## $H<sub>c2</sub>$  of MgCNi<sub>3</sub> Determined From the Specific **Heat and Resistivity Measurements**

## **J.-Y. Lin,<sup>1</sup> P. H. Lin,<sup>1</sup> P. L. Ho,<sup>2</sup> H. L. Huang,<sup>2</sup> Y.-L. Zhang,<sup>3</sup> R.-C. Yu,<sup>3</sup> C.-Q. Jin,<sup>3</sup> and H. D. Yang<sup>2</sup>**

 $H<sub>c2</sub>$  of MgCN<sub>13</sub> has been determined from the specific heat *C* and resistivity  $\rho$  measurements *in the same sample*. The results from  $\rho$  are nearly identical with those determined from the anomaly in *C*. Furthermore, utilizing the relation  $\gamma(H) \propto H$  and the value of  $d\gamma/dH$ , the obtained value of  $H_{c2}$  is the same as that by the WHH model, if the spin paramagnetic effect and the spin–orbit interaction are taken into account. The results of this comparison have strong implications on the order parameter of MgCNi<sub>3</sub>.

**KEY WORDS:** MgCNi<sub>3</sub>; *H*<sub>c2</sub>; specific heat; order parameter; mixed state.

Very recently, several new superconductors with possible ferromagnetic spin fluctuations have been reported  $[1-3]$ . Among them, MgCNi<sub>3</sub> has highest  $T_c \leq 8$  K. MgCNi<sub>3</sub> can be regarded as fcc Ni with only one quarter of Ni replaced by Mg and with *C* sitting on the octahedral sites. With the structure so similar to that of ferromagnetic Ni, the occurrence of superconductivity in  $MgCNi<sub>3</sub>$  is really surprising. Supposed that ferromagnetic fluctuations exist in a superconductor, *p*-wave order parameter is generally expected. Therefore, MgCNi<sub>3</sub> could have been a *p*wave superconductor with highest  $T_c$ . Accordingly, there has been theoretical speculation about *p*-wave pairing in MgCNi<sub>3</sub> [4]. Specific heat  $(C)$  [5] and NMR experiments [6], however, provided evidence of the full energy gap (possibly due to s-wave pairing) in MgCNi3. On the other hand, there were tunneling spectra suggesting an unconventional pairing state [7].

The upper critical field  $H_{c2}$  is one of the important fundamental properties of a type II superconductor. It is related to important parameters like the coherence length  $\xi$  and many magnetic properties. The value of  $H<sub>c2</sub>$  is also one of the crucial criteria for applications of the superconducting magnets.  $H_{c2}$  of MgCNi<sub>3</sub> was reported in Ref. 8 by resistivity ( $\rho$ ) measurements. *H*<sub>c2</sub> can also be deduced from the magnetic field dependence of *C*. It is of interest to see if the values of  $H_{c2}$ by distinct techniques are consistent with each other. Moreover, the magnetic field dependence of the linear coefficient  $\gamma$  in *C* from the electronic contribution was reported to be proportional to  $H$  in MgCNi<sub>3</sub>, and this was taken as one of the strong evidences for *s*wave pairing [5]. This conclusion in principle could be further examined by the relevant data of  $H_{c2}$ . In this paper, both *C* and ρ were measured *using the same sample*. This would allow eliminating any possible uncertainty from individual samples leading to more conclusive results.

The MgCNi<sub>3</sub> sample was prepared on the basis of procedure described in Ref. 1, and the X-ray diffraction pattern revealed the nearly single phase of MgCNi<sub>3</sub> structure. It is well known that  $T_c$  significantly depends on the real carbon content in the nominal  $MgCNi<sub>3</sub>$  [1,9]. Magnetization, specific heat, and resistivity measurements all showed a superconducting onset at about 7 K in the present sample, while thermodynamic  $T_c$  determined from  $C(T)$  was 6.4 K.  $C(T)$  was measured using a <sup>3</sup>He thermal relaxation

<sup>1</sup>Institute of Physics, National Chiao Tung University, Hsinchu 300, Taiwan, Republic of China.

<sup>2</sup>Department of Physics, National Sun Yat-Sen University, Kaohsiung 804, Taiwan, Republic of China.

<sup>&</sup>lt;sup>3</sup>Institute of Physics, Center for Condensed Matter Physics and Beijing High Pressure Research Center, Chinese Academy of Sciences, P. O. Box 603, Beijing 100080, People's Republic of China.



**Fig. 1.**  $C(T, H)/T$  vs.  $T^2$  of MgCNi<sub>3</sub> for  $H = 0-8$  T.

calorimeter from 0.6 to 10 K with magnetic fields *H* up to 8 T. Detailed description of the specific heat measurements can be found in Ref. 10. Resistivity  $\rho(T)$  was measured by the four-probe method. The change of *H* was applied at  $T = 12$  K for both  $\rho$  and *C* measurements, and the data were taken in field cooling.

 $C(T)$  of MgCNi<sub>3</sub> with  $H = 0-8$  T is shown in Fig. 1 as  $C/T$  vs.  $T^2$ . The superconducting anomaly at  $H = 0$  is much sharper than that in Ref. 1, indicating high quality of the sample, and clearly persists even with *H* up to 8 T. It is noted that  $C/T$  shows an upturn at very low temperatures. This upturn disappears in high *H*, which is a manifestation of the paramagnetic contribution like the Schottky anomaly. The normal state  $\gamma_n = 33.6$  mJ mol K<sup>2</sup> and the Debye temperature  $\Theta_{\rm D} = 287$  K can be extracted from data in Fig. 1 [5]. To derive  $H_{c2}(T)$  from  $C(T, H)$ , it is of interest to figure out  $\Delta C(T, H) = C(T, H) - C_{\text{lattice}}(T) - \gamma_n T$ .  $T_c(H)$  is thus determined by the conservation of entropy around the transition in the  $\Delta C/T$  vs. *T* plots at various *H*. The case of  $H = 0$  has been demonstrated in Ref. 5.

Figure 2 shows the transition of  $\rho(T)$  in *H* up to 8 T. The normal state  $\rho(T)$  of the present sample is slightly smaller than that in Ref. 8. It is noted that *H* did not broaden the transition width, probably due to the low temperatures investigated here.  $T_c$  deter-

mined by the criterion of 50% of the normal state  $\rho$ just above  $T_c$ .

 $H<sub>c2</sub>$  vs. *T*, resulting from both *C* and  $\rho$  measurements, is shown in Fig. 3. As expected  $T_c$  determined



**Fig. 2.** MgCNi<sub>3</sub>  $\rho(T)$  in *H* near the transition. The applied magnetic fields from right to left are 0, 0.1, 0.2, 1, 1.5, 2, 4, 6, 8 T, respectively.



Fig. 3.  $H_{c2}$  of MgCNi<sub>3</sub>. Open and solid circles denote the results from  $C$  and  $\rho$  measurements, respectively. The straight lines are the linear fits of data with  $H \leq 4$  T (roughly corresponding to  $0.8 \leq T/T_c < 1$ ). The zero field data were excluded for the fitting.

by *C* is slightly lower than that by  $\rho$ . Other than that, both techniques yielded very similar results.  $H = 8$  T lead to about 50% of  $T_c$  suppression. The data show a downward curvature in Fig. 3 as in conventional superconductors. The slopes  $(dH_{c2}/dT)$ <sub>*T*c</sub> derived from the linear fit of both *C* and  $\rho$  data for  $0.8 \le (T/T_c) < 1$  are very close to each other. The values of  $(dH_{c2}/dT)_{T_c}$ are  $2.96 \pm 0.08$  and  $2.88 \pm 0.03$  T/K from *C* and  $\rho$ measurements, respectively. These values are slightly larger than that of 2.67 T/K in Ref. 8. The relation of  $H_{c2} \approx 0.69 T_c (dH_{c2}/dT)_{T_c}$ , which does not take into account the effects of the spin–orbit interaction and the spin paramagnetic term [11], was used in Ref. [8] to estimate  $H_{c2}$  at  $T = 0$ . In this way, it led to  $H_{c2} = 13.2 \pm 0.7$  T in the present sample. However, failure to include the spin–orbit and the spin paramagnetic effects is known to significantly overestimate  $H_{c2}$ . To derive  $H_{c2}$  accurately, one has to utilize the numerical analysis, which is beyond the scope of this paper. However, certain plausible assumption can be made. It is found that the physical properties of MgCNi<sub>3</sub> are very similar to those of  $Nb<sub>0.5</sub>Ti<sub>0.5</sub>$  [5]. It seems plausible for both compound to have a similar relation between  $H_{c2}$  and  $(dH_{c2}/dT)_{T_c}$ . According to Ref. [11],  $H_{c2}$  ≈ 0.59 $T_c(dH_{c2}/dT)_{T_c}$  for Ni<sub>0.5</sub>Ti<sub>0.5</sub>. Fol-

**Table I.** Estimates of  $H_c$ <sup>2</sup> in MgCNi<sub>3</sub> by Different Approaches

Estimate approaches	$H_{c2}$ (T)
$T_c$ transition by C and $\rho$ measurements	
Spin-orbit interaction and spin	$13.2 \pm 0.7$
paramagnetic effect not included	
Spin-orbit interaction and spin	$11.2 \pm 0.6$
paramagnetic effect included	
dγ/dH	$11.5 \pm 0.6$

lowing this relation,  $H_{c2} = 11.2 \pm 0.6$  T in the present sample. It is intriguing to derive  $H_{c2}$  from  $\gamma(H)$ . The high field  $\gamma(H)$  can be estimated from the linear extrapolation to  $T = 0$ , and is found to be proportional to *H* with  $d\gamma/dH = 2.91$  mJ mol K<sup>2</sup> T, which is slightly smaller than the estimate by  $\delta C(T, H)/T$  at  $T = 0.6$  K [5]. For  $\gamma_n = 33.6$  mJ mol K<sup>2</sup>,  $H_{c2} = 11.5 \pm 1.5$ 0.6 T can be estimated, amazingly close to the above value estimated from the  $T_c$  transition in  $H$ . Estimates of  $H_{c2}$  for the present sample are summarized in Table I.

In conclusion, we have estimated  $H_{c2}$  from the  $T_c$  transition in *H* by *C* and  $\rho$  measurements. Both lead to identical value of  $H_{c2}$ . Furthermore, this value is consistent with that derived from linear  $\gamma(H)$ with respect to *H*. Since  $\gamma(H) \propto H$  is characteristic of conventional superconductors (see discussions in Refs. 12–14), the results presented in this paper have intriguing implications on the order parameter of MgCNi<sub>3</sub>.

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