Study of Nb$_{0.18}$Re$_{0.82}$ non-centrosymmetric superconductor in the normal and superconducting states

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Abstract

We examine the evidence for multiband superconductivity and non s-wave pairing in the non-centrosymmetric superconductor (NCS) Nb$_{0.18}$Re$_{0.82}$, using electrical transport, magnetization and specific heat measurements. In the normal state, the evolution of electrical resistivity with temperature and magnetic field support the phonon-assisted interband scattering and multiband picture. In the superconducting state, the temperature dependence of the upper critical field, $H_{c2}(T)$, is found linear and cannot be described within the Werthamer, Helfand and Hohenberg model over the whole temperature range measured. In addition, the observed $H_{c2}(0)$ exceeds the Pauli limit, suggesting non-s-wave pairing. Interestingly, the Kadowaki–Woods ratio and Uemura plot reveal a behavior in Nb$_{0.18}$Re$_{0.82}$ which is similar to that found in unconventional superconductors. The normalized superfluid density ($\rho_s$), estimated using the temperature dependence of the lower critical field, $H_{c1}(T)$, is well explained with the help of the multiband description. Phase fluctuation analysis conducted on the reversible magnetization data, reveals a significant deviation from the mean-field conventional s-wave behavior. This trend is interpreted in terms of a non s-wave spin-triplet component in the pairing symmetry as might be anticipated in a NCS where anti-symmetric spin–orbit coupling plays a dominant role. Recently, time reversal symmetry breaking observed in Nb$_{0.18}$Re$_{0.82}$ supports this picture.

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(Some figures may appear in colour only in the online journal)

1. Introduction

The inversion and the time reversal symmetry of the ground state of the superconducting wave function defines the pairing state of the Cooper pairs, which may be categorized in terms of even-parity spin-singlet or odd-parity spin-triplet pairing [1]. The lack of either one of these symmetries in a system gives rise to unconventional superconductivity. The lack of inversion symmetry is controlled by the strength of anti-symmetric spin–orbit coupling (ASOC), which leads to the spin splitting of the electronic states at the Fermi level and
may give rise to a mixture of spin-singlet and spin-triplet pairing states in the superconducting wave function [1, 2]. In addition, non-centrosymmetric superconductors (NCS) can also exhibit a variety of other unconventional properties, such as, magneto-electrical effects, multigap behavior, helical vortex-state, nodes in the superconducting gap, non-trivial topological effects and time reversal symmetry breaking (TRSBR) [3]. These copious exotic behaviors in NCS are topical to investigate fundamentally and could even underpin the development of new technological device concepts, such as, those based on Majorana fermions [1–4].

The discovery of superconductivity in a heavy fermion non-centrosymmetric compound, CePt3Si, ignited the rapid growth in the field of NCS [3, 5–7]. Most of the heavy fermion NCS show superconductivity under finite applied pressure (except CePt3Si) and the effect of strong electron correlations and spin fluctuations are responsible for the observation of unconventional superconductivity in these systems [3, 8]. Interestingly, due to the absence of magnetic correlations, the weakly-correlated electron systems, such as, Nb–Re [9], LaNiC2 [10], Li2(Pd, Pt), B [11, 12], (Rh, Ir)Ga9 [13] and Mg1014Ir916B16, [14], are particularly interesting because they better enable the isolation of the impact of ASOC in NCS. Most of the weakly-correlated electron systems appear to show s-wave behavior, although, in some cases, they also show nodal superconducting gap structure [2, 3, 15]. It has also been observed that even in the case of fully gaped s-wave behavior, µ-SR measurements found TRSB in La3Ir5, LaNiC2 [16, 17].

Nb0.18Re0.82 is a phonon-mediated NCS having superconducting transition temperature, Tc, in the vicinity of 9 K [9]. Literature suggests that electronic correlations are not strong and magnetic correlations are also absent, which makes this system suitable to investigate the role of ASOC [9, 18]. It is also observed that the upper critical field, Hc2, in Nb0.18Re0.82 superconductor reaches the Pauli limit, which suggests the possibility of unconventional pairing [9, 18]. A recent study of the specific heat and Andreev point contact spectroscopy on a single crystal of Nb0.18Re0.82 concluded that the system is a nodeless two-gap superconductor [19]. However, it has been argued that there are a variety of different scenarios based on mixed spin singlet-triplet pairing that may also explain these observations [19]. Very recently, TRSB has been observed in Nb0.18Re0.82 [20], which is indicative of unconventional superconductivity in this system. Various superconducting properties in Nb0.18Re0.82, such as, upper critical field [9, 18], energy gap behavior [19] and TRSB [20], are not well described within the conventional Bardeen–Cooper–Schrieffer (BCS) theory. Therefore, further investigation in the normal and superconducting states is required to explore the unconventional nature of Nb0.18Re0.82 in terms of pairing symmetry and physical properties.

In the present study, a detailed characterization of a polycrystalline Nb0.18Re0.82 superconductor is performed in the normal and superconducting states. Electrical resistivity measurements in the normal state provides evidence of phonon-assisted interband scattering. The two-gap picture is confirmed using electrical transport and magnetization results [19]. We find that the Kadowaki–Woods (KW) ratio and Uemura plot suggest unconventional behavior, which is inconsistent with the previous claims [18], and in agreement with the recently observed TRSB that supports the unconventional nature in Nb0.18Re0.82. The temperature dependence of upper critical field, Hc2(T), is not explained using the conventional Werthamer, Helfand and Hohenberg (WHH) model and the observed Hc2(0) exceeds the Pauli limit, indicating the unconventional pairing. The phase analysis we have performed using reversible magnetization data does not agree with a mean-field s-wave picture.

2. Experimental details

A polycrystalline sample of Nb0.18Re0.82 was prepared by melting the constituent elements (99.8% purity of Nb and 99.99% purity of Re, Alfa Aesar) in an arc furnace with a constant supply of 99.999% pure argon atmosphere. The sample was flipped and re-melted six times to ensure homogeneity. A mass loss of about 1% was observed after melting. In addition, the as-cast sample was wrapped in Ta foil and sealed in a quartz ampoule with argon atmosphere for annealing. The annealing was performed at 800 °C for 7 days, followed by slow cooling, down to the room temperature. The sample homogeneity was confirmed using energy dispersive x-ray spectroscopy. The metallographic characterization was performed using a high power optical microscope. The x-ray diffraction (XRD) of the sample was performed using a standard diffractometer (Model D2 Phaser, Bruker) using Cu–Kα(λ = 1.5406 Å) radiation in the 2θ range from 20°–80° with Δ2θ = 0.02°.

Electrical transport, specific heat, and magnetic susceptibility measurements were performed at low temperatures (down to 2 K) and high magnetic fields (up to 9 T), using a physical properties measurement system (Quantum Design, USA). The temperature and magnetic field dependence of electrical resistivity, ρ(H, T), was measured on a rectangular parallelepiped shaped sample, using a standard four probe technique. The electrical contacts were made using thin gold wires and silver paint. The contact resistance was observed to be R ≈ 0.3 Ω. A widely used thermal relaxation technique was exploited to measure the temperature and magnetic field dependence of specific heat, C(H, T), in the superconducting as well as in the normal state. The sample was also characterized using ac-susceptibility measurements performed with 0.05 mT excitation magnetic field and 500 Hz frequency. DC magnetization measurements were performed using a SQUID-vibrating sample magnetometer (VSM) (Quantum Design, USA), where a small sample was mounted within a quartz sample holder.

3. Results and discussion

Figure 1(a) shows the x-ray powder diffraction pattern of the polycrystalline Nb0.18Re0.82 Sample. A silicon thin film was used as a substrate, giving a well defined XRD peak at 68.8 2θ value. The silicon XRD peak was excluded from the
Rietveld refinement analysis and shown as the shaded portion in the XRD spectra. The experimental data is well explained using the $\alpha$-Mn type cubic crystal structure. The lattice parameter estimated using the Rietveld refinement analysis is 9.653 Å, which is consistent with the literature [9, 18]. No secondary phase peaks are observed in the XRD spectra, consistent with the inspection by optical metallography shown in figure 1(b). A schematic picture of the $\alpha$-Mn type cubic crystal structure is shown in figure 1(c), where the distribution of the Nb and Re atoms at different atomic positions are shown with different colors (see the caption of 1(c)).

The superconducting transition temperature is measured using various experimental techniques. In figure 2(a), the temperature dependence of electrical resistivity, $\rho(T)$, at zero applied magnetic field shows a sharp superconducting transition with $\Delta T_{c} = 0.06$ K. The inset of figure 2(a) shows the derivative of $\rho(T)$ and the peak position is taken as the $T_{c}$ of the sample. Figure 2(b) shows the bulk superconducting transition observed in the temperature dependence of specific heat, $C(T)$, measured in the absence of magnetic field. The $\Delta T_{c} = 0.27$ K, is estimated as the temperature difference between the transition onset and completion. The arrow indicates the $T_{c}$ of the sample. The real part of ac-susceptibility ($\chi'$) in figure 2(c) shows a sharp superconducting transition with $\Delta T_{c} = 0.15$ K, where the arrow indicates the $T_{c}$ of the sample. The nearly perfect shielding confirms the good quality of the sample. The imaginary part of the ac-susceptibility, $\chi''$, (inset of figure 2(c)) shows a sharp peak. The dc-susceptibility in the zero field cooled (zfc) and field cooled cooling (fcc) protocols measured in 1 mT magnetic field, is shown in figure 2(d). The nearly perfect shielding and ~12% of Meissner fraction is indicated by zfc and fcc curves respectively, and the arrow indicates the $T_{c}$ of the sample. All four measurements shown in figure 2 are consistent with each other and show $T_{c} = 8.63$ K ± 0.05 K. The $T_{c}$ of the sample observed in our measurements is in agreement with the literature within the error of 0.1 K [9, 18, 19].

Figure 3 shows the electrical resistivity in the normal state as a function of temperature and magnetic field. The residual resistivity, $\rho_{0}$ is observed to be 1.02 $\mu\Omega m$ and the residual resistivity ratio (RRR) is defined as, $\rho_{300K}/\rho_{10K} = 1.16$. A small value of RRR suggests a dominant role of disorder in the sample. The mean free path ($l$) estimated using the Drude’s theory is 2.12 Å and the Pippard’s coherence length obtained using the expression, $\xi_{0} = 1.781h\nu/(\pi^{2}k_{B}T_{c}) \approx 4364$ Å, implying that the sample is in the dirty limit ($l \ll \xi_{0}$).


Figure 2. (a) Temperature dependence of resistivity, $\rho(T)$, measured in zero magnetic field, showing a sharp superconducting transition. The inset shows the derivative, $d\rho/dT$. (b) Temperature dependence of the specific heat, $C(T)$, measured at $\mu_0 H = 0$. (c) Real part of temperature dependence of ac-susceptibility ($\chi'$) in zero applied magnetic field showing almost perfect shielding. The inset shows the imaginary part of ac-susceptibility ($\chi''$). (d) Temperature dependence of dc magnetization in 1 mT magnetic field, measured using the zero field cooled (zfc) and field cooled (fc) protocols. The zfc curve shows the nearly perfect shielding state and the arrow indicates the $T_c$ onset. The observed $T_c$ in all the measurements is of the order of 8.63 K ± 0.05 K.

Figure 3(a) shows that the zero field $\rho(T)$ at low temperature (14–50 K) follows a quadratic behavior, $\rho = \rho_0 + A T^2$. The solid line is a fit of this expression to the experimental data, which yields the coefficient of the $T^2$ term, $A = 0.000 \pm 0.000109 \pm 4.4 \times 10^{-8} \ \mu\Omega\text{m}$ $K^{-2}$. The small value of the coefficient $A$ indicates that the electronic correlations in the Nb$_{0.18}$Re$_{0.82}$ alloy are not strong [9, 18]. Figure 3(b) shows the zero field $\rho(T)$ where the room temperature value is 1.2 $\mu\Omega$ m and the trend is a metal-like behavior down to 10 K. The qualitative behavior of $\rho(T)$ in the whole temperature range is similar to the one observed in [9]. The experimental data is well explained using an empirical relation provided by Woodard and Cody [21], $\rho(T) = \rho_0 + \rho_1 T^n + \rho_2 \exp(-T_0/T)$, where $\rho_0$ is the residual resistivity, $T_0$ in the exponential term is defined as a characteristic temperature of a certain phonon mode. The exponential term has been explained in terms of phonon-assisted interband scattering or intraband Umklapp scattering, while the origin of the non-exponential term is not defined [22]. The empirical relation discussed above describes the data reasonably well over the whole temperature range and the estimated fitted parameters are $\rho_0 = 1.02 \ \mu\Omega$ m, $\rho_1 = 0.00025 \ \mu\Omega$ m/$K^n$, $n = 1$, $\rho_2 = 0.126 \ \mu\Omega$ m and $T_0 = 103.4$ K. Nb$_{0.18}$Re$_{0.82}$ is considered to be a multiband superconductor, and the exponential term in the resistivity which can be interpreted in terms of phonon-assisted interband scattering in the normal state, is certainly consistent with this picture. Similar behavior has been observed in the Mo–Re multiband superconductor [23, 24].

Figure 3(c) shows the magnetic field dependence of the transverse magnetoresistance, $MR = (\rho(H) - \rho_0)/\rho_0$, measured above $T_c$, at different temperatures ranging from 9 K–16 K. In low magnetic fields the MR follows a linear behavior, while in higher magnetic fields it saturates. It is also observed that the MR does not follow the conventional metallic quadratic magnetic field dependence ($MR \propto H^2$) [25]. As Nb$_{0.18}$Re$_{0.82}$ is considered to be a multiband superconductor [19], we explore whether the MR is explained using the following expression for a simple two-band model [25]

$$\frac{\Delta \rho}{\rho_0} = \frac{\mu_0 H^2}{\alpha + \beta \times (\mu_0 H)^2},$$

where $\Delta \rho$ is the magnetoresistance, $\rho_0$ is the residual resistivity, $\mu_0$ is the magnetic permeability, $H$ is the magnetic field, and $\alpha$ and $\beta$ are the fitted parameters.
Figure 4. (a) Temperature dependence of the resistivity, \( \rho(T) \), in different applied magnetic fields in the superconducting state. (b) The fluctuation conductivity \( (\Delta \sigma) \) as a function of \( (T - T_c/T_c) \) in the temperature range 8.7 K–20 K, shows the fitting of the experimental data using the function \( (\Delta \sigma_{GL} = Pr^{-\eta}) \) [26, 35]. The parameter \( \eta \approx 0.5 \), suggests the existence of three-dimensional fluctuations in the sample. The inset shows the rounding-off behavior of \( \rho(T) \) in zero field, from 8.7 to 20 K. (c) The temperature dependence of specific heat, \( C(T) \), in different applied magnetic fields. Inset (i) shows the \( C(T) \) measured in 9 T magnetic field, and fitted using the expression, \( C(T) = \gamma T + \beta T^3 + \delta T^5 \), where \( \gamma \) represents the electronic part and \( \beta, \delta \) are the phonon terms [18]. Inset (ii) shows the specific heat measured in the normal state in zero magnetic field which approaches the Dulong–Petit value near 300 K. (d) The coefficient of the \( T^3 \) term \( A \) in \( \rho(T) \) plotted as a function of the square of the coefficient of the electronic specific heat \( (\gamma^2) \) for comparing different classes of materials. This plot is known as the KW plot.

where \( \alpha \) and \( \beta \) are the fitting parameters related to the conductance and mobility of the charge carriers in the associated two bands. In figure 3(c), the solid lines represent the fit of equation (1) to the experimental data showing a comparatively better fit to the experimental data in higher magnetic fields. The dotted line in figure 3(c) shows the linear dependence of the MR in low magnetic fields. The deviation between the two-band model and the experimental data may imply the contribution of more than two bands in the transport mechanism.

Figure 4(a) shows the temperature dependence of the electrical resistivity, \( \rho(T) \), in the superconducting state, measured in the presence of different applied magnetic fields. It is observed that the magnetic field does not play a significant role in the transition broadening. A sharp transition occurs even in high magnetic fields (9 T), with \( \Delta T_c = 0.06 \) K. The superconducting transition shows a rounding behavior in zero field, as well as in higher magnetic fields. This behavior for \( \mu_0H = 0 \), is due to the effect of thermal fluctuations [26–28], which is generally characterized in terms of the Ginzburg number \( (G_i) \) [29]. For an isotropic 3D superconductor, the Ginzburg number \( (G_i) \) is obtained using the expression, \( G_i = 32\pi^4(k_BT_c/k_B\lambda_{GL}(0))/\Phi_0^2 \), where \( k_B \) is the Boltzmann constant, \( T_c \) is the superconducting transition temperature, \( \kappa \) is the Ginzburg–Landau (GL) parameter, \( \lambda_{GL}(0) \) is the GL penetration depth and \( \Phi_0 \) is the magnetic flux quantum [29]. In the case of Nb0.18Re0.82, \( G_i \sim 10^5 \) is obtained, which is significantly higher than the \( G_i \) value in the conventional low-\( T_c \) superconductors \( (G_i \sim 10^{10}) \) but much lower than the \( G_i \) value reported in the high-\( T_c \) cuprates \( (G_i \sim 10^{-2} - 10^{-3}) \) [30]. The same order of magnitude of \( G_i \) is also observed for MgB2 [31], Ti–V [26], RNNi2B2C \( (R = Y, Lu) \) [32], and iron-pnictides \( (G_i \sim 10^{-3}) \) [33] superconductors, where fluctuation conductivity is well studied. Fluctuation conductivity analysis can provide a measure of the dimensionality of the fluctuations in a superconductor [34]. Figure 4(b) shows the fluctuation conductivity \( (\Delta \sigma = \sigma_{exp} - \sigma_e) \) as a function of \( (T - T_c)/T_c \) in the temperature range from 8.7 K–20 K. The normal state conductivity \( (\sigma_n) \) was estimated by extrapolating the conductivity from above \( 3T_c \). The inset of figure 4(b), clearly shows the rounding-off behavior for the zero field \( \rho(T) \) data below 20 K. The observed experimental
fluctuation conductivity ($\Delta\sigma$) can be explained in terms of the Aslamazov–Larkin model, $\Delta\sigma_{ML} = P\gamma^2$, where $T = (T - T_c)/T_c$, $P$ is a constant and $\gamma = 2 - D/2$ is a critical exponent, where $D$ defines the dimensionality of superconducting fluctuations [26, 35]. In figure 4(b), the straight line fits to the experimental data ($\Delta\sigma$) in the temperature range 8.7 K–9.5 K, and gives $\gamma \approx 0.5$, which suggests the 3D character of the superconducting fluctuations.

The temperature dependence of the specific heat, $C(T)$, in different magnetic fields is shown in figure 4(c). In the inset (i), showing $C(T)$ measured in 9 T magnetic field, the solid line is a fit to the expression $C(T) = \gamma T + \beta T^3 + \delta T^5$, where $\gamma$ is the coefficient of electronic specific heat and $\beta, \delta$ are the phonon terms [18, 36]. The fitting parameters are $\gamma = 5.60 \pm 0.04$ mJ m$^{-1}$ K$^{-2}$, $\beta = 0.045 05 \pm 0.001 24$ mJ m$^{-1}$ K$^{-4}$ and $\delta = 2.50 \times 10^{-4} \pm 8.52 \times 10^{-6}$ mJ m$^{-1}$ K$^{-6}$, which are consistent with the values obtained in the literature [18, 19]. The $\gamma$ value shows that the electronic contributions in the sample are not strong. On the other hand, the parameter $\beta$ is used to obtain the characteristic Debye temperature ($\Theta_D$), $\beta = N(12/5)\pi^2 R\Theta_D^3$, where $N$ is the number of atoms in a unit cell and $R$ is the gas constant. For $N = 1$, the $\Theta_D$ is 350 K and it is observed that the specific heat approaches the classical Dulong–Petit value near room temperature ($T = 300$ K), which suggests that the higher vibrational energy modes are populated near room temperature. The specific heat jump, $\Delta C/\gamma T_c = 1.64$ is larger than the weak coupling BCS limit which suggests a moderate coupling in the superconductor [37]. The electron–phonon coupling constant ($\lambda_{ep}$) is estimated using the McMillan expression [38], $\lambda_{ep} = \frac{1.04 + \mu^*/(\ln \Theta_D/1.45T_c)}{(1 - 0.62\mu^*/(1.45T_c))\ln \Theta_D/1.45T_c - 1.04}$. In this expression, $\mu^*$ is the Coulomb pseudopotential, which takes into account the direct Coulomb repulsion between electrons, $\Theta_D$ is the Debye temperature and $T_c$ is the superconducting transition temperature. For transition metals, $\mu^* = 0.13$ and taking $\Theta_D = 350$ K and $T_c = 8.63$ K, the $\lambda_{ep}$ is estimated as 0.73, which is consistent with the intermediate coupling limit seen in [18] for Nb–Re superconductors. The zero field $C(T)$ below $T_c$ is better explained in terms of a two-band model (see the supplementary information available online at stacks.iop.org/SUST/32/055003/mmedia), consistent with the single crystal study [19]. A power law behavior in $C(T)$ below $T_c$ would indicate a non s-wave behavior. However, we do not see power law behavior in our data (see supplementary information), nevertheless to draw a conclusion about the symmetry of the gap from this measurement alone is rather tenuous and possibly other techniques, such as, magnetic resonance probes might be needed to determine the spin symmetry of the superconducting pairing unambiguously [19]. The quadratic dependence of $\rho(T)$ at low temperatures indicates the dominance of electronic correlations over electron–phonon scattering. In figure 4(d), the coefficient of the $T^2$ term, $A$, in zero field $\rho(T)$ is plotted against the square of the coefficient of electronic specific heat ($\gamma^2$) for Nb$_{0.18}$Re$_{0.82}$ and compared with various other classes of materials. This is widely known as the KW plot and provides a unified picture of electronic correlations in the strongly correlated systems [39]. It is clearly seen in figure 4(c), that Nb$_{0.18}$Re$_{0.82}$ follows a similar trend as exhibited by the heavy fermion compounds in the KW plot and suggests the possibility of spin fluctuations in Nb$_{0.18}$Re$_{0.82}$ [40]. Similar behavior is also observed in the case of the moderately coupled Ti–V superconductors, where spin fluctuations are observed [26, 41].

The temperature dependence of the upper critical field, $H_{c2}(T)$, is estimated from the peak value in $d\rho/dT$ and the onset of the jump in $C(T)$, as shown in figure 5(a). The determination of $H_{c2}(T)$ is consistent between the two independent measurements. The $H_{c2}(T)$ data shows linear variation

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**Figure 5.** (a) Temperature dependence of the upper critical field, $H_{c2}(T)$ estimated using the temperature dependences of specific heat and electrical resistivity. The dashed line shows the orbital limit of $H_{c2}$ estimated using the WHH model by exploiting the initial slope of the $H_{c2}(T)$ curve. The solid line is obtained using the WHH model by varying the $T_c$ and the slope of $H_{c2}(T)$ to obtain the zero field limit of $H_{c2}$. (b) Isothermal $H$ in low magnetic field for different temperatures below the $T_c$. The $H$ values are corrected for the demagnetization effects and the straight line (slope $\approx -1$) is fit to the experimental data to estimate $H_{c1}$. (c) The magnetic field dependence of $\Delta M$ in the temperature range 2–8 K, where the $\Delta M$ is the difference between the experimental data $M(H)$ and the fitted straight line in low fields. The dotted horizontal line, $\Delta M = 0.05$ mT, represents the criterion to estimate the lower critical field value ($H_{c1}$).
as also observed in [18]. The \( H_c^2(T) \) is not well described by the GL expression, \( H_c^2(T) = H_c^2(0)[(1 - t^2)/(1 + t^2)] \), where \( t = T/T_c \) (not shown). The derivative, \((dH_c^2/dT)_{T=T_c} = -2.2 \pm 0.03 \text{ T}^{-1} \), is estimated by fitting a straight line to the data points below \( T_c \). The \( H_c^2(T) \) data is examined in terms of the WHH model in the dirty limit [42] using the equation given below, which is widely used to describe the upper critical field in different superconductors [43–45]

\[
\ln \left( \frac{1}{t} \right) = \sum_{\nu=-\infty}^{\infty} \left[ \frac{\nu}{2(\nu+1)} - \left( \frac{\nu}{2(\nu+1)} + \frac{(\alpha_M/\tau)^2}{(\nu+1)} \right)^{-1} \right],
\]

(2)

where \( t = T/T_c \), \( \tau = 2eH(\nu^2/\sigma T_c) / (\rho/\tau) \), \( \nu = \text{Fermi velocity} \), and \( \tau = \text{relaxation time of electrons} \). \( \alpha_M = 3/(2m^2 \nu^2/\tau) = H_c^2(0)/1.84 \sqrt{2} T_c \). is the Maki parameter and \( \lambda_{so} = 1/3 \pi T_c T_2 \), is the spin–orbit coupling parameter, with the relaxation time of electrons for the spin–orbit interaction \( T_2 \). Using equation (2), \( H_c^2(T) \) may be obtained by providing fixed values for \( T_c \) and \((dH_c^2/dT)_{T=T_c} \), and then varying \( \lambda_{so} \) and \( \alpha_M \). In the case of \( \lambda_{so} = \alpha_M = 0 \), the \( H_c^2(0) \) is defined as the orbital limit of the upper critical field. This is shown as the dashed line in figure 5(a) and it clearly does not describe the experimental data. To obtain the \( H_c^2(0) \) we use a phenomenological fit routine to extrapolate to lower temperatures by allowing the WHH expression and allowing \( T_c \) and \( dH_c^2/dT \) to vary. The fit parameters are not meaningful in themselves but this allows a more realistic estimation of \( H_c^2(0) \) of 16.5 T (solid line in figure 5(a)). It is to be noted that for Nb0.18Re0.82, the \( \lambda_{so} \approx 0.7 \), which is close to the intermediate coupling limit. Therefore, the Chandrasekhar–Clogston or the Pauli paramagnetic limit for the upper critical field, \( H_p(0) \) is estimated \( \sim 16 \text{ T} \). A value of the upper critical field, comparable to or larger than the Pauli paramagnetic limit is considered a potential signature for unconventional superconductivity [46, 47]. In real materials, the upper critical field of a system is generally influenced by both orbital and paramagnetic effects. The relative importance of these two competing effects may be defined in terms of the Maki parameter, \( \alpha_M = \frac{H_{C2}^2(0)}{\sqrt{2} H_{C2}(0)} \) and usually \( \alpha_M \ll 1 \) [42]. In the case of heavy fermions and the systems with multiple small Fermi pockets, the \( E_F \) may be quite small which results in \( \alpha_M \gg 1 \) and yields a possibility for the Fulde–Ferrell–Larkin–Ovchinnikov state [42, 48]. However, in our study, \( \alpha_{so} \approx 0.6 \), which lies in the intermediate limit of weakly-correlated and heavy fermion materials.

We used the isothermal dc magnetization measured at different temperatures below the \( T_c \) to estimate the lower critical field, \( H_{c1} \). The experimental \( M(H) \) data in the Meissner state is corrected for the demagnetization effects using the procedure of a linear fit near \( H_{app} = 0 \). This provides the slope \( (M/H_{app}) \) of the raw \( M(H) \) data in the Meissner region. It is known that the magnetic flux density, \( B \), inside a magnetic material is defined as \( B = \mu_0 (H_{eff} - \alpha M) \), where \( \alpha \) is the demagnetization factor and \( H_{eff} = H_{app} - \alpha M \). Hence, for a superconductor in the Meissner state, \( B = 0 \), then,

\[
M/H_{app} = 1/(\alpha - 1), \quad \text{where} \quad M/H_{app} \quad \text{is the slope of the raw} \quad M(H) \quad \text{data in the Meissner state, and thus we estimated} \quad \alpha \approx 0.34. \quad \text{Then, the effective magnetic field is estimated as} \quad H_{eff} = H_{app} - \alpha M \quad \text{and is plotted with the magnetization}, \quad M, \quad \text{which is shown in figure 5(b)}. \quad \text{To estimate the lower critical field, the} \quad M(H) \quad \text{data in figure 5(b) is fitted with a straight line in the low magnetic field region. The deviation from linearity is considered to be the lower critical field value. The straight line fit to the low magnetic field section of the} \quad M(H) \quad \text{curve is subtracted from the data and the resulting} \quad \Delta M \quad \text{is plotted for different isotherms in figure 5(c)}. \quad \text{The dashed line in the figure indicates the threshold, where} \quad \Delta M \quad \text{decreases to the noise level,} \quad \Delta M = 0.05 \text{ mT}, \quad \text{is used as the criterion to estimate the} \quad H_{c1} \quad \text{value}. \quad \text{This method used widely in literature can provide a good indication of the trend of the} \quad H_{c1}(T) \quad \text{curve, although may not be relied upon for accurate absolute values} [49].

Inset of figure 6(a), shows that the temperature dependence of the extracted lower critical field, \( H_{c1}(T) \), follows a
cubic temperature dependence in the whole temperature range of measurement instead of the usual quadratic behavior seen previously [9]. However, it is to be noted that in [9] the magnetization measurements were performed using VSM (on an as-cast sample), whereas, in the present study, the measurements were performed using a SQUID-VSM (on a well annealed sample), which has at least two order of magnitude higher sensitivity than the VSM. The zero field limit of the lower critical field, $H_{c1}(0) = 9.3$ mT, is almost twice than the value observed previously [9]. Using the expression, $\lambda(0) = (\phi_0 \ln\kappa/4\pi H_{c1})^{1/2}$, the zero field limit penetration depth is estimated to be $\sim 305$ nm, where $\kappa = 192$, is the GL parameter and is defined as $\kappa = H_{c2}/\sqrt{2}H_c$. The thermodynamic critical field, $\mu_0 H_{c1}(0) = 66.4$ mT, is estimated using the expression, $H_{c1}(0) = 4.23\gamma^{1/2}T_c$, provided in [30]. In the framework of the local London model, making use of the lower critical field, $H_{c1}(T)$, the normalized superfluid density $\rho_s(T)$ is estimated using the following expression [23, 51]

$$\rho_s(T) = \frac{\lambda(0)}{\lambda(T)} = \frac{H_{c1}(T)}{H_{c1}(0)}, \tag{3}$$

where $\lambda(T)$ is the temperature dependence of the penetration depth and $\lambda(0)$ is the zero field limit of $\lambda$. The temperature dependence of the $\rho_s$ for the Nb$_{0.18}$Re$_{0.82}$ superconductor is shown in figure 6(a). The open symbols with error bars represent the experimental data. Nb$_{0.18}$Re$_{0.82}$ is a multiband (two-gaps) superconductor as reported in [19]. Hence, we used the two-gap model to explain the superfluid density, which is widely used to explain the multiband behavior in superconductors [23, 51]. For a two-gap superconductor, the normalized superfluid density may be expressed by the following relation [52]

$$\rho_s(T) = 1 + 2\left(\int_{\Delta_S(T)}^{\infty} \frac{dF(E)}{dE} D_s(E)dE\right) + \left(1 - c\right)\left(\int_{\Delta_L(T)}^{\infty} \frac{dF(E)}{dE} D_L(E)dE\right), \tag{4}$$

where $F(E)$ is the Fermi function, $D_{S,L}(E) = E/\sqrt{E^2 - \Delta_S^2(T)^2}, \Delta_S$ and $\Delta_L$ are the small and large superconducting gap respectively. The parameter $c$ is the fraction that the small gap contributes to the superconductivity. Equation (4) is used to fit the normalized superfluid density data, where the values of the gap parameters, $\Delta_L = 1.86$ meV, $\Delta_S = 1.36$ meV and $c = 0.32$, have been taken from [19] to generate the curve (for consistency). The data is well explained using the two-energy gap model and the fitted curve is shown in figure 6(a). For comparison, a single band BCS model is also used to explain the experimental data, where the same energy gap value ($\Delta = 1.9T_c$) is considered to generate the fitting curve as observed in [18]. However, figure 6(a) shows that the single band BCS model is insufficient to explain the data. Our study with polycrystalline sample provides further confirmation that Nb$_{0.18}$Re$_{0.82}$ is a two-gap superconductor as previously observed for the single crystal sample in [19].

In order to classify the non-centrosymmetric Nb$_{0.18}$Re$_{0.82}$ compound as a conventional or unconventional superconductor, we compared it with the other classes of superconductors, using the Uemura plot [55], as shown in figure 6(b). In the Uemura plot, the superconducting transition temperature ($T_c$) is plotted as a function of the Fermi temperature ($T_F$) estimated using the superfluid density. The shaded portion in figure 6(b) represents the unconventional superconductors, such as, heavy fermion superconductors, Fe- pnictide and high temperature superconductors. Most of the elemental superconductors, e.g Sn, Al, are well outside the shaded region. The Fermi temperature ($T_F$) for Nb$_{0.18}$Re$_{0.82}$ is obtained using the relation, $k_BT_F = h^2/(3\pi^2/2)^{3/2}v_F^2/\mu_0e^2\lambda_{ph}$, where $k_B$ is the Boltzmann constant, $n_s$ is the superfluid density, $n_e$ is the electron mass and $\lambda_{ph}$ is the electron–phonon coupling constant [53]. The superfluid density $\sim 5.27 \times 10^{26}$ m$^{-3}$ is estimated using the relation, $n_s = \frac{m_e}{\mu_0e^2\lambda_{ph}}$ [53], where $\lambda$ is the penetration depth. The same order of magnitude is also obtained for the superfluid density, using, $n_s = n_lT_0$, where $l$ is the mean free path and $\xi_0$ is the Pippard coherence length. The estimated value of $T_F$ is $\sim 2740$ K, which is plotted with the superconducting transition temperature of the sample ($T_c$), in figure 6(b). It is seen that the data point for the Nb$_{0.18}$Re$_{0.82}$ in the Uemura plot, lies in close proximity of the unconventional superconductors. A similar behavior is observed in the weakly-correlated (Ca/Sr)$_3$Ir$_2$Sn$_2$S$_5$ system [54]. It is argued that in the presence of competing orders or multiband behavior, a phonon-mediated BCS superconductor may also show the characteristics of an unconventional superconductor [54]. A recent observation of TRSB in Nb$_{0.18}$Re$_{0.82}$ superconductor indicates that the superconductivity in this system is not of conventional nature [20].

The search for a spin-triplet component in NCS is of great interest at present. The non-centrosymmetric lattice structure leads to an ASOC in the system. The strength of the ASOC is responsible for the component of spin-triplet pairing, which leads to line nodes in the superconducting energy gap [56]. The phase and the amplitude fluctuations of the superconducting order parameter have different contributions in the node and anti-node regions, which may alter the density of states near $T_c$ [57]. Consequently, it may modify the behavior of the superconducting order parameter as compared to the conventional mean field dependence ($\sim(T - T_c)^{1/2}$) near $T_c$. The amplitude and the phase fluctuation analysis relies on the isofield reversible magnetization data, $M(T)$, and is a convenient tool to investigate the non-s-wave behavior in the unconventional superconductors [58–60].

According to the conventional theory of the upper critical field, $H_{c2}$ [61], the magnetic induction, $B$, obtained from the GL equation may be expressed as [62]

$$B = H - \frac{4\pi e^2 h}{mc} |\psi|^2, \tag{5}$$

where $\psi$ is the order parameter. Using, $M = (\delta - H)/4\pi$, we may write

$$M = -\frac{e^2 h}{mc} |\psi|^2. \tag{6}$$
This relation shows that the $\sqrt{M}$ is directly proportional to the amplitude of the superconducting order parameter. Hence, near the $T_c$, the magnetization may be expressed as $\sqrt{M} \propto (T_c(H) - T)^m$, where $T_c(H)$ is the mean field transition temperature. Within the GL theory, for both s-wave and d-wave superconductors, the exponent $m$ is equal to 1/2 [62, 63]. It is known that in the low-$T_c$ superconductors, the phase of the order parameter is unimportant and the superconducting transition is well described using mean-field theory with an exponent, $m = 1/2$ [59]. On the other hand, superconductors with small superfluid density, such as, the high-$T_c$ oxides, bears a relatively small phase stiffness and poor screening, which leads to phase fluctuations playing a significant role [64]. It is thought that, due to the spin-triplet component, the presence of line nodes may make the phase fluctuations relevant in NCS [58]. Consequently, the exponent $m$ may differ significantly from the mean-field value, as observed in the non-centrosymmetric Li$_2$(Pd-Pt)$_3$B superconductor [58].

Figure 7(a) shows the isofield temperature dependence of the magnetization, $M(T)$, in the zfc and ffc protocols, which are used to obtain the reversible (equilibrium) magnetization. Each isofield $M(T)$ curve in figure 7(a) is corrected for the background magnetization using a linear relation, as demonstrated in the inset of figure 7(a) for 1 T magnetic field. Figure 7(b), shows the temperature dependence of $\sqrt{M}$, where $M$ represents the background corrected value. We are primarily focusing on the reversible region below $T_c(H)$ to analyze the phase fluctuations behavior. The region above $T_c(H)$ is important to study the amplitude fluctuations, which generates an anomalous enhancement of magnetization above $T_c(H)$. The phase-mediated superconducting transition is well described by fitting the reversible magnetization region below $T_c(H)$ using the relation, $\sqrt{M} \propto (T_c(H) - T)^m$, where $T_c(H)$ is the apparent transition temperature and $m$ is the fitting exponent [58–60]. Deviation of $m$ from the mean field value ($m = 1/2$), suggests the phase-mediated nature of the transition. Results for fitting of each isofield $\sqrt{M}$ versus $T$ curve is shown in figure 7(b). In 0.1 T magnetic field, the extracted value of the exponent $m = 0.78$, which is larger than the mean field value. This suggests that the superconducting transition is phase mediated, indicating the possibility of a spin-triplet component [58]. However, the inset (ii) of figure 7(b) shows that the exponent, $m$, decreases with applied magnetic field and depicts the mean field value, $m = 0.5$ for $\mu_0H = 1$ T. We speculate that for high magnetic fields and at low temperatures, the lowest Landau levels could mask the effect of the phase fluctuations resulting in the mean field type transition being the correct description in high magnetic fields. The inset (i) of figure 7(b) shows the linear variation of apparent transition temperature $T_a$ with magnetic field $H$. The phase fluctuation analysis suggests the possible admixture of spin-singlet and spin-triplet pairing components in the Nb$_{0.18}$Re$_{0.82}$ NCS.

4. Summary and conclusion

In conclusion, we presented a detailed investigation of the Nb$_{0.18}$Re$_{0.82}$ superconductor using electrical transport, specific heat and magnetization measurements. Structural characterization is performed using XRD and optical metallography techniques, and both confirm the high quality of the sample. Electrical resistivity in the normal state is interpreted using an empirical relation, which includes the presence of phonon-assisted interband scattering. The magnetoresistance data is explained in terms of two-band model. The upper critical field, $H_{c2}$, varies linearly with temperature in the range studied and does not follow the WHH model over the whole temperature range of measurement and the zero temperature limit of $H_{c2}$ exceeds the Pauli limit, both suggesting the possibility of unconventional pairing. The KW and the Uemura plots suggest unconventional behavior which is also strengthened by the recent observations of TRSB in this system. The normalized superfluid density estimated using the lower critical field, $H_{c1}(T)$, is well explained within the multiband picture. The phase of the superconducting order parameter is analysed using the reversible magnetization data, which indicates that the superconducting transition is phase mediated. This implies the possibility of an admixture of spin singlet and spin triplet pairing consistent with the anticipated influence of anti-symmetric spin–orbit coupling in this NCS.
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