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Physica B 378–380 (2006) 411–412
PHYSICA B
www.elsevier.com/locate/physb

Phase diagram of $ZrZn_2$ at high pressure: Low-temperature features and elusive superconductivity

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Abstract

Studies of the AC magnetic susceptibility and electrical resistivity of polycrystalline samples of $ZrZn_2$, synthesized at high pressure, were performed at pressures up to 4.5 GPa and temperatures down to 0.4 K. The evolution with pressure of the line of ferromagnetic phase transformations qualitatively agrees with numerous previous data, though the transition temperature is highly sensitive to the quality and history of samples. Upon approaching zero temperature, the transition line bends toward the pressure axis as dictated by the Nernst theorem. An additional feature of the phase diagram was discovered in the samples with the highest Curie temperature (25–26 K). The electrical resistance of these samples drastically decreases near 1.4–1.8 K at ambient pressure. The temperature of this resistive transition does not change much with pressure and crosses the Curie line at a pressure near 1.2 GPa, seemingly forming some sort of tetracritical point. Application of magnetic fields up to 2 T suppresses the transition that one may expect if superconductivity is involved. However, heat capacity measurements do not show any anomaly at the transition, which resembles the case described by Pfliederer et al. [Nature (2001) 58].

PACS: 74.70.Ad; 75.50.Cc; 74.25.Fy; 74.70.−b

Keywords: Phase transitions; Superconductivity

The phenomenon of coexisting ferromagnetism and superconductivity is now the subject of intensive experimental and theoretical efforts. The itinerant ferromagnet $ZrZn_2$ is one of a few materials that has been reported to become superconducting in the ferromagnetic state [1]. Two other known examples are UGe_2 under high pressure [2] and URhGe [3]. The superconducting state was reported to disappear in $ZrZn_2$ at the same pressure where magnetism is suppressed [1]. This claim stimulated the appearance of theoretical models that explained superconductivity of $ZrZn_2$ as p-wave triplet pairing mediated by spin fluctuations (see, for example [4]). A heat capacity anomaly at the superconducting transition in $ZrZn_2$ is absent, though it was found in UGe_2 and URhGe.

We prepared 30 polycrystalline samples of $ZrZn_2$ by melting a stoichiometric amounts of Zr (99.93%) and Zn (99.999%) at high pressure of 8 GPa, as described earlier [5]. Samples with composition of ZrZn and $ZrZn_3$ also were prepared under the same conditions. Most of the samples prepared have a rather broad ferromagnetic transition with the midpoint varying in the range 10–20 K. These samples do not show any sign of superconductivity down to 0.4 K. Three samples have a sharp ferromagnetic transition at 25–26 K comparable with single crystals [1]. All of them exhibit an appreciable drop of resistance starting at 1.4–1.8 K. A zero resistance state is not reached above 0.4 K, but magnetic field suppresses the resistive transition, as if superconductivity is involved (Fig. 1).

After annealing samples in vacuum at 1023 K for 29 h, the ferromagnetic transition becomes sharper and its midpoint temperature increases by 1–2 K, but a sign of

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0921-4526/$ - see front matter
doi:10.1016/j.physb.2006.01.144

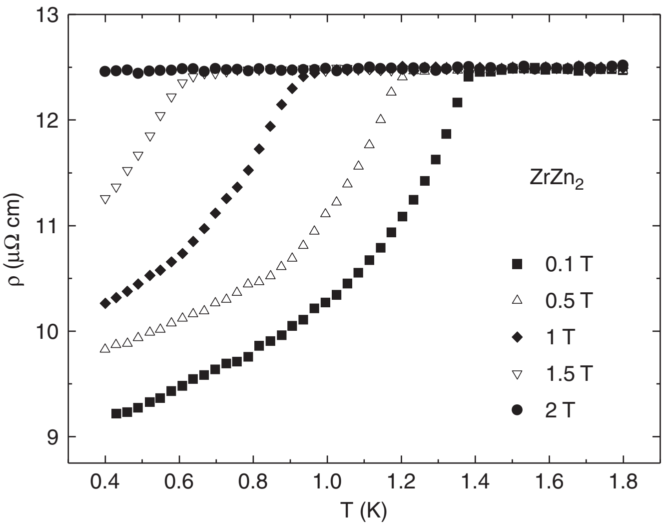

Fig. 1. Low-temperature resistivity of $ZrZn_2$ sample, having a Curie point $T_C = 25\,K$, as determined by AC susceptibility and Arroit plots in magnetization measurements.

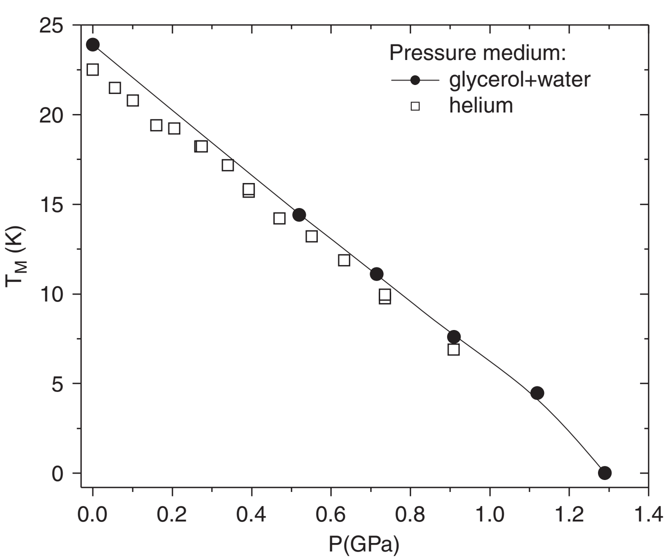

Fig. 2. Pressure dependence of the Curie point of $ZrZn_2$.

superconductivity disappears in all three samples. The sample preparation procedure [5] includes rapid cooling of the melt, and, taking into account very close ionic radii of Zr and Zn, it may result in partial site disordering. In this case, magnetism is suppressed as observed for most of the samples. But if site disorder is not pronounced, the d-band density of states of Zr at the Fermi level changes, favoring a superconducting state. Further site ordering on annealing destroys this metastable state, which exhibits superconductivity. At present we do not have a clue if there is any connection between this observation and indications for the surface superconductivity in $ZrZn_2$ as a result of cutting by the spark erosion [7].

To construct a *P–T* phase diagram, we have measured the electrical resistivity and AC susceptibility of $ZrZn_2$ at high hydrostatic pressures to 4.5 GPa using a toroidal anvil

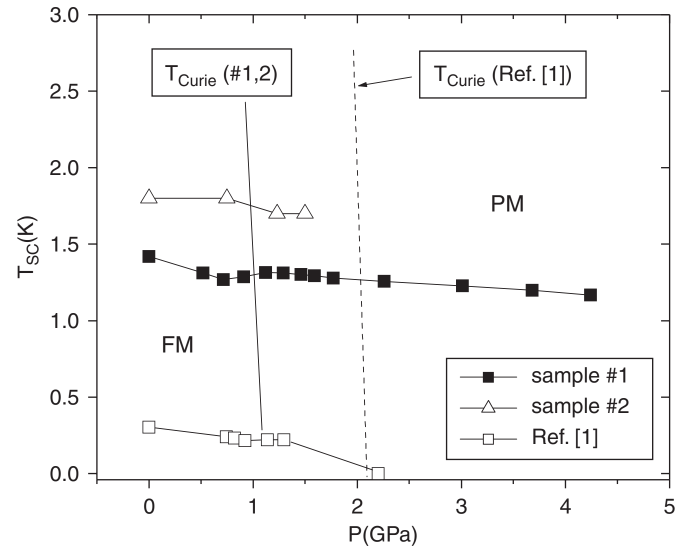

Fig. 3. Pressure dependence of superconducting transition temperature of $ZrZn_2$.

cell, and up to 1 GPa using a helium gas apparatus [6] (Fig. 2).

Pressure suppresses ferromagnetism in accordance with previous data (see [1] and references therein), and above ~1.3 GPa our samples remain paramagnetic. The disappearance of ferromagnetism has a negligible effect on superconductivity of $ZrZn_2$ (Fig. 3). This implies that superconductivity in $ZrZn_2$ is probably not mediated by ferromagnetic spin fluctuations as was proposed in Ref. [1] and subsequently in numerous other theoretical papers.

X-ray diffraction analysis revealed the presence of a few percent of ZrZn in our $ZrZn_2$ samples, exhibiting superconducting features. Resistivity measurements on ZrZn samples, prepared under the same high-pressure conditions, showed no evidence of a resistive transition of the type depicted in Fig. 1. So, an impurity phase of ZrZn is not the origin of superconductivity in $ZrZn_2$. Heat capacity measurements do not show an anomaly in the region of the resistive transition, similar to [1], suggesting that only a small fraction of the $ZrZn_2$ sample is superconducting.

This work was supported by the Russian Foundation for Basic Research (03-02-17119) and by special programs of the Department of Physical Sciences, Russian Academy of Sciences. Work at Los Alamos was performed under the auspices of the US DOE, Office of Science.

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