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Crystal growth of the non-centrosymmetric superconductor $Nb_{0.18}Re_{0.82}$

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ABSTRACT

Single crystals of the non-centrosymmetric superconductor $Nb_{0.18}Re_{0.82}$ have been synthesized by the floating zone technique using a four mirror optical furnace equipped with Xe arc lamps. The crystal shows a superconducting transition at ~8.8 K. The quality and phase purity of the crystals were confirmed by x-ray Laue and Energy dispersive x-ray spectroscopy (EDAX) measurements. Large volumes of crystal obtained are suitable for detailed magnetic, transport and neutron scattering experiments.

1. Introduction

In conventional superconductors, inversion symmetry plays an important role in the formation of Cooper pairs. The discovery of non-centrosymmetric superconductors lacking inversion symmetry has attracted considerable attention both experimentally and theoretically [1–6]. The lack of inversion symmetry raises the possibility of an anti-symmetric spin–orbit coupling (ASOC) which can lift the degeneracy of the conduction band electrons and may cause the Cooper pairs of non-centrosymmetric superconductors to contain an admixture of spin-singlet and spin-triplet states [7]. This mixed pairing may lead non-centrosymmetric superconductors (NCS) to display significantly different properties from conventional superconducting systems.

The discovery of superconductivity in the non-centrosymmetric heavy fermion compound $CePt_3Si$ by Bauer et al. [1] revived interest in NCS. $CePt_3Si$ shows evidence of unconventional behaviour. However, several non-centrosymmetric superconductors have been reported to show conventional s-wave behaviour e.g. Ru_7B_3, $AMSi_3$ (A=Ca, Sr and Ba; M=Co, Rh, Ir, Ni, Pd and Pt), $Mg_{10}Ir_{19}B_{16}$, Mo_3Al_2C, $Nb_{0.18}Re_{0.82}$ and Re_3W [8–16] to name a few. It is believed that the superconducting wave function of some of these non-centrosymmetric superconducting compounds has two components, which correspond to spin-singlet and triplet pairing, but with a triplet component having a low superfluid density. There are also reports that the vortex dynamics of NCS show anomalous behaviour [17]. In order to perform detailed measurements to understand the issues related to mixed pairing and for investigations of the vortex state, high quality single crystals of NCS are highly desirable.

In this communication, we report the growth of large single crystals of the non-centrosymmetric superconducting compound, $Nb_{0.18}Re_{0.82}$ by the floating zone technique using an optical mirror furnace. Binary phase diagrams of Nb–Re [18–20] suggests that they have two intermediate phases, one with *sigma* (σ) structure and other with non-centrosymmetric α-Mn structure with the space group $I43m$ (No. 217). The *sigma* phase has been shown to exist over a narrow composition range at elevated temperatures only and forms peritectically at ~57 at% of Re. The phase with the α-Mn structure exists for around 62–87 at% of Re and melts congruently. Due to the wide stability range of the α-Mn structure, a slight variation in stoichiometry does not alter the crystal structure of compound, but results in a change in the superconducting transition temperature [21]. Nb_xRe_{1-x} has an optimal superconducting transition temperature, T_c~8.8 K, for x~0.18. Previous studies on the Nb_xRe_{1-x} compounds have been only on polycrystalline samples, and suggest that $Nb_{0.18}Re_{0.82}$ is a conventional s-wave superconductor [15]. The melting point for Nb–Re binary alloys ranges from ~2200 to 3000 °C depending upon the relative composition of Nb and Re. $Nb_{0.18}Re_{0.82}$ melts close to ~3000 °C [18–19]. In order to attain these high melting temperatures for crystal growth, either radio frequency (r.f.) heating or very powerful lamps are required when using the floating zone technique. We have used a four mirror optical furnace equipped with Xe arc lamps to grow single crystals of optimal superconductor $Nb_{0.18}Re_{0.82}$. This furnace has the capability of melting materials with melting points of up to a maximum of ~3000 °C. The crystals obtained by this technique are large enough for most physical property measurements and

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ideal for neutron scattering experiments, where large volumes of single crystals are essential.

2. Experimental

Polycrystalline samples were prepared by arc melting in a tetra-arc furnace (Cyberstar, Grenoble, France) on a water cooled copper hearth. The starting materials, high purity (5N) Nb and Re ingots, were melted under an argon gas (5N) atmosphere. The buttons were melted and flipped several times to ensure phase homogeneity. The observed weight loss during the melting was negligible. The phase purity of the polycrystalline buttons was checked by powder x-ray diffraction. Several polycrystalline buttons obtained in this manner were then melt cast in the shape of a rod ($\sim$40 mm in length and $\sim$3 mm in diameter) in a tri-arc furnace under an argon atmosphere. These rods were used as feed rods for the crystal growth in a high temperature optical furnace equipped with four 3 kW Xe arc lamps (CSI Model FZT-1200-X-VI VP). A tungsten rod of the same diameter was used as the seed rod for the first crystal growth and the crystal obtained was used as the seed for subsequent growths. The growth chamber was evacuated to a vacuum of $\sim 10^{-6}$ mbar (10^{-4} Pa) prior to starting the growth and then filled with argon gas to a pressure of 0.3 MPa for the growth. The crystal growth was carried out under a flow (1.0 l/min) of high purity argon gas. The feed and seed rods were rotated at 25–30 rpm and crystal growths were carried out at speeds of 3–6 mm/h. The crystals obtained were typically $\sim$30–35 mm long and $\sim$3 mm in diameter.

3. Results and discussion:

Fig. 1 shows an as-grown boule of $Nb_{0.18}Re_{0.82}$. The boule is shiny with a metallic lustre. $Nb_{0.18}Re_{0.82}$ melts congruently and therefore lends itself to crystal growth by the floating zone method. The molten zone was stable throughout and the best crystals were obtained for slow growth rates $\sim$3 mm/h. The crystals could be cut using a low speed diamond saw and did not require the use of a spark cutter. Prior to cutting, the crystals were examined using x-ray Laue back reflection. X-ray Laue photographs were collected along the length of each crystal at 1.5 mm intervals on several faces to confirm the crystal quality. The lower panel of Fig. 1 shows the x-ray Laue patterns obtained at three different positions along the length of the crystal grown at 3 mm/h. The photographs show sharp consistent patterns for the whole sample, confirming the high quality of single crystal. By use of the x-ray Laue patterns, crystals oriented along particular crystallographic axes were cut from the as-grown crystal for measurements. X-ray powder diffraction pattern taken on a small portion of powdered single crystal suggests that the compound is single phase and lattice parameters agree well with those reported for the polycrystalline $Nb_{0.18}Re_{0.82}$ [15]. Composition analysis was carried

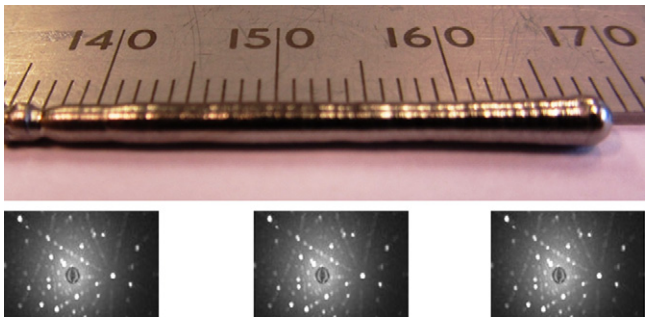

Fig. 1. As-grown single crystal of $Nb_{0.18}Re_{0.82}$ (upper panel). Lower panels show the x-ray Laue patterns taken from the lower, middle and upper regions of the single crystal.

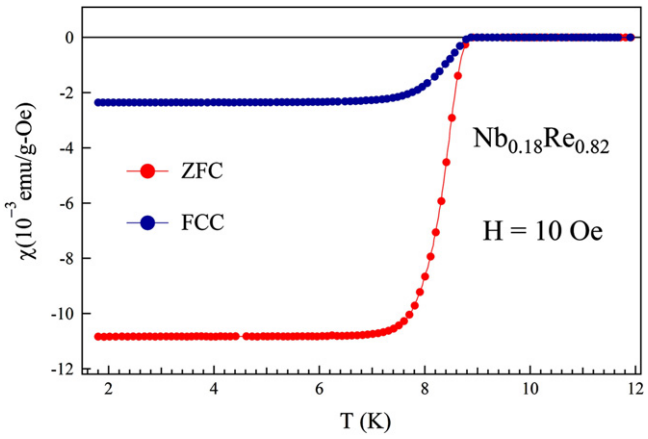

Fig. 2. Variation of magnetic susceptibility with temperature in an applied field along the [100] axis.

out by EDAX measurements using a SEM (JEOL 6100) to confirm the stoichiometry of the single crystal. The results obtained confirm that the average stoichiometry of the boule (Re = 84 ± 2 and Nb = 16 ± 2 at%) is very close to that of the feed rod (Re = 81.9 ± 2.9 and Nb = 18.1 ± 2.9 at%) used for the crystal growth. In order to check the superconducting properties of the crystal, dc magnetic susceptibility measurements were made using a Quantum Design Magnetic Properties Measurement System. Fig. 2 shows the magnetic susceptibility as a function of temperature for a field applied along the [100] axis of the crystal. The observed superconducting transition temperature, $T_c \sim 8.8$ K (T_c onset), in our single crystals is higher than that previously reported for polycrystalline ingots of the same composition measured by dc susceptibility (T_c onset $\sim$8.0 K) [15]. Detailed magnetization, transport and heat capacity measurements on the single crystals are currently in progress. Muon spin rotation and small angle neutron scattering studies are planned on the crystals to investigate the coupling mechanism and vortex state respectively in this superconductor.

In conclusion, we have been able to grow high quality single crystals of $Nb_{0.18}Re_{0.82}$ by the floating zone method using a four mirror optical furnace equipped with Xe arc lamps. Crystals of large volume can be obtained by this method. The crystals exhibit a sharp superconducting transition at $\sim$8.8 K. Further investigations of the superconducting properties of this non-centrosymmetric crystal are currently in progress and will be published separately.

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