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Specific heat of the non-centrosymmetric superconductor Re_3W*

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The alloys of non-centrosymmetric superconductor, Re_3W, which were reported to have an α-Mn structure [P. Greenfield and P. A. Beck, J. Metals, N. Y. **8**, 265 (1959)] with $T_c = 9\,K$, are prepared by arc melting. The values of ac susceptibility and the low-temperature specific heat of these alloys are measured. It is found that there are two superconducting phases coexisting in the samples with $T_{c1} \approx 9\,K$ and $T_{c2} \approx 7\,K$, which are both non-centrosymmetric in structure as reported previously. By analysing the specific heat data measured in various magnetic fields down to a temperature of 1.8 K, we find that the absence of the inversion symmetry does not lead to an obvious deviation from an s-wave pairing symmetry in Re_3W.

Keywords: Re_3W, non-centrosymmetric, superconductivity, specific heat
PACC: 6155H, 7125, 7430E, 7420F

1. Introduction

Very recently the scientific community has paid much attention to understanding the so-called non-centrosymmetric superconductivity in heavy atom compounds, such as LiPtB, MgIrB, since the superconducting properties of such materials are expected to be unconventional.[1−7] Generally, in a lattice with inversion symmetry, the orbital wave function of the cooper pair has a certain symmetry and the spin paring will be simply in either a singlet or a triplet state. On the contrary, the absence of centrosymmetry in the lattice may bring a complexity to the symmetry of orbital wave function. On the other hand, the effect of the antisymmetric spin-orbital coupling in heavy atomic compounds gives rise to the break of the spin degeneracy, thus the existence of the mixture of spin singlet and triplet may become possible.[2,5] Most recently, a spin-triplet pairing component has been demonstrated in Li_2Pt_3B both by penetration depth measurement[4] and by nuclear magnetic resonance (NMR)[5] as ascribed to the large atomic number of Pt which enhances the spin-orbit coupling.

Re_3W is one of the members of the rhenium and tungsten alloy family. Up to now, two superconducting phases of Re_3W have been reported to have $T_c \approx 9\,K$[8] and $T_c \approx 7\,K$.[9] These two phases belong to the α-Mn phase (A12, space group $I\bar{4}3m$),[10] which is non-centrosymmetric in structure. Moreover, atomic numbers of Re and W are 75 and 74, respectively, close to that of Pt ($Z = 78$) in which it is predicted to has a strong spin-orbital coupling effect. Therefore, a similar spin-triplet pairing component to that in Li_2Pt_3B is expected in Re_3W theoretically. However, very recently, the superconducting phase of Re_3W with $T_c \approx 7\,K$ has experimentally been found to be that of a weak-coupling s-wave BCS superconductor by both penetration depth[9] and Andreev reflection measurements.[11]

In the present paper, we report on a low-temperature specific heat measurement on Re_3W alloys in which two superconducting phases coexist with $T_c = 9\,K$ and 7 K, separately, and the high temperature phase near 9 K accounts for nearly 78%–82% of total volume. The specific heat data down to 1.8 K can be fitted very well by the simple two-component model, which is based on the isotropic s-wave BCS theory. Furthermore, a linear relationship is found between zero-temperature electronic specific heat coefficient and applied magnetic field. These results sug-

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gest that the absence of the inversion symmetry does not result in novel pairing symmetry in Re_3W.

2. Experiment

The Re_3W alloys are prepared by arc melting rhenium (Re) and tungsten (W) powders (both with a purity of 99.9%) with a nominal component ratio of 3 : 1 in a Ti-guttered argon atmosphere. Normally, the obtained alloy is a hemisphere in shape with a dimension of 5 mm (diameter) × 5 mm (height). Some samples (e.g. sample ♯1 and sample ♯2) were pieces cut down from the original bulky alloys. The ac susceptibility and the resistive transition of these samples have been measured to identify their superconducting phases. Both samples have two superconducting transitions at about 9 K and 7 K as shown in Fig.1. The specific heat measurement is performed in a Physical Property Measurement System (PPMS, Quantum Design) down to a limited temperature of 1.8 K. The temperature dependence of specific heat under a magnetic field is obtained by cooling the sample in the field from a temperature well above T_c to the lowest temperature and then collecting data with temperature increasing. The data of two samples: #1 and #2 are ultimately the same except for a difference in superconducting volume for high T phase, i.e. 52% and 82% for samples #1 and #2, respectively.

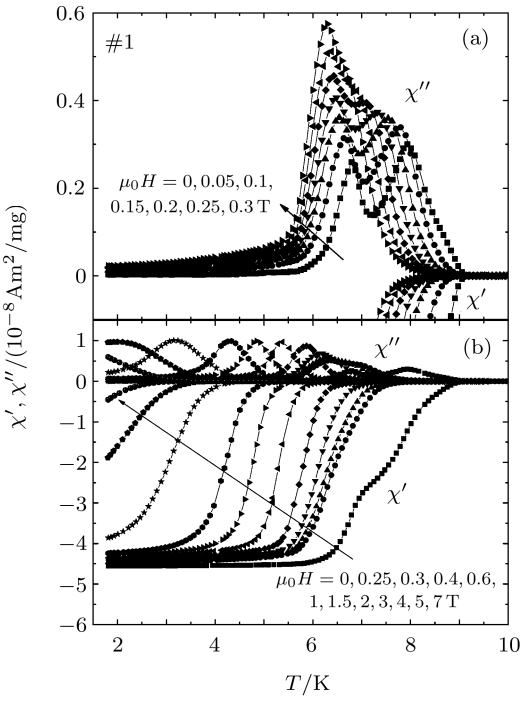


Fig.1. Temperature dependence of ac susceptibility on sample ♯1 under different dc magnetic fields, with ac field $h = 1$ Oe (1 Oe = 80 A/m) and frequency $f = 333$ Hz.

3. Results and discussion

The temperature dependences of ac susceptibility of sample ♯1 [$\chi(T) = \chi'(T) + \mathrm{i}\chi''(T)$] under dc magnetic field $\mu_0 H$ ranging from 0 to 7 T are shown in Fig.1. One can see that two distinct superconducting transitions occur at $T_{c1} \approx 9$ K and $T_{c2} \approx 7$ K in $\chi'(T)$ curve at $H = 0$ [Fig.1(b)], and double peaks in $\chi''(T)$ show up at their corresponding temperatures. The high-T phase with $T_c = 9$ K has been reported by Blaugher and Hulm,[8] and identified to be of α-Mn structure. On the other hand, the low-T phase with $T_c \approx 7$ K belongs to the symmetry group $I\bar{4}3m$, which includes an inversion axis of fourth order and therefore lacks the inversion centre.[9] Surprisingly, these two non-centrosymmetrical phases with different values of T_c coexist in our samples. As $\mu_0 H$ increases to about 7 T, the $\chi(T)$ curves are completely flat in a temperature limit of $T > 1.8$ K, showing no sign of superconducting transition. This implies that the upper critical field at $T = 1.8$ K is less than 7 T for both phases. Similar results are obtained from sample ♯2.

The specific heat of sample ♯2 has been measured and the data are presented in Fig.2 in the form of C/T versus T^2 at various magnetic fields. On each curve under low fields, i.e. $\mu_0 H < 1$ T, there are two jumps related to the superconducting transitions, which is consistent with the measurements of ac susceptibility. From the zero field data in a low temperature region, one can see that the residual specific heat coefficient γ_0 is close to zero, implying the absence of non-superconducting phase. The superconducting anomaly is suppressed gradually with H increasing, and the curve at 7 T shows no sign of superconductivity above 1.8 K. The low temperature part of the normal state specific heat plot at $\mu_0 H = 7$ T in Fig.2 is not a straight line, implying that the specific heat of phonon does not satisfy the Debye's T^3 law. A T^7 term may fit the normal state specific heat curve well

$$C_n/T = \gamma_n + \beta_3 T^2 + \beta_5 T^4 + \beta_7 T^6. \qquad (1)$$

The first term is the electronic specific heat in the normal state, and the others are the contributions of phonons to the specific heat. Fitting the data at 7 T to expression (1), the coefficients are determined as $\gamma_n =$

$17 \pm 0.1\,\mathrm{mJ/mol\cdot K^2}$, $\beta_3 = 0.185 \pm 0.001\,\mathrm{mJ/mol\cdot K^4}$, $\beta_5 = (1.63 \pm 0.01) \times 10^{-3}\,\mathrm{mJ/mol\cdot K^6}$, and $\beta_7 = (-2.087 \pm 0.005) \times 10^{-6}\,\mathrm{mJ/mol\cdot K^8}$. From the relation, $\beta_3 = \dfrac{12\pi^4}{5}\dfrac{N_\mathrm{A}k_\mathrm{B}Z}{\Theta_\mathrm{D}^3}$, where $N_\mathrm{A} = 6.02 \times 10^{23}$ is the Avogadro's constant and $Z = 4$ the number of atoms in one unit cell, we obtain the Debye temperature of our alloys, $\Theta_\mathrm{D} = 347.9\,\mathrm{K}$. These coefficients and Debye temperature are all very close to the results of other studies on Re-W alloys.[12−18]

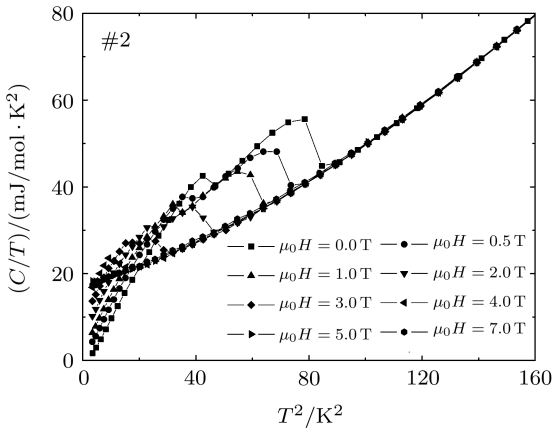


Fig.2. Specific heat data of sample ♯2 plotted as C/T versus T^2 at various fields.

By subtracting the phonon contribution, the electronic specific heat C_e is obtained, which is shown in Fig.3 as C_e/T against T. Before a quantitative analysis, we should notice that the low temperature specific heat under low fields already shows a strong evidence that Re_3W has a nodeless gap function. For a nodal superconductor (expected by the strong mixing of spin-singlet and spin-triplet pairing components in a heavily non-centrosymmetric superconductor such as Li_2Pt_3B), the low temperature C/T versus T relation should be power-law-like. However, as denoted by the dashed lines in Fig.3, if a linear relationship is assumed, the specific heat at zero field would be negative when the temperature approaches to zero.

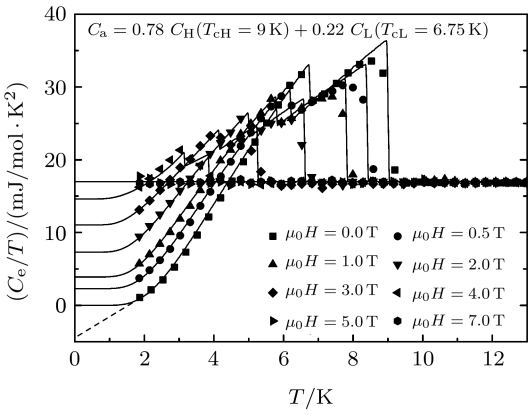


Fig.3. Specific heat of electron plotted as C_e/T versus T, where solid lines represent the calculated results which separate the electronic specific heat into two components with different T_c by using specific heat formula based on BCS theory.

Then we extract the low temperature $\Delta C_\mathrm{e}/T = [C_\mathrm{e}(H) - C_\mathrm{e}(0)]/T$ which is shown in Fig.4 to investigate the field-induced density of states (DOS).[19] It is found that $\Delta C_\mathrm{e}/T$ at 1.8 K has an almost linear dependence on magnetic field. A slight deviation from the linearity can be seen, which is due to the fact that quasiparticle excitation cannot be ignored at these finite temperatures. Such a linear dependence will be further proved by the value of $\Delta C_\mathrm{e}/T$ extrapolated to zero temperature in the following analysis (refer to Fig.6). This result presents a direct evidence that the field-induced DOS depends only on the density of the vortex cores in Re_3W which is a unique property of an *s*-wave superconductor. In order to further gain an insight into this point, we try to use a simple quantitative *s*-wave model to analyse our data in what follows.

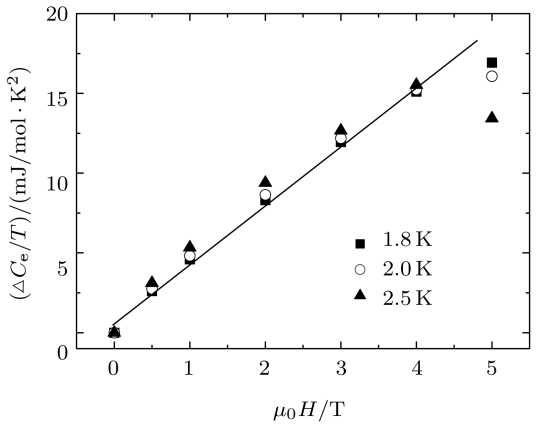


Fig.4. Field dependences of $\Delta C_\mathrm{e}/T = [C_\mathrm{e}(H) - C_\mathrm{e}(0)]/T$ at the lowest temperatures (1.8 K, 2.0 K, and 2.5 K) in our measurements.

Figure 5 shows the ac susceptibility and the specific heat data at zero dc field measured from the same sample (♯2). The ac susceptibility data have been normalized. The high-T phase occupies nearly $82 \pm 1\%$ of the whole superconducting volume. In order to fit the zero field electronic specific heat, we attempt to use the formula derived from thermodynamic relations based on the BCS theory[20]

$$C_\mathrm{es} = \frac{4N(0)}{k_\mathrm{B}T^2}\int_0^\infty \frac{\mathrm{e}^{\zeta/k_\mathrm{B}T}}{(1+\mathrm{e}^{\zeta/k_\mathrm{B}T})^2} \times \left(\varepsilon^2 + \Delta^2(T) - \frac{T}{2}\frac{\mathrm{d}\Delta^2(T)}{\mathrm{d}T}\right)\mathrm{d}\varepsilon, \quad (2)$$

where $\zeta = \sqrt{\varepsilon^2 + \Delta^2(T)}$ and $\Delta(T)$ is an isotropic s-wave gap which depends on temperature in the same way as expected by BCS theory. Since there are two phases coexisting in our samples, we use two separate terms of $C_{\rm H}$ and $C_{\rm L}$ to take into account the contributions of the high-T and the low-T phases, respectively. Thus the total specific heat can be expressed as follows:

$$C_{\rm e} = \omega_{\rm H} C_{\rm H} + \omega_{\rm L} C_{\rm L}, (\omega_{\rm H} + \omega_{\rm L} = 1), \qquad (3)$$

in which $\omega_{\rm H}$ and $\omega_{\rm L}$ indicate the weights of the contributions from the two phases.

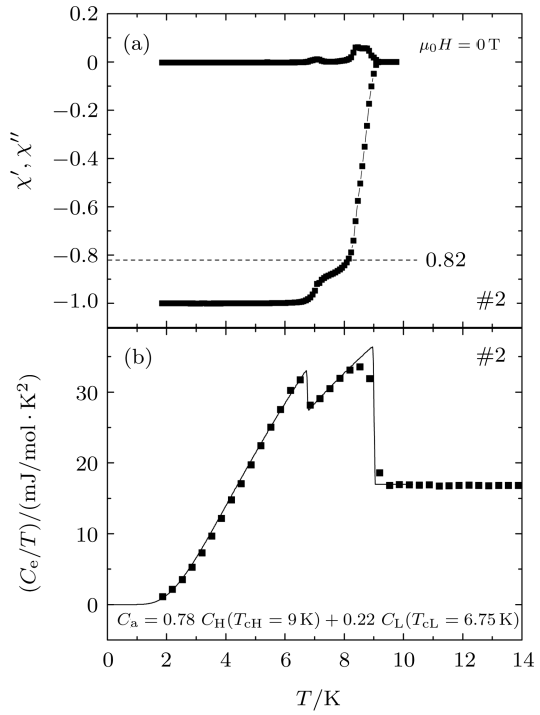


Fig.5. (a) the ac susceptibility of sample ♯2 on which the specific heat has been measured and (b) the zero field specific heat data, where the thick line is for the calculated result from the BCS theory.

According to expressions (2) and (3), we can nicely simulate the experimental data as represented by a solid line in Fig.5(b). The parameters for the best fit are $\Delta_{0\rm H} = 1.4\,\mathrm{meV}$ and $\omega_{\rm H} = 0.78$ for $T_{\rm cH} = 9\,\mathrm{K}$; $\Delta_{0\rm L} = 1.1\,\mathrm{meV}$ and $\omega_{\rm L} = 0.22$ for $T_{\rm cL} = 6.75\,\mathrm{K}$, where Δ_0 denotes the gap value at zero temperature. With these fitting parameters, it is obtained that $2\Delta_{0\rm H}/k_{\rm B}T_{\rm cH} = 3.6$ and $2\Delta_{0\rm L}/k_{\rm B}T_{\rm cL} = 3.7$, and both values are close to that of $2\Delta_0/k_{\rm B}T_{\rm c} = 3.52$ obtained from the BCS weak coupling theory. Interestingly, $\omega_{\rm H} = 0.78$ found here is very close to the relative weight 82% of the high-T phase which is obtained from the ac susceptibility measurement in Fig.5(a). Furthermore, $\Delta_{0\rm L} \approx 1.1\,\mathrm{meV}$, which is in good agreement with those obtained from the penetration depth and Andreev reflection measurements.[9,11] These results give strong evidence that there is no novel pairing symmetry in our alloys.

To acquire further evidence for this argument, we have carried out similar calculations for the specific heat in the mixed state by using the same weights obtained from the zero field calculation for the two phases. In the mixed state, there are two different regions, namely the core region and the outside core region. Therefore we adopt a simple two-component model,[21–23] in which the electronic specific heat is separated into two components. The electronic specific heat is thus written as

$$C_{\rm e} = \alpha \frac{H}{H_{\rm c2}(0)} \gamma_{\rm n} T + \left(1 - \alpha \frac{H}{H_{\rm c2}(0)}\right) C_{\rm es}, \qquad (4)$$

where α is an adjustable parameter. The first part on the right-hand side is the quasi-particle DOS from the normal vortex core region, and the second part comes from the superconducting region outside the core. The calculations are plotted as solid lines in Fig.3, and they are in good agreement with the experimental results for all magnetic fields.

In the superconducting state, $C_{\rm e} = \gamma T$, where γ is the electronic specific heat coefficient that is dependent on temperature and field. According to expression (4), the zero temperature electronic specific heat coefficient $\gamma(H)$ is equal to $\alpha H/H_{\rm c2}(0)\gamma_{\rm n}$, which is shown in Fig.6 as solid squares, and the solid line is a linear fitting to the data. The obvious linear relationship of γ-H presents further evidence that Re_3W is a conventional superconductor in which $\gamma(H)$ is proportional to the number of vortex cores and hence

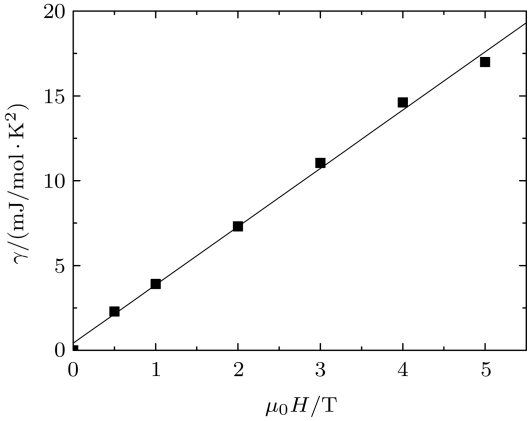


Fig.6. Electronic specific coefficient γ versus applied field $\mu_0 H$ at zero temperature, calculated from the BCS theory.

to applied field. For a nodal superconductor with novel pairing symmetry, on the other hand, a nonlinear relation $\gamma(H)$ should be expected, which is obviously not the case in our present samples.[24,25]

4. Conclusions

We have synthesized Re_3W alloys by arc melting. From the measurements of ac susceptibility and specific heat of the alloys, two distinct superconducting phases are found and identified to be noncentrosymmetrical in structure. Both the qualitative and quantitative analyses on the specific heat data in zero field and the mixed state are performed. We find that the simple two-component model based on the BCS theory with an isotropic *s*-wave gap can fit our experimental data down to 1.8 K very well, and a linear relation $\gamma(H)$ is obtained. All these results indicate that the absence of the inversion symmetry perhaps does not result in any novel pairing symmetry in Re_3W for both $T_c \approx 7$ K and $T_c \approx 9$ K phases.

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