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Pressure induced changes in the antiferromagnetic superconductor $YbPd_2Sn$

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Low temperature ac magnetic susceptibility measurements of the coexistent antiferromagnetic superconductor $YbPd_2Sn$ have been made in hydrostatic pressures ≤ 74 kbar in moissanite anvil cells. The superconducting transition temperature is forced to $T_{SC} = 0$ K at a pressure ~ 58 kbar. The initial suppression of the superconducting transition temperature is corroborated by lower hydrostatic pressure ($p \leq 16$ kbar) four point resisitivity measurements, made in a piston cylinder pressure cell. At ambient pressure, in a modest magnetic field of ~ 500 G, this compound displays reentrant superconducting behaviour. This reentrant superconductivity is suppressed to lower temperature and lower magnetic field as pressure is increased. The antiferromagnetic ordering temperature, which was measured at $T_N = 0.12$ K at ambient pressure is enhanced, to reach $T_N = 0.58$ K at $p = 74$ kbar. The reasons for the coexistence of superconductivity and antiferromagnetism is discussed in the light of these and previous findings. Also considered is why superconductivity on the border of long range magnetic order is so much rarer in Yb compounds than in Ce compounds. The presence of a new transition visible by ac magnetic susceptibility under pressure and in magnetic fields greater than 1.5 kG is suggested.

PACS numbers: 74.25.Dw, 74.62.Fj, 75.30.Kz

I. INTRODUCTION

Heavy fermion superconductivity has now been observed in over twenty cerium based compounds (for recent reviews see Refs. [1–4]). All of these materials are examples of superconductors on the border of antiferromagnetic order. In the Ce based heavy fermion superconductors, the Ce has nominally one f-electron (i.e. $4f^1$). Corresponding ytterbium heavy fermion systems have around one electron missing from their f-shell, thereby being the hole equivalent of the Ce in the heavy fermion compounds. One may therefore expect to find heavy fermion superconductors based on Yb in the $4f^{13}$ electron configuration. However, such behaviour in Yb ($4f^{13}$) systems actually proves to be exceedingly rare. As such, the search for heavy fermion Yb based superconductors in the presence of strong magnetic fluctuations continues.

One rare example of an Yb based superconductor with coexistent long-range antiferromagnetic order is $YbPd_2Sn$ [5]. In the case of the Ce heavy fermion compounds (with $4f^1$), strong hybridisation between the f-electrons and conduction electrons is a key ingredient to the occurrence of unconventional superconductivity. Here, the electrons responsible for magnetism are the same ones which cause superconductivity. However, at first sight the behaviour of $YbPd_2Sn$ appears like it could be more reminiscent of other compounds, i.e. the rare earth ternary compounds, including the Chevrel phase $\mathbf{M}Mo_6S_8$ ($\mathbf{M}$ = Gd, Tb, Dy, and Er), some borocarbides ($\mathbf{R}NiB_2C$ with $\mathbf{R}$ = Tm, Er, Ho and Dy) and the rhodium boride $SmRh_4B_4$ [6]. Here, the electrons on different sublattices are responsible for the two types of behaviour. $YbPd_2Sn$ is additionally in the same class of materials as $ErPd_2Sn$ [7]. It is a Heusler compound which has the cubic Cu_2MnAl-type structure [8]. It contains a significantly larger percentage of magnetic rare earth ions than other magnetic superconductors of its type. It is a particularly interesting case due to the relatively short distance between nearest neighbour magnetic ions (4.7 Å) compared to 5.3 Å for $\mathbf{R}Rh_4B_4$ and 6.5 Å for $\mathbf{R}Mo_6S_8$. The proximity between the magnetic ions prevents a clear isolation between the magnetic and superconducting sublattices [9].

$YbPd_2Sn$ at ambient pressure undergoes a

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transition into superconductivity at $T_{SC} = 2.3$ K, and a further transition into antiferromagnetism at $T_N = 0.22$ K [8]. The antiferromagnetic order (measured using neutrons at T = 30 mK [10]) has a commensurate structure (FCC-type I) with an ordering wavevector of $\mathbf{k} = [0, 0, 1]$. Within the ab-plane the moments are ordered ferromagnetically, but adjacent planes order antiferromagnetically. The ordered Yb magnetic moments have magnitude m = 1.4(1) μ_B and are aligned along the [1, 1, 1] direction.

The superconductivity has been shown to display reentrant behaviour in a modest magnetic field of 500 G (0.05 T) [8], whose origin is believed to be due to the competition between magnetic and superconducting ordering fluctuations [11].

II. EXPERIMENTAL METHODS

The ac magnetic susceptibility measurements under pressure were undertaken in a standard BeCu Dunstan type anvil cell employing moissanite anvils with a 1.0 mm beveled culet. The pre-indented gaskets were made of BeCu or phosphorus bronze. The pressure medium used in these cells was Daphne oil. To ensure that the samples were not squashed between the anvils or by the measurement coils, they had typical dimensions 150 μm x 100 μm x 50 μm.

Magnetic susceptibility measurements were made using a microcoil setup based on the design of Alireza [12]. The diameter of the 10 turn balanced pick-up coil was 250 - 300 μm. All wiring was insulated from the gasket using cured Stycast 1266 with alumina. At room temperature, ruby was used as a pressure gauge for all measurements. Additionally at low temperatures for p $\geq$ 45 kbar, the superconducting transition temperature of lead was used as a pressure gauge [13].

The four point electrical resistivity measurements were made in a piston cylinder cell [14]. In this case the pressure medium was a mixture of 50 % n-pentane with 50 % iso-pentane, and lead was used to measure the pressure.

Electrical measurements were amplified using low temperature transformers. The signal was multiplied further by Brookdeal EG&G 5006 ultra-low noise pre-amplifiers. The measurement was taken with a lock-in amplifier. The low temperatures were achieved using a CMR adiabatic demagnetisation refrigerator, whose optimal base temperature was 55 mK. The polycrystalline samples measured were cleaved from the same ingot.

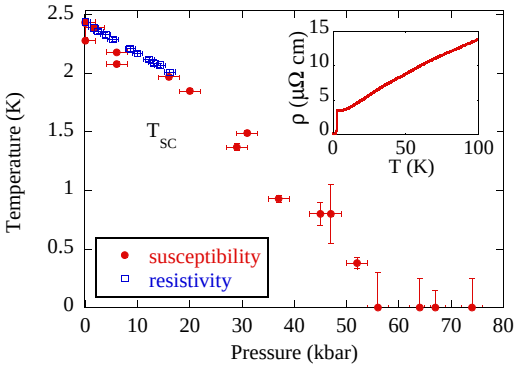


FIG. 1: (Color online) Main: The suppression of the superconducting transition temperature of $YbPd_2Sn$ as a function of pressure in zero applied magnetic field. The filled circles are measurements from magnetic susceptibility, and the open squares are from four point resistivity. Inset: An example of the drop observed in the ac magnetic susceptibility at the onset of superconductivity in $YbPd_2Sn$ [15]. This data was taken at ambient pressure and with zero applied magnetic field.

III. RESULTS

Ambient pressure four-point resistivity measurements indicate that the RRR of this ingot was 12, which is comparable to some of the better samples reported in literature.

At ambient pressure there was a clear drop in the magnetic susceptibility signal at T = 2.4 K, which has been attributed to the superconducting transition observed in other published work. An example of this signal drop is given in the inset of Fig. 1. Note that the y-axes of all of the insets to figures shown in this section display the measured ac magnetic susceptibility signal in arbitrary units, on a linear scale.

The superconducting transition was observed as a function of pressure using both magnetic susceptibility and four point resistivity [15] (Fig. 1). In this figure, the superconducting transition temperature, which was estimated as the 90 % point of the drop, is seen to be suppressed with pressure. The transition appears to be forced to below our minimum readable temperature, typically 100 mK, for pressures above 58 kbar. Relatively large error bars are depicted on data points at pressures above the expected critical pressure. These arise because the antiferromagnetic peak in the magnetic susceptibility data at higher temperatures (i.e. when $T_N \geq T_C$) means that it is difficult to unambiguosly rule out the possibility of superconductivity at the lowest temperatures measured.

The superconductivity displayed reentrant behaviour, and was observed as a function of

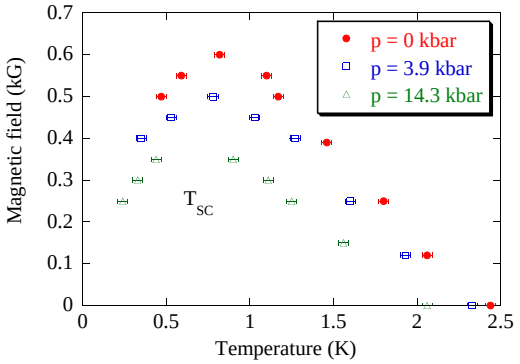


FIG. 2: (Color online) The reentrant superconducting behaviour of $YbPd_2Sn$ as a function of magnetic field for p = 0 kbar (filled circles), 3.9 kbar (open squares) and 14.6 kbar (open triangles), as measured by four point resistivity [15]. The plotted data indicates the temperature of the 90 % point of the drop at the onset or return from superconductivity.

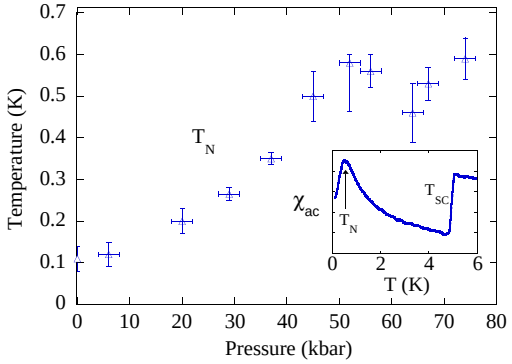


FIG. 3: (Color online) Main: The enhancement of the antiferromagnetic transition temperature of $YbPd_2Sn$ as a function of pressure in zero applied magnetic field. Inset: The relative sizes of antiferromagnetic peak of $YbPd_2Sn$ and the superconducting transition of lead at T = 4.9 K (indicating a pressure of 67 kbar). The lead was $\sim$ 12 times smaller by volume than the $YbPd_2Sn$ sample.

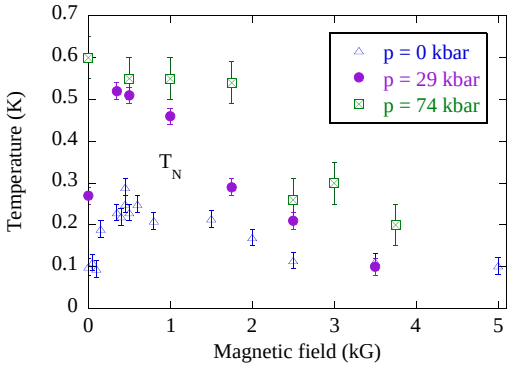


FIG. 4: (Color online) The behaviour of the antiferromagnetic peak as a function of magnetic field at p = 0, 29 and 74 kbar. In the two lower pressure measurements (open triangles and squares respectively) the behaviour of T_N with field shows an initial increase until B = 1 kG, then the transition temperature falls with increasing field. The data taken at p = 74 kbar shows that the transition temperature continuously falls as magnetic field is increased.

pressure up to 14.6 kbar by four point resistivity [15]. A selection of these results are shown in Fig. 2. It is clearly seen that as the pressure is increased the field required to cause reentrance reduces. Along with this the superconducting transition temperatures for both entry into superconductivity, and reentrance to the normal state, reduce with pressure.

A lambda-like peak in the magnetic susceptibility has been attributed to the onset of antiferromagnetic order, T_N, which has been observed elsewhere [8], [10], [5]. In this work, at ambient pressure this was measured to occur at 0.12 K in both unpressurised moissanite anvil cells and in standard ambient pressure balanced coils. The value for T_N at ambient pressure increases to approximately 0.25 K in a magentic field of 500 G. This compares to a temperature of 0.22 - 0.23 K in zero magnetic field in other work [5, 8]. It is speculated here that the discrepancy made be due to differences in the crystals measured. The motion of the antiferromagnetic transition as a function of pressure may also be observed in Fig. 3. At ambient pressure $T_N = 0.12$ K, and rises monotonically to reach $T_N = 0.58$ K at p = 74 kbar. Figure 3, suggests that the gradient of the increase in the transition temperature with pressure shows the hint of decreasing at the higher pressures. Further measurements at higher pressures would be required to confirm this idea.

The relative magnitude of the antiferromagnetic peak of $YbPd_2Sn$ is shown in the inset of Fig. 3. A clear drop in the susceptibility signal at T = 4.9 K is due to the onset of superconductivity in the lead pressure gauge. The temperature of this transition indicates that the measurement is at a pressure of 64 kbar [13]. The $YbPd_2Sn$ has a volume approximately 12 times greater than the lead in this measurement.

Measurements of the antiferromagnetic transition were taken in applied magnetic fields of 0 - 5 kG. In the lower pressures measured it was observed that the peak in the magnetic susceptibility goes to a higher temperature in magnetic fields of $\sim$ 500 G - 1 kG, to then be gradually suppressed. This is demonstrated in Fig. 4. This behaviour changed noticeably at higher pressures. Instead of having an increase in the Néel temperature as a function of field, the Néel temperature continuously decreased.

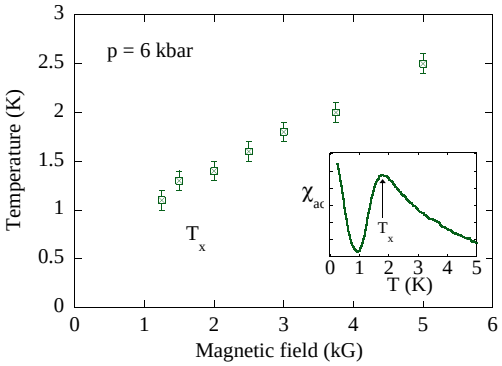


FIG. 5: (Color online) Main: The motion of the unidentified transition labelled 'T_x' as a function of magnetic field at p = 6 kbar. Inset: Raw ac magnetic susceptibility data showing the shape of T_x. The sharp increase below T = 1 K is due to the residue of the lambda-like peak of the antiferromagnetic transition. T_x has been attributed to the peak at T = 1.8 K.

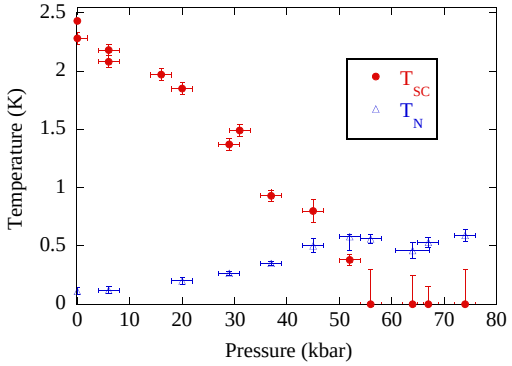


FIG. 6: (Color online) The phase diagram of $YbPd_2Sn$ in zero magnetic field. The superconducting transition temperature is shown as T_{SC}, and the antiferromagnetic transition is shown by T_N.

An additional feature was observed in the magnetic susceptibility signal at all pressures above ambient pressure. An example of this feature is shown in the inset of Fig. 5. This data was taken in an applied magnetic field of 3 kG. To the far left of the figure, the slope is due to the residue of the lambda-like antiferromagnetic transition. The new feature was the peak that is clearly visible at T = 1.8 K. This peak was normally observed initially in magnetic fields $\sim$ 1.5 kG. It is perceived to exist up to B $\sim$ 5 kG. At p = 6 kbar, the new peak is observed to occur at higher temperature as the magnetic field is increased. This is shown in Fig. 5. The transition was observed in all of the samples that were pressurised in a moissanite anvil cell, and in a sample measured by ac susceptibility in a piston cylinder cell. Often the signal for this transition was fairly weak, and thus difficult to track. At all pressures measured between p = 6 – 74 kbar, in B = 2.5 kG, the transition occurred at T_x = 1.6 – 1.8 K. With magnetic field the transition moved to higher temperature in a similar way to the data shown at p = 6 kbar. The type of transition that this feature may be attributed to is unknown thus far.

IV. CONCLUSIONS AND DISCUSSION

The application of pressure to $YbPd_2Sn$ causes the superconducting transition temperature to be suppressed, such that the transition appears to occur at zero temperature at p $\geq$ 58 kbar. The superconducting reentrance is also forced to lower temperatures and magnetic fields as pressure is increased. Magnetic susceptibility measurements at pressures between 58 kbar and 74 kbar do not indicate any further evidence of superconductivity above T = 0.2 K.

By considering the critical magnetic field associated with superconductivity, a simple estimate [16] gives the coherence length of this compound to be $\sim$ 600 $\AA$. Further, using the residual resistivity, it is possible to estimate the mean free path [17] to be $\sim$ 200 $\AA$. As the mean free path is found to be significantly smaller than the coherence length, it is unlikely that the superconductivity is heavy fermion in nature. This does not, however, mean that the superconductivity is necessarily of a standard BCS nature. One theory presented by Machida and co-workers [11] suggests that to explain the reentrant superconducting behaviour, a more stable ground state may involve a Q-dependent superconducting order parameter in the antiferromagnetic state [6].

The antiferromagnetic transition temperature of $YbPd_2Sn$ is enhanced as a function of pressure, starting at 0.12 K at ambient pressure and rising to 0.58 K at p = 74 kbar. This fact is interesting because neutron data at ambient pressure suggests that at T = 0.6 K a small magnetic moment is observed, which increases significantly as the temperature reduces to $\sim$ 0.1 K, at which point the moment doesn't increase much further [18].

The phase diagram of $YbPd_2Sn$ in zero magnetic field is shown in Fig. 6. It is interesting that superconductivity survives in the presence of strong magnetic fluctuations in the temperature range $T_C \geq T \geq T_N$. In this temperature range the long range magnetic order has not fully set in, but the magnetic fluctuations will be significant. This is thought to be because

the Yb sublattice, responsible for magnetism, is decoupled from the Pd sublattice, which carries the superconducting electrons. This idea is corroborated by band structure calculations undertaken by Nevidomskyy [19].

To address the question of the lack of heavy fermion superconductivity in Yb based compounds, one may consider the magnitude of T_N in these materials. The data shown here for $YbPd_2Sn$ suggests that T_N reaches a peak at around T = 0.6 K or slightly higher. Compare this with the situation in $YbRh_2Si_2$, which displays a maximum magnetic ordering temperature of around T = 1 K at p = 40 kbar [20]. The analogous Ce based compound is $CeRh_2Si_2$. This compound has an antiferromagnetic transition at p = 0 kbar at $T_N = 36$ K [21]. As the strength of the relevant RKKY magnetic coupling constant, J_{RKKY}, is related to the maximum value of T_N (as in the Doniach phase diagram [22]), the measurements of $\mathbf{R}Rh_2Si_2$ ($\mathbf{R}$ = Yb, Ce) suggest that the RKKY coupling constant in Yb systems may be lower than in Ce systems. This is consistent with Yb systems having nearly local moment magnetism. This is supported by the data in this work, showing the possibility of another Yb compound with a low T_N maximum value with pressure. As heavy fermion superconductivity relies on a larger value of J_{RKKY} [23], the lower J_{RKKY} values in Yb compounds may account for the absence of superconductivity in the temperature and compound purity ranges currently possible experimentally.

Evidence in the ac magnetic susceptibility data for a possible new transition in $YbPd_2Sn$ has been found at p $\geq$ 6 kbar and in magnetic fields between $\sim$ 1.5 kG and 5 kG. The nature of this transition is not known.

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