Multiband superconductivity in Lu\textsubscript{3}Os\textsubscript{4}Ge\textsubscript{13}

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Abstract

Intermetallic R\textsubscript{3}T\textsubscript{4}X\textsubscript{13} series consist of a cage-like structure and have attracted attention due to their unconventional electronic ground states. In this work, we report the normal and superconducting state properties of a high-quality single crystal of Lu\textsubscript{3}Os\textsubscript{4}Ge\textsubscript{13}. Lu\textsubscript{3}Os\textsubscript{4}Ge\textsubscript{13} belongs to the above-mentioned series and crystallizes in a cubic crystal structure with the space group $Pm\overline{3}n$. Using electrical transport, magnetization and heat capacity measurements, we show that Lu\textsubscript{3}Os\textsubscript{4}Ge\textsubscript{13} is a type-II multi-band superconductor ($T_c = 3.1$ K) with unusual superconducting properties. The analysis of the low-temperature heat capacity data suggests that Lu\textsubscript{3}Os\textsubscript{4}Ge\textsubscript{13} is a moderately coupled multi-band BCS superconductor with two gaps ($2\Delta/k_B T_c = 3.68 \pm 0.04$ & $0.34 \pm 0.02$) in the superconducting state. The dc-magnetization $(M-H)$ shows a large reversible region in the superconducting state similar to the vortex liquid phase observed in high-$T_c$ superconductors. The value of the Ginzburg number $G_I$ suggests that the thermal fluctuations, though small compared to those in high-$T_c$ cuprates, may play an important role in the unpinning of the vortices in this compound. The electronic band structure calculations show that three bands cross the Fermi level and constitute a complex Fermi surface in Lu\textsubscript{3}Os\textsubscript{4}Ge\textsubscript{13}.

Keywords: superconductivity, multi-band, multi-gap, vortex liquid, band structure, heat capacity, resistivity, magnetization

1. Introduction

Ternary intermetallic compounds with the formula R\textsubscript{3}T\textsubscript{4}X\textsubscript{13}, where R is a rare-earth element, T transition metal and X semi-metallic/semiconducting element, have significantly contributed to the understanding of the physics of strongly correlated materials. These compounds show a variety of unusual magnetic ground states and superconductivity at low temperatures [1]. One such structure with no metalloids was reported by Remeika et al [2]. These compounds (R\textsubscript{3}T\textsubscript{4}X\textsubscript{13}) crystallize in a cubic structure (space group $Pm\overline{3}n$) and feature a cage-like environment within the unit cell. Amongst these, La-, Yb-, and Th-based compounds are superconducting, whereas Gd- and Eu-based compounds show magnetic transition. By replacing Sn with Ge and Rh with Ru in R,Rh\textsubscript{4}Sn\textsubscript{13}, Segre and Braun [3] reported superconductivity and magnetic ordering. Our studies on polycrystalline samples of R\textsubscript{3}Ru\textsubscript{4}Ge\textsubscript{13} series (R = Y, Ce, Pr, Nd, Ho, Er, Dy, Yb, Lu) showed [4–6] that the series exhibit unusual physical properties. These properties could be considered as those belonging to the semi-metals or low band-gap semiconductors. Studies on Lu- and Ru-based polycrystalline samples [6] reported superconductivity below 2.3 K and 2.8 K respectively. These compounds are also reported as good thermo-electric materials [7]. The multi-valley nature of the band structure [8], cage-like coordination in the crystal structure and occurrence of superconductivity make iso-structural Lu\textsubscript{3}Os\textsubscript{4}Ge\textsubscript{13} an interesting compound in which to look for unusual superconductivity. Bulk studies show that Lu\textsubscript{3}Os\textsubscript{4}Ge\textsubscript{13} is a multi-band superconductor and that it shows a large reversible region in the magnetization (M-H) data, which has typically been observed in high-$T_c$ superconductors [10]. In this report, we present a detailed study of the anomalous superconducting properties of a Lu\textsubscript{3}Os\textsubscript{4}Ge\textsubscript{13} single crystal, supported by the electronic band structure calculations.
2. Methods

2.1. Sample preparation and characterization

The single crystal of Lu₃Os₄Ge₁₃ was grown using the Czochralski crystal pulling method in a tetra-arc furnace under inert argon atmosphere. A stoichiometric mixture (10 g) of highly pure elements (Lu: 99.99%, Os: 99.99%, Ge: 99.99%) was melted 4–5 times in the same furnace to make a homogeneous polycrystalline sample. The single crystal was pulled from the polycrystalline melt using a tungsten seed rod at the rate of 10 mm h⁻¹ for about 6 h to get 5–6 cm long cylindrical shaped crystals with 3–4 mm diameter. Lu₃Os₄Ge₁₃ crystallizes in a cubic crystal structure with space group Pm₃n (space group number 223) with 40 atoms per unit cell (2 formula unit). The structure is similar to that of the isostructural compound Y₃Ru₄Ge₁₃ [18]. The unit cell consists of a cage-like crystal structure as shown in the left panel. The Rietveld analysis and circular spots in the Laue diffraction confirm the high-quality, single-phase and single-crystalline nature of the grown crystal.

The single crystal was characterized using various experimental techniques such as room temperature powder x-ray diffraction (PXRD) and energy dispersive x-ray spectroscopy (EDX) and was confirmed to be a well-defined single crystal with no trace of impurity phases. The EPMA and EDX characterizations were done on the well-polished surfaces and confirmed the proper stoichiometry (3: 4: 13) and single-phase nature of the Lu₃Os₄Ge₁₃ compound. The single crystals were oriented along the crystallographic direction [100] via the Laue back reflection method using a Huber Laue diffractometer and cut to the desired shape and dimensions using a spark erosion cutting machine.

2.2. Measurement techniques

The electrical resistivity was measured using the standard four-probe technique in a homemade setup. We used 40 μm diameter Au wires for making the electrical connections using indium solder. The contact resistance is of the order of 10 mΩ. The zero magnetic field data was recorded in the temperature range 1.6–300 K using an LR700 resistance

![Figure 1.](image-url)
bridge. The electronic resistivity was measured in constant magnetic fields (0–5.5 T) in a Cambridge magnetic refrigeration (CMR) mFridge refrigerator setup from 0.1–4.2 K for determining the upper critical field \( \mu_0 H_{c2}(T) \). The CMR setup can reach a base temperature of \( \approx 100 \text{ mK} \) using adiabatic demagnetization of paramagnetic salt pills. Magnetic susceptibility was measured using a commercial superconducting quantum interferometer device (SQUID) magnetometer (MPMS5, Quantum Design, USA) in a constant magnetic field of 10 mT; the sample was cooled down to 1.8 K in a zero-magnetic field and then the magnetic field was applied, followed by warming to 5 K (this is called the zero field cooled or ZFC data). Then the sample was cooled down to 1.8 K in the magnetic field of 10 mT to take field cooled (FC) data. The heat capacity was measured using a physical property measurement system (PPMS) equipped with a He\(^3\) dilution refrigerator in the temperature range 0.05–4 K and 1.8–300 K by a time-relaxation method in different magnetic fields from 0 to 7 T.

### 2.3. Band structure calculations

The electronic band structure calculations were performed by density functional theory (DFT) using WIEN2k code with a full-potential linearized augmented plane-wave and local orbitals (FP-LAPW + lo) basis [20, 21], together with Perdew–Burke–Ernzerhof (PBE) parametrization [23] of the generalized gradient approximation (GCA), with no spin–orbit coupling. The plane-wave cutoff parameter \( R_{MF} K_{\text{MAX}} = 7 \) was taken with 5000 k-points (the choice of number of k-points varies with symmetry of the crystal structure, and lower symmetry structure, may require a larger number of k-point samplings). The program xCrysden was used for calculating and plotting the bands and Fermi surface.

### 3. Results and discussion

Figure 2(a) shows the electrical resistivity of Lu\(_3\)Os\(_4\)Ge\(_{13}\) for current parallel to the [100]-direction measured in the temperature range from 2 to 300 K. A negative temperature coefficient of resistivity \( (\frac{\Delta \rho}{\Delta T} < 0) \) is observed in the normal state. This can result from several mechanisms such as site disorder, electronic correlations and low-energy phonon scattering. The compound under study has a cage-like structure with a loosely bound Ge atom within the cage, which gives rise to the low-energy soft phonon modes resulting in significant electron–phonon scattering at low temperatures. The resistivity becomes zero at the onset of superconductivity below 3.1 K. The inset in figure 2(a) shows the change in the superconducting transition temperature in different magnetic fields. The width of the superconducting transition \( (\Delta T) \) increases with increasing magnetic field. The transition temperature is taken at the point where the resistivity drops to 50% of the normal state value. Figure 2(b) shows the temperature dependence of the upper critical field \( \mu_0 H_{c2} \). A linear relationship is observed between \( H_{c2} \) and temperature in the proximity of the transition temperature \( (T_c, H = 0) \).

In type-II superconductors, an external magnetic field leads to the Cooper pair breaking via two mechanisms, i.e. the orbital and spin paramagnetic effects [9, 26, 28] (the latter is also known as the Pauli paramagnetic limiting effect). The orbital pair breaking is related to the emergence of Abrikosov vortices. The orbital limiting field refers to the magnetic field at which the vortex cores fill the whole volume and is given by the formula

\[
H_{c2}^{\text{orb}}(0) = \frac{\Phi_0}{2\pi \xi^2},
\]

where \( \xi \) is the Ginzburg–Landau coherence length and \( \Phi_0 \) (= \( hc/2e = 2.07 \times 10^{-15} \text{Tm}^2 \)), is the magnetic flux quantum. For the single-band BCS superconductors, \( H_{c2}^{\text{orb}}(0) \) is
derived from the slope of the \( H_c(T) - T \) phase boundary at \( T_c \), given by,

\[
\mu_0 H_{c2}^{\text{orb}}(0) = -0.69 T_c \frac{dH_{c2}}{dT} \bigg|_{r=r_0}, \quad (2)
\]

in the dirty limit and

\[
\mu_0 H_{c2}^{\text{orb}}(0) = -0.73 T_c \frac{dH_{c2}}{dT} \bigg|_{r=r_0}, \quad (3)
\]

in the clean limit of the Werthamer–Helfand–Hohenberg (WHH) theory [11].

The spin paramagnetic pair-breaking mechanism is related to the Zeeman splitting of the spin singlet Cooper pairs due to the interaction of the magnetic field with electron spins. For a BCS superconductor, the Pauli limiting field is given by \( H_{P}^{\text{Pauli}} = 1.84 T_c \). The upper critical field \( \mu_0 H_{c2} \) is influenced by both orbital and spin paramagnetic effects. Orbital pair breaking is the dominant mechanism at low magnetic fields and the Pauli paramagnetic effect dominates the upper critical field at very high magnetic fields. The relative importance of the orbital and Pauli limiting fields is described by the Maki parameter \( \alpha \) [12] defined as

\[
\alpha = \sqrt{2} H_{c2}^{\text{orb}}(0) / H_{P}^{\text{Pauli}}(0). \quad (4)
\]

The value of the orbital critical field calculated using equation (2) is 5.45 T. The Ginzburg–Landau coherence length, \( \xi(0)_{GL} \), is given by,

\[
\xi(0)_{GL} = \sqrt{\frac{\Phi_0}{2\pi H_{c2}^{\text{orb}}(0)}}. \quad (5)
\]

Using equation (5), we get \( \xi(0)_{GL} = 78 \AA \). Using the values of \( \mu_0 H_{c2}^{\text{orb}}(0) \) and \( \mu_0 H_{P}^{\text{Pauli}} \), the value of the Maki parameter comes out to be \( \alpha = 1.33 \).

The temperature dependence of \( H_{c2} \) for single-band, dirty limit superconductors is given by the WHH formula

\[
\ln \left( \frac{1}{1} \right) = \sum_{\nu=-\infty}^{\infty} \left\{ \frac{1}{|2\nu + 1|} - \left[ \frac{\nu + 1}{t} + \frac{\hat{R}}{t} \right]^{2} \right\} \quad (6)
\]

where \( t = T/T_c, \hat{R} = 4H_{c2}(T)/(\pi^2 T_c) \left( \frac{dH_{c2}(T)}{dT} \right) \), \( \alpha \) is the Maki parameter, and \( \lambda_{so} \) is the spin–orbit scattering constant. When \( \lambda_{so} = 0, H_{c2}(0) \) obtained from the WHH formula satisfies the relation

\[
H_{c2}(0) = H_{c2}^{\text{orb}}(0) \sqrt{1 + \alpha^2}. \quad (7)
\]

In figure 2(b), we note that the experimental \( H_{c2} \) versus \( T \) data is best described by the WHH formula for \( \alpha = 0 \) \( (H_{c2}^{\text{orb}}(0) \ll H_{P}^{\text{Pauli}}(0)) \), \( \lambda_{so} = 0 \) (red dashed curve). If we use \( \alpha = 1.33 \), as derived above, the WHH formula does not explain the data except when \( \lambda_{so} \approx 100 \), which is clearly unphysical. This suggests that single-band models are not enough to describe the temperature dependence of upper critical field \( H_{c2}(T) \) and multi-band effects have to be included in the models to describe the data. The multi-band models [27] need parameters like inter-band coupling constants and knowledge of Matsubara frequencies for the specific compound and these are not yet known for Lu$_4$Os$_8$Ge$_{13}$. Assuming moderate multi-band effects present in this compound, if we do a linear extrapolation of the \( H_{c2}(T) \) at \( T = 0 \) in figure 2(b), we get an estimate of the \( H_{c2}(0) = 6.8 T_c \), which is \( \approx 1 T \) more than the value obtained from WHH theory. With this observation, to a first-order approximation, we can use single-band models to get approximate values of superconducting state parameters, like coherence length, penetration depth etc.

Figure 3(a) shows the zero field heat capacity data of Lu$_4$Os$_8$Ge$_{13}$ in the temperature range 1.8–300 K. The experimental value of heat capacity \( C(T) = 504 \) Jmol$^{-1}$K$^{-1}$ is very close to the Dulong–Petit high-temperature limit of the lattice heat capacity \( C_l = 3N_{R} = 498.9 \) Jmol$^{-1}$K$^{-1}$. The electronic contribution to the heat capacity \( C_{el}(T) \) can be calculated by subtracting the total heat capacity contribution from the total heat capacity \( C(T) \), i.e., \( C_{el}(T) = C(T) - \beta T^3 \). Figure 3(b) shows a sharp jump in the heat capacity at 3.1 K which confirms the bulk superconductivity in the compound. Figure 3(e) shows the heat capacity data measured in different magnetic fields \( (H \parallel [100]) \). The magnitudes of the heat capacity jump as well as \( T_c \) decrease with increasing magnetic field.

The superconductivity is fully suppressed in a magnetic field of \( 7T \) and the data is fitted to the equation \( C(T) = \gamma_{ph} T + \beta T^3 \) as shown in figure 3(b), where \( \gamma_{ph} T \) represents the electronic contribution and \( \beta T^3 \) describes the lattice–phonon contribution to the specific heat in the normal state. We find the electronic specific heat coefficient \( \gamma_{el} = 25.4 \pm 0.3 \) mJmolK$^{-2}$ and the phonon/lattice contribution coefficient \( \beta = 2.30 \pm 0.05 \) mJmolK$^{-4}$ from the fit. The Debye temperature \( (\Theta_D) \), calculated using the formula

\[
\Theta_D = \left( \frac{12\pi^4 R N / 5\beta}{T_c} \right)^{1/2}, \quad (8)
\]

where \( R \) is the molar gas constant and \( N (= 20) \) is the number of atoms per formula unit (f.u.), is 256.6 ± 0.7 K. The density of states at the Fermi level calculated using the formula \( \gamma_{el} = (\pi^2 k_{B}^2 / 3) D(E_F) \), is \( D(E_F) = 21.3 \) states/eV-f.u. for both spin directions. This density of state contains quasiparticle mass enhancement by many-body electron–phonon interaction and is related to bare density of states \( D_{band}(E_F) \) by \( D(E_F) = (1 + \lambda_{eph})D_{band}(E_F) \), where \( \lambda_{eph} \) is the dimensionless electron–phonon coupling constant. The value \( \lambda_{eph} \) is related to the phonon spectrum and density of states in Eliashberg theory [13] and represents the strength of electron–phonon coupling. The value \( \lambda_{eph} \) can be calculated using McMillan’s formula [14],

\[
\lambda_{eph} = \frac{1.04 + \mu^* \ln(\Theta_D / 1.45T_c)}{(1 - 0.62\mu^*)\ln(\Theta_D / 1.45T_c) - 1.04}, \quad (9)
\]

where \( \mu^* \) is the repulsive screened coulomb parameter. The competition between \( \mu^* \) and \( \lambda_{eph} \) is the determining factor for Cooper pairing in conventional superconductors [15]. The value of \( \mu^* \) is taken as 0.13. Using equation (9), we obtain
This suggests a moderately enhanced electron–phonon coupling in Lu$_3$Os$_4$Ge$_{13}$. Combining the values of \( \lambda_{\text{eph}} \) and \( D(E_F) \), we get \( D_{\text{band}}(E_F) = 13.48 \text{ states/eV-f.u.} \) for both spin directions. The effective mass of the quasiparticles \( (m^*) \), calculated using the relation, \( m^* = (1 + \lambda_{\text{eph}}) m_{\text{band}} \) is \( m^* = 1.58 m_e \), assuming effective band mass \( m_{\text{band}} = m_e \), the free electron mass.

The sharp jump in the electronic heat capacity \( \Delta C_{el}(T) \) at \( T_c \) is 89.6 mJ/mol K, giving the ratio \( \Delta C_{el}/\gamma N T_c = 1.15 \). This ratio can be used to understand the strength of the

\[ \gamma_N(H) = 13.07^*H^{0.38\pm0.03} \]
electron–phonon coupling. This ratio is smaller than the weak-coupling limit value of 1.43 for a conventional BCS superconductor. If we consider a single-band model [16] for a BCS superconductor, such a reduction in the heat capacity jump can be either due to the presence of an anisotropic superconducting energy gap or the presence of multiple gaps at the Fermi surface in the superconducting state.

Apart from the reduced jump in the heat capacity at \( T_c \), the low-temperature electronic heat capacity is higher than the expected value for a single gap s-wave BCS superconductor (see figures 3(c) and (d)). To analyze the suppression in the heat capacity jump and the low temperature data in more detail, we have used an empirical (and not self-consistent) one-band one-gap model [16, 22] that accounts for the anisotropy in the order parameter \( \Delta_0 \) at the Fermi surface and a two-band two-gap model [16, 22]. In these models, the superconducting energy gap \((\Delta(t))\) is parametrized in terms of the normalized BCS gap \((\delta(t))\), \(\Delta(t) = \Delta_0 \delta(t)\), where \( t = T/T_c \) is the reduced temperature. The normalized BCS gap as a function of reduced temperature is taken from Muhlschlegel’s paper [29]. The entropy \((S)\) and the heat capacity \((C)\) for a system of independent fermionic-quasiparticles can be written as

\[
\frac{S}{\gamma_n T_c} = -\frac{6}{\pi^2 k_B T_c} \int_0^\infty \left[ f \ln f + (1-f) \ln(1-f) \right] \frac{1}{\Delta_0} \, dy,
\]

\[
\frac{C_{el}}{\gamma_n T_c} = \int \frac{d(S/\gamma_n T_c)}{dt},
\]

where \( f = \left[ e^{\beta E} + 1 \right]^{-1}, \beta = (k_B T)^{-1}\) and \( y = \epsilon/\Delta_0\). The quasiparticle energy \((E)\) is \(\sqrt{\epsilon^2 + \Delta^2(t)}\), where \(\epsilon\) is the energy of the normal electrons measured from the Fermi level. Using equations (10) and (11), we can write the electronic heat capacity for the one-gap \(\alpha\)-model as

\[
C_{el}(\alpha, \epsilon, t) = a \int_0^\infty \left[ \frac{x^3}{t^2} + \alpha^2 \left( \frac{\delta(t)}{t} \right)^2 \right]
- \alpha^2 \left( \frac{\delta(t)}{t} \right) \left( \frac{d\delta(t)}{dt} \right)
\times \operatorname{sech}^2 \left( \frac{x}{t} + \alpha \left( \frac{\delta(t)}{t} \right)^2 \right) \, dx,
\]

where \( a = 12\gamma_n T_c/\pi^2, x = \epsilon/2k_BT_c\) and \( \alpha = \Delta/2k_BT_c\). In the two-gap model, the total electronic heat capacity, \(C_{el}(a_1, a_2, a_2, a_2, t)\), is taken as the sum of the independent contributions from two-bands with different superconducting energy gaps, each following BCS-type temperature dependence (if we neglect inter-band transitions due to scattering by impurities or phonons, and assume that \(\gamma_{a_1} + \gamma_{a_2} = \gamma_n\)). For the two-gap \(\alpha\)-model, the total heat capacity is given by

\[
C_{tot}(a_1, a_1, a_2, a_2, a_2, t) = C_{el}(a_1, a_1, t) + C_{el}(a_2, a_2, t),
\]

where \( a_1 = 12\gamma_{a_1} T_c/\pi^2, a_1 = \Delta_1/2k_BT_c, a_2 = 12\gamma_{a_1} T_c/\pi^2 \) and \( a_2 = \Delta_2/2k_BT_c\). The subscripts ‘1’ and ‘2’ stand for large and smaller gap respectively.

We estimate the fitting parameters \((\alpha, \gamma)\) for the one-gap \(\alpha\)-model, and \((\alpha_1, \alpha_2, \alpha_2, \alpha_2)\) for the two-gap \(\alpha\)-model using a nonlinear least-squares fit for two different samples in order to confirm the reproducibility of the results. To estimate the uncertainties in the fitted parameters, we repeat the whole process for 1000 realizations of the data obtained by adding random perturbations to the data with the standard deviation equal to \(1\sigma\) uncertainty in the corresponding data point for both samples. The median value of each parameter for 1000 realizations was accepted as the fitted value, while the \(\pm 1\sigma\) uncertainty in the fitted value was estimated from the range covering 34\% of the area on either side of the median in the distribution function of the fitted parameter. Figures 3(c) and (d) show the fit to the electronic specific heat for the two samples using a one-gap \(\alpha\)-model and two-gap \(\alpha\)-model. The two-gap \(\alpha\)-model fails to fit the data at the low temperatures with large values of chi-square per degree of freedom \((\chi^2_{\text{df}} = 18.8\) and 20.1 for sample #1 and sample #2 respectively). The low-temperature electronic heat capacity of both samples is best described by the two-gap \(\alpha\)-model with \(\chi^2_{\text{pdf}} = 1.31(1.43)\). Clearly, the two-gap \(\alpha\)-model fits the data significantly better than the one-gap \(\alpha\)-model. We find the two gaps to be \(2\Delta_1/k_BT_c = 3.68 \pm 0.04 (3.69 \pm 0.08)\) and \(2\Delta_2/k_BT_c = 0.34 \pm 0.02 (0.31 \pm 0.05)\) for sample #1 (sample #2). The contributions of \(\gamma_{a_1}\) and \(\gamma_{a_2}\) to \(\gamma_n\) are 81.5\% and 18.5\% respectively.

The study of vortex excitations in the mixed state provides insight into the understanding of the superconducting order parameter. Figure 3(e) shows the low-temperature electronic heat capacity in different magnetic fields \((H_{dC} < H < H_{c2})\). The linear extrapolations to zero temperature give the heat capacity contribution due to normal electrons in the mixed state, and are determined in terms of the electronic specific heat coefficient \(\gamma_n(H)\). This electronic contribution is attributed to the normal state electrons present in the vortex cores. In s-wave superconductors, the vortex cores contribute to the electronic heat capacity as normal metals. This contribution is proportional to the number of vortices and hence proportional to the applied magnetic field, i.e. \(\gamma_n(H) \propto H\). However, we find that \(\gamma_n(H) = 13.07 H^{0.38} \pm 0.03\) as shown in the figure 3(f). A nonlinear dependence of \(\gamma_n(H)\) on the magnetic field has been argued to be intrinsic property of the multi-band, multi-gap superconductors [25]. This analysis also suggests the presence of multiple gaps at the Fermi level in the superconducting state and indicates multi-band superconductivity in the Lu$_3$Os$_4$Ge$_{13}$ single crystal.

Figure 4(a) shows a diamagnetic transition into the superconducting state at 3.1 K in the low-temperature dc-susceptibility data. A significant amount of vortex pinning can be observed by comparing the zero field cooled (ZFC) and the field cooled (FC–Meissner) data below the transition temperature. The sample is weakly paramagnetic at room temperature \((\chi = 3 \times 10^{-4} \text{ emu/mol at } T = 300 \text{ K})\). The
temperature dependence of the susceptibility is of Pauli paramagnetic type with a small rise at low temperature \((c = -81.03 \text{ emu/mol at } T = 3.2 \text{ K})\) possibly due to the presence of small magnetic rare-earth impurities (ppm level) in Lu. We estimate the value of the lower critical field \(H_{c1}(1.8 \text{ K}) = 20 \text{ mT}\) from magnetization measurements. The value of the lower critical field \(H_{c1}(0) = 30 \text{ mT}\) is obtained using the following expression

\[
H_{c1}(0) = H_{c1}(T) \left(1 - \left(\frac{T}{T_c}\right)^2\right).
\]

Substituting the values of \(\xi_{\text{GL}}(0)\) and \(H_{c1}(0)\) in the following expression:

\[
H_{c1}(0) = \frac{\Phi_0}{4\pi \lambda_{\text{GL}}(0)} \ln \left(\frac{\lambda_{\text{GL}}(0)}{\xi_{\text{GL}}(0)}\right),
\]

we obtain \(\lambda_{\text{GL}}(0) = 4736 \text{ Å}\). The Ginzburg–Landau parameter \(\kappa_{\text{GL}}(0) = \frac{\lambda_{\text{GL}}(0)}{\xi_{\text{GL}}(0)} = 61\). Using the formula

\[
H_{c1}(0) = \frac{H_{c1}(0)}{\sqrt{2\kappa}} (\ln \kappa + 0.5),
\]

Figure 4. (a) DC magnetic susceptibility data as function of temperature. The superconducting transition temperature, as determined from susceptibility measurement is in excellent agreement with the resistivity data. The ZFC and FC susceptibility data indicate a significant number of pinning of vortices in the compound. (b) Magnetization as a function of magnetic field for \(H \parallel [100]\) direction. The \(M-H\) loop is closed (\(\delta M = 0\)) at magnetic fields \(H > 0.45 \text{ T}\) at 1.8 K, which suggests possible melting of vortices at 0.45 T field.

Figure 5. (Left panel) Band structure of Lu$_3$Os$_4$Ge$_{13}$ near Fermi level. The band structure shows a multi-valley type character. Three bands shown in bold lines cross the Fermi surface. (Right panel) Analysis of the density of states of Lu$_3$Os$_4$Ge$_{13}$. The total DOS curve has a local maximum at the edge of the Fermi energy. The partial density of states curves show that the major contribution to the total density of states is coming from Os and Ge atoms. Lu atoms have the least contribution to the total density of states.
we obtain the value of the thermodynamic critical field $H_i(0) = 564 \text{ mT}$. The magnetization ($M-H$) data as shown in figure 4(b) shows that the $M-H$ loop is closing at magnetic fields $H > 0.45$ T at 1.8 K, suggesting the unpinning (melting) of vortices in magnetic fields much smaller than the upper critical field ($\mu_0H_u(1.8 \text{ K}) = 3.2$ T). An unpinned vortex phase exists in the magnetic field region $0.45 \text{ T} \leq H \leq \mu_0H_u(1.8 \text{ K}) = 3.2$ T, which may be a vortex liquid phase. A similar vortex liquid phase is observed in high-$T_c$ superconductors [10] and is an unusual phenomenon in low-$T_c$ superconductors. Though the melting of the vortices in high-$T_c$ superconductors is attributed to quantum and thermal fluctuations [10, 17], there is no clear understanding of the origin of the vortex liquid phase in low-$T_c$ superconductors. The strength of the thermal fluctuations, which leads to the vortex unpinning, is described in terms of the Ginzburg number given by

$$G_i = \frac{1}{2} \left( \frac{k_B \mu_0 \Gamma H_{c1}(0)}{4\pi \xi^3(0) H_{c2}(0)} \right)^2,$$  (17)

where $\Gamma$ is the anisotropy parameter ($\approx 1$ for cubic Lu$_3$Os$_4$Ge$_{13}$). Using equation (17), we get the value of the Ginzburg number $G_i = 4.1 \times 10^{-6}$. The value of Ginzburg number for Lu$_3$Os$_4$Ge$_{13}$ is larger than low-$T_c$ superconductors ($\approx 10^{-8}$) but smaller than high-$T_c$ superconductors ($\approx 10^{-2}$) [10, 17]. This suggests that the thermal fluctuations, though weak, may play an important role in the unpinning of the vortices in this compound.

Figure 6. Contributions of the bands crossing the Fermi level to the Fermi surface. All three bands are plotted together make a complex surface.
Table 1. Crystal structure parameters obtained from the Rietveld refinement of the room temperature powder x-ray diffraction data of Lu₂Os₄Ge₁₃. Profile reliability factor Rp = 17.5%, weighted profile R-factor Rw = 17.2%, Bragg R-factor = 8.46% and Rp-factor = 7.44% were obtained from the best fit.

| Structure | Cubic |
|-----------|-------|
| Space group | Pm3n (No. 223) |
| Lattice parameters | |
| a (Å) | 8.94585(22) |
| Vcell (Å³) | 715.921(0.030) |
| Atomic coordinates | |
| Atom | Wyckoff position | x | y | z |
|-------|-----------------|----|---|---|
| Lu | 6c | 0.25 | 0 | 0.50 |
| Os | 8e | 0.25 | 0.25 | 0.25 |
| Ge1 | 24k | 0 | 0.31206(30) | 0.14965(32) |
| Ge2 | 2a | 0 | 0 | 0 |

Type-II superconductors in which H₁ and H₂ are well separated (H₂/ H₁ ~ κ²) are classified as strongly type-II superconductors. This (H₁ ≪ H₂) leads to a situation in which the magnetic fields associated with vortices overlap and the superposition becomes nearly homogeneous, while the order parameter characterizing superconductivity is still inhomogeneous [17]. Detailed penetration depth (λGL) measurements are required to obtain correct values of H₁ and κ.

The analysis of the electronic band structure and density of states of Lu₂Os₄Ge₁₃, based on the electronic structure calculations using Wien2K, is presented below. The left panel in figure 5 shows the band structure of Lu₂Os₄Ge₁₃ near the Fermi level. The band structure has a multi-valley type character [8]. Three bands cross the Fermi level and account for the metallic nature of the compound. The right panel in figure 5 shows the calculated density of states (DOS) for one formula unit (20 atoms) of Lu₂Os₄Ge₁₃.

The Fermi level is located near the edge of a local maxima in the total density of states. The value of the DOS at E_F is ~17 states/eV-f.u for both spin directions. This value is smaller than the value (~21 states/eV-f.u for both spin directions) calculated using the value of γω obtained from heat capacity measurements, which indicates the mass-enhancement in the compound, since no electronic correlations were taken into account in the band structure calculations. The partial DOS shows that the total DOS is dominated by contributions from Os and Ge. Figure 6 shows the calculated Fermi surfaces for the three bands (as well as all the bands plotted together) which cross the Fermi level. The combination of these bands leads to a complex Fermi surface (figure 6).

4. Conclusion

In summary, we have studied the superconducting properties of Lu₂Os₄Ge₁₃ in detail using electrical transport, magnetization and heat capacity measurements. We show that Lu₂Os₄Ge₁₃ is a multi-band type-II superconductor (Tc = 3.1 K) by analyzing the low-temperature heat capacity data using an empirical two-gap multi-band α-model. The single-band WHH model does not fully explain the temperature dependence of the upper critical field Hc2(T), suggesting the presence of multi-band effects in the Lu₂Os₄Ge₁₃. The analysis of the low-temperature heat capacity data for two different samples of Lu₂Os₄Ge₁₃ single crystal using a two-gap α-model confirms the presence of two superconducting gaps (2Δ₁/kB Tc = 3.68 ± 0.04(3.69 ± 0.08) and 2Δ₂/kB Tc = 0.34 ± 0.02(0.31 ± 0.05)) in the compound. The magnetization measurements show a large reversible region in the mixed state, similar to the vortex liquid phase observed in high-Tc superconductors. The estimation of the Ginzburg number GI suggests that thermal fluctuations (though small) may play an important role in the unpinning of the vortices in this compound. Electronic band structure calculations along with heat capacity measurements suggest that the electronic correlations are not significant in Lu₂Os₄Ge₁₃. Band structure calculations show a very complex structure in the Fermi surface which might play a significant role in enhancing the multi-band effects in Lu₂Os₄Ge₁₃.

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