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Specific heat of superconducting MgCNi$_3$ single crystals

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Abstract. The ac microcalorimetry measurements with light emitting diode used as a heater has been performed on high quality MgCNi$_3$ single crystals with $T_c$ of 6.9 K. The measurements have been realized in the temperature range down to 0.7 K and magnetic fields up to 8 T. All the measurements show sharp and well defined specific heat anomaly at the transition. The low-temperature electronic specific heat in superconducting state shows a classical exponential decrease confirming an s-wave pairing. Analysis of the data in the framework of classical BCS theory points to the moderate coupling in this material. The upper critical field $H_{c2}$ has also been derived from specific heat measurements in different magnetic fields. The temperature dependence of $H_{c2}$ is discussed.

1. Introduction
Even though MgCNi$_3$ consists of large amount of Ni atoms it clearly shows superconductivity [1] rather than ferromagnetism. That makes it a good candidate for unconventional superconductivity. However there is a lot of discrepancy in results of different experimental techniques for polycrystalline MgCNi$_3$. The specific heat [2, 3] and tunneling spectroscopic studies [4] have shown the s-wave BCS-type superconductivity while the penetration depth measurements exhibit a non-s-wave behavior [5]. Recently the two-band model has been proposed to explain consistently different properties of this material. To resolve this controversial situation single crystals of good quality are highly desirable. Here we present a detailed study of high quality MgCNi$_3$ single crystals. We performed specific heat measurements in the temperature range down to 0.7 K and in magnetic fields up to 8T. The superconducting transition in zero magnetic field was detected at 6.85 K. The electronic specific heat in superconducting state was inspected closely down to the lowest temperatures in order to find out the strength of coupling in this superconductor. The height of the specific heat anomaly at critical temperature and exponential decrease of the data with the temperature point to a moderate coupling in this system with the ratio $2\Delta(0)/k_B T_c \approx 3.9$. Moreover we present the temperature dependence of the upper critical field $H_{c2}$ which follows a classical Werthamer-Helfand-Hohenberg (WHH) behavior.
2. Measurements

Recently the long-standing problems of MgCNi$_3$ single-crystals preparation have been finally overcome. The samples were fabricated in a high-pressure closed system. Details of the synthesis can be found elsewhere [6]. Using an X-ray micro analyzer it was proved that carbon deficiencies in 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 $T_c \approx 6.9$ K [6] compared to the highest $T_c \approx 7.3$ K of polycrystalline samples.

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]. In our case an optical fiber is used to guide the heating power emitted from the diode towards the sample. Absence of a contact heater reduces the total addendum to the total specific heat. The temperature of the sample is recorded by a sensitive thermocouple. A precise in situ calibration of the thermocouple in magnetic field was obtained from measurements on ultrapure silicon. The magnetoresistence of the Cernox thermometer was precisely inspected and corrections were included in the data treatment. Although an ac calorimetry is not capable to measure the absolute values of the heat capacity it is very sensitive technique for measurements of relative changes on minute samples.

3. Results and discussion

Figure 1 shows the low temperature total heat capacity of the sample plus addenda in zero magnetic field measured in temperature range down to 0.7 K and in the normal state (part of 8 T measurement) down to 4 K. Dashed line is extrapolation of the normal state background. Since our ac calorimetry technique only measures relative values, the measured heat capacity is given in arbitrary units. Specific heat in zero magnetic field shows a well-defined superconducting transition and relatively small total addendum.

![Figure 1](image1.png)  
**Figure 1.** Total specific heat of the sample plus addendum measured in zero magnetic field and 8 T. Dashed line is calculated normal state background.

![Figure 2](image2.png)  
**Figure 2.** Electronic specific heat in zero magnetic field. Dashed line is entropy conservation construction around critical temperature.
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. \[ \Delta C(T)/T = C(T)/T - C_n(T)/T. \] At low temperatures $C_n(T)/T$ can usually be described as $a\gamma_n + bT^2$. In case of MgCNi$_3$ higher order term is necessary to be considered - normal state data can be consistently fitted with the formula $C_n(T)/T = a\gamma_n + bT^2 + cT^4$. Since magnetic field of 8 T suppresses superconductivity in MgCNi$_3$ only down to 4 K we used this formula to calculate the normal state specific heat in order to get a background in the whole temperature range. Figure 2 represents the resulting electronic specific heat in zero field after subtraction of normal state background. The anomaly at the transition is sharp ($T_c \approx 0.2$ K) indicating the high quality and homogeneity of our samples. The transition temperature from entropy conservation construction around anomaly (dashed line in the figure 2) is 6.85 K. The low temperature electronic specific heat shows a classical temperature dependence as expected for a $s$-wave superconductor. The entropy conservation rule is well satisfied.

It is known that the parameter $\Delta C(T_c)/\gamma_n T_c$ ($\gamma_n$ is the Sommerfeld coefficient) can be used to measure the strength of the electron coupling. Taking the height of the jump at anomaly 8.41 and $\gamma_n \approx 4.3$, both in arbitrary units, we obtained ratio of $\Delta C(T_c)/\gamma_n T_c$ approximately 1.96 which is larger than that for the BCS weak coupling limit (1.43).

Moreover, to estimate the coupling strength of MgCNi$_3$ we compared measured dependence of $\Delta C(T)/T$ with a so-called alpha model [8] based on the BCS theory. In this model the only adjustable parameter is the gap ratio $2\Delta(0)/k_BT_c$. Our comparison yields a ratio $2\Delta(0)/k_BT_c \approx 4$, which is again higher than the ratio 3.52 for the BCS weak coupling limit.

**Figure 3.** Specific heat anomaly measured in magnetic fields 0, 0.1, 0.2, 0.3, 0.5, 1, 2, 3, 4, 5, 6, 7 and in 8 Tesla. Dashed line corresponds to the normal state specific heat as calculated from the formula $C_n(T)/T = a\gamma_n + bT^2 + cT^4$.

**Figure 4.** Temperature dependence of the upper critical field in MgCNi$_3$ from specific heat measurements. Dashed line is corresponding temperature dependence of $H_{c2}$ from WHH theory.

Figure 3 displays specific heat measurements in magnetic fields up to 8 T, all showing a well resolved and sharp anomaly gradually shifted to lower temperatures with increasing magnetic field. Dashed line corresponds to the normal state background calculated as described above. After subtraction of the normal state specific heat we determined values of the critical temperature for the measurement in each magnetic field. Figure 4 shows resulting temperature dependence of the upper critical field in MgCNi$_3$. It shows a linear increase close
to critical temperature and gradual deviation from linearity in low temperature range. It can be satisfactorily described in the frame of WHH theory represented by dashed line in figure 4. Within this theory $H_{c2}(T=0 \text{ K})$ can be estimated from the slope of $H_{c2}$ temperature dependence close to critical temperature which leads to value of 12 T in our case. The temperature dependence of $H_{c2}$ does not show any positive curvature contrary to expectation of magnetic fluctuations. Absence of deviation from linearity in the temperature range close to $T_c$ indicates that magnetic fluctuations in MgCNi$_3$ do not play an important role.

4. Conclusions

We performed ac calorimetry measurements on MgCNi$_3$ single crystals of very good quality. We found a well resolved superconducting transitions in zero field measurements as well as in measurements in magnetic fields up to 8 T. The low temperature electronic specific heat clearly shows a classical exponential decrease which is a strong indication for s-wave superconductivity in MgCNi$_3$. We found the coupling ratio $2\Delta(0)/k_B T_c \approx 4$ and reduced specific heat jump at the transition in zero field $\Delta C(T_c)/\gamma_n T_c \approx 1.96$ being higher than that for BCS weak coupling limit. Presented temperature dependence of $H_{c2}$ clearly shows a classical WHH behavior with absence of any positive curvature close to $T_c$ and can possibly rule out the magnetic fluctuations in MgCNi$_3$. The extrapolation of upper critical field to the lowest temperatures gives the value $H_{c2}(T=0 \text{ K}) \approx 12 \text{ T}$. In conclusion MgCNi$_3$ is a classical s-wave superconductor with moderate coupling.

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