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Search for superconductivity of magnetic metals

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Abstract

A search for superconductivity of magnetic elemental metals is performed. A successful discovery of the onset of superconductivity is reported in the case of iron under pressure. By electrical resistance measurement, a maximum value of the superconducting transition temperature T_c of 2 K and the upper critical magnetic field H_c of 0.2 T are observed under pressure of 20 GPa where iron is in the crystallographic hcp phase and non-magnetic. Further confirmation of the superconducting transition of hcp iron was obtained by the detection of the diamagnetic signal due to the Meissner effect in accordance with the results of the electrical resistance measurements.

1. Introduction

Among various elemental metals, magnetic metals such as iron, nickel, cobalt and manganese are expected not to show superconductivity because of the pair breaking interaction between conduction electrons and the magnetic moment. However, it is also known that the magnetism disappears under certain high pressures.

In the case of iron, there exists a pressure-induced crystal phase transition under pressure between 10 and 14 GPa from the ferromagnetic bcc phase (α-Fe) to the non-magnetic hcp phase (ε-Fe). Therefore, we may expect the superconducting transition of Fe at a certain low temperature where the iron is non-magnetic [1]. This is predicted by theories [2, 3] but there have been no experimental reports on the observation of superconductivity of iron in its non-magnetic state.

We have searched for superconductivity in Fe, as well as Mn, over wide range of temperature and pressure, using a diamond-anvil cell (DAC) and a ^{3}He/^{4}He dilution refrigerator. Quite recently, we managed to detect the appearance of superconductivity of Fe by means of electrical resistance as well as magnetization measurement [4].

2. Experimental details

High-purity iron (from Johnson-Matthey), 5N, was purified further by heating up to the melting point in an ultrahigh-vacuum chamber. The purified iron was confined in a sample hole of

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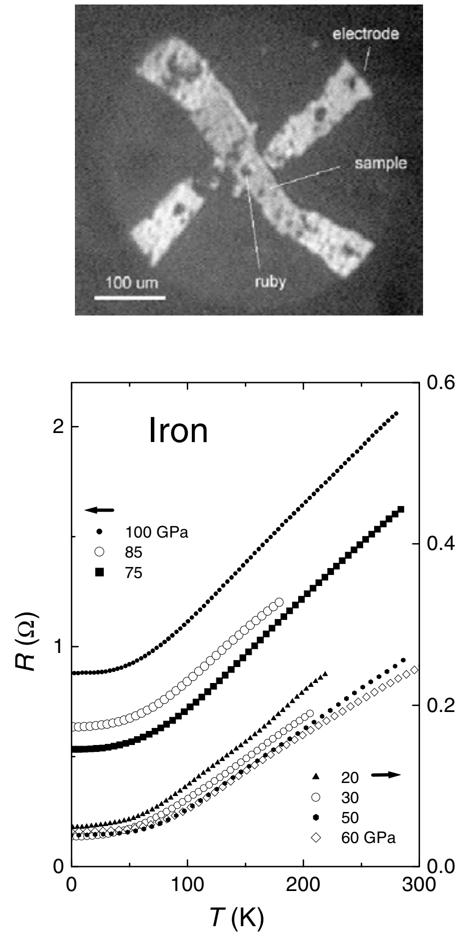

Figure 1. A photograph of the arrangement of the sample under pressure and the temperature dependence of the electrical resistance of iron under pressure. There is no sign of superconductivity in these runs with non-hydrostatic pressure.

an insulated BeCu metal gasket of DAC. NaCl was used as a pressure medium so as to avoid excess uniaxial stress on the sample iron. Four Pt electrodes were spot welded to the sample for electrical resistance measurement. The DAC was assembled on our powerful dilution refrigerator, which enabled us to produce complex extreme conditions of high pressure up to 200 GPa and low temperature down to 30 mK. For magnetization measurement, pick-up coils around the sample were connected to the standard SQUID magnetometer, which has enough sensitivity to detect the diamagnetic signal due to the possible superconducting transition of the extremely small amount of Fe.

We have also searched for the superconductivity of manganese under pressure in the case of stable α-Mn as well as quenched β-Mn. The magnetism of β-Mn at low temperature, especially under pressure, has not been studied in detail and in fact a high-purity and/or high-quality sample is hard to obtain. Results of electrical resistance measurements under pressure are not reliable in many cases from the viewpoint of reproducibility. On the other hand, in the case of α-Mn which is stable at ambient pressure, we were able to obtain reliable results.

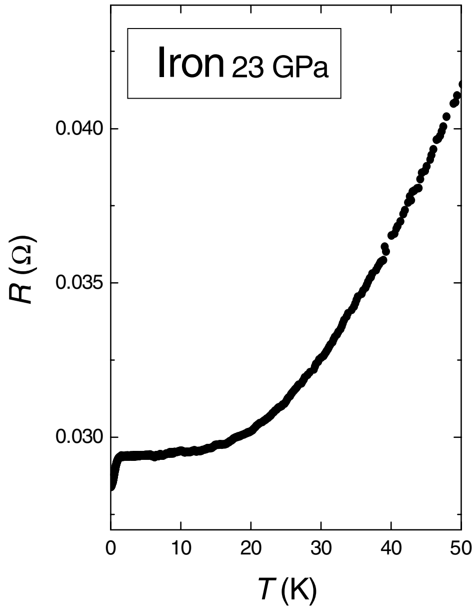

Figure 2. The electrical resistance versus temperature curve for iron at 23 GPa which shows a drop of the resistance at low temperature around 2 K.

Therefore we focused our attention on α-Mn, but we have not succeeded yet in purifying the sample as in the case of iron.

3. Experimental results

The ferromagnetic-to-non-magnetic phase transition has a large pressure hysteresis that depends very much on the quality of the applied pressure (e.g. the hydrostaticity). Also, high-pressure Mössbauer experiments report that the amount of remaining low-pressure-phase iron strongly depends on the hydrostaticity. It is expected that even a very small amount of remaining ferromagnetic phase may suppress the superconductivity. In fact, with non-hydrostatic conditions, no superconductivity has been detected in the course of increasing pressures up to 100 GPa (figure 1).

We studied the temperature dependence of the electrical resistance of Fe under various pressures exceeding 14 GPa and at temperatures down to 50 mK. We observed that the resistance decreases linearly with decreasing temperature in a relatively high-temperature region under observed pressures up to 100 GPa. At low temperatures below 5 K, the resistance value becomes constant just like for the typical elemental metals. The above-described behaviours were observed commonly in Fe, independently of which pressure media were used.

By detailed investigation of resistance at temperatures below 2 K, however, we were able to observe a small drop in electrical resistance as shown in figure 2. The amount of the drop was less than 10% of the total resistance but was found to be dependent on the external magnetic field and pressure. Further examination shows that the observed small drop is due to the superconducting transition of iron under pressure in its non-magnetic state. This is confirmed also by the detection of the Meissner effect, giving a diamagnetic signal in accordance with the resistance measurement. The results are summarized as follows.

The maximum T_c is observed at the pressure of 20 GPa and the superconducting state appears just after the crystal phase transition from the bcc to the hcp phase. T_c appears at 15 GPa and increases with increasing pressure up to 20 GPa where T_c takes the maximum value of 2 K, and then T_c decreases with increasing pressure and disappears at around 35 GPa. The maximum critical field H_c is 0.2 T at 20 GPa. The superconductivity has not been observed for non-purified iron in the cases with and without pressure media. For a purified sample, but without pressure media, we were able to observe a much smaller T_c of around 1 K under pressure of 20 GPa—and that was in the process of decreasing pressure after application up to 90 GPa.

The above results may show the importance, firstly, of purification of the sample and, secondly, of employing an appropriate pressure medium. Further examination and the establishing of the phase diagram of the superconducting state with respect to temperature and magnetic field are now proceeding.

We have also studied the high-pressure low-temperature properties of antiferromagnetic manganese. It has already been reported that antiferromagnetism is suppressed by applying pressure [5, 6]. We have also confirmed the pressure dependence of the Néel temperature, T_N, by electrical resistance measurement. Simple extrapolation of the T_N–pressure phase diagram shows that the antiferromagnetism disappears at around 2.5 GPa in accordance with previous experimental works. Therefore, we searched for the superconductivity of manganese in its non-magnetic state up to 6 GPa and for temperature down to 50 mK.

Up to now, however, we have not observed any sign of superconductivity in our electrical resistance measurements; instead, we observed an anomaly at 50 K which is pressure independent. At present, no interpretation is given for the origin of the observed anomaly. Both purification of the sample and employing appropriate pressure media appear essential.

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