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AC Microcalorimetry of Superconducting $MgCNi_3$ Single Crystals

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The low-temperature specific heat of single-crystal samples of superconducting $MgCNi_3$ with typical dimensions 200 μm were measured for the first time. A computer controlled ac microcalorimeter using an optical fiber and an infrared light-emitting diode as the heat source was used down to 2 K at magnetic fields up to 8 T. The specific heat data suggest a moderate coupling in $MgCNi_3$.

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1. Introduction

The low-temperature specific heat measurements of the single-crystalline samples of superconducting $MgCNi_3$ by ac microcalorimetry are presented. This highly sensitive technique is very well adapted to measure specific heat of very small samples (with typical dimensions of few hundred microns in our case) and to carry continuous measurements during either temperature or magnetic field sweeps.

Despite the strong expectation for ferromagnetism in $MgCNi_3$, it shows superconductivity rather than magnetism [1]. However, the superconducting origin of this material has not yet been clarified. The large ratio of Ni (60% in molar ratio)

makes $MgCNi_3$ an intriguing compound to investigate the possible magnetic interaction with superconductivity. There are experiments supporting s-wave [2, 3] but some other propose non-s-wave gap symmetry [4]. All the previous reports on the specific heat of $MgCNi_3$ polycrystals [1–3, 5, 6] show conventional phonon mediated pairing mechanism, though electron–phonon coupling constants from various groups were different. To resolve this controversial situation measurements on single crystals of good quality are highly desirable.

2. Experimental details

We have built an ac microcalorimeter which uses a diode as the heater. An optical fiber is used to drive the heating power onto the sample. The absence of a contact between the heater and the sample reduces total addendum. The temperature of the sample is measured by a thermocouple. The basics of the ac microcalorimetry technique consist of applying periodically modulated sinusoidal power and measuring the resulting sinusoidal temperature response. In the proper frequency regime, the heat capacity of the sample is inversely proportional to the amplitude of the temperature oscillations. The experimental method and instrument are similar to the ones described in [7].

It is extremely difficult to synthesize desired stoichiometry of $MgCNi_3$ even in polycrystalline samples. These difficulties were solved for $MgCNi_3$ single-crystals preparation using a high-pressure closed system. Details of the synthesis of the single crystals can be found elsewhere [8]. X-ray analysis proved that carbon deficiencies from stoichiometry are negligible. However, in contrast to polycrystalline $MgCNi_3$, which has usually local carbon deficiency, in these single crystals the Ni site was partly deficient. This leads to lower critical temperature from transport measurements $T_c \approx 6.7$ K [8] compared to the highest T_c of 7.3 K for polycrystalline samples.

We performed ac specific heat measurements of $MgCNi_3$ single crystals in the temperature range between 2 and 10 K and magnetic fields up to 8 T. The samples used for the measurements were with typical dimensions of few hundred microns.

3. Results and discussion

The samples have shown a well-defined superconducting transition in the specific heat. Figure 1a shows the low temperature total heat capacity of the sample plus addenda. Since our ac calorimetry technique only measures relative values, the measured heat capacity is given in arbitrary units. The anomaly in zero field is clearly visible, representing around 40% of the total signal which is much smaller than in polycrystalline samples. This indicates that addenda contribution is comparable to our crystal heat capacity. The anomaly is sharp ($\Delta T_c \approx 0.2$ K) indicating the high quality and homogeneity of our samples.

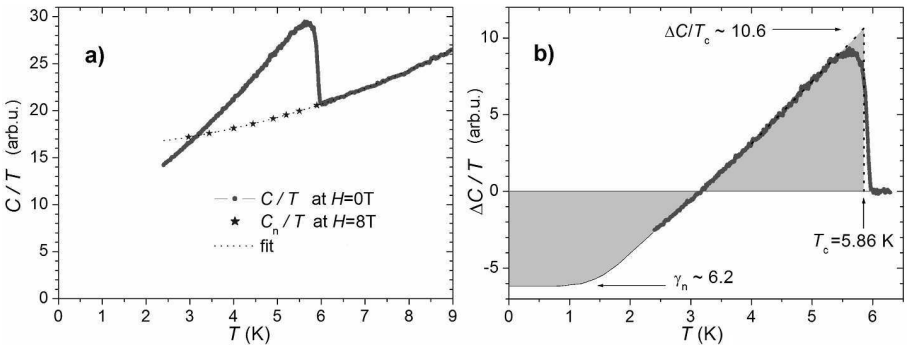

Fig. 1. (a) Total heat capacity of $MgCNi_3$ sample at zero field together with the background including the normal state specific heat data C_n for field of 8 T. (b) Electronic specific heat data in the superconducting state. The dotted lines are determined by conservation of entropy around the anomaly.

To clarify superconductivity in $MgCNi_3$, it is of interest to derive the electronic specific heat by subtracting the normal state specific heat C_n i.e. $\Delta C(T)/T = C(T)/T - C_n(T)/T$. Magnetic field of 8 T suppresses superconductivity down to 3 K — and in the temperature range from 3 to 9 K, $C(8\ \mathrm{T})$ follows very well the behavior (see Fig. 1a): $C(8\ \mathrm{T})/T = C_n/T = A + BT^2$. This formula can then be safely extrapolated to the whole temperature range of the measurements.

The resulting electronic specific heat at $H = 0$ is shown in Fig. 1b. From an entropy conserving construction (see the dotted line in Fig. 1b), the transition temperature T_c was estimated to be 5.86 K. We remark that T_c values deduced from specific heat are lower than those deduced from transport measurements on the samples of the same batch [8].

It is known that the parameter $\Delta C(T_c)/\gamma_n T_c$ (γ_n is the Sommerfeld coefficient) can be used to measure the strength of the electron coupling. Since we do not have specific heat data at lower temperatures than 2.4 K we cannot determine γ_n directly. To estimate γ_n we made a construction of $\Delta C(T)/T$ down to zero temperature obeying the entropy conservation law, demanding that two shaded areas must have equal surfaces (see Fig. 1b). The obtained ratio of $\Delta C(T_c)/\gamma_n T_c$ estimated from our data is approximately 1.71 which is larger than the BCS weak coupling limit (1.43). Moreover, to estimate the coupling strength of $MgCNi_3$ we compared measured dependence of $\Delta C(T)$ with a so-called alpha model [9] based on the BCS theory. In this model the only adjustable parameter is the gap ratio $\Delta(0)/k_B T_c$. Our comparison yields a ratio $2\Delta(0)/k_B T_c \approx 3.8$, which is again higher than the ratio ≈ 3.52 for the BCS weak coupling limit. These values suggest that the $MgCNi_3$ may be a moderate-coupling superconductor.

4. Conclusion

We have performed specific heat study of $MgNi_3$ single crystals by means of highly sensitive ac microcalorimetry. We found the critical temperature of the system to be 5.86 K. The specific heat measurements give $\Delta C(T_c)/\gamma_n T_c \approx 1.71$ and $2\Delta(0)/k_B T_c \approx 3.8$ suggesting that $MgCNi_3$ may be a moderate-coupling superconductor.

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