1. Introduction

Understanding the mechanism of unconventional superconductivity, where the structure lacks an inversion symmetry has been a tough challenge ever since the discovery of the heavy fermion noncentrosymmetric (NCS) superconductor CePt₃Si [1, 2]. The lack of an inversion center, in the crystal structure, of an NCS superconductor makes parity an unconserved quantity. Therefore, the superconducting ground state of an NCS superconductor may exhibit a mixing of spin-singlet and spin-triplet pair states [3–11]. A parity mixed superconducting ground state can give rise to several anomalous superconducting properties, e.g. upper critical field exceeding the Pauli limit, nodes in the superconducting gap, a helical vortex state, and time-reversal symmetry breaking (TRSB).

Several NCS superconducting systems have been investigated to study the effects of broken inversion symmetry [12–28], but majority of them appear to show s-wave superconductivity. Theoretical predictions suggest that NCS superconductors are prime candidates to exhibit TRSB due to its admixed superconducting ground states. To date only a few NCS superconductors Re₆Zr [29], LaNiC₂ [30], SrPtAs [31] and La₇Ir₃ [32] have been reported to show TRSB. It is a rarely observed phenomena and apart from NCS superconductors, it has only been observed in a few unconventional superconductors e.g. Sr₂RuO₄ [33, 34], UPt₃ [35, 36], PrPt₄Ge₁₂ [37], LaNiGa₂ [38], and Lu₅Rh₀S₁₈ [39]. The discrepancy between theory, experiment and the possibility of realizing an unconventional superconducting state having TRSB in NCS superconductors are of great interest. To understand the superconducting mechanism, it is necessary to investigate new NCS superconducting systems by combining bulk measurements such as transport, magnetization, specific heat, etc and local probe techniques like muon spectroscopy. Muon spectroscopy
is one of the direct methods of detecting the unconventional superconducting ground state. This technique can accurately determine the temperature dependence of the magnetic penetration depth and the onset of TRSB in superconductors.

Here we report the properties of the superconducting state of a binary NCS compound (α-Mn structure) Nb0.5Os0.5, which has a superconducting transition at \( T_c = 3.07 \) K. Resistivity, magnetization, and specific heat measurements were carried out to explore the superconducting properties of Nb0.5Os0.5. \( \mu \)SR measurements in transverse-field (TF) and zero-field (ZF) were used to probe the flux line lattice and TRSB respectively.

2. Experimental details

The polycrystalline sample of Nb0.5Os0.5 was prepared by standard arc melting technique. The stoichiometric amounts of Nb (99.95%, Alfa Aesar) and Os (99.95%, Alfa Aesar) were placed on the water cooled copper hearth in an ultrapure argon gas atmosphere. The sample was inverted and remelted several times to ensure the sample homogeneity. There was a negligible weight loss during the sample synthesis. The sample was sealed inside an evacuated quartz tube and annealed at 1000 °C for 15 d in order to remove any thermal stresses if present in the sample. The phase analysis for Nb0.5Os0.5 was done using x-ray diffraction (XRD) at room temperature on an X′pert PANalytical diffractometer. The magnetization and ac susceptibility measurements were performed using a superconducting quantum interference device (MPMS 3, Quantum Design Inc.). The electrical resistivity and specific heat measurements were done using the physical property measurement system (PPMS, Quantum Design Inc.). The \( \mu \)SR measurements were carried out using the MuSR spectrometer at the ISIS facility, Rutherford Appleton Laboratory, Didcot, U.K. in both longitudinal and transverse geometries.

3. Results and discussion

3.1. Sample characterization

The powder x-ray diffraction pattern for Nb0.5Os0.5 was collected at room temperature. Rietveld refinement was performed using the High Score Plus Software. As observed from figure 1, the Nb0.5Os0.5 sample has no impurity phase. It can be indexed by cubic, noncentrosymmetric α-Mn structure (space group \( I4_3m \), No. 217) with the lattice cell parameter \( a = 9.765(3) \) Å. Also, the energy dispersive x-ray spectroscopy confirmed above-mentioned composition and showed that there are no other phases present in the sample to within the limits of these analysis techniques.

3.2. Normal and superconducting state properties

3.2.1. Electrical resistivity. The electrical resistivity measurement was done by the ac transport technique in the temperature range of 1.85 K \( \leq T \leq 300 \) K in zero field as displayed in figure 2. The superconducting transition temperature was observed around \( T_c = 3.1 \) K (see inset: (a)). The normal state resistivity remains almost temperature independent up to the highest measured temperature, indicating that Nb0.5Os0.5 exhibit poor metallicity. Similar behavior was found in other compounds with the same structure and space group (\( I4_3m \)) \cite{12–15, 40}. The resistivity measurements as a function of temperature were also done under different applied magnetic fields up to 3 T in a temperature range of 1.9 K to 12 K as shown in inset: (b) of figure 2. The superconducting transition temperature moves to the lower temperature with the increase in magnetic field, with the transition width becoming broader.

3.2.2. Magnetization. The magnetization measurement was done in zero-field cooling (ZFC) and field-cooled cooling (FCC) mode in an applied field of 1 mT. The strong diamagnetic signal due to the superconducting transition appears around \( T_{\text{onset}} = 3.07 \) K, as shown in figure 3(a). The value of \( 4\pi\chi \) at 1.8 K exceeds –1 due to the demagnetization effect. The magnetization measurement was done as a function of field at various temperatures to calculate the temperature dependence of the lower critical field \( H_{c1}(T) \). Lower critical field is defined as the first deviation from linearity in low-field regions in \( M \) versus \( H \) curves (see inset figure 3(b)). The measurements were done up to a field range of 8 mT, at different temperatures from 1.9 K to 3 K. The main panel of figure 3(b) shows the \( H_{c1}(T) \) plot, where the lower critical field decreases with the increase in temperature. The temperature variation of \( H_{c1}(T) \) can be described by the Ginzburg–Landau (GL) formula

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

When fitted with the experimental data it yields \( H_{c1}(0) = 3.06 \pm 0.05 \) mT.

The temperature dependence of the upper critical field \( H_{c2}(T) \) was obtained by measuring the field dependence of superconducting transition \( T_c \) in magnetization, ac susceptibility, resistivity, and specific heat measurements. It is evident...
from figure 4 that $H_c2$ vary linearly with the temperature and can possibly be best fitted by Ginzburg–Landau relation given by

$$H_{c2}(T) = H_{c2}(0)\left(1 - \frac{T}{T_c}\right).$$  

Figure 2. (a) The resistivity measurement $\rho(T)$ for Nb$_{0.5}$Os$_{0.5}$ taken in zero field in a temperature range of 1.85 K $\leq T \leq 300$ K showing poor metallicity. Inset: (a) the superconducting transition temperature is $T_c = 3.1$ K (b) $\rho(T)$ measurements done at several applied magnetic fields up to 3 T.

Figure 3. (a) The magnetization data for Nb$_{0.5}$Os$_{0.5}$ taken in 1 mT field shows the superconducting transition at $T_c = 3.07$ K. (b) The lower critical field $H_{c1}$ was estimated to be 3.06 ± 0.05 mT. Inset: the magnetization curves as a function of applied magnetic field at various temperatures.

Figure 4. The upper critical field $H_{c2}(T)$ obtained from magnetization, ac susceptibility, resistivity, and specific heat measurements. The dotted lines show the GL fits, yielding $H_{c2}(0) = 5.4 \pm 0.1$ T for Nb$_{0.5}$Os$_{0.5}$.

where $t = T/T_c$. By fitting above equation in the $H_{c2}$-T graph, the specific heat and magnetization measurements give $H_{c2}(0) = 5.4 \pm 0.1$ T, whereas resistivity and ac susceptibility measurements give $H_{c2}(0) = 4.6 \pm 0.1$ T. $H_{c2}(0)$ can be used to estimate the Ginzburg Landau coherence length $\xi_{GL}$ from the relation [41]

$$H_{c2}(0) = \frac{\Phi_0}{2\pi\xi_{GL}^2}.$$  

where $\Phi_0$ is the quantum flux ($\hbar/2e$). For $H_{c2}(0) \approx 5.4 \pm 0.1$ T, we obtained $\xi_{GL}(0) = 78.12 \pm 0.72$ Å. For a type-II BCS superconductor in the dirty limit, the orbital limit of the upper critical field $H_{c2}^{orbital}(0)$ is given by the Werthamer–Helfand–Hohenberg expression [42, 43]

$$H_{c2}^{orbital}(0) = -0.693T_c\frac{dH_{c2}(T)}{dT}_{T=T_c}.$$  

Using initial slope 2.10 ± 0.02 T K$^{-1}$ from the $H_{c2}$-T phase diagram, $H_{c2}^{orbital}(0)$ in the dirty limit was estimated to be 4.46 ± 0.04 T. Within the $\alpha$-model the Pauli limiting field is given by [44]

$$H_{c2}^{\alpha}(0) = 1.86T_c\left(\frac{\alpha}{\alpha_{BCS}}\right).$$  

Using $\alpha = 1.81$ (from the specific heat measurement), it yields $H_{c2}^{\alpha}(0) = 5.86$ T. The upper critical field $H_{c2}(0)$ calculated above is close to both the orbital limiting field and Pauli limiting field. Detailed investigations of the upper critical field in high quality single crystals of Nb$_{0.5}$Os$_{0.5}$ at low temperatures is highly desirable due to the possibility of enhanced upper critical field due to grain boundaries in polycrystalline samples. The Ginzburg Landau penetration depth $\lambda_{GL}(0)$ can be obtained from the $H_{c1}(0)$ and $\xi_{GL}(0)$ using the relation [41]

$$H_{c1}(0) = \frac{\Phi_0}{4\pi\lambda_{GL}^2(0)}\left(\ln\frac{\lambda_{GL}(0)}{\xi_{GL}(0)} + 0.12\right).$$  

Using $H_{c1}(0) = 3.06$ mT and $\xi_{GL}(0) = 78.12$ Å, we obtained $\lambda_{GL}(0) \approx 4774 \pm 50$ Å. The Ginzburg Landau parameter is given by the relation [41]
\[ \kappa_{GL} = \frac{\lambda_{GL} (0)}{\xi_{GL} (0)} \]  

(7)

For \( \xi_{GL} (0) = 78.12 \pm 0.72 \text{ Å} \) and \( \lambda_{GL} (0) = 4774 \pm 50 \text{ Å} \), it yields \( \kappa_{GL} = 61.11 \pm 0.97 \). This value of \( \kappa_{GL} \gg \frac{1}{\sqrt{T}} \), indicating that Nb0.5Os0.5 is a strong type-II superconductor. Thermodynamic critical field \( H_c \) can be estimated from \( \kappa_{GL} (0) \) and \( H_{c2} (0) \) using the relation

\[ H_c = \frac{H_{c2}}{\sqrt{2} \kappa_{GL}}. \]  

(8)

which for \( H_{c2} = 5.4 \pm 0.1 \text{ T} \) and \( \kappa_{GL} = 61.11 \pm 0.07 \) yields \( H_c = 62.4 \pm 0.1 \text{ mT} \).

3.2.3. Specific heat. The low temperature specific heat measurement \( C(T) \) was taken in zero applied field. Figure 5(a) shows the \( C/T \) versus \( T^2 \) data in the temperature range \( 3 \text{ K} \leq T^2 \leq 100 \text{ K} \). The discontinuity in the specific heat data shows the onset of superconducting transition below \( T_c = 3.0 \text{ K} \), confirming the bulk superconductivity in Nb0.5Os0.5. The temperature dependence of the specific heat was done under different applied fields, where the increase of applied field suppresses the superconductivity, manifested by the shifting of transition temperature to lower temperature with the reduction in the specific heat jump as shown in the inset of figure 5(a). The normal state low temperature specific heat data above \( T_c \) can be fitted with the equation given below, where the extrapolation to the limit \( T \to 0 \) extracts the electronic and phonon contribution to the specific heat

\[ \frac{C}{T} = \gamma_n + \beta_3 T^2 + \beta_5 T^4. \]  

(9)

Here \( \gamma_n \) is the normal state Sommerfeld coefficient related to the electronic contribution to the specific heat whereas \( \beta_3 \) and \( \beta_5 \) are the coefficients related to the lattice contribution to the specific heat. The solid red line in the main panel of figure 5(a) shows the best fit to the data which yields \( \gamma_n = 3.42 \pm 0.01 \text{ mJ mol}^{-1} \text{ K}^{-2} \), \( \beta_3 = 0.039 \pm 0.002 \text{ mJ mol}^{-1} \text{ K}^{-4} \), and \( \beta_5 = 0.205 \pm 0.004 \text{ mJ mol}^{-1} \text{ K}^{-6} \). The Debye temperature \( \theta_D \) can be calculated from \( \beta_3 \) coefficient through formula

\[ \theta_D = \left( \frac{12 \pi^2 R N}{5 \beta_3} \right)^{\frac{1}{3}}, \]  

(10)

where \( R \) is the molar gas constant (=8.314 J mol\(^{-1}\) K\(^{-1}\)), gives \( \theta_D = 367 \text{ K} \). The Sommerfeld coefficient is proportional to the density of states \( D(E_F) \) at the Fermi level given by

\[ \gamma_n = \left( \frac{\pi^2 k_B^2}{3} \right) D_c(E_F), \]  

(11)

where \( k_B \approx 1.38 \times 10^{-23} \text{ J K}^{-1} \). Using \( \gamma_n = 3.42 \pm 0.01 \text{ mJ mol}^{-1} \text{ K}^{-2} \), we obtained \( D_c(E_F) = 1.45 \text{ states/eV f.u.} \).

The electron–phonon coupling constant can be calculated using the McMillan equation [45]

\[ \lambda_{e-ph} = \frac{1.04 + \mu^* \ln(\theta_D / 1.45 T_c)}{(1 - 0.62 \mu^*) \ln(\theta_D / 1.45 T_c)} - 1.04. \]  

(12)

The magnitude of the specific heat jump for Nb0.5Os0.5 in \( \Delta C/T_c \) data is 5.06 mJ mol\(^{-1}\) K\(^{-2}\). The normalized specific heat jump \( \frac{\Delta C}{\gamma_n T_c} \) is 1.48 for \( \gamma_n = 3.42 \text{ mJ mol}^{-1} \text{ K}^{-2} \), which is close to the value for a BCS superconductor (=1.43) in the weak coupling limit. The temperature dependence of the normalized entropy \( S \) in the superconducting state for a single-gap BCS superconductor is given by

\[ \frac{S}{\gamma_n T_c} = -\frac{6}{\pi^2} \left( \frac{\Delta (0)}{k_B T_c} \right) \left[ \ln(f) + (1 - f) \ln(1 - f) \right] \text{d}y. \]  

(14)
where \( f(\xi) = [\exp(E(\xi)/k_B T) + 1]^{-1} \) is the Fermi function, \( E(\xi) = \frac{\xi^2}{2} + \Delta^2(t) \), where \( \xi \) is the energy of normal electrons measured relative to the Fermi energy, \( y = \xi/\Delta(0) \), \( t = T/T_c \), and \( \Delta(t) = \tanh[1.82(1.018/(1/t) - 1)]^{0.5} \) is the BCS approximation for the temperature dependence of the energy gap. The normalized electronic specific heat is then calculated from the normalized entropy by

\[
\frac{C_{el}}{\gamma_n T_c} = \frac{dS/\gamma_n T_c}{dT}.
\]

The \( C_{el} \) below \( T_c \) is described by equation (15) whereas above \( T_c \) its equal to \( \gamma_n T_c \).

In the single-band \( \alpha \) model, for systems where \( \Delta C_{el}/\gamma_n T_c \neq 1.43 \), the BCS parameter \( \sigma_{BCS} \) is replaced by \( \alpha \) which can be used as an adjustable parameter to fit the experimental superconducting specific heat data [44]. This parameter can be calculated using the equation

\[
\frac{\Delta C_{el}}{\gamma_n T_c} = 1.426 \left( \frac{\alpha}{\sigma_{BCS}} \right)^2.
\]

Substituting the value of normalized specific heat jump \( \Delta C_{el}/\gamma_n T_c = 1.48 \) for our sample, we get \( \alpha = 1.8 \). The temperature dependence of single band \( \alpha \)-model superconducting-state heat capacity \( C_{el}(T) \) calculated for \( \alpha = 1.81 \) is shown by the solid blue curve in figure 5(b) together with that of the BCS prediction for \( \alpha = \sigma_{BCS} \). It can be clearly seen that the curve for \( \alpha = \sigma_{BCS} \) deviates around the temperature region near \( T_c \), whereas it was much more improved for \( \alpha = 1.81 \). All the superconducting and normal state parameters are summarized in table 1.

3.2.4. Muon spin relaxation and rotation. The superconducting ground state of Nb0.5Os0.5 was further analyzed by \( \mu \)SR relaxation and rotation measurements. The zero-field muon spin relaxation (ZF-\( \mu \)SR) spectra was collected below (3.5 K) and above (3.5 K) the transition temperature. The solid lines are the fits to Gaussian KT function given in equation (18).

The conclusion that the time-reversal symmetry is preserved in Nb0.5Os0.5 within the detection limit of \( \mu \)SR.

Transverse-field muon spin rotation (TF-\( \mu \)SR) measurements were done to gain information on the superconducting gap structure of Nb0.5Os0.5. Asymmetry spectra was recorded above (3.5 K) and below (0.1 K) the transition temperature \( T_c \) in a transverse field of 30 mT as shown in figure 7. The TF-\( \mu \)SR precession signal were fitted using an oscillatory decaying Gaussian function

\[
G_{TF}(t) = A_1 \exp \left( -\frac{\sigma_2 \omega^2 t^2}{2} \right) \cos(w_1 t + \phi) + A_2 \cos(w_2 t + \phi),
\]

where \( w_1 \) and \( w_2 \) are the frequencies of the muon precession signal and background signal respectively, \( \phi \) is the initial phase offset and \( \sigma \) is the Gaussian muon-spin relaxation rate. Figure 7(a) shows the signal in the normal state where depolarization rate is small, attributed to homogeneous field distribution throughout the sample. The significant depolarization rate in the superconducting state shown in the figure 7(b) is due to the flux line lattice (FLL) in the mixed state of the superconductor, which gives rise to the inhomogeneous field distribution. The depolarization arising due to the static fields from the nuclear moments \( \sigma_N \) is assumed to be temperature independent and adds in quadrature to the contribution from the field variation across the flux line lattice \( \sigma_{FLL} \):

\[
\sigma^2 = \sigma_N^2 + \sigma_{FLL}^2.
\]

The muon-spin rotation rate in the superconducting state \( \sigma_{FLL} \) is related to the London magnetic penetration depth \( \lambda \) and thus to the superfluid density \( n_s \) by the equation

\[
\frac{\sigma_{FLL}(T)}{\sigma_{FLL}(0)} = \frac{\lambda^{-2}(T)}{\lambda^{-2}(0)},
\]

For an \( s \)-wave BCS superconductor in the dirty limit, the temperature dependence of the London magnetic penetration depth is given by

\[
\lambda^{-2}(T) = \frac{\lambda^{-2}(0)}{\tanh \left( \frac{\Delta(T)}{2k_B T} \right)}.
\]
Table 1. Normal and superconducting properties of Nb0.5Os0.5.

| Parameter   | Unit | Value  |
|-------------|------|--------|
| $T_c$       | K    | 3.07   |
| $H_{c1}(0)$ | mT   | 3.06   |
| $H_{c2}(0)$ | T    | 5.4    |
| $H_c(0)$    | mT   | 62.4   |
| $H_c^{(0)}$ | T    | 4.46   |
| $\xi(0)$   | A    | 78.12  |
| $\lambda(0)$ | A | 4774   |
| $\kappa(0)$ |       | 61     |
| $\gamma(0)$ | mJ mol$^{-1}$ | 3.42  |
| $\beta(0)$  | mJ mol$^{-1}$ | 0.039  |
| $\theta_D$  | K    | 367    |
| $\lambda_{\text{GL}}$ | states/ev f.u | 1.45  |
| $D_{\text{GL}}$ |       | 1.48   |
| $\Delta(0)/\gamma T_c$ |       | 1.81   |

where $\Delta(T) = \Delta_0 \delta(T/T_c)$. The temperature dependence of the gap in the BCS approximation is given by the expression $\delta(T/T_c) = \tanh[1.82(1.018((T_c/T) - 1))]^{0.51}$. By combining equations (20)–(22), a model was obtained for a dirty limit single-gap s-wave superconductor, where $\sigma(T)$ above $T_c$ is equal to $\sigma_N$ and below $T_c$ is given by equation (23) which contain contributions from both $\sigma_N$ and $\sigma_{\text{FLL}}$.

$$
\sigma(T) = \sqrt{\sigma_{\text{FLL}}^2(0) \frac{\Delta^2(T)}{\Delta^2(0)} \tanh^2 \left( \frac{\Delta(T)}{2k_B T} \right) + \sigma_N^2}. \quad (23)
$$

The temperature dependence of muon depolarization rate $\sigma$ was collected in an applied field of 30 mT as shown in Figure 8. The depolarization rate $\sigma$ remains temperature independent up to $T_c$ attributing to random nuclear magnetic moments, then after $T_c$, $\sigma$ increases due to the formation of well-ordered FLL. The best fit to the $\sigma(T)$ data were obtained with the single-gap BCS model (equation (23)) shown by the solid blue line in Figure 8, where we have obtained $\sigma_N = 0.366 \pm 0.002 \, \mu s^{-1}$, $\sigma(0) = 0.444 \pm 0.001 \, \mu s^{-1}$, and $\Delta(0) = 0.50 \pm 0.02 \, \text{meV}$. The value of $\alpha = \Delta(0)/k_B T_c = 1.89$ is close to the value ($\alpha = 1.81$) obtained from the low temperature specific heat measurement. Thus, the TF-$\mu$SR measurements together with the specific heat measurement confirm that Nb0.5Os0.5 is a s-wave superconductor.

The GL penetration depth $\lambda_{\mu}(0)$ at $T = 0$ K can be directly calculated from $\sigma_{\text{FLL}}(0) = 0.251 \pm 0.001 \, \mu s^{-1}$ from muon measurements by the relation [49, 50] and $\Phi_0$ is the magnetic flux quantum. The value of GL penetration depth $\lambda_{\mu}(0)$ is 6538 ± 13 Å. The estimated value is higher than the $\lambda_{\text{GL}}(0)$ calculated earlier, which could be due to the dirty limit superconductivity in Nb0.5Os0.5.

Uemura et al showed in 1991 that the superconductors can be classified into a conventional/unconventional superconductor [51, 52] based on the ratio of the transition temperature ($T_c$) to the Fermi temperature ($T_F$). It was shown that the unconventional, exotic superconductors fall in the range of $0.01 \leq T_c/T_F \leq 0.1$. The Fermi temperature can be calculated using the relation

$$
\frac{\sigma_{\text{FLL}}^2(0)}{\gamma_{\mu}} = 0.00371 \frac{\Phi_0^2}{\lambda_{\mu}^2(0)}. \quad (24)
$$

where $\gamma_{\mu}/2\pi = 135.53 \, \text{MHz T}^{-1}$ is the muon gyromagnetic ratio and $\Phi_0$ is the magnetic flux quantum.
The sample of Nb$_{0.5}$Os$_{0.5}$ was prepared by standard arc-melting technique. XRD analysis confirms the phase purity and cubic, NCS α-Mn structure (space group no.217) with lattice cell parameter $a = 9.765(3)$ Å. The transport, magnetization, and specific heat measurements confirm type-II, s-wave superconductivity in Nb$_{0.5}$Os$_{0.5}$, having transition temperature $T_c = 3.07$ K. The upper and lower critical fields estimated to be $H_{c1} = 3.06$ mT and $H_{c2} = 5.4$ T respectively. The TF-μSR measurements further confirm s-wave superconductivity. The ZF-μSR measurements confirm that time-reversal symmetry is preserved in Nb$_{0.5}$Os$_{0.5}$ within the detection limit of μSR. In order to understand the superconducting ground state of NCS compounds, it is clearly important to search for new NCS superconductors.

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