Superconducting properties of the non-centrosymmetric superconductors TaXSi (X = Re, Ru)

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Abstract
Ternary noncentrosymmetric superconductors TaXSi (X = Re, Ru) investigated by magnetization, resistivity, and specific heat measurements. It crystallize in the orthorhombic TiFeSi-type structure with superconducting transition $T_c = 5.32$ K and 3.91 K for TaReSi and TaRuSi, respectively. A low value of specific heat jump and the upper critical field concave nature suggests a nontrivial superconducting gap.

Keywords: noncentrosymmetric superconductor, unconventional superconductors, ternary compounds

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

1. Introduction

Superconductivity in noncentrosymmetric (NCS) systems has sparked a renewed research interest owing to their fascinating properties of fundamental interest, such as anisotropic superconducting gap, time reversal symmetry breaking, and the presence of Majorana quasiparticles [1–3]. Systems with strong spin–orbit coupling have been the prime candidate to show nontrivial band topology, leading to topologically protected zero-energy surface modes [4–6]. NCS materials are remarkable in this regard with their intrinsic Rashba-type antisymmetric spin-orbit (ASOC) interactions that lift the spin degeneracy of the electronic bands at the Fermi level and generate complex spin textures [7–10]. This, in general, can lead to Cooper pair of mixed singlet-triplet character, leading to a broken time reversal symmetry and anisotropy in the superconducting gap [1, 11–14]. Akin to topological insulators, this nontrivial pairing results in various types of protected zero energy states at the edge or surface of NCS materials [15]. Furthermore, topologically protected zero-energy boundary modes also occur in NCSs with an anisotropic superconducting gap [3, 16, 17].

The current research in noncentrosymmetric materials is focused on finding new materials with high ASOC and establishing a relation between the strength of ASOC and its influence on the superconducting ground state. The discovery of CePt$_3$Si [1] with nodes in the superconducting gap has revived interest in this field. Experimental evidence suggests a strong ASOC (50–200 meV) [12] in this material has triggered the presence of line nodes. Another remarkable evidence of ASOC dependence on the gap structure was visible when Pd was replaced with Pt in Li$_2$(Pd, Pt)$_3$B [18, 19]. It has shown the presence of triplet along with singlet, which challenges the primary concepts explaining the superconducting phenomenon. Several other members of the noncentrosymmetric family have also shown anisotropic [20–24] gap structure, while only a handful of compounds has shown time reversal symmetry breaking [25–32]. Despite evidence supporting the ASOC dependence on the superconducting ground state, several materials have shown conventional isotropic BCS superconductivity [33–39]. Among which, LaPt$_3$Si with a similar structure as CePt$_3$Si and very strong ASOC has failed to show any unconventional behavior [40]. At the same time, few noncentrosymmetric materials with very low ASOC have also shown triplet presence, and nodal superconductivity [41]. It is also suggested for CePt$_3$Si that the ferromagnetic ordering might have caused the nodal behavior, which is absent for the case of LaPt$_3$Si. It raises questions on the
selective observation of unconventional superconductivity in NCS systems and their explicit dependence on the strength of ASOC.

Ternary noncentrosymmetric materials give an excellent platform to investigate the role of ASOC on the superconducting ground state similar to the case of Li$_2$(Pd, Pt)$_3$B. It is easy to play with the strength of ASOC, which can be tuned for the case of ternary materials by replacing the constituent elements. For the present study, we have selected TaXSi, where X represents Re/Ru. Both the materials crystallize into a noncentrosymmetric orthorhombic TiFeSi-type structure (space group $Ima_2$). The TiFeSi-type structure is a superstructure modification of ordered hexagonal Fe$_2$P structure. This structural transition occurs due to small displacements of atoms from their ideal hexagonal position, with a reduction in symmetry from hexagonal to body centered orthorhombic structure. The crystal structure strongly influences the superconducting properties of ternary equiatomic systems. Among these, ZrRuP with hexagonal Fe$_2$P structure has shown the highest $T_c$ at 13 K, while the TiFeSi family, in general, has shown low $T_c$. The high $T_c$ in hexagonal ZrRuP is expected to originate from the strong electron-phonon interaction, where the electron-phonon coupling constant has a value of 1.25 [42]. Furthermore, the initial band structure calculation revealed the enforced semi-metal nature of TaRuSi with possible topological nature. Hence, it will be interesting to look for the implications of the band structure on the superconducting ground state [43, 44]. Also, the TiFeSi-type structure falls under the globally stable nonsymmorphic symmetry, which is favorable for topological material [45]. Superconducting transition in TaXSi materials was reported in 1985, while the nature of the superconductivity and normal state property remained unexplored [46]. Being a heavy transition element, Re can induce a strong ASOC in TaReSi, compared to TaRuSi where Ru is a comparatively light element. This difference is expected to have effects on the superconducting properties as well as the ground state. In this paper, we have studied the superconducting and the normal state properties of both the samples using resistivity, specific heat and magnetization measurements.

2. Experimental details

Polycrystalline TaXSi ($X = \text{Re, Ru}$) samples were prepared using a standard arc melting technique. High purity Ta (99.99%), Re (99.99%), (or Ru (99.99%)), and Si (99.99%) were taken in a stoichiometric ratio and melted on a water-cooled copper hearth under high purity Argon gas. During the synthesis Ta and $X$ were melted together at the first step, and then melted with Si. This method reduces the weight loss in the melting process. The resulting ingot formed with the negligible mass loss was flipped and remelted several times to improve the homogeneity. The samples phase purity and crystal structure was confirmed by room temperature x-ray diffraction measurement using a PANalytical diffractometer equipped with CuK$_\alpha$ radiation ($\lambda = 1.5406 \text{Å}$). Magnetization measurements were done using a superconducting quantum interference device (MPMS 3, quantum design) at various temperatures and field ranges. The electrical resistivity and heat capacity measurements were performed using a physical property measurement system (PPMS, quantum design).

3. Results and discussion

3.1. Sample characterization

Figure 1 shows the x-ray diffraction pattern for the TaXSi samples. The samples crystallize into noncentrosymmetric orthorhombic TiFeSi-type structure (Space group $Ima_2$). The obtained diffraction pattern fits very well with the reported data showing the phase purity of the samples. Figure 1(c) shows the body-centered orthorhombic structure of TaXSi. The fitted lattice cell parameters for both the compounds are enlisted in table 1.

3.2. Resistivity

The figure 2 displays the temperature dependence of resistivity for TaReSi and TaRuSi in the range 1.8 K $\leq T \leq 300.0$ K. Both the samples showed a decrease in resistivity as temperature decreases, showing metallic behavior in the whole temperature
range. A drop in resistivity was observed at $T_c^{\text{onset}} = 5.62(5)$ K and $3.92(5)$ K, respectively, suggesting the onset of superconductivity. The low residual resistivity ratio indicates resistive property in the normal state region is sensitive to defects and other scattering centers present in the polycrystalline samples.

### 3.3. Magnetization

A temperature-dependent DC magnetic susceptibility measurement has shown a marked drop at around $T_c^{\text{onset}} = 5.32(4)$ K and $3.91(4)$ K at 1.0 mT (figure 3), suggesting the occurrence of superconductivity for Re and Ru variant, respectively. A type-II nature of both the samples is visible from the flux pinning nature during FCC measurement. The rectangular cuboid shape samples used for magnetization measurement, and the superconducting volume fraction is close to 100%, corresponding to a full diamagnetic shielding. A low field magnetization was carried out to estimate the lower critical field $H_c(0)$ for the compounds. The deviation from linear behavior in the magnetization curve is taken as the $H_c(0)$ at that particular temperature. An extrapolation of $H_c(0)$ using G-L equation $H_c(0) = H_c(0)(1 - T^2)$, where $T = T_c / T_f$, gives $H_c(0) = 4.64(8)$ mT and $6.27(4)$ mT for TaReSi and TaRuSi, respectively.

### 3.4. Upper critical field

The upper critical field $H_c(2)$ for the compounds is determined by both magnetization as well as resistivity measurements in the field range 10.0 mT $\leq H \leq 1.0$ T. The transition temperature was seen shifting towards lower temperatures as the field increases, with transition becoming broader. The onset of superconductivity in magnetization/resistivity at each field is taken as the value of $H_c(2)$. The $H_c(2)$ curve obtained from magnetization data in the 0.0–$T_c$ range can be extrapolated using the WHH model, considering the effects of orbital breaking. Pauli spin paramagnetism ($\alpha$), and spin-orbit scattering parameter ($\lambda_{\text{so}}$) [47, 48]. According to this model, $H_c(2)$ can be implicitly explained by the expression,

$$
\ln \left( \frac{1}{t} \right) = \left( \frac{1}{2} + \frac{i \lambda_{\text{so}}}{4 \gamma} \right) \psi \left( \frac{1}{2} + \frac{h + i \lambda_{\text{so}} + i \gamma}{2t} \right)
$$

$$
+ \left( \frac{1}{2} + \frac{i \lambda_{\text{so}}}{4 \gamma} \right) \psi \left( \frac{1}{2} + \frac{h + i \lambda_{\text{so}} - i \gamma}{2t} \right) - \psi \left( \frac{1}{2} \right)
$$

(1)

where $t = \frac{T_c}{T}$ is the reduced temperature, $\lambda_{\text{so}}$ is the spin-orbit scattering parameter, $\psi$ is the digamma function, $\gamma = \sqrt{(\alpha m h^2 - \frac{1}{4} \lambda_{\text{so}}^2}$, $\alpha m$ is the Maki parameter, and $h$ is the dimensionless form of the upper critical field given by $h = \frac{(4/\pi^2)}{H_c(2) / dH_c(2)/dT|_{T_c}}$. Extrapolating temperature dependence of $H_c(2)$ for the two samples with $\alpha m = 0.19$, 0.24 and $\lambda_{\text{so}} = 0$, respectively for TaReSi and TaRuSi gave a best fit using the model and is shown in figure 4. The upper critical field using the WHH model can be approximated by

$$
H_c^{\text{whh}}(0) = -0.693 \times T_c \times \frac{dH_c(2)(T)}{dT} \bigg|_{T=T_c}
$$

(2)

where $\alpha = 0.528 \frac{dH_c(2)(T)}{dT}|_{T=T_c}$. Combining the expressions, we get $H_c(2)(0) = 1.35$ T for TaReSi and 1.25 T by WHH model. However, this model is insufficient to reproduce the data points due to concave upward nature $H_c(2)$ for both samples, prominent for TaRuSi, giving an underestimated value of $H_c(2)(0)$. This can be arise from various reasons such as localization effects [49], twisting of electron orbits by a magnetic field [50], dimensional cross over [51], multi-gap behavior etc [52].

The slight upturn nature of $H_c(2)(T)$, prominent for the case of TaRuSi, is similar to the case reported for the two-gap superconductors MgB$_2$, YNi$_2$B$_2$C, LuNi$_2$B$_2$C, 2H-NbSe$_2$ [53–56]. Hence, we have attempted to describe the $H_c(2)(T)$ curve using the two-gap model, according to which $H_c(2)(T)$ is described by the parametric equation,

$$
\ln \left( \frac{1}{t} \right) = \left[ U(s) + U(\eta s) + \frac{\lambda_0}{w} \right]
$$

$$
+ \left( \frac{1}{4} \right) \left[ U(s) - U(\eta s) - \frac{\lambda_0}{w} \right] - \lambda_{hh} \lambda_{he} \left( \frac{w}{w^2} \right)^{1/2}
$$

$$
H_c(2) = \frac{2 \phi_0 T_s}{D_e} \eta = \frac{D_h}{D_e}
$$

(3)

$$
U(s) = \psi(s + 1/2) - \psi(1/2)
$$

where, $\lambda_{ee} = \lambda_{hh} - \lambda_{he}$, $\lambda_0 = (\lambda_{ee}^2 + 4 \lambda_{eh} \lambda_{he})$, $w = \lambda_{ee} \lambda_{hh} - \lambda_{he} \lambda_{eh}$. The variables, $\lambda_{ee}$, $\lambda_{hh}$, $\lambda_{he}$, $\lambda_{eh}$ are the matrix elements of the BCS coupling constants. $D_h$ and $D_e$ are the electron and hole diffusivity, respectively. $\phi_0$ is the flux quantum and $\psi(s)$

| Parameters | TaReSi | TaRuSi |
|-----------|--------|--------|
| a         | 6.972(7) | 7.132(4) |
| b         | 11.574(1) | 11.292(2) |
| c         | 6.657(6)  | 6.547(6) |

\[ \alpha = \beta = \gamma = 90^\circ \]
A fitting employed using this relation gave $H_{c2}(0) = 1.76(3)$ T and $1.46(2)$ T, respectively for TaReSi and TaRuSi. The coherence length is calculated to be $137(2)$ Å and $114(2)$ Å, respectively for Re and Ru variant using $\xi_{GL} = (\phi_0/2\pi H_{c2}(0))^{1/2}$ ($\phi_0 = 2.07 \times 10^{-15}$ Tm$^2$). And the magnetic penetration depth for the sample $\lambda_{GL}(0)$ is estimated using the relation

$$H_{c2}(0) = \frac{\phi_0}{4\pi\lambda_{GL}^2(0)} \left( \ln \frac{\lambda_{GL}(0)}{\xi_{GL}(0)} + 0.12 \right)$$

which is obtained as 3373(87) Å and 2766(62) Å, respectively for TaReSi and TaRuSi. Following the penetration and coherence length, the Ginzburg–Landau parameter for the samples can be found out as 25(1) and 18(1), respectively for TaReSi and TaRuSi.

However, the temperature dependence of $H_{c2}$ determined from resistivity measurements has shown a relatively high value. Such a high value can arise due to surface or filamentary effects. Here, much stronger scattering of electrons at grain boundaries can reduce the mean free path, reducing the coherence length and increasing the upper critical field. Also, a higher residual resistivity value ($\rho = 559.0$ and $647.0 \mu\Omega$ cm for TaReSi and TaRuSi respectively) indicates higher density of defects/disorder in the system. The magnetic flux line, in this case, can pin to these defects and hence reducing the effects of the orbital pair breaking, increasing the upper critical field. Similar high upper critical field is reported for LaPtSi, BaPtSi$_3$, LaInSi$_3$ [33, 39, 58]. We have extrapolated the data using both the G-L model and the two-gap model for the magnetization data. Similar to magnetization data, we have obtained better fitting of the data points using the two-gap model, giving rise to $H_{c2}(0) = 6.55$ T and 2.84 T respectively for TaReSi and TaRuSi.

### 3.5 Specific heat

Heat capacity measurement was carried out in the temperature range $1.9 \, K \leq T \leq 15 \, K$ showing the bulk nature of superconductivity in both compounds with a jump at $T_{\text{mid}} = 5.40(1)$ K and $3.86(4)$ K, respectively for TaReSi and TaRuSi. The

### Figure 3.

(a), (b) Temperature dependence of magnetic moment collected via zero field cooled warming (ZFC) and field cooled cooling (FCC) methods under an applied field of 1 mT. Onset of diamagnetic signal was observed at $T_{\text{ZFC}} = 5.32$ K and 3.91 K for TaReSi and TaRuSi respectively. (c) The lower critical field is estimated from the M-H curve using the G-L equation.

### Figure 4.

Estimation of the upper critical field by magnetization and resistivity measurements. The $H_{c2}(T)$ determined from magnetization is fitted by WHH, G-L, and two-gap model. The WHH model has failed to trace the data points, while the G-L and the two-gap model has successfully estimated the $H_{c2}(T)$. The $H_{c2}(T)$ determined from resistivity measurements has shown comparatively high value, probably due to surface effects. The resistivity data is fitted using the two-gap and G-L model, as shown.

is the digamma function. Though the fitting seems to be in good agreement with the experimental data, we must admit that there remains questionable reliability of the fitting parameters since the fit was done for a large number of parameters. However, extrapolating to zero temperature yields the values of $H_{c2}(0)$ as 1.81 T and 1.46 T, respectively for TaReSi and TaRuSi, close to that obtained from G-L fitting.

According to Maki theory [57], the the upper critical field at 0 K is related to $\alpha$ by the relation, $H_{c2}(0) = \alpha H_P(0)/\sqrt{2}$ where $H_P$ is the zero temperature Pauli limiting field. $H_P(0)$ can be relate to $H_P^{\text{BCS}}$, the BCS value for paramagnetic limiting field by the equation $H_P(0) = H_P^{\text{BCS}} \sqrt{T + \lambda_{e-ph}}$. Substituting $\alpha = 0.19, 0.24$ and $\lambda_{e-ph} = 0.63$ and 0.58 for TaReSi and TaRuSi respectively, we get $H_{c2}(0) = 1.63$ T and 1.52 T. This value is in close agreement with prediction from the G-L formula which describe the temperature dependence of $H_{c2}$ as

$$H_{c2}(T) = H_{c2}(0) \left[ \frac{1 - T^2}{(1 + T^2)} \right],$$

which is obtained as 3373(87) Å and 2766(62) Å, respectively for TaReSi and TaRuSi. Following the penetration and coherence length, the Ginzburg–Landau parameter for the samples can be found out as 25(1) and 18(1), respectively for TaReSi and TaRuSi.

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Table 2. Superconducting and normal state parameters for TaXSi (X = Re, Ru).

| Parameters | Unit | TaReSi | TaRuSi |
|-----------|------|--------|--------|
| $T_c$     | K    | 5.32   | 3.91   |
| $H_a(0)$  | mT   | 4.64   | 6.27   |
| $H_c(0)$  | T    | 1.76   | 1.46   |
| $H_c^p(0)$| T    | 9.73   | 7.15   |
| $\lambda_{GL}(0)$ | Å | 3373 | 2766 |
| $\xi_{GL}(0)$ | Å | 137  | 114   |
| $\theta_D$ | K | 338   | 296   |
| $D_e(E_F)$ | eV T.u. | 2.28 | 3.34  |
| $\Delta_0/n_B T_c$ | 1.4 | 1.36 |
| $m^*/m_e$ | 5.2 | 6.6   |

where $\mu^*$ is 0.13 for intermetallic superconductors. Inserting the value of Debye temperature $\theta_D$, the obtained values are 0.63(2) and 0.58(2), respectively for TaReSi and TaRuSi. This places both the superconductors in the moderately coupled family. The value of the Sommerfeld coefficient can be inserted in the equation

$$\gamma_n = \left(\frac{n^2 \pi^2}{6}\right) D_C(E_F)$$

(9)

to determine the density of states at the Fermi surface $D_C(E_F)$. This gives 2.28(3) and 3.34(1) $\text{state} / \text{eV T.u}$ for TaReSi and TaRuSi, respectively. The electronic contribution to the specific heat can be calculated by directly subtracting the phononic contribution, using the relation $C_n = C - \beta T^3 - \beta_s T^4$. Figure 6 shows the electronic specific heat, $C_n$ plotted against normalized temperature. A normalised jump in electronic specific heat, $\Delta C_n$ for both the samples are close to 1 (1.07 and 0.91 for TaReSi and TaRuSi respectively) which is less than the BCS approximation. An isotropic s-wave model [60] in the dirty limit regime can be used to trace the data points in the superconducting region, giving a normalized superconducting gap as $\Delta(0)/k_B T_c = 1.40(4)$ and 1.36(4).

A set of four equations as explained in [61, 62] is simultaneously solved to get BCS coherence length $\xi_0$, mean free path $\ell$, Fermi velocity $v_F$, superconducting carrier density $n$, and effective mass $m^*$. The obtained ratio $\xi_0/v_F = 9.66$ and 11.75 for TaReSi and TaRuSi indicate the dirty limit nature of the samples. The calculated parameters are tabulated in Table 2. Using the value of $n$, the Fermi temperature for the system can be extracted from the relation,

$$k_B T_F = \frac{\hbar^2}{2} \left(3/2 \right) \frac{n^{2/3}}{m^{*2/3}}$$

(10)

This gave the Fermi temperatures for the system as $T_F = 4997$ K and 5066 K respectively for TaReSi and TaRuSi.
The ratio, \( T_c / T_F \) for high \( T_c \) and unconventional superconductors fall in the range, \( 0.01 \leq \frac{T_c}{T_F} \leq 0.1 \) [63–66]. Here, this ratio \( \frac{T_c}{T_F} \approx 0.001 \) for both samples, placing them in the conventional family as shown in figure 7.

4. Conclusion

We have synthesized TaXSi (\( X = \text{Re}, \text{Ru} \)) and characterized it by XRD, resistivity, magnetization, and specific heat measurements. The samples showed a type-II superconducting nature below transition temperatures 5.32 K and 3.91 K, respectively, for TaReSi and TaRuSi. The temperature dependence of the upper critical field determined from magnetization and resistivity has shown an upward curvature, reminiscent of a two-gap nature. A relatively high value of the upper critical field \( T_c \) and unconventional superconductors with unconventional properties. The dashed line in the figure represents the Bose–Einstein condensation temperature. The positions of TaReSi and TaRuSi indicate a conventional nature of these materials.

The specific heat jump around \( T_c \) for both samples, placing them in the conventional family as shown in figure 7.

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