \quad (3a)$$

$$\gamma_{-k} = u_k c_{-k} + v_k c_k^+ \quad (3b)$$

$$\gamma_k^+ = u_k c_k^+ - v_k c_{-k} \quad (3c)$$

$$\gamma_{-k}^+ = u_k c_{-k}^+ + v_k c_k \quad (3d)$$

the inverse transformation of equations (3) are used.

$$c_k = u_k \gamma_k + v_k \gamma_{-k}^+ \quad (4a)$$

$$c_{-k} = u_k \gamma_{-k} - v_k \gamma_k^+ \quad (4b)$$

$$c_k^+ = u_k \gamma_k^+ + v_k \gamma_{-k} \quad (4c)$$

$$c_{-k}^+ = u_k \gamma_{-k}^+ - v_k \gamma_k \quad (4d)$$

The Hamiltonian for the  $s$ -wave superconductivity model from equation (2) is

$$\begin{aligned}
H = & \sum_k \varepsilon_k \left\{ 2v_k^2 + (u_k^2 - v_k^2)(m_k + m_{-k}) + 2u_k v_k (\gamma_k^+ \gamma_{-k}^+ + \gamma_{-k} \gamma_k) \right\} \\
& - \sum_k t_k (4u_k v_k (m_k + m_{-k}) - 1) + (2v_k^2 - 2u_k^2)(\gamma_{-k} \gamma_k + \gamma_k^+ \gamma_{-k}^+) \\
& + \sum_k \varepsilon_f \left\{ 2v_k^2 + (u_k^2 - v_k^2)(m_k + m_{-k}) + 2u_k v_k (\gamma_k^+ \gamma_{-k}^+ + \gamma_{-k} \gamma_k) \right\} \\
& - \mu \sum_k \left( 2u_k^2 m_k + 2v_k^2 (1 - m_{-k}) + 2u_k v_k (\gamma_k^+ \gamma_{-k}^+ + \gamma_{-k} \gamma_k) \right) \\
& + \sum_k U_k \left( v_k^4 (1 - m_k)(1 - m_{-k}) + 2v_k^3 u_k (\gamma_{-k} \gamma_k + \gamma_k^+ \gamma_{-k}^+) \right)
\end{aligned} \quad (5)$$

### B. Diagonalization of the s-wave wave pairing Hamiltonian

To diagonalize the Hamiltonian in equation (5), we put the sum of the off-diagonal terms equal to zero and simplify the resultant expression and determine the values of  $u_k$  and  $v_k$  to be used for the diagonalization. Thus

$$4u_k v_k + 2v_k^3 u_k + 2u_k^2 - 2v_k^2 = 0 \quad (6)$$

By solving equation (6) we obtain  $u_k = 0.41$  and

$v_k = 0.91$  by invoking the constraint (7) for fermions

$$v_k^2 + u_k^2 = 1 \quad (7)$$

The diagonalized Hamiltonian from equation (5) is

$$\begin{aligned}
H_{diag} = & \sum_k 2v_k^2 \varepsilon_k + \varepsilon_f \sum_k 2v_k^2 - \sum_k t_k (4u_k v_k (m_k + m_{-k}) - 1) \\
& - \mu \sum_k \left( 2v_k^2 (1 - m_{-k}) \right)
\end{aligned} \quad (8)$$

At the lowest energy state of this system, both  $m_k$  and  $m_{-k}$  are zero. Hence to carry out the Bogoliubov-Valatin transformation for a superconductor in its ground state,  $m_k$  and  $m_{-k}$  are set to zero in eq.(8)

$$H_{diag} = \sum_k 2\varepsilon_k v_k^2 + \varepsilon_f \sum_k 2v_k^2 - \mu \sum_k 2v_k^2 + \sum_k 4t_k u_k v_k + \sum_k U_k v_k^4 \quad (9)$$

### C. Superconducting energy for s-wave superconducting state.

Substituting for the values of  $u_k = 0.41$  and  $v_k = 0.91$  in equation (9) the magnitude of the ground state energy of the system is obtained as;

$$E_o = 1.66\varepsilon_k + 1.66\varepsilon_f - 1.66\mu + 0.37t_k + 0.69U_k \quad (10)$$

The energy of the system  $E$  at any temperature is found by multiplying the ground state energy  $E_o$  by the thermal

activation factor  $e^{\frac{-\Delta E}{K_B T}}$  [20], where  $K_B$  is Boltzmann's constant and  $-\Delta E$  is the energy gap. The energy gap for superconductors is a very small quantity and it is generally 1% of the minimum energy of the system [20]. Thus  $-\Delta E = 0.01E_o$  so at any temperature  $T$ , the energy of the system is given as

$$E = E_o e^{\frac{-E_o}{100K_B T}} \quad (11)$$

Substituting equation (10) in equation (11), we obtain the magnitude of energy of the system at any given temperature as;

$$E = (1.66\varepsilon_k + 1.66\varepsilon_f - 1.66\mu + 0.37t_k + 0.69U_k) e^{\frac{-(1.66\varepsilon_k + 1.66\varepsilon_f - 1.66\mu + 0.37t_k + 0.69U_k)}{100K_B T}} \quad (12)$$

### D. Specific heat capacity of s-wave superconducting state.

The specific heat capacity at constant volume  $C_V$  of the system is obtained by determining the first derivative of the energy of the system with respect to the temperature. Hence, using equation (12), we calculate the magnitude of  $C_V$  as follows

$$\begin{aligned}
C_V = & \frac{\partial E}{\partial T} = \frac{\partial}{\partial T} (1.66\varepsilon_k + 1.66\varepsilon_f - 1.66\mu + 0.37t_k + 0.69U_k) \\
& \times \left( e^{\frac{-(1.66\varepsilon_k + 1.66\varepsilon_f - 1.66\mu + 0.37t_k + 0.69U_k)}{100K_B T}} \right)
\end{aligned} \quad (13a)$$

Hence, according to this model the superconducting specific heat capacity at constant volume is given as;

$$\begin{aligned}
C_V = & \frac{-(1.66\varepsilon_k + 1.66\varepsilon_f - 1.66\mu + 0.37t_k + 0.69U_k)^2}{100K_B T^2} \\
& \times \left( e^{\frac{-(1.66\varepsilon_k + 1.66\varepsilon_f - 1.66\mu + 0.37t_k + 0.69U_k)}{100K_B T}} \right)
\end{aligned} \quad (13b)$$

Equation (13b) is the expression for determining specific heat capacity in s-wave superconducting model.

### E. Transition temperature of s-wave superconducting state

Transition temperature of the superconducting state,  $T_c$  is calculated from the condition;

$$\left[ \frac{\partial C}{\partial T} \right]_{T=T_c} = 0 \quad (14)$$

$$T_c = \frac{(1.66\varepsilon_k + 1.66\varepsilon_f - 1.66\mu + 0.37t_k + 0.69U_k)}{200K_B} \quad (15)$$

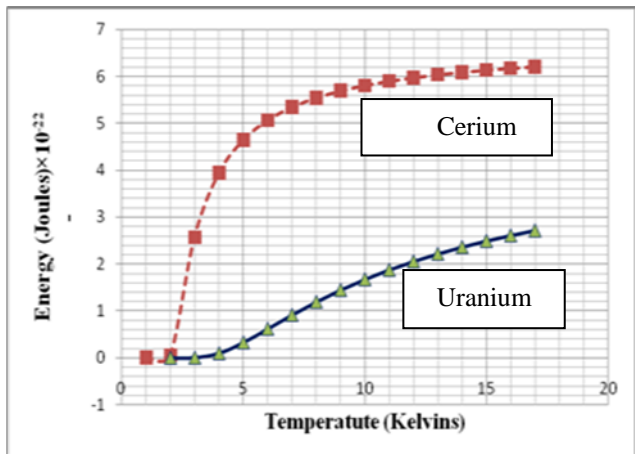
Equation (15) is the expression for determining the transition temperature for heavy fermion systems.

## III. RESULTS AND DISCUSSION

The heavy fermion system total specific heat at constant volume  $C_V$  is the specific heat of the gas of Cooper-pairs plus the specific heat of the gas of electrons (fermions). One of the characteristics quantities in the thermodynamic properties of heavy fermion systems is the specific heat, which can be obtained by evaluating the temperature-derivative of the internal energy as in equation (12). In Fig. 1, we plot the specific heat  $C_V$  as a function of temperature for both the Cerium and Uranium based heavy fermion systems.

The parameters used for Cerium compounds are  $E_k=0.1\text{eV}, E_f= -3\text{eV}, \mu= -0.52\text{eV}, t_k= -0.4\text{ eV}, U=6\text{ eV}$  and for Uranium compounds  $E_k=0.1\text{eV}, E_f= -3\text{eV}, \mu= -0.52\text{eV}, t_k= -0.43\text{ eV}, U=6\text{ eV}$  [21].

A graph depicting the variation of system energy versus temperature is as shown in figure 1



**Figure1: Variation of System Energy against Temperature for Cerium and Uranium compounds.**

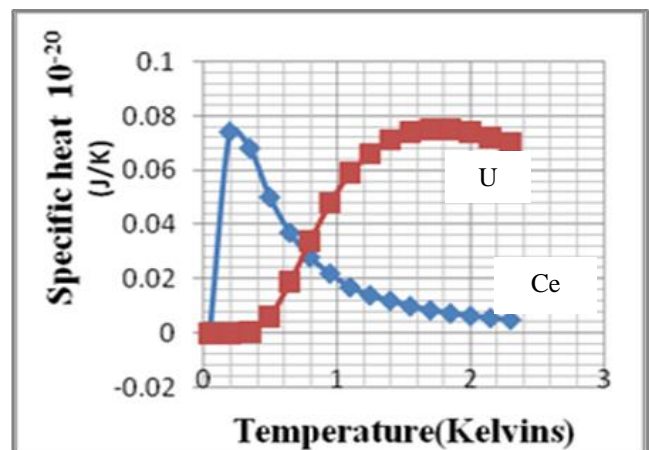
The value of E decreases below  $T_c$  (K) and goes to zero at  $T=0$  K and this is consistent with the nature of super-fluid state. The total energy of the system increases with increase in temperature of the system.

The Uranium based compounds show a system energy that is lower than that of Cerium based compounds both at low and high temperatures. Cerium based compounds show a low system energy at low temperatures and a higher one at higher temperatures which is in agreement with their atomic properties [23] when the two compounds are studied comparatively.

There exists an exponential increase in the energy of the system as the temperature increases approaching a plateau like state dependent on type of the material. It was noted that energy of interaction between Cooper pair is a stretched sigmoid shaped curve. Similar shapes of curves relating energy and temperature has been noted by other scientists [24], [25] and [26]. The rate of increase of energy with temperature for Uranium is lower than that of Cerium. The variation of specific heat with temperature is studied using equation (13b).

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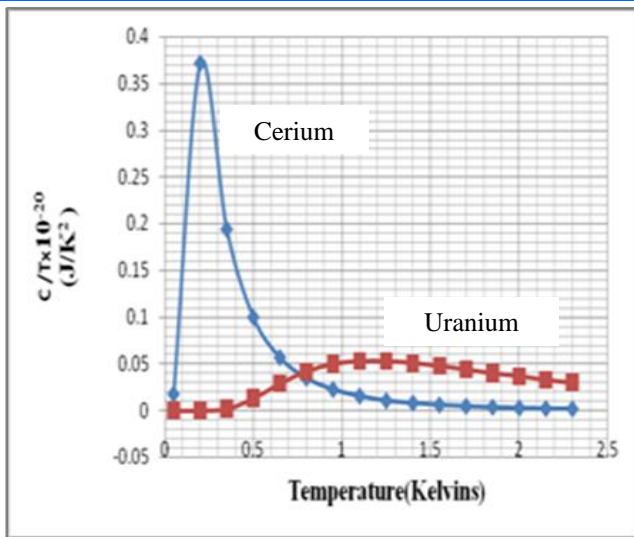
**Figure 2: The graph of specific heat against temperature**

The heat capacity shows a turning point at  $T=1.74$ K and  $T=0.24$ K for Uranium and Cerium compounds respectively and decreases as  $T \rightarrow 0$ K. The heat capacity is exponentially small at low temperatures. The results compares with that of Sakwa et al, 2013 on their study on a grand-canonical binary system at low temperatures. From the graph of specific heat in figure 2, Gaussian shaped curves relating specific heat for Cerium and Uranium based compounds is noted. This type of shape was observed by other scientists [24], [25] and [26]. The maximum value of specific heat capacity for the Uranium-based compounds is maintained at  $0.006$ eV/K and  $0.005$ eV/K for Cerium based compounds. The specific heat jump of these compounds is strongly influenced by the presence of magnetic impurities. The reduction of the specific heat jump corresponds to pair breaking situations. The jump corresponds to  $T_c$ . At  $T \gg T_c$  the impurity spin is essentially free to fluctuate driven by thermal agitation; the strongly temperature dependent pair breaking mechanism acts upon the system in the superconducting state and leads to the strong reduction of the specific heat jump. At  $T \ll T_c$  the impurity spin is locked into a singlet bound state and no longer takes part in dynamics.

$$\text{At the peak or turning point of each graph, } \left[ \frac{\partial C}{\partial T} \right]_{T=T_c} = 0$$

hence the temperature corresponding to the peak is critical temperature of the superconductor. Clearly,  $T_c$  for Uranium-based compounds is approximately  $1.74$ K and  $0.24$ K for Cerium based compounds which are approximately equal to the experimental values by Chen & Wang, 2016. However the Cerium based compounds seem to show strong agreement with the experimental results. The values of electronic specific heat in response to changes near zero Kelvin temperature are studied as shown;





**Figure 3: The graph of specific heat divided by temperature against temperature.**

The electronic density of states near the Fermi surface is predicted to be higher for Cerium based compounds than the Uranium based compounds by this model.

### CONCLUSIONS

The temperature dependence of specific heat at  $T < T_c$  for Cerium based compounds is strongly reminiscent of that of an isotropic  $s$ -wave. We have explicitly demonstrated that superconductivity is a bulk phenomenon as exhibited by a hump-like feature on the specific heat Figure 2: Variation of specific heat with temperature for Cerium and Uranium curves. The peak value of the Gaussian curves of specific heat represents the superconducting transition temperature of the heavy fermion systems. At this point, a condensate is formed and  $C_v$  remains fairly constant. This depicts that the system is unstable at the peak and a second order phase transition (normal metal to superconducting state) occurs due to absence of latent heat. In general, the total specific heat of any system is the sum of several different excitations. Specific heat need to be explored in order to unravel the magnitude of different contributions to the total specific heat.

We have demonstrated that the pairing in Cerium compounds is basically  $s$ -wave superconductivity. However the uranium based compounds need to be further investigated as it will be shown in our future publication. Some heavy-fermion compounds, such as  $CeRu_2$ ,  $CeCo_2$ , and the recently reinvestigated  $CeCu_2Si_2$ , are known to exhibit  $s$ -wave superconductivity which is in good agreement with our working [15].

### ACKNOWLEDGEMENTS

The authors would like to express their gratitude to Masinde Muliro University of Science and Technology for the opportunity and enabling environment to conduct this research.

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