Ultra-low temperature ac susceptibility of the heavy-fermion superconductor YbRh$_2$Si$_2$

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Abstract. Recently, we reported on the discovery of superconductivity in YbRh$_2$Si$_2$ at the very low temperature $T_c = 2$ mK. We present here complementary measurements of the temperature and field dependence of the ac susceptibility $\chi_{ac}(T)$ of this heavy-fermion system for temperatures down to 1 mK and magnetic fields in the basal plane up to 75 mT. The in-phase response $\chi_{ac}(T)$ shows a steep drop at $T_c$, but it does not become negative, possibly because of the presence of the earth field that was not compensated. The out-of-phase response $\chi_{ac}(T)$ shows an increase right below $T_c$ indicating that the transition is first order. We also observe two kinks in the field dependence of $\chi_{ac}(B)$ at $B_1 \approx 50$ mT and $B_2 \approx 60$ mT, which imply a more complex $T - B$ phase diagram for YbRh$_2$Si$_2$ with $B \parallel c$. These features possibly indicate two second-order phase transitions or domain reorientation at $B_1$ and a phase transition from the antiferromagnetic to the paramagnetic state at $B_2$.

1. Introduction

YbRh$_2$Si$_2$ is a canonical quantum critical heavy-fermion (HF) compound [1, 2]. It is characterized by a Kondo temperature $T_K = 25$ K and antiferromagnetic (AF) order below $T_{AF} = 70$ mK with Yb-derived ordered moments of the order of $10^{-3} \mu_B$ [3, 4, 5]. The AF phase can be continuously suppressed by a magnetic field applied within the basal tetragonal (“easy magnetic”) plane. The critical field in the basal plane was estimated to be $B_N \approx 50 - 60$ mT [4, 6]. At $B_N$ isothermal magnetotransport [7, 8] and thermodynamic measurements [4, 6] have provided evidence that an unconventional quantum critical point (QCP) exists, but no superconductivity was found down to 20 mK, which is typically observed in several Ce-based HF compounds [9, 10, 11, 12, 13, 14].

Recently, we reported on the discovery of superconductivity in this system at the very low temperature of 2 mK [16]. YbRh$_2$Si$_2$ is only the second Yb-based HF superconductor, following $\beta$-YbAlB$_4$ with $T_c = 80$ mK [17]. A $T_c$ of 2 mK is much smaller than the typical $T_c$ of Ce-based HF superconductors ($\lesssim 2.5$ K). Such a ratio is also found for the magnetic ordering temperatures, e.g., $T_{AF} = 70$ mK for YbRh$_2$Si$_2$ compared to $T_{AF} \lesssim 4$ K in CeRhIn$_5$; this can be attributed to the much smaller radius of the 4f electrons in Yb$^{3+}$ compared to that in Ce$^{3+}$ [18].
addition, we could identify a rich $T - B$ phase diagram for YbRh$_2$Si$_2$ (with $B \perp c$), by means of magnetisation, susceptibility and heat capacity measurements. This phase diagram has a new A + SC phase below $T_A \gtrsim 2$ mK which includes superconductivity. In addition, there is evidence for strong AF (A-phase) fluctuations below $T_B \simeq 10$ mK. While the AF phase is of purely electronic nature, the new A phase involves a coupling between the magnetic moments due to the Yb-derived 4$f$ electrons and the Yb nuclear moments. The size of the critical field $B_A$ at which $T_A \to 0$ (30–60 mT) indicates that the A phase must be dominantly of nuclear origin. This led us to conclude that the superconductivity develops from quantum critical fluctuations associated with the disappearance of the AF order induced by the nuclear spin order.

Here, we present new complementary field-dependent measurements of the ac susceptibility $\chi_{ac}(B)$ taken at constant temperatures (down to 11 mK) and in fields as high as 75 mT. We have discovered new features in $\chi_\prime_{ac}(B)$ in form of kinks at $B_1 \approx 50$ mT and $B_2 \approx 60$ mT, which point to a more complex $T - B$ phase diagram for YbRh$_2$Si$_2$ with $B \perp c$.

Figure 1. Temperature dependence of the ac susceptibility of YbRh$_2$Si$_2$ measured in earth magnetic field and earth magnetic field plus 28 mT. With this setup it was possible to measure temperature sweeps from 1 mK up to about 1000 mK and scale the data with those taken in a standard Kelvinox 400 dilution refrigerator (Oxford Instruments) down to 20 mK (black points) [15]. We observe a kink when entering the AFM phase at $T_{AF} \approx 80$ mK, a weak signature at $T_B$, i.e. at a crossover into the regime with A-phase fluctuations, and a strong drop of $\chi_{ac}(T)$ at the superconducting transition temperature $T_c = 1.9$ mK (onset). Here, $\chi_{ac}(T)$ shows a clear increase, suggesting that the transition is of the first order.
2. Experimental setup

We have carried out our experiments in the ultra-low-temperature adiabatic demagnetisation refrigerator (ADR) at the Walther Meissner Institute in Garching. It consists of a 0.9 mole PrNi$_5$ nuclear demagnetisation stage with a final temperature of 0.4 mK. The temperature of the nuclear stage was determined by a simulated warm-up curve, which was verified to be in excellent agreement with temperatures obtained by pulsed nuclear magnetic resonance on $^{63}$Cu and $^{195}$Pt nuclei. This method results in a temperature accuracy of 2 – 3% even down to 0.8 mK (Ref. [16]). Temperatures above 10 mK were measured with a resistive thermometer.

A 5.44 mg YbRh$_2$Si$_2$ single crystal of very high quality (RRR ≈ 50) was used in our experiments. This thin YbRh$_2$Si$_2$ platelet was clamped on a 5N silver wire attached to a 5N silver rod which by itself was screwed to the nuclear stage. The crystal was placed in the pick-up coil of the susceptometer with its flat surface (the basal tetragonal plane) aligned parallel to the coil axis. Since the electrical conductivity of the samples is high (of the order of 5000 S) we have no indication that, after thermalisation, its temperature deviated from that of the nuclear stage by more than a few tens of µK, even at the lowest temperatures. Unfortunately, heating the sample to temperatures above 600 mK resulted in a thermal drift of the signal due to a warm-up of the mixing chamber of the dilution refrigerator, making measurements unreliable at higher temperatures.

The ac susceptibility $\chi_{ac}(T)$ was measured by a conventional mutual inductance setup thermally coupled to the nuclear stage and mounted in the centre of a superconducting 8 T magnet. The left and right pick-up coils consist of 18 (1694 windings) and 19 (1786 windings) layers, respectively, of 5 µm copper wire, with a total resistance of 1176 kΩ at 300 K. For the excitation coil we used 3 layers (761 windings) of 0.1 mm superconducting NbTi wire in CuNi matrix which had a resistance of 1.439 kΩ at 300 K. The susceptometer was balanced systematically at 300, 77 and 4.2 K. The in-phase $\chi_{ac}'(T)$ and out-of-phase $\chi_{ac}''(T)$ responses were measured with a two-channel lock-in amplifier. This allowed frequencies up to a few hundred Hz. Here the smallest field was the earth field: According to the international geomagnetic reference field (IGRF), the vertical component of the earth field at the location of the laboratory is -0.0433 mT. In Ref. [16] we presented a measurement of $\chi_{ac}(T)$ obtained by modulating a rf SQUID system at which the earth field could be compensated with an extremely small dc field between 2 and 5 nT. This allowed us to see negative values of $\chi_{ac}'(T)$ below $T \approx 2$ mK which are not observed in the measurements presented here.

3. Results

Fig. 1 shows the temperature dependence of the in-phase $\chi_{ac}'(T)$ and out-of-phase $\chi_{ac}''(T)$ responses of the ac susceptibility measured in earth magnetic field and at 28 mT (with $B \perp c$). The measurements were taken with increasing temperature. The conventional mutual inductance setup allowed us to measure $T$-sweeps from 1 mK up to about 1 K and to scale the data with those taken in a standard Kelvinox 400 (Oxford Instruments) down to 20 mK (black points in Fig. 1) [15]. We used an excitation field of 2.5 µT and a frequency of 117 Hz. With higher excitation fields it was not possible to cool the sample to below 10 mK (see, e.g., magenta points in Fig. 1).

The results observed here are very similar to those seen in other and better crystals investigated in Ref. [16]: In particular, we observe i) a clear kink in $\chi_{ac}'(T)$ at $T_{AF} \approx 80$ mK when entering the AF phase, ii) a weak signature at $T_B$, i.e. at the crossover into the regime with A-phase fluctuations and iii) a strong drop of $\chi_{ac}'(T)$ at the superconducting transition temperature $T_c \approx 1.9$ mK. Although the drop in $\chi_{ac}'(T)$ is quite large we do not reach negative values for $T \rightarrow 0$, but the susceptibility saturates at about $3.5 \times 10^{-6}$ m$^3$/mol. With this setup it was not possible to screen the earth magnetic field, and this might be the reason why the superconducting transition is not manifested by negative values. We tried to use a µ-metal...
Figure 2. Isothermal field sweeps of the ac susceptibility $\chi'_{ac}(B)$ of YbRh$_2$Si$_2$ taken at selected temperatures. The data are plotted with those taken in a standard Kelvinox 400 dilution refrigerator (Oxford Instruments) at 20 and 65 mK (black empty points) on a sample of similar quality [20]. The fields $B_1$ and $B_2$ mark the positions of kinks in $\chi'_{ac}(B)$ (observed only at 11 and 13 mK) which possibly indicate phase transitions of the second order since $\chi''_{ac}(T)$ is not showing any feature at those fields. However, domain orientation might also be possible at $B_1$.

At higher temperatures $T < T_{AF}$ we observe a single kink at $B_1$ which decreases with increasing $T$ as previously observed.
Figure 3. Extended phase diagram of YbRh$_2$Si$_2$ [16]. We added new points and a new phase boundary line (magenta) inferred from the measurements in this work. PM and AF indicate the paramagnetic and antiferromagnetic states, respectively. The hatched light-blue area indicates the onset of A-phase fluctuations (phase B) which give rise to a reduction of the staggered magnetisation and the beginning of shielding due to superconducting fluctuations. The A + SC phase represents the concurring (dominantly) nuclear AF order and superconductivity. Only at $B = 0$ almost full shielding is observed. The two red dashed lines mark the range within which the A-phase boundary line may end. The green circle indicates the superconducting transition temperature seen in the $ac$ susceptibility at $B = 0$ in Ref. [16] while the yellow circles (partially covered by the green point) result from the shielding signals in the zero-field-cooled dc magnetisation. In the inset, these shielding transitions are shown separately on an enlarged scale. The superconducting transition temperature (onset) observed with earth field in our set-up is $T_c \approx 1.9 \text{ mK}$, slightly lower than that measured at $B = 0$.

at selected temperatures. The data are plotted with those taken in a standard Kelvinox 400 dilution refrigerator (Oxford Instruments) at 20 and 65 mK (black empty points) on a sample of similar quality [20]. Remarkably, the measurement obtained in the Kelvinox 400 at 20 mK overlaps quite well with that obtained in our ADR at 45 mK and shows a single peak at $B_1 \approx 50 \text{ mT}$. This was interpreted as the critical field $B_N$ [20]: In fact, $B_1$ decreases with increasing $T$. However, at the lowest temperature of 11 and 13 mK we see two clear features (kinks) in $\chi''_{ac}(B)$ at the fields $B_1 \approx 50 \text{ mT}$ and $B_2 \approx 60 \text{ mT}$. They possibly indicate phase transitions of the second order since $\chi''_{ac}(T)$ is not showing any feature at those fields.

4. Discussion
The features observed in the field dependent ac susceptibility suggest the presence of an additional phase line in the high-field side of the AF phase. This is indicated by the magenta coloured points at $B_1$ that we added to the $T - B$ phase diagram of Ref. [16] (Fig. 3). The points at $B_2$ are surely related to the transition from the AF phase into the field-polarised state. Although phase diagrams can be extremely complex, a comparison with the parent compound YbCo$_2$Si$_2$ could help to elucidate the nature of this new line [22]. In the case of YbCo$_2$Si$_2$ the
Yb moments are fully localized and the magnetic anisotropy is about 10 times smaller than in YbRh$_2$Si$_2$, which makes the analysis of the magnetic order much more easier. The magnetic phase diagrams of YbCo$_2$Si$_2$ with magnetic field along the [100], [110] and [001] directions have been intensively investigated [23] and they are all different. Most importantly, in YbCo$_2$Si$_2$ there are some lines in the phase diagrams within the basal plane ($B \parallel [100]$ and [110]) which correspond to a depopulation of unfavoured AF domains, without modifying the propagation vector. On the other hand, phase lines which indicate a change of the ordering wave vector were observed to be first order. In our data we definitely see the transition at $B_1$ to be of the second order, and this might be just due to domain reorientation. Since we did not check the exact orientation of our samples - crystal platelets grow preferentially with one side parallel to the [100] or [110] direction - and because of a possible strong in-plane anisotropy in YbRh$_2$Si$_2$ as in YbCo$_2$Si$_2$, the phase diagrams with $B \parallel [100]$ might differ from that with $B \parallel [110]$, with slightly different critical fields of 50 and 60 mT. This could explain why measurements performed in our ADR at 13 mK are apparently different from those measured in the Kelvinox 400 at 20 mK.

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